

In-situ 3D Analysis of Hydrogen Induced Cracking Behavior in Linepipe Steels to Investigate Sour Environmental Severity

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ABSTRACT

Hydrogen Induced Cracking (HIC) is a major issue of the linepipe exposed to sour environment. NACE MR0175 / ISO15156 classifies the severity of sour conditions as a function of pH and H₂S partial pressure, based on Sulfide Stress Cracking (SSC) susceptibility of low carbon steel from experiments. Recently, the sour environmental severity diagrams for HIC and SSC were proposed based on the equal hydrogen concentration concept. From this concept, HIC behavior corresponds well with the steady-state hydrogen permeability, while SSC behavior is close related to the maximum hydrogen permeability. In this study, HIC behavior was closely investigated using the phased-array ultrasonic testing techniques to create three-dimensional (3D) images of cracks. HIC initiation and propagation behaviors were clearly visualized and investigated. It was found that HIC susceptibility was strongly affected by the hydrogen diffusion and corresponded with the steady value of hydrogen permeability.

Key words: sour environment, HIC, hydrogen permeability, in-situ, ultrasonic inspection, linepipe

INTRODUCTION

In the oil and gas industry, oil country tubular goods (OCTG) or linepipes are exposed to wet H₂S environments (Sour environments) in some cases. The presence of H₂S in the sour environment enhances hydrogen entry into the steel due to catalytic action of H₂S. The absorbed hydrogen enhanced by H₂S affects hydrogen embrittlement. Hydrogen Induced Cracking (HIC) and Sulfide Stress Cracking (SSC) forms of environmentally assisted cracking caused by hydrogen embrittlement.

To evaluate susceptibility for these environmentally assisted cracking, NACE TM0284 solution A, B, C and 5 wt% NaCl + 0.93 N CH₃COOH/CH₃COONa solution (HLP solution), which comprises sodium chloride, acetic acid, and/or HCl dissolved in distilled water with various pH are used.¹ However, various H₂S partial pressures and pH values exist in actual oil and gas wells around the world. Therefore, corrosion tests often require solutions with H₂S partial pressure and pH close to those of actual sour environments as the European Federation of Corrosion (EFC)⁽¹⁾ specifications.² The severity of sour environment determined in accordance with EFC 16 and NACE MR0175 / ISO15156 for carbon and low-alloy steels has been proposed as indicated in Figure 1.^{2,3} Material properties such as yield strength and hardness are required depending on the severity of sour environment. The critical line of each region which was indicated as regions 0, 1, 2 and 3 in Figure 1 based on experimental SSC results of API grade N80 and P110.⁴ P110 is not applicable to the region below the upper diagonal (region 2 and 3) and N80 is not applicable to the region below the lower diagonal (region 3). Some researchers have investigated the susceptibility of HIC under various sour environments.^{5,6,7,8} However, HIC susceptibility does not necessarily correspond to SSC susceptibility. Hence, hydrogen assisted environmental cracking (HIC and SSC) domains as functions of pH and H₂S partial pressure have not been fully generalized.

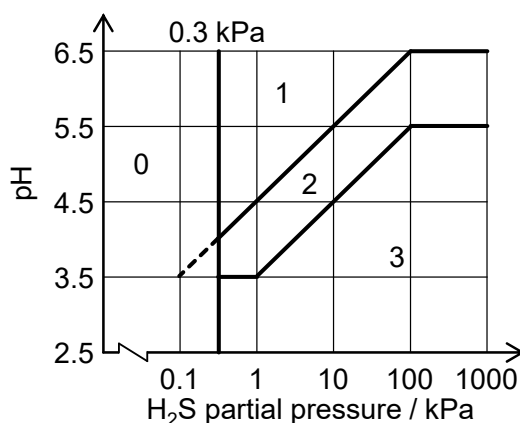


Figure 1: Regions of environmental severity with respect to the SSC of carbon and low-alloy steels.³

Many researchers have investigated the behavior of hydrogen entry for various sour environments.^{5,6,9,10,11,12,13,14} HIC or SSC, which is a type of hydrogen embrittlement, occurs if hydrogen concentration entering into steel from the environment. Thus the critical line of each region should be determined based on hydrogen concentration in steel from the environment. In a precise sense, the local hydrogen concentration at the crack tip of the HIC or SSC should be evaluated. However, it is so difficult to evaluate the local hydrogen concentration because the local hydrogen concentration is affected by both environmental factors and material factors, such as a hydrogen diffusion coefficient of steels.

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The extensive research on the relationship between the severity of sour environment differing in pH and H₂S partial pressure and HIC/SSC susceptibility was investigated and severity diagrams for HIC and SSC were proposed based on the equal hydrogen concentration concept.¹⁵ From this concept, HIC behavior corresponds well with the steady-state hydrogen permeability, while SSC behavior is closely related to the maximum hydrogen permeability. In this study, HIC behavior was precisely investigated using the phased-array ultrasonic testing techniques to create three-dimensional (3D) images of cracks. HIC initiation and propagation behaviors were clearly visualized and investigated. Hydrogen diffusion behavior was also investigated by Finite Element Analysis (FEA). It was found that HIC susceptibility was strongly affected by the hydrogen diffusion and corresponded well with the steady value of hydrogen permeability.

EXPERIMENTAL PROCEDURE

Materials

In this study, low-alloy carbon steel plates for non-sour service were prepared. Chemical composition of the test steels are shown in Table 1. Steels A and B were produced in an actual plate mill for commercial linepipe products. Steel A meets the requirements of strength grade X80 and is expected to have higher HIC susceptibility than Steel B of X65. Steel H was produced in the laboratory. These steel plates were produced by Thermo-Mechanical Control Process (TMCP). Figure 2 shows optical micrographs of the test steels. All steels used in this study have a bainitic microstructure. Steels A and B were used to investigate the HIC behavior by an in-situ ultrasonic inspection. Steel H was used to investigate the hydrogen permeation behavior by an electro-chemical hydrogen permeation test.

Table 1: Strength grade and chemical compositions of test steels.

Steel	Grade	Chemical compositions /mass%					
		C	Si	Mn	P	S	others
A	API X80	0.05	0.1	1.6	<0.006	<0.001	Cu, Ni,Cr, Mo, Nb, Ti,Ca
B	API X65	0.06	0.1	1.9	<0.006	<0.001	
H	API X65	0.05	0.2	1.5	<0.006	<0.001	Nb, Ti, Al

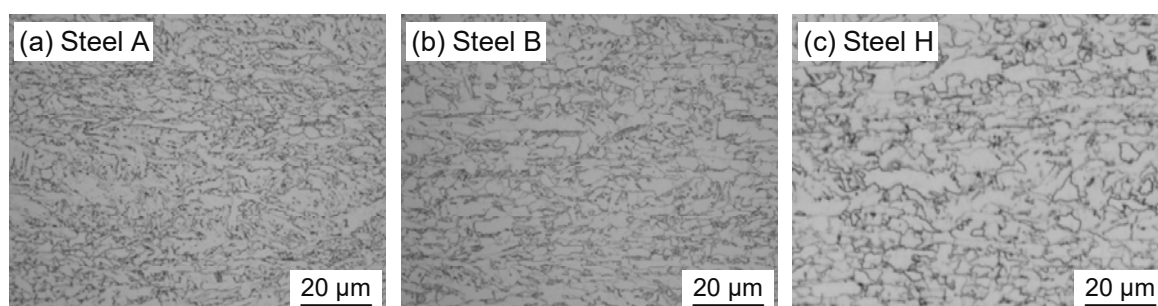


Figure 2: Optical microphotographs of (a) Steel A, (b) Steel B and (c) Steel H.

In-situ Ultrasonic Inspection during HIC Test

In-situ ultrasonic inspection during HIC test was established to investigate HIC initiation and propagation behavior.¹⁶ Figure 3 shows a schematic illustration of the test apparatus. The array probes were immersed in the test solution and the specimens were uniaxially moved to inspect the cracks. Ultrasonic scanning was carried out every 30 minutes in this study. As shown in Figure 4 (a), a 3D image was reconstructed to visualize the temporal change of the crack distribution. The projection image of the crack from the normal direction to the measured surface was also reconstructed, and

cracking area ratio (CAR) was defined as the ratio of the crack area to specimen surface area as shown in Figure 4 (b).

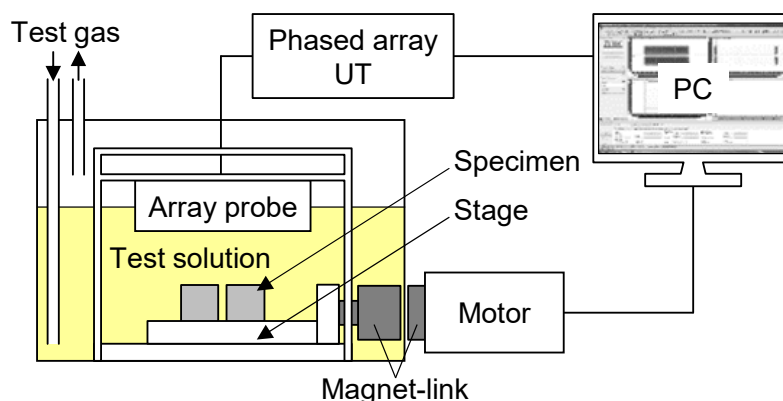


Figure 3: Schematic illustration of in-situ ultrasonic inspection system during HIC test.

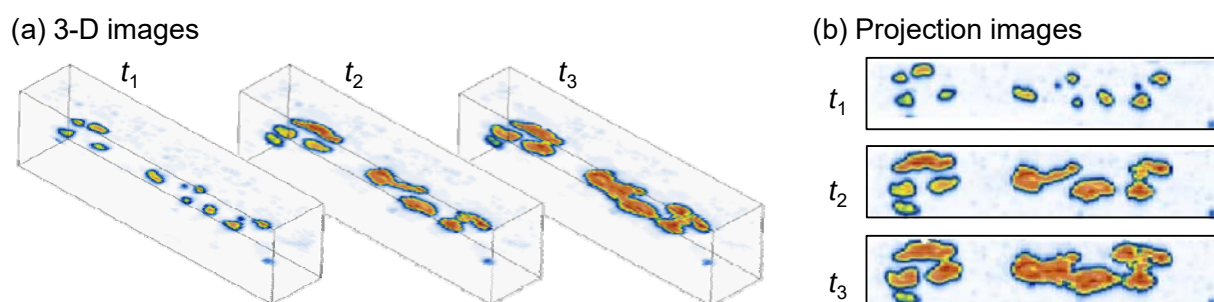


Figure 4: Examples of (a) 3D image and (b) projection image of crack distribution.

The HIC test procedure basically complied with NACE TM0284-2016.¹ Specimens were taken from a middle-thickness of steels A and B to include center-segregation zones with higher HIC susceptibility. The dimensions of the specimens were 24 mm in thickness, 20 mm in width and 100 mm in length. All specimen surfaces were wet-polished with # 600 emery paper and degreased prior to immersion in the test solution. In this study, NACE solution A (5 wt% NaCl + 0.5 wt% CH₃COOH, initial pH 2.7) and HLP solution were prepared. The HLP solution was developed by High-strength Line Pipe (HLP) Committee of The Iron and Steel Institute of Japan (ISIJ)⁽²⁾ and was designed as an optional solution alternative to Solution C for mildly sour HIC test in NACE TM0284-2016.¹⁷ The initial pH of the HLP solution was adjusted to 3.0 by mixing acetic acid and sodium acetate in an appropriate volume ratio. H₂S/CO₂ mixed gas with the H₂S partial pressure of 100 kPa and 10 kPa was introduced into the solution A and HLP solution after purging N₂ gas, respectively.

Hydrogen Permeation Method

Hydrogen permeability and corrosion potential were measured using an electrochemical hydrogen permeation method using a specimen with nickel-plating on the anode-side (hydrogen-withdraw side) surface.¹⁸ The ISIJ/HLP committee has established a highly reproducible hydrogen permeation test method in sour environment.¹⁴ The specimen surface was wet-polished with # 1000 emery paper and electro-polished. Ni metallic coating was plated on the hydrogen-withdraw side of the specimen in the

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Watts bath.¹⁸ Figure 5 shows a schematic illustration of the test apparatus of this experiment. The hydrogen-withdraw side was filled with 0.1 N NaOH solution and the potential was set to +0.148 V (vs Ag/AgCl). After the current measured in the hydrogen-withdraw side decreased less than 0.1 $\mu\text{A}/\text{cm}^2$ due to passivation of Ni coating, the hydrogen-entry side was filled with the test solution.

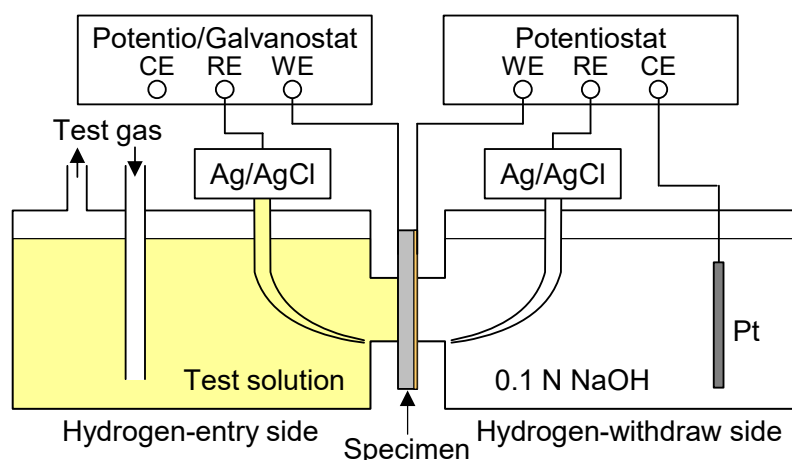


Figure 5: Schematic illustration of hydrogen permeation system.

In this study, hydrogen permeability was investigated at different H_2S partial pressure. The specimens with a thickness of 1 mm were taken from a quarter-thickness of Steel H. The HLP solution was used as a test solution. The initial pH of the solution was 3.0 by mixing acetic acid and sodium acetate in an appropriate volume ratio. Before pouring it into the hydrogen-entry side, the test solution was pre-purged and saturated with a $\text{H}_2\text{S}/\text{CO}_2$ mixed gas. The H_2S partial pressures of the mixed gas were 100 kPa and 10 kPa. The $\text{H}_2\text{S}/\text{CO}_2$ mixed gas was also bubbled into the test solution during the hydrogen permeation test. The permeation current was observed during the test, and the maximum permeation current density (J_{MAX}) and the steady-state permeation current density at 48 hours after the start of the test ($J_{48\text{h}}$) were measured. Hydrogen permeability, described as the rate of hydrogen permeation through the steel multiplied by the specimen thickness, was calculated.^{19,20}

$$[\text{Per}] = J \times L \quad (1)$$

where Per is the hydrogen permeability, J is the permeation current density, and L is the specimen thickness through which hydrogen permeates.

A hydrogen permeation tests by cathodic charging were also carried out to determine the apparent hydrogen coefficients and solubilities of steels A and B. The test apparatus was a Devanathan type cell as shown in Figure 5. The platinum counter electrode was used for both hydrogen-entry side and hydrogen-withdraw side. The hydrogen-withdraw side was filled with 0.1 N NaOH solution and the potential was set to +0.148 V (vs Ag/AgCl). After the current measured on the hydrogen-withdraw side reached 0.1 $\mu\text{A}/\text{cm}^2$ or less, the hydrogen-entry side was filled with 0.5 wt% NaCl solution. Thereafter, a constant current density was applied to generate a hydrogen absorption reaction at the specimen surface. At this time, it was reported that the permeation current in the 1st build-up was unstable due to inactive surface, and could be slower than the 2nd and/or subsequent build-ups due to irreversible trapping of non-diffusible hydrogen. Therefore, the 2nd build-up was used in this study. The following equation holds in the transition period of hydrogen permeation curve.^{21,22}

$$\log(\sqrt{t} \cdot J_t) = \log \frac{2CF\sqrt{D}}{\sqrt{\pi}} - \frac{\log e \cdot L^2}{4D} \cdot \frac{1}{t} \quad (2)$$