

A new oligobenzodithiophene end-capped with 3-ethyl-rhodanine groups for organic solar cells with high open-circuit voltage

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A new solution-processable small-molecule donor material, named DRBDT₃, comprised of oligobenzo[1,2-b:4,5-b'] dithiophene as the backbone and 3-ethyl-rhodanine as the end-capped group has been developed and synthesized for application in organic photovoltaic cells. The oligobenzodithiophene derivative exhibits absorption band from 300 to 640 nm. The film of DRBDT₃ shows highly long-range ordering assembly and high mobility of $1.1 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. The new molecule shows a deep highest-occupied molecular orbital energy level. The device based on DRBDT₃ as the donor and PC₇₁BM as the acceptor exhibits a power conversion efficiency of 4.09% with a high open-circuit voltage of 0.99 V under AM.1.5G illumination (100 mWcm⁻²).

Keywords: small molecule, organic solar cell, oligobenzodithiophene, high open-circuit voltage

1 Introduction

Organic photovoltaic cells (OPVs) have attracted an increasing amount of attention during the past decade, due to their advantages of solution processability, light weight, low cost, and potential in the fabrication of flexible devices [1-9]. Many efforts have been made to improve the power conversion efficiency (PCE) of polymer-based OPVs (P-OPVs) [10-15]. PCEs over 9% have been achieved [16-21] in the past few years. Compared to polymer materials, small molecules exhibit competitive advantages such as well-defined structure and therefore less batch-to-batch variation, easier band structure control, etc. [22-31]. To date, solution-processed small-molecule-based OPVs (SM-OPVs) have achieved PCEs higher than 8% [32-35] through design and synthesis of new small-molecule materials and optimization of device fabrication [36-41].

Benzo[1,2-b:4,5-b'] dithiophene (BDT) as an electron-donating unit has been widely used in P-OPVs

[3,42-45]. The symmetric and plain conjugated structure could facilitate the formation of π - π stacking. Devices based on BDT-based polymer are able to afford high PCEs of more than 9% [16, 17]. A series of BDT-based polymers including homopolymers and copolymers has been reported by Yang's group [40]. Among these BDT-based polymers, BDT-based homopolymer shows the deepest highest-occupied molecular orbital (HOMO) energy level, which could lead to a high V_{oc} in OPV devices because open circuit voltage (V_{oc}) mainly depends on the difference between the HOMO energy level of the donor material and the lowest unoccupied molecular orbital (LUMO) energy level of the acceptor material [5, 46]. John P. Ferraris *et al.* reported a BDT-based homopolymer, named O-PBDT, which exhibits a PCE of 1.56% with a high V_{oc} of 0.83 V and a relatively low short-circuit current density (J_{sc}) of 4.18 mA cm^{-2} [47]. The relatively low J_{sc} of O-PBDT is believed to be due to the narrow absorption band with absorption onset at 560 nm. To enhance the J_{sc} , it is essential to reduce the band gap, which is decided by the difference between the HOMO en-

was spin-coated (3000 rpm, ca. 40 nm thick) onto the ITO surface. After being baked at 150°C for 20 min, the substrates were transferred into an argon-filled glove box. Then the active layer was spin-coated from different ratios of donor (8 mg/mL):PC₇₁BM-blend chloroform solution at 1700 rpm for 20 s each. Next, ZnO particle suspension [49] was used to spin-coat the ZnO layer on top of the active layers. Thermal annealing was carried out on a digitally controlled hot plate at various temperatures after ZnO spin-coating in an argon-filled glove box. Finally, a 50 nm Al layer was deposited onto the ZnO film under high vacuum ($< 2 \times 10^{-4}$ Pa). The thickness of the ZnO and the active layer were measured using a Dektak 150 profilometer. The effective area of each cell was 4 mm², defined by masks for the solar-cell devices discussed in this work.

3 Results and discussion

3.1 Synthesis and thermal property

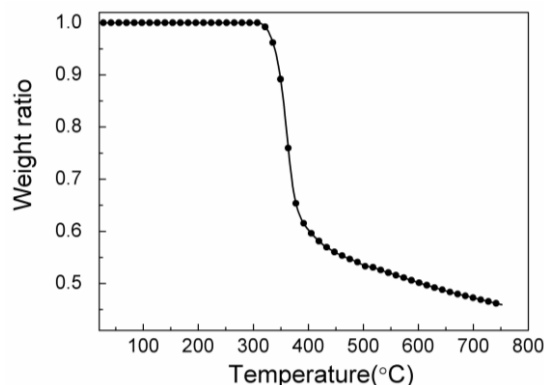


Figure 1 TGA curves of DRBDT₃ with a heating rate of 10°C/min under N₂ atmosphere.

As shown in Scheme 1, the intermediate DCHOBDT₃ was synthesized by a Stille coupling reaction between **2** and **3** under argon atmosphere in the presence of Pd(PPh₃)₄ as the catalyst for 24 h. The target molecule DRBDT₃ was then prepared by the Knoevenagel condensation of DCHOBDT₃ with 3-ethyl-rhodanine under argon atmosphere in the presence of piperidine as the catalyst for 12 h. DRBDT₃ shows good solubility in common solvents. The thermal property of DRBDT₃ was investigated by TGA. As shown in Figure 1, compound DRBDT₃ exhibited good thermal stability with 5% weight-loss temperature at 337°C under N₂ atmosphere.

3.2 Optical properties and electrochemical properties

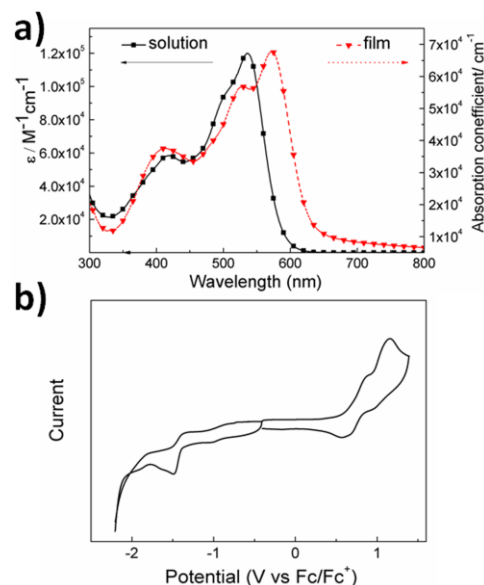


Figure 2 a) Absorption spectra of DRBDT₃ in chloroform solution and in as-cast film; b) Cyclic voltammogram of DRBDT₃ in a dichloromethane solution of 0.1 mol/L [NBPF₆] with a scan rate of 100 mV s⁻¹.

UV–vis absorption spectra of DRBDT₃ in diluted chloroform solution and a solid state are shown in Figure 2a. DRBDT₃ in the solution shows a maximum absorption peak at 536 nm with a maximal coefficient of 1.20×10^5 M⁻¹cm⁻¹. The DRBDT₃ film cast from CHCl₃ shows a red-shifted maximum absorption peak at 572 nm with a broader absorption band from 300 to 640 nm. The optical band gap of DRBDT₃ is 1.97 eV, estimated from the onset of the absorption spectra. A cyclic voltammogram was used to investigate the electrochemical properties of DRBDT₃. Ferrocene/ferrocenium of the (Fc/Fc⁺) redox couple (4.8 eV below the vacuum level) was used as the internal calibration. The HOMO and LUMO energy levels of DRBDT₃ were estimated based on the onset oxidation potential and the onset reduction potential of the redox curve as shown in Figure 2b; they are -5.34 and -3.40 eV, respectively. Note that, compared with O-PBDT, the new oligobenzodithiophene derivative end-capped with 3-ethyl-rhodanine indeed shows a decreasing LUMO energy level, which also demonstrates that the LUMO energy level of the A-D-A-structure small molecule mainly depends on the acceptor moiety. The electrochemical band gap of DRBDT₃ is 1.94 eV, which is consistent with the value of the optical band gap. The data for the optical and electrochemical properties are summarized in Table 1.

Table 1 Optical and electrochemical data of DRBDT₃.

Compound	λ_{max} solution/nm	ϵ solution/M ⁻¹ cm ⁻¹	λ_{max} film/nm	ϵ film/cm ⁻¹	E_g^{opt} film/eV	E_g^{CV} /eV	HOMO/eV	LUMO/eV
DRBDT ₃	536	1.20×10^5	572	6.8×10^4	1.97	1.94	-5.34	-3.40

3.3 X-Ray diffraction (XRD) and mobility

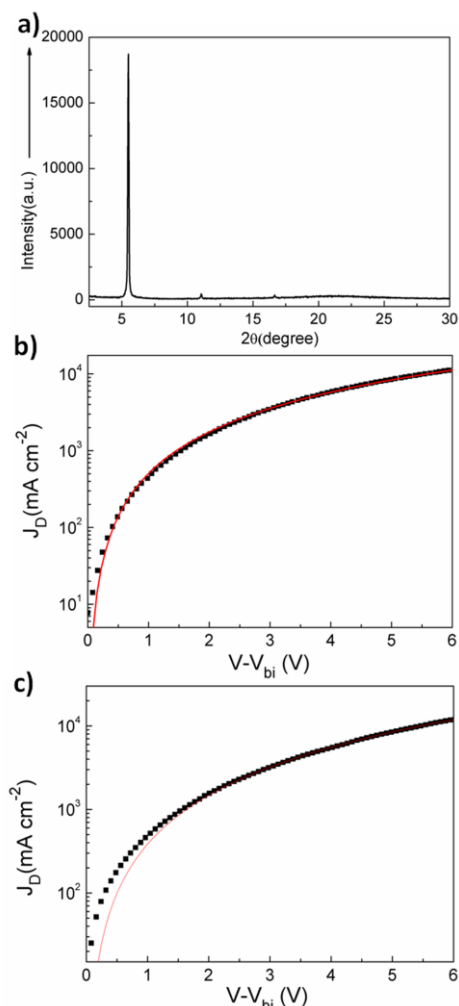


Figure 3 a) XRD pattern of pristine DRBDT₃ film spin-coated from CHCl₃ onto the glass substrate. b) J - V characteristics of a hole-only device with the configuration ITO/PEDOT:PSS (30 nm)/DRBDT₃/PC₇₁BM/Au (30 nm). c) J - V characteristics of an electron-only device with the configuration ITO/Al (30 nm)/DRBDT₃/PC₇₁BM/Au (30 nm). The solid line represents the fit using a model of single carrier SCLC with field-independent mobility. The J_D - V characteristics are corrected for the built-in voltage V_{bi} that arises from the work-function difference between the contacts.

The structural order of the pristine DRBDT₃ film spin-coated from CHCl₃ solution was investigated by XRD analysis. As shown in Figure 3a, we observed a strong diffraction peak at $2\theta=5.49^\circ$, corresponding to a d_{100} -spacing value of 16.09 Å for first order. The d_{100} -spacing value is the distance between the planes of the main conjugation chains of DRBDT₃, which are separated by alkoxy side-chains. The second- and third-order diffraction peaks, at $2\theta=11.05^\circ$ and 16.65° , are also clearly evident. The results of XRD show that DRBDT₃ has some long-range ordering at the solid state. The hole mobility of DRBDT₃/PC₇₁BM (w/w, 1:0.8) blend film was measured by SCLC method. As plotted in Figures 3b and 3c,

the calculated hole mobility and electron mobility of the blend films were $1.21 \times 10^{-4} \text{ cm}^2\text{V}^{-1}\text{S}^{-1}$ and $1.43 \times 10^{-4} \text{ cm}^2\text{V}^{-1}\text{S}^{-1}$, respectively. The blend films showed balanced hole and electron transport which could be beneficial for charge collection. The high hole mobility of DRBDT₃ was ascribed to the strong and ordered packing of DRBDT₃, as demonstrated by the XRD results above.

3.4 Photovoltaic properties

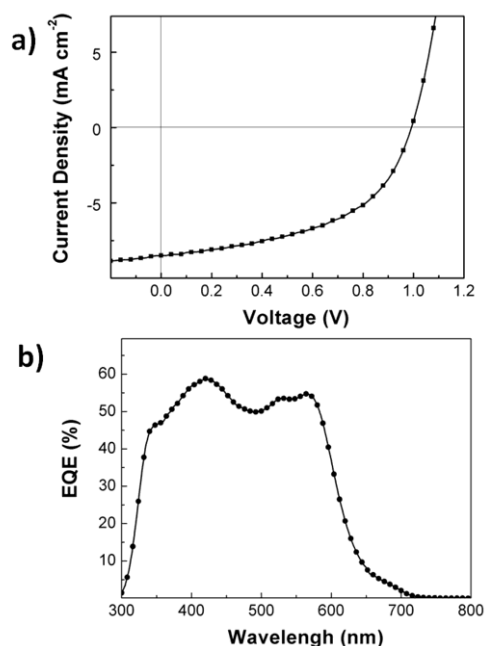


Figure 4 a) J - V curve of optimized device based on DRBDT₃:PC₇₁BM (1:0.8, w/w). b) EQE of devices based on DRBDT₃:PC₇₁BM (1:0.8, w/w) with or without thermal annealing.

Table 2 Device-performance parameters of the BHJ solar cells based on DRBDT₃:PC₇₁BM-blend films with different donor:acceptor blend ratios.

Blend ratio	V_{oc}/V	$J_{sc}/\text{mA cm}^{-2}$	FF	PCE (%)
1:0.5	0.98	7.90	0.48	3.72
1:0.8	0.99	8.26	0.50	4.09
1:1	0.99	7.96	0.49	3.86

SM-OPV devices were fabricated using DRBDT₃ as the electron donor material and PC₇₁BM as the electron acceptor material with a conventional device structure of glass/ITO/PEDOT:PSS/DRBDT₃:PC₇₁BM/ZnO/Al, using the solution spin-coating process. The typical J - V curve of the test results is displayed in Figure 4a and the results are summarized in Table 2. The optimized device based on DRBDT₃:PC₇₁BM-blend film shows a PCE of 4.09% with a high V_{oc} of 0.99 V, a J_{sc} of 8.26 mA cm⁻² and FF of 0.50. The high V_{oc} of the device based on DRBDT₃ is consistent with its deep HOMO energy level. The external quantum efficiency (EQE) spectrum of the optimized device is shown in Figure 4b. From the EQE curve, we can see that the DRBDT₃-based device exhibits a moderate response, from 330 nm to 640 nm, with the maximum EQE value of 58% at 420 nm.

3.5 Morphology of the DRBDT₃:PC₇₁BM-blend film

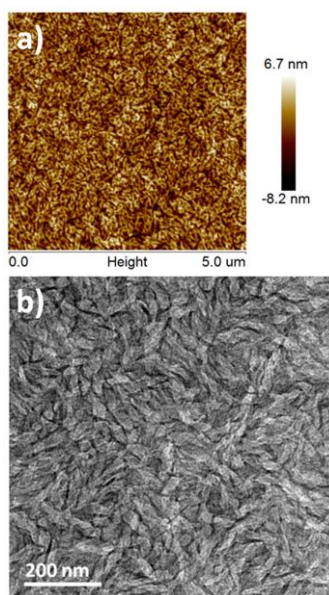


Figure 5 AFM (a) and TEM (b) images of DRBDT₃:PC₇₁BM-blend film.

The morphology of DRBDT₃:PC₇₁BM blend film was investigated by atomic-force microscopy (AFM) and transmission-electron microscopy (TEM). As shown in Figure 5a, root mean square (*rms*) roughness of DRBDT₃:PC₇₁BM blend film was 1.72 nm. The surface of the blend film was uniform and smooth, indicating the good film quality of the DRBDT₃:PC₇₁BM-blend film. From Figure 5b, we can see that the DRBDT₃:PC₇₁BM film exhibited an interpenetrating network with the width of 50~60 nm. The domain size is much larger than the hole charge diffusion length (10~20 nm), [50] leading to a relatively low photocurrent, which is consistent with the result of EQE.

4 Conclusions

In conclusion, we designed and synthesized a new oligo-benzo[1,2-b:4,5-b'] dithiophene derivative comprising of three benzo[1,2-b:4,5-b'] dithiophene units as the central building block and 3-ethyl-rhodamine as terminal units. The new molecule shows a broad absorption, from 300 to 640 nm, and a deep HOMO energy level of -5.34 eV. The optimal device based on DRBDT₃:PC₇₁BM-blend film shows a PCE of 4.09% with a high V_{oc} of 0.99V, a short-circuit current of 8.26 mA cm⁻², and a fill factor of 0.50. Furthermore, this new oligobenzo[1,2-b:4,5-b'] dithiophene derivative exhibits a much better photovoltaic performance than BDT-based homopolymer. These results demonstrate that oligo-benzodithiophene derivatives would be potential small-molecule donor materials for OPVs.

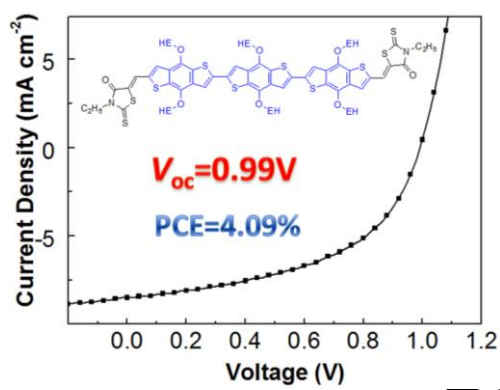
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