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# SUSCEPTIBILITY OF HYDROGEN EMBRITTLEMENT OF AL-ALLOY CIRCULAR BARS SUBMITTED TO COLD DRAWING

**Frederico Tavares**

**Gilberto Caldeira Mendes**

**Diego Camargos Sampaio**

Pontifical Catholic University of Minas Gerais, Department of Mechanical Engineering, R. Rio Comprido, 4580, Contagem (MG) 32010-025, Brazil.

[fredkarklin@hotmail.com](mailto:fredkarklin@hotmail.com), [gilberto@sirq.com.br](mailto:gilberto@sirq.com.br), [diegocamargos93@hotmail.com](mailto:diegocamargos93@hotmail.com)

**Diego Fonseca Silva**

**Carlos Trivellato de Carvalho Filho**

**Pedro Brito**

Pontifical Catholic University of Minas Gerais, Department of Mechanical Engineering, Av. Dom José Gaspar 500, Belo Horizonte (MG) 30535-901, Brazil.

[profdiegofsilva@gmail.com](mailto:profdiegofsilva@gmail.com), [carlos.trivellato.filho@gmail.com](mailto:carlos.trivellato.filho@gmail.com), [pbrito@pucminas.br](mailto:pbrito@pucminas.br)

**Abstract.** *In the present work, an analysis of the susceptibility to hydrogen embrittlement of AA6351 Al-alloy bars as a function of the reduction in the cold-drawing process was performed. The materials were initially annealed submitted to cold-work levels of 0.11, 0.18, 0.25, and 0.32% in order to introduce different levels of strain-hardening. The average hardness along the samples cross section varied between 82 and 107HV for the 0.11 and 0.32 reductions, respectively and similar hardness values were observed at the surface and interior of samples prepared from the same bars. Tensile test specimens were obtained from the cold-drawn bars were then prepared and the materials were submitted to tensile testing before and after hydrogen permeation performed by cathodic charging in a 0.6M NaCl solution. It was possible to notice a positive correlation between the amount of strain-hardening induced by cold-drawing and the susceptibility to hydrogen embrittlement. The samples submitted to the largest reductions in the drawing process exhibited a 0.80 loss in ductility and the fracture mode changed from ductile to brittle, with no signs of necking.*

**Keywords:** hydrogen embrittlement, cold drawing, fracture, ductility.

## 1. INTRODUCTION

Aluminum alloys are currently the non-ferrous metallic materials most widely employed. They exhibit low cost, good corrosion resistance, low density, as well as elevated mechanical strength and ductility. For these reasons, Al-alloys are used in various numerous applications, especially in the automotive and aerospace industries, where the constant drive for reducing structural weight creates incentives of employment of lightweight metals.

An important factor that affects the structural integrity of Al-alloy components is environmental embrittlement caused by hydrogen (Barnoush and Vehoff, 2008; Scully *et al.*, 2012). The incorporation of hydrogen in Al-alloys is known to take place in association with corrosion processes (Kamoutsi *et al.*, 2006). In aqueous environments, atomic hydrogen is generated by the following reaction (Young and Scully, 2002):



The atomic hydrogen produced at the surface of the alloy is then absorbed in the metal lattice and is trapped in the vicinities of crystal defects such as dislocations (Safyari *et al.*, 2021) or vacancies (Lu and Kaxiras, 2005).

Because of their versatility as structural materials, Al-alloy products are supplied in numerous conditions: as-cast, hot and cold-rolled sheets, extruded or cold-drawn bars or tubes, etc. Among these, cold-drawing is one of the main mechanical forming processes currently used for producing seamless tubes, and can be used in finishing passes after extrusion since it allows adequate dimensional accuracy and good surface finish (Béland *et al.*, 2011). Although cold-drawing does not in principle produce significant microstructural changes, it does lead to strain-hardening, with an increase in crystal defect density and a decrease in ductility.

Considering the importance of the interaction between hydrogen and a materials microstructure, in the present work an experimental investigation concerning the susceptibility to hydrogen embrittlement of Al-alloy AA6351 as a function of drawing reduction in cold drawing is presented. The alloy was selected for investigation because of its widespread use,

and it is expected that this study may contribute to the understanding of manufacturing and environment conditions on the mechanical behavior of a common engineering alloy.

## 2. EXPERIMENTAL PROCEDURE

The material used in the present work was an AA6351 Al-alloy circular bar, with 25.4 mm diameter initially received in the extruded condition. The chemical composition of the material was determined by mass spectroscopy and is presented in Table 1. The microstructure of the received materials revealed the presence of fine precipitates, as seen in Figure 1(a). In order to homogenize the initial materials, the samples were initially annealed at 600°C for 120 min and the resulting microstructure is shown in Figure 1(b). The microstructure of the AA6351 is composed of an Al-rich matrix and dispersed precipitates, which are highlighted in Figure 1(c). Chemical composition analysis performed by EDS suggested AlFeMnSi and AlMnSi precipitates, but the exact stoichiometry could not be inferred.

Table 1. Chemical composition of the AA6351 alloy (values in weight percent).

	Si (%)	Mn (%)	Ti (%)	Fe (%)	Zn (%)	Mg (%)	Cu (%)	Al (%)
AA6351	1.00	0.606	0.019	0.117	0.007	0.652	0.029	Bal.

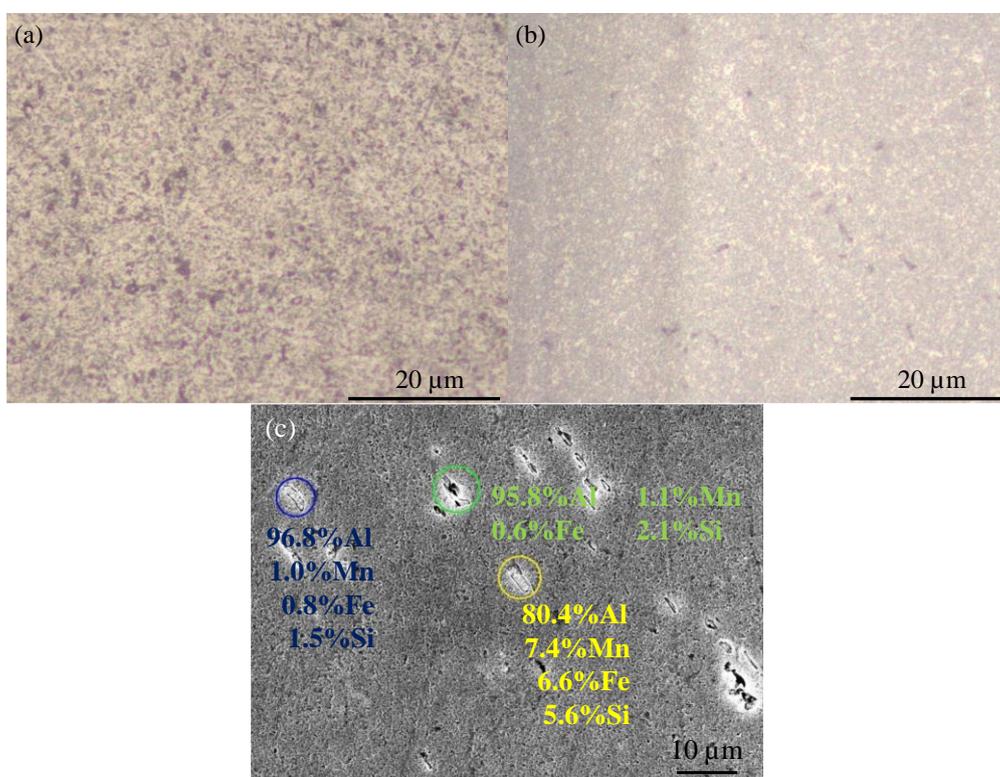


Figure 1. Microstructure of the as-received (a) and annealed (b) AA6351 Al-alloy. Optical microscopy, Keller etchant (2.5 ml HNO<sub>3</sub>, 1.5 ml HCl, 1.0 ml HF and 95 ml distilled water). Identification of precipitates by SEM/EDS (c).

After annealing, the 25.4 mm bars were cold-drawn and reduced to 24, 23, 22 and 21 mm final diameters (equivalent reductions of 0.11, 0.18, 0.25 and 0.32). The operations were performed in the drawing bench depicted in Figure 2, as in previous investigations (OLIVEIRA *et al.*, 2014; SILVA *et al.*, 2014). After each drawing pass, test specimens were taken for further analyses. Cross-section hardness measurements were performed using a HMV-1 Shimadzu microhardness tester with a 0.025 gf load and 20 s load time load in conformity with the ASTM E384-17 standard, and tensile test specimens were prepared for evaluating tensile strength, following ASTM E8-21 standard. The tensile testes were performed on pristine samples and samples exposed to hydrogen by cathodic polarization. The cathodic polarization was performed in a 0.6M NaCl solution containing 0.25g/l NaAsO<sub>2</sub> (Klimovicz and Latanision, 1978) with a current density of 15 mA/cm<sup>2</sup> for 4 hours.

## 3. RESULTS AND DISCUSSION

The results of hardness measurements performed along the cross-section of different cold-drawn specimens is presented in Figure 3, while average hardness values obtained after each drawing pass are shown in Figure 4. As expected,

the drawing process led to an increase in hardness of the AA6351 alloy, with average hardness values of  $82.0 \pm 0.8$ ,  $90.6 \pm 0.6$ ,  $97.7 \pm 1.5$  and  $107.2 \pm 2.0$  HV0.025 for reductions of 0.11, 0.18, 0.25 and 0.32, respectively. for pre-strain values of 0.0, 6.0 and 12.0%, respectively. The samples did not exhibit large variations in hardness over the cross-section, which can be seen by the relatively small data dispersion shown in Figure 3 and by the reduced standard deviation values reported in Figure 4. A linear relation between drawing reduction (R) and the average hardness (HV) was identified:

$$HV = 69.63 + 116.08R, \quad (2)$$



Figure 2. Drawing bench used for performing the cold-forming operations on the AA6351 Al-alloy bars.

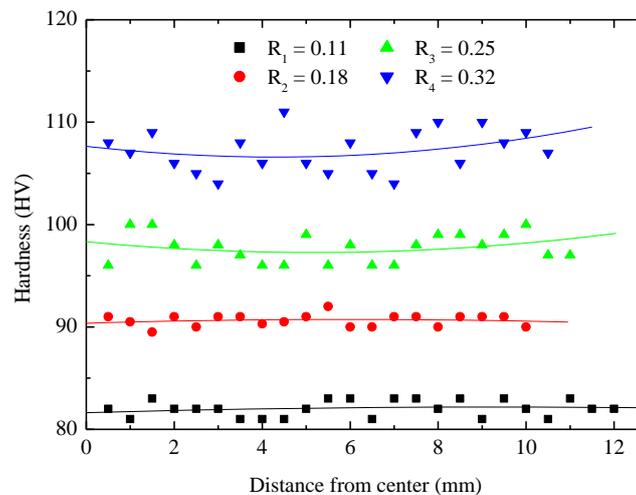


Figure 3. Cross-section radial hardness profiles for drawing reductions of 0.11, 0.18, 0.25 and 0.32.

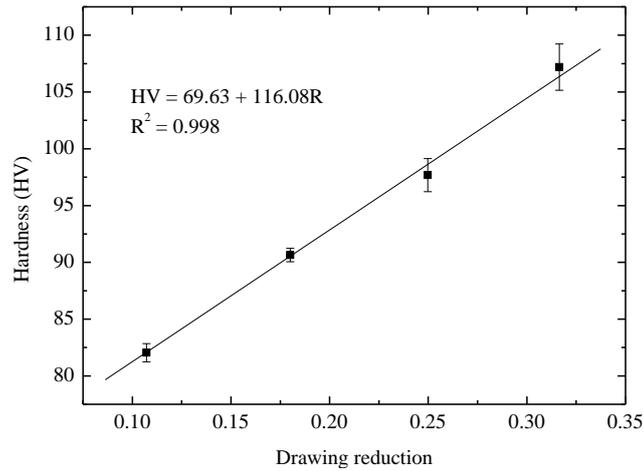


Figure 4. Average cross-section hardness as a function of drawing reduction (R).

After initial hardness evaluation, the materials were submitted to cathodic charging and tensile tests. The results obtained are presented in Figure 5(a-d) for the 0.11, 0.18, 0.25 and 0.32 drawing reductions, respectively, with and without hydrogen charging. It was possible to notice that in all cases, the presence of hydrogen was detrimental to the mechanical behavior of the tested materials, inducing significant reductions in ductility which is expected given the increase in dislocation density caused by the cold-working process. The loss of mechanical strength, given the observed increase in hardness registered in Figs. 3 and 4 is not, however, expected. In the present case, it is possible to speculate that surface residual stresses or surface integrity might have compromised the material ability to withstand uniaxial tensile loads, but further investigations would be required to properly account for the observed behavior.

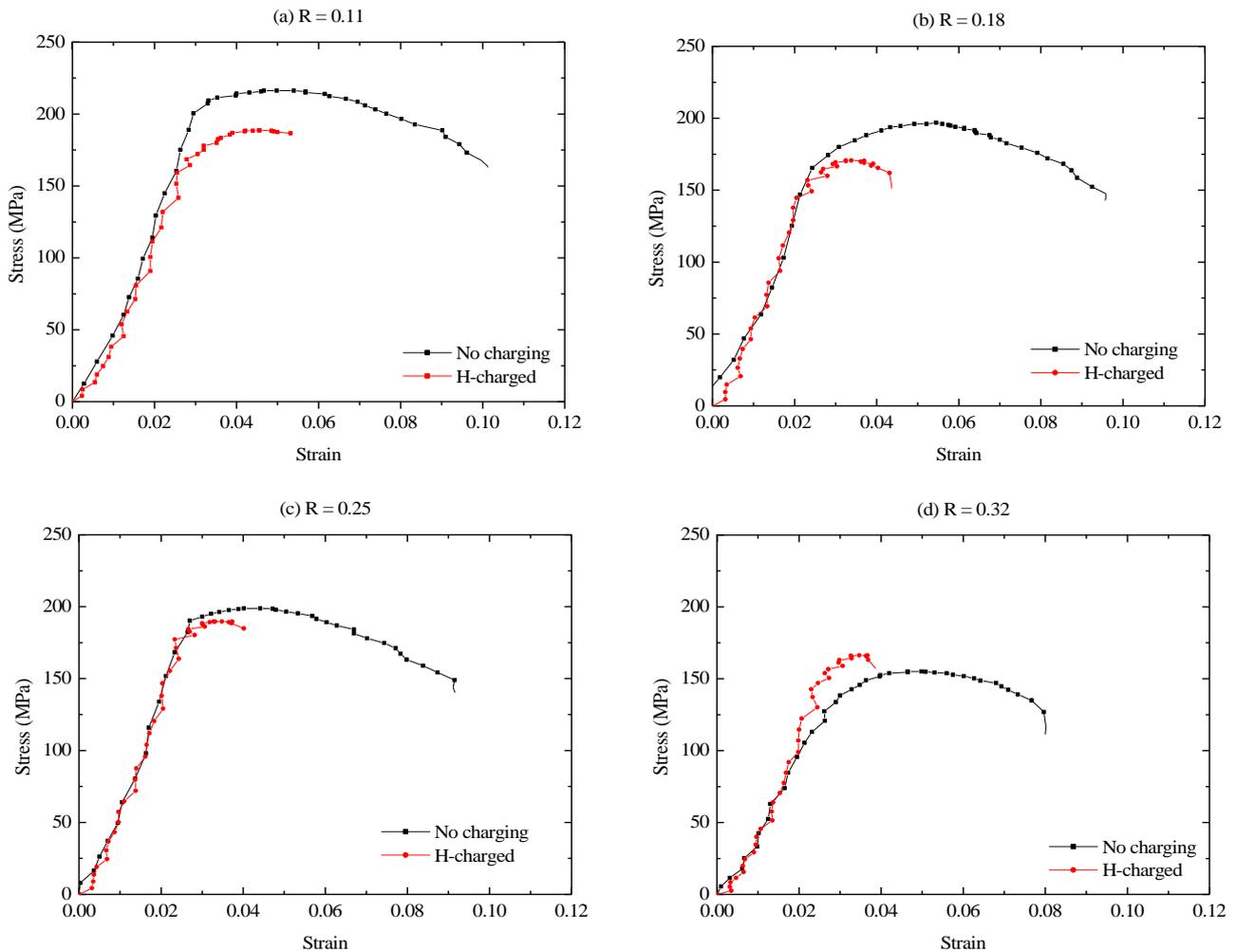


Figure 5. Stress-strain diagrams obtained for different drawing reductions, before and after hydrogen charging.

It was also possible to show that by introducing hydrogen by cathodic charging, the fracture surface of the tested samples was also modified. In Figure 6(a) and 6(b), the fractured specimens obtained after the 0.32 reduction before and after hydrogen permeation are presented, respectively. Prior to hydrogen charging, the specimens exhibited a ductile fracture with the characteristic 45° plane. In addition, it is possible to see that the cross section at the fracture surface is reduced, an indication of necking prior to rupture. On the other hand, after exposure to hydrogen the sample 0.32 cold-drawn sample exhibited very little plasticity, as can be seen in Figure 5(d) and no necking, as shown in Figure 6(b). Indeed, the fracture surface was found in this case to be perpendicular to the applied stress in tensile testing, an indication of brittle fracture in the otherwise ductile material.

In order to provide a quantitative assessment of the influence of hydrogen charging on the mechanical behavior of the cold-drawn AA6351 alloys, the hydrogen embrittlement (HE) number was determined (Depover *et al.*, 2014) by applying equation (3):

$$HE = \frac{l_0 - l_H}{l_0} \quad (3)$$

In the above equation,  $l_0$  and  $l_H$  are the post-yield relative elongation values before and after hydrogen permeation, respectively. The values obtained by applying equation (3) for the 0.11, 0.18, 0.25 and 0.32 reductions were 0.65, 0.74, 0.80 and 0.80, respectively.

The relation between hydrogen embrittlement, quantified by the parameter defined in equation (3) and the level of cold-drawing reduction is explored in Figure 7, which reveals a positive relation between cold-work by drawing and hydrogen embrittlement. The experimental data were fitted to a first order exponential decay model, as shown in Figure 7, and the following relation was obtained:

$$HE = 0.81 - 0.59\exp(-11.64R) \quad (3)$$

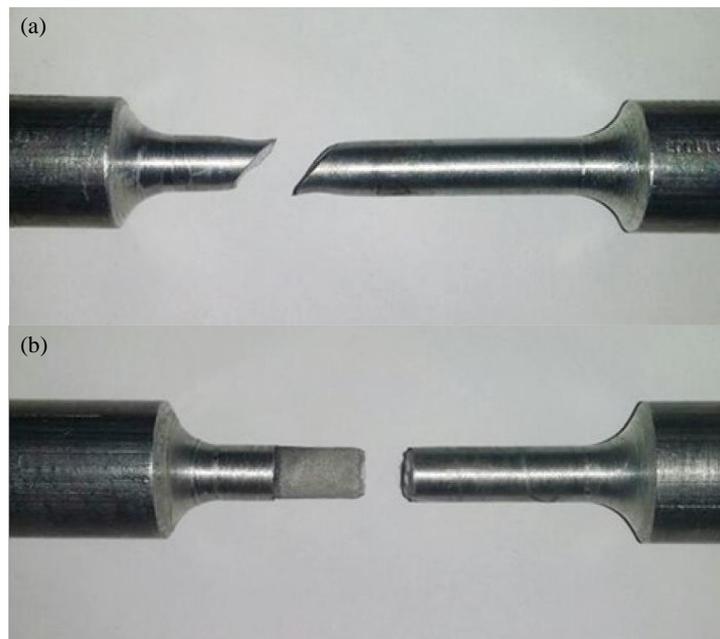


Figure 6. Fractured tensile specimens tested before (a) and after (b) hydrogen charging ( $R = 0.32$ ).

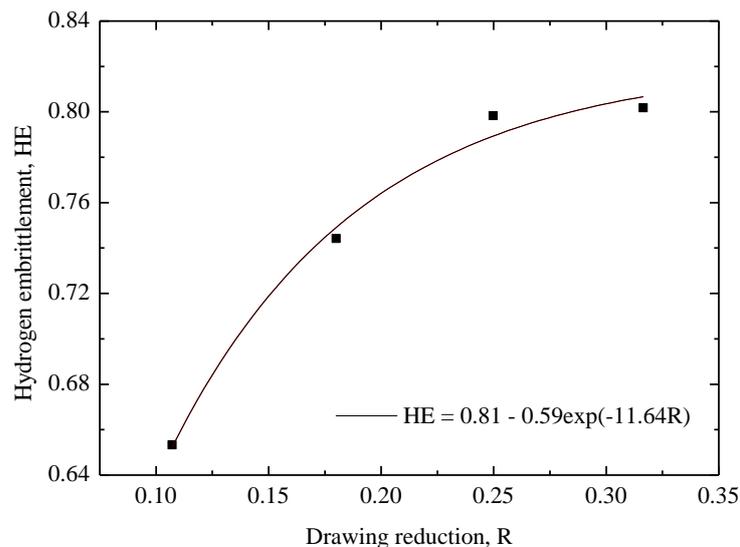


Figure 7. Hydrogen embrittlement as a function of drawing reduction.

#### 4. CONCLUSIONS

In the present investigation, the susceptibility to hydrogen embrittlement of a AA6351 Al-alloy submitted to cold-drawing was studied. The following conclusions could be drawn:

1. The incremental cold-drawing process lead to a linear increase in hardness of the initially annealed AA6351 alloy.
2. Hydrogen charging in 0.6M NaCl solution (containing 0.25mg/l NaAsO<sub>2</sub>) for at 30 mA/cm<sup>2</sup> for 4 hours led to significant embrittlement in all tested materials.
3. Reduction in the cold-drawing process led to an increase in hydrogen embrittlement susceptibility, following a first order exponential decay model.

#### 5. ACKNOWLEDGEMENTS

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