SUPPLEMENTARY INFORMATION

Dual pH-triggered physical gels prepared from mixed dispersions of oppositely charged pH-responsive microgels

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Derivation for equation (4)

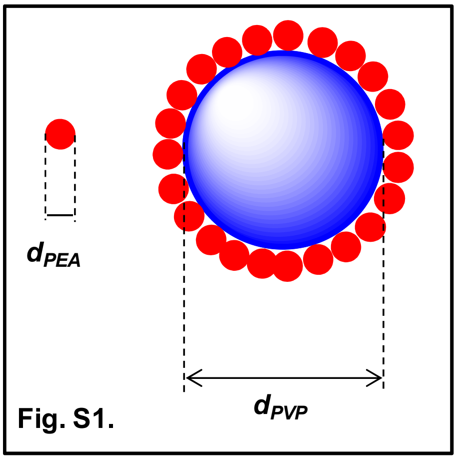

Fig. S1.

Consider PVP particles coated with a monolayer of PEAMAA (abbreviated to PEA) particles. The total surface area of the PVP particles is $A_{PVP(Tot)}$. The total cross-sectional area of the PEA particles is $A_{X,PEA(Tot)}$. The nominal fractional coverage is given by:

$$\theta_{nom} = \frac{A_{X,PEA(Tot)}}{A_{PVP(Tot)}} \qquad \text{(S1)}$$

For simplicity we have assumed that the maximum packing fraction is unity. The following equations apply for PEAMAA

$$A_{X,PEA(Tot)} = n_{PEA} A_{X,PEA(P)} \qquad \text{(S2)}$$

$$n_{PEA} = \frac{m_{PEA(Tot)}}{m_{PEA(P)}} \qquad \text{(S3)}$$

$$m_{PEA(P)} = \pi \rho_{PEA} \frac{{d_{PEA(hs)}}^{3}}{6} \qquad \text{(S4)}$$

For the above equations: n_{PEA}, $m_{PEA(Tot)}$, $m_{PEA(P)}$, ρ_{PEA} are number of PEA particles, total mass of PEA, mass of a hard sphere PEA particle and density of PEA, respectively. $A_{X,PEA(P)}$ is the cross-sectional area of a PEA particle. Equivalent equations can be derived for the PVP particles; however, in that case the surface area of a PVP particle ($A_{PVP(P)}$) is used to determine $A_{PVP(Tot)}$. It follows from equations (S1) to (S4) that:

$$\theta_{nom} = \left(\frac{V_{PVP(P,hs)}\rho_{PVP}}{V_{PEA(P,hs)}\rho_{PEA}}\right)\left(\frac{m_{PEA}}{m_{PVP}}\right)\left(\frac{A_{X,PEA(P)}}{A_{PVP(P)}}\right) \qquad \text{(S5)}$$

$V_{PVP(P,hs)}$ is the volume of a hard sphere PVP particle. Using standard geometric equations for $A_{X,PEA(P)}$ and $A_{PVP(P)}$), $R_{hs} = (d_{PEA(hs)} / d_{PVP(hs)})$ and equation (1), we obtain:

$$\theta_{nom} = \frac{1}{4}\left(\frac{Q_{PEAMAA}}{Q_{PVP}}\right)^{2/3}\left(\frac{1}{R_{hs}}\right) \qquad \text{(S6)}$$

This derivation assumed that $m_{PEA} = m_{PVP}$ and $\rho_{PEA} = \rho_{PVP}$.

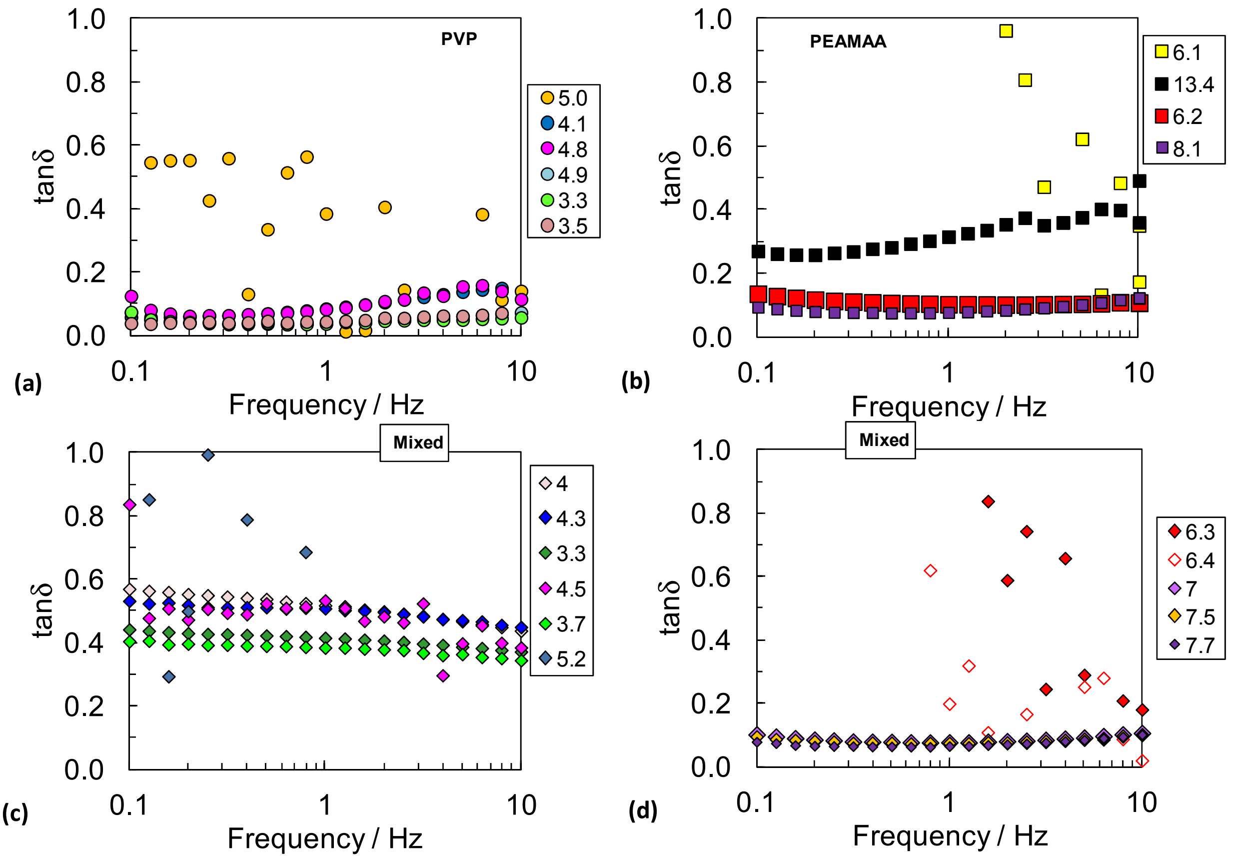

Fig. S2 Frequency sweep rheology data for mixed and heterodispersions Loss factor data for (a) PVP and (b) PEAMAA homodispersions. (c) and (d) show data for PEAMAA / PVP heterodispersions. The legends show the pH values used.

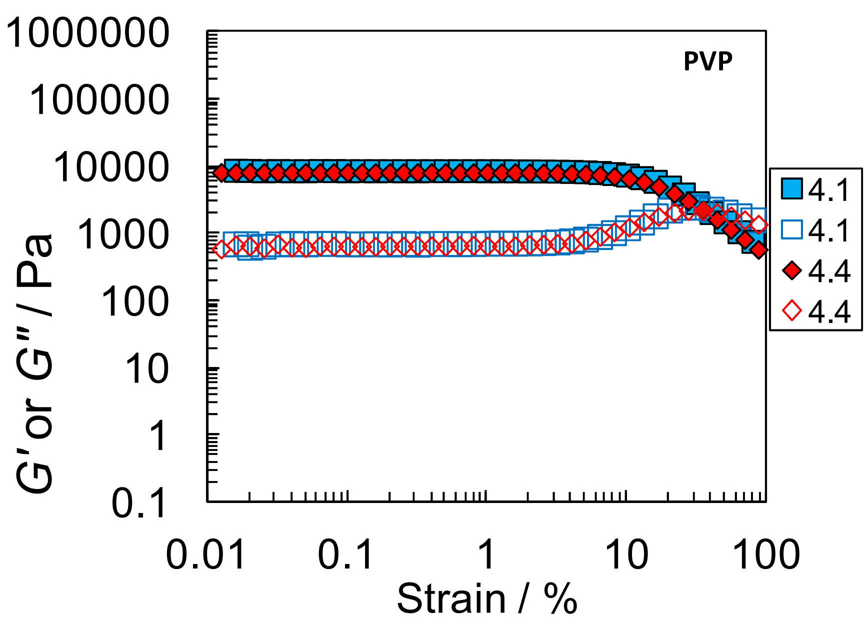

Fig. S3. The variation of *G'* (closed symbols) and *G"* (open symbols) with strain for PVP homodispersions at pH of 4.1 and 4.4.