Effect of 1,8-Diiodooctane Content on the Performance of P$_3$HT:PC$_{61}$BM Bulk Heterojunction Photodetectors

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Abstract A series of polymer photodetectors with device configuration of ITO/PEDOT:PSS/P$_3$HT:PC$_{61}$BM/C$_{60}$/Al were prepared by using P$_3$HT as the donor material and PC$_{61}$BM as the acceptor material. By regulating the content of 1,8-diiodooctane (DIO) (W/V: 1%, 3%, 5%) as a processing additive, the morphology of the active layer can be greatly improved. With C$_{60}$ as the hole blocking layer, the dark current density of the device can be reduced by about an order of magnitude. When employing 3% DIO (W/V) in the active layer processing, the photodetectors present the best performance, and the detectivity of the device is 1.52×10$^{12}$ Jones at 540 nm under a bias of −0.1 V. Moreover, it also has a wider linear dynamic range of 60 dB as well as faster response speed ($\tau_r$/$\tau_f$=0.53/0.71 $\mu$s) than those of devices with other content of DIO additives.

Keywords Polymer photodetector; Configuration; 1,8-Diiodooctane; Morphology

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INTRODUCTION

Photovoltaic device is a solid-state electrical device made of various semiconductive materials that can convert optical signals into electrical signals. They are widely used in solar energy generation, industrial inspection, military guidance and other fields.$^{[1−4]}$ Organic photovoltaic materials have been developed rapidly in recent years due to their tunable bandgap, low cost, light weight, flexibility and solution processability for large area fabrication.$^{[5−8]}$

Photodetector is an important kind of photovoltaic device. The structure of an organic photodetector (OPD) usually consists of an anode, a hole transport layer, an active layer, a hole blocking layer and a cathode. To improve the performance of photodetector, researchers focus on the study of the transport layer, the active layer, and even the electrode.$^{[9−12]}$ However, the active layer is always the most critical part of an organic photodetector, since most of the optoelectronic processes, such as photon absorption, exciton formation, exciton diffusion, exciton separation, and carrier transmission, occur in this layer. At present, solution processing is still the common method for preparing bulk heterojunction (BHJ) active layer. That is, the donor material and the acceptor material are mixed at a certain ratio and an active layer of the device is fabricated with spin-coating, blade-coating, ink-jet printing and other solution processing methods. Although the solution processing process is simple, the ratio of donor and acceptor materials, the selection and content of additives, the rotate speed of spin coating, and the type of solvents cannot be ignored for the great performance of the device.$^{[13−15]}$

Chen et al. prepared efficient P3HT:PC$_{61}$BM solar cells by employing trichlorobenzene (TCB) as the processing additives. The authors found that the orientation of poly(3-hexylthiophene-2,5-diyl) (P3HT) can be induced by TCB. By adjusting the processing content of TCB, the power conversion efficiency (PCE) of the device can be improved from 3.84% to 4.74%.$^{[16]}$ 1,8-Diiodooctane (DIO) is one of the common additives which can effectively adjust the surface morphology of the active layer. The DIO additive can increase the solubility of the fullerene material in the processing solution, allowing the active layer to form an ideal interpenetrating network structure, which facilitates the diffusion, transfer and separation of excitons.$^{[17]}$ Qi et al. synthesized a diketopyrrolopyrrole-based polymer (PDPP) and prepared a BHJ photodetector by blending PDPP and [6,6]-phenylC$_{71}$-butyric acid methyl ester (PC$_{71}$BM) as the active layer. They investigated the device performance with or without DIO additives. The result shows that the responsivity of the device is improved by nearly 7 times, and the dark current is also reduced by an order of mag-
magnitude after adding DIO additives.\textsuperscript{[18]}

P3HT is very popular in the field of photovoltaic devices due to its simple chemical structure and low synthesis cost. In just ten years, the PCE of P3HT:PCBM based organic solar cells has achieved a breakthrough from 2.8\% to 7.4\%.\textsuperscript{[19−22]}

However, this material has not been fully studied in the field of organic photodetectors, and the characterization of device performance is usually only focused on the improvement of responsivity ($R$) and detectivity ($D^*$).\textsuperscript{[23−26]} It should be noted that linear dynamic range (LDR) and response time (the rising time $\tau_r$ and the falling time $\tau_f$) are also two important parameters which determine whether the organic photodetectors can be used. Compared with the LDR of inorganic photodetectors (66 dB for InGaAs, 120 dB for Si), the LDR of organic photodetectors is often less than 60 dB. Due to the unbalanced carrier transfer caused by traps in the active layer, the response speed of organic photodetectors often falls behind inorganic photodetectors.\textsuperscript{[27]}

Therefore, in recent years, many researchers focus on improving the comprehensive performance of the organic photodetectors.\textsuperscript{[28−30]}

Herein, we report the effects of the content of DIO additives and C$_{60}$ layer on the performance of the devices. Polymer photodetectors (PPDs) with a forward configuration of ITO/PEDOT:PSS/P3HT:PC$_{61}$BM/C$_{60}$/Al were fabricated. We found that high detectivity of 1.52×10\textsuperscript{12} Jones, wide linear dynamic range of 60 dB, and fast response speed ($\tau_r/\tau_f=0.53/0.71$ µs) can be achieved through device configuration and morphology of the active layer.

**EXPERIMENTAL**

**Materials**

Poly(3-hexylthiophene-2,5-diyl) (P3HT) was purchased from Rieke Metals. (6,6)-Phenyl-C$_{60}$-butyricacidmethylester (PC$_{61}$BM) was obtained from Xi’an Polymer Light Technology Corp. (Xi’an, China). Poly(3,4-ethylenedioxy thiophene):poly(styrene sulfonate) (PEDOT:PSS, Clevious Al 4083) was purchased from Heraeus. 1,8-Dioiodooctane (DIO) was obtained from Alfa Aesar. C$_{60}$ was purchased from Energy Chemical (Shanghai, China). Other chemicals were of analytical grade and used without further purification.

**Fabrication of Organic Photodetectors**

A mixture of P3HT and PC$_{61}$BM in a weight ratio of 1:1.5 were dissolved in solvent (chloroform containing 1\%, 3\%, 5\% (V/V) DIO) with a total concentration of 20 mg/mL and stirred under nitrogen atmosphere in a glove box at 55°C for more than 8 h. For device fabrication, patterned indium tin oxide (ITO) glass substrates were cleaned by ultrasonic treatment in detergent, deionized water, acetone, and isopropyl alcohol, sequentially, then dried in an oven at 120 °C for 1 h, and treated with O$_2$ and Ar plasma for 20 min. PEDOT:PSS was filtered through a 0.45 µm poly(tetrafluoroethylene) (PTFE) filter, and then spin-coated on the ITO glass substrates at 4000 r/min for 60 s, followed by annealing at 120 °C for 20 min, forming a film with a thickness of about 40 nm (Fig. S1 in the electronic supplementary information, ESI). Then the active layer solution was filtered through a 0.45 µm PTFE filter, and then spin-coated on the PEDOT:PSS film at 1500 r/min for 60 s, forming a film with a thickness of about 150 nm (Fig. S2 in ESI). 10 nm of C$_{60}$ and 100 nm of Al were successively deposited on the active layer at a vacuum of 4×10\textsuperscript{−4} Pa by thermal evaporation using a shadow mask (the active area of every device was 16 mm\textsuperscript{2}) on QHV-R20 metal/organic evaporation equipment from Shenyang Qihui Vacuum Technology Co., Ltd.

**Characterizations**

Responsivity ($R$), dark current density ($J_d$), linear dynamic range (LDR) and response time ($\tau_r/\tau_f$) measurements were conducted under ambient conditions in air using a detector test system from Jilin Yuanda Optical Inspection Technology Co., Ltd. A calibrated silicon photodetector (Thorlabs’ FDS010) was used as a reference to measure polymer photodetectors. The certificate of calibration of the standard detector is provided by Thorlabs GmbH manufacturer (Fig. S3 in ESI). The maximum light source power for linear dynamic range test is 0.37 W, and the minimum light source power was 9.78×10\textsuperscript{−14} W. The response time of the photodetector was obtained by measuring the output curve and data of the detector’s electrical signal with pulse radiation (640 nm monochromatic light). The UV-Vis absorption spectrum was tested on a Shimadzu UV3600 spectrophotometer. The film thickness of the active layer was measured by a KLA Tencor D-100 step meter. Atomic force microscopy images were obtained by Xe-70 Park System.

**RESULTS AND DISCUSSION**

**Configuration of Photodetectors**

To evaluate the photodetector performance of the solution processing with different contents of DIO additive (V/V: 1\%, 3\%,
5%), the devices with a configuration of ITO/PEDOT:PSS/active layer/C60/Al were fabricated (Fig. 1). The lowest unoccupied molecular orbital (LUMO) energy level between P3HT and PC61BM is 1.0 eV (Fig. 2), which is much higher than 0.3 eV. Such a large offset is beneficial to exciton separation. A thin layer of C60 as hole blocking layer (HBL) can prevent holes entering the active layer from the Al cathode, which avoids the recombination of electrons and holes. In order to explore the influence of C60 on the photodetector, we prepared a device without C60 for comparison. Four polymer photodetectors (PPD-1, PPD-2, PPD-3 and PPD-4) were obtained by different device configurations or solution processing conditions (Table 1).

**Responsivity and Dark Current Density**

Responsivity (\(R\)) and dark current density (\(J_d\)) are two basic parameters for characterizations of photodetector devices. \(R\) is the photocurrent (\(I_{ph}\)) divided by the incident light power (\(P_{light}\)) at a given wavelength (Eq. 1), which is a physical quantity describing the photoelectric conversion capability of the device,

\[
R = \frac{I_{ph}}{P_{light}}
\]

The unit of \(I_{ph}\) and \(P_{light}\) is A and W, respectively, so the unit of \(R\) is A/W. The responsivity of the four devices under a bias of \(-0.1\) V are displayed in Fig. 3(a). The spectral response range of the PPDs is 300–800 nm, covering visible light and part of the ultraviolet light, due to the complementary spectral absorption of P3HT and PC61BM. The absorption range of P3HT is in the visible light region, and PC61BM mainly absorbs ultraviolet light.

The absorption spectrum can be broadened after these two materials are mixed (Fig. S4 in ESI). The polymer photodetector PPD-2 (with 3 vol% DIO) achieved a maximum responsivity \((R_{max})\) of 152.84 mA/W at 540 nm. While the \(R_{max}\) of PPD-1 and PPD-3 based on 1 vol% and 5 vol% DIO is 145.69 and 132.72 mA/W, respectively. The external quantum efficiency (EQE) can be calculated by \(R\) as Eq. (2):

\[
EQE = 1240 \times \frac{R}{\lambda}
\]

where \(\lambda\) is the wavelength, \(R\) is the responsivity at a specific wavelength. The \(R\) and EQE curves of the PPDs under different bias are shown in Figs. S5 and S6 (in ESI). It can be seen that the photoelectric conversion capability of the three devices increases with the increase of the bias. Dark current density (\(J_d\)) refers to the current per unit area in the photodetector at a given bias range without light. As displayed in Fig. 3(b) and Fig. S7 (in ESI), the \(J_d\) of the three devices containing C60 layer (PPD-1, PPD-2 and PPD-3) is one order of magnitude lower than that of the device without C60 layer (PPD-4). This indicates that the C60 layer can prevent holes from entering the active layer, which effectively prevents bimolecular recombination, thereby reducing the dark current of the device and increasing the noise resistance of the device.

**Specific Detectivity and Linear Dynamic Range**

The specific detectivity (\(D^*\)) is a key figure-of-merit to

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**Table 1**: Device configuration and solution processing conditions of polymer photodetectors.

| Device | Configuration | Additive | Solvent | Spin-coating speed (r/min) |
|--------|---------------|----------|---------|---------------------------|
| PPD-1  | ITO/PEDOT:PSS/P3HT:PC61BM/C60/Al | 1 vol% DIO | Chloroform | 1500 |
| PPD-2  | ITO/PEDOT:PSS/P3HT:PC61BM/C60/Al | 3 vol% DIO | Chloroform | 1500 |
| PPD-3  | ITO/PEDOT:PSS/P3HT:PC61BM/C60/Al | 5 vol% DIO | Chloroform | 1500 |
| PPD-4  | ITO/PEDOT:PSS/P3HT:PC61BM/Al | 3 vol% DIO | Chloroform | 1500 |

**Fig. 3** (a) Responsivity (−0.1 V), (b) dark current density (−0.1 V) and responsivity (−0.1 V, 540 nm) of the polymer photodetectors.
characterize the performance of a photodetector. If the shot noise generated by dark current is considered to be the main cause of the noise signal,[35] $D$ can be calculated as Eq. (3):

$$D = \frac{R}{(2qJ_d)^{1/2}}$$  \hspace{1cm} (3)

where $q$ is the elementary charge ($1.6\times10^{-19}$ C), $R$ is the responsivity and $J_d$ is dark current density. The unit of $D$ is cm·Hz$^{1/2}$·W$^{-1}$, which is also recorded as Jones in the field of photodetectors. Fig. 4(a) shows the specific detectivity ($D^*$) of these four devices. Due to the smaller dark current density (Table 2), the devices with C$_{60}$ layer PPD-1, PPD-2 and PPD-3 present higher specific detectivity of $1.43\times10^{12}$, $1.52\times10^{12}$, and $1.20\times10^{12}$ Jones, respectively. The device PPD-2 has the lowest dark current density, so the calculated specific detectivity is the highest. The $D^*$ of the device PPD-2 with 3 vol% DIO is higher than $10^{11}$ Jones in the range of 300–710 nm under $-0.1$ V bias. The detectivity of PPD-1, PPD-2 and PPD-3 under $-1.0$ V or $-2.0$ V is relatively lower than that of $-0.1$ V (Fig. S8 in ESI), because large bias makes the dark current density increase.

The linear dynamic range (LDR) describes the range of light intensity that a photodetector can detect. It is measured by recording the linear change of photocurrent with light intensity.[36] The LDR can be expressed as Eq. (4):

$$\text{LDR} = 20 \log \left( \frac{J_{\text{max}}}{J_{\text{min}}} \right)$$  \hspace{1cm} (4)

where $J_{\text{max}}$ and $J_{\text{min}}$ are the corresponding photocurrent measured at the maximum and minimum light intensity without deviating from the linear relationship between photocurrent and light intensity, respectively.[37] The LDR curves of the polymer photodetectors are shown in Fig. 4(b) and Fig. S9 (in ESI). It can be seen that the device PPD-2 with 3 vol% DIO has the widest LDR of 60 dB, much wider than the LDR values of other three devices (PPD-1, PPD-3 and PPD-4) of 49, 40 and 46 dB, respectively. The photocurrent density of the PPD-2 decreased linearly from $4.63\times10^{-3}$ mA/cm$^2$ to $5.52\times10^{-6}$ mA/cm$^2$ when decreasing the light intensity from 0.35 mW/cm$^2$ to 1.02×10$^{-4}$ mW/cm$^2$ (Fig. 4b).

### Transient Photocurrent Response

The transient photocurrent response is also a crucial parameter for photodetector. It represents the speed at which the photodetector responds to the light signal stimulation. From the inside of the device, the transient response time describes the generation and transport time of electrons and holes in the active layer.[38] The transient photocurrent response of a photodetector is usually evaluated by the rising time ($\tau_f$) and the falling time ($\tau_r$). $\tau_f$ represents the time for the photocurrent of the photodetector to rise from 10% to 90% after pulsed optical signal excitation. $\tau_r$ is the time for the photocurrent of the photodetector to fall from 90% to 10% after the pulsed optical signal disappears.[39] Fast response speed reflects the efficient transfer of charge from the internal circuit to the external circuit.

Fig. 5(a) displays the transient photocurrent response curves of the device PPD-1, PPD-2 and PPD-3 chopped at a frequency of 4000 Hz. It can be seen that PPD-2 exhibits the fastest response speed. The $\tau_f$ and $\tau_r$ of the device PPD-2 are 0.53 and 0.71 $\mu$s, respectively. The response speed of the device PPD-3 is slower than that of PPD-2 ($\tau_f/\tau_r = 0.54/1.25 \mu$s). However, the device PPD-1 with 1 vol% DIO additive shows a longer $\tau_r$ of 0.89 $\mu$s and an obvious tailing $\tau_f$ of 1.42 $\mu$s, which is twice of that of PPD-2. It can be attributed to the large number of charges trapped on the donor and acceptor interface. The unbalanced carrier transport promotes a bimolecular recombination which leads to the photocurrent tail.[40] When the chopper frequency is increased to 8000 Hz, all the three devices show a fast response speed (Fig. 5b), which may be due to the fact that the time resolution of the equipment has no influence on the response of the photodetectors.[41] The $\tau_f$ and $\tau_r$ of the devices are all below 1 $\mu$s, which can meet the requirements of light detection in various fields.

### Film Morphology

It is well known that the performance of optoelectronic devices is greatly affected by the morphology of the active layer.[42,43] Appropriate nano-scale domain facilitates the diffusion and separation of excitons and the transport of carriers, which is critical to the optimization of device. Atomic force microscope (AFM) was used to characterize the morphology of the BHJ active layers with different DIO additive contents. Figs. 6(a), 6(b) and 6(c) display AFM topography of the active layers with DIO.

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**Fig. 4** (a) Specific detectivity ($-0.1$ V) and (b) liner dynamic range of the polymer photodetectors.

**Table 2** Characteristics of PPDs under $-0.1$ V bias.

| Devices | $R_{\text{max}}$ ($\text{mA/W}$) | $J_{\text{max}}$ ($\text{A/cm}^2$) | $D_{\text{max}}^*$ (Jones) | LDR ($\text{dB}$) |
|---------|-----------------|-----------------|-----------------|-----------------|
| PPD-1   | 145.69          | 3.26×10$^{-2}$  | 1.43×10$^{12}$  | 49              |
| PPD-2   | 152.84          | 3.16×10$^{-8}$  | 1.52×10$^{12}$  | 60              |
| PPD-3   | 132.72          | 3.81×10$^{-8}$  | 1.20×10$^{12}$  | 40              |
| PPD-4   | 149.94          | 1.52×10$^{-7}$  | 6.03×10$^{11}$  | 46              |

*a Responsivity ($-0.1$ V, 540 nm); $b$ Dark current density ($-0.1$ V); $c$ Specific detectivity ($-0.1$ V, 540 nm); $d$ Linear dynamic range of the polymer photodetectors.

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content of 1 vol%, 3 vol% and 5 vol%, respectively. The active layer with 5 vol% DIO shows obviously large particles (~35 nm). This rough surface reduces the efficiency of carrier transport, which is an important reason for its low responsivity. The morphology of the 3 vol% DIO based active layer was improved significantly, and the surface of the film formed proper topography of 10−20 nm, which promotes the transmission of excitons and avoids the recombination of excitons. Therefore, the performance of the device PPD-2 is the best. The surface of the active layer based on 1 vol% DIO is quite smoother than those of the former two films. This film prevents the donor and acceptor materials from forming a nano-scale interpenetrating network structure,\cite{44} which results in the ineffective separation of excitons, low responsivity and high dark current density of the corresponding device. The root mean square roughness (RMS) of the three active layers with 1 vol%, 3 vol% and 5 vol% DIO are calculated as 3.4, 4.4 and 7.3 nm, respectively. According to the performance of the three devices and the morphology of the active layers, it can be summarized that tens of nano-scale domain of the active layer is favorable for good photovoltaic.

Fig. 5  Transient photocurrent response of the polymer photodetectors chopped at a frequency of (a) 4000 Hz and (b) 8000 Hz.

Fig. 6  AFM topography images: (a) 1 vol% DIO, (b) 3 vol% DIO and (c) 5 vol% DIO; Phase images: (d) 1 vol% DIO, (e) 3 vol% DIO, (f) 5 vol% DIO; 3D topography images: (g) 1 vol% DIO, (h) 3 vol% DIO, (i) 5 vol% DIO of the active layer based on P3HT:PC61BM (2.5 μm × 2.5 μm).

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response. Figs. 6(d), 6(e) and 6(f) show the phase diagrams of the three active layers, and the BHJ film with 3 vol% DIO exhibits moderate phase separation and domain sizes. The difference between the three active layers can be seen more intuitively from the 3D topography (Figs. 6g, 6h and 6i). The film with 5 vol% DIO has uneven morphology in 0−40 nm. On the contrary, the film with 1 vol% DIO remains smooth at 0−15 nm. The optimal film with 3 vol% DIO has a proper topography in 0−25 nm, which tends to form an interpenetrating network structure between the donor and acceptor materials. The results indicate that fine control of the active layer morphology is an important method for improving the performance of the device.

CONCLUSIONS

The bulk heterojunction photodetectors based on P3HT and PC_{61}BM were fabricated. By appropriately regulating the content of DIO additives, the active layer with favorable morphology can be obtained. The device PDD-2 containing 3 vol% DIO exhibits the best performance. The specific detectivity of the device is 1.52×10^{9} Jones at 540 nm under 3 vol% DIO exhibits the best performance. The specific detectivity of the device is 1.52×10^{12} Jones at 540 nm under 0.1 V, the linear dynamic range is 60 dB, and the response speed is also fast (r/τ<0.03/0.73 μs). This work demonstrates that high detectivity, wide linear dynamic range and fast response speed can be achieved through some subtle changes in solvent processing.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at http://doi.org/10.1007/s10118-021-2548-5

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