



## Planar-diffused photovoltaic device based on the MEH-PPV/PCBM system prepared by solution process

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### ABSTRACT

A planar-diffused photovoltaic device based on Poly[2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylenevinylene] (MEH-PPV) and [6,6]-phenyl C<sub>61</sub>-butyric acid methyl ester (PCBM) has been prepared by a simple solution process, in which the PCBM organic solution was spin-coated onto the underlying MEH-PPV layer to fabricate the MEH-PPV/PCBM planar-diffused active layer. Investigation of the effects of active layer thicknesses and solvents on the performance of planar-diffused photovoltaic devices indicates that, with increasing the underlying MEH-PPV layer thickness, a gradual transition from absolutely penetrated to planar-diffused active layer structure occurs and the best power conversion efficiency is obtained for the device prepared by spin-coating a non-aromatic chloroform solution of PCBM onto the MEH-PPV layer, rather than device prepared by spin-coating the mixed solution of chloroform and chlorobenzene or aromatic chlorobenzene solution of PCBM onto MEH-PPV layer. Based on the photovoltaic performance, a structural model of planar-diffused active layer is proposed.

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### 1. Introduction

In the last decade, organic polymer solar cells, having the advantages of low cost, flexibility, large area and light weight, have received great interest in the domains of academia and industry. With the common goal of increasing the power conversion efficiency (PCE), much effort has been spent on material synthesis and device structure design. Materials, such as organic small molecules [1–3], star-shaped oligomers [4,5] and conjugated low band gap polymers [6–9], have attracted more attention in worldwide research because of their inexpensive, easily processable and tailored functionality by molecular design and chemical synthesis. At the same time, the concept of the photovoltaic (PV) device structure has also evolved from single layer [10], double layer [11] to bulk heterojunction (BHJ) [12], which is now the most successful and widely adopted organic PV device structure. The BHJ device structure, which exhibits a largely increased interfacial area where charge separation occurs and recombination is reduced, can be obtained by simple spin-coating from a solution of homogeneous blends. Recently, PCEs approaching 5% have been reported for BHJ devices based on rr-P3HT blended with [6,6]-phenyl C<sub>61</sub>-butyric acid methyl ester (PCBM) [13,14]. To date, the highest efficiency of 6.5% has been realized by using promising materials of PCPDTBT, P3HT, PCBM

and a tandem structure [15]. In order to successfully apply the organic polymer solar cells in commercial production and real life, not only the high PCE but also the large area [16,17] and stability [18–20] are equally important in solar cell technologies. Efforts on large-area and stable organic solar cells have been made in recent years but are still very limited [21]. Polymers have been examined so far for stability in solar cells, mainly the Poly[2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylenevinylene] (MEH-PPV) [22], P3HT [23] and P3CT [24].

Apart from BHJ device structure, several groups have dedicated themselves to developing different processing methods for fabricating various PV devices based on different donor–acceptor material systems to improve PV performance. For example, Granstrom et al. [25] applied a lamination technique to fabricate a diffuse bilayer heterojunction PV device based on a polymer/polymer material system, and a PCE of 1.9% under a simulated solar spectrum was reported. Drees et al. [26,27] created a gradient BHJ device based on a polymer/C60 material by a thermal-controlled diffusion processes. Fujii et al. [28] prepared interpenetrating conducting polymer/C60 heterojunction using a solution casting method, in which the polymer solution was spin-coated onto the underlying insoluble C60 layer. Osasa et al. [29] fabricated organic solar cells with a bulk-heterojunction structure by annealing stacked aromatic amine and C60 layers.

Actually, the PCE of PV devices is determined by four main factors: the efficiency of geminate electron–hole pair (exciton) generation, the efficiency of exciton separation into free charges, the efficiency of uninterrupted charges transport to respective

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electrodes and the efficiency of charges collection by electrodes. Therefore, an ideal PV device structure is a planar-diffused structure, which has an enlarged donor–acceptor interface and a spatially uninterrupted pathway for transporting the opposite charge carriers to their corresponding electrodes. Chen et al. [30] prepared a stratified multilayer PV device based on MDMO-PPV and PCBM. In their case, the solvent selected is a poor solvent for the underlying MDMO-PPV layer, which results in a lower PCE of 0.5% due to the limited donor–acceptor interface.

Here, we prepared planar-diffused PV devices based on MEH-PPV and PCBM with the structure of ITO/PEDOT:PSS/MEH-PPV/PCBM/LiF/Al, completely utilizing a simple solution process. We selected chloroform (CF), a mixture of CF and chlorobenzene (CB) (1/1, v/v), and CB as the solvents for spin-coating the PCBM layer, all of which are good solvents for the underlying MEH-PPV layer. On increasing the underlying MEH-PPV layer thickness, a gradual transition from absolutely penetrated to planar-diffused active layer structure occurred. The effects of active layer thicknesses and solvents on the PV performance are also investigated via UV–VIS absorption spectra, photoluminescence (PL) spectra, atomic force microscopy (AFM) measurement, incident photon-to-electron conversion efficiency (IPCE) and current–voltage ( $J$ – $V$ ) characteristics. According to the PV performance, a structural model of planar-diffused active layer has been proposed to explain the solvent-dependent PV performance.

## 2. Experimental detail

MEH-PPV and PCBM were synthesized in our group according to the literature [31,32] and confirmed by  $^1\text{H}$  NMR measurement. The molecular weight ( $M_n$ ) of MEH-PPV  $2.25 \times 10^5$  g/mol and a polydispersity of 3.6 were determined by gel permeation chromatography. Planar-diffused PV devices with the structure of ITO/PEDOT:PSS/MEH-PPV/PCBM/LiF/Al were fabricated in the following way. ITO glass (sheet resistance of  $20 \Omega/\text{sq}$ ) serving as the transparent anode was cleaned by conventional methods. A 50 nm layer of poly-[ethylene dioxy-thiophene]:(poly-(styrene sulphonic acid) (PEDOT:PSS) (Bayer PVP Al 4083) was spin-coated onto the pre-cleaned ITO glass substrate and then dried at  $110^\circ\text{C}$  for 20 min in a vacuum oven. MEH-PPV solution was only prepared in CF solvent at the weight concentration ranging from 5 to 6.5 mg/ml and stirred overnight under ambient condition to ensure that it is fully dissolved. PCBM was dissolved in three different solvents, namely CF, a mixture of CF and CB (1/1, v/v) and CB at a constant weight concentration of 5 mg/ml. For the fabrication of a planar-diffused active layer, MEH-PPV was first spin-coated from the CF solution onto the PEDOT:PSS layer and the thickness of the MEH-PPV layer was controlled by changing the concentration of MEH-PPV solution and spin speeds. Second, PCBM dissolved in three different solvents was spin-coated on the top of the MEH-PPV layer at a constant speed of 2000 rpm for 20 s and additionally dried at 1000 rpm for 1 min. Finally, a back electrode of LiF (0.6 nm) and aluminum (100 nm) was thermally evaporated in high vacuum at pressure better than  $5 \times 10^{-4}$  Pa, providing devices with the active area of  $2 \times 2.5 \text{ mm}^2$ . Fifteen different PV devices were prepared and classified into three sets according to the solvent from which the PCBM was spin-coated on the top of MEH-PPV layer. The corresponding sets of devices were named Device CF, Device MIX and Device CB. Each set consisted of five devices with different active layer thickness, which was adjusted by changing the underlying MEH-PPV layer thickness. For simplicity, devices in each set were coded. For example, in the Device CF set, five devices were marked as Device CF-1, Device CF-2, Device CF-3, Device CF-4 and Device CF-5. Table 1 shows the thicknesses of the underlying MEH-PPV layers and the planar-

**Table 1**

Thickness of the underlying MEH-PPV layer and planar-diffused active layer in Device CF, Device MIX and Device CB.

	The underlying MEH-PPV layer (nm)	Planar-diffused active layer (nm)		
		Device CF	Device MIX	Device CB
1	45	55	50	20
2	50	65	55	35
3	100	75	70	45
4	150	95	80	55
5	200	120	100	80

diffused active layers in three sets of planar-diffused PV devices. Devices with the same sequence number have the same thickness of the underlying MEH-PPV layer; however, after PCBM solution spin-coating, the thickness of planar-diffused active layers in Device CF, Device MIX and Device CB varies. These results correspond to the fact that the MEH-PPV film is eroded by additional PCBM solution. In the following part, this will be confirmed by the reduction of the absorption of the MEH-PPV film after spin-coating the PCBM solution. Both the thicknesses of the first pure MEH-PPV layer and the final planar-diffused active layer were measured on an Ambios Tech. XP-2 profilometer. UV–VIS absorption and PL spectra were recorded using a Shimadzu UV-3100 Spectrometer and an RF-5301PC spectrometer, respectively. The surface morphology of the active layer was characterized using a Nanoscope IIIa Dimension 3100 AFM operating in the tapping mode. Current density–voltage ( $J$ – $V$ ) measurement of the devices was conducted on a computer-controlled Keithley 2400 source meter measurement system. Air Mass 1.5 global (AM 1.5G)  $100\text{-mW}/\text{cm}^2$  illumination from a SCIENCETECH 500-W solar simulator acted as a light source. The spectral response of IPCE was measured under illumination of monochromatic light from the xenon lamp using a monochromator (Jobin Yvon, TRIAX 320) and detected by a computer-controlled Stanford SR830 lock-in amplifier with a Stanford SR540 chopper. A calibrated Si photodiode (UDT Instruments) was used as a standard for the determination of IPCE. All the measurements were performed under ambient atmosphere at room temperature.

## 3. Results and discussion

### 3.1. Absorption of the planar-diffused active layer

Fig. 1 shows the UV–VIS absorption spectra of pristine films and the active layers of Device CF, Device MIX and Device CB. A 100 nm underlying MEH-PPV film was spin-coated from a CF solution onto a quartz substrate. The absorption spectrum of the pristine MEH-PPV film shows a strong absorption peak at 493 nm and a weak absorption peak at 330 nm, whereas the pristine PCBM film shows a strong absorption peak at 334 nm and small peaks located at 400–500 nm. Three planar-diffused active layers were prepared by spin-coating the PCBM dissolved in three different solvents on the 100 nm underlying MEH-PPV layer, respectively. The concentration of PCBM solution is 5 mg/ml and the spin speed is 2000 rpm for 20 s and additionally dried at 1000 rpm for 1 min, which is in accordance with the processing condition for preparation of the active layer of the planar-diffused PV device. The absorption of the MEH-PPV film reduces noticeably after the PCBM solution is spin-coated on the top, which corresponds to the fact that a large amount of polymer molecules near the surface are eroded by the PCBM solution during spin-coating. The appearance of PCBM absorption peak at 334 nm in three absorption spectra of planar-diffused active layers indicates the existence of PCBM

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