Electronic Supplementary Information

Effects of Shear-Induced Crystallization on the Complex Viscosity of Lamellar-Structured Concentrated Surfactant Solutions

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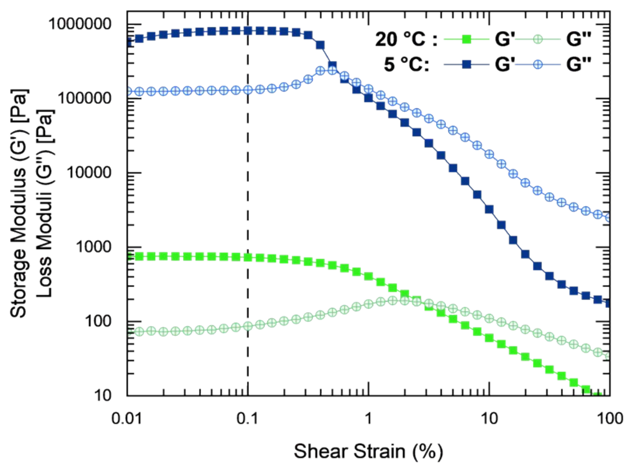

Figure S1. Amplitude sweeps at different temperatures. A strain amplitude (γ_0) = 0.1%, shown by the dotted line is within the linear viscoelastic range (LVER).

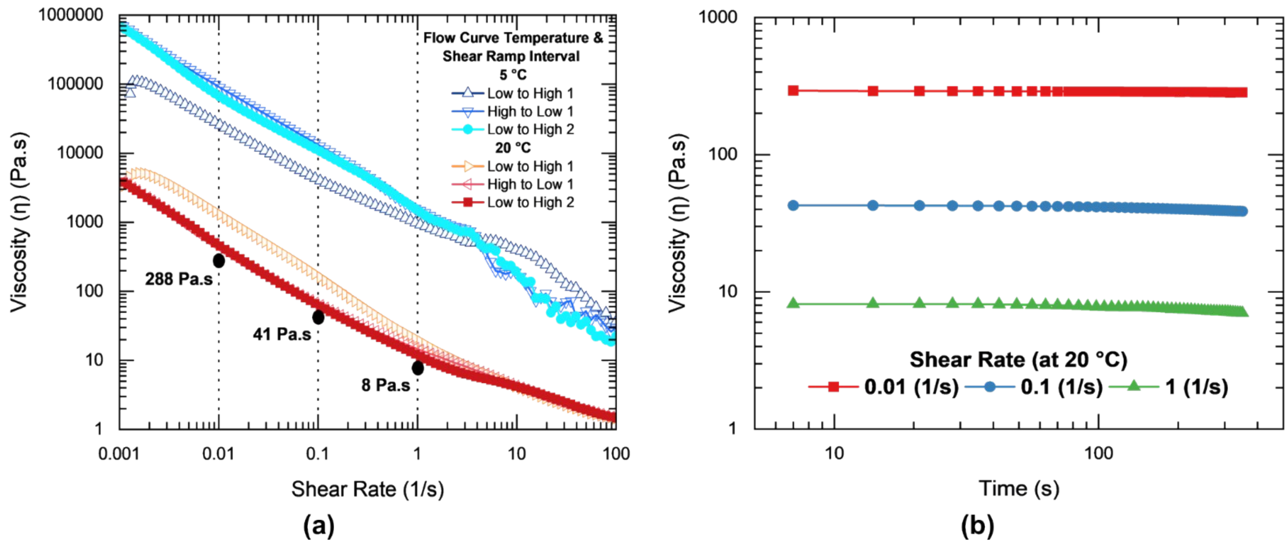

Figure S2. (a) Low-to-high and high-to-low shear rate sweeps. Data from the second low-to-high ramp is presented. (b) Time sweep experiments at constant shear rate after low-to-high and high-to-low shear rate sweep. The average of viscosities at each shear rate is shown in (a). Figures (a) and (b) reveal that the procedure used to create the continuous shear rate ramp flow curve achieves steady state conditions.

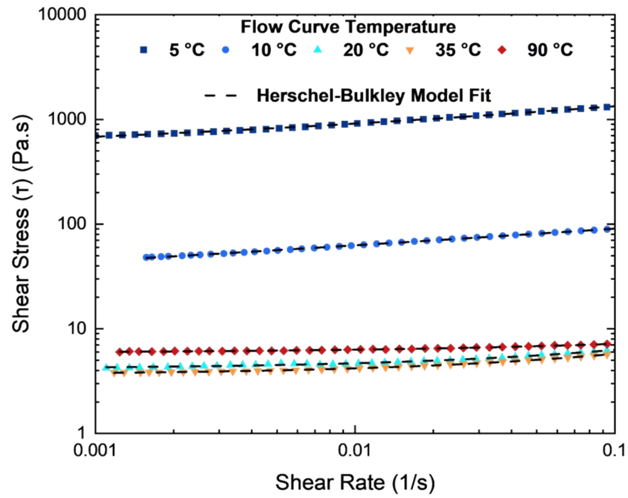

Figure S3. Herschel - Bulkley fits at low shear rates for flow curves at different temperatures. The data is presented as shear stress vs shear rate. Samples were loaded into the fixture at 20 °C, cycled to the predetermined temperature with applied oscillations, and rested for 2 min before shearing.

The Herschel - Bulkley model[1,2] is as follows: $\sigma = \sigma_y + K * (\dot{\gamma}^n)$, where σ is the shear stress, σ_y is the yield stress, K is the consistency coefficient, $\dot{\gamma}$ is the shear rate and n is the flow behavior index which varies as follows. If n=1, the Herschel - Bulkley model is equivalent to the Bingham Plastic model. If $\sigma_y = 0$, it is equivalent to the Ostwald–de Waele power law model. The yield stresses, consistency coefficients and flow index are summarized in the table below.

Herschel - Bulkley Fits:				
Temperature (°C)	**Yield Stress (σ_y) (Pa)**	**Consistency coefficient (K)**	**Flow behavior index (n)**	**R-square (COD)**
5	415	1709	0.268	0.999
10	9.80	123	0.183	0.999
20	5.86	3.83	0.462	0.996
35	4.15	8.54	0.600	0.993
90	3.65	8.26	0.592	0.999

As the temperature increases, the dynamic yield stress predicted by the Herschel – Bulkley model decreases. This is consistent with other observations in the manuscript. At 5 °C, the crystalline phase has high viscosity and high yield stress. The concentrated surfactant solution has different microstructures at 20 °C, 35 °C and 90 °C, but the flow behavior and magnitude of yield stress are similar.

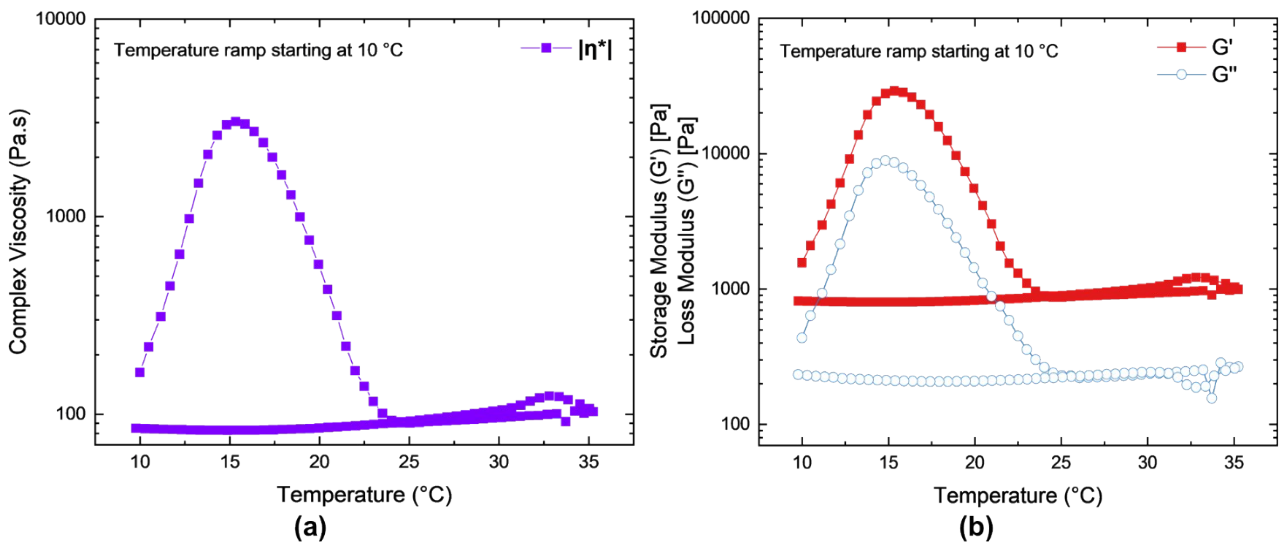

Figure S4. (a) Complex viscosity and (b) G′ and G″ profiles for temperature ramp starting at 10 °C. Samples at 20 °C were loaded into the fixture set to a pre-determined temperature, pre-sheared and rested before the temperature sweep. The samples were heated up to 35 °C and immediately cooled back to the temperature of interest.

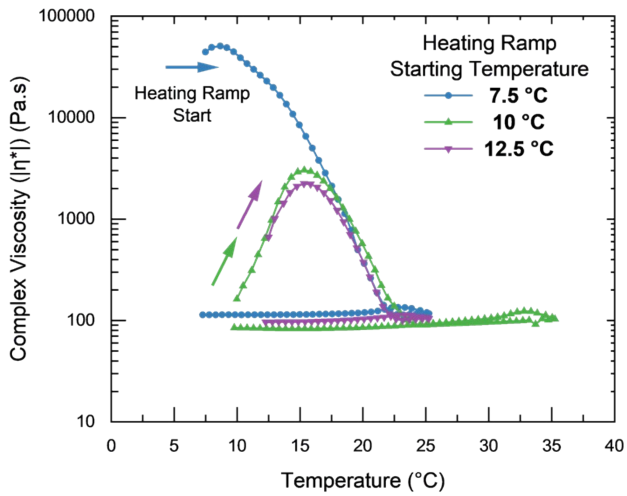

Figure S5. Effect of starting temperature. Samples at 20 °C were loaded into the fixture set to a pre-determined temperature, pre-sheared and rested before the temperature sweep. The samples were heated up to 35 °C and immediately cooled back to the temperature of interest.

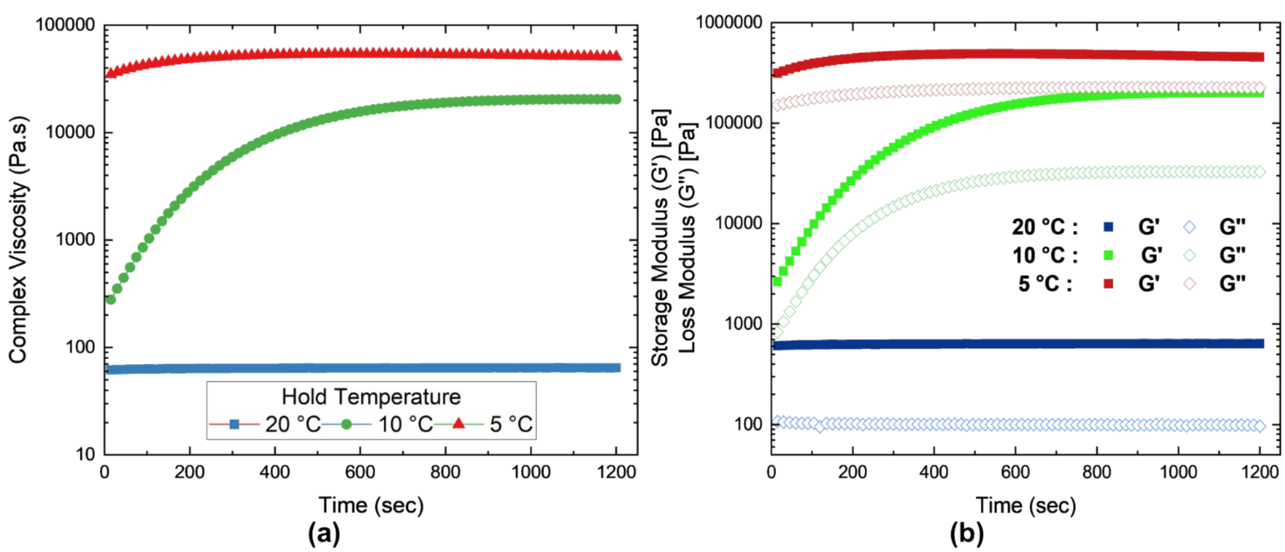

Figure S6. (a) Complex viscosity and (b) G′ and G″ profiles for thermal aging of specimens at pre-determined temperatures for 20 mins. The specimens were cooled from 20 °C to a pre-determined temperature (e.g., 5 °C) at 0.5 °C/min without oscillation, pre-sheared, and rested before the aging experiment.

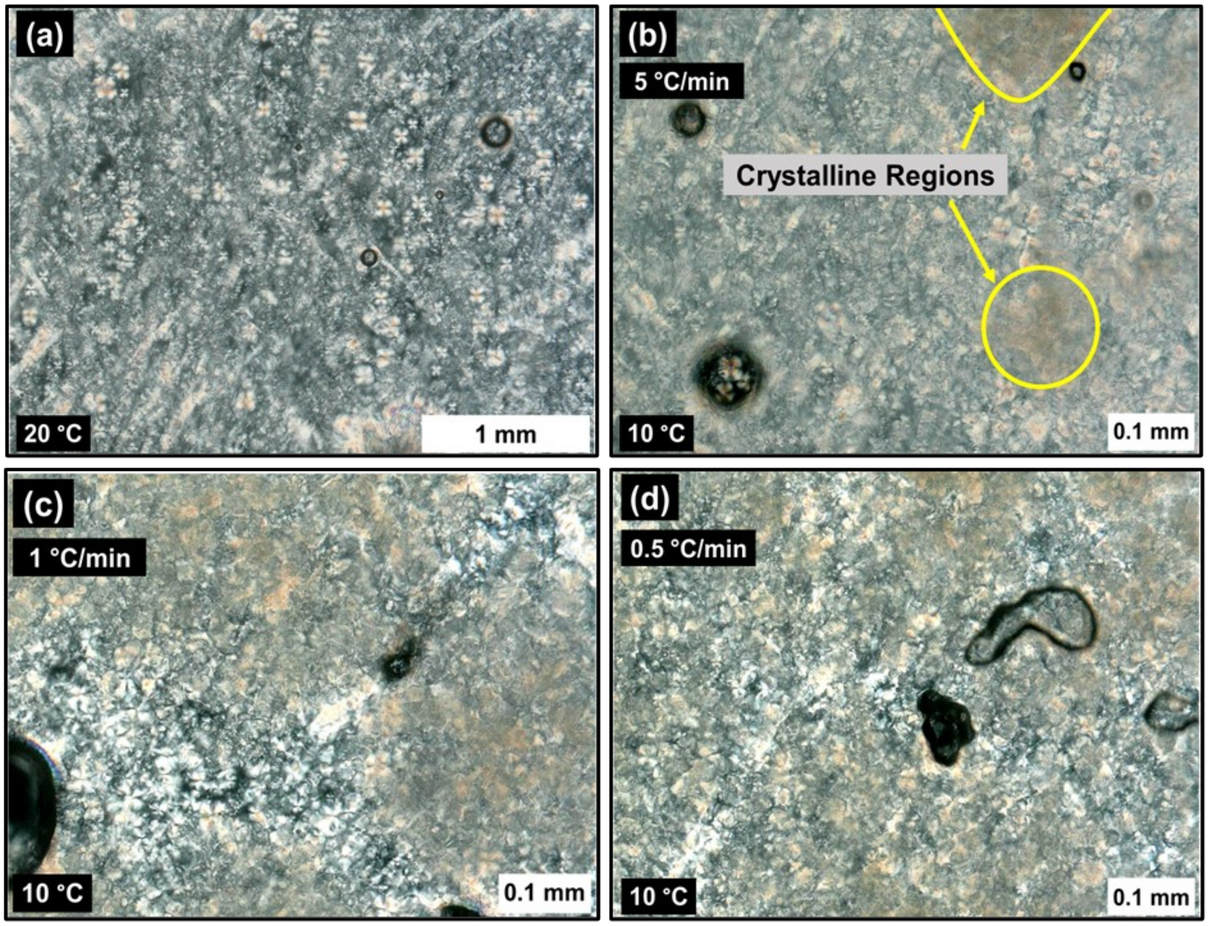

Figure S7. Cross-polarized micrographs showing the effect of pre-oscillation quiescent cooling rate on the microstructure. The specimens were cooled from 20 °C to 10 °C at different cooling rates (b) 5 °C/min, (c) 1 °C/min and (d) 0.5 °C/min.

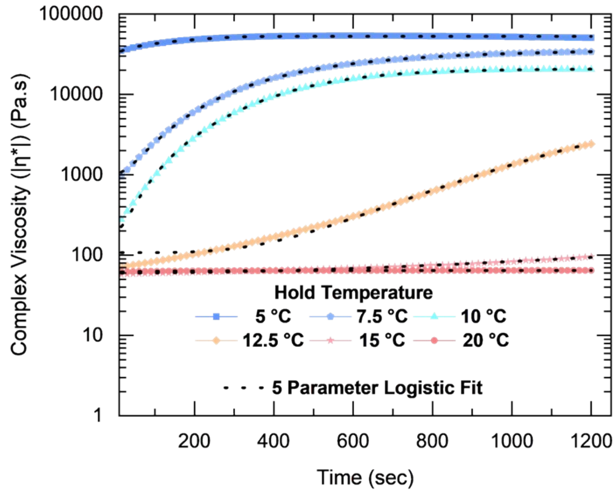

Figure S8. 5 Parameter Logistic Fits for isothermal shear-induced crystallization. The specimens were cooled from 20 °C to a pre-determined temperature (e.g., 5 °C) at 0.5 °C/min without oscillation, pre-sheared, and rested before the aging experiment – with oscillations.

The modified five-parameter logistic (5PL) model is as follows.

$$[|\eta^*|(t)]_{Temperature} = |\eta^*_0| + \frac{|\eta^*|_{max} - |\eta^*_0|}{\left(1 + \left[\frac{t}{t_{inflection}}\right]^{-h}\right)^{s}} \qquad \text{Eqn. 1}$$

where $|\eta^*_0|$ and $|\eta^*|_{max}$ respectively are the complex viscosities at time (t) = 0 and infinity, $t_{inflection}$ is the time at which curvature changes direction (time at which $|\eta^*|(t) = (|\eta^*|_{max} - |\eta^*_0|)/2)$, h is the slope of the curves before the plateau and s is the asymmetry factor (when s = 1, the curve is symmetric). The model parameters are summarized in the table below.

5 Parameter Logistic Fits

Temperature (°C)	$\lvert\eta^*_0\rvert$ (Pa.s)	$\lvert\eta^*\rvert_{max}$ (Pa.s)	$t_{inflection}$ (sec)	h	s	R-square (COD)
5	28048	52797	325	10.43	0.04	0.96
7.5	1033	37767	567	2.48	0.74	0.99
10	200	20965	606	4.51	0.40	0.99
12.5	107	2707	1223	34.34	0.1	0.99
15	60	1905	8814	7.27	0.27	0.99
20	60	65	322	1.01	0.37	0.98

The Avrami Fit Parameters are summarized in the table below:

Temperature (°C)	Intercept (K)	Slope (n)	R-square (COD)
7.5	-9.83	1.46	0.99
10	-11.77	1.68	0.99
12.5	-20.91	2.49	0.96

REFERENCES

1 W. H. Herschel and R. Bulkley, *Kolloid-Zeitschrift*, 1926, **39**, 291–300.

2 T. Divoux, C. Barentin and S. Manneville, *Soft Matter*, 2011, **7**, 8409.