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Optimization of inverted tandem organic solar cells

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ABSTRACT

Inverted tandem organic solar cells, consisting of two bulk heterojunction sub-cells with identical poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C_{61} (PCBM) active layer and a MoO_3/Ag/Al/Ca intermediate layer, have been presented and optimized. Indium tin oxide (ITO) modified by Ca acts as a cathode for electron collection and Ag is used as the anode for hole collection for the tandem device. A proper thickness of Ca (3 nm) forms a continuous layer, working as a cathode for the top sub-cell. MoO_3 as the anode buffer layer prevents exciton quenching and charge loss at the anode side, which could result in increase in interfacial resistance. The variance of sub-cell thickness adjusts the optical field distribution in the entire device, facilitating light absorption and good current matching in both sub-cells. The optimal inverted tandem device achieves a maximum power conversion efficiency of 2.89% with a short-circuit current density of 4.19 mA/cm^2, an open-circuit voltage of 1.17 V, and a fill factor of 59.0% under simulated 100 mW/cm^2 (AM 1.5G) solar irradiation.

1. Introduction

Investigation on organic solar cells (OSCs) has intensified in the past few years due to greatly increased demands for green energy and their unique properties compared to inorganic solar cells [1–7]. A few intrinsic properties of organic semiconductors, such as their narrow absorption range, short exciton diffusion length, and low charge carrier mobility, limit the device efficiency [8,9]. The bulk heterojunction (BHJ) structure employed in OSCs exhibits remarkable superiority over single layer or bilayer structures [10–12], primarily due to the decreased exciton diffusion distance to donor/acceptor interfaces and increased interfacial areas, facilitating exciton dissociation and yielding photocurrent enhancement. The charge transport in such a structure depends on the nanoscale bi-continuous percolated pathways, and subsequently the charge carriers can be extracted by their corresponding electrodes [3,13,14]. It has been found that the poly(3-hexylthiophene) (P3HT):1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C_{61} (PCBM) based BHJ exhibits vertical phase separation [12,15], i.e. PCBM-rich at the bottom (ITO anode side) and P3HT-rich atop (cathode side), which obstructs efficient charge transport in conventional devices. The inverted structure with the direction of charge transport/collection reversed (ITO is modified as cathode and a high work function metal is used as anode) is beneficial for charge transport and collection in these circumstances. Moreover, this inverted structure can also overcome the interface instability between poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) and low work function metals in conventional devices [16–22].

On the other hand, in order to improve absorption efficiency, organic materials with lower band gap and broad absorption range have been synthesized [9–11]. In addition, increasing active layer thickness can lead to more solar light absorption. However, the thickness has to be limited to within ~100 nm due to the relatively small charge carrier mobility, which leads to a high recombination rate [23–25]. Therefore, a tandem structure consisting of two or more cells with complementary absorption spectra is believed to have the potential to further boost the efficiency by means of maximization of light absorption [3,26–33]. In conventional tandem devices, intermediate layers are of great significance to effectively connect the sub-cells [30,34], which are usually comprised of metal nanoclusters [29,35], metal/PEDOT:PSS combination [26,36], metal oxides/PEDOT:PSS combination [28,37,38], and metal/metal oxide combination [27,30,31]. However, these

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intermediate layers cannot be applied in inverted tandem OSCs, where ITO acts as the cathode at the bottom and the high work function metal atop as the anode.

Recently, we compared the intermediate layers with various combinations of metals and metal oxides used in inverted tandem OSCs [39]. As a result, the combination of a MoO_3 layer and Ag/Al/Ca metal layers has been found to function well as an intermediate layer to efficiently connect two BHJ polymer sub-cells. In this work, we present a detailed study on optimizing the intermediate layer and the two BHJ sub-cell thicknesses through experiments and simulations.

2. Experiment

All cells in this study were fabricated on indium tin oxide (ITO) coated glass substrates with a sheet resistance of 20 Ω/sq. A Ca layer was first thermally evaporated in a base vacuum of 9.0×10^{-5} Pa.

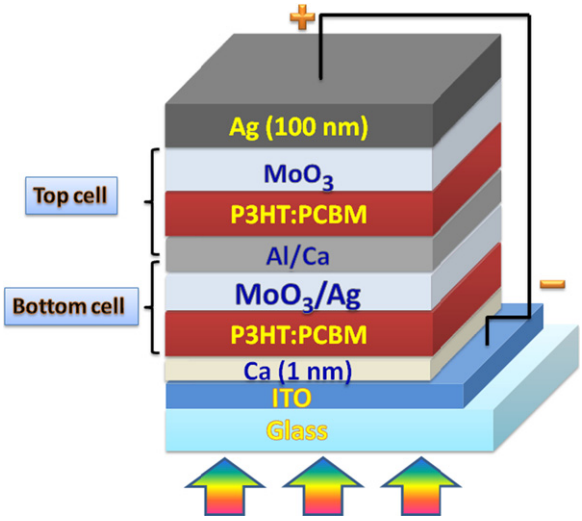

Fig. 1. Device structure of inverted tandem OSCs.

Then, a blend solution made of P3HT (Rieke Metals, Inc.) and PCBM (American Dye Sources Inc.) with a weight ratio of 1:0.8 in chlorobenzene (30 mg/ml) was spin-coated to form the active layers for both bottom and top sub-cells in a glove box filled with N_2. MoO_3, Ag, Al, and Ca were evaporated in sequence to assigned thicknesses to form the intermediate layer (9.0×10^{-5} Pa). Evaporated MoO_3/Ag with a variable thickness of MoO_3 was used as the anode for all tandem cells in this paper. Post-annealing at 160 °C for 10 min was performed on all devices after the final MoO_3/Ag anode fabrication. All devices had an active area of 0.1 cm^2 and were encapsulated before being taken out from the glove box.

The current–voltage (I–V) characteristics were measured with a Keithley 2400 sourcemeter under simulated 100 mW/cm^2 (AM 1.5G) irradiation from a solar simulator (Solar Light Company Inc.). The film thickness was measured with a surface profiler (Tencor P15). A tapping mode atomic force microscope (tapping mode AFM) (D5000, Veeco) was used to characterize the surface roughness of metal oxide layer, metal layers, and spin-coated polymer on Ca coated ITO glass. Fig. 1 shows the structure of the inverted tandem device in this study, consisting of ITO/Ca/P3HT:PCBM/MoO_3/Ag/Al/Ca/P3HT:PCBM/MoO_3/Ag. Devices with variable thicknesses of Ca in the intermediate layer, MoO_3 buffer layer, and P3HT:PCBM active layers in sub-cells were fabricated for comparison.

3. Results and discussion

3.1. Morphology of intermediate layer

Fig. 2 shows the surface morphology (tapping mode AFM images) of P3HT:PCBM (80 nm), P3HT:PCBM (80 nm)/MoO_3 (7.5 nm), P3HT:PCBM (80 nm)/MoO_3 (7.5 nm)/Ag(1 nm), P3HT:PCBM (80 nm)/MoO_3 (7.5 nm)/Ag (1 nm)/Al (1 nm), and P3HT:PCBM (80 nm)/MoO_3 (7.5 nm)/Ag (1 nm)/Al (1 nm)/Ca (3 nm) films used in the inverted tandem cells. The P3HT:PCBM (80 nm) film displays a rather smooth surface with a root-mean-square (RMS) surface roughness of 0.703 nm. Obviously, the deposited MoO_3 layer (7.5 nm) does not cause an increase in surface roughness (0.740 nm). With the deposition of metal layers (Ag, Al, and Ca) used for the intermediate layer, the RMS surface

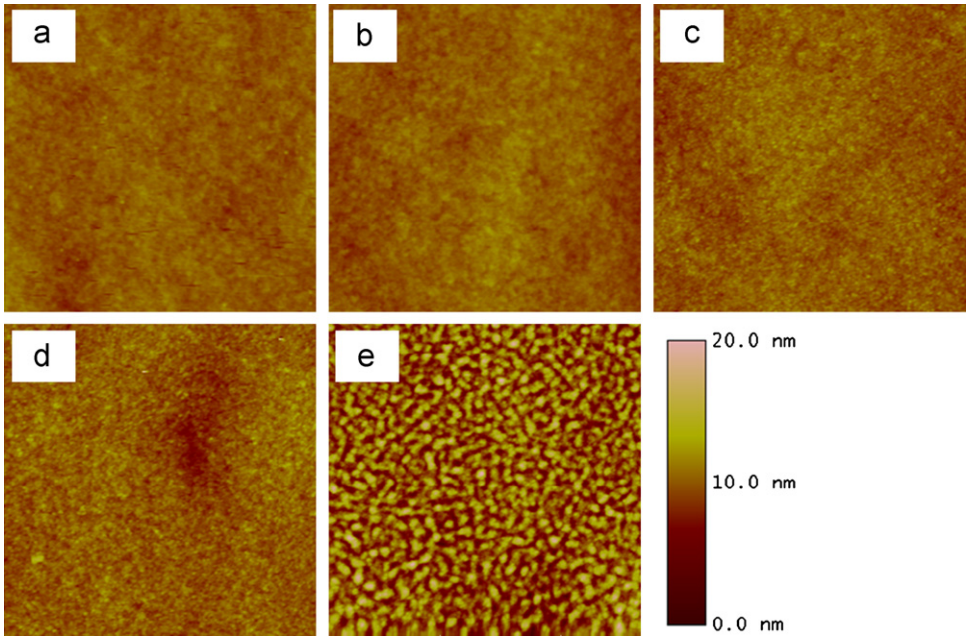

Fig. 2. Surface morphology (AFM images) of different films used for the intermediate layer: (a) P3HT:PCBM, (b) P3HT:PCBM/MoO_3, (c) P3HT:PCBM/MoO_3/Ag, (d) P3HT:PCBM/MoO_3/Ag/Al, and (e) P3HT:PCBM/MoO_3/Ag/Al/Ca.