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Inverted polymer solar cells with a boron-doped zinc oxide layer deposited by metal organic chemical vapor deposition



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ABSTRACT

We report on the photovoltaic properties of inverted polymer solar cells (IPSCs) where the transparent indium tin oxide (ITO) electrode was modified by a ZnO layer using metal organic chemical vapor deposition (MOCVD). The intrinsic ZnO (i-ZnO) layers were deposited with varying thicknesses from 0 to 1500 nm. The work function and surface morphology of ITO/i-ZnO were found to be dependent on the i-ZnO thickness. When the thickness of the i-ZnO layer was 80 nm, optimized IPSCs with a power conversion efficiency (PCE) of 2.93% was achieved. Furthermore, the i-ZnO layer doped with boron (BZO) was investigated. The best IPSC with a BZO layer showed a PCE of 3.26%, which is higher than that (2.93%) of the device with the i-ZnO layer. The better performance is due to combined effects of improvement in charge collection and conductivity of BZO/ITO electrode.

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1. Introduction

Organic photovoltaic (OPV) devices based on bulk-heterojunction (BHJ) concept are considered to be a promising candidate for the next generation solar cells due to their appealing properties such as mechanical flexibility, light weight and potential low cost [1–3]. Current focus of OPV research includes the development of low-bandgap polymers for better sunlight absorption, the improvement of long term stability and easy process as well as device structure suitable for cost-effective large-scale product [4–9]. In a conventional structure of an OPV device, indium tin oxide (ITO) modified with p-type poly(3,4-ethylene dioxythiophene):(polystyrene sulfonate) (PEDOT:PSS) and a low work function metal are used as anode and cathode, respectively. However, the acidic PEDOT:PSS is detrimental to the transparent conducting oxide (TCO) anode and the low work function metallic cathode is oxidized easily in air, which makes devices exhibit poor lifetime [10–12]. In addition, it has been reported that the spontaneous phase separation of donor and acceptor in OPV leads to higher concentration of acceptor at the TCO side. Such a vertical concentration distribution of donor and acceptor disfavors charge transport and collect in the direction required in conventional OPV [13].

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** Corresponding author. Tel./fax: +86 22 23508032. E-mail addresses: huziyangyong@gmail.com, huziyang@nbu.edu.cn (Z. Hu), A very effective way to circumvent this problem was proposed with the so-called inverted device. By taking advantage of the ability to reverse the polarity of charge collection in an inverted configuration, an airstable high work function metal is used as the anodic electrode for hole collection, while metal oxides such as TiO_x and ZnO act as the electron transporting layer (ETL) [14–16]. For high performance device, an important key appears to be the appropriate selection of the ETL, which is also to provide hole blocking capability and a low resistance pathway for efficient electron extraction.

Recently, there have been lots of studies characterizing inverted solar cells that use ITO/ZnO transparent electrodes. In these studies, many methods have been employed for producing the ZnO layers, including solution processed sol-gel [17-18], nanoparticles [19-20], and sputtering [21]. Furthermore, different nanostructures, such as ZnO nanorods [16,22-23], ZnO nanoridges [24], or ZnO nanoripples [25] have been used to improve the electron transport in BHJ solar cells. Unfortunately, inverted polymer solar cells (IPSCs) using modified ITO electrodes have shown severely reduced fill factors, owing to the appearance of an S-shaped kink in the current–voltage (I-V) characteristics, which is commonly associated with surface recombination and imbalanced carrier transport [26-27]. Additional resistance resulting from the ETL leads to high series resistance in the device and reduced short circuit current density (Jsc) [28–29]. Hence, further improvement of inverted architecture can be expected by reducing the resistivity of the ETL layer and optimizing the energy alignment between acceptor/ETL interfaces [30].

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ZnO can also be deposited by metal organic chemical vapor deposition (MOCVD) [31-32]. MOCVD is a kind of chemical vapor deposition technique with the precursor gas reactants. Pinholefree films can be deposited when the precursor gas reactants are introduced into the reaction chamber separately and sequentially. Therefore, film depositon by MOCVD not only precisely controls the film thickness but also offers superior film quality. Moreover, the amount of dopant is also tunable by controlling the amount of precursors [31-32], which can improve the film conductivity. Consequently, MOCVD-grown ZnO may be a promising candidate for ETL in OPV devices. In this paper, MOCVD was firstly employed to prepare a ZnO ETL used in IPSCs. The effects of laver thickness and boron doping concentration of ZnO on the device performance were also investigated. The performance of IPSC is enhanced, especially in J_{sc} , by replacing an intrinsic ZnO layer with an appropriately boron doped ZnO layer.

2. Experimental

The ZnO thin films were deposited on cleaned ITO-coated glass substrates by the MOCVD technique. The precursors were diethylzinc (DEZ) and water, introduced with high purity argon into the chamber. Diborane is used as the dopant. During the growth, deposition temperature and total pressure were kept constant to 175 °C and 1 Torr, respectively. The flow rates of DEZ and water were kept constant at 180 and 110 sccm, respectively. The intrinsic ZnO (i-ZnO) layers of various thicknesses (40, 60, 80, 150, 500 and 1500 nm) were deposited by the different reaction time. The boron-doped ZnO (BZO) layers with different doping



Fig. 1. A schematic drawing of the MOCVD setup.

concentrations were deposited by the various DEZ flow rates (1 sccm, 3 sccm and 5 sccm). The schematic drawing of the MOCVD setup was shown in Fig. 1.

To fabricate an inverted device, the active layer with a thickness of ~100 nm was fabricated by spin coating the blend solution, made of poly (3-hexylthiophene)(P3HT) (Rieke Metals, Inc.) and 6, 6-phenyl C₆₁ butyric acid methyl ester (PCBM) (Nano-C) with a weight ratio of 1:0.8 in chlorobenzene (10 mg/ml), onto the ZnO layer previously deposited. Finally, a 10 nm MoO₃ and an 80 nm Al were deposited under 4×10^{-4} Pa by thermal evaporation through a shadow mask. The structure of the inverted device is as follows: ITO/ZnO/P3HT:PCBM/MoO₃/Al as show in Fig. 2(a). Devices with conventional structure of ITO/PEDOT:PSS/P3HT:PCBM/LiF/Al were also made for comparison. A ~40 nm thick PEDOT:PSS (Baytron P VP AI 4083, H.C. Stark) buffer layer was spin coated on clean substrates at 5000 rpm for 60 s and baked at 140 °C in air. The blend solution comprising P3HT and PCBM was then spin-coated on the top of the PEDOT:PSS layer. The P3HT:PCBM solution was identical to the one used in the inverted devices. Subsequently, a LiF(1 nm)/Al(100 nm) film was deposited on the top of the active layer in vacuum(10^{-4} Pa).

The surface morphologies of ZnO layers were taken in a tapping mode with an atomic force microscope (Seiko SPA-400 SPM UNIT). The carrier concentration was obtained by Hall-effect measurements using Van der Pauw method (Accent HL5500 PC). The film thickness was measured by step profilometer (AMBIOS-XP2). The work function of ITO modified by ZnO was measured in air by a Kelvin single probe. Device current density–voltage characteristics were measured using a Keithley 2420 under 100 mW/cm² simulated AM 1.5G solar illumination_(Oriel 96000, 150 W). Device fabrication and characterization were performed in ambient condition.

3. Results and discussion

Fig. 3 shows the dependence of the work function of the modified ITO as on the thickness of the i-ZnO layer. The work function of cleaned ITO was measured to be 4.87 eV. After modified by the i-ZnO layers, the work function of ITO/ZnO was reduced and fluctuated at 4.45 eV for all ZnO thicknesses as shown in Fig. 3(a). These results indicate that a thin ZnO deposited by MOCVD is sufficient to change the work function of ITO. The work function of ITO/i-ZnO electrode is close to the lowest unoccupied molecular orbital (LUMO) energy level (3.8 eV) of PCBM as shown in Fig. 2(b). Therefore, ZnO layers of any thickness can be used to modify ITO, to serve as a transparent ETL in organic solar cells. However, the role of ITO/i-ZnO electrode is not only determined by



Fig. 2. (a) Device structure and (b) energy diagram of IPSCs with ZnO layer.

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