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Some Mechanistic Observations on the Crack Growth Characteristics of Pressure Vessel and Piping Steels in PWR Environment*

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Abstract

The fatigue crack growth behavior of A533B and A508 pressure vessel steel and AISI Types 304 and 316 steels used in reactor coolant piping have been studied in a pressurized water reactor environment at 288°C(550°F). The influence of stress ratio (P_{min}/P_{max}), frequency, ramp times, specimen orientation and material microstructures were included in the study. While none of the materials showed evidence of static crack growth in the environment, the ferritic steels did show an enhanced fatigue crack growth rate at test frequencies of five cycles per minute and lower. Based on fractographic examinations the enhanced growth rate is not the result of environmentally induced intergranular or cleavage modes of crack propagation. Instead, striation spacing measurements were found to agree with the macroscopic crack growth rate, demonstrating a time dependent environmental interaction which introduces a frequency dependent enhancement of the mechanically developed striations. Crack growth experiments using hold times have confirmed the absence of any superimposed contribution of static crack growth components. Fatigue crack growth tests were conducted in an environment of Hydrogen Sulfide gas to establish the contribution of hydrogen embrittlement and will also be described.

Introduction

For the past several years a number of programs have been conducted to study the corrosion fatigue crack growth characteristics of pressure vessel and piping steels used in pressurized water reactor systems. The goals of the work are to characterize the crack growth behavior and to gain some understanding of the mechanisms which cause the behavior. It is evident that to

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satisfy the first goal without the second would be ill-advised, particularly in view of the complex environmental interactions which are produced in the pressurized water reactor environment. The purpose of this paper is to review the data now available for pressure vessel and piping steels in this environment, and to suggest some explanation for the observed behavior.

Two classes of materials will be discussed, pressure vessel steels, and reactor coolant piping steels. The pressure vessel steels are medium strength ferritic steels (SA508 Cl 2 and SA533B Cl 1), while the piping steels are austenitic stainless steels (SA304 and SA316). Pressure vessels are fabricated of plate and forging material, while the piping may be forged or centrifugally cast stainless steel.

The results discussed herein were obtained from sinusoidal load controlled tests of specimens taken from production heats of these materials. Specimens have been primarily two inch (5.08 cm) thick precracked fracture specimens of the compact and WOL configurations, and have been tested in an environment of PWR primary water at 288°C (550°F) and 14 MPa (2000 psi). Coupled with the testing program has been microscopic examination of the fracture surfaces of the tested specimens.

Experimental Results

The crack growth rate tests were conducted in environmental chambers which are actually small pressure vessels heated to the desired temperature with electrical heaters embedded in the chamber wall. The PWR chemistry is maintained by a flowing system with carefully monitored oxygen and thermal conductivity. The PWR water chemistry is essentially demineralized water with carefully limited Chlorides and Fluorides to which is added lithium hydroxide and Boron, which are used to control the nuclear reaction. A hydrogen overpressure is also added to the system, in both the operating reactor and the test autoclave. The resulting test environment remains essentially demineralized water, however, because the Boron dissociates at 288°C (550°F). Oxygen is controlled to be less than 0.1 ppm.

Crack growth is monitored continuously with an externally mounted linear variable differential transformer (LVDT) which reflects the face opening displacement of the specimen. The LVDT reading is then converted to crack length by a compliance technique, based on a calibration between LVDT and crack length. The resulting crack length vs. cycles data is then converted to crack growth rate data, using the incremental polynomial method recommended by ASTM. This method has the dual advantages of being the least subjective method of data processing, as well as producing a large amount of data, which aids in establishing trends and making comparisons.

Ferritic Steels

Fatigue crack growth rates for the pressure vessel steels in PWR environment are considerably enhanced over those obtained in air [1,2]. The degree of enhancement is related to the cyclic frequency of the test, with growth rates increasing as the frequency is decreased, presumably because this allows the environment more time to affect the crack growth process. A summary of the effect of frequency on crack growth rates for these steels is contained in Figure 1. This figure shows results for steels tested at R = 0.2, but similar behavior has been obtained at R = 0.7 and would be expected at other R ratios. The most enhanced crack growth occurs at cyclic frequencies of 0.5 and 1.0 cycle per minute, and this behavior shows a distinct curvature for the crack growth rate when portrayed on a logarithmic plot versus the range of stress intensity factor ΔK. At faster cyclic frequencies, i.e. 5 cycles per minute, the crack growth is enhanced over the behavior in air, but much less than the one cycle per minute data. Also the crack growth behavior seems to curve over to become parallel to the air data at a lower value of ΔK than does the 1 cpm data. For the 5 cpm data this occurs at such a low value of ΔK that the behavior appears to be mostly parallel to the air data.

In tests that were conducted at very low frequencies of 0.1 cpm, the behavior is less clear because there is much more scatter. The overall behavior indicates that the crack growth is less enhanced than the data at 1.0 cpm, indicating that a saturation is occurring at between 0.5 and 1.0 cpm. The effect of frequency on crack growth is presently thought to be most strongly dependent on the loading rate, and substantially less dependent on hold time or unloading rates [1,2].

At a given loading frequency the most important effect on crack growth rate is R ratio, which causes increased growth rates at a given value of applied stress intensity factor range ΔK. An example of this effect is given in Figure 2. As with the frequency effect, the R ratio effect is peculiar to the water environment; neither is observed in tests of the same material in air, although such is not the case for the austenitic stainless steels.

Data obtained for forging and plate material show no discernable difference, and welds display the same or slightly lower growth rates as the base metal. Frequent small reversals are observed in the crack growth rates in welds, as shown for example in Figure 3.

Austenitic Stainless Steels

Unlike the ferritic steels, these steels have essentially the same behavior in the PWR environment as in air [3]. There is an important R ratio effect, as shown in Figure 4, but the test frequency (or loading rate) appears to have little or no effect on the crack growth rates. Growth rates observed in welds are slightly lower than the base metal [3,4].

Crack growth behavior in both the forged (SA304) and centrifugally cast (SA316) materials was indistinguishable, even through these two materials have a widely differing grain size. Data from the literature [4] indicate that SA304 and SA316 plate material also have similar behavior in air.

Behavior of the Ferritic Vessel Steels

In attempting to understand the mechanisms of corrosion fatigue crack growth in the PWR environment, a comprehensive review of the literature was undertaken, and extensive microscopic examination of the crack surfaces of tested specimens was conducted. Also an auxiliary series of crack growth rate tests was conducted on these steels in an environment of 60 psi hydrogen sulfide gas. These tests were conducted at room temperature and 93°C (200°F) and results were obtained at R = 0.2 and 0.7. The crack growth rate behavior was very similar to that obtained in PWR environment at the same R ratio, at low and intermediate values of ΔK, as shown for example in Figure 5. At the higher values of ΔK the crack growth rate in the PWR environment increases at a slower rate with ΔK, and thus the data tend to curve over, but the growth rate in hydrogen sulfide continues to increase steadily in this region, as shown in Figure 5.

Crack growth rate data now available on pressure vessel steels in PWR environment indicate that there are two distinctly different regimes of behavior for the most accelerated crack growth rate data. At low values of ΔK the crack growth rate increases rapidly with ΔK, but at higher values of ΔK the rate of increase declines, and the data curve over, as shown in Figures 1 and 2 for example. These two regimes will be discussed separately, after which the results of microscopic examination of the fracture surfaces will be presented.

Low ΔK Behavior

The present data indicate that there is an apparent threshold ΔK below which there is little or no crack growth. This apparent threshold is dependent on the R ratio of the test, as shown in Figure 2. From the two R ratios tested (0.2, 0.7) the data can be correlated to a function of K_{max}, the maximum applied stress intensity factor. This agrees with the model for threshold ΔK developed by Richie [5], using a critical stress criterion for crack growth. On the basis of this model the load ratio R only influences threshold ΔK (ΔK_{th}) in an environment where the influence of hydrogen in reducing the fracture stress is related to the stress assisted accumulation of hydrogen in the region of hydrostatic tension near the crack tip. For a given yield strength, temperature, and environment, the amount of hydrogen accumulated is influenced only by K_{max}. The model further predicts that ΔK_{th} should be influenced by the environment. The greater the unstressed equilibrium concentration of hydrogen that the environment introduces into the steel, the lower ΔK_{th}.

Based on this model it would be expected that the hydrogen sulfide environment would produce a lower ΔK_{th} than the high temperature PWR water environment. The present data for the H_2S environment has a finite slope, and results are not available at very low growth rates where ΔK_{th} would be defined, so a precise value has not been obtained. It is clear from the data now available (Figure 5 for example) that the data extend beyond the apparent ΔK_{th} observed for the PWR environment, and are thus consistent with the model.

The model predicts that threshold ΔK should be independent of frequency or loading rate, since it is based on equilibrium segregation behavior. The data now available are consistent with this model, as are other data obtained for similar material - environment combinations in the literature [6,7,8].

Other models for environmental threshold ΔK based on the idea of a critical COD to expose fresh metal to the environment do not explain this dependence on environment. Although the lack of any static stress corrosion cracking [1] implies the necessity for cracking a protective film to allow hydrogen to enter the material, the crack growth enhancement as well as the threshold behavior appear to be directly related to the amount of hydrogen present.

The apparent threshold ΔK occurs at a value well above the threshold ΔK measured for these steels in an air environment of approximately 2 ksi $\sqrt{in}$ [9].

Although no deliberate attempt has been made in the present study to determine ΔK_{th} for the PWR environment, specimens have been cycled for up to a month at lower values of ΔK than the apparent threshold, with no observable growth, which would indicate that the effect is real.

Apparent thresholds for fatigue crack growth in the environment greater than the value in air and corrosion fatigue crack growth rates higher than those in air have been frequently observed in previous work [6,7,10,11,12] on fatigue crack growth of steels below K_{ISCC}, although no attempt has been made to study this effect in detail using the precautions necessary for ΔK_{th} determination. One of the major reasons for this lack of information is that the environmental enhancement only occurs at low frequencies and thus the time required for such tests would be prohibitive. Several investigations indicate that the apparent threshold measured coincides with the initially applied ΔK [8,12]. This agrees with behavior observed in the present study, when higher initial values of ΔK were used than those shown in Figures 1 through 3. This effect is illustrated in Figure 6, which shows that specimens started at different levels of ΔK show a different crack growth behavior. The effect of higher initial loading is to retard the environmental effect for some time, and this time can often last the full period of the test, as shown in Figure 6. The cause of this behavior appears to be related to the kinetics of the water-steel system, but is not yet well understood. Such behavior is not obtained in systems with slower kinetics (steel-air) or systems with faster kinetics (steel-Hydrogen sulfide) [13].

Behavior at High ΔK

Above the threshold ΔK the crack growth rate increases with ΔK much faster in the PWR environment than in air, as illustrated in Figures 1 and 2. As in static stress corrosion cracking, the synergistic effect of increasing K and increasing the available hydrogen combine to give this effect.

The steep rise does not continue, but at a particular value of ΔK the growth rate slows down, becoming nearly independent of ΔK. The point at which this occurs is dependent on the frequency, increasing with decreasing frequencies. This effect was also observed by Vosikovsky [6]. It is also dependent on the environment - in the hydrogen sulfide atmosphere no plateau region is developed, as seen in Figure 5.

Although plateau regions in fatigue crack growth are frequently seen in Aluminum, magnesium and titanium alloys [14], their incidence in low alloy steels is only occasionally reported [6,7,10], with more often results being parallel to air, or a slight bending over [8,11,15,16].

This plateau region analogous to stress corrosion cracking behavior can be related to an environmentally controlled phenomenon, the requirement that the crack must wait for the hydrogen to build up to the concentration required for propagation. Testing at a slower frequency allows more hydrogen to accumulate per cycle, and thus allows a faster crack growth rate. By adding a kinetically faster environment to produce hydrogen - the hydrogen sulfide - no plateau was seen in the growth rate because hydrogen can be supplied at a fast enough rate to sustain growth.

One aspect of this plateau region is the tendancy of the cracks to branch, while in the absence of a plateau region crack branching is self limited due to decreasing K at the tip when a branch forms, and the high K dependence of the crack growth. With the plateau region the strong K dependence no longer exists, and when the applied ΔK level is sufficiently high (about $3 \Delta K_{th}$) then macro-branched cracks have sufficient driving force to allow propagation.

The macrobranching is also strongly dependent on the material being tested. Macrobranches have not been observed in forgings for example, and seem to be limited to plate material, which displays some effect of the rolling process. Earlier specimens from HSST plate 02 tested at R = 0.2 with the crack plane parallel to the rolling direction showed extensive macrobranching at a value of $\Delta K \cong 2.3\ \Delta K_{th}$ [13]. Subsequent tests on the same plate with a crack plane perpendicular to the rolling direction with R = 0.67 showed much less propensity for macrobranching, with only one specimen (02GB4) showing an extensive branch, although microbranching was observed on all specimens, both plate and forging. One explanation for the lack of extensive branching is that at the higher R ratio the test must be concluded before the applied ΔK reaches the required level of approximately $3\ \Delta K_{th}$.

It is not clear why specimen 02GB4 branched, but it appears to have originated at a beach mark produced at 10 Hz to correlate compliance and crack length before returning to the 0.1 cycle per minute testing rate. Since no data were taken during the beach marking process, a check on the plateau requirement could not be made. More limited crack branching events were also apparent in specimen 02GB-5, originating during the loading at 1 Hz used to mark the specimen, and continuing once the specimen was returned to the test frequency of one cycle per minute. A similar behavior occurred in specimen 04A102, from HSST plate 04 as shown in Figure 7. Again the branch was initiated at the 1 Hz beach mark, and although this mark was only 0.025 cm (0.01 inches) long the branch continued for 0.508 cm (0.2 inches) into the specimen after the test conditions were returned to one cycle per minute. The crack growth behavior for this specimen was well developed and shows a gradual development of the plateau region as seen in Figure 8, however some arrests were seen.

The hydrogen sulfide specimens, which did not show any plateau region, also were completely free of macrobranches. This is consistent with the proposed model.

Fractography

The mechanisms of fatigue crack growth in the present experiments are dependent on both the material and environment. Examination of the fracture surfaces indicates that different mechanisms of crack growth are occurring in the base metal and the weld specimens, as shown in Figure 9.

In the base metal, both A533B and A508 Cl. 2, all the crack propagation in the PWR environment occurs by ductile striation formation. The striation spacings measured agree completely with the macroscopic growth throughout the range of applied stress intensity factor where measurements could be made. This agreement held for all values of R ratio and cyclic frequency, confirming the absence of any other environmentally induced cracking mechanism.

Another mechanism of crack growth occurs in the weld specimens, void coalescence. This mechanism occurs in both air and PWR water environment, but the crack growth in the PWR environment is considerably enhanced, as has been observed previously in both base metal [18,19] and welds [20]. In the present experiments the transition in fractography from striations to void coalescence is related to the much higher density of large carbide particles in the weld metal as shown in Figure 10. Voids form at particle interfaces as a result of plastic strain at the crack tip, and the strain required decreases with increasing particle density.

The influence of hydrogen on the nucleation and growth of microvoids in the plastic zone has been recently reviewed [21], and could include increased nucleation through a decrease in the particle matrix cohesive energy or growth due to a buildup of hydrogen pressure in the void. The enhancement of growth rates in the water environment implies that hydrogen is able to penetrate the plastic zone to the extent of about the crack opening displacement (COD) during the cyclic test, which is approximately one microinch (2.5×10^{-6} cm) for an applied stress intensity factor of 25 ksi $\sqrt{\text{in}}$ (27.5 MPa$\sqrt{\text{m}}$). With a diffusion coefficient of 10^{-7} to 10^{-9} cm^2/sec only 0.06 to 6 seconds would be required for hydrogen transport by diffusion, so clearly this does not impose a kinematically limiting effect at the test frequencies imposed (1 cycle per minute imposes a 30 second rise time). In view of this, it is not surprising that although the microvoid coalescence represents a much different mechanism, it results in a similar crack growth rate.

The mechanism whereby the environment is able to assist the crack growth occurring by the plastic flow process in ductile striation formation is not clear, in view of the minor role played by hydrogen on the plastic flow properties of low alloy steels [22]. The observed effect is, however, consistent with the proposal of Beacham [23] whereby hydrogen is envisioned to aid whatever process the crack tip wants to take, implying hydrogen lowers the stress required for dislocation motion. This idea is recently supported by experiments of Bernstein [34], which could be interpreted to imply that hydrogen reduces the frictional stress and thereby makes dislocation motion easier.

Another mechanism that has been advanced which could explain the influence of hydrogen is the reduction in surface energy associated with the creation of a slip step on the surface [25]. This model also considers the effect of hydrogen in lowering the surface energy associated with cleavage crack propagation. The mechanism of crack advance by ductile plastic deformation is shown to be in delicate balance with the cleavage crack propagation, as illustrated schematically in Figure 12.

In a PWR water environment at 288°C (550°F) the specific surface energy associated with slip γ_G is lower than that required for cleavage $\gamma_{C\ell}$, resulting in ductile striations. As the temperature of the test is lowered the crack growth mechanism begins to switch to a cleavage mode. A specimen tested at 93°C (200°F) in the water environment showed a mixture of ductile striations and cleavage [26], indicating that $\gamma_{C\ell}$ and γ_G were nearly equal. Other tests conducted in room temperature demineralized water have produced almost entirely the cleavage mode of crack growth [27], which agrees well with the model, in that $\gamma_{C\ell}$ is lower than γ_G.

Hydrogen sulfide gas lowers the value of $\gamma_{C\ell}$ to a much greater extent than γ_G, resulting in a cleavage mode of propagation at both room temperature and 93°C (200°F). Although the diagram of Figure 11 is schematic, and drawn arbitrarily to be consistent with observations, it does predict that, assuming the environment is able to induce sufficient hydrogen, the mode of propagation will revert to ductile striations at higher temperatures in the hydrogen sulfide.

Austentic Steels Behavior

In the present experiments a PWR water environment has no measurable influence on the fatigue crack growth rates in either 304 or 316 stainless steels[3], for tests conducted at frequencies as low as one cycle per minute. A small frequency effect was noted by Rabbe, et.al. [28] but it is not clear whether this effect was real, or within the scatter of the data. Thus the frequency has at most a small impact on the crack growth behavior.

All crack growth occurs by a ductile striation mechanism, as shown in Figure 12, which unlike the ferritic material is not enhanced by the environment. Lacking a definite model for enhancement of the striated growth in the ferritic materials, it is not yet clear why no effect is seen in the austenitic steels. In general austenitics are less influenced by hydrogen than ferritics. Several factors may be important-the greater solubility of hydrogen in austenite, the slower diffusion kinetics, and a difference in the specific surface energies which may be associated with such events such as cleavage and slip step emergence.

Summary and Conclusions

The fatigue crack growth behavior of SA533B and SA508 pressure vessel steel and AISI Types 304 and 316 stainless steels has been studied in a pressurized water reactor environment, through tests as well as fractographic examination of the specimen surfaces. The following conclusions were reached.

1. Fatigue crack growth rates for the ferritic steels are accelerated in water environments over those rates obtained in air. Growth rates are dependent on both test frequency and R ratio.

2. Fatigue crack growth rates for the 300 series austentic stainless steels are not affected by the water environment. Growth rates are strongly dependent on R ratio, and only slightly if at all affected by test frequency.

3. The effect of frequency on growth rates in the ferritic steels appears to saturate between 0.5 and 1.0 cpm.

4. Crack growth behavior of ferritic steel in hydrogen sulfide is similar to that in the PWR environment, but there is no frequency effect - only the R ratio effect.

5. Fractography of the crack growth process in the ferritic steels depends on temperature, with the mode of growth changing from ductile striated growth to cleavage as the temperature decreases.

6. Similar trends to (5) occur for ferritic steels in hydrogen sulfide.

7. The transition from ductile to cleavage modes is well explained by a surface energy model.

8. Fractography of the austenitic stainless steels shows entirely ductile striations.

9. Fractography of ferritic welds shows a different mode of propagation, void coalescence, than exists in the base metal, although the crack growth rates are comparable.

10. The major cause for crack growth rate enhancement for the ferritic steels in the water environment was found to be hydrogen embrittlement.

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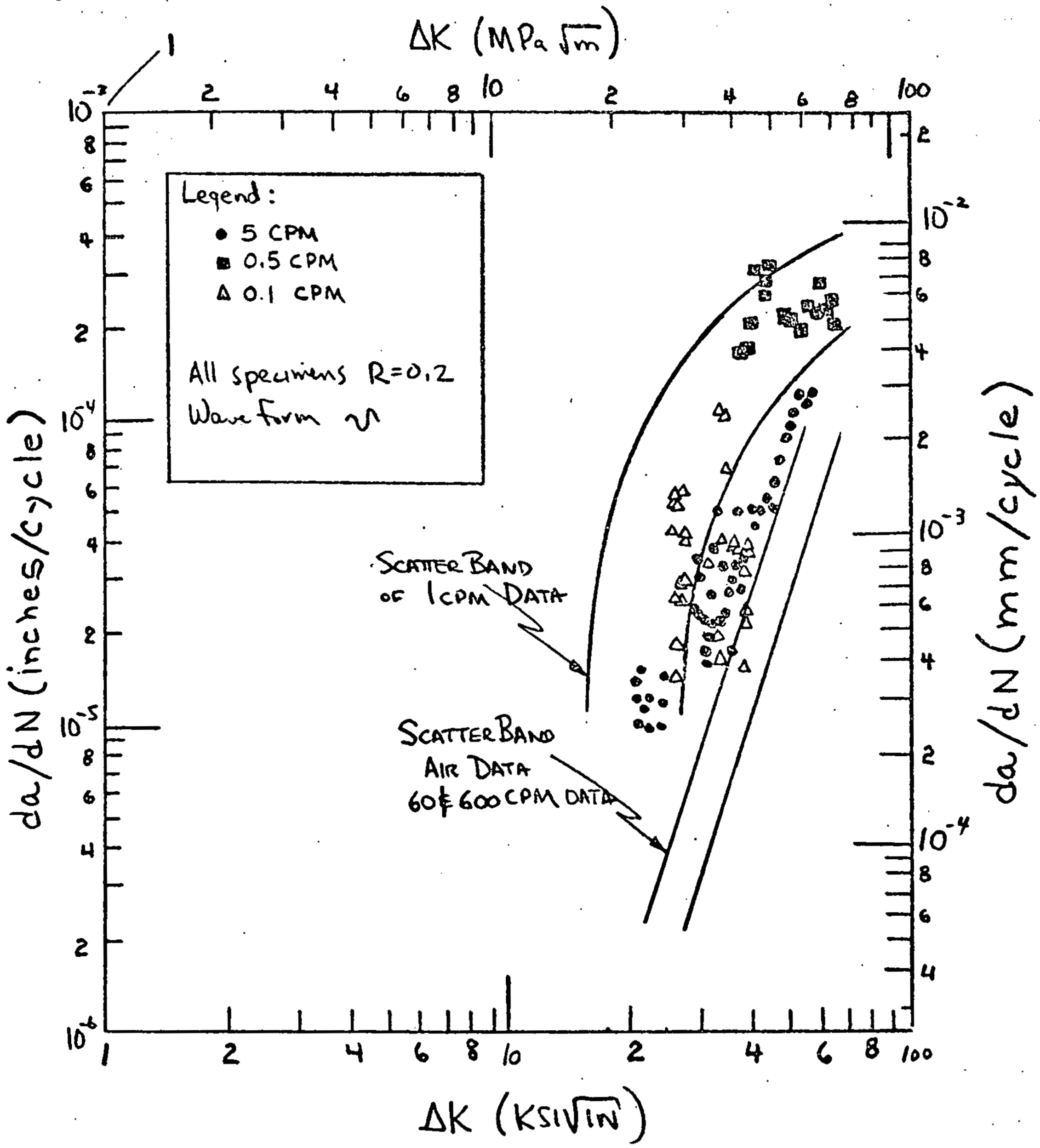

Figure 1: Effect of Cyclic Frequency on Crack Growth Rate - Pressure Vessel Steels & Welds in PWR Environment

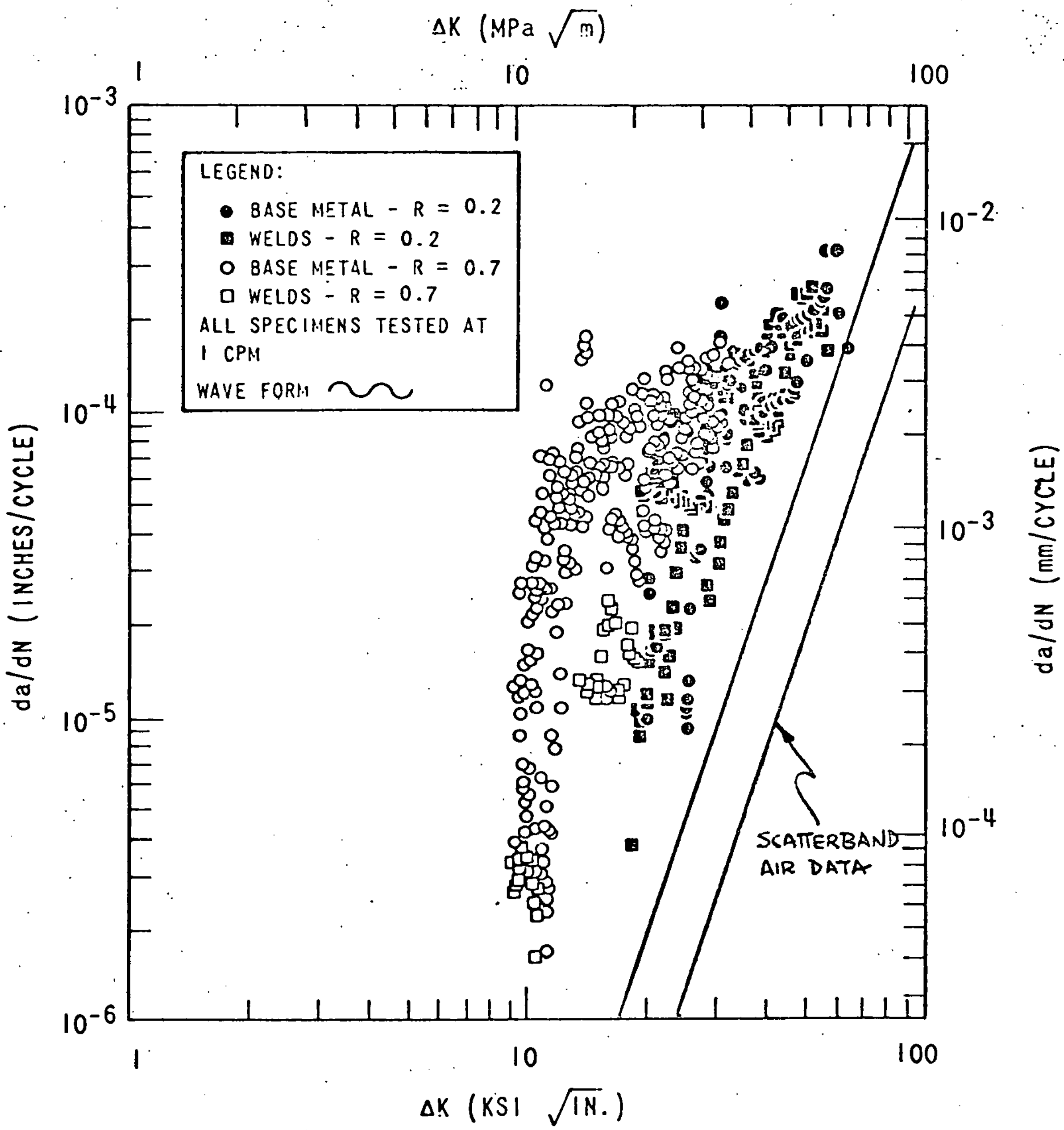

Figure 2: Effects of R Ratio - Base Metal and Welds in PWR Environment

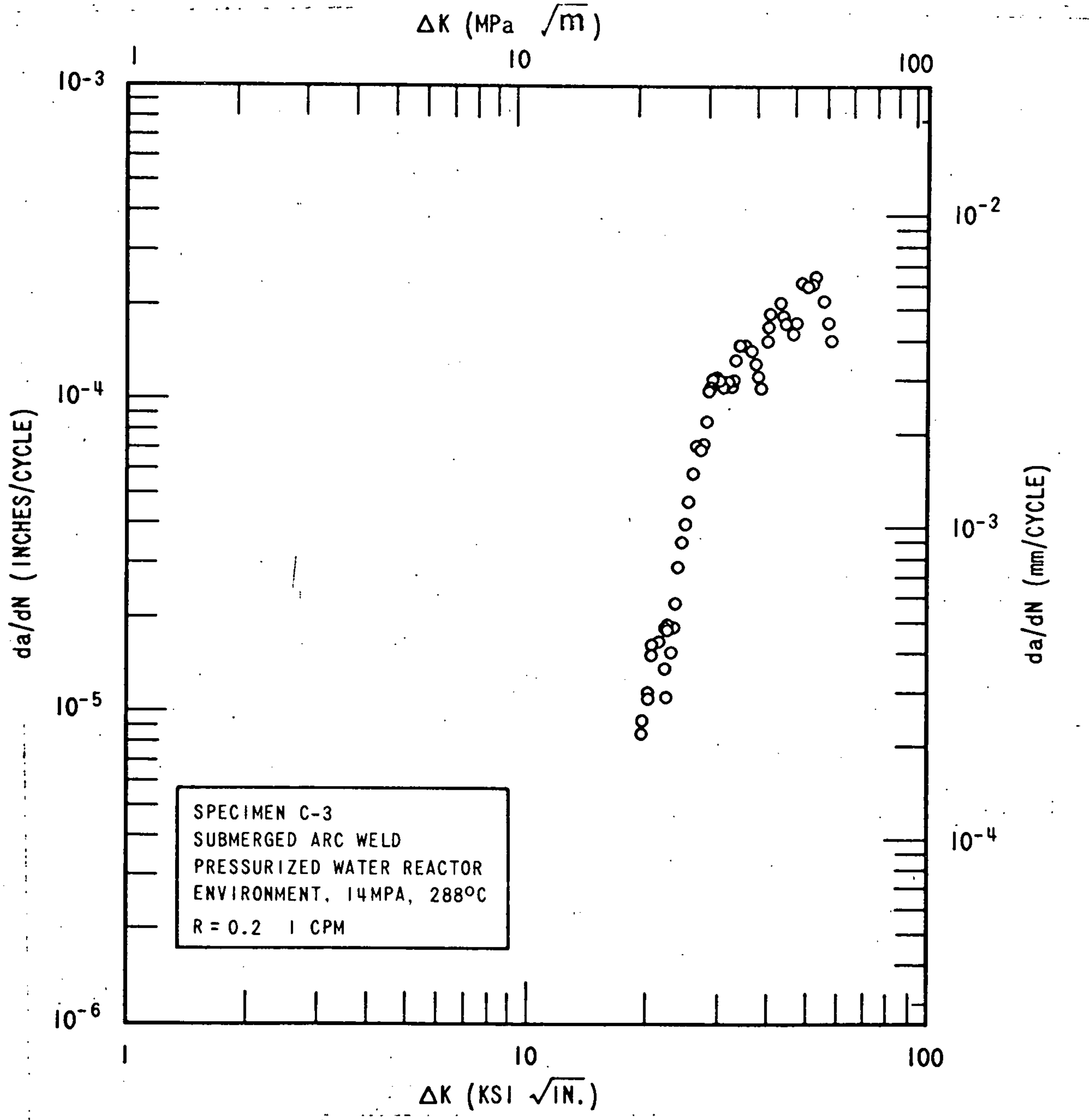

Figure 3: Fatigue Crack Growth Rate Results for Specimen C-3

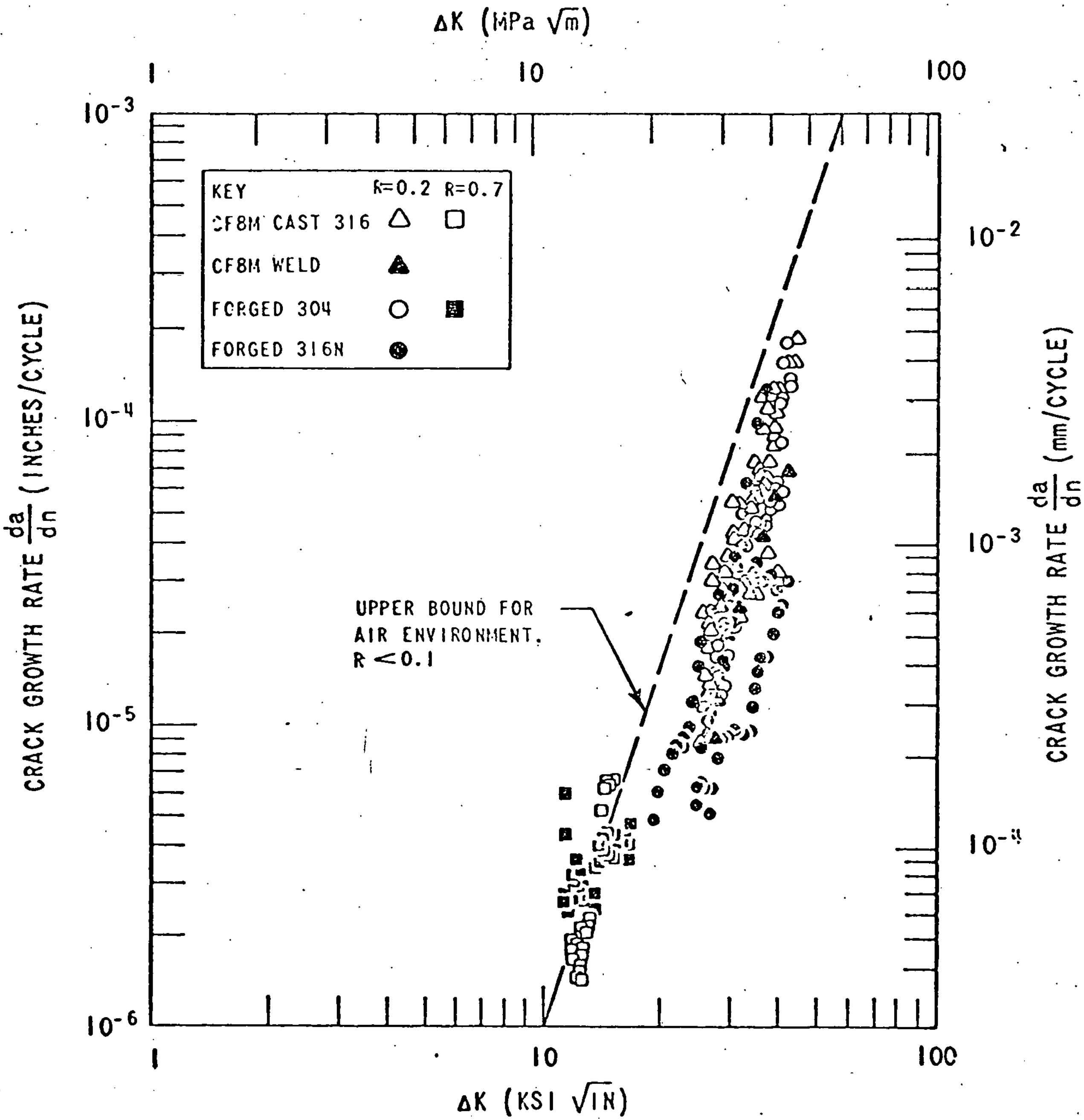

Figure 4: Fatigue Crack Growth Results - Stainless Steels in Pressurized Water Reactor Environment

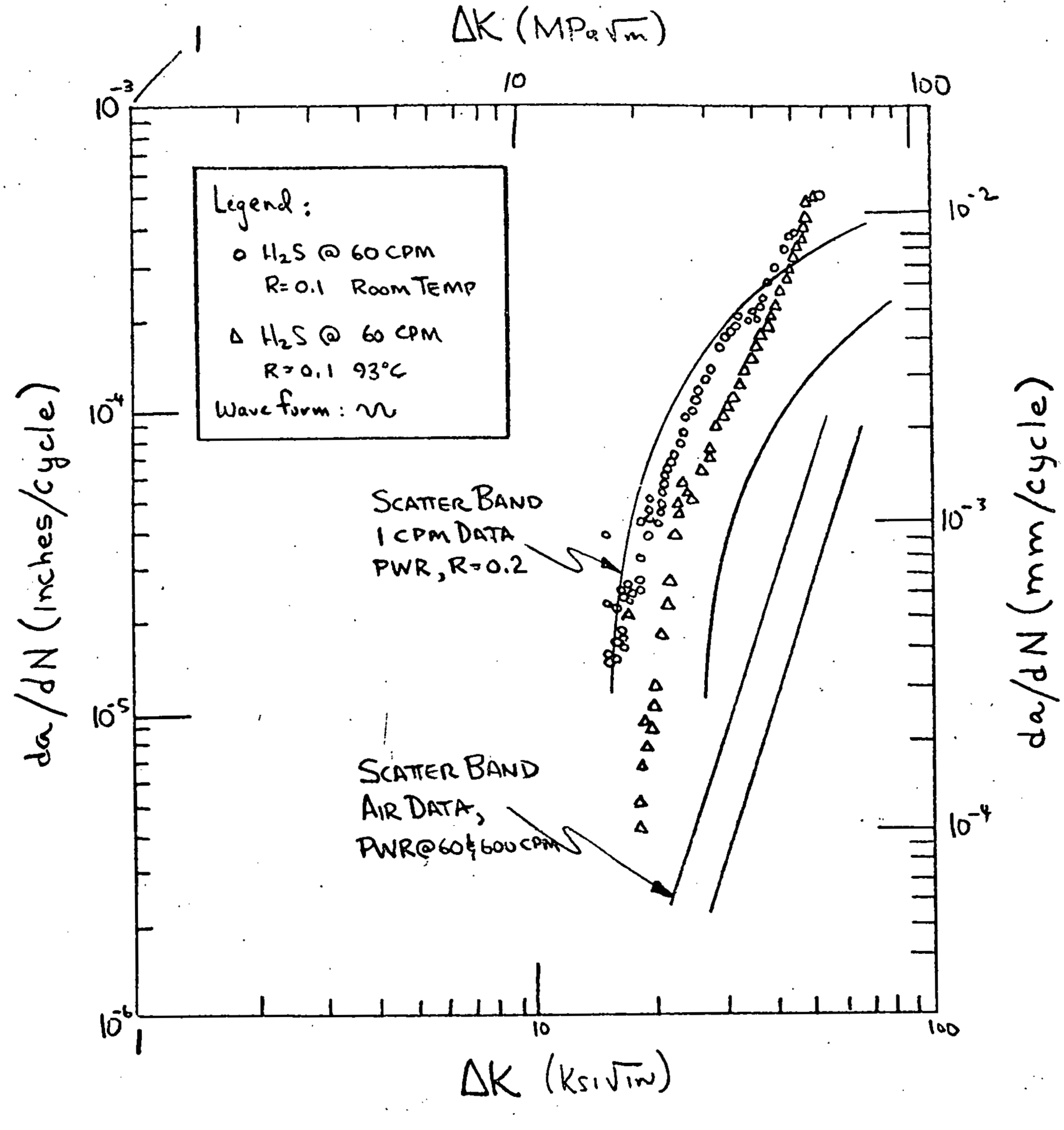

Figure 5: Fatigue Crack Growth Rate Results - Pressure Vessel Steels in PWR and Hydrogen Sulfide Environments

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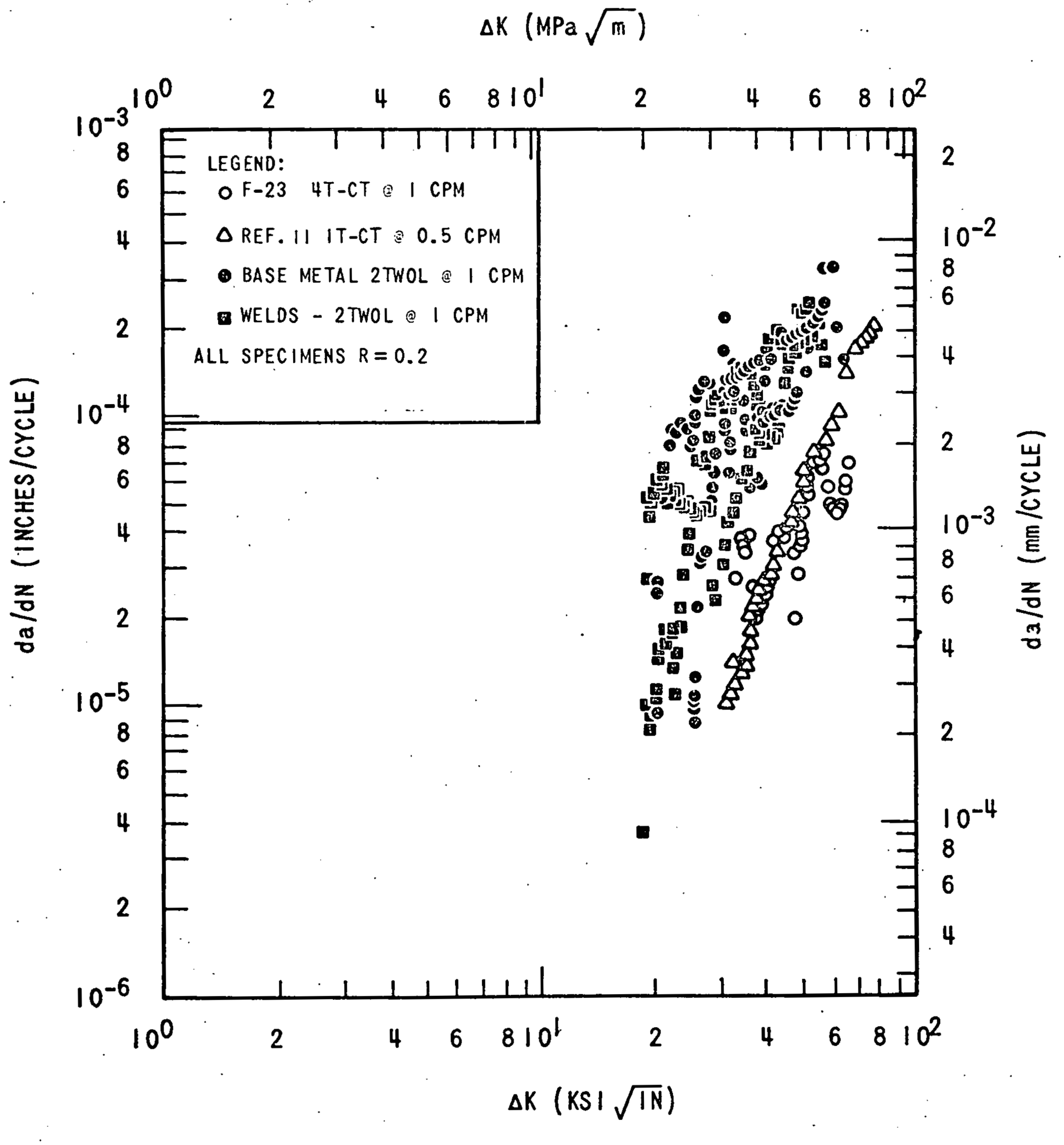


Figure 6: Example of the Effect of Starting Conditions on Fatigue Crack Growth - Pressure Vessel Steel in PWR Environment

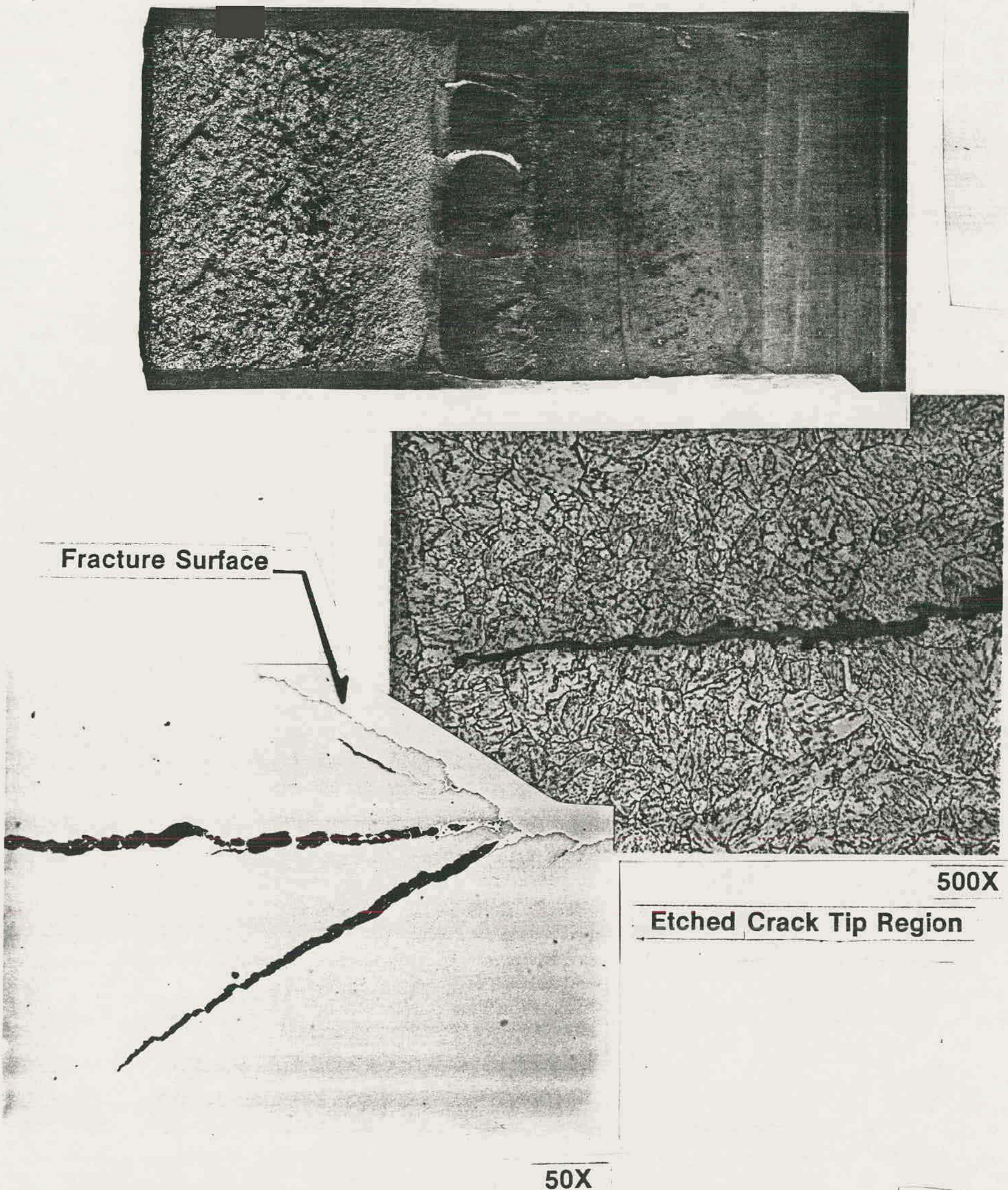


Figure 7: Branching Originating at a Beach Mark Specimen 04A-102, HSST Plate 04 in PWR Environment - 1 CPM, R=0.7

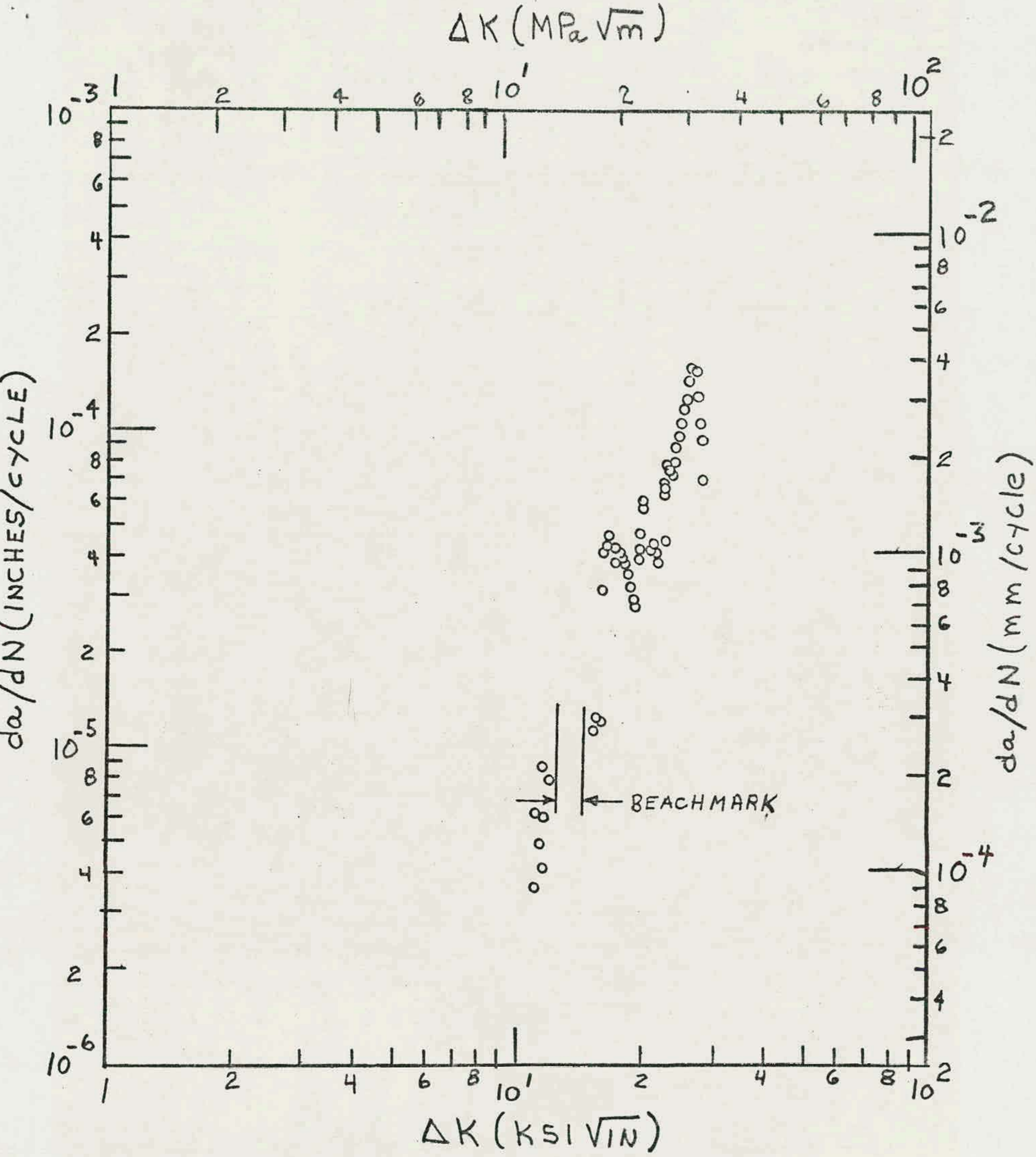
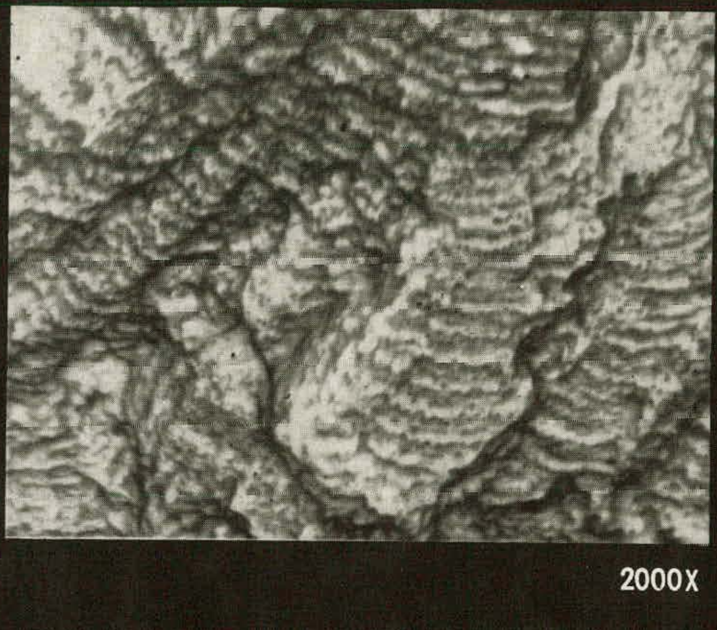


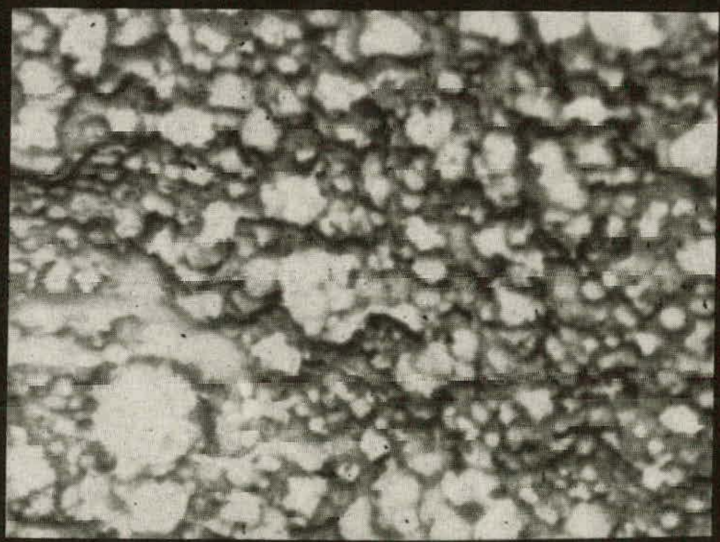
Figure 8: Fatigue Crack Growth--Pressurized Water Reactor Environment
Specumen 04A-102, A533B Cl 1 1.0 CPM, R=0.7

11984-17



2000X

SPECIMEN F-1 PWR ENVIRONMENT, 1CPM, R = 0.7



2000X

SPECIMEN C-1 PWR ENVIRONMENT, 1CPM, R = 0.7

Figure 9: Comparison of Fracture Surfaces - Weld (C-1) and Base Metal (F-1)

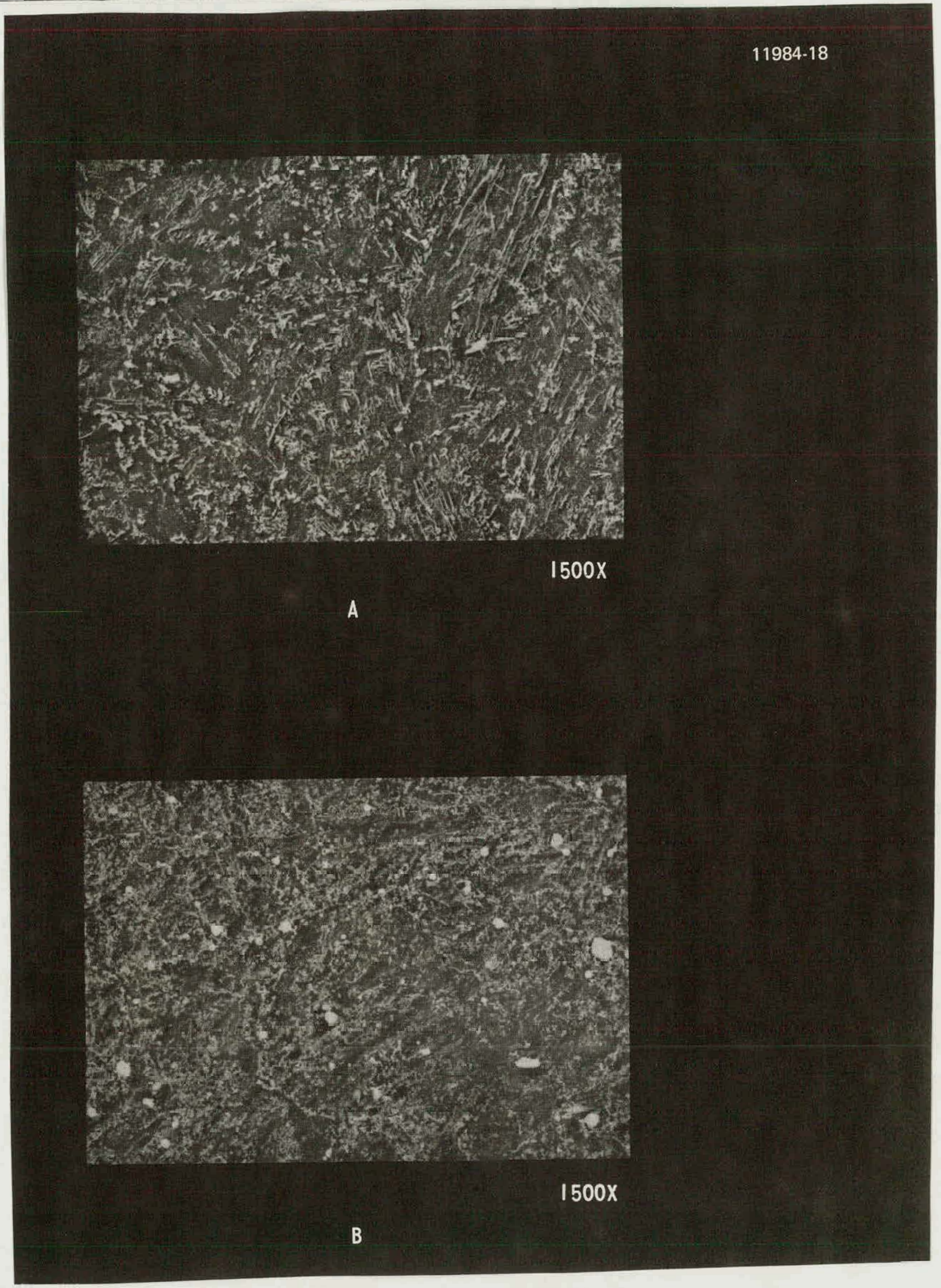


Figure 10: Extraction Replica of Carbides in (A) Base Metal Specimen 02GB-3, (B) Weld Metal Specimen C-2 Showing Larger Spherical Particles in the Weld Metal

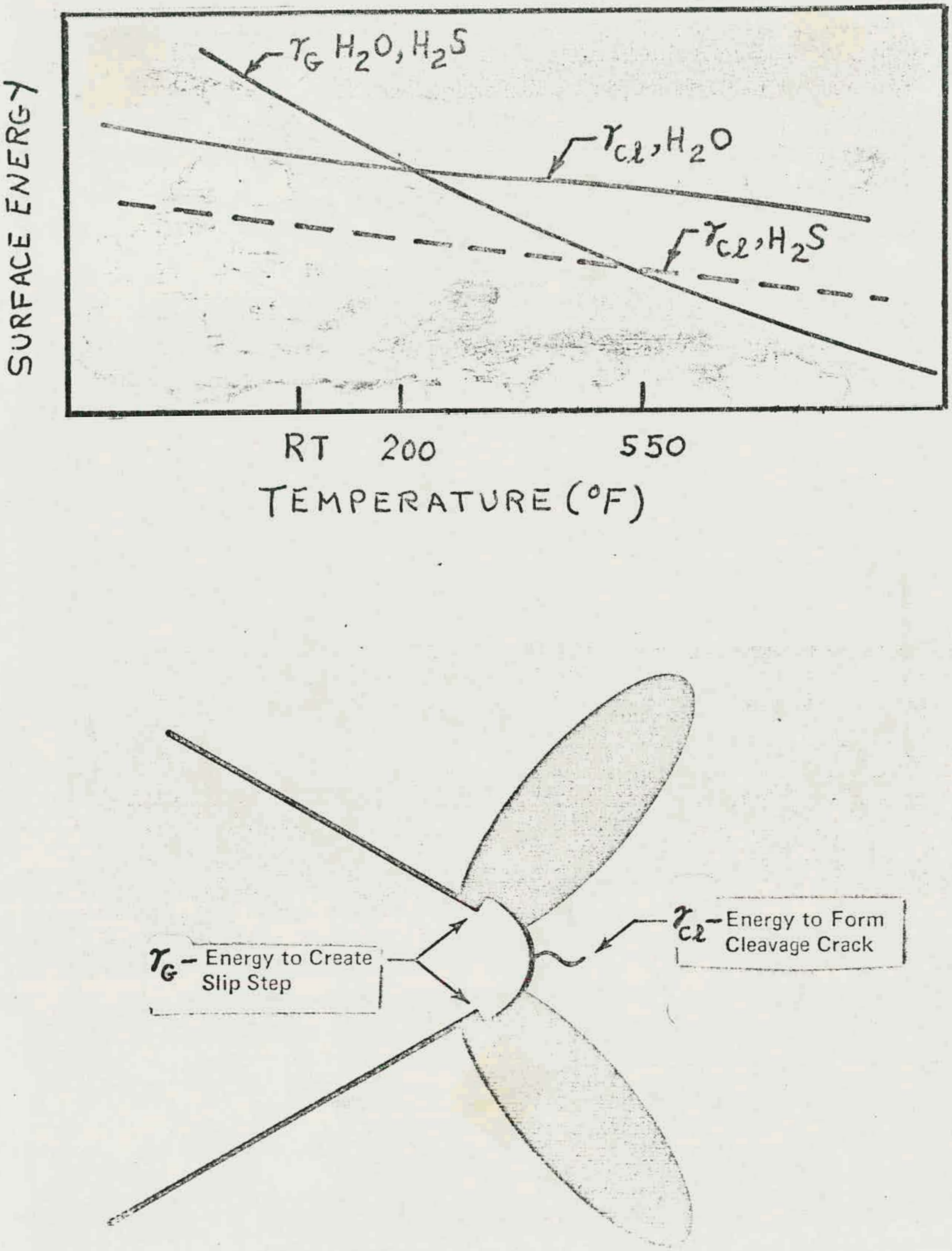


Figure 11: Influence of Environment on Crack Tip Behavior

Comments

1. (pp. 1 & 2) On p. 2, it is stated that only sinusoidal loading tests are to be discussed. However, on p. 1, both frequency and ramp time are mentioned (in the abstract) and hold time is also mentioned (in the text).

2. (p. 3) The fact that growth rate reaches a maximum as a function of frequency implies that something like crack-tip blunting due to anodic dissolution (see also ref. 6, pp. 303 & 304).

3. (p. 7) There appears to be an error in the COD value and the diffusion time calculations. Using

$$\delta = \epsilon_Y \left(\frac{K_I}{\sigma_Y} \right)^2 ,$$

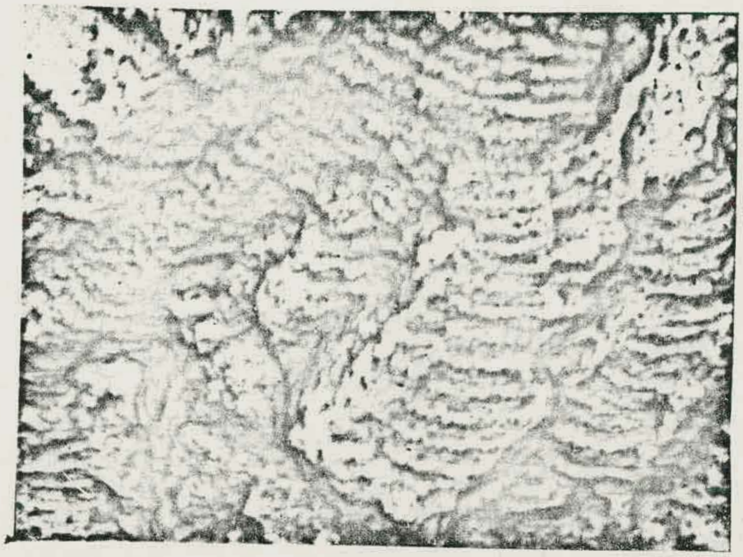
$K_I = 25 \text{ KSI} \sqrt{\text{IN}}$ and $\sigma_Y = 70$ KSI, $\delta = 7.6 \times 10^{-4}$ cm. Then, from

$$t = \frac{\delta^2}{\alpha} ,$$ where α is the diffusion

coefficient,

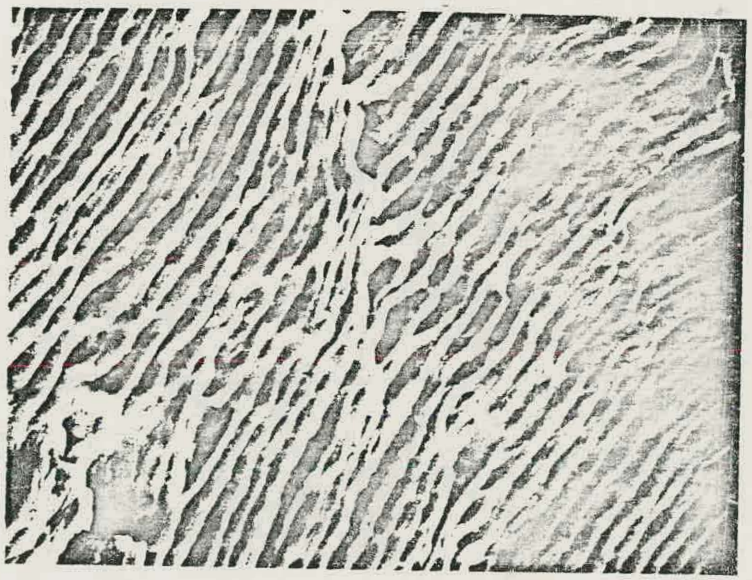
$$5.78 < t \quad 578 \text{ seconds.}$$

4. (p. 7) See the error in figure number at the end of the second to last paragraph.



2000X

SPECIMEN F-1



2000X

SPECIMEN SW-25

Figure 12. Comparison of Fracture Surfaces - Ferritic (F-1) and Stainless (SW-25) Steels in PWR Environment