



Natural drying effect on active layer for achieving high performance in polymer solar cells



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ABSTRACT

The self-organization of the polymer in solar cells based on regioregular poly(3-hexylthiophene)(RR-P3HT):[6, 6]-phenyl C61-butyric acid methyl ester is studied systematically as a function of the room temperature (RT) (varied from 300 K to 290 K). Optimal self-organized structures within the RR-P3HT:PCBM films are achieved by varying spin speed and time as well as the temperature at which the spin casting process occurs. These blend films are characterized by UV–vis absorption spectroscopy, atomic force microscopy, and X-ray diffraction measurements. The optimum device efficiency can be achieved in naturally dried devices when spin coating within the temperature range 292–294 K. Both the power conversion efficiency (PCE) and fill factor (FF) of the optimum devices show a plateau region, with PCEs exceed 4% and FFs close to 0.70. For RT < 290 K, the corresponding devices show a wide distribution of performance parameters for the unhomogeneous active layer. While for RT > 296 K, the short current density, FF and PCE of the corresponding devices are gradually decreased, suggesting that there is a major change in the ordered structure of the polymer. Based on the results, it is demonstrated that high performance device can be achieved just by natural drying the active layer at RT condition in air condition without further thermal treatments.

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1. Introduction

Bulk heterojunction (BHJ) polymer solar cells (PSCs), whose active layer is comprised of semiconducting polymer and fullerene, afford the potential advantages of easy processability, low material costs and mechanical flexibility [1–6]. Generally, the performance of BHJ PSCs can be maximized by controlling the active layer morphology, because efficient photoinduced charge generation, transport, and collection at each electrode chiefly depend on the nanoscale morphology of the active layer [2,3]. Charge generation takes place at the donor/acceptor heterojunction, so the donor and acceptor need to be well-mixed so as to maximize the interface area of the heterojunction. On the other hand, charge transport requires continuous donor and acceptor domains throughout the active layer, so that electrons and holes can flow smoothly to their respective electrodes. Particularly, the transport of holes is often

the factor that limits the obtainable photocurrent, as the hole mobility of most used donors is lower than the electron mobility of commonly used acceptors [4–6]. Therefore, the optimal morphology of the active layer has been recognized as one that contains large donor/acceptor interface area, continuous donor and acceptor domains, and high molecular ordering or crystallinity of the donor.

Over the last decade, many research groups have systematically analyzed external treatments in order to optimize the BHJ morphology of regioregular poly(3-hexylthiophene) (RR-P3HT):[6, 6]-phenyl C61-butyric acid methyl ester (PCBM) films for high-efficiency solar cells. These treatments enable both components to evolve self-organized crystallization and achieve a well-ordered interpenetrating network of individual materials. The most notable of these treatments involve: (1) post thermal annealing (TA) of the active layer to activate rearrangement of the morphology [7–12]; (2) slowing down the drying rate (solvent annealing, SA) to allow the active layer to a desirable morphology [13–18]; and (3) the use of cosolvent and additive to manipulate the active layer morphology [19–22]. Although impressive efficiencies are achieved with these morphology-controlling techniques, the optimal

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morphology can only be attained with careful control of the processing conditions. Consequently, implementations of identical processing conditions have often resulted in wide-ranging device performance values and time-consumed. Furthermore, all highest-performance devices have been fabricated and characterized entirely in inert atmospheres [7–11,13–19], suggesting the importance of device processing in an oxygen-free environment. However, it is difficult for most laboratories to maintain the oxygen free environment in the whole fabricating process. On the other hand, device fabrication in air condition may decrease the cost for mass production of organic solar cells.

In our previous report [23], high-performance and air-processed polymer solar cells were achieved by room temperature drying of the active layer. In this paper, we present a systematic study of the natural drying effect of the BHJ active layer afforded by spin-coating speed and time at room temperature (RT). Similar to TA and SA approach, we use the term “natural drying (NA)” to describe this process here. The RR-P3HT:PCBM active layer was investigated by NA treatment combined with the variable spin-coating speed (S_s) and time (T_s) from 300 K to 290 K. By systematically varying S_s and T_s at different RT, we have conducted a comprehensive investigation of the evolved morphology of the RR-P3HT:PCBM films and the performance of the PSCs. The solvent evaporation rate was controlled by RT, S_s and T_s . The spin coating conditions at different RT were then optimized in terms of the cell efficiency. Post SA was applied in conjunction with the NA process to further manipulate the morphology. The resultant morphology was analyzed with UV–vis spectroscopy, X-ray diffraction (XRD), and atomic force microscopy (AFM), which was in turn correlated to the device characteristics.

2. Experimental details

2.1. Device fabrication

The layer structure of the solar cells was glass/indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS)/P3HT:PCBM/LiF/Al. PEDOT:PSS (Baytron P VP Al 4083), regioregular (90–93%) P3HT (MW = 68000 g/mol, purity: 99%), and PCBM were purchased from H.C. Starck, Rieke Metals, Inc., and Nano C, Inc., respectively. All materials for the fabrication of the polymer solar cells were used as received. Before device fabrication, the ITO coated glass substrates were cleaned by sequential ultrasonic treatment in detergent, deionized water, acetone, and isopropyl alcohol. A thin layer (~40 nm) of PEDOT:PSS was spin-coated to modify the ITO surface and then the substrates were baked at 403 K for 20 min. The blend solutions of P3HT and PCBM (weight ratio of 1:0.8, 20 mg/ml in chlorobenzene) were spin-coated onto PEDOT:PSS-coated ITO glass substrates in air condition. LiF (0.8 nm)/Al (100 nm) electrodes were deposited by thermal evaporation under high vacuum with shadow masks determining the photoactive areas (6 mm²).

2.2. Natural drying

Natural drying treatment of the P3HT:PCBM composite films were carried out before the deposition of the LiF/Al electrodes. The P3HT:PCBM solution was stirred on the heating plate for ~24 h at 283 K. In order to achieve the uniform films, prior to spinning the substrate was static and coated with the P3HT:PCBM solution completely. Immediately, the active layer was obtained by spin-coating the solution in a single spinning process with a fixed spinning speed. This process was performed at different RT from 300 K to 290 K, as measured by a hygrothermograph (Testo, Ger.) in the open lab. The resulting P3HT:PCBM film was then natural

drying at RT after spin coating. The S_s and T_s of the spin coating process were alternatively changed to optimize the device performance at a given temperature. As an example, spin coating for 10 s at 300 rpm and spin coating for 12 s at 230 rpm may result in the same device performance for natural drying at the same RT. For solvent annealing, the spin coated films (either liquid or solid, depending on S_s , T_s and RT) were then left in a grease-sealed cylinder (volume: 10 cm³) at the same RT. At the same time, small amounts of chlorobenzene solvent were added into the cylinder with a lid in order to prevent rapid evaporation of the solvent.

2.3. Device characterizations

The P3HT:PCBM composite films used for optical and morphological characterization were prepared using the same method as used for the device fabrication. The samples for absorption measurements were prepared by the same procedure before the cathode deposition step, and then measured using a Varian Cary 5000 UV–vis spectrophotometer. AFM images were obtained with a Digital Instruments Multimode scanning probe microscope operated in the tapping mode. All the films with the same thickness as measured by a Dektak profilometer, were prepared for measurement. The current-density–voltage (J – V) characteristics of the devices were measured with a Keithley 2400 source meter under simulated AM1.5G irradiation (100 mW/cm²) in air condition. For J – V measurements, the light intensity was calibrated using a reference silicon solar cell. The hole mobility of the active layer was obtained from the dark J – V characteristics of hole-only devices operated in the space-charge limited current (SCLC) regime, where the devices were obtained by replacing the LiF/Al cathode of the regular devices with thermally evaporated MoO₃/Ag.

3. Results and discussion

The experiments have been designed based on several considerations. Firstly, both fast drying and slow drying processes transform the polymer solution into a solid film. To obtain optimized performance, TA is commonly performed after fast drying the film, which requires the inert atmosphere [7–11]. NA is free of thermal treatment after fast drying the film, so the detrimental reactions can be avoided during post treatment. Therefore, the high performance can be expected to achieve in air condition. Secondly, the P3HT chains would induce the self-organization or crystallization by slow drying the film, which results in the improved performance [13,14,18]. The evaporation rate of the solvent is subject to the influence of temperature, the vapor pressure, the surface to volume ratio, the flow rate of air flowing through the surface. Therefore, it is realizable that the P3HT self-organization is performed by controlling RT, the selected solvent, S_s and T_s . Thirdly, chlorobenzene (CB) has been deliberately chosen as the solvent because its moderate boiling point allows a wide window of spin-coating process at RT condition. By systematically varying S_s and T_s at different RT, we have conducted a comprehensive investigation of the evolution of the RR-P3HT:PCBM films and the device performance.

At elevated temperature, the solvent will be removed more rapidly and the film will lose its crystallinity. Here, the representative experimental conditions were chosen and the corresponding data are presented. RT was changed from 300 K to 290 K. The four combinations of S_s and T_s ((800 rpm, 30 s), (500 rpm, 20 s), (300 rpm, 10 s) and (230 rpm, 12 s)), which were named by #1, #2, #3, and #4, respectively, are selected. At the same time, the SA treatment was also performed for comparison. The SA process was described in the experimental section. RT was first fixed at 298 K, and conducted a comprehensive investigation of the evolution of

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