

## **Feasibility of Natural Gas Storage on Carbon Nanotubes – A Review**

**Naomi A. Ogolo<sup>1\*</sup>, Ogechi C. Anih<sup>2</sup> and Mike O. Onyekonwu<sup>1</sup>**

<sup>1</sup>Institute of Petroleum Studies, University of Port Harcourt, Rivers State, Nigeria.

<sup>2</sup>Department of Chemical Engineering, University of Port Harcourt, Rivers State, Nigeria.

\*Corresponding author's e-mail address: amoniogolo@yahoo.com

### **Abstract.**

There is a growing demand for the use of natural gas as a clean source of energy but a more efficient and economic means of its storage and transportation is a challenge that constitutes an area of research interest. Natural gas can be stored as liquefied natural gas (LNG) or compressed natural gas (CNG) at about 20-30MPa in heavy steel cylinders, but carbon nanotubes (CNT) have been proposed to be more advantageous in storing and transporting natural gas in light weight containers at a pressure of about 4Mpa. There are two kinds of CNT; single walled carbon nanotubes (SWCNT) and multi-walled carbon nanotube (MWCNT) and their mechanism of gas storage is adsorption. Reports have shown that there is a considerable increase in methane adsorption on SWCNT in the presence of structural defects than in a perfect structure. Studies have also shown that the capacity of MWCNT to store natural gas can be enhanced by treating the nanotubes with acid. It has been generally observed that the methane adsorption capacity of CNT increases as pressure increases but decreases as temperature increases. CNT is speculated to become a prospective candidate for effective storage and transportation of natural gas and may compete favorably with LNG and CNG in future.

**Keywords:** Natural gas, carbon nanotube, adsorption, methane, storage, transportation

### **1. Introduction**

The vast potential application of nanotechnology has propelled numerous researches in various fields including the oil and gas sector. There are several challenges in the oil and gas industry that nanotechnology could address if articulately harnessed through research and one of such challenges is natural gas storage and transportation. Several types of porous media for gas storage have been proposed, developed and studied, and these include molecular sieve, activated carbon, zeolite and carbon nanotubes <sup>[1]</sup>. Research has shown that carbon nanotube (CNT), products of nanotechnology have the capacity to store natural gas through a comparatively cheap and efficient method, though investigations are still ongoing in solving

the challenges facing this method. This paper is thus aimed at reviewing the prospects of storing and transporting natural gas in CNT and it highlights some factors that can enhance the gas storing capacity of methane in nanotubes.

Presently, natural gas can be stored and transported through compression (compressed natural gas – CNG) and liquefaction (liquefied natural gas – LNG) but these methods are complex and expensive. In compression, the gas is stored as a supercritical fluid at room temperature but at high pressures of about 20-30MPa, reaching a density which is about 230 times higher (230 v/v) than the density of natural gas at standard temperature and pressure (STP) conditions <sup>[2, 3]</sup>. In this case, the energy density is approximately 25% of that of gasoline. The disadvantages associated with this storage method include the risk of carrying highly pressurized tank in transit, high energy requirement for raising the pressure to 20-30MPa, the costs associated with acquiring the heavy thick walled steel cylinders that can withstand such pressure, safety valves requirements and the cost of transporting these heavy cylinders containing highly pressurized gas. In liquefaction, natural gas is stored at the critical temperature, 112K (-161°C) in a cryogenic tank at a pressure of 0.1MPa where the energy density is approximately 72% of the total of gasoline. This means that one volume of LNG corresponds to 600 volumes of natural gas under STP (600v/v) conditions <sup>[2, 4]</sup>. Natural gas can be stored in CNT which are light weight containers stuffed in a pressure vessel of about 2 - 4Mpa. This method reduces cost; it reduces the risks associated with other storage methods and is a possible alternative for large scale transportation of natural gas. A comparison between the natural gas storing capacity of CNT and CNG has shown that although CNT stores less amount of natural gas, it however does this at 83% storage pressure lower than CNG <sup>[5]</sup> which is a huge advantage that can be exploited to efficiently and economically store and transport natural gas.

## **2. Types and Characteristics of Carbon Nanotubes (CNT)**

Carbon nanotubes are allotropes of carbon with a concentric cylindrical shape of diameter in the order of nanometer and length of micrometer. They are tubular cylinders of carbon atoms with extraordinary mechanical, electrical, thermal, optical, and chemical properties <sup>[6]</sup>. CNT were first discovered by Iijima in 1991<sup>[7]</sup> however, the first microscopic production of CNT was made by two researchers at Nippon Electric Company Limited fundamental research laboratory <sup>[8]</sup> and since then, there have been several developments of CNT. There are basically three methods of producing high quality CNT namely the arc discharge, <sup>[9, 10, 11]</sup> laser vaporization <sup>[12]</sup> and chemical vapor deposition <sup>[13, 14]</sup> methods but the most popular and most widely used method of synthesis is the chemical vapor deposition method.

There are two types of carbon nanotube based on the number of layers or walls; the single-wall carbon nanotubes (SWCNT) and the multi-wall carbon nanotube (MWCNT). The SWCNT can be described as a graphene sheet rolled into single cylindrical shape so that the structure is one dimensional with axial symmetry <sup>[15]</sup>. Most SWCNT typically have diameters

in the range of 1 - 1.3nm and a few micrometers long. SWNT can be formed in three different designs; Armchair, chiral and Zig-Zag. The design depends on the way the graphene is wrapped into a cylinder which could be represented by a pair of indices  $(n,m)$ . The integers  $n$  and  $m$  denote the number of unit vectors along two directions in the crystal lattice of graphene. If  $m = 0$ , the nanotubes are called zigzag nanotubes, if  $n = m$ , they are called armchair nanotubes and if  $m \neq n$  they are called chiral. The values of the integers  $n$  and  $m$  greatly affect the property of SWNT. The MWCNT is a multi-layer of graphene rolled and superimposed on each other <sup>[16]</sup>. The outer diameters are typically in the range of 2 - 100nm while the inner diameters are in the range of 1- 3nm and the length is one to several micrometers <sup>[17]</sup>.

SWCNT are more flexible than MWCNTs. They can be twisted, flattened and bent into small circles or around a sharp bend without breaking, thereby increasing its applicability <sup>[18]</sup>. SWCNT have the unique electronic and mechanical properties which can be used in applications such as field-emission displays, nano-composite material, nano-sensors and logical elements. MWCNT exhibit some advantages over SWCNT such as higher surface to volume ratio, <sup>[19]</sup> they are easier to produce in high volume quantities, the product cost per unit is low and its thermal and chemical stability are enhanced. However, MWCNTs have regions of structural imperfection which may reduce its desirability for application <sup>[20]</sup>.

CNT are light in weight and have the strongest tensile strength as compared to any synthetic fiber. This strength results from the covalent  $Sp^2$  bonds formed between the individual carbon atoms <sup>[21]</sup>. A standard SWCNT can withstand a pressure of 25Gpa without deformation <sup>[22]</sup>. In terms of thermal conductivity, nanotubes are good conductors along the tube axis but good insulators lateral to the tube axis <sup>[23]</sup>. Measurements carried out shows that SWCNT have better thermal conductivity compared to copper at the same conditions <sup>[24, 25]</sup>. MWCNT exhibit a striking telescoping property whereby an inner nanotube core can slide almost without friction within its outer nanotube shell, thus creating an atomically perfect linear or rotational bearing. The symmetry and unique electronic structure of graphene strongly affect its electrical properties. All CNT have a large surface area and a high level of adsorption.

### **3. Natural Gas as a Clean Energy Source**

Natural gas is a homogeneous mixture, having variable proportions of hydrocarbons. The general composition of natural gas includes methane ( $CH_4$ ) constituting the major part and it generally ranges from 55 - 98% in volume, ethane ( $C_2H_6$ ), propane ( $C_3H_8$ ) and other heavy constituents. There is a growing interest in the use of natural gases as an alternative source of energy especially because it is clean. Its thermal efficiency is higher than that of other fuels and it produces mainly  $CO_2$  and water vapor. The emissions of  $CO_2$  are 25-30% lower than that generated from fuel-oil and 40-50% lower than coal per unit of produced energy <sup>[26]</sup>.

As regards its use as a fuel, natural gas has many advantages. These include reduction in post combustion contaminants, reduction in maintenance costs compared to other fuels, <sup>[3]</sup> reduction in suspended solid particles which are associated with combustion of gasoline, absence of sulfur and sulfur dioxide (SO<sub>2</sub>) emissions which are typical contaminants from transportation vehicles. Compared to liquid fuels, the emissions from natural gas vehicle combustion is 76% less in carbon monoxide, 75% less in nitrogen compounds, 88% less in hydrocarbons and 30% less in Carbon dioxide. Furthermore, the physiochemical properties of natural gas enable the use of catalysts for the combustion of gases, obtaining excellent results and minimizing emissions <sup>[27]</sup>. In spite of the numerous advantages associated with the use of natural gas as a fuel over other forms of hydrocarbon fuels, its efficiency and economics in storage and transportation has constituted a major barrier to its usage. This could be attributed to its low energy density (heat of combustion/volume) at standard temperature and pressure conditions <sup>[28]</sup>.

#### 4. Storage Mechanism of Gas in Carbon Nanotubes

CNT store gas through the process of adsorption. Adsorption of a gas is a process that gains one or more constituents of the gas in the region of the gas-solid interface where the molecules of the gas are bounded to the surface of the adsorbent as illustrated in Figure 1. Adsorption is a superficial process and the phenomenon involves an increment of the gas density in the neighborhood of the contact surface and since the process is spontaneous, the change in the free energy of Gibbs is smaller than zero. Given that the entropy change is also below zero (a decrease in the freedom degree of the gas molecules during the process), the enthalpy change is lower than zero thus, the process is exothermic <sup>[29]</sup>.

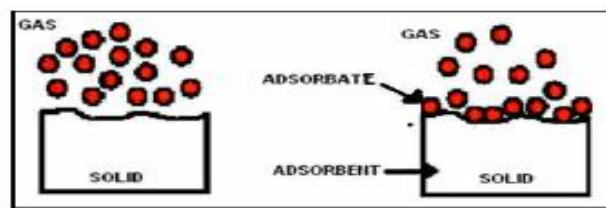


Figure 1: Representation of the adsorption process of a gas on a solid surface

There are two types of adsorption; physisorption and chemisorption. In physisorption, the bounding of the gas molecules is superficial because the gaseous molecules are not chemically bounded to the walls of the adsorbent. Weak Van der Waals forces are responsible for holding the molecules of the gas to the adsorbent. The adsorption of methane on CNT is most likely through the process of physisorption where the adsorbents require an elevated exposed surface per gram of material called specific surface area and it is expressed in cubical centimeters of adsorbate per gram of adsorbent. When the elemental constituents of the solid get smaller, the specific surface area gets larger. CNT that have diameters in the order of nanometers and with hollow cylindrical surfaces present good specific surface areas and are

an excellent adsorbent for adsorption of gases. As the pores of a CNT surface decrease, the gas storage capacity increases <sup>[28]</sup>.

### **5. Factors that Affect the Storage Capacity of Natural Gas in CNT**

In a conducted simulation work to study the storage capacity of SWCNT at different temperatures and pressures, it was observed that methane is weakly adsorbed in SWCNT. Results showed that as pressure increases, the amount of methane adsorbed on SWCNT increased but as temperature increased, the amount of methane adsorbed on SWCNT decreased <sup>[29]</sup>. It was reported that the binding energies for methane on the defected SWCNT increased by about 56% over the defect free SWCNT showing that the presence of defects on the structure of nanotubes increases its methane adsorption capacity. Furthermore, for the encapsulated methane molecules inside the defected (10,0) nanotubes, results showed about 68% increase in binding energy compared to the confined molecules in the defect free (10,0) nanotubes. It was pointed out that introducing surface curvatures in the nanotubes could reduce the binding energy between the methane molecules and the substrate <sup>[30]</sup>. Thus, some factors that affect methane adsorption in SWCNT are pressure, temperature, structural defects and curvatures.

The methane storage capacity of MWCNT has been studied by several researchers. One set of results showed that a type of MWCNT strongly adsorbed methane at a maximum value of 5.44mmol/g at a temperature of 283.15K and a pressure of 40bars. It was also reported that increasing pressure increased the amount of methane adsorbed, while increasing temperature decreased the amount of adsorbed methane <sup>[31, 32]</sup>. Another report has it that treating MWCNT with acids such as HCl and HNO<sub>3</sub> improves its methane adsorption capacity. The results of experiments conducted using acid treated MWCNT and untreated MWCNT at the same pressures revealed that acid treatment of nanotubes enhances methane adsorption capacity especially at low pressures <sup>[33]</sup>. In another work, the methane adsorptions on MWCNT treated with sulfuric and nitric acids, nitric acid and alkaline were compared with the methane adsorption capacity of untreated MWCNT. Reported results showed that sulfuric and nitric acid treated MWCNT adsorbed more methane than all other treated and untreated nanotubes. This was followed by nitric acid treated case before the alkaline treated nanotubes. The methane adsorption capacity on all the treated nanotubes was higher than the untreated cases showing that treating MWCNT with acids enhances its methane adsorption capacity. This work also showed that increase in pressure increases methane adsorption while increase in temperature decreases methane adsorption on MWCNT <sup>[32]</sup>.

### **6. Prospects of CNT as a Natural Gas Storage Device**

Carbon nanotubes have many industrial applications <sup>[34]</sup> but more attention is currently given to it as a material for natural gas storage. Several researches have been conducted to investigate the adsorption behavior of methane which is a major component of natural gas on SWNT and MWNT. The key to successful commercialization of CNT for gas storage is the

adsorption capacity of the nanotubes at standard conditions, that is, the ratio of the volume of adsorbed natural gas to the volume of storage container ( $V_g/V_s$ ). There are indications that substantial volume of methane can be stored in activated carbon pellets at atmospheric conditions, <sup>[35]</sup> in fact an adsorption capacity of  $126V_g/V_s$  has been reported <sup>[36]</sup>. Commercial development of CNT for adsorption of natural gas requires high storage gas capacity greater than  $150 V_g/V_s$ <sup>[37]</sup>. There are other advances in gas storage on CNT <sup>[38, 39, 40]</sup> that can be leveraged on to improve the natural gas storage capacity on nanotubes. It should however be noted that a volumetric capacity of about  $160 V_g/V_s$  for methane adsorption on SWNT has been reported <sup>[41]</sup>.

One of the challenges facing the wide use of CNT as a major adsorbent for natural gas storage is achieving consistency in storage densities and replicable manufacturing capacity. The methods used to manufacture the carbon nanotubes that give the required capacity are quite rigorous, and research in its reproducibility are still ongoing. There is need to control the pressure, temperature and the flow rate at which gas is efficiently filled and released from the CNT tanks for automotive application. These conditions affect the performance of the storage system since the adsorption process is exothermic <sup>[42]</sup>. The cost of producing, purifying and tuning the CNT to obtain the required diameter in large quantity makes the existing methods economically viable.

## **7. Conclusions**

Conclusions drawn from this review are as follows:

1. Natural gas is a clean source of energy but an efficient and economic means of its storage and transportation is a challenge that constitutes an area of growing research interest.
2. CNT have the potential to store natural gas at low pressures and they are prospective economic and efficient candidates for storing and transporting natural gas.
3. CNT store natural gas through the process of Adsorption.
4. Structural imperfection on SWCNT improves its methane adsorption capacity.
5. Treating MWCNT with acids enhances its methane adsorption capacity.
6. Increasing pressure increases the methane adsorption capacity of CNT while increasing temperature decreases the methane adsorption capacity of CNT.
7. Natural gas storage and transportation using CNT may compete favourably with LNG and CNG in future.

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