Supporting Information

Semitransparent organic solar cells exhibiting 13.02% efficiency and 20.2% average visible transmittance

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1. Detailed experimental section

1.1 Cell fabrication

The used D18-Cl and Y6-1O were designed and synthesized by He Yan's group in Hong Kong University of Science and Technology, the two materials were purchased from the eFlexPV limited company owned by Yan. The Y6 was purchased from Solarmer Materials Inc. The D18-Cl and Y6-1O were dissolved in chloroform to prepare 11 mg/ml and 16 mg/ml solutions, respectively, which were mixed by different volume ratios to obtain binary blend solutions of D18-Cl_x: Y6-$1O_{1.6}$ (x=1.1, 0.9, 0.7, 0.5, 0.3, 0.1, x represents D18-Cl content ratios in binary blend solutions), the Y6-1O content is maintained at 8mg/ml. The D18-Cl and Y6 with 0.7:1.6 weight ratio was dissolved in chloroform to prepare 11.5 mg/ml binary blend solution. The ternary blend solutions of D18-Cl:Y6-$1O_{1-n}$:$Y6_n$ (n=10, 30, 50, 70 wt%, the n represents Y6 content in acceptors) were obtained by mixing optimized binary blend solution with different volume ratios. N,N'-bis(3-(dimethylamino)propyl)perylene-3,4,9,10-tetracarboxylic diimide (PDIN, purchased from Solarmer Materials Inc.) was dissolved in methanol with addition of 0.25 vol% acetic acid to prepare 2 mg/ml solution.

The patterned indium tin oxide (ITO) coated glass substrates (sheet resistance 15 Ω/sq) were consecutively cleaned in ultra-sonic baths containing detergent, de-ionized

water, ethanol, respectively. The cleaned ITO substrates were blow-dried by high-purity 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 rounds per minute (RPM) for 30 s, and then annealed at 150 ℃ for 10 min under atmospheric condition. PEDOT:PSS solution was purchased from H.C. Starck co. Ltd. The ITO substrates coated with PEDOT:PSS films were transferred to a high-purity nitrogen-filled glove box to prepare active layers. The prepared binary and ternary blend solutions were spin-coated on PEDOT:PSS modified ITO substrates at 2300 RPM for 30s in high-purity nitrogen-filled glove box, and then the prepared active layers were annealed with CS_2 solvent vapor for 30 s. PDIN was purchased from Solarmer Materials Inc. Afterwards, the prepared PDIN solution was spin-coated onto active layers at 5000 RPM for 30 s. Finally, 100 nm Ag or Au (1 nm)/Ag (20, 15, 10 nm) 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 metal electrodes. Opaque or semitransparent organic solar cells (OSCs) are fabricated with the structure of ITO/PEDOT:PSS/active layer/PDIN/Ag (100 nm) or ITO/PEDOT:PSS/active layer/PDIN/Au(1 nm)/Ag(20, 15, 10 nm), respectively.

1.2. General characterizations

The current-voltage (*J-V*) characteristics of all OSCs 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 OSCs were measured in air conditions by a Zolix Solar Cell Scan 100. Photoluminescence (PL) spectra of neat and blend films were measured by a HORIBA Fluorolog®-3 spectrofluorometer system. The ultraviolet-visible (UV-Vis) absorption spectra of neat and blend films, transmission spectra of blend films and semitransparent PSCs

were obtained using a Shimadzu UV-3101 PC spectrometer. 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). It is highlighted that light is incident from ITO side during the measurement processes of *J-V* curves, EQE or AVT spectra.

2. Average visible light transmittance calculation

The average visible light transmittance (AVT) are calculated according to the equation:

$$AVT = \frac{\int T(\lambda) \times V(\lambda) \times AM1.5G(\lambda)\, d(\lambda)}{\int V(\lambda) \times AM1.5G(\lambda)\, d(\lambda)}$$

Where $T(\lambda)$ is the transmission spectrum, $V(\lambda)$ is the photopic response of human eye, $AM1.5G(\lambda)$ is photon flux.

Table S1 Typical progress of recent semitransparent OSCs.

Active layers	J_{sc} [mA cm^{-2}]	V_{oc} [V]	FF [%]	PCE [%]	AVT [%]	Spectrum range of AVT	Highest QUE [%]	Ref.
PBT1-C-2Cl:Y6	19.51	0.83	70.2	11.7	19.7	380~780 nm	82	*Adv. Funct. Mater.* 2020, 30, 2002181
PBDB-T:PTAA:Y1	19.7	0.86	69.1	11.7	20.1	400~700 nm	81	*Adv. Mater.* 2020, 32, 2003891
PTB7-Th:IEICO-4F	22.5	0.720	69.3	11.25	21.5	380~780 nm	83	*Joule* 2019, 3, 2241
PM6:Y6:DTNIF	22.71	0.847	70.16	13.49	22.58	380~780 nm	95	*Energy Environ. Sci.* 2020, 13, 5177
D18-Cl:Y6-1O:Y6	19.56	0.884	75.31	13.02	20.2	370~740 nm	82	*This work*

3. Additional experimental results

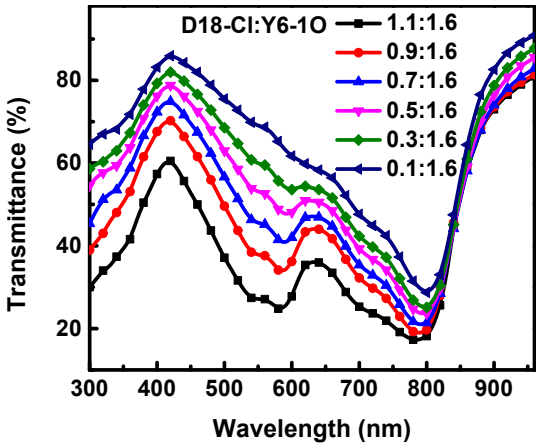

Fig. S1 Transmission spectra of blend films with distinct D18-Cl:Y6-1O weight ratios.

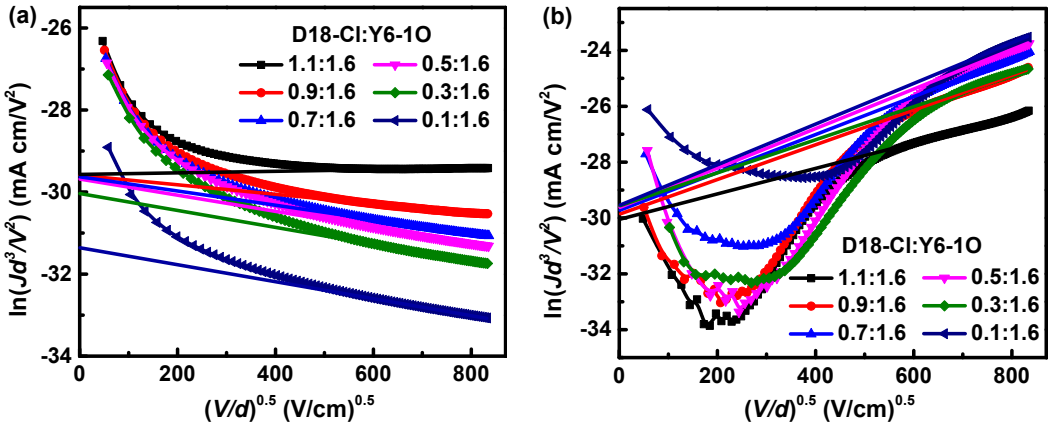

Fig. S2 The $ln(Jd^3/V^2)$ versus $(V/d)^{0.5}$ curves of (a) hole-only cells, (b) electron-only cells.

The charge mobility is described by the Mott-Gurney equation:

$$J = \frac{8}{9}\varepsilon_r\varepsilon_0\mu\frac{v^2}{d^3}exp[0.89\gamma\sqrt{\frac{v}{d}}]$$

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.

Table S2 Hole mobility (μ_h), electron mobility (μ_e) and their ratios (μ_h/μ_e) of the opaque OSCs with distinct D18-Cl:Y6-1O weight ratios.

D18-Cl:Y6-1O ratios	μ_h ($cm^2\ v^{-1}\ s^{-1}$)	μ_e ($cm^2\ v^{-1}\ s^{-1}$)	μ_h/μ_e
1.1:1.6	4.82×10^{-4}	3.01×10^{-4}	1.60

0.9:1.6	4.70×10^{-4}	3.68×10^{-4}	1.28
0.7:1.6	4.54×10^{-4}	4.23×10^{-4}	1.07
0.5:1.6	4.26×10^{-4}	4.63×10^{-4}	0.92
0.3:1.6	3.01×10^{-4}	4.91×10^{-4}	0.61
0.1:1.6	8.04×10^{-5}	5.01×10^{-4}	0.16

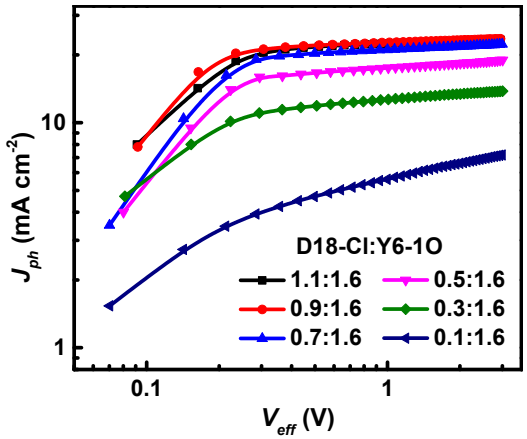

Fig. S3 (a) J_{ph}-V_{eff} curves of opaque OSCs with distinct D18-Cl:Y6-1O weight ratios.

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 effective applied voltage (V_{eff}) is defined as $V_{eff} = V_0 - V$, V_0 is the voltage at where $J_{ph} = 0$, V is the applied voltage. 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 V_{eff} of 2.5 V. The saturation current density (J_{sat}) should be mainly determined by the harvested photons of active layers. 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.

Table S3 J_{ph}, J_{sat} and J_{ph}/J_{sat} values of opaque OSCs with distinct D18-Cl:Y6-1O weight ratios.

D18-Cl:Y6-1O	J_{ph}^a [mA/cm^2]	J_{ph}^b [mA/cm^2]	J_{sat} [mA/cm^2]	J_{ph}^a/J_{sat} [%]	J_{ph}^b/J_{sat} [%]
1.1:1.6	22.49	19.35	23.20	96.94	83.41

0.9:1.6	22.74	20.01	23.50	96.77	85.15
0.7:1.6	21.17	18.91	22.06	95.97	85.72
0.5:1.6	17.59	15.90	18.65	94.32	85.25
0.3:1.6	12.77	11.11	13.65	93.55	81.39
0.1:1.6	5.76	4.37	6.93	83.12	63.06

[a] short-circuit condition, [b] maximal power output condition.

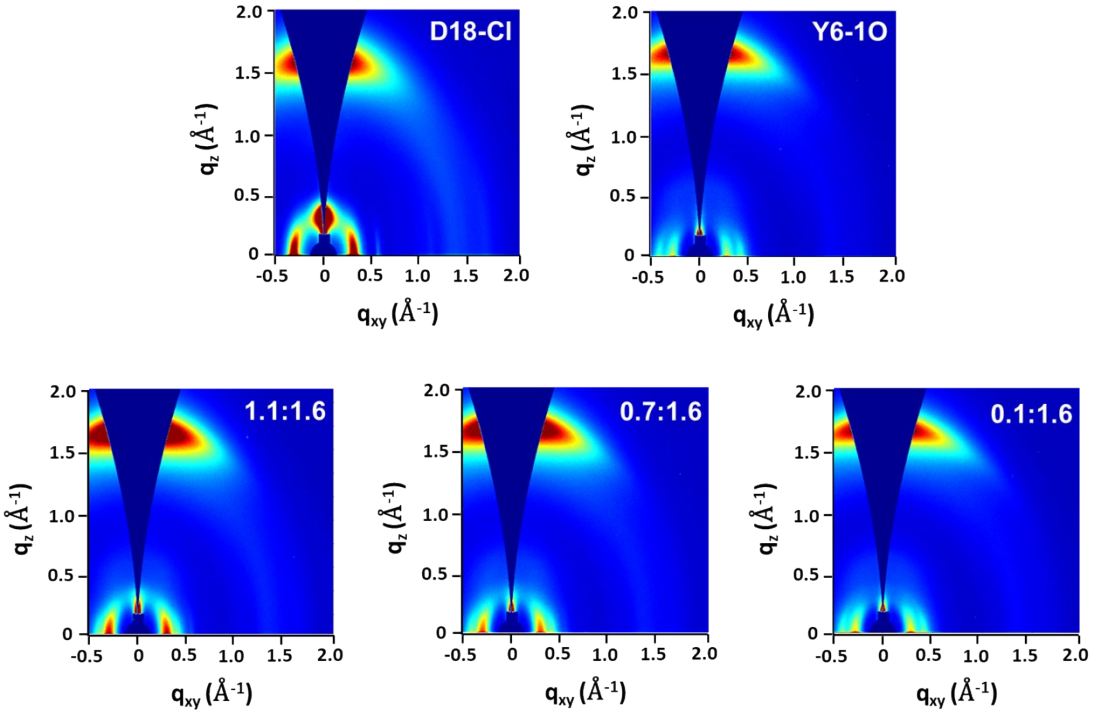

Fig. S4 2D GIWAXS patterns of a) neat D18-Cl, Y6-1O films and b) their blend films with typical ratios of 1.1:1.6, 0.7:1.6 and 0.1:1.6 (wt/wt).

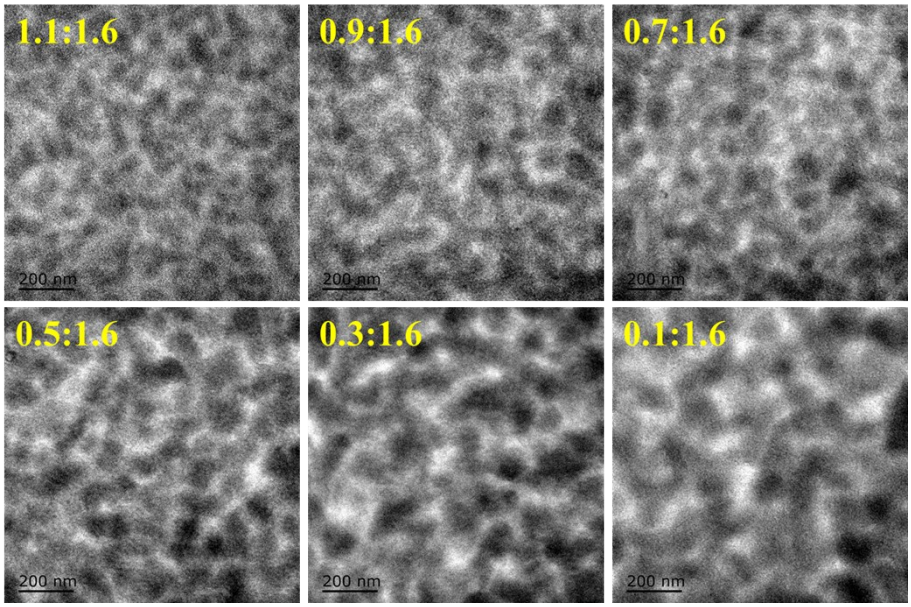

Fig. S5 TEM images of blend films with distinct D18-Cl:Y6-1O weight ratios.

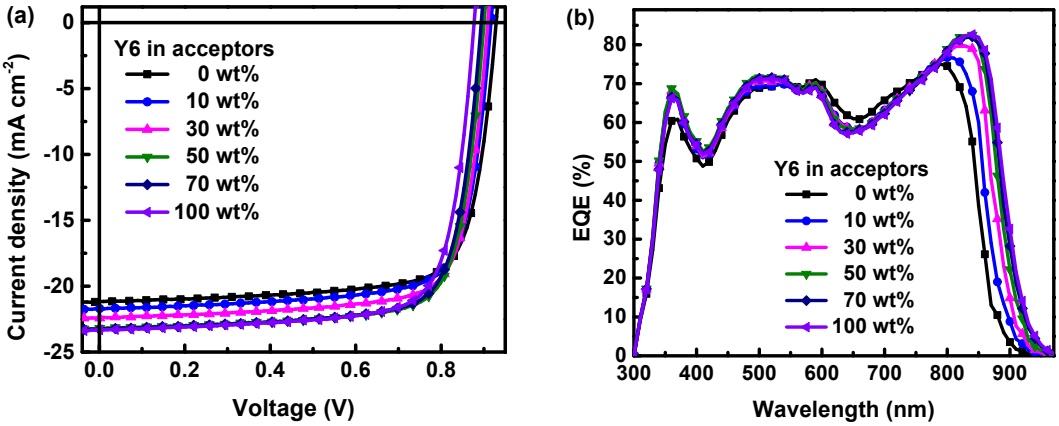

Fig. S6 The *J-V* curves (a) and EQE spectra (b) of OSCs with distinct Y6 content in acceptors.

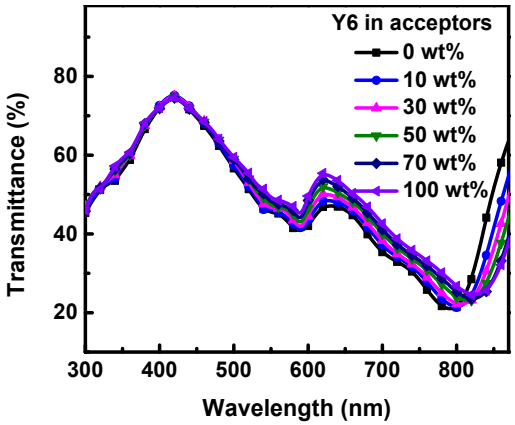

Fig. S7 Transmission spectra of the blend films with distinct Y6 content in acceptors.