Supporting Information

SI Figures

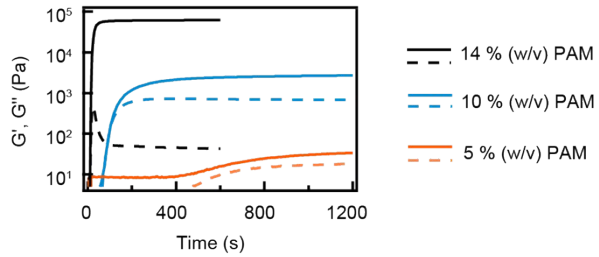

Fig. S1. Polymerization of PAM. Polymerization monitored by measuring G' (lines) and G'' (dashed lines) at $f = 1\ Hz$ and $\gamma = 0.1$ for different PAM concentrations. The asymptotic value G'_{max} of G' is approximately reached at $t = 20$ min.

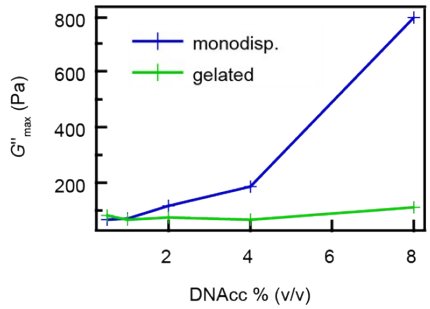

Fig. S2. Loss modulus of hybrid gels at high PAM concentrations. The loss modulus increases significantly with higher DNAcc concentrations for monodisperse colloids but stays approximately constant for the gelated colloidal system. This indicates that the monodisperse colloids lead to additional energy dissipation in the system.

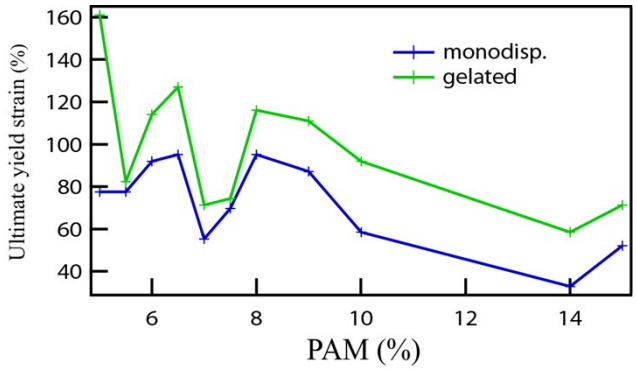

Fig. S3. Ultimate yield strain for gelated and monodispersed (4% (v/v)) at different PAM concentrations. As the second derivative of the stress strain responses relies on the smoothness of the curves, we also determined the ultimate strain, where the first derivative is zero – which can be achieved with much less numeric error. The gelated colloids have consistently higher values for all PAM concentrations and can preserve more mechanical stability.

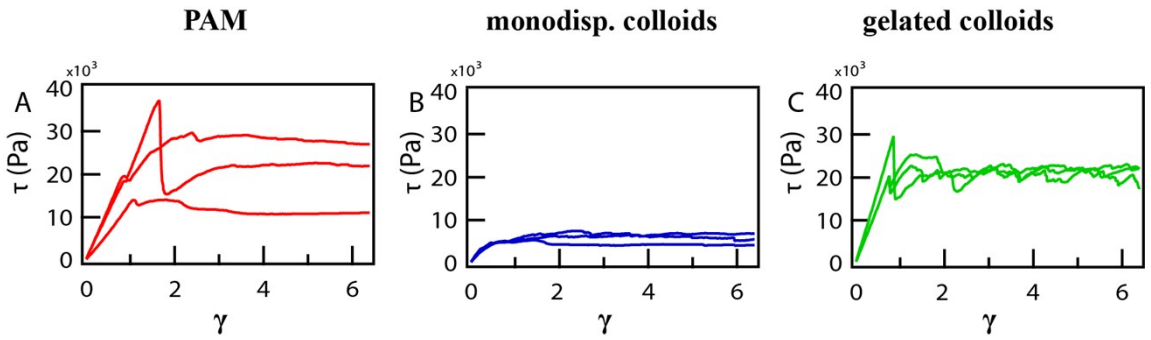

Fig. S4. Strain stress measurements for PAM, monodisperse and gelated colloids. a,b and c, The yield strain was determined for pure PAM (10%), monodispersed and gelated colloids (embedded in PAM 10%) by analyzing stress strain curves. The yield strain is defined as the first turning point in the strain stress curve, equivalent to the second derivative to be zero. The yield strain of the PAM $(\gamma = 84.0 \pm 11.2)$ reaches the highest value. Gelated colloids $(\gamma = 77.1 \pm 6.0)$ are more than 6 times larger than the monodispersed $(\gamma = 12.1 \pm 1.6)$.

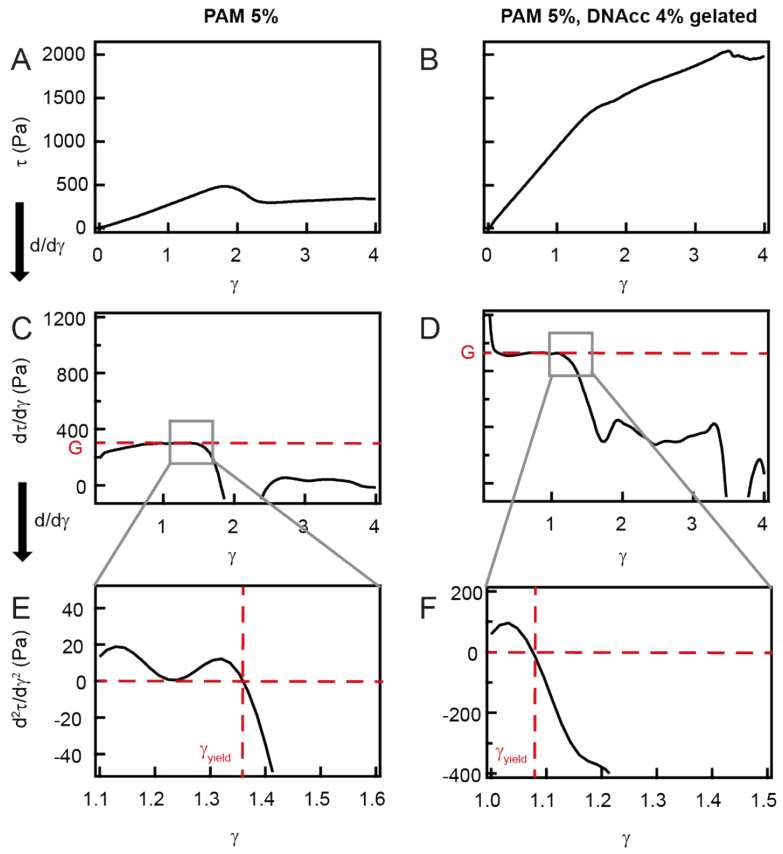

Fig. S5. Analysis of stress-strain curves. a and b, stress-strain curve of 5 % (w/v) PAM without DNAcc and with gelated DNAcc. **c and d,** derivatives of a and b that were used to derive the shear modulus G in the linear regime. **e and f,** derivatives of c and d. At the inflection point of the stress-strain curves, the second derivative of τ crosses the zero-line, yielding the yield strain γ_{yield}. Curves were smoothed with a binomial IGOR PRO algorithm before derivation to minimize noise artefacts.

DNA Sequences

The streptavidin coated beads were functionalized with biotinylated DNA strands *Dock A* and *Dock B*. The $\bar{A}B$ Linker is complementary to the docking strands and used to enable the aggregation. Following DNA sequences were used (5'→3'):

Dock A:

CAC CCA CCC ACA CCA ACC AAC TTT TTT TTT TTT TTT TTT TTT TTT TTT TTT TTT TTT TTT T – Biotin

Dock B:

Biotin - TT TTT TTT TTT TTT TTT TTT TTT TTT TTT TTT TTT TTT TTA TCT AAT ACA TTA C

$\bar{A}B$ Linker:

GTT GGT TGG TGT GGG TGG GTG TTT GTA ATG TAT TAG AT