FRACTURE PREVENTION IN ENERGY AND TRANSPORT SYSTEMS

IODINE STRESS CORROSION CRACKING IN ZIRCALOY

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The subcritical growth of iodine-induced cracks in unirradiated Zircaloy plates is investigated as a function of the stress intensity factor K. The testing variables are: crystallographic texture (f-Number), microstructure (grain directionaly), heat treatment (stress relieved vs recrystallized plate), and temperature. The iodine partial pressure is 40Pa.

INTRODUCTION

The Zircaloys have been developed for improved corrosion resistance up to 623K and their typical use is in fuel cladding of LWR nuclear plants. Cladding tubes for PWRs is used in a cold-worked and stress-relieved condition (CWSR). Annealed Zircaloy generally is used for BWRs cladding and others parts of the reactor system.

The Pellet-Cladding Interaction (PCI) phenomenon which affects the Zircaloy cladding of LWR fuel rods is a manifestation of stress corrosion cracking (SCC) with the causative agent being most likely iodine. During SCC, cleavage of Zircaloys is prominent only on planes near the basal planes. Thermodynamic data by Cubicciotti et al (1) leads to the conclusion that solid iodides on the Zircaloy surface are involved in the mechanism of iodine-induced SCC. It is shown also that ZrI_4 must be the gaseous species involved in any mechanism in which vapor transport of Zr is important.

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Two metallurgical parameters which can have an influence on SCC of Zircaloys are crystallographic texture and heat treatment condition.

The crystallographic texture, i.e. the development of a preferred orientation of the grains in the final product is one consequence of the fabrication process of the zirconium-based alloys. The concept of an f-parameter introduced by Kearns (2) can be used as a quantitative index of the effective texture, where the single f-number represents the effective fraction of basal poles aligned in any one reference direction.

Syrett et al (3) showed that the greater the tangential texture of the Zircaloy fuel cladding, the greater the susceptibility to SCC. Knorr and Pelloux (4) found that texture has a strong influence on K_{ISCC}.

This work was undertaken to determine the kinetics of iodine-induced crack growth in Zircaloy, using a fracture mechanics approach. Crack propagation occurs by a mixture of cleavage and ductile tearing (fluting) in CWSR materials but may involve intergranular processes in unirradiated, recrystallized materials (5).

MATERIALS AND TEST METHODS

Double cantilever beam (DCB) type of specimen from plate material was used in this work. They have dimensions of 12.7 mm width by 12.7 mm height (2h) by 40.6 mm total length with a machined notch 16.5 mm long. The specimens are machined from different orientations in the plate. In the designation of orientation, the first letter gives the direction of specimen loading (i.e., crack opening) while the second letter indicates the direction of crack growth. Table 1 shows the chemical composition of these plates.

The fabrication schedules of the plates were controlled in a way which resulted in two different initial textures. Plate "S" texture representated that which results from standard commercial plate rolling procedure. The texture consists of one with basal planes parallel to the rolling plane and basal poles in the normal-transverse plane tilted approximately 40 degrees to the transverse direction from the normal direction. Plate "P" is warm cross-rolled to give a normal (0^o) texture; (0002) pole figures for both materials are shown in (4). Texture f-numbers are listed in Table 2.

The plates were subjected to two different heat treatments. The grains of the CWSR material are flattened with the smallest grain dimension measured through the plate thickness; for both plates that grain dimension is 4-6 μm. The dimensions in the longitudinal direction are in the range of 20-25 μm and in the transverse direction 12-15 μm. The grain size for the recrystallized plate is 8-10 μm.

SCC growth tests were conducted at 298, 373, and 583 K in highly purified argon + 40Pa iodine flowing gas environment. The gas purification system illustrated in Figure 1 was able to deliver a partial pressure of iodine of minimal water vapor and metal iodides content. The system was built with materials known to be inert to iodine attack. The test chamber was made with glass and gold-plated stainless steel to prevent volatile metal iodides from the test environment. The chamber has mechanical feed-through to permit the application of an external load to the specimen on a universal testing machine.

Stress intensity factor for DCB-type specimen can be calculated using the expression:

$$K_I = (2p/Bh^2).(3h(a+.6h)^2 + h^3)^{\frac{1}{2}} \quad (1)$$

To perform a SCC test using this specimen, the fatigue pre-cracked specimen is exposed to the environment and loaded to an initial displacement. The load drops due to crack growth and the crack length a is measured on the side of the specimen as a function of time. The crack tip velocity can be calculated from the slope of the crack length versus time plot. Stress intensities corresponding to each measured velocity are obtained from the expression above if the corresponding a and p are known.

RESULTS

Iodine-induced stress corrosion crack growth was observed for all orientations tested and at all temperatures investigated.

The general appearance of da/dt vs. K behavior at 583K of the S-plate material for three different orientations is shown in Figure 2. A low-K region (regime I) exhibited strong K dependence SCC growth, and a high-K region (regime II) where the K dependence of da/dt is weaker. The data obtained by Cox and Wood (6) at 573K is plotted in this figure for comparison.

Figure 3 shows the effect of the heat treatment on the crack growth behavior of specimens of S-plate, NL orientation. For the same K value, the crack growth rate was higher for the stress--relieved material. At low K values there was some evidence of intergranular cracking in the recrystallized material.

Figure 4 gives the crack growth rate as a function of K for specimens of CWSR S-plate material (NT orientation) tested at 298, 373, and 583K. Scanning electron microscopy revealed a tendency for extensive "fluting" in the transgranular fractures at room temperature and also at 373K.

DISCUSSION

The hypothesis of an adsorption-type mechanism which reduces the Zr-Zr bond strength in certain crystallographic directions at the crack tip is a reasonable explanation for the predominantly transgranular crack propagation in the iodine-induced SCC of Zircaloy. The study done by Krishnan et al (7) provides a better understanding of the chemisorption process of iodine on clean zirconium surfaces. Their results show that iodine chemisorbed on zirconium is chemically more stable than combined in the zirconium iodides. So, iodine can initially react with exposed zirconium surface and the solid iodide available to be transferred to fresh zirconium surface, when the metal is strained. Possible mechanisms of transference are surface diffusion of iodides species or vapor transport as ZrI_4 gas.

Due to the mostly crystallographic transgranular cracking of iodine-induced SCC of Zircaloy, it is not surprizing that texture influences the velocity and morphology of crack propagation as shown by our results. Hence, texture that aligns basal poles toward the tensile stress axis should increase the rate of subcritical crack growth at a given K value. The crack growth rate for the P-plate, NT orientation (f_N = 0.70) is the highest among those specimens tested at 583K. Also, the rates of crack growth for specimens of S-plate in both NL or NT orientation (f_N = 0.56) are higher than that for the TL orientation (f_T = 0.33) as shown in Figure 2.

Independently of the texture effects, the microstructural directionality of the grains also affects the rate of crack growth. The grains tend to become elongated in the rolling direction during fabrication leading to anisotropy of the crack growth rate, i.e. the velocity of cracks in specimens of NL orientation (crack growth parallel to the rolling direction) are higher than those for specimens in the NT orientation (crack growth normal to the rolling direction) for all values of K in tests at 583K.

As shown in Figure 3, the crack velocities for SR Zircaloy are higher than those for recrystallized Zircaloy at a given K value. In a qualitative sense, this is consistent with the higher mechanical strength and lower ductility of the SR material. In addition, the extrapolation of the curves corroborates the results of (4) who have found that K_{ISCC} for CWSR Zircaloy is lower than for the recrystallized material.

In a number of iodine-containing environments (8), the SCC growth in Zircaloy appears to start as an intergranular chemical process and changes to a transgranular propagation process. In this work some evidence of intergranular cracking is observed in the recrystallized material at low K values (near K_{ISCC}) as shown in Figure 3. The crack growth rates for intergranular processes are much slower than for transgranular cracking. Intergranular cracking seems to require a combination of a suitable K value and heat treatment condition (recrystallized material).

There is often a significant occurrence of fluting and other ductile processes during tests at lower temperatures (Figure 4); this is indicative of the high stress levels (high K values for a given crack depth) reached in the experiments since the fluted regions are associated with high local tensile strain (8). The flute spacing for a specimen tested at room temperature, for instance, is about 5 μm whereas the grain size is relatively small (15-25 μm). The occurrence of fluting requires considerable expenditure of strain energy in the process of crack propagation leading to a much slower growth rate as evidenced in Figure 4. This result is in conflict with results by (6) who have found little or no difference in crack velocity for Zircaloy tested in iodine between 298 and 573K. It is also apparent in Figure 4 that the extrapolated K_{ISCC} value is displaced to higher K values as the temperature increases.

SYMBOLS USED

LWR = Light Water Reactor

PWR = Pressurized Water Reactor

BWR = Boiling Water Reactor

p = Load (kg)

h = One-half the specimen height (m)

L = Longitudinal direction in the plate

N = Normal direction in the plate

T = Transverse direction in the plate

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TABLE 1 - Chemical Composition of Zircaloy Plates

Chief Alloying Elements	S-Plate(Zircaloy 4)	P-Plate(Zircaloy 2)
Sn	1.52w/o	1.51w/o
Fe	.20w/o	.14w/o
Cr	.11w/o	.10w/o
Ni	35 ppm	.05w/o
O	1160 ppm	1310 ppm

TABLE 2 - Kearns' f-Number for Zircaloy Plates

f-Number	S-Plate(Zry 4) SR	S-Plate(Zry 4) RX	P-Plate(Zry 2) SR
f_N	.56	.59	.70
f_T	.33	.32	.19
f_L	.09	.09	.09

S-Plate: SR-Stress Relieved at 785K for 4 hours
RX-Recrystallized at 898K for 4 hours

P-Plate: SR-Stress Relieved at 770K for 4 hours

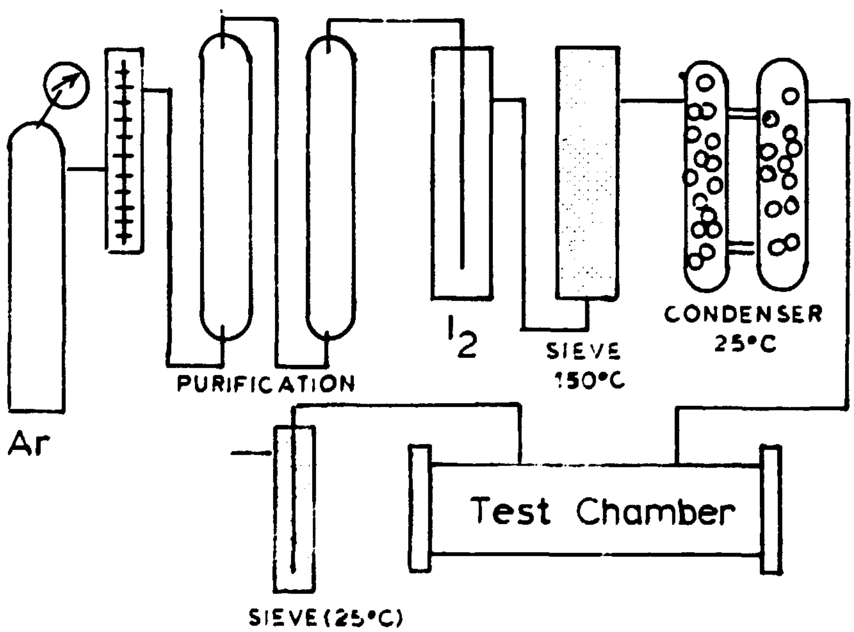

Figure 1 Diagram of the flowing gas (Ar + 40Pa I_2) system

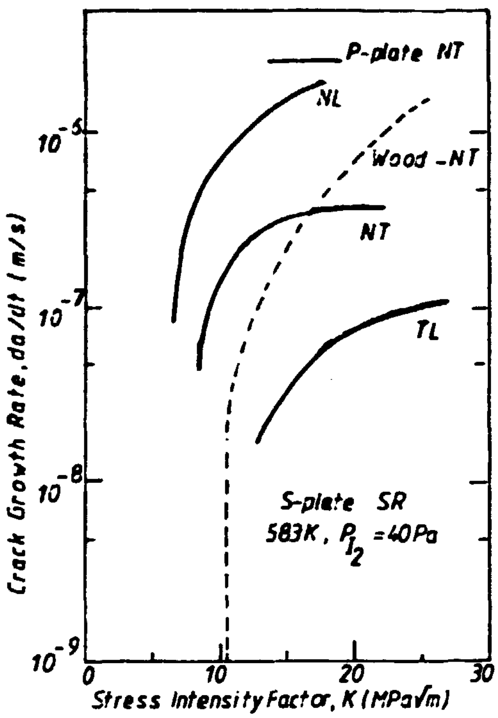

Figure 2 da/dt vs K behavior at 583K

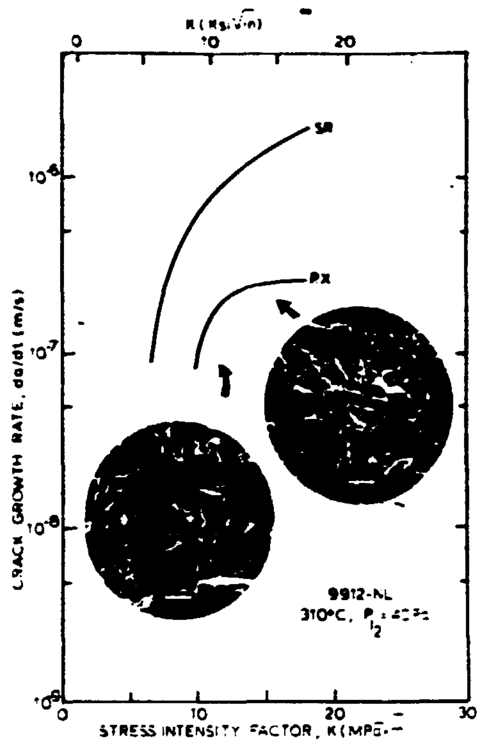

Figure 3 Effect of heat treatment on crack growth of S-plate

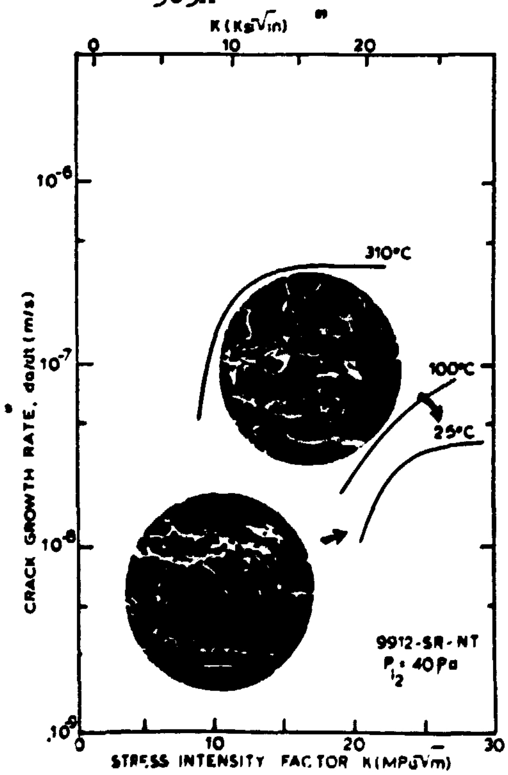

Figure 4 Effect of temperature on crack growth of S-plate material