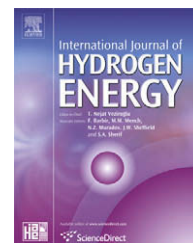


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# Hydrogen transport in solution-treated and pre-strained austenitic stainless steels and its role in hydrogen-enhanced fatigue crack growth

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## ABSTRACT

Hydrogen solubility and diffusion in Type 304, 316L and 310S austenitic stainless steels exposed to high-pressure hydrogen gas has been investigated. The effects of absorbed hydrogen and strain-induced martensite on fatigue crack growth behaviour of the former two steels have also been measured. In the pressure range 10–84 MPa, the hydrogen permeation of the stainless steels could be successfully quantified using Sieverts' law modified by using hydrogen fugacity and Fick's law. For the austenitic stainless steels, hydrogen diffusivity was enhanced with an increase in strain-induced martensite. The introduction of dislocation and other lattice defects by pre-straining increased the hydrogen concentration of the austenite, without affecting diffusivity. It has been shown that the coupled effect of strain-induced martensite and exposure to hydrogen increased the growth rate of fatigue cracks.

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## 1. Introduction

In the future, hydrogen-based fuel cell systems are expected to play an increasing important role in reducing society's dependency on fossil fuels and significantly reducing carbon dioxide emissions. Stainless steels are commonly used for fuel cell system components such as liners of high-pressure vessels of 70 MPa class, piping, valves, springs,

bearings, etc. These components are directly exposed to hydrogen gas environments and are subjected to cyclic loading.

It has been reported that hydrogen easily penetrates metallic materials and that dissolved hydrogen degrades the mechanical properties of the materials. Loss of ductility for metals subjected to hydrogen has received significant attention. Vennet and Ansell [1], and Benson et al. [2] reported that

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hydrogen embrittlement occurred in austenitic stainless steels strained while exposed to high-pressure hydrogen gas environments. Birnbaum et al. used a hydrogen environmental cell TEM to study *in-situ* the effect of hydrogen on the behaviour of numerous materials [3]. Based on these experimental results, Birnbaum and Sofronis [4] proposed a mechanism of hydrogen-enhanced localized plasticity (HELP). Even though the detrimental influence of hydrogen has been studied primarily with respect to static properties, in many applications the degradation of fatigue properties of metallic materials due to hydrogen exposure is more critical. In previous investigations [5–8], hydrogen was artificially (electro-chemically) charged into specimens of several candidate materials for hydrogen fuel systems. It was revealed that hydrogen influences the slip band morphology and the fatigue crack growth behaviour.

In medium carbon steels subjected to cyclic deformation [7], slip bands in the uncharged materials were widely distributed among many grains and occurred on many slip systems within a single grain. In the hydrogen-charged materials, the slip bands were more discrete and localized inside ferrite grains. Corresponding to the slip localization due to hydrogen charge and cyclic loading, the overall strain amplitude was reduced in the cyclic stress–strain hysteresis loops of the hydrogen-charged specimens.

In austenitic stainless steels [8], hydrogen increased the fatigue crack growth rates and enhanced slip localization near the fatigue crack tip. Type 304 and Type 316 stainless steels, in which strain-induced martensite is easily produced, exhibited greater increase in crack growth rate due to hydrogen diffusion and localized slip than did a Type 316L stainless steel which has significantly greater austenitic stability. In the case where a significant portion of martensite phase was induced by pre-straining the Type 316L stainless steel at 203 K, a significant increase in the fatigue crack growth rate due to hydrogen was measured. Thus, the coupled effect of martensite phase and hydrogen exposure plays an important role in fatigue crack growth. Since the diffusion rate of hydrogen through martensite is significantly higher than that through austenite, even a small amount of strain-induced martensite in the neighbourhood of a crack tip can act as “hydrogen diffusion highway” [6]. However, the degree of hydrogen penetration into metallic materials exposed to high-pressure hydrogen gas and how the dissolved hydrogen influences the fatigue strength properties is still unknown. San Marchi et al. [9] have recently reviewed the permeability, solubility and diffusivity of hydrogen in austenitic stainless steels at high gas pressures. According to this report, many of the data were typically measured by hydrogen permeation

method at pressures near-ambient and temperatures greater than 473 K. The hydrogen permeation method provides the solubility of hydrogen within the austenite lattice. Thus, the solubility does not reflect the effect of hydrogen trapped with lattice defects, such as dislocation, vacancy, and grain boundary. On the fatigue mechanism, however, the concentration of hydrogen including both the lattice hydrogen and the trapped hydrogen is of importance. To investigate the weak trapping effects on the solubility and diffusivity of austenitic stainless steels, the thermal desorption spectrometry (TDS) is more effective in the measurement of the hydrogen concentration. The objectives of this study are to investigate the role of martensite and lattice defects such as dislocation produced by pre-straining in the mechanism of hydrogen transport in austenitic stainless steels, in high-pressure hydrogen gas environments and to clarify its effect on hydrogen-induced fatigue crack growth.

## 2. Materials and experimental methods

The materials used in this study were Type 304 (JIS-SUS304), Type 316L (JIS-SUS316L) and Type 310S (JIS-SUS310S) austenitic stainless steels. Materials were received in the form of 30 mm thick plates. The chemical compositions and the characteristic temperatures of strain-induced martensitic transformation,  $M_{d30}$ , are given in Table 1. The  $M_{d30}$  is defined as the temperature at which 50% of the microstructure is transformed into martensite by a true tensile strain of 0.30. The  $M_{d30}$  values were calculated with Angel's equation [10],

$$M_{d30}(\text{K}) = 413 - 462([\text{C}] + [\text{N}]) - 9.2[\text{Si}] - 8.1[\text{Mn}] - 13.7[\text{Cr}] - 9.5[\text{Ni}] - 18.5[\text{Mo}] + 273.15.$$

Measured Vickers hardness,  $HV$ , of the three steels is listed in Table 2. These values represent the average of 20 measurements with a force of 9.8 N. Hydrogen contents were measured by thermal desorption spectrometry (TDS). Type 304 stainless steel was solution treated at 1353 K for 3 min, Type 316L stainless steel was solution treated at 1373 K for 3 min and Type 310S stainless steel was solution treated at 1403 K for 15 min.

To investigate the effect of pre-strain on hydrogen absorption, the initially thick plates were cold-rolled to predetermined thicknesses representing pre-strain (true strain) values  $-0.16$ ,  $-0.36$ ,  $-0.60$  and  $-0.92$ . Cold rolling was accomplished using multiple passes corresponding to a plate thickness reduction of about 1% per pass. Plastic forming temperature is known to influence strain-induced martensitic transformation. Therefore, plate temperatures were monitored after each cold-rolling

**Table 1 – Chemical compositions and characteristic Temperatures of strain-induced martensitic transformation,  $M_{d30}$ , of austenitic stainless steels used in this study.**

	(Mass%, *mass ppm)									$M_{d30}$ [K]
	C	Si	Mn	P	S	Ni	Cr	Mo	H*	
JIS-SUS304	0.05	0.58	1.24	0.025	0.003	8.09	18.54	–	2.2	316.8
JIS-SUS316L	0.010	0.53	0.77	0.023	0.001	12.13	17.16	2.86	1.5	267.2
JIS-SUS310S	0.04	0.42	0.38	0.019	<0.001	20.31	24.69	–	4.7	129.5

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