

Supplementary information

Fluid separation and network deformation in wetting of soft and swollen surfaces

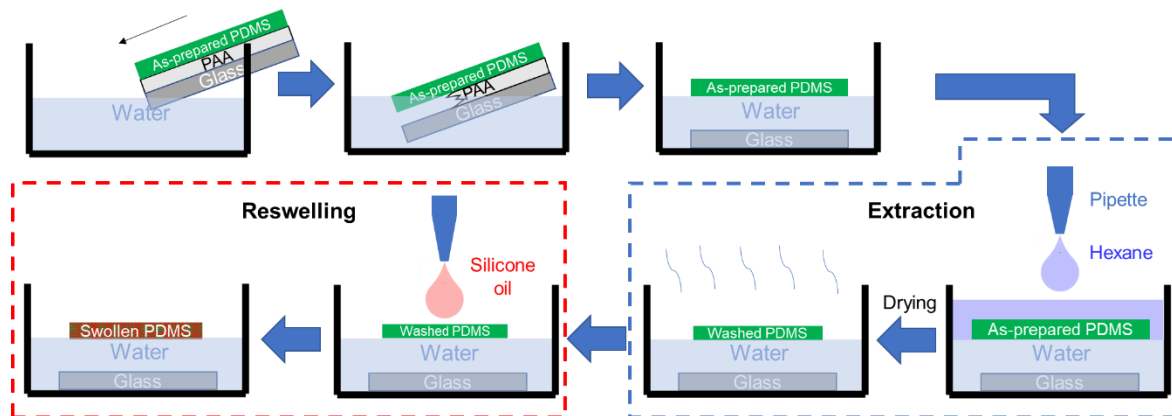
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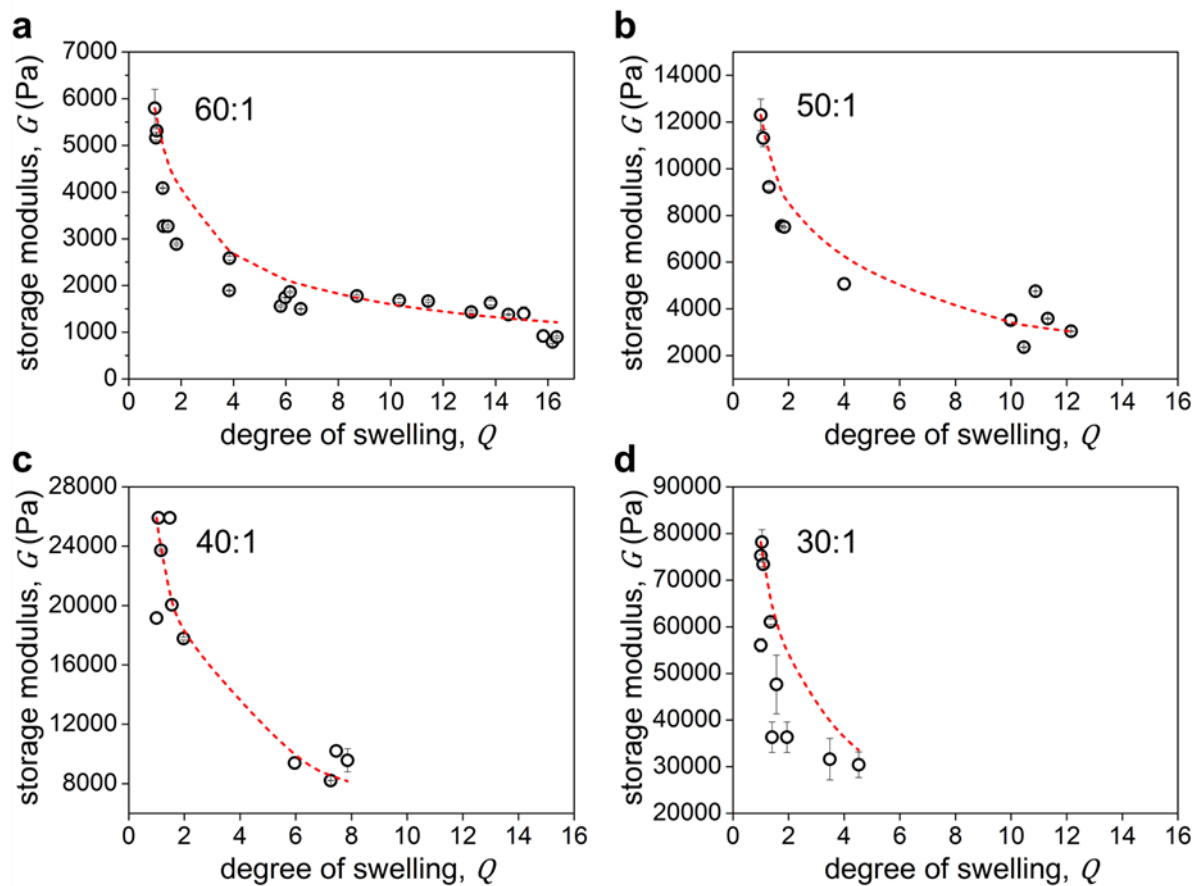
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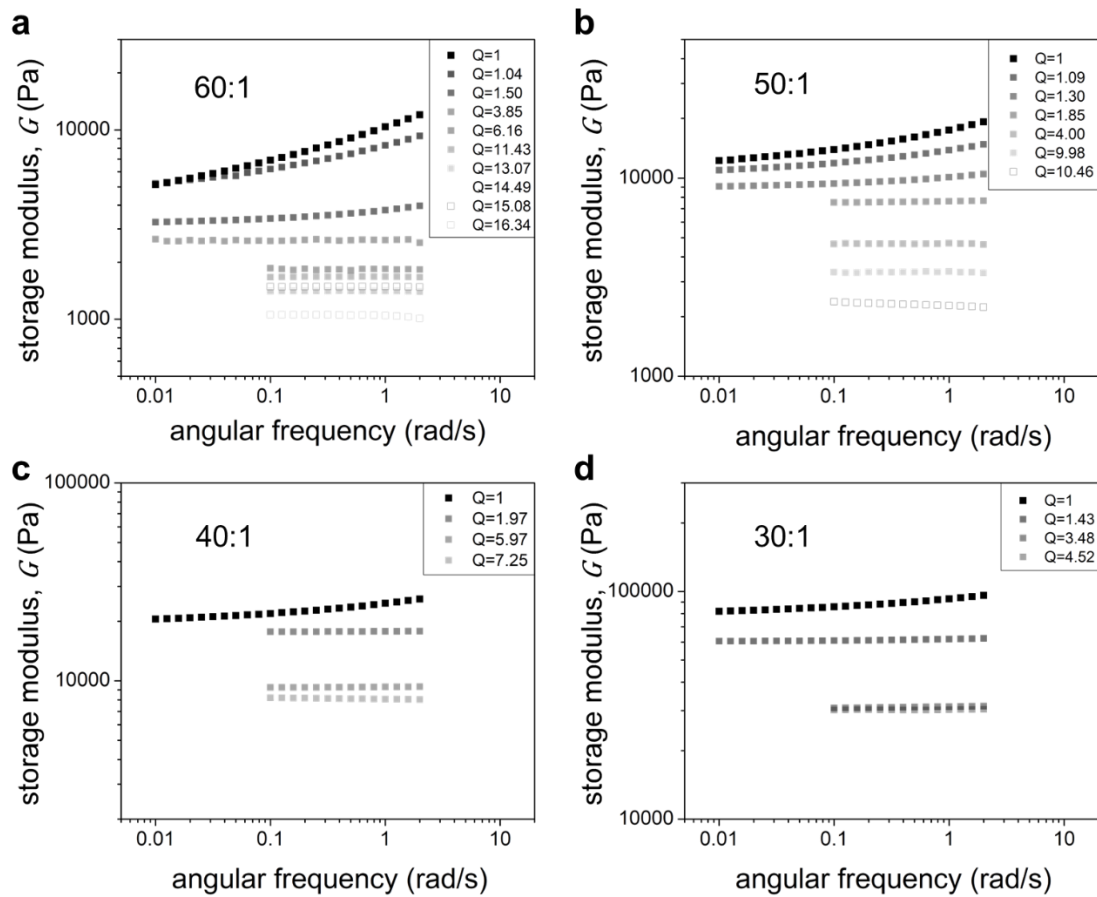
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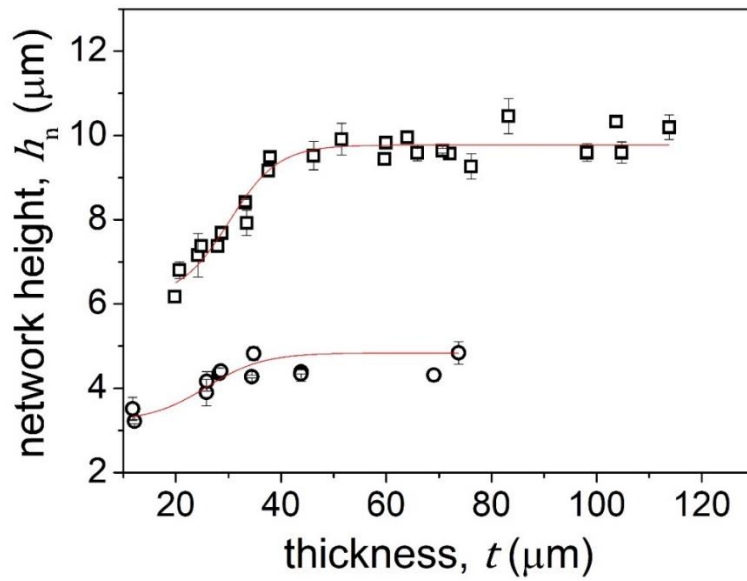
Supplementary Figure 1 | Schematic of interfacial extraction and swelling method. The as-prepared PDMS films are released on a water surface by dissolving the sacrificial PAA underlayer. For extraction, hexane was then carefully added to the petri dishes to swell the network and allow uncrosslinked molecules to migrate into the surrounding media. During this procedure, the film remains at the hexane–water interface. To swell the sample with the desired silicone oil fluid, drops of dyed silicone oil are added directly onto the extracted film when it is still floating on the water surface.



Supplementary Figure 2 | Shear modulus. Plots of shear modulus vs. swelling ratio of (a) 60:1, (b) 50:1, (c) 40:1 and (d) 30:1 base/crosslinker mixing ratios. The dashed lines are calculated values given by $G = G_{\text{dry}}\phi^{0.56}$.

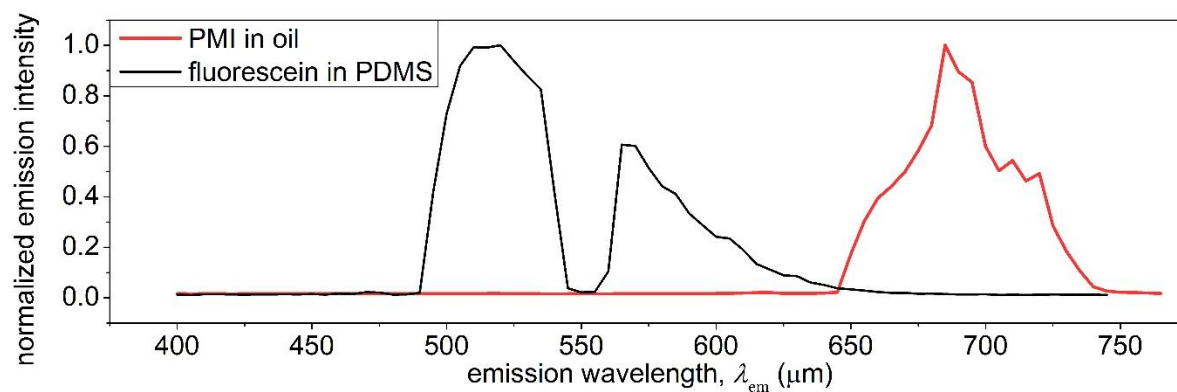


Supplementary Figure 3| Storage modulus vs. frequency. Plots of shear storage modulus (G) vs. frequency for the different degrees of swelling Q with as-prepared mixing ratios of (a) 60:1, (b) 50:1, (c) 40:1 and (d) 30:1.

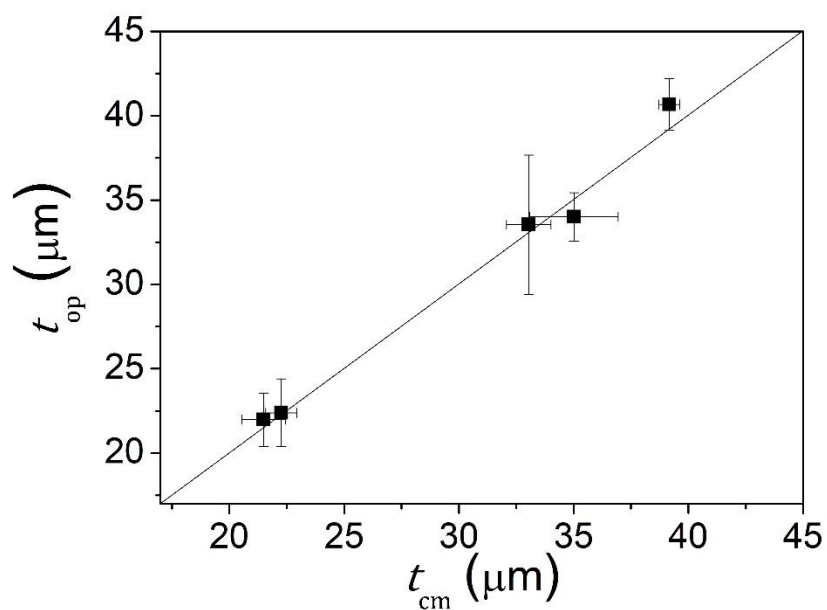


Supplementary Figure 4 | Height of wetting ridge as a function of substrate thickness.

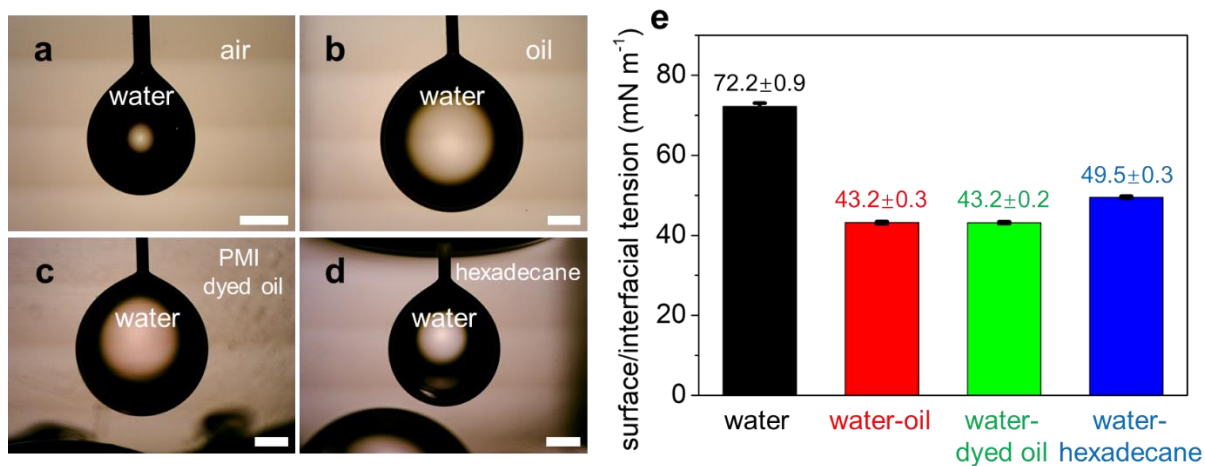
Examples of wetting ridge heights for 60:1 and 50:1 as-prepared samples. The red line shows that the height of the wetting ridge is first increasing with thickness and then remains constant at thickness values larger than $\sim 50 \mu\text{m}$ for 60:1 and $\sim 40 \mu\text{m}$ for 50:1.



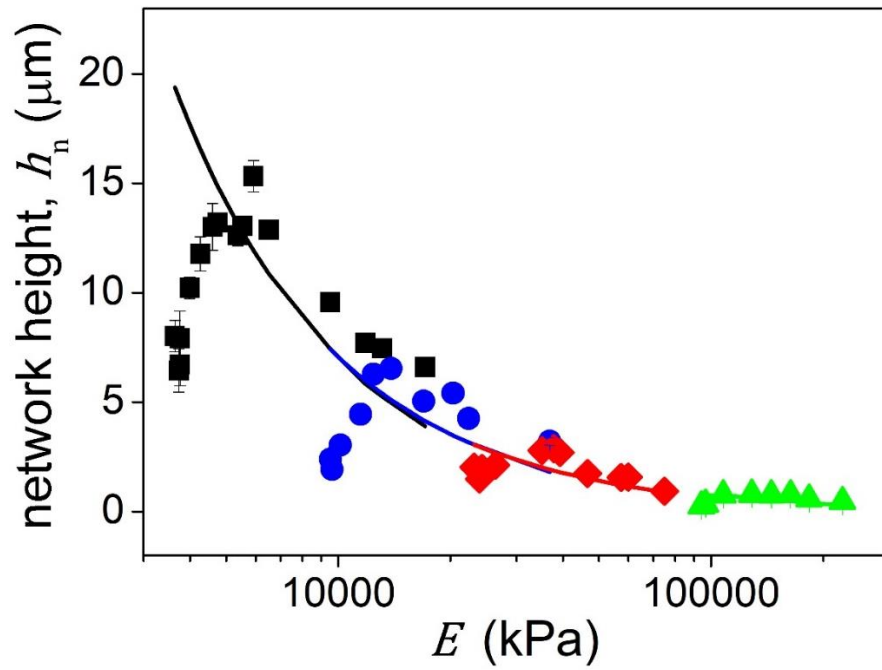
Supplementary Figure 5 | Emission spectrum. Fluorescein in PDMS is excited with a 488 nm laser (black line) and Perylene monoimide in silicone oil is excited with 638 nm laser (red line). This shows a non-overlapping emission of the two dyes.



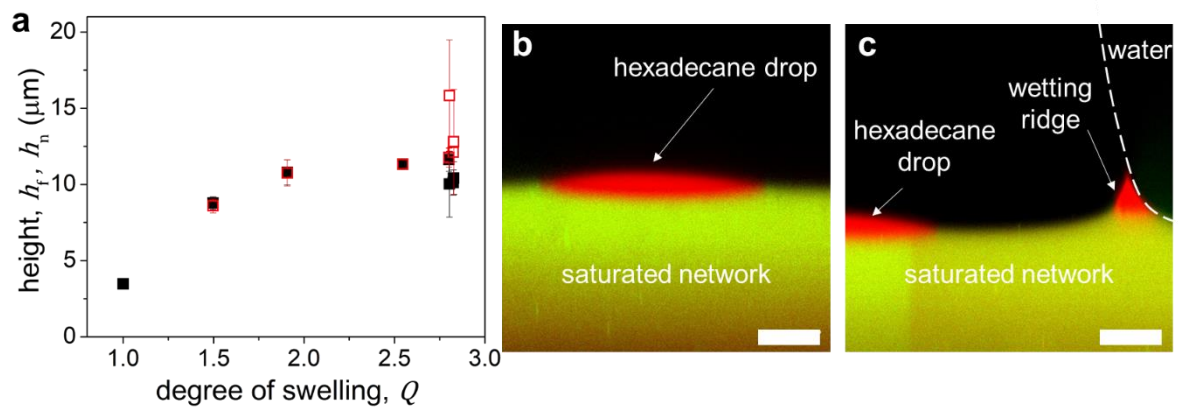
Supplementary Figure 6| Thickness measurements by optical profilometry and confocal microscopy. A plot comparing the thickness measurements from confocal microscopy to optical profilometry. t_{op} : the thickness measured by the optical profilometry, and t_{cm} : the thickness measured by the confocal microscopy. Optical profilometry measurements were taken on a Filmetrics Profilm 3D.



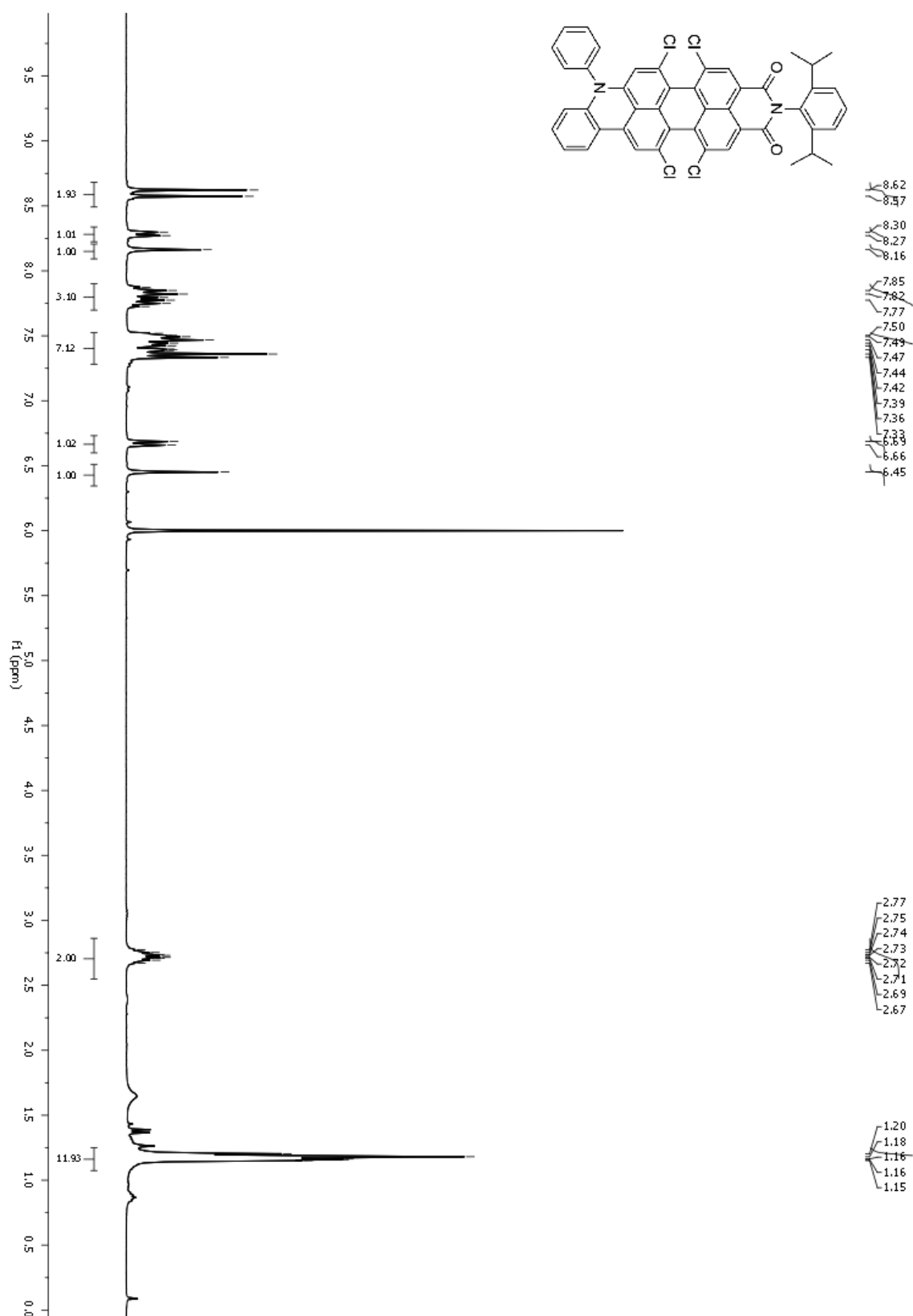
Supplementary Figure 7 | Surface/interfacial tensions. A pendant water drop in (a) air, (b) silicone oil, (c) PMI dyed silicone oil and (d) n-hexadecane. Scale bars: 1mm. (e) A bar plot showing the surface/interfacial tensions of water-air, water-oil, water-PMI dyed oil, and water-hexadecane.



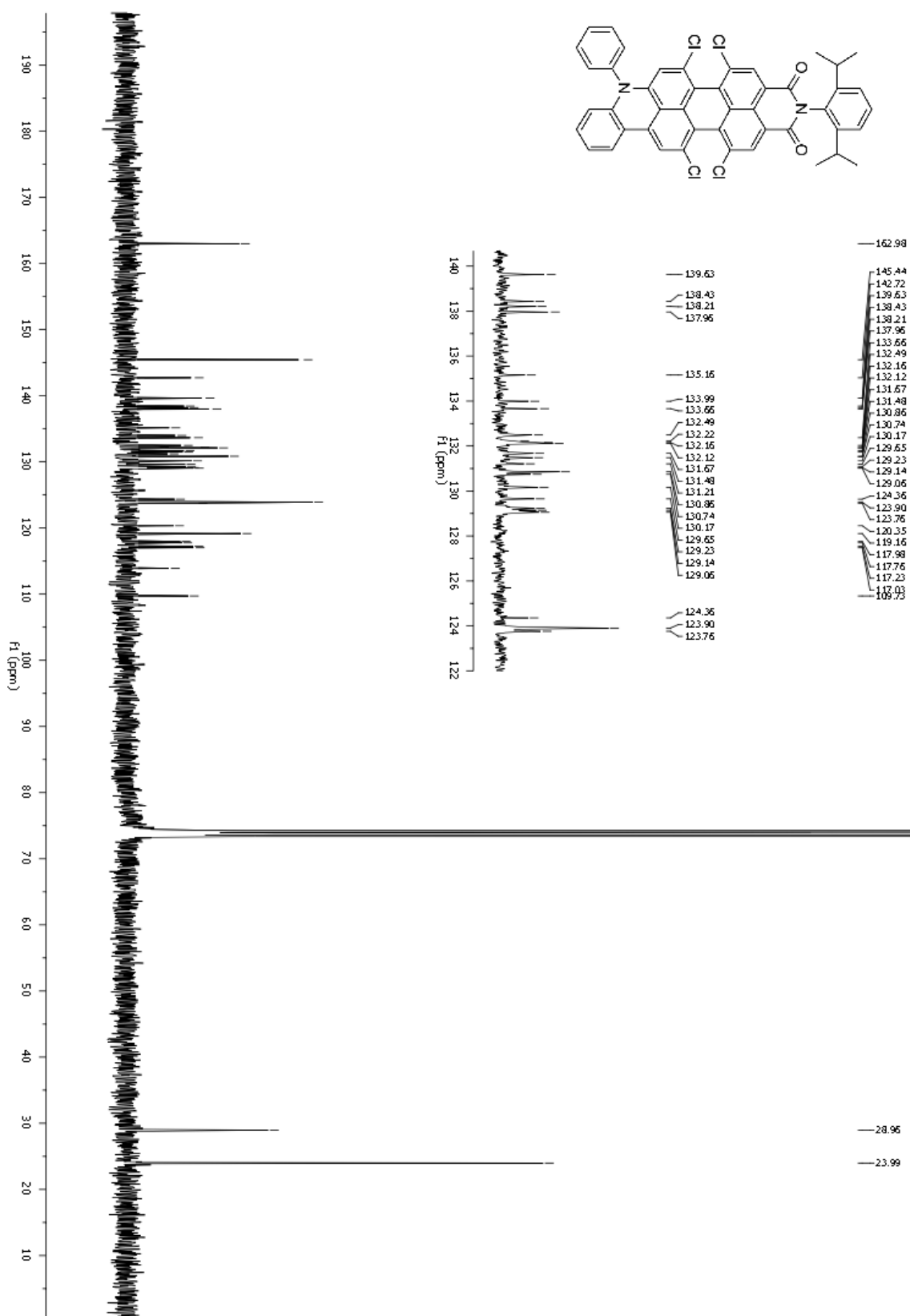
Supplementary Figure 8 | Network height vs. modulus. A plot of the network height (h_n) vs. modulus (E) for 60:1 (black squares), 50:1 (blue circles), 40:1 (red diamonds), and 30:1 (green triangles) samples. The solid lines are calculated values given by $h_n = \gamma_w \sin\theta / E$.



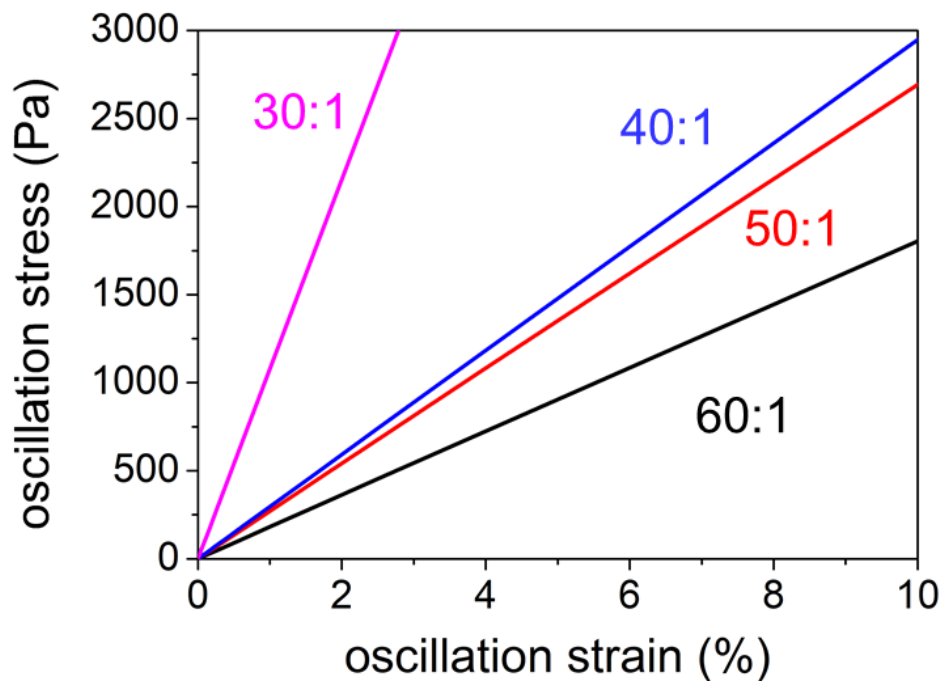
Supplementary Figure 9 | Hexadecane swollen samples. (a) A plot of network height h_n (black) and fluid height h_f (red) as a function of the degree of swelling (Q) of hexadecane swollen samples. (b) Confocal image showing a small excess hexadecane droplet on a saturated network. (c) The hexadecane droplets can be found near a wetting ridge. The fluid zone of the wetting ridge likely comes from the excess droplets. Scale bars: $20 \mu\text{m}$.



Supplementary Figure 10 | ^1H NMR spectrum. ^1H NMR spectrum of Compound 2 in 1,1,2,2-tetrachloroethane- d_2 (see Supplementary Note 1).



Supplementary Figure 11 | ^{13}C NMR spectrum. ^{13}C NMR spectrum of Compound 2 in 1,1,2,2-tetrachloroethane- d_2 (see Supplementary Note 1).

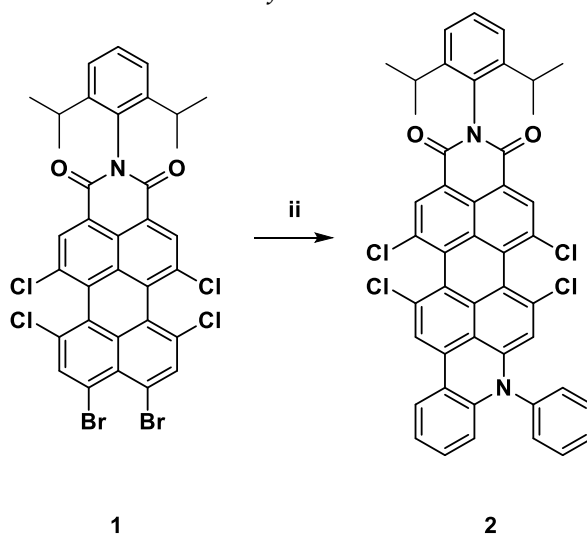


Supplementary Figure 12 | Stress vs. strain. A plot of stress vs. strain for dry 60:1, 50:1, 40:1, and 30:1 samples with at a frequency of 10 Hz. The samples remain in the linear region up to 10% strain, which is used for the frequency sweeps to quantify modulus values.

Supplementary Note 1 | PMI preparation and characterization

Experimental details of PMI modification. The progress of all reactions was monitored by thin layer chromatography (Macherey- Nagel F 254 silica gel sheet) with appropriate solvents described for each compound in the synthetic procedures. Column chromatography was performed with dichloromethane (Fisher Scientific), hexane (Sigma-Aldrich) on silica gel (Macherey-Nagel, Si60). FD mass spectra were performed with a VG-Instrument ZAB 2-SE-FDP. ^1H and $^{13}\text{C}\{^1\text{H}\}$ NMR spectra were recorded on Bruker AC300 NMR spectrometers using the residual proton or the carbon signal of the deuterated solvent as an internal standard. Chemical shifts are reported in parts per million. The high-resolution mass spectra were carried out by the Microanalytical Laboratory of Johannes Gutenberg University. UV/Vis absorption spectra were recorded on a Perkin Elmer Lambda 900 spectrophotometer. Fluorescence emission spectra were recorded on a J&M Tidas spectrometer. The fluorescence quantum yield measurement was using rhodamine 800 in ethanol as standard (25%). The absorbance of each sample in the 10 mm fluorescence cuvette was < 0.1 at the excitation wavelength. All chemicals and solvents were purchased from commercial suppliers and used without further purification unless otherwise specified. The 8,9-dibromo-5,6,11,12-tetrachloro-2-(2,6-diisopropylphenyl)-1H-benzo[5,10]anthra[2,1,9-def]isoquinoline-1,3(2H)-dione (**1**) was synthesized according the literature¹. All reported yields are isolated yields.

Preparation Procedures and characterization of the red PMI (2)



5,6,14,15-tetrachloro-2-(2,6-diisopropylphenyl)-8-

phenylisoquinolino[6',5',4':10,5,6]anthra[2,1,9-mna]acridine-1,3(2H,8H)-dione (2):

A solution of 8,9-dibromo-5,6,11,12-tetrachloro-2-(2,6-diisopropylphenyl)-1H-benzo[5,10]anthra[2,1,9-def]isoquinoline-1,3(2H)-dione (0.78 g, 1.00 mmol), bis(phenyl)amine (1.10 mmol), Pd(OAc)₂ (5 mmol%), Pd₂(dba)₃ (5 mmol%) sodium-*tert*-butoxide (0.48g, 5.02 mmol), tricyclohexylphosphine (10 mol%), tri-*tert*-butylphosphine (10 mol%) in 50 ml toluene was stirred at 100°C under argon atmosphere for 24 h. The solvent was removed under reduced pressure. The crude product was purified by column chromatography using dichloromethane/hexane (9:1) as eluent on silica. Yield: 0.50 g (63%).

¹H NMR (300 MHz, C₂D₂Cl₄, 300K): 1.15-1.20 (m, 12H, CH₃); 2.67-2.77 (m, 2H, CH); 6.45 (s, 1H); 6.67 (d, 1H, ³J_{HH} = 8.2 Hz); 7.33-7.52 (m, 7H); 7.72-7.87 (m, 3H); 8.16 (s, 1H); 8.28 (d, 1H, ³J_{HH} = 7.9 Hz); 8.57 (s, 1H); 8.62 (s, 1H). ¹³C NMR (75.0 MHz, C₂D₂Cl₄, 300K): 23.99 (4C, CH₃); 28.96 (2C, CH); 109.73 (1C) 113.92 (1C); 117.03 (1C); 117.23 (1C); 117.76 (1C); 117.98 (1C); 119.16 (1C); 120.35 (1C); 123.76 (1C); 123.90 (2C); 124.36 (1C); 129.06 (1C); 129.14 (1C); 129.23 (1C); 129.65 (1C); 130.17 (1C); 130.74 (1C); 130.86 (1C); 131.21 (1C); 131.48 (1C); 131.67 (1C); 132.12 (1C); 132.16 (1C); 132.22 (1C); 132.49 (1C); 133.66 (1C); 133.99 (1C); 135.16 (1C); 137.96 (1C); 138.21 (1C); 138.43 (1C); 139.63 (1C); 142.72(1C); 145.44 (2C); 162.98 (2C, C=O). HRMS *m/z* calculated for C₄₆H₃₀Cl₄N₂O₂ 782.1061 found 782.1067. UV-Vis (CH₂Cl₂): λ_{abs} = 679 (58 200) nm (M⁻¹cm⁻¹), λ_{em} = 746 nm (Φ_f =62%).

Supplementary References

1. Abul-Futouh, H. *et al.* Toward a Tunable Synthetic [FeFe]-Hydrogenase H-Cluster Mimic Mediated by Perylene Monoimide Model Complexes: Insight into Molecular Structures and Electrochemical Characteristics. *Organometallics* **37**, 3278–3285 (2018).