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III.5 Hydrogen Embrittlement of Structural Steels

Brian Somerday
Sandia National Laboratories
P.O. Box 969
Livermore, CA 94550
Phone: (925) 294-3141
Email: bpsomer@sandia.gov

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DOE Manager

Erika Sutherland
Phone: (202) 586-3152
Email: Erika.Sutherland@ee.doe.gov

Project Start Date: January, 2007

Project End Date: Project continuation and direction determined annually by DOE

Overall Objectives

- Demonstrate the reliability/integrity of steel hydrogen pipelines subjected to cyclic pressure operating conditions, emphasizing welded regions
- Identify pathways for reducing the cost of steel hydrogen pipelines without compromising reliability/integrity

Fiscal Year (FY) 2014 Objectives

- Complete triplicate measurements to establish reliable fatigue crack growth relationships for X65 girth weld fusion zone in 3000 psi (21 MPa) hydrogen gas
- Complete duplicate measurements of fatigue crack growth relationships in hydrogen gas for model iron-carbon alloys with two different grain sizes in collaboration with I2CNER

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Delivery section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan (section 3.2.5):

(B) Reliability and Costs of Gaseous Hydrogen Compression
(D) High As-Installed Cost of Pipelines
(K) Safety, Codes and Standards, Permitting

Technical Targets

The principal targets addressed by this project are the following (Table 3.2.4):

- Pipeline Reliability/Integrity

One salient reliability/integrity issue for steel hydrogen pipelines is hydrogen embrittlement. For steel pipelines, the central unresolved issue is the pipeline performance under extensive pressure cycling. One of the objectives of this project is to enable safety assessments of steel hydrogen pipelines subjected to pressure cycling through the use of structural integrity models in design codes, e.g., American Society of Mechanical Engineers (ASME) B31.12. This structural integrity analysis can determine limits on design and operating parameters such as the allowable number of pressure cycles and pipeline wall thickness. Efficiently specifying pipeline dimensions such as wall thickness also affects pipeline cost through the quantity of material required in the design.

FY 2014 Accomplishments

- Reproducible fatigue crack growth rate (da/dN) vs. stress-intensity factor range (ΔK) relationships were measured for X65 weld fusion zones in 3,000 psi (21 MPa) hydrogen gas. Comparing with results from FY 2013 suggests that the weld fusion zone and heat-affected zone are no more susceptible to hydrogen-accelerated fatigue crack growth compared to the base metal.
- Modified specimen geometry ESE(T) used in this study expands fatigue crack growth testing capabilities by allowing thinner walled pipes to be examined as well as different orientations of materials that would otherwise be precluded by employing the conventional C(T) specimen.

Introduction

Carbon-manganese steels are candidates for the structural materials in hydrogen gas pipelines; however, it is well known that these steels are susceptible to hydrogen embrittlement. Decades of research and industrial experience have established that hydrogen embrittlement compromises the structural integrity of steel components. This experience has also helped identify the failure modes that can operate in hydrogen containment structures. As a result, there are tangible ideas for managing hydrogen embrittlement in steels and quantifying safety margins for steel hydrogen containment structures. For example, fatigue crack growth aided by hydrogen embrittlement is a well-established failure mode for steel hydrogen containment structures subjected to pressure cycling. This pressure cycling represents one of the key differences in operating conditions between current hydrogen pipelines and those anticipated in a hydrogen delivery infrastructure. Applying structural integrity models in design codes coupled with measurement of relevant material properties allows quantification of the reliability/integrity of steel hydrogen pipelines subjected to pressure cycling. Furthermore, application of these structural integrity models is aided by the development of physics-based predictive models, which provide important insights such as the effects of microstructure on hydrogen-assisted fatigue crack growth. Successful implementation of these structural integrity and physics-based models enhances confidence in the design codes and enables decisions about materials selection and operating conditions for reliable and efficient steel hydrogen pipelines.

Approach

The approach of this project is to apply the core capability in materials characterization at Sandia to measure the fatigue crack growth rates of technologically relevant pipeline steels in high-pressure hydrogen gas. These properties must be measured for the base materials but more importantly for the welds, which are likely to be most vulnerable to hydrogen embrittlement. Such measurements are necessary for enabling the application of structural integrity models in design codes. For example, the new ASME B31.12 code for hydrogen pipelines includes a fracture mechanics-based integrity management option, which requires material property inputs such as the fatigue crack growth relationship in hydrogen gas.

Following the establishment of reliable fatigue crack growth relationships for pipeline steel base metal, weld heat-affected zone, and weld fusion zone in hydrogen gas, a secondary approach of this project is to apply analytical techniques such as electron microscopy to define the mechanisms of hydrogen embrittlement for the purpose of developing physics-based predictive models. Such predictive models can provide quantitative insight into the effects of environmental, material, and mechanical variables on hydrogen embrittlement. For example, quantifying the effect of microstructure on hydrogen-accelerated fatigue crack growth can aid in the qualification of line pipe steels and their welds for hydrogen service.

Results

The fatigue crack growth rate (da/dN) vs. stress-intensity factor range (ΔK) relationship is a necessary material-property input into structural integrity models applied to steel hydrogen pipelines. One such integrity assessment methodology for steel hydrogen pipelines was recently published in the ASME B31.12 code. The measurement of fatigue crack growth relationships in this task supports the objective of establishing the reliability/integrity of steel hydrogen pipelines by enabling application of the ASME B31.12 code.

Low-strength line pipe steels such as X52, X60, and X65 were selected for this task because of their stakeholder-recognized technological relevance for hydrogen pipelines. Generally, lower-strength steels are selected for hydrogen pipelines since these steels are less susceptible to hydrogen embrittlement. A section of X65 steel pipe containing a girth-oriented gas metal arc weld (GMAW) was provided by an industry partner. An optical-microscope image revealing the base metal (BM), weld fusion zone (FZ), and weld heat-affected zone (HAZ) is shown in Fig. 1. This image demonstrates that the material regions have distinctly different metallurgical structures, particularly the base metal and fusion zone. In FY 2013, the da/dN vs. ΔK relationships for the heat-affected zone (HAZ) in 3,000 psi (21 MPa) hydrogen gas were measured and compared to the base metal. The emphasis for FY 2014 was measurement of the weld fusion zone (FZ) in 3,000 psi (21 MPa) hydrogen gas. As specified in ASME B31.12, the da/dN vs. ΔK relationship was measured following ASTM Standard E647. Since the maximum pressure specified for hydrogen gas pipelines in the ASME B31.12 code is 3,000 psi (21 MPa), this upper-bound pressure was selected for the testing. The load-cycle frequency selected for the testing was 1 Hz, consistent with previous testing on X52 line pipe steel in high-pressure hydrogen gas.

As demonstrated in FY 2013, non-uniform crack fronts were observed in HAZ and FZ compact tension (C(T)) specimens. To mitigate deviation of the crack fronts during testing, C(T) specimens were thinned from their original 0.5 in (13 mm) to 0.25 in (6 mm) for the HAZ and FZ specimens. While this specimen modification improved results for the HAZ C(T) specimens, this thinning did not rectify non-uniform crack advance in the FZ C(T) specimens; therefore, both an alternate specimen geometry, i.e. the eccentrically loaded, single edge notched tension (ESE(T)), and crack propagation direction were adopted for the weld FZ, which improved crack front uniformity. Non-uniform crack fronts in the FZ were attributed to residual stress gradients [1] across the crack front, which prompted the alternate crack propagation orientation in the weld FZ specimens. Figure 2 shows a schematic of the orientations and specimen geometries tested in this study with respect to the welded pipe. The first letter in parenthesis refers to the direction of applied load and the second letter refers to the crack propagation direction. Because of the residual stress gradient, the preferred orientation for the FZ specimens was the L-R. The ESE(T) specimens were extracted not only from the FZ but also from the BM (L-R and L-C orientations) for comparison. Uniform crack fronts were observed in all ESE(T) specimens.

The results from testing the X65 BM, HAZ, and FZ in 3,000 psi (21 MPa) hydrogen gas are shown in Fig. 3. Tests performed in air at 10 Hz are also shown for comparison. The specimen orientation relative to the pipe is identified in parenthesis for each specimen tested and can be referenced in Fig. 2, and tests performed in triplicate are identified by (x3). The FZ and BM (C-L) exhibited similar fatigue crack growth rates (FCGR) in hydrogen gas over the entire ΔK range. The HAZ exhibited slightly lower crack growth rates in hydrogen gas for the lower ΔK range but shows similar trends to the BM (C-L) and FZ in the higher range of ΔK. These results indicate that the weld FZ and HAZ are no more susceptible to hydrogen-accelerated fatigue crack growth compared to the base metal.

The effect of orientation on hydrogen-assisted FCGR is not well documented for line pipe steels, mainly because geometrical constraints preclude testing in all orientations; however, the ESE(T) specimen geometry allows for BM samples to be extracted in the L-R orientation (see Fig. 2). Additional tests were performed on BM ESE(T) specimens for the L-R orientation in both air and hydrogen gas as shown in Fig. 3. The FCGR of BM (L-R) was 2 to 4 times lower in air and nearly an order of magnitude lower in hydrogen throughout the ΔK range as compared to BM (C-L) tests in the respective environments. Microstructural banding is prevalent in pipeline steels with features elongated in the longitudinal and circumferential directions. Cracks propagating perpendicular to the banding (i.e. in L-R orientation) will encounter sequential planes of ferrite and pearlite which may affect FCGR. Previous work [2] showed lower hydrogen-assisted FCGR in fully pearlitic microstructures as compared to pure iron. This microstructural banding appears to play an important role in hydrogen-assisted FCGR for the X65 base metal when crack growth is perpendicular to the banded direction as demonstrated by the significantly lower FCGR in the BM (L-R) orientation.

In collaboration with the International Institute for Carbon-Neutral Energy Research (I2CNER), custom heats of two high-purity Fe-C materials were procured and delivered to Kyushu University (Fukuoka, Japan): a fine grain size (15 μm) heat and a coarse grain size (70 μm) heat. The goal was to assess whether the grain size in these model ferrite-pearlite steels affects the onset of hydrogen-accelerated cracking. Tests on the model steels were performed at 10 Hz and R=0.1 in 2.1 MPa hydrogen gas. Figure 4 shows the FCGR curves for the fine and coarse grain size steels along with data for a commercial ferrite-pearlite steel (SA516) in both air (15 Hz) and hydrogen gas (1 Hz) for comparison. Both model ferrite-pearlite steels exhibited the onset of hydrogen-accelerated fatigue crack growth at similar ΔK ranges, suggesting that grain size does not have a pronounced effect. However, further testing (i.e. at higher load ratio, R) is required to conclusively demonstrate such insensitivity of hydrogen-assisted FCGR to grain size.

Conclusions and Future Directions

- Reliable da/dN vs. ΔK relationships were measured for the X65 weld FZ in 3,000 psi (21 MPa) hydrogen gas. Comparison of results shows that weld FZ and HAZ are no more susceptible to hydrogen-accelerated fatigue crack growth compared to the base metal.
- (future) Orientation was shown to have a pronounced effect on crack growth rate, therefore, in order to compare the BM and HAZ results directly similar orientations should be tested. Tests on C(T) specimens from the BM in L-C orientation will provide a more direct comparison to results from completed tests on C(T) specimens in the HAZ.
- (future) ESE(T) specimens will be extracted from a friction stir welded X52 pipe provided by ORNL for triplicate testing in 3,000 psi (21 MPa) hydrogen gas.

Special Recognitions & Awards/Patents Issued

1. Best Poster Award: "Assessing Hydrogen Pipeline Reliability: Quantifying Susceptibility of Pipeline Steels to Hydrogen Gas-Accelerated Fatigue Crack Growth," Brian Somerday and Joe Ronevich, ASME 12th Fuel Cell Science, Engineering and Technology Conference Boston, MA, June 30 - July 2, 2014.
2. Brian Somerday and Chris San Marchi, DOE Hydrogen and Fuel Cells Program Awards, Hydrogen Delivery and Safety, Codes and Standards, 2014.

FY 2014 Publications/Presentations

1. "Effect of Hydrogen Gas Impurities on the Hydrogen Dissociation on Iron Surface", A. Staykov, J. Yamabe, and B.P. Somerday, *International Journal of Quantum Chemistry*, vol. 114, 2014, pp. 626-635
2. "Measurement of Fatigue Crack Growth Relationships for Steel Pipeline Welds in High-Pressure Hydrogen Gas", J.A. Ronevich and B.P. Somerday, submitted to Second International Conference on Metals & Hydrogen, 2014.
3. "Measurements of H_2-Assisted Crack Growth in Pipeline Steels at SNL", J. Ronevich, B. Somerday, C. San Marchi, and K. Nibur, Joint DOC/DOE/DOT Meeting on Hydrogen Fuel Research, NIST, Boulder, CO, Dec. 2013.
4. (invited) "Modeling of Gaseous Impurity Inhibition of Hydrogen Environment Embrittlement", B. Somerday, A. Staykov, P. Sofronis, and R. Kirchheim, TMS 2014 Annual Meeting & Exhibition, San Diego, CA, Feb. 2014.
5. "Structural Materials Challenges in the Deployment of Hydrogen Pipelines", B. Somerday, Hydrogen Transmission and Distribution Workshop, National Renewable Energy Laboratory, Golden, CO, Feb. 2014.
6. (invited) "Addressing Critical Issues for Hydrogen Gas-Accelerated Fatigue Crack Growth in Pipeline Steels: Effects of Welds and Oxygen Gas Impurities", B. Somerday, University of Virginia, Charlottesville, VA, March 2014.
7. "Assessing Hydrogen Pipeline Reliability: Quantifying Susceptibility of Pipeline Steels to Hydrogen Gas-Accelerated Fatigue Crack Growth", B.P. Somerday and J.A. Ronevich, ASME 12th Fuel Cell Science, Engineering and Technology Conference Boston, MA, June 30 - July 2, 2014.

References

[1] Neeraj, T., Gnaupel-Herold, T., Prask, H.J., Ayer, R., "Residual stresses in girth welds of carbon steel pipes: neutron diffraction analysis," Science and Technology of Welding and Joining. Vol. 16, No.3, pp. 249-253, 2011.

[2] Cialone, H.J. and Holbrook, J.H. "Microstructure and fractographic features of hydrogen-accelerated fatigue crack growth in steels" in Microstructural Science Vol. 14, Eds. M.R. Louthan, I. LeMay, and G.F. Vander Voort, ASM, Metals Park, OH, (1987) pp. 407-422.

Acronyms

ASME – American Society of Mechanical Engineers

BM – base metal

C - circumferential

C(T) – compact tension

da/dN – fatigue crack growth rate

ΔK – stress-intensity factor range

ESE(T) – eccentrically loaded, single edge tension

FCGR – fatigue crack growth rate

FZ – fusion zone

HAZ – heat-affected zone

L – longitudinal

R – radial

Figure Captions

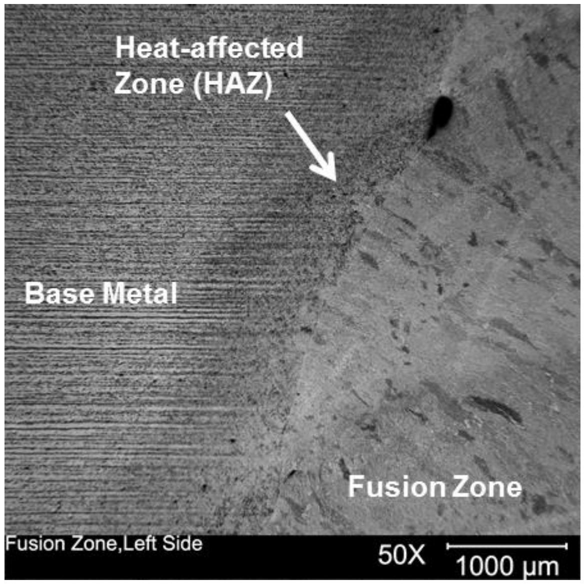

Figure 1. Optical-microscope image showing base metal, fusion zone, and HAZ of X65 weld.

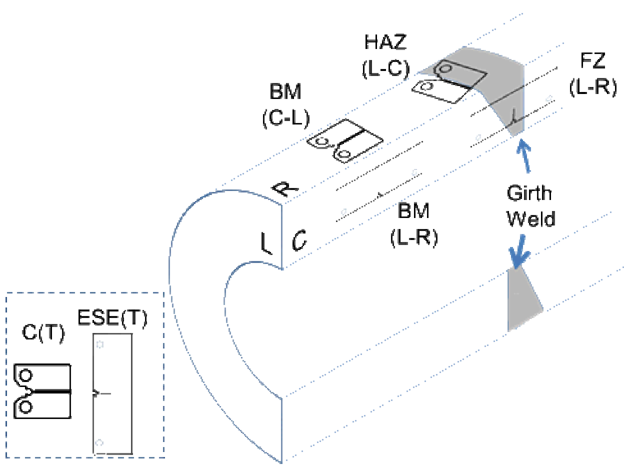

Figure 2. Schematic of gas metal arc welded pipeline with ESE(T) and C(T) specimens superimposed to show orientation tested for base metal (BM), heat-affected zone (HAZ), and fusion zone (FZ).

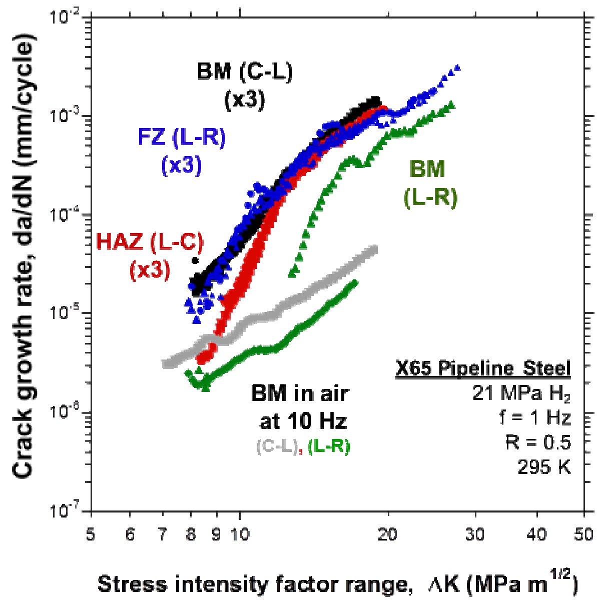

Figure 3. Fatigue crack growth rate relationships (da/dN vs. ΔK) for X65 base metal (BM), heat-affected zone (HAZ), and fusion zone (FZ) in 3,000 psi (21 MPa) hydrogen gas. The results are compared to measurements for X65 BM in air.

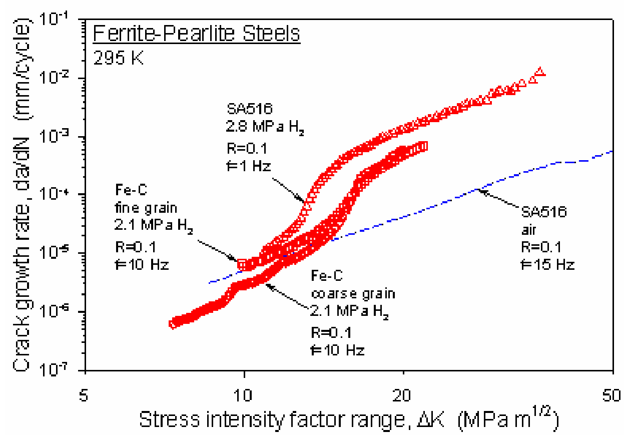

Figure 4. Fatigue crack growth rate relationships (da/dN vs. ΔK) for fine and coarse grained model ferrite-pearlite steels. Data for a commercial ferrite-pearlite steel (SA516) is shown for comparison in air and hydrogen gas.