Supporting Information

Manipulating Crystallization Kinetics and Vertical Phase Distribution via Small Molecule Donor Guest for Organic Photovoltaic Cells with 20% Efficiency

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Experimental Section

1. Materials

All chemicals and solvents were reagent grade and purchased from Alfa Aesar and TCI. The donor BTR-SCl, and PM6 were synthesized according to previous reports.^{1, 2} L8-BO and D18 were purchased from Solarmer Materials Inc. The number average molecular weight (Mn) of PM6 and D18 are 49.9 and 59.1 kDa with polydispersity index (PDI) of 2.4 and 2.3, respectively.

2. Characterization Techniques and Instrument

UV-Vis-NIR and Photoluminescence (PL) measurements: The UV-Vis-NIR absorption spectra of films were recorded using a SHIMADZU UV-1900i UV-Vis-NIR spectrophotometer. Photoluminescence (PL) spectra were obtained on an Edinburgh Instrument FLS 980 spectrofluorometer.

Cyclic voltammetry (CV): Electrochemical cyclic voltammetry (CV) measurements were carried out on a electrochemical workstation (Squidstat Plus, Admiral Instrument) with a glassy carbon disk, Pt wire, and Ag/Ag⁺ electrode as the working electrode, counter electrode, and reference electrode, respectively, in a 0.1 M tetrabutylammonium hexafluorophosphate (Bu₄NPF₆) acetonitrile solution. The reference electrode was checked versus ferrocenium-ferrocene (Fc⁺/Fc) as the internal standard as recommended by IUPAC (vacuum energy level: 24.8 eV). All the solutions were deaerated by bubbling nitrogen gas for a few minutes prior to the electrochemical measurements. HOMO energy levels were calculated from the equation $E_{\text{HOMO}} = -(E_{\text{onset(ox)}} + 4.80)$ eV and LUMO was calculated from $E_{\text{LUMO}} = -(E_{\text{onset(re)}} - 4.80)$ eV.

Film-depth-dependent Light Absorption Spectroscopy (FLAS): Film-depth-dependent light absorption spectra were acquired by an in-situ spectrometer (PU100, Shaanxi Puguang Weishi Co. Ltd.) equipped with a soft plasma-ion source. Oxygen plasma etching technology was used to collect the depth-resolved absorption spectra.³

Charge mobility measurement: The hole and electron mobilities of devices were evaluated from the space-charge-limited current (SCLC) method with the hole-only structure of ITO/PEDOT:PSS/blend films/MoO₃/Ag and the electron-only structure of ITO/ZnO/blend films/PDINN/Ag, respectively. The corresponding charge mobilities were calculated by fitting the Mott-Gurney square law $J = 9\varepsilon_r\varepsilon_0\mu V^2/(8L^3)$, where J is the current density, ε_r is the dielectric permittivity of the active layer, ε_0 is the vacuum permittivity, L is the thickness of the active layer, and μ is the hole or electron mobility.

J-V and EQE measurements: The PCE values of all devices were measured under an illumination of AM 1.5G (100 mW cm⁻²) using a Keithley 2400 source meter generating from LSS-55 solar simulator (50 × 50 mm spot size) of LightSky Technology CO., LTD. A 2 × 2 cm² monocrystalline silicon reference cell (LRC-KG5-012) was purchased from LightSky Technology CO. The EQE spectra were recorded utilizing the spectral response measurement system LST-QE of LightSky Technology CO., LTD. The light intensity at each wavelength was calibrated with a standard singlecrystal Si photovoltaic cell.

TPC, TPV and CE measurements: Transient photovoltage (TPV), transient photocurrent (TPC), and charge extraction (CE) measurements were obtained by the all-in-one characterization platform, Paios (Fluxim AG, Switzerland).

Calculation of bimolecular charge recombination rate constant: The carrier density (*n*) and carrier lifetimes (τ) can be obtained from CE and TPV measurements. The relationship between *n* and τ is determined using the equation:

$$\tau = \tau_0 n^{-\lambda} \tag{1}$$

where τ_0 is constant and λ is the recombination exponent. The recombination order $R = \lambda + 1$. Furthermore, the bimolecular recombination rate constants k_{rec} are calculated from τ and n according to the formula:

$$k_{rec} = \frac{1}{(\lambda + 1)n\tau(n)} \tag{2}$$

sEQE, EQE_{EL}, and EL measurements: sEQE, EQE_{EL}, and EL measurements were measured using an integrated system (LST-QE, LightSky Technology CO., LTD).

Energy loss measurements: According to the SQ limit model, E_{loss} in OSCs can be classified into three segments, as illustrated by the following equation:

$$E_{loss} = E_g - qV_{OC}$$

= $(E_g - qV_{OC}^{SQ}) + (qV_{OC}^{SQ} - qV_{OC}^{rad}) + (qV_{OC}^{rad} - qV_{OC})$
= $(E_g - qV_{OC}^{SQ}) + qV_{OC}^{rad} + qV_{OC}^{non - rad}$
= $\Delta E_1 + \Delta E_2 + \Delta E_3$ (3)

where ${}^{qV_{OC}^{SQ}}$ is the thermodynamic limit of ${}^{V_{OC}}$ in the SQ, ${}^{V_{OC}^{rad}}$ is the radiative limit of ${}^{qV_{OC}}$, ${}^{\Delta E_1}$ is the loss between E_g and ${}^{V_{OC}^{SQ}}$, which is commonly inevitable for any type of solar cell, and ${}^{\Delta E_2}$ is the loss resulted from radiative recombination under the optical gap. ${}^{\Delta E_3}$ is the nonradiative recombination, which is defined as follows:

$$\Delta E_3 = q V_{OC}^{non-rad} = -k_B T \ln(EQE_{EL}) \tag{4}$$

where k_B is the Boltzmann constant.

Calculation of trap density:

The frequency axis can be scaled to the energy axis through the following equation:

$$E_{\omega} = kT \ln\left(\frac{2\nu_0}{\omega}\right) \tag{5}$$

Where ω is the angular frequency calculated by $\omega = 2\pi f$, v_0 is the attempt-to-escape frequency of 10⁹ Hz. The trap density at energy E_{ω} can be acquired as:

$$N_t(E_\omega) = -\frac{V_{bi} dl C \,\omega}{q d d \omega k T} \tag{6}$$

d is the thickness of the active layer and V_{bi} is the built-in voltage measured through Mott–Schottky characterization. A Mott-Schottky plot (1/ C^2 versus V) is used to extract the built-in voltage V_{bi} :

$$\frac{1}{C_j^2} = \frac{2(V_{bi} - V)}{q\varepsilon_0 \varepsilon N_{a_p}}$$
(7)

Where $N_{\rm ap}$ is the apparent doping profile.

Then the energy distribution can be described with Gaussian shape distribution:

$$N_t(E) = \frac{N_t}{\sqrt{2\pi\delta}} \exp\left[-\frac{(E_t - E)^2}{2\delta^2}\right]$$
(8)

Where N is the total density, E_t is the center of the DoS, and δ is the disorder parameter.

In-situ UV-Vis measurement: In situ fast response ultraviolet-visible absorption spectroscopy measurements were obtained on a multi-spectrometer (DU-200, Shaanxi Puguang Weishi Co. Ltd.). According to the preparation process of the devices, the active layer materials dissolved in chloroform solution were spin-coated on clean quartz flakes. In this process, the in-situ UV-vis absorption spectra were obtained on a multi-spectrometer. The sampling intervals and integration times were set as 1 and 10 ms, respectively.

Contact angles measurements: Contact angles were measured on the neat donor and acceptor films with the contact angle meter (Dataphysics OCA40 Micro) by using two different solvents (water and diiodomethane).

Calculation methods of Flory-Huggins interaction parameter (χ): The Flory–Huggins interaction parameter $\chi_{D:A}$ was used to evaluate the miscibility between the donor and acceptor, which could be calculated as follow:

$$\chi_{D:A} = K \left(\sqrt{\gamma_D - \gamma_A} \right)^2 \tag{9}$$

where *K* is a positive constant, and γ_D and γ_A are the respective surface energies of the donor and acceptor materials.

GIWAXS measurements: Grazing incidence wide-angle X-ray scattering (GIWAXS) measurements were conducted at a Xeuss 3.0 SAXS/WAXS laboratory beamline at Vacuum Interconnected Nanotech Workstation (Nano-X) in China with Kα X-ray of Cu source (operated at 50kV, 0.6 mA, 1.5419 Å). GIWAXS patterns were recorded by a two-dimensional X-ray detector (Eiger2 R 1M, Dectris). The incident angle was 0.18°. The crystalline coherence length (CCL) was

defined as CCL = $0.9 \times (2\pi/\text{FWHM})$ (Å) = $0.09 \times (2\pi/\text{FWHM})$ (nm), where FWHM is the full width at half maximum of the corresponding diffraction peak.

AFM and TEM characterization: Atomic force microscopy (AFM) images of blend films were recorded using a Dimension 3100 (Veeco) atomic force microscope in tapping mode. Transmission electron microscopy (TEM) images were obtained by using a TePNai G2 F20 S-TWIN instrument at an accelerating voltage of 200 kV, in which the blend films were prepared using the following processing technique: first, the blend film to be tested was spin-coated on an ITO/PEDOT:PSS substrate; then, the obtained blend film on the substrate was immersed in deionized water, and the separate blend film was peeled off by the interaction between PEDOT:PSS and water; finally, the blend film floating on the water surface was removed using an unsupported 200 mesh copper mesh and used for TEM measurements.

3. Device Fabrication

The ITO-coated glass substrates were alternately washed three times with deionized water, acetone, and isopropanol and then sprayed dry with nitrogen gas. After surface oxygen enrichment with UVozone cleaner, PEDOT:PSS (Heraeus Clevios P VP Al 4083) was spin-coated onto the clean ITO substrate at 6000 rpm for 40 s. Subsequently, the films were annealed on a hot plate at 150 °C for 15 min. Then ITO substrates coated with PEDOT:PSS films were transferred into a high-purity nitrogenfilled glove box. The solutions of PM6:L8-BO (1:1.2 w/w, 16.5 mg/mL in total), PM6:BTR-SCI:L8-BO (0.9:0.1:1.2 w/w, 16.5mg/mL in total), and BTR-SCI:L8-BO (1:1.2 w/w, 20 mg/mL in total) in chloroform (CF) was stirred at 50 °C for 2 hours. Before the spin coating process, 0.25% 1,8diiodooctane (v/v) was added to the solutions. The blend solution was spin-cast at 3000-4000 rpm for 40 s over the PEDOT: PSS layer to form an active layer. The solutions of D18:L8-BO (1:1.2 w/w, 11mg/mL in total) and D18:BTR-SCI:L8-BO (1:0.1:1.2 w/w, 11mg/mL in total) in CF stirred at 80 °C for 1 hours in advance and then spin-coated on top of the PEDOT:PSS layer at 2500-3000 rpm for 40 s to form an active layer. All active layers were thermal annealed at 90°C for 10 min. The optimized thickness of the BHJ is ~100 nm, which was measured by Bruker Dektak-XT profilometer. Subsequently, a thin layer of PNDIT-F₃N (0.5 mg ml⁻¹ in methanol with 0.5% acetic acid (vol%) was spin-coated onto the active layer at a rate of 3500 rpm for 30 s. Finally, 100 nm Ag was vapour deposited on the films under a ca. 4×10^{-5} Pa pressure with a shadow mask to maintain the active area of the devices (0.049 cm²).



Fig. S1 Film absorption spectra of (a) PM6 and (b) L8-BO films with and without the addition of 10 wt% BTR-SC1.



Fig. S2 (a) *J*–*V* curves and (b) EQE spectra of PSCs based on PM6: BTR-SCl: L8-BO BHJ with various ratio of donor and acceptor.



Fig. S3 (a) The molecular structure of D18. (b) J-V curves and (c) EQE spectra of PSCs based on D18:L8-BO and D18:BTR-SCI:L8-BO.



Fig. S4 Normalized PCEs of the unencapsulated PSCs based on PM6:L8-BO, and PM6:BTR-SCl:L8-BO under 85 °C in a N₂-filled glove box.



Fig. S5 J_{SC} as functions of light intensity for the three PSCs



Fig. S6 PL intensities of neat films and related blend films, excited at (a) 580 nm, (b) 642 nm, and (c) 800nm.



Fig. S7 TPC curves of binary and ternary PSCs.



Fig. S8 The $J^{0.5}-V$ curves of the (a) hole-only diodes and (b) electron-only diodes based on binary and ternary films.



Fig. S9 (a) The capacitance-voltage curves and (b) the capacitance-frequency curves for PM6:L8-BO and PM6:L8-BO:BTR-SCl blend films.



Fig. S10 DSC profiles for the heating process of (a) PM6, BTR-SCl, and PM6:BTR-SCl films. (b)L8-BO, BTR-SCl, and BTR-SCl:L8-BO films.



Fig. S11 Contact angle images of water and diiodomethane (DIM) droplets on PM6, L8-BO, and BTR-SCl neat films.



Fig. S12 (a) GIWAXS patterns of L8-BO film. (b) The corresponding in-plane (IP) and out-of-plane (OOP) line-cuts.



Fig. S13 (a) AFM height images, (b) AFM phase images, and (c) TEM phase images of the binary and ternary films.



Fig. S14 In situ UV-vis absorption spectra of the (a) PM6:L8-BO and (b) PM6:BTR-SCI:L8-BO films during spin coating.

	V _{OC}	$J_{ m SC}/J_{ m cal}$	FF	РСЕ
PM6:BTR-SCI:L8-BO	(V)	(mA cm ⁻²)	(%)	(%)
1:0:1.2	0.88	26.2/25.0	78.1	17.9
0.9:0.1:1.2	0.89	26.8/25.6	81.5	19.4
0.8:0.2:1.2	0.89	26.6/25.3	80.0	18.9
0.7:0.3:1.2	0.89	25.8/24.8	79.2	18.2
0:1:1.2	0.90	17.7/17.4	61.7	9.9

Table S1 The performance parameters of the champion devices based on PM6:L8-BO with differentBTR-SCl components.

 Table S2 Summary of T-PSCs with PCEs over 18% through guest small molecule donor.

Ref.	Donor	Acceptor	Small molecule donor	FF (%)	РСЕ (%)	
14	D18-Cl	Y6	G19	77.7	18.53	
2 ⁵	D18	Y6	ZW1	76.9	18.50	
36	D18	CH-6F	CBTSeHR	76.7	18.41	
47	D18	CH-6F	FSBTSeEHR	76.1	18.55	
5 ⁸	D18	Y6	BTR	80.28	19.08	
6 ⁸	D18	Y6	BTR	81.16	19.41	
7 ⁹	D18	BTP-EC9-4F	LJ1	79.45	19.43	
8 ⁹	D18	L8-BO	LJ1	79.12	19.78	
98	PM6	Y6	BTR	78.84	18.39	
10 ¹⁰	PM6	Y6	DB-1	77.9	18.07	
11 ¹⁰	PM6	Y6	DB-2	77.4	18.20	
12 ¹¹	PM6	Y6	TTBT-R	76.1	18.07	

1312	PM6	Y6	pPh2F	77.29	18.40
14 ¹³	PM6	BTP-eC9	BPR-SC1	77.6	18.02
15 ¹⁴	PM6	BTP-eC9	Z2	77.8	18.12
16 ¹⁵	PM6	BTP-eC9	ADA	77.3	18.32
17 ¹⁶	PM6	BTP-eC9	BTTC	79.40	19.18
18 ¹⁷	PM6	L8-BO	BTC	76.2	18.41
19 ¹⁸	PM6	L8-BO	BTID-2F	77.5	18.52
20 ¹⁹	PM6	L8-BO	BT-ER	74.7	18.11
21 ²⁰	PM6	L8-BO	aBTC1	79.2	18.96
22 ²⁰	PM6	L8-BO	βBTC1	77.1	18.55
23 ²¹	PM6	L8-BO	SeDZ-3TR	78.55	18.59
24 ¹⁶	PM6	L8-BO	BTTC	78.42	18.80
25 ²²	PM6	L8-BO	SD-T	78.8	18.7
26 ²²	PM6	L8-BO	SD-EDOT	81.5	19.3
This work	PM6	L8-BO	BTR-SCI	81.5	19.4
This work	D18	L8-BO	BTR-SCI	80.9	20.0

Table S3 Exciton dissociation efficiency (P_{diss}) and charge collection efficiency (P_{coll}) of devices with different structures.

Blends	P _{diss}	P _{coll}
PM6:L8-BO	98.6%	91.4%
PM6:BTR-SCI:L8-BO	99.8%	93.6%
BTR-SCI:L8-BO	88.4%	61.9%

Blends	$\mu_{\rm h}$ (cm ² V ⁻¹ s ⁻¹)	μ _e (cm ² V ⁻¹ s ⁻¹)	$\mu_{\rm h/}\mu_{\rm e}$
PM6:L8-BO	7.26×10^{-4}	6.54×10^{-4}	1.11
PM6:BTR-SCI:L8-BO	1.07×10^{-3}	1.04×10^{-3}	1.03
BTR-SCI:L8-BO	3.23×10^{-4}	2.29×10^{-4}	1.41

Table S4 The hole mobilities (μ_h) and electron mobilities (μ_e) of the devices measured by the SCLC method.

Table S5 Detailed voltage loss parameters of the PSCs.

Active layer	Eg (eV)	qV _{oc} (eV)	E _{loss} (eV)	Δ <i>E</i> ₁ (eV)	$\frac{\Delta E_2}{(\text{eV})}$	EQE _{EL} (%)	Δ <i>E</i> ₃ a (eV)
PM6:L8-BO	1.441	0.880	0.561	0.267	0.054	9.40×10 ⁻³	0.240
PM6:BTR- SCl:L8-BO	1.446	0.890	0.556	0.268	0.058	1.40×10 ⁻²	0.230

 $\Delta E_{3^{a}}$ is calculated from the EQE_{EL} measurements.

Table S6 Key parameters of contact angle measurements by using water and diiodomethane (DIM).

	Contact	rt angels		4.0	
Sample	θ _{water} (°)	θ _{DIM} (°)	$(\mathbf{mN} \mathbf{m}^{-1})$	χ^{A-D} $\chi^{D} 1^{-D} 2$ 0.39 κ / / /	χ^{ν} 1 ^{-ν} 2
PM6	101	55	32.05	0.39 к	/
L8-BO	97	42	39.53	/	/
BTR-SCI	98	54	32.28	0.37 ĸ	0.0004 κ

Sample	Pe	ak	q (Å ⁻¹)	<i>d</i> -spacing (Å)	CCL (Å)
PM6	IP	(100)	0.29	21.30	81.54
	OOP	(010)	qd-spacing $(Å^{-1})$ $(Å)$ 0.2921.301.693.710.4812.951.813.470.3219.571.853.400.3120.201.803.480.3219.511.823.450.3219.511.833.430.3418.591.773.55	20.59	
I 8-RO	IP	(100)	0.48	12.95	88.70
	OOP	(010)	1.81	3.47	14.52
RTR-SCI	IP	(100)	0.32	19.57	127.94
DTR-5C1 _	OOP	(010)	1.85	3.40	27.40
PM6·RTR·SCI	IP	(100)	0.31	20.20	84.39
	OOP	(010)	1.80	3.48	21.45
PM6·I 8-BO	IP	(100)	0.32	19.51	107.42
	OOP	(010)	1.82	3.45	21.57
PM6-RTD SCI-LS RO	IP	(100)	0.32	19.51	93.42
1 WIO.DI K-SCI.LO-DO _	OOP	(010)	1.83	3.43	22.15
RTR-SCI-L& RO	IP	(100)	0.34	18.59	69.96
DIR-5CI.L0-D0 –	OOP	(010)	1.77	3.55	14.82

Table S7 Structure parameters of the various films obtained from GIWAXS data.

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