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Stress-dependent hardening-to-softening transition of hydrogen effects in nanoindentation of a linepipe steel

Dong-Hyun Lee [a], *Jung-A Lee* [a], *Moo-Young Seok* [a], *Un Bong Baek* [b], *Seung Hoon Nahm* [b], *Jae-il Jang* [a,*]

[a] *Division of Materials Science and Engineering, Hanyang University, Seoul 133-791, South Korea*
[b] *Division of Industrial Metrology, Korea Research Institute of Standards and Science, Daejeon 305-340, South Korea*

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ABSTRACT

We explored the influences of hydrogen on small-scale strength of a linepipe steel through nanoindentation experiments with four pyramidal indenters. Interestingly, a transition from hydrogen-induced hardening to softening was observed as indenter sharpness increases. The transition was analyzed based on the enhancement in hydrogen's elastic shielding effects for a sharper indenter, which could be indirectly evidenced by the stress effects on indentation pile-up, dislocation density, and rate dependency of hardness.

1. Introduction

Recently, the influence of hydrogen on the mechanical behavior of linepipe steels has gathered much research interests [1–5]. The motivations are two-fold. First, natural gas transmission pipelines can be exposed to hydrogen atmosphere (especially in the sour environment) [1,2,4]; i.e., hydrogen can be absorbed either from hydrogen sulfide (H_2S) included in natural gas or from localized corrosion and cathodic protection (in the buried pipeline). Second, such hydrogen research may be required for preparing the upcoming era of "hydrogen economy". It has been reported that, transporting gaseous hydrogen (in a form of either pure hydrogen or a blend of natural gas and hydrogen) through the existing natural gas pipelines may be the most cost-effective and energy-efficient way to transport large amounts of hydrogen over long distance [3]. Thus, understanding of the hydrogen effects on the mechanical performance of linepipe steels is essential for ensuring the safety and integrity of the hydrogen pipeline system.

Although it is well accepted that sufficient hydrogen deteriorates ductility and toughness in a linepipe steel [4,5] (which is often termed as hydrogen embrittlement [6]), there are contradictory aspects in the hydrogen effects on the plastic flow and dislocation mobility in a steel [6,7]; that is, sometimes hydrogen induces hardening [8,9] and sometimes softening [10,11], depending on the materials, hydrogen

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[*] *Corresponding author.*
E-mail address: jijang@hanyang.ac.kr (J.-i. Jang).

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concentration, and temperature [6,7]. For example, in austenitic stainless steels, many articles reported the hydrogen-induced increase in macroscopic yield strength [8,9], whereas hydrogen-induced decrease in microscopic strength was also observed through in-situ transmission electron microscopy observation [12] or internal friction measurement [13]. Matsui et al. [10] also reported that both H-induced hardening and softening can occur in high purity iron at low temperatures (170–300 K).

On the other hand, nanoindentation tests have been extensively performed for evaluating the hydrogen (H)-induced change in local mechanical properties [14–16] since they have strong advantage in addition to simple and easy procedure; i.e., nanoindentation tests require only small volumes of test material, making it possible to obtain statistically meaningful data from an identical sample. It is noteworthy that most of the nanoindentation studies to date have reported H-induced hardening [14–16] (that are explained by general solid solution hardening mechanisms such as dislocation dragging or pinning [16] and H-enhanced slip planarity [15]), but almost no nanoindentation research has been performed on the H-induced softening.

Plastic deformation mechanisms of metallic materials are known to be strongly dependent on the level of applied stresses and plastic strains, which can be also true for the H-affected deformation in hydrogenated samples. In this regard, it is somewhat interesting to note that (to the best of our knowledge) almost no efforts have been made on the issue. This is mainly due to the difficulty in changing stresses during nanoindentation tests with a typically-used three-sided pyramidal indenter (such as Berkovich indenter); i.e., from a continuum plasticity concept, the stresses and strains underneath a sharp indenter are unique and independent of indentation load or displacement due to the so-called geometrical self-similarity of the tip. This difficulty may be overcome by varying the sharpness of the pyramidal indenter which is characterized by its centerline-to-face angle, ψ. Generally, sharper indenters induce larger stresses and strains in the material due to the larger volume of material that is displaced [17–20]. Thus, indentations made with different ψ lead to different level of stresses and plastic strains, allowing a systematic evaluation of the effects of the imposed stresses and strains.

In this work, we systematically analyzed the influence of external stresses and plastic strains on the H-induced change in small-scale strength through a series of nanoindentation experiments with four triangular pyramidal indenters having different ψ from 35.3° to 75°. The purpose of this letter is to analyze our interesting observations that, with increasing the indentation stresses and strains, there is a clear transition from H-induced hardening to H-induced softening.

2. Experimental

The material under investigation was a commercial grade API X70 steel, one of the most popular natural gas pipeline steel, whose nominal chemical composition (in wt.%) is 0.071C–0.25Si–1.55Mn–0.25Cu–0.2Ni–0.04Nb–0.03V–0.015Ti–0.03Al and (balance) Fe. The microstructure of the steel mainly consists of ferrite with very small fraction of pearlite. Surfaces of the specimens were ground with 2000-grit SiC paper and then electrolytically polished at 40 V for 60 s in a solution of 80% Ethanol, 14% distilled water, and 6% perchloric acid (according to ASTM E1558-09) in order to remove any hardened surface layer produced during grinding.

Electrochemical hydrogen charging was performed at room temperature with a potentiostat (HA-151A, Hokuto Denko, Tokyo, Japan) using a 0.25 g/L As_2O_3 in a 1 N H_2SO_4

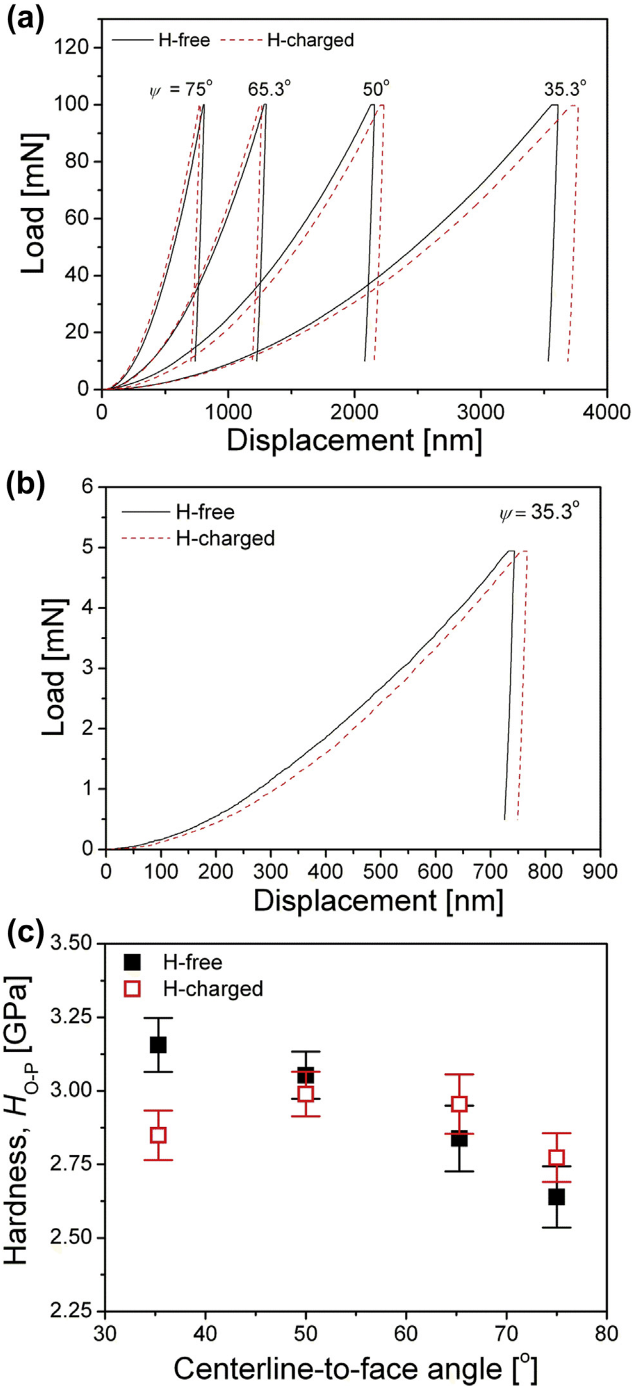

Fig. 1 – Influence of indenter angle on the nanoindentation results (obtained under an indentation strain rate of 0.05/s); (a) typical load-displacement curves conducted at P_{max} = 100 mN with various indenters; (b) the curves for ψ = 35.3° and P_{max} = 5 mN; (c) nanoindentation hardness.