Fracture Mechanics Analysis of Iodine-Induced Crack Growth in Zircaloy-4 Tubing Between 500 and 700 °C

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Abstract

Low-ductility failure of Zircaloy tubing due to iodine-induced stress corrosion cracking (SCC) can occur up to about 700°C. The time-to-failure behavior of Zircaloy-4 cladding tubes containing iodine has been described by the elastic-plastic fracture mechanics model CEPFRAME for the temperature region 500 to 700°C. The model includes an empirically-determined computation method for the incubation period of crack formation, as a portion of the time-to-failure, as well as an elastic-plastic model for describing crack growth due to iodine-induced SCC. The total life time of the cladding tube is obtained by adding the crack initiation and crack propagation periods. The incubation period is a temperature-dependent function of both the depth of surface damage (both fabrication pits and machined notches) and the applied load, and is 40 to 90% of the time-to-failure.

The elastic-plastic crack growth model is a modified version of the stress intensity K_I -concept of linear-elastic fracture mechanics. The extensions of this concept take into account a plastic strain zone ahead of the crack tip, which effectively increases the crack depth, and in addition, a dynamic correction factor for the crack geometry which is essentially a function of the effective crack depth. Unstable crack growth is predicted to occur when the residual cross section reaches plastic instability.

Model results show good agreement with experimental data of tube burst tests at 500, 600 and 700°C. The crack growth velocity at all three temperatures is a power function of stress intensity ahead of the crack tip; the exponent is 4.9. The model can estimate time-to-failure of as-received cladding tubes containing iodine within a factor of 2. Application of the model to temperatures below 500°C is possible in principle. Due to the increasing scatter in experimental data, the structural transformation of the cladding by recrystallization, and the growing importance of creep strain, CEPFRAME has an upper temperature limit of approximately 650°C. The model is suitable for use in computer codes describing LWR fuel rod behavior during reactor transients and accidents.

1. Introduction

It is a generally established fact that zircaloy cladding can fail due to stress corrosion cracking (SCC) when the power is increased too quickly in light-water reactors (LWR). The prerequisites for SCC failure of fuel rods are the combined effects of tensile stress, caused by mechanical interaction between fuel and cladding, and chemical attack of the cladding inner surface, primarily by the volatile fission product iodine. These conditions may exist during very fast power increases under normal reactor operation conditions, but may also occur during reactor incidents as a result of mechanical interaction between fuel and cladding as in "Anticipated Transients without Scram" (ATWS) or "Reactivity Initiated Accidents" (RIA), or as a result of the difference between system and internal rod pressure as in a "Loss-of-Coolant Accident" (LOCA).

The results of out-of-pile creep rupture and burst tests performed between 500 and 900°C with Zircaloy-4 tube specimens containing small amounts of simulated fission products have clearly shown that only iodine or volatile iodine compounds (ZrI_4) can induce low-ductility failure due to SCC up to temperatures of about 800°C /1,2,3,4/. In some cases, the time-to-failure of the tubing is strongly reduced, especially in preflawed specimens. However, low-ductility cladding failure due to SCC only occurs after a well defined critical iodine concentration and a threshold hoop stress (both temperature-dependent) have been exceeded /3,12/.

Investigations of other authors have been limited to the study of iodine-induced SCC behavior of zircaloy tubing below 500°C. Various fracture mechanics models have been developed to describe the propagation of stress corrosion cracks under normal reactor operating conditions and to evaluate the critical values of stress, stress intensity, deformation, and iodine concentration, as functions of temperature, which can lead to cladding failure /5-11, 16-19/. This paper discusses the possibility of using fracture mechanics methods to describe the SCC behavior of zircaloy cladding above 500°C. The objective was to give a clear answer, if and up to which temperature a modified stress intensity concept can be applied to describe the iodine-induced stress-corrosion crack growth in the temperature range 500 to 700°C and to predict times-to-failure.

2. Construction of an Appropriate Fracture Mechanics Model

In the literature, the stress intensity (K_I) concept of linear-elastic fracture mechanics (LEFM) has been used to describe zircaloy cladding fracture due to iodine-induced SCC /5,6,7, 9,10, 16-19/. Efforts have been made to include a plastic deformation zone and the influence of cohesive forces between atoms in the region ahead of the crack tip in more realistic crack models /8,9,13/. Crack initiation has been discussed extensively /5,9,10,16,17/ but was not taken into account in the models.

A modified LEFM model was used as the basis of our own investigations in the temperature range 500 to 700°C. Since low-ductility ("brittle") failure of zircaloy due to iodine-induced SCC has been observed at all temperatures, especially for preflawed specimens, predominantly elastic material behavior can be assumed. Therefore, a plastic zone correction is taken into account ahead of the crack tip in the model CEPFRAME (Computation of elasticplastic fracture mechanics). Three phases of the SCC failure mechanism can be defined: (a) crack initiation or incubation, (b) crack propagation, and (c) spontaneous rupture of the residual wall thickness.

2.1. Crack Initiation

Crack initiation is a defect-creating process which is operative before stress-intensity-controlled crack growth begins. If the critical values of iodine concentration and hoop stress in a zircaloy tube specimen are exceeded, cracks will form at the inner cladding surface or at the notch tip. The cracks will then grow with increasing velocity until spontaneous ductile failure of the remaining wall cross section occurs. The total time-to-failure t_F is thus composed of the crack initiation period t_I plus the stable crack propagation time t_p ($t_F = t_I + t_p$). Final rupture of the cladding takes place so quickly that no corresponding time portion is assumed in the model. The crack initiation time t_I is a function of the depth and geometry of the notch, and of the applied load.

$$t_I = A(T) \cdot F(a_o/w, \sigma_n/\sigma_B, T) \quad (1)$$

where $A(T)$ is a temperature-dependent factor, a_o is the initial notch depth, w is the wall thickness, σ_n is the nominal hoop stress, and σ_B is the rupture stress. Since t_I could not be directly determined during the SCC experiments, a power law between t_I and the applied hoop stress combined with an exponential function between t_I and the initial notch depth a_o were assumed. Influences due to the deformation rate of the tubing and the iodine concentration were not considered. However, they are taken into account indirectly by temperature-dependent factors.

2.2. Crack Propagation

The crack depth (a) increases according to:

$$\frac{da}{dt} = C \cdot K_I^n, \quad (2)$$

where C and n are experimentally determined constants for a given temperature. The stress intensity factor K_I at the crack tip increases with increasing crack (or notch) depth according to the basic LEFM equation:

$$K_I = \sigma_n \sqrt{\pi a} Y(a) \quad (3)$$

The function $Y(a)$ describes the influence of the geometry of the crack front and the size of the crack with respect to the wall thickness and was calculated using the method of Newman and Raju /14/.

The length ω of the plastically deformed zone ahead of the crack tip (in the direction of crack growth) is expressed by /15/:

$$\omega = \frac{2}{\beta \sqrt{\pi}} \left(\frac{K_I}{\sigma_F} \right)^2. \quad (4)$$

If plane-stress in the thin-walled cladding tubes is assumed ahead of the crack front, then β equals 2. The flow stress σ_F is equal to $0.5 (\sigma_{0.2} + \sigma_B)$. An effective crack is then defined whose tip lies in the middle of the plastic zone:

$$a_{eff} = a + \frac{\omega}{2} = a + \frac{1}{\beta \sqrt{\pi}} \cdot \left(\frac{K_I}{\sigma_F} \right)^2. \quad (5)$$

This gives an effective stress intensity of:

$$K_{I,eff} = \sigma_n \sqrt{\pi a_{eff}} Y(a_{eff}). \quad (6)$$

The actual crack grows until it attains a critical depth a_c , which is the depth at which the stress in the remaining cross section is equal to the ultimate tensile stress. At this depth, the threshold load for plastic instability is reached in the remaining cross section and spontaneous ductile rupture occurs. The critical crack depth a_c can therefore be defined as the point at which crack growth becomes unstable (plastic breakoff criterion); a_c can not be determined by LEFM.

The relationship between the effective stress intensity and the crack propagation rate can be represented by equation (2) in the modified form:

$$\frac{da}{dt} = C(T) \cdot K_{I,eff}^n \quad (7)$$

Where C and n are constant for fixed environmental conditions (temperature and iodine concentration). Two values of the stress-intensity factor ($K_{I,eff}$) can be determined, one at the beginning ($K_{I,eff,o}$) and one at the end ($K_{I,eff,c}$) of the stable crack propagation period /20/.

The crack growth time t_p can be calculated by integrating equation (7) between the limits a_o and a_c .

$$t_p = \frac{1}{C \sigma_n^n \eta^{n/2}} \int_{a_o}^{a_c} \frac{da}{a_{eff}^{n/2} Y(a_{eff})^n} \quad (8)$$

One method to examine whether this model is suitable for describing Zircaloy cladding failure due to iodine-induced SCC is to determine the degree of agreement between the measured time-to-failure (t_M) and the calculated time-to-failure (t_F).

3. Experiment Conduct

The test program was conducted under isothermal test conditions as follows:

1. The stress intensity K_I was varied by varying the internal gas pressure of the tube specimens without changing the initial geometry a_o of the machined internal notch;
2. The average crack velocity da/dt was varied with respect to the effective stress intensity $K_{I,eff}$ by varying the initial notch depth without changing the time-to-failure (by applying different gas pressures in the tube specimens).

The objective of the test program was to determine the relationship between the SCC propagation rate da/dt and the stress intensity factor K_I .

Isothermal, isobaric creep rupture experiments were performed with unirradiated Zircaloy-4 tube specimens at 500, 600, and 700°C in helium. The initial iodine concentration was 10 mg/cm³ (well above the critical concentration with respect to SCC /3/) in all experiments. As-received and machine-notched tube specimens with axial notches of various initial depths ($a_o = 50$ to 200 μ m) and lengths ($2c = 100 \times a_o$) on the cladding inside surface were used. The specimens were about 100 mm long, with an outer diameter of 10.76 mm and a wall thickness of 0.72 mm. The time-to-failure varied between 5 and 3600 s for internal pressures between 50 and 500 bar. The burst apparatus and experiment conduct are described in reference /1/.

4. Test Results and Discussion

The failure behavior is determined primarily by two processes: (a) plastic creep strain in the whole cladding wall which takes place predominantly during the crack initiation phase (incubation period); and (b) crack propagation which is influenced by both the temperature-dependent transport velocity of gaseous iodine or volatile iodine compounds and the tendency of iodine to be adsorbed at the crack tip. The temperature-dependent combination of the two processes and additional local plastic strains in the vicinity of the crack front, particularly at 600°C, determine the strain and time-to-failure behavior of the specimens /20/.

4.1. Burst Behavior

Burst pressure is shown versus time-to-failure at 500, 600, and 700°C in figure 1. The time-to-failure of the tube specimens increases with decreasing pressure. The initial notch depths have nearly the same influence at 600°C for all applied burst pressures. This is different from 500 and 700°C. At 500°C, spontaneous ductile failure occurs at high pressure, and at 700°C, ductile failure after creep deformation occurs at low pressure. Figure 2 shows circumferential burst strain as a function of burst pressure at 600°C. The strong influence of iodine and of the initial notch depth on the burst strain are clearly apparent. The circumferential burst strain is in general less than 10% and for deep notches less than 1 %. The results at 500 and 700°C are similar. This low-ductility ("brittle") failure behavior of the zircaloy tubing due to iodine-induced SCC justifies the application of a modified LEFM model. Circumferential burst strain versus average strain rate is shown in figure 3. A burst strain minimum clearly exists for notched specimens at an average deformation rate of about 10^{-4} s^{-1} at 600°C (the average circumferential deformation rate is not directly related to the crack growth rate).

4.2. Fracture Surface Appearances

Scanning electron microscope (SEM) examination of the cladding inside surface and fracture surfaces show strongly temperature-dependent crack initiation behavior and fracture surface appearances /20/. In as-received (unnotched) tube specimens, both microscopic and macroscopic axially-oriented incipient cracks formed over the entire cladding inside surface. The number of incipient cracks increased mainly with temperature. Crack initiation was intergranular at all temperatures. In notched tube specimens at 500 and 600°C, incipient cracks formed only at the root of the notches. At 700°C, a few cracks also formed in the undamaged regions for initial notch depths smaller than 100 μm .

The type of fracture observed within the SCC zones was found to depend mainly on temperature and to be nearly independent of the applied stress. The fracture surface at 500°C showed transgranular fracture, at 700°C intergranular fracture, and at 600°C a mixture of both types of fracture in the as received specimens /3,20/.

5. Fracture Mechanics Calculations

Calculation of the crack initiation and propagation periods and a discussion of the results are presented in the following sections.

5.1. Crack Initiation

The crack incubation period t_I was calculated using equation (1). The correlation was developed empirically from plausibility considerations (stress dependence), from an analysis and comparison between measured and calculated times-to-failure (notch depth dependence), and from iterative calculations of the equation factors simultaneously with the determination of C and n of equation (8). The resulting correlation was supported by some crack initiation experiments performed at 600°C. As a fraction of the total lifetime, iodine-induced SCC initiation varied between 40 and 90%, depending on the initial notch depth.

5.2. Crack Propagation

After an iodine-induced crack has formed, the crack propagation obeys a power law on the stress intensity according to equation (7). The exponent n was assumed to be constant for all temperatures (500 to 700°C). The parameters σ_n , a_o , a_c , ω , $Y(a_{eff})$, and t_F were measured or directly calculated. C and n were determined iteratively for each temperature as follows:

1. A value was chosen for the exponent n (the same for all experiments) and the corresponding C_i calculated from equation (8) for each individual test i .
2. The value of n was optimized until scattering of the C_i values around a (logarithmic) mean value C_m reached a minimum.
3. Using the optimum pairs of values C_m and n corresponding values for (da/dt) were determined for $K_{I,eff,o}$ and $K_{I,eff,c}$ for each test (these values represent bounding conditions and serve as a check on the calculation). At a given temperature, $\log (da/dt)$ versus $\log (K_{I,eff})$ represents the equation

$$da/dt = C_m(T) \cdot K_{I,eff}^n \quad (8.1)$$

and thus constitutes an approximation to the desired crack growth relationship. Figure 4 shows the results at 600°C.

4. The measured and calculated times-to-failure (t_M and t_F) were then compared by calculating t_F for each test from C_m and n by equation (8) and the correlation

$$t_F = t_P (1 - t_I/t_F)^{-1} \quad (9)$$

and plotting these values versus t_M . Figure 5 shows the results at 600°C.

The growth rate for iodine-induced SCC can be described by an Arrhenius-Type rate equation

$$\left(\frac{da}{dt}\right)_{500-600^\circ\text{C}} = 1.17 \cdot 10^7 \exp\left(\frac{-2.39 \cdot 10^5}{R \cdot T}\right) K_I^{4.9} \quad (10)$$

where da/dt is in [m/s], $K_{I,eff}$ in [$\text{MPa} \cdot \text{m}^{1/2}$], and R is 8.3143 [J/mol·K].

The time-to-failure predictions for as-received tube specimens, with an assumed initial notch depth of 30 μm , show that the iodine-induced crack growth can be satisfactorily described with the model CEPFRAME (fig. 5, solid circles). The correlation factors are: 0.918 at 500°C, 0.827 at 600°C, and 0.818 at 700°C. The prediction improves with decreasing temperature.

The results of the present fracture mechanics analyses are shown with similar literature data at lower temperatures /7,10,16-19/ in figure 6. For a given stress intensity factor the crack growth rate increases with increasing temperature. The crack growth rates from 500 and 700°C agree well with those from 300 to 400°C, which underlines the reliability of the CEPFRAME results. The consideration of a plastic zone ahead of the crack tip is probably an important prerequisite for the successful analysis of crack propagation at higher temperatures. Consideration of a separate crack initiation phase also supports the calculation of growth rates of the appropriate order of magnitude. The influence of a continuously changing crack-geometry factor on the growth rate is less clear.

6. Summary and Conclusions

- Iodine in as-received Zircaloy-4 cladding tubes causes low-ductility ("brittle") failure due to SCC between 500 and 700°C. Circumferential burst strain is further reduced in preflawed (notched) specimens. The circumferential burst strain depends on temperature, burst pressure, and initial crack depth. At 500°C the burst strain varied between 0 and 2 % and at 600 and 700°C between 1 and 10 %; the deeper the initial notch depth the smaller the failure strain.
- Due to the iodine-induced low-ductility failure behavior of zircaloy tubing a modified version of the stress intensity factor K_I -concept of LEFM can be applied (CEPFRAME). SCC growth under static loading conditions obeys a power law of the form $da/dt = C(T) \cdot K_{I,eff}^n$. A plastic region in front of the crack tip, which effectively increases the crack depth ($a_{eff} = a + w/2$), and the continuous change in crack geometry are considered in CEPFRAME. The constants C and n were determined from the actual crack growth time, neglecting the crack initiation period. C(T) is an exponential function of temperature with an activation energy of $2.39 \cdot 10^5$ J/mol between 500 and 600°C, and the exponent n is 4.9 for all temperatures.
- The fracture surface appearance depends on temperature. At 500°C the cracks were primarily transgranular to a depth of 40 to 60% of the wall thickness before ductile rupture occurred. At 600°C the cracks were initially intergranular, followed by a transgranular region to a depth of 75 to 90% of the wall thickness before ductile rupture occurred. At 700°C the cracks were almost always completely intergranular with no ductile rupture.
- CEPFRAME time-to-failure predictions (crack initiation plus crack propagation periods) show fair agreement with experimental data. The model can estimate the lifetime of iodine-containing Zircaloy-4 cladding tubes within a factor of 2. Application of the model to temperatures below 500°C is possible; the upper temperature limit is about 650°C.

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Literature

- / 1 / HOFMANN, P., "Influence of iodine on the burst strain of Zircaloy-4 cladding tubes under simulated reactor accident conditions", J. Nucl. Mater. 87 (1979) 49-69.
- / 2 / HOFMANN, P., SPINO, J., "Chemical interactions between simulated fission products and Zircaloy-4 at elevated temperatures, including influence on ductility and time-to-failure", J. Nucl. Mater. 102 (1981) 117-134.
- / 3 / HOFMANN, P., SPINO, J., "Determination of the critical iodine concentration for SCC failure of Zircaloy-4 tubing between 500 and 900°C", J. Nucl. Mater. 107 (1982) 297-310.
- / 4 / HOFMANN, P., SPINO, J., "Chemical aspects of the iodine-induced SCC failure of Zircaloy-4 tubing above 500°C", J. Nucl. Mater. 114 (1983) 50-65.
- / 5 / SHANN, S.H., OLANDER, D.R., "Correlation of failure times for iodine SCC of Zircaloy", Nuclear Technology, 53 (1981) 407-409.
- / 6 / TUCKER, R.P., KREYNS, P.H., KEARNS, J.J., "The effect of internal surface flaws, iodine concentration and temperature on the SCC behavior of Zircaloy-4 tubing", WAPD-TM-1248 (1976).
- / 7 / KREYNS, P.H., SPAHR, G.L., MCCAULEY, J.E., "An analysis of iodine SCC of Zircaloy", J. Nucl. Mater. 61 (1976) 203-212.
- / 8 / SMITH, E., MILLER, A.K., "Stress corrosion fracture of Zircaloy cladding in fuel rods subjected to power increases. A model for crack propagation and the failure threshold stress", J. Nucl. Mater. 80 (1979) 291-302.
- / 9 / ROBERTS, J.T.A., et.al., "An SCC model for PCI failures in LWR fuel rods", Proceedings 4th Intl. Conf. on Zirconium in the Nuclear Industry, Stratford-upon-Avon, 1978, ASTM-STP 681 (1979) 285-305.
- / 10 / MILLER, A.K. OCKEN, H., TASOOJI, A., "Iodine SCC of Zircaloy: Laboratory data, a phenomenological model, and predictions of in-reactor behavior", J. Nucl. Mater. 99 (1981) 254-268.
- / 11 / SMITH, E., "Some implications of recent developments in plastic fracture mechanics on SCC in engineering materials", Mat. Sci. Eng. 44 (1980) 205-211.
- / 12 / HOFMANN, P., SPINO, J., "Can one expect low ductility failure of Zircaloy-4 tubing due to iodine-induced SCC in LOCA transients?" Proceedings ANS Topical Meeting on Reactor Safety Aspects of Fuel Behavior, Sun Valley, Idaho, USA, 1981, 2/410.
- / 13 / GARLICK, A., GRAVENOR, J.G., "A criterion and mechanism for power ramp defects", Proceedings ANS Topical Meeting on LWR Fuel Performance, ST. Charles, Illinois, USA, 1979, Vol. 2, 410-421, AED-Conf. 667-000.
- / 14 / NEWMAN, J.C., RAJU, I.S., "Stress analysis and fracture of surface cracks in cylindrical pressure vessels", Transactions of the 5th Internat. Conf. on SMiRT, Berlin, Germany (1979), Paper G7/3.
- / 15 / MUNZ, D., "Linear-elastische und elastisch-plastische Bruchmechanik", DFVLR-79-31 (1979).
- / 16 / VIDEM, K., LUNDE, L., "SCC initiation and growth in Zircaloy", Proceedings Enlarged HPG Meeting on LWR Fuel Performance, Sanderstolen, Norway, 1977, HPR 211

- /17/ VIDEM, K., et.al., "Cracking of cladding tubes caused by power ramping and by laboratory stress corrosion experiments", J. Nucl., Mater. 87 (1979) 259-267
- /18/ BUSBY, C.C., TUCKER, R.P., McCauly, J.E., "Halogen SCC of Zircaloy-4 tubing", WAPD-TM-1149 (1974)
- /19/ WOOD, J.C., "Factors affecting SCC of Zircaloy in iodine vapors", J. Nucl. Mater., 45 (1972/73) 105-122
- /20/ DIENST, W., ECKSTEIN, H., HOFMANN, P., "Fracture mechanics analysis of iodine-induced crack growth in Zircaloy-4 tubing between 500 and 700°C", submitted for publication in Nucl. Engineering and Design.

Fig. 1: Burst pressure versus time-to-failure of as received and preflawed Zircaloy-4 tubing at 500, 600 and 700°C (without and with iodine)

Fig. 2: Circumferential burst strain versus burst pressure of as-received and preflawed Zircaloy-4 tubing at 600°C (without and with iodine)

Fig. 3: Circumferential burst strain versus average strain rate of as-received and preflawed Zircaloy-4 tubing at 600°C (without and with iodine).

Fig. 4: Crack growth rate versus stress-intensity factor for Zircaloy-4 tubing failed at 600°C due to iodine-induced SCC.

Fig. 5: Calculated versus measured time-to-failure for Zircaloy-4 tubing failed at 600°C due to iodine-induced SCC.

Fig. 6: Crack growth rate versus stress-intensity factor for Zircaloy-4 tubing failed due to iodine-induced SCC; comparison with literature data (A/7/, B/19/, C/16/, D/17/, E/10/).