



Suppressed charge recombination in polymer solar cells based on perylene diimide derivative acceptors via solvent vapor annealing

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ABSTRACT

A singly-linked perylene bisimide dimer (di-PBI) and a low band gap polymer based on dithienocarbazole and isoindigo moieties (P(IID-DTC)) are selected as electron acceptor and donor respectively to fabricate non-fullerene based polymer solar cells. The combination of di-PBI acceptor and P(IID-DTC) donor provides well overlapped absorption profile with solar light in the visible region. The P(IID-DTC):di-PBI blend morphology and its effect on charge transport and recombination have been investigated in detail. The dominant bimolecular recombination in commonly processed P(IID-DTC):di-PBI blend results in a low power conversion efficiency of merely 1%. We find that CH₂Cl₂ vapor annealing can effectively improve bi-continuous phase separation and boost the electron transport by more than two orders of magnitude due to the detrap of di-PBI molecules from amorphous P(IID-DTC) matrix. More importantly, the dominant bimolecular recombination in the P(IID-DTC):di-PBI blend films is strongly suppressed. Finally, the power conversion efficiency of non-fullerene P(IID-DTC):di-PBI blend solar cell is dramatically improved to 2.95%.

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1. Introduction

Polymer solar cells (PSCs) have been drawing extensive studies in the past ten years because of their potential applicability in lightweight, flexible and even transparent colorful large-area photovoltaic devices [1–6]. In a bulk heterojunction (BHJ) active layer comprising a donor and an acceptor, the photo-generated excitons are dissociated into free charge carriers at donor/acceptor interfaces, followed by migrating to the respective electrodes via interpenetrating donor/acceptor networks [7–9]. Fullerene and its derivatives are widely used as electron acceptors in

PSCs due to their high electron affinity and excellent electron-transporting capability [10]. Unfortunately, fullerene derivatives have poor absorption in the visible region and thus have negative effect on the optical density of the light-absorbing active layer [11]. As a thin-film type photovoltaic device, increasing the active layer thickness of PSCs toward enhancing its optical density and absorption would lead to significant charge recombination due to limited charge mobilities of organic semiconductors. Thus, the acceptor materials with suitable energy levels and strong absorption in the visible region are highly desired. Some cheap and easily produced non-fullerene acceptor materials including both polymers and small molecules have been attempted to fabricate non-fullerene PSCs in recent years [12–17]. More recently, Jen et al. reported

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an inverted non-fullerene solar cell with power conversion efficiency (PCE) as high as 5.9% by using a simple bay-linked perylene bisimide as acceptors, which is among the highest results of non-fullerene PSCs reported to date [18]. Nevertheless, the photovoltaic performance of non-fullerene PSCs is still lower than those of fullerene-based devices [19,20]. Different from the polymer donor/fullerene derivative BHJ, the BHJ morphology based on non-fullerene acceptors is more complicated, and resultant charge generation as well as recombination needs to be clearly disclosed.

Among the reported non-fullerene acceptor materials, perylene diimides (PDIs) are promising candidates for electron acceptors due to their strong absorption in the visible region, suitable energy levels and favorable electron mobility [21–25]. Throughout the history of organic photovoltaics, PDIs have always been one of the most important acceptor materials for non-fullerene based photovoltaic devices [24]. However, one major issue limiting the application of PDI and its derivatives in solution-processed BHJ PSCs is their strong crystallization capability which always leads to large PDI aggregates in the BHJ active layers. The photo-induced excitons on large PDI aggregates would quickly relax into stabilized and essentially immobile intermolecular states rather than undergo charge transfer, finally leading to an energy loss [26]. Both chemical [27–32] and physical [33,34] approaches have been attempted to optimize the polymer donor:PDI derivatives blend morphology toward depressing the intermolecular state formation. Addition of small amount of 1,8-diiodooctane (DIO) is an effective approach to tune miscibility and inhibit aggregation of PDIs in the non-fullerene blend films [34]. On the other hand, the fast bimolecular recombination would be presented when PDI molecules are finely dispersed in conjugated polymer matrix, severely limiting the photovoltaic performance of the resultant devices [26]. In this case, the isolated PDI molecules serve as electron traps that would facilitate bimolecular recombination of charges [24,26]. Thus, how to suppress both intermolecular state formation and bimolecular recombination toward enhancing charge generation as well as charge extraction is a focus in non-fullerene blend system.

Herein, a singly-linked perylene bisimide dimer (di-PBI) and a dithienocarbazole and isoindigo-based low band gap polymer (P(IID-DTC)) are selected as electron acceptor and donor to fabricate non-fullerene PSCs. The complementary absorption of di-PBI acceptor and P(IID-DTC) donor renders the resultant blend strong absorption and full coverage to solar light in the visible region and thus has a potential to improve photon harvesting with thin BHJ blend film. It is found that there exists severe bimolecular recombination in commonly processed P(IID-DTC):di-PBI blend films. CH_2Cl_2 vapor treatment to the P(IID-DTC):di-PBI blend can effectively improve bi-continuous phase separation and boost the electron transport by more than two orders of magnitude due to the de-trap of di-PBI molecules from amorphous P(IID-DTC) matrix. As a result, the charge recombination in P(IID-DTC):di-PBI blend films is strongly suppressed. Finally, the PCE of the non-fullerene P(IID-DTC):di-PBI blend solar cell is significantly improved to 2.95%.

2. Experimental

2.1. Materials

The chemical structures of organic materials used in this study are shown in Fig. 1a. The low band gap polymer P(IID-DTC) donor and di-PBI acceptor are provided by Y.H. Geng and Z.H. Wang, respectively, and their synthetic procedures are reported elsewhere [9,28]. Poly(ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) (Baytron P Al 4083) was purchased from H.C. Starck and used as received.

2.2. Device fabrication

The non-fullerene PSCs have a structure of ITO/PEDOT:PSS/P(IID-DTC):di-PBI/PDI/LiF/Al and their energy level alignment is shown in Fig. 1b. The patterned indium tin oxide (ITO)-coated glass substrates were cleaned with detergent, and were ultrasonicated in de-ionized water, acetone, and isopropyl alcohol sequentially. The cleaned ITO substrates were subsequently dried at 120 °C in an oven overnight and then subject to UV-Ozone treatment before use. A thin layer of PEDOT:PSS is spin-coated on ITO substrate at 3000 rpm for 1 min, followed by drying at 150 °C for 10 min in vacuum oven. The active layer comprising a blend of P(IID-DTC):di-PBI is spin-coated on top of the PEDOT:PSS layer from *o*-dichlorobenzene solution in a nitrogen-filled glove box. The samples are then transferred into a chamber filled with saturated CH_2Cl_2 vapor and stay for different times to undergo the solvent annealing treatment. The sample is subsequently transferred into an evaporator to thermally deposit a structure of PDI (5 nm)/LiF (1 nm)/Al (100 nm) under a base pressure of 10^{-6} Torr. The active area of the devices is about 0.12 cm^2 , which was defined by the overlapping area of the ITO and Al electrodes. The devices are encapsulated in glove box and measured in ambient condition.

2.3. Photovoltaic measurement

The illuminated current density–voltage (J – V) characteristics of the devices are measured by using a Keithley 2400 source meter. A solar simulator with an AM 1.5G filter is used to provide simulated solar light and the light intensity is determined by a calibrated silicon diode with KG-5 visible color filter. The external quantum efficiency (EQE) measurement was performed with QE-R 3011 equipment (Enli Tech. Co.) with a lock-in amplifier (Stanford, SR830) at a chopping frequency of 133 Hz during illumination with monochromatic light from a Xenon lamp. A 150W quartz-tungsten-halogen lamp is used to provide a background illumination for the EQE measurement.

2.4. Thin film characterization

The atomic force microscopy (AFM) measurements were carried out on SPA-300HV instrument with an SPI3800 controller (Seiko Instruments) in a tapping mode. Transmission electron microscopy (TEM) images were obtained on a JEOL JEM-1011 transmission electron micro-

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