Improving the efficiency of inverted organic solar cells by introducing ferrocenedicarboxylic acid between an ITO/ZnO interlayer (Postprint)

Authors: Li, BR, Jiu, TG, Kuang, CY, Chen, QS, Ma, SS, Li, JS, Hou, XL, Fang, JF

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Abstract

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Full Text

Preamble

Improving the efficiency of inverted organic solar cells by introducing ferrocenedicarboxylic acid between an ITO/ZnO interlayer†

Bairu Li,ab Tonggang Jiu,*b Chaoyang Kuang,b Qiushan Chen,b Sushuang Ma,b Jiangsheng Li,b Xueling Houa and Junfeng Fang*b

In this study, ferrocenedicarboxylic acid (FDA) has been introduced between an ITO electrode and ZnO interlayer to improve the performance of inverted polymer solar cells. The highest power conversion efficiency (PCE) of 9.06% is achieved among the measurements. Besides, the average PCE of FDA/ZnO based devices is observed with 11.9% enhancement (8.73% vs. 7.80%) compared to ZnO-only devices.

Electrical characterization, surface morphology, wetting properties, as well as exciton generation rate and dissociation probability were investigated to understand the impact of FDA insertion on the interfacial properties. It was found that exciton dissociation efficiency and charge collection efficiency were significantly improved after inserting FDA, while the surface morphology, average roughness and water contact angle of the ZnO film were not changed. It was thought that FDA connected to the ITO electrode and ZnO film because of its carboxyl groups, which lead to a compact interfacial contact and reduced charge recombination. In addition, the devices based on the FDA/ZnO interlayers displayed improved stability in the argon-filled glove box without any encapsulation for about 1000 h compared to the ZnO-only devices. This study provides a new idea to introduce materials with functional groups between ITO/metal oxides interfaces to achieve more efficient charge collection and device performance.

Introduction

Bulk heterojunction (BHJ) polymer solar cells (PSCs) based on -conjugated polymers and different transport materials have attracted more and more attention recently due to their easy solution-based fabrication, large area, low cost and promising flexibility.1-6 Nowadays the highest power conversion efficiency (PCE) of a single junction bulk heterojunction PSC has exceeded 10%,7 and a triple-junction PSC can reach a PCE of 11.55%.8 Interfacial layers which locate between the active layer and the electrodes play a key role in improving the performance of PSCs.9-11 This is because they can block unwanted charge recombination and facilitate selective contacts for charge collection.9,12,13 In inverted PSCs, n-type semiconducting metal oxides such as ZnO14-16 and TiO2 17-19 are commonly used as effective interfacial electron transport layers owing to their excellent optical transparency, high electron mobility and chemical stability. However, these metal oxides interfaces often show leakage currents and interfacial recombination issues because of high surface defects and poor interfacial contact, which have seriously blocked the further improving of device performance.4,20 It is a simple and effective method by introducing an interface modification layer between metal oxides layer and active layer, or between ITO and metal oxides layer to reduce the charge recombination in cathode interlayer.

Materials such as poly(ethylene glycol) (PEG)21,22, poly(ethylene oxide) (PEO),20,23 conjugated polyelectrolytes24,25 and fullerene derivatives26,27 have been widely used to optimize the interfacial contact between metal oxides layer and active layer. Besides, insertion of polyoxyethylene tridecyl ether (PTE), copper hexadecafluorophthalocyanine (F16CuPc) interface modification layer between ITO and metal oxides layer has been also developed to improve the electron transport and increased device performance. Choulis group showed that the incorporation of polyoxyethylene tridecyl ether (PTE) between the ITO electrode and TiOx interlayer does not modify the energy levels but clearly affects the morphology, wetting properties and hydrophilicity of the TiOx, which led to the increased device performance.5 Marks group reported that the insertion of copper hexadecafluorophthalocyanine (F16CuPc) between the ITO electrode and ZnO layer reduced the energetic loss associated with unfavorable electron transport continuity and increased the interfacial area.28 Lee group achieved the higher device efficiency by using self-assembled monolayers with the terminal groups -NH2, -CH3, and -CF3 to modify ITO.29 Ouyang group reported that the zwitterion-modified ITO sheets were used as the cathode for the electron collection of inverted PSCs.30 All these results present the great potential to increase the PCE by introducing new interfacial materials between the electrode and interfacial layer. Also, the surface modification of ITO electrode is still very important. So far the introduction of interfacial materials with different anchoring group lay in the interface between metal oxide and ITO electrode is rarely reported and necessary to figure out the intrinsic mechanism of interfacial modification and the effect on the device performance.

Ferrocene derivatives are widely used in the field of catalyst,31 drug delivery,32 electrochemistry33 because of their unique chemical structure, high thermal stability and chemical stability. There is a potential to apply them in PSCs for improving the device performance and stability due to its excellent physical and chemical properties. In this work, ferrocenedicarboxylic acid (FDA) was inserted between ITO electrode and ZnO interlayer to enhance electron transport and improve the stability of inverted PSCs. Low bandgap polymer thieno[3,4-b]thiophene/benzodithiophene (PTB7) was used as the electron donor and [6,6]-phenyl-C71 butyric acid methyl ester (PC71BM) was used as the electron acceptor. Photovoltaic devices based on an ITO/FDA/ZnO/PTB7 : PC71BM/MoO3/Al structure (Fig. 1 [Figure 1: see original paper]) showed significant enhancement in short-circuit current density (Jsc) and fill factor (FF), which led to a best PCE of 9.06% and an average PCE of 8.73%. Electrical characterization, surface morphology, wetting properties, as well as exciton generation rate and dissociation probability were investigated to understand the impact of FDA insertion on the interfacial properties.

2.1 Materials

Zinc oxide nanoparticles were prepared using an adapted procedure based on the work of Janssen et al.34 After a centrifugation step, the precipitate was dissolved in chloroform and methanol (1 : 1 vol%). FDA were purchased from Sinopharm Chemical Reagent Co. Electron donor material PTB7 and electron acceptor material PC71BM were purchased from 1-Material Chemscitech Inc. All these commercially available chemicals were used as received.

2.2 Device fabrication

The indium tin oxide (ITO) glass substrates were cleaned by sequential ultrasonic treatment in detergent, deionized water, acetone and isopropanol for 15 minutes respectively, and then blown dry with nitrogen. Subsequently the substrates were treated by UV-ozone for 20 minutes. Ferrocene derivative FDA in dimethyl sulfoxide solutions with various concentration (0.5 mg mL ^{1}, 1 mg mL ^{1}, 2 mg mL ^{1}) were spin-coated at 1500 rpm for 5 min on the top of ITO substrates and then the films were annealed at 100 °C for 10 min. The thicknesses of FDA layers were determined to be ~8.9 nm and ~13.2 nm corresponding to the 1 mg mL ^{1} and 2 mg mL ^{1} concentrations, respectively. The FDA film from 0.5 mg mL ^{1} was too thin to measure its thickness.

Next, the ZnO nanoparticles solution with a concentration of 15 mg mL ^{1} was spin-coated at 2500 rpm on the FDA-coated ITO substrates and then annealed at 80 °C for 10 min. All substrates were transferred to the argon-filled glove box for further processing. Then the active layer was made by spin-coating the active materials in mixed solvent of chlorobenzene/1,8-diiodoctane (97 : 3 vol%) at 2000 rpm for 120 s on the top of interfacial layers. The blend ratio of PTB7 : PC71BM was 1 : 1.5 by weight (25 mg mL ^{1}). The device fabrication was completed after thermal evaporation of 10 nm of MoO3 and 100 nm of Al as the anode under vacuum at a base pressure of about 1 × 10 Torr. The device area was 0.04 cm². All devices were fabricated from the same solution under the same condition.

2.3 Measurements and characterization

The current density-voltage (J-V) characteristics of the photovoltaic devices were measured using a computer-controlled Keithley 2400 sourcemeter. And the characterization was performed in glove box under a simulated AM 1.5 G solar illumination (100 mW cm ^{2}) with an Oriel So13A solar simulator. The EQE measurements were performed with the Newport IQE-200 measurement system, which equipped a Xe lamp, a monochromator, a current-voltage preamplifier, and a lock-in amplifier. Atomic force microscope (AFM) was operated in tapping mode by using a Veeco dimension atomic microscope at room temperature.

3 Results and discussion

Fig. 1 shows the device configuration and the chemical structure of FDA, PTB7, and PC71BM. The current density-voltage (J-V) characteristics of both devices with and without FDA interfacial modification layer between ITO/ZnO were demonstrated in Fig. 2a. The detailed device parameters were summarized in Table 1 . The control device based on ZnO-only layer exhibited an open circuit voltage (Voc) of 0.749 V, a Jsc of 16.5 mA cm ^{2}, a FF of 65.0% with a corresponding PCE of 8.03%. However, when FDA was inserted between ITO electrode and ZnO interlayer, the Jsc and FF were enhanced significantly. The device with an optimized concentration 1 mg mL ^{1} (see Table S1 in the ESI†) exhibited improved PCE of 9.06%, which was higher than that of ZnO-only device (8.03%). A higher Jsc and FF than ZnO-only device, indicating that incorporating FDA can effectively reduce leakage current and enhance electron transport to ITO, which can be proved to some extent by the decrease of series resistance (Rs) from 7.0 Ω cm² to 5.4 Ω cm² in Table 1.

In order to ensure the enhanced performance by inserting FDA interlayer between ITO and ZnO, 15 devices with and without FDA interlayer were fabricated under the same condition to examine the reproducibility. The distribution of different parameters of these devices are shown in Fig. 3 [Figure 3: see original paper]. As can be seen, the Jsc, FF and PCE based on FDA/ZnO devices were indeed higher than those of ZnO-only devices. The average PCE of devices with FDA interfacial modification layer was 8.73% (Table 1), which was 11.9% higher than ZnO-only devices (7.80%). This suggested the device could present excellent performance and favourable reproducibility by using FDA/ZnO bilayer as cathode interfacial layer.

The external quantum efficiency (EQE) of devices with and without FDA interfacial modification layer was measured to verify the Jsc (Fig. 2b). It is apparent that the EQE of FDA/ZnO interlayer based device is slightly higher than that of ZnO-only device in the wavelength region of 350-700 nm, which is in agreement with the measured Jsc. It can be noticed that the Jsc calculated from the EQE (14.83 mA cm ^{2} for ZnO-only device and 15.23 mA cm ^{2} for FDA/ZnO device) are lower than those obtained from J-V curves (16.5 mA cm ^{2} and 17.7 mA cm ^{2}, respectively). This arises from the degradation of PTB7 : PC71BM based devices due to the oxygen and moisture in the EQE test environment. Fig. 2c depicts the dark J-V characteristic of devices with and without FDA interfacial modification layer.

Evidently, the ZnO-only device showed a high leakage current in the reverse direction. In addition, the device incorporating FDA interfacial modification layer exhibited favourable diode characteristics with low leakage current and higher rectification ratio compared with the ZnO-only devices. All of these results demonstrated the superior interface properties and the improved electron transfer ability of FDA/ZnO interlayer. As shown in Table 1, the series resistance Rs in the FDA/ZnO based devices decreased when a thin FDA film was introduced (5.4 Ω cm^2 vs. 7.0 Ω cm^2). In addition, the rectification ratio (RR) at ±1.5 V increased significantly (1.42×10 vs. 1.49×10^3), indicating improved diode characteristics.35 The good electrical characteristics of the FDA/ZnO based devices are consistent with the performance data in Table 1, which are responsible for the enhanced FF in FDA/ZnO based devices.

The performance of inverted PSCs is often closely associated with the surface morphology of cathode interlayer.36-38 In order to investigate the effect of FDA insertion on the surface morphology of ZnO, we used atomic force microscope (AFM) measurement to examine the surface morphology of ZnO with and without FDA insertion, as shown in Fig. 4 [Figure 4: see original paper]. From the AFM images, we can see that the morphology of ZnO with and without FDA interfacial modification layer is almost the same. The root-mean-square roughness (RMS) of FDA/ZnO film was 3.83 nm, which was similar to the ZnO-only layer (3.77 nm). This implied that both of the ZnO film with and without FDA insertion possess typical uniformly smooth surface. This is corresponding to the favorable device performance in this work.

Besides, the water contact angle of water on FDA/ZnO surface as well as ZnO surface were conducted, as displayed in Fig. 4 inset. The contact angle on FDA/ZnO surface was not changed compared with the ITO/ZnO surface. Therefore, active layer with the same thickness and morphology could be obtained on ZnO layer and FDA/ZnO bilayer. Hence, we could conclude that surface morphology of ZnO interlayer is not influenced by FDA insertion and it is not the main factor that enhance the Jsc and FF in devices.

In order to figure out whether the electron mobility has been changed after the insertion of FDA, the electron mobility was approximated using space charge limited current (SCLC) method. Electron only devices with structures of ITO/ZnO/Ca/Al and ITO/FDA/ZnO/Ca/Al were constructed. And current-voltage data were plotted as ln(JL^{3}/V^{2}) versus (V/L) • by fitting to the Mott-Gurney law, as shown in Fig. 5a [Figure 5: see original paper]. The electron mobility which was calculated from the intercept of the corresponding lines on the axis of ln(JL^{3}/V^{2}) is 7.203 × 10^{3} cm^{2} V^{1} s^{1} for ZnO based device and 1.366 × 10^{1} cm^{2} V^{1} s^{1} for FDA/ZnO based one, respectively. This result suggested that the charge transport mobility of cathode interlayer increased after inserting FDA. Fig. S2† shows the impedance spectra (measured in the dark) of devices with and without FDA interlayer at open-circuit voltage. Two semicircles were observed in the Nyquist plots, which presented the charge transfer resistant (Rct) related to the recombination of electrons in the interlayer. Larger Rct after inserting of FDA means less electron recombination at the surface which suggests FDA has prohibited the electron-hole recombination and improved the electron transport. It thus contributes to the FF enhancement.

To further analyze the origin of the higher Jsc and FF in FDA/ZnO based device, photocurrent density (Jph) versus effective voltage (Veff) characteristics of devices with and without FDA interlayer were plotted in Fig. 5b. The Jph-Veff curves were used to investigate the effect of interlayers on the charge collection efficiency.39-42 Photocurrent density (Jph) is given by Jph = JL -JD, where JL and JD are the current density under illumination and in the dark, respectively. Effective voltage (Veff) is given by Veff = V0 -V, where V0 is the compensation voltage at which Jph = 0 and V is the applied voltage. As we can see from Fig. 5b, Jph reaches saturation for both devices at a large reverse voltage (i.e., Veff = 2 V, Jph 175 A m^{2}), suggesting all photogenerated excitons are dissociated into free carriers and all carriers are collected at the electrodes without any bimolecular recombination. In this situation, saturation current density Jsat is only limited by the amount of absorbed incident photons. Therefore the maximum exciton generation rates (Gmax) which could be given by Jsat = qGmaxL (where q is the electronic charge and L is the thickness of the active layer) are almost the same (about 1.09 × 10^{2}) in both devices. But it is noticed that the ratios between the net photocurrent density and the observed saturation photocurrent density (Jph/Jsat) in the devices at lower effective voltage (Veff < 0.3 V) are obviously different. For example, at lower effective voltage (Veff = 0.2 V), the value of Jph/Jsat for device with FDA/ZnO interlayer is 84.6%, while it is only 76.7% for ZnO-only device. Since Jph/Jsat is essentially the product of exciton dissociation efficiency and charge collection efficiency, a higher Jph/Jsat value indicates an increased exciton dissociation efficiency. It suggests that the exciton recombination has been suppressed when inserting FDA between ITO and ZnO and thus leads to a higher Jsc and FF. The higher exciton dissociation efficiency indicated that the FDA have better interfacial modification role. This can be supposed that the interaction between the FDA and ITO,43 as well as

ZnO27,44 bridged by the two carboxyl groups results in a better interface contact and reduces exciton recombination at ITO/ZnO interface so as to increase the electron transport. Scheme which illustrate the mechanism of the reduced charge recombination is shown in Fig. S3.† In addition UPS measurements (see Fig. S4 in the ESI†) indicate that FDA modification do change the working function of ITO surface. But the open circuit voltage didn' t change which indicate the FDA modification only play a role of electron transfer during the device working. It can be concluded that FDA layer didn' t change the surface morphology but improve the interfacial contact. Correspondingly it helps electron transport from ZnO to ITO due to the improvement of the interfacial contact by carboxyl bridges.

In order to investigate the effect of FDA insertion on device stability, we performed the lifetime measurement of both ZnO-only and FDA/ZnO devices by monitoring the parameters versus storage time in an argon filled glovebox. The results have been shown in Fig. 6 [Figure 6: see original paper]. As we can see, the PCE of devices using FDA/ZnO as the interlayer decays only 22.38% of the original value after storage for 1000 hours, which is obviously superior to that of ZnO-only device (28.98%). It indicates that FDA/ZnO based devices degraded slower than the ZnO-only devices did. This is attributed to the favorable thermal stability and chemical stability of FDA. It is suggested that interfacial modification by inserting FDA layers between ITO electrode and ZnO interlayer is beneficial for device stability.

4 Conclusion

In conclusion, we successfully fabricated PSCs with best efficiency of 9.06% by inserting the ferrocene derivatives (FDA) containing two carboxyl groups between ITO electrode and ZnO interlayer. Solar cells exhibited enhanced Jsc and FF compared to ZnO only devices. Several measurements such as AFM measurement and Jph–Veff curves were conducted to give detailed analysis of the improved device performances. AFM results revealed that surface morphology, average roughness of ZnO film with and without FDA insertion were not changed. But Jph–Veff curves showed that charge recombination was significantly reduced and exciton dissociation efficiency was improved. The results indicated that the improved FF in the FDA/ZnO based device rises from the reduced Rs and the better electrical characteristics. The enhancement of Jsc in the FDA/ZnO based device results from the higher exciton dissociation efficiency and charge collection efficiency. It is suggested that insertion of FDA could effectively improve the interfacial contact of ITO electrode and ZnO film, so as to enhance the device performance. Besides, FDA/ZnO based devices displayed significant long term stability. All these results indicated that inserting ferrocenedicarboxylic acid modified layers provides a promising strategy for achieving efficient and stable inverted polymer solar cells.

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