Supporting Information

Optimization of Local Orientation and Vertical Phase Separation by Adding a Volatilizable Solid Additives to J51:N2200 Blend to Improve its Photovoltaic Performance

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Fig. 1 The photovoltaic properties of J51:N2200 blend for the varying content of Bipy.

Table 1. The summary of parameters of photovoltaic properties for J51:N2200 devices varying content of Bipy.

	$V_{ m oc}$ (V)	J _{sc} (mA cm ⁻²)	FF (%)	PCE (%)
Blend+0.5 wt% Bipy	0.83	15.04	65.58	8.22
Blend+0.75 wt% Bipy	0.85	15.02	68.63	8.73
Blend+1 wt% Bipy	0.84	15.09	66.97	8.48
Blend+2 wt% Bipy	0.84	14.04	65.80	7.73



Fig. 2 The hole and electron mobility of films at different conditions: Blend, Blend/TA, Blend+Bipy, Blend+Bipy/TA.



Fig. 3 The photovoltaic properties of PBDB-T:N2200 blend on different conditions (a) The *J*-*V* curves. (b) The EQE data.

Table 2. The summary of corresponding parameters of the dependence of photovoltaic properties of PBDB-T:N2200 on different conditions.

PBDB-T:N2200	$V_{ m oc}$ (V)	J _{sc} (mA cm ⁻²)	FF (%)	PCE (%)
Blend	0.82	14.73	57.40	6.89
Blend/TA	0.81	15.37	60.70	7.61
Blend+Bipy	0.86	14.93	58.97	7.56
Blend+Bipy/TA	0.86	15.95	62.92	8.59



Fig. 4 (a) The GIXRD pattern of Bipy thin film. (b) Thermogravimetric Analysis (TGA) of Bipy, the scan rate is 10 °C min⁻¹, the black line corresponding heating from 25 °C to 300 °C, while the red line corresponding heating from 25 °C to 200 °C and the temperatures were held for 1h at 100, 115, 130 °C, respectively. (c) Photograph of the top of crucible after the heating and cooling of Bipy. (d) The UV-vis absorption spectra of Bipy film before and after annealing at 130 °C for 10 min. (e) Morphology of Bipy spin-coated on Si substrates, (f) The Bipy films were thermal annealing at 130 °C for 10 min characterized by optical profiler (Insert photos are taken by the camera.)



Fig. 5 (a-c) AFM height images of films processed under different conditions. (d-f) The roughness for corresponding films.



Fig.6 The GIXRD spectra of different films. (a) J51 film, (b) N2200 film, (c) The 1D linecuts for 2D GIXRD.



Fig. 7 IR spectra of film. All tested films were spin-coated on KBr pellets. (a) The IR spectra of pure component films for J51, N2200, and Bipy. (b) The enlarged of Figure (a) from 1800-550 cm⁻¹. (c) The IR spectra of blend films at different treatments, Blend, Blend/TA, Blend+Bipy, Blend +Bipy/TA. (d) The enlarged of Figure (c) from 1800-550 cm⁻¹.



Fig. 8 Absorption spectrum of films (100 nm thick) during incremental etching. (a) Blend, (b) Blend/TA, (c) Blend+Bipy, (d) Blend+Bipy/TA.



Fig. 9 Absorption spectra of pure component film. (a) J51, J51/TA, J51+Bipy, J51+Bipy/TA. (b) N2200, N2200/TA, N2200+Bipy, N2200+Bipy/TA.