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A new A2-π-A1-π-A2-type small molecule denoted as BDD-IN was synthesized employing a strong electron-withdrawing unit, 3bis(4-(2-ethylhexyl)-thiophen-2-yl)-5,7-bis(2ethylhexyl)benzo[1,2-:4,5-c']-dithiophene-4,8-dione (BDD) as the central acceptor unit (A₁), thiophene-alkoxy benzene-thiophene as the π -bridge, and indenedione (IN) as the end-capped unit (A2). An optimal power conversion efficiency (PCE) of 8.70% with a high open-circuit voltage (Voc) of 0.965 V and a high fill factor (FF) of 72.3% was achieved.

organic solar cells

Organic solar cells (OSCs) have emerged as a promising renewable energy source to address the energy crisis and environment-related issues.¹ Through great efforts devoted into the photoactive materials innovation and device optimization, power conversion efficiency (PCE) of over 14% has been achieved for single-junction polymer non-fullerene solar cells.² Compared with polymers, small molecules have many unique advantages, such as well-defined molecular structure and high reproducibility, simple synthesis and so on.³ And small molecules show great potential for high performance solar cells.⁴ Among various types of small molecules containing donor-acceptor (D-A) structure, A-π-D-π-Atype small molecules have presented outstanding photovoltaic performance whether used as electron donors or electron acceptors, which incorporate an electron-donating unit as the central core, two conjugated π bridges, and two electron-accepting terminal units.⁵ For example, Chen's group reported a small molecule DR3TSBDT with dialkylthiol substituted BDT as the core and 3-ethylrhodanine as the end-capped unit which exhibited a high PCE of 9.95%.⁶ Wei's group reported a suit of small molecules which incorporated thiophene-substituted benzodithiophene as the core and fluorinated 1H-indene-1,3(2H)-dione as the end-capped units, among which a high PCE of 11.08% was achieved.⁷ In addition, since Zhan's group reported ITIC based on a bulky fusedring core (indacenodithieno[3,2-b]thiophene) end-capped with 2-(3oxo-2,3-dihydroinden-1-ylidene)malononitrile unit, a large number of chemists focused on the A-D-A or A-π-D-π-A-type non-fullerene acceptors, and excellent photovoltaic performance has been obtained.8

Then how about substituting the central electron-donating unit with an electron-accepting unit, namely A₂-π-A₁-π-A₂-type small molecules?9 It has been demonstrated that the open-circuit voltage $(V_{\rm oc})$ mainly depends on the energy level difference between the highest occupied molecular orbital (HOMO) level of the donor materials and the lowest unoccupied molecular orbital (LUMO) level of the acceptor materials.^{3a, 3b} Previous researches indicate that the HOMO energy level and the LUMO energy level are generally dependent on the electron-donating ability of the donor units and the electron-withdrawing ability of the acceptor units, respectively.^{5, 10} Therefore, it is expected that larger V_{oc} could be obtained via increasing the electron-accepting ability of the whole molecule when the small molecule is used as the donor materials. 3-Bis(4-(2-ethylhexyl)-thiophen-2-yl)-5,7-bis(2ethylhexyl)benzo[1,2-:4,5-c']-dithiophene-4,8-dione (BDD) unit is a strong electronwithdrawing and planar group, which is widely used for constructing D-A alternate copolymer donors.¹¹ Notably, the polymer PBDB-T based on BDD unit developed by Hou's group has exhibited excellent performance as wide bandgap donors in nonfullerene polymer solar cells.12

Incorporating noncovalent intramolecular interactions into molecular backbone is an effective way to enhance chain coplanarity, which could lead to enhanced molecular order beneficial for exciton diffusion and charge carrier transport.13 Specially, noncovalent intramolecular O····S interactions between alkoxy substituents and thiophene rings have been demonstrated to be effective for minimizing torsional angles within polymer backbones.¹⁴ Herein, we designed and synthesized a new A_2 - π - A_1 - π - A_2 -type small molecule BDD-IN which incorporated BDD as the central building block (A1),

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thiophene-alkoxy benzene-thiophene as the π -bridge and indenedione (IN) as the terminal units (A₂). Its chemical structure was depicted in Fig. 1a. Bulk-heterojunction OSCs using BDD-IN as the donor and [6,6]-phenyl-C71-butyric acid methyl ester (PC₇₁BM) as the acceptor were prepared and the photovoltaic properties were optimized. A high PCE of 8.70% was achieved along with a high V_{oc} of 0.965 V, a short-circuit current density (J_{sc}) of 12.44 mA cm⁻² and a high fill factor (FF) of 72.3%.

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The target molecule could be obtained via Stile crosscoupling reaction and Knoevenagel condensation within a few steps, and the detailed synthetic procedures and characterization data are provided in the Electronic Supplementary Information. The chemical structure of BDD-IN was confirmed by NMR spectroscopy and mass spectrometry (Fig. S1-S12). The compound showed good solubility in common organic solvents, such as chloroform, chlorobenzene. BDD-IN shows good thermal stability with the decomposition temperature (T_d, 5% weight loss) of 351 °C under N₂ atmosphere (Fig. S13a). Clear melting temperatures (T_m) of 216 °C upon heating and recrystallization points (T_{cr}) of 197 °C upon cooling for the compound could be observed on the differential scanning calorimeter (DSC) curves, indicative of its crystallinity (Fig. S13b).

The UV-Vis absorption spectra of BDD-IN in chloroform solution and thin film are illustrated in Fig. 1b. The absorption of BDD-IN in diluted CHCl₃ solution exhibits a peak at 532 nm. The maximum extinction coefficient is 1.12×10^5 M⁻¹ cm⁻¹. From solution to solid state, the absorption spectrum was broadened and red-shifted with maxima at 569 nm. Besides, there exists a pronounced shoulder peak at 616 nm, suggesting effective π - π stacking between molecular backbones. The optical band gap of BDD-IN is 1.83 eV calculated from the absorption edge. Theoretical calculation by density functional theory (DFT) at the B3LYP/6-31G(d) level were also performed to evaluate the distribution of the frontier molecular orbitals and the molecular geometries. As indicated from Fig. S14, the electron densities of HOMO levels are mainly delocalized along the entire molecular backbone except the end-capped groups, while the electron densities of the LUMO levels are delocalized along the entire molecular backbone. The extended LUMO wave function results in enhanced overlap with the HOMO wave function, which could contribute to its higher extinction coefficients.¹⁵ From Fig. S15, BDD-IN showed a dihedral angle of 21.62° between the BDD unit and its adjacent thiophene unit. The indicated dihedral angle might break the great aggregation tendency of fully planar molecules, which would contribute to the nanoscale bicontinuous interpenetrating network morphology.¹⁶

The electrochemical properties of the compound were studied by cyclic voltammograms (Fig. 1c). The potentials were internally calibrated using ferrocene/ferrocenium (Fc/Fc⁺) redox couple (4.8 eV below the vacuum level). The HOMO and LUMO levels calculated from the onset oxidation and reduction potentials are -5.40 and -3.51 eV, respectively. The electrochemical band gap of BDD-IN is estimated to be 1.89 eV, which is consistent with its optical band gap. The lower HOMO

level indicates that larger V_{oc} could be expected by using the compound as donor materials.



Fig. 1 (a) Chemical structure of BDD-IN. (b) UV-vis absorption spectra of BDD-IN in solution and thin film. (c) Cyclic voltammogram of BDD-IN in tetrahydrofuran solution with 0.1 mol L^{-1} n-Bu₄NPF₆ at a scan rate of 100 mVs⁻¹.

To evaluate the photovoltaic performance of BDD-IN, conventional devices using BDD-IN as the donor material with the configuration of ITO/PEDOT:PSS/BDD-IN:PC71BM/PrC60MA/ Al were fabricated, where PrC₆₀MA represented a fulleropyrrolidinium iodide derivative material developed by Alex K.-Y and its structure is shown in Fig. S16.17 Typical current density-voltage (J-V) curves of the as-cast and the optimized devices are illustrated in Fig. 2a. After a series of optimization, it was found that solvent vapour annealing (SVA) treatment could deliver the optimal performance (Table S1-S4). Without any treatment, the devices delivered a very low PCE of 1.70%, with a V_{oc} of 1.042 of V, a J_{sc} of 5.75 mA cm⁻² and an FF of 28.3%. The high $V_{\rm oc}$ is attributed to the low HOMO energy level of BDD-IN. After SVA treatment, the PCE was sharply enhanced to 8.70%, with a high V_{oc} of 0.965 V, a J_{sc} of 12.44 mA cm⁻² and a high FF of 72.3%. The reduction in the V_{oc} after SVA treatment may be due to the increasing intermolecular interaction between the donor and acceptor.18 The strength of intermolecular interactions could be represented by J_{so}. As shown in Figure S18 and Table S5, in

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comparison with the as-cast devices, the devices with SVA treatment exhibit a higher J_{so}, which is in accordance with the decreased V_{oc} . Notably, the FF is up to 76.3% when the donor concentration is 8 mg ml⁻¹ (Table S1). The FF values are higher than those of previously reported similar molecules.^{9a, 9b, 15} The external quantum efficiency (EQE) spectra of the as-cast and optimized devices are shown in Fig. 2b. From Fig. 2b, the ascast devices exhibit relatively low EQE response with maximum value of 37.5%. After SVA treatment, uniform enhancement of the spectral response across the 300-700 nm wavelength regions are clearly observed and the maximum value is 76.1%. The calculated J_{sc} estimated from the integration of the EQE curves are 5.51 and 11.90 mA cm⁻² for the as-cast and the optimized devices, respectively, which are in agreement with the values got from the J-V curves within an acceptable deviation of ~5%.



Fig. 2 (a) Typical *J-V* curves and (b) EQE curves of the as-cast and SVA-treated devices based on BDD-IN.

To investigate the charge generation, collection and recombination behaviours, the photocurrent density (J_{ph}) as a function of effective voltage (V_{eff}) is plotted in double logarithmic coordinates shown in Fig. 3. The J_{ph}/J_{sat} ratio can be employed to evaluate the overall exciton separation and charge collection efficiency. Under the short-circuit conditions, the J_{ph}/J_{sat} ratios of the as-cast and optimal devices are 48% and 97%, respectively. At the maximal power output conditions, the J_{ph}/J_{sat} ratios of the as-cast and optimal devices are 25% and 83%, respectively. These results suggest higher exciton dissociation and charge collection efficiency, and less bimolecular recombination for devices with SVA treatment, thus enhanced J_{sc} and FF.



Fig. 3 Variation of J_{ph} with V_{eff} for the as-cast and SVA-treated BDD-IN:PC₇₁BM based devices.

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To understand the effect of SVA treatment on the device performance, atomic force microscopy QAFM 102245915882565 characterize the morphologies of the active layers. From Fig. 4a and 4b, the surface root-mean-square (RMS) roughness for the as-cast and SVA-treated BDD-IN:PC71BM blend films are 0.67 and 0.79 nm, respectively. The blend films under different conditions all showed nano-scaled phased separation, which may benefit from the twisted backbones of the small molecule donor compared with previously reported molecules with nearly planar backbone.^{9b, 15} Nevertheless, for the as-cast film, there are many aggregates with size about 50 nm, which are larger than the exciton diffusion length and adverse to exciton dissociation, and thus lead to poor J_{sc}. The discontinuous phase aggregation could cause severe bimolecular recombination, which leads to low FF. After SVA treatment, the largely segregated donor and acceptor phases became adequately intermixed to form a good interpenetrating network, which could be beneficial for exciton dissociation and charge transport, and therefore J_{sc} and FF were greatly enhanced. Besides, from the X-ray diffraction (XRD) patterns for the neat BDD-IN film (Fig. S19a), BDD-IN exhibited a very weak diffraction peak (100) at 2θ = 5.57°. After SVA treatment, a distinct diffraction peak (100) at $2\theta = 5.73^{\circ}$ was observed. Similarly, for the as-cast BDD-IN:PC71BM blend film, no obvious diffraction peaks appeared. However, a diffraction peak at 20 = 5.60° was present after SVA treatment (Fig. S19b). These results indicate that SVA treatment enhanced the crystallinity of BDD-IN, which are good for exciton diffusion and charge transport.



Fig. 4 AFM height images of BDD-IN:PC_71BM blend films (a) as cast (b) SVA treatment. The scale bar is 5.0 $\mu m.$

The hole mobility (μ_h) and electron mobility (μ_e) of the blend films were tested by the SCLC method (Fig. S20).¹⁹ For the as-cast devices, the hole/electron mobilities are 2.48 × 10⁻⁴ and 7.49 × 10⁻⁴ cm² V⁻¹ s⁻¹, respectively. For the SVA-treated devices, the hole/electron mobilities are 1.83 × 10⁻³ and 5.33 × 10⁻³ cm² V⁻¹ s⁻¹, respectively. After SVA treatment, higher mobilities are good for efficient charge transport and thus excellent J_{sc} and FF.

In conclusion, a new A_2 - π - A_1 - π - A_2 -type small molecule BDD-IN using BDD with strong electron-accepting ability as the central unit was designed and synthesized. This compound exhibited strong absorption in the wavelength range of 300-700 nm and deeper HOMO energy levels. After SVA treatment, the BDD-IN:PC₇₁BM based devices delivered a high PCE of 8.70%, with a high V_{oc} of 0.965 V, a J_{sc} of 12.44 mA cm⁻² and a

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high FF of 72.3%. The results suggest that the A_2 - π - A_1 - π - A_2 -type small molecules are very promising to construct more efficient photovoltaic devices.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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A new $A_2-\pi - A_1-\pi - A_2$ -type small-molecule donor using a strong electron-withdrawing unit as the system of t