Electronic Supplementary Material (ESI) for Soft Matter.
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Supplementary Information

Collapse-induced phase transitions in binary interfacial microgel monolayers

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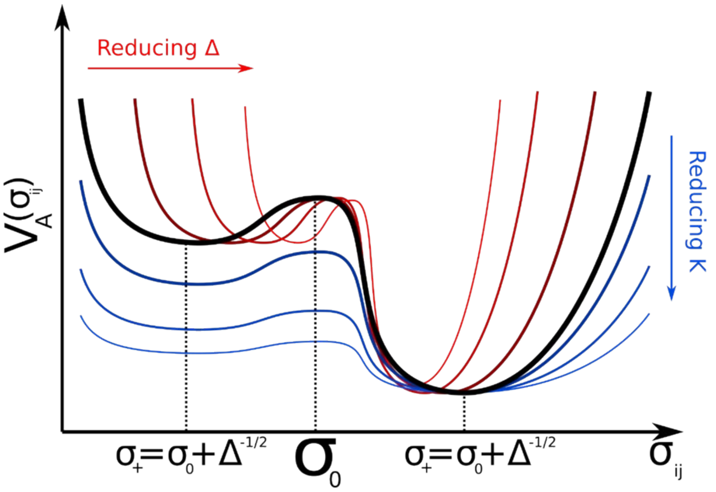

Figure S1: Effect of the size Delta and the stiffness K over the augmented potential defined in eq.1 . To model different particle types, we select Delta (and σ_0) that reproduces the experimental sizes. The stiffness K controls which type of particle collapses first because it tunes the energy barrier between the expanded ($\sigma_{ij}=\sigma_+$) and the collapsed ($\sigma_{ij}=\sigma_-$) states.

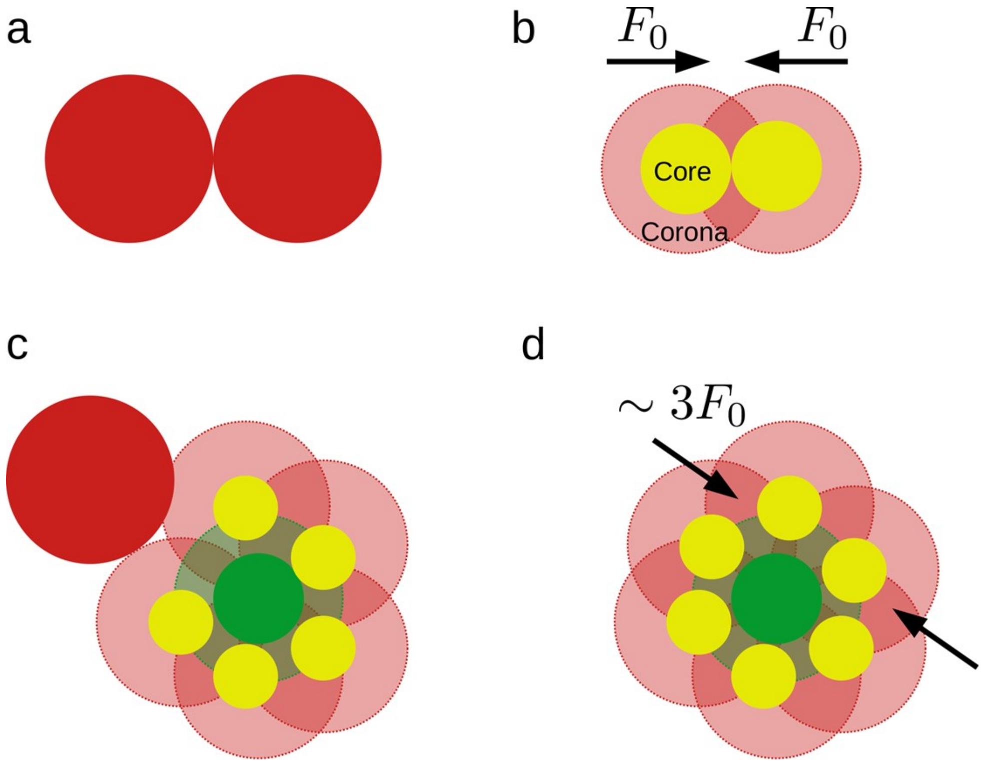

Figure S2: Flower-like defects in simulations. A force of magnitude F0 is required to compress a pair of microgels (a) into core-core contact (b). However, a much larger force is required to create a prefect flower-like defect, because it requires simultaneously many corona-corona overlaps. For example, to add the last petals to the flower in (c), the pushing force has to compress at least 3 corona-corona contacts, so the force has to be at least $3F_0$. In the manuscript, we partially facilitate the formation of such defects, by using the augmented potentials with additional many-body interactions that make the collapsed particles (the first petals) more likely to collapse again, thus lowering the energy required to add petals to the flower.

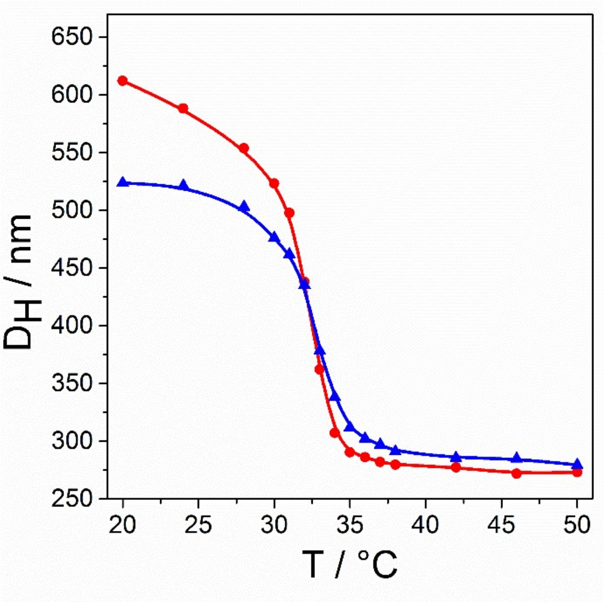

Figure S3: Hydrodynamic diameter (D_H) as a function of temperature measured by dynamic light scattering (DLS) for PNiPAm microgels with 2.5 mol% BIS (red) and 5.0 mol% BIS (blue) as crosslinker.

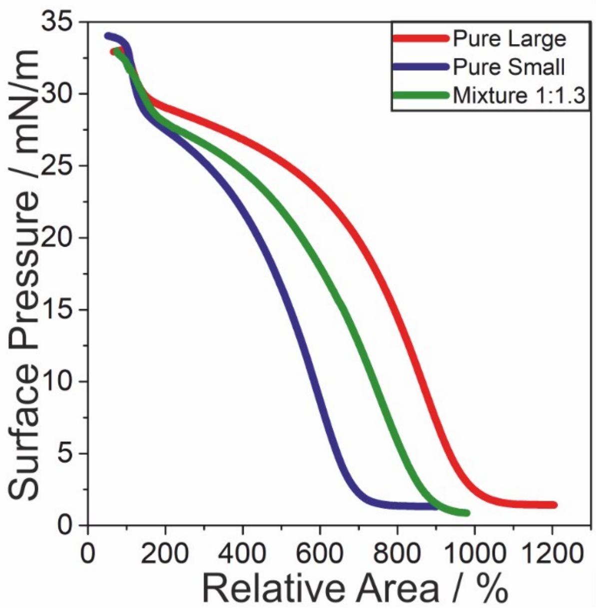

Figure S4: Surface pressure-area isotherms of the pure small and large microgels as well as a 1:1.3 mixture.

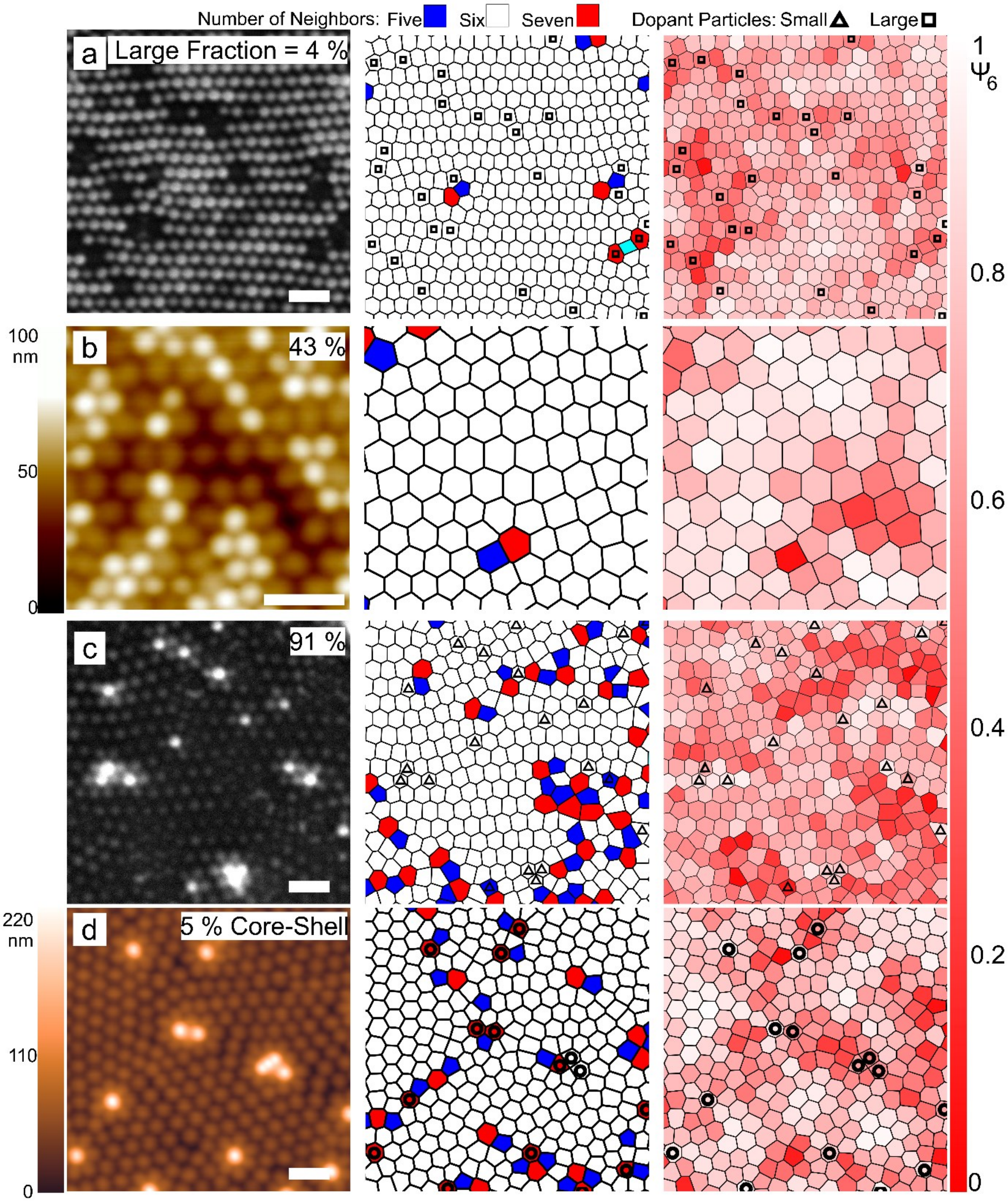

Figure S5: Scanning electron microscopy and atomic force microscopy images of close-packed monolayers at different mixing ratios with corresponding Voronoi tessellations colored according to number of neighbors and Ψ_6 parameters. a) Ratio large:small = 1:24, b) Ratio large:small = 1:1.3, c) Ratio large:small = 10:1, d) Ratio microgel:core-shell = 19:1. Scale bar = 1 µm.