

Selecting Hydrogen Embrittlement Resistant Materials by Means of the Disc Rupture Test

E. Leunis, L. Duprez

This document appeared in

Detlef Stolten, Thomas Grube (Eds.):

18th World Hydrogen Energy Conference 2010 - WHEC 2010

Parallel Sessions Book 5: Strategic Analyses / Safety Issues / Existing and Emerging Markets

Proceedings of the WHEC, May 16.-21. 2010, Essen

Schriften des Forschungszentrums Jülich / Energy & Environment, Vol. 78-5

Institute of Energy Research - Fuel Cells (IEF-3)

Forschungszentrum Jülich GmbH, Zentralbibliothek, Verlag, 2010

ISBN: 978-3-89336-655-2

Selecting Hydrogen Embrittlement Resistant Materials by Means of the Disc Rupture Test

Elke Leunis, Lode Duprez, OCAS NV, ArcelorMittal Global R&D Gent, Belgium

1 Introduction

In industry, metals and alloys are often used in hydrogen rich environments. Hydrogen sometimes has a detrimental effect on the mechanical properties of metals, depending on the alloy, the microstructure, the environment, the stress state etc.

In many cases hydrogen related studies on laboratory scale are performed by introducing the hydrogen artificially into the metal via cathodic charging [1]. For applications with high pressure hydrogen gas it is however important to test the alloys in more realistic conditions.

The disc rupture test, a standardized test to select materials for hydrogen pressure vessels, is used to evaluate high strength materials in contact with gaseous hydrogen under pressure. In this test a circular specimen is loaded up to rupture. The rupture pressure ratio of helium to hydrogen is determined for different pressure increase rates, which results in the embrittlement index of the material. The disc rupture test was already discussed many years ago to evaluate the hydrogen effect on materials [2,3,4,5]. In many cases cathodically charged specimens were tested under He and compared with uncharged specimens. The disc rupture test is a fast and reliable tool to screen the hydrogen embrittlement behaviour of lab as well as industrial materials.

In this study, different metallic alloys are tested with the disc rupture test. The rupture appearance is studied to obtain a more profound understanding of the fracture mechanisms. Additionally, the materials are electrolytically charged to compare their behaviour. The comparison of the different methods will result in an evaluation of the use of the disc rupture test for selecting hydrogen embrittlement resisting materials.

2 Experimental

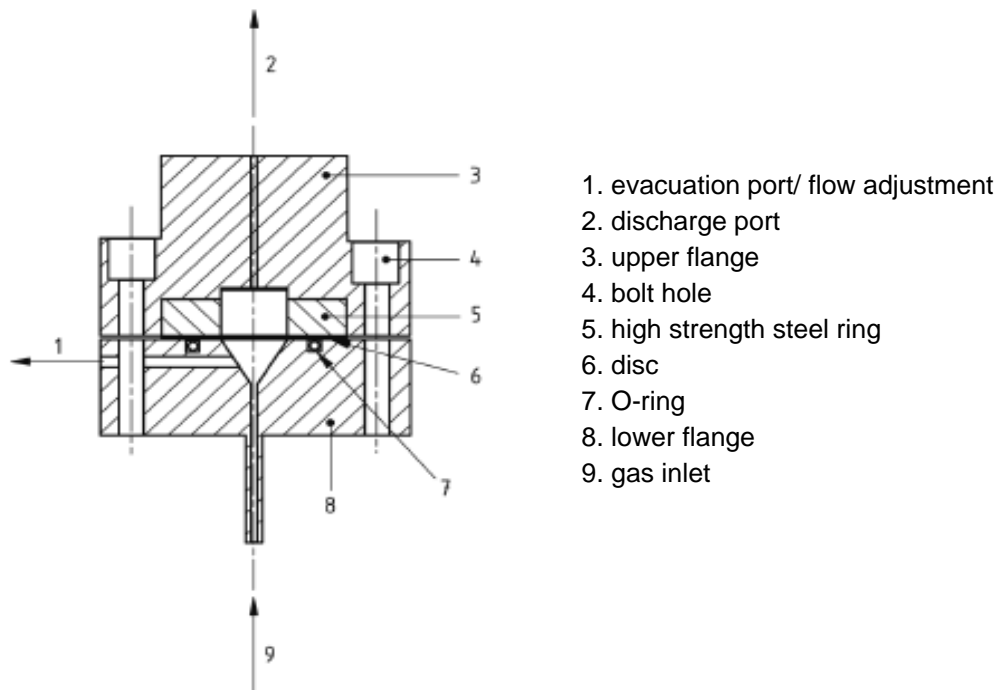
Materials. A selection of ferrous and non-ferrous alloys is used in this study: stainless steel, aluminium, nickel, titanium and copper based alloys. The mechanical properties are summarized in Table 1. Industrially produced materials are used. Diffusion coefficients from literature are given in Table 2.

Table 1: Mechanical properties.

| Grade | Material | Thickness | Rp0.2 | Rm | A50 |
|-------------|----------------------------|-----------|-------|-------|-----|
| | | [mm] | [MPa] | [MPa] | [%] |
| SS 304 | austenitic stainless steel | 0.8 | 290 | 676 | 54 |
| SS 316 | austenitic stainless steel | 1.0 | 279 | 613 | 48 |
| SS 430 | ferritic stainless steel | 1.0 | 306 | 497 | 29 |
| Inconel 718 | Ni-alloy | 1.0 | 457 | 898 | 44 |
| OFHC Copper | Copper | 1.0 | 200 | 269 | 36 |
| A6061 | aluminum alloy | 1.6 | 272 | 336 | 19 |
| Ti-6Al-4V | titanium alloy | 1.0 | 1053 | 1063 | 10 |

Table 2: Literature data on the diffusion of hydrogen.

| | D_0 | H_D | $D = D_0 \exp(-H_D/RT)$ | Ref |
|----------------------|-----------------------|---------------------|-------------------------|------|
| | $m^2 \cdot s^{-1}$ | $kJ \cdot mol^{-1}$ | $m^2 \cdot s^{-1}$ | |
| Austenitic SS | 5.79×10^{-7} | 53.62 | 2.3×10^{-16} | [6] |
| Ferritic SS | | | 1.73×10^{-12} | [7] |
| Inconel 718 | 4.06×10^{-7} | 48.63 | 1.2×10^{-15} | [8] |
| OFHC copper | 1.06×10^{-6} | 38.5 | 1.9×10^{-13} | [9] |
| Pure Aluminum | 1.75×10^{-8} | 16.2 | 2.5×10^{-11} | [10] |

**Figure 1: Lay-out of the disc pressure test.**

Disc rupture test. In the disc rupture test a mounted disc sample is subjected to increasing gas pressure at a constant pressure increase rate (Figure 1). The embrittling effect of hydrogen is evidenced by comparing the hydrogen rupture pressures P_{H_2} with the helium rupture pressures P_{He} , helium being chosen as a reference gas. The rupture pressure ratio P_{He}/P_{H_2} shall be determined. The lower this ratio, the better the material behaves in the presence of hydrogen. The discs with diameter 58 mm are punched from the original sheet. No additional surface preparation or thinning was performed, except for the A6061 alloy which was polished down to 0.75 mm. Pressure increase rates between 1 bar/min and 100 bar/min are applied to compare the materials. The test temperature is 20°C in all cases.

The fractured surface for samples tested under helium and hydrogen are examined with the scanning electron microscope (SEM).

Cathodic charging. Samples of 10 x 6 mm were charged cathodically in a poly-carbonate cell between two symmetric Pt anodes. The electrolyte used in the tests is a sulfuric aqueous solution containing thiourea (1 g CSN₂H₄ + 0.5M H₂SO₄ / 1l H₂O). The samples were charged for 1h at room temperature using a varying current density up to 100 mA/cm² to evaluate the influence of the charging conditions. After charging, the samples were cleaned with distilled water and acetone, the surface and edges were polished to remove the electrolyte and cleaned again with distilled water and acetone. Immediately after charging, the hydrogen content was determined via melt extraction with a Bruker G8 Galileo equipment.

3 Results & Discussion

Disc rupture test. The rupture pressure ratios for the ferrous and non-ferrous alloys as a function of the pressure rise rate are given in Figure 2. A pressure ratio of 1 means no effect of the hydrogen, while all higher values indicate a HE effect.

The test results reveal that the high pressure hydrogen atmosphere does not have any embrittling effect on the austenitic 316L SS, while it is significantly affecting the properties of the 304 SS. For the 430 SS grade the hydrogen only has an effect for low pressure rise rates, i.e. low deformation rates.

In general austenitic stainless steels are less susceptible to HE due to the low hydrogen diffusivity (see Table 2) and the high hydrogen solubility. In the 316L SS the austenite is stable, even after deformation. In the 304 SS however the austenite is metastable and transforms to strain-induced α' -martensite during deformation. This martensite enhances hydrogen uptake since hydrogen diffuses more rapidly in BCC phases and make the material more vulnerable to HE.

In the ferritic 430 SS the embrittlement is dependent on the strain rate which is determined by the pressure rise rate. The local strains enhance the local uptake of hydrogen and micro-cracks are formed. For slow strain rates, the exposure time to the hydrogen gas is higher and also the hydrogen has more time to diffuse and to recombine in the deformed matrix.

Most of the tested non-ferrous alloys seem to be insensitive to HE under high pressures. In the Inconel 718 alloy the HE effect increases significantly with lower deformation rates, which can be explained by the same mechanism as for the 430 SS discussed above. It is known from literature that the Inconel 718 alloy is sensitive to HE [8,11]. This embrittlement is related to the presence of strengthening phases γ' and γ'' and the grain boundary phase δ .

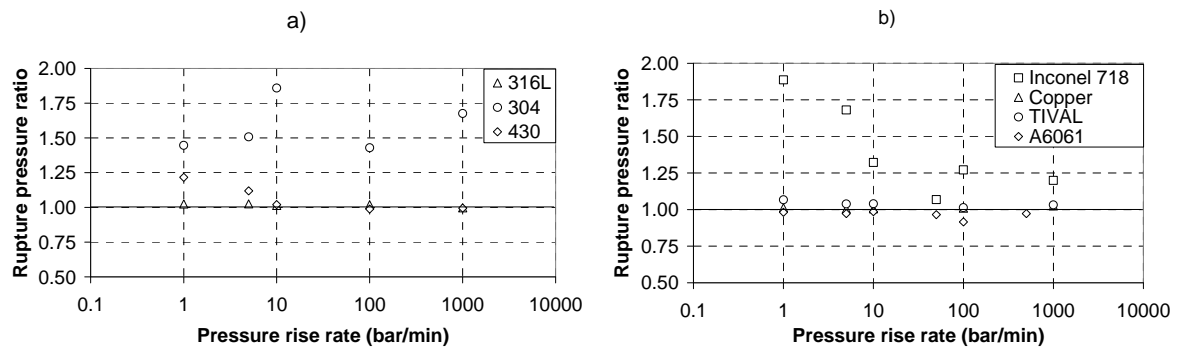


Figure 2: Rupture pressure ratio as a function of the pressure rise rate a) ferrous alloys b) non-ferrous alloys.

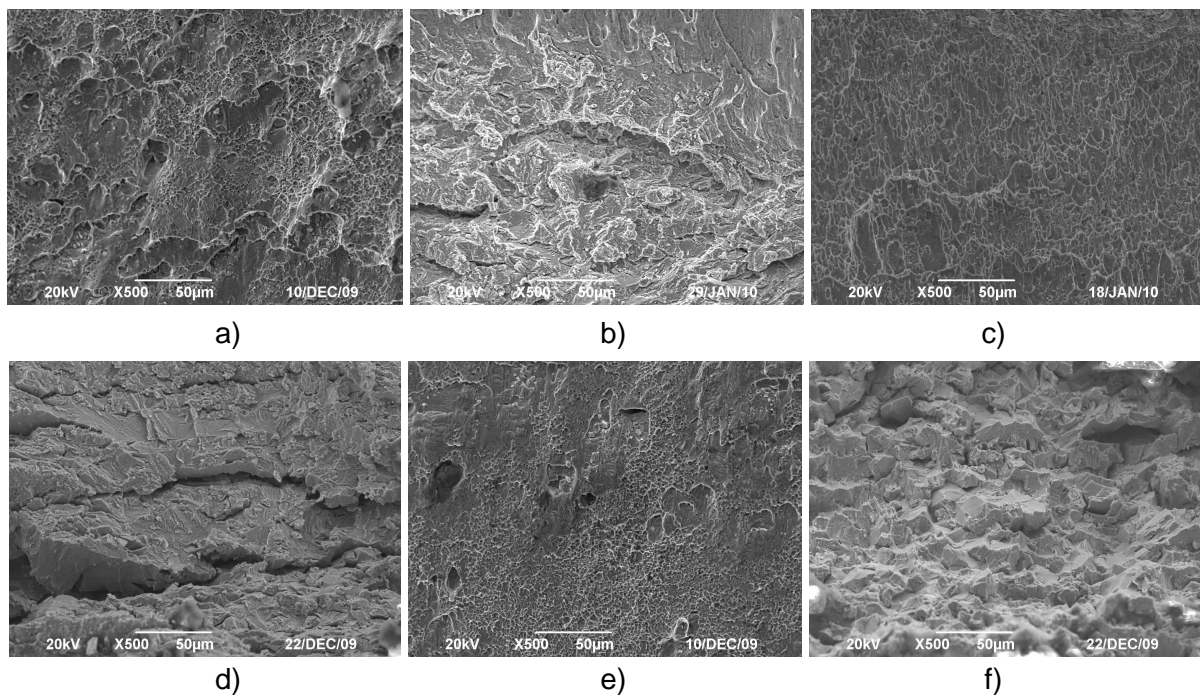


Figure 3: Fracture surface after disc rupture test: a) Inconel 718 at 1 bar/min in He; b) SS304 at 1 bar/min in H₂; c) SS430 at 1000 bar/min in H₂; d) SS430 at 1 bar/min in H₂; e) Inconel 718 at 1000 bar/min in H₂; f) Inconel 718 at 5 bar/min in H₂.

Fracture analysis. Results from the scanning electron microscope observations on fracture surface after disc rupture testing are presented in Figure 3. All specimens exhibited ductile fracture after helium testing for which an example of Inconel 718 is given in Figure 3.a. The samples that not revealed any HE effect, such as the 316L SS, copper, Ti-6Al-4V and A6061, had a similar ductile fracture after testing in hydrogen.

For 304 SS the fracture surface in hydrogen was brittle and transgranular for all testing conditions. For the alloys 430 SS and Inconel 718, the hydrogen sensitivity is strain rate dependent. Ductile fractures are observed for high pressure rise rates and brittle fractures for low pressure rise rates. In the SS430 a brittle cleavage fracture occurred, while the fracture in Inconel 718 is clearly intergranular.

Cathodic charging. During cathodic charging, the samples were saturated with hydrogen. After charging, hydrogen is assumed to be present in both interstitial lattice places as well as near lattice defects, *i.e.* at hydrogen traps. Figure 4 gives an overview of the hydrogen content of the samples charged for 1h with different current densities.

It is clear from Figure 4 that for identical charging conditions, the hydrogen content in the materials is different. This is related to the difference in solubility of hydrogen in the crystal lattices, the hydrogen diffusion coefficients and the surface status of the alloys. Diffusion coefficients from literature are given in Table 2.

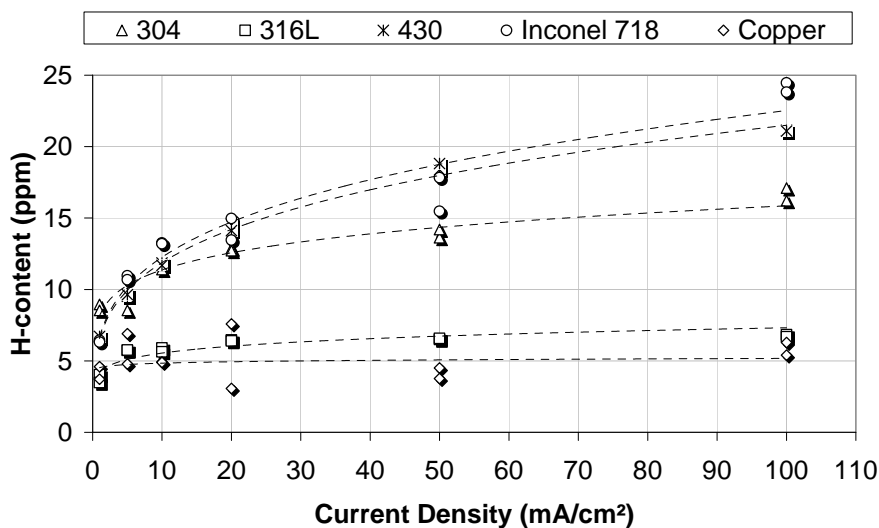


Figure 4: Overview of the hydrogen content of the samples charged for 1h with different current densities.

In the disc rupture test, the hydrogen uptake takes place from the high pressure gaseous phase. In the cathodic charging, the hydrogen is electrochemically forced to enter the material which makes the testing conditions more severe. However the same tendencies as for the disc rupture test are noticed with cathodic charging. Almost no hydrogen is taken up in the 316L SS and copper material. They didn't show any embrittlement during disc rupture testing neither. In the ferritic 430 SS and the Inconel 718 alloys, the hydrogen charging is high. For the austenitic 304 SS in the non-deformed state however, the same behaviour is expected as for the 316 SS, because of the very low hydrogen diffusivity in austenite. However, the samples are much more charged than the 316 SS. This makes us assume that surface effects are playing. This needs further investigations.

4 Conclusions

Disc rupture tests were performed on different ferrous and non-ferrous alloys in helium and hydrogen atmosphere. The ratio of the rupture pressures in both gases gives an idea of the HE and allows to select HE resistant materials for gaseous applications.

The stable austenitic 316 SS does not show any HE with the disc rupture test. The metastable 304 SS embrittles after gaseous hydrogen exposure combined with deformation because strain induced martensite forms. The HE of the ferritic 430 grade is dependent on the strain rate.

The tested copper, aluminium and titanium alloys did not show any HE during disc rupture testing. The Inconel 718 does show strain rate dependent HE. The obtained results are in agreement with literature.

The disc rupture test results were in agreement with results obtained after cathodic charging.

References

- [1] Mertens G., Duprez L., De Cooman B.C., Verhaege M., *Advanced Materials Research*, Vols.15-17 (2007), pp. 816-821
- [2] Fidelle, J. P., Broudeur, R., Pirrovani, C., and Roux, C., *ASTMSTP543*, 1974, pp. 34-47.
- [3] Fidelle, J. P., Bernardi, R., Broudeur, R., Roux, C., and Rapin, M., *ASTM STP 543*, pp. 221-253.
- [4] Fidelle, J.-P., *ASTM STP 962*, 1988, pp. t53-172.
- [5] Beghini M. ; Benamati G. ; *J. Eng Mat Technol* (1996) Vol. 118, 179-185
- [6] XK Sun, J Xu and YY Li. *Mater Sci Eng A114* (1989) 179-187
- [7] S.K. Yen, I.B. Huang / *Materials Chemistry and Physics* 80 (2003) 662–666
- [8] L. Fournier, D. Delofosse, T. Magnin, *Mater Sci Eng A269* (1999) 111-119
- [9] WG Perkins. *J vac Sci Technol* 10 (1973) 543-556
- [10] GA Young and JR Scully, *Acta Mater* 46 (1998) 6337-6349
- [11] L.Liu, K. Tanaka, A. Hirose, K.F. Kobayashi, *Sci Technol Adv Mat* 3 (2002) 335-344