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Supporting Information

High-performance nonfullerene polymer solar cells based on fluorinated wide bandgap copolymer with a high open-circuit voltage of 1.04 V

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Materials

All chemicals and solvents were reagent grades and purchased from Alfa Aesar and TCI. ITIC was purchased from Solarmer Materials Inc. PM6 was synthesized according to the procedure reported in the literatures.[1]

Experimental Section

Measurements: UV-vis absorption spectra were taken on an Agilent Technologies Cary Series UV-Vis-NIR Spectrophotometer. Photoluminescence (PL) spectra were taken on an Edinburgh Instrument FLS 980. Atomic force microscopy (AFM) measurements were performed on a Dimension 3100 (Veeco) Atomic Force Microscope in the tapping mode. Transmission electron microscopy (TEM) was performed using a Tecnai G2 F20 S-TWIN instrument at 200 kV accelerating voltage. TEM was performed using a

Tecnai G2 F20 S-TWIN instrument at 200 kV accelerating voltage, in which the blend films were prepared using a processing technique, as following: first, the blend films were spin-cast on the PEDOT:PSS/ITO substrates; second, the resulting blend film/PEDOT:PSS/ITO substrates were submerged in deionized water to make these blend films float onto the air-water interface; finally, the floated blend films were taken up on unsupported 200 mesh copper grids for a TEM measurement.

Fabrication and characterization of polymer solar cells. Polymer solar cells with a conventional device structure of ITO/ZnO/PM6:ITIC/MoO_3/Al were fabricated under conditions as follows: patterned indium tin oxide (ITO)-coated glass with a sheet resistance of 10-15 ohm/square was cleaned by a surfactant scrub and then underwent a wet-cleaning process inside an ultrasonic bath, beginning with deionized water followed by acetone and isopropanol. After oxygen plasma cleaning for 10 min, then the ZnO layer with a thickness of 30 nm was deposited by spin-coating under 5000 rpm for 60 s on top of the ITO substrate and then dried by baking in the titanium plate oven at 200 ^{o}C for 1 h. The ZnO precursor was prepared by dissolving zinc acetate dihydrate (Sigma-Aldrich, 99.999%, 1 g) and ethanolamine (Sigma-Aldrich, 99.5%, 0.28 g) in 2-methoxyethanol (Sigma-Aldrich, 99.3%, 10 mL) under vigorous stirring at 60 ^{o}C for 12 h for the hydrolysis reaction in argon. The active layer was then deposited on top of the ZnO layer by spin-coating a chlorobenezene (CB) solution of PM6:ITIC. The thicknesses of the active layers are 100-110 nm and measured on a KLA Tencor D-100 profilometer. Finally, 10 nm MoO_3 and 80 nm Al were successively deposited on the photosensitive layer under vacuum at a pressure of ca. 4×10^{-4} Pa, and through a shadow mask to determine the active area of the devices (~4 × 5 mm^2). The PCE values of the PSCs were measured under a illumination of AM 1.5G (100 mW/cm^2) using a SS-F5-3A solar simulator (AAA grade, 50 × 50 mm^2 photobeam size) of Enli Technology CO. Ltd. A 2 × 2 cm^2 monocrystalline silicon reference cell (SRC-00019) was purchased from Enli Technology CO. Ltd. Mask made by laser beam cutting technology with a defined area of 9 mm^2 was used to determine the effective area for accurate measurement. All the measurements with mask or without mask gave consistent results with relative errors within 2%. PCE statistics were obtained using 20 individual devices fabricated under the same conditions. The EQE was measured by Solar Cell Spectral Response Measurement System QE-R3011 of Enli Technology CO., Ltd. The light intensity at each wavelength was calibrated with a standard single crystal Si photovoltaic cell.

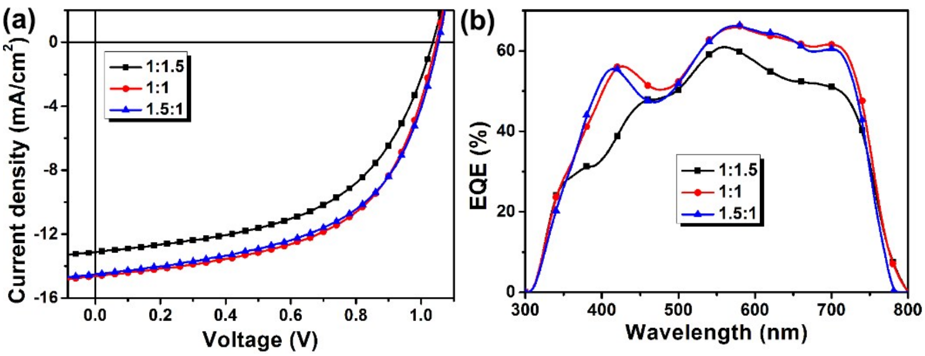

Fig. S1. (a) The *J-V* curves under the illumination of AM 1.5G, 100 mW cm^{-2}, and (b) EQE curves of the PSCs based on PM6:ITIC with the different D/A ratios.

Table S1. Photovoltaic performances of the PSCs based on PM6:ITIC with the different D/A ratios under the illumination of AM 1.5G, 100 mW cm^{-2}.

D/A	V_{oc} (V)	J_{sc} [$J_{sc}{}^{a}$] (mA cm^{-2})	FF (%)	PCE [PCE$_{ave}{}^{b}$] (%)

1:1.5	1.03	13.1 [12.4]	53	7.2 [7.0]
1:1	1.04	14.7 [14.2]	56	8.6 [8.4]
1.5:1	1.05	14.5 [13.9]	55	8.3 [8.2]

[a] The J_{sc} values calculated from the EQE curves. [b] The average PCEs were gained from 20 devices and showed in parentheses.

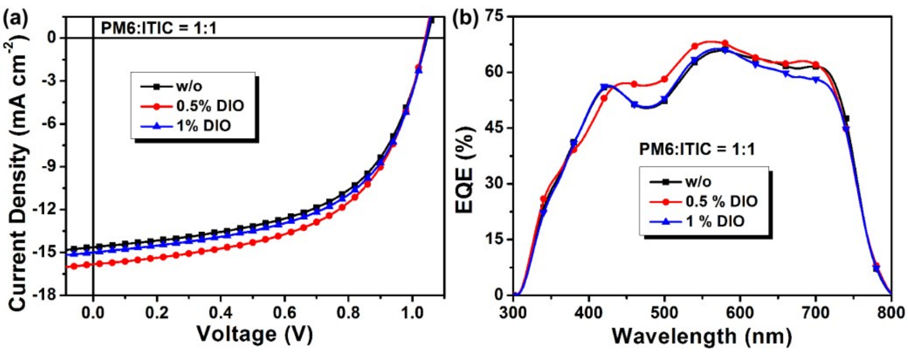

Fig. S2. (a) The *J-V* curves under the illumination of AM 1.5G, 100 mW cm^{-2}, and (b) EQE curves of the PSCs based on PM6:ITIC (1:1, w/w) with the different DIO contents.

Table S2. Photovoltaic performances of the PSCs based on PM6:ITIC (1:1, w/w) with the different DIO contents under the illumination of AM 1.5G, 100 mW cm^{-2}.

DIO (v/v%)	V_{oc} (V)	J_{sc} [J_{sc}^{a}] (mA cm^{-2})	FF (%)	PCE [PCE_{ave}^{b}] (%)
w/o	1.04	14.7 [14.2]	56	8.6 [8.4]
0.5	1.04	15.8 [15.0]	57	9.3 [9.2]
1	1.04	15.0 [14.4]	57	8.9 [8.7]

[a] The J_{sc} values calculated from the EQE curves. [b] The average PCEs were gained from 20 devices and showed in parentheses.

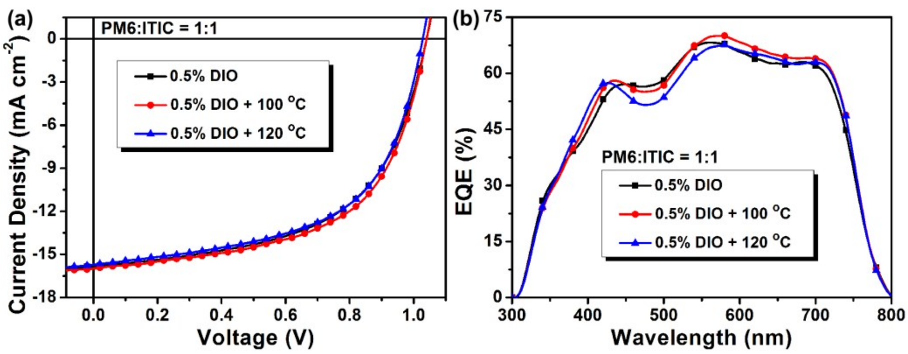

Fig. S3. (a) The *J*-*V* curves under the illumination of AM 1.5G, 100 mW cm^{-2}, and (b) EQE curves of the PSCs based on PM6:ITIC (1:1, w/w) with 0.5% DIO under different annealing temperatures.

Table S3. Photovoltaic performances of the PSCs based on PM6:ITIC (1:1, w/w) with 0.5% DIO under different annealing temperatures under the illumination of AM 1.5G, 100 mW cm^{-2}.

Temperature (°C)	V_{oc} (V)	J_{sc} [J_{sc}^{a}] (mA cm^{-2})	FF (%)	PCE [PCE$_{ave}$^{b}] (%)
w/o	1.04	15.8 [15.0]	57	9.3 [9.2]
100	1.04	16.0 [15.3]	58	9.7 [9.5]
120	1.03	15.7 [14.8]	57	9.2 [9.0]

[a] The J_{sc} values calculated from the EQE curves. [b] The average PCEs were gained from 20 devices and showed in parentheses.

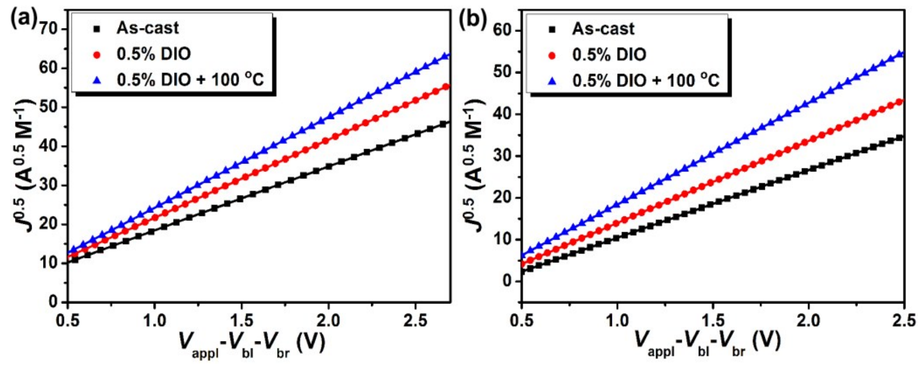

Fig. S4. The *J-V* curves of (a) the hole-only devices with the the structure of ITO/PEDOT:PSS/PM6:ITIC/MoO_3/Al; and (b) the electron-only devices with the structure of ITO/ZnO/ PM6:ITIC/Ca/Al according to the SCLC model.

Table S4. Photovoltaic results of polymer donors and NF-acceptors.

Polymer:NF-acceptor	E_g^{opt} [eV]	V_{oc} [V]	eE_g^{opt} - eV_{oc} [eV]	EQE_{max} [%]	PCE_{max} [%]	Ref.
PBPD-Th:ITIC	1.55	1.01	0.54	79	10.8	2
PBDBT:IT-M	1.60	0.94	0.66	78	12.05	3
PDBT-T1:SdiPBI-Se	1.77	0.96	0.81	73.3	8.42	4
J61:ITIC	1.57	0.89	0.68	78	9.53	5
J71:ITIC	1.59	0.94	0.65	76.5	11.41	6
PBDB-T:FDICTF	1.63	0.94	0.69	75	10.06	7
J51:IDSe-T-IC	1.52	0.91	0.61	66	8.58	8
J51:IDTIDSe-T-IC	1.52	0.91	0.61	65	8.02	9
PDBT-T1:IDIC	1.62	0.89	0.73	76	8.71	10
PTFBDT-BZS:IDIC	1.62	0.905	0.715	80	11.03	11

PDBT-T1:ITIC-Th	1.60	0.88	0.72	80	9.6	12
P3TEA:SF-PDI_2	1.72	1.11	0.61	66	9.5	13
J61:*m*-ITIC	1.58	0.912	0.668	81	11.77	14
PTB7-Th:TPB	1.61	0.79	0.82	75	8.47	15
PBDB-T:ITIC	1.57	0.90	0.67	75	11.2	16
PTB7-Th:ATT-1	1.54	0.87	0.67	74	10.07	17
PffQx-PS:ITIC	1.57	0.97	0.60	66	9.12	18
3MT-Th:ITIC	1.57	0.95	0.62	78	9.73	19
PBDB-T:FTIC-C6C8	1.63	0.93	0.70	80	11.12	20
PBDB-T:IDT-BOC6	1.63	1.01	0.62	72	9.60	21
PFBZ:ITIC	1.57	0.89	0.68	84	10.4	22
PTZ6:ITIC	1.59	1.01	0.58	62	10.3	23
PBDB-T-SF:IT-4F	1.54	0.88	0.66	83	13.1	24
PBDB-T:IT-OM-2	1.59	0.93	0.66	78	11.9	25
PBDB-T:ITCC	1.67	1.01	0.66	78	11.4	26[26]
PB3T:IT-M	1.61	1.00	0.61	82	11.9	27
PDCBT:ITIC	1.57	0.94	0.63	73	10.16	28
PBDTTT-E-T:IEICO	1.34	0.82	0.52	66	8.4	29
PBDTTT-EFT:IEICO-4F	1.24	0.739	0.501	74	10.0	30
PBQ-4F:ITIC	1.57	0.95	0.62	82	11.34	31
PBDB-T:NFBDT	1.56	0.868	0.692	75	10.42	32

HFQx:ITTC	1.61	0.88	0.73	73	10.4	33
J71:BT-IC	1.43	0.90	0.53	68	10.46	34
PBDB-T1:ITTIC	1.46	0.92	0.54	65	9.12	35
PTB7-Th:NDP-V	1.61	0.74	0.87	80	8.59	36
PvBDTTAZ:O-IDTBR	1.63	1.08	0.55	71	11.6	37
PffBT4T-B:ITIC-Th	1.60	0.972	0.628	72	9.4	38
PTzBI:ITIC	1.57	0.87	0.70	75	10.24	39
PTzBI:N2200	1.44	0.844	0.596	74	9.16	40
FTAZ:ITIC-Th1	1.55	0.849	0.701	80	12.1	41
FTAZ:INIC3	1.48	0.852	0.628	77	11.5	42
PTB7-Th:ATT-2	1.32	0.73	0.59	71	9.58	43
PBT1-EH:ITCPTC	1.58	0.95	0.63	72	11.8	44
PDBT-T1:TPH-Se	1.78	1.0	0.78	75	9.28	45
PBDTS-Se:SdiPBI-S	1.77	0.91	0.86	72	8.22	46
J51:ITIC	1.57	0.82	0.75	74	9.26	47
PBDTS-DTBTO:ITIC	1.57	0.843	0.727	78	9.09	48
PffBT4T-2DT:IDTBR	1.63	1.07	0.56	76	9.95	49[49]
PSEHTT:DBFI-EDOT	1.77	0.93	0.84	81.6	8.10	50
J51:N2200	1.48	0.83	0.65	75	8.27	51
FTAZ:ITIC2	1.53	0.925	0.605	78.2	11.0	52
PTZ1:IDIC	1.60	0.92	0.68	73.8	11.5	53

PTBTz-2:ITIC	1.56	0.888	0.672	82	10.92	54
PBDB-T:ITCPTC-Th	1.60	0.856	0.744	80	10.61	55
PBDT-T:ITDI	1.53	0.94	0.59	63.2	8.0	56
HFQx-T:ITIC	1.57	0.92	0.65	72	9.4	57
PMOT16:IDIC	1.61	0.925	0.685	70	10.04	58
PTPDBDT:Cl-ITIC	1.56	0.94	0.62	67	9.5	59
PBDT(T)[2F]T:ITIC	1.60	0.94	0.66	78	9.8	60
J81:*m*-ITIC	1.57	0.96	0.61	78	11.05	61

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