Acceptor-rich bulk heterojunction polymer solar cells with balanced charge mobilities

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A B S T R A C T

In this work, we reported efficient polymer solar cells with balanced hole/electron mobilities tuned by the acceptor content in bulk heterojunction blend films. The photovoltaic cells were fabricated with two new wide band-gap D-A polymers PBDDIDT and PBDDIDTT as the donor material. The molecular conformations of new polymers are carefully evaluated by theoretical calculations. The results of photovoltaic studies show that two devices reach their optimal conditions with rich PC71BM content up to 80% in blend films, which is uncommon with most of reported PSCs. The as-cast devices based on PBDDIDT and PBDDIDTT reveal good photovoltaic performance with PCE of 7.04% and 6.40%, respectively. The influence of PC71BM content on photovoltaic properties is further detailed studied by photoluminescence emission spectra, charge mobilities and heterojunction morphology. The results exhibit that more efficient charge transport between donor and acceptor occurs in rich PC71BM blend films. Meanwhile, the hole and electron mobilities are simultaneously enhanced and afford a good balance in rich PC71BM blend films (D/A, 1:4) which is critical for the improvement of current density and fill factors.

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1. Introduction

During the past decade, polymer solar cells (PSCs) with bulk heterojunction (BHJ) structure have made great achievements owing to their low-cost, low weight and easy fabrication of large area devices [1–4]. The optoelectronic properties of active-layer materials of PSCs with blend of donor material and acceptor material largely determined the power conversion efficiencies (PCEs) of solar cells. Up to date, the study of PSCs has achieved great progress with PCEs exceeding 10% in single- and multi-junction bulk heterojunction (BHJ) devices [5–10]. The electron acceptors including fullerene derivatives (PC61BM, PC71BM et al.) and fullerene-free n-type semiconductors (N2200, ITIC et al.) have been developed and proved efficient [11–16]. To match well with special acceptor materials, donor materials play an important role and the molecular structures and basic optoelectronic properties should be carefully regulated.

Donor-acceptor (D-A) conjugated polymers composed of electron-donating unit (D) and electron-accepting unit (A) are widely investigated as donor materials for easy-tuning of frontier energy levels, absorption spectra and charge mobilities via construction of intramolecular charge transfer (ICT) effect [17,18]. To harvest more solar photons, narrow band-gap (NBG, \( E_g < 1.8 \) eV) polymers with broad absorption spectra were designed and intensively studied. PSCs based on narrow band-gap polymers can exhibit favorable short-circuit current densities (\( J_{SC} \)) benefiting from the broad and strong ICT absorption bands, yet the devices often exhibit decreased open-circuit voltages (\( V_{OC} \)) owing to the high-lying highest occupied molecular orbitals (HOMO). For example, the D-A polymers composed of strong electron-accepting diketopyrrolopyrrole (DPP) unit often possess narrow band-gaps around 1.40 eV and broad absorption [19–21]. DPP-based D-A polymers reported by Janssen exhibit decent \( J_{SC} \) of 15.9 mA cm\(^{-2}\), relatively low \( V_{OC} \) of 0.75 V and the overall power conversion efficiencies (PCEs) of 8.0% [22]. Furthermore, Janssen and coworkers reported a series of small-band gap D-A polymers between 1.13 eV...
and 1.34 eV, composed of strong electron-donating dithieno[3,2-b:2',3'-d]pyrrole (DTP) unit and DPP unit. The PSCs devices reveal high near-infrared photo-response, presenting particularly high $J_{SC}$ up to 23.0 mA cm$^{-2}$; while low $V_{OC}$ of 0.44 V are obtained, leading to a moderate efficiency of 5.3% [23]. Yan and coworkers developed novel narrow band-gaps D-A polymers around 1.6 eV based on difluorobenzothiazole (FBT), the resulting PSCs exhibit high $J_{SC}$ up to 18.8 mA cm$^{-2}$, enhanced $V_{OC}$ of 0.77 V and state of the art PCE of 10.8% [24]. Nevertheless, the open-circuit voltages of photovoltaic devices based on narrow band-gap polymers are difficult to be further improved which inevitably restrict the further development of polymer solar cell efficiencies.

To solve the above challenges, medium band-gap (MBG, $E_{g}$:1.6–1.8 eV) and wide band-gap (WBG, $E_{g}$ > 1.8 eV) D-A polymers were designed to explore a balance between $J_{SC}$ and $V_{OC}$ of PSCs. Currently, benzodithiophene (BDT) moieties and analogues including one-dimension (1D), two-dimension (2D) and asymmetric 1D-2D derivatives are efficient electron-donating building blocks to construct medium or wide band-gap D-A photovoltaic donor materials, due to its high stability against oxidation ability and planar molecular framework [25–30]. Recently, our group developed a series of D-A polymers based on asymmetric 1D-2D and planar molecular framework [25]. Among them, the polymer PBDD/IDT2BrBDD based on strong electron-donating moiety IDT and IDTT and withdrawing moiety benzo[1,2-c:4,5-c']dithiophene-4,8-dione (BDD) to couple weak electron-donating moiety IDT and IDTT and synthesize two wide band-gap D-A conjugated polymers PBDDIDT and PBDDIDTT with optical band-gap over 1.85 eV. Two polymers show similar absorption profiles which are synthetically determined by different polymer structures and distortion dihedral angles between D and A moieties. Polymer solar cells were fabricated with PC$_{71}$BM as acceptor material. The results reveal that both additive-free devices based on PBDDIDT and PBDDIDTT exhibit favorable photovoltaic properties with PCE up to 7.04% and 6.40%, respectively. The photovoltaic performance based on PBDDIDT is one of the highest efficiencies achieved with WBG IDT-based polymers. Notably, the polymer/PC$_{71}$BM weight ratios have a significant impact on FF values and fullerene-rich devices exert better performance when the optimal weight ratios for two devices are determined to be 1:4. This is very different from other IDT-based polymer devices and most of PSCs. The correlations between donor/acceptor weight ratios and fill factors are investigated and proved to be mainly affected by high and balanced hole/electron charge transport. Photoluminescence (PL) spectra were carried out.

Scheme 1. Synthesis and chemical structures of PBDDIDT and PBDDIDTT.
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