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Research paper

Effect of molecular coverage on the electric conductance of a multi-walled carbon nanotube thin film

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ABSTRACT

We investigated the influence of water adsorption on a CNT thin film. When we assumed that the magnitude of the change in electrical resistance was correlated with the surface coverage of the adsorbed molecules, this phenomenon could be explained by two-layer adsorption. The first layer was expressed by Langmuir adsorption and that on the second layer was expressed by Fowler–Guggenheim adsorption, which was derived by Bragg–Williams approximation and involved a lateral molecular interaction. The adsorption energy estimated by this assumption was on the same order as derived by DFT calculation.

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1. Introduction

The common use of carbon nanotubes [1,2] (CNTs) in gas sensors is underwhelming in comparison to their use in other applications [3–9]. The most popular type of gas sensor is the semiconductor-type gas sensor, which has a high sensitivity but requires heating to enhance the responsivity and uses a considerable amount of energy. CNTs have a large specific surface area and show practical changes against gas adsorption, even at room temperature; therefore, a lot of researches have been studied so far such as NO_2 [10,11], NH_3 [12–14], CO_2 [15,16], H_2 [17,18], and others [19–34]. Kong et al. [35] and Collins et al. [36] performed much of the frontier work using isolated or thin bundles of single-walled CNTs (SWCNTs) as a sensor component. Many other researchers preferred and employed isolated SWCNTs to analyze the detection mechanism, and proposed Schottky barrier [37] and charge transfer [38] models, depending on the conditions. The use of isolated SWCNTs simplifies this phenomenon and is useful for a basic understanding of interaction between surface and adsorbed molecules, and analysis of detection mechanism. However, advanced technologies and high initial expenditure will be required for practical applications. In a previous paper, we demonstrated a trial for a simple gas sensor made from as-grown CNTs and showed their potential for practical use. As for a film of CNTs, there is no consensus on the detection mechanism, which is expected to be complicated. To understand the surface interaction between the CNTs and gas molecules, we could study the interaction with a variety of gas molecules; however, we cannot ignore the influence of humidity (water molecules) on the gas sensor for practical use. Therefore, we focused on the surface interaction for absorbed water vapor. We can explain this interaction using a simple adsorption model in terms of the sensor response, which is defined as the electric conductance change with respect to the gas pressure. Furthermore, we showed that the adsorption energy estimated from an adsorption equilibrium constant was similar to that from a previous report. In addition, to understand the electronic states of an adsorbed molecule qualitatively, we calculated the charge transfer between the water molecule and graphene by using the Gaussian09 software package with the HF/6-31G** method. Here, graphite was used instead of the multi-walled carbon nanotubes (MWCNT) for simplicity.

2. Experimental

We prepared a MWCNT film on a quartz substrate by spin coating. Before spin coating, the surface of the substrate was reformed by an argon plasma to remove contamination and enhance the wettability. A solution of MWCNTs (3 wt%, Ohashi Kasuga Tsusho Inc., CF163CP), which was dispersed in distilled water, was employed and the 56 μL solution was added to a burette. The solution was dropped onto the substrate, which was rotating at 4000 rpm for 200 s. A cross sectional view and the surface morphology of the MWCNT film were observed by scanning electron microscopy (Fig. 1). The profile of the MWCNT was uniform and an intertwisted structure was observed. The transmission line method (TLM) [39,40] was used to measure the contact resistance and sheet resistance. The substrate was set in the TLM holder, and its schematics is shown in Fig. 2. By scanning the transmission length of the current, we derived each resistance as:

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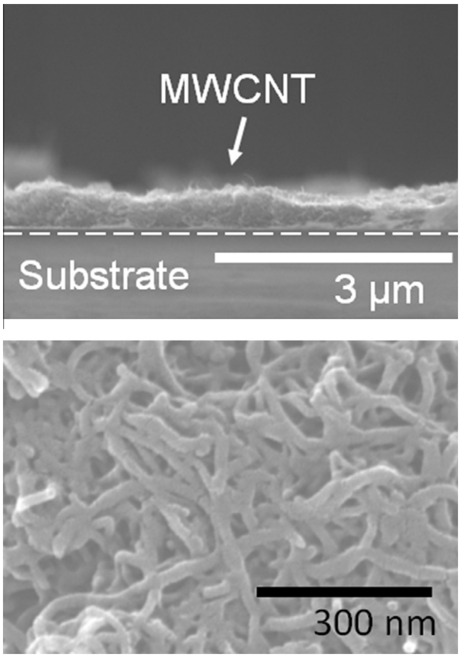

Fig. 1. Cross-sectional view and surface morphology of the MWCNT thin film fabricated by a spin coating method. Although there are some clumps of CNTs locally, the morphology is uniform overall based on our results explained by Fig. 4.

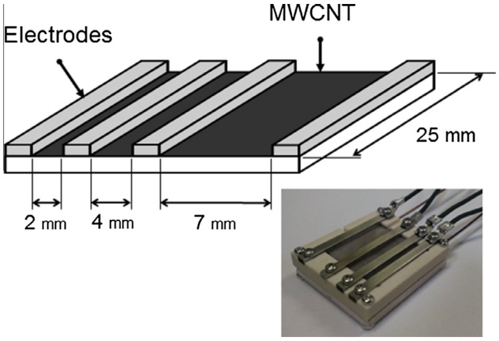

Fig. 2. Schematic of the measurement system for the transmission line method (TLM).

$$R_T = 2R_c + \frac{R_{sh}}{W} d, \tag{1}$$

where R_T is the total resistance, R_C is the contact resistance, R_{sh} is the sheet resistance, W is the width of the electrodes, and d is the electrode distance. Here, we must assume that the contact resistance and the sheet resistance were constant, but as shown later, the resistance of the transmission length exhibited a linear relation to the electrode distance, so that the MWCNT film was homogeneous. To investigate the interaction between the MWCNTs and a water molecule, the MWCNT film was set in the measurement section that is shown in Fig. 3. The vacuum chamber was evacuated using a rotary vacuum pump and the pressure was measured using a capacitance manometer. The electrical resistance was measured by a DC power supply, which applied a constant voltage to the electrode and measured the current. Before the measurement, the chamber was evacuated to less than 1 Pa and left for typically 30 min; water vapor was then introduced using a mass flow controller.

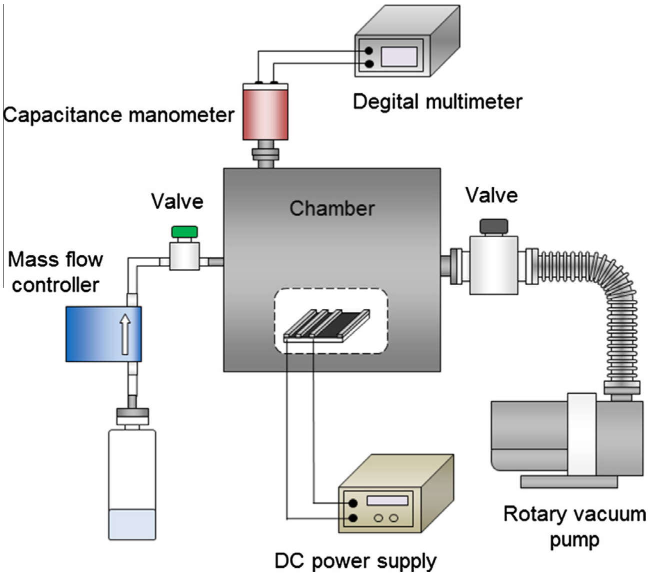

Fig. 3. Schematic of the experimental setup.

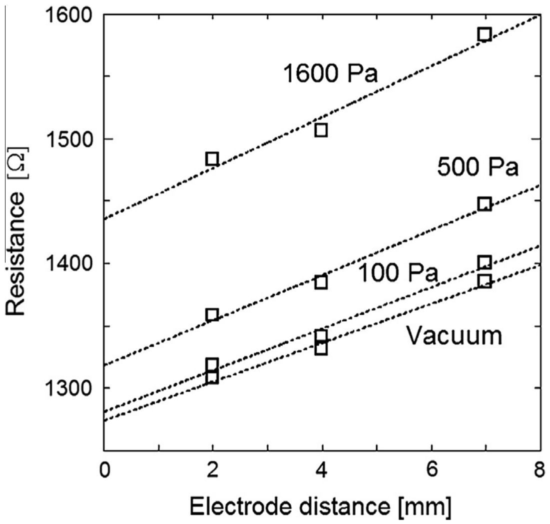

Fig. 4. Total electrical resistances with respect to the electrode distance and vapor pressure. The MWCNT thin film could be considered uniform because the relationship between the sheet resistance and distance showed a linear correlation. The *y*-intercept indicates the contact resistance, which increases with the pressure. The sheet resistance (shown by the slope of the line) also increases with pressure. Sheet resistances derived from these slopes are 390.3, 416.5, 451.3, and 511.3 Ω/sq. for vacuum (0.8 Pa), 100, 500, and 1600 Pa, respectively.

3. Results and discussion

Fig. 4 shows the relationship between the electrode distance and total resistance. Each data point is the average of three values. The maximum error in this experiment was less than 0.2%, so we omitted error bars. As for the contact resistance (distance = 0), as mentioned in previous studies. However, when the thickness of the CNT film was more than 120 nm or for a high-density CNT network, the contact resistance was hardly affected by the gas adsorption. It was inferred that a gas molecule can hardly reach the contact surface and it does not affect the resistance change. Referring to Fig. 1, the thickness was more than 400 nm, and the network density seems to be high; therefore, our result is different

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