

Donor–Acceptor Molecule as the Acceptor for Polymer-Based Bulk Heterojunction Solar Cells

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We fabricated polymer-based bulk heterojunction (BHJ) solar cells using a donor–acceptor (D–A) molecule *N*-propyl-3,6-bis[2-(3-dicyanomethylene-5,5-dimethylcyclohex-1-enyl)vinyl]carbazole (PDHC) as the acceptor. The strong photoluminescence (PL) quenching of poly[2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylene vinylene] (MEH-PPV) when mixed with PDHC means that efficient charge transfer has happened between MEH-PPV and PDHC. We used MEH-PPV and poly (3-hexylthiophene) (P3HT) as the donors and PDHC as the acceptor to fabricate BHJ solar cells. The cells with the structure of ITO/PEDOT/polymer:PDHC/LiF/Al exhibited a power conversion efficiency (PCE) about 0.2% under 100 mW/cm² white light illumination. The open-circuit voltages (V_{oc} s) of 1.14 V of the cells based on MEH-PPV:PDHC and about 0.76 V of the cells based on P3HT:PDHC were achieved, which are higher than the V_{oc} s of the corresponding solar cells with phenyl-C-butyric acid methyl ester (PCBM) as the acceptor. In comparison with the cells based on PCBM, the relatively lower PCE of solar cells based on PDHC is due to its low electron mobility of $1.15 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ S}^{-1}$ estimated by the space charge limited current method.

1. Introduction

Polymer solar cells have been attracting great attention in recent years due to its offering a promising route to flexible, large-area, low-cost, and sustainable electricity generation.^{1–3} Bulk heterojunction (BHJ) solar cells based on intimate blends of donor and acceptor materials are of particular interest.^{4,5} During the past decade, researchers put great efforts on materials synthesis^{6–8} and optimization of active layers^{9,10} and device structure^{11–14} to enhance the efficiency of the BHJ solar cells. Recently the BHJ solar cells have achieved the power conversion efficiency (PCE) of about 5% for single-junction polymer solar cells^{6,15} and about 6.5% for double-junction cells.¹⁶ In most of the BHJ solar cells, the electron acceptor is fullerene derivatives such as mostly used phenyl-C-butyric acid methyl ester (PC₆₀BM and PC₇₀BM), since ultrafast charge transfer was found between conjugated polymer and C60 in 1992.¹⁷ Nonetheless, fullerene derivatives are nonideal materials for photovoltaic applications due to their weak absorption in the visible spectrum and their extremely deep-lying lowest-unoccupied molecular orbital (LUMO) levels, which results in lower open-circuit voltage (V_{oc}).¹⁸ Therefore it is important to search an acceptor material which can absorb photons in the solar spectrum efficiently and possesses higher-lying LUMO level than PCBM. On the basis of these demands, perylene diimide (PDI) derivatives,^{19–22} benzothiadiazole-containing small molecules²³ and some novel conjugated polymers^{24,25} have ever been used as the acceptor for organic and polymer BHJ solar cells. Their V_{oc} s were enhanced efficiently compared with the solar cells using PCBM as the acceptor.

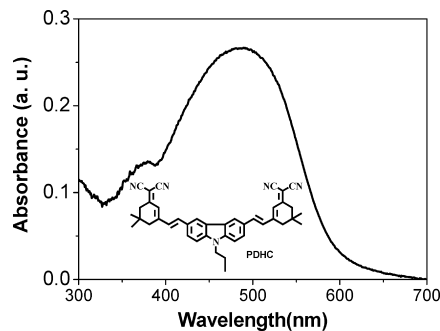


Figure 1. Chemical structure of the D–A molecule (PDHC) and its absorption in film.

Donor–acceptor (D–A) materials consist of strong electron-donating and electron-withdrawing units, which usually possess relatively low-lying LUMO levels and absorb toward long-wavelength from internal charge transfer (ICT) transition. Recently D–A molecules have been widely used as donors and PCBM as acceptor for solution-processable BHJ solar cells^{26–28} because of their advantages, including their easier synthesis and purification process than polymers, their longer wavelength absorption for matching the solar irradiation spectrum and the easier adjustment of the highest-occupied molecular orbital (HOMO) level for higher V_{oc} . However, few works used the D–A molecules as the acceptor for polymer-based solar cells.²⁹ Previously we used three D–A molecules as donors mixed with PCBM for BHJ solar cells.³⁰ Their photovoltaic performance was strongly dependent on the molecular structure, i.e., the donor unit and acceptor unit which determine their energy levels and absorption band.

Here we used the D–A molecule of *N*-propyl-3,6-bis[2-(3-dicyanomethylene-5,5-dimethylcyclohex-1-enyl)vinyl]carbazole (PDHC, Figure 1) as the acceptor for BHJ solar cells due to its

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relatively long wavelength absorption band (Figure 1) and low-lying LUMO level (-3.42 eV)³⁰ (higher-lying than the LUMO level of PCBM). The commonly used polymers of poly[2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylene vinylene] (MEH-PPV) and poly(3-hexylthiophene) (P3HT) were used as donors. The strong photoluminescence (PL) quenching of MEH-PPV when mixed with PDHC means efficient charge transfer has happened between MEH-PPV and PDHC. The solar cells with a structure of indium tin oxide (ITO)/PEDOT:PSS/polymer:PDHC/LiF/Al exhibit a V_{oc} of 1.14 V, a short-circuit current (J_{sc}) of 0.7 mA/cm², a fill factor (FF) of 0.27, and a PCE of 0.2% based on MEH-PPV:PDHC, while a V_{oc} of 0.74 V, a J_{sc} of 0.5 mA/cm², a FF of 0.40, and a PCE of about 0.14% based on P3HT:PCBM under 100mW/cm² white light illumination.

2. Experimental

MEH-PPV and P3HT were synthesized by our group. The D-A molecule (PDHC) was synthesized as reported before.³¹ PEDOT:PSS (CLEVIO S VP AI 4083) was purchased from H.C. Starck GmbH. Solvents of chloroform (CF) and chlorobenzene (CB) were purchased from Aldrich.

We prepared blend films of MEH-PPV:PDHC with different blend ratios by spin coating in order to investigate the PL quenching. Eight totally different blend ratios were prepared where the concentrations of PDHC were 0, 1, 2, 5, 10, 20, 50, and 100%. The PL measurement was done on a fluorescence spectrophotometer from Shimadzu.

As for device fabrication and measurement, ITO glass was cleaned by detergent, acetone and boiled in H₂O₂. A thin PEDOT:PSS (~40 nm, measured by Veeco DEKTAK 150 surface profilometer) layer was spin coated on cleaned ITO and then annealed at 120 °C for 20 min on a hot plate. Then the active layers MEH-PPV:PDHC (1:1 and 1:2, weight ratio) and P3HT:PDHC (1:1 and 1:2, weight ratio) were spin coated from CB solution and mixed solvents (CB:CF) solution. The thickness of MEH-PPV:PDHC is about 40 nm, and P3HT:PDHC is about 50 nm. For some devices, "solvent" annealing was done on P3HT:PDHC active layers for 20 min in closed container by heating CB it at 50 °C. Finally, the cathode of LiF(0.6nm)/Al(80nm) was thermally deposited to finish the device fabrication. The device area is about 5 mm². Afterward, some devices with P3HT:PDHC as active layer was annealed at 110 or 120 °C for 5 and 10 min before measurement. Current–voltage (J – V) characteristics were recorded using Keithley 2400 Source Meter in the dark and under 100 mW/cm² white light illumination (from Xeon light). The spectral response was recorded by SR830 lock-in amplifier under short circuit condition when devices were illuminated with a monochromatic light from a Xeon lamp. All fabrication and characterizations were performed in an ambient environment except thermal annealing was done in N₂ atmosphere.

3. Results and Discussion

3.1. PL Quenching. First, we investigate the PL quenching between donor and PDHC to check if efficient charge transfer happens or not, which is prerequisite for efficient photovoltaic devices. Because MEH-PPV gives strong emission while P3HT emits very weakly in film, we used MEH-PPV as the representative to investigate the PL quenching between MEH-PPV and PDHC. Eight blend films with different blend ratios were prepared where the concentrations of PDHC are 0, 1, 2, 5, 10, 20, 50, and 100%. Their thicknesses were controlled around 30–40 nm. Figure 2a shows the PL spectra of the blend films. Figure 2b is the PL intensity of blend films at 570 nm as a

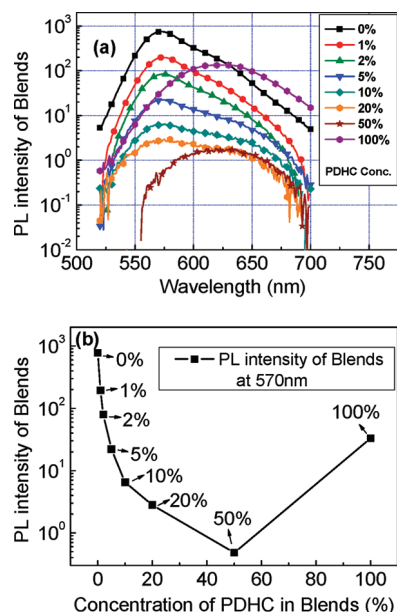


Figure 2. (a) PL spectra of the blend films where the concentrations of PDHC are 0, 1, 2, 5, 10, 20, 50, and 100%. (b) the PL intensity of blend films at 570 nm as a function of the concentration of PDHC.

function of the concentration of PDHC (data extracted from Figure 2a). It can be seen that the PL intensity was quenched very strongly as the concentration of PDHC increases. When the concentration of PDHC is 1, 10, and 50%, the PL intensity was quenched by about 4, 200, and 2000 times, respectively, compared with the pure MEH-PPV film. This result means efficient charge transfer happens between MEH-PPV and PDHC. Efficient photovoltaic performance of MEH-PPV:PDHC could be expected. P3HT possesses similar energy levels to MEH-PPV, therefore efficient charge transfer between P3HT and PDHC and efficient photovoltaic performance could also be expected.

3.2. Photovoltaic Properties. By using the D-A molecule PDHC as the acceptor, we fabricated BHJ solar cells using MEH-PPV and P3HT as donors separately. The structure of the solar cells is ITO/PEDOT:PSS/MEH-PPV (or P3HT):PDHC/LiF/Al. We optimized the active layers by trying different composition of donors and PDHC, solvent annealing, and thermal annealing on active layers and using different solvents for solution preparation and film forming of the active layers. In total, 11 types of solar cells (defined as devices 1–11) were fabricated with PDHC as the acceptor. For comparison, we fabricated reference solar cells with MEH-PPV and P3HT as the donors and PCBM as the acceptor (defined as device R1–R3). Their photovoltaic data are listed in Table 1.

Two solar cells were fabricated using MEH-PPV as the donor and PDHC as the acceptor where the blend ratio of MEH-PPV:PDHC is 1:1 and 1:2 (w/w). The two cells (devices 1 and 2 in Table 1) show similar performance, the V_{oc} of 1.14 V, the J_{sc} of about 0.68 mA/cm², the FF of about 0.25, and the PCE of about 0.19%. The PCE is about 20 times higher than that of solar cells based on pure MEH-PPV, which due to the efficient charge transfer between MEH-PPV and PDHC. The reference cell R1 based on MEH-PPV:PCBM exhibits a V_{oc} of 0.83 V, a J_{sc} of 4.42 mA/cm², a FF of 0.40, and a PCE of 1.46%. The higher V_{oc} of devices 1 and 2 (1.14 V) than device R1 and other reported results based on MEH-PPV:PCBM (typically about 0.8–0.85 V³²) is because the LUMO level of PDHC (-3.42 eV) is higher lying than that of PCBM (about -4.1 eV). The lower J_{sc} and FF than the counterparts of solar cells based on

TABLE 1: Photovoltaic Performance of Solar Cells with PDHC as the Acceptor Where the Active Layers Were Treated under Different Processing Conditions (Including Different Composition of Donor and PDHC, Annealing on Active Layer, and Using Different Film-Forming Solvents) (Devices 1–11) and the Reference Cells (R1–R3) with PCBM as the Acceptor under 100 mW/cm² White Light Illumination

device no.	donor (polymer)	blend ratio (D:A)	solvent	annealing	V_{oc} (V)	J_{sc} (mA/cm ²)	FF	PCE (%)
1	MEH-PPV	1:1	CB	N/A	1.14	0.68	0.25	0.19
2	MEH-PPV	1:2	CB	N/A	1.14	0.62	0.23	0.16
3	P3HT	1:1	CB	N/A	0.54	0.11	0.41	0.024
4	P3HT	1:1	CB	solvent ^b	0.66	0.024	0.21	0.0033
5	P3HT	1:1	CB	120 °C ^c	0.68	0.49	0.38	0.13
6	P3HT	1:2	CB	N/A	0.52	0.10	0.40	0.021
7	P3HT	1:2	CB	solvent ^b	0.68	0.032	0.19	0.004
8	P3HT	1:2	CB	120 °C ^c	0.74	0.40	0.35	0.11
9	P3HT	1:1	CB:CF ^a	N/A	0.56	0.26	0.38	0.054
10	P3HT	1:1	CB:CF ^a	110 °C ^c	0.76	0.49	0.30	0.11
11	P3HT	1:1	CB:CF ^a	110 °C ^d	0.78	0.66	0.27	0.14
R1 ^e	MEH-PPV	1:4	CB	N/A	0.83	4.42	0.40	1.46
R2 ^e	P3HT	1:1	CB	solvent ^b	0.54	6.58	0.58	2.06
R3 ^e	P3HT	1:1	CB	120 °C ^d	0.60	6.11	0.54	1.98

^a The blend ratio of mixed solvents is 3:1 (chlorobenzene:chloroform). ^b Solvent annealing was done in chlorobenzene atmosphere for 20 min in a closed container. ^c Thermally annealed for 5 min. ^d Thermally annealed for 10 min. ^e The reference cells R1–R3 with PCBM as the acceptor.

MEH-PPV:PCBM is possibly due to the lower electron mobility of PDHC than that of PCBM. The electron mobility of PDHC was estimated as shown behind.

The solar cells based on P3HT:PCBM blend films have been widely investigated, which were optimized by thermal annealing and “solvent” annealing because P3HT can self-organize under annealing to achieve a more ordered structure with higher hole mobility and absorbing more photons.^{9,10} The reference cells R2 and R3 based on P3HT:PCBM were optimized by thermal annealing and “solvent” annealing separately. Their PCEs are around 2% under white light illumination. As for the solar cells based on P3HT:PDHC, we also optimized them by thermal annealing and “solvent” annealing. In total, three groups of solar cells based on P3HT:PDHC were fabricated. The first group (group 1, devices 3–5) are solar cells with blend ratio of 1:1 (P3HT:PDHC) formed from CB solution. The second group (group 2, devices 6–8) are solar cells with blend ratio of 1:2 (P3HT:PDHC) formed from CB solution. The third group (group 3, devices 9–11) are solar cells with blend ratio of 1:1 (P3HT:PDHC) formed from CB:CF (3:1) solution. And each group of solar cells was annealed under different conditions, including thermal annealing for different time and solvent annealing. The difference between group 1 and group 2 is the blend ratio of P3HT:PDHC, i.e., 1:1 for group 1 and 1:2 for group 2. The photovoltaic performance of the two groups is similar when they were annealed under the same conditions. This result is similar to that of device 1 and 2 based on MEH-PPV:PDHC, which means that the photovoltaic performance of the solar cells is not dependent on the composition from 1:1 to 1:2 too much. By comparison of the three solar cells in group 1 (devices 3–5): device 3 with nonannealed active layer, device 4 with active layer processed by solvent annealing, and device 5 with active layer thermal annealed for 5 min at 120 °C, we found that device 3 exhibited a V_{oc} of 0.54 V, a J_{sc} of 0.11 mA/cm², a FF of 0.41, and a PCE of 0.024%, which could be enhanced efficiently to a higher V_{oc} of 0.68 V, a higher J_{sc} of 0.49 mA/cm², a FF of 0.38, and a PCE of 0.13% in device 5 after thermal annealing for 5 min at 120 °C, because P3HT self-organized and achieved a more ordered structure with higher hole mobility and absorbing more photons under thermal annealing.⁹ Since the photovoltaic performance of P3HT:PCBM blend system could be enhanced by solvent annealing,¹⁰ here we tried solvent annealing on P3HT:PDHC blend film (device 4). To our surprise, the PCE of the

solvent annealed device 4 became much poorer (about 8 times lower than the device 3 without post production treatment) after treatment by solvent annealing in CB atmosphere. This is due to the poor solubility of PDHC in CB but good solubility of P3HT in CB, which results in the aggregate of P3HT and large-scale phase segregation between P3HT and PDHC as observed under optical microscopy of surface profiler (not shown here), accordingly the photovoltaic performance decreased.

Because of the relatively poor solubility of PDHC in CB but good solubility in CF, we used blend solvents of CB:CF (3:1) for solving the mixed P3HT and PDHC. The solar cells fabricated from blend solvents were defined as group 3 (devices 9–11). The blend ratio of P3HT:PDHC is 1:1. Device 9 with nonannealed active layer exhibits a PCE of 0.054%, more than twice as high as the PCE of device 3, fabricated with pure CB solvent. After thermally annealed at 110 °C for 5 min (device 10) and 10 min (device 11), the V_{oc} was enhanced to be 0.76 V, J_{sc} was enhanced to be twice that of device 9, and PCE was enhanced to be about 0.14%. The photovoltaic performance of device 10 and 11 fabricated from mixed solvents after treated by thermal annealing is quite similar to that of device 5 and 8 fabricated from pure CB solvent.

Parts a and b of Figure 3 show the J – V curves of device 1 (MEH-PPV:PDHC, 1:1) and device 5 (P3HT:PDHC, 1:1, with thermal annealing) under 100 mW/cm² white light illumination and in the dark. The J – V curve of device 1 in the dark exhibits a high rectification of 10^3 at ± 2 V, and device 3 exhibits a higher rectification of 3×10^3 , which means devices have high quality, few drawbacks or pin holes. Figure 3c shows the equivalent quantum efficiency (EQE) of devices 1 and 5 and light intensity spectra of the Xeon lamp we used for J – V measurement under illumination. The light intensity spectra of Xeon lamp is quite mismatched with EQEs of devices. Therefore the PCE of the devices can be enhanced if illuminated under AM1.5 simulated solar light, which the EQEs of devices are quite matched with. Even so, the PCEs of device based on PDHC as acceptor are still much lower than those of devices based on PCBM as acceptor, which could probably due to the lower electron mobility of PDHC than that of PCBM.

3.3. Mobility. To investigate the reason why the photovoltaic performance of devices based on P3HT (or MEH-PPV):PDHC is much lower than that of devices based on P3HT (or MEH-PPV):PCBM, we estimated the electron mobility of PDHC

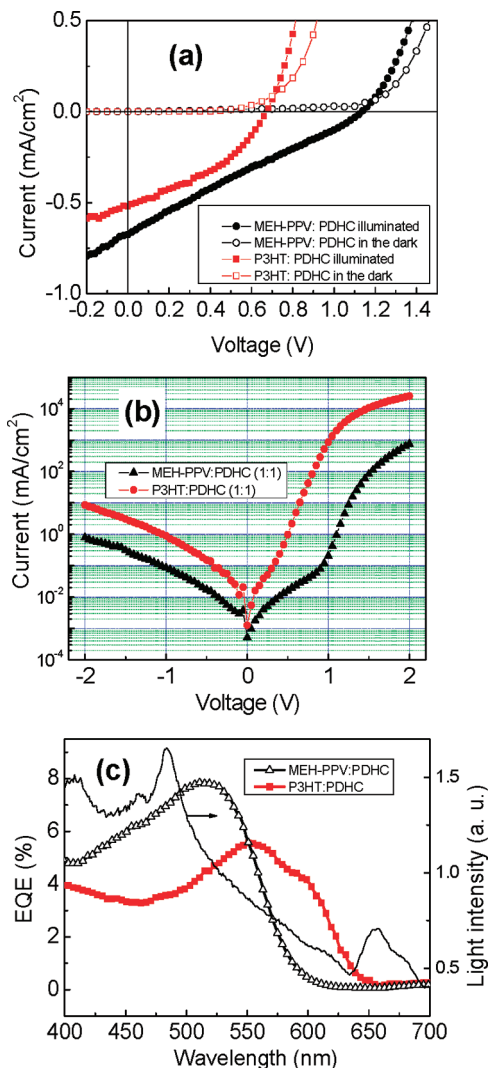


Figure 3. (a) J - V curves of ITO/PEDOT/MEH-PPV:PDHC (1:1)/LiF/Al (device 1) and ITO/PEDOT/P3HT:PDHC (1:1)/LiF/Al (thermally annealed, device 5) in the dark and under 100 mW/cm² white light illumination. (b) J - V curves of device 1 and 5 in the dark in semilog axis. (c) EQEs of devices 1 and 5. The solid line is the Xeon lamp spectra we used for J - V measurement under illumination.

approximately by the space charge limited current (SCLC) method.³³ Devices with a structure of ITO/PDHC/LiF/Al were fabricated. Their J - V curves in the dark are shown in Figure 4. It can be seen that in the low-voltage area (from 0.01 to 0.60 V) $\log J$ is linearly dependent on $\log V$ with a slope of 1. In the relatively high-voltage area (from 0.6 to 1 V) $\log J$ can also be fitted to be linearly dependent on $\log V$ and the slope is 2. The J - V in this high-voltage area is corresponding to the SCLC behavior. The work function of ITO is about -4.3 to -4.4 eV, and that of LiF/Al is about -3.7 eV. The HOMO level and LUMO level of PDHC are -5.47 and -3.42 eV, respectively (the alignment of energy level shown in the inset of Figure 4). Therefore the device can be regarded as electron-dominating device. We estimated the electron mobility of PDHC according to the equation

$$J = \frac{9}{8} \epsilon_r \epsilon_0 \mu_e \frac{(V - V_{bi})^2}{L^3}$$

Where V is the applied voltage, J is the current density, ϵ_r and ϵ_0 are the relative dielectric constant and the permittivity of the free space (8.85×10^{-12} F/m), respectively, μ_e is electron

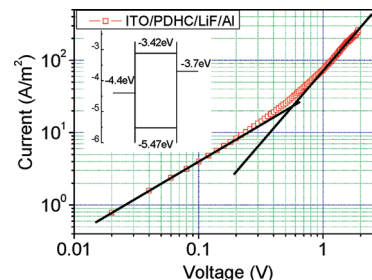


Figure 4. J - V curve in the dark of an ITO/PDHC/LiF/Al device in log axis for estimating the electron mobility of PDHC. The thickness of PDHC is around 45 nm. The open rectangle symbols are the experimental data. The solid line from 0.01 to 0.60 V means $\log J$ is fitted linearly dependent on $\log V$ with a slope of 1. For the solid line from 0.6 to 1 V $\log J$ is fitted linearly dependent on $\log V$ with a slope of 2 (SCLC area). The inset is the alignment of the energy levels of PDHC and electrodes.

mobility and L is the thickness of the organic layer. Here we fit ϵ_r to be 3. V_{bi} is fitted to be about 0.6 V, which is determined by the difference between the work functions of cathode and anode. Then the electron mobility of PDHC is estimated to be about 1.15×10^{-5} cm² V⁻¹ S⁻¹, which is a quite high electron mobility but about two orders lower than the μ_e of PCBM, about 2×10^{-3} cm² V⁻¹ S⁻¹.³⁴ That is why the solar cells based on PDHC as the acceptor performed poorer than those based on PCBM as the acceptor.

Conclusion

In this work, we used a D-A molecule as the acceptor for BHJ solar cells. The cells exhibit higher V_{oc} s than the corresponding cells with PCBM as the acceptor due to the higher-lying LUMO energy level of the D-A molecule. The PCEs of the cells based on MEH-PPV:PDHC are around 0.2% and P3HT:PDHC are around 0.13%, which is due to the lower electron mobility of the D-A molecule than PCBM. This work will draw researchers' attention to use the D-A molecules (now widely used as donors) with higher electron mobility as acceptors for efficient BHJ solar cells.

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