

Fabrication of Highly Conductive Graphene/ITO Transparent Bi-Film through Chemical Vapor Deposition (CVD) and Organic Additives-Free Sol-Gel Techniques

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Abstract : Indium tin oxide (ITO) remains the industrial standard transparent conducting oxides with better performances. Recently, graphene becomes as a strong material with unique properties to replace the ITO. However, graphene/ITO hybrid composite material is a newly born field in the electronic world. In this study, the graphene/ITO composite bi-film was synthesized by a two steps process. 10 wt.% tin-doped, ITO thin films were produced by an environmentally friendly aqueous sol-gel spin coating technique with economical salts of $\text{In}(\text{NO}_3)_3 \cdot \text{H}_2\text{O}$ and SnCl_4 without using organic additives. The wettability and surface free energy (97.6986 mJ/m^2) enhanced oxygen plasma treated glass substrates were used to form voids free continuous ITO film. The spin-coated samples were annealed at 600°C for 1 hour under low vacuum conditions to obtain crystallized, ITO film. The crystal structure and crystalline phases of ITO thin films were analyzed by X-ray diffraction (XRD) technique. The Scherrer equation was used to determine the crystallite size. Detailed information about chemical composition and elemental composition of the ITO film were determined by X-ray photoelectron spectroscopy (XPS) and energy dispersive X-ray spectroscopy (EDX) coupled with FE-SEM respectively. Graphene synthesis was done under chemical vapor deposition (CVD) method by using Cu foil at 1000°C for 1 min. The quality of the synthesized graphene was characterized by Raman spectroscopy (532nm excitation laser beam) and data was collected at room temperature and normal atmosphere. The surface and cross-sectional observation were done by using FE-SEM. The optical transmission and sheet resistance were measured by UV-Vis spectroscopy and four point probe head at room temperature respectively. Electrical properties were also measured by using V-I characteristics. XRD patterns reveal that the films contain the In_2O_3 phase only and exhibit the polycrystalline nature of the cubic structure with the main peak of (222) plane. The peak positions of $\text{In}_{3d5/2}$ (444.28 eV) and $\text{Sn}_{3d5/2}$ (486.7 eV) in XPS results indicated that indium and tin are in the oxide form only. The UV-visible transmittance shows 91.35 % at 550 nm with $5.88 \times 10^{-3} \Omega\text{cm}$ specific resistance. The G and 2D band in Raman spectroscopy of graphene appear at 1582.52 cm^{-1} and 2690.54 cm^{-1} respectively when the synthesized CVD graphene on SiO_2/Si . The determined intensity ratios of 2D to G (I_{2D}/I_G) and D to G (I_D/I_G) were 1.531 and 0.108 respectively. However, the above-mentioned G and 2D peaks appear at 1573.57 cm^{-1} and 2668.14 cm^{-1} respectively when the CVD graphene on the ITO coated glass, the positions of G and 2D peaks were red shifted by 8.948 cm^{-1} and 22.396 cm^{-1} respectively. This graphene/ITO bi-film shows modified electrical properties when compares with sol-gel derived ITO film. The reduction of sheet resistance in the bi-film was 12.03 % from the ITO film. Further, the fabricated graphene/ITO bi-film shows 88.66 % transmittance at 550 nm wavelength.

Keywords : chemical vapor deposition, graphene, ITO, Raman Spectroscopy, sol-gel

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