



Efficient organic solar cells employing ytterbium ion-doped zinc oxide as cathode transporting layer

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ABSTRACT

Rare earth ions possess highly conductive, magnetic, electrochemical and luminescent properties, multiple valence electrons and long lifetime of excited state, thus could be alternatives as dopant to tune and promote the cathode interlayer property in solar cells. In this work, we study the modification of cathode interlayer by rare earth ion doping, based on the model of Yb doped ZnO by sol-gel method. A relative low doping concentration (below 1%) can improve the electron transport of ZnO and the solar cell performance. This is also confirmed by the obtained photocurrent density, excitons generation rate and electron mobility of device. The best PCE 11.04% (with V_{OC} 0.940 V, J_{SC} 16.58 mW cm^{-2} , FF 70.7%) is achieved with 0.5% Yb doping, based on PBDB-T:IT-M active layer, compared to the reference PCE of 10.19% with pure ZnO ETL. While with concentration over 1%, the Yb doping decreases the device performance.

1. Introduction

The interfacial layer between active layer and electrode play a crucial role in organic solar cells (OSCs) [1,2], as they may impact the device polarity by selectively transporting the charge carriers, energy barrier at the contact interface, light propagation and distribution, electrode surface affinity and the active layer morphology. Imperfect interface results in depressed device performance. Thus doping has been one of the effective strategy to tune the interface property and improve the solar cell performance [3–6]. Organic molecules and metal ions are often been used as dopant for metal oxide interfacial layer in OSCs [1,7], as a thin layer on top of metal oxide or doped into metal oxide compound of the whole interlayer respectively [6,8–10]. Doping helps to passivate the defects, reduce the charge carrier recombination, increase the charge carrier concentration and thus conductivity of the interlayer [11–14]. Organic molecules usually have been deposited on top of interlayer by simple spin-cast from a solution. It can further tune the electrode energy level and reinforce the actual device built-in potential due to the introduced interfacial dipole [15]. However, thin film modification is limited only at the interlayer surface but not the whole

interlayer. While doping with metal ions can provide uniform modification for the whole bulk interlayer, enhanced N/P type property of material, i.e. better conductivity and improved device stability [16–18].

As one of the most commonly used cathode interfacial materials, zinc oxide (ZnO) possesses advantages like suitable energy levels, easy synthesis, low cost, and environmental stability compared with other metal oxides [19–23]. However, ZnO synthesized by solution method usually exhibits moderate electron mobility, and tends to form trap states associated with defects [24–26], resulting in relatively inferior device performance. Thus efforts by doping strategy with metal ions have been conducted to solve the above problems [5,27–32]. Among which, Al doping is the most classical model. It can effectively enhance the N type property of ZnO by providing one more valence electron than the Zn atom [33]. In 2013, Tobias Stubhan and co-workers reported Al doped ZnO transparent interfacial layer [27]. Improved conductivity of the interlayer and promoted device performance were achieved. In 2016, our group reported ‘thickness-insensitive’ Al-doped ZnO cathode interlayer (CIL) in OSCs with a champion PCE of 10.42% [18]. In 2014, Mg doped ZnO (ZnMgO) cathode interlayer was prepared and applied in PTB7-Th:PC₇₁BM solar cell by Zheng and co-workers, a

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high PCE of 9.39% was presented due to the reduced interface energy losses and improved electron collection [16]. Solution processed Ga doped ZnO (GZO) interfacial layer presented higher electron conductivity and favorable surface morphology, thus improved hugely the OSC as well [30,33–35].

Besides, rare earth ion Yb has often been used as dopants (or sensitizer) for numerous applications due to their highly conductive, magnetic, electrochemical and luminescent properties [36–39]. It has multiple valence electrons (including 4f and outer valence electrons), and especially is stable in the trivalent state Yb [40]. We presume that Yb could be alternative as dopant to enhance the N type property of CIL in solar cells, like ZnO. The excited states of rare earth elements have separation levels and long lifetime [11,36,41–44]. Thus electrons have more chance to be transferred or extracted to electrode before they get recombined through radiative/thermal relaxation, hence benefiting device performance. Moreover, Yb does not absorb photons in visible but shows specific high extinction coefficient in IR range [45]. And Yb doped ZnO (YZO) shows low resistivity and high carrier mobility [46,47]. All these imply a good alternative of YZO for interfacial layer in photovoltaics. However, to the best of our knowledge, few work about YZO as CIL has been reported in literature.

Thus, in this work YZO as CIL was studied as a model, with the aim to explore how rare earth element doping will influence the ZnO interlayer property. Different mole ratio of Yb/Zn (0%, 0.05%, 0.25%, 0.5%, 1.0%, 2.0%) was applied to prepare the YZO by sol-gel method [18]. The influence of the Yb doping on the material's optical and electronic properties were tested. And the corresponding device performances were also studied and discussed based on the polymer non-fullerene active blend PBDB-T:IT-M.

2. Experimental section

2.1. Materials and reagents

Zinc acetate dehydrate ($\text{Zn}(\text{Ac})_2 \cdot 2\text{H}_2\text{O}$) were bought from Sinopharm Chemical Reagent Co. Ytterbium (III) nitrate pentahydrate ($\text{Yb}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$), Chlorobenzene (CB), ethanolamine (EA), 1,8-diiodoctane (DIO), and MoO_3 were obtained from Sigma-Aldrich. Patterned ITO glass substrates ($R_s \leq 15 \Omega^{-1}$, Transmittance $\geq 86\%$) were provided by Shenzhenhuananxiangcheng Technology Co. PBDB-T and IT-M were purchased from Solar Materials Inc. (China). All the available chemical reagents were used without further purification. ZnO and YZO were synthesized by sol-gel method. $\text{Zn}(\text{Ac})_2 \cdot 2\text{H}_2\text{O}$ (0.01 mol) and doping source of $\text{Yb}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ (molar ratio of Yb/Zn:0%, 0.05%, 0.25%, 0.5%, 1.0%, 2.0%) were added into anhydrous ethanol (100 mL) in a round-bottom flask. After stirring for 10 min at 80 °C, 0.61 mL ethanolamine (as stabilizer) was added into the reaction solution, this transparent solution was refluxed at 80 °C for 2 h [18,48].

2.2. Device fabrication

The device structure ITO/CIL/Active layer/ MoO_3 /Al was adopted. The ITO glass substrates were pre-cleaned by ultrasonic, sequentially in detergent, deionized water, acetone, and isopropyl alcohol for 15 min, and then dried by nitrogen stream and treated with UV-ozone for 10 min. Then, the solution of YZO with different Yb concentration (0%, 0.05%, 0.25%, 0.5%, 1.0%, 2.0%) were spin-coated on the ITO substrates under 2000 rpm for 1 min (ca. 25 nm). Then annealed at 140 °C for 20 min in air. Subsequently, the ITO substrates with ZnO (or YZO) film were transferred into a N_2 -filled glovebox. Then solution of PBDB-T:IT-M (1:1 w/w, 20 mg/ml) in chlorobenzene:DIO (99:1 v/v) was spin-coated on ITO/CIL under 3000 rpm for 2 min to obtain the active layer (100 nm). PTB7-Th:PC₇₁BM (1:1.5 w/w, 25 mg/ml) solution in chlorobenzene:DIO (97:3 v/v) was spin-coated on ITO/CIL substrate under 2000 rpm for 2 min to obtain the active layer (100 nm). Eventually, the device fabrication was finished by thermally

evaporating 10 nm MoO_3 and 100 nm Al under vacuum. The electron-only device is ITO/CIL/PBDB-T:IT-M/Ca/Al.

2.3. ZnO and YZO film characterization

Transmittance spectra were obtained by a GS54T spectrophotometer (Shanghai Lengguang Technology Co. China) in 300–800 nm range. Powder X-ray diffractometer (XRD) analysis were carried out on a Bruker D8 Advance X-ray diffractometer (XRD, Bruker D8 Advance, Germany) equipped with $\text{Cu K}\alpha$ radiation ($\lambda = 0.154 \text{ nm}$) and diffraction patterns were collected in range of 20–80°. X-ray photoelectron spectroscopy (XPS) measurement was performed on AXIS ULTRADALD XPS/ultraviolet photoelectron spectroscopy (UPS) system (Kratos, England). XPS was carried out under monochromatized $\text{Al K}\alpha$ ($h\nu = 1486.8 \text{ eV}$) excitation and UPS was carried out under helium discharge lamp excitation with $h\nu = 21.2 \text{ eV}$. The photoluminescence emission (PL) spectra was recorded by a fluorescence spectrometer (FL3-111, HORIBA, France) under 310 nm excitation. The film surface morphologies of interfacial and active layer were obtained by AFM (Veeco, Dimension 3100, America) in tapping mode. All measurements were performed at room temperature.

2.4. Device characterization

The *J-V* characteristics of devices were measured by a Keithley 2400 sourcemeter under AM1.5 solar spectra. The EQE curves were recorded through a Newport quantum efficiency measurement system (ORIEL IQE 200TM) with a lock-in amplifier in atmosphere. Each wavelength light intensity was calibrated with a standard Si/Ge solar cells. The electron mobility was obtained by fitting the dark current to the model of a single carrier space charge limited current (SCLC) following the equation $J = (9/8)\epsilon_0\epsilon_r\mu((V^2)/L^3)$, *J* is current density, μ is zero-field mobility, ϵ_0 is permittivity of free space, ϵ_r is the relative permittivity, *L* is thickness of active layer and *V* is effective voltage [16,18].

3. Results and discussion

3.1. Device structures and molecular structure as well as energy level diagram

The transmittance spectra of ZnO and YZO with different doping concentration on ITO glass substrate are shown in Fig. S1. High transmittance value over ~98% (the ITO glass background subtracted) in the visible range is recorded for different samples, ensuring big photon flux at the active layer after substrate. The adopted inverted solar cell structure, molecular structures of donor PBDB-T and non-fullerene acceptor IT-M, and the energy levels of the materials included in device are shown in Fig. 1. The HOMO/LUMO levels of the pure ZnO and 0.5% YZO interlayer were determined to be $-7.51/-4.17$ and $-7.65/-4.31 \text{ eV}$ respectively from the UPS and absorption spectra (Fig. S2). And the energy levels of ITO, PBDB-T and IT-M were taken from literature [49]. The YZO shows slightly deeper LUMO and HOMO than ZnO, implying better electron extracting and hole blocking ability from the active layer, but could slightly reduce the open circuit voltage (V_{OC}) of the device [28,50].

3.2. Device performance

Fig. 2a shows the current density–voltage (*J-V*) curves of the inverted solar cells with different doping concentration of Yb (0%, 0.05%, 0.25%, 0.5%, 1.0%, 2.0%) under AM1.5 G solar irradiation (100 mA cm^{-2}). The best performance was obtained when 0.5% Yb doping was applied. The photovoltaic parameters of devices with different doping concentration are shown in Fig. 2b. It shows that the J_{SC} , FF and PCE increases with Yb at low concentration, reach the maximum at concentration of ~0.5%, and beyond that the parameters start to

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