Direct correlation between work function and oxygen concentration of Graphene Oxide - Reduced Graphene Oxide films on ITO

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Work function (WF) is a fundamental electronic property of any material and provides understanding of the relative position of the Fermi surface level. WF tuning of the contact electrodes is a key requirement in several device technologies, including organic photovoltaics (OPVs), organic light-emitting diodes (OLEDs), and complementary metal oxide semiconductor (CMOS) transistors [1]. Work function of grapheme and its derivatives and the ability to control its value is a very important factor in applying these as an electrode material. Very recently, interest in a thin film of graphene oxide (GO) and reduced GO as an efficient hole transporting layer (HTL) for high-performance polymer solar cells (PSCs) has been emerged [2].

The presence of carboxylic acid, phenolic-hydroxyl and epoxide groups on graphene oxide allows them to be stably dispersed either in water or polar organic solvents. In addition, the oxygen-containing groups attached onto the graphene surface disrupt the conjugation of graphitic lattice, create a lot of structural defects on graphene and result in poor electrical properties. Therefore, the post-treatment that leads to removal of oxygen-based moieties, either by thermal or by chemical treatment, may partially recover its parent electrical properties. The conductivity, mobility and the oxygen content of GO and reduced GO chemically and thermally treated as well as a combination of both treatments was investigated [3,4]. In the case of certain chemical reduction protocols, additional heteroatoms have been shown to decorate the graphitic surface. The type and concentration of such functionalities have a decisive influence on the WF of graphene. The latter parameter can be controlled by varying the oxygen concentration which is dependent on the rGO thickness and the reduction process at different temperatures [1].

The aim of this work is to correlate the absolute WF value with the oxygen functionalities in a thin GO and rGO films. The GO film was deposited on ITO substrate, reduced with thermal treatment (heating in ultra high vacuum), chemical treatment or a combination of chemical and thermal process in vacuum. In order to investigate the effect of GO synthetic strategy , two different GO synthetic protocols which lead to differences in flake size and oxygen content were investigated. Additionally, the effect of GO thickness was also investigated. For this reason, two different thicknesses studied, ~4 and ~7nm, which are the most commonly used in devices [5]. To our knowledge this is the first experimental work where a direct correlation of oxygen functionalities in GO and rGO films with the WF values was performed in relation with: (a) different reduction methods frequently reported in literature [6], (b) film thickness and (c) GO synthesis. In order to correlate the WF with oxygen content and oxygen functionalities X-ray and Ultraviolet Photoelectron Spectroscopies (XPS/UPS) used in each reduction step.

The results showed that, as the oxygen content decreases upon thermal reduction of GO/ITO films, the work function decreases up to ~1eV. The combination of both chemical and subsequent thermal reduction leads to reduction of WF at even lower values depending on the presence of heteroatoms on the surface.

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