Highly crystalline bilayer electron transport layer for efficient conjugated polymer solar cells

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ABSTRACT

Solution processed solar cells are a promising renewable energy technology due to the low fabrication costs. The most commonly used electron transport layer for solution processed organic solar cells is ZnO. However, sol-gel derived ZnO is amorphous, which limits interfacial charge transport. In this study, we demonstrate a ZnO bilayer, composed of a nanoparticle ZnO and sol-gel derived ZnO layer, as the electron transport layer in polymer solar cells incorporating the novel polymer poly[(5,6-difluoro-2,1,3-benzothiadiazol-4,7-diyl)-alt-(3,3″-di-(2-octyldodecyl)-2,2″; 5′,2′; 5″,2″-quaterthiophen-5,5″-diyl)] (PBBiBT4T-2OD). Compared with the single layer sol-gel ZnO, the bilayer displayed enhanced crystallinity. Consequently, the interfacial transport from the active layer was improved, as evidenced by dark J-V and PL spectroscopy measurements. Solar cells incorporating this bilayer ZnO layer achieved PCE values exceeding 10%, a relative improvement of 25% compared to the sol-gel ZnO devices.

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1. Introduction

Polymer bulk heterojunction (BHJ) solar cells possess multiple desirable properties, such as the potential for low-cost, high throughput manufacturing, which has caused significant interest in the technology [1]. The efficiency of these solution processed solar cells has increased rapidly, with many recent reports demonstrating power conversion efficiency (PCE) values higher than 10% [2–4]. Many different conjugated polymers, each with unique properties, have been explored as the electron donor material in polymer solar cells. Of these, one conjugated polymer, poly[(5,6-difluoro-2,1,3-benzothiadiazol-4,7-diyl)-alt-(3,3″-di-(2-octyldodecyl)-2,2″; 5′,2′; 5″,2″-quaterthiophen-5,5″-diyl)] (PBBiBT4T-2OD) has demonstrated high performance with a range of acceptor materials [5]. In particular, this polymer forms small, highly crystalline domains, which is conducive to excellent charge transport properties [3,5,6]. Typically, these devices are fabricated using an inverted device structure, whereby the photoactive layer is sandwiched in between a cathode buffer layer (hole blocking layer) and an anode buffer layer (electron blocking layer). These buffer layers play a significant role in the successful operation of the device [7,8].

Zinc oxide (ZnO) is one of the most commonly used electron transporting layer (ETL) in many solution-processed photovoltaic devices such as organic solar cells and perovskite solar cells, due to its high transparency in the visible region, wide bandgap of 3.3 eV [9–12], and simple processing techniques [13]. Multiple methods can be used to deposit the ZnO layer, of these, sol-gel processing has been widely studied. This technique allows for low temperature processing which makes it conducive to the roll-to-roll manufacturing ultimately desired for this technology [10,14,15]. However, sol-gel processing can lead to ZnO films with poorly crystallinity and a high density of trap states, which hinders interfacial charge transfer and device stability. Liang et al. reported that the morphology of sol-gel ZnO is heavily dependent on annealing temperatures [9]. When processed at temperatures below 200 °C, the ZnO film tends to be amorphous rather than crystalline, which significantly reduces electron mobility (4 × 10⁻³ - 2.0 × 10⁻⁴ cm² V⁻¹ s⁻¹) compared to that of crystalline ZnO (1–2 cm² V⁻¹ s⁻¹) [9,10,16,17]. Thus, high-temperature annealing or post-treatment of the ETL will be required to achieve high efficiency solar cells. By carefully controlling the processing temperatures from 130 to 200 °C, Sun et al. have reported a ca. 18% improvement in PCE [10]. In another report, Richardson et al. achieved a PCE of 8.32% for PTB7-polymer based solar cells by modifying the surface morphology of ZnO ETLs via different
annealing conditions [16]. These previous reports suggest that the sol-gel ZnO layer, although advantageous due to the low temperature processing, must be modified to improve the device PCE.

Here, we report a modified low temperature processing method for fabricating the ZnO layer in PFBT4T-2OD-based polymer solar cells. ZnO NPs were combined with a sol-gel derived ZnO layer to form a bilayer as the ETL. XRD measurements showed enhanced crystallinity in this bilayer, when compared to sol-gel derived ZnO. Additionally, the top ZnO layer resulted in a smoother surface, with fewer pinholes. These combined desirable effects resulted in improved interfacial charge transfer to the anode, leading to device efficiencies exceeding 10%.

2. Experimental section

2.1. Device fabrication

PFBT4T-2OD (high molecular weight) and PC71BM ([6,6]-Phenyl C71 butyric acid methyl ester) (high purity) were purchased from 1-Material. Zinc acetate dihydrate (purity ≥99.0%), Ethanolamine (purity ≥99.5%), 2-methoxyethanol (99.8% purity), ZnO dispersion (particle size < 130 nm) chlorobenzene (purity 99.8%), 1,2-dichlorobenzene (purity 99.9%), reagent alcohol (anhydrous, <0.003% water) and 1,8-diiodooctane were all purchased from Sigma-Aldrich. To prepare the ZnO precursor solution 108 mg of zinc acetate dihydrate was mixed in 1 mL of 2-methoxyethanol with 32 µL of ethanolamine, then stirred overnight at room temperature. For the nanoparticle ZnO precursor, the ZnO dispersion stock solution was diluted with reagent alcohol to achieve the desired concentration. The PFBT4T-2OD:PC71BM blend (1:1.2 wt%, 33 mg/ml) was dissolved in chlorobenzene:dichlorobenzene (1:1, v/v), with 3 vol% DIO and stirred for at least 4 h on a hotplate at 110 °C.

Devices were fabricated using an inverted architecture with the following structure: glass/ITO/sol-gel ZnO or NPs ZnO or bilayer ZnO/PFBT4T-2OD:PC71BM/MoO3/Ag. This device architecture is shown schematically in Fig. 1. The devices were built on patterned ITO substrates, purchased from Lumtec. All substrates were cleaned by ultrasonication in and ultrasonic bath using Hellmanex III, DI water, acetone and isopropanol. Cleaning with each solvent occurred for 15 min. Then, the ETL was deposited onto the cleaned ITO substrate by spin coating with different approaches: (i) Sol-gel ZnO solution was spin coated onto the ITO substrate at 5000 rpm and annealed in air at 120 °C for 10 min (iii). NPs ZnO solution was spin coated onto the ITO substrate at 5000 rpm and annealed in air at 120 °C for 10 min. Then, the bilayer ZnO layer was formed by spin coating Sol-gel ZnO on top of the NPs ZnO at 5000 rpm and annealing in air at 160 °C for 50 min. The average thickness for the resulting sol-gel ZnO, NPs ZnO and bilayer ZnO were ~34 nm, ~50 nm and ~72 nm, respectively.

Subsequently, a thick PFBT4T-2OD:PC71BM active layer (~350 nm) was deposited on different electron transport layers. Both the active layer solution and ITO substrate were preheated on a hot plate at a temperature of 110 °C for 5 min, which was required to allow for a uniform film formation. Then, the coated ITO substrate was annealed at 85 °C for 5 min inside a nitrogen glovebox. Detailed spin casting parameters are described in previous reports [3,5]. The inverted devices were completed by a thermal deposition of 11.5 nm thick MoO3 (hole transport layer) and 100 nm thick Ag (cathode electrode) at a vacuum level of 3 × 10⁻⁶ Torr. The effective device area for each cell was 0.13 cm².

2.2. Device characterization

The device performance was measured by a calibrated Keithley 2400 Source Meter under 100 mW cm⁻² (AM 1.5G) solar spectra illumination. The external quantum efficiency (EQE) was obtained from a spectral response equipment QEX10 (PV Measurements). The absorption spectrum of interfacial layers was measured using a UV–Vis–NIR spectrometer (Perkin Elmer lambda 950). X-ray diffraction (XRD) patterns were detected by an Empyrean MRD system, equipped with a CuKα radiation, with a step-size of 0.02°. Samples were prepared on polished silicon wafers. The X-ray wavelength used in the system is 0.154 nm. The surface morphology properties were investigated using Bruker dimension icon SPM equipment to capture atomic force microscopy (AFM) images operating in the tapping mode. A Carl Zeiss AURIGA Crossbeam was applied to acquire scanning electron microscopy (SEM) images.

**Fig. 1.** Schematic illustration of the inverted polymer solar cell device structure, ITO/ Sol-gel ZnO or NPs ZnO or Bilayer ZnO/PFBT4T-2OD:PC71BM/MoO3/Ag.

**Fig. 2.** XRD patterns of Sol-gel ZnO, NPs ZnO and Bilayer ZnO films grown from polished silicon wafers. The "(111)" peak corresponds to the ITO substrate. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)
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