Supplementary Information (ESI) for Soft Matter

Probing the stability of sterically-stabilized polystyrene particles by centrifugal sedimentation

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S1. Determination of Compressive Yield Stress at Initial Conditions, $P_y(ø_0)$

The initial compressive yield stress, $P_y(ø_0)$, can be determined according to the approach of Buscall and White[1,2], as proposed in Equation S1.

$$\left.\frac{dH}{dt}\right|_0 = -\frac{(1-\emptyset_0)u_0}{r(\emptyset_0)}\left(1-\frac{P_y(\emptyset_0)}{\Delta\rho g\emptyset_0 H_0}\right) \tag{S1}$$

Where $\left.\frac{dH}{dt}\right|_0$ is the initial rate of change of sediment height, $ø_0$ is the initial (uniform) volume fraction of particle dispersion, $r(ø_0)$ is the volume fraction dependent hindered settling factor, which is governed by $r(ø) \to 1$ as $ø \to 0$ and $r(ø) \to \infty$ as $ø \to 1$, u_0 is the sedimentation rate of an isolated particle, $P_y(ø_0)$ is the compressive yield stress when volume fraction of particle dispersion is $ø_0$, $\Delta\rho$ is the density difference between particle and liquid, g is the centrifugal acceleration at the bottom of the bed and H_0 is the initial sediment height.

For an initial particle volume fraction of 0.05 (in our experiments), the influence of $(1 - ø_0)/r(ø_0)$ is approximated to be of order 1. For simplification, $P_y(ø_0)$ is represented as P_y subsequently. Therefore,

$$\left.\frac{dH}{dt}\right|_0 = -u_0 + \frac{u_0 P_y}{\Delta\rho g\emptyset_0 H_0}$$

Rearranging into a straight line equation ($y = mx + c$) by multiplication with $-\frac{1}{g}$ and plotting $\left(-\frac{1}{g}\left.\frac{dH}{dt}\right|_0\right)$ against $(\Delta\rho g\emptyset_0 H_0)^{-1}$,

$$\left.\frac{dH}{dt}\right|_0\left(-\frac{1}{g}\right) = \frac{u_0}{g} - \frac{u_0 P_y}{\Delta\rho g\emptyset_0 H_0}\left(\frac{1}{g}\right)$$

$$\left.\frac{dH}{dt}\right|_0\left(-\frac{1}{g}\right) = \left(-\frac{P_y u_0}{g}\right)\frac{1}{\Delta\rho g\emptyset_0 H_0} + \frac{u_0}{g} \tag{S2}$$

Hence, Equation S2 can be plotted as a straight line of $\left(-\frac{1}{g}\left.\frac{dH}{dt}\right|_0\right)$ against $(\Delta\rho g\emptyset_0 H_0)^{-1}$, with y-axis intercept $\frac{u_0}{g}$ (denoted as I) and slope $-\frac{P_y u_0}{g}$ (denoted as $-IP_y$). As a result, the initial compressive yield stress, $P_y(ø_0)$, can be determined accordingly.

[1] R. Buscall and L. R. White, *J. Chem. Soc. Faraday Trans 1*, 1987, **83**, 873-891.
[2] R. G. de Krester, D. V. Boger and P. J. Scales, *Rheol. Rev.*, 2003, 125-165.

S2. Compressive yield stress data at equilibrium, $P_y(\phi_{eq})$

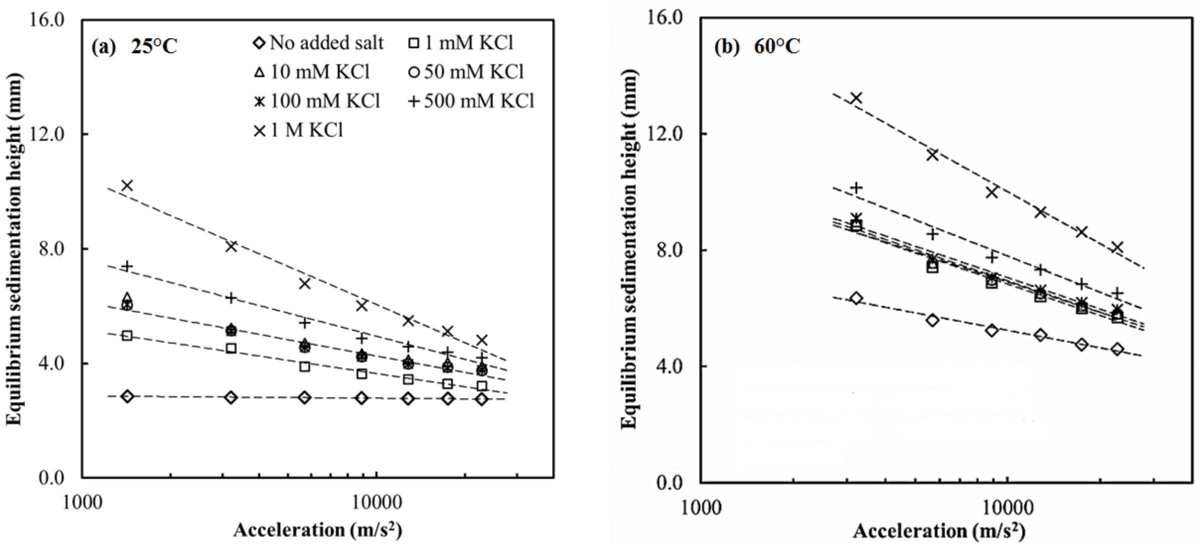

Figure S2.1: Equilibrium sedimentation height as a function of acceleration for 5 vol% PEGMA-stabilized polystyrene particle dispersions at varying electrolyte concentration at (a) 25°C and (b) 60°C, confirming the logarithmic decay of H_{eq} with acceleration in centrifugal field (Legend applicable to both graphs)