SPENT FUEL CLADDING CONTAINMENT CREDIT TESTS

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ABSTRACT

Preliminary tests are being conducted to evaluate the effectiveness of defected cladding as a barrier to radionuclide release from spent fuel rods stored in a geologic repository. These tests are in support of the Nevada Nuclear Waste Storage Investigations (NNWSI) tuff repository project. Leach tests are performed in a text matrix that includes spent pressurized water reactor (PWR) fuel rod specimens with artificially induced cladding defects, specimens with intact cladding, and bare fuel with the cladding removed. The artificially induced cladding defects are made by laser drilling or sawing to give defect areas in the 10^4 μm^2 to 10^6 μm^2 range. Periodic samples of the leach solution and fused quartz rods contained in the test vessels are analyzed. Results for the first 180 days of testing are presented.

INTRODUCTION

The most likely mechanism for release of radionuclides from a geologic repository to the biosphere is dissolution into ground water and migration as a result of ground water flow. The repository horizon for the Nevada Nuclear Waste Storage Investigations (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. Most of the available quantitative data on the radionuclide release from spent fuel is derived from leach testing of bare fuel particles. Using such data to model the NNWSI repository, the "source term" for radionuclide transport may be unnecessarily conservative. The amount of conservatism would depend upon: 1) the portion of stored rods breached at a given time during the post-containment period, 2) the amount of inhibition to radionuclide release provided by breached cladding, and 3) the long-term stability of spent fuel rods in the repository environment.

For fuel currently being discharged from light water reactors (LWR), the portion of rods containing breaches is estimated to be in the 10^{-3} to 10^{-4} range.[1] The typical in-reactor breach is a series of small pin holes or a fine crack. The pin holes are typically only a few microns in size, and the cracks are typically several microns wide and a few millimeters to a few centimeters in length. However, larger cladding failures may occur. Spent fuel pool storage experience[2] suggests that radionuclide release rates from rods containing small breaches may be limited during repository storage if longer time processes (such as cladding corrosion or fuel swelling due to oxidation) do not cause a progressive loss of cladding integrity under NNWSI-proposed terminal storage conditions.

The Spent Fuel Cladding Containment Credit Tests[3] are being conducted at the Hanford Engineering Development Laboratory (HEDL) for the Lawrence Livermore National Laboratory's (LLNL) NNWSI Waste Package Task. The objective of these tests is to determine the effectiveness of cladding containing a defined and stable breach as a barrier to radionuclide release. The tests are conducted as a scoping study to provide initial data and to provide direction for future studies if a sigificant barrier effect for breached cladding is indicated.

TEST DESCRIPTION

Sections of pressurized water reactor (PWR) spent fuel rods with artificially induced cladding defects are comparatively leach tested in deionized water under air at ambient hot cell temperature (22°C to 28°C). The test matrix includes four specimen types: 1) rod segments with undefected cladding, 2) rod segments with small laser-drilled holes through the cladding to the fuel, 3) rod segments with a fine axially machined slit through the cladding, and 4) rod segments which are split open and the bare fuel and cladding separated. Cladding defect sizes per kg of fuel are $\sim 10^6$ μm^2/kg for the laser-drilled defects and $\sim 10^8$ μm^2/kg for the slit defect. This defect range compares with an expected 1 μm^2/kg to 10^5 μm^2/kg range for a typical in-reactor breach. Additional characteristics of the fuel and test specimens are given in Table I. A 60-day post-test metallographic section through a laser-drilled cladding defect is shown in Fig. 1.

The test apparatus is shown in Fig. 2. The bare fuel and cladding hulls are placed in a fused quartz basket. As a result of thermal cracking of the UO_2 pellets during irradiation, the bare fuel material is present as pellet pieces. The mean particle size of the pellet pieces is in the 2 mm to 3 mm range. The undefected, laser-drilled and slit-cladding specimens are fitted with end caps. These end caps provide a water-tight seal by compressing ethylene propylene O-rings against the cladding exterior. The upper end cap is vented to allow water entering the defect to fill the internal free volume of the specimen and completely water log the fuel. Several fused quartz rods are contained in each test vessel. These rods can be periodically removed to monitor radionuclide plateout during the tests.

Each test vessel is initially filled with 250 ml of deionized water to the approximate solution level shown in Fig. 2. Solution samples (10 ml) are removed at day 1, 5, 15, 30, and every 30 days thereafter. To maintain a constant solution level, 10 ml

TABLE I

Spent Fuel Specimen Characteristics[3]

Fuel Type:	PWR 15 x 15
Burnup:	25,500 to 27,700 MWd/MTU
Discharge Date:	November 1975
Fission Gas Release:	~0.3%
Initial Enrichment:	2.559 wt% ^{235}U
Initial UO_2 Density:	92% TD
Specimen Length:	5 inches
Hole Defect Specimen:	Two ~200-μm diameter laser-drilled holes
Slit Defect Specimen:	Axial slit, ~2 cm long by 150 μm wide

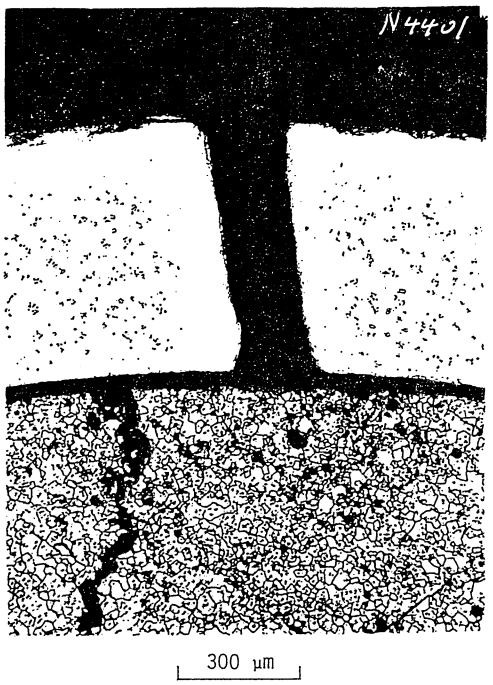

Fig. 1. Metallographic Section of a Laser-Drilled Cladding Defect After 60-Days Test Duration. No visable attack of the fuel by the leaching solution was observed. (Etched to reveal fuel microstructure.)

of fresh deionized water is added for each solution sample removed. Fused quartz rods are removed at day 5, 30, 60, 120, and every 60 days thereafter. The rod samples are rinsed with fresh deionized water, stripped with HNO_3, and the strip solution analyzed. Each sample is radiochemically analyzed for alpha and gamma emitters by spectrographic methods, and for uranium by the laser-excited fluorescence method. Additional radiochemical analyses requiring chemical separations and/or filtering are being made on selected samples.

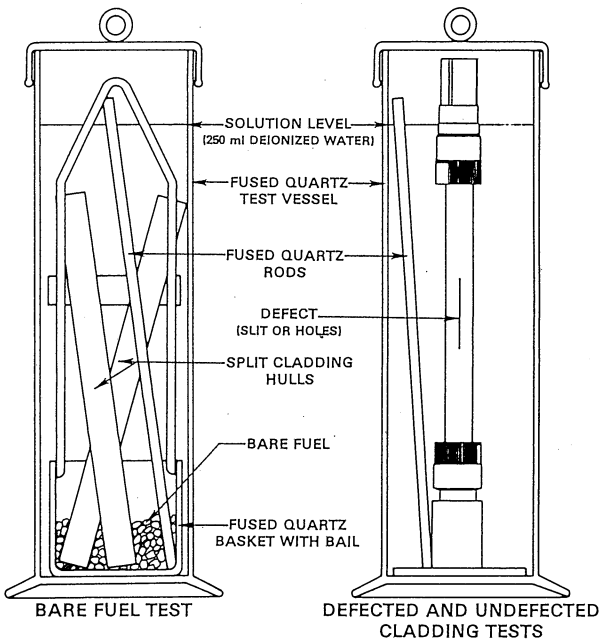

Fig. 2. Test Apparatus.

RESULTS AND DISCUSSION

Uranium, ^{239}Pu plus ^{240}Pu and ^{137}Cs results for solution and fused quartz rod samples through 180 days are given in Fig. 3. The solution concentration for each species that would result if 10^{-5} of the test specimen inventory were dissolved in the 250 ml of test solution is indicated on each of the solution data plots. The quantity of each species corresponding to 10^{-5} of the test specimen inventory is indicated on each fused quartz rod data plot. The 10^{-5} inventory values are given to relate the data to geologic repository release rate limits given in 10 CFR 60,[4] and are established using ORIGEN code-calculated inventory data for PWR fuel 10 years after discharge.[5] The 10 CFR 60 limits are based on 1000-year inventories. For uranium and plutonium, the 10-year and 1000-year inventories are similar; ^{137}Cs has decayed to a negligible level by 1000 years after disposal.

Data given in each plot for the "undefected" specimen indicate release levels for each species from cladding exterior surface contamination. The primary source of the cladding exterior contamination is sectioning and handling of the fuel specimens in contaminated hot cells. One exception is ^{60}Co, which is an activation product occurring in cladding exterior crud deposits. The cladding exterior surfaces were decontaminated to ~50-dpm smearable alpha prior to final specimen preparation in a clean hot cell. However, the remaining exterior contamination levels are most likely greater than the levels expected on whole fuel rods received at a repository.

It is interesting to note from the Fig. 3 data that most of the release in both bare fuel and defected cladding tests appears to have occurred during the first 30 days. Substantially reduced releases of uranium and plutonium were observed in the defected cladding tests in comparison with the bare fuel test.

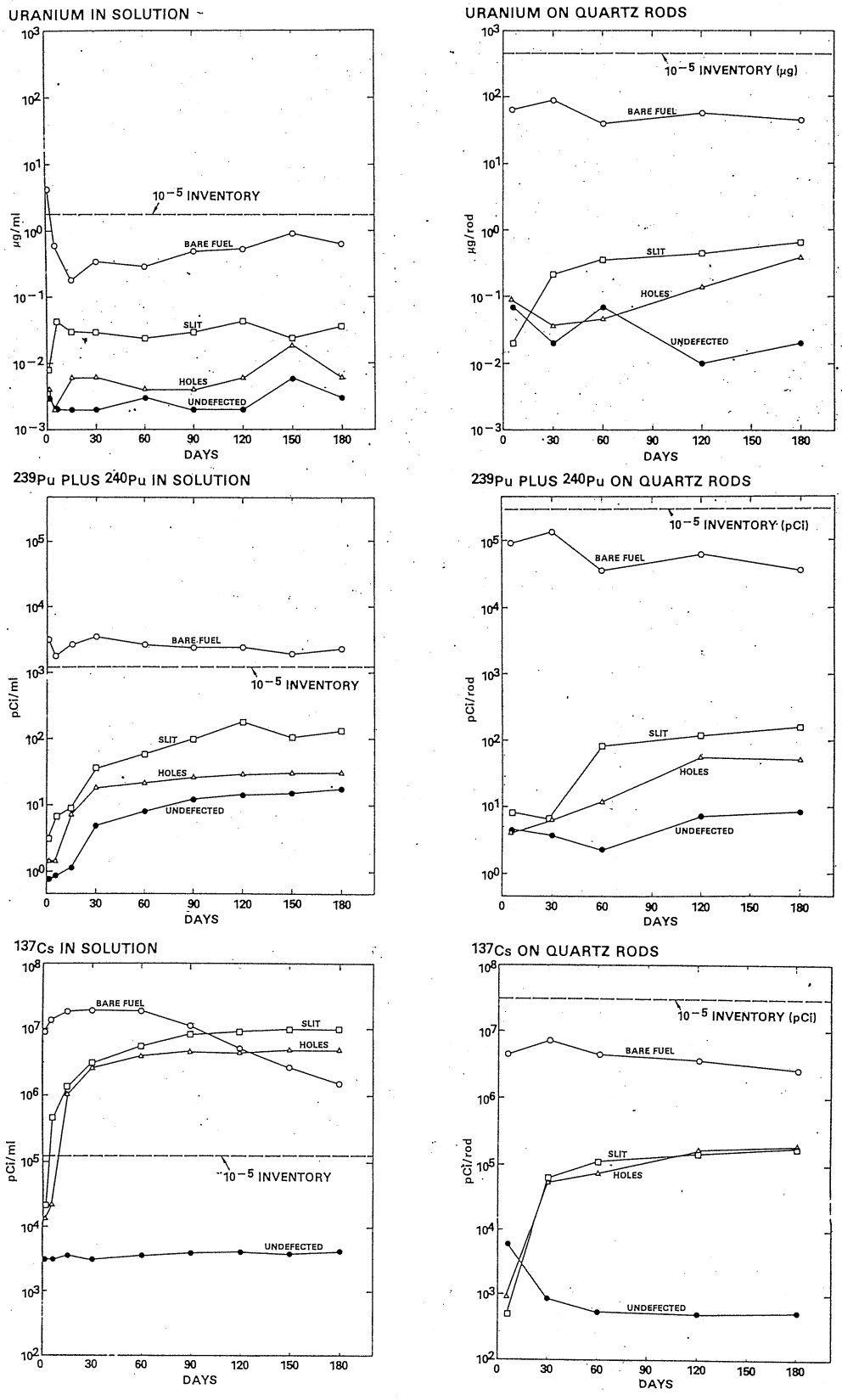

Fig. 3. Uranium, ^{239}Pu plus ^{240}Pu and ^{137}Cs Measured in Solution Samples and on Fused Quartz Rod Samples During the First 180 Days of Testing.

Reduced uranium release with the defected cladding specimens is particularly noteworthy since, after an initial preferential release of highly soluble fission products such as cesium, it is expected that UO_2 matrix dissolution will control radionuclide release. Localized solution oxygen depletion and saturation effects within the defected cladding may be factors contributing to the observed lower actinide release from these specimens.

The absence of well-defined solubility limits when "solution" sample results are compared to fused quartz rod sample results, suggests that the quantities of fuel source species measured in solution samples are not solution concentrations in equilibrium with a "plateout" phase. A possible explanation is that a portion of the fuel source species measured in solution samples is in a colloidal state. (However, all solution samples were clear.) A solution sample taken from the bare fuel test (202 days) was filtered through 0.4-μm and 18-Å filters to identify what portion of the radionuclide species released was in a colloidal state. Results are given in Table II. Most of the plutonium, the antimony, and approximately half the cesium passed through the 18-Å filter. However, only about 1% of the uranium, curium, europium, and cobalt passed the 18-Å filter, and most of the uranium was collected on the 0.4-μm filter. A substantial portion of the total radionuclide release from spent fuel to water may be tied up in colloidal phases. Planned future tests and sample analyses will further investigate the state of radionuclides released from spent fuel.

TABLE II

Radiochemical Data for 202-Day Solution Sample from Bare Fuel Test pCi/ml or (μg/ml)*

	Not Filtered	Filtered 0.4 μm	Filtered 18 Å
^{244}Cm	2.1×10^{4}	2.9×10^{3}	1.6×10^{2}
$^{239}Pu + ^{240}Pu$	2.2×10^{3}	2.2×10^{3}	1.8×10^{3}
^{154}Eu	5.5×10^{4}	7.7×10^{3}	7.7×10^{2}
^{137}Cs	1.4×10^{6}	9.4×10^{5}	5.6×10^{5}
^{125}Sb	2.0×10^{4}	2.3×10^{4}	2.4×10^{4}
^{60}Co	1.1×10^{5}	8.7×10^{4}	6.6×10^{2}
Uranium	(0.30)*	(0.003)*	(0.002)*

The relatively high solubility of cesium is reflected in its higher initial solution levels relative to the 10^{-5} inventory concentration in the bare fuel and defected cladding tests. Two unexpected observations are the quantities of cesium observed on fused quartz rods and the decrease in solution cesium content over the bare fuel after 60 days. Cesium on the rod samples may be contained in fine colloidal particles adsorbed on the rods. Partial association of cesium with colloidal particles is suggested by the filtered sample data in Table II. It is interesting to note that cesium coming out of the "solution" phase over the bare fuel is not coming out onto the fused quartz rods. The reaction of cesium with the leached fuel particles is a possible explanation, since the only other materials in the bare fuel test (zircaloy and fused quartz) are common to the other tests, and the fuel particle surfaces are more highly leached and exposed in the bare fuel test.

SUMMARY

A series of tests has been described in which radionuclide releases from PWR spent fuel rod segments containing artificially induced cladding defects are compared with radionuclide releases from an equal quantity of bare unclad fuel. Results for the first 180 days indicate that: 1) the quantity of radionuclides released from the fuel through the cladding defects was lower in comparison to the quantity released from the bare fuel, and 2) most of the observed release from both defected cladding specimens and the bare fuel specimen appears to have occurred during the first 30 days. Testing is continuing, and future sample analyses are planned to more comprehensively evaluate the state of released radionuclides and the degree to which defected cladding is a barrier to radionuclide release.

REFERENCES

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