Supporting Information

Semitransparent polymer solar cells with 9.06% efficiency and 27.1% average visible transmittance by employing a smart strategy

Zhenghao Hu[a], Zhi Wang[a] and Fujun Zhang*[a]

[a]Key Laboratory of Luminescence and Optical Information, Ministry of Education, Beijing Jiaotong University, Beijing 100044, P. R. China

E-mail: fjzhang@bjtu.edu.cn (Fujun)

1. Detailed experimental section

1.1. Cell fabrication

The patterned indium tin oxide (ITO) coated glass substrates (sheet resistance 15 $\Omega/\square$) were consecutively cleaned in ultrasonic baths containing detergent, de-ionized water and ethanol, respectively. The cleaned ITO substrates were blow-dried by high pure nitrogen gas and then treated by oxygen plasma for 1 min to improve its work function and clearance. Subsequently, PEDOT:PSS thin films were fabricated on the cleaned ITO substrates by spin-coating method at 5000 round per minute (RPM) for 30 s, and then annealed at 150 °C for 10 minutes in atmospheric air. After annealing treatment, ITO substrates coated with PEDOT:PSS films were transferred to a high-purity nitrogen-filled glove box to prepare active layers. The PTB7-Th and IEICO-4F were dissolved in C_6H_5Cl to prepare 30 mg/ml solutions, respectively, which were mixed by different volume ratios to obtain binary blend solutions of PTB7-Th$_x$: IEICO-4F$_{1.5}$ (x=1.5, 1.4, 1.3, 1.2, 1.1, 1.0, 0.9, 0.8, 0.7, 0.6, 0.5, x represents PTB7-Th content ratios in binary blend solutions, the IEICO-4F content is 15mg/ml). 2 vol% 1-chloronaphthalene (CN) were added to the prepared binary blend solutions. Subsequently, the prepared binary blend solutions were spin-coated on PEDOT:PSS modified ITO substrates at different RPM for same active layers thickness in high-purity nitrogen-filled glove box. The prepared active layers were suffered from thermal

annealing at 100 ℃ for 10 min (TA). Afterwards, a PDIN cathode interlayer was spin-coated onto active layers at 5000 RPM for 30 s. Finally, 100 nm Al or 1 nm Au/11 nm Ag electrode were deposited by thermal evaporation under vacuum (10^{-5} Pa) with a shadow mask. The effective area of cell is ~3.8 mm^2, which is defined by the vertical overlap of ITO and Ag electrodes. Opaque or semitransparent polymer solar cells (PSCs) are fabricated with the structure of ITO/PEDOT:PSS/active layer/PDIN/Al(100 nm) or ITO/PEDOT:PSS/active layer/PDIN/Au(1 nm)/Ag(11 nm), respectively.

1.2. General characterizations

The current-voltage (J-V) characteristics of all PSCs were measured in high-purity nitrogen-filled glove box using Keithley 2400 source meter. AM 1.5G irradiation at 100 mW/cm^2 was provided by An XES-40S2 (SAN-EI Electric Co. Ltd.) solar simulator (AAA grade, 70 × 70 mm^2 photo beam size), which was calibrated by standard silicon solar cells (purchased from Zolix INSTRUMENTS CO., LTD). The external quantum efficiency (EQE) spectra of PSCs were measured in air conditions by a Zolix Solar Cell Scan 100. The ultraviolet-visible (UV-Vis) absorption spectra of neat and blend films, transmission spectra of glass/ITO substrate, active layers with anode buffer layer, 1nm Au/11nm Ag ultrathin electrode and semitransparent PSCs were obtained using a Shimadzu UV-3101 PC spectrometer. The thickness of active layers was measured by Ambios Technology XP-2 stylus Profiler. Transmission electron microscopy (TEM) images of blend films were obtained by using a JEOL JEM-1400 transmission electron microscope operated at 80 kV. The Comission Internationale de I'Eclairage (CIE) 1931 color coordinate of the transmitted light were obtained by using PR-655 (Photo Research).

1.3. J_{ph}-V_{eff}

Here, the J_{ph} is defined as $J_{ph} = J_L$ - J_D, where J_{ph} is photogenerated current density,

J_L and J_D represent the current density under light illumination or in dark, respectively. The V_{eff} is defined as $V_{eff} = V_0 - V$, V_0 is the voltage at where $J_{ph} = 0$, V is the applied bias. Assuming that all excitons can be dissociated into free charge carriers and the photogenerated charge carriers can be efficiently collected by the individual electrode at a high V_{eff} of 2.5 V. The saturation current density (J_{sat}) should be mainly determined by the harvested photons of active layers (J_{sat} = qLG$_{max}$, where q is elementary charge and L is the active layer thickness, G_{max} is the maximal exciton generation rate). The exciton dissociation efficiency (η_{diss}) and charge collection efficiency (η_{coll}) can be evaluated by the J_{ph}/J_{sat} values at short circuit condition or maximal power output condition, respectively.

1.4. Charge mobility measurement

Electron-only or hole-only devices were fabricated with the structure of ITO/ZnO/active layers/Al or ITO/PEDOT:PSS/active layers/MoO$_3$/Ag, respectively. The active layers were prepared under the same conditions compared with those in PSCs. The charge mobility was extracted by fitting the current density-voltage curves using space charge limited current methods (SCLC). The charge mobility is described by the Mott-Gurney equation:

$$J = \frac{8}{9}\varepsilon_r\varepsilon_0\mu\frac{v^2}{d}exp[0.89\gamma\sqrt{\frac{v}{d}}]$$

Here, ε_r is dielectric constant of organic materials, ε_0 is the free space permittivity, μ is charge mobility, V is the applied voltage and d is the active layer thickness (~100 nm).

2. Additional experimental results
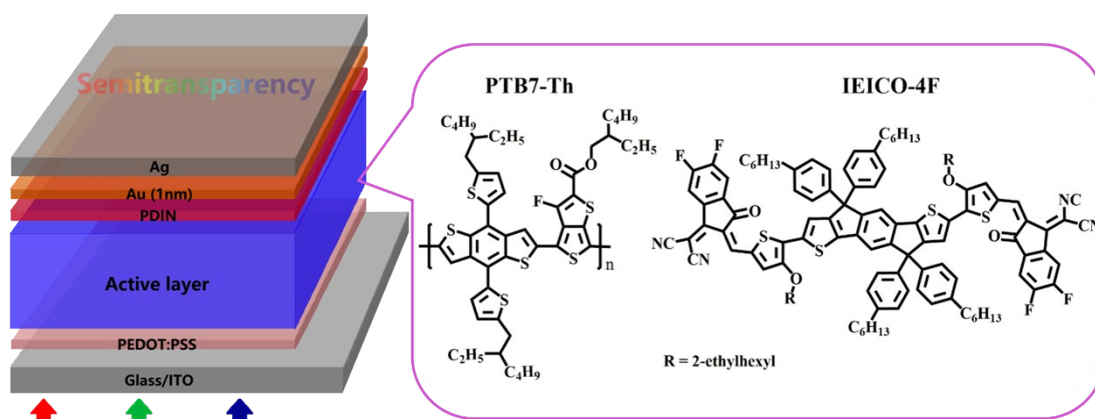
Fig. S1 Schematic diagram of semitransparent PSCs and chemical structures of used materials.

Table S1 J_{ph}, J_{sat} and J_{ph}/J_{sat} values of typical PSCs.

D:A ratios	J_{ph}^{a} [mA/cm²]	J_{ph}^{b} [mA/cm²]	J_{sat} [mA/cm²]	J_{ph}^{a}/J_{sat} [%]	J_{ph}^{b}/J_{sat} [%]
1.5:1.5	24.03	19.29	26.04	92.28	74.08
1.2:1.5	24.58	20.24	26.81	91.68	75.49
1.0:1.5	23.60	19.84	25.89	91.15	76.63
0.8:1.5	22.48	19.15	24.80	90.65	77.22
0.5:1.5	18.62	15.91	20.70	89.95	76.85

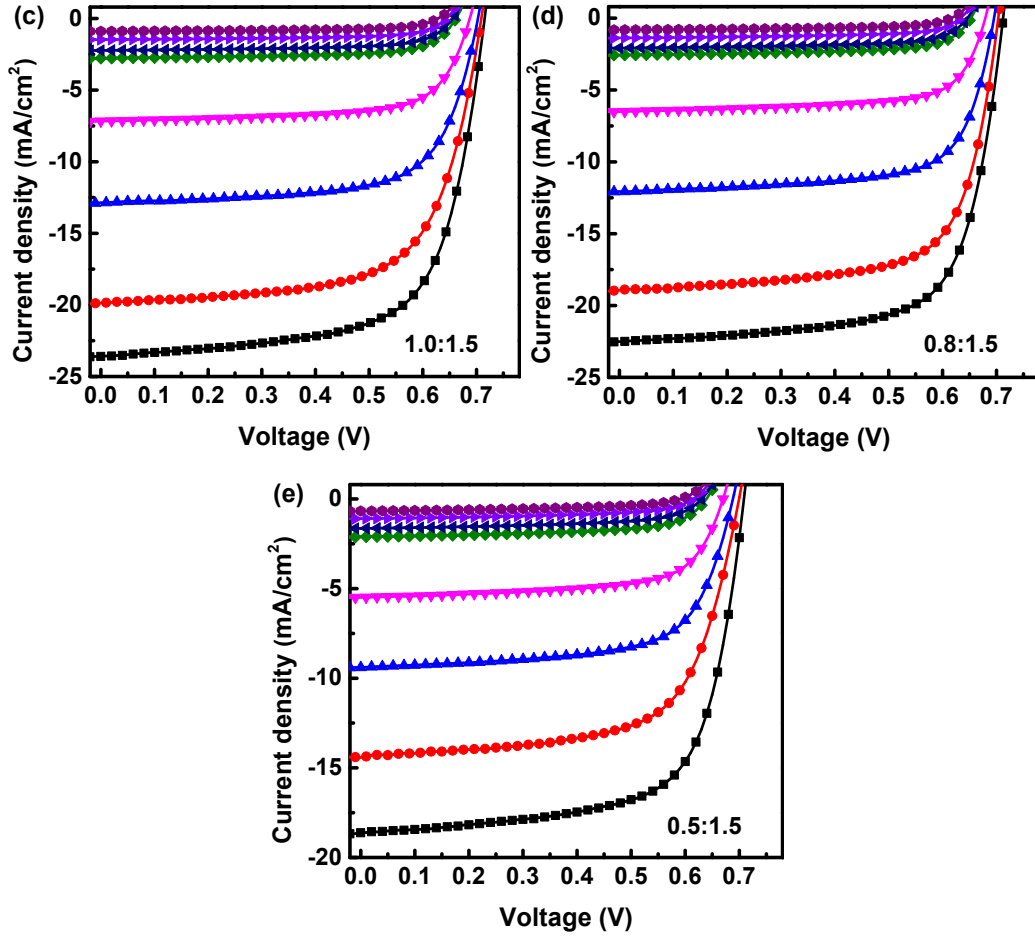[a] short-circuit condition, [b] maximal power output condition.
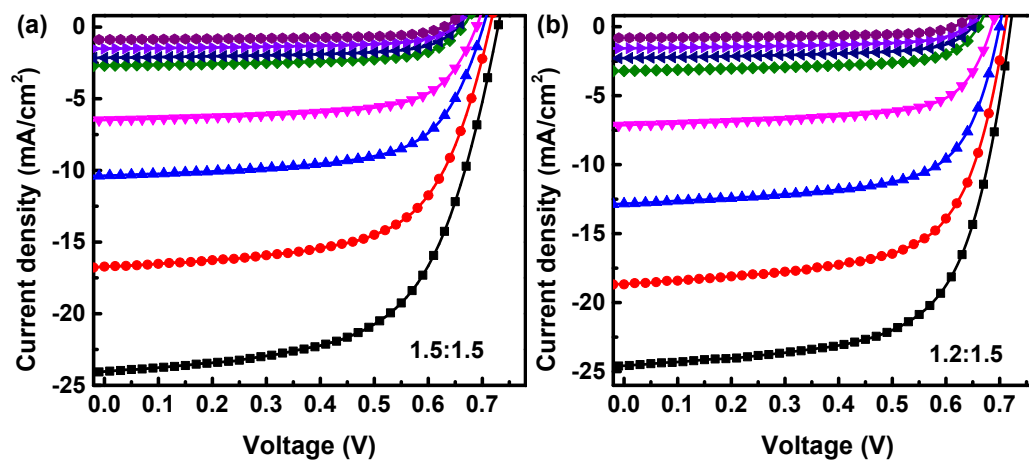
Fig. S2 *J-V* curves of PSCs with different D:A ratios under AM 1.5G illumination with light intensity of 100, 80, 50, 25, 10, 8, 5 or 2.5 mW/cm^2, respectively.
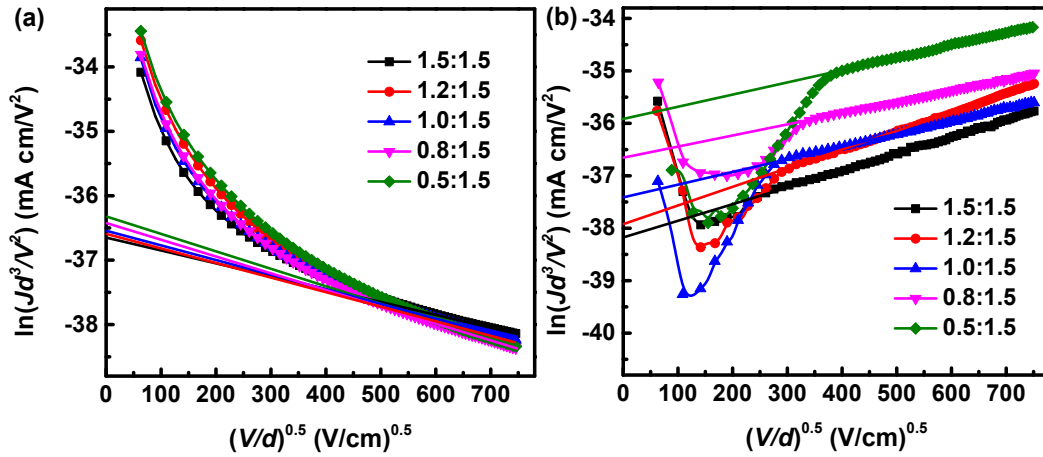

Fig. S3 The *ln(Jd^3/V^2)* versus *(V/d)$^{0.5}$* curves of (a) hole-only devices, (b) electron-only devices.

The relationship of AVT values can be described as:

$$AVT_{sub} \times AVT_{alb} \times AVT_{ele} = AVT_{ST-PSCs} \tag{1}$$

Where the AVT_{sub} is AVT value of glass/ITO substrate, AVT_{alb} is AVT value of active layer with buffer layers, AVT_{ele} is AVT value of Au/Ag electrode and $AVT_{ST\text{-}PSCs}$ is AVT value of semitransparent PSCs. According to the formula (1), the calculated $AVT_{ST\text{-}PSCs}$ are summarized in **Table S2**.

Table S2 The AVT values of glass/ITO substrate, 1 nm Au/11 nm Ag ultrathin electrode, active layers with buffer layers, measured semitransparent PSCs and calculated semitransparent PSCs.

	AVT (%)
Glass/ITO substrate	91.7
Au/Ag electrode	51.4
Active layers	
D:A =1.2:1.5	52.2
D:A =1.0:1.5	57.6
D:A =0.8:1.5	60.0
Measured PSCs	
D:A =1.2:1.5	23.7
D:A =1.0:1.5	25.7
D:A =0.8:1.5	27.1
Calculated PSCs	
D:A =1.2:1.5	24.6
D:A =1.0:1.5	27.1
D:A =0.8:1.5	28.3

An objective parameter quantum utilization efficiency (QUE) was proposed to evaluate the PCE and AVT of semitransparent PSCs, which is defined as the sum of EQE and transmittance (denoted as T) of semitransparent PSCs. The QUE is described as:

$$QUE = EQE + T \tag{2}$$

Table S3 The coloring rendering index (CRI) and correlated color temperature (CCT) of the semitransparent PSCs.

D:A ratios	CIE(x, y)	CCT(K)
1.2:1.5	(0.2650, 0.2996)	11053
1.0:1.5	(0.2680, 0.2990)	10746
0.8:1.5	(0.2694, 0.2920)	11073