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RESULTS FROM CYCLES 1 AND 2 OF NNWSI SERIES 2 SPENT FUEL DISSOLUTION TESTS

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NNWSI SERIES 2 SPENT FUEL DISSOLUTION TESTS

C. N. Wilson

ABSTRACT

Pressurized Water Reactor (PWR) spent fuel rod segments from the H. B. Robinson Unit 2 and Turkey Point Unit 3 reactors were leach tested in Nevada Nuclear Waste Storage Investigations (NNWSI) reference J-13 water under ambient hot cell conditions. The test matrix included bare fuel plus the cladding, rod segments with artificially induced cladding defects, and undefected rod segments. The testing procedures are described and comprehensive radionuclide release results are presented and discussed.

The actinides Pu, Am, Cm and Np appear to have been released congruently as the UO₂ oxide fuel matrix dissolved. Preferential U release measured in certain tests may be related to dissolution of oxidized UO_{2,x} from the fuel surface, and/or greater solubility (and mobility) of U relative to the other actinides within defected cladding specimens. Uranium solubility measured in the J-13 water was much greater than that measured in deionized water in previous tests. All of the principal fission products analyzed (¹³⁷Cs, ¹²⁹I, ⁹⁹Tc and ⁹⁰Sr) were released preferentially relative to the actinides. Preferential release of activation product ¹⁴C was also measured, with a portion of the ¹⁴C release appearing to originate from the cladding exterior surface. Much greater fractional fuel dissolution appeared to have occurred with bare fuel particles than from fuel contained in defected cladding. Measured actinide release from test specimens containing small (~200 μm) laser-drilled holes through the cladding was not significantly greater than that observed from undefected specimens.

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Spent fuel test specimens were prepared by H. H. Larson and M. E. Freed. Post-test ceramographic sections were prepared by R. D. Bell and C. E. Chamberlin. Scanning electron microscopy of fuel specimens was performed by B. Mastel. Hot cell setup and sampling operations were performed by R. T. Steele and D. V. Archer. Data base management activities for this work were performed by J. R. Stuart, E. Yatabe and B. Whitten. Technical editing of this report was provided by N. E. Kenny. Technical direction was provided by the author, who gratefully recognizes the good work and support contributed by the above and many additional unnamed HEDL personnel.

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ACRONYMS

ATM	Approved Testing Material
BCL	Battelle Columbus Laboratories
CFR	Code of Federal Regulations
EBS	Engineered Barrier System
EPA	Environmental Protection Agency
HEDL	Hanford Engineering Development Laboratory
HLW	High-Level Waste
IC	Ion Chromatography
ICP	Inductively Coupled Plasma
LLNL	Lawrence Livermore National Laboratory
MCC	Materials Characterization Center
NAA	Neutron Activation Analysis
NNWSI	Nevada Nuclear Waste Storage Investigations
NRC	Nuclear Regulatory Commission
PCI	Pellet-Cladding Interaction
PNL	Pacific Northwest Laboratory
PWR	Pressurized Water Reactor
SCC	Stress Corrosion Cracking
SEM	Scanning Electron Microscope
WHC	Westinghouse Hanford Company

RESULTS FROM CYCLES 1 AND 2 OF
NNWSI SERIES 2 SPENT FUEL DISSOLUTION TESTS

1.0 INTRODUCTION

The Nevada Nuclear Waste Storage Investigations (NNWSI) Project is investigating the suitability of the volcanic tuff at Yucca Mountain, Nye County, Nevada, for potential use as a disposal site for high-level nuclear waste. Lawrence Livermore National Laboratory (LLNL) is the lead contractor for the Waste Package Task of the NNWSI Project. Westinghouse Hanford Company (WHC) is a subcontractor to LLNL assisting them in determining the requirements for successful disposal of spent fuel rods at the Yucca Mountain Site. In the Spent Fuel Leaching/Dissolution Task at WHC, laboratory tests are being conducted with spent fuel specimens to characterize radionuclide release under NNWSI-relevant conditions.

The repository horizon under study by the NNWSI Project is in a densely welded and devitrified volcanic tuff formation that lies approximately 200 to 400 meters above the water table in the unsaturated zone. Contact of the spent fuel by water will not occur until the repository has cooled to 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 occurred. Radionuclide release from spent fuel with failed cladding to water that may enter a failed storage container is being studied in the WHC tests, which are the subject of this report. The potential for transport of dissolved radionuclides from a failed waste package to the external environment is being evaluated by other NNWSI tasks.

The Nuclear Regulatory Commission (NRC) has stated the engineered barrier system (EBS) performance requirements in 10 CFR 60⁽¹⁾ licensing procedures for geologic repositories. The two principal EBS performance requirements contained in 10 CFR 60 (Section 60.113) are:

- 1) "Containment of HLW (high-level waste) within the waste packages will be substantially complete for a period ... not less than 300 years nor more than 1000 years after permanent closure of the geologic repository (the containment period).
- 2) "The release rate of any radionuclide from the engineered barrier system (waste packages and the underground facility) following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1000 years following permanent closure" (post-containment period).

In addition, the Environmental Protection Agency (EPA) has specified⁽²⁾ cumulative release limits for radionuclides from a geologic repository (see Table 6).

In the WMC Spent Fuel Leaching/Dissolution Task, specimens of pressurized water reactor (PWR) spent fuel in configurations representing various degrees of cladding failure, have been tested in fused quartz vessels under ambient hot cell conditions. In the Series 1 Tests,* specimens prepared from Turkey Point Reactor Unit 3 fuel were tested in deionized water.⁽³⁾ Cycle 1 of the Series 1 tests was initiated in July 1983 and terminated at approximately 240 days. At termination, the test apparatus was stripped with 8 M HNO₃ and the strip solutions analyzed along with the final test solution. The test specimens were then restarted for a second cycle, which was terminated at 128 days during July 1984. Final results from both cycles of the Series 1 tests are reported in Reference 4.

The Series 2 tests, which are the subject of this report, were similar to the Series 1 tests except that: 1) the Series 2 tests were run in NRSI reference J-13 well water, and 2) the test matrix duplicated each specimen configuration using both Turkey Point and H. B. Robinson spent fuels. Cycle 1 of the Series 2 tests was started in June 1984 and terminated at

*Originally entitled, "Spent Fuel Cladding Containment Credit Tests."

181 days for the Turkey Point fuel and at 223 days for the H. B. Robinson fuel. Cycle 2 of the Series 2 tests was terminated at 195 days for the Turkey Point fuel and at 202 days for the H. B. Robinson fuel. The two Series 2 bare fuel tests were continued for a third and fourth cycle. This report discusses the results from Cycles 1 and 2 of the Series 2 tests.

Series 3 tests⁽⁵⁾ are being run in J-13 water in sealed 304 stainless steel vessels at 85°C using the same specimen configurations used in Series 1 and Series 2 tests.

The spent fuel used in these first three test series is young (~10 years from discharge) and is representative of fuel to be initially placed in the repository. However, this fuel may possibly become degraded in the post-containment period repository environment. The possibility of degradation of the spent fuel by oxidation is suggested by thermodynamic considerations. Spent fuel oxidation under NNWSI repository relevant conditions is being studied in another task of the WHC program in support of NNWSI.⁽⁶⁾ Use of spent fuel that has been degraded by oxidation is anticipated in a future spent fuel leaching/dissolution test series pending results from the ongoing WHC spent fuel oxidation studies.

2.0 TEST DESCRIPTION

The first cycle of the Series 2 tests was initiated and run in accordance with an approved test plan.⁽⁷⁾ Supplemental procedures for test Cycle 1 termination and the subsequent Cycle 2 restart are given in Reference 8. The testing procedures are summarized here.

2.1 TEST CONFIGURATION AND SAMPLING

Two basic test configurations were used as shown in Figure 1: one for bare fuel specimens and one for clad fuel specimens (with or without artificially induced cladding defects) with water tight end fittings. The tests were conducted in fused quartz test vessels with loose fitting lids under ambient hot cell air and temperature (~25°C) conditions. The tests were run in 250 ml of NHWSI reference J-13 well water. The tests were "semi-static" in that the leachate solution was periodically sampled and the sample volume replenished with fresh J-13 water. The semi-static method provides solution radionuclide content as a function of time and simulates limited flow through a breached waste container. Fused quartz rods were also periodically removed and stripped with 8 M HNO₃ to monitor progression of radionuclide plateout. The rod samples were gently rinsed with J-13 water before acid stripping to remove adhering test solution.

After several months, the tests were terminated and restarted in fresh J-13 water using the same test specimens. The following four types of samples were taken at test cycle termination:

- Final solution
- Final fused quartz rod
- Vessel and specimen rinse solution
- Vessel acid strip solution

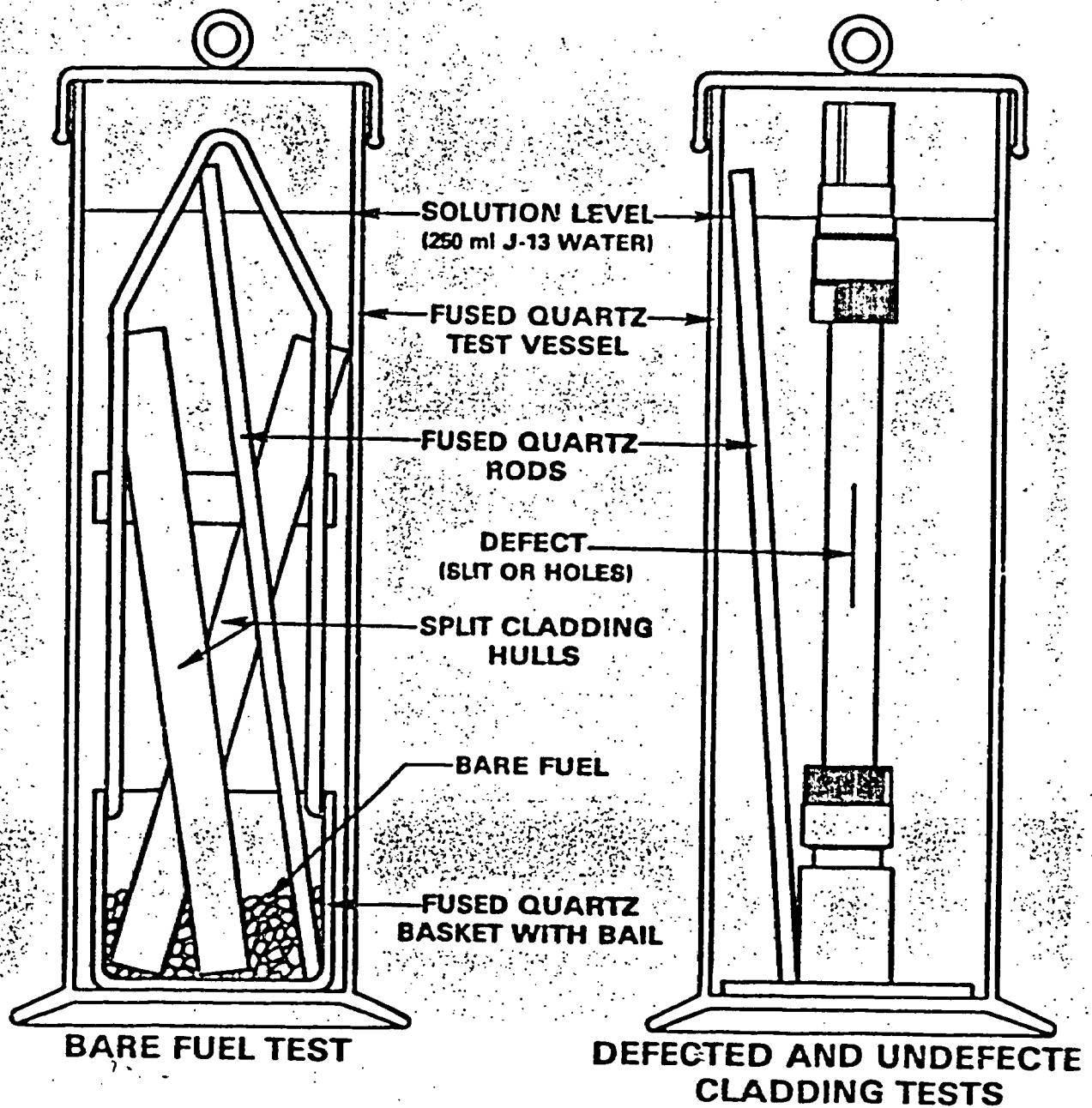


FIGURE 1. Test Configurations.

HEDL 8401-17.

The final solution sample was taken by the same procedure as periodic solution samples but was considered, during subsequent radionuclide release evaluation, to represent the full 250 ml test solution volume. After final solution removal, the inside of the test vessels, test specimens, and specimen holders (bare fuel basket or clad specimen pedestal) were rinsed with fresh J-13 water. Bare fuel particles were rinsed by covering the particles with J-13 water in a small beaker, gently rocking, and decanting the rinse water; this procedure was repeated several times. All rinse water from a given test was collected as a combined rinse solution made up to 600-ml volume with additional J-13 water. After rinsing, the test vessels containing their respective bare fuel baskets or specimen pedestals were filled with 300 ml of 8 M HNO_3 and left to sit overnight. The acid strip solution was sampled the next day, after pouring the acid back and forth between the test vessel and an appropriate clean container.

Two bare fuel particles were removed from each bare fuel test at cycle termination for mounting, sectioning and optical ceramographic examination. Rinse solutions from bare fuel tests were allowed to settle and samples of the residue (fine particulate sediment) were obtained for scanning electron microscopy (SEM) examination.

2.2 TEST SPECIMENS

The test matrix included eight test specimens, one each of four specimen configurations of two fuel types. The test specimens are identified in Table 1. All specimens were prepared from five-inch-long fuel rod sections. The four specimen configurations are:

- Undefected specimens with intact cladding and water-tight end fittings were run as control samples to indicate the amount of radionuclide release originating from residual cladding surface contamination and cladding crud deposits.
- Hole defect specimens contained two small (~200- μm diameter) laser-drilled holes through the cladding near the center of the specimens. The ends of the hole defects specimens were also sealed with water-tight end fittings. The hole defects specimens

are intended to be representative of fuel rods containing small breaches, such as may result from pellet-cladding interaction (PCI) or stress corrosion cracking (SCC) cladding failures.

- Slit defect specimens contained a 0.006-inch wide by 1-inch long machined slit through the cladding near the specimen center and were sealed at the ends with water-tight end fittings. The slit defect specimens are intended to represent a more severe cladding failure, such as may occur if an SCC defect progressed to a relatively large crack, or cladding cracks that may result from postirradiation handling.
- Bare fuel specimens were prepared by machining axial slits through the cladding from end-to-end on opposite sides of the specimens, opening the split cladding, and removing the bare fuel particles from the cladding. The cladding hulls are included with the fuel particles in these tests as part of the test specimen. The bare fuel specimens are intended to represent the worst-case cladding failure, where the cladding has split open and the fuel has fallen out.

TABLE 1
SERIES 2 TEST SPECIMENS

<u>Specimen Identification</u>	<u>Specimen Configuration</u>	<u>Fuel Type</u>	<u>Fuel Weight (grams)</u>
C5C-A	Undefected	H. B. Robinson	81.82
C5C-C	Holes Defect	H. B. Robinson	84.41
C5C-E	Slit Defect	H. B. Robinson	85.84
C5C-H	Bare Fuel	H. B. Robinson	83.10(a)
19-1	Undefected	Turkey Point	50.00(b)
19-12	Holes Defect	Turkey Point	50.93
19-19	Slit Defect	Turkey Point	48.98
19-24	Bare Fuel	Turkey Point	27.21(c)

(a) 81.92 g for Cycle 2.

(b) Estimated weight.

(c) 26.66 g for Cycle 2.

An important part of the specimen preparation procedures was the removal from the cladding exterior surface of fine particulate contamination that results from sectioning and handling in contaminated hot cells. Such contamination would seriously bias the test results if not removed. The primary purpose of the undefected test specimens in the test matrix was to provide an indication of the released radioactivity originating from residual contamination on the cladding exterior surface. Before installing end fitting hardware on the undefected, laser-drilled, and slit defect specimens, the cladding surface was decontaminated to less than 50 cpm smearable alpha and less than detectable beta/gamma above the HEDL 327 Building background (~150 cpm in a lead shielded cave). The cladding exterior of the bare fuel specimens was also decontaminated to less than 50 cpm smearable alpha before axially splitting the cladding to remove the bare fuel.

The ends of the undefected, laser-drilled, and slit defect specimens were sealed using water-tight end fittings fabricated from modified Cajon Model SS-8-UT-A-10 Ultra Torr® vacuum adaptors. The end-fitting seal was made using ethylene propylene O-rings chosen for their radiation resistance and water compatibility. The top fittings of both the hole defects and slit defect specimens contained a small vent hole above the test solution level to allow the defected cladding specimens to fill with solution to the external test solution level.

2.3 FUEL CHARACTERISTICS AND HISTORY

The two fuel types used in the test matrix (see Table 1) were similar PWR fuels as indicated by the relevant fuel characteristics listed in Table 2. Both fuels were low gas release PWR fuels from the same vendor and approximately the same vintage.

The H. B. Robinson fuel was obtained through the Pacific Northwest Laboratory (PNL) Materials Characterization Center (MCC) as an "approved testing material" (ATM) for geological repository testing and was identified by

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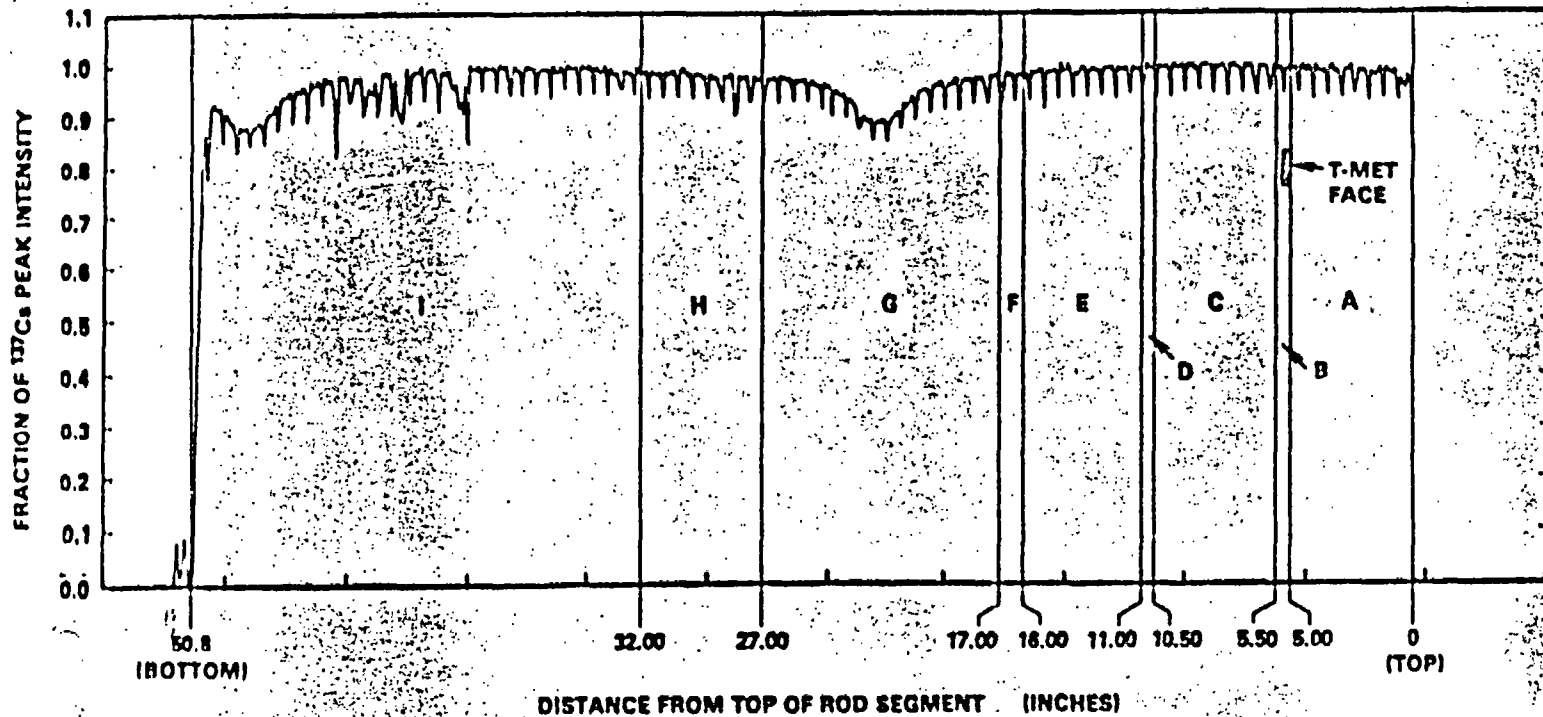
TABLE 2

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
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	--

PNL-MCC as ATM-101.⁽⁹⁾ All four of the Series 2 test specimens, plus samples for burnup and metallographic characterization were sectioned from the ATM-101 rod segment identified as segment C5C, as shown in Figure 2. The C5C segment was the central one-third of the C5 rod from the B0-5 H. B. Robinson spent fuel assembly. The specimen axial locations from the C5C segment were chosen to avoid regions of nonuniform burnup at spacer grid locations.

The Turkey Point Reactor Unit 3 spent fuel specimens were sectioned from the I-9 rod of the B-17 assembly at Battelle Columbus Laboratories (BCL) in 1979. Sectioning diagrams and characterization data for the I-9 rod sections and other B-17 assembly fuel rods sectioned at this time are contained in Reference 10. Approximately one inch of fuel was removed from each end of the 5-inch-long rod sections at the time of sectioning. Therefore, the fuel weights as indicated in Table 1 are less than indicated for the H. B. Robinson sections, which contained a full 5 inches of fuel.



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|------------------------------|--------------------------------|
| A. Undeformed Specimen | F. Longitudinal-Metallographic |
| B. Transverse-Metallographic | G. Spare (MCC Archive) |
| C. Laser-Drilled Specimen | H. Bare Fuel Specimen |
| D. Burnup | I. Spare (used in Series 3) |
| E. Slit Specimen | |

FIGURE 2. Sectioning Diagram for H. B. Robinson Spent Fuel Rod Segment C5C Superimposed on ^{137}Cs Gamma Scan.

In addition, some fuel had been lost from Section I-9-24, accounting for its lighter weight relative to Sections 12 and 19. Fuel weights were calculated by subtracting the weight of 5 inches of cladding (16.40 grams) from the specimen weight. Section 1 was from the bottom of the fuel rod, and its fuel weight was estimated (since an accurate weight of the bottom end cap plus cladding was not available) to be approximately 50 grams. The use of an estimated fuel weight for the I-9-1 undefected specimen is not considered significant since ^{137}Cs release data indicated no water entered the specimen during testing, and all release was assumed to be from cladding surface contamination. The bare fuel specimens were reweighed at the end of Cycle 1, after removing particles for radiometallurgical examination.

Grain size is probably the most significant difference between the two fuel types relative to potential leaching behavior. Initial as-fabricated grain size was estimated to be approximately 6 μm for the H. B. Robinson fuel versus 25 μm for the Turkey Point fuel, based on post-test radiometallographic examination of Series 2 specimens (see Figure 3). Both fuels had low gas release and exhibited little restructuring during irradiation. However, a small amount of central grain growth was observed in the finer-grained H. B. Robinson fuel. The additional specimen internal free volume in the Turkey Point holes and slit defect specimens relative to the H. B. Robinson specimens (which contained a full 5 inches of fuel) may also have been significant for these test configurations because of the resulting differences in specimen internal fuel-to-water ratios. Another possibly significant difference between the two fuel types is the length of time between specimen sectioning and testing. The H. B. Robinson specimens were sectioned from segment C5C a few weeks before Cycle 1 startup, and original C5 rod sectioning was within the same year. The I-9 rod Turkey Point specimens had been sectioned approximately 5 years before Cycle 1 startup and stored in air in a sealed metal can.

Radionuclide inventories for each fuel type were calculated by linear interpolation of ORIGEN-2 inventory data given for a 10-year old PWR fuel in Reference 9. A burnup value of 27.7 Mwd/MTM reported in Reference 10 was

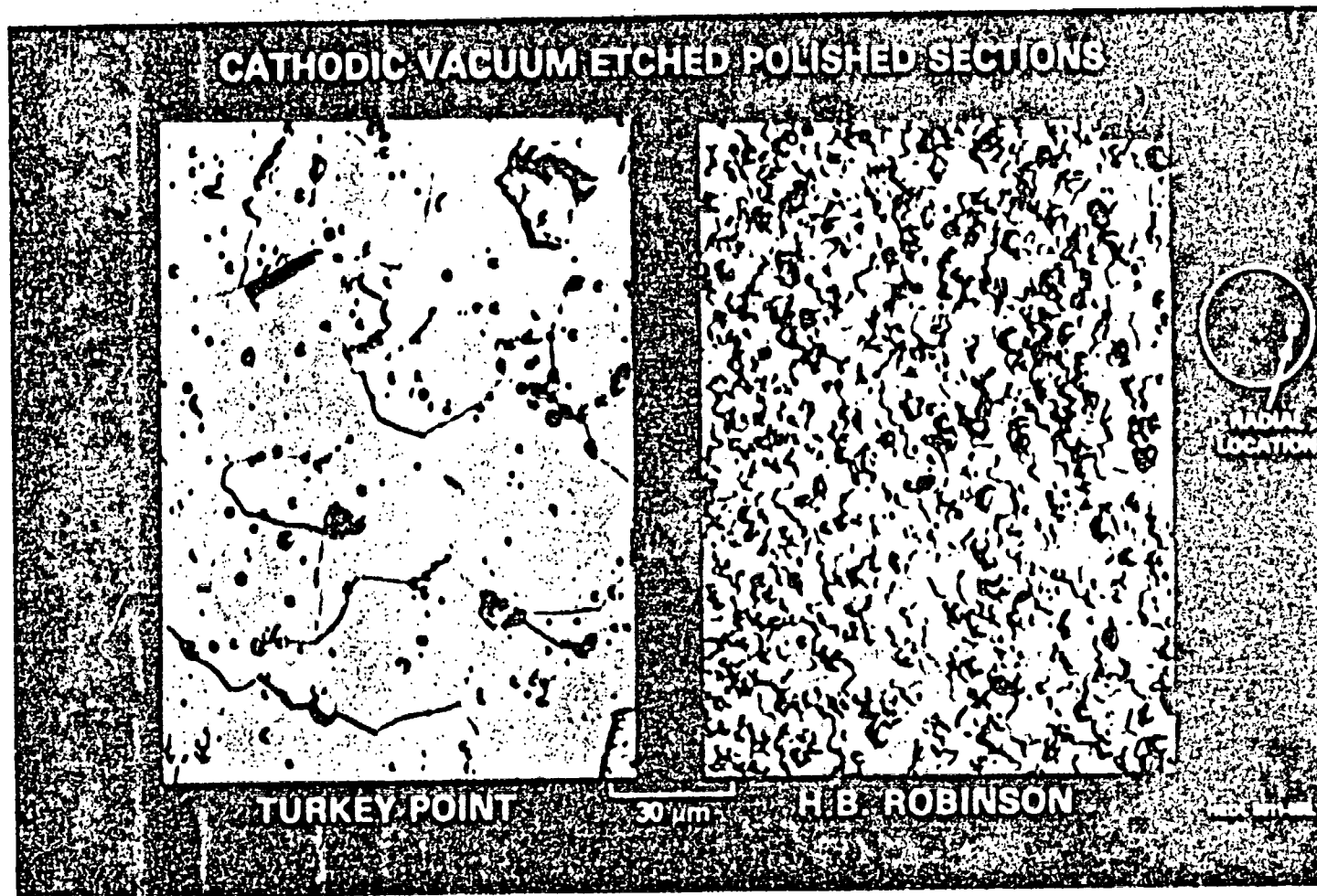


FIGURE 3. Microstructures of Irradiated Turkey Point and H. B. Robinson Spent Fuels in the Unrestructured Zone Near the Cladding. Cathodic vacuum etched to reveal grain structure, Neg 8507895-1cn

used for the Turkey Point fuel. A burnup value of 30.2 MWD/MTM was radiochemically determined by WHC on the C5C-D section from the H. B. Robinson fuel. Inventory values for ^{244}Cm , ^{241}Am , $^{239+240}\text{Pu}$, ^{237}Np , ^{137}Cs , and ^{99}Tc were also directly measured by radiochemical methods during burnup analysis on the C5C-D section. ^{14}C was also directly measured on two H. B. Robinson sections. For consistency, the ORIGEN-2 calculated inventories were used for fractional release calculations on both fuel types. The ORIGEN-2 based inventory data for both fuels and radiochemically determined inventories for the H. B. Robinson fuel are given in Table 3.

2.4 SAMPLE ANALYSES

All solution chemical and radiochemical analyses performed by WHC followed approved procedures prepared for testing in support of geologic repositories. Principal nuclides analyzed are listed in Table 4 with detection limits. The ability to analyze less than one part in 100,000 of the test specimen inventory dissolved in the 250 ml of test solution was desired for all radiochemical methods used based on the NRC 10 CFR 60⁽¹⁾ requirement that "the release rate of any radionuclide ... shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1000 years." Relative 1000-year inventories in spent fuel for radionuclides with half-lives greater than one year and activity equal to or greater than ^{129}I are given in Table 5. Except for radium, thorium, and tin isotopes, an isotope of each radionuclide element listed in the EPA 40 CFR 191⁽²⁾ cumulative release limits (see Table 6) is included in the Table 4 list of nuclides analyzed. The ^{226}Ra , ^{230}Th and ^{232}Th isotope activities build up in spent fuel as actinide decay chain products and are not a significant component of spent fuel activity until after several thousand years, and were not practical to analyze for release in the current tests. ^{126}Sn will replace ^{79}Se in Table 4 for specifically analyzed radionuclides in the Series 3 tests.

TABLE 3
 SERIES 2 SPECIMEN RADIONUCLIDE INVENTORIES(a)

Inventories ($\mu\text{Ci/g}$ oxide fuel)	H. B. Robinson ORIGEN-2	H. B. Robinson Measured ^(b)	Turkey Point ORIGEN-2
Burnup (Mwd/kgM)	30.2(b)	30.2	27.7(c)
Uranium ($\mu\text{g/g}$ oxide fuel)	8.40×10^5	--	8.48×10^5
^{244}Cm	1.38×10^3	1.43×10^3 (d)	1.01×10^3
^{241}Am	1.56×10^3	1.63×10^3	1.45×10^3
$^{239+240}\text{Pu}$	7.44×10^2	7.16×10^2	7.04×10^2
^{237}Np	2.42×10^{-1}	2.35×10^{-1}	2.18×10^{-1}
^{137}Cs	6.67×10^4	6.57×10^4	6.11×10^4
^{129}I	2.65×10^{-2}	--	2.42×10^{-2}
^{99}Tc	1.05×10^1	8.34×10^0	9.74×10^0
^{90}Sr	4.37×10^4	--	4.08×10^4
^{79}Se	3.71×10^{-1}	--	3.02×10^{-1}
^{14}C	--	(e)	--

(a) Calculated from ORIGEN-2 data in PNL-5109(9) assuming 10-year old fuel, unless otherwise noted.

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

(c) Red I-9 reported burnup, Reference 10.

(d) Actually $^{244}\text{Cm} + ^{241}\text{Am}$ since both isotopes have similar alpha energies, ORIGEN-2 data indicate that ^{244}Cm is ~1% of $^{244}\text{Cm} + ^{241}\text{Am}$.

(e) ^{14}C average of values measured on samples C5C-J and C5B-C:

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

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

TABLE 4
RADIOCHEMISTRY METHODS

Radionuclide	Method	Detection Limits		10 ⁻⁵ Inventory (pCi/ml)*
		(pCi/ml)	(ppb)	
²⁴⁴ Cm	α-spectrometry	0.2	3 x 10 ⁻⁶	4580
²⁴¹ Am	α-spectrometry following separation	0.2	6 x 10 ⁻⁵	4700
²³⁹⁺²⁴⁰ Pu	α-spectrometry	0.2	0.003	2350
²³⁷ Np	α-spectrometry following separation	0.2	0.3	0.9
¹³⁷ Cs	γ-spectrometry	200	0.002	2.4 x 10 ⁵
¹²⁹ I	Neutron activation analysis	10 ⁻⁵	0.0001	0.09
⁹⁹ Tc	β-proportional counting following separation	20	1.2	40
⁹⁰ Sr	β-proportional counting following separation	20	0.0001	1.4 x 10 ⁵
⁷⁹ Se	Liquid scintillation counting following separation	20	0.3	1.2
⁶⁰ Co	γ-spectrometry	200	0.0002	**
¹⁴ C	Liquid scintillation counting following separation	20	0.004	2
U	Fluorescence	--	1	(3 ppm)

* Assumes 10⁻⁵ of H. B. Robinson test specimen inventory released to 250 ml.
** ⁶⁰Co inventory is variable.

TABLE 5

PWR SPENT FUEL RADIONUCLIDE INVENTORIES AT 1000 YEARS^(a)

Radionuclide ^(b)	Ci/1000 MTM	% of Total 1000-Year Activity	Cumulative %
Am-241	894,500	51.33	51.33
Am-243	31,080	1.78(c)	53.11
Pu-240	476,900	27.37	80.48
Pu-239	304,700	17.45	97.96
Pu-242	1,755	0.10	98.07
Pu-238	967	0.06	98.12
Tc-99	13,030	0.75	98.87
Ni-59	5,150	0.295	
Ni-63	354	0.020	
Zr-93	1,933	0.111	
Nb-93m	1,836	0.105	
Nb-94	1,240	0.071	
C-14	1,372	0.079(d)	
U-234	1,984	0.114	
U-238	317	0.016	
U-236	271	0.018	
Np-237	1,000	0.057	
Sn-126	772	0.044	
Se-79	405	0.023	
Cs-135	345	0.020	
Sm-151	163	0.009	
Pd-107	112	0.006	
I-129	32	0.0018	

(a) Based on ORIGEN-2 data reported in ORNL/TM-7431(11) for 33,000 Mwd/MTM burnup PWR spent fuel, actinides plus fission products plus activation products.

(b) Radionuclides with 1000-year activity less than 129I or half-life less than 1 year omitted.

(c) Includes activity of ^{239}Np daughter products.

(d) ^{14}C activity may vary considerably depending on as-fabricated nitrogen impurities.

TABLE 6
EPA RELEASE LIMITS

Radionuclide	Cumulative Release Limit (Ci/1000 MTHM)
^{241}Am or ^{243}Am	100
^{14}C	100
^{137}Cs or ^{135}Cs	1,000
^{129}I	100
^{237}Np	100
^{238}Pu , ^{239}Pu , ^{240}Pu , or ^{242}Pu	100
^{226}Ra	100
^{90}Sr	1,000
^{99}Tc	10,000
^{230}Th or ^{232}Th	10
^{126}Sn	1,000
^{233}U , ^{234}U , ^{235}U , ^{236}U , or ^{238}U	100
Any other alpha-emitting radionuclide with a half-life greater than 20 years	100
Any other radionuclide with a half-life greater than 20 years that does not emit alpha particles	1,000

A brief summary of each radionuclide analysis procedure follows:

$^{240}\text{Pu} + ^{239}\text{Pu}$ (and ^{244}Cm) -- An accurately measured quantity (i.e., 100 to 500 $\mu\text{g} \pm 1 \mu\text{g}$) of solution was evaporated onto a stainless steel "source disk," which was then counted for total alpha activity and analyzed by alpha energy spectrometry. The alpha spectrometry results were used to calculate the portions of the total alpha counts originating from each alpha energy peak. Three significant alpha spectrometry peaks generally resulted: $^{239}\text{Pu} + ^{240}\text{Pu}$, $^{238}\text{Pu} + ^{241}\text{Am}$ and ^{244}Cm . Results for all three alpha activities were reported.

^{241}Am -- Since ^{241}Am and ^{238}Pu have similar alpha decay energies, a separation is required for ^{241}Am analysis. A 500- μl solution sample is reacted with 200 μl of 5 M hydroxylamine hydrochloride and 100 μl of 8 M HNO_3 and passed through an anion exchange resin. The Pu is loaded onto the resin while the Am passes through the resin. The ^{241}Am is then measured by alpha counting and alpha spectrometry after evaporating onto a stainless steel source disc that has been heated to dull red by flame from the bottom side to burn off organic and volatile residues.

The Am separation was not performed on periodic solution samples during Cycle 1 of the tests. A one-time determination of $^{238}\text{Pu}/(^{239}+^{240}\text{Pu})$ ratio was made on test solution from all but the undefected tests, and this ratio was used to calculate ^{241}Am from $^{238}\text{Pu} + ^{241}\text{Am}$ and $^{239}+^{240}\text{Pu}$ data on the Cycle 1 periodic samples. Starting with the Cycle 1 termination samples, direct ^{241}Am analysis following separation was performed on all samples.

Gamma Spectrometry -- ^{137}Cs and ^{134}Cs were measured in nearly all samples by gamma spectrometry. ^{60}Co was also measured in many samples by gamma spectrometry, and some rod and strip samples from bare fuel tests also showed indications of other gamma-emitting isotopes. Quantitative gamma counting data were calculated from gamma energy peaks based on daily measurement of control standards of ^{241}Am , ^{137}Cs and ^{60}Co mixtures run under the same geometry as the test samples.

^{237}Np -- Since ^{237}Np activity in 10-year old spent fuel is much lower than other alpha-emitting isotopes and is interfered with by ^{234}U alpha decay, a separation is required. Np is separated by cation exchange of NpO_2^+ followed by solvent extraction of Np^{+4} into thenoyltrifluoroacetone in xylene. The ^{237}Np activity is then evaporated onto a stainless steel source disk that is heated to "a high heat" on a hot plate to burn off organic residues. The ^{237}Np is counted using alpha counting and alpha spectrometry. The volume of sample used was 0.5 ml for Cycle 1 ^{237}Np analyses. Volumes for Cycle 2 were: 2.0 ml for 20-day, 0.5 ml for 62-day, 1.0 ml for 154-day, and 1.0 ml for all termination sample analyses.

^{99}Tc -- Since ^{99}Tc decays solely by beta decay, spectrometry methods are not applicable and the ^{99}Tc must be separated from other beta emitters to be counted. The Tc is oxidized to TcO_4^- with sodium dichromate and separated from most other radioactive species that adsorb onto a cation exchange resin. Tc is then extracted from the cation exchange effluent into hexone as tetraphenylarsonium pertechnetate. A measured portion of the hexone is evaporated onto a stainless steel source disc under a heat lamp. The ^{99}Tc activity is then counted using a gas flow beta proportional counter.

^{90}Sr -- Analysis for ^{90}Sr was begun during Cycle 2. Since ^{90}Sr is a beta emitter, separation is required. The ^{90}Sr is separated from other radioactive species by selective elution from a cation exchange resin using 2-methylactic acid. Following separation, the growth of yttrium-90 is measured by beta counting. The ^{90}Sr is then calculated, based on the growth of the ^{90}Y daughter over a measured period of time. The technique of Roberts (1961) is used. (12)

^{79}Se -- Since ^{79}Se is a beta emitter, separation is required before counting. The test sample is mixed with 0.5 M HNO_3 saturated with bromine and a Se carrier solution prepared by dissolving Se metal in HNO_3 and diluting to 0.5 M HNO_3 . ^{79}Se is separated from other radioactive species by passing the prepared solution through a cation plus anion exchange resin column. The selenium in the column effluent is distilled from hydrobromic acid and precipitated as metal by reducing it with hydroxylamine hydrochloride. The metal is dissolved in nitric acid, and the ^{79}Se is measured using liquid scintillation counting. (^{79}Se was below detectable limits in all Series 2 samples analyzed for ^{79}Se and was later deleted from the list of radio-nuclides analyzed and replaced by ^{126}Sn .)

^{14}C -- Carbon-14 is separated from other radioactive species by distillation of carbon dioxide from an acidic oxidizing solution. The carrier gas (air) is passed through Ascarite II* to remove carbon dioxide. The distilled

*Ascarite II is a registered trademark of Arthur H. Thomas Co., Philadelphia, PA.

^{14}C dioxide is trapped in a liquid scintillation cocktail made basic with sodium hydroxide.

^{129}I -- Because of its low concentration and long half-life (17,000,000 yrs), ^{129}I activity in spent fuel leach solutions is much too low to detect by direct counting methods after separation. Therefore, a neutron activation analysis (NAA) procedure (performed by PNL) is used for ^{129}I . A measured volume of test solution and a "spike" solution containing ^{125}I are absorbed into quartz wool in a gas phase separation column. The sample is evaporated from the quartz wool by flaming the outside of the column while flowing air through the column. The oxidized iodine vapor species are trapped by absorption onto activated charcoal. The column is then evacuated, the charcoal trap valved off from the vacuum, and then the charcoal trap is heated, reducing the iodine to I_2 . The I_2 vaporizes from the heated charcoal trap and is condensed in a liquid N_2 cold trap. The column is sealed off above the liquid N_2 cold trap creating a glass ampoule containing the separated I_2 . The ampoules are then activated by neutron irradiation in a nuclear reactor (Hanford N Reactor when available). The irradiated ampoules are then broken in CCl_4 , in which I_2 is soluble. ^{126}I and ^{130}I decay are then counted by gamma spectrometry over several ^{130}I half-lives (12.4 hours), and the original preirradiation ^{129}I is calculated.

Uranium -- Uranium is determined using a Scintrex* model UA-3 uranium analyzer. Diluted aqueous solutions containing a buffered complexing reagent are excited by a UV-emitting nitrogen pulsed laser, and the green uranium fluorescence emitted from the solution is measured. An increase in signal from the addition of a known uranium-containing standard solution to the diluted test solution is used to calculate uranium concentration. The method gives reproducible results for sample uranium concentrations down to 1 ppb.

*Scintrex Ltd, Concord, Ontario, Canada

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3.0 RESULTS AND DISCUSSION

A complete tabulation of radiochemical results reported in pCi activity units (μg for uranium) is contained in Appendix A. Tables of conversion factors and equations for calculating isotopic and elemental concentrations from pCi activity data and discussion of radiochemistry error estimates are also included in Appendix A. A complete tabulation of solution chemistry data (pH, cation and anion concentrations) is contained in Appendix B. Data for major radionuclides discussed below are presented in plots showing activities (concentration for U) in the unfiltered solution with time, and in tabular form showing the distribution of "measured release" components between the different sample types.

Unfiltered solution concentrations for major radionuclides are plotted in Figures 4 through 10 for the four test configurations. Data points in these plots with arrows pointing downward are data reported as "less than" values. Circled data points have identical reported coordinate values but are vertically offset within the circle to show the individual data point symbols. Data lines are drawn to the actual data coordinates for the circled data points. When all data points within a circle are "less than" values, a downward arrow is drawn from the circle. If a single data point within a circle is a "less than" value, the downward arrow is drawn from that data point. Lines to data points within a circle are drawn into the circle, and lines not associated with data points within a circle through which the lines pass are blanked within the area of the circle (for an example, see Figure 10).

The activity (concentration for U) level that would be obtained if 10^{-5} of the test specimen radionuclide inventory dissolved in the 250 ml of test solution is shown on each plot. Since the test specimen fuel masses varied significantly in the Turkey Point fuel tests, the activity corresponding to 10^{-5} of the inventory dissolved in 250 ml of solution is shown for all three defected cladding configurations in the Turkey Point periodic solution sample plots. One 10^{-5} inventory level is shown in the solution sample plots for the H. B. Robinson fuel tests since the fuel mass was nearly equal

(84 ± 2 g) for all four specimens. The 10^{-5} inventory level was chosen as a reference value to compute "fractional releases" and for comparison of release data since this value falls within the plotted range for all the radionuclides. This value cannot be directly compared to the NRC release-rate limit of 1 part in 10^5 per year of the 1000-yr inventory.

An accounting of the total measured release for uranium, actinide radionuclides and fission product radionuclides is given in Tables 7 through 16, based on radiochemical results tabulated in Appendix A. Data reported as "less than" values (below detection limit) are included in values given in Tables 7 through 16, and are indicated by a "less than" symbol where a significant portion of the value given is based on "less than" data. Totals that include "less than" values are not given as less than if the included "less than" values are less than 5% of the total value. A summary of Cycle 1 and 2 "total measured fractional release" values from Tables 7 through 16 is contained in Table 17. Activities of ^{14}C measured in test samples during Cycles 1 and 2 are given in Table 18. Less than data are not included in the release values calculated for ^{14}C and ^{60}Co activation products in Tables 19 and 20. Tables 7 through 20 are at the end of their respective radionuclide section.

The tabulated values listed for "Solution Samples" are the sum of the products of unfiltered solution activity/ml (concentration for U) times sample volume for all periodic solution samples taken during a test cycle, excluding the final solution sample taken at termination of the test cycle. The terminal "Final Solution" value is the activity/ml (concentration for U) of the unfiltered final solution sample taken at the end of a test cycle times 250 ml. The concentration of the "Final Solution" sample is given in parentheses below the "Final Solution" value in units indicated in the left

*The Cycle 1 "Rod Samples" values also contain activity (mass for U) measured in the rod rinse solution from the 6-day rod sample. The 6-day Cycle 1 rod rinse sample was the only rod rinse solution analyzed and is reported in units per rod in Appendix A.

column. The "Rod Samples" value is the sum of the activity (μg mass for U) stripped from all rod samples taken during a test cycle.* The terminal "Rinse" value is the activity/ml ($\mu\text{g}/\text{ml}$ for U) measured in the collected terminal rinse solution times the 600-ml terminal rinse volume. The "Acid Strip" value is the activity/ml ($\mu\text{g}/\text{ml}$ for U) measured in the 8 M HNO_3 vessel strip solution at cycle termination times the 300-ml acid strip volume. The "Total Release" value is the total measured release obtained by summing all the above release quantities for each sample type. Total measured fractional release (total release \div 10^{-5} inventory) is calculated for each radionuclide for both test cycles and for the sum total release for Cycles 1 and 2.

Specimen inventory values used for fractional release calculations were obtained by multiplying the Table 3 ORIGEN-2 based per gram fuel inventories times the fuel weights given in Table 1. The "% in Solution" value is the sum of the "Solution Samples" plus "Final Solution" values ($\times 100\%$) divided by the "Total Release" value for each test cycle.

3.1 URANIUM

Solution uranium concentrations as shown in the Figure 4 semi-log plots reached relatively stable levels after a few days, with the most notable exception being an early peaking of uranium concentration in the Cycle 1 H. B. Robinson bare fuel test. Uranium concentrations generally correlated with the severity of the cladding defect, with concentration being greatest in the bare fuel tests, followed by the slit defect tests, followed by the hole defects tests. Greater uranium concentration in the undefected test versus hole defects in the Cycle 1 H. B. Robinson tests is most likely the result of greater residual cladding exterior surface contamination on the undefected specimen at the start of the tests. Much of the cladding exterior contamination present on the undefected specimen appears to have been removed during Cycle 1 by dissolution during the test and/or by postcycle rinsing. During Cycle 2, the uranium concentrations were approximately equal for the undefected and hole defects tests for both fuel types.

Greater uranium concentrations for the Cycle 1 Turkey Point "slit" and "holes" tests versus the respective H. B. Robinson tests may be related to the greater internal water-to-fuel volume ratio for the Turkey Point specimens, and also to the initial presence of a more oxidized fuel surface in the Turkey Point specimens, which experienced much more extensive exposure to air before testing.

A difference in the extent of initial fuel surface oxidation may be responsible for the observed differences in uranium concentration behavior between the H. B. Robinson and Turkey Point bare fuel tests, as indicated by the linear unfiltered uranium concentration plots in Figure 5. Uranium appears to have supersaturated during Cycle 1. Uranium concentration in the H. B. Robinson bare fuel test peaked at 4.5 $\mu\text{g/ml}$ on Day 6 and decreased to 1.2 $\mu\text{g/ml}$ at Cycle 1 termination as uranium apparently equilibrated with a phase having lower solubility than the phase initially present on the fuel surface. Uranium concentration in the Turkey Point bare fuel tests

began to decrease only after 120 days from 4.8 $\mu\text{g/ml}$ to 4.0 $\mu\text{g/ml}$ at Cycle 1 termination, suggesting that more of the higher solubility phase was initially present. The bare fuel was dried in air between Cycles 1 and 2.

Uranium concentration in both bare fuel tests tended towards $\sim 1 \mu\text{g/ml}$ in subsequent test cycles as the higher solubility surface phases were apparently depleted. During Cycle 2, unfiltered uranium concentration in the H. B. Robinson bare fuel test was 2.4 $\mu\text{g/ml}$ at day 20 and decreased to 2.0 $\mu\text{g/ml}$ at the end of Cycle 2. Uranium concentration in Cycle 2 of the Turkey Point bare fuel test was 1.4 $\mu\text{g/ml}$ in the initial 20-day sample, peaked at 2.6 $\mu\text{g/ml}$ in the 154-day sample, and dropped to 2.4 $\mu\text{g/ml}$ in the 195-day final solution sample. At the end of Cycle 3 (224 days), unfiltered uranium concentration was 1.4 $\mu\text{g/ml}$ and 1.2 $\mu\text{g/ml}$, respectively, for the H. B. Robinson and Turkey Point bare fuel tests. Cycle 3 was terminated and Cycle 4 started on the same day; this did not allow the fuel to dry. The initial Cycle 4 sample (7 day) showed unfiltered uranium concentrations of 0.65 $\mu\text{g/ml}$ and 0.37 $\mu\text{g/ml}$, respectively, for the H. B. Robinson and Turkey Point bare fuel tests. The 63-day Cycle 4 unfiltered uranium concentrations were 1.2 $\mu\text{g/ml}$ and 0.71 $\mu\text{g/ml}$, respectively, for these two tests.

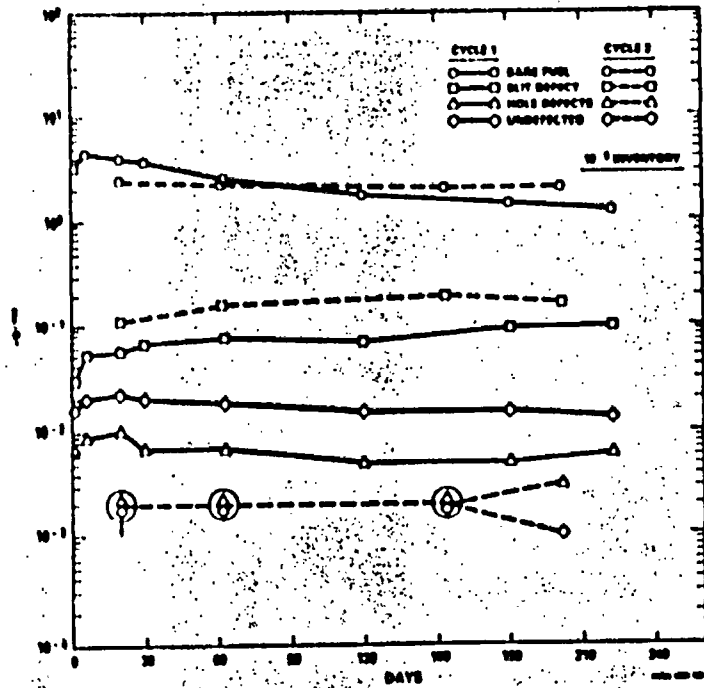
The uranium solubility behavior was significantly different in J-13 water than in deionized water in the Series 1 tests. Essentially all uranium measured in unfiltered J-13 water Series 2 solution samples passed through 0.4- μm and 18- λ filters. In the Series 1 deionized water tests, real uranium solubility appeared to be $\sim 0.001 \mu\text{g/ml}$ versus 1 to 2 $\mu\text{g/ml}$ in later Series 2 filtered samples. In Cycle 1 of the Series 1 bare fuel test, uranium peaked in unfiltered solution at 4.3 $\mu\text{g/ml}$ on Day 1, decreasing to the 0.3 to 0.6 $\mu\text{g/ml}$ range through Day 202, and then dropped to 0.003 $\mu\text{g/ml}$ in the 250-day final solution sample. (Series 1 Cycle 1 samples were not filtered until Day 202.) Uranium concentrations ranged from 0.001 to 0.004 $\mu\text{g/ml}$ in later Cycle 1 and all Cycle 2 0.4- μm and 18- λ filtered samples, suggesting that true uranium solubility in the deionized water tests was on the order of 0.001 $\mu\text{g/ml}$ or less and that excess uranium above this level was likely in a colloidal state that precipitated with time. Higher uranium solubility

in the Series 2 tests is attributed to the ~120 $\mu\text{g/ml}$ of HCO_3^- ion in J-13 water that complexes with the uranium (likely with UO_2^{+2}), stabilizing uranium in solution.

The uranium release accounting data in Table 7 indicate greater measured fractional release from Turkey Point fuel relative to H. B. Robinson fuel in all three defected cladding configurations. Again, more extensive pre-test exposure of the Turkey Point fuel to air may be involved. The larger specimen internal solution volume with good communication to external solution (through a relatively large defect) is probably also a contributing factor to the approximate order of magnitude greater "total measured fractional release" observed in the Turkey Point slit defect test versus the H. B. Robinson slit defect test.

Total release values tabulated for actinides in bare fuel tests in Tables 7-11 are likely to be conservative since fine undissolved fuel particles may have been components of the acid strip samples. Summing the uranium contained in solution samples plus the peak measured uranium concentration multiplied by 250 ml gives the minimum amount of uranium known to have been in solution. Dividing by 10^{-5} of specimen inventory yields "soluble" fractional release values of 2.0 and 6.6, respectively, for Cycle 1 of the H. B. Robinson and Turkey Point bare fuel tests and 1.1 and 3.5, respectively, for Cycle 2 of these tests. The "% in solution" given for the Cycle 1 H. B. Robinson bare fuel test would increase from 14% to 35% based on the amount known to have been in solution rather than "solution samples" plus "final solution." Subtracting the soluble fractional release values from the total fractional release values listed in Table 7 ($\pm 10^{-5}$ Inv.) leaves ~3.7 and ~5.0, respectively, in the H. B. Robinson and Turkey Point Cycle 1 bare fuel tests, which may never have been in solution. Much of this component of the total release reported in Table 7 most likely results from fine undissolved fuel particles.

H.B. ROBINSON FUEL IN J-13 WATER



TURKEY POINT FUEL IN J-13 WATER

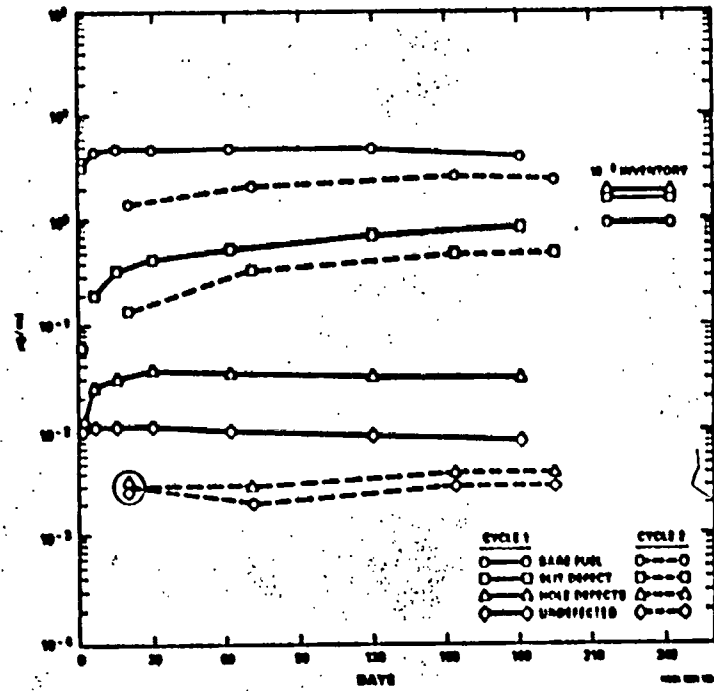


FIGURE 4. Uranium Concentration in Unfiltered Solution.

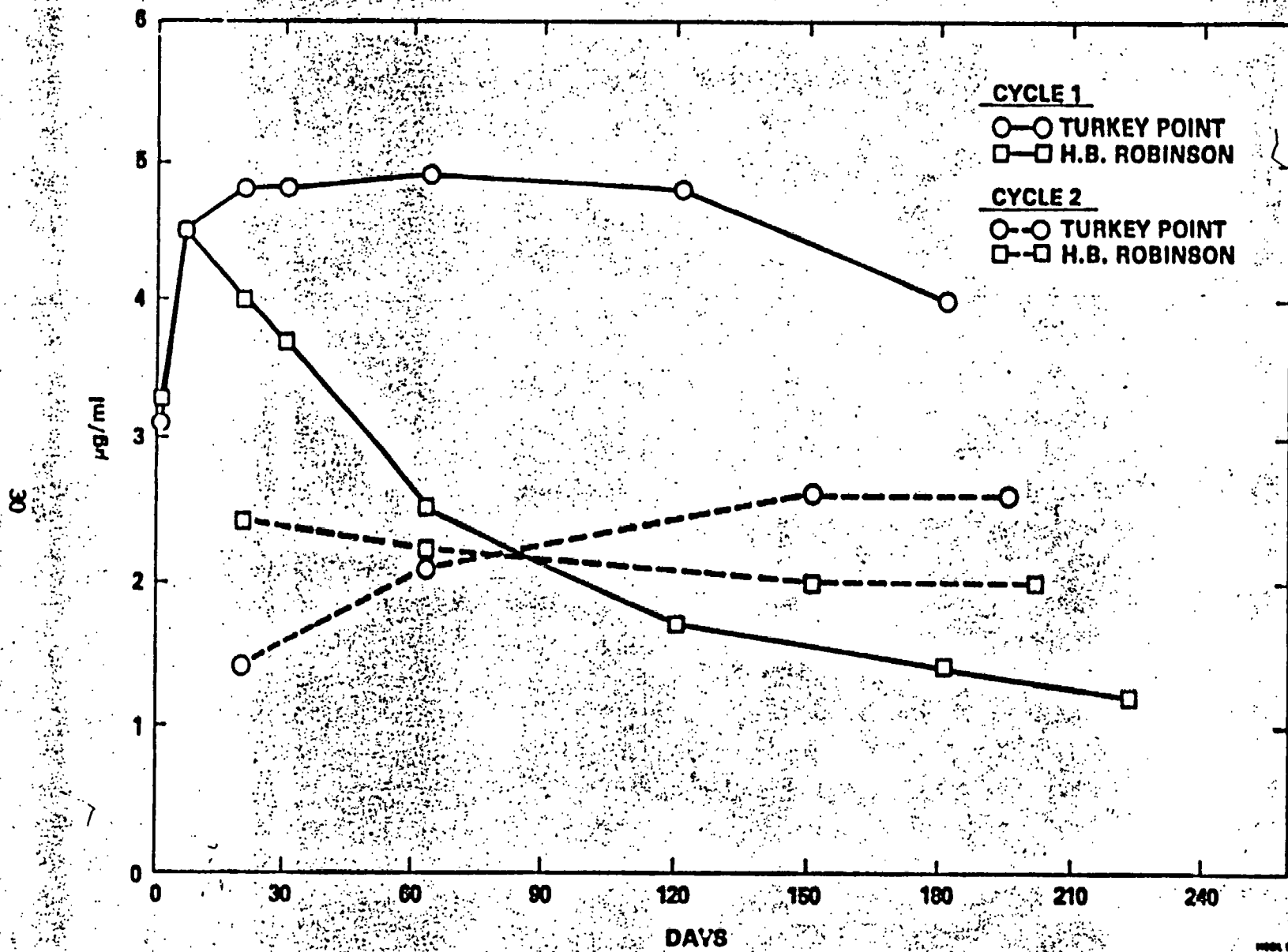


FIGURE 5. Uranium Concentrations for Turkey Point and H. B. Robinson Bare Fuels Tests in J-13 Water.

TABLE 7
URANIUM RELEASE DATA (ug)

	Bare Fuel		Slit Defect		Holes Defect		Undefected	
	HSR	TP	HSR	TP	HSR	TP	HSR	TP
<u>Cycle 1</u>								
Solution Samples	253	351	6.59	35.8	0.63	2.27	1.67	0.76
Final Solution	300	1000	23.80	212.5	1.50	7.25	3.25	2.00
[U (ppm)]	(1.2)	(4.0)	(0.09)	(0.85)	(0.005)	(0.029)	(0.013)	(0.008)
Rod Samples	36	15	0.31	0.54	<0.18	<0.19	<0.22	<0.10
Rinse	660	366	1.80	10.2	0.60	<0.60	<0.60	<0.6
Acid Strip	2700	960	1.50	15.9	0.60	2.70	0.60	<0.3
Total Release	3949	2692	34.00	274.9	3.51	<13.01	<6.34	<3.76
+ 10 ⁻⁵ Inv.	5.66	11.67	0.047	0.662	0.005	<0.030	<0.009	<0.009
% in Solution	14.00	50.19	89.38	90.32	60.86	73.17	77.60	--
<u>Cycle 2</u>								
Solution Samples	142	135	10.15	22.1	<0.13	0.22	<0.13	0.18
Final Solution	500	600	40	125.0	0.75	1.00	0.25	0.75
[U (ppm)]	(2.0)	(2.4)	(0.16)	(0.50)	(0.003)	(0.004)	(0.001)	(0.003)
Rod Samples	18	3	<0.08	<0.054	<0.06	<0.05	<0.06	<0.05
Rinse	102	39	1.20	3.6	0.60	<0.60	0.60	<0.6
Acid Strip	300	156	1.20	0.3	4.50	0.30	--	<0.3
Total Release	1062	933	52.63	151.0	6.04	<2.17	<1.04	<1.88
+ 10 ⁻⁵ Inv.	1.54	4.13	0.073	0.363	0.008	<0.005	<0.0015	<0.004
% in Solution	60.45	78.78	95.29	97.42	14.57	56.22	36.54	--
<u>Summary Cycle 1 & 2</u>								
Total Release	5011	3625	86.63	425.9	9.55	<15.18	<7.38	<5.64
+ 10 ⁻⁵ Inv.	7.20	15.80	0.120	1.025	0.013	<0.035	<0.010	<0.015

3.2 PLUTONIUM

Plutonium isotopes account for ~45% of the gross ORIGEN-2 calculated⁽¹¹⁾ activity of spent fuel at 1000 years and ~90% of the gross activity at 10,000 years. The ORIGEN-2 calculated $^{239+240}\text{Pu}$ inventories are 744 $\mu\text{Ci/g}$ and 704 $\mu\text{Ci/g}$, respectively, for the H. B. Robinson and Turkey Point spent fuels tested.

As indicated in the Figure 6 semi-log plots, unfiltered Pu activity was much greater in the bare fuel tests than in the slit defect and hole defects tests for both fuel types. Plutonium activities were also generally greater in the slit defect tests than in the hole defects tests. Plutonium activity in the hole defects test was greater than the undefected test for the Turkey Point fuel, but not significantly different in the respective H. B. Robinson fuel tests. With the exception of the H. B. Robinson 62-day Cycle 2 bare fuel test sample and the Turkey Point slit defect test, Pu activities in solution samples were generally lower for Cycle 2 compared with Cycle 1.

The high $^{239+240}\text{Pu}$ activities measured in the initial unfiltered sample aliquots in the Cycle 1 bare fuel tests were most likely caused by fine undissolved fuel particles. After 30 days in the H. B. Robinson Cycle 1 bare fuel test, unfiltered $^{239+240}\text{Pu}$ activity settled to approximately 200 pCi/ml, later falling off to 112 pCi/ml at the end of the test cycle. After the initial 1-day sample from the Turkey Point bare fuel test, unfiltered $^{239+240}\text{Pu}$ stabilized at approximately 500 pCi/ml for the duration of Cycle 1.*

The hypothesis that the initially high unfiltered activities were caused by undissolved fuel particles is supported by data from the 6-day H. B. Robinson sample (the only initial sample with high unfiltered Pu activity to be

*The unfiltered alpha activity data points for $^{239+240}\text{Pu}$, ^{241}Am and ^{244}Cm from the Cycle 1 15-day sample from the Turkey Point bare fuel test were not plotted in Figures 6, 7, and 8 since these data appear quite high, indicating that this sample aliquot contained an undissolved fuel particle(s).

filtered), where $^{239+240}\text{Pu}$ activity peaked at 2130 pCi/ml in the unfiltered aliquot and was 178 pCi/ml in the 0.4- μm filtered aliquot. The 178 pCi/ml value is in the range in which the unfiltered $^{239+240}\text{Pu}$ activity settled, beginning with the 30-day sample in this test. The undissolved particle hypothesis is also supported by the ^{241}Am and ^{244}Cm data, which were measured from the same alpha counting "source disks" as the $^{239+240}\text{Pu}$ data, and exhibited similar behavior to the $^{239+240}\text{Pu}$ data. High unfiltered uranium values were not detected in the initial Cycle 1 bare fuel test samples since the laser fluorescence method used for uranium does not measure uranium in undissolved particles.

The approximate 500 pCi/ml $^{239+240}\text{Pu}$ activity measured in unfiltered and 0.4- μm filtered samples from the Cycle 1 Turkey Point bare fuel test corresponds to about 5 ppb Pu, which is much less than the 430 ppb EQ3/6 calculated Pu solubility in J-13 water reported by Kerrisk.⁽¹³⁾ The Pu activities measured in the bare fuel tests may actually be just the Pu released to solution when the UO_2 fuel matrix dissolves in a system controlled by uranium solubility rather than a Pu solubility limit. However, the fraction of uranium inventory in solution in these tests was significantly greater than the fraction of $^{239+240}\text{Pu}$ inventory in solution. A slow decrease in Pu activity in later samples (during Cycles 1 and 2 in the H. B. Robinson bare fuel test) also suggests that Pu may have been equilibrating with a lower chemical activity "plate out" phase in these tests. Measured $^{239+240}\text{Pu}$ activities were generally lower in the H. B. Robinson bare fuel test than in the Turkey Point bare fuel test. Differences in apparent Pu solubility with the two bare fuel types are not currently explained.

Except for the previously discussed initial Cycle 1 bare fuel test samples, most $^{239+240}\text{Pu}$ activity measured in unfiltered samples was also measured in 0.4- μm filtered samples. A lesser fraction of the unfiltered activity passed through the 18- μm filters. Solution activities measured in filtered solution samples from the bare fuel tests were summed and divided by the

^aCalculated from $^{238}\text{Pu} + ^{241}\text{Am}$ and $^{233+240}\text{Pu}$ activities measured on the same alpha source disks, see Section 3.3.

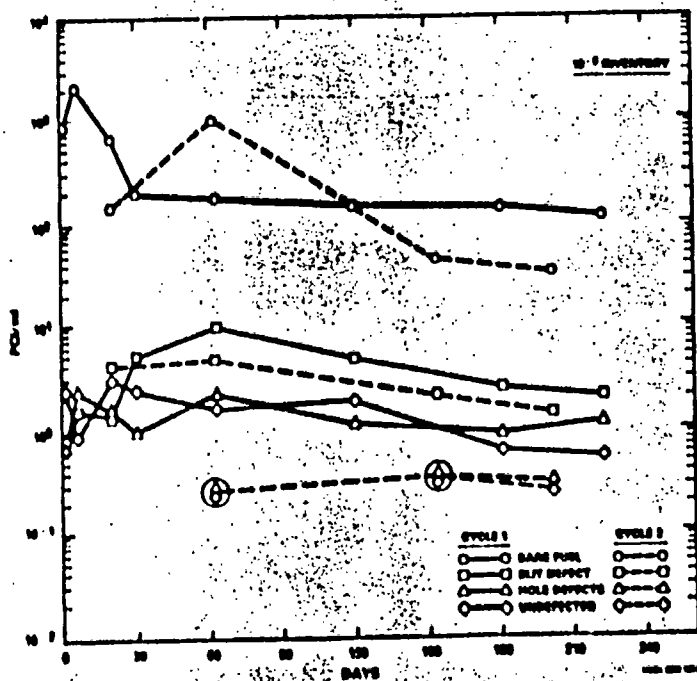
unfiltered sum for the same samples to calculate average activities passing each filter size. The 6-day filtered sample from the H. B. Robinson bare fuel test was not included since it was not representative of the other samples that exhibited relatively consistent filtering behavior. The results are:

<u>Fuel Test</u>	<u>Solution Sample</u>	<u>Cycle 1</u>	<u>Cycle 2</u>
H. B. Robinson	\int 0.4 μ m \int unfiltered	80%	80%
	\int 18 A \int unfiltered	30%	34%
Turkey Point	\int 0.4 μ m \int unfiltered	92%	95%
	\int 18 A \int unfiltered	42%	76%

Comparison of filtered Pu activities with unfiltered activities was somewhat erratic in the slit, holes, and undefected tests, since activities in these tests generally ranged from below the ~ 0.2 pCi/ml detection limit to 10 pCi/ml. Greater activities were reported for some filtered samples in these tests than for the unfiltered fraction of the same sample, which is presumably a result of counting statistics. Filtered sample data for Cycle 2 slit defect tests for both fuel types were relatively consistent and indicated that most Pu activity passed both the 0.4- μ m and 18-A filters at the few pCi/ml activity levels in these samples.

The Pu release accounting data tabulated in Table 8 indicates similar total measured fractional release for both bare fuel tests (8.4×10^{-5} for H. B. Robinson and 8.88×10^{-5} for Turkey Point). Total measured fractional release values for the slit and hole defects test were much less, ranging from 0.016×10^{-5} to 0.033×10^{-5} for these 4 tests. The percentage of measured Pu release in solution was generally lower than that for U. During Cycle 1, total measured Pu release from the H. B. Robinson hole defects specimen was actually less than from the undefected control. However, Pu release from this specimen did increase substantially during Cycle 2.

H.B. ROBINSON FUEL IN J-13 WATER



TURKEY POINT FUEL IN J-13 WATER

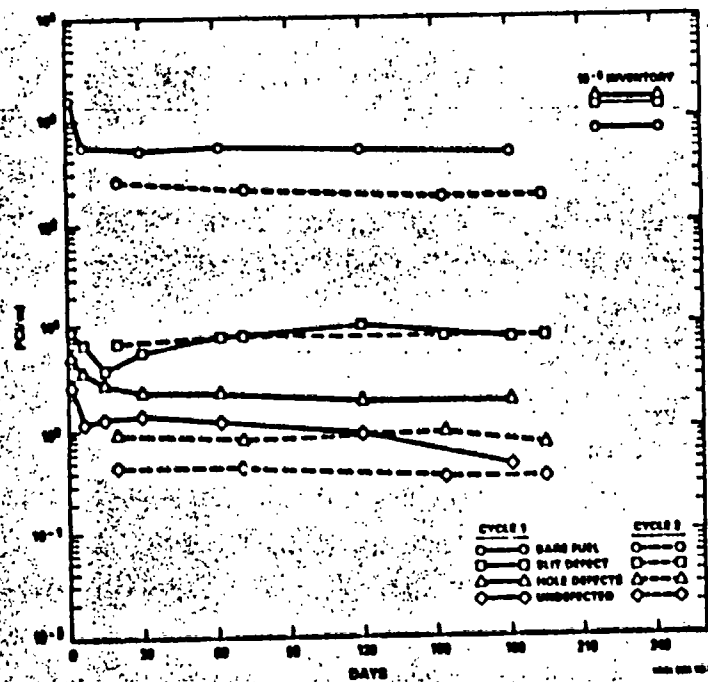
FIGURE 6. $^{239+240}\text{Pu}$ Activity in Unfiltered Solution.

TABLE 8
239+240Pu RELEASE DATA (nC1)

	Bare Fuel		Slit Defect		Holes Defect		Undefected	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
<u>Cycle 1</u>								
Solution Samples	41	62.7	0.454	0.556	0.140	0.171	0.154	0.092
Final Solution	28	114	0.495	1.800	0.405	0.450	0.135	0.113
[Pu (ppb)]	(1.2)	(4.9)	(0.021)	(0.077)	(0.017)	(0.019)	(0.006)	(0.005)
Rod Samples	59	21.6	0.332	0.621	<0.05	0.511	<0.02	0.029
Rinse	254	62.7	1.70	11.6	--	1.35	0.189	<0.27
Acid Strip	4054	1140	2.57	23.1	0.527	5.27	0.662	1.31
Total Release	4436	1401	5.551	37.68	1.122	7.75	1.16	1.54
+ 10 ⁻⁵ Inv.	7.18	7.31	0.0087	0.109	0.0018	0.0216	0.0019	<0.0052
% in Solution	1.56	12.61	17.10	6.25	48.57	8.01	24.91	~13
<u>Cycle 2</u>								
Solution Samples	23.8	13.4	0.223	0.480	0.014	0.058	0.014	0.027
Final Solution	8.3	45.0	0.350	1.910	0.079	0.180	0.068	0.090
[Pu (ppb)]	(0.36)	(1.9)	(0.015)	(0.082)	(0.003)	(0.008)	(0.003)	(0.004)
Rod Samples	173	5.4	0.059	0.151	0.014	0.032	0.009	0.021
Rinse	235	26.5	1.46	0.351	0.786	0.108	0.351	<0.054
Acid Strip	339	204.0	2.84	2.16	6.890	1.010	--	0.338
Total Release	779	294.3	4.932	5.052	7.783	1.388	0.442	<0.53
+ 10 ⁻⁵ Inv.	1.28	1.57	0.0077	0.0147	0.0124	0.0039	0.0007	<0.0015
% in Solution	4.12	19.84	11.62	47.31	1.20	17.15	18.55	~22
<u>Sum Cycle 1 & 2</u>								
Total Release	5215	1695.3	10.483	42.73	8.905	9.140	1.60	<2.34
+ 10 ⁻⁵ Inv.	8.46	8.88	0.0164	0.124	0.0142	0.0255	0.0026	<0.0066

3.3 AMERICIUM AND CURIUM

Americium-241 activity in spent fuel increases during the first ~100 years after reactor discharge as a result of ^{241}Pu decay. ^{241}Am accounts for approximately half of the gross activity of spent fuel at 1000 years. At 10,000 years, ^{241}Am with a 432-year half-life has decayed out and the only significant Am isotope remaining is ^{243}Am , which accounts for approximately 3% (including ^{239}Np daughter) of the total 10,000-year Ci activity. Curium-244 is a principal component of the actinide activity in the 10-year old fuel tests but, with its 18-year half-life, decays away before the end of the 300-year minimum containment period specified in 10 CFR 60. Curium is discussed with Am because of its similar behavior in the current tests.

The Cycle 1 ^{241}Am * and ^{244}Cm unfiltered sample activity plots (Figures 7 and 8) are quite similar to that of the $^{239+240}\text{Pu}$ data (Figure 6), all three of which were measured from the same alpha source disks. The peak observed in the 62-day Cycle 2 H. B. Robinson bare fuel test $^{239+240}\text{Pu}$ and ^{244}Cm data was not observed in the ^{241}Am data, which was measured on different sample aliquots in Cycle 2.

The bare fuel test ^{241}Am and ^{244}Cm data for 0.4- μm filtered and 18- μm filtered solution sample fractions were summed for each test cycle and divided by the sum of the unfiltered activities for the same samples. (Same calculation performed on the $^{239+240}\text{Pu}$ data omitting the 6-day H. B. Robinson sample for the same reason.) The results are:

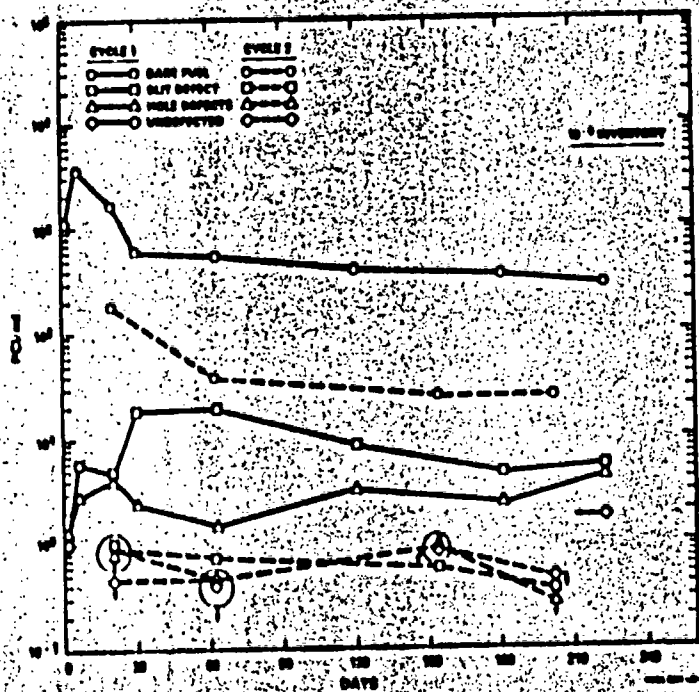
*Cycle 1 ^{241}Am data up to (but not including) the final solution samples were calculated from $^{241}\text{Am} + ^{239}\text{Pu}$ and $^{239+240}\text{Pu}$ data using $^{239}\text{Pu}/^{239+240}\text{Pu}$ ratios radiochemically measured for all except the undefected specimen tests. Therefore, only the final solution sample data points are plotted for the Cycle 1 undefected tests (shown with an arrow extending to the left).

<u>Bare Fuel Test</u>	<u>Solution Sample</u>	<u>Cycle 1</u>		<u>Cycle 2</u>	
		<u>²⁴¹Am</u>	<u>²⁴⁴Cm</u>	<u>²⁴¹Am</u>	<u>²⁴⁴Cm</u>
H. B. Robinson	<u>0.4 μm</u> unfiltered	75%	74%	58%	66%
	<u>18 A</u> unfiltered	3.2%	0.7%	6.0%	4.0%
Turkey Point	<u>0.4 μm</u> unfiltered	96%	93%	76%	79%
	<u>18 A</u> unfiltered	2.9%	12%	6.2%	4.1%

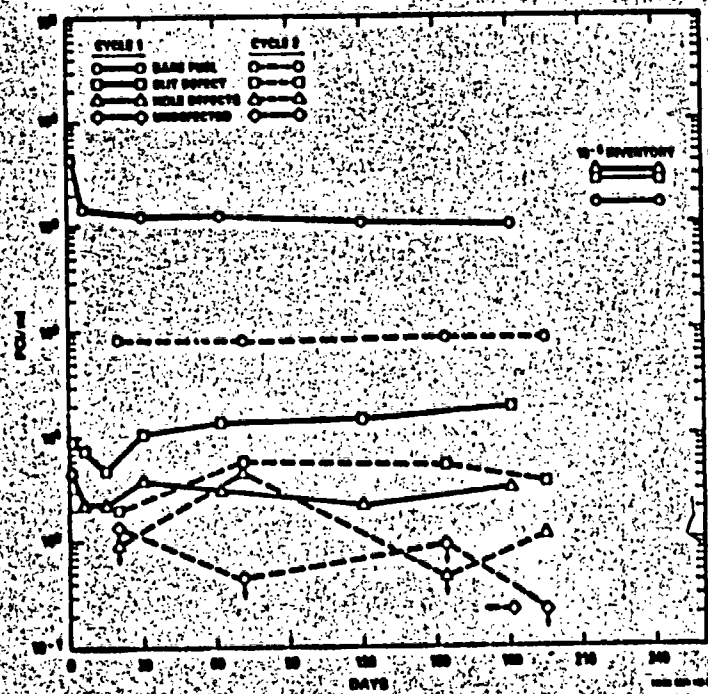
As with Pu, most of the Am and Cm passed through the 0.4-μm filters. The percentage of Am and Cm passing through the 18-A filters was substantially less than with Pu. The Am and Cm may have been associated as large complexes or colloids prohibiting it from passing the finer filter. However, Am concentration even in the bare fuel tests was on the order of 0.1 ppb and Cm concentration was on the order of a few ppt (parts per trillion). At such low concentrations, Am and Cm may have been removed by sorption of very small amounts of Am and Cm on the 18-A membrane filters.

As shown in Tables 9 and 10, total measured fractional releases and percentages in solution for ²⁴¹Am and ²⁴⁴Cm were similar for most of the test cycles. The two highest unfiltered ²⁴¹Am activities were the 15-day Cycle 1 Turkey Point bare fuel sample (7340 pCi/ml calculated, but not plotted in Figure 7 because of the questionably high value) and the 6-day H. B. Robinson bare fuel sample (3710 pCi/ml calculated). The respective Am concentrations for these activities would be ~2.6 ppb and ~1.4 ppb, which are close to the calculated 2.4 ppb (1.0×10^{-8} M) Am solubility in J-13 water reported by Kerrisk.⁽¹³⁾ However, these samples are thought to have contained undissolved fuel particles. Calculated 0.4-μm filtered ²⁴¹Am activity for most of Cycle 1 of the Turkey Point bare fuel tests was about 1000 pCi/ml, corresponding to an Am concentration of 0.35 ppb concentration, which is about the highest filtered sample Am concentration determined in the Series 2 tests. As with Pu, the greatest fraction of specimen ²⁴¹Am inventory measured in solution was in the Turkey Point bare fuel test.

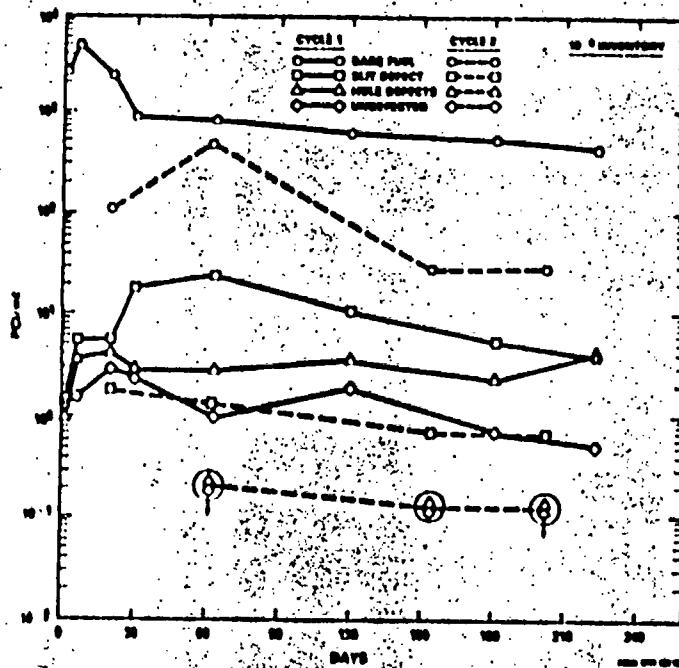
H.B. ROBINSON FUEL IN J-13 WATER



TURKEY POINT FUEL IN J-13 WATER

FIGURE 7. ^{241}Am Activity in Unfiltered Solution.

H.B. ROBINSON FUEL IN J-13 WATER



TURKEY POINT FUEL IN J-13 WATER

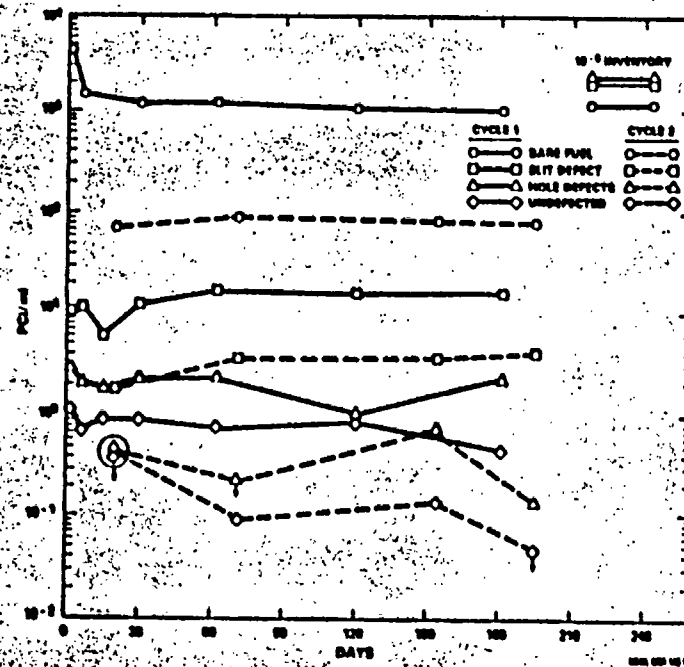
FIGURE 8. ²⁴⁴Cm Activity in Unfiltered Solution.

TABLE 9

 ^{241}Am RELEASE DATA (nCf)

	Bare Fuel		Slit Defect		Holes Defect		Undeformed	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
<u>Cycle 1</u>								
Solution Samples	91.9	138	1.07	0.87	0.22	0.22	--	--
Final Solution [Am (ppt)]	71.4 (105)	243 (348)	1.35 (2.0)	4.62 (6.62)	1.07 (1.57)	0.79 (1.13)	0.428 (0.61)	0.563 (0.81)
Rod Samples	132	42.1	0.73	1.18	0.05	0.98	--	--
Rinse	532	111	2.70	32.7	<0.54	4.05	1.05	0.541
Acid Strip	9595	2180	5.94	49.7	0.81	11.20	2.16	1.35
Total Release	10422	2714	11.79	89.07	<2.69	17.24	3.64	2.45
+ 10^{-5} Inv.	8.04	6.88	0.0088	0.126	<0.002	0.023	0.0029	0.0034
% in Solution	1.57	14.04	20.53	6.16	~48	5.86	--	--
<u>Cycle 2</u>								
Solution Samples	3.6	5.4	0.05	0.29	<0.05	<0.12	<0.040	<0.058
Final Solution [Am (ppt)]	6.2 (9.1)	21.1 (30.1)	0.09 (0.13)	0.90 (1.32)	<0.07 (<0.1)	0.28 (0.40)	0.113 (0.17)	<0.056 (<0.08)
Rod Samples	59.4	38.4	0.34	0.38	0.09	0.13	0.072	0.035
Rinse	139	50.5	<0.16	2.05	0.43	0.32	<0.16	0.243
Acid Strip	773	400	6.35	3.24	17.6	1.89	--	0.878
Total Release	981	515	6.99	6.86	18.2	2.74	<0.39	<1.27
+ 10^{-5} Inv.	0.77	1.33	0.0052	0.0097	0.014	0.0037	<0.0003	<0.0018
% in Solution	1.00	5.15	2.00	17.35	--	13.65	--	--
<u>Sum Cycle 1 & 2</u>								
Total Release	11403	3229	18.78	95.93	20.9	19.98	4.03	<3.72
+ 10^{-5} Inv.	8.81	8.21	0.014	0.136	0.016	0.027	0.0032	<0.005

TABLE 10
 ^{244}Cm RELEASE DATA (nCi)

	Bare Fuel		Slit Defect		Holes Defect		Undefected	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
<u>Cycle 1</u>								
Solution Samples	125	138	1.23	1.02	0.26	0.14	0.141	0.064
Final Solution [Cm (ppt)]	102 (3.5)	250 (8.6)	0.98 (0.05)	3.94 (0.20)	1.00 (0.05)	0.56 (0.03)	0.158 (0.008)	0.113 (0.006)
Rod Samples	145	38.3	0.81	1.14	0.02	0.88	0.023	0.044
Rinse	565	104	2.68	24.6	--	3.25	0.459	0.270
Acid Strip	8973	1610	5.81	36.9	0.81	9.05	1.89	1.76
Total Release + 10^{-5} Inv.	9910 8.64	2140 7.79	11.51 0.0097	67.60 0.137	2.09 0.0018	13.88 0.027	2.67 0.0024	2.25 0.0045
% in Solution	2.29	18.13	19.23	7.34	60.29	5.05	11.20	7.87
<u>Cycle 2</u>								
Solution Samples	11.8	5.4	0.08	0.20	<0.008	0.031	0.008	<0.014
Final Solution [Cm (ppt)]	7.0 (0.24)	20.3 (0.97)	0.17 (0.009)	1.01 (0.05)	0.034 (0.002)	0.034 (0.002)	<0.034 (<0.002)	<0.011 (<0.0006)
Rod Samples	388	34.4	0.25	0.37	0.049	0.067	0.049	0.041
Rinse	678	36.8	4.32	0.40	2.41	0.135	1.27	<0.054
Acid Strip	732	285	6.62	2.97	17.30	1.30	--	0.595
Total Release + 10^{-5} Inv.	1817 1.61	382 1.42	11.44 0.0097	4.95 0.010	19.80 0.0170	1.567 0.003	1.36 0.0012	<0.715 <0.0014
% in Solution	1.04	6.73	2.19	24.44	0.19	3.35	--	--
<u>Sum Cycle 1 & 2</u>								
Total Release + 10^{-5} Inv.	11727 10.25	2522 9.21	22.95 0.0194	72.55 0.147	21.69 0.0188	15.45 0.030	4.03 0.0036	<2.96 <0.0059

3.4 NEPTUNIUM

The primary Np isotope of concern for repository storage of spent fuel is ^{237}Np . Activity of ^{237}Np (2.14×10^6 year half-life) increases with time for several thousand years as a product of ^{241}Am (432-years half-life) decay. At 1000 years and 10,000 years, ^{237}Np accounts for about 0.06% and 0.25%, respectively, of the total ORIGEN-calculated⁽¹¹⁾ Ci activity of PWR fuel. Although ^{237}Np is a small portion of the total activity for the first 10,000 years, its expected solubility in groundwater and long half-life give cause for concern relative to long-term repository release. ^{239}Np represents a greater fraction of the calculated total activity (~0.9% at 1000 years, 1.4% at 10,000 years). However, ^{239}Np with its short (~2 day) half-life is a transient state in the ^{243}Am to ^{239}Pu decay chain and is considered as part of the ^{243}Am activity in this report.

Release of Np from spent fuel would be expected to be congruent with U and the other actinides (Pu, Am and Cm). The bulk concentration of Np in the oxide fuel matrix in the 10-year old fuels tested is only ~0.03% by weight and should be soluble in the oxide matrix phase at this low concentration. As previously stated, ^{237}Np concentration increases as a result of ^{241}Am decay, which in turn increases for ~100 years as a result of ^{241}Pu decay (13-year half-life). However, since neither Am or Pu are expected to phase segregate from the initial oxide fuel matrix, ^{237}Np should remain uniformly distributed in the oxide fuel matrix until such time this matrix is disrupted by reaction with external components (i.e., oxidation).

Because of its low concentration in the fuel and long half-life, ^{237}Np activity was very low when analyzed. Only samples from the bare fuel tests provided sufficient ^{237}Np activity to allow an estimate of fractional release. Based on the measured and estimated release data for the bare fuel tests given in Table 11, and comparison with the Pu, Am, and Cm tabulated release data, Np does appear to be released congruently with the other actinides. The summed Cycles 1 and 2 total ^{237}Np releases for either bare fuel test corresponds to <28 pCi/gm.

TABLE 11
²³⁷Np RELEASE DATA (pCi)

	Bare Fuel		Slit Defect		Holes Defect		Undefected	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
<u>Cycle 1</u>								
Solution Samples	<25	55						
Final Solution	<112	112						
[Np (ppb)]	(<0.64)	(0.64)						
Rod Samples	<10	<						
Rinse	<270	<135						
Acid Strip	946	135						
Total Release	<1363	<437						
+ 10 ⁻⁵ Inv.	<7.0	<7.4						
% In Solution	--	--						
<u>Cycle 2</u>								
Solution Samples	20	<14						
Final Solution	90	90						
[Np (ppb)]	(0.5)	(0.5)						
Rod Samples	2	2						
Rinse	<135	<135						
Acid Strip	68	<68						
Total Release	<315	<309						
+ 10 ⁻⁵ Inv.	<1.6	<5.2						
% In Solution	--	--						
<u>Sum Cycle 1 & 2</u>								
Total Release	<1678	<746						
+ 10 ⁻⁵ Inv.	<8.6	<12.6						

²³⁷Np activity was generally below the detection limit in samples from the slit defect, hole defects and undefected tests.

3.5

CESIUM

^{137}Cs and ^{134}Cs account for ~20% of the total Ci activity and most of the gamma activity in the ~10-year old spent fuel tested. At 1000 years, the only significant Cs activity remaining is ^{135}Cs , which has a 3×10^6 year half-life and accounts for ~0.02% of total Ci activity at 1000 years and about 0.08% of the Ci activity at 10,000 years.

Fission product Cs is partially released from the fuel during irradiation and segregates to grain boundaries and to the fuel-cladding gap. "Gap inventory" Cs dissolves immediately when contacted by water. The very low ^{137}Cs activities measured in solution for the undefected tests relative to bare fuel and defected cladding tests plotted in Figure 9 indicate that the end fitting seals did not leak in the undefected tests, and that release from these two tests can be assumed to be from exterior cladding contamination. Most of the measured ^{137}Cs release occurred at the beginning of Cycle 1 and was retained in solution. As indicated in Table 12, ~98% to 99% of the Cs in the slit defect and hole defects tests was in solution. The slightly lower percentages indicated in solution in the bare fuel tests were likely caused by undissolved fuel particles contributing to the ^{137}Cs activity measured in the rinse and acid strip samples.

Cesium release in the Turkey Point bare fuel tests was ~0.3% of specimen inventory, which is the approximate fractional fission gas release value reported⁽¹⁰⁾ for this fuel. Fractional ^{137}Cs release in the Turkey Point hole defects test (<0.01%) was much lower than expected and not currently explained. (Perhaps the I-9-12 specimen had been contacted by water or alcohol during previous undocumented handling at BCL.) Fractional release for the H. B. Robinson bare fuel, slit, and hole defects specimens (~0.8%, 0.8%, and 0.4%, respectively) was greater than the reported 0.2% fission gas release value⁽⁹⁾ for ATM-101 and greater than measured with the Turkey Point fuel. The ~250% greater fractional ^{137}Cs release with H. B. Robinson fuel is attributed to its finer grain size (6 μm for

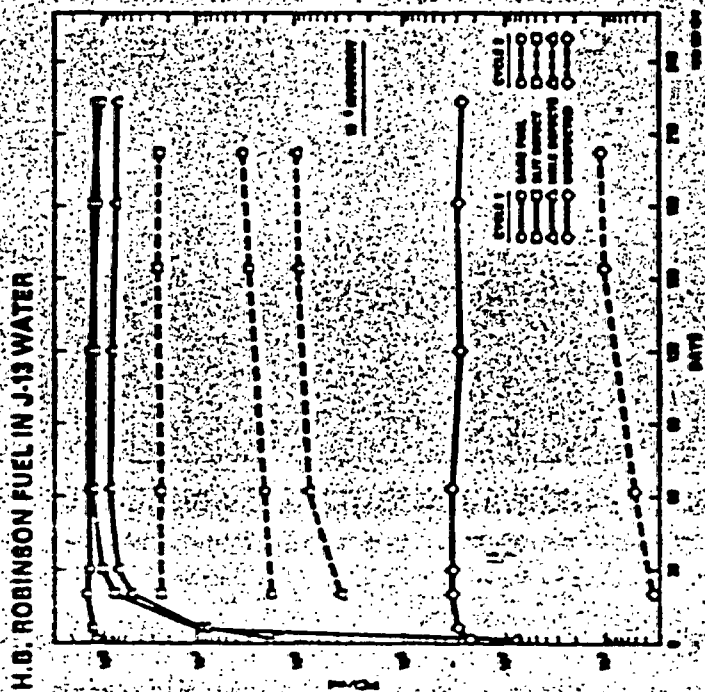
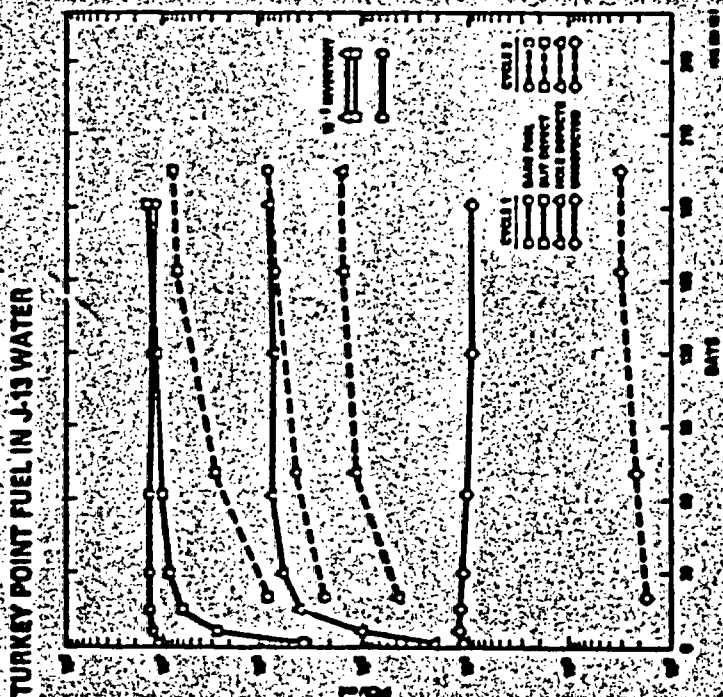


FIGURE 9. ¹³⁷Cs Activity in Unfiltered Solution.

H. B. Robinson versus 25 μm for Turkey Point). The greater grain boundary area and shorter diffusion distances to grain boundaries likely resulted in greater grain boundary ^{137}Cs inventories, and possibly greater fuel-cladding gap inventories as Cs was transported along grain boundaries to the gap during irradiation.

TABLE 12
 ^{137}Cs RELEASE DATA (μCi)

	Bare Fuel		Slit Defect		Holes Defect		Undeformed	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
<u>Cycle 1</u>								
Solution Samples	12600	1010	9650	644	6130	41.7	2.98	0.81
Final Solution	28200	3080	28000	3525	17500	195	6.80	2.33
[Cs (ppb)]	(3277)	(357)	(3248)	(409)	(2024)	(23)	(0.78)	(0.27)
Rod Samples	28	2.6	7	1.4	10	0.1	0.03	0.007
Rinse	1560	786	357	120	240	3.2	0.25	0.15
Acid Strip	612	138	28	9.6	36	1.5	0.12	0.08
Total Release	43000	5017	38042	4300	23916	241.5	10.2	3.38
+ 10^{-5} Inv.	776	308	664	144	425	7.76	0.19	0.11
% in Solution	94.88	81.52	98.97	96.95	98.80	98.01	95.88	92.90
<u>Cycle 2</u>								
Solution Samples	151	30	1630	249	44.5	7.0	0.04	0.015
Final Solution	840	204	6300	1960	241	39	0.28	0.08
[Cs (ppb)]	(98)	(24)	(731)	(227)	(28)	(4.5)	(0.32)	(0.009)
Rod Samples	2.0	0.3	1.0	0.3	0.1	0.02	<0.002	<0.002
Rinse	50.5	14.2	82.4	41.9	4.7	0.6	<0.05	0.05
Acid Strip	24.9	13.4	4.9	2.0	1.8	0.27	—	0.03
Total Release	1070	262	8018	2253	292	47	<0.37	0.18
+ 10^{-5} Inv.	19.6	16.1	140	75	5.2	1.51	<0.007	0.006
% in Solution	92.80	89.31	98.90	98.05	97.77	98.09	—	54.29
<u>Sum Cycle 1 & 2</u>								
Total Release	44070	5279	46060	6553	24208	288	10.6	3.9
+ 10^{-5} Inv.	796	324	804	219	430	9.27	0.20	0.12

3.6 TECHNETIUM

Technetium-99, with a 213,000-year half-life, accounts for 0.75% of the total Cf activity of spent fuel at 1000 years and ~3% at 10,000 years. (11) Release of ^{99}Tc from spent fuel is of particular interest since Tc compounds are predicted to be quite soluble in NNWSI groundwater. (13)

The radiochemical detection limit for ^{99}Tc was just adequate for analysis of 10^{-5} of specimen inventory in solution. Significant ^{99}Tc activities were reported for most solution samples analyzed during Cycles 1 and 2 of the bare fuel and slit defect tests. ^{99}Tc activities were generally below the detection limit in samples from the hole defects and undefected tests. ^{99}Tc activities measured in unfiltered solution samples are plotted in Figure 10. Essentially all of the ^{99}Tc activity measured in unfiltered samples was also measured in the 0.4- μm and 18- λ filtered samples, indicating that the ^{99}Tc activity was in true solution, most likely as TcO_4^- .

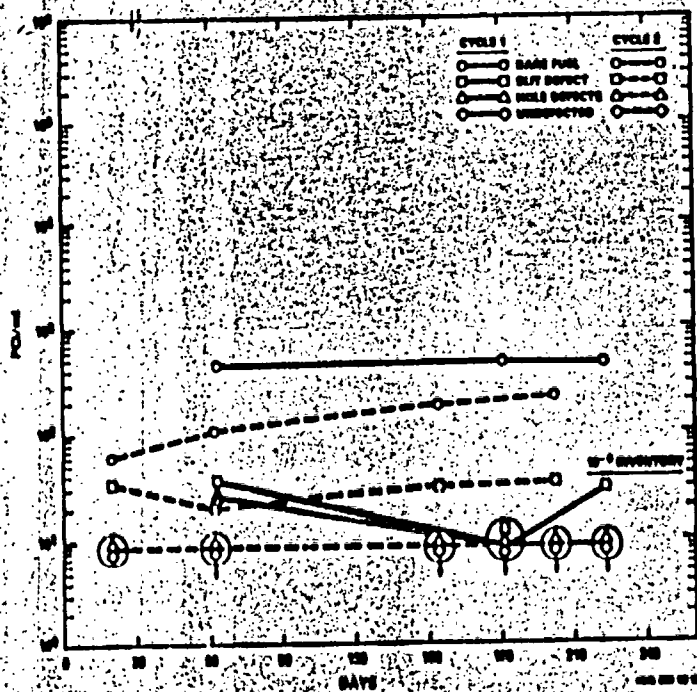
Since ^{99}Tc was not analyzed on all Cycle 1 solution samples, the "Solution Samples" release value given for the Cycle 1 bare fuel and slit defect tests in Table 13 is the total volume for all solution samples taken times an average activity for samples on which ^{99}Tc was measured. (^{99}Tc was measured on all unfiltered Cycle 2 solution samples.) Total measured fractional ^{99}Tc release (Cycles 1 and 2) is estimated at $\sim 32 \times 10^{-5}$ and $\sim 40 \times 10^{-5}$, respectively, for the H. B. Robinson and Turkey Point bare fuel tests versus ~ 8 to 10×10^{-5} for the actinides (Pu, Am, Cm, and Np) in the bare fuel tests, corresponding to a ^{99}Tc preferential release factor of 4 to 5 in these tests. A total fractional ^{99}Tc release of $\sim 20 \times 10^{-5}$ was estimated for the Turkey Point slit defect test (Cycles 1 and 2) versus $\sim 0.1 \times 10^{-5}$ for Pu, Am, and Cm, corresponding to a ^{99}Tc preferential release factor of about 150. Relative to uranium, Tc was preferentially released by a factor of ~ 20 in this test. Similar preferential Tc release relative to the actinides was observed in the H. B. Robinson slit defect test.

The preferential release observed for Tc may result from grain boundary leaching. Very high preferential Tc release (~12 times actinide release) was observed in the Series 1 bare fuel tests where visible grain boundary dissolution was observed in post-test fuel particle characterization.⁽⁴⁾ Dissolution of phase-separated Tc from open porosity is another possible source for preferential Tc release. The stable thermodynamic state for Tc should be the metallic state at the oxygen potentials that are present in LWR fuel during irradiation. Tc should show a tendency to phase separate with other metallic fission products when the temperature during irradiation is sufficiently high to allow diffusion and secondary phase formation. Fission product Mo (which also tends to separate into metallic phase during irradiation) was measured at slightly above detectable levels by inductively coupled plasma (ICP) emission spectrometry analysis of solution samples. Although the Mo ICP data are not precise enough for quantitative comparison, they do suggest preferential Mo release on the order of that observed for Tc in the Cycle 1 H. B. Robinson bare fuel test.

Preferential dissolution factors were previously calculated⁽¹⁴⁾ for ^{99}Tc in the two Cycle 1 bare fuel tests by comparing the fractional ^{99}Tc inventories known to have been in solution with the fractional inventory of uranium known to have been in solution. For uranium, the "known to have been in solution" value was calculated by summing the "Solution Samples" values (given in Table 7) and the maximum solution concentration times 250 ml. For ^{99}Tc , it was assumed that no precipitation from solution occurred and the "known to have been in solution" value was the sum of the "Solution Samples" and "Final Solution" values given in Table 13. Since similar fractional ^{99}Tc and U inventories were calculated for the sum of the "Rinse" and "Acid Strip" values, these values were assumed to be primarily composed of undissolved fine fuel particles. This evaluation resulted in Cycle 1 preferential ^{99}Tc dissolution factors of 8.7 for the H. B. Robinson bare fuel test and 3.8 for the Turkey Point bare fuel test. The greater preferential ^{99}Tc dissolution factor calculated for the H. B. Robinson bare fuel was attributed to the finer grain size of this fuel. Performing the above described calculation

for the Cycle 2 bare fuel tests gave ^{99}Tc preferential dissolution factors of 6.9 and 1.5, respectively, for the H. B. Robinson and Turkey Point bare fuel tests. These results are similar to the Cycle 1 results, indicating greater preferential ^{99}Tc release with the finer grained H. B. Robinson fuel. The ^{99}Tc preferential dissolution factors calculated for the H. B. Robinson and Turkey Point slit defect tests are 28.5 and 23.8, respectively, for Cycle 1 and 14.3 and 13.7, respectively, for Cycle 2. These values are not much different for the two fuel types tested in the slit defect tests where less fractional fuel dissolution occurred than in the bare fuel tests.

H.B. ROBINSON FUEL IN J-13 WATER



TURKEY POINT FUEL IN J-13 WATER

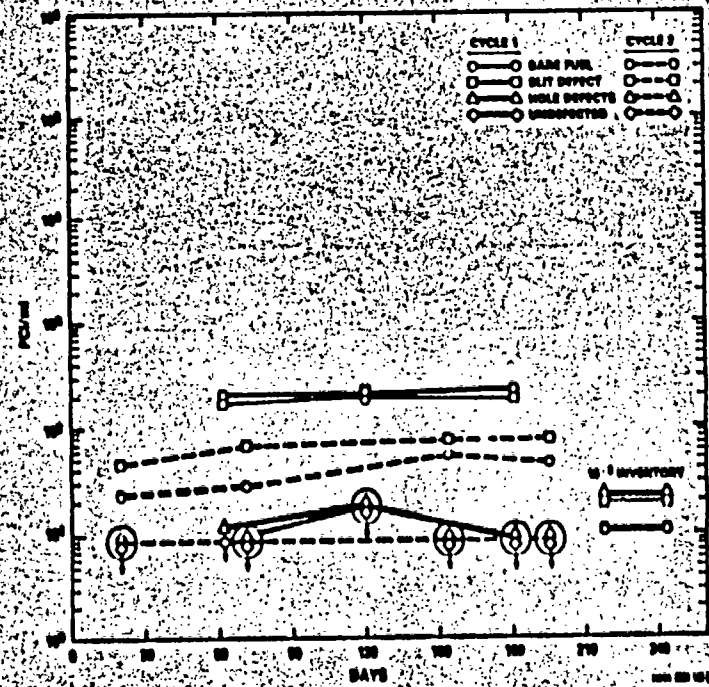


FIGURE 10. ⁹⁹Tc Activity in Unfiltered Solution.

TABLE 13
⁹⁹Tc RELEASE DATA (nCi)

	Bare Fuel		Slit Defect		Holes Defect		Undefected	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
<u>Cycle 1</u>								
Solution Samples	43(a)	15(b)	3.5(c)	13.9(d)				
Final Solution [⁹⁹ Tc (ppb)]	113 (26)	53 (12.4)	7.3 (1.7)	50.7 (11.9)				
Rod Samples	<0.3	--	<0.3	--				
Rinse	18.1	11.6	10.5	<5.4				
Acid Strip	28.4	6.1	3.5	<2.7				
Total Release + 10 ⁻⁵ Inv.	~203 ~23.3	~86 ~32	~25 ~2.8	~73 <15.3				
% in Solution	~77	~79	~43	--				
<u>Cycle 2</u>								
Solution Samples	8.0	2.4	1.86	4.2				
Final Solution [⁹⁹ Tc (ppb)]	56 (13)	11.3 (2.6)	8.45 (2.0)	19.1 (4.5)				
Rod Samples	<0.1	<0.1	<0.1	<0.1				
Rinse	<5.4	<5.4	<5.4	<5.4				
Acid Strip	4.0	<2.7	<2.7	<2.7				
Total Release + 10 ⁻⁵ Inv.	~74 ~8.6	~22 ~8.3	~18.5 ~2.1	~32 ~6.6				
% in Solution	~87	--	--	--				
<u>Sum Cycle 1 & 2</u>								
Total Release + 10 ⁻⁵ Inv.	~277 ~32	~108 ~41	~44 ~4.9	~105 ~22				

⁹⁹Tc activity was generally below the detection limit in samples from the holes defect and undefected tests.

- (a) Assumes 0.45 nCi/ml in all solution samples (approximate amount measured in 63,181 and 223-day samples).
 (b) Assumes 0.2 nCi/ml in all solution samples.
 (c) Assumes 36.5 pCi/ml in all solution samples (63-day value).
 (d) Assumes 185 pCi/ml in all solution samples (62-day value).

3.7 IODINE

Iodine-129 (half-life 17,000,000 years) is the lowest activity radionuclide measured in the present study (~ 0.025 $\mu\text{Ci/g}$ oxide fuel) and required neutron activation analysis for detection. Although ^{129}I has a relatively low activity, it may be relatively mobile in either the vapor phase or in solution and may become incorporated into the biosphere.

Activities of ^{129}I measured in Cycle 1 and 2 test samples are given in pCi/ml in Table 14. The activity in solution is 10^{-5} of the ^{129}I inventory in a H. B. Robinson specimen dissolved in the 250 ml of test solution would be ~ 0.09 pCi/ml. All of the solution samples analyzed for ^{129}I taken from bare fuel tests, the Turkey Point slit defect test, and Cycle 1 of the H. B. Robinson slit defect test exceeded 0.09 pCi/ml. ^{129}I was not measured on the Cycle 1 vessel strip solution because it was thought that the iodine would be lost as I_2 in the HNO_3 stripping procedure. However, the quantity of ^{129}I found in the Cycle 2 acid strip samples indicates that some of the ^{129}I activity does "plate out" of solution and is retained in the HNO_3 strip solution. The measured ^{129}I activities are considered to be a lower limit for these tests since the actual state of iodine in the test solutions is not known and some portion of the released ^{129}I may be lost from the unsealed test vessels as a volatile species (possibly I_2). The higher relative ^{129}I activity measured in an initial solution sample from H. B. Robinson bare fuel, which was tested at 25°C in sealed vessels in the Series 3 tests, further suggests that some ^{129}I may have been lost and not measured in the presently reported Series 2 tests.

The total activities of ^{129}I contained in the different sample types on which ^{129}I was measured were calculated to estimate the minimum "measured" release. ^{129}I activity measured on a single periodic solution sample was assumed for all periodic solution samples taken during the test cycle. The final solution activity for Cycle 1 Turkey Point fuel tests was assumed for all Cycle 1 Turkey Point periodic solution samples since ^{129}I was not determined for any of these samples. The resulting release and fractional release data are given

in Table 15. The greatest "measured" fractional release was 41.2×10^{-5} of specimen inventory for the Turkey Point bare fuel test. Much less fractional release was observed in the holes defect and undefected tests than in the bare fuel and slit defect tests. Releases from the holes defected specimens are not significantly greater than that from the undefected specimens. This suggests that much of the measured release was from cladding crud and/or residual cladding contamination in the holes defect tests and not from the fuel.

TABLE 14
 ^{129}I ACTIVITIES MEASURED IN SAMPLES (pCi/ml)

Fuel*	Cycle	Days	Sample Type	Bare Fuel	Slit	Holes	Undefected
HBR	1	63	Solution	0.523	0.292	0.00191	0.0060
HBR	1	223	Solution	0.718	0.377	0.00617	0.00541
HBR	2	154	Solution	0.432	0.0357	0.00471	0.0072
HBR	2	202	Solution	0.466	0.0473	0.00517	0.00893
HBR	2	202	Rinse	0.0141	---	---	---
HBR	2	202	Strip	0.0298	0.00485	0.00179	---
TP	1	181	Solution	0.590	0.289	0.0260	0.0076
TP	2	154	Solution	0.186	0.169	0.00533	0.0061
TP	2	195	Solution	0.219	0.217	0.00658	0.0117
TP	2	195	Rinse	0.0073	---	---	---
TP	2	195	Strip	0.0191	0.010	0.00136	0.0017

*HBR = H. B. Robinson, TP = Turkey Point.

TABLE 15
¹²⁹I RELEASE DATA (pCi)

	Bare Fuel		Slit Defect		Holes Defect		Undefected	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
<u>Cycle 1</u>								
Solution Samples*	50	44	27.7	21.7	0.18	1.95	0.57	0.57
Final Solution [I (ppb)]	180 (5.8)	148 (4.8)	94.3 (3.0)	72.3 (2.3)	1.54 (0.05)	6.50 (0.21)	1.35 (0.04)	1.90 (0.06)
Total Release + 10 ⁻⁵ Inv.	230 10.5	192 29.2	122 5.4	94 7.9	1.72 0.077	8.45 0.69	1.92 0.088	2.47 0.20
<u>Cycle 2</u>								
Solution Samples*	28	12	2.3	11.0	0.31	0.35	0.47	0.40
Final Solution [I (ppb)]	117 (3.8)	55 (1.8)	11.8 (0.4)	54.0 (1.8)	1.29 (0.04)	1.65 (0.05)	2.23 (0.07)	2.93 (0.09)
Rinse	8.5	4.4	--	--	--	--	--	--
Acid Strip	8.9	5.7	1.45	3.0	0.54	0.41	--	0.51
Total Release + 10 ⁻⁵ Inv.	162 7.5	77 12.0	15.6 0.69	68.0 5.7	2.14 0.096	2.41 0.20	2.70 0.12	3.84 0.32
% in Solution	89	87	90	96	75	83	--	--
<u>Sum Cycle 1 & 2</u>								
Total Release + 10 ⁻⁵ Inv.	392 18	269 41.2	138 6.1	162 13.6	3.86 0.17	10.86 0.88	4.62 0.21	6.31 0.52

*Final solution activities assumed for Cycle 1 Turkey Point periodic solution samples.

3.8 STRONTIUM

⁹⁰Sr accounts for ~14% of the total Ci activity of the ~10-year old fuel tested and is a pure beta emitter. Because of its relatively short 28.1-year half-life, ⁹⁰Sr will have essentially decayed out before the end of minimum 300-year 10 CFR 60 containment period.

Radiochemical separation for and measurement of ⁹⁰Sr was not started until the 154-day sample of Cycle 2. Cycle 2 ⁹⁰Sr release data were calculated and are given in Table 16. The ⁹⁰Sr activity measured in the 154-day sample was assumed for prior Cycle 2 samples in calculating the "solution samples" release value. Cycle 2 "total release" values indicate that ⁹⁰Sr was released preferentially relative to the actinides. The greatest preferential release was in the H. B. Robinson slit defect test where, during Cycle 2, ⁹⁰Sr fractional release was ~370 times the measured uranium fractional release and ~3000 times that measured for plutonium. Since, like Cs, Sr is relatively soluble, it is likely that even greater preferential release of ⁹⁰Sr occurred during Cycle 1, when most of the "free" Sr would have been dissolved.

TABLE 16
⁹⁰Sr CYCLE 2 RELEASE DATA

Cycle 2	Bare Fuel		Slit Defect		Holes Defect		Undeformed	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Solution Samples*	155	22.2	195	8.52	0.46	0.08	0.01	0.01
Final Solution [Sr (ppb)]	630 (32.6)	89.0 (4.6)	800 (41.4)	75.50 (3.9)	2.83 (0.146)	0.19 (0.010)	0.50 (0.026)	0.25 (0.013)
Rod Samples	1	0.3	0.4	0.03	<0.01	<0.01		
Rinse	71	5.4	13.0	4.60	<0.46	0.43		
Acid Strip	39	15.7	1.2	0.35	0.66	<0.23		
Total Release x 10 ⁻⁵ Inv.	887 24.8	132.6 12.2	1010 26.9	89.00 4.45	<4.41 <0.12	<0.94 <0.05		
% in Solution	88.5	83.9	98.6	94.4	>75	>30		

*Based on 154-day sample.

3.9 ACTINIDE AND FISSION PRODUCT RELEASE SUMMARY

The radionuclides discussed in the preceding sections are either actinides produced by progressive neutron activation of uranium and subsequent decay, or fission products. As such, these radionuclides are produced uniformly* throughout the UO_2 fuel matrix during irradiation. If all of these radionuclides remained fixed at their original sites of production in the fuel matrix, they would be released congruently as the fuel matrix dissolves. However, many of the fission products are only sparingly soluble in the oxide fuel matrix and tend to segregate into secondary phases during irradiation. The extent of secondary phase segregation increases with increasing temperature during irradiation. Relative radionuclide yields, and the variety of different elements with differing chemical properties represented, are shown in Figure 11. A summary of fractional release (total release divided by 10^{-5} inventory) values calculated in Tables 7 through 16 for the actinides and fission products is given in Table 17.

Within the probable accuracy of the current testing methods, the actinides Pu, Am, Cm, and Np appear to have been released congruently in all of the tests. Preferential uranium release was indicated relative to the other actinides in the Turkey Point bare fuel test and the two slit defect tests. An important factor probably contributing to these apparent preferential releases is the higher solubility of U relative to the other actinides in these tests. Much of the Pu, Am, Cm and Np released as the oxide fuel matrix dissolved, either did not dissolve, or may have precipitated back onto the fuel surface or within the defected cladding specimens, and was not measured. This hypothesis is supported by the "% in solution" values calculated for the actinides in Tables 7-10, which show a greater fraction of the measured uranium release to be in solution where it is readily measured. Greater apparent preferential U dissolution of the Turkey Point fuel relative to the H. B. Robinson fuel is attributed to much more extensive exposure of the Turkey Point fuel to air before testing. Dissolution of

*Disregarding minor radial distribution gradients arising from radial neutronic flux and spectral variations within the fuel.

UO_{2+x} from the fuel surface probably resulted in initial supersaturation of U relative to the bulk fuel matrix. Much of the initially dissolved U precipitated in the H. B. Robinson bare fuel test and may not have been recovered from the fuel in the rinse samples, possibly explaining the lack of measured preferential U release in this test. In the holes defect tested, measured actinide release was not much greater than in the undefected tests, so any preferential release from within the holes defect tested specimens may have been masked by release from residual cladding surface contamination. Also, actinide levels measured in the holes and undefected tests were near or often below detection levels, reducing the significance of conclusions that can be drawn from these data.

The fractional release data do not conclusively indicate the existence of any actinide phase segregation that would cause non-congruent actinide release from fuel as it dissolves from its as-irradiated state. Nevertheless, degradation of the oxide fuel matrix phase by exposure to an oxidizing environment before contact of the fuel with water may lead to actinide segregation, and differences in actinide solubilities may affect transport by solution from the fuel surface.

All of the fission product radionuclides measured showed preferential release relative to actinides. The greatest preferential release was observed with Cs, in which the several tenths of a percent "gap inventory" was rapidly released at the beginning of Cycle 1. An additional lesser preferential Cs release occurred in Cycle 2, which may have been associated with grain boundary leaching and exposure of additional segregated phases as fuel dissolution progressed. Although based on limited data, ^{90}Sr appeared to show preferential release similar to that of Cs in Cycle 2 of the bare fuel tests and may have also shown a substantial early preferential release in Cycle 1 had it been measured. Although there is some uncertainty concerning the portion of released fission product iodine actually measured, quantities indicating preferential ^{129}I release were measured in both Cycles 1 and 2. Preferential ^{99}Tc release was confirmed in Cycles 1 and 2 of the bare fuel and slit defect tests and is thought to occur primarily as a result of grain boundary leaching and the dissolution from exposed phase-segregated metallic particles.

Of the fission products measured, ^{137}Cs , ^{134}Cs and ^{90}Sr will decay out during the containment period. ^{135}Cs , ^{129}I and ^{99}Tc will be present at nearly the same activities during the post-containment period. Fission product ^{79}Se was also analyzed for but not detected. If ^{79}Se were released congruently with the uranium and remained in solution, then ^{79}Se activity in the bare fuel test solution samples would have been just slightly less than its detection limit, suggesting that there may not have been a significant preferential release of ^{79}Se in these tests.

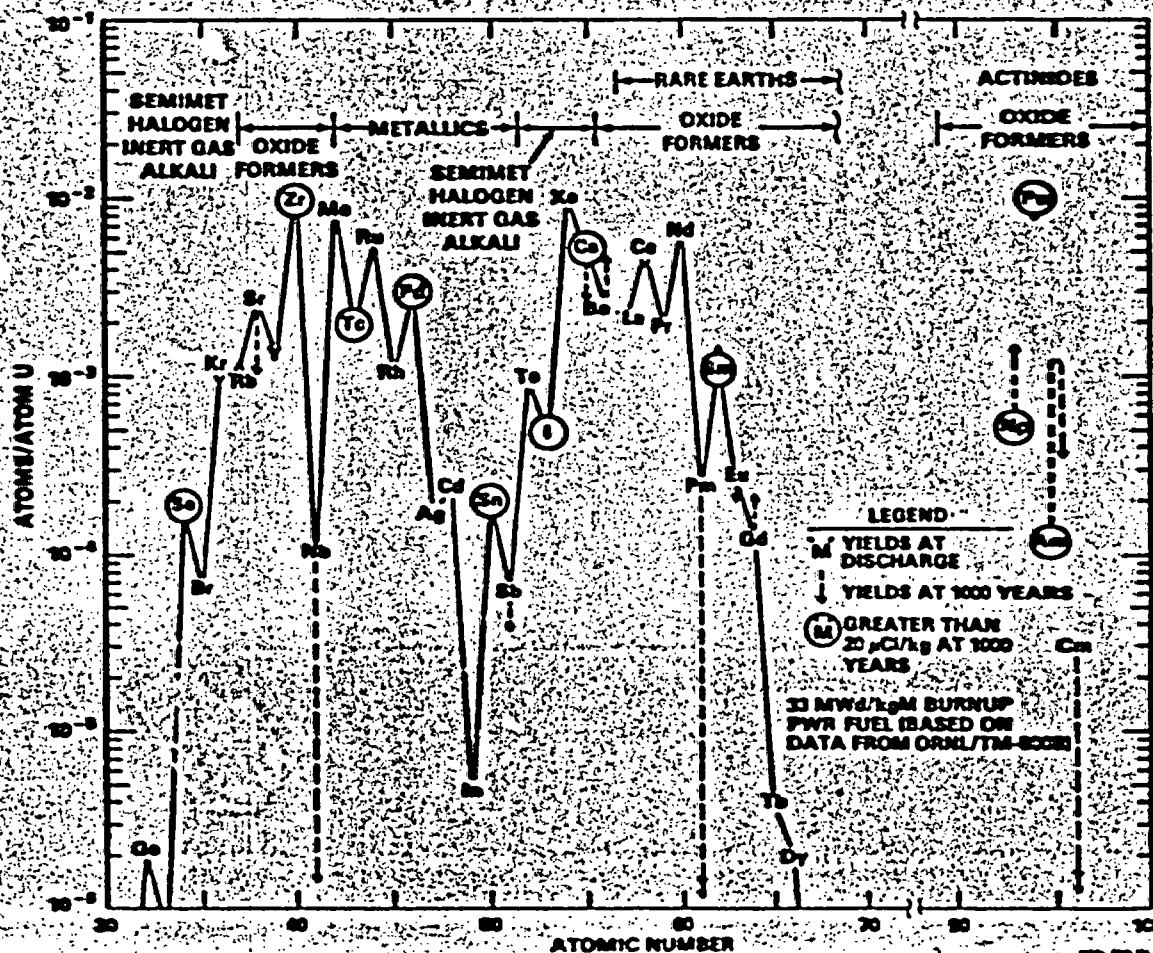


FIGURE 11. Elemental Yields for Actinides and Fission Products in Spent Fuel at Discharge and at 1000 years. Circled elements have isotopes with greater than $20 \mu\text{Ci/kg}$ activity at 1000 years.

TABLE 17

SUMMARY OF TOTAL MEASURED FRACTIONAL RELEASE
FOR ACTINIDES AND FISSION PRODUCTS*

Nuclide & Cycle	Bare Fuel		Slit Defect		Holes Defect		Undeformed	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Uranium								
Cycle 1	5.66	11.67	0.047	0.662	0.005	0.030	0.009	<0.009
Cycle 2	1.54	4.13	0.073	0.363	0.008	0.005	0.002	<0.006
Sum	7.20	15.80	0.120	1.025	0.013	0.035	0.010	<0.015
239+240Pu								
Cycle 1	7.18	7.31	0.009	0.109	0.002	0.022	0.002	<0.005
Cycle 2	1.28	1.57	0.008	0.015	0.012	0.004	0.001	<0.002
Sum	8.46	8.88	0.017	0.124	0.014	0.026	0.003	<0.007
241Am								
Cycle 1	8.04	6.88	0.009	0.126	<0.002	0.023	0.0029	0.003
Cycle 2	0.77	1.33	0.005	0.010	0.014	0.004	<0.0003	<0.002
Sum	8.81	8.21	0.014	0.136	0.016	0.027	0.0032	<0.005
244Cm								
Cycle 1	8.64	7.79	0.010	0.137	0.002	0.027	0.0024	0.0045
Cycle 2	1.61	1.42	0.010	0.010	0.017	0.003	0.0012	<0.0016
Sum	10.25	9.21	0.020	0.147	0.019	0.030	0.0036	<0.0059
237Np								
Cycle 1	<7.0	<7.4						
Cycle 2	<1.6	<5.2						
Sum	<8.6	<12.6						
137Cs								
Cycle 1	776	308	664	144	425	7.76	0.19	0.11
Cycle 2	19.6	16.1	140	75	5.2	1.51	<0.007	0.006
Sum	796	324	804	219	430	9.27	0.20	0.12
129I								
Cycle 1	10.5	29.2	5.4	7.9	0.077	0.69	0.09	0.20
Cycle 2	7.5	12.0	0.7	5.7	0.096	0.20	0.12	0.32
Sum	18	41.2	6.1	13.6	0.17	0.88	0.21	0.52
99Tc								
Cycle 1	~23	~32	~2.8	<15.3				
Cycle 2	<8.6	<8.3	<2.1	<6.6				
Sum	<32	<41	<4.9	<22				
90Sr								
Cycle 2	24.8	12.2	26.9	4.45	<0.12	<0.05		

*Units in parts per 100,000 of test specimen inventory; less than (<) symbols indicate totals containing >5% data reported as less than values, approximately (~) symbols indicate totals containing estimated values.

3.10

CARBON

Carbon-14 (half-life of 5730 years) is an activation product produced during irradiation by the (n,p) reaction on nitrogen impurities, and from the (n, α) reaction on ^{17}O . (15) ORIGEN calculations for spent fuel ^{14}C inventories depend on the assumed initial ^{14}N impurity levels in the fuel and cladding, which are not generally known and may vary between individual fuel samples. Carbon-14 was radiochemically measured separately on fuel and cladding for two samples from the H. B. Robinson C5 spent fuel rod (same rod used for Series 2 test specimens). The average of the analyses gave 0.53 $\mu Ci/g$ for the cladding and 0.49 $\mu Ci/g$ for the fuel. This quantity of ^{14}C corresponds to 530 Ci/1000 MTH. ^{14}C is of particular concern since it is mobile in the gas phase as CO_2 and in groundwater as HCO_3^- , and has a high potential for incorporation into the biosphere.

Activities of ^{14}C measured in test samples during Cycles 1 and 2 are given in Table 18. These activities are considered the lower limits of the actual ^{14}C activities, since ^{14}C in solution as HCO_3^-/CO_3^{2-} would be in equilibrium with CO_2 in the air over the test solution and the test vessels were capped (C52)

with loose fitting lids that may have allowed ^{14}C loss to the atmosphere. There was no correlation between ^{14}C release and defect severity in the H. B. Robinson Cycle 1 tests, suggesting that the ^{14}C may have been released by the cladding rather than the fuel. The H. B. Robinson Cycle 2 and Turkey Point fuel data in Table 18 suggest that part of the ^{14}C release in these test cycles did originate from the fuel. However, lower ^{14}C activity in the Turkey Point undefected test (Cycles 1 and 2) may be due to the fact that the 1-9-1 fuel section used for this specimen was the bottom end of the fuel rod and may have had less ^{14}C on its cladding surface.

Carbon-14 fractional release data were calculated for the H. B. Robinson fuel tests using the radiochemically measured fuel and cladding ^{14}C inventories and are given in Table 19. (Fractional release data for the Turkey Point fuel tests are omitted from Table 19 because ^{14}C inventory data were not obtained

HCO₃⁻

10

for this fuel.) Since ^{14}C was only measured on one periodic solution sample prior to cycle termination, this single value is assumed for all solution samples in calculating the "solution samples" values for Table 19. Approximately twice as much ^{14}C release was measured in the H. B. Robinson slit and holes defect tests as in the bare fuel and undefected tests.

The undefected specimen data suggest that a significant portion of the ^{14}C release originates from the cladding external surface. Activation of cladding nitrogen impurities and ^{17}O in the ZrO_2 passive layer on the cladding surface are possible sources for this ^{14}C . Another probable source is the reactor primary cooling water, which contains a relatively large ^{17}O inventory. ^{14}C deposited onto the cladding external surfaces from the primary coolant during irradiation may be the predominant source of ^{14}C released from the undefected fuel specimens, since dissolution of the cladding or the very insoluble Zircaloy or ZrO_2 surface layer would not be required for such release. As a component of deposited cladding "crud," ^{14}C inventory on the cladding surface may be quite variable.

TABLE 18
 ^{14}C ACTIVITIES MEASURED IN SAMPLES (pCi/ml)

Fuel*	Cycle	Days	Sample Type	Bare Fuel	Slit	Holes	Undefected
HBR	1	63	Solution	19.8	13.1	5.41	22.5
HBR	1	223	Solution	25.7	55.9	96.4	19.8
HBR	2	154	Solution	22.5	75.7	68.0	14.0
HBR	2	202	Solution	24.3	68.9	44.6	7.2
HBR	2	202	Rinse	5.41	3.6	4.9	9.9
TP	1	181	Solution	52.7	72.1	16.7	9.0
TP	2	154	Solution	54.1	34.7	32.9	7.7
TP	2	195	Solution	45.0	35.6	34.2	11.3
TP	2	195	Rinse	5.86	2.25	2.70	2.7

*HBR = H. B. Robinson, TP = Turkey Point.

TABLE 19
¹⁴C RELEASE DATA (nCi)
 FROM H. B. ROBINSON FUEL TESTS

	<u>Bare Fuel</u>	<u>Slit Defect</u>	<u>Holes Defect</u>	<u>Undeformed</u>
<u>Cycle 1</u>				
Solution Samples*	1.9	1.2	0.5	2.1
Final Solution [¹⁴ C (ppt)]	6.4 (5.8)	14.0 (12.5)	24.1 (21.6)	5.0 (5.0)
Total Release + 10 ⁻⁵ Inv.	8.3 17	15.2 30	24.6 49	7.1 15
<u>Cycle 2</u>				
Solution Samples*	1.5	4.9	4.4	0.9
Final Solution [¹⁴ C (ppb)]	6.1 (5.4)	17.2 (17.0)	11.2 (10.0)	1.8 (1.6)
Rinse	3.2	2.2	2.9	5.9
Total Release + 10 ⁻⁵ Inv.	10.8 21	24.3 48	18.5 37	8.6 18
<u>Sum Cycle 1 & 2</u>				
Total Release + 10 ⁻⁵ Inv.	19.1 38	39.5 78	43.1 86	15.7 32

*¹⁴C measured in 63-day Cycle 1 sample and 154-day Cycle 2 sample assumed for all periodic solution samples for those Cycles.

3.11 COBALT

Cobalt-60 is an activation product produced by neutron activation of ^{59}Co and (n,p) reaction on ^{60}Ni . With a 5.26-year half-life, ^{60}Co will decay out long before the end of the containment period. However, the relatively high gamma energy (1.3 MeV) associated with its decay requires heavy shielding for attenuation. Since ^{60}Co is an activation product, its inventory and distribution in young spent fuel may vary considerably depending on cobalt and nickel impurity levels in the fuel and cladding, the use of nickel containing alloys for in-reactor hardware, and upon the efficiency of ^{60}Co removal from the reactor coolant.

A tabulation of ^{60}Co activity actually measured in various samples is contained in Table 20. Much greater release of ^{60}Co occurred in the Turkey Point fuel tests than in the H. B. Robinson fuel tests. Also, greater release occurred from the defected specimens than from the undefected specimens, indicating the fuel, the gap, or the cladding inner surface as a significant source. Excess release from the defected specimens relative to undefected specimens was particularly notable with the Turkey Point fuel. The 287 μCi of ^{60}Co release measured in the Turkey Point bare fuel test is greater than the measured release for all other radionuclides except ^{137}Cs in these tests. This is interesting since "crud" layers plated out from the reactor coolant are usually thought of as primary sources of ^{60}Co activity. Considering the relative increase in ^{60}Co release going from undefected to holes to slit to bare fuel tests in comparison with the actinide release data, the data suggest that cobalt or nickel may have been a significant impurity in the as-fabricated Turkey Point fuel. Another possible source of excess ^{60}Co inventory in the Turkey Point fuel specimens may have been prior handling in contaminated hot cells at BCL where the specimens were originally prepared for pressurized stress rupture testing. Turkey Point fuel from the same assembly showed similar ^{60}Co release in the Series I tests conducted a year earlier. (4)

TABLE 20
⁶⁰Co RELEASE DATA (uCi)

	Bare Fuel		Slit Defect		Holes Defect		Undeformed	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP
<u>Cycle 1</u>								
Solution Samples	bd	34.7	bd	3.69	bd	0.45	0.013	0.07
Final Solution	bd	154	bd	11.7	bd	1.85	0.037	0.21
[⁶⁰ Co (ppt)]	--	(544)	--	(41)	--	(6.5)	(0.13)	(0.7)
Rod Samples	0.03	0.4	bd	0.04	bd	0.004	bd	0.004
Rinse	bd	47.6	bd	0.68	bd	0.09	bd	bd
Acid Strip	bd	5.0	bd	2.12	bd	0.29	bd	0.07
Total Release	--	242	--	18.2	--	2.68	0.05	0.35
% in Solution	--	78	--	34	--	86	100	79
<u>Cycle 2</u>								
Solution Samples	0.21	9.1	bd	0.40	bd	0.04	bd	0.003
Final Solution	1.16	32.9	bd	1.84	bd	0.12	bd	0.018
[⁶⁰ Co (ppt)]	(4.1)	(116)	--	(6.5)	--	(0.4)	--	(0.06)
Rod Samples	bd	0.6	bd	0.02	bd	0.001	bd	0.001
Rinse	bd	1.6	bd	0.14	bd	bd	bd	bd
Acid Strip	bd	1.1	bd	0.27	bd	0.04	bd	bd
Total Release	1.37	45	--	2.7	--	0.20	--	0.022
% in Solution	--	93	--	84	--	80	--	95
<u>Sum Cycle 1 & 2</u>								
Total Release	1.40	287	--	20.9	--	2.88	0.05	0.37

bd = below detection limit
 ppt = parts per 10⁻¹² weight

3.12 SOLUTION CHEMISTRY

The starting J-13 water and selected periodic solution samples from each test were analyzed by ICP emission spectrometry for metal cations and by ion chromatography (IC) for anions. All solution samples were measured for pH. Total and inorganic carbon were also measured on most samples receiving ICP and IC analyses. Inorganic carbon ($\mu\text{g/ml}$ carbon) was assumed to be 100% CO_3^{2-} , and equivalent CO_3^{2-} was calculated by multiplying the inorganic carbon value by 5 (i.e., to convert from 12 g atomic weight to 60 g mole weight for CO_3^{2-}). Complete solution chemistry data (pH, ICP, IC and CO_3^{2-}) for all Cycle 1 and 2 solution samples receiving ICP and IC analyses are given in Appendix B. ICP, IC and carbon analyses were performed on 0.4- μm filtered sample fractions only.

The solution chemistry data indicate that the overall chemistry of the J-13 water changed very little during any of the tests. The initial pH of the J-13 water barrel (fresh full barrel) was 7.2 when first sampled at HEDL just before Cycle 1 start. The chemistry shifted slightly basic to pH \sim 8.5 during Cycle 1. After using several gallons of J-13 water from the barrel, the water in the barrel also apparently shifted slightly basic because of equilibration with air, and the pH of the J-13 water at the start of Cycle 2 was \sim 8.0. The pH shifted to \sim 8.5 during the Cycle 2 tests (the same pH value reached during Cycle 1).

Silicon concentration in solution was of particular interest since the tests were run in fused silica vessels. Except for a 20% to 30% drop in Si in the 30-day and 120-day Cycle 1 H. B. Robinson samples, Si concentration remained relatively constant at 30 to 32 $\mu\text{g/ml}$ during the tests. Later SEM characterization of residue rinsed from fuel particle surfaces indicated that Si (most likely as a silica gel) deposited onto the fuel surfaces. Apparently, silica dissolved from the vessel at about the same rate that it deposited on the fuel particles, maintaining a constant (equilibrium?) silica concentration in solution.

Another minor solution change that occurred, apparently because of radiolysis, was conversion of some of the initial NO_3^- to NO_2^- . The NO_2^- values reported are best estimates since NO_2^- calibration standards were not used in the IC analyses. The source of NO_2^- is thought to be primarily NO_3^- radiolysis rather than dissolved air radiolysis, since the measured NO_3^- in solution usually decreased with increasing NO_2^- .

The initial ICP analyses through Cycle 1 of the H. B. Robinson tests included Al, B, Ca, Cr, Fe, Gd, K, Li, Mg, Mo, Na, Si, U and Zn. (These initial 14 elements are those analyzed on solution samples from spent fuel testing in support of the basalt repository.) Two sets of ICP analyses were run on the 223-day Cycle 1 H. B. Robinson samples. In the second set, Cr, Gd, Li, U, and Zn were replaced by Nd, Pd, Ru, Sr and Zr. Cr and Gd were deleted since they were always below detection limits and Li was deleted because it was usually reported as a "less than" value, but Li concentration was apparently at about its 0.016 $\mu\text{g}/\text{ml}$ detection limit. Uranium was deleted since the laser fluorescence method used on all samples was more sensitive. The Nd, Pd, Ru and Sr are major fission product elements that were not measured radiochemically. (Radiochemical measurement of ^{90}Sr was started later in the Cycle 2 tests). Zr was also of interest since the tests included the cladding.

The final Cycle 1 solution samples, and all Cycle 2 samples, used the second 14-element ICP analyses set. Nd, Pd, Ru and Zr measured in the later ICP analyses are not included in Appendix B since they were always below detection limits. Cr, Gd, and Li measured in the initial ICP analyses are also excluded from Appendix B for the same reason.

3.13 SOLIDS CHARACTERIZATION

Four types of solids characterization were performed: 1) SEM of small fractured fuel particles, 2) SEM examination with energy dispersive x-ray (EDX) microanalysis of filters used to filter solution samples, 3) SEM and EDX analysis of bare fuel rinse residues obtained at the end of test cycles, and 4) post-test radiometallurgical examination of polished sections from test specimens.

3.13.1 SEM Characterization of Fuel Particles

SEM photographs of the fracture surface on small particles of the Turkey Point and H. B. Robinson spent fuels are compared with the fracture surface of an unirradiated UO_2 fuel pellet in Figure 12. Unirradiated UO_2 is a hard ceramic material that exhibits transgranular cleavage when fractured. The irradiated fuel is friable and commonly fractures along grain boundaries, as shown in Figures 12B and 12C. Accumulation of fission products at the grain boundaries during irradiation is thought to cause the change in fracture properties. Areas of cleavage fracture and mixed cleavage and grain boundary fracture (see Figure 13) were observed in some spent fuel particles thought to be from near the outer radial regions in the fuel where irradiation temperature is lower and less fission product migration to grain boundaries occurs.

The non-homogeneous chemical microstructure resulting from fission product migration and secondary phase separation is thought to cause the incongruent dissolution behavior observed with spent fuel. The spent fuel dissolution data reported here suggest congruent release of actinides as the oxide fuel matrix dissolves and indicate preferential leaching of fission products Tc, Cs, Sr and I, and activation product ^{14}C . Fission products expected to have low solubility in the oxide fuel matrix are metallic fission products: Mo, Tc, Ru, Rh, Pd, Ag, Cd and Sn; inert gases: Kr and Xe; and others, including Se, Cs, Rb, Sr, Ba, Te and I. (See Figure 11.) Very small fission gas bubbles in the fuel matrix and along grain boundaries may provide nucleation sites for condensed fission product phases, which may coalesce and grow, depending on local irradiation temperature.

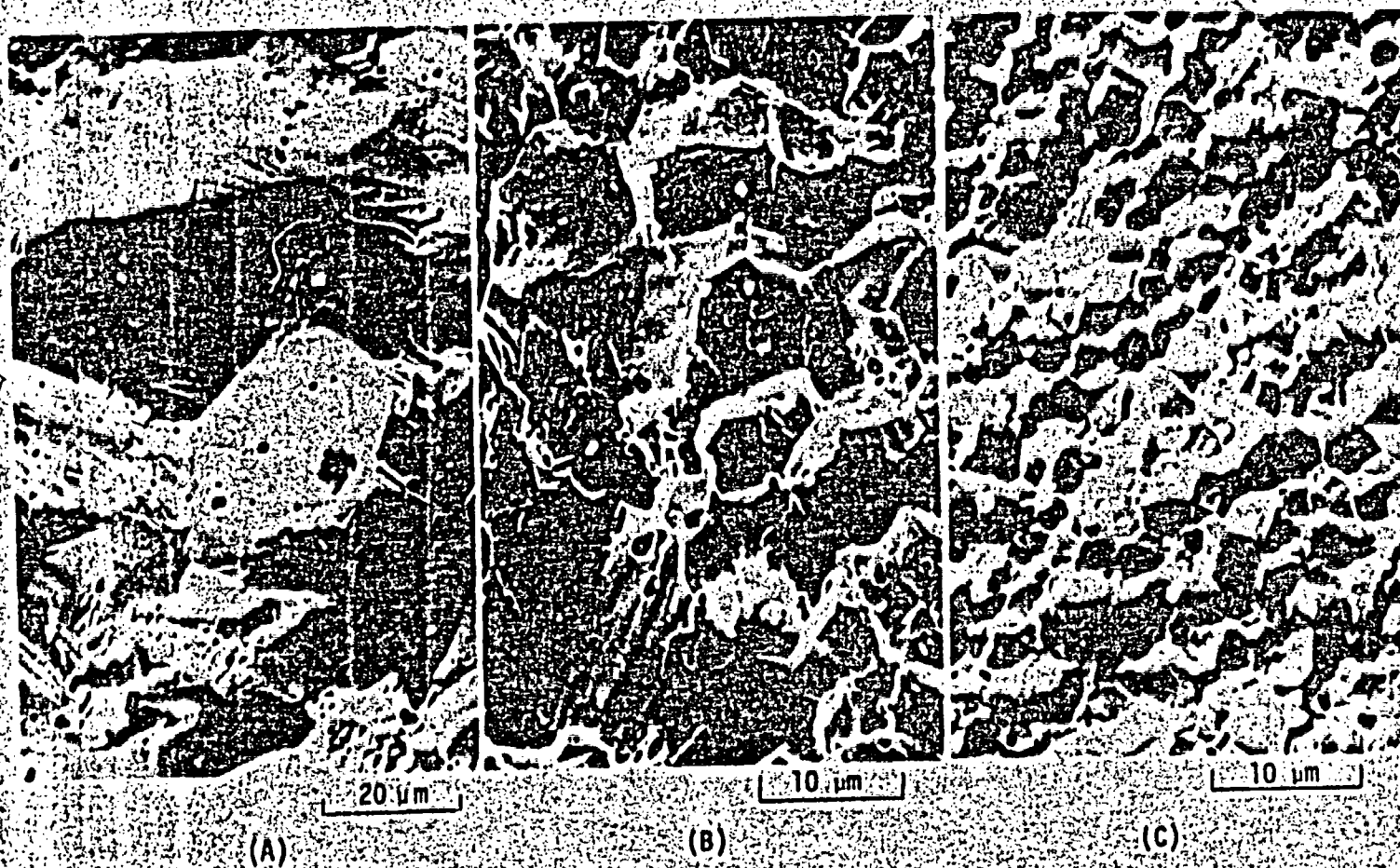


FIGURE 12. SEM Photographs of Fracture Surfaces of (A) Unirradiated UO_2 Fuel Pellet, (B) Turkey Point Fuel After Cycle 1 Testing, and (C) H.B. Robinson Fuel Before Testing.



FIGURE 13. SEM Photograph of Pre-test H. B. Robinson Spent Fuel Fracture Surface Showing Region of Mixed Cleavage and Grain Boundary Fracture.

SEM examination of post-test fuel particles did not reveal substantial alteration of fuel surface characteristics in dense regions exhibiting cleavage fracture as a result of testing. Several regions exhibiting open grain boundaries and possibly enhanced porosity as a result of testing were examined. However, more extensive examination of both pre-test and post-test fuel is needed to verify the extent of microstructural alteration that may have occurred as a result of testing.

3.13.2 Filters Examination

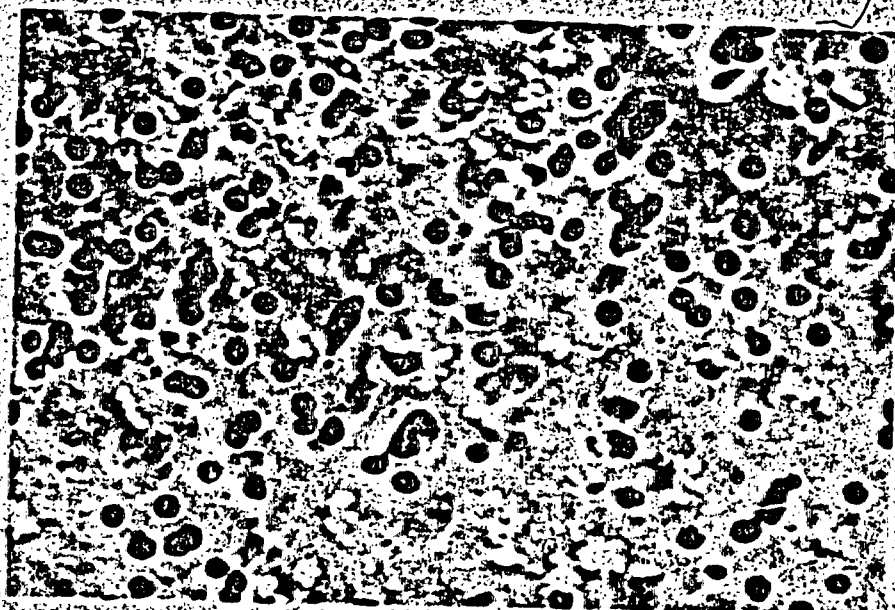
Selected filters used to filter periodic solution samples were examined by SEM. The 0.4- μm polycarbonate filter used on the 181-day Cycle 1 H. B. Robinson bare fuel test solution sample is shown in Figure 14A. EDX microanalysis of the small particle agglomerates visible on the filter showed only the element Si (lighter elements present, such as H, C and O, are not detectable by EDX).

SEM examination data on a portion of the 18-A membrane cone filter used on the same 181-day Cycle 1 H. B. Robinson bare fuel test solution sample are shown in Figure 15. The filter portion shown is near the bottom tip of the filter where most of the filtered material would collect as solution is centrifugally forced through the filter. The filter surface is nearly completely covered by the fine filtered floc. EDX analysis showed only the elements Si, Au and S. The low intensity of the Si K-line peak relative to the Au M-line peak ($\sim 200 \text{ \AA}$ of Au was sputtered onto the specimen surface) and the sulfur K-line peak originating from the underlying filter membrane, suggests that the dried silica floc has very low density.

3.13.3 Rinse Residue Examination

When rinsing the H. B. Robinson bare fuel particles in a 250-ml beaker at the end of Cycle 1, the rinse water became dark and turbid. The fuel particles were rinsed with just enough J-13 water in the beaker to cover the particles. The beaker was rocked back and forth allowing the particle to slide side-to-side in the water. The rinse procedure was repeated 10 times and the rinse water became slightly grey even after the tenth rinse. The collected rinse water was allowed to settle overnight, sampled, and then decanted from the settled residue in the bottom of the 1000-ml rinse water collection beaker. The dried particulate residue weight was 186 mg. The rough loose granular appearance of the H. B. Robinson bare fuel particle surface shown in Figure 14B suggests that many fuel grains may have been released from the surfaces of these particles.

When Cycle 1 of the Turkey Point bare fuel test was terminated, the rinse procedure was modified and the fuel was only rinsed five times in the 250-ml beaker. Residue weight from the Cycle 1 Turkey Point bare fuel rinse was only 12.3 mg. Lesser quantities of residue appeared to have settled from the bare fuel rinse solutions at the end of Cycle 2. Although rinse solution residue was visible at the end of Cycle 2, the quantity appeared to be much less than that from Cycle 1 and was not collected and weighed.



(A)

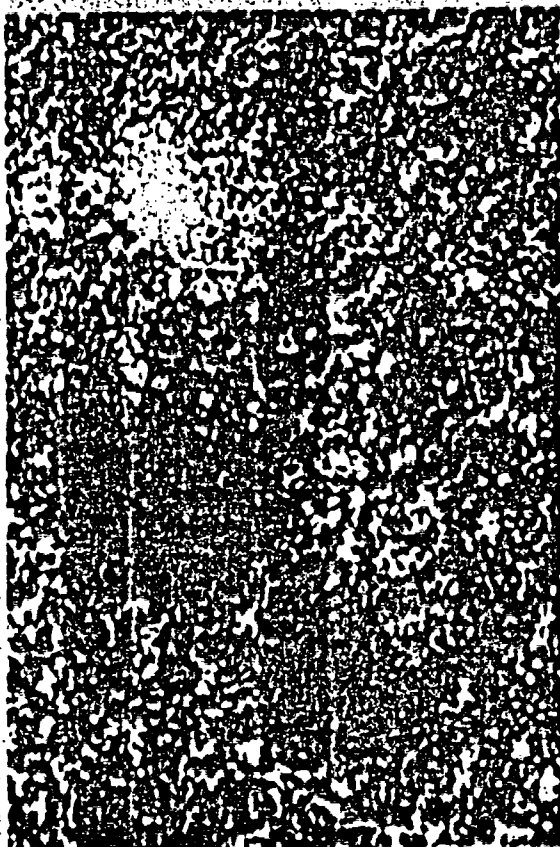
2 μm



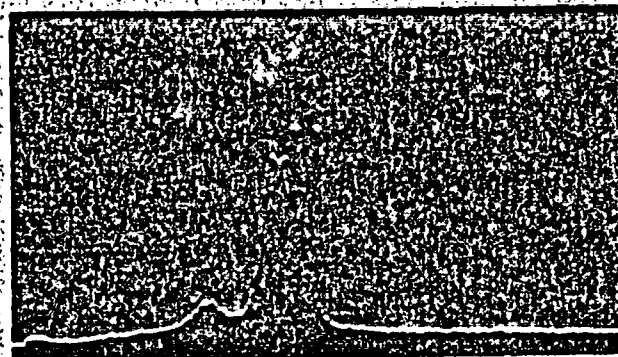
(B)

10 μm

FIGURE 14. (A) SEM Photograph of 0.4- μm Polycarbonate Filter Used to Filter 181-Day H. B. Robinson Cycle 1 Bare Fuel Test Solution Sample. EDX analysis showed only the element Si in the small particles visible on the filter (assumed to be colloidal silica flocs). (B) Silica Flocs on an H. B. Robinson Fuel Particle.



2 μ m



SI Au S

EDX SPECTRUM
SI - FROM SOLUTION SAMPLE
S - FROM FILTER
Au - GOLD COATED SPECIMEN

HEDL 8502 021.20

FIGURE 15: SEM Photograph of Bottom Inner Surface of 18-A Filter Cone used to Filter 181-Day Cycle 1 H; B; Robinson Bare Fuel Test Solution Sample. Energy dispersive x-ray (EDX) spectra shows only Si in the filtered material, plus gold from specimen coating and S from the filter. Neg 8500845-2

An SEM image of the larger particles in the Cycle 1 H. B. Robinson bare fuel test rinse residue is shown in Figure 16. The particle is a small fuel fragment that has been coated by a surface layer of material that cracked and partially spalled when dried. EDX analysis of the surface layer showed primarily the element Si. The layer appears to be a silica gel layer ranging from 10 μm to 25 μm in thickness. Similar layers were observed on several other fuel particles. Figure 17 shows flakes of the silica material with fine UO_2 particles adhering to the surfaces which had apparently interfaced with fuel. Similar appearing silica layers were also observed on fuel particles in the Cycle 1 Turkey Point bare fuel rinse residue. The quantity of Si involved in the deposited layers would appear to be a significant portion of (or greater than) the ~ 7.5 mg of Si contained in the 250-ml of J-13 water. Dissolution of the fused silica test vessel must have occurred during the test to maintain the ~ 30 $\mu\text{g/ml}$ Si in the test solution.

The effect of a silica gel coating on the leaching behavior of fuel is not known. Most previous ambient temperature spent fuel leaching has been conducted with bare fuel particles in fused silica vessels. Series 3 NWSI spent fuel leach testing will be conducted in stainless steel vessels. One vessel will be run at ambient hot cell temperature to provide a comparison with the Series 2 results.

3.13.4 Metallographic Examination

At the end of each test cycle, particles of bare fuel were removed from the test, metallographically mounted, polished, and optically examined. Particular attention was given to examination of the particle section edge for evidence of grain boundary dissolution. Significant localized grain boundary dissolution was observed in Series 1 Turkey Point bare fuel particles tested in deionized water. (4) However, no significant evidence of grain boundary dissolution was visible by optical metallographic examination of Series 2 H. B. Robinson or Turkey Point bare fuel particles at the ends of Cycles 1 or 2. Micrographs of bare fuel particle sections at the particle surface are shown in Figure 18. The probable explanation for the failure to find evidence of grain boundary dissolution in polished sections of Series 2 bare



2

100 μm

● SPOT 1 EDX SPECTRUM



Na Mg Si S Cl K

● SPOT 2 EDX SPECTRUM SHOWS ONLY URANIUM

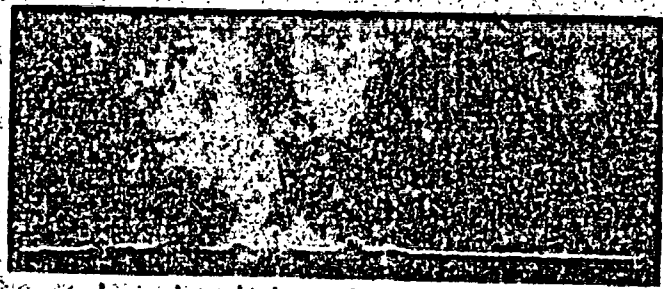
HEDL 8502-021.21

FIGURE 16. Fine Fuel Particle from H. B. Robinson Bare Fuel Test Showing Remnants of Silica Layer Deposited on the Particle Surface During the Test. Neg 8500845-3



50 μ m

● SPOT 1 EDX SPECTRUM



Na Si S Cl K Ca

● AREA 2 (5X)



3

● AREA 3 EDX SPECTRUM SHOWS ONLY URANIUM

HEDL 8502 021.23

FIGURE 17. Particles from Cycle 1 H. B. Robinson Bare Fuel Rinse Residue Showing an Apparent Silica Gel Flake on Edge and a Particle Coated with Very Fine UO_2 Grains. Neg. 8500845-5

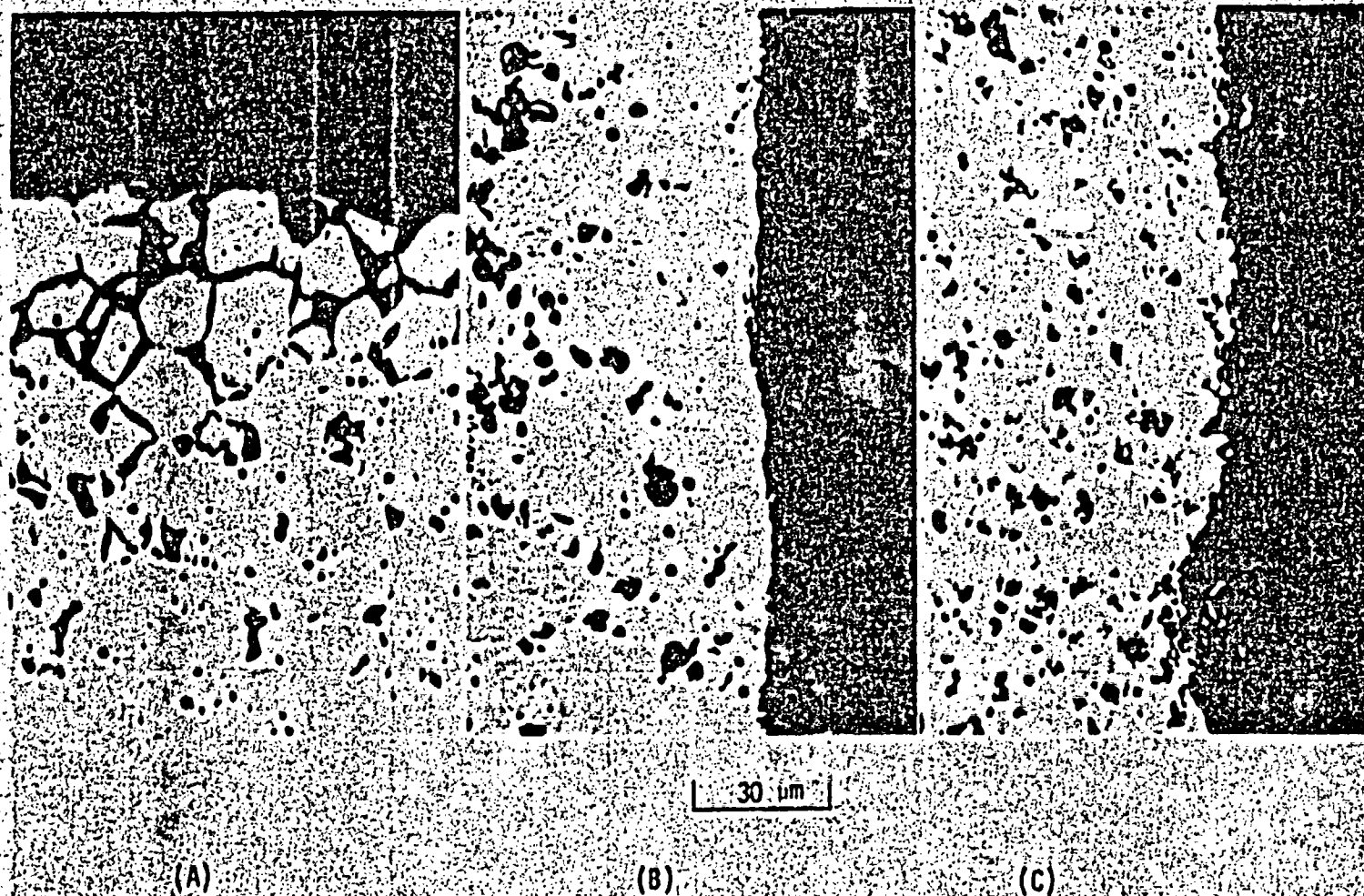


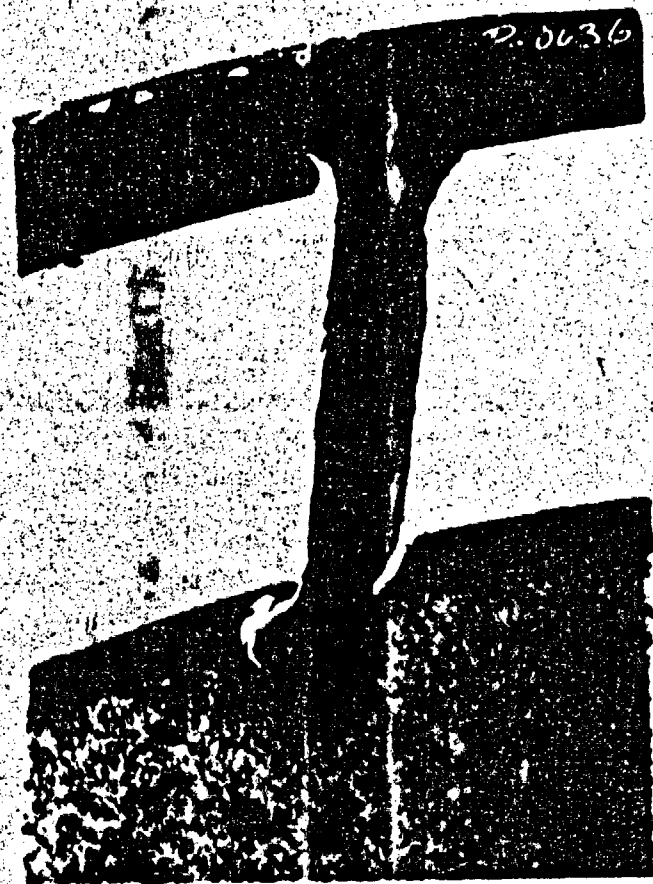
FIGURE 18. Polished Sections at Bare Fuel Particle Surfaces, (A) Turkey Point Fuel Tested for 250 days in Deionized Water, (4) (B) Turkey Point Fuel Tested 181 Days in J-13 Water, and (C) H. B. Robinson Fuel Tested 223 Days in J-13 Water. (All tested at ambient hot cell temperature; all micrographs taken at 750X.)

fuel particles may be that loose grains were removed from the fuel particle surface by the more aggressive rinsing procedure used in the Series 2 tests.

Micrographs showing polished sections through the H. B. Robinson and Turkey Point slit defect specimens are shown in Figure 19. Examination of the fuel at the fuel-cladding gap and along fuel cracks did not reveal any unusual appearing features that could be related to fuel dissolution in these sections from the slit defect tests.

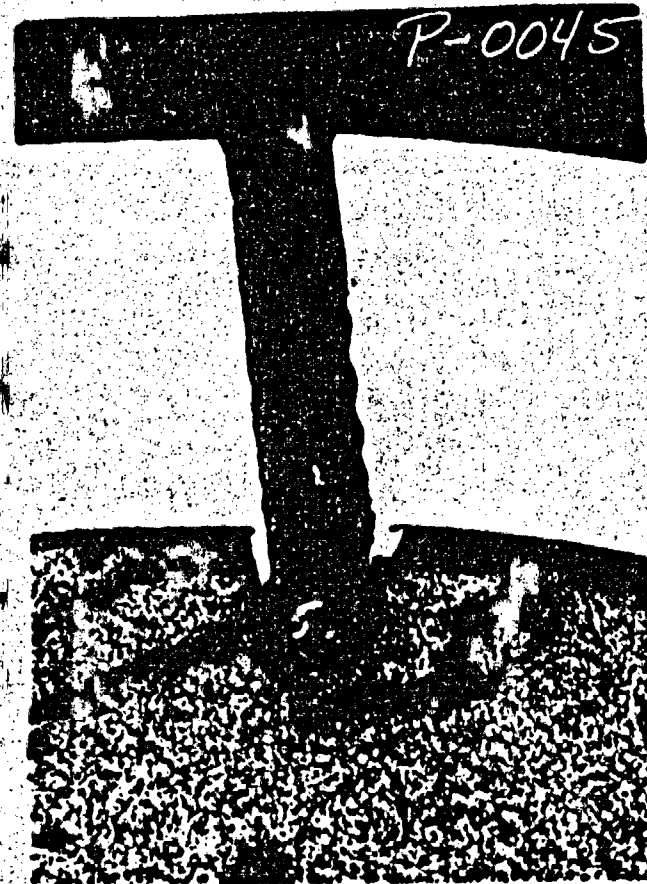
Micrographs of cathodic vacuum etched sections from the slit defect specimens (see Figure 3), and SEM data indicated different as-fabricated initial grain size for the Turkey Point and H. B. Robinson fuels. Average initial grain size appeared to be $\sim 6 \mu\text{m}$ for the H. B. Robinson fuel versus $\sim 25 \mu\text{m}$ for the Turkey Point fuel. Some grain growth (i.e., to a 7-10 μm range) appears to have occurred in the central region of the H. B. Robinson fuel during irradiation. No significant grain growth was observed in the Turkey Point fuel.

79/80



(A) Turkey Point

300 μm



(B) H. B. Robinson

300 μm

FIGURE 19. Polished Sections Through Slit Defect on (A) Turkey Point 1-9-19 Specimen and (B) H. B. Robinson CSC-C Specimen.

4.0 SUMMARY AND CONCLUSIONS

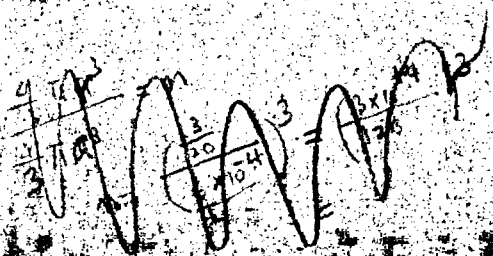
PWR spent fuel specimens were tested in NNWSI reference J-13 well water under ambient hot cell air and temperature conditions in fused quartz vessels. Four specimen configurations and two fuel types were tested. The specimen configurations were: 1) bare fuel plus the cladding, 2) fuel rod segments with water tight end fittings and a machined slit (0.006 inches wide by one inch long) through the cladding, 3) fuel rod segments with water-tight end fittings and small laser-drilled holes (~200 μm diameter) through the cladding, and 4) undefected fuel rod segments with water-tight end fittings. The four specimen configurations were tested using both H. B. Robinson and Turkey Point reactor spent fuels. The tests were semi-static in that periodic solution samples were taken and replenished with fresh J-13 water.

After approximately 6 months, the first cycle of the tests was terminated, the specimens and apparatus rinsed, the vessels stripped in 8M HNO_3 , and the tests restarted in fresh J-13 water for a second cycle. Periodic solution samples, periodic fused quartz rod samples, terminal rinse samples, and terminal vessel acid strip samples were radiochemically analyzed and release values determined for major radionuclides of importance for spent fuel disposal. Results from test Cycles 1 and 2 of the Series 2 tests are reported.

The principal observations and conclusions from the Series 2 spent fuel dissolution tests are summarized below:

- 1) The actinides Pu, Am, Cm, and Np appear to be released congruently. Observed preferential releases of U relative to the other actinides were attributed to higher relative U solubility (i.e. higher percentage of released U in solution where it is readily measured) and to initial preferential U dissolution from oxidized UO_{2+x} on the fuel surface. There was no conclusive evidence indicating actinide phase segregation in as-irradiated spent fuel that may result in non-congruent actinide release.

- 2) Highest observed uranium concentration was 4.8 $\mu\text{g/ml}$. However U concentration tended to equilibrate towards a 1 to 2 $\mu\text{g/ml}$ range, apparently as a higher solubility UO_{2+x} phase was depleted from the fuel surface and U solubility became controlled by a phase having lower U activity.
- 3) True (filtered) uranium solubility was much higher in J-13 water than in the deionized water used in the Series 1 tests. Most uranium measured in J-13 water Series 2 unfiltered samples was also measured in 0.4- μm and 18- μm filtered samples. Most Pu, Am, and Cm activity in unfiltered samples passed through the 0.4- μm filter, and much of the Pu activity also passed through the 18- μm filters.
- 4) Most of the measured radionuclide release to solution occurred during the first few days of a test cycle with some species (e.g., U) then decreasing in concentration as lower chemical activity phases begin to control solubility.
- 5) All of the principal fission product radionuclides measured (^{137}Cs , ^{129}I , ^{99}Tc , and ^{90}Sr) were found to be released preferentially relative to the actinides.
- 6) Activation product ^{14}C was preferentially released (H. B. Robinson tests) relative to the actinides. A portion of the measured ^{14}C release appears to originate from the external surface of the cladding.
- 7) Much lower fractional actinide release was measured in the slit and holes defect tests than in the bare fuel tests.
- 8) Actinide release measured in the tests with small laser-drilled holes through the cladding was not significantly greater than with undefected specimens.



82

Handwritten calculations and notes:

- 20.7
- $= m \frac{4\pi}{3} \left(\frac{3}{20}\right)^3$
- $= m \cdot 0.1413$
- $m = 14.2$
- 100 mg (circled)
- 2.4×10^{-4}
- 10
- 4×10^{-4}
- 2.3×10^{-4}
- 2×10^{-4}
- 1.7×10^{-4}
- 1.5×10^{-4}
- 1.3×10^{-4}
- 1.1×10^{-4}
- 0.9×10^{-4}
- 0.7×10^{-4}
- 0.5×10^{-4}
- 0.3×10^{-4}
- 0.1×10^{-4}
- 0.05×10^{-4}
- 0.02×10^{-4}
- 0.01×10^{-4}
- 0.005×10^{-4}
- 0.002×10^{-4}
- 0.001×10^{-4}
- 0.0005×10^{-4}
- 0.0002×10^{-4}
- 0.0001×10^{-4}
- 0.00005×10^{-4}
- 0.00002×10^{-4}
- 0.00001×10^{-4}
- 0.000005×10^{-4}
- 0.000002×10^{-4}
- 0.000001×10^{-4}

- 9) A significant quantity of loose fuel grains was rinsed from the surface of bare fuel particles during test cycle termination, possibly loosened by grain boundary dissolution during the test.
- 10) Silica, which appears to dissolve from the fused silica test vessels, deposited onto the bare fuel specimens during the tests.
- 11) The J-13 water chemistry remained relatively stable during the tests.

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A P P E N D I X A

RADIONUCLIDE INVENTORY AND RADIOCHEMICAL DATA

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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 10 years after reactor discharge. Since the Turkey Point fuel was very similar (same vendor, same design, similar vintage and same 2.55% ²³⁵U initial enrichment), these ORIGEN-2 data were considered appropriate for both fuels. A factor of 0.8815 was then applied 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 3 of the text. Specimen weights required for calculating per specimen radionuclide inventories are contained in Table 1 of the text.

A.2 RADIOCHEMICAL DATA

With the exception of uranium, all radiochemical data were reported as disintegrations per minute (dpm) per ml of solution or per rod sample. Data were converted from dpm to pCi units using the conversion factor of 1 pCi equal to 2.22 dpm. Since the primary concern was radioactivity release, most data evaluations were based on the pCi activity levels of each radionuclide rather than concentrations. The pCi activity results for all radionuclide determinations, and uranium results as ug/ml (ppm) or ug/rod, are given for each individual test in Tables A-2 through A-9 of this appendix.

For chemical and equilibria considerations, it is usually more useful to consider elemental concentrations rather than radionuclide activities. Concentrations may be calculated using Equations (A.1) and (A.2). The ug/pCi and isotope/element conversion factors for Equation A.1 are contained in Table A.1.

TABLE A.1

ACTIVITY-CONCENTRATION CONVERSION FACTORS

<u>g/pCi</u>	<u>Turkey Point*</u>	<u>H. B. Robinson*</u>
2.24×10^{-7}	--	--
8.82×10^{-10}	--	--
1.44×10^{-5}	0.105	0.106
7.07×10^{-9}	0.546	0.546
5.87×10^{-5}	1.000	1.000
6.13×10^{-3}	0.76	0.76
1.16×10^{-8}	0.40	0.40
1.42×10^{-3}	0.999	0.99999
5.72×10^{-8}	0.012	0.014
1.63×10^{-5}	0.595	0.577
4.41×10^{-6}	0.255	0.263
3.09×10^{-7}	0.863	0.840
1.20×10^{-8}	0.930	0.930

Element mass ratio based on ORIGEN-2 data in PNL-5109 inter-
 comparison of 7.7 MWd/kgM for Turkey Point and 30.2 MWd/kgM for
 H. B. Robinson on burnup, 10 years after discharge.

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

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 Pu concentration from $^{239}\text{Pu} + ^{240}\text{Pu}$ pCi/ml data using Equation (A.1), the ^{239}Pu (or ^{240}Pu) pCi/ml value is needed. The $^{239}\text{Pu}/(^{239}\text{Pu} + ^{240}\text{Pu})$ activity ratio should be 0.374 for the H. B. Robinson fuel and 0.389 for the Turkey Point fuel based on the PNL-5109 ORIGEN-2 data.

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:

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

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

Volume Errors: +2%

Recovery: 100% (no separation required)

Counting Statistics at +1σ:

1 dpm/ml (0.45 pCi/ml)	= <u>+60%</u>
10 dpm/ml (4.5 pCi/ml)	= <u>+8%</u>
100 dpm/ml (45 pCi/ml)	= <u>+2.5%</u>
1000 dpm/ml (450 pCi/ml)	= <u>+1.5%</u>

The counting statistics for 1, 10 and 100 dpm/ml are based on a 100- μ l aliquot plate counted for 480 minutes with a background of 0.2 cpm. The 1000 dpm/ml counting statistic is based on a 100- μ l aliquot plate counted for 100 minutes with a background of 1 cpm. (Higher activity plates are counted on higher background counters, saving newer, lower-background counters for low activity samples.)

• ²⁴¹Am

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

Volume Errors: +3%

Recovery: 97 ± 2%

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

• ²³⁷Np

Method: Separation by cation exchange and solvent extraction.

Volume errors: +2%

Recovery: $98 \pm 2\%$

Counting Statistics at 1σ :

1 dpm/ml (0.45 pCi/ml) = $\pm 30\%$
10 dpm/ml (4.5 pCi/ml) = $\pm 6\%$
100 dpm/ml (45 pCi/ml) = $\pm 4\%$
1000 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 minutes with a background count of 0.2 cpm. Counting statistics for 100 and 1000 dpm/ml are based on a 100-minute 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:

Lower limit 20 dpm/ml at 2σ
100 dpm/ml (45 pCi/ml) = $\pm 11\%$ at 1σ
1000 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 minutes with a background of 30 cpm.

• ^{137}Cs , ^{134}Cs , ^{60}Co

Method: Gamma spectrometry

Volume Errors: +2%

Recovery: 100% (no separation required)

Counting Statistics at $\pm 1\sigma$:

1000 dpm/ml (450 pCi/ml)	= $\pm 20\%$
10,000 dpm/ml (4500 pCi/ml)	= $\pm 8\%$
100,000 dpm/ml (45,000 pCi/ml)	= $\pm 2\%$
10^6 dpm/ml	= $\pm 1\%$

Based on 1-ml aliquot counted for 60 minutes.

• 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 optimum range. The lower limit is 0.001 $\mu\text{g/ml}$ ($\pm 0.001 \mu\text{g/ml}$) using a 100 μl sample aliquot.

A.4

REFERENCES

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

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TABLE A.2
 CSC-H BARE FUEL TEST RADIOCHEMICAL DATA

DAY	SAMPLE TYPE	VOLUME ml	pH	U			Pu-239/Pu-240			Am-241/Pu-238	
				UNFILTERED	0.4 um	18 A	UNFILTERED	0.4 um	18 A	UNFILTERED	0.4 um
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	1.70E+02	9.82E+01	8.29E+03	1.03E+03
6	Rod			5.20E+00			8.15E+03			2.64E+04	
6	Rinse	**		1.70E+00			2.82E+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	9.59E+02
30	Rod			4.90E+00			1.02E+04			4.36E+04	
63	Sol	25	8.221	2.50E+00	2.40E+00	2.10E+00	1.78E+02	1.29E+02	4.80E+01	9.32E+02	8.31E+02
83	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	2.68E+01	7.21E+02	5.81E+02
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	8.58E+02	4.82E+02
181	Rod			7.90E+00			1.35E+04			5.86E+04	
223	Sol	250	8.485	1.20E+00	1.20E+00	1.20E+00	1.32E+02	7.52E+01	2.81E+01	1.11E+03	2.91E+02
223	Rod			6.90E+00			9.41E+03			3.99E+04	
223	Rinse	600			1.10E+00			4.23E+02			1.82E+03
223	Strip	300		9.00E+00			1.35E+04			6.22E+04	

CYCLE 2, TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.427	2.40E+00	2.40E+00	2.40E+00	1.47E+02	1.40E+02	8.94E+01	4.50E+02	3.83E+02
20	Rod			7.50E+01			3.8E+03			1.51E+04	
82	Sol	20	8.460	2.20E+00	2.10E+00	2.00E+00	9.86E+02	7.75E+02	3.06E+02	2.46E+03	2.09E+03
82	Rod			1.40E+01			1.88E+03			7.70E+03	
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	9.19E+01
202	Sol	250	8.580	2.00E+00	2.00E+00	2.00E+00	3.33E+01	2.66E+01	1.62E+01	1.09E+02	8.94E+01
202	Rod			3.50E+00			1.06E+03			4.82E+03	
202	Rinse	600			1.70E+01			3.92E+02			1.94E+03
202	Strip	300		1.00E+00			1.13E+03			5.32E+03	

DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			Sr-90			Pu-237	
				UNFILTERED	0.4 um	18 A	UNFILTERED	0.4 um	18 A	UNFILTERED	0.4 um
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	3.60E+01
63	Rod			1.43E+03						2.70E+00	
120	Sol	15	8.542	<3.38E+04	<2.93E+04	<2.93E+04					
120	Rod			<1.08E+04						<4.50E+00	
181	Sol	20	8.458							<4.50E+01	<4.50E+01
181	Rod			1.07E+04						2.25E+00	
223	Sol	250	8.485	<2.94E+04	<3.35E+04	<2.94E+04				<4.50E+01	<4.50E+01
223	Rod			1.45E+04							
223	Rinse	600			<5.22E+03						<4.50E+01
223	Strip	300		<2.84E+03						9.15E+00	

CYCLE 2, TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.427	2.86E+03	2.98E+03	2.57E+03				5.41E+01	4.50E+01
20	Rod			<1.04E+04							
82	Sol	20	8.460	3.50E+03	3.94E+03	3.26E+03				4.50E+01	4.50E+01
82	Rod										
154	Sol	25	8.200	3.44E+03	4.41E+03	2.61E+03	2.30E+06				
202	Sol	250	8.580	4.64E+03	3.81E+03	3.07E+03	2.52E+06			3.60E+01	4.05E+01
202	Rod						9.01E+05			2.21E+00	
202	Rinse	600						1.18E+05			<2.25E+01
202	Strip	300					8.91E+04			2.25E+01	

UNITS: Solution samples (Sol) in pCi/ml for all but Uranium, ug/ml for Uranium.
 Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.
 Rinse in same units as solution samples, sample was rinsed in 600 ml J-13 water.
 Strip in same units as solution samples, test vessel was stripped with 300 ml 8N HNO3.
 * Am-241 values through 181-day rod sample calculated from Pu-239/Pu-240 and Am-241/Pu-238 values using Pu-238.
 ** Rod rinse sample reported in pCi/rod (ug/rod for Uranium).

TABLE A3
CSCC SLIT DEFECT TEST 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
1	Sol	5	7.792	3.00E-02			6.11E-01			2.70E+00	
6	Sol	10	7.806	5.20E-02	5.60E-02	5.90E-02	1.53E+00	8.91E-01	9.01E-01	8.01E+00	4.14E+00
6	Rod			5.70E-02			6.49E+01			3.17E+02	
6	Rinse	60		2.00E-02			9.81E+00			4.55E+01	
20	Sol	5	8.247	5.50E-02			1.40E+00			7.66E+00	
30	Sol	15	8.406	6.50E-02	5.80E-02	5.70E-02	4.95E+00	1.45E+00	1.89E+00	2.88E+01	7.21E+00
30	Rod			5.60E-02			8.29E+01			3.34E+02	
63	Sol	25	8.195	7.30E-02	7.30E-02	6.70E-02	9.46E+00	1.26E+01	1.26E+01	3.92E+01	6.72E+01
63	Rod			8.30E-02			5.41E+01			1.85E+02	
120	Sol	15	8.530	6.80E-02	7.60E-02	6.40E-02	4.59E+00	3.87E+00	4.64E+00	1.83E+01	1.35E+01
120	Rod			3.20E-02			2.25E+01			8.11E+01	
181	Sol	20	8.460	9.10E-02	9.10E-02	8.60E-02	2.39E+00	2.81E+00	3.15E+00	8.59E+00	1.16E+01
181	Rod			2.30E-02			5.50E+01			2.51E+02	
223	Sol	250	8.496	8.50E-02	8.00E-02	8.00E-02	1.98E+00	1.94E+00	3.33E+00	6.11E+00	8.78E+00
223	Rod			3.60E-02			4.32E+01			1.81E+02	
223	Rinse	600		3.00E-03			2.84E+00			6.88E+00	
223	Strip	300		8.00E-03			8.54E+00			3.42E+01	

CYCLE 2. TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.436	1.10E-01	1.30E-01	1.10E-01	4.05E+00	4.05E+00	4.05E+00	1.04E+01	8.01E+00
20	Rod			2.00E-02			2.70E+01			1.20E+02	
62	Sol	20	8.474	1.60E-01	1.70E-01	1.60E-01	4.80E+00	4.50E+00	3.15E+00	1.13E+01	1.04E+01
62	Rod			2.90E-02			3.11E+01			1.92E+02	
154	Sol	25	8.210	1.90E-01	1.90E-01	1.80E-01	2.67E+00	2.21E+00	2.07E+00	4.19E+00	4.95E+00
202	Sol	250	8.550	1.60E-01	1.30E-01	1.50E-01	1.40E+00	1.53E+00	1.40E+00	3.83E+00	3.63E+00
202	Rod			3.20E-02			4.50E+01			1.23E+02	
202	Rinse	600		2.00E-03			2.43E+00			1.08E+01	
202	Strip	300		4.00E-03			8.46E+00			3.92E+01	

DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			Sr-90			Pu-237	
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um
1	Sol	5	7.792								
6	Sol	10	7.806								
6	Rod										
6	Rinse	60									
20	Sol	5	8.247								
30	Sol	15	8.406								
30	Rod										
63	Sol	25	8.195	<1.80E+04	<1.80E+04	<1.80E+04				<4.50E-01	<4.50E-01
63	Rod									8.01E+00	
120	Sol	15	8.530	<2.93E+04	<2.93E+04	<2.93E+04					
120	Rod			<1.04E+04						<4.50E+00	
181	Sol	20	8.460							<4.50E-01	<4.50E-01
181	Rod									<4.50E+00	
223	Sol	250	8.496	<2.95E+04	<2.96E+04	<2.94E+04				<4.50E-01	<4.50E-01
223	Rod			<1.04E+04						<4.50E-01	
223	Rinse	600		<1.18E+03						<4.50E-01	
223	Strip	300		<1.04E+03						<4.50E-01	

CYCLE 2. TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.436	<6.22E+03	<6.22E+03	<6.22E+03				<2.25E-01	<2.25E-01
20	Rod			<1.04E+04						<2.25E-01	<2.25E-01
62	Sol	20	8.474							<2.25E-01	<2.25E-01
62	Rod										
154	Sol	25	8.210				3.01E+04				
202	Sol	250	8.550				3.20E+08			<2.25E-01	<2.25E-01
202	Rod						4.41E+05			8.01E-01	
202	Rinse	600					2.16E+04			<2.25E-01	
202	Strip	300					3.96E+03			<2.25E-01	

UNITS: Solution samples (Sol) in pCi/ml for all but Uranium, ug/ml for Uranium.
Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.
Rinse in same units as solution samples, sample was rinsed in 600 ml J-13 water.
Strip in same units as solution samples, test vessel was stripped with 300 ml EN HNO3.

* Am-241 values through 181-day rod sample calculated from Pu-239+Pu-240 and Am-241+Pu-238 values using Pu-239+Pu-240 Rod rinse sample reported in pCi/rod (ug/rod for uranium).

A-11/A-0

Am-241			Cs-244			Cs-137			Cs-134		
UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A
1.00E+00			1.31E+00			1.50E+06			8.38E+04		
1.80E+00	2.07E+00		5.41E+00	1.88E+00		9.10E+06	8.29E+06	7.79E+08	5.32E+05	4.86E+05	4.50E+05
1.81E+02			1.88E+02			6.25E+04			3.94E+03		
2.47E+01			2.88E+01			2.82E+05			1.56E+04		
4.73E+00			5.41E+00			7.82E+07			4.27E+08		
1.84E+01	4.28E+00		1.80E+01	4.37E+00	<4.50E-C1	1.01E+08	1.00E+08	9.30E+07	5.72E+06	5.54E+06	5.18E+06
1.60E+02			1.91E+02			5.41E+05			2.32E+04		
1.94E+01	3.08E+01	2.99E+01	2.43E+01	3.74E+01	3.74E+01	1.31E+08	1.30E+08	1.25E+08	7.43E+06	7.52E+06	7.16E+06
8.48E+01			1.22E+02			1.33E+06			7.07E+04		
8.55E+00	5.65E+00		1.05E+01	8.69E+00	<4.50E-C1	1.23E+08	1.27E+08	1.21E+08	6.62E+06	6.49E+06	6.22E+06
3.39E+01			4.23E+01			1.32E+06			4.95E+04		
4.59E+00	8.14E+00	2.40E+00	8.27E+00	8.32E+00	<4.50E-C1	1.23E+08	1.03E+08	1.17E+08	6.09E+06	4.91E+06	5.62E+06
1.36E+02			1.42E+02			2.04E+06			8.50E+04		
5.41E+03	4.95E+00	<1.35E+00	3.92E+00	3.69E+00	<4.50E-C1	1.12E+08	9.82E+07	1.11E+08	5.90E+06	4.68E+06	5.61E+06
1.07E+02			1.01E+02			1.82E+06			1.67E+04		
4.80E+00			4.46E+00			8.92E+05			2.83E+04		
1.98E+01			1.94E+01			9.23E+04			4.06E+03		
8.01E-01	4.50E-01	<1.35E+00	1.80E+00	8.01E-01	<4.50E-C1	2.45E+07	2.36E+07	2.27E+07	1.08E+06	1.06E+06	1.00E+06
8.11E+01			8.51E+01			1.00E+05			47.21E+03		
7.21E-01	<8.01E-01	<8.01E-01	1.35E+00	8.01E-01	<2.25E-01	2.42E+07	2.51E+07	2.51E+07	1.07E+06	1.12E+06	1.12E+06
1.17E+02			1.84E+02			3.82E+05			41.21E+04		
8.86E-01	<4.80E-01	<4.80E-01	7.21E-01	8.31E-01	<4.80E-C2	2.63E+07	2.54E+07	2.40E+07	1.04E+06	1.07E+06	9.80E+05
3.60E-01	8.01E-01	<2.25E-01	8.76E-01	8.61E-01	<2.25E-01	2.82E+07	9.32E+05	2.36E+07	8.89E+05	3.78E+04	9.37E+03
1.42E+02			8.01E-01			8.07E+05			2.58E+04		
41.70E-01			7.21E+00			1.37E+05			4.31E+03		
3.12E+01			2.21E+01			1.84E+04					
Tc-99			Se-78			C-14			I-129		
UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A
3.65E+01	3.87E+01	3.56E+01	<1.58E+01			1.31E+01			2.92E-01		
<8.01E+01			<1.35E+02								
<8.01E+01											
8.01E+00	<8.01E+00	2.48E+01									
8.01E+01			<8.01E+01			5.55E+01			3.77E-01		
2.93E+01	1.81E+01	2.78E+01	<8.01E+00								
1.78E+01											
1.17E+01											
3.39E+01	2.33E+01	2.84E+01									
2.07E+01	3.33E+01	3.06E+01									
3.06E+01						7.57E-01			3.57E-02		
3.38E+01	2.88E+01	2.88E+01	<4.50E+00			8.89E-01			4.73E-02		
<8.01E+01			<4.50E+01								
<8.01E+00			<4.50E+00			3.62E+00			4.85E-03		
<8.01E+00			<4.50E+00								

39-Pa-240) activity ratio of 2.097.

2

TABLE A.4
CSC-C HOLES DEFECT TEST 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
1	Sol	5	7.851	7.00E-03			7.21E-01			2.86E+00		
6	Sol	10	7.835	9.00E-03	8.00E-03	8.00E-03	2.30E+00			7.21E+00	Alpha<1.8 Alf	
6	Rod			<2.00E-02						Alpha<18		
6	Rinse	00		1.00E-02			<9.01E+00			<9.01E+00		
20	Sol	5	8.228	1.00E-02			1.53E+00			7.21E+00		
30	Sol	15	8.171	7.00E-03	7.00E-03	7.00E-03	9.91E-01	8.78E-01	7.21E+00	4.28E+00	3.51E+00	2.2
30	Rod			3.70E-02			5.41E+00			3.47E+01		
51	Sol	2							2.70E+00			7.2
63	Sol	25	8.179	7.00E-03	6.00E-03	7.00E-03	2.25E+00	1.80E+00	3.15E+00	5.85E+00	9.91E+00	1.3
63	Rod			6.20E-02						Alpha<14		
120	Sol	15	8.563	5.00E-03	5.00E-03	5.00E-03	1.13E+00	1.82E+00	1.58E+00	5.41E+00	6.17E+00	3.4
120	Rod			<2.00E-02						Alpha<18		
181	Sol	20	8.408	5.00E-03	5.00E-03	5.00E-03	9.01E-01	9.01E-01	<4.50E-01	4.05E+00	3.15E+00	4.8
181	Rod			<2.40E-02			1.35E+00			1.12E+01		
223	Sol	250	8.497	6.00E-03	5.00E-03	5.00E-03	1.62E+00	1.35E+00	8.56E-01	8.78E+00	8.78E+00	2.1
223	Rod			<2.00E-02						Alpha<14		
223	Rinse	600		1.00E-03						Alpha<1.4		
223	Strip	300		2.00E-03			1.78E+00			5.86E+00		

CYCLE 2, TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.420	<2.00E-03	<2.00E-03	<2.00E-03				Alpha<1.4	Alpha<1.4	Alpha
20	Rod			<2.00E-02			4.50E+00			2.84E+01		
62	Sol	20	8.440	2.00E-03	2.00E-03	1.00E-03	2.70E-01	3.60E-01		8.11E-01	8.56E-01	Alf
62	Rod			<2.00E-02			9.01E+00			4.77E+01		
154	Sol	25	8.200	2.00E-03	2.00E-03	2.00E-03	3.80E-01	3.60E-01	4.10E-02	9.91E-01	8.56E-01	1.3
202	Sol	250	8.540	3.00E-03	2.00E-03	3.00E-03	3.15E-01	2.25E-01	1.80E-01	9.91E-01	8.78E-01	4.8
202	Rod			2.40E-02			9.01E-01			3.15E+00		
202	Rinse	600		1.00E-03			1.31E+00			7.21E+00		
202	Strip	300		1.80E-02			2.30E+01			1.09E+02		

DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			Sr-90			Np-237		
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A
1	Sol	5	7.851									
6	Sol	10	7.835									
6	Rod											
6	Rinse	00										
20	Sol	5	8.228									
30	Sol	15	8.171									
30	Rod											
51	Sol	2										
63	Sol	25	8.179	<1.80E+04	<1.80E+04	<1.80E+04				<4.50E-01	<4.50E-01	<4.5
63	Rod									<4.50E+00		
120	Sol	15	8.563	<1.53E+04	<1.53E+04	<1.53E+04				<4.50E+00		
120	Rod			<1.04E+04						<4.50E-01	<4.50E-01	<4.5
181	Sol	20	8.408							<4.50E+00		
181	Rod									<4.50E-01	<4.50E-01	<4.5
223	Sol	250	8.497	<1.51E+04	<1.54E+04	<1.54E+04				<4.50E-01	<4.50E-01	<4.5
223	Rod			<1.04E+04						<4.50E-01		
223	Rinse	600		<1.04E+03						<4.50E-01		
223	Strip	300		<1.04E+03						<4.50E-01		

CYCLE 2, TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.420	<1.04E+03	<1.04E+03	<1.04E+03				<4.50E-02	<2.25E-01	<2.2
20	Rod			<1.04E+04								
62	Sol	20	8.440							<4.90E-01	<2.25E-01	<2.2
62	Rod											
154	Sol	25	8.200				7.03E+03					
202	Sol	250	8.540				1.13E+04			<2.25E-01	<2.25E-01	<2.2
202	Rod						<7.84E+03			5.41E+00		
202	Rinse	600					<7.86E+02			<2.25E-01		
202	Strip	300					2.21E+03			<2.25E-01		

UNITS: Solution samples (Sol) in pCi/ml for all but Uranium, ug/ml for Uranium.
Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.
Rinse in same units as solution samples. Test was rinsed with 600 ml of J-13 water.
Strip in same units as solution samples. Test vessel was stripped with 300 ml 2N HNO3.

* Am-241 values through 181-day rod sample calculated from Pu-239+Pu-240 and Am-241+Pu-238 values using Pu-238/Am-241 ratio.
** Rod rinse sample reported in pCi/rod (ug/rod for Uranium).

2

TABLE A.5
CSC-A UNDEFECTED TEST RADIOCHEMICAL DATA

DAY	SAMPLE TYPE	VOLUME ml	pH	Pu-239/Pu-240			Am-241/Pu-238					
				UNFILTERED	0.4 um	10 A	UNFILTERED	0.4 um	10 A			
1	Sol	5	7.718	1.00E-02			2.40E+00					
8	Sol	10	7.944	2.00E-02	2.00E-02	1.90E-02	8.80E-01	1.25E+00	2.52E+00			
8	Rod			2.00E-02								
8	Rinse	∞		2.00E-02			<2.00E-00		<2.00E+00			
20	Sol	5	8.156	2.20E-02			1.90E+00		1.08E+01			
30	Sol	15	8.076	2.00E-02	1.90E-02	1.90E-02	2.40E+00	2.00E+00	1.53E+00			
30	Rod			5.60E-02			2.70E+00					
83	Sol	25	8.906	1.80E-02	1.70E-02	1.80E-02	2.60E+00	1.80E+00				
83	Rod			8.00E-02								
120	Sol	15	8.530	1.50E-02	1.50E-02	1.40E-02	1.80E+00	1.40E-01	1.67E+00			
120	Rod			<2.00E-02			4.30E+00					
181	Sol	20	8.405	1.60E-02	1.60E-02	1.40E-02	5.70E-01	1.17E+00	1.22E+00			
181	Rod			<2.40E-02			1.30E+00					
223	Sol	250	8.428	1.30E-02	1.30E-02	1.30E-02	8.60E-01	9.40E-01	8.56E-01			
223	Rod			<2.00E-02					Alpha<14			
223	Rinse	600		<1.00E-03			3.70E-01		1.22E+00			
223	Strip	300		2.00E-03			1.70E+00		1.17E+01			
CYCLE 2, TEST RESTARTED IN FRESH J-12 WATER												
20	Sol	20	8.426	<2.00E-03	<1.00E-03	<2.00E-03	3.60E+00					
20	Rod			<2.00E-02			2.70E-01	3.80E-01	5.80E-01			
82	Sol	20	8.385	2.00E-03	2.00E-03	2.00E-03	5.40E+00					
82	Rod			<2.00E-02			5.40E+00					
154	Sol	25	8.18	2.00E-03	2.00E-03	1.00E-03	3.60E-01	4.90E-01	1.23E-01			
202	Sol	250	8.52	1.00E-03	1.00E-03	1.00E-03	2.70E-01	4.80E-01	3.80E-01			
202	Rod			1.70E-02			<2.20E-01		<1.90E+00			
202	Rinse	600		1.00E-03			8.80E-01		3.47E+00			
202	Strip											
DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			Sr-90			Mn-237		
				UNFILTERED	0.4 um	10 A	UNFILTERED	0.4 um	10 A	UNFILTERED	0.4 um	10 A
1	Sol	5	7.718									
8	Sol	10	7.944									
8	Rod											
8	Rinse	∞										
20	Sol	5	8.156									
30	Sol	15	8.076									
30	Rod											
83	Sol	25	8.906	1.44E+01	2.11E+02	1.90E+02				<4.50E-01	<4.50E-01	
83	Rod									<4.50E+00		
120	Sol	15	8.530	2.53E+02	1.54E+02	1.40E+02						
120	Rod			<1.04E+04						<4.50E+00		
181	Sol	20	8.405	2.87E+02	1.83E+02	1.50E+02				<4.50E-01	5.41E-01	
181	Rod									<4.50E+00		
223	Sol	250	8.428	1.47E+02	1.89E+02	8.91E+01				<4.50E-01	<4.50E-01	
223	Rod			<1.04E+03								
223	Rinse	600		<1.04E+02						<4.50E-01		
223	Strip	300		<1.04E+02						<4.50E-01		
CYCLE 2, TEST RESTARTED IN FRESH J-12 WATER												
20	Sol	20	8.426	<1.04E+02	<1.04E+02	<1.04E+02				<4.50E-02	<4.50E-02	
20	Rod			<1.04E+03						<4.50E-01	<4.50E-01	
82	Sol	20	8.385							<4.50E-01	<4.50E-01	
82	Rod											
154	Sol	25	8.18				1.40E-02					
202	Sol	250	8.52				1.80E-03			<2.25E-01	<2.25E-01	
202	Rod						<7.60E-03			9.61E-01		
202	Rinse	600					<7.60E-02			<2.25E-01		
202	Strip											

UNITS: Solution samples (Sol) in pCi/ml for all but Uranium, ug/ml for Uranium.
 Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.
 Rinse in same units as solution samples. Sample was rinsed in 600 ml J-12 water.
 Strip in same units as solution samples. Test vessel was stripped with 300 ml 8N HNO3.

∞ Rod rinse sample reported in pCi/rod (ug/rod for Uranium).
 The CSC-A vessel broke during Cycle 2 terminal rinsing and was not stripped.

A-15/A-16

Am-241

Cm-244

Cf-252

Cf-254

UNFILTER 0.4 mm 18 A

UNFILTER 0.4 mm 18 A

UNFILTER 0.4 mm 18 A

UNFILTER 0.4 mm 18 A

1.44E+00
1.56E+00 8.41E-012.27E+04
2.38E+04 2.51E+04 2.24E+041.13E+03
1.46E+03 1.38E+03 1.23E+032.89E+00
2.47E+00 7.86E-01 <4.50E-01

3.35E+04 3.46E+04 3.20E+04

1.81E+03 1.82E+03 1.83E+03

8.11E+00
9.91E-01 1.04E+00

4.72E+03 3.45E+04 3.47E+04 3.24E+04

4.83E+03 1.84E+03 1.85E+03 1.82E+03

1.89E+00 9.01E-01 <4.50E-01

2.72E+03 2.82E+04 2.95E+04 2.69E+04

4.83E+03 1.81E+03 1.37E+03 1.39E+03

1.31E-01
7.21E-01 9.91E-01 <4.50E-01

8.60E+03 2.89E+04 2.84E+04 2.68E+04

4.83E+03 1.80E+03 1.37E+03 1.12E+03

2.25E+00
6.31E-01 1.08E+00 <4.50E-01

2.51E+03 2.72E+04 2.75E+04 2.89E+04

4.83E+03 1.82E+03 1.37E+03 1.01E+03

1.71E+00 1.08E+00 <1.35E+00
5.41E+007.66E-01
6.31E+004.18E+02
4.62E+024.83E+03
4.83E+031.70E+00
7.21E+007.66E-01
6.31E+004.18E+02
4.62E+024.83E+03
4.83E+03

<4.50E-01 <4.50E-01 <9.01E-01

1.40E-01

3.40E+02 4.24E+02 2.36E+02

<6.08E+01 <6.31E+01 <6.31E+01

2.12E+01
<4.50E-01 <4.50E-01 <4.50E-01

4.22E-01 <2.25E-01 <2.25E-01

4.83E+02 4.80E+02 4.82E+02

4.83E+02 4.83E+02 4.83E+02

2.97E-01
<9.01E-01 <9.01E-01 <9.01E-013.81E-01
1.35E-01 1.80E-01 <4.50E-02

8.73E+02 1.02E+03 9.23E+02

4.83E+02 4.83E+02 4.83E+02

4.50E-01 <2.25E-01 <2.70E-01

<1.35E-01 1.80E-01 <1.35E-01

1.10E+03 1.02E+03 1.19E+03

7.49E+01 <6.31E+01 <6.76E+01

2.97E+01
<2.70E-014.83E-01
2.12E+004.83E+02
4.78E+014.83E+02
4.83E+01

Tc-99

Pu-239

C-14

X-135

UNFILTER 0.4 mm 18 A

UNFILTER 0.4 mm 18 A

UNFILTER 0.4 mm 18 A

UNFILTER 0.4 mm 18 A

1.67E+01 1.67E+01 2.79E+01
<9.01E-01<1.50E-01
<1.35E-02

2.25E+01

6.04E-03

<9.01E-01

<1.35E-02

1.80E+01

5.41E-03

<9.01E+00 <9.01E+00 <9.01E+00

<9.01E-01

1.60E+01

7.21E-03

<9.01E+00 <9.01E+00 <9.01E+00

<9.01E-01

7.21E+00

8.93E-03

1.60E+01
<9.01E+00

<9.01E-01

9.91E+00

<9.01E+00 <9.01E+00 <9.01E+00

<4.50E-00

1.60E+01

7.21E-03

<9.01E+00 <9.01E+00 <9.01E+00

<4.50E-01

7.21E+00

8.93E-03

<9.01E+00

<4.50E-00

9.91E+00

<9.01E+00 <9.01E+00 <9.01E+00

<4.50E-00

9.91E+00

<9.01E+01

<4.50E-01

9.91E+00

<9.01E+00

<4.50E-00

9.91E+00

TABLE A.8
1-24 BARE FUEL TEST RADIOCHEMICAL DATA

DAY	SAMPLE TYPE	VOLUME ml	pH	U			Pu-239+Pu-240			Am-241+Pu-238		
				UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A
1	Sol	5	7.926	3.10E+00			1.55E+03			7.30E+03		
6	Sol	10	8.162	4.50E+00	4.40E+00	4.20E+00	5.55E+02	4.80E+02	2.64E+02	2.45E+03	2.04E+03	5.38E+02
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		
20	Sol	15	8.320	4.80E+00	4.80E+00	5.40E+00	4.80E+02	5.00E+02	1.82E+02	2.11E+03	1.88E+03	3.33E+02
20	Rod			2.40E+00			4.14E+03			1.85E+04		
82	Sol	20	8.288	4.80E+00	4.80E+00	4.00E+00	5.27E+02	4.55E+02	1.88E+02	2.17E+03	1.73E+03	3.40E+02
82	Rod			1.60E+00			2.91E+03			1.05E+04		
120	Sol	20	8.429	4.80E+00	4.80E+00	4.20E+00	4.88E+02	4.59E+02	2.07E+02	1.93E+03	1.70E+03	3.80E+02
120	Rod			1.10E+00			2.04E+03			8.18E+03		
181	Sol	250	8.458	4.00E+00	4.00E+00	4.00E+00	4.85E+02	4.11E+02	2.17E+02	1.77E+03	1.57E+03	4.64E+02
181	Rod			2.00E+00			3.32E+03			1.30E+04		
181	Rinse	600			8.10E-01			1.05E+02			3.78E+02	
181	Strip	300		3.20E+00			3.78E+03			1.44E+04		
CYCLE 2, TEST RESTARTED IN FRESH J13 WATER												
20	Sol	20	8.462	1.40E+00	1.40E+00	1.40E+00	2.48E+02	2.36E+02	1.94E+02	5.18E+02	5.00E+02	3.85E+02
20	Rod			1.80E+00			1.93E+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	2.49E+02
71	Rod			3.80E-01			1.90E+03			1.15E+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	3.48E+02	2.16E+02
195	Sol	250	8.490	2.40E+00	2.20E+00	2.10E+00	1.90E+02	1.71E+02	1.44E+02	3.78E+02	3.38E+02	2.30E+02
195	Rod			1.10E+00			1.95E+03			1.47E+04		
195	Rinse	600			8.50E-02			4.41E+01			1.57E+02	
195	Strip	300		8.20E-01			8.80E+02			2.48E+03		
DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			Sr-90			Pu-237		
				UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A	UNFILTER	0.4 um	10 A
1	Sol	5	7.926	3.41E+04								
6	Sol	10	8.162	1.83E+05	1.82E+05	1.42E+05						
6	Rod			6.74E+02								
15	Sol	5	8.144	3.53E+05								
20	Sol	15	8.320	4.85E+05	4.86E+05	2.81E+05						
20	Rod			5.41E+04								
82	Sol	20	8.288	5.58E+05	5.50E+05	4.28E+05				1.80E+00		
82	Rod			2.95E+04								
120	Sol	20	8.429	6.17E+05	6.26E+05	5.32E+05				9.46E-01	1.04E+00	1.17E+00
120	Rod			3.05E+04								
181	Sol	250	8.458	6.17E+05	6.04E+05	5.54E+05				4.50E-01	4.50E-01	4.50E-01
181	Rod			2.30E+05								
181	Rinse	600			7.93E+04					4.25E-01		
181	Strip	300		1.46E+04						4.50E-01		
CYCLE 2, TEST RESTARTED IN FRESH J13 WATER												
20	Sol	20	8.462	1.21E+05	1.22E+05	1.07E+05				4.25E-01	4.25E-01	3.80E-01
20	Rod			6.26E+04								
71	Sol	20	8.485	1.30E+05	1.55E+05	1.22E+05				4.50E-01	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+03					
195	Sol	250	8.490	1.32E+05	1.36E+05	1.13E+05	3.54E+03			3.60E-01	4.85E-01	3.80E-01
195	Rod			4.73E+05			2.61E+05			1.80E+00		
195	Rinse	600			2.68E+03			9.01E+03			4.25E-01	
195	Strip	300		3.52E+03			5.23E+04			4.25E-01		

UNITS: Solution samples (Sol) in pCi/ml for all but Uranium, ug/ml for Uranium.
 Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.
 Rinse in same units as solution samples, sample was rinsed in 600 ml J-13 water.
 Strip in same units as solution samples, test vessel was stripped with 300 ml 5M HNO3.

Am-241 values through 120-day rod sample calculated from Pu-239+Pu-240 and Am-241+Pu-238 values using Pu 238/(Pu-239+Pu-240) = 0.15

Am-241			Cm-246			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
4.46E-03			4.46E-03			1.12E-07			9.32E-05		
1.47E-03	1.16E-03	6.30E-01	1.50E-03	1.00E-03	3.06E-01	1.30E-07	1.33E-07	1.25E-07	1.09E-06	1.09E-06	1.05E-06
1.55E-04			1.53E-04			6.35E-05			4.45E-04		
7.34E-03			7.52E-03			1.42E-07			1.16E-06		
1.24E-03	1.08E-03	1.34E-01	1.20E-03	1.07E-03	1.83E-00	1.42E-07	1.41E-07	1.36E-07	1.13E-06	1.12E-06	1.08E-06
9.04E-03			7.84E-03			3.97E-05			3.23E-04		
1.23E-03	9.16E-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		
1.06E-03	8.79E-02	8.86E-00	1.06E-03	8.87E-02	2.39E-01	1.22E-07	1.32E-07	1.30E-07	9.50E-05	9.73E-05	9.65E-05
4.41E-03			4.04E-03			9.27E-05			3.84E-04		
8.73E-02	8.24E-02	7.81E-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
8.80E-03			8.17E-03			7.07E-05			4.91E-04		
7.25E-03	1.85E-02		5.32E-03	1.73E-02		4.59E-05	1.31E-06		3.17E-04	9.05E-04	
8.15E-01	5.86E-01	4.50E-00	7.21E-01	5.77E-01	3.50E-00	2.26E-05	2.20E-05	2.17E-05	1.46E-04	1.26E-04	1.34E-04
1.38E-04			1.10E-04			1.14E-05			1.02E-04		
8.11E-01	7.21E-01	8.41E-00	8.01E-01	7.66E-01	4.05E-00	4.23E-05	4.27E-05	3.99E-05	2.87E-04	2.78E-04	2.48E-04
1.04E-04			1.03E-04			3.79E-04			1.24E-04		
8.51E-01	8.69E-01	3.33E-00	8.47E-01	7.03E-01	2.23E-00	8.94E-05	8.78E-05	8.40E-05	4.18E-04	4.22E-04	3.70E-04
8.42E-01	8.17E-01	7.21E-00	8.11E-01	8.41E-01	3.80E-00	8.15E-05	8.24E-05	7.93E-05	4.84E-04	5.14E-04	4.73E-04
1.45E-04			1.25E-04			1.24E-05					
1.33E-03	8.42E-01		9.50E-02	8.13E-01		4.45E-04	2.37E-04		2.08E-03	1.63E-03	
Tc-99			Sg-79			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
1.82E-02											
2.03E-02	2.25E-02	2.03E-02									
2.12E-02	1.85E-02	1.89E-02	<9.01E-00			5.27E-01			5.90E-01		
	1.94E-01										
2.03E-01											
2.30E-01	1.89E-01	2.34E-01									
2.78E-01	2.03E-01	4.19E-01									
8.41E-01						5.41E-01			1.86E-01		
4.50E-01	7.21E-01	5.96E-01	<9.01E-00			4.50E-01			2.19E-01		
<9.01E-01			<9.01E-00								
<9.01E-00	<9.01E-00		<9.01E-00	<9.01E-00		5.86E-00			7.31E-03		
			<9.01E-00						1.91E-02		

Am-241 activity ratio of 1.792.

2

TABLE A.7
1-8-19 SLIT DEFECT TEST 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
1	Sol	5	8.021	8.00E-02			8.50E-00			2.86E-01		
8	Sol	10	8.157	2.80E-01	2.00E-01	1.80E-01	6.31E-00	4.32E+00	1.22E+00	2.03E-01	1.24E-01	2.12E-01
8	Rod			4.30E-01			4.64E-02			1.77E-03		
15	Sol	5	8.169	3.40E-01			3.60E-00			1.22E-01		
30	Sol	15	8.325	4.40E-01	4.40E-01	4.30E-01	5.41E-00	5.41E+00	1.80E+00	2.16E-01	1.88E-01	3.15E-01
30	Rod			4.30E-02			1.00E-02					
62	Sol	20	8.315	5.40E-01	5.30E-01	4.90E-01	7.86E-00	7.21E+00	2.84E+00	3.85E-02	2.97E-01	8.98E-01
62	Rod			1.80E-02			1.26E-01			5.34E-01		
120	Sol	20	8.448	7.20E-01	6.90E-01	6.60E-01	9.91E-00	9.01E+00	4.23E+00	3.47E-01	2.88E-01	7.66E-01
120	Rod			3.60E-02			1.49E-01			6.82E-01		
181	Sol	250	8.473	8.50E-01	8.10E-01	8.20E-01	7.21E-00	9.01E+00	3.60E+00	2.84E-01	2.75E-01	8.76E-01
181	Rod			2.60E-02			2.97E-01			1.25E-02		
181	Rinse	600		1.70E-02			1.94E-01			8.60E-01		
181	Strip	300		5.30E-02			7.70E-01			2.60E-02		
CYCLE 2. TEST RESTARTED IN FRESH J-13 WATER												
20	Sol	20	8.460	1.40E-01	1.30E-01	1.40E-01	8.78E-00	4.50E+00	4.50E+00	1.35E-01	9.91E-00	8.46E-01
20	Rod			1.40E-02			2.57E-01			1.20E-02		
71	Sol	20	8.460	3.50E-01	3.50E-01	3.70E-01	7.86E-00	7.66E+00	4.50E+00	1.62E-01	1.67E-01	7.86E-01
71	Rod			2.60E-02			5.32E-01			2.08E-02		
154	Sol	25	8.500	4.90E-01	4.80E-01	4.20E-01	7.86E-00	7.66E+00	3.80E+00	1.62E-01	1.62E-01	8.18E-01
195	Sol	250	8.480	5.00E-01	5.00E-01	5.00E-01	7.66E-00	8.31E+00	3.33E+00	1.71E-01	1.35E-01	8.41E-01
195	Rod			2.60E-02			7.21E-01			2.97E-02		
195	Rinse	600		6.80E-03			5.86E-01			1.62E-00		
195	Strip	300		1.80E-03			7.21E-00			2.68E-01		
CYCLE 3. TEST RESTARTED IN FRESH J-13 WATER												
1	Sol	5	8.021	1.43E+03								
8	Sol	10	8.157	3.59E+03	2.88E+03	4.01E+03						
8	Rod			1.04E+03								
15	Sol	5	8.169	1.75E+04								
30	Sol	15	8.325	4.73E+04	4.20E+04	2.37E+04						
30	Rod			3.57E+03								
62	Sol	20	8.315	7.82E+04	7.52E+04	8.04E+04						
62	Rod			3.92E+03								
120	Sol	20	8.448	8.35E+04	6.58E+04	5.77E+04						
120	Rod			8.11E+03								
181	Sol	250	8.473	4.82E+04	4.42E+04	4.62E+04						
181	Rod			2.62E+04								
181	Rinse	600		1.16E+03								
181	Strip	300		7.07E+03								
CYCLE 4. TEST RESTARTED IN FRESH J-13 WATER												
20	Sol	20	8.460	5.05E+03	5.41E+03	4.38E+03						
20	Rod			2.77E+03								
71	Sol	20	8.460	8.78E+03	7.07E+03	5.14E+03						
71	Rod			4.25E+03								
154	Sol	25	8.500	6.40E+03	7.16E+03	3.78E+03	1.31E+05					
195	Sol	250	8.480	7.34E+03	4.95E+03	6.44E+03	3.02E+05					
195	Rod			1.48E+04			3.15E+04					
195	Rinse	600		2.37E+02			7.66E+03					
195	Strip	300		9.05E+02			1.17E+03					

UNITS: Solution samples (Sol) in pCi/ml for all but Uranium, ug/ml for Uranium.
 Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.
 Rinse in same units as solution samples, sample was rinsed in 600 ml J-13 water.
 Strip in same units as solution samples, test vessel was stripped with 300 ml 8N HNO3.

* Am-241 values through 120-day rod sample calculated from Pu-239+Pu-240 and Am-241+Pu-238 values using Pu-239/(Pu-239+Pu-240) = 0.45

A-19/120

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	
0.24E+00				1.04E+01				3.58E+05				2.92E+04			
7.45E+00	7.38E+00			1.13E+01	8.31E+00	2.25E-01		2.73E+06	2.80E+06	2.70E+06		2.23E+03	2.14E+03	2.1E+03	
8.35E+02				8.38E+02				6.58E+04				4.73E+03			
4.88E+00				5.88E+00				6.17E+06				3.14E+03			
1.07E+01	1.28E+01			1.22E+01	1.13E+01	4.50E-01		8.85E+06	8.80E+06	8.33E+06		6.94E+03	7.03E+03	8.6E+03	
1.82E+02				1.82E+02				2.08E+05				1.64E+04			
1.38E+01	1.67E+01	1.23E+00		1.67E+01	1.49E+01	1.78E+00		1.01E+07	1.05E+07	9.64E+06		8.20E+03	8.33E+03	7.7E+03	
2.81E+01				2.16E+01				2.85E+05				2.35E+04			
1.45E+01	1.89E+01			1.58E+01	1.31E+01	5.85E-01		1.26E+07	1.25E+07	1.22E+07		9.64E+03	9.50E+03	9.3E+03	
3.81E+01				3.33E+01				2.52E+05				2.01E+04			
1.25E+01	1.49E+01	1.35E+00		1.58E+01	1.31E+01	9.01E-01		1.41E+07	1.41E+07	1.33E+07		1.05E+06	1.07E+06	1.6E+06	
1.03E+02				6.26E+01				8.50E+05				3.85E+04			
8.45E+01				4.10E+01				2.00E+05				1.40E+04			
1.66E+02				1.23E+02				3.19E+04				2.23E+03			
2.03E+00	1.58E+00	<1.35E+00		1.80E+00	9.01E-01	4.50E-01		8.24E+05	8.38E+05	7.88E+05		5.54E+04	5.88E+04	5.41E+04	
7.48E+01				8.78E+01				1.14E+04				<9.46E+02			
9.86E+00	4.50E+00	<1.35E+00		3.80E+00	2.70E+00	4.50E-01		2.81E+06	2.72E+06	2.68E+06		1.88E+05	1.81E+05	1.74E+05	
1.22E+02				1.27E+02				2.05E+04				<1.34E+03			
8.36E+00	3.78E+00	<4.50E-01		3.80E+00	3.15E+00	4.91E-02		7.07E+06	8.88E+06	8.62E+06		4.30E+05	4.27E+05	4.97E+05	
3.80E+00	5.81E+01	<4.50E-01		4.05E+00	2.97E+00	4.25E-01		7.84E+06	7.88E+06	7.70E+06		4.77E+05	4.64E+05	4.94E+05	
1.80E+02				1.71E+02				2.80E+05				1.41E+04			
3.42E+00				6.78E+01				8.98E+04				3.83E+03			
1.08E+01				9.91E+00				6.71E+03				4.12E+02			
Tc-99				Sr-90				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	
1.85E+02															
2.67E+02	1.94E+02	2.07E+02													
2.03E+02	1.98E+02	2.21E+02		<9.01E+00				7.21E+01				2.89E+01			
<9.01E+00															
<9.01E+00															
4.80E+01	4.95E+01	4.50E+01													
6.76E+01	6.76E+01	6.31E+01													
7.65E+01								3.47E+01				1.89E+01			
7.64E+01	8.55E+01	8.11E+01		<9.01E+00				3.54E+01				2.17E+01			
<9.01E+01				<9.01E+01											
<9.01E+00				<9.01E+00				2.25E+00							
<9.01E+00				<9.01E+00								1.01E+02			

2

TABLE A.8
I-9-12 HOLES DEFECT TEST 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
				1	Sol	5	7.752	1.20E-02	2.40E-02	2.10E-02	4.95E+00	3.47E+00
8	Sol	10	8.138	1.10E-01	2.90E-02	2.10E-02	4.28E+00	1.89E+00	1.55E+03	8.78E+00	7.86E+00	
15	Sol	5	8.170	2.90E-02	3.30E-02	3.90E-02	2.65E+00	3.60E+00	2.25E+00	7.86E+00	9.48E+00	2.70E+00
30	Sol	15	8.315	2.90E-02	3.30E-02	3.90E-02	2.25E+00	3.60E+00	2.25E+00	7.86E+00	9.48E+00	2.70E+00
30	Rod			<2.00E-02			1.98E+01			5.54E+01		
62	Sol	20	8.343	3.30E-02	3.20E-02	3.00E-02	2.16E+00	1.94E+00	8.56E-01	6.76E+00	6.58E+00	1.80E+00
62	Rod			1.80E-02			3.07E-01			1.02E+02		
120	Sol	20	8.462	3.10E-02	3.10E-02	3.70E-02	1.08E+00	2.34E+00	8.56E-01	4.05E+00	6.76E+00	1.62E+00
120	Rod			<2.00E-02			1.08E+01			3.42E+01		
181	Sol	250	8.478	2.90E-02	2.90E-02	2.90E-02	1.80E+00	2.25E+00	<4.50E-01	6.31E+00	6.31E+00	<9.01E-00
181	Rod			<2.00E-02			2.39E+01			9.85E+01		
181	Rinse	600		<1.00E-03			2.25E+00			1.05E+01		
181	Strip	300		9.00E-03			1.76E+01			8.86E+01		

CYCLE 2. TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.453	3.00E-03	3.00E-03	3.00E-03	9.01E-01	1.35E+00		2.70E+00	2.25E+00	Alphac.
20	Rod			1.10E-02			8.56E+00			3.97E+01		
71	Sol	20	8.425	3.00E-03	3.00E-03	3.00E-03	9.11E-01	7.21E-01	7.66E-01	2.21E+00	1.62E+00	1.67E+00
71	Rod			<2.00E-02			1.04E+01			5.41E+01		
154	Sol	25	8.510	4.00E-03	4.00E-03	3.00E-03	9.66E-01	9.46E-01	5.41E-01	2.57E+00	1.88E+00	1.04E+00
195	Sol	250	8.540	4.00E-03	4.00E-03	3.00E-03	7.21E-01	9.31E-01	4.95E-01	1.55E+00	1.49E+00	9.64E-01
195	Rod			<2.00E-02			1.28E+01			5.77E+01		
195	Rinse	600		<1.00E-03			1.80E-01			4.05E-01		
195	Strip	300		1.00E-03			3.38E+00			1.22E+01		

DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			Sr-90			Pu-239		
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A
				1	Sol	5	7.962	9.23E-02	7.97E-02	9.88E-02		
8	Sol	10	8.138	1.04E-03	7.12E-02	9.88E-02						
15	Sol	5	8.170	1.71E-03								
30	Sol	15	8.315	2.95E-03	2.94E-03	1.63E-03						
30	Rod											
62	Sol	20	8.343	9.77E-03	9.86E-03	6.48E-03				<4.50E-01		
62	Rod			8.56E-02								
120	Sol	20	8.462	9.37E-03	8.78E-03	7.70E-03				<9.01E-01	<8.01E-01	<9.01E-01
120	Rod			2.09E-03								
181	Sol	250	8.478	7.39E-03	7.61E-03	8.44E-03				<2.25E-01	<2.25E-01	<2.25E-01
181	Rod			1.27E-03								
181	Rinse	600		1.53E-02						<4.50E-01		
181	Strip	300		9.73E-02						<2.25E-01		

CYCLE 2. TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.453	5.99E-02	5.39E-02	4.23E-02				<2.25E-01	<2.25E-01	<2.25E-01
20	Rod			<1.04E-03								
71	Sol	20	8.425	6.53E-02	6.26E-02	3.95E-02				<2.25E-01	<2.25E-01	<4.50E-01
71	Rod			<1.32E-03								
154	Sol	25	8.510	5.50E-02	8.11E-02	4.04E-02	1.22E-03					
195	Sol	250	8.540	4.73E-02	4.10E-02	4.47E-02	7.68E-02			<2.25E-01	<2.25E-01	<2.25E-01
195	Rod			9.41E-02			<8.11E-03			2.72E+00		
195	Rinse	600					7.21E-02			<2.25E-01		
195	Strip	300		1.40E-02			<7.68E-02			<2.25E-01		

UNITS: Solution samples (Sol) in pCi/ml for all but Uranium, ug/ml for Uranium.
 Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.
 Rinse in same units as solution samples, sample was rinsed in 600 ml J-13 water.
 Strip in same units as solution samples, test vessel was stripped with 300 ml J-13 water.
 Am-241 values through 120-day rod sample calculated from Pu-239/Pu-240 and Pu-239/Pu-238 values using Pu-239/Pu-238 = 0.115.

As-241g			Cs-266			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
4.67E+00			2.88E+00			2.13E+04			1.82E+03		
2.24E+00	2.85E+00		2.07E+00	1.31E+00		1.00E+05	9.55E+04	1.06E+05	8.06E+03	7.16E+03	8.02E+03
8.28E+02			7.52E+02			3.14E+03			<6.31E+02		
2.20E+00			1.80E+00			3.91E+05			2.94E+04		
3.84E+00	3.36E+00		2.25E+00	2.70E+00	<4.50E-01	8.72E+05	8.72E+05	5.36E+05	4.59E+04	4.43E+04	4.25E+04
2.19E+01			2.12E+01			2.33E+04			5.77E+04	5.90E+04	5.36E+04
3.10E+00	3.30E+00	3.53E-01	2.30E+00	1.58E+00	<4.50E-01	7.52E+05	7.75E+05	8.85E+05	1.48E+03		
3.12E+01			4.55E+01			2.45E+04			5.23E+04	5.14E+04	5.09E+04
2.22E+00	2.78E+00	1.73E-01	1.04E+00	2.18E+00	1.01E-02	7.16E+05	6.98E+05	6.94E+05	2.92E+03		
1.59E+01			1.71E+01			3.99E+04			5.27E+04	5.32E+04	5.14E+04
3.15E+00	1.35E+00	9.01E-01	2.25E+00	1.80E+00	<4.50E-01	7.78E+05	7.78E+05	7.66E+05	3.04E+03		
8.31E+01			4.46E+01			4.95E+04			3.25E+02		
6.78E+00			5.41E+00			5.36E+03			2.76E+02		
3.74E+01			3.02E+01			5.14E+03					

8.01E-01	<1.35E+00	<1.35E+00	<4.50E-01	<4.50E-01		4.88E+04	4.73E+04	4.49E+04	3.23E+03	3.18E+03	2.83E+03
5.09E-01			2.12E+01			2.12E+03			<8.40E+02		
4.80E+00	<9.01E-01	<4.50E-01	<2.25E-01	<2.25E-01	<2.25E-01	1.16E+05	1.13E+05	1.10E+05	7.84E+03	7.81E+03	6.89E+03
4.23E-01			2.34E+01			5.95E+03			<6.08E+02		
<4.50E-01	<4.50E-01	<4.50E-01	7.21E-01	<2.25E-01	<2.25E-01	1.46E+05	1.46E+05	1.36E+05	8.92E+03	8.87E+03	8.47E+03
1.13E+00	<4.50E-01	<4.50E-01	1.35E-01	<2.25E-01	<4.50E-01	1.84E+05	1.82E+05	1.47E+05	8.60E+03	9.10E+03	8.42E+03
3.47E-01			2.21E+01			7.93E+03					
8.41E-01			2.25E-01			1.01E+03					
8.31E+00			4.32E+00			8.05E+02					

Tc-99			Sr-79			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
1.17E+01											
<1.80E+01	<1.80E+01	<1.80E+01									
<9.01E+00	1.35E+01	<9.01E+00	<9.01E+00			1.87E-01			2.60E-02		
<9.01E+00											
<9.01E+00											
<9.01E+00	<9.01E+00	<9.01E+00									
<9.01E+00	<9.01E+00	<9.01E+00									
<9.01E+00						3.28E-01			5.33E-03		
<9.01E+00	<9.01E+00	<9.01E+00	<9.01E+00			3.42E-01			6.58E-03		
<9.01E+00			<9.01E+00			2.70E-00			1.36E-03		
<9.01E+00			<9.01E+00								

2

TABLE A.9
I-8-1 UNDEFECTED TEST 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
1	Sol	5	7.932	1.00E-02			2.52E+00			4.50E+00		
6	Sol	10	8.060	1.10E-02	1.10E-02	1.10E-02	1.13E+00	4.95E-01	8.11E-01	2.75E+00	1.28E+00	1.93E+00
6	Rod			2.00E-02						Alpha<12		
15	Sol	5	8.128	1.10E-02			1.28E+00			3.20E+00		
30	Sol	15	8.253	1.10E-02	1.00E-02	1.00E-02	1.54E+00	9.01E-01		3.15E+00	2.25E+00	Alpha<8
30	Rod			<2.00E-02						Alpha<6		
82	Sol	20	8.308	1.00E-02	1.00E-02	9.00E-03	1.17E+00	9.91E-01		3.65E+00	2.87E+00	Alpha<1.4
82	Rod			1.80E-02			2.85E+00			7.97E+00		
120	Sol	20	8.424	9.00E-03	8.00E-03	9.00E-03	9.01E-01	7.21E-01	4.95E-01	3.15E+00	2.30E+00	1.05E+00
120	Rod			<2.00E-02			1.89E+01			8.80E+01		
181	Sol	250	8.440	8.00E-03	9.00E-03	9.00E-03	4.80E-01	9.01E-01	1.35E+00	1.35E+00	2.70E+00	9.01E-01
181	Rod			<2.00E-02			8.78E+00			1.98E+01		
181	Rinse	800		<1.00E-03			4.80E-01			1.17E+00		
181	Strip	300		<1.00E-03			4.37E+00			1.80E+01		

CYCLE 2, TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.480	3.00E-03	3.00E-03	3.00E-03	4.50E-01	9.01E-01	4.50E-01	1.35E+00	1.80E+00	9.01E-01
20	Rod			1.10E-02			8.11E+00			3.81E+01		
71	Sol	20	8.480	2.00E-03	2.00E-03	2.00E-03	4.80E-01	4.80E-01	8.88E-01	9.01E-01	9.01E-01	7.88E-01
71	Rod			<2.00E-02			8.41E+00			2.34E+01		
154	Sol	25	8.490	3.00E-03	3.00E-03	3.00E-03	3.80E-01	3.15E-01	1.80E-01	7.21E-01	6.31E-01	2.70E-01
185	Sol	250	8.450	3.00E-03	2.00E-03	2.00E-03	3.80E-01	2.70E-01	2.70E-01	4.95E-01	4.05E-01	4.80E-01
185	Rod			<2.00E-02			7.21E+00			3.87E+01		
185	Rinse	800		<1.00E-03			4.80E-01			2.70E-01		
185	Strip	300		<1.00E-03			1.13E+00			4.19E+00		

DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			Sr-90			Np-237		
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A
1	Sol	5	7.932	1.05E+03								
6	Sol	10	8.060	1.12E+03	1.07E+03	7.16E+02						
6	Rod			<1.04E+03								
15	Sol	5	8.128	1.14E+03								
30	Sol	15	8.253	9.28E+02	9.59E+02	5.54E+02						
30	Rod											
82	Sol	20	8.308	9.32E+02	1.01E+03	7.25E+02				4.80E-01		
82	Rod			<1.04E+03								
120	Sol	20	8.424	7.39E+02	7.75E+02	6.35E+02				4.80E-01	4.80E-01	4.80E-01
120	Rod			1.87E+03								
181	Sol	250	8.440	8.20E+02	8.71E+02	4.92E+02				4.80E-01	4.25E-01	4.25E-01
181	Rod			2.32E+03								
181	Rinse	800		<1.04E+02						4.80E-01		
181	Strip	300		2.42E+02						4.25E-01		

CYCLE 2, TEST RESTARTED IN FRESH J-13 WATER

20	Sol	20	8.480	<1.22E+02	<1.04E+02	<1.06E+02				4.25E-01	4.25E-01	4.25E-01
20	Rod			<1.04E+03								
71	Sol	20	8.480				8.59E-01			4.25E-01	4.25E-01	4.25E-01
71	Rod			<1.04E+03								
154	Sol	25	8.490	1.11E+02						1.82E+02		
185	Sol	250	8.480	7.18E+01			9.74E-01	9.91E-02		4.25E-01	4.25E-01	4.25E-01
185	Rod			8.29E+02						4.80E-01		
185	Rinse	800								4.25E-01		
185	Strip	300								4.25E-01		

UNITS: Solution samples (Sol) in pCi/ml for all but Uranium, ug/ml for Uranium.
 Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.
 Rinse in same units as solution samples, sample was rinsed in 800 ml J-13 water.
 Strip in same units as solution samples, test vessel was stripped with 300 ml 8M HNO3.

APPENDIX B

SOLUTION CHEMISTRY

TABLE B.1

SOLUTION CHEMISTRY* FOR THE
C5C-H H. B. ROBINSON BARE FUEL TEST

	Cycle 1				Cycle 2		
	Starting J-13 Water	30 Days	120 Days	223 Days	Starting J-13 Water	154 Days	202 Days
pH	7.2	8.2	8.5	8.5	8.0	8.2	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**	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

*Units in µg/ml, 0.4 µm filtered.

**Low value attributed to analytical error.

TABLE B.2

SOLUTION* CHEMISTRY FOR THE
C5C-E H. B. ROBINSON SLIT DEFECT TEST

	Starting J-13 Water	Cycle 1			Cycle 2		
		30 Days	120 Days	223 Days	Starting J-13 Water	154 Days	202 Days
pH	7.2	8.4	8.5	8.5	8.0	8.2	8.55
Al	0.11	0.11	0.10	<0.08	<0.08	<0.08	<0.08
B	<0.10	<0.01	0.12	0.26	0.21	0.25	0.26
Ca	15.0	12.7	11.9	12.8	11.2	12.4	13.1
Fe	--	0.82	0.68	0.57	<0.01	<0.01	<0.01
K	5.5	8.3	4.5	2.9	1.95**	5.6	5.0
Mg	2.1	1.9	1.9	2.1	0.93	2.0	2.1
Mo	0.08	0.06	0.03	<0.02	<0.02	0.085	0.089
Na	49.5	40.4	41.4	46.5	43.1	43.0	45.2
Si	31.9	22.0	23.6	31.0	30.0	31.4	33.4
Sr	--	--	--	0.061	0.04	0.061	0.065
Cl	7.3	7.1	6.3	7.2	7.4	7.8	7.4
F	2.7	2.4	2.1	2.2	2.3	2.4	2.1
PO ₄	2.8	--	--	--	--	--	--
NO ₂	--	~2.7	~1.9	~1.4	--	~1.6	~1.8
NO ₃	8.7	4.5	5.6	5.7	8.3	6.6	5.7
SO ₄	18.8	18.2	18.3	18.2	18.6	18.7	19.0
CO ₃	118.0	--	123	122	121.5	115	114.5

*Units in µg/ml, 0.4 µm filtered.

**Low value attributed to analytical error.

TABLE B.3

SOLUTION* CHEMISTRY FOR THE
CSC-C H. B. ROBINSON HOLE DEFECTS TEST

	Starting J-13 Water	Cycle 1			Starting J-13 Water	Cycle 2	
		30 Days	120 Days	223 Days		154 Days	202 Days
pH	7.2	8.2	8.56	8.5	8.0	8.20	8.54
Al	0.11	0.09	0.08	<0.08	<0.08	<0.08	<0.08
B	<0.10	<0.01	0.05	0.19	0.21	0.21	0.20
Ca	15.0	13.0	12.0	12.8	11.2	12.7	13.0
Fe	--	0.44	0.32	0.30	<0.01	<0.01	<0.01
K	5.5	4.8	2.7	<0.5	1.95**	4.2	6.4
Mg	2.1	1.9	2.0	2.1	0.93	2.0	2.0
Mo	0.08	0.05	<0.02	<0.02	<0.02	<0.02	<0.02
Ka	49.5	44.2	44.4	43.5	43.1	41.4	43.6
Si	31.9	23.6	24.5	30.1	30.0	33.6	32.4
Sr	--	--	--	0.042	0.04	0.04	0.042
Cl	7.3	7.0	7.5	7.4	7.4	7.8	7.3
F	2.7	2.4	2.2	2.2	2.3	2.4	2.1
PO ₄	2.8	--	--	--	--	--	--
NO ₂	--	~2.8	~2.3	~1.9	--	~2.1	~3.2
NO ₃	8.7	4.4	5.4	5.5	8.3	6.4	5.7
SO ₄	18.8	18.2	18.7	18.5	18.6	19.2	19.3
CO ₃	118.0	--	122	118.5	121.5	113	113.5

*Units in µg/ml, 0.4 µm filtered.

**Low value attributed to analytical error.

TABLE B.4

SOLUTION* CHEMISTRY FOR THE
C5C-A H. B. ROBINSON UNDEFECTED TEST

	Cycle 1				Cycle 2		
	Starting J-13 Water	30 Days	120 Days	223 Days	Starting J-13 Water	154 Days	202 Days
pH	7.2	8.08	8.53	8.43	8.0	8.15	8.52
Al	0.11	0.08	<0.08	<0.08	<0.08	<0.08	<0.08
B	<0.10	<0.01	0.06	0.22	0.21	0.20	0.18
Ca	15.0	12.7	12.3	13.0	11.2	12.9	13.0
Fe	--	0.06	<0.01	<0.01	<0.01	0.024	<0.01
K	5.5	3.4	3.5	3.9	1.95**	5.8	6.7
Mg	2.1	2.1	2.0	2.1	0.93	2.1	2.1
Mo	0.08	0.04	<0.02	<0.02	<0.02	<0.02	<0.02
Na	49.5	40.9	47.7	46.5	43.1	42.3	41.3
Si	31.9	23.4	26.2	31.0	30.0	32.4	32.5
Sr	--	--	--	0.04	0.04	0.041	0.041
Cl	7.3	7.1	7.0	7.5	7.4	7.7	7.2
F	2.7	2.3	2.2	2.2	2.3	2.4	2.1
PO ₄	2.8	--	--	--	--	--	--
NO ₂	--	~3.4	~3.4	~2.8	--	~1.8	~2.0
NO ₃	8.7	3.6	4.3	4.5	8.3	6.5	5.7
SO ₄	18.8	18.1	18.6	19.1	18.6	19.8	21.6
CO ₃	118.0	--	121	120.5	121.5	113	114.5

*Units in µg/ml, 0.4 µm filtered.

**Low value attributed to analytical error.

TABLE B.5

SOLUTION* CHEMISTRY FOR THE
I-9-24 TURKEY POINT BARE FUEL TEST

	Cycle 1			Cycle 2		
	Starting J-13 Water	30 Days	181 Days	Starting 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**	4.7	4.8
Hg	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

*Units in µg/ml, 0.4 µm filtered.

**Low value attributed to analytical error.

TABLE B.6

SOLUTION* CHEMISTRY FOR THE
I-9-19 TURKEY POINT SLIT DEFECT TEST

	Cycle 1			Cycle 2		
	Starting J-13 Water	30 Days	18 [†] Days	Starting J-13 Water	154 Days	195 Days
pH	7.2	8.32	8.47	8.0	8.50	8.48
Al	0.11	<0.08	0.16	<0.08	<0.08	<0.08
B	<0.10	<0.01	0.19	0.21	0.21	0.16
Ca	15.0	11.6	12.6	11.2	12.6	12.0
Fe	--	0.32	0.26	<0.01	0.01	<0.01
K	5.5	5.1	11.7	1.95**	5.9	4.2
Mg	2.1	2.0	2.0	0.93	2.1	2.1
Mo	0.08	<0.02	0.115	<0.02	0.03	0.03
Na	49.5	48.8	47.2	43.1	44.1	43.5
Si	31.9	31.0	32.3	30.0	31.7	28.8
Sr	--	--	0.041	0.04	0.041	0.039
Cl	7.3	6.0	7.4	7.4	7.4	7.1
F	2.7	2.3	2.2	2.3	2.4	2.2
PO ₄	2.8	--	--	--	--	--
NO ₂	--	--	~1.6	--	~1.3	~1.2
NO ₃	8.7	3.8	5.9	8.3	7.9	6.8
SO ₄	18.8	19.9	18.8	18.6	19.0	18.7
CO ₃	118.0	--	123	121.5	119	118

*Units in µg/ml, 0.4 µm filtered.

**Low value attributed to analytical error.

TABLE B.7
SOLUTION* CHEMISTRY FOR THE
I-9-12 TURKEY POINT HOLE DEFECTS TEST

	Cycle 1			Cycle 2		
	Starting J-13 Water	30 Days	181 Days	Starting J-13 Water	154 Days	195 Days
pH	7.2	8.31	8.48	8.0	8.51	8.54
Al	0.11	<0.08	0.15	<0.08	<0.08	<0.08
B	<0.10	<0.01	0.19	0.21	0.19	0.15
Ca	15.0	11.7	13.0	11.2	12.8	11.8
Fe	--	0.19	0.19	<0.01	0.017	<0.01
K	5.5	5.1	5.6	1.95**	6.7	5.4
Mg	2.1	1.9	2.0	0.93	1.9	2.1
Mo	0.08	<0.02	0.027	<0.02	<0.02	<0.02
Na	49.5	50.0	46.4	43.1	43.9	43.6
Si	31.9	29.9	34.1	30.0	32.3	30.5
Sr	--	--	0.041	0.04	0.041	0.039
Cl	7.3	6.0	7.4	7.4	7.8	7.2
F	2.7	2.3	2.2	2.3	2.4	2.2
PO ₄	2.8	--	--	--	--	--
NO ₂	--	--	~1.8	--	~1.5	~1.4
NO ₃	8.7	4.5	5.6	8.3	7.9	6.5
SO ₄	18.8	19.7	18.8	18.6	19.5	18.9
CO ₃	118.0	--	114.5	121.5	118	117

*Units in µg/ml, 0.4 µm filtered.

**Low value attributed to analytical error.

TABLE B.8

SOLUTION* CHEMISTRY FOR THE
I-9-1 TURKEY POINT UNDEFECTED TEST

	Cycle 1			Cycle 2		
	Starting J-13 Water	30 Days	181 Days	Starting J-13 Water	154 Days	195 Days
pH	7.2	8.25	8.44	8.0	8.49	8.46
Al	0.11	<0.08	0.14	<0.08	<0.08	<0.08
B	<0.10	<0.01	0.21	0.21	0.22	0.16
Ca	15.0	12.6	12.6	11.2	12.3	12.2
Fe	--	0.34	0.34	<0.01	<0.01	<0.01
K	5.5	4.7	9.3	1.95**	6.0	6.0
Mg	2.1	1.9	2.0	0.93	2.1	2.2
Mo	0.08	<0.02	0.035	<0.02	<0.02	<0.02
Na	49.5	47.6	46.8	43.1	43.1	45.6
Si	31.9	32.1	33.5	30.0	32.1	29.1
Sr	--	--	0.041	0.04	0.042	0.038
Cl	7.3	6.2	7.5	7.4	7.4	7.2
F	2.7	2.4	2.2	2.3	2.4	2.2
PO ₄	2.8	--	--	--	--	--
NO ₂	--	~2.4	~1.9	--	~1.8	~1.9
NO ₃	8.7	7.1	5.6	8.3	6.1	6.1
SO ₄	18.8	21.1	19.7	18.6	19.0	18.9
CO ₃	118.0	--	121	121.5	118	119

*Units in µg/ml, 0.4 µm filtered.

**Low value attributed to analytical error.

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