

Characterizing semiflexible network structure of wormlike micelles by dynamic techniques

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S1. Determination of Overlap Concentration

The overlap concentration can be determined from the peak in the static correlation length ξ_0 obtained from the Zimm plot in the SLS measurements:^{1,2}

$$\left(\frac{Kc}{Rq}\right)_{c=0} = \frac{1}{M_w} (1 + \xi_0^2 q^2 + \dots)$$

where $K = (4\pi^2 n^2 / \lambda_0^4 N_A) (dn/dc)^2$ is an optical constant (n : refractive index, λ_0 : wavelength of the incident light, N_A : Avogadro constant), c is the mass concentration of SDS, R_q is the Rayleigh ratio, M_w is the weight average molecular weight, and q is the scattering vector. The overlap concentration is estimated to be around $C_{\text{SDS}}^* = 0.15$ wt% from Fig. S1.

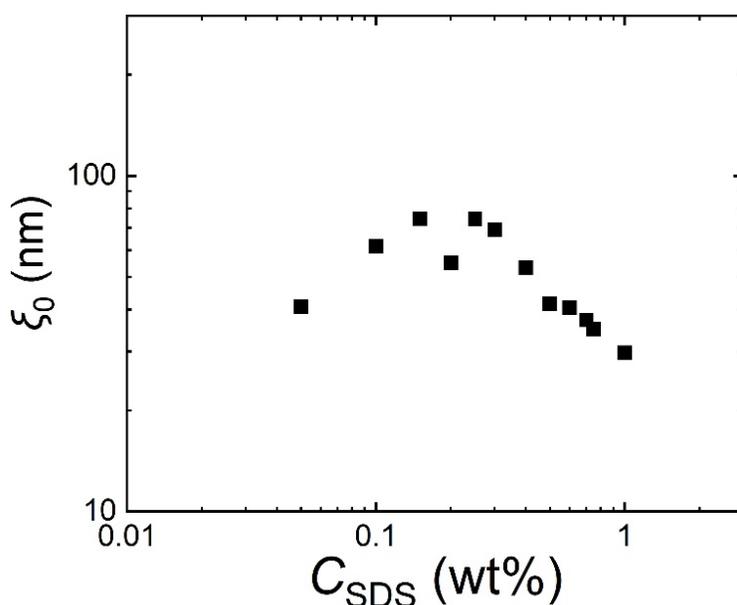


Figure S1. Static correlation length ξ_0 as a function of C_{SDS} obtained from SLS measurements.

S2. Determination of Relaxation Times in DLS

Normalized *intensity* autocorrelation functions for C_{SDS} below 1 wt% are shown in Fig. S2. At 0.1 wt%, below the overlap concentration 0.15 wt%, the function exhibits single mode of decorrelation. Above the overlap concentration, a slower mode appears and its amplitude increases with concentration. For the wormlike micelles in semidilute regime, two main decorrelation modes and one more mode can be found.² We fitted thus the normalized *field* autocorrelation functions $g^{(1)}(t)$ with a three-component stretched exponential function, which is empirically known to fit well the multimodal autocorrelation of polymer solutions and gels:³

$$g^{(1)}(t) = A_1 \exp\left\{-\left(\frac{t}{\tau_1}\right)^{\alpha_1}\right\} + A_2 \exp\left\{-\left(\frac{t}{\tau_2}\right)^{\alpha_2}\right\} + (1 - A_1 - A_2) \exp\left\{-\left(\frac{t}{\tau_3}\right)^{\alpha_3}\right\}$$

where A_i is the amplitude, τ_i is the characteristic time, α_i is the stretch exponent related to the polydispersity of the characteristic time. The mode having the smallest characteristic time is taken as fast mode, and the mode having the slowest relaxation time is taken as slow mode. Between these two larger modes, an intermediate mode with a small amplitude (A less than 0.1 at most of the concentration and angle) is found. This mode presumably corresponds to the Rouse mode,² however, experimental precisions do not allow us to further investigate the mode. As an example, the fitting curve for the *intensity* autocorrelation $g^{(2)}(t) - 1 = \{g^{(1)}(t)\}^2$ for $C_{\text{SDS}} = 6 \text{ wt}\%$ found in Fig. 1a is shown in Fig.S3.

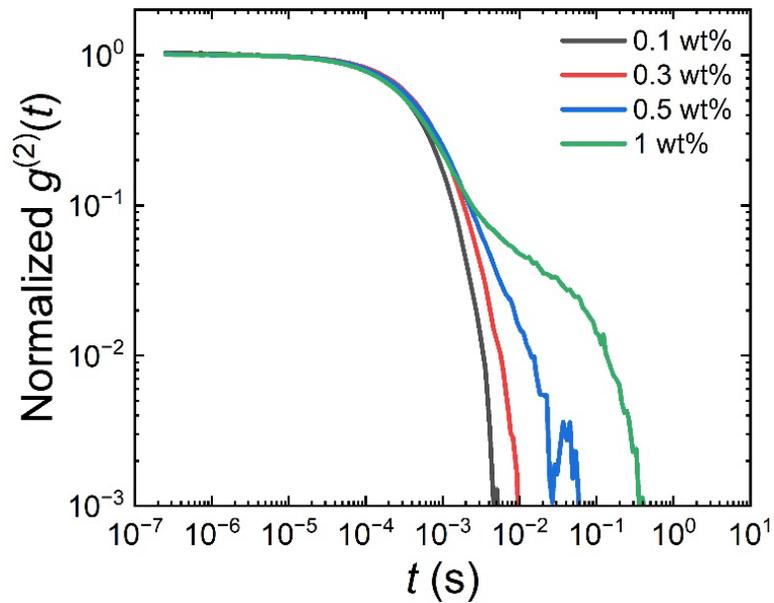


Figure S2. Normalized intensity autocorrelation function $g^{(2)}(t)$ of SDS/Al(NO₃)₃ aqueous solutions at different SDS concentrations. Scattering angle: 90°.

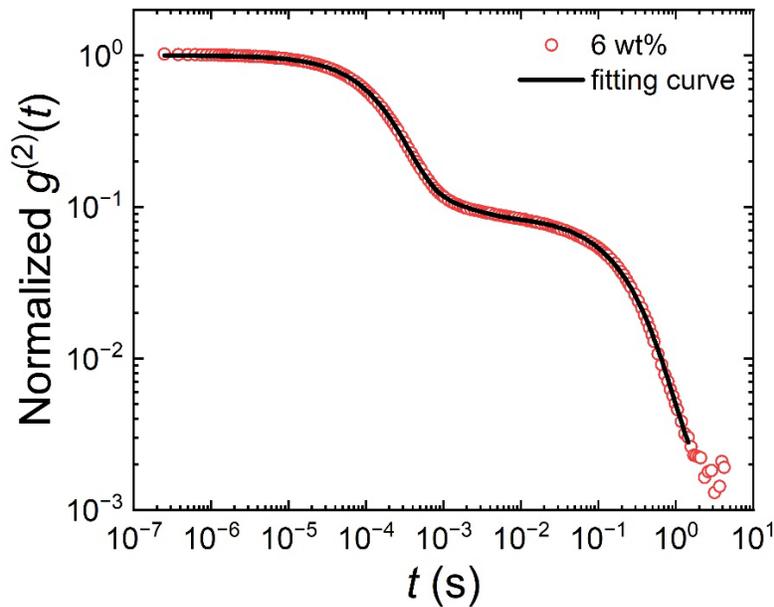


Figure S3. Normalized intensity autocorrelation function $g^{(2)}(t)$ of SDS/Al(NO₃)₃ aqueous solutions at $C_{\text{SDS}} = 6 \text{ wt}\%$. Solid curve shows the fitting curve with the multicomponent stretched exponential function. Scattering angle: 90°.

S3. Supporting Figures

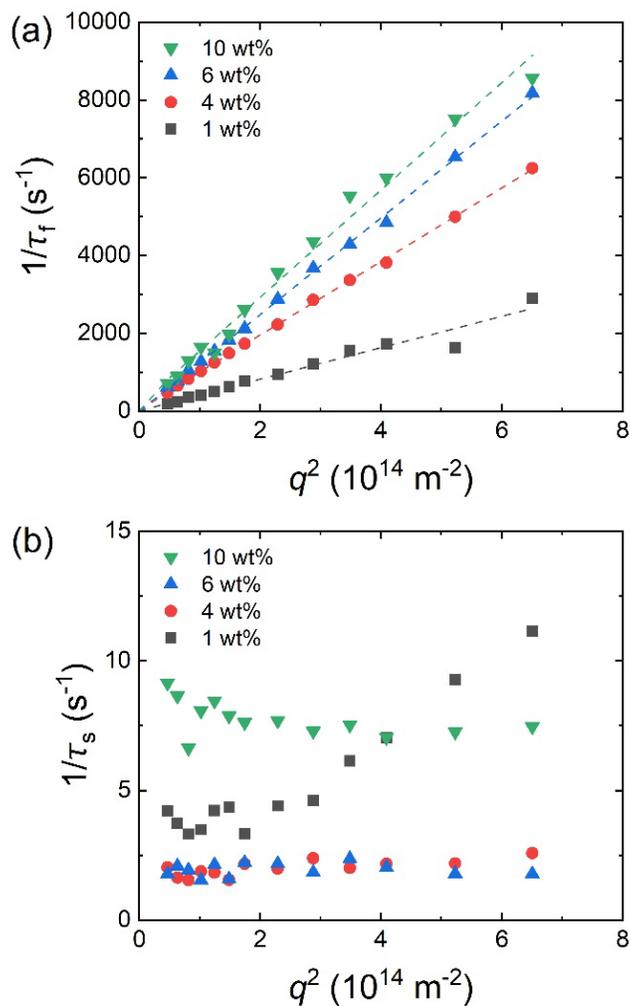


Figure S4. (a) $1/\tau_f$ and (b) $1/\tau_s$ as functions of q^2 . In (a), the slope represents the collective diffusion coefficient D .

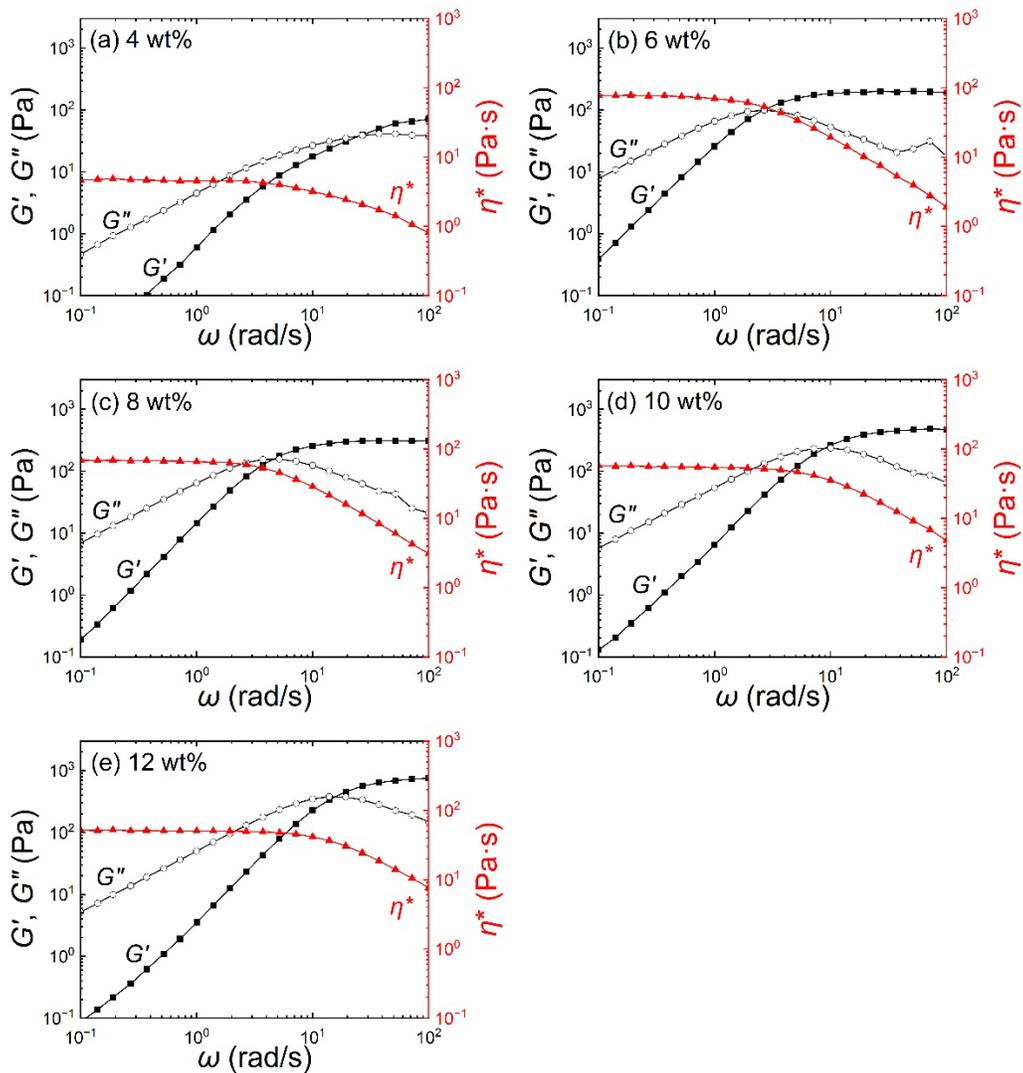


Figure S5. Viscoelastic moduli and complex viscosity obtained from oscillatory shear macrorheology.

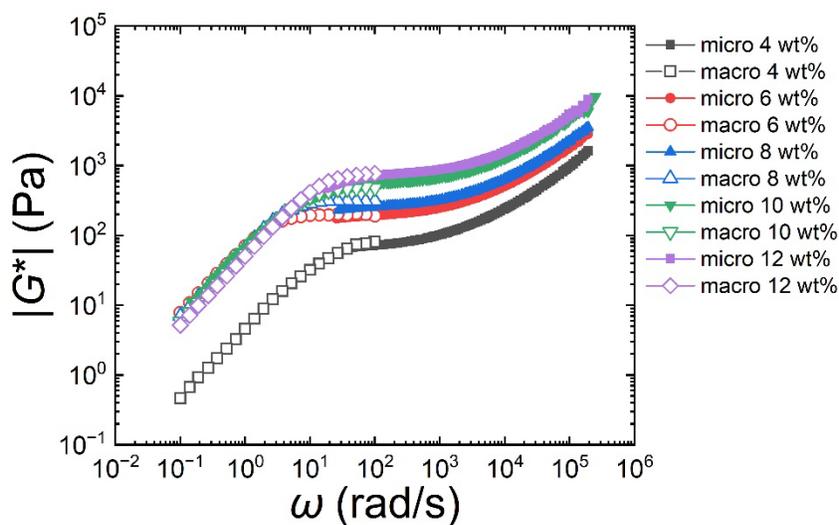


Figure S6. Polymer contribution to complex modulus obtained by DWS microrheology and macrorheology.

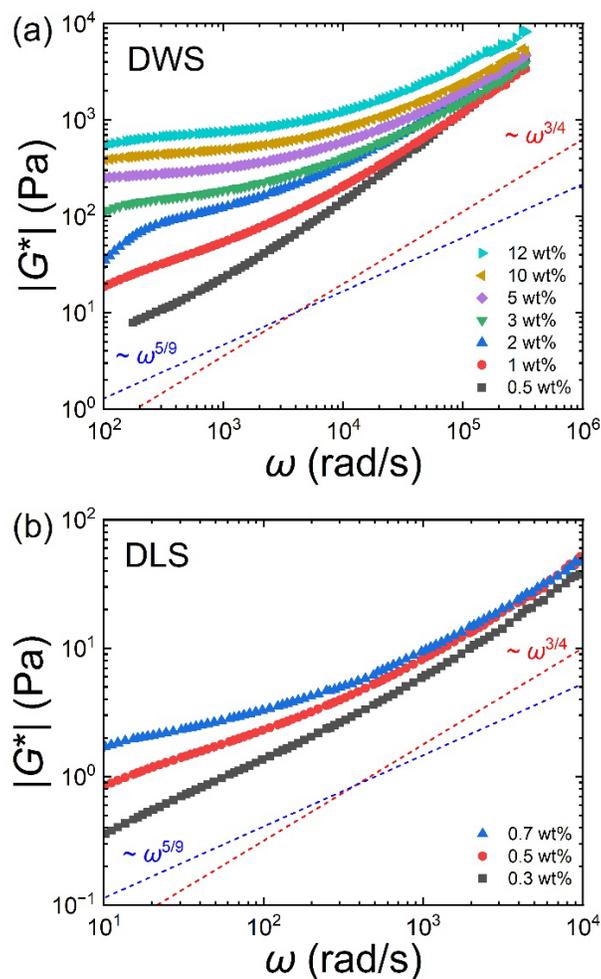


Figure S7. Polymer contribution to complex modulus obtained from (a) DWS and (b) DLS measurements. Dashed lines serve as visual guides for the Zimm mode (5/9 power law) and the bending mode (3/4 power law).

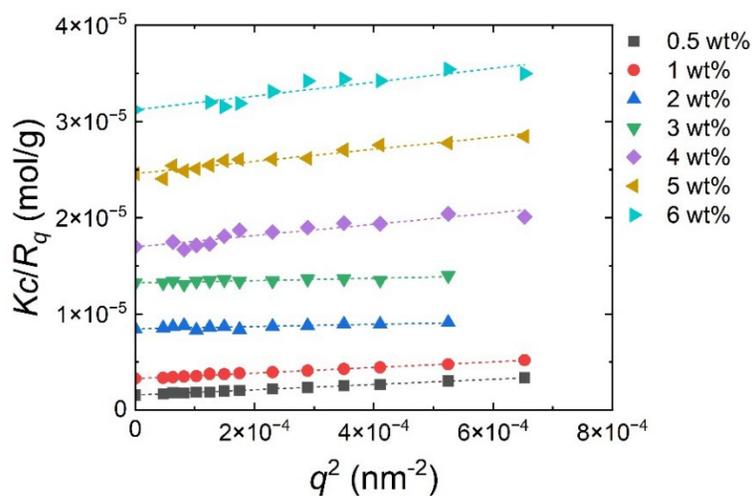


Figure S8. Zimm plots from static light scattering measurements of SDS/ $\text{Al}(\text{NO}_3)_3$ solutions.

REFERENCES

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- (2) E. Buhler, J. P. Munch and S. J. Candau, *J. Phys. II*, 1995, **5**, 765-787.
- (3) S. Rose, A. Marcellan, D. Hourdet and T. Narita, *Macromolecules*, 2013, **46**, 5329-5336.