LABEL-FREE IMMUNOSENSORS USING HORIZONTALLY ALIGNED CARBON NANOTUBES

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Direct growth of horizontally aligned single-walled carbon nanotubes (SWNTs) on single-crystalline quartz substrates offers several advantages for the application of carbon nanotube field-effect transistors (CNTFETs). The multiple, parallel transport pathways in CNTFETs provide large drain current (I_D) and active areas together with statistical averaging effect in properties, even with tubes that individually have widely different properties. In addition, the aligned arrays have advantages compared to the random networks because they avoid percolation transport pathways, unusual scaling of device properties, tube/tube junction resistances. Recently, guided growth of SWNTs on single-crystalline quartz substrates has been reported. They have found that SWNTs are aligned along the specific crystalline directions by angle-dependent van der Waals interactions between the SWNTs and the quartz lattice. This is one of the most facile routes to obtain horizontally aligned SWNTs. In this study, we demonstrate label-free immunosensing based on CNTFETs using horizontally-aligned CNTs as channels of FET.

In the experiments, aligned CNTs were grown on ST-cut Quartz substrates by alcohol CVD. The substrates were annealed at 900ºC for 12 hours in air to re-crystalize the surfaces before the growth. Source and drain electrodes of Ti (2 nm)/Au (30 nm) were formed. To measure protein adsorption in solution, the device was surrounded by a silicone rubber barrier attached to the substrate. The target protein is human immunoglobulin E (IgE), which plays an important role in allergy. To enable electrical detection of IgE, IgE aptamers were chemically modified on CNT channels.

First, we measured specific sensing characteristics of aptamer-modified CNTFETs. Time dependence of I_D was monitored in phosphate-buffered solution (PBS). Nontarget protein, namely, bovine serum albumin (BSA 4 μm) and avidin (4 μm), and the target IgE (500 nM) was added into PBS. Upon addition of nontarget proteins, I_D remained almost constant. In contrast, when the target IgE was introduced into the SWNT channels, I_D suddenly decreased. This result indicates that selective detection of IgE was successfully performed.

Next, we estimated the detection range of IgE concentration by monitoring I_D at various IgE concentrations. The target IgE at concentrations of 50, 75, 155, 250, 350, 400, 500 nM was introduced into an aptamer-modified CNTFET while I_D was monitored in real time. I_D decreased stepwise after injection of IgE at various concentrations. Then, the change in net source-drain currents (ΔI_D) before and after introduction of IgE (C_{IgE}) at each concentration is plotted. The experimental results were fitted well by the Langmuir adsorption isotherm, which is given by

$$\frac{\Delta I}{\Delta I_{max}} = \frac{C_{IgE}}{K_d + C_{IgE}}$$

where K_d is the dissociation constant of the reaction between IgE molecules and IgE aptamers and ΔI_{max} is the amount of saturated drain current. From the fitted curve, the detection range was estimated to be from 16.4 nM to 1.18 μM.

In conclusion, we have investigated an aptamer-modified CNTFET with multi channel as a label-free immunosensor. Dense, well-aligned SWNTs grown on quartz substrates were utilized as channels of CNTFETs. The CNTFET electrically detected IgE protein, whereas other proteins were not detected. The detectable range of IgE concentration was from 16.4 nM to 1.18 μM, which is a clinically relevant concentration regime. These results indicate that CNTFETs with multi channel is promising platforms for label-free immunosensors.