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Supplementary Information to:

Solution Structure: Defining Polymer Film Morphology and Optoelectronic Device Performance

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Table S1 Relative viscosities of the three PCDTBTs measured at different solution concentrations. Relative viscosities increase with increasing concentration and molecular weight.

	$c_{Polymer}$ mg/ml	relative viscosity -- slow cooling	relative viscosity -- fast cooling	change %
PCDTBT-1	6	1.15 ±0.03	1.14 ±0.03	-1
PCDTBT-2	7	1.91 ±0.05	2.07 ±0.05	+8
	15	4.04 ±0.08	5.21 ±0.09	+29
PCDTBT-3	4	4.65 ±0.11	5.46 ±0.13	+17
	6	17.86 ±0.23	48.84 ±2.67	+173

Table S2 Relative viscosities of the three PCDTBTs and of corresponding PCDTBT:$PC_{71}BM$ 1:4 w/w blends. Very similar behavior for pristine polymers and for blends was found when comparing the effect of different solution treatments, *i.e.* slow and fast cooling rates, on the resulting solution viscosities.

	$c_{Polymer}$ mg/ml	relative viscosity -- slow cooling	relative viscosity -- fast cooling	change %
PCDTBT-1	6	1.15 ±0.03	1.14 ±0.03	-1
Blend-1	6	1.24 ±0.04	1.23 ±0.04	-1
PCDTBT-2	7	1.91 ±0.05	2.07 ±0.05	+8
Blend-2	7	2.11 ±0.04	2.29 ±0.03	+9
PCDTBT-3	4	4.65 ±0.11	5.46 ±0.13	+17
Blend-3	4	4.90 ±0.07	5.70 ±0.13	+16

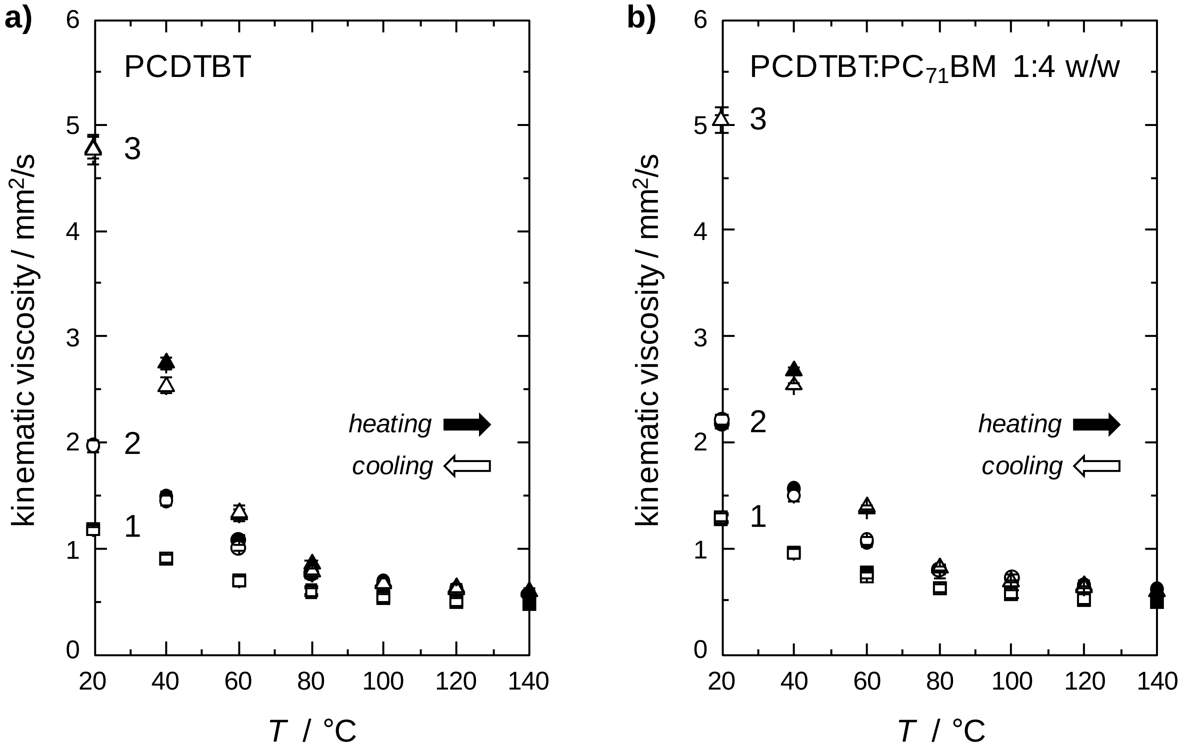

Fig. S1 Thermal evolution of kinematic viscosities for PCDTBTs 1-3. Full and open symbols were recorded during a heating and cooling cycle, respectively. No hysteresis in viscosity was found when applying a slow cooling rate of about 1.5 °C/min.

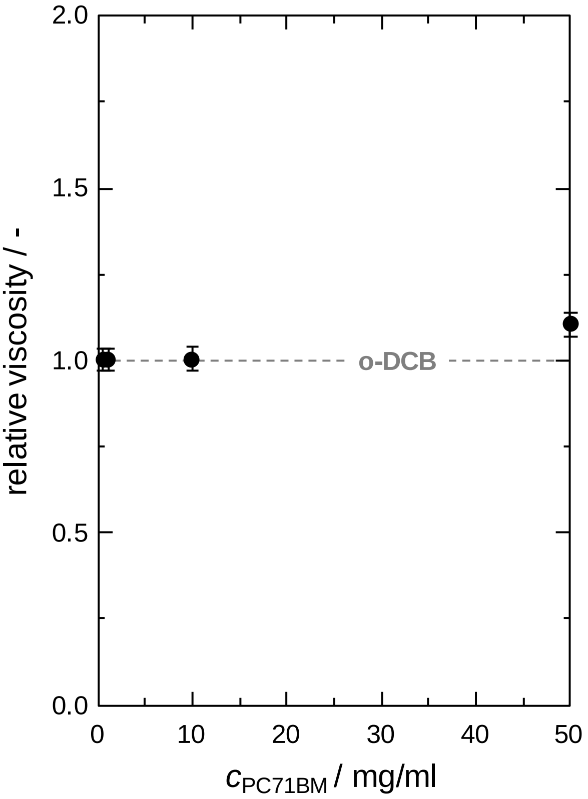

Fig. S2 Solution concentration dependent relative kinematic viscosities of $PC_{71}BM$. Over a wide range of concentrations, addition of $PC_{71}BM$ to the solvent (o-DCB) only has a very minor effect on resulting solution viscosities.

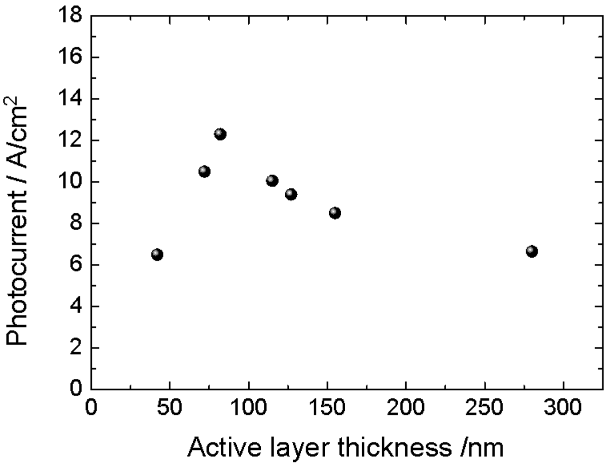

Fig. S3 Short circuit photocurrent of PCDTBT:PC$_{71}$BM solar cells as a function of active layer thickness. A maximum in the photocurrent is observed for an active layer thickness between 80 and 90 nm.

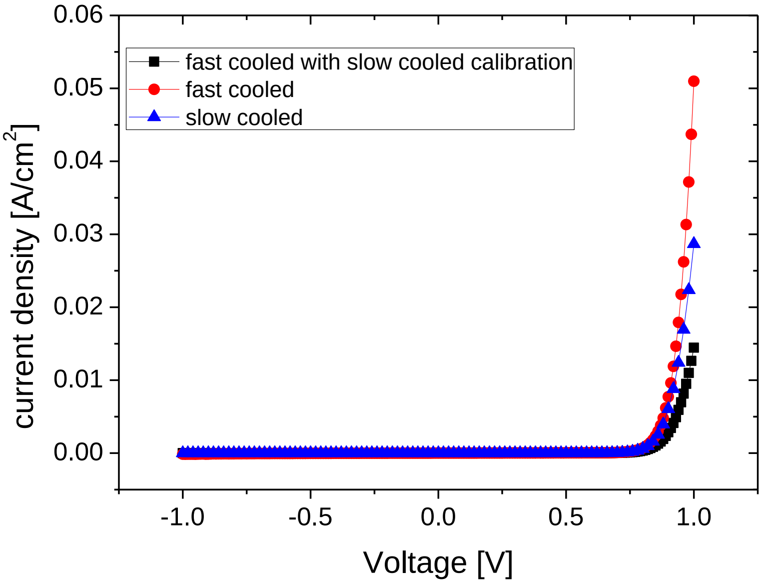

Fig. S4 Dark IV characteristics of PCDTBT:$PC_{71}BM$ devices that were fabricated from fast- and slow-cooled solutions and calibrated to a target thickness of 85±5 nm. For comparison, the IV of a fast cooled solution is presented that was fabricated according to the processing protocol elaborated for slow-cooled solutions.

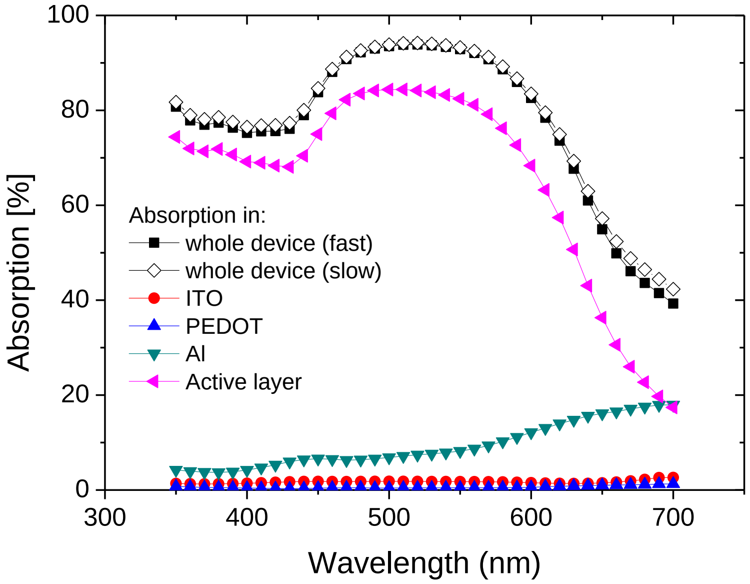

Fig. S5 Light absorption within the different layers of a PCDTBT:$PC_{71}BM$ solar cell. The absorption in the whole device is measured by performing spectroscopic reflectometry for devices fabricated with slow and fast cooled solutions. The absorption of the whole device is found to be identical for both processing pathways. The parasitic absorptions within Al, PEDOT and ITO layers are simulated by the transfer matrix method and the absorption within the active layer is evaluated by subtracting the simulated parasitic absorption from the total absorption within the device as determined with reflectometry.