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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 In(NO3)3.H2O and SnCl4 without using organic additives. The wettability and surface free energy (97.6986 mJ/m2) enhanced oxygen plasma treated glass substrates were used to form voids free continuous ITO film. The spin-coated samples were annealed at 600 0C for 1 hour under low vacuum conditions to obtained 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 0C 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 In2O3 phase only and exhibit the polycrystalline nature of the cubic structure with the main peak of (222) plane. The peak positions of In3d5/2 (444.28 eV) and Sn3d5/2 (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 x 10-3 Ωcm 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 SiO2/Si. The determined intensity ratios of 2D to G (I2D/IG) and D to G (ID/IG) 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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