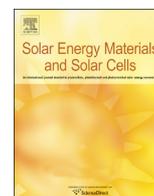




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Graphene quantum dots-incorporated cathode buffer for improvement of inverted polymer solar cells



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ABSTRACT

Graphene quantum dots (GQDs) are an emerging class of nanomaterials with unique photonic and electric properties. In this study, GQDs were prepared by a facile, inexpensive and high-yield hydrothermal method and were further used as a cathode buffer additive for inverted polymer solar cells due to a wide band gap (~ 3.3 eV) and well-matched energy level between GQDs–cesium carbonate (GQDs– Cs_2CO_3) modified indium tin oxide (3.8 eV) and high occupied molecular orbit of [6,6]-phenyl-C61-butyrac acid methyl ester (3.7 eV). In comparison to inverted polymer solar cells using cesium carbonate (Cs_2CO_3) buffer layer, the power conversion efficiency of GQDs– Cs_2CO_3 based device showed 22% enhancement from 2.59% to 3.17% as a result of enhanced exciton dissociation and suppressed free charge recombination at cathode/polymer active layer interface by GQDs. This work provides a new application of GQDs in organic electronic devices.

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1. Introduction

Solution processable bulk heterojunction (BHJ) polymer solar cells (PSCs) are considered as a promising alternative to conventional silicon based ones [1–3] because of their low cost of roll-to-roll production and flexible device applications [4–6]. Over the past years, tremendous efforts have been made on development of efficient and stable BHJ PSCs for higher power conversion efficiency. Small bandgap polymers with suitable band structures were synthesized to increase the visible light absorption and thus photocurrent [7,8]. New device structure and novel charge transport layer were rationally designed to enhance photo-generated charge carrier extraction from organic active layer to electrodes and increase stability of interfaces [9–12]. Among various PSCs, inverted PSCs have received growing research attention due to their superior device stability and manufacturing compatibility. In an inverted PSC, modification of indium tin oxide (ITO) with a buffer layer is needed to reduce ITO work function and enhance electron collection. The most commonly used cathode buffer in inverted BHJ PSCs is solution-processed ZnO or TiO_2 [13]. Very recently, Cs_2CO_3 became a promising cathode buffer because of its good electron injection and facile fabrication process [14–16]. However, the poor

hole blocking ability of the Cs_2CO_3 -modified ITO cathode significantly increases the recombination loss at the cathode/polymer active layer interface and reduces the overall power conversion efficiency.

Graphene quantum dots (GQDs) are a new class of nanomaterials, which have unique photonic and electronic properties and have been widely used in biological science [17], photocatalysis [18], light-emitting [19] and photovoltaic devices [20–22]. In photovoltaics, GQDs were used as electron acceptors in organic/inorganic hybrid solar cells and as sensitizers in nanocrystalline QD-sensitized solar cells, however the power conversion efficiencies of the respective solar cells are quite low.

In this work, we investigated GQDs as a cathode buffer additive in inverted PSCs, showing that GQDs can act as an excellent cathode buffer additive in inverted PSCs to increase the power conversion efficiency from 2.57% to 3.17% due to enhanced exciton dissociation and suppressed free charge recombination at the cathode/polymer active layer interface with the introduced GQDs in the buffer layer.

2. Experimental details

2.1. Preparation of GQDs

GQDs were synthesized using a hydrothermal method developed in the literature [23]. In a typical synthesis, 2 g of citric acid monohydrate (9.5 mmol) and 1 g of L-cysteine (8.3 mmol) were dissolved in 5 mL of deionized water. The solution was evaporated

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until dry at 70 °C for 12 h. The resulting viscous syrup was heated in a Teflon-lined stainless steel autoclave at 200 °C for 3 h. Finally, the black syrup product was collected and dissolved in 2-ethoxyethanol.

2.2. Fabrication of BHJ PSCs

All devices with the structure of ITO/cathode buffer (GQDs, Cs₂CO₃ or GQDs–Cs₂CO₃)/P3HT:PCBM/V₂O₅/Au were fabricated on pre-cleaned ITO patterned glass substrates. The cathode buffer was deposited on ITO substrates by spin-coating the GQDs (1 mg/ml), Cs₂CO₃ (1 or 2 mg/ml) or GQDs (2 mg/ml)–Cs₂CO₃ (2 mg/ml) (1:1) in 2-ethoxyethanol at 4000 rpm for 60 s. Following, the buffer layer coated ITO substrates were baked on a hotplate at 120 °C in air for 15 min. Afterwards, Poly(3-hexylthiophene-2,5-diyl) (P3HT, from Aldrich, regioregularity ≥90.0%) and [6,6]-Phenyl-C61-butyric acid methyl ester (PCBM), from Nano-C (99.5% purity) blend in 1:1 weight ratio at a P3HT concentration of 20 mg/ml in chlorobenzene was spin-coated on the baked ITO substrates at 700 rpm for 25 s (the active layer thickness was estimated to be ~180 nm), and dried in a covered Petri dish for 20 min. Subsequently, the vanadium oxide (V₂O₅) anode buffer was deposited on top of the polymer layer by spin-coating a vanadium precursor solution, vanadium oxytriisopropoxide in isopropanol (1:100 v/v), at 5000 rpm. The device fabrication was completed by thermal evaporation of 50 nm gold film on top of the V₂O₅ buffer in vacuum ($P < 1.5 \times 10^{-5}$ Pa) as anode electrode. The illumination area of each device is defined by the area of gold electrode which is ~7.06 mm². The as-prepared devices were then annealed at 150 °C for 20 min in vacuum to improve the crystallinity of the P3HT:PCBM blend.

2.3. GQDs and device characterization

The size and size distribution of GQDs was examined with atomic force microscopy (Nanoman, Veeco, Santa Barbara, CA) using tapping mode and transmission electron microscopy (HRTEM JEOL 2100). The

optical properties of GQDs were studied using a UV–vis spectrophotometer (UV-2450, Shimadzu). The photoluminescence (PL) of P3HT film was recorded using an LP920-KS instrument from Edinburgh Instruments, equipped with an iCCD camera from Andor and a photomultiplier tube. The work function of ITO electrodes modified with different cathode buffers was examined using ultraviolet photoelectron spectroscopy with the Helium line (21.2 eV).

The current density (J)–voltage (V) characteristics were recorded with a Keithley 2420 source meter in the dark and under illumination using a Sun 2000 solar simulator (Abet) with AM1.5 spectrum (100 mA/cm²). The intensity of the solar simulator was determined by a standard Si photovoltaic cell calibrated by the National Renewable Energy Laboratory. External Quantum Efficiency (EQE) measurements were performed without bias illumination with respect to a calibrated silicon diode. The EQE was calculated using $EQE(\lambda) = (hc/\lambda) \times (I(\lambda) - I_{\text{dark}}) / P(\lambda)$, where $I(\lambda)$ and I_{dark} are the short circuit current recorded under illumination and in the dark, respectively. No corrections were made for reflection from the glass substrate. The monochromatic light used for EQE measurement was supplied from xenon light, which passes through a Cornerstone monochromator. A chopper was placed after the monochromator and the signal was collected by Merlin lock-in radiometry after amplification through the current preamplifier. All measurements mentioned above were performed in air at room temperature without encapsulation.

3. Results and discussion

Fig. 1a shows the atomic force microscopy (AFM) image of GQDs distributed on mica substrate. The as-synthesized GQDs have sizes in a range from 0.5 to 3.5 nm with a low degree of aggregation, which is consistent with the TEM measurement. The chemical structural information of GQDs was examined with Raman spectroscopy. Two major features of D and G bands were observed at around 1380 and

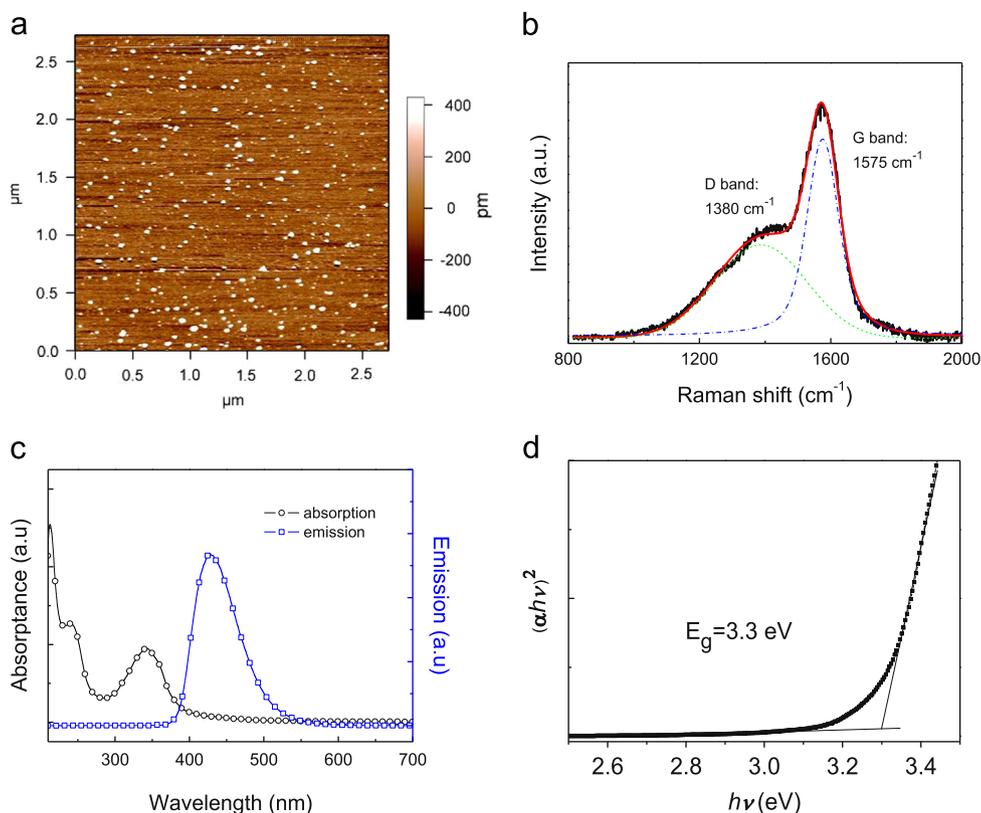


Fig. 1. AFM image (a), Raman (b), UV–vis absorption and photoemission spectra (c) of as-prepared GQDs and the corresponding Tauc plot (d).

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