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Improving performance of inverted organic solar cells using ZTO nanoparticles as cathode buffer layer

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ABSTRACT

In this study, a low-temperature solution-processed zinc tin oxide (ZTO) films are successfully utilized as the cathode buffer layer in the inverted organic P3HT:PCBM bulk heterojunction solar cells. ZTO film cathode buffer layer with an appropriate Sn-doping concentration outperforms the zinc oxide (ZnO) film with an improved power conversion efficiency (1.96% (ZTO film) vs. 1.56% (ZnO film)). Furthermore, ZTO nanoparticles (NPs) are also synthesized via low-temperature solution route and the device with ZTO NPs buffer layer exhibits a significant improvement in device performance to reach a PCE of 2.60%. The crystallinity of the cathode buffer layer plays an influential factor in the performance. From impedance spectroscopy analysis, a correlation between short circuit current (J_{sc}), carrier life time (τ_{avg}) and, thus, PCE is observed. The interplay between composition and crystallinity of the cathode buffer layers is discussed to find their influences on the solar cell performance.

1. Introduction

Organic bulk-heterojunction solar cells have drawn extensive attention due to its low-cost, large scale production and the compatibility to flexible electronics. However, the conventional organic solar cell devices comprising a low work function metal electrode are prone to degrade in ambient air without proper encapsulation. One of the promising solutions is inverted structure [1], in which layer sequence is reversed. In addition, for organic photovoltaic devices, problems associated with oxygen, moisture, and metal diffusion into the polymer active layers can be alleviated by inserting a buffer layer between the active layer and cathode electrode. A metal oxide buffer layer is deposited on transparent conducting oxide such as indium tin oxide (ITO) and a high work function metal like Au was utilized as a top electrode in inverted devices [1]. The commonly used metal oxide cathode buffer layers [2–4] can act as hole blocking contact and facilitating electron extraction. ZnO is one of the n-type metal oxides with conduction band edge close to the Lowest Unoccupied Molecular Orbital (LUMO) of organic electron acceptor; therefore, it has been applied as the cathode buffer layer in air-stable organic photovoltaic (OPV) cells [5]. However, the choice of cathode buffer layer remains limited.

On the other hand, stoichiometry of the oxide semiconductors is a key parameter in controlling the carrier concentration and electrical properties. It has been reported that metal ions with high ionic potential can effectively attract oxygen and suppress the generation of oxygen deficiency, which consequently changes the oxide stoichiometry [6]. Because Sn has a higher ionic potential than Zn, we have demonstrated that adding Sn into ZnO to form zinc tin oxide (ZTO) will effectively reduce the oxygen vacancy and improve field-effect mobility as a channel material for thin film transistors [7]. Therefore, ZTO would be a suitable buffer material for photovoltaic applications. ZTO was a widely investigated n-type semiconducting material in thin film transistor [8–10] and it was exploited as electron transporting buffer layer in different types of solar cells [11–13]. But, the correlation between the Sn/Zn composition in ZTO film buffer layers and the performance of OPV devices is still not well understood. To optimize the ZTO film composition as cathode buffer layers in inverted organic solar cells, ZTO buffer layers are chosen in this paper. To focus on the function of ZTO buffer layers, we adopt the widely investigated P3HT:PCBM bulk heterojunction system. P3HT is poly(3-hexylthiophene-2,5-diyl) and is served as donor; on the other hand, fullerene-derived PCBM is [6,6]-phenyl C61 butyric acid methyl ester and is served as acceptor. PEDOT:PSS is used as anode buffer layer [14]. The ZTO film cathode buffer layers were prepared via solution route and only a low temperature (200 °C) annealing was needed. To optimize the composition in the ZTO film buffer

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layers, we vary the Sn/Zn molar ratios and find that the Sn concentration in ZTO plays an important role in solar cell performance. In addition, ZTO nanoparticles (NPs) synthesized through solution process is also demonstrated to compare the performance of ZTO film and ZTO NPs cathode buffer layers. The differences among all ZTO cathode buffer layers are discussed.

2. Material and methods

The solar cell devices were fabricated on patterned ITO glass with a sheet resistance of 7 Ω/□. The substrates were sequentially cleaned in an ultrasonic bath with detergent, acetone, and isopropyl alcohol each for 15 min, and then dried in a nitrogen stream. The zinc acetate dehydrate ($Zn(CH_3COO)\cdot 2H_2O$, SHOWA) and tin(II) chloride ($SnCl_2\cdot 2H_2O$, Aldrich) were dissolved in 2-methoxy alcohol (Merck) and the solutions were stirred overnight. The concentration of ZTO solution was kept at 0.1 M and the Sn/Zn molar ratios are 0, 0.1, 1, 2, and 4. The ZTO solution was spin-coated on ITO substrate at 4000 rpm for 30 s followed by 10 min of thermal annealing at 200 °C on hot plate in air to prevent damaging the ITO electrode. On the other hand, the ZTO nanoparticles (NPs) suspension was spin-coated onto the ITO substrate with a spin rate of ~7000 rpm. ZTO NPs were prepared following the method described by Jen's group [5] but the precursors were replaced with zinc acetate dehydrate and tin chloride dehydrate, and a 10 nm thick ZTO NPs film was obtained without any further post thermal treatment. P3HT (Rieke metals, UR-P3H001) and PCBM (Nano-C, UR-PCBM001) were dissolved in 1,2-dichlorobenzene (Sigma–Aldrich, 99%) in 1:1 w/w ratio to form a 40 mg/ml solution. P3HT:PCBM solution was spin-coated on the top of the buffer layers at 500 rpm for 10 s and 1500 rpm for 1 min, and then dried at 110 °C for 20 min on a hot plate in the glove box. The PEDOT:PSS isopropyl alcohol dilution was spin-coated on P3HT:PCBM active layer with spin rate at 5000 rpm for 60 s. Finally, Au/Ag bilayer top electrodes were deposited through thermal evaporation. The thickness is 60 nm and 80 nm for Au and Ag, respectively. The working area of the devices was 0.16 cm^2 defined by a shadow mask.

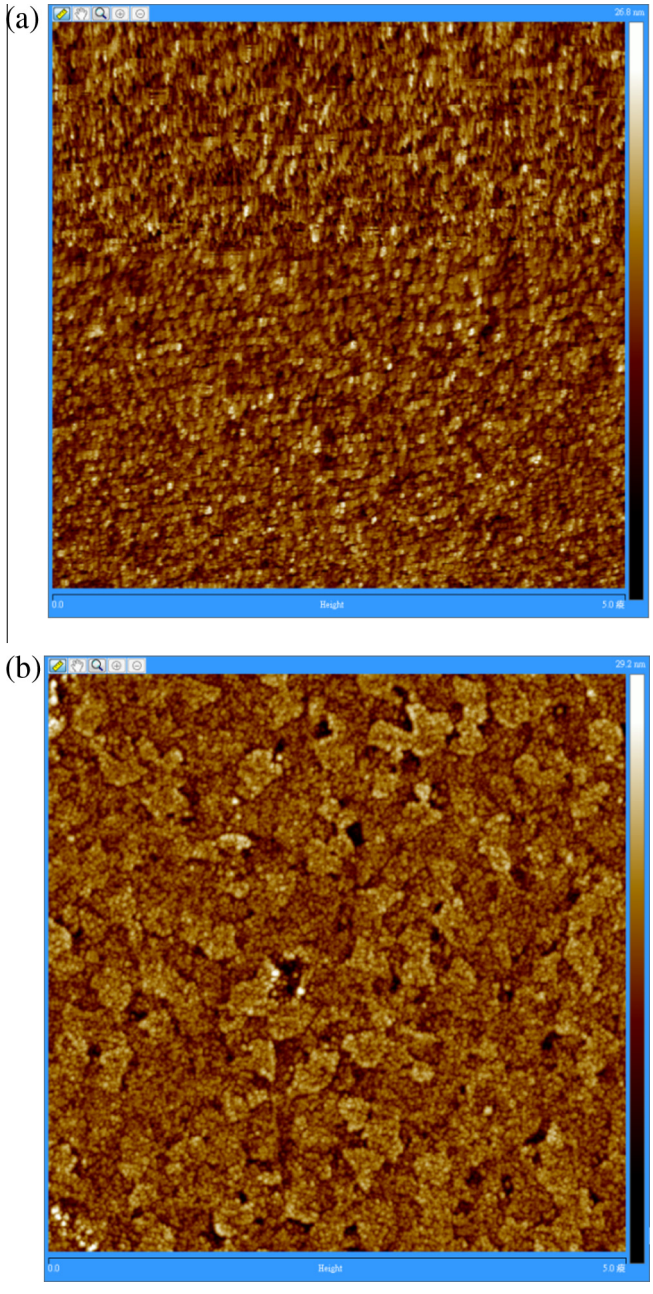

Fig. 1. AFM topography of (a) the ZTO (Sn/Zn = 1) film on ITO substrate and (b) ZTO NPs on ITO substrate (scan area: 5 μm × 5 μm).

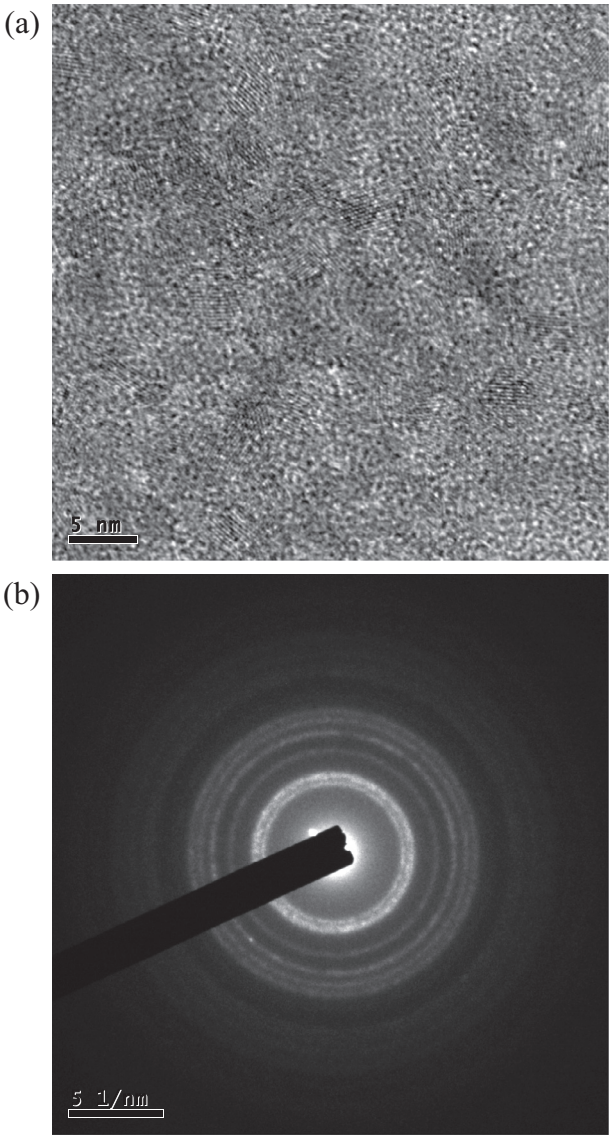

Fig. 2. (a) TEM image of ZTO NPs and (b) diffraction pattern of ZTO NPs.