Synthesis of high-quality graphene films by plasma chemical vapor deposition and doping process

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Abstract

Conventional transparent electrodes make use of indium tin oxide (ITO) and are commonly used in touch screens, flat panel displays and solar cells. Nearly 90% of ITO film market is for the touch screen application, which is expected to grow more and more in the future. Graphene is potential candidates for transparent conductive films for electrical and optoelectronic devices and various other applications due to its high electrical conductivity, chemical and physical stability. Graphene has been prepared by several methods, including precipitation on a silicon carbide surface, mechanical exfoliation from graphite, reduction of exfoliated graphene oxide, and growth by thermal chemical vapor deposition (CVD) on catalytic metal surfaces [1]. Bae et al, reported the synthesis of graphene by thermal CVD on a copper substrate at high deposition temperature of 1000 degree C and fabrication of transparent graphene electrodes [2]. Recently, we reported the low-temperature (below 400 degree C) synthesis of large-area graphene transparent conductive films using a slot antenna type surface-wave plasma CVD [3]. But the high sheet resistance of its graphene was a big problem for the touch screen application. We succeeded the improvement of the electrical conductivity of graphene by doping with HNO3. A 33μm-thick copper foil with A4 (211 mm X 297 mm) size was used as substrate. The substrate temperature was about 400 degree C. Total microwave power is 18 kW. The pressure of CH4, H2 and Ar atmosphere was 3 Pa. Few-layer graphene was deposited on the copper foil for 2 minutes. The transfer of the graphene films to a desired target substrate is enabled by the wet-etching of the underlying copper foil. This is carried out by treating the film with an aqueous (NH4)2S2O8 solution after a support material is covered on the graphene/copper surface, in our case a surface protective sheet (Nitto Denko Co.). The surface protective sheet is attached to the graphene/copper surface by using a film laminating roller with applying pressure. The result in a free-standing graphene/sheet film that can be handled easily and rinsed with deionized water to remove residual etchant. The graphene/sheet film is placed on the 100-μm thick polyethylene terephthalate (PET) substrate (graphene facing the surface). Finally, the surface protective sheet is removed from a sheet/graphene/PET film. We measured the transmittance and sheet resistance of the graphene/PET by using a haze meter (Nippon denshoku Co.) and four probe method, respectively. The transmittance was 93.6% (except PET substrate) and the sheet resistance was about 10000 ohm/square. Subsequently**,** graphene/PET film was p-doped with HNO3 to decrease its sheet resistance. After the graphene/PET film is dipped in 63 wt% HNO3 for 10 minutes, the sheet resistance is decreases to 2000 ohm/square and the transmittance is no change. By using the HNO3 doping, the electrical conductivity of graphene synthesized by surface-wave plasma CVD was improved substantially.

References

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