Various Techniques and Challenges of Hydrogen Storage: A Review

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Abstract-*With increase in human population present day situation has constrained us to face dual problems of fossil fuel depletion and environmental degradation simultaneously. The researchers are working towards finding out the best replacement for the fossil fuel; if not at least to offset the total fuel demand. Hydrogen is the most abundant element in the universe and a promising solution to the world's oil dependence. Since 1970s, hydrogen has been considered as a possible energy career for the storage of renewable energy. H2 is a promising replacement energy storage molecule because it has the highest energy density of all common fuels by weight.By observing the various properties of fuel; hydrogen is considered as a best option. Developing safe, reliable, compact, and cost-effective hydrogen storage technologies is one of the most technically challenging barriers to the widespread use of hydrogen as a form of energy. It can be produced by various processes but the challenging task is storage of hydrogen. This paper aims to study various techniques of hydrogen storage. Due to imminent release of commercial FCEVs by automobile manufacturers in regional markets, a dual strategy is being pursued to (a) lower the cost and improve performance of high-pressure compressed hydrogen storage systems while (b) continuing efforts on advanced storage technologies that have potential to surpass the performance of ambient compressed hydrogen storage.*

Keywords-Hydrogen storage, Mechanical storage, Metal hydrides, Nanostructures.

I. INTRODUCTION

Now a day, human society depends primarily on fossil fuels for energy production. So, this energy needed to be stored in effective and efficient way. By reviewing the present situation, it is clear that fossil fuels cannot continue to be the primary energy source because resources are depleting rapidly and due to burning of fossil fuels greenhouse gases are emitted continuously [1]. Motorized vehicles are the largest users of fossil fuels which burn gasoline in an internal combustion engine (IC). For limiting the use of fossil fuels first effort in automobiles was hybrid vehicle development. It contains a battery that carries an electric charge in addition to an internal combustion engine. Hybrid vehicles had some great achievements but they cannot be a solution because they still need some input of fossil fuels. Due to this, the next generation of automobiles will likely be fuel cells vehicles (FCV's). FCVs require hydrogen gas as fuel to power a hydrogen fuel cell. FCV's produce no environmentally damaging emissions, hence these are preferred. Also, once a reliable method for H2 gas production is developed, FCVs will supply fuel constantly [2]. Energy density by weight of hydrogen fuel is 142.0 MJ/kg which is three times larger than gasoline [3]. Space available on vehicle to store hydrogen is limited so tank need to be strengthened enough to prevent explosion during accidents. A compact, safe, reliable, effective and energy efficient technique of hydrogen storage is required so that FCV's will be commonly used.

It has been widely reported that hydrogen will be the fuel of the future, because its abundance on earth, it burns cleanly with water being the only by product, and it contains the highest energy on weight basis. In reality hydrogen is only a fuel carrier and energy is required to produce elemental hydrogen. In addition, hydrogen contains very small amount of energy on a volume basis, and volume counts the most when fuel is stored and transported. From the point when hydrogen is converted to work, energy consumed in storage, transport and other must be at a minimum to maximize the energy carried to do work. Storage is probably the most important step in this process. Hydrogen gas storage is challenging task because it is smallest molecule. Hydrogen has been stored in gaseous form for centuries and as a liquid since the 1940's but these techniques are financially expensive. As a result, many alternative methods of hydrogen storage are researched like storage within carbon nanotubes as well as in metal hydrides. Currently there is not a single hydrogen storage technology which meets DOE standards because each storage method has limits. Gaseous hydrogen requires large volume; liquid hydrogen evaporates easily; metal hydrides add significant weight to the system and carbon nanotubes are inflammable. Therefore, research is constantly underway to improve these storage methods and develop new storage techniques [4]. The current options for storage on board FCVs are discussed in the following sections.

II. MECHANICAL STORAGE

Mechanical hydrogen storage techniques are the techniques which stores hydrogen in promising and unchanged form. For increasing the volumetric and gravimetric capacity of hydrogen methods like compression and cryogenic cooling (Sections 2.1 and 2.2) were used significantly. Recently, this has improved to cryo compression mechanism (Section 2.3).

2.1 High pressure Tanks

The most common storage system is high pressure gas tanks, which are operated at a maximum pressure of 20 MPa. In fuel cell vehicles compressed hydrogen tanks are used to store hydrogen. This method is selected as at present situation it is simplified and best understood. The variables like weight, cost, safety etc. needed to be considered before high-pressure storage becomes a reliable option [2]. The most important factor which needed to be taken in account is the material composing the high-pressure vessel. It must withstand the embrittlement possible with hydrogen in addition to being inexpensive and easy to work with. At first all those vessels were manufactured from aluminium which has high conductivity, but after some observations researchers noted that vessels made from aluminium could not comply with safety standards and often were not strong enough. Due to this, in recent years researchers turned towards fibre composite i.e. carbon fibre reinforced plastic (CFRP) [2, 5]. Basically, CFRP vessels have two main advantages: (1) They are lightweight, (2) They are very strong, but have relatively low thermal conductivity. So, the material needed to be kept below 358 K at all times to match with safety requirements. During filling the compressed hydrogen tank, there is exothermic compression of hydrogen gas which increases temperature extremely. This temperature constraint can be an issue. The CRPF is fire sensitive unit which changes mechanical properties. There is a large concern that an explosion would result if the H2 storage vessel was subjected to a fire, such as might occur in a car crash [6, 7]. For the storage hydrogen in tanks cost is considered the greatest barrier. Even if the cost is lowered, there is concern that high pressure tanks will not be accepted by the general public because they are perceived as unsafe [2]. Compressed hydrogen tanks are bulky so they are used mainly for static applications where weight does not matter. Compressed hydrogen tank for automotive applications is shown in fig. 1.

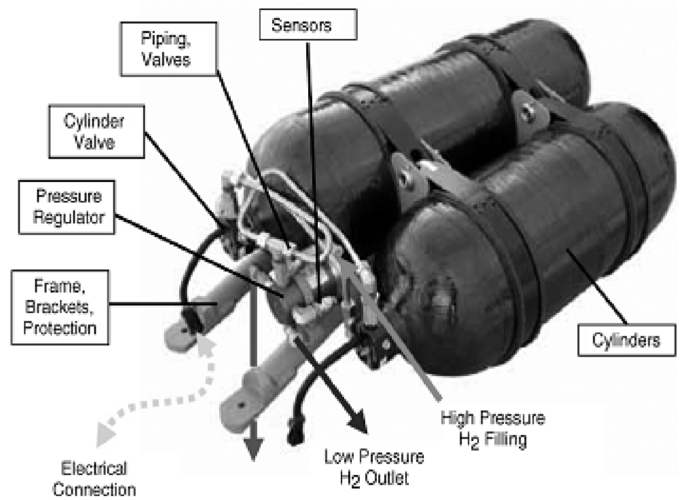

Fig. 1:Compressed hydrogen tank for automotive applications[37]

2.2 Cryogenic storage: liquid hydrogen

To store hydrogen as a liquid, the energy required to cool hydrogen to the liquid state is critical. Liquefying hydrogen is means of increasing volumetric energy density. It is also known as cryogenic storage as liquefaction is performed by cooling hydrogen to 20 K. Although, it is an energy expensive process, it increases hydrogen volumetric energy density from 2.5 or 5 MJ/L (for compressed H2 at 345 and 690 atm respectively) to 8 MJ/L (for liquid hydrogen LH2) [8]. As a result, less volume is required for storage so a smaller, lighter container can be used. This allows longer distances to be driven. Because of this great potential, cryogenic hydrogen storage was extensively studied in the 1980's and 1990's [9].

If hydrogen is stored in the liquid form on board a FCV's, it needed to be maintained below its boiling point of 20 K. Therefore, heat intrusion must be kept at the lowest possible level. To accomplish this, the original liquid hydrogen tanks were metallic vessels having two walls. The inner vessel had multilayer insulation composed of several metallic foil layers separated by glass wool; space between the inner and outer vessels was evacuated to create a vacuum [10]. In recent years, pressure release valves have been added for safety reasons. In addition, new tanks must be able to release gaseous hydrogen as well as liquid hydrogen. A liquid hydrogen tank was most recently tested in a GMHydroGen3; it was found to hold 4.6 kg of LH2 with a total system weight of 90 kg [11, 12].

Despite important improvements to volumetric density, LH2 storage is not preferably used for several reasons: (1) At least 35% of the fuel's energy content is used to liquefy it which is three times more energy than is needed to compress H2 to 690 atm, (2) LH2 evaporates very easily during delivery and refuelling, (3) LH2 pressurizes quickly while on-board vehicles as it absorbs heat from the environment. Therefore, the tank must be vented every 3-5 days during inactivity to prevent dangerous and costly boil-off losses; if boil-off is not controlled, the entire hydrogen store will evaporate in 2 weeks. Even if these problems could be overcome, the volumetric energy density of LH2 is 8 MJ/L which is substantially less than that of gasoline which requires 32 MJ/L and diesel fuel 36 MJ/L. As a result, studies of mechanical hydrogen storage have largely shifted to cryocompressed H2 (Section 2.3) in recent years [4, 13].

2.3 Cryocompressed hydrogen

Cryocompressed hydrogen storage is the technique in which two hydrogen storage methods i.e. compression and cryogenic storage of hydrogen are combined. This includes pressurized liquid hydrogen, cooled compressed hydrogen gas and two phase systems of liquid hydrogen with vapour in the headspace [14].

The storage density of LH2 is higher in insulated pressure vessels as it is slightly compressible: at 21 K, its density increases from 70 g/L at 1 atm to 87 g/L at 237 atm [15]. Cryocompressed vessels are accepted over traditional LH2 storage containers as they are better able to withstand heat since H2 within the vessel can be vented at a higher temperature. Venting stops when the tank reaches ambient temperature. When this occurs, the pressure within the tank is maintained so that the H2 density remains at 30% of the initial LH2 density. Cryocompressed hydrogen vessels have the ability to increase the volumetric energy density by eliminating unused space in container. This can be done because H2 becomes a supercritical fluid at pressures greater than 13 atm before the tank must be vented [16].

A sophisticated cryogenic capable pressure vessel was recently developed at the Lawrence Livermore National Laboratory (LLNL) in California, USA. It has advantages like: (1) high storage capacity (5-10 times greater than conventional LH2 tanks), (2) long thermal endurance and flexible refuelling. Also, this vessel eliminates the evaporative losses common in traditional liquid hydrogen tanks [17].

Cryogenic pressure vessel is shown in fig. 2.

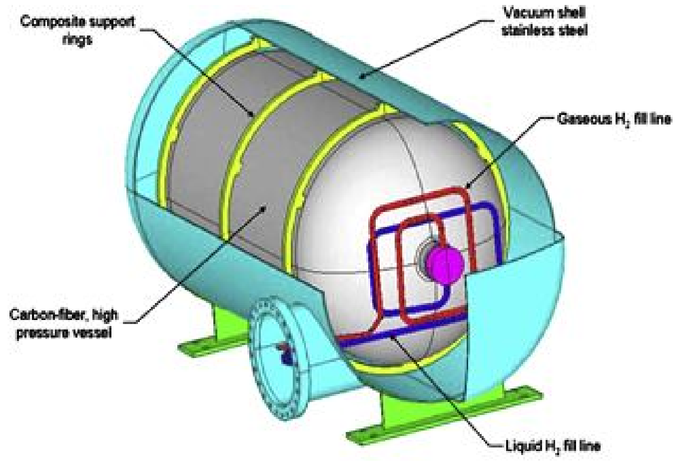

Fig. 2: Cryogenic pressure vessel [4].

III. METAL HYDRIDES

Metal hydrides have higher volumetric and gravimetric content than mechanical storage methods; they have been given a lot of attention recently. Hydrides have tendency to operate at the relatively low temperatures and pressures required in fuel cell vehicles, (the optimum ranges being 1-10 atm and 25-393 K for a polymer electrolytic membrane fuel cell [18].

3.1 Simple metal hydrides

Simple metal hydrides are the metal complexes which incorporates hydrogen into their crystal structure. They have been investigated since 1866 when Thomas Graham observed hydrogen absorption onto palladium. Metal hydrides are divided into two classes: binary hydrides, which contain only one metal in addition to hydrogen, and intermetallic hydrides, which contain two or more metals. Two-metal hydrides have the general formula AmBnHx where A and B are metals; they are then further subdivided into AB5 (CaCu5 structure type), AB2 (Laves phase), AB (CsCl structure type) or A2B (AlB2 structure type) where metal A has a strong affinity for hydrogen and forms a stable binary hydride while metal B does not interact with hydrogen [19].For a simple metal hydride to be practical for hydrogen storage, it must be produced by an exothermic reaction. The kinetics of the reaction must favour low energy hydride formation and H2 desorption [20].

3.2 Binary metal hydrides

There are many binary metal hydrides. However, most are not discussed here because they have such low gravimetric capacity that their use is unrealistic. The most promising binary metal hydride is alane (aluminium hydride, AlH3), which contains10.1wt% hydrogen with a density of

1.48 g/mL. There are at least seven phases of thermodynamically unstable alane that are metastable at room temperature and so do not decompose rapidly. Some level of instability is necessary for hydrogen storage materials because they must be able to release hydrogen without a large energy input [21]. The size of the metal hydride particles has a large effect on the thermodynamics and kinetics of hydrogen adsorption and desorption. It had previously been noted that reducing the particle size to the nano-scale lowers the desorption temperature and accelerates reaction kinetics. This occurs because (i) the surface-to-volume ratio increases, which increases surface reactions such as adsorption and desorption, and (ii) defects are formed, which cause the material to become more amorphous (less crystalline), which leads to more favourable thermodynamics [22].

Further research into the effect of size on hydrogen storage in alane lead to density functional theory (DFT) calculations performed on clusters containing 1-20 aluminium atoms. These studies found that, with the exception of very small clusters, desorption energy decreases steadily as cluster size increases until it reaches the low value of 0.19 eV/H2 forAl20H60. This suggests that bulk alane is unstable. In contrast, desorption energies of 0.4-0.6 eV/H2 are observed for Al8 H24 to Al16H48; these are desirable for hydrogen storage applications [23]. Further research into alane and other binary metal hydrides is on-going.

3.3 Intermetallic hydrides

Intermetallic hydrides are hydrides that contain at least two metals in addition to hydrogen. They were the first hydrides investigated because they can adsorb and desorb hydrogen under mild conditions. This is very important because many hydrogen storage materials do not work in the narrow range available in fuel cell vehicles (1-10 atm, 298-393 K. Although several intermetallic hydrides operate in the desired range, most have a gravimetric capacity too low and cost too high for vehicular applications [24]. As a result, LaNi5H6 is one of the few that is currently commercially available.

IV. NANOSTRUCTURES

4.1 Single walled carbon nanotubes (SWNT's)

Pores at the level near the molecular dimensions can adsorb quantities of gases having enhanced density of material which is adsorbed in those pores. The CNTs having diameters in the range of few nanometres can draw up the liquid by capillary. SWNTs were developed by co-evaporation of cobalt and graphite by the process of electric arc. In Transmission electron microscopy it is observed that amorphous carbon-coated fibers are having dimensions if several micrometre's. Those fibers are having bundle of 7-14 SWNTs which are individually ~12Å in diameters. And also the cobalt Nano particles embedded in amorphous carbon having diameter of 5-50nm and cobalt contents were observed up to ~20wt% by electron microprobe analysis [25] and remaining are the amorphous carbon and planer graphite observed in each sample. The adsorption of hydrogen on SWNTs and activated carbon was probed in a ultra-high vacuum chamber under a spectroscopy called temperature programmed desorption with the liquid-nitrogen-cooled cryostat and mass spectrometer [26].

The weight of sample is ~1mg which is packed in the packet of platinum foil packet having pinholes for the gas diffusion. Activated carbons are not having that much effect in storing hydrogen because only a small fraction of the pores in the typically wide pore-size distribution. Which are small enough to interact heavily with hydrogen molecules at normal temperature and moderate pressure. In recent years absorbents of carbon nano structures have been produced which includes graphite Nano fibers and carbon multi-walled and single walled Nanotubes [27]. The United States Department of energy (DOE) has set two targets, the first one is to acquire a ratio of hydrogen weight/tank weight which is 0.065, and the target is going to limit the weight of tank [28]. Second target requires a hydrogen volumetric density higher than 62kg/m3 in order to limit the volume of tank. Activated carbons are not having that much effect in storing hydrogen because only a small fraction of the pores in the typically wide pore-size distribution [29]. There are small enough to interact heavily with hydrogen molecules at normal temperature and moderate pressure. In recent years absorbents of carbon Nano structured have been produced which includes graphite Nano fibers and carbon multi-walled and single walled Nanotubes. The United States Department of energy (DOE) has set two targets, the first one is to acquire a ratio of hydrogen weight/tank weight which is 0.065, and the target is going to limit the weight of tank [30]. Second target requires a hydrogen volumetric density higher than 62kg/m^3 in order to limit the volume of tank.

There are two mechanism called physisorption and chemisorption used for hydrogen storage in carbon nanotubes. Chemisorption's use a catalyst for dissociation of molecular hydrogen allowing it to bond with some of the unsaturated carbon bonds along the tube. But during the early research in the potential means of hydrogen storage in CNTs focused on physisorption as the primary storage mechanism.

4.2 Spherical fullerenes

In 1991 it is predicted that hydrogen molecules can be trapped within a C60 cage. Although it is usually unfavourable for hydrogen to be stored in such a space, the high energy barrier is required to break the cage open stabilizing the hydrogen molecule inside. In the later studies it was found that one hydrogen molecule in the C60 can be synthesized experimentally with high yield by molecular surgery and by surrounding C60 cage in excitation of laser, with high pressure [31]. And it is difficult to put a laser amount into fullerene cages, so before the task has been undertaken many problems must be answered theoretically. This can include how hydrogen is put inside the cage; how it can be released and controlled, and also the properties required for storing the hydrogen [32].

Many scientists claim that only one hydrogen molecule will stay in C60 because of there are high formation energies for complexes containing larger amounts of hydrogen, But in some other studies it is found that 23-25 H2 molecule can be stored within a single C60 cage. There are also some disagreements about the mechanism in storage of hydrogen whether it chemisorbs to interior surface of the carbon cage or if they exist in the molecular form. The most densely populated C60 cage is having the capacity of containing 29H2 molecule, has a gravimetric capacity of 7.5 wt%, which is crossing the 2010 DOE target of 6 wt%. [33]. Some observations have been made like that when there are less than 10 H2 molecule inside the cage, H2 molecule exists in the molecular form and are in well-organized geometry.[34] when the number is increased beyond, it is observed that some hydrogen atoms form covalent bonds with carbon atoms in the fullerene. Rather than its uptake and release mechanism, putting H2 in a fullerene is still largely unfavourable. So many methods have been investigated for fullerene enhancement. One of those is charging a fullerene. This can be done by electrochemical doping, ion/electron impact, laser desorption or chemical doping. Chemical doping can be done through substitutional, endohedral or exohedral doping of the fullerene with metal atoms [35]. Charged fullerenes have a potential hydrogen storage capacity of 8 wt%. Fullerenes which have an endohedral metal atom did not shows a significant enhancement of hydrogen binding but systems having exohedral metal atoms are promising [19]. C60 containing 5 (left, a), 15 (middle, b) and 25 (right, c) H2 molecules showing the deformation of the fullerene and the formation of H3 and covalently bound hydrogen in fig. 3.

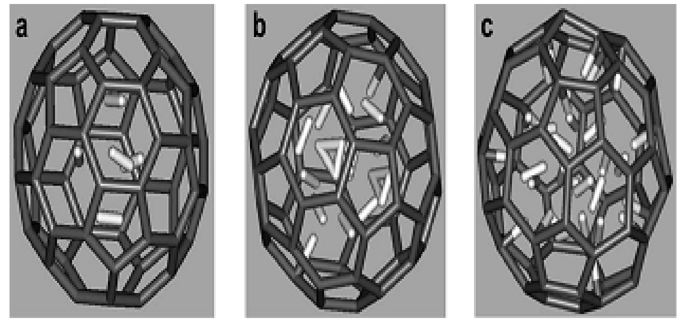

Fig. 3: C60 containing 5 (left, a), 15 (middle, b) and 25 (right, c) H2 molecules showing the deformation of the fullerene and the formation of H3 and covalently bound hydrogen [26].

V. CONCLUSION

Now a day, hydrogen storage has become very challenging and necessary task as fossil fuel resources are diminishing continuously and they causes pollution in large extent so. Clean, reliable, safe and efficient replacement is needed. The present paper discussed hydrogen gas storage methods from the last several decades. The general categories of hydrogen storage discussed in this paper include mechanical techniques, such as cooling and compressing the gas, chemical hydrides which contain hydrogen chemically bonded to non-hydrogen atoms and storage in single walled carbon nanotubes. These techniques are at various stages in their development and show a wide range of gravimetric and volumetric hydrogen storage capacities. There are many encouraging theoretical results, but an experimental system capable of meeting the targets set out by the U.S. Department of Energy has not yet been found. Therefore, extensive research in this field continues. Storage in high pressure tanks having hydrogen of low density is a challenging task. Some important points in hydrogen storage are:

(1) Storage of hydrogen in Cryogenic tanks is compact system which requires more energy than compressed system to store.
(2) Compressed hydrogen storage system is preferred for a static application where weight does not matter as tanks tends to be heavy.
(3) Metal hydrides such as MgH2, NaAlH4, LiAlH4 and TiFeH2 with varying degrees of efficiency can be used as storing medium for hydrogen, often reversibly. These materials have good energy density by volume, although energy density by weight is often worse than hydrocarbon fuels.
(4) Metal hydrides store hydrogen in solid form under moderate temperature and pressure that gives them safety advantage.

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