Electronic Supplementary Information

High Efficiency All-Polymer Solar Cells: the Role of Donor Polymer Structure and Solvent Additive

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Hole and Electron Mobility Measurement (SCLC)

Hole-only and electron-only devices were fabricated to measure the hole mobility using the space charge limited current (SCLC) method with device structure of ITO/PEDOT/polymer:N2200/MoO_x/Al and ITO/Al/polymer:N2200/LiF/Al. The mobility was determined by fitting the dark current to the model of a single carrier SCLC, described by the equation:

$$J = \frac{9}{8}\varepsilon_0\varepsilon_r\mu_h\frac{V^2}{d^3}$$

where J is the current density, μ_h is the mobility under zero field, ε_0 is the permittivity of free space, ε_r is the material relative permittivity, d is the active layer thickness, and V is the effective voltage. The effective voltage can be obtained by subtracting the built-in voltage (Vbi) and the voltage drop (Vs) from the substrate's series resistance from the applied voltage (Vappl), $V=V_{appl} - V_{bi} - V_s$. The hole-mobility can be calculated from the slope of the $J_{1/2} \sim V$ curves.

Figure S1 $J^{1/2}$~V characteristics of the hole-only devices based on polymer:N2200 blend films cast from chloroform with 0.5% DIO

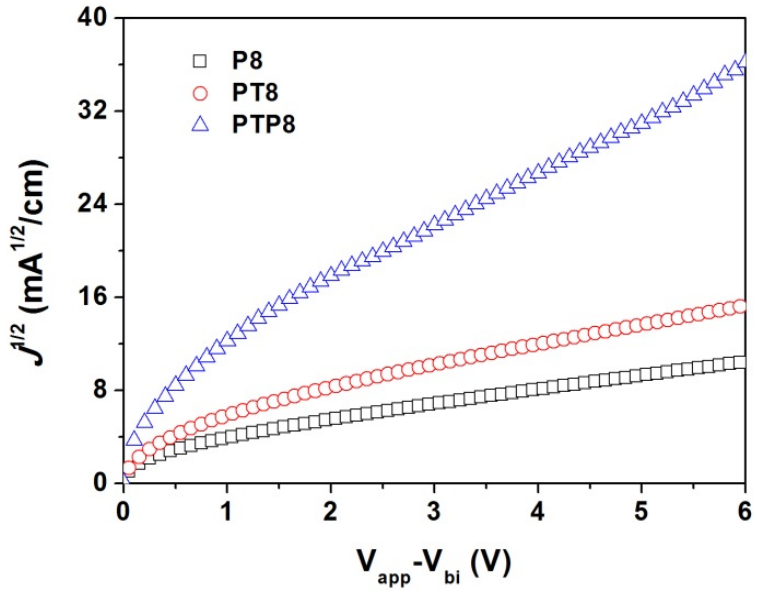

Figure S2. AFM height images (5.0 μ m × 5.0 μ m) of PTP8:N2200 blend films cast from chloroform with varying DIO content.

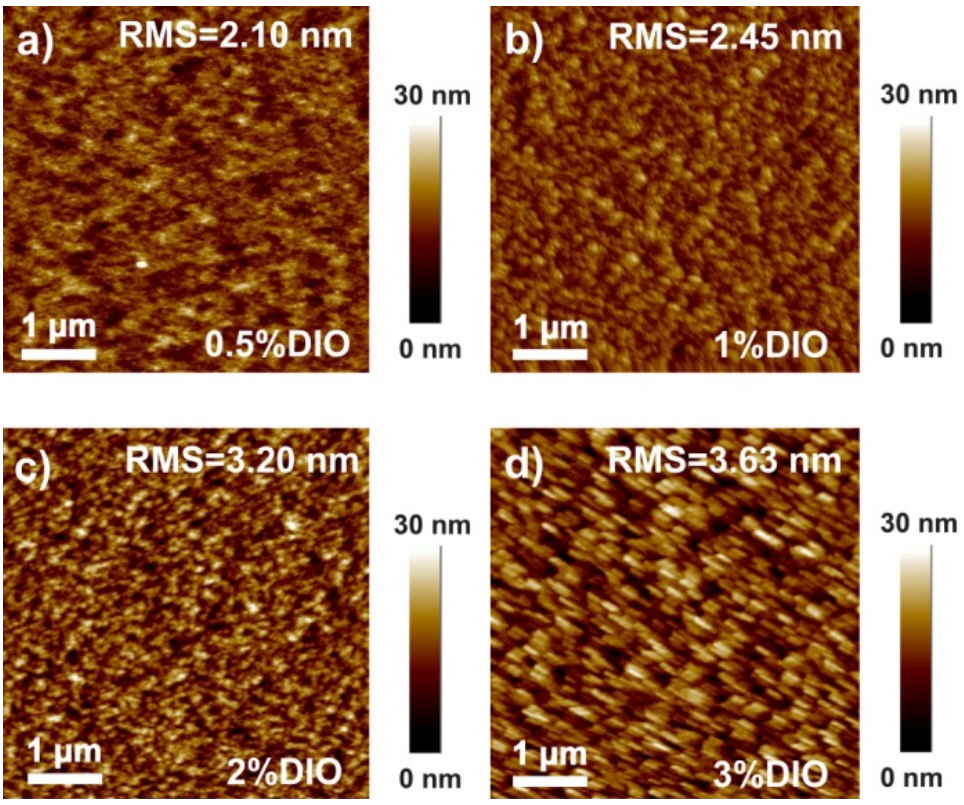

Figure S3 $J^{1/2}$~V characteristics of the hole-only (a), and electron-only (b) devices based on PTP8:N2200 blend films cast from chloroform with varying DIO concentration.

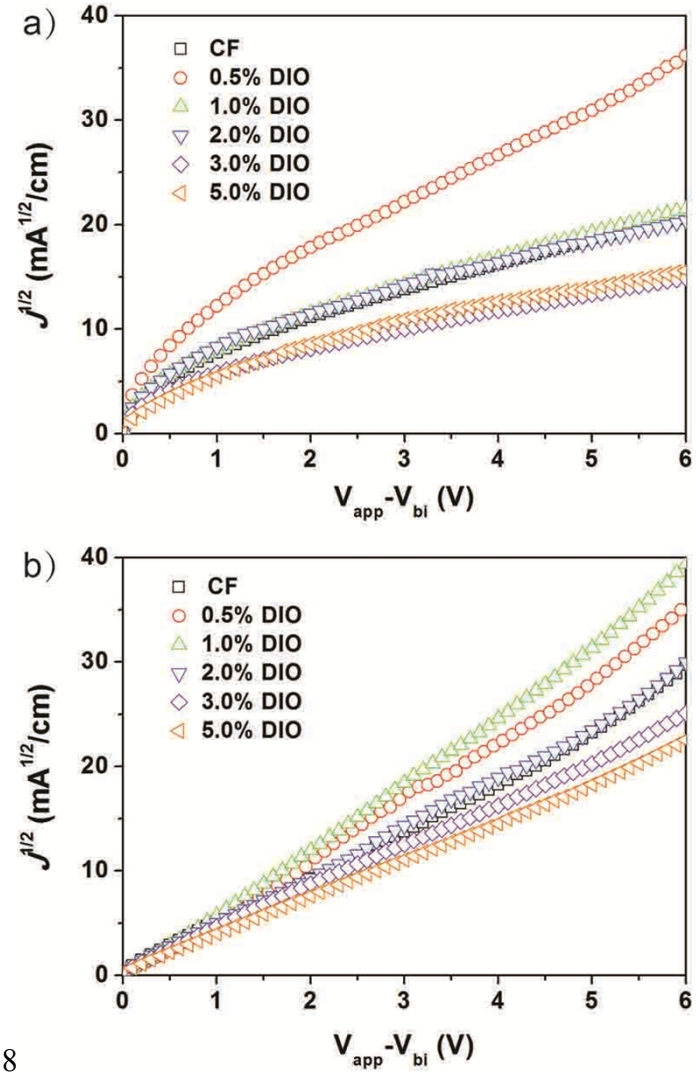

Figure S4. GIXD patterns of pristine polymer cast from chloroform as well as polymer:N2200 blend films cast from chloroform w/wo 0.5% DIO concentration. Note that the 2D spectra have not been corrected for the "missing wedge" of data along the out-of-plane direction.

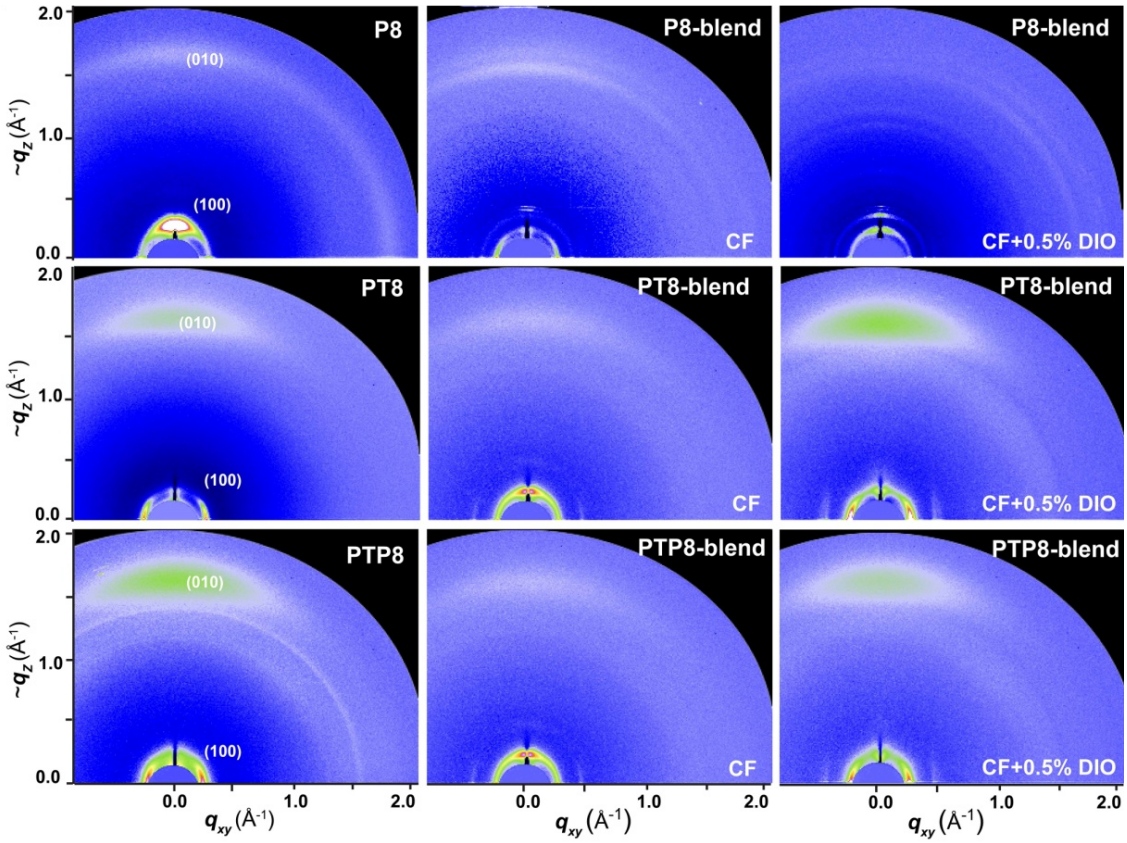

Table S1 Effect of spin speed and blend ratio on the performance of devices based on PTP8:N2200, with CF and 0.5% DIO as the processing solvent.

Spin-coating Speed	Ratio *w/w*	V_{oc} *(V)*	J_{sc} [mA/cm^2]	*FF (%)*	*PCE (%)*[a]	*Thickness (nm)*
1000 rpm	2:1	0.950	5.57	36.9	1.96 (1.80 ± 0.16)	190 ± 5
1500 rpm	2:1	0.938	5.88	44.3	2.44 (2.20 ± 0.24)	156 ± 5
2000 rpm	2:1	0.950	6.92	53.5	3.52 (3.42 ± 0.10)	124 ± 5
2500 rpm	2:1	0.975	7.38	54.3	3.91 (3.70 ± 0.21)	105 ± 5
3000 rpm	2:1	0.978	8.43	52.8	4.35 (4.20 ± 0.15)	92 ± 5
3500 rpm	2:1	0.977	8.18	51.0	4.07 (4.00 ± 0.07)	87 ± 5
3000 rpm	1:2	0.913	6.72	50.9	3.12 (3.00 ± 0.12)	-
3000 rpm	1:1	0.950	6.82	54.7	3.55 (3.50 ± 0.05)	-
3000 rpm	3:2	0.963	7.25	54.8	3.83 (3.60 ± 0.23)	-
3000 rpm	2:1	0.977	8.44	50.3	4.15 (4.05 ± 0.10)	-
3000 rpm	3:1	0.970	7.15	52.4	3.64 (3.55 ± 0.09)	-

[a] Averages values based on 6 parallel devices

Table S2 Optimized Devices performance of Polymer:N2200 blend films cast from chloroform w/wo thermal annealing and 0.5% DIO.

Donor	Treatment	V_{oc} *(V)*	J_{sc} [mA/cm^2]	*FF (%)*	*PCE (%)*[a]
P8	None	0.950	3.53	42.1	1.41 (1.35 ± 0.06)
P8	TA 120 ^{o}C[a]	0.945	2.65	40.0	1.00 (0.95 ± 0.05)
P8	0.5% DIO	0.945	3.74	43.0	1.52 (1.50 ± 0.02)
PT8	None	1.020	1.89	51.2	0.99 (0.90 ± 0.09)
PT8	TA 120 ^{o}C[a]	0.990	1.65	45.8	0.75 (0.70 ± 0.05)
PT8	0.5% DIO	1.000	3.45	50.8	1.75 (1.68 ± 0.07)
PTP8	None	0.975	6.53	44.4	2.83 (2.75 ± 0.08)
PTP8	TA 120 ^{o}C[a]	0.975	6.67	49.1	3.19 (3.09 ± 0.10)
PTP8	0.5% DIO[b]	0.978	8.43	52.8	4.35 (4.20 ± 0.15)

[a] Averages values based on 6 parallel devices

Table S3 The SCLC mobility of PTP8/N2200 blend with different DIO content.

DIO content	Electron mobility (cm ^{2}V^{-1}s^{-1})	Hole mobility (cm ^{2}V^{-1}s^{-1})
None	$6.12*10^{-5}$	$1.35*10^{-5}$
0.5 %	$8.48*10^{-5}$	$4.78*10^{-5}$
1.0 %	$1.08*10^{-4}$	$2.78*10^{-5}$
2.0 %	$6.01*10^{-5}$	$1.50*10^{-5}$
3.0 %	$3.78*10^{-5}$	$7.21*10^{-6}$
5.0 %	$2.56*10^{-5}$	$5.42*10^{-6}$