

Hydrogen Embrittlement in ferrous and non-ferrous metals (ACEA7010)

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1. INTRODUCTION

As the world shifts towards a net zero energy policy, the necessity to eliminate carbon-based fuels and reducing agents becomes increasingly crucial. Renewable energy sources are being leveraged to produce hydrogen through electrolysis, which can then be used as a clean fuel and a potential reducing agent in processes such as iron ore reduction. However, storing and transporting hydrogen presents significant challenges due to its small atomic size, high reactivity, and impact on material structures. Additionally, hydrogen poses issues throughout the metal processing lifecycle, from extraction to casting and fabrication. The following paper presents a review of some of the challenges considering Hydrogen embrittlement in metals.

2. Origins of Hydrogen

Hydrogen is the oldest, lightest, and most abundant element in the universe, which plays an essential role on today's earth. [1] Hydrogen and helium atoms emerged around 379,000 years after the Big Bang as hot plasma of protons began to cool and expand, electrons and protons gathered to form atoms. Four Hundred million years later, stars, such as the sun, evolved from gravitationally collapsed clouds of hydrogen gas, providing heat necessary to sustain life. A third breakthrough, 4.4 billion years ago, was when temperature on earth dropped below 100°C allowing dihydrogen oxide to condense at its surface, allowing the emergence of life in the new aqueous environment. [1]

Today hydrogen is estimated to account for 90% of all atoms in the universe, and it is essential to the material world. Close to two-thirds of the human body exists out of hydrogen. The first element of the periodic table makes for an excellent chemical fuel — one that has been attracting increasing attention. The early Earth's atmosphere was rich in hydrogen, and bacterial enzymes called hydrogenases evolved to generate energy from molecular H₂ or H₂O [2].

Van Helmont was the first to discover that while hydrogen was combustible in air, it could not support combustion by

itself. In 1671, Robert Boyle described the formation of gas bubbles from the reaction of iron filings with acid, but it was Henry Cavendish who recognized hydrogen (referred to as 'inflammable air') as a distinct substance. He discovered that burning hydrogen in 'dephlogisticated air' (oxygen) produced water. This finding led Lavoisier to name the substance 'hydrogen,' meaning water-former, in 1783. In 1800, Nicholson and Carlisle, followed by Ritter, managed to decompose water into its elemental constituents using electrolysis. This is a process we still strive to optimize today, aiming for lower energy consumption through photochemical methods. The evolved hydrogen gas is an excellent, ultra-light energy carrier and holds great promise as an abundant, environmentally friendly fuel, since its oxidation produces only water. Molecular hydrogen was used to fill one of the first balloons for human flight in 1783 and later fueled rockets, enabling further exploration. [2]

For practical applications, hydrogen must be stored in a compressed, liquefied, or solid state. In 1970, researchers at Philips Research laboratories accidentally discovered that hydrogen could be reversibly absorbed by intermetallic compounds, forming hydrides. This discovery led to significant advancements in electrochemical hydrogen storage, culminating in the mass production of nickel-metal-hydride battery-powered vehicles in Japan in 1997. Alongside this, there has been vigorous development in hydrogen-oxygen fuel cells and solid proton conductors [3,4].

Because hydrogen (H) and molecular hydrogen (H₂) represent the prototypical atom and molecule, respectively, they have been extensively studied by theoreticians since the inception of quantum mechanics over a century ago. These two species have served as crucial test beds for rigorous evaluations of diverse quantum mechanical models and approximations. The oxidation states of hydrogen range from -1 (hydride), through 0 (elemental), to +1 (proton), each exhibiting distinct physicochemical properties. The H₂ molecule, isoelectronic with the closed-shell helium (He) atom in the unified atom model, is notably inert. Kubas [5] described the coordination of molecular H₂ to transition metals. In contrast, the H⁻ anion is a very strong base and reducing agent, while H⁺ is a potent acid and powerful

oxidizer; slightly hydrated protons in a superacidic environment readily convert alkanes into carbocations. Hydrogen has been pivotal in developing theories of acidity and basicity, which are now understood as proton transfer reactions in the Brønsted-Lowry theory [6].

3. Structural changes due to hydrogen absorption into metals

The *ASM Materials Handbook* [7] lists five specific types of hydrogen-induced damage to metals and alloys. These types are:

- (1) hydrogen embrittlement,
- (2) hydrogen-induced blistering,
- (3) cracking from precipitation of internal hydrogen,
- (4) hydrogen attack, and
- (5) cracking from hydride formation.

Except for hydrogen embrittlement, a phase transformation is coupled to each of the listed hydrogen damage process. [8]

3.1 Solid State Metals

Hydride Formation:

Intermetallic Hydrides: Metals such as palladium, titanium, and zirconium form intermetallic hydrides (MH_x) when they absorb hydrogen. These hydrides have distinct crystal structures compared to the base metals. For example, palladium forms palladium hydride (PdH_x) with an expanded face-centered cubic (FCC) structure. [9,10]

Brittle Hydrides: The formation of brittle hydrides, such as titanium hydride (TiH₂), leads to a significant reduction in ductility and toughness. [10]

Lattice Expansion:

Interstitial Occupation: Hydrogen atoms occupy interstitial sites (tetrahedral or octahedral) within the metal lattice, causing lattice expansion. This expansion induces internal stresses and distorts the metal lattice. [10]

Volume Increase: The overall volume of the metal increases due to lattice expansion, which can affect the dimensional stability of metal components. [11]

Phase Transformation:

Martensitic Transformations: In some steels, hydrogen absorption can induce martensitic transformations, changing the phase composition without significant diffusion. This can lead to the formation of hard and brittle martensite phases. [12,13]

Hydride Stabilization: Hydrogen can stabilize certain phases that are not stable at room temperature, leading to phase changes that alter the mechanical properties of the metal. [14,15]

Hydrogen Embrittlement:

Crack Initiation and Propagation: Hydrogen embrittlement occurs as hydrogen atoms localize at areas of high stress (grain boundaries, dislocations, and crack tips), leading to crack initiation and propagation. This significantly reduces the ductility and fracture toughness of the metal. [16]

Delayed Fracture: Metals can experience delayed fracture where cracks develop and propagate over time under constant stress due to hydrogen diffusion and accumulation. [17,18]

Microstructural Changes:

Dislocation Density Increase: Hydrogen absorption increases the dislocation density in metals, as hydrogen atoms interact with dislocations and impede their movement. [19,20]

Microvoid Formation: Hydrogen can promote the formation of micro voids and cracks, especially at grain boundaries and other defects, leading to a decrease in mechanical integrity. [21,22]

3.2 Liquid State of Metals

Solubility Effects:

High Solubility: Hydrogen has a higher solubility in liquid metals compared to solids. This can lead to a significant absorption of hydrogen in the molten state. [23]

Gas Bubble Formation: If the hydrogen concentration exceeds its solubility limit during solidification, gas bubbles can form, leading to porosity and defects in the solidified metal. [24,25]

Viscosity and Density Changes:

Viscosity Reduction: Hydrogen absorption generally reduces the viscosity of liquid metals, which can affect casting processes and fluid dynamics. [26,27]

Density Variations: The absorption of hydrogen can change the density of the liquid metal, impacting buoyancy and the behavior of the melt during solidification. [28]

Structural Inhomogeneity:

Segregation: Hydrogen can cause structural inhomogeneity in the liquid metal, leading to segregation of hydrogen-rich and hydrogen-poor regions. This segregation can result in non-uniform properties in the solidified metal. [29]

Phase Separation: In some alloy systems, hydrogen absorption can lead to phase separation in the liquid state, forming distinct hydrogen-rich and hydrogen-poor phases. [30]

Solidification Effects:

Nucleation and Growth: Hydrogen affects the nucleation and growth of crystals during solidification. It can promote the formation of finer grains or lead to dendritic growth, depending on the alloy system. [31,32]

Defect Formation: Hydrogen can cause defects such as porosity, microcracks, and segregation during solidification. These defects are detrimental to the mechanical properties of the solidified metal. [33,34]

4. Metallographic Examination using Microscopy

Metallographic examination using optical microscopy and electron microscopy is essential for identifying and analyzing the structural changes in metals due to hydrogen absorption. Both techniques provide complementary information that helps in understanding the microstructural alterations and defects induced by hydrogen.

4.1 Optical Microscopy

Optical microscopy involves using visible light and lenses to magnify the surface and internal features of a material sample, allowing for the observation of microstructural elements such as grain boundaries, phases, and defects at magnifications up to around 1000x. It is useful for identifying larger-scale features and overall microstructural organization in materials.

Grain Boundaries: Changes such as embrittlement or cracking due to hydrogen absorption. [35]

Micro voids and Cracks: Hydrogen-induced microvoids and microcracks, particularly in high-stress regions or the heat-affected zone (HAZ) of welds. [36]

Hydride Formation: In metals like titanium and zirconium, optical microscopy shows hydride phases as distinct regions within the grains or along grain boundaries. [37]

4.2 Electron Microscopy

Electron microscopy, including both Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM), uses a beam of electrons to achieve much higher magnifications and resolutions compared to optical microscopy. SEM provides detailed surface morphology and compositional information, while TEM allows for atomic-scale imaging and analysis of internal structures, revealing fine details such as dislocations, grain boundaries, and precipitates.

Scanning Electron Microscopy (SEM):

High resolution: SEM provides higher resolution imaging, revealing detailed surface features and microstructural defects. [38]

Surface Morphology: SEM shows surface morphology changes due to hydrogen, such as blistering or surface cracks. [39]

Fractography: Examination of fracture surfaces using SEM identifies features indicative of hydrogen embrittlement, such as intergranular fracture patterns. [40]

Transmission Electron Microscopy (TEM):

Atomic-Scale Resolution: TEM offers atomic-scale resolution to observe hydrogen's effects on the crystal lattice. [41]

Dislocation Structures: TEM reveals changes in dislocation structures and densities due to hydrogen absorption. [42]

Hydride Precipitation: TEM provides detailed images of hydride precipitates and their interactions with the metal matrix. [43]

5. Effect of Hydrogen Absorption on mechanical properties on ferrous and non-ferrous metals.

5.1 Ferrous metals

Steels

Hydrogen Embrittlement (HE): Hydrogen embrittlement is a major issue in steels. It causes a reduction in ductility and toughness, leading to brittle fracture. This occurs because hydrogen atoms diffuse into the steel and localize at areas of high stress, such as grain boundaries, dislocations, and crack tips, leading to crack initiation and propagation. [44]

Decreased Tensile Strength: The presence of hydrogen can decrease the tensile strength of steel by promoting crack initiation and growth under tensile loading conditions. [45, 46]

Delayed Fracture: Steels exposed to hydrogen can experience delayed fracture, where the material fails under a constant load after a period of time due to hydrogen diffusion and accumulation at crack tips. [47,48]

Lattice Expansion and Phase Transformation: Hydrogen can cause lattice expansion and promote the formation of brittle phases such as martensite in certain types of steels, further reducing ductility. [49,50]

Cast Irons

Graphitisation: In cast irons, hydrogen can interact with carbon to form methane (CH₄), which can lead to porosity and reduce the mechanical strength and impact resistance. [51]

Hydrogen-Induced Cracking: Similar to steels, cast irons can also suffer from hydrogen-induced cracking, although they are generally more resistant due to the presence of graphite

flakes or nodules which can act as barriers to crack propagation. [52]

5.2 Non-Ferrous Metals

Aluminium Alloys:

Hydrogen Porosity: During solidification of aluminum alloys, hydrogen can form gas porosity, which severely reduces the mechanical properties such as strength, ductility, and fatigue resistance. [53]

Hydride Formation: Aluminum can form aluminum hydride (AlH₃), which is brittle and can negatively impact the mechanical integrity of the alloy. [54]

Reduced Weldability: Hydrogen absorption can lead to weld defects such as porosity and cracks in aluminum welds, compromising the structural integrity of welded joints.

Titanium Alloys:

Hydride Formation: Titanium readily absorbs hydrogen and forms brittle titanium hydrides (TiH_x), which significantly reduce ductility and toughness. [55]

Embrittlement: Similar to steels, titanium alloys can also experience hydrogen embrittlement, leading to a loss of ductility and the risk of sudden brittle failure.

Delayed Cracking: Hydrogen absorption can cause delayed cracking in titanium alloys under sustained loads, similar to delayed fracture in steels.

Nickel Alloys:

Hydrogen Embrittlement: Nickel alloys, particularly those used in high-temperature and corrosive environments, can suffer from hydrogen embrittlement, resulting in reduced ductility and premature failure. [56]

Hydride Formation: Some nickel alloys can form nickel hydrides, which are brittle and can degrade mechanical properties. [57]

Copper Alloys:

Hydrogen Embrittlement: While copper and its alloys are generally less susceptible to hydrogen embrittlement compared to other metals, certain high-strength copper alloys can experience embrittlement under specific conditions, leading to reduced ductility and toughness. [58]

Hydrogen-Induced Cracking: Hydrogen can cause intergranular cracking in some copper alloys, particularly in environments containing ammonia.

Summary of Mechanical Property Effects:

Ductility and Toughness: Hydrogen absorption generally decreases ductility and toughness across both ferrous and

non-ferrous metals by promoting brittle fracture mechanisms.

Strength: While the ultimate tensile strength may remain unaffected or even slightly increase due to hydrogen-induced phase transformations, the overall structural integrity is compromised due to embrittlement.

Fatigue Resistance: Hydrogen absorption reduces fatigue resistance by facilitating crack initiation and propagation under cyclic loading conditions.

Fracture Toughness: The presence of hydrogen lowers fracture toughness, making the material more prone to catastrophic failure under stress.

6. Hydrogen Absorption Effect on Processing and Weldability

Hydrogen absorption in metals can significantly affect various processing techniques, especially welding. These effects can lead to defects, reduced mechanical properties, and challenges in maintaining the integrity of welded structures.

Cold Cracking: Also known as delayed cracking or hydrogen-assisted cracking, cold cracking occurs after the weld has cooled to room temperature. This is primarily due to the presence of diffusible hydrogen in the weld metal and heat-affected zone (HAZ). Hydrogen atoms migrate to regions of high stress and combine to form molecular hydrogen, creating pressure that initiates cracks. [59,60]

Heat-Affected Zone (HAZ) Cracking: The HAZ is particularly susceptible to hydrogen-induced cracking due to the combined effects of thermal cycles and the presence of hydrogen. This region can develop microcracks that propagate under stress, compromising the structural integrity of the weld. [61]

Hydrogen Porosity: During welding, hydrogen can be absorbed into the molten weld pool and subsequently form gas bubbles as the weld solidifies. These bubbles become trapped in the solidified weld, leading to porosity. This significantly reduces the mechanical strength and toughness of the weld. [62]

Reduced Weld Quality: Porosity and gas entrapment lead to a reduction in the fatigue resistance and overall quality of the weld, making it more susceptible to failure under cyclic loading conditions. [63]

Microstructural Changes: Hydrogen can cause microstructural changes in the weld metal and HAZ, such as grain boundary embrittlement and the formation of brittle phases. These

changes can adversely affect the mechanical properties of the weld, including its toughness and ductility. [64]

Increased Residual Stresses: The absorption and subsequent release of hydrogen can induce residual stresses within the weld and the surrounding base metal. These stresses can lead to warping, distortion, and an increased likelihood of stress corrosion cracking. [65]

Process considerations:

Preheating and Post-Heating: To mitigate the effects of hydrogen absorption, welding procedures often require preheating and post-heating treatments. Preheating helps to reduce the cooling rate of the weld, allowing hydrogen to diffuse out of the weld metal before it solidifies. Post-heating or hydrogen bake-out treatments help to remove residual hydrogen from the weld. [66]

Use of Low-Hydrogen Consumables: Using low-hydrogen welding consumables (such as electrodes and filler materials) is essential to minimize hydrogen absorption during welding. These consumables are designed to produce less hydrogen when heated. [67]

Weld Repair and Maintenance:

Difficulty in Repairs: Hydrogen-induced defects in welds, such as cracks and porosity, can complicate repair efforts. Removing these defects often requires grinding out the affected areas and rewelding, which can be time-consuming and costly. [68]

Inspection and Monitoring:

Regular inspection and monitoring of welds are necessary to detect hydrogen-induced defects early. Non-destructive testing methods, such as ultrasonic testing and radiographic inspection, are commonly used to identify and assess the extent of these defects. [65]

6. CONCLUSIONS

Hydrogen plays a crucial role in the transition to a net zero energy policy, offering a clean, efficient, and abundant energy source. However, its storage, transport, and integration into existing processes pose significant challenges. Hydrogen's small atomic size and high reactivity lead to issues such as hydrogen embrittlement, microstructural changes, and defect formation in metals. These challenges are particularly evident in welding and other fabrication processes, where hydrogen-induced cracking and porosity can significantly degrade material properties.

Advancements in hydrogen storage, such as the discovery of reversible hydrides and the development of nickel-metal-hydride batteries, demonstrate the potential for practical applications. However, to fully realize hydrogen's benefits, further research and development are needed to optimize storage methods, improve material compatibility, and reduce costs.

Recommendations include continued innovation in electrochemical and photochemical hydrogen production methods to enhance efficiency and lower energy consumption. Employing low-hydrogen consumables and implementing rigorous preheating and post-heating procedures in welding can mitigate hydrogen-related defects. Additionally, regular inspection and non-destructive testing of welded structures are essential to ensure long-term integrity and safety.

In summary, while hydrogen presents promising opportunities for sustainable energy, addressing its associated challenges through advanced materials science and engineering practices is vital for its successful implementation.

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