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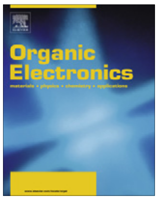

Non-peripheral octahexylphthalocyanine doping effects in bulk heterojunction polymer solar cells

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ABSTRACT

The improvement of long-wavelength sensitivity in bulk heterojunction organic thin-film solar cells based on poly(3-hexylthiophene) (P3HT) by the addition of the soluble phthalocyanine derivative, 1,4,8,11,15,18,22,25-octahexylphthalocyanine ($C6PcH_2$) is reported. $C6PcH_2$ possesses near-infrared absorption and can be mixed with a P3HT:1-(3-methoxycarbonyl)-propyl-1-1-phenyl-(6,6)C61 (PCBM) bulk heterojunction active layer. By doping $C6PcH_2$, the photosensitivity in the long-wavelength region was improved, and the energy conversion efficiency reached 3.0% at a composition ratio of P3HT:$C6PcH_2$:PCBM = 10:3:10. We discuss the principle of photoconversion in the bulk heterojunction solar cell based on the P3HT:$C6PcH_2$:PCBM active layer by taking into consideration the existence of both highly ordered P3HT domains and hexagonal columnar structures of $C6PcH_2$, and the micro-phase separation of P3HT and $C6PcH_2$ in the active layer.

1. Introduction

Since the discovery of photoluminescence quenching [1] and photoconductivity enhancement [2] based on photoinduced charge transfer [3] between the π-conjugated main chain in conducting polymers and fullerenes, conducting polymer–fullerene systems have been investigated as donor–acceptor type organic solar cells. Donor–acceptor junction structures utilizing conducting polymers, such as bulk heterojunction [4] or interpenetrating heterojunction [5], could be fabricated by wet processes because of the high solubility of conducting polymers in organic solvents.

Poly(3-hexylthiophene) (P3HT) is known as one of the conventional active layer materials used in organic thin-film solar cells, and its high efficiency was reported in previous works [6–10]. However, the photoabsorption of P3HT is limited to the visible range at wavelengths shorter than 600 nm, and infrared light contained in solar light is transmitted through the P3HT film, resulting in absorption loss. Therefore, narrow-band-gap conducting polymers [11] and tandem solar cells [12] have been developed for the purpose of obtaining efficient absorption over a wide wavelength range.

On the other hand, the improved long-wavelength sensitivity of organic thin-film solar cells utilizing P3HT upon the addition of dye materials exhibiting long-wavelength absorption has been reported [13–16]. However, in organic thin-film solar cells based on a bulk heterojunction system utilizing P3HT and a fullerene derivative, 1-(3-methoxycarbonyl)-propyl-1-1-phenyl-(6,6)C61 (PCBM), the alignment of P3HT is disordered by the doping of additives. Although the fabrication of the active layer structure of P3HT and PCBM, in which dye molecules such as phthalocyanine derivatives are dispersed, and the long-wavelength sensitivity has been discussed previously [14–16], the marked improvement of energy conversion efficiency by the addition of materials exhibiting long-wavelength

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absorption has not been achieved. In the previous reports, the absorption peak width of the phthalocyanine derivative was narrow because of the low π-stacking characteristic of phthalocyanine derivatives, and the absorbance was low because of the small composite amount of phthalocyanine derivatives. For efficient carrier generation and transport with absorption over a wide wavelength range, a carrier path based on a dye molecule with high crystallinity and high absorption are important.

Recently, we have demonstrated high hole and electron drift mobilities of 1.4 and 0.5 cm^2/Vs, respectively, in the crystalline phase of mesogenic non-peripheral octahexylphthalocyanine, 1,4,8,11,15,18,22,25-octahexylphthalocyanine ($C6PcH_2$) [17]. $C6PcH_2$ exhibits high absorption in the wavelength range of 600–800 nm corresponding to the Q-band, and $C6PcH_2$ forms a hexagonal columnar structure owing to its high self-organization and π-stacking properties. We also reported a simple organic solar cell with a bulk heterojunction of $C6PcH_2$ and PCBM that was fabricated by the spin-coating method and demonstrated its high energy conversion efficiency [18,19]. It is considered that $C6PcH_2$ is a candidate dopant material for P3HT:PCBM bulk heterojunction solar cells.

In this study, we report on the improvement of long-wavelength sensitivity in P3HT:PCBM bulk heterojunction solar cells by doping $C6PcH_2$ and discuss their photovoltaic properties by taking into consideration the microphase separation and photoinduced charge separation.

2. Experimental details

Regioregular P3HT and PCBM were used as purchased from Merck & Co., Inc., and Frontier Carbon Ltd., respectively. $C6PcH_2$ was synthesized in accordance with the reported method [20] with slight modifications and purified by column chromatography (silicagel with toluene as an eluent) followed by repetitive recrystallization from toluene–methanol (1:2) solution. The molecular structures of P3HT, PCBM and $C6PcH_2$ are shown in Fig. 1.

The fabrication of solar cells in this study was carried out in the following manner. A hole transport layer of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, Baytron P VP AI 4083) was spin-coated onto an indium-tin-oxide (ITO)-coated quartz substrate at 3000 rpm for 60 s using an aqueous solution diluted with the same volume of isopropanol, and dried at 100 °C for 10 min in an oven under atmospheric conditions. The thickness of the PEDOT:PSS layer was estimated to be approximately 30 nm.

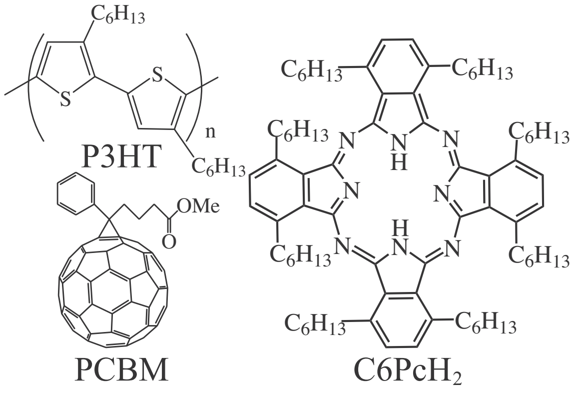

Fig. 1. Molecular structures of P3HT, PCBM and $C6PcH_2$.

10 mg of P3HT, x mg of $C6PcH_2$ (x = 0, 1, 2, 3, 4, 5, 6) and 10 mg of PCBM were dissolved in 1 ml of chloroform. The P3HT:$C6PcH_2$:PCBM mixed solution was spin-coated onto the PEDOT:PSS layer at 1000 rpm for 60 s in a glove box filled with argon gas. The thickness of the active layer was estimated to be approximately 180 nm.

0.8 nm-thick lithium fluoride (LiF) and 80 nm-thick aluminum (Al) layers as a counter electrode to the ITO were deposited onto the composite layer through shadow masks by thermal evaporation under a pressure of approximately 10^{-4} Pa. The evaporation speeds of the LiF and Al layers were approximately 0.1 and 5.0 Å/s, respectively. The active area of the solar cell was 2×2 mm^2.

The absorbance spectra were measured using a spectrophotometer (Shimadzu UV-3150). X-ray diffraction (XRD) patterns were measured using a Rigaku X-ray diffractometer (RINT 1100). The highest occupied molecular orbital (HOMO) levels of P3HT and $C6PcH_2$ films were measured using a photoelectron spectrometer (RIKEN KEIKI AC-2), and the energy band gaps of P3HT and $C6PcH_2$ films were estimated from the absorption edge energy.

Current–voltage characteristics and photocurrent spectra were measured in vacuum at room temperature. The photocurrent spectra were measured with a programmable electrometer (Keithley 617S) using xenon lamp light passing through a monochromator as a light source, and the external quantum efficiency (EQE) was estimated for each incident light wavelength using EQE (%) = $1240 \times I$ (A/cm^2) $\times 100/(\lambda$ (nm) $\times P_{in}$ (W/cm^2)), where I is the photocurrent density and λ is the wavelength of incident light. The current–voltage characteristics were measured with a high-voltage-source measurement unit (Keithley 237) under AM1.5G (100 mW/cm^2) solar-illuminated conditions. From the current–voltage characteristics under AM1.5G, the fill factor (FF) and energy conversion efficiency (η_e) were estimated according to the following definitions: $FF = I_{max}V_{max}/I_{sc}V_{oc}$ and $\eta_e = I_{sc}V_{oc}FF/P_{in}$, where I_{max} and V_{max} are the current density and voltage at the maximum output power, respectively, I_{sc} is the short-circuit current density, V_{oc} is the open-circuit voltage, and P_{in} is the intensity of the incident light.

3. Results and discussion

Fig. 2 shows the EQE spectra of the solar cells with P3HT:PCBM and P3HT:$C6PcH_2$:PCBM active layers. In the solar cell without $C6PcH_2$, although a high EQE of 74% was obtained at the wavelength of 540 nm, corresponding to the absorption peak of P3HT, a low EQE was obtained at wavelengths longer than 650 nm because of the low absorbance of the active layer. On the other hand, in the solar cell with $C6PcH_2$ (composite ratio of 10:3:10), a high EQE of 46% at 730 nm originating from the absorption of $C6PcH_2$ was obtained as well as a high EQE of 66% at

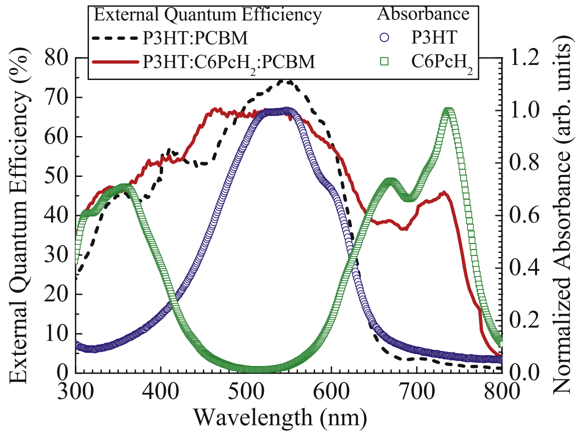

Fig. 2. EQE spectra of the solar cells with P3HT:PCBM and P3HT:C6PcH$_2$:PCBM (composite ratio of 10:3:10) active layers, and normalized absorbance spectra of P3HT and C6PcH$_2$ thin films on a quartz substrate.

540 nm. That is, the photosensitivity of the bulk heterojunction organic solar cell with the P3HT:PCBM active layer was improved in the near-infrared region.

Fig. 3 shows typical current–voltage characteristics of the solar cells with active layers of P3HT:C6PcH$_2$:PCBM, the composition ratios of which were 10:0:10, 10:3:10, and 10:6:10, and the composition ratio dependence of C6PcH$_2$ of the photovoltaic properties of the solar cell with the P3HT:PCBM active layer. V_{oc} was almost independent of the composition ratio of C6PcH$_2$, and FF gradually decreased with increasing amount of C6PcH$_2$ as clearly shown in Fig. 3(b). On the other hand, I_{sc} markedly depended on the composition ratio of C6PcH$_2$, and was 1.4 times higher at the composition ratio of P3HT:C6PcH$_2$ = 10:3 than in the case of without C6PcH$_2$. The long-wavelength sensitivity in the EQE spectrum, as shown in Fig. 2, was improved by doping C6PcH$_2$, resulting in the enhancement of I_{sc}. The energy conversion efficiency of the solar cell without C6PcH$_2$ was 2.3% with V_{oc} of 0.56 V, I_{sc} of 8.6 mA/cm^2 and FF of 0.48, and that of the solar cell containing C6PcH$_2$ with the composition ratio of P3HT:C6PcH$_2$ = 10:3 was improved to 3.0% with V_{oc} of 0.56 V, I_{sc} of 12.1 mA/cm^2 and FF of 0.44.

In our previous work on C6PcH$_2$:PCBM bulk heterojunction solar cell, the V_{oc} and FF were 0.81 V and 0.40, respectively[18]. From the results in this study, the HOMO–LUMO gap at the P3HT:PCBM bulk heterojunction must be the dominant factor in achieving an V_{oc} of 0.56 V and FF must be suppressed by doping C6PcH$_2$ in the P3HT:C6PcH$_2$:PCBM system. It is considered, therefore, that the enhanced I_{sc} strongly contributes to the improvement of the energy conversion efficiency. In the solar cell without C6PcH$_2$, it is considered that the energy conversion efficiency was lower than that of P3HT:PCBM solar cells reported previously [21] because the used solvent for spin-coating the active layer was chloroform in this study. In P3HT:PCBM solar cells, the typical solvents for spin-coating the active layer are chlorobenzene or *o*-dichlorobenzene, however, these solvents are inappropriate for C6PcH$_2$ because of low

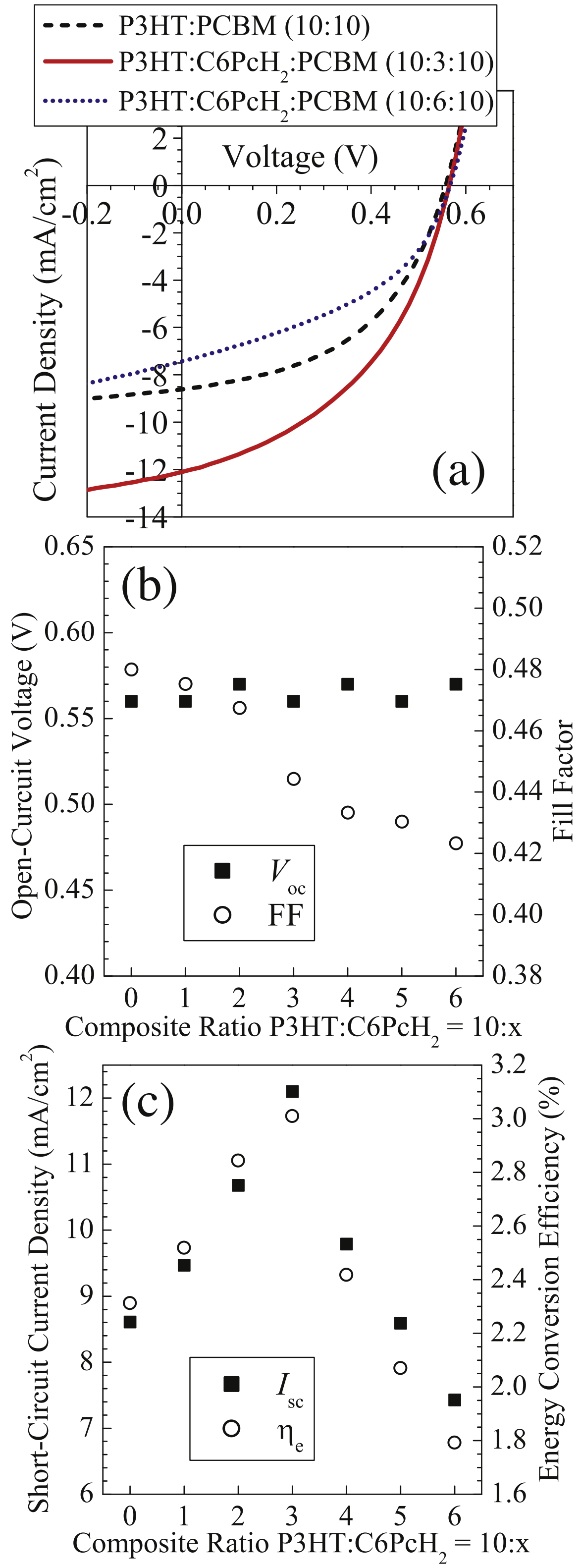

Fig. 3. (a) Current–voltage characteristics of the solar cells with P3HT:PCBM and P3HT:C6PcH$_2$:PCBM (composite ratio of 10:3:10 and 10:6:10) active layers under AM1.5 (100 mW/cm^2) solar-illuminated conditions, and C6PcH$_2$ composite ratio dependences of (b) V_{oc} and FF, and (c) those of I_{sc} and η_e.

wettability on the substrates. Therefore, chloroform was adopted as a solvent for the film fabrication at this stage. In the C6PcH$_2$:PCBM solar cell, a high V_{oc} of 0.81 V is observed in spite of the absorption loss in the visible region, and the energy conversion efficiency is about 3% [18]. On the other hand, in the P3HT:C6PcH$_2$:PCBM solar cell, a high I_{sc} of 12.1 mA/cm^2 is observed by the achievement of wide-wavelength absorption although a V_{oc} is about 0.56 V, and the energy conversion efficiency is also about 3%. In the previous reports on P3HT:PCBM solar cells, the energy conversion efficiency of about 4% was demonstrated [21], therefore, it is considered that the further improvement of energy conversion efficiency must be expected by the selection of organic solvents and thermal annealing treatment in the P3HT:C6PcH$_2$:PCBM solar cell. The detailed studies on the optimization of the solar cells utilizing P3HT, C6PcH$_2$ and PCBM still remain to be carried out and will be reported elsewhere.

Fig. 4(a) shows the XRD patterns of bulk heterojunction active layers with P3HT:C6PcH$_2$:PCBM, where the composition ratios are 10:x:10, x = 0,1,2,3,4,5,6. The diffraction peak around $2\theta = 5.4°$ corresponds to the distance of the main chain of P3HT (16.3 Å), and that around $2\theta = 4.9°$ corresponds to the distance of the hexagonal column of C6PcH$_2$ (18.0 Å). The schematic diagram of the microphase separation in the P3HT:C6PcH$_2$:PCBM active layers considered from the results of XRD patterns is shown in Fig. 4(b), (c) and (d). At the P3HT:C6PcH$_2$ composition ratio of 10:2, the diffraction of C6PcH$_2$ appeared, which gradually increased in intensity with increasing composition amount of C6PcH$_2$. Although the diffraction intensity of P3HT was maintained for composition ratios from 10:0 to 10:3, it decreased in the case of larger amounts of C6PcH$_2$. Therefore, it is considered that both P3HT and C6PcH$_2$ aggregate to form mutual microphase separation and that highly ordered P3HT domains and hexagonal columnar structures of C6PcH$_2$ coexist in the P3HT:C6PcH$_2$:PCBM bulk heterojunction thin film, especially with the P3HT:C6PcH$_2$ composition ratio of 10:3. At composition ratios from 10:4 to 10:6, it is considered that the alignment of P3HT was disordered by the addition of a large amount of C6PcH$_2$, resulting in the marked decrease of I_{sc}.

To interpret the results in this study, schematic diagrams of charge generation and transfer in the solar cells with a P3HT:C6PcH$_2$:PCBM bulk heterojunction active layer and energy diagrams of P3HT, C6PcH$_2$ and PCBM in

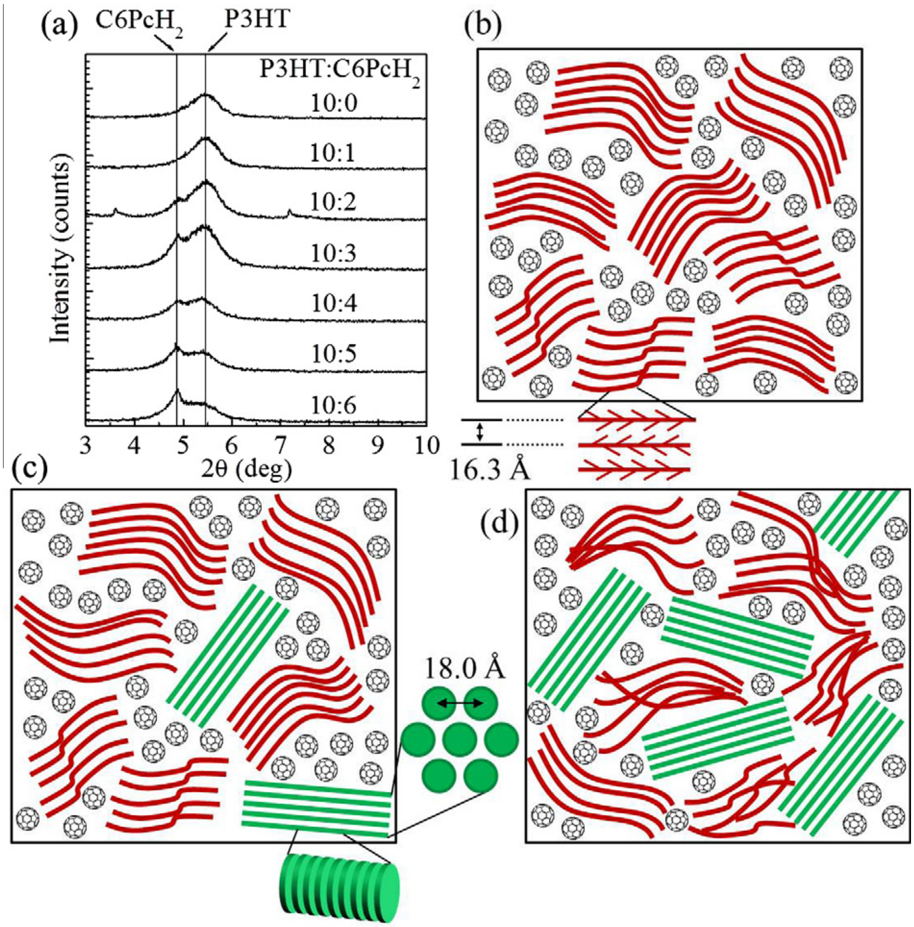

Fig. 4. (a) XRD patterns of the P3HT:C6PcH$_2$:PCBM bulk heterojunction films and schematic diagrams of micro phase separation in the P3HT:C6PcH$_2$:PCBM active layers, the composition ratios of which are (b) 10:0:10, (c) 10:3:10, and (d) 10:6:10.

the solar cells are proposed as shown in Fig. 5. From the photoelectron spectroscopy measurement and absorption edge energy, the HOMO and lowest unoccupied molecular orbital (LUMO) levels of P3HT were estimated to be −4.8 and −2.8 eV, respectively. Those of $C6PcH_2$ were estimated to be −5.3 and −3.7 eV, respectively. Therefore, P3HT acts as a donor molecule and $C6PcH_2$ acts as an acceptor molecule at the heterojunction of P3HT and $C6PcH_2$. Moreover, $C6PcH_2$ has high ambipolar drift mobilities in the direction of its columnar axis, which are higher than those of conventional organic semiconductors [17], and could efficiently transport electrons. Therefore, it is considered that the photoinduced charge transfer in the donor–acceptor system occurs at the heterojunction between P3HT and $C6PcH_2$ as well as at those between P3HT and PCBM and between $C6PcH_2$ and PCBM. Actually, we confirmed photoinduced charge transfer in the P3HT:$C6PcH_2$ bulk heterojunction solar cell without PCBM, which would be reported elsewhere.

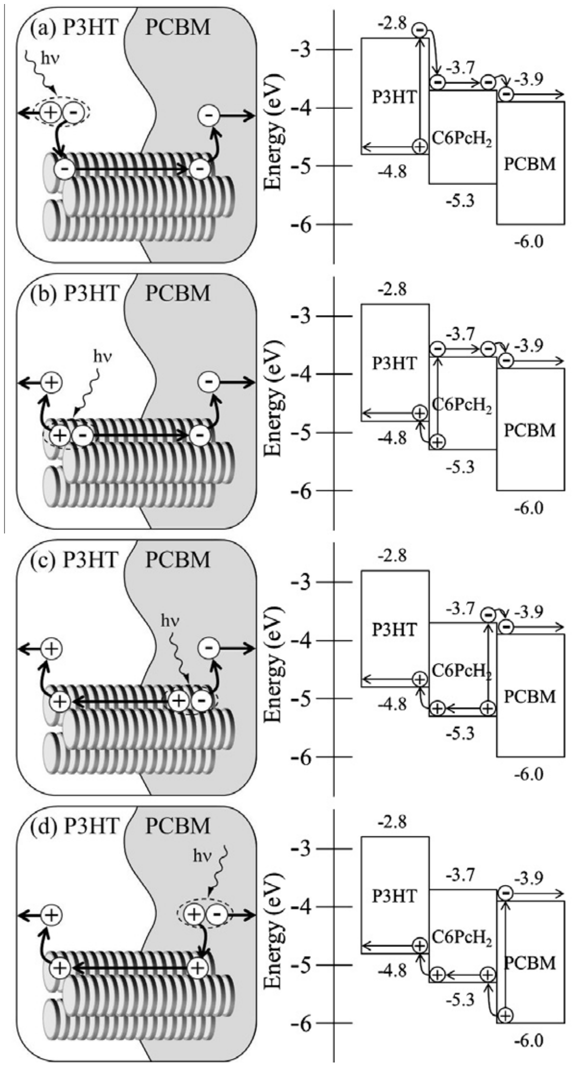

Fig. 5. Schematic diagrams of charge generation and transfer in the solar cells with P3HT:$C6PcH_2$:PCBM bulk heterojunction active layer (left side) and energy diagrams of P3HT, $C6PcH_2$ and PCBM in the solar cells (right side). (a) and (b) indicate the charge separation between P3HT and $C6PcH_2$ in the case of excitation at P3HT and $C6PcH_2$, respectively. (c) and (d) indicate the charge separation between $C6PcH_2$ and PCBM in the case of excitation at $C6PcH_2$ and PCBM, respectively.

In the case of exciting P3HT, $C6PcH_2$ accepts electrons from P3HT and transfers electrons through the columnar structures to PCBM (Fig. 5(a)). In the case of exciting $C6PcH_2$, $C6PcH_2$ donates holes to P3HT and transfers the remaining electrons through the columnar structures to PCBM (Fig. 5(b)). Photoinduced charge transfer also occurs between P3HT and PCBM. The area of the interface between P3HT and PCBM should decrease upon the addition of $C6PcH_2$, but the doped $C6PcH_2$ works as an acceptor and electron transport material. Therefore, the EQE at the absorption wavelength range of P3HT was maintained in spite of the decrease of the area of the interface between P3HT and PCBM by doping $C6PcH_2$. Moreover, a high EQE in the near-infrared region was also obtained as shown in Fig. 2.

A similar explanation can be adopted for the photoinduced charge transfer between $C6PcH_2$ and PCBM, which act as a donor and acceptor, respectively [18,19]. In the case of exciting $C6PcH_2$, $C6PcH_2$ donates electrons to PCBM and transfers the remaining holes through the columnar structure to P3HT (Fig. 5(c)). In the case of exciting PCBM, $C6PcH_2$ accepts and transports holes from PCBM and transfers holes to P3HT (Fig. 5(d)).

$C6PcH_2$ formed the columnar structure owing to its high self-organization properties, even in the bulk heterojunction film, and behaved as both a donor and an acceptor because of its high ambipolar properties. Therefore, it is considered that the long-wavelength sensitivity of the bulk heterojunction polymer solar cell could be improved without decreasing the donor–acceptor interface area.

4. Conclusion

We reported on the improvement of the long-wavelength sensitivity in P3HT:PCBM bulk heterojunction organic thin-film solar cells by the doping of $C6PcH_2$, which is a soluble phthalocyanine derivative exhibiting near-infrared absorption, into the P3HT:PCBM bulk heterojunction active layer. At the composition ratio of P3HT:$C6PcH_2$:PCBM = 10:3:10, the photosensitivity in the wavelength region from 650 to 800 nm was improved, and I_{sc} was enhanced from 8.6 to 12.1 mA/cm^2. As a result, the energy conversion efficiency was improved from 2.3% to 3.0%. It was found that the bulk heterojunction was composed of both highly ordered P3HT domains and hexagonal columnar structures of $C6PcH_2$ at the P3HT:$C6PcH_2$ composition ratio of 10:3 and that P3HT and $C6PcH_2$ underwent mutual microphase separation in the active layer. We have discussed the mechanisms of photoconversion in the bulk heterojunction organic thin-film solar cell based on the P3HT:$C6PcH_2$:PCBM active layer.

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