High Open-circuit Voltage and Low Voltage Loss in All-polymer Solar Cell with a Poly(coronenediimide-vinylene) Acceptor

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Abstract Reducing the voltage loss ($V_{oc,loss}$) is a critical factor in optimizing the open-circuit voltage ($V_{oc}$) and overall power-conversion efficiency (PCE) of polymer solar cells. In the current work, by designing a novel electron-accepting unit of coronenediimide (CDI) and using it as the main functional building block, a new polymer acceptor CDI-V is developed and applied to fabricate all-polymer solar cells. Compared with the perylenediimide-based polymer acceptors we previously reported, the current CDI-V polymer possesses a noticeably elevated lowest unoccupied molecular orbital (LUMO). Thereby, virtue of the enlarged energy gap between the donor HOMO and acceptor LUMO, a high $V_{oc}$ value of 1.05 V is achieved by the all-polymer photovoltaic device, along with an impressively low $V_{oc,loss}$ of 0.55 V. As remarkably, in spite of an extremely small LUMO level offset of 0.01 eV exhibited by the donor and acceptor polymers, effective charge separation still takes place in the all-polymer device, as evidenced by a proper short-circuit current ($J_{sc}$) of 9.5 mA·cm$^{-2}$ and a decent PCE of 4.63%.

Keywords All-polymer solar cell; Coronenediimide; Voltage loss; Acceptors

INTRODUCTION

Compared with solar cells based on polymer donor allied with fullerene or non-fullerene small-molecule acceptors (SMAs), bulk-heterojunction (BHJ) all-polymer solar cells (all-PSCs) featuring an active layer of a p-type polymer donor blended with n-type polymer acceptor boast the potential advantages of superior morphological stability, mechanical robustness, and better compatibility with large-area printing techniques.[1−6] Although the best power-conversion efficiency (PCE) of all-PSCs has so far lagged behind the SMA-based devices,[7−11] very rapid advancement has been witnessed in this area for the past few years.[12−16]

Our group has been focusing on harnessing polycyclic aromatic dicarboximides, particularly perylenediimide (PDI) derivatives, as the main functional building block to develop high-performance acceptor materials.[17−21] In comparison to the more widely studied naphthenediimide (NDI)-based polymer acceptors, PDI polymers possess considerably higher light absorbance coefficients in the visible light regime. By tuning the steric feature of the side chains, we have designed and achieved PDI polymers exhibiting a lower tendency to self-aggregation and over-crystallization, and thus materials more amenable to facile solution-processing procedures.[22−24] By screening a variety of spacer units, the slim vinylene group was found to suit well with the relatively congested bay-region of PDI moiety, to confer the polymer with a relatively planar conjugated backbone and more compact chain packing in the condensed state.[19]

Despite the desirable PCEs achieved by our previously designed PDI-polymer acceptors, the open-circuit voltage ($V_{oc}$) of relevant all-PSC devices was only 0.7 V to 0.8 V, which greatly limited the overall PCE. Hence, improving $V_{oc}$ via material structure optimization arose as an imperative issue. While enlarging the energy gap between the highest occupied molecular orbital (HOMO) of the donor and the lowest unoccupied molecular orbital (LUMO) of the acceptor is a well-known practice for enhancing $V_{oc}$, minimizing the voltage loss ($V_{oc,loss}$)[25] is apparently of equal importance.[26−30] To this end, Liu and coworkers designed a high-LUMO polymer acceptor P-BNBP-T comprising a double B−N bridged bipyrindyl functional group, with which a remarkable $V_{oc}$ of 1.3 V (3.2% PCE) was realized, with a small $V_{oc,loss}$ of 0.58 V.[31] Jenekhe et al. recently reported a set of biselenophene- and
bisselenophene-linked random NDI copolymers. Blending one of such NDI acceptors ($E_g = 1.45$ eV) with a donor polymer of PBDB-T enabled a notable PCE of 10.1%, featuring a small $V_{oc}$ of 0.59 V. This result represented an attractive example of combining high PCE with small $V_{oc}$ in all-PSCs.[15] In order to enhance $V_{oc}$ of PDI-based all-PSCs, we have now designed a novel aromatic dicarboximide and used it to construct a new polymer acceptor exhibiting an elevated LUMO. Coronenediimide (CDI) is a derivative from PDI by fusing a vinylene unit to the bay regions on both sides, resulting in an expanded polycyclic aromatic framework.[32] Such a chemical modification to PDI brings about distinct electronic effects to the monomer and related polymers. Density functional theory (DFT) calculations (Fig. S1 in the electronic supplementary information, ESI) estimated the LUMO of CDI-V polymer to be $-3.25$ eV, which is about 0.45 eV higher than that calculated for PDI-V. We anticipate such a raise in the LUMO of acceptor to promote a growth in $V_{oc}$. Besides, the discotic geometry of the polycyclic framework featured by CDI may as well induce a favourable molecular packing motif for improving the carrier transport.[33–37] As expected, the all-PSC based on an actively layer of PTB7-Th:CDI-V polymer blend achieves an impressive $V_{oc}$ of 1.05 V, nearly 0.31 V higher than that shown by devices using PDI-V as the acceptor.[19] Furthermore, it is worth noting that a fairly small $V_{oc}$ of 0.55 V is manifested by this system, featuring an extremely small LUMO level energy gap between the donor PTB7-Th and acceptor CDI-V. Hence, with a respectable PCE value of 4.63%, the current result represents one of the smallest $V_{oc}$ values accomplished by all-PSCs exhibiting a $V_{oc}$ of over 1 V.

**EXPERIMENTAL**

**General Methods**

The NMR spectra were recorded with a Bruker-400 (400 MHz) using CDCl$_3$ as the solvent. The chemical shifts in $^1$H- and $^{13}$C-NMR spectra are reported in parts per million (ppm) with TMS (0 ppm) and CDCl$_3$ (77.23 ppm) as the references for $^1$H- and $^{13}$C-NMR, respectively. MALDI-TOF mass spectra were recorded with an ABI 4800 Plus MALDI TOF/TOF Analyzer using CHCA as the matrix, or a Bruker BIFLEX III MALDI TOF spectrometer using CHCA as the matrix. The elemental analyses were performed with a vario EL elemental analyzer (Elementar Analysensysteme GmbH). UV-Vis absorption spectra were acquired with a Hitachi U-4100 spectrophotometer. The cyclic voltammetry (CV) was conducted with a Gamry Interface 1000 electrochemical workstaton with three electrodes configuration.

**Materials**

All reagents were purchased from commercial sources and used without further purification unless otherwise specified. Trans-1,2-bis(tri-n-butylstannyl)ethylene was purchased from Alfa Aesar and PTB7-Th was purchased from 1-Material. Dichloromethane and toluene were distilled prior to use.

**Synthesis of the Polymer Acceptor**

**Synthesis of the monomer CDI-Cl$_2$**

Dichloro-d(triethylsilyl)coronenediimide (CDI-Cl$_2$-TMS$_2$) was prepared via a modified procedure from a previous report of our group,[38] and the yield was improved to 96%. CDI-Cl$_2$-TMS$_2$ (0.396 g, 0.39 mmol) was dissolved in dry dichloromethane (30 mL). Then, a solution of TBAF (tetra-n-butylammonium fluoride) in THF (1 mol·L$^{-1}$, 1.56 mmol) was added to it, and the reaction mixture was stirred in an ambient environment for 30 min. The resultant mixture was washed with water and dried over anhydrous Na$_2$SO$_4$ before it was concentrated under reduced pressure. The residue was purified with silica gel column chromatography eluted with dichloromethane/petroleum ether (1:1) to afford CDI-Cl$_2$, as a dark orange solid (0.345 g, 0.38 mmol, 98%).$^1$H-NMR (400 MHz, CDCl$_3$, $\delta$ ppm): 10.02 (m, 2H), 9.56 (m, 2H), 8.87 (m, 2H), 5.47 (m, 2H), 2.51 (m, 4H), 2.16 (m, 4H), 1.63–1.38 (m, 16H), 1.31 (m, 16H), 0.85 (m, 12H).$^{13}$C-NMR (100 MHz, CDCl$_3$, $\delta$ ppm): 163.4, 162.5, 132.6, 128.1, 127.5, 127.0, 126.6, 125.0, 121.5, 120.9, 119.9, 118.0, 117.4, 116.3, 55.5, 32.6, 32.0, 29.5, 27.5, 22.8, 14.2. MALDI-TOF-MS: Calcd. for C$_{44}$H$_{38}$Cl$_2$N$_2$O$_4$ [M + H$^+$]: 871.3. Found: 871.3. Elemental Anal.: Calcd. for C$_{44}$H$_{38}$Cl$_2$N$_2$O$_4$: C, 74.38; H, 6.94; N, 3.21. Found: C, 74.31; H, 6.96; N, 3.19.

**Synthesis of CDI-V**

A mixture of CDI-Cl$_2$ (0.109 g, 0.12 mmol), trans-1,2-bis(tri-n-butylstannyl)ethylene (0.076 g, 0.12 mmol), Pd(0.109 g, 0.12 mmol), 74.31; H, 6.96; N, 3.19. Calcd. for C$_{30}$H$_{26}$Cl$_2$N$_2$O$_4$+ [M  +  H$^+$]: 625.2. Found: 625.2. Elemental Anal.: Calcd. for C$_{30}$H$_{26}$Cl$_2$N$_2$O$_4$: C, 74.38; H, 6.94; N, 3.21. Found: C, 74.31; H, 6.96; N, 3.19.

**Device Fabrication and Characterization**

Pre-patterned ITO-coated glasses with a sheet resistance of $\sim 15$ OMS$^{-1}$ were used as the substrates. They were cleaned by sequential cycles of sonication in soap water, deionized water, acetone, and isopropanol for 15 min each. After UV/ozone treatment for 60 min, ZnO electron transport layers were prepared on top of the ITO by spin-coating a ZnO precursor solution (diethyl zinc) at 5000 r·min$^{-1}$. The active-layer solutions were prepared in chlorobenzene (CB, with a polymer concentration of 9 mg·mL$^{-1}$ and a D/A weight ratio at 1/1). To completely dissolve the polymers, the solutions were stirred on a hot plate at 110 °C for at least 3 h. The active layers were spin-coated from the cooled polymer solution on the ZnO-coated substrates in a glove box under N$_2$ atmosphere at 1700 r·min$^{-1}$ to obtain a thickness of $\sim 120$ nm. The blend films were then transferred to the vacuum chamber with a thermal evaporator in the same glove box. At a vacuum level of $\leq 4 \times 10^{-6}$ Pa, a thin layer ($\sim 7$ nm) of Al$_2$O$_3$ was deposited as the anode interlayer, followed by the deposition of 100 nm of Al as the top electrode. All cells were encapsulated using epoxy inside the glove box. The device $J$-$V$ characteristics were measured under AM1.5G (100 mW·cm$^{-2}$) using a Newport solar simulator. The light

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intensity was calibrated using a standard Si diode (with a KG5 filter purchased from PV Measurement) to bring spectral mismatch to unity. The J-V characteristics were recorded using a Keithley 2400 source meter unit. Typical cells have a device area of 5.9 mm², which is defined by a metal mask with an aperture aligned with the device area. External quantum efficiencies (EQEs) were measured using a Newport EQE system equipped with a standard Si diode. Monochromatic light was generated from a Newport 300 W lamp source.

RESULTS AND DISCUSSION

Synthesis and Characterization

The synthesis route of polymer acceptor CDI-V is shown in Scheme 1. As a critical precursor, CDI-Cl-TMS₂ was prepared from a regio-isomer mixture of 1,6/1,7-dibromo-PDI (the 1,6/1,7 ratio is about 1/4), following a modified procedure from our previous report. The annulation step was optimized by reducing the reaction time at −78 °C to 5 min before an aqueous solution of Na₂S₂O₅ was added to quench the excess ICl. This new protocol helped preventing the produced CDI-Cl₂-TMS₂ from further substitution by ICl. TBAF was then used as the desilylation reagent to offer CDI-Cl₂ in a quantitative yield. Polymer CDI-V was prepared via a Stille cross-coupling reaction between CDI-Cl₂ and trans-1,2-bis(tri-n-butylstannyl)ethylene, using Pd₂(dba)₃ and P(t-Bu)₃ as the catalyst. The crude product was purified with a size-exclusion chromatography column to remove oligomers and impurities, followed by precipitation from methanol to afford polymer CDI-V as a red solid in 71% yield. According to the high-temperature gel-permeation chromatography (GPC), the number-average molecular weight (Mₙ) and polydispersity index (Mₙ/Mₚ) of the product were 8.9 kDa and 2.3, respectively. CDI-V showed good solubility in a variety of organic solvents including chloroform, chlorobenzene (CB), and o-dichlorobenzene at room temperature. The polymer also exhibited optimal thermal stability, as evidenced by a decomposition temperature of 363 °C (with 5% weight loss) under nitrogen atmosphere shown by the thermogravimetric analysis (TGA) profile (Fig. S2 in ESI).

Photophysical and Electronic Properties

The electronic and electrochemical properties of CDI-V were examined with the UV-Vis absorption spectroscopy and cyclic voltammetry (CV). The absorption spectra were collected from both chloroform solution and thin film (Fig. 1 and Table 1). A broad absorption band ranging from 300 nm to 600 nm with two major peaks shown at ca. 370 and 540 nm was observed. According to CV (Fig. 1 and Fig. S3 in ESI), the LUMO level of CDI-V was calculated to be −3.68 eV, which was about 0.27 eV higher than that of PDI-V. Such a LUMO difference was consistent with the DFT calculation results (Fig. S1 in ESI). Nonetheless, it was noticed that such an elevated LUMO of acceptor negatively entailed a very small energy difference than that of donor polymer, PTB7-Th. Namely, the LUMO energy gap (ΔE_{LUMO}) between the donor and acceptor polymers was narrowed to merely 0.01 eV. It was interesting to inspect whether such a small driving force was sufficient to overcome the binding energy of excitons and promote efficient charge separation. To help analyse the charge separation efficiency in the active layer, the photoluminescence (PL) quenching experiments were conducted with the PTB7-Th:CDI-V blend films (Fig. S4 in ESI). The fluorescence emission intensity of the acceptor and donor was quenched by over 90% and 65%, respectively, suggesting that the exciton collection at the donor/acceptor interface was fairly efficient despite the extremely small ΔE_{LUMO} exhibited by the donor and acceptor polymers.

![Scheme 1 Synthesis of CDI-V.](https://doi.org/10.1007/s10118-020-2426-6)
Device Performance

As the donor polymer, the absorption of PTB7-Th with a relatively narrow bandgap ($E_g = 1.60$ eV) [39,40] very well complemented the spectrum of the newly designed acceptor CDI-V. The all-PSC solar cells were then fabricated with an inverted device structure of ITO/ZnO/active layer/V$_{2}$O$_{5}$/Al. In the absence of CN, the electron and hole mobilities were improved to $5.6 \times 10^{-4}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. With the addition of 1% CN, the electron and hole mobilities were measured with a space charge limited current (SCLC) experiments, the hole mobility was measured with a device structure of ITO/V$_{2}$O$_{5}$/PTB7-Th:CDI-V/V$_{2}$O$_{5}$/Al, and the electron mobility was measured with a configuration of ITO/ZnO/PTB7-Th:CDI-V/Ca/Al. In the absence of CN, the electron and hole mobilities were calculated to be $2.4 \times 10^{-5}$ and $8.3 \times 10^{-4}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. With the addition of 1% CN, the electron and hole mobilities were improved to $5.6 \times 10^{-5}$ and $1.4 \times 10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively.

The EQE spectra confirmed that the CDI-V-based devices exhibited strong photo-responses in a broad range of 300 nm to 700 nm (Fig. 2b). The current values calculated by integrating the EQE profiles recorded from cells with and without the solvent additive were 9.94 and 8.40 mA cm$^{-2}$, respectively. These data matched well with the $J_{sc}$ values measured from the J-V experiments. Upon the addition of CN, a pronounced photo-response increment was perceivable around 500 nm to 700 nm, which covered the absorption ranges of both the acceptor (CDI-V) and donor (PTB7-Th) polymers. It was thus suggested that the solvent additive helped optimizing the morphologies in both the donor- and acceptor-phase domains.

Atomic force microscopy (AFM) was employed to characterize the surface morphology of the active layer (Fig. 3). The phase images revealed that nanoscale phase separations favourable for charge transport were formed in both blend films with and without the solvent additive, and the root-mean-square roughness (RMS) in the height images revealed relatively smooth and uniform surfaces. Nonetheless, evident differences in the bulk charge-transport capability were detected upon the addition of solvent additive to the active layer (Table 2). In the space charge limited current (SCLC) experiments, the hole mobility was measured with a device structure of ITO/V$_{2}$O$_{5}$/PTB7-Th:CDI-V/V$_{2}$O$_{5}$/Al, and the electron mobility was measured with a configuration of ITO/ZnO/PTB7-Th:CDI-V/Ca/Al. In the absence of CN, the electron and hole mobilities were calculated to be $2.4 \times 10^{-5}$ and $8.3 \times 10^{-4}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. With the addition of 1% CN, the electron and hole mobilities were improved to $5.6 \times 10^{-5}$ and $1.4 \times 10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively.

### Table 1: Summary of optical and electrochemical properties of CDI-V.

| Polymer | $\lambda_{max, sol}$ (nm) | $\lambda_{max, film}$ (nm) | $\lambda_{em, sol}$ (nm) | $\lambda_{em, film}$ (nm) | $E_g^{opt}$ (eV) | LUMO $^b$ (eV) | HOMO $^c$ (eV) |
|---------|--------------------------|-----------------------------|-------------------------|--------------------------|-----------------|----------------|---------------|
| CDI-V   | 371, 540                 | 370, 539                    | 569                     | 579                      | 2.14            | $-3.68 \pm 0.03$ | $-5.82 \pm 0.03$ |

$^a$ Estimated from the onset of absorption band of CDI-V in thin film; $^b$ Estimated from CV using Fc$^+$/Fc as the external reference; $^c$ Calculated using the equation: HOMO = LUMO − $E_g^{opt}$.

### Table 2: Device parameters of PTB7-Th:CDI-V-based all-PSCs.

| D/A     | Additives | $V_{oc}$ (V) | $J_{sc}$ (mA cm$^{-2}$) | FF   | PCE $^a$ (%) | $\mu_e$ (cm$^2$ V$^{-1}$ s$^{-1}$) | $\mu_h$ (cm$^2$ V$^{-1}$ s$^{-1}$) |
|---------|-----------|--------------|--------------------------|------|-------------|---------------------------------|---------------------------------|
| 1/1     | 0         | 1.04 ± 0.01  | 8.43 ± 0.05 (8.40 ± 0.06) | 0.45 ± 0.01 | 3.96 ± 0.12 (4.09) | 2.4 × 10$^{-5}$ | 8.3 × 10$^{-4}$ |
| 1/1     | 1% CN     | 1.05 ± 0.01  | 9.50 ± 0.15 (9.94 ± 0.12) | 0.46 ± 0.01 | 4.54 ± 0.07 (4.63) | 5.6 × 10$^{-5}$ | 1.4 × 10$^{-3}$ |

$^a$ Integrated current from EQE profile shown in parentheses; $^b$ Average value from 10 solar cell devices and the best value shown in parentheses.

![Fig. 1](https://doi.org/10.1007/s10118-020-2426-6)
and 1.4 × 10^{-3} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}, respectively. Meanwhile, the ratio of $\mu_h/\mu_e$ decreased from 35 to 25, implying more balanced charge transport abilities. These data reinforced the increased $J_{sc}$ and FF values measured in the device processed with the solvent additive, which presumably helped improving the binary blend morphology and/or phase purity in the active layer. Such desirable effects induced by the solvent additive in the current system presented a stark contrast to our previous results from PDI-V and NDP-V polymer acceptors, with which solvent additives did not bring about favourable device performance.

**CONCLUSIONS**

In order to optimize the $V_{oc}$ in all-PSCs, a new electron-acceptor unit, coronenediimide (CDI), was designed and synthesized. Owing to the unique electronic effect brought about by the two vinylene units fused to the bay regions of PDI, this polycyclic aromatic dicarboximide molecule featured a noticeably raised LUMO level. When such a new functional building block was joined into a polymer structure by vinylene linkers, a new potent polymeric electron acceptor was developed, showing an enlarged band gap compared to its analogue of PDI-V. Hence, when combined with the same polymer donor PTB7-Th, a much widened energy gap between the acceptor LUMO and donor HOMO necessarily produced a substantially enlarged $V_{oc}$ of 1.05 V in all-PSC devices. Such a favourable $V_{oc}$ value also reflected an impressively small voltage loss of 0.55 V. Most remarkably, an extremely small LUMO level gap of 0.01 eV was manifested by the donor and acceptor polymers in this system. In spite of such a small charge-separation driving force, a respectable $J_{sc}$ value of 9.50 mA·cm$^{-2}$ with a decent

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**Fig. 2** (a) $J$-$V$ curves and (b) EQE spectra of photovoltaic devices with or without the solvent additive.

**Fig. 3** (a, b) Height and (c, d) phase images of PTB7-Th:CDI-V blend films (a, c) with or (b, d) without 1% CN.
PCE of 4.63% was achieved. Hence, the current results have not only demonstrated a new promising polymer acceptor for all-PSCs featuring high V_{oc} but also a unique platform for studying the distinctive charge-separation mechanism in organic photovoltaic devices.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at https://doi.org/10.1007/s10118-020-2426-6.

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