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## **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<sup>1</sup>:

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

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 cosurfactants 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<sup>2</sup>. 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.

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