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Effect of hydrogen on mechanical degradation and fatigue in 7075 Aluminium alloy with in-situ hydrogenation

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Abstract

The present study investigated the effect of hydrogen on the mechanical degradation and the fatigue crack growth rate of 7075 T6 Aluminium alloy by in-situ cathodic hydrogen charging. The effect of strain rates on tensile deformation in air and by in-situ hydrogen charging was studied. The fatigue crack growth tests were carried out at constant stress intensity factor ranges (ΔKs). The effects of different parameters, like stress intensity factor range (ΔK), stress ratio (R), and frequency have been investigated. The fatigue crack growth rate was found to increase in hydrogenating environment compared to the crack growth rate in air at different stress ratios. The effect of hydrogen on the fatigue crack growth rate was more at lower ΔK in comparison to higher ΔK. Fracture surfaces and crack morphologies were studied using scanning electron microscopy analysis.

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1. Introduction

High strength Aluminium alloys of 7xxx series have been found extensive use in the aerospace industry, owing to their high specific strength levels. The susceptibility of Aluminium alloys to environmentally assisted cracking is directly proportional to their strength levels. Since 7075 Aluminium alloy is used in aircraft industry due to its high strength, environmental degradation is generally observed. It is well known that hydrogen degrades mechanical properties of metals, such as ductility, fracture toughness, fatigue resistance, etc. This phenomenon is termed “hydrogen embrittlement”. The role of hydrogen in material degradation of Aluminium alloy has been studied since hydrogen embrittlement was suggested as one of the dominant mechanisms of their stress corrosion cracking. Due to the complex mechanical, metallurgical and environmental issues involved in synergistic corrosion-fatigue process, the failure stresses and failure times in a corrosive environment are lower than those in a non-corrosive environment.

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[1]. Takano [2] studied the effect of hydrogen on the mechanical properties of 7075 Aluminium alloy and hydrogen diffusivity at 318K using the electrochemical cathode charge method. Watson, et al. [3] studied the effect of cathodic hydrogen charging on the mechanical properties of pure Aluminium and they found that the cathodic charging reduced the ductility and increased the yield and tensile stresses of pure Aluminium. Research work has also been carried out on hydrogen-environment embrittlement (HEE) response of a high-tensile strength 7075 Aluminium alloy subjected to slow-strain rate tensile (SSRT) tests in humid air [4-8].

In studies on environments, the environmental issues of importance to crack growth are anodic dissolution, crack closure, adsorption of deleterious species and hydrogen enhanced cracking or hydrogen embrittlement [9-10]. Wei and Simmons [11] attributed the crack growth enhancement in moisture primarily to the process of bulk hydrogen embrittlement. R.G. Song et al. [12] studied the mechanism of Mg and hydrogen induced grain boundary fracture on 7050 Aluminium alloy. The effects of microstructure and environment on the fatigue crack growth resistance of 7075 Aluminium alloy were studied by S. Suresh et al. [13]. Menan, et al. [14] investigated the influence of frequency and exposure to saline water (3.5% NaCl) on corrosion fatigue crack growth rate in 2024 Aluminium alloy. They observed that the corrosion fatigue crack growth rate in alternate immersion accelerated the crack propagation up to an order of magnitude in the loading range compared to the propagation in a permanent immersion.

The aim of this work was to investigate the effect of hydrogen on the mechanical properties of 7075 T6 Aluminium alloy by in-situ cathodic hydrogen charging with slow strain rate technique. A comparative study has been made on the stress-strain curve, time to failure at different strain rate ($10^{-5}$ s$^{-1}$ and $10^{-6}$ s$^{-1}$) in air and by in situ hydrogen charging. Furthermore, the effect of hydrogen on the fatigue crack growth rate (FCGR) was studied by in-situ cathodic hydrogen charging. The fatigue crack growth tests were carried out at constant stress intensity factor range ($\Delta K$). The effects of different parameters, like stress intensity factor range ($\Delta K$), stress ratio (R), and frequency have been investigated.

### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>$\Delta K$</td>
<td>Stress intensity factor range</td>
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<tr>
<td>R</td>
<td>Stress ratio</td>
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<tr>
<td>$da/dN$</td>
<td>Crack growth rate</td>
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</tbody>
</table>

2. **Experimental**

2.1. **Material and specimens**

The material used for this study was 7075 Aluminium alloy. Its chemical composition (in wt.%) is Zn 5.4, Mg 2.2, Cu 1.56, Fe 0.12, Cr 0.22, Mn 0.02, Si 0.04, Ti 0.04, and balance is Al. The cylindrical block of Aluminium alloy was forged, rolled and then used to prepare round tensile specimens and three point bend specimens. The round specimens with 28mm gauge length and 7mm diameter of gauge length were used. The tensile samples were used for in-situ hydrogen embrittlement studies. Three point bend specimens were used for fatigue crack growth studies. After sample making all the specimens were heat treated in peak-aged (T6) temper condition. For T6 temper, the specimens were first solution annealed at 480°C for 1h and then aged at 120°C for 24h.

2.2. **Mechanical degradation**

The round tensile samples have been used for tensile tests. The tensile test has been carried out in air at different strain rates ($10^{-5}$ s$^{-1}$ & $10^{-6}$ s$^{-1}$) to determine the mechanical properties of 7075 T6 Aluminium alloy. The tensile tests were also performed with in-situ hydrogen charging by varying cathodic charging current density. All tests were carried out in a Perspex cell attached to the lower ram of a servo-electric loading frame. For hydrogenating environment, 3.5 wt% NaCl solution was used. For hydrogen charging, only the gauge length of the specimen was exposed in solution and other portions of the specimen were covered with Teflon tape. The specimen was made the cathode and a platinum wire was used as anode in the electrolytic cell. The slow strain rate tensile tests were performed
at different cathodic charging current density for different strain rates.

After the tensile test, specimens were cleaned in an ultrasonic bath. The fracture surfaces of the tensile tested specimens were studied using scanning electron microscopy (SEM) to identify the mode of fracture.

2.3. Fatigue crack growth rate test

The fatigue crack growth tests were conducted using notched three point bend specimens with nominal dimensions of 100mmx20mmx10mm. Before conducting the fatigue tests, all specimens were pre cracked up to 2 mm in air. The fatigue crack growth tests were carried out under decreasing ΔK control at two stress ratios and at 1Hz loading frequency in air to assess the stress ratio effect in this ΔK regime. Subsequently the fatigue crack growth tests were carried out using the constant ΔK protocols at two stress ratios and at different frequencies in air and in in-situ hydrogenating environment. Hydrogenating environment was created by using 3.5% NaCl solution. The hydrogen was introduced by applying 1mA/cm$^2$ cathodic charging current density. All the fatigue crack growth tests were conducted at room temperature according to the ASTM standard E-647.

The fracture surface morphologies of fatigue crack growth were studied using scanning electron microscopy. The fractographs were taken along the direction of the crack growth. The chemical composition of particles found on the fracture surface was determined qualitatively using EDS (energy dispersive X-ray spectroscopy), attached with the scanning electron microscope.

3. Results and discussion

3.1. Mechanical degradation

The tensile deformation in air as well as in hydrogenating environment was studied at two strain rates (10$^{-5}$s$^{-1}$ and 10$^{-6}$ s$^{-1}$) and at different cathodic charging current densities. The presence of hydrogen reduces the mechanical properties of the material. The slow strain rate effect is also observed in hydrogenating environment. Figs. 1a and 1b show that the total elongation to failure and tensile strength in uncharged and hydrogen charging environment at two strain rates. These figures reveal that the tensile deformation decreases with progressive increase in cathodic charging current density. It is also evident that the hydrogen reduces the tensile deformation at a very slow strain rate of 10$^{-6}$s$^{-1}$ compared to strain rate of 10$^{-5}$s$^{-1}$. This decrease in deformation is due to the increase in hydrogen embrittlement, because at a strain rate of 10$^{-6}$s$^{-1}$, sufficient time is available for substantial hydrogen entry. However, at higher strain rate (10$^{-5}$s$^{-1}$) the enhancement in ductility (compared to air) was found only at the lowest charging current density ($I_c$=1mA/cm$^2$). The lowest charging current density corresponds to longer exposure time to the environment which is responsible for a protective (oxide/hydroxide) film formation. It is obvious that the presence of an oxide film will hinder hydrogen diffusion into the matrix. For higher current densities at higher strain rate (10$^{-5}$s$^{-1}$), there was a progressive loss in ductility. Moreover time seems to catalyze opposing influences on hydrogen embrittlement. At the slowest strain rate (10$^{-6}$s$^{-1}$), there is progressive hydrogen embrittlement with increasing cathodic charging current density. The time available at these strain rates is sufficient for film growth, but it is also sufficient for substantial hydrogen entry in spite of the film.
Fig. 2 provides the fractographs of the hydrogenated samples tested at a strain rate of $10^{-6}\text{s}^{-1}$ at cathodic charging current density of 2 mA/cm$^2$. The fracture surface (Fig. 2a) reveals that the crack initiated from the edge of the fracture surface. Large and long secondary cracks are often observed on the fracture surface (Figs. 2b & 2c). The periphery also shows flat facets (Figs. 2b & 2c) which are indicative of hydrogen embrittlement (HE) at the periphery. The observations of secondary cracks are similar to that reported by Takano [2] for both tests in the air as well as with cathodic charging for the same alloy used here.

3.2. Fatigue crack growth rate (FCGR) test in air & hydrogenating environment

The effect of hydrogen on the fatigue crack growth rate (FCGR) was studied by in-situ cathodic hydrogen charging. The fatigue crack growth tests were carried out using the constant $\Delta K$ protocols at two stress ratios and at different frequencies in air and in in-situ hydrogenating environment. Before performing the FCGR test at constant $\Delta K$, the FCGR tests were carried out in air with decreasing $\Delta K$ protocol and at two stress ratios (R), which is shown in Fig. 3. This figure reveals that at a higher stress ratio (R=0.5), the crack growth rate is greater than the lower stress ratio (R=0.1).

The constant $\Delta K$ tests were conducted in air as well as in hydrogen charging conditions with two different $\Delta K$s, ($\Delta K$=12MPa.m$^{0.5}$ and 15MPa.m$^{0.5}$), at two stress ratios (R=0.5 and 0.1) and at different loading frequencies. Figs. 4 and 5 show the FCGR test in air and hydrogenating environment at constant $\Delta K$ of 12MPa.m$^{0.5}$, at two stress ratios,
R = 0.5 and R = 0.1 respectively. These figures demonstrate that the crack growth rate of the hydrogenated samples increases compared to air, indicating hydrogen assisting the cracking process.

Fig. 3. Crack growth rate (da/dN) versus stress intensity factor range (ΔK) in air at different stress ratios (R) and at 1Hz loading frequency

Fig. 4 also reveals that at R = 0.5, the corrosion fatigue crack growth rate increases with decreasing loading frequency from 1Hz to 0.25Hz. Bonakadar et al. [9] discussed the effect of moisture on FCG to be strongly dependent on the loading frequency and water vapour pressure. The residence time for hydrogen increases with decrease in loading frequency, which would explain the higher crack growth at lower frequencies. The FCGR in air is much lower than in NaCl (Fig. 5) due to the absence of hydrogen. Fig. 5 also illustrates that at low R ratio, the crack growth rate increases with crack length in air, but in NaCl environment at the same frequency, the rate of crack growth slightly reduces after 14mm crack length due to the crack closure at the crack tip. It seems that the crack closure is due to the oxide film formation after long exposure time to the hydrogenating environment. It also appears that at low ΔK and lower stress ratio for longer exposure time, two possible phenomena are occurring during the crack growth. The hydrogen entry is enhanced at low ΔK while the formation of an oxide/hydroxide film hinders the hydrogen diffusion into the matrix.

Fig. 4. Crack growth rate (da/dN) with crack length in air and hydrogen charging (1mA/cm²) at different loading frequencies at ΔK = 12 MPa.m.0.5 and R = 0.5
3.2.1. Effect of stress ratios (R) on fatigue crack growth rate

The effect of stress ratio plays a significant role in the fatigue crack growth rate. In the present investigation the effect of R ratios was studied at two constant ΔK values. A comparison has been made on the effect of R ratios at ΔK=12 MPa.m0.5 and at 1Hz loading frequency, which is shown in Fig. 6. It is clear from the figure that hydrogen enhances the fatigue crack growth rate in both the stress ratios. The enhancement of the crack growth rate in hydrogenated samples compared to air is more at a lower stress ratio (R=0.1) than the higher stress ratio (R=0.5). An explanation for this “R-ratio” effect could not be developed. Fig. 7 shows the effect of R ratios at ΔK=15 MPa.m0.5 and 1Hz loading frequency. This figure also reveals that the FCGR was found to be substantially higher in hydrogenated samples in comparison to the crack growth rate in air at corresponding R ratios. Comparing the Figs. 6 and 7, at R=0.5, the enhancement of crack growth rate at both the ΔKs are not significantly changed, but at a lower R ratio (R=0.1), the enhancement of crack growth rate at low ΔK, (ΔK=12MPa.m0.5), is more compared to the higher ΔK, (ΔK=15MPa.m0.5). This indicates that the hydrogen influence was prominent only at low ΔK and low R ratio.

Fig. 5. Crack growth rate (da/dN) with crack length in air and hydrogen charging at ΔK=12 MPa.m0.5 and R=0.1, f=1Hz

Fig. 6. Crack growth rate with crack length as a function of R ratios at ΔK=12MPa.m0.5

Fig. 7. Crack growth rate with crack length as a function of R ratios at ΔK=15MPa.m0.5
3.2.2. Effect of ΔKs on fatigue crack growth rate

The effect of ΔKs was studied by normalization of the FCGR of hydrogenated samples to the FCGR of air tested samples. Figs. 8a and 8b show the effect of ΔKs at R=0.5 and R=0.1 at 1Hz frequency respectively. These figures illustrated that the normalized crack growth rate at low ΔK is greater compared to the higher ΔK value in both R ratios. The ΔK effect on the normalized crack growth of HSLA steel has been provided by A. Roy, et. al [15]. They also observed that the hydrogen effect was greatest for the lowest ΔK value and least for the highest ΔK value. They also found that the normalized crack growth rate increased with increase in crack length indicating a higher hydrogen activity at higher crack lengths. But in Fig. 8b it is found that at low ΔK and at a low stress ratio (R=0.1), the normalized crack growth rate decreases with increase in crack length. This decrease was due to the formation of oxide/hydroxide at the crack tip, leading to crack closure. The production of hydrogen results from the reaction of Aluminium and moisture resulting in elemental hydrogen and Aluminium oxide. Hydrogen diffuses into the crack tip and the grain boundaries where the embrittling reaction takes place, causing the crack advance.

3.2.3. Examination of fatigue fracture surface

The fracture surfaces of the 7075 T6 Aluminium alloy subjected to cathodic hydrogen charging are shown. Fig. 9 shows the fracture surface of low ΔK (ΔK=12MPa.m⁰.⁵) and at R=0.5 tested specimen. The fracture surfaces produced in 3.5% NaCl solution show a transgranular fracture along with striations and secondary cracks, especially at a high R ratio (Fig. 9). But at low R ratio, the crack growth rate slightly decreases after 14mm crack length, because some oxide/hydroxide formed on the fracture surface, which is marked in Fig. 10a. The marked area is shown in Fig. 10b. EDS confirms that this oxide/hydroxide layer is Aluminium oxide or Aluminium hydroxide (Fig. 10c).
4. Conclusions

The present study highlights the effect of strain rate on tensile deformation of 7075 T6 Aluminum alloy in air and by in situ hydrogen charging. Furthermore, the study was extended to the fatigue crack growth rate of same alloy for constant ΔK protocols with in-situ cathodic hydrogen charging. The following conclusions can be drawn from the above study.

- The hydrogen reduces the tensile deformation at a very slow strain rate of 10^-6 s^-1 in comparison to the strain rate of 10^-5 s^-1. The ductility decrease was found to increase with increase in cathodic charging current density.

- In the FCGR test, overall FCGR was found to be substantially higher in hydrogenated samples in comparison to the crack growth rate in air. An inverse frequency effect was observed, which is possibly related to the residence time of hydrogen in the samples.

- In the FCGR study of decreasing ΔK and constant ΔK tests, crack growth rate at higher stress ratio is more compared to the lower stress ratio both in air and hydrogenating environment respectively.

- At lower R ratio (R= 0.1), enhancement of crack growth rate at lower ΔK is more compared to the higher ΔK. This indicates that the hydrogen influence was prominent only at low ΔK and low R ratio.
The effect of hydrogen observed through normalized crack growth rates is found to be greatest for the lowest \( \Delta K \) value and least for the highest \( \Delta K \) value.

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References