Supporting Information for:

"Aqueous solution behavior of stimulus-responsive poly(methacrylic acid)-poly(2-hydroxypropyl methacrylate) diblock copolymer nanoparticles"

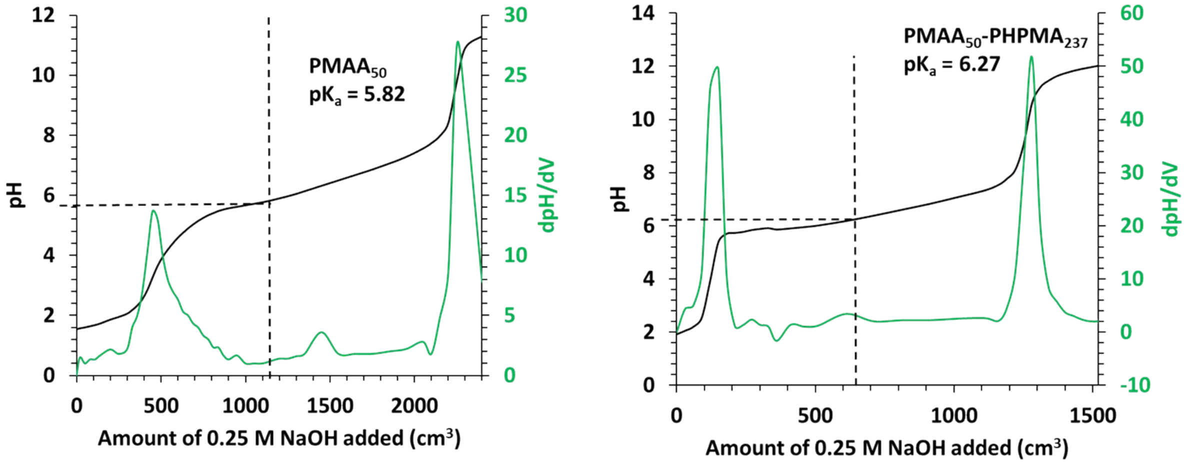

Figure S1. Potentiometric acid titration studies on the $PMAA_{50}$ precursor and the $PMAA_{50}$-$PHPMA_{237}$ diblock copolymer to determine their pK_a values.

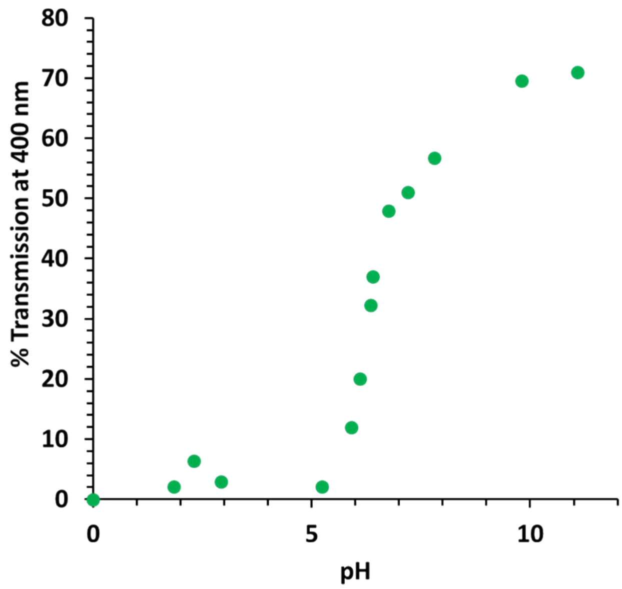

Figure S2. Turbidimetry studies using visible absorption spectroscopy to measure the light transmittance at 400 nm. The large increase in turbidity at approximately pH 6 indicates the onset of macroscopic precipitation.

Calculation of the Mean Aggregation Number (N_{agg}) for the $PMAA_{50}$-$PHPMA_{237}$ nanoparticles

Equation S1. The volume (*V*) of the core-forming PHPMA block was calculated using the equation:

$$V_{PHPMA} = \frac{DP.M_{(HPMA)}}{N_A.\rho}$$

Where *DP* is the mean degree of polymerization of the PHPMA block determined by UV spectroscopy, *M* is the molecular weight of the monomer (HPMA), N_a is Avogadro's constant, and ρ is the density of the PHPMA block.

Equation S2. The mean aggregation number (N_{agg}) is calculated using the volume of the core-forming block from **Equation S1**, as follows.

$$N_{agg} = \frac{\frac{4}{3}.\pi.r^3}{V_{PHPMA}}$$

Here *r* is the mean radius of the $PMAA_{50}$-$PHPMA_{237}$ spheres as determined from TEM studies.

Equation S3. The following equation was used to estimate the mean aggregation number for the worm-like nanoparticles, using the volume of the core-forming PHPMA block from **Equation S1.**

$$N_{agg} = \frac{\pi.r^2.L}{V_{PHPMA}}$$

Here *r* is the mean core radius (or half of the mean worm width) and *L* is the mean worm length.

Shear-Induced Polarized Light Imaging (SIPLI) studies of $PMAA_{50}$-$PHPMA_{237}$

The instrument design and general experimental set-up has been previously reported by Mykhaylyk and co-workers.[1] SIPLI experiments were conducted on a 20% w/w aqueous dispersion of $PMAA_{50}$-$PHPMA_{237}$ nano-objects at an applied shear rate of 250 s^{-1} during temperature ramp experiments conducted at a heating/cooling rate of 1.0 °C min^{-1}.

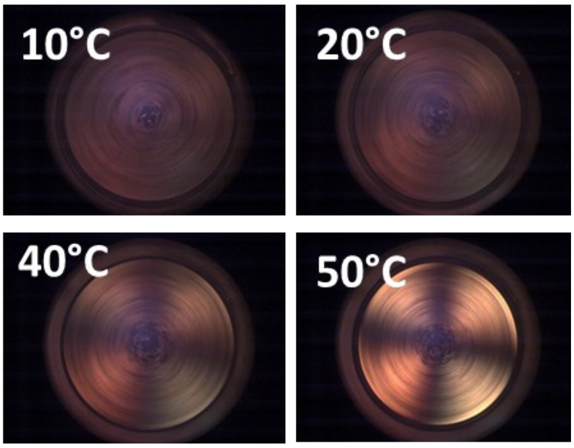

Figure S3. SIPLI images obtained at 10 °C, 20 °C, 40 °C and 50 °C during a temperature ramp experiment (heating cycle). The featureless images recorded at 10 °C and 20 °C are consistent with the presence of isotropic spheres and/or dissolved copolymer chains. A characteristic Maltese cross is formed at 50 °C, owing to the birefringence caused by the alignment of anisotropic worm-like nanoparticles in the direction of shear flow.

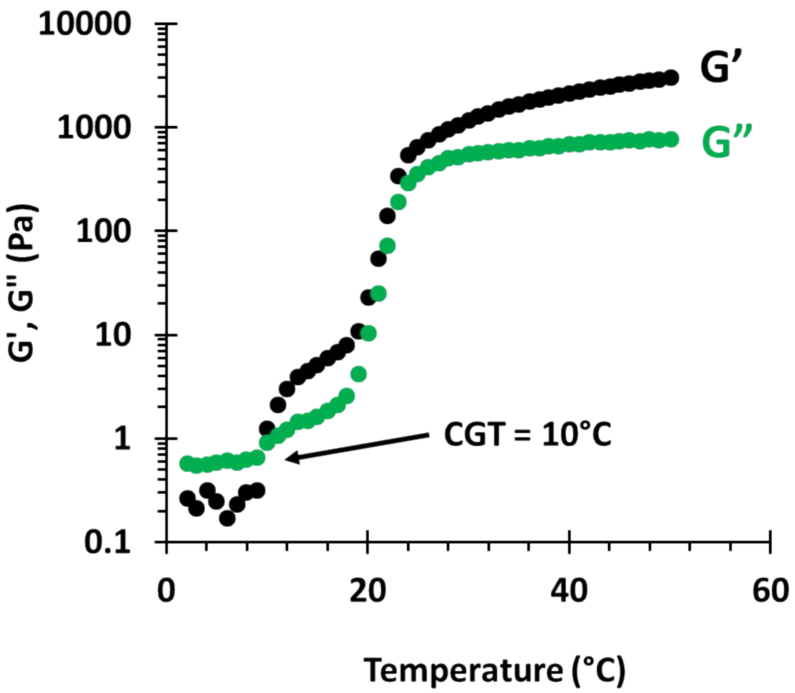

Figure S4. Temperature-dependent oscillatory rheology studies obtained on heating a 20% w/w aqueous copolymer dispersion of $PMAA_{50}$-$PHPMA_{237}$ nanoparticles from 2 °C to 50 °C. Measurements were conducted at an angular frequency of 1.0 rad s^{-1} and an applied strain of 1.0%, with an equilibration time of 5 min being allowed at each temperature. A critical gelation temperature was observed at around 10 °C, as judged by the cross-over point for the G’ and G” curves.

(1) Mykhaylyk, O. O.; Warren, N. J.; Parnell, A. J.; Pfeifer, G.; Laeuger, J. Applications of Shear-Induced Polarized Light Imaging (SIPLI) Technique for Mechano-Optical Rheology of Polymers and Soft Matter Materials. *J. Polym. Sci. Part B Polym. Phys.* **2016**, *54* (21), 2151–2170.