



Stability enhancement of polymer solar cells in trilayer configuration



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ABSTRACT

Inverted organic solar cells by bulk-heterojunctions were fabricated using TiO₂ and ZnO as electron extraction layers. Electrospray deposition was applied to fabricate trilayer structures with thin donor and acceptor layers sandwiching bulk-heterojunction layers. Comparing to single layer devices, trilayer devices show significantly enhanced stability in device performances under continuous illumination in air. Conventional structure devices with Al as electron extraction layer were also compared showing how the improvement achieved with the inverted architecture is surprisingly higher. Our results, on a basis of 4 h of continuous operations, show how combining inverted device architecture with trilayer configuration is a viable approach in achieving highly stable organic solar cells with 70% retainment of the initial PCE to be compared to the 13% in the case of direct trilayer BHJ.

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1. Introduction

As a cheap alternative for clean energy, organic solar cells have been a topic of intense research in the last decade. Material engineering and better understanding of the device operation mechanism have led to steady improvement of the device performance reaching above 10% power conversion efficiency (PCE) [1]. However, stability of device performance still remains an important issue. In common architecture of bulk heterojunction conventional cells, donor materials such as poly(3-hexylthiophene) (P3HT) and acceptor materials such as (6,6)-phenyl C61 butyric acid methyl ester (PCBM) are blended to form mixed layers and deposited on the hole transporting poly(3,4-ethylenedioxythiophene):polystyrene sulfonic acid (PEDOT:PSS) buffer layer. Low work function metals (such as Al, Ca/Al) are used as the electron collecting layer on the top of active layer [2]. Both PEDOT:PSS and low work function metals are not stable in air because of etching and oxidization. In order to avoid these two shortcomings, inverted device architectures were proposed where high work function transition metal oxides, covered by stable Au or Ag were used as the hole extracting electrodes, while suitable oxide layers such as TiO₂ and ZnO were used as electron extracting electrodes [3–6] (cathodes) by depositing them on tin-doped indium oxide (ITO). Consequently, inverted devices show much better device operation stability compared to conventional cells [7].

Bulk heterojunction photoactive layer is another component which is sensitive to ambient conditions and interfacial degradation in direct contact with electrodes. Protecting it with pure donor or acceptor layers can be envisaged. However, realizing such a multilayer film configuration is not trivial using common solution processing techniques.

A possible solution is the use of the electrospray deposition (ESD) technique as a semi-wet processing technique, which can be widely applied in various organic materials into current material technologies, micro-electronics, and nanotechnology [8–19]. We recently used electrospray deposition (ESD) to acquire flexibility in polymer film fabrication [20–22,24,25]. The quite dry nature of the solutes while approaching the substrate allows deposition of subsequent layers from the same or different solvent avoiding orthogonal dissolution. Using electrospray deposition, recently we have reported trilayer solution processed polymer solar cells which exhibited significant better performance than single layer devices [22,25]. Here, we show that organic inverted solar cells with trilayer configuration have exceedingly better operational stability than their single layer counterparts and even more interestingly show an improvement more significant than that obtained with the trilayer direct cell.

2. Experimental

Poly(3-hexylthiophene) (P3HT) and (6,6)-phenyl C61 butyric acid methyl ester (PCBM) commercially available solar cell materials were purchased from Plextronics and Solaris respectively and were used

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without any further purification (see Fig. 1a and b for chemical structures). ITO-coated glass substrates were used as transparent electrodes ($10 \Omega/\text{sq}$, Kintec). Substrates were cleaned using standard procedures by immersing successively in solutions of acetone, ethanol, iso-propanol and de-ionized water in ultrasound bath for 15 min, followed by 15 min of UV-Ozone treatment. For conventional cells, hole transport buffer layer (30 nm) poly(3,4-ethylenedioxythiophene) doped with polystyrene sulfonic acid (PEDOT:PSS) was spin coated on ITO/glass substrates and subsequently dried in air for 30 min at 110°C . Pedot was purchased from HARAEUS with solid content of about 1%, conductivity of 0.1–1.0 S/cm and viscosity of 8–30 mPa s. Electro spray deposition setup used for active layers was described in detail in our previous works [20,21]. P3HT and PCBM (1:1 weight ratio) were dissolved in 1,2-dichlorobenzene with solution concentration of 0.5 mg/ml and deposited using flow rate of $1.3 \mu\text{l/s}$ and applied electric field of 2 kV. Films were pre-annealed in glove box at 150°C for 10 min before thermally depositing 100 nm thick aluminum electrode with a shadow mask to define an active device area of 8.6 mm^2 (see Fig. 1c for device structure). For trilayer devices, the same amount of total solution was used (2 ml), the ratio of used total solution for the separate layers of P3HT/P3HT:PCBM/PCBM was 0.3/1/0.3. The active layer thickness was about 90 nm (see Fig. 1d for device structure). For inverted cells, ZnO or TiO_2 , amorphous layer were processed at low temperatures for electron transport layers on ITO surface as shown in the following: 1) For ZnO, 0.75 M zinc acetate dihydrate and 0.75 M monoethanolamine in 2-methoxyethanol solution was spin coated at 2000 rpm for 40 s and then the films were immediately placed onto a hot plate that was pre-heated to 275°C and annealed for 5 min. The resulting ZnO films were rinsed in de-ionized water, acetone, and isopropanol and then dried to remove residual organic material. All these processes were performed in ambient condition. 2) For TiO_2 layer, absolute ethanol (10 ml) and an amount of $150 \mu\text{l}$ of tetraisopropylorthotitanate (TIPT) purchased from Sigma Aldrich were mixed together in nitrogen glove box. A quantity of $10 \mu\text{l}$ of HCl was added in air and then kept the resulting solution on roller overnight to ensure better mixing of solution. The solution was then spin coated on ITO substrates at 1000 rpm for 60 s and then kept in air for 90 min to dry the film. The substrates were then transferred to nitrogen glove box and pre-heated at 110°C for 10 min. As a result of the low temperature involved in the process both TiO_2 and ZnO films are amorphous.

Active layers were deposited in similar fashion as for conventional cells. Only in trilayer devices, thin P3HT and PCBM layers were switched as the charge collection directions are opposite in these two device configurations (see Fig. 1e and f). Finally, a 8 nm layer of MoO_3 as a buffer layer for hole collection, followed by 60 nm of Ag were deposited by thermal evaporation without any further treatment to complete the device.

Four single cell were fabricated per each $1.5 \times 1.5 \text{ cm}^2$ ITO coated glass substrate.

Atomic Force Microscopy studies were carried out using Digital Instruments Dimension 3100 in tapping mode. AFM quantitative morphological analysis was performed using WSxM software. Current-voltage (J–V) characteristics of devices were examined under inert atmosphere using a Keithley 4200 semiconductor analyzer under illumination of an AM 1.5 G set at 100 mW/cm^2 calibrated by an IL1400BL radiometer. Then the devices were tested in air under continuous illumination for 9 h without sealing and/or encapsulation.

3. Results and discussions

An inspection of device morphological quality is performed by looking at Fig. 2. In Fig. 2a) and b) and in Fig. 2c) and d) the single organic layer and trilayer on TiO_2 and ZnO respectively were reported. In particular it is shown how the trilayer architecture does not introduce rougher surfaces, but in some case like in the TiO_2 trilayer a smoother layer is obtained because Ti oxide layer maintains some crystalline reminiscence while grown at low temperatures. The rms roughness is always lower than 3 nm in trilayer devices.

In Fig. 3a) and b) are reported the energy diagrams for the inverted devices fabricated with a single and a trilayer architecture, where it is clear the role of pure P3HT and PCBM layers as protecting the oxidation of the active blend induced by the contact with the electrodes, and by hindering the reverse sign currents (holes collection from ITO and electrons from the metal).

J–V characteristics of ESD single layer and trilayer devices under illumination at AM 1.5 G (100 mW/cm^2) were presented in Fig. 4. We observed similar improvement with respect to the single layer by the trilayer devices in inverted architecture as in the direct case devices we reported in our previous study [22].

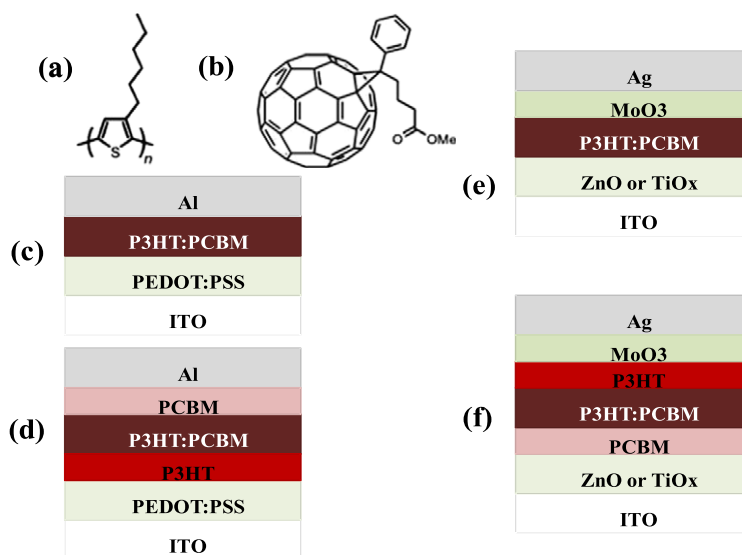


Fig. 1. Chemical structures of P3HT (a) and PCBM (b); solar cell device structures of conventional single layer (c), conventional trilayer (d), inverted single layer (e) and inverted trilayer (f).

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