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Fabrication of integrated field-effect transistors and detecting system based on CVD grown graphene

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ABSTRACT

Based on large-area multilayer graphene produced by chemical vapor deposition, an integrated field-effect transistor was fabricated in this work. A planar Au electrode was used as gate electrode integrated with graphene field-effect transistors (FETs) to generate perpendicular electrical fields between the gate and the graphene substrate. Graphene film was then transferred to cover indium tin oxide films which were used as drain and source electrodes. Via this process, the contact resistance between graphene and electrodes which were caused by polymethylmethacrylate (PMMA) residues in chemical vapor deposition process was almost eliminated. An electrical detecting system was designed to detect equivalent resistance of the FETs, which indicated that the performance of the FETs was associated with the width of conducting channel, the electrical field intensity between gate and graphene, and the ion concentration of electrolyte. Adenosine triphosphate (ATP) was selected as model electrolyte to verify the detecting system in this study. It was demonstrated that the integrated FETs could achieve high sensitivity to detect ATP as low as 10 pM and the equivalent resistance of the FETs showed a good correlation with ATP concentration from 10 pM to 10 μM. These results may provide a better direction and a common detecting platform for the design of integrated graphene FETs biosensors.

1. Introduction

As an interesting field, applications of biochemical sensors based on graphene have been investigated utilizing the interaction of chemical and biological species with pristine or functionalized graphene surfaces [1,2]. Interactions between biomolecules or adsorption of biomolecules on graphene surfaces could modulate electrical charge transport via changes of the carrier's concentrations and carrier's mobilities caused by electrostatic gating or charge doping effect in the channel [3,4]. For this reason, several theoretical and experimental works involving solutions gated graphene field effect transistors (FETs), in which the channel conductance is modulated by applying a gate potential through electrolyte [5], have been reported for pH sensing [6], DNA [7], proteins [8], and cellular activities detection [9]. Since large area graphene films grown by chemical vapor deposition (CVD) exhibit better reproducibility compared to the chemical exfoliation processes [10] and mechanical exfoliation, the CVD processes are more favorable for graphene FETs fabrication [7].

However, several limitations in graphene FETs fabrication and detecting system have been identified in various laboratories. Firstly, the development of graphene-based electronics is limited by the quality of the contacts between the graphene and the electrodes [11,12] which can significantly affect the electronic transport properties of the devices. A high contact resistivity limits the total on-state current, and has a severe impact on transistor performance, negatively influencing the peak trans-conductance as well as the linearity of the current versus gate-voltage characteristics [13]. Secondly, while Ag/AgCl electrode was adopted generally as gate electrode in graphene FETs [14] for stable gate voltage, however, Ag/AgCl electrode was apart from graphene FETs and might not be very suitable for transistor integration and practical application.

In this study, we constructed an integrated graphene FETs (IGFETs) which adopted planar Au sheet as gate electrode, indium tin oxide (ITO) electrode as drain and source electrodes respectively. The planer Au gate electrode was then integrated with graphene FETs and was connected with an adjustable constant voltage source which was to generate stable electrical field between gate and graphene to modulate channel conductance as well as Ag/AgCl electrodes. So far as selected drain and source, CVD grown graphene films were transferred and covered on ITO films. Via this process, it was anticipated that the graphene could contact with ITO directly without polymethylmethacrylate (PMMA) residues to decrease the contact resistance. Considering that the channel conductance of graphene FETs could be modulated, the equivalent resistance of the FETs was acquired as signal [15] by a bridging circuit and an instrumentation amplifier. The width of conducting

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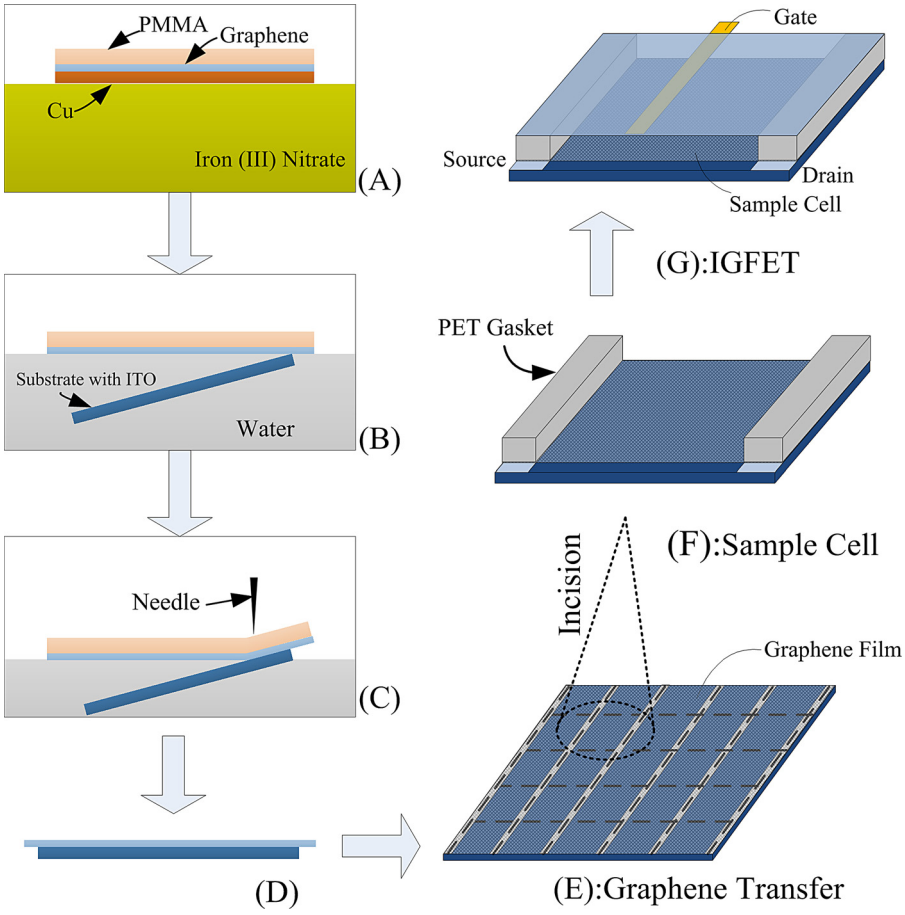

Fig. 1. Schematic of the graphene transfer and IGFETs fabrication. (A–D) Graphene film transfer onto the glass substrate with ITO. (E–G) IGFETs fabrication.

channel of FETs, the distance between gate and graphene, and the ion concentration of electrolyte were studied. We also investigated the effect of the sensing performance and surface charge state of the devices.

Adenosine triphosphate (ATP) is not only a universal energy currency, but also an important signaling molecule regulating many biological functions in various cells [16]. In this study, ATP was selected as model electrolyte to verify the performance of the IGFETs. Generally, ATP concentrations in various cells would be within reasonably constant content of approximately micromole order [17]. ATP has been reported to be detected by fluorescence and electrical method respectively [16–18]. Interestingly based on graphene biosensor, as low as pM order of ATP were detected in a number of published research papers [19–21] .The graphene FETs design in this work could also achieve even higher sensitivity to detect as low as 10 pM of targeted ATP. The equivalent resistance of FETs showed a good correlation with ATP concentrations varied from 10 pM to 10 μM. The results could provide better understanding and useful information for the design of integrated graphene FETs transistors.

2. Materials and methods

2.1. Materials and reagents

Glass substrate (~40 mm × 40 mm) with indium tin oxides (ITO) electroconductive film was purchased from Hua Nan Xiang Cheng Ltd. (Shenzhen, China). Phosphate buffered saline (P5368-10PAK, pH = 7.4) and adenosine 5′-triphosphate disodium salt hydrate was purchased from Sigma–Aldrich (Shanghai, China). ATP hydrate was dissolved and diluted by phosphate buffered saline to electrolyte with concentrations from 10 pM to 10 μM. Silver conductive paint was purchased from CAIG Laboratories Inc. (CW-200, USA).

2.2. Preparation and transfer of graphene by CVD method

Centimeter-scale graphene films (~100 mm × 100 mm) were grown using a CVD method as shown in Fig. 1(A–D) [22]. PMMA solution (20 mg/mL) was spin-coated on graphene/copper foils at 4000 rpm for 30 s and dried in vacuum. The copper was etched with iron (III) nitrate solution (0.05 g/mL in water), and the PMMA/graphene film was floated on the surface of the solution. After rinsing the PMMA/graphene film with distilled water, the glass substrate with ITO electrode was placed in the water just under the floating PMMA/graphene film (Fig. 1(B)). The water was sucked out using a syringe to lower the PMMA/graphene film onto the glass substrate. Since the floating PMMA/graphene film would drift in the water, a needle was used to adjust and position the floating film right above the glass substrate (Fig. 1(C)). Since the size of the floating film was larger than the glass substrate, the glass substrate would be covered completely by the floating film with no need for positioning accurately. After vacuum drying for 3 h, the substrate with PMMA/graphene film was heated at 180 °C in air for over 30 mins to enable the flattening of the graphene film on the substrate and remove the water completely (Fig. 1(D)). Through this method, the ITO electrode on glass substrate was contacted directly with graphene film without PMMA residues. An additional annealing process was performed at 400 °C with Ar (~500 sccm) and H_2 (~500 sccm) for ~1 h to remove further