

## Performance of polymer solar cells fabricated by dip coating process

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### ABSTRACT

We demonstrate the use of dip coating technology as a fabrication tool for highly efficient polymer solar cells. We investigate the critical parameters for dip coating deposition of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C<sub>61</sub>-butyric acid methyl ester (PCBM) to obtain optimized active layers. These parameters include the selection of organic solvent, solution concentration and room temperature. Structure and morphology of dip coated films are characterized via X-ray diffraction, UV–visible absorption spectra and atomic force microscopy.

We achieve dip coated devices with power conversion efficiency of 3.93%, with fill factor, short circuit current and open circuit voltage of 0.63, 10.62 mA cm<sup>-2</sup> and 0.60 V, respectively, even though the devices were fabricated in air without any annealing treatment.

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### 1. Introduction

Polymer solar cells (PSCs) have recently attracted more attentions due to their advantages of low cost, ease of fabrication and their potential for the manufacture of flexible and large area solar cells [1–3]. In particular, polymer bulk heterojunction (BHJ) solar cells based on interpenetrating networks of an electron donor and an acceptor, resulting in an efficient photo-induced charge separation, have gained great interest. So far, the major issues in the field of PSC research have been focussed on two fields. First, the device efficiency must be improved for commercialization through a new material design. Second, a roll-to-roll compatible, high-yielding process must be developed for low cost. Great efforts have been made to improve the power conversion efficiency (PCE) of the PSCs. The device efficiency has exceeded to 7% based on new materials [4,5]. Therefore, along with the stable rise of efficiencies, the development of novel solution processes that are compatible with a roll-to-roll production is one of the crucial requirements for practical application.

For solution-processed solar cells, spin coating method is considered as the most reliable and reproducible process in the majority of the research for high-efficiency PSCs [6]. However, the spin coating process has several detrimental problems in its application to mass production. This method is only suitable for

a limited substrate size, with large materials waste and an incompatibility with roll-to-roll processing. For low cost production and commercialization, a high throughput, continuous process, and large area devices on flexible substrates are necessary to better utilize the great advantages of PSCs. Many deposition techniques for fabrication of PSCs have been demonstrated, including ink-jet printing [7,8], spray coating [9–11], screen printing [12–14], and doctor blading [15]. Each method possesses its own advantages better than the spin coating method.

Dip coating process is a commonly used method for conventional dyeing and can provide easy and fast deposition of polymer films over a large area. It may be a suitable technology for large scale production of organic solar cells. A particular advantage of dip coating process compared to other spray coating and inkjet print processes is the prompt and single pass formation of the film. The formed films are free-pinhole. Furthermore, the formation of the dip coated film is a slow natural drying process, offering the opportunity to develop an active layer morphology and obtain the desired film properties in PSCs. Previous, organic materials, such as poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C<sub>61</sub>-butyric acid methyl ester (PCBM), can be effectively crystallized when the solvents are slow drying by dip coating [16,17]. The utilization of the dip coating technique for the RR-P3HT:PCBM system can facilitate its self-assembly into a nanofibrillar lamellar structure after evaporation of the solvent. A power conversion efficiency of 3.6% using only this approach without thermal treatment has been reported [18].

The focus of this study was to investigate the parameters necessary to optimize the film quality for dip coating deposition

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methods with the goal to produce uniform, continuous films comparable to spin coated films. We employed P3HT and PCBM, currently the most widely used active layer in polymer photovoltaics. Films were characterized via X-ray diffraction, UV–visible absorption spectra and atomic force microscopy. We achieve dip coated devices with power conversion efficiency of 3.93%. As comparison, the drop coated and spin coated devices were also investigated. Efficiencies were 3.96% and  $\sim 0.51\%$  to 3.70% for spin and drop coatings, respectively.

## 2. Experimental details

ITO-coated glass substrates ( $R_s = 10 \Omega \text{ sq}^{-1}$ ) were cleaned with a special detergent followed by ultrasonication in acetone and isopropyl alcohol and then kept in a  $70^\circ\text{C}$  oven overnight. For spin coated devices, pristine PEDOT:PSS (Baytron P VP Al 4083) was spin coated with a thickness of  $\sim 40 \text{ nm}$  from an aqueous solution after passing a  $0.45 \mu\text{m}$  PTFE filter and subsequently dried at  $135^\circ\text{C}$  for 20 min in air. P3HT (Rieke Metals, 90–93% regioregular) and PCBM (Nano-C, 99%) were dissolved in chlorobenzene to make  $15 \text{ mg mL}^{-1}$  and  $12 \text{ mg mL}^{-1}$  solutions, respectively. These solutions were stirred for at least 24 h at room temperature and then were passed through a  $0.22 \mu\text{m}$  PTFE filter before spin coating. The active layers were fabricated at the rotated speed 230 rpm for 10 s. The active layers were natural slow drying under room temperature of  $20^\circ\text{C}$ . After complete drying of the active layer, a LiF/Al ( $0.8 \text{ nm}/80 \text{ nm}$ ) was thermally evaporated on the top of the active layer under vacuum lower than  $10^{-4} \text{ Pa}$ . The top metal electrode area, determining active area of the devices, was evaluated to be  $\sim 0.08 \text{ cm}^2$ .

For dip coated active layer, P3HT and PCBM were dissolved in chlorobenzene to make  $10 \text{ mg mL}^{-1}$  and  $8 \text{ mg mL}^{-1}$  solutions, respectively. First, the substrate coated PEDOT:PSS film ( $\sim 40 \text{ nm}$ ) was upright soaked fully, allowing full coverage of the P3HT:PCBM solution. Then, the substrate was removed slowly from the solution with a lifting rate of  $3 \text{ mm s}^{-1}$ , allowing the liquid flow by the self-gravity of the solution. The formation of the active layer is a slow natural drying process. After the solvent evaporates, a free-pinhole solid film was left on the substrate. The backside film of the substrate was carefully erased using the tampon moistened with alcohol. We judged the complete drying of the dip coated film by the transition of the film color. The liquid film was brown, and when the solvent was evaporated, the solidified film became purple. For drop coated active layer, the solution drop on the substrate coated PEDOT:PSS film followed by a natural drying. The following process for the finished device

fabrication was the same as the spin coated device. The spin coated, dip coated and drop coated devices were processed under ambient condition in air without any annealing treatment.

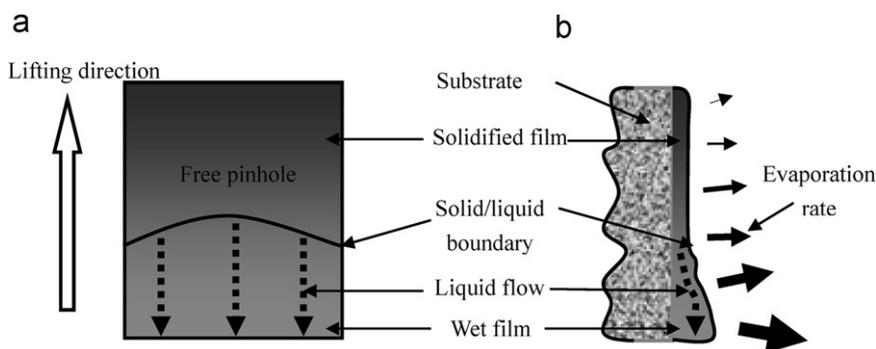
The UV–visible absorption spectra of dip coated or spin-coated P3HT:PCBM films were measured by a Cary spectrophotometer (Cary 5000 UV–vis). The morphology of the active layer films were observed by SEM (JSM-6700F) and AFM (Seiko SPA-400 SPM UNIT). The thickness of thin film was measured by a surface profiler (Veeco dektak 150 profilometer) and SEM. The crystallinity of active layer was investigated by X-ray diffractometer ( $\lambda_{\text{Cu}} = 1.542 \text{ \AA}$ ). The photovoltaic performance was measured under ambient condition in air. The current density–voltage ( $J$ – $V$ ) characteristics were measured with a Keithley 4200 source meter under AM 1.5 G ( $100 \text{ mW cm}^{-2}$ ) simulated by a Newport–Oriel solar simulator. The light intensity was calibrated using a certified photodiode and light source meter prior to each measurement.

## 3. Results and discussion

### 3.1. Film optimization

Dip coating is the single pass method for preparation films. The schematic evolution of the liquid layer during the slow natural drying process is illustrated in Fig. 1. In general, each film-preparation method requires a unique set of deposition parameters. The operating temperatures, solution concentrations, drying kinetics and optimal solvents are quite different from those in the spin coating method. The thin quality of the PSCs depending on the deposition parameters influences the performance of the PSCs. A new optimization is required for each method, though the other fundamental issues are still applicable.

In order to obtain the optimized quality of active layer for high performance device, the quality of the active layer covered with the substrate and the drying behavior of the coated film are controlled by the solvent selection, solution concentration and room temperature. We used different organic solvents for preparation the P3HT:PCBM film by dip coating. We find the chlorobenzene (CB) solvent is optimal when the experiment is conducted at room temperature ( $17$ – $22^\circ\text{C}$ ) in air. It is easy to cause the formation of liquid droplet and picture framing effect when the high boil solvent such as dichlorobenzene (DCB) was used. Even the macroscopically uniform film was hard to obtain when the improper solvents were employed. We found that the room temperatures above  $22^\circ\text{C}$  can result in the accumulation of the P3HT:PCBM in the center of the substrate when the CB solvent



**Fig. 1.** Schematic evolution of the liquid layer during the slow natural drying process: (a) top view of the coverage of the substrate and its surroundings, together with the involved solidified film and wet film (liquid layer). The solid/liquid boundary is not level but lunular due to the different evaporation rate of the solvent and flow velocity of the solution. Both the evaporation rate of the solvent and flow velocity of the solution at the sides of the substrate are larger than that at the middle of the substrate. (b) Side view of the coverage of the substrate and its surroundings. The evaporation rate of the solvent was increased from the top to the bottom of the substrate, which was indicated by the size of the arrows. The purple (at the top of the substrate) is representative of the solid film and the brown (at the bottom of the substrate) is representative of the liquid film.

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