Supporting information for

Highly deformable hydrogels constructed by pH-triggered polyacid nanoparticle disassembly in aqueous dispersions

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Scheme S1. Depiction of method used to prepare GMA-functionalised nanoparticles from precursor nanoparticles.



Figure S1. Potentiometric titration data for two vinyl-functionalised nanoparticle (NP) dispersions (PMAA-EA-GMA and PMAA-MMA-GMA) and a PEA-MAA-GMA microgel (MG) dispersion. The MG is used as a control in this study.



Figure S2. Larger area TEM images obtained for PMAA-EA-GMA NPs ((a) and (d)), PMAA-MMA-GMA NPs ((b) and (e)) and PEA-MAA-GMA MG ((c) and (f)) deposited from aqueous pH 4.0 (top row) and pH 7.4 (bottom row) solutions. The arrows highlight some incompletely disassembled NPs. Scale bars: 200 nm.



Figure S3. Variation of z-average diameter (d_z) and PDI with pH for (a) PMAA-EA-GMA NPs, (b) PMAA-MMA-GMA NPs and (c) PEA-MAA-GMA MGs.



Figure S4. Variation of electrophoretic mobility with pH for PMAA-MMA-GMA NPs.



Figure S5. Images of concentrated NP dispersions that were used to prepare the three gels: (a) Gel-MMA-5.8, (b) Gel-MMA-7.4 and (c) Gel-MMA-10. The pH values used were (a) 5.8, (b) 7.4 and (c) 10.0, respectively. The scale bars are 10 mm.



Figure S6. (a) Volume-swelling ratio variation with buffer pH for the gels shown in (b). The lines in (a) are guides for the eye. (b) Images of the gels after reaching equilibrium swelling in buffers with various pH values (shown). The scale bar is 10 mm.



Figure S7. Cyclic compressive stress-strain data obtained for (a) Gel-MMA-5.8 and (b) Gel-MMA-10.0. The hysteresis is the area between the loading and unloading curves. This accounted for 13 and 17% of the respective loading curves in (a) and (b).



Figure S8. Lower magnification TEM image obtained for PMAA-MMA-GMA NPs deposited from a solution with a pH value of 10. The red arrows highlight NPs that had not fully disassembled - which were infrequent. The scale bar represents 200 nm.

Video

Tensile with knot (Knotting video_fn.mp4).