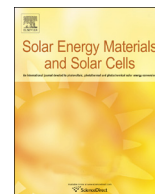




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Enhanced performance of polymer solar cells by dipole-assisted hole extraction

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ABSTRACT

Polymer solar cells (PSCs) with different anode buffer layers were fabricated to investigate dipole-assisted hole extraction on the performance improvement of PSCs. The power conversion efficiency (PCE) of PSCs was increased from 4.77% to 6.18% with 29.6% improvement due to the increased short circuit current density (J_{sc}) from 12.00 to 15.65 mA/cm² induced by the oriented LiF dipole-assisted hole extraction. More hole could be swept-out from the active layers by oriented LiF dipole/PEDOT:PSS combined anode buffer layers. The dipole-assisted hole extraction could be further demonstrated from the J - V curves of hole-only devices with different anode buffer layers.

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1. Introduction

Polymer solar cells (PSCs) have obtained extensive investigations and dramatic progress in the last ten years due to their huge potential application as the next green and renewable energy source [1,2]. The power conversion efficiency (PCE) of PSCs based on single bulk heterojunction (BHJ) has arrived ~8% [3,4]. Krebs et al. have put forward that the key 10–10 targets (10% PCE and 10 years of stability) of large-scale processed PSCs are threshold for its application as the next energy source [5–7]. Much effort has been carried out to reach the threshold, including enhancing exciton dissociation and carrier transport by incorporation of multiwall carbon nanotubes (MWCNTs) in the active layer [8–10], improving the charge carrier transport and optical field strength in the active layer by adding solvent additive [11], enlarging the optical absorption range by employing ternary BHJ active layer [12–14], and modifying the interfaces of electrode/active layer with appropriate interfacial materials [15–17]. Among the approaches, interfacial functional layers could serve some unique functions that mainly include: (i) tuning the energy level alignment or energetic barrier height at the electrode/active layer interface, (ii) defining polarity of electrodes and forming a selective collection for hole or electron, (iii) controlling surface properties to alter the morphology of

the active layer, (iv) acting as optical spacer or introducing plasmonic effects to modulate light absorption in the active layer, (v) improving interfacial stability by prohibiting a chemical or physical reaction between the active layer and electrodes [18–20].

The core function of interfacial buffer layer in PSCs is to increase charge carriers collection. Among the interfacial materials for hole collection, poly-(3,4-ethylenedioxythiophene)-poly-(styrenesulfonate) (PEDOT:PSS) is the most widely used due to its excellent performances: (i) forming Ohmic contact with the indium tin oxide (ITO) anode; (ii) tunable conductivity to reduce resistive losses by changing the doping ratios between PEDOT and PSS; (iii) high light transmittance in the Vis–NIR wavelengths [20,21]. For electron collection, Lithium fluoride (LiF) is widely used as cathode buffer layer with ultrathin thickness (~1 nm) due to its high electron injection or collection resulting from the tunneling effect induced by LiF dipole layer [22,23]. Recently, Hu et al. pointed out that the PCE of PSCs was increased by adapting PEDOT:PSS doped with methoxyethanol (ME) or dimethyl sulfoxide (DMSO) as anode buffer layer for the better hole collection [24]. It means that the purposely enhanced hole collection may be an effective method to improve the performance of PSCs. In this letter, the combined anode layers with LiF/PEDOT:PSS was designed to form the oriented LiF dipole array for further extracting hole from the active layer, the positive effect of dipole-assisted hole extraction was demonstrated from the improvement performance of PSCs and electric properties of hole-only devices with different anode buffer layers.

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2. Experimental section

Patterned indium tin oxide (ITO) glass substrates (sheet resistance $15 \Omega/\square$) were consecutively cleaned in ultrasonic baths containing glass lotion, de-ionized water, ethanol and then blow-dried by nitrogen. All pre-cleaned ITO substrates were treated by UV–ozone for 6 min for improving work function and cleaning the surface. PEDOT:PSS (Clevios AI 4083) films were prepared by spin-coating method at 3000 rounds per minute (rpm) with 50 s and annealed at 120°C for 10 min. Electron donor poly(diketopyrrolopyrrole-terthiophene) (PDPP3T, purchased from Organtec Materials Inc., Mn > 240k) and electron acceptor [6,6]-phenyl-C₇₁-butyric acid methyl ester (PC₇₁BM, purchased from Luminescence Technology Corp.) were co-dissolved in *o*-dichlorobenzene (*o*-DCB) at a concentration of 15 mg/ml with weight ratio of 1:2 to form hybrid solution. Solvent additive 1,8-diiodooctane (DIO) with 5% volume of *o*-DCB was added into the hybrid solution. The active layer of PDPP3T:PC₇₁BM films were fabricated by spin-coating method at 2500 rpm with 60 s. The 1 nm LiF and aluminum (Al) thin films were deposited under 10^{-4} Pa vacuum conditions. The active area is defined by the vertical overlap of ITO anode and Al cathode about 3.8 mm^2 .

The absorption spectra of films were measured with a Shimadzu UV-3101 PC spectrophotometer. The current density–voltage (*J*–*V*) characteristics of the PSCs were measured using a Keithley 4200 source measurement unit and an ABET Sun 2000 solar simulator with light power 100 mW/cm^2 under air conditions. The external quantum efficiency (EQE) spectra were measured by a Zolix Solar Cell Scan 100. The architecture of the PSCs and chemical structures of used materials are shown in Fig. 1.

3. Results and discussion

The absorption spectra of pristine PDPP3T, PC₇₁BM and PDPP3T:PC₇₁BM (wt/wt, 1:2) blend films were measured and are shown in Fig. 2. The electron donor PDPP3T material shows near-infrared absorption with an absorption peak at 853 nm and a shoulder peak at 770 nm. The highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels of PDPP3T are around 5.35 and 3.81 eV with a relative low band gap about 1.54 eV [25]. The electron acceptor PC₇₁BM has apparent absorption in the range from 350 to 700 nm, with the HOMO and LUMO energy levels of about 6.0 and 4.3 eV [26]. The absorption of

PDPP3T:PC₇₁BM blend film shows a wide and strong absorption in the range from 350 to 900 nm, which is beneficial to photon harvesting for performance improvement of PSCs. In addition, the LUMO–LUMO offset between PDPP3T and PC₇₁BM is about 0.49 eV (enough larger than the typical value 0.3 eV) for providing an effective driving force for exciton dissociation, as shown in Fig. 4 (a) [27]. It was known that LiF dipole could be formed in LiF ultrathin film due to the LiF molecule polarity. The oriented LiF dipole that F[−] directed towards the active layer and Li⁺ directed towards ITO anode are beneficial to hole extraction from the active layer [28,29]. However, the orientation of LiF dipole may be random during the thermal deposition process of LiF ultrathin film. In order to control LiF dipole orientation, ultrathin LiF layer was designedly prepared on PEDOT:PSS film or sandwiched by ITO and PEDOT:PSS films. A series of PSCs with different anode buffer layers were fabricated to investigate the effect of dipole-assisted hole extraction on the performance of PSCs.

The *J*–*V* characteristic curves of PSCs with different anode buffer layers are shown in Fig. 3. The key parameters of PSCs were summarized according to *J*–*V* curves and are listed in Table 1. The open circuit voltage (V_{oc}) of all PSCs was kept almost constant about 0.69 V, which is mainly determined by the energy barrier between the HOMO of donor and the LUMO of acceptor. It should be highlighted that the short circuit current density (J_{sc}) of PSCs with combined buffer layers (LiF/PEDOT:PSS or PEDOT:PSS/LiF)

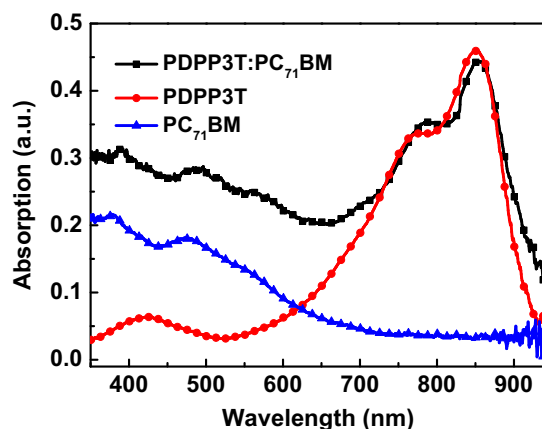


Fig. 2. The absorption spectra of pristine PDPP3T, PC₇₁BM and PDPP3T:PC₇₁BM blend films.

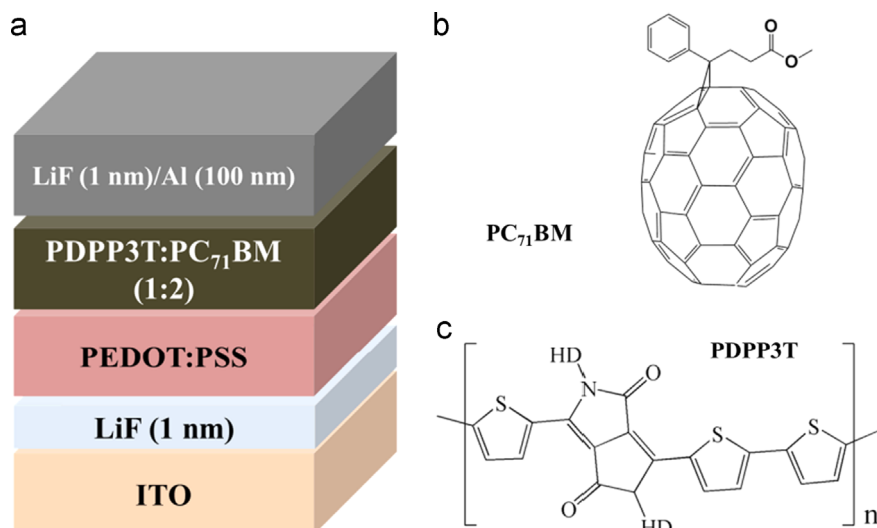


Fig. 1. (a) Device architecture of the PSCs; (b), (c) chemical structures of used materials PDPP3T and PC₇₁BM.

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