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THE EFFECT OF SOLUTION VOLUME OF GRAPHENE OXIDE FOR THE APPLICATION ON ELECTRODE FOR BIOSENSOR DETECTION

A. Rashid Diyana[1], A. Rahim Ruslinda[1], M. F. Fatin[1], Saeed S. Ba Hashwan[1], U. Hashim[1] and M. K. Md Arshad[1, 2]
[1]Institute of Nano Electronic Engineering, Universiti Malaysia Perlis (UniMAP), Kangar, Perlis, Malaysia
[2]School of Microelectronic Engineering, Universiti Malaysia Perlis (UniMAP), Kangar, Perlis, Malaysia
E-Mail: ruslinda@unimap.edu.my

ABSTRACT

The effect of different volume of graphene oxide (GO) were demonstrated to provide a suitable biosensor platform. A simple technique using spray method was presented to deposit GO on silicon substrate. This method is a promising method due to ideal coating on a variety of substrates with high production speed. Thermal reduction process was selected to reduce the GO and produce even flakes of the coating material. Reduced graphene oxide (RGO) was obtained after heating on hot plate fixed at temperature of 250˚C. A surface morphology of RGO using Scanning electron microscopy were observed imply that the morphology obtained were rely on the parameters such as reduction temperature, volume of GO solution and height of spray nozzle. In this study, the height of 10 cm with a 0.3 ml GO solution was successful presented for RGO deposition on electrode for biosensor. The electrical were conducted for Tat protein detection revealed that shift of current occurred during each step of immobilization and detection.

Keywords: Reduced graphene oxide (RGO), electrode, biosensor, SEM, I-V.

INTRODUCTION

Carbon-based material including carbon nanotubes (CNTs), diamond (Burghard *et al.* 2009) and graphene are central materials in nanoscience. Their unique electrical, physical, mechanical (Pumera 2011), (Yeon *et al.* 2014) and chemical properties (Kwak *et al.* 2012) are widely studied so as to develop biosensor devices. Biosensor is an analytical device that utilize biological component to act as molecular recognition unit. It combines the biological component and modern microelectronic to form a powerful analytical tool for the application in pharmaceutical, medical, environmental and food analysis. Graphene is a 2 dimensional (2D) single layer of carbon atoms that is surrounded by six electrons around the nucleus are arranged in the electron configuration of. The outer orbitals combine or hybridize to form hybrid orbitals (Pham *et al.* 2010).

Graphene material can be produced by mechanical exfoliation (Kochmann *et al.* 2012) which needs the repeated pealing of pyrolytic graphite. Chemical vapor deposition (CVD) (Kuila *et al.* 2011), (You & Pak 2014) is a cost and time effective method for producing high quality graphene in large quantities. The process of graphene is performed under vacuum and uses heat to break apart the atoms in a gaseous hydrocarbon such as methane. Graphene may be prepared by thermal decomposition on silicon carbide (SiC) wafers (Sharma & Ahn 2013), grown by chemical vapor deposition (CVD), spray deposition process or chemically synthesized by various methods. Recent studies of graphene based biosensors have been performed using reduced graphene oxide (r-GO) due to its easy functionalization procedure (Hasegawa *et al.* 2012).

In a reduced graphene oxide modified electrode, graphene sheet acts as the semiconducting channel between two metal source and drain electrodes which lie atop an electrical insulator such as SiO2.In this study, a fast, low cost, simple method to fabricate conductive RGO films by spray deposition was developed. When spraying on the preheated substrate, GO sheets were reduced to form a RGO thin film. The RGO thin films were measured and characterized by scanning electron microscopy (SEM), and the electrical measurements were carried out to test the performance and the output current.

EXPERIMENTAL

Fabrication of RGO-electrode

The fabrication process of RGO-electrode device was done by using 1cm x 1cm glass substrate. The glass was cleaned with distilled water for 5 minutes and then sonicated with isopropyl alcohol (IPA) for 30 minutes. After that, Physical Vapor Deposition (PVD) process was used for deposition of Aluminium as a source and drain. To fabricate graphene films by spray process, graphene oxide (GO) solution at a volume of 0.3ml, 0.4ml and 0.5ml were sprayed onto the glass substrate placed on the hot plate fixed at 250˚C. Here, the distance between nozzle and the substrate was fixed at 10cm. Then, 2% of aptes solution was dropped into the channel and let it dry for 1 hour at ambient temperature. After that, the device is washed with phosphate buffer solution (PBS) and the electrical measurement was recorded. Terephthalic acid was dropped into the channel for 1 hour. After wash with PBS, the electrical measurement was recorded. 10 of DNA solution was dropped into the channel and incubated at 100˚C in the incubator. After 1 hour, the device was washed with PBS and electrical measurement was recorded. 10 of aptamer was dropped into the channel and incubated at 100˚C for 1 hour. After washing with PBS, the device underwent electrical measurement to test the performance and the output current. Then, 10 of 100nM

Tat Protein was dropped into the channel and incubated for 1 hour at 100˚C. Electrical measurement was recorded after washing with PBS.

Characterization of RGO film

Surface morphologies of RGO were examined using SEM. During the spray process, the critical parameters such as substrate temperature, distance between nozzle to substrate and volume of graphene oxide giving a significant effect on the morphology of the deposited graphene oxide. The electrical measurements were carried out using sourcemeter to test the performance and the output current of the device.

RESULTS AND DISCUSSION

Scanning Electron Microscopy (SEM)

The SEM images were compared to identify the changes occur when volume of GO solution sprayed are varied. By changing the deposition parameters, it can be seen that there is a variation in the morphology of reduced graphene oxide (RGO) on the substrate. At ambient temperature, GO contains flakes of monolayer and a few layer of graphene (Figure-1(a)). But as the substrate temperature was increased to 250˚C, the layer of GO decrease due to the carbon and oxygen react and outgas as CO2 at high temperature. With the increase of the GO solution volume, after reduction for 2 hours, there were different in the morphology of the RGO film obtained. In Figure-1, it can be observed that as the volume increased from 0.3ml to 0.5ml, there is increase in the thickness of the RGO film indicating overlapping of the RGO flakes.

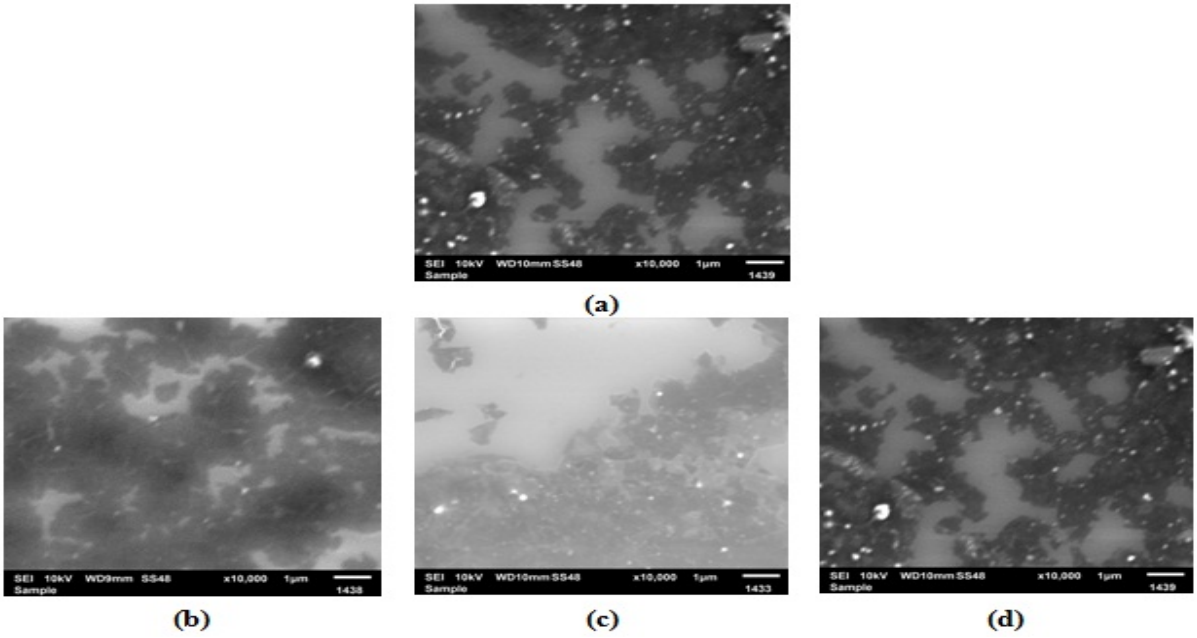

Figure-1. SEM images of (a) GO film and RGO film (reduced at 250˚C) using solution volume of (b) 0.3ml, (c) 0.4ml and (d) 0.5ml. The nozzle to substrate height was fixed at 10cm.

From the morphology of the RGO film, we have chosen the film produced by using 0.3ml of the GO solution. A good distribution of RGO flakes is needed to produce a biosensor platform. This RGO flakes will act as the base for the biomolecules immobilization in field effect transistor. A thin layer of RGO will give less resistance and large surface area for reaction.

Electrical characterization using sourcemeter

The electrode were characterized by using sourcemeter and the current to voltage (I-V) value was recorded. Figure-2 shows the relationship between voltage and current produced by the electrode. The RGO on the electrode surface was modified by using 10ml of 2% APTES solution. The soluttion was dropped into the channel to modified the RGO surface with amine functional group. APTES is highly reactive and silanizes the surface by forming covalent bonds with the hydroxyl group on the RGO. The changes in the electrical current when aptes was functionalized on the RGO proved that aptes was successfully reacted with surface hydroxyl groups to form amine-functionalization. 0.2M of Terephthalic Acid solution was dropped into the channel and acts as a linker that connects molecules between substrate and biomolecules. The increase in current was due to the presence of two negatively charged carboxyl functional group in terepthalic acid molecule. 10µl of aminated DNA was dropped after chemical treatment and show an increase in current value. This was due to the negatively charge DNA. We can confirm there are amide

binding occur between aminated DNA and terepthalic acid linker. Then, probe was introduce on the device by incubation with 100nM RNA Tat aptamer for 2 hours. The RNA Tat-tail aptamer form hydrogen bond with the DNA and immobilized on the device, increasing the resistance on the device and reduce the current output. Besides, both DNA and RNA aptamer are negatively charged due to its phoosphate backbone and tend to repel each other leaving the aptamer probe standing and accessible for reaction [14]. When 100nM Tat protein solutions were dropped into the sensing area, the current value decrease further from 3.3×10^{-5}A to 2.6×10^{-5}A. It might be the effect from RNA Tat aptamer folded and trapped the protein in the middle of the folding structure by electrostatic interaction with the positively charged Tat protein and leads to increase in resistance due to formation of the bulky folding structure on the sensing surface.

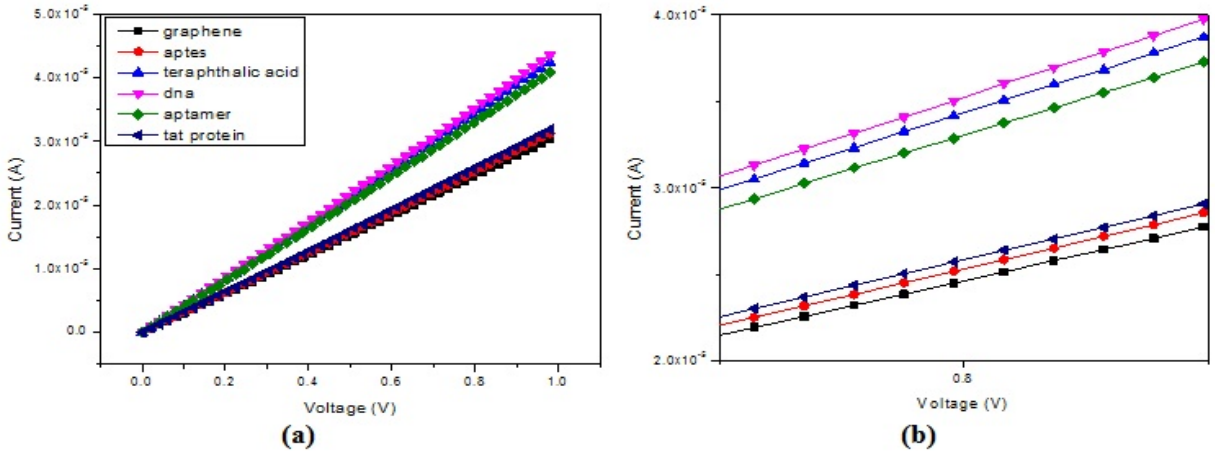

Figure-2. Electrical characteristics of the device after each immobilization and detection steps. (a) Full scale and (b) Enlarged scale.

This work has shown the capability of the RGO modified electrode in sensing of biomolecule interaction. Further work can be done to optimize the performance of the electrode in terms of stability, specificity and limit of detection.

CONCLUSIONS

Reduced graphene oxide modified electrode was successfully fabricated by using spray method. All the fabrication process for the device had been carried out and yield a good signal for each modification step. The current between aptamer and Tat protein was shifted by $0.7\text{x}10^{-5}$A and it shows that the RNA aptamer immobilization step on RGO has successfully detected HIV-1 Tat protein as a target in biosensor application.

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