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## Introduction

Organic solar cells (OSCs) with bulk-heterojunction (BHJ) structures have developed rapidly over the past decade owing to their advantages such as light weight, low cost, mechanical flexibility, *etc.*<sup>1-9</sup> The adequate pairing of newly designed donor and acceptor materials with additional device optimizations leads to a significant boost in the power conversion efficiencies (PCEs).<sup>10-16</sup> Compared with traditional fullerene derivatives as acceptor materials, non-fullerene acceptors (NFAs), especially those with the acceptor-donor-acceptor (A–D–A) architectures, with easily tuned energy levels and broad absorption have gained great attention and developed rapidly in recent years.<sup>17–23</sup> With the precise structure design of NFAs and suitable selection of donor materials with matched energy levels, absorptions, *etc.*, the PCEs of NFA based OSCs have reached 12–13%.<sup>24–28</sup> Although these inspiring results have been obtained, detailed

# Fine-tuning the side-chains of non-fullerene small molecule acceptors to match with appropriate polymer donors†

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Side-chain engineering of donor and acceptor materials is an important topic in the field of organic photovoltaics. The influence of side-chains in active layer molecules on the corresponding photovoltaic device performances is still elusive, especially for the devices based on non-fullerene small molecule acceptors. In this work, we designed and synthesized two non-fullerene acceptors (IDTT-BH and IDTT-OBH) using an indacenodithieno[3,2-b]thiophene (IDTT) moiety as the central building block and (2-3-oxo-2,3-dihydro-1H-cyclopenta[b]naphthalen-1-ylidene)malononitrile (NINCN) as the end-group, which have similar energy levels and optical absorption spectra, and differ in the side-chains. We systematically investigated the effect of the side-chains on the device performance based on these two non-fullerene acceptors pairing with different donor materials. For the devices with J71 and PDCBT as donor materials, IDTT-BH showed better PCEs of 11.05% and 10.35%, respectively. Notably, 10.35% efficiency is among the top values in PDCBT-based OSCs. While, the devices based on PBDB-T:IDTT-OBH showed a better PCE of 10.93% than that of IDTT-BH based devices. It was found that the side-chains of non-fullerene acceptors have an effect on tuning the morphology of blend films and thus affect the photovoltaic performance, and different donor materials should be paired with the acceptors with the most suitable side-chains to achieve the best photovoltaic performance.

> information about the relationship between the chemical structure of NFAs and device performance needs to be thoroughly investigated.

> Basically, from the viewpoint of the chemical structure of NFAs,<sup>29-43</sup> three key factors should be considered, *i.e.* central aromatic fused rings to guarantee the charge transport, electron-withdrawing end-groups to adjust the energy level of the lowest unoccupied molecular orbital (LUMO), and bulky side-chains to prevent over self-aggregation and provide the solubility for solution processing. Compared with the central rings and end-groups, the effect of side-chains of A-D-A type NFAs on the device performance of OSCs has been rarely investigated.44-49 Li et al. have reported an NFA m-ITIC in which side-chain isomerization with meta-alkyl-phenyl substitution govern the morphology of the blend film. Compared to ITIC, a significant improvement of PCE was achieved for *m*-ITIC based devices.<sup>50</sup> Recently, a series of NFAs (ITIC-SC6, ITIC-SC8 and ITIC-SC2C6) were designed by the Bo group. They focused on investigating the effect of side-chains on the energy levels of acceptors, the film morphologies and the device performances.<sup>51</sup> These pioneering results reveal the fact that side-chain engineering on NFA molecules is a convenient and effective way to further improve the device PCEs. In addition, the effect of side-chains on the device

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performance is still elusive for pairing NFAs with different polymer donors.

For NFA based devices, it is a fundamental prerequisite for the acceptor materials paired with the matched donor materials to achieve better performances. Firstly, both donor and acceptor materials possess appropriate energy levels. That is, donor materials have a low-lying highest occupied molecular orbital (HOMO) energy level and acceptor materials have a high-lying LUMO energy level to obtain high open-circuit voltage  $(V_{oc})$ .<sup>52,53</sup> Indeed, the values of HOMO and LUMO can be regulated by the structural variation of donors and acceptors. As a typical and successful case, Hou and co-workers designed a new polymer donor (PBDB-T-SF) and NFA (IT-4F), and the corresponding device achieved a record high efficiency of 13.1% with a  $V_{\rm oc}$  of 0.88 V,<sup>24</sup> in which they used fluorine atoms to modify the structure of the donor material with the result of a down-shifted HOMO energy level. Secondly, donor and acceptor materials with complementary absorptions could ensure efficient harvesting of solar photons.<sup>54</sup> Meanwhile, the π-conjugation structure and end-groups of donor and acceptor materials could conveniently control the absorption region and optical bandgap to ensure the desired absorption. Based on these considerations, careful investigation of the relationship between the material structure and device performance is typically necessary.

Herein, we designed and synthesized two NFAs, named **IDTT-BH** and **IDTT-OBH**, in which an indacenodithieno[3,2-b] thiophene (**IDTT**) moiety was used as the central unit<sup>24,31,50,55,56</sup> and (2-3-oxo-2,3-dihydro-1*H*-cyclopenta[*b*]naphthalen-1-

ylidene)malononitrile (NINCN) was employed as the endgroup (Fig. 1). These two molecules have extended planar backbones with NINCN end-groups which were firstly introduced by our group<sup>57</sup> and have showed great potential for high efficiency NFA based devices.58,59 Meanwhile, in order to investigate the effect of side-chains on the device performances and other issues, we chose 4-alkylphenyl and 4alkoxyphenyl as the side-chains on IDTT-BH and IDTT-OBH, respectively. Three widely used polymer donors J71, PDCBT and PBDB-T were blended with the two NFAs to fabricate the devices and used to investigate the effect of side-chains on the morphology of the active layer and photovoltaic performance of the devices. Although the two molecules had little chemical structure difference and showed nearly the same energy levels and absorptions, they achieved different device performance when blended with the above three polymers. When J71 and PDCBT were used as donor materials, IDTT-BH based devices gave PCEs of 11.05% and 10.35%, respectively, which were higher than those of IDTT-OBH based devices with PCEs of 8.02% and 8.24%, respectively. Notably, 10.35% efficiency is among the top values in PDCBT-based OSCs. For PBDB-T as the donor material, IDTT-OBH based devices showed better results than those of IDTT-BH with the PCEs of 10.93% and 9.92%, respectively. It is found that different side-chains have significant impacts on the morphologies for the corresponding blend films, thus leading to the difference in PCEs when different polymer donors were used. Our results demonstrate that the side-chains should be carefully considered not only in the NFA design but also for the donor choice in the device fabrication.

## Result and discussion

#### Synthetic route and thermal properties of acceptor materials

The synthetic route of **IDTT-BH** and **IDTT-OBH** is shown in Scheme S1 (ESI<sup>†</sup>). Starting compound **1** was obtained in good yield according to the literature.<sup>60</sup> The dialdehyde compound **2** was prepared by the Vilsmeier–Haack reaction with POCl<sub>3</sub> and *N*,*N*-dimethylformamide (DMF). The target materials **IDTT-BH** and **IDTT-OBH** were prepared through the condensation of compound **2** with **NINCN** catalyzed by chlorotrimethylsilane in 72% and 68% yields, respectively. The chemical structure of **IDTT-BH** and **IDTT-OBH** was characterized by <sup>1</sup>H NMR, <sup>13</sup>C NMR, and mass spectra detailed in the ESI.<sup>†</sup> These two acceptors easily dissolved in chloroform (CF), dichloromethane (DCM) and *o*-dichlorobenzene (DCB).

The thermal properties of these two acceptors were obtained by thermogravimetric analysis (TGA). As shown in Fig. S1,† the 5% weight loss temperature is 392 and 372 °C for **IDTT-BH** and **IDTT-OBH**, respectively. It is indicated that these two NFAs have good thermal stability and meet the requirements of OSC device fabrications.

#### Photophysical and electrochemical properties

To investigate the photophysical properties of these two acceptors, the ultraviolet-visible (UV-vis) absorption spectra of **IDTT-BH** and **IDTT-OBH** in chloroform solution and films are depicted in Fig. 2a and b. It can be found that these two acceptors possess a strong absorption in the range of 550– 800 nm with a main absorption peak at 710 nm in dilute chloroform solution and significant red-shifts of 23 nm in films, which indicated that side-chains have little effect on the photophysical properties. The absorption onsets of **IDTT-BH** and **IDTT-OBH** in solid films are 802 and 787 nm and the corresponding optical band gaps ( $E_g^{opt}$ ) are 1.54 and 1.57 eV, respectively (Table 1). As shown in Fig. 1b, **J71**, **PDCBT** and **PBDB-T** show strong absorption in the region of 400–600 nm, and the combination of these donors and the two acceptors provides a complementary absorption in the visible and nearinfrared region, which is critical to acquire high photocurrent.

A cyclic voltammetry (CV) method was adopted to explore the effect of side-chains on the molecular energy levels of these two acceptors. The plots are displayed in ESI, Fig. S2<sup>†</sup> and the energy levels of donors and acceptors are summarized in Fig. 2c. The energy levels of HOMO/LUMO estimated from the onset oxidation/reduction potential are -5.42 eV/-3.86 eV of **IDTT-BH** and -5.41 eV/-3.86 eV of **IDTT-OBH**, respectively. The results show that the energy levels have slight changes with the variation of side-chains and similar  $V_{\text{oc}}$  will be obtained when the corresponding acceptors paired with the same donor material in the OSC devices.

#### Photovoltaic properties

Photovoltaic devices with the conventional OSC device structure of ITO/PEDOT:PSS/donor:acceptor/PDINO/Al were fabricated to



Fig. 2 (a) UV-vis absorption spectra of IDTT-BH and IDTT-OBH in chloroform solution; (b) UV-vis absorption spectra of J71, PDCBT, PBDB-T, IDTT-BH and IDTT-OBH in the solid film state. (c) Energy levels of the donors and acceptors. (d) The structure of devices.

Table 1 Optical and electrochemical data of IDTT-BH and IDTT-OBH

Compound	$\lambda_{\max}^{sol}\left(nm\right)$	$\lambda_{max}^{\mathrm{film}}\left(nm\right)$	$\lambda_{edge}^{film}\left( nm\right)$	$E_{g}^{\mathrm{opt}}\left(\mathrm{eV}\right)$	HOMO (eV)	LUMO (eV)	$E_{\mathrm{g}}^{\mathrm{CV}}\left(\mathrm{eV}\right)$
IDTT-BH	710	733	802	1.54	-5.42	-3.86	1.56
IDTT-OBH	710	733	787	1.57	-5.41	-3.86	1.55

investigate the effect of side-chains on the photovoltaic performance in different polymer donor systems. The current densityvoltage (I-V) curves of the devices are shown in Fig. 3a-c and the corresponding photovoltaic parameters are summarized in Table 2. As displayed in Table 2, the devices based on J71 as the donor material gave the PCEs of 11.05% for IDTT-BH and 8.02% for **IDTT-OBH**, respectively. The  $J_{sc}$  of 14.75 mA cm<sup>-2</sup> and FF of 61.0% for IDTT-OBH based devices were obviously lower than those of **IDTT-BH** based devices with a  $J_{sc}$  of 17.77 mA cm<sup>-2</sup> and FF of 69.1%. For the devices using PDCBT as the donor material, the result showed the same tendency. PDCBT:IDTT-BH based devices gave a PCE of 10.35% with a  $V_{\rm oc}$  of 0.88 V,  $J_{\rm sc}$  of 17.15 mA cm<sup>-2</sup> and FF of 68.6%, while a PCE of 8.24% was obtained for the PDCBT:IDTT-OBH devices with relatively lower  $J_{\rm sc}$  and FF. Notably, the PCE of 10.35% is among the top values in PDCBT-based OSCs. However, the device performances were reversed when PBDB-T was used as the donor material. The devices based on PBDB-T:IDTT-OBH delivered a PCE of 10.93% with a  $V_{\rm oc}$  of 0.87 V,  $J_{\rm sc}$  of 17.46 mA cm<sup>-2</sup> and FF of 72.0%, whose three parameters were all higher than those of PBDB-T:IDTT-BH based devices with a PCE of 9.92%. From Table 2, it was found that IDTT-OBH based devices had a slightly higher  $V_{\rm oc}$  of 0.020.03 V than that of the devices based on **IDTT-BH**, though **IDTT-BH** and **IDTT-OBH** have identical CV LUMO levels. This might originate from the different charge recombinations of the two acceptor based systems, which led to different energy losses and gave different  $V_{\text{oc}}$ .<sup>61</sup> These results indicated that the side-chains of NFAs are closely related to the performance of devices and different donor materials should be paired with the acceptors with favorable side-chains to achieve high performance.

Fig. 3d–f show the external quantum efficiencies (EQEs) of the **IDTT-BH** and **IDTT-OBH** based optimized devices. The devices exhibited broad photocurrent response region from 300 to 800 nm and it can be found that both the donors and acceptors made effective and complementary contribution to the  $J_{sc}$  values. As shown in Table 2, the  $J_{sc}$  values calculated from the EQE values were consistent with those from J-V measurements within 5% mismatch. For **J71** and **PDCBT** systems, the highest EQE values of devices based on **IDTT-BH** reached nearly 70%, which is much higher than those of **IDTT-OBH**. The photoresponse range of **PBDB-T:IDTT-OBH** was slightly broader than that of **PBDB-T:IDTT-BH** with a highest EQE value of 66.5%.

To further gain insight into the exciton dissociation and charge collection behavior, we measured the photocurrent



Fig. 3 J–V curves and EQE curves of J71:IDTT-BH/IDTT-OBH (a and d), PDCBT:IDTT-BH/IDTT-OBH (b and e), and PBDB-T:IDTT-BH/IDTT-OBH (c and f) based devices.

Table 2 Photovoltaic parameters of the OSCs based on the optimized devices of different donors and acceptors under AM1.5G illumination, 100 mW cm $^{-2}$ 

Donor	Acceptor	$V_{ m oc}$ (V)	$J_{\rm sc} ({ m mA}{ m cm}^{-2})$			PCE (%)		Thielenoor
			J-V	EQE calculated	FF (%)	Best	Average <sup><i>a</i></sup>	(nm)
J71	IDTT-BH	0.90	17.77	17.01	69.1	11.05	10.89	100
	IDTT-OBH	0.92	14.75	13.97	61.0	8.02	7.93	100
PDCBT	IDTT-BH	0.88	17.15	16.76	68.6	10.35	10.14	120
	IDTT-OBH	0.91	13.97	13.55	64.8	8.24	8.09	100
PBDB-T	IDTT-BH	0.85	16.86	16.37	69.2	9.92	9.87	90
	IDTT-OBH	0.87	17.46	16.82	72.0	10.93	10.79	90

density  $J_{\rm ph}$  ( $J_{\rm ph} = J_{\rm L} - J_{\rm D}$ ) as a function of the effective voltage  $V_{\rm eff}$ ( $V_{\rm eff} = V_0 - V_a$ ) of the devices.  $J_{\rm L}$  is the current density under illumination,  $J_{\rm D}$  is the current density in the dark,  $V_0$  is the voltage at which  $J_{\rm ph} = 0$  and  $V_a$  is the applied voltage in the dark. As shown in Fig. 4a and b, the saturation photocurrent ( $J_{\rm sat}$ ) of the **IDTT-BH** devices were reached earlier than that of **IDTT-OBH**, suggesting that **IDTT-BH** was the favorable acceptor paired with **J71** and **PDCBT**. While, from Fig. 4c, we found that the result was reverse for the **PBDB-T** based devices and **IDTT-OBH** was the favorable acceptor. The charge dissociation and charge collection probability (P(E, T)) can be calculated using the value of  $J_{\rm ph}/J_{\rm sat}$ . Under short-circuit and maximal power output conditions, the devices based on **J71:IDTT-BH** and **PDCBT:IDTT-BH** exhibited the P(E, T) values of 93.3%/81.8% and 97.2%/84.2%, respectively, which were all higher than those for the **IDTT-OBH**. However, the P(E, T) values of **PBDB**-**T:IDTT-OBH** based devices (95.1%/80.3%) were higher than those of **PBDB-T:IDTT-BH**. These results suggested that the optimal devices have a more efficient exciton dissociation and charge collection process. And the side-chains are thought to be the crucial factor for these data.

#### Morphology characterization

Atomic force microscopy (AFM) and transmission electron microscopy (TEM) were used to measure the morphologies of the optimal blend films as shown in Fig. 5. A significant difference of the morphologies for those blend films could be clearly observed. When **J71** and **PDCBT** were used as donor materials, the blend films with **IDTT-OBH** showed poor surface morphologies with large phase separations and room-mean-



Fig. 4 (a)–(c)  $J_{ph}$  versus  $V_{eff}$  of the corresponding devices; photoluminescence spectra of neat acceptors (excitation at 733 nm) and donors:acceptors (excitation at 733 nm) in films: (d) J71:IDTT-BH/IDTT-OBH, (e) PDCBT:IDTT-BH/IDTT-OBH, and (f) PBDB-T:IDTT-BH/IDTT-OBH.



Fig. 5 AFM height images (3  $\mu m \times$  3  $\mu m)$  of donor:acceptor blended films.

square (RMS) roughness values of 4.36 and 3.60 nm, respectively. In contrast, blending films using **IDTT-BH** as the acceptor displayed much smoother surface morphologies with RMS values of 1.33 and 1.84 nm, respectively. When we chose **PBDB-T** as the donor material, the blend film of **IDTT-OBH** showed a suitable morphology with a RMS roughness of 2.25 nm, which is much better than those of **IDTT-BH**. As shown in the TEM images (Fig. 6), some nanofibril-like networks with good phase separation were formed in the **J71:IDTT-BH** and **PDCBT:IDTT-BH** blend films. While the **J71:IDTT-OBH** and **PDCBT:IDTT-**



Fig. 6 TEM images of donor:acceptor blended films. The scale bars are 100 nm.

**OBH** blend films show clearly larger aggregations compared with those of IDTT-BH based films. When PBDB-T was chosen as the donor material, the IDTT-OBH blend film showed better phase separation with some fibrillar structures. The distinct difference in morphologies can easily explain the effect caused by the side-chains of NFAs. Indeed, a blend film morphology with a better phase separation is favorable to charge transportation, and thus leads to a higher device performance, which is consistent with the results of photoluminescence (PL) quenching experiments at an excitation wavelength of 733 nm (Fig. 4d-f). The blend films with the better phase separation exhibited higher PL quenching efficiency, illustrating that more efficient photoinduced charge transfer occurred in the active layer. The above results demonstrate that the side-chains of NFAs could distinctly control the morphologies of the active layer. Different donors should be paired to the acceptor with most favorable side-chains to obtain a suitable morphology of blend films and then to achieve the best photovoltaic performance.

Two-dimensional grazing-incidence X-ray diffraction (2D-GIXD) was employed to investigate the influence of sidechains on the molecular packing and packing orientation of the active layer. As shown in Fig. S11,† the J71:IDTT-BH blend film showed higher (h00) diffraction and obvious (010) diffraction which indicated that the J71:IDTT-BH blend film had ordered molecular packing, which led to the better device performance based on J71:IDTT-BH. The blend films of PDCBT:IDTT-BH exhibited a more obvious (010) diffraction peak than PDCBT:IDTT-OBH, suggesting that the IDTT-BH system had better  $\pi$ - $\pi$  stacking diffraction and a preferred faceon orientation. When these two acceptors blended with PBDB-T, the IDTT-OBH system displayed better  $\pi$ - $\pi$  stacking diffraction than that of IDTT-BH. The favorable molecular packing should be beneficial for charge transport, which is consistent with the results of good FF values of the corresponding devices.

The change transport properties of the optimized devices were studied by the space-charge-limited current (SCLC) method with an electron and hole only device. The hole mobility  $(\mu_h)$  of the blend films were investigated with the device structure of ITO/PEDOT/donors:acceptors/Au and the electron mobility  $(\mu_e)$  was measured with the device structure of ITO/ ZnO/donors:acceptors/Al. As the data shows in Table 3, the devices based on J71:IDTT-BH, PDCBT:IDTT-BH and PBDB-T:IDTT-OBH showed higher and more balanced hole and

Blend fili	ns			
Donor	Acceptor	$\mu_{\rm e}  ({\rm cm}^2  {\rm V}^{-1}  {\rm s}^{-1})$	$\mu_{\rm h} \left( {\rm cm}^2 \ {\rm V}^{-1} \ {\rm s}^{-1} \right)$	$\mu_{\rm h}/\mu_{\rm e}$
J71	IDTT-BH	$1.63 imes10^{-4}$	$2.02 imes10^{-4}$	1.24
	IDTT-OBH	$4.09\times 10^{-5}$	$1.52\times 10^{-4}$	3.72
PDCBT	IDTT-BH	$1.33 imes 10^{-4}$	$1.82 \times 10^{-4}$	1.37
	IDTT-OBH	$8.18\times 10^{-5}$	$1.72\times 10^{-4}$	2.10
PBDB-T	IDTT-BH	$8.88\times10^{-5}$	$1.88\times 10^{-4}$	2.12
	IDTT-OBH	$9.78\times10^{-5}$	$1.85\times10^{-4}$	1.89

electron mobilities, which are consistent with the morphology analysis. All these contribute to the higher FF and  $J_{sc}$  for the corresponding devices.

## Conclusions

In conclusion, we designed and synthesized two new NFAs, IDTT-BH and IDTT-OBH. These two acceptors possess the same core and end-groups, but have different side-chains of 4-alkylphenyl in IDTT-BH and 4-alkylphenol in IDTT-OBH. They exhibit similar photophysical properties and energy levels. Three polymer donors J71, PDCBT and PBDB-T were blended with these two NFAs to investigate the effect of side-chains on the morphology of the active layer and device performance. For the devices based on J71 and PDCBT as donor materials, IDTT-BH exhibited the PCEs of 11.05% and 10.35%, respectively, which were higher than those of IDTT-OBH based devices. While, the devices based on PBDB-T:IDTT-OBH showed a better PCE of 10.93% than those of IDTT-BH based devices. It has been found that the side-chains have an obvious effect on the morphology of blend films and the corresponding photovoltaic performance. These results demonstrate that the side-chains should be carefully considered not only in the design of NFAs but also in the choice of donors for device fabrications. The work provides a valuable insight for acceptor materials design and device optimization to further boost the PCEs of OSCs.

## Conflicts of interest

There are no conflicts of interest to declare.

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