

Supplementary Information

Bithiophene Imide-Based Polymer Donor for Alloy-like Ternary Organic Solar Cells with Over 20.5% Efficiency and Enhanced Stability

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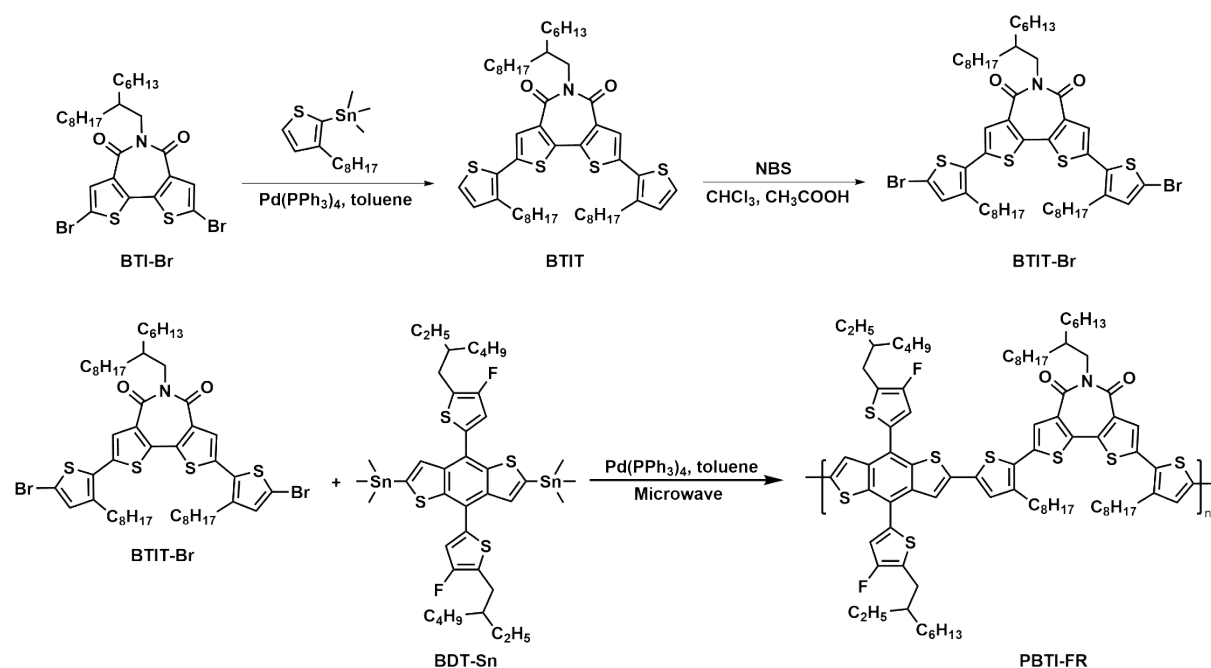
[†]These authors contribute equally.

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1. Supplementary Notes

PM6, L8-BO, BTP-eC9, Y6 and D18-Cl were purchased from Solarmer Energy Inc. PEDOT:PSS was purchased from eFlexPV. All chemicals and agents were commercially available and were used without further purification unless otherwise stated. Anhydrous toluene was distilled from Na/benzophenone under argon flow. ^1H and ^{13}C NMR spectra of monomers and their precursors were recorded on Bruker Ascend 400 MHz spectrometers. Polymer molecular weights were measured by Agilent 1260 Infinity II high-temperature gel permeation chromatography (GPC) System, using 1,2,4-trichlorobenzene as the eluent at 160 °C, relative to polystyrene standards. Room temperature UV-vis absorption spectra of polymer solutions and films were collected on a Shimadzu UV-3600 UV-VIS-NIR spectrophotometer. Cyclic voltammetry measurements were performed on a CHI660A electrochemical workstation with 0.1 M tetra(*n*-butyl)ammoniumhexafluorophosphate in acetonitrile as the supporting electrolyte, a platinum disk as the working electrode, a platinum wire as the counter electrode, and Ag/AgCl as the reference electrode. Ferrocene/ferrocenium (Fc/Fc^+) redox couple was used as the external reference for all measurements.



Scheme 1. Synthetic routes to monomers and polymers. The dibrominated bithiophene imide (BTI-Br) was synthesized following the procedure outlined in the reference.¹

Synthesis of 5-(2-hexyldecyl)-2,8-bis(3-octylthiophen-2-yl)-4H-dithieno[3,2-c:2',3'-e]azepine-4,6(5H)-dione, (BTIT). BTI-Br (620 mg, 1 mmol, 1 eq), trimethyl(3-octylthiophen-2-yl)stannane (1.08 g, 3.01 mmol, 3 eq), PdCl₂(PPh₃)₂ (10%) and toluene (10 mL) were added to a 25 mL air-free flask, and the reaction mixture was then stirred at 120 °C for 12 h. After cooling to room temperature, the reactant was poured into a potassium fluoride solution (1 g mL⁻¹, aq) and stirred for 24 h. Then the mixture was extracted with 20 mL CH₂Cl₂ for three times. The combined organic layer was dried over anhydrous Na₂SO₄ and then filtrated. The obtained organic solution was evaporated under reduced pressure to afford an orange-red liquid, which was purified by column chromatography over silica gel with CH₂Cl₂/petroleum ether (1:5) as eluent to afford an orange-red solid (426 mg, 50%). ¹H NMR (400 MHz, CD₂Cl₂) δ 7.70 (s, 2H), 7.29 (d, *J* = 5.2 Hz, 2H), 7.00 (d, *J* = 5.2 Hz, 2H), 4.22 (d, *J* = 7.2 Hz, 2H), 2.86 – 2.78 (m, 4H), 1.90 (s, 1H), 1.66 (dd, *J* = 15.1, 7.6 Hz, 4H), 1.39 - 1.24 (m, 44H), 0.85 (dd, *J* = 6.9, 5.0 Hz, 12H). ¹³C NMR (101 MHz, CDCl₃) δ 162.25, 141.68, 136.28, 135.44, 133.36, 130.98, 130.51, 128.78, 125.55, 49.79, 36.68, 32.13, 32.09, 30.72, 30.52 - 30.36, 30.17, 29.93 - 29.36, 26.69, 22.88, 14.32. C₅₀H₇₃NO₂S₄, Calcd: 847.4524; Found: 848.45984 [M+H]⁺.

Synthesis of 2,8-bis(5-bromo-3-octylthiophen-2-yl)-5-(2-hexyldecyl)-4H-dithieno[3,2-c:2',3'-e]azepine-4,6(5H)-dione, (BTIT-Br). BTIT (400 mg, 0.047 mmol, 1 eq) was dissolved in a mixed solvent of CHCl₃/CH₃COOH (4:1, 40 mL), then the solution NBS (184.62 mg, 1.04 mmol, 2.2 equiv.) was added. The reaction was stirred at room temperature for ~6 h and quenched with 5 mL water. The mixture was extracted by CH₂Cl₂, and then the combined organic layer was dried over anhydrous Na₂SO₄. The solvent was evaporated to afford the crude products, which were then purified by column chromatography over silica gel with CH₂Cl₂/ petroleum ether (1:5) as eluent to afford an orange-red solid (427 mg, 90 %). ¹H NMR (400 MHz, CD₂Cl₂) δ 7.65 (s, 2H), 6.97 (s, 2H), 4.21 (d, *J* = 7.3 Hz, 2H), 2.80 - 2.71 (m, 4H), 1.89 (s, 1H), 1.63 (dd, *J* = 15.3, 7.6 Hz, 6H), 1.41 - 1.21 (m, 44H), 0.86 (t, *J* = 6.2 Hz, 12H). ¹³C

NMR (101 MHz, CDCl₃) δ 161.97, 142.29, 136.22, 134.08, 133.51, 133.15, 131.41, 130.10, 112.70, 49.80, 36.65, 32.08, 32.05, 30.75 - 30.68, 30.45, 29.89, 29.75 - 29.25, 26.66, 22.90, 22.88, 14.32. C₅₀H₇₁Br₂NO₂S₄, Calcd: 1003.2734; Found: 1004.28040 [M+H]⁺.

Synthesis of polymer PBTI-FR

To a 10 mL microwave reaction tube equipped with a stirring bar, a distannylated monomer of BDT-Sn (0.05 mmol), dibrominated monomer (**BTIT-Br**) (0.05 mmol) and Pd(PPh₃)₄ (1%). The tube was purged with argon three times, and toluene (5 mL) was added via syringe. The tube was sealed and transferred to a microwave reactor to be heated to 140 °C, reacting for 3 h. After cooling to 80 °C, the reaction solution was poured into 80 mL methanol. The polymer was collected by filtration and dried under reduced pressure to afford a deep-colored solid. Then, the precipitated solid was extracted via sequential Soxhlet extraction with methanol, acetone, hexane, dichloromethane, and chloroform. The main polymer component was extracted using chloroform as the final solvent. The corresponding solutions were then concentrated to ~20 mL and poured into 100 mL methanol under vigorous stirring. The polymer was collected by filtration and dried under reduced pressure to give a solid as the product polymer (75%).

Device fabrication

Conventional-structured binary and ternary OSCs were fabricated using the following configuration: ITO/PEDOT:PSS/Active Layer/PNDIT-F3N/Ag. The glass substrate coated with ITO film was wiped clean with detergent-infused water, then cleaned through a series of steps including deionized water, acetone and isopropanol by using ultrasound cleaning instrument for 15 min. Then, it was dried in a vacuum system and subjected to UV-ozone treatment for 15 min. The PEDOT: PSS was spin-coated at a speed of 5000 rpm for 20 s to deposit onto the UV-ozone-treated ITOs and then annealed at 150°C for 10 min. The ITO substrate was then transferred into a nitrogen-filled glove box (O₂ <10 ppm, H₂O <10 ppm) for subsequent procedures. For

binary and ternary OSCs treated with DIO (0.25 vol.%), the donor and acceptor ratio of the active layer was maintained at 1:1.25, and the total concentration of all systems remained constant at 16mg mL⁻¹ in CF solvent, the spin coating speed was 4000 rpm for 30 s. All solutions were fully dissolved at 45°C and deposited onto the PEDOT: PSS layer, followed by thermal annealing at 100°C for 10 min. The PNDIT-F3N was dissolved in methanol at a concentration of 0.5 mg mL⁻¹ with 0.5 vol.% acetic acid and was spin-coated onto the active layer at a speed of 2000 rpm for 30 s. Finally, these semi-finished cells were transferred to a thermal evaporation chamber with an essential pressure of 1×10⁻⁶ Pa, and a layer of Ag with a thickness of 110 nm was successively coated onto the electron transport layer using thermal evaporation.

Calculation of the surface free energy and miscibility

According to the Harmonic Mean formula, the surface energy (γ) of the film can be obtained by utilizing the different contact angles of two solvents with different polarities on the film surface. Based on the measured surface tension, Neumann's formula can be employed to calculate the interfacial energy (γ^{X-Y}) between the blended films. Subsequently, Young's formula can be applied to compute the wetting coefficient (ω), thereby deducing the distribution of the third component at the donor-acceptor interface. Alternatively, based on the surface tension of the film, the interaction parameter (χ) between different active materials can be derived using the Flory–Huggins theory.

Harmonic Mean Formula:

$$\gamma_1(1 + \cos \theta_1) = \frac{4\gamma_1^d\gamma_s^d}{\gamma_1^d + \gamma_s^d} + \frac{4\gamma_1^p\gamma_s^p}{\gamma_1^p + \gamma_s^p}$$

$$\gamma_2(1 + \cos \theta_2) = \frac{4\gamma_2^d\gamma_s^d}{\gamma_2^d + \gamma_s^d} + \frac{4\gamma_2^p\gamma_s^p}{\gamma_2^p + \gamma_s^p}$$

$$\gamma = \gamma_s^d + \gamma_s^p$$

In this context, γ_s^d represents the dispersive component, and γ_s^p represents the polar

component of the surface energy. θ_1 and θ_2 denote the contact angles of ultrapure water and glycerol, respectively, when dropped onto the film surface. According to the literature, the surface energy of water (γ_1) is 72.8 mJ m⁻², with γ_1^d (the dispersive part) being 21.8 mJ m⁻² and γ_1^p (the polar part) being 51 mJ m⁻². For glycerol, the surface energy (γ_1) is 48.0 mJ m⁻², with γ_2^d and γ_2^p being 29.0 mJ m⁻² and 19 mJ m⁻², respectively.

The Neumann's formula:

$$\gamma_{X-Y} = \gamma_X + \gamma_Y - 2\sqrt{\gamma_X \gamma_Y} e^{[-\beta(\gamma_X - \gamma_Y)]^2}$$

γ_{X-Y} is the interfacial surface energy between materials X and Y, γ_X and γ_Y are the surface energies of the films made from materials X and Y, respectively. β is a coefficient with a value of 0.000115 m⁴ MJ⁻².

The Flory–Huggins interaction parameter:

$$\chi^{X-Y} = (\sqrt{\gamma^X} - \sqrt{\gamma^Y})^2$$

Device characterization

The J - V characteristics of all devices were evaluated under simulated AM1.5G irradiation (100 mW cm⁻²) using an SS-F5-3A Solar Simulator (Enli Technology, Inc.) equipped with an Xe lamp. The white light source utilized an Xe lamp with an AM1.5G filter, while the light irradiance was regulated by the National Renewable Energy Laboratory (NREL)-calibrated Si solar cell with a KG-5 filter. The EQE curves were evaluated with a QE-R3011 measurement system (Enli Technology, Inc.).

DFT calculation

All density functional theory (DFT) calculations were carried out using Gaussian 16 package. The optimized ground-state geometry and the single point properties at the ground state were calculated by the DFT method at the B3LYP-D3/6-31G** level. The S1 geometry was optimized by time-dependent DFT (TD-DFT) at the B3LYP-D3/6-

31G** level. The decomposition of reorganization energies calculations for S0 and S1 of the molecules have been performed.

MD simulation

The partial charge of PBTI-FR, PM6, and L8-BO molecules was calculated using Gaussian 16 code, and the 6-311g(d,p) basis functions were applied.² The GAFF force field³ and Auxiliary Tools of Force Field (AuToFF) were used to parametrize all atoms, such as the bond parameters, angle parameters, dihedral angles, etc.

The interaction energy between molecules of different components was studied using molecular dynamics (MD) simulation. In system 1, 40 PBTI-FR and 130 L8-BO molecules were randomly added into a $12 \times 12 \times 12$ nm³ simulation box; In system 2, 40 PM6 and 108 L8-BO molecules were randomly added into a $12 \times 12 \times 12$ nm³ simulation box; In system 3, 4 PBTI-FR, 40 PM6, and 108 L8-BO molecules were randomly added into a $12 \times 12 \times 12$ nm³ simulation box. To better obtain a stable accumulation structure, a high-temperature equilibrium simulation of 15 ns is performed at 373.15 K, and then the temperature is lowered to 298.15 K within 3 ns simulation time. Finally, the dynamic equilibrium of 50 ns is performed at 298.15 K.

The MD simulations were performed in the GROMACS 2021 software package.⁴⁻⁶ The steepest descent method was applied to minimize the initial energy for each system with a force tolerance of 1 kJ (mol⁻¹ nm⁻¹) and a maximum step size of 0.001 ps before MD calculations. In all the three directions, periodic boundary conditions were imposed. Leapfrog algorithm was used to integrate the Newtonian equation of motion.⁷ In NPT simulations, the pressure was maintained at 1 bar by the Parrinello-Rahman barostat in an isotropic manner,⁸ and the temperature was maintained by the V-rescale thermostat. The Particle-Mesh-Ewald (PME) with a fourth-order interpolation was used to evaluate the electrostatic interactions,⁹ and a cutoff of 1.2 nm was employed to calculate the short-range van der Waals interactions.

Empirical relationship between the V_{oc} and FF

The correlation between the V_{oc} and the maximum fill factor (FF_{max}) in a specific

device can be characterized as follows:

$$FF_{max} = \frac{\gamma_{oc} - \ln(\gamma_{oc} + 0.72)}{\gamma_{oc} + 1}$$

$$\gamma_{oc} = \frac{qV_{oc}}{nkT}$$

Where q is the elementary charge, n is the diode ideality factor, k is the Boltzmann constant, and T is the temperature. It has been observed that the FF achieved in high-performance organic solar cells (OSCs) typically falls within the range of FF_{max} (0.10 $\pm$ 0.04), as documented in prior studies.

Energy loss analysis

According to the Shockley-Queisser (SQ) limit theory, energy losses are typically categorized into three parts: radiative losses above the optical bandgap (ΔE_1), radiative losses below the optical bandgap (ΔE_2), and non-radiative recombination losses (ΔE_3). The quantitative expressions for these energy losses can be formulated as:

$$\begin{aligned} E_{loss} &= E_g - qV_{oc} \\ &= (E_g - qV_{oc}^{SQ}) + (q\Delta V_{oc}^{rad, below\ gap}) + (q\Delta V_{oc}^{non-rad}) \\ &= \Delta E_1 + \Delta E_2 + \Delta E_3 \end{aligned}$$

V_{oc}^{SQ} is the maximum open-circuit voltage calculated according to the SQ theory, $\Delta V_{oc}^{rad\ below\ gap}$ denotes the radiative recombination open-circuit voltage loss below the optical bandgap, and $\Delta V_{oc}^{non-rad}$ signifies the non-radiative recombination open-circuit voltage loss.

The SQ limit open-circuit voltage can be expressed as:

$$V_{oc}^{SQ} = \frac{kT}{q} \ln \left(\frac{J_{sc}}{J_0^{SQ}} + 1 \right) = \frac{kT}{q} \ln \left(\frac{q \cdot \int_0^\infty EQE_{PV}(E) \cdot \Phi_{AM1.5}(E) dE}{q \cdot \int_{E_{gap}}^\infty \Phi_{BB}(E) dE} \right) + 1$$

The voltage loss below the optical bandgap ($\Delta V_{oc}^{rad \text{ below gap}}$) can be calculated using Equation below:

$$\Delta V_{oc}^{rad \text{ below gap}} = V_{oc}^{SQ} - V_{oc}^{rad}$$

The non-radiative voltage loss ($V_{oc}^{non \text{ rad}}$) can be calculated using the following formula:

$$\Delta V_{oc}^{non \text{ rad}} = V_{oc}^{rad} - V_{oc}$$

ΔV_{oc}^{rad} is the open-circuit voltage due to radiative recombination, and V_{oc} is the open-circuit voltage measured through I - V testing of the solar cell.

The formula can also be expressed as:

$$V_{oc}^{non \text{ rad}} = -\frac{kT}{q} \ln (EQE_{EL})$$

Morphological characterization

The film morphology of active layers was performed using an Atomic Force Microscopy (AFM) technique, employing a Dimension Icon Scanning Probe Microscope (Asylum Research, MFP-3D-Stand Alone) in tapping mode. TEM images were captured from a Hitachi HT7700 microscope running at 100 kV and outfitted with an AMF-5016 charge-coupled device (CCD) detector. GIWAXS measurements. GIWAXS measurements were performed at the PLS-II 9A U-SAXS (Ultra-Small-Angle X-ray Scattering) beamline of the Pohang Accelerator Laboratory in Republic of Korea.

SCLC measurement

The charge carrier mobilities were characterized by employing the SCLC technique. The structure of hole-only devices consisted of ITO/PEDOT: PSS/active layer/Au, while for electron-only devices, the architecture comprised ITO/ZnO/active layer/F3N/Ag. The mobilities were derived using the MOTT-Gurney formula, given by

$$J = \frac{9}{8} \epsilon_r \epsilon_0 \mu \frac{V^2}{L^3}$$
, Where J represents the current density, ϵ_r denotes the relative dielectric constant of photoactive material. ϵ_0 refers to the permittivity of space, μ represents the mobility of either holes or electrons, and d is the thickness of the active layer. V denotes the internal voltage within the device, and $V = V_{\text{appl}} - V_{\text{bi}}$, where V_{appl} signifies the applied voltage and V_{bi} represents the built-in voltage derived from the relative difference in work function between the two electrodes (in the electron-only and hole-only devices, the V_{bi} values can be disregarded).

Transient photovoltage (TPV) and transient photocurrent (TPC) measurements

During TPV evaluations, devices were subjected to ambient illumination via a Quartz Tungsten-Halogen Lamp, ensuring a light intensity comparable to that of operational devices, thereby aligning the device's voltage with the V_{oc} under solar simulation. The photo-excitation was induced using 8 ns pulsed laser emissions from the NLD520 system by Oriental Spectra, with the excitation wavelength precisely set at 518 nm and a narrow spectral bandwidth of 3 nm. The TPV responses were captured under open-circuit conditions using a high-resolution digital oscilloscope. Concurrently, TPC measurements were conducted under short-circuit conditions, employing identical excitation parameters but in the absence of background illumination.

FTPS-EQE and EL-EQE measurements

The FTPS-EQE measurements were conducted using the integrated system (PECT-600, Enlitech), which facilitated the amplification and modulation of the

photocurrent by a lock-in instrument. For the EL-EQE assessments, external voltage/current sources were applied across the devices through the (REPS-Pro, Enlitech) setup. All devices were meticulously prepared in accordance with the optimal fabrication conditions for EL-EQE measurements.

2. Supplementary Figures

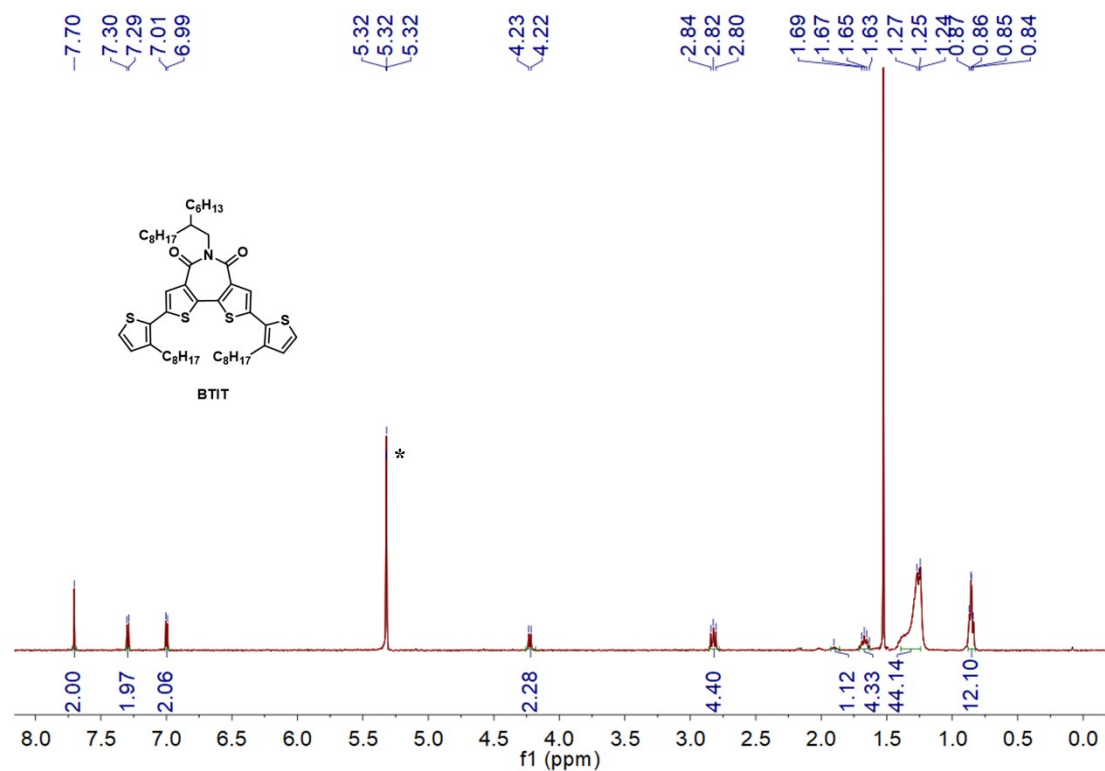


Fig. S1 ¹H NMR spectrum of compound BTIT (r.t., in CD₂Cl₂).

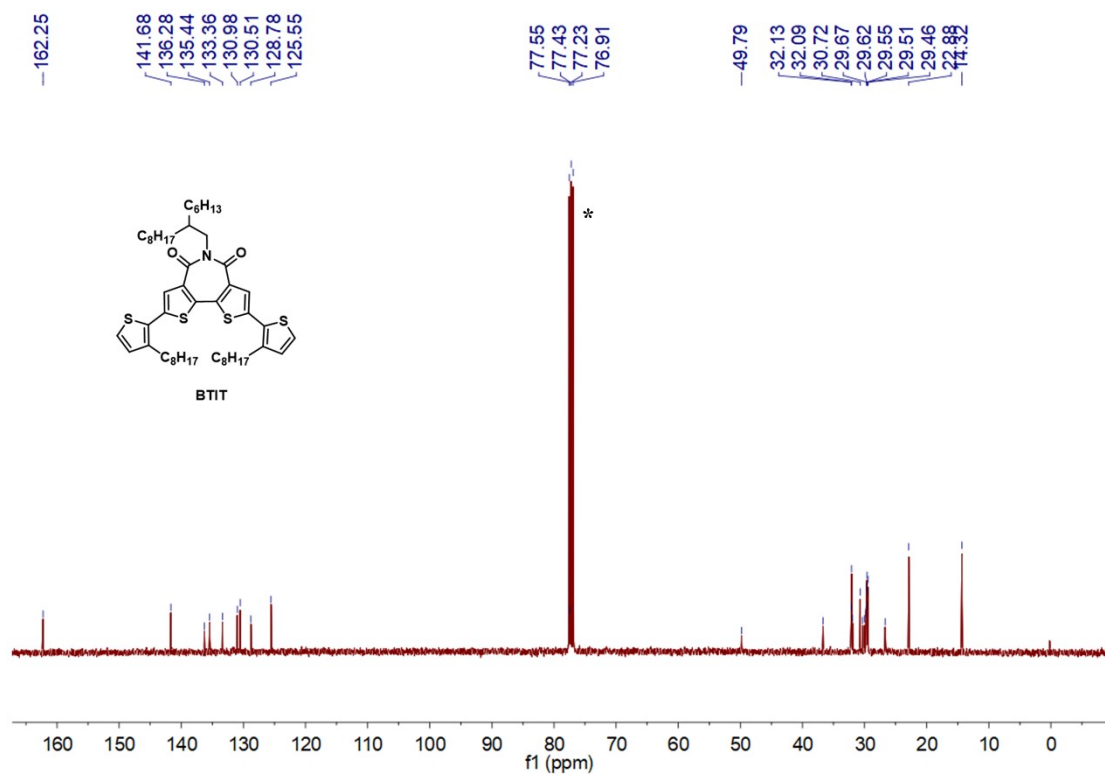


Fig. S2 ¹³C NMR spectrum of compound BTIT (r.t., in CDCl₃).

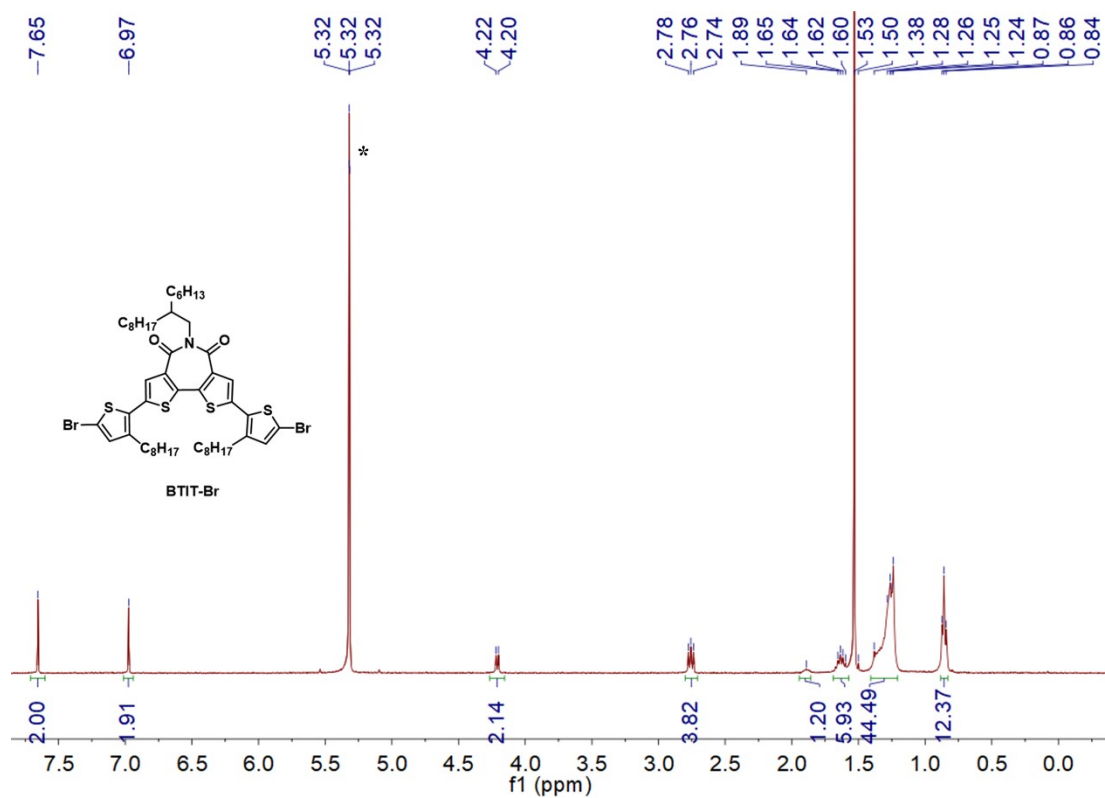


Fig. S3 ¹H NMR spectrum of compound BTIT-Br (r.t., in CD₂Cl₂).

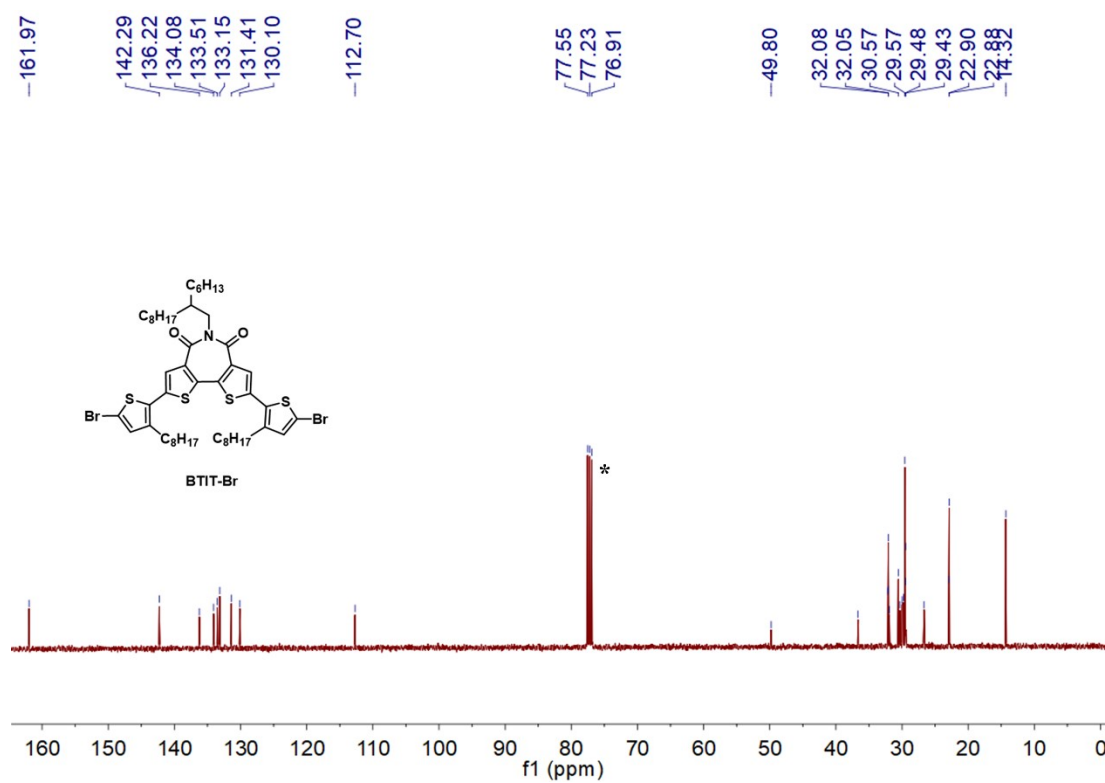


Fig. S4 ¹³C NMR spectrum of compound BTIT-Br (r.t., in CDCl₃).

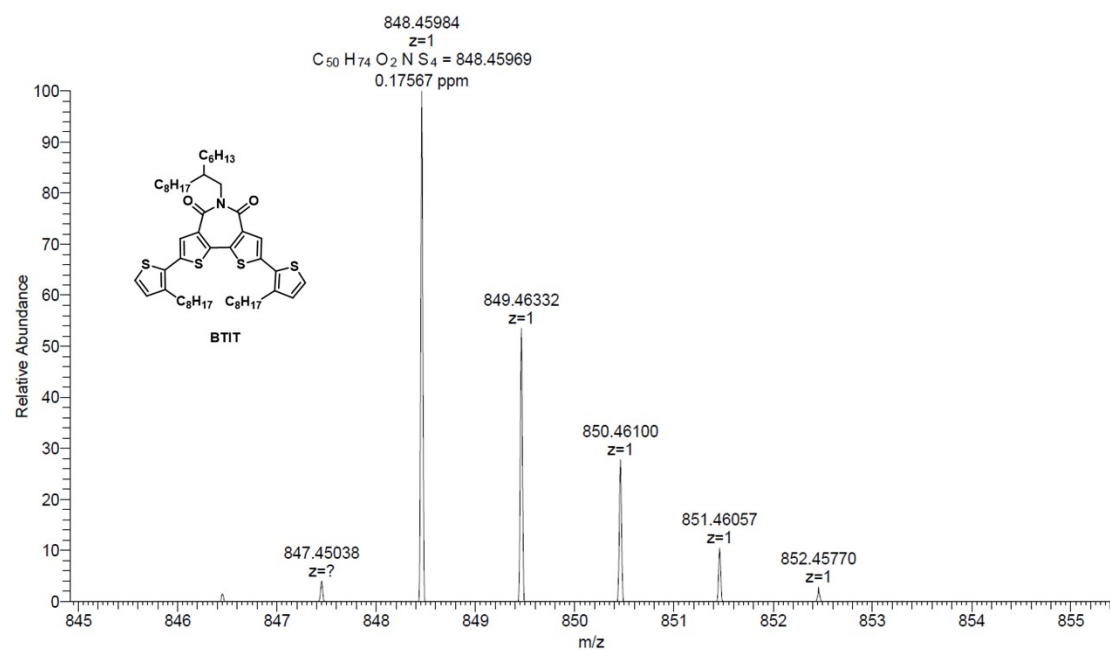


Fig. S5 HRMS of compound BTIT.

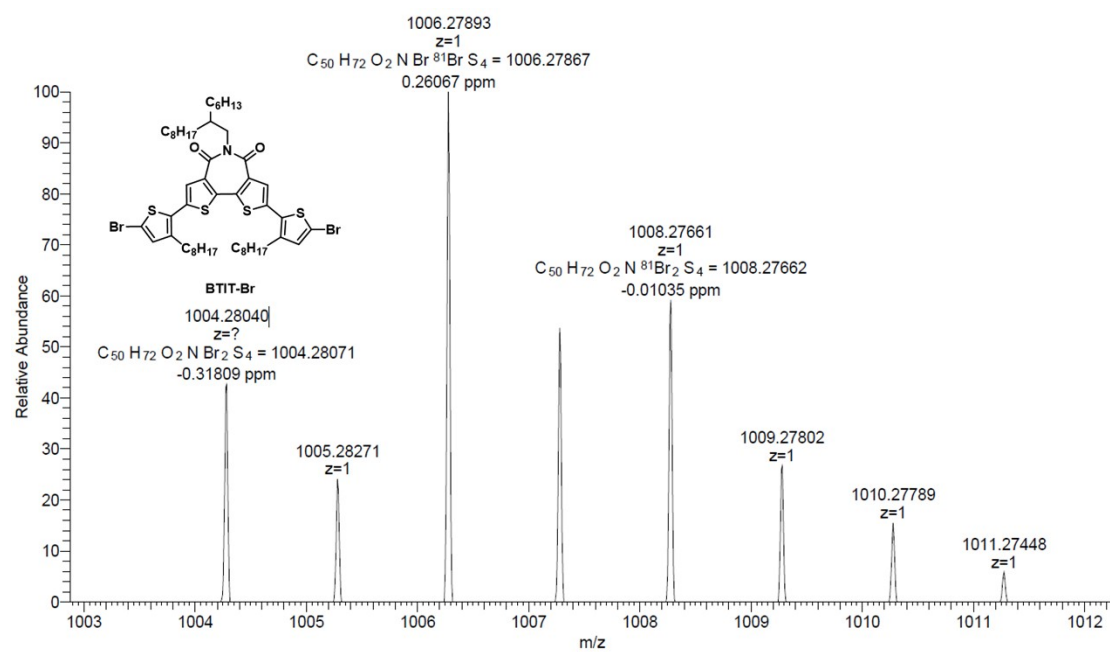
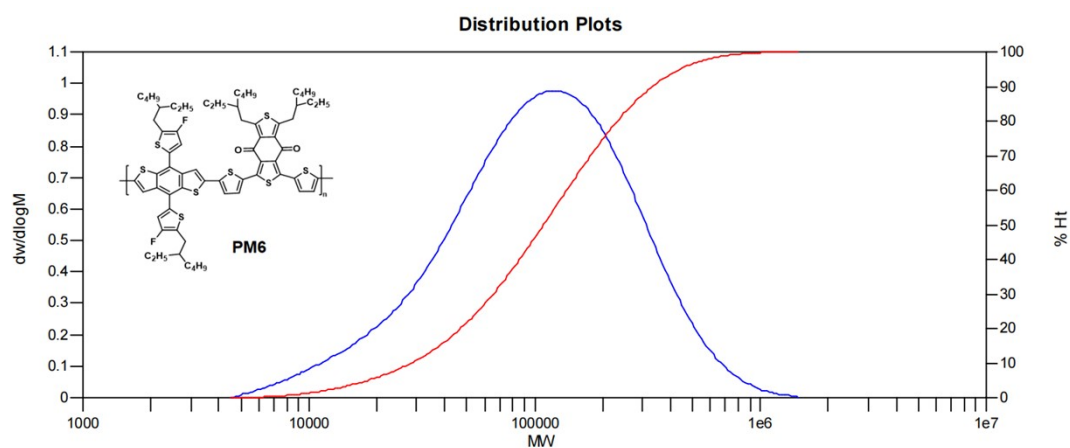


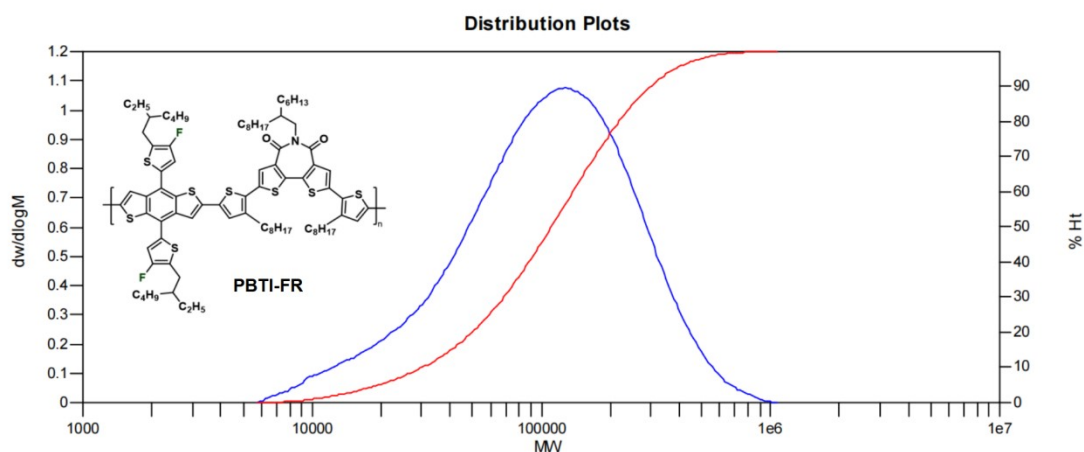
Fig. S6 HRMS of compound BTIT-Br.



MW Averages

Peak No	Mp	Mn	Mw	Mz	Mz+1	Mv	PD
1	121324	62021	152121	290595	460365	135481	2.45273

Fig. S7 GPC curve and molecular weight of polymer PM6.



MW Averages

Peak No	Mp	Mn	Mw	Mz	Mz+1	Mv	PD
1	127474	65204	142117	242222	351560	128987	2.17957

Fig. S8 GPC curve and molecular weight of polymer PBTI-FR.

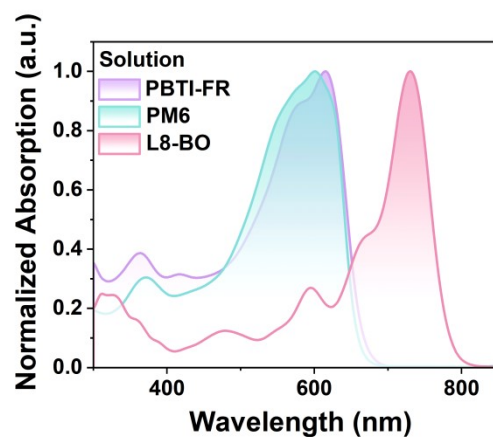


Fig. S9 Normalized UV-vis absorption spectra for PBTI-FR, PM6 and L8-BO in chloroform solution state.

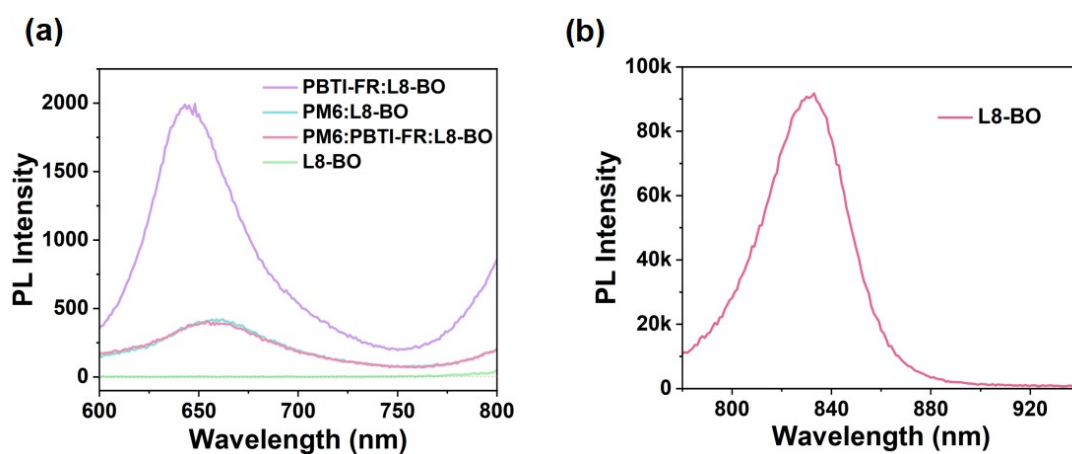


Fig. S10 (a) PL intensities of the blend films and L8-BO neat film under 560 nm excitation. (b) PL spectrum of the neat L8-BO film when excited at 750 nm.

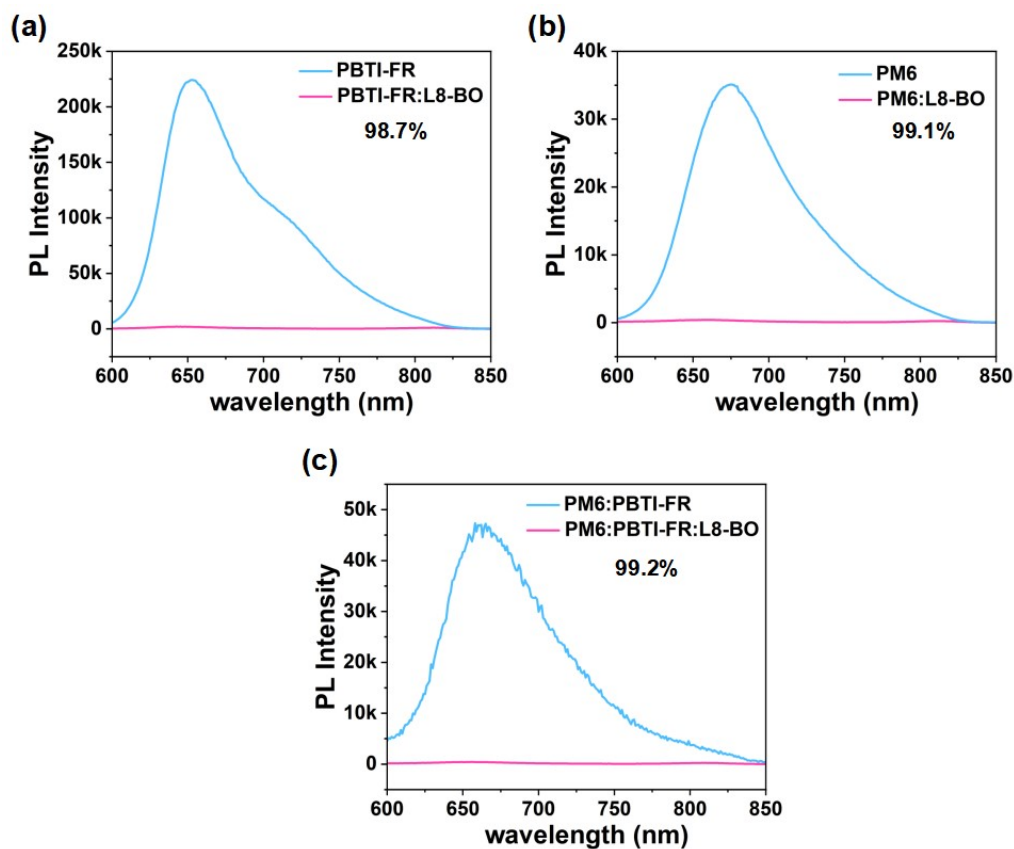


Fig. S11 The PL quenching efficiencies for (a) PBTI-FR:L8-BO, (b) PM6:L8-BO, and (c) PM6:PBTI-FR:L8-BO films, respectively, at 560 nm excitation.

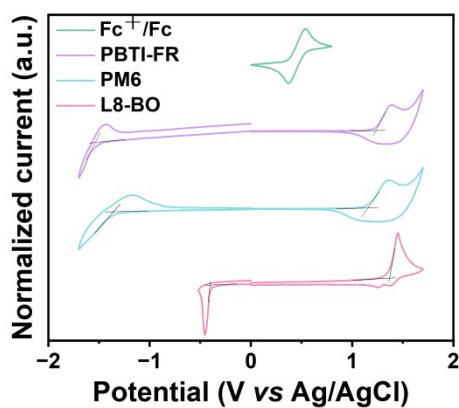


Fig. S12 Cyclic voltammogram curves of donor and acceptor films relative to Fc/Fc^+ as the reference.

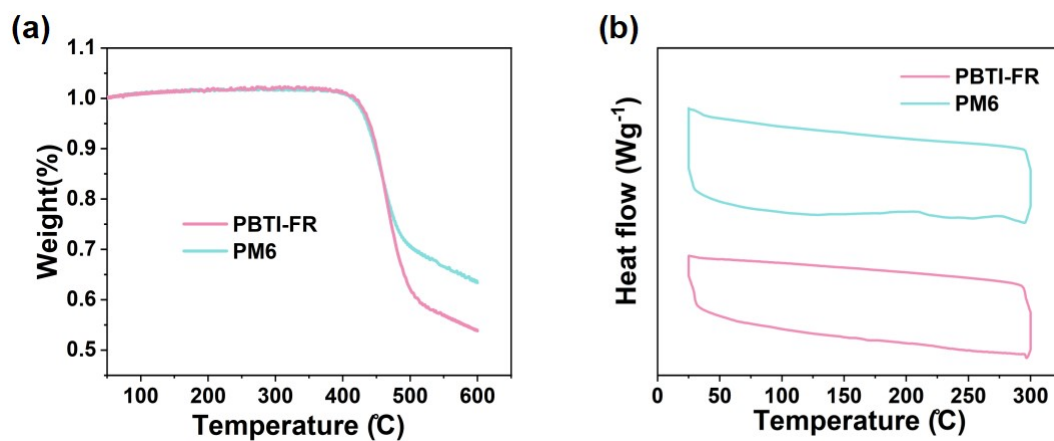


Fig. S13 (a) TGA and (b) DSC curves of PM6 and PBTI-FR.

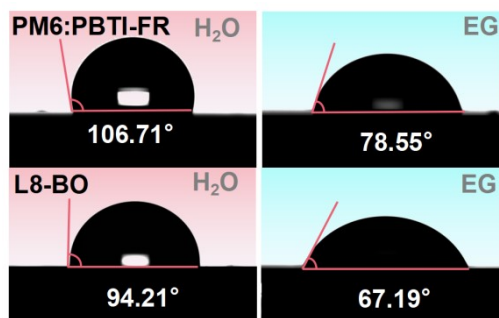


Fig. S14 Contact angle images of blend PM6:PBTI-FR film and neat L8-BO film.

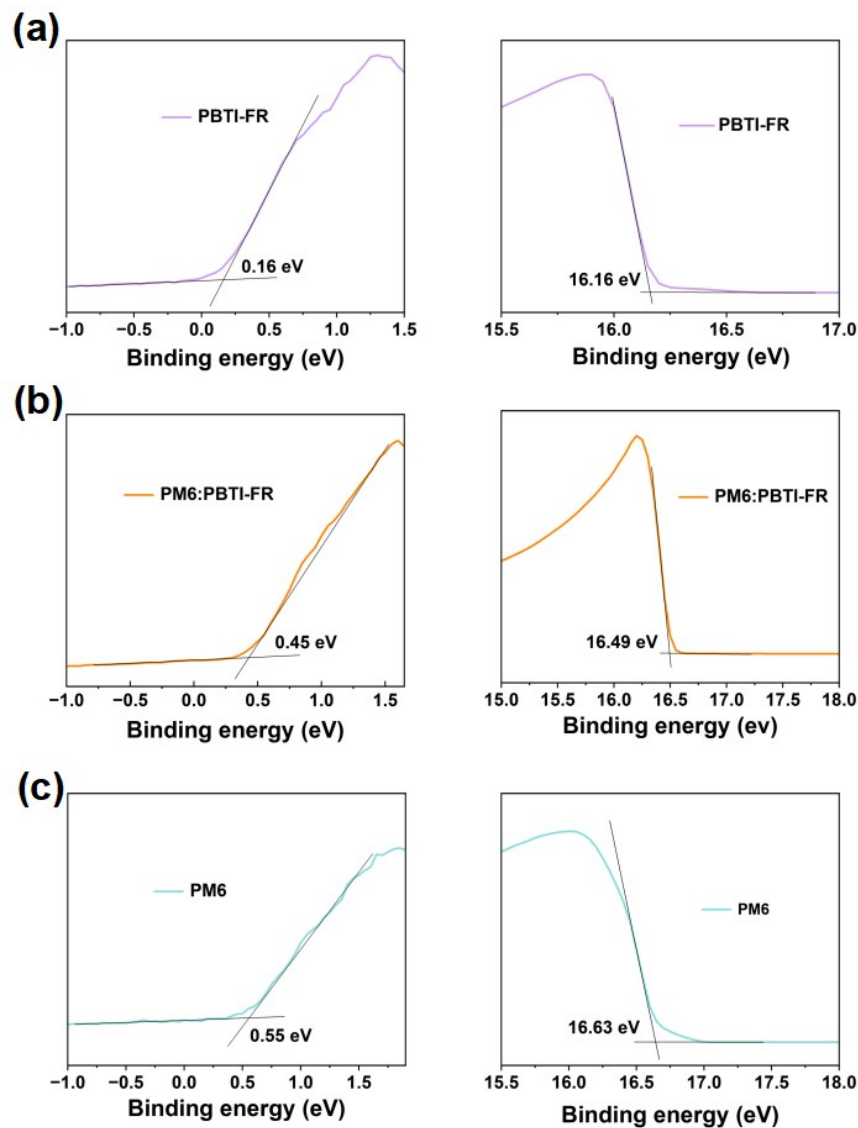


Fig. S15 UPS measurement for donor films UPS results of (a) PBTI-FR, (b) PM6:PBTI-FR and (c) PM6, which is obtained by the tangent extrapolation methods. The PM6:PBTI-FR blend film is fabricated with a ratio of 0.9:0.1 w/w, which is in accordance with the device fabrication condition. The HOMO energy levels of PBTI-FR, PM6:PBTI-FR, and PM6 obtained by testing are -5.22, -5.18, and -5.14 eV, respectively.

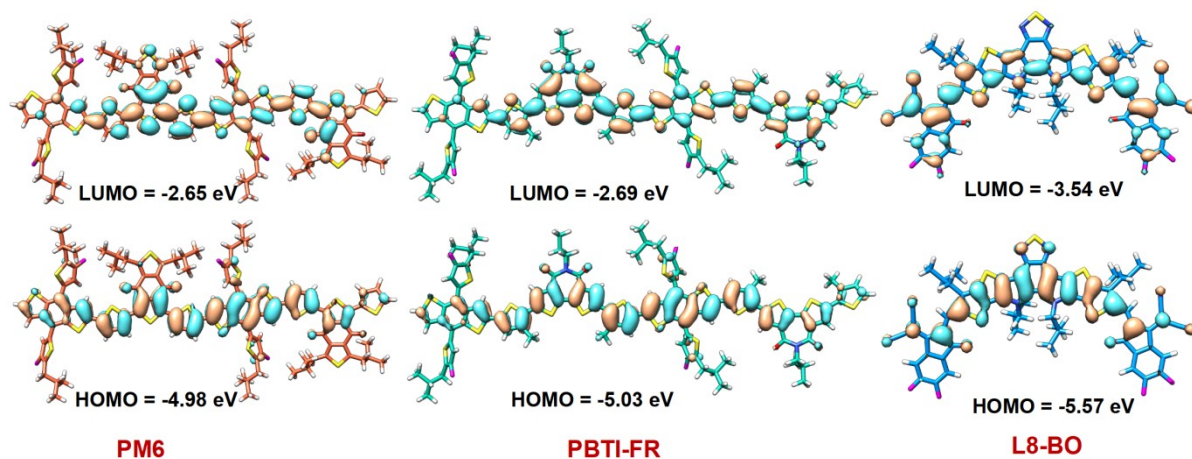


Fig. S16 DFT-calculated (at the B3LYP-D3/6-31G** level) HOMO and LUMO energy levels, along with their corresponding orbital topologies, for dimeric models of PM6 and PBTI-PBTI-FR polymers and L8-BO monomers.

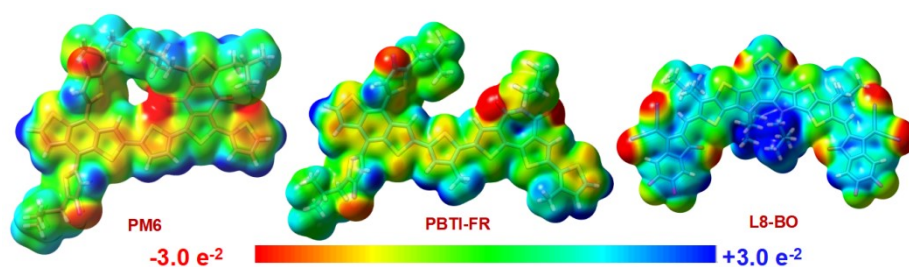


Fig. S17 DFT-calculated (at the B3LYP-D3/6-31G** level) HOMO and LUMO energy levels, along with their corresponding orbital topologies, for dimeric models of PM6 and PBTI-PBTI-FR polymers and L8-BO monomers.

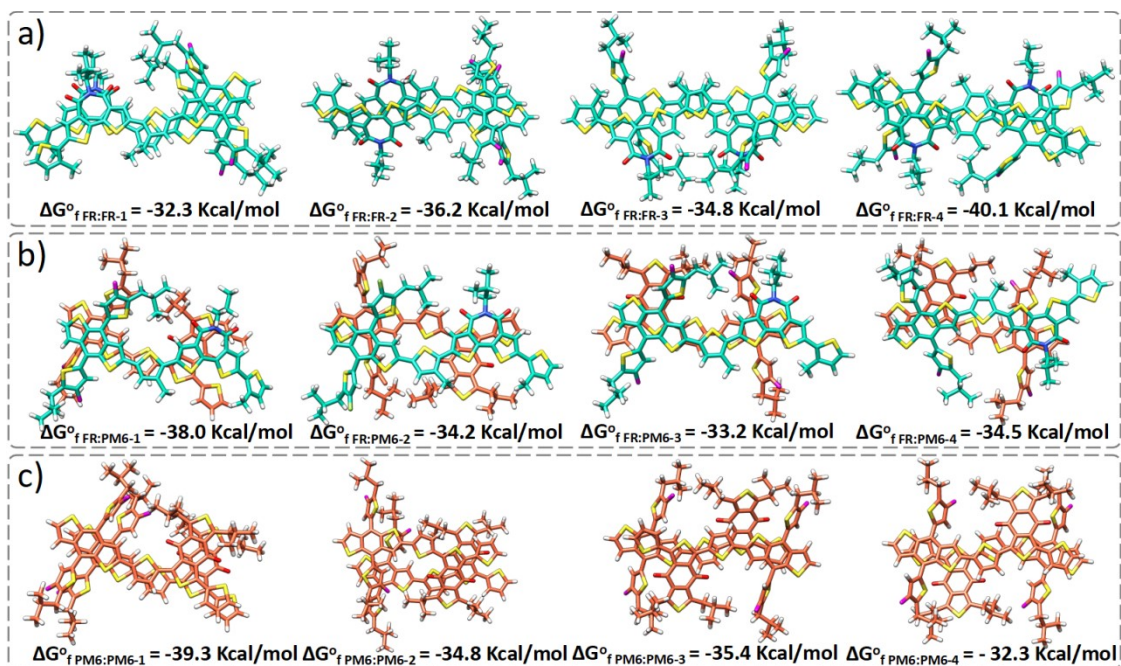


Fig. S18 Top views and Gibbs free energies of formations for the (a) PM6:PM6, (b) PM6:PBTI-FR, and (c) PBTI-FR:PBTI-FR molecular complexes. Four potential molecular arrangements within the π - π dimers were evaluated: parallel (conformation 1), flipped-parallel (conformation 2), antiparallel (conformation 3), and flipped-antiparallel (conformation 4). Calculations were performed at the B3LYP-D3/6-31G** level of theory.

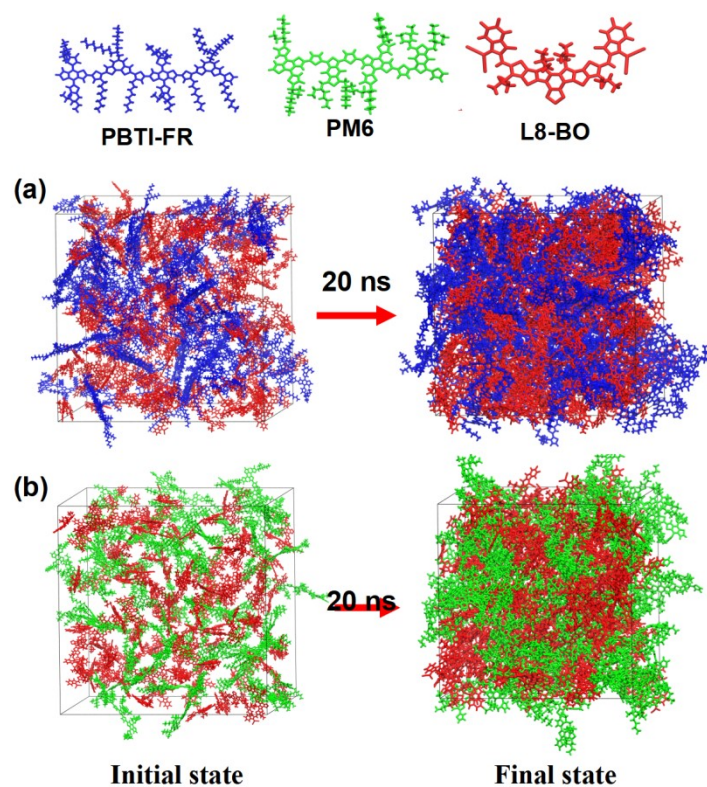


Fig. S19 MD simulated molecular dynamics snapshots of the initial and final states of (a) PBTI-FR:L8-BO and (b) PM6:L8-BO-based systems.



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1. 本中心是国家市场监督管理总局在华南地区设立的国家法定计量检定机构, 本中心的质量管理体系符合 ISO/IEC 17025:2017 标准的要求。

This laboratory is the National Legal Metrological Verification Institution in southern China set up by the State Administration for Market Regulation. The quality system is in accordance with ISO/IEC 17025:2017.

2. 本中心所出具的数据均可溯源至国家计量基准和/或国际单位制 (SI)。

All data issued by this laboratory are traceable to national primary standards and/or International System of Units (SI).

3. 校准地点、环境条件:

Location and environmental conditions of the calibration:

地点 本院二基地A1-402

Place

温度 $(25 \pm 2) ^\circ\text{C}$

Temperature

相对湿度 $(30 \pm 5) \%$

R.H.

4. 本次校准的技术依据:

Reference documents for the calibration:

JJF1622-2017 太阳能电池校准规范: 光电性能 C.S. for Solar Cells: Photoelectric Properties

5. 本次校准所使用的主要计量标准器具:

Major standards of measurement used in the calibration:

设备名称/型号规格/测量范围
Name of Equipment
/Model/Type/Range

编号
Serial No.

证书号/有效期/溯源单位
Certificate No./Due Date
/Traceability to

计量特性
Metrological
Characteristic

ABET 稳态太阳模拟器
1000W full spectrum solar
simulator

374

NYX202400537
/2025-12-03
/本中心

光谱匹配度: A级
辐照度不均匀性: A级
辐照度不稳定性: A级

标准太阳能电池
Standard Solar Cell
/RR_257_0/ $I_{sc}: (1 \sim 200) \text{mA}$

13/01/2014

GXgf2024-06295
/2025-09-24
/中国计量院

$U_{ref}=2.2\%, k=2$

标准源表
Standard Source Meter
/2420/ $(0 \sim 60) \text{V}, (0 \sim 3) \text{A}$

4051271

DBB202403401
/2025-02-26
/本中心

电压: $U_{ref}=0.1\%$, 电
流: $U_{ref}=0.1\%$ ($k=2$)
DCV: $U_{ref}=0.1\%$, DCA: $U_{ref}=0.1\%$ ($k=2$)

— 本说明页以下空白 —

注: 1. 本证书校准结果只与受校准仪器有关。The results relate only to the items calibrated.

Note: 2. 未经本机构书面批准, 不得部分复制此证书。 This certificate shall not be reproduced except in full, without the written approval of our laboratory.

3. “客户名称”、“联络信息”由委托方提供, “制造厂”、“型号规格”、“出厂编号”以及“设备编号”为仪器上标注, 委托方对上面内容如有异议, 须在收到证书后二十个工作日内提出。

The information Name of the Customer and Contact Information are provided by client, and the Manufacturer, Model/Type, Serial No. and Equipment No. are marked on the items. Client shall submit any objection within 20 working days after receiving the certificate for the information above.



校准结果 RESULTS OF CALIBRATION

证书编号 NYX202500017
Certificate No.

原始记录号 NYX202500017
Record No.

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Page of

1、外观检查: 符合要求
Apparent Inspection: Pass.

2、测试条件: 温度 $(25\pm 2)^{\circ}\text{C}$; 辐照度 $1000\text{W}/\text{m}^2$ 。
Test conditions: Temperature: $(25\pm 2)^{\circ}\text{C}$; Irradiance: $1000\text{W}/\text{m}^2$.

3、电流-电压特性曲线和功率-电压特性曲线:
I-V and P-V curves:

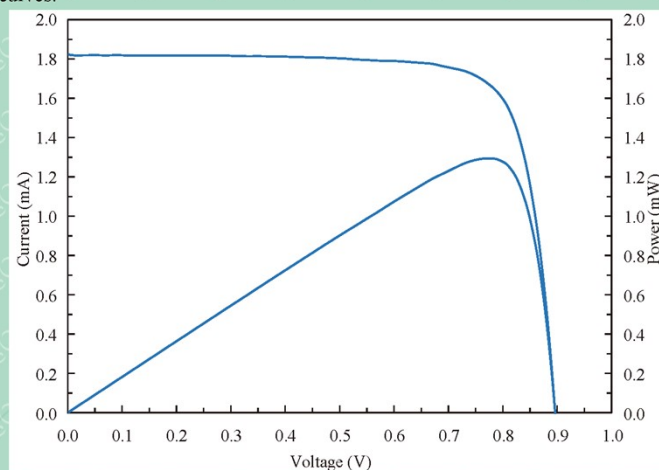


图1 电流-电压特性曲线和功率-电压特性曲线
Figure1 I-V and P-V characteristic curves

4、光电性能参数:
Results of photoelectric properties:

表1(Table 1)

面积	短路电流 密度 J_{sc}	短路电流 I_{sc}	开路电压 V_{oc}	填充因子 FF	最大功率 P_m	最佳工作 电流 I_m	最佳工作电 压 V_m	转换效率 η
Area	Short circuit current density	Short circuit current	Open circuit voltage	Fill factor	Maximum power	Optimum working current	Optimum working voltage	Efficiency
cm^2	mA/cm^2	mA	V	%	mW	mA	V	%
0.0647	28.12	1.819	0.896	79.10	1.289	1.632	0.790	19.92

Fig. S20 Certification report by South China National Center of Metrology.

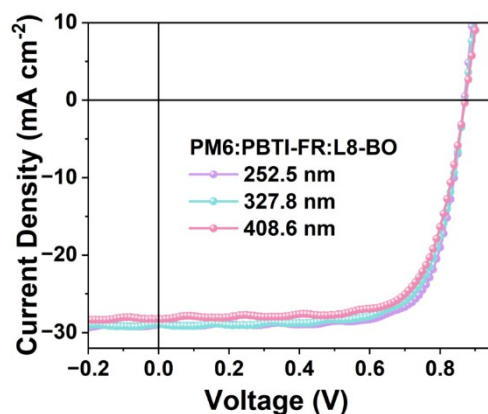


Fig. S21 J - V characteristics of the PM6:PBTI-FR:L8-BO-based devices with distinct active layer thicknesses under AM 1.5G at 100 mW cm^{-2} .

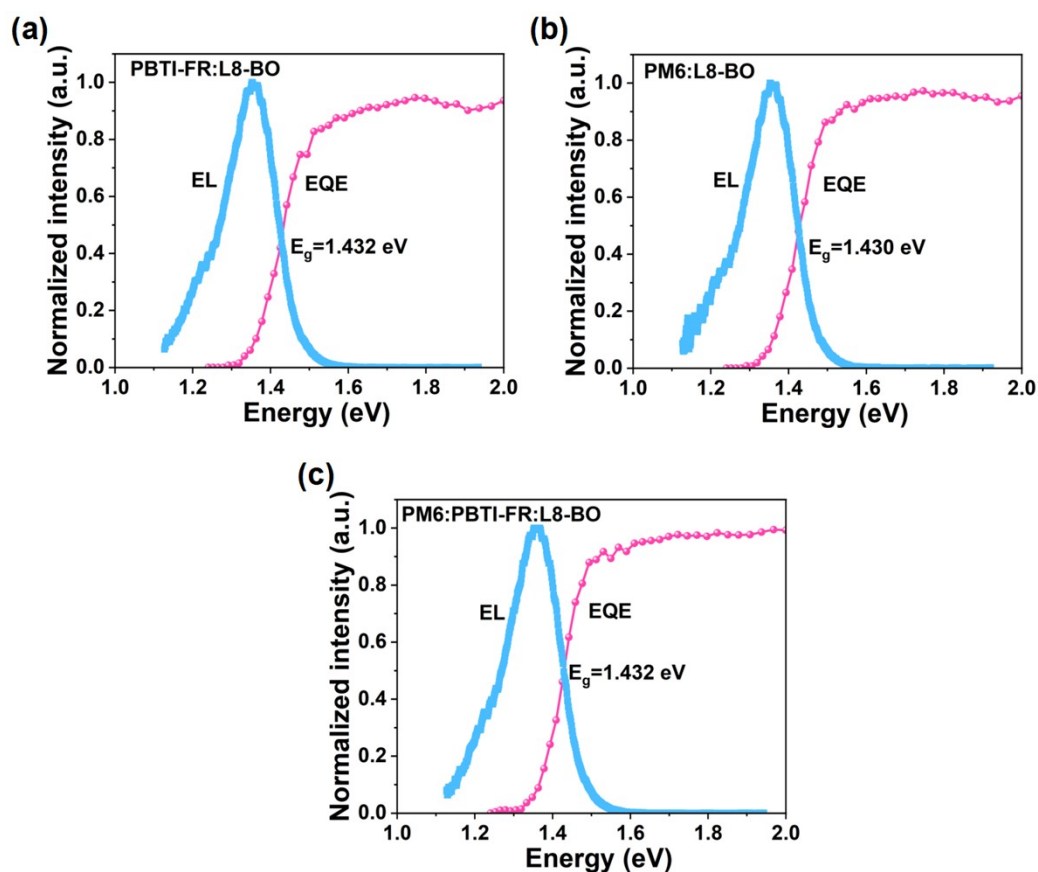


Fig. S22 The determination of E_g s by EL and EQE spectra of (a) PBTI-FR:L8-BO, (b) PM6:L8-BO and (c) PM6:PBTI-FR:L8-BO-based systems.

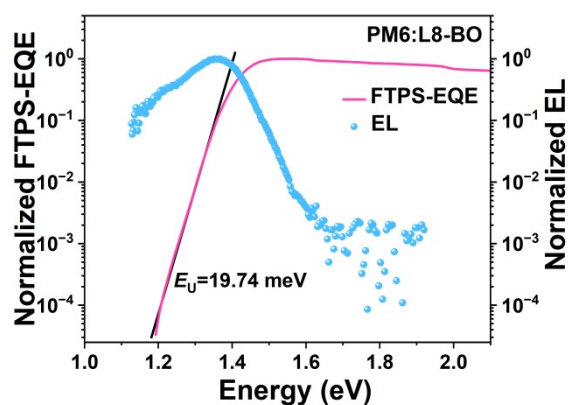


Fig. S23 The FTPS-EQE and EL spectra of the PM6:L8-BO-based OSCs.

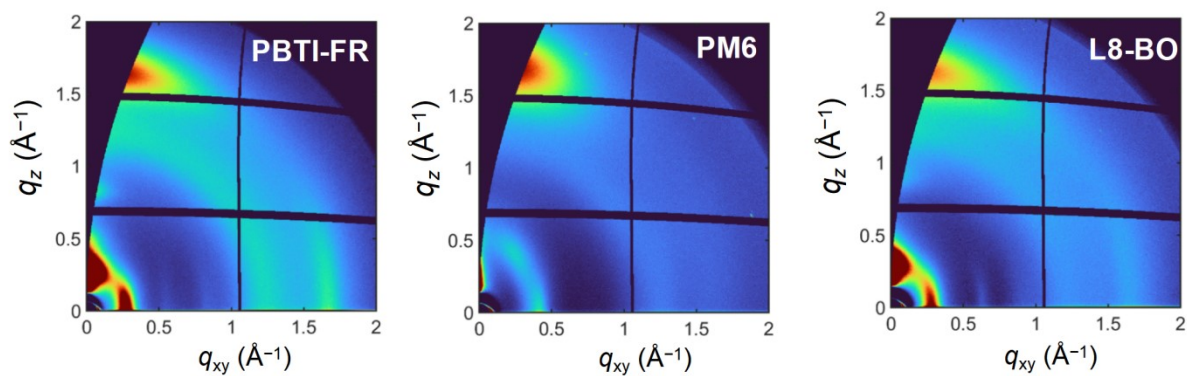


Fig. S24 2D-GIWAXS patterns of the neat films.

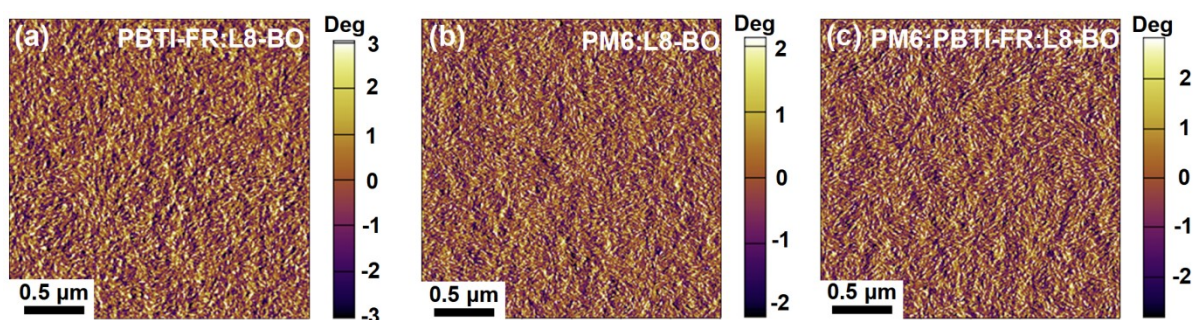


Fig. S25 AFM phase images of (a) PBTI-FR:L8-BO, (b) PM6:L8-BO and (c) PM6:PBTI-FR:L8-BO.

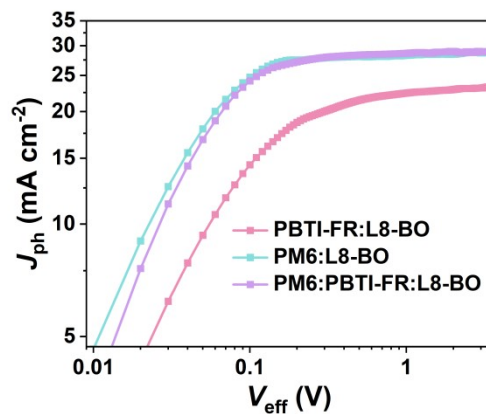


Fig. S26 Plots of photocurrent density (J_{ph}) versus effective voltage (V_{eff}).

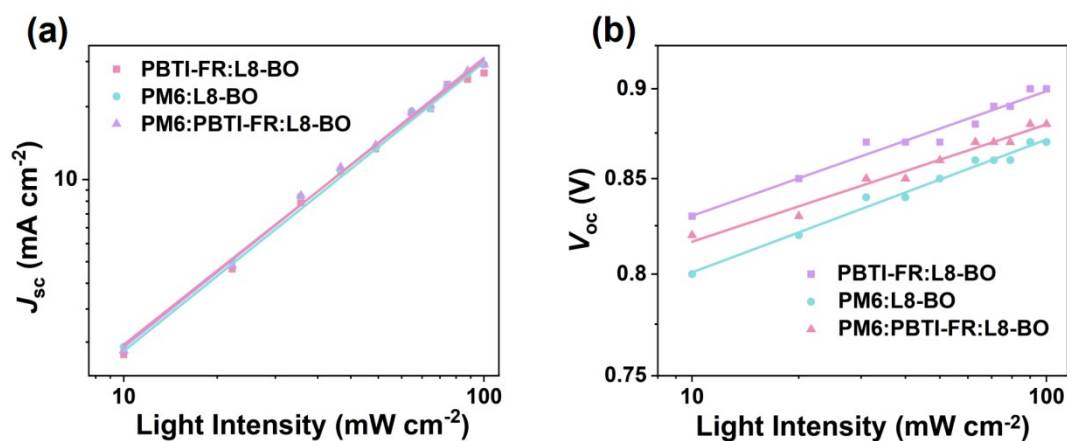


Fig. S27 The dependence of (a) J_{sc} and (b) V_{oc} on light intensity (P_{light}) for binary and ternary devices.

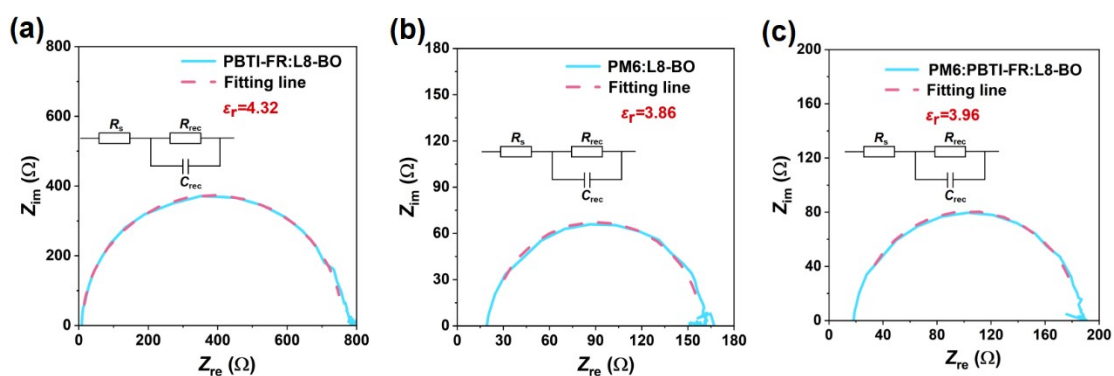


Figure S28. The electrochemical impedance spectroscopy of (a) PBTI-FR:L8-BO, (b) PM6:L8-BO and (c) PM6:PBTI-FR:L8-BO blends, respectively.

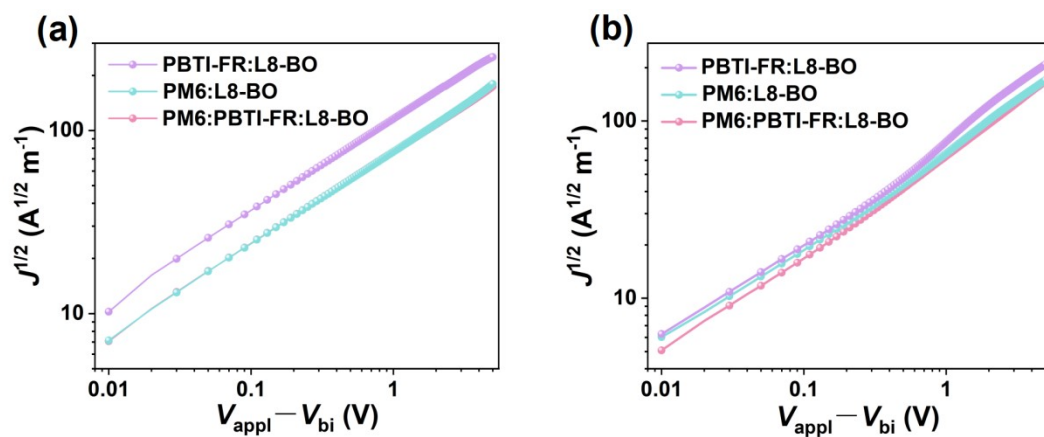


Fig. S29 J - V curves of (a) the hole-only and (b) electron-only devices.

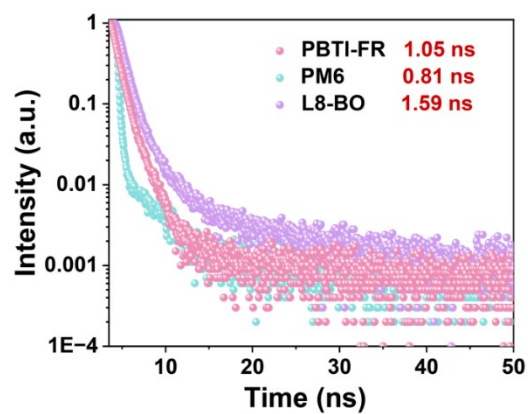


Fig. S30 Normalized TRPL dynamics of neat films.

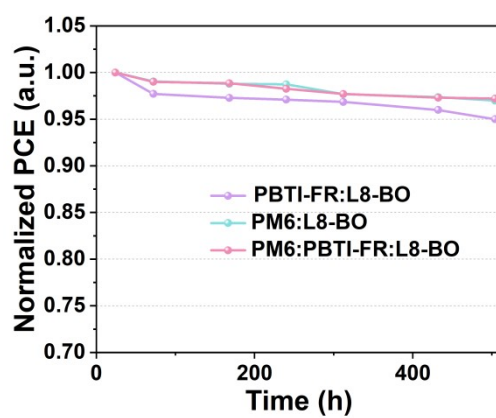


Fig. S31 PCE evolution of storage stability for the binary and ternary devices.

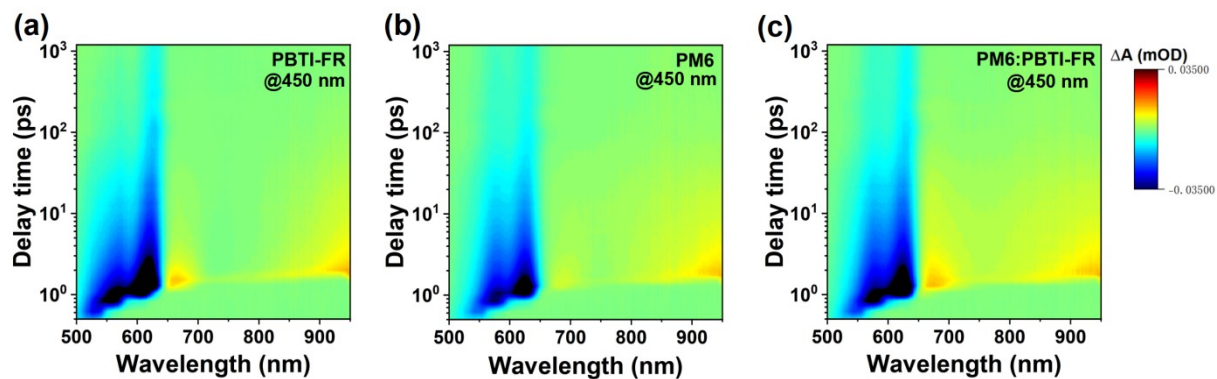


Fig. S32 Femtosecond TA spectra of (a) PBTI-FR, (b) PM6, and (c) PM6:PBTI-FR donor films, pump wavelength @450 nm.

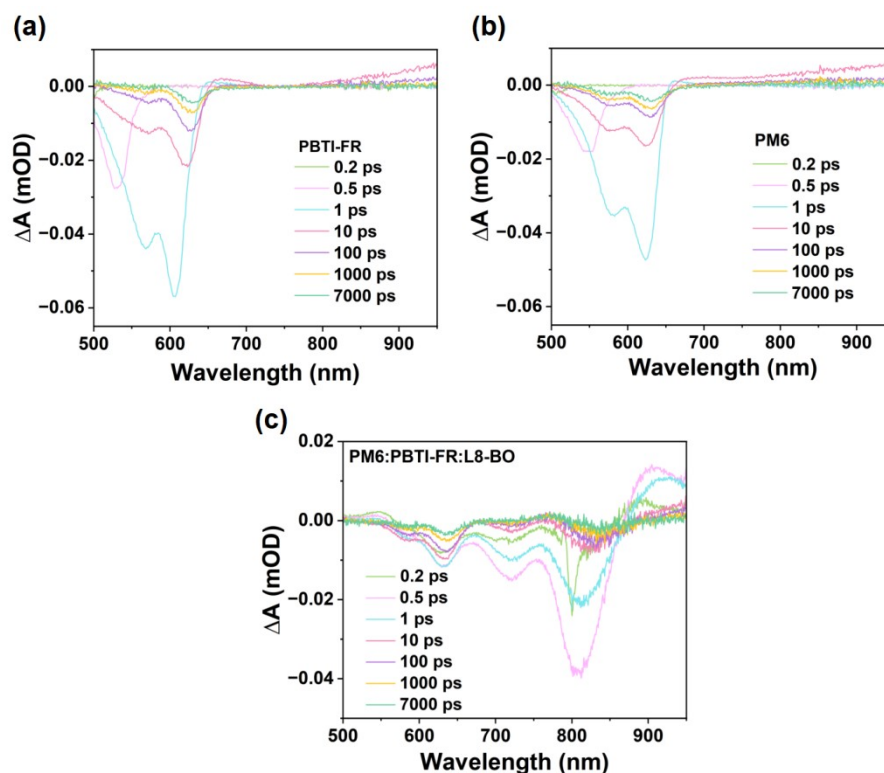


Fig. S33 TA spectra of (a) PBTI-FR, (b) PM6, and (c) PM6:PBTI-FR donor films at various delay times, pump wavelength @450 nm.

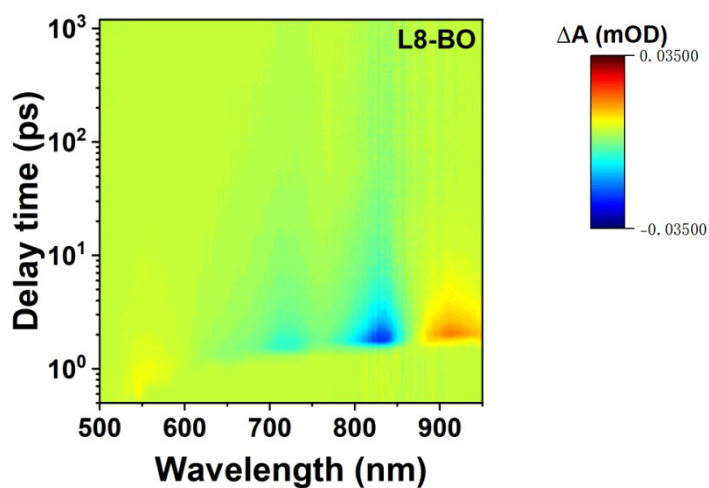


Fig. S34 Femtosecond TA spectra of the L8-BO film, pump wavelength @780 nm.

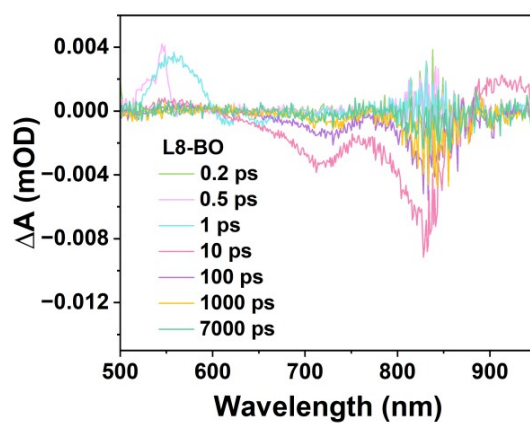


Fig. S35 TA spectra of the L8-BO film at various delay times, pump wavelength @780 nm.

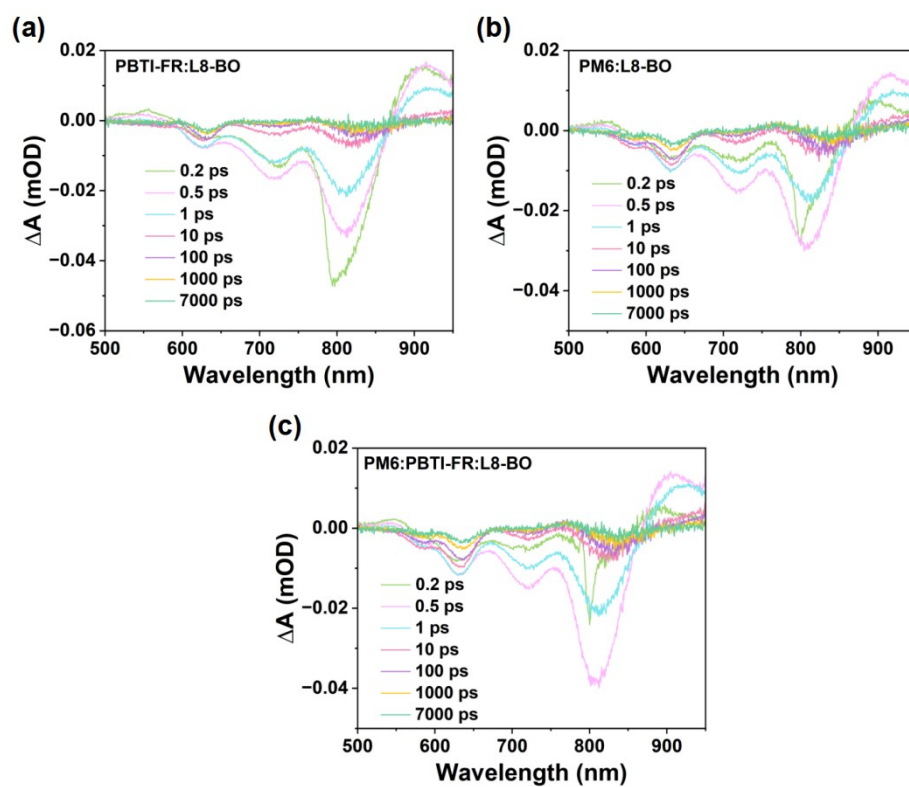


Fig. S36 TA spectra of (a) PBTI-FR:L8-BO, (b) PM6:L8-BO and (c) PM6:PBTI-FR:L8-BO films at various delay times, pump wavelength @800 nm.

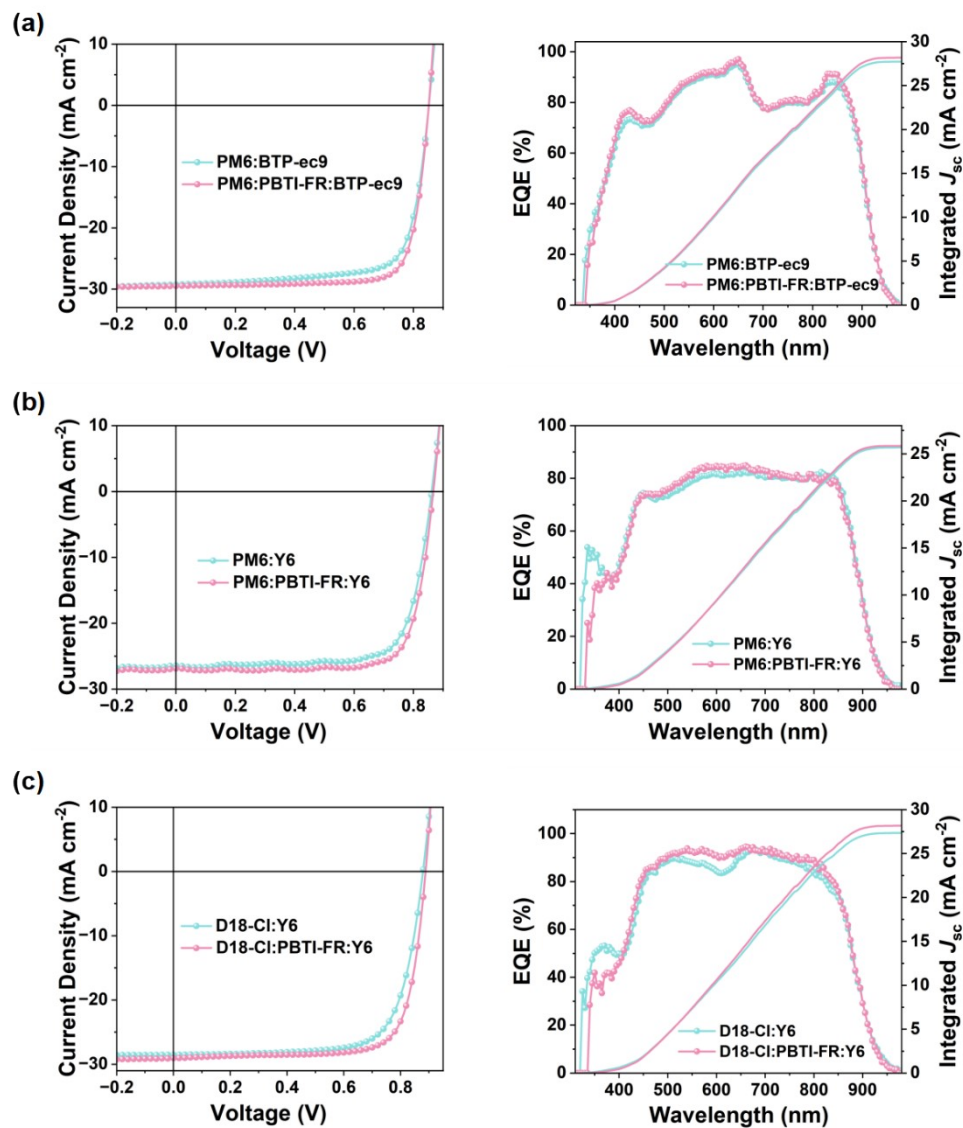


Fig. S37 J - V characteristics and EQE spectra with the integral J_{sc} curves of (a) PM6:BTP-ec9, (b) PM6:Y6 and (c) D18-Cl:Y6-based devices incorporating the PBTI-FR polymer donor.

3. Supplementary Tables

Table S1 The surface-energy parameters of the neat and PM6:PBTI-FR films.

Surface	θ_1 ($^{\circ}$)	θ_2 ($^{\circ}$)	γ_s^d (mJ cm $^{-2}$)	γ_s^p (mJ cm $^{-2}$)	γ_s (mJ cm $^{-2}$)
PBTI-FR	107.49	79.49	25.64	0.06	25.70
PM6	105.70	77.47	26.30	0.13	26.43
L8-BO	94.21	67.19	25.16	2.08	27.24
PM6:PBTI- PBTI-FR	106.71	78.55	26.03	0.09	26.12

Table S2 χ^{A-B} , γ^{X-Y} and ω parameters of the interfaces between different materials.

Interface	χ^{A-B}	γ^{X-Y}	ω
PBTI-FR:PM6	0.0051	0.0083	
PBTI-FR:L8-BO	0.0226	0.0373	2.79 > 1
PM6:L8-BO	0.0063	0.0104	
(PM6:PBTI-FR):(L8- BO)	0.0119	--	--

Table S3 DFT-calculated Gibbs free energies of formation (ΔG°_f), intermolecular interaction energies ($E_{\text{interaction}}$), and intermolecular binding energies (E_{bonding}) for the different PM6:PM6, PM6:PBTI-FR, and PBTI-FR:PBTI-FR complexes.

Sample	ΔG°_f (kcal mol ⁻¹)	E_{bonding} (kcal mol ⁻¹)	$E_{\text{interaction}}$ (kcal mol ⁻¹)
PM6:PM6-1	-39.34	-66.53	-71.55
PM6:PM6-2	-34.81	-60.51	-66.00
PM6:PM6-3	-35.36	-58.80	-61.15
PM6:PM6-4	-32.25	-55.89	-59.17
PM6:PBTI-FR-1	-38.03	-68.62	-77.55
PM6:PBTI-FR-2	-34.23	-62.10	-68.71
PM6:PBTI-FR-3	-33.23	-60.95	-66.77
PM6:PBTI-FR-4	-34.47	-60.30	-65.82
PBTI-FR:PBTI-FR-1	-32.28	-63.15	-72.19
PBTI-FR:PBTI-FR-2	-36.23	-65.76	-72.21
PBTI-FR:PBTI-FR-3	-34.80	-63.06	-65.13
PBTI-FR:PBTI-FR-4	-40.05	-69.77	-74.44

Table S4 Photovoltaic performance of the TOSCs processed by different D1:D2 ratios in the active layer (AM 1.5 G illumination at 100 mW cm⁻².)

D1:D2	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)
0.8:0.1	0.89	27.84	79.98	19.87
0.9:0.1	0.89	28.07	82.55	20.52
1:0.1	0.88	27.22	81.92	19.70

Table S5 Comparison of our results with the reported PCEs of ternary devices.

Active layer	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)	Reference
D18:BTP-eC9-4F:SM16	0.899	27.3	78.54	19.28	10
D18:N3:YIS-6F	0.865	28.4	79.1	19.43	11
PM6:eC9:5-IDT	0.863	29.23	79.1	19.96	12
PM6:L8-BO:Qx-p-N4F	0.887	27.83	78.91	19.48	13
D18:N3-BO:F-BTA3	0.924	26.77	81.85	20.25	14
PBD-10:PBTz-F:L8-BO	0.897	27.27	79.49	19.45	15
PM6:L8-BO:BTP-OS	0.898	27.01	79.72	19.34	16
PM6 : eC9 : BTP-2FCIO	0.863	27.96	80.15	19.34	17
PM6:BTP-eC9:Qx-5Cl	0.862	29.04	79.24	19.83	18
PM6:BTP-eC9:BTP-9F	0.85	28	79.9	19.1	19
PM6:oPhFO:BTP-ec9	0.878	27.6	80.6	19.5	20
PM6 : L8-BO : BTP-eC9	0.873	28.07	81.6	20	21
PM6:D18-Cl:NA3	0.911	27.55	78.7	19.75	22
PBDB-TF:AA-2:BO-4I	0.917	26	81	19.3	23
PM6:CH8-6:L8-BO	0.884	27.46	78.6	19.2	24
PM6:BTP-eC9:SMA	0.863	29.1	80.52	20.22	25
D18:Z8:L8-BO	0.92	27.2	80	20.2	26
D18:LJ1:L8-BO	0.924	26.84	79.12	19.78	27
D18 : L8-BO : DM-F	0.92	26.98	81	20.09	28
PTB7-Th : CA-CN : DICTF	0.95	21.78	72	14.91	29
D18:N3:ANF-3	0.844	27.9	80.4	18.93	30
D18:PM6:L8-BO	0.9	27.5	80.4	19.9	31
PM6:BTP-C9:o-BTP-eC9	0.86	28.75	80.41	19.88	32

PM6:BTP-eC9:PBB-Cl	0.853	28.53	78.23	19.04	33
PM6:Y6 :BTP-PIO	0.848	27.31	73.9	17.1	34
D18:N3:DP-BTP	0.87	27.95	78.5	19.07	35
PM6:L8-BO:PC71BM (LbL)	0.9	26.77	75.56	18.2	36
PBBTz-Cl:PY-IT:BTP-2T2F	0.922	25.82	78.11	18.6	37
D18-Fu:Y6-IO:PC71BM	0.9	23.62	80.4	17.07	38
PM6:Y6:ITIC-M	0.859	26.35	80.1	18.13	39
PM6:PBTI-FR:L8-BO	0.89	28.07	82.55	20.52	this work

Table S6 Photovoltaic performance parameters of the ternary organic solar cells with distinct active layer thicknesses under AM1.5G at 100 mW cm⁻² illumination.

Film thickness	V_{oc}	J_{sc}	FF	PCE
(nm)	(V)	(mA cm ⁻²)	(%)	(%) ^a
253	0.87	28.94	74.77	18.80 (18.52±0.16)
328	0.87	28.92	72.06	18.14 (18.06±0.12)
409	0.87	28.21	71.42	17.55 (17.36±0.13)

^a Average values with standard deviation from 10 devices.

Table S7 The lamellar distances (q_{xy} profile, 100) and (q_z profile, 010) of the neat and blend films with L8-BO.

Sample	$d_{(100)}^{\text{IP}}$ (Å ⁻¹)	$\text{CCL}_{(100)}^{\text{IP}}$ (nm)	$d_{\pi-\pi}^{\text{OOP}}$ (Å ⁻¹)	$\text{CCL}_{(010)}^{\text{OOP}}$ (nm)
PBTI-FR	22.9	10.3	3.8	2.6
PM6	22.2	8.2	3.8	1.5
L8-BO	15.48	5.0	3.7	1.6
PBTI-FR:L8-BO	22.8	13.5	3.7	3.6
PM6:L8-BO	20.9	7.9	3.7	2.5
PM6:PBTI-FR:L8-BO	20.9	10.7	3.7	3.4

Table S8 P_{diss} , P_{coll} , α (J_{sc}) and α (V_{oc}) parameters for the binary and ternary devices.

Active layer	P_{diss} (%)	P_{coll} (%)	α (J_{sc})	α (V_{oc})
PBTI-FR:L8-BO	94.72	80.07	0.956	1.26
PM6:L8-BO	95.85	88.25	0.983	1.20
PM6:PBTI-FR:L8-BO	98.02	90.32	0.998	1.10

Table S9 The μ_h and μ_e parameters via the SCLC measurement.

Active layer	Film thickness	μ_h	μ_e	μ_h/μ_e
	(nm)	($\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$)	($\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$)	
PBTI-FR:L8-BO	111	4.86×10^{-4}	3.48×10^{-4}	1.40
PM6:L8-BO	117	2.97×10^{-4}	3.26×10^{-4}	0.91
PM6:PBTI-FR:L8-BO	118	3.25×10^{-4}	3.29×10^{-4}	0.99

Table S10 Fitted parameters of photoluminescence decay curves for neat films.

Film	τ_1 (ns)	τ_2 (ns)	τ (ns)
PBTI-FR	0.78	2.20	1.05
PM6	0.24	3.72	0.81
L8-BO	1.00	5.37	1.59

Table S11 Fitted parameters of photoluminescence decay curves for binary and ternary films.

Active Layer	τ_1 (ns)	τ_2 (ns)	τ (ns)
PBTI-FR:L8-BO	0.24	4.28	0.69
PM6:L8-BO	0.22	4.06	0.80
PM6:PBTI-FR:L8-BO	0.25	6.06	1.07

Table S12 Normalized thermal stability evolution data for PBTI-FR:L8-BO under 65 °C heating for 1008 h.

Time (h)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)
0	100.00	100.00	100.00	100.00
48	100.00	103.03	95.90	98.35
96	100.00	103.40	93.63	97.06
168	100.00	101.89	94.22	96.08
192	100.00	100.37	95.39	95.47
216	98.90	101.15	94.14	94.67
264	98.90	102.42	92.35	93.93
288	100.00	99.47	93.87	93.75
312	100.00	101.68	92.42	93.50
336	100.00	101.35	92.02	93.14
360	100.00	101.19	91.90	92.89
432	98.90	101.72	91.97	92.10
456	98.90	100.45	92.01	91.30
504	98.90	97.09	93.76	89.77
528	98.90	97.05	92.68	88.97
576	98.90	96.27	92.39	88.24
648	98.90	93.23	93.65	86.58
744	98.90	95.04	91.90	86.46
840	98.90	92.21	93.38	85.23
912	98.90	93.15	92.44	85.23
984	98.90	92.70	91.17	84.68
1008	98.90	93.36	91.64	84.31

Table S13 Normalized thermal stability evolution data for PM6:L8-BO under 65 °C heating for 1008 h.

Time (h)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)
0	100.00	100.00	100.00	100.00
48	100.00	101.05	98.68	99.73
96	100.00	98.26	99.17	97.47
168	98.82	100.00	97.37	96.45
192	98.82	99.24	96.92	95.05
216	98.82	100.00	95.28	94.40
264	98.82	100.76	95.02	93.97
288	98.82	99.20	95.23	93.21
312	98.82	100.51	93.94	93.05
336	98.82	98.18	94.64	92.46
360	98.82	98.37	94.65	92.35
432	98.82	96.73	96.42	91.60
456	98.82	95.46	96.17	90.79
504	98.82	93.79	95.28	88.31
528	97.65	94.12	95.18	87.88
576	98.82	94.37	94.60	87.83
648	97.65	94.41	94.72	87.61
744	98.82	93.43	94.33	87.08
840	98.82	91.87	95.63	86.81
912	96.47	94.59	93.80	85.62
984	97.65	96.77	89.96	85.35
1008	97.65	96.59	89.01	84.87

Table S14 Normalized thermal stability evolution data for PM6:PBT-FR:L8-BO under 65 °C heating for 1008 h.

Time (h)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)
0	100.00	100.00	100.00	100.00
48	100.00	101.04	97.65	99.19
96	100.00	101.04	95.61	97.11
168	100.00	102.62	93.56	96.50
192	100.00	100.75	95.10	96.10
216	100.00	100.36	95.09	95.95
264	100.00	100.72	95.10	95.80
288	100.00	101.26	93.68	95.34
312	100.00	100.54	94.24	95.24
336	100.00	100.72	94.22	95.14
360	100.00	101.40	94.00	94.88
432	98.86	101.55	94.17	94.63
456	100.00	101.40	93.11	94.53
504	98.86	101.80	94.37	94.38
528	98.86	100.36	93.10	94.17
576	98.86	100.00	94.41	93.97
648	98.86	101.47	93.14	93.92
744	100.00	99.86	93.08	93.41
840	100.00	98.74	93.43	93.11
912	100.00	100.11	92.73	92.86
984	100.00	99.17	92.63	92.35
1008	98.86	99.21	93.72	92.20

Table S15 Photovoltaic performance parameters for PM6:BTP-ec9, PM6:Y6 and D18-Cl:Y6-based devices incorporating the PBTI-FR polymer donor under AM 1.5G at 100 mW cm⁻².

Active layer	V_{oc} (V)	J_{sc} (mA cm ⁻²)	Cal. J_{sc} (mA cm ⁻²) ^a	FF (%)	PCE (%) ^b
PM6:BTP-ec9	0.85	29.25	27.75	74.53	18.57 (17.00±0.38)
PM6:PBTI-FR:BTP-ec9	0.85	29.50	28.17	79.46	19.96 (19.80±0.14)
PM6:Y6	0.86	26.43	25.68	75.55	17.20 (16.89±0.20)
PM6:PBTI-FR:Y6	0.87	26.89	25.84	78.67	18.34 (17.85±0.22)
D18-Cl:Y6	0.88	28.54	27.34	72.83	18.27 (17.84±0.29)
D18-Cl:PBTI-FR:Y6	0.89	29.07	28.18	76.41	19.72 (19.39±0.22)

^b Integrated current density obtained from EQE spectra.

^a Average values from 10 devices with standard deviation included in parenthesis.

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