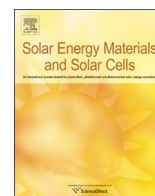




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Investigating coating method induced vertical phase distribution in polymer-fullerene organic solar cells

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ABSTRACT

The morphologies of bulk heterojunction (BHJ) films of blended semiconductor of poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C61 (PCBM) deposited by doctor blading and spin coating were investigated. Controlled phase separation in BHJ organic/polymer solar cells (OSCs) were achieved using doctor blading. The results revealed that the coating method induced a significant difference in the film morphology. The vertical phase distribution of the BHJ layer fabricated by doctor blading was uniform, while uneven phase distribution was observed in spin coated BHJ layer. The uniform phase distribution in the doctor blading coated film resulted in higher charge carrier mobility and power conversion efficiency in solar cell than those in films fabricated from spin coating. The results demonstrated that doctor blading has advantages not only in large area high throughput and roll-to-roll compatibility, but also in better control of film morphology to achieve high efficiency consistently in OSC devices.

1. Introduction

Organic/polymer solar cells (OSCs) have numerous advantages due to light weight, low cost and ease of large area scaling up via roll-to-roll coating/printing processes [1–4]. Some roll-to-roll compatible processes have been explored for the fabrication of OSCs, such as screen printing [5], inkjet printing [6,7], gravure and flexographic printing [8], spray coating [9], brush painting [10,11], doctor blading and slot-die coating [12–14]. Among them, doctor blading and slot-die coating are the most promising technologies for OSC fabrication as it is capable of high speed coating, good film morphology control and tolerance to the rheological properties of the coating solutions. The power conversion efficiency (PCE) of a photovoltaic (PV) module is highly dependent on the uniformity of the film morphology and nanoscale phase separation in the active layer. Phase separation has been proven to be a crucial factor in realizing high efficiency bulk heterojunction (BHJ) OSCs [15,16]. With optimized phase separation, the donor and acceptor domains form an inter-penetrating network, which helps to create a large donor/acceptor interface for efficient charge separation and transportation. For solution processes, some factors are believed to affect phase separation, such as the donor/acceptor crystallinity [17], blend composition [18], solvent evaporation rate [19], solvent mixture/additives [20], and post thermal or solvent annealing [21,22].

Although numerous efforts have been undertaken to understand nanoscale phase separation and morphology optimization in spin cast polymer films for OSC devices [15–23], more efforts are needed for controlling/optimizing the film morphology in doctor blading process in large area fabrication. Morphology difference may be significant in the BHJ layers fabricated using different solution process methods, e.g. doctor blading and spin coating processes. Understanding the morphology difference induced by coating methods will be helpful for process development of large area, roll-to-roll fabrication of OSCs. In this paper, the morphology in a doctor bladed BHJ film of poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C61 (PCBM) blend was studied and compared to that of the spin coated film. It was found that films deposited using doctor blading resulted in a more uniform vertical phase distribution of the P3HT and PCBM components and higher ambipolar charge carrier mobility in the BHJ layer as compared to films prepared by spin coating. The inverted OSC device with P3HT:PCBM active layer fabricated by doctor blading achieved higher photocurrent and PCE as compared to the device with a spin coated active layer. We believed that this arose from a better controlled morphology and optimum nanoscale phase separation in the doctor bladed active layer.

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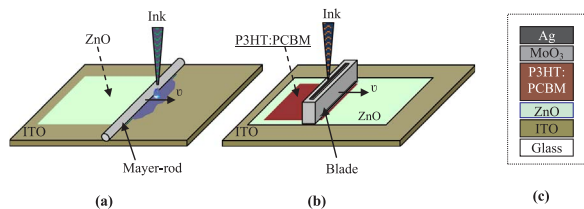


Fig. 1. Schematics showing (a) Mayer-rod coating of ZnO film on ITO substrate, (b) subsequent doctor blading of P3HT:PCBM film, and (c) the structure of the inverted OSCs (glass/ITO/ZnO/P3HT:PCBM/MoO₃/Ag).

2. Experimental procedure

ITO-coated glass substrates were cleaned by detergent, de-ionized water, acetone, and iso-propanol in an ultrasonic bath for 10 min each. After that, a thin ZnO layer (25–30 nm) was deposited on the top of ITO via Mayer-rod coating with the ZnO ink prepared by dissolving 0.45 M of ZnO powder in aqueous ammonium solution (29 wt%). The coating was conducted in a Coatmaster-510 system (ERICHSEN LABTECH) at a heated substrate temperature of 40 °C. The ZnO film thickness was controlled and determined by the moving speed of the rod. The coated ZnO film was annealed in air at 150 °C for 10 min. Next, the P3HT:PCBM active layer was deposited on top of the ZnO layer by either spin coating or doctor blading a solution containing 25 mg/ml of P3HT:PCBM (1:0.8 wt./wt.) blend in a solvent mixture of chlorobenzene (CB) and 1, 2-dichlorobenzene (DCB) (CB:DCB = 3:1 v/v). The spin coating was conducted in a N₂-filled glovebox with a spin speed of 500 rpm for 60 s. The doctor blading was performed in ambient conditions in a fume hood with the Coatmaster-510 system, at 20 mm/s coating speed and a 60 μm gap (between the substrate and die). All the P3HT:PCBM films were dried in N₂ glovebox at 80 °C for 2 h. For some films, further thermal annealing was undertaken at 150 °C for 10 min. Finally, a MoO₃ layer (10 nm) and Ag layer (100 nm) were sequentially deposited on the BHJ layer by vacuum thermal evaporation. A metal shadow mask was used for the Ag deposition to define the device area of 12 mm². Fig. 1 illustrates the coating schematics of (a) ZnO film and (b) P3HT:PCBM film. The layer structure of the inverted OSC devices (Glass/ITO/ZnO/P3HT:PCBM/MoO₃/Ag) is also sketched in Fig. 1c.

The film absorbance spectra were recorded using a UV–visible-near IR scanning spectrophotometer (UV-3101PC). Film thicknesses were measured using a surface profiler (KLA-TENCOR P-10). The surface morphologies of the active layers were studied using atomic force microscopy (AFM) (SII SPI3800). Depth profile analysis of the BHJ layers and devices was performed using a time-of-flight secondary ion mass spectrometer (TOF-SIMS) (ION-TOF GmbH, TOF-SIMS IV), with 3 keV Cs sputtering beam on a 200 × 200 μm² surface area of the sample and 25 keV Ga analysis beam. The current-voltage (I-V) characteristics of

devices were measured with a Keithley 2400 source meter in dark and under 100 mW/cm² (AM1.5 G) illumination using a 300 W solar simulator (Newport 91160). The intensity of the light source was determined by a Si reference cell (calibrated by Fraunhofer Institute for Solar Energy, ISE) and corrected according to the spectral mismatch calculation. The double injection (DoI) current transient spectroscopy measurements were used to further understand the charge transport characteristics in the devices. The DoI current transients were measured by applying square shaped voltage pulses to the devices in forward bias condition. The time derivative of the injected current transients were calculated in order to estimate the charge carrier transit time and mobility. The details of the DoI measurement are elaborated elsewhere [24,25].

3. Results and discussion

To obtain uniform polymer-fullerene films over a large area, the solvents used for P3HT:PCBM and the corresponding drying process for doctor blading were investigated and optimized. Chlorobenzene (CB) and dichlorobenzene (DCB) were used for dissolving P3HT and PCBM as they have high solubility in the chlorinated solvents. However, the use of pure CB or pure DCB resulted in non-uniform films, as seen in Fig. 2a and b where coloured patterns and coffee-ring like circles are observed. It was found that the coating of CB-based polymer solution had a fast drying time of less than 5 s. Drying a polymer film too fast may cause uneven film morphology as insufficient time is given for self-flattening of the wet film surface. On the other hand, the coating of DCB-based polymer solution had a much slower drying time of more than 40 s. Agglomeration and dewetting were observed in the slow drying film, giving rise to poor film morphology. An optimized mixture of CB/DCB (3:1 v/v) was found to be ideal in controlling the film drying process and hence the film morphology. This enabled us to obtain a uniform P3HT:PCBM film (Fig. 2c). The film thickness was controlled by solution concentration, gap separation between the substrate and die, coating speed and substrate temperature. A uniform P3HT:PCBM film with 200 nm thickness was achieved by doctor blading a solution of 2.5 wt% P3HT:PCBM in CB:DCB (3:1) at a heated substrate temperature of 40 °C, with a 20 mm/s coating speed and a 60 μm gap (between the substrate and die). An increased air flow was introduced after the coating, to further control the film drying time (~10 s). The second step of fast drying by air flow ensured a uniform drying over the large area and also to prevent dewetting and agglomeration. For comparison, using the same batch of P3HT:PCBM solution, spin coating process was also used to fabricate the BHJ layer with similar thickness. The spin coating was done in a N₂-filled glovebox at 500 rpm for 60 s on a heated substrate of 40 °C, resulting in a film thickness of 200 nm. By visual inspection, the spin coated film appeared uniformly coated. However, optical characteristics of the doctor bladed and spin coated

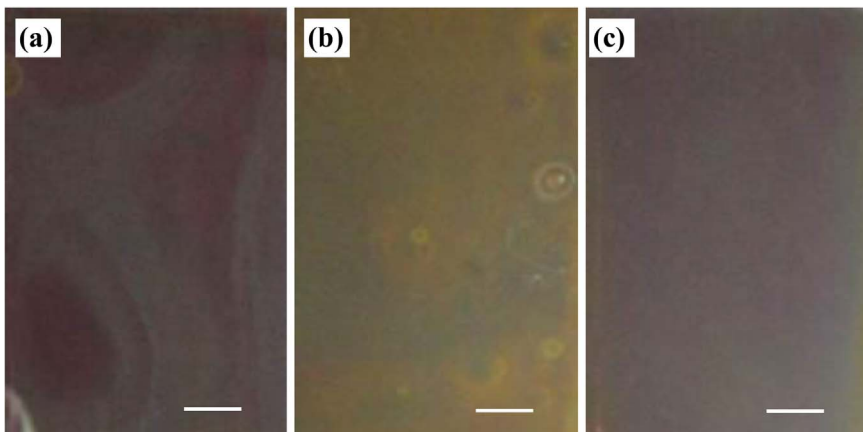


Fig. 2. Photo images of the P3HT:PCBM films fabricated by doctor blading with (a) CB, (b) DCB and (c) CB:DCB = 3:1 as the solvents. Scale bar: 5 mm.

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