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An efficient polymer solar cell using graphene oxide interface assembled via layer-by-layer deposition

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ABSTRACT

Graphene oxide (GO) is widely used as an interfacial material in applications such as organic light emitting diodes and photovoltaic devices. Herein we report a layer-by-layer (LbL) assembled GO thin film as an anode interfacial layer (AIL) for efficient polymer solar cells (PSCs). The GO thin film is fabricated by alternately depositing cationic polyelectrolyte poly(diallyldimethylammonium chloride) (PDDA) and GO on ITO/glass substrate, which possesses controllable thickness by adjusting LbL deposition frequency. The presence of ultrathin GO films improves the work function of ITO, leading to a better contact between the active layer and ITO anode. With the optimized number of deposition times, the efficiency of 6.04% for the PSC with PDDA-GO bilayer (GO-2) as the AIL was achieved.

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1. Introduction

Polymer solar cells (PSCs) have attracted worldwide attention because of their versatility for large-scale fabrication on flexible substrate at low temperature through roll-to-roll processing to achieve low cost and lightweight [\[1–3\]](#page--1-0). With the development of novel organic semiconductors and interfacial materials [\[4–6\],](#page--1-0) optimization of the morphology of active layer [\[7–9\]](#page--1-0) and device configurations [\[10,11\],](#page--1-0) and improvement of fabrication techniques [\[12\]](#page--1-0), the power conversion efficiency (PCE) of PSCs has steadily increased to 10% [\[13–16\]](#page--1-0). In a typical PSC, there is a large energy barrier between the active layer and the electrode, which is detrimental to the charge transport and collection. Therefore, an interfacial layer is vital to effectively minimize the energy barrier, selectively extract specific charge carriers and block the opposite ones, and consequentially improve the device performance [\[17\].](#page--1-0) The most commonly used anode interfacial layer (AIL), poly (styrene sulfonate)-doped poly (3,4-ethylenedioxy-thiophene) (PEDOT:PSS), suffers from poor stability due to its acidic and hygroscopic nature $[18]$. To overcome these deficiencies, a few metal oxides (MoO₃, V₂O₅, and WO₃) have been successfully employed as AILs to improve the anode contact [\[19,20\].](#page--1-0) However, fabrication of these oxides usually needs vacuum deposition, which is incompatible with the solution-processed PSCs.

Therefore, it is still of great significance to implement alternative interfacial materials for large-scale fabrication and practical application of PSCs.

Recently, the quasi two-dimensional graphene oxide and its derivatives have emerged as a new class of interfacial materials for PSCs due to its unique chemical and electronic structures, along with their solution processability [\[21,22\]](#page--1-0). The GO thin film prepared by spin-coating was shown preliminarily to be AIL in PSC [\[23,24\].](#page--1-0) It is demonstrated that the performance of PSCs is very sensitive to the thickness of GO thin films due to the insulating property of GO [\[24–26\].](#page--1-0) Therefore, it is essential to prepare thickness controllable GO thin film as AILs for PSCs. Although Langmuir–Blodgett (LB) assembly technique has been successfully employed to form thickness-controllable GO films as interlayers for PSCs [\[27\],](#page--1-0) the LB method suffers from the requirement for rather expensive instruments and has several limitations with respect to substrate size and is very sensitive to impurities. Compared to the methods mentioned above, layer-by-layer (LbL) assembly is one of the most versatile techniques for multilayer film fabrication with well-controlled structure and thickness [\[28\].](#page--1-0) Also, LbL technique can realize large area thin film fabrication under the condition of low solution concentrations. Over the last decade, a large number of multilayers composed of GO have been fabricated through LbL self-assembly and applied to transparent conducting films, field effect transistors, and lithium ion batteries [\[29\]](#page--1-0). To the best of our knowledge, the application of LbL assembled GO thin films in PSCs has not been reported yet.

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In this work, we demonstrate a direct LbL method to fabricate thickness controllable GO film, which is further applied as AIL for PSCs. A series of PSCs based on poly(3-hexylthiophene) (P3HT) and indene- C_{60} bisadduct (ICBA) integrated with LbL assembled GO films with different thickness as AILs are fabricated. Without an AIL, the device shows a poor efficiency of 2.97%. By inserting the thickness optimized GO thin film as the AIL, the work function of ITO improved, leading to a better contact between the active layer and ITO anode. Therefore, the power conversion efficiency (PCE) of PSC increases to 6.04%, which is comparable to that of the device with PEDOT:PSS. The results suggested that it is highly relevant for realizing photovoltaic (PV) devices by using LbL assembled GO film as interfacial layers.

2. Experimental

2.1. Layer-by-layer assembly of GO nanosheets

Polyelectrolyte poly(diallyldimethylammonium chloride) (PDDA) (M_w = 200,000–350,000 g mol⁻¹, 20 wt.% in H₂O) was purchased from Aldrich and diluted into water to a weight concentration of 0.2 wt.%. GO was purchased from Nanjing Jcnano Technology Co., and was suspended in water with sonication at a power of 200 W for 30 min. Then, GO aqueous dispersion was centrifuged at 10,000 rpm. GO solution with a concentration about \sim 1 mg mL $^{-1}$ was obtained. Substrates of ITO/glass slides were cleaned by ultraviolet-ozone (UV- O_3) to remove any organic contamination and introduce hydrophilic surface. To assemble GO on the substrates, ITO were first immersed in a 0.2 wt.% PDDA solution for 20 min to get a positively charged surface, then rinsed thoroughly with ultra-pure water and dried in a nitrogen gas flow. Then the PDDA pre-coated substrates were immersed in GO solution (\sim 1 mg mL⁻¹) for another 20 min, followed by the same rinsing step. Then, one bilayer of PDDA-GO film (GO-1) was obtained. The above procedures were repeated to achieve the desired number of bilayers (GO-n).

2.2. Device fabrication

Polymer solar cells were fabricated on ITO/glass substrates with a sheet resistance less than 10 Ω sq $^{-1}$. ITO substrates were cleaned via sonication in acetone, isopropyl alcohol and ethanol sequentially, followed by UV-O₃ treatment for 15 min prior to thin film deposition. PEDOT:PSS (Clevios 4083) was spin-coated onto the cleaned substrates at 4500 rpm for 30 s (thickness \sim 45 nm), and baked on a hot plate at 150 \degree C for 15 min in air. The as prepared LbL GO films were annealed on a hot plate at $120 \degree C$ for 15 min in air, then treated with UV-O₃ for 4 min. A mixed solution of P3HT and ICBA with 1:1 weight ratio and a total concentration of $40 \:\rm{mg} \: \rm{m} \rm{L}^{-1}$ in ortho-dichlorobenzene (o-DCB) was spin-casted at

1000 rpm for 30 s either on PEDOT:PSS or on GO films under nitrogen atmosphere, then annealed at 150 \degree C for 10 min. Finally, the top electrodes, 20 nm Ca and 100 nm Al films, were thermally evaporated with a shadow mask at a base pressure below 5×10^{-4} Pa. The effective area of the devices was 0.066 cm².

2.3. Measurement and characterization

Current density–voltage (J–V) characteristics were performed using a computer controlled Keithley 2400 source measure unit under the illumination intensity of 100 mW cm^{-2} (AAA Oriel solar simulator with AM 1.5G filter). External quantum efficiency (EQE) was characterized on the QTest Station 2000ADI system (Crowntech. Inc.). The UV–Vis transmission spectra were recorded on a Cary 5000 UV–Vis–NIR spectrophotometer. Atomic force microscopy (AFM) topography images and CPD images were obtained by using a Multimode Nanoscope III–D atomic force microscope (Bruker) in the tapping mode. The thickness of films was determined by the Bruker 150 surface profiler. Raman scattering spectra were recorded on an Acton Raman spectrometer with 532 nm semiconductor laser as excitation source.

3. Results and discussion

Fig. 1 shows the schematic diagram for the process of LbL assembly of cationic polyelectrolyte PDDA and GO on ITO substrate. The atomic force microscopy (AFM) topography image in Fig. 1 shows the morphology of the spin-coated GO nanosheets on mica substrate, indicating that they are mostly single layer with a thickness of \sim 1 nm and lateral size ranges from hundreds of nanometers to several micrometers. To deposit GO on ITO substrate by LbL electrostatic assembly technique, the substrate was firstly treated with UV- O_3 to introduce hydrophilic surface. Then, positively charged PDDA and negatively charged GO nanosheets were alternately assembled on ITO substrates.

AFM was used to investigate the deposition of graphene oxide nanosheets and the morphology of the multilayer films. [Fig. 2a](#page--1-0)–d shows the AFM topography images of bare ITO, 1-bilayer (GO-1), 2-bilayer (GO-2), and 4-bilayer (GO-4) PDDA-GO on ITO/glass substrates, respectively. The samples are measured after annealing on a hot plate for 15 min at 120 \degree C. The GO nanosheets are lying on the ITO/glass substrates, and some folds also formed according to the images. The thickness of the GO-n $(n = 1, 2, 4)$ films are about 2 nm, 4 nm, 9 nm as estimated by AFM height analysis. And the surface root-mean-square (RMS) roughness of the samples is 1.202 nm, 1.440 nm, 1.613 nm and 2.127 nm, respectively. The increasing deposition cycles enhance the surface roughness of the films due to the increasing amount of absorbed PDDA and GO nanosheets on substrates by electrostatic interaction. The optical transmission spectra of GO films with various thicknesses

Fig. 1. Device configuration of PSC with LbL assembled GO as the AIL. The left images show the AFM topography image of GO nanosheets on mica substrate and the schematic structure of GO and PDDA.

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