SIMULATED DRY STORAGE TEST OF A SPENT PWR NUCLEAR FUEL ASSEMBLY IN AIR

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ABSTRACT

A test simulating dry storage of a pressurized water reactor (PWR) nuclear spent fuel assembly was conducted at the Nevada Test Site (NTS) in the Engine Maintenance and Disassembly (EMAD) shielded facility. The purpose was to investigate the behavior of Zircaloy-clad spent fuel in air between 200°C and 275°C. Electric heaters provided an axial temperature gradient consistent with that predicted for light-water reactor (LWR) fuel in dry storage. Thermocouples in the assembly were used to measure temperatures, which were programmed to simulate the declining decay heat with time in a spent fuel assembly.

Atmospheric air was used for the cover gas due to the interest in establishing regimes where air inleakage into an initially inert system would not cause potential fuel degradation. Cover gas samples were extracted monthly to determine fission gas concentrations as a function of time. The oxygen concentration was monitored to detect oxygen depletion, which could signal oxidation of the fuel.

The gas analyses indicated very low but detectable levels of krypton-85 during the first month of the test. A large increase (five orders of magnitude) in krypton-85 and the appearance of helium in the cover gas indicated that a fuel rod had breached during the second month of the test. Stress rupture calculations showed that the stresses and temperatures were too low to expect breaches to form in defect-free cladding; it is theorized that the breach occurred in a fuel rod weakened by a cladding or end cap defect.

Calculations based on the rate of krypton-85 release suggest that the diameter of the initial breach was ~0.3 μm. A post-test fuel examination will be performed to locate and investigate the cause of the cladding breach and to determine if detectable fuel degradation progressed after the breach occurred. This test demonstrated that the prospect for a small number of fuel rod breaches to develop during dry storage cannot be ruled out. The number is expected to be very small; this rod was the first to breach in over 6000 Zircaloy-clad rods that have been monitored in dry storage tests and demonstrations. The post-test evaluation will define the consequences of a fuel rod breach occurring in air cover gas at 270°C, followed by subsequent exposure to air at a prototypic descending temperature.

INTRODUCTION

Dry storage is an attractive option for spent fuel because of low maintenance requirements compared with wet storage. It has lower initial costs and is perceived to be a safe storage method. Dry storage will probably require an inert cover gas until the fuel cools below temperatures that could lead to spent fuel oxidation through cladding breaches. If a temperature is defined below which fuel oxidation is not significant, monitoring for inert gas verification can be discontinued. The reference temperature limit for spent fuel storage in air was proposed to be 250°C during the planning stages of this test.(1) Therefore, this test was initiated at 275°C to address overtemperatures in a dry storage system.

Spent fuel assembly B02 was selected for this test because of the relatively low storage temperatures following reactor discharge. B02 and other sibling assemblies were characterized in pretest examinations and did not show evidence of leaking fuel rods.(2,3) Assembly B02 was judged to be a good candidate for air storage because the pretest characterizations would provide data for evaluation of changes that might occur during dry storage testing. Assembly B02 was stored in a concrete silo for 3.5 years and in a dry storage vault for ~0.5 year.(4) Gas samples taken at the termination of B02 storage in the concrete silo and during the period of vault storage verified that no measurable fission gases were leaking from the fuel rods.

This air storage test was performed at the EMAD test facility at NTS. The hot cell capabilities at this facility include the fuel temperature test (FTT) stand, fuel handling, and an atmosphere control system (ACS).

EXPERIMENTAL DESCRIPTION

Assembly B02 is one of 17 Turkey Point 15 x 15 PWR spent fuel assemblies received by EMAD for dry storage testing. Assembly B02 resided in the core of Turkey Point Unit 3 during the first two cycles of operation. It was discharged November 25, 1975, after 827 full-power days of operation. The average core burnup was 25,665 MWd/MTU. Comprehensive nondestructive testing performed on Assembly B02 at Battelle Columbus Laboratories (BCL) prior to its shipment to EMAD was described by Davis.[2] Five rods (G7, G9, J8, I9, and H6) were removed from B02 for detailed nondestructive examination and then replaced. Qualitative results of nondestructive examinations performed on Assembly B02 and the five rods at BCL are described in Table I. A destructive examination of Rods G7, G9, J8, I9, and H6 from a sibling assembly (B17) yielded values of 2.28 to 2.77 MPa (331 to 401 psia) for the internal rod pressure at ambient temperature;[5] void volumes ranged from 21.70 to 24.26 cm^3. Assembly B02 was characterized at EMAD by visual and photographic examination to document that no detectable damage resulted during shipping and handling operations following the examination at BCL.

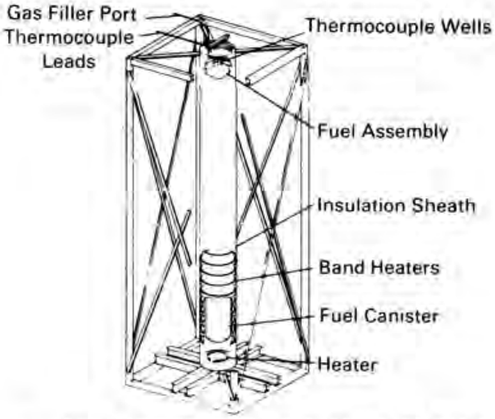

Fig. 1. Fuel Temperature Test Stand at NTS-EMAD.

The EMAD FTT stand used for these tests is shown in Fig. 1. It consists of a stainless steel canister surrounded by external heaters that are separated into 20 individually controlled zones. Thermocouples are located next to the heaters, on the exterior of the canister, and inside the fuel assembly guide tubes and are monitored continuously. The operation criteria are maintained by monitoring seven thermocouples in the center instrumentation tube of the fuel assembly.

The test assembly is protected against overheating by an adjustable automatic alarm/shutdown feature that shuts down the entire test and sounds an alarm if the preselected temperature is exceeded. The alarm/shutdown temperature selected for the test was 288°C. In addition, each of the 20 controlled zones is equipped with an adjustable overtemperature trip to shut off the power to the controller if temperatures

TABLE I

Nondestructive Examinations Performed by BCL on Assembly B02(2)

Examination	Results
Sipping	No evidence of breached rods.
Visual	Gray loose powder coating at top; black spalling crud layers in middle; dark adherent crud layer at bottom. All peripheral rods appeared in contact with the bottom tie plate. No damage or gross distortion were found in the structural components.
Neutron dosimetry	79 n/(mm^2-s), 1.3 MeV average energy.
Gamma dosimetry	58,800 R/h.
Assembly length	3.8947 m (153.338 in.).
Assembly bow	0 to 1.65 mm (0 to 0.065 in.).
Flat-to-flat distance at midspacer grid	210.4 to 210.9 mm (8.285 to 8.302 in.).
Weight	665 kg (1465 lb).
Eddy current	Indication of incipient cladding defects.
Rod profilometry	Ridging up to 0.05 mm (0.002 in.) on cladding; no blisters or bulges.
Rod ovality	~0.05 mm (0.002 in.) nominal; 0.17 mm (0.007 in.) maximum.
Rod diameter	10.6 mm (0.419 in.) average; 10.7 mm (0.423 in.) maximum.
Rod gamma scan	Correlated with ridging data from cladding profilometry.

exceed the limits. The temperature limit selected for heater cutoff on all heater controllers was 316°C.

The temperature history of Assembly B02 during dry storage at EMAD prior to and during the FTT is shown in Fig. 2. The peak initial temperature during the test was 275°C. The temperature was reduced monthly during the test to simulate the cooling of fuel placed in dry storage 5 years after reactor discharge. The sharp temperature reduction spikes shown in Fig. 2 were due to power outages and maintenance operations. The axial temperature distribution was imposed to simulate that expected for storage of spent fuel in a vertically oriented metal storage cask containing air.

Samples were taken of the gas atmosphere surrounding Assembly B02 while it was stored for ~30 months in a concrete silo, during its ~6 months of storage in the vault, and at least monthly during its 2 years in the FTT. Gas sampling in the FTT was accomplished using the ACS. Since tubing connects the FTT canister and the ACS, a 25-L accumulator was used to assure that representative samples of the canister atmosphere were collected. Two 500-mL samples were taken from the accumulator and analyzed for radionuclides (using gamma spectroscopy) and composition (using mass spectroscopy).

A gas sample was taken and analyzed prior to initiation of the test to provide baseline data for comparison with test data. Samples were taken approximately monthly during the test, usually closely coinciding with the changing of fuel assembly temperatures. Four sampling techniques were used during the test. During months 0 to 6, samples of the test atmosphere were taken without replacing the volume removed for samples. During months 7 to 15, the canister was vented to atmosphere after samples were taken to allow the system pressure to return to atmospheric. For months 15 to 22, the entire system atmosphere was evacuated through a 0.45-μm filter; and after samples were collected, the system was vented to atmosphere. Gas sampling of the FTT was then repeated immediately after backfilling the system, which permitted monitoring the krypton release rate from the fuel assembly. During month 23, the system was held at -200 torr gage for 6 days and sampled, held at -300 torr gage for 4 days and sampled, and held at -400 torr gage for 4 days and sampled. The system atmosphere was then evacuated and replaced with fresh air, and the 14-day sampling cycle was repeated. This method of sampling was used to characterize the nature and size of the fuel rod breach by observing the krypton-85 release rates at different system pressures. At month 24, the system atmosphere was exchanged; gas samples were collected before and after the exchange.

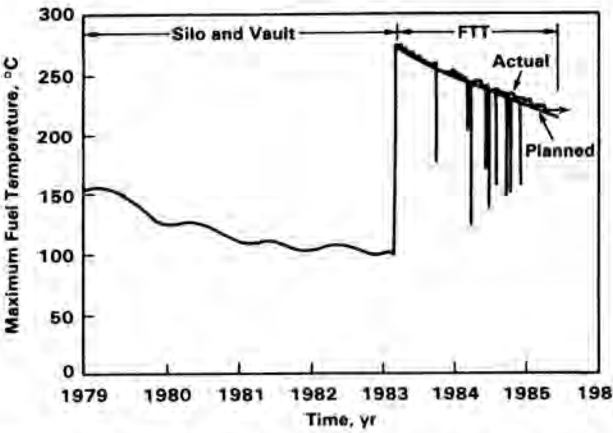

Fig. 2. Maximum Fuel Cladding Temperature History for Assembly B02 During Dry Storage at NTS-EMAD.

RESULTS

The results of the cover gas analyses are shown in Table II. Samples of the cover gas surrounding Assembly B02 were reported for standard temperature and pressure (STP) conditions. Corrections from actual to STP conditions were made by applying the ideal gas law and using the measured pressures and temperatures. Corrections were also made to account for the perturbations introduced by sample removal and dilution due to air replenishment after sampling. To make these corrections, it was necessary to compute the cumulative component concentrations. A correction was also made to the krypton-85 data to account for radioactive decay. The krypton-85 results shown in Fig. 3 indicate a large increase in krypton-85 concentration between months 2 and 3 (the second and third analyses of the gas atmosphere) coincident with a large increase in helium (Fig. 4). The interpretation of the data is discussed in the next section.

DISCUSSION

The corrected results are compared with the as-received chemical analyses in Figs. 4 through 8. The cask atmosphere initially had a composition similar to that of air (see Table II). The gas sample taken at month 2 of the test shows increases in CO_2 (Fig. 6) and H_2O (Table II) and a small decrease in O_2 (Fig. 7). One possible explanation is that carbonaceous material oxidized in the presence of O_2, elevated temperature, and a radiation field of about 10^4 R/h. It is postulated that a substantial fraction of this carbonaceous material originated from surfaces within the system.[6]

The gas analysis at month 2 showed detectable concentrations of N_2O. As shown in Fig. 8, the concentration of N_2O increased with time throughout the test until month 14. This gas is a known product of the gamma irradiation of air, and the production rate observed here is consistent with that observed in other studies.[7]

The krypton-85 concentration increased slightly prior to the third gas sample, when the krypton-85 level increased significantly (Fig. 3). This increase indicates that a cladding breach occurred. The total krypton-85 released to date is less than 1% of the estimated free krypton-85 available in one fuel rod (Fig. 3). The helium concentration also increased greatly in the third gas sample (Fig. 4), but the amount released did not exceed the estimated helium inventory from one fuel rod. From these observations, it was concluded that only one fuel rod had failed by the third month.

TABLE II

Results of Mass Spectrometric and Radiometric Analysis of Gas Samples from the Fuel Temperature Test

Sample Date	Month	Mass Spectrometric Analysis, vol%								Krypton-85 Radiometric Analysis, pCi/cm^3
		N_2	O_2	Ar	He	CO	CO_2	N_2O	H_2O	
3-01-83	1	78.4	20.6	0.93	ND[(a)]	tr[(a)]	0.06	ND	0.05	<0.05
4-08-83	2	78.9	18.2	0.94	ND	ND	1.96	0.03	0.23	0.52
5-12-83	3	79.0	17.5	0.94	0.27	ND	2.22	0.05	0.30	122,000
6-03-83	4	79.4	16.9	0.96	0.32	ND	2.31	0.08	0.32	147,000
7-18-83	5	79.6	16.7	0.94	0.32	ND	2.25	0.16	0.45	147,100
8-17-83	6	79.7	16.5	0.95	0.34	ND	2.33	0.19	0.36	157,000
9-20-83	7	79.9	16.1	0.95	0.34	ND	2.35	0.25	0.53	160,800
10-18-83	8	79.2	18.3	0.94	0.17	ND	1.14	0.17	0.07	72,800
11-11-83	9	79.1	18.6	0.94	0.15	ND	1.01	0.16	0.08	63,900
12-09-83	10	79.3	18.5	0.94	0.12	0.07	0.87	0.19	0.07	53,700
1-16-84	11	78.9	19.0	0.94	0.135	ND	0.80	0.19	0.08	50,400
2-17-84	12	79.0	18.98	0.94	0.12	0.068	0.70	0.181	0.07	43,700
3-22-84	13	79.1	19.2	0.92	0.08	0.04	0.52	0.173	0.06	30,500
4-26-84	14	79.2	19.0	0.921	0.076	ND	0.515	0.225	0.10	33,200
6-26-84	15	79.2	19.0	0.944	0.066	ND	0.481	0.243	0.02	27,600
6-27-84	15	78.5	20.4	0.926	ND	ND	0.038	ND	0.04	21.1
7-06-84	16	78.5	20.5	0.928	ND	ND	0.064	ND	0.03	125
7-06-84	16	78.5	20.5	0.926	ND	ND	0.052	ND	0.03	3.46
8-14-84	17	78.6	20.4	0.926	ND	ND	0.071	0.011	0.08	494
8-15-84	17	78.4	20.6	0.931	ND	ND	0.034	0.011	0.08	14.3
9-18-84	18	78.6	20.4	0.927	ND	ND	0.075	0.013	0.11	613
9-18-84	18	78.5	20.5	0.923	ND	tr	0.049	ND	0.10	4.67
10-22-84	19	78.4	20.6	0.933	ND	tr	0.069	ND	0.07	491
10-22-84	19	78.3	20.7	0.931	ND	tr	0.043	ND	0.04	3.15

(a) ND - not determined
tr - trace.

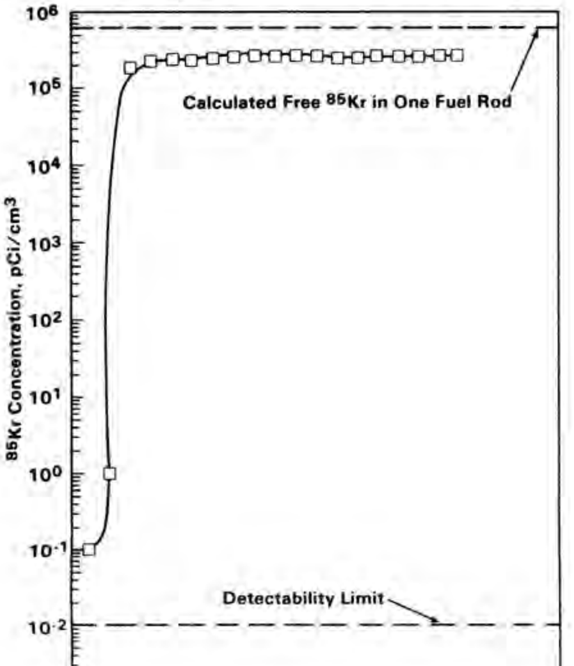

Fig. 3. Corrected Krypton-85 Concentration Increase in Canister Plenum Gas for Assembly B02.

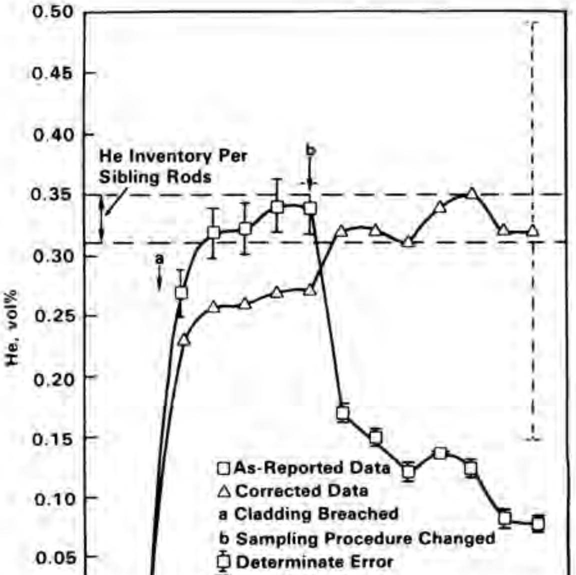

Fig. 4. He from Reported Chemical Analyses and Corrected for Sample Removal and Air Replenishment.

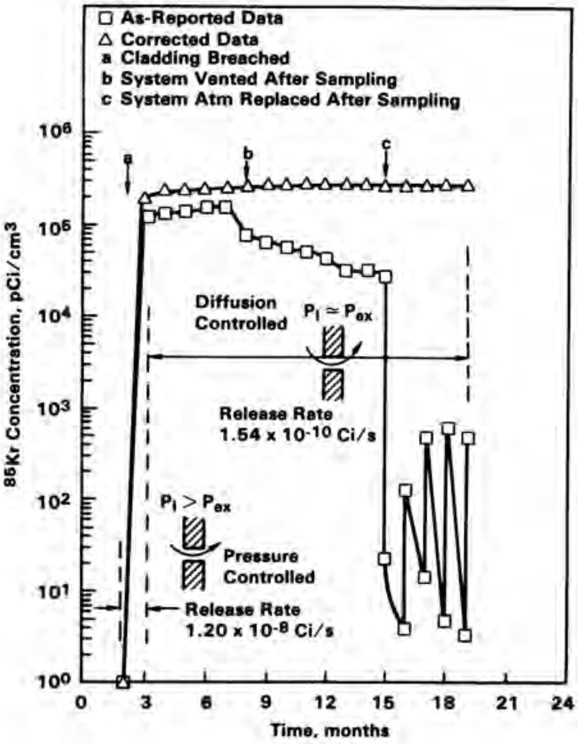

Fig. 5. Krypton-85 Concentration from Reported Chemical Analyses and Corrected for Pressure, Temperature, and Radioactive Decay.

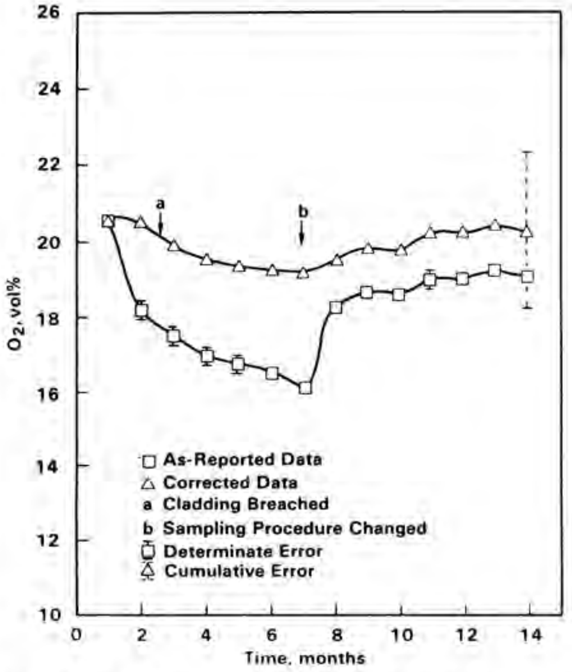

Fig. 6. O_2 from Reported Chemical Analyses and Corrected for Sample Removal and Air Replenishment.

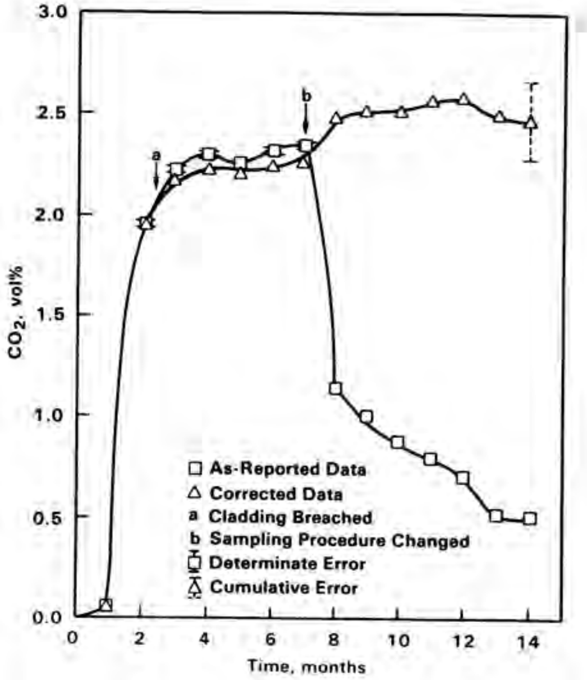

Fig. 7. CO_2 from Reported Chemical Analyses and Corrected for Sample Removal and Air Replenishment.

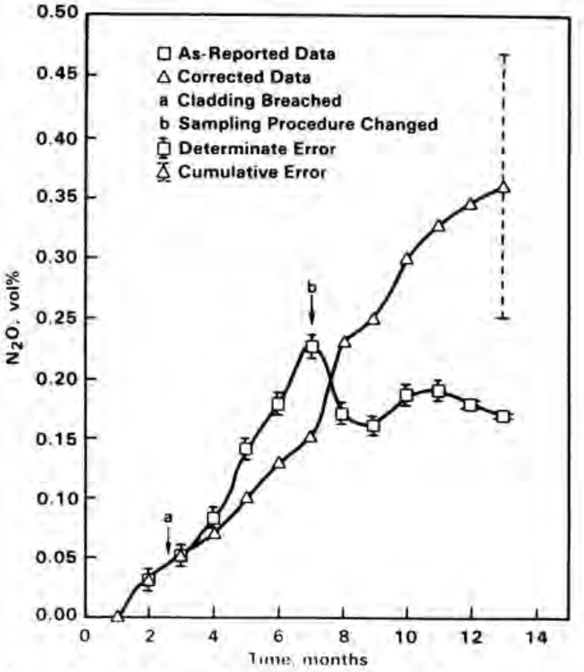

Fig. 8. N_2O from Reported Chemical Analyses and Corrected for Sample Removal and Air Replenishment.

The release rate of krypton-85 into the FTT atmosphere was computed from the results of the gas analyses. The release rate was analyzed hydrodynamically under assumptions of viscous flow dominated by gaseous molecular interactions to determine the flow rate of krypton-85. Using this flow rate, the estimated initial internal fuel rod pressure, and the external fuel rod pressure, the size of an ideal hole in the cladding was calculated. The Poiseuille equation(8) was used to obtain an approximate hole size using the following assumptions: 1) the gas is incompressible; 2) the flow is fully developed; 3) there is no turbulent motion of the gas; and 4) the flow velocity at the tube walls is zero. The Poiseuille equation is:

$$Q = \pi \, (a^4) \; P_a \, (P_2 - P_1)/8nL$$

where $Q = P \; \partial V/\partial t = kT \; \partial N/\partial t$

Q = volumetric flow rate (microns Hg cm^3/s)
a = hole radius (cm)
P_a = arithmetic mean of P_1 and P_2 (microns Hg)
P_1 = external fuel rod pressure (microns Hg)
P_2 = internal fuel rod pressure (microns Hg)
n = viscosity of the gas (poise)
L = cladding thickness (cm)
P = pressure for determination of Q (microns Hg)
∂V = partial differential of flowing gas (cm^3)
∂t = partial differential with respect to time (s)
k = Boltzmann constant (ergs/K)
T = absolute temperature (K)
∂N = partial differential of atoms of flowing gas (atoms).

Using the Q value obtained from krypton-85 release rates (1.2×10^{-8} Ci/s), a cladding breach diameter of ~0.3 μm was calculated. If the value of Q obtained from the helium release rate were used, a cladding breach diameter of ~1.3 μm would be calculated. Two possible kinetics models were examined in an attempt to explain this difference. The first model involved a pressurized release through an orifice penetrating the fuel cladding such that the hole size was greater than the krypton or helium molecular mean-free paths. For this case, illustrated in Fig. 9, the rate of helium and krypton-85 release must be the same. Since this is contradictory to observations, a second kinetics model was investigated.

In the second model, the smaller helium atom (atomic radius 0.49 Å) diffuses more rapidly than the larger krypton-85 atom (atomic radius 1.03 Å) through small intergranular cladding cracks that are probably generated by stress corrosion cracking (SCC) (Fig. 9). The observed difference in release rates between the krypton and helium atoms leads to the conclusion that the SCC cladding breach model is more appropriate. The differences in krypton-85 release rates between months 2 to 3 and the following months indicate that the initial krypton-85 release was pressure controlled (1.2×10^{-8} Ci/s) followed by a lower release rate (1.5×10^{-10} Ci/s) that may be diffusion controlled, as illustrated in Fig. 5.

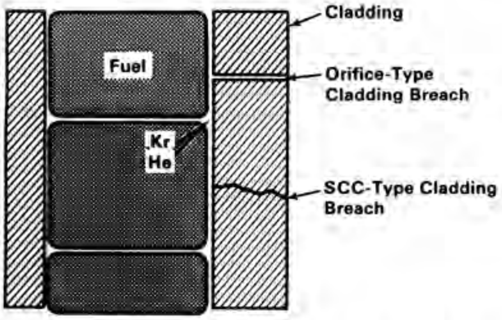

Fig. 9. Orifice- and SCC-Type Cladding Breaches.

The oxygen data shown in Fig. 6 indicate depletion of oxygen early in the test, which could result from reaction between oxygen and UO_2 at the site of the cladding breach. However, the depletion of oxygen shown in Fig. 6 corresponds to the increase in CO_2 shown in Fig. 7. There is potential for undetectable fuel oxidation to the extent of the uncertainty in the oxygen analyses. A computation of the amount of fuel that could be oxidized by the amount of oxygen represented by the uncertainty bounds in Fig. 6 leads to the conclusion that several centimeters of fuel could be converted to U_3O_8 without detectable oxygen depletion.

Over 6000 spent fuel rods have been monitored for fission gas release during dry storage tests and demonstrations.(9) This test with Assembly B02 represents the first reported case of a spent fuel rod breach during dry storage. Calculations from creep rupture properties suggest that the in-storage breach of a spent fuel rod in Assembly B02 required an incipient crack extending at least 90% through the cladding wall. Pretest eddy current inspection of five spent fuel rods in Assembly B02 indicated that the cladding contained incipient defects. The cause of the cladding breach will be investigated in the posttest examination.

CONCLUSIONS

- Breaching of a spent fuel rod in Assembly B02 during dry storage testing represents the first such case in over 6000 spent fuel rods that have been monitored during dry storage tests and demonstrations.
- Breaching of spent fuel cladding during dry storage can be determined through analyses of the storage atmosphere.
- Determination of the extent of fuel oxidation from analysis of oxygen depletion in the storage atmosphere has limited sensitivity because of the relatively small quantities of oxygen required to oxidize significant amounts of spent fuel.
- Diffusion-controlled krypton-85 release rates through the cladding breach are two orders of magnitude less than initial pressure-controlled krypton-85 release rates.

- Differences in the release rates of krypton and helium from the breached fuel rod suggest that the breach geometry consists of cracks similar to those produced by SCC. The breach is postulated to have occurred at the site of an incipient defect that was present prior to the dry storage test.

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