

Research Article

Characterization of Organic Thin Film Solar Cells of PCDTBT : PC₇₁BM Prepared by Different Mixing Ratio and Effect of Hole Transport Layer

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The organic thin film solar cells (OTFSCs) have been successfully fabricated using PCDTBT : PC₇₁BM with different mixing ratios (1 : 1 to 1 : 8) and the influence of hole transport layer thickness (PEDOT : PSS). The active layers with different mixing ratios of PCDTBT : PC₇₁BM have been fabricated using *o*-dichlorobenzene (*o*-DCB). The surface morphology of the active layers and PEDOT : PSS layer with different thicknesses were characterized by AFM analysis. Here, we report that the OTFSCs with high performance have been optimized with 1 : 4 ratios of PCDTBT : PC₇₁BM. The power conversion efficiency (PCE = 5.17%) of the solar cells was significantly improved by changing thickness of PEDOT : PSS layer. The thickness of the PEDOT : PSS layer was found to be of significant importance; the thickness of the PEDOT : PSS layer at 45 nm (higher spin speed 5000 rpm) shows higher short circuit current density (J_{sc}) and lower series resistance (R_s) and higher PCE.

1. Introduction

The present world population exceeds seven billion and is expected to increase beyond nine billion per year by 2050. As the population growth, energy consumption was double in the year 2010 and it would be increased in the future. The energy demand is big issue, which would be widening the gap between rich and poor countries [1, 2]. In order to reduce energy problem, solar energy is the best alternative of fossil fuel. Total amount of solar energy might be approximately estimated about 1000 TW. Considering the current amount of total energy consumption (10 TW) for mankind, solar energy could be regarded as an infinite source of energy. Besides, solar energy generation does not cause CO₂ emission; this would be to reduce environment problem. However, the silicon based solar cells fabrication has high cost, because

of that supply of resource materials could be limited to the material processing.

Therefore, the researchers have extensively used novel organic molecules or materials to overcome the above problems. Organic solar cell has unique advantages such as lightweight, flexibility, cost-effectiveness due to solution processes, and adaptability for mobile applications [3–7]. Also, in the past, significant advances have been made in the OSC using the state-of-art organic bulk heterojunction (BHJ) polymer solar cells [3–9]. Hence, the BHJ structures composed of electron-donating polymers and electron-withdrawing fullerenes are used as active layers and reached the highest PCE over 7–9% [4–7] for single junction and over 9–12% PCE for tandem cell [8, 9], which is still inferior to those of poly- and amorphous-silicon-based solar cells (efficiencies of 15–20%). Moreover, the development of

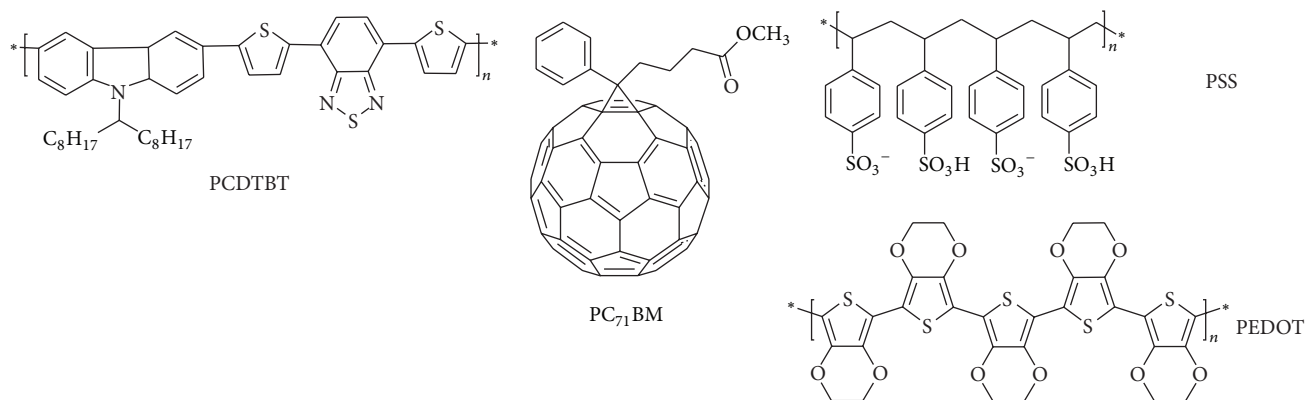


FIGURE 1: Molecular structure of PCDTBT, PC₇₁BM, and PEDOT : PSS.

new material has been focusing on high efficiency and stability of the device lifetime. Currently, the poly[[9-(1-octylonyl)-9H-carbazole-2,7-diyl]-2,5-thiophenediyl-2,1,3-benzothiadiazole-4,7-diyl-2,5-thiophenediyl] (PCDTBT) is used as a p-type organic semiconductor in the organic thin film solar cell (OTFSC) due to higher efficiency with good solubility [10–14]. A fullerene derivative of [6,6]-phenyl C₇₁ butyric acid methyl ester (PC₇₁BM) was widely used as n-type organic semiconductor due to the superior electron transporting properties [8–14]. For the optimization, the mixing ratio of PCDTBT:PC₇₁BM has been accomplished with the superior performance of OTFSC. In order to improve the performance of OTFSC, a poly(3,4-ethylenedioxythiophene) : poly(styrenesulfonate) [PEDOT:PSS] was used as a hole transport layer. The OTFSC performance was sensitive to PEDOT:PSS film thickness and surface morphology [8, 9, 15–23].

In this present investigation, we demonstrated that the PCDTBT:PC₇₁BM with different mixing ratios (1:1 to 1:8) and changing thickness of the hole transporting layers (PEDOT:PSS) for high performance OTFSCs have been studied. The PCDTBT:PC₇₁BM active layers with four different ratios (1:1, 1:2, 1:7, and 1:8) have been prepared using o-dichlorobenzene as a solvent, which was compared with mixing ratio of 1:3, 1:4, 1:5, and 1:6 to our previous reports [21]. The surface morphologies of different blending ratios of active layers and different thickness PEDOT:PSS films were observed by AFM analysis. The electrical properties of different mixing ratio of active layers and PEDOT:PSS layer thickness on the performance of OTFSC were examined by measuring the open circuit current voltage (V_{oc}), J_{sc} , R_s , and PCE using current density-voltage (J - V) analysis. The open circuit current voltage (V_{oc}), short circuit current density (J_{sc}), series resistance (R_s), and power conversion efficiency (PCE) have been carried out for the different thickness of PEDOT:PSS layers.

2. Experimental Work

2.1. Materials. PCDTBT and PC₇₁BM were purchased from Ossila, England, and used as electron donor, electron acceptor materials, respectively. PEDOT:PSS (Clevios PHI000)

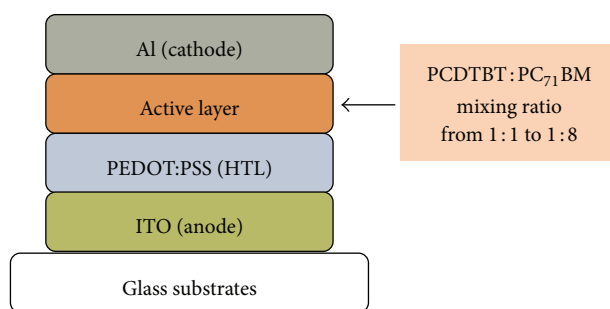


FIGURE 2: Schematic diagram of the OTFSCs.

was purchased from Heraeus, Germany, and used as a hole transport layer between ITO and active layer (PCDTBT:PC₇₁BM). Chlorobenzene, o-dichlorobenzene, ethanol, and acetone were purchased from Aldrich and used without further purification. Figure 1 shows the molecular structures of the PCDTBT, PC₇₁BM, and PEDOT:PSS.

2.2. Fabrication of Organic Thin Film Solar Cells. Indium doped tin oxide (ITO) conducting glass substrate (Aldrich; 8–12 Ω/sq) was used to fabricate the OTFSC device with a structure of ITO/PEDOT:PSS/PCDTBT:PC₇₁BM/Al. Figure 2 shows schematic diagram of the OTFSC device. ITO conducting glass substrate was ultrasonicated thoroughly in acetone, ethanol, and distilled water for 10 min for each step to remove the organic contaminations. The cleaned ITO conducting glass substrate was then exposed to UV Ozone for 10 min. PEDOT:PSS was coated at spin speed of 3000 rpm for 30 sec followed by heat treated at 150°C in atmospheric air for 10 min. Four different active layers were prepared by changing the mixing ratio of PCDTBT and PC₇₁BM (1:1, 1:2, 1:7, and 1:8) in o-DCB solvent. Al was deposited by vacuum evaporation technique on the active layer and used as cathode electrode in OTFSCs. The effect of the hole transporting layer thickness (PEDOT:PSS) on photovoltaic performance was studied by different thickness of PEDOT:PSS layers. The different thickness of PEDOT:PSS layers was achieved by changing the spin speed

TABLE 1: Surface roughness parameters of the PCDTBT:PC₇₁BM prepared with different mixing ratios of 1:1~1:8 in o-DCB and CB.

PCDTBT:PC ₇₁ BM blend ratio	P-V (nm)	RMS (nm)
1:1 (oDCB)	19.64	1.0450
1:2 (oDCB)	8.005	0.6804
1:3 (oDCB)	6.513	0.5983*
1:4 (oDCB)	9.845	0.7437*
1:5 (oDCB)	6.513	0.5858*
1:6 (oDCB)	8.403	0.5957*
1:7 (oDCB)	10.94	0.7530
1:8 (oDCB)	9.944	0.7984
1:4 (CB)	15.16	1.95*

*Reference [21].

from 2000 to 5000 rpm and the Al thin films were deposited with 5 nm (effective area of the device: 25 mm²).

2.3. Characterization Techniques. The thickness of hole transport layer (PEDOT:PSS) was measured using a Dektak II profilometer. Surface morphologies of the different blend of active layer and PEDOT:PSS layer are observed by atomic force microscopy (AFM: Seiko Instruments SPA400-SPI4000). All AFM images were taken in dynamic force mode at optimal force. Absorption characteristics of eight different active layers of PCDTBT:PC₇₁BM, pristine PCDTBT, and PC₇₁BM are measured using UV-visible spectroscopy (Shimazu, UV2450). The photovoltaic current density-voltage (*J-V*) characteristic of PCDTBT is as follows: PC₇₁BM based OTFSCs were performed in standard Newport 66902 150 W solar simulator with an Oriel Cornerstone 130 monochromator and a Xe lamp equipped with an air mass of 1.5 G (AM 1.5 G) filter as the white-light source (100 mW/cm²). An Advantest-R-6441 A.C. source meter was used for measuring the *J-V* parameter such as V_{oc} , J_{sc} , fill factor (FF), R_s , and PCE. All measurements were carried out in air without any encapsulation.

3. Results and Discussion

3.1. Morphological Studies. Surface morphology and the layer thickness play vital role on the photovoltaic characteristics of OTFSC. The morphologies of active layer with different mixing ratios (1:1, 1:2, 1:3, 1:4, 1:5, 1:6, 1:7, and 1:8) of PCDTBT:PC₇₁BM have been studied by AFM analysis. Figure S1 (in Supplementary Material available online at <http://dx.doi.org/10.1155/2015/687678>) shows the AFM image of the active layer of PCDTBT:PC₇₁BM with ratio of 1:1 to 1:8 in o-DCB and 1:4 ratio prepared using CB. Surface roughness parameters such as peak-to-valley (P-V) and root-mean-square (RMS) are summarized in Table 1. AFM images and surface parameter do not disclose any noticeable change in the surface of the active layer (PCDTBT:PC₇₁BM) with different blend ratio in o-DCB compared to CB solvent. Table 1

TABLE 2: Surface roughness parameters obtained from AFM analysis for different thickness of PEDOT:PSS layer.

PEDOT:PSS layer (thickness)	P-V (nm)	RMS (nm)
5000 rpm (~45 nm)	15	1.34
3000 rpm (~62 nm)	21	1.45
2000 rpm (~92 nm)	35	1.57

shows the surface roughness parameters with smaller P-V of PCDTBT:PC₇₁BM surface which suggests smoother surface with nanomorphology (Figure S1) [21]. The 1:1 ratio of PCDTBT:PC₇₁BM shows slightly higher P-V and RMS values when compared with other ratios. It revealed that the mixing ratio of 1:1 with PC₇₁BM molecules is relatively weaker with diffusivity and lower dispersibility in the bulk heterojunction of PCDTBT:PC₇₁BM. However, the other mixing ratios (1:2, 1:3, 1:4, 1:5, 1:6, 1:7, and 1:8) exhibit lower P-V and RMS roughness values. It implies that PC₇₁BM could play major role in the surface roughness to resulting bulk heterojunction of PCDTBT:PC₇₁BM. In addition, all the active layers of PCDTBT:PC₇₁BM prepared in o-DCB (Figure S1, RMS = 1.04~0.6) do not aggregate with PC₇₁BM compared with the surface of PCDTBT:PC₇₁BM layer prepared with CB (Figure S1, RMS = 1.95) due to higher solubility of PC₇₁BM in o-DCB compared with CB [11–14, 21]. In addition, o-DCB has higher boiling point at 180°C and evaporation rate of o-DCB is slow during drying processes. Slow evaporation prevented the growth of the rough structure during the drying processes, resulting in smoother surface with nanomorphology, which could improve the device performance [21, 24, 25].

The effect of the PEDOT:PSS layer thickness on the surface morphology was studied using AFM analysis. Figure 3 shows the AFM image of PEDOT:PSS layer with different thickness. The thickness of PEDOT:PSS layer is controlled by different spin speed. Surface roughness parameters obtained from AFM analysis for different thickness of PEDOT:PSS layer are given in Table 2. AFM image of PEDOT:PSS layer at different thickness is shown in Figure 3.

Surface roughness parameters show that there is increase in the RMS roughness of 1.34, 1.45, and 1.57 nm and P-V of 15, 21, and 35 nm when increasing the thickness 45, 62, and 90 nm, respectively. The AFM result shows that the PEDOT:PSS layer thickness about 45 nm is smoother surface due to higher spinning speed (5000 rpm) and evaporation rate of the solvent is higher. The smoother surface of PEDOT:PSS layer enables more uniform interfacial contact between active layer and PEDOT:PSS layer, which might increase the charge transport and extraction results improvement in the device performance [16, 20, 22]. The AFM result shows that the RMS value increases when the thickness of PEDOT:PSS layer was increased around 62 nm, which implies that the uniformity of the PEDOT:PSS layer is less. In particular, a ring of solution is formed at the edge of the substrate due to incomplete centrifugation of the applied solution and there is some variation in film thickness across the substrate [16–20], which would lead to changes in charge

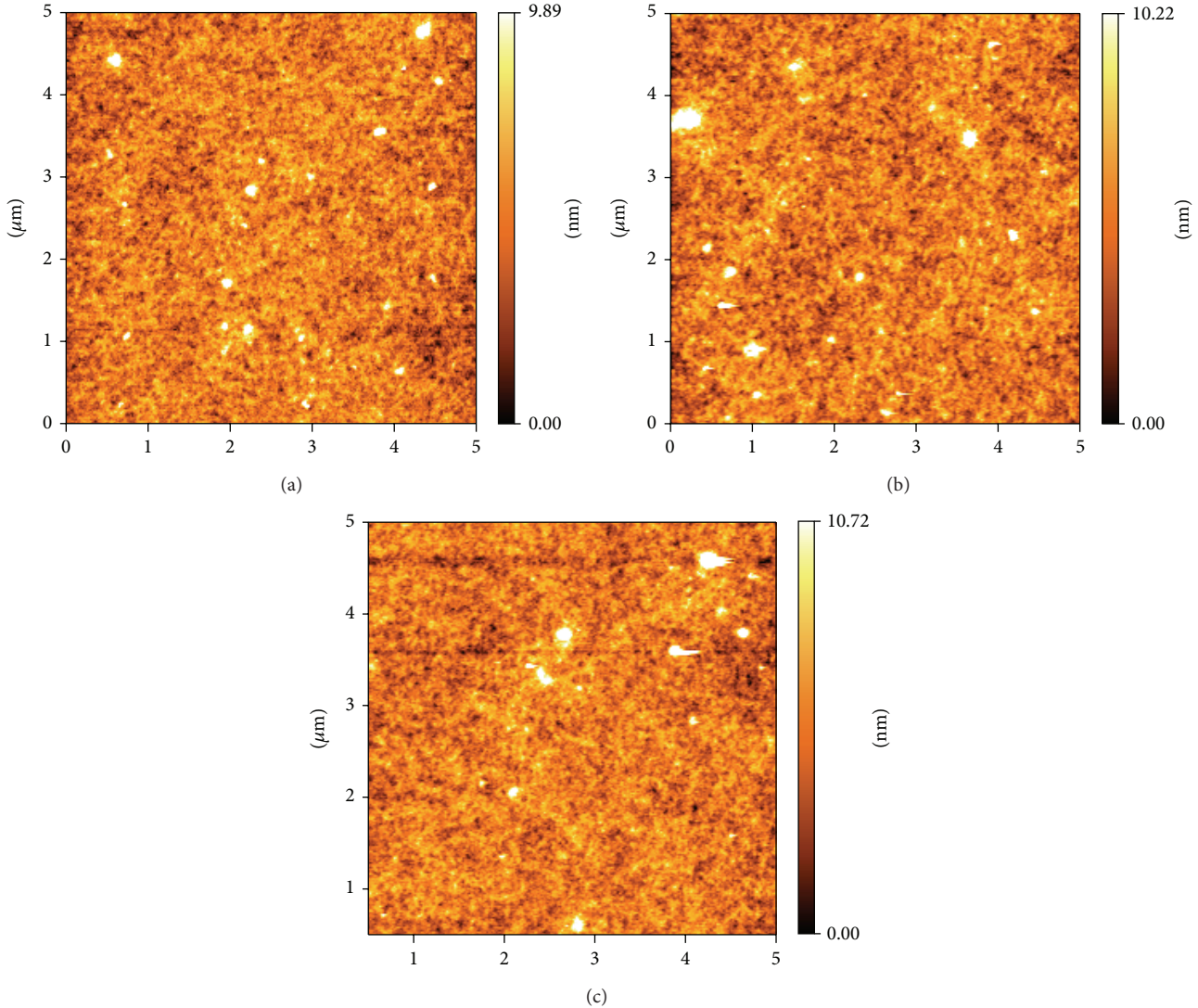


FIGURE 3: AFM image of PEDOT : PSS layer at different thickness: (a) 45, (b) 62, and (c) 90 nm.

carrier transport property [16, 18, 22]. Further increasing the thickness, the drying of the film becomes significantly slower and possible for different drying dynamics are responsible for a change in orientation of the PEDOT grains within the film and surface properties, which would lead to changes in charge transport property [16, 18–20, 22]. Thus it might be suggested that the surface roughness increases at higher thickness due to PEDOT : PSS aggregation [14, 16, 17, 20]; it might decrease the charge transport and extraction properties and also reduce the PCE [16, 18, 19, 22].

3.2. Photocurrent Density-Voltage (J - V) Characteristics. Characterization of OTFSCs of PCDTBT : PC₇₁BM prepared by different mixing ratio (1:1~1:8) and thickness of the hole transporting layer of PEDOT : PSS. The PCDTBT : PC₇₁BM based OTFSCs are prepared by different mixing ratios 1:1, 1:2, 1:7, and 1:8, which is compared with our earlier report (mixing ratios 1:3, 1:4, 1:5, and 1:6) [21]. Figure 4 shows

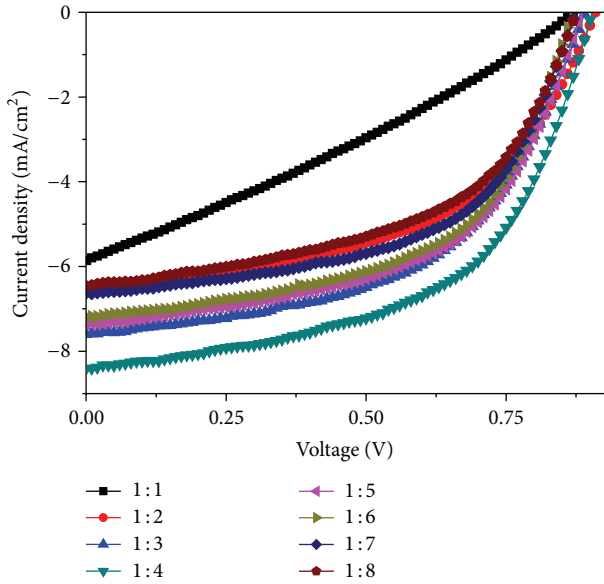
the J - V characteristics for the OTFSC of PCDTBT : PC₇₁BM prepared with different mixing ratios in o-DCB. Performance parameters such as J_{sc} , V_{oc} , FF, and PCE are obtained from the J - V measurement as given in Table 3.

The OTFSC device with 1:1 ratio of PCDTBT : PC₇₁BM reveals distinctively lower PCE = 1.53% compared with higher composition of PC₇₁BM of other devices. It can be assumed that a lower composition of PC₇₁BM would be relatively smaller number of electron-path, causing a lot of electron-hole recombination, which reduces the charge collection efficiency as well as increasing the series resistance ($R_s = 80 \Omega \text{ cm}^2$) [26, 27]. The higher $R_s = 80 \Omega \text{ cm}^2$ decreases the $J_{sc} = 5.86 \text{ mA/cm}^2$ as well as FF = 30%, resulting in lowest PCE = 1.53% for 1:1 ratio of PCDTBT : PC₇₁BM device. Whereas, 1:4 ratio of PCDTBT : PC₇₁BM the OTFSC device shows that there is an increase of $J_{sc} = 8.41 \text{ mA/cm}^2$ and a decrease of $R_s = 21 \Omega \text{ cm}^2$. As a result, a higher PCE = 4.15% with good FF = 55% was achieved [21, 23, 24]. It implies that

TABLE 3: J - V parameters of OTFSCs device with PCDTBT:PC₇₁BM prepared with different mixing ratios 1:1~1:8 in o-DCB.

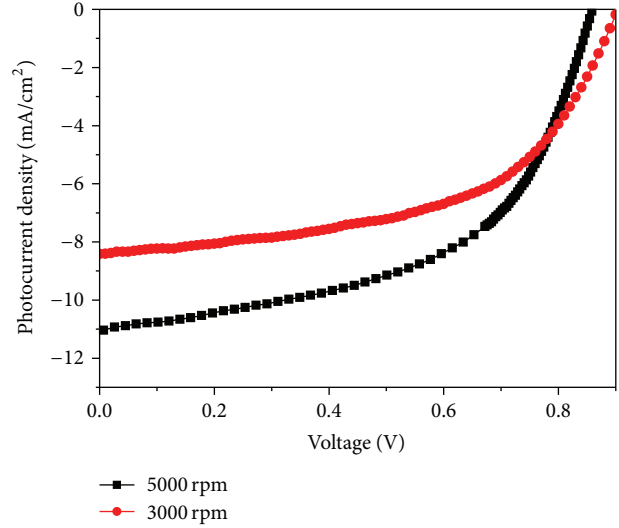
PCDTBT:PC ₇₁ BM mixing ratio	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	PCE (η) (%)
1:1	5.86	0.87	30	1.53
1:2	6.6	0.91	51	3.04
1:3	7.6	0.89	53	3.60*
1:4	8.41	0.9	55	4.15*
1:5	7.36	0.89	55	3.56*
1:6	7.19	0.87	55	3.45*
1:7	6.63	0.87	55	3.21
1:8	6.47	0.88	52	2.94

*Reference [21].

FIGURE 4: J - V characteristics of OTFSCs of PCDTBT:PC₇₁BM prepared with different mixing ratios 1:1~1:8 in o-DCB.

when the relative composition of PC₇₁BM ratio was increased which lead to increase in the number of electron-path and a decrease in the electron-hole recombination have been optimized in the case of 1:4 ratio of PCDTBT:PC₇₁BM. This reveals an increase in the charge carrier transport properties as well as the increase in J_{sc} and decrease in the R_s results the enhancement in the device performance. From this observation, when the relative composition of PC₇₁BM ratio was increased from 1:1 to 1:4, which lead to increases in the J_{sc} , FF as well as the PCE. This might be attributed to increasing electron-path; it prevents the recombination and enhances the charge collection results improving the device efficiency. Further, increasing the relative composition of PC₇₁BM ratio from 1:5 to 1:8 reveals a gradual decrease in J_{sc} as well as PCE for the resulting OTFSC devices [21, 23, 24, 26]. It seems that a decrease of relative composition of PCDTBT over PC₇₁BM would cause unbalanced hole-electron-path (or excessive hole-path) according to increase of relative composition of PC₇₁BM over PCDTBT [23, 24, 26, 27].

The effect of the hole transporting layer (PEDOT:PSS) thickness on photovoltaic performance was studied by

FIGURE 5: J - V characteristics of OTFSCs device of PCDTBT:PC₇₁BM ratios (1:4) prepared in o-DCB with different PEDOT:PSS layer thickness.

J - V measurement. The thickness of PEDOT:PSS layers was varied by changing the spin speed from 3000 rpm to 5000 rpm and active layer thickness kept constant with 1:4 ratio of PCDTBT:PC₇₁BM. The J - V parameter and device performance of PCDTBT:PC₇₁BM based OTFSC device with respect to PEDOT:PSS layer thickness are given in Table 4. Figure 5 shows the J - V characteristics of PCDTBT:PC₇₁BM based OTFSC device with different thicknesses of PEDOT:PSS layer.

The photovoltaic performance of the OTFSC device of PEDOT:PSS layers with a thickness of 45 nm shows significantly improved PCE = 5.15% when compared with higher thickness of PEDOT:PSS layer at 62 nm (PCE = 4.15%) obtained for lower spin speed with 3000 rpm as per our earlier report [21]. When the spin speed was increased about 5000 rpm, PEDOT:PSS layer thickness as well as surface roughness will decrease. The enhancement of the device performance might be due to decreasing the series resistance $R_s = 16 \Omega/\text{cm}^2$ resulting in enhanced J_{sc} of 11.01 mA/cm², FF 55%. The R_s is closely related with active layer morphology, intrinsic resistance, and thickness of the active layer.

TABLE 4: J - V parameter of OTFSCs device of PCDTBT : PC₇₁BM ratios (1 : 4) prepared in o-DCB with different thickness of PEDOT : PSS layer.

PCDTBT : PC ₇₁ BM (1 : 4) PEDOT : PSS (thickness)	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	R_s (Ω cm ²)	PCE (η) (%)
5000 rpm (~45 nm)	0.85	11.05	55	13	5.17
3000 rpm (~62 nm)	0.9	8.41	55	21	4.15*

*Reference [21].

The enhancement of the device performance (PCE = 5.15%) due to the change in the properties of the PEDOT : PSS layer thickness itself directly or indirectly influences of the PEDOT : PSS surface properties on the properties of the blend spin-coated on top [16, 17, 19, 20, 22]. This suggests that the thickness of PEDOT : PSS layer was decreased at 45 nm which leads to the decrease in the series resistance and the results will enhance the charge carrier transport properties and suppress the charge carrier recombination at the interface with an improvement in the device efficiency. This implies that the decreases in the series resistance as well as increase in the shunt resistance and FF might be depending on PEDOT : PSS layer thickness and influence of the PEDOT : PSS surface properties. The AFM results reveal that the surface of the PEDOT : PSS is smoother with the thickness of 45 nm, which enables more uniform interfacial contact between the active and PEDOT : PSS layer, which might reduce the local fluctuation and enhance the charge collection efficiency [16, 18, 19, 22, 28]. The PEDOT : PSS layer thickness has increased around 62 nm and the results slightly increase in $V_{oc} = 0.9$ V. However, there are increases in the $R_s = 21 \Omega$ cm² and decreases in the $J_{sc} = 8.41$ mA/cm², as a result of moderate performance of PCE = 4.15%. The decrease in R_s can be attributed to recombination loss. Further, increase in the PEDOT : PSS layer thickness about 92 nm will increase the $R_s = 29 \Omega$ cm² and decrease the device performance of PCE = 3.52 %. The device performance depends on PEDOT : PSS layer thickness and its surface. These results suggested that device performance significantly improved by changing thickness of PEDOT : PSS layer 45 nm at higher spin speed with 5000 rpm as well as decreasing the surface roughness and hence improvement in the OTFSCs performance. However, at higher thickness of PEDOT : PSS layer, the surface roughness will increase which results in a poor device performance due to PEDOT : PSS aggregation which leads to decrease in the charge transport and extraction [16–22].

3.3. Absorption Studies. The solar energy conversion efficiency mainly depends on its light absorption property of active layers. The steady state absorption spectral properties of the PCDTBT : PC₇₁BM active layer were investigated by UV-visible spectroscopy. Figure 6 shows that the absorbance spectra for the PCDTBT : PC₇₁BM layer prepared with different ratios of PCDTBT and PC₇₁BM in o-DCB and active layer show similar shape of light absorption. The spectra reveal that difference in ratio of PCDTBT and PC₇₁BM could hardly be contributed to formation of different morphology in the resulting bulk heterojunction of PCDTBT : PC₇₁BM.

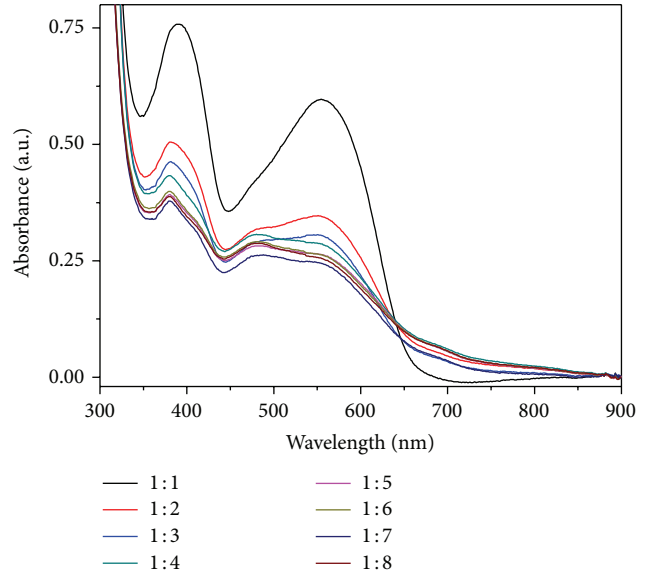


FIGURE 6: UV-visible absorption spectra for the active layer with different ratio of PCDTBT and PC₇₁BM.

The 1:1 ratio of PCDTBT : PC₇₁BM layer was higher than that of other ratios. This result indicates that light absorption property of PCDTBT : PC₇₁BM could be strongly dependent on relative quantity of PCDTBT [21, 24, 26].

To verify that supposition, absorption studies were carried out for a single layer of PCDTBT and single layer of PC₇₁BM. 18 mg of PCDTBT was dissolved in 1 mL of o-DCB and 18 mg of PC₇₁BM was dissolved in 1 mL of o-DCB. Figure 7 shows light-absorbance spectra of the resulting individual layer of PCDTBT and PC₇₁BM, respectively. Pristine PCDTBT layer shows strong light absorption peaks at wavelength of about 390 nm and 557 nm, while pristine PC₇₁BM shows weak absorption peaks at wavelength of about 377 nm and 552 nm. Pristine PCDTBT shows four times higher light absorption peak at 390 nm than that of PC₇₁BM at 377 nm, and about five times higher light absorption peak at 557 nm than that of PC₇₁BM at 552 nm. Even though the weight percentage of PCDTBT and PC₇₁BM is same, the PCDTBT layer reveals drastically higher light-absorbance than that of PC₇₁BM layer. Therefore, the difference in the absorption property according to ratio of PCDTBT and PC₇₁BM in o-DCB (Figure 7) is originated from the difference of quantity of PCDTBT. Absorption spectra for the PCDTBT : PC₇₁BM with mixing ratios of 1:1 show higher absorption than the other pristine PCDTBT and PC₇₁BM layers. The absorption

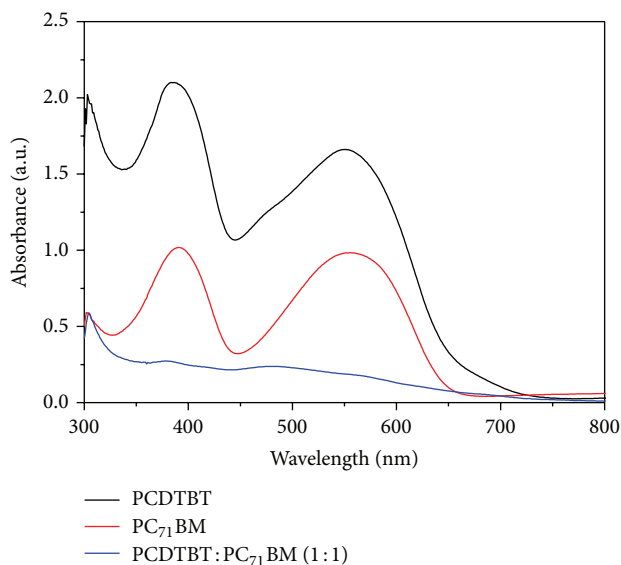


FIGURE 7: UV-vis absorption spectra of pristine PCDTBT and PC₇₁BM and PCDTBT : PC₇₁BM layer in o-DCB.

spectra of PCDTBT : PC₇₁BM with mixing ratios of 1 : 1 show two absorbance maxima at 384 nm, 554 nm and a notable one shoulder band was observed around 440 nm, which could not be seen for the single layer of PCDTBT and PC₇₁BM [21, 24, 26, 27]. It was reported that bulk heterojunction layer of PCDTBT/PC₇₁BM revealed a strong absorption at 440 nm [26], which could play an important role to improve power conversion efficiency of organic thin film solar cells. The increase in the light absorption property of active layer leads to enhance the power conversion efficiency of organic thin film solar cell [21, 24, 26, 27].

4. Conclusions

The characteristics of OTFSCs device have been fabricated using PCDTBT:PC₇₁BM with mixing ratios of 1:1~1:8 and the influences of PEDOT : PSS film thicknesses on the photovoltaic performances have been studied and demonstrated. The mixing ratio of 1 : 4 device shows higher PCE of 4.15% with PEDOT : PSS layer at 3000 rpm; it confirms the optimized mixing ratio which results in a higher PCE compared with the other mixing ratios. by changing thickness of PEDOT : PSS layer about 45 nm than PCE = 4.15% with higher thickness of 62 nm. The improvement in the device performance is due to the change in the PEDOT : PSS layer thickness and influence of the PEDOT : PSS surface properties. The optimized thickness of PEDOT : PSS layer at 45 nm shows uniform interfacial surface which leads to enhance the interfacial properties of the PEDOT : PSS layer and an increase in the charge transport and extraction. As a result, an improvement in the device performance was achieved.

Conflict of Interests

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

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