

## Research Article

# Characterization of the Organic Thin Film Solar Cells with Active Layers of PTB7/PC<sub>71</sub>BM Prepared by Using Solvent Mixtures with Different Additives

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Organic thin film solar cells (OTFSCs) were fabricated with blended active layers of poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]] (PTB7)/[6,6]-phenyl-C<sub>71</sub>-butyric (PC<sub>71</sub>BM). The performances of active layers are prepared in chlorobenzene (CB) with different additives of 1-chloronaphthalene (CN) and 1,8-Diiodooctane (DIO) by a wet process with spin coating technique. The effects of different solvent additives on photovoltaic parameters such as fill factor, short circuit current density, and power conversion efficiency of active layers are reported. The absorption and surface morphology of the active layers are investigated using UV-visible spectroscopy and atomic force microscopy, respectively. The results indicate that structural and morphological changes were induced by the additives with solvent. The current density-voltage (*J-V*) characteristics of photovoltaic cells were measured under the illumination of simulated solar light with 100 mW/cm<sup>2</sup> (AM 1.5 G) by an Oriel 1000 W solar simulator. The OTFSCs of PTB7/PC<sub>71</sub>BM prepared with organic solvent additives of DIO+CN show more improved PCE of 4.96% by spin coating method.

## 1. Introduction

Recently, a great paradigm shift has occurred in the generation and consumption of energy. The so-called “clean” energy is consequently an utmost critical issue for all the nations and entire civilized societies. In the 34th G8 Toyako summit, it has been decided to cut the carbon emission up to 50% by the year 2050 [1], and realization of the “low-carbon-society” has been worked towards on global level. Moreover, the recent “Great East Japan Earthquake” has aroused a great attention to severe riskiness of nuclear power, so that recommencement of electricity generation by the nuclear power and construction of new nuclear power plant are rather reluctant. Consequently, a large scale electricity generation by new and renewable energy source has been raised as an urgent issue [2]. It has been reported [3] that electricity generation in Japan in November, 2012, was comprised of 2.7% from

nuclear power, 90.6% from fossil fuel, 6.2% from hydraulic power, and only 0.6% from new energy source, respectively, and the portion of renewable energy was still in the minute level of 10%. All of those kinds of situations have caused a critical status in Japan in terms of supply and demand of energy. Therefore, the Japanese government has devised a target in the general energy planning, aiming for elevating the proportion of “zero-emission-power source” up to 70% by the year 2030, so that introduction of the new and renewable energy source should be accelerated as fast as possible [4].

Solar light power generation could have a promising possibility for contributing to elevate the self-sufficiency of energy needs by domestic energy resource in Japan. It has been planned, therefore, that the mass of introduction for solar light power generation should be raised by ten times by the year 2020 and forty times by the year 2030. Additionally, an innovative solar light power generation is

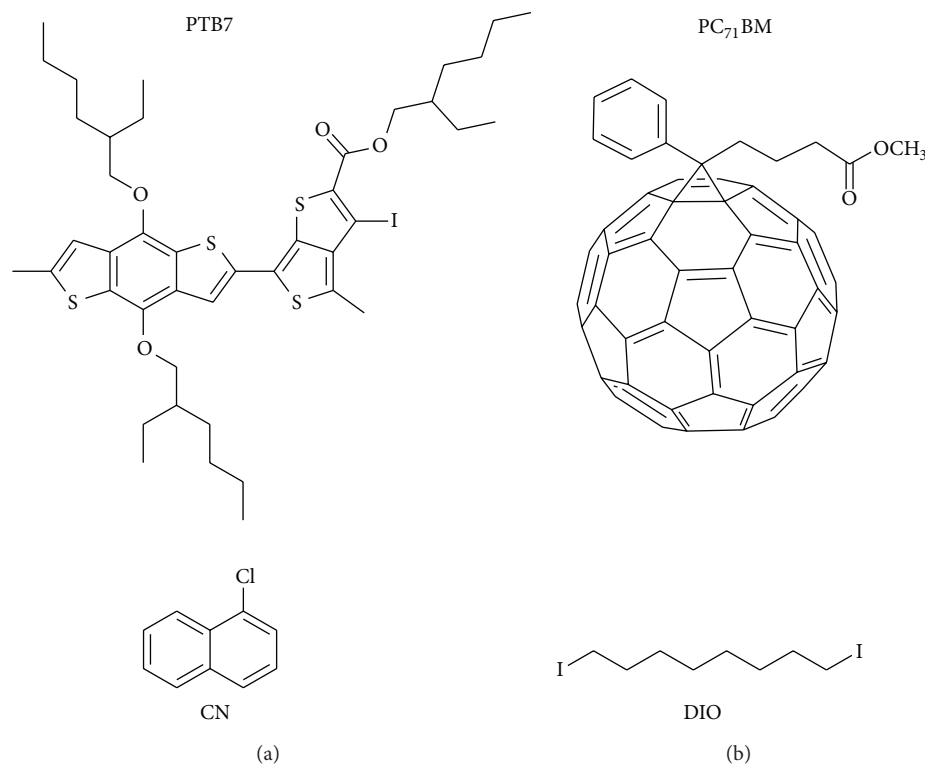


FIGURE 1: Molecular structures of PTB7, PC<sub>71</sub>BM, CN, and DIO.

aimed by establishing a sophisticated technology of solar cell through usage of new materials and device structure, so that power generation efficiency of over 40% and power generation cost of 7 Yen/kWh should be achieved after the year 2030 [5].

First-generation solar cells based on Si have demerits of high cost and limitation of installing location due to heavy weight. Second-generation solar cells based on inorganic compounds (amorphous/poly crystalline Si and CIGS) have been introduced in commercial market. Although second-generation solar cells show still relatively inferior power conversion efficiency, they are advantageous from a cost perspective. It is acutely expected for third-generation solar cells that they would have improved power conversion efficiency of over 40% and a merit of low cost comparable to those of the second-generation solar cells. Generally, organic thin film solar cells (OTFSCs) are acknowledged that they are lighter than any other counterparts and suitable for low-cost fabrication [6–9]. OTFSC materials have been focused towards the energy-consumption and environment friendly viewpoint. Recently, power conversion efficiency of the OTFSCs has been improved up to 10–11% [8–10]. Moreover, they could be formed to any shape due to flexible characteristic, so that they could be optimized for portable and/or wearable application [5]. Similar to the case of liquid crystal displays (LCDs) [11], light emitting diodes (LEDs), inorganic solar cells, and organic light emitting diodes (OLEDs) are based on similar materials and structures to those of OTFSCs. Recent rapid commercialization of the OLEDs is mainly concentrated on mobile display applications [12], but it is expected that

the market for OLEDs could be expanded in the near future to large-size TVs and lighting applications. The success of the OLEDs has motivated vigorous developments of OTFSCs based on devices and related materials [13].

In this present work, we report the performance of PTB7/PC<sub>71</sub>BM based OTFSCs prepared in chlorobenzene (CB) solvent with different additives. The active layers of PTB7/PC<sub>71</sub>BM were prepared by wet processes with spin coating technique. Organic solvent additives were varied to prepare the organic active layer, and the effect of the solvent additives was investigated in relation to surface topology of PTB7, PC<sub>71</sub>BM, and PTB7/PC<sub>71</sub>BM thin films. The performance of the resulting OTFSC devices and surface topology of the active layers were comparatively discussed.

## 2. Experimental Procedure

Active layers of organic thin film solar cell device were prepared using Poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]] (PTB7), as organic electron donor material, and [6,6]-Phenyl C<sub>71</sub> butyric acid methyl ester (PC<sub>71</sub>BM) used as electron acceptor material. Figure 1 shows the molecular structure of the PTB7, PC<sub>71</sub>BM, 1-chloronaphthalene (CN), and 1,8-Diiodooctane (DIO). PC<sub>71</sub>BM is an organic material chemically modified to be dissolved in an organic solvent. To enhance the hole transport characteristic, a thin film of

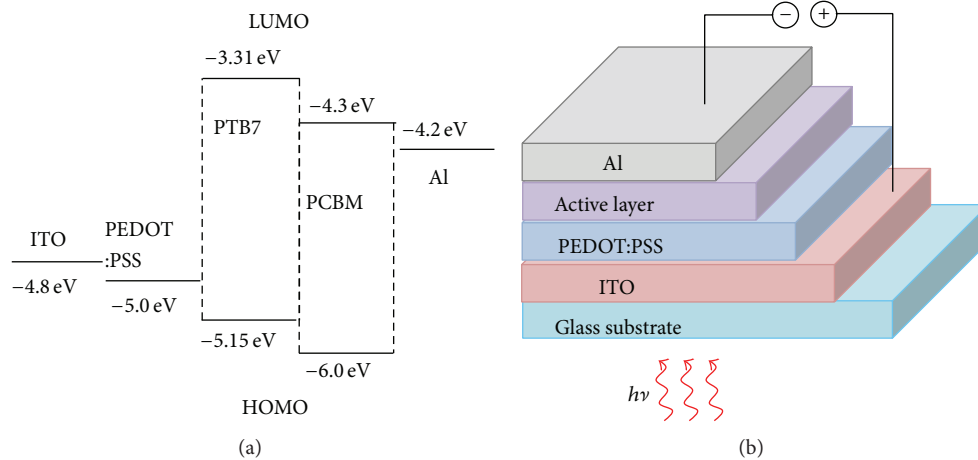


FIGURE 2: Schematics of the (a) energy band structure of the OTFSC with hole transfer layer of PEDOT:PSS and blended active layer of PTB7:PC<sub>71</sub>BM and (b) cross section view of OTFSC device.

poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) [PEDOT:PSS] was prepared by using a commercially available material of PH1000 (Heraeus Clevios), which serves as a hole transfer layer interfaced with the anode and electron donor layer of PTB7. While many organic materials could only be dissolved in organic solvents, PEDOT:PSS could be dissolved in water, and it could be easily used in wet process to prepare thin films. Moreover, PEDOT:PSS would serve for planarization of the unevenness on the ITO substrate surface as well as to enhance the interface characteristic of anode substrate and thin film.

As for the process to prepare the thin film components of OTFSC device, a spin coating technique was adopted for compatibility to large-area-sized device fabrication. Firstly, indium tin oxide (ITO) coated glass substrates were cleaned ultrasonically in a sequential step by using a neutral detergent, distilled water, acetone, and ethanol for 10 min, respectively. The cleaned substrates were exposed to UV in vacuum for 30 min, aiming for a hydrophilic state of the treated ITO substrates. Then hole transfer layer of PEDOT:PSS thin film was prepared on the substrate by spin coating technique: rotation speed of 5000 rpm and rotating time of 30 s. The PEDOT:PSS thin film was then annealed for 15 min at a temperature of 140°C. Composite organic active layers of PTB7:PC<sub>71</sub>BM with a relative ratio of 1:1.5 were deposited also by spin coating technique, where different organic solvents were used to prepare the precursor solutions: (1) 2 mL of pristine chlorobenzene (CB), (2) 2 mL of CB added with an additive of DIO (3 v%), (3) 2 mL of CB added with an additive of CN (3 v%), and (4) 2 mL of CB added with DIO (3 v%) and CN (3 v%). The PTB7:PC<sub>71</sub>BM (9 mg:13.5 mg) was dissolved in each organic solvent mixture and the spin coating process was carried out with a rotation speed of 800 rpm and rotation time of 60 s. The thickness of the active layer was ~86 nm, as measured using a Dektak II profilometer. Aluminum (Al) electrode (~100 nm) for the organic thin film solar cell device was then prepared by a thermal vacuum evaporation through a shadow mask in a high vacuum of

$1.3 \times 10^{-4}$  Pa and the effective area of the resulting device was 9 mm<sup>2</sup>.

The organic thin film solar cell device presented in this report is based on an electron donor layer of PTB7 with deep highest occupied molecular orbital (HOMO) level and low band gap interfaced with a fullerene derivative of PC<sub>71</sub>BM as a representative electron acceptor. Figure 2 shows a schematic of energy band diagram and device structure of the OTFSC. Performance of the OTFSC device was investigated by measuring the current density-voltage (*J-V*) characteristics in a solar simulator illuminated by a light source of Xn lamp combined with an air mass filter: 100 mW/cm<sup>2</sup>, AM 1.5. A shadow mask made of a thick black sheet was placed in front of the active device area in order to avoid the overestimation of the current density and power conversion efficiency. All measurements were carried out in air without any encapsulation.

As shown in Figure 2, PTB7 has the lowest unoccupied molecular orbital (LUMO) level of -3.31 eV and a HOMO level of -5.15 eV. It is generally acknowledged that P3HT has a LUMO level of -2.9 eV and HOMO level of -5.0 eV [14, 15]. It could be expected that PTB7 would reveal relatively narrow band gap and excellent light absorption characteristic. In addition, open circuit voltage of the OTFSC with PTB7:PC<sub>71</sub>BM would be similar to that of the OTFSC with P3HT:PCBM [16].

### 3. Results and Discussions

**3.1. Current Density- (*J*-) Voltage (*V*) Characteristics.** The current density-voltage (*J-V*) characteristics of OTFSC devices were measured under the illumination of simulated solar light with 100 mW/cm<sup>2</sup> (AM 1.5G) by an Oriel 1000 W solar simulator. *J-V* characteristics of the four different OTFSC devices based on blended active layer of PTB7:PC<sub>71</sub>BM are presented in Figure 3. Photovoltaic parameters of the four different OTFSC devices along with series resistance (*R<sub>s</sub>*)

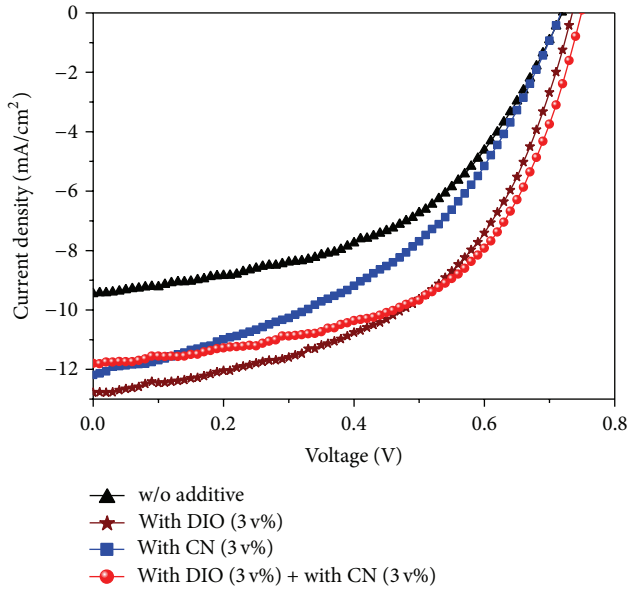


FIGURE 3:  $J$ - $V$  characteristics of the four different organic thin film solar cell devices based on blended active layer of PTB7:PC<sub>71</sub>BM prepared with different organic solvent mixture.

and shunt resistance ( $R_{sh}$ ) are given in Table 1: short circuit current density ( $J_{sc}$ ), open circuit voltage ( $V_{oc}$ ), fill factor (FF), and power conversion efficiency (PCE;  $\eta$ ).

The OTFSC of PTB7/PC<sub>71</sub>BM prepared with pristine organic solvent of CB reveals a PCE of 3.38%, which is distinctively lower than those of the OTFSC of PTB7/PC<sub>71</sub>BM prepared with organic solvent of CB with additives [6, 7, 17, 18]. On the other hand, the OTFSC of PTB7/PC<sub>71</sub>BM prepared with organic solvent of CB with additive of DIO (3 v%) shows high PCE of 4.89%. It may be attributed to the formation of interpenetrating network with better morphology by the addition of DIO, which decrease the series resistance and enhance the charge carrier transport properties, as well as increase of equivalent parallel resistance due to suppression of charge carrier recombination in the active layer. The OTFSC of PTB7/PC<sub>71</sub>BM prepared with organic solvent of CB with additive of CN (3 v%) shows a moderately improved PCE of 3.86% with low FF of 0.443. An additive of DIO with high boiling point (b.p) of 169°C could influence the formation of nanoscale morphology of molecules, decrease of  $R_s$ , and increase of shunt resistance ( $R_{sh}$ ) due to suppression of hole-electron recombination. However, relatively higher b.p. of the CN (263°C) than that of the DIO might have resulted in a moderate formation of the nanomorphology of molecules. In the device prepared with CB+CN, the  $R_s$  is increased and  $R_{sh}$  is decreased. The decrease in  $R_{sh}$  is attributed to the leakage current induced by pinholes and traps in the active layer morphology and the increase in  $R_s$  is due to recombination loss. Therefore, FF of the device is reduced as consequences of both  $R_{sh}$  and  $R_s$ . The OTFSC of PTB7/PC<sub>71</sub>BM prepared with organic solvent additives of DIO+CN shows more improved PCE of 4.96%, which implies that the effect of the additive of DIO is dominant for enhancement of the device performance through better intermixing between PTB7 and PC<sub>71</sub>BM.

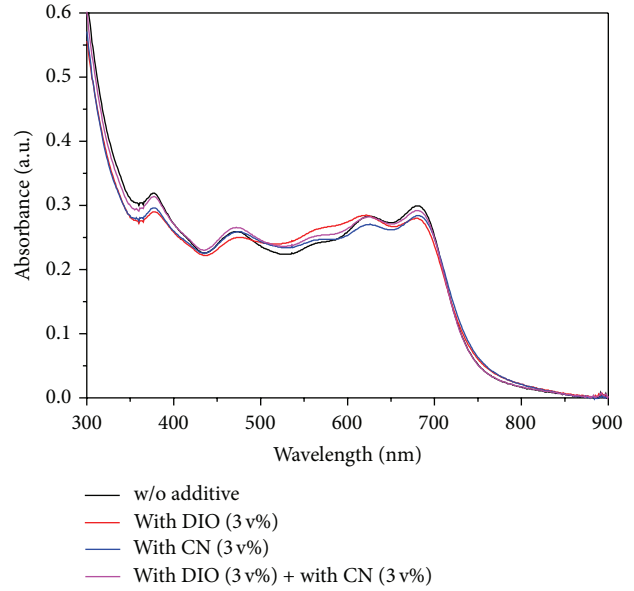


FIGURE 4: UV/Vis spectra of four different organic active layers of PTB7/PC<sub>71</sub>BM prepared with different organic solvent mixture.

The increase in FF (0.556) originates from the reduction in  $R_s$  by the additives of DIO+CN. Because of the smaller  $R_s$  and higher  $R_{sh}$ , the FF of the OTFSC prepared with DIO+CN additives was higher than that of the OTFSC prepared with DIO and OTFSC prepared with CN additives.

**3.2. Absorption and Atomic Force Microscopy Studies.** Absorption characteristic of four different organic active layers of PTB7/PC<sub>71</sub>BM was investigated by UV/Vis spectrophotometer and depicted in Figure 4. It can be seen that there is no shift of the absorption peak for the samples prepared with organic additives from that of the sample prepared with pristine CB. It indicates that the thin films prepared with additives have amorphous phase. Topology of the organic active layers was investigated by atomic force microscopy (AFM). The thin films of PTB7 were prepared by using the three varieties of organic solvent mixture: (1) PTB7 with pristine CB, (2) PTB7 with CB and DIO (3 v%), and (3) PTB7 with CB and CN (3 v%). Figure 5 shows the AFM images, and roughness parameters obtained from the AFM analysis are given in Table 2: peak-to-valley value and room-mean-square value. All the three samples show a minute formation of micrograins (Figure 5) and root-mean-square roughness of one-nanometer level. It indicates again that the thin films do not show any crystalline nature.

Effects of the organic solvent on the surface morphology of the PC<sub>71</sub>BM thin films were also investigated by AFM. Figure 6 shows the AFM images of the PC<sub>71</sub>BM thin films prepared by using pristine organic solvent of CB, CB with additive of DIO (3 v%), and CB with additive of CN (3 v%), respectively. Surface roughness parameters obtained from the AFM analysis are presented in Table 3: peak-to-valley value and root-mean-square value. All the PC<sub>71</sub>BM thin films (Table 3) reveal distinctively smaller parameters of surface

TABLE 1: Photovoltaic parameters of the four different organic thin film solar cell devices based on blended active layer of PTB7:PC<sub>71</sub>BM prepared with different organic solvent mixture.

PTB7:PC <sub>71</sub> BM in organic solvent mixture	$J_{sc}$ [mA/cm <sup>2</sup> ]	$V_{oc}$ [V]	FF	PCE( $\eta$ ) [%]	$R_s$ [ $\Omega$ ]	$R_{sh}$ [ $\Omega$ ]
CB without additive	9.46	0.72	0.496	3.38	167	3163
CB with DIO (3 v%)	12.78	0.74	0.516	4.89	74	3304
CB with CN (3 v%)	12.11	0.72	0.443	3.86	133	2256
CB with DIO (3 v%)+CN (3 v%)	11.90	0.75	0.556	4.96	63	5035

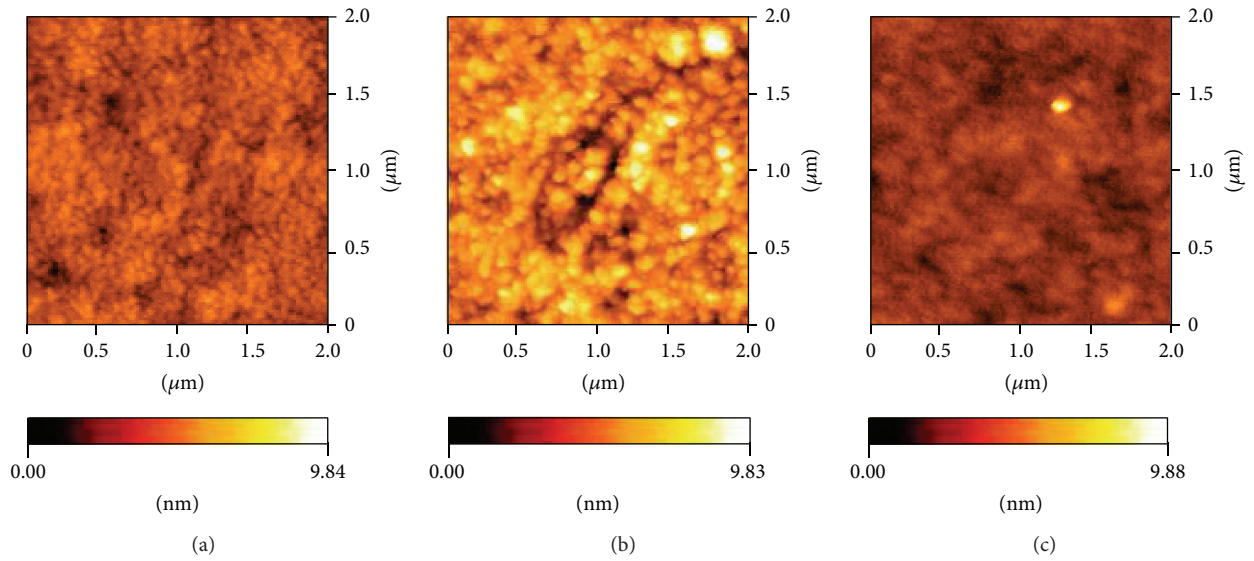


FIGURE 5: AFM images of the PTB7 thin films prepared with (a) pristine CB, (b) CB and additive of DIO (3 v%), and (c) CB and additive of CN (3 v%).

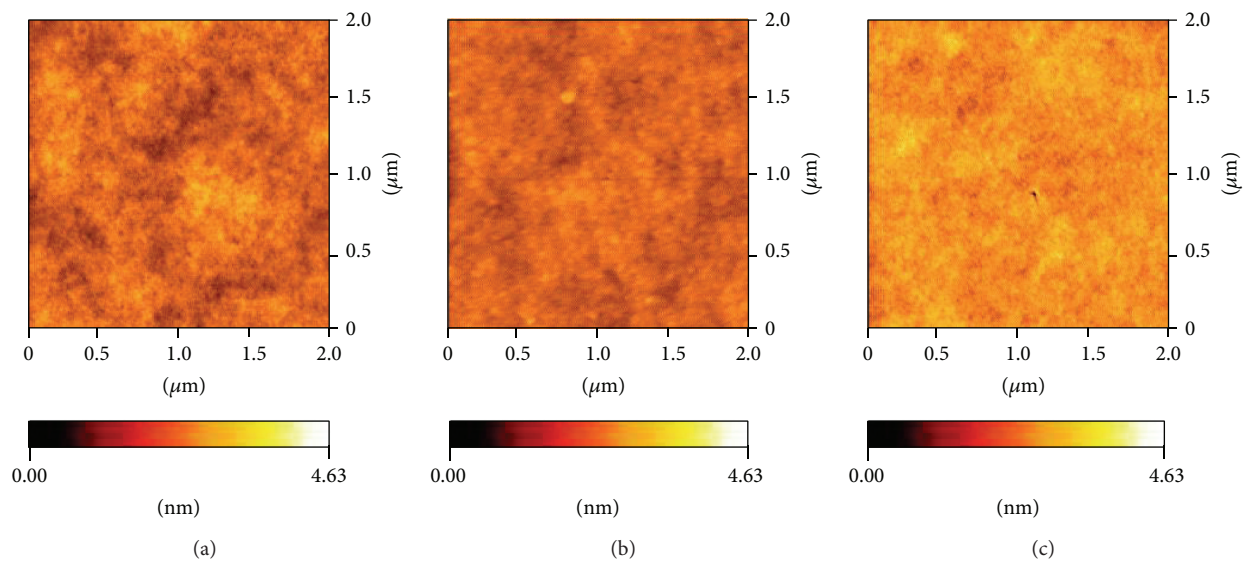


FIGURE 6: AFM images of the PC<sub>71</sub>BM thin films: (a) prepared with pristine CB, (b) prepared with CB and additive of DIO (3 v%), and (c) prepared with CB and additive of CN (3 v%).

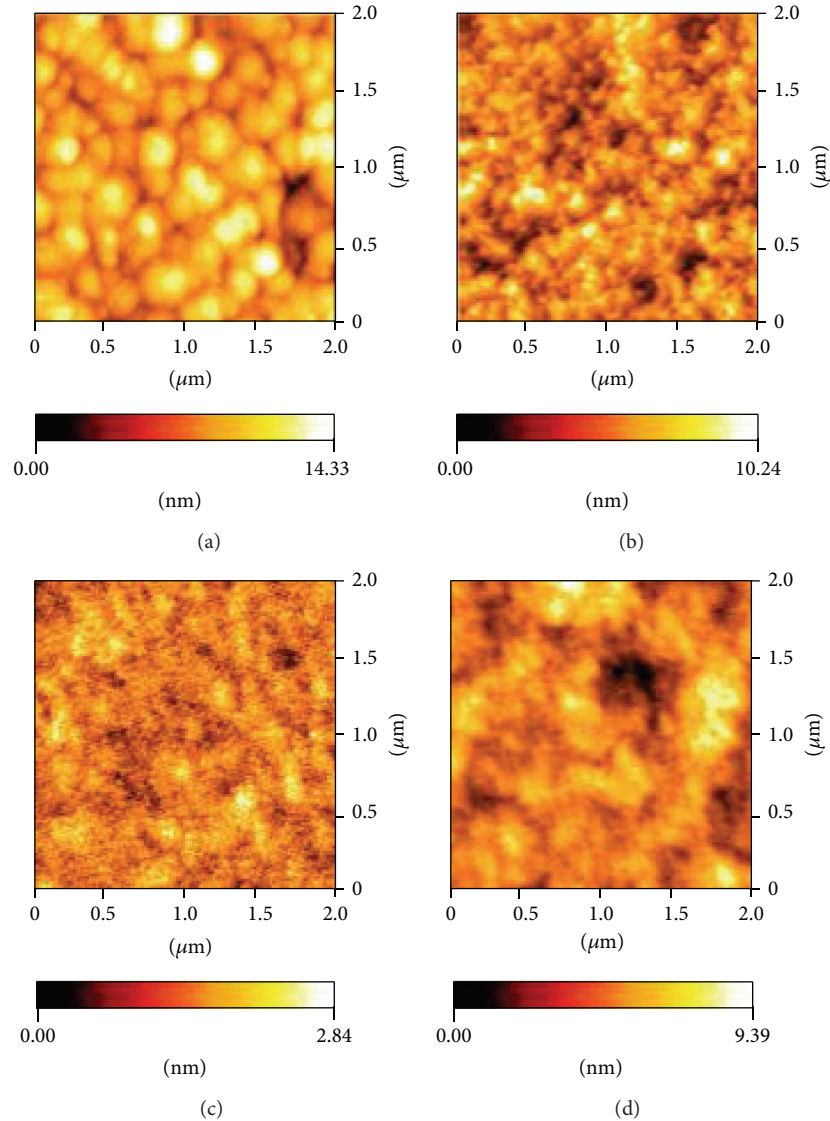


FIGURE 7: AFM images of the PTB7/PC<sub>71</sub>BM thin films: (a) prepared with pristine CB, (b) prepared with CB and DIO (3 v%), (c) prepared with CB and CN (3 v%), and (d) prepared with CB and DIO (3 v%)+CN (3 v%).

TABLE 2: Surface roughness parameters of the PTB7 thin films obtained from AFM analysis.

Layer	Peak-to-valley [nm]	Root-mean-square roughness [nm]
PTB7 with pristine CB	7.0	0.7
PTB7 with CB and DIO (3 v%)	12.7	1.5
PTB7 with CB and CN (3 v%)	9.3	0.6

roughness, which is quite different from the cases of PTB7 (Table 2). It can be observed (Figure 6) that PC<sub>71</sub>BM reveals excellent solubility in all the organic solvent mixtures of pristine CB, CB with additive of DIO (3 v%), and CB with additive of CN (3 v%) [19].

Finally, the effect of the organic solvent on the surface morphology of the resulting thin films of blended

TABLE 3: Surface roughness parameters of the PC<sub>71</sub>BM thin films obtained from AFM analysis.

Layer	Peak-to-valley [nm]	Root-mean-square roughness [nm]
PC <sub>71</sub> BM with pristine CB	3.4	0.4
PC <sub>71</sub> BM with CB and DIO (3 v%)	3.3	0.3
PC <sub>71</sub> BM with CB and CN (3 v%)	3.8	0.3

PTB7/PC<sub>71</sub>BM was investigated by AFM, which could play a decisive role in the performance of organic thin film solar cell. Figure 7 shows the surface topology of three different active layers of blended PTB7/PC<sub>71</sub>BM [14]. Surface roughness parameters of the PTB7:PC<sub>71</sub>BM prepared in CB

TABLE 4: Surface roughness parameters of the PTB7:PC<sub>71</sub>BM active layer obtained from AFM analysis.

Layer	Peak-to-valley [nm]	Root-mean-square roughness [nm]
PTB7:PC <sub>71</sub> BM with pristine CB	13.4	1.18
PTB7:PC <sub>71</sub> BM with CB and DIO (3 v%)	10.7	0.53
PTB7:PC <sub>71</sub> BM with CB and CN (3 v%)	11.8	0.72
PTB7:PC <sub>71</sub> BM with CB and DIO (3 v%) + CN (3 v%)	7.4	0.64

with different solvent additives are presented in Table 4. The PTB7/PC<sub>71</sub>BM thin film prepared by using pristine organic solvent of CB shows a large micrograin of 1  $\mu\text{m}$  (Figure 7(a)). It implies for the case that molecules of PTB7 and PC<sub>71</sub>BM might have insufficient mutual solubility during the thin film formation process, and a mutual interaction between the molecules might have an influence on such kind of thin film formation [20]. On the contrary, the PTB7/PC<sub>71</sub>BM thin film prepared with a mixture of CB and DIO (3 v%) reveals conspicuously smaller micrograins on the surface (Figure 7(b)). An interpenetrated blending through excellent percolation of micrograins might have been achieved for the molecules of PTB7 and PC<sub>71</sub>BM through the effect of organic solvent mixture of CB and DIO, which could be contributed to the distinctively enhanced PCE (4.89%) of the resulting OTFSC (Table 1) [6, 17]. The PTB7/PC<sub>71</sub>BM thin film prepared with a mixture of CB and CN (3 v%) (Figure 7(c)) shows smaller micrograins similar to the case of PTB7/PC<sub>71</sub>BM thin film prepared with a mixture of CB and DIO (3 v%) (Figure 7(b)), but it reveals an inferior connectivity between the micrograins. It might be a reason why the OTFSC prepared with a mixture of CB and CN (3 v%) reveals a moderate PCE of 3.86%, which is distinctively lower than that of the OTFSC with PTB7/PC<sub>71</sub>BM prepared with a mixture of CB and DIO, although it is rather higher than the OTFSC with PTB7/PC<sub>71</sub>BM prepared with pristine CB. The OTFSC of PTB7/PC<sub>71</sub>BM was prepared with organic solvent (CB) with additives of CN + DIO (3 v%), and the addition of DIO might have resulted in a moderate formation of the nanomorphology of molecules (Figure 7(d)), which implies that the PCE of the OTFSC further improved to 4.96%. The effect of the additive of DIO is dominant for enhancement of the device performance, so that it is similar to that of the OTFSC of PTB7/PC<sub>71</sub>BM prepared with organic solvent mixture of CB and DIO.

#### 4. Conclusions

The OTFSC devices were fabricated with blended active layers of PTB7/PC<sub>71</sub>BM with a relative ratio of 1:1.5. The active layers of PTB7/PC<sub>71</sub>BM were prepared by a spin coating process with four different kinds of organic solvents: (1) pristine CB, (2) CB with additive of DIO (3 v%), (3) CB with additive of CN (3 v%), and (4) CB with additives of DIO

(3 v%)+CN (3 v%). Solubility of the PTB7 and PC<sub>71</sub>BM in the organic solvent mixtures was investigated by surface topology of the resulting thin films through AFM images. PC<sub>71</sub>BM revealed excellent solubility in the organic solvent of pristine CB as well as in mixtures solvents of CB/DIO, CB/CN, and CB/DIO+CN. For the case of pristine CB, the resulting composite thin film of PTB7/PC<sub>71</sub>BM revealed relatively large micrograins of 1  $\mu\text{m}$  on the surface, which indicates an inferior mutual solubility and insufficient interpenetration of PTB7 and PC<sub>71</sub>BM in the organic solvent of CB. Conspicuously smaller micrograins of one-tenth level were observed for the PTB7/PC<sub>71</sub>BM thin film prepared with organic solvent mixture of CB and DIO (3 v%), which is possibly caused by improved percolation between PTB7 and PC<sub>71</sub>BM due to the effect of DIO. Consequently, PCE of 4.89% was achieved for the resulting OTFSC based on the interpenetrated active layer of PTB7/PC<sub>71</sub>BM prepared with organic solvent mixture of CB and DIO. Enhanced transport of charge carriers might have been achieved through formation of nanomorphology in the blended active layer of PTB7/PC<sub>71</sub>BM due to the effect of DIO added in organic solvent of CB. Although the PTB7/PC<sub>71</sub>BM prepared with organic solvent mixture of CB and CN (3 v%) showed improved nanomorphology compared to the sample prepared with pristine CB, sufficient connectivity of the micrograins might not be achieved, which could be resulted in suppression of charge-carrier transport. The moderate increase of PCE up to 3.86% might have been caused by the effect of organic solvent mixture of CB with CN. However, the OTFSC of PTB7/PC<sub>71</sub>BM prepared with organic solvent additives of DIO + CN shows formation of the intermixed nanomorphology, which implies that the PCE of the OTFSC further improved to 4.96% by spin coating method.

#### Conflict of Interests

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

#### Authors' Contribution

Masakazu Ito and Palanisamy Kumar contributed equally to this work.

#### Acknowledgments


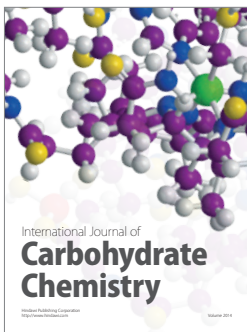
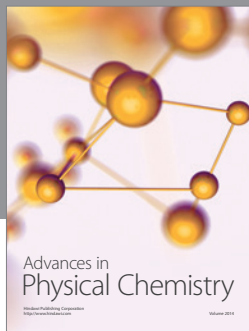
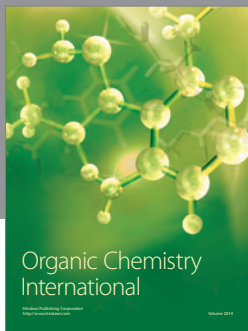
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