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**RESULTS FROM NNWSI SERIES 1  
SPENT FUEL LEACH TESTS**

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C.N. Wilson

Date Published: May 1985

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ABSTRACT

*Pressurized water reactor spent fuel rod segments containing small artificially induced cladding defects were leach tested in a test matrix that also included rod segments with undefected cladding and rod segments which were split open to fully expose the bare fuel. Results from the first test series conducted in deionized water under air at ambient hot cell temperature are presented and discussed.*

*The amounts of measured fuel dissolution were two to three orders of magnitude less for fuel contained in artificially defected cladding than for the bare fuel removed from the cladding. Post-test microstructural examination of fuel particles from the bare fuel test revealed significant grain boundary dissolution. Total measured fractional release relative to inventory was calculated for several radionuclides. The actinides, plutonium, americium, curium, and probably neptunium, appeared to be released congruently with uranium. Fission products cesium and technetium were both preferentially released relative to their inventory.*

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Radiochemical analyses described in this report were performed by, or under the direction of, A. C. Leaf of the HEDL Chemistry and Analysis Section. These analyses are a major portion of the results reported.

Spent fuel test specimens were prepared by N. 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 C. E. Saari, R. T. Steele and D. V. Archer. Specimen preparation, testing, and examination were directed 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

|       |  |
|-------|--|
| BCL   | Battelle Columbus Laboratories                     |
| DOE   | Department of Energy                               |
| EDX   | Energy-Dispersive X-Ray                            |
| HEDL  | Hanford Engineering Development Laboratory         |
| ICP   | Inductively Coupled Plasma (Emission Spectrometry) |
| LLNL  | Lawrence Livermore National Laboratory             |
| LMFBR | Liquid Metal Fast Breeder Reactor                  |
| LWR   | Light Water Reactor                                |
| NNWSI | Nevada Nuclear Waste Storage Investigations        |
| ORNL  | Oak Ridge National Laboratory                      |
| PNL   | Pacific Northwest Laboratory                       |
| PWR   | Pressurized Water Reactor                          |
| SEM   | Scanning Electron Microscope                       |
| WHC   | Westinghouse Hanford Company                       |

## RESULTS FROM NNWSI SERIES 1 SPENT FUEL LEACH TESTS

### 1.0 BACKGROUND

The US Department of Energy (DOE), through the Office of Civilian Radioactive Waste Management, is actively studying the technical feasibility of permanent disposal of high-level nuclear waste in geologic formations. A schedule for siting, licensing, and construction of a geologic repository is established by the Nuclear Waste Policy Act of 1982.<sup>(1)</sup> Principal geologic formations under consideration for the first repository include tuff, salt, and basalt. The Nevada Nuclear Waste Storage Investigations (NNWSI) Project is investigating the suitability of the Topopah Spring Member of the Paintbrush Tuff at Yucca Mountain, Nye County, Nevada for development of a repository. Lawrence Livermore National Laboratory (LLNL) is the lead contractor for the Waste Package Task in support of the NNWSI Project. LLNL has chosen Westinghouse Hanford Company (WHC) as a subcontractor to assist them in determining the requirements for successful disposal of spent fuel rods in the Yucca Mountain Site.

A primary objective of the NNWSI Waste Package Task is to determine if and how the Nuclear Regulatory Commission requirements contained in 10 CFR 60<sup>(2)</sup> can be satisfied. Additional applicable standards are being prepared by the US Environmental Protection Agency in 40 CFR 191.<sup>(3)</sup> Two principal waste package 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."

The principal mechanism for release of radionuclides from a failed waste package is leaching of the radionuclides into groundwater and migration as a result of groundwater flow. The candidate repository horizon for the NNWSI Project is a welded devitrified tuff above the water table. However, a limited amount of water infiltrating the rock may provide a potential transport mechanism for radionuclides to the underlying water table. Contact between this water and the waste package is not expected to occur during the first several hundred years when the repository temperature is greater than the 95°C boiling temperature for water at the repository elevation. After the thermal period, one scenario for water contact with stored spent fuel is the collection of a near static volume of water in a failed waste package. The condition of stored fuel rods at this time will be difficult to determine. Many of the rods may remain intact, some rods may contain breaches but not be catastrophically failed, and others may be broken or more severely degraded.

The objective of the Spent Fuel Leaching/Dissolution Tests being conducted by WHC in support of NNWSI is to gain a better understanding of probable spent fuel behavior and radionuclide release under NNWSI-proposed repository conditions. The initial approach is to perform "semi-static" leach tests with specimens representing a range of fuel physical degradation under conditions applicable to NNWSI. The Series 1 tests described in this report were conducted in deionized water. The Series 2 tests are similar except that they use NNWSI J-13 well water.<sup>(4)</sup> Higher temperature is planned for the Series 3 tests. Future tests using fuel that has been degraded by air/steam oxidation and testing with different fuel types are also planned.

## 2.0 TEST DESCRIPTION

Details of test specimen preparation, testing, and sample analysis are given in the test plan.<sup>(5)</sup>

### 2.1 TEST SPECIMENS

Four specimen types, representing a range of potential states of spent fuel rod degradation under NNWSI-proposed conditions, were leach-tested in deionized water under air at ambient hot cell temperature ( $\sim 25^{\circ}\text{C}$ ). The four specimen types were:

- Spent fuel rod segments with water-tight end fittings and undefected cladding.
- Fuel rod segments with water-tight end fittings and laser-drilled holes through the cladding.
- Fuel rod segments with water-tight end fittings and a sawed slit through the cladding.
- Bare fuel removed from a split fuel rod segment plus the split cladding hulls.

Test specimens used for the Series 1 deionized water tests were prepared from Turkey Point Unit 3 pressurized water reactor (PWR) spent fuel rods. These fuel segments had been previously sectioned at Battelle Columbus Laboratories (BCL) under air, packaged in pipe plug-sealed metal tubes, and stored at Hanford Engineering Development Laboratory (HEDL, operated by WHC for DOE) until the present specimens were prepared during June 1983. Fuel rod sectioning diagrams and characterization data are contained in Reference 6. Relevant fuel characteristics are summarized in Tables 1 and 2. Identification and descriptions for all Series 1 test specimens are given in Table 3.

Test specimen radionuclide inventories and spent fuel composition used in the evaluation of results in this report were calculated from ORIGEN-2 data for 10-year old spent PWR fuel compiled in Appendix E of PNL-5109.<sup>(7)</sup> Calculation of specimen radionuclide inventories is described in Appendix A.1.

TABLE 1  
FUEL CHARACTERISTICS

|                        | <u>Turkey Point</u>       |
|------------------------|---------------------------|
| Fuel Type              | PWR 15 x 15               |
| Discharge Date         | November 1975             |
| Nominal Burnup         | 26 MWd/kgM                |
| Fission Gas Release    | 0.3%                      |
| Initial Enrichment     | 2.55% <sup>235</sup> U    |
| Rod Diameter           | 10.7 mm OD                |
| Cladding Thickness     | 0.62 mm                   |
| Cladding Material      | Zircaloy-4                |
| Initial Pellet Density | 92% TD (UO <sub>2</sub> ) |

Radionuclide inventory estimates for 1000-year old spent fuel are based on ORIGEN data contained in ORNL/TM-6008.<sup>(8)</sup>

## 2.2 CLADDING SURFACE CONTAMINATION

A primary purpose of testing fuel rod specimens with undefected cladding was to provide an estimate for that portion of released radionuclides originating from the cladding exterior surface in tests with defected cladding. Two types of cladding exterior surface activity were present on the specimens: 1) contamination picked up during postirradiation sectioning and handling in contaminated hot cells, and 2) "crud" deposits picked up in-reactor during irradiation. The surface contamination picked up during postirradiation sectioning and handling is primarily fine particulate material much of which can be wiped off. A large amount of this type of postirradiation surface contamination should not be expected on whole rods received at the repository. Crud deposits are more adherent and are expected to occur on fuel received for repository storage. However, much of the crud deposit activity (i.e., <sup>60</sup>Co) is relatively short lived in comparison to the repository waste package containment period.

TABLE 2  
CALCULATED SPENT FUEL SPECIMEN BULK COMPOSITION\*

| <u>Element</u>   | <u>wt%</u>    | <u>Element</u> | <u>wt%</u>    |
|------------------|---------------|----------------|---------------|
| Se               | 0.0047        | U              | 84.808        |
| Br               | 0.0016        | Np             | 0.0309        |
| Kr               | 0.0256        | Pu             | 0.7399        |
| Rb               | 0.0254        | Am             | 0.0492        |
| Sr               | 0.0548        | Cm             | <u>0.0013</u> |
| Y                | 0.0324        |                |               |
| Zr               | 0.2615        | Actinides      | 85.629        |
| Mo               | 0.2456        |                |               |
| Tc               | 0.0574        | Oxygen         | <u>11.852</u> |
| Ru               | 0.1640        |                |               |
| Rh               | 0.0366        | TOTAL          | 99.995        |
| Pd               | 0.1084        |                |               |
| Ag               | 0.0063        |                |               |
| Cd               | 0.0086        |                |               |
| In               | 0.0002        |                |               |
| Sn               | 0.0071        |                |               |
| Sb               | 0.0016        |                |               |
| Te               | 0.0368        |                |               |
| I                | 0.0181        |                |               |
| Xe               | 0.3934        |                |               |
| Cs               | 0.1788        |                |               |
| Ba               | 0.1263        |                |               |
| La               | 0.0894        |                |               |
| Ce               | 0.1730        |                |               |
| Pr               | 0.0820        |                |               |
| Nd               | 0.2950        |                |               |
| Pm               | 0.0008        |                |               |
| Sm               | 0.0597        |                |               |
| Eu               | 0.0096        |                |               |
| Gd               | 0.0086        |                |               |
| Tb               | 0.0002        |                |               |
| Dy               | <u>0.0001</u> |                |               |
| Fission Products | 2.514         |                |               |

\*Elements comprising less than 0.0001% (1 ppm) of composition have been omitted.  
Reference 7 (PNL-5109), 10-year data interpolated to 27.7 Mwd/kgU burnup, corrected for oxygen weight.

TABLE 3  
SERIES 1 TEST SPECIMENS

| <u>Identification*</u> | <u>Description</u>                                  | <u>Test Duration</u>            |
|------------------------|---|---------------------------------|
| H-6-19                 | Bare fuel plus split cladding hulls                 | 250 days, restarted<br>128 days |
| H-6-12                 | Slit defect ~150 $\mu\text{m}$ wide by 2 cm long    | 252 days, restarted<br>128 days |
| J-8-24                 | Two laser-drilled holes ~200 $\mu\text{m}$ diameter | 244 days, restarted<br>128 days |
| J-8-19                 | One laser-drilled hole ~200 $\mu\text{m}$ diameter  | 60 days                         |
| H-6-1                  | Undefected  | 252 days, restarted<br>128 days |
| H-6-24**               | Undefected  | 271 days                        |
| J-8-12                 | Undefected  | 60 days                         |

\*All specimens from Turkey Point Unit 3 assembly B17 rods H-6 and J-8, sections numbered from bottom of rod, see HEDL-TME 80-85<sup>(6)</sup> for sectioning diagrams.

\*\*Test H-6-24 was contaminated with ~60 ppm  $\text{H}_2\text{SO}_4$  and pH was ~3.0 for duration of test.

Because of the expected low solubility of the fuel matrix in deionized water, an initial concern was test result bias from radionuclides leached from the cladding exterior contamination picked up during postirradiation sectioning and handling. To avoid this potential problem, specimen cladding exterior surfaces were cleaned several times by wiping with alcohol-dampened wipes. Initial cotton swab smears from the cladding surface read 8000 cpm to 20,000 cpm alpha and 7 mR/h to 15 mR/h beta/gamma. The cladding of the bare fuel and slit defect specimens were decontaminated to a few hundred cpm smearable alpha before slitting in a low contamination hot cell. Final decontamination and assembly of the undefected, laser-drilled, and slit

defect specimens was performed in a clean hot cell. Final cladding smears on these specimens read 20 cpm to 50 cpm alpha with beta/gamma levels undetectable above the laboratory background.

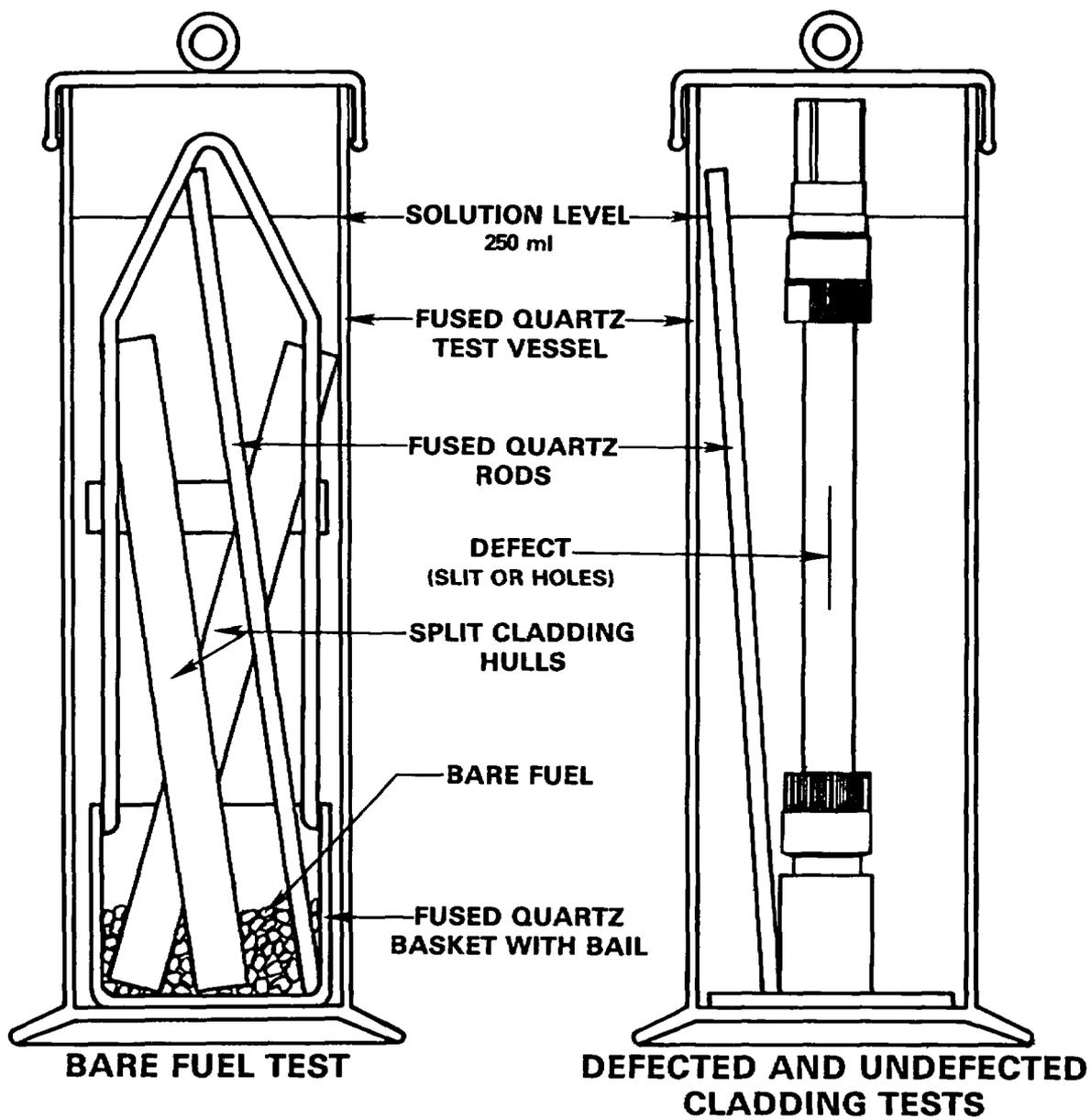
### 2.3 TEST APPARATUS

The test specimen configurations shown in Figure 1 were used. The sectioned ends of the laser-drilled, slit and undefected specimens were sealed using specially fabricated 316 stainless steel fittings incorporating ethylene propylene O-rings compressed against the cladding by a compression nut. The top end fitting contained a small vent hole above the solution level allowing the internal free volume of the defected cladding test specimen to fill with solution up to the external solution level. Each test was started by adding 250 ml of deionized water. Each test vessel also initially contained six fused quartz rods that could be periodically removed and analyzed for radionuclide plate-out. The tests were run in the HEDL 325 Building Shielded Analytical Laboratory.

### 2.4 TESTING AND SAMPLING

The tests were of a "semi-static" nature, in that only small portions of the test solutions were periodically sampled and replenished. The semi-static test allowed monitoring of solution characteristics with time while allowing the bulk of the solution to form a "stabilized" system with the test specimen and apparatus surfaces. This type of test is representative of NNWSI proposed conditions, where a finite volume of water with a very low flow rate would contact a potentially failed waste package during the post-containment and post-thermal period.

Solution samples of 10-ml volume were taken using a fresh pipette attached to a syringe for each sample. Constant solution volume was maintained by adding fresh deionized water for each sample removed. Approximate solution sampling schedule was to sample at days 1, 5, 15, 30, 60, 90, 120, 180 and 240. Fused



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FIGURE 1. Test Apparatus.

quartz rod samples were taken approximately at days 5, 30, 60, 120, 180 and 240. The rod samples were briefly rinsed with fresh deionized water to remove loose particulate or colloidal material, and then immersed in a fused quartz tube filled with 10 ml of 8 M HNO<sub>3</sub>. The 8 M HNO<sub>3</sub> was then analyzed for radionuclides that "plated out" on the fused quartz rods.

After the terminal solution and rod samples were taken, the test specimens were removed and rinsed with fresh deionized water. The bare fuel, the bare fuel baskets, the 316 stainless steel test specimen pedestals, and the fused quartz test vessels were also briefly rinsed with deionized water to remove loose particulate or colloidal particles. The pedestals and bare fuel baskets were placed back into their respective test vessels and each vessel filled with 300 ml of 8 M HNO<sub>3</sub> to strip off radionuclide plate-out.

The following resulting sample types were submitted for analysis:

- Periodic test solution samples
- Periodic 8 M HNO<sub>3</sub> rod strip solutions
- Final vessel 8 M HNO<sub>3</sub> strip solutions\*
- Bare fuel rinse solution

The four longest term tests, one of each specimen type (see Table 2), were then restarted in fresh deionized water. Both solution and rod samples were taken from the "second run" tests on days 15, 70 and 128. The second run tests were terminated on day 128, the components rinsed with fresh deionized water, and stripped with 8 M HNO<sub>3</sub> as before. The final test specimens were then sectioned, mounted, and examined ceramographically for evidence of fuel corrosion or other microstructural effects.

---

\*The pedestals and test vessels were acid stripped separately in the J-8-12 and J-8-19 tests.

## 2.5 SAMPLE ANALYSES

### 2.5.1 Radionuclides of Interest

Based on 10 CFR 60 requirements, significant radionuclides present in 1000-year old spent fuel are those of most interest for the current studies. By 1000 years, most of the shorter lived fission product activity (i.e.,  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{90}\text{Sr}$ , etc.), which represents a majority of the activity in the 10-year old fuel tested, has decayed out. Most of this activity decays out during the 300-year minimum containment period during which time "substantially complete" waste package integrity is required. Table 4 gives ORIGEN-calculated relative inventories for radionuclides with half-life greater than one year and relative activity greater than that of  $^{129}\text{I}$  for a 1000-year old 33,000 Mwd/MTM burnup PWR spent fuel. It should be noted from Table 4 that  $^{129}\text{I}$  activity is an order of magnitude below that given for  $^{238}\text{U}$ , which originates from the ore removed from the earth to originally fabricate the fuel. Approximately 98% of the calculated 1000-year activity is due to Am and Pu isotopes, with an additional  $\sim 0.77\%$  originating from  $^{99}\text{Tc}$  decay.  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$ ,  $^{94}\text{Nb}$ ,  $^{14}\text{C}$  and  $\sim 8\%$  of the  $^{93}\text{Zr}$  are light element activation products.  $^{14}\text{C}$  activity, in particular, may vary since it originates primarily from activation of as-fabricated fuel and cladding nitrogen impurities, which are variable and not well characterized.

### 2.5.2 Radionuclides Analyzed

All samples were analyzed by alpha spectrometry, gamma spectrometry, and laser-excited fluorescence (for uranium). The pH was also measured on all solution samples as-sampled from the test vessels. After pH determination and subsequent filtering (for later test solution samples), sample fractions were acidified with ultra-high purity  $\text{HNO}_3$  to stabilize the samples and prevent plate-out. Alpha spectrometry gave results for  $^{239}\text{Pu}$  plus  $^{240}\text{Pu}$ ,  $^{238}\text{Pu}$  plus  $^{241}\text{Am}$  and  $^{244}\text{Cm}$  activities. Gamma spectrometry gave results for

TABLE 4

## PWR SPENT FUEL ASSEMBLY RADIONUCLIDE INVENTORIES AT 1000 YEARS(a)

| <u>Radionuclide</u> <sup>(b)</sup> | <u>% of Total<br/>1000-Year<br/>Activity</u> | <u>Cumulative %</u> |
|------------------------------------|--|---------------------|
| Am-241                             | 51.84  | 51.84               |
| Am-243                             | 1.75(c)                                      | 53.59               |
| Pu-240                             | 26.87  | 80.46               |
| Pu-239                             | 17.37  | 97.83               |
| Pu-242                             | 0.10   | 97.93               |
| Pu-238                             | 0.06   | 97.99               |
| Tc-99                              | 0.77   | 98.76               |
| Ni-59                              | 0.252  |                     |
| Ni-63                              | 0.021  |                     |
| Zr-93                              | 0.181  |                     |
| Nb-94                              | 0.074  |                     |
| C-14                               | 0.076(d)                                     |                     |
| U-234                              | 0.113  |                     |
| U-238                              | 0.018  |                     |
| U-236                              | 0.015  |                     |
| Np-237                             | 0.058  |                     |
| Sn-126                             | 0.045  |                     |
| Se-79                              | 0.023  |                     |
| Cs-135                             | 0.022  |                     |
| Sm-151                             | 0.013  |                     |
| Pd-107                             | 0.006  |                     |
| I-129                              | 0.0018                                       |                     |

(a) Based on ORIGEN data reported in ORNL/TM-6008(8) for 33,000 MW<sub>e</sub>/MTM burnup PWR Assembly.

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

(c) Includes activity of <sup>239</sup>Np daughter products.

(d) <sup>14</sup>C activity may vary considerably depending on as-fabricated nitrogen impurities.

$^{137}\text{Cs}$  and  $^{134}\text{Cs}$  on all samples measured and for  $^{60}\text{Co}$  for most test solution samples. Other activities measured by gamma spectrometry samples included  $^{125}\text{Sb}$ ,  $^{106}\text{Ru}$ ,  $^{144}\text{Ce}$  and  $^{154}\text{Eu}$ .

Selective separations and analyses were performed on later samples for  $^{99}\text{Tc}$ ,  $^{237}\text{Np}$ ,  $^{79}\text{Se}$ ,  $^{14}\text{C}$  and  $^{129}\text{I}$ . Aliquots for  $^{14}\text{C}$  and  $^{129}\text{I}$  analysis were removed from solution sample fractions before acidification. Am-Pu separations were performed on later samples to determine  $^{241}\text{Am}$  activities. The  $^{238}\text{Pu}$  to  $^{239}\text{Pu}$  plus  $^{240}\text{Pu}$  activity ratios were determined in order to calculate  $^{241}\text{Am}$  activity from  $^{241}\text{Am}$  plus  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  plus  $^{240}\text{Pu}$  activities for early samples from three tests. As testing progressed, sensitivity of some of the radiochemical methods was improved by increasing counting times and using larger sample volumes for specific counting source preparations. A comparison of estimated detection limits, and levels in solution if  $10^{-5}$  of specimen inventory were dissolved in the 250 ml of test solution, are given in Table 5.

### 2.5.3 Filtering

After the 180-day initial run samples from the four longest term tests, solution samples were split and filtered to give unfiltered, 0.4  $\mu\text{m}$  filtered (Nuclepore stock number 110407 polycarbonate disc filters), and 18  $\text{\AA}$  filtered (Amicon Corporation Model CTS-1 membrane cone centrifuge filters) fractions. Analyses for radionuclides that routinely produced "below detectable limits" results ( $^{79}\text{Se}$ ,  $^{14}\text{C}$ ,  $^{129}\text{I}$ ) were performed only on unfiltered fractions. After filtering, all sample fractions were then stabilized by adding  $\sim 2$  vol% high purity  $\text{HNO}_3$  to prevent loss of radionuclides from solution. ( $^{14}\text{C}$  and  $^{129}\text{I}$  sample aliquots taken before acidification.)

TABLE 5  
RADIOCHEMISTRY METHODS

| Radionuclide                           | Method  | Detection Limits |        | 10 <sup>-5</sup> Inventory<br>(pCi/ml)* |
|--|---|------------------|--------|---|
|  |   | (pCi/ml)         | (ppb)  |   |
| <sup>241</sup> Am                      | α-spectrometry<br>following separation                            | 2                | 0.0006 | 2400                                    |
| <sup>238</sup> Pu                      | α-spectrometry<br>following separation                            | 2                | 0.0001 | 3200                                    |
| <sup>239</sup> Pu<br><sup>240</sup> Pu | α-spectrometry  | 2                | 0.03   | 1200                                    |
| <sup>237</sup> Np                      | α-spectrometry<br>following separation                            | 0.2**            | 0.3    | 0.3                                     |
| <sup>137</sup> Cs                      | γ-spectrometry  | 1000             | 0.01   | 1.0 x 10 <sup>5</sup>                   |
| <sup>129</sup> I                       | Liquid scintillation<br>counting following<br>separation          | 100              | 560    | 0.04                                    |
| <sup>129</sup> I                       | Neutron activation<br>analysis                                    | 10 <sup>-5</sup> | 0.0001 | 0.04                                    |
| <sup>99</sup> Tc                       | β-proportional<br>counting following<br>separation                | 20**             | 1.2    | 16                                      |
| <sup>79</sup> Se                       | Liquid scintillation<br>counting following<br>separation          | 100              | 1.4    | 0.5                                     |
| <sup>14</sup> C                        | Liquid scintillation<br>following CO <sub>2</sub><br>distillation | 100              | 0.02   | --                                      |
| U                                      | Fluorescence  | --               | 1      | (1.8 ppm)                               |

\*Assumes 10<sup>-5</sup> of test specimen inventory released to 250 ml.  
\*\*Using 3-ml sample and 100 minutes of counting time.

### 3.0 RESULTS AND DISCUSSION

A complete tabulation of radiochemical results reported as pCi activities ( $\mu\text{g/ml}$  for U) is contained in the Appendix. Tables of conversion factors and instruction for conversion from pCi activities to isotopic and elemental concentrations are also contained in the Appendix.

#### 3.1 URANIUM RELEASE

Uranium measured in unfiltered solution samples is plotted in Figure 2A. The open data points connected by lines in Figure 2A are for the initial test runs on each specimen. The solid data points are for the second runs in which each specimen was restarted in fresh deionized water. The  $10^{-5}$  inventory line is the concentration that would result in solution if  $10^{-5}$  of the uranium in the test specimen were dissolved in the 250-ml deionized water test solution. Approximately one order of magnitude more uranium was measured in unfiltered solution samples from the bare fuel test than from the slit defect test during the first  $\sim 200$  days, and approximately two orders magnitude greater uranium was measured in the bare fuel test relative to the laser-drilled test during this time. About 200 days after initial test start, uranium began dropping out of solution and was reduced to 1 ppb to 3 ppb levels in all unfiltered solution samples by 250 days. When the tests were restarted with fresh deionized water, nearly identical behavior was observed except that the uranium dropped out sooner.

Filtered and unfiltered solution uranium levels for the last samples from the initial test runs and the second test runs are given in Table 6. These data suggest that actual uranium solubility in these tests was  $\sim 0.001 \mu\text{g/ml}$  (1 ppb). An initial supersaturation may have occurred early in the tests accounting for the slightly higher  $0.007 \mu\text{g/ml}$  and  $0.006 \mu\text{g/ml}$  18 A filtered uranium levels, respectively, in the 15-day second run samples from the slit and hole defect tests. With time, the colloids apparently become unstable and settle out. A much higher solubility limit of  $\sim 50 \text{ ppm}$  has been

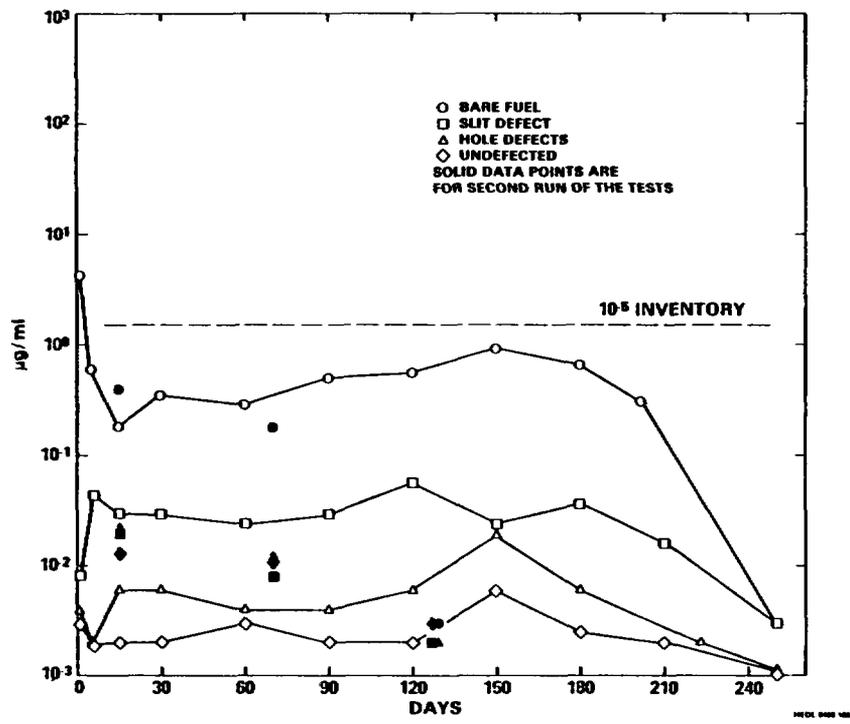


FIGURE 2A. Uranium in Unfiltered Solution.

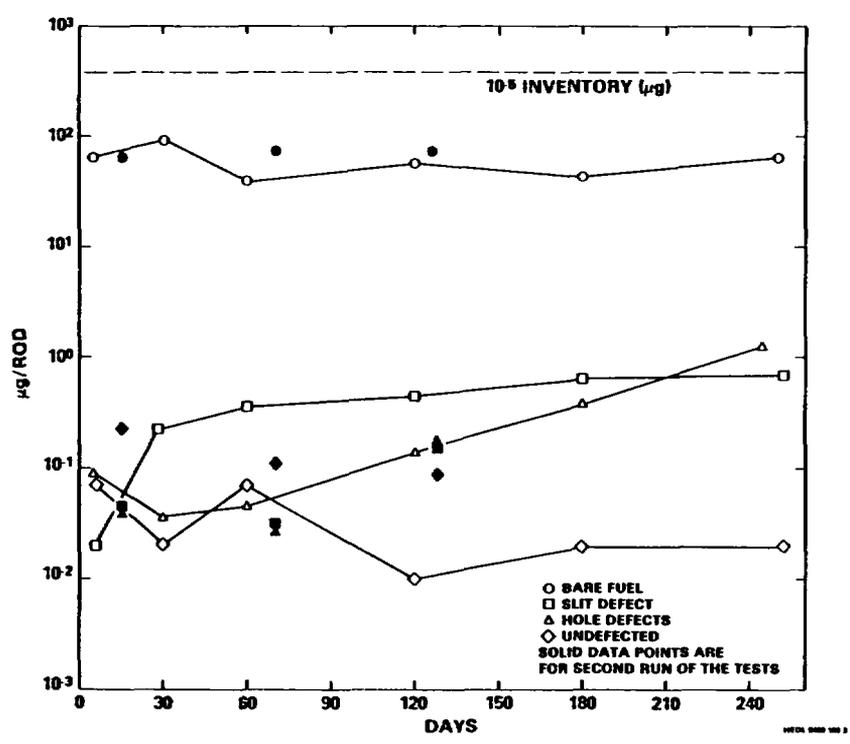


FIGURE 2B. Uranium on Fused Quartz Rod Samples.

TABLE 6

## URANIUM MEASURED IN UNFILTERED AND FILTERED SOLUTION SAMPLES

| Days                   | Filter            | Uranium ( $\mu\text{g/ml}$ )* |        |       |            |
|------------------------|-------------------|-------------------------------|--------|-------|------------|
|                        |                   | Bare Fuel                     | Slit   | Holes | Undelected |
| 202                    | Unfiltered        | 0.30                          | --     | --    | --         |
|                        | 0.4 $\mu\text{m}$ | 0.003                         | --     | --    | --         |
|                        | 18 A              | 0.002                         | --     | --    | --         |
| ~250                   | Unfiltered        | 0.003                         | 0.003  | 0.002 | 0.002      |
|                        | 0.4 $\mu\text{m}$ | <0.001                        | <0.001 | 0.002 | <0.001     |
|                        | 18 A              | <0.001                        | <0.001 | 0.002 | <0.001     |
| <u>Second Test Run</u> |                   |                               |        |       |            |
| 15                     | Unfiltered        | 0.40                          | 0.020  | 0.023 | 0.013      |
|                        | 0.4 $\mu\text{m}$ | 0.004                         | 0.012  | 0.011 | 0.005      |
|                        | 18 A              | 0.001                         | 0.007  | 0.006 | 0.003      |
| 70                     | Unfiltered        | 0.18                          | 0.008  | 0.011 | 0.011      |
|                        | 0.4 $\mu\text{m}$ | 0.004                         | 0.003  | 0.004 | 0.004      |
|                        | 18 A              | 0.001                         | 0.001  | 0.002 | 0.002      |
| 128                    | Unfiltered        | 0.003                         | 0.002  | 0.002 | 0.003      |
|                        | 0.4 $\mu\text{m}$ | 0.003                         | 0.002  | 0.002 | 0.003      |
|                        | 18 A              | 0.001                         | 0.001  | 0.002 | 0.002      |

\*Best reported detection limit was 0.001  $\mu\text{g/ml}$  (1 ppb).

predicted for U in NNWSI J-13 well water.<sup>(9)</sup> The higher solubility is attributed to uranium complexing by carbonate in the J-13 water.\*

The filters used to filter the 202-day solution sample from the bare fuel test were examined by scanning electron microscope (SEM) and SEM-EDX (energy-dispersive x-ray). Nothing was found on the filter surfaces. The ~1  $\mu\text{g}$  of

\*Uranium measured in initial samples from Series 2 bare fuel tests with J-13 water ranged from ~2 to 5 ppm. Essentially all of the uranium in the initial Series 2 samples passed both the 0.4  $\mu\text{m}$  and 18 A filters.

uranium removed from this solution sample by the 0.4- $\mu\text{m}$  filter was apparently too diffuse for detection in the SEM.

The pH during all but one of the tests remained in a range of 5.4 to 6.7. One test with undefected cladding was initially contaminated with  $\sim 60$  ppm  $\text{H}_2\text{SO}_4$ . The pH in this test ranged from  $\sim 2.9$  to 3.3. Uranium, most likely from cladding surface contamination, was about 130 ppb for most unfiltered solution samples from this test. Filtering of the last two samples from this test (250 and 271 days) indicated that most of this uranium was in true solution (110 ppb U in both 18 A filtered samples versus 130 ppb before filtration).

Uranium measured on fused quartz rod samples (Figure 2B) correlated with the unfiltered uranium solution data. Rod samples from the bare fuel test showed much more uranium plate-out than did rod samples from the other three test types, which exhibited much less apparent colloidal uranium in the "solution phase." Most of the plate-out apparently occurred before the first rod samples were taken at 5 days in the initial tests and at 15 days in the second run tests. The  $10^{-5}$  inventory line in the Figure 2B plot is  $10^{-5}$  of the total uranium inventory in the initial test specimens (each specimen contained  $\sim 40$  g of uranium).

An accounting of the measured released uranium for the various sample types taken is given in Table 7. Two primary observations from the Table 7 data are: 1) total measured uranium release from the bare fuel test was much greater than from the slit or holes defected tests, and 2) only a very small fraction of the total measured uranium release was in solution. Total measured uranium releases from the slit and holes defected specimens were only  $\sim 4$  and 2 times, respectively, the release measured from the undefected specimen. Total measured release from the bare fuel specimen was about 360 times that measured with the slit defect specimen and about 700 times that measured with the holes defect specimen. Greater release during the initial test run may be due to early attack and depletion of higher activity material such as exposed grain boundaries, fine particles, or minute exposed secondary phases (i.e., Cs-U-O phases).

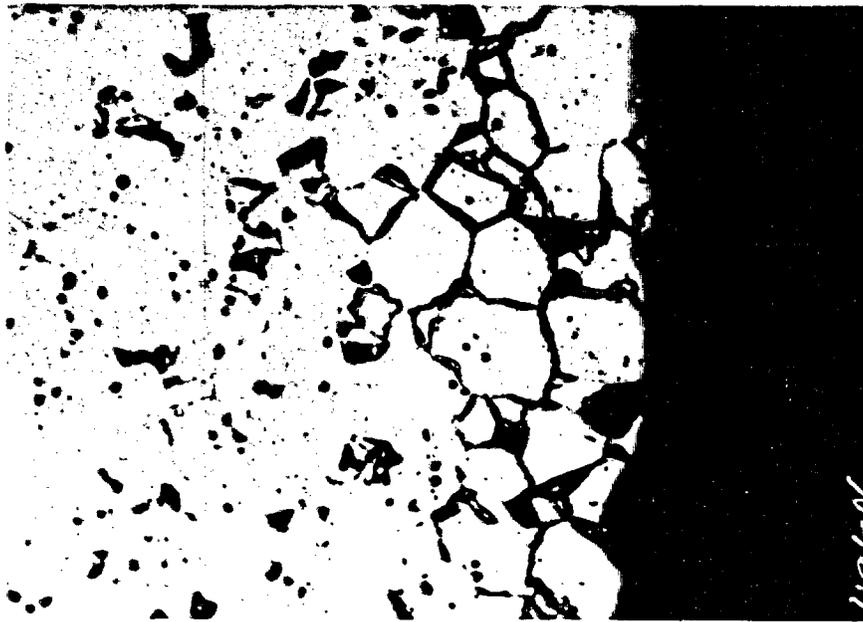
TABLE 7  
URANIUM RELEASE DATA ( $\mu\text{g}$ )

|                                  | <u>Bare Fuel</u> | <u>Slit Defect</u> | <u>Holes Defect</u> | <u>Undefected</u> |
|----------------------------------|------------------|--------------------|---------------------|-------------------|
| <u>Initial Test Run</u>          |                  |                    |                     |                   |
| $\Sigma$ Solution Samples*       | 87               | 2.92               | 0.60                | 0.27              |
| $\Sigma$ Rod Samples             | 363              | 2.44               | 2.00                | 0.19              |
| Final Solution                   | 0.75             | 0.75               | 0.25                | 0.50              |
| Test Vessel Strip                | 6300             | 17.40              | 8.40                | 2.70              |
| Bare Fuel Rinse**                | 800              | --                 | --                  | --                |
| <u>Second Test Run</u>           |                  |                    |                     |                   |
| $\Sigma$ Solution Samples*       | 6                | 0.28               | 0.34                | 0.24              |
| $\Sigma$ Rod Samples             | 213              | 0.96               | 0.85                | 0.43              |
| Final Solution                   | 0.75             | 0.50               | 0.50                | 0.75              |
| Test Vessel Strip                | 1530             | 3.00               | <1.50               | <1.50             |
| Bare Fuel Rinse**                | 210              | --                 | --                  | --                |
| <u>Total Measured Release</u>    |                  |                    |                     |                   |
| $\Sigma$ Above ( $\mu\text{g}$ ) | 9510             | 28.25              | <14.44              | <6.58             |
| Divided by $10^{-5}$ Inventory   | 28.0             | 0.078              | <0.0406             | <0.0185           |

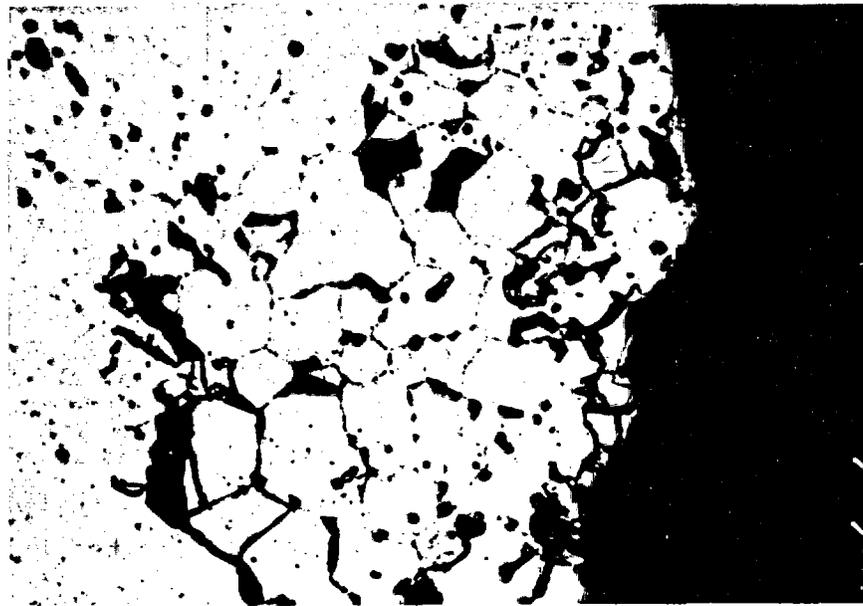
\*Last solution sample is included in "Final Solution" value.

\*\*Bare fuel was ultrasonically cleaned in 500 ml deionized water when the run was terminated to produce the "Rinse" sample.

Post-test ceramographic examination of bare fuel particles from the H-6-19 test showed clearly visible corrosion of the particles. As shown in Figure 3, the attack occurred preferentially at grain boundaries. Grain boundaries, not visible in pre-test "as-polished" ceramographic sections, were resolved up to  $100 \mu\text{m}$  in from the particle surfaces. Another indication of the grain boundary attack was the amount of grain "pull out" during ceramographic section preparation observed near the bare fuel particle surfaces. The total measured release of  $\sim 0.028\%$  of the uranium in the bare fuel test corresponds to an equivalent uniform corrosion depth of  $\sim 0.1 \mu\text{m}$  (assuming 2-mm diameter spherical particles). This amount of total fuel dissolution is consistent with the post-test particle appearance in ceramographic sections.



30  $\mu\text{m}$



30  $\mu\text{m}$

FIGURE 3. As-Polished Ceramographic Section of Fuel Particles from H-6-19 Bare Fuel Test Showing Grain Boundary Corrosion.

An as-polished section through one of the two J-8-24 specimen laser-drilled hole defects is shown in Figure 4A. The fuel surface  $\sim 45^\circ$  from the laser-drilled hole is shown in Figure 4B. The  $0.04 \times 10^{-5}$  of inventory total measured release from this specimen corresponds to an estimated equivalent uniform corrosion depth of only 2 Å from the specimen internal fuel particle surfaces. No visible evidence of fuel dissolution at the fuel cladding gap or along fuel cracks was observed in this specimen. Similarly, little evidence of fuel dissolution was observed in the fuel-cladding gap or along tight cracks in a post-test ceramographic section of the H-6-12 slit defect specimen. However, one apparent intergranular attack area was observed at the slit defect location (Figure 5B), and areas of corroded-appearing fuel particle surfaces were observed (Figure 6) adjacent to a large void (1 to 2 mm) on the opposite side of the specimen from the slit. The void area shown in Figure 6 probably resulted from fuel movement when the specimens were originally prepared at BCL. (At this time, the fuel column was pushed up and down in the cladding to remove  $\sim 1$  in. of fuel from each end of the specimens.) Intergranular penetrations equivalent to those observed on bare fuel test particles were not observed in the slit defect section.

### 3.2 PLUTONIUM RELEASE

The Turkey Point fuel tested is  $\sim 0.8\%$   $\text{PuO}_2$ , which is thought to be in solid solution with  $\text{UO}_2$ . At 1000 years, Pu isotopes account for  $\sim 44\%$  of spent fuel Ci activity as calculated by ORIGEN.<sup>(8)</sup> At 10,000 years,  $^{239}\text{Pu}$  plus  $^{240}\text{Pu}$  accounts for  $\sim 90\%$  of the activity. Plutonium release was evaluated using  $^{239}\text{Pu}$  plus  $^{240}\text{Pu}$  activity levels. Activity of these isotopes in unfiltered solution samples is plotted as a function of sampling time in Figure 7A. Plutonium plate-out on fused quartz rods is shown in Figure 7B. Total measured Pu release and the distribution of measured release between different sample types is given in Table 8.

Most of the Pu activity measured in unfiltered solution was also measured in  $0.4 \mu\text{m}$  and  $18 \text{ Å}$  filtered solution sample fractions. The few percent of the Pu activity lost on the filters appeared to be independent of solution Pu

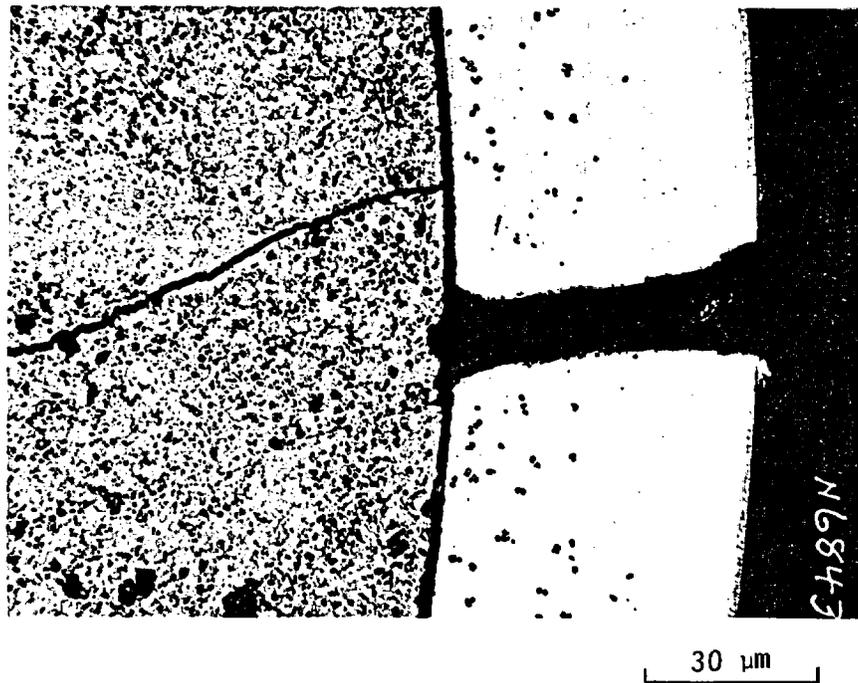


FIGURE 4A. Laser-Drilled Defect in Test Specimen J-8-24 (As-Polished).



FIGURE 4B. Fuel Surface at Fuel-Cladding Gap Near the Laser-Drilled Defect Showing No Visible Evidence of Fuel Dissolution (As-Polished).

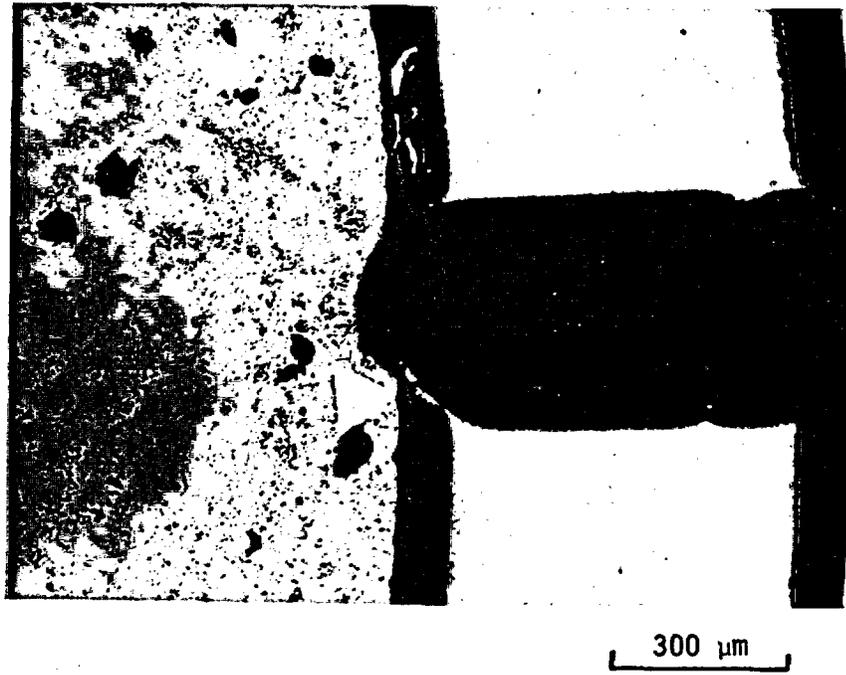


FIGURE 5A. Slit-Defect in Test Specimen H-6-12.

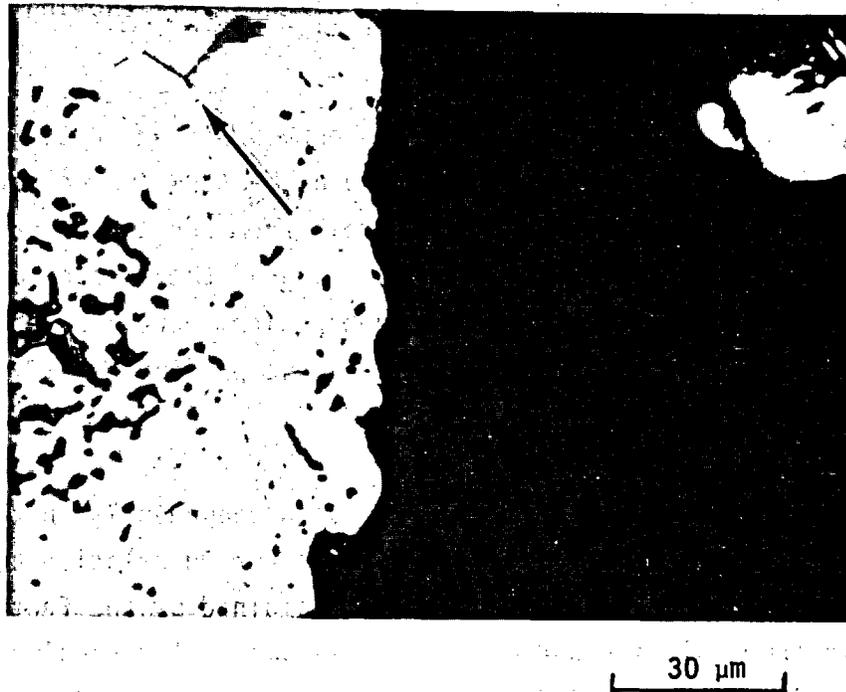


FIGURE 5B. As-Polished Fuel Surface Near the Slit Defect Showing Visible Intergranular Attack.

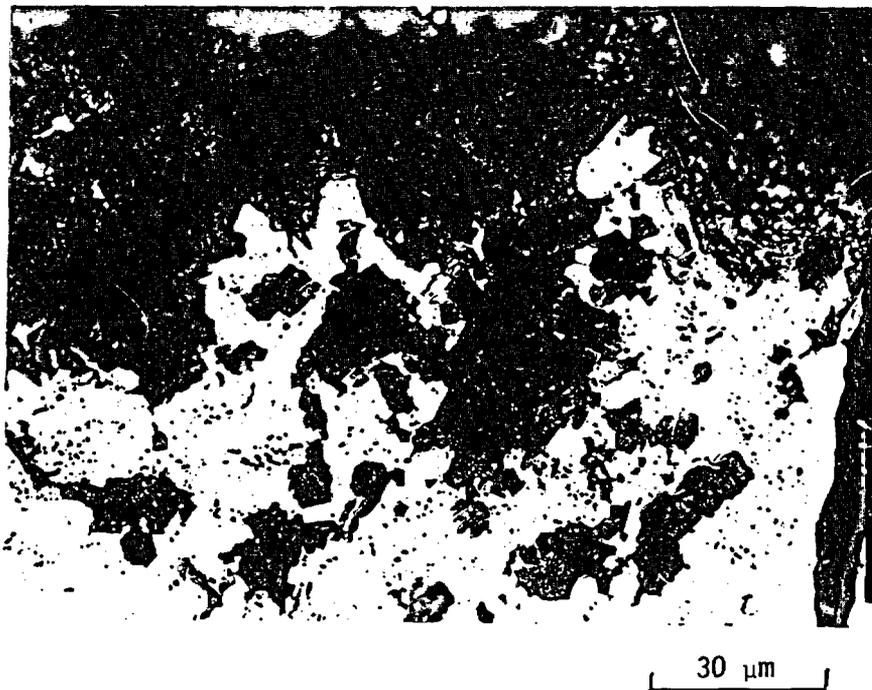


FIGURE 6. Void Area in Slit Defect Specimen H-6-12 Showing Corroded Fuel Surface Appearance (As-Polished).

content, suggesting the loss may have been due to adsorption by the filters or some other systematic artifact. The highest Pu concentration measured was 46 ppb for the 15-day solution sample from the second run of the bare fuel test. Without knowledge of the nature of the "plate-out" phases, it is difficult to know if measured Pu concentrations were controlled by a solubility limit. However, 46 ppb is less than the predicted ~430 ppb solubility for Pu in NNWSI groundwater.<sup>(9)</sup>

Uranium and Pu appear to have been released congruently in the bare fuel test. Greater apparent fractional release for Pu relative to U in the slit defect and holes defect tests could be explained by the fact that released Pu was in true solution and could diffuse out of the specimen much easier than U, which formed colloids. As with U, measured Pu fractional release from the bare fuel specimen was 2 to 3 orders of magnitude greater than from

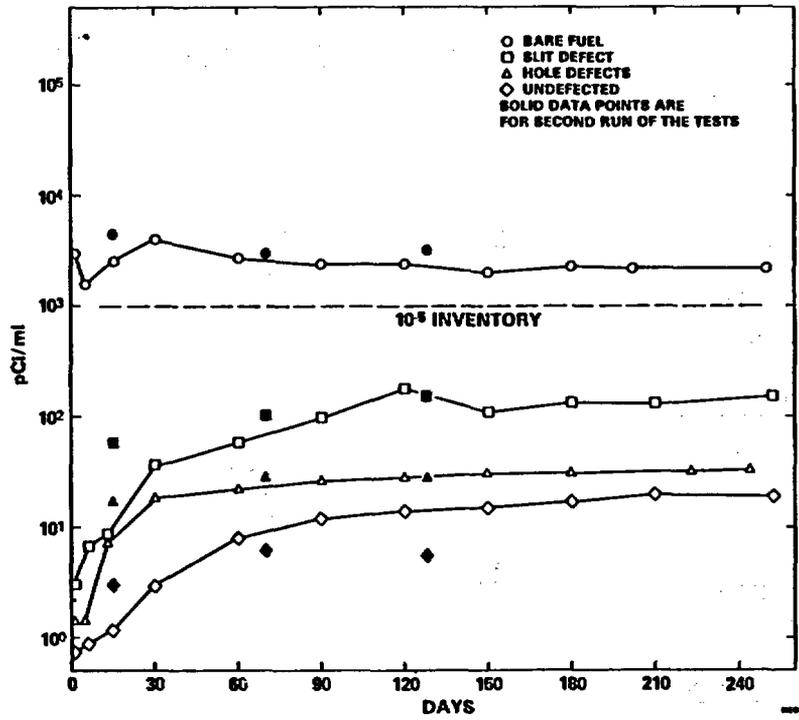


FIGURE 7A. <sup>239</sup>Pu Plus <sup>240</sup>Pu in Unfiltered Solution.

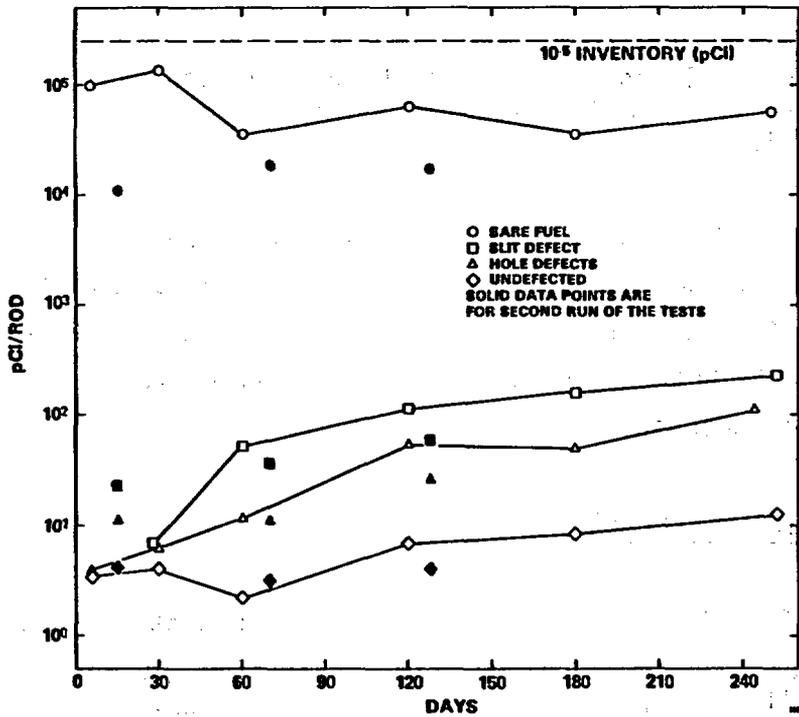


FIGURE 7B. <sup>239</sup>Pu Plus <sup>240</sup>Pu on Fused Quartz Rod Sample.

TABLE 8  
 $^{239}\text{Pu}$  PLUS  $^{240}\text{Pu}$  RELEASE DATA (nCi)

|                                | <u>Bare Fuel</u> | <u>Slit Defect</u> | <u>Holes Defect</u> | <u>Undefected</u> |
|--------------------------------|------------------|--------------------|---------------------|-------------------|
| <u>Initial Test Run</u>        |                  |                    |                     |                   |
| $\Sigma$ Solution Samples*     | 250              | 7.6                | 1.98                | 0.96              |
| $\Sigma$ Rod Samples           | 430              | 0.6                | 0.24                | 0.04              |
| Final Solution                 | 540              | 38.5               | 8.33                | 4.84              |
| [Pu (ppb)]**                   | (23.1)           | (1.64)             | (0.356)             | (0.207)           |
| Test Vessel Strip              | 4750             | 13.2               | 2.03                | 0.46              |
| Bare Fuel Rinse                | 370              | --                 | --                  | --                |
| <u>Second Test Run</u>         |                  |                    |                     |                   |
| $\Sigma$ Solution Samples*     | 70               | 1.6                | 0.45                | 0.09              |
| $\Sigma$ Rod Samples           | 50               | 0.1                | 0.05                | 0.01              |
| Final Solution                 | 860              | 37.6               | 7.09                | 1.46              |
| [Pu (ppb)]**                   | (36.7)           | (1.60)             | (0.304)             | (0.062)           |
| Test Vessel Strip              | 340              | 4.6                | 0.46                | 0.12              |
| Bare Fuel Rinse                | 280              | --                 | --                  | --                |
| <u>Total Measured Release</u>  |                  |                    |                     |                   |
| $\Sigma$ Above (nCi)           | 7940             | 103.8              | 20.63               | 7.98              |
| Divided by $10^{-5}$ Inventory | 27.96            | 0.341              | 0.0690              | 0.0267            |

\*Last solution sample is included in "Final Solution" value.

\*\*Calculated plutonium (all isotopes) concentration in the fuel unfiltered solution sample in ppb ( $\mu\text{g/g}$ ).

the slit defect and holes defect specimens. The amount of both U and Pu release measured in the holes defect test may not be significant in comparison to release from the undefected specimen, since the differences observed could also be attributed to variations in the effectiveness of pretest specimen surface decontamination.

### 3.3 AMERICIUM RELEASE

The activity of  $^{241}\text{Am}$  in spent fuel increases substantially during the first ~100 years after reactor discharge as a result of  $^{241}\text{Pu}$  beta decay (13-year half-life).  $^{241}\text{Am}$ , with a 458-year half-life, accounts for approximately half of the total Ci activity in 1000-year old spent fuel. Americium was

separated by anion exchange and  $^{241}\text{Am}$  activity was determined by alpha counting plus alpha spectrometry in the later samples from the four longest term tests. The  $^{238}\text{Pu}/(^{239}\text{Pu} + ^{240}\text{Pu})$  ratios were radiochemically determined for samples from the bare fuel, slit defect, and holes defect specimens. This ratio was then used to calculate  $^{241}\text{Am}$  from  $^{239}\text{Pu} + ^{240}\text{Pu}$  and  $^{241}\text{Am} + ^{238}\text{Pu}$  data for the remaining initial run samples from these tests. Calculated  $^{241}\text{Am}$  values for initial run samples from the H-6-1 undefected cladding test were not obtained because the required alpha activities in this test were too low to obtain reliable results.

The calculated and measured  $^{241}\text{Am}$  activities in unfiltered solution samples are plotted in Figure 8A. The overall  $^{241}\text{Am}$  release data are given in Table 9. These data suggest that Am was released congruently with U and Pu. The observed differences in fractional total measured release between Am and Pu and U could be explained by the apparently very low solubility of Am in these tests and the probability that colloidal or plate-out Am was not totally accounted for. Congruent  $^{241}\text{Am}$  release with U and Pu is consistent with its expected state as an oxide in solid solution with the  $\text{UO}_2$  fuel matrix.

Most of the measured  $^{241}\text{Am}$  release was in the test vessel strip solutions. The highest  $^{241}\text{Am}$  activities measured radiochemically in unfiltered and 0.4- $\mu\text{m}$  filtered bare fuel test solution samples were  $\sim 14$  nCi/ml ( $\sim 5$  ppb) in the 15-day second run unfiltered sample and  $\sim 3.7$  nCi/ml ( $\sim 1.3$  ppb) in the 128-day second run 0.4- $\mu\text{m}$  filtered sample. The highest  $^{241}\text{Am}$  activity measured in 18 A filtered solution samples was  $\sim 100$  pCi/ml ( $\sim 35$  ppt). However, Am absorption by the 18 A membrane filters may account for the much lower  $^{241}\text{Am}$  activities measured in the 18 A filtered samples. Am solubility of  $\sim 2.4$  ppb has been predicted<sup>(9)</sup> in NNWSI groundwater.

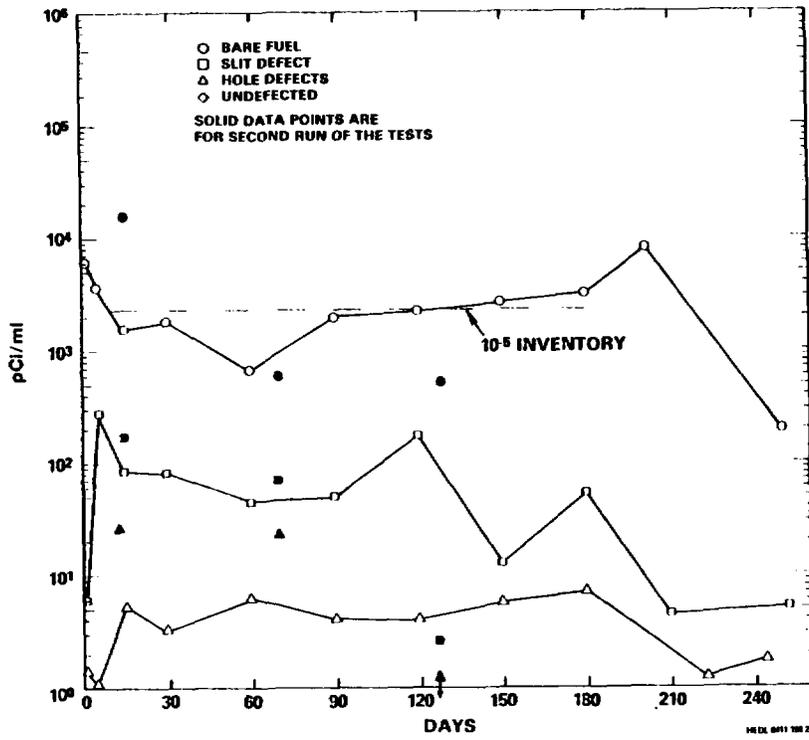


FIGURE 8A. <sup>241</sup>Am in Unfiltered Solution.

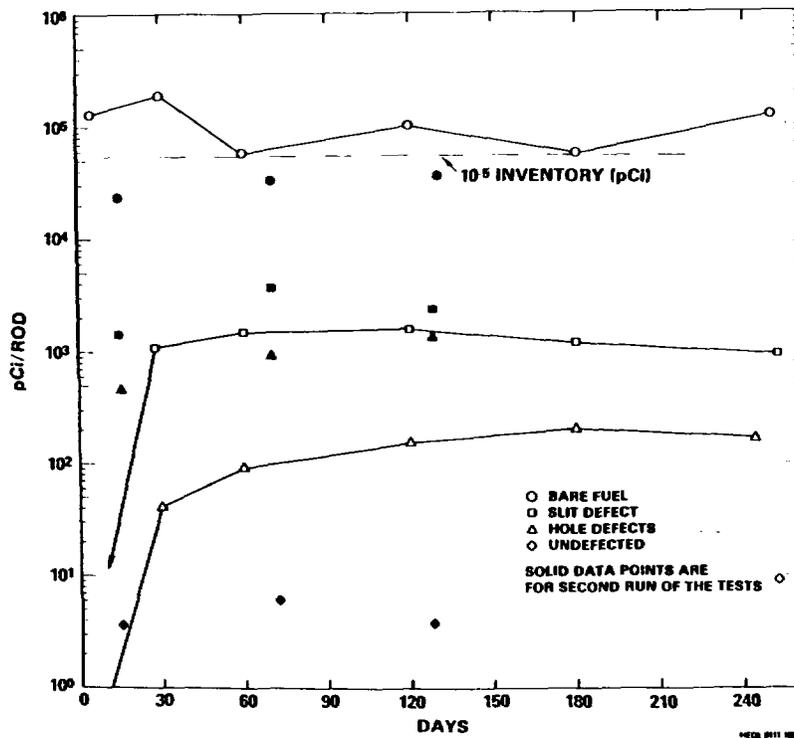


FIGURE 8B. <sup>241</sup>Am on Fused Quartz Rod Samples.

TABLE 9  
<sup>241</sup>Am RELEASE DATA (nCi)

|                                       | <u>Bare Fuel</u> | <u>Slit Defect</u> | <u>Holes Defect</u> | <u>Undefected</u> |
|---------------------------------------|------------------|--------------------|---------------------|-------------------|
| <u>Initial Test Run</u>               |                  |                    |                     |                   |
| Σ Solution Samples*                   | 336              | 7.92               | 0.400               | --                |
| Σ Rod Samples                         | 660              | 6.36               | 0.668               | --                |
| Final Solution                        | 49               | 1.58               | 0.450               | <0.225            |
| [Am (ppt)]**                          | (70)             | (2.3)              | (0.64)              | (<0.32)           |
| Test Vessel Strip                     | 8490             | 77.97              | 9.865               | 4.595             |
| Bare Fuel Rinse                       | 430              | --                 | --                  | --                |
| <u>Second Test Run</u>                |                  |                    |                     |                   |
| Σ Solution Samples*                   | 197              | 2.43               | 0.505               | 0.095             |
| Σ Rod Samples                         | 92               | 7.58               | 2.851               | 0.309             |
| Final Solution                        | 1273             | 0.56               | <0.338              | <0.338            |
| [Am (ppt)]**                          | (1822)           | (0.81)             | (<0.50)             | (<0.50)           |
| Test Vessel Strip                     | 552              | 26.08              | 3.514               | 0.811             |
| Bare Fuel Rinse                       | 525              | --                 | --                  | --                |
| <u>Total Measured Release</u>         |                  |                    |                     |                   |
| Σ Above (nCi)                         | 12,604           | 130.48             | <18.591             | <6.373            |
| Divided by 10 <sup>-5</sup> Inventory | 21.7             | 0.208              | < 0.0302            | --                |

\*Last solution sample is included in "Final Solution" value.

\*\*Calculated Americium concentration (all isotopes) in the final unfiltered solution sample in ppt (ng/g).

### 3.4 CURIUM RELEASE

Relative to Pu and Am isotopes, curium activity from spent fuel will probably not be significant during the repository post-containment period. However, <sup>244</sup>Cm, with a half-life of 17.6 years, represents a significant portion of the total actinide activity in the ~8-year old spent fuel used in these tests. <sup>244</sup>Cm activity measured in unfiltered solution samples is plotted in Figure 9A, and <sup>244</sup>Cm activity measured on fused quartz rod samples is plotted in Figure 9B. The overall measured <sup>244</sup>Cm release data are summarized in Table 10.

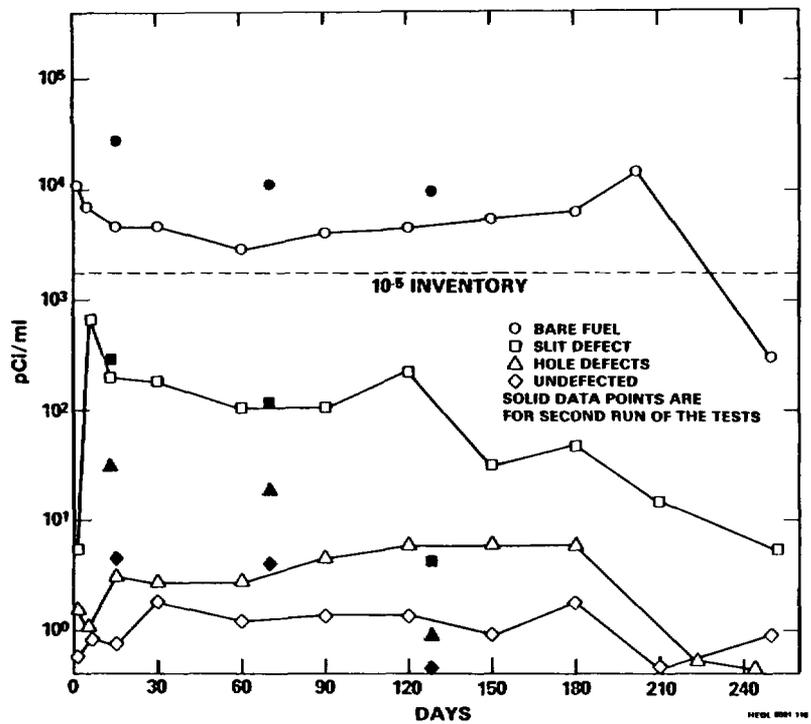


FIGURE 9A.  $^{244}\text{Cm}$  in Unfiltered Solution.

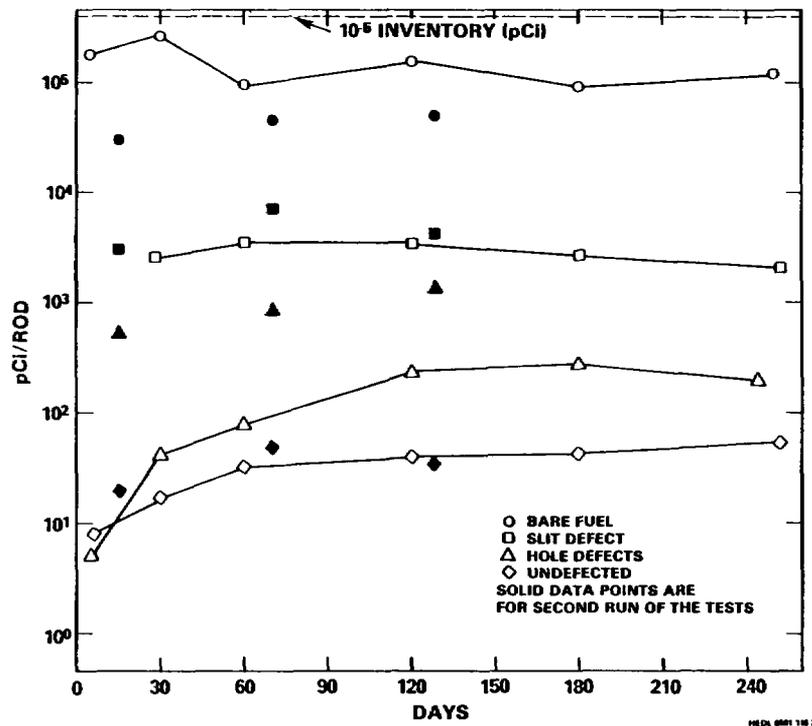


FIGURE 9B.  $^{244}\text{Cm}$  on Fused Quartz Rod Samples.

TABLE 10  
<sup>244</sup>Cm RELEASE DATA (nCi)

|                                       | <u>Bare Fuel</u> | <u>Slit Defect</u> | <u>Holes Defect</u> | <u>Undefected</u> |
|---------------------------------------|------------------|--------------------|---------------------|-------------------|
| <u>Initial Test Run</u>               |                  |                    |                     |                   |
| Σ Solution Samples*                   | 512              | 16.5               | 0.338               | 0.110             |
| Σ Rod Samples                         | 887              | 14.1               | 0.818               | 0.197             |
| Final Solution                        | 73               | 1.6                | 0.113               | <0.225            |
| [ <sup>244</sup> Cm (ppt)]**          | (3.5)            | (0.08)             | (0.005)             | (0.011)           |
| Test Vessel Strip                     | 7500             | 144.6              | 10.680              | 2.568             |
| Bare Fuel Rinse                       | 565              | --                 | --                  | --                |
| <u>Second Test Run</u>                |                  |                    |                     |                   |
| Σ Solution Samples*                   | 393              | 4.1                | 0.491               | 0.086             |
| Σ Rod Samples                         | 124              | 14.4               | 2.730               | 0.105             |
| Final Solution                        | 2487             | 1.0                | 0.225               | 0.113             |
| [ <sup>244</sup> Cm (ppt)]**          | (119)            | (0.05)             | (0.011)             | (0.005)           |
| Test Vessel Strip                     | 588              | 39.0               | 2.703               | 0.513             |
| Bare Fuel Rinse                       | 171              | --                 | --                  | --                |
| <u>Total Measured Release</u>         |                  |                    |                     |                   |
| Σ Above (nCi)                         | 13,300           | 235.3              | 18.098              | < 3.917           |
| Divided by 10 <sup>-5</sup> Inventory | 30.0             | 0.49               | 0.039               | < 0.008           |

\*Last solution sample is included in "Final Solution" Value.

\*\*<sup>244</sup>Cm concentration in the final unfiltered solution samples in ppt (ng/l).

A notable observation on <sup>244</sup>Cm data plotted in Figures 9A and 9B is its similarity to <sup>241</sup>Am data plotted in Figures 8A and 8B. Curium appears to have been released congruently with the actinides U, Pu and Am. (The lower apparent fractional U release relative to Pu, Am and Cm in the slit defect specimen may be explained if much of the U released from the fuel was tied up by colloid particles that did not move out of the specimen.) The highest <sup>244</sup>Cm activities measured in solution samples from the bare fuel test corresponded to ~330 ppt (15-day second run unfiltered sample), of which only ~0.14 ppt (~12 pCi/ml) passed the 18 A filter.

### 3.5 CESIUM RELEASE

$^{137}\text{Cs}$  and  $^{134}\text{Cs}$  account for  $\sim 20\%$  of the total Ci activity and most of the gamma activity in the  $\sim 8$ -year old spent fuel tested. At 1000 years, the only significant Cs activity remaining is  $^{135}\text{Cs}$ , which has a  $3 \times 10^6$  year half-life and accounts for  $\sim 0.02\%$  of total Ci activity at 1000 years.

$^{137}\text{Cs}$  activity measured in unfiltered solution samples is plotted in Figure 10A. Approximately 0.1% of the bare fuel specimen  $^{137}\text{Cs}$  inventory entered the solution on the first day of the initial test run, and  $\sim 0.2\%$  of the bare fuel  $^{137}\text{Cs}$  activity was in solution a few days later. The highest measured  $^{137}\text{Cs}$  solution sample activity in the bare fuel test corresponds to a 220 ppb concentration. After 60 days, the  $^{137}\text{Cs}$  activity in unfiltered solution over the bare fuel began to decline to levels less than that measured in the slit defect and holes defect tests. Much of the  $^{137}\text{Cs}$  that left the solution phase was not accounted for in the final vessel strip or bare fuel rinse solutions. Less  $^{137}\text{Cs}$  was measured in solution during the second run. However, Cs was still preferentially released relative to inventory during the second run, suggesting that not all of the "free" Cs (at fuel cladding interface, cracks and grain boundaries) had been released during the first run. As in the first run,  $^{137}\text{Cs}$  activity in solution over the bare fuel decreased with time during the second run of the test.

In filtered samples, the fraction of the  $^{137}\text{Cs}$  activity passing the 18  $\mu$  filter ranged from about 40% to 81%. Because of its high solubility, loss of Cs from solution with time or filtering was not expected. The fraction of Cs removed by filtering was independent of the amount in the unfiltered solution. The only correlation with the Cs filtering data noted was that a larger fraction of Cs tended to be removed by filtration of samples from which a large portion of the uranium was also removed by filtration. This suggests that a portion of the Cs may become associated with the colloidal uranium phase.

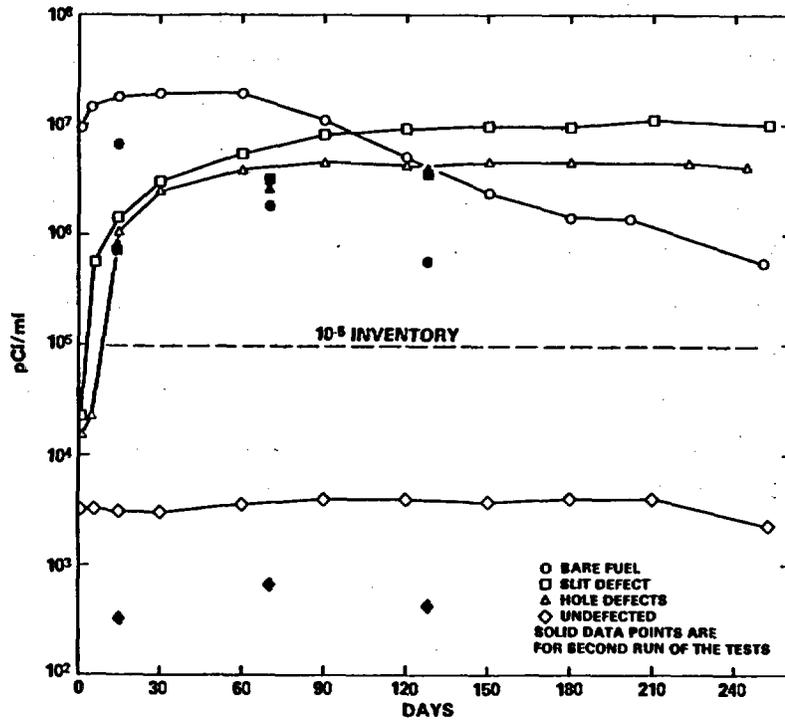


FIGURE 10A.  $^{137}\text{Cs}$  in Unfiltered Solution.

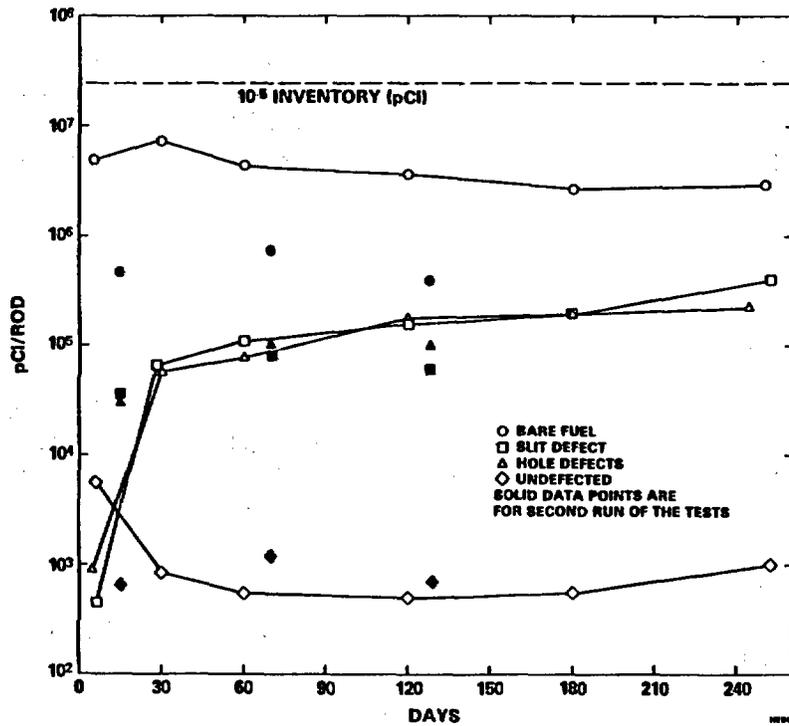


FIGURE 10B.  $^{137}\text{Cs}$  on Fused Quartz Rod Samples.

Cumulative measured  $^{137}\text{Cs}$  data for each test and sample type are given in Table 11. The 0.075% of inventory total measured released value given for the bare fuel test is known to be low since 0.2% of the specimen inventory was in solution at 30 days. Correcting for the  $^{137}\text{Cs}$  that dropped out of solution and was not accounted for in the terminal strip and rinse solutions, total  $^{137}\text{Cs}$  release in the bare fuel test is estimated to be  $\sim 7600 \mu\text{Ci}$  or  $\sim 0.3\%$  of inventory. Fission gas release for the H-6 Turkey Point rod from which the bare fuel specimen was prepared is reported to be 0.3%.<sup>(6)</sup> This agrees with results previously reported for CANDU fuel by Johnson et al.,<sup>(10)</sup> in which fast Cs release approximately equivalent to the fractional fuel fission gas release during irradiation was observed.

TABLE 11  
 $^{137}\text{Cs}$  RELEASE DATA ( $\mu\text{Ci}$ )

|                                   | <u>Bare Fuel</u> | <u>Slit Defect</u> | <u>Holes Defect</u> | <u>Undefected</u> |
|-----------------------------------|------------------|--------------------|---------------------|-------------------|
| <u>Initial Test Run</u>           |                  |                    |                     |                   |
| $\Sigma$ Solution Samples*        | 1003.6           | 579.73             | 296.77              | 0.363             |
| $\Sigma$ Rod Samples              | 25.4             | 2.10               | 1.64                | <0.020            |
| Final Solution                    | 129.5            | 2432.43            | 1033.78             | 0.538             |
| [ $^{137}\text{Cs}$ (ppb)]**      | (6.0)            | (112.9)            | (48.0)              | (0.025)           |
| Test Vessel Strip                 | 281.1            | 10.77              | 6.92                | 0.057             |
| Bare Fuel Rinse                   | 84.9             | --                 | --                  | --                |
| <u>Second Test Run</u>            |                  |                    |                     |                   |
| $\Sigma$ Solution Samples*        | 83.2             | 38.24              | 34.10               | 0.011             |
| $\Sigma$ Rod Samples              | 1.5              | 0.39               | 0.53                | <0.006            |
| Final Solution                    | 144.1            | 872.75             | 948.20              | 0.102             |
| [ $^{137}\text{Cs}$ (ppb)]**      | (6.7)            | (40.5)             | (44.0)              | (0.005)           |
| Test Vessel Strip                 | 19.3             | 2.36               | 4.98                | 0.019             |
| Bare Fuel Rinse                   | 165.3            | --                 | --                  | --                |
| <u>Total Measured Release</u>     |                  |                    |                     |                   |
| $\Sigma$ Above ( $\mu\text{Ci}$ ) | 1937.9           | 3938.8             | 2326.9              | <1.116            |
| Divided by $10^{-5}$ Inventory    | 75.1***          | 142.1              | 85.6                | 0.041             |

\*Last solution sample is included in "Final Solution" value.

\*\* $^{137}\text{Cs}$  concentration in the final unfiltered solution sample in ppb ( $\mu\text{g/g}$ ).

\*\*\*Estimated to be  $\sim 300 \times 10^{-5}$  of inventory ( $\sim 7600 \mu\text{Ci}$ ) based on highest solution activities measured.

### 3.6 TECHNETIUM RELEASE

Technetium-99 with a 213,000-year half-life, accounts for ~1% of the total Ci activity of spent fuel at 1000 years and ~3% at 10,000 years.<sup>(8)</sup> Release of <sup>99</sup>Tc from spent fuel is of particular interest since Tc compounds are predicted to be quite soluble in NNWSI groundwater.<sup>(9,11)</sup> As a pure beta emitter, radiochemical separation is required for <sup>99</sup>Tc measurement. Release of 10<sup>-5</sup> of the specimen inventory into solution in the current tests gives <sup>99</sup>Tc activity just sufficient to count at the estimated detection limit.

Technetium analyses were conducted on the final test solutions and the 8 M HNO<sub>3</sub> test vessel strip solutions at the ends of the initial and second test runs. Results from these analyses are given in Table 12.

The most significant observation from the limited <sup>99</sup>Tc release data obtained was that fractional release relative to inventory appears to be very high in comparison to that of the actinides. Essentially all of the <sup>99</sup>Tc measured in unfiltered solution samples passed through 0.4 μm and 18 Å filters, most likely in solution as TcO<sub>4</sub><sup>-</sup>. In the bare fuel test, <4% of the <sup>99</sup>Tc activity was found as plate-out in the vessel strip solutions. The fractional total measured release values (total measured release divided by 10<sup>-5</sup> of specimen inventory) for <sup>99</sup>Tc were 8.2, 35.5, and <98 times higher, respectively, for the bare fuel, slit defect, and holes defect tests than measured for plutonium. The same <sup>137</sup>Cs-to-Pu fractional release ratios were 2.7 (~11)\*, 416, and 1240, respectively, for the bare fuel, slit defect, and holes defect tests. Greater fractional release of Cs relative to Tc in the slit and holes defect tests versus the bare fuel test may be explained if much of the released Cs was from the fuel cladding gap or soluble phases at fuel particle surfaces, while preferential Tc release resulted from grain boundary dissolution, which occurred to a much greater extent in the bare fuel test.

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\*The Cs-to-Pu fractional release ratio is estimated to be ~11 for the bare fuel test based on highest <sup>137</sup>Cs activities measured in solution.

TABLE 12  
<sup>99</sup>Tc RELEASE DATA (nCi)

|  | <u>Bare Fuel</u> | <u>Slit Defect</u> | <u>Holes Defect</u> |
|--|------------------|--------------------|---------------------|
| <u>Initial Test Run</u>                      |                  |                    |                     |
| Final Solution<br>[ <sup>99</sup> Tc (ppb)]* | 563<br>(132)     | 28.2<br>(7)        | <4.5<br>(<1)        |
| Test Vessel Strip                            | 33.8             | 5.4                | <1.4                |
| <u>Second Test Run</u>                       |                  |                    |                     |
| Final Solution<br>[ <sup>99</sup> Tc (ppb)]* | 302<br>(71)      | 15.8<br>(4)        | 18.0<br>(4.2)       |
| Test Vessel Strip                            | 1.6              | <1.4               | 3.8                 |
| <u>Total Measured Release</u>                |                  |                    |                     |
| Σ Above (nCi)                                | 900.4            | <50.8              | <27.7               |
| Divided by 10 <sup>-5</sup> Inventory        | 230              | 12.1               | <6.7                |

\*Calculated <sup>99</sup>Tc concentration in unfiltered solution in ppb (μg/g).

To be preferentially leached, Tc must be locally enriched in regions of the fuel. It is likely that Tc partitions to the grain boundaries in the hotter central portions of the fuel. As grain boundary attack (which is clearly visible in Figure 3) progresses in these regions, enhanced Tc release would then be observed. Noble metal group fission products are not soluble in the UO<sub>2</sub> fuel matrix under the relatively reducing fuel oxygen potential during irradiation and will segregate when the temperature is high enough for them to diffuse. Technetium is found along with elements Mo, Ru, Rh, and Pd in electron microprobe examination of metallic fission product phases in the central restructured regions in liquid metal fast breeder reactor (LMFBR) fuels.<sup>(12)</sup> LMFBR fuel center temperatures are ~2000°C versus 1200°C to 1300°C in PWR fuels, resulting in more extensive restructuring and noble metal phase separation on a large enough scale for characterization by the electron microprobe. (For an overview of fission product thermochemical behavior in oxide fuels, see Chapter 12 of Reference 13.)

Molybdenum in 0.4- $\mu\text{m}$  filtered solution at the end of the initial bare fuel test run was determined to be 91 ppb by inductively coupled plasma (ICP) emission spectrometry analysis. The 91 ppb corresponds to  $\sim 23 \times 10^{-5}$  of the specimen fission product Mo inventory in solution at this time. The Tc concentration in the same solution sample was 132 ppb (determined radiochemically) corresponding to  $143 \times 10^{-5}$  of the specimen inventory. A lesser fraction of the fission product Mo might be expected to segregate from the oxide fuel matrix relative to Tc, since the oxygen potential threshold for Mo oxidation is nearer to the oxygen potential of the fuel during irradiation than is the higher oxygen potential for Tc oxidation. Lower fractional Mo release relative to Tc and Cs, but higher than U and Pu, may have been confirmed had a more accurate accounting of released Mo been made. Some additional insight on the state of preferentially leached Tc may be acquired in future tests by more accurate correlation to total release of Mo, Ru, Rh, and Pd. However, ICP accuracy at the predicted release concentrations may limit this approach.

### 3.7 NEPTUNIUM RELEASE

The primary Np isotope of concern for repository storage of spent fuel is  $^{237}\text{Np}$ . Activity of  $^{237}\text{Np}$  ( $2.14 \times 10^6$  year half-life) increases with time for about 10,000 years as a product of  $^{241}\text{Am}$  (458-year half-life) decay. At 1000 years and 10,000 years,  $^{237}\text{Np}$  accounts for about 0.06% and 0.25%, respectively, of the total ORIGEN-calculated<sup>(8)</sup> Ci activity of PWR fuel. Although  $^{237}\text{Np}$  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.  $^{239}\text{Np}$  represents a greater fraction of the calculated total activity ( $\sim 0.9\%$  at 1000 years). However,  $^{239}\text{Np}$  with its short ( $\sim 2$  day) half-life is a transient state in the  $^{243}\text{Am}$  to  $^{239}\text{Pu}$  decay chain and is considered as part of the  $^{243}\text{Am}$  activity in this report.

Release of  $^{237}\text{Np}$  from spent fuel could be expected to be congruent with U, Pu, and Am. Neptunium concentration is predicted to be only  $\sim 0.3\%$  on an

oxide weight basis and should be soluble in the  $\text{UO}_2$  fuel matrix as an oxide during irradiation at this concentration. After discharge from the reactor,  $^{237}\text{Np}$  increases slowly as a result of  $^{241}\text{Am}$  decay (which also increases for  $\sim 100$  years as a result of  $^{241}\text{Pu}$  beta decay with a 13.2-year half-life). The  $^{237}\text{Np}$  resulting from  $^{241}\text{Am}$  decay after discharge should also be uniformly distributed in the  $\text{UO}_2$  matrix if  $^{241}\text{Am}$  exists in solid solution as expected.

Neptunium was separated by a solvent extraction method and  $^{237}\text{Np}$  activity determined by alpha counting plus alpha spectrometry. Samples analyzed for  $^{237}\text{Np}$  were the final solution samples and test vessel strip solution samples at the ends of the both the initial and second runs of the four longest term tests. Results from these analyses are given in Table 13. Detection limit was only  $\sim 2$  pCi/ml for the first analyses on the initial run final test solutions accounting for the  $< 500$  pCi values given for these samples in Table 13. Detection limits were improved afterward by using larger sample portions and longer counting times. Results from initial run vessel strip and the second run samples from the bare fuel and slit defect tests appear to be significant, and suggest that  $^{237}\text{Np}$  may be preferentially released relative to U, Pu, Cm and Am. Preferential Np release would suggest partial segregation of Np from the irradiated  $\text{UO}_2$  matrix contrary to expectations. True leaching of Np from exposed fuel surfaces (with exposed surface area increasing as grain boundary dissolution progresses) could also explain the observed results. Possible errors, including ORIGEN calculation of  $^{237}\text{Np}$  inventory and analytical errors, may also be involved.

### 3.8 $^{14}\text{C}$ , $^{79}\text{Se}$ , AND $^{129}\text{I}$ RELEASE

Radiochemical separations and analyses for  $^{14}\text{C}$ ,  $^{79}\text{Se}$ , and  $^{129}\text{I}$  were made on unfiltered solution samples at the end of the initial run of the four longest term tests. Analysis for  $^{79}\text{Se}$  was also conducted on the 8 M  $\text{HNO}_3$  initial run test vessel strip solutions ( $^{14}\text{C}$  and  $^{129}\text{I}$  are lost from  $\text{HNO}_3$ ). Results for all of these analyses were reported as less-than values. The less-than values are the estimated detection limits for each analysis.

TABLE 13

 $^{237}\text{Np}$  RELEASE DATA (pCi)

|                                | <u>Bare Fuel</u> | <u>Slit Defect</u> | <u>Holes Defect</u> | <u>Undefected</u> |
|--------------------------------|------------------|--------------------|---------------------|-------------------|
| <u>Initial Test Run</u>        |                  |                    |                     |                   |
| Final Solution*                | <500             | <500               | <500                | <500              |
| Test Vessel Strip              | 1162             | 18                 | <3                  | <4                |
| <u>Second Test Run</u>         |                  |                    |                     |                   |
| Final Solution                 | 3266             | 90                 | 34                  | --                |
| [ $^{237}\text{Np}$ (ppb)]     | (18)             | (0.5)              | (0.2)               | --                |
| Test Vessel Strip              | 297              | 95                 | <130                | <130              |
| <u>Total Measured Release</u>  |                  |                    |                     |                   |
| $\Sigma$ Above (pCi)           | 4725             | 203                | --                  | --                |
| Divided by $10^{-5}$ Inventory | 54               | 2.2                | --                  | --                |

\*Detection limit improved after this analysis, initial run final solution not included in " $\Sigma$  Above (pCi)" value.

$^{14}\text{C}$  results from the (n,p) reaction on nitrogen present as an impurity in as-fabricated fuel and from the (n, $\alpha$ ) reaction on  $^{17}\text{O}$ . Fuel nitrogen impurities are not well characterized and reported for commercial fuels, and code-calculated  $^{14}\text{C}$  inventories for spent fuel are not reliable. Based on Reference 7 ORIGEN-2 data, specimen  $^{14}\text{C}$  inventory should be  $\sim 10^{-4}$   $\mu\text{Ci/gU}$ . Based on Reference 8 ORIGEN data, specimen inventory should be  $\sim 1.2$   $\mu\text{Ci/gU}$ . For H. B. Robinson spent fuel (same vendor and vintage as the Turkey Point fuel in this test),  $^{14}\text{C}$  values ranging from 0.14  $\mu\text{Ci/gU}$  to 0.29  $\mu\text{Ci/gU}$  were reported by Campbell and Buxton.<sup>(14)</sup> A specimen of H. B. Robinson fuel from the same assembly was analyzed for  $^{14}\text{C}$  by WHC with the following results: 0.33  $\mu\text{Ci/g}$  for the fuel and 0.6  $\mu\text{Ci/g}$  for the cladding.

The reported less-than values for  $^{14}\text{C}$  in the present tests correspond to <18 nCi (16 ppt) in the 250-ml test solutions. Assuming the above WHC-determined  $^{14}\text{C}$  values for H. B. Robinson fuel and cladding for the specimens

in these tests, specimen  $^{14}\text{C}$  inventory is estimated to be  $\sim 20 \mu\text{Ci}$ . The reported less-than value (detection limit) would therefore correspond to  $\sim 10^{-3}$  of specimen inventory in solution.

The reported less-than values for  $^{79}\text{Se}$  would correspond to  $< 9 \text{ nCi}$  (0.5 ppb) in the test solution and  $< 11 \text{ nCi}$  as vessel plate-out. The  $< 20 \text{ nCi}$  sum of the reported less-than values for the test solution and vessel strip solution corresponds to  $< 1.6 \times 10^{-3}$  of the calculated specimen  $^{79}\text{Se}$  inventory.

The reported less-than values for  $^{129}\text{I}$  would correspond to  $< 8.4 \text{ nCi}$  ( $\sim 0.2 \text{ ppm}$ ) in solution, or  $\sim 8.5 \times 10^{-3}$  of specimen inventory. If  $^{129}\text{I}$  were released congruently with  $^{137}\text{Cs}$ , as reported for CANDU fuel,  $^{129}\text{I}$  would still probably not be measurable by the liquid scintillation method used.  $^{129}\text{I}$  by neutron activation analysis is planned for future tests.

### 3.9 SOLUTION CHEMISTRY

Samples of  $0.4\text{-}\mu\text{m}$  filtered solution were analyzed by ICP and ion chromatography at the end of the first run of the tests. The ICP (Bausch and Lomb Model 3510 sequential ICP) was set up to analyze for: Al, B, Ca, Cr, Fe, Gd, K, Li, Mg, Mo, Na, Si, U, and Zn. Less-than values were reported for Al, Cr, Gd, and U for all four tests. These less-than detection limit values were 0.075, 0.010, 0.050, and  $0.500 \mu\text{g/ml}$ , respectively. Lithium just slightly greater than its  $0.015 \mu\text{g/ml}$  detection limit was reported for the bare fuel and undefected tests. The anions  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^-$  were measured by ion chromatography on solution samples from all four test types. The pH of all solution samples was measured immediately after sampling and before filtration. Significant results from these analyses are given in Table 14.

The two highest level impurities measured in the deionized water test solutions at the termination of the first run were  $\text{NO}_3^-$  and Si. The  $\sim 1 \mu\text{g/ml}$  of Si most likely came from the fused quartz test apparatus. The  $\sim 2 \mu\text{g/ml}$   $\text{NO}_3^-$  is though to have resulted from radiolysis of air, forming  $\text{NO}_2$ , which dissolves

TABLE 14  
 FINAL SOLUTION\* CHEMISTRY  
 (Units: pH and  $\mu\text{g/ml}$ )

|                 | <u>Bare Fuel</u> | <u>Slit Defect</u> | <u>Hole Defects</u> | <u>Undefected</u> |
|-----------------|------------------|--------------------|---------------------|-------------------|
| pH              | 6.03             | 5.87               | 6.15                | 6.05              |
| B               | 0.19             | 0.18               | 0.15                | 0.13              |
| Ca              | 0.33             | 0.29               | 0.26                | 0.41              |
| K               | <0.5             | 1.3                | 0.9                 | <0.5              |
| Fe              | 0.07             | 0.06               | 0.06                | 0.10              |
| Mo              | 0.09             | <0.02              | <0.02               | <0.02             |
| Na              | 0.76             | 0.78               | 0.69                | 0.18              |
| Si              | 0.43             | 1.06               | 0.90                | 1.11              |
| Zn              | 0.13             | 0.10               | 0.10                | 0.19              |
| F               | 0.09             | 0.13               | 0.09                | 0.08              |
| Cl              | 0.21             | 0.26               | 0.24                | 0.34              |
| NO <sub>3</sub> | 2.0              | 2.1                | 2.2                 | 1.6               |
| SO <sub>4</sub> | 0.85             | 0.66               | 0.63                | 0.66              |

\*At the end of the first test run (~250 days) with each specimen type; 0.4  $\mu\text{m}$  filtered.

into the test solution as HNO<sub>3</sub>. This small amount of radiolysis-induced HNO<sub>3</sub> would explain the slightly acidic pH values, ranging from ~5.4 to 6.4, observed in the tests.

### 3.10 STRUCTURAL CONSIDERATIONS

Incongruent release of U and radionuclides implies nonhomogeneous microstructural characteristics for spent fuel. Fuel restructuring (extensive grain growth) and metallic phase separation is easily observable in LMFBR fuels where centerline temperatures exceed 2000°C and fission gas release may exceed 50%. Fission product reaction phases containing Cs, Mo, Te, and I

are also found in the fuel-cladding gap, and alkaline earth oxide phases may be observed in the restructured regions of LMFBR fuels. Very little restructuring or phase separation is observable by optical ceramography of spent PWR fuel such as that used in the current tests. Centerline peak temperatures in these fuels are typically less than 1300°C and gas release is typically less than 0.5%. Cesium, which is relatively volatile at typical LWR fuel irradiation temperatures and reducing oxygen potential, is thought to be released from the fuel as "gap inventory" in quantities comparable to the fuel fractional fission gas release. A portion of the fission product  $^{129}\text{I}$  may be similarly released.<sup>(10)</sup> Little is known about the state of other nonvolatile fission products in LWR fuels, many of which would not be expected to be stable in solid solution with the oxide fuel matrix phase.

Preferential release of nonvolatile radionuclides suggests localized partitioning to fuel surfaces or grain boundaries. Figure 11A shows a SEM photograph of a fracture surface from a particle of the Turkey Point Fuel used in this study. An unirradiated  $\text{UO}_2$  fracture surface is shown in Figure 11B for comparison. The spent fuel is very friable and fractures along its grain boundaries. The unirradiated fuel is much tougher and exhibits primarily transgranular cleavage fracture. The spent fuel has open appearing grain boundaries, while the grain boundaries are not resolvable in the unirradiated fracture surface. Progression of spent fuel dissolutions by grain boundary attack, as indicated by Figure 3, is consistent with the spent fuel morphology shown in Figure 11A. Incongruent leaching of  $^{99}\text{Tc}$  and other preferentially released radionuclides in relatively short-term leach tests is probably due to dissolution of minute phases that have segregated to the grain boundaries. Figure 11 also suggests limitations in applying physical and chemical data obtained from studies using unirradiated  $\text{UO}_2$  as a surrogate for actual spent fuel.

An additional process that could significantly alter the microstructure of spent fuel is oxidation. In the humid air (or oxygen containing steam) atmosphere likely to be present in the NNWSI repository, oxidation of  $\text{UO}_2$  through  $\text{U}_3\text{O}_8$  to  $\text{UO}_3$  is thermodynamically favored. The kinetics of  $\text{UO}_2$  oxidation beyond  $\text{U}_3\text{O}_7$  become too slow to measure on unirradiated  $\text{UO}_2$  below

~200°C in reasonable laboratory time scales. However, based on thermodynamic considerations, one could argue that over repository relevant time scales, the fundamental question of spent fuel degradation by oxidation (with failed cladding in a failed waste package) is not a question of if, but a question of when. The open microstructure of spent fuel shown in Figure 11A should have greater oxidation rates as well as greater leaching rates in comparison to the unirradiated microstructure. A better understanding of spent fuel microstructure, the state of radionuclides in the microstructure, and the long-term stability of the fuel structure will be needed if any credit is to be taken for the fuel structure by itself as a barrier to long-term radionuclide release.

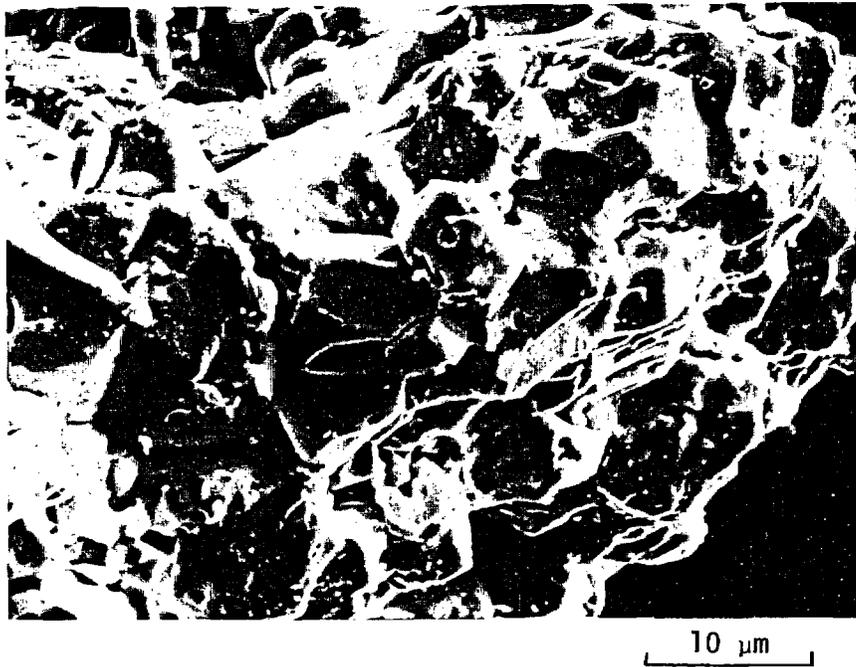


FIGURE 11A. Fracture Surface of Turkey Point Spent Fuel Exhibiting Fracture Primarily Along Grain Boundaries.



FIGURE 11B. Fracture Surface of Unirradiated Fuel Exhibiting Primarily Cleavage Fracture Through Grains.

#### 4.0 SUMMARY AND CONCLUSIONS

A comparative leach test of PWR spent fuel specimens was conducted in deionized water under ambient hot cell conditions. The Series 1 tests described are the first of several tests planned to characterize potential radionuclide release from and behavior of spent fuel stored under NNWSI-proposed conditions. The Series 1 test specimen types included: 1) bare fuel plus the cladding, 2) a rod segment with capped ends and a slit defect through the cladding, 3) rod segments with capped ends and laser-drilled holes ( $\sim 200 \mu\text{m}$  diameter) through the cladding, and 4) undefected rod segments with capped ends. The tests were "semi-static," in that small sample volumes of the test solution were removed periodically and replenished with fresh deionized water. After  $\sim 250$  days, a test of each type was terminated, radionuclide "plate-out" stripped from the test apparatus, and the tests restarted in fresh deionized water for a second run. An accounting of released radionuclides in solution and stripped from quartz rods and the test vessels was attempted. Selected solution samples were filtered to characterize the state of radionuclides in the "solution." Measured releases were compared to the 10 CFR 60  $10^{-5}$  of 1000-year inventory maximum annual release rate requirement. Total measured release and total measured release as a fraction of inventory times  $10^5$  are summarized in Table 15.

#### CONCLUSIONS

The principal observations and conclusions from these spent fuel leaching tests are summarized below:

- 1) Within the probable accuracy of total release measurements and specimen inventory calculations, the actinides U, Pu, Am, and Cm appear to have been released congruently.
- 2) Limited data suggest that  $^{237}\text{Np}$  may have been preferentially released rather than being congruently released with other actinides as expected.

TABLE 15  
TOTAL MEASURED RELEASE AS A FRACTION  
OF INVENTORY  $\times 10^5$ (a)

| <u>Component</u>                             | <u>Bare Fuel</u>                 | <u>Slit Defect</u>              | <u>Holes Defect</u>            | <u>Undefected</u>              |
|--|----------------------------------|---------------------------------|--------------------------------|--------------------------------|
| Uranium<br>( $\mu\text{g}$ )                 | 28.0<br>(9510)                   | 0.078<br>(28)                   | <0.041<br>(<14)                | <0.018<br>(<6.6)               |
| $^{239}\text{Pu} + ^{240}\text{Pu}$<br>(nCi) | 28.0<br>(7940)                   | 0.341<br>(104)                  | 0.069<br>(20)                  | 0.027<br>(8)                   |
| $^{241}\text{Am}$<br>(nCi)                   | 21.7<br>(12,604)                 | 0.208<br>(130)                  | <0.030<br>(<18.6)              | <0.011<br>(<6.4)               |
| $^{244}\text{Cm}$<br>(nCi)                   | 30.0<br>(13,300)                 | 0.76<br>(362)                   | 0.039<br>(18.1)                | 0.008<br>(<3.9)                |
| $^{237}\text{Np}$ (b)<br>(nCi)               | 54<br>(4.73)                     | 2.2<br>(0.2)                    | --<br>--                       | --<br>--                       |
| $^{137}\text{Cs}$<br>(nCi)                   | 300(c)<br>( $1.94 \times 10^6$ ) | 142.1<br>( $3.94 \times 10^6$ ) | 85.6<br>( $2.33 \times 10^6$ ) | 0.041<br>( $1.1 \times 10^3$ ) |
| $^{99}\text{Tc}$ (d)<br>(nCi)                | 230<br>(900)                     | 12.1<br>(51)                    | <6.7<br>(<28)                  | --<br>--                       |

- (a) Total measured release given in parentheses.  
 (b)  $^{237}\text{Np}$  includes only vessel strip from initial and second runs and final solution from second run.  
 (c) Estimate based on maximum  $^{137}\text{Cs}$  activities measured in solution.  
 (d)  $^{99}\text{Tc}$  includes only final solution and vessel strip from initial and second runs.

However, these data are too limited to be conclusive. Inaccuracies in ORIGEN-2 calculated  $^{237}\text{Np}$  inventory and radiochemical analysis could also account for those results.

- 3) A rapid fractional release of cesium on the order of the fractional fission gas release was observed for the bare fuel, slit defect, and holes defect tests. Additional preferential cesium release, possibly from grain boundary inventory, was also noted in the second run on these specimens.

- 4) Observed fractional  $^{99}\text{Tc}$  release ranged from one order of magnitude greater release relative to the actinides in the bare fuel test to almost three orders of magnitude greater fractional release relative to the actinides in the holes defect test.
- 5) For the actinides U, Pu, Am, and Cm, approximately two orders of magnitude less total fractional release was measured in the slit defect test relative to the bare fuel test. An additional approximate one order of magnitude reduction in actinide release was observed in the holes defect test relative to the slit defect test.
- 6) Apparent uranium saturation occurred at  $\sim 1$  ppb in all tests. Uranium in excess of a few ppb was removed by 18 Å filtration. Most of the U, Am, and Cm in solution samples from the bare fuel test was removed by filtration.
- 7) Grain boundary dissolution appeared to be a major source of release. Preferential release of  $^{99}\text{Tc}$  is likely a result of Tc segregation to the grain boundaries. Grain boundaries in the spent fuel are relatively wide and easily resolved by SEM. Grain boundaries in unirradiated  $\text{UO}_2$  are tight and not resolvable on a fracture surface by SEM.
- 8) Spent fuel leaching behavior, as well as other chemical and mechanical behavior, is influenced by microstructural phenomena, such as localized segregation of some elements to the grain boundaries. The extent of localized radionuclide segregation is influenced by irradiation temperature and may be correlated to fission gas release. Additional segregation of radionuclides into more easily leached phases could possibly occur if the fuel structure is degraded by oxidation during long-term repository storage.

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A P P E N D I X

RADIONUCLIDE INVENTORY AND RADIOCHEMICAL DATA

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## APPENDIX

### 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 for a H. B. Robinson Unit 2 PWR fuel 10 years after reactor discharge. The H. B. Robinson fuel is very similar (same vendor, same design, similar vintage and same 2.55% initial  $^{235}\text{U}$  enrichment) as the Turkey Point fuel used in the present tests. A burnup of 27.7 MWd/kgM was assumed for the Turkey Point fuel, and radionuclide inventories were calculated by linear interpolation between 25 MWd/kgM and 30 MWd/kgM data given in PNL-5109. The 27.7 MWd/kgM burnup value used is the actual radiochemical burnup analysis results reported for the Turkey Point H-6 and J-8 fuel rods from which the specimens were made (Reference 6, burnup samples 70 inches from rod bottom). Since the age after discharge of the fuel was ~8 years during the Series 1 tests, inventories for shorter-lived isotopes were corrected for decay from 8 to 10 years. 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 A.1. Specimen radionuclide inventories used for calculation of fractional release values in this report were obtained by multiplying the Table A.1 inventories by the specimen fuel weights. The fuel weights for the four primary specimens discussed in this report are listed below.

|   |        |                   |         |
|---|--------|-------------------|---------|
| • | H-6-19 | Bare Fuel         | 40.33 g |
| • | H-6-12 | Slit Defect       | 43.20 g |
| • | J-8-24 | 2 Laser Punctures | 42.50 g |
| • | H-6-1  | Undefected        | 42.50 g |

TABLE A.1  
 RADIONUCLIDE INVENTORIES USED IN FRACTIONAL  
 RELEASE CALCULATIONS

| <u>Radionuclide</u> | <u>Inventory (<math>\mu\text{Ci/g}</math>)</u> |
|---------------------|--|
| $^{244}\text{Cm}^*$ | $1.10 \times 10^3$                             |
| $^{241}\text{Am}$   | $1.45 \times 10^3$                             |
| $^{240}\text{Pu}$   | $4.31 \times 10^2$                             |
| $^{239}\text{Pu}$   | $2.73 \times 10^2$                             |
| $^{237}\text{Np}$   | $2.18 \times 10^{-1}$                          |
| $^{137}\text{Cs}^*$ | $6.40 \times 10^4$                             |
| $^{129}\text{I}$    | $2.42 \times 10^{-2}$                          |
| $^{99}\text{Tc}$    | $9.74 \times 10^0$                             |
| $^{79}\text{Se}$    | $3.02 \times 10^{-1}$                          |

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\*Corrected from 10 to 8 years after reactor discharge.

## 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  $\mu\text{g/ml}$  (ppm) or  $\mu\text{g/rod}$ , are given for each individual test in Tables A-3 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  $\mu\text{g/pCi}$  and isotope/element conversion factors for Equation A.1 are contained in Table A.2.

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

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  $^{239}\text{Pu}$  (or  $^{240}\text{Pu}$ ) pCi/ml value is needed. The  $^{239}\text{Pu}/(^{239}\text{Pu} + ^{240}\text{Pu})$  activity ratio should be 0.389 for the tested fuel based on the PNL-5109 ORIGEN-2 data.

TABLE A.2  
ACTIVITY-CONCENTRATION CONVERSION FACTORS

| <u>Radionuclide</u> | <u>μg/pCi</u>            | <u>Isotope/Element*</u>   |
|---------------------|--------------------------|---------------------------|
| <sup>14</sup> C     | $2.24 \times 10^{-7}$    | --                        |
| <sup>60</sup> Co    | $8.82 \times 10^{-10}$   | --                        |
| <sup>79</sup> Se    | $1.44 \times 10^{-5}$    | 0.105                     |
| <sup>99</sup> Tc    | $5.87 \times 10^{-5}$    | 1.000                     |
| <sup>106</sup> Ru   | $\sim 3 \times 10^{-10}$ | $\sim 3 \times 10^{-4**}$ |
| <sup>125</sup> Sb   | $9.43 \times 10^{-10}$   | $\sim 0.1**$              |
| <sup>129</sup> I    | $6.13 \times 10^{-3}$    | 0.76                      |
| <sup>134</sup> Cs   | $7.68 \times 10^{-10}$   | $\sim 0.003**$            |
| <sup>135</sup> Cs   | $1.13 \times 10^{-3}$    | 0.13                      |
| <sup>137</sup> Cs   | $1.16 \times 10^{-8}$    | 0.4                       |
| <sup>144</sup> Ce   | $3.1 \times 10^{-10}$    | $\sim 1 \times 10^{-4**}$ |
| <sup>154</sup> Eu   | $6.89 \times 10^{-9}$    | 0.13                      |
| <sup>237</sup> Np   | $1.42 \times 10^{-3}$    | 0.9999                    |
| <sup>238</sup> Pu   | $5.72 \times 10^{-8}$    | 0.012                     |
| <sup>239</sup> Pu   | $1.63 \times 10^{-5}$    | 0.595                     |
| <sup>240</sup> Pu   | $4.41 \times 10^{-6}$    | 0.255                     |
| <sup>241</sup> Am   | $3.09 \times 10^{-7}$    | 0.863                     |
| <sup>244</sup> Cm   | $1.20 \times 10^{-8}$    | 0.93                      |

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\*Based on ORIGEN-2 data in PNL-5109 interpolated to 27.7 MWd/kgM burnup, 10 years after discharge unless otherwise noted.  
 \*\*<sup>106</sup>Ru, <sup>125</sup>Sb, <sup>134</sup>Cs, and <sup>144</sup>Ce, half-lives are relatively short (1.0, 2.7, 2.0 and 0.8 years, respectively) causing these values to change significantly with time; values given have been corrected from 10 to 8 years after reactor discharge.

TABLE A.3

TEST TYPE: BARE FUEL

TEST IDENTIFICATION: H-6-19.

| DAY | SAMPLE TYPE | pH    | U                            |             | Pu-239+Pu-240              |             |                            | Am-241+Pu-238 |                            | Am-241*     |                            | Cm-244      |                            |             | Cm-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 | 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   | Sol         | 5.899 | 4.32E+00                     |             | 2.97E+03                   |             | 1.25E+04                   |               | 6.08E+03                   |             | 1.03E+04                   |             | 9.50E+06                   |             | 1.09E+06                   |             |          |             |
| 5   | Sol         | 6.050 | 6.00E-01                     |             | 6.00E-01                   |             | 1.59E+03                   |               | 7.03E+03                   |             | 3.60E+03                   |             | 6.89E+03                   |             | 1.45E+07                   |             |          |             |
| 5   | Rod         |       | 6.50E+01                     |             | 9.73E+04                   |             | 3.41E+05                   |               | 1.32E+05                   |             | 1.77E+05                   |             | 4.82E+06                   |             | 5.14E+05                   |             |          |             |
| 15  | Sol         | 6.462 | 1.80E-01                     |             | 2.52E+03                   |             | 6.98E+03                   |               | 1.57E+03                   |             | 4.43E+03                   |             | 1.80E+07                   |             | 2.00E+06                   |             |          |             |
| 30  | Sol         | 6.288 | 3.50E-01                     |             | 4.03E+03                   |             | 1.05E+04                   |               | 1.80E+03                   |             | 4.64E+03                   |             | 1.90E+07                   |             | 2.21E+06                   |             |          |             |
| 30  | Rod         |       | 9.10E+01                     |             | 1.35E+05                   |             | 4.77E+05                   |               | 1.07E+05                   |             | 2.59E+05                   |             | 7.39E+06                   |             | 7.88E+05                   |             |          |             |
| 60  | Sol         | 6.347 | 2.90E-01                     |             | 2.69E+03                   |             | 6.44E+03                   |               | 6.57E+02                   |             | 2.81E+03                   |             | 1.83E+07                   |             | 1.93E+06                   |             |          |             |
| 60  | Rod         |       | 3.90E+01                     |             | 3.47E+04                   |             | 1.32E+05                   |               | 3.77E+04                   |             | 9.10E+04                   |             | 4.33E+06                   |             | 4.41E+05                   |             |          |             |
| 90  | Sol         | 6.083 | 5.00E-01                     |             | 2.44E+03                   |             | 7.21E+03                   |               | 1.97E+03                   |             | 4.01E+03                   |             | 1.10E+07                   |             | 1.14E+06                   |             |          |             |
| 120 | Sol         | 5.927 | 5.30E-01                     |             | 2.42E+03                   |             | 7.39E+03                   |               | 2.19E+03                   |             | 4.59E+03                   |             | 4.86E+06                   |             | 5.14E+05                   |             |          |             |
| 120 | Rod         |       | 5.70E+01                     |             | 6.31E+04                   |             | 2.34E+05                   |               | 1.01E+05                   |             | 1.53E+05                   |             | 3.71E+06                   |             | 3.67E+05                   |             |          |             |
| 150 | Sol         | 6.218 | 9.30E-01                     |             | 1.95E+03                   |             | 4.88E+03                   |               | 2.65E+03                   |             | 5.32E+03                   |             | 2.38E+06                   |             | 2.45E+05                   |             |          |             |
| 180 | Sol         | 5.909 | 6.50E-01                     |             | 2.27E+03                   |             | 8.11E+03                   |               | 3.22E+03                   |             | 6.08E+03                   |             | 1.43E+06                   |             | 1.47E+05                   |             |          |             |
| 180 | Rod         |       | 4.40E+01                     |             | 3.55E+04                   |             | 1.33E+05                   |               | 5.68E+04                   |             | 8.92E+04                   |             | 2.64E+06                   |             | 2.34E+05                   |             |          |             |
| 202 | Sol         |       | 3.00E-01 3.00E-03 2.00E-03   |             | 2.24E+03 2.18E+03 1.81E+03 |             | 1.47E+04 6.13E+03 3.99E+03 |               | 1.43E+03 9.68E+01          |             | 2.11E+04 2.86E+03 1.63E+02 |             | 1.43E+06 9.37E+05 5.63E+05 |             | 1.36E+05 8.83E+04 5.18E+04 |             |          |             |
| 250 | Sol         | 6.028 | 3.00E-03 <1.00E-03 <1.00E-03 |             | 2.16E+03 2.14E+03 1.93E+03 |             | 4.82E+03 4.73E+03 4.17E+03 |               | 1.95E+02 1.23E+02 4.95E+01 |             | 2.92E+02 1.47E+02 4.05E+00 |             | 5.18E+05 5.05E+05 3.47E+05 |             | 4.59E+04 4.53E+04 3.09E+04 |             |          |             |
| 250 | Rod         |       | 6.70E+01                     |             | 5.95E+04                   |             | 2.14E+05                   |               | 1.25E+05                   |             | 1.18E+05                   |             | 2.52E+06                   |             | 2.16E+05                   |             |          |             |
| 250 | Strip       |       | 2.10E+01                     |             | 1.59E+04                   |             | 6.22E+04                   |               | 2.83E+04                   |             | 2.50E+04                   |             | 9.37E+05                   |             | 8.02E+04                   |             |          |             |
| 250 | Rinse       |       | 1.60E+00                     |             | 7.34E+02                   |             | 2.44E+03                   |               | 8.60E+02                   |             | 1.13E+03                   |             | 1.70E+05                   |             | 1.48E+04                   |             |          |             |

TEST RESTARTED IN FRESH DEIONIZED WATER

|     |       |       |                            |  |                            |  |                            |  |                            |  |                            |  |                            |  |                            |  |  |
|-----|-------|-------|----------------------------|--|----------------------------|--|----------------------------|--|----------------------------|--|----------------------------|--|----------------------------|--|----------------------------|--|--|
| 15  | Rod   |       | 6.40E+01                   |  | 1.08E+04                   |  | 4.43E+04                   |  | 2.27E+04                   |  | 3.00E+04                   |  | 4.39E+05                   |  | 3.74E+04                   |  |  |
| 15  | Sol   | 6.050 | 4.00E-01 4.00E-03 1.00E-03 |  | 4.59E+03 4.39E+03 3.94E+03 |  | 2.54E+04 1.18E+04 8.20E+03 |  | 1.38E+04 2.64E+03 4.95E+01 |  | 2.76E+04 4.42E+03 1.17E+01 |  | 6.53E+06 5.72E+06 3.60E+06 |  | 5.77E+05 5.05E+05 3.16E+05 |  |  |
| 70  | Rod   |       | 7.50E+01                   |  | 1.84E+04                   |  | 7.07E+04                   |  | 3.33E+04                   |  | 4.89E+04                   |  | 7.15E+05                   |  | 4.17E+04                   |  |  |
| 70  | Sol   | 6.340 | 1.80E-01 2.00E-03 1.00E-03 |  | 2.87E+03 2.67E+03 2.51E+03 |  | 1.23E+04 7.16E+03 5.32E+03 |  | 5.84E+03 7.84E+02 1.89E+01 |  | 1.17E+04 1.50E+03 7.21E+00 |  | 1.79E+04 1.44E+04 8.78E+05 |  | 1.54E+05 1.15E+05 7.25E+04 |  |  |
| 128 | Sol   | 5.902 | 3.00E-03 3.00E-03 1.00E-03 |  | 3.43E+03 3.33E+03 3.03E+03 |  | 1.29E+04 1.12E+04 8.58E+03 |  | 5.09E+03 3.73E+03 9.91E+00 |  | 9.95E+03 7.61E+03 6.76E+00 |  | 5.77E+05 5.90E+05 4.22E+05 |  | 4.53E+04 4.68E+04 3.31E+04 |  |  |
| 128 | Rod   |       | 7.40E+01                   |  | 1.71E+04                   |  | 6.80E+04                   |  | 3.58E+04                   |  | 4.84E+04                   |  | 3.82E+05                   |  | 2.95E+04                   |  |  |
| 128 | Strip |       | 5.10E+00                   |  | 1.12E+03                   |  | 3.93E+03                   |  | 1.84E+03                   |  | 1.96E+03                   |  | 6.44E+04                   |  | 4.91E+03                   |  |  |
| 128 | Rinse |       | 4.20E-01                   |  | 5.54E+02                   |  | 1.33E+03                   |  | 1.05E+03                   |  | 3.42E+02                   |  | 3.31E+05                   |  | 2.60E+04                   |  |  |

| DAY | SAMPLE TYPE | pH    | Co                         |             | Sb-125                     |             | Pu-104   |             | Ce-144   |             | Eu-154                     |             | Nd-237    |             | Tc-99                      |             |
|-----|-------------|-------|----------------------------|-------------|----------------------------|-------------|----------|-------------|----------|-------------|----------------------------|-------------|-----------|-------------|----------------------------|-------------|
|     |             |       | UNFILTER                   | 0.4 um 18 A | UNFILTER                   | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER                   | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER                   | 0.4 um 18 A |
| 1   | Sol         | 5.899 | 5.95E+03                   |             |                            |             |          |             |          |             |                            |             |           |             |                            |             |
| 5   | Sol         | 6.050 | 9.10E+03                   |             |                            |             |          |             |          |             |                            |             |           |             |                            |             |
| 5   | Rod         |       |                            |             | 6.13E+03                   |             | 1.02E+05 |             | 6.49E+04 |             | 1.94E+05                   |             |           |             |                            |             |
| 15  | Sol         | 6.462 | 1.85E+04                   |             |                            |             |          |             |          |             |                            |             |           |             |                            |             |
| 30  | Sol         | 6.288 | 3.18E+04                   |             |                            |             |          |             |          |             |                            |             |           |             |                            |             |
| 30  | Rod         |       |                            |             | 1.10E+05                   |             |          |             | 8.87E+04 |             | 3.42E+05                   |             |           |             |                            |             |
| 60  | Sol         | 6.347 | 5.45E+04                   |             |                            |             |          |             |          |             |                            |             |           |             |                            |             |
| 60  | Rod         |       | 4.42E+03                   |             | 6.53E+04                   |             | 1.38E+05 |             | 4.64E+04 |             | 2.21E+05                   |             |           |             |                            |             |
| 90  | Sol         | 6.083 | 6.44E+04                   |             |                            |             |          |             |          |             |                            |             |           |             |                            |             |
| 120 | Sol         | 5.927 | 7.25E+04                   |             |                            |             |          |             |          |             |                            |             |           |             |                            |             |
| 120 | Rod         |       | 5.81E+03                   |             | 5.50E+04                   |             | 1.09E+05 |             | 3.02E+04 |             | 2.32E+05                   |             |           |             |                            |             |
| 150 | Sol         | 6.218 | 7.88E+04                   |             |                            |             |          |             |          |             |                            |             |           |             |                            |             |
| 180 | Sol         | 5.909 | 9.10E+04                   |             | 2.43E+04                   |             |          |             |          |             | 1.52E+04                   |             |           |             |                            |             |
| 180 | Rod         |       | 6.94E+03                   |             | 4.26E+04                   |             |          |             | 2.28E+04 |             | 1.99E+05                   |             |           |             |                            |             |
| 202 | Sol         |       | 1.13E+05 8.69E+04 6.62E+02 |             | 2.02E+04 2.36E+04 2.45E+04 |             | 8.92E+04 |             |          |             | 5.50E+04 7.64E+03 7.66E+02 |             |           |             |                            |             |
| 250 | Sol         | 6.028 | 9.95E+04 9.55E+04 1.53E+04 |             | 1.81E+04 1.84E+04 1.83E+04 |             |          |             |          |             |                            |             | <2.25E+00 |             | 2.25E+03 2.25E+03 2.20E+03 |             |
| 250 | Rod         |       | 2.32E+04                   |             | 5.48E+04                   |             |          |             | 2.14E+04 |             | 1.95E+05                   |             |           |             |                            |             |
| 250 | Strip       |       | 1.34E+03                   |             | 1.34E+04                   |             | 1.39E+04 |             | 4.85E+03 |             | 4.36E+04                   |             | 3.87E+00  |             | 1.13E+02                   |             |
| 250 | Rinse       |       | 7.88E+02                   |             |                            |             |          |             |          |             | 3.35E+03                   |             |           |             |                            |             |

TEST RESTARTED IN FRESH DEIONIZED WATER

|     |       |       |                             |  |                            |  |  |  |          |  |                             |  |          |  |                            |  |
|-----|-------|-------|-----------------------------|--|----------------------------|--|--|--|----------|--|-----------------------------|--|----------|--|----------------------------|--|
| 15  | Sol   | 6.050 | 1.68E+04 1.36E+04 <4.14E+02 |  |                            |  |  |  |          |  |                             |  |          |  |                            |  |
| 70  | Sol   | 6.340 | 3.49E+04 2.53E+04           |  |                            |  |  |  |          |  |                             |  |          |  |                            |  |
| 128 | Sol   | 5.902 | 4.50E+04 4.44E+04 2.37E+03  |  | 8.69E+03 8.92E+03 8.60E+03 |  |  |  |          |  | 2.77E+04 2.11E+04 <3.90E+02 |  | 1.31E+01 |  | 1.21E+03 1.23E+03 1.16E+03 |  |
| 128 | Rod   |       | <1.40E+03                   |  | <1.70E+04                  |  |  |  |          |  | 1.25E+03                    |  |          |  |                            |  |
| 128 | Strip |       | 1.05E+02                    |  |                            |  |  |  | 1.57E+03 |  | 5.77E+03                    |  | 9.91E-01 |  | 5.41E+00                   |  |
| 128 | Rinse |       | 7.52E+02                    |  |                            |  |  |  |          |  | 1.40E+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 500 ml deionized water. Strip in same units as solution samples, test vessel was stripped with 300 ml of 8 M HNO3.

\* Am-241 values through 202 day solution sample calculated from Pu-239+Pu-240 and Am-241+Pu-238 values using Pu-238/(Pu-239+Pu-240) activity ratio of 2.148.

TABLE A.4

TEST TYPE: SLIT DEFECT

TEST IDENTIFICATION: H-6-12

| DAY                                     | SAMPLE TYPE | pH    | U         |             | Pu-239+Pu-240 |             |          | Am-241+Pu-238 |          | Am-241*     |          |             | Ce-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 | UNFILTER | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER | 0.4 um 18 A |          |          |          |
| 1                                       | Sol         | 5.760 | 8.00E-03  |             | 3.06E+00      |             | 1.08E+01 |               | 5.67E+00 |             | 5.41E+00 |             | 2.23E+04  |             | 2.47E+03  |             |           |             |          |             |          |          |          |
| 4                                       | Sol         | 6.620 | 4.30E-02  |             | 6.76E+00      |             | 2.91E+02 |               | 2.79E+02 |             | 6.98E+02 |             | 5.34E+05  |             | 6.31E+04  |             |           |             |          |             |          |          |          |
| 6                                       | Rod         |       | <2.00E-02 |             | Alpha<9       |             |          |               |          |             |          |             | <4.50E-01 |             | <4.05E+00 |             |           |             |          |             |          |          |          |
| 15                                      | Sol         | 6.585 | 3.00E-02  |             | 8.56E+00      |             | 9.91E+01 |               | 8.47E+01 |             | 1.97E+02 |             | 1.40E+06  |             | 1.63E+05  |             |           |             |          |             |          |          |          |
| 28                                      | Rod         |       | 2.20E-01  |             | 6.76E+00      |             | 1.14E+03 |               | 1.15E+03 |             | 2.59E+03 |             | 6.49E+04  |             | 6.53E+03  |             |           |             |          |             |          |          |          |
| 30                                      | Sol         | 6.073 | 2.90E-02  |             | 3.65E+01      |             | 1.42E+02 |               | 8.07E+01 |             | 1.77E+02 |             | 2.98E+06  |             | 3.45E+05  |             |           |             |          |             |          |          |          |
| 60                                      | Sol         | 6.667 | 2.40E-02  |             | 5.81E+01      |             | 1.42E+02 |               | 4.44E+01 |             | 1.03E+02 |             | 5.59E+06  |             | 6.49E+05  |             |           |             |          |             |          |          |          |
| 60                                      | Rod         |       | 3.60E-01  |             | 5.32E+01      |             | 1.42E+03 |               | 1.53E+03 |             | 3.49E+03 |             | 1.10E+05  |             | 1.16E+04  |             |           |             |          |             |          |          |          |
| 90                                      | Sol         | 6.458 | 2.90E-02  |             | 9.82E+01      |             | 2.13E+02 |               | 4.79E+01 |             | 1.04E+02 |             | 8.24E+06  |             | 8.87E+05  |             |           |             |          |             |          |          |          |
| 120                                     | Sol         | 6.505 | 5.30E-02  |             | 1.80E+02      |             | 4.77E+02 |               | 1.75E+02 |             | 2.26E+02 |             | 8.74E+06  |             | 9.53E+05  |             |           |             |          |             |          |          |          |
| 120                                     | Rod         |       | 4.50E-01  |             | 1.14E+02      |             | 1.75E+03 |               | 1.56E+03 |             | 3.40E+03 |             | 1.59E+05  |             | 1.60E+04  |             |           |             |          |             |          |          |          |
| 150                                     | Sol         | 6.252 | 2.40E-02  |             | 1.07E+02      |             | 1.91E+02 |               | 1.15E+01 |             | 2.06E+01 |             | 9.55E+06  |             | 1.01E+06  |             |           |             |          |             |          |          |          |
| 180                                     | Sol         | 5.795 | 3.60E-02  |             | 1.35E+02      |             | 2.78E+02 |               | 5.13E+01 |             | 9.73E+01 |             | 9.73E+06  |             | 1.00E+06  |             |           |             |          |             |          |          |          |
| 180                                     | Rod         |       | 6.50E-01  |             | 1.57E+02      |             | 1.46E+03 |               | 1.20E+03 |             | 2.61E+03 |             | 1.92E+05  |             | 1.83E+04  |             |           |             |          |             |          |          |          |
| 210                                     | Sol         | 5.730 | 1.60E-02  |             | 1.30E+02      |             | 2.21E+02 |               | 3.16E+00 |             | 1.44E+01 |             | 1.12E+07  |             | 1.12E+06  |             |           |             |          |             |          |          |          |
| 252                                     | Sol         | 5.868 | 3.00E-03  | <1.00E-03   | <1.00E-03     | 1.54E+02    | 1.52E+02 | 1.45E+02      | 2.51E+02 | 2.52E+02    | 2.34E+02 | 6.31E+00    | 2.70E+00  | 4.05E+00    | 6.31E+00  | <4.50E-01   | <4.50E-01 | 9.73E+06    | 9.82E+06 | 8.02E+06    | 9.64E+05 | 9.59E+05 | 7.48E+05 |
| 252                                     | Rod         |       | 7.40E-01  |             | 2.32E+02      |             | 1.36E+03 |               | 1.56E+02 |             | 9.46E+02 |             | 2.02E+03  |             | 4.03E+04  |             |           |             |          |             |          |          |          |
| 252                                     | Strip       |       | 5.80E-02  |             | 4.41E+01      |             | 3.32E+02 |               | 2.60E+02 |             | 4.82E+02 |             | 3.59E+04  |             | 3.33E+03  |             |           |             |          |             |          |          |          |
| TEST RESTARTED IN FRESH DEIONIZED WATER |             |       |           |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 15                                      | Rod         |       | 5.00E-01  |             | 2.34E+01      |             | 1.81E+03 |               | 1.45E+03 |             | 3.05E+03 |             | 3.52E+04  |             | 3.26E+03  |             |           |             |          |             |          |          |          |
| 15                                      | Sol         | 5.896 | 2.00E-02  | 1.20E-02    | 7.00E-03      | 5.72E+01    | 5.68E+01 | 4.91E+01      | 2.77E+02 | 1.20E+02    | 9.59E+01 | 1.76E+02    | 9.46E+00  | <9.00E-01   | 2.89E+02  | 1.71E+01    | 9.01E-01  | 7.52E+05    | 4.77E+05 | 3.25E+05    | 6.67E+04 | 4.22E+04 | 2.88E+04 |
| 70                                      | Rod         |       | 3.00E-01  |             | 3.69E+01      |             | 4.37E+03 |               | 3.77E+03 |             | 7.16E+03 |             | 7.97E+04  |             | 7.66E+03  |             |           |             |          |             |          |          |          |
| 70                                      | Sol         | 5.780 | 8.00E-03  | 3.00E-03    | 1.00E-03      | 1.04E+02    | 9.46E+01 | 8.11E+01      | 2.57E+02 | 1.76E+02    | 1.49E+02 | 6.71E+01    | 7.66E+00  | 4.05E+00    | 1.17E+02  | 6.76E+00    | 9.01E-01  | 3.07E+06    | 2.18E+06 | 1.27E+06    | 2.68E+05 | 1.86E+05 | 1.02E+05 |
| 128                                     | Sol         | 5.826 | 2.00E-03  | 2.00E-03    | 1.00E-03      | 1.50E+02    | 1.49E+02 | 1.30E+02      | 2.59E+02 | 2.62E+02    | 2.21E+02 | 2.25E+00    | 1.35E+00  | 4.05E+00    | 2.70E+00  | <4.50E-01   | 3.49E+06  | 3.50E+06    | 2.12E+06 | 2.80E+05    | 2.83E+05 | 1.67E+05 |          |
| 128                                     | Rod         |       | 1.60E-01  |             | 5.68E+01      |             | 2.63E+03 |               | 2.36E+03 |             | 4.22E+03 |             | 5.95E+04  |             | 4.59E+03  |             |           |             |          |             |          |          |          |
| 128                                     | Strip       |       | 1.00E-02  |             | 1.49E+01      |             | 1.05E+02 |               | 8.69E+01 |             | 1.30E+02 |             | 7.88E+03  |             | 6.58E+02  |             |           |             |          |             |          |          |          |
| DAY                                     | SAMPLE TYPE | pH    | Co-60     |             | Sb-125        |             | Pu-106   |               | Ce-144   |             | Eu-154   |             | Mn-257    |             | Tc-99     |             |           |             |          |             |          |          |          |
|   |             |       | UNFILTER  | 0.4 um 18 A | UNFILTER      | 0.4 um 18 A | UNFILTER | 0.4 um 18 A   | UNFILTER | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A |          |             |          |          |          |
| 1                                       | Sol         | 5.760 | 1.86E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 4                                       | Sol         | 6.620 | 5.41E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 6                                       | Rod         |       | <6.76E+02 |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 15                                      | Sol         | 6.585 | 1.26E+04  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 28                                      | Rod         |       |           |             |               |             |          |               |          |             |          |             | 5.05E+03  |             |           |             |           |             |          |             |          |          |          |
| 30                                      | Sol         | 6.073 | 2.87E+04  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 60                                      | Sol         | 6.667 | 8.42E+04  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 60                                      | Rod         |       | 2.51E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 90                                      | Sol         | 6.458 | 1.40E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 120                                     | Sol         | 6.505 | 1.73E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 120                                     | Rod         |       | 9.41E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 150                                     | Sol         | 6.252 | 2.23E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 180                                     | Sol         | 5.795 | 2.48E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 180                                     | Rod         |       | 3.22E+04  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 210                                     | Sol         | 5.730 | 3.20E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 252                                     | Sol         | 5.868 | 3.52E+05  | 3.58E+05    | 8.60E+04      |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 252                                     | Rod         |       | 1.74E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 252                                     | Strip       |       | 5.41E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| TEST RESTARTED IN FRESH DEIONIZED WATER |             |       |           |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 15                                      | Rod         |       | 1.38E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 15                                      | Sol         | 5.896 | 3.81E+04  | 2.91E+04    | 8.96E+02      |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 70                                      | Sol         | 5.780 | 1.12E+05  | 9.32E+04    |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 128                                     | Sol         | 5.826 | 1.95E+05  | 1.78E+05    | 4.91E+03      |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 128                                     | Rod         |       | 3.64E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |
| 128                                     | Strip       |       | 4.22E+02  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |          |          |

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.  
 Strip in same units as solution samples, test vessel was stripped with 300 ml of 8 M HNO3.

\* Am-241 values through 210 day solution sample calculated from Pu-239+Pu-240 and Am-241+Pu-238 values using Pu-238/(Pu-239+Pu-240) activity ratio of 1.677.

TABLE A.4

TEST TYPE: SLIT DEFECT

TEST IDENTIFICATION: H-6-12

| DAY                                     | SAMPLE TYPE | pH    | U         |             | Pu-239+Pu-240 |             |          | Am-241+Pu-238 |          |             | Am-241*  |             |           | Ce-244      |           |             | Ce-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 | UNFILTER | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER | 0.4 um 18 A |          |
|   |             |       |           |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 1                                       | Sol         | 5.760 | 8.00E-03  |             | 3.06E+00      |             | 1.08E+01 |               | 5.67E+00 |             | 5.41E+00 |             | 2.23E+04  |             | 2.47E+03  |             |           |             |          |             |          |             |          |
| 4                                       | Sol         | 6.620 | 4.30E-02  |             | 6.76E+00      |             | 2.91E+02 |               | 2.79E+02 |             | 6.98E+02 |             | 5.54E+05  |             | 6.31E+04  |             |           |             |          |             |          |             |          |
| 6                                       | Rod         |       | <2.00E-02 |             | Alpha<9       |             |          |               |          |             |          |             | <4.50E-01 |             | <4.05E+00 |             |           |             |          |             |          |             |          |
| 15                                      | Sol         | 6.585 | 3.00E-02  |             | 8.56E+00      |             | 9.91E+01 |               | 8.47E+01 |             | 1.97E+02 |             | 1.40E+06  |             | 1.63E+05  |             |           |             |          |             |          |             |          |
| 28                                      | Rod         |       | 2.20E-01  |             | 6.76E+00      |             | 1.14E+03 |               | 1.13E+03 |             | 2.59E+03 |             | 6.49E+04  |             | 6.53E+03  |             |           |             |          |             |          |             |          |
| 30                                      | Sol         | 6.073 | 2.90E-02  |             | 3.65E+01      |             | 1.42E+02 |               | 8.07E+01 |             | 1.77E+02 |             | 2.98E+06  |             | 3.45E+05  |             |           |             |          |             |          |             |          |
| 60                                      | Sol         | 6.667 | 2.40E-02  |             | 5.81E+01      |             | 1.42E+02 |               | 4.44E+01 |             | 1.03E+02 |             | 5.59E+06  |             | 6.49E+05  |             |           |             |          |             |          |             |          |
| 60                                      | Rod         |       | 3.60E-01  |             | 5.32E+01      |             | 1.62E+03 |               | 1.53E+03 |             | 3.49E+03 |             | 1.10E+05  |             | 1.16E+04  |             |           |             |          |             |          |             |          |
| 90                                      | Sol         | 6.458 | 2.90E-02  |             | 9.82E+01      |             | 2.13E+02 |               | 4.79E+01 |             | 1.04E+02 |             | 8.24E+06  |             | 8.87E+05  |             |           |             |          |             |          |             |          |
| 120                                     | Sol         | 6.505 | 5.30E-02  |             | 1.80E+02      |             | 4.77E+02 |               | 1.75E+02 |             | 2.26E+02 |             | 8.74E+06  |             | 9.35E+05  |             |           |             |          |             |          |             |          |
| 120                                     | Rod         |       | 4.50E-01  |             | 1.14E+02      |             | 1.73E+03 |               | 1.54E+03 |             | 3.40E+03 |             | 1.59E+05  |             | 1.60E+04  |             |           |             |          |             |          |             |          |
| 150                                     | Sol         | 6.252 | 2.40E-02  |             | 1.07E+02      |             | 1.91E+02 |               | 1.15E+01 |             | 3.06E+01 |             | 9.55E+06  |             | 1.01E+06  |             |           |             |          |             |          |             |          |
| 180                                     | Sol         | 5.795 | 3.60E-02  |             | 1.35E+02      |             | 2.78E+02 |               | 5.13E+01 |             | 9.73E+01 |             | 9.73E+06  |             | 1.00E+06  |             |           |             |          |             |          |             |          |
| 180                                     | Rod         |       | 6.50E-01  |             | 1.57E+02      |             | 1.44E+03 |               | 1.20E+03 |             | 2.61E+03 |             | 1.92E+05  |             | 1.83E+04  |             |           |             |          |             |          |             |          |
| 210                                     | Sol         | 5.730 | 1.60E-02  |             | 1.30E+02      |             | 2.21E+02 |               | 5.16E+00 |             | 1.44E+01 |             | 1.12E+07  |             | 1.12E+06  |             |           |             |          |             |          |             |          |
| 252                                     | Sol         | 5.868 | 3.00E-03  | <1.00E-03   | <1.00E-03     | 1.54E+02    | 1.52E+02 | 1.45E+02      | 2.51E+02 | 2.52E+02    | 2.34E+02 | 6.31E+00    | 2.70E+00  | 4.05E+00    | 6.31E+00  | <4.50E-01   | <4.50E-01 | 9.73E+06    | 9.82E+06 | 8.02E+06    | 9.64E+05 | 9.59E+05    | 7.48E+05 |
| 252                                     | Rod         |       | 7.40E-01  |             | 2.32E+02      |             | 1.36E+03 |               | 9.46E+02 |             | 2.02E+03 |             | 4.19E+05  |             | 4.03E+04  |             |           |             |          |             |          |             |          |
| 252                                     | Strip       |       | 5.80E-02  |             | 4.41E+01      |             | 3.32E+02 |               | 2.60E+02 |             | 4.82E+02 |             | 3.59E+04  |             | 3.33E+03  |             |           |             |          |             |          |             |          |
| TEST RESTARTED IN FRESH DEIONIZED WATER |             |       |           |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 15                                      | Rod         |       | 5.00E-01  |             | 2.34E+01      |             | 1.81E+03 |               | 1.45E+03 |             | 3.05E+03 |             | 3.52E+04  |             | 3.26E+03  |             |           |             |          |             |          |             |          |
| 15                                      | Sol         | 5.896 | 2.00E-02  | 1.20E-02    | 7.00E-03      | 5.72E+01    | 5.68E+01 | 4.91E+01      | 2.77E+02 | 1.20E+02    | 9.59E+01 | 1.76E+02    | 9.46E+00  | <9.00E-01   | 2.89E+02  | 1.71E+01    | 9.01E-01  | 7.52E+05    | 4.77E+05 | 3.25E+05    | 6.67E+04 | 4.22E+04    | 2.88E+04 |
| 70                                      | Rod         |       | 3.00E-01  |             | 3.69E+01      |             | 4.37E+03 |               | 3.77E+03 |             | 7.16E+03 |             | 7.97E+04  |             | 7.64E+03  |             |           |             |          |             |          |             |          |
| 70                                      | Sol         | 5.780 | 8.00E-03  | 3.00E-03    | 1.00E-03      | 1.04E+02    | 9.44E+01 | 8.11E+01      | 2.57E+02 | 1.76E+02    | 1.49E+02 | 6.71E+01    | 7.64E+00  | 4.05E+00    | 1.17E+02  | 6.76E+00    | 9.01E-01  | 3.07E+06    | 2.18E+06 | 1.27E+06    | 2.68E+05 | 1.84E+05    | 1.02E+05 |
| 128                                     | Sol         | 5.826 | 2.00E-03  | 2.00E-03    | 1.00E-03      | 1.50E+02    | 1.49E+02 | 1.30E+02      | 2.59E+02 | 2.62E+02    | 2.21E+02 | 2.25E+00    | 1.35E+00  | 4.05E+00    | 2.70E+00  | <4.50E-01   | 3.49E+06  | 3.50E+06    | 2.12E+06 | 2.80E+05    | 2.83E+05 | 1.67E+05    |          |
| 128                                     | Rod         |       | 1.60E-01  |             | 5.68E+01      |             | 2.63E+03 |               | 2.36E+03 |             | 4.22E+03 |             | 5.95E+04  |             | 4.59E+03  |             |           |             |          |             |          |             |          |
| 128                                     | Strip       |       | 1.00E-02  |             | 1.49E+01      |             | 1.08E+02 |               | 8.69E+01 |             | 1.30E+02 |             | 7.88E+03  |             | 6.58E+02  |             |           |             |          |             |          |             |          |
| DAY                                     | SAMPLE TYPE | pH    | Co-60     |             | Sb-125        |             | Ru-106   |               | Ce-144   |             | Eu-154   |             | Mn-237    |             | Tc-99     |             |           |             |          |             |          |             |          |
|   |             |       | UNFILTER  | 0.4 um 18 A | UNFILTER      | 0.4 um 18 A | UNFILTER | 0.4 um 18 A   | UNFILTER | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A |           |             |          |             |          |             |          |
| 1                                       | Sol         | 5.760 | 1.86E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 4                                       | Sol         | 6.620 | 5.41E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 6                                       | Rod         |       | <6.76E+02 |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 15                                      | Sol         | 6.585 | 1.26E+04  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 28                                      | Rod         |       |           |             |               |             |          |               |          |             |          | 5.05E+03    |           |             |           |             |           |             |          |             |          |             |          |
| 30                                      | Sol         | 6.073 | 2.87E+04  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 60                                      | Sol         | 6.667 | 8.42E+04  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 60                                      | Rod         |       | 2.51E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 90                                      | Sol         | 6.458 | 1.40E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 120                                     | Sol         | 6.505 | 1.73E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 120                                     | Rod         |       | 9.41E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 150                                     | Sol         | 6.252 | 2.23E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 180                                     | Sol         | 5.795 | 2.48E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 180                                     | Rod         |       | 3.22E+04  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 210                                     | Sol         | 5.730 | 3.20E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 252                                     | Sol         | 5.868 | 3.32E+05  | 3.58E+05    | 8.60E+04      |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 252                                     | Rod         |       | 1.74E+05  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 252                                     | Strip       |       | 5.41E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| TEST RESTARTED IN FRESH DEIONIZED WATER |             |       |           |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 15                                      | Rod         |       | 1.38E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 15                                      | Sol         | 5.896 | 3.81E+04  | 2.91E+04    | 8.96E+02      |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 70                                      | Sol         | 5.780 | 1.12E+05  | 9.32E+04    |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 128                                     | Sol         | 5.826 | 1.95E+05  | 1.78E+05    | 4.91E+03      |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 128                                     | Rod         |       | 3.64E+03  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |
| 128                                     | Strip       |       | 4.22E+02  |             |               |             |          |               |          |             |          |             |           |             |           |             |           |             |          |             |          |             |          |

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.  
 Strip in same units as solution samples, test vessel was stripped with 300 ml of 8 M HNO3.

\* Am-241 values through 210 day solution sample calculated from Pu-239+Pu-240 and Am-241+Pu-238 values using Pu-238/(Pu-239+Pu-240) activity ratio of 1.677.

TABLE A.5

TEST TYPE: 2 LASER PUNCTURES

TEST IDENTIFICATION: J-8-24

| DAY                                     | SAMPLE TYPE | pH    | U         |          |           | Pu-239+Pu-240 |          |          | Am-241+Pu-238 |          |          | Am-241*   |           |           | Ca-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      | UNFILTER | 0.4 um    | 18 A      | UNFILTER  | 0.4 um   | 18 A     | UNFILTER  | 0.4 um   | 18 A     |
| 1                                       | Sol         | 5.622 | 4.00E-03  |          |           | 1.40E+00      |          |          | 4.28E+00      |          |          | 1.40E+00  |           |           | 1.49E+00 |           |           | 1.51E+04  |          |          | 1.60E+03  |          |          |
| 5                                       | Sol         | 6.333 | 2.00E-03  |          |           | 1.40E+00      |          |          | 3.92E+00      |          |          | 1.04E+00  |           |           | 1.04E+00 |           |           | 2.27E+04  |          |          | 2.07E+03  |          |          |
| 5                                       | Rod         |       | 9.00E-02  |          |           | 4.01E+00      |          |          | 8.56E+00      |          |          | 2.70E-01  |           |           | 4.95E+00 |           |           | <9.00E+02 |          |          | <7.00E+02 |          |          |
| 15                                      | Sol         | 6.480 | 6.00E-03  |          |           | 7.21E+00      |          |          | 2.03E+01      |          |          | 5.32E+00  |           |           | 5.13E+00 |           |           | 1.05E+06  |          |          | 1.23E+05  |          |          |
| 30                                      | Sol         | 6.484 | 6.00E-03  |          |           | 1.80E+01      |          |          | 4.05E+01      |          |          | 3.20E+00  |           |           | 2.70E+00 |           |           | 2.54E+06  |          |          | 2.98E+05  |          |          |
| 30                                      | Rod         |       | 3.60E-02  |          |           | 4.31E+00      |          |          | 5.41E+01      |          |          | 4.10E+01  |           |           | 4.19E+01 |           |           | 5.90E+04  |          |          | 6.71E+03  |          |          |
| 60                                      | Sol         | 6.389 | 4.00E-03  |          |           | 2.16E+01      |          |          | 5.09E+01      |          |          | 4.08E+00  |           |           | 2.70E+00 |           |           | 3.81E+06  |          |          | 4.28E+05  |          |          |
| 60                                      | Rod         |       | 4.50E-02  |          |           | 1.17E+01      |          |          | 1.19E+02      |          |          | 9.50E+01  |           |           | 7.79E+01 |           |           | 7.75E+04  |          |          | 6.98E+03  |          |          |
| 90                                      | Sol         | 6.106 | 4.00E-03  |          |           | 2.61E+01      |          |          | 5.86E+01      |          |          | 4.41E+00  |           |           | 4.50E+00 |           |           | 4.43E+06  |          |          | 4.86E+05  |          |          |
| 120                                     | Sol         | 5.958 | 6.00E-03  |          |           | 2.84E+01      |          |          | 6.31E+01      |          |          | 4.23E+00  |           |           | 5.86E+00 |           |           | 4.18E+06  |          |          | 4.55E+05  |          |          |
| 120                                     | Rod         |       | 1.40E-01  |          |           | 5.41E+01      |          |          | 2.66E+02      |          |          | 1.54E+02  |           |           | 2.37E+02 |           |           | 1.73E+05  |          |          | 1.62E+04  |          |          |
| 150                                     | Sol         | 5.829 | 1.90E-02  |          |           | 3.06E+01      |          |          | 6.94E+01      |          |          | 5.86E+00  |           |           | 5.86E+00 |           |           | 4.59E+06  |          |          | 4.91E+05  |          |          |
| 180                                     | Sol         | 5.875 | 6.00E-03  |          |           | 3.06E+01      |          |          | 7.07E+01      |          |          | 7.21E+00  |           |           | 5.86E+00 |           |           | 4.64E+06  |          |          | 4.77E+05  |          |          |
| 180                                     | Rod         |       | 3.90E-01  |          |           | 5.00E+01      |          |          | 3.15E+02      |          |          | 2.67E+02  |           |           | 2.67E+02 |           |           | 1.94E+05  |          |          | 1.86E+04  |          |          |
| 223                                     | Sol         | 5.859 | 2.00E-03  | 2.00E-03 | 1.00E-03  | 3.24E+01      | 3.20E+01 | 2.93E+01 | 6.85E+01      | 6.76E+01 | 6.08E+01 | 1.23E+00  | 1.27E+00  | 1.17E-01  | 6.31E-01 | <5.00E-01 | <5.00E-01 | 4.40E+06  | 4.34E+06 | 3.24E+06 | 4.18E+05  | 4.10E+05 | 3.03E+05 |
| 244                                     | Sol         | 6.156 | 1.00E-03  | 1.00E-03 | 1.00E-03  | 3.33E+01      | 3.33E+01 | 2.97E+01 | 6.71E+01      | 6.94E+01 | 6.35E+01 | 1.80E+00  | 3.15E+00  | 1.35E+00  | 4.50E-01 | 9.01E-01  | <0.5E+00  | 4.14E+06  | 4.13E+06 | 3.12E+06 | 3.79E+05  | 3.82E+05 | 2.92E+05 |
| 244                                     | Rod         |       | 1.30E+00  |          |           | 1.16E+02      |          |          | 3.98E+02      |          |          | 1.67E+02  |           |           | 1.89E+02 |           |           | 2.34E+05  |          |          | 2.05E+04  |          |          |
| 244                                     | Strip       |       | 2.80E-02  |          |           | 6.74E+00      |          |          | 4.50E+01      |          |          | 3.29E+01  |           |           | 3.56E+01 |           |           | 2.31E+04  |          |          | 2.06E+03  |          |          |
| TEST RESTARTED IN FRESH DEIONIZED WATER |             |       |           |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 15                                      | Rod         |       | 3.90E-01  |          |           | 1.26E+01      |          |          | 5.05E+02      |          |          | 4.82E+02  |           |           | 5.14E+02 |           |           | 3.07E+04  |          |          | 2.96E+03  |          |          |
| 15                                      | Sol         | 6.094 | 2.30E-02  | 1.10E-02 | 6.00E-03  | 1.71E+01      | 1.76E+01 | 6.31E+00 | 6.40E+01      | 3.83E+01 | 1.26E+01 | 2.70E+01  | 2.70E+00  | <0.5E+00  | 3.06E+01 | 2.25E+00  | <5.00E-01 | 7.79E+05  | 4.77E+05 | 3.14E+05 | 7.03E+04  | 4.28E+04 | 2.79E+04 |
| 70                                      | Rod         |       | 2.80E-01  |          |           | 1.26E+01      |          |          | 9.01E+02      |          |          | 9.89E+02  |           |           | 8.56E+02 |           |           | 1.05E+05  |          |          | 7.75E+03  |          |          |
| 70                                      | Sol         | 6.088 | 1.10E-02  | 4.00E-03 | 2.00E-03  | 2.84E+01      | 2.48E+01 | 2.48E+01 | 7.30E+01      | 5.09E+01 | 4.93E+01 | 2.34E+01  | 4.05E+00  | 2.25E+00  | 1.85E+01 | 1.33E+00  | 9.01E-01  | 2.63E+06  | 1.90E+06 | 1.09E+06 | 2.17E+05  | 1.63E+05 | 9.10E+04 |
| 128                                     | Sol         | 5.403 | 2.00E-03  | 2.00E-03 | 2.00E-03  | 2.84E+01      | 2.97E+01 | 2.66E+01 | 5.99E+01      | 6.08E+01 | 5.23E+01 | <1.40E+00 | <1.40E+00 | <1.40E+00 | 9.01E-01 | 9.01E-01  | <5.00E-01 | 3.79E+06  | 3.79E+06 | 2.52E+06 | 3.12E+05  | 3.15E+05 | 2.08E+05 |
| 128                                     | Rod         |       | 1.80E-01  |          |           | 2.61E+01      |          |          | 1.48E+03      |          |          | 1.41E+03  |           |           | 1.36E+03 |           |           | 1.01E+05  |          |          | 8.65E+03  |          |          |
| 128                                     | Strip       |       | <5.00E-03 |          |           | 1.53E+00      |          |          | 1.26E+01      |          |          | 1.17E+01  |           |           | 9.01E+00 |           |           | 1.66E+04  |          |          | 1.41E+03  |          |          |
| DAY                                     | SAMPLE TYPE | pH    | Co-60     |          |           | Sb-125        |          |          | Pu-106        |          |          | Ca-144    |           |           | Eu-154   |           |           | Mn-237    |          |          | Tc-99     |          |          |
| 1                                       | Sol         | 5.622 | 1.56E+03  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 5                                       | Sol         | 6.333 | 1.81E+03  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 5                                       | Rod         |       | <1.30E+03 |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 15                                      | Sol         | 6.480 | 2.19E+03  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 30                                      | Sol         | 6.484 | 2.84E+03  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 30                                      | Rod         |       | 6.89E+03  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 60                                      | Sol         | 6.389 |           |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 60                                      | Rod         |       |           |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 90                                      | Sol         | 6.106 | 1.42E+04  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 120                                     | Sol         | 5.958 | 1.64E+04  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 120                                     | Rod         |       | 1.13E+03  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 150                                     | Sol         | 5.829 | 2.71E+04  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 180                                     | Sol         | 5.875 | 3.07E+04  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 180                                     | Rod         |       | 1.42E+03  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 223                                     | Sol         | 5.859 | 3.93E+04  | 3.49E+04 | 2.64E+03  |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 244                                     | Sol         | 6.156 | 3.32E+04  | 3.27E+04 | 5.81E+03  |               |          |          |               |          |          |           |           |           |          |           |           | <2.20E+00 |          |          | <1.80E+01 |          |          |
| 244                                     | Rod         |       | 4.49E+03  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 244                                     | Strip       |       | 5.41E+02  |          |           |               |          |          |               |          |          |           |           |           |          |           |           | <3.00E+02 |          |          | <9.00E-03 |          |          |
| TEST RESTARTED IN FRESH DEIONIZED WATER |             |       |           |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 15                                      | Sol         | 6.094 | 2.43E+03  | 1.85E+03 |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 70                                      | Sol         | 6.088 | 4.77E+03  | 3.83E+03 |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 128                                     | Sol         | 5.403 | 7.84E+03  | 6.13E+03 | <8.00E+02 |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 128                                     | Rod         |       | <1.00E+03 |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |
| 128                                     | Strip       |       | 1.04E+02  |          |           |               |          |          |               |          |          |           |           |           |          |           |           |           |          |          |           |          |          |

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. Strip in same units as solution samples, test vessel was stripped with 300 ml of 8 N HNO3.

\* Am-241 values through 223 day solution sample calculated from Pu-239+Pu-240 and Am-241+Pu-238 values using Pu-238/(Pu-239+Pu-240) activity ratio of 2.073.

TABLE A.6

TEST TYPE: 1 LASER PUNCTURE

TEST IDENTIFICATION: J-8-19

| DAY | SAMPLE TYPE | pH    | U         |             | Pu-239+Pu-240 |             | Am-241+Pu-238 |             | Am-241   |             | Pu-238   |             | Ce-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 | 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   | Sol         | 5.636 | 3.00E-03  |             | 1.44E+00      |             | 4.93E+00      |             |          |             |          | 2.21E+00    |          | 1.65E+04    |          | 1.68E+03    |          |             |
| 5   | Sol         | 6.147 | 4.00E-03  |             | 2.21E+00      |             | 5.86E+00      |             |          |             |          | 2.39E+00    |          | 2.40E+05    |          | 2.47E+04    |          |             |
| 5   | Rod         |       | 3.90E-02  |             | 2.88E+00      |             | 1.80E+01      |             |          |             |          | 1.17E+01    |          | 5.86E+03    |          | <9.00E+02   |          |             |
| 15  | Sol         | 6.240 | 1.70E-02  |             | 7.07E+01      |             | 1.95E+02      |             |          |             |          | 9.59E+01    |          | 6.40E+05    |          | 7.43E+04    |          |             |
| 30  | Sol         | 6.577 | 7.00E-03  |             | 8.42E+01      |             | 2.53E+02      |             |          |             |          | 1.13E+02    |          | 9.41E+05    |          | 1.07E+05    |          |             |
| 30  | Rod         |       | 5.00E-01  |             | 8.33E+02      |             | 2.95E+03      |             |          |             |          | 1.77E+03    |          | 8.33E+04    |          | 1.01E+04    |          |             |
| 60  | Sol         | 6.693 | 8.00E-03  |             | 5.36E+01      |             | 1.16E+02      | 3.56E+01    |          | 8.06E+01    |          | 1.76E+01    |          | 2.09E+06    |          | 2.43E+05    |          |             |
| 60  | Rod         |       | 1.60E-01  |             | 1.48E+02      |             | 6.89E+02      | 4.82E+02    |          | 2.11E+02    |          | 4.50E+02    |          | 5.72E+04    |          | 5.59E+05    |          |             |
| 61  | Strip #1    |       | 5.90E+00  |             | 1.27E+04      |             | 4.77E+04      | 2.92E+04    |          | 1.86E+04    |          | 2.86E+04    |          | 2.00E+06    |          | 2.11E+05    |          |             |
| 61  | Strip #2    |       | 1.30E+00  |             | 4.23E+02      |             | 2.75E+03      | 2.03E+03    |          | 7.21E+02    |          | 1.94E+03    |          | 5.05E+06    |          | 5.36E+05    |          |             |
| 61  | Strip #3    |       | 2.90E-01  |             | 3.11E+02      |             | 1.04E+03      | 5.72E+02    |          | 4.50E+02    |          | 5.41E+02    |          | 7.25E+04    |          | 8.51E+03    |          |             |
|     |             |       |           |             |               |             |               |             |          |             |          |             |          |             |          |             |          |             |
| DAY | SAMPLE TYPE | pH    | Co-60     |             | Sb-125        |             | Ru-106        |             | Cs-144   |             | Eu-154   |             | Mn-237   |             | Tc-99    |             | Br-90    |             |
|     |             |       | UNFILTER  | 0.4 um 18 A | UNFILTER      | 0.4 u 18 A  | UNFILTER      | 0.4 u 18 A  | UNFILTER | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER | 0.4 um 18 A |
| 1   | Sol         | 5.636 | 8.96E+02  |             |               |             |               |             |          |             |          |             |          |             |          |             |          |             |
| 5   | Sol         | 6.147 | 1.18E+03  |             |               |             |               |             |          |             |          |             |          |             |          |             |          |             |
| 5   | Rod         |       | <1.30E+03 |             |               |             |               |             |          |             |          |             |          |             |          |             |          |             |
| 15  | Sol         | 6.240 | 1.08E+03  |             |               |             |               |             |          |             |          |             |          |             |          |             |          |             |
| 30  | Sol         | 6.577 | 1.04E+03  |             |               |             |               |             |          |             |          |             |          |             |          |             |          |             |
| 30  | Rod         |       |           |             |               |             |               |             |          |             |          |             |          |             |          |             |          |             |
| 60  | Sol         | 6.693 | 1.84E+03  |             |               |             |               |             |          |             |          |             |          |             |          | <2.30E+01   |          | 3.23E+04    |
| 60  | Rod         |       |           |             |               |             |               |             |          |             |          |             |          |             |          | <4.10E+02   |          | 7.21E+03    |
| 61  | Strip #1    |       | 7.61E+03  |             |               |             |               |             |          | 3.49E+04    |          |             |          |             |          | <1.40E+03   |          | 5.81E+05    |
| 61  | Strip #2    |       |           |             |               |             |               |             |          |             |          |             |          |             |          | <5.90E+03   |          | 3.80E+04    |
| 61  | Strip #3    |       |           |             |               |             |               |             |          |             |          |             |          |             |          | <6.30E+02   |          | 1.36E+04    |

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.  
 Strip #1 in same units as solution samples, specimen pedestal, 50 ml of 8 M HNO3.  
 Strip #2 in same units as solution samples, test vessel, 250 ml of 0.1 M HNO3.  
 Strip #3 in same units as solution samples, test vessel, 25 ml of 8 M HNO3 following Strip #2.

TABLE A.7

TEST TYPE: UNDEFECTED

TEST IDENTIFICATION: H-6-1

| DAY                                     | SAMPLE TYPE | pH    | U         |           |           | Pu-239+Pu-240 |          |          | Am-241+Pu-238 |          |          | Am-241    |           |           | Ce-244    |           |           | Ce-137   |          |           | Ce-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      | UNFILTER  | 0.4 um    | 18 A      | UNFILTER | 0.4 um   | 18 A      | UNFILTER | 0.4 um | 18 A      |
| 1                                       | Sol         | 5.875 | 3.00E-03  |           |           | 7.66E-01      |          |          | 1.76E+00      |          |          |           |           | 5.86E-01  |           |           | 3.21E+03  |          |          |           |          |        | 2.95E+02  |
| 4                                       | Sol         | 6.430 | 2.00E-03  |           |           | 8.56E-01      |          |          | 2.57E+00      |          |          |           |           | 8.56E-01  |           |           | 3.37E+03  |          |          |           |          |        | 3.72E+02  |
| 4                                       | Rad         |       | 7.00E-02  |           |           | <4.50E+00     |          |          | 3.33E+01      |          |          |           | 3.33E+02  |           |           |           | 5.86E+03  |          |          |           |          |        | 5.41E+02  |
| 15                                      | Sol         | 6.210 | 2.00E-03  |           |           | 1.13E+00      |          |          | 2.70E+00      |          |          |           |           | 7.21E-01  |           |           | 3.05E+03  |          |          |           |          |        | 3.08E+02  |
| 30                                      | Sol         | 6.311 | 2.00E-03  |           |           | 4.95E+00      |          |          | 9.91E+00      |          |          |           |           | 1.80E+00  |           |           | 3.15E+03  |          |          |           |          |        | 3.06E+02  |
| 30                                      | Rad         |       | <2.00E-02 |           |           | <4.50E+00     |          |          | 3.11E+01      |          |          |           |           | 1.67E+01  |           |           | 8.56E+02  |          |          |           |          |        | 2.89E+02  |
| 60                                      | Sol         | 6.205 | 3.00E-03  |           |           | 8.11E+00      |          |          | 1.49E+01      |          |          |           |           | 1.17E+00  |           |           | 3.59E+03  |          |          |           |          |        | 2.89E+02  |
| 60                                      | Rad         |       | 7.10E-02  |           |           | 2.25E+00      |          |          | 5.45E+01      |          |          |           |           | 3.38E+01  |           |           | <5.40E+02 |          |          |           |          |        | <3.90E+02 |
| 90                                      | Sol         | 6.137 | 2.00E-03  |           |           | 1.22E+01      |          |          | 2.21E+01      |          |          |           |           | 1.33E+00  |           |           | 3.94E+03  |          |          |           |          |        | 3.68E+02  |
| 120                                     | Sol         | 6.085 | 2.00E-03  |           |           | 1.44E+01      |          |          | 2.57E+01      |          |          |           |           | 1.33E+00  |           |           | 4.07E+03  |          |          |           |          |        | 3.57E+02  |
| 120                                     | Rad         |       | <1.00E-02 |           |           | 7.21E+00      |          |          | 8.11E+01      |          |          |           |           | 4.05E+01  |           |           | <5.00E+02 |          |          |           |          |        | <3.80E+02 |
| 150                                     | Sol         | 5.898 | 6.00E-03  |           |           | 1.53E+01      |          |          | 2.52E+01      |          |          |           |           | 9.01E-01  |           |           | 3.73E+03  |          |          |           |          |        | 2.51E+02  |
| 180                                     | Sol         | 6.140 | 3.00E-03  |           |           | 1.76E+01      |          |          | 2.97E+01      |          |          |           |           | 1.80E+00  |           |           | 4.20E+03  |          |          |           |          |        | 3.34E+02  |
| 180                                     | Rad         |       | 2.00E-02  |           |           | 8.56E+00      |          |          | 8.42E+01      |          |          |           |           | 4.37E+01  |           |           | 5.41E+02  |          |          |           |          |        | <3.70E+02 |
| 210                                     | Sol         | 5.935 | 2.00E-03  |           |           | 2.03E+01      |          |          | 3.15E+01      |          |          |           |           | <4.50E-01 |           |           | 4.03E+03  |          |          |           |          |        | 3.01E+02  |
| 252                                     | Sol         | 6.054 | 2.00E-03  | <1.00E-03 | <1.00E-03 | 1.94E+01      | 1.98E+01 | 1.67E+01 | 3.06E+01      | 2.93E+01 | 2.43E+01 | <9.00E-01 | <9.00E-01 | <9.00E-01 | <9.00E-01 | <9.00E-01 | <9.00E-01 | 2.15E+03 | 2.15E+03 | 1.60E+03  |          |        | <1.60E+02 |
| 252                                     | Rad         |       | 2.00E-03  |           |           | 1.33E+01      |          |          | 1.11E+02      |          |          | 9.44E+01  |           | 5.41E+01  |           |           | 1.00E+03  |          |          |           |          |        | <6.30E+02 |
| 252                                     | Strip       |       | 9.00E-03  |           |           | 1.53E+00      |          |          | 1.38E+01      |          |          | 1.53E+01  |           | 8.56E+00  |           |           | 1.89E+02  |          |          |           |          |        | <6.30E+01 |
| TEST RESTARTED IN FRESH DEIONIZED WATER |             |       |           |           |           |               |          |          |               |          |          |           |           |           |           |           |           |          |          |           |          |        |           |
| 15                                      | Rad         |       | 2.30E-01  |           |           | 4.50E+00      |          |          | 3.42E+01      |          |          | 3.87E+01  |           | 2.07E+01  |           |           | <6.30E+02 |          |          |           |          |        | <6.30E+02 |
| 15                                      | Sol         | 6.054 | 1.30E-02  | 3.00E-03  | 3.00E-03  | 3.15E+00      | 2.70E+00 | 1.33E+00 | 1.08E+01      | 4.50E+00 | 2.25E+00 | 4.05E+00  | 1.33E+00  | <9.00E-01 | 4.50E+00  | 4.50E-01  | <4.50E-01 | 3.32E+02 | 1.60E+02 | <1.00E+02 |          |        | <7.20E+01 |
| 70                                      | Rad         |       | 1.10E-01  |           |           | 3.15E+00      |          |          | 7.64E+01      |          |          | 6.31E+01  |           | 4.95E+01  |           |           | <1.30E+03 |          |          |           |          |        | <1.20E+03 |
| 70                                      | Sol         | 6.185 | 1.10E-02  | 4.00E-03  | 2.00E-03  | 6.31E+00      | 4.95E+00 | 3.15E+00 | 1.67E+01      | 7.64E+00 | 4.95E+00 | 5.41E+00  | 9.01E-01  | <9.00E-01 | 4.05E+00  | 3.15E-01  | <9.00E-01 | 6.76E+02 | 2.89E+02 | <3.20E+02 |          |        | <1.20E+02 |
| 128                                     | Sol         | 5.659 | 3.00E-03  | 3.00E-03  | 2.00E-03  | 5.86E+00      | 6.31E+00 | 4.95E+00 | 9.91E+00      | 9.44E+00 | 7.21E+00 | <1.40E+00 | <1.40E+00 | <1.40E+00 | <4.50E-01 | <4.50E-01 | <4.50E-01 | 4.09E+02 | 3.96E+02 | 2.56E+02  |          |        | <8.10E+01 |
| 128                                     | Rad         |       | 8.70E-02  |           |           | 3.92E+00      |          |          | 7.12E+01      |          |          | 3.74E+01  |           | 3.51E+01  |           |           | 7.03E+02  |          |          |           |          |        | <6.30E+02 |
| 128                                     | Strip       |       | <5.00E-03 |           |           | 4.05E-01      |          |          | 3.24E+00      |          |          | 2.70E+00  |           | 1.71E+00  |           |           | 6.22E+01  |          |          |           |          |        | <6.30E+01 |

| DAY                                     | SAMPLE TYPE | pH    | Co-60     |          |           | Bb-125   |        |      | Pu-106   |        |      | Ce-144   |        |           | Eu-154   |        |           | Np-237   |        |      | Tc-99    |        |           |
|---|-------------|-------|-----------|----------|-----------|----------|--------|------|----------|--------|------|----------|--------|-----------|----------|--------|-----------|----------|--------|------|----------|--------|-----------|
|   |             |       | UNFILTER  | 0.4 um   | 18 A      | UNFILTER | 0.4 um | 18 A | UNFILTER | 0.4 um | 18 A | UNFILTER | 0.4 um | 18 A      | UNFILTER | 0.4 um | 18 A      | UNFILTER | 0.4 um | 18 A | UNFILTER | 0.4 um | 18 A      |
| 1                                       | Sol         | 5.875 | 8.33E+02  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 4                                       | Sol         | 6.430 | 1.12E+03  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 4                                       | Rad         |       | <4.80E+02 |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 15                                      | Sol         | 6.210 | 1.07E+03  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 30                                      | Sol         | 6.311 | 1.07E+03  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 30                                      | Rad         |       | 9.91E+02  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 60                                      | Sol         | 6.205 | <6.30E+02 |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 60                                      | Rad         |       | 9.91E+02  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 90                                      | Sol         | 6.137 | 1.07E+03  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 120                                     | Sol         | 6.085 | 1.10E+03  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 120                                     | Rad         |       | <6.30E+02 |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 150                                     | Sol         | 5.898 | 8.83E+02  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 180                                     | Sol         | 6.140 | 1.14E+03  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 180                                     | Rad         |       | <6.30E+02 |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 210                                     | Sol         | 5.935 | 1.01E+03  |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 252                                     | Sol         | 6.054 | 8.92E+02  | 1.02E+03 | 1.38E+02  |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 252                                     | Rad         |       | <1.00E+03 |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 252                                     | Strip       |       | <1.00E+02 |          |           |          |        |      |          |        |      |          |        | <2.90E+02 |          |        | <1.40E-02 |          |        |      |          |        | <4.50E+00 |
| TEST RESTARTED IN FRESH DEIONIZED WATER |             |       |           |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 15                                      | Rad         |       |           |          |           |          |        |      |          |        |      |          |        | <2.90E+03 |          |        |           |          |        |      |          |        |           |
| 15                                      | Sol         | 6.054 | 1.53E+02  | 1.26E+02 | <1.20E+02 |          |        |      |          |        |      |          |        | <2.90E+02 |          |        |           |          |        |      |          |        |           |
| 128                                     | Sol         | 5.659 | 2.75E+02  | 1.44E+02 | <1.00E+02 |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 128                                     | Rad         |       | <1.00E+03 |          |           |          |        |      |          |        |      |          |        |           |          |        |           |          |        |      |          |        |           |
| 128                                     | Strip       |       | <1.00E+02 |          |           |          |        |      |          |        |      |          |        |           |          |        | <4.50E-01 |          |        |      |          |        | <4.50E+00 |

UNITS: Solution samples (Sol) in pCi/ml for all but Uranium, ug/ml for Uranium.  
 Rad samples in pCi/rad for all but Uranium, ug/rad for Uranium.  
 Strip in same units as solution samples, test vessel was stripped with 300 ml of 8 M HNO3.



TABLE A.9

TEST TYPE: UNDEFECTED

TEST IDENTIFICATION: J-8-12

| DAY | SAMPLE TYPE | pH    | U        |             | Pu-239+Pu-240 |             | Am-241+Pu-238 |             | Am-241   |             | Pu-238   |             | 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 | UNFILTER | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER  | 0.4 um 18 A | UNFILTER | 0.4 um 18 A | UNFILTER |
| 1   | Rod         |       |          |             | (Alpha <11.7) |             |               |             |          |             |          |             |          |             |           |             |          |             |          |
| 1   | Sol         | 6.480 |          |             | 3.60E+00      |             | 1.17E+01      |             |          |             |          | 4.95E+00    |          |             | 1.02E+03  |             |          | <4.30E+02   |          |
| 5   | Sol         | 6.395 |          |             | 8.54E+00      |             | 3.06E+01      |             |          |             |          | 1.13E+01    |          |             | 8.29E+02  |             |          | 1.23E+02    |          |
| 5   | Rod         |       |          |             | 1.17E+00      |             | 1.71E+01      |             |          |             |          | 1.35E+01    |          |             | <3.70E+02 |             |          | 1.55E+02    |          |
| 15  | Sol         | 6.419 | 1.00E-02 |             | 1.13E+01      |             | 3.24E+01      |             |          |             |          | 8.11E+00    |          |             | 2.22E+03  |             |          | <4.30E+02   |          |
| 29  | Sol         | 6.790 | 1.60E-02 |             | 2.30E+01      |             | 5.54E+01      |             |          |             |          | 5.86E+00    |          |             | 3.00E+03  |             |          | 2.65E+02    |          |
| 29  | Rod         |       | 4.80E-02 |             | 3.51E+00      |             | 5.86E+01      |             |          |             |          | 4.50E+01    |          |             | <4.10E+02 |             |          | <4.10E+02   |          |
| 60  | Sol         | 6.055 | 6.00E-03 |             | 3.78E+01      |             | 8.15E+01      |             |          |             |          | 1.35E+00    |          |             | 4.68E+03  |             |          | 4.43E+02    |          |
| 60  | Rod         |       | 3.90E-01 |             | 1.98E+02      |             | 9.32E+02      |             |          |             |          | 5.54E+02    |          |             | 2.50E+04  |             |          | 2.43E+03    |          |
| 90  | Sol         | 6.586 | 6.00E-03 |             | 5.09E+01      |             | 1.14E+02      |             | 6.31E+00 |             | 1.08E+02 | 2.25E+00    |          |             | 5.81E+03  |             |          | 5.50E+02    |          |
| 90  | Rod         |       | 3.90E-01 |             | 5.05E+01      |             | 3.05E+02      |             | 1.95E+02 |             | 1.11E+02 | 1.73E+02    |          |             | 1.08E+03  |             |          | <3.90E+02   |          |
| 91  | Strip #1    |       | 2.70E+00 |             | 1.69E+03      |             | 6.80E+03      |             | 2.31E+03 |             | 4.50E+03 | 2.18E+03    |          |             | 1.56E+05  |             |          | 1.69E+04    |          |
| 91  | Strip #2    |       | 5.70E+00 |             | 7.12E+02      |             | 4.73E+03      |             | 3.21E+03 |             | 1.55E+03 | 2.66E+03    |          |             | 3.96E+04  |             |          | <1.00E+04   |          |
| 91  | Strip #3    |       | 5.30E+00 |             | 2.68E+03      |             | 1.54E+04      |             | 9.32E+03 |             | 6.13E+03 | 8.29E+03    |          |             | 2.06E+05  |             |          | 2.09E+04    |          |

| DAY | SAMPLE TYPE | pH    | Co-60     |             | Sb-125   |            | Ru-106   |            | Ce-144   |             | Eu-154   |             | Nb-237   |             | Tc-99    |             | SrY-90   |             |          |
|-----|-------------|-------|-----------|-------------|----------|------------|----------|------------|----------|-------------|----------|-------------|----------|-------------|----------|-------------|----------|-------------|----------|
|     |             |       | UNFILTER  | 0.4 um 18 A | UNFILTER | 0.4 u 18 A | UNFILTER | 0.4 u 18 A | UNFILTER | 0.4 um 18 A | UNFILTER |
| 1   | Rod         |       | <7.20E+02 |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 1   | Sol         | 6.480 | 1.79E+02  |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 5   | Sol         | 6.395 | 5.05E+02  |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 5   | Rod         |       | <7.20E+02 |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 15  | Sol         | 6.419 | 6.98E+02  |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 29  | Sol         | 6.790 | 6.58E+02  |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 29  | Rod         |       | <6.80E+02 |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 60  | Sol         | 6.055 | 7.52E+02  |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 60  | Rod         |       |           |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 90  | Sol         | 6.586 | 9.37E+02  |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 90  | Rod         |       | <6.30E+02 |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 91  | Strip #1    |       | <1.50E+03 |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 91  | Strip #2    |       | <1.50E+04 |             |          |            |          |            |          |             |          |             |          |             |          |             |          |             |          |
| 91  | Strip #3    |       | 6.76E+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.  
 Strip #1 in same units as solution samples, specimen pedestal, 50 ml of 8 M HNO3.  
 Strip #2 in same units as solution samples, test vessel, 250 ml of 0.1 M HNO3.  
 Strip #3 in same units as solution samples, test vessel, 25 ml of 8 M HNO3 following Strip #2.

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