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An acceptor-donor-acceptor type non-fullerene acceptor with an asymmetric backbone for high performance organic solar cells[†]

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An A–D–A type acceptor CC10 with an asymmetric D unit has been designed and synthesized by introducing an alkylbenzene unit into a symmetric acceptor CC5. Compared with the symmetric CC5, the asymmetric CC10 showed a similar absorption range and energy level, but better π – π stacking, enhanced electron mobility and optimized microscopic morphology. As a result, the CC10-based organic solar cells demonstrated a high PCE of 11.78%, better than that of 6.91% for CC5-based devices. The significantly improved device performance caused by only a minor modification in the molecular backbone indicates that there is substantial potential of the asymmetric strategy for designing high performance active layer materials.

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1. Introduction

Organic solar cells (OSCs) have drawn much attention over the past decade due to the advantages of light weight, low cost, and mechanical flexibility.^{1–7} Recently, great progress has been made in OSCs with power conversion efficiencies (PCEs) of 16–17%,^{8–17} which is mainly attributed to the rapid development of active layer materials, especially non-fullerene small molecule acceptors (NF-SMAs).^{18–23} Among various NF-SMAs, molecules with an acceptor-donor–acceptor (A–D–A) architecture have demonstrated great success since their molecular electronic and optical properties and even solid packing can be effectively tuned through careful chemical structure optimization.^{24–31} Presently, most of NF-SMAs with A–D–A structure have a symmetric D unit.

In contrast, NF-SMAs with asymmetric D cores are expected to exhibit larger dipole moment and stronger intermolecular binding energy than symmetric NF-SMAs, which may contribute to forming ordered molecular packing with intense intermolecular interaction, and thus a high fill factor (FF) and PCE for OSC devices might be expected.^{32–35} Therefore, the study of A–D–A acceptors with asymmetric D cores has drawn much attention, which presents a possible strategy for the design and optimization of A-D-A molecules aiming to maintain the advantages of symmetric NF-SMAs and bring further efficiency enhancement.³⁶⁻⁴² For example, Sun et al.43 employed a cutting strategy to design an asymmetric acceptor TTPT-T-2F. Compared with the control symmetric acceptor IT-2F, TTPT-T-2F showed red-shifting absorption, elevated molecular energy levels, improved electron mobility and better OSC performance. Yang et al.44 reported an asymmetric acceptor a-BTTIC by flanking one side of D cores with one thiophene from a symmetric acceptor BTTIC. The asymmetric acceptor a-BTTIC based device showed favourable micromorphology and reduced bimolecular recombination compared with that of the control symmetric molecule BTTIC. Zhan et al.45 reported a fused-8ring based NF-SMA AOIC through a unidirectional extension strategy. The unidirectional extension increases the molecular dipole moment and dielectric constant, as well as reduces the exciton binding energy. In addition, an asymmetric A-D-A acceptor a-IT-2OM with different end groups has also been reported by Bo et al. and a promising device performance of 12.07% was achieved.46 These results have demonstrated that an asymmetric molecule might be a promising strategy for the design of high efficiency active layer materials.

With these in mind, we herein reported an asymmetric acceptor CC10 by tuning the molecular backbone of the symmetric acceptor CC5.^{47,48} As shown in Fig. 1, the only difference between CC5 and CC10 is an alkylbenzene unit on the backbone. Compared with the symmetric CC5, the asymmetric CC10 possesses a similar absorption range and energy levels but greatly different molecular packing and blend microscopic morphology. The optimized PM6:CC10 based device showed a PCE of 11.78%, with an open-circuit voltage (V_{oc}) of 0.771 V,

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Fig. 1 (a) Chemical structure of CC5 and CC10; (b) chemical structure of PM6; (c) energy level diagrams for PM6, CC5 and CC10.

a short-circuit current (J_{sc}) of 22.70 mA cm⁻² and a fill factor (FF) of 0.673. In contrast, a PCE of 6.91% was obtained for PM6:CC5 based device with a V_{oc} of 0.652 V, a J_{sc} of 18.03 mA cm⁻² and a FF of 0.588. It has been found that the asymmetric acceptor CC10-based device manifested more enhanced and balanced charge transport properties, favourable morphology and energy transfer process, and thus achieved a higher FF, J_{sc} and V_{oc} , compared with that of the symmetric molecule CC5-based device.

2. Results and discussion

2.1 Synthesis and characterization

As depicted in Scheme 1, the synthetic routes towards the corresponding central units (TT-TT and BDT-TT) are as simple as only a two-step procedure (Stille cross-coupling and Friedel–Crafts intramolecular cyclization). The detailed synthesis procedures and characterization data are summarized in the ESI.† The subsequent Vilsmeier–Haack reaction afforded the corresponding dialdehyde intermediates in high yields. The targeted molecules CC5 and CC10 were finally synthesized *via* Knoevenagel condensation using bis(5,6-dichloro-3-oxo-2,3-dihydro-1*H*-indene-2,1-diylidene)dimalononitrile (IC-2Cl). CC5 and CC10 show good solubility in common solvents such as chloroform and

chlorobenzene and are fully characterized by nuclear magnetic resonance (NMR) and high-resolution Fourier transform mass spectrometry (HR-FTMS).

2.2 Optical and electrochemical properties

The UV-vis absorption spectra of CC5 and CC10 in chloroform solution and thin films are provided in Fig. S3 (ESI⁺) and Fig. 3a, and the detailed data are summarized in Table 1. In chloroform solution, asymmetric CC10 exhibited a maximum absorption peak located at 736 nm, which is slightly red-shifted by 11 nm compared with that of CC5 (725 nm). From the solution to the film state, CC10 demonstrates less red shifted absorption (736 to 797 nm) than CC5 (725 to 800 nm). As a result, CC5 and CC10 show similar absorption peaks and absorption edges, corresponding to the similar optical bandgaps of 1.37 and 1.38 eV, respectively. Cyclic Voltammetry (CV) measurements were carried out to investigate the electrochemical properties of the two molecules (Fig. S4, ESI⁺), and the corresponding energy level diagram is shown in Fig. 1c. The highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) energy levels are summarized in Table 1. The LUMOs/HOMOs of CC5 and CC10 were estimated to be -4.09/-5.75 and -4.07/-5.72 eV, respectively. Clearly, compared with the symmetric CC5, the asymmetric CC10 shows no clear difference in the UV-vis absorption range and energy levels at the molecular level.

2.3 Theoretical calculation

Density functional theory (DFT) (B3LYP/6-31G*) calculations were applied to evaluate the molecular geometry and the frontier molecular orbitals. To simplify the calculations, we replaced the alkyl chains by a methyl group. The optimized electron-state density distributions in the molecular orbitals (LUMO and HOMO) are shown in Fig. S2 (ESI†). The ground state dipole moment (μ_g) for CC5 and CC10 is 2.48 D and 2.52 D, respectively. The dipole moment direction of CC5 is parallel to the molecular plane and forms an angle of $\approx 90^{\circ}$ with the molecular long axis. However, the dipole moment direction of CC10 has an angle of $\approx 11^{\circ}$ with the molecular long axis, which is induced by the asymmetry of the donor core. And the excited state dipole moment (μ_e) for CC5 and CC10 is 2.37 D and 6.80 D, respectively.



Scheme 1 Synthetic route towards CC5 and CC10.

 Table 1
 The optical and electrochemical properties of CC5 and CC10

	λ_{\max}^{CF} (nm)	$\lambda_{\max}^{\mathrm{film}}$ (nm)	$\lambda_{ m edge}^{ m film}$ (nm)	HOMO (eV)	LUMO (eV)	$E_{\rm g}^{\rm CV}$ (eV)	$E_{ m g}^{ m opt}$ (eV)
CC5 CC10	725 736	800 797	908 899	$\begin{array}{c} -5.75 \\ -5.72 \end{array}$	$\begin{array}{c} -4.09 \\ -4.07 \end{array}$	$\begin{array}{c} 1.66 \\ 1.65 \end{array}$	1.37 1.38

The change in the dipole moment from the ground to the excited state ($\Delta \mu_{ge}$) for CC5 and CC10 is 0.11 D and 7.97 D, respectively. The slightly larger ground state μ_g of CC10 is beneficial to reinforce intermolecular interaction and the larger $\Delta \mu_{ge}$ indicates enhanced intramolecular charge transfer properties, which are expected to increase FF and PCEs.32,49 To gain further understanding of the transport channels for the two acceptors, we studied the local intermolecular π - π stacking between terminal acceptor units by Gaussian 16.⁵⁰ Gimme's D3 dispersion correction with Becke-Johnson damping was used during the optimization. The corresponding intermolecular binding energies were calculated by correction of the basis set superposition error (BSSE) according to the literature method.³⁵ Packing geometry configurations of the two dimer molecules and their corresponding intermolecular binding energies are displayed in Fig. 2. The CC5 dimer exhibited only one kind of packing form, while there were three packing forms for the asymmetric CC10 dimer. Moreover, the CC10 dimer showed two stronger intermolecular binding energies and two more stable geometry configurations than the CC5 dimer, suggesting that CC10 demonstrates enhanced π - π stacking in theoretical calculation and will possess higher electron mobility than CC5.

2.4 Photovoltaic device performance

Inverted devices with the structure of ITO/ZnO/PFN-Br/active layer/MoO_x/Ag were fabricated and optimized to evaluate the photovoltaic properties using CC5 and CC10 as acceptors. The wide-bandgap polymer PM6⁵¹ was employed as the donor due



Fig. 2 Optimized geometries and the corresponding intermolecular binding energies by DFT calculations of CC5 and CC10 dimers.



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Fig. 3 (a) Normalized UV-vis absorption spectra of PM6, CC5, and CC10 in thin film; (b) EQE curves, (c) current density–voltage (J-V) curves and (d) photocurrent *versus* effective voltage $(J_{ph}-V_{eff})$ characteristics of CC5- and CC10-based devices.

to its matched energy level and complementary absorption with CC5 and CC10. The device parameters are summarized in Tables S1-S6 (ESI⁺), including the donor/acceptor weight ratio, additive, etc. The optimized weight ratio was 1:0.7 for PM6: CC5 and 1:1.2 for PM6:CC10. The current density-voltage (J-V) curves of the optimized devices and their corresponding external quantum efficiency (EQE) are presented in Fig. 3, and the related photovoltaic data are summarized in Table 2. The optimized PM6:CC5 based cells showed a PCE of 6.91%, with a $V_{\rm oc}$ of 0.652 V, a $J_{\rm sc}$ of 18.03 mA cm⁻² and a FF of 0.588. In contrast, the optimized PM6:CC10 based OSCs achieved a higher PCE of 11.78%, with the simultaneously improved $V_{\rm oc}$ of 0.771 V, $J_{\rm sc}$ of 22.70 mA cm⁻² and FF of 0.673. It is worth noting that the asymmetric CC10-based device showed a lower energy loss $(E_{loss} = E_g^{opt} - eV_{oc})$ with a value of 0.61 eV compared with that of the CC5-based device with a value of 0.72 eV. In addition, despite the similar absorption range, the CC10-based OSC achieved a larger I_{sc} than the CC5-based device, which can be observed from their EQE results shown in Fig. 3b. The CC10based OSCs showed clearly higher EQE values than CC5-based OSCs from 300 to 900 nm. The PM6:CC10 based device showed the maximum EQE value of 77% at 800 nm, which is higher than that of the PM6:CC5 based device (63% at 630 nm). The integrated current density from the EQE measurements was 21.75 mA cm⁻² for the PM6:CC10 based devices and 17.46 mA cm⁻² for the PM6:CC5 based devices, which agrees well with the values from J-V measurements.

2.5 Device characterization

To study the charge transport properties, the space-charge-limited current (SCLC) method was employed to measure the charge mobility of the blend films of PM6:CC5 and PM6:CC10 with the electron-only and hole-only devices, respectively (Fig. S5, ESI†).⁵² The electron and hole mobility of the PM6:CC5 blend film are calculated to be 5.18×10^{-5} and 1.48×10^{-4} cm⁻² V⁻¹ s⁻¹, respectively. On the other hand, the electron and hole mobility of

Table 2 The optimized photovoltaic data of CC5- and CC10-based devices under the illumination of AM 1.5G (100 mW cm⁻²)

Devices	$V_{\rm oc}{}^a$ (V)	FF^{a}	$J_{ m sc}{}^a ({ m mA}~{ m cm}^{-2})$	PCE^{a} (%)	$J_{ m sc}^{ m EQE}~({ m mA~cm^{-2}})$
PM6:CC5 PM6:CC10	$\begin{array}{c} 0.650 \pm 0.006 \ (0.652) \\ 0.775 \pm 0.004 \ (0.771) \end{array}$	$\begin{array}{c} 0.597 \pm 0.039 \ (0.588) \\ 0.657 \pm 0.016 \ (0.673) \end{array}$	$\begin{array}{c} 17.09 \pm 0.94 \ (18.03) \\ 22.64 \pm 0.44 \ (22.70) \end{array}$	$\begin{array}{c} 6.63 \pm 0.28 \ (6.91) \\ 11.52 \pm 0.26 \ (11.78) \end{array}$	(17.46) (21.75)

^a The PCE value was calculated from 20 devices; the values in parentheses are the parameters of the best CC5- and CC10-based devices.

the CC10-based blend film increase to 1.02×10^{-4} and 1.60×10^{-4} cm⁻² V⁻¹ s⁻¹, respectively. In comparison with a μ_h/μ_e ratio of 2.86 for the CC5-based device, the CC10-based device had a more balanced μ_h/μ_e ratio of 1.57. The higher and more balanced charge transport properties can promote charge transport and suppress bimolecular recombination, which contribute to the higher FF and J_{sc} of the PM6:CC10 based devices.

As shown in Fig. 3d, the plots of photocurrent density $(J_{\rm ph})$ *versus* effective voltage $(V_{\rm eff})$ were measured to investigate the exciton dissociation and charge collection properties.^{53,54} Herein, $J_{\rm ph} = J_{\rm L} - J_{\rm D}$, where $J_{\rm L}$ and $J_{\rm D}$ are photocurrent densities under illumination and dark conditions, respectively. $V_{\rm eff} = V_0 - V_{\rm bias}$, where V_0 is the voltage at which $J_{\rm ph}$ is zero and $V_{\rm bias}$ is the applied external bias voltage. The value of $P_{\rm diss}$ ($P_{\rm diss} = J_{\rm ph}/J_{\rm sat}$, where $J_{\rm sat}$ is the saturation photocurrent density) represents charge dissociation and collection probability. The $P_{\rm diss}$ values for CC5and CC10-based OSCs were calculated to be 89.39% and 95.82%, respectively. These results indicate that the CC10-based device offers more efficient exciton dissociation.

To further study the behaviour of charge recombination of the two devices, the plots of light-intensity dependence (P) of J_{sc} $(J_{\rm sc} \propto P^{\alpha})$, where the exponent of α being close to 1 reflects a weak bimolecular recombination) were measured and are displayed in Fig. S6 (ESI[†]). The J_{sc} values of CC5- and CC10-based devices were highly linearly correlated with P, with α values of 0.894 and 0.958, respectively. The above results illustrate that devices based on the asymmetric CC10 showed more efficient charge dissociation and less bimolecular recombination. By plotting Voc versus light intensity, the trap-assisted recombination behaviours were investigated. CC5-based OSCs feature a slope of 1.266 KT/q, whereas a smaller slope of 1.175 KT/q was generated for CC10-based OSCs (Fig. S6, ESI[†]). The smaller slope for the asymmetric NF-SMA is indicative of weak trap-assisted recombination, which suggests that CC10-based OSCs possess less trap-assisted recombination, resulting in a higher level of FF and J_{sc} .

From the perspective of mobility, bimolecular recombination and trap-assisted recombination, positive effects on the device working process occurred for the asymmetric acceptor CC10based device, indicating that modulation of the backbone of CC5 by introducing an alkylbenzene unit is indeed favourable for improving the device efficiency.

2.6 Morphology analysis

Atomic force microscopy (AFM) and transmission electron microscopy (TEM) were used to characterize the difference between the two symmetric and asymmetric acceptor CC5- and CC10-based blend films. From the AFM images in Fig. 4, both blend films had smooth surface morphologies, with root-mean-square roughness (R_q) values of 1.63 and 3.58 nm for CC5- and CC10-based blend films, respectively. The larger R_q of PM6:CC10 blend films should be attributed to the enhanced molecular aggregation of the asymmetric acceptor, which is consistent with the X-ray scattering results as will be discussed below. In addition, compared with the PM6:CC5 blend film, the PM6:CC10 blend film exhibited a more obvious interpenetrating network with nanoscale phase separation. The distinctive morphologies for the PM6:CC10 blend film are highly favourable for exciton dissociation and charge transport, which result in the improvement of J_{sc} and FF and thus enhancement of the device performance for the asymmetric acceptor based OSCs.

Grazing incidence wide-angle X-ray scattering (GIWAXS) was used to further study the influence of molecular stacking characteristics for these two acceptors.^{55,56} As shown in Fig. S7 (ESI†), in the neat film, CC10 shows a clear π - π stacking diffraction peak (010) at 1.84 \AA^{-1} with a distance of 3.51 \AA in the out of plane (OOP) direction. On the other hand, there is no π - π stacking diffraction for the CC5 neat film. The result shows that the asymmetric structure of CC10 makes it tend to form an ordered packing with efficient π - π stacking. The 2D GIWAXS patterns of PM6:CC5 and PM6:CC10 optimized blend films are shown in Fig. 4e and f, and the corresponding in-plane and out-of-plane line-cut profiles are shown in Fig. S7 (ESI⁺). Both the blend films exhibit a lamellar diffraction peak (100) along the IP direction and a π - π stacking diffraction peak (010) in the OOP direction. The PM6:CC5 blend film showed 100 and 010 peaks at 0.31 \AA^{-1} and 1.80 Å⁻¹, corresponding to an alkyl-to-alkyl distance of 20.13 Å and a π - π stacking distance of 3.58 Å, respectively. As for the PM6:CC10 blend film, the significantly sharper and stronger 100 and 010 diffraction peaks are observed at 0.32 $Å^{-1}$



Fig. 4 TEM images for (a) PM6:CC5 and (b) PM6:CC10 optimized blend films. AFM images for (c) PM6:CC5 and (d) PM6:CC10 optimized blend films. 2D-GIWAXD diffraction images of (e) PM6:CC5 and (f) PM6:CC10 optimized blend films.

and 1.86 Å⁻¹, corresponding to the smaller alkyl-to-alkyl distance and π - π stacking distance of 19.5 Å and 3.42 Å, respectively. The crystal coherence length (CCL) in the (010) diffraction direction was estimated to be 17.27 Å for the CC5-based blend film and 21.96 Å for the CC10-based blend film. These results indicate the enhanced crystallinity and intense molecular packing in the PM6:CC10 blend film, which is consistent with the theoretical simulation results and mobility measurements as discussed above.

3. Conclusions

In this work, we have designed and synthesized an asymmetric acceptor CC10 by tuning the molecular backbone of a symmetric acceptor CC5. With similar absorption and energy levels as CC5, the asymmetric acceptor CC10 has an ordered packing with efficient π - π stacking, improved active layer morphology, and higher and balanced charge mobility. Consequently, quite different device performances have been obtained for these two acceptor-based devices. The asymmetric acceptor CC10-based device achieved a higher PCE of 11.78% compared to the CC5based device with a PCE of 6.91%. These results demonstrate that modification of the A-D-A acceptor with an asymmetric molecular backbone D unit has great potential to design and achieve high efficiency new active layer materials. It is believed that higher efficiency asymmetric A-D-A molecules including donor and acceptor could be obtained through careful design based on the great success of symmetric A-D-A molecules.

Conflicts of interest

There are no conflicts of interest to declare.

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