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FRACTURE BEHAVIOR OF NICKEL-BASED ALLOYS IN WATER

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Abstract

The cracking resistance of Alloy 600, Alloy 690 and their welds, EN82H and EN52, was characterized by conducting J_{IC} tests in air and hydrogenated water. All test materials displayed excellent toughness in air and high temperature water, but Alloy 690 and the two welds were severely embrittled in low temperature water. In 54°C water with 150 cc H_2/kg H_2O, J_{IC} values were typically 70% to 95% lower than their air counterparts. The toughness degradation was associated with a fracture mechanism transition from microvoid coalescence to intergranular fracture. Comparison of the cracking response in water with that for hydrogen-precharged specimens tested in air demonstrated that susceptibility to low temperature cracking is due to hydrogen embrittlement of grain boundaries. The effects of water temperature, hydrogen content and loading rate on low temperature crack propagation were studied. In addition, testing of specimens containing natural weld defects and as-machined notches was performed to determine if low temperature cracking can initiate at these features. Unlike the other materials, Alloy 600 is not susceptible to low temperature cracking as the toughness in 54°C water remained high and a microvoid coalescence mechanism was operative in both air and water.

Introduction

Alloy 600 and its weld, EN82H, are used in commercial primary water reactors where they are exposed to water that can lead to stress corrosion cracking (SCC). Alloy 600 components that have failed due to SCC include piping, auxiliary penetrations, instrument nozzles and steam generator tubes.[1-3] Alloy 690 and its weld, EN52, are attractive candidates for replacing Alloy 600 components due to their superior high temperature SCC resistance.[4-6] Indeed, Alloy 690 has replaced Alloy 600 in pressurizer heater sleeves, instrument nozzles and steam generator tubes.[1,7] The expected failure processes for these components involve crack initiation and propagation due to high temperature SCC or corrosion fatigue, followed by stable or unstable tearing when the crack depth reaches a critical size. This work focuses on the final step in the tearing process.

Previous fracture mechanics tests[8,9] showed that Alloy 600, Alloy 690 and their welds possess excellent toughness in air and elevated temperature water with high hydrogen content. In low temperature water, however, the cracking resistance was reduced. The reduction in toughness for Alloy 600 was relatively small, whereas Alloy 690, EN82H welds and EN52 welds experienced a dramatic toughness degradation. The loss of toughness is associated with a fracture mechanism transition from ductile tearing to hydrogen-induced intergranular cracking in low temperature water. This paper summarizes the fracture behavior of these materials in water and presents new data concerning water temperature, hydrogen content and loading rate effects. It also examines the environmental cracking response for natural welding defects and as-machined notches.

Elastic-plastic J_{IC} test methods are needed to characterize the cracking behavior of these low strength materials. A fully ductile response is observed for all materials in air and high temperature water, while the behavior in 54°C water approaches a brittle, linear-elastic response. Elastic-plastic methodology provides a continuous scale for representing the environmental cracking response under all test conditions. The two main parameters obtained in elastic-plastic fracture test are J_{IC}, the fracture toughness at the onset of cracking, and the tearing modulus, T, which is a dimensionless measure of a material's resistance to cracking after J_{IC} is exceeded. To provide some insight into the propensity for component failure, cracking resistance can be classified into the following three broad categories:[10]

- Category I corresponds to low toughness materials with J_{IC} values less than 30 kJ/m^2 (K_{IC}<75 MPa$\sqrt{m}$) and tearing moduli less than 10 (dimensionless). At these toughness levels, fracture can occur at or below yield strength loadings for relatively small flaw sizes. For this class of materials, linear-elastic fracture mechanics assessments should be an integral part of design and operational analyses.

- Category II corresponds to intermediate toughness materials with J_{IC} values ranging from 30 to 150 kJ/m^2 (75<K_{IC}<160

MPa√m) and tearing moduli between 10 and 100. Unstable or stable fracture can occur at approximately yield strength loadings for components containing small to medium crack sizes. Fracture control based on a fracture mechanics approach should be considered, especially for materials with relatively high stress intensity limits. Elastic-plastic fracture mechanics methods may be required, particularly at the higher toughness levels.

- Category III corresponds to high toughness materials with J_{IC} values greater than 150 kJ/m^2 and tearing moduli greater than 100. Fracture typically involves stable tearing at stresses well above the yield strength; tearing instabilities are unlikely except after gross plastic deformation. Engineering fracture mechanics evaluations are generally not required because conventional stress limits are adequate for guarding against ductile fracture.

Although the low strength nickel-based alloys discussed herein are inherently ductile and exhibit Category III behavior, exposure to low temperature water negates their high toughness and renders these materials susceptible to Category I / II nonductile fracture.

Materials And Experimental Procedure

The wrought materials used in this program include 76 mm and 64 mm diameter bars of Alloy 690 (Heat "A"--9486-3 and Heat "B"--9518-1) and a 50.8 mm thick plate of Alloy 600 (Heat "A"--NX5853G) in the as-received condition. Limited testing was also performed on cold worked Alloy 600 Heats "A" (10% and 16% cold work) and "B"--NX4650G (14% cold work). EN82H and EN52 welds were fabricated by three manufacturers (denoted by the letters "A", "B" and "C" in the weld identification) using a manual gas-tungsten-arc (GTA) process. Chemical compositions and mechanical properties for the test materials, as well as welding parameters for the welds, are provided in References 8 and 9.

Fracture tests were performed on deeply precracked compact tension (CT) specimens with a width (W) of 30.5 mm and a thickness (B) of 15.2 mm. All specimens except a few Alloy 600 specimens contained 20% side grooves to increase constraint and minimize crack tunneling. Weld specimens were tested in the as-fabricated conditions with either a longitudinal orientation, where the notch was parallel to the welding direction, or transverse orientation, where the notch was normal to the welding direction.

Specimens were tested in air and water under displacement control conditions. Testing in water was conducted at low loading rates, 4 MPa√m/h at 54°-149°C and 0.4 to 2 MPa√m/h at 338°C, to assure sufficient time to produce environmental cracking. Loading rates for a few 54°C water tests were varied between 0.4 and 26,000 MPa√m/h to characterize loading rate effects. The water environment had a room temperature pH of 10.1 to 10.3, an oxygen concentration between 3 and 17 ppb and a nominal hydrogen concentration of 15, 50 or 150 cc H_2/kg H_2O.

Multiple-specimen heat-tint and single-specimen normalization J-R curve test procedures[8,11] were used to establish J_{IC} and tearing modulus values. Specifically, J-R curves were constructed by plotting J values against corresponding crack extension values (Δa) and fitting the J-Δa data with a power-law regression line. The J_{IC} toughness corresponds to the value of J at the intersection of the power-law curve with the 0.2 mm offset blunting line for high strain hardening materials:[12]

$$\Delta a = \frac{J}{4\sigma_f} + 0.2 \quad (mm) \tag{1}$$

where σ_f is the flow strength, which is equal to the average of the yield and ultimate strength levels [i.e., $\sigma_f = (\sigma_{YS} + \sigma_{UTS})/2$]. Values of the tearing modulus (T) were calculated from the following equation:[13]

$$T = \frac{dJ}{da}\frac{E}{\sigma_f^2} \tag{2}$$

where dJ/da is the average slope of the J-R curve (at $\Delta a \approx 1.3$ mm) and E is the elastic modulus. For specimens that exhibit relatively little plasticity prior to the onset of environmental cracking, equivalent critical stress intensity factors (K_{JC}) were computed from experimental J_{IC} values using the equation:[14]

$$K_{JC} = \sqrt{EJ_{IC}} \tag{3}$$

Hydrogen-precharged CT specimens were also tested in 24°C air and hydrogenated water at a nominal stress intensity rate of 3 MPa√m/h. The resulting maximum stress intensity factors (K_{Pmax}), computed by substituting the maximum load and initial crack length into the K solution in ASTM E1737-96, were compared with those obtained for non-precharged specimens tested in water to determine if hydrogen precharging could reproduce the embrittlement observed in low temperature water.

Broken specimen halves were examined on an SEM to characterize the fracture surface morphology to compare operative cracking mechanisms for hydrogen-precharged and non-precharged specimens.

Results

Fracture Toughness

Values of J_{IC} and T for EN82H welds, EN52 welds, Alloy 690 and Alloy 600 tested in 54° to 338°C air and water are summarized in Figures 1 through 7. Most of the environmental testing was performed in 150 cc H_2/kg H_2O, but limited testing was also performed in 15 and 50 cc H_2/kg H_2O. The overall cracking behavior for these materials is discussed below:

- All test materials exhibit exceptional fracture resistance in air and 338°C water [Figures 1-3, 5 and 7]. For the wrought metals, J_{IC} ranges from 358 to 520 kJ/m^2 and T ranges from 242 to 488. For the welds, J_{IC} ranges from 350 to 1060 kJ/m^2 and T ranges from 245 to 540. These exceptionally high toughness values demonstrate that this class of alloys exhibits Category III behavior so fracture control is not a concern.

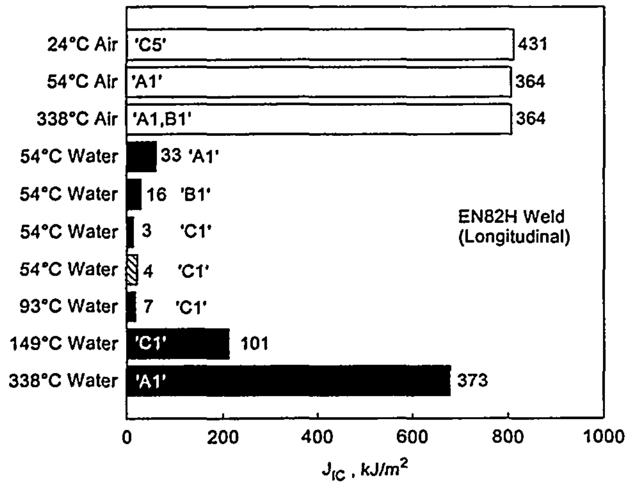

Figure 1. J_{IC} response for longitudinal-oriented EN82H welds "A1", "B1", "C1" and "C5". Values of T are provided beyond each bar. Solid and hatched bars denote tests conducted in water with 150 and 15 cc H_2/kg H_2O, respectively.

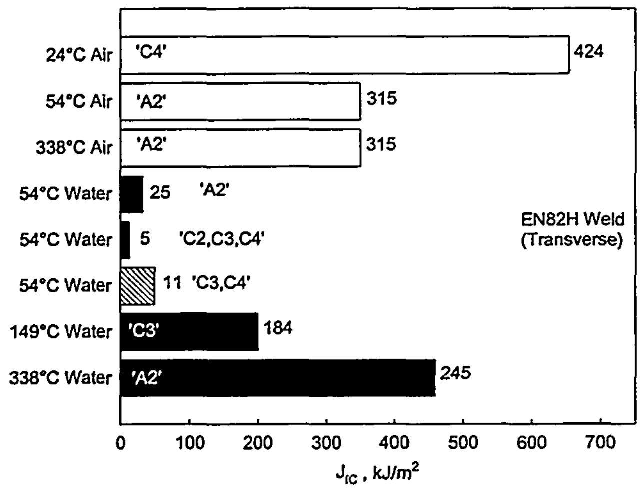

Figure 2. J_{IC} response for transverse-oriented EN82H welds "A2" and "C2"-"C4". Values of T are provided beyond each bar. Solid and hatched bars denote tests conducted in water with 150 and 15 cc H_2/kg H_2O, respectively.

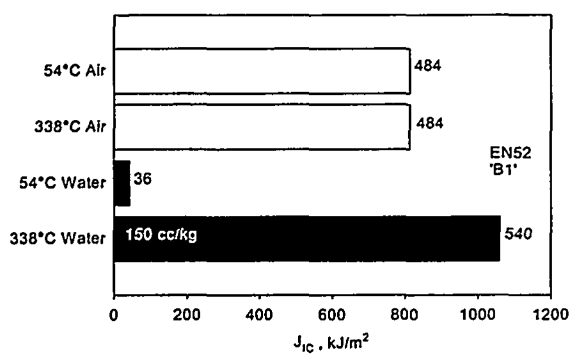

Figure 3. J_{IC} response for EN52 weld "B1". Values of T are provided beyond each bar. The hydrogen content of the water was 150 cc H_2/kg H_2O.

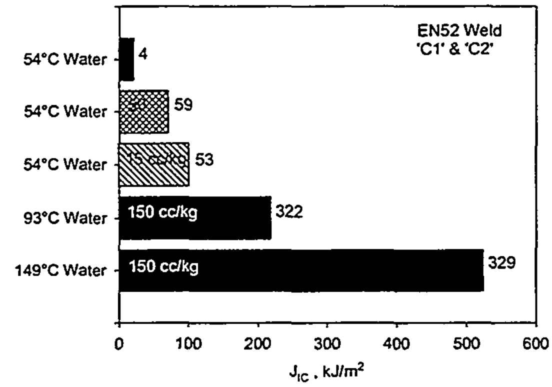

Figure 4. J_{IC} response for EN52 welds "C1" and "C2". Values of T are provided beyond each bar. Solid, cross-hatched and hatched bars denote tests conducted in water with 150, 50 and 15 cc H_2/kg H_2O, respectively.

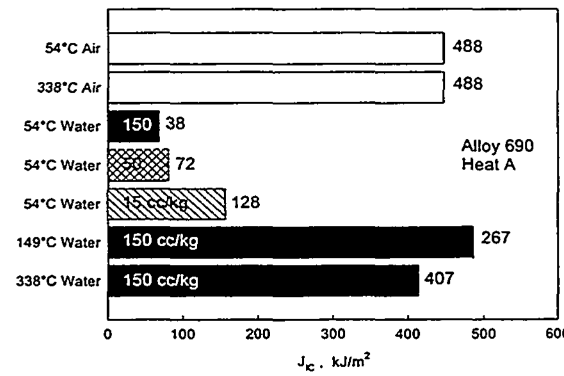

Figure 5. J_{IC} response for Alloy 690 Heat "A". Values of T are provided beyond each bar. Solid, cross-hatched and hatched bars denote tests conducted in water with 150, 50 and 15 cc H_2/kg H_2O, respectively.

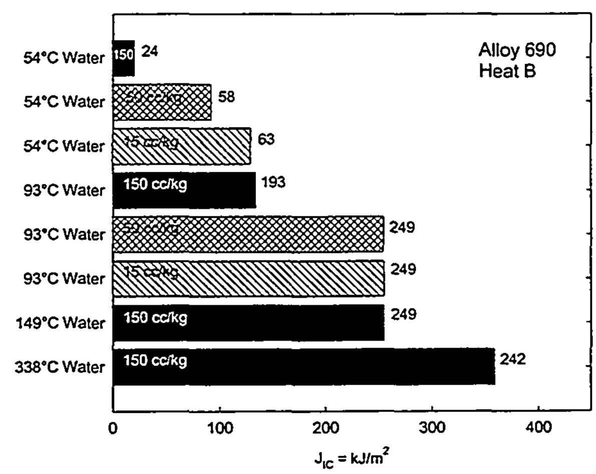

Figure 6. J_{IC} response for Alloy 690 Heat "B". Values of T are provided beyond each bar. Solid, cross-hatched and hatched bars denote tests conducted in water with 150, 50 and 15 cc H_2/kg H_2O, respectively.

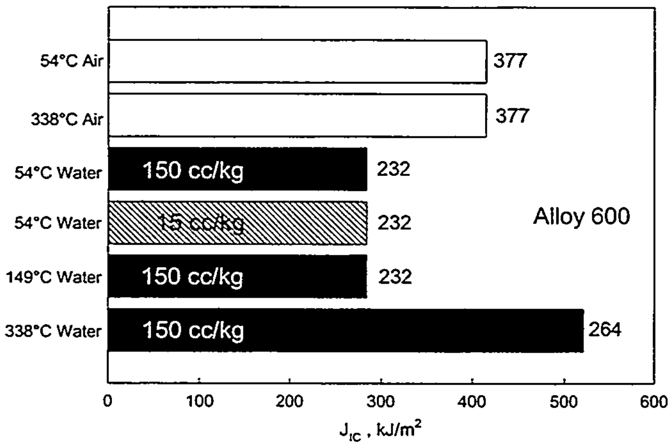

Figure 7. J_{IC} response for Alloy 600 Heat "A". Values of T are provided beyond each bar. Solid and hatched bars denote tests conducted in water with 150 and 15 cc H_2/kg H_2O, respectively.

- EN82H, EN52 and Alloy 690 experience a dramatic degradation in cracking resistance in 54°C water with 150 cc H_2/kg H_2O. J_{IC} and T values are at least an order of magnitude lower than their air counterparts [Figures 1, 2, 3 and 5]. The welds exhibit a Category I / II response so low temperature crack propagation (LTCP) is a potential concern. Alloy 690 exhibits a Category II response with some plastic deformation preceding the onset of environmental tearing. The modest tearing moduli (24-38) in low temperature water indicate that Alloy 690 possesses modest resistance to LTCP so fracture is not a primary design concern.

- For the most embrittled welds, K_{JC} values computed from experimental J_{IC} values agree with K_{Pmax} values. This agreement suggests that cracking initiates near maximum load under nearly linear-elastic conditions. As a result, lower-bound K_{Pmax} values can be used to assess LTCP resistance for components. Lower-bound K_{Pmax} values for EN82H and EN52 are 40 and 53 MPa√m, respectively.

- The dramatic toughness degradation in 54°C water is attributed to a fracture mechanism transition from ductile dimple rupture to intergranular cracking with limited evidence of crystallographic facets.[8,9]

- Alloy 600 does not appear to be susceptible to LTCP [Figure 7]. A Category III response is observed in 54°C hydrogenated water as J_{IC} and T values are reduced by only 30%; hence, sufficient cracking resistance is retained to preclude fracture design concerns. Moreover, the operative cracking mechanism in air and water is ductile dimple rupture, regardless of test temperature. The superior low temperature cracking performance of Alloy 600 was reinforced by conducting tests on Alloy 600 Heats "A" and "B" after introducing 10% to 16% cold work. The cold work failed to induce intergranular LTCP in 54°C water, even though it severely degraded high temperature SCC resistance.[15]

Effect of Hydrogen Pre-Charging

Comparison of the cracking behavior for hydrogen-precharged specimens tested in air with results for non-precharged specimens tested in low temperature water show that a hydrogen embrittlement mechanism is responsible for the LTCP phenomenon.

- Figure 8 shows that the degree of embrittlement exhibited by the welds and Alloy 690 in 54°C water is reproduced in hydrogen-precharged specimens tested in air. Moreover, the intergranular cracking morphology observed in 54°C water is reproduced in hydrogen-precharged specimens tested in air [Figure 9]. These findings demonstrate that intergranular LTCP is associated with hydrogen embrittlement of grain boundaries ahead of a crack tip.

- Testing of hydrogen-precharged specimens indicates that the critical local hydrogen content ahead of a crack, due to hydrogen enrichment in the peak triaxial stress region, is on the order of 120 to 150 ppm.[8,9] This is slightly higher than the 50 to 100 ppm level required to cause LTCP in Alloy X-750,[16] indicating that X-750 grain boundaries are more susceptible to hydrogen embrittlement.

- Figure 8 shows that the lowest cracking resistance is observed when hydrogen-precharged specimens are tested in water. This indicates that hydrogen from the water is added to hydrogen already in the metal to further reduce cracking resistance. It is also noted that the presence of hydrogen in the water minimizes the loss of precharged hydrogen from the crack tip region.

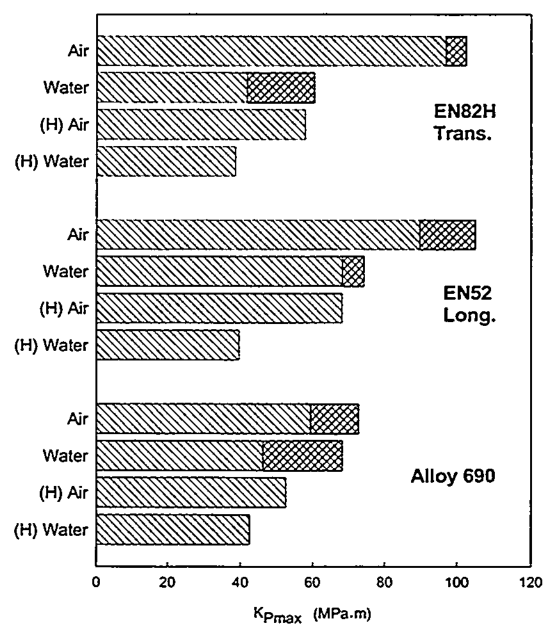

Figure 8. K_{Pmax} values for EN82H, EN52 and Alloy 690. Non-precharged specimens were tested in 54°C air or water; hydrogen-precharged [H] specimens were tested in 24°C air or water. Narrow cross-hatching represents a range of K_{Pmax} values.

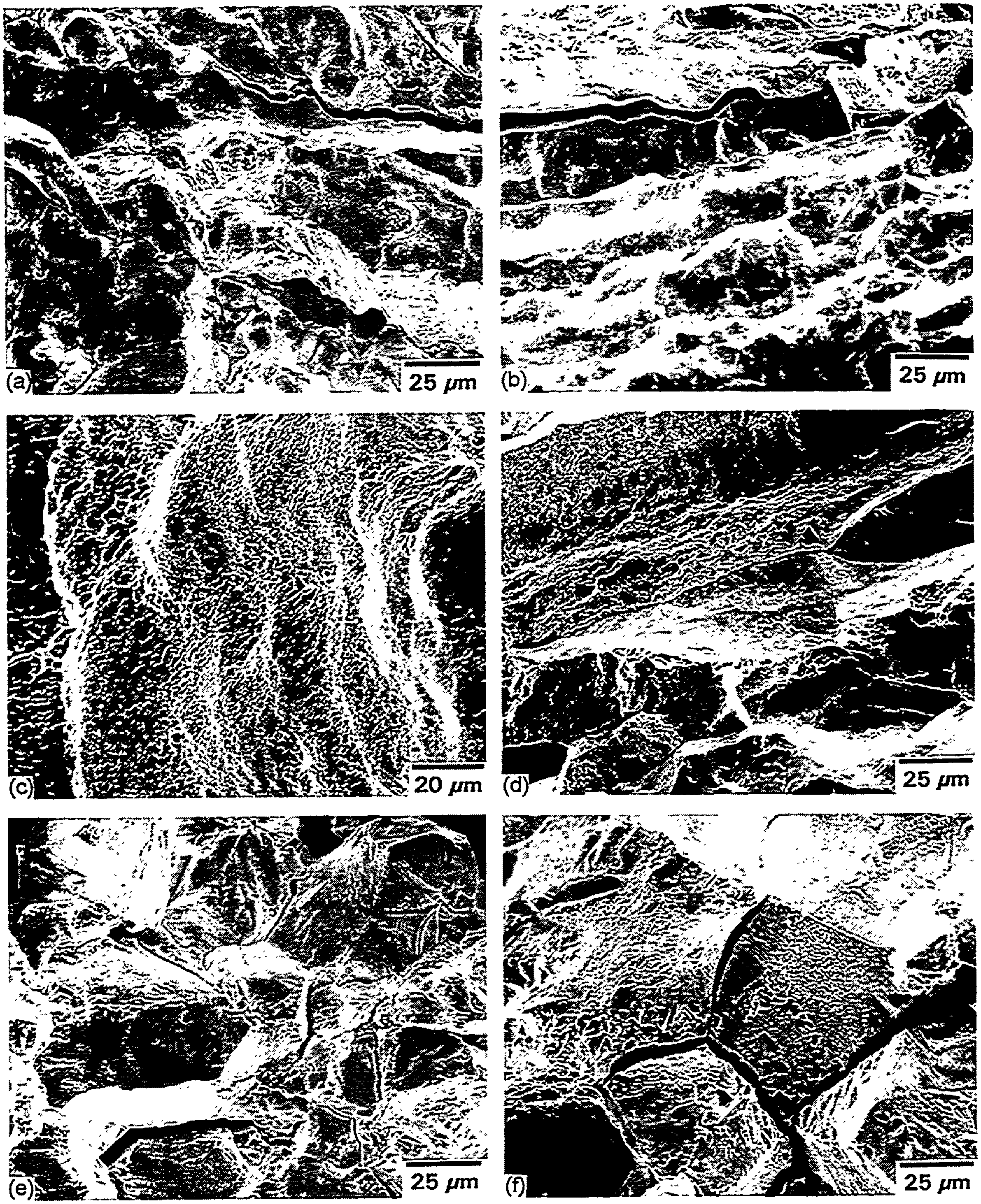

Figure 9. SEM fractographs showing representative intergranular cracking for: (a) Non-precharged EN82H tested in 54°C water. (b) Hydrogen-precharged EN82H tested in air. (c) Non-precharged EN52 tested in 54°C water. (d) Hydrogen-precharged EN52 tested in air. (e) Non-precharged Alloy 690 tested in 54°C water. (f) Hydrogen-precharged Alloy 690 tested in air.

Effect of Displacement Rate

The effect of displacement rate on the LTCP performance of Alloy 690, EN82H welds and EN52 welds was studied to understand the time dependency of the hydrogen embrittlement process. Specifically, tests were performed at increasing displacement rates to determine the conditions where insufficient time was available to embrittle the crack tip region. Displacement rate effects on LTCP, which are shown in Figure 10, are summarized below.

- LTCP properties are insensitive to displacement rate below 1.3 mm/h (<100 MPa√m/hr).[8,9]
- At 15 mm/h (1300 MPa√m/h), LTCP resistance improves significantly for the welds, but not Alloy 690. Based on these findings, LTCP is not an issue for welded components subjected to rapid transients where loading rates exceed ~1000 MPa√m/h.
- A full recovery in cracking resistance for all materials occurs at 305 mm/h (26,000 MPa√m/h) because there is insufficient time to embrittle grain boundaries ahead of a crack.

Effect of Temperature and Hydrogen Content

Extensive testing was performed at various temperatures and dissolved hydrogen levels to establish the conditions under which LTCP is a concern. These findings show that EN82H welds are susceptible to LTCP over a greater range of temperatures and hydrogen levels than either EN52 welds or Alloy 690. Specific findings are summarized below

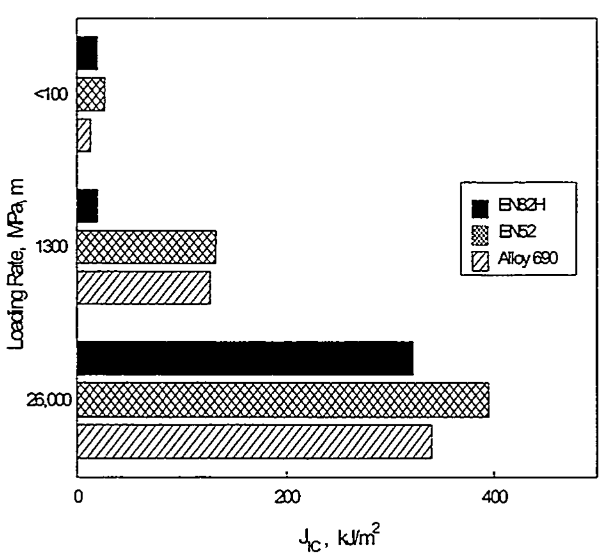

Figure 10. Effect of displacement rate on the fracture toughness of EN82H, EN52 and Alloy 690 in 54°C water. Values of T are given beyond each bar.

EN82H:

- LTCP behavior is relatively insensitive to dissolved hydrogen content between 15 and 150 cc H_2/kg H_2O. For the EN82H longitudinal weld [Figure 1], degassing has little effect on cracking susceptibility as a Category I response is observed at both hydrogen levels (i.e., decreasing hydrogen content from 150 to 15 cc H_2/kg H_2O causes J_{IC} to increase from 14 to 23 kJ/m^2 and T increases from 3 to 4). A slightly greater effect is observed for the transverse weld (Figure 2), as degassing produces a transition from Category I behavior (J_{IC} = 13 kJ/m^2 and T = 5) to Category I / II behavior (J_{IC} = 50 kJ/m^2 and T = 11).
- Increasing the water temperature from 54° to 93°C has little effect on LTCP susceptibility [Figure 1]. At 93°C, EN82H welds continue to show a Category I response with a J_{IC} of 19 kJ/m^2 and T of 7.
- Figures 1 and 2 show that a significant recovery in cracking resistance occurs at 149°C. The Category III behavior observed at 149° and 338°C demonstrates that fracture control is not an important issue in this temperature regime.

EN52 and Alloy 690:

- Decreasing the hydrogen content of water produces a significant recovery in cracking resistance for EN52 welds [Figure 4] and Alloy 690 [Figures 5 and 6]. In fact, degassing to 15 cc H_2/kg H_2O produces a Category II / III response for both materials. The high toughness values in low hydrogen water show that LTCP is not an issue under degassed conditions.
- Figures 4 and 6 show that increasing the water temperature from 54° to 93°C produces a dramatic increase in cracking resistance. Both Alloy 690 and EN52 welds exhibit Category III behavior at and above 93°C, even in water with 150 cc H_2/kg H_2O.

Effect of Crack Geometry

The cracking response of EN82H specimens containing natural weld defects and as-machined notches was characterized to determine if these features are susceptible to LTCP. Specific findings are summarized below.

- In 54°C high hydrogen water (150 cc H_2/kg H_2O), specimens with weld root defects displayed the same degree of embrittlement as fatigue precracked specimens.
- Intergranular LTCP does not initiate from as-machined notches because the stress state is less severe and the diffusion distance to the peak stress location is greater than those for a crack. Since the diffusion distance ahead of a notch is about five times that for a crack,[17] a relatively small fraction of the hydrogen from the water reaches the peak stress location due to hydrogen loss to the surrounding matrix. However, once a ductile tear initiates near maximum load, subsequent cracking in 54°C water occurs by an intergranular mechanism, as shown in Figure 11. Apparently, the tear acts as a crack and increases local stresses and hydrogen concentrations, which induce intergranular LTCP.

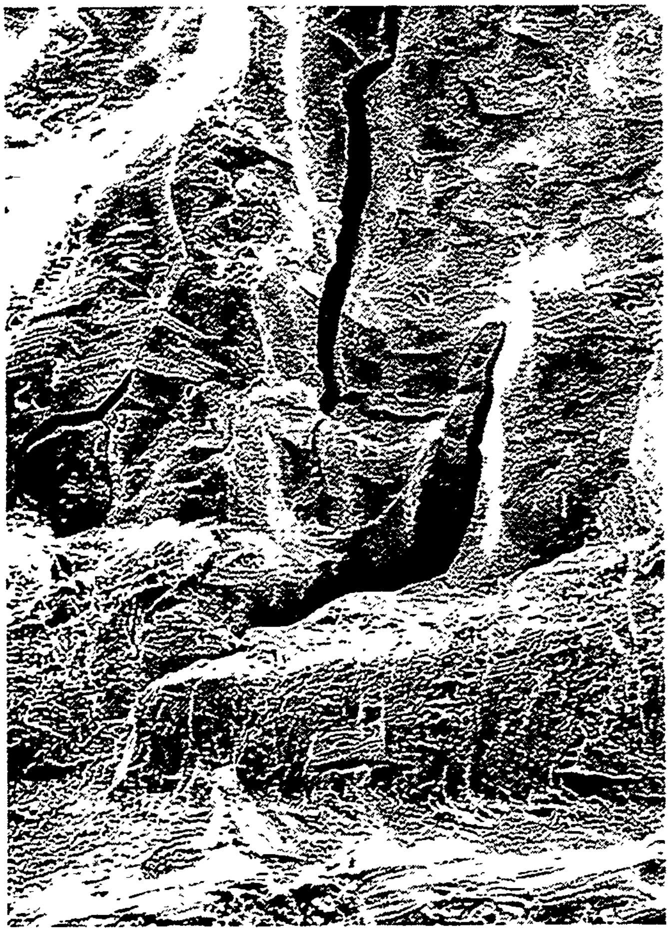

Figure 11. SEM fractograph of as-notched EN82H specimen tested in 54°C water showing ductile tearing between the base of the notch (bottom) and intergranular cracking region (upper half).

Discussion And Conclusions

Low strength nickel-based alloys display exceptional fracture resistance in air and elevated temperature water, so fracture control is not an engineering concern under these conditions. In low temperature water, however, susceptibility to LTCP at K levels as low as 40 MPa√m for EN82H and 53 MPa√m for EN52 negates the inherent high cracking resistance of these welds. The dramatic loss of toughness is due to a fracture mechanism transition from ductile dimple rupture to hydrogen-induced intergranular cracking.

Although the cracking resistance of Alloy 690 is degraded in low temperature water, modest toughness values are retained as some plasticity precedes environmental cracking. As a result, LTCP is not a primary design concern for Alloy 690. The fracture toughness of Alloy 600 is exceptionally high in low temperature water, so LTCP in Alloy 600 is not an issue.

Decreasing the hydrogen content of the water to 15 cc H_2/kg H_2O produces a substantial increase in cracking resistance for EN52 welds and Alloy 690. At temperatures above 93°C, EN52 welds and Alloy 690 are immune to LTCP, even in water with 150 cc H_2/kg H_2O. Therefore, LTCP is not an issue for these materials under degassed conditions or when the water temperature is above 93°C.

Degassing produces a smaller benefit for EN82H welds. Although lower hydrogen contents cause a slight increase in toughness, the welds continue to display a rather brittle Category I or I / II response in water with 15 cc H_2/kg H_2O. In addition, a water temperature of 149°C is required to turn off the LTCP phenomenon. Hence, LTCP is a potential concern for EN82H welds at temperatures below 149°C, even under degassed conditions. Degassing is still worthwhile, however, because it provides some benefit.

Low temperature cracking does not initiate at notches or smooth surfaces; hence, the failure scenario for materials that are susceptible to LTCP involves subcritical crack initiation and growth by either fatigue or high temperature SCC. As the crack grows, K levels increase until a critical crack depth is reached at which point LTCP can occur (i.e., when the applied K exceeds K_{Pmax} or K_{JC}) when the component is exposed to low temperature water.

Acknowledgement

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