An Overview of Hydrogen Storage Technologies

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ABSTRACT

How to store hydrogen efficiently, economically and safely is one of the challenges to be overcome to make hydrogen an economic source of energy. This paper presents an overview of present hydrogen storage technologies, namely, high-pressure gas compression, liquefaction, metal hydride storage, and carbon nanotube adsorption. The energy efficiency, economic aspect, environmental and safety issues of various hydrogen storage technologies were compared. Presently, high-pressure gas compression is favorable due to its high energy efficiency as well as low capital and operation costs. Liquefaction is mainly employed in space applications because of its high volumetric and gravimetric efficiency. The disadvantages are low energy efficiency and high cost. Due to their high volumetric efficiency, metal hydride storage and carbon nanotube adsorption are promising hydrogen storage technologies and are expected to play a key role in hydrogen economy in the future.

Keywords: Hydrogen economy; Hydrogen energy; Hydrogen storage; Renewable energy; Environment pollution; Fuel cell

1. INTRODUCTION

Using fossil fuels as the primary energy source has led to serious energy crisis and environmental pollution on a global scale (Demirbas et al. 2004; Kaygusuz, 2004; Balat M., 2005b). In Hong Kong, fossil fuels consumed directly for vehicle transportation and generation of electricity amount to approximately 36% and 48%, respectively, of the estimated total of 290,000 terajoules per year (Ni et al. 2005a). About 40 million tons of greenhouse gases and toxic gases are emitted every year due to combustion of fossil fuels (Ni et al. 2005b).

In order to resolve the above environmental problems, it is of paramount importance to develop clean, renewable energy technologies (Kaygusuz, 2001). Solar, wind, biomass, hydrothermal and tidal energies are potential renewable energy sources (Vamvuka and Tsoutsos, 2002; Demirbas, 2002a, 2002b; Demirbas, 2003; Demirbas M.F., 2004; Balat M., 2004; Balat M., 2005a; Balat H., 2005). Especially, solar, biomass, and wind energy are abundant resources and may dominate the energy

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market in the future as the technological development has brought down the costs to a competitive level. However, solar and wind energy are site-specific, intermittent, and thus, not reliable. 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. Therefore, in order to better utilize renewable energy, hydrogen has been identified as a potential alternative fuel as well as an energy carrier for the future energy industry. Powered by solar cells or wind turbines, hydrogen can be produced from water via electrolysis. When hydrogen is converted into electricity via a fuel cell, the only by-product is water (Ni, 2005c). Thus, from its life cycle point of view, hydrogen is environmentally friendly. In many countries, such as the United States, China, Japan, Australia, Germany, Turkey, UK, et al., the importance of hydrogen economy has been recognized (Tseng et al., 2005; Sherif et al., 2005; Winter, 2005). In order to realize hydrogen economy, one of the challenges need to be resolved is to store hydrogen efficiently, safely, and economically. Presently, there are four candidate hydrogen storage technologies available: (1) high-pressure gas compression, (2) liquefaction, (3) metal hydride storage, and (4) carbon nanotube adsorption. This paper attempted to give an overview of these hydrogen storage technologies. Their scientific aspect, economic consideration, and environmental as well as safety issues are compared and summarized in this paper.

2. HIGH-PRESSURE GAS COMPRESSION

2.1. Scientific basis

Gas compression to low volume and high pressure is a commonly used storage method for gaseous fuels. The apparent difference between compression of hydrogen and compression of other conventional fuel gases, such as natural gas and town gas, is the energy requirement. As hydrogen has a lower specific gravity than other fuel gases, it takes more energy to compress hydrogen for given mass and compression ratio (Ananthachar and Duffy, 2005).

The volumetric storage density (H2-kg/m^3) of hydrogen at 25^{o}C can be calculated by 0.0807P, based on thermodynamics. The expression is derived from the ideal gas law, where P is the storage pressure in bars. For example, at typical P = 350 bars, the volumetric density is 28 H2-kg/m^3. For a storage capacity of 5 kg of hydrogen in a vehicle, that can travel 500 to 700 km before refilling, the high-pressure storage vessel should be sized to 0.18 m^3 (Leung et al., 2004).

The efficiency of energy storage by compressed hydrogen gas is about 94% (Leung et al., 2004). This efficiency can compare with the efficiency of battery storage around 75% (Chan, 2000; Linden, 1995). It is noted that increasing the hydrogen storage pressure increases the volumetric storage density (H2-kg/m^3), but the overall energy efficiency will decrease.

Steel vessels are commonly used for high-pressure gas compression storage with operating pressure as high as 700 bars. However, for hydrogen storage, steel is not a desirable material. It is because the diffusion of hydrogen into steel causes hydrogen embrittlement failure, especially when the vessels undergo frequent charge and discharge. In the case of rupture, steel projectiles may cause serious injuries. Furthermore, the gravimetric storage density, defined as the ratio of the mass of stored gas to the mass of vessel, is low, normally in an order of 0.01 H2-kg/kg. Steel vessels are too heavy for practical use in vehicles. The hydrogen embrittlement problem can be resolved by using vessels made of composite materials comprised of polyethylene, or carbon fiber and epoxy resin with thin aluminum liner (Takeichi et al. 2003).

2.2. Application and future development

As pressure vessels are simple to build and use, they are popular for hydrogen storage, especially for small-scale storage with frequent charge/discharge cycles. For vehicle application, pressure vessels of high gravimetric and volumetric storage densities are important. Kruse et al. (2002) presented a number of trial applications with pressure vessels in cars and buses from various manufacturers, including Ford, General Motors, Daimler-Chrysler, BMW, and Toyota. The common limitation of these vehicles is the short traveling range due to the limited amount of hydrogen that can be stored. More recently, GM announced that a GM/Opel prototype van equipped with a pressure vessel operating at maximum 700 bars could travel a maximum range of 270 km (Leung et al. 2004).

Continuous research aims to develop high-strength and light-weight materials for making pressure vessels with increased volumetric and gravimetric storage densities. The new materials should also be chemically inert with hydrogen to avoid hydrogen embrittlement. Mitlitsky et al. (2000) reported a long-term goal of achieving a gravimetric storage density of 0.12 H_2-kg/kg at 345 bars. The US DOE goal for vehicular hydrogen storage is to achieve a gravimetric storage density of 0.065 H_2-kg/kg efficiency and volumetric storage density of 62 H_2-kg/m^3 (Dillon et al., 2000).

2.3. Environmental and safety issues

Steel, aluminum, carbon fiber, epoxy resins, and polyethylene are materials commonly used for production of pressure vessels. These materials, consumed at a rate of about 20 to 100 kg for every 1-kg hydrogen storage capacity, are neither environmentally detrimental nor hazardous.

In general, the safety concerns for hydrogen storage are same as those for storage of common fuel gases. As hydrogen gas is much lighter than air, any hydrogen leak will flow upward and disperse quickly. Accumulation of hydrogen around the source of leakage is less likely in comparison with other fuel gases. Therefore, hydrogen is less hazardous. The flammability range of hydrogen in dry air at 1 bar is 4.1 to 74.8% by volume of hydrogen. For a safe operation, sufficient ventilation is required for hydrogen storage to ensure that any hydrogen leaks can be diluted to less than 1% by volume (Utgikar and Thiesen, 2005).

2.4. Economic aspect

The major components of a high-pressure gas compression storage system are the compressor and pressure vessel. Based on the data reported by Prince-Richard et al. (2000), Mitlitsky et al. (2000), and Amos (1998), the following cost figures could be deduced:

- Capital cost of compressor is US$80-380/kW of H_2.
- Capital cost of pressure vessel is US$40-1,300/$H_2$-kg. It is noted that the costs of compressed gas vessels reported by Amos (1998) are considerably higher than those by Mitlistsky et al. (2000) and Prince-Richard et al. (2000).

- Energy consumption is about 2.2 kWh/H2-kg.
- The annual operating and maintenance costs are about 0.01-0.05% of the capital cost.

3. LIQUEFACTION

3.1. Scientific basis

Hydrogen gas can be liquefied and stored in a thermally insulated vessel. Storage in liquid, hydrogen has higher volumetric as well as gravimetric storage densities than storage in compressed hydrogen gas. Hydrogen gas is compressed and cooled below the inversion temperature of 202 K. Subsequent expansion causes the formation of cryogenic hydrogen liquid at boiling point of -253^{o}C (20 K). The energy storage density has been estimated to be 5 MJ/liter (Thomas and Keller, 2003). With a calorific value of 120 MJ/kg, the volumetric storage density of hydrogen liquefaction is about 40 H_2-kg/m^3. Takeichi et al. (2003) reported the volumetric and gravimetric storage densities of about 20 to 50 H_2-kg/m^3 and 8 to 25 H_2-kg/kg, respectively.

The energy required to liquefy hydrogen for storage in an ideal Linde thermodynamic cycle has been calculated to be 11.88 MJ/H_2-kg (Leung et al., 2004), about 64% higher than the energy required for high-pressure hydrogen gas compression. Taking into account the caloric value of hydrogen of 120 MJ/H_2-kg, the energy efficiency of hydrogen liquefaction storage is 91%. Amos (1998) reported that the energy consumption would be 10 kWh/H_2-kg (36 MJ/H_2-kg), equivalent to an energy efficiency of 77% for hydrogen storage. It is possible to increase this efficiency by modification of the thermodynamics of the Linde cycle with multiple heat exchangers, compressors, and expansion valves. However, the equipment cost and corresponding maintenance cost will increase accordingly.

As the storage vessel gains heat from the ambient, the stored liquid hydrogen will gradually evaporate. For safety reasons, the hydrogen vapor is vented when the pressure exceeds the critical operating pressure, resulting in a boil-off loss (Gursu et al., 1992). In order to minimize the boil-off, the storage vessels are thermally insulated by materials of low thermal conductivity, evacuated double walls, and reflective metallic foils to reduce heat transfer by conduction, convection, and radiation, respectively (Bracha et al., 1994; Sherif et al., 1997). Typically, boil-off occurs about 3 days after a vessel is charged with liquid hydrogen and the boil-off rate ranges from 0.1% to 3% (Amos 1998). Smaller vessels tend to have a higher boil-off rate.

3.2. Application and future development

Due to the high cost and low energy efficiency, hydrogen liquefaction storage is only attractive when high gravimetric and volumetric storage densities are required, such as road vehicles and space applications. Road vehicles with 5-kg liquid hydrogen storage have been tested by automotive manufacturers including General Motors, DaimlerChrysler, and BMW (Kruse et al. 2002). The tests have shown the promise for further development of such applications.

Presently, hydrogen liquefaction plants in operation have liquefaction capacity range from 380 to 2,300 H_2-kg/hour (Amos, 1998). As high cost is one of the major drawbacks, current research works aim to improve the energy efficiency of liquefaction. Alternative magnetic liquefaction, that has the potential to achieve higher energy efficiency, is now being developed (Padro and Putsche, 1999). Better thermal insulation and higher operating pressure for minimizing boil off in liquid hydrogen storage have also been considered.

3.3. Environmental and safety issues

Hydrogen liquefaction storage is less environmental than high-pressure hydrogen gas compression because of lower energy efficiency. From the life cycle point of view, low energy efficiency means high pollutant emission. The methods to improve energy efficiency have been discussed in previous section.

In general, hydrogen liquefaction storage is more hazardous than high-pressure hydrogen gas compression storage (Hord, 1978; Edeskuty et al., 1979; Knowlton, 1984; Peschka, 1987), as: (1) Unlike hydrogen gas, liquid hydrogen is heavier than air. Therefore, in case of leakage, liquid hydrogen flows downwards and accumulates before it vaporizes; (2) Boil-off hydrogen vapor must be vented to a safe location clear from any source of ignition; (3) The safety valve and vent of a storage vessel may be clogged by ice due to the cooling of moist air. The subsequent pressure build-up may cause the vessel to rupture; (4) Liquid hydrogen is subject to contamination of air condensed onto equipment during repeated charging and discharging. The mixture can then be easily ignited. It is important to prevent air condensation or oxygen enrichment by proper insulation and sealing. Condensed air mist must not be allowed to drop onto combustible materials such as tar and asphalt to avoid creation of a local explosive mixture; (5) Air leaking into liquid hydrogen can lead to fire or explosion. The pressure of the storage vessel must be maintained above the atmospheric pressure to prevent air from entering the vessel.

3.4. Economic aspect

The cost of liquefier dominates the capital cost of hydrogen liquefaction storage. Amos (1998) estimated the capital cost per unit of liquid hydrogen production rate to be US$44,100/kg/hour. For example, a hydrogen liquefaction storage system of 300-kg/hour capacity, that can instantaneously charge 5 kg of liquid hydrogen in one minute, will cost about US$13 million. The operating cost, including the energy for compressor and cooling liquid nitrogen, is about US$½/kg.

4. METAL HYDRIDE STORAGE

4.1 Scientific basis

In metal hydride storage, hydrogen molecules are chemically bonded with metals or alloys to form metal hydrides. When the hydrogen to metal atomic ratio is small (< 0.1), the hydrogen can be exothermically dissolved into the metal. The hydrogen atoms occupy the interstitial sites of the metal lattice structure to form interstitial hydrides. Heat is generated during hydrogen charging of the hydride storage (absorption of hydrogen) and the same heat is needed to discharge the hydrogen (desorption of hydrogen). The metal hydride formed must be chemically and thermally stable under frequent charging and discharging cycles. Storage materials include Mg, Ti, Ti_2Ni, Mg_2Ni, MgN_2, NaAl et al., and various combinations, such as

$Nd(Ni_{1-x}Cu_x)(In_{1-y}Al_y)$ (Riabov et al., 2005), $Ti_{0.64}Zr_{0.36}Ni$ (Cuevas et al., 2005), $LaNi_{4.7}Sn_{0.3}$ (Stange et al., 2005), $MmNi_{4.6}Fe_{0.4}$, $MmNi_{4.6}Al_{0.4}$ (Muthukumar et al., 2005).

As hydrogen is absorbed, the heat generated must be removed or recovered simultaneously to minimize the loss in storage capacity due to the increase in temperature. The heat removed may be stored and later used to release the hydrogen from the hydride storage during discharging. Depending on the hydrogen absorbing material used, the heat generation ranges from 9,300 to 23,250 kJ/kg of hydrogen. The hydrogen desorption temperature can be above 500^{o}C (Amos 1998). The waste heat from fuel cells may be used to release the hydrogen. The operating pressure can exceed 100 bars. For practical and economic reasons, the charging pressure should preferably be less than 30 bars. The discharging process should be conducted at a pressure of about 2 bars and a temperature below 100^{o}C (Heung, 2003).

Although metal hydride storage has very high volumetric storage density (> 100 H_2 -kg/m^3), the gravimetric storage density is very low due to the heavy metals or alloys. Amos (1998) reported that the gravimetric storage density of hydrides storage is between about 0.02-0.06 H_2-kg/kg while Barbir and Veziroglu (2003) stated that the maximum gravimetric storage density is about 0.07 H_2-kg/kg. Hydrides storage working around ambient temperature and pressure would normally have gravimetric storage density of about 0.03 H_2-kg/kg.

Heat of reaction, will be 23,250 kJ/kg for absorption of hydrogen. Assuming that 50% of this energy can be recovered for desorption use, then the energy for one charging/discharging cycle will be about 11,600 kJ/kg. Assuming a pressure of 20 bars for hydrogen in the reactor vessel, the calculation gave the compression energy of 4,123 kJ/kg. The total energy required for hydrides storage will thus be (11,600 + 4,123) kJ/kg or 14.7 MJ/kg. With a CV of 120 MJ/kg for hydrogen, this can be translated into an energy storage efficiency of about 88% for metal hydrides storage.

4.2. Application and future development

Story (2000) tested a metal hydride storage using Hydralloy C15 (a German product) for a prototype fuel cell power plant for mining/tunneling locomotives. The gravimetric storage density is about 0.015 H2-kg/kg. Heung (2003) reported the demonstration of a 15-kg metal hydride storage for a city transit bus and a 2-kg storage for a John Deere Gator utility vehicle. The storage materials tested were LaNi4.96Al0.04, La1.06Ni4.96Al0.04, and Fe0.9Mn0.1Ti. The material cost was US$1,000/kg using Fe0.9Mn0.1Ti. It takes 1 to 2 hours to fully charge up the system at refueling pressure at 20 bars. The hydrogen discharge pressure and temperature were about 8 bars and 50oC, respectively. Sapru et al. (2002) claimed a successful development of a proprietary metal hydride storage device. The device was tested on an 80 c.c. scooter and the maximum speed achieved was 33 km/h.

Metal hydride storage is relatively new compared with pressure vessel storage and liquid hydrogen storage. The weight of storage materials is too heavy for commercial use in vehicles with reasonable traveling range. Metal hydride storage may be a suitable option when weight is not a major concern, i.e. stationary power plant. Further research and development in the following areas are needed: (1) Light-weight metals or alloys should be developed; (2) The speed of charging and discharging of hydrogen should be increased with more effective heat transfer. Jensen and Takara (2000) demonstrated that the use of catalyst could significantly enhance the reaction kinetics for NaAl storage; (3) Minimize deterioration in hydrogen storage capacity due to cycling of charging and discharging. (Gross et al. 2000); (4) Use hydrides storage in hybrid with pressure vessels to meet both volumetric and gravimetric storage constraints (Takeichi et al. 2003).

4.3. Environmental and safety issues

The main material consumptions will be the metal and alloys for hydrides storage, besides those required for compressor and storage vessel. Energy consumption will be the need to power the compressor and for heat transfer during the absorption and desorption process.

Metal or alloys hydrides would have to be disposed at the end of their life and if these metals or alloys were not hazardous, then they would not contribute to any hazardous wastes issues. The other environmental considerations as those for pressure vessel will also be valid for hydrides storage.

A metal hydride tank is considered to be a very safe fuel storage system. In the event of a collision or tank crack, the loss of pressure in the punctured tank will cool down the metal hydride, which will then cease to release hydrogen. Hydrides storage materials will expand when hydrogen is absorbed and 20% allowance for expansion should be considered in the system design (Heung 2003).

4.4. Economic aspect

The dominating capital cost is the cost of the storage materials. Therefore, the overall cost is very much linearly proportional to the storage capacity. Amos (1998) presented a number of capital cost estimates for hydrides storage ranging from US$820 to US$22,000 per kg. The estimates are consistent with the figures given by Heung (2003). Padro and Putsche (1999) deduced that the total capital investment per annual throughput is about US$18/MJ, which is equivalent to US$2,160/kg.

5. CARBON NANOTUBE ADSORPTION

5.1 Scientific aspect

Adsorption is such a phenomenon that gaseous molecules are diffused into the microspores of an adsorbent. The adsorption mechanism may involve both van der Waals adsorption and chemical bonding (Dillon, 2000). The amount of hydrogen that can be adsorbed is practically proportional to the specific surface area of the adsorption substrates. Activated carbon that has high specific surface area is a popular adsorbent. However, it has a wide distribution of pore sizes and only a fraction of the pores are of similar size to hydrogen molecules for effective hydrogen storage.

The use of carbon nanotubes with pores of a few nanometers is a promising candidate for hydrogen storage (Yoo et al., 2005; Panella et al., 2005). Hydrogen molecules can be adsorbed into the micropores and remain trapped in the cavities. Hydrogen can be released when the temperature is elevated. The most effective adsorption temperature for hydrogen is usually below 0°C and the operating pressure is below 100 bars (Ye et al., 1999, Padro and Putsche, 1999). Dillon et al. (2000), Padro and Putsche (1999) and Kruse et al. (2002) reported that 5 to 10% [0.1 kg/kg] gravimetric storage density at room temperature has been demonstrated. Dillon et al. (2000) further pointed out that the volumetric storage density could be up to 65 H_2-kg/m^3.

5.2. Application and technology status

The potential of carbon nanotube storage applied to vehicles is highly promising as: (1) the adsorbent material used is light-weight; and (2) the volumetric efficiency of hydrogen storage is high. Presently, the high cost of carbon nanotubes is the main barrier to overcome for commercialization. Active R&D efforts are being made to develop methods for producing nanotubes more economically.

Dillon et al. (2000) reported arc-generated carbon nanotube soots produced by co-evaporating cobalt/carbon mixtures in a spar-gap evaporator. Kruse et al. (2002) mentioned the use of laser technology to produce nanotubes with precise diameter and high level of purity needed.

6. SUMMARY OF HYDROGEN STORAGE TECHNOLOGIES AND CONCLUDING REMARKS

Among the hydrogen storage technologies discussed in the previous sections, their characteristics are summarized in Table 1 for easy comparisons. It is found that the pressure vessel technology is favorable because it is easy to implement with high storage energy efficiency and at low cost. The main drawback is low volumetric storage density. The problem will be lessened if the technology can be further developed to safe operation at higher storage pressure. Liquefaction is suitable for space applications because of its high volumetric and gravimetric efficiency. The disadvantages are the high cost and low energy efficiency. Metal hydride and carbon nanotube adsorption are promising hydrogen storage technologies as both the volumetric efficiency is very high and the gravimetric efficiency is comparable with high-pressure gas compression method. Therefore, metal hydride and carbon nanotube adsorption should receive more research efforts to realize the sustainable hydrogen economy.

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Table 1. Characteristics of candidate hydrogen storage technologies

Technology Types	High-pressure compressed gas vessels	Liquid hydrogen tank	Metal hydride	Carbon nanotube adsorption
Development Status	Commercially proven	Commercially proven	Quite developed but not yet commercially proven	Ongoing active research and development
Energy efficiency	About 93%	Less than 70%	About 88%	Unknown at this stage
Gravimetric efficiency	5 - 10%	8 - 25%	2 - 7%	5-10%
Volumetric efficiency	20 H2-kg/m3 at 350 bars storage pressure	20 - 50 H2-kg/m3	Above 100 H2-kg/m3	Up to 65H2-kg/m3
Applicability	Stationary and road vehicles	Mainly space vehicles	Target for road vehicles	Promise for road vehicles
Economic Considerations	Relatively least capital & operation costs	Very expensive capital and operation costs for liquefaction	More costly than pressure vessels in capital & operating expenses	Uncertain at this stage, manufacturing process yet to be developed
Environmental Considerations	Small impacts from construction and operation	Moderate impacts due to liquefaction in addition to those due to pressure vessel	Slightly higher impacts than those of pressure vessels due to hydrides disposal	Uncertain at this stage, manufacturing process yet to be developed
Safety Considerations	Typical pressure vessel precautions, monitoring & good ventilation needed	In addition to precautions for pressure vessels, care needed to avoid condensed air mixed with hydrogen	Relatively safe	Relatively safe