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Supplementary Information for

The Contraction of PEDOT Films Formed on a Macromolecular Liquid-like Surface

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The main vibration of PEDOT are list in Tale S1. The results show the asymmetric and symmetric C=C vibrations are shifted from 1527 and 1416 cm⁻¹ to 1488 and 1439 cm⁻¹, respectively, which are due to structure change from quinoid to benzenoid structure¹. The peaks at 1668 and 1644 cm⁻¹ are due to the bending of H-O-H from absorbed water.²⁻⁴

Assignment	cm ⁻¹			
	contracted	"stretched"	PEDOT+Oxidizer	shift
	PEDOT	PEDOT		
v (HOH)			1668 and 1644	
v _{as} (C=C) _{Th}	1546 (sh)	1546 (sh)	1541	5
v _{as} (C=C) _{Th}	1527	1527	1526 and 1488	39
v _s (C=C) _{Th}	1416	1416	1439	23
v(C-C) _{Th}	1373	1373	1365	8
v (CC) _{inter-ring}	1272	1272		
v _{as} (S=O) _{Tos}	1242	1242		
v (COC) _{ED}	1163	1163		
v _s (S=O) _{Tos}	1122	1122		
v _s (S=O) _{Tos}	1112	1112		
v (COC) _{ED}	1061	1061		
v (S-	1012	1012		
phenyl) _{Tos}				
v (CSC) _{Th}	954	954		
γ (CH) _{ED}	923	923		
v(CSC) _{Th}	840	840		

 Table S1.
 Infrared band assignments for PEDOT.^{1, 4}

(Note: δ bending; v stretching; as asymmetric; s symmetric; and sh shoulder.)

The peak assignment of PEG-PPG-PEG is listed below in Table S2.

Table S2. Infrared band assignment for PEG-PPG-PEG.⁵⁻⁷

Assignment	PEG-PPG-PEG (cm ⁻¹)
δ (CH ₃)+ γ(CH ₃)	1648 and 1601
δ(CH ₂)	1497 and 1453
ν(CC)+ ω(CH ₃)	1374
ν(CC)+ ω(CH ₃)	1349
τ(CH ₂)	1248
v(CC)+ v(COC)	1146
v(COC)	1129, 1098, 1033 and 1007
$\rho(CH_3) + \rho(CH_2)$	844, 816 and 680

(Note: δ bending; ω wagging; ν stretching; ρ rocking; τ twisting; γ deformation)

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Supplementary Figure S1. (A) Contracted PEDOT films after washing. (B) A contracted PEDOT stripe shows 16% contraction in length after washing. The blue line marks its original length on the PDMS substrate. (C) PEDOT:tosylate on PDMS cracks at 10% strain.



Supplementary Figure S2. Surface reflectance of (A) contracted and (B) "stretched" PEDOT films at different thicknesses. (C) Reflectance of stretched and collapsed PEDOT films. (D) absorption spectra of the two films in (C).



Supplementary Figure S3. UPS spectra of contracted and "stretched" PEDOT.



Supplementary Figure S4. XRD spectra of PEDOT films