“N-π-N” Type Oligomeric Acceptor Achieves an OPV Efficiency of 18.19% with Low Energy Loss and Excellent Stability

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A novel “N-π-N” type oligomeric acceptor of 2BTP-2F-T, constructed by two small non-fullerene acceptor (NFA) units linked with a thiophene π bridge is reported. The 2BTP-2F-T not only combines the advantages of small NFA and polymeric acceptors (PYF-T-o) with similar units but also exhibits superior characteristics of high absorption coefficient and high electron mobility ($\mu_o$) with less dependence on molecular packing. Using PM6 as the donor, a remarkable efficiency of 18.19% is obtained with an open circuit ($V_{oc}$) of 0.911 V, short current circuit ($J_{sc}$) of 25.50 mA cm$^{-2}$, and fill factor (FF) of 78.3%, which is much better than that of the corresponding monomer (16.54%) and PYF-T-o (15.8%) based devices. The much-improved efficiency results from two aspects: 1) an enhanced FF due to the largely improved $\mu_e$ and well-controlled morphology; 2) a higher value of ($J_{sc} \times V_{oc}$) due to its higher absorption coefficient and efficient charge generation at a similar low energy loss. Furthermore, the PM6/2BTP-2F-T device possesses the longest $T_{80}$ lifetime to light-soaking and comparable high thermal stability with PM6/PYF-T-o. The results indicate that the “N-π-N” type oligomeric acceptor has a great application prospect due to its superior high efficiency and improved stability in organic solar cells.

1. Introduction

Organic solar cells (OSCs) show excellent application prospects in flexible and portable devices because of their light-weight property, flexibility, and low cost. The polymer-donor-based OSCs, according to the compositions of the active layer, fall into two categories: polymer-small molecule acceptor and all-polymer OSCs. The invention of small molecular non-fullerene acceptors (NFA), such as ITIC, Y6, and their analogs, has pushed the OSC efficiency to higher than 18%, and all-polymer OSCs efficiency over 17% with high thermal stability.

An oligomer-type acceptor with higher molecular weight (MW) would be expected to combine the advantages of both, because 1) compared to its small NFA counterpart, it is anticipated to decrease the diffusion rate in the blend films by the vastly increased MW, improving the device stability; 2) compared to its polymeric counterparts, it could obtain definite molecular structure, absence of long chains entanglements and increased diffusion coefficient, leading to an easier morphology control and improved device performances.

Then, the main point is how to construct an oligomeric acceptor to achieve the advantages mentioned above. The traditional oligomers feature identical structures with definite repeating units, and their MW should be located between 1000 of the “small molecules” and 10 000 of the polymers. Currently, small molecules of typical NFA and small molecular donors sometimes are also called oligomeric molecules with MWs of 1500–2000. Obviously, the MW of oligomers with less than 2500 can hardly meet our requirements. Oligomeric conjugated molecules with three or more definite arms are also reported. However, their MW is still hardly larger than 2500, due to their small repeating units such as the perylene diimine, naphthalene diimide, diketopyrrolopyrrole, or their derivatives; additionally, their poorer absorption spectrum or non-ideal charge transport resulted from their non-planar molecular backbone or non-ideal morphology limit their device performances.

That is a similar situation for ladder-core armed with electron-deficient end groups. Motivated by the excellent device performance of the polymer acceptor with a moderate MW ($\approx$20 000) and also the rigid molecular backbone of Y-series NFA, linking the efficient Y-series NFA in a proper sequence might be a possible strategy to construct our objective oligomer-type acceptors.

In this article, we designed and successfully synthesized an “N-π-N” type of oligomeric acceptor 2BTP-2F-T (Figure 1a) with
a high MW (3359) through linking two efficient Y-series NFA molecules (N type) by the thiophene \( \pi \) bridge. Compared to corresponding monomer and polymeric acceptor (PYF-T-o) with similar building units, the N-\( \pi \)-N oligomeric acceptor 2BTP-2F-T exhibits not only a combined advantage of them, but also superior molecular properties, including an in-between absorption range but with a higher absorption coefficient, and enhanced electron mobility with moderate molecular packing. Using PM6 as the donor, 2BTP-2F-T obtained a remarkable power conversion efficiency (PCE) of 18.19% with an open-circuit voltage \( (V_{oc}) \) of 0.911 V, a short-circuit current \( (J_{sc}) \) of 25.50 mA cm\(^{-2}\), and a fill factor (FF) of 78.3%, which is much higher than that of the corresponding monomer (16.5%) and PYF-T-o (15.9%) based devices. The excellent device performance for PM6/2BTP-2F-T is a combined result of improved FF and enhanced value of \( (J_{sc} \times V_{oc}) \) with a small energy loss (0.53 eV), due to the joint contributions of its excellent material property, and easily well-tuned morphology with a small Urbach energy of 22.74 meV. Additionally, the encapsulated PM6/2BTP-2F-T device possesses the longest \( T_{20} \) lifetime (7 times longer than PM6/monomer, and 2.4 times longer than PM6/polymer) under illumination in the air condition, and its thermal stability is also comparable to that of PM6/PYF-T-o. The result indicates that the “N-\( \pi \)-N” type oligomeric acceptor has promising application due to its ability to obtain maximized efficiency and improved stability in OSCs.

2. Results and Discussion

2.1. Material Design and Molecular Properties of 2BTP-2F-T

The monomer was synthesized through an asymmetrical Knoevenagel reaction from its precursor BTP-CHO (Scheme S1, Supporting Information). To improve the yield of the reported asymmetrical Knoevenagel reaction, we adopted a two-step heating method (reacted at room temperature for 3 h, then heated to 65 °C overnight) to ensure the selectivity of the reaction, and the yield was slightly improved to 41.5% \([24,25]\). The oligomeric acceptor 2BTP-2F-T was prepared through Stille coupling with monomer and linker with a high yield of 70%. Consequently, compared to the symmetrical NFA or the monomer, the synthesis of 2BTP-2F-T only takes an extra step but with a high yield; in contrast to the polymer (PYF-T-o), the yield of 2BTP-2F-T was probably
higher due to its easier purification, no MW distribution, and no batch-to-batch variations. Noteworthy, because the performance of commercial PJ1 (23) is not as good as PYF-T-o (7), we use PYF-T-o as its corresponding control polymer.

As seen in the top view of 2BTP-2F-T (Figure 1a), the dihedral angle between the linker and monomer is ∼20°, which is decreased by ∼15° than the compound without the linker (Figure S1, Supporting Information). This result demonstrates that the thiophene linker well alleviates the crowded space between the two end-capped acceptors, which leads to a more planar molecular backbone and a longer effective conjugated length. Additionally, 2BTP-2F-T exhibited good solubility in common solvents, including chloroform (CF), tetrahydrofuran (THF), and chlorobenzene (CB), demonstrating its good solution process-ability.

The UV–vis absorption spectra of 2BTP-2F-T, monomer, and PYF-T-o were investigated and compared both in the CF solutions and neat films, and the detailed data are summarized in Table S1, Supporting Information. As shown in Figure 1b, there is a big disparity in their absorption spectrums for the three molecules in the CF solution. Compared to 2BTP-2F-T, PYF-T-o exhibits the red-shifted absorption while the monomer shows the blue-shifted absorption spectrum, which should be due to easier pre-aggregation for the polymer and the increased effective length for the dimer. The tendency of their absorption-onset variation in the film was consistent with their solution; however, their differences become smaller, which is ascribed to the largest red-shifts for the monomer and the least red-shifts for the polymer (Figure 1c). The variations in the solution and film differences result from their different assembly abilities, which will be discussed in the following. The absorption shape of 2BTP-2F-T is more similar to the monomer than that of PYF-T-o, indicating that 2BTP-2F-T retains the absorption properties of the monomer. Importantly, 2BTP-2F-T presents the highest photon absorption capacity due to its most broadened absorption spectrum from 600 to 900 nm and its highest absorption coefficient (1.33 × 10⁵ cm⁻¹) at the maximized absorption peak. The superior absorption for 2BTP-2F-T should be the combined effect of its higher absorption co-efficient of dimer in solution due to its increased molecular conjugation (Figure S2, Supporting Information) and well-assemble ability, indicating a probable high J_rc for the devices. Noteworth- y, although PYF-T-o exhibits a slightly red-shifted absorption, it exhibits a much lower absorption coefficient in the range of 600–800 nm than that of monomer and 2BTP-2F-T.

The square-wave voltammetry (SWV; Figure S3, Supporting Information) was carried out to measure their electrochemical properties, and their frontier energy levels are aligned in Figure 1d. The frontier energy levels were calculated from the onset oxidation (E₁ox) and reduction (E₁red) potentials using the following equations: $E_{HOMO} = -\epsilon (E_{1ox} - E_{Fc/Fc^+} + 4.8)$ and $E_{LUMO} = -\epsilon (E_{1red} - E_{Fc/Fc^+} + 4.8)$. The calculated highest occupied molecular orbital (HOMO) energy levels were −5.68, −5.67, and −5.64 eV for monomer, 2BTP-2F-T, and PYF-T-o, respectively. And their corresponding lowest unoccupied molecular orbital (LUMO) energy levels were −3.81, −3.84, and −3.82 eV. In comparison to the monomer, 2BTP-2F-T shows a slightly higher HOMO but lower LUMO energy level, which is consistent with its slightly red-shifted absorption and indicates a probable decreased $V_{OC}$. Summarily, 2BTP-2F-T exhibits similar in-between energy levels compared with monomer and PYF-T-o, which is a matched energy level with PM6 (Figure 1d).

The grazing incidence wide-angle X-ray scattering (GIWAXS) and contact angle measurements were carried out to characterize the molecular packing at thermal annealing (TA) condition (TA = 100 °C), and surface tension of three materials. As shown in their 2D images and 1D curves (Figure 2a,b), the 2BTP-2F-T adopts a preferential face-on packing mode, the same as that of the PYF-T-o and monomer, illustrated from their prominent (010) peak in out-of-plane (OOP) direction and (100) peak in-plane (IP) direction. The calculated π–π stacking distance for (010) peak in the OOP direction is 3.64, 3.79, and 3.87 Å, for monomer, 2BTP-2F-T, and PYF-T-o, respectively; their corresponding coherence crystal length (CCL) is 24.9, 18.6, and 16.9 Å, respectively. The d-spacings and CCLs of 2BTP-2F-T are located in the middle of monomer and PYF-T-o, indicating its in-between molecular assemble ability, which well explains its intervening absorption red-shifts from solution to film as we discussed above. This inferior molecular packing of 2BTP-2F-T than that of monomer is probably due to its slightly un-planar molecular backbone resulted from the joint of the two monomers; its more ordered packing than that of polymer should be attributed to its much weaker or absence of molecular chain entanglement, which could be observed from the increased q value of the (100) peak (Figure 2b).

Out of our expectation, although 2BTP-2F-T exhibits moderate molecular packing ability and π–π stacking distance, however, it obtains the highest averaged electron mobility ($\mu_\text{E}$) of 1.55 × 10⁻³ cm² V⁻¹ s⁻¹, which is slightly higher than that of the monomer (9.17 × 10⁻⁴ cm² V⁻¹ s⁻¹) and much higher than that of PYF-T-o (2.20 × 10⁻⁴ cm² V⁻¹ s⁻¹) (Figure 2c). Compared to the $\mu_\text{E}$ of the monomer, the higher value for 2BTP-2F-T with less molecular packing ability is probably due to its more contribution of charge transport in intra-molecules, resulting from its double prolonged molecular backbone in 2BTP-2F-T. [26,27] Importantly, the $\mu_\text{E}$ with less dependence on molecular packing ability indicates that the N-aryl-N type oligomeric acceptor owns a better tolerance to morphology, which is a prominent advantage over its monomer and polymer due to the morphology control being a tough-nut in all currently reported type of OSCs.

The change of the molecular backbone would also impact the molecular surface tensions and lead to a different Flory–Huggins interaction parameter $\chi$, and ultimately act on the morphology. The surface tensions calculated from contact angle (Figure 2d and Figure S4, Supporting Information) were 15.24, 15.92, and 16.42 mN m⁻¹ for monomer, 2BTP-2F-T, and PYF-T-o, respectively, all of which were close to that of PM6 (13.08 mN m⁻¹). The in-between surface tension for 2BTP-2F-T leads to an intervening Flory–Huggins interaction parameter $\chi$ for PM6/2BTP-2F-T ($\chi = 0.14$), compared to that of PM6/monomer (0.08) and PM6/PYF-T-o (0.19).

2.2. Fabrication and Device Performance of the OSCs

Using PM6 as the donor, a conventional device with a structure of ITO/2-(9H-carbazol-9-yl)ethylphosphonic acid (2PACz)/active layer/poly[(9,9-bis(3′-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-5,5′-bis(2,2′-thiophene)-2,6-naphthalene-1,4,5,8-tetracaboxlyc-N,N′-di(2-ethylhexyl)imide] (PNDIT-F3N)/Ag was
fabricated to investigate the device performance of 2BTP-2F-T (Figure 3a). The fabrication conditions were carefully tuned by adjusting the ratios between donor and acceptor, thermal annealing temperatures, concentrations of additives, and different hole/electron transport layers to improve the device performance. The detailed optimization results and conditions are summarized in Tables S2–S4, Supporting Information. Surprisingly, the device based on PM6/2BTP-2F-T exhibits a superior PCE of 18.19% with a $V_{oc}$ of 0.911 V, a $J_{sc}$ of 25.50 mA cm$^{-2}$, and an FF of 78.3%, which is much better than that of PM6/monomer.
and PM6/PYF-T-o. The best PCE for PM6/monomer is 16.5% with a $V_{oc}$ of 0.925 V, a $J_{sc}$ of 24.4 mA cm$^{-2}$, and an FF of 73.4%; and the best PCE for PM6/PYF-T-o was 15.9% with a $V_{oc}$ of 0.889 V, a $J_{sc}$ of 24.8 mA cm$^{-2}$, and an FF of 71.9%.

The slightly higher PCE of PM6/PYF-T-o in this study compared to the reported literature should be attributed to the changed electron/hole interlayer. The best $J-V$ curves and statistical distributions of PCEs of 20 pieces of devices are shown in Figure 3b,c for the three systems. Compared to the other two, the apparent advantage of PM6/2BTP-2F-T is its high $J_{sc}$ and FF. The tendency for the $V_{oc}$ variation has mainly resulted from their differed energy bandgap rather than the energy loss, which we will discuss in the following section. As seen from the external quantum efficiency (EQE) curves of the best devices (Figure 3d), the disparity in $J_{sc}$ for the three devices is mainly in the range of 600–800 nm, which is in the absorption range of the acceptor. Hence, the higher $J_{sc}$ of PM6/2BTP-2F-T should be a joint contribution to its higher absorption coefficient and the efficient utilization of the absorbed photons. Noteworthy, the calculated $J_{sc}$ from EQE is in good agreement with that measured from the $J-V$ tests within an error of 5%, and the best photovoltaic parameters are summarized in Table 1.

The efficient utilization of the absorbed photons and high FF in PM6/2BTP-2F-T was convinced by the photo-physics characterization. The photocurrent density ($J_{ph}$, the current density difference under illumination and in the dark) dependent on the effective voltage ($V_{eff}$, the voltage difference between the voltage at $J_{sc} = 0$ and applied bias) (Figure 3e) was applied to evaluate the efficiency of charge generation and collection. At a sufficiently high $V_{eff}$, all excitons are hypothesized to separate into free charges and the photo-generated current reaches the maximum saturation $J_{sat}$, and the exciton dissociation ($\eta_{diss}$) and charge collection efficiency ($\eta_{coll}$) could be calculated from the following functions: $\eta_{diss} = J_{sat} / J_{ph}$ (under short-circuit condition) and $\eta_{coll} = J_{sat} / J_{ph}$ (under maximum power output condition). The $\eta_{diss}$ is calculated to be 97.43% and 99.45%, and 96.94% for the devices based on PM6/monomer, PM6/2BTP-2F-T, and PM6/PYF-T-o, respectively, and their corresponding $\eta_{coll}$ is 85.01%, 91.23%, and 83.15%, respectively. The highest value of $\eta_{diss}$ and $\eta_{coll}$ demonstrates that the 2BTP-2F-T-based device wins the most efficient charge generation and the best charge collection compared to those of monomer- and polymer-based devices.

The power index $\alpha$ (fitted from the function of $J \propto (P_{light})^\alpha$; $P_{light}$ represents the incident light intensity) and the ideal factor $n$ (fitted from the function of $V_{oc} \propto n kT/q \ln(P_{light})$; $k$ is Boltzmann constant, $T$ is the temperature in Kelvin, and $q$ is the elementary charge) were applied to estimate the recombination mechanism for the three devices. For the device based on PM6/2BTP-2F-T, $\alpha$ is equal to 1 (Figure S5, Supporting Information), indicating that PM6/2BTP-2F-T is almost free from bimolecular recombination, and the other two systems also show negligible bimolecular recombination with an $\alpha$ value of 0.99. What is more, the lowest $n$ value of PM6/2BTP-2F-T (Figure 3f, $n = 1.15$ for PM6/monomer, 1.08 for PM6/2BTP-2F-T, and 1.26 for PM6/PYF-T-o), demonstrates that the device based on PM6/2BTP-2F-T effectively inhibits the trap-assisted recombination. The decreased $n$ value is probably due to its smallest energetic disorder as deduced from its smallest Urbach energy ($E_U$) value, which is described in the following parts. The photon-physical result suggested that the N-π-N type oligomeric acceptor 2BTP-2F-T could effectively enhance the efficiency of charge generation and charge collection while suppressing trap-assisted recombination simultaneously. Thus, higher $J_{sc}$ and FF are obtained.

### 2.3. Molecular Packing, Morphology, and Charge Properties

The molecular packing and morphology in the optimized blend were further used to confirm the efficient charge generation and collection in the PM6/2BTP-2F-T blends. As shown in 2D GI-WAXS (Figure S6, Supporting Information) and corresponding 1D curves (Figure 4a), all the blends reserved a preferential face-on packing mode of pristine films; in view of the lamellar peak of PM6, the (010) peak in OOP direction is a combination of the donor and acceptor; therefore, the (100) peak in IP direction could be applied to separate their crystalline ability in the blends. As deduced from the CCL of (100) peak, PM6 exhibited decreased packing ability for PM6/monomer (112 Å), PM6/2BTP-2F-T (95 Å), and PM6/PYF-T-o (93 Å), respectively. This trend is totally opposite to the change in their miscibility, which is probably due to the different diffusion speeds of their corresponding acceptors escaping from their mixed regions originating from their differed MW. Consequently, although PM6/2BTP-2F-T shows smaller miscibility ($\chi = 0.14$) than that of PM6/monomer ($\chi = 0.08$), however, it forms a more interpenetrating film than that of PM6/monomer, which could be intuitively seen in height atomic force microscopy (AFM) images (Figure 4b) and confirmed by its slightly decreased root-mean-square (RMS) surface roughness value (RMS = 1.38 nm). The opposite is also true for the PM6/2BTP-2F-T and PM6/PYF-T-o.

The above results indicate that PM6/2BTP-2F-T blends present an easier morphology control than PM6/monomer and PM6/PYF-T-o. Furthermore, as shown in the transmis-
Figure 4. Morphology properties and charge properties: a) 1D GIWAXS curves of optimized blends; b) AFM height images and TEM images for the optimized blends; c–e) charge transport, TPV, and TPC curves for the optimized blend.

2.4. Energy Loss and Stability

2.4.1. Energy Loss

Energy loss ($\Delta E$) measurements were carried out to elucidate the potential of the N-$\pi$-N type oligomeric acceptors. The total $\Delta E$ can be split into three parts: 1) $\Delta E_1$ represents radiative recombination loss above the bandgap, which is based on the Shockley–Queisser (SQ) theory limits. $\Delta E_1$ is directly decided by the energy bandgap. 2) $\Delta E_2$ represents radiative recombination loss below the bandgap; 3) $\Delta E_3$ represents non-radiative recombination loss below the bandgap, which could be quantified by external quantum electroluminescence efficiency (EQE_EL).

As shown in Figure 5a and Table S5, Supporting Information, the $\Delta E$ for the three devices has a very small difference within a value of 0.01–0.02 eV, and the value of PM6/2BTP-2F-T (0.53 eV) is in between that of PM6/monomer (0.54 eV) and PM6/PYF-T-o (0.52 eV). The energy band-gap ($E_g$) is calculated from the precise measurement of the EQE, with a value of 1.48, 1.45, and 1.42 eV for monomer, 2BTP-2F-T, and PYF-T-o blend (Figure S7, Supporting Information). Hence, their Voc differences mainly result from their different $E_g$ rather than their $\Delta E$. Interestingly, there is a delicate trade-off between $\Delta E_2$ and $\Delta E_3$ in the three devices. That is, the PM6/monomer-based device obtains...
the largest $\Delta E_2$ ($0.093$ eV), while the smallest $\Delta E_3$ ($0.184$ eV) and PM6/PyF-T-o exhibits exactly the reverse result with an $\Delta E_2$ of $0.049$ eV and an $\Delta E_3$ of $0.208$ eV. As for PM6/2BTP-2F-T, both the values of $\Delta E_2$ ($0.068$ eV) and $\Delta E_3$ ($0.199$ eV) are in between. This is probably because their small energy offset between $E_g$ and energy of charge transfer (CT) states estimated from normalized fourier-transform photocurrent spectroscopy external quantum efficiency (FTPS-EQE) curves and electroluminescence (EL) spectra of devices (as shown in Figure S8 and Table S5, Supporting Information) creates an equilibrium state between the CT states and acceptor singlet, which is believed to enhance the efficiency of radiative decay, while suppressing its non-radiative decay.[30] Hence, the previous analysis indicates there is negligible difference in the $\Delta E$ for the three systems, and their disparity is due to their slightly different energy levels of themselves and complex energy alignments due to the morphology optimization. Further applying a more rigid molecular backbone or monomer, and proper HOMO energy level of the donor would be a potential approach to decrease the $\Delta E$ of N-x-N oligomeric type OSCs.

The low degree of energetic disorder at interfaces in the active layers enables free charge separation to outcompete trap-assisted non-radiative recombination of the CT state, gaining excellent device performance with small $\Delta E$.[28,29,31] Hence, Urbach energy ($E_U$) is obtained through an exponential fit to the FTPS-EQE spectra to evaluate the degree of energetic disorder (Figure 5b and Figure S9, Supporting Information).[32] The PM6/2BTP-2F-T obtains the smallest $E_U$ with a value of $22.74$ meV, compared to that of PM6/monomer ($E_U = 23.92$ meV) and PM6/PyF-T-o ($E_U = 24.79$ meV), indicating the smallest width of the tail of the electronic density of disorder (DOS). It should be mentioned that all the $E_U$ for the three devices is comparable to the thermal energy $k_B T$ (25.8 meV) at room temperature. The small $E_U$ for the three devices guarantees their minimal loss of $\Delta E_2 + \Delta E_3$ (<0.28 eV). The smallest $E_U$ of PM6/2BTP-2F-T enables less trap-assisted recombination of the charge, and a much longer charge lifetime, benefiting the improvements of FF and $J_{sc}$.

Hence, PM6/2BTP-2F-T exhibits more efficient charge generation, higher absorption coefficient, and an optimal morphology with a smaller energetic disorder, leading to ultimately a high $J_{sc}$. The much higher $J_{sc}$ with similar $\Delta E$ for PM6/2BTP-2F-T leads to a maximized ($V_{oc} \times J_{sc}$) in the three devices (Figure 5c), which is a bottleneck for the current photovoltaic materials design and device optimization. Together with excellent charge properties, it wins the champion PCE in the three systems. Furthermore, its small $E_U$, high tolerance of $\mu_e$ to the morphology, and easily tuned morphology indicate a potential of low $\Delta E$, efficient charge generation, and excellent charge transport properties. If a more matched polymer in energy level could be found, further improved device performances in OSCs would be expected based on N-x-N oligomeric acceptors.

2.4.2. Stability

The much higher MW of 2BTP-2F-T than monomer would lead to a lower relative diffusion ability and better stability, as proved in polymer acceptors.[15] The $T_{50}$ (sustained 80\% of the initial efficiency) of the three encapsulated devices to light soaking (100 mW cm$^{-2}$) under air conditions and thermal stability at 80 °C under a nitrogen atmosphere were investigated. Conventional devices with ITO/poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)/active layer/PNDIT-F3N/Ag structure were used in the light-soaking stability and thermal stability test. Figures 5d
and 5e show the trend of PCE decay over time in light-soaking stability and thermal stability test, respectively. The curves in Figure 5d were fitted by a double exponential decay function to obtain a relatively accurate $T_{90}$, while curves in Figure 5e were obtained by the b-spline algorithm.

Expectedly, the lifetime of $T_{90}$ to light soaking under air condition based on PM6/2BTP-2F-T device exhibits 7 times longer (443 h) than that of PM6/monomer (62 h); beyond belief, it also demonstrates 2.4 times longer than that of PM6/PYF-T-o (185 h). Besides the morphology factors, the higher optical chemical stability of the 2BTP-2F-T was also observed in a recently reported result measured under a nitrogen atmosphere.\textsuperscript{[13]} The lifetime of $T_{90}$ to heat treatment for the PM6/2BTP-2F-T is also much longer than that of PM6/monomer (54 h vs 20 h) and comparable to that of all-polymer OSCs PM6/PYF-T-o (57 h). Noteworthy, further prolonging the heating time, the PM6/2BTP-2F-T shows a slightly decreased rate of device attenuation than that of PM6/PYF-T-o (Figure 5e). The above results demonstrate that PM6/2BTP-2F-T possesses comparable thermal stability but better light-soaking stability than PM6/PYF-T-o, and is much better than PM6/monomer. This difference in stability was further convinced by the less varied morphology of PM6/2BTP-2F-T films under the heating and light-soaking process, as shown in Figure S10, Supporting Information.

### 3. Conclusion

In this article, we designed an N-$\pi$-N type oligomeric acceptor 2BTP-2F-T. Using its corresponding monomer and polymer as references, we profoundly demonstrated the advantages of 2BTP-2F-T, including a higher absorption coefficient, improved electron mobility with less dependence on molecular packing, and easier morphology control with polymer donors. Using PM6 as the donor, the PM6/2BTP-2F-T blend exhibits superior advantages compared with their monomeric and polymeric counterparts: 1) More favorable morphology, including the smallest energetic disorder, moderate $\pi$-$\pi$ stacking, and appropriate interpenetrating network; 2) Improved charge properties, including higher hole/electron mobility, longer charge lifetime, and faster charge extraction; 3) Efficient charge generation with similar energy loss; 4) Better light-soaking stability and comparable thermal stability to its polymeric counterparts. The high efficiency of over 18% in OSCs, together with its improved stability, suggests a bright future for the N-$\pi$-N oligomeric acceptor type OSCs.

### 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 from the corresponding author upon reasonable request.

### Keywords

energy loss, N-$\pi$-N type, oligomeric acceptor, organic solar cells, thermal stability

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