

## Research Article

# Homogenized Poly(3-hexylthiophene)/Methanofullerene Film by Addition of End-Functionalized Compatibilizer and Its Application to Polymer Solar Cells

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A uniformed poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C<sub>61</sub>-butyric acid methyl ester (PCBM) blended film prepared by using spin-coating process is achieved by addition of 10% hydroxyl group end-functionalized P3HT (HOC-P3HT-COH) as a compatibilizer. Ratio of P3HT/PCBM on a spin-coated film has been improved from 1:1.22 on the edge and 1:0.85 in the center of substrate (substrate size, 2 cm × 3 cm) to 1:1.03 on the edge and 1:0.94 in the center for P3HT/HOC-P3HT-COH/PCBM film (1:0.1:1). Homogeneous and reproducible polymer solar cell with an average of 3.71% power conversion efficiency under AM1.5G irradiation is fabricated with 10% HOC-P3HT-COH in P3HT/PCBM layer.

## 1. Introduction

Power conversion efficiency ( $\eta$ ) of organic solar cells has increased from 0.04% of a bilayer device [1] to more than 8% of bulk heterojunction (BHJ) devices [2]. A major breakthrough is the introduction of BHJ morphology [3]. The BHJ morphology creates large interfacial area between the electron donors and electron acceptors that increases the efficiency of exciton dissociation and generates higher current density ( $J_{sc}$ ) of the BHJ devices. Evidently, morphology of the active layer plays a very important role in the performance optimization of polymer solar cells [4]. Among the electron donors and the electron acceptors, P3HT and PCBM have been widely used as model materials in the active layer for polymer solar cells. Researchers [5–12] find the performance of P3HT/PCBM based polymer solar cells highly relied on the ratio of P3HT/PCBM. Domain size of PCBM increases significantly after thermal annealing as the content of PCBM increases in the P3HT/PCBM film. This will lead to decreased interfacial area between P3HT and PCBM and will end up with lower  $J_{sc}$ . Additionally, hole mobility and electron mobility also vary with P3HT/PCBM ratio. A balanced hole and electron mobility in P3HT/PCBM blend occurs as the P3HT/PCBM ratio is between 1:0.8 and 1:1 [6, 7]. Crystalline

structure of P3HT/PCBM film characterized by scattering techniques [8–10] also reveals a high dependency on the P3HT/PCBM ratio. Generally high performance solar cells are fabricated with P3HT/PCBM ratios between 1:0.8 and 1:1 [11, 12].

Based on the above information, a uniformed distribution of P3HT/PCBM is crucial if a reproducible large area photovoltaic module is going to be processed. It comes to our attention to examine the distribution of P3HT/PCBM ratio across a glass substrate (2 cm × 3 cm) by using spin-coating process, which is the most commonly used technique in laboratory. In this study, P3HT/PCBM ratio across a 2 cm × 3 cm glass substrate is determined by using a UV-Vis spectroscopy technique. Effect of P3HT/PCBM ratio on the annealed P3HT/PCBM film is characterized by optical microscopy (OM). A synthesized hydroxyl group end-functionalized P3HT (HOC-P3HT-COH) is added to the P3HT/PCBM film to control the P3HT/PCBM ratio across the glass substrate.

## 2. Materials and Methods

2.1. Materials. P3HT ( $\overline{M}_n = 25,000$ ) and PCBM were purchased from Aldrich Corporation and were used without

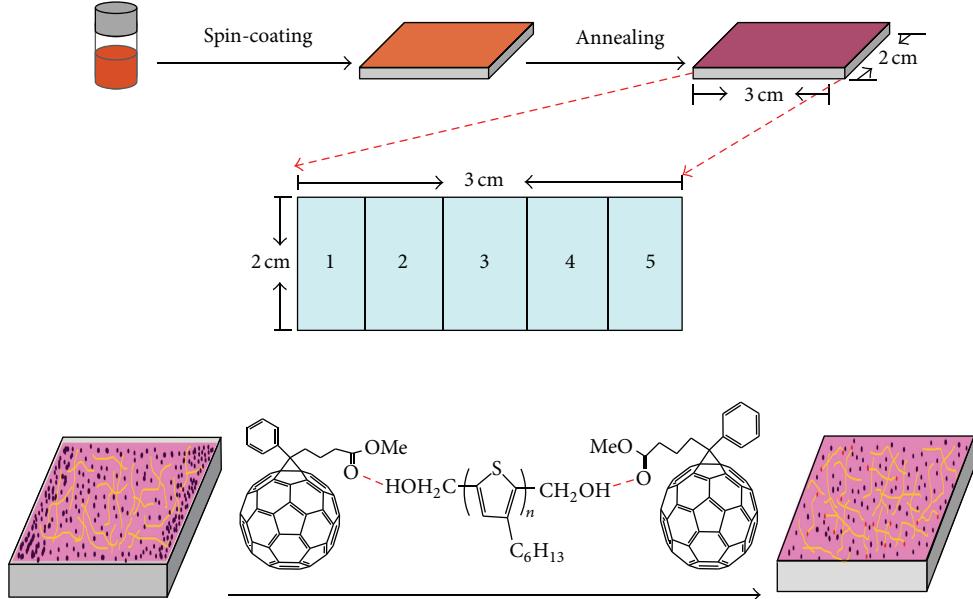


FIGURE 1: Illustration of sample preparation and identification of spin-coated P3HT/PCBM (1:1) and P3HT/HOC-P3HT-COH/PCBM (1:0.1:1) films from 1% dichlorobenzene solutions.

further purification. HOC-P3HT-COH was synthesized following a reported method [13]. The synthesized HOC-P3HT-COH has a  $M_n$  of 28,700, and a polydispersity index (PDI) of 1.89. NMR analysis [14] showed the regioregularity of the HOC-P3HT-COH was 95% and that for the purchased P3HT was 93%.

**2.2. Sample Preparation.** 1% P3HT/PCBM (1:1) and 1% P3HT/HOC-P3HT-COH/PCBM (1:0.1:1) in o-dichlorobenzene (ODCB, spectroscopy grade, purchased from Aldrich) were spin-coated on 2 cm × 3 cm glass substrates (500 rpm 3 seconds and 1000 rpm 10 seconds). The spin-coated films were annealed at 150°C for 30 minutes for UV-Vis spectroscopy and OM measurements.

**2.3. Device Fabrication.** ITO substrate (sheet resistance =  $12 \Omega/\square$ ) was cleaned in an ultrasonic machine following by treatment with O<sub>2</sub> plasma. A 30 nm PEDOT:PSS (Baytron PH from HC Starck) layer was coated on the ITO substrate and dried at 140°C for 10 min under N<sub>2</sub> atmosphere. A 100 nm P3HT/PCBM (1:1) or P3HT/HOC-P3HT-COH/PCBM film (1:0.1:1) was spin-coated and annealed at 150°C for 10 min. Ca (10 nm)/Al (100 nm) film was thermally evaporated as cathode at  $1.0 \times 10^{-6}$  torrs.

**2.4. Characterization.** The molecular weight of P3HT and HOC-P3HT-COH was measured by a Viscotek DM400/LR40 Gel Permeation Chromatography (GPC) using standard polystyrene as reference. Regioregularity was determined by a Bruker AV-300 Nuclear Magnetic Resonance. UV-Visible absorption measurements were carried out with a Shimadzu UV-2101C Spectrometer. The

morphology of the P3HT/PCBM and P3HT/PCBM/HOC-P3HT-COH films was studied using an Olympus Optical Microscopy (OM model: BH2). *I-V* parameters of the fabricated devices were measured by an Oriel Class A Solar Simulator 91160A under AM1.5G irradiation ( $100 \text{ mW/cm}^2$ ) calibrated by a NREL certified reference solar cell.

### 3. Results and Discussion

Spin-coating process [15] is a widely used technique in the research of polymer solar cells in laboratory. A blended photoactive layer such as P3HT/PCBM is spin-coated on a PEDOT:PSS precoated ITO substrate. This film is subject to different processes such as thermal annealing [16–18] and solvent annealing [19] before deposition of cathode materials. A polymer solar cell is ready for testing after these processes. Many factors such as composition of P3HT/PCBM [6, 7, 11, 12], annealing conditions [16–18], and solvent selection [20] affect the final performance of polymer solar cells. Composition of the P3HT/PCBM active layer is one of the key factors that are highly correlated with the device performance. It is shown that performance of an optimized P3HT/PCBM based polymer solar cell has a P3HT/PCBM ratio between 1:0.8 and 1:1.0 [6, 7, 11, 12] and the performance deteriorates significantly outside this range. It becomes very important if a consistent P3HT/PCBM ratio across substrate can be achieved for a large size polymer solar cell. To examine this issue, a P3HT/PCBM (1:1) solution in dichlorobenzene is spin-coated on a 2 cm × 3 cm substrate for evaluation. This substrate is divided into five regions symmetrically (Figure 1). Thicknesses of regions 1, 2, and 3 are 120 nm, 109 nm, and 95 nm, respectively, for P3HT/PCBM film. Thicknesses of regions 1, 2, and 3 for the P3HT/PCBM/10% HOC-P3HT-COH film are 111 nm, 113 nm, and 112 nm, respectively.

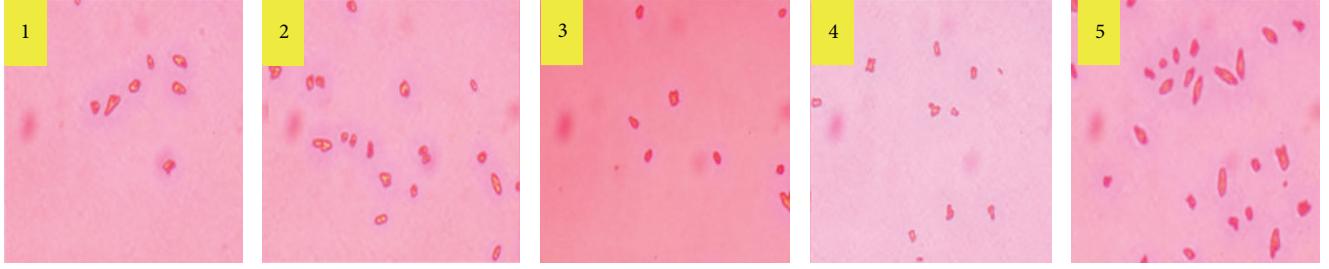


FIGURE 2: Optical microscopy photos ( $\times 100$ ) of the distribution of aggregated PCBM domains in annealed ( $150^\circ\text{C}$ , 30 min) P3HT/PCBM (1:1) film across a  $2\text{ cm} \times 3\text{ cm}$  glass substrate (edge: regions 1 and 5; center: region 3).

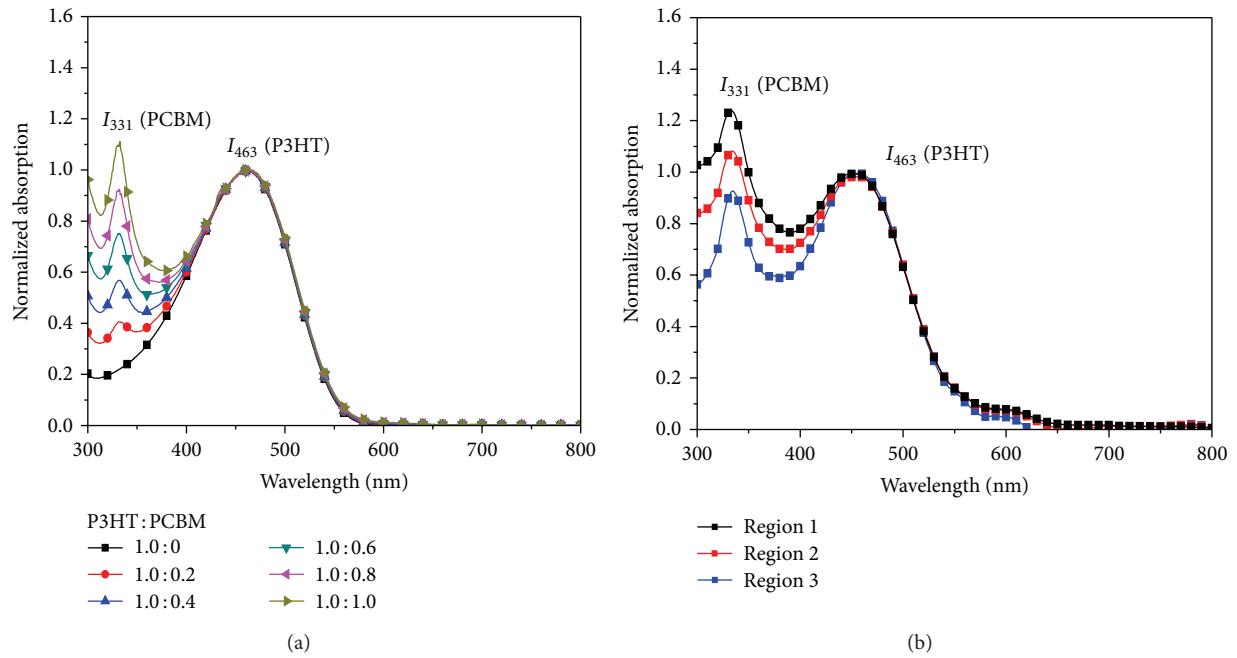


FIGURE 3: UV-Vis spectra of (a) P3HT/PCBM (1: $X$ ) (calibration curve) and (b) P3HT/PCBM (1:1) absorption in regions 1, 2, and 3.

Compositions of P3HT/PCBM between center and edge are characterized.

As reported in literatures, PCBM tends to aggregate and form large domain size in a P3HT/PCBM film upon thermal treatment [16–18]. Therefore, it will be possible to evaluate if the distribution of PCBM/P3HT is uniformed by using an optical microscope [21]. As shown in Figure 2, density of aggregated PCBM domains increases gradually from center of the substrate (region 3) to the edge of the substrate (regions 1 and 5). This indicates a higher PCBM concentration on the edge than that in the center after the spin-coating process. To further find out the actual P3HT/PCBM ratios in these regions, a UV-Vis calibration curve is constructed (Figure 3(a)). P3HT/PCBM in different regions (1, 2, and 3) is dissolved in dichlorobenzene and UV-Vis spectra are measured (assuming the coating is symmetry during the spin-coating process [15]). As shown in Figure 3(b), the relative intensity of  $I_{331}$  (absorption of PCBM and P3HT) to  $I_{463}$  (absorption of P3HT) increases from region 3 (center) to region 1 (edge), indicating higher PCBM concentration on

TABLE 1: List of ratio of  $I_{\text{PCBM}}$  to  $I_{\text{P3HT}}$  and actual P3HT/PCBM compositions (1:1, theoretically, without compatibilizer) in regions 1, 2, and 3 of the substrate.

	Region 1 (edge)	Region 2	Region 3 (center)
$I_{\text{PCBM}}/I_{\text{P3HT}}$	1.07	0.91	0.75
P3HT : PCBM	1:1.22	1:1.04	1:0.85

the edge (region 1). The relative intensity of  $I_{\text{PCBM}}$  to  $I_{\text{P3HT}}$  in different regions is calculated by

$$\frac{I_{\text{PCBM}}}{I_{\text{P3HT}}} = \left[ \frac{(I_{331} - 0.2191)}{I_{463}} \right], \quad (1)$$

where 0.2191 is the absorption intensity of P3HT at 331 nm. The corresponding P3HT/PCBM ratios obtained by using calibration curve are listed in Table 1. The 1% P3HT/PCBM solution in dichlorobenzene is 1:1. Ideally, the ratio of P3HT/PCBM will be 1:1 across the substrate. The actual P3HT/PCBM ratio in the center is 1:0.85 and that on the edge

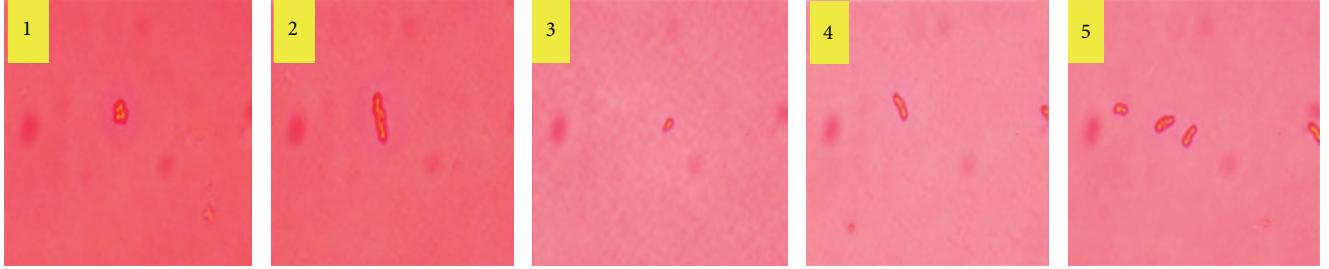


FIGURE 4: Optical microscopy photos ( $\times 100$ ) of the distribution of aggregated PCBM domains in annealed ( $150^{\circ}\text{C}$ , 30 min) P3HT/HOC-P3HT-COH/PCBM (1:0.1:1) film across a  $2\text{ cm} \times 3\text{ cm}$  glass substrate (edge: regions 1 and 5; center: region 3).

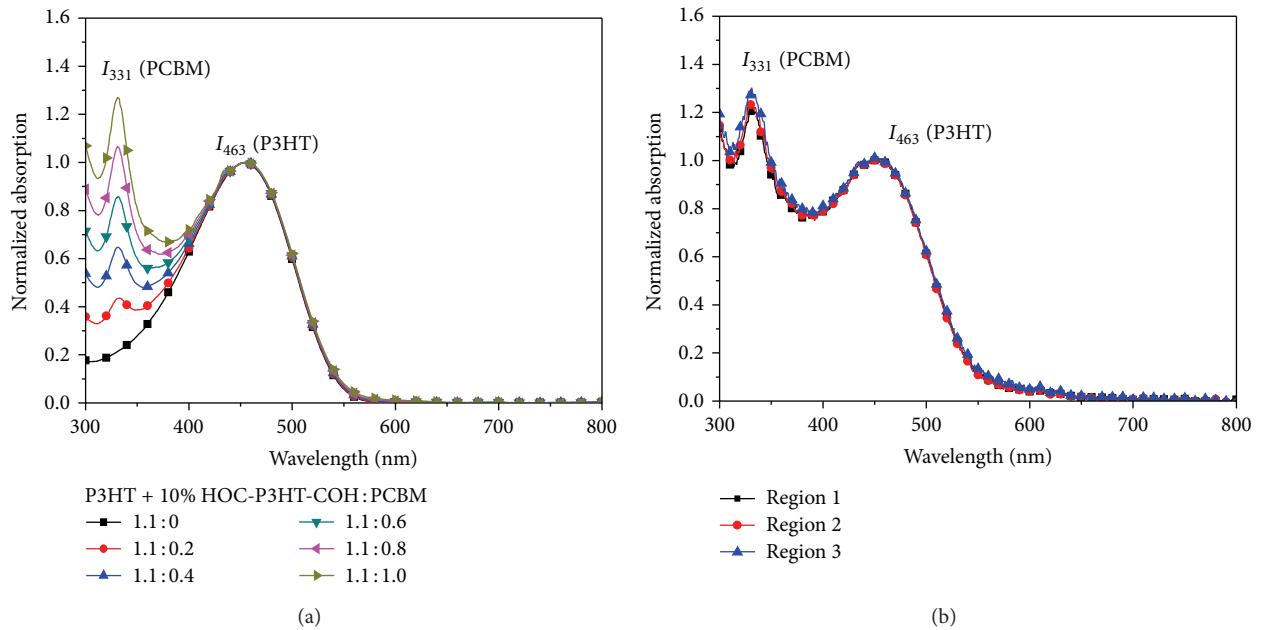


FIGURE 5: UV-Vis spectra of (a) P3HT/HOC-P3HT-COH/PCBM (1:0.1: X) (calibration curve) and (b) P3HT/HOC-P3HT-COH/PCBM absorption in regions 1, 2, and 3.

is 1:1.22. Parts of the PCBM molecules are moved further outside the center region during the spin-coating process. P3HT and PCBM are homogeneously dissolved in dichlorobenzene and they are distributed independently from each other in the solution. However, P3HT is a polymeric material with many entanglements between molecules and PCBM is a small molecule with no such entanglements in solution. Upon spin-coating from a common solvent-dichlorobenzene, P3HT and PCBM bear with different extent of centrifugal forces [15] and exhibit different coating behavior. Parts of the PCBM are taken away from the center region and are moved to the outer region. The difference in coating behavior of P3HT and PCBM ends up with a composition variation in P3HT/PCBM ratio across the substrate.

Hydroxyl group end-functionalized P3HT (HOC-P3HT-COH) has been proved as an effective compatibilizer in the stabilization of P3HT/PCBM blend [13]. H-bonds between the hydroxyl group of HOC-P3HT-COH and ester group of PCBM stabilize the morphology of P3HT/PCBM blend. As HOC-P3HT-COH is added to the P3HT/PCBM solution,

P3HT and PCBM molecules are physically linked through the H-bond formation. P3HT/HOC-P3HT-COH/PCBM can be treated as a physically bonded molecule during the spin-coating process. As a 1% P3HT/HOC-P3HT-COH/PCBM solution is spin-coated onto a substrate, P3HT, HOC-P3HT-COH, and PCBM do not separate. They form a uniformed coating across the substrate. As shown in Figure 4, distribution of aggregated PCBM domains is uniformed across the substrate that indicates a consistent P3HT/PCBM composition across the substrate. It is very important that HOC-P3HT-COH not only stabilizes the P3HT/PCBM morphology, but also homogeneously disperses the PCBM in P3HT. A UV-Vis absorption calibration curve of P3HT/HOC-P3HT-COH/PCBM (1:0.1: X) is constructed (Figure 5(a)) to determine the P3HT/PCBM ratio in regions 1, 2, and 3. As shown in Figure 5(b), the relative intensity ratios of  $I_{331}$  to  $I_{463}$  in the region 1, region 2, and region 3 are very similar revealing the information that the ratios of P3HT/PCBM are consistent in these three regions. The actual P3HT/PCBM ratios determined by calibrated UV-Vis absorption curve are

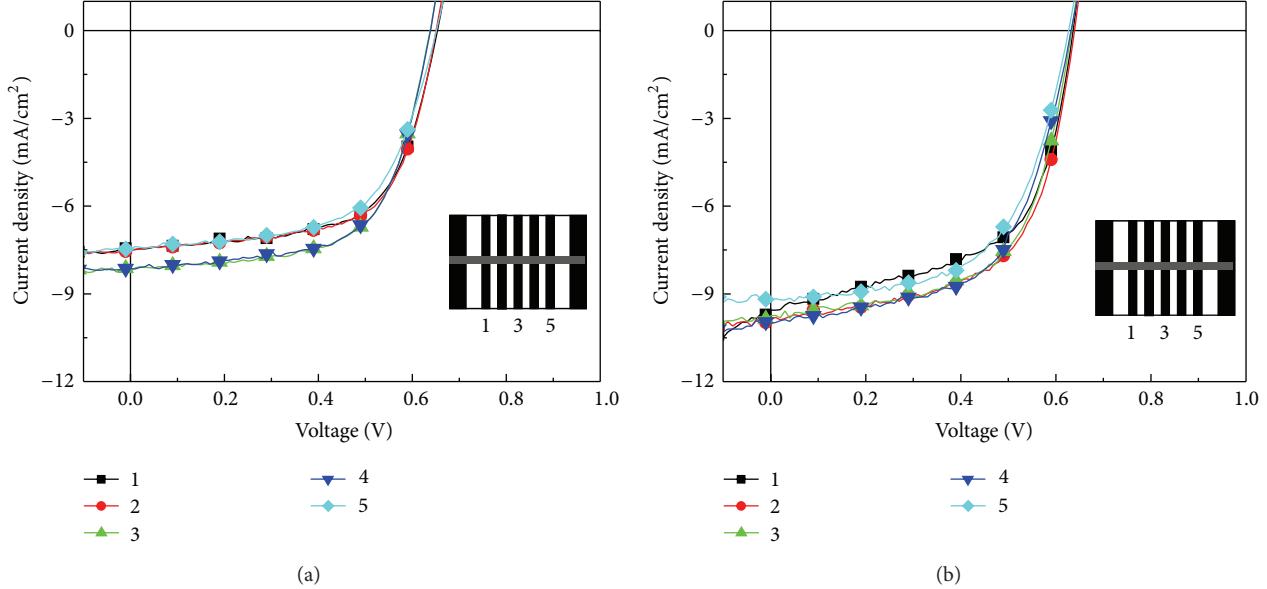


FIGURE 6:  $J$ - $V$  curves of (a) P3HT/PCBM (1:1) and (b) P3HT/HOC-P3HT-COH/PCBM (1:0.1:1) based polymer solar cells with a device structure of ITO/PEDOT:PSS (40 nm)/active layer (100 nm)/Ca/Al in different regions (1–5) of the same substrate.

TABLE 2: List of ratio of  $I_{\text{PCBM}}$  to  $I_{\text{P3HT}}$  and actual PCBM/P3HT\* compositions (1:1.1, theoretically, with compatibilizer) in regions 1, 2, and 3 of the substrate.

	Region 1 (edge)	Region 2	Region 3 (center)
$I_{\text{PCBM}}/I_{\text{P3HT}}$	1.00	1.01	1.02
P3HT*: PCBM	1:0.94	1:0.95	1:1.03
P3HT*: P3HT + 10 wt% HOC-P3HT-COH.			

1:0.94 (region 1), 1:0.95 (region 2), and 1:1.03 (region 3) (Table 2). These values are close to the P3HT/PCBM ratio (1:0.91) in solution if the absorption of 10% HOC-P3HT-COH is taken into consideration.

To examine how the P3HT/PCBM ratio affects performance of polymer solar cells in the different regions of the same substrate, an ITO substrate is patterned in the form as illustrated in inlet of Figure 6. Five devices are symmetrically designed on a 2 cm  $\times$  3 cm substrate. As shown in Figure 6,  $J_{\text{sc}}$  of devices with 10% HOC-P3HT-COH in the active layer are all higher than those without HOC-P3HT-COH, respectively. It has been shown [13] that reduction of PCBM aggregation is one of the factors leading to higher  $J_{\text{sc}}$  value in a polymer solar cell with HOC-P3HT-COH in the active layer. Moreover, thermal stability of the P3HT/PCBM layer is also improved with addition of HOC-P3HT-COH in the P3HT/PCBM layer.

In this study, consistency of device performance in different regions on a 2 cm  $\times$  3 cm substrate has been improved as shown in Tables 3 and 4. Standard deviation of  $J_{\text{sc}}$ ,  $V_{\text{oc}}$ , FF, and  $\eta$  for the devices (devices 1–5 as illustrated in Figure 6) with HOC-P3HT-COH in the active layer is all lower than that without HOC-P3HT-COH in the active layer, respectively. These results reveal that incorporation of HOC-P3HT-COH as compatibilizer improves performance deviation of devices in a 2 cm  $\times$  3 cm substrate, which is

TABLE 3: Solar cells performance of P3HT/PCBM (1:1) based polymer solar cells in different regions of a 2 cm  $\times$  3 cm substrate. The active layer is thermally treated at 150°C for 10 minutes.

(a) P3HT/PCBM	$J_{\text{sc}}$ (mA/cm²)	$V_{\text{oc}}$ (V)	FF	$\eta$ (%)
Region 1	7.49	0.66	0.62	3.07
Region 2	7.52	0.66	0.63	3.13
Region 3	8.22	0.64	0.63	3.29
Region 4	8.13	0.64	0.63	3.28
Region 5	7.47	0.65	0.6	2.93
Average value	7.77	0.65	0.62	3.14
Standard deviation	0.375	0.010	0.013	0.151

TABLE 4: Solar cells performance of P3HT/HOC-P3HT-COH/PCBM (1:0.1:1) based polymer solar cells in different regions of a 2 cm  $\times$  3 cm substrate. The active layer is thermally treated at 150°C for 10 minutes.

P3HT/PCBM/10% HOC-P3HT-COH	$J_{\text{sc}}$ (mA/cm²)	$V_{\text{oc}}$ (V)	FF	$\eta$ (%)
Region 1	9.56	0.64	0.6	3.66
Region 2	9.89	0.64	0.6	3.79
Region 3	9.75	0.64	0.61	3.77
Region 4	10	0.64	0.59	3.77
Region 5	9.27	0.63	0.61	3.56
Average value	9.70	0.64	0.60	3.71
Standard deviation	0.289	0.005	0.008	0.098

highly related to P3HT/PCBM ratio across the substrate. As mentioned previously, the P3HT/PCBM ratio has a high deviation from the center of the substrate (1:0.85) to the edge of the substrate (1:1.22) without HOC-P3HT-COH in the

film (Table 1). This deviation is diminished by addition of HOC-P3HT-COH to the P3HT/PCBM layer (Table 2) that matches the results from the device performance as shown in Tables 3 and 4. A high performance polymer solar cell with an average of 3.71% power conversion efficiency and low standard deviation under AM1.5G irradiation is prepared with 10% HOC-P3HT-COH in P3HT/PCBM (1:1).

## 4. Conclusions

Compositions of P3HT/PCBM in a 2 cm × 3 cm have been found to be inconsistent from the center part of substrate to the edge of the substrate. Concentration of PCBM tends to be higher on the edge of substrate. This can be solved by adding a hydroxyl group end-functionalized P3HT as a compatibilizer to the P3HT and PCBM. Composition variation across the substrate becomes consistent across the substrate. This will possibly improve the performance of a large area polymer solar cell because of homogeneous distribution of P3HT/PCBM on the substrate.

## Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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