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Effect of tensile stress in elastic and plastic range on hydrogen permeation of high-strength steel in sour environment

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ABSTRACT

The new hydrogen permeation technique developed for the electrochemical permeation method makes it possible to evaluate the effect of applied tensile stress in both elastic and plastic range on hydrogen permeation behavior in high-strength steels. This study suggests that the iron sulfide film acts as sites for hydrogen reduction reaction and that the applied elastic stress weakens the stability of the sulfide film due to lots of cracks forming in the local sulfur deficient region, resulting in increase in both the anodic dissolution and hydrogen reduction reaction. In addition, the effect of applied stress in plastic range on hydrogen permeation behavior was clearly evaluated by excluding other side effects such as the reduced rate of hydrogen oxidation caused by the disruption of the Pd layer under stress. Furthermore, by evaluating hydrogen permeation behavior in the transient range from elastic to plastic stress, the effect of newly generated dislocation on the permeation current was accurately investigated without any side effect.

1. Introduction

The hydrogen assisted cracking (HAC) problem has been one of the major technical issues for development of high-strength metals and alloys since the possibility of HAC problem increases proportionally with the level of their strength [1–5]. Depending on the source of hydrogen and level of stress, HAC problem occurs in different manner. When the linepipe steel is used for transportation of oil and gas containing H_2S, it may experience either the hydrogen induced cracking (HIC) under no applied stress [6,7] or the sulfide stress cracking (SSC) under applied tensile stress or residual stress [8,9]. In this case, the source of hydrogen is the atomic hydrogen which is reduced from H^+ ion dissociated from H_2S. A similar type of either HIC or SSC may also occur in the high-strength pressure vessel steel when used in sour environment containing H_2S [10]. The high-strength bolt steel and automotive steel may fail by the hydrogen delayed cracking when used in ambient atmosphere. In such cases, the source of hydrogen is the atomic hydrogen which is reduced from the neutral water condensed on the steel surface [3,4]. The hydrogen delayed cracking behavior is also affected by the level of plastic deformation applied during manufacturing process [11].

To understand clearly HAC problem, the combined effect of hydrogen and stress should be evaluated together. The accurate hydrogen permeation data provide the critical information to mitigate HAC problems occurring in various industries. The Devanathan–Stachurski cell [12] is widely used to measure electrochemically the hydrogen permeation through the metallic membrane. The successful use of this electrochemical method requires the palladium (Pd) plating on the hydrogen exit side of the metallic membrane. For the high-strength metals and alloys, however, the hydrogen permeation data evaluated electrochemically under applied stress have been controversial due to pre-mature failure of Pd-plated layer when the applied stress level is higher than the yield strength (YS) of Pd (<205 MPa) [13–15]. To overcome such technical problem, we have developed a new and simple permeation technique which can be used with no problem of pre-mature failure of the Pd coating layer under both elastic and plastic stress of the base metal [16].

Even though the hydrogen permeation data still lead to controversy due to pre-mature failure of the Pd layer, much attention has been given to the combined effect of the tensile stress and environment of the hydrogen source on the hydrogen permeation current. Some researchers have evaluated the permeation current under elastic tensile stress and reported that the applied stress results in a slight increase in the permeation rate due to the lattice expansion [17–19].

In most permeation tests, the hydrogen charging has been carried out under cathodic polarization condition to avoid formation of

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Table 1
Chemical composition of the tested steel.

Chemical composition (wt%)							
C	Mn	P	S	Al	Cr + Mo	Cu + Ni	Ti + Nb
0.1–0.2	1.16	0.003	<0.002	0.043	0.1	0.199	0.117

any corrosion product acting as a barrier against the hydrogen diffusion. The accurate diffusion parameters can be determined by the cathodic charging method, however, such data are not enough to understand the actual HAC problems occurring in the industry. For the high-strength steel used in sour environment such as linepipe steel and pressure vessel steel, the iron sulfide film naturally forms on the steel surface and it affects critically the hydrogen evolution rate and consequently the hydrogen diffusion behavior. Since the iron sulfides (FeS_{1-x}, FeS and FeS_2) have a good electrical conductivity [20,21], they can act as a site for the hydrogen reduction reaction ($H^+ + e^- \rightarrow H$), suggesting that the hydrogen diffusion and permeation behavior can also be influenced by the characteristics of the sulfide film. Although the effect of alloying element on the hydrogen permeation has been discussed in terms of change in the characteristics of the sulfide film [22], no one has paid particular attention to the effect of elastic tensile stress on the characteristics of the sulfide film and subsequent permeation current.

Furthermore, a number of considerable efforts have been directed to evaluate the effect of applied tensile stress in plastic range on the permeation current. Especially, in body-centered cubic materials under the tensile stress in plastic range, a significant decrease in the permeation current has been observed by several researchers and it has been ascribed to the enhanced hydrogen trapping by dislocation generated under plastic stress [17,23,24]. However, it is difficult to assure that the considerably slower permeation rate is caused solely by the generated dislocation. This means that the decrease in the permeation current can also be ascribed to the combined effect of the reduced rate of hydrogen oxidation ($H \rightarrow H^+ + e^-$) by disruption of the Pd layer and accelerated dissolution of the steel substrate and subsequent decrease in current by passivation. Although our previous investigation has proven that applied tensile stress greater than YS of Pd affects the stability of the Pd layer, resulting in unstable background current [16], there is no clear experimental evidence for the change in permeation current caused by the side effects developed by disruption of the Pd layer under tensile load, such as the reduced rate of hydrogen oxidation.

In the present study, the effect of the applied tensile stress in both elastic and plastic range was investigated with respect to the hydrogen permeation behavior and characteristics of the sulfide film by utilizing the new permeation technique reported in our previous work [16]. In addition, for a clear understanding of the relationship between the dislocation generated by tensile stress in plastic range of the steel and hydrogen atom diffusion in the steel, hydrogen permeation behavior in the transient range from elastic to plastic stress was systematically evaluated by excluding other side effects such as the reduced rate of hydrogen oxidation caused by disruption of the Pd layer under the tensile stress.

2. Experimental

2.1. Test materials and specimen preparation

The test materials used in this study was 64 mm thick pressure vessel steel (ASTM A516) plate. Table 1 lists the chemical composition of the steel specimens. The test specimens were austenitized by heating to 910 °C for 30 min and subsequently cooled to room temperature by using a mixture of oil and water. Since the center region of the thick steel plate contains some low temperature transformation microstructures such as bainite and martensite/austenite (M/A) constituents caused by the center segregation [25], a tempering heat treatment was additionally carried out at 650 °C for 100 min to toughen the test specimen which has a YS of 338 MPa. The sheet-type tensile specimens, with 1 mm thickness, were then machined from the center region of the steel plate and the detailed specification of the specimen is shown in Fig. 1.

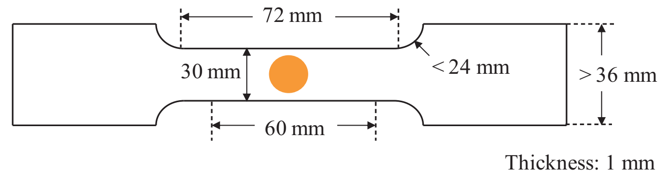

Fig. 1. Specification of designed tensile specimen.

The test specimens were ground to 2000 grit paper and polished with 0.25 μm diamond suspension. And then, the specimens were degreased with ethanol and etched with 5% nital solution. The microstructure was examined with optical microscope (OM) and field emission-scanning electron microscope (FE-SEM).

2.2. Electrochemical hydrogen permeation test under tensile stress

The hydrogen permeation test cell equipped with a constant load testing device is shown in Fig. 2. According to the aforementioned new and simple method of Pd plating technique, the hydrogen permeation behavior was evaluated in various ranges of elastic and plastic stress without the rupture of the Pd film. The

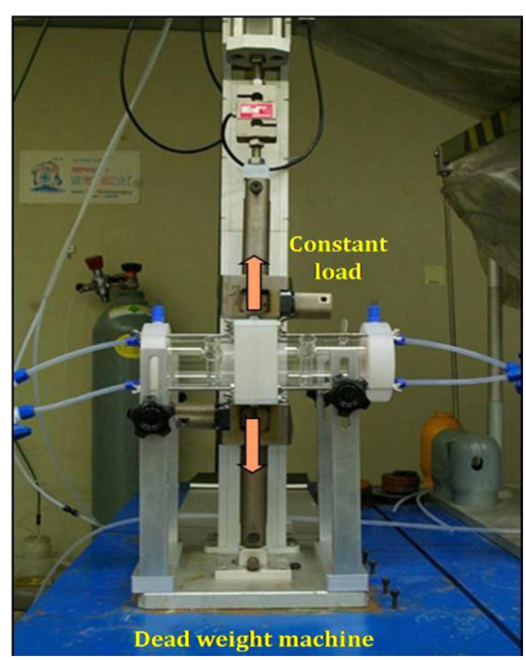

Fig. 2. A modified electrochemical permeation cell equipped with a constant load testing device.