

# Homogeneous phase separation in polymer:fullerene bulk heterojunction organic solar cells



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## ARTICLE INFO

### Article history:

Received 30 May 2015

Received in revised form 21 June 2015

Accepted 28 June 2015

Available online 3 July 2015

### Keywords:

Phase separation  
Homogeneous distribution  
Bulk heterojunction  
Organic solar cells

## ABSTRACT

Phase separation in donor/acceptor bulk heterojunction (BHJ) significantly affects the exciton dissociation and charge collection in organic solar cells (OSCs). In this work, the dynamics of phase separation in poly[[4,8-bis[(2-ethylhexyl)oxy] benzo[1,2-b:4,5-b'] dithiophene-2,6-diyl] [3-fluoro-2-[(2-ethylhexyl) carbonyl] thieno [3,4-b] thiophenediyl]] (PTB7):[6,6]-phenyl C71 butyric acid methyl ester (PC<sub>70</sub>BM) based BHJ is analyzed. Solvent additive diiodooctane selectively dissolves PC<sub>70</sub>BM aggregation to form homogeneous distribution in PTB7 matrix. Quantitative analyses show that the size of PC<sub>70</sub>BM domains decreases from 67 nm to 38 nm with the addition of solvent additive. The vertical phase separation in PTB7:PC<sub>70</sub>BM BHJ was studied by the X-ray photoelectron spectroscopy measurement and its effect on the performance of the regular and inverted OSCs was examined. Compare to the vertical phase separation occurred in the poly(3-hexylthiophene):PCBM system, the homogeneous vertical phase separation observed in the PTB7:PC<sub>70</sub>BM BHJ favors an enhanced exciton dissociation process for attaining high performing OSCs.

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## 1. Introduction

Organic solar cells (OSCs) have attracted extensive attention owing to the low cost, flexibility and easy processability for large area device fabrication [1–5]. The heart of the bulk heterojunction (BHJ) is made by blending the donor and acceptor materials. The photogenerated exciton in the BHJ diffuses to the donor/acceptor interface where the exciton dissociates into free charge carriers. The free charges are transported via the percolation pathways and extracted by the electrodes. Therefore, optimization of the phase separation between the donor and acceptor in the BHJ is critical to the performance of OSCs [6–11]. A series of processing techniques, such as solvent vapor annealing [12,13], thermal annealing [14] and solvent additive treatment [15–18] have been applied to achieve controllable morphology. Among these strategies, solvent additive processing is a simple and low cost way that can be easily integrated into solution-processed techniques, such as ink-jet printing or roll-to-roll coating. Recently, a record power conversion efficiency (PCE) of around 10% for single junction OSCs with

inverted structure was achieved by adding 3% volume diiodooctane (DIO) into poly[[4,8-bis[(2-ethylhexyl)oxy] benzo[1,2-b:4,5-b'] dithiophene-2,6-diyl] [3-fluoro-2-[(2-ethylhexyl) carbonyl] thieno [3,4-b] thiophenediyl]] (PTB7): [6,6]-phenyl C71 butyric acid methyl ester (PC<sub>70</sub>BM) blend solution [19,20]. However, the phase mixture and segregation in PTB7:PC<sub>70</sub>BM are still not fully understood yet due to the small length-scales and low contrast between the two organic materials.

Vertical phase separation in the donor:acceptor BHJ controls the charge transportation and collection in OSCs. Therefore, various techniques [21–24] have been employed to optimize the vertical phase separation in poly(3-hexylthiophene) (P3HT): [6,6]-phenyl-C<sub>61</sub>-butyric acid methyl ester (PCBM) BHJ based OSCs. It was demonstrated that vapor or thermal annealing resulted in an increase of P3HT concentration at the blend/anode interface [21]. Several groups [23,25] have reported that solvent additive induces P3HT segregation at the top surface of the P3HT:PCBM blend film. Thus, the inverted configuration favors charge collection at the cathode/PCBM interface and also possesses a slow degradation behavior as compared to the regular OSCs in the accelerated aging test in air [26]. This kind of morphology in P3HT blends was widely employed in the high efficient PTB7:PC<sub>70</sub>BM BHJs in recent reports

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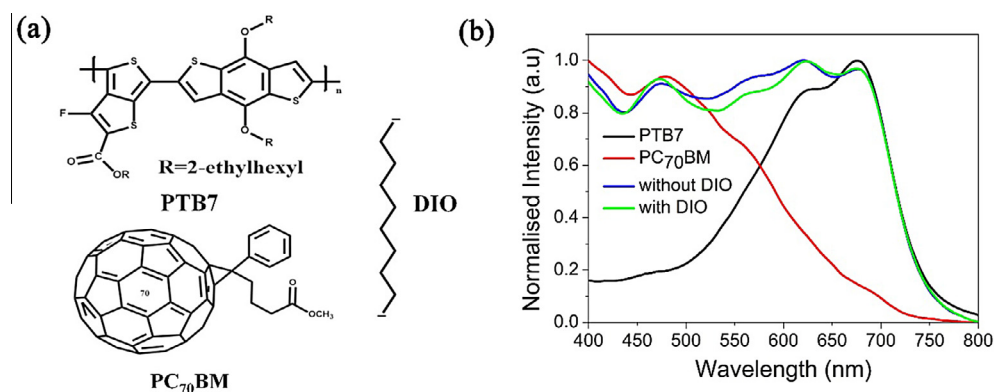
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[19,27]. Nevertheless, the vertical phase distribution in PTB7:PC<sub>70</sub>BM system has not been systematically clarified.

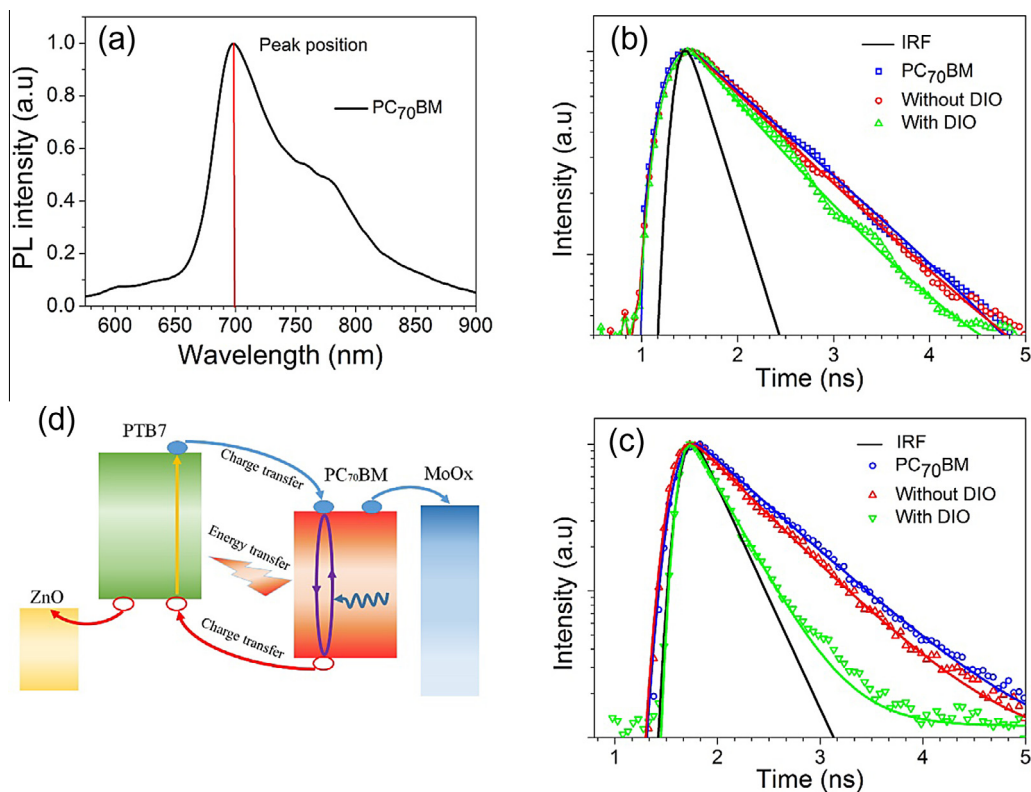
In this work, the nanoscale phase separation in the PTB7:PC<sub>70</sub>BM BHJ was examined using atomic force microscopy (AFM), time-resolved photoluminescence (PL) and grazing incidence small angle X-ray scattering (GISAXS) measurements. The vertical phase separation in PTB7:PC<sub>70</sub>BM system was analyzed with X-ray photoelectron spectroscopy (XPS) measurement. The effect of vertical phase distribution on the performance of structurally identical OSCs with regular and inverted configurations is studied.

## 2. Experimental section

PTB7, PC<sub>70</sub>BM and DIO were purchased from 1-Material, Nano-C and Sigma–Aldrich and used as received without further purification. PTB7 and PC<sub>70</sub>BM were blended in a weight ratio of 1:1.5 and dissolved in chlorobenzene at a concentration of 25 mg ml<sup>-1</sup> for device fabrication. PTB7:PC<sub>70</sub>BM samples for the time-resolved PL measurements were prepared using a lower concentration of 5 mg ml<sup>-1</sup>. The solutions were stirred at 70 °C for 24 h. DIO was then mixed in the PTB7:PC<sub>70</sub>BM solution in a 3% volume ratio prior to the spin-coating. The uniform blend films



**Fig. 1.** (a) Molecular structures of PTB7, PC<sub>70</sub>BM and the DIO additive; (b) The normalized absorption spectra measured for the thin films of PTB7, PC<sub>70</sub>BM, PTB7:PC<sub>70</sub>BM prepared using the solutions with or without DIO additive.



**Fig. 2.** (a) The emission spectrum of PC<sub>70</sub>BM; Time-resolved PL spectra measured for pure PC<sub>70</sub>BM, PTB7:PC<sub>70</sub>BM blends with or without DIO additive in solution (b) and solid film (c). IRF stands for instrumental response function. It should be noted that no PTB7 fluorescence is observed even at the timescales as short as 2 ps with the selective excitation wavelength 670 nm, as reported by Hedley et al. [6] Hence, PC<sub>70</sub>BM emission was chosen to analyze the exciton dynamics. (d) Excited dynamics between PC<sub>70</sub>BM and PTB7.

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