Supplementary Information

Two-Step Yielding Behavior of Densely Packed Microgel Mixtures with Chemically Dissimilar Surfaces and Largely Different Sizes

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Supporting information 1. $\sigma_{c1}^{\text{LAOS}}$ and $\sigma_{c1}^{\text{flow}}$ as a function of $\phi_N$ for N/NM-0.9, 0.3 and N/(N)NM-0.09. The values of $\sigma_c$ determined from the two different methods (LAOS and steady flow measurements) agree well with each other.
**Supporting Information 2.** \( \omega \) dependence of \( G' \) and \( G'' \) in linear response regime for N/NM-0.3 with \( \phi_0 = 0.4 \) before and after the heating treatment. In the heating treatment, the specimen was heated to 55 \( ^\circ \)C in the shrunken state above the LCST, and then cooled to 25 \( ^\circ \)C in the swollen state. No appreciable difference in linear viscoelasticity before and after heating ensures that the packing at 25 \( ^\circ \)C corresponds to the equilibrium state.
Supporting Information 3. Effect of ionic strength ($I$) on (a) linear dynamic viscoelasticity, (b) steady state flow for the pastes ($\phi = 1$) and (c) particle dimension using the N-microgels in which a finite amount (0.5 mol%) of fumaric acid at pH = 11 ($>>$ pKa for Fac; pKa$_1 = 3.1$ and pKa$_2 = 4.4$) at 25 °C. No appreciable effect of $I$ is observed in (a)-(c).