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Effect of Graphite Oxide Solution Concentration on the Properties of Multilayer Graphene

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Abstract. This paper reports the influence of graphite oxide (GO) solution concentration on the optical and electrical properties of multilayer graphene (MLG) films. Graphene oxide (GrO) films were deposited on the glass substrates by spin coating aqueous solutions of GO with different concentrations (7, 8, 9, 10 and 11 mg/ml). The GrO films were then thermally reduced at temperature of 500^{0}C in argon flow for half an hour to form MLG films. Both the transmittance and sheet resistance decreased with the GO concentration from 8 mg/ml to 9 mg/ml, possibly due to thicker and uniform coverage of MLG over the substrate. However, the transmittance and sheet resistance increased rapidly as the GO concentration reached 11 mg/ml, which can be attributed to poor film quality. The MLG film obtained at concentration of 10 mg/ml showed the highest transmittance/sheet resistance ratio with 69 % transmittance and sheet resistance of 292 ± 63 kΩ/sq. The optimum MLG film was utilized as counter electrode in dye sensitized solar cells based on ZnO nanorods.

Keywords: Dye sensitized solar cells; sheet resistance; transmittance.
PACS: 68.65.Pq; 78.67.Wj; 81.05.uf; 81.20.Ka

INTRODUCTION

For the past two decades, graphene has been attracting attention due to its good electrical property and high optical transmission [1-5]. Graphene as a monolayer of carbon in two dimensional honeycomb lattices can be easily prepared by reduction of graphene oxide (GrO) [6-8]. Hummers method is a famous, low cost and simple technique to obtain large quantity of graphite oxide (GO) which can be exfoliated to form GrO film [9].

It is well known that both sheet resistance and transmittance of graphene film decrease with film thickness. Weber at al. reported that the transmittance of 24 nm-thick graphene film at 300 nm is ~55 % which is much lower than that of 8 nm-thick graphene film (~80 %) [10]. Meanwhile, graphene film should be made thicker to minimize the boundaries between individual graphene sheets which normally result in higher sheet resistance [11]. The thickness of the graphene film could be controlled by manipulating the GO solution concentration or coating cycle [12]. This present work reports the effect of GO solution concentration (7, 8, 9, 10 and 11 mg/ml) on the optical and electrical properties of multilayer graphene (MLG) films. The highest transmittance to sheet resistance ratio of MLG film was obtained at GO concentration of 10 mg/ml. The corresponding MLG film has been successfully applied as transparent counter electrode in dye sensitized solar cell (DSSC).

EXPERIMENTAL

The procedures to produce GO flake using modified Hummers method were described elsewhere [13]. The GO flakes were dissolved in deionized water at various concentrations of 7, 8, 9, 10, and 11 mg/ml by sonication for 20 min followed by stirring for 2 hours. Then, the resultant solution was spin-coated on the glass substrates to form GrO films. Next, the GrO films were annealed at 500^{o}C in argon flow for half an hour to form MLG films.

Halo DB-20 UV-Vis spectrophotometer was used to investigate the optical transmission of MLG films. The sheet resistance of MLG films was characterized by four point probe measurement using Keithley 2401 source

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meter. Lastly, DSSC consisted of ZnO nanorod arrays as photoanode and optimum MLG as counter electrode with an iodine electrolyte was fabricated as shown in FIGURE 1. The ZnO nanorod arrays were grown on FTO glass substrates following the method described elsewhere [14-15]. The DSSC with active area of 0.23 cm^2 was characterized by current–voltage measurement under 100 mW/cm2 simulated AM 1.5 G sunlight using Keithley 237 source measurement unit.

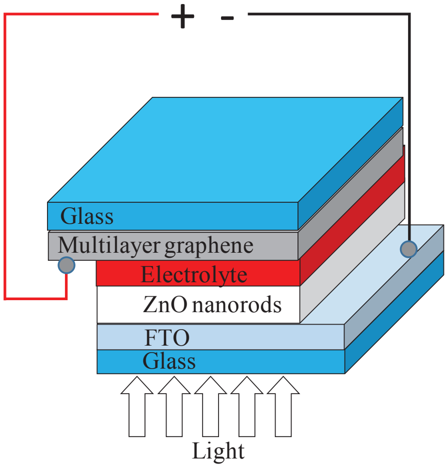

FIGURE 1. The DSSC with ZnO nanorods/electrolyte/MLG structure.

RESULTS AND DISCUSSION

The optical transmission spectra of MLG films prepared at different GO concentrations are shown in FIGURE 2. It can be clearly seen that the transmittance decreased significantly from 69-71 % (λ=550 nm) to 62 % when the GO concentration increased from 7-8 mg/ml to 9 mg/ml. This can be explained by the formation of thicker MLG film and uniform coverage of MLG over the substrate. However, the transmittance started to increase as the GO concentration further increased to 10 and 11 mg/ml due to poor film quality. The crack and void can be clearly observed from the photograph of the corresponding MLG films deposited from concentrated solution (FIGURE 3).

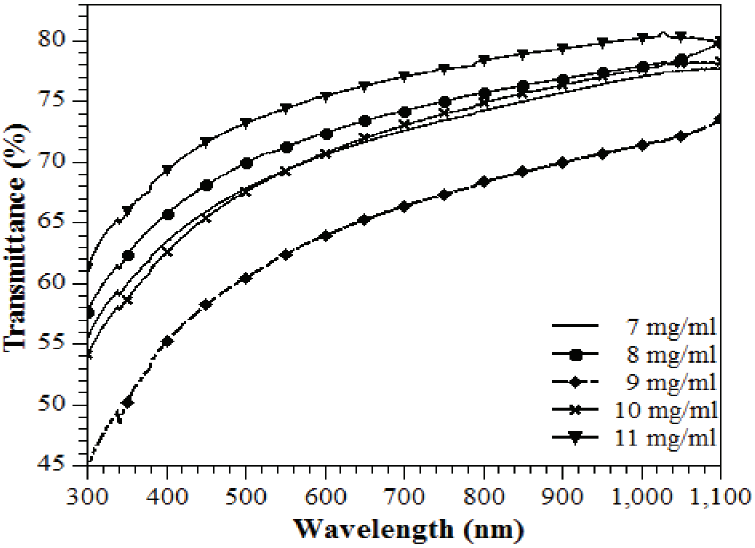

FIGURE 2. The optical transmission spectra of MLG films prepared at different GO concentrations.

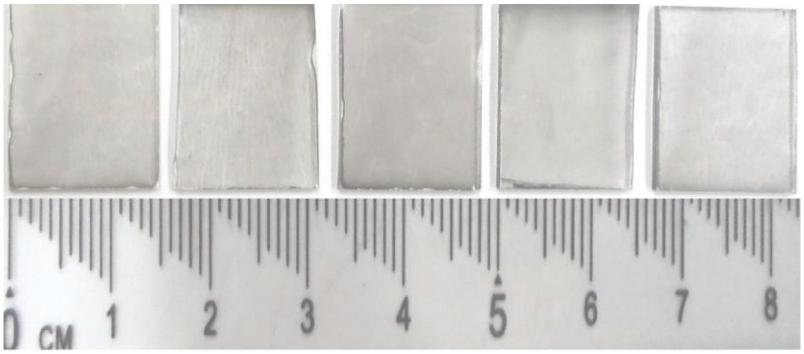

FIGURE 3. The photograph of MLG films prepared at GO concentrations of 7, 8, 9, 10, and 11 mg/ml (from left to right).

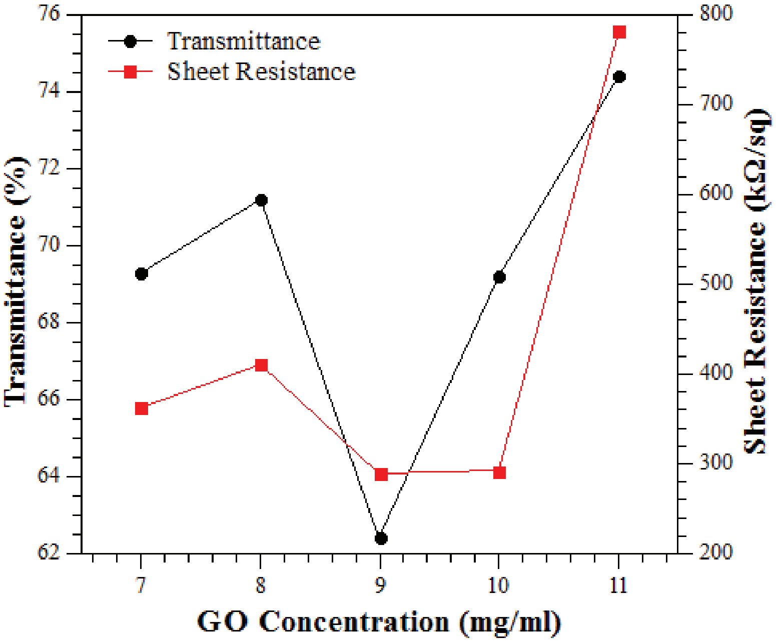

FIGURE 4. The transmittance and sheet resistance of MLG films as a function of GO concentration.

TABLE (1). The transmittance and sheet resistance of MLG films prepared at various GO concentrations

Concentration (mg/ml)	Transmittance (T, %)	Sheet Resistance (SR, kΩ/sq)	T/SR
7	69.3	362.14 ± 49.44	0.19
8	71.2	410.09 ± 35.69	0.17
9	62.4	289.79 ± 12.81	0.22
10	69.2	291.95 ± 63.07	0.24
11	74.4	781.96 ± 299.7	0.10

FIGURE 5 shows the current density–voltage (J–V) graph of DSSC constructed with 10 mg/ml MLG film as counter electrode in dark and under illumination of a simulated AM 1.5 G sunlight at 100 mW/cm^2. The DSSC showed power conversion efficiency (PCE) of 0.0027 % with short circuit current density (Jsc) of 26 μA/cm^2, open circuit voltage (Voc) of 0.42 V, and fill factor (FF) of 25 %. It should be noted that the present PCE was smaller than that of a similar DSSC with MLG as counter electrode reported recently because the substrate and thermal reduction temperature are different [13].

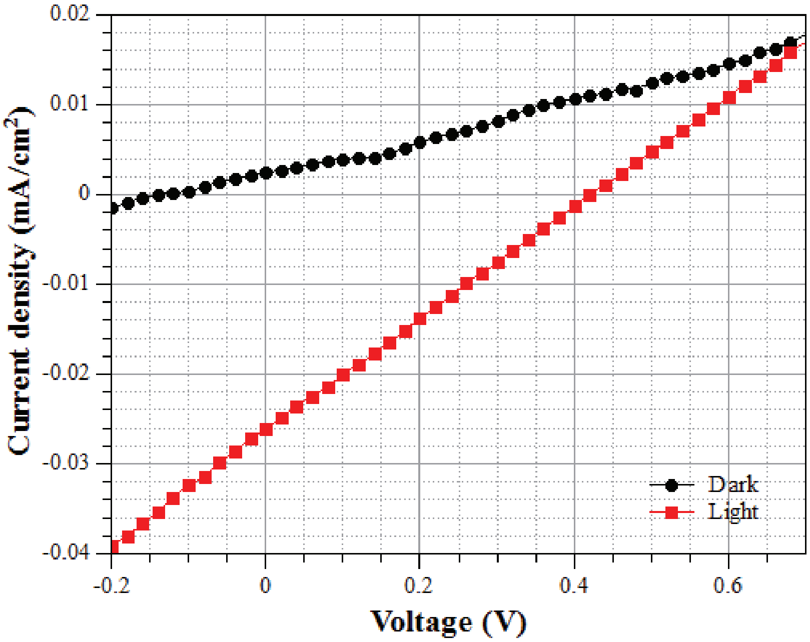

FIGURE 5. The J–V characteristic of DSSC in dark and under illumination.

CONCLUSION

The effect of GO solution concentration on the optical and electrical properties of MLG films has been investigated. The transmittance and sheet resistance decreased when the GO concentration increased from 8 mg/ml to 9 mg/ml as a result of formation of thicker MLG film and uniform coverage of MLG over the substrate. Further increase in GO concentration to 11 mg/ml results in higher transmittance and sheet resistance due to poor film quality. The MLG prepared at GO concentration of 10 mg/ml which showed the highest transmittance to sheet resistance ratio was studied as a transparent counter electrode in DSSC. The DSSC yielded Jsc of 26 µA/cm^2, Voc of 0.42 V, and FF of 25 %, resulting in PCE of 0.0027 %.

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