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ZIRCALOY

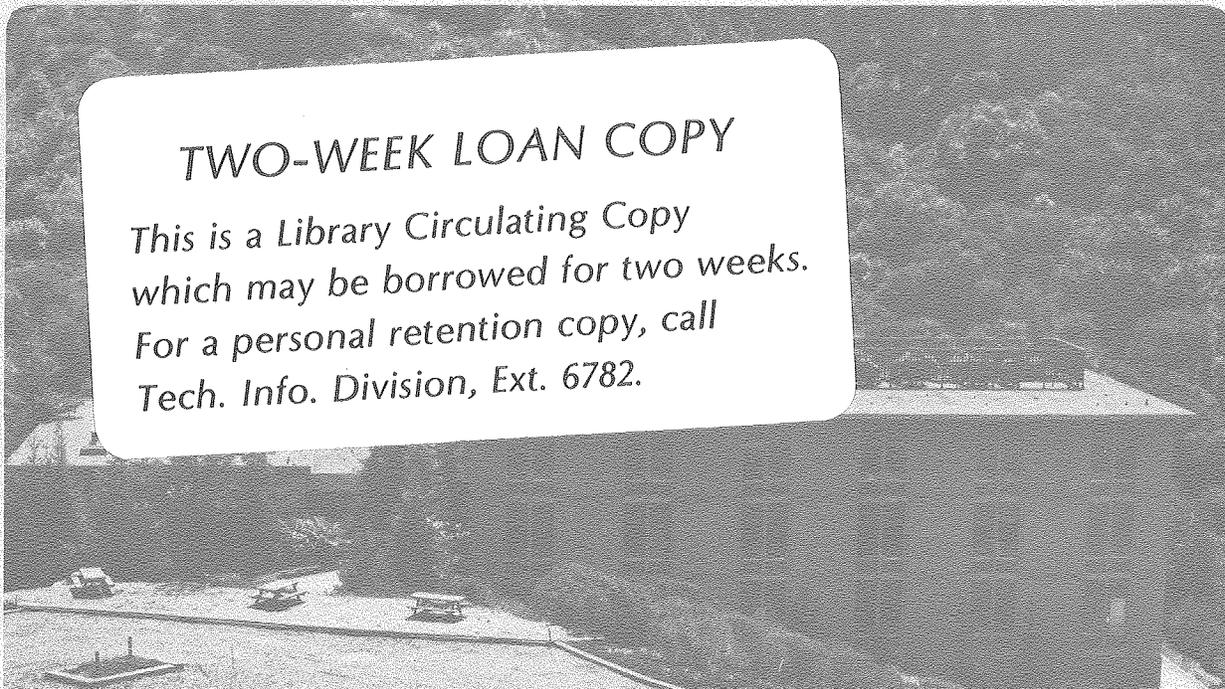
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CORRELATION OF FAILURE TIMES FOR IODINE SCC OF ZIRCALOY

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ABSTRACT

A crack-growth model of stress corrosion cracking has been successfully applied to predict times-to-failure of zircaloy specimens exposed to iodine vapor. Data for two types of tests were analyzed using the model. The first was a variable loading experiment in which failure occurred after the specimen had been subjected to two distinct stresses in succession. The second was a series of tests in which surface roughness, and probably residual stress as well, was reduced by chemical polishing of the specimens. The success of the crack growth model in dealing with these situations suggests that crack propagation rather than crack initiation is the rate-controlling step in iodine stress corrosion cracking of zircaloy. Furthermore, the metal in the vicinity of the growing crack is apparently so embrittled by iodine that a model originally intended for ceramics applies.

Recently, Jones et al.(1) reported that the linear damage accumulation rule fails to predict the rupture lifetime of unirradiated Zircaloy-4 tubes subject to iodine stress corrosion cracking (SCC) under variable stress conditions. In a related study, Syrett et al.(2) tested the iodine SCC susceptibilities of two lots of Zircaloy-2 tubing. The two lots were manufactured to the same nominal specifications (dimensions, chemical composition, mechanical properties, hydride distribution and steam corrosion) but some properties (surface roughness, texture, and residual stress) were different. In this note, we show that a crack propagation model (3) of iodine SCC can satisfactorily rationalize the results of the studies.

In this model (3) the crack growth rate da/dt in a solid embrittled by SCC is given in chemical reaction rate terminology

$$\frac{da}{dt} = A_0 p^n \exp(-E^*/RT) \exp(B_0 K/T) \quad (1)$$

where A_0 and B_0 are constants, T is the temperature and p is the partial pressure of the chemically active species responsible for SCC (I_2 in the present case). E^* and n are the activation energy and the order, respectively, of the chemical reaction. The crack length is a and R is the gas constant. The stress intensity factor K is:

$$K = Y\sigma\sqrt{a} \quad (2)$$

where Y is a geometrical factor of order unity and σ is the applied hoop stress in the internally pressurized tube. The last exponential term in Eq.(1) arises from the stress effect on the free energy of activation of the chemical step.

Under conditions of constant stress, temperature and iodine exposure, Eqs(1) and (2) can be combined and integrated to give the time-to-failure t_F :

$$t_F = \frac{1}{A} \int_{a_0}^{a_c} e^{-B_0 Y \sigma \sqrt{a}} da = \frac{2 \left[e^{-B_0 Y \sigma \sqrt{a_0} (1+B_0 Y \sigma \sqrt{a_0})} - e^{-B_0 Y \sigma \sqrt{a_c} (1+B_0 Y \sigma \sqrt{a_c})} \right]}{AB^2 \sigma^2} \quad (3)$$

where $A = A_0 p^n \exp(-E^*/RT)$ and $B = B_0 Y/T$. In Eq (3), a_0 is the depth of the pre-existing cracks in the specimen (either fabricated or present due to normal manufacturing operations) and a_c is the crack length at which the net section stress equals the ultimate tensile stress as determined from:

$$\frac{\sigma w}{w - a_c} = \sigma_{UTS} \quad (4)$$

where w is the thickness of the tube wall. The model assumes that stress corrosion causes propagation of the crack from length a_0 to length a_c at which point ductile failure occurs.

Variable Stresses

The crack propagation model has been successfully applied to iodine SCC under constant loading conditions(4) and should be equally applicable to the constant-loading data reported in Ref. 1. Selecting $a_0 = 5 \mu\text{m}$ and $\sigma_{UTS} = 531 \text{ MPa}$ (5), the constant-pressure test results of Ref.1 were fitted to Eq(3). The best fit was obtained for the parameters $A = 5 \times 10^{-5} \mu\text{m}/\text{ks}$ and $B_0 Y = 6.2 \text{ K/MPa}\sqrt{\mu\text{m}}$. The latter result compares favorably with the value $B_0 Y = 6.0 \text{ K/MPa}\sqrt{\mu\text{m}}$ obtained in the experiments reported in Ref. 4. The values of the coefficients A cannot be compared because the iodine loading reported by Jones et al(1) used units of concentration (mg/cm^2) different from and not convertible to the partial pressure units used in Ref. 4. Table 1 shows that the constant pressure data reported by Jones et al(1) can be quite well reproduced with these A and B values and Eq(3).

In the pressure-change tests of Ref. 1, the specimen was loaded a stress σ_1 for a time Δt_1 and then at a stress σ_2 for an additional time Δt_2 at the end of which failure occurred. Application of the crack propagation model to this two-stage loading gives the crack length at the end of the first stage (a_1) as the solution of the equation:

$$\Delta t_1 = \frac{2 \left[e^{-B\sigma_1 \sqrt{a_0} (1+B\sigma_1 \sqrt{a_0})} - e^{-B\sigma_1 \sqrt{a_1} (1+B\sigma_1 \sqrt{a_1})} \right]}{AB^2 \sigma_1^2} \quad (5)$$

and the second time interval to failure by:

$$\Delta t_2 = \frac{2 \left[e^{-B\sigma_2 \sqrt{a_1} (1+B\sigma_2 \sqrt{a_1})} - e^{-B\sigma_2 \sqrt{a_c} (1+B\sigma_2 \sqrt{a_c})} \right]}{AB^2 \sigma_1^2} \quad (6)$$

where a_1 is obtained from Eq(5) and a_c is given by Eq(4) with $\sigma = \sigma_2$. The values of A and B determined from the constant stress results were used in predicting Δt_2 so no disposable constants remain. Table 2 compares the measured times-to-failure in the second stage with those calculated from Eqs(5) and (6) for four two-stage pressure-change tests. The multiple experimental values in column 4 refer to repeated experiments conducted under the same nominal conditions. The large variation in some test results is attributed to the variability of the applied stresses, for which $\pm 5\%$ uncertainties were reported(1). In the first test shown in Table 2, if both σ_1 and σ_2 are decreased by 5%, Δt_2 is calculated to be 14.8 ks. Similarly, for the last test, an increase of both stresses by the same percentage decreases the calculated values of Δt_2 to 0.5 ks. Thus, the calculations appear to predict the experimental results to within the variability of the latter.

Table 1 Constant-Pressure Tests (Ref. 1)*

σ , MPa	t_F , ks	
	experimental	Model
445	1.12 \pm 0.03	1.12
396	3.80 \pm 0.09	3.80
348	12.6 \pm 2.3	12.6

*Stress-relieved Zircaloy -4 tubing with 0.64 mm wall thickness at 633 ± 5 K.
Iodine availability ~ 6 mg/cm²

Table 2 Pressure-Change Tests (Ref. 1)

σ_1 , MPa	Δt_1 , ks	σ_2 , MPa	Δt_2 , ks	
			experimental	predicted
445	0.61	348	6.1, 17.5, 9.2	6.9
348	6.01	445	0.8, 0.4, 0.9	0.5
445	0.61	396	2.2, 2.1	1.9
348	6.01	396	1.4, 0.7	1.8

Surface Roughness

The two lots of Zircaloy-2 used in the tests reported in Ref. 2 were both manufactured under ASTM B 353 standards. The material designated as lot no. 1 (lot A in Ref. 2) had a surface roughness of 4 μm (peak-to-peak) and a residual stress of ~ 100 MPa, while the corresponding figures for lot no. 2 (lot B in Ref. 2) were 9 and ~ 6 MPa, respectively. We have fitted these data to Eqs(3) and (4) in order to determine the iodine SCC constants A and B, which should be the same for the two lots. However, following the arguments in Refs. 2 and 5, test results from lot no. 1 at applied stresses less than 400 MPa were not included because the high residual stresses in this material render estimation of the true stresses uncertain. With these omissions, texture and surface roughness are the only remaining differences in the two lots. The latter parameter appears in the model as the quantity a_0 in Eq.(4). The best fit line shown in Fig. 1 corresponds to $A = 4 \times 10^{-5} \mu\text{m}/\text{ks}$ and $B_0 Y = 5.1 \text{ k/MPa} \cdot \sqrt{\mu\text{m}}$. The spread due to the 5% variability of the applied stress (6) is indicated by dashed lines. The fit is seen to be satisfactory, and the observed effect of surface roughness is adequately modeled.

Syrett, Cubicciotti and Jones(2,6) also polished a specimen from each lot to reduce the surface roughness to 3 μm (the wall thickness was reduced at the same time). After polishing, they tested the specimens under the same iodine availability and temperature conditions as were used for the as-fabricated specimens and at an applied stress of 346 MPa. Failure times for the treated specimens were computed from Eqs. (3) and (4) using the new values of a_0 and w but with the same iodine SCC parameters A and B that were determined for the as-fabricated tubes. The results are shown in Table 3. The specimen from lot no. 2 showed a dramatic effect of polishing, which reduced a_0 from 9 μm to 3 μm . The factor of ~ 20 increase in t_F is predicted by the model. The stress used in the tests of the polished specimens was lower than that judged necessary to eliminate residual stress by plastic strain. However, the fair agreement between theory and experiments for the treated specimen from the lot no. 1 supports the suggestion of Cubicciotti et al. (6) that removal of the surface layers of metal during the polishing operation may also have reduced the residual stress.

Summary

Despite the successes of the model, the basis of its applicability to iodine SCC is still uncertain. First, the model contains no provision for a threshold stress below which iodine does not affect the rupture lifetime or the fracture mode. Equation (1) is valid only for stresses in Eq(2) larger than the threshold value, which can only be determined empirically. A threshold

stress may be required to initiate cracks, but once done, their propagation is governed by the growth theory utilized here.

Second, the threshold stresses are a substantial fraction of the yield stress, so that departure from linear elasticity undermines the applicability of the stress intensity factor concept. However, the very low strains at failure which characterize iodine SCC suggest that the region of the metal in front of the crack is far less ductile than unaffected zones. The physical state of the solid in region where stress amplification occurs may be as brittle as a ceramic, which is the type of material for which the crack growth model was originally developed (3).

Third, the model does not explicitly account for variations in specimen texture, although this variable may influence the parameters A_0 and B_0 in Eq(1).

The crack propagation model, although originally intended for use with brittle ceramics, appears to be applicable to Zircaloy embrittled by iodine SCC. Although Eq.(1) has only tenuous fundamental underpinnings, its integrated form appears to provide a better correlation of iodine SCC with variable stresses than does the linear damage accumulation rule. The model also quantitatively accounts for the effect of surface roughness on the time-to-failure.

Table 3. Effect of Surface Polishing on Failure Times

Lot No.	Burst Strength MPa [†]	Surface Roughness μm^{\dagger}	Residual Stress MPa [†]	time-to-failure at 346 MPa, ks			
				as fabricated		after surface polishing	
				expt'l [†]	predicted	expt'l [†]	predicted
1	496	4	~ 100	(347) [*]	109	180	207
2	490	9	~ 6	10	8	220	205

[†] Reported in Ref. 2

* Result probably affected by high residual stress

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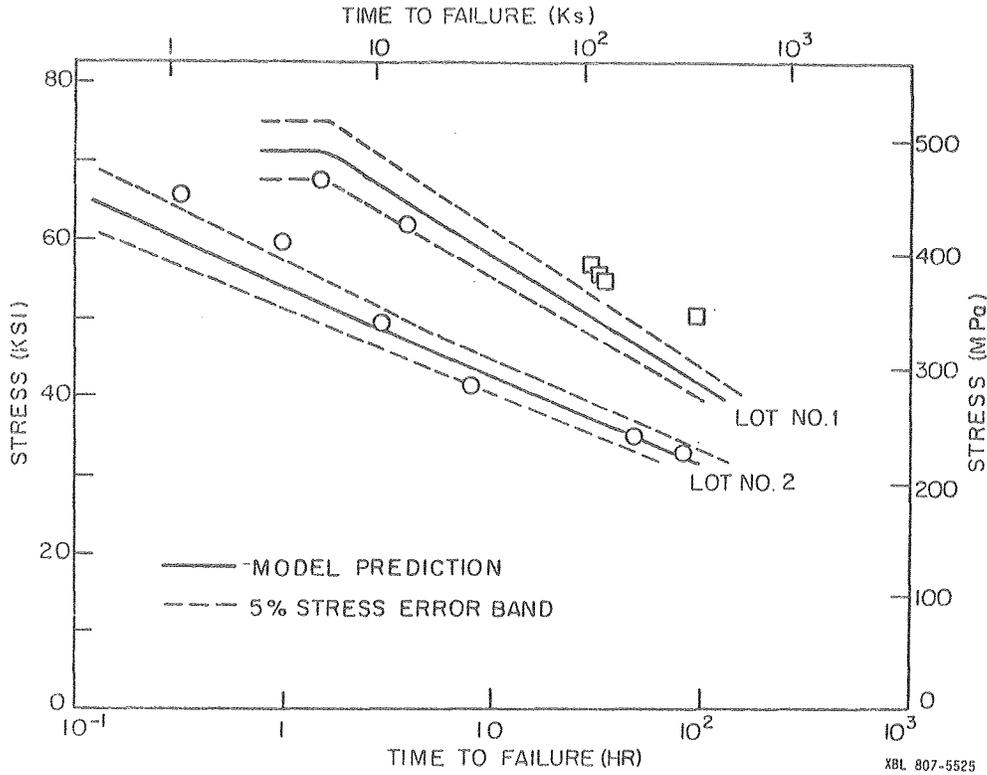


Figure 1

Surface roughness effect on time-to-failure of two lots of zircaloy-2 tubing (Ref.2); testing temperature = 590°K, iodine availability = 6.0 mg/cm²

- o = points with either low or relaxed residual stressed (used in curve fitting)
- = points not used in curve fitting because of high residual stress.

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