Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Chain Stiffness Regulates Entropy-Templated Perfect Mixing of Polymer-Tethered Janus Nanoparticles at Interfaces

The file includes:

Figures S1-S5

Supplementary Videos 1-3

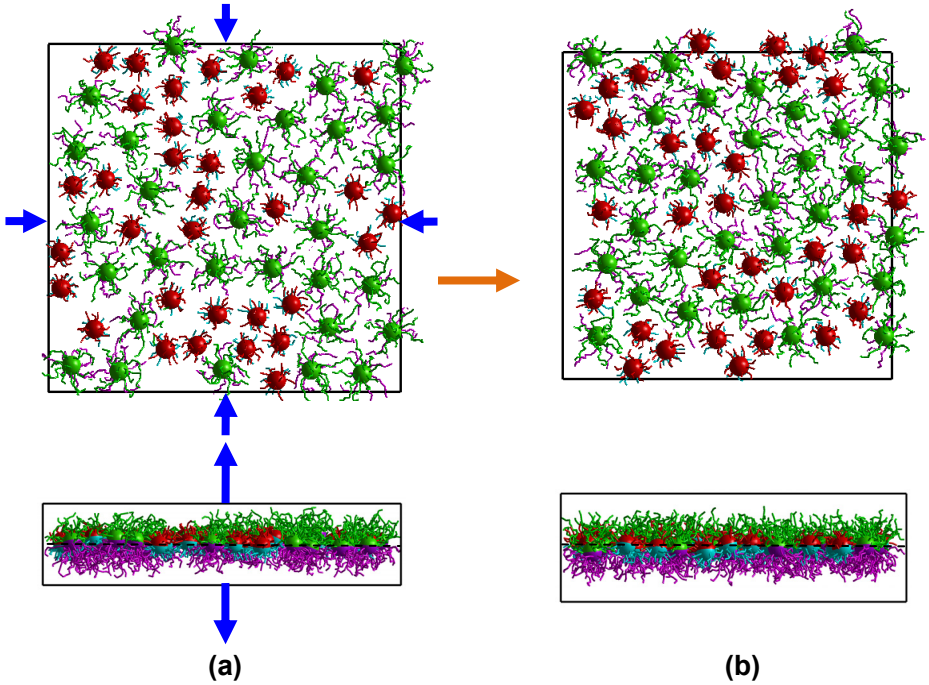

Figure S1. The structural change of the simulation box before (a) and after (b) a mechanical pressure is applied. In (a), the arrows indicate the lateral and vertical changes of the simulation box. The dashed black lines denote the fluid interface whereas the fluid beads are not shown for clarity.

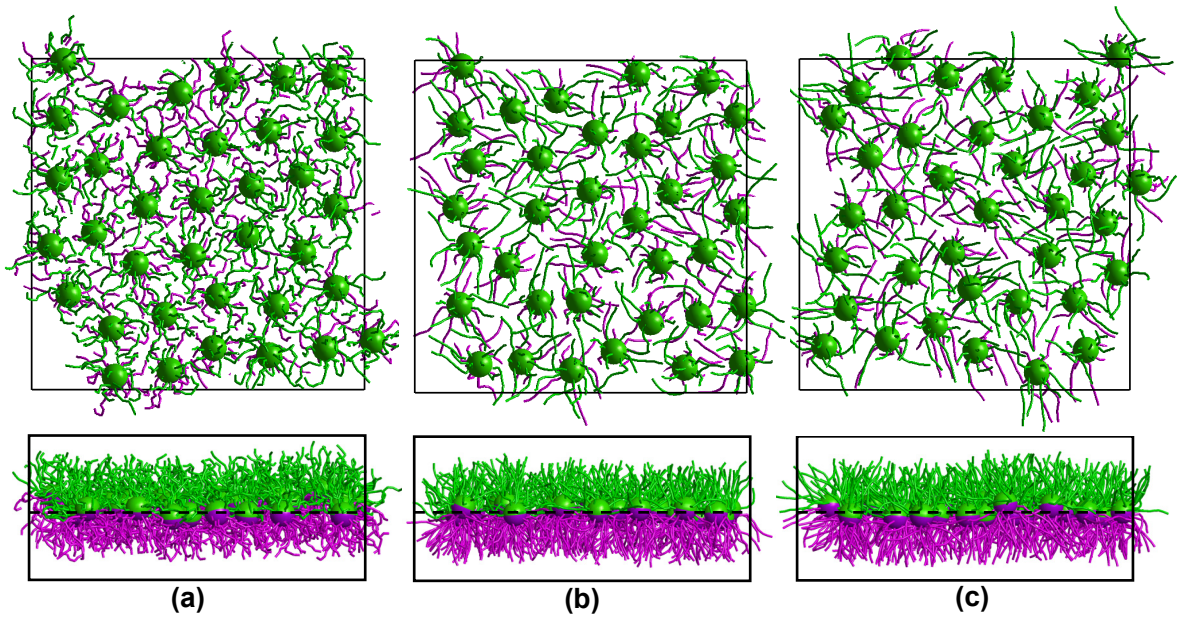

Figure S2. The top- and side-view snapshots show representative interfacial nanopatterns for the systems of pure long-tether nanoparticles in response to a lateral compression with λ=0.92, where l_p=0.50r_c (a), l_p=3.28r_c (b), and l_p=5.90r_c (c). The dashed black lines denote the fluid interface whereas the fluid beads are not shown for clarity. Detailed dynamical processes can be found in Supporting Videos 1-3.

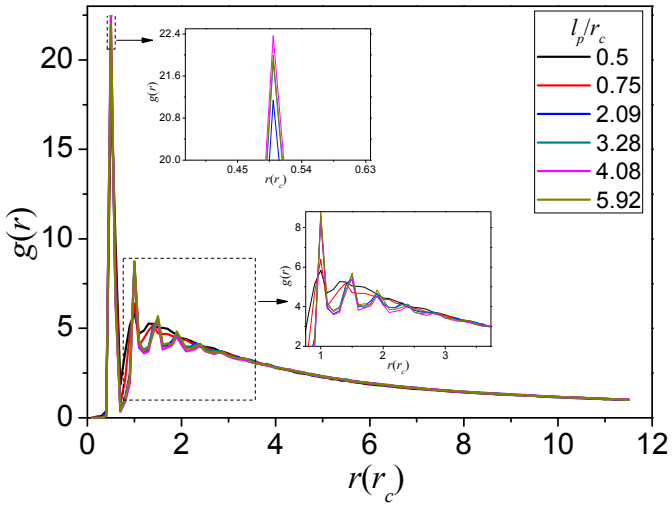

Figure S3. The pair correlation function $g(r)$ at various persistent length, l_p. The insetting images enlarge the local structures as marked by the dashed rectangles.

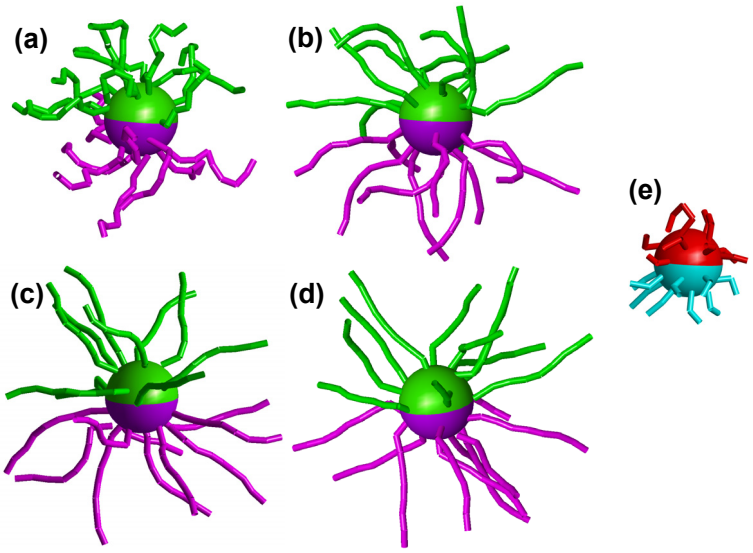

Figure S4. (a)-(d) Janus nanoparticles tethered with long ligand chains of various persistence length, l_p: (a) l_p=0.50r_c, (b) l_p=2.09r_c, (c) l_p=3.28r_c, and (d) l_p=7.36r_c. The contour length of the long tethers is L=4.0r_c. (e) Janus nanoparticles tethered with short ligand chains of a contour length 1.0r_c. Color scheme: (green) long tether preferring the *A* phase, (pink) long tether preferring the *B* phase, (red) short tether preferring the *A* phase, (cyan) short tether preferring the *B* phase.

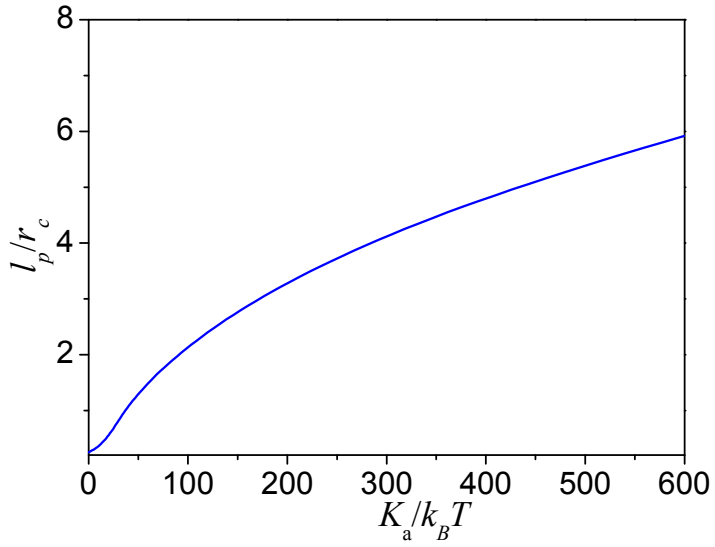

Figure S5. Persistence length of the polymer chain, l_p, as a function of K_a. The line is to guide the eye. Here l_p is calculated through systematical DPD simulations of a single polymer chain, as the method reported in Ref.S1. The relationship between K_a and l_p indicates a detailed correlation between our simulations results and the possible experimental systems.

Ref.S1: Micka, U.; Kremer, K. *J. Phys.*: *Condens. Matter* **1996**, *8*, 9463-9470.

Supporting Videos:

Video S1. The temporal evolution of the interfacial nanoparticle configuration for the system of pure long-tether nanoparticles in response to a lateral compression of $\delta \approx 0.1$, where $\boldsymbol{l_p = 0.50 r_c}$.

Video S2. The temporal evolution of the interfacial nanoparticle configuration for the system of pure long-tether nanoparticles in response to a lateral compression of $\delta \approx 0.1$, where $\boldsymbol{l_p = 3.28 r_c}$.

Video S2. The temporal evolution of the interfacial nanoparticle configuration for the system of pure long-tether nanoparticles in response to a lateral compression of $\delta \approx 0.1$, where $\boldsymbol{l_p = 5.90 r_c}$.