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# Potential of carbon nanotubes as hydrogen storage medium for vehicular applications

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## ABSTRACT

Increasing energy requirements of our present day society are generally met by fossil fuel hydrocarbon sources. These sources are depleting at a faster rate and their excessive use in automobiles is causing global environmental problems. There is need, therefore, to develop alternative energy sources which are cleaner, more efficient and environmental friendly. Hydrogen is one such energy source which has a potential to power zero-emission automobiles via a proton-exchange membrane fuel cell. However, the use of this energy source is hampered by lack of a viable hydrogen storage and release system. Carbon nanomaterials play a pivotal role in several energy related applications. Since their development during the last one decade, carbon nanotubes have been considered a promising hydrogen storage material for use in fuel cells. Consequently, a considerable amount of research has been directed in this direction. However, less than 1 wt% of hydrogen storage has been found to be practical. According to U.S. department of energy, the carbon material should be able to store 5.5% of its weight of hydrogen to make fuel cells practical in automobiles. The main objective of this review paper is to overview challenges of hydrogen storage technologies and potential of carbon nanotubes as hydrogen storage medium.In this review, recent developments in the preparation, purification, modification methods of CNT to enhance hydrogen storage as well as sorption–desorption kinetics and hydrogen uptake mechanisms in nanotubes are also discussed.

***Keywords: Carbonnanotubes, hydrogen storage, energy source, fuel cell, vehicular applications***

## I. INTRODUCTION

Energy is the integral part of human life since ancient time. Today's world relies heavily on the non-renewable fossil fuels such as coal, oil and natural gas, but their availability is declining. The conventional energy sources are not likely to meet the peak energy demand of growing world population which necessitates alternate energy sources. Moreover, their intensive use is leading to serious global environmental problems which are affecting the health of living beings on the earth. Therefore, there is need to shift from carbon based to carbon neutral technologies like solar, hydro, wind, biomass & biofuel based technologies etc. With accelerating demand of cleaner, sustainable and more efficient energy sources, research on hydrogen-based energy systems has attracted considerable interest in scientific community.

### 1.1. Hydrogen as alternative energy source

Hydrogen has been widely recognized as a promising energy carrier because of its abundance, environmental friendliness and high energy efficiency. Hydrogen is a potential non-carbon based energy resource, which could be generated from clean and green sources such as nuclear energy, natural gas and coal with carbon capture, biomass and renewable energy sources (wind, hydro, solar, geothermal). Currently, global hydrogen production is 48% from natural gas, 30% from oil, 18% from coal and 4% from water electrolysis [1]. Hydrogen is a fully renewable energy carrier and is harmless to environment because of its clean combustion. Energy density of hydrogen (38 KHW/Kg) is much higher than conventionally used gasoline (14 KHW/Kg). In addition, it has the highest energy content per weight unit (120 MJ/kg) of any known fuel [2]. Despite of several advantages, it exhibits certain major drawbacks in its utilization as fuel. The most important is that it is gaseous under ambient conditions and has a very low density (10 times lower than air). This results in severe storage difficulties. Secondly, due to high inflammability, adequate safety measures should be taken for $H_2$ handling in refilling station and automobiles. In the case of on-board storage of hydrogen for vehicular applications, automobile manufacturers require lightweight, compact, safe, and cost-effective storage with the ability to achieve a driving range of at least 300 miles. Typically, 6 kg of hydrogen is able to allow a light-duty vehicle to run for 500 km [3]. Unfortunately, the main problem associated with hydrogen technology is availability of safe and practically possible hydrogen storage device which could easily load and unload hydrogen to provide supply to fuel cell. For practicality, the 2020 targets by the US DOE for system gravimetric and volumetric densities are set to 5.5 wt.% and 40 g of $H_2$/L at an operating temperature of 40 to 60 $^{o}$ C under a maximum delivery pressure of 12 bar [4]. During the last fifteen years, significant efforts have been made for finding a solution to the hydrogen storage problem. Although various hydrogen storage technologies are presently available, no approach satisfies all of the efficiency, size, weight, cost and safety requirements for transportation use.

### 1.2. Hydrogen Storage Technologies

Presently, hydrogen may be stored by following six different techniques [5-7] :

(i) High-pressure gas cylinders (up to 800 bar)

(ii) Liquid hydrogen in cryogenic tanks (at 21K)

(iii) Physiosorbed hydrogen on materials with a large specific surface area (at temperatures < 100 K)

(iv) Chemisorbed on interstitial sites in host metals at ambient temperature and pressure

(v) Chemically bonded in covalent and ionic compounds

(vi) Through oxidation of reactive metals such as. Li, Na, Mg, Al, Zn with water.

A comprehensive list of hydrogen storage methods along with their volumetric and gravimetric densities is given in Table 1 [5]. Compressed gas storage requires the container to be strong and bulky whereas liquid storage of hydrogen requires a cryogenic temperature (20 K). In metal hydride storage, their low mass, energy inefficiency, stable thermodynamics and slow kinetics hinder their practical application Although the solid state hydrides have high volumetric energy density but low energy density by weight i.e. 1.5 wt% which is only

marginal particularly for vehicular applications. Nanoadsorbents with high surface areas are being actively developed as another option to overcome kinetics and thermodynamics drawbacks of metal hydrides and provide solutions for producing, transmitting and storing energy. Among these adsorbents, carbon nano materials are inherently safe, cleaner and more energy efficient than chemical or metal hydrides and compressed gas storage. Consequently, the hydrogen storage capacities of CNTs have been intensively investigated.

**Table 1: Hydrogen storage methods[5]**

| Storage method | $\rho_m$ (mass %) | $\rho_v$ [$kgH_2m^{-3}$] | T ($^oC$) | P (bar) | Phenomena and remarks |
|---|---|---|---|---|---|
| High pressure gas cylinders | 13 | < 40 | RT | 800 | Compressed gas (molecular $H_2$) in light weight composite cylinders (tensile strength of the material is 2000 MPa) |
| Liquid hydrogen in Cryogenic tanks | size dependent | 70.8 | -252 | 1 | Liquid hydrogen (molecular $H_2$), continuous cryogenic tanks loss of a few % per day of hydrogen at RT |
| Adsorbed hydrogen | ≈ 2 | 20 | -80 | 100 | Physisorption (molecular $H_2$) on materials e.g. carbon with a very large specific surface area, fully reversible |
| Absorbed on interstitial sites in a host metal | ≈ 2 | 150 | RT | 1 | Hydrogen (atomic H) intercalation in host metallic hydrides working at RT are fully reversible |
| Complex compounds | < 18 | 150 | >100 | 1 | Complex compounds ($[AlH_4]^-$ or $[BH_4]^-$), desorption at elevated temperature, adsorption at high pressures |
| Metals and complexes together with water | < 40 | >150 | RT | 1 | Chemical oxidation of metals with water and liberation of hydrogen, not directly reversible |

### 1.3. Carbon nanotubes

Carbon nanotubes were discovered in 1991 by Ijima [8]. Carbon nanotubes are tubular forms of carbon that can be envisaged as graphitic sheets rolled into cylindrical form. These nanotubes have diameters of few nanometers and their lengths are up to several micrometers. These are produced by Arc-discharge, Laser-abalation and CVD methods. There are two variations of CNT's i.e. single walled nanotubes (SWCNT) and multiwalled (MWNT). SWCNT are composed of individual graphene sheets rolled into seamless hollow cylinder with diameter ranging from 1-20 nm. MWNT by constrast, comprises of several concentric graphene cylinders. Due to unique

structural, mechanical and electrical properties of single wall carbon nanotubes (SWNTs), they have been proposed as potential hydrogen medium for automotive applications. CNTs have high surface area and highly developed microporous structure and can provide large external surface and internal hollow cavity for storage of hydrogen.

## II. HYDROGEN STORAGE IN CARBON NANOTUBES –REVIEW OF LITERATURE

In 1997, Dillon et. al. [9] reported the first experimental evidence for hydrogen storage in single wall carbon nanotubes. The high hydrogen uptake of these materials suggested that carbon nanotubes could be effective as hydrogen storage material for vehicular applications. Thereafter, several groups of researchers initiated work towards adsorption of hydrogen by carbon nanotubes. Dillon et. al. [9] used unpurified soot containing 0.1-0.2 wt % of single wall carbon nanotubes and obtained 0.01 wt% of $H_2$ storage at 133 K temperature and 300 Torr pressure. From these results, it was extrapolated that a sample of highly pure SWNTs could reach hydrogen adsorption capacity of 5 to 10 wt %. Following up on these findings, many research groups studied the influence of purity of CNTs on hydrogen adsorption. Tarasov et. al. [10] produced SWNTs by arc evaporation of graphite electrodes with the use of two different catalysts, 3Co/Ni and $YNi_2$ and then purified these samples upto purity level of 75%. These workers observed the reversible storage capacity of 2.4 wt. % $H_2$ on purified samples at cryogenic temperatures below −150 °C and at a pressure of 25 bar $H_2$. Liu et al. [11] measured the hydrogen storage capacity of single walled carbon nanotubes synthesized by semi-continuous hydrogen arc discharge method at ambient temperature and under 10 MPa pressure and found that SWNTs could adsorb upto 4.2 wt% $H_2$ after purification with aqueous HCl. Ioannatos and Verykios [12] investigated the adsorption of hydrogen on single-walled and multi-walled carbon nanotubes (CNTs) at 77 K and 298 K, in the pressure range of 0–1000 Torr. It was seen that the hydrogen uptake capacity of CNTs was increased with the increasing purity of the materials. Similarly, According to Tekkaya and Karatepe [13] purification by acid treatment is the most important factor affecting the hydrogen uptake. The maximum capacity was obtained with purified SWCNTs produced on Fe catalyst whereas purified SWCNTs grown on Fe–Co catalyst had the minimum hydrogen uptake. Gundiah et al [14 ] also reported that maximum hydrogen storage capacity of 3.7 wt% on densely aligned bundles of MWCNTs treated with acid. In the same way, experimental study by Ritschel et al. [15] showed that the purified SWNTs had a reversible hydrogen storage capacity of 0.63 wt% at room temperature and 45 bar which was higher than that of MWNTs and CNFs. Darkrim et al [16] also observed that high degree of purity ensures high hydrogen adsorption in carbon nanotubes. All these were of view that both the material synthesis and purification will have to be optimized in order to improve the hydrogen uptake.

On the other hand, Luxembourg et al [17] prepared samples of single wall carbon nanotubes (SWNTs) with different diameter distributions using a solar reactor. For purification, these samples were treated with HCl and were oxidized thermally in air and with $H_2O_2$ and $HNO_3$. The highest adsorption capacity (0.7 wt%) was found on raw soot. Further, a correlation between textural properties and hydrogen storage capacities was also discussed. HCl protocol clearly increased the BET surface area ($S_{BET}$) and the microporous volume

whereas $HNO_3$ protocol decreases them. Muthu et al [18] reported the hydrogen storage performance of multi-walled carbon nanotubes /hexagonal boron nitride nanocomposites (MWCNT/h-BN) synthesized by ultrasonication method. Hydrogen storage was enhanced from 0.15 wt% to 2.3 wt% with acid treatment. Moreover, the calculated binding energy (0.42 eV) of stored hydrogen of acid treated-MWCNT with 5 wt% of h-BN nanocomposite lies in the recommended range of binding energy (0.2–0.6 eV) for fuel cell applications. Also, TG study showed that 100% desorption is achieved at the temperature range of 120–410 °C. Chen et al. [19] investigated the hydrogen adsorption on alkali- doped carbon nanotubes and found that potassium-doped carbon nanotubes demonstrated hydrogen uptake of 14 wt% at room temperature while lithium-doped CNTs showed 20 wt% hydrogen storage at elevated temperatures (473 K- 673 K). However, Yang et al [20] reexamined their results and reported that this high $H_2$ weight uptake is mainly attributed to the moisture. Moisture drastically increased the weight gain by reactions with the alkali species on carbon. However, in dry hydrogen atmosphere, alkali-doped carbon nanotubes adsorbed nearly 2 wt. % hydrogen. Ding et al [21] also observed that high $H_2$ weight uptake of alkali metal doped carbon nanotubes uptake was due to presence of water which causes error in evaluation of the storage capacity of doped nanotubes.

Some research groups conducted the density functional theory calculations to investigate hydrogen storage applications of carbon nanotubes doped with various elements and to predict the binding energy of hydrogen with CNT system. Tian and Dong [22] predicted that Yttrium-dispersed capped-carbon nanotubes (CCNY) can serve as a high-capacity hydrogen storage material. The interaction of $H_2$ molecules with the CCNT can be significantly enhanced upon decoration by Y atoms. The $H_2$ molecules binding to the Y-CCNT system with six Y atoms has an average binding energy of −0.48 eV which is suitable for reversible hydrogen storage at ambient conditions. Further, the gravimetric density of the cap was found to be 7.51 wt% which exceeded the 6.5 wt % target of the U.S. Department of Energy. Molani et al [23] studied the adsorption of hydrogen atom on Ca-decorated (8, 0) $C_3$NNT by spin-polarized DFT calculations. Interaction of Ca atom with the nanotubes and $H_2$ molecules were explained by Dewar coordination and Kubas interaction, respectively. It was found that the Ca-decorated on defective nanotube can adsorb up to eight $H_2$ molecules with the average binding energy of 0.11 eV/$H_2$. Therefore, Ca-decorated $C_3$NNT has been proposed as a suitable candidate for hydrogen storage. Similarly, Seenithurai et al [24] investigated that hydrogen storage capacity of Al-decorated SWCNT reaches to 6.15 wt%, making it a good hydrogen storage medium for onboard automobile applications. Each Al atom in (8,0) CNT-8Al adsorbs four $H_2$ molecules and the average adsorption binding energy of $H_2$ in (8,0) CNT-8(Al+4$H_2$), i.e. 0.214 eV/$H_2$, lies between 0.20 and 0.60 eV/$H_2$ which is required for adsorbing and desorbing $H_2$ molecules at near ambient conditions. Li et al [25] found that Ni or Fe embedment enhanced the SWNT interactions significantly for hydrogen due to increase in adsorption energies. Thus Ni- and Fe-embedded capped (5, 5) single-walled carbon nanotubes (SWNTs) can benefit the hydrogen storage applications.

Furthermore, $H_2$ adsorption uptake was measured using the different measurement methods and under different experimental conditions of temperature and pressure. Lan and Mukysan [26] developed an accurate gravimetric apparatus based on a contactless magnetic suspension microbalance to measure the hydrogen storage capacity

for a variety of carbon nanotubes (CNTs) at room temperature and hydrogen pressures up to 11.5 MPa. The results showed that regardless of their synthesis methods, purities, and nanostructures all investigated CNT products possess relatively low hydrogen storage capacities ( < 0.2 wt %). Ströbel et al [27] measured the hydrogen adsorption from the gas phase by isothermal gravimetric analysis, using a microbalance at hydrogen pressures up to 125 bar at 23°C. In this work, the hydrogen adsorption reached values of approximately 1.5 wt. % at ambient temperature and 125 bar. Anson et al [28] studied hydrogen adsorption on as-grown and heat-treated single walled carbon nanotubes (SWNTs) by a volumetric procedure using a Quantachrome Autosorb-1 equipment. They found that amounts of hydrogen adsorbed at atmospheric pressure reach approximately 0.01 wt.% at 298 K and 1 wt.% at 77 K. Barghi et al [29] utilised a powerful measurement technique based on a magnetic suspension balance coupled with a residual gas analyzer and found that measured sorption capacity was less than 0.2 wt %. Li et al [30] introduced tangent mass method to measure and calculate hydrogen storage capacity of CNT using high pressure microbalance. This method can calibrate the effect of buoyancy.

Zubizarreta et al [31] evaluated $H_2$ storage capacities by both volumetric and gravimetric methods at different temperatures and pressures. The differences between two methods at various operating conditions were related to the textural properties (i.e. surface area, pore size distribution, etc.) of the carbon-based adsorbents. The morphological characteristics had no influence on gravimetric storage capacity. The results also showed that temperature has a greater influence on the storage capacity of carbons than pressure. Furthermore, hydrogen storage capacity seems to be proportional to surface area and pore volume. Panella et al [32] also obtained a linear relation between hydrogen uptake and specific surface area (SSA) for all carbonaceous samples independent of the nature of the carbon material. The carbon material with a SSA of 2560 $m^2/g$ showed the highest storage capacity of 4.5 wt% at 77 K. Zhu et al [33] found that well aligned CNTs exhibited higher hydrogen capacity of (3 wt%) compared with randomly ordered CNTs. This was attirbuted to narrow pore distribution of well-aligned carbon nanotube bundles which was favorable for hydrogen uptake. Darkrim et al [16] were also of view that optimizing the nanotube structure by controlling tube diameters and lengths may enable a maximal hydrogen adsorption. Systematic experimental investigation by Gogotsi et al [34] on a large number of CDCs with controlled pore size distributions and specific surface area (SSA) showed that the effect of pore size was stronger than the effect of surface chemistry on the hydrogen uptake. Pores larger than ~1.5 nm contribute little to hydrogen storage while pores of 0.6–0.7 nm in diameter provided the largest $H_2$ uptake per unit SSA at both ambient as well as elevated pressures and liquid nitrogen temperatures. Jordá-Beneyto [35] carried out hydrogen adsorption measurements at different temperatures (298 K and 77 K) and high pressure of carbon materials. At 298 K, the hydrogen adsorption capacity depends on both the micropore volume and the micropore size distribution while At 77 K, hydrogen adsorption depends on the surface area and the total micropore volume of the activated carbon. A. Züttel [36] observed the effect of temperature on amount of adsorbed hydrogen. It was seen that the nanotube samples adsorbed hydrogen up to 5.5 mass% at cryogenic temperatures (77 K). However, at room temperatures this value drops to ≈ 0.6 mass%. The electrochemical experiments on the carbon samples showed a maximum discharge capacity of 2.0 mass% at room temperature

(298 K). Xu et al [37] also investigated the hydrogen adsorption capacity of various carbon materials at 303 K and 77 K. It was seen that hydrogen adsorption was increased significantly by lowering adsorption temperature. Further, it was directly proportional to their specific surface are and micropore volume. The results by Thomas [38] also indicated that the amounts of hydrogen adsorbed in porous materials at ambient temperature was much lower (< 0.5 wt %) while at 77K, upto 5 wt% of hydrogen can be stored.

Thermodynamics and kinetics studies were carried out by different research groups in order to understand the uptake mechanisms. Rather et al [39] compared the hydrogen adsorption of commercial, milled, and $MgH_2$composite. Hydrogen adsorption capacity of commercial $MgH_2$, milled $MgH_2$, and $MgH_2$/CNT composite are found to be 0.04, 0.057, and 0.059 g ($H_2$)/g ($MgH_2$) at 673 K and hydrogen pressure of 4.6 MPa. Further, addition of 5 wt% of CNTs to $MgH_2$ enhanced the hydrogen adsorption capacity and improved its kinetics. Rather also [40] studied hydrogen uptake of cobalt and copper oxide-multiwalled carbon nanotube composites prepared by in situ reduction method. Hydrogen uptake enhancement of composites was 10-fold compared to pristine MWCNTs. Enhancement of hydrogen storage of both composites was attributed to the spillover mechanism due to decoration of Co and Cu-oxide nanoparticles on the outer surface of MWCNTs. Similarly, Pyle et al [41] reported the experimental and theoretical studies on hydrogen storage by transition metal doped carbon nanostructures via the spillover i.e. by dissociating molecular hydrogen and allowing adsorption via chemical means. The most promising materials are found to be high surface area hexagonal system carbons for which the $\pi$-conjugation is broken by well dispersed oxygen functional groups or lattice dopants. Callejas et al [42] measured adsorption isotherms at 77 K and 300 Torr on raw and modified SWNTs produced by arc discharge using Ni/Y as catalyst at different percentages. The samples after hydrogen reduction at 350 $^{o}$C showed hydrogen adsorption 40% higher than that of unreduced materials. This suggested the hydrogen dissociation by the reduced metal nanoparticles and subsequent spillover to the SWNTs. Ruse et al [43] explored the hydrogen storage kinetics of Pd-Mg composites upon addition of different carbonaceous spillover agents (activated carbon and a wide spectrum of carbon nanotube types). They found that the hydrogen (loading or release) kinetics is strongly dependent on the nanocarbon morphology and configuration (e.g., length, diameter and Pd distribution). The fastest kinetics was obtained for our Pd-decorated carbon nanotubes having the largest diameter. Hwang and Chiang [44] investigated the hydrogen storage performance of ball-milled $MgH_2$ with 5 wt% $ZrO_2$ + 5 wt% single-walled carbon nanotubes as additives. It was seen that the hydrogen sorption kinetics of magnesium was markedly improved by these co-additives. The sample absorbed 4.00 wt% $H_2$ within 700 s under ambient temperature conditions (298 K). Zacharia [45] investigated the hydrogen storage capacities of palladium- and vanadium-doped carbon nanotubes (CNTs) at room temperature using the Sieverts apparatus. The storage capacity of Pd- and V-doped CNTs at 2 MPa were found to be 0.66 and 0.69 wt%, which were nearly 30% more than that of the pristine-CNTs. The doped-CNTs exhibited faster initial hydrogen adsorption kinetics and higher storage capacity compared with the pristine samples. All these studies revealed that metal particles enhanced the storage capacity via the spill-over mechanism.

The natures of interaction between hydrogen and carbon nanotubes were studied using experimental and theoretical modeling. It is proposed that hydrogen may be stored through physical or chemical adsorption. Physiosorption involves weak vanderwaal forces of interaction and occurs via condensation of hydrogen in the cavity of the nanotube. Early studies have focused on physical sorption as the primary storage mechanism, but it could not explain the high hydrogen uptake capacities of CNT at ambient temperatures. However, hydrogen can be stored reversibly through physiosorption due to low binding energies and the process is associated with fast kinetics [31,46-48 ] . In constrast, chemisorption is characterized by dissociation of hydrogen molecules and formation of stable C-H bonds. Though consideration of chemisorption mechanism can account for high wt% of hydrogen under ambient conditions, it requires high temperatures (above 450 $^{o}$ C) to release $H_2$ for technical applications by breaking of covalent C-H bond [31, 46-48 ]. Optimal interaction energies for significant but reversible storage under ambient temperature and pressure should be around 7 Kcal/mol i.e. inbetween those of physiosorption and chemisorptions [49 ]. Dillon et al [9] and Xu et al [37] suggested that physical adsorption of hydrogen mainly occurred within the inner hollow cavities of SWNTs due to condensation inside narrow pores. Experimental studies by Panella et al [32] showed physisorption interactions between hydrogen molecules and the carbon nanostructure as indicated by the fast kinetics and complete reversibility of the process. The strong temperature dependence of hydrogen adsorption on porous materials also indicated the physiosorption as hydrogen uptake mechanism [38]. Zuttel [50] also proposed that hydrogen get adsorbed by condensation in the cavity of the nanotube or by formation of monolayer at the surface of the tube. Further, the hydrogen storage density due to condensed hydrogen was found to be 1.5 mass% while surface adsorption of a monolayer of hydrogen led to a maximum storage capacity of 3.3 mass%. While in another studies [36], they found that hydrogen tends to bind covalently to carbon at elevated temperatures ( > 573). On the other hand, Barghi et al [29] observed that the interactions of hydrogen with multi-walled CNTs are due to a combination of weak sorption (physisorption) and strong sorption (chemisorption). The maximum amount of hydrogen adsorbed by physisorption and chemisorption were found to be 0.13 wt % and 0.058 wt %, respectively. Thermal desorption studies by Tarasov et al [10] revealed the presence of weakly bonded physisorbed hydrogen (90%) and chemically bonded hydrogen (10%). Nam et al [51] also observed both the physisorption and chemisorption sites on SWCNTs using thermal desorption spectroscopy (TDS). However, Ioannatos and Verykios [12] suggested through TPD studies that a single type of adsorption site exists on the solid surface which binds the adsorbent and adsorbate through relatively strong adsorption bonds. Muthu [18] and Molani et al [23] indicated the chemical nature of interactions between hydrogen and doped adsorbent via spillover mechanism. Zacharia [45] suggested that nearly 70–85% of the spilled hydrogen occupies the physisorption binding sites such as external-walls or groove-sites of CNTs. Hydrogen was released at temperatures above 450 °C as a result of breaking of the covalent C–H bonds. Nikitin et al [48, 52] were also of view that hydrogen is chemisorbed in single-walled carbon nanotubes through the formation of reversible C−H bonds. A method of chemisorption was also proposed by researchers at Penn State [53], in which clusters of metal nanoparticles are chemically affixed to the surface of carbon nanotubes and act as doorways into the surface of the tubes. Some of

the hydrogen is absorbed by the metal, converting them to metal hydrides, while the bulk is absorbed into the CNT where it adheres to the walls.

## III.CONCLUSIONS

The above perusal of literature have shown that carbon nanotubes could serve as ideal storage materials for hydrogen for use in fuel cell powered vehicles due to high surface area, tunable porosity, reasonable tube diameter and cavity volume. Although published amounts of hydrogen uptake appear to vary between 0.1 to 67 wt.% of carbon, it is generally accepted that the available hydrogen for practical use is still less than 1%. Many of these results have been subject of considerable debate as these are not confirmed by other research groups. Furthermore, Carbon nanotubes showed high $H_2$ uptake capacities only under cryogenic conditions, making them unsuitable for automotive applications. At room temperature and around moderate pressures, the amount of hydrogen adsorbed by carbon nanostructures is low due to low interaction energies between adsorbed molecules and nanotube walls. However, alkali and transition metal doped –CNTs exhibited high hydrogen storage capacities even at ambient conditions due to large adsorption energies. In order to improve the hydrogen storage performance of CNTs, emphasis should be given on large-scale production of purified carbon nanotubes with ultrahigh surface areas, and controlled microporous structure. In addition, chemical structure of carbon nanomaterials can be modified through incorporation of metals and surface functionalities. Attempts should be made for development of carbon materials which could store sufficient hydrogen at ambient conditions in terms of gravimetric and volumetric densities and at the same time possess suitable thermodynamic and kinetic properties for quick uptake and release of hydrogen. If above challenges are met, the carbon materials will revolutionise the technological world through commercialization of hydrogen economy.

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