

SI:

Efficient All-Polymer Solar Cells Enabled by Interface Engineering

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Device characterization.

SCLC Measurements: The electron- and hole-mobilities were measured using space charge limited current (SCLC) method. The device architecture of the electron-only devices were ITO/PNDIT-F3N/active layer/PNDIT-F3N/Ag and ITO/PDINN/active layer/PDINN/Ag. The device architecture of the hole-only devices was ITO/PEDOT:PSS/active layer/Au. The charge carrier mobilities were determined by fitting the dark current into the model of a single carrier SCLC according to the equation: $J = 9\epsilon_0\epsilon_r\mu V^2/8d^3$, where J is the current density, d is the film thickness of the active layer, μ is the charge carrier mobility, ϵ_r is the relative dielectric constant of the transport medium, and ϵ_0 is the permittivity of free space. $V = V_{app} - V_{bi}$, where V_{app} is the applied voltage, V_{bi} is the offset voltage. The carrier mobilities were calculated from the slope of the $J \sim V^2$ curves.

Table S1 EQE, μ_{th} and μ_e of PM6:PYF-T-*o*/PNDIT-F3N, PM6:PYF-T-*o*/PDINN, PM6:PY-IT/PNDIT-F3N and PM6:PY-IT/PDINN.

Samples	J_{EQE} (mA/cm ²)	μ_{th} [cm ² V ⁻¹ s ⁻¹]	μ_e [cm ² V ⁻¹ s ⁻¹]
PM6:PYF-T- <i>o</i> /PNDIT-F3N (BHJ)	23.90	1.83E-04	1.34E-04
PM6:PYF-T- <i>o</i> /PDINN (BHJ)	24.30	1.83E-04	1.76E-04
PM6 :PY-IT/PNDIT-F3N (BHJ)	23.45	2.65E-04	1.17E-04
PM6 :PY-IT/PDINN (BHJ)	23.51	2.65E-04	2.91E-04

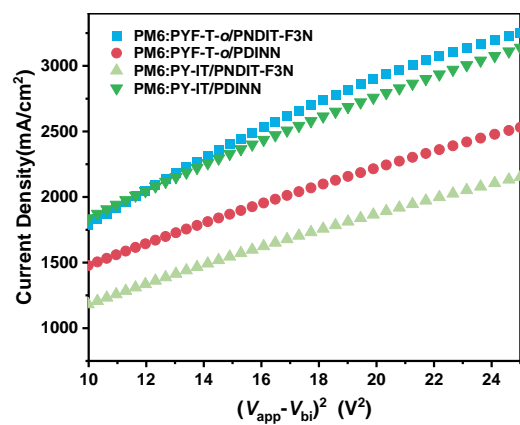


Figure S1 Electron mobility curves of PM6:PYF-T-*o*/PNDIT-F3N, PM6:PYF-T-*o*/PDINN, PM6:PY-IT/PNDIT-F3N and PM6:PY-IT/PDINN.

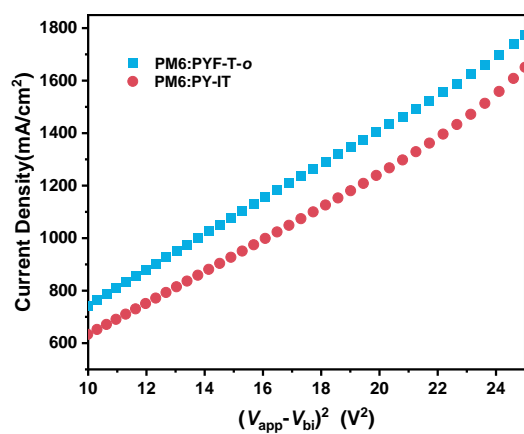


Figure S2 Hole mobility curves of PM6:PYF-T-*o* and PM6:PY-IT.

Table S2 Device optimization: The spin coating parameters of active layer are compared.

Devices	V_{oc} [V]	J_{sc} [mA/cm ²]	FF [%]	PCE [%]
PM6:PYF-T-o(2000rpm) / PNDIT-F3N	0.910	24.4	66.8	14.8
PM6:PYF-T-o (3000rpm) /PNDIT-F3N	0.912	23.8	66.8	14.4
PM6:PYF-T-o(2000rpm) /PDINN	0.911	24.6	65.6	14.8
PM6:PYF-T-o (3000rpm) /PDINN	0.912	23.8	66.4	14.5
PM6:PYIT(2500rpm) /PNDIT-F3N	0.911	24.1	66.4	14.6
PM6:PYIT(3000rpm) /PNDIT-F3N	0.911	24.1	66.4	14.6
PM6:PYIT(2500rpm) /PDINN	0.931	23.9	69.2	15.4
PM6:PYIT(3000rpm) /PDINN	0.931	23.9	70.1	15.6

Table S3 Device optimization results. Different ETL processing conditions are compared.

Devices	V _{oc} [V]	J _{sc} [mA/cm ²]	FF [%]	PCE [%]
PM6:PYF-T- <i>o</i> / PDINN (0.5mg/ml、1500rpm)	0.898	24.766	65.321	14.443
PM6:PYF-T- <i>o</i> / PDINN (0.5mg/ml、2000rpm)	0.902	25.250	65.057	14.725
PM6:PYF-T- <i>o</i> / PDINN (0.5mg/ml、2500rpm)	0.908	23.563	67.586	14.365
PM6:PYF-T- <i>o</i> / PDINN (1.0mg/ml、2500rpm)	0.912	25.640	65.712	15.266
PM6:PYF-T- <i>o</i> / PDINN (1.0mg/ml、3000rpm)	0.907	25.856	66.660	15.527
PM6:PYF-T- <i>o</i> / PDINN (1.0mg/ml、3500rpm)	0.907	25.091	66.863	15.119
PM6:PYF-T- <i>o</i> / PDINN (2mg/ml、3000rpm)	0.911	24.036	66.253	14.518
PM6:PYF-T- <i>o</i> / PDINN (2mg/ml、4000rpm)	0.912	24.735	66.625	15.027
PM6:PYF-T- <i>o</i> / PDINN (2mg/ml、5000rpm)	0.913	23.408	68.307	14.598
PM6:PYF-T- <i>o</i> / PDINN (3mg/ml、4000rpm)	0.910	24.026	67.046	14.669
PM6:PYF-T- <i>o</i> / PDINN (3mg/ml、5000rpm)	0.912	24.780	66.933	15.123
PM6:PYF-T- <i>o</i> / PDINN (3mg/ml、6000rpm)	0.911	24.103	66.170	14.529

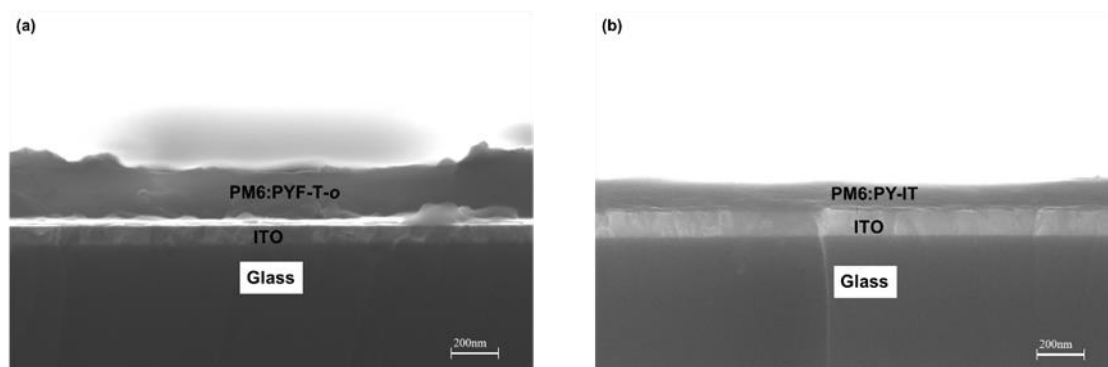


Figure S3. cross-sectional SEM image of each layer.