



Low-temperature, solution-processed aluminum-doped zinc oxide as electron transport layer for stable efficient polymer solar cells



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ABSTRACT

A simple low-temperature solution-processed zinc oxide (ZnO) and aluminum-doped ZnO (AZO) were synthesized and investigated as an electron transport layer (ETL) for inverted polymer solar cells. A solar cell with a blend of poly(4,8-bis-alkyloxy-benzo[1,2-b:4,5-b'] dithiophene-alt-alkylcarbonyl-thieno [3,4-b] thiophene) and (6,6)-phenyl-C71-butyric acid methyl ester as an active layer and AZO as ETL demonstrates a high power conversion efficiency (PCE) of 7.36% under the illumination of AM 1.5G, 100 mW/cm². Compared to the cells with ZnO ETL (PCE of 6.85%), the PCE is improved by 7.45% with the introduction of an AZO layer. The improved PCE is ascribed to the enhanced short circuit current density, which results from the electron transport property of the AZO layer. Moreover, AZO is a more stable interfacial layer than ZnO. The PCE of the solar cells with AZO as ETL retain 85% of their original value after storage for 120 days, superior to the 39% of cells with ZnO ETL. The results above indicate that a simple low-temperature solution-processed AZO film is an efficient and economical ETL for high-performance inverted polymer solar cells. Due to its environmental friendliness, good electrical properties, and simple preparation approach, AZO has the potential to be applied in high-performance, large-scale industrialization of solar cells and other electronic devices.

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1. Introduction

Polymer solar cells (PSCs) were intensively studied in the past decades due to their low-cost, mechanical flexibility, light-weight and ease for large area manufacturing on flexible substrates [1,2]. Recently, PSCs showed impressive power conversion efficiency (PCE) of over 10% due to intense research in low band-gap polymers [3]. While the optoelectronic properties of a photoactive layer determine the theoretically maximum photocurrent generation, interfacial layers are of equal importance. They have to provide efficient charge carrier extraction towards the electrodes thereby avoiding losses such as non-ohmic contact, charge carrier recombination and exciton quenching at interfaces [4,5]. Poly(styrenesulfonate)-doped poly(ethylenedioxythiophene) (PEDOT:PSS) [6] and transition metal oxides, such as nickel oxide (NiO), molybdenum oxide (MoO₃), and vanadium oxide (V₂O₅) are attractive choices for p-type interfaces as a hole transport layer (HTL) for conventional PSCs [7–11]. Nevertheless, the poor stability of conventional PSCs is one of crucial obstacles to commercialization. Further tremendous efforts are required to develop environmentally stable and mechanically robust interfaces.

Inverted polymer solar cells (IPSCs) have rapidly evolved with the progress of n-type inorganic metal oxides as electron transport layers (ETLs), such as titanium oxide (TiO_x), zinc oxide (ZnO) or aluminum doped zinc oxide (AZO) [12–18]. They have emerged because of their intrinsically higher environmental stability as compared to PEDOT:PSS or calcium (Ca) charge transport layers [19]. Intrinsic ZnO and TiO_x show low conductivities and therefore are limited in IPSCs. Recent reports showed that using doped metal oxides with enhanced conductivity is a viable path to overcome this restriction [20,21]. ZnO is a prototype material system for doping investigations. Puetz et al. used an indium-doped ZnO nanoparticle solution to process ETL [20]. Stubhan et al. demonstrated the benefits of AZO layers over ZnO, however, the formation of AZO is under a high temperature of 260 °C for the calcinations of the film [17,21]. By employing the Fullerene-SAMs, Stubhan et al. [22] show a modification of the electronic properties of AZO. Until very recently, L. K. Jagadamma et al. [23] reported a low-temperature (125 °C) solution-processed AZO buffer layer yielding power conversion efficiency in excess of 10%.

Here we introduce a simple solution-processing method for the preparation of ZnO and AZO with a relatively low annealing temperature of 150 °C as ETL for IPSCs. Poly(4,8-bis-alkyloxy-benzo[1,2-b:4,5-b'] dithiophene-alt-alkylcarbonyl-thieno [3,4-b] thiophene) (PBDTTT-C)

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and (6,6)-phenyl-C₇₁-butyric acid methyl ester (PC₇₁BM) were applied as donor and acceptor, respectively.

2. Experimental section

2.1. Materials

Zinc acetate (Zn(CH₃COO)₂·2H₂O) and aluminum nitrate (Al(NO₃)₃·9H₂O) were purchased from Aladdin. 2-Ethoxyethanol and ethanolamine were bought from Alfa Aesar and Sinopharm Chemical Reagent Co., Ltd. respectively. Indium tin oxide (ITO) glass with a sheet resistance of 15 Ω/sq was supplied by Shenzhen Display Co., Ltd. (China). PBDTTT-C and PC₇₁BM were purchased from Solarmer and American Dye Sources (ADS), respectively. 1,8-Diiodooctane (DIO) was purchased from Sigma Aldrich. All materials were used directly without any further purification.

2.2. Synthesis of ZnO/AZO precursors

The solution-processable ZnO precursor was prepared by adding 4 mmol of zinc acetate to 10 mL of 2-ethoxyethanol mixed with 0.24 mL ethanolamine. The ethanolamine was used as stabilizer. Then the mixed solution was stirred at 60 °C for 2 h in a round-bottom flask. With the same approach, the AZO (0.25 at.%) was obtained by adding 0.02 mmol of aluminum nitrate as the aluminum source.

2.3. Device fabrication

ITO glass substrates were cleaned sequentially with detergent, de-ionized water, acetone, and isopropanol, followed by drying with N₂ flow and oxygen treatment for 5 min. Then, ZnO or AZO was formed via spin coating at 4000 rpm for 40 s followed by heating at 150 °C for 24 h. The substrate was then transferred to a N₂ filled glove box. Then, the solution of PBDTTT-C:PC₇₁BM was prepared by spin coating (1300 rpm) its o-dichlorobenzene solution of 1:1.5 w/w, polymer concentration of 12.5 mg/mL with 3% volume ratio of DIO additive on

the ITO/ETL electrode. Finally, 5 nm MoO₃ and 100 nm Ag were thermally deposited on the photoactive layer under a base pressure of 5 × 10⁻⁴ Pa. The active area of the device is 0.1 cm² defined by a shadow mask.

2.4. Characterization and measurement

A scanning electron microscope (SEM, Hitachi S4800) was performed to characterize the morphology of ZnO and AZO films. X-ray diffraction (XRD) was recorded on a powder X-ray diffractometer (Rigaku D/max-rA; Japan) equipped with a rotating anode and a Cu-Kα radiation source at a step width of 0.02°. For chemical analysis, X-ray photoelectron spectroscopy (XPS, VG ESCALab220i-XL) measurements with 300 W Al Kα radiation were carried out and were calibrated with C 1s peak (284.8 eV). UV-vis spectra were recorded from 300 to 800 nm on a Hitachi U-4100. Current density–voltage (*J*–*V*) characteristics of the devices were measured in a N₂-filled glove box by an AM 1.5 solar simulator with a light intensity of 100 mW/cm² calibrated using a standard mono-crystalline silicon solar cell (Newport, Oriel Instruments, Model: 91150V) and a computer-controlled digital source meter (Keithley, Model: 2420). Current–voltage characteristics of ZnO and AZO films were used to investigate their conductivity. External quantum efficiencies (EQEs) of solar cells were analyzed by using a certified Newport incident photon conversion efficiency (IPCE) measurement system.

3. Results and discussion

Fig. 1 shows SEM images of ZnO (a) and AZO (b) samples. The films possess a uniform and dense microstructure with granular grains of about 20 nm. The SEM images indicate the continuous and compact structure of as-prepared films, which benefited from improvements on device fabrication and performance. Fig. 1(c) shows a comparison of the XRD pattern of AZO with that of a ZnO film. Obvious characteristic peaks are identified as (100), (002), (101), (102), and (110) planes of the ZnO structure. A little shift to a lower angle is found on the (002)

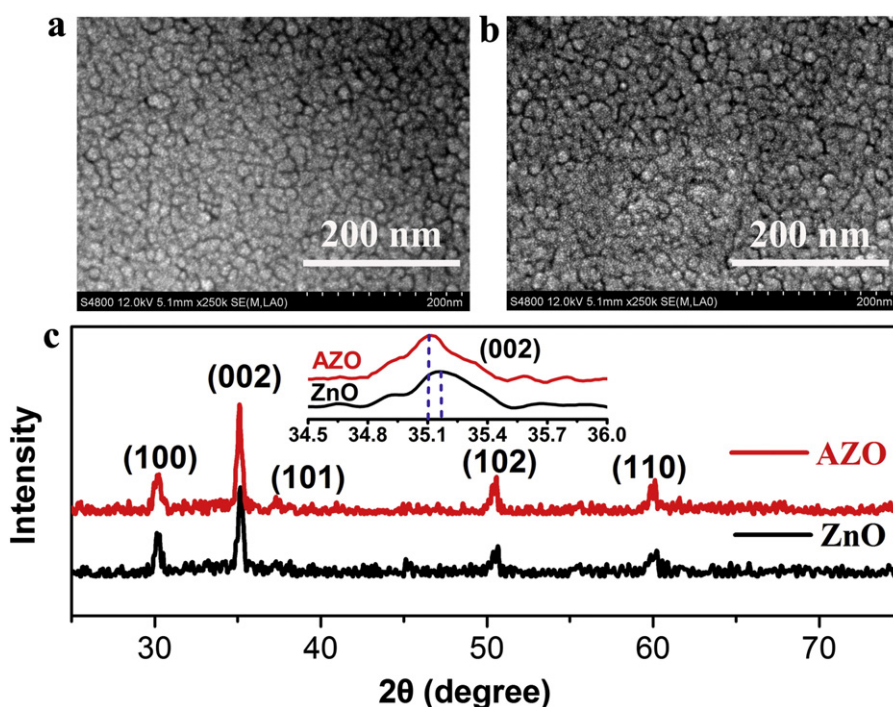


Fig. 1. SEM images of ZnO (a) and AZO (b) annealed at 150 °C. The particle sizes are approximately the same. (c) The X-ray diffraction pattern of AZO and ZnO. (Partial range at the peak position of (002) plane is inserted for comparison.)

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