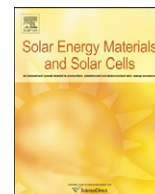




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## Mechanism for dimethylformamide-treatment of poly(3,4-ethylenedioxythiophene): poly(styrene sulfonate) layer to enhance short circuit current of polymer solar cells

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### ABSTRACT

Dimethylformamide (DMF), an organic solvent, was used to treat the poly(3, 4-ethylenedioxythiophene): poly(styrene sulfonate) (PEDOT: PSS) layer in poly(3-hexylthiophene) (P3HT): [6,6]-phenyl C61-butyric acid methyl ester (PCBM) polymer solar cells, resulting in significant enhancement of photocurrent and power conversion efficiency (PCE) improvement by 70%. Analyses of *I*–*V* characteristics reveal that the change in the active layer rather than that of the PEDOT: PSS buffer layer is ascribed to performance improvement. AFM images indicate that the roughness of PEDOT: PSS layer has been increased after the treatment. We argue that the protrudent PEDOT: PSS could serve as the centers for an initial crystallization of P3HT chains leading to a better alignment of P3HT: PCBM domains for a greatly enhanced photocurrent.

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### 1. Introduction

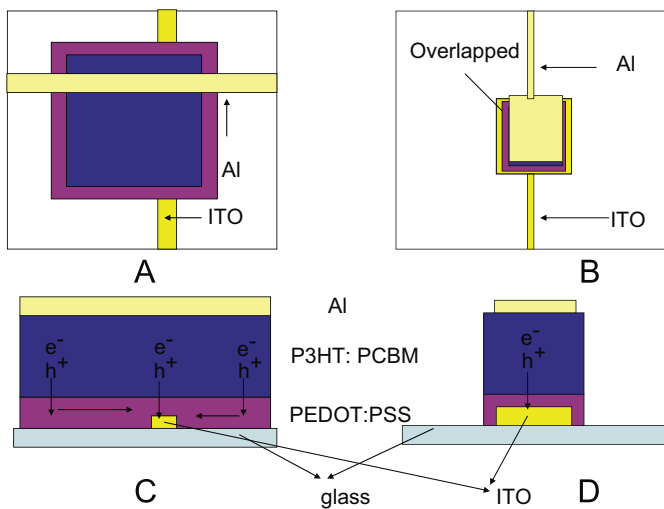
Since its discovery in 1986 [1], the organic photovoltaic (OPV) device has aroused great interests in both fundamental research and practical applications [2–9]. In most OPV devices, poly(3,4-ethylenedioxythiophene): poly(styrene sulfonate) (PEDOT: PSS) is used as an important buffer layer for charge transport or electrical interconnect due to its good transparency, high charge transport ability, excellent stability and solution-based fabrication process [10–18]. It has been reported that the conductivity of PEDOT: PSS layer can be enhanced by organic solvent treatment [19–21], the conductivities of PEDOT: PSS films treated with dimethyl sulfoxide (DMSO), N,N-dimethyl acetamide (DMAc), N,N-dimethyl formamide (DMF) and dichloromethane (DMC) were reported to be  $85 \pm 15$ ,  $45 \pm 10$ ,  $36 \pm 7$  and  $20 \pm 6$  S/cm, respectively, much higher than the value of an untreated PEDOT: PSS film, which is only  $2 \pm 0.05$  S/cm [22]. The organic solvent treated PEDOT: PSS layer has been used in polymer solar cells and significantly improved the solar cell performance [23–25]. Different concentrations of ethylene glycol are added into PEDOT: PSS layer to optimize the solvent treatment and obtain 29% higher power conversion efficiency (PCE) than has been obtained by

a pristine device [23]; the improved conductivity of PEDOT: PSS film has been proposed to be responsible for the enhanced short circuit current ( $J_{sc}$ ) and fill factor (FF) that result in higher PCE. Ethanol, 2-propanol and acetonitrile–water are also used to treat PEDOT: PSS layer to have higher PCE than that of devices without treatment, and the improvement is attributed to improved conductivity and surface morphology as well [24,25]. Although the enhancement effect of solvent treated PEDOT: PSS films on device performance is well confirmed, the mechanism is not completely proved due to the improved film conductivity. Experimental results reported by Kim et al. [26] show that the four types of PEDOT: PSS films with different conductivities are used in polymer solar cell and the PCE is independent of the conductivity of the PEDOT: PSS layer. Thus, there is a need to further investigate the enhancement mechanism. In addition, the active layer of a polymer solar cell is neighbored with the PEDOT: PSS layer, and whether the solvent treatment affects its performance or not has not been studied. In this work, dimethylformamide (DMF), an organic solvent, is used to treat PEDOT: PSS layer for the study of its performance enhancement mechanisms. The effect of volume ratio of DMF to PEDOT: PSS aqueous solution on performance is explored. In particular, AFM images of the treated PEDOT: PSS layer and solar cell parameters extracted from *I*–*V* curves are used to study the enhancement mechanism.

As shown in Fig. 1, in the most commonly used crossbar type cells (Fig. 1A and C), since the illuminated area is always larger than the overlapped area of the crossbar electrodes, excess

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**Fig. 1.** Top-view (A, B) and cross section (C, D) of the crossbar-type device (A, C) and the structure (B, D) used in this study.

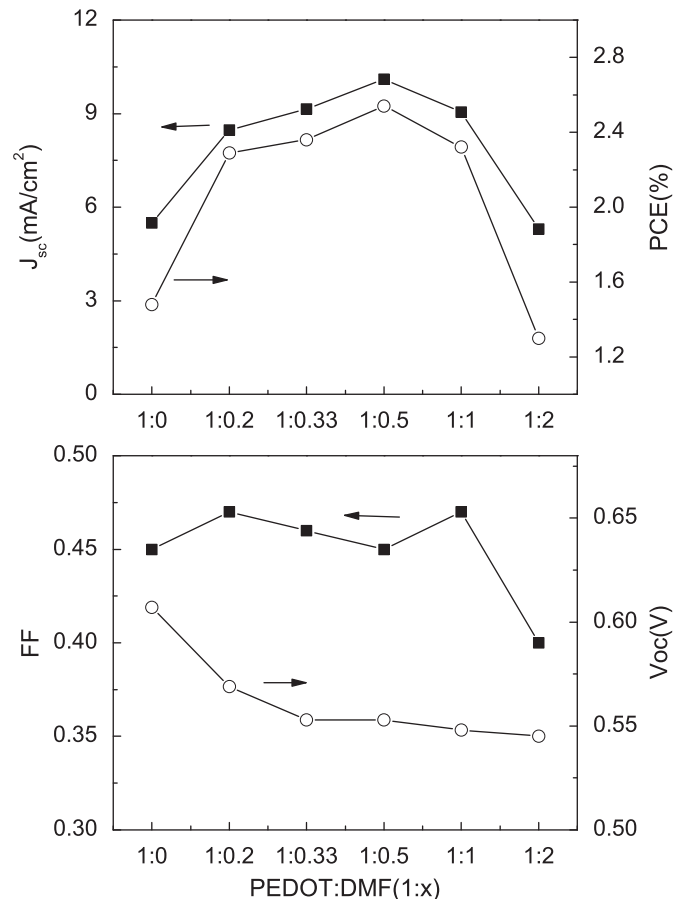
photocurrent can be generated from the regions outside the overlap area, where the conductive PEDOT: PSS layer plays the role of an effective anode [25,26]. In this type of device, as shown in Fig. 1C and D, electrons ( $e^-$ ) and holes ( $h^+$ ) can be generated in the side areas where no ITO electrode is coated on the glass. It is often that the side area is comparable to the overlapped area. Obviously, the device is not suitable to study the effect of the solvent treatment on photocurrent generation since the enhanced photocurrent from the solvent treatment can be magnified by the photocurrent produced from the side areas when the conductivity of PEDOT: PSS increases. Thus, a structure is designed here to exclude the photocurrent generated from the side areas. As shown in Fig. 1B and D, our devices precisely defined a large active area ( $1\text{ cm} \times 1\text{ cm}$ ) through etching the ITO electrode and shielding the extra aluminum deposited on the top of the devices by adhesive tape. Therefore, the side area (around  $0.1\text{ cm} \times 1\text{ cm}$ ) is much smaller to the active area ( $1\text{ cm} \times 1\text{ cm}$ ). The side area photocurrent effect in such a device is negligible.

## 2. Experimental

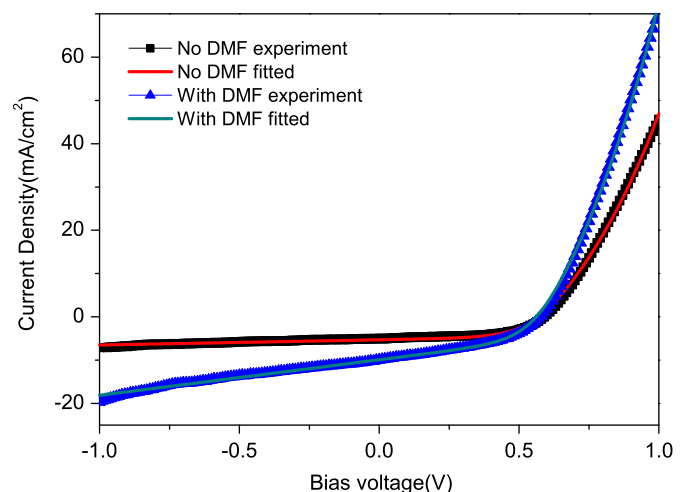
P3HT (Aldrich) and PCBM (American Dye Source) were dissolved in chlorobenzene (Aldrich) by heating and ultrasonating at  $80\text{ }^\circ\text{C}$  for 30 min. Hydrochloric acid (Aldrich,  $\sim 5\%$ ) was used to etch the indium tin oxide (ITO) coated glass substrate to form a  $1\text{ cm} \times 1\text{ cm}$  ITO pattern as shown in Fig. 1B. Then these substrates were cleaned by detergent and de-ionized water. Highly conductive PEDOT: PSS (Aldrich, 1.3 wt% dispersion in  $\text{H}_2\text{O}$ ) blended with DMF (Aldrich) at various ratios was spin-casted (5000 rpm for 30 s) onto the cleaned ITO patterned glass substrate (with a resistivity of  $12\ \Omega/\text{square}$ ). After drying the PEDOT: PSS layer at  $140\text{ }^\circ\text{C}$  on a hotplate in air for 10 minutes, the chlorobenzene solution containing 20 mg/ml P3HT and 16 mg/ml PCBM was spin-casted, followed by deposition of a  $\sim 100\text{ nm}$  aluminum film in vacuum ( $< 1.5 \times 10^{-5}\text{ Pa}$ ) with adhesive tape-defined surface area. Then the tapes were removed and thermal annealing was carried out in a digitally controlled vacuum oven (VOS-201SD, EYELA). The current–voltage ( $I$ – $V$ ) characteristics were measured by Keithley 2420 under illumination of a Newport solar simulator with AM1.5G filter. AFM images were obtained from a Dimension 3100 machine. All the measurements were carried out in air at room temperature.

## 3. Results and discussion

The effect of concentration of DMF in the PEDOT: PSS solution shown in Fig. 2 reveals that open circuit voltage ( $V_{oc}$ ) decreases continuously with the increasing DMF, while the fill factor (FF) is relatively stable before the amount of DMF exceeds PEDOT: PSS solution. However, short circuit current density ( $J_{sc}$ ) significantly



**Fig. 2.** Relationship between photovoltaic parameters and PEDOT: DMF composition (plotted as DMF volume ratio): (a)  $V_{oc}$ , (b)  $J_{sc}$ , (c) FF and (d) PCE vs. DMF fraction.



**Fig. 3.**  $J$ – $V$  characteristics of ITO/PEDOT-PSS/P3HT: PCBM/Al devices with (volume ratio is 1:0.5) and without DMF treatment under  $100\text{ mW}/\text{cm}^2$  AM1.5G simulated illumination with the fitted data.

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