Electronic Supplementary Material (ESI) for Soft Matter. This journal is © The Royal Society of Chemistry 2020

## **Supplementary Information**

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

Saori Minami,<sup>†</sup> Takumi Watanabe,<sup>‡</sup> Yuma Sasaki,<sup>‡</sup> Haruka Minato,<sup>‡</sup> Atsushi Yamamoto,<sup>†</sup> Daisuke Suzuki,\*<sup>,‡,¶</sup> and Kenji Urayama<sup>\*,†</sup>

<sup>†</sup> Department of Macromolecular Science and Engineering, Kyoto Institute of Technology,

Sakyo-ku, Kyoto 606-8585, Japan

<sup>‡</sup>Graduate School of Textile Science & Technology, <sup>¶</sup>Research Initiative for Supra-Materials, Interdisciplinary Cluster for Cutting Edge Research, Shinshu University, Ueda 386-8567, Japan



**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  $\phi_N = 0.4$  before and after the heating treatment. In the heating treatment, the specimen was heated to 55 °C in the shrunken state above the LCST, and then cooled to 25 °C in the swollen state. No appreciable difference in linear viscoelasticity before and after heating ensures that the packing at 25 °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<sub>1</sub> = 3.1 and pKa<sub>2</sub> = 4.4) at 25 °C. No appreciable effect of *I* is observed in (a)-(c).