Supporting Information

1. Synthesis of core-shell particles

Three samples with different NIPAM/PS ratios (PS-N 1, PS-N 2 and PS-N 3) were synthesized. A detailed recipe is shown in table 1s.

Table 1s. Synthesis of the core-shell particles (the hydrodynamic radius of the PS core is 52nm)

Sample name	PS latex	Water	NIPAM	BIS	KPS	R_h(25°C)
PS-N 1	24.00g	200mL	1.00g	0.07g	0.02g	61nm
PS-N 2	24.00g	200mL	2.00g	0.14g	0.04g	94nm
PS-N 3	24.00g	200mL	4.00g	0.28g	0.08g	104nm

2. Oscillatory frequency sweep for PS-N 1 at different temperatures

We studied the storage and loss modulus for PS-N 1 sample at the same weight fraction 23wt% at temperatures ranging from 25°C to32°C. In Figure 1s, both the storage and loss modulus decrease dramatically due to the shrinking of the PNIPAM shell as temperature increases from 25°C to 30°C. However, when it reaches 32°C, the modulus increases abruptly. This occurrence can be attributed the overall interaction between particles becoming attractive, which causes the particles to form a gel. Because the modulus decreases dramatically at 28°C and 30°C, we can conclude that

no gel has formed and the overall interaction is still repulsive at these temperatures.

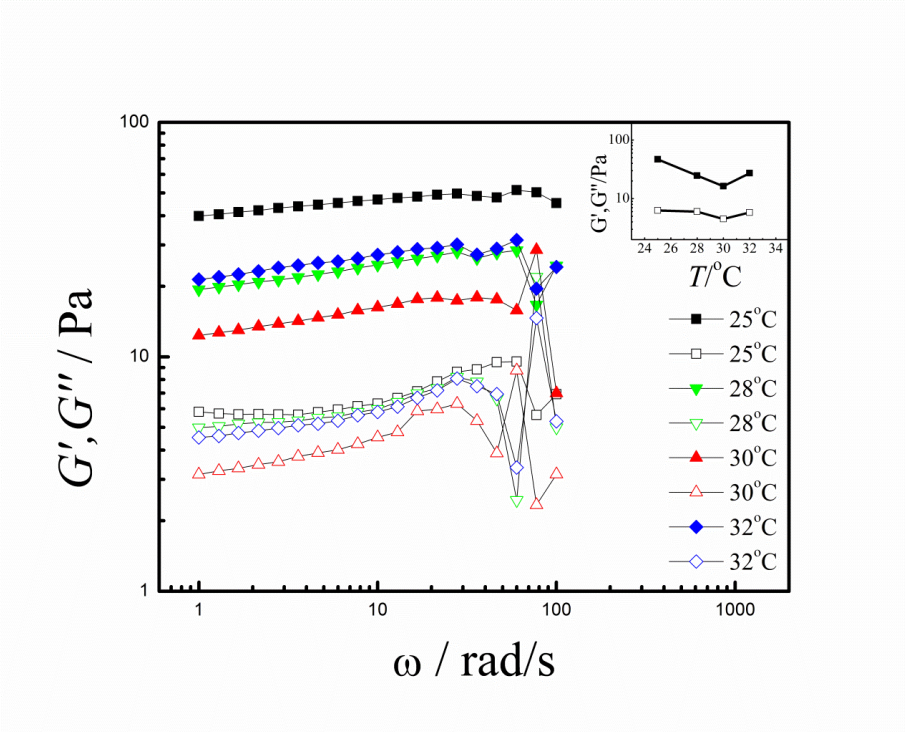

Figure 1s.Frequency dependence of the storage modulus G'(filled symbols) and loss modulus G" (open symbols) of PS-N 1 suspensions at different temperatures. The weight fraction is 23 wt%, and the amplitude is 1Pa. The inset is the storage modulus G' (filled square) and loss modulus G'' (open square) at 10 rad/s for different temperatures.

3. Particle anisotropy of microgel suspensions under shear

Rheo-SANS experiments of core-shell particles at 2100 s^{-1} shows that the form factor of particles under shear is no longer symmetric; anisotropy appeared at high shear rates. Sector averaged scattering intensity I(q) was obtained separately along the horizontal and vertical direction, respectively, as shown in Figure 2s. The second peak, which is attributed to the form factor of the particle (N. Dingenouts et al., *Phys. Chem.*

Chem. Phys., 2001, 3, 1169-1174), in the vertical direction was at higher q than that in the horizontal direction, which indicated the soft PNIPAM shell is elongated along the shear direction. Further SANS experiments and modeling are done to obtain a better understanding of the structure of hydroclusters and their interactions.

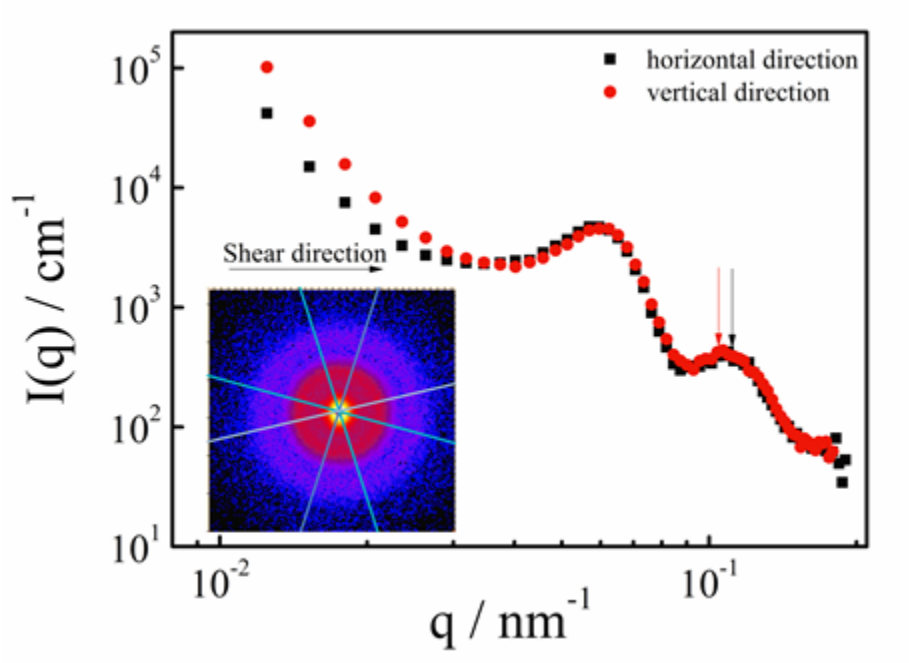

Figure 2s. Plot of the sector averaged scattering intensity along the horizontal and vertical direction measured in the radial direction against the magnitude of scattering vector at 2100 s^{-1}. The inset is the corresponding scattering pattern.