Simultaneously improved efficiency and average visible transmittance of semitransparent polymer solar cells with two ultranarrow bandgap nonfullerene acceptors

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Experimental section

Device Fabrication: The patterned indium tin oxide (ITO) coated on glass substrates (15 Ω per square) were cleaned via sequential sonication in detergent, de-ionized and ethanol and then blow-dried by high-purity nitrogen. All pre-cleaned ITO substrates were treated by oxygen plasma for 1 minute to improve their work function and clearance. The Zinc oxide (ZnO) solution was spin coated on ITO substrates at 4000 rounds per minute (RPM) for 30 s and annealed at 200°C for 1 hour in atmospheric air. Then ITO substrates coated with ZnO layer were transferred into a high-purity nitrogenfilled glovebox. The polymer donor PTB7-Th was purchased from 1-Materials Inc. The acceptor CO_i8DFIC was synthesized by Ding et al. from National Center for Nanoscience and Technology. The acceptor IEICO-4F was purchased from Organtecsolar Materials Inc. The PTB7-Th: COi8DFIC and PTB7-Th: IEICO-4F with 1:1.5 weight ratio were dissolved in chlorobenzene (adding 1.0% v/v 1,8-diiodooctane, DIO) to prepare 25 mg/mL binary blend solutions, respectively. The proportions of CO_i8DFIC: IEICO-4F are 1:0, 0.95:0.05, 0.85:0.15, 0.7:0.3, 0.5:0.5, 0:1. The mixed solutions were spin-coated onto the ZnO/ITO substrates at 2000 RPM for 40 s to prepare the active layers. A thin molybdenum trioxide (MoO₃) layer (10 nm) and a silver (Ag) layer were sequentially deposited by thermal evaporation. For opaque PSCs, the Ag thickness is appropriate 100 nm. For semitransparent PSCs, the Ag thickness was 10 nm, 15 nm, or 20 nm. The active area is approximate 3.8 mm², which is defined by the overlapping area of ITO anode and Ag cathode.

Device Measurement: The current density-voltage (*J-V*) curves of all the polymer solar cells (PSCs) were measured by a Keithley 2400 unit in high-purity nitrogen-filled glovebox. The AM 1.5G irradiation was provided by an XES-40S2 (SAN-EI ELECTRIC Co., Ltd) solar simulator (AAA grade, 70×70 mm² photobeam size) with light intensity of 100 mW cm⁻². The external quantum efficiency (EQE) spectra of PSCs were measured by a Zolix Solar Cell Scan 100. The absorption spectra of films were measured with a Shimadzu UV-3101 PC spectrometer. Grazing incidence X-ray diffraction (GIXD) images were measured by a 5-circle Huber diffractometer at the Beijing Synchrotron Radiation Facility (BSRF). A bent triangle silicon crystal was used

to select the X-rays of a wavelength of 1.54 Å. A grazing incidence angle of 0.4° was chosen to increase GIXD peak intensity for investigating the crystallinity and orientation that prevail throughout the active layers. Transmission electron microscopy (TEM) images of blend films were obtained by a JEOL JEM-1400 transmission electron microscope operated at 80 kV. The hole and electron mobility were calculated according to the space charge limited current (SCLC) method. The charge mobility is generally described by the Mott-Gurney equation:

$$J=\frac{9}{8}\,\varepsilon_0\varepsilon_r\mu\frac{V^2}{L^3}$$

Where *J* is the current density, ε_0 is the permittivity of free space (8.85×10⁻¹⁴ F/cm), ε_r is the dielectric constant of used materials which is assumed to be 3, a typical value for organic materials, μ is the charge mobility, *V* is the applied voltage and *L* is the active layer thickness. The hole-only and electron-only devices were fabricated with the configurations of ITO/PEDOT:PSS/active layers/MoO₃/Ag and ITO/ZnO/active layers/PDIN/Al, respectively.



Fig. S1 The reflection spectra of PSCs with different IEICO-4F content and a special device ITO/ZnO/PMMA (~120 nm)/MoO₃/Ag.

The reflection measurement of all devices was performed on a commercial QE measurement system (QE-RT3011, Enlitech) by using an integrating sphere. The absorption spectra of active layers were calculated by subtracting the parasitic absorptions $(1-R_1)$ from the total absorption in PSCs $(1-R_2)$, where R₁ is the reflection spectrum of device ITO/ZnO/PMMA (~120 nm)/MoO₃/Ag, R₂ is the reflection spectra of PSCs ITO/ZnO/active layers (~120 nm)/MoO₃/Ag. The 120 nm PMMA layer in the

special cell is used to simulate the optical path in real PSCs because PMMA has negligible photon harvesting in the whole spectral range, and the light absorption of glass/ITO/ZnO layers in real PSCs can be obtained from this special cell.



Fig. S2 The *J-V* curves of the PSCs with 0 wt% (a), 15 wt% (b), and 100 wt% (c) IEICO-4F content in acceptors under different light illumination intensity, obtained from standard AM $1.5G (100 \text{ mW cm}^{-2})$ illumination using a set of neutral optical filters.



Fig. S3 The plotted $\ln(Jd^3/V^2)$ versus $(V/d)^{0.5}$ curves of the devices with different

IEICO-4F content.

IEICO-4F content (wt %)	μ_h (cm ² V ⁻¹ s ⁻¹)	μ_e (cm ² V ⁻¹ s ⁻¹)	μ _h /μ _e
0	7.77×10 ⁻⁴	4.81×10 ⁻⁴	1.62
5	1.63×10 ⁻³	1.06×10 ⁻³	1.54
15	1.82×10 ⁻³	1.43×10 ⁻³	1.27
30	1.44×10 ⁻³	9.20×10 ⁻⁴	1.57
50	1.19×10 ⁻³	7.31×10 ⁻⁴	1.63
100	9.87×10 ⁻⁴	5.99×10 ⁻⁴	1.65

Table S1 Hole, electron mobility, and μ_h/μ_e in PSCs with different IEICO-4F content.



Fig. S4 In-plane (a) and out-of-plane (b) GIXD profiles for neat films.



Fig. S5 Transmission spectra of Ag thin films with different thickness.



Fig. S6 *J-V* curves of semitransparent binary and ternary PSCs with 10 nm (a), 15 nm (b), and 20 nm (c) Ag as electrode.



Fig. S7 The sheet resistance of Ag films with different thickness.



Fig. S8 Transmission spectra of (a) PTB7-Th: COi8DFIC, (b) PTB7-Th: COi8DFIC: IEICO-4F (15 wt% IEICO-4F in acceptors), and (c) PTB7-Th: IEICO-4F based active layers and semitransparent PSCs with different Ag thickness.