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Effect of Strain Rate and Temperature on the Susceptibility of 304 Austenitic Stainless Steel to Hydrogen Embrittlement

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أثر الحرارة ومعدل الانفعال على قابلية الفولاذ المقاوم للتآكل نوع 304 للتصدع الهيدروجيني

فوزي الشاوش

عدد من عينات الشدّ المصنوعة من الفولاذ المقاوم للتآكل نوع 304 شحنت بالهيدروجين كهروكيميائياً في وسط مائي يحتوي على 1 عياري حمض الكبريتيك مضافاً إليه قليل من زرنيخ الصوديوم أثناء تعرضها لمعدلات إنفعال عند درجة حرارة الغرفة (22±2C°) وأخرى عالية (70±2C°). نتائج إختبارات الشد عند درجة حرارة الغرفة بيّنت حدوث إنخفاض كبير في لدونة الصلب عند معدلات إنفعال بطيئة، كما أن الفحص المجهري لأسطح الكسور لعينات الشد (Fracture Morphology) أوضح بأنها هشة وأن إنتشارها كان بين وعبر حبيبات الصلب.

عندما شحنت عينات الشد عند درجة حرارة عالية (70±2C°) حدث إنخفاض كبير في قابلية الصلب للتصدع الهيدروجيني حيث أن عينات الشد كانت لها لدونة أعلى من التي كانت لعينات الشد التي تم إختبارها عند درجة حرارة الغرفة، كما أن نسبة اللدونة بها تزداد بزيادة معدلات الانفعال. أوضح الفحص المجهري لأسطح الكسور بأنها شبه هشة وأن نسبة اللدونة بها تزداد عند معدلات الإنفعال السريعة والبطيئة جداً.

بصفة عامة نتائج إختبار الشد أكدت بأن قابلية الصلب المقاوم للتآكل للتصدع الهيدروجيني تعتمد كلياً على درجة الحرارة ومعدل الإنفعال (strain rate).

Abstract: *Cathodic charging of notched 304 austenitic stainless steel specimens was carried out in aqueous solution of 1N H_2SO_4 containing 250 mg/l $NaAsO_2$ at room temperature and 70±2°C while undergoing tensile strain over a wide range of crosshead speeds of 833 µm/s, 83 µm/s, 8.3 µm/s, 833 nm/s, 83 nm/s and 9.8 nm/s.*

Test at room temperature 22±2°C resulted in a marked reduction in the elongation to fracture ratio (Esol/Eair) with reduction of the crosshead speed. However, little reduction was observed for stress to fracture ratio (σsol/σair). Cleavage and intergranular where the predominant fracture mode when tests were carried out at a low crosshead speed. The extent of these modes of fracture was observed to increase with reducing of crosshead speed.

Cathodic charging of 304 austenitic stainless steel at 70±2°C caused less reduction in the elongation to fracture ratio compared to the tests carried out at room temperature. Consistent with the room temperature test results, the reduction in the elongation to fracture ratio was found to increase with reduction of the crosshead speed. However, restoration in the elongation to fracture ratio was exhibited by 304 austenitic stainless steel specimens tested at the lowest crosshead speed of 9.8 nm/s. These results are in good

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agreement with the finding that the hydrogen embrittlement is temperature and strain dependent.

Cleavage fracture associated with the plastic deformation was the predominant fracture mode exhibited by 304 austenitic stainless steel specimens tested at 70±2°C over low crosshead speed.

INTRODUCTION

Austenitic stainless steels were observed to have good resistance to hydrogen embrittlement and, therefore, they were frequently chosen as the suitable material in environments containing hydrogen. However, a number of studies have shown that austenitic stainless steels are susceptible to hydrogen embrittlement, though not to the extent of ferritic stainless steels. This owing to the differences in the solubilities and-diffusivities of hydrogen within these two alloys.[1]

For austenitic stainless steels, it has been established that the degree of embrittlement in tensile tests increases as the time of hydrogen charging increases and is dependent on strain rate and temperature[1]. The most severe embrittlement was observed to occur at low strain rates and at temperatures below or at room temperature[2]. Despite many previous studies on hydrogen assisted cracking in austenitic stainless steel, there are no reliable, consistent or uniform data about the the effect of hydrogen on the mechanical properties as functions of strain rate and temperature.

In the current work, specimens of 304 austenitic stainless steel were cathodically charged at current density of 0.4 mA/cm^2 in aqueous solution of 1N H_2SO_4 containing $NaAsO_2$ at room temperature and 70±2°C while undergoing tensile strain over a wide range of crosshead speeds. The effect of hydrogen on the mechanical properties (ductility and strength) and change of the fracture morphology for 304 austenitic stainless steel as function of temperature and strain rate (crosshead speed) will be presented and discussed.

EXPERIMENT

A commercial 304 austenitic stainless steel specimens with the required dimension was machined from 0.8 mm thick sheet supplied in the annealed condition. A 60°V notch were machined at the middle of all tested specimens in order to promote the initiation and propagation of single crack to enable convenient investigation of the tested specimens (Fig.1). In order to remove the stresses induced during specimens preparation, all machined specimens were given solution treatment at 1050°C for 1 hour. The chemical composition of the received sheet is shown in Table 1.

Table 1. Chemical composition (wt%) of 304 austenitic stainless steel specimen used throughout the work.

Materials	C	Mn	Si	P	S	Cr	Ni
304 S.S	0.04	1.42	0.32	0.024	0.002	18.36	8.73

The all tested specimens were cathodically charged at a current density of 0.4 mA/cm^2 in aqueous solution of 1N H_2SO_4 containing $NaAsO_2$ while undergoing tensile strain over a wide range of crosshead speeds of 833 µm/s, 83 µm/s, 8.3 µm/s, 833 nm/s, 83 nm/s and 9.8 nm/s at room temperature and 70±2°C. For comparison, the same number of specimens were tested in air at room temperature and 70±2°C (in oil). The degree of embrittlement for 304 austenitic stainless steel was evaluated by dividing the value of elongation {Esol} and stress {σsol} to fracture in solution over the elongation{Eair}and stress{σair} to fracture in air at eash used crosshead speed. The obtained values of Esol/Eair and σsol/σair are plotted versus the all-used crosshead speeds.

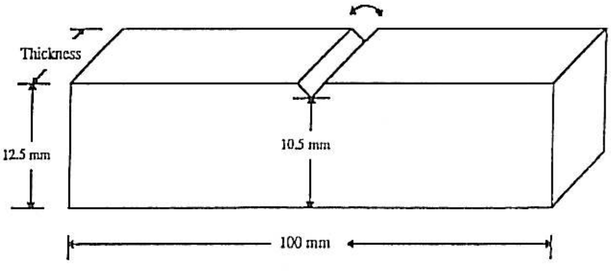

Fig. 1. Schematic representation showing the shape and dimensions (mm) of the type of specimen used in the hydrogen embrittlement test.

The resulted fracture surfaces were cleaned in acetone using ultrasonic and then dried. A scanning electron microscope (SEM) was then used to examine the changes in the fracture morphology as a result of hydrogen embrittlement.

RESULTS AND DISCUSSION

Mechanical Properties (ductility)

The results of elongation to fracture ratio for 304 austenitic stainless steel specimens tested at room temperature and 70±2°C are shown in Figure 2 as a function of crosshead speed. It can be observed that 304 austenitic stainless steel specimens is susceptible to hydrogen embrittlement when tested at room temperature and 70±2°C over low crosshead speeds. Testing at 70±2°C markedly reduce the susceptibility of 304 austenitic stainless steel to hydrogen embrittlement. However, little restoration in the embrittlement was observed when the test was carried out at 70±2°C over the lowest used crosshead speed of 83 nm/s (Fig. 2).

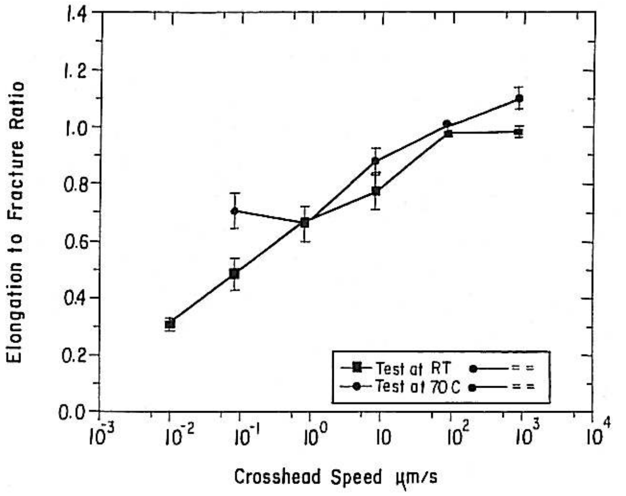

Fig. 2. Relationship between the elongation to fracture ratio and the crosshead speed for 304 austenitic stainless steel specimens fractured at room temperature and 70±2°C in aqueous solution of 1N H$_2$SO$_4$ containing NaAsO$_2$.

The results of stress to fracture ratio for 304 austenitic stainless steel specimens tested at room temperature and 70±2°C are shown in Figure 3. Consistent with the elongation ratio results, the stress ratio was observed to decrease with reduction in the crosshead speed. Since the resulting elongation ratio was observed to be more sensitive to hydrogen embrittlement than the stress ratio, during the discussion, the emphasis will be placed on the elongation to fracture ratio.

The reduction in the elongation to fracture ratio with lowering of crosshead speeds is consistent with the finding that the straining at slow crosshead speeds enhances the formation of high localized hydrogen concentration which are required to cause severe embrittlement[3,4]. The high potential sites for hydrogen concentration are dislocations, grain boundaries, and inclusions. Therefore, during straining high amounts of hydrogen is expected to be interacted or carried out by the mobile dislocations at the crack tip. These mobile dislocations are expected to tangle or pile up against any obstacles (*e.g* inclusions and grain boundaries) encountered its movement. Therefore, high hydrogen concentration, well in excess of the equilibrium content, can develop leading to severe embrittlement[3,5,6].

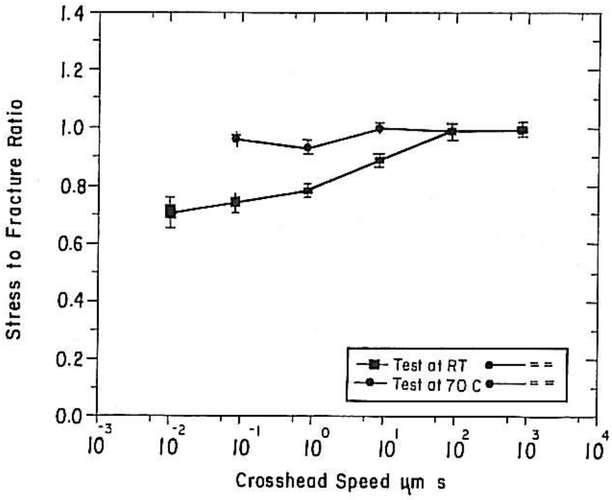

Fig. 3. Relationship between the stress to fracture ratio and the crosshead speed for 304 austenitic stainless steel specimens fractured at room temperature and 70±2°C in aqueous solution of 1NH$_2$SO$_4$ containing NaAsO$_2$.

When the test was carried out at 70±2°C$_9$, marked reduction in the susceptibility of 304 austenitic stainless steel to hydrogen embrittlement was observed. This can be attributed to the reduction in the localized hydrogen concentration in the test specimens when the test was carried out at 70±2°C[7,8]. As stated above, if the hydrogen embrittlement by dislocation is assumed to be vital to the hydrogen embrittlement mechanism, the hydrogen concentration on the mobile dislocations is thought to be low due to enhanced diffusion of hydrogen with increases in temperature[7,8]. In other words, increase in temperature results in destabilization of hydrogen laden with dislocation and homogenize its very rapid[3]. This, in turn, results in a low hydrogen concentration at potential sites such as inclusions and dislocations. at which dislocation pile-ups takes place Hence, less embrittlement is attained. High localized hydrogen concentration is required in order to cause severe embrittlement[3].

Louthan *et al.*,[3] have demonstrated the effect of temperature on the susceptibility to the hydrogen embrittlement as follows.

The amount of hydrogen transported (C_T) for any given amount of dislocation motion is dependent on $C_\perp$ through the following equation[3].

$$C_T \, \alpha \sigma_m \, C_\perp \, V \, b \, L \text{ ———— } (1)$$

Where C_T is the amount of hydrogen transported and ?m is the density of the mobile dislocation which have a hydrogen atmosphere and a burger vector, b. These dislocation are assumed to move with an average velocity of V, through an average distance L. Increasing the temperature tends to decrease the ratio of the hydrogen concentration on the dislocation core $C_\perp$ with respect to the lattice hydrogen concentration C_L. The relationship between the lattice hydrogen concentration of the dislocations is as follows[3].

$$C\perp = C_L \exp\left(-G_B / RT\right) \text{ ———— } (2)$$

Where G_B is the binding energy between the hydrogen and the dislocation.

The restoration in elongation reduction may be attributed to the diffusion-out of hydrogen from the mobile dislocations at the lowest used crosshead speed of 83 nm/s. This is obviously due to the faster diffusivity of hydrogen from dislocations compared with dislocation velocity. Therefore, little amounts of hydrogen are expected to be dumped at obstacles at which dislocations piled-up, hence, less embrittlement resulted. As stated above, high localized hydrogen is required in order to embrittle the 304 austenitic stainless steel[3].

FRACTOGRAPHY

The effect of hydrogen, strain rate and temperature on the changes of fracture morphology of 304 austenitic stainless steel was studied and the results can be given as follows. Generally four features were observed on the fracture surfaces of tested specimens. The all air tested specimens at room temperature and 70±2°C (oil) exhibited ductile fracture as shown in Figure 4. Marked change in the fracture morphology was observed when 304 specimens tested at room temperature over low crosshead speeds of 83 nm/s and 9.8 nm/s. Cleavage and faceted intergranular fracture were the predominant fracture modes (Fig. 5). As the crosshead speed was increased above 83 nm/s, the percentage of ductile fracture to brittle fracture (cleavage) starts to increase (Fig. 6), for specimen tested at crosshead speed of 833 nm/s.

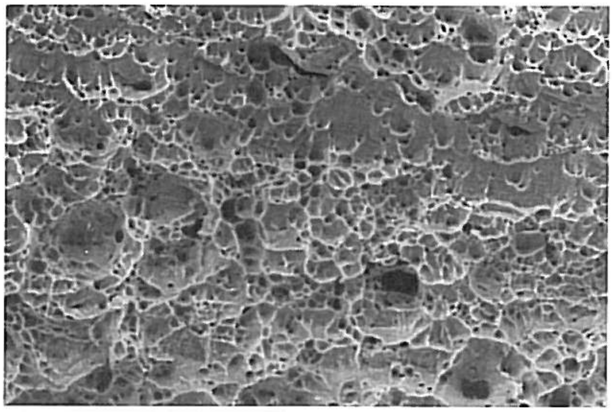

Fig. 4. Typical appearance of S.E.M. fractograph for a 304 austenitic stainless steel specimen strained in air over a wide range of crosshead speeds.

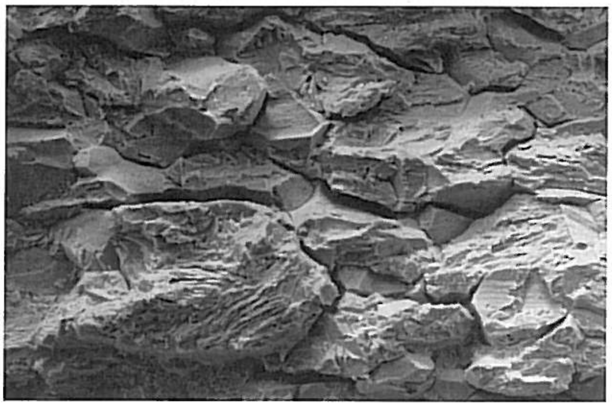

Fig. 5. S.E.M. fractograph of a 304 austenitic stainless steel specimen strained in solution at a crosshead speed of 9.8 nm/s. Fractograph shows both cleavage and faceted intergranular fracture.

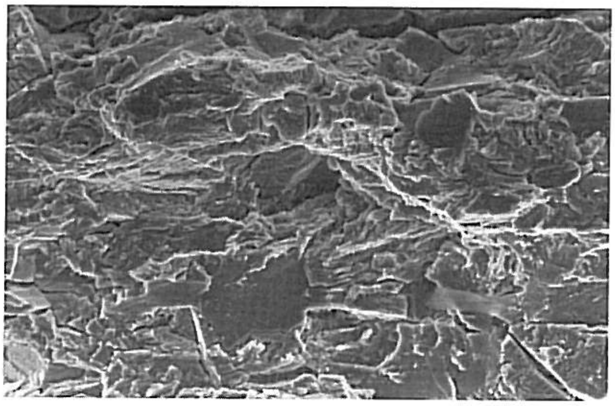

Fig. 6. S.E.M. fractograph for a 304 austenitic stainless steel specimen strained in solution at a crosshead speed of 833 nm/s. Fractograph shows that cleavage fracture is predominant. Little plastic deformation is visible.

The results of fracture morphology are consistent with the elongation ratio results when the test was carried out at 70±2°C. At a low crosshead speed of 83 nm/s, cleavage or quasi-cleavage like fractures were predominant (Fig.7) while above it, the percentage of ductile fracture to quasi cleavage fractures increases.

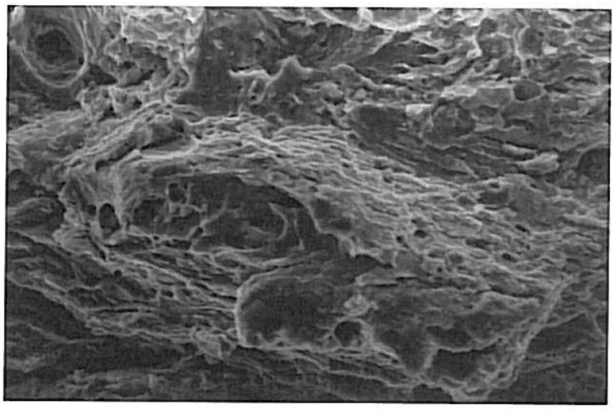

Fig. 7. S.E.M. fractograph for a 304 austenitic stainless steel specimen strained in solution at 70±2°C at the lowest crosshead speed of 83 nm/s. Fractograph shows quasi-cleavage fracture associated with some plastic deformation.

The observation of cleavage and intergranular fractures at the lowest crosshead speeds of 83 nm/s and 9.8 nm/s may be attributed to the high localized hydrogen concentration in the test specimens[3,9]. This may be owing to the pile-ups of mobile dislocations saturated with hydrogen against the obstacles (e.g inclusions, grain boundaries etc..) at the strained crack tip(3,9(. As stated above, high hydrogen conentration are required in order to embrittle the 304 austenitic stainless steel[3]. Increases of crosshead speed increase the percentage of ductile to cleavage fractures. This is mainly due to formation of low hydrogen concentration when the test was carried out at a high crosshead speed[4].

Cleavage fracture had been observed by many workers[10,11]. However, mixed mode, cleavage and faceted intergranular fracture of annealed 304 austenitic stainless steel, as observed in the current work, has not been previously shown.

Hydrogen charging at 70±2°C reduce the percentage of the ductile fracture with respect to the cleavage fracture owing to the low localized hydrogen concentration in the test specimens[3,5]. Generally tested at a temperature higher than room temperature, resulted in homogenization of hydrogen concentrated at obstacles present at crack tip such as dislocations and inclusion[3]. Therefore, low hydrogen concentration is attained, hence, less embrittlement was resulted.

CONCLUSIONS

1- The 304 austenitic stainless steel is significantly susceptible to hydrogen embrittlement when tested at room temperature over low crosshead speeds of 83 nm/s and 9.8 nm/s.

2- Marked decrease in the susceptibility of 304 austenitic stainless steel to hydrogen embrittlement was observed when the test was carried out at 70±2°C over the all-used crosshead speeds.

3- Transgranular and cleavage types of fracture modes were observed when 304 austenitic stainless steel was tested at room temperature over low crosshead speeds of 83 nm/s and 9.8 nm/s.

4- Transgranular cleavage, associated with plastic deformation, was the predominant fracture when 304 austenitic stainless steel was tested at 70±2°C over low crosshead speed of 83 nm/s.

5- The susceptibility of 304 austenitic stainless steel to hydrogen embrittlement was observed to decrease with the increase of test temperature and decrease of crosshead speed.

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