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of Laboratory Experiments

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FOR NNWSI FROM THE RESULTS  
OF LABORATORY EXPERIMENTS

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ABSTRACT

Results are presented for the dissolution of Turkey Point pressurized water reactor (PWR) spent fuel in J-13 well water at ambient hot cell temperatures. These results are compared with those previously obtained on Turkey Point fuel in deionized water, on H. B. Robinson PWR fuel in J-13 water, and by other workers using various fuels in dilute bicarbonate groundwaters. A model is presented that represents the conditions under which maximum dissolution of spent fuel could occur in a repository sited at Yucca Mountain, Nevada. Using an experimentally determined upper limit of 5 mg/l for uranium solubility in J-13 water, a fractional release rate of  $6.4 \times 10^{-8}$  per year is obtained by assuming that all water entering the repository carries away the maximum amount of uranium.

INTRODUCTION

The Nevada Nuclear Waste Storage Investigations Project (NNWSI) is investigating the suitability of the volcanic rocks at Yucca Mountain, Nye County, Nevada, for potential use as a disposal site for high-level nuclear waste. The repository horizon under study is the Topopah Spring Member of the Paintbrush Tuff. At the potential repository level, the tuff is densely welded and devitrified, and lies approximately 200 to 400 meters above the water table in the unsaturated zone [1].

Laboratory tests on spent fuel are being conducted to determine the release rates of radionuclides from waste packages emplaced in a repository at Yucca Mountain. Four specimen types are used to represent a range of fuel conditions that might occur: (1) fuel rod sections split open to expose bare fuel particles to the liquid phase, (2) rod sections with a 2.5-cm long by 150- $\mu$ m wide slit through the cladding, (3) rod sections with two 200- $\mu$ m laser-drilled holes through the cladding, and (4) undefected rod segments. The last three specimen types are fitted with water-tight end fittings containing a vent in the upper fitting to aid water entry through the defects. The undefected specimen type provides a control to show the portion of radionuclide release originating from residual contamination on the cladding exterior surfaces. The details of the test procedure are given in Reference 2.

The first series of tests used Turkey Point PWR fuel discharged in November 1975, with a burnup of approximately 27 MWd/kgM and estimated fission gas release of 0.3%. These tests were conducted in deionized water (DIW) with periodic sampling of the solution phase and rods contained in the test vessel to monitor the progression of radionuclide "plate-out." The tests were run for 8 months, terminated, and restarted for a further four-month testing period in fresh DIW. At the termination of both testing periods, the test specimens and test vessels were rinsed in DIW, and the test vessels were subjected to an acid strip procedure using 8M nitric acid. Uranium and the radionuclides recovered in each phase of the testing were separately measured so that a complete accounting of these species could be made. The results from this test series (DIW/IP) are contained in Reference 3.

The second series of tests is being conducted in J-13 well water, a dilute bicarbonate groundwater that is believed to be representative of the water to be encountered at the potential repository horizon [4]. Spent fuel from the H. B. Robinson reactor, discharged in May 1974, with a burnup of approximately 31 MWd/kgM and 0.2% gas release, was used in the first part of the Series 2 tests (J-13/HBR). Turkey Point fuel from the same pins that provided material for the DIW/IP Series 1 tests was used in the second part of the Series 2 tests (J-13/IP). All tests are conducted under air at ambient hot cell temperatures. Results for the H. B. Robinson fuel in J-13 water through the first termination are given in Reference 5. A summary of selected results from the Turkey Point tests in J-13 water through the first termination is given below. The restarted tests for both fuels are still in progress.

#### RESULTS OF TESTING TURKEY POINT FUEL IN J-13 WELL WATER

Figure 1 shows the uranium concentration in unfiltered solutions for each of the four specimen types as a function of time. Samples filtered through 0.4- $\mu$ m and 1.8-nm filters gave identical concentrations, showing that all of the uranium was in true solution. Fuel contained in cladding with induced defects produced much lower concentrations of uranium in solution than did bare fuel. In particular, much lower uranium concentrations were produced by the specimens with the laser-drilled holes. Reduced solution concentrations for the actinides Pu, Am, and Cm were also observed with the slit and the laser-drilled specimens relative to the bare fuel specimens. Similar results were also obtained in the DIW/IP and J-13/HBR test series, indicating that some control on the dissolution rate was provided when the fuel was contained in its original tight geometry in the defected cladding.

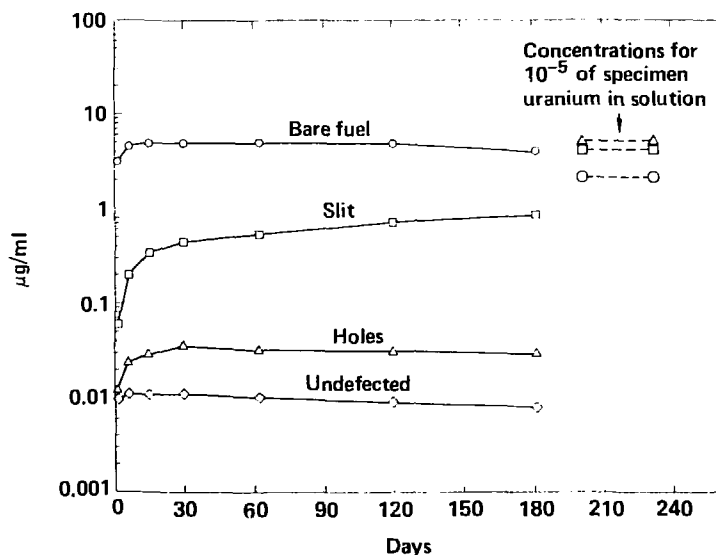


FIGURE 1. Uranium Concentrations (Unfiltered) for the First 181 Days for Tests Using Turkey Point Fuel in J-13 Water.

About half of the total uranium was recovered in periodic solution samples plus the final solution sample and half in the rinse and strip solutions. In contrast, for the other actinides only 12 to 18% was in solution and the bulk of the material was recovered in the acid strip. The initial release of material from the Turkey Point sample appears to be dominated by dispersion of fine particles of fuel, possibly produced during the sample preparation. The smallest of these appear to have rapidly dissolved while the larger settled rapidly to the bottom of the test vessel, since there is no evidence in the filtration data for any particulate matter in suspension. If this explanation is correct, there should be less uranium and actinides in the vessel rinse and strip samples on termination of the second portion of the tests. In the tests using Turkey Point fuel in deionized water the amount of material recovered in the rinse and strip on second termination was only 1/4 of that found in the first termination [3], suggesting much less dispersion of fines on restarting of the test.

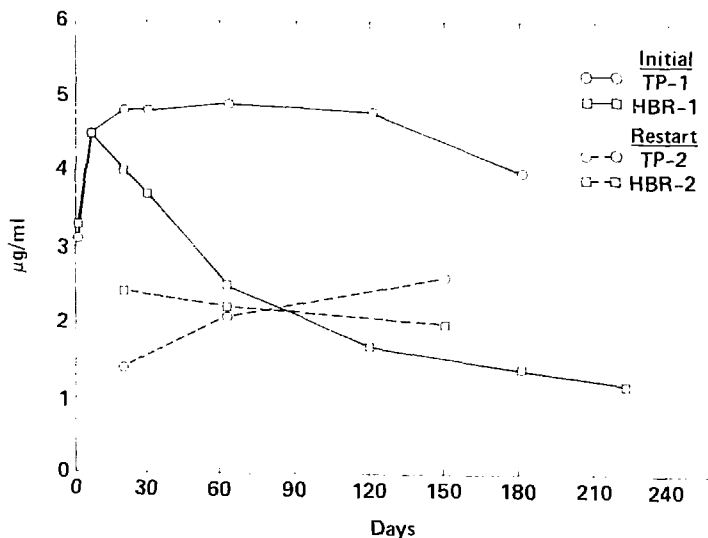


FIGURE 2. Uranium Concentration for Turkey Point and H. B. Robinson Bare fuels tested in J-13 Water.

Figure 2 compares the uranium concentration observed with the Turkey Point bare fuel in J-13 water to that observed with the H. B. Robinson bare fuel in J-13 water. The Turkey Point fuel produced a nearly constant concentration of 4.8 mg/l for 4 months, but decreased to 4.0 mg/l at 180 days. The H. B. Robinson fuel produced a maximum concentration of 4.5 mg/l after 6 days of testing, followed by a steady decrease to 1.4 mg/l at 181 days and 1.2 mg/l at 223 days. When the tests were restarted, the uranium concentrations were 2.4 mg/l for H. B. Robinson and 1.4 mg/l for Turkey Point fuels at 20 days; 2.2 mg/l and 2.1 mg/l, respectively, at 2 months; and 2.0 mg/l and 2.6 mg/l, respectively, at 5 months. The higher initial solution concentrations reached over both bare fuel types is thought to be due to a thin oxidized layer on the surface of the fuel formed by air exposure of the fuel after the fuel rods were sectioned. In this hypothesis, as the more highly oxidized surface layer dissolves and a steady-state condition is approached with an intermediate oxidation state or "plate-out" phase, uranium concentration decreases. The time between rod sectioning and

testing was approximately 5 years for the Turkey Point fuel versus a few months for the H. B. Robinson fuel. The more rapid decrease in uranium concentration over the H. B. Robinson fuel is thought to be due to the shorter time between rod sectioning and testing resulting in less of the higher oxidation state. After the first exposure to J-13 water, the surface oxidation states of the two fuels may be similar, since they exhibited more similar solubility behavior when the tests were restarted.

Data for total release of actinides, technetium, and cesium during the initial six-month testing period for the bare fuel samples are given in Table I. Total release of Pu, Am, and Cm was congruent and lower than that found for uranium. The preferential release observed for uranium relative to the actinides may be due to leaching of oxidized uranium from the surface of the fuel. If this is the case, the total release of uranium measured on termination of the restarted tests should be more similar to the actinide release.

TABLE I: RELEASE DATA FOR TURKEY POINT BARE FUEL IN J-13 WATER

	Uranium ( $\mu\text{g}$ )	$^{239+240}\text{Pu}$ (nCi)	$^{241}\text{Am}$ (nCi)	$^{244}\text{Cm}$ (nCi)	$^{99}\text{Tc}$ (nCi)	$^{137}\text{Cs}$ ( $\mu\text{Ci}$ )
Solution Samples	350.5	63	138	138	15 <sup>a</sup>	1010
Rod Samples	15.4	22	42	38	n.m.	2.7
Final Solution	1000	114	243	250	53	3090
Water Rinse	366	63	111	104	11.6	786
Acid Strip	960	1140	2180	1610	6.1	138
TOTAL	2691.9	1401	2714	2140	86	5027
Fractional Release (parts in 100,000)	11.67	7.30	6.87	7.44	32.4	295
% in Solution	50.2	12.6	14.0	18.1	79.1	81.5

<sup>a</sup> Includes 8 nCi in solutions on which Tc was actually measured and an estimate of 7 nCi removed in other solutions.

Technetium is expected to be very soluble under the conditions of these tests where there is free access of air to the solutions. It is, therefore, reasonable to assume that the technetium recovered from the sample rinse and acid strip solutions represents undissolved fine particles of spent fuel. If this is the case, we should be able to predict the uranium recovered in these samples by assuming they contain an inventory of technetium and uranium that is representative of the average in the fuel sample. The 17.7 nCi of Tc found in these samples for the bare fuel test implies that 1540  $\mu\text{g}$  of U should be in the rinse and acid strip samples; 1326  $\mu\text{g}$  was recovered in these samples, in good agreement with the prediction. A similar calculation using the technetium data from the H. B. Robinson bare fuel test gives the predicted amount of uranium as 3585  $\mu\text{g}$  as compared with 3300  $\mu\text{g}$  actually recovered. This evidence supports the arguments given above that the sample preparation method for bare fuel samples causes fine particles of fuel to become dislodged. Thus, the first run of a test using bare fuel will somewhat overestimate the "dissolution" rate of the fuel. The slit defect and laser puncture sample results should be much less affected by sample preparation.

The data for technetium in the solution samples may be compared with the uranium recovered from solution samples plus that calculated to have precipitated from solution to obtain an estimate of the degree of preferential dissolution of technetium relative to the bulk of the matrix. Using a peak uranium concentration of 4.8 mg/l and a final value of 4.0 mg/l for the first run of the Turkey Point bare fuel tests gives an estimate of 200  $\mu\text{g}$  of uranium precipitation. If this uranium is added to that recovered from solution samples, we estimate that 1550  $\mu\text{g}$  of uranium were once in solution. Assuming the average fuel inventory ratio of technetium to uranium, this corresponds to 17.8 nCi of technetium. The amount of technetium in solution was 68 nCi, indicating a preferential dissolution factor of 3.8 for technetium in the first run of the test. A similar calculation for the H. B. Robinson bare fuel test gives a factor of 8.7 for preferential technetium dissolution. Microstructural examination of cathodic vacuum etched ceramographic sections from both fuel types indicated a finer pre-irradiation grain size for the H. B. Robinson fuel (6  $\mu\text{m}$  for H. B. Robinson versus 25  $\mu\text{m}$  for Turkey Point). Partial diffusion of technetium from the oxide fuel matrix to the grain boundaries likely occurs during irradiation. The higher preferential dissolution factor for technetium from the H. B. Robinson fuel correlates well with the higher grain boundary fractional technetium inventory expected for the finer grain fuel.

Total measured fractional release for the initial run of the J-13/TP tests are compared to that of the J-13/HBR and DIW/TP tests in Table II. Actinide releases from both bare fuel tests in J-13 water were nearly identical. The slightly lower fractional uranium release with the H. B. Robinson fuel may be explained by precipitation (as suggested by Figure 2) onto the bare fuel bed covering the bottom of the vessel where it would not necessarily be completely recovered in the rinse or acid strip solutions. Higher actinide and  $^{99}\text{Tc}$  release in the DIW/TP tests have been correlated to extensive grain boundary dissolution, which was not observed in the J-13 water bare fuel tests [5]. Higher fractional release from Turkey Point fuel (both J-13 and DIW) slit and holes defect specimens relative to the corresponding J-13/HBR tests may be related to the longer pretesting exposure to air for the Turkey Point fuel.

The release of cesium from the bare fuel was comparable to the amount of fission gas release for the fuel, in agreement with the results of Johnson et al. for CANDU fuel [6]. The cesium release for the H. B. Robinson bare fuel was nearly four times the estimated gas release. Forsyth et al. [7] have found cesium release for boiling water reactor (BWR) high burn-up fuel to be 1.5 times the fission gas release. Variations in reported cesium release to fission gas release ratios may be due to a number of factors, including specimen preparation and storage conditions, and irradiation parameters for different fuel types. Calculation of the preferential release factor for Cs based on U that once was in solution (as done above for Tc) gives a factor of 36 for Turkey Point fuel versus 358 for H. B. Robinson fuel. As with technetium, the greater fractional Cs release with the H. B. Robinson fuel may be correlated to the finer grain size in this fuel.

The summary of release data for the first run of all sample types in both series of tests (Table II) shows that there is a larger total release from the bare fuel in DIW than in J-13. This is thought to reflect a difference in the amount of fine particles dispersed at the start of the test and not a true difference in "solubility" of the fuel in the two water types. The cesium release for Turkey Point bare fuel is similar in both waters and is probably a true reflection of the amount of readily soluble Cs in the fuel. The release of technetium appears to depend on the water type, with much lower release occurring in the J-13 water. Total release of actinides from the slit defect samples is generally two orders of magnitude less than that of the bare fuel, and the release from the hole defect specimens is another order of magnitude

TABLE II: SUMMARY OF TOTAL MEASURED FRACTIONAL RELEASE DATA  
(Units: parts per 100,000 of the test specimen inventory)

<u>Nuclide</u>	<u>Series<sup>a</sup></u>	<u>Bare Fuel</u>	<u>Slit</u>	<u>Holes</u>	<u>Control</u>
Uranium	J-13/HBR	5.63	0.05	0.005	0.009
	J-13/TP	11.7	0.65	0.030	0.011
	DIW/TP	21.2	0.07	0.032	0.010
239+240Pu	J-13/HBR	7.04	0.01	0.003	0.002
	J-13/TP	7.30	0.11	0.022	0.006
	DIW/TP	22.4	0.20	0.042	0.021
241Am	J-13/HBR	7.76	0.01	0.002	0.003
	J-13/TP	6.87	0.12	0.023	0.004
	DIW/TP	17.2	0.15	0.019	0.008
244Cm	J-13/HBR	6.54	0.01	0.001	0.002
	J-13/TP	7.44	0.13	0.026	0.005
	DIW/TP	21.5	0.35	0.026	0.006
237Np	J-13/HBR	<6.4	--	--	--
	J-13/TP	<7.4	--	--	--
	DIW/TP	18	0.2	--	--
137Cs	J-13/HBR	739	633	400	0.18
	J-13/TP	295	137	7.6	0.13
	DIW/TP	230	110	50	0.04
99Tc	J-13/HBR	22.2 <sup>b</sup>	2.4	<1.5	---
	J-13/TP	32.4	<13.7	<2	---
	DIW/TP	152	8.1	---	--

<sup>a</sup> J-13/HBR -- H. B. Robinson fuel tested in J-13 water [5].

J-13/TP - Turkey Point fuel tested in J-13 water.

DIW/TP -- Turkey Point fuel tested in deionized water [3].

<sup>b</sup> Includes an estimate of 22.5 nCi for Tc removed in solutions on which direct measurement was not made.

lower. Cesium release is only slightly lower for the defected samples in comparison to the bare fuel, an indication of the readily accessible nature of the rapid release component and the high solubility of Cs.

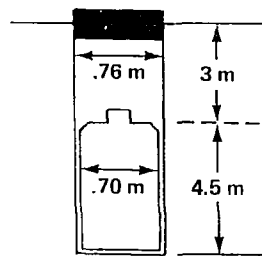
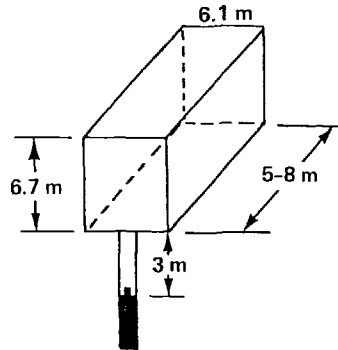
#### UPPER BOUND TO RELEASE RATES FROM SPENT FUEL UNDER NNWSI CONDITIONS

Based on our data and that of others [6,7,8], we have developed a model to estimate the upper bound to the release rate of radionuclides from spent fuel under NNWSI conditions. General corrosion rates for the stainless steel containment barrier are sufficiently low that the container will remain substantially intact for more than 10,000 years [8]. Localized corrosion mechanisms, such as stress corrosion cracking or crevice corrosion, may cause containers to develop breaches that allow water to enter the container. The most likely location for the breach to occur is at or near the final closure weld [8].

Figure 3 shows a schematic representation of a spent fuel container in a vertical emplacement hole. The hole is assumed to be lined with a carbon steel liner that allows water to collect in the hole. In the absence of the liner, water would be expected to drain away and not fill the hole [9]. If we assume that the container has a breach large enough



to allow free access of water to the interior and that the liner is intact, approximately 1800 liters of water could accumulate in the hole. The weight of fuel in the container is 3140 kg, giving a fuel-to-water ratio of 1.74 kg/l. Our tests using Turkey Point bare fuel had a ratio of 0.11 kg/l, and those using H. B. Robinson fuel had a ratio of 0.33 kg/l. The solution concentration of uranium for both fuels on the restarted tests was about 2 mg/l, suggesting that solubility of uranium in the system was controlling the solution concentration and the dissolution of the matrix after initial dissolution of any oxidized surface films and equilibration of the system.



- ~ 40 m<sup>2</sup> average open area per package

- Water volume to open area per year

Influx rate	Volume per year
1 mm/yr	40ℓ
2 mm/yr	80ℓ
8 mm/yr	320ℓ

- For 1 mm/yr infiltration rate, water volume directly to borehole is 0.5ℓ/yr

- Void volume per hole: 1800ℓ

- Fuel weight per package: 3140 kg UO<sub>2</sub>

- When borehole is full of water: R = 1.74 kg/ℓ

FIGURE 3. Water-to-Fuel Ratio and Vertical Emplacement Configuration.

Forsyth et al. [7] used a fuel-to-water ratio of 0.08 kg/l, similar to our Turkey Point tests, and found a concentration of about 1 mg/l in water with similar bicarbonate concentration to J-13. Johnson [10] found uranium concentrations of 1.2 mg/l to 4.5 mg/l in "KBS" water with bicarbonate 2.5 times higher than J-13, and 0.2 mg/l to 3 mg/l in granite groundwater with bicarbonate half of the J-13 value. Both sets of experiments used a fuel-to-water ratio of 0.72 kg/l. Taken together, the data strongly suggest that the ambient temperature solubility of uranium over spent fuel in dilute bicarbonate solutions is 5 mg/l or less. We will use this value as the matrix solubility control for dissolution of spent fuel in our model calculations.

Control of matrix solubility at less than 5 mg/l of uranium does not necessarily imply that matrix dissolution ceases when solution concentrations reach that value. The dissolution might still continue, accompanied by precipitation of a secondary uranium-bearing phase. In our tests, technetium can be used to monitor whether dissolution of the matrix continues once the uranium in solution has reached the solubility limit. Table III shows data for technetium in solution for the first run of both the H. B. Robinson and Turkey Point bare fuel tests. Solution concentrations are constant to within the precision of the measurements for both tests. Since solution samples have been taken at 2, 4, and

6 months in each case, we would expect decreasing concentrations of technetium if no further dissolution of the fuel matrix occurred when fresh J-13 water was added to replace the volume removed during sampling. The fact that solution concentrations are essentially constant implies some matrix dissolution has occurred in the added water. For H. B. Robinson, the total technetium increase from day 63 to day 223 was 27 nCi; for Turkey Point the increase from day 62 to day 181 was 8 nCi. Applying the preferential dissolution factors calculated above, this would correspond to 249 and 182  $\mu\text{g}$  of uranium dissolution respectively. In the H. B. Robinson test, a total of 60 ml of new water was added between day 63 and day 223. If this water had dissolved 5 mg/l of uranium, a total of 300  $\mu\text{g}$  of uranium should have dissolved. In the Turkey Point test, 40 ml of water was added, implying that 200  $\mu\text{g}$  of uranium should have dissolved. In both cases the results based on assuming that uranium dissolved until the new water reached 5 mg/l concentration are close to, and slightly higher than, those based on assuming that technetium was added to solution with the average preferential release factor remaining constant throughout the test. We would expect the preferential release factor to decrease with time as the grain-boundary inventory is depleted, and eventually to observe congruent technetium and uranium release. Once congruent release is achieved we will be able to base our calculations on measured matrix dissolution rates and will revise the release rate model accordingly.

TABLE III: CONCENTRATION OF TECHNETIUM IN SOLUTION  
FOR TURKEY POINT AND H. B. ROBINSON TESTS IN J-13 WATER,  
In pCi/ml.

H. B. Robinson Bare Fuel		Turkey Point Bare Fuel	
Sampling Day	[Tc]	Sampling Day	[Tc]
63	450	62	198
181	468	120	203
223	450	181	212

Our initial model also assumes that the oxide fuel matrix is not substantially degraded by oxidation or other mechanisms that are not observed in short-term laboratory leaching tests and that all actinides are therefore released congruently. Under the assumptions of the model, the rate at which the matrix of the spent fuel will dissolve depends linearly on the rate of delivery of water to the fuel. The infiltration rate at Yucca Mountain has been estimated to be less than 1 mm/yr [9]. If we assume a flow rate of 1 mm of water per year through the repository horizon and that all of the water that intersects the emplacement drift excavation area is able to collect into the package emplacement holes, we can calculate a rate of influx of new water into the borehole. The flow rate calculated by this method will be far higher than that expected to actually occur in the unsaturated zone, since water flow should be channeled around rather than through the large openings in the rock [11].

Figure 3 shows a model of the emplacement drift with a vertical package borehole. The excavation area is 40  $\text{m}^2$  per package, corresponding to a delivery rate of 40 l/yr of new water to the borehole. Dissolution of U to give a concentration of 5 mg/l would result in a fractional dissolution rate of  $6.4 \times 10^{-8}$  per year. Using a borehole plus container void volume of 1800 l and an inflow rate of 40 l/yr, it would take 45 years for the borehole to fill. During this time, no release would occur under the model assumption of an intact borehole liner (inflow opening near the top of the liner). After the borehole

plus container are filled with water, new water is assumed to displace U-saturated water and to itself reach saturation. This would produce a long-term release rate of  $6.4 \times 10^{-8}$  per year for those nuclides whose release is controlled by matrix solubility.

The time of 45 years to fill the void volume is much longer than our longest test interval of 223 days. The present ratio of model extrapolation time to experiment length is 74; by conducting a 3 year test this ratio can be decreased to 15, and a 5 year test reduces the ratio to 9.

There are four types of release that are not accounted for by the model discussed above. First,  $^{14}\text{C}$  present in the Zircaloy can be released because of contact with air as soon as the container is breached. Measurements at 275°C on Turkey Point fuel showed an initial release of about 0.3% of the calculated  $^{14}\text{C}$  inventory [12]. This material is thought to come from the oxidized layer on the cladding surface. Further release was very low. The pulsed  $^{14}\text{C}$  release will be controlled by the container failure rate. A container failure rate of 0.1% per year would control the release rate of  $^{14}\text{C}$  to 0.3 parts in 100,000 per year.

The second component of release from non-fuel-matrix sources is the release of activation products contained in the cladding matrix and in fuel assembly parts. The rate of corrosion of Zircaloy under expected Yucca Mountain conditions is being determined to provide data on the release rate of  $^{14}\text{C}$  and other activation products. The release rate of activation products from other metal components will be determined using the corrosion rates obtained as part of the metal containment barrier testing program.

The third component of the release rate model is that for the gap/grain boundary components that are released initially at a rapid rate, but then more slowly until finally controlled by the matrix fuel solubility. The highest initial release rate is for cesium, and the amount of release is similar to the fission gas release for the fuels. However, after the 300-year minimum 10 CFR 60 required [13] containment period,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  will have essentially decayed away, and the only remaining radioactive  $^{135}\text{Cs}$  isotope accounts for only about 0.01% of the total 300-year curie activity of a spent PWR fuel assembly [14]. We will assume that the average gap/grain boundary inventory is 0.5% and that all of this material is released in the first year following breach of the cladding. For a cladding breach rate of 0.1% per year, the release rate from the repository ensemble is dominated by the pulsed release for the 1000-year period over which the pins are being breached. The rate would be  $5.55 \times 10^{-6}$  per year. After this component is exhausted, the release rate would rapidly decrease to the much lower rate controlled by matrix dissolution. Release rates for technetium are expected to be intermediate between those of cesium and the matrix.

A fourth component of the release rate model to be developed is the possibility of slow degradation of the oxide fuel matrix by oxidation. Such oxidation of the  $\text{UO}_2$  spent fuel matrix phase would provide a mechanism for accelerated release of soluble long-lived radionuclides such as  $^{99}\text{Tc}$ ,  $^{135}\text{Cs}$ ,  $^{129}\text{I}$ ,  $^{237}\text{Np}$ ,  $^{79}\text{Se}$  and  $^{14}\text{C}$ . The kinetics for oxidation of the spent fuel matrix are known to be extremely slow at post-containment repository temperatures (<130°C) and may not be significant; however, studies have been initiated to better characterize the low temperature oxidation behavior of spent fuel.

In order to improve the model described above, several types of data are needed. The most important are (1) the dependence of uranium solubility on temperature, (2) the rate of failure of the containment barrier, (3) the rate of breach of cladding and (4) the rate of oxidation of the  $\text{UO}_2$  fuel matrix. Experiments and tests are in progress to improve our understanding of all of these processes and to provide the needed data.

## REFERENCES

1. S. Sinnock, Y. T. Lin, and J. P. Brannen, SAND84-1492, Sandia National Laboratories, Albuquerque, NM (1984).
2. C. N. Wilson, HEDL-TC-2353-3, Hanford Engineering Development Laboratory, Richland, WA (1984).
3. C. N. Wilson, HEDL-TME-84-30, Hanford Engineering Development Laboratory, Richland, WA (1985).
4. V. M. Oversby, UCRL-53629, Lawrence Livermore National Laboratory, Livermore, CA (1985).
5. C. N. Wilson, and V. M. Oversby, UCRL-91464, Lawrence Livermore National Laboratory, Livermore, CA (1985).
6. L. H. Johnson, K. I. Burns, H. Joling, and C. J. Moore, Nucl. Technol 63, 470 (1983).
7. R. S. Forsyth, K. Svanberg, and L. Werme, in Proceedings of the Third Spent Fuel Workshop, L. Werme editor, KBS Technical Report No. 83-76, Swedish Nuclear Fuel Supply Co., 1983 (issued March 1984).
8. V. M. Oversby, and R. D. McCright, UCRL-91257, Lawrence Livermore National Laboratory, Livermore, CA (1984).
9. P. Montazer, and W. E. Wilson, USGS-WRI-84-4345, U.S. Geological Survey Water-Resources Investigations, Lakewood, CO (1984).
10. L. H. Johnson, AECL-6837, Atomic Energy of Canada Limited, Pinawa, Manitoba, Canada (1982).
11. J. A. Fernandez, and M. D. Freshley, SAND83-1778, Sandia National Laboratories, Albuquerque, NM (1984).
12. R. A. Van Konyenburg, C. F. Smith, H. W. Culham, and C. H. Otto, Jr., UCRL-90855, Rev. 1, Lawrence Livermore National Laboratory, Livermore, CA (1984).
13. Code of Federal Regulations, 10CFR60, Title 10, Chapter 1, Part 60 (June 30, 1983).
14. C. W. Alexander et al., ORNL/TM-6008, Oak Ridge National Laboratory, Oak Ridge, TN (October, 1977).

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