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# Calibration method for a carbon nanotube field-effect transistor biosensor

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**Abstract**
An easy calibration method based on the Langmuir adsorption theory is proposed for a carbon nanotube field-effect transistor (NTFET) biosensor. This method was applied to three NTFET biosensors that had approximately the same structure but exhibited different characteristics. After calibration, their experimentally determined characteristics exhibited a good agreement with the calibration curve. The reason why the observed characteristics of these NTFET biosensors differed among the devices was that the carbon nanotube (CNT) that formed the channel was not uniform. Although the controlled growth of a CNT is difficult, it is shown that an NTFET biosensor can be easy calibrated using the proposed calibration method, regardless of the CNT channel structures.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Biosensors represent one of the important applications of nanostructured carbon materials [1–9] such as carbon nanotubes (CNTs) [10–13], carbon nanowires [14], and carbon nanohorns [15, 16]. The carbon nanotube field-effect transistor (NTFET) biosensor [17–19] can be easily prepared by using a conventional device fabrication process. In an NTFET biosensor, the CNT functions as the channel of the field-effect transistor (FET). NTFET biosensors sense the electric charge of living molecules such as proteins [20, 21], glucose [22], and DNAs [23, 24]. For example, in an antigen sensor, a specific target protein (antigen) is adsorbed on the antibodies that are fixed on the gate of an NTFET by an antigen–antibody reaction. Since the target protein carries a charge, it changes the drain current after adsorption because its electric charges modulate the CNT channel.

For NTFET biosensors, the most important challenge that should be overcome is developing a precise technique for controlling the CNT growth; this is because the characteristics of an NTFET are determined by the CNT channel. In order to produce devices with uniform characteristics, it is essential to control the growth of the CNT in terms of chirality, location, number, and direction; however, these controls are difficult to achieve.

Although many researchers have attempted to achieve the control of chirality, location, number, and direction [25–27], limited results have been reported; furthermore, these methods were unsuitable for the mass production of CNTs.

Our aim is not to achieve a precisely controlled growth of CNTs but to propose an easy calibration method for NTFET biosensors. By using the proposed method, we can quantitatively detect living molecules using any NTFET biosensor without precise control of the CNT growth.

In our previous research, we found that the adsorption behaviour of an antigen on the antibodies fixed on a gate

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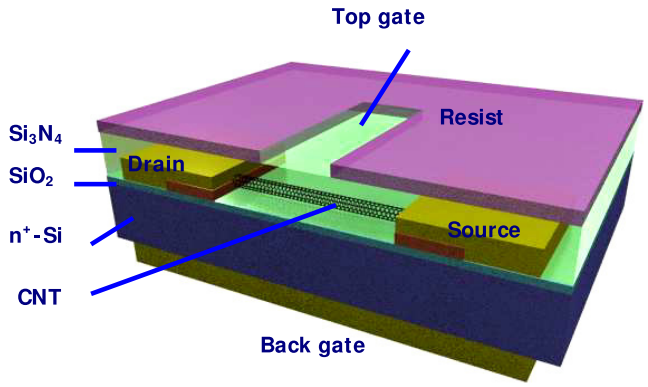


**Figure 1.** Schematic of an NTFET biosensor

electrode of a CNT obeyed the Langmuir adsorption mechanism [18]. Hence, in this study, we will show that an NTFET biosensor can be calibrated using the Langmuir adsorption equation.

## 2. Experimental set-up

For this study, we used three n-type NTFET biosensors of the same structure, as illustrated in figure 1. The details of the fabrication process and device structure have been given in a previous article [18]. We attached an antibody to the top gate of NTFETs and converted them into antigen sensors. A pig serum albumin (PSA) and an anti-PSA (a-PSA) were selected as the antigen and antibody, respectively. By physical adsorption, a-PSA was attached to the top gate of the NTFET.

Because the isoelectric point of the PSA molecule is pH 4.8, it carries a negative charge above this pH. Then, the pH of the PSA solutions was adjusted to 8.0 using a buffer solution. Thus, the PSA adsorbed on the top gate lowered the source–drain current.

For measurements using the antigen sensor, the environmental conditions are important. For precise measurements, static electricity must be removed because it gives rise to a marginal error in the results. The temperature and humidity in the measurement field were maintained at 25 °C and at least more than 60%, respectively. For these purposes, a manual prober (705B-6, Micronics Japan) and a precision semiconductor parameter analyser (4156A, Hewlett-Packard) were set up in an electrically sealed room.

The measurement system of the PSA sensor is shown in figures 2(a) and (b). A silicone rubber wall was set around the top gate. A PSA solution was poured onto the top gate of the NTFET. A gate voltage was applied to the top gate from an Ag/AgCl reference electrode. The drain current was measured as a function of the gate voltage for several concentrations of the PSA solution. The occurrence of the PSA/a-PSA reaction was detected by a decrease in the drain current from its original value corresponding to the nonoccurrence of the reaction. The detailed experimental procedures were reported previously [18].

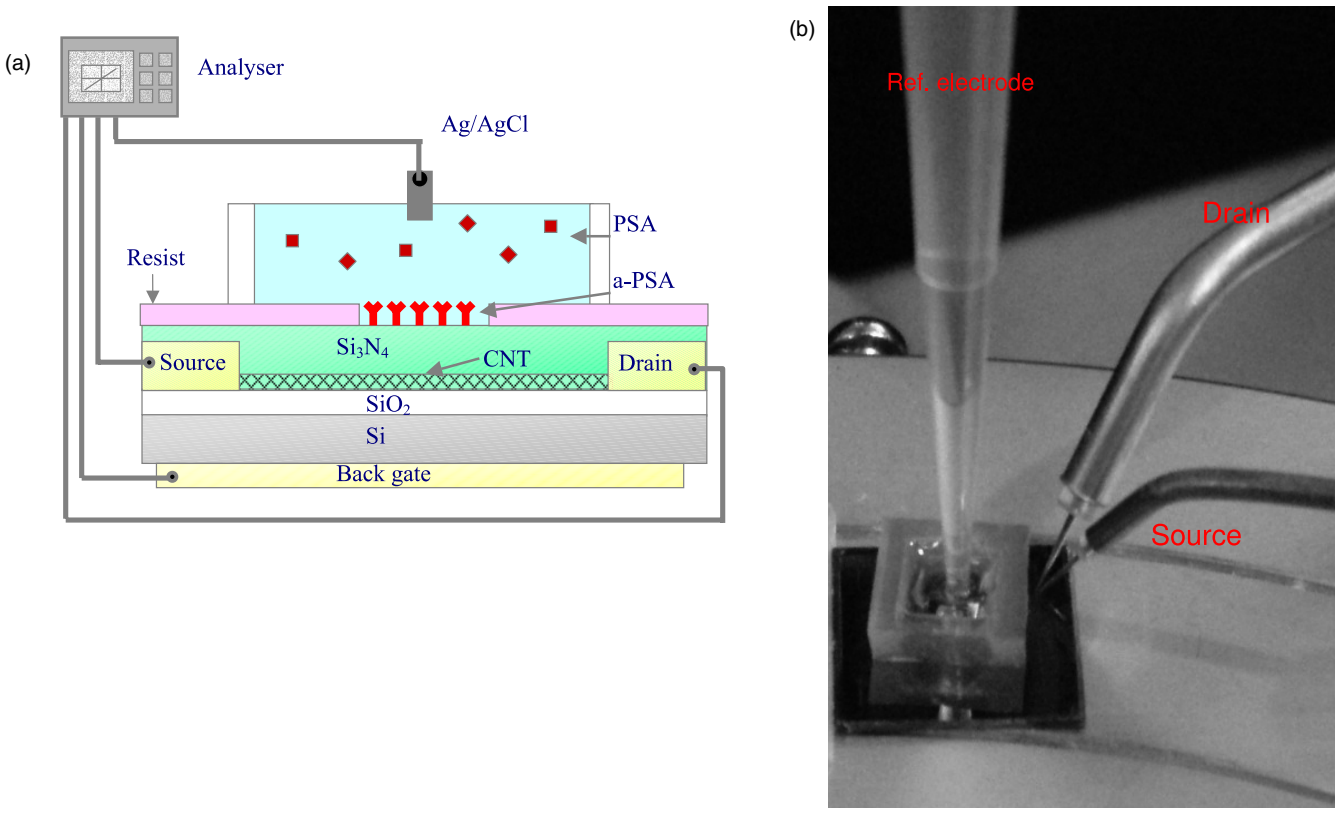


**Figure 2.** Experimental set-up of PSA sensing: (a) schematic and (b) photographic image.

## 3. Calibration method

Because the interaction between a macromolecule such as a protein and a solid surface is always strong, these adsorption isotherms are classified as type L of the Giles classification [28] (or type I of the IUPAC classification [29]). In many cases, these type-L adsorption isotherms follow the Langmuir equation without depending on the adsorption mechanism [30, 31]. Thus, a calibration method which uses the Langmuir equation is highly versatile. In addition, the adsorption of a PSA molecule on the a-PSA fixed on the top gate of an NTFET is considered to follow the Langmuir adsorption mechanism because the assumptions of this mechanism are satisfied; that is, the PSA molecule adsorbs 1:1 at an adsorption site, resulting in a monomolecular layer adsorption.

If the interactant molecule adsorbs on ligands fixed on the top gate of an NTFET following the Langmuir equation, the NTFET biosensor can be easily calibrated. The reason is that the Langmuir adsorption equation can be described by only two parameters—$\Delta I_{\mathrm{st}}$ and $K_{\mathrm{eq}}$. Here, $\Delta I_{\mathrm{st}}$ is the change in the drain current when the adsorption of the interactant molecules reaches saturation, and $K_{\mathrm{eq}}$ is the equilibrium constant of adsorption for the interactant molecules. When interactant molecules adsorb on specific ligands that are fixed on the sensor head, the Langmuir equation for the FET derived in [18] is given by

$$C/\Delta I = C/\Delta I_{\mathrm{st}} + 1/\Delta I_{\mathrm{st}} K_{\mathrm{eq}}, \tag{1}$$

where $C$ is the concentration of the interactant solution at equilibrium and $\Delta I$ is the change in the drain current. In these experiments, the interactant and ligand are PSA and a-PSA molecules, respectively.

$K_{\mathrm{eq}}$ is indicated as the strength of interaction between the interactant and the ligand because it is related to the Gibbs free energy. Hence, for the same pair of interactant and ligand, $K_{\mathrm{eq}}$ can be considered to remain constant. Thus, $K_{\mathrm{eq}}$ can be replaced with its expected value ($\langle K_{\mathrm{eq}} \rangle$). Here, we define the normalized drain current change $\theta$ as $\theta = \Delta I/\Delta I_{\mathrm{st}}$, and the calibrated Langmuir equation is then given by

$$C/\theta = C + 1/\langle K_{\mathrm{eq}} \rangle, \tag{2}$$

where $\Delta I_{\mathrm{st}}$ can be easily determined experimentally.

## 4. Results and discussion

The sensing results of the PSA molecules are shown in figure 3. In all the measurements, both the top gate and drain voltages were set at +1 V. Although the same concentration of the PSA solution was used, the values of $\Delta I$ significantly differed for the three devices. For example, when $1.0 \times 10^4$ nM of the PSA solution was measured using devices B and C, $\Delta I$ observed for device B was approximately 3.3 times larger than that observed for device C.

The reason why the observed values of $\Delta I$ differed among the devices is that the CNT that forms the channel is not uniform. In other words, the chirality of the CNT which

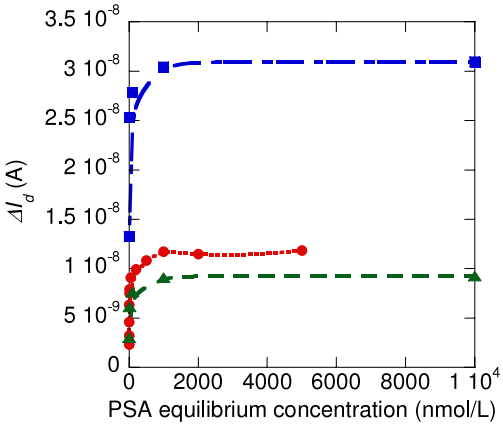


**Figure 3.** Characteristic curves of relationship between $\Delta I$ and equilibrium concentration of PSA. (●), (■), and (▲) denote devices A, B, and C, respectively.

**Table 1.** Characterization of NTFET biosensor for PSA sensing.

| Device | $g_{\mathrm{m}}$ (nS) | $\Delta I_{\mathrm{st}}$ (nA) | $1/K_{\mathrm{eq}}$ |
|---|---|---|---|
| A | 34.4 | 11.8 | $1.6 \times 10^{-8}$ |
| B | 164 | 31.0 | $8.1 \times 10^{-9}$ |
| C | 62.0 | 9.26 | $1.2 \times 10^{-8}$ |

formed the channel differed among the devices. In fact, the values of transconductance ($g_{\mathrm{m}}$) significantly differed among the devices (see table 1). Though devices A and B have the same structure, they have significantly different values of $g_{\mathrm{m}}$. This implies that the control of CNT growth is impossible. Furthermore, the experimental values of $\Delta I_{\mathrm{st}}$ also differed among the devices. On the other hand, it was confirmed that the variation in $1/K_{\mathrm{eq}}$ among the devices was small since the reaction with the same pair of interactant and ligand was measured. The value of $K_{\mathrm{eq}}$ obtained agreed well with that obtained by other measurement methods such as surface plasmon resonance [32] and infrared absorption spectrometry [33].

We then calibrated these devices using the above-mentioned method. Figure 4 shows the calibrated plots of figure 3; that is, $\Delta I$ is normalized by $\theta$. The solid curve in figure 4 was calculated using equation (2). $1/\langle K_{\mathrm{eq}} \rangle$ is regarded as the mean value of $1/K_{\mathrm{eq}}$ of the three devices: A, B, and C.

The experimental results obtained from the three devices were in good agreement with the solid curve. The low-concentration region of figure 4 is enlarged in figure 5. The experimental values and solid curve also coincided well in this region. Therefore, it was shown that an NTFET biosensor can be calibrated by using the Langmuir adsorption equation.

## 5. Conclusion

The usefulness of the method of calibrating NTFET biosensors using the Langmuir adsorption equation was demonstrated. This result demonstrates the possibility of developing a practical NTFET biosensor, even if the precisely controlled growth of a CNT with regard to its chirality, location, number,

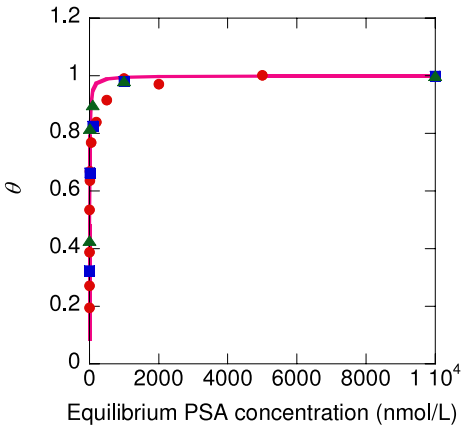


**Figure 4.** Normalized characteristic curves denoting the relationship between $\Delta I$ and the equilibrium concentration of PSA. (●), (■), and (▲) denote devices A, B, and C, respectively. The solid curve represents the calibration curve calculated using equation (2).

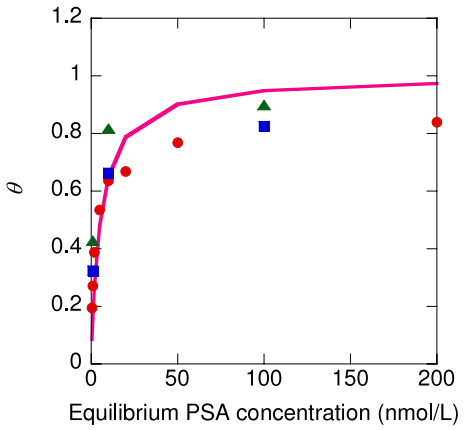


**Figure 5.** Low-concentration region of normalized characteristic curves denoting the relationship between $\Delta I$ and the equilibrium concentration of PSA. (●), (■), and (▲) denote devices A, B, and C, respectively. The solid curve represents the calibration curve calculated using equation (2).

and direction cannot be achieved due to technical difficulties. Hence, the present calibration method is useful practically.

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