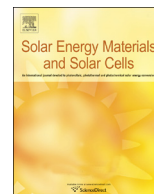




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A quasi-ordered bulk heterojunction of P3HT/PCBM solar cells fabricated by zone-casting



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ABSTRACT

In this paper, a quasi-ordered bulk heterojunction of P3HT/PCBM solar cells (Q-O BHJ device) was fabricated by zone-casting from the solutions with different aging times. P3HT nanofibers aligned in an oriented manner due to the shear stress and solute concentration gradient during zone-casting process. The orientation degree of P3HT nanofibers increased when the aging time of solution was prolonged. Meanwhile, P3HT had a high crystallinity due to the slow evaporation rate of solvent during the film-forming process. The oriented alignment of nanofibers could facilitate the connection between nanofibers by increasing the bridging chains. Furthermore, the bridging chains tended to adopt an untwisted conformation, which results in a reduced energy barrier of hole transfer. As a consequence, the Q-O BHJ device had a higher fill factor (*FF*) and short-circuit current density (J_{sc}) compared with the device fabricated by spin coating (BHJ device), leading to an increase by almost more than 50% in device efficiency and a PCE of 3.26% without any annealing treatment.

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1. Introduction

Polymer solar cells (PSCs) are currently being investigated as a potential low-cost, lightweight and flexible large-area candidate for sustainable solar energy conversion [1–4]. Since the original work of Tang [5], efforts have been made in the development of solar cells based on conjugated polymers. In particular, polymer bulk heterojunction (BHJ) solar cells based on interpenetrating networks of electron-donating material and electron-accepting material with a large donor/accepter interface have gained considerable interest [6–9].

Employing BHJ structure, device performance of solar cells depends critically on processing techniques which affect the blend morphology of active layer. So far, all published work concerned with BHJ has been done using spin coating, spray coating, doctor blading, screen printing or inkjet printing [10]. Among these film-forming techniques, the most attractive PSCs BHJ model, the blend of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61butyric acid methyl ester (PCBM) have an active layer morphology in which the P3HT self-organizes into an interconnected nanofiber network surrounded by a continuous PCBM phase. However, P3HT nanofibers are dispersed in a disordered manner in this structure, which is not suitable for the charge transport due to grain

boundary [11]. According to recent studies, charge transport is dominated by grain-boundary effects in semicrystalline and polycrystalline materials [12]. In order to reduce the activation energy for charge transport, one can increase the amount of bridging chains in disordered regions, which act as tie molecules and link different ordered nanofibers [11,13]. In addition, the conformation of bridging chains also has a great influence on the carrier mobility. If bridging chains are forced to undergo sharp bends or twists when they cross grain boundaries, there may be localized states associated with breaks in conjugation electron cloud, adverse to the carrier transport [12]. However, in P3HT/PCBM active layer fabricated by conventional techniques mentioned above, disorderly dispersed nanofibers restrict the quantity of bridging chains and also increase their distorting extent, which increase activation energy of charge transport across grain boundary [14,15]. As known in P3HT/PCBM system, hole mobility is lower than electron mobility. As a result, the space charges would deteriorate the photocurrents of the BHJ device [16,17]. Therefore, increasing the carrier mobility of P3HT is a key to improve device performance. Fortunately, researchers have shown that crystals aligned in an oriented manner could facilitate the connection between adjacent crystals by increasing bridging chains with an unbenet conformation [18]. Zone-casting, also referred to as directional casting, is a commonly used technique for deposition of functional materials and can provide an easy deposition of polymer films over a large area [19,20]. In addition, zone-casting is very promising because it could induce a high orientation

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degree with no need for special substrates, external fields or exotic treatments after casting [20]. However, to the best of our knowledge, no researchers have reported on efficient polymer solar cells fabricated by zone-casting.

In this contribution, we combined the zone-casting technique with aging solutions to fabricate P3HT/PCBM active layer. During the film forming process, induced by shear stress and concentration gradient, P3HT nanofibers aligned in an oriented manner and dispersed in PCBM matrix, which we call as quasi-ordered bulk heterojunction (Q-O BHJ). In Q-O BHJ, the uniaxially aligned nanofibers facilitated an increase of P3HT chains bridging neighboring nanofibers and decreased the distortion of the bridging chains, which would reduce the transport barrier across grain boundaries. In addition, the crystallinity of P3HT in Q-O BHJ was also higher than that in BHJ fabricated by spin coating because of slower evaporation rate of solvent in zone-casting process. Compared with the reference BHJ fabricated by spin coating, the hole mobility of Q-O BHJ device was improved, resulting in a higher fill factor (*FF*) and short-circuit current density (J_{sc}). As a consequence, the Q-O BHJ yielded solar cells with a PCE of 3.26% without any thermal or vapor annealing treatment, which increased by almost more than 50% compared with the reference BHJ device (PCE=2.2%).

2. Experimental section

2.1. Materials

Regioregular HT-HT P3HT (>98.5%; HT: head-to-tail; M_w =68 kDa) was purchased from Nichem Fine Technology Co. Ltd. PCBM (99.0%) was purchased from Solenne Co. Ltd. Toluene (Tol), tetralin (THN) and chloroform (CF) were purchased from Beijing Chemical, China. THN was purified by column chromatography (neutral aluminum oxide), and rest of the materials were used as received.

2.2. Preparation of P3HT/PCBM solution and film

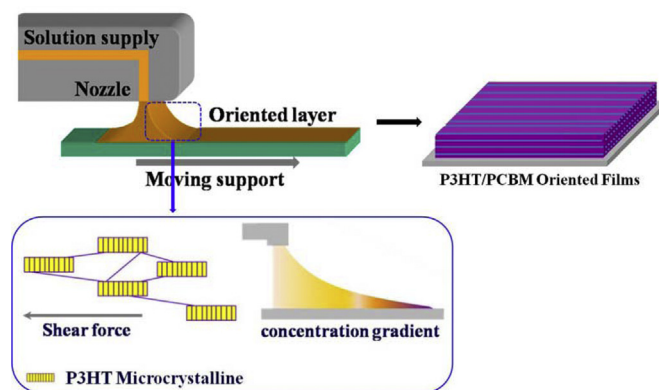
First, P3HT and PCBM were blended in a 1:0.8 weight ratio and dissolved in a CF/Tol/THN solvent mixture (the ratio of the solvent volume was CF:Tol:THN=3:2:1) at 27 mg/ml. Then the solution was stirred at 70 °C for 30 min in a nitrogen-filled glovebox to achieve ample dissolution. After that, the solution was stored at 15 °C for different aging times (1 h, 2 h or 3 h). The resulting solution was used to prepare the P3HT/PCBM films via spin coating or zone-casting for studies. It is noted that the P3HT/PCBM solution was deposited without any filtration.

Zone-casting consists in the deposition of P3HT/PCBM from solution on a moving substrate [20]. The casting process is schematically presented in Scheme 1. The detail description of zone-casting process and the photo of zone-casting equipment are shown in supporting information (Fig. S1). The solution is continuously supplied to the evaporation zone through a flat nozzle. The solvent evaporates from the meniscus zone and the solute is deposited on the moving substrate. During the film forming process, P3HT/PCBM films were prepared by zone-casting solutions onto substrate with a solution supplying rate of 20 μ L/s and a substrate velocity of 60 μ m/s at 40 °C by using a self-made devices. The zone-casting process was executed in ambient.

The spin-coating P3HT/PCBM films were prepared using a commercial spin-coater (KW-4A, chemat Technology Inc). The spin coating process was executed in ambient.

2.3. Fabrication of solar cells

Solar cell devices were built on transparent indium tin oxide (ITO) coated glass substrates with a sheet resistance of 10 Ω^2 .



Scheme 1. Schematic illustration of the zone-casting process and the driving force of film orientation: shear force from the nozzle and the concentration gradient of solute.

Glasses were cleaned for 10 min in acetone and another 10 min in isopropyl alcohol in an ultrasonic bath. Before use, substrates were surface treated for 15 min in an ozone oven. Subsequently, a 25 nm thin layer of poly(3,4-ethylene dioxythiophene) doped with polystyrene sulfonic acid (PEDOT:PSS, Baytron P4083) was deposited by spin coating on top of the ITO bottom electrode and baked at 140 °C for 10 min in ambient. A 240-nm-thick P3HT/PCBM layer was deposited on top of the PEDOT:PSS layer from the prepared P3HT/PCBM solutions by zone-casting/spin coating in ambient. Finally, the samples were transferred into an evaporator and pumped down to 2×10^{-4} Pa to deposit 30-nm-thick calcium (Ca) and 100-nm-thick aluminum (Al) cathodes, respectively. The active surface area of a typical device is 0.09 cm².

2.4. Characterization

Viscosity measurements of solutions with different aging times were carried out with ARES G2 rotational rheometer. The solution was fed into the cylinder rheometer and measured under constant angular shear frequency ($\omega=1 \text{ s}^{-1}$) at 25 °C.

UV-vis absorption spectroscopy (UV-vis) of the solution was recorded using a Lambda 750 spectrometer (Perkin-Elmer, Wellesley, MA).

The orientation of the P3HT/PCBM films was proven by cross-polarized optical microscopy and polarized UV-vis absorption spectroscopy. Cross-polarized optical microscopy (Carl Zeiss A1m microscope) was used to record the brightness of the films at different angles. To calculate the order parameter of P3HT/PCBM films quantitatively, polarized UV-vis absorption spectroscopy was also recorded with a Lambda 750 spectrometer (Perkin-Elmer, Wellesley, MA) equipped with a green prism.

The morphology of the P3HT/PCBM films was characterized using transmission electron microscopy (TEM). TEM was used to probe the vertical direction information of the P3HT/PCBM film. Images were obtained using a JEOL JEM-1011 transmission electron microscope operated at 100 kV. Thin films were first floated on deionized water and then transferred onto a copper grid. The samples were dried at room temperature for 12 h before TEM experiments.

The crystal structure of P3HT was analyzed using out-of-plane grazing incidence X-ray diffraction (GIXD) measurements. The GIXD profiles were obtained by using a Bruker D8 Discover reflector with an X-ray generation power of 40 kV tube voltage and 40 mA tube current. The measurements were obtained in a scanning interval of 2θ between 4° and 30°. To increase the GIXD peak intensity for investigating the crystallinity and orientation

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