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Gelation phase diagrams of colloidal rod systems measured over a large composition space †

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Electronic supplementary material

Bulk rheological measurements of HCO and PA



Fig. S1 Strain sweep measurement used to determine the linear viscoelastic regime for (a) HCO and (b) PA at a constant angular frequency (6.28 rad s^{-1}).

Characterization of PA colloidal rods



Fig. S2 Differential interference contrast microscopy (DIC) image of 1 wt% PA rods.



Fig. S3 Atomic force microscopy (AFM) image of 0.02 wt% PA rods.

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Microrheological measurements of HCO and PA colloidal rod systems with increasing component concentrations



Fig. S4 Measured ensemble-averaged mean-squared displacement of (a) HCO and (b) PA with increasing colloid concentration from 0-0.8 wt%. The magnitude of the MSD decreases as colloid concentration increases, while the value of $\alpha \approx 1$ for all measurements. This indicates that the diffusivity of the particles decreases due to an increase in solution viscosity.



Fig. S5 Measured ensemble-averaged mean-squared displacement of (a) HCO and (b) PA with increasing PEO concentration from $0-0.75 \ c/c^*$. Both the magnitude of the MSD and α values decrease as PEO concentration increases. This indicates that probe particles motion is being restricted as depletion interactions increase.



Fig. S6 Measured ensemble-averaged mean-squared displacement of (a) HCO and (b) PA with increasing LAS concentration from 0.8-12.8 wt%. The magnitude of the MSD decreases as LAS concentration increases, this indicates that the solution viscosity is increasing. The value of α changes non-monotonically with LAS concentration, which indicates that the microstructure of the system is changing.



Fig. S7 Microrheological measurements of HCO with increasing PEO concentration, which increases depletion interactions. The dashed line is the critical relaxation exponent, *n*, for *LAS* : *colloid* \leq 16. The shaded area is the sol-gel transition region. In all measurements the LAS concentration is held constant at 1.6 wt%. The logarithmic slope of the MSD, α , decreases and passes through the sol-gel transition region for all colloidal gel systems as depletion interactions are increased, indicating that the system transitions from a sol to a gel. No data are provided for PA since this low concentration of LAS is not achievable.



Fig. S8 Microrheological measurements of HCO with increasing PEO concentration, which increases depletion interactions. The dashed line indicates the critical relaxation exponent, *n*, for *LAS* : *colloid* \leq 16. The shaded area is the sol-gel transition region. In all measurements the LAS concentration is held constant at 2.4 wt%. The logarithmic slope of the MSD, α , decreases and passes through the sol-gel transition region for all colloidal gel systems as depletion interactions are increased, indicating that the system transitions from a sol to a gel. No data are provided for PA since this low concentration of LAS is not achievable.



Fig. S9 Microrheological measurements of (a) HCO and (b) PA with increasing PEO concentration, which increases depletion interactions. The dashed line indicates the critical relaxation exponent, *n*, for *LAS* : *colloid* \leq 16. The shaded area is the sol-gel transition region. In all measurements the LAS concentration is held constant at 3.2 wt%. The logarithmic slope of the MSD, α , decreases and passes through the sol-gel transition region for all colloidal gel systems as depletion interactions are increased, indicating that the system transitions from a sol to a gel.



Fig. S10 Microrheological measurements of (a) HCO and (b) PA with increasing PEO concentration, which increases depletion interactions. The dashed line indicates the critical relaxation exponent, *n*, for *LAS* : *colloid* \leq 16. The shaded area is the sol-gel transition region. In all measurements the LAS concentration is held constant at 6.4 wt%. The logarithmic slope of the MSD, α , decreases and passes through the sol-gel transition region for all colloidal gel systems as depletion interactions are increased, indicating that the system transitions from a sol to a gel.



Fig. S11 Microrheological measurements of (a) HCO and (b) PA with increasing PEO concentration, which increases depletion interactions. The dashed lines indicate the critical relaxation exponent, *n*, for either *LAS* : *colloid* \leq 16 or *LAS* : *colloid* > 16. The shaded area is the sol-gel transition region. In all measurements the LAS concentration is held constant at 9.6 wt%. The logarithmic slope of the MSD, α , decreases and passes through the sol-gel transition region for all colloidal gel systems as depletion interactions are increased, indicating that the system transitions from a sol to a gel.



Fig. S12 MPT measurements of (a) HCO and (b) PA with increasing colloid concentration at 0.14 c/c* PEO and 12.8 wt% LAS. The logarithmic slope of the MSD, α , is constant with increasing colloid concentration from 0-0.8 wt%, indicating that the system remains in the sol phase ($\alpha \approx 1$).



Fig. S13 MPT measurements of (a) HCO and (b) PA with increasing PEO concentration at 0.2 wt% colloid and 12.8 wt% LAS. The dashed line indicates the value of *n* for *LAS* : *colloid* > 16. The shaded area is the sol-gel transition region. The logarithmic slope of the MSD, α , decreases and passes through the sol-gel transition region as depletion interactions are increased, indicating that the system transitions from a sol to a gel.



Fig. S14 MPT measurements of (a) HCO and (b) PA with increasing LAS concentration at 0.2 wt% colloid and 0.52 c/c^* PEO. For both systems, α increases as LAS concentration increases. Because the *LAS* : *colloid* ratio is not fixed, the sol-gel transition line is not plotted.



Fig. S15 Bulk rheology measurements of (a) HCO and (b) PA with increasing colloid concentration at 0.14 c/c^* PEO and 12.8 wt% LAS. The storage modulus increases as colloid concentration increases.



Fig. S16 Bulk rheology measurements of (a) HCO and (b) PA with increasing PEO concentration at 0.2 wt% colloid and 12.8 wt% LAS. The storage modulus remains constant as PEO concentration increases.



Fig. S17 Bulk rheology measurements of (a) HCO and (b) PA with increasing LAS concentration at 0.2 wt% colloid and 0.52 c/c^* . The storage modulus remains constant as LAS concentration increases.