



Morphology-dependent charge recombination through localized states in polymer/polymer blend solar cells



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ABSTRACT

Polymer solar cells (PSCs) based on the PBDTTT-EFT (polymer donor):N2200 (polymer acceptor) blend have been fabricated with the active layer processed with various solvents including *o*-dichlorobenzene (*o*-DCB), chlorobenzene (CB), chloroform (CF), and CF:*p*-xylene (PX) mixed solvent. A highest power conversion efficiency (PCE) of 5.37% is achieved with the active layer processed with CF:PX mixed solvent. The dependence of short-circuit current density (J_{SC}) on incident light intensity indicates that the charge recombination is to some extent suppressed in the device with the active layer processed with the CF:PX mixed solvent and results in an enhanced J_{SC} . Morphology studies disclosed that the domains are preferential face-on orientation in the active layers processed with *o*-DCB, CB and CF, while it shows a combined face-on and edge-on orientation in CF:PX-processed film. The long-lived trap-assisted charge recombination originated from the active layer morphological variance has been focused on and investigated. And the nanosecond transient absorption experiment further demonstrated that the PBDTTT-EFT:N2200 film processed with *o*-DCB shows obvious long-lived trap-assisted charge recombination, while the trap-assisted charge recombination is effectively suppressed in the PBDTTT-EFT:N2200 film processed with CF:PX mixed solvent. This implies that deep localized traps correlated with the PBDTTT-EFT:N2200 morphology are reduced by using the CF:PX mixed solvent to process the active layer. In addition, the domains with a mixed orientation in the active layer may also enhance the three-dimensional charge transport and hence improve the charge-collection efficiency.

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1. Introduction

Bulk heterojunction (BHJ) polymer solar cells (PSCs) composed of conjugated polymer donor and fullerene derivative acceptor have achieved significant progresses by fine modification of polymer donors and interface engineering with the power conversion efficiencies (PCEs) over 10% in recent years [1–6]. Meanwhile, PSCs based on non-fullerene acceptors have attracted intensive research interest in recent years due to their potential to overcome the drawbacks of conventional fullerene-based PSCs (e.g., poor solar spectra coverage, large output voltage loss and metastable morphology of photoactive layer, etc.) [7–22] and the highest PCE of 8.4% for evaporated single-junction organic solar cells has been

achieved with multilayer cascade structures [23]. Polymer:polymer blend solar cells in which an n-type semiconducting polymer is used as electron acceptor to replace fullerene derivative are of particular importance owing to the capabilities of tuning energy levels, improved light absorption and stable morphology [14–22]. The PCE of polymer blend PSCs has increased to a competitive value of 7.7% very recently [24]. In contrast to the polymer:fullerene BHJ solar cells, the polymer:polymer blends show more complicated morphology and severe charge recombination behavior, resulting in a relatively poor photovoltaic performance, such as relatively low fill factor (FF) and depressed external quantum efficiency (EQE) or short-circuit current density (J_{SC}). The spherical fullerenes are prone to form nanoscale phase separation with polymer donors and the delocalized pi-orbitals of fullerene aggregates render electrons to lose the spatial correlation quickly with the holes located within polymer donor domains [25]. However, the situation is more complicated in disordered polymer:polymer blend solar

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cells. In polymer blend BHJ, the flexible side chains of conjugated polymers would lead to an increased chain entanglement and suppress the crystallization of the blend film; on the other hand, the polymer donor and the acceptor would tangle together intimately without sharp interfaces, and thus many of the interfacial excitons cannot be split on account of the weak internal electric field due to the decreased intermolecular distance at the donor-acceptor (D-A) interfaces [12]. A lot of work has been focused on developing novel donor or acceptor materials toward optimizing its photovoltaic performance. However, understanding the charge carrier recombination process and the correlation between morphology and photovoltaic performance are of great significance to develop high-efficiency polymer:polymer BHJ solar cells.

Among polymer acceptors reported so far, the most promising n-type polymers are the ones based on strong electron-withdrawing naphthalene di-imides (NDIs) units. A representative n-type polymer is poly[N,N'-bis(2-octyldodecyl)-1,4,5,8-naphthalenedicarbonximide-2,6-diyl]-alt-5,5'-(2,2'-bithiophene) (Polyera ActivInk N2200), which exhibits efficient photovoltaic performance in polymer:polymer blend BHJ solar cells [26–30]. Herein, we selected N2200 as an acceptor and a benzodithiophene-based copolymer poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)-benzo[1,2-b; 4,5-b']dithiophene-2,6-diyl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-b] thiophene-)-2-carboxylate-2-6-diyl] (PBDTTT-EFT) as a donor to fabricate polymer:polymer BHJ solar cells. The long-lived trap-assisted charge recombination originated from the active layer morphological variance has been focused on and investigated. The results indicate that charge recombination is more sensitive to the localized states caused by the morphology of the photoactive layer in PSCs [31,32]. Generally, the loss mechanisms of BHJ solar cells include geminate [33–35], first-order [26,37], and second-order recombination [38]. The geminate recombination is defined as the recombination of Coulombically bound exciton or charge-transfer exciton before creating free carriers, while the last two non-geminate recombination involves free charge pairs originated from two different excitons [39]. A mobile charge recombining with a trapped charge is regarded as a first-order process described by the Shockley-Read-Hall equation [40,41]. If two kinds of mobile charges are involved, the recombination is a second order process (bimolecular, Langevin-type recombination) [42]. Illuminated light intensity-dependent measurement was employed to analyze the charge recombination in PBDTTT-EFT:N2200 blend PSCs processed with various solvents. The results indicate that the loss mechanism in PBDTTT-EFT:N2200 polymer blend solar cells is mainly dominated by a combination of trap-assisted and bimolecular recombination. More importantly, the trap-assisted recombination through localized states is dramatically suppressed within the photoactive layer processed with CF:PX mixed solvent.

2. Experimental

2.1. Materials

The polymer donor PBDTTT-EFT with a weight-average molecular weight (Mw) of 114 kg/mol and a polydispersity index (PDI) of 2.2 was purchased from 1 MATERIAL Co. The polymer acceptor N2200 with an Mw of 61.95 kg/mol and a PDI of 2.1 was obtained from Polyera Inc. Both of the two polymers were used as received. The chemical structures of the two polymers are shown in Fig. 1(a).

2.2. Fabrication and characterization of polymer blend solar cells

The polymer blend solar cells were fabricated with a regular device configuration of glass substrate/ITO anode/PEDOT:PSS

(Baytron 4083) (40 nm)/PBDTTT-EFT:N2200 (1:1 in weight)/Ca (5 nm)/Al (100 nm). A PEDOT:PSS layer was spin-coated on top of the pre-cleaned patterned ITO glass substrate and subsequently baked at 130 °C in air for 30 min. The PBDTTT-EFT:N2200 active layer was spin-coated on the top of the PEDOT:PSS layer from various solvents including *o*-dichlorobenzene (*o*-DCB), chlorobenzene (CB), chloroform (CF), and the CF:*p*-xylene (PX) mixed solvents, respectively. The thicknesses of the PBDTTT-EFT:N2200 active layers were kept at ca. 100 nm by tuning the solution concentration and the spin-coating rate. The thicknesses of various films were determined by DEKTA 6M Stylus profilometer. The Ca/Al cathode was thermally deposited atop the active layer through a shadow mask in a vacuum chamber under a base pressure of 2×10^{-4} Pa. The effective area of the devices was 0.08 cm². The illuminated current density-voltage (*J*-*V*) characteristics were measured under a nitrogen atmosphere using a Keithley 2400 source measure unit under AM1.5G simulated solar light illumination. EQE measurement was carried out at room temperature under ambient condition on QE-R equipment (Enli Technology, Taiwan).

2.3. Thin film characterization

The atomic force microscopy (AFM) images were obtained on an SPI3800N AFM instrument (Seiko Instrument Inc.) in a tapping mode. Transmission electron microscopy (TEM) measurement was performed on a JEOL JEM-1011 TEM instrument operating at an acceleration voltage of 100 kV. All of the samples were taken from the solar cell devices directly. Out-of-plane XRD profiles of thin films were obtained by using a Bruker D8 Discover Reflector and the in-plane data were recorded by using a Rigaku SmartLab instrument ($\lambda = 1.54 \text{ \AA}$). The 2D-XRD images were taken on BL1W1A at Shanghai Synchrotron Radiation Facility ($\lambda = 1.24 \text{ \AA}$). For the XRD measurements, the films were deposited on silicon wafers with a natural oxide and the film thicknesses were ca. 250 nm. The transient absorption decays were measured with a laser flash photolysis spectrometer (Applied Photophysics Ltd., LKS 80). The used excitation wavelength was 600 nm for the PBDTTT-EFT:N2200 blend films and the probe wavelength was 750 nm. The 250-nm-thick samples were encapsulated in a glove box filled with nitrogen.

3. Results and discussion

The PBDTTT-EFT:N2200 blend PSCs were fabricated with a regular device configuration as shown in Fig. 1(b). The energy level alignment in Fig. 1(c) exhibits large offsets between LUMO levels of donor and acceptor, indicating that efficient charge transfer can occur upon photoexcitation. The device configuration and fabrication conditions are the same except that the PBDTTT-EFT:N2200 blend layers are processed with different solvents for these PSCs. The illuminated *J*-*V* characteristics under 100 mW/cm² AM1.5G simulated solar light and EQE curves of the PSCs with the PBDTTT-EFT:N2200 active layer processed with different solvents are shown in Fig. 2(a) and (b), respectively. The photovoltaic parameters of the PSCs are summarized in Table 1. It can be seen from Fig. 2(a) that the PBDTTT-EFT:N2200 PSCs demonstrate a same open-circuit voltage (V_{OC}) of 0.79 V, but different J_{SC} s depending on the processing solvents. The PBDTTT-EFT:N2200 PSC processed with *o*-DCB shows a lowest J_{SC} of 10.21 mA/cm², leading to an overall PCE of 4.45%. When the CF:PX (9:1, v/v) mixed solvent is used to deposit the PBDTTT-EFT:N2200 active layer, the J_{SC} of the resultant PSC was increased to 12.37 mA/cm² and the PCE was finally increased to 5.37%. This value is close to the reported highest PCE of 5.7% for the PBDTTT-EFT:N2200-based PSCs [14]. The EQE curves in Fig. 2(b) show similar trend to the measured J_{SC} s. We can see that the maximum peak of EQE is increased from 54% to 63% with the CF:PX

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