

Sensing Property of Horizontally Aligned Carbon Nanotube Field-Effect Transistor on Quartz Substrate

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1. Introduction

In recent years, highly sensitive and label-free biosensors are required in various fields such as early diagnosis, home care, and practical pharmacy. Carbon nanotube field-effect transistors (CNTFETs) are one of the most promising platforms for electronic bio-detection owing to their unique electrical and physical properties. The detection of biomolecules such as DNA and proteins using the CNTFETs has been successfully performed [1, 2]. However, high bias can not be applied between electrodes because of the oxidation-reduction reaction of biomolecules in solutions. Hence, the current that flows in one CNT channel is less than tens of nA, and it is comparable to the noise level. It is the reason why the current changes are small when biomolecules are adsorbed onto the CNT channels. The use of multiple nanotube channels in a FET offers several advantages such as higher uniformity, lower noise, and higher reproducibility [3].

In this abstract, horizontally-aligned CNTs were grown on quartz substrates and CNTFETs with multi CNT channels were fabricated. Aligned arrays of CNTs increase the drain current (I_D) of CNTFETs. Therefore, the performance of CNTFET based biosensors will be improved. In this study, we carried out solution pH sensing and label-free immunosensing based on CNTFETs.

2. Experimental Procedure

Quartz substrates were annealed at 900°C for 8 hours in air to re-crystallize the surfaces. 0.5-nm-thick Fe catalysts were patterned by conventional photolithography and liftoff process. CNTs were synthesized by alcohol CVD. Ethanol as a carbon source gas was introduced for 20 min at 900 °C. Then, source and drain electrodes of Ti (2 nm)/Au (30 nm) were formed. To measure pH value and protein adsorption in solution, the device was surrounded by a silicone rubber barrier attached to the substrate. Figure 1 shows a schematic image of the fabricated CNTFET based sensor on quartz substrate. A Ag/AgCl reference electrode was used as a top-gate electrode to minimize the effects of environment.

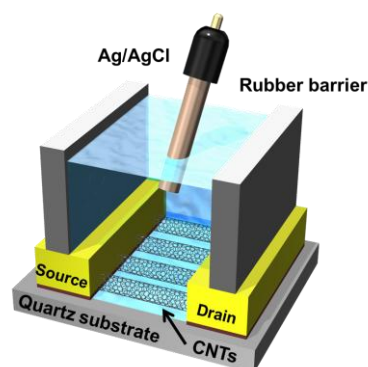


Fig. 1. Schematic illustration of CNTFET-based sensor.

3. Results and Discussion

Figures 2 (a) and (b) show scanning electron microscopy (SEM) images of CNTs grown on Si/SiO₂ and quartz substrates, respectively. The random networks of CNTs were observed on the Si/SiO₂ substrate (Fig. 2 (a)). In contrast, aligned CNTs were observed on the quartz substrate (Fig. 2 (b)). It has been reported that CNTs are aligned along the specific crystalline directions of quartz substrate [4]. This alignment enables fabricating highly density CNT arrays between electrodes and minimizes the tube-to-tube contact resistance associated with network devices. Therefore, it is expected that the performance of the CNTFET is improved using aligned CNTs grown on quartz substrate.

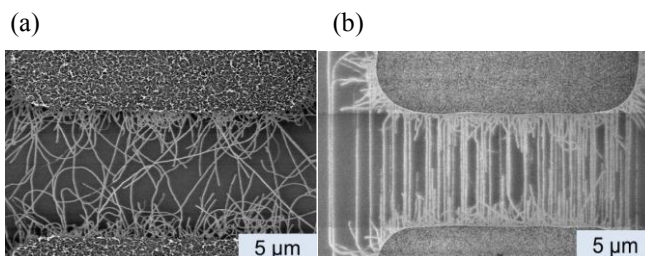


Fig. 2. SEM images of CNTs grown on Si/SiO₂ substrate (a) and quartz substrate (b).

Subsequently, the aligned CNTs were applied to CNT-FET-based electrolyte-gated sensors. First, the change in pH in buffer solution was measured. Figure 3 (a) shows a kinetic measurement to observe a real-time response recorded by tracking I_D at fixed top-gate voltage (V_{TG}) and source-drain voltage (V_D). Every 5 min, the pH of the buffer solution was changed from 4.0 to 8.3. The drain current clearly showed stepwise increases. Figure 3 (b) shows the I_D plotted as a function of pH value. This result indicates that the relationship between pH and I_D is linear over the range from 4.0 to 8.3. The detection limit (resolution, signal/noise=3) for changes in pH was estimated to be 0.015. This is superior characteristics than that of CNTFET with one CNT channel which showed the detection limit of 0.67 owing to the small I_D in solution (~ 10 nA) [5]. On the other hand, in our device, large changes in I_D were observed against pH changes in solution because highly density CNT arrays improved the transconductance in CNTFETs.

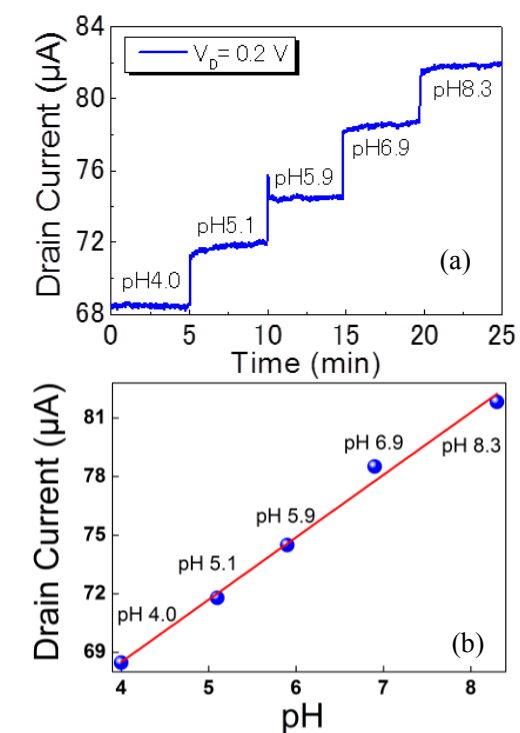


Fig. 3. (a) Time dependence of I_D with various pH values at V_D of 0.2 V and V_{TG} of -0.15 V. (b) I_D as a function of pH. A red dashed line corresponds to a linear fit to the data points.

Finally, we demonstrated label-free immunosensing based on aptamer-modified CNTFETs with multi CNT channels. The target protein is immunoglobulin E (IgE), which plays an important role in allergy. To enable IgE sensing, IgE aptamers were immobilized on the CNT channels as described previously [1]. Then, the electrical properties of the CNTFET during introduction of the target IgE were measured. To measure IgE concentration dependence, target IgE at concentrations of 28, 222, and 351 nM was introduced into the CNTFET while I_D was moni-

tored in real time (Fig. 4). I_D decreased stepwise after injection of the target IgE at each concentration. It is considered that the positively charged IgE molecules were connected to the negatively charged IgE aptamers, resulting in the decreases in I_D . The result indicates that we have demonstrated label-free immune sensing using aptamer-modified CNTFETs with aligned CNTs on quartz substrates.

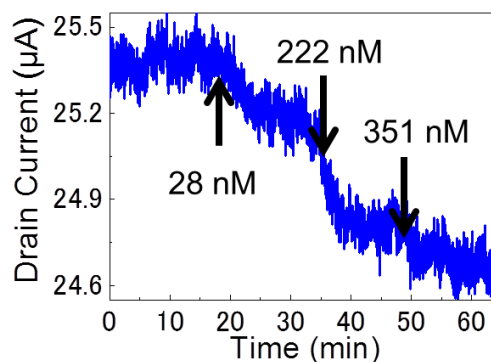


Fig. 4. Time dependence of I_D at V_D of 0.2 V and at V_{TG} of -0.07V after introduction of target IgE at various concentrations onto the IgE aptamer-modified CNTFET.

4. Conclusions

In summary, we demonstrated a simple and high performance biosensing platform using horizontally well-aligned CNTs grown on quartz substrates. Changes in the solution pH were electrically detected with a lower detection limit of 0.015. Besides, detection of nM quantities of IgE was accomplished. These results indicate that aligned CNTFET based biosensors will provide the production of sensitive label-free electronic biosensors to detect clinically important biomarkers for disease diagnosis.

Acknowledgements

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References

- [1] K. Maehashi, T. Katsura, K. Kerman, Y. Takamura, K. Matsumoto, and E. Tamiya: *Anal. Chem.* **79** (2007) 782.
- [2] K. Maehashi, K. Matsumoto, K. Kerman, and E. Tamiya: *Jpn. J. Appl. Phys.* **43** (2004) 1558.
- [3] E. S. Snow, F. K. Perkins, and J. A. Robinson: *Chem. Soc. Rev.* **35** (2006) 790.
- [4] C. Kokabas, S. Hurm A. Gaur, M. A. Meitl, M. Shim, and J. A. Rogers: *small* **1** (2005) 1110.
- [5] Y. Yamamoto, Y. Ohno, K. Maehashi, and K. Matsumoto: *Jpn. J. Appl. Phys.* **48** (2009) 06FJ01.