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Fabrication of graphene FETs combined with fluorescence and its Double Read-Out System

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ABSTRACT

Based on the fact that graphene films grown by Chemical Vapor Deposition (CVD) have unique electronic and optical properties concurrently, a Graphene Field-Effect Transistor (GFET) which combined electronics and fluorescence was proposed and fabricated in this work. Comparing conventional GFETs, fluorescent probes were dissolved in the electrolyte of GFETs. Consequently, the combined GFETs could not only be detected by common electrical method but also be detected by fluorescent method. A Double Read-Out System (DROS) was constructed to monitor the combined GFETs by electrical method and fluorescent method synchronously. The combined GFETs and DROS have been evaluated by sensing pH value of a Phosphate Buffered Solution (PBS). Both the electrical signal and the fluorescence intensity of the combined GFETs were recorded in real-time and synchronously. Through data processing, the combined effect of electrical signal and fluorescence intensity lead to a combined sensitivity that is higher than each isolate method. These results may provide a new feasible direction for CVD graphene FETs research.

1. Introduction

Graphene and its derivatives have attracted a great deal of attention because of its unique structure [1] as well as its electronic [2] and optical properties [3–5]. The electrical properties of graphene are highly sensitive to the interaction between the graphene surface and adsorbed foreign molecules or ions [6] and this makes them ideal as conducting channel in Field-Effect Transistors (FETs) [7,8]. Since the channel conductance of the Graphene FETs (GFETs) could be modulated by electronic-doping through solution electrolytes, GFETs have attracted considerable attention in pH [9], DNA [10] and protein sensing [11], as well as cellular activities [12]. Compared to the other processing methods such as chemical exfoliation and mechanical exfoliation, graphene films grown by Chemical Vapor Deposition (CVD) method are more favorable for device fabrication because of its intrinsic properties in terms of 2D electrical conductivity, large surface area and superior reproducibility [13].

Another advantage of graphene is its intrinsic optical properties. Firstly, since the energy transfer from dyes to graphene, graphene can be an excellent quencher of electronic excited states of dyes [14]. Therefore, it is possible to realize sensitive fluorescent sensing using graphene as a fluorescence quencher. Fluorescent probes based on graphene and its derivatives have been investigated in sensing DNA [15], ATP [16], and small cells [17]. Secondly, since CVD graphene has ultra-high light transmittance (>96% for single layer CVD graphene films) [18], they are also widely used in optical fields, such as transparent electrodes [19] and ultrafast pulsed lasers [20].

Graphene has unique electronic and optical properties concurrently, it may be feasible to fabricate a combined device which could be monitored through optical and electronic method synchronously. In order to verify this assumption, GFETs combined with fluorescence and a Double Read-Out System (DROS) have been fabricated and examined through pH sensing in this work. In the combined GFETs, CVD graphene films were transferred onto a substrate as a conducting channel in a similar way to conventional GFETs. The GFETs have exhibited clear pH-dependent conductance characteristics since hydroxyl (OH−) or hydroxonium (H_3O+) may be able to modulate the channel conductance by doping "holes" or "electrons"[9,21]. The main difference here from conventional GFETs was that fluorescent dyes were dissolved in the solution electrolyte as pH indicator. Via this way, the combined GFETs could generate an additional fluorescence comparing with conventional GFETs. Since the CVD graphene films have high light transmittance, the exciting light and emission fluorescence could be detected through the transparent CVD graphene films. A DROS has also been constructed for collecting electrical and fluorescence

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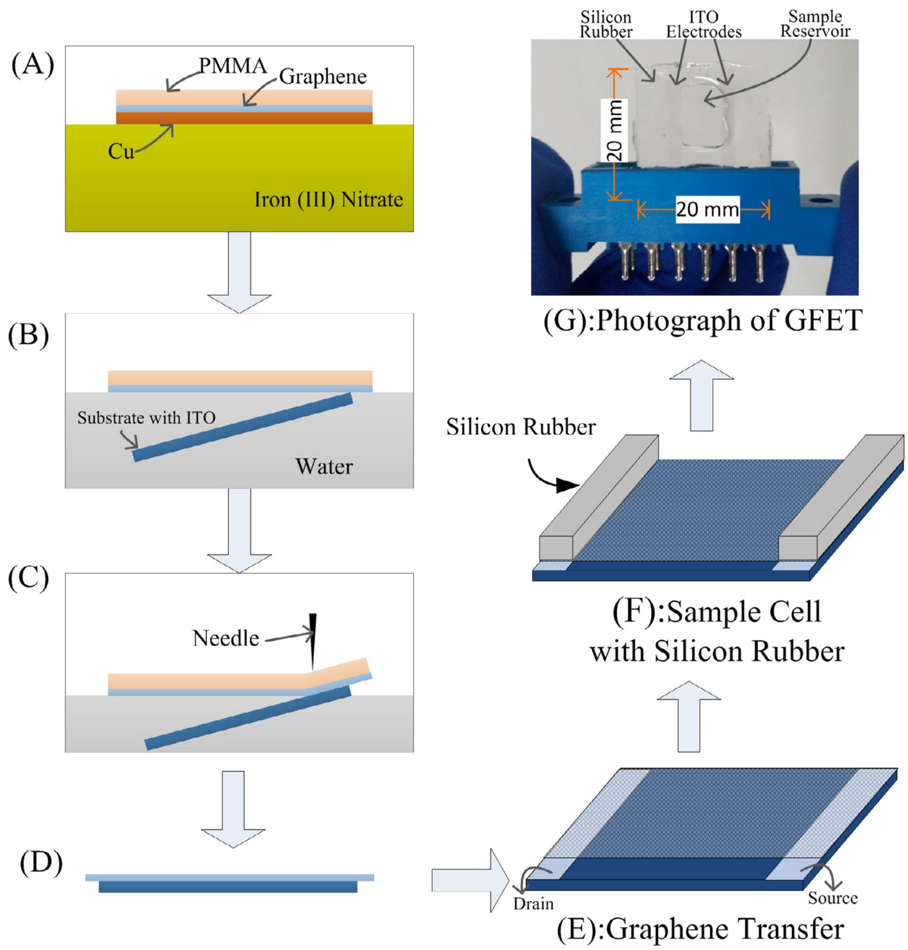

Fig. 1. Schematic of the graphene FETs fabrication.

signal simultaneously. Utilizing the DROS and the combined GFETs, pH changing in Phosphate Buffered Solution (PBS) has been monitored in real time. Through processing the combined data, the combined GFETs have shown a higher sensitivity than each isolate method. In this paper, the fabrication of combined GFETs and DROS, the electrical and fluorescence characteristics of GFETs and the combined data processing method were all described in detail.

2. Experimental

2.1. Materials and instruments

Glass substrates (20 mm × 20 mm × 1 mm) with indium tin oxide (ITO) electroconductive film were purchased from Hua Nan Xiang Cheng Ltd. (Shenzhen, China) as a substrate for graphene FETs. Phosphate buffered saline's (PBS) (pH = 6.24, 6.55, 6.90, 7.35, 7.65, 8.01, 8.50) were purchased from O'BioLab Technology Ltd. (Beijing, China) as test samples. Carboxyfluorescein (6′-FAM) was purchased from Life Technologies Ltd. (Beijing, China) as pH fluorescence probes. A LED SMB490R (EPITEX, Japan) was used as exciting light source for 6′-FAM and a photomultiplier tube (PMT, CH253, Hamamatsu Photonics (China) Co., Ltd.) was selected as an optical–electrical converter device. Optical guide fiber (Chun Hui Optics Ltd., Nanjing, China), optical filters (FF01-520, Semrock) and convergent lens (Zolix, Beijing, China) were used to construct the fluorescence optical unit. Ag/AgCl micro-electrode (R0303, AiDaHengSheng, Tianjin, China) was adopted as gate electrode in GFETs.

The quality of transferred graphene films to the glass substrate was determined by confocal Raman microscopy (SPEX-1403, SPEX). The emission spectrum of the exciting LED was measured by an optical fiber spectrometer (USB2000, Ocean Optics). The absorption and emission spectra of 6′-FAM were scanned by a fluorescence spectrophotometer (LS55, PerkinElmer). A homemade DROS was used to collecting current and fluorescence signal simultaneously.

2.2. Fabrication of graphene FETs

The preparation and transfer of graphene by CVD method have been reported in our previous work [23] as in Fig. 1.

Briefly, PMMA solution was spin-coated on graphene/copper foils and then the copper was etched with iron (III) nitrate solution (A). The glass substrate with two ITO electrodes (taken as drain and source electrodes respectively in (E)) was placed in the water just under the floating PMMA/graphene film and then the water was sucked out using a syringe to lower the PMMA/graphene film onto the glass substrate (B, C). After vacuum drying and heated, the water and the PMMA was eliminated (D). The ITO electrodes covered by graphene films were taken as source and drain electrodes respectively (E). Silicone rubber was used to build a reservoir for sample solutions (F). The size of the fabricated GFETs was 20 mm × 20 mm × 3 mm and the volume of the sample reservoir was about 10 mm × 5 mm × 2 mm (100 μL). The GFETs were