Assessing Hydrogen-Assisted Cracking Fracture Modes in High-Strength Steel Weldments

Test results substantiate and extend the Beachem theory on hydrogen embrittlement

BY S. A. GEDEON AND T. W. EAGAR

ABSTRACT. The stress intensity that causes crack propagation in high-strength steel weldments was quantified as a function of the hydrogen content at the crack location. This relationship was used to assess previously proposed theoretical hydrogen-assisted cracking mechanisms. Indeed, it was found that the microplasticity theory of Beachem can best describe how the stress intensity factor and hydrogen content affect the modes of intergranular, quasi-cleavage and microvoid coalescence fracture.

Implant test results were analyzed with the aid of fracture mechanics to determine the stress intensity associated with various modes of fracture. Diffusible weld hydrogen results were analyzed with the aid of a hydrogen distribution model developed by Coe and Chano to determine the amount of hydrogen present at the crack location at the time of fracture.

The stress intensity and hydrogen content responsible for the microvoid coalescence fracture mode have been quantified for the high-strength steel used in this study. The resulting relationship agrees with the results of Beachem but extend his theory to a wider range of hydrogen contents.

Introduction

It is known that hydrogen-assisted cracking is a complex function of the amount of hydrogen, the stress, the temperature and the microstructure of the steel. The purpose of this study is to quantify the amount of hydrogen that causes crack propagation as a function of stress intensity for a specific material and temperature. This relationship is then compared to existing cracking mechanism theories.

Previous literature concerning cracking theories, stress intensity, determination and hydrogen content determination is briefly reviewed.

Hydrogen-Assisted Cracking Mechanisms

The results of theoretical studies of hydrogen embrittlement mechanisms proposed by physical metallurgists have rarely been applied to the field of welding. Sawhill's study (Ref. 1) of HY-130 steel weldments, however, provides a good background for the ensuing analysis of the most often proposed hydrogen embrittlement mechanisms. Even though the problem of hydrogen embrittlement has been studied extensively, no one theory has become generally accepted.

The planar pressure theory, proposed by Zapfe (Ref. 2), is based on the decrease in solubility of hydrogen as the temperature is lowered. The atomic hydrogen is postulated to reassociate into diatomic hydrogen in pores and microvoids. The pressure of diatomic hydrogen then builds to very high values, which adds to the applied external stresses. By applying Sievert's law, it is estimated that a steel with 5 ppm hydrogen would have over 17,000 atmospheres pressure in the voids at 20°C (68°F). However, several experimental observations conflict with this mechanism. Hydrogen embrittlement can be eliminated by degassing even after exposure to room temperature. The low temperature of the degassing would not be high enough to dissociate the diatomic hydrogen into monatomic hydrogen which could diffuse out of the steel. Also, the observation of hydrogen-induced cracks growing on a free surface excludes an internal pressure gradient as the driving force for crack growth.

The adsorption theory of Petch and Stable (Ref. 3) and further modifications (Ref. 4) propose a lowering of the surface free energy by hydrogen so that a crack can grow under a lower applied stress. This theory has been criticized on the basis that the small but finite plastic deformation observed on hydrogen-induced fracture surfaces requires more energy than could be explained by the adsorption theory. In addition, fracture surfaces indicate rapid void formation and coalescence at low temperatures where rate of surface migration would be negligible.

A theory proposed by Troiano (Ref. 5) suggests that hydrogen interacts with dislocation pileups in areas of triaxial stress to lower the cohesive strength. It is known that hydrogen will diffuse toward regions of high triaxial stress such as those associated with a stress riser. When the concentration reaches a given level, the interaction of hydrogen with dislocation array ahead of the stress riser is postulated to be sufficient to cause fracture. Troiano suggests that this interaction is due to the valence electrons from hydrogen atoms entering the unfilled "d" shells of the iron and modifying the repulsive forces which determine the interatomic spacing in transition metals.

KEY WORDS

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Others have modified the planar pressure theory and the adsorption theory by assuming that hydrogen atoms are transported to the void or crack tip as Cottrell atmospheres. Bastin (Ref. 6) has proposed that hydrogen atoms are carried along by the movement of dislocations during plastic deformation. Thus, he reasons, dislocation pileups at structural defects will produce an oversaturation of hydrogen, which will result in an increase in pressure which in turn produces triaxial stresses and embrittlement. Research by Graville (Refs. 7, 8) supports the hypothesis that hydrogen transport by dislocations to the site of crack initiation is a necessary part of the embrittlement process.

Beachem (Ref. 9) has proposed a theory of hydrogen-assisted cracking, based on a microplasticity mechanism rather than embrittlement. He suggests that the hydrogen in the lattice ahead of the crack tip assists whatever microscopic deformation processes the microstructure will allow. Thus, intergranular, quasi-cleavage or microvoid coalescence fracture modes will operate depending on the microstructure, the crack tip stress intensity, and the concentration of hydrogen. The model unifies several theories, but shows that the planar pressure and adsorption theories are unnecessary. He proposes that the basic hydrogen-steel interaction appears to be an easing of dislocation motion or generation, or both.

In all of the above studies, the specimen was charged with hydrogen in order to examine the effect on fracture. However, Bonisewski and Moreton (Ref. 10) have observed that hydrogen introduced by this means will not behave in the same way as hydrogen introduced by an actual welding process.

Quantification of the Stress Intensity in a Weld

Among the various testing methods for assessing hydrogen embrittlement, the implant test has become one of the most popular for scientific investigations of the cracking phenomenon in welds. This is due to the fact that the stress, hydrogen level and microstructure can be independently varied and controlled. Crack susceptibility using this test is typically defined as the lower critical stress (LCS). The LCS is the maximum stress at which fracture does not occur for an arbitrarily long period of time (usually 1 to 3 days).

Fracture mechanics can be used to determine the stress intensity associated with fracture in the implant specimens. Since the helical notch used on the implant specimens is too blunt to use linear elastic fracture mechanics (LEFM), the crack initiation process is difficult to quantify. However, once hydrogen embrittlement occurs, the embrittled region itself will act as a sharp crack tip, and one can use LEFM to investigate the fracture of the remaining area, at least in high-strength welds.

Daoud, et al. (Refs. 11, 12), have determined the stress concentration factor for an edge cracked circular bar in tension, and since modified this to include the effect of the crack geometry. Even though their analysis does not include the effect of a restraining weld close to the crack, it can be used to give an approximation of the $K_{IC}$ of the final fractured area.

The diffusible hydrogen test can be used to determine the amount of hydrogen initially solidified into the weld pool. However, since fracture in the implant specimens will occur sometime after the weld has cooled down, and some hydrogen will have been lost by diffusion, these results must be analyzed to determine the amount of hydrogen remaining in the cracking zone at the instant of fracture.

The amount and distribution of hydrogen remaining in an implant specimen as a function of time after welding can be estimated with the aid of a model initially developed by Coe and Chano (Ref. 13). They used an iterative procedure using small time-temperature increments to calculate the effect of time on the hydrogen distribution. The results are presented as hydrogen as a function of the nondimensional parameter $\theta$. This value is defined as:

$$\theta = \frac{D \theta}{\theta_0}$$

where $D$ is the diffusivity of hydrogen in solid iron, $\theta$ is time, and $\theta_0$ is the weld bead depth. A sample of their distribution plots is shown in Fig. 1, which shows the hydrogen distribution as a function of distance in the weld for various values of $\theta$.

A better plot for the purposes of this research is shown in Fig. 2, which shows the hydrogen concentration as a function of $\theta$ at various weld locations.

It has been postulated that dislocation sweeping will increase the actual amount of hydrogen at the crack tip. The increased solubility of hydrogen under an applied axial tensile stress has been estimated to be 5 times higher than the nominal solubility by Louthan, et al. (Ref. 14).

Anderson (Ref. 15) used a finite element technique to estimate that the hydrogen in front of the crack tip is about 1.2 times the nominal bulk hydrogen value. Schulte and Adler (Ref. 16), using nuclear reaction analysis of deuterium distribution, determined that the maximum hydrogen will be about 1.4 times the nominal bulk hydrogen concentration.

Experimental Procedure

Although no standard procedure exists for the implant method (Ref. 17), the IW published a document (Ref. 18) containing guidelines for performing this test. These procedures were followed using the 165 notch geometry and a helical notch.
A loading time of 5 min was chosen based on research by Peng (Ref. 19), who showed that variations in loading time from 2 to 7 min after welding did not affect the lower critical stress (LCS). A 24-hour loading time was also chosen so that the hydrogen distribution model of Coe and Chano (Ref. 13) could be used to determine the amount of hydrogen in the cracking zone.

The material studied in this investigation is a high-strength steel conforming to MIL-A-46100C (Ref. 20). Its main use is for armor in military applications and it is the main structural steel used in the M1 tank. Chemical composition requirements in MIL-A-46100C are very broad as the main performance criteria are good hardenability and ballistic integrity. Due to its extremely high hardness, this material is very susceptible to hydrogen embrittlement.

The composition of the 46100 used throughout this investigation is listed in Table 1. It is composed primarily of tempered martensite with some banding. Due to the high hardness (53 RC), this steel had to be normalized to 35 RC in order to be machined into implant specimens. The specimens were then austenitized in vacuum, quenched in oil, and tempered in air to their original condition. The implant specimens were machined longitudinal to the rolling direction.

Diffusible hydrogen testing was performed in accordance with AWS A4.3-86. The gas chromatography method was used with a Yanaco hydrogen analyzer model G-1006.

By varying the amount of hydrogen and oxygen in argon GMAW shielding gas, time to loading, and preheat temperature, amount of hydrogen in the weldment at the time of cracking can be varied. A matrix of seven conditions was studied for each shielding gas composition:

1) Diffusible weld hydrogen content (as per AWS A4.3-86).
2) Hydrogen remaining 24 h after welding (the AWS specimen was allowed to cool for 24 h before being analyzed).
3) Hydrogen remaining 24 h after welding with preheat.
4) LCS when loaded 5 min after welding.
5) LCS when loaded 24 h after welding.
6) LCS when loaded 5 min after welding with preheat.
7) LCS when loaded 24 h after welding with preheat.

Seven different shielding gas compositions were studied although not every gas was evaluated both with and without preheat.

A scanning electron microscope (SEM) was used to examine the fractured surfaces of the implant specimens. The initial fracture in the majority of specimens was due to hydrogen embrittlement, with the remaining area failing due to microvoid coalescence. Quantitative fractography was performed to map the various failure zones across the failed surfaces of the implant specimens, and the crack geometries were compared with those modeled by Daoud, et al. Other details of the experimental procedures, materials used, weld parameters and results have been published elsewhere (Ref. 21).

**Experimental Results**

Table 2 summarizes the experimental results acquired in this portion of the research program. Each LCS value was determined from a plot of time-to-fracture vs. applied stress. Two such plots show the effect of preheat (Fig. 3) and hydrogen in the shielding gas (Fig. 4). In the majority of specimens, fracture occurred in the fusion zone.

The ratio $Q/Q_0$ in Fig. 2 is approximately equal to the ratio of the hydrogen content at 24 h divided by the initial hydrogen content. Using the data from Table 2, this ratio averages 0.25 for welds made without preheat. From Fig. 2, this...
corresponds to a $\Theta$ of 0.9, which is very close to the value of 1.0 found if $\Theta$ is calculated directly from the cooling curve and diffusivity vs. temperature data.

Based on the experimentally determined $\Theta$ value of 0.9, the amount of hydrogen located at the fusion line ($l = l_0 = 0.3$ in Fig. 2) will be equal to 12.5% of the initial hydrogen content in the weld. For welds made with 250°F ($121^\circ$C) preheat, $Q/Q_0$ averages 0.13, 24 h after welding, which corresponds to a $\Theta$ of 1.9.

In this way, the amount of hydrogen at the cracking zone during the final fracture can be found for each of the implant specimens. This amount of hydrogen is termed the bulk hydrogen in the cracking zone and does not include any increased amount which may be due to the increased stress state at the crack tip.

In order to determine the stress intensity that caused cracking, a fractographic analysis of the fractured implant specimens was performed. The cracking zone in all of the implant fractures studied was at the weld fusion line.

Figure 5 shows a typical overall view of the fractured surface of an implant specimen. Figure 6 shows the location of each of the magnified photos taken of this surface. Figure 7 shows the fracture morphology typical of hydrogen embrittlement as evidenced by intergranular faceting. The fracture morphology associated with microvoid coalescence is shown in Fig. 8 as evidenced by the ductile dimples. The transition region showing areas of intergranular faceting below or next to areas of microvoid coalescence is shown in Fig. 9. The resulting quantitative fracture map developed for this specimen is depicted in Fig. 10.

Not all of the specimens exhibited such a clear distinction between the different fracture zones. For example, a number of...
the low-hydrogen samples had areas of fisheyes. A fisheye is an inclusion that is locally surrounded by an area of hydrogen embrittlement. The local area of hydrogen embrittlement surrounding a fisheye is presumed to be due to hydrogen trapping at the inclusion. Numerous investigators (Refs. 22-24) have found that hydrogen can be trapped at inclusions.

Of 140 fractured implant specimens, only 60 had cracks starting from one edge. Of the fracture maps developed for these 60 specimens, the 12 that very closely approximated the crack geometry studied by Daoud, et al. (i.e., circular-arc edge crack), were used to determine the fracture toughness (as estimated by $K_{IC}$) of the area which fractured due to microvoid coalescence. Table 3 gives a presentation of the results for these 12 specimens. The hydrogen values shown in Table 3 were determined by estimating $r$ from the cooling curve the time at which fracture occurred, and finding the corresponding hydrogen concentration from Fig. 2. The $a/D$ ratio corresponds to the region of intergranular fracture, which was assumed to approximate a crack.

The resulting plot of stress intensity vs. amount of hydrogen in the cracking zone is shown in Fig. 11. As can be seen, the stress intensity (an approximation of $K_{IC}$ at which microvoid coalescence occurs) decreases with increasing hydrogen in the crack zone. However, at very high hydrogen contents, intergranular fracture will
be more energetically favorable than microvoid coalescence until very high stress intensities are reached.

**Discussion**

The implant specimens that were welded with a high-hydrogen content in the shielding gas (2% H₂), had a \( K_{IC} \) of about 16 MPa m\(^{1/2} \) (15 kips in.\(^{1/2} \)). This value agrees quite closely with the work of Herman and Campbell (Ref. 25), who used fracture toughness samples of this identical type of steel and determined the stress corrosion cracking toughness \( K_{SCC} \). Herman and Campbell found that the fracture toughness of this material was 16 MPa m\(^{1/2} \) (15 kips in.\(^{1/2} \)) when exposed to distilled water.

At low hydrogen levels (0% H₂ added to the shielding gas), the final fractures had a toughness of approximately 71 MPa m\(^{1/2} \) (65 kips in.\(^{1/2} \)) when exposed to distilled water. In a few cases, very small amounts of hydrogen seemed to increase the \( K_{IC} \) of microvoid coalescence fracture above the \( K_{IC} \) of hydrogen-free specimens. Hydrogen-induced strengthening has been documented by others (Ref. 14). White (Ref. 26) also noticed some slight strengthening of her implant specimens when welding with 0.05% hydrogen in the shielding gas. This phenomena may be due to hydrogen pinning the dislocations.

There were a number of specimens that had both high hydrogen contents and high \( K_{IC} \) values. These hydrogen values were much higher than in specimens with \( K_{SCC} \) values of 16 MPa m\(^{1/2} \) (15 kips in.\(^{1/2} \)). At higher loads, the value increased to approximately the nominal \( K_{IC} \) value of 71 MPa m\(^{1/2} \) (65 kips in.\(^{1/2} \)). In a few cases, very small amounts of hydrogen seemed to increase the \( K_{IC} \) of microvoid coalescence fracture above the \( K_{IC} \) of hydrogen-free specimens. Hydrogen-induced strengthening has been documented by others (Ref. 14). White (Ref. 26) also noticed some slight strengthening of her implant specimens when welding with 0.05% hydrogen in the shielding gas. This phenomena may be due to hydrogen pinning the dislocations.

One of the main features of the Beacham theory is the classification of fracture modes with respect to stress and hydrogen level. At relatively high stresses, hydrogen-assisted cracking can propagate by microvoid coalescence, which is normally thought of as a ductile failure mechanism. Beacham proved that hydrogen can be responsible for microvoid coalescence by partially fracturing a sample in hydrogen, then freezing the sample in liquid nitrogen and sectioning the sample to find evidence of the processes occurring ahead of the crack tip. As the stress intensity decreases, crack propagation proceeds by the lower plastic deformation processes of quasi-cleavage, and finally, intergranular separation. Increasing hydrogen concentration at the crack tip has the effect of decreasing the stress intensity at which these fracture processes occur.

Beachem's model adequately explains the presence of plastic deformation preceding hydrogen cracking in the HAZ of welds (Ref. 27) and plastic deformation in other systems as well (Refs. 28-30). Also, the qualitative experimental results postulated by Beachem in Fig. 12 bear a remarkable resemblance to the quantitative results of the present investigation.

The major difference between the fracture map proposed by Beachem and the one found in the present investigation, is...
that this investigation shows that intergranular failure can still occur at much higher values of hydrogen. It makes sense that intergranular failure will occur at high-hydrogen contents, but Beachem suggests that microvoid coalescence will occur faster, and thus predominate. The three points at high hydrogen concentrations are beyond the range investigated by Beachem. Thus, Fig. 11 shows a modification to the original work by Beachem, namely, that high hydrogen concentrations can suppress the microvoid coalescence fracture mode, and that intergranular fracture will still be operative. The Beachem model appears to be the most comprehensive model to date, and accounts for most experimental observations of hydrogen cracking.

The present investigation did not quantify the hydrogen concentration that caused intergranular or quasi-cleavage fracture. There were not enough specimens that exhibited the proper amount of quasi-cleavage fracture along with a crack geometry which approximated the fracture mechanical analysis of Daoud, et al.

An attempt was made to quantify the relationship between stress intensity and hydrogen content for which no hydrogen-assisted cracks will propagate. In an unfractured specimen, it is assumed that a very small crack exists for which a/D is less than 0.2. From Daoud, et al., the stress intensity factor will be approximately unity. Here, the maximum hydrogen present in the lower critical stress specimens can be used along with the applied stress (the LCS value) and the assumed a/D ratio to develop the "no hydrogen-assisted cracking" region in Fig. 13. The numbers for this region should be considered to be tentative now since the assumptions are not necessarily justified.

The current research has attempted to quantify both the stress intensity factor and the amount of hydrogen responsible for causing microvoid coalescence. This is the first time this has been attempted. Discrepancies may arise due to the fact that the implant specimens were not well suited to KIC measurements. Another shortcoming may be that the bulk hydrogen in the cracking zone was determined rather than the hydrogen due to dislocation sweeping or stress concentrations at the crack tip. However, the relationships developed in this investigation may be accurate since the increased amount of hydrogen due to stress concentrations may be only a factor of about 1.4.

Hopefully, future research will enable anticipated hydrogen levels to be used to quantify the allowable defect size, which will result in a stress intensity factor lower than that which causes hydrogen-assisted cracking. Thus, very low hydrogen welds can be designed to either allow higher stresses or larger defects than high-hydrogen welds.

## Conclusions

The fracture modes of high-strength steel welds have been characterized as a function of the stress intensity and hydrogen content at the cracking zone in implant tested welds. The results indicate that the hydrogen embrittlement theory originally proposed by Beachem can be used to explain the effect of hydrogen on cracking of high-strength steels. The results of the present study increase the range of hydrogen above that used in the original Beachem study to show that large amounts of hydrogen will increase the propensity for intergranular fracture rather than microvoid coalescence.

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## References


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A Preliminary Evaluation of the Elevated Temperature Behavior of a Bolted Flanged Connection
By J. H. Bickford, K. Hayashi, A. T. Chang and J. R. Winter

This Bulletin consists of four Sections that present a preliminary evaluation of the current knowledge of the elevated temperature behavior of a bolted flanged connection.

Section I—Introduction and Overview, by J. H. Bickford; Section II—Historical Review of a Problem Heat Exchanger, by J. R. Winter; Section III—Development of a Simple Finite Element Model of an Elevated Temperature Bolted Flanged Joint, by K. Hayashi and A. T. Chang; and Section IV—Discussion of the ABACUS Finite Element Analysis Results Relative to In-the-Field Observations and Classical Analysis, by J. R. Winter.

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By A. W. Pense

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