III.20 Hydrogen Permeability and Integrity of Steel Welds

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Objectives

- Quantify the effects of welding and joining on the resistance to hydrogen embrittlement (HE) of high-strength pipeline and other structural steels under high-pressure hydrogen.
- Determine the hydrogen transport behavior (absorption, diffusion, trapping, etc.) in steels.
- Develop the technical basis and guidelines to manage hydrogen, stress and microstructure in the weld region to ensure structural integrity and safety of hydrogen delivery systems.
- Develop welding/joining technology that greatly reduces the capital cost and eliminate HE concerns in constructing new pipelines and converting existing pipelines for high-pressure hydrogen delivery.
- Develop risk assessment based approach to manage the integrity and safety of hydrogen pipelines including weld joints.

Technical Barriers

This project addresses the following technical barriers from the Delivery section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

(D) High Capital Cost and Hydrogen Embrittlement of Pipelines
(F) Gaseous Hydrogen Storage and Tube Trailer Delivery Costs
(G) Storage Tank Materials and Costs
(K) Safety, Codes and Standards, Permitting

Technical Targets

This project is to develop the scientific understanding, technical basis and cost-effective engineering solutions to control and mitigate hydrogen embrittlement in the steel weld region of pipelines and other high-pressure hydrogen delivery infrastructure systems. Insights gained from this project will be applied toward the hydrogen delivery infrastructure that meets the following DOE 2017 hydrogen pipeline delivery targets:

- Capital cost: $490K/mile for transmission pipeline and $190K/mile for distribution pipeline.
- Cost of delivery of hydrogen <$1.00/gasoline gallon equivalent (gge).
- Reliability/Integrity: Acceptable for H_2 as a major energy carrier.

Accomplishments

- Baseline measurements of hydrogen permeation, diffusion and trapping in pipeline steels have been completed. Preliminary investigations on the effects of weld microstructure on the previously mentioned hydrogen transport behavior have been completed.
- Initial studies on friction stir welding of pipeline steels have completed. The results indicated considerable improvement of weld toughness and strength over the conventional arc welds.
- Completed development of a new testing method specific to quantify and rank the sensitivity of different microstructures in the weld region to hydrogen embrittlement.

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Introduction

The hydrogen energy delivery infrastructure will require extensive use of steels and other cost-effective structural and functional materials under high-pressure gaseous hydrogen (H_2) exposure. For example, high pressure (up to 3,000 psi) hydrogen pipelines are presently considered to be one of the most cost-effective and energy-efficient means to transport very large amounts of hydrogen to much of the market as is done currently for natural gas [1]. Under high hydrogen pressures, there are concerns about HE of steel pipelines and its potential catastrophic consequences [2]. Concerns regarding HE are not limited to steel pipelines; according to a recent DOE Basic Energy Science Office report [3], HE needs to be addressed for a variety of hydrogen storage and delivery system parts made of metallic materials that are exposed to hydrogen.

As in the case of natural gas and other energy carrier transmission pipelines, welding will be used to construct steel pipelines for high-pressure hydrogen delivery. Welding will be also widely used in fabrication of other system components for hydrogen production, storage, and delivery. However, welds in pipeline steels and other engineering materials are often the most susceptible regions to HE due to the formation of unfavorable microstructures and high tensile residual stresses. Indeed, recent studies [4] on pipeline steels have shown that the weld region exhibits delayed cracking (signature of HE) when exposed to high-pressure hydrogen gas. Furthermore, the weld region typically has substantially lower resistance to hydrogen crack initiation and higher crack growth rates, when compared to the baseline pipeline steel (base metal). *In this regard, the weld region can be the weakest link for the structural integrity and safety of hydrogen pipelines and hydrogen delivery infrastructure.* A systematic approach to deal with weld property degradation under high-pressure hydrogen gas is critical to ensure the safe, cost-effective operation and long-term reliability of the hydrogen delivery infrastructure.

Approach

While there have been extensive studies in the past on HE and hydrogen-induced material property degradation of pipeline steels, the high-pressure hydrogen delivery pipeline presents some unique issues that are seldom addressed in the past. At the center of these issues is the hydrogen transport behavior in metal – the absorption, diffusion, and trapping of hydrogen in metal. At the present time, very limited knowledge is available about the rate of diffusion and amount of hydrogen in steel under high-pressure gaseous environment relevant to the hydrogen delivery infrastructure. Therefore, the first major effort in this project is directed toward high-pressure hydrogen permeation and mechanical performance tests to systematically study the hydrogen permeation behavior and to evaluate the tolerance level to hydrogen of different steels before considerable mechanical property degradation would occur.

The weld joint in steel pipeline is expected to be a critical region mostly susceptible to hydrogen embrittlement due to the formation of unfavorable microstructure and the high residual stresses. The second major activity of this project focuses on developing new welding technology and/or improve existing welding technology to optimize weld microstructure and proactively control the weld residual stress for hydrogen pipeline construction and repair. In addition, special testing methods need to be developed to quantify the degradation of mechanical properties in the weld region with complex microstructure and HE resistance gradients. Finally, risk assessment based approach will be developed to manage the integrity and safety of hydrogen pipelines including the weld joints.

Results

The Fiscal Year 2009 activities focused on the development of testing methods and system for quantifying the degree of weld property degradation under high-pressure hydrogen pressure.

Current standard methods for testing hydrogen-induced mechanical property degradation of base metal have shown to be inadequate for the weld metal region due to the highly inhomogeneous microstructure and property gradients of the weld region [4]. In this project, we designed and fabricated miniaturized and self-loading testing devices for in situ measurement of mechanical property degradation of weld metal in high-pressure gaseous hydrogen environments. Two types of the mechanical testing devices were devised. The first one was a special multiple-notched tensile test where the notch is precisely located in different regions of weld. This test is intended as a quick yet quantitative screening test of weld property changes under high-pressure hydrogen. The second type test was a novel fracture toughness test based on the spiral notch torsion test (SNTT) method. The compact size and self-loading mechanisms employed in both testing devices make it possible that the entire loading assembly and the test specimen be placed inside a relatively small high-pressure chamber. This minimizes the capital cost of the testing system and allows for multiple testing devices to be operated in a single high-pressure vessel to cost-effectively study and quantify the effects of the weld microstructure on HE resistance of different steels. The effects of high-pressure hydrogen on the stability of load sensors immersed in hydrogen have been solved with a novel sensor design.

In FY 2009, we focused on the development of the in situ SNTT method under high-pressure hydrogen. A unique miniature self-loading device was successfully developed that can be placed inside the autoclave. The long-term stability of the load-monitoring system with temperature and pressure self-compensation capability and resistance to hydrogen permeation was successfully evaluated under high-pressure hydrogen. Figure 1 shows the design of the spiral notch specimen

FIGURE 1. SNTT specimen for K_{th} Measurement under High-Pressure Hydrogen

used in this project for fracture toughness threshold (K_{th}) measurement under high-pressure hydrogen environment. A U.S. patent application has been submitted for the in situ SNTT method under high-pressure hydrogen developed in this project.

In addition, a 3-dimensional finite element model has been developed to determine the stress intensity factor for the specific SNTT configuration used in this work. Figure 2 shows the circumferential stress (Mode I stress) distribution in the specimen, from which the stress intensity can be calculated. This model will be used to calculate Kth for high-pressure hydrogen embrittlement test planned in next quarter.

The validity of the SNTT method was first confirmed by testing American Iron & Steel Institute (AISI) 4340 steel in air, by comparing with the fracture toughness (K_{IC}) results of Bandyopadhyay et al. [5] on the same steel obtained using the standard compact tension (CT) specimen. It is important to note that, in order to compare with the CT testing results, the entire SNTT specimen was heat-treated to a uniform microstructure by furnace heating and oil quenching. The AISI 4340 steel was heat treated to fully hardened microstructure with hardness of 50-53 Rockwell Scale C, following the heat treatment procedure by Bandyopadhyay. The K_{IC} value from the SNTT was averaged at 67.5 ksi-in½, which is consistent with reported 65-75 ksi-in½ by standard CT specimen.

Under 2,000 psi high-pressure hydrogen, the threshold fracture toughness for crack initiation (Kth) reduced to 36–39 ksi-in½ (obtained from multiple samples). The reduction is about 55% of K_{IC} values in air. The broken sample shown in Figure 3 confirmed that the Mode I fracture is achieved in the SNTT test under high-pressure hydrogen.

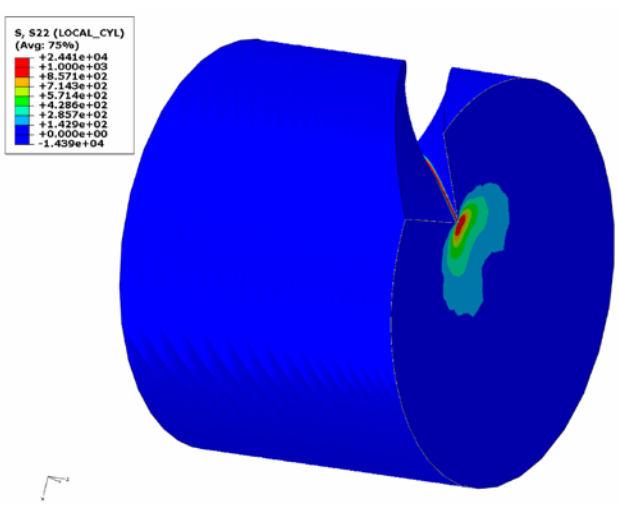

FIGURE 2. Circumferential Stress Distribution in SNTT Specimen

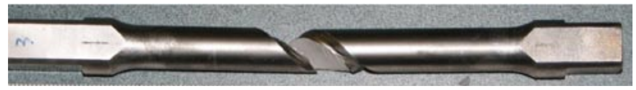

FIGURE 3. Broken SNTT Specimen Tested in High-Pressure Hydrogen for K_{th} Measurement

In addition, SNTT steel specimens with non-uniform microstructure distributions were produced with the Gleeble thermophysical simulator. These specimens will be tested in air and in high-pressure hydrogen to study the effect of inhomogeneity of microstructure/property on fracture toughness of steels. SNTT specimens made from actual welded pipelines will be then tested to determine the degree of property degradation of pipeline steel welds under high-pressure hydrogen.

In summary, we have successfully demonstrated the SNTT method for Kth testing under high-pressure hydrogen. The SNTT test has been performed using uniform microstructure specimen. We will continue to develop the SNTT method to test non-uniform microstructures in the heat-affected zone and weld region in the remaining of the project. The research on weld property testing under high-pressure hydrogen environment in this project has resulted in three U.S. patent applications in FY 2009.

Conclusions and Future Directions

This project focuses on the resistance of HE in the weld region of steels, the region regarded as the weakest link for the structural integrity and safety of hydrogen pipeline and hydrogen delivery infrastructure. We have accomplished the followings milestones in FY 2009:

- Baseline measurements of hydrogen permeation, diffusion and trapping in pipeline steels and preliminary investigations on the effects of weld microstructure on hydrogen transport.
- Initial work on friction stir welding on pipeline steels which showed considerable improvement of weld toughness and strength over the conventional arc welds.
- Development of a new testing method specific to quantify and rank the sensitivity of different microstructures in the weld region to hydrogen embrittlement.

The project plan for FY 2010 includes (1) develop the understanding and the relationship between hydrogen permeation and diffusion and various microstructures in pipeline steels, (2) continue on welding technology development to reduce the weld residual stress and manage the weld microstructure to improve the resistance to HE, and (3) complete the development of testing methods to quantify the HE resistance in the weld region under both static and cyclic loading conditions.

Special Recognitions & Awards/Patents Issued

Three U.S. patent applications have been filed in FY 2009 in this project on the subject of weld mechanical property testing under high-pressure hydrogen subjecting to static and low-cycle cyclic loading conditions.

FY 2009 Publications/Presentations

1. Feng, Z. Steel, R. Packer, S. and David, S.A. 2009. "Friction Stir Welding of API Grade 65 Steel Pipes." – ASME PVP Conference, Prague, Czech Republic.

2. Feng, Z. Anovitz, L.M., Armstrong, T.A. 2009. "hydrogen permeation and diffusion in steels under high pressure hydrogen (Invited)." – TMS Annual Meeting, San Francisco, CA.

3. 2009 DOE Hydrogen Program Review – Washington, DC, May 2009. PDP 43.

References

1. U.S. Department of Energy, "National Hydrogen Energy Roadmap," Nov. 2002, http://www.eere.energy.gov/hydrogenandfuelcells/pdfs/national_h2_roadmap.pdf.

2. U.S. Department of Energy, "Hydrogen, Fuel Cells & Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan," January 21, 2005, page 3-40, http://www.eere.energy.gov/hydrogenandfuelcells/mypp/.

3. U.S. Department of Energy, "Basic Research Needs for the Hydrogen Economy," *Basic Energy Sciences Workshop on* Hydrogen *Production, Storage, and Use*, Second Print, February, 2004.

4. Xu, K. "Evaluation of API 5L X80 in High Pressure Hydrogen Gas," ASTM G1.06 Hydrogen Embrittlement Workshop, Nov 8, 2005, Dallas, TX.

5. Bandyopadhyay et al, *Metallurgical Transactions A*, **14**(5), 881-888 (1983).