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Effect of aluminium on hydrogen permeation of high-manganese twinning-induced plasticity steel

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To investigate the hydrogen mobility in high-manganese twinning-induced plasticity (TWIP) steels, high-temperature permeation tests were conducted. They showed that the addition of aluminium to TWIP steel decreases the permeability and diffusivity of hydrogen and increases the solubility, which implies that one of the effects of aluminium in suppressing the hydrogen-induced mechanical degradation comes from a reduction in hydrogen mobility. The findings on diffusivity and solubility are consistent with an earlier calculation that considered aluminium–hydrogen interactions.

Keywords: Hydrogen; Permeation test; TWIP steel; Delayed fracture

Twinning-induced plasticity (TWIP) steels have been the subject of intensive studies in the last decade thanks to the excellent combination of strength and elongation that they offer [1–3]. Such alloys are expected to contribute to weight reduction in automobiles, but delayed fracture has been a hurdle to industrial exploitation. Delayed fracture is the failure of a component after the forming or welding process, which leaves residual stress. This type of mechanical degradation is thought to originate from the presence of hydrogen. In practice, delayed fracture in TWIP steel is known to be mitigated by aluminium addition. There have been many studies on the underlying mechanism of the aluminium effect [4–11]. Most of them concern the change in microstructural evolution caused by aluminium, such as the suppression of martensite formation and twinning during deformation. It is also suggested that the magnitude of the residual stress can be reduced by decreasing the flow stress with aluminium. There is no doubt that all of these phenomena contribute to the aluminium effect. Nevertheless, it is worth investigating the influence of aluminium on the intrinsic behaviour of hydrogen, which has attracted little attention to date. Indeed, assuming 1.5 wt.% of aluminium – the typical concentration in TWIP steels – the site occupancy of aluminium in the lattice point is around 3.1%, which is sufficiently large to induce atomic interaction with hydrogen.

One of the most reliable ways to investigate the aluminium effect on hydrogen mobility is the permeation test, which gives information on a number of parameters, such as diffusivity, permeability and solubility [12]. However, there is considerable difficulty in maintaining consistent experimental conditions for the duration of the test of TWIP steels. This is because the diffusion of hydrogen in austenite is very slow at ambient temperature, being some six orders of magnitude slower than in ferrite. It therefore takes an extremely long time for hydrogen to penetrate through the steel [13]. To finish the permeation test in a reasonable time, the sample thickness is reduced or the diffusion of hydrogen is accelerated by increasing the test temperature. Since the penetration depth of hydrogen is reported to be only several tens of micrometers under typical electrolytic charging conditions at ambient temperature [14,15], the thickness of the steel membrane has to be comparable to that. However, with such a thin sample, a small variation in sample thickness may have an unexpected influence on the permeation result, therefore, high-temperature permeation tests are conducted in this study. This approach has not been attempted before for TWIP steel, particularly to investigate the role of aluminium in preventing the mechanical degradation caused by hydrogen.

The investigated alloys are austenitic TWIP steels containing 0.6C–18Mn wt.%, with different aluminium contents of 0 (TW1) or 1.5 wt.% Al (TW2), respectively. The detailed chemical compositions are listed in Table 1. Cold-rolled 1.4 mm thick sheets were heat-treated at 900 °C for 15 min at ± 10 °C s^{-1} heating and cooling rates.

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Table 1. Chemical composition of the tested materials (wt.%).

	C	Mn	Al
TW1	0.60	18.3	0.001
TW2	0.58	18.4	1.48

The annealed samples were ground up to 1200 grit SiC paper and polished with 0.25 μm diamond suspension for microstructural characterisation using field-emission scanning electron microscopy and orientation imaging. For hydrogen permeation measurements, the mechanically polished sheets were prepared as disc-shaped membranes with a thickness of 1.4 mm and a diameter of 20 mm.

The diffusion behaviour of hydrogen was examined using a hydrogen-isotope permeation measurement system (HPMS) that was designed and constructed in-house. Details of the HPMS are found in the literature [16–18]. Hydrogen permeation tests were conducted at 400, 500, 550 and 600 °C, with heating and cooling rates of 10 °C min^{-1}, and the charging pressure of hydrogen was fixed at 1 bar. The hydrogen flux through the steel membrane was measured as a function of time until the flux reached a steady state, from which the permeability, Φ (mol m^{-1} s^{-1} $Pa^{-0.5}$) and diffusivity, D (m^2 s^{-1}) were determined, assuming Fick's laws as well as Sievert's law. The solubility of hydrogen in the alloys, S (mol m^{-3} $Pa^{-0.5}$), was calculated directly from the following relationship [17–20]:

$$S = \Phi/D \tag{1}$$

Typical initial microstructures are shown in Figure 1, taken from the normal direction of the rolled sheet. Orientation images indicate that the alloys are composed of only austenite with average grain sizes of 11.5 ± 1.1 and 11.3 ± 2.8 μm in TW1 and TW2, respectively. There is no significant difference in microstructure due to the aluminium addition. It is noted that the microstructures after the permeation tests do not show any sign of grain growth. This is because the initial microstructures are fully annealed by heat treatment at 900 °C for 15 min, so that grain growth hardly occurs with further exposure to the permeation temperature in range of 400–600 °C.

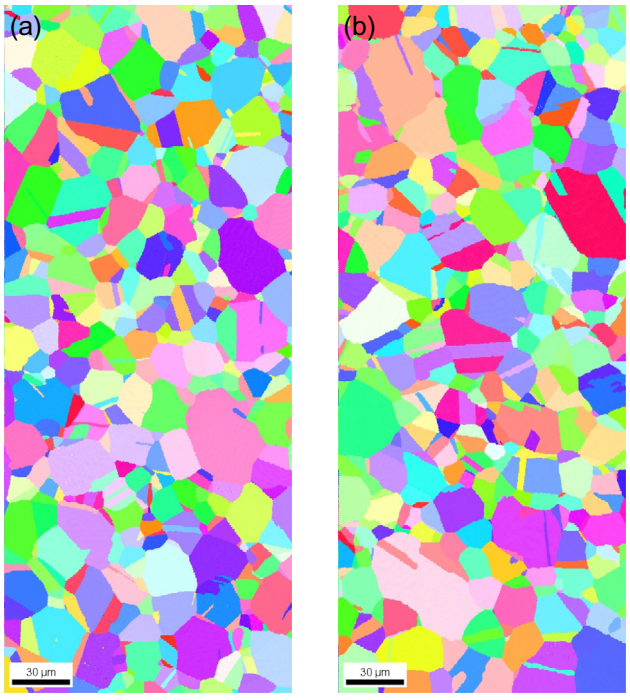

Figure 1. Typical EBSD orientation images of annealed (a) TW1 and (b) TW2 alloys.

Figure 2a and b show the hydrogen permeation flux through the alloys at 400, 500, 550 and 600 °C. The saturated hydrogen flux becomes higher as the test temperature increases in both alloys. In addition, at the same temperature, the saturated flux through TW2 alloy is lower than that through TW1. The fact that the saturated flux is proportional to the permeability implies that aluminium addition reduces the hydrogen permeability in the TWIP steel. Figure 3 shows the hydrogen permeability determined from the following relationship between the permeability and the hydrogen flux [18–20]:

$$J = \frac{n}{A} = \frac{\Phi \cdot P_{H_2}^{0.5}}{d} \tag{2}$$

where J is the permeation flux, n is the rate of permeation flow, A is the exposed area of the sample, d is the thickness of the sample, Φ is the permeability and P_{H_2} is the hydrogen pressure on the charging side. The dotted lines are plotted using Arrhenius fitting, which gives

$$\Phi(\mathrm{TW1}) = 1.87 \times 10^{-7} \exp(-63.9 \times 10^3/RT)$$

$$\Phi(\mathrm{TW2}) = 1.56 \times 10^{-7} \exp(-64.2 \times 10^3/RT)$$

where Φ (mol m^{-1} s^{-1} $Pa^{-0.5}$) is the hydrogen permeability, R (J mol^{-1} K^{-1}) is the gas constant, and T is the absolute temperature. As expected from the steady-state flux, the hydrogen permeability is lower in the TW2 alloy, which contains aluminium, at all test temperatures. The hydrogen permeabilities extrapolated to ambient temperature are 1.19×10^{-18} mol m^{-1} s^{-1} $Pa^{-0.5}$ for TW1 alloy and 8.59×10^{-19} mol m^{-1} s^{-1} $Pa^{-0.5}$ for TW2, which indicates a 28% decrease in the permeability with the presence of aluminium. The diffusivity of hydrogen in the alloys is also estimated from the hydrogen flux. The total amount of hydrogen that has permeated through the alloy, $Q(t)$, is obtained by integrating the permeation flux with time:

$$Q(t) = \int_0^t J(t)Adt \tag{3}$$

The time lag, t_0, is determined by line fitting on the linear region of the $Q(t)$, t_0 being the time when the fitting line intersects the time axis [18,20,21]. The hydrogen diffusivity, D, is then obtained by applying following equation [18,20,21]:

$$D = \frac{d^2}{6t_0} \tag{4}$$

The diffusivity of hydrogen is shown in Figure 4. The Arrhenius plots assuming equivalent activation energy are given as well. Similar to the permeability, the presence of aluminium lowers the diffusivity of hydrogen at all test temperatures. By extrapolation, the diffusivity of hydrogen at room temperature is 7.11×10^{-15} m^2 s^{-1} in TW1 and 4.62×10^{-15} m^2 s^{-1} in TW2, respectively, indicating a decrease of diffusivity by 35% with aluminium addition. This is comparable to a previous calculation using first principles indicating a 56% decrease in hydrogen diffusivity