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Ambient stable large-area flexible organic solar cells using silver grid hybrid with vapor phase polymerized poly (3,4-Ethylenedioxythiophene) cathode

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ABSTRACT

The presence of PSS in solution processed high conducting polymer PEDOT:PSS (PH1000) limits the reliability and lifetime of organic photovoltaic devices due to its acidic and hygroscopic nature. We have developed an alternative PSS-free transparent electrode, based on vapor-phase polymerized (VPP) PEDOT in combination with a current collecting silver grid. The hybrid electrode exhibits a low sheet resistance of 1.6 Ω/□ with an excellent bending proof performance. The power conversion efficiency (PCE) is 2.63% in a 1.21 cm^2 area device with a stacking structure of PET/Ag-grid/VPP-PEDOT/ZnO/poly[(9,9-bis(3-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)](PFN)/poly(3-hexylthiophene):[6,6]-phenyl-C61 butyric acid methyl ester (P3HT:$PC_{61}BM$)/MoO_3/Al. This efficiency is lower than, but comparable to the PCE (3.36%) of a control device with similar structure PET/Ag-grid/PH1000/ZnO/PFN/P3HT:$PC_{61}BM$/MoO_3/Al. A striking advantage using VPP-PEDOT to replace PH1000 is the high ambient stability of the device. The PCE of un-encapsulated devices after 120 h continuous exposure to ambient oxygen and moisture is retained at a 75% level of its initial value. These results suggest that the Ag grid/VPP-PEDOT is a promising alternative to ITO or high conducting PEDOT:PSS for realization of high efficiency, low cost and stable organic solar cells (OSCs).

1. Introduction

Effective utilization of solar power has become an intensively studied field due to the environmental effect and security concerns of fossil fuels [1–3]. Organic solar cells (OSCs) is a highly promising technology because of its light weight, easy process and low cost. The power conversion efficiency (PCE) of OSCs has lately exceeded 10% [4,5]. However, there are still two critical steps toward the commercialization of OSCs, one is to achieve large area OSCs on flexible substrate [6] and the other is to extend the working lifetime of OSCs to a rather acceptable time range [7].

Indium tin oxide (ITO), the most commonly used transparent electrode in OSCs, however, has several inherent drawbacks. The high cost of rare element indium [8], relatively high sheet resistance and poor mechanical property [9–11] hinder the effort of industrializing OSCs. The brittle ITO is apt to crack under tension, which makes it incompatible with low cost technologies like roll-to-roll printing and coating [12]. Various materials such as carbon nanotubes [13], graphene [14–16], metallic nanowires [17–20] and conductive polymer [21–23] thus have been proposed to replace ITO, but few of these alternatives alone can possess high conductivity and transmittance simultaneously, which are two essential factors for a material considered as electrode of OSCs.

Recently, we have presented a hybrid electrode consisted of high conducting poly (3,4-Ethylenedioxythiophene):poly(styrenesulfonate) (HC-PEDOT:PSS) and current collecting silver nanogrid [24,25]. The high transmittance and low sheet resistance of the hybrid electrode result in superior performance of OSC

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devices as large as 1.21 cm^2. However, the hygroscopic nature of PSS in PEDOT:PSS film allows absorption of moisture from the atmosphere and results in an increased resistivity [26–28]. The PEDOT:PSS is also known to be easily photo-oxidized [29,30]. Moreover, the acidic PSS used in PEDOT:PSS may cause gradual erosion of the adjacent components such as the metal grid and the active material [31,32]. Therefore, the using of high conducting PEDOT:PSS as a component of the transparent electrode is detrimental to the stability of OSCs.

In comparison with solution processed PEDOT:PSS, the vapor-phase polymerization (VPP) PEDOT thin film usually shows enhanced conductivity and a smooth morphology [33,34]. Once incorporated with current collecting silver nanogrid, an electrode with low square resistance could be expected. The most appealing advantage of VPP-PEDOT is that it does not contain PSS, and thus shall be beneficial to stable OSCs with long lifetime.

Here we used VPP-PEDOT to replace HC-PEDOT:PSS in the hybrid cathode of large area, flexible OSCs with inverted structure of PET/Ag-grid/VPP-PEDOT/ZnO/poly [(9,9-bis(3-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)](PFN)/poly(3-hexylthiophene):[6,6]-phenyl-C61 butyric acid methyl ester (P3HT:$PC_{61}BM$)/MoO_3/Al. The resulting solar cells present a satisfactory power conversion efficiency (PCE) of 2.63%. Very importantly, the completely unencapsulated device retains 75% of its initial PCE after 120 h continuous exposure to ambient oxygen and moisture environment. We believe these results are highly intriguing for practical application of OSCs.

2. Experimental

The Ag-grid embedded PET (Suzhou NanoGrid Technology Co., Ltd.) was used as the substrate for VPP of the hybrid electrode. The structure of Ag-grid embedded in PET can be seen in our recent publication [24]. The hybrid electrodes were prepared by a VPP technique through procedures illustrated in Fig. 1(a). A 20 wt% solution of Fe(III) tosylate (technical grade, Aldrich) in n-butanol was used as the oxidizing agent. 0.5 mol of pyridine per mole of oxidant was used for base-inhibited VPP. To reduce the crystal formation of Fe(III) tosylate, the solution was spin-coated at 4500 rpm on Ag-grid embedded PET in a N_2 filled glovebox, and then baked at 55 °C overnight under vacuum. The monomer 3,4-ethylene dioxythiophene (EDOT) was obtained from Aldrich with a purity of 97%, and other chemicals were of analytical grade. The VPP process was conducted in the room humidity of ~40% with a N_2 flow at 150 mL/min. A jacketed reaction flask was used as the VPP chamber and its temperature was 55 °C, controlled by a super constant temperature bath (± 0.01 °C). The same VPP conditions were used to prepare a VPP-PEDOT on PET sample (PET/VPP-PEDOT) to evaluate the conductivity and optical transparency of VPP-PEDOT films.

Fig. 1(b) illustrates the structure of OSCs prepared in this study. ZnO nanoparticles and subsequent poly [(9,9-bis(3-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN) layers were employed as the electron extraction layer. P3HT as donor material purchased from Luminescence Technology Corporation, $PC_{61}BM$ as acceptor material purchased from American Dye Source, Inc. were dissolved in 1,2-dichlorobenzene with a mixing ratio of 1:0.8 by weight and stirred for 12 h at 45 °C. Then, the active layers were obtained by spin coating of the blend solution at 600 rpm for 120 s followed by baking at 110 °C for 10 m. Then, MoO_3 and metal Al were thermally evaporated in a vacuum chamber through a shadow mask. The thickness of MoO_3, employed as the hole extraction, was about 10 nm; The thickness of Al, worked as top electrode, was about 100 nm. The effective cell area defined by the geometrical overlap between the bottom cathode electrode and top anode electrode was 1.21 cm^2.

Current density–voltage (*J*–*V*) characteristics of the solar cells under illumination of 100 mA/cm^2 white light from a Hg–Xe lamp filtered by a Newport 81094 Air Mass Filter were obtained using a Keithley 2635 A source meter. All the measurements were performed under ambient atmosphere at room temperature. Then without any encapsulation, these devices were stored in air and tested at intervals in 120 h to determine the device stability and the electrical performance, according to ISOS-L-1 conditions [35].

3. Results and discussion

The current-collecting silver nanogrid used in this study has been reported in our previous publication [24]. This investigation focuses on the application of VPP-PEDOT in the hybrid electrode. The VPP-PEDOT is used as an electrical connection between the cathode interlayer (ZnO/PFN) and Ag-grid, therefore we intend to make the VPP-PEDOT film as thin as possible for high optical transmittance and low electrical resistance. A 50 nm VPP-PEDOT film is the thinnest film we can obtain through VPP method. The transmittance spectra of Ag-grid, 50 nm VPP-PEDOT/Ag-grid and 150 nm PH1000/Ag-grid are presented in Fig. 2(a). A layer of 50 nm VPP-PEDOT blocks nearly 15% of the incident light, which is

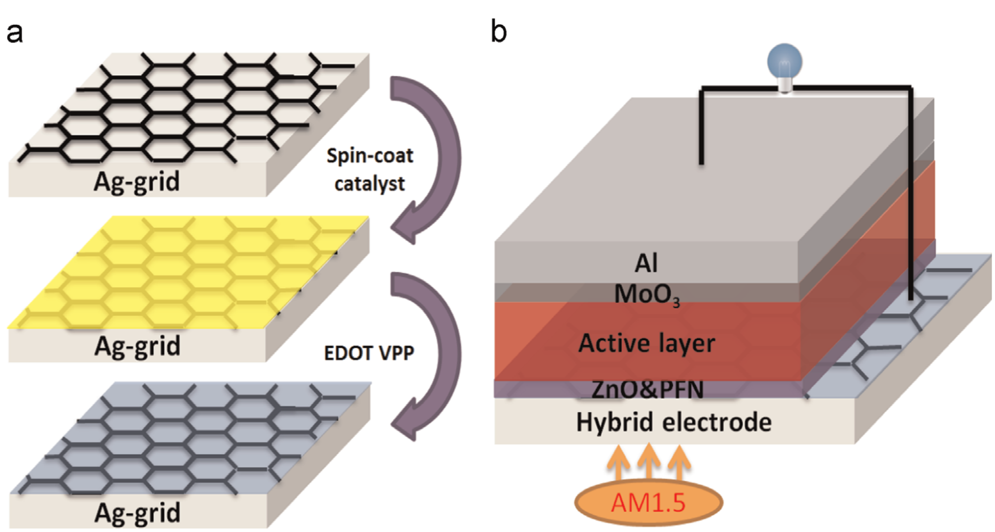

Fig.1. Schematic illustration of (a) procedures of preparing hybrid electrode; and (b) device configuration of hybrid electrode based flexible OSCs.