In a high performance PTB7:PC71BM bulk-heterojunction (BHJ) solar cell, the commonly optimized polymer:fullerene (D:A) weight ratio is about 1:1.5, when PC71BM is used as the acceptor. This report explores alternative D:A weight ratios. We describe how to enrich the polymer contents of these BHJ solar cells to achieve high power conversion efficiencies (PCEs). The concentration of 1,8-diiodooctane (DIO), a solvent additive for the BHJ precursor solutions, is increased in order to re-optimize the BHJ cells. The PCEs of the re-optimized cells are improved for the PTB7 cells. Detailed charge transport measurements were carried out to examine the polymer-rich BHJs. We observed enhanced hole mobilities for the PTB7 BHJs. Additionally, the electron mobilities are preserved due to the dispersion of fullerene domains by increased DIO concentrations. Two other well-known polymer donors PCDTBT and PDTSTPD have been also investigated, and the improvements of hole mobilities and PCEs can be obtained for both polymer-rich BHJ solar cells. © 2016 Elsevier B.V. All rights reserved.

1. Introduction

Organic photovoltaic (OPV) cells have attracted tremendous attentions [1–5]. They are light, flexible, and have huge potential in large area fabrication by roll-to-roll processing [6–8]. In a polymer-based OPV cell, the polymer acts as the light absorber, electron donor, and hole transporter, whereas fullerene molecules typically have less optical absorption and mainly serve as electron acceptors and transporters [9]. If the polymer and fullerene are blended together intimately, the resulting bulk heterojunction (BHJ) is a very effective active layer for optical absorption and charge carrier transport [10–12]. Among many tested BHJs, the PTB7:PC71BM BHJ blend with 1,8-diiodooctane (DIO) as the additive in casting solution is one of the well-studied and efficient OPV systems [13–15]. A certified power conversion efficiency (PCE) of 9.2% has been reported [16]. Other BHJ devices such as PCDTBT:PC71BM and PDTPS:PC71BM have been reported with the PCEs of 6.1% and 7.3% [10,17]. Of the BHJs listed above, the optimal donor-acceptor (D:A) weight ratios are all larger than 1:1. However, the fullerene acceptors have weak absorbances in solar spectrum. Therefore, it should be highly desirable to enrich the polymer content in the BHJ in order to harvest more photon energy. Furthermore, a higher polymer content should improve hole transport, lead to better balance of electron and hole mobilities, and improve the PCE.

Recently, novel polymers have been designed to accomplish BHJs with high polymer contents. Qian et al. designed a novel polythiophene derivative (PBT1) [18]. With a D:A ratio of 1:0.66, an impressive PCE of 6.88% was achieved. More recently, Yuan et al. synthesized a polymer PTP8 with a high Voc (~1.0 V) and PCE of 6% [19]. The D:A weight ratio is 1:0.5. In both of these examples, the main strategy for reducing the content of fullerene is to reduce the free volume between the side chains of the high performance polymers. As proposed previously by McGehee et al. [20], large spacings in between side chains may be conducive to fullerene intercalations and lead to a high level of fullerene loading. Thus, dense polymer side-chains should be able to suppress fullerene loading without compromising the power conversion efficiencies. This concept was realized from Refs. [18,19], using bulky and rigid side chains.

In this report, we adopt a different strategy to realize OPV cells with enhanced polymer contents. We employ a well-known BHJ system PTB7:PC71BM and explore how to fabricate polymer-rich OPV devices with better performances. The chemical structures of the materials are shown in Fig. 1. Our general strategy is as follows. Due to increased polymer content, the reduced fullerene concentration...
in the BHJ is expected to suppress electron mobilities. To compensate this effect, we increase the DIO additive concentration in the processing solution. Although the fullerene concentration is reduced, a higher DIO concentration further disperses the fullerene domains, and helps to retain the electron mobility for electron extraction. With this strategy, we re-optimized the BHJs which have enriched polymer contents. For the BHJs using PTB7 as the donor polymer, their polymer-rich solar cells have improved PCEs relative to their reference cell. To look into the origins of the improved PCEs for the polymer-rich cells, we investigate the optical and transport properties of the BHJ films. The polymer-rich BHJ film exhibits an improved optical absorption and hole mobility. The polymer-rich strategy also works for other two BHJ systems based on PCDTBT:PC71BM and PDTSTPD:PC71BM. The enhancements of PCE, hole mobility and UV–visible optical absorption can be also observed in both devices with higher polymer contents. Our results provide insight on the existing limitation of D:A ratio on the photovoltaic performance, and suggest alternatives to optimize the BHJ devices.

2. Result and discussion

2.1. Impacts of donor-acceptor ratio on OPV devices performance

Fig. 2(a) shows the current-voltage (J-V) characteristics of optimized PTB7:PC71BM bulk-heterojunction OPV devices with different donor-acceptor mass ratios under 100 mWcm⁻² AM1.5G illumination. The polymer-rich device was processed from a casting solution consisting of higher DIO additive concentrations for the balance of carrier mobilities. For the PTB7:PC71BM solar cells, 4 vol% DIO additive was doped into the casting solution of the polymer-rich (1:1 D:A mass ratio) devices, comparing with only 3 vol% DIO in the casting solution of the control devices (1:1.5 D:A mass ratio). The PCE of the polymer-rich cell can be found to be enhanced by a factor of 10%, from a control device with a PCE of 7.0%–7.7%. The Jsc is the main contribution of the PCE enhancement, increasing from 15.1 mA/cm² of the control device to 16.8 mA/cm² of the polymer-rich device. However, the Voc and FF have no significant changes in both D:A ratio devices with various DIO concentrations. Table 1 lists the performances of the polymer-rich and control PTB7:PC71BM devices. Our result suggests that the polymer/fullerene OPV solar cell can be fabricated with reduced fullerene content. The Jsc of device can be improved by the reduction of PC71BM content with a higher DIO concentration in casting solution.

Fig. 2(b) shows the external quantum efficiency (EQE) spectra of the PTB7:PC71BM devices with different D:A ratios. The optimized thickness of PTB7:PC71BM bulk-heterojunction active layer is around 90 nm, where the PCE performance is the best for the organic solar cells. The polymer-rich device shows an EQE enhancement in the wavelength region from 530 nm to 800 nm, in which PTB7 mainly contributes the light absorption. The EQE of the polymer-rich cell maintains above 70% from 490 nm until 695 nm, while that of the control cell decreases gradually to only 56% in 700 nm. Fig. 2(c) shows the UV–visible absorption spectra of the PTB7 BHJs. The polymer-rich BHJ film has an distinct enhanced absorbance from 500 to 700 nm arising from the enhanced polymer content, when compared to the control sample (D:A ratio of 1:1.5).

In order to understand how the polymer fraction impacts on the carrier transport characteristics, hole-only devices were fabricated for the J-V measurements. The general device structure is ITO/PEDOT:PSS/BHJ/CuPc:spiro-TPD/Au. The PEDOT:PSS layer plays the role of the hole injection layer. The electron blocking layer (EBT) is a mixed layer of spiro-TPD and CuPc in a ratio of 30:1 [21,22]. The JV data of PTB7:PC71BM hole-only devices at room temperature are shown in Fig. 2(d). The hole mobilities of the BHJs can be obtained by the space-charge-limited current (SCLC) equation [23–25].

$$J_{SCL}d = \frac{9}{8}\varepsilon_0\varepsilon_r\mu_0\exp\left(0.89\beta\sqrt{F}\right)F^2$$  \hspace{1cm} (1)

where J_{SCL} is the space-charge-limited current density, d is the thickness of the BHJ film, \varepsilon_0 is the permittivity of a vacuum, \varepsilon_r is the relative permittivity of the polymer, \mu_0 is the zero-field mobility, \beta is the field-dependent coefficient, and F is the average electrical field. It can be seen that the polymer-rich BHJ has an enhanced hole current with respect to the control devices, and therefore the hole mobility. It increases gradually from around $1.3 \times 10^{-4}$ cm²V⁻¹s⁻¹ to around $2.2 \times 10^{-4}$ cm²V⁻¹s⁻¹, when the D:A ratio increases from 1:1.5 to 1:1. Table 1, last column, is a summary of the SCLC fittings.

2.2. Electron and hole transports measurements of polymer-rich BHJ of PTB7:PC71BM

To probe further the carrier transport properties, we use PTB7:PC71BM system as a model system and employ admittance
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