OBSERVATIONS ON DAMAGE ACCUMULATION DURING IODINE-INDUCED STRESS CORROSION CRACKING OF ZIRCALOY CLADDING

ROBIN L. JONES Electric Power Research Institute 3412 Hillview Avenue, Palo Alto, California 94303

EDWIN SMITH Manchester University, Department of Metallurgy Manchester, United Kingdom, M17HS

ALAN K. MILLER Stanford University Department of Materials Science and Engineering Stanford, California 94305

Received December 18, 1978 Accepted for Publication January 2, 1979

A predictive model has recently been proposed for pellet-cladding interaction (PCI) failures of water reactor fuel rods.¹⁻³ The model is based on the assumption that PCI failures are attributable to stress corrosion cracking (SCC) of the Zircaloy cladding induced by fission product iodine. Because the kinetics of iodine-induced SCC of irradiated Zircaloys are not well-documented at present, the PCI model¹⁻³ is based largely on failure-time data from constant stress tube pressurization experiments.⁴⁻⁶ The following procedure was used to apply these constant stress laboratory data to the variable-stress situation that exists during PCI failure. The computed hoop stress history of the cladding was divided into discrete time increments of length Δt . For each time step, a damage increment ΔD was calculated from

$$\Delta D = \frac{\Delta t}{t_f} \quad , \tag{1}$$

where t_f was defined as the failure time in a constant stress test on irradiated Zircaloy cladding at the average hoop stress and iodine availability prevailing during the time increment. PCI failure was predicted when

$$\Sigma \Delta D = 1 \quad , \tag{2}$$

where $\Sigma \Delta D$ was obtained by simple addition of the damage increments for successive time steps. In this Letter, we report the results of tests on unirradiated tubing that were designed to provide a preliminary assessment of the validity of this linear treatment of damage accumulation during iodine SCC of Zircaloys.

Specimens of reactor-grade unirradiated Zircaloy-4 tubing were pressurized at 633 ± 5 K with a mixture of helium and iodine. The metallurgical characteristics of this stress-relieved tubing and the details of the experimental procedure are described elsewhere.⁴ The test conditions and results are summarized in Tables I and II. An initial group of constant pressure tests was used to deter-



mine average values of t_f at three hoop stresses. As seen in Table I, the behavior in these tests was quite reproducible, with at most a $\pm 20\%$ scatter in failure times. In the subsequent pressure-change tests (Table II), each specimen was first held at one of the three stresses (σ_1) for a time Δt_1 , equal to about half the average failure time at that stress in the constant-pressure tests. Then the hoop stress was suddenly increased or decreased to another of the stress levels used in the constant pressure tests and held at this second stress (σ_2) until specimen failure occurred (Δt_2). Table II lists the values of ΔD_1 (at σ_1), ΔD_2 (at σ_2), $\Sigma \Delta D$ at failure (= $\Delta D_1 + \Delta D_2$) calculated using Eqs. (1) and (2), and σ_2/σ_1 .

The average values of $\Sigma \Delta D$ at failure are plotted against σ_2/σ_1 in Fig. 1. The dashed line at $\Sigma \Delta D = 1$ illustrates the relationship expected for the linear damage accumulation case. Although the individual values of $\Sigma \Delta D$ in Table II

TABLE I

Results of	Constant-Pressure	Tests*
------------	-------------------	--------

Specimen Number	Hoop Stress (MPa)	Failure Time (ks)	Average Failure Time (ks)
Zr-4-29	445	1.08	1.12
Zr-4-30	445	1.15	
Zr-4-31	396	3.89	3.80
Zr-4-32	396	3.71	
Zr-4-33	348	10.6	12.6
Zr-4-34	348	14.9	
Zr-4-35	348	12.4	

*Tests on stress-relieved Zircaloy-4 tubing at 633 ± 5 K. Iodine availability ~6 mg/cm² Zircaloy surface.

Specimen Number	σ ₁ (MPa)	Δt_1 (ks)	ΔD_1	σ ₂ (MPa)	Δt_2 (ks)	ΔD_2	$\Sigma \Delta D$	Average $\Sigma \Delta D$	σ_2/σ_1
Zr-4-36 Zr-4-37 Zr-4-38	445 445 445	0.61 0.61 0.61	0.55 0.55 0.55	348 348 348	6.05 17.5 9.29	0.48 1.39 0.74	1.03 1.94 1.29	1.42	0.78
Zr-4-39 Zr-4-40 Zr-4-41	348 348 348	6.01 6.01 6.01	0.47 0.47 0.47	445 445 445	0.83 0.43 0.94	0.74 0.38 0.84	1.21 0.85 1.31	1.12	1.28
Zr-4-42 Zr-4-43	445 445	0.61 0.61	0.55 0.55	396 396	2.16 2.05	0.57 0.54	1.12 1.09	1.11	0.89
Zr-4-44 Zr-4-45	348 348	6.01 6.01	0.47 0.47	396 396	1.37 0.65	0.36 0.17	0.83 0.64	0.74	1.14

TABLE II Results of Pressure-Change Tests*

*Tests on stress-relieved Zircaloy-4 tubing at 633 ± 5 K. Iodine availability ~6 mg/cm² Zircaloy surface.



Fig. 1. Damage accumulation behavior of stress-relieved Zircaloy-4 tubing in pressure-change iodine SCC tests at 633 ± 5 K.

show considerable scatter, it seems clear from Fig. 1 that the average values do not support the use of a linear damage accumulation rule for iodine-induced SCC of unirradiated Zircaloy-4 tubing.

The nonlinear behavior evident in Fig. 1 is not entirely unexpected. One implication of a linear cumulative damage rule is that each increment of damage depends only on the currently imposed external conditions and not on the current internal state of damage. In general, it is more likely that the current state of damage will affect the rate at which further damage is built up, resulting in nonlinear damage accumulation. Moreover, the qualitative form of Fig. 1 can be rationalized as follows. We observed that the stress corrosion crack depth leading to plastic instability failure of the remainder of the tube wall between the crack tip and the outside tube surface increased significantly with decreasing hoop stress: At a stress of 445 MPa, the crack reached a depth of ~100 μ m before instability occurred, whereas at 348 MPa, the crack depth at instability was ~250 μ m. As illustrated schematically in Fig. 2, this tends to lead to failure at values of $\Sigma \Delta D < 1$ in tests of the present type if $\sigma_2/\sigma_1 > 1$ but at values of $\Sigma \Delta D > 1$ if $\sigma_2/\sigma_1 < 1$. The behavior depicted in Fig. 2 was chosen arbitrarily, but the same prediction follows from most physically sensible crack formation and growth assumptions.

As seen in Fig. 1, the measured values of $\Sigma \Delta D$ at failure obey the expected trend for values of σ_2/σ_1 between 0.78 and 1.14. However, the value of $\Sigma \Delta D$ obtained at $\sigma_2/\sigma_1 =$ 1.28 is not in accordance with the expected behavior. The arrow labeled "Onset of Yielding" in Fig. 1 shows the maximum stress increase that could be imposed before σ_2 exceeded the short-time yield stress of the tubing. The data point at $\sigma_2/\sigma_1 =$ 1.28 is the only one in the present series obtained from tests in which σ_2 exceeded the yield stress. Therefore, the observed deviation from predicted behavior probably occurred because the stress corrosion cracks formed in these specimens at the lower stress σ_1 were partially blunted by plastic deformation during the stress increase. This would result in somewhat larger-than-expected values of $\Sigma \Delta D$ at failure, as was observed. (Complete blunting would require stress corrosion crack reinitiation at σ_2 , leading to values of $\Sigma \Delta D$ at failure approaching 1.5.)

The present results indicate that a linear damage accumulation rule does not provide accurate failure predictions for unirradiated Zircaloy-4 tube specimens undergoing iodine SCC under variable stress conditions. The data suggest that the use of a linear damage accumulation rule in our PCI failure model¹⁻³ may result in underestimates of the failure probability under rising stress and overestimates under falling stress. Tests are currently in progress to evaluate the damage accumulation behavior of irradiated Zircaloy cladding during iodine SCC. Depending on the magnitude of the departures from linear damage accumulation behavior observed in those tests, it may be necessary to develop a nonlinear treatment for incorporation in future versions of the PCI failure model.

REFERENCES

1. J. T. A. ROBERTS, R. L. JONES, D. CUBBICIOTTI, A. K. MILLER, H. F. WACHOB, E. SMITH, and F. L. YAGGEE, "An SCC Model for Pellet-Cladding Interaction Failures in LWR Fuel Rods,"



Fig. 2. Schematic illustration of the damage accumulation behavior expected in pressure-change tests when the stress corrosion crack depth at instability increases with decreasing stress. (a) Pressure decrease: $\sigma_2/\sigma_1 < 1$; $\Delta D_1 = 0.5$; $\Delta D_2 > 0.5$; $\Sigma \Delta D$ at failure >1; (b) $\sigma_2/\sigma_1 > 1$; $\Delta D_1 = 0.5$; $\Delta D_2 < 0.5$; $\Sigma \Delta D$ at failure <1.

Proc. 4th Int. Conf. Zirconium in the Nuclear Industry, Stratfordupon-Avon, England, June 26-29, 1978.

2. A. K. MILLER, K. D. CHALLENGER, E. SMITH, G. V. RANJAN, and R. C. CIPOLLA, "Zircaloy Cladding Deformation and Fracture Analysis," EPRI NP-856, Electric Power Research Institute (Aug. 1978).

3. A. K. MILLER, R. L. JONES, E. SMITH, K. D. CHALLENGER, and J. T. A. ROBERTS, "Modeling Cladding Deformation and Fracture in the SPEAR Code System," *Trans. Am. Nucl. Soc.*, **30**, 171 (1978).

4. R. L. JONES and D. CUBICCIOTTI, "Stress Corrosion Cracking of Zircaloys," EPRI NP-717, Electric Power Research Institute (Mar. 1978).

5. F. L. YAGGEE, R. F. MATTAS, and L. A. NEIMARK, "Iodine SCC in Irradiated LWR Cladding," *Trans. Am. Nucl. Soc.*, **30**, 199 (1978).

6. F. GARZAROLLI, R. MANZEL, M. PEEHS, and H. STEHLE, "KWR-Observations and Hypothesis of PCI Failures," *Proc. IAEA Specialists' Mtg. Pellet-Cladding Interaction for Water Reactors*, International Atomic Energy Agency, Vienna (1977).