



Dramatically enhanced performances and ideally controlled nano-morphology via co-solvent processing in low bandgap polymer solar cells



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ARTICLE INFO

Article history:

Received 5 March 2016

Received in revised form

6 April 2016

Accepted 7 April 2016

Available online 14 April 2016

Keywords:

Bulk heterojunction solar cell

Polymer solar cell

Co-solvent

Nanoscale morphology

Phase separation

ABSTRACT

The device performance of photovoltaics with a polymer:fullerene bulk heterojunction (BHJ) structure, consisting of DT-PDPP2T-TT donor polymer and poly(3-hexylthiophene):[6,6]phenyl-C₆₁-butyric acid methyl ester (PC₆₁BM) acceptor compound, was investigated as a function of co-solvent composition. An enhancement of the photocurrent density and fill factor is observed in diodes made by spin-coating with chloroform mixed with *ortho*-dichlorobenzene, which allows a significantly higher device efficiency of 5.55% compared to diodes made from neat chloroform (efficiency = 3.61%). To clarify the role of the co-solvent, we investigated the nanoscale morphology with AFM, TEM and 2D-GIWAXS techniques and also the free-charge carrier mobility via space-charge limited current theory. We obtained the result that, under such supersaturated conditions, co-solvents induce increased polymer crystalline aggregation into a 3D phase structure and boost charge-carrier transport characteristics. This provides a rational basis for the development of ideally-controlled BHJ films that yield efficient DT-PDPP2T-TT:PCBM solar cells. Therefore, carefully selecting solvent mixtures provides an approach toward efficient low bandgap polymer solar cells.

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1. Introduction

Polymer solar cells (PSCs) have attracted considerable interest in recent years, due to their potential for rapid energy payback time and low-cost fabrication of flexible plastic solar modules [1–3]. Bulk heterojunction PSCs, typically consisting of an active layer with a donor-acceptor blend film (electron-donating polymers, electron-accepting fullerenes), have garnered significant attention for reaching a high power conversion efficiency (PCE) of up to 9–10% [4,5]. The strategy for power conversion efficiency (PCE) improvement of BHJ PSCs could be summarized as the following: the design and synthesis of polymeric electron-donating materials [6–10]; the development of novel device architectures [11–13]; the control of nanoscale BHJ morphology [14,15]; and, selecting

interfacial layers for better charge carrier collection [16].

Among the promising device configurations, controlling the degree of phase separation for the BHJ morphology is one of the best strategies for boosting the performance of PSCs. Therefore, several methods have been applied to modify or control the morphology of the BHJ blends such as solvent (vapor) annealing, thermal annealing, and selection of the processing solvent or mixed solvent [17–23].

For example, very recently, Janssen et al. reported the realization of desired polymer aggregation via a co-solvent in diketopyrrolopyrrole (DPP)-based solar cells [24,25]. In a similar period, Cheng et al. reported that using mixed chloroform:dichloromethane solvents rather than chloroform is better for the improvement of efficiency due to resultant ordered microstructural features of active layer materials [26]. Moreover, Jenekhe et al. studied all-polymer PSEHTT/PNDIS-HD films with which a co-solvent controlled phase-separated domain sizes [27]. According to these works, it can be concluded that crystallinity as well as domain size in the blends can be effectively tuned by using binary solvent mixtures. As a result, how to screen for a suitable solvent system with which to

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adjust the morphology of the active layer is one of the key issues in the quest for high performance PSCs.

Based on the aforementioned considerations, we introduced two functional solvents as processing solvents for low bandgap DT-PDPP2T-TT:PCBM solar cells, to create efficient bulk heterojunction PSC devices (device structure; Fig. 1a). Indeed, we recently reported a DT-PDPP2T-TT:PCBM system in tandem organic solar cells that exhibits superior photovoltaic properties with a PCE of 7.4% [28]. Although this copolymer shows excellent performances, the detailed understanding and information on the effects of the co-solvents are not in that study.

Therefore, for the present article on the DT-PDPP2T-TT:PCBM blend we systematically studied the effect of the co-solvent on device performance using two commonly-used mixed solvents: chloroform (CF) and *ortho*-dichlorobenzene (DCB). To clarify the role of the co-solvents, the morphological evolution of the blends was demonstrated by atomic force microscopy and grazing incidence wide angle X-ray scattering. Moreover, free-carrier transport characteristics were investigated using the space-charge limited current model.

2. Results and discussion

Before investigating for the effects of co-solvents on device performance, we have first rechecked the energy levels of each component's band structure, as shown in Fig. 1b. The difference between DT-PDPP2T-TT and PC₆₁BM of the highest occupied molecular orbital (HOMO) energy was ca. 1.0 eV, which is sufficient to block the electrons diffused from the DT-PDPP2T-TT polymer [29]. Furthermore, considering the large lowest unoccupied molecular orbital (LUMO) offset (0.6 eV) between the two active materials in the solid-state films, the charge separation process from the excitons in the DT-PDPP2T-TT domain to the LUMO level of the PC₆₁BM domain is expected to occur efficiently [29].

2.1. The effect of co-solvents on device efficiency

A series of polymer solar cells with DT-PDPP2T-TT:PCBM as the active layer were fabricated to investigate the effect of mixed solvents on photovoltaic performance. Fig. 2a exhibits the distinct differences in the current density-voltage (*J*-*V*) characteristics for layers processed using chloroform (CF) with and without 50, 100 and 200 vol% DCB as co-solvent (detailed device parameters are in Table 1). For the solar cell devices processed with neat CF as solvent, an optimized PCE of 3.61% was obtained with an open-circuit

voltage (*V*_{oc}) of 0.71 V, a short-circuit current (*J*_{sc}) of 8.28 mA cm⁻² and a fill factor (FF) of 62%. By using CF:DCB (1:0.5, v/v) mixed solvents, an enhanced photovoltaic performance with a PCE of 4.76% was obtained. Furthermore, further increases in the *J*_{sc} (from 11.24 mA cm⁻² to 11.79 mA cm⁻²) and FF (from 62% to 65%) were obtained by using the co-solvent (CF:DCB = 1:1) and thus the PCE was increased to 5.55%. However, when the concentration of DCB was further increased (CF:DCB = 1:2), both *J*_{sc} and FF were decreased, (but not the *V*_{oc} value)— below the values that were obtained by using CF only. As a result, the champion device was obtained when a CF:DCB solvent blend with the optimal composition of 1:1 was processed. We expected that by processing CF:DCB co-solvents, particularly with 1:1 vol ratios, the main impact would be the enhanced photocurrent and fill factor values. For these *J*_{sc} improvements to have occurred, we believe that the differences in the photovoltaic performances are mainly due to the differences in network morphology and molecular packing structures, as is discussed below.

In addition, to be able to investigate the impact on the fullerene compounds, the DT-PDPP2T-TT:PC₇₁BM device spin-coated from CF:DCB 1:1 v/v was recorded; they show no significant effect on the PC₇₁BM compound, which suggests that considerable variation of the device performances possibly due to processing solvents.

In order to further confirm the effect of the co-solvent on PSC performance, incident photon to current efficiency (IPCE) spectra were measured and are shown in Fig. 2b. The IPCE spectra of DT-PDPP2T-TT:PC₆₁BM PSCs made with CF and with CF:DCB show that devices processed with CF:DCB as solvent have quite higher IPCE values in the entire wavelength range. Moreover, according to the IPCE curves, the calculated *J*_{sc} is 8.36 mA cm⁻² for CF-devices, 12.19 mA cm⁻² for CF:DCB-polymer:PC₆₁BM devices, and 12.36 mA cm⁻² for CF:DCB-polymer:PC₇₁BM devices, which is in good agreement with the *J*_{sc} results obtained from *J*-*V* characteristics curves within a 3% error [30]. And also, with regard to fullerene derivatives (PC₆₁BM and PC₇₁BM), the IPCE spectra show that the IPCE value of PC₇₁BM is superior to that of PC₆₁BM in the short-wavelength range (400 nm–700 nm), but inferior in the long-wavelength range (700 nm–900 nm). The effect of using fullerene derivatives is inadequate to enhance the PCE, and so controlling the co-solvent system for devices made of DT-PDPP2T-TT is more meaningful than changing the fullerene compounds.

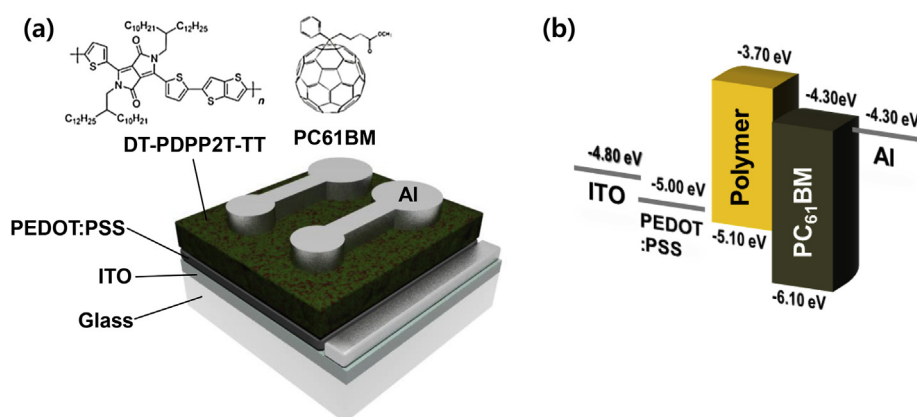


Fig. 1. (a) Conventional structure of ITO/PEDOT:PSS/DT-PDPP2T-TT:PCBM/Al and the molecular structures of the polymer and PCBM. (b) Schematic diagram for band energy levels of the proposed device structure.

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