

Supporting Information

Tuning the bridging attraction between large hard particles by the softness of small microgels

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Figure S1 shows the storage moduli G' (filled symbols) and loss moduli G'' (open symbols) of mixtures with $\Phi_L = 0.30$ and various volume fraction of S(0). For samples without microgels ($\Phi_{S(0)}/\Phi_L = 0$, Fig.S1a) or samples with saturated adsorption of microgels ($\Phi_{S(0)}/\Phi_L = 0.6$, Fig.S1b), the G' and G'' can be scaled as $G' \sim \omega^2$ and $G'' \sim \omega$, which are typical characteristics of viscoelastic liquid at the low frequency region. For mixtures with $0.0025 \leq \Phi_{S(0)}/\Phi_L \leq 0.53$, solid-like characters are clear in the rheological data: G' is almost frequency-independent and $G' > G''$ in the experimental linear viscoelastic regime ($0.1 < \omega < 100$ rad/s). It is obvious that, addition of small particles first triggers a liquid-to-gel transition, and the gel becomes stronger with increasing amount of small particles (Fig. S1a). Up to a certain value, further increasing $\Phi_{S(0)}$ gradually weakens the gel, and eventually samples regain fluidity at saturated adsorption (Fig. S1b). Black and red symbols correspond to liquid and gel state, respectively. The arrows indicate the direction of increasing $\Phi_{S(0)}$.

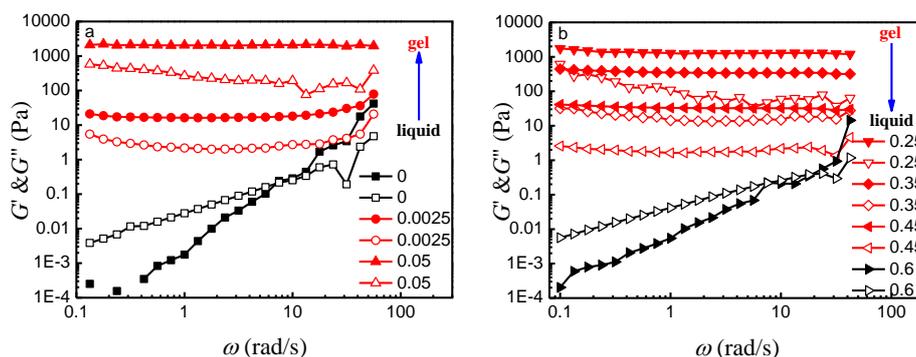


Figure S1. Frequency sweep data for concentrated mixed suspensions with $\Phi_L = 0.30$ at various volume fraction of S(0). The legends indicate the mixing ratio $\Phi_{S(0)}/\Phi_L$. Filled symbols correspond to storage moduli G' and open symbols correspond to loss moduli G'' . The stress amplitude is set to be $\sigma = 0.1$ Pa.

Figure S2 shows the storage moduli G' (filled symbols) and loss moduli G'' (open symbols) of mixtures with $\Phi_L = 0.30$ and various volume fraction of S(100). A liquid-gel transition could be triggered by the addition of small amount of S(100) as shown in Fig. S2a. Addition of more $\Phi_{S(100)}$ decrease the mechanical properties of gels as can be seen in Fig. S2b. Unlike the case shown in Fig.S1, where samples regain its

fluidity, samples here show no signs to get into liquid-like state at high $\Phi_{S(100)}$ but re-entrance another solid state (Fig. S2c) by further increasing $\Phi_{S(100)}$. This may be due to the jamming effect when the total particle volume fraction, $\Phi_L + \Phi_S \geq 0.58$.

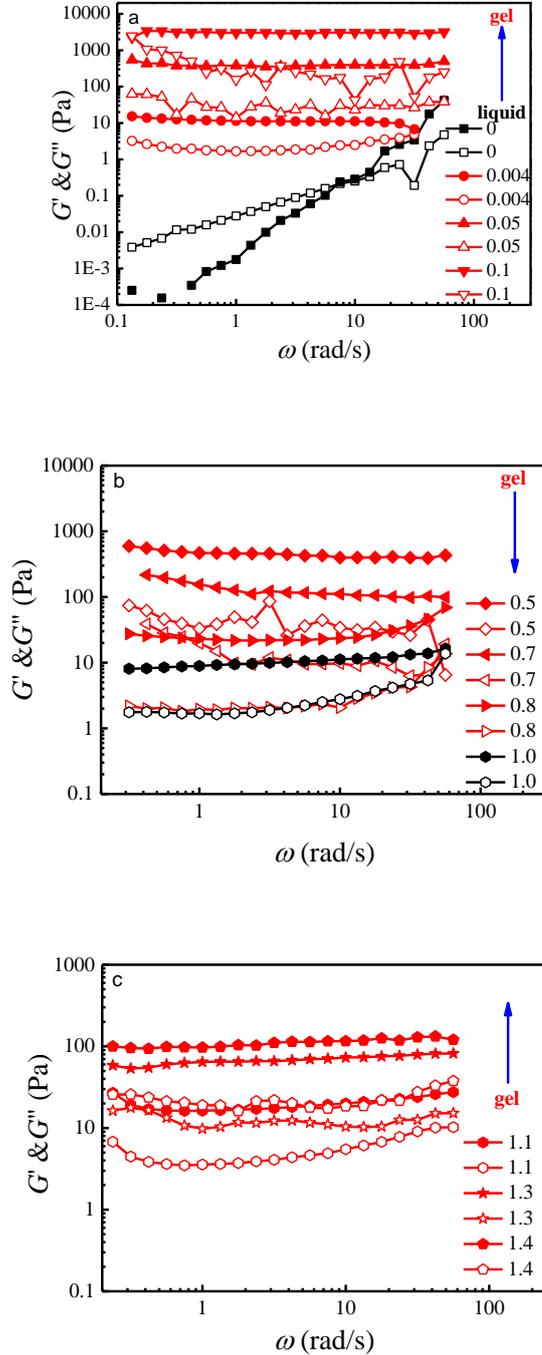


Figure S2. Frequency sweep data for concentrated mixed suspensions with $\Phi_L = 0.30$ at various volume fraction of S(100). The legends indicate the mixing ratio $\Phi_{S(100)}/\Phi_L$. Filled symbols correspond to storage moduli G' and open symbols correspond to loss

moduli G'' . The stress amplitude is set to be $\sigma = 0.1$ Pa.

Acknowledgements / Disclaimer

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