III.15 Integrity of Steel Welds in High-Pressure Hydrogen Environment

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Fiscal Year (FY) 2011 Objectives

- Develop and demonstrate cost-effective welding technology that can mitigate hydrogen embrittlement (HE) concerns in constructing new pipelines and converting existing pipelines for high-pressure hydrogen delivery.
- Quantify the effects of welding and joining on the resistance to HE of high-strength pipeline and other structural steels under high-pressure hydrogen.
- Develop the technical basis and guidelines to manage the weld region to ensure structural integrity and safety of hydrogen delivery systems.
- Determine the hydrogen transport behavior (such as diffusion, trapping, etc.) in steels.

Technical Barriers

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

(D) High Capital Cost and Hydrogen Embrittlement of Pipelines

(K) Safety, Codes and Standards, Permitting

Technical Targets

This project aims at developing the scientific understanding, technical basis and cost-effective engineering solutions to control and mitigate hydrogen embrittlement in the weld region of steel 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 technical targets:

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

FY 2011 Accomplishments

- Baseline high-pressure hydrogen permeation measurements established the effects of weld microstructure, surface conditions, temperature and hydrogen pressure on hydrogen permeation, diffusion and trapping in selected pipeline steels.
- The in situ spiral notch torsion test (SNTT) and the associated finite element fracture mechanics modeling were applied to quantify the fracture toughness degradation of steel welds exposed to high-pressure hydrogen. For American Iron & Steel Institute (AISI) 4340 high-strength steel, a common material for hydrogen storage tank, preliminary testing results showed the fracture toughness of weld region in high-pressure hydrogen is as low as 25% of that in air (i.e., a degradation of 75%).
- Preliminary multi-notch tensile testing of American Petroleum Institute (API) 5L Grade X-65 pipeline steel in hydrogen showed encouraging results for the improved resistance to HE achieved by friction stir welding (FSW). Additional tests on X-65 FSW are ongoing and are expected to be completed by September-November 2011 timeframe.

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Introduction

The hydrogen energy delivery infrastructure will require extensive use of steels and other cost-effective structural materials under high-pressure gaseous hydrogen 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 for natural gas [1]. Under high hydrogen pressures, there are concerns about HE of steel pipelines and its potentially catastrophic consequences [2]. Concerns on HE are not limited to steel pipelines; according to a recent DOE Basic Energy Science report [3], HE needs to be addressed for a variety of hydrogen storage and delivery systems 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 also be 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. 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 is oftentimes 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 exposure is critical to ensure the safe, cost-effective operation and long-term reliability of the hydrogen delivery infrastructure. Insights gained from this project will provide the scientific and technical basis for hydrogen pipelines that meet relevant DOE 2017 technical targets on total capital investment and reliability/integrity.

Approach

Currently, there exists only a limited amount of knowledge on the diffusion rate and amount of hydrogen in steels under high-pressure gaseous environment relevant to the hydrogen delivery infrastructure. Therefore, the first major effort of this project is directed toward the systematical study of (1) high-pressure H_2 permeation behavior in steels and their welds, and (2) the tolerance level to hydrogen of different steels before considerable mechanical property degradation would occur. A special hydrogen permeation testing system has been developed to test H_2 permeation and diffusion in coupon-size steel discs under high pressure (up to 5,000 psi). The effect of steel composition, microstructure, and sample surface conditions on H_2 transport behavior is studied.

As discussed earlier, the weld joint in steel pipeline is expected to be a critical region highly susceptible to HE due to the existence of unfavorable microstructure and high tensile residual stresses. Hence, the second major activity of this project focuses on generating weld property data for fracture-mechanics based pipeline design. To account for the highly inhomogeneous nature of the weld region, two special tests have been developed and validated. A tensile test is used for relative ranking of weld microstructure susceptibility to HE, while the spiral notch torsion test is used for quantifying fracture toughness degradation in steel welds.

The third major effort involves the development and validation of new welding technology for H_2 delivery steel pipelines. In particular, solid-state FSW is evaluated for improved mechanical property and reduced cost for pipeline construction and repair.

Results

The results achieved in this FY (2011) are highlighted as follows.

Quantitative Understanding of Hydrogen Embrittlement in Steel Weld Using In Situ SNTT

Although they may work for testing of base metal in hydrogen, current standard methods such as compact tension for testing hydrogen-induced mechanical property degradation have shown to be inadequate when applied to the weld due to the highly inhomogeneous microstructure and property gradients of the weld region [4]. Special, miniature and self-loading devices for in situ mechanical testing in high-pressure hydrogen were designed and fabricated previously. The FY 2011 activity is focused on fracture toughness measurement of welds exposed to a high-pressure gaseous hydrogen environment using in situ SNTT.

Figure 1 shows the in situ device for fracture toughness testing in high-pressure hydrogen. In the test, the specimen with a spiral notch is loaded in the load frame where a pure torsional load is applied to the specimen. The load frame containing the pre-stressed specimen is placed inside a high-pressure vessel. The strain gages attached to the load frame are connected to the data acquisition box via feedthroughs in the vessel cover/lid. After the assembly, the vessel is tightly sealed and is charged with hydrogen gas to the desired pressure (typically around 1,900 psi or 13.1 MPa). During the test, the strain gage readings are monitored for the variation of torsional load on the specimen, which is an indicator for hydrogen-induced cracking initiation. As the hydrogen permeation in steel can be slow at room temperature, the specimen is immersed in H_2 and tested for

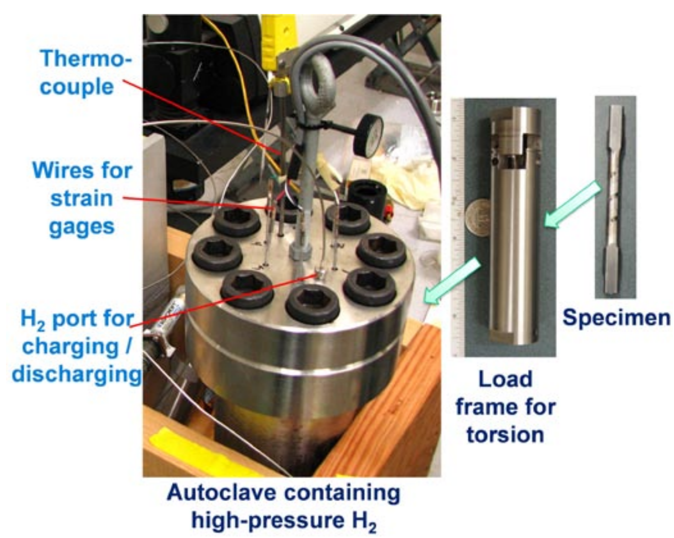

FIGURE 1. Special, in situ testing apparatus for measuring fracture toughness degradation of materials exposed to high-pressure hydrogen

one to three days to ensure sufficient time for hydrogen to diffuse to the crack tip.

As shown in Figure 1, the compact size and self-loading mechanism employed in the in situ SNTT device make it possible to accurately measure mechanical property degradation in high-pressure hydrogen without the use of a dedicated mechanical testing system. This minimizes the capital cost of the testing system and allows cost-effective study of the effect of weld microstructure and its inhomogeneity on HE resistance.

The steel studied using in situ SNTT is AISI 4340 high-strength steel, a material commonly used for hydrogen storage tank. Simulated weld heat-affected zone (HAZ), which is the most critical region to HE, is prepared using a Gleeble® thermal-mechanical system. Each specimen is subject to a heating and cooling cycle mimicking that encountered in practical welding condition. Using Gleeble to produce simulated weld specimens eliminates the need for fabricating a 4340 steel weld and machining specimens from the weld, which can be costly and time-consuming. After heat-treatment in Gleeble, the simulated weld specimens are cyclic-fatigued in air to induce sharp crack tips before testing in hydrogen.

Figure 2(a) shows the surface appearance of a fractured specimen after exposure to hydrogen at 1,900 psi. Figure 2(b) is the scanning electron microscope (SEM) image of fracture surface near the notch root, where primarily intergranular fractures (i.e., brittle) are observed. As it locates just underneath the surface directly exposed to hydrogen, the region near the notch root is likely saturated with hydrogen. The tensile stress field in front of the notch tip further enhances the hydrogen diffusion. The presence of hydrogen severally deteriorates the local microstructure resistance to HE, thus resulting in a brittle fracture. Figure 2(c) is the SEM image of fracture surface away from the notch root. Both dimples (ductile) and intergranular fractures (brittle) are observed, indicting a mixed ductile and brittle fracture behavior. Detailed microstructure analyses of this region is ongoing to understand the extent of hydrogen embrittlement there.

The preliminary testing results obtained on fatigue pre-crack specimens show that the fracture toughness of 4340 steel weld HAZ drops from 38.7 ksi$\cdot\sqrt{in}$ in air to only 9.6 ksi$\cdot\sqrt{in}$ in gaseous hydrogen at 1,900 psi. The measured fracture toughness values are consistent with those reported in the literature [5]. Exposure to high-pressure hydrogen results in significant reduction in fracture toughness for the 4340 steel weld HAZ. Such hydrogen embrittlement is indeed severe, though not unexpected. The 4340 steel relies on a martensitic microstructure for its high strength. The martensitic microstructure and associated high carbon content make 4340 steel prone to HE [5]. On the other hand, new generation pipeline steels often utilize micro-alloying for improved strength and favorable microstructure for HE resistance.

Cost-Effective FSW of Pipeline Steel for Improved HE Resistance

Figure 3(a) is a snapshot of FSW of X-65 steel pipe sections. The resulting pipe girth weld is shown in

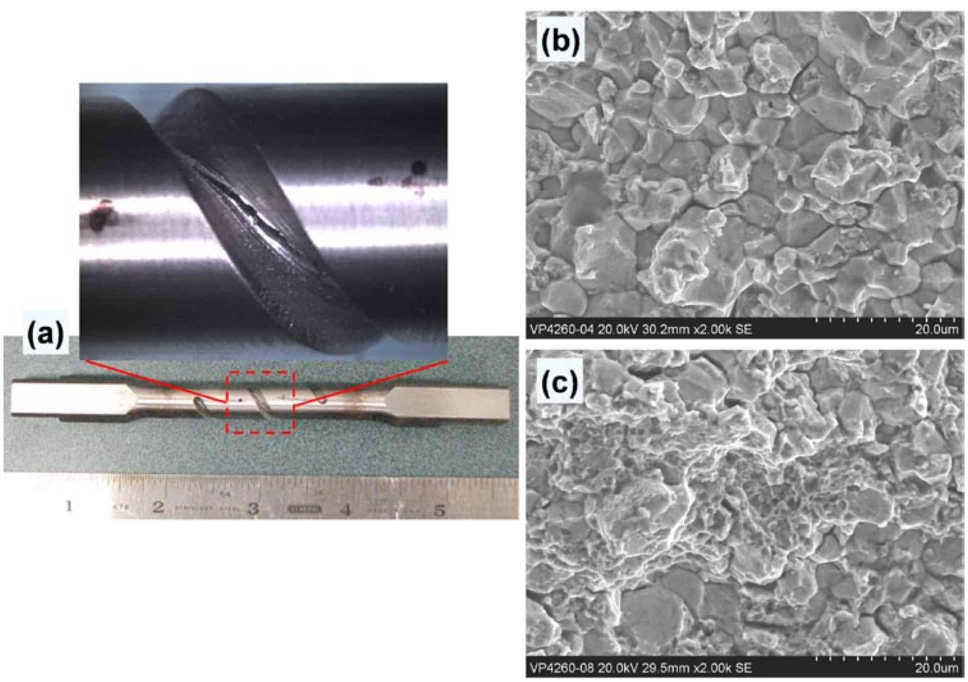

FIGURE 2. (a) Surface appearance of a fractured sample after hydrogen exposure, (b) SEM image of fracture surface near the notch root, and (c) SEM image of fracture surface away from the notch root

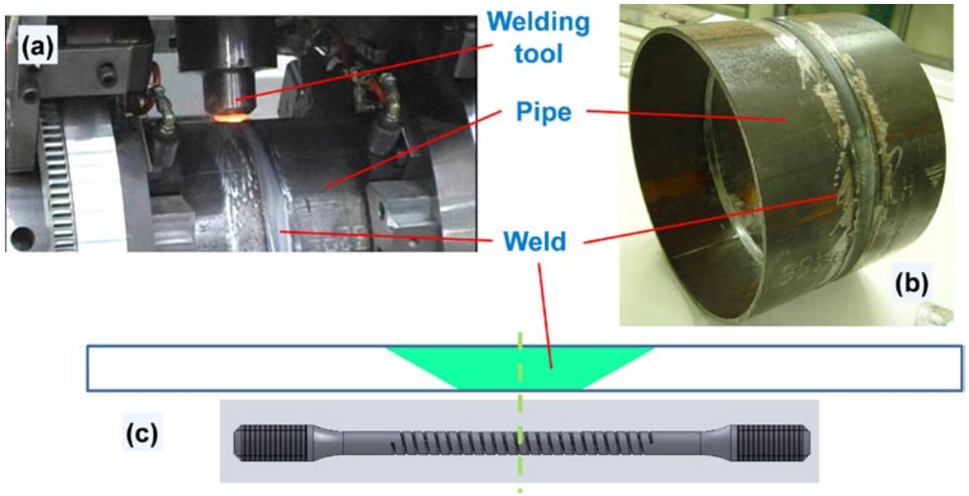

FIGURE 3. (a) Snapshot of FSW of X-65 steel pipe sections, (b) resulting pipe girth weld, and (c) schematic drawing of multi-notch tensile specimen

Figure 3(b). Previous studies on FSW of pipeline steel demonstrated considerable improvement of weld toughness and strength over the conventional arc weld in air [6]. In FY 2011, the in situ multi-notch tensile test is applied to study the resistance to HE of friction stir weld exposed to high-pressure hydrogen. Figure 3(c) is a schematic of the transverse cross-section of friction stir girth weld from which the multi-notch specimen is machined. The helical notch samples various weld regions including the stir zone, thermo-mechanically affected zone, HAZ and base metal. During testing, the notched sample is put into a load frame similar to that shown in Figure 1, where a tensile load rather than a torsional load is applied to the sample. The tensile load frame is placed inside the pressure vessel, charged to the desired hydrogen pressure, to measure the threshold tensile load for crack initiation in hydrogen.

It is noted that the multi-notch tensile test gives a qualitative ranking of microstructure sensitivity to HE. It is used because the notched samples can be readily machined from the relatively thin X-65 pipe (wall thickness = 0.24 in., or 6.1 mm). Nevertheless, such tests can provide insights into the relative performance of friction stir welds over conventional arc welds.

Preliminary tests have shown some encouraging results, as the friction stir weld sample exposed to hydrogen does not fracture at a load that is 70% of the fracture load in air. Additional tests on X-65 FSW are ongoing and are expected to be completed by September-November 2011 timeframe.

Improving Capability of Testing Apparatus

Since the current testing apparatus directly utilizes the regular hydrogen cylinder for pressuring, the maximum pressure is thus limited to about 1,900 psi. A new hydrogen charging system capable of maintaining or varying H_2 pressure up to 10,000 psi has been designed and assembled. The schematic drawing of the charging system is shown in Figure 4. The system will enable the study of the effect of hydrogen on material properties under very high pressures that are relevant to hydrogen production, delivery, and storage infrastructure.

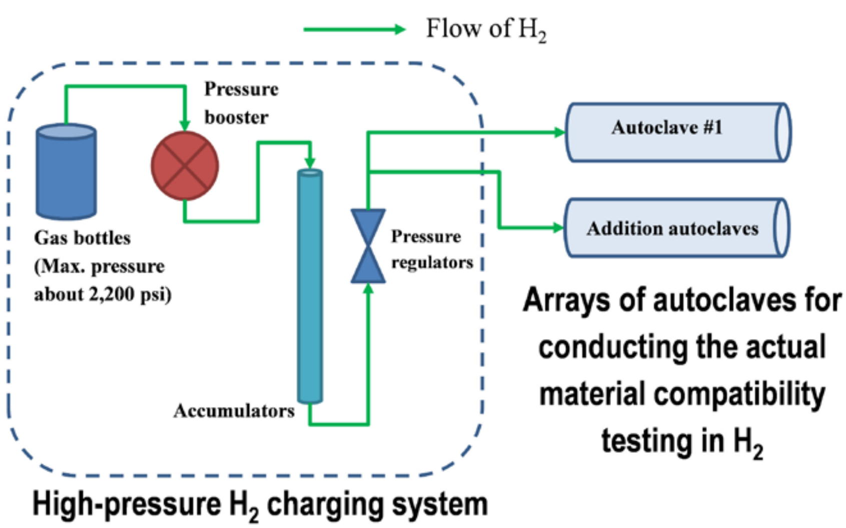

FIGURE 4. Schematic of a hydrogen charging system capable of maintaining or varying pressure up to 10,000 psi

Conclusions and Future Directions

- Demonstrated in situ SNTT for cost-effective and accurate measurement of the fracture toughness degradation in steel welds exposed to high-pressure hydrogen. The quantitative knowledge provides the technical basis for managing the weld region, i.e., the weakest link for the structural integrity and safety of hydrogen delivery infrastructure.
- Preliminary results showed FSW of pipeline steel could result in favorable microstructure with superior properties compared to the conventional arc welding.
- For the duration of FY 2011, we plan to complete (1) the study on HE resistance of X-65 steel pipe FSW, (2) the microstructure characterization of fracture surface of 4340 steel weld exposed to H_2, and (3) the assembly of high-pressure hydrogen testing system.
- At the discretion of DOE, future direction may include the application of special in situ testing methods and apparatus to study hydrogen-induced property degradation in other important materials and their welds relevant to hydrogen infrastructure.

FY 2011 Publications/Presentations

1. Wang, J., Ren, F., Zhang, W., Feng, Z., Anovitz, L.M., Chen Z. and Xu, H. 2011, "Development of in-situ Techniques for Torsion and Tension Testing in Hydrogen Environment," 2011 ASME PVP Conference, Baltimore, MD.

2. 2011 DOE Annual Merit Review, Fuel Cell Technologies Program, Washington, D.C., May 2011.

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3. U.S. Department of Energy, "Basic Research Needs for the Hydrogen Economy," Basic Energy Sciences Workshop on Hydrogen Production, Storage, and Use, Feb. 2004, http://science.energy.gov/~/media/bes/pdf/reports/files/nhe_rpt.pdf.

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6. 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.