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Graphene Multi-layer on Plastic Substrate

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Abstract. Vacuum filtration method was used in order to fabricate a homogeneous and uniform thin film of multi-layer graphene on plastic substrate. This self-regulating technique allows the number of graphene layer to be controlled thus controlling the film thickness by simply varying either the concentration of the graphene in the suspension or the filtration volume. Measurement of the sheet resistance as a function of graphene concentration in solution shows the percolation behavior of multi-layer graphene films.

1. Introduction

Graphene is a single layer of carbon atom arranged in honeycomb lattice like structure. It has been proved that graphene exhibits fast response and high sensitivity for detection of small concentration of toxic gases in room temperature due to very high volume-exposure ratio [1]. It has improved the sensitivity of the device by increasing charge carrier concentration [1].

Recently, the demand on low cost and flexible device has improved greatly. The use of plastic substrate become preferable as reduce the production cost and make it easy to wear this device as an 'easy-to-wear' item that can be used in any locations and activities for safety purposes. In this paper, the fabrication of graphene on plastic substrate is discussed in details.

2. Experimental Details

2.1. Graphene oxide production

First, graphene oxide (GO) was produced from graphite flake (NGS Naturgraphit GmbH, carbon contents >85%) using the modified Hummers method. The graphite powder (2.0 g) was added into a solution of concentrated hydrogen sulphide (H₂SO₄) (20 mL), potassium persulfate (K₂ S₂O₈) (1.0 g) and phosphorus pentoxide (P₂O₅) (1.0 g) mixture that were completely dissolved at 80 °C. Then the mixture was kept at 80°C for 4 and half hours using an oil bath. After that, the mixture was cooled to room-temperature and diluted with 1 L de-ionised (DI) water and left overnight. Subsequently, the resulting product was filtered (0.2 µm Milipore membrane) and washed with DI water until the pH of filtrate water become neutral in order to remove any residual acid. Then the oxidation process of Hummers method was performed. The pre-oxidised graphite was then dispersed by stirring into cold H₂SO₄ (80 mL) in an ice bath. Then, potassium permanganate (KMnO₄) (10 g) was added by stirring it slowly to keep the temperature of reaction mixture below 20 °C. After that, the mixture was stirred at 35 °C for 2 hours and diluted with DI



water (160 mL) to give a dark brown solution. The addition of water in concentrated H_2SO_4 released a large amount of heat. Hence water was added into an ice bath in order to keep the temperature below $50^\circ C$. Then, the mixture was further diluted by DI water (500 mL) and stirred for 2 hours. After that, the hydrogen peroxide (H_2O_2) (30%, 8 mL) was added slowly into the mixture and the colour of the mixture turned into yellowish from dark brownish previously. The resulting product was then washed with 10% hydrochloric acid (HCl) (1L) by stirring and the supernatant was decanted after overnight. The remaining product was then centrifuged and washed with DI water until the pH of the solution became neutral. This yellow – brown homogeneous suspension was stable for several months and used for reduction. This GO suspension was then further diluted with DI water to produce a set of different concentration that varied from 0.00004 mg/ml up until 0.01 mg/ml. The variation of concentration was critical in order to investigate the film conductivity as a function of concentration.

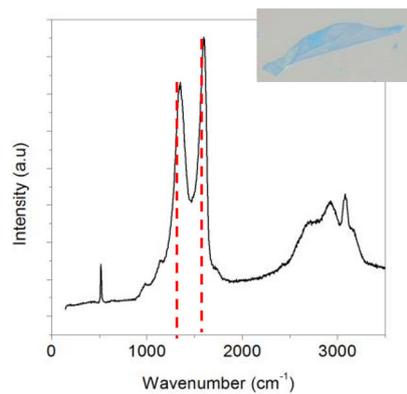


Figure 1. Raman spectrum shows the D and G peaks. (Inset) Graphene sheet on SiO_2 .

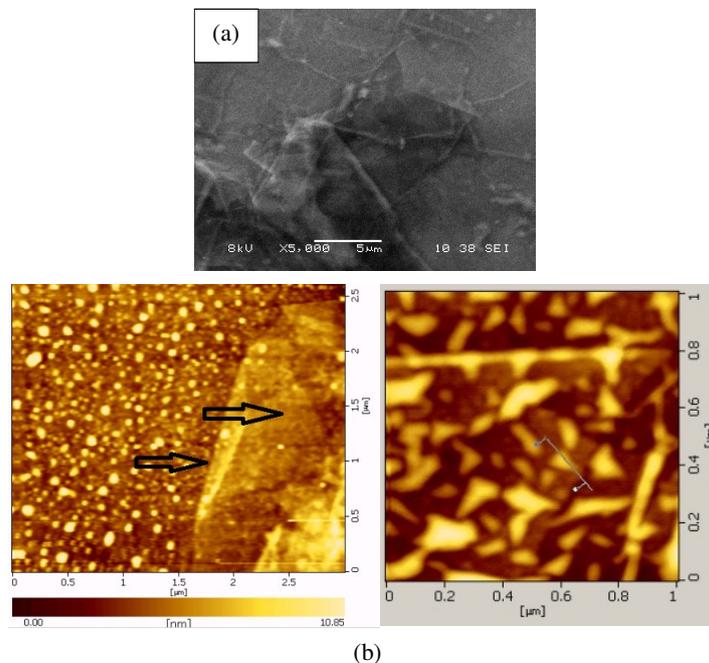


Figure 2. (a) SEM and (b) AFM image of few layers graphene oxide.

2.2. Thermal treatment

Peracetic Formation GO is electrically insulating because of the disruption of the sp_2 bonded graphitic structure by the oxygen atoms. A reduction process is needed to make GO conducting by removing oxygen functional groups. By exposing GO to reduction agent such as hydrazine [7] or through thermal treatment [8], GO can become conducting. Hydrazine reduction is believed to remove the oxygen functional groups [9]. However in doing so, hydrazine also introduces a significant amount of nitrogen-containing functional groups to the GO. The presence of these groups may decelerate the sensing response of the device when expose to various adsorbates [9]. Hence, the reduction process using thermal treatment method was preferred in order to increase GO electrical conductivity. Thermal reduction of GO was carried out in vacuum oven (Technico Ltd., India). The GO solutions were then placed in vacuum oven and treated at 100 °C, 200 °C and 300 °C for 1, 2 and 3 hours. After heating, samples were cooled to room temperature.

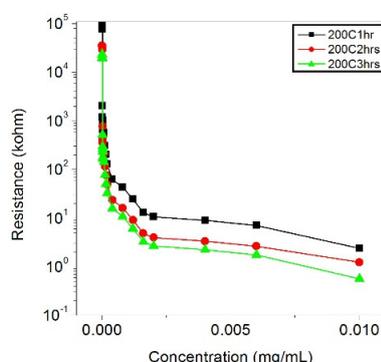


Figure 3. Electrical resistance of different concentration of graphene oxide solution for different annealing time.

2.3. Sample preparation

The production of homogeneous and thin film of r-GO was crucial. There are several methods to produce uniform film. Methods such as drop casting [12], rapid freezing by spraying [13] and dip coating [14] from r-GO colloidal suspension were used to obtain a thin and homogeneous film. In order to reproducibly achieve uniform thin films with a controllable layer (thickness) over large areas, vacuum filtration method was used. This method was widely used to deposit highly uniform r-GO thin films [15]. This method involves the filtration of r-GO suspension through a commercial polycarbonate membrane (0.1 μm pore size). As the solvent passes through the pores, the r-GO sheets were trapped on the membrane surface thus forming an interconnected network (Figure 2(a)). The permeation rate of the solution was controlled by the accumulation of r-GO sheets on the pores so as the number of tubes increases at a given location on the porous membrane, the rate of filtration decreases, but does so to a lesser degree at thinner or uncovered regions. The process is therefore self-regulating, which allows reasonably good nanoscale control over the r-GO sheets by simply varying either the concentration of the r-GO in the suspension or the filtration volume. Here, 0.5 ml of each solution was sucked through a membrane, coating a circular area with a diameter of 13 mm. Hence, the density of the r-GO sheets or thickness can be controlled by varying the concentration of the suspension filtered through the membrane.

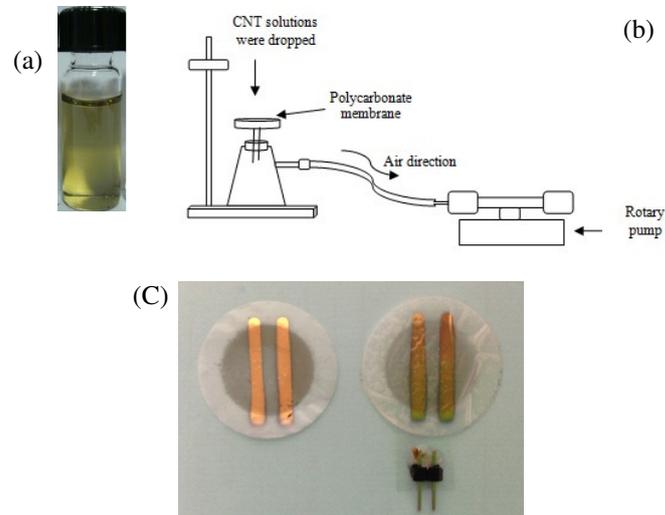


Figure 4. Graphene oxide suspension in 100% concentration. b) Illustration image of vacuum filtration set-up. c) Plastic membrane that was used as a substrate with graphene layer and gold electrodes for further potential applications.

3. Conclusions

A colloidal suspension of reduced-graphene oxide was produced by modified Hummer's method. The graphene sheets images was observed using SEM and AFM. Vacuum filtration method was successfully applied to produce conductive and uniform thin film of reduced-graphene oxide sheets. The optimum temperature for annealing treatment was observed at 200 °C for 3 hours.

Acknowledgment

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