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Electrical characterization of graphene oxide and organic dielectric layers based on thin film transistor

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ABSTRACT

We have studied the electrical characteristics of graphene oxide based thin flim transistor with the polymer insulators such as polymethyl methacrylate (PMMA) and poly-4-vinylphenol (PVP). Graphene oxide (GO) nanosheets were prepared by using modified Hummers method. The structural properties of GO nanosheets were characterized with Ultraviolet Visible (UV–vis), FT-IR spectroscopy and X-rays diffraction (XRD). Graphene oxide based thin flim transistor (GO-TFT) was prepared by a spin-coating and thermal evaporation technique. The electrical characterization of GO-TFT was analyzed by output and transfer characteristics by using Keithley-4200 semiconductor characterization system (SCS). The graphene oxide based thin flim transistor devices show *p*-type semiconducting behavior. The mobility, threshold voltage, sub-threshold swing value and I_{on}/I_{off} of GO-TFT were found to be 0.105 $cm^2 V^{-1} s^{-1}$, −8.7 V, 4.03 V/decade and 10, respectively.

1. Introduction

Graphene is one of the most exciting materials being investigated today, not only out of academic curiosity but also for its potential applications. Graphene, a two-dimensional (2-D) crystal of sp^2-bonded carbon atoms [1], has become the focus of intense fundamental research due to its extraordinary properties, but even more so has spurred massive interest into studies regarding nanotechnology applications [2,5]. An area of immense importance for the latter is the study of the metal–graphene interactions, because metals have to be used in every single application of graphene as functional material [3,5]. The effects of metals on the transport, electronic, optical, magnetic and structural properties [1,6–8] of graphene have been investigated both experimentally [3] and theoretically by means of density functional theory [4] with arguably more emphasis on theoretical than on experimental studies. Those characteristics graphene make graphene an ideal nanomaterial for nanoelectronics, nanodevices, and nanocomposites. Recently, graphene researchers have been focused on transistors and thin film applications, but the interest in different applications of graphene is growing rapidly [1,7]. Practical applications of graphene require a reliable high-throughput method of graphene identification and quality control, which can be used for large-scale substrates and wafers. One of the alternate methods to prepare graphene oxide (GO) is achieved by oxidizing graphite via modified Hummers method. GO is a very valuable material. Unlike graphene, GO is highly soluble in water and possesses many reactive groups (e.g., carboxyl and hydroxyl) but still maintains the basic framework of graphene. Thus, GO provides an excellent platform for preparing functional graphene materials or graphene composites [9]. Graphene oxide (GO) as chemical structure consists of sheets of sp^2 hybridized carbon atoms arranged in hexagonal fashion with few sp^3 hybridized carbon atoms attached to hydroxyl or epoxide moieties. The edge areas of these sheets bear carboxyl or carbonyl groups [4]. In flexible and large area electronic applications, the easy processability by solution based methods, dielectric properties, transparency and tunability of electronic properties widen the scope of graphene oxide (GO) [10,11].

The semiconducting properties of organic materials, combined with their ease of synthesis and processing, make them promising candidates for application in a variety of electronic and optoelectronic devices [12]. For the low-cost and large-area electronic applications, graphene oxide based organic thin film transistors on flexible plastic substrates are of interest. In the organic semiconductor research area, two of the commonly used insulator layers are polymethylmethacrylate (PMMA) and polyvinyl phenol (PVP). Both PMMA and PVP can be solution processed, have high resistivity and have high thermal/mechanical stability. Thus they tend to be very effective organic insulators. However, it is to be noted that their different chemical structure have resulted in significantly different electronic properties when used as dielectric in graphene oxide based organic thin film transistor configurations [13–15].

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A lot of research efforts have been concentrated thin film transistors based on bilayer oxide semiconductors which have attracted considerable attention because of their shown excellent electrical characteristics, large field effect mobility values, high optical transparency and environmental stability [16]. Proper attention to device structure design and materials integration is crucial for optimizing TFT performance. In particular, bilayer oxide based TFTs, where two metal oxide films are used as the channel layer, offer significant performance advantages by combining the properties of two semiconducting materials. Furthermore, when compared to the corresponding any single layer devices, bilayer devices effectively enhance the photostability and suppress normalized current noise spectral density [17].

The basic function of a field-effect transistor is to modulate the current flowing in a conducting channel between two source and drain electrodes by means of a variable voltage applied at a third electrode, the gate. When no potential is applied to the plates, the free carriers are homogeneously distributed within the entire layer of the organic semiconductor. Owing to their low density, the conductance of the layer is very small and the current between the source and drain electrodes is very low. Now, when polarizing the gate negatively, an excess of holes will be attracted at the surface of the *p*-type semiconductor and will be concentrated there within a very thin layer [18–20]. The density of charge carriers in this very thin layer will be largely increased, allowing a significant conductivity to be created in this conducting channel. On the other hand, when applying a positive bias on the gate, a depletion of charge carriers occurs in the semiconductor, which, under sufficiently high gate voltage, can be extended over the whole thickness of the semiconducting layer. Thus the external voltage applied to the gate allows either the accumulation of charge carriers at the semiconductor–insulator interface or the depletion of this interface, leading to a modulation of the charge carrier density in this conducting channel. This modulation of the carrier density in the conducting channel is read by two other electrodes, source and drain. A field-effect transistor (FET) is thus a three electrode unipolar device that allows one to monitor, through the gate bias, the conductance of a thin channel at the semiconductor–insulator interface. The critical parameters of thin-film transistors (TFTs) are the channel width, *W*, the channel length, *L*, and the capacitance per unit area of the insulator layer of thickness [18,19]. In a traditional OFET, the charge carriers travel in the channel parallel to the surface of the gate dielectric. The source and drain electrodes provide access to the channel and are engineered to inject charge well under all bias conditions. It is the gap between the source and drain–the channel–whose conductance is switched, adjusting the conductance of the whole device. The gate is separated from the channel by the gate dielectric forming a capacitor to the channel charge sheet. It is the gate dielectric that allows the creation of a field across the semiconductor and the resultant accumulation and depletion of carriers without the need for a DC current [19,20].

The goal of the present study is to produce graphene oxide based thin flim transistor and investigate of their properties by using obtained the graphene oxide (GO). For this aim, it has been chosen Ag/pentacene/poly-4-vinylphenol (PVP) and poly(methyl methacrylate) (PMMA)/GO/SiO_2/Si/Al structures. The electrical measurement of GO-TFT was analyzed by output and transfer characteristics by using Keithley-4200 semiconductor characterization system (SCS).

2. Materials and methods

Graphite powders were bought from the company "Sigma–Adrich". The graphene oxide (GO) particles were synthesized by a modified Hummers method [21] using expandable graphite powders as the starting material. Graphite (0.5 g) was mixed with 0.5 g of $NaNO_3$ and 23 mL of concentrated H_2SO_4. The mixture was cooled down to 0 °C in an ice bath and stirred for 2 h. Then, 3.0 g of $KMNO_4$ was added slowly (temperature was maintained at <5 °C throughout the mixing), and continuously stirred for another hour. The cooling bath was then removed and the mixture was cooled down to room temperature. To this, ~100 mL of distilled water was added and the temperature was increased to 90 °C. After reaching 90 °C, 150 mL of water was added again and continuously stirred for another hour and a half. This mixture was then treated with 10 mL of 35% H_2O_2 and ~1 L of hot water was added and diluted. The mixture was further washed with excess water and 5% HCl aqueous solution until the pH of the filtrate was nearly neutral, finally it was dried at 60 °C in vacuum oven for 48 h to yield the graphene oxide (GO) powder.

To make a monolayer of graphene oxide (GO) nanosheets on *p*-SiO_2 (300 nm), a well dispersed solution of powder graphene oxide in deionised water having concentration of 0.4 mg/mL was prepared, which was spin coated at 3000 rpm for 40 s. This is followed by annealing the film at 80 °C for 15 min in air. In the case of 0.4 mg/mL GO solution, the total coverage of GO nanosheets was around 75% with the average sheet size of about 500 nm with a step height of 1.2 nm indicative of single GO nanosheets [22]. The insulator layers PMMA is deposited from a toluene solution having the concentration of 10 mg/ml and spin coated at 3000 rpm for 40 s. The film is then annealed at 120 °C for 15 min in the nitrogen atmosphere. To deposit PVP films, a 1 wt.% solution of PVP with a 0.5 wt.% crosslinking agent in propylene glycol methyl ether acetate (PGMEA) was spin coated at 3000 rpm for 40 s followed by annealing in vacuum at 200 °C for 2 h. The thickness of PMMA and PVP thus obtained were nearly 10 nm. About 50 nm thick pentacene (Aldrich) active layer was deposited by thermal evaporation at 10^{-7} Torr with the substrates held at room temperature. Finally, 50 nm thick silver source and drain electrodes were defined with a shadow mask (W = 2000 μm, L = 100 μm) by thermal evaporation at a pressure of 10^{-7} Torr. Finally, graphene oxide (GO) based organic thin film transistor, Ag/pentacene/PVP/PMMA/GO/SiO_2/Si/Al structures was obtained by a spin coating and thermal evaporation technique.

The structures of graphene oxide were examined by XRD using Philips X'Pert PRO diffractometer with CuK_α radiation (λ = 0.154 nm) at 40 kV and 30 mA. The UV–vis spectroscopy was studied using spectrophotometer (SHIMADZU-UV-3600). FT-IR spectroscopy measurement was conducted in FT-IR spectrometer (model: Bruker IFS 66/S). The electrical characterization of graphene oxide based thin film transistors were measured by Keithley-4200 semiconductor characterization system (SCS).

3. Results and discussion

Fig. 1 presents the X-ray diffraction patterns of graphene oxide nanoparticles by using modified Hummers method. The insert shows XRD patterns of graphite in Fig. 1 shows that the distinguishable (0 0 2) peak of graphite at 26.56 has an interlayer distance of 0.334 nm (0 0 2). This implies that graphite is a highly oriented carbon material. In the XRD pattern of GO, the strong and sharp peak at 2θ = 11.7° corresponds to an interlayer distance of 0.76 nm (0 0 2). The increase in interlayer distance of GO is due to the existence of oxygen functional groups and some other structural defects [23]. Fig. 2 shows the UV–vis spectra of graphene oxide (GO). The spectrum of GO has an absorption peak at about 223 nm which is attributed to π–π* transition of aromatic C—C bonds. FT-IR measurement was employed to investigate the bonding interactions in graphene before and after the oxidation process. In Fig. 3, the results are shown that a broad absorption bands at 1730 C=O (carbonyl/carboxy), 1616 C=C (aromatics), 1048 C—O (alkoxy) for GO

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