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Side-chain modification of non-fullerene acceptors for organic solar cells with efficiency over 18%†

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Three non-fullerene acceptors BTP-OC4, BTP-OC6 and BTP-OC8 with different 4-alkyloxyphenyl side-chains were designed and synthesized through a new synthetic route without organotin reagent. The length of the alkyloxy attached to the benzene ring has no influence on the energy levels and absorption of these small-molecular acceptors. However, the active layer morphology is modulated by the different crystallinity caused by the different chain's length. Among them, the device based on the PM6:BTP-OC6 system exhibited the lowest energy loss of 0.513 eV and the highest power conversion efficiency (PCE) of 17.59%, as well as excellent photo-soaking/thermal/storage stability. Moreover, when Y6 was introduced into PM6:BTP-OC6 to fabricate a ternary device, the PCE was promoted to over 18%. Our results provide a feasible strategy to develop high-performance acceptors through fine tuning the aromatic side chain, which can be easily realized by our reported synthetic route.

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1. Introduction

Organic solar cells (OSCs) have received great attention as forming one of the most promising classes of renewable energy platforms owing to their advantages of low cost, light weight, flexibility and roll-to-roll manufacturing capability.^{1–10} In the past few years, impressive power conversion efficiencies (PCEs) over 19% of OSCs have been reported.^{10–14} These remarkable results are mainly attributed to the design of novel active layer materials, together with the improved understanding of the

mechanism and device optimization.^{15–24} Due to the acceptor-donor-acceptor (A–D–A) architecture, non-fullerene acceptors (NFAs) have the desirable characteristics of easy molecular tailoring, tunability of energy levels, and strong absorption in the visible and near-infrared (NIR) regions.^{25–27} Although many breakthroughs have been achieved for OSCs, there still remains much room for improving device performance according to the theoretical predictions.^{28–32}

The current mainstream of NFAs can be roughly thought of as consisting of backbones, side chains and end groups, which can be finely-tuned to adjust the properties of the NFAs. In 2019, the representative molecule Y6, which employs an electron-deficient-core-based central fused ring with a benzo-thiadiazole core, was reported by Zou and coworkers.²⁵ Afterwards, many modifications have been carried out based on the Y6 skeleton to regulate the energy levels and enhance molecular planarity as well as intramolecular charge transfer. Among them, the changes of the side chains have a great impact on the photo-voltaic performance of solar cells.^{33,34} For example, by gradually shortening the side chain length, the Hou group synthesized BTP-eC11, BTP-eC9 and BTP-eC7. Among them, BTP-eC9 maintains good solubility as well as enhanced intermolecular ordering, resulting in an enhanced PCE of 17.8%.³⁵ An acceptor material named L8-BO with a 2-butyloctyl side chain, which was reported by Sun *et al.*, achieved a PCE of 18.2% and a high fill factor (FF) over 81%.³⁶ Yan *et al.* changed the alkyl side chains to aromatic chains, and realized a significant increase

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in PCE.^{1,37} Those studies revealed the fact that fine-tuning of the side chains could influence the absorption spectrum, molecular configuration and intermolecular packing of NFAs effectively. Meanwhile, through comparing two acceptors with different side-chains of 4-alkyl-phenyl in IDTT-BH and 4-alkylphenol in IDTT-OBH in our previous work, it was found that the introduction of an oxygen atom in ITIC derivatives can significantly change the film morphology property in OSCs.³⁸ However, the influence of the length of the aromatic chains and the commercially accessible benzene epoxy chain are seldom reported for Y6 derivatives before.

In this work, we changed the traditional synthetic route, and picked 4-alkoxy-phenylboronic acid as a raw material of Suzuki-coupling, thus introducing an oxygen atom to the benzene side chain on the backbone of Y6. Three acceptors, named BTP-OC4, BTP-OC6 and BTP-OC8, with different alkoxy lengths (butoxy, hexyloxy and octyloxy) were successfully reported. The acceptors share similar UV-vis absorption and energy levels, while the device based on BTP-OC6 shows the best PCE of 17.59% along with the best device stability. The best PCE based on BTP-OC6 was attributed to the fine-tuned crystalline feature and best device performance of charge separation and recombination. When Y6 was chosen as the third component, the PCE was further improved to over 18%. This work provided a route for the synthesis of Y6 derivatives with versatile aromatic side-chains from cheap raw materials, and highlighted the impact of oxyalkyl chain length on benzene rings in Y6 analogues and OSC performance.

2. Results and discussion

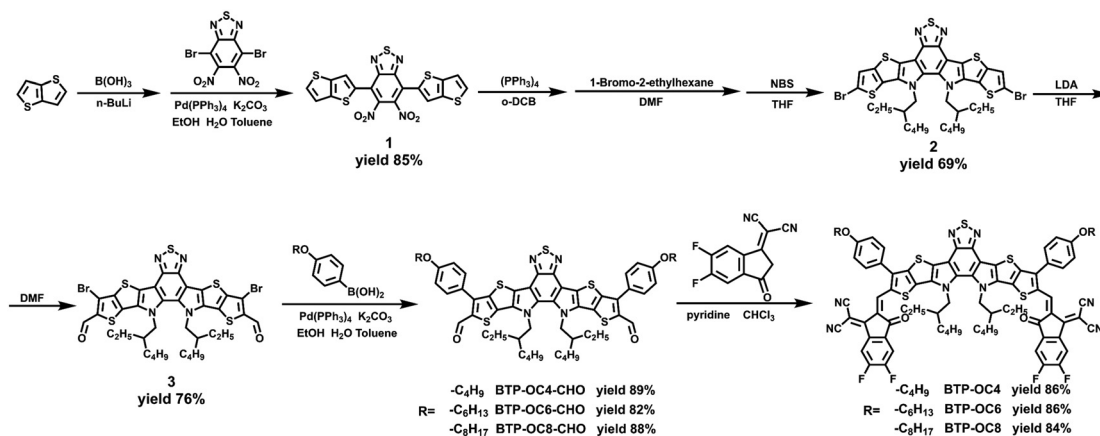
2.1 Synthesis and optoelectronic properties

The synthetic routes of BTP-OC4, BTP-OC6 and BTP-OC8 are shown in Scheme 1. Starting from thieno[3,2-*b*]thiophene, compound **1** was obtained through similar synthetic routes of Y6, and then compound **3** was successfully synthesized by brominated reaction and bromine migration reaction.³⁹ Finally, the targeted products BTP-OC4, BTP-OC6 and BTP-OC8 were

obtained. It should be noted that organotin reagents were not involved in the synthetic routes, which is important for the large-scale production of organic photovoltaic materials. Through this method, many kinds of aromatic side chains can be introduced easily, and thus the fine adjusting of energy levels and molecular stacking can be easily realized by changing the raw material of the penultimate step, without going through complex and repetitive reaction routes. The detailed synthesis procedures and characterizations including ¹H NMR and ¹³C NMR are provided in the ESI.†

The normalized UV-vis absorption spectra of the BTP-OC4, BTP-OC6 and BTP-OC8 were investigated and compared in their chloroform (CF) solutions and neat films (Fig. 1b and c and Fig. S2, ESI†), and the detailed data are summarized in Table 1. As shown in Fig. 1b, the absorption coefficients of three acceptors in chloroform solutions are calculated to be around 2.24, 2.22 and 2.26 × 10⁵ M^{−1} cm^{−1}, respectively. The negligible difference in their absorption coefficients could be attributed to their same conjugated backbones. In neat films, BTP-OC4, BTP-OC6 and BTP-OC8 have rather similar absorption profiles with maximum absorption peaks around 820 nm, consistent with the trend of their solution absorptions. The optical bandgaps of BTP-OC4, BTP-OC6 and BTP-OC8 calculated from the onset of the neat films are around 1.38 eV.

Cyclic voltammetry (C-V) measurements were performed to evaluate the energy levels of the three acceptors. From the onset of the oxidative and reductive peaks of the C-V curves (Fig. S3, ESI†), the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy levels of BTP-OC4, BTP-OC6 and BTP-OC8 are calculated and summarized in Table 1. The HOMO/LUMO values of BTP-OC4, BTP-OC6 and BTP-OC8 are −5.53/−4.02 eV, −5.51/−3.99 eV and −5.54/−3.97 eV, respectively. Clearly, those acceptors show similar UV-vis absorption patterns and HOMO/LUMO values, indicating that the length of the alkyl chains on the benzene has little influence on their optoelectronic properties in neat films. As the energy level alignment shows in Fig. 1d, all acceptors are well-matched with the donor PM6. Though the HOMO energy level offsets between PM6 and the two acceptors are



Scheme 1 Synthetic route of acceptors.

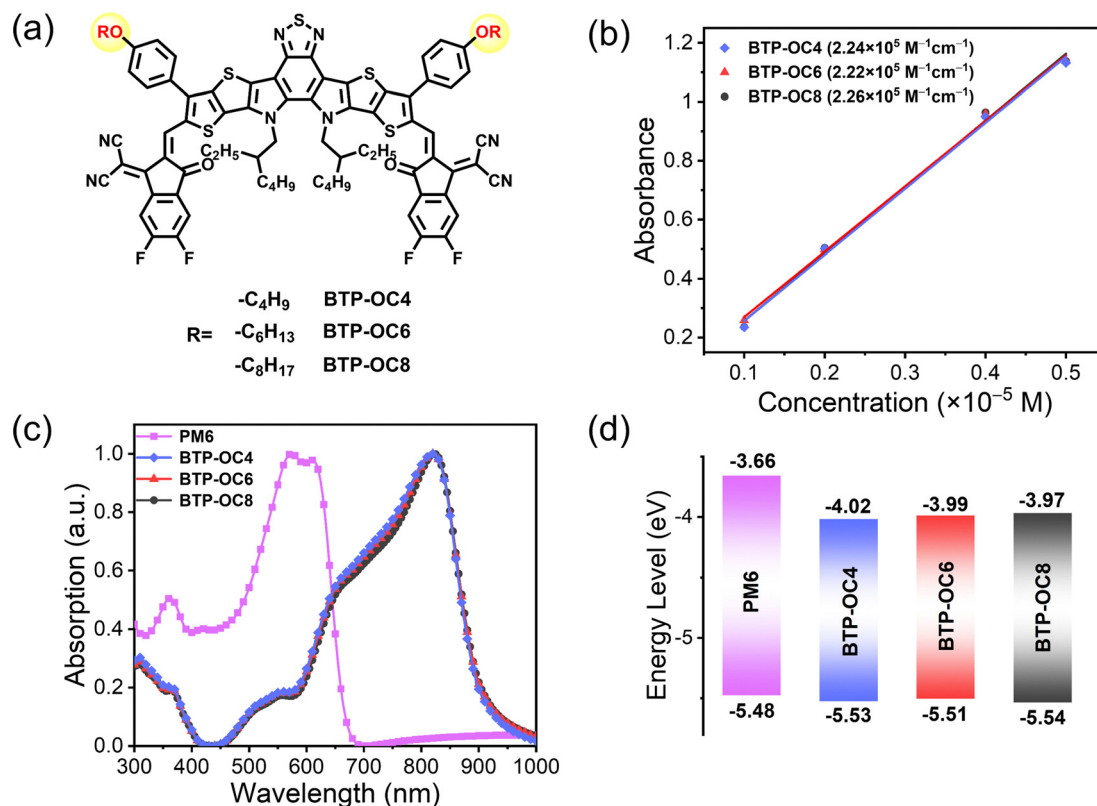


Fig. 1 (a) Chemical structures of BTP-OC4, BTP-OC6 and BTP-OC8, (b) absorption coefficients of the NFA materials in chloroform solutions, (c) absorption spectra on neat films and (d) energy level diagrams of donors and acceptors.

Table 1 Photoelectric properties of BTP-OC4, BTP-OC6 and BTP-OC8 in neat films

Compound	$\lambda_{\text{max}}^{\text{CF}}$ (nm)	$\lambda_{\text{max}}^{\text{film}}$ (nm)	$E_{\text{g}}^{\text{opt}}$ (eV)	E_{HOMO} (eV)	E_{LUMO} (eV)
BTP-OC4	728	820	1.39	-5.53	-4.02
BTP-OC6	728	820	1.38	-5.51	-3.99
BTP-OC8	728	822	1.38	-5.54	-3.97

around 0.05 eV, this is still large enough for the exciton separation in the nonfullerene systems.⁴⁰

2.2 Photovoltaic performance

All devices were fabricated to evaluate the photovoltaic properties using BTP-OC4, BTP-OC6 and BTP-OC8 as acceptors, PM6 as the donor, and PDINO was used as the interface layer.⁴¹ And the conventional structure of ITO/PEDOT:PSS/active layer/PDINO/Ag was adopted. Afterwards, the device optimization details, such as donor/acceptor weight ratio and thermal annealing temperature are summarized in the ESI† (Tables S1–S3). The current density–voltage (J – V) curves of the optimized devices are illustrated in Fig. 2b, and the related photovoltaic data are summarized in Table 2. The optimized PM6:BTP-OC4 based cell exhibited a PCE of 16.45%, with a V_{oc} of 0.885 V, a J_{sc} of 24.86 mA cm $^{-2}$, and an FF of 74.76%. With a longer alkyl chain of hexoxy, the PCE of the PM6:BTP-OC6 based cell was greatly improved to 17.59% with similar V_{oc} but obviously improved FF of 77.14% and J_{sc} of 25.66 mA cm $^{-2}$.

When the length of the alkyl chain was further improved to octyloxy, the optimized device of PM6:BTP-OC8 showed a decreased PCE of 16.88%, with a V_{oc} of 0.882 V, a J_{sc} of 25.31 mA cm $^{-2}$, and an FF of 75.58%. Among the three acceptors, devices based on BTP-OC6 demonstrated slightly higher V_{oc} , together with the highest J_{sc} and FF simultaneously, which might be related to its excellent film morphology and compact molecular stacking. Besides, the E_{loss} of the three OSC devices was calculated based on the reported methods (Table S4, ESI†),⁴² and the E_{gap} was calculated from the cross point between the normalized UV-vis absorption spectra and the normalized photoluminescence (PL) spectra of the neat films as shown in Fig. S4 (ESI†). Combined with their V_{oc} , the E_{loss} was calculated to be around 0.51 eV, which was significantly lower than reported NFAs.⁴²

The external quantum efficiency (EQE) spectra of the devices are shown in Fig. 2d. All three devices yielded high EQE values in their absorption ranges. The PM6:BTP-OC6 based cell achieved the highest EQE value of 88% at 530 nm, which is higher than the other two devices (85% at 530 nm). The integrated J_{sc} values of the devices determined from the EQE curves are 24.22, 25.08 and 24.68 mA cm $^{-2}$, respectively, which are in good agreement with the J_{sc} values measured in the J – V curves.

Due to the excellent performance of the BTP-OC6 based device, we further fabricated the corresponding ternary device using Y6 as the third component, which achieved a PCE of

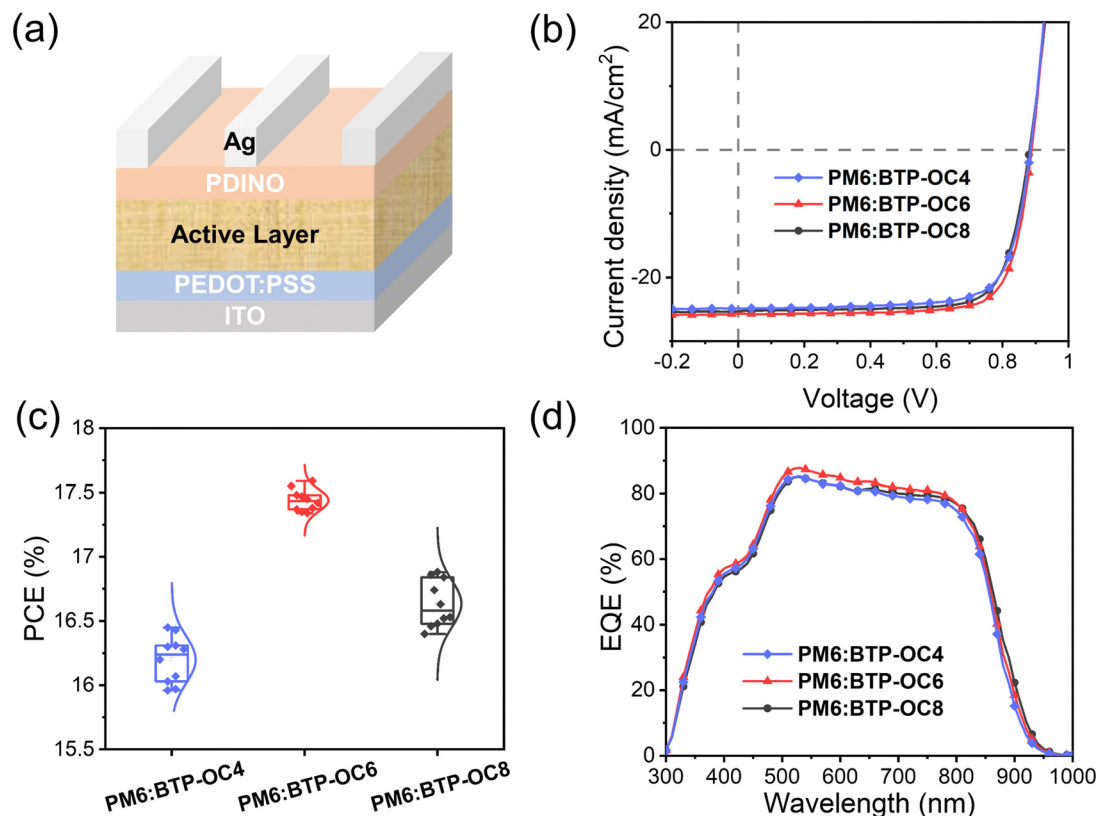


Fig. 2 (a) Device structure, (b) J - V curves, (c) PCE distribution image and (d) EQE curves of BTP-OC4, BTP-OC6 and BTP-OC8 based devices.

Table 2 Photovoltaic performance of acceptor-based devices

Active layer	V_{oc} (V)	J_{sc} (mA cm ⁻²)	J_{sc}^{EQE} (mA cm ⁻²)	FF (%)	PCE ^a (%)
PM6:BTP-OC4	0.885 (0.885 ± 0.002)	24.86 (24.53 ± 0.31)	24.22	74.76 (74.63 ± 0.54)	16.45 (16.20 ± 0.20)
PM6:BTP-OC6	0.889 (0.884 ± 0.004)	25.66 (25.56 ± 0.13)	25.08	77.14 (77.16 ± 0.39)	17.59 (17.44 ± 0.08)
PM6:BTP-OC8	0.882 (0.879 ± 0.002)	25.31 (25.07 ± 0.18)	24.68	75.58 (75.50 ± 0.47)	16.88 (16.63 ± 0.17)
PM6:BTP-OC6:Y6	0.866 (0.866 ± 0.002)	27.00 (26.66 ± 0.22)	26.01	77.24 (76.97 ± 0.55)	18.0 (17.77 ± 0.18)

^a The average parameters included in the brackets were calculated from 10 devices.

18.06%, with an integrated J_{sc} value of 26.01 mA cm⁻². The related J - V curves and EQE spectra are shown in Fig. S5 (ESI†).

2.3 Charge transport, exciton dissociation and charge generation

To understand the different device performance in FF and J_{sc} , the dependence of photocurrent density (J_{ph}) on effective voltage (V_{eff}) was measured to study exciton dissociation and charge collection properties, as shown in Fig. 3a. Herein, $J_{ph} = J_L - J_D$, where J_L and J_D are photocurrent densities under illumination and dark conditions, respectively. $V_{eff} = V_0 - V_{bias}$, where V_0 is the voltage at which J_{ph} is zero and V_{bias} is the applied external bias voltage. The value of η ($\eta = J_{ph}/J_{sat}$, where J_{sat} is the saturation photocurrent density) under the short-circuit and the maximum power output conditions represents charge dissociation (η_{diss}) and collection (η_{coll}) probability, respectively. When the V_{eff} is over 2 V, the J_{ph} of all the devices

approached saturation (J_{sat}), signifying complete exciton dissociation and charge collection. The η_{diss} values for BTP-OC4, BTP-OC6 and BTP-OC8 based OSCs were calculated to be 96.39%, 97.33% and 97.24%, respectively, which means that devices based on all three acceptors possess efficient exciton dissociation, especially for BTP-OC6. Meanwhile, the PM6:BTP-OC6 device possessed the highest charge collection efficiency as supported by the η_{coll} over 88%.

To study the charge recombination in the devices, the light-intensity dependence (P_{light}) of V_{oc} or J_{sc} was measured. The semi-logarithm plots of P_{light} -dependent V_{oc} for the three devices are illustrated in Fig. 3b. The fitted slopes of the BTP-OC4, BTP-OC6 and BTP-OC8 based devices are 1.22 kT/q, 1.15 kT/q and 1.19 kT/q, respectively, in which a slope (n) close to unity means low degree of trap-assisted charge recombination in the OSCs. In addition, the dependence of J_{sc} on P_{light} follows the power-law equation: $J_{sc} \propto P^\alpha$, where α reflects the degree of

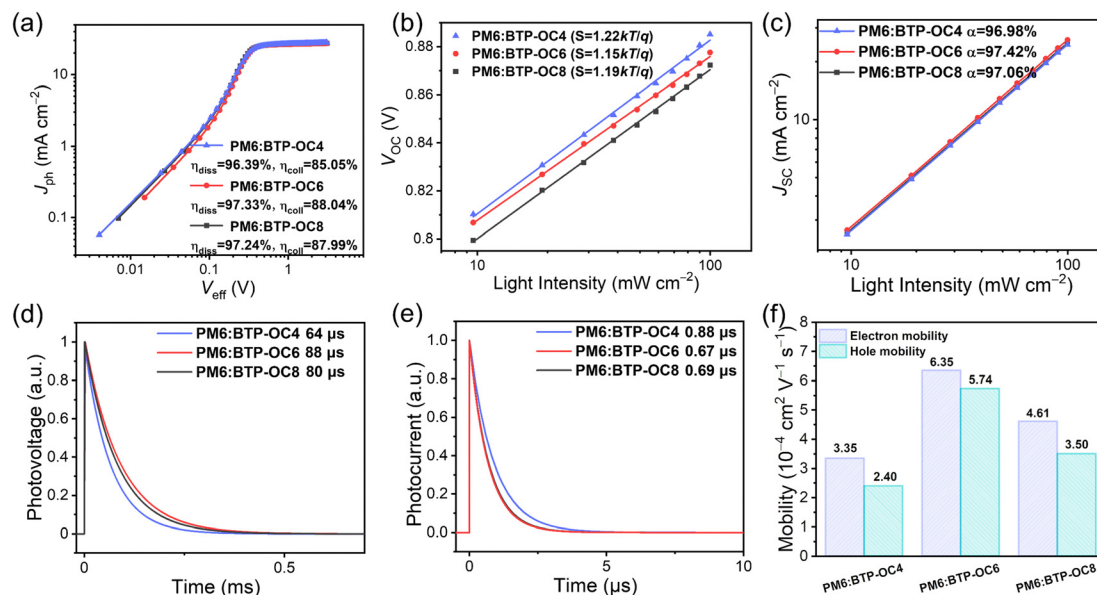


Fig. 3 (a) J_{ph} versus V_{eff} , (b) V_{OC} and (c) J_{sc} versus light intensity, (d) transient photovoltage (TPV), (e) transient photocurrent (TPC) and (f) hole mobility and electron mobility of BTP-OC4, BTP-OC6 and BTP-OC8 based devices.

bimolecular recombination. As shown in Fig. 3c, the α values of the devices are found to be 96.98%, 97.42%, and 97.06%, respectively. The α values close to unity indicate low extent of bimolecular recombination and efficient charge collection of the three devices, among which the PM6/BTP-OC6 based device suffers from the least extent of trap-assisted recombination and bimolecular recombination compared with the other two devices, resulting in higher FF and J_{sc} .

Fig. 3d and e display the transient photovoltage (TPV) and transient photocurrent (TPC) decay traces of the BTP-OC4, BTP-OC6 and BTP-OC8 based devices. The photogenerated carrier lifetimes derived from TPV measurements are 64, 88 and 80 μ s for BTP-OC4, BTP-OC6 and BTP-OC8 based OSCs, respectively, indicating that charge carrier recombination has been suppressed in PM6:BTP-OC6 based OSCs. For TPC measurements, PM6:BTP-OC6 based OSCs displayed a charge extraction time of 0.67 μ s, shorter than that of BTP-OC4 and BTP-OC8 based OSCs (0.88 and 0.69 μ s, respectively). The fastest charge sweep-out process of BTP-OC6 based OSCs indicates the most efficient charge transport. Furthermore, as shown in Fig. S6 and Table S5 (ESI[†]), the hole mobility and electron mobility of the PM6:BTP-OC6 blend were calculated to be 5.74×10^{-4} and 6.35×10^{-4} cm² V⁻¹ s⁻¹, respectively, both of which are higher than those of the PM6:BTP-OC4 blend and PM6:BTP-OC8 blend, as shown in Fig. 3f. The higher and more balanced hole and electron mobility for the PM6:BTP-OC6 blend should account for its faster charge sweep-out process, consistent with the TPC results.

Based on the above results, the BTP-OC6 based device achieved the most effective charge dynamic behaviors from the perspective of exciton dissociation, charge recombination and transportation, thus leading to its best PCE.

2.4 Morphology analysis

Atomic force microscopy (AFM) measurements were carried out to investigate the microstructure morphologies of the blend films. As shown in Fig. 4 and Fig. S7 (ESI[†]), an obvious uniform fibrillar structure can be observed for all the blend films from their AFM images, which suggests interpenetrating networks in the active layers. From the AFM height images, the root-mean-square surface (RMS) roughness was calculated to be 1.13 nm, 0.955 nm, and 0.886 nm, respectively. The gradually decreased RMS values of the three blend films imply that the longer sidechain brought out weaker molecular aggregation.

Grazing incidence wide-angle X-ray diffraction (GIWAXS) was employed to further investigate the crystallization properties, as shown in Fig. S8 (ESI[†]). All the neat films showed a lamellar peak at ~ 0.25 Å⁻¹ in the in-plane (IP) direction and π - π stacking peak at ~ 1.69 Å⁻¹ in the out-of-plane (OOP) direction, an indicator of face-on molecular orientation. After being blended with PM6, their face-on orientation can be well maintained. As observed in Table S6 (ESI[†]), the π - π stacking distance in the BTP-OC4 and BTP-OC6 blend films is quite similar (3.722 Å), while it slightly became larger for the BTP-OC8 (3.751 Å) featuring the longest alkyloxy side chain, which is in agreement with the conclusion acquired from AFM images. In addition, the PM6:BTP-OC6 blend film possessed the largest crystal correlation length (CCL) of 25.32 Å. The smaller π - π stacking distance along with the largest CCL in the PM6:BTP-OC6 blend film is an advantage for the charge transportation, which can be matched up with the high FF and J_{sc} of PM6:BTP-OC6 based OSCs.

2.5 Device stability

The device stability is one of the most challenging issues for OSCs, especially in conventional-structure devices. As shown in

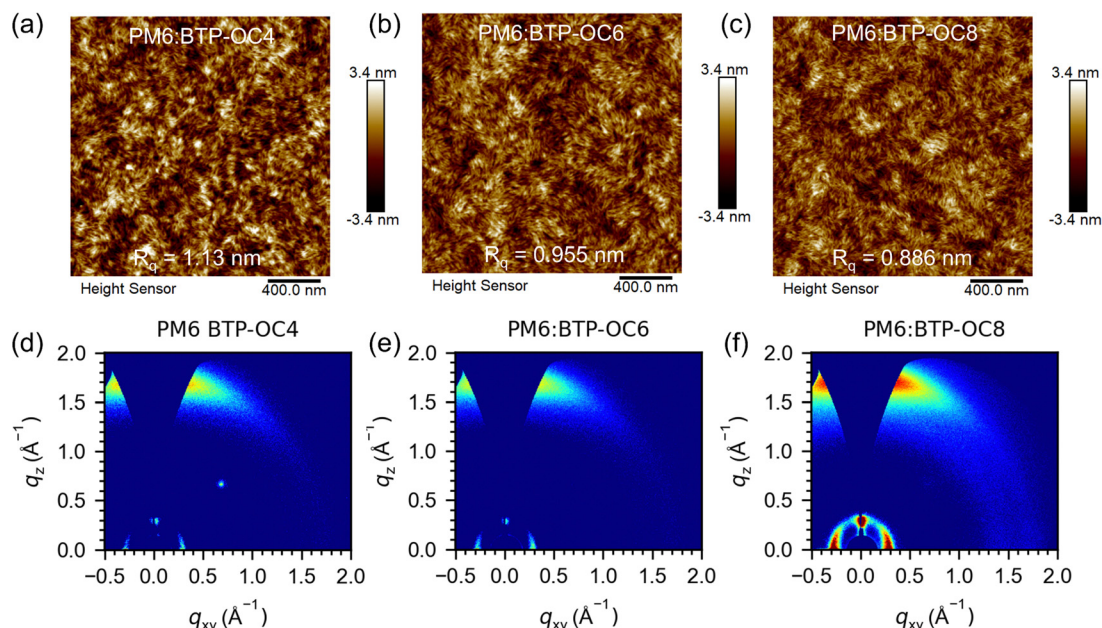


Fig. 4 (a–c) The AFM height images and (d–f) 2D GIWAXS images for the blend films based on PM6:BTP-OC4, PM6:BTP-OC6 and PM6:BTP-OC8.

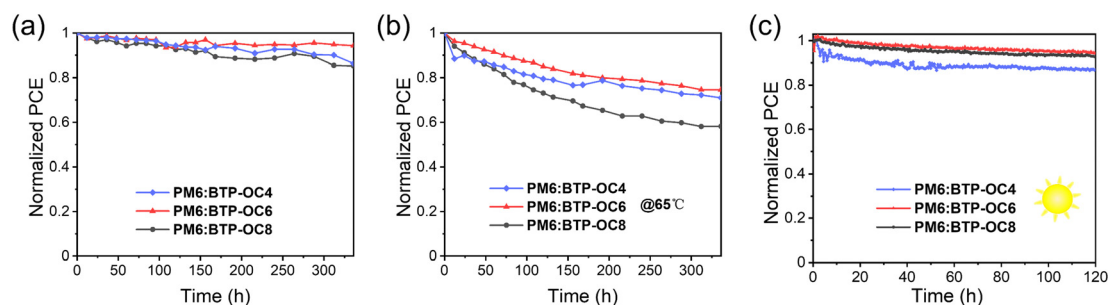


Fig. 5 (a) Normalized PCE of storage stability in a nitrogen-filled glovebox, (b) thermal stability at 65 °C, and (c) light-soaking stability under MPP tracking of BTP-OC4, BTP-OC6 and BTP-OC8 based devices.

Fig. 5 and Table S7 (ESI[†]), it was found that devices based on BTP-OC4, BTP-OC6 and BTP-OC8 all exhibited decent storage stability when kept in a glovebox filled with nitrogen, in which the BTP-OC6 based device maintained over 94% of its initial PCE after 350 h. Besides, thermal stability and light-soaking stability under max power point (MPP) tracking of the OSCs were tested. Eventually, 74.6% PCE was maintained after continuously heating at 65 °C for 350 h by BTP-OC6 based devices. And the light-soaking stability of the PM6:BTP-OC6 system is evidently the highest among them. In conclusion, devices based on BTP-OC6 not only get the best photovoltaic performance but also possess the best stability performance.

Generally, the lifetime of the device is controlled by the photoactive layer, which might be affected by several factors, such as oxygen and water, irradiation, mechanical stress, and so on.^{43–45} Here, the different kinetic stability performance with the related device would be owing to the small molecular acceptor (SMA) diffusions and crystallization. That is, the higher diffusion coefficients lead to higher nucleation density of crystals and faster crystal growth and lower stability.⁴⁶ A high

glass transition temperature (T_g) that prevents molecular diffusion is conducive to kinetic stabilization of the morphology.⁴⁷ As depicted in Fig. S9 (ESI[†]), the T_g values of thin films measured by UV-vis spectroscopy⁴⁸ are 101 °C, 102 °C and 81 °C for BTP-OC4, BTP-OC6 and BTP-OC8, respectively. The higher T_g of BTP-OC6 partially contributes to its better storage and thermal stability. To explain the difference of the light-soaking stability, we tested the stability of the SMAs under continuous UV illumination (365 nm, 40 mW cm⁻²). As shown in Fig. S10 (ESI[†]), the absorption intensity of the BTP-OC4 film decreased sharply as time went on, while the BTP-OC6 exhibited an excellent photostability, which is in agreement with the tendency of the corresponding device under MPP tracking.

3. Conclusion

In this work, three acceptors (BTP-OC4, BTP-OC6 and BTP-OC8) with different lengths of side chain on the benzene ring were designed and synthesized with cheap raw materials and

without organotin reagent. The length of the alkyloxy on the aromatic phenyl side chain has little impact on their absorption, energy levels and molecular packings. Meanwhile, after being blended with PM6, the blend film of PM6:BTP-OC6 achieved better morphological characteristics with the largest CCL. As a result, the OSC based on BTP-OC6 shows the best PCE of 17.59% along with the best device stability, attributed to the fine-tuned crystalline feature, more efficient exciton separation process and less charge recombination properties. Furthermore, the PCE was improved over 18% when Y6 was chosen as the third component, suggesting its potential in fabricating high-performance OSCs. We believe that our synthetic method provides an option for developing more high-performance acceptors with different aromatic side-chains.

Conflicts of interest

The authors declare no conflict of interest.

Acknowledgements

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