PRJ, Vol. 7 (January 1995); P. 31–34, 4 FIGS.

HYDROGEN EMBRITTLEMENT IN LOW CARBON STEELS

H.L. Rizkalla* and A. Abdulwahed*

القصافة الهيدروجينية في الصلب منخفض الكربون

هاني رزق الله و عياد عبدالواحد

تم شحن مجموعات من عينات الصلب المنخفض الكربون بالهيدروجين بطريقة مهبطية. وقيست صلادة جميع العينات عبر مقطعها العرضي بطريقة ميكرز وذلك لتعيين عمق تغلغل الهيدروجين. أظهرت نتائج الدراسة أن عدد الدورات التي تتحملها العينة تحت ظروف الكلال تتناقص بتزايد مقدار الهيدروجين المشحون بالعينات في نفس الوقت الذي تتزايد فيه المقاومة القصوى للشد نتيجة التأثير الإصلادي للهيدروجين. وقد أجريت عملية استعادة الهيدروجين من العينات واستخدمت الفترة الزمنية لإستعادة الهيدروجين لإسترجاع عدد الدورات تحت ظروف الكلال الى 50% من قيمتها الأولى عند درجات حرارة مختلفة لحساب طاقة التحفيز لهذه العملية التي تبين أنها تتفق إلى حد كبير مع النتائج المذكورة في المصادر المختلفة.

ABSTRACT

Sets of low carbon steel (AISI 1015) fatigue specimens were cathodically charged with hydrogen. Transverse Vickers hardness profile was measured for each specimen to estimate the hydrogen penetration depth. The fatigue life and ductility decreased with increasing hydrogen content, while the UTS increased due to the hardening effect of hydrogen. Hydrogen baking time, to restore 50% of the fatigue life, was utilized in calculating the activation energy for the process. The estimated value for the activation energy through the use of this proposed technique proved to be in close agreement with the reported values in the literature.

INTRODUCTION

The damaging effect of hydrogen has long been recognized in ferrous metals. The phenomenon of hydrogen embrittlement (HE) of a metal is manifested primarily as a loss in the ability of a metal to undergo plastic deformation under the applied stress. Normally, it is measured as a decrease in the ductility of the metal [1]. One of the most intriguing aspects of the hydrogen embrittlement phenomenon is its sensitivity to strain-rate and temperature. Hydrogen embrittlement is enhanced by slow strain rate and moderately elevated temperature [2–3]. This is the opposite of the most common behavior where embrittlement is enhanced by high strain rate and low temperature. This slow strain rate and temperature sensitivity suggest that hydrogen embrittlement is mainly controlled by lattice diffusion of hydrogen. In addition, calculations have shown that hydrogen transport rates in association with dislocation motion can be several orders of magnitude than that associated with lattice diffusion [3]. Consequently, hydrogen embrittlement would have a major contribution in the cracking process. Generally, the presence of hydrogen does not change the flow stress or ultimate tensile strength, but it squeezes the true strain to failure. Hydrogen might have a hardening effect leading to an increase in the strength of low-alloy, low strength steels [4, 5]. On the contrary, maraging steels showed a decrease in strength when charged with hydrogen. Such behavior was attributed to the effect of stacking fault energy (SFE) and slip planarity where hydrogen is believed to lower (SFE) [6–7]. Different hypotheses have been proposed to explain the mechanism of hydrogen embrittlement [8–9]. A trial to calculate the activation energy for the process is attempted in the present work through the use of the fatigue behavior of the samples.

---

*Department of Materials and Metallurgical Engineering. Al Fateh University, P.O. Box 13292, Tripoli, Libya.

EXPERIMENTAL PROCEDURE

Fatigue Test

Sets of fatigue specimens were machined and shaped according to ASTM standards [10]. Some of the fatigue specimens were cathodically charged with different amounts of hydrogen in an electrolytic cell. The cell consisted of two similar electrodes (fatigue specimens) immersed in an electrolyte of 4% H_2SO_4 solution. The specimen to be charged was negatively polarized using constant voltage DC power supply, while the other specimen served as an anode to minimize contamination. All specimens were hydrogen charged under constant current density of 15 mA/Cm^2 for different times (10, 15, 20, 25 and 30 minutes). The fatigue life of the as received specimens was determined by using a rotary bending fatigue machine (Shimadzo). On the average, a minimum of three specimens were tested on each stress level. Thus, the stress vs. number of cycles up to failure (S–N curve) was established. Secondly, some of the cathodically charged specimens for different times were tested at a fixed stress of 275 MPa, which is above the experimentally determined endurance limit of the as received specimens. Then, the fatigue life as a function of the hydrogen content (charging time) was obtained. Thirdly, uncharged samples were heat treated at 225°C for 24 hours. Then, some of these heat treated samples were hydrogen charged for 30 minutes and their fatigue behavior was investigated. Finally, hydrogen charged specimens for 30 minutes, were baked as 125°C and 225°C for different baking periods (0.5, 7, 12 and 24 hours). The activation energy for the baking process was calculated provided that the time and temperature to restore 50% of the uncharged fatigue life were known.

Hardness Test

Small cylindrical sections ($\phi = 12$ mm) were cut from the gage length of untested hydrogen charged fatigue specimens. The transverse surface hardness was measured on a Zwick 3212 tester. The Hardness is taken at 1 mm intervals, starting from the edge towards the center of the specimen. The specimen was rotated at 90° and measurements were repeated. The change in hardness was used to give an indication about the hydrogen penetration depth.

Tensile Test

The ultimate tensile strength was measured as a function of charging time for the as received, and hydrogen charged specimens. Also, the percentage reduction in area as a function of the hydrogen content was calculated.

RESULTS AND DISCUSSION

Fatigue Results

The S–N curve for the as received samples showed that the endurance limit (fatigue life > 10^7 cycles) is 265 MPa which is 0.6 of the ultimate tensile strength. The deleterious effect of hydrogen appeared as a drop in the fatigue life, under constant stress of 275 MPa. Increasing the charging time up to 10 minutes did not produce a significant decrease in the fatigue life as can be shown in Fig. 1. Specimens charged for 15–25 minutes suffered from a remarkable decrease in the fatigue life. Further increase in charging time did not affect the fatigue life. This behavior is attributed to diffusion limitations where a period of 25 minutes of charging time would correspond to the amount of maximum hydrogen that can be absorbed by the steel due to the hydrogen fugacity that prevailed over the surface of the specimen under the prevailed charging conditions [7]. Consequently, this would represent the maximum degree of embrittlement beyond which little or no further embrittlement is observed. In addition, the fatigue behavior for the specimens that were heat treated at 225°C then charged with hydrogen for 30 minutes did not show any significant difference as compared to the fatigue behavior of the non heat treated specimens. Such behavior is in agreement with the previous argument.

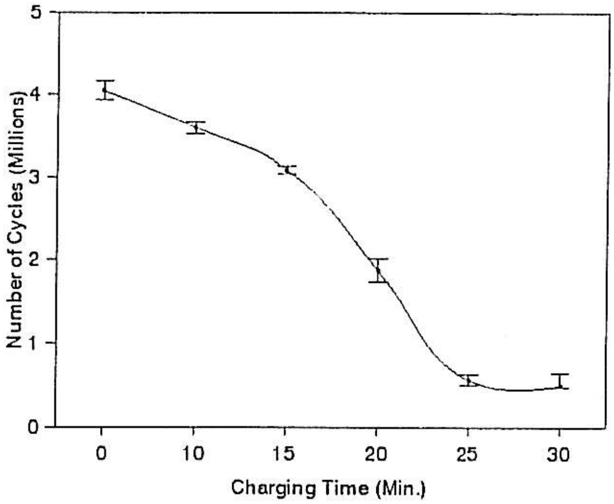

FIG. 1. The effect of hydrogen content (charging time) on fatigue life.

Transverse Hardness

The surface hardness increased with increasing the charging time, Fig. 2. This effect is mainly due to lattice distortion and hardening effect resulting from hydrogen diffusion. Hydrogen would be accumulated in regions of triaxial stress-state, or be associated with

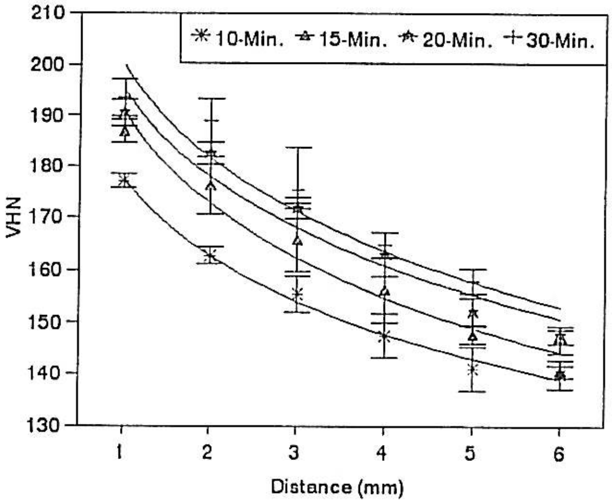

FIG. 2. Transverse hardness profile for different charging time.

dislocations. Hydrogen atoms can recombine and form H_2 molecules, accompanied by a pressure build up. Consequently, local regions would be under compressive state of stress, causing an increase in hardness. For constant charging time, hardness gradually decreased as the center of the specimen was approached, but the hardness level increased with increasing charging time as can be shown in Fig. 2.

Tensile Properties

An increase of approximately 20% in ultimate tensile strength (UTS) was observed between the as received and the maximum hydrogen charged specimens. The increase in UTS was accompanied by a 13% decrease in percent reduction in area as shown in Fig. 3. Similar behavior was reported by Oriani [11] in case of medium carbon steel (AISI 1035), where an increase in the required stress to form and propagate Luders bands was observed. In general low-alloy, low strength steels show an increase in strength associated with decreased ductility due to hydrogen induced hardening effect. On the other hand, high strength steels exhibit a decrease in strength when charged with hydrogen [6–7].

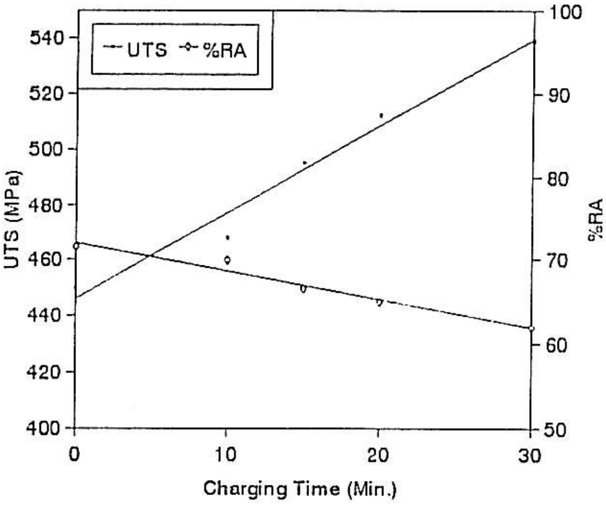

FIG. 3. The change in the tensile properties with increasing hydrogen content.

Activation Energy

Hydrogen embrittlement is a reversible damage process. Therefore, charged specimens can restore their ductility if heated up in vacuum or left long enough in ambient conditions. Fig. 4 represents the restored fatigue life of the charged specimens when baked for different times at 125°C and 225°C. The baking time required to restore 50% of the fatigue life at different temperature was utilized in calculating the activation energy for the process where

$$\Delta H = R \ln \left[\frac{t_2}{t_1} \right] \left[\frac{T_1 \times T_2}{T_2 - T_1} \right]$$

and ΔH = activation energy, t_1, t_2 are the baking times at temperatures T_1 and T_2 respectively and R is the gas constant. The calculated activation energy $\Delta H = 3.7$ Kcal/mole turned out to be in good agreement with the reported values of ΔH for hydrogen diffusion in steel (ΔH reported = 4.2 KCal/mole [12]). Keeping in mind the statistical nature of the fatigue test and it's dependence on many variables other than the hydrogen content the above calculated ΔH would be a good approximation.

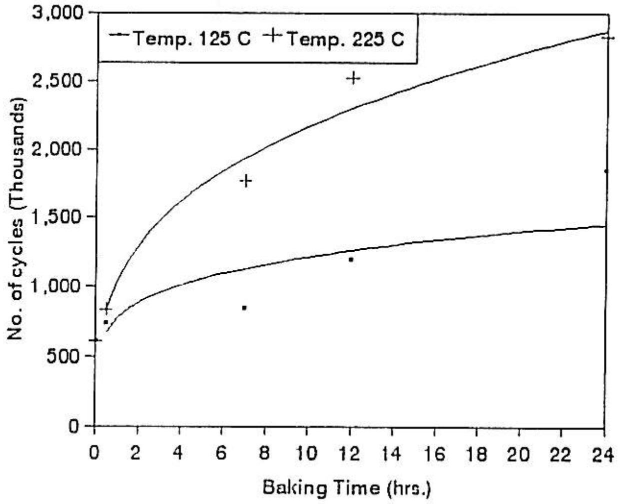

FIG. 4. The effect of baking time and temperature on the fatigue life.

CONCLUSION

1. The ultimate tensile strength of AISI 1015 increased with increasing the hydrogen content in

the samples; while the ductility and the fatigue life decreased.

2. The transverse hardness measurements proved to be effective in probing the hydrogen penetration into the sample.
3. The proposed technique for measuring the activation energy for hydrogen diffusion proved to be successful.

ACKNOWLEDGEMENT

The authors would like to thank the Department of Materials & Metallurgical Engineering for providing the facilities to carry out this work.

REFERENCES

[1] Troiano, A.R., 1973, Hydrogen in Metals, Proceedings of an international conference of the effect of hydrogen on materials properties and selection, ASM, p. 3.

[2] Dieter, G.E., 1986, Mechanical Metallurgy, 3/e., McGraw Hill, London.

[3] Tetelman, A.S. and McEvily, J., 1967, Fracture of structure Materials, Wiley, New York.

[4] Gosh, A. and Basu, A., 1988 U.K. Chatterjee and B. Sasmal, Trans. Ind. Inst. Metals 41, 519.

[5] Bowker, J., Piggett, M.R. and Wheatherly, G.C., 1984, Proceedings of the 6th International Conference on Fracture, New Delhi, Vol. 4 edited by K.N. Raju *et al.* p. 2371.

[6] Thompson, A.W. and Bernstein, I.M., 1980, Advances in Corrosion Science and Technology, Vol. 7, edited by M.G. Fontana, R.W. Staehle, Plenum Press, New York, p. 53.

[7] Reddy, K.G., Arumugam, S. and Lakshmanan, T.S., 1992, Hydrogen embrittlement of marging steel, J. Mats. Sci. 27, p. 5159–5162.

[8] Oriani, R.A., 1977, A review of proposed mechanisms for hydrogen assisted cracking in Metals, in Hydrogen Damage, edited by Cedric Beachem edited, Metals Park, Ohio, p. 301.

[9] Gilman, J.J., The Role of surface Hydrides in stress Corrosion Cracking, ibid.

[10] Dave, H.E., Troxell, C.E. and Wiskocil, C.T., 1964, The testing and inspection of Engineering Materials, McGraw Hill, New York.

[11] Oriani, R.A. and Josephic, P.H., 1980, Met. Trans. A, p. 1809–1820.

[12] Hertzber, R.W., 1986, Deformation and Fracture Mechanics of Engineering Materials, Wily, New York.