RESEARCH OF THE PLASTIC FLOW OF ELECTROLYTICALLY SATURATED WITH HYDROGEN (He) Al-Cu-Mg ALLOY

Received – Prispjelo: 2016-07-29 Accepted – Prihvaćeno: 2016-11-01 Original Scientific Paper – Izvorni znanstveni rad

The effect of hydrogen embrittlement on the plastic flow of Al-Cu-Mg alloy was investigated (HE). The studies were performed for the test samples of aluminum alloy subjected to electrolytic hydrogenation in a three electrode electrochemical cell. It is found that the mechanical properties and plastic flow curves of aluminum alloy are affected adversely by HE. These are found to show all the plastic flow stages: the linear, parabolic and pre-failure stages. It is established that the hydrogenation enhances the localization of straining leads to significant changes in the characteristics distances between local straining zones. The patterns of localized plasticity appear to be useful for a detailed analysis of plasticity exhibited by aluminum alloys.

Key words: AI-Cu-Mg alloy, plastic deformation, hydrogen, electrolytic process, localization

INTRODUCTION

The presence of hydrogen (H) in solid solution in metals and alloys is related mainly to the small diameter of this element and its capacity to diffuse with certain ease in solid state. Different factors contribute to elevate or diminish the solubilization and/or diffusion of H in alloys. The main ones are temperature, alloy composition, crystalline structure and substructure. Nevertheless, the presence of H in metals and specifically in aluminum alloys is not desired in most of the cases, since H alters considerably the mechanical–metallurgic properties of these materials with the possibility of fracture [1-4]. The most typical damage caused by H in alloys embrittlement.

Since the beginning of the past century, there is known a special type of plastic deformation instability called intermittent flow [5], when the deforming stress oscillates during deformation at a constant strain rate. It was established, that each drop of stress is accompanied by the formation of mesoscopic deformation bands. A series of experimental studies has been conducted to investigate plastic flow kinetics of solids [6]. In the course of its development the plastic deformation is found to exhibit space-time non-homogeneities. The findings presented in [7] give a clue to a most complex problem one comes up against when dealing with the plastic deformation process in solids, namely, its strong tendency to localization. This view finds support in recent studies of other workers [8-10]. However, it is at present unknown how the visible uniformity of deformation is supported at the macro-level, whether there appear zones of macrolocalized deformation in this case, and whether there is a connection between the stages of strain hardening and the type of serrations of the intermittent flow upon hydrogen electrolytic saturation. In this connection, the effect of electrolytic hydrogenation on the plastic flow and fracture of aluminum alloys subjected to thermal strengthening has to be elucidated. Among thermally hardened aluminum alloys, duralumins hold a firm place due to ready availability, satisfactory strength characteristics and high corrosion resistance owing to the protective surface inert oxide film [11].

MATERIAL AND EXPERIMENTAL METHODS

The investigation was performed for the test samples of duralumin D1, which had been subjected to thermal strengthening. The test samples having dog-bone shape were stamped out from hot-rolled 2 mm sheet. These were annealed for 1 h at temperature 340 °C with subsequent cooling down in the furnace to remove internal stresses. After such manipulations, the alloy was in a naturally aged state. [11].

The mechanical tests were carried on at the rate $6,67 \times 10^{-5}$ s⁻¹ at room temperature in a universal testing machine LFM-125 (Switzerland). Then the plastic flow curves plotted for the samples tested in tension were analyzed to single out individual flow stages. The electrolytic hydrogenation of alloy samples was carried on

S. Barannikova, L. Zuev, Institute of Strength Physics and Materials Science of Siberian Branch Russian Academy of Sciences (ISPMS SB RAS) and National Research Tomsk State University «TSU»; A. Bochkareva, A. Lunev, ISPMS SB RAS and National Research Tomsk Polytechnic University «TPU»; Yu. Li, ISPMS SB RAS; G. Shlyakhova, ISPMS SB RAS and Seversk Technological Institute - branch of State Autonomous Educational Institution of Higher Professional Education, Russian Federation

for 100 h under a controlled cathode potential of 600 mV relative to a reference electrode of silver chloride in 1N solution of sulphuric acid solution with an addition of 20 mg/L of thiourea [4,12].

The fractography observations were carried on using a raster electron microscope LEO EVO 50 (Zeiss, Germany) with adapter INCA for the energy-dispersive analysis. Shared use center "Nanotech" ISPMS SB RAS.

Using digital image correlation method (DIC) and an automated complex 'ALMEC-tv' [6,7], the $\sigma(\varepsilon)$ diagram was recorded for the test sample of D1; simultaneously, local strain patterns were being registered for the D1 sample face.

RESULT AND DISCUSSION

The curves obtained for the samples of the original alloy D1 (1) and the counterpart of the same subjected to hydrogenation for t = 100 h (2) tested by uniaxial tension have a characteristic serrated shape, which is indicative of plastic flow instability (Figure 1). The plastic flow would exhibit an unsteady-state behavior from the yield limit up to material failure. Hydrogen charging for 100 h (2) has only minor effect on the yield stress and tensile strength of studied alloys. It is found that a 15% decrease in the elongation to rupture is observed for the counterpart subjected to hydrogenation for 100 h (2) relative to the original alloy (1).



Figure 1 Loading curves obtained for the original alloy D1 (1) and the counterpart of the same subjected to hydrogenation for t=100 h (2)

In the deforming sample of the original alloy (1), the onset of necking takes place, which is a forerunner of viscous fracture. The deformation curves plotted for the original alloy were represented in functional logarithmic coordinates as $\ln(s-s_0) = f(\ln e)$ (here s is the true stress which takes no account of reduction in the work cross-section, s_0 is critical shear stress and e is the true deformation). Three distinct rectilinear segments are distinguished on the flow curve of investigated alloys for strains ε in the different ranges for a constant value *n* (here is the exponent of deformation hardening in

Ludwik equation). These segments correspond to the linear and parabolic work hardening stages and to the prefracture stage, which occur for n = 1; $n \approx \frac{1}{2}$ and $n \approx 0,3$, respectively. The stress-strain curve after electrolytic hydrogenation for 100 h (2) exhibits the stages of linear and parabolic work hardening, and the prefracture stage. In the case of counterpart hydrogenated for 100 h fracture occurs in the absence of necking.

D1 sample without hydrogen manifest a ductile dimpled fracture surface. Hydrogen charging of D1 alloy results in remarkable changes of their fracture surface morphology [2]. In this case the mixed fracture modes with ductile and brittle cleavage fraction were observed [12]. One can assume that presence of hydrogen the second phase precipitation β - phase at the grain boundaries may decrease the grain boundary cohesion resulting in intergranular fracture of the D1 alloy. Such a mechanism of hydrogen embrittlement requires that the hydrogen-induced damage (hydrogen-filled nanovoids or material decohesion) nucleates at the precipitation β - phase on the grain boundaries [13-15].

The measurements of local strain distributions showed that the strain is macroscopically localized at all stages of plastic flow in duralimin D1 polycrystals. An analysis of these patterns revealed that, in the initial (hydrogen-free) state of polycrystals, the tensile plastic strain from proof stress in the deforming material there travel a set of solitary fronts of plastic deformation, which separated the strained and unstrained zones of the sample (Figure 2). The deformation fronts travel at varying rates along the deformed sample.



Figure 2 The localized deformation zones moving at different strain levels in uncharged D1 sample: 1,1% (a) and 5,1% (b)

To study the kinetics of the evolution of the macrolocalization zones, we used the representation of the positions of local deformation zones X, in the sample as functions of strains ε_{xx} or time t (at active tension, $\varepsilon \sim t$ (Figure 3); the coordinate X is counted off from the immobile grips of the tensile machine). In [6-8], it has been shown that the use of this method for the distributions with a time-spatial periodicity (wave distributions) makes it possible to determine the spatial and



Figure 3 The localized deformation zones moving at different strain levels in uncharged D1 sample (a) and hydrogen charged D1 sample (b)

temporal periods of the process, as well as the velocities of the motion of the deformation zones V = dX/dt.

An analysis of the distributions of local strain ε_{xx} in tensile tested D1 polycrystals saturated hydrogen during 100 hours showed that, in this case in the material there emerge a set of deformation fronts (Figure 3). At the pre-fracture stage, the deformation zones started moving consistently with a tendency to merge into a high-amplitude focus of localized straining, where a neck-like narrowing of the sample cross section was formed.

The motion rates of localized plasticity fronts were determined for the test samples of D1 using the complex ALMEC-tv; the values obtained are in the range $(0,2...1,8)\cdot10^{-3}$ m/s in uncharged sample (Figure 3, a) and in the range $(0,6...2,5)\cdot10^{-3}$ m/s in hydrogen charged sample (Figure 4, b).

It was found that localized plasticity fronts velocity decreases with plastic deformation of D1. The decrease of the deforming force at the end of the stretching process is well known to be associated with the formation of a macroscopic neck and is characterized with a zero values in the deformation fronts velocity. The change in the front's velocity was attributed to an increasing dislocation density during plastic flow with the process of hydrogen charging [1].

It is found, particularly, that hydrogen provides not only the remarkable softening effect on the alloy D1 [10], but it initiates also significant reconstruction in the spatial scale for the distribution and magnitude of plastic strain in the shear bands (Figure 4). Analysis of the surface relief obtained with atomic-force microscope observations of the alloy D1 with and without hydrogen after deformation by tension clearly evidences, that hy-



Figure 4 AFM-images of D1 surfaces after deformation: a – hydrogen-free specimen; b– specimen with hydrogen

drogen markedly enhances the magnitude of the plastic shear on the meso-scale level. It is seen from Figure 4 that the characteristic scale of the shear modulation or hydrogen-induced mesoscopic strain localization in duralumin can be estimated to be about 5 μ m. It seems that hydrogen promotes dislocation-dislocation reactions, which result in the dislocation source coupling and in generation of excessive vacancies [4].

CONCLUSION

Hydrogen provides not only the remarkable softening effect on the alloy D1, but it initiates also significant reconstruction in the spatial scale for the distribution and magnitude of plastic strain in the shear bands. Change in the microstructure of hydrogen charged duralimin D1 affects on the stress-strain curves and also affects on the plastic strain localization patterns. Hydrogen enhances localization and changes the quantitative parameters of the macroscopic plastic strain localization: the velocity of autowaves plastic strain localization.

Using the analysis of the loading curve three work hardening stages on the flow curve were distinguished. In the case of counterpart hydrogenated for 100 h fracture occurs in the absence of necking. Hence, the probability of hydrogen-induced crack growth forming in as-treated material is high.

Acknowledgments

The work was performed in the Program of Fundamental Research of State Academies of Sciences for the period 2013-2020 yrs.; in the frame of the Tomsk State University Academic D.I. Mendeleev Fund Program and supported in part by the Russian Foundation for Basic Research (project no. 16-08-00385-a)

References

- G.M. Bond, I.M. Roberston, H.K. Birnbaum, Effect of hydrogen on deformation and fracture processes in highpurity aluminum, Journal Acta Metallurgica 36 (1988), 2193–2197.
- [2] E. Pouillier, A.-F. Gourgues, D. Tanguy, E.P. Busso, G.M. Bond, I.M. Roberston, H.K. Birnbaum, A study of intergranular fracture in an aluminium alloy due to hydrogen embrittlement, International Journal of Plasticity 34 (2012), 139–153.
- [3] J.R. Scully, G.A. Young, Jr., and S.W. Smith, Hydrogen solubility, diffusion and trapping in high purity aluminum and selected Al-base alloys, Materials Science Forum 331-337 (2000), 1583-1600.
- [4] Y. Yagodzinskyy, O. Todoshchenko, S. Papula, H. Hänninen, Hydrogen Solubility and Diffusion in Austenitic Stainless Steels Studied with Thermal Desorption Spectroscopy, Steel Research International 82 (2011), 20-25.
- [5] A. F. Portevin and F. Chatelier, Sur un phenomene observe lors de l'essai de traction d'alliages en cours de transformation, Comptes Rendus de l'Académie des Sciences 176 (1923), 507–510.
- [6] L.B. Zuev, V.I. Danilov, S.A. Barannikova, V.V. Gorbatenko, Autowave model of localized plastic flow of solids, Physics of Wave Phenomena 17 (2009), 1-10.
- [7] L.B. Zuev, S.A. Barannikova, Experimental study of plastic flow macro-scale localization process: pattern, propa-

gation rate, dispersion International Journal of Mechanical Sciences 88 (2014), 1-8.

- [8] A. Roth, T.A. Lebedkina, M.A. Lebyodkin, On the critical strain for the onset austenitic FeMnC steel, Materials *Science* and *Engineering* 539 (2012), 280-84.
- [9] C.C. Aydıner, M.A. Telemez, Multiscale deformation heterogeneity in twinning magnesium investigated with *in situ* image correlation, International Journal of Plasticity 56 (2014), 203–218.
- [10] J.F. Hallai, S. Kyriakides, Underlying material response for Lüders-like instabilities, International Journal of Plasticity 47 (2013), 1–12.
- [11] N.J. H. Holroyd, A.K. Vasudevan, L. Christodolou, Stress Corrosion of High-Strength Aluminum Alloys, Academic Press, London, 1989, 463.
- [12] A.V. Bochkaryova, A.G. Lunev, S.A. Barannikova, L.B. Zuev, The effect of electrolytic hydrogenation on the plastic flow of aluminum alloy, Applied Mechanics and Materials 756 (2015), 59-64.
- [13] E. Lunarska, O. Chernyaeva, Effect of precipitates on hydrogen transport and hydrogen embrittlement of aluminum alloys, Journal of Materials *Science* 40 (2004), 399-407.
- [14] M.B. Kannan, V.S. Raja, Hydrogen embrittlement susceptibility of over aged 7010 Al-alloy, Journal of Materials *Science* 41 (2006), 5495-5499.
- [15] S.J. Kim, M.S. Han, S.K. Jang, Kor. J. Electromicanical characteristics of Al-Mg alloy in seawater for leisure ship: Stress corrosion cracking and hydrogen embrittlement, Korean Journal of Chemical Engineering 26 (2004), 250-257
- Note: The response for English language is YU.V Stankina the translation professional of National Research Tomsk State University «TSU»