Australian Journal of Basic and Applied Sciences, 7(5): 329-335, 2013
ISSN 1991-8178

Failure Modes of Hydrogen Damage on Metal Tubes

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Abstract: Hydrogen damage commonly occurs on metal tubes with the presence of hydrogen and residual or applied stress. Hydrogen damage understanding and collections of hydrogen damage failure modes are important for constructing a metallurgical failure analysis task. Besides collecting the process background and failure history occurrence in a metallurgical failure analysis task, failures can be predicted base on the fracture surface, surface defects and microstructure change. This paper explains the common hydrogen damage mechanism and consists of general failure modes and may be used as reference in constructing metallurgical failure analysis of metal tubes.

Key words: Failure Analysis; Hydrogen Damage; Hydrogen Embrittlement; Failure Mode.

INTRODUCTION

Hydrogen damage is a form of environmentally assisted failure that results most often from the combined action of hydrogen, and residual or applied tensile stress. Hydrogen damage to specific alloys or groups of alloys manifests itself in many ways such as cracking, blistering, hydride formation and loss of tensile ductility. For many years, these failures have been collectively termed hydrogen embrittlement; this term persists even though it is improperly used to describe a multitude of failure modes involving hydrogen, several of which do not demonstrate the classical features of embrittlement.

In most metals the solubility of hydrogen at room temperature is very low. Nevertheless, its presence strongly influences their mechanical properties. A well-known phenomenon in this context is hydrogen embrittlement. Even at low temperatures, the small hydrogen atoms defuse rapidly through the crystal lattice and can thus be trapped at defects possessing a sufficient binding energy. Among other hydrogen-defect interactions, the interaction with dislocations is of particular importance because it is generally quoted as the origin of embrittlement (Herlach, D., 2000).

Hydrogen Damage Theories:

Various materials used in a power plant such as carbon steel, low and high alloy steel and stainless steel are susceptible to hydrogen damage. It is a very common failure in fossil boiler tubes (Dayal, R.K., N. Parvathavarthini, 2003). During their service, these steels pick up hydrogen from the surrounding environment, which migrates in to the matrix and causes damage. The pickup of hydrogen may be due to corrosion in aqueous medium including pickling, excessive cathodic protection, electroplating without baking, welding using damp electrodes and hydrogen gas if moist used as coolant in generators (Dayal, R.K., N. Parvathavarthini, 2003). Hydrogen can initially be present either externally or internally within the bulk of a structural alloy. Whenever steel surfaces come in to contact with an aqueous environment the following reactions take place (Dayal, R.K., N. Parvathavarthini, 2003).

$$x\mathrm{Fe} + y\mathrm{H_2O} \longrightarrow \mathrm{Fe}x\mathrm{O}y + 2y\mathrm{H}^+$$

$$\mathrm{H}^+ + e \longrightarrow \mathrm{H(ads)}$$

The adsorbed hydrogen (in atomic form) migrates further into the metal matrix causing metal–hydrogen interaction. The molecular hydrogen formed on the external surface then gets evolved as a gas in the aqueous solution. The mechanism of hydrogen embrittlement is illustrated in figure 1.

Hydrogen embrittlement results from acid corrosion either due to condenser leaks allowing ingress of chloride or improper rinsing of chemical cleaning solutions. The nascent hydrogen generated by corrosion reactions diffuses into the metal and reacts with carbon present in the iron carbides to form methane bubble. The large methane molecules trapped produce very high localized stress leading to microfissures which link up and form cracks. Sometimes pitting caused by high oxygen level as well as chlorides is also associated with

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hydrogen damage (Dayal, R.K., N. Parvathavarthini, 2003). With the presence of hydrogen, it lowers the yield stress and initial flow stress of pure iron at room temperature, more specifically at temperatures greater than 200K (Hirth, J.P., 1985). Hydrogen embrittlement usually happens with high temperature that can be explained by graph in figure 2. It begins in the metallurgical process because the hydrogen solubility in the molten metal is much higher than it is in the solid condition.

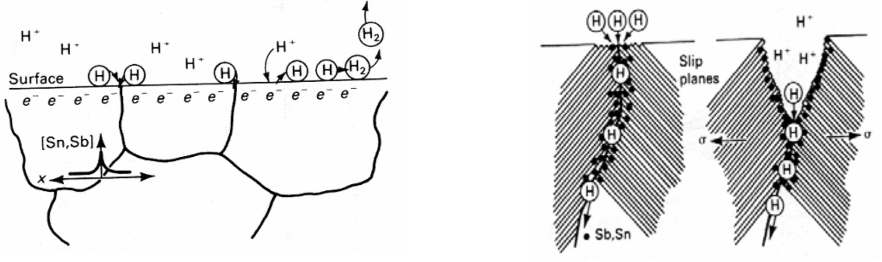

Fig. 1: Mechanism of hydrogen embrittlement (Ralph M. Davidson, 1987).

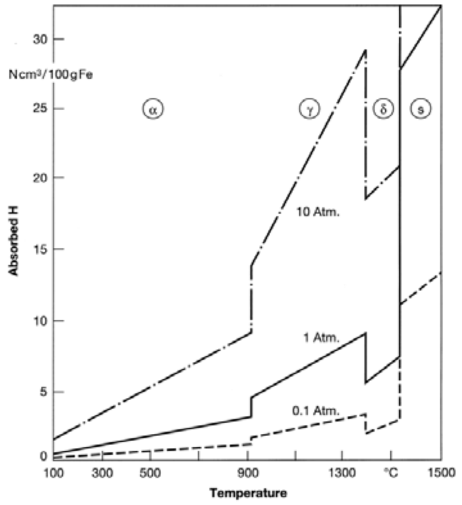

Fig. 2: Solubility of Hydrogen in Iron as a function of temperature and of pressure (Woodtli, J., R. Kieselbach, 1999).

A characteristic feature of hydrogen embrittlement is the lowering of the ultimate tensile strength. Fracture may occur either by shear or cleavage. Certainly both shear strength and cleavage strength are lowered by hydrogen. The lower the strain hardening of material, the more lowering of the ultimate tensile strength will decrease the ductility. When temperature increases with the hydrogen pressure increases, the ultimate tensile strength will decrease. According to Griffth theory, the virgin material contains numerous small cracks. Fracturing will start at an external stress at which an incremental enlarging of a crack can result in a decrease of the total energy. This involves release of elastic strain energy in excess of the energy required to build up the new crack surface, so that the net energy set free in the incremental process is positive. When the crack can continue to spread at constant external stress with further decrease in total energy of the sample, sudden fracturing will occur at a very high rate of crack propagation. Hydrogen embrittlement is less under impact tests or under tensile tests at low temperatures. When temperature is lowered the hydrogen diffusivity decreases, resulting in a decreased rate of crack propagation (de Kazinczy, F., 1954).

The transport of hydrogen from its original form and location to another form and location within the alloy where degradation can occur is probably the most complex aspect of the hydrogen embrittlement process. When hydrogen is normally present in the bulk of the alloy, its transport is a simple process and most often controlled by lattice diffusion under the influence of stress gradient. There are several specific locations in a material where the presence of hydrogen may be critical to the fracture behavior. These include the lattice itself (hydrogen in solution) as well as grain boundaries, incoherent and coherent precipitates, voids and dislocations as shown in figure 3.

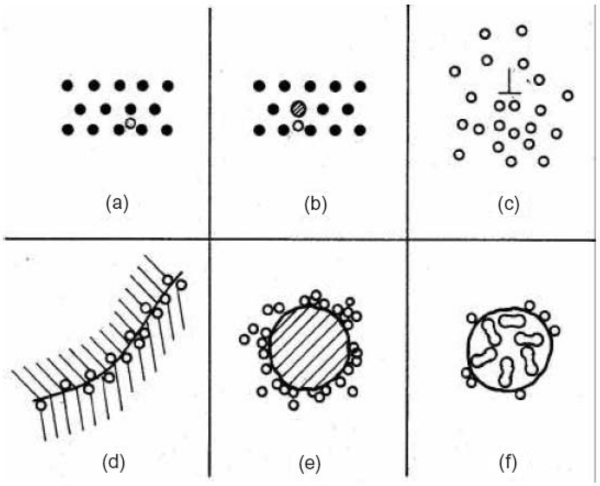

Fig. 3: Schematic view of destinations for hydrogen in a metal microstructure: (a) solid solution; (b) solute–hydrogen pair; (c) dislocation atmosphere; (d) grain boundary accumulation; (e) particle-matrix interface accumulation; (f) void containing recombined hydrogen (Dayal, R.K., N. Parvathavarthini, 2003).

Hydrogen Damage Failure Modes:

The figure indicates the way in which hydrogen from a variety of sources transported by dislocations or lattice diffusion can accumulate at any one or jointly with other sites (traps) in the metal matrix. These traps are classified as 'irreversible' if they act purely as hydrogen sinks or reversible if they accept hydrogen under some circumstances but act as a hydrogen source otherwise. The so-called irreversible traps liberate hydrogen at a sufficiently elevated temperature which depends on the trap energy (Dayal, R.K., N. Parvathavarthini, 2003). The local hydrogen concentration at a potential crack site must reach a critical level for a given stress intensity factor *(Kt)* before the initiation of cracking. The hydrogen traps influence the likelihood of cracking by controlling the availability of hydrogen to the critical cracking locations. An increase in temperature decreases the trap energy, thus decreasing their tendency to hinder hydrogen diffusion. Above about 400°C, the apparent diffusion coefficient is close to the diffusion coefficient of hydrogen by lattice diffusion while below this temperature the diffusion coefficient is affected by hydrogen trapping. The number of reversible traps is strongly affected by the transformation products formed on cooling. For example a tempered martensite has more trapping sites than a pearlite. This is due to the increased surface area of the finer carbides in the martensite (Dayal, R.K., N. Parvathavarthini, 2003).

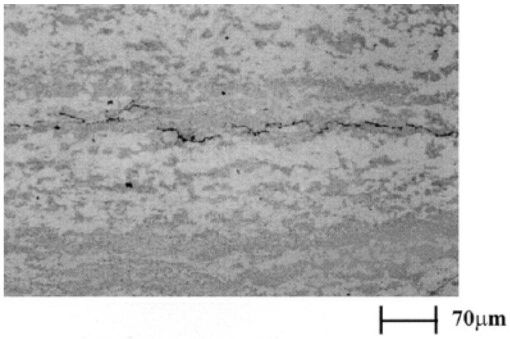

Fig. 4: Microcracking in hydrogen-charged specimens of X60 steel (Hardie, D., 2006).

Hydrogen embrittlement can be observed by metallurgical examination with result in either intergranular or transgranular by brittle cleavage or interface separation, depending on the relative strength of the boundary. At the region in contact with hydrogen fracture, intergranular occurs by interface separation of the grain boundaries where it depends on the relative strength of the grain boundary (Eliaz, N., 2000). Figure 4 shows an example of intergranular microcracks in hydrogen-charged specimens of X60 steel. Steels with reduced yield strengths when damaged by hydrogen charging at high fugacity can be characterized either by enhanced dislocation formation or void or fissure formation at grain boundaries and interfaces of second phase particles.

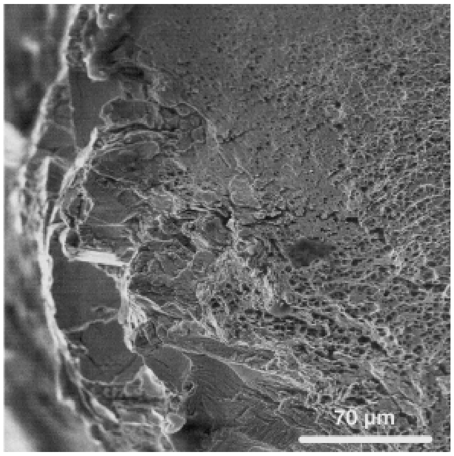

Fig. 5: Transition between brittle and ductile fracture (Herms, D.E., 1999).

Fracture surface of ruptured steel cause by hydrogen embrittlement often can be seen in brittle appearance or together with ductile appearance where it can be seen in figure 5 and figure 6. Figure 5 shows the transition between ductile and brittle surface of 316L steel caused by hydrogen emmbrittlement (Herms, D.E., 1999). Fractures by hydrogen are promoted by the presence of P, S and other metalloids and impurities where the crack surfaces include ductile dimple-rupture, ductile tearing, quasi cleavage and cleavage forms (Hirth, J.P., 1985).

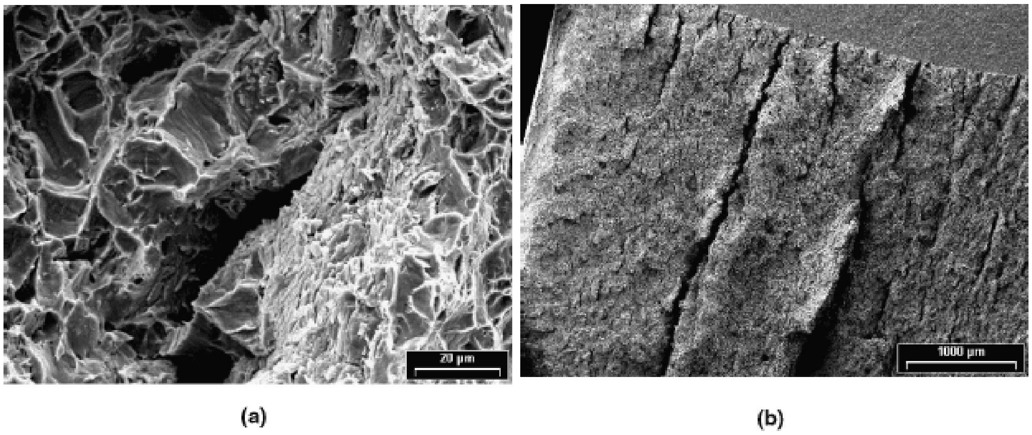

Fig. 6: (a) Fracture surface observed in a specimen that fractured by brittle mode. (b) Large longitudinally oriented cracks (Bertolino, G., 2002).

Figure 7 shows an example of an aluminium alloy 2024 that was corroded with corrosion-inducedhydrogen embrittlement where combination of intergranular, quasi cleavage and ductile fracture. When tensile stresses are applied to hydrogen embrittled component it may fail prematurely. Hydrogen embrittlement failures are frequently unexpected and sometimes catastrophic. An externally applied load is not required as the tensile stresses may be due to residual stresses in the material. The threshold stresses to cause cracking are commonly below the yield stress of the material.

High strength steel, such as quenched and tempered steels or precipitation hardened steels are particularly susceptible to hydrogen embrittlement. Hydrogen can be introduced into the material in service or during materials processing. Tensile stresses, susceptible material, and the presence of hydrogen are necessary to cause hydrogen embrittlement. Residual stresses or externally applied loads resulting in stresses significantly below yield stresses can cause cracking. Thus, catastrophic failure can occur without significant deformation or obvious deterioration of the component. Very small amounts of hydrogen can cause hydrogen embrittlement in high strength steels. Common causes of hydrogen embrittlement are pickling, electroplating and welding, however hydrogen embrittlement is not limited to these processes. Hydrogen embrittlement is an insidious type of failure as it can occur without an externally applied load or at loads significantly below yield stress. While high strength steels are the most common case of hydrogen embrittlement all materials are susceptible.

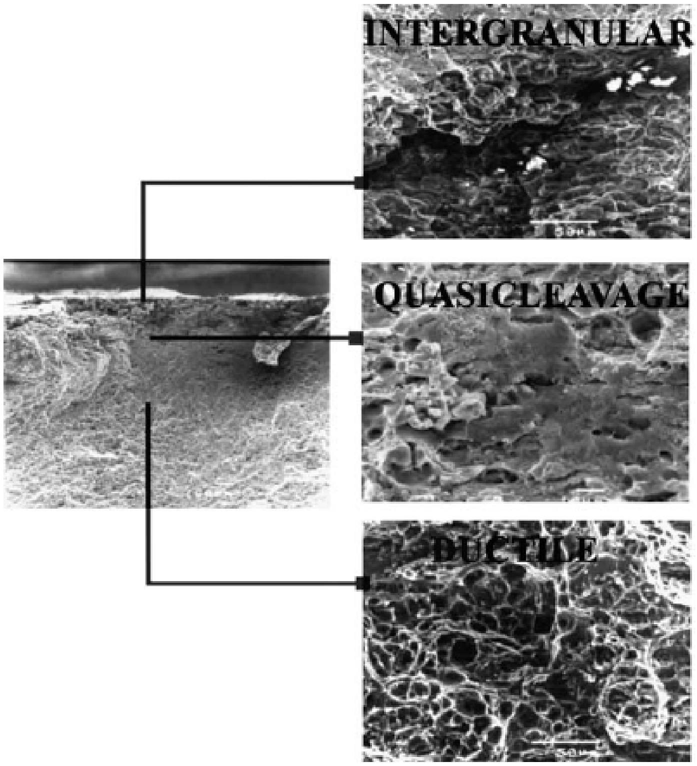

Fig. 7: Fracture Surfaces of Corrosion-induced Hydrogen Embrittlement in Aluminium Alloy 2024 (Kamoutsi, H., 2006).

Hydrogen embrittlement can be also recognized by fractographically from the cleavage aureole around the pores and non-metallic inclusions, originated from humid feed material. Where when the material is tested with tensile testing, the fracture of a tensile specimen is illustrated in which a large number of fish eyes can be detected as in figure 8. The fracture happened during a tensile test which had a reduced tensile strength from 487 to 529 N/mm2 (specified value: 540 N/mm2) (figure 9).

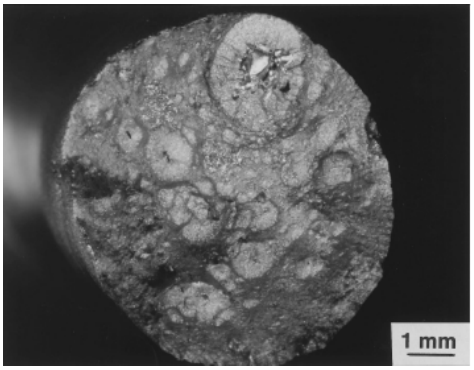

Fig. 8: Fish eyes on the fracture surface (Woodtli, J., R. Kieselbach, 1999).

One of the hydrogen embrittlement indications is blistering phenomena. The embrittlement is due to the fact that the excess dissolved hydrogen disperses into inclusions, pores or microcracks and because of high pressure where it is in the form of blistering flakes.

Figure 10 shows blistering phenomena on a galvanised steel surface (unalloyed carbon steel tempered to 460 HV5) (Woodtli, J., R. Kieselbach, 1999). Apparently these washers were not fully degassed. They failed after a relatively short time in operation due to low ductility cracks and fractures even though the load at a torque of 70 Nm was very low. The fracture surface (figure 11) indicates typical characteristics of hydrogen embrittlement in the form of a partially intercrystalline fracture with ductile markings on the grain boundaries (crow's feet). This fracture pattern may be a typical indication but it is nevertheless not a satisfactory proof of hydrogen embrittlement. In this particular case the residual hydrogen content of up to 1.49 ppm was determined by a gas chromatograph. This hydrogen concentration was above the lower brittleness threshold which lies between 0.3 and 3 ppm due to the particular material (Woodtli, J., R. Kieselbach, 1999).

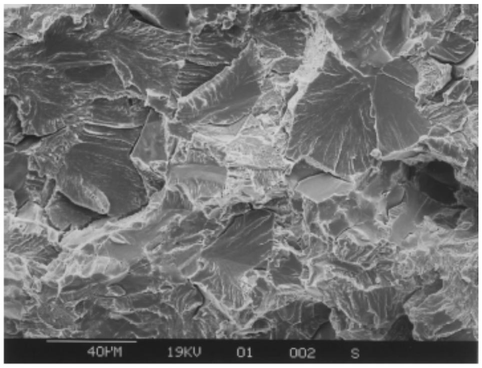

Fig. 9: Cleavage fracture of the aureole of a fish eye region (Woodtli, J., R. Kieselbach, 1999).

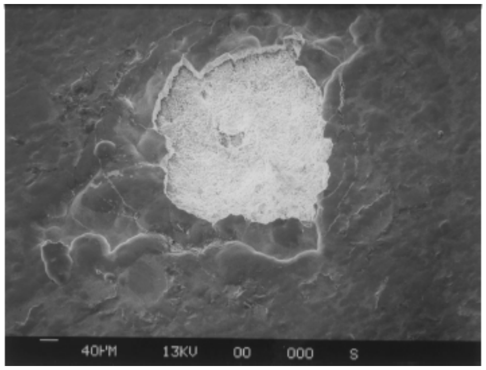

Fig. 10: Opened bubble on a galvanized steel surface (Woodtli, J., R. Kieselbach, 1999).

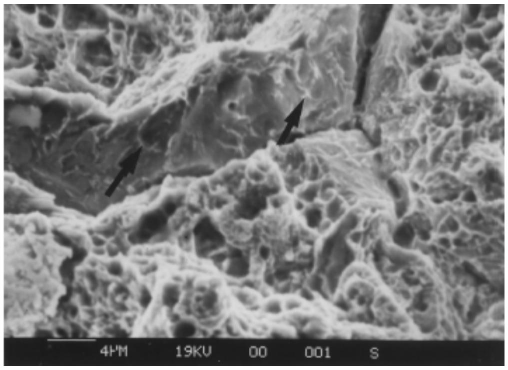

Fig. 11: Partially intercrystalline fracture with ductile marks on the grain faces (Woodtli, J., R. Kieselbach, 1999).

With the presence of deposit layer in internal tubes, the deposit corrosion will release atomic hydrogen which can migrate into the tube wall metal. The hydrogen atom will react with carbon in the steel and this causes intergranular separation. The reaction between the hydrogen atom and carbon in steel will decrease the carbon content of steel and this is called decarburization. Decarburization can be identified by metallographic examinations and results with microstructural degradation of the tube material (Eyckmans, M., 1999). Decarburization results in a reduction of tensile strength and an increase in ductility and creep rate. Steel will tend to lose strength after a long term of exposure to hydrogen at elevated temperature (Eyckmans, M., 1999). Figure 12 shows an example of microstructure of 1060 steel that showed decarburization. Carbon has a large influence on the mechanical properties of steel. The decreasing carbon content causes a degradation of these properties, as the hardness as well as the strength decrease. However the elongation of the metal when subjected to a tensile stress increases. On the other hand, increasing the carbon (carburization) decreases the toughness of the steel (Mars G. Fontana, 1987).

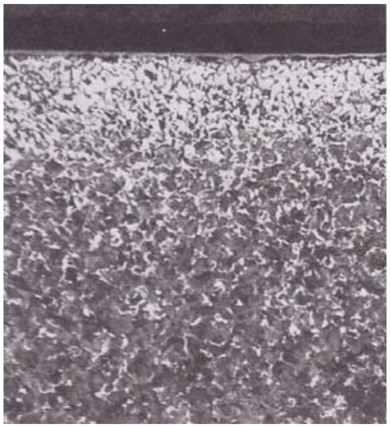

Fig. 12: Decarburized 1060 steel heated at 1205°C at 100X magnification (Arlan O. Benscoter, 1998).

Conclusions:

It can be concluded that the contribution factors for a metal tube to fail consists of temperature, pressure and environment that contains hydrogen. From visual examination, hydrogen damage can be identified by rough fracture features, presence of oxide scale on internal wall, either brittle/ ductile fracture or both and cleavage fracture. For further details of metallographic examination, hydrogen damage can be identified by either transgranular crack or intergranular crack, cleavage facets with ductile markings, blistering marks and indication of decarburization.

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