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Contents lists available at ScienceDirect

Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Enhancement of the performance of organic solar cells by electro spray deposition with optimal solvent system



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ARTICLE INFO

Article history:

Received 19 June 2013

Received in revised form

5 October 2013

Accepted 21 October 2013

Available online 23 November 2013

Keywords:

Electrospray

Organic photovoltaics

Additive

Electrical conductivity

Morphology

ABSTRACT

Electrospray (ES) as a thin film deposition method that is uniquely suited for manufacturing organic photovoltaic cells (OPVs) with desired characteristics of atmospheric pressure fabrication, roll-to-roll compatibility, less material loss, and possible self-organized nanostructures. The additional solvent with high electrical conductivity plays an important role in ES deposition process to fabricate OPVs with active layer composed of polymer mixture poly(3-hexylthiophene):[6,6]-phenyl C61-butyric acid methyl ester (P3HT:PC₆₁BM). Here we introduced acetic acid, which possesses high electrical conductivity, as additive solvent in ES process. The dependence of device performance on the concentration of acetic acid was investigated, and optimal ratio was obtained. To further demonstrate the influence of additive solvents with different electrical conductivity, OPV devices with active layer deposited by ES method using solutions containing acetic acid, acetone or acetonitrile were fabricated. The characteristics of active layers were revealed by optical microscope, atomic force microscopy, UV–vis spectroscopy and X-ray diffraction. Compared with additive solvents of acetone and acetonitrile, the active layer formed by electro spraying solvent containing acetic acid demonstrated enhanced vertical segregation distribution and improved P3HT crystallinity, which resulted in better device performance. OPV device using acetic acid as additive achieved power conversion efficiency (PCE) of $2.99 \pm 0.08\%$ under AM 1.5 solar simulation, which is on par with that of the spin coated device ($PCE 3.12 \pm 0.07\%$).

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1. Introduction

Organic photovoltaics (OPVs) have gained broad interest in the past two decades due to its potential to fabricate flexible, lightweight, low-cost and large-area photovoltaic devices. Most recently, power conversion efficiency (PCE) of OPVs has achieved over 8% [1,2]. To date, the reported OPVs with high efficiencies

have been fabricated by spin coating, which is a process only suited for laboratory scale and rigid substrate with significant material waste [3–5]. Therefore, there is an urgent need to develop fabrication methods that are scalable and roll-to-roll compatible.

Recently, electro spray (ES) has emerged as an attractive approach for fabricating OPVs. ES is an electrohydrodynamic liquid atomizing technique that can generate monodisperse droplets with diameter of a few nm to 100 μm. Studies on using ES as deposition method in the field of organic thin-film electronic devices have been reported in fabrication of organic light-emitting diodes (OLEDs) [6,7] and organic photoconductive devices [8]. These studies showed that ES was competitive to other solution-process techniques for organic thin-film electronic device fabrication. ES has several unique advantages for OPV production, such as compatibility with roll-to-roll process, operation in atmospheric pressure, less material loss, and possible self-organized nano-structures [9,10]. Most recently, ES has been used in OPV fabrication to deposit P3HT:PCBM active layers with

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bulk heterojunction [11–15] and multilayer structure [16], as well as to deposit poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) as hole transport layer [15] or electrode [17]. Notably, the structural and electronic properties study of electro-sprayed multilayer active film [16] showed ES is capable of creating organic thin films with complex heterostructures by using different materials and/or solvents, or differently aggregated phase of the same active material, which is in sharp contrast to spin coating. Cells fabricated by electro-spray have reached PCEs that are comparable to spin coated devices [10,15]. In addition, due to the Coulombic attraction force between the droplet and the conducting substrate, nearly 100% materials utilization rate is achievable, while spin coating suffers material loss as high as 95% in our spin coating tests.

Among numerous physical properties of the solution used in ES, the electrical conductivity plays the most critical role because it directly affects the droplet diameter and subsequently the film nanostructure and morphology. The most commonly used solvents to dissolve the OPV active materials are dichlorobenzene (DCB) and chlorobenzene (CB). However, both DCB and CB are non-polar and their electrical conductivities are too low for ES process. Therefore, small amounts of additives are usually used to boost the conductivity of the solutions. Typical polar solvents such as 1,1,2,2-tetrachloroethane (TCE) [10], acetone [11], dimethyl sulfoxide (DMSO), *N,N*-dimethylformamide (DMF), and acetonitrile [12] have been used as additives.

The polarity of the above mentioned additives is relatively weak and the electrical conductivity enhancements are still modest. This prompts us to consider organic solvents with strong polarity such as acetic acid, which has not been reported as conductivity booster for electro-spray fabrication of OPV devices. In this study, we used DCB as primary solvent for active materials, and acetic acid as solvent additive to fabricate the bulk-heterojunction active layers based on P3HT:PCBM blend. We optimized the concentration of the acetic acid, and also compared the effect of acetic acid with two other polar solvents (acetone and acetonitrile). We characterized the morphology, the surface roughness, the light absorption, and the nanoscale domain organization of the electro-spray fabricated films using various techniques including optical microscopy (OM), UV-vis spectroscopy, and high-resolution X-ray diffraction (XRD). We correlated the solution properties, morphology, and the current density–voltage (*J*–*V*) characteristics of the ES devices. We found the PCE of the ES OPV device fabricated in air was comparable to that of the devices fabricated in pure nitrogen environment.

2. Experimental

Patterned indium tin oxide (ITO) coated glass substrates were cleaned by sonication in a sequence of diluted Hellmanex solution, deionized (DI) water, acetone, and isopropyl alcohol (IPA) baths for 15 min each and then dried at 80 °C for 1 h prior to use. After drying, the substrates were UV/ozone treated for 10 min. Then the PEDOT:PSS (Clevios P VP Al 4083) solution was spin-coated at 4000 rpm for 60 s onto the ITO/glass substrates and dried at 120 °C for 10 min. The additives of acetone, acetonitrile or acetic acid were mixed with the P3HT:PCBM solution (with the ratio of 1 mg:0.8 mg in 1 mL of DCB) separately.

Fig. 1(a) shows the ES setup, which consisted of a syringe pump, a DC high voltage power supply, a metal nozzle and a substrate stage. The electro-spray nozzle was a metal needle with the tip tapered to 100 μm outer diameter (New Objective Company). The distance between the needle tip and the substrate was kept at 5 cm. A high voltage of 4–6 kV was applied between the nozzle and the substrate to generate stable electro-spray in the

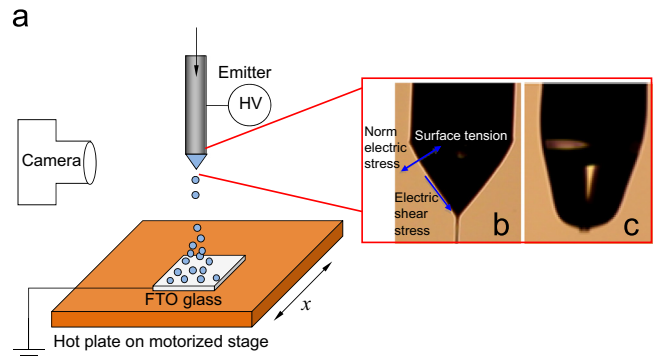


Fig. 1. (a) The electro-spray setup; (b) cone-jet mode in electro-spray; and (c) the unstable mode because of low electrical conductivity.

cone-jet mode. The P3HT:PCBM solution was injected through the nozzle at a rate of 5 μL min⁻¹. The ES process was performed in air at room temperature. For the spin coated reference devices, 20 mg of P3HT and 16 mg of PCBM were dissolved in 1 mL of DCB and then stirred for more than 12 h prior to use. The solution was spin coated on the PEDOT:PSS coated ITO/glass at 500 rpm for 120 s and dried for more than 2 h. Then one device is spin coated in a glove box filled with nitrogen (N₂) and another device is spin coated in air. All the samples were transferred in a thermal evaporator for metal electrode (100 nm of Al) deposition at the pressure of 4 × 10⁻⁶ mbar. The active area of each device was 9 mm². Then the devices were post-annealed at 120 °C for 10 min in the glove box. The current density–voltage (*J*–*V*) characteristics were examined using a Keithley 2400 source measuring unit under simulated AM 1.5 illumination (100 mW/cm²) with a solar simulator.

The conductivity of the solutions was tested by a megohmmeter. The optical microscopy and atomic force microscopy were used for the morphological investigation. The absorption spectra of the active films were obtained using a UV-vis-NIR spectrometer. The XRD results were obtained using a high-resolution X-ray diffractometer.

3. Results and discussions

3.1. Optimization of acetic acid concentration

To produce quasi-monodisperse droplet with electro-spray process, the liquid at the nozzle tip should take a conical shape termed cone-jet mode (Fig. 1(b)), which results from a balance between surface tension and electric stress normal at the liquid–gas interface [18,19]. The liquid electrical conductivity plays a key role in the cone-jet phenomenology. The cone-jet mode may not be obtained if the liquid electrical conductivity is too low with certain nozzle inner diameter and liquid flow rate. The electrical conductivity of the solution composed of 1 mg P3HT and 0.8 mg PCBM dissolved in 1 mL DCB in our experiment is only 1 × 10⁻⁷ S/cm and could not support the cone-jet mode (Fig. 1(c)). One way to increase the electrical conductivity is to mix another solvent with high electrical conductivity with the solution.

As a non-toxic solvent with high electrical conductivity (3.18 × 10⁻⁴ S/cm at 20 °C), acetic acid is a popular additive solvent to increase the solution electrical conductivity in electro-spray. When 5, 10 or 15 vol% acetic acid are added into the P3HT:PCBM:DCB (1 mg:0.8 mg:1 mL) solutions, the electrical conductivity of the solutions are increased to 1.55 × 10⁻⁶, 2.46 × 10⁻⁶ and 3.41 × 10⁻⁶ S/cm, respectively, and a stable cone-jet mode can be obtained. Concentration of 20 vol% or more resulted in active

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