

Role of zinc oxide thickness on the photovoltaic performance of laminated organic bulk-heterojunction solar cells

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ABSTRACT

A comprehensive study of zinc oxide (ZnO) film thickness and morphology on the electronic properties of inverted cells is reported. The complete conversion of zinc acetate precursor to 3–5 nm particles of ZnO with no residual acetate is obtained after 10 min at 300 °C. The work-function determined by Ultraviolet Photoelectron Spectroscopy (UPS) was 4.3 eV. and is independent of the thickness of the ZnO layer or with the planarization of the ITO surface topology. However, the efficiency varies from 0.6% to 1.7% as the ZnO thickness varies from 17 nm to 28 nm (assuming full density for the ZnO layer) in a laminated device, with both the shunt and series resistance showing a strong variation with ZnO thickness. A relatively thick, mixed phase ZnO/bulk hetero-junction in which the bulk hetero-junction penetrates into the porous ZnO layer is proposed to explain the observed performance trends.

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1. Introduction

Solution processed bulk hetero-junction organic photovoltaic (OPV) have attracted extensive research and development due to their potential to be a low cost renewable energy source, mechanically flexible, light weight and their production scalability [1,2]. In the last few years, significant effort has focused on improving the performance and lifetime of these devices resulting in efficiencies of more than 8% being reported [3], leading to realistic commercialization opportunities of the OPV technology in the near future. However, a number of significant challenges remain, especially regarding interfacial stability, such as the etching of ITO by Poly (3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) in which indium and tin migrate into the PEDOT:PSS layer resulting in reduced device efficiencies and lifetimes [4,5].

Inverted devices, as shown in Fig. 1, in which the polarity has been reversed, have the potential to overcome the inherent instability of the ITO-PEDOT:PSS interface. This structure is also highly compatible with roll-to-roll manufacturing processes [6–10] to enable large scale production through various printing and coating methods [7,11,12]. Lamination has the added potential to allow optimal treatment of the layers in the device prior to fusing as well as overcoming wetting issues in the sequential fabrication of these devices.

If ITO is to be used as the cathode, the work function must be lowered. This can be achieved by the application of low work-function materials such as zinc oxide (ZnO) [13], cesium carbonate (Cs₂CO₃) [14], or titania (TiO₂) [15]. Recent research has focused on ZnO buffer layers, due to its high transparency, electron mobility [16] and more importantly the ability to synthesize ZnO films by solution processing [16,17] making it applicable to roll-to-roll processing of inverted OPV's [18]. However, there are no systematic studies of the optimal morphology or understanding of the critical performance parameters for this system, with a wide range of performance being reported due to morphology, [19,20] and film thicknesses of ZnO [21,22]. The final process conditions used in a roll-to-roll process will be limited by the dimensional stability of the substrate (especially PET), so it is especially important to understand the mechanisms limiting optimal device performance prior to low temperature processes being developed and utilised to minimise performance degradation.

In this paper, we report a comprehensive study of the impact of film thickness and morphology of the ZnO layer on the performance of inverted OPV's produced by lamination and propose a mechanism to explain the thickness dependent properties.

2. Experimental

2.1. Device fabrication

ITO coated glass substrates with a sheet resistance of 15 Ω sq⁻¹ (purchased from Kintec) were used. The ITO glass was cleaned by

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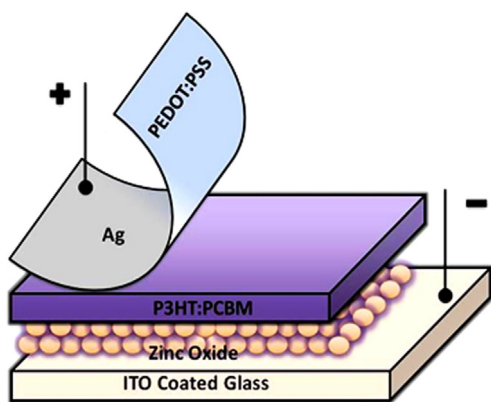


Fig. 1. Schematic of an inverted device fabricated using lamination technique.

immersing it in a well-stirred solution of 5% Pyroneg (supplied by Johnson Diversey) at 80 °C for 20 min. Samples were then rinsed in DI water and were successively sonicated in deionized (DI) water, acetone and iso-propanol for 10 min each. Subsequently the substrates were then cleaned with UV-ozone for 20 min.

Zinc oxide sol-gel was prepared as follows: zinc acetate dihydrate ($\text{Zn}(\text{C}_2\text{H}_3\text{O}_2)_2 \cdot (\text{H}_2\text{O})_2$ 99.9% chemical purity, supplied by Sigma Aldrich) (250 mg) was dissolved in absolute ethanol (5 ml) at room temperature (0.23 M). The resulting mixture was stirred at 70 °C for two hours to form a clear homogeneous solution. Upon cooling to room temperature, undissolved zinc acetate salt was seen to settle at the bottom of the vessel.

The zinc acetate layer thickness was varied by changing the temperature of the zinc acetate solution and thus the solubility of zinc acetate in ethanol. The homogenous solution was used at 70 °C at different spin speeds as well as the saturated room temperature solution.

The resulting ZnO films were prepared by spin-coating the solution (filtered using a 0.45 μm syringe filter) on cleaned ITO substrates (using a PWM32 spincoater from Headway Research, Inc.) at various spin speeds followed by annealing at 300 °C for 10 min in air, and then cooled at room temperature. The detailed recipes for various experiments are summarized in Table 1.

P3HT and PCBM were mixed in a 1:1 weight ratio (20 mg ml^{-1}) and dissolved overnight with constant stirring in the dark in chlorobenzene with 2% bromooctane to promote phase separation [23]. This solution was then spin-coated on ZnO coated ITO substrates in ambient conditions at 1000 rpm for 30 s followed by annealing at 115 °C for 10 min (first part of the device).

For the second half of the laminated device, 100 nm thick silver electrodes were sputter coated on a Kapton film and PEDOT:PSS (Clevios P VP AL 4083) doped with d-sorbitol (15% by weight, purchased from Sigma Aldrich) was doctor bladed. PEDOT:PSS coated substrates were then annealed at 115 °C for 60 min in ambient conditions on a laminar flow hood.

The two parts of the device were brought together and laminated for 10 min under contact pressure, in a hydraulic hot press preheated at 130 °C. The press was cooled to 40 °C before releasing the pressure and recovering the laminated devices.

2.2. Atomic force microscopy (AFM)

The surface morphology of the samples was studied using VEECO multimode AFM operated in tapping mode.

2.3. Electron spectroscopy

The investigations of the ZnO seed layer on ITO substrates with UPS were performed in an ultra high vacuum (UHV) apparatus

Table 1

Recipes for spin-coating various zinc oxide layers on ITO.

Sample	Spin speeds (rpm)	Solutions used
Sample A	5000	Saturated solution @ ambient
Sample B	5000	Fresh clear (0.23 M) solution @ 70 °C
Sample C	3000	Fresh clear (0.23 M) solution @ 70 °C

built by SPECS (Berlin, Germany). The apparatus is equipped with a two-stage cold cathode gas discharge from MFS (Clausthal-Zellerfeld, Germany) to generate simultaneously metastable helium atoms ($\text{He}^* \ ^3\text{S}_1$) and UV light (He I line). The spectra of the electrons emitted from the samples are recorded at a pass energy of 10 eV and 2 eV, with a hemispherical Phoibos 100 energy analyser from SPECS. At the pass energy of 10 eV the analyser has an energy resolution of 400 meV as evaluated from the Fermi edge of polycrystalline silver.

In a UPS experiment the sample is irradiated with UV photons leading to photoionization via the photoelectric effect. The energy of emitted electrons is given by

$$E_{KE} = E(h\nu) - E_{bin} - \phi_{spec} \quad (1)$$

where E_{KE} is the kinetic energy of the emitted electron, $E(h\nu)$ the photon energy (21.22 eV for the He I line used here), E_{bin} the binding energy of the electron before excitation and ϕ_{spec} the spectrometer work-function. UPS spectra can be used to determine the work-function and the density of states in near surface region of a material. The work-function of the samples is determined as the difference between the excitation energy and the length of the spectrum. The latter is given as the difference of the high binding energy cut-off and the cut-off of the spectrum at the lowest binding energy.

The apparatus is further equipped with a non-monochromatic X-ray source for Mg and Al $K\alpha$ radiation with the first one used in the present study. The angle between the He^*/UV light irradiation and the analyzer and the X-ray irradiation and the analyzer are both 54°. The base pressure of the UHV chamber is a few 10^{-10} mbar. High-resolution XPS spectra were obtained to determine the chemical state of zinc and oxygen in the surface near region of the ZnO thin films. The XPS spectra were referenced to the carbon 1 s peak, which is due to the adventitious hydrocarbons adsorbed on the ZnO surface. The carbon 1 s peak was set to 285 eV.

2.4. Rutherford backscattering spectroscopy (RBS)

RBS was performed on an HVEE 2MV Tandem accelerator with 1.8 MeV He^+ directed perpendicular to the sample surface, with a rectangular beam shape of 2 mm width and 4 mm height and a beam current of around 5 nA. The energy of backscattered He atoms were measured with a surface barrier detector, in a vacuum environment of less than 1×10^{-7} mbar, and the data was processed using a commercial package (SIMNRA) [24]. The scattering angle of the He^+ was 170°.

2.5. Thermogravimetric analysis (TGA)

TGA was performed using a TA Instruments Hi-Res modulated TGA 2950-Thermogravimetric Analyser. For TGA measurements, a fresh clear solution of zinc acetate was placed in the pan. The TG analysis was performed in air by ramping temperature from room temperature to 300 °C at a rate of 20 °C min^{-1} under an air flow of 60 mL min^{-1} , and an isothermal hold at 300 °C, as shown in Figs. 4 and 5 to mimic device processing.

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