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Hydrogen-Assisted Fracture of a Cr-Mo Steel for High-Pressure Gas Containment

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Abstract

The proposed hydrogen energy infrastructure has motivated the need for fracture mechanics data of structural metals in high-pressure hydrogen gas. The objective of this study was to measure the threshold stress-intensity factor (K_{TH}) for the Cr-Mo steel SA 372 Gr. J in 100 MPa hydrogen gas. The K_{TH} measured using wedge-opening-load specimens was nearly 100 MPa·m$^{1/2}$. This threshold is greater than values reported previously for low-alloy steels in high-pressure hydrogen gas and is comparable to the threshold for A-286 stainless steel. These results suggest that SA 372 Gr. J could be an attractive structural metal for components used to contain high-pressure hydrogen gas.

Introduction

Hydrogen is well known to degrade the mechanical properties of structural metals, including steels used for high-pressure hydrogen gas containment. Although low-alloy steels can exhibit significant susceptibility to hydrogen-assisted fracture [1], these structural metals have been used to contain hydrogen gas at pressures up to 45 MPa [2]. Despite the successful service of low-alloy steels in high-pressure hydrogen gas, these materials must be used judiciously since hydrogen-assisted fracture can be sensitive to steel strength, alloy composition, and gas pressure [1, 3].

The proposed hydrogen energy infrastructure could require the storage and transport of hydrogen gas at pressures up to 100 MPa. Low-alloy steels are attractive materials for structures such as pressure vessels, since these materials are low cost and their mechanical properties can be widely varied through heat treatment. However, data on the hydrogen-assisted fracture susceptibility of low-alloy steels are limited at high hydrogen gas pressures, particularly fracture-mechanics data such as the threshold stress-intensity factor (K_{TH}) for hydrogen-assisted fracture [1].

The objective of this study was to measure K_{TH} values for the Cr-Mo steel SA 372 Gr. J in high-pressure hydrogen gas. The SA 372 Gr. J steel has been used in seamless pressure vessels for the containment of hydrogen gas at pressures up to 15 MPa [2]. Measurements of K_{TH} were conducted on wedge-opening-load (WOL) specimens exposed to 100 MPa hydrogen gas.

Experimental Procedures

The SA 372 Gr. J steel was obtained as a 38 mm thick curved plate, which was removed from the wall of a seamless pipe. The steel from this quenched-and-tempered pipe had yield and tensile strengths of 715 MPa and 855 MPa, respectively. The composition of the steel is provided in Table 1. The SA 372 Gr. J is a Cr-Mo steel similar to AISI 4147.

Table 1: Composition (wt%) of SA 372 Gr. J steel.

Cr	Mo	Mn	Si	Al	C	P	S	Fe
0.96	0.18	0.92	0.30	0.026	0.48	0.010	0.002	bal.

Wedge-opening-load (WOL) specimens similar to those described in ASTM Standard E1681 [4] were machined from the steel plate. The WOL specimens had a length of 71 mm, a height of 55 mm, and a thickness (B) of 22 mm. The distance from the load line to the back face (i.e., width dimension, W) of the specimen was 57 mm. Sidegrooves reduced the thickness by 15% in the plane of the precrack starter notch. The WOL specimens were oriented so that the precrack starter notch was parallel to the longitudinal axis of the steel pipe.

Each WOL specimen was prepared for testing by first growing a fatigue precrack from the starter notch. Cyclic loading was conducted in air over a constant load range. At the termination of precracking, the crack length-to-width ratio was approximately 0.55 and the maximum stress-intensity factor was about 25 $MPa \cdot m^{1/2}$. The fatigue-precracked specimens were placed in a glove box, which contained flowing argon gas at 0.1 MPa pressure. Oxygen and water vapor levels were controlled in the argon gas; concentrations of these trace gases were approximately 1 ppm and 7 ppm, respectively. Load was applied to each WOL specimen in the glove box using a bolt, which threaded into the specimen and reacted against a custom-designed load cell. Load and front-face crack opening displacement were monitored as the bolt was torqued. The final load and crack-opening displacement were determined based on a prescribed initial stress-intensity factor for each WOL specimen.

While the bolt-loaded WOL specimens were still contained in the glove box, the specimens were sealed in a thick-walled, A-286 stainless steel pressure vessel. Subsequently, the pressure vessel was connected to a gas manifold. The gas manifold was purged of impurity gases by using several sequences of evacuating and backfilling, where helium and then hydrogen were used as the backfill gases. After the gas manifold was purged, the pressure vessel was evacuated and then filled with 99.9999% hydrogen gas to 100 MPa. The pressure vessels resided in a laboratory at ambient temperature.

The load signals from the WOL specimens were monitored while the specimens were exposed to hydrogen gas. Since a constant crack-opening displacement was maintained on the WOL specimens by the loading bolts, the load decreased continuously during hydrogen-assisted crack propagation and approached a constant value as crack propagation arrested. Once the cracks arrested, the specimens were maintained in the hydrogen gas environment for another 75 hours.

After disassembling the pressure vessel, the WOL specimens were unloaded by removing the bolts. The front-face crack opening displacement was monitored as the specimens were unloaded. Each WOL specimen was then loaded in a mechanical test frame until the front-face displacement equaled the value measured during removal of the bolt. The stress-intensity factor at crack arrest, i.e., the threshold stress-intensity factor, K_{TH}, was calculated using two methods: 1) K_{TH} was calculated using the final load measured in the mechanical test frame, and 2) K_{TH} was calculated using the final load inferred from the initial crack-opening displacement coupled with the compliance relationship for the WOL specimen. The initial and final crack lengths were measured directly from the fracture surface of each WOL specimen.

Results and Discussion

Values of K_{TH} were measured from two WOL specimens. The K_{TH} values calculated using the final measured loads were modestly higher than K_{TH} values calculated using the final inferred loads. These K_{TH} values are listed in Table 2 along with the initial stress-intensity factor (K_o) and final crack length-to-width (a/W) ratio for each specimen.

Table 2: Initial stress-intensity factor, final crack length-to-width ratio, and threshold stress-intensity factor for WOL specimens of SA 372 Gr. J steel.

Specimen	K_o (MPa·m$^{1/2}$)	Final a/W	K_{TH} - measured load (MPa·m$^{1/2}$)	K_{TH} - inferred load (MPa·m$^{1/2}$)
1	144	0.81	102	95
2	136	0.77	106	92

The K_{TH} values measured for SA 372 Gr. J steel in 100 MPa hydrogen gas are approximately 100 MPa·m$^{1/2}$. These K_{TH} values are considerably higher than thresholds for similar low-alloy steels in high-pressure hydrogen gas. Figure 1 compares K_{TH} for SA 372 Gr. J to thresholds for the quenched-and-tempered Cr-Mo steels AISI 4145 and AISI 4147 [1]. The alloy composition and yield strength of the AISI 4147 are particularly close to those for SA 372 Gr. J. Figure 1 shows that K_{TH} for SA 372 Gr. J is about two-fold higher than the threshold for AISI 4147 at 100 MPa hydrogen gas pressure. The only known difference between the AISI 4147 and SA 372 Gr. J steels that could affect hydrogen-assisted fracture is the sulfur content. Sulfur can act in concert with hydrogen to promote fracture at grain boundaries in low-alloy steels [5]. The sulfur concentration in the AISI 4147 (0.011 wt% [1]) was greater than the sulfur content in SA 372 Gr. J (0.002 wt%, Table 1). Heat treatment parameters can also affect sulfur segregation, and it is possible that the two steels were heat treated differently.

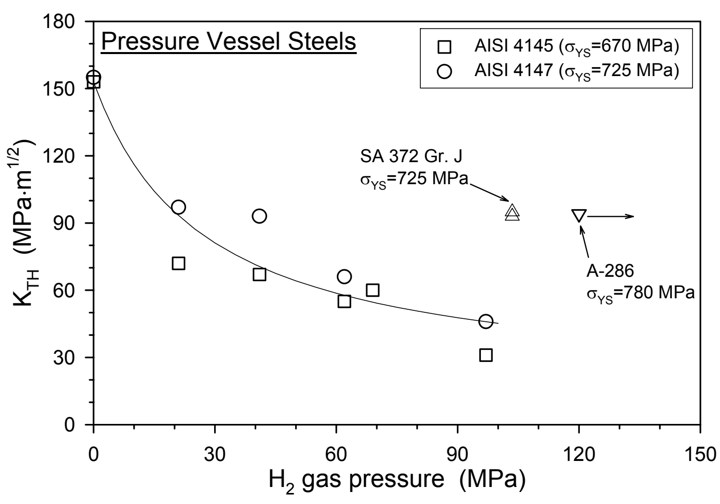

Figure 1: Threshold stress-intensity factor data for SA 372 Gr. J compared to data for low-alloy steels AISI 4145 and AISI 4147 [1] as well as for the stainless steel A-286 [6].

The performance of SA 372 Gr. J in high-pressure hydrogen gas compares favorably with structural metals that are generally considered more resistant to hydrogen-assisted fracture. For example, the stainless steel A-286 is one of the few high-strength alloys considered for high-pressure hydrogen gas service. The K_{TH} for SA 372 Gr. J is similar to the threshold for A-286 measured in 200 MPa hydrogen gas (94 MPa·m$^{1/2}$) [6]. Although the K_{TH} value for A-286 was measured at a much higher gas pressure than the value for SA 372 Gr. J, the threshold for A-286 is not expected to vary in the gas pressure range between 100 and 200 MPa [6]. The K_{TH} for A-286 is plotted in Fig. 1 with data for the low-alloy steels.

The fracture mode of SA 372 Gr. J in high-pressure hydrogen gas is consistent with the high K_{TH}. Figure 2 shows an image of the fracture surface from the region of hydrogen-assisted fracture in SA 372 Gr. J. The fracture path in Figure 2 is transgranular, and virtually no evidence for intergranular cracking was found. Results for hydrogen-assisted fracture in low-alloy steels demonstrate that transgranular fracture is associated with high K_{TH} values [3].

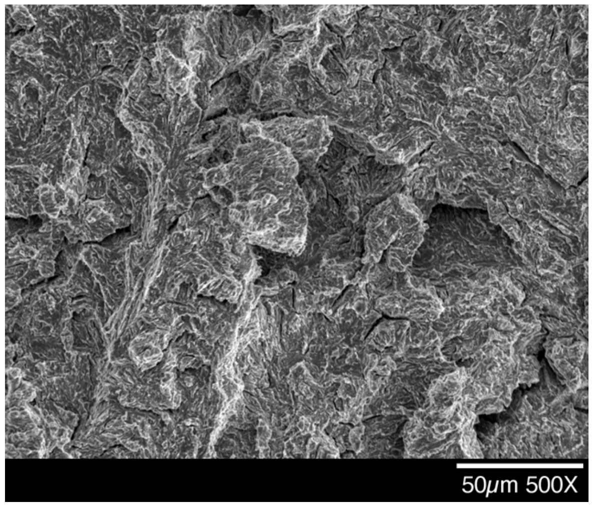

Figure 2. Fracture surface from region of hydrogen-assisted fracture in SA 372 Gr. J.

Although SA 372 Gr. J is clearly more resistant to hydrogen-assisted fracture than other low-alloy steels, the magnitude of K_{TH} may be affected by mechanical conditions in the WOL specimens. Linear elastic and plane-strain conditions in fracture-mechanics specimens are commonly assessed by comparing the non-cracked ligament and thickness to the quantity $2.5(K/\sigma_{YS})^2$ [7]. In this case, when $K=K_{TH}$ the quantity $2.5(K/\sigma_{YS})^2 \approx 47$ mm, which exceeds both the non-cracked ligament and thickness. This suggests that the K_{TH} for SA 372 Gr. J may be lower, since linear elastic and plane-strain conditions may not prevail in the WOL specimens.

Hydrogen-assisted fracture in low-alloy steel WOL specimens typically does not proceed immediately after exposing the specimen to hydrogen gas; rather, an incubation time precedes hydrogen-assisted fracture [1]. Incubation times can be thousands of hours in duration for low-alloy steels [1]. This incubation time has been associated with surface oxides, which serve as a kinetic barrier to hydrogen uptake [1]. The incubation times for the two SA 372 Gr. J specimens were less than 24 hours in duration. Since the SA 372 Gr. J specimens were prepared in a glove box, it is possible that crack-surface oxides ruptured during specimen loading and did not reform prior to hydrogen gas exposure. As a result, hydrogen uptake in SA 372 Gr. J was not significantly impeded.

Conclusions

The threshold stress-intensity factor (K_{TH}) for the low-alloy steel SA 372 Gr. J was measured as approximately 100 MPa·m$^{1/2}$ in 100 MPa hydrogen gas. This threshold is greater than values reported previously for low-alloy steels in high-pressure hydrogen gas and is comparable to the threshold for A-286 stainless steel. Since Cr-Mo steels are relatively low cost, these results suggest that SA 372 Gr. J could be an attractive structural metal for components in the hydrogen energy infrastructure that must contain 100 MPa hydrogen gas.

References

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