"Turning up the heat on worm-like micelles with a hydrotopic salt in microfluidics" By Joshua J Cardiel; Ya Zhao; Pablo de la Iglesia; Lilo D Pozzo; Shen, Amy Supporting Information

## Concentration regime of the precursor solution

The rheological properties of wormlike micelles have been reported to depend on the micellar volume fraction in the semi-dilute regime [1], in which the scaling law holds. The scaling law for dilute and semi-dilute micellar solutions renders  $G_0 \sim \phi^{2.3\pm0.2}$ , where  $\phi$  is the surfactant concentration and  $G_0$  is the plateau modulus of the solution. We plotted  $G_0$  against the surfactant concentration for 3 precursor solutions at CTAB concentrations of 45 mM, 60 mM, and 75 mM, while keeping the salt to surfactant concentration ratio fixed at 0.32. The slope of the dash line yields the power of m = 2.4 (see Figure 1), similar to the value ( $m \sim 2.3\pm0.2$ ) predicted by the scaling law, suggesting that the precursor solution investigated in this work falls under the semi-dilute regime. Moreover, the weakly viscoelastic behavior of our precursor solution also suggests that the solution is within the semi-dilute regime.



Figure 1: Semi-log plot of  $G_0$  against the surfactant concentration. The slope of the dash line yields the power of m = 2.4.

## Elastic modulus measurements at different temperatures

Figure 2 below shows that the plateau modulus  $G_0$  of the 45 mM SHNC/CTAB precursor solution remains constant ( $G_0 \sim 3.6$  Pa) from 25 °C to 30 °C under small-amplitude oscillatory shear. However, the elastic modulus G' decreases at 45 °C for frequencies ranging from 0.01–50 Hz. Nevertheless, G' exhibits a turn-up trend at higher frequencies and reaches ~3.6 Pa at 100 Hz.

We could not obtain accurate measurements of G' at frequencies higher than 100 Hz at 45 °C, due to instrument limitations at high frequencies for weakly viscoelastic solutions. Raghavan *et al.* observed that  $G_0$  of cationic surfactants with long (C<sub>22</sub>) mono-unsaturated tails was independent of temperature variations at frequencies larger than 100 Hz [2]. In fact, similar trend was observed for more concentrated SHNC/CTAB precursor solutions. Therefore, we assume here that  $G_0$  of the SHNC/CTAB precursor solution reaches a similar value above 100 Hz at 25 °C, 30 °C, and 45 °C. At 60 °C, 45 mM SHNC/CTAB precursor solution exhibits Newtonian-like behavior, which can be verified by the SANS spectra, showing the presence of short cylindrical micelles (see main manuscript).



Figure 2: Elastic modulus (G') of the precursor solution at 25 °C, 30 °C and 45 °C, measured by small-amplitude oscillatory shear procedure.

## Sample preparation for neutron scattering measurements

The precursor was prepared by volume in deuterated water containing 99.9% D<sub>2</sub>O (Cambridge Isotope Laboratories, Andover, MA). Since the conversion rate from the precursor to the FISP scaffold is not 100% from the microfluidic process, we collected the sample from the microchannel containing the FISP scaffold and liquid precursor residues. We then added 15 mL of D<sub>2</sub>O and centrifuged the diluted solution at 1500 RPM for 30 minutes. We left the centrifuged solution without agitation for 5 hours before experiments. A supernatant thin layer containing the concentrated FISP structure was formed in the stored solution at room temperature. We then loaded ~100  $\mu$ L of the FISP suspended in D<sub>2</sub>O in a demountable, 2 mm titanium cell with quartz windows (NIST). The demountable cell was filled with the dispersion and capped with a screw. The cell was gently shaken to obtain homogenous dispersion before samples were exposed to the neutron beam in the SANS instrument. Subsequently the samples were again agitated before in the cell before the USANS measurement. The scattering contrast was determined from the scattering length density (SLD) of the deuterated solvent ( $6.39 \times 10^{-6} \text{ Å}^{-2}$ ) and the precursor ( $-1.54 \times 10^{-7} \text{ Å}^{-2}$ ) by using SANSview tool bar [3].



Figure 3: FISP structure suspended in  $D_2O$  for SANS experiments at ~25 °C.

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