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REVIEW

Carbon nanotubes for production and storage of hydrogen: challenges and development

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Abstract

Carbon nanotubes have garnered significant interest due to their promising applications and facile synthesis. This study highlights the applications of CNTs in the field of hydrogen production and storage. Hydrogen energy attracted researchers because of its clean, renewable and sustainable energy with low impact on the environment around the globe. It is expected hydrogen energy systems replace the prevailed fossil fuels in the coming years. Hydrogen systems exhibit many disadvantages such as production costs and storage aspects. CNTs have the greater capability as support for the manufacture of effectual contrasting catalysts in hydrogen production systems. The main focus of this article is their different manufacturing methods along with their models and the purification techniques to obtain the best quality CNT's and then use them in different applications. Some of them are best suited to provide the quantity while on the other hand, some can provide the better quality of CNT's. Moreover, by using different techniques the different classifications of CNT's like SWCNT and MWCNTs can be obtained according to our needs and preferences. This paper reviewed the methodologies used in the production and storage of hydrogen. Our concern is basically to review the dares in production and storage of hydrogen compare their results, to study recent developments in modifications of CNTs to increase production and storage of hydrogen.

Keywords Hydrogen · Carbon nanotubes (CNTs) · Adsorption · Storage · Production

Introduction

Nanotechnology is one of the emerging fields in the recent era. It allows us to tackle things at a vast level. It tells us the anatomy of the objects around us is different, at macro- and nanolevels (Dresselhaus et al. 2000a, b). Nanotechnology allows us to handle things beyond our thoughts. If we can manipulate and control things at such a small level, this can be beneficial for many fields involving military, medical sciences, cosmetics, and engineering, etc. These nanomaterials can be created by objects with a size range of less than 100 nm (Dresselhaus et al. 1998). Nanotubes are one of the best-suited tools in this regard. Hydrogen has taken great curiosity as it is the cleanest, sustainable energy transporter with less effect on the environment (Tahir et al. 2020a, b). In the future, hydrogen is considered as alternative fuels that will replace fossil fuels as these give rise to global warming and pollute the environment (Tahir et al. 2019a, b, 2020a, b; Dresselhaus et al. 1995, 1998). One of the major applications of hydrogen is its utilization as fuel in power transfer vehicles. Although hydrogen has many advantages, it also shows major drawbacks such as storage characteristics and production costs. CNTs and CNFs have very fascinating properties and are considered versatile materials in nanoscience (Dresselhaus et al. 2000a, b). Being adsorbent material, CNTs are used in hydrogen storage systems and as support of catalysts in the production of hydrogen. In this article, synthesis methods of CNTs, their demand in both production and storage of hydrogen, and at last to talk over and contrast the different conclusions published.

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Carbon nanotubes

Carbon nanotubes were discovered by Dresselhaus et al. (1995). These are long thin macromolecules shaped as cylinders of carbon. So, carbon with the atomic number 6 is the main crucial element of these cylindrical tubes. These tubes have gained abundant interest (Dresselhaus et al. 2000a, b, 2005) due to their unique size, shape, and extraordinary properties that they possess (Dresselhaus et al. 2000a, b, 2005). Recent studies found a hexagonal lattice of carbon rolled into a cylinder deep within the structure. The properties of tubes are quite intriguing to the recent researches for science and large investments have been made for the sake of nanoscience. But the thing is, due to their extremely small size, their physical properties are still disputed (Popov 2004). These nanotubes can be of different types depending on their diverse properties regarding their composition, like broad thermal and electronic properties. These tubes have a single sheet of atoms rolled into a cylinder but they can have multiple layers in-cylinder within the cylinders to make things more interesting. Not every object or particle falls in this category. Some conditions should be fulfilled. In nanoworld, we deal with some 0D, 1D, 2D as well as 3D particles and materials (Dresselhaus et al. 1998; Peigney et al. 2001). These particles have at least one dimension less than 100 nm and that's the main condition. So, the nanotubes are the best in this regard. Their length and width are about 1 or 2 nm. They are just like molecules with distinct chemical and physical properties. These properties are manipulated by their distinct manufacturing methods. Due to unique properties, they exhibit many promising applications (Baughman et al. 2002). These tubes have best-suited electrical and thermal conductivities with versatile mechanical abilities. They have a large L to D ratio so they can be considered as a good electron emitter. The main element of these nanotubes is carbon, so due to pure carbon polymers they can be changed during their preparation. The carbon nanotubes have attracted researchers because they can replace many types of building blocks of different devices and can modify and increase their performance such as nanoelectronic sensors (Lu 1997). In 2018, there has been a research paper published focusing on the utilization of CNT's in the production of nanoelectronic sensors. This interest developed due to their better sensitivity and accuracy (Peigney et al. 2001). Before this, there were only experimental studies of sensors but in 2018, they provide analytical models that can be comparable to the experimental investigations (Dresselhaus et al. 2005; Mitsuyama et al. 2014). It was FET based on the CNT's that provide the transport parameters of CNT-based gas sensors. In this proposed model, the target analytic NH_3 is absorbed by the CNTs surface during a reaction to affect the conductance of the CNTs (Dai 2002; O'connell 2018). So, I–V characteristics of CNT-based FET sensors can be used to predict the effect of NH_3 gas on CNT surface not only this but carboxylic functionalized multi-walled carbons impact on trypsin's nanotube dispersion along with their deposition from catalytic C–H–O has been studied as well (Serp et al. 2003). Balachandra et al. (2018) have studied that due to the high sensitivity and better response of CNTs towards external stimulus (Balachandra, Darsanasiri, and Soroushian), they exhibit many promising applications including soft robotics, artificial muscles, and other electronic devices. The CNTs that can be used as gene carriers can interact with many biological structures to achieve potential applications in biomedical fields (Shiraishi and Ata 2001).

In 2015, as we go further back in the years, we can see plenty of work related to CNTs in biomedical fields. In this year, a work came into the observation that suggests the CNTs can be used to produce bone-like structures but with better firmness and durability (Ebbesen 1996). This can help in bone regeneration and enhance their strength as well. This work further suggests the interaction of mechanical and biological functions of bones. In the same year, it was explained that how CNTs can replace the silicon transistors to fulfill Moore's law (Bottini et al. 2006; Chen et al. 2011). Moore's law states that with every passing couple of years the number of transistors would be doubled. With the help of carbon nanotubes, it is possible to minimize the size of the transistors as we go further the width or size would be up to only a few atoms. So, in this way smaller sized can support Moore's law with promising future progress (Dresselhaus et al. 2005; Salvetat et al. 1999).

In 2014, the drug design its development and cancer therapy, etc. has been reported. It uses the CNTs as the modified drug delivery agents to the tumors that show the (MDR) or multidrug resistance (Bandaru et al. 2005). MDR can lead a drug or therapy useless against a tumor that factor can be overcome by shortened CNTs that are proved to be the real-time reversal of tumor (Long and Yang 2001; Ishigami et al. 2000). Along with it, there have been many other works reported related to CNTs applications and their importance. In 2013, cell imaging using CNTs have been researched (Meyyappan 2004). SWCNTs have been used in cell imaging and as a diagnostic agent for many biological issues (Hiura et al. 1993). So, they can be used for targeting and imaging cells as well. Studies have mainly focused on biomedical problems and their solutions using CNTs. Due to their small size and structure, CNTs use in different biomedical areas has been increased (Planeix et al. 1994; Salvetat et al. 1999; Chen et al. 2011). Considering this, there has been reported about the CNTs exposure to the human. In 2012, it was analyzed and determined by Raman spectroscopy for

the experiment, mice have been used were made to inhale the CNTs that were SWCNTs (Su et al. 2000). After that their effect on the circulatory system has been studied by using Raman spectroscopy (Tsai et al. 2009). In the 1950s, Michael Kasha, a chemist proposed a rule in which he said that when a molecule is exposed to light it will emit energy from its lowest energy excited state (Chen et al. 1999a, b). In 2011, the breaking of the Kasha rule has been reported, by CNTs. In which emission occurs from a higher excited state that is opposite of Kasha's rule (Saito and Uemura 2000). There are many more researches where carbon nanotube-related technology has been developing and making its progress as we go further back in history and considering the recent era its importance has been revealed (White and Todorov 1998) as highlighted in Fig. 1.

CNTs have attained great interest in the recent few years due to its importance in the world of science (Wang et al. 2011). Both types of CNTs exhibit unique properties depending on their preparation techniques.

SWCNT

The thickness of these types of tubes is only about a single atom thick while it is a hollow cylinder of graphene. While most of the credit for their discovery is given to Iijima due to his publications in 1991 but its roots are very deeper than this. Due to the numerous properties, they have gained a lot of interest that is still growing by the time, in the field of nanoscience. Han and coworkers have introduced first SWCNT in 1998 (Han et al. 2006). Recent studies have revealed that CNTs presents significant properties at macro- and micro-levels, So the axis with which we measure the length and width of nanotubes can vary its mechanical properties enormously (Sano et al. 2012). By that, we can vary their Young's Moduli ranging up to 1TPa and their strength about the range of 100 GPa. The SWCNT is oriented in the lattice structure upon which their electrical properties depend (Lee and Parpura 2009). Fan et al. have reported that the structure of SWCNTs is like hexagonal benzene rings of carbon (Fan and Advani 2007). The graphene sheets are arranged as the honeycomb lattice. The usual circumference of these tubes is only 10 atoms. The distance-to-diameter quotient is around 1000. We can consider them as one-dimensional (1D) structures (Neubauer et al. 2010).

MWCNT

Multi-walled carbon nanotubes (MWCNTs) are nothing but the quantity of single-walled nested into each other. The tubes can be different in number in different multi-walled CNTs (Li et al. 2012; Neubauer et al. 2010). The least of them could be three tubes into each other. Most of them could be 20 nested insides. That's the reason for their huge diameter compared to single-walled (Roch et al. 2007). If the most centered tube has a diameter of 2 nm, then it might be possible for the outer one to have a diameter of 50 nm or above. Depending on the structure, they can exhibit various properties of mechanical, electrical, and thermal conduction (Roch et al. 2007; Thostenson et al. 2009). As there are several tubes nested inside of each other so that there are much more possibilities of flaws and defects compared

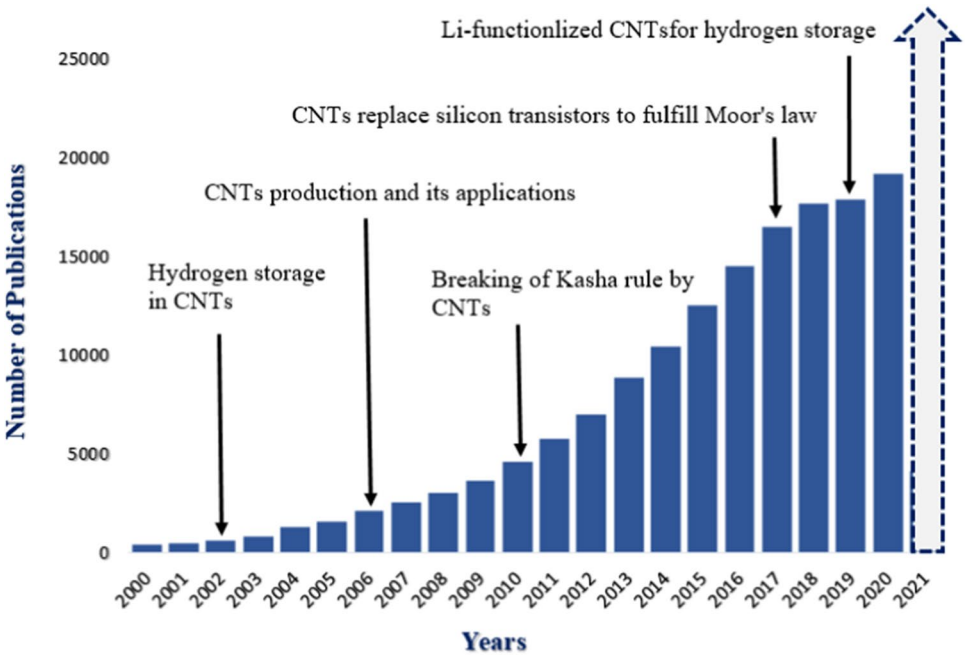

Fig. 1 Year-wise advancement in the utilization of CNTs in hydrogen production

to SWCNTs (Coleman et al. 1999). It can affect their performances as well. These properties of MWCNTs and their flaws can be reduced by using different physical or chemical methods (Al-Khedher et al. 2007; Baughman et al. 2002). The separation between the tubes is about 0.34–0.39 nm. In contrast to single-walled, multi-walled CNTs are easy to deal with. They can be purified easily and can be produced in a high amount (Al-Khedher et al. 2007; Malik et al. 2011). These have attractive applications due to unique properties including cost-effectiveness. Niranjana et al. have reported that these tubes are bound to have an outer diameter of less than 15 nm (Niranjana et al. 2009). If they cross their restriction, then they are no more tubes but carbon nanofibers instead (Niranjana et al. 2009; Sacha et al. 2009). Multi-walled CNTs have two types of models: One model is known as Russian doll model when one nanotube with a relatively greater diameter contains another nanotube nested inside of it. They are in the form of concentric cylinders (Al-Khedher et al. 2006). About the size, we can say that a (0 and 8) single-walled CNT could be placed inside of a (0 and 17) single-walled CNT (Qian et al. 2002). When one sheet of graphene is being folded throughout itself like a roll it's called a parchment model. The spacing between the layers is about the space inside the graphene sheets in graphite, which is about 3.4 Å (Vigolo et al. 2000). Riggs and coworkers have found that carbon nanotubes have gained significant attention to develop future most efficient devices to get the most out of them (Riggs et al. 2000). They have different types and each has unique properties so that we can choose from according to requirement (Li et al. 1999). Table 1 presents some of these properties that have been discussed along with how they are different for each type.

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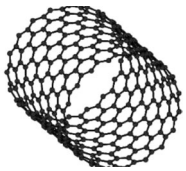
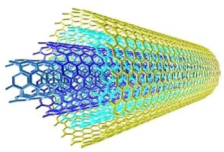
Production of hydrogen

Now a day's scientists are focusing on two major problems, i.e., environment and energy sources. The combustion of petroleum-related fuels causing major problems in earth's climate. In recent few years, scientific society working on alternative fuels to reduce global warming effects (Gayathri et al. 2010; Li et al. 2008; Verdinelli et al. 2014). Hydrogen is recognized as an alternative fuel of the future because it has less effect on the environment (Iqbal et al. 2019), efficient energy carrier, lightweight and large production as compared to other renewable sources (Jamal and Wyszynski 1994). The main application of hydrogen is in power transfer vehicles where it is utilized as fuel rightly in internal combustion engines (Amphlett et al. 1994; Eswaramoorthi et al. 2006). In past decades, researchers have faced many problems about its production at low cost and its storage. Rather et al. have reported that it is very difficult to transport and store hydrogen because of its combustible nature and high volume (Rather et al. 2009) but CNTs have fascinating properties and have shown to be an all-around substance in the area of nanoscience. For the production of systematic heterogeneous catalysts (Ijaz and Zafar 2021) in the production of hydrogen process, carbon nanotubes play a significant role (Eswaramoorthi et al. 2006; Ndungu et al. 2008). Hydrogen can be produced by four methods such as hydrocarbon steam reforming reaction, water electrolysis, gasified reactions, and oxidation reactions of dense oil (Faungnawakij

Table 1 Differences between SWCNT and MWCNT

SWCNT	MWCNT	References
It's a layer of single atoms of graphene	It's multi-layered graphene	Wang et al. (2011)
It needs a catalyst to be created during the chemical reactions	It does not need any type of catalyst to be created	
It's difficult to synthesis as a bulk material	It is easy to be created as a bulk material	
It can't be dispersed easily to the full extent	It is dispersed homogeneously	
It has appeared as bundled structure	No bundled structures	
The range of resistivity is about 10^{-4} to 10^{-3} Ωm	The range of resistivity is about 1.8×10^{-5} to 6.1×10^{-5} Ωm	
Single characterization	Complex characterization	
Flexible	Not flexible	
Poor purity	High purity	
		

et al. 2007; Wang et al. 2009a, b). Water electrolysis is the most promising method to produce hydrogen by renewable energy sources (Khalid et al. 2020). Water is a rich source of hydrogen but its splitting is difficult in the initial stage of development (Oriňáková and Filkusová 2010). Besides many advantages, water electrolysis is not considered the proper route for the creation of carbon monoxide-free hydrogen (Azadi et al. 2010). Another renewable source (Hassan et al. 2019) for hydrogen production is biomass. Biomass is receiving great attention because of its lavish energy applications (Serrano et al. 2009) but it has many disadvantages including its low efficiency (Ni et al. 2006). By the process of reforming, a large number of biomaterials have attracted researchers to produce hydrogen (Serrano et al. 2009). Biomass fermentation gives rise to bioethanol which is considered as an alternative route for hydrogen production. The presence of fast deactivation of metal catalysts (Nadeem Riaz et al. 2019) and other competitive reactions imposes limitations on this technology (Duprez et al. 1984; Tahir et al. 2019a, b). Gasification hydrogen production, biological dark fermentation, and thermochemical pyrolysis are the hydrogen production methods for its commercial utilization in the future (Ni et al. 2006). In the future, among all technology's biomass will play a vital role in hydrogen production. In fuel cell vehicles (FCVs) for onboard storage of H_2, squashed gas, liquid, or hydrogen storage substances are utilized (Eswaramoorthi et al. 2006). Hydrogen in mobile fuel cell applications is not used because of its limitations of storage and refueling. These problems are solved by producing hydrogen from hydrogen-rich liquid fuel. Ethanol is a suitable fuel for hydrogen production through reforming and decomposition as it has low toxicity, low cost, and easy generation from renewable resources (Laosiripojana et al. 2007). Many researchers explained hydrogen production by ethanol decomposition but only some of them explained the creation of carbon nanotubes lead by hydrogen through the decomposition of hydrogen (Davidian et al. 2007; Laosiripojana et al. 2007).

Methanol is also transformed into hydrogen with the use of a catalytic reactor through reforming, decomposition, and partial oxidation. Among all, the production of hydrogen through partial oxidation of methanol has more benefits as it is an exothermic reaction and has a high amount of energy to complete reaction (Jiang et al. 1993; Murcia-Mascarós et al. 2001). In recent few years, natural gas is also utilized as a source of hydrogen for fuel cells. The cost of hydrogen production is reduced from natural gas by steam reforming. It consists of some stages including high-temperature catalytic reforming of methane to produce gas, conversion of CO into CO_2 small temperature water gas shifting (WGS) reaction, separation of H_2–CO_2 mixture (Otsuka et al. 2004; Serrano et al. 2009). During discharging of hydrogen because of adiabatic expansion CO_2 and H_2O are condensed so there is a potential need to remove these impurity gases to avoid any blocking. Methane decomposition has taken much attention due to its unique applications in the CNTs and CNFs production (Serrano et al. 2009; Verdinelli et al. 2014). Yuguo and coworkers have studied that hydrocarbon-based catalytic decomposition is a suitable replacement to create carbon-free hydrogen and carbon nanotubes (Wang et al. 2005). Catalytic methane decomposition (CMD) is a short route to produce carbon-free hydrogen at a condition where CO, H_2O, and O_2 are absent (Muradov 2001). Moreover, Serrano et al. have reported that CMD is a moderately endothermic process and the amount of hydrogen production in it is less than steam refining but this quantity is remunerated with less fuel processor. The H_2 products in CMD do not produce pollutants during burning so it can be used in proton exchange membrane fuel cells (PEMFC). It was also analyzed that hydrogen is a major product produced by carbon fibers (CNFS) as a by-product (Serrano et al. 2009; Weizhong et al. 2004). So CMD is an appropriate method for producing hydrogen. From ethanol decomposition, creation of H_2 and carbon nanotubes are affected by the catalytic effect of the catalysts (Serrano et al. 2009).

Application of CNTs in hydrogen production

The creation of hydrogen and carbon from the decomposition of methane is carried by low-cost catalysts that display less deactivation throughout the reaction. Carbon-based catalysts are not high priced so their recreation is not required. To produce hydrogen free of CO and CO_2 impurities oxygen group from carbon is detached before the coming temperature of methane decomposition (Pinilla et al. 2007; Serrano et al. 2009). Catalyst decomposition is always a major problem as a result of the formation of a large amount of carbon during methane decomposition. Carbon black is better than activated carbon for methane decomposition (Huang et al. 2008). Seeing the structure and characteristics of CNTs these are used as support of catalysts. For their use as support surface of carbon nanotubes is changed to produce functional groups of definite requirements (Azadi et al. 2010; Ndungu et al. 2008). Catalysts that are supported on CNTs for hydrogen production are produced by different methods such as precipitation, chemical depletion, and impregnation and by a method of hydrothermal (Wang et al. 2006). Decomposition of ethylene over Ni assisted on CNTs provides a piece of vital information about many facts like loading of Ni metal on the activity of catalyst, catalyst synthesis method, and chemical composition of support. Catalytic performance depends upon Ni metal loading. An increase in the production of hydrogen has been seen for both decompositions of ethylene and methane (Hou et al. 2009; Reshetenko et al. 2003). The behavior of Ni-assisted catalysts could be changed with both geometric and electronic features

of support. Geometric factor and electronic factor give rise to the formation of Ni clusters and modification of electronic properties because of interaction with support, respectively. With Ni loading production of hydrogen has reached 92.5% over catalyst (Azadi et al. 2010). Catalysts of Ni–Cu alloys are produced by depletion of formaldehyde in steam refining of methanol supported on CNTs. To prepare imperfections and functional groups on its top at first CNTs were pretreated with nitric and other acids to increase their hydrophilicity. By co-depletion of Cu and Ni Precursors Ni and Cu, alloys stayed on the surface of CNTs with the use of tetra-n-methyl ammonium hydroxide to deplete the accumulation of Ni and Cu particles (Azadi et al. 2010; Hou et al. 2009; Panella et al. 2005). Ni and Cu catalyst assisted on activated carbon and CNTs were considered separately in methanol steam reforming. Production of hydrogen for Ni–Cu/CNTs was 100% at 360 °C (Sankaran et al. 2008). The catalytic behavior of Ni–Cu/CNTs was more than Ni–Cu/C also lower accumulation of particles of metal was seen by utilizing CNTs as support than utilizing activated carbon as support. Irrespective of the high surface area of activated C activity of Ni–Cu/C depleted remarkably as Ni–Cu alloy accumulated to form bigger particles (Eswaramoorthi et al. 2006). For partial oxidation of methanol, CNTs were utilized as the holder of Cu–Zn catalyst to produce hydrogen. Good quality of CNTs is produced by chemical vapor deposition method by utilizing Al_2O_3 as template and acetylene as the origin of carbon. The co-precipitation method is used to prepare Cu–Zn/CNTs catalyst (Wang et al. 2004). Strong Lewis acid cites are created when deposition of Cu over CNTs happens. Hydrogen production and methanol conversion rate increased with reaction temperature. Cu^{o} is an active species for the production of hydrogen with reduced production of carbon monoxide while unaltered Cu^{+} and Cu^{2+} ions slow down hydrogen creation from methanol (Liao and Yang 2008). Wet impregnation and chemical reduction methods are used to prepare Cu/ZnO–carbon nanotubes nanocatalyst for steam refining of methanol (Yang and Liao 2007). A good deposition of Cu nanoparticles with a size of 10 nm on the surface of CNTS is obtained. By using Cu/ZnO–CNTs as a catalyst for steam reforming of methanol production of hydrogen has been increased up to 100% at a temperature higher than 320 °C (Wang et al. 2006). For limited dehydrogenation of cyclohexane and methylcyclohexane, CNTs are utilized as support to make pt and Pd catalysts for the production of pure hydrogen. Platinum/Scandium–carbon nanotube has more catalytic efficiency and a large amount of hydrogen production for dehydrogenation of cyclohexane and methylcyclohexane (Hou et al. 2009; Wang et al. 2006). Pd supported on SC–CNTs is not much better than pt supported on SC–CNTs for dehydrogenation of cyclohexane and methylcyclohexane. HRTEM studying characteristics of catalysts showed to us that pt has more ability to disperse on SC–CNTs as support than any other supports. The main challenges that scientists are facing about the utilization of CNTs are its high cost and its production by a CVD method (Eswaramoorthi et al. 2006). However, its applications as support of catalysts are more effective regarding its employment irrespective of its high cost.

Storage of hydrogen

Volcanic nature and the high volume of hydrogen are major obstacles for safe and cheap storage to use in applications. Many technologies have been developed for secure and cheap storage of hydrogen such as liquefaction, compressed gas, underground and glass microsphere storage, in form of metal hydrides, and adsorption on large surface area substances like storage of hydrogen chemically using electrochemical hydrogen storage and complex hydrides (Panella et al. 2005; Shaijumon and Ramaprabhu 2005). Some of them are expensive and are hindered to many storing techniques like loss due to evaporation and lavishes in H_2 energy and compression requirements. Metal hydrides are the best method to store hydrogen and solving problems of pollution and reduction of fossil fuels (Zhang et al. 2004). By solid-state storage metal, hydrides are formed with metals and alloys under temperature and pressure that is better than liquid and gas storage methods. Metal hydrides have more capacity to store hydrogen density than liquid and gas hydrogen. On the other hand, metal hydrides have disadvantages such as high cost, high temperature of decomposition, and large weight (Wang et al. 2009a, b). Hydrogen storage electrochemically is an easy process in which throughout electrochemical decomposition of wet medium atomic hydrogen is adsorbed on hydrogen storage substances. This process has a high amount of capacity of storage and therefore has applications in nickel/metal hydride accumulators (Verdinelli et al. 2014). All the above methods have some disadvantages so there is a need for to latest storage techniques as physical adsorption on nano substances. Storage of H_2 on surfaces having large surface area is the best technique to store hydrogen with a capacity of 6.5% by the department of energy in the US despite its disadvantage of low weight (Panella et al. 2005; Zhou 2005). Nanomaterials possess large physical and chemical properties and have many demands in the storage of hydrogen techniques especially carbon substances such as CNTs, and CNFs have large hydrogen storage capacity (Cao et al. 2009).

Mechanism of CNTs in hydrogen storage

Due to porous structure and high surface area, CNTs adsorb H_2 for its storage on the top layer due to physical forces also known as Van der Waals forces that are present between carbon atoms and hydrogen molecules. This phenomenon is called physisorption (Lauerhaas et al. 1997). Some models

show hydrogen storage on CNTs like the atomistic model and continuum model. In the atomistic model, both C atoms and H_2 molecules are discrete. This model explains covalent bond; the interaction between carbon–carbon atoms and Van der Waal interactions, between carbon–hydrogen and hydrogen–hydrogen molecules (Dillon et al. 1997). Hydrogen molecules are randomly added then total energy is minimized that tells us about the position of C atoms and H_2 molecules. For large CNTs, continuum model is used in which CNTs are denoted by the continuum model and hydrogen molecules as internal pressure as shown in Fig. 2. As pressure increase spacing between H_2–H_2 and C–H_2 decrease but a radius of CNTs increase that leads to high hydrogen storage (Liu et al. 1999).

Effect of modifications in CNTs

CNTs being large molecules are practically insoluble in all solvents so it is hard to handle them. To solubilize them in organic solvents, functionalization of CNTs is very necessary task as functional groups can stop the accumulation of nanotubes. At first CNTs are purified with sonication that is technique for separating CNTs from catalyst impurities and carbon amorphous. In this method, –COOH and –OH groups are attached with CNTs at sidewall defects sites (Gabriel et al. 2006). Hydrogen storage in CNTs is explained by both physisorption and chemisorption processes. US department of energy (DOE) suggested a target for hydrogen storage systems that volumetric hydrogen storage density and weight storage density should not be less than 63 kg m^{-3} and 65wt%, respectively, at pressure up to 100 bar but at same time conventional hydrogen storage technologies did not match these two conditions so scientists are trying to modify materials for maximum storage capacity of hydrogen especially in CNTs. Storage capacity of hydrogen by physisorption is explained by both capacity due to compression in voids and due to adsorption on solid surfaces. Specific surface area, pore size, and porosity have a great impact on hydrogen storage capacity in CNTs as molecular sized pores have large ability to absorb hydrogen. Defects present on surface of CNTs not only decrease diffusion path but also increase pore size and surface area of CNTs to increase hydrogen storage capacity. For modifications of CNTs, physical and chemical treatments are used. Ion irradiation and milling are commonly used physical treatment methods. By increasing γ irradiation doses, hydrogen storage capacity can be increased up to 1.2 wt% at ambient pressure and 100C temperature as interaction of Compton electrons with atoms of carbon nanotubes increase defects in MWCNTs (Lyu et al. 2020). Milling also increases the hydrogen storage capacity by breaking and decreasing length of CNTs and increasing defects and surface area in CNTs. Acid, base, and heat treatments are used as chemical treatment. Chemical treatment to modify CNTs gives more selectivity and modifications are done by oxidations from defects on surfaces or from reactive end portions. Many oxidants such as sulfuric acid, nitric acid, hydrogen peroxide per chloric acid, and many others are used to form functional groups on CNTs (Çalışır and Çiçek 2020). In chemical treatments, mostly microwave reactions are preferred because these are fast, and efficient reactions also these decrease volumes of solvent, and increase selectivity, surface area and yield or reaction (Caddick and Fitzmaurice 2009). Bromine and fluorine are also used to enhance hydrogen storage capacity in CNTs because of high electronegativity of fluorine and bromine.

Applications of CNTs in hydrogen storage

Carbon can adsorb gases. This is because of its ability to have a fine powdered form with a porous structure. Many developments have been done to find out new microporous carbon materials that have high adsorbent properties (Darkrim et al. 2002). Because of the porous structure of adsorbent materials, these pores adsorb a large number of gases that give rise to a high density of adsorbent materials. Many carbon materials have been developed but carbon

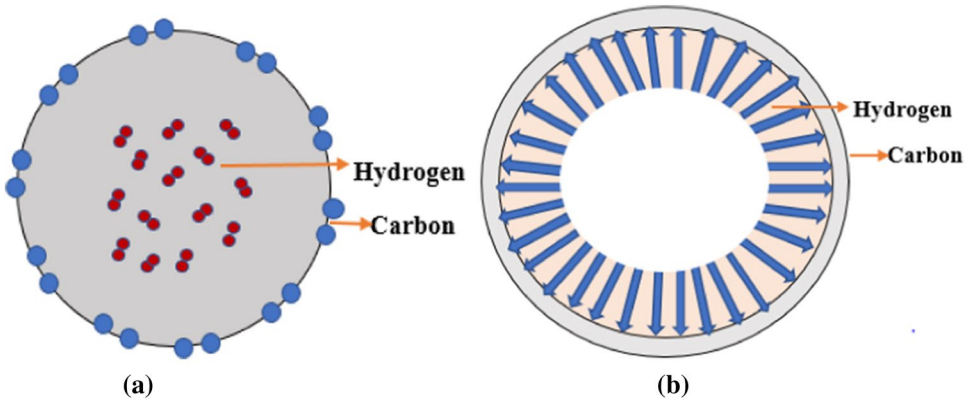


Fig. 2 Hydrogen storage in CNTs: **a** atomistic model in which both hydrogen and carbon are discontinuous **b** continuum model in which CNTs are constituted by continuum shell and H_2 by internal pressure [Represented this with permission from (YL Chen et al. 2008)].

nanotubes having large surface area, chemical stability, hollowness, and wetting properties are widely used as an adsorbent for other materials. So CNTs have elevated and reversible H_2 storage capacity (Shaijumon and Ramaprabhu 2005). Having elevated density of energy and transition elasticity at room temperature CNTs are used in onboard hydrogen storage. Distinct results of adsorption of hydrogen in CNTs substances show that by controlling microscopic parameters gas adsorption is controlled (Cheng et al. 2001; Darkrim et al. 2002). Experiment and theoretical results show to us that hydrogen is stored on CNTS by two methods one is physisorption and the other is chemisorption. In physisorption, there are weak Vander Waal forces between carbon atoms and hydrogen molecules. It is a reversible process in its gas adsorption is a function of pressure at a given temperature and desorbed as pressure decrease (H. Zhang et al. 2004). In chemisorption, there is a chemical bond between carbons of nanotubes and atoms of hydrogen. The binding energy of physisorption and chemisorption is 0.1 eV and 2–3 eV, respectively (Bénard and Chahine 2007; Jhi 2007). In metal hydrides as the interaction between hydrogen and adsorbent substance increases, molecular hydrogen is converted into atomic hydrogen, and then the rate of adsorption increases due to more diffusion of hydrogen into adsorbent (Jhi 2007; Liu et al. 2003). Many studied have been done about the interaction between hydrogen and CNTs during adsorption yet operation is not very transparent (Darkrim et al. 2002). It is considered that H_2 was stored in the inner surface of carbon nanotubes making a cylindrical form that is monolayer, or in the outer top layer or among nanotubes when there are bundles of CNTs. By studying surface modifications of CNTs, it is seen that adsorption of hydrogen favors only on some particular sites (Verdinelli et al. 2014). Three distinct sites of binding are observed where the molecular axis of hydrogen can take place: (1) above and parallel carbon ring (2) normal and over carbon ring (3) parallel and above bond of carbon (Yürüm et al. 2009). According to density functional estimations, there are two sites at end of the tube and one site in the hollow tube of CNTs. Many factors like chemical composition of the surface, surface area, size, impurities, given temperature and pressure, and defects largely affect H_2 adsorption in CNTs (Panella et al. 2005). Because of the pore structure of single-walled nanotubes, these have excellent hydrogen storage capacity. Experiments show that the capacity of H_2 adsorption for refined SWNTs is in the scope of 5–10 wt% and by changing conditions and different factors this storage capacity can be enhanced (Dillon and Heben 2001). The hydrogen storage capacity of 6.5 wt% is got when SWNTs have a diameter of 2 nm, 7 wt% for end opened SWNTs, 8 wt% for crystalline SWNTs at temperature and pressure of 80 K and 12 MPa respectively, and 4.2 wt% at room temperature by arc discharge method having a diameter of 1.85 nm (Liu et al. 1999). The hydrogen storage capacity of 20% can be got in a small milligram amount of Lithium-doped multi-walled nanotubes (Chen et al. 1999a, b). All above discussion of H_2 storage in CNTs was at starting stage and stimulated further investigation on hydrogen storage capacities. In further experiments from the pure SWNT storage capacity of 0.43wt% and 3.7wt% from acid-treated aligned MWNTs at 300 K temperature and 145 bar pressure (Chen et al. 1999a, b; Ritschel et al. 2002) was obtained as shown in Table 1. By using a CVD method, it was investigated that a storage capacity of 0.631% that is reversible was obtained at room temperature and 45 bar hydrogen gaining pressure. By studying at same time hydrogen storage properties of open-capped MWNTs, close-capped MWNTs and SWMTs at two cases one at room temperature and second at 77 K we see that in all cases open capped MWNTs have a high storage capacity of 6.46 wt% at 77 K and 1.12 wt% at room temperature (Gao et al. 2003). For SWNTs, adsorption capacity of 4 wt% was got at 1 bar and 77 K and 0.2 wt% at 1 bar and room temperature. It was investigated that structure of single-walled nanotubes favor deposition of hydrogen on graphitic structures that are found in large surface areas ACs (Poirier et al. 2004). Studying desorption properties of CNFs and MWNTs with the CVD method, it was seen that their thermal desorption was 0.7 wt% and 8.6 wt%, respectively. Hydrogen starts to desorb at 773 K but MWNTs coated with Ni have low hydrogen desorption temperature because of the catalytic effect of Ni. From the above results, we cannot conclude that which is best SWNTs or MWNTs for storage of hydrogen so further structural properties, synthesis techniques are studied that affect the storage behavior of any type of carbon nanotubes (Yamanaka et al. 2004). Now we see the effect of structural properties on hydrogen storage capacity in CNTs. By using the plasma-assisted hot filament CVD method CNTs having a diameter of 50–100 nm give a hydrogen storage capacity of 6–7 wt% (Chen et al. 2001). Comparison of aligned MWNTs and randomly oriented MWNTs at different conditions shows that aligned MWNTs are the best domain for hydrogen storage (Cao et al. 2001; Zhu et al. 2001). By using the arc discharge method aligned SWNTs showed molding ability so can be easily compressed into large tablets these tablets have H_2 the storage capacity of 4.0 wt% at ambient temperature and 11 MPa (Lee and Lee 2002). Studying H_2 capacity in MWNTs by using levitating catalyst method in both as-grown position and elevated temperature annealing showed that annealed MWNTs have a large storage capacity of 3.98 wt% at 2473 K than as grown MWNTs having 1.29 wt% at room temperature. An annealing process is done to detach functional groups from the as-grown state as after annealing C layers were bitterly piled along the axis of nanotubes (Ci et al. 2003). CNFs having a large degree of defects and herringbone structure possessed not only high hydrogen adsorption capacity but also high desorption capacity at

3.8 wt% at room temperature and 69 bar. To achieve significant levels of hydrogen storage in CNFs in-situ pre-treatment of CNFs is done as it removes moisture from it (Lueking et al. 2004). Results got from nuclear magnetic resonance and electron spin resonance showed that both SWNTs and MWNTs behave like physisorption and an upper limit of 1 wt% was guessed at hydrogen pressure up to 1480 kPa (Shen et al. 2004). SWNTs and MWNTs having large diameters with open ends and low crystallinity, respectively, showed preferable hydrogen adsorption capacity. For purified SWNTs, storage capacity of 2.4 wt% is got at 25 bar and 77 K but if pressure is changed from 25 to 35 bar storage capacity becomes doubled (Tarasov et al. 2003). When CNTs are produced by CVD method, then maximum hydrogen adsorption on CNTs with 80% purity was 0.4 wt%. Adsorption capacity of CNTs can be increased by further purification and acid washing. So, more purification steps increase the adsorption capacity up to 0.8 wt% (Rashidi et al. 2010) as shown in Table 2. Based on structural properties, SWNTS with large diameters and CNFs and MWNTs with fewer structural defects are suitable for hydrogen storage. Synthesize methods also affect hydrogen storage capacity likeCNTS made from AB_2 or AB_5-type metal hydride as catalyst indicates a high capacity of hydrogen storage. Acid and alkali treatments do not improve storage capacity in CNTs samples. A suitable activated method is required to produce imperfections on the surface of CNTs by increasing their hydrogen storage properties (Anson et al. 2004). The surface area of CNTs is very small but its hydrogen storage capacity is very high at room temperature because of the orifice structure of CNTs (Lawrence and Xu 2004).

Many scientists have attempted to explain the storage of hydrogen capacity of CNTs from theoretical estimations. Three methods are used generally such as DFT calculations, computer simulation by using a method of grand canonical Monte Carlo (GCMC), and geometrical models-based estimations. From geometrical models, it was estimated that amount of one layer of hydrogen packed on the graphene layer shows capacities of 4.1 wt% and 2.8 wt% (Dresselhaus et al. 1999). By using DFT calculations, it was estimated that storage of H_2 in SWNTs was a chemisorption reaction. Binding energy of H_2 storage of (10, 10) SWNTs at distinct rates was studied (Lee and Lee 2002). Results represented that at 50% coverage C–H having unbreakable energy of 57.3 kJ mol^{-1} was a steady state with a storage capacity of 4 wt% (Bauschlicher 2001). From DFT calculations it was said that the physisorption process in carbon nanotubes may take importance at a temperature of liquid nitrogen not at room temperature. The chemisorption process had an estimated limit of 7.7 wt% but experimentally it was hard to get because of slow kinetics (Li et al. 2003). It was seen that the physisorption of hydrogen outside SWNTs above the center of the hexagon surface was the most steady state with very low binding energy. Although chemisorption on SWNTs was known, reactions from physisorption to chemisorption did not happen impulsively which was a big problem to use SWNTs in practical for hydrogen storage (Han and Lee 2004). Hydrogen storage activity of SWNTs increased by increasing pressure and has a maximum value of 1 wt% at 20 MPa for isolated SWNTs sample. Maximum capacities of 7.1 wt% and 9.5 wt% were got for bundled and isolated SWNTs, respectively, at 77 K and 4 MPa pressure with a diameter of 2.719 nm. So, by changing different conditions and parameters storage capacity of hydrogen on CNTs can be changed by using DFT calculation theoretically (Zhang et al. 2003a, b). Maximum adsorption capacity of hydrogen with activated MWCNTs was 0.54 wt% at 77 K and1bar. Hydrogen storage affected with pore volume and pore size in activated MWCNTs is desirable for hydrogen adsorption. By using chemical activation method, hydrogen adsorption volume can be increased as by using it surface area and pore size of activated MWCNTs can be enhanced. Storage capacity can also be increased by increasing temperature (Lee and Park 2012). In chemical synthesis microwave-assisted cross-coupling method has gained popularity and is used to functionalize MWCNTs. For maximum storage capacity several modifications on surface of MWCNTs are done by using Suzuki cross-coupling reaction. Volumetric method was used to investigate hydrogen storage capacity at cryogenic temperature. To study hydrogen storage properties in MWCNTs, several compounds were synthesized by using different methods and conditions that shows different capacities (Çalışır et al. 2021). Wakayama studied storage characteristics of milled and unmilled carbon materials prepared by plasma chemical vapor deposition without catalyst and at different pressures. Hydrogen storage capacity of unmilled materials was 0.3 wt% that is less than milled materials that have storage capacity of 1 wt%. The main difference between two storage capacities is that milling increase the surface area and reduced the crystallites size. So, hydrogen storage capacity varied with type of CNTs, pressure and temperature. In plasma CVD crystallite size and surface area can be controlled by controlling and changing pressure (Wakayama 2020). By using GCMC simulations, physisorption of hydrogen on the surface of CNTs was studied. Both SWNTs and MWNTs showed different results at different conditions and methodologies (Zhang et al. 2003a, b). Purification and oxidation treatments are important ways to increase hydrogen storage capacity as these exploits merged the effect of top layer curvature and high surface area. Different theoretical and experimental hydrogen storage capacities that are shown in Table 2 were reported during 2003–2004 accompanied by different temperatures, pressure properties of a sample, and methodologies applied (Conte et al. 2004; Dillon and Heben 2001). Hydrogen storage capacities are still scattered ranging from 0.02 to 17 wt% as demonstrated in given tabular

Table 2 Utilization of CNTs in Hydrogen Production and Storage

Type of CNT	Synthesis/storage technique	Size (nm)	Storage conditions	Efficiency experimental/theoretical findings (wt%)	References
SWNT	Synthesis Technique: HiPco	–	Volumetric	Purified Experimental findings: 0.43 (adsorption)	Kajiura et al. (2003)
SWNT	Synthesis Technique: HiPco	–	Gravimetric	Unpurified Experimental findings: 1.2 (Adsorption)	Smith et al. (2003)
SWNT	Synthesis Technique: Arc discharge	1.2–1.5	Volumetric	– Experimental Findings: 0.02 and 1.58 (Adsorption)	Anson et al. (2004)
The mixture of SWNT and DWNT	Synthesis Technique: CVD	–	Volumetric	Unpurified Experimental findings: 0.51 (Adsorption)	Bacsa et al. (2004)
SWNT	Synthesis Technique: Laser deletion	1.41	Volumetric	Purified Experimental findings: 0.3 (Adsorption)	Shiraishi et al. (2002)
SWNT	Synthesis Technique: HiPco	–	Gravimetric	Purified Experimental findings: 0.2 and 1.7 (adsorption)	Poirier et al. (2004)
SWNT	Synthesis Technique: arc discharge	1.2–1.5	Volumetric	Unpurified Experimental findings: 3 (Adsorption)	Callejas et al. (2004)
SWNT	Synthesis Technique: HiPco	–	Volumetric	Purified Experimental findings: 0.25 (adsorption)	Takagi et al. (2004)
SWNT	Synthesis Technique: HiPco	–	Volumetric	90 wt% purified Experimental findings: 0.91 (adsorption)	Lawrence and Xu (2004)
SWNT	Synthesis Technique: arc ejection	1.20–1.50	Volumetric	74 wt% purified Experimental findings: 2.4 (adsorption)	Tarasov et al. (2003)
SWNT	Synthesis Technique: arc ejection	0.7–1.2	Thermal desorption spectrum	12–15 wt% purified Experimental findings: 0.6 (release)	Sudan et al. (2003)
SWNT	Synthesis Technique: CVD	0.8–1.1	Natural Gas Decomposition	Experimental findings: 0.8 wt%	Rashidi et al. (2010)
CNF	Synthesis Technique: CVD	–	Volumetric	Experimental findings: – 3.8 (release)	Lueking et al. (2004)

Table 2 (continued)

Type of CNT	Synthesis/storage technique	Size (nm)	Storage conditions	Efficiency experimental/theoretical findings (wt%)	References
CNF	Synthesis Technique: CVD	250	Volumetric	Experimental findings: – 17 (adsorption)	Gupta et al. (2004)
MWNT	Synthesis Technique: CVD	20–30	Volumetric	Purified Experimental findings: 3.3 (adsorption)	Shaijumon and Ramaprabhu (2003)
MWNT	Synthesis Technique: CVD	10–60	Volumetric	– Experimental findings: 3.98 (adsorption)	Ci et al. (2003)
MWNT	Synthesis Technique: CVD	53	Volumetric	– Experimental findings: 4.6 (adsorption), 3.2 (release)	Hou et al. (2003)
MWNT	Synthesis Technique: CVD	30	Volumetric	– Experimental findings: 0.45 (adsorption)	Zhou et al. (2003)
MWNT	Synthesis Technique: CVD	40	Volumetric	– Experimental findings: 6.46 (adsorption)	Gao et al. (2003)
MWNT	Synthesis Technique: CVD	10	Thermal desorption spectrum	Unpurified Experimental findings: 8.6 (release)	Yamanaka et al. (2004)
MWNT	Synthesis Technique: CVD	25–30	Volumetric	Purified Experimental findings: 0.69 (adsorption)	Liu et al. (2003)
MWNT	Synthesis Technique: CVD	60–100	Volumetric	– Experimental findings: 5.0 wt%	Zhang et al. (2003a, b)
MWNT	Synthesis Technique: CVD	10–30	Volumetric	85 wt% purified Experimental findings: 0.272 (release)	Ning et al. (2004)
Activated MWNT	Synthesis Technique:	Pore size approximately 0.73 nm	Volumetric	Experimental findings: 0.54 wt%	Lee and Park (2012)
Aryl Functionalized MWCNTs	Synthesis Technique: Microwave—assisted cross coupling	–	Volumetric	Experimental findings: 0.45 wt%	Çalışır et al. (2021)
Mechanically Milled Carbon Material	Synthesis Technique: plasma CVD	Crystallite size along C-axes approximately 1 nm	25 MPa	Experimental findings: 1.0 wt%	Wakayama (2020)
Un-Milled Carbon Material	Synthesis Technique: Plasma CVD	Several nanometers	25 MPa	Experimental findings 0.3 wt%	Wakayama (2020)

Table 2 (continued)

Type of CNT	Synthesis/storage technique	Size (nm)	Storage conditions	Efficiency experimental/theoretical findings (wt%)	References
SWNT	Storage Technique: Classical potential and DFT	2.71	4 MPa and 77 K	– Theoretical findings: 9.5 wt%	Zhang et al. (2003)
SWNT	Storage Technique: Classical potential and DFT	2.719	300 K and 20 MPa	Isolated Theoretical findings: 1.0 wt%	Zhang et al. (2003a, b)
SWNT	Storage Technique: Tight binding MD and GCMC	0.7	293 K and 10 MPa	Theoretical findings: 3.4 wt%	Volpe and Cleri (2003)
Li doped SWNT	Storage Technique: DFT and GCMC	0.9	50 bar RT	Theoretical findings: 6 wt%	Deng et al. (2004)
CNT and CNF	Storage Technique: GCMC	–	10 MPa and 293 K	– Theoretical findings: 0.6 wt%	Guay et al. (2004)

data. Results varied because of factors that affect hydrogen storage capacities such as structural characteristics of CNTs, surface area, pressure and methodologies of measurements, and post-synthesis treatment of CNTs (Cheng et al. 2001).

Conclusion

In this review article, the importance of the CNTs, their main element and Moore's law have been considered briefly. It's the most emerging technology nowadays in which science is making its progress. The types of CNTs that are differentiated due to the variations in their manufacture, their conditions and models to fall in a certain category are also discussed here. Further, we approached some of their manufacturing methods. Major progress of applications of CNTs in both production and storage of hydrogen are reviewed. Use of carbon nanotubes as catalyst support in H_2 production is very helpful because of its properties and structure morphology. Catalysts that supported CNTs for hydrogen production are produced by chemical reduction, precipitation, and impregnation. To produce H_2 free of C oxides and CNTs by decomposition of hydrocarbons that is non-oxidative catalytic is very effective, easy, and green chemical process and has very important technological applications. Hydrogen storage shows many challenges to producing a clean-burning hydrogen economy many efforts have been done to optimize the current status of production as well as storage technologies. H_2 the storage capacity of CNTs can be enhanced by different methods such as creating defects, ball milling, alignment of CNTs, producing carbon materials, purification, exposing heteroatoms, and doping. Now scientists are trying to store hydrogen at room temperature on CNTs but by seeing its practical applications there is a need to study further on characteristics and properties of CNTs.

Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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