

REVIEW ARTICLE

HYDROGEN STORAGE: A REVIEW

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ABSTRACT

Hydrogen storage is concerned with the process of storing hydrogen safely and efficiently. This is crucial because hydrogen may hold the potential as a green clean energy source to better exploit the energy obtained via renewable sources, although it is also highly flammable and can be difficult to store. In order to enrich the literature and further enhance the current understanding of hydrogen storage details, this review paper shortly discusses use of compression of hydrogen to high pressures, hydrogen liquefaction and metal hydride(s) for the purpose of storage of hydrogen. Results of the reviewed work have indicated that each process has its own advantages and disadvantages in terms of storage density, safety and energy requirements, etc., although; in general, the advantages defeat disadvantage. Proper hydrogen storage is a significant component in hydrogen utilization for different applications to help decarbonize related sectors.

KEYWORDS

Renewable, compression, liquefaction, metal hydride, gravimetric storage density.

1. INTRODUCTION

Fossil fuels, that comprise crude oil, natural gas and coal, are by no-means sustainable nor renewable but are with a destructive effect(s) on the environment (Demirbas et al, 2004; Kaygusuz, 2004; Balat, 2005). Annually, large amounts of such fossil fuels are consumed for electricity generation, vehicle transportation and other industrial applications, etc., to mention a few. Accordingly, large amounts of greenhouse gases and other toxic gases such as carbon mono/dioxide, methane, nitrous oxides, nitrogen oxides, halogenated gases, sulphur dioxide, black and organic carbon and ammonia are inappropriately emitted directly into the environment despite continuous effort to reduce their concentration(s). Such gases trap part of the heat that results when sunlight heats the earth's surface, i.e., global warming. In order to render this a history, renewable energies have been proposed as an alternative to such fossil fuels worldwide. Such energies, on the other hand, are clean, sustainable and as the name implies, renewable. Among renewable sources are solar, biomass, wind and hydrothermal/ tidal energies, etc. Of these sources, solar, biomass and wind are quite abundantly available; thus, they have dominated the energy sector if it is acceptable to disregard site-specificity and intermittence of solar and wind energies, respectively. Another positive factor with these sources is that the technological advancements related to them have led to great cost reductions. However, the storage of energy obtained out of such sources is not fully a well-established technology. Although battery can be used as a storage medium, it has several disadvantages, such as low storage capacity, short equipment life and a large amount of waste generated.

Hydrogen has gained significant attention as a potential green clean energy source to better exploit the energy obtained via renewable sources. Hydrogen has also been used as an energy carrier on its own. Technologies of hydrogen production have been reviewed elsewhere (Bidattul Syirat Zainal et al., 2024). Environmentally friendly hydrogen can be produced from water through an electrolysis process powered by solar panels or

wind turbines. In this review, it was shown that produced hydrogen can be converted into electricity via a fuel cell with no bad emissions/waste but water. Therefore, the significance of economy of hydrogen has been well-recognized in some countries including Australia, China, Germany, Japan, UK and US. Nevertheless, efficient, safe and economic storage of light weight gaseous hydrogen, that's to store hydrogen in a reversible manner at a high gravimetric and volumetric density, has still been a challenging task. In fact, it is not only hydrogen production that can help in the decarbonization of energy and industrial sectors, etc., but also proper hydrogen storage. Storage of hydrogen has been performed via technologies include but not limited to: high-pressure gas compression, liquefaction or metal hydride storage. It is the subject of this review paper to review these technologies.

2. HIGH- PRESSURE GAS COMPRESSION

Depending on the pressure and temperature, hydrogen molecule(s) can exist in several forms as indicated in the phase diagram, Fig. 1. This figure illustrates the different states that the hydrogen molecule (H₂) can take based on temperature and pressure. At low temperatures, hydrogen is a solid with a density of 70.6 kg/m³ at -262 °C and is a gas at higher temperatures with a density of only 0.089886 kg/m³ at 0°C and a pressure of 1 bar. The liquid hydrogen at a temperature of -253 °C is shown to exist in a narrow zone that stretches from the triple point to the critical point. Its density is 70.8 kg/m³ (Andreas Zuttel, 2004). Hydrogen is a gas at room temperature (298.15 K), and the Van der Waals equation describes it as following:

$$P(V) = \frac{nRT}{V - nb} - a \cdot \frac{n^2}{V^2}$$

Where: P is the gas pressure, V the volume, T the absolute temperature, n the number of moles, R the gas constant, a the dipole interaction or

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repulsion constant and b is the volume occupied by the hydrogen molecules (Weast RC (ed), 1976). It is due to strong repulsion interaction between hydrogen molecules, the low critical temperature ($T_c = 33\text{K}$) of hydrogen in the gas state. In principle, storage of hydrogen refers to reducing the massive volume of hydrogen gas as 1 kg of hydrogen at the

room temperature and atmospheric pressure occupies 11 m^3 . Either work must be performed to compress hydrogen, the temperature must be lowered below the critical point, or hydrogen must interact with another material to reduce repulsion in order to increase the density of hydrogen in a storage system (Andreas Zuttel, 2004).

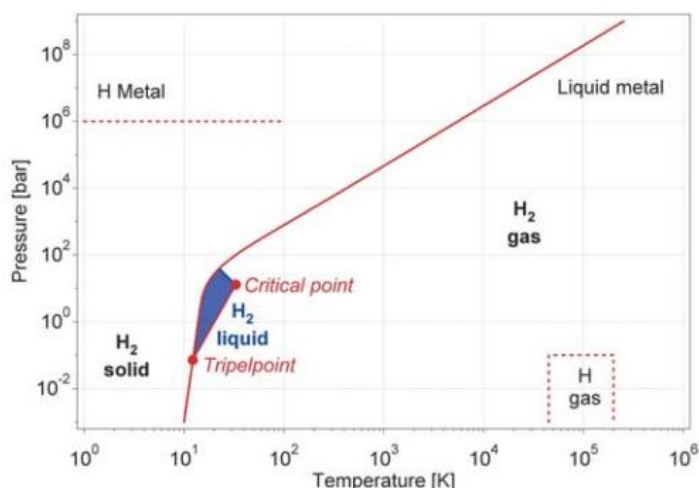


Figure 1: Phase Diagram for Hydrogen (Andreas Zuttel, 2004).

The development of the hydrogen infrastructure depends heavily on the methods of hydrogen storage. Not only does the mode of storage of hydrogen influence how it is transported, but it also influences how hydrogen is used. Thus, the field of hydrogen uses can be further expanded and promoted by advancements in hydrogen storage technology. It is the aim of compressing gaseous fuels, such as natural/town gas and hydrogen, to extremely elevate their pressure while lowering their volume which can be translated into an increased storage capacity in the gas state. For hydrogen compression for the purpose of hydrogen storage, two different compression modes (mechanical and non-mechanical compressors) are generally utilized in order to boost the volume density. Mechanical compressors, which use more than half of the capital expenditure of a hydrogen re-fueling station (Sdanghi et al., 2020). Include: reciprocating piston compressors, diaphragm compressors, screw compressors and centrifugal compressors. On the other hand, non-mechanical compressors, which do not have any moving parts, consist of metal hydride compressors, electrochemical compressors, adsorption compressors and cryogenic compressors (Orlova et al., 2023). Discussing such compressors requires a mechanical engineering background which is not the case. Hence, it has been decided to be sufficed with what has already been mentioned. However, there is a detailed discussion in a study carried out by Orlova, et al. for which interested readers are directed to. The study compared the performance data of mechanical and non-mechanical compressors considering some key factors such as efficiency, deliverable flow rates, pressure capability, cost and the due required maintenance (Orlova et al., 2023). Once compressed, hydrogen is ready for storage in a storage vessel of a certain type, as would be explained in two paragraphs ahead, the choice among them is a trade-off between the performance and cost.

Filling such a highly compressed hydrogen gas in hydrogen storage vessels in a re-fuelling station requires rigorous safety measures, obviously due to high pressure characteristics involved. To this end, several investigations have been carried out on the behavior of the process of charging of high-pressure gaseous hydrogen into a storage vessel aiming to propose a number of computational fluid dynamic models that consider flow and temperature field (Heitsch et al., 2009; Melideo et al., 2017; Li et al., 2019; Gonin et al., 2022; Kim et al., 2022; Xue et al., 2022; Zhao et al., 2022). Nevertheless, it is beyond the scope of this review to further consider this point. In re-fuelling stations, the stored high pressure compressed hydrogen can; then, be dispensed as a fuel for fuel cell vehicles via a hydrogen dispenser, much alike to the traditional gasoline/diesel pump.

Initially, the first type of hydrogen storage vessels were made out of either steel or aluminum requiring up to 500 Kg to build a steel vessel to store 25 Nm^3 of hydrogen at a pressure of no any higher than 12 MPa (Miao Yang et al., 2023). High pressure storage vessels has been built using materials including aluminum, steel, carbon fiber, polyethylene and epoxy resin, etc. If steel vessels are to be used for hydrogen storage for fuelling a vehicle, they are considered too heavy (Takeichi et al., 2003). A storage capacity of 5 Kg of hydrogen in a vehicle that last for 500 to 700 Km, requires a high-pressure storage vessel with a volume of 0.18 m^3 (Leung et al., 2004). In

fact, in this application both gravimetric density and volumetric density must be high in order to increase the amount of hydrogen that can be stored in an effort to widen the travelling range (Kruse et al., 2002). Furthermore, use of steel vessels to store high-pressure hydrogen, as the case with storing gases at high- pressures of up to 700 bar, is not a good practice, due to hydrogen embrittlement failure due to hydrogen diffusion into steel. It should be noted that embrittlement failure can be further pronounced in case of a frequent charge and discharge of vessel(s) with hydrogen. In order to eliminate the embrittlement failure issue, steel storage vessels can be replaced with vessels built from composite materials while chemically inert with hydrogen such as polyethylene/carbon fiber or epoxy resin coupled with thin aluminum liner (Takeichi et al., 2003). Also, with the use of such vessels to store high-pressure hydrogen, the gravimetric storage density is quite low ($0.01\text{ Kg H}_2/\text{Kg}$). On the contrary, use of this technology to store hydrogen is linked to an energy storage efficiency of 94% (Takeichi et al., 2003). Compared to only 75% when battery storage is used (Linden, 1995; Chan, 2000). However, the energy storage efficiency may decline with excessive pressure increase, although this might boost the volumetric storage density.

Today's first type hydrogen storage vessels, compared to other types, still withstands the lowest pressure (15-30 MPa), and is the cheapest as it costs \$240/Kg hydrogen stored, and with the lowest gravimetric storage density (which accounts for the ratio of the mass of gas stored to that of the vessel (Sdanghi, 2020; Barthelemy et al., 2017; Biaeck et al., 2018; Legault, 2012; Rivard et al., 2019; Kruse et al., 2002). Due to low gravimetric density, this vessel type is merely used for stationary applications to store hydrogen as an industrial gas (Miao Yang et al., 2023; Barthelemy et al., 2017). For this type, it is recommended to hoop-wrap the central part of the vessel by a resin-impregnated fibre in order to cope with higher pressures (Miao Yang et al., 2023). The second type comprises a metal with a composite of glass and fibre (Sdanghi et al., 2020; Miao Yang et al., 2023; Biaeck et al., 2018). This type exhibits the highest-pressure tolerance with a quite wide range of pressure (10-100 MPa), which may justify its high cost at \$360/Kg hydrogen stored (Legault, 2012). It is used for stationary applications such as refueling stations (Parks et al., 2014). Fully composite wrapped with a metal liner and fully composite with a high-density polyethylene inner supported by glass/carbon fibre, presents the third and fourth types of hydrogen storage vessels which are quite light in weight. They both can accommodate pressures between 30 and 70 MPa (Sdanghi et al., 2020; Miao Yang et al., 2023; Biaeck et al., 2018). Both are more expensive than the other two types and can be used for hydrogen tube trailer and for on-board hydrogen storage (Elgowainy et al., 2014; Miao Yang et al., 2023). In terms of gravimetric storage density, it improves as we head up from the second type towards the forth. Further details on processes of manufacture of pressure vessels can be found in a study by Yang and co-workers (Miao Yang et al., 2023).

In fact, it is not only hydrogen embrittlement, as pointed out above, what might result in the damage of steel hydrogen storage vessels of either generations, but also hydrogen-induced cracking and high-temperature

hydrogen attack, although hydrogen embrittlement stands out as the most hazardous among them, posing the most severe potential harm (Ohaeri et al., 2018; Hao Li et al., 2022). Due to their significance, they were a target of several research work studies (Ohaeri et al., 2018; Hao Li et al., 2022; Hardie et al., 2006; Kane, 2008; Moro et al., 2010; Melaina et al., 2013; Slifka et al., 2014; Drexler et al., 2016; Meng et al., 2017; Djukic et al., 2019). It has been reported that these phenomena are all caused by a common cause, that's the tendency of hydrogen molecules to react with the steel. The phenomenon of hydrogen embrittlement in steel storage vessels is a multifaceted occurrence where hydrogen interacts with the metal storage vessel or metal additives to form solid solutions, metal hydrides, molecular hydrogen as well as gaseous products, e.g., methane. This interaction can diminish the bonding strength of metal grain boundaries as a result of a reduced vessel plasticity, leading to a brittle fracture, microscopic cracking or pitting as indicated in Figure 2 (Hao Li et

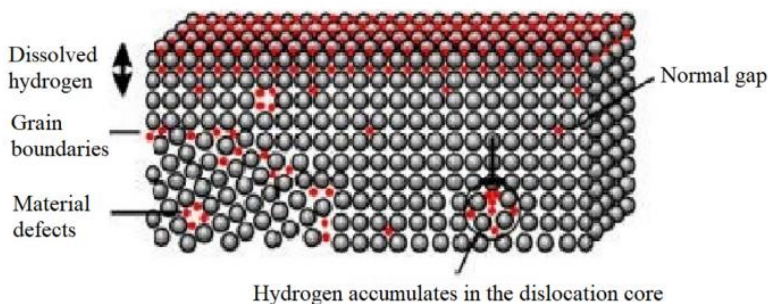


Figure 2: Material Defects and Hydrogen Accumulation Location (Hao Li et al., 2022).

Furthermore, the phenomenon of hydrogen-induced cracking in steel storage vessels is frequently associated with stress corrosion cracking which results in vessel failure (steel cracks). This is attributed to the collaborative interaction between hydrogen and steel vessel stress (serious hydrogen embrittlement) (Ohaeri et al., 2018). The third important phenomenon in this regard is high-temperature hydrogen attack which can be explained as following. If hydrogen contacts the steel metal at a high temperature and pressure, hydrogen molecules/atoms attack the metal and becomes absorbed (dissolved) within the metal internals due a synergetic effect of this high temperature ($> 200^{\circ}\text{C}$) and pressure. This absorbed hydrogen interacts with the alloy and/or impurity elements found within the microstructure of the metal to produce insoluble gaseous product, high-temperature hydrogen attack, as the name implies. This dissolved hydrogen within the metal microstructure is so mobile and tends to constantly accumulate at the grain boundaries, inclusions or other microstructure defect pores. In these pores, the atoms of dissolved hydrogen unite again to produce hydrogen molecules which; then, at the grain boundaries, react with impurities carbon to give methane in addition to the reaction of decarboximization and cementite decomposition of steel at high temperatures ($> 200^{\circ}\text{C}$). Hence, the produced methane gas holds an internal pressure and creates bulges and cracks in the steel metal storage vessel in the form of elongated holes. This can seriously trigger the hydrogen embrittlement effect leading to catastrophic damage to hydrogen storage vessels (Djukic, et al., 2016).

However, the energy requirements for hydrogen compression, for a given mass and at a given compression ratio, are higher due to its lower specific gravity compared to other gaseous fuels (Ananthachar and Duffy, 2005). Also, in terms of the environmental and safety issues of both the materials used to construct a high pressure storage vessel as well as the stored hydrogen, they are neither environmentally damaging nor dangerous. In fact, in case of a hydrogen leakage into the atmosphere, it flows upwards and quickly disappears, due to its lighter weight than air. Thus, it is likely that hydrogen would accumulate at the sources of leakage. However, care should be paid regarding the flammability of hydrogen, since its flammability range, by hydrogen volume, lies between 4.1 and 74.8% at only at 1 bar and in a dry air. For a safe operation; therefore, sufficient ventilation should be in place to ensure hydrogen dilution to avoid ignition (Utgikar and Thiesen, 2005).

3. LIQUEFACTION

In comparison to the storage of hydrogen in the gas form via high pressure hydrogen gas compression, liquefaction of hydrogen gas is to store it in a safer liquid state to benefit from the higher volumetric and gravimetric storage densities (Miao Yang et al., 2023). A liquefied hydrogen can be used as a fuel for internal combustion engines and fuel cells, etc. According to a study by Takeichi et al., the volumetric and gravimetric storage densities of a liquefied hydrogen lie between 20 to 50 kg H_2/m^3 and 8 to

al., 2022). According to a study by Melaina and others, the occurrence of hydrogen embrittlement is a function of pressure, purity and moisture content of the stored hydrogen pressure as well as strength level and deformation rate of the storage vessel. Surrounding temperature also plays a role in hydrogen embrittlement. Generally, as the pressure in a storage vessel is increased, the risk of hydrogen embrittlement would accordingly increase (Haeseldonckx and D'haeseleer, 2007). However, it was reported that such a relationship is assessed on a case-by-case basis in a way that there is no consensus among specialists on all hydrogen embrittlement mechanisms and their interactions (Hao Li et al., 2022; Melaina et al., 2013). Despite this; nevertheless, the application of molecular simulations and some cutting-edge technologies, e.g., transmission electron microscopy and atom probe technology, has led to a gradual clarification of the mechanism of hydrogen embrittlement (Hao Li et al., 2022).

25 kg H_2/kg ; respectively, with a calorific value of 120 MJ/Kg (Takeichi et al., 2003). However, it should be made crystal clear that liquefaction of hydrogen is a complicated and energy-intensive process if compared to the process of liquefaction of other gases or the previously discussed compression of hydrogen process (Miao Yang et al., 2023). Low evaporation temperature at the atmospheric pressure and critical point of hydrogen at 20.28 K and 33 K; respectively, are what in part justify the difficulty of liquefaction of hydrogen. The last is also affected by the small size of a molecule of hydrogen to the extent it is comparable to the size of molecules of an ideal gas but at a higher temperature. Its Joule-Thomson coefficient can only be positive if the temperature is less than 202 K. Otherwise, the temperature would increase, a paradox to the principle of liquefaction. At last, what further complicates the liquefaction of hydrogen is the lower enthalpy of vaporization of para-hydrogen at 447 KJ/Kg at the critical point of hydrogen (20 K) than that of exothermic conversion of normal hydrogen to equilibrium hydrogen which is 532 KJ/Kg at the same temperature as a result of conversion of ortho-to-para hydrogen (Valenti, 2016)

The simplest liquefaction cycle is the Joule-Thomson cycle. Further developments, resulted in the invention of some other newer cycles and systems for hydrogen compression such as Linde-Hampson cycle and Claude system, etc., refer to Figure 3. In order to liquefy hydrogen, it undergoes compression followed by cooling using heat exchanger(s) down to under the phase inversion temperature (202 K). In order to attain a storable cryogenic liquid hydrogen with a boiling temperature of 20 K, liquefied hydrogen is fed to an expansion unit (a throttle valve, where Joule-Thomson expansion occurs). To avoid complication, further details of the liquefaction cycle are omitted from this review and can be found elsewhere (Andreas Zuttel, 2004; Michel et al., 1998; Barron, 2000; Krasae-in et al., 2010; Leachman, 2015; Cardella et al., 2017; Swanger et al., 2017; Petitpas et al., 2018; Derking, 2019; Matveev and Leachman, 2021)

To avoid any phase inversion of the stored liquefied hydrogen, i.e. evaporation (boil-off loss) due to heat absorbed from the ambient, thermally insulated vessels are usually used. For this purpose, materials of the support system with a low thermal conductivity and with a high mechanical strength are used to minimize heat transfer via conduction. Similarly, to reduce heat transfer by conduction through interconnecting piping system, it is suggested to reduce the cross section while increase the length of pipes (Miao Yang et al., 2023). To reduce heat transfer by natural convection, heat flows should be kept minimal and evacuated double walls are used, whereas reflective metallic foils are used to cut off radiation (Andreas Zuttel, 2004; Bracha et al., 1994). Protecting the storage vessel with baffles, optimizing the design and avoiding direct exposure of warm parts of the vessel can help mitigate radiation strength. Another approach to further reduce hydrogen boil-off down to only 0.01% and 0.04% per day for large and small storage vessel; respectively, is to implement a multilayer insulation technology coupled with a high vacuum

of about 10^{-4} mbar (Miao Yang et al., 2023). Zero hydrogen boil-off loss can be achieved, if a refrigeration system is integrated with such a multilayer insulation technology (Swanger et al., 2017). For safety considerations since the metallic inner vessel is by no means designed to hold higher pressures (Miao Yang et al., 2023). However, it is necessary to arrange for a ventilation away from any ignition source to accommodate hydrogen

vapor (Gursu et al., 1992). Recently, it has become more and more popular to effectively exploit hydrogen vapor for cooling the hot compressed hydrogen gas, reducing the boil-off loss to less than 1% per day (Petitpas, 2018; Derking, 2019). For hydrogen vapor to form (boil-off), it requires nearly 3 days after charging the vessel with liquid hydrogen and it accounts for 0.1 to 3% of the total liquid hydrogen stored (Amos, 1998).

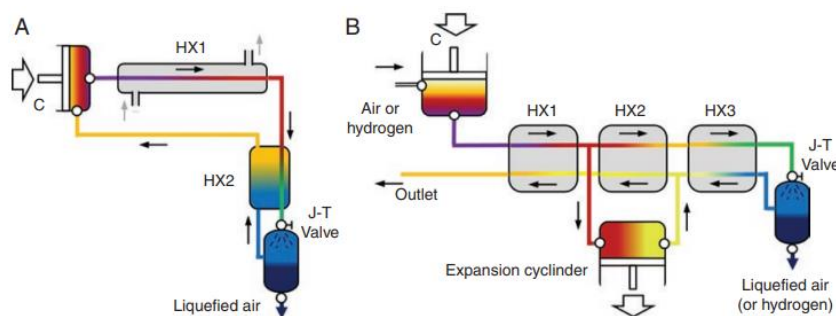


Figure 3: (A) Linde-Hampson Cycle and (B) Claude System for Hydrogen Liquefaction (Miao Yang et al., 2023).

Hydrogen boil-off depends on the shape, size and thermal insulation of storage vessel of liquid hydrogen. Small spherical storage vessels are the best choice to reduce hydrogen boil-off due to their least surface to volume ratio and due to their uniform distribution of stress and strain. Large spherical storage vessels; however, have seen lesser applications due to their high cost. As the boil-off is proportional to the surface to volume ratio, it largely decreases as the size of the storage tank increases. For a double-walled vacuum-insulated spherical Dewar vessel with a storage capacity of 50, 100 and 20000 m³, the daily boil-off losses are: 0.4, 0.2 and 0.06%, respectively (Andreas Zuttel, 2004). In terms of energy storage density per liter of hydrogen stored, it is estimated at 5 MJ/ liter (Thomas and Keller, 2003). However, this process requires higher energy requirements than high pressure hydrogen gas compression (Andreas Zuttel, 2004; Amos, 1998). In addition, continuous boil-off of hydrogen limits the applicability of liquid hydrogen storage systems (Andreas Zuttel, 2004). Also, unless high gravimetric and volumetric storage densities are essential as the case with vehicles as well as space applications, storage of hydrogen through liquefaction is not an attractive choice due to low energy efficiency, high cost and high energy requirements (Andreas Zuttel, 2004; Meng Ni, 2006). In terms of the environmental and safety issues, the process of storage of hydrogen through liquefaction is less environmental considerate than high-pressure hydrogen gas compression process due to lower energy efficiency which is a synonym of high pollutant emissions (Meng Ni, 2006). Furthermore, it has been reported in several studies that storage of hydrogen through liquefaction is more hazardous than high-pressure hydrogen gas compression storage (Hord, 1978; Edeskuty et al., 1979; Knowlton, 1984; Peschka et al., 1987). This can be attributed to the following: In case of leakage of liquid hydrogen, which is heavier than air which is heavier than hydrogen gas, it flows downwards and accumulates prior to vaporization. Also, it requires an arrangement for ventilation to accommodate any hydrogen vapor (Gursu et al., 1992). Third, it suffers from clogging of safety valves and the vent of the storage vessel by ice as a result of cooling of moist air. This has the sequence of rupture of the storage vessel due to pressure build-up. In this process, there is a high risk of ignition, fire or explosion if the stored liquid hydrogen is contaminated with air during charging and discharging, although maintaining the storage vessel at a pressure higher than the atmospheric pressure may help prevent this happening (Meng Ni, 2006).

4. METAL HYDRIDE STORAGE

Among all chemical elements, hydrogen is the lightest. It is capable to produce a hydride with all chemical elements apart with inert gases. To produce a metal hydride, hydrogen is dissolved exothermically into a metal/alloy (host metal) when the ratio of hydrogen atoms to metal atoms is minimal (< 0.1) (Andreas Zuttel, 2004; Sanusi Kazeem Olajide, 2020). Metal hydrides are fundamental to the following processes: chemical processing (reducing agents, strong bases, strong reductants, catalysts), physical separation processing (desiccants, isotope separation, gas separation, and hydrogen purification), nuclear engineering (neutron moderators, reflectors, and shields), thermal applications (heat pumps), energy storage (hydrogen fuel tanks and secondary batteries) and energy conversion by means of alkaline fuel cells, etc. (Young, 2013). Since the topic of renewable energy is at the foundation of many of these applications, metal hydrides and their uses have been a busy area of research in recent years.

Metal hydrides can be synthesized via reactions in the gaseous phase, in

solution or in solids formed from other hydrides. Several metal hydrides can be synthesized as a product of a hydrogenation reaction that involves directly reacting hydrogen with an elemental metal, intermetallic compounds or as alloy. A typical hydrogenation reaction advances the step of initial heating to an elevated temperature of up to 1100 K at which the rate of hydrogen absorption is small although fast. In order to attain the temperature that balances kinetics and capacity of hydrogenation reaction and at which the hydride forms, a step of slow cooling in hydrogen environment is required. However, owing to its exothermic nature, a hydrogenation reaction can be completed despite heat loss during the cooling step. Occasionally, it is required to use a spark to 'ignite' (overcome the activation barrier) the hydrogenation reaction. The dependence of a hydrogenation reaction on pressure is that some metal hydrides to be synthesized require high hydrogen (equilibrium) pressures between 103 and 109 Pa, while other hydrides do not require such a high pressure to form depending on the metal or intermetallic compounds used. Furthermore, employment of high pressures in a hydrogenation reaction helps stabilizes new phases of metal hydrides having high coordination numbers or high metallic oxidation states. In numerous instances, the kinetic of a hydrogenation reaction negatively affected by lower temperatures, although low temperatures are with a positive effect on hydrogen storage (Sanusi Kazeem Olajide, 2020). In a study by Young, K., it was found that metal systems with larger unit cell volumes tend to create more stable hydrides. On the other hand, metal systems with high levels of disorder can exhibit a variety of local cell volumes, i.e., a better storage capacity and adsorption/desorption kinetics (Young, 2013).

Metal hydride hydrogen storage is somewhat new compared to storing hydrogen via high pressure gas compression and liquefaction. Storage of hydrogen by metal hydride(s) is a method to store hydrogen within a solid form. This method is the safest among other methods and with which the highest volumetric hydrogen storage density can be attained (Miao Yang et al., 2023; Sanusi Kazeem Olajide, 2020). Because of their special ability to both absorb and desorb hydrogen, metal hydrides can be utilized to store or release hydrogen on demand. This ability largely depends on the hydriding conditions. Undoubtedly, the most crucial characteristic of a metal hydride is its ability to reversibly store hydrogen, allowing it to be utilized as a solid hydrogen storage material (Sanusi Kazeem Olajide, 2020). Hydrogen molecules and metals, alloys or some intermetallic compounds via chemical bonding generate metal hydride(s). Intermetallic compounds, such as LaNi₅ and Ti-based body-centered cubic alloys such as FeTi alloy, suffer from very low gravimetric hydrogen storage density of 1.28 wt.% and 1.9 wt.%; respectively, due to which their application for hydrogen storage has been limited. Therefore, selected metal and metal alloys materials have been limited to those light metals with a reasonable hydrogen storage density such as Al, B, Be, Li, Mg, Na and Pd, Mg₂Ni, MgN₂, NaAl, Ti and Ti₂Ni (Miao Yang et al., 2023; Stange et al., 2005; Shashikala, 2012). Other combinations such as are also in use: LaNi_{4.7}Sn_{0.3}, MmNi_{4.6}Fe_{0.4}, MmNi_{4.6}Al_{0.4}, Nd (Ni_{1-x} Cu_x)(In_{1-y} Al_y), LaNi_{4.96}Al_{0.04}, La_{1.06}Ni_{4.96}Al_{0.04} and Fe_{0.9}Mn_{0.1}Ti and Ti_{0.64}Zr_{0.36}Ni, to mention a few (Stange et al., 2005; Muthukumar et al., 2005; Riabov et al., 2005; Heung, 2003; Cuevas et al., 2005). Historically, use of metals in this application goes back to 1866 when Graham, T. observed the capability of Pd metal to absorb a large quantity of hydrogen. Later, metal hydrides were also involved in this area following realizing reversible hydrogen absorption and desorption on intermetallic compounds (Shashikala et al., 2012). Among these metal hydrides, metal hydrides based on magnesium (MgH₂) and related alloys are the most promising materials for solid-state

hydrogen storage along with the high hydrogen absorption potential, relative affordability and good reversibility (Zaluska et al., 1999). However, in order to ensure the cycle stability of MgH_2 formed, magnesium particles are to be well protected from gaseous impurities such as O , N_2 , CO and CO_2 , as otherwise, they are capable to inhibit or slow down hydrogen absorption (Pedersen and Larsen, 1993).

Hydrogen can be dissolved exothermically into the metal/alloy when the ratio of hydrogen atoms to metal atoms is minimal (< 0.1). Within the metal lattice structure, the hydrogen atoms occupy the interstitial sites to produce interstitial hydrides leading to the absorption of large amounts of hydrogen at a constant pressure, into the metal hydride storage by which heat is generated which ranges between 9300 and 23250 KJ/ Kg of hydrogen absorbed (Andreas Zuttel, 2004). This heat can be simultaneously recovered in order to lessen the loss in the capacity of storage caused by temperature increase. On the other hand, equal heat is on demand while desorption of hydrogen, which occurs at a temperature greater than 500 °C, during hydrogen discharge (Amos, 1998). This required heat can be secured by using the recovered heat from the adsorption step or via using the wasted heat of fuel cells. Although the operating pressure can be higher than 100 bar, charging and discharging pressures should not exceed 30 and 2 bar, respectively, due to practical and economic considerations (Heung, 2003). For all metal-hydrogen systems, the principles governing the hydrogen-metal interaction are invariably replica. Because atomic hydrogen is injected into a matrix of host metal atoms of the metal hydrides suitable for hydrogen storage, this kind of metal hydride is also known as an interstitial hydride. However, certain sites can be simultaneously occupied by hydrogen. For hydrogen with an atomic radius of 0.53 \AA to occupy a site, the site must meet certain criteria that its radius should be greater than 0.40 \AA and that the distance between two consecutive occupied sites is greater than 2.10 \AA (Young, 2013). Additional information about the chemistry of storing hydrogen in metal hydride storage can be found elsewhere (Andreas Zuttel, 2004).

In this method of hydrogen storage, it is not only the gravimetric hydrogen storage density, as stated above, that matters, but also the following: safe operation while fast kinetics, chemical and thermal stability of the generated metal hydride(s) during multiple cycles of charge/discharge (Meng Ni, 2006; Sakintuna et al., 2007). Utmost care should be practiced while handling those metal hydrides comprising heavy alkaline metals as they are tremendously reactive. Also, due to the sensitivity of several metal hydrides, high stability against oxygen/air and moisture should also be a feature of metal hydrides; otherwise, they should be stored under inert atmosphere (Sanusi Kazeem Olajide et al., 2020). Low dissociation temperature with moderate pressure, low heat of dissipation during exothermic formation of the hydride, low heat of formation to reduce the energy required for hydrogen release and low cost of the infrastructure of recycling and charging, are all also important factors. In addition, energy loss during hydrogen charge/discharge should be kept minimal to help limit spontaneous hydrogen release (Sakintuna et al., 2007).

Storing of hydrogen via metal/alloy hydride is a suitable choice for stationary power plants where the heavy weight of the storage material is not a major issue. However, the volumetric storage density of metal/alloy hydride storage is so high, greater than $100 \text{ kg H}_2/\text{m}^3$ (Meng Ni., 2006). Nevertheless, its gravimetric storage density is quite low ($0.015 \text{ kg H}_2/\text{kg}$, $0.02\text{-}0.06 \text{ kg H}_2/\text{kg}$, $0.03 \text{ kg H}_2/\text{kg}$ and $0.07 \text{ kg H}_2/\text{kg}$) (Story, 2000; Meng Ni., 2006; Amos, 1998; Barbir and Veziroglu, 2003). This can be attributed to the heavy internal structure of the metal/alloy used. Another issue with metal hydride hydrogen storage is the ultra-heavy weight of the storage material for use in mobile applications (vehicles). On the contrary, use of this technology to store hydrogen is linked to an energy storage efficiency of 88%. In terms of the environmental and safety issues associated with the process of storage of hydrogen through metal hydride, are the disposal of spent materials of metals/alloys and those of compressor and storage vessel, while used for storing hydrogen at a high pressure, as well as the energy requirements (Meng Ni, 2006).

5. CONCLUSION

Among hydrogen storage processes reviewed are compression of hydrogen to high pressures, hydrogen liquefaction and metal hydride(s). Different storage hydrogen processes can be applied in different hydrogen applications. Highly compressed hydrogen gas can be used for both stationary and mobile applications for industrial applications and for on-board hydrogen uses, respectively. Liquefaction of hydrogen offers storage of hydrogen in a liquid state for internal combustion engines and fuel cells. Storing of hydrogen via metal hydride(s) is a suitable choice for stationary power plants where the heavy weight of the storage material is not a major issue. Of these, use of chemically stable metal hydride(s) to store hydrogen, seems the efficient and safest. It is also advantageous over

other two processes reviewed due to its high volumetric storage density, although its gravimetric storage density, mainly due to heavy internal structure of the metal/alloy used, is otherwise.

Environmentally, use of high-pressure compression process to store hydrogen in a storage vessel is environmentally benign. However, the process of storage of hydrogen through liquefaction is less environmentally considerate and more hazardous than high-pressure hydrogen gas compression process due to lower energy efficiency which is a synonym of high pollutant emissions. Similarly, use of metal hydride(s) is associated with the issues of disposal of spent materials of metals/alloys and those of compressor and storage vessel, while used for storing hydrogen at a high pressure, as well as the energy requirements.

To sum up, for each of reviewed storage processes, there are advantages and disadvantages in terms of storage density, safety and energy requirements, etc., although the former far outweighs the latter. Storage vessels employed for of storage of high-pressure hydrogen gas; in particular, suffer from embrittlement, hydrogen-induced cracking and high-temperature hydrogen attack, which collectively can lead to catastrophic failure of storage vessel(s).

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