Supporting Information

Manipulating the crystal structure of a conjugated polymer for efficient sequentially processed organic solar cells

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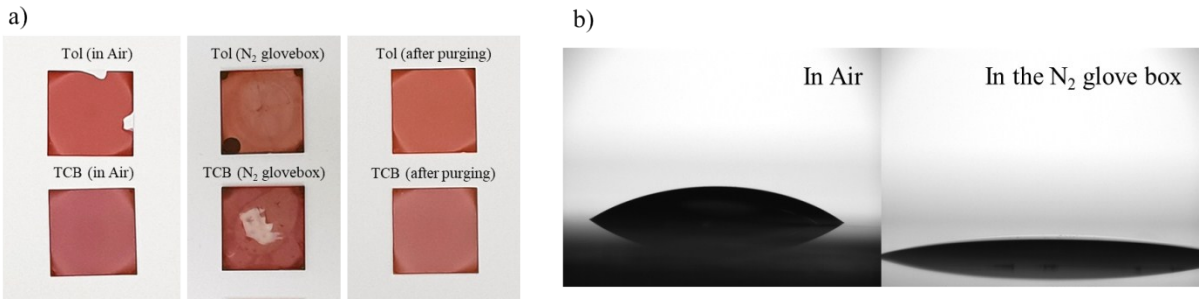


Figure S1. a) Details of sequential processed films in three different conditions for the experiment. b) Water contact angle images of the films (TCB).

We prepared the films and performed the Sq-process in three different conditions. As mentioned in the manuscript, we could obtain the fine quality with the process in air while the films were peeled off with the process in the N_2 glove box since the residual solvent resulted from the process in the sealed glove box dissolved sequence DCM solvent. (All P3HT samples were dried about one hour.) To prove this, we additionally fabricated the Sq-processed films in the glove box after purging with N_2 gas about one hour to flow out the residual solvent in the glove box, and then the films showed good quality like in air process due to the absence of the residual solvent. The contact angle images also clearly show the residual solvent effect dissolving the bottom layer.

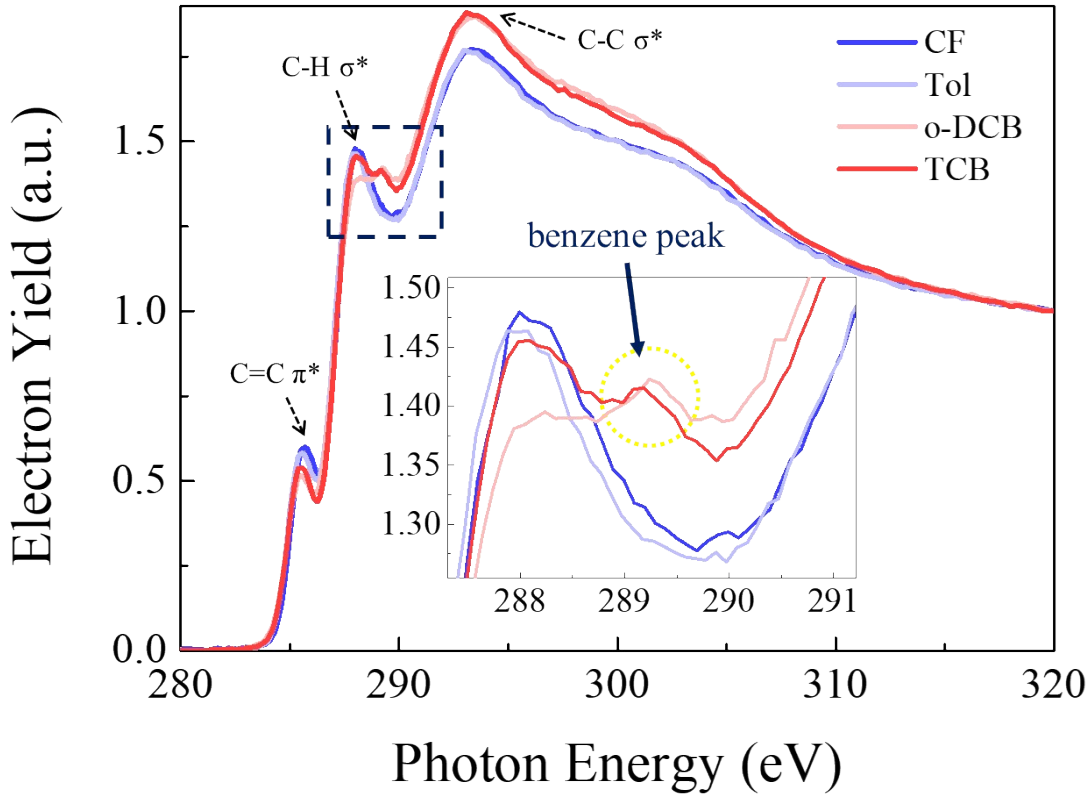


Figure S2. NEXAFS spectra of P3HT thin films using various solvents in incident angle (55º).

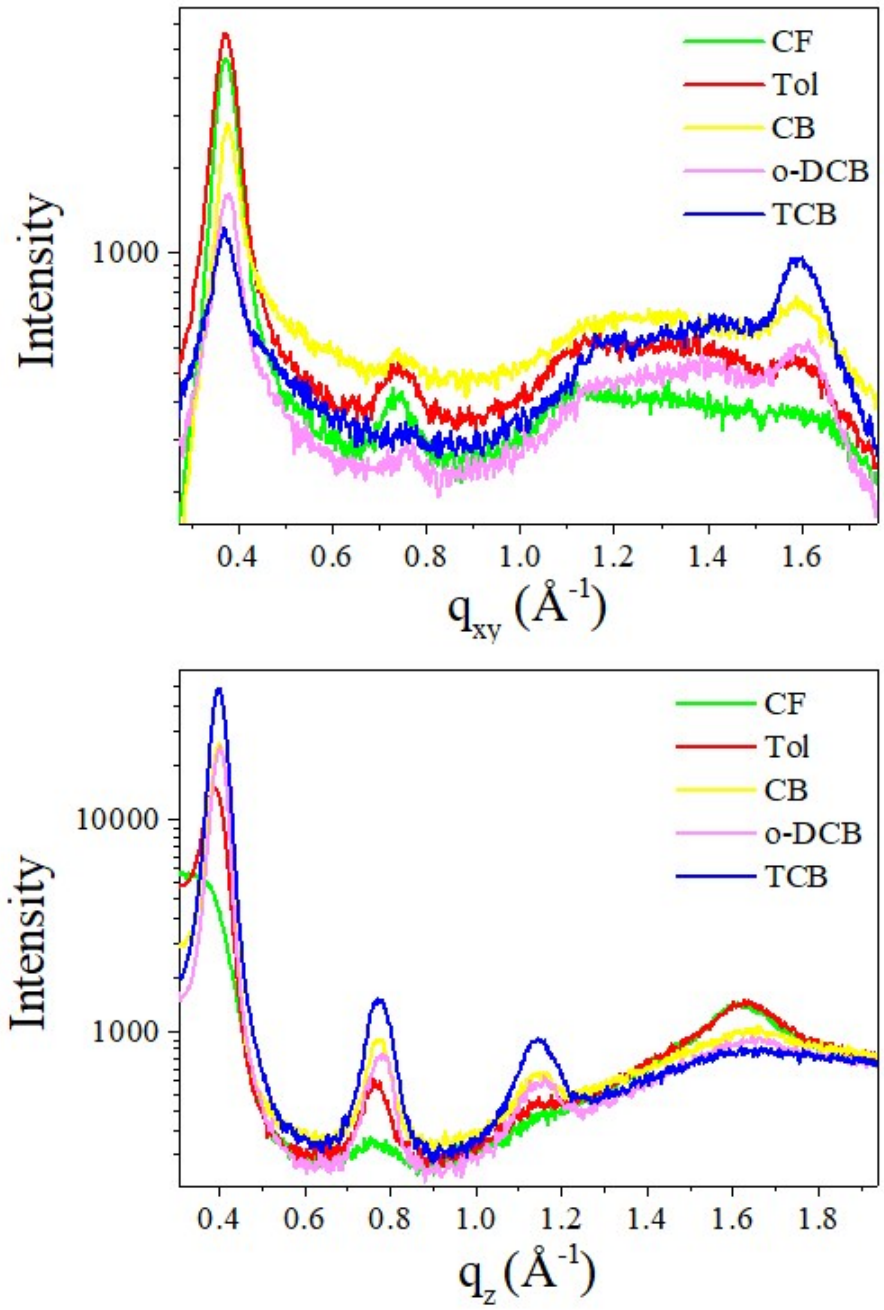


Figure S3. High resolution grazing incidence diffraction data of P3HT films using CF, Tol, CB, o-DCB and TCB. (Incident angle = 0.13 º)

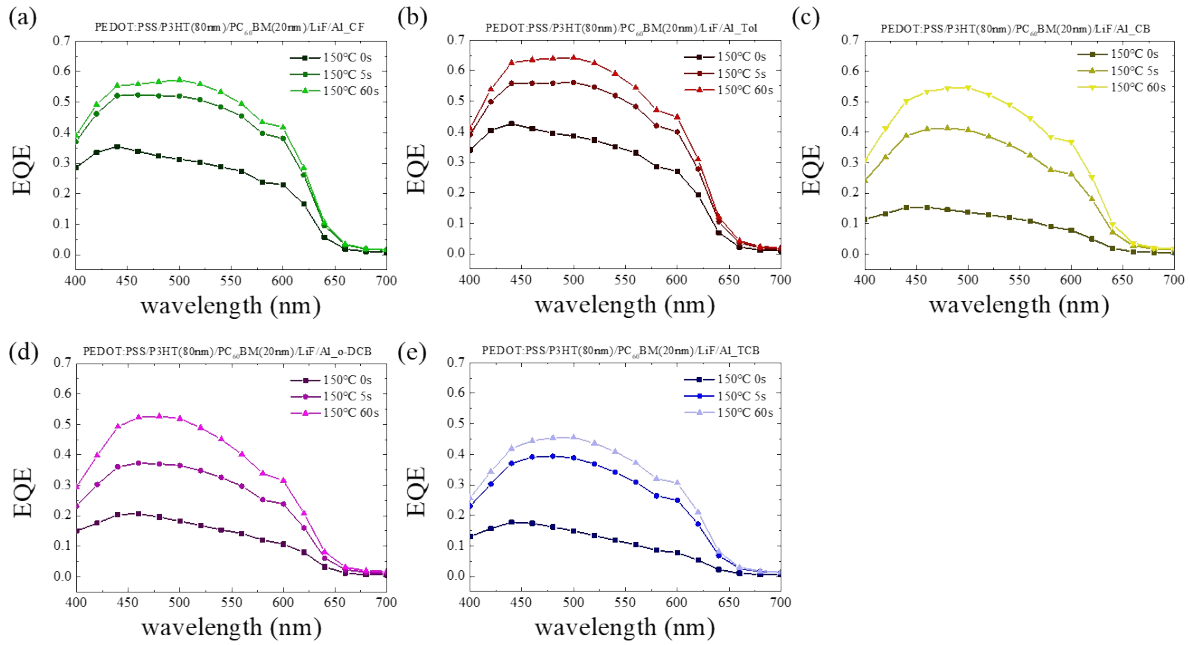
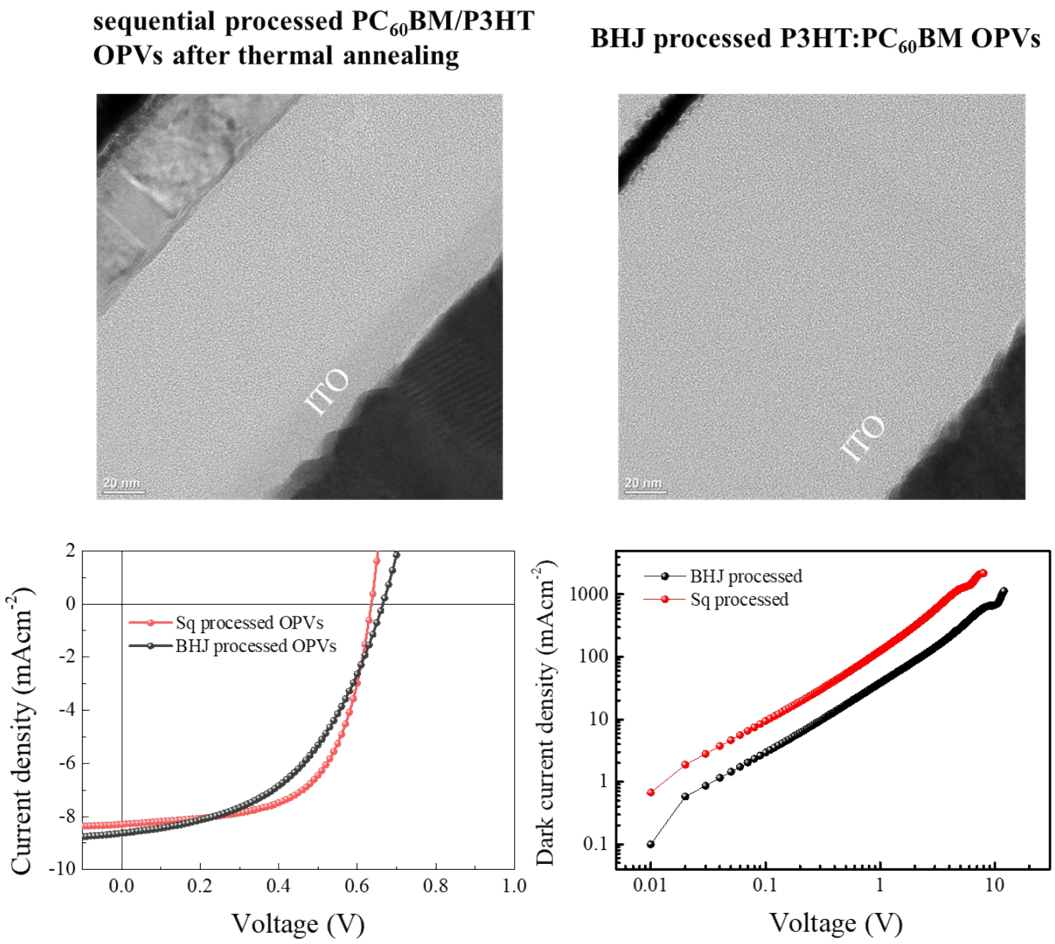


Figure S4. (a-e) The external quantum efficiency (EQE) spectra of the sequential processed OPVs using different solvents and the annealing process.

solvent	TA	*EQE-*J_{SC} [mA/cm²]	J_{SC} [mA/cm²]
CF	150°C 0s	4.43	4.98
	150°C 5s	6.93	6.91
	150°C 60s	7.49	7.61
Tol	150°C 0s	5.30	5.29
	150°C 5s	7.34	7.29
	150°C 60s	8.30	8.30
CB	150°C 0s	1.89	1.84
	150°C 5s	5.16	5.16
	150°C 60s	6.86	6.88
o-DCB	150°C 0s	2.52	2.50
	150°C 5s	4.63	4.55
	150°C 60s	6.37	6.39
TCB	150°C 0s	1.96	1.81
	150°C 5s	4.91	4.62
	150°C 60s	5.74	5.75

Table S1. Comparison with EQE and measured Jsc from the I-V curves of the devices.



solvent	Thermal Annealing	μ_h [cm²/V s]	J_{SC} [mA/cm²]	V_{OC} [V]	*FF* [%]	PCE [%]
Tol	150°C 60s (Sq processed)	3.73×10^{-3}	8.30 (8.12)	0.64 (0.63)	61.11 (61.31)	3.25 (3.13)
	150°C 10min (BHJ processed)	1.34×10^{-4}	8.63 (8.21)	0.67 (0.67)	48.24 (48.56)	2.79 (2.66)

Figure S5. TEM images and I-V curves of sequential processed and BHJ processed devices.

we measured the hole mobility of the active layers by using space charge limited current (SCLC) method with the hole only device configuration of ITO (145 nm) / PEDOT:PSS (42 nm) / active layer (100 nm) / MoO3 (8 nm) / Ag (100 nm).

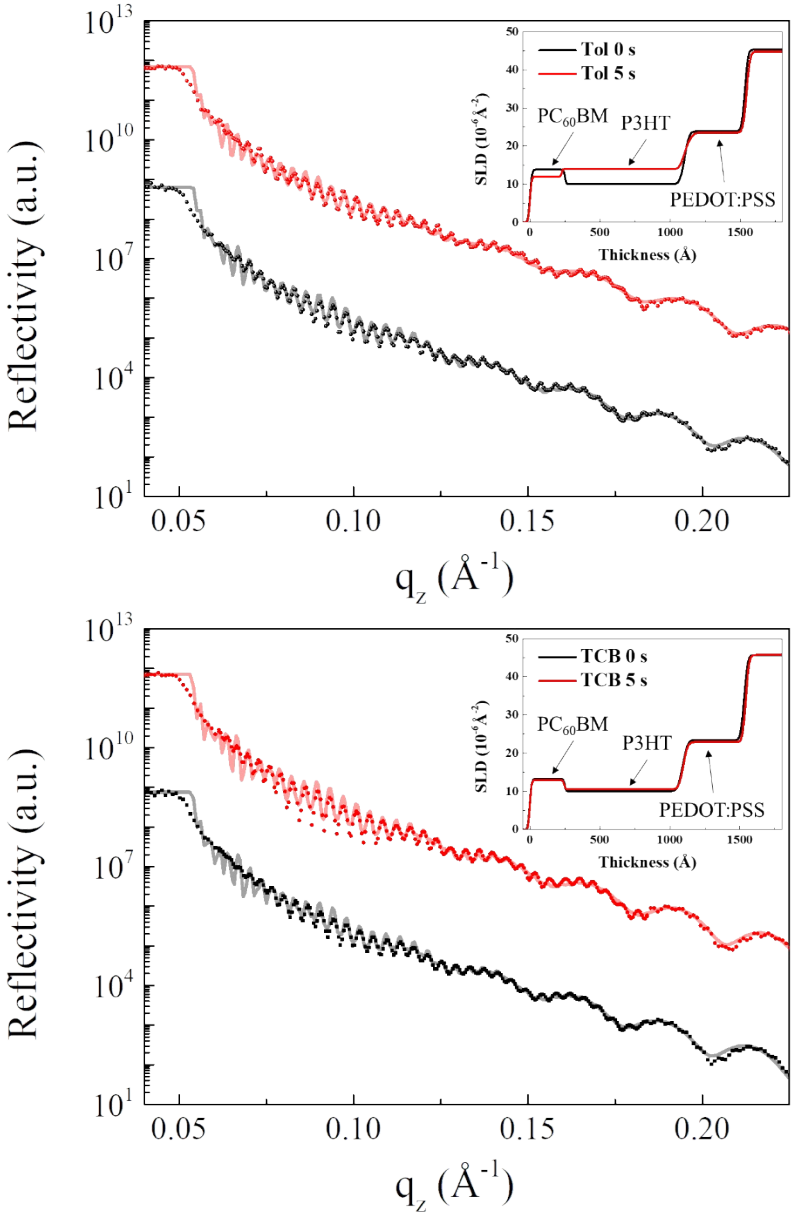


Figure S6. X-ray reflectivity (XRR) for sequentially processed $PC_{60}BM$/P3HT samples using Tol and TCB. The solid lines are fits that yields the scattering length density (SLD) profiles shown in the inset.

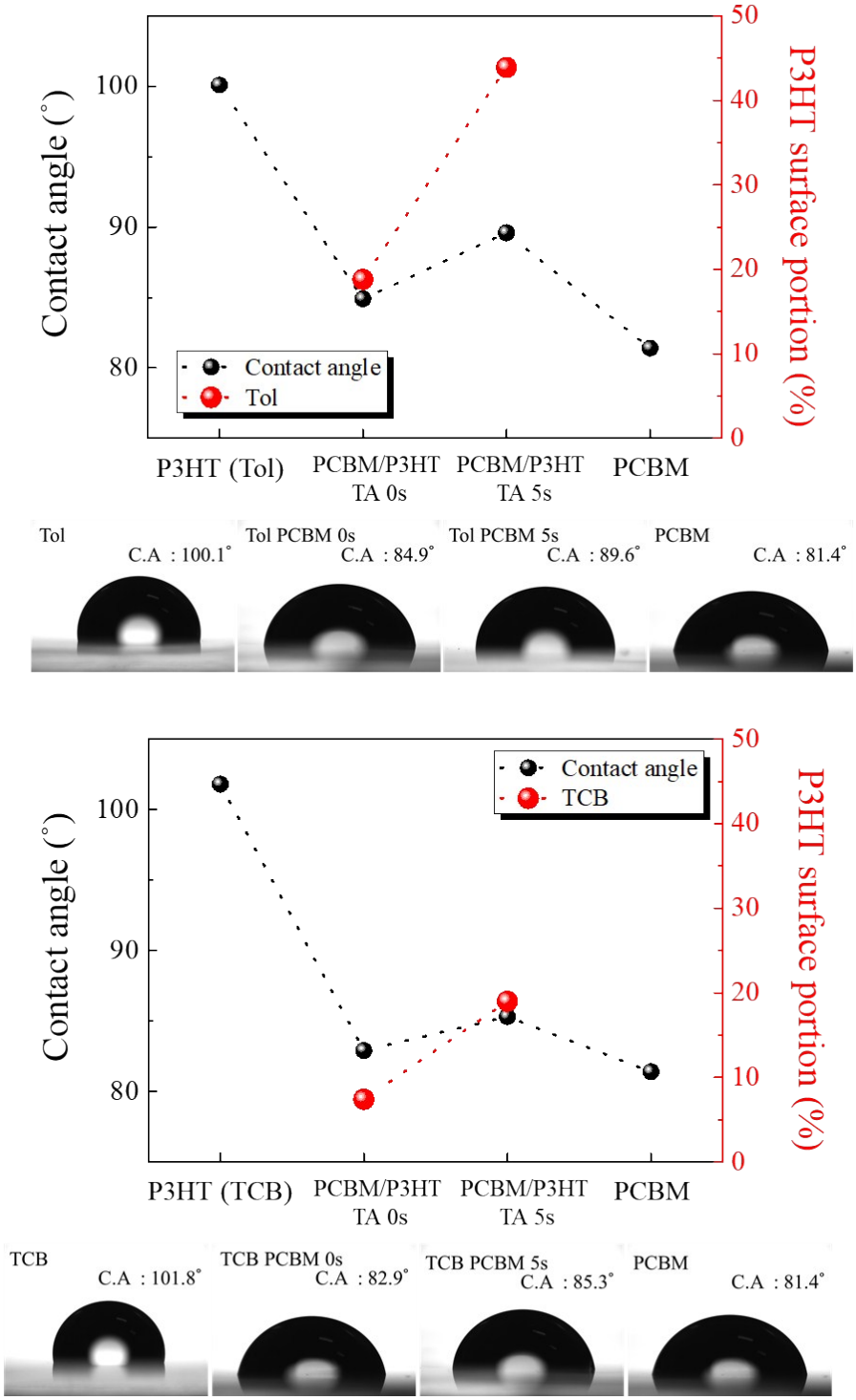
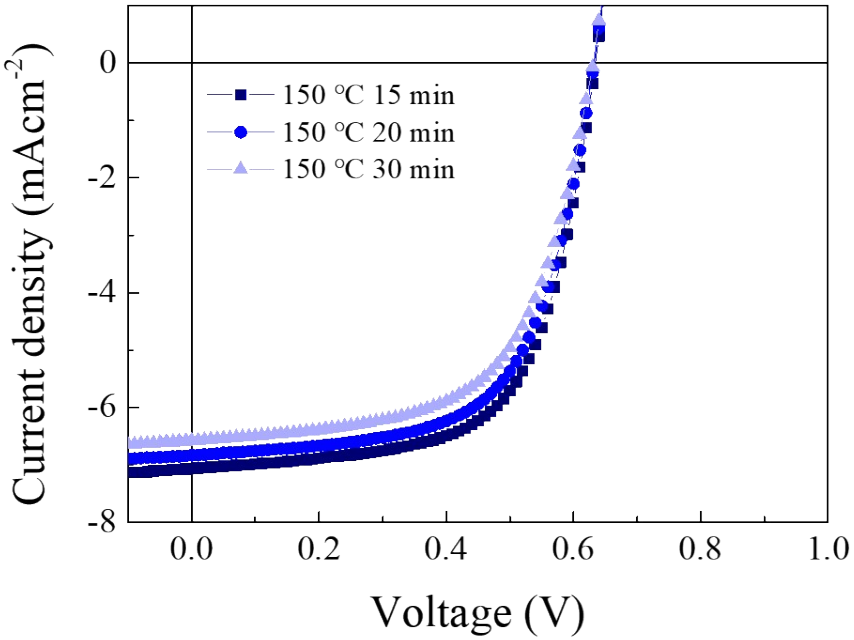


Figure S7. Contact angle measurement data for sequentially processed $PC_{60}BM$/P3HT samples using Tol and TCB.



ITO / PEDOT:PSS / P3HT-TCB / PCBM / LiF / Al	J_{sc} [mA/cm²]	V_{oc} [V]	FF [%]	PCE [%]
TA 15 min	7.067(6.859)	0.630(0.626)	64.34(63.51)	2.86(2.73)
TA 20 min	6.833(6.767)	0.630(0.626)	62.91(62.70)	2.71(2.66)
TA 30 min	6.579(6.495)	0.630(0.623)	60.88(60.93)	2.50(2.40)

Figure S8. Photovoltaic properties of the $PC_{60}BM$/P3HT (TCB)-based bilayer OPVs. Current density-voltage (*J-V*) curves under an AM 1.5G condition (100 mW/cm²). Summary of the PCEs of the devices made using the different annealing times in the underlying table.