An electron acceptor with a truxene core and ring-fused perylene diimide (PDI) tripolymer annulated by selenium (Se) branch, named as FTr-3PDI-Se, is designed and synthesized. FTr-3PDI-Se exhibits large conjugated planar conformation, strong absorption spectra in the regions of 300–400 and 450–550 nm, the deep HOMO energy level of 6.10 eV, and high decomposition temperature above 400°C. The FTr-3PDI-Se: PBDB-T-2Cl based device achieved a disappointing power conversion efficiency (PCE) of 1.6% together with a high $V_{oc}$ of 1.12 V. The low PCE was due to the large aggregates of blend film, the imbalanced hole/electron transport and low PL quenching efficiencies. The high $V_{oc}$ can be attributed to the high-lying LUMO level of FTr-3PDI-Se and the low-lying HOMO level of PBDB-T-2Cl. Our research presents an interesting and effective molecule-designing method to develop non-fullerene acceptor.

Keywords: organic solar cells, non-fullerene acceptor, truxene, perylene diimides, selenium

INTRODUCTION

Organic solar cells (OSCs) have attracted boundless interest over the past few decades owing to the advantages of light weight, low cost, wide source, and large-scale roll-to-roll printing process (Kang et al., 2016; Hou et al., 2018). Recently, the fullerene acceptors, due to their numerous of disadvantages of weak absorption, limited structural modifications and electronic energy levels non-tunability, were marginalized by non-fullerene acceptors (NFAs) (Cheng et al., 2018; Yan et al., 2018). Significant progress in NFAs-based OSCs has been achieved with power conversion efficiency (PCE) over 18% (Lin et al., 2020; Liu Q. et al., 2020; Zhan et al., 2021). Among the widely reported NFAs, fused-ring electron acceptors (FREAs) and perylene diimide derivatives (PDIs) are the two main study directions.

Because of the strong electron affinity, high absorption coefficient and electron mobility, as well as energy-level tunability, PDIs are widely developed (Chen et al., 2018; Li and Wonneberger, 2012; Liu et al., 2016; Sun et al., 2016; Feng et al., 2018; Agnieszka and Frank, 2019; Li M. Y. et al., 2020). The large conjugated skeleton of PDI exhibits strong aggregation tendency, which may result in self-trapping of light excitons and afterwards generate fast bimolecular recombination of charge...
carriers, limiting the high performance of OSCs (Sharenko et al., 2013; Liu S. Y. et al., 2015). The researcher verified that changing the planarity of the PDIs is the popular methods to avoid this strong aggregation (Zhong et al., 2014, 2016; Lin et al., 2016; Zhang et al., 2016; Duan et al., 2017a; Liu X. et al., 2017; Liu et al., 2018). For example, various 3D electron acceptors with the central aromatic core (atom) and twisted PDI trimers or tetramer were investigated (Liu Y. H. et al., 2015; Lee et al., 2016; Zhan et al., 2017; Zhang A. D. et al., 2017; Lin et al., 2018a; Liu W. X. et al., 2020). A twisted configuration of PDIs is confirmed effectively to avoid large aggregation. However, the single bonding connection between central core and PDIs would weaken charge mobility due to an excessive twist geometry, giving a low OSCs performance. Therefore, the proper twisted non-planar structures, i.e., good balance of desirable film morphology with proper domain size and sufficient charge transport ability seems to be the key point for developing high-performance PDI electron acceptors (Lin et al., 2018a).

Interestingly, oxidative ring-fusion between the central aromatic core and the PDI branches was verified to be an effective strategy to achieve an exquisite balance aforesaid for high OSCs performance (Hartnett et al., 2016; Meng et al., 2016a, 2017; Zhong et al., 2016; Wang et al., 2017; Zhang J. Q. et al., 2017; Lin et al., 2018a; Chen et al., 2020). The fused PDI NFAs all exhibited better planarity than non-fused counterparts, since the aromatic core and PDI branches were locked by the adjacent benzene. Meanwhile, the fused PDI NFAs showed stronger intermolecular π-π stacking and higher electron mobility (Lin et al., 2018a). Moreover, these fused PDI NFAs generated proper phase separation with proper domain size and high domain purity when blended with donors (Chen et al., 2018; Hu et al., 2018; Wu et al., 2019). Therefore, the fused PDI NFAs displayed better OSCs properties compared with unfused ones (Li et al., 2016; Meng et al., 2016a, 2017; Liu X. F. et al., 2017; Wang et al., 2017; Zhang J. Q. et al., 2017; Lin et al., 2018a; Yin et al., 2019; Carlotti et al., 2020).

Recently, several studies showed that five-membered heteroatom-annulated (nitrogen/chalcogen-fused in bay regions) of PDIs has been regarded as the most effective molecular design strategy to achieve high performance OSCs (Sun et al., 2015; Meng et al., 2016a; Cann et al., 2017). The five-membered heteroatom-annulated PDI NFAs reinforced intra- and intermolecular interactions, leading to high electron mobility, which achieved improved PCEs. Among the varied nitrogen/chalcogen, the selenium atom (Se), since its enormous and loose electron cloud, is much easier to realize orbital overlap between the adjacent PDI NFAs, afterwards enhance the charge carrier mobility (Meng et al., 2016b; Li et al.,

![Scheme 1](image1.jpg)

**Scheme 1** | Chemical structure and synthetic routes of FTr-3PDI-Se.

![Figure 1](image2.jpg)

**Figure 1** | Views of the optimized geometries of FTr-3PDI-Se, and the LUMO/HOMO electron distribution obtained using DFT calculations at the B3LYP/6-31G(d) level.
Moreover, due to the natural easy-polarizing characteristic of the Se atom, the Se-annulated PDIs exhibit the stronger intra- and intermolecular interactions, which also confirmed the important application foreground of Se-annulation PDIs in non-fullerene OSCs (Duan et al., 2017b; Yin et al., 2018; Li et al., 2019; Luo et al., 2019; Qureshi et al., 2020; Wang et al., 2020).

Truxene has been demonstrated as a promising skeleton to construct high performance NFAs (Nielsen et al., 2013, 2014; Lin et al., 2018b; Wu et al., 2018). Inspired by the above achievements of Se-annulated PDIs herein, we report the design and synthesis of truxene functionalized star-shaped NFAs with fused selenium-annulated PDIs, named FTr-3PDI-Se (Scheme 1). The devices based on poly[(2,6-(4,8-bis(5-(2-ethylhexyl-3-chloro)thiophen-2-yl)benzo[1,2-b:4,5-b']dithiophene))-alt-(5,5-(1′,3′-di-2-thienyl-5′,7′-bis(2-ethylhexyl)benzo[1′,2′-c:4′,5′-c']dithiophene-4,8-dione)] (PBDB-T-2Cl): FTr-3PDI-Se exhibited a PCE of 1.6% with a high open-circuit voltage ($V_{oc}$) of 1.12 V. The FTr-3PDI-Se exhibited large conjugated planar skeleton that can effectively promote the blend films to form large aggregates, which may lead to bimolecular recombination, limiting the OSCs performance.

### RESULT AND DISCUSSION

#### Material Synthesis and Characterization

The synthetic routes of FTr-3PDI-Se was presented in Scheme 1 and the detailed synthetic procedure was provided in the Supporting Information. Compounds FTr-3PDI was synthesized according to the reported method (Lin et al., 2018a). FTr-3PDI-NO$_2$ was prepared with a high yield of 95% using the fuming HNO$_3$. Finally, the three fused selenium-annulated PDIs branches based on truxene, FTr-3PDI-Se, was synthesized by reductive cyclization reaction with Se powder. The as-synthesized FTr-3PDI-NO$_2$ and FTr-3PDI-Se were fully characterized by $^1$H NMR, $^{13}$C NMR, and MALDI-TOF mass spectrometry (Supplementary Figures 1–6). Although large conjugated planar conformation, FTr-3PDI-Se electron acceptor displays moderate solubility in section of organic solvents such as chloroform, toluene, and chlorobenzene at room temperature. We ascribe it to the six hexyl chains of the truxene core.

#### Theoretical Calculations

The geometry and electron distribution of FTr-3PDI-Se was presented by employing the density functional theory (DFT) method at the B3LYP/6-31G(d,p) level in the Gaussian 09 software, where the long alkyl chain (–C$_6$H$_{13}$ of the truxene core and –C$_5$H$_{11}$ of the PDIs branches) was simplified to methyl groups (Figure 1). Obviously, FTr-3PDI-Se exhibits an overall planarity structure from the top view and side view. According to the optimized geometry, the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) electron distribution were calculated. The LUMO is distributed on two fused selenium-annulated PDIs sub-group. The HOMO is localized on one two fused selenium-annulated PDIs and truxene. The different wave function distributions between HOMO and LUMO are attributed to the degenerate orbital/multiple...
resonance configurations of the three fused selenium-annulated PDI groups. Furthermore, the calculated LUMO and HOMO levels were $-3.30$ and $-6.00$ eV.

**Thermodynamic, Optical, and Electrochemical Properties**

Thermogravimetric analysis (TGA) measurement (Figure 2A) demonstrated that FTr-3PDI-Se showed outstanding thermal stability along with a high decomposition temperature ($T_d$, 5% weight loss) exceeding $400^\circ$C under nitrogen atmosphere, benefiting from large conjugated planar conformation. Afterwards, differential scanning calorimetry (DSC) was performed without obvious endo- and exothermal peaks from room temperature to $320^\circ$C in the second heating cycle (Figure 2B). The spectrum of FTr-3PDI-Se in chloroform solution showed two sets of absorption bands in the range of $300–600$ nm. The short wavelength region displayed a maximal sharp peak of $360$ nm with two broad shoulder peak, while the longer wavelength region exhibited the maximal peak of $500$ nm with two broad shoulder peak as well (Figure 2C). FTr-3PDI-Se in thin film showed similar absorption spectra outline to their solution ones, indicating that the intermolecular aggregation is effective suppressed. Meanwhile, FTr-3PDI-Se demonstrated a slightly large optical bandgap of $2.24$ eV with optical absorption onsets $555$ nm ($E_{opt} = 1240/\lambda_{onset}$ eV). The absorption profiles of FTr-3PDI-Se is complementary to the strong absorption of PDBT-T-2Cl donor, which was exhibited in Supplementary Figure 7. The electrochemical property of FTr-3PDI-Se in chloroform solution was investigated by CV, as shown in Figure 2D. The half-wave potential of Fc/Fc$^+$ was measured to be $0.40$ V, and the energy levels of HOMO and LUMO were estimated from the onset oxidation ($E_{onset}^{ox}$) and reduction ($E_{onset}^{red}$) potentials by equations:

$$E_{HOMO} = -e(E_{onset}^{ox} - E_{Fc/Fc^+} + 4.8)$$

$$E_{LUMO} = -e(E_{onset}^{red} - E_{Fc/Fc^+} + 4.8)$$

respectively (Li et al., 1999). The HOMO/LUMO levels are $-6.10/−3.65$ eV. The slightly high-lying LUMO level cooperate with low-lying HOMO level of donor will contribute to achieve a high $V_{oc}$. Meanwhile, the down-shifted HOMO level maintain the excellent chemical durability, and is favorable for hole transfer from excited acceptor to donor in OSCs (Duan et al., 2016, 2017b; 2018; Jia et al. 2017).
Photovoltaic Properties

The OSCs devices were prepared and measured with a conventional device structure of ITO (indium-tin oxide)/PEDOT:PSS (poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate))/PBDB-T-2Cl: FTr-3PDI-Se/PFN-Br (poly[(9,9-bis(3’-((N,N-dimethyl)-N-ethylammonium)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)])/Ag (Figure 3A). PDBT-T-2Cl was picked as the medium-bandgap donor to matched FTr-3PDI-Se acceptor benefiting from their complementary absorption and appropriate energy levels. The devices were fabricated and evaluated in terms of donor/acceptor weight ratios, solvent additives, and thermal annealing. All the device parameters under the mentioned above conditions are listed in Supplementary Tables 1–3. The optimal devices fabrication is that chlorobenzene as the main processing solvent with 1% chloronaphthalene solvent additives, and the annealing temperature is 120°C. The total concentration of PDBT-T-2Cl and FTr-3PDI-Se was optimized to be 20 mg mL\(^{-1}\) with the donor:acceptor weight ratio of 1.5:1. The optimized device parameters are summarized in Table 1, and the corresponding J–V curves are shown in Figure 3B. The optimized OSC device based on PBDB-T-2Cl: FTr-3PDI-Se exhibited a PCE of 1.6% with a high \(V_{oc}\) of 1.12 V, but a relatively poor short-circuit current density \(J_{sc}\) of 3.6 mA cm\(^{-2}\) and a fill factor (FF) of 38.9%. The high \(V_{oc}\) is consistent with the high-lying LUMO level of FTr-3PDI-Se and low-lying HOMO level of PDBT-T-2Cl.

The external quantum efficiency (EQE) spectra of PBDB-T-2Cl: FTr-3PDI-Se blend films were collected from the above optimized devices and displayed in Figure 3C. The calculated \(J_{sc}\) of 3.5 mA cm\(^{-2}\) from the EQE spectra was consistent with the measured \(J_{sc}\) (Table 1). The continuous EQE responses between 300 and 700 nm for the PBDB-T-2Cl: FTr-3PDI-Se based device results from the complementary absorption of PBDB-T-2Cl: FTr-3PDI-Se blend film (Supplementary Figure).

Charge Transport and Recombination

The charge transport were acquired by single-carrier devices with a device structure of ITO/ZnO/PBDB-T-2Cl: FTr-3PDI-Se/Ca/Al for electron only devices and ITO/PEDOT:PSS/PBDB-T-2Cl: FTr-3PDI-Se/MoO\(_3\)/Ag for hole only devices, respectively (Supplementary Figure 8). The hole mobilities \(\mu_h\) of PBDB-T-2Cl: FTr-3PDI-Se blend film was estimated to be 4.5 \(\times\) 10\(^{-6}\) cm\(^2\) V\(^{-1}\) s\(^{-1}\). In contrast, the electron mobility \(\mu_e\) was measured to be 2.2 \(\times\) 10\(^{-4}\) cm\(^2\) V\(^{-1}\) s\(^{-1}\), which are two orders of magnitude higher than \(\mu_h\). The low hole mobility and highly imbalanced \(\mu_e/\mu_h\) seriously suppress the charge transport and give rise to more bimolecular recombination, which in turn acquire low FF and \(J_{sc}\).

The photoluminescence (PL) quenching experiments were proceeded to study the charge transfer efficiency. As shown in Supplementary Figure 9, the PL quenching efficiencies of PBDB-T-2Cl: FTr-3PDI-Se blend films are 83.3 and 47.7% as compared to the neat PBDB-T-2Cl and FTr-3PDI-Se films, respectively, suggesting a moderate exciton dissociation efficiency.

Morphology

The surface morphology of PBDB-T-2Cl: FTr-3PDI-Se blend films were investigated using atomic force microscopy (AFM). The film exhibited obvious phase separation with nanofibrillar structures (Figure 4), forming a relative coarse surface with a RMS surface roughness of 3.97 nm. The large planar conformation of FTr-3PDI-Se, can effectively promote the blend films to form large aggregates.
CONCLUSION
In summary, FTr-3PDI-Se was synthesized and employed as electron acceptors for organic solar cells. The optimized devices based on PBDB-T-2Cl: FTr-3PDI-Se displayed a PCE of 1.6%, which was attributed to the following reasons. The conjugated planar conformation of FTr-3PDI-Se, verified by the DFT quantum calculation, can effectively promote the blend films to form large aggregates, which impeded the charge transport. Meanwhile, the imbalanced hole/electron transport and low PL quenching efficiencies seriously obstruct the charge transport and reduce exciton dissociation efficiency. Obviously, this research missed the balance between the highly twisted non-planar reduce exciton dissociation efficiency. Taking the excellent advantages into consideration and discard the disadvantages, we expect that the combination of the fused selenium-annulated PDIs with other conformation cores will create more promising and practical acceptors.

DATA AVAILABILITY STATEMENT
The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding authors.

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AUTHOR CONTRIBUTIONS
KL and FH: designed experiments. KL, YC, and CD: analyzed experimental results. KL and CD: wrote the manuscript. All authors contributed to the article and approved the submitted version.

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SUPPLEMENTARY MATERIAL
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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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