Supporting information

Mechanical reinforcement of lamellar bilayer hydrogels by small amounts of co-surfactants

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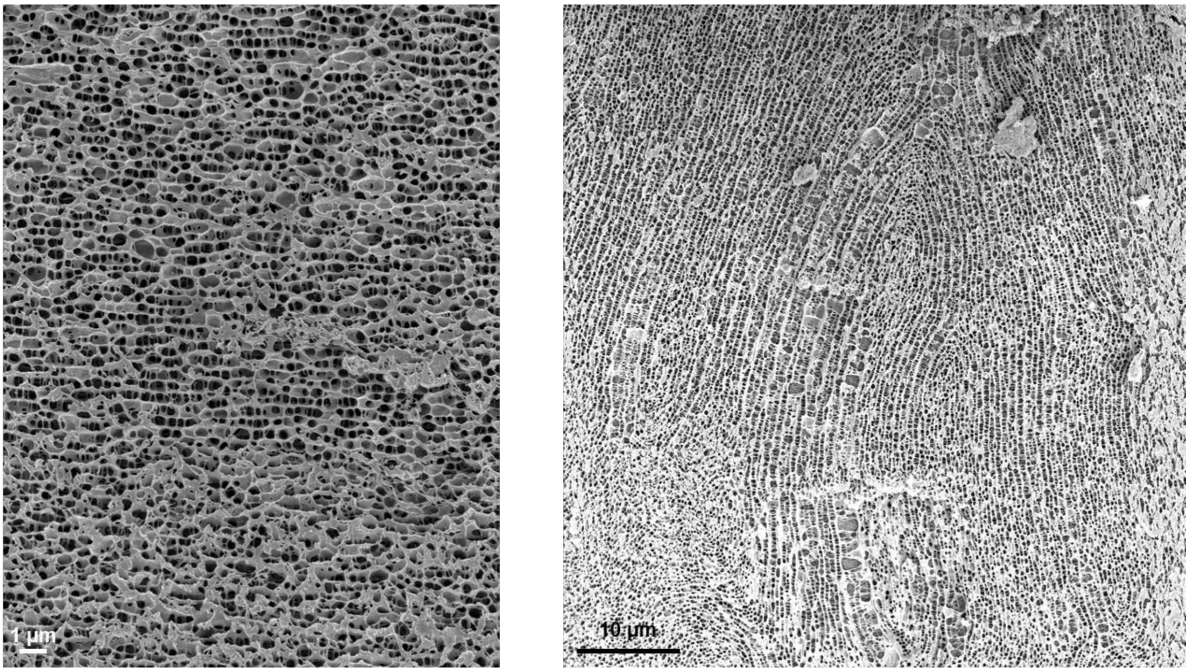

Figure S1. Cryo-scanning electron micrograph of the cross-section of the sample PDGI/PAAm with NBA at R=1:64. Left: PDGI bilayers showing good alignment parallel to the glass plates of the mold. Right: another part of the cross-section displaying PDGI bilayers curved like digital imprints, mostly perpendicular to the glass plates of the mold.

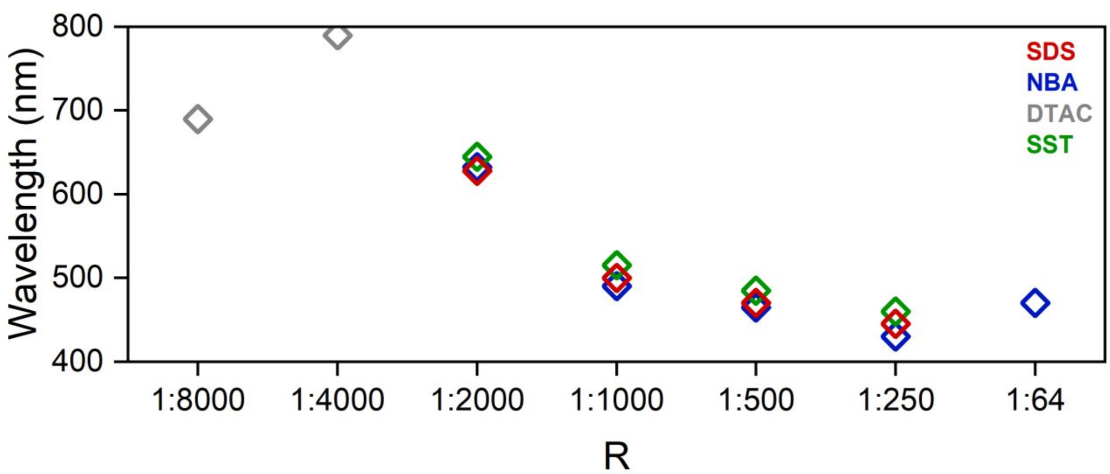

Figure S2. Wavelength at maximum reflectance intensity of PDGI/PAAm hydrogels exhibiting structural colors.

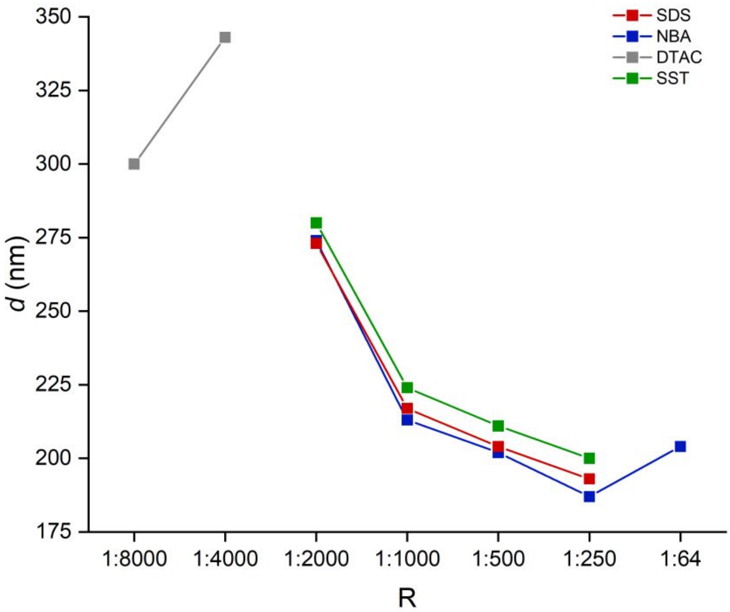

Figure S3. Lamellar bilayers *d*-spacing in PDGI/PAAm hydrogels with different co-surfactants to DGI molar ratios.

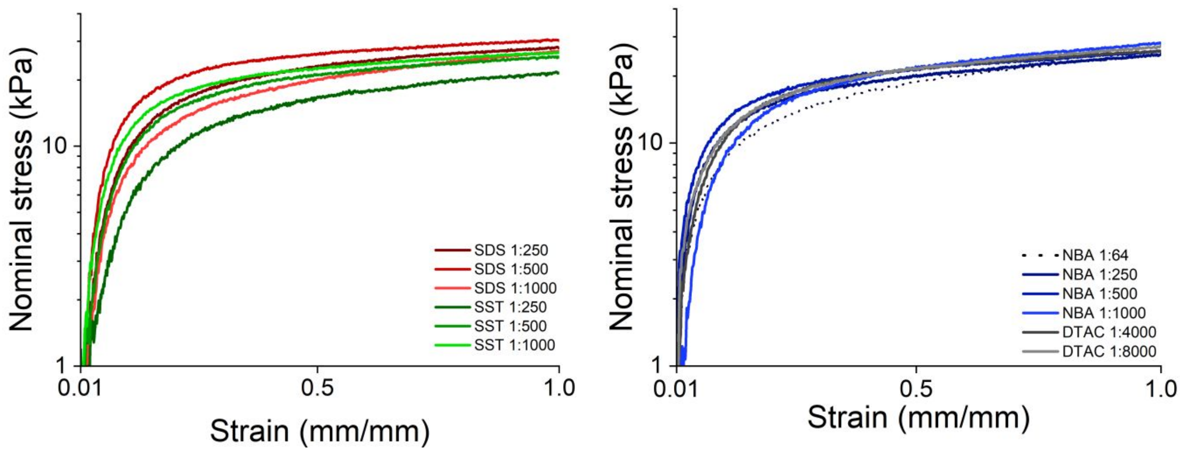

Figure S4. Semi-log plots of the small strain tensile behavior of PDGI/PAAm hydrogels with SDS or SST co-surfactants (left) or NBA or DTAC co-surfactants (right) depicted in Figure 3a and 3b as insets, respectively.

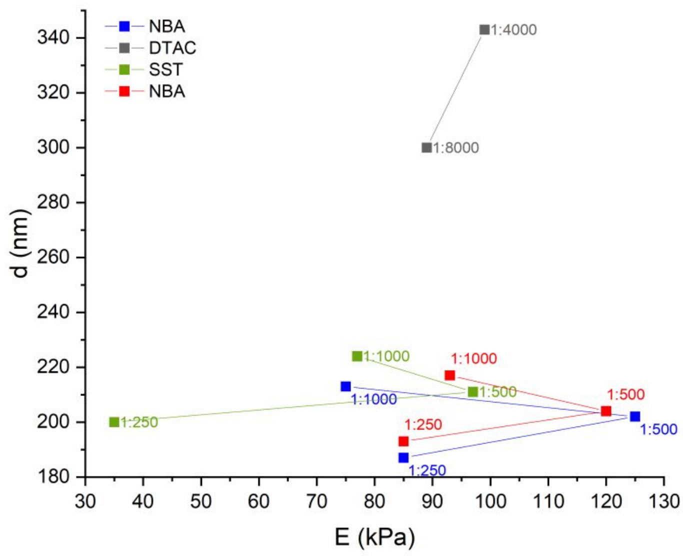

Figure S5. Interlamellar distance plotted against Young's modulus of lamellar bilayer hydrogels at different R.

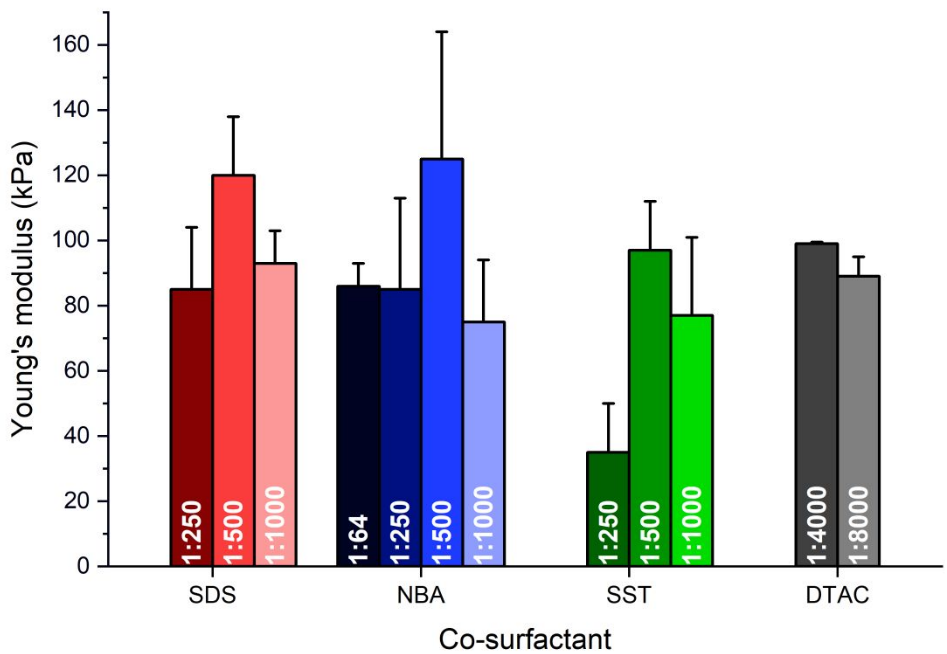

Figure S6. Young's moduli of PDGI/PAAm hydrogels with different co-surfactants at different R, at equilibrium swelling state.

The Young modulus of the bilayers E_b was calculated using the following equation[1]:

$Ed = E_b t_b + E_g t_g$ (1)

Here, E is the Young modulus of the PDGI/PAAm hydrogels and E_g that of pure PAAm hydrogel, t_b=5 nm is the thickness of a bilayer, t_g is the thickness of a PAAm gel layer that can be estimated from the inter-bilayer distance at equilibrium swelling (Figure S3) $d = t_b + t_g$. E_g was estimated as 10kPa.

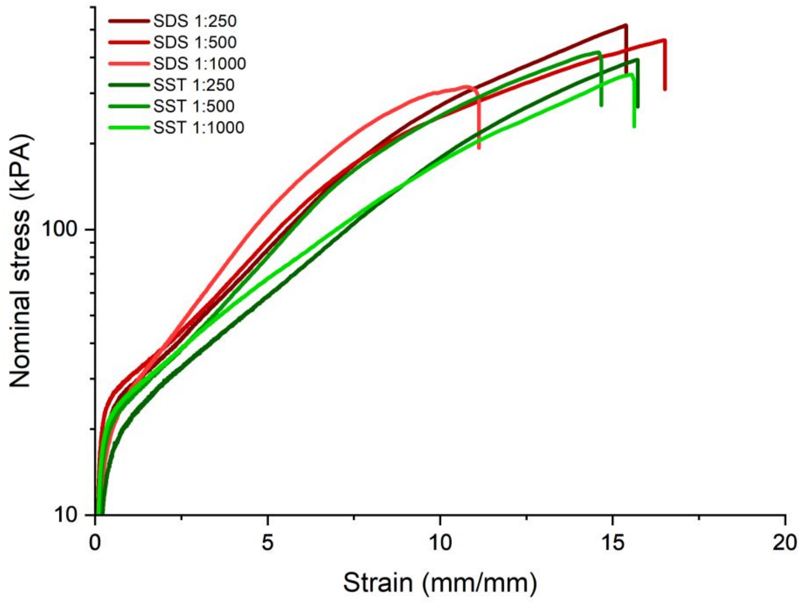

Figure S7. Semi-log plot of the large strain tensile behavior of PDGI/PAAm hydrogels with SDS or SST co-surfactants depicted in Figure 3a.

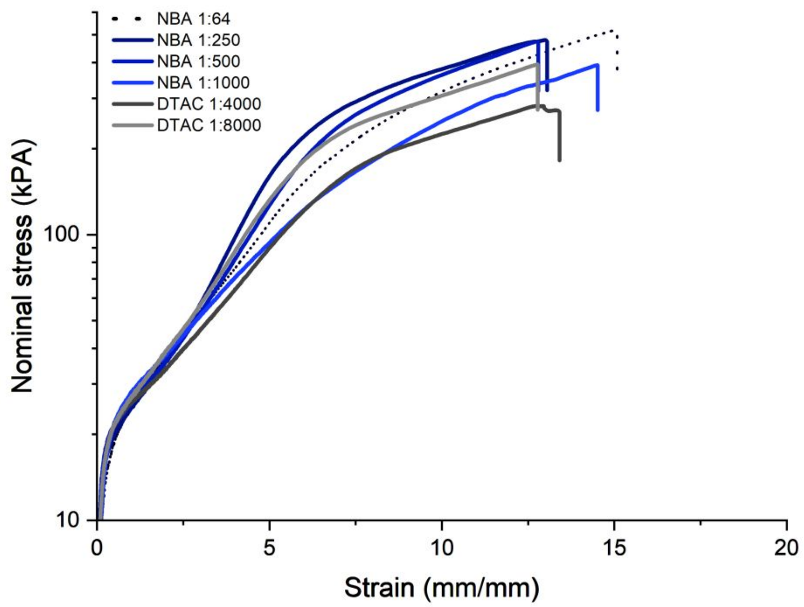

Figure S8. Semi-log plot of the large strain tensile behavior of PDGI/PAAm hydrogels with NBA or DTAC co-surfactants depicted in Figure 3b.

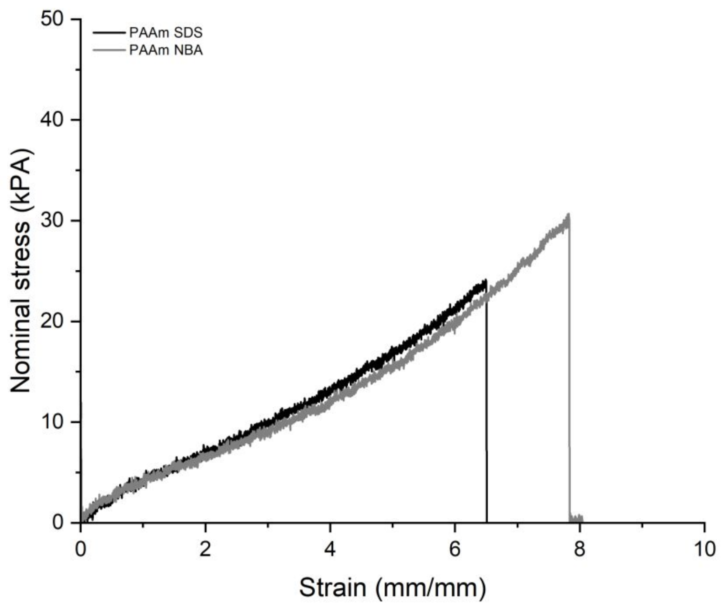

Figure S9. Tensile behavior of PAAm hydrogels without DGI at equilibrium swelling state, synthesized either with SDS or NBA at “R=1:250” exhibiting identical mechanical behavior.

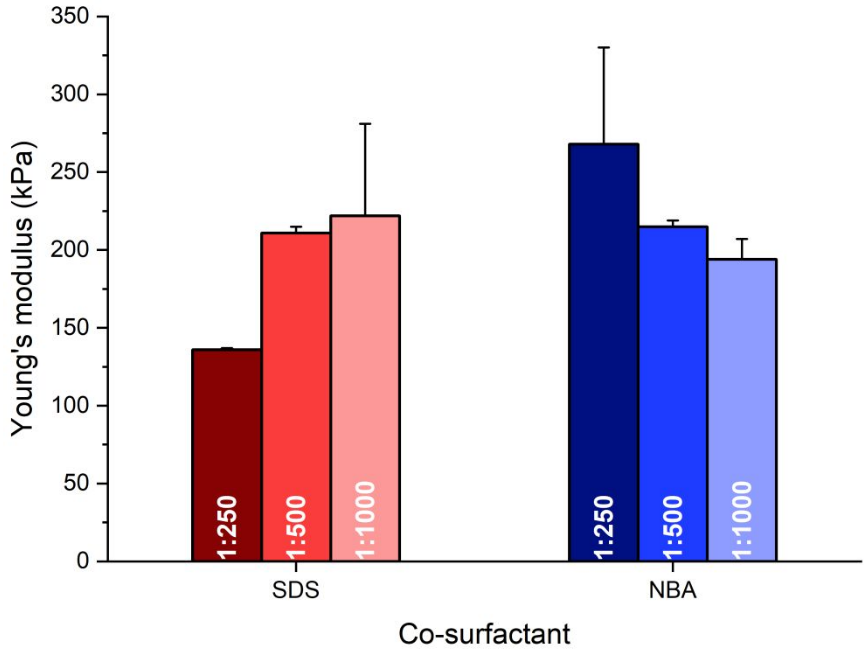

Figure S10. Young's moduli of as-prepared PDGI/PAAm lamellar hydrogels with co-surfactants SDS or NBA at different R.

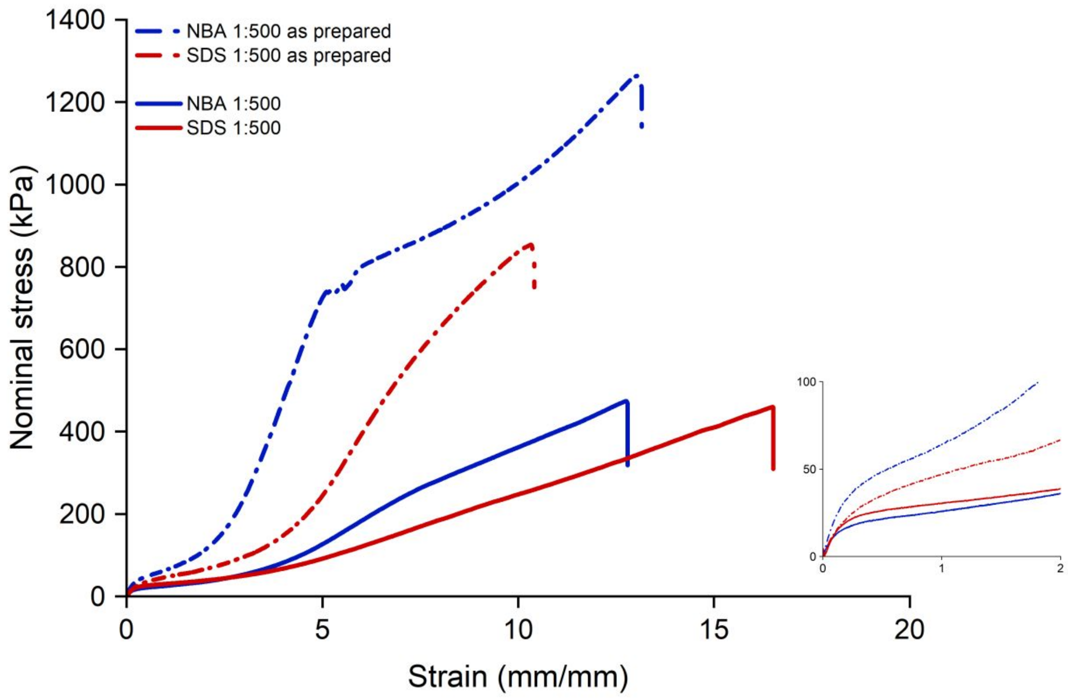

Figure S11. Tensile behavior of PDGI/PAAm hydrogels with SDS or NBA co-surfactants at same R=1:500 compared at as-prepared state and equilibrium swelling state before correction of swelling. Inset shows the early stages of deformation.

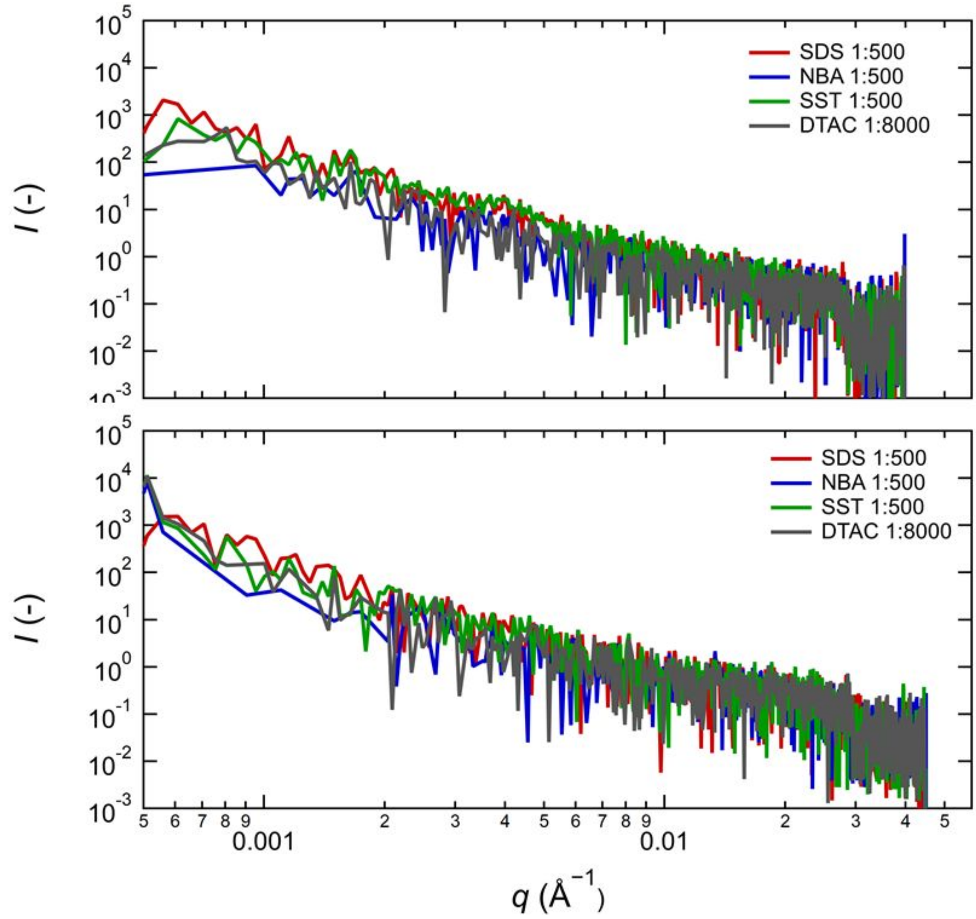

Figure S12. 1D USAXS intensity *I vs*. scattering vector, *q* obtained from 2D USAXS images of the top view of un-stretched samples (strain 0%). Top: integration along the equatorial direction; bottom: integration perpendicular to the equatorial direction. No correlation peaks were observed.

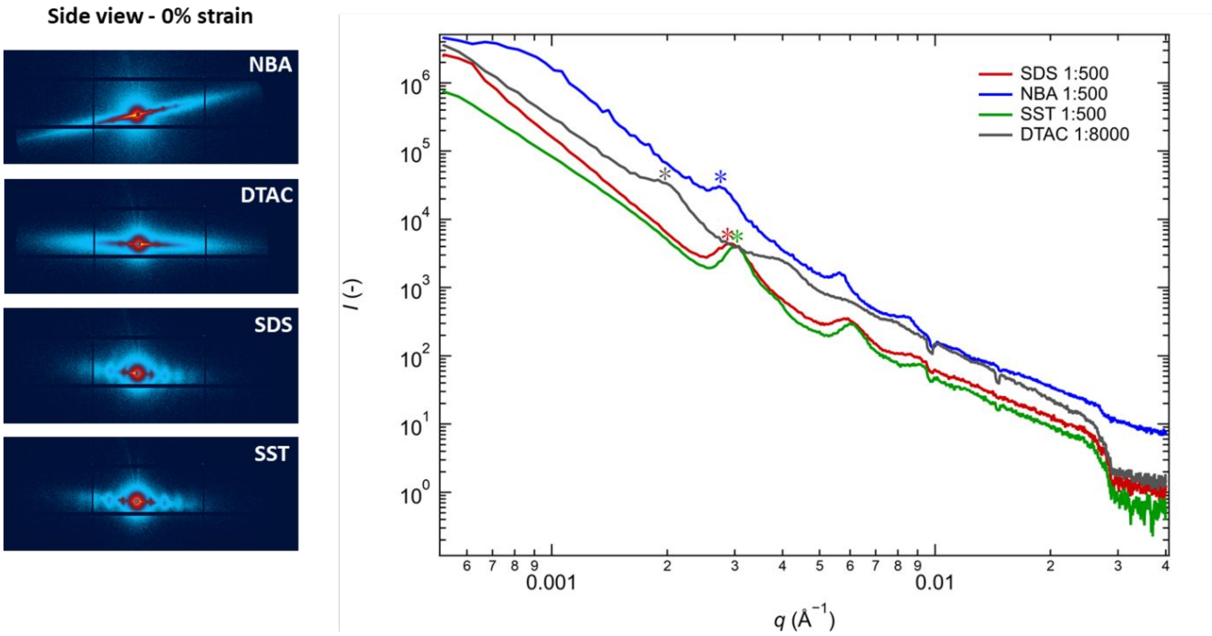

Figure S13. Left: 2D USAXS images of the side view of un-stretched samples (perpendicular to the bilayers) showing patterns typical of lamellar structure[2]. Right: 1D USAXS intensity *I* *vs*. scattering vector, *q*. Stars highlight the first order scattering peak.

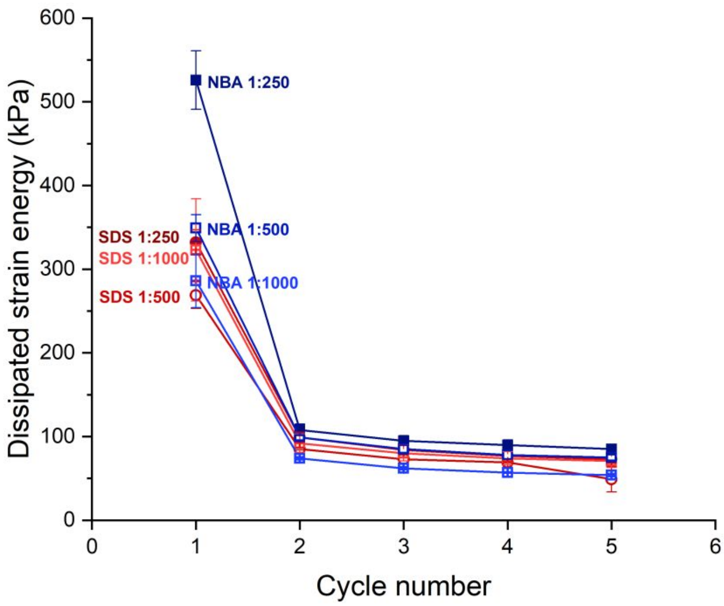

Figure S14. Dissipated strain energy (calculated from the area of the hysteresis loop) from cycles 1 to 5 of PDGI/PAAm hydrogels with SDS or NBA co-surfactants (R=1:250, 1:500 or 1:1000) during cyclic tensile loading.

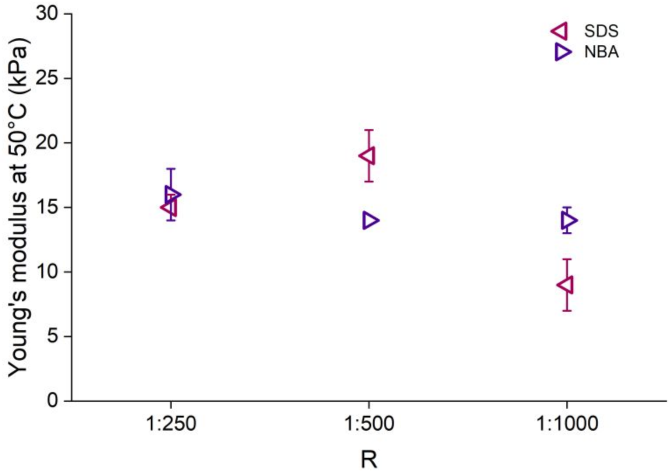

Figure S15. Young's modulus of PDGI/PAAm hydrogels with SDS or NBA co-surfactants at different R, at 50°C.

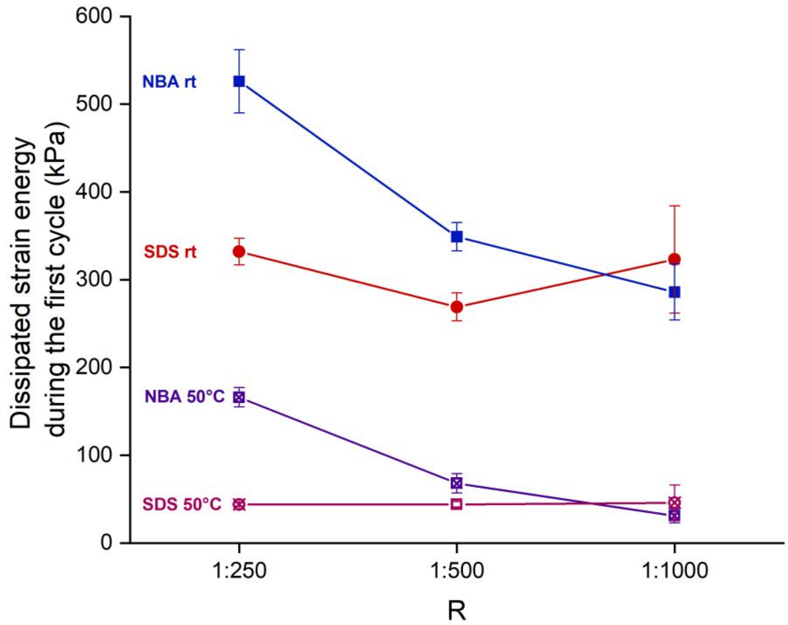

Figure S16. Dissipated strain energy of PDGI/PAAm hydrogels with various amounts of SDS or NBA co-surfactants during the first cycle of tensile cyclic test at room temperature or 50°C.

References

(1) Haque, M. A.; Kamita, G.; Kurokawa, T.; Tsujii, K.; Gong, J. P. Unidirectional Alignment of Lamellar Bilayer in Hydrogel: One-Dimensional Swelling, Anisotropic Modulus, and Stress/Strain Tunable Structural Color. *Adv. Mater.* **2010**, *22* (45), 5110–5114. https://doi.org/10.1002/adma.201002509.

(2) Haque, M. A.; Cui, K.; Ilyas, M.; Kurokawa, T.; Marcellan, A.; Brulet, A.; Takahashi, R.; Nakajima, T.; Gong, J. P. Lamellar Bilayer to Fibril Structure Transformation of Tough Photonic Hydrogel under Elongation. *Macromolecules* **2020**, *53* (12), 4711–4721. https://doi.org/10.1021/acs.macromol.0c00878.