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Single-Walled-Carbon-Nanotube-Based Field-Effect Transistors with Biosensing Functions for Prostate-Specific-Antigen

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Abstract

Single-walled carbon nanotube (SWCNT) field-effect transistors (FETs) were fabricated on strontium titanate ($SrTiO_3$) substrate through a wet-process by using amide-functionalized SWCNT. The SWCNT-FET exhibited good gate-modulation for drain current at low operating voltages (-3 V). The hole mobility was 0.19 cm^2/Vs with an on/off current ratio of 1.3. After immobilization of prostate-specific-antigen (PSA) antibody the SWCNT-FET clearly responded against the PSA. The drain current at -3 V of both drain and gate voltage almost linearly increased with increasing the concentration of the PSA.

Keywords: Singe-walled carbon nanotube; Field-effect transistor; Prostate-specific-antigen

Introduction

Carbon nanotubes (CNTs) [1] have become attractive electronic materials to date. Single-walled carbon nanotube (SWCNT)-based field-effect transistors have been extensively studying since applications to future electric circuits [2,3] and bio-sensing chips [4] were proposed. Since the SWCNTs have high surface-to-volume ratios high sensitivities are expected in application to biosensors. Li, et al. proposed biosensing SWCNT-FET for detection of prostate-specific-antigen (PSA) [5] which is an oncological marker for the presence of prostate cancer. They, however, did not show well-defined FET characteristic and advantages of FET sensors compared with resistance ones, although high sensitivity (5 ng/ml) was obtained.

In this letter, we present the fabrication and well-defined output/transfer characteristics of SWCNT-FETs by a simple wet-processing technique and their application to bio-sensor for detection of PSA.

Experimental

Chemicals and materials

SWCNTs with amide functional side groups was purchased from Aldrich Co., Ltd. PSA (0.5~100 ng/ml) and mouse PSA-antibody (1.0 ng/ml) were purchased from Fujireio Inc. and Mikuri Laboratory Co., Ltd., respectively. $SrTiO_3$/Si wafers were provided by Tokyo Electron Ltd. The $SrTiO_3$ wafer is excellent dielectric insulator for low-voltage organic FETs [6,7].

Raman spectroscopic measurement of the SWCNT

The Raman spectroscopic measurement was conducted on the SWCNT with JASCO NRS-2100 spectroscopic equipment. Argon (488 nm) laser was used for measurement of G and D bands, while Green (532 nm) lasers for radial breathing mode (RBM). The calibration of wave number was carried out by using standard silicon.

Fabrication of SWCNT-based field-effect transistors

The amide-functionalized SWCNT [8] was dispersed in methanol (0.1 w% SWCNT) by sonication and stirring. Finally the SWCNT dispersion was filtered by a disc filter with pore size of 5 µm. The $SrTiO_3$/Si wafer was cut into 1.0 cm x 1.0 cm area. The substrate was cleaned by solvent (water and acetone)-washing and UV-ozoning treatment. Gold was thermally deposited in vacuum on the substrate for FET channel pattern. The channel length (100 µm) was controlled by using copper wire with a diameter of 100 µm as a shadow mask. Seven drops (14 µl) of the SWCNT dispersion were drop-casted on the channel-patterned $SrTiO_3$ substrate. The deposited SWCNTs on the FET channel were observed by a scanning electron microscope (SEM, S-4500 Hitachi). Output characteristics were measured with a semiconductor parameter analyzer (4200-SCS, Keithley) in atmosphere or in vacuum (6 x 10^{-1} Pa) and the FET mobility was calculated from the linear regime [9].

Biosensing measurements using the singe-walled carbon nanotube-based field-effect transistors

PSA-antibody (1.0 ng/ml) was dropped on the channel pattern of the SWCNT-FET device and incubated for 12 h. The channel pattern was washed with deionized water to remove free PSA-antibody, and then dried in vacuum for overnight. After confirmation of FET characteristics PSA with a certain concentration was dropped on the antibody-immobilized channel pattern and incubated for 15 h. After measurement of the FET characteristics the experiment repeatedly continued with a higher concentration. The sensing performance against the PSA concentration was evaluated by using the drain current at -3 V of drain voltage and at -3 V of gate voltage.

Results and Discussion

Performance of SWCNT-based field-effect transistors

Raman spectrum of the amide-functionalized SWCNT was shown in Figure 1. Clear and sharp G band of the SWCNT was observed at

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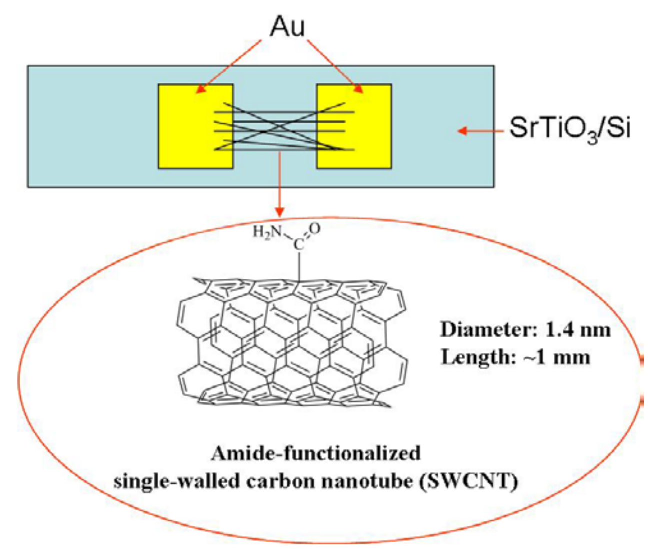
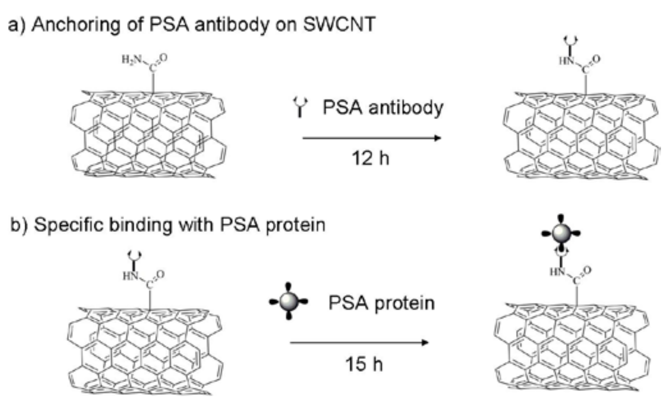


Scheme 1: Device structure of the SWCNT-FET (top) and chemical structure of the amide-functional SWCNT (bottom).



Scheme 2: Procedures of antibody immobilization on the SWCNTs (a) and PSA binding to the antibody on the SWCNTs (b).

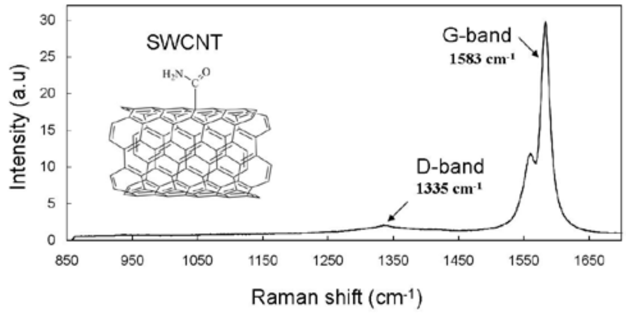


Figure 1: Raman spectrum of the amide-functional SWCNT.

1583 cm^{-1}, being agreement with value in literature [10]. On the other hand, very weak D band was observed at 1335 cm^{-1}, indicating high quality of the SWCENT with diameter of 1.4 nm which was determined by RBM of the Raman spectrum, 172.3 cm^{-1}.

The SWCNT was dispersed in methanol thanks to the hydrophilic amide functional groups. It is considered that the SWCNTs are physically adhered on surfaces of the $SrTiO_3$ and gold electrodes during the drop-and-dry process, as shown in Scheme 1. Figure 2 shows SEM image of the SWCNT-FET channel fabricated by the drop-and-dry process. The SEM image clearly indicated that the SWCNTs formed bundles with an average diameter of 13.4 nm, and the bundles strongly adhered on both $SrTiO_3$ and gold electrode surfaces. The output and transfer characteristics of the SWCNT-FET with a channel length of 100µm were shown in Figure 3. The FET showed good gate-modulation at low gate voltage of -3 V (Figure 3 left hand). We evaluated the performance of the FET using the dielectric constant and thickness values of the $SrTiO_3$ (ε_r = 12.1; d = 87 nm). In Figure 3

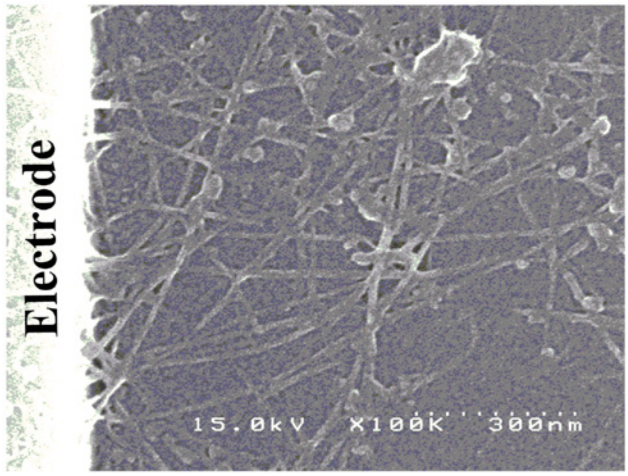


Figure 2: SEM image of SWCNT-FET channel fabricated by drop-and-dry process.

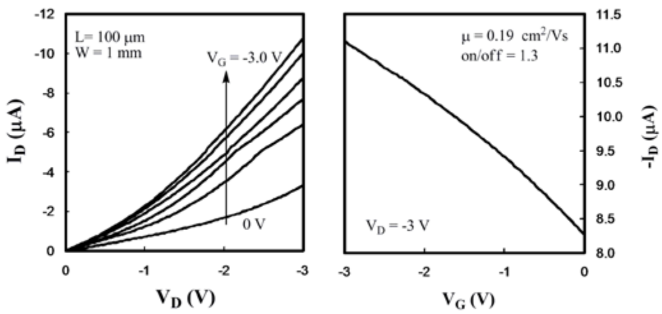


Figure 3: Output (left) and transfer (right) characteristics of the SWCNT-FET.

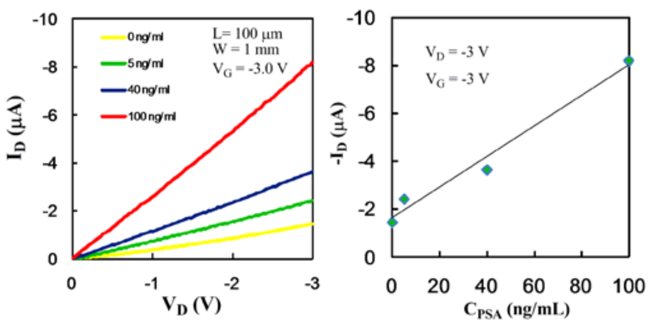


Figure 4: Output characteristics of the SWCNT-FET at a gate voltage of -3 V (left), and drain current of the SWCNT-FET at -3 V of drain and gate voltages (right) after treatment with the PSA of various concentration.

(right hand), the I_D evidently increased with increasing the V_G. The hole mobility can be calculated from the plot is 0.19 cm^2/Vs (linear regime) and on/off current ratio is 1.3 for the V_G between -3 and 0 V. Noting that the mobility is relatively higher among the devices using soluble SWCNTs [8].

PSA-sensing of SWCNT-based field-effect transistors

PSA-antibody-immobilization was carried out directly through the amide-functional groups covalently binding on the SWCNT, as shown in Scheme 2a. It is noted that this process is simpler than method reported by Li et al. [4] in which succinimidyl ester and pyrenyl terminal groups serves as anchoring the PSA-antibody and the SWCNET, respectively [4]. Binding reaction of PSA to the antibody is the same with the method reported by Li et al. [4] as shown in Scheme 2b.

The antibody-immobilized SWCNT-FET showed clear FET characteristics and the drain current was regarded as value when concentration of PSA is zero. After treatment of the channel area of the FET with the PSA the SWCNT-FET still showed FET characteristic and the drain current evidently increased. The drain current at -3 V of both drain and gate voltages increased with increasing the concentration of PSA, as shown in Figure 4 (left hand). It is considered that the PSAs specifically bind to sites of the antibody, and then affect electronic state of the SWCNTs which form FET channel, resulting in change of the drain current [4]. The increase of the drain current is almost quantitatively proportional to the concentration of PSA (Figure 4, right hand), indicating that the SWCNT-FET has a basic function for biosensing of the PSA. For lower concentration of PSA (1 ng/ml) no drain current increase was observed, indicating other chemical materials in the buffer solution of the PSA do not induce such evident influence on the SWCNT-FET. The current increase was eventually and evidently observed for the concentration higher than 5 ng/ml, therefore, it is considerable that the sensitivity of the SWCNT-FET is roughly 5 ng/ml which is similar value to that reported by Li et al. [4].

It is noted that further improvements of reproducibility and sensitivity, for example, by alignment of the SWCNTs on the channel or by single SWCNT channel, may need to utilize such SWCNT-FETs as practical biosensors.

Conclusion

In conclusion, SWCNT-FETs were fabricated on $SrTiO_3$ substrate through a wet-process by using amide-functionalized SWCNT. The SWCNT-FET exhibited good gate-modulation for drain current at low operating voltages (-3 V). The hole mobility of 0.19 cm^2/Vs with an on/off current ratio of 1.3. After immobilization of prostate-specific-antigen (PSA) antibody the SWCNT-FET clearly responded against the PSA. The drain current at -3 V of both drain and gate voltage almost linearly increased with increasing the concentration of the PSA. The results indicated that the amide-functionalized SWCNT is potentially good candidate for the PSA-sensing FETs.

Acknowledgement

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