31st Annual International Conference of the IEEE EMBS
Minneapolis, Minnesota, USA, September 2-6, 2009

A Carbon Nanotube Gas Sensor Fabricated by Dielectrophoresis and Its Application for NH_3 Detection

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Abstract—**Multi-walled carbon nanotubes (MWNTs) were successfully manipulated by dielectrophoresis (DEP) to form electrical connection between interdigitated gold electrodes (IDEs) and were demonstrated to serve as gas sensor for NH_3 detection. The MWNTs were suspended in ethanol and deposited on the IDEs under the effect of DEP. After the evaporation of ethanol, the MWNTs remained between the gaps of the IDEs. The electrical conductivity of the DEP-fabricated MWNTs sensor decreased when exposed to NH_3 at room temperature. There is a good linear correlation between the decreasing amplitude of conductance and the NH_3 concentration, and the detection limit of 10ppm NH_3 could be achieved.**

I. Introduction

The discovery of carbon nanotubes (CNTs) in 1991 by Ijima [1] has generated great interest among researchers to explore their unique electrical, physical, mechanical and chemical properties to develop high performance devices. CNTs-based gas sensors have received considerable attention because of their outstanding properties, such as faster response, higher sensitivity, lower working temperature, and a wider variety of detectable gas than other types of gas sensors [2-10]. Application of CNTs-based gas sensors in NH_3 detection have been already reported on both single-walled carbon nanotubes (SWNTs) [2-5] and multi-walled carbon nanotubes (MWNTs) [6-10].

Currently, there are mainly two methods to position CNTs on metal electrodes [11]. One is directly growing CNTs on the sensor platform via controlled chemical vapor deposition (CVD), this fabrication method is complex with low yield. The other is drop-casting CNTs suspension or solution in water or in organic solvent on top of prefabricated electrodes by micropipettor, followed by a drying process, this method is simple but sometimes CNTs can't be distributed symmetrically on the electrodes. Inexpensive, high yield and reproducible fabrication techniques are essential for the success of CNTs-based sensors. In this paper, we use dielectrophoresis (DEP) to fabricate CNTs-based gas sensor. DEP is the electrokinetic motion of dielectrical polarized materials in non-uniform electric fields and has been successfully applied to manipulation of nanomaterials including CNTs, metallic nanoparticles and semiconducting nanowires [12]. Using DEP technique can precisely quantify the amount of trapped nanomaterials by monitoring the amplitude, frequency of the AC voltage and the deposition time [13]. DEP ensured a uniform distribution of the deposited CNTs on the interdigitated gold electrodes (IDEs).

II. Experimental Details

A. Fabrication of Gas Sensor

The MWNTs used were obtained from Shenzhen Nanotech Port Co.Ltd. They were grown by the Chemical Vapor Depositon(CVD) method, had a purity higher than 98% ,were 1 to 2 μm in length and their diameter ranged from 10 to 20nm.

The MWNTs were suspended in ethanol (1mg/ml final concentration), and some $Al(NO_3)_3$ was added to the solution as supporting electrolyte, then the solution was ultrasonicated for 2h. The system of fabricating the MWNTs-based gas sensor using DEP is depicted in Fig.1. The DEP trapping of MWNTs on the IDEs was performed with an AC voltage of 5MHz frequency and 10V amplitude (peak-to-peak value). The IDEs has a ceramic substrate, where an interdigitated array of gold tracks had been previously evaporated and photolithographically defined (the distance and width of tracks were both 40 μ m). After 1min, the DEP process was stopped and the ethanol was evaporated, the MWNTs remained on the IDEs.

Manuscript received April 1, 2009. This work was supported in part by the National Science & Technology Pillar Program in the Eleventh Five-year Plan Period of China (No. 2006BAD30B03) and Science & Technology Program of Zhejiang Province (2008C23081)

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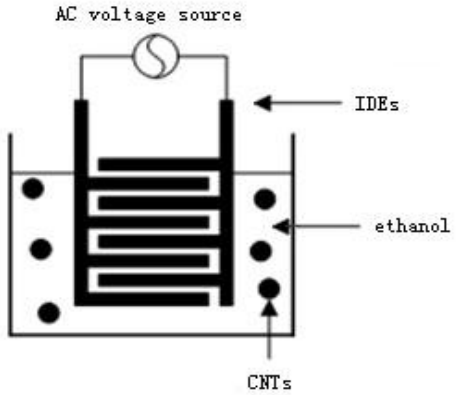

Fig. 1. The system of fabricating the MWNTs-based gas sensor using DEP.

B. Measurement of the Sensing Characteristics

Fig.2 shows the experimental device used to characterize the sensing properties of MWNTs –based gas sensor fabricated by DEP, located in a sealed test chamber (500 ml) with an inlet valve and an outlet valve, and connected with an Electrochemical Analytical Instrument (CHI660).N_2 was used as reference gas and carrier gas. Saturated vapor of NH_3 was diluted by dry N_2 to prepare tested NH_3 of 10, 20, 30, 40, and 50 ppm.

Inflate dry N_2 into the testing chamber for 5 min to expel the air inside. Close both the inlet valve and outlet valve, and then run the CHI electrochemical analytical program using "i-t" method. Inject the tested NH_3 respectively when the output current-time curve responses stable. Open the inlet valve and outlet valve when the output current-time curve returns stable. Inflate dry N_2 into the test chamber to expel NH_3 inside. All the sensing measurements of the sensor were carried out at room temperature and atmospheric pressure.

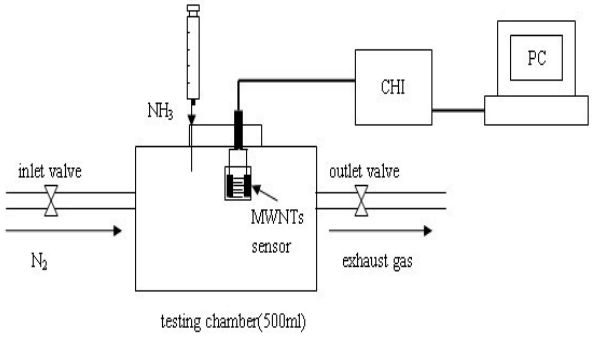

Fig. 2. Experimental setup used to characterize the sensing properties of MWNTs-based gas sensor.

III. RESULTS

A. DEP Fabrication

The surface morphology of MWNTs on the IDEs was examined using a SIRION-100 field emission scanning electron microscopy (SEM, America). Fig.3 is the SEM image of IDEs without MWNTs. Fig.4 shows SEM images of MWNTs sensors fabricated by DEP. The SEM observation revealed that MWNTs were trapped in the electrodes gap to make connection with the IDEs, and the MWNTs formed entangled networks rather than individual trapped.

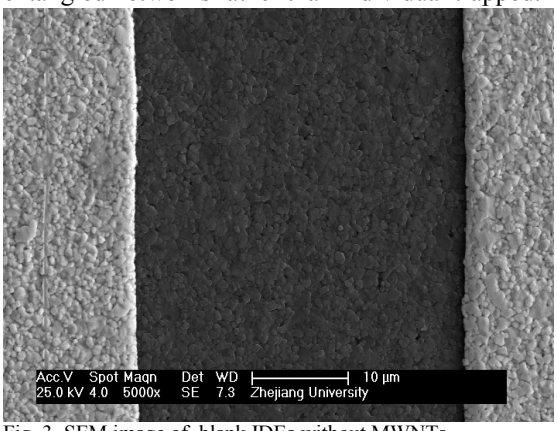

Fig. 3. SEM image of blank IDEs without MWNTs.

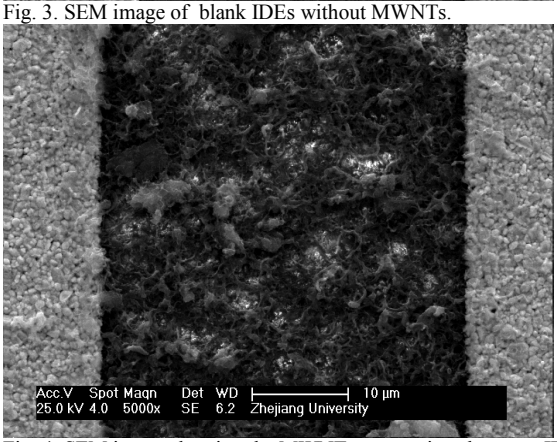

Fig. 4. SEM image showing the MWNTs connections between IDEs.

B. NH_3 Sensing

The successive dynamic responses of MWNTs-based gas sensor exposure to NH_3 with the concentrations from 10ppm to 50ppm at room temperature are shown in Fig.5. The fall of current in the curves indicates the falling of the conducting ability of the sensor. The sensitivity (S) of the sensor is defined as: $S=(R-R_0)/R_0*100\%$ where R_0 is the resistance in the presence of N_2, and R is the resistance after NH_3.Since the working voltage is fixed at 0.1V, the sensitivity could be simplified to: $S=(I_0-I)/I*100\%$.

It can be observed that the sensing responses increase with the increment of NH_3 concentrations and the sensor exhibits a fast response time of 1~3 minutes (defined as the time for 80% conductance change to take place).The detection limit of 10ppm NH_3 could be achieved. However, the baseline of current in Fig.5 drifts downwards, indicating that the conductance of the sensor cannot return to the initial value after purging with N_2 with only a partial recovery. Fig.6 displays a good linear correlation between the sensitivity of the MNWTs gas sensor fabricated by DEP and the NH_3 concentration at room temperature.

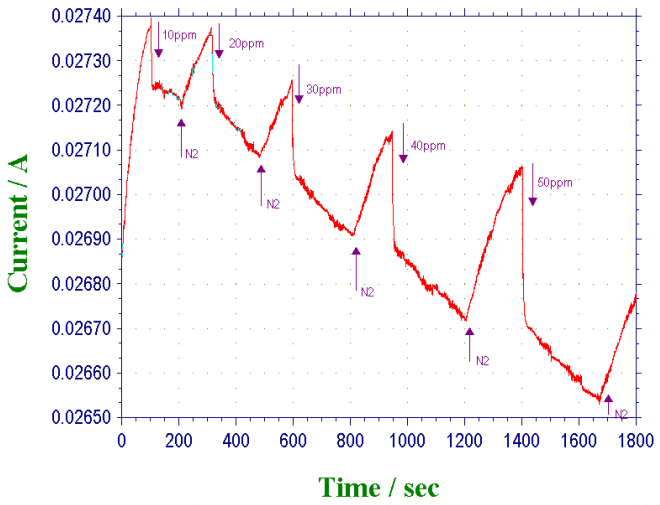

Fig.5. The successive dynamic responses of the sensor exposure to NH_3 with the concentrations from 10ppm to 50ppm at room temperature

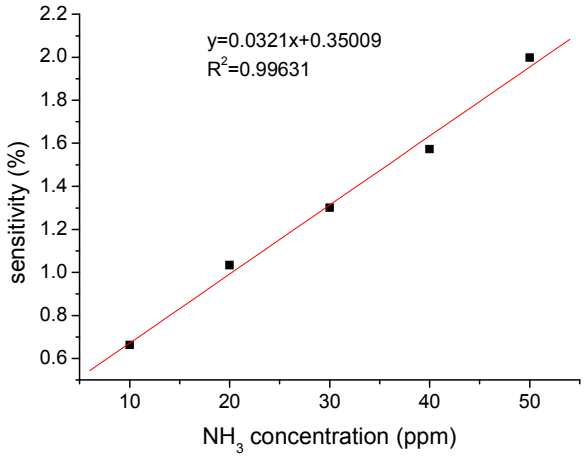

Fig. 6. The linear correlation between the sensitivity of the sensor and the NH_3 concentration at room temperature.

IV. Discussion

As shown in Fig.4, the MWNTs were manipulated by DEP and uniformly adhered to the gap of IDEs. The DEP-trapped MWNTs formed a sensing membrane, which had the network structure with large surface area and hollow tubule. When the MWNTs sensing membrane exposed to NH_3, the NH_3 molecule could be adsorbed into the surface of MWNTs and make the electrical conductivity of MWNTs decrease. The mechanism by which the conductance decreases hasn't came to a general theory. Kong [2] demonstrated that exposure to NH_3 effectively shifts the valence band of CNTs away from the Fermi level, resulting in hole depletion and reduced conductance. Wang [7] held the point that the adsorption of NH_3 could enhance the interlayer interaction of nanotubes, which may cause the MWNTs change from metallic to semiconducting , resulting in the decrease of conductance.

It can be observed from Fig.5 that NH_3 could not be completely desorbed from MWNTs and the conductance of the sensor was partly recovered after purging N_2. The binding energy of NH_3 and MWNTs is so large that the recovery process usually takes a long time at room temperature, ranged from several minutes to a few hours [14]. To accelerate the recovery process of CNT sensors, some researchers have attempted to lower the adsorption energy barrier by implementing external energy, such as heating the CNTs to high temperatures (~200 ℃), or using ultraviolet light illumination[11]. Espinosa [15] reported SnO_2/MWNTs-based sensors showed faster recovery at 150 ℃ than at room temperature. Qi [3], Suehiro [12] used the UV light to illuminate the CNTs-based sensor recover and made a good recovery.

V. Conclusion

In this paper, a MWNTs-based sensor has been fabricated by DEP and applied for NH_3 detection at room temperature. Performed with an AC voltage of 5MHz frequency and 10V amplitude (peak-to-peak value) for 1min, the MWNTs were uniformly trapped in the gap of IDEs and formed a sensing membrane. The DEP-fabricated MWNTs sensor exhibited fast response to NH_3 at room temperature, and the detection limit of 10ppm NH_3 could be achieved. DEP may be a promising technique for the application of fabricating CNTs-based sensors.

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