

Entanglements in Marginal Solutions: A Means of Tuning Pre-Aggregation of Conjugated Polymers with Positive Implications for Charge Transport

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Table S1: The viscosity of four different solvents measured at room temperature.

Solvent	CHCl ₃	Tol	CB	oDCB
Viscosity (mPa·s)	1.335±0.006	0.546±0.005	0.773±0.006	0.524±0.003

Table S2: The viscosity of P3HT (68 kDa) solution in four different solvents with constant concentration (5 mg/ml) before and after disentanglement process at room temperature.

Solvent		CHCl ₃	Tol	CB	oDCB
Viscosity (mPa·s)	pristine	0.746±0.007	0.826±0.013	1.075±0.001	1.835±0.007
	Sonicated	0.727±0.004	0.775±0.004	1.074±0.024	1.835±0.006

Table S3. The viscosity of P3HT solution in Tol under constant concentration (5 mg/ml) with different Mw before and after disentanglement process at room temperature

M _w of P3HT (kDa)		22	32	68	100	120	180
Viscosity (mPa·s)	pristine	0.63±0.01	0.73±0.01	0.83±0.02	6.05±0.04	7.29±0.21	13.42±0.29
	sonicated	0.62±0.01	0.68±0.01	0.78±0.04	4.27±0.04	4.73±0.18	9.51±0.52

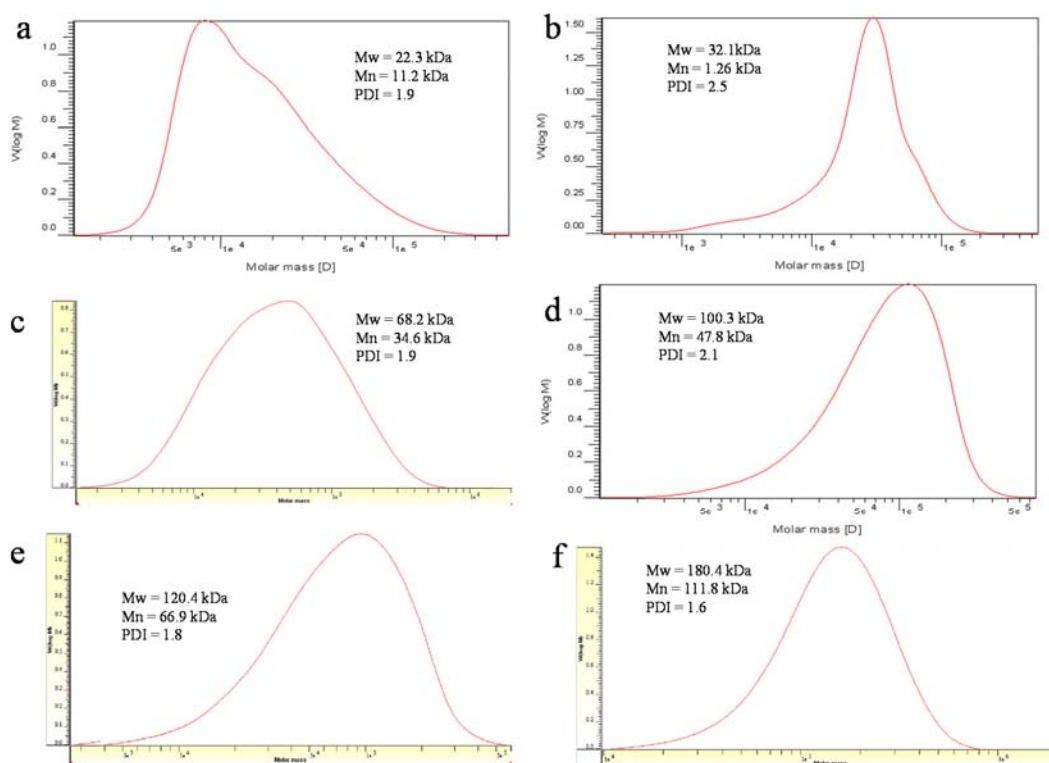


Figure S1: Gel permeation chromatography (GPC) data obtained on different Mw dissolved in THF.

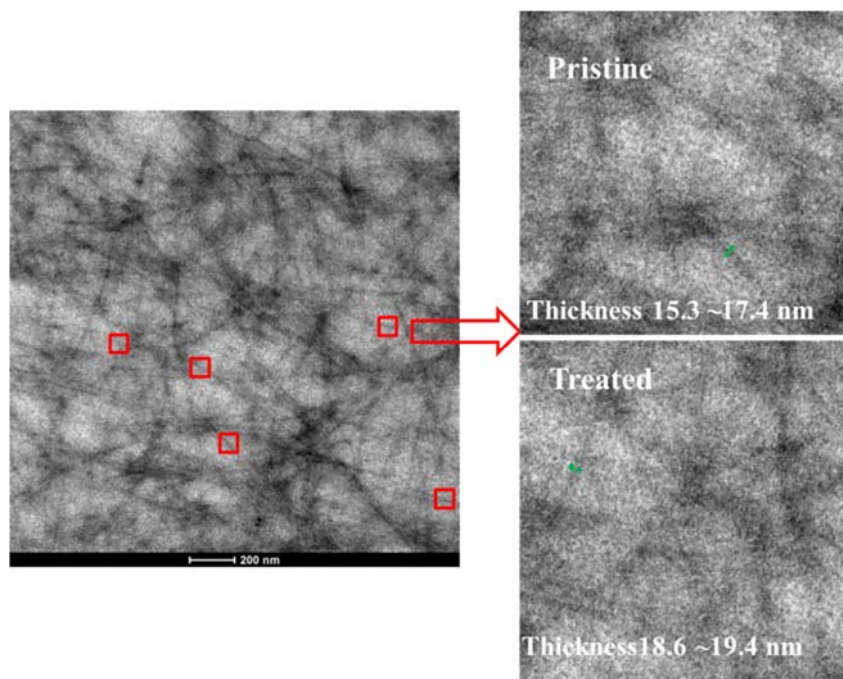


Figure S2: The average lamellar thickness measured at least for 5 clearly isolated fibrils (marked with red squares) for P3HT (68 kDa) thin-film spin-cast from pristine and ultrasonication solutions in Tol.

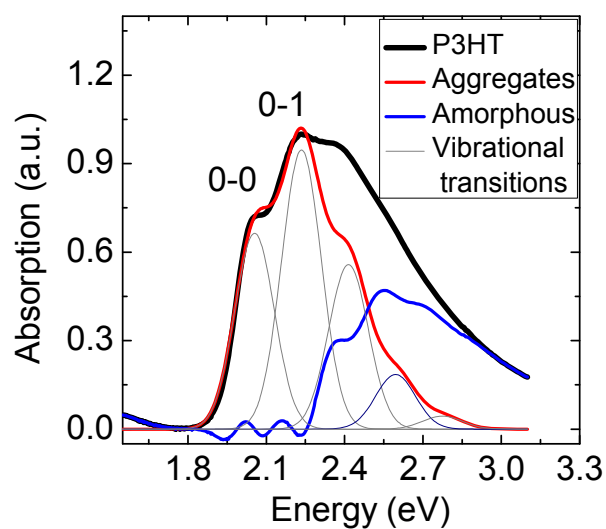


Figure S3: Absorption spectrum of P3HT thin-films with Spano fit using Equation 1

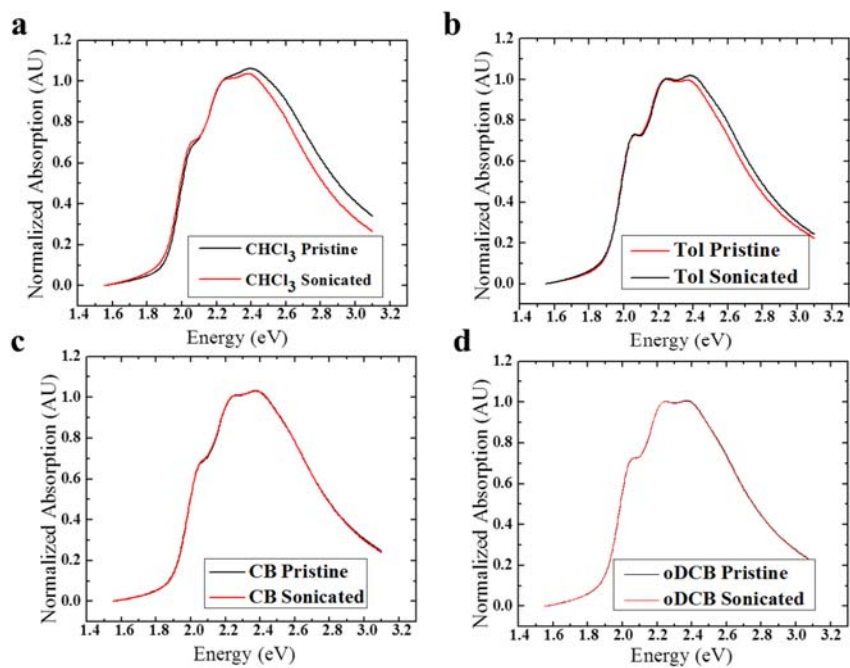


Figure S4: Normalized UV-Vis absorption spectra of P3HT thin film from different solution with/without disentanglement process with constant concentration (5 mg/ml): a) CHCl₃, b) Tol, c) CB, d) oDCB.

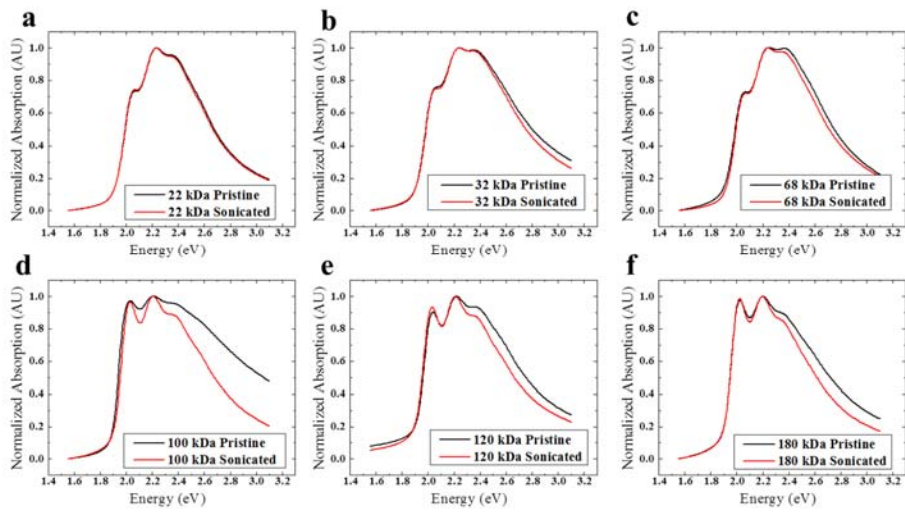


Figure S5: Normalized UV-Vis absorption spectra of P3HT (different Mws) thin film from Tol with/without disentanglement process with constant concentration (5 mg/ml). a) 22 kDa, b) 32 kDa, c) 68 kDa, d) 100 kDa, e) 120 kDa, f) 180 kDa.

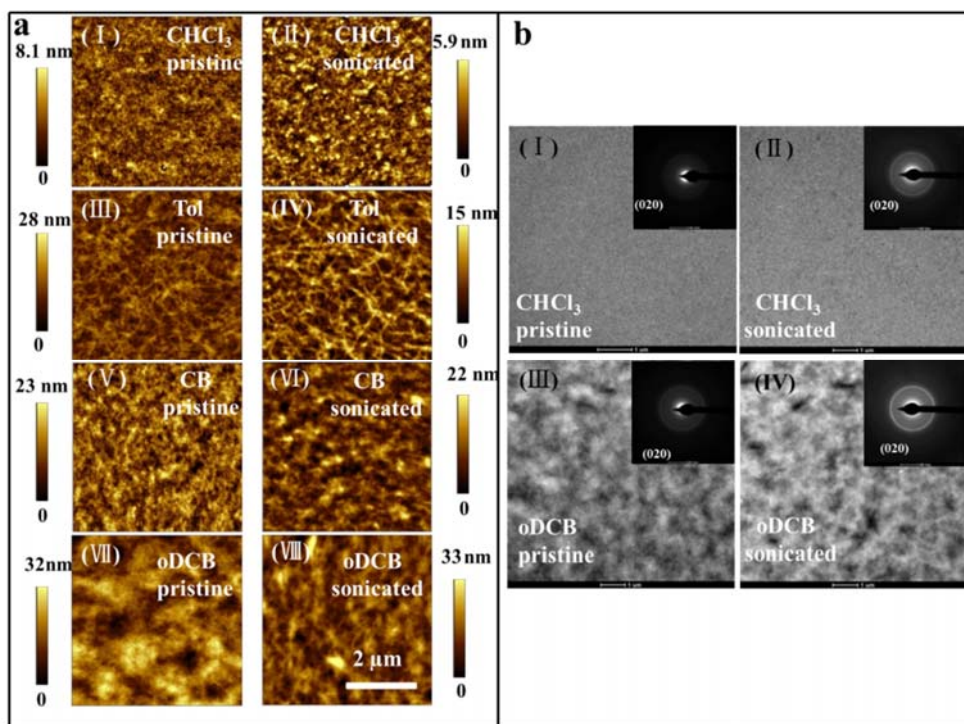


Figure S6: (a) AFM morphology images of P3HT (68 kDa) thin-films by spin coating with same concentration (5 mg/ml) in different solvents before and after disentanglement process: I) in CHCl_3 , II) in CHCl_3 sonicated, III) in Tol, IV) in Tol sonicated, V) in CB, VI) in CB sonicated, VII) in oDCB, VIII) in oDCB sonicated. (b) Bright-field TEM images of P3HT (68 kDa) thin-films by spin coating with concentration of 5 mg/ml in CHCl_3 and oDCB before and after disentanglement process: I) in CHCl_3 , II) in CHCl_3 sonicated, III) in oDCB, IV) in oDCB sonicated. The insets are ED images.

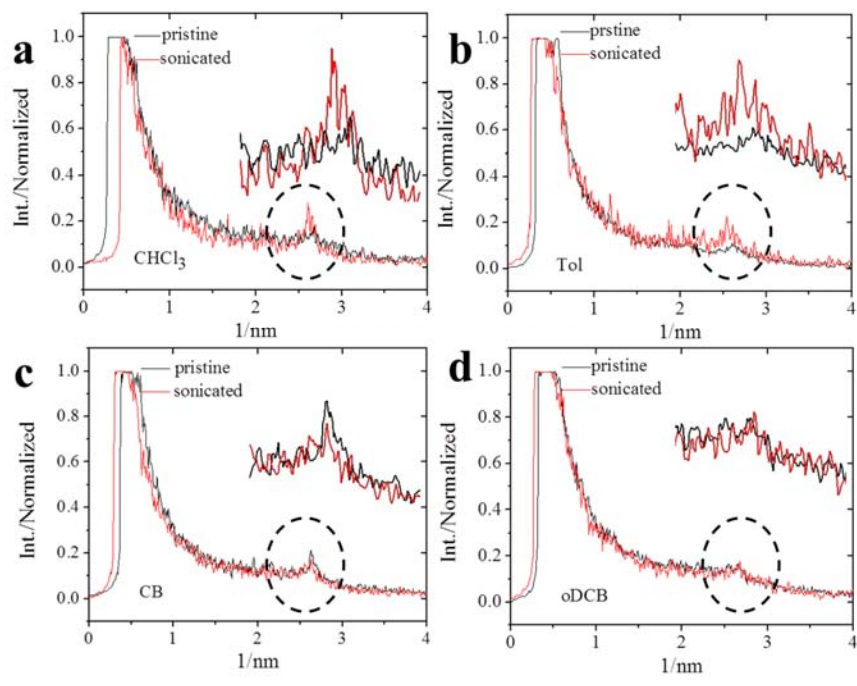


Figure S7: Normalized electron diffraction (ED) profiles obtained from plan-view TEM of P3HT (68 kDa) with same concentration (5 mg/ml) from CHCl₃ (a), Tol (b), CB (c) and oDCB (d) before and after disentanglement process.

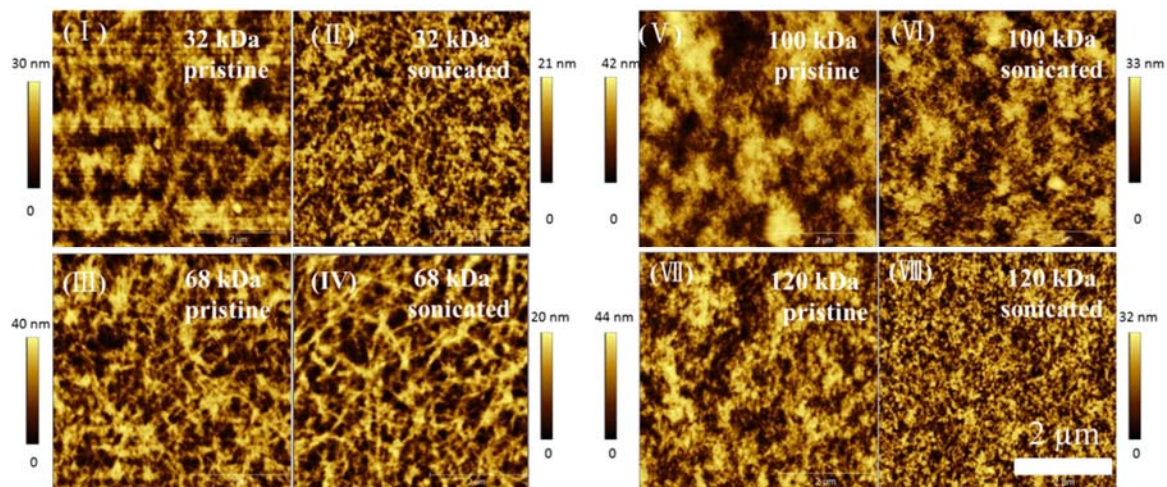


Figure S8: AFM morphology images of P3HT thin-films by spin coating with same concentration (5 mg/ml) in Tol with different Mw before and after disentanglement process: I) 32 kDa, II) 32 kDa sonicated, III) 68 kDa, IV) in 68 kDa sonicated, V) 100 kDa, VI) 100 kDa sonicated, VII) 120 kDa, VIII) 120 kDa sonicated.

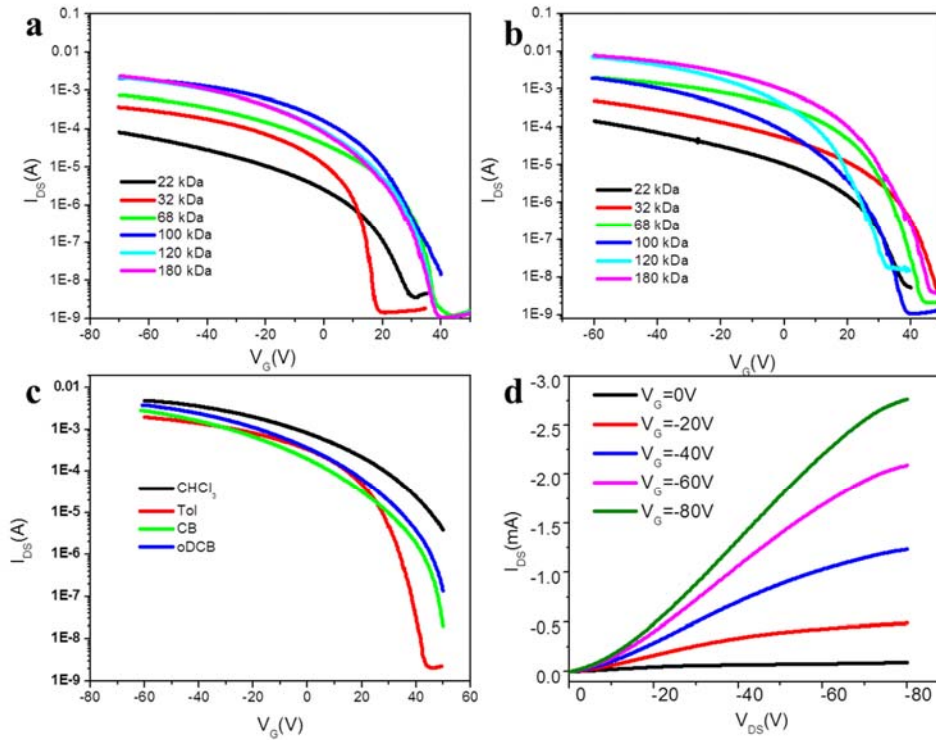


Figure S9: Electrical characteristics in ambient conditions of a representative OTFT with 5mg/ml cast P3HT organic semiconductor thin-films on 240 nm SiO₂ dielectric and W/L (10000 μ m/2.5 μ m) of 4000. All devices were fabricated in ambient conditions and measured in glove box filled with Nitrogen after one night. (a) Transfer characteristics (I_{DS} vs. V_G) with a constant V_{DS} fabricated in ambient from P3HT solution in Tol with different Mw, (b) from P3HT solution in Tol with different Mw after disentanglement process. (c) From P3HT (68 kDa) solution from different solvents after disentanglement process. (d) Output characteristics (I_{DS} vs. V_{DS}) with various V_G measured from film spin-cast from a P3HT (100 kDa) solution in Tol ultrasonicated for 4 minutes.

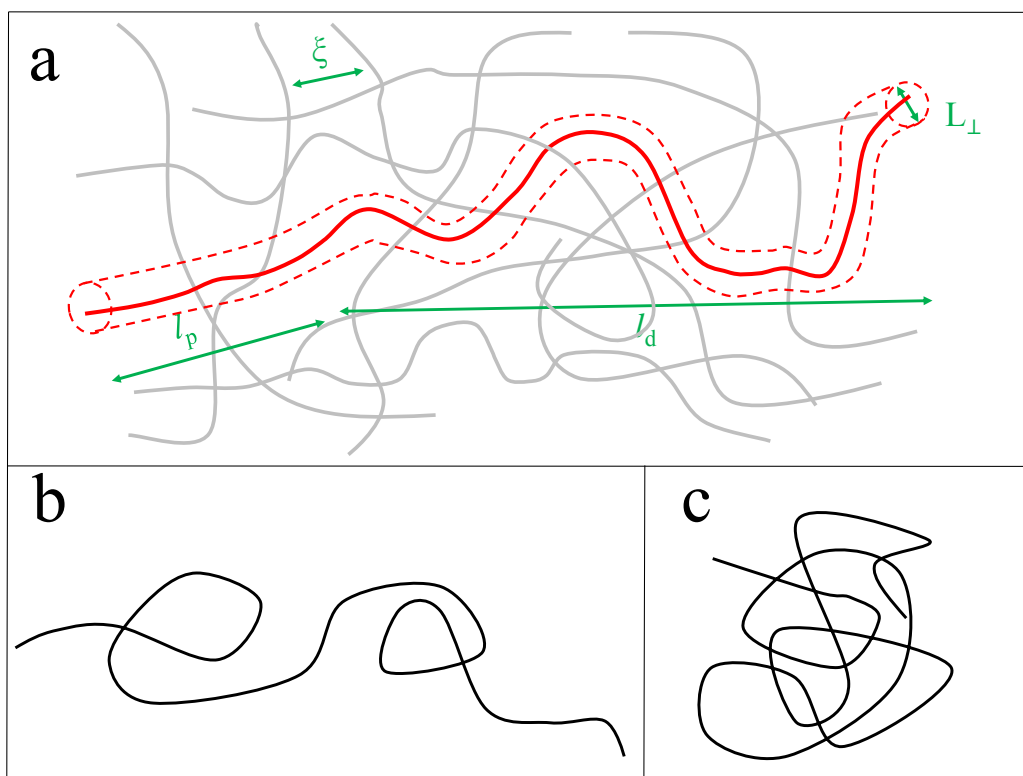


Figure S10: (a) illustration of relevant length scales in entangled networks of semiflexible polymer: tube diameter L_{\perp} , mesh size ξ , deflection length l_d and persistence length l_p . (b) Schematic images of a semiflexible polymer molecule in extension state and (c) Schematic images of a semi-flexible polymer molecule in coiled state.