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Carbon nanotubes based gas sensor

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ABSTRACT

Carbon nanotube (CNT) based Gas sensors are at- tracting huge research interest as it gives high sensitivity, quick response, and stable sensors for industry, biomedicine, and more. The development of nanotechnology has opened a new gateway to build highly sensitive, cheap, portable sensors those who have low power consumption. The extremely high surface to volume ratio and the hollow structure of nanomaterials is perfect for the adsorption of gas molecules. Mainly, the advent of carbon nano- tubes has boosted the inventions of gas sensors that exploit CNTs unique morphology, geometry, and properties. Upon exposure to some gases, the changes in carbon nanotubes properties can be determined by many methods. Therefore, carbon nanotube-based gas sensors and their mechanisms are widely studied. In this paper, a broad survey of current carbon nanotubes based gas sensing technology is presented. Few experimental works done are reviewed. The types, fabrication, and the sensing mechanisms of the carbon nanotubes based gas sensors are discussed. The challenges of the research up to some extent are also addressed in this paper.

Keywords: *Carbon Nanotubes, ARC Discharge, Laser Ablation, CVD, Adsorption.*

1. INTRODUCTION

The demand for small-sized devices and systems is increasing day by day. The advantages like the small area and less power consumption have grasped the attention of various research institutes. One of them is the Carbon nanotubes, these nano structures have found their application in various fields that include biomedical, actuators, sensors, small devices etc. Carbon nanotubes are formed of the allotropic form of carbon Graphene. Structurally CNT appears like rolled up sheets of graphene with length: diameter ratio of up to 1.32*108: 1 , which is very high compared to many materials. Among its numerous application CNT gas sensors have found their way through to their vast commercial and industrial application. CNT have grabbed attention to its impressive properties like small size high thermal and electrical conductivity indicating its wide application in various fields.

CNT based ammonia gas sensors have increasing demand since keeping track of ammonia in industrial areas is essential to ensure a safe workplace. In addition to detection of hazardous gas and ensuring safety, C02 based gas sensors are used in the health industry to ensure avoiding and spoilage. It has been observed that CNT experience change in conductivity on exposure to different gas for example conductivity of CNT changed up to 3 times when exposed to nitrogen dioxide as compared to metal oxide, this inturn directed towards CNT enormous application in gas sensors.

Basic criteria for an efficient gas sensing device include -

(i) Selectivity high sensitivity, (ii) fast response time recovery time, (iii) low analyst consumption, (iv) low operating temper- ature temperature independence, (v) stability in performances, of which most of them are satisfied by CNT based gas sensors. Most of the gas sensors are based upon absorption of gas molecules and then detection of changes in sensor properties is inferred to detects its effect, incase of CNT based senors mainly change in I-V characteristics is studied since CNT experience change in conductivity on exposure to different gases other methods are also available to sense the gas offering CNT based gas sensors an edge over other sensors. In this paper, we will study about CNT and how they are applied in Gas sensors.

2. CNT AND ITS TYPES

Single wall carbon nanotubes Single-wall nanotubes are formed by an only single sheet of graphene. If we wrap graphene sheet to form a tube SWCNT but that's not how they are produced though, their production involves various different complex processes.

Single wall Nanotubes have an average diameter 0.1 to 2 to 3nm and average length in micrometers. The properties of these nano tubes depend upon chirality(a type of wrapping) and diameter of the tube.

The SWNT can be further classified as an armchair, zigzag or chiral based on the pair of indices (n,m) that describe the chiral vector and various properties. Chirality plays a major role in deciding the electrical properties of nanotubes.

when m= 0, the nanotubes are named zigzag nanotubes and when n = m, the nanotubes are named armchair nanotubes, and other states are called chiral. This chirality also play role in deciding whether the SWNT will act as metallic or semiconductor. If the value of n-m=3i that multiple of 3 then the nanotube will act as metallic carbon nanotube if it's not a multiple of 3 it acts as semiconducting.

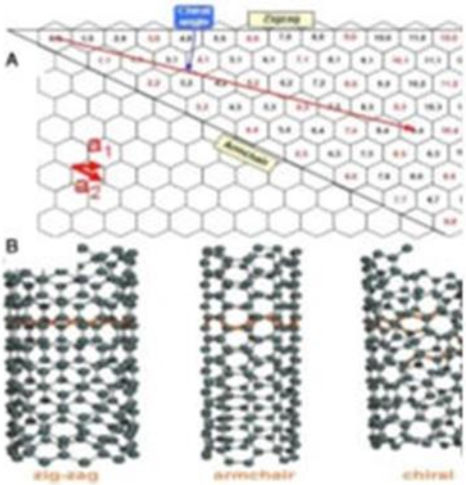

Fig. 1. Chirality of CNT

Multiple walls cnt The Multi wall carbon nanotubes consist of single wall carbon nano tubes laid coaxially or concentric layers of graphene with interlayer distance 0.34nm. There are two types of MWNT:

1) Russian doll model look like concentric layers of the graphene sheet

2) Parchment model- appear like a rolled up around itself like a rolled up paper.

Smaller diameter mwnt can be effectively produced through arch discharge rather than CVD.

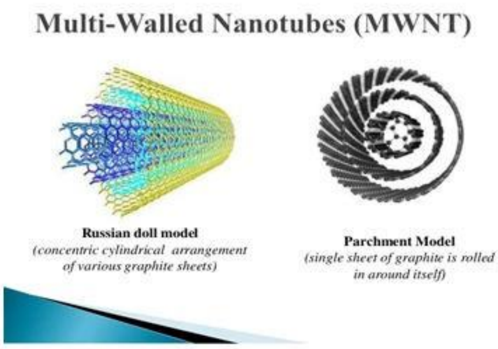

Fig. 2. Multiwalled Carbon Nanotubes

Comparison between SWNT and MWNT

SWNT	MWNT
Single layer of graphene	Multiple layers of graphene
Catalyst is required for synthesis	Can be produced without catalyst
Bulk synthesis is difficult as it requires	Bulk synthesis is easy
Purity is poor	Purity is high
A chance of defect is more	A chance of defect is less
Less accumulation in the body	More accumulation in the body
Characterization and evaluation is easy	It has very complex structure
It can be easily twisted and is more pliable	It cannot be easily twisted

3. CARBON NANOTUBES PROPERTIES

A. Electronic Structure

Carbon is an element with atomic number 6 with electronic configuration 1s2,2s2,2p2, hence can form sp,sp2 and sp3 hybridisation. The forms of carbon in sp2 hybridization can yield different shapes like honey comb or graphite-like struc- tures example -graphene ,fullerene and carbon nano tubes. In Carbon nanotubes, the carbon atom is covalently bonded with 3 neighboring atoms using sp2 hybridization and thus forming the honeycomb-like structure.

B. Electrical properties

The chiral vector C = na1 + ma2 (a1 and a2 are the base cell vectors of graphite) determines the tube diameter, and this vector finds out the direction of rolling a graphene sheet. The electrical property of a CNT depends on diameter and chirality of cnt. Depending on the chiral indices, CNTs exhibit both metallic or semiconducting properties.

The electrical conductivity of MWNTs is quite complex as their inter-wall interactions non-uniformly distribute the current over individual tubes. But in metallic SWNT uniform distribution of current can be seen. Electrodes are placed in order to measure the conductivity and resistivity of different parts of SWNT rope. The measured resistivity of the SWNT ropes is in the order of 104 cm at 27 C.

C. Strength and Elasticity

SWNT have higher elastic modulus than steel due to which they are highly resistant. It has been observed that youngs modulus of carbon nanotubes depends less on chirality and more on disorders in the CNT. Young's modulus of up to 1.8 Tpa has been reportedly achieved with other SWNT raging with 1.22-1.26 TPa.

Deformation in case of CNT is not permanent as they tend to regain their shape but deformation above a particular threshold may lead to permanent deformation. Each carbon atom in a single sheet of graphite is connected through the strong chemical bond to its three neighboring atoms. Hence, CNTs can exhibit the strongest basal plane elastic modulus and so are expected to be an ultimate high strength fiber.

D. Thermal Conductivity

CNTs posses higher thermal conductivity and large in-plane expansion thus implying its large application in sensing and actuating devices. Superconductivity can be observed in CNT below 20 K due to strong in-plane C-C bonds, this also allows high strength and stiffness against the axial strain. CNT has also been able to improve thermo mechanical and thermal properties of the composite material. CNT can with stand temperature of up to 750 C at normal atmospheric pressure and up to 2800 C in a vacuum which is pretty high-temperature standards.

4. FABRICATION OF CNT

A. Growth of CNT

There are many techniques that are used to fabricate CNT structures, and most of them involve gas phase processes. Most commonly used processes for CNT fabrication are:

(1) The chemical vapor deposition (CVD) technique[3,4,5]

(2) The laser ablation technique[3,4,5], (3) The carbon arc- discharge technique [3,4,5,6] . Low-temperature chemical vapor deposition (CVD) methods have replaced High temper- ature preparation methods like laser ablation and arc-discharge

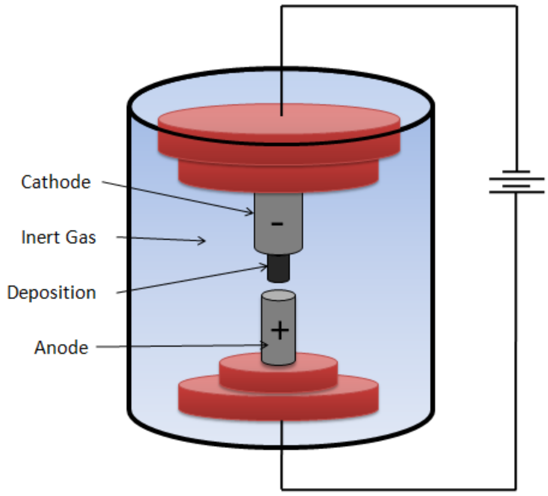

Fig. 3. Electric Arc Discharge

1) Electric arc discharge: This is the first technique that was used to grow CNTs [1,3,7]. Very high temperature is required (above 1700oC) for CNT synthesis. It is carried out in a vacuum chamber, there are two carbon electrodes as a carbon source. Inorder to increase the speed of carbon deposition Inert gas is supplied (typically helium).after applying high DC voltage between a carbon anode and carbon cathode, plasma of the inert gas is generated and evaporates the carbon atom. These ejected carbon atoms are then deposited on the negative electrode to form CNTs. SWCNT and MWCNT can be grown using this method, the catalyst is required for the production of SWCNT, synthesis of MWCNT can be done without the use of catalyst precursors. Studies have shown that Ni-y-graphite mixture can produce high yields (¡90

The main advantage of This method is its ability to produce a large number of nanotubes.the disadvantage is that we cannot control the chirality of created carbon nanotubes, which in turn helps to define its property.

2) Chemical Vapour Decomposition: In this method, a gas hydrocarbon source (usually methane, ethylene or acetylene flows into the chamber. this is one of the standard methods. There are many different types of CVD such as catalytic chemical vapor deposition (CCVD) plasma enhanced (PE) oxygen assisted CVD, water assisted CVD, microwave plasma (MPECVD), radio frequency CVD (RF-CVD). currently, the standard technique for the synthesis of carbon nanotubes is cat- alytic chemical vapor deposition. In CVD hydrocarbons molecules are broken into reactive species at a temperature range of around 500-1000oC. these species react in presence of a catalyst that is present on the substrate, which leads to the formation of CNTs. In this method CNT can be synthesised at relatively lower temperature as compared to above two methods, there- fore this method is more efficient.high quality SWCNT can be obtained by optimizing catalyst. The one disadvantage of this method is that we get high defect density in MWCNTs.

In all the methods discussed above, CNT comes with a number of impurities, which may have a negative effect on

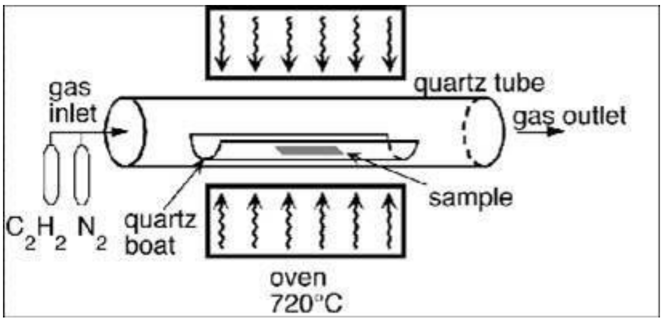

Fig. 4. Chemical Vapour Decomposition

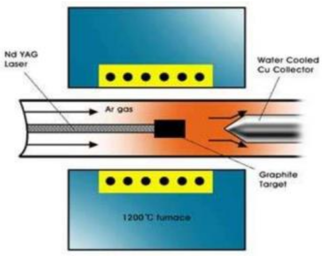

Fig. 5. Laser Ablation

properties of CNT. so purification is needed so as to make them efficient. Mainly the impurities are metallic impurities.

3) Laser Ablation: In this method, Intense laser pulse is ablated on the carbon target in a furnace in presence of a catalyst and inert gas. temperature required of this method is 1200oC, Studies have shown that the diameter of nanotubes depends on laser power. more the power thinner the diameter, a large quantity of nanotubes can be obtained using this method. In this method, there are certain parameters which when changed can have an effect on the properties of carbon nanotubes like chemical composition and structure of target material, the distance between target and substrate, laser properties. This method can be used to produce high quantity and quality of SWCNT. mechanism of arc discharge and laser ablation is almost similar, in this method the energy is provided by the laser that is focused on target[3].

The advantage of this method is that it gives very high yield and also the impurities are less, and the disadvantage is that the nanotubes formed contains branching i.e they are not uniformly straight. But it is not economically advantageous as it contains graphite rods, high laser power and also the yield per day is less.

B. Fabrication of CNT Gas Sensor:

There are many methods to integrate CNT to gas sensor structures. Li et al. developed a resistive gas sensor by simply casting SWCNTs on electrodes (IDEs). The electrodes were fabricated by photolithography, evaporation of Ti and Au on silicon oxide. As grown SWCNTs were purified with acid first, then by air oxidation before being integrated with the IDEs. As a result, final SWCNTs had a high purity up to 99.6

Fabrication of CNT sensors is also achieved by a method of

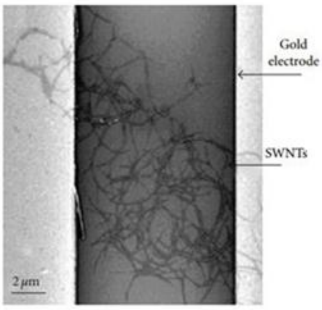

Fig. 6. SEM image of SWCNT across two gold Electrodes

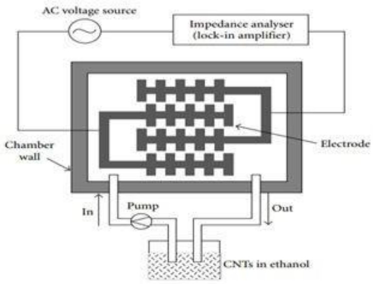

Fig. 7. Electric Arc Discharge

dielectrophoresis. DEP is the electrokinetic motion of dielectri- cally polarized materials in a nonuniform electric fields also has been used to manipulate CNTs for separation, positioning, and orientation of CNTs. Suehiro et al. [8] demonstrated that DEP fabrication could establish a good electrical connection between CNTs and electrodes. During fabrication, the CNTs with high purity were suspended in ethanol and ultrasonicated for around 60 minutes. The system to fabricate the CNTs- the based gas sensor is depicted in Figure 2. An interdigitated microelectrode was designed on a glass substrate. the electrode was with castle wall pattern in order to form high - low electric field regions periodically. the castle-wall electrode was surrounded by a silicon rubber spacer so as to form a sealed chamber in which CNTs suspension was continuously fed from the reservoir with a peristaltic pump.

The DEP trapping of MWCNTs on the microelectrode was performed with ac voltage. After a desired period of time, the DEP process stopped and ethanol was evaporated at room temp. The DEP fabricated CNT gas sensors successfully detected various vapors like NH3, NO2, SO2, and HF. By following this method, the amount of trapped CNTs can be controlled just by monitoring the impedance of the sensor and also different metal materials can also be used as the electrodes [9].

To obtain well-aligned CNTs for better sensor behavior, directly growth of the CNTs mats on the sensor substrates is required. Huang et al. [10] fabricated three terminal N2 sensor which was vertically aligned CNTs. The CNT was grown by thermal CVD on (100) n-type silicon substrate at temp 700C. A 30 nm thick Fe layer was also sputtered on the substrate as catalyst, C_2H_2 was used as the carbon source. preheating process was carried out before the growth of CNTs, the substrate was cooled down at room temperature in N_2 ambient.

5. GENERAL THEORY ON CNT BASED GAS SENSORS

A. Molecule Adsorption on SWCNTs

Owing to the unique surface structure of carbon nanotubes, the entire weight is mostly concentrated on the surface of the CNT. It has been proved in many experiments that electrochemical properties of CNT change considerably when exposed to the environment due to its high sensitivity for gas molecule adsorption.[1-2] This extraordinary adsorption capacity and sensitive electrochemical properties of the CNT gives us the new way of designing sensors on the basis of nanotubes. Peng and Cho studied the adsorption of NO_2 on to SWNTs by using rst-principles calculations using density functional theory (DFT) [3]. The adsorption of gas molecules on CNTs surface is studied. They also observed the binding energy, tube-molecule distance, and change.

Their work shows that most of the studied molecules (except for NO_2 and O_2) are charge donors with small charge transfer (0.01 0.035e) and weak binding (0.2eV). While O_2 and NO_2 being charge acceptors, it they both are with considerably large charge transfer and adsorption energies.

B. Effects on Electronic Properties of CNTs

Collins observed the electrical resistance, thermoelectric power (TEP), and local density of states of SWNTs by SEM and TEM method [4]. Generally, the electrochemical property changes of CNTs upon exposure to gas molecules are due to the charge transfer between the molecules and the nanotubes. But Sumanasekera showed that both of the thermopower and the resistance of the degassed SWNT R can be very sensitive to inert gases [5]. Without charge transfer, it is believed that the resistance changes are due to the change in the electron and hole free carrier lifetimes. These changes in the carrier lifetime can be caused either by the increased carrier scattering from temporary defect states due to adsorbed gas. Based on these observations, it is concluded that the changes in the thermoelectric properties of SWCNTs can be either caused by a change of Fermi energy caused by charge transfer or by the additional scattering channel.

In an SWCNT, at low temperatures, electrons pass through the nanotube without scattering, according to the quantum ballistic mechanism of conductivity. In this case, the resistance of an SWNT is expressed by the following equation -

$$R = h4e^2 \qquad (1)$$

6. GAS SENSORS BASED ON CARBON NANOTUBES

Owing to the limitation of geometries of sensor fabrication, single SWCNT is rarely used for sensor fabrication. Instead of a single SWCNT, dense SWCNT lm is mostly used. They

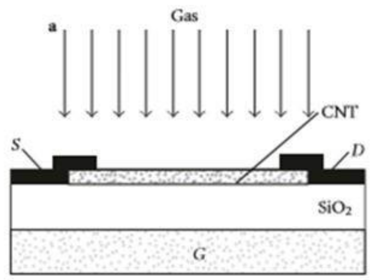

Fig. 8. SWCNTs based FET sensor

have a very fast response time, but long recovery time associated with high binding energy between gas molecules which makes desorption little slow and difficult. Currently, there are 4 major types of gas sensors based on CNT which are in use.

A. Adsorption based gas sensors:

Since sensors based on a change in properties like resistance are easy to design, adsorption-based gas sensors are the most used and common type of sensors. The basic principle of adsorption based CNT gas sensors is that when gas molecules get adsorbed by the surface of CNT, there is an exchange of electrons between gas molecules and CNT, which prompts the changes in electrochemical properties of CNT which can be detected easily by observing either the resistance or conductivity of it. Two methods are discovered so far for this purpose.

1) Semiconducting CNTs FET Gas Sensors: In CNT FET a P-type single CNT is connected between two metal contacts. Such devices can be designed by controlled growth of SWCNT using CVD method on SiO_2 substrates. Kong observed that when CNT FET is exposed to NO_2, there was an increase in conductance of CNT by the order of three[7]. The response time for the same was 2-10 seconds.

2) CNTs-Based Two Terminal Resistor Gas Sensors: This sensor generally consists of a CNT connected to two elec- trodes at its two ends. The change in resistance of CNT is detected at the two electrodes by applying DC voltage to them. Generally, interdigitated electrodes are designed so that larger sensing areas and sucient contacts between the electrodes and the CNTs are

provided. Several experiments were done using the same method to detect other gases too. It was observed that NO2 decreases the electrical resistance while other gases like NH3, H2O, C6H6, and ethanol on exposure increases the electrical resistance. The response time for these sensors is considerably high. For NO2, it was around 10 hr, but it can be reduced using UV light.

B. CNTs Based Gas Sensing Capacitor

Yeow and She designed a capacitance sensor consisting array of disoriented nanotubes grown on a SiO2 layer as a sensitive element. CNT array worked as one plate of sensor and silicon as another one [11]. When an external voltage is applied across the device, high magnitude electric eld is generated at the CNT terminations. This results in the polarization of

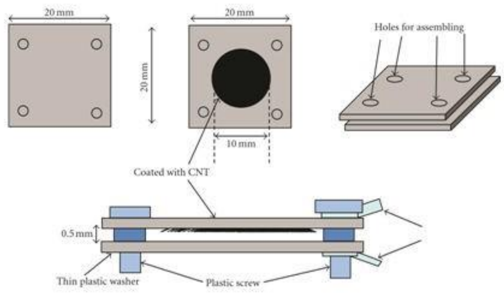

Fig. 9. Parallel plate capacitor having one plate with CNT on it

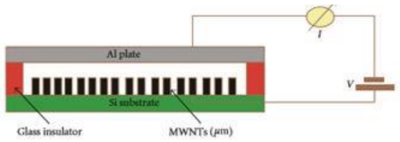

Fig. 10. CNT based Ionization chamber setup

adsorbed molecules and an increase in the capacity. Snow also developed a same capacitor based device and obtained the same results [12]. These sensors possess high sensitivity for a number of gases. Despite having such advantage, its applications are limited. The sensing action causes some irreversible changes in the CNT associated with gas adsorp- tion. These sensors require constant replacement of sensitive element.

C. Ionization gas sensors

Forementioned methods are not adequate to detect gases with low adsorption energies or inert gases. In such cases, ionization method is best conducted where gas ionization pa- rameters are observed during a collision between gas molecules and ion.

Modi in his experiment designed a device consisting of MWCNT lm as an anode, aluminum plate as a cathode and a 150m thick glass plate between them [13]. The MWCNT films of about 25 30nm diameters and 30 m length were grown vertically using CVD on SiO2 substrate. A very strong nonlinear electric eld is formed by MWCNT after applying a voltage between anode and cathode which creates the conduc- tion path of ionized gas around the tip of CNT. This results in the development of self-sustained electrode discharge even at lower voltages. Using this method, breakdown voltages were brought down considerably. These unique breakdown voltages for each gas provides a solid way for detecting various gases because the breakdown voltages rarely change for a gas molecule according to its concentration.

Another theory was mentioned which contradicts the afore- mentioned theory. According to this theory, the breakdown voltage decreases with increasing concentration and then in- crease again. The decrease in breakdown voltage at rst was attributed to the change in discharge current based on the number of neutral molecules available. At higher concentration values, the mean free path determines the iodization rate. Due

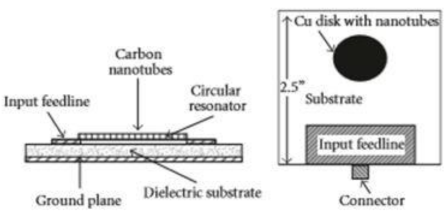

Fig. 11. The resonator circuit coated with nanotubes

to the short mean free path, the iodization rate decreases which results in the increase of breakdown voltages [14].

Despite having low consumption power and low break- down voltages, its applications are limited due to the requirement of high sensitivity signal processing devices.

D. Resonant frequency shift gas sensors

Scientist was in a search of new reliable and effective way to sense gas molecules other than the traditional ways which generally rely on the resistance measurements as the sensor output. The traditional methods were dependent upon the surrounding environment, temperature and they were also inadequate for the detection of gases with low adsorption energies or in the cases of inert gases. These limitations encouraged scientists to develop nanoscale gigahertz and ter- ahertz range devices for detection of gas atoms [15].

The working principle of this method is to detect the induced shift in resonant frequencies or the wave propagation velocity in nano sensors associated with attachments of gas atoms or molecules on the surface of the nano-sensor. The major drawback faced by this method is that such high-frequency vibrations in an order of gigahertz may be affected easily by environmental disturbances resulting in the non-precise measure- ment of frequency shifts in the nanosensors.

After exposing to gas molecules, the dielectric permeability of the substrate disk changes which causes a shift in its resonant frequency. Such precise measurements make them more selective and sensitive to a wide range of gases. The necessity of additional analyzing equipment poses as another drawback of this method.

7. CONCLUSION

The discovery of carbon nanotubes and the development in its related field has brought a revolution in the manufacturing and electronics industry. The extraordinary properties of CNT especially electrochemical properties has led the scientist community on a new way of designing sensor applications. From the literature survey done so far, we have found that electronic properties of CNT are governed by the surface structure of it or the chirality it posses. This dependence of CNTs properties on its structure and its interaction with the surrounding environmental factors made CNT a valuable active sensing element for sensors.

CNT gas sensors made using adsorption properties are common and easy to design and contribute to large part of CNT based gas sensors. This is associated with the change in resistance of CNT due to charge transfer between CNT and gas molecules or the change in carrier lifetime due to scattering. Though this kind of sensors is common, they are in adequate for inert gases or gases with low adsorption properties. Hence newer and technologically advanced methods for designing CNT based gas sensors were developed such as ionization based gas sensors or resonant frequency shift based gas sen- sors. These methods are effective and very efficient to detect a wide range of gases but they require additional equipment for measurement. Hence it is very difficult to commercialize them. Sensing properties of CNTs can be enhanced further by functionalizing them with various functional groups to enable them to detect a wide range of elements. These studies has shown that CNT as a sensor has very promising future in the field of biology, medicine, chemical, astronomy, etc.

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