

Effect of vertical morphology on the performance of silole-containing low-bandgap inverted polymer solar cells

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ABSTRACT

The vertical distribution of blended components in the active layer of a bulk heterojunction solar cell composed of poly[(4,4'-bis(2-ethylhexyl)dithieno[3,2-b:2',3'-d]silole)-2,6-diyl-alt-(2,1,3-benzothiadiazole)-4,7-diyl] (PDTS-BTD) and [6,6]-phenyl-C71 butyric acid methyl ester (PC₇₁BM) was studied using X-ray photoemission spectroscopy (XPS) and Auger electron spectroscopy (AES) depth profile. We found that the vertical profile of the PDTS-BTD:PC₇₁BM thin films varies, with a PC₇₁BM rich region near the bottom surface and a polymer rich on the top surface. This suggests that the inverted device geometry may be superior to the conventional device geometry for the fabrication of efficient polymer solar cells. Significant enhancement in the power conversion efficiency (PCE) of an inverted polymer solar cell was observed compared to the conventional cell. The enhancement in device performance is attributed to the efficient charge extraction due to the exploitation of vertical morphology of the active layer using the inverted device geometry.

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1. Introduction

Polymer solar cells (PSCs) have become an active area of research in recent years due to their potential as an economically competitive sustainable energy source [1–3]. It has been demonstrated that the performance of PSCs can be enhanced using many approaches, such as synthesizing new narrow band-gap semiconducting polymers [4–7], optimization of morphology in the bulk hetero-junction layers [8,9], adopting suitable interlayers for efficient charge carrier extraction [10,11], and the design of novel device geometries [12–14]. However, the power conversion efficiency (PCE) of PSCs that has been achieved so far is not yet sufficient for large scale commercialization.

Morphology optimization of the active layer is one of the factors determining the efficiency of PSCs [15,16]. Many studies have shown that the active layer morphology such as packing of the molecules and the domain formation of donor and acceptor phases at nano-scale control the device performance [14,17]. Optimum nano-scale phase morphology can be achieved using various process

conditions such as solvent additives [8,16,18], thermal annealing, vapor annealing, and slow drying of spin-coated films [19–22]. Besides the lateral phase separated morphology, the vertical morphology plays a crucial role in determining the exciton dissociation efficiency and charge extraction efficiency, and thus the overall PCE of the photovoltaic (PV) devices [14,23]. In an ideal device structure, the region close to the anode contact should be donor-rich while the region close to the cathode region should be acceptor-rich. However, some material systems such as poly(3-hexylthiophene) (P3HT) donor and fullerene derivative [6,6]-phenyl-C61 butyric acid methyl ester (PCBM) acceptor exhibit vertical phase morphology resulting in polymer enrichment at the top surfaces and fullerene enrichment at the bottom [14,17]. This inhomogeneous vertical phase separation is more favorable for the inverted device geometry than the conventional device geometry and it has been demonstrated to improve the performance of PSC upon changing from conventional to inverted devices [24,25].

In this work, we sought to determine whether or not the vertical phase segregation that occurs in P3HT/PCBM solar cells also occurs in low bandgap polymer/fullerene blends. To this end, we studied the vertical phase separation of spin coated poly[(4,4'-bis(2-ethylhexyl)dithieno[3,2-b:2',3'-d]silole)-2,6-diyl-alt-(2,1,3-benzothiadiazole)-4,7-diyl] (PDTS-BTD) and fullerene derivative blends of active layers using X-ray photoemission spectroscopy (XPS) and depth profiling using Auger electron spectroscopy (AES). The molecular structures of PDTS-BTD and [6,6]-phenyl-C71

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butyric acid methyl ester (PC₇₁BM) are shown in Fig. 1a. In order to determine whether or not the vertical phase separation of active layer influences the device performance, we have fabricated both conventional and inverted polymer solar cells using identical processing conditions for the active layer. Compared to the conventional PV device, the PCE of the inverted cell increased from 4.42% to 5.51%.

2. Experimental details

The polymer PDTS-BTD was synthesized according to a previously reported method [26]. The sol-gel ZnO was prepared by dissolving zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ (Aldrich, 0.11 g) and poly(vinylpyrrolidone) (Aldrich, 0.03 g) in 10 ml ethanol under vigorous stirring for 2 h at 70 °C. Both conventional and inverted PV devices are fabricated [18] as shown in Fig. 1b and c. To fabricate conventional solar cells, first a thin layer (30 nm) of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS; Baytron Al 4083 from HC Starck) was spin-coated on an ultrasonically cleaned ITO substrate, followed by baking on a hot plate at 180 °C for 10 min. For cells with a MoO₃ interlayer, a 10 nm thick MoO₃ film was thermally evaporated on the ITO substrate at vacuum of 10⁻⁷ Torr. Subsequently, the photoactive layer was produced by spin coating films of the blend

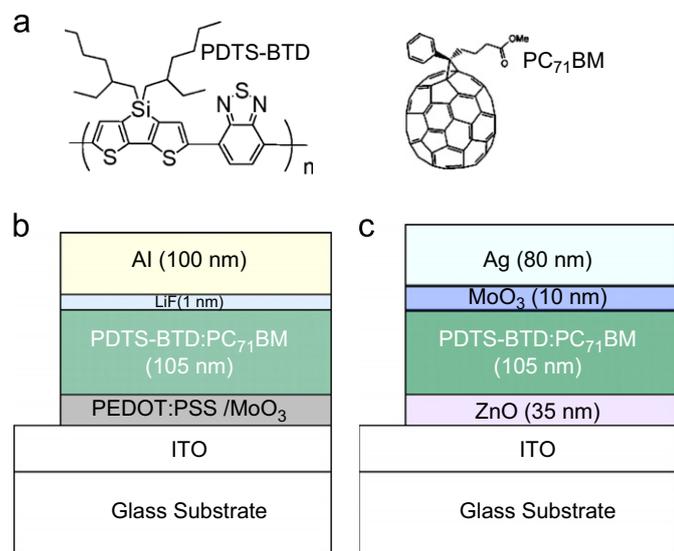


Fig. 1. (a) Molecular structure of PDTS-BTD polymer and PC₇₁BM. (b) Schematic diagram of the PV devices with (b) conventional and (c) inverted geometry.

PDTS-BTD:PC₇₁BM (1:1.5 weight ratio) from chlorobenzene solutions on top of the PEDOT-PSS layer. The device was subsequently heated on a hot plate at 150 °C for 5 min. LiF (1 nm) and aluminum (100 nm) were thermally evaporated at vacuum of 10⁻⁷ Torr on top of the active layer as a cathode.

Inverted solar cells were fabricated by spin coating sol-gel ZnO on top of the ITO glass substrate and then annealed at 200 °C for 30 min. An active layer solution of PDTS-BTD and PC₇₁BM with a concentration of 20 mg/ml was spin coated on top of the ZnO film followed by annealing at 150 °C for 5 min. Thin films of MoO₃ (10 nm) and silver (80 nm) were sequentially evaporated at a vacuum pressure of 10⁻⁷ Torr on top of the active layer as an anode. The device area was 0.04 cm². The thickness of the polymer/fullerene layer was 105 nm, as determined by a profilometer. Current density versus voltage measurements were carried out using a Keithley 4200 semiconductor characterization system under AM1.5 G solar simulator with an irradiation intensity of 100 mW/cm². Device fabrication was done under nitrogen atmosphere and characterizations were performed in an ambient environment without any encapsulation.

A Perkin-Elmer PHI 5100 X-ray photoelectron spectrometer with MgK α X-rays (1256.6 eV) was used to determine the chemical composition of PDTS-BTD:PC₇₁BM films. To study the vertical composition of PDTS-BTD:PC₇₁BM film, a Perkin-Elmer PHI 660 scanning Auger spectroscopy system in combination with Ar-ion beam milling was used. In this system, the surface layer of a film can be tested for its atomic constituents. After the Auger scan, the surface layer is milled off with an Ar-ion beam, and the new surface layer can be tested. Surface morphology of the active layer was investigated by using a Veeco Nanoscope 3100 Atomic force microscope (AFM).

3. Results and discussion

3.1. Vertical morphology of active layer

X-ray photoemission spectroscopy (XPS) was performed to investigate the composition of the active layer at the top and the bottom surfaces of the PDTS-BTD/PC₇₁BM blend films. Here, we inspect the differences in XPS intensity of 2p and 1s bands of sulfur and carbon, respectively, on both sides (top and bottom) of the active layer film surface to determine the relative composition of PDTS-BTD polymer and PC₇₁BM in the film. In order to investigate the bottom surface of the active layer, the samples were prepared using a lift-off technique in which the blended (PDTS-BTD:PC₇₁BM) films spin coated on PEDOT:PSS layer are lifted off from the surface using water and the floated films are

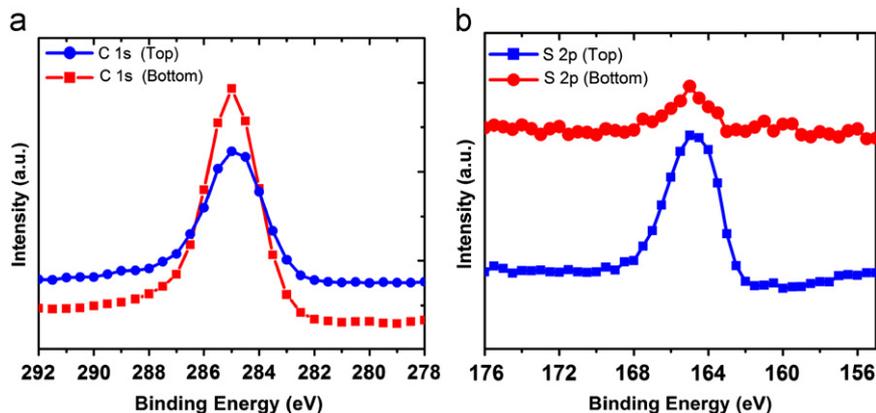


Fig. 2. XPS spectra of (a) C 1s region and (b) S 2p region obtained from top and bottom surfaces of the PDTS-BTD:PC₇₁BM films.

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