



Moderately reduced graphene oxide via UV-ozone treatment as hole transport layer for high efficiency organic solar cells

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ABSTRACT

This work demonstrates a new hole transport layer (HTL) via UV-ozone assisted moderate reduction of graphene oxide (GO) in organic solar cells (OSCs). The XPS and Raman measurements confirmed the successful reduction/removal of oxygen functional groups after UV-ozone treatment. Remarkably enhanced performance of the OSCs was demonstrated by the 15 min treated device with power conversion efficiency (PCE) of 4.07%, followed by the 10 min device with almost similar PCE of 4.03%, as compared to the device with untreated GO HTL exhibiting a much lower PCE of 2.80%. The significant enhancement in the device performance is attributed to the moderate reduction of oxygen functional groups, leaving behind highly conductive graphene as well as the decreased series resistance, increased shunt resistance and comparatively smooth HTL morphology. Further, it is also ascribed to enhanced short circuit current density, open circuit voltage and fill factor of UV-ozone treated GO which directly contributed to the improved PCE. Moreover, the OSCs with the UV-ozone treated GO HTL showed very good stability over the 250 h of exposure to ambient atmosphere.

1. Introduction

Organic solar cells (OSCs) have been the subject of tremendous research over the recent years due to their facile processing advantages [1]. The OSCs functional layers can be deposited by solution processed techniques, which allow low-cost, lightweight, flexible and large area (roll to roll) production [2–4]. Bulk heterojunction (BHJ) OSCs are one of the material systems extensively explored, where a random blend of a conjugated polymer (donor) and a small molecule fullerene (acceptor) form the photoactive layer, which is sandwiched between the conductive indium tin oxide (ITO) anode and a low work-function (WF) metallic cathode [5,6]. The devices with BHJ architecture have possessed power conversion efficiencies (PCEs) of more than 10% in recent years [7,8], but a significant amount of work is required to translate the lab-scale progress into the large scale industrial production [9,10].

Recently, interface engineering of OSCs has been receiving significant attention, and the buffer layer at the anode-photoactive interface is introduced as an integral part of the device structure [11]. It possesses electron blocking properties, reduces leakage current,

improves carrier mobility, reduces contact resistance, improves Ohmic contacts, suppresses carrier recombination as well as increases selectivity of the charge carriers to maximize the extraction of holes towards the anode in OSCs for higher performance [12–14]. Poly (3, 4-ethylenedioxythiophene): poly (styrenesulfonate) or (PEDOT:PSS) is one of the most widely used hole transport layer (HTL) in OSCs due to its high conductivity, high transparency and solution processability [15,16]. However, it limits the device lifetime and performance, owing to its hygroscopicity and inability to block electrons [17,18]. To overcome these inherent instabilities associated with PEDOT:PSS, various types of materials have been employed to function as HTLs in BHJ OSCs, mainly focusing on inorganic semiconductor metal oxides [19–21]. However, the deposition of oxides is usually carried out by cost-intensive vacuum techniques which ultimately makes them incompatible with cost-effective, solution-processable and large scale production [16,22]. Solution-processable metal oxides have been reported to serve as HTLs in BHJ OSCs, but the high annealing temperatures (typically higher than 200 °C) for the achievement of fully stoichiometric metal oxides restricts their applications [23–25].

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Therefore, development of cost-effective, solution-processable and large scale production modules is urgently demanded.

In this context, solution processable carbonaceous materials such as graphene oxide (GO) and reduced-GO (r-GO) are promising class of candidates with their high conductivity, good structural stability and tunable functionality [22]. In case of GO, it can be obtained by a simple sonication of graphite oxide [26], however, it possess insulating characteristics due to substantial sp^3 fraction, as the carbon atoms bonded with oxygen are mostly sp^3 hybridized [27], consequently, it disrupts the sp^2 conjugation of the hexagonal graphene lattice [28]. The cell performance with GO HTL is highly thickness dependent due to its insulating properties [29], and therefore the interests in r-GO have been increased in recent years [30]. Jung et al. [31] and Yingdong et al. [32] reported that removal of oxygen in GO leads to transition of GO from an electrical insulator to semiconductor and ultimately to a graphene-like semimetal, which can effectively compliment the drawbacks of GO. The r-GO can be used as an efficient HTL by the chemical processing using the toxic reagents such as hydrazine and the solvothermal processing that normally uses harsh solvents such as N,N-dimethylformamide or N-methyl-2-pyrrolidinone [33–35], however, reduction of GO by these methods is inadequate for mass production; toxicity of chemical reducing agent and multiple-steps or even ultra-high vacuum and high-temperature (up to 1050 °C) approaches under argon and dihydrogen in case of solid heating reduction [36,37] are inappropriate to the simple and cost-efficient production of OSCs on various substrates [29]. Therefore, facile low cost and low temperature methods such as UV-ozone reduction of GO are highly desirable.

To circumvent these processes, the present work reports a facile, low-cost and fast route to reduce oxygen content in GO by UV-ozone treatment of spin-coated GO HTLs in OSCs. Changes in the chemical composition, structural, morphological and optical properties were systemically investigated to better understand the effect of the UV-ozone treated GO HTLs on OSC performance and the relationship between the degree of changes in these properties due to UV-ozone treatment and the device performance. The UV-ozone treated GO based OSCs showed significantly enhanced photovoltaic performance, primarily due to the fact that the ozone molecules can partially reduce GO leading to highly conductive graphene. The treated films also possessed smoother surface morphology and as well as high transmittance.

2. Materials and methods

2.1. Materials

Pre-patterned ITO coated (Sheet resistance, 15 Ω /sq) glass substrates with the dimensions of 20 × 15 mm (six active pixels of 4.5 mm² on each) were purchased from Ossila Ltd., UK. Graphite flakes were obtained from Asbury Inc. (USA). Potassium permanganate (KMnO₄, > 99%), sulphuric acid (H₂SO₄, 98%), phosphoric acid (H₃PO₄, 98%), and hydrochloric acid (HCl, 35%) for GO synthesis, were obtained from R&M Chemicals, Malaysia. Both poly [N-9'-heptadecanyl-2,7 carbazole-alt-5,5-(4', 7'-di-2-thienyl-2',1,3'-othiadiazole)] or PCDTBT and (6,6)-Phenyl C71 butyric acid methyl ester or PC₇₁BM were purchased from Lumtec Corp. Taiwan.

2.2. Preparation of GO HTLs

The synthesis of GO can be seen in our previously reported work [38]. The GO solution was prepared by dissolving it in deionized (DI) water at an optimized concentration of 1 mg/mL. The solution was ultrasonicated for few minutes and continuously stirred at room temperature for several hours to get a homogeneous mixture. The freshly prepared GO solution was spin-coated on cleaned, pre-patterned ITO/glass substrates at 6000 rpm for 60 s. After the spin-casting process, the films were further treated with UV-ozone for 0, 5, 10 and 15 min.

2.3. Fabrication of PCDTBT:PC₇₁BM based solar cells

Normal architecture solar cells were fabricated on pre-patterned ITO coated glass substrates. Prior to the fabrication, the substrates were sequentially cleaned with detergent, ultrasonicated in DI water, acetone and isopropyl alcohol followed by oxygen plasma cleaning for 5 min. GO HTLs were deposited and treated as described above, before transferring to the nitrogen filled glovebox. The PCDTBT:PC₇₁BM blend solution was prepared by dissolving it in the ratio of 1:4 with a concentration of 10 mg/mL in chloroform for each and keeping under stirring overnight at room temperature. The prepared solution was filtered with 0.25 μ m PTFE filters (Whatman, Germany) prior to spin-casting. The photoactive layers were spun-cast at an optimized speed of 2000 rpm for 30 s. Further, the Aluminum (Al) electrodes were thermally evaporated in vacuum (10⁻⁶ Torr.) using a shadow mask to complete the normal architecture devices in ITO/GO/PCDTBT:PC₇₁BM/Al configuration. All the devices were encapsulated with glass covering the active area in the ambient air by using UV-curable epoxy before the characterization. A total of six fabrication cycles were repeated for each experimental condition to ensure the repeatability and reproducibility of the fabricated OSCs.

2.4. Photovoltaic characteristics of solar cells

The current density-voltage (*J-V*) characteristics of the devices were measured using a Keithley 236 (Keithley Co.) source measurement unit, in the dark and under an Air Mass 1.5 Global (AM 1.5 G) solar simulator with an irradiation intensity of 100 mW/cm². The light intensity was calibrated using silicon reference cell to reduce the spectral mismatch.

2.5. Characterization of UV-ozone treated GO interlayer

For the characterization of HTLs, GO films were replicated on ITO coated glass substrates using same parameters as described in section 2.2. The surface morphology of the films was characterized using field emission scanning electron microscopy (FESEM; JEOL JSM-7600F, Japan) at an operating voltage of 5 kV, and atomic force microscopy (AFM; SPM PROBE VT AFM XA 50/500, Omicron, Germany). Chemical changes on the surface of GO films due to UV-ozone treatment were analyzed using X-ray photoelectron spectroscopy (XPS; PHI 5000 VersaProbe II, Ulvac-PHI, USA) equipped with the monochromatized Al K α ($h\nu$ = 1486.6 eV) anode, in ultrahigh vacuum chamber ($\sim 10^{-10}$ mbar). Curve fittings for XPS core-level spectra were done using Multipack software (version 9, ULVAC-PHI, Inc.). Raman measurements were carried out using DXR Raman Microscope (Thermo Scientific, USA), by using green light excitation (532 nm) laser source with 6 mW power. The transmittance was recorded using Perkin Elmer UV-visible diffuse reflectance spectrophotometer (Lambda 650) in the range of 250–800 nm.

3. Results

This work demonstrates the possibility of moderate reduction of GO as an HTL for high efficiency solar cells by simple UV-ozone treatment of GO films. The GO is a derivative of the one-atom thick graphene functionalized with oxygen groups in the form of epoxy and hydroxyl groups on the basal plane and various other groups, such as carboxylic acid at the edges [39,40]. It consists of a mixed sp^2 - and sp^3 -hybridized carbon atoms. Particularly, manipulation of the size, shape and relative fraction of the sp^2 -hybridized domains of GO provides opportunities for tailoring its optoelectronic properties [16,41,42]. In case of GO HTLs, the efficiency is highly sensitive to the thickness of GO layers due to its insulating characteristics; therefore, the exposure of GO to the UV-ozone could potentially lead to the reduction of oxygen groups due to the fact that ozone molecules can moderately reduce GO which ultimately cause partial conversion of GO into highly conductive graphene.

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