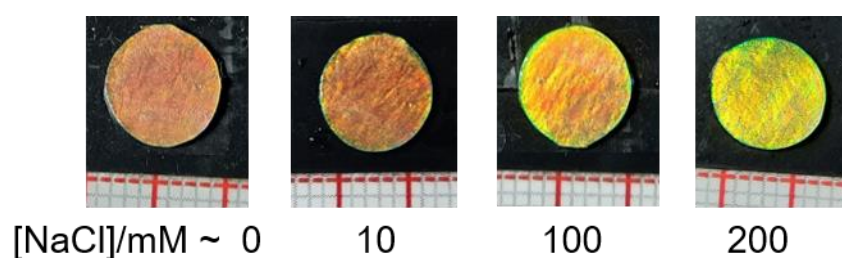


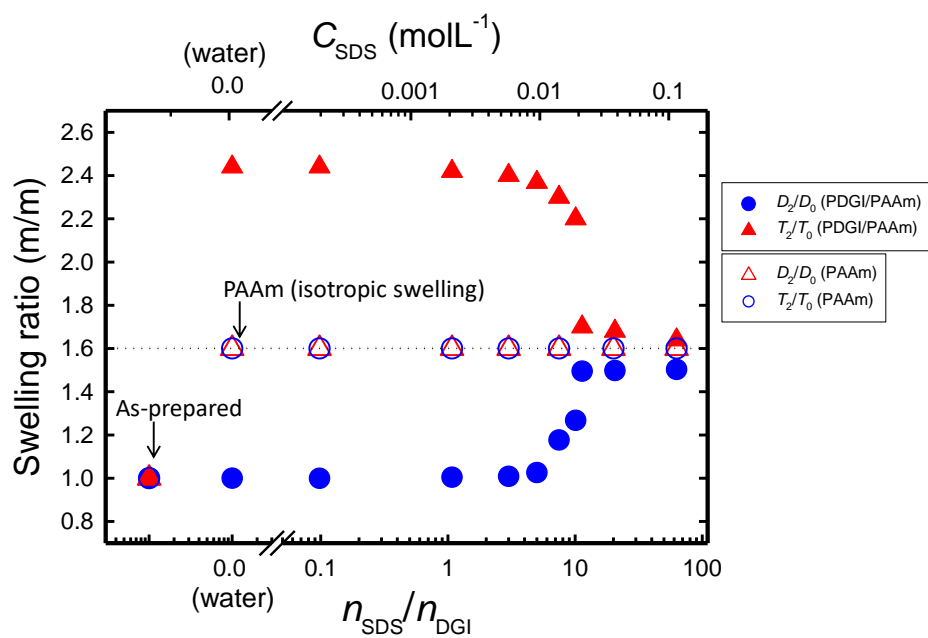
### Supplementary Information

## Surfactant induced bilayer-micelle transition for emergence of functions in anisotropic hydrogel

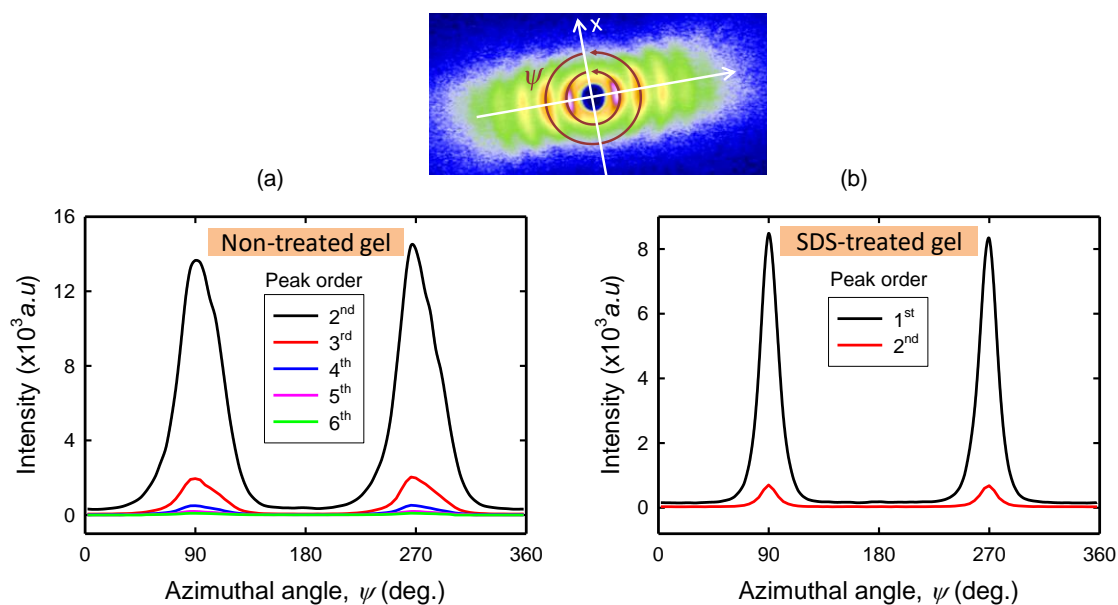
Md Anamul Haque<sup>a,b</sup>, Takayuki Kurokawa<sup>b,c</sup>, Tasuku Nakajima<sup>b,c</sup>, Gen Kamita<sup>d</sup>, Zannatul Fatema<sup>a</sup>, and Jian Ping Gong<sup>b,c,\*</sup>



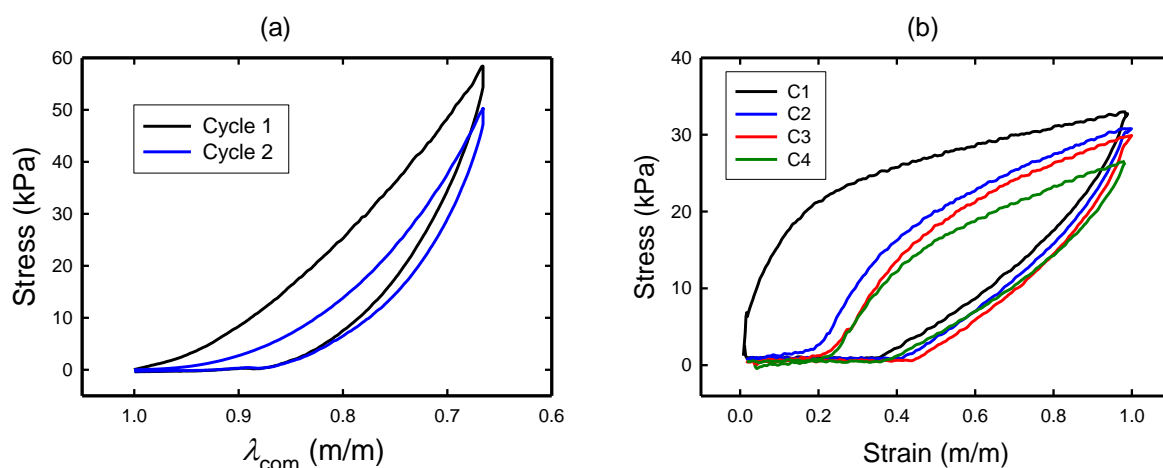
**Figure S1:** Photographs of the PDGI/PAAm gel treated with various NaCl concentrations (0 to 100 mM). The water swollen PDGI/PAAm gel was immersed and kept sequentially in 10, 100, 200 mM NaCl solution for 7 days. The color and diameter of the gel remain unchanged at 10 mM NaCl solution. At 100 and 200 mM NaCl solutions, small blue shift of color from orange to yellow was noticed whereas the diameter of the gel remains nearly unchanged. In contrast, the PDGI/PAAm gel shows significant change in color and diameter at SDS concentration of 10 mM and above. The slight color change is apparently due to decrease of the electrostatic repulsion (as a result of used-SDS present in the preparation of gel) between the bilayers. The ion shielding effect of NaCl on the SDS present in the bilayers. Therefore, in comparison with the gel treated with SDS solution, ion shielding or NaCl has no significant effect on the lamellar structure.



**Figure S2:** Swelling ratio of PDGI/PAAm gel normalized to the as prepared state ( $D_2/D_0$ ,  $T_2/T_0$ ). The gels swelled only along the thickness direction ( $T_2/T_0 \sim 2.4$ ) while the diameter remains constant ( $D_2/D_0 \sim 1$ ) at in water and in low SDS concentration ( $n_{\text{SDS}}/n_{\text{DGI}} < 7$ ). With increasing SDS concentration, thickness decreases ( $T_2/T_0 \sim 1.6$ ) while the diameter increases ( $D_2/D_0 \sim 1.6$ ) until reaching the isotropic swelling ( $T_2/T_0 = D_2/D_0 \sim 1.6$ ), the same as that of the PAAm gel.



**Figure S3:** 1D SAXS intensity vs. azimuth angle ( $\psi$ ) curves of (a) non-treated and (b) SDS-treated ( $n_{\text{SDS}}/n_{\text{DGI}} \sim 100$ ) PDGI/PAAm gels for various scattering peak orders. 1D curves for azimuth profile were obtained by integrating 2D SAXS images at small  $q$  range ( $\pm 0.005 \text{ nm}^{-1}$ ) corresponding to peak order (1<sup>st</sup> to 2<sup>nd</sup> for SDS-treated gel and 2<sup>nd</sup> to 6<sup>th</sup> for non-treated gel).



**Figure S4:** Results showing that gel with low SDS treatment showed large hysteresis and is not recoverable with immediate subsequent loading-unloading cycles. (a) Compressive and (b) tensile stress-strain cyclic behavior for the non-treated gel to reveal the time dependent recovery. The compression is applied perpendicular to, and the tensile deformation is applied parallel to the lamellar layers. The loading-unloading cycles were performed at a velocity of 5 and 200 mm/min for compression (a) and tensile stretching (b), respectively. Initial thickness of the gel sample was 1.2 mm for the compression and gauge length was 12 mm for the tensile test.