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Hydrogen-induced toughness drop in weld coarse-grained heat-affected zones of linepipe steel

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ABSTRACT

In this study, hydrogen effects on the impact toughness of simulated coarse-grained heat-affected zones (CGHAZs) in a linepipe steel were investigated in search of the possible "local brittle zone phenomenon" in hydrogen pipeline welds. After hydrogen charging, the inter-critically reheated and unaltered CGHAZ exhibited very low impact energies as well as occurrence of splitting. This hydrogen-induced toughness drop is discussed in terms of combined effects of brittle microstructures and hydrogen trapping.
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1. Introduction

In the context of this century's major energy-related issues arising from the non-renewable nature of fossil fuels and serious air pollution, recent interest in the use of hydrogen as an alternative main energy source has increased explosively. For preparing the up-coming era of the so-called "hydrogen economy," development of hydrogen infrastructures is essential. Particularly, hydrogen delivery is a critical issue for the widespread use of hydrogen. It has been reported that, among various transportation methods, a pipeline network infrastructure may be the most cost-effective and energy-efficient way to transport large amounts of hydrogen over long distance [1,2]. However, new design and construction of hydrogen pipelines may require high initial capital costs. A low-cost option for partly overcoming this issue is transporting gaseous hydrogen (in a form of either pure hydrogen or a blend of natural gas and hydrogen) using the existing natural gas pipelines [1,2].

One of the major concerns arising from hydrogen transportation through the existing natural gas pipelines under very high pressure level (up to ~3000 psi or ~21 MPa) is the influences of hydrogen entry and permeation on the mechanical performance of the pipeline steels. It is widely accepted that hydrogen almost always deteriorates the mechanical properties of materials by various ways including hydrogen embrittlement (HE), hydrogen-induced cracking (HIC), hydrogen attack (HA), sulfide stress cracking (SSC), and stress corrosion cracking (SCC) [3]. These hydrogen effects can be also available for the natural gas pipeline steels (that are specified by the American Petroleum Institute, API). Aside from the hydrogen pipeline issue, the influence of hydrogen in API steels has been studied for different purposes such as assessment of API steels' resistance to sour environment [4,5].

An important issue remaining unsolved yet may be the influence of hydrogen in weld heat affected-zones (HAZs) of the pipeline steels. The hydrogen effects in base metal may

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be very different from that in HAZ since welding can seriously alter (and generally degrade) the metallurgical and thus mechanical properties of materials. Especially, the coarse-grained HAZs (CGHAZs) adjacent to the weld fusion line often exhibit abnormally low fracture resistance and thus are referred to as local brittle zones (LBZs) in multi-pass welded joints [6–9]. Thus, analysis of the hydrogen effects in the CGHAZs may be essential for ensuring the safety and long-term reliability of the hydrogen transmission pipeline infrastructure. Nevertheless, somewhat surprisingly, little systematic research on the issue (i.e., hydrogen effects in CGHAZs) is available in the literature. As the first step to shed light on the issue, here we systematically explore (for the first time, to the best of our knowledge) the hydrogen effects on the toughness of pipeline steel CGHAZs to find out the possible LBZ phenomenon in hydrogen pipelines.

2. Experimental

The material examined in this study was a commercial grade API X70 steel, one of the most popular natural gas pipeline steel, whose chemical composition is 0.07C–0.25Si–1.55Mn–0.25Cu–0.2Ni–0.03V–0.015Ti–0.04Nb–0.03Al and balanced Fe (wt.%).

For systematic and reproducible evaluation of the local properties in HAZ, HAZ simulations were performed with a Gleeble 1500 thermo-mechanical simulator (DSI, Poestenkill, NY). The thermal cycles for HAZ simulation are typically characterized by both the peak temperature (T_P) and cooling time from 800 to 500 °C ($\Delta t_{8/5}$) which represent distance from heat source and welding condition, respectively. After reaching the first peak temperature (T_{P1}) of 1350 °C, the specimens were cooled down for a given $\Delta t_{8/5}$. Subsequently, the specimens were reheated to the second peak temperature (T_{P2}), and then cooled down under the same rate. Through a modified equation based on Rosenthal's heat flow formula [10], the $\Delta t_{8/5}$ was calculated as 18 s that is approximately equivalent to the cooling rate of a shielded metal arc welding (SMAW) of 17-mm-thick plate with a heat input of 30 kJ/cm. Microstructures of both base metal (BM) and simulated HAZ specimens were observed with an optical microscopy (CK40M, Olympus, Tokyo, Japan) using samples whose surfaces were mechanically-polished and then etched with 3% nital. To identify the strength variation within the CGHAZs, Vickers hardness tests were performed at maximum load of 9.8 N with HMV-2 equipment (Shimadzu, Tokyo, Japan).

Electrochemical hydrogen charging was performed on a potentiostat (HA-151A, Hokuto Denko, Tokyo, Japan) using a 0.25 g/L As_2O_3 in a 1 N H_2SO_4 solution. The As_2O_3 was added for enhancing the hydrogen atom permeation and avoiding their recombination. While platinum and the specimen were used as anode and cathode respectively, hydrogen was charged into specimens at an electrochemical current density of 100 mA/cm^2 for 24 h at room temperature (RT).

Standard Charpy V-notch impact tests were performed on both hydrogen (H)-free and H-charged samples at RT and −40 °C using a 500 J-capacity tester (CI-500D, TTM, Tokyo, Japan). The H-charged samples were tested within 20 min after charging. Fracture surfaces were observed through a scanning electron microscopy (SEM, JCM 5700, JEOL Ltd, Tokyo, Japan).

3. Results and Discussion

Generally, the CGHAZs are roughly subdivided into four characteristic zones according to the peak temperature of subsequent thermal cycles in a multi-pass welding procedure [7,8]; (1) the unaltered (UA) CGHAZ, the region reheated above the specific temperature of grain growth or not reheated at all, (2) the super-critically reheated (SCR) CGHAZ, the region reheated to the temperature just above A_{C3}, (3) the inter-critically reheated (IC) CGHAZ, the region reheated to austenite/ferrite two phase region ($A_{C1} < T_{P2} < A_{C3}$), and (4) the sub-critically reheated (SC) CGHAZ, the region for $T_{P2} < A_{C1}$. Among them, the SCR CGHAZ is often treated as fine-grained HAZ (FGHAZ) due to its recrystallized fine grains. The present study was focused on the UA, SCR, and IC CGHAZ. The SC CGHAZ is not considered here because its properties are expected to be similar (or a little superior) to the UA CGHAZ due to its low peak temperature and tempering effects [8]. For the HAZ simulations, the A_{C1} and A_{C3} were determined as ~713 and ~860 °C, respectively, based on Andrews' empirical formula [11]. Thus, in this study, the T_{P2} was taken as 1350, 1050, and 800 °C for the UA, SCR, and IC CGHAZ, respectively.

Vickers hardness values and representative microstructures of three simulated CGHAZs are presented in Fig. 1. While microstructure of BM mainly consists of ferrites with very small fraction of pearlite (not shown here), both the UA and IC CGHAZ exhibit bainitic laths within coarsened prior austenite grain. The SCR CGHAZ has very fine matrix (and thus often called FGHAZ, as mentioned before) since its second thermal cycle above A_{C3} induced grain refinement by recrystallization. Although the UA and IC CGHAZ have coarsened structure, they show higher hardness than the SCR CGHAZ and BM, which may be due to their bainitic structure having very high matrix strength.

Fig. 2a summarizes the room-temperature Charpy impact test results of both the H-free and H-charged CGHAZs as a function of T_{P2}. Before hydrogen charging, the SCR CGHAZ shows the highest impact energy (~429 ± 30 J) that is even higher than that of the BM (~400 ± 40 J). In contrast, the H-free UA and IC CGHAZ exhibit much lower average impact energy with a larger scatter (~83 ± 51 J and ~ 106 ± 65 J, respectively)

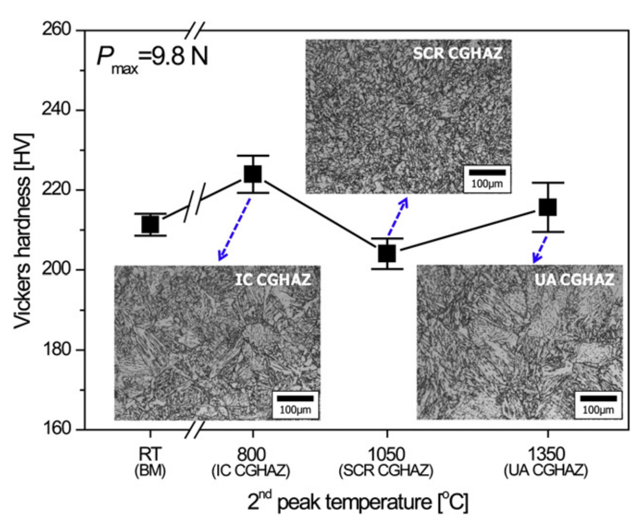

Fig. 1 – Microstructures and hardness of the three simulated CGHAZs.

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