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SYNTHESIS OF CARBON NANOTUBES FOR ACETYLENE DETECTION

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Abstract

A gas sensor, utilizing carbon nanotubes (CNTs) in a pellet form for acetylene detection has been developed. This research was carried out to investigate the absorption effect of acetylene (C2H2) towards the change of resistance of carbon nanotubes pellet as sensor signal. Source Measurement Unit (SMU) was used to study the gas sensing behaviour of resistance based sensors employing carbon nanotubes pellet as the active sensing element. Studies revealed that the absorption of acetylene into the carbon nanotubes pellet resulting in increase in pellet resistance. The changes are attributed to p-type conductivity in semiconducting carbon nanotubes. Carbon nanotubes used in this research was synthesized by means of Floating Catalyst Chemical Vapor Deposition (FC-CVD) method. Benzene was used as a hydrocarbon source while ferrocene as a source of catalyst with Hydrogen and Argon as carrier and purge gas respectively. From the research, it was shown that carbon nanotubes show high sensitivity towards acetylene. The highest sensitivity recorded was 1.21, 1.16 and 17.86 for S1, S2 and S3 respectively. It is expected that many applications of CNT-based sensors will be explored in future as the interest of the nanotechnology research in this field increases.

Keywords: Carbon Nanotubes, Acetylene Sensor.

Nomenclatures

R_{argon}	Resistance of sample in argon gas
R_{gas}	Resistance of sample in tested gas
S	Sensitivity

1. Introduction

CNTs are new carbon materials discovered recently. They are cylindrical carbon molecules with properties such as low density, high tensile strength and elastic modulus. Metallic carbon nanotubes have a high electric current density, and all of these properties make carbon nanotubes potentially useful in extremely small-scale electronic and mechanical applications. There are two main types of carbon nanotubes; single walled carbon nanotubes (SWNTs) and multi walled carbon nanotubes (MWNTs). SWNTs can be considered as a sheet of graphene that has been rolled up into a seamless cylinder, while MWNTs consist of nested coaxial arrays of SWNT constituents. Their structures are unique, only a few nanometers in diameter, but up to hundreds of microns long [1, 2, 3].

Carbon nanotubes possess very unique characteristics due to their hollow center, nanometer size and large surface area, and are able to change their electrical resistance drastically when exposed to alkalis, halogens and other gases at room temperature. Hence, carbon nanotubes have the potential to be a better chemical sensor [4].

2. Experimental Details

The carbon nanotubes used in this research is produced by means of floating catalyst Chemical Vapour Deposition (FC-CVD) method. Benzene was used as a hydrocarbon source while ferrocene as a source of catalyst with Hydrogen and Argon as carrier and purge gas respectively. The reaction temperature is in the range of 850°C-750°C, while the reaction time is 30 minutes.

A simplified schematic diagram for the apparatus used for carbon nanotubes production is shown in Fig.1

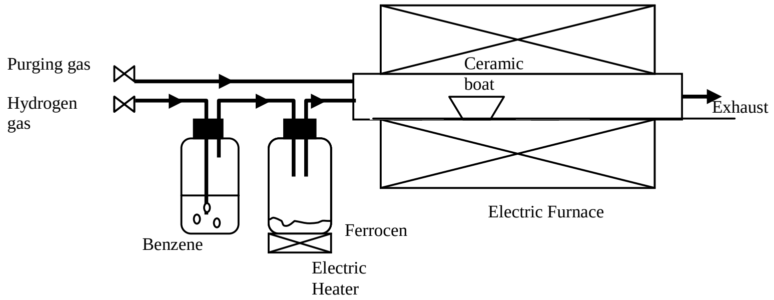

Fig. 1. Schematic of Chemical Vapour Deposition System.

The study is carried out to investigate the potential application of carbon nanotubes as a gas sensor by measuring the change of electrical resistance of the carbon nanotubes upon acetylene (C_2H_2) absorption.

2.1 Sample preparation

100mg of carbon nanotubes powder was mixed with a non-conductive binder (ethyl cellulose) to form a thick paste. The binder was prepared by dissolving one gram of ethyl cellulose powder in ethyl alcohol to produce a viscous liquid. The mixture of carbon nanotubes and binder was then placed in a rectangular mould (5 mm width, 15 mm length and 1.5 mm thick) and left for about 1 hour at room temperature to dry and solidify. Sample of this pellet is shown in Fig. 2.

Fig. 2. Carbon Nanotubes Pellets with Binder.

2.2 Gas sensor setup

Three sets of carbon nanotubes pellets with binder were made for gas sensing application. Pellet S1,S2 and S3 were made using bulk carbon nanotubes produced at temperatures of 850°C, 800°C and 750°C respectively. Silver paint was used to create contact at both surfaces of the pellets. Four strips of copper wires were soldered onto the silver paint on the pellet surface.

The wires from the pellet were then connected to the Source Measurement Unit, SMU (Keithley 4200 SCS, Department of Electrical and Electronic Engineering, UPM) used to obtain I-V (current-voltage) curve and to extract the value of resistance, R and conductivity, G, of the samples. The range of current source used was 0 – 50mA.

The pellet was placed in a sealed conical flask with electrical feedthrough. Carrier gas, Argon, was fed continuously into the flask. The resistances of carbon nanotubes were recorded every ten seconds interval until the system reached steady state, i.e. when the resistance of the carbon nanotubes gave a constant reading. Then, the tested gas was injected periodically for duration of three seconds into the flask. The resistance of the carbon nanotubes was recorded before and after the injection at the interval of ten seconds. The interval time was chosen based on the time taken for the carbon nanotubes' resistance to reset to the original reading. All the measurements were at room temperature of 25°C. The tests were repeated three times for samples S1, S2 and S3 in order to get the precise results.

3. Results and Discussion

3.1 SEM and HRTEM characterization

The scanning electron microscope (SEM) and High Resolution Transmission Electron Microscope (HRTEM) are used to characterize the carbon nanotubes produced.

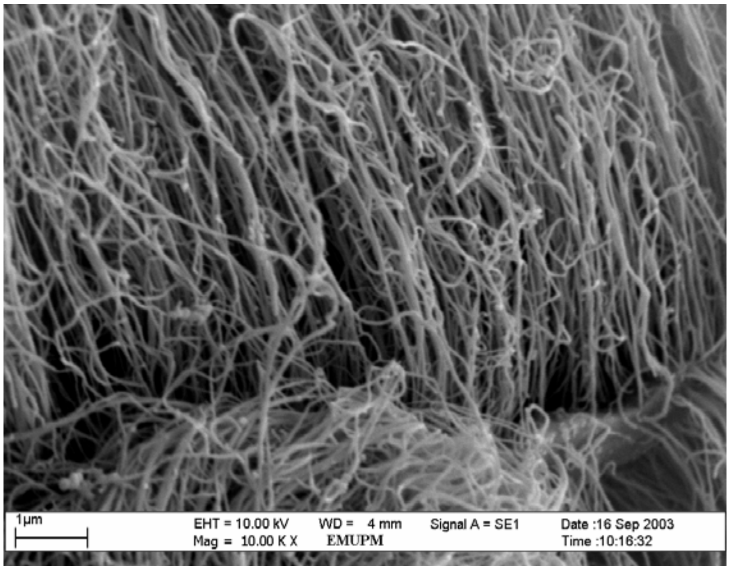

Fig. 3. SEM Micrograph of Carbon Nanotubes.

SEM micrographs have a characteristic three-dimensional appearance and are useful for judging the surface structure of the sample. In Fig. 3, the SEM observation shows that the carbon nanotubes are of high purity with tens of microns long and rather uniform diameters. The bulk morphology of the long carbon nanotubes is film like, randomly oriented and some of them are entangled. From the HRTEM micrograph in Fig. 4, the carbon nanotube is multiwalled with diameter of 43.57nm and wall thickness of 9.45nm.

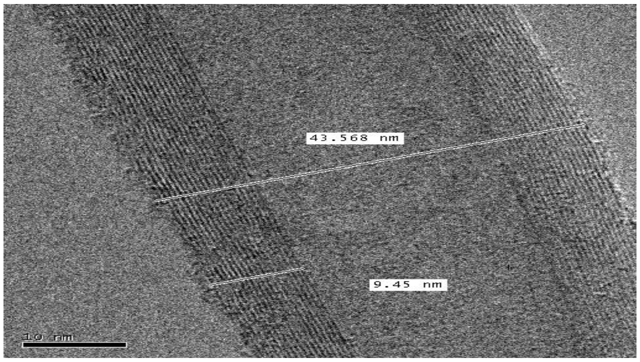

Fig. 4. HRTEM Micrograph of Carbon Nanotubes.

3.2 Acetylene absorption

Having loosely held pi-electrons, acetylene is a reducing gas and will readily donate electrons to the carbon nanotubes sample. Absorption of the gas into the sample results in increasing the electrical resistance of carbon nanotubes. In Fig. 5 it can be seen that the resistance of carbon nanotubes sample S1 shoots from 507 milliohm to near 513 milliohm at the first injection of acetylene. Consecutively the same increment can be observed for the second, third and fourth injection. The tests were repeated three times for every each of the carbon nanotubes samples, indicated by R1, R2 and R3.

Similarly for sample S2, immediate response can be observed upon injection of the acetylene gas at time 30, 80, 130 and 180 second as presented in Fig. 6. This indicates that the sensor is sensitive towards acetylene absorption. The duration of injection is three seconds and it can be observed that the resistance of the sample immediately subsides after every injection

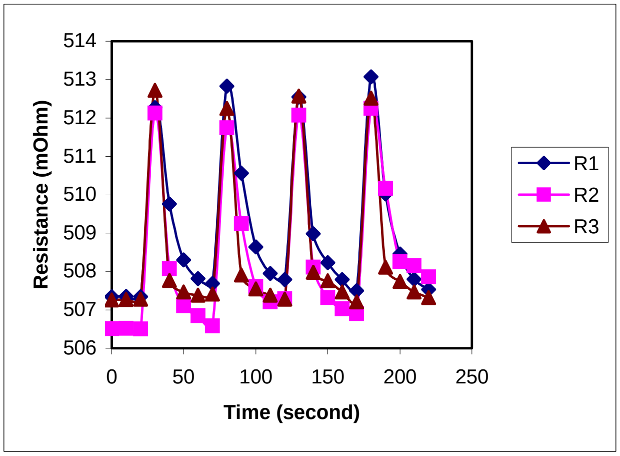

Fig. 5. Electrical Resistance Variations of S1 upon Injection of C_2H_2 Gas.

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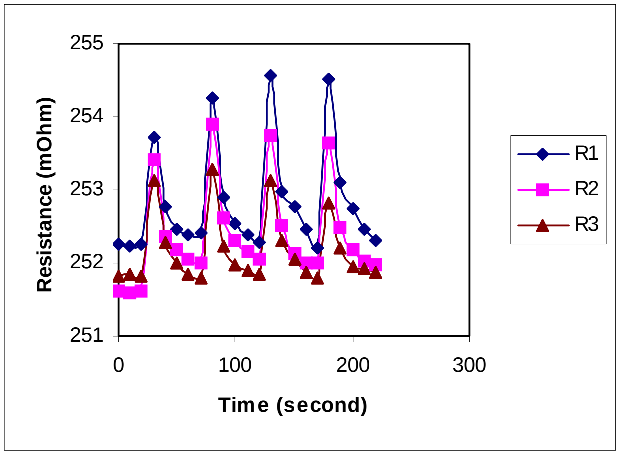

Fig. 6. Electrical Resistance Variations of S2 upon Injection of C_2H_2 Gas.

Figure 7 shows the response of sample S3 upon absorption of acetylene gas. The pattern is comparable with sample S1 and S2, resulting in a peak after each successive injections.

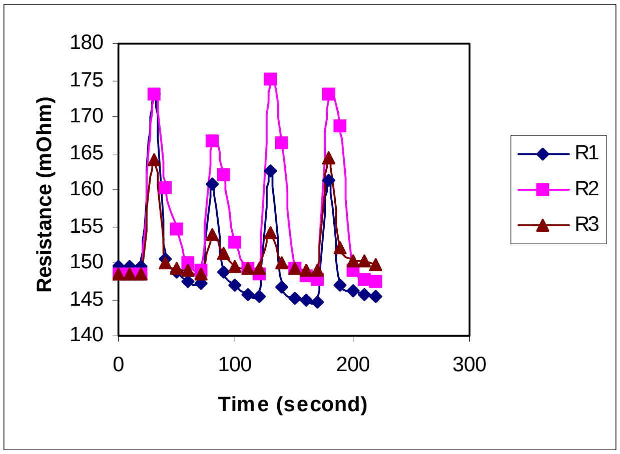

Fig. 7. Electrical Resistance Variations of S3 upon Injection of C_2H_2 Gas.

Sensitivity of the carbon nanotubes sample is estimated using the equation 1 [8]:

$$S = \frac{R_{gas} - R_{arg\,on}}{R_{arg\,on}} \times 100 \tag{1}$$

Sensitivity of S1, S2 and S3 are summarised in Table 1. The highest sensitivity recorded was 1.21, 1.16 and 17.86 for S1, S2 and S3 respectively.

Table 1. Sensitivity of S1, S2 and S3 in Acetylene.

Time	S1			S2			S3		
	R1	R2	R3	R1	R2	R3	R1	R2	R3
30	1.05	1.03	1.14	0.81	0.70	0.58	16.52	16.51	10.47
80	1.18	0.96	1.06	1.05	0.91	0.66	8.23	12.19	3.52
130	1.11	1.02	1.12	1.16	0.83	0.59	9.47	17.86	3.72
180	1.21	1.05	1.10	1.13	0.78	0.45	8.51	16.56	10.55

A review of the available literature revealed that there is no work reported in acetylene sensor using carbon nanotubes. However, looking at the chemistry of the acetylene molecule, it can be predicted that the response will be in a similar way with any reducing gases. Acetylene has excess electrons and prone to act as an electron-donating agent. Donation of electrons to a p-type semiconductor will result in holes depletion and consequently give rise to the resistance.

4. Conclusions

The aim of this study was to investigate the electronic sensor application of carbon nanotubes upon absorption of acetylene. Based on the experimental results, it was proven that the carbon nanotubes have the capability to detect acetylene at room temperature. Therefore, it can be concluded that the gas sensing characteristics carried out in this work has shown that carbon nanotubes have potential to be an excellent acetylene sensor material at room temperature.

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