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Efficient inverted polymer solar cells integrated with a compound electron extraction layer



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ABSTRACT

We constructed an effective electron extraction layer (EEL) used for polymer solar cells by integrating one new kind of organic material of 4,4'-(1,4-phenylene) bis(2-phenyl-6-p-tolylnicotinonitrile) (p-PPtNT) and cesium carbonate (Cs_2CO_3) used as a compound EEL (CEEL). The CEEL based device exhibits an ideal PCE of 4.15%, corresponding to an enhancement 220% in contrast to that of control device without EEL, which is also comparable to that of ZnO based device. Our analyses indicated that the remarkably improved PCE for CEEL based device is mainly ascribed to the Ohmic contact and the negligible electron extraction barrier at cathode/active layer by inserting CEEL.

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1. Introduction

Bulk-heterojunction polymer solar cells (PSCs) are currently attracting a great deal of attention because of great promise as renewable, lightweight and low-cost energy sources [1-3]. Recently, the viable application for this emerging photovoltaic technology is promising with the power conversion efficiency (PCE) exceeding 10% as reported in the scientific literature [4]. Regardless of the competitive PCE, limitations of short-term stability and practical applications are still the major challenges for PSC being a commercially available technology compared with traditional inorganic based solar cells [5]. To achieve these goals, significant progress in PCE and short-term stability of PSC has been made in recent years by employing new materials [6,7], new device fabrication methods [8], and special interface engineering [9]. For example, Mark G. Kuzvk's group has done some work on self-healing organic dye-doped polymers to improve the stability of PSC by considering that the short-term stability is related to photodegradation of organic materials [10]. Some researchers also try to increase the carrier extraction by employing interface

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http://dx.doi.org/10.1016/j.optcom.2015.08.049 0030-4018/© 2015 Elsevier B.V. All rights reserved. engineering, which includes interface modification and P, N doping [11–13]. The most commonly used interface modification material for efficient electron extraction is some inorganic compound films, such as titanium oxide (TiO_x) [14], zinc oxide (ZnO) [15], aluminum oxide (Al_2O_3) [16] or cesium carbonate (Cs_2CO_3) [17], due to their superiority in large band gaps and high electron mobility. However, a relatively complicated preparation procedures and the high-temperature annealing process are usually needed for these inorganic compound films [18], which limit their application field especially in the flexible devices [19].

Recently, some doping technologies (P or N doped organic material) have been explored by combining organic small molecules and alkali metal compounds, such as Cs_2CO_3 doped Bphen or Alq₃ used as an efficient electron extraction layer (EEL) in PSCs [20,21]. Nevertheless, these doping methods are not adopted intensively due to their complexity and poor repeatability during the fabrication procedures. One new kind of organic material, 4,4'-(1,4-phenylene) bis(2-phenyl-6-p-tolylnicotinonitrile) (p-PPtNT) [22], has been demonstrated as an efficient electron injection layer in OLEDs [23] because of its merits in good thermal stability, a suitable lowest unoccupied molecular orbital (LUMO) level of 3.57 eV, relatively high electron mobilities (in the order of 10–5 cm² V⁻¹ s⁻¹), large ionization potentials (IP > 6.31 eV) and a low thermally evaporated temperature. In this work, we designed

a compound electron extraction layer (CEEL) by employing p-PPtNT as an organic electron collection layer and Cs₂CO₃ as the interface layer between active layer and ITO cathode. We fabricated the CEEL of Cs₂CO₃/p-PPtNT through sequential thermal evaporation technology, which is easier and more conveniently controllable at low processing temperature compared to those doping methods. Besides, small molecules exhibit good stability even though in an air environment, while those oxides especially the solution processed materials is very sensitive to humidity. A remarkably improved photoelectric properties was achieved in the CEEL based device with a high PCE of 4.15%, while it is only 1.28% for the control device without any EEL, which is also higher than that of device with Cs₂CO₃ doped Bphen as EEL with PCE of 4.12% [21]. Our results shed light on that the CEEL of Cs₂CO₃/p-PPtNT can be used as an efficient EEL in PSCs via a simple evaporation processing.

2. Experiment

The inverted PSCs with a structure of ITO/EEL/active layer/MoO₃/Al were fabricated (shown in Fig. 1(b)). Prior to the film deposition, the ITO glass substrates were cleaned by detergent, acetone, ethanol and DI water, for 10 min, respectively, and dried in an oven. Then the CEEL consisting of 0.6 nm Cs₂CO₃ and a thin layer of p-PPtNT with different thickness range from 10 to 25 nm was deposited at a base pressure of 2×10^{-6} Torr. For comparison, we also fabricated device with ZnO as the EEL, and the ZnO precursor solution was prepared by dissolving 0.5 M zinc acetate and 0.5 M monoethanolamine in 2-methoxyethanol with vigorous stirring for 12 h for the hydrolysis reaction at ambient conditions. The sol-gel-derived ZnO laver was prepared by spincoating the ZnO precursor solution onto an UV-ozone treated ITO substrate at a rate of 4000 rpm for 40 s, and then thermally annealing at 150 °C for 5 min in ambient air, during which the precursor was converted to a dense ZnO film by hydrolysis. And then, Poly(3-hexylthiophene): [6,6]-phenyl C61 butyric acid methyl ester (P3HT:PCBM) blend was used as photoactive layer, a 1:0.8 weight ratio of 10 mg/mL P3HT and PCBM was spin coated and annealed at 110 °C for 10 min inside the glove box. Finally, 5 nmthick MoO₃ was deposited as the hole collection layer (HCL), and 100 nm-thick Al film was used as anode. The photoactive area of the devices was about 0.1 cm² as determined by the overlap of ITO cathode and Al anode.

Surface morphology and roughness were characterized with atomic force microscopy (AFM) (Veeco MultiMode V) in tapping mode. The UV-vis absorption and transmission spectra were recorded at room temperature with a Perkin Elmer Lambda 750 UV/ Vis/NIR spectrophotometer. Elemental composition and the electronic structures of the film were determined by ultraviolet photoelectron spectroscopies (UPS) using a Kratos AXIS Ultra-DLD ultrahigh vacuum (UHV) surface analysis system with a monochromatic aluminum Ka source (1486.6 eV) and UPS analysis were carried out with an unfiltered HeI (21.2 eV) gas discharge lamp and a hemispherical analyzer. Details and the experimental setup for UPS measurements have been given elsewhere [21]. All spectra were measured at room temperature. Fermi level (E_F) was referred as the zero binding energy (BE) in UPS spectra. Photovoltaic measurements were conducted at room temperature in air with device encapsulation under the illumination of a 150 W Newport 91160 solar simulator using an air mass (AM) 1.5 G filter. The simulated light intensity was adjusted to be 100 mW/cm², which was calibrated with a calibrated silicon solar cell. The current density-voltage (J-V) characteristics of the inverted PSC devices were examined with a programmable Keithley 2612 source meter. The incident photons to current conversion efficiency (IPCE)



Fig. 1. (a) *J–V* characteristics of inverted P3HT:PCBM PSCs integrated with $Cs_2CO_3(0.6 \text{ nm})/p$ -PPtNT (20 nm) (CEEL), ZnO as EEL and bare-ITO based device under 100 mW/cm² AM 1.5G simulated solar illumination and (b) IPCE spectra of these inverted PSCs.

Table 1

Device performance parameters of inverted PSCs with the structure of ITO/EEL/P3HT:PCBM/MoO₃/Al, in which bare-ITO, ZnO and $Cs_2CO_3(0.6 \text{ nm})/p$ -PPtNT with various thickness were used as EEL.

| EEL | J_{SC} (mA/cm ²) | $V_{OC}\left(\mathbf{V}\right)$ | FF (%) | PCE (%) |
|-----------------|--------------------------------|---------------------------------|--------|---------|
| Bare ITO | 8.49 | 0.33 | 0.46 | 1.28 |
| ZnO | 10.13 | 0.63 | 0.63 | 4.02 |
| p-PPtNT (10 nm) | 9.01 | 0.53 | 0.52 | 2.47 |
| p-PPtNT (15 nm) | 9.40 | 0.61 | 0.59 | 3.37 |
| p-PPtNT (20 nm) | 10.07 | 0.64 | 0.64 | 4.15 |
| p-PPtNT (25 nm) | 9.53 | 0.63 | 0.62 | 3.69 |

spectra were measured with a photo-modulation spectroscopic setup (Newport monochromator).

3. Results and discussion

To investigate the interfacial function of CEEL, the current density–voltage (J-V) characteristics of the devices with p-PPtNT

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