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Fully spray-coated inverted organic solar cells

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ABSTRACT

This study evaluated the possibility of utilizing a spray-coating method for all spray-coated inverted organic solar cells (IOSCs). The spray-coating process was applied to deposit electron selective, active and hole selective layers. The IOSCs fabricated by fully spray-coating method showed a power conversion efficiency of 3.17 and 1.33% at a cell area of 0.36 and 15.25 cm², respectively, under AM.1.5 simulated illumination, resulting in similar performances with that of spin-coated devices.

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1. Introduction

Development in the field of organic solar cells (OSCs) has been growing due to low production costs and roll-to-roll manufacturing on flexible substrate [1–8]. In recent years, the organic solar cell systems, poly(3-hexylthiophene) (P3HT) as electron donor and: [6,6]-p-phenyl-C61 butyric acid methyl ester (PCBM) as electron acceptor blends have been made and power conversion efficiency (PCE) of up to 5–6% [9–11]. Recently the highest PCE of 10.6% was successfully demonstrated a solution-processed tandem organic solar cells [12]. For solution processed cells, mostly OSC layers are fabricated by spin-coating method, however, it is not suitable for roll-to-roll fabrication and has large materials losses with a limited substrate size [6]. To overcome these problems, several groups have explored various coating techniques such as doctor blading, [13] screen printing, [14] and inkjet printing [15,16] and spray-coating [17–21].

The spray-coating technique with potential advantages of enabling a no-limitation in substrate size and low utilization of polymers can be a promising substitute for overcoming the drawbacks of the conventional spin-coating process. Outstanding of these merits, spray-coating method has been developed for the fabrication of OSCs with many advantages, such as large-area coating, uniform coating, roll-to-roll compatible and low-cost process [17,18]. Furthermore, the spray-coating method is able to access a broad spectrum of fluids with various rheologies, offering the opportunity to tune the system to deposit practically almost any kind of solution and obtain the desired film properties [20]. Recently, spray-coating technologies

have been reported the fabrication of conventional OSCs structure (ITO/hole selective layer/active layer/Al) and inverted OSCs (IOSCs) structure (ITO/electron selective layer/active layer/hole selective layer/Ag) focused on controlling the surface roughness and coating thickness of hole selective and active layers, resulting in similar device performance with spin-coated OSCs [17,19–25].

For the fabrication of all-printed OSCs, an inverted architecture is usually used to avoid the vacuum process for the deposition of the low-work function metal electrode (such as Al and Ca) in which a high work function anode (such as Ag and Au) to collect holes and electron selective layer on the surface of ITO to collect electrons are utilized [26–29]. One critical issue in this fabrication process is how to form hole selective layer, poly(3,4-ethylenedioxythiophene):poly(styrene-sulfonate) (PEDOT:PSS), on top of active layer by using spray-coating process with a good film quality. PEDOT:PSS was not coated well onto active layer because PEDOT:PSS aqueous solution is highly hydrophilic and P3HT:PCBM film is hydrophobic, resulting in very poor device performance.

This paper reports the fabrication of inverted OSCs (IOSCs) with a ITO/ZnO/P3HT:PCBM/PEDOT:PSS/Ag structure utilizing fully spray-coated electron selective, active and hole selective layers to demonstrate large-area, air-stable and highly efficient devices. The effects of the spray-coated layers on the device performance and life time were investigated, and the performance of the IOSCs with fully spray-coated layers was then compared with that of the all spin-coated device.

2. Experimental details

The IOSCs fabricated on the ITO-coated glass with a sheet resistance of $\sim 10 \Omega/\text{sq}$. First, ITO substrates underwent a routine

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cleaning procedure, which included sonication and repeated rinsing in acetone and isopropyl alcohol. The substrates were then dried in an oven at 120 °C and treated with UV-ozone for 5 min. The spray-coating conditions for three layers were optimized to minimize the surface roughness by varying the solution injection rate, carrier gas flow, nozzle to substrate distance and printing speed. In this study, the carrier gas flow and printing speed was optimized as 50 psi, and 6 cm/min at x -axis and 1800 cm/min at y -axis for the spray-coating of three layers, respectively.

For the preparation of the ZnO sol-gel solution, zinc acetate (16.40 mg, Aldrich) was dissolved in 2-methoxyethanol (100 ml, Aldrich) using a magnetic stirrer. Ethanolamine (5 ml, Aldrich) was then added to the zinc acetate solution and the resulting solution was kept at 60 °C for 1 h under ambient conditions with vigorous stirring. Before spray-coating, the ZnO solution was diluted with methanol (ZnO sol-gel: methanol=1:10) because a solution without dilution was not well distributed from the nozzle or the layer was not coated uniformly. The ZnO thin film as electron selective layer was spray-coated on ITO glass with a thickness of approximately 40 nm (in an injection rate of 0.5 ml/min and nozzle to substrate distance of 8.5 cm), followed by thermal annealing at 300 °C for 10 min. The P3HT:PCBM blend solution was prepared at 1:1 mass ratio in 1,2-dichlorobenzene (10 mg/ml P3HT and 10 mg/ml PCBM). The photoactive materials were spray-coated with a thickness of 180–320 nm in an injection rate of 0.2 ml/min and nozzle to substrate distance of 3.5 cm. Slow evaporation and pre-annealing (150 °C for 20 min on hot plate) were carried out in a glove box. A buffer layer of PEDOT:PSS (AI 4083, Stark):isopropyl alcohol (IPA) (PEDOT:PSS:IPA=1:6) was prepared using spray-coating with an injection rate of 0.5 ml/min and nozzle to substrate distance of 5 cm after passing through a 0.45 μ m filter with a thickness of approximately 20–100 nm on the temperature of 25–80 °C. The coated PEDOT:PSS film was dried at 150 °C for 1 min on a hot plate in a glove box. In control device, the 40-nm-thick ZnO, 270-nm-thick P3HT:PCBM and 40-nm-thick PEDOT:PSS layers were spin-coated with a spin speed of 1000, 600 and 5000 rpm for 40 s, respectively. Finally, a 120-nm-thick Ag electrode was evaporated at 3×10^{-6} Torr. The effective area of the active layer for the solar cell prepared in this approach was 0.36 cm², which was determined using a shadow mask. The fabricated IOSCs was characterized using restricted illumination by inserting a shadow mask to eliminate excess photocurrent from conductive PEDOT:PSS layer [30]. The current-voltage (J - V) characteristics were measured under AM 1.5 simulated illumination with an intensity of 100 mW/cm² (Pecell Technologies Inc., PEC-L11 model). The intensity of sunlight illumination was calibrated using a standard Si photodiode detector with a KG-5 filter. The J - V curves were recorded automatically with a Keithley SMU 2400 source meter by illuminating the IOSCs.

3. Results and discussion

A schematic diagram of the spray-coating apparatus is shown in Fig. 1(a). The spray-coating system has two nozzles as the core and clad. The core nozzle was connected to the injection pump for the solution (ZnO, P3HT:PCBM or PEDOT:PSS) and the clad nozzle was linked an N₂ for the carrier gas. A computer controlled xy -stage (nozzle and substrate moving stage) and injection pump allows a reproducible thickness of the deposited film [22,25]. The spray-coating conditions were optimized to minimize the surface roughness. The stage was vacuumed on top of hot-stage to control of substrate temperature. To compare the surface morphologies of each layer, atomic force microscopy (AFM) images were taken as

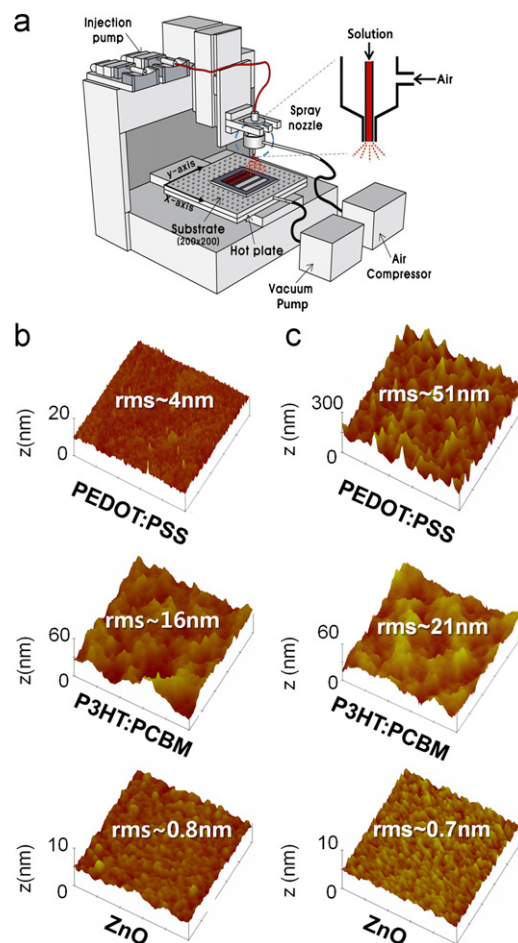


Fig. 1. (a) Schematic diagram of the spray-coating apparatus. AFM images and rms roughness of the three different layers in the inverted organic solar cell coated by (b) spin- and (c) spray-coating process. AFM image scans are $5 \times 5 \mu$ m.

shown in Fig. 1(b) and (c). The surface morphology and roughness of ZnO and P3HT:PCBM layers prepared by spray-coating method at 25 °C were similar in spin-coated layers. To form uniform PEDOT:PSS film on top of P3HT:PCBM layer in spray-coating process, the PEDOT:PSS was mixed with IPA solution and the substrate was heated up to 80 °C to improve wettability. The surface morphology of the PEDOT:PSS layer in optimized spray-coating process on top of the active layer shows root-mean-square (rms) roughness values of 4 and 51 nm using the spin- and spray-coating process, respectively.

Fig. 2 shows the photovoltaic response IOSCs produced with a different spray-coated P3HT:PCBM thickness with a spray-coated 40-nm-thick ZnO and spin-coated 40-nm-thick PEDOT:PSS. The thickness of the P3HT:PCBM layer in the IOSCs plays a major role in the device performance.[31] With increasing P3HT:PCBM thickness from 180 to 320 nm, the short circuit current density (J_{sc}) of the IOSCs increase from 7.74 to 9.61 mA/cm² with a constant open circuit voltage (V_{oc}) of about 0.59 V. The performance of the spray-coated devices does not decrease substantially with thickness and the PCE of the cells remains around 3.0% in the thickness of 270–320 nm. This thickness has relevant industrial applications, since it could increase the production yield and relax the constraints in thickness control [20]. The highest PCE of 3.12% is achieved at 270 nm, where a J_{sc} reaches 9.33 mA/cm² and the fill factor (FF) is 0.56.

In order to demonstrate fully spray-coated IOSCs, the PEDOT:PSS layer was spray coated onto spray-coated P3HT:PCBM and ZnO layers. Fig. 3 shows the effects of coating temperature on the

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