



Reducing charge recombination of polymer solar cells by introducing composite anode buffer layer



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ARTICLE INFO

Keywords:

Interface engineering
Trap passivation
Hole transport layer
Defect state
Boundary topography

ABSTRACT

An interface engineering strategy is employed to reduce charge recombination in inverted polymer solar cells by simply modifying active layer with an electrolyte PEDOT:PSS solvent between photon-active layer and MoO₃ hole transfer layer. The incorporated PEDOT:PSS electrolyte can improve interfacial contact between PTB7:PC₇₁BM and electrode via establishing a composite PEDOT:PSS/MoO₃ hole transfer layer. The composite interface layer can optimize boundary topography and adjust the anode energy levels alignment, leading to a uniform and ordered hole transmission channel. The tailored boundary morphology eliminates the defect states and interfacial barrier, resulting in the simultaneously enhanced short-circuit current, open-circuit voltage, and fill factor (FF). These results demonstrate that PEDOT:PSS/MoO₃ composite layer owns a versatile potential using in high-performance organic photovoltaic devices.

1. Introduction

Over the past decade, much study has been devoted into the solution processed polymer solar cells (PSCs) as a promising alternative to traditional photovoltaic application (Li et al., 2005; Peet et al., 2007; Bronstein et al., 2011; Sun et al., 2012; Li et al., 2015; Chen et al., 2013; Guo et al., 2014; Li et al., 2012). Among them, the typical bulk heterojunction (BHJ) PSCs comprising an interpenetrating conjugated polymer (electron donor)/fullerene derivatives (electron acceptor) as photoactive layer have attracted extensively attention due to its unique merits of simply process, high power conversion efficiency (PCE), and long-term stability (Coffey et al., 2007; Vandewal et al., 2008; Liang et al., 2010; Brabec et al., 2010; Wang et al., 2011). However, the insufficient absorbed photons, unsatisfactory exciton migration and dissociation, charge transport, and charge recombination are hampering the improvement of PCE (Scharber et al., 2006; Li et al., 2007; Pivrikas et al., 2007; Chen et al., 2009). To reach the threshold value for mass commercial application, one widely adopted strategy is to develop novel photosensitive material. Particularly, new donor and acceptor materials with the high absorption overlapping the solar spectrum are related to the number of absorbed photons and photo-generated current, but this approach requires a complex functional design and technology process (Chen et al., 2009; Huang et al., 2009; He et al., 2010; He and Li, 2011). Another effective way to improve the device performance is to optimize the device structure and interface

engineering (Yu et al., 1995; Park et al., 2014; Holman et al., 2013; Xiang et al., 2013; Blum and Shaked, 2015; Su et al., 2012; Kim et al., 2007; Ma et al., 2005; Yang et al., 2005; Li et al., 2005). The heterogeneous physical contact of interface and the energy level offsets between donor (acceptor) and electrode usually produce traps and defects, which will result in a pronounced potential loss and current decrease (Chen et al., 2012; Graetzel et al., 2012). Morphology and interface control could increase the charge dissociation in a low band gap polymer:fullerene blend and decrease interfacial bimolecular recombination, leading to the charge transport and extraction promotion. Furthermore, incorporating nanocomposites, (Li et al., 2015; Li et al., 2016) solvent additives, (Lee et al., 2008) molecular doping (Li et al., 2017) are usually used to control the morphology of BHJ active layer. New interfacial layers and composite buffer layers between donor(acceptor) and electrodes have also been developed to improve device performance (Zhang et al., 2016; Li et al., 2016; Li et al., 2017; Li et al., 2017). Composite transport layer is considered as an effective way to improve thin film morphology and interfacial contact. Developing multiple layers by solvent treatment during the spin-coating process to reduce interfacial recombination and optimize boundary topography is an ongoing topic of research (Huynh et al., 2003; Moulé and Meerholz, 2009; Liu et al., 2012).

In this study, a composite PEDOT:PSS/MoO₃ hole transport layer is used to reduce interfacial recombination by solvent treating during the PTB7:PC₇₁BM phase forming. This multiplex electrolyte layer could not

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only facilitate the boundary topography transformation but also improve the interfacial contact with the anode electrode. The optimized boundary morphology and adjusted energy alignment could passivate traps, decrease the defects states, and boost charge carrier transport, leading to the reduced interfacial recombination. Consequently, the simultaneously enhanced short-circuit current (J_{sc}), open-circuit voltage (V_{oc}), and fill factor (FF) of 20.01 mA/cm², 0.743 V, and 64.58% were achieved, yielding an improved PCE of 9.60%.

2. Experimental section

The donor and acceptor materials of PTB7, PC₇₁BM were purchased from 1-Material Chemsctech without purification. PEDOT:PSS was brought from Xi'an p-OLED Corp. and used as received. Firstly, the commercial ITO glass was cleaned by detergent, deionized water, and isopropanol (IPA) in turn, then dried by nitrogen. Secondly, PEI was dissolved in water solvent and spin-coated onto the precleaned ITO substrate, and the devices were dried in an atmosphere at 100 °C for 15 min. Next, PEDOT:PSS and IPA were mutually dissolved in a certain proportion of 1 mL:8 mL, 1 mL:6 mL, 1 mL:5 mL, and 1 mL:4 mL, and stirred for 24 h in room temperature. The PTB7 and PC₇₁BM with the weight proportion of 10 mg/mL:15 mg/mL were mutually dissolved in a mixed solvent of chlorobenzene/1,8-dioctane (97:3% by volume) and spin-coated on the top of PEI at 1000 rpm for 2 min. At the end of the spin-coating, 350 μL PEDOT:PSS/IPA mixed solution was dropwise added on the top of PTB7:PC₇₁BM blend. Finally, a 4 nm MoO₃ layer and a 100 nm Ag film were subsequently evaporated through a shadow mask to form a top anode. The active area of the devices is about 4.4 mm². For the purpose of comparison, the control device consisting of ITO/PEI/active layer/MoO₃/Ag was also made, which is named as Type A. Meanwhile, the devices treated with different concentration of IPA:PEDOT:PSS were listed as Type B (1 mL:8 mL), Type C (1 mL:6 mL), Type D (1 mL:5 mL) and Type E (1 mL:4 mL), respectively. Fig. 1 shows the schematic of fabrication process of PSCs in our experiment.

3. Results and discussion

The device structure and the molecular structure of PC₇₁BM, PTB7 and PEDOT:PSS are presented in Fig. 2a, and the energy levels alignment of all the materials is shown in Fig. 2b. To explore the influence of the PEDOT:PSS/MoO₃ composite electrolyte interlayer on device performance, the current density-voltage curves (J–V) of PSCs are shown in Fig. 2c, and all the photovoltaic parameters of the control and optimized devices are summarized in Table 1 (more than 32 devices were made from over 8 individual experiments from 4 tests for each type of devices). For the control devices (Type A), an average PCE of 7.18 ± 0.19% (with V_{oc} of 0.724 V, J_{sc} of 17.18 mA cm⁻², FF of 58.09%, and maximum PCE value of 7.22%) was achieved, which is

analogous with the previous reports (Li et al., 2017; Liu et al., 2017). Device efficiency begins to increase when the PEDOT:PSS was incorporated into the cell. And a simultaneous enhancement of V_{oc} (0.743 V vs. 0.724 V), J_{sc} (20.01 mA cm⁻² vs. 17.18 mA cm⁻²), and FF (64.58% vs. 58.09%) was observed in the optimized devices with IPA:PEDOT:PSS of 1:5, resulting in a high PCE of 9.54 ± 0.11% (the highest value of 9.60%). The device efficiency tends to decrease with continuous increasing PEDOT:PSS concentration (more than 1:4) because the thickness of the surface are inappropriate.

In order to make a realistic evaluation on the apparent J_{sc} improvement of the devices, the absorption spectrum of the device was measured and shown in Fig. 3a. The modified devices show a higher absorption in the whole visible light spectrum, and the light-harvesting improves with the increased PEDOT:PSS concentration. The incident photon-to-electron conversion efficiency (IPCE) spectrum is shown in Fig. 3b. An increased IPCE in range of 350–700 nm can be observed for the optimized devices due to the improved charge collection of composite hole transport layer. The IPCE decreases when the PEDOT:PSS concentration is more than 1:4 because the charge transport would be damped by the thick buffer layer, which perfectly match the results of J–V characteristics. IPCE is decided by diffusion of the photo-generated excitons to the donor/acceptor interface, excitons separation and charge carriers collection at the electrodes. The modified donor/acceptor interpenetrating interface decreases the loss of excitons that is caused by trapping sites or ‘dead ends’, and the excitons separation is facilitated. The improved interfacial contact of active layer with MoO₃ layer reduces interfacial defects and fluctuation, and photo-generated charge carriers will have less chance to recombine in the interface before they reach the electrodes. Therefore, the modified devices own a higher IPCE. The improved absorption and IPCE spectra mean that more photons could be converted into charge carriers then collected by the electrodes, which resulted in the enhanced photocurrent (Peet et al., 2007; Kim et al., 2007). The integral current density of IPCE spectrum is included in Fig. 3b, which is consistent with J_{sc} .

The microscopic morphology of film was investigated by atomic force microscopy (AFM) to find the physical mechanism of improved performance. Fig. 4 presents the AFM image of PEDOT:PSS/PTB7:PC₇₁BM thin films. The thin PTB7:PC₇₁BM film without any treatment (Fig. 4a for Type A) exhibited rough and uneven, and part of the film assembled to form the hill and low-lying aggregation, which made against the balanced current carrier transport in the whole region. The surface of PTB7:PC₇₁BM thin films (Fig. 4b for Type B) became uniform after IPA:PEDOT:PSS (8:1) treating was carried out, and a small quantity of aggregation could be found. The surface of PTB7:PC₇₁BM films treated by IPA and PEDOT:PSS in the ratio of 6 mL:1 mL became more evenly distributed, and polymerization parts tended to be smaller, thus some micro-cracks appeared (Fig. 4c for Type C). After the ratio of IPA and PEDOT:PSS decreased to 5 mL:1 mL

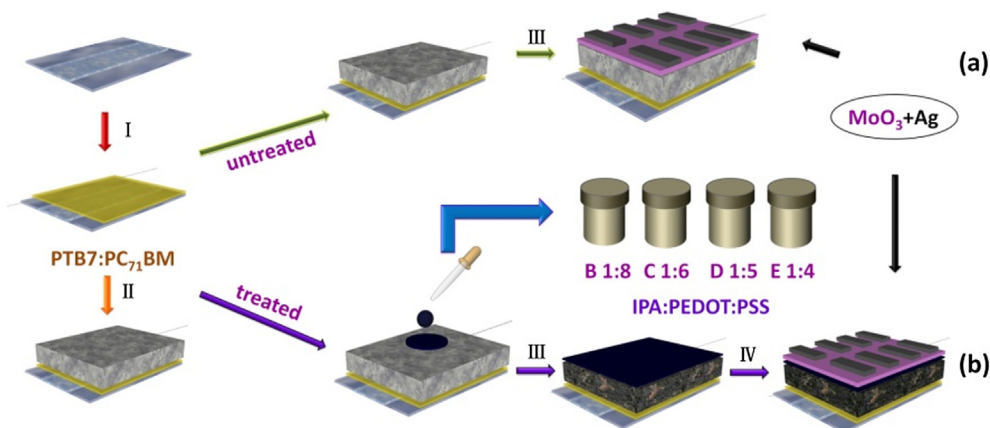


Fig. 1. Schematic of fabrication process of device.

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