Hydrogen Permeability and Integrity of Hydrogen Transfer Pipelines

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Background

Hydrogen production and supply methodologies are important for hydrogen based transportation infrastructure. Transportation of hydrogen from reforming plants to dispensers includes liquid hydrogen transport through trucks and transportation of gaseous hydrogen through high-pressure pipelines. A recent case study of hydrogen infrastructure development indicated that transfer of gaseous hydrogen could be a economical option if the flow rates of hydrogen can be increased from 1 million standard cubic feet (scf) per day to 20 million scf per day in a 3 ft. diameter pipeline rated for 1000 psi pressure [1]. It is important to note that the flow rate is related to pressure differentials and the cross-sectional area of the pipes. Therefore, one could envision reduction of wall thickness and / or increase of pressure differential to increase the flow rates in these pipes [2]. Under these circumstances, current mild-steel pipeline infrastructure may prove inadequate and may require high strength steels.

For example, one may consider replacing Grade A (API 5L specification) steels with Grade X-52 or Grade X-80 type steels. The X-52 and X-80 steels have higher manganese concentrations (1.3 to 1.8 wt.% Mn) compared to Grade A (0.3 to 0.6 wt.% wt.% Mn). Due to increased manganese concentrations, these steels can readily form hard martensite under rapid weld cooling conditions in the heat-affected-zone (HAZ) region of the weld. If a filler wire with identical composition was used to join these steels, bainite and martensite microstructures may form in the weld metal (WM) region. Moreover, traditional fusion welding processes such as submerged arc welding and manual metal arc welding also lead to high residual stresses in the welded region. It is well known that with the presence of hard microstructure and high residual stress, the welds will be prone to hydrogen induced cracking (HIC) [3]. The HIC is enhanced with the presence of diffusible hydrogen. For many decades, the above problem has been addressed by careful choice of filler metal compositions to reduce the source of hydrogen and by employing post-weld heat treatment to remove hydrogen. However, the application of high strength steel welded pipes to hydrogen transport leads to new challenge as illustrated below.

In traditional piping applications of natural gas, the welded high strength steel piping is not exposed to very high hydrogen pressures and therefore hydrogen pick up during service is not a major issue. In the present case, the inner surfaces of tubes are exposed to pure hydrogen at large pressures where as the outer surfaces of tubes are exposed to atmospheric pressure with low-partial pressure of hydrogen. This leads to a large chemical potential gradient for hydrogen diffusion from inner surface to outer surface. This will lead to a flux of hydrogen or permeation through the steel given by Fick's first law [4, 5].

$$J_\infty = -D\frac{\Delta c}{\Delta x}, \tag{1}$$

where the J_∞ is the steady state flux of hydrogen, D is the diffusivity (1 X 10^{-5} $cm^{-2}s^{-1}$), and Δc is the concentration difference over Δx distance. Using equation (1), and by assuming a concentration difference of 1 wt.PPM a steady state hydrogen flux is estimated to be 960 pL $cm^{-2}s^{-1}$. In hydrogen transfer applications; the above flux may be increased by many orders of magnitude. Under these conditions, if the underlying microstructure in welded region is very hard and with the presence of residual stresses, the welded joints may exhibit HIC after certain service time. As a result, there is a need for designing base metal composition (modified X-52 or X-80 compositions), filler metal composition (matching composition with hydrogen traps), welding process (to minimize the heat-affected-zone), and process parameters (minimize the residual stresses) for optimum performance of reduced HIC risk as well as reduced hydrogen permeability.

Research Approach

The goals of the research are to minimize the HIC in the base metal, HAZ, and WM regions while reducing the steady state flux through the pipe. To reach this challenging goal, the proposed research will leverage extensive knowledge base that is available on hydrogen embrittlement phenomena and weld microstructure development in steels. With this knowledge, innovative process-material combinations will be derived for high strength steel piping for high-pressure hydrogen transport. Three proposed research approaches are briefly discussed below.

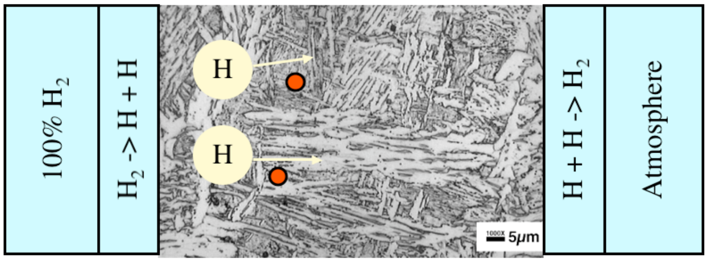

Fig. 1 Schematic diagram shows the hydrogen transport from the interior of the pipe through a steel microstructure and escape to the atmosphere. The hydrogen trapping sites (red circles) and microstructure (background image from a weld) need to be designed for welds with good integrity for hydrogen transport.

Hydrogen Management: The first phase of the research must consider the hydrogen solubility into steel. The hydrogen diffuses into steel in the form of monatomic hydrogen [see Fig. 1]. On entering the steel, the hydrogen may be present in two forms (1) trapped at sites like inclusions and grain/phase boundaries and (2) diffusible form in the interstitial positions within the ferrite lattice [6]. The HIC is mostly caused by the diffusible hydrogen. Current research in this area focuses on increasing trapped hydrogen by increasing the number density of trapping sites. [7, 8, 9, 10]. These trapping sites can be either reversible or irreversible [11, 12]. Recent research has correlated the weakly binding trapping sites to environmental degradation [13]. Moreover, the presence of elastic stresses appears to increase the hydrogen permeation and plastic deformation appears to reduce the hydrogen permeation [14]. This reduction is related increased to dislocation density that acts as trapping sites for hydrogen. The above research shows that the hydrogen management can be achieved by careful control of microstructure in steels. In this ORNL will collaborate with Lincoln Electric on filler metal design for the use in pipeline industries. This design will be focused on the various aspects of hydrogen management and modifying the dynamic microstructure evolution. In this task, conventional fusion welding processes including submerged arc welding, manual metal arc welding, gas metal arc welding and flux cored arc welding processes will be considered and one of the ideal process will be selected as a candidate based on initial scoping studies.

Weld Stress Management: In the second phase, stresses that are present in the weld metal region. Recently, it has been shown that in the presence of stress fields around the crack tip, the presence of diffusible hydrogen. Recent in-situ TEM analyses have shown that diffusible hydrogen increases the velocity of dislocations resulting in localized plasticity that leads to embrittlement [15]. It is well known that welding leads to large residual stresses and prolonged exposure to hydrogen rich atmosphere may lead to localized plasticity initiation even without any external load. Therefore, it is imperative that welds are designed such a way that they do not lead to catastrophic cracking under service conditions. Thermal stress management in welds is an active research area and the existing knowledge base at ORNL will be leveraged in this research. State of the art thermo-mechanical-metallurgical models will be developed that consider the interactions between thermal fields, dynamic microstructure evolution and effect of temperature and microstructure on the thermophysical properties [16]. In addition, new fusion welding processes, such as laser-assisted arc welding processes, will be considered to minimize the heat-affected-zone and to refine the weld metal microstructure [17].

Interface Barrier Design: In the final phase, it is possible to reduce the hydrogen concentration gradient by physically separating the steel and hydrogen rich atmosphere. This approach is currently being used in the oil pipelines for avoiding the corrosion of exterior surfaces of pipelines by coating with epoxy resin. Similar approaches can be used on the interior of the pipelines to retard the absorption of hydrogen into the steel, thus reducing the concentration gradient and peak concentration.

Evaluation of Welds and Base Material: The welds produced in this study will be compared with the performance of X-52 or X-80 line pipe steel. The evaluation will focus on microstructural characterization, hydrogen permeability, toughness (under both hydrogen charged and uncharged condition in both HAZ and WM region), and residual stress measurement. This task will utilize extensive characterization facilities that are available at ORNL.

Deliverables

At the end of three-year research effort, this research will lead to following deliverables:

(1) A suitable welding process – process parameter – filler wire – physical interface barrier design for welding high strength steel pipe lines
(2) A fundamental understanding of the effect of trapping sites and stresses on the hydrogen permeability in welds
(3) A thermo-mechanical-metallurgical model to evaluate the hydrogen cracking sensitivity in pipelines as a function of composition and thermal cycles.
(4) The final knowledge base can be actually used as guidance for the production of base material steel composition that will improve the overall performance of pipeline (reduce leakage and minimize HIC), not only, in the welds.

Required Level of Support

We envision the about outlined research would require three year research effort at a cost of $350 K per year. The total cost of the research for three year would be $1,050 K. Some of the research will be performed in collaboration with Lincoln Electric Corporation.

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