Field-Effect Sensor Based on Graphene Thin Films Fabricated by Layer-by-Layer Stacking

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Graphene based field-effect transistors and electrochemical electrodes have recently attracted a great deal of attention [1,2]. Graphene is a two-dimensional sheet comprising sp^2–hybrided carbon atoms. With its charge transport properties being extremely sensitive to local environment, graphene is considered a promising channel material for ion and biomolecule sensing in electrolyte. Unfortunately, large-scale fabrication of uniform graphene remains a challenge at the present. Graphene sheets derived from widely used approaches, such as exfoliation of graphite and vapor phase growth, are normally randomly located, with a wide distribution of electronic properties. As a result, great efforts are needed for construction of biosensors based on the available graphene. Furthermore, large diversities in sensor performance also make it difficult to interpret the results from different sensing experiments.

In 2010, Kim and colleagues reported a layer-by-layer stacking technology which can produce large scale (30 inch) graphene thin films with a controlled number of layers by chemical vapor deposition on a Cu substrate [3]. Excellent uniformity and stability make these films promising channel materials in field effect sensing applications. Here we use the same approach to fabricate graphene films on plastic substrate and subsequently construct a field-effect transistor (FET) as shown in Figure 1(a). Ion sensing properties of the graphene FET (GFET) are evaluated by means of current-voltage (I_{DS}-V_{GS}) and capacitance-voltage (C-V) methods. Schematic representations of the experimental arrangement for the characterization schemes are shown in Figure 1(b) and Figure 3(a).

The derived films comprise 4 stacking layers in average with a transmittance of 85% and a sheet resistance of about 300 ohm/□. As found in Figure 2(a), the I_{DS}-V_{GS} curves of the GFET in different NaCl solutions show similar an I_{on}/I_{off} ratio of 1.5. Furthermore, a 150-mV negative shift of the minimum conductance point is observed when increasing NaCl concentration from 0.01 to 10 mM. However, the I_{DS}-V_{GS} shift is negligible when changing the pH value of electrolyte, i.e., [H^+], as seen in Figure 2(b). The C-V curves measured in different NaCl solutions are depicted in Figure 3(b). The electrical double layer capacitance, i.e., C_{EDL}, can be viewed as a Stern layer capacitance (C_{Stern}) and a diffuse layer capacitance (C_D) connected in series and calculated as $1/C_{EDL}=1/C_{Stern}+1/C_D$ [4]. As NaCl concentration increases, the diffuse layer becomes more compact. Consequently, C_D, C_{EDL} and the gate capacitance (C_G) should increase. However, the measured C_G of GFET increases when increasing NaCl concentration from 0.01 mM to 0.1 mM, and then decreases when the concentration further increases from 1 mM to 10 mM. The results indicate that the NaCl solution contains ions that can specifically absorbed to graphene surface and thereby modifying C_{Stern}. As shown in Figure 3(c), a negative shift of the C-V curve by increasing NaCl concentration demonstrates that the specifically absorbed species are positively charged, i.e., Na^+ ions. On the contrary, H^+ can neither specifically adsorb to graphene surface nor chemically react with the functional groups on a graphene surface. Thus, the GFET is insensitive to H^+ in this case.

References

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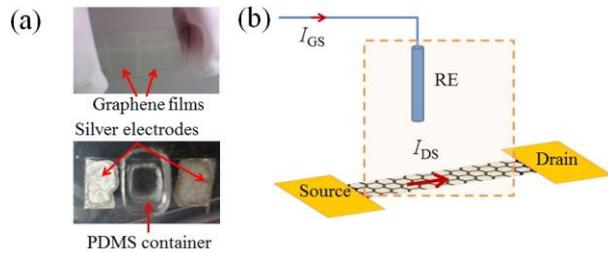
Figures

Figure 1 (a) Photo pictures of graphene films transferred onto a PET substrate (upper) and a GEFT with PDMS container and manually coated silver electrodes (lower), RE: Ag/AgCl reference electrode, (b) schematic representation of I_{DS}-V_{GS} measurement arrangement.
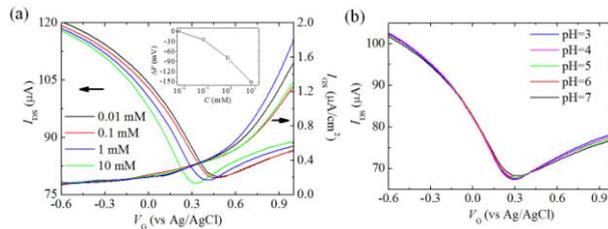

Figure 2 (a) I_{DS}-V_{GS} and I_{GS}-V_{GS} (gate leakage current) curves measured in different NaCl solutions, insert: shift of the minimum conductance point as a function of NaCl concentration, and (b) I_{DS}-V_{GS} curves measured in 0.1 M NaCl solution with different pH values, all at V_{DS}=0.1 V.
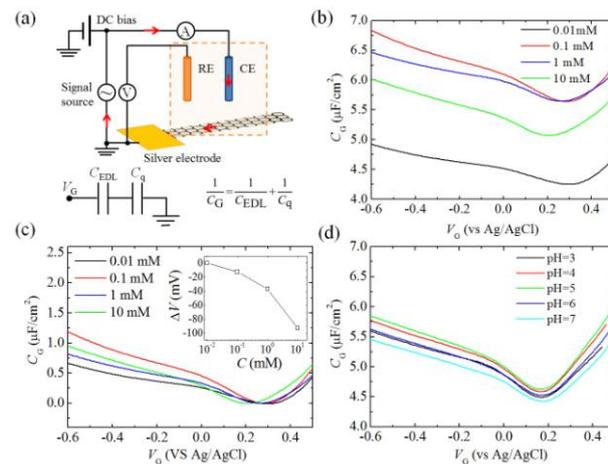

Figure 3 (a) Schematic representation of the C-V measurement arrangement, CE: platinum counter electrode, (b) measured and (c) vertically shifted C-V curves in different NaCl solutions, insert: shift of the minimum capacitance point as a function of NaCl concentration, and (d) C-V curves measured in 0.1 M NaCl solution with different pH values.