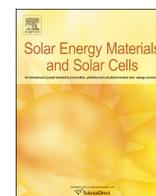




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## Solution processed n-type mixed metal oxide layer for electron extraction in inverted polymer solar cells

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### ABSTRACT

In this paper we demonstrate an efficient inverted polymer bulk heterojunction (BHJ) solar cells using solution processed interfacial layer by mixing zinc oxide (ZnO) with cesium carbonate ( $\text{Cs}_2\text{CO}_3$ ) for ideal electron extraction and transport. The optimized inverted BHJ solar cell with n-type mixed ( $\text{ZnO}:\text{Cs}_2\text{CO}_3$ ) electron transport layer and blend of poly (3-hexylthiophene):Indene-C60 bisadduct as a photoactive layer shows a power conversion efficiency as high as 5.29%. This performance enhancement is attributed to improved interfacial contact between photoactive layer and electron extraction layer, low leakage current, and a suitable surface morphology for electron extraction and transport.

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### 1. Introduction

Photovoltaic offers a clean and renewable energy source for solving the future energy crises. Among all available solar cells, polymer BHJ solar cells have received a strong attention due to their advantages being lower cost, light weight, easily tunable optical properties, mechanical flexibility and simple fabrication process [1–4]. Recently, a highest efficiency of 9.2% for polymer based single junction solar cell was reported [5]. Although, the PCE of reported solar cell is impressive, however prime efforts are needed to improve the lifetime yield and performances of BHJ solar cells in order for them to be commercialized.

Nowadays, solution processed inverted BHJ polymer solar cells demonstrate a widespread interest due to their superior life-time yield and vertical phase separation [6–8]. In general, the inverted solar cell architecture comprises the most widely used transparent indium tin oxide (ITO) electrode to collect electrons and metal electrodes such as Au, Ag, or Al to collect holes from the photoactive layer. The ITO has a respectable conductivity and very high transparency in the visible wavelength region. However, ITO displays a low work function ( $\sim 4.8$  eV), which is not well enough to collect electrons efficiently from the acceptor of photoactive layer, thus resulting in poor solar cell performance index [9]. It is also well known that the energy level matching or barrier less contact in optoelectronic devices help to improve the charge carrier transport and extraction properties [10,11]. Hence, various efforts have been applied to improve their electron collection

competency such as modification of work function by using metal oxide buffer and conjugated polyelectrolyte layers respectively [9,12,13].

In polymer solar cells, in order to efficiently collect and transfer a charge carrier from photoactive layer to their respective electrode, an effectual charge extraction layer is needed. The purpose of cathode buffer layer is not only to decrease the work function of ITO electrode but also to form an ohmic contact with the hydrophobic photoactive layer. This could have properties such as easy-processing, worthy surface morphology, air stability, hole blocking capability and well matched energy level with the acceptor of photoactive layer. Recently, we have demonstrated an efficient and air stable transition metal oxide layer for hole extraction and transport from the photoactive layer in inverted BHJ solar cells [14]. Among various n-type metal oxides, ZnO is the most widely researched and a promising candidate for electron transport and hole blocking in organic solar cells due to their favorable characteristics such as good electron mobility, environmental stability, high transparency in the visible range and easy film processing ability [15].

The charge extraction layer and their interface with the electrode and photoactive layer have always been the decisive element for improving the performances of polymer solar cells. Mixed electron transport layer (mixed-ETL) is also one of the important techniques for achieving reasonable solar cell performances. However, solution processed mixed-ETL has not been adequately studied in the literature. There are few studies that have been reported on mixed-ETL for polymer solar cells. Xu et al. reported  $\text{Cs}_2\text{CO}_3$ :BPhen based hybrid ETL for inverted polymer solar cells and demonstrated an enhanced performances due to easy control of ETL morphology, chemical composition and

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conductivity at low processing temperature [16]. Similarly, ZnO and non-conjugated polymer based composite layer for electron extraction was reported by Small et al., they investigated improved electron collection ability by applying a post-UV ozone treatment on their studied ETL film [17].

In this paper, we demonstrate a facile solution processed n-type interfacial layer by blending ZnO and Cs<sub>2</sub>CO<sub>3</sub> for inverted BHJ solar cells. The Cs<sub>2</sub>CO<sub>3</sub> is used in this study due to its effective electron extraction characteristic, high transmission in the visible range, easy and low temperature processing. It is easily soluble in polar solvents. This material was reported as an effective electron selective layer for organic solar cells [18]. The main idea behind incorporation of mixed-ETL in BHJ solar cell due to their ideal electrical and optical characteristics such as deep HOMO level, high optical transparency, good electron transport property and decent mobility. Enhanced performances were obtained for the fabricated solar cells with the mixed-ETL composed of optimized concentration of Cs<sub>2</sub>CO<sub>3</sub> into intrinsic ZnO. The effect of Cs<sub>2</sub>CO<sub>3</sub> mixing concentration on the performances and film properties of polymer solar cells are studied. Such facile blended cathode buffer layer provides appropriate control over morphology, ease of processing and low annealing treatment.

## 2. Experimental method

### 2.1. Device fabrication

The commercially available patterned ITO coated glass substrates with a sheet resistance of 10–15 Ω/sq were used for the fabrication of solar cells. Prior to the spin coating of electron extraction layer, ITO substrates were cleaned in ultrasonic bath filled with water, followed by cleaning with acetone and isopropyl alcohol (IPA), each for 15 min and finally treated with UV-ozone for 10 min. The ZnO:Cs<sub>2</sub>CO<sub>3</sub> layer with different mixing concentrations (0 wt%, 0.2 wt%, 0.6 wt%, and 1 wt%) of Cs<sub>2</sub>CO<sub>3</sub> (99.995%, Sigma-Aldrich) in ZnO for 1:1 ratio were spin coated on top of the pre-cleaned ITO coated glass substrates. The thickness of mixed-ETL layer for different concentrations of Cs<sub>2</sub>CO<sub>3</sub> was kept at a constant value of ~30 nm by controlling the rpm of spin coater and then annealed on hot plate at 150 °C for 30 min in ambient conditions. Pristine ZnO (~30 nm) and ZnO (~30 nm)/Cs<sub>2</sub>CO<sub>3</sub> (~8 nm) double interlayers were used as a reference electron transport layer. The ZnO/Cs<sub>2</sub>CO<sub>3</sub> bilayer has been reported as an electron selective layer with enhanced solar cell performances [19]. In case of double interlayer, ZnO was spin coated onto ITO substrate followed by annealing at 150 °C for 30 min. Subsequently 0.2 wt% of Cs<sub>2</sub>CO<sub>3</sub> solution was spin coated on top of ZnO layer and resulting film was annealed at 150 °C for 10 min inside the glove box. The ZnO solution was prepared using a method reported by Heegar et al. [15]. The Cs<sub>2</sub>CO<sub>3</sub> was dissolved in 2-ethoxyethanol for the concentration of 0.2 wt%, 0.6 wt% and 1 wt% and stirred overnight in nitrogen filled glove box. Later, both well prepared ZnO (0.1 M) and Cs<sub>2</sub>CO<sub>3</sub> solutions were mixed together at 1:1 volume ratio. The precursor zinc acetate dehydrate, Cs<sub>2</sub>CO<sub>3</sub>, solvent 2-ethoxyethanol, and monoethalamine were purchased from Sigma-Aldrich and used as received. After depositing different ETLs on ITO substrates, all samples were transferred into nitrogen filled glove box for the deposition of photoactive layer. The blend of poly (3-hexylthiophene) (P3HT) (from Sigma-Aldrich) and indene-C60 bisadduct (ICBA) (from Luminescence Technology Corporation) was formed in 1,2-dichlorobenzene for 1:0.6 concentration (1.5 wt%) and spin coated on top of ETL layer at 1000 rpm for 30 s. The ~80 nm film of photoactive layer was consequently annealed on a hot plate at 150 °C for 10 min inside the glove box. Afterward, 5 nm of tungsten trioxide (WO<sub>3</sub>) (from Sigma-Aldrich)

and 100 nm of Al were thermally deposited on top of the active layer through shadow mask at a base pressure of 10<sup>-7</sup> Torr and deposited at the rates of 0.3 Å/s and 2.5 Å/s respectively. The overlap photoactive area between the patterned ITO and evaporated Al was 0.04 cm<sup>2</sup>. Finally all devices were encapsulated inside the glove box using transparent glass cover and curable UV resin.

### 2.2. Device characterization

The current density–voltage (*J*–*V*) characteristics of the fabricated inverted solar cells were obtained using a computer controlled Keithley 2400 source-meter in the dark and under an illumination intensity of 100 mW/cm<sup>2</sup> with a calibrated AM 1.5G sun simulator (McScience, K3000-LAB50) in ambient conditions. All measurements were carried out at room temperature, under relative humidity of 60%. The EQE measurements were performed using EQE system (Model 74000) obtained from Newport Oriel Instruments USA and HAMAMATSU calibrated silicon cell photodiode was used as a reference diode. The wavelength was controlled with a mono-chromator in the range 200–1600 nm. The morphology, surface roughness and chemical composition of the studied ETL were analyzed using atomic force microscopy (AFM) (SPA-400, Japan), X-ray photoelectron spectroscopy (XPS) (Thermo Electron, K-Alpha), respectively. The UV–vis characteristics of the samples were obtained using UV–vis spectrophotometer (JASCO, S-4100). The thickness measurements were carried out by using profilometer (alpha-step).

## 3. Results and discussion

The schematic representation of fabricated device architecture and molecular structure of donor and acceptor materials used in this study are shown in Fig. 1. The fabricated inverted BHJ solar cells include a blend of P3HT and ICBA based photoactive layer, WO<sub>3</sub> as a hole extraction layer and n-type mixed metal oxide layer for electron extraction. For valid comparison of the electrical properties of inverted solar cells, devices with pristine ZnO and ZnO/Cs<sub>2</sub>CO<sub>3</sub> double interlayers were fabricated and used as reference cells. The *J*–*V* characteristics of fabricated P3HT:ICBA based solar cells with pristine ZnO, ZnO/Cs<sub>2</sub>CO<sub>3</sub> bilayer and mixed ZnO:Cs<sub>2</sub>CO<sub>3</sub> electron transport layers (ETL) were measured under AM 1.5G illumination with light intensity of 100 mW/cm<sup>2</sup> are shown in Fig. 2. The solar cell devices with n-type mixed ZnO:Cs<sub>2</sub>CO<sub>3</sub> (0.2 wt%) ETL exhibits enhanced power conversion efficiency (PCE) as high as 5.29%, an open circuit voltage (*V*<sub>oc</sub>) of 0.87 V, a short circuit current density (*J*<sub>sc</sub>) of 8.78 mA/cm<sup>2</sup> and a fill factor (FF) of 69.04%. In contrast, the inverted solar cells with intrinsic ZnO and ZnO/Cs<sub>2</sub>CO<sub>3</sub> double ETL show a PCE of 5.06% and 5.14% respectively. The comparative electrical parameters of fabricated inverted BHJ solar cells are listed in Table 1. To confirm the reproducibility of

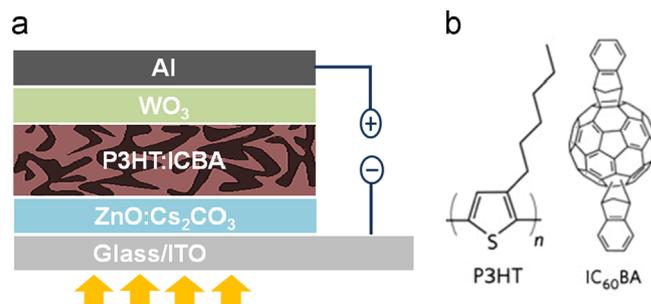


Fig. 1. (a) Schematic representation of fabricated inverted solar device structure and (b) molecular structure of donor (P3HT) and acceptor (ICBA) used as a photoactive layer.

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