

A Low-Energy-Gap Organic Dye for High-Performance Small-Molecule Organic Solar Cells

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 Supporting Information

ABSTRACT: A novel donor–acceptor–acceptor (D–A–A) donor molecule, **DTDCTB**, in which an electron-donating ditolylaminothienyl moiety and an electron-withdrawing dicyanovinylene moiety are bridged by another electron-accepting 2,1,3-benzothiadiazole block, has been synthesized and characterized. A vacuum-deposited organic solar cell employing **DTDCTB** combined with the electron acceptor C₇₀ achieved a record-high power conversion efficiency (PCE) of 5.81%. The respectable PCE is attributed to the solar spectral response extending to the near-IR region and the ultracompact absorption dipole stacking of the **DTDCTB** thin film.

Organic solar cells (OSCs) have garnered considerable research interest because of their prominent merits, such as low cost, light weight, and mechanical flexibility. At present, solution-processed bulk heterojunction (BHJ) solar cells¹ based on bicontinuous interpenetrating networks of π -conjugated polymers and soluble fullerene derivatives have demonstrated remarkable achievements, with power conversion efficiencies (PCEs) in excess of 7%.² Beyond that, the small-molecule counterparts, particularly p-type organic semiconductors utilized for OSCs, have also shown exceptional promise. The competitive nature of small molecules relative to polymeric materials can be ascribed to the predominant advantages including well-defined molecular structures, easier purification, and better batch-to-batch reproducibility. Therefore, tremendous research endeavors have been devoted to developing small-molecule OSCs (SMOSCs),³ which exhibit appreciable PCEs of >5%, by using either solution-processing or vacuum-deposition fabrication techniques.⁴ Although solution processing is generally considered to be more cost-effective than vacuum deposition, vacuum-deposited SMOSCs are emerging as competitive OSCs because of the advantage of easy fabrication of multilayer tandem architectures.⁵ In this regard, a tandem SMOSC device with a PCE of up to 8.3% has been disclosed recently.⁶

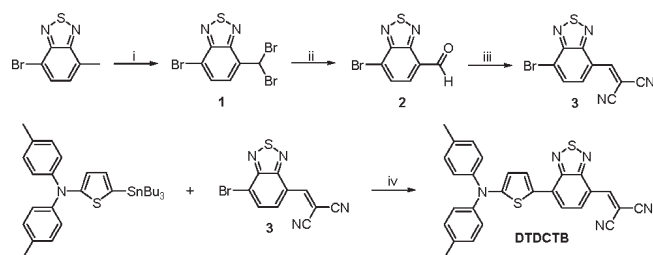
To date, the molecular architectures of most donor materials for SMOSCs fabricated by vacuum-evaporation processes can be classified into two main categories: acceptor–donor–acceptor (A–D–A) and donor–acceptor (D–A) systems. A–D–A systems, in which electron-rich oligothiophenes are end-capped with various electron-withdrawing groups such as dicyanovinylene, 2,1,3-benzothiadiazole, and thiadiazolo[3,4-c]pyridine, are currently among the most successful molecular architectures.

Tailoring of the conjugation length of the oligothiophene core unit as well as the length and location of pendant alkyl side chains has enabled this class of active materials to demonstrate reliable PCEs.⁷ One distinctive feature of such materials is that they possess deep-lying highest occupied molecular orbital (HOMO) energy levels and thus afford SMOSCs with extraordinary open-circuit voltages (V_{oc}) of ~ 1 V. On the other hand, D–A-type molecules incorporating arylamines as electron-donating groups also appear to be attractive candidates because of their effective intramolecular charge transfer (ICT) characteristics. Moreover, taking advantage of the versatility of structural modifications in such systems enables the frontier orbital energy levels to be readily tuned through judicious combinations of different electron-donating and/or -accepting functional groups. Along this line, a series of triphenylamine-based D–A materials have been reported to exhibit PCEs of up to 2.2%.⁸ However, both of these types of materials generally suffer from insufficient light-harvesting capabilities. They usually have absorption maxima at less than 600 nm, which may be one of the main impediments to further improvement of their efficiencies. Although a few dyes, such as squaraine⁹ and merocyanine,¹⁰ have been explored to address this issue, the progress still lags behind that for the A–D–A counterparts. Therefore, it is highly desired to design new molecular architectures that can readily allow donor materials to extend the spectral responses to the far-red and even near-IR regions.

In this communication, we report a novel donor–acceptor–acceptor (D–A–A)-type donor molecule, **DTDCTB** (Scheme 1), whose thin-film absorption can extend into the near-IR region. In this molecule, an electron-donating ditolylaminothienyl moiety and an electron-withdrawing dicyanovinylene moiety are bridged by another electron-accepting 2,1,3-benzothiadiazole (BT) block. Specifically, **DTDCTB** was designed with the following structural characteristics: (i) The ditolylaminothienyl block behaves as a stronger electron-donating moiety in comparison with the conventional ditolylaminophenyl congener because of its fortified quinoidal character. The quinoidal resonance structure is energetically less stable than the aromatic form, and thus, the adoption of the ditolylaminothienyl donor endows **DTDCTB** with a smaller band gap.^{1c} (ii) The BT moiety represents the most ubiquitous acceptor utilized in optoelectronic materials (e.g., low-band-gap polymer donors,¹ nonfullerene acceptors,¹¹ and n-type field-effect transistors¹²) because of its fascinating features, including low-

Received: June 7, 2011

Published: September 09, 2011

Scheme 1. Synthetic Route to DTDCTB^a

^a (i) NBS, azobis(isobutyronitrile), chlorobenzene, 80 °C, 83%. (ii) AgNO₃, H₂O/MeCN, reflux, 92%. (iii) Malononitrile, Al₂O₃, toluene, 70 °C, 67%. (iv) PdCl₂(PPh₃)₂, toluene, 110 °C, 55%.

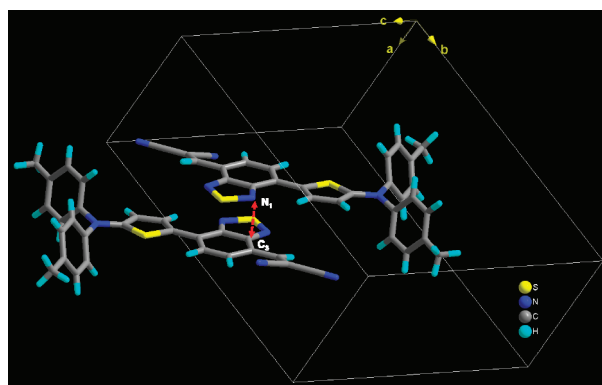


Figure 1. X-ray-analyzed molecular structure and crystal packing of DTDCTB.

band-gap character, high absorption coefficient, and appropriate energy levels. (iii) According to our previous study,¹³ the innovative combination of two acceptors (BT and dicyanovinylene used here) would effectively narrow the optical band gap of the resulting molecule while maintaining a relatively low-lying HOMO energy level, and thus, an enhanced short-circuit current (J_{sc}) and V_{oc} value can be concurrently anticipated. As a proof of concept, our preliminary investigation of vacuum-deposited SMOSCs employing DTDCTB as the electron donor and C₇₀ as the electron acceptor revealed a remarkable PCE as high as 5.81%. To the best of our knowledge, this efficiency is among the highest values ever reported for vacuum-deposited single cells with organic donor molecules. This encouraging result indicates the great potential of such D–A–A systems in creating high-performance donor materials for SMOSCs.

The synthesis of DTDCTB is described in Scheme 1. Benzylic bromination of 4-bromo-7-methyl-2,1,3-benzothiadiazole¹⁴ with *N*-bromosuccinimide (NBS) afforded **1**. Silver nitrate-promoted hydrolysis of **1** gave aldehyde **2**, which was then condensed with malononitrile to give the key intermediate **3** via Knoevenagel reaction in the presence of basic Al₂O₃. Finally, Stille coupling of 5-(*N,N*-ditolylamino)-2-(tri-*n*-butylstannyl)thiophene¹⁵ and **3** yielded DTDCTB in good yield. It is noteworthy that this synthetic route provides a versatile method for further engineering of the molecular structure through the combination of the unsymmetrical intermediate **3** and other electron-donating groups.

The molecular structure of DTDCTB was analyzed by X-ray crystallography. As shown in Figure 1, DTDCTB displays an

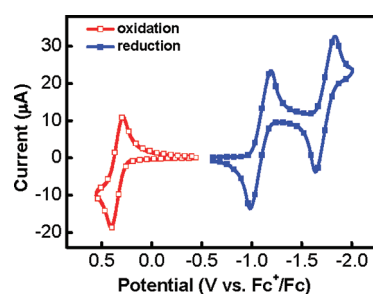


Figure 2. Cyclic voltammograms of DTDCTB in solution.

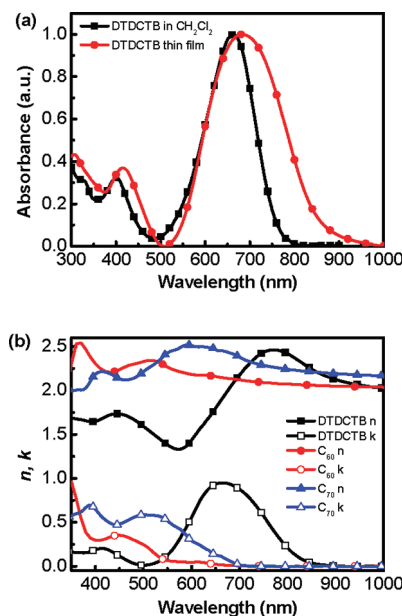


Figure 3. (a) Normalized absorption spectra of DTDCTB in CH₂Cl₂ and a thin film. (b) Optical constants (refractive index, n , and extinction coefficient, k) of DTDCTB, C₆₀, and C₇₀ thin-film spectra.

almost coplanar conformation between the thiophene and BT rings with a dihedral angle of 5.5°. This coplanarity facilitates the electronic coupling between the electron-donating and electron-withdrawing blocks, enhancing the ICT efficiency and thus ensuring a distinctive bathochromic shift of the spectral responses. The strong polar character and coplanar conformation of the heteroaryl components lead DTDCTB crystals to pack in an antiparallel manner along both molecular axis directions. The cofacial arrangement of two neighboring BT rings with a shortest point-to-point (N1–C3) distance of 3.55 Å indicates non-negligible π – π interactions, which may facilitate charge-carrier hopping in the solid state.

The electrochemical properties of DTDCTB were probed by cyclic voltammetry in solution. As shown in Figure 2, DTDCTB exhibits one reversible oxidation potential at 0.35 V vs ferrocene/ferrocenium (Fc/Fc⁺), corresponding to oxidation of the ditolylaminothienyl donor. On the other hand, two reversible reduction waves at –1.09 and –1.74 V vs Fc/Fc⁺ were observed in the cathodic potential regime. The first reduction potential can be ascribed to the reduction of the dicyanovinylene block and the second to reduction of the BT fragment. By reference to the Fc/Fc⁺ redox couple, where the HOMO of Fc is assigned to be 4.8 eV below the vacuum level, the HOMO and lowest

unoccupied molecular orbital (LUMO) energy levels of **DTDCTB** were calculated to be -5.15 and -3.71 eV on the basis of the oxidation potential and the first reduction potential, respectively. In spite of the presence of the strong ditolylaminothienyl donor, **DTDCTB** shows a relatively low-lying HOMO level because of the strong electron-withdrawing character of the BT and dicyanovinylene blocks. Given energy level shifts due to intermolecular interactions, the HOMO level of the **DTDCTB** thin film was determined by UV photoelectron spectroscopy to have a value of -5.30 eV. The fairly low-lying HOMO would result in a large energy level offset to the LUMOs of fullerenes, ensuring relatively large V_{oc} values.

Figure 3a shows the electronic absorption spectra of **DTDCTB** in a vacuum-deposited thin film and in CH_2Cl_2 solution. Surprisingly, with such a short effective conjugation length, the absorption spectrum of **DTDCTB** in solution shows a band at $\lambda_{\text{max}} = 663$ nm accompanied by a high extinction coefficient (k) of up to $41\,660\text{ M}^{-1}\text{ cm}^{-1}$. To gain more insight into the electronic and optical properties of **DTDCTB**, density functional theory (DFT) and time-dependent DFT (TDDFT) calculations were performed for the molecule in CH_2Cl_2 solution [Figure S1 and Tables S1 and S2 in the Supporting Information (SI)] and gave a computed λ_{max} value of 673 nm that is close to the experimental results. On the other hand, the thin-film absorption of **DTDCTB** exhibits $\lambda_{\text{max}} = 684$ nm. The significant broadening of the thin-film absorption spectrum is possibly due to intermolecular π - π stacking as evidenced in the crystal packing. Apparently, the thin-film absorption of **DTDCTB** effectively extends the useful photon-harvesting range down to near-IR wavelengths (700–800 nm). Optical constants of vacuum-deposited thin films of **DTDCTB**, C_{60} , and C_{70} are shown in Figure 3b. The **DTDCTB** thin film exhibits high k values across the 550–800 nm wavelength range, with $k_{\text{max}} \approx 0.95$ at $\lambda = 670$ nm, which is coincident with the absorption spectrum shown in Figure 3a. Notably, the k_{max} value is among the highest reported for solar-absorbing organic thin films, as compared with the commonly used nano/microcrystalline poly(3-hexylthiophene) (P3HT) and copper phthalocyanine (CuPc), which show k_{max} values of ~ 0.65 and ~ 0.95 , respectively.^{16,17} This result indicates that the efficient ICT gives the **DTDCTB** molecule high polar character, which favors the formation of ultracompact absorption dipole packing in the thin film upon vacuum deposition. In addition, the k spectra of the commonly used acceptor molecules C_{60} and C_{70} show complementary wavelength coverage (<550 nm) with respect to the **DTDCTB** thin film. As a result, fullerenes C_{60} and C_{70} were selected as acceptor materials to be paired with **DTDCTB** for subsequent device fabrication.

In photovoltaic characterizations, we adopted the vacuum-deposited planar mixed heterojunction (PMHJ) structure incorporating one layer of mixed donor/acceptor materials sandwiched between pure donor and acceptor layers.¹⁸ The optimized device structures were configured as follows: Device I: ITO/MoO₃ (30 nm)/**DTDCTB** (7 nm)/1:1 (v/v) **DTDCTB**: C_{60} (40 nm)/ C_{60} (20 nm)/2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) (10 nm)/Ag (150 nm). Device II: ITO/MoO₃ (5 nm)/**DTDCTB** (7 nm)/1:1 (v/v) **DTDCTB**: C_{70} (40 nm)/ C_{70} (7 nm)/BCP (10 nm)/Ag (150 nm). In these devices, BCP was employed as both an electron-transporting layer and an exciton-blocking layer. More importantly, MoO₃ was chosen as the hole-transporting layer because of its superior performance in comparison with several other hole-transporting materials.¹⁹ It was noted that BHJ polymer solar cells incorporating MoO₃ usually

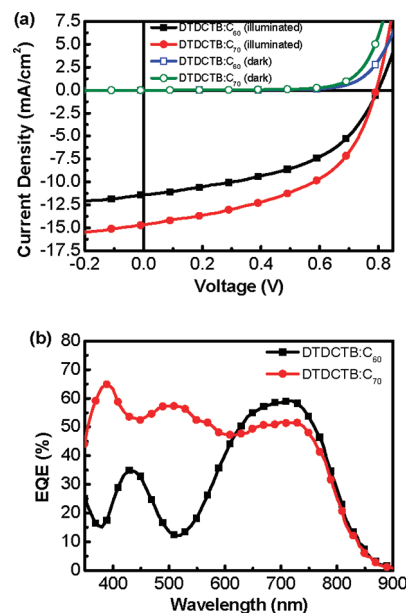


Figure 4. (a) J - V characteristics and (b) EQE spectra of **DTDCTB**: C_{60} PMHJ (squares) and **DTDCTB**: C_{70} PMHJ (circles) solar cells.

exhibit better device stability than those fabricated with poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS).²⁰ Optimizations to fine-tune the thicknesses of the **DTDCTB** and MoO₃ layers are shown in the SI. It was found that the devices with a 7 nm **DTDCTB** donor layer demonstrated superior performance. Moreover, through spectrum-response modeling of the simplified bilayer cells, the exciton diffusion length in the **DTDCTB** thin film was found to be ~ 6 nm (Figure S4),²¹ which is within the typical range of 5–10 nm for organic semiconductors. Figure 4a shows the current density–voltage (J - V) characteristics of **DTDCTB**:fullerene PMHJ solar cells. The **DTDCTB**: C_{60} PMHJ device gave a V_{oc} of 0.80 V, a J_{sc} of 11.40 mA/cm^2 , and fill factor (FF) of 0.48, yielding a PCE of 4.41% under AM 1.5 G 1 sun ($100\text{ mW}/\text{cm}^2$) simulated solar illumination. Remarkably, the **DTDCTB**: C_{70} device delivered higher performance, with a V_{oc} of 0.79 V, J_{sc} of 14.68 mA/cm^2 , FF of 0.50, and PCE of 5.81%, which is the highest one ever reported for SMOSCs. The high V_{oc} values of these devices are due to the moderately low-lying HOMO level of **DTDCTB**. However, the V_{oc} value (~ 0.8 V) still leaves room for future improvement. The FF values of the **DTDCTB**: C_{60} and **DTDCTB**: C_{70} devices are similar, suggesting similar blending layer morphologies and charge-carrier percolation networks in the two devices. The higher J_{sc} of the **DTDCTB**: C_{70} device can be attributed to the higher extinction coefficient of C_{70} relative to C_{60} , which is fully consistent with the external quantum efficiency (EQE) spectra shown in Figure 4b. The EQE spectrum of the **DTDCTB**: C_{70} device shows impressive high values of $\sim 50\%$ throughout the UV–vis to near-IR range (350–770 nm), resulting in the high J_{sc} . It is noteworthy that the integrated EQE values are in agreement with the measured short-circuit currents (within 3–5% error; see Tables S3 and S4). Although the **DTDCTB**: C_{70} films show amorphous morphologies (see the X-ray diffractogram in Figure S5), the donor–acceptor phase separation can be clearly observed in the phase image of the **DTDCTB**: C_{70} film (Figure S6). The effective phase separation may provide carrier transportation pathways and contribute to high quantum efficiencies.

In conclusion, a novel D–A–A-type donor material, **DTDCTB**, in which an electron-donating ditolylaminothienyl moiety is connected to an electron-withdrawing dicyanovinylene moiety through another electron-accepting 2,1,3-benzothiadiazole block, has been synthesized and applied in the fabrication of vacuum-deposited SMOSCs. The innovative structural design strategy enables **DTDCTB** to exhibit distinguished light-harvesting abilities with spectral responses close to the near-IR region. A vacuum-deposited SMOSC employing **DTDCTB** as the electron donor and C₇₀ as the electron acceptor demonstrated an exceptional PCE of up to 5.81% in initial trials. This efficiency is among the highest ever obtained for organic vacuum-deposited single cells. The high efficiency is primarily attributed to the broad and intensive absorption (giving high J_{sc}) and a reasonably low-lying HOMO level (giving high V_{oc}) of the **DTDCTB** thin film. Our results may advance efforts to develop new organic dyes to boost the device performance of SMOSCs.

■ ASSOCIATED CONTENT

S Supporting Information. Synthesis, characterization, copies of ¹H and ¹³C NMR spectra, CIF for **DTDCTB**, procedures for fabricating devices and performing measurements, quantum-mechanical calculations, an atomic force microscopy image, and an X-ray diffractogram. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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■ ACKNOWLEDGMENT

We thank the National Science Council of Taiwan (NSC 98-2112-M-007-028-MY3 and NSC 98-2119-M-002-007-MY3) and the Low Carbon Energy Research Center, National Tsing-Hua University, for financial support. We are also grateful to the National Center for High-Performance Computing for computer time and facilities.

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