

Poly(3-hexylthiophene):indene-C₆₀ bisadduct morphology improvement by the use of polyvinylcarbazole as additive

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ABSTRACT

Bis-indene fullerene (ICBA) is a highly symmetric molecule that tends to diffuse and aggregate into larger clusters, resulting in a huge horizontal phase separation when the master solvent vaporizes, in addition to a significant drop in power conversion efficiency (PCE). We found that Poly(3-hexylthiophene) (P3HT): indene-C₆₀ bisadduct (ICBA) blended with a high-molecular-weight polyvinylcarbazole (PVK) film could effectively prevent the clustering of ICBA crystalline molecules, thereby improving the phase separation. Furthermore, PVK dispersed the ICBA:P3HT active layer in a more uniform manner, increasing open circuit voltage (V_{oc}) and reducing photocurrent loss. As a result, the PCE of the device can be improved from 2.2% to 4.5%.

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1. Introduction

Polymer solar cells (PSCs) have the potential for roll-to-roll (R2R) process and are light-weight, low-cost, and rich in tunable synthetic materials [1–3]. In the development of PSCs, inverted structures have become increasingly important. It can avoid the unstable interface between indium tin oxide (ITO) and Poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS), as well as the oxidation of air-sensitive metal Al, thus improving the long-term stability of PSCs. To fix the above-mentioned problems, an air-stable metal that has a higher work-function compared to Al is used as the top anode; moreover, metal oxide (MO) interlayers are placed between the active layer and the electrodes in order to increase charge extraction and energy-level matching. Recently, the air-stable structure with high work-function metal is combined with low-cost solution-processed p-type and n-type metal oxides, which have become the strategic method for fabricating blocking layers on PSCs. The metal oxides include P-NiO, MoO_x, V₂O₅, ZnO, and TiO_x [4–7], etc.

On the other hand, the synthesis of the active materials is the most important part of PSCs in the process of improving power conversion efficiency (PCE) of [6], 6-phenyl-C₆₁-butyric acid methyl ester (PCBM), which is the acceptor material of the most

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developed bulk-heterojunction (BHJ) of poly(3-hexylthiophene) (P3HT):PCBM blending system. PCBM has several advantages, such as good solubility (in chlorobenzene or dichlorobenzene), high electron transportability, and compatibility in a P3HT-based system [8–10]. In the past few years, PCE of P3HT:PCBM-based highly stable inverted PSCs have been mostly in the range of 3–4%, but V_{oc} of 0.5–0.6 V is obviously inferior to that of low bandgap polymer-based PSCs such as poly((9-(1-octylnonyl)-9H-carbazole-2,7-diyl)-2,5-thiophenediyl-2,1,3-benzothiadiazole-4,7-diyl-2,5-thiophenediyl) (PCDTBT) or poly(4,8-bis-alkyloxybenzo(1,2-b:4,5-b)dithiophene-2,6-diyl-alt-(alkyl thieno(3,4-b)thiophene-2-carboxylate)-2,6-diyl) (PBDDTT-C) (V_{oc} of 0.7–0.8 V) [11–12]. The relatively low V_{oc} results from a small energy difference between the lowest unoccupied molecular orbital (LUMO) of PCBM and the highest occupied molecular orbital (HOMO) of P3HT. To circumvent the low V_{oc} problems, Yongfang Li et al. have synthesized a novel C₆₀ derivative for P3HT-based donors; indene-C₆₀ bisadduct (IC₆₀BA) is reported to reveal a higher LUMO energy level of –3.74 eV, which is 0.17 eV higher than that of PCBM. The inverted PSCs, which are based on P3HT incorporating ICBA as the acceptor showed a high V_{oc} of 0.84 V and a higher PCE of over 6% [13–17].

Unfortunately, although the premium property of ICBA greatly improves the performance of P3HT-based PSCs, the highly symmetric molecule ICBA could result in serious horizontal phase separation of the incompatible organic–inorganic ICBA/ZnO interface during the fabricating process of inverted PSCs devices without another modified interlayer. This phase separation problem apparently reduced

the V_{oc} and J_{sc} of devices with a low PCE of 2–3%. In order to solve the phase separation problem, PVK, a conjugate polymer with high carrier mobility, was blended with P3HT:ICBA as an additive to disperse the ICBA clustering in this work. We found that blending a PVK concentration of 13% by the weight ratio in P3HT:ICBA (P3HT:ICBA:PVK=1:1:0.3 weight ratio) could form an acceptor/donor (A/D) morphology fairly uniform throughout, as shown in the AFM phase images. Furthermore, the J_{sc} and V_{oc} of the devices can be optimized simultaneously under the same PVK concentration. In addition, we found that the active layer of optimized A/D morphology combined with solution processed CuO_x [4] as HTL between the active layer and Ag anode, forming an ohmic contact between the interfaces of the active layer and the Ag anode, which could effectively reduce the current leakage effectively.

2. Experiment

The fabrication procedure of the device is based largely on a solution process; its architecture is shown in Fig. 1a. The cathode buffer layer chosen here is a sol–gel derived zinc oxide (ZnO) thin film that shows signs of reasonable electron mobility and selectivity. In the n-type ZnO thin film, the doping of an amount of oxygen vacancy provides high mobility of the majority carrier (electron). In addition, 4.4 eV of ZnO conduction band edge is close to the LUMO level of fullerene electron-acceptors. On the top side of the active layer, a sol–gel derived copper oxide (CuO_x) is used, serving as the electron blocking layer. Fig. 1b shows the energy level diagram of the carrier transport layer and the active materials blended with PVK [13,23], as well as the electrodes.

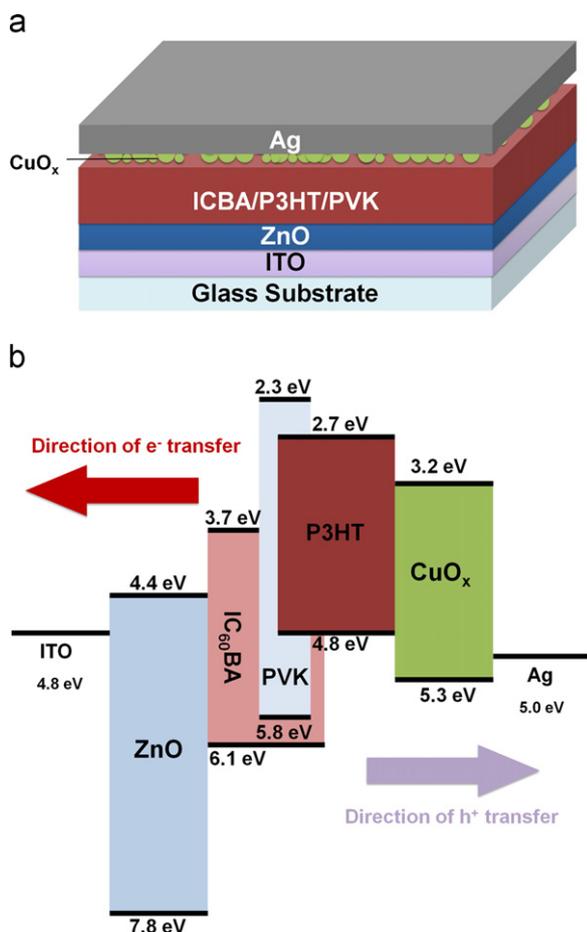


Fig. 1. (a) Device configuration of the inverted IC60BA:P3HT:PVK solar cell. (b) The energy level diagram of the device.

ITO-coated glass (250 nm, $7 \Omega/\square$) as a transparent electrode was cleaned by de-ionized water, acetone, and isopropanol. The ZnO sol–gel from a 0.5 M solution of zinc acetate in 2-methoxyethanol was then deposited on the ITO glass by a spin-coating method at 4000 rpm. Afterward, it was dried in the oven at 200 °C to create a thin film. The P3HT was purchased from Rieke Metals, and the IC₆₀BA was purchased from Lumtec Corp. The active solution contains a mixture of P3HT:IC₆₀BA (1:1 20 mg/ml weight ratio) in 1,2-dichlorobenzene (o-DCB) with different weight ratios (0%, 5%, 9%, 13% and 20% wt% of the combined weights of P3HT and ICBA). Here, the 5% weight ratio means the ratio of the PVK weight to the total weight of the P3HT and the ICBA. PVK (Aldrich-sigma, Mw=1,100,000) was then spin-cast onto the ZnO seed layer, which was spun at 600 rpm. After spinning the active layer, the CuO_x layer was spin-coated at 2000 rpm from a 0.25 M solution of copper (II) acetate monohydrate in isopropanol mixed with monoethanolamine and de-ionized water. The active layer and the CuO_x layer were annealed/dried at 150 °C for 10 min on a hotplate inside a nitrogen-filled glove box. The controlled HTL PEDOT:PSS (Clevios™ P, purchase from H.C. Starck GmbH) was then spin-cast at 2000 rpm on top of the active layer using X-triton 100 (1 wt%) as the processing additive for improving the affinity of the active/PEDOT:PSS interlayer and then pre-annealed at 140 °C in the glove box. Finally, 250 nm thick Ag cathodes were deposited over the top through thermal evaporation under a vacuum (3×10^{-4} Pa), and the active surface area was 0.06 cm².

The unencapsulated solar cells were stored and tested under ambient conditions using a Keithley 2400 SMU and an Oriel xenon lamp (450 W) with an AM1.5 filter. The ICBA:P3HT surface morphologies of the photoactive layers were measured by atomic force microscopy (AFM).

3. Results and discussion

To facilitate recognizing the morphology and the distribution of the ICBA:P3HT domain, atomic force microscopy (AFM) was used to scan the active surface layer for catching phase-difference signals to draw the phase diagrams [24,25]. The phase distribution in Fig. 2a clearly shows major horizontal phase separations on the ICBA:P3HT surface. From the statistical counts in the result of Fig. 2b, the two peaks also quantitatively demonstrate the serious horizontal phase separations. The phase separations are caused by the incompatibility of the ICBA/ZnO non-polarized/polarized interface, which will result in pin-holes and large-scale damages, further rendering non-homogeneous the blending of the acceptor/donor (A/D) morphology.

The physical phenomena of horizontal phase separations result from the highly symmetric ICBA molecules, which tend to cluster together with expanding delocalized electron orbitals, reducing the overall system energy of the active layer [18–22]. To solve the incompatible contact problem of the organic ICBA/inorganic ZnO, and the aggregation of the highly symmetric molecule ICBA in the active layer, Chain-Shu Hsu et al. reported that a cross-linkable fullerene material, (6,6)-phenyl-C61-butyric styryl dendron ester (PCBSD) [23,26], in the C-PCBSD interlayer modifies the inorganic ZnO interface with close contact for enhancing electrical coupling, then undergoes spontaneous vertical phase separation of ICBA on bottom contact. Furthermore, in the previous literatures regarding poly(2-methoxy-5-(3',7'-dimethyloctyloxy)-1,4-phenylenevinylene) (MDMO-PPV), poly(3-octylthiophene-2,5-diyl) (P3OT):C₆₀ blending system [27–28], C.J. Brabec et al. demonstrate the phenomenon of serious horizontal phase separation with the highly symmetric molecular C₆₀ and showed that blended MDMO-PPV, P3OT:C₆₀ with small amounts of conventional polymer hosts, such as polystyrene (PS), polyvinylcarbazole (PVK) or polyvinylbenzylchloride (PVBC)

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