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# Achieving Organic Solar Cells with an Efficiency of 18.80% by Reducing Nonradiative Energy Loss and Tuning Active Layer Morphology

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The efficiency of organic solar cells (OSCs) is primarily limited by their significant nonradiative energy loss and unfavorable active layer morphology. Achieving high-efficiency OSCs by suppressing nonradiative energy loss and tuning the active layer morphology remains a challenging task. In this study, an acceptor named CH-ThCl is designed, featuring an extended conjugation central core, dichlorodithienoquinoxaline. The incorporation of chlorine-substituted extended conjugation in the central core enhances the acceptor's rigidity and promotes J-aggregation, leading to improved molecular luminescent efficiency and a reduction in nonradiative energy loss. A binary device based on PM6: CH-ThCl demonstrates a power conversion efficiency (PCE) of 18.16% and exhibits a high open-circuit voltage ( $V_{oc}$ ) of 0.934 V, attributed to the remarkably low nonradiative energy loss of 0.21 eV. Furthermore, a ternary device is fabricated by incorporating CH-6F as the third component, resulting in a significantly enhanced PCE of 18.80%. The ternary device exhibits improvements in short-circuit current (J<sub>sc</sub>) and fill factor (FF) while maintaining the  $V_{oc}$ , primarily due to the optimized active layer morphology. These results highlight the effectiveness of combining the reduction of nonradiative energy loss and precise tuning of the active layer morphology as a viable strategy for achieving high-efficiency OSCs.

# 1. Introduction

Organic solar cells (OSCs) have garnered significant research interest and demonstrated potential applications in building-integrated photovoltaics, greenhouses, wearable devices, and more, owing to their advantages of flexibility, light weight, and large-area solution processing.<sup>[1-4]</sup> In recent years, the power

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conversion efficiencies (PCEs) of OSCs have substantially increased primarily due to innovations in active layer materials, particularly the A-D-A type of non-fullerene acceptors such as ITIC, Y6, and their derivatives.[5-8] However, the efficiencies of OSCs are limited by their substantial energy loss, especially the nonradiative energy loss.<sup>[9-14]</sup> Recent results indicate that nonradiative energy loss can be reduced by improving the luminescent efficiency of the low bandgap component in the active layers.<sup>[15–18]</sup> In typical OSCs with a bulk heterojunction structure, the active layer materials consist of a wide bandgap donor and a low bandgap acceptor. Therefore, it is crucial and necessary to design low bandgap acceptor materials with highly luminescent properties to minimize nonradiative energy loss and achieve high open voltage  $(V_{\alpha c})$ . Additionally, active layer morphologies play a crucial role in enhancing OSC performance, particularly the photovoltaic parameters of short-circuit current  $(I_{sc})$  and fill factor (FF).<sup>[19-23]</sup> Over the past decade, various strategies, such as thermal or solvent

annealing, additives, and ternary approaches, have been employed to finely tune active layer morphologies.<sup>[24-30]</sup> The ternary strategy, in particular, has been extensively investigated and has achieved significant success in morphology control and energy loss suppression.<sup>[31-36]</sup> Clearly, active layer materials play a fundamental role in energy loss suppression and morphology control. Considerable research efforts have been dedicated to acceptor material design in recent years.<sup>[2,22]</sup> The Y-series acceptors, characterized by banana structures and dense packing modes, have substantially boosted OSC efficiencies to impressive levels.<sup>[8,27,37-42]</sup> However, many devices based on Y6 derivatives still suffer from relatively high energy loss and trade-offs among the photovoltaic parameters of  $V_{\rm oc}$ ,  $J_{\rm sc}$ , and FF. Therefore, maximizing the PCE of OSCs by reducing energy loss and controlling morphology poses an essential and challenging task from a material design perspective.[43,44]

In our recent work, we have designed a series of CH acceptors with extended conjugation central cores, which form strong intermolecular interactions and exhibit low energy loss in corresponding photovoltaic devices. These CH acceptor molecules

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**Figure 1.** a) Chemical structure of CH-ThCl. b) The absorption spectra and c) energy level diagram of PM6 and CH-ThCl. d) The single crystal structure of CH-ThCl from a top view and side view. e) The molecular packing patterns and f-i) the three types of intermolecular packing modes in the single crystal of CH-ThCl.

have achieved PCEs exceeding 18%.[45-47] Building upon these previous achievements, we herein reported a new acceptor called CH-ThCl, featuring an even more extended conjugation central core, dichlorodithienoquinoxaline. The further extension of the conjugation central core with chlorine substitution enhances the acceptor's rigidity and promotes J-aggregation, thereby enhancing the molecular luminescent efficiency and suppressing nonradiative energy loss.<sup>[9,15]</sup> The binary device based on PM6: CH-ThCl demonstrated a PCE of 18.16%, with a high  $V_{oc}$  of 0.934 V,  $J_{\rm sc}$  of 25.43 mA cm<sup>-2</sup>, and an FF of 76.45%. Introducing another acceptor, CH-6F, as the third component in a ternary device resulted in a high PCE of 18.80%. The ternary device showed improved  $J_{sc}$  and FF without sacrificing  $V_{oc}$  compared to the binary device, thanks to the improved active layer morphology. These results serve as a successful example of careful molecule design to minimize nonradiative energy losses and achieve morphology control using the ternary strategy.

#### 2. Results and Discussion

#### 2.1. Synthesis and Characterization

The synthesis of CH-ThCl followed a similar method as we have previously reported,<sup>[46]</sup> and detailed procedures and characterization can be found in the Supporting Information (SI).

CH-ThCl exhibits a high solubility of  $\approx$ 148 mg mL<sup>-1</sup> in chloroform (CF). The UV-vis absorption spectra of CH-ThCl are presented in Figure 1b. In CF solution, CH-ThCl exhibits a maximum absorption ( $\lambda_{\rm max})$  peak at 728 nm. The solid film absorption of CH-ThCl is red-shifted by 73 nm, with  $\lambda_{\rm max}$  at 801 nm compared to its solution absorption. This red-shift suggests the presence of strong intermolecular interactions in the solid film. The energy levels of CH-ThCl were estimated using electrochemical cyclic voltammetry (CV). From the CV curves (Figure S1, Supporting Information), the lowest unoccupied molecular orbital (LUMO) and highest occupied molecular orbital (HOMO) levels of CH-ThCl were determined to be -3.78 and -5.67 eV, respectively (Figure 1c). Compared to Y6, the energy levels of CH-ThCl are upshifted due to the weaker electron-withdrawing ability of the central core, which incorporates thiophene units and weakens the intramolecular charge transfer (ICT) effect. These upshifted energy levels of CH-ThCl reduce the energy offset with donor materials, thus favoring the suppression of radiative loss during exciton dissociation.[9,48]

The single crystal of CH-ThCl was grown by diffusing methanol into a chloroform solution of CH-ThCl. As shown in Figure 1d, CH-ThCl exhibits good planarity, with a twist angle of 3.0° between the end group and the molecular backbone. Unlike the Y6 crystal, CH-ThCl demonstrates a one-dimensional arrangement through  $\pi$ - $\pi$  stacking in the crystal. There are three



Figure 2. a) Architecture of the binary and ternary devices. b) J-V and c) EQE curve for the optimized devices.

types of intense  $\pi$ - $\pi$  interactions in the crystal: the end group to end group mode (E/E) and two types of the end group to the central core dichlorodithienoquinoxaline unit mode (E/C-1 and E/C-2). CH-ThCl demonstrates the intense E/E  $\pi$ - $\pi$  stacking (J- aggregation) with a distance of 3.366 Å (Figure 1h), similar to that in the single-crystal of Y6. In addition, owing to the extended conjugation center unit, the E/C-1 and E/C-2 also show strong  $\pi$ - $\pi$  interactions with the distances similar to that of E/E  $\pi$ - $\pi$  stacking.

#### 2.2. Photovoltaic Performance

То evaluate the photovoltaic performance of CH-ThCl, binary OSCs with a conventional structure of ITO/PEDOT:PSS/PM6:CH-ThCl/PNDIT-F3N/Ag (Figure 2a) were fabricated. Various conditions were employed to optimize the devices, and the details are provided in Table S1 (Supporting Information). The optimized binary device exhibited a power conversion efficiency (PCE) of 18.16% with a  $V_{\rm oc}$  of 0.934 V, a  $J_{\rm sc}$  of 25.43 mA cm<sup>-2</sup>, and a FF of 76.45%. To further enhance the device performance, we introduced an acceptor CH-6F (Figure S2, Supporting Information), previously reported by our group, as the third component to fabricate ternary devices. After optimization, the ternary device achieved a high PCE of 18.80% with a  $V_{\rm oc}$  of 0.931 V, a  $J_{\rm sc}$  of 26.20 mA cm<sup>-2</sup>, and an FF of 77.09%. Compared to the binary device, the ternary device maintained a high  $V_{oc}$  while exhibiting improvements in  $J_{sc}$ and FF. The optimized photovoltaic parameters of the binary and ternary devices are listed in Table 1, and the corresponding current density-voltage (J-V) curves are depicted in Figure 2b. The external quantum efficiency (EQE) curves of the optimized binary and ternary devices are shown in Figure 2c. The ternary device demonstrated higher photon responses across nearly the entire light absorption range compared to the binary device. In

particular, a distinct valley can be observed in the EQE curve of the binary device within the wavelength range of 650–800 nm. However, this valley is almost absent in the EQE curve of the ternary device. The diminished valley in the ternary device is primarily attributed to its higher exciton dissociation efficiency and lower charge recombination, as will be discussed below. Furthermore, the ternary film exhibits slightly enhanced absorption within that range (see Figure S3, Supporting Information), which also contributes to the improved EQE. The calculated  $J_{sc}$  values from the EQE curves of the binary and ternary devices were 24.46 and 25.24 mA cm<sup>-2</sup>, respectively, which align well with those measured in the *J*–V curves.

#### 2.3. Energy Loss Analysis

The above binary and ternary devices all showed high  $V_{0c}$  over 0.93 V. The energy loss analysis was carried out to investigate the underlying reasons. First, the overall  $E_{loss}$  of the two devices together with the device PM:CH-6F were calculated from the equation  $E_{\rm loss} = E_{\rm gap} - qV_{\rm oc}$ , where the  $E_{\rm gap}$  values were estimated by the intersections between the absorption and emission spectra of the low bandgap component.<sup>[49]</sup> As listed in **Table 2**, the  $E_{\text{loss}}$ of the binary and ternary devices are 0.519 and 0.508 eV, respectively, which are all lower than that of PM6:Y6 system with a value of  $\approx 0.55$  eV. Then, the detailed  $E_{\rm loss}$  components for the three devices were investigated following the established methods, in which the  $E_{\rm loss}$  can be divided into three parts,  $E_{\rm loss} = \Delta E_1 + \Delta E_2 + \Delta E_3$ .<sup>[50]</sup> The first part is the radiative recombination loss above the bandgap ( $\Delta E_1$ ), which is unavoidable for any type of solar cells according to the Shockley-Queisser (SQ) theory. The second part is the radiative recombination loss below the bandgap ( $\Delta E_2$ ). The third part  $\Delta E_3$ , also called nonradiative recombination energy loss, is the determined loss in OSCs, which can be calculated from the EQE of EL (EQE  $_{\rm EL})$  following the

 Table 1. Photovoltaic parameters of the optimized binary and ternary devices.

Active layer V <sub>oc</sub> [V]		$J_{\rm sc}$ [mA cm <sup>-2</sup> ]	$\int_{sc}^{a)}$ [mA cm <sup>-2</sup> ]	FF [%]	PCE [%]	
PM6:CH-ThCl	0.934	25.43	24.46	76.45	18.16	
PM6:CH-6F	0.872	25.31	24.97	75.99	16.77	
PM6:CH-ThCl:CH-6F	0.931	26.20	25.24	77.09	18.80	

<sup>a)</sup> Current densities calculated from EQE data.

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Table 2. Energy loss analysis of the binary and ternary devices.

Active layer	<i>V</i> <sub>oc</sub> [V]	$E_{g}^{a)}$ [eV]	E <sub>loss</sub> [V]	V <sub>oc,sq</sub> <sup>b)</sup> [V]	$\Delta E_1^{c)}$ [eV]	$V_{\rm oc,rad}$ <sup>d)</sup> [V]	$\Delta E_2  [\mathrm{eV}]$	$\Delta E_3^{e)}$ [cal. V]	$\Delta E_3^{\text{f}}$ [exp. V]
PM6:CH-ThCl	0.936	1.455	0.519	1.190	0.265	1.137	0.053	0.201	0.211
PM6:CH-6F	0.872	1.429	0.557	1.167	0.264	1.108	0.059	0.234	0.236
PM6:CH-ThCl:CH6F	0.932	1.440	0.508	1.176	0.264	1.134	0.042	0.202	0.208

 $E_{g}$  was estimated via the crossing points between normalized absorption and PL spectra of films;  $^{b)}V_{oc,sq}$  is calculated according to the SQ limit;  $^{c)}\Delta E_{1} = E_{g} - V_{oc,sq}$  $\Delta E_{g}$  was estimated via the clossing points octation hormanics accurate in the response of  $\Delta E_{g}$  ( $\Delta E_{g} = q\Delta V_{nr}$ ) is determined by two approaches:  $e^{(t)} V_{oc,rad}$  is the  $V_{oc}$  when there is only radiative recombination and are calculated from EL and sEQE measurements.  $\Delta E_{g}$  ( $\Delta E_{g} = q\Delta V_{nr}$ ) is determined by two approaches:  $e^{(t)} c_{oc,rad} = -kT \ln EQE_{EL}$  by measuring the device EQE<sub>EL</sub>.

equation of  $\Delta E_3 = -kT \ln(EQE_{EI})$ . In this work, as summarized in Table 2, the binary and ternary devices show the nearly same  $\Delta E_1$  (0.265/0.264 eV) and very small  $\Delta E_2$  below 0.1 eV. As shown in Figure S4 (Supporting Information), the PM6:CH-ThCl and ternary devices show high EQE<sub>FI</sub> (8.4  $E^{-4}$ ) and afford  $\Delta E_3$  with values of 0.211 and 0.208 eV, respectively according to the equation of  $\Delta E_3 = -kT \ln(EQE_{EL})$ , which are smaller than that of the PM6:CH-6F binary device. As reported by Gao et al., the limit of  $\Delta E_3$  in OSCs is defined by the photoluminescence efficiencies of the pristine material components.<sup>[15]</sup> To explain the low  $\Delta E_3$ values of the PM6:CH-ThCl and ternary devices, the photoluminescence quantum vield (PLQY) of CH-ThCl, CH-6F, and their blend were measured under the same conditions. As shown in Figure S5 (Supporting Information), the blending film of CH-ThCl and CH-6F with the same ratio in the ternary device demonstrates a PLQY of 5.96%, higher than that of CH-ThCl with a value of 5.55%. The high PLOY of CH-ThCl and its blend film might originate from their enhanced J-aggregations in the solid films, which has proved to favor high luminescence efficiencies according to the established results.<sup>[51]</sup>

#### 2.4. Excition and Charge Dynamics Analysis

The exciton and charge dynamics were analyzed to uncover the underlying reasons for the different performances of the binary and ternary devices. First, the space-charge-limited current (SCLC) method was employed to measure the charge mobilities of the blend films, PM6:CH-ThCl and PM6:CH-ThCl:CH-6F. As depicted in Figure 3a, the ternary blend exhibited higher and more balanced electron/hole mobilities, with values of 4.46  $\times$  $10^{-4}/4.42 \times 10^{-4} \text{ cm}^{-2} \text{ V}^{-1} \text{ s}^{-1}$ , compared to the binary blend. This enhanced mobility favored the achievement of high FF and  $J_{\rm sc}$  in the ternary device.<sup>[52]</sup> Next, the dependence of photocurrent density  $(J_{ph})$  on effective voltage  $(V_{eff})$  was measured to investigate the exciton dissociation properties of the two devices (Figure 3b).<sup>[53]</sup> The exciton dissociation probability ( $P_{diss}$ ), calculated from  $J_{\rm ph}$  under the short-circuit condition divided by the saturated photocurrent density ( $J_{sat}$ ), were 97.1% and 98.1% for the binary and ternary devices, respectively. Furthermore, the behavior of charge recombination in the two devices was studied by measuring the light-intensity dependence (P) of  $J_{sc}$ ,  $J_{sc}$ 



Figure 3. a) The carrier transport properties of the optimized devices. b)  $J_{ph}$  versus  $V_{eff}$  characteristics, c)  $J_{sc}$ , and d)  $V_{oc}$  versus light intensity of the optimized devices.

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Figure 4. a) AFM height image of the binary device. b) AFM height image of the ternary device. c) TEM image of the binary device. d) TEM image of the ternary device. e) GIWAX pattern of the binary device. f) GIWAX pattern of the ternary device.

 $\propto P\alpha$ , was measured.<sup>[54]</sup> As shown in Figure 3c, the  $\alpha$  values were 0.991 and 0.993, respectively, indicating less bimolecular charge recombination for the binary and ternary devices. Additionally, the trap-assisted recombination behaviors were investigated from the plotting of the  $V_{\rm oc}$  versus light intensity. The binary and ternary devices featured small slopes with values of 1.109 and 1.091 kT/q, respectively, suggesting a reduced occurrence of trap-assisted recombination (Figure 3d).<sup>[55]</sup>

#### 2.5. Morphology Analysis

The morphologies of the PM6:CH-ThCl and PM6:CH-ThCl:CH-6F blends were characterized using atomic force microscopy (AFM) and transmission electron microscopy (TEM). As depicted in Figure 4a,b, both the binary and ternary blend films exhibited fibrillar structures, with root-mean-square roughness  $(R_{a})$  values of 1.32 and 1.19 nm, respectively. The ternary blend film displayed a smoother surface morphology compared to the binary blend film, which was consistent with the TEM results. As shown in Figure 4c,d, the PM6:CH-ThCl blend film exhibited clear aggregates and non-uniform domains, whereas the ternary film of PM6:CH-ThCl:CH-6F exhibited a fine and uniform structure without large domains and aggregates. This uniform structure provides sufficient interfacial areas for efficient exciton dissociation and charge transport pathways.<sup>[56]</sup> Grazing incidence wideangle X-ray scattering (GIWAXS) was employed to investigate the molecular stacking and orientation in the two blend films. As shown in Figure 4e,f, both blend films exhibited a face-on orientation with distinct  $\pi$ - $\pi$  stacking (010) signals in the out-of-plane (OOP) direction, located at  $\approx$ 1.717 and 1.712 Å<sup>-1</sup>, respectively. This indicates that the ternary film maintained the same stacking distance as the binary film. As summarized in Table S7 (Supporting Information), the ternary film exhibited a slightly larger CCL (41.27 Å) in the (010) OOP direction compared to the PM6:CH-ThCl binary film (38.37 Å). This suggests that the presence of CH-6F improved the molecular packing and carrier dynamics in ternary devices, resulting in simultaneous improvements in  $J_{\rm sc}$  and FF values for the ternary device.

Active layer morphologies are significantly influenced by the miscibility between donors and acceptors, which can be evaluated empirically using the Flory-Huggins interaction parameter ( $\chi$ ). The contact angles of water and ethylene glycol droplets on the surfaces of various films were measured, and the surface tensions and Flory-Huggins interaction values ( $\chi$ ) were calculated and listed in Table S8 (Supporting Information). The Flory-Huggins interaction between PM6 and CH-ThCl is 0.38, which is higher than the  $\chi$  value of 0.18 for PM6:CH-6F. In contrast, a lower  $\chi$  value of 0.17 was observed for PM6 between CH-ThCl:CH-6F, indicating better miscibility in the ternary blend. In addition, the  $\chi$  value for CH-6F:CH-ThCl was calculated to be 0.036. To check whether the two acceptors form an alloy, we conducted the differential scanning calorimetry (DSC) measurement (see Figure S8, Supporting Information). Upon heating, CH-6F and CH-ThCl exhibited melting transition peaks (T<sub>m</sub>) at 257 and 281 °C, respectively. In the CH-6F:CH-ThCl blend, there still exist the characteristic  $\rm T_m$  of CH-6F and CH-ThCl. The results indicate that there is no alloy-like phase present in the CH-6F:CH-ThCl

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blend.<sup>[57]</sup> These results are consistent with the findings from photoluminescence (PL) measurements. As depicted in Figure S9 (Supporting Information), the steady-state PL quenching efficiency was calculated by comparing the PL intensity ratio of the blend films to that of the neat films. The PL quenching efficiency serves as an indicator of exciton dissociation and charge transfer at the donor-acceptor interface and reflects the miscibility and morphology of the blend films. In Figure S9a,b (Supporting Information), the binary film PM6:CH-ThCl and ternary film PM6:CH-ThCl:CH6F exhibited PL quenching efficiencies of 94.1% and 97.1%, respectively, relative to the neat films. For the binary and ternary films relative to the PM6 neat film, PL quenching efficiencies of 98.6% and 99.0% were obtained for CH-ThCl and CH-ThCl:CH6F, respectively. The higher PL quenching efficiency of the ternary film can be attributed to the improved miscibility of the components in the ternary blend.[58,59]

# 3. Conclusion

In summary, we have designed an acceptor CH-ThCl, featuring an extended conjugation central core dichlorodithienoquinoxaline. CH-ThCl exhibits a high photoluminescence quantum yield (PLQY) attributed to its extended and rigid conjugation structure, which effectively suppresses nonradiative energy loss in photovoltaic devices. The binary device based on PM6:CH-ThCl achieved a remarkable PCE of 18.16% with a high  $V_{oc}$  of 0.934 V, thanks to its low nonradiative energy loss of 0.21 eV. To further enhance device performance, we introduced another acceptor. CH-6F, as the third component, resulting in a ternary blend film with improved morphology, mainly due to enhanced miscibility between the acceptors and PM6. Compared to the binary device, the ternary device exhibited an impressive PCE of 18.80% with enhanced  $J_{sc}$  and FF, while maintaining a high  $V_{oc}$ . These findings emphasize the significance of suppressing nonradiative energy loss and optimizing morphology for achieving highperformance OSCs. It is anticipated that combining active layer design and morphology regulation will enable the development of even more efficient OSCs in the future.

# **Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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# **Conflict of Interest**

The authors declare no conflict of interest.

# **Data Availability Statement**

The data that support the findings of this study are available in the supplementary material of this article.

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