Electronic Supplementary Material (ESI) for Soft Matter.

Interfacial self-assembly of SiO2-PNIPAM core-shell particles with varied crosslinking density

Supplementary Information

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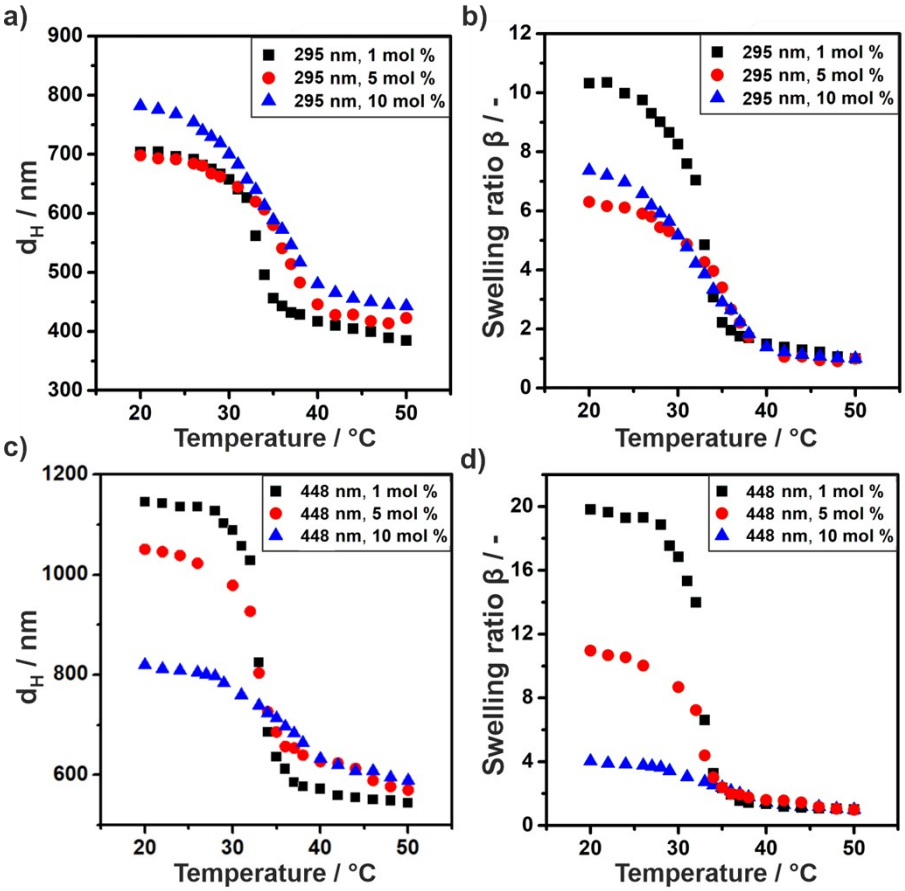

Figure S1: a) Hydrodynamic diameter d_H of core-shell particles with a 295 nm core as a function of temperature . b) Swelling ratio of core-shell particles with a 295 nm core as a function of temperature. c) Hydrodynamic diameter d_H of core-shell particles with a 448 nm core as a function of temperature . b) Swelling ratio of core-shell particles with a 448 nm core as a function of temperature.

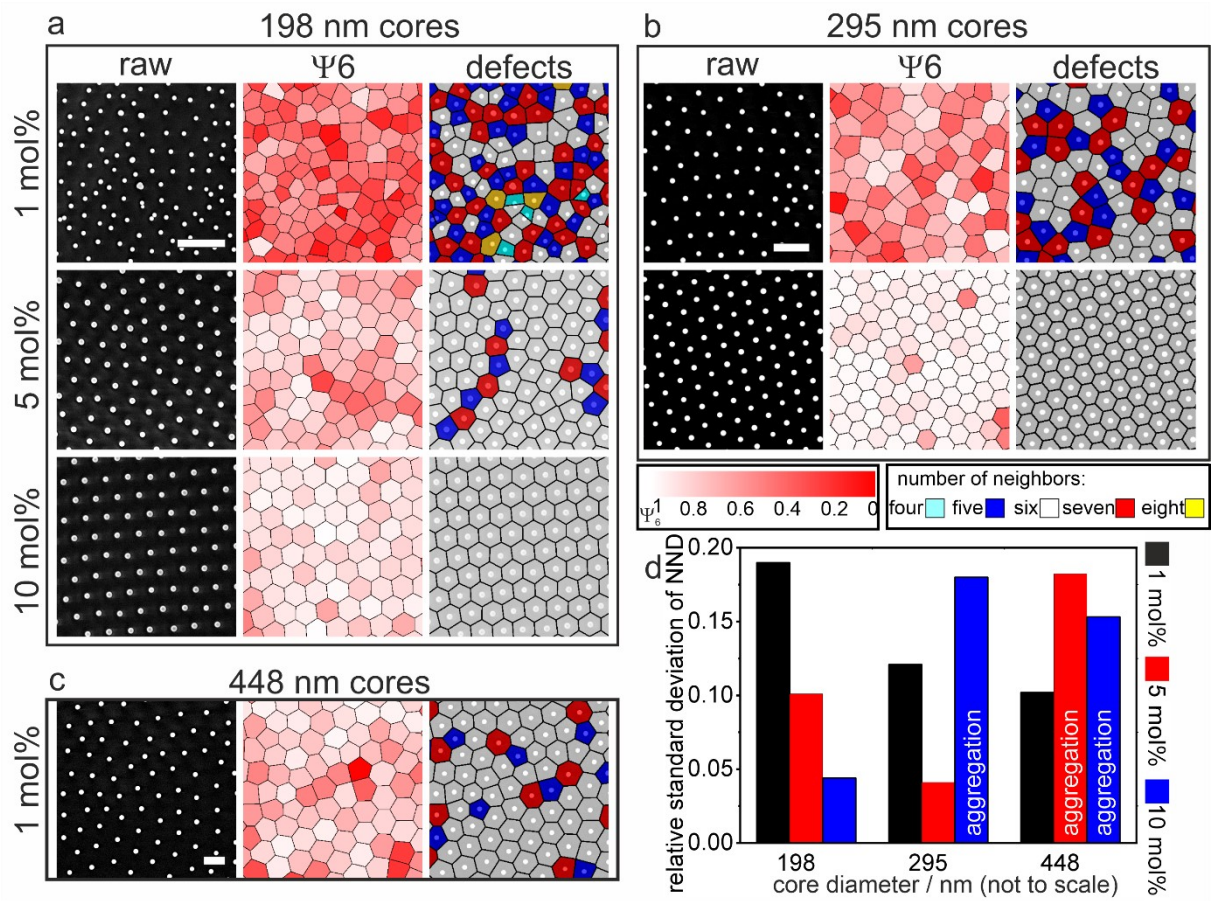

Figure S2: Order analysis. a-c) Raw and post-processed SEM images of $\Psi 6$-parameter and defect analysis for core-shell particles with 198 nm (a), 295 nm (b) and 448 nm (c) cores. Due to aggregation, the 295nm-10% particles and the 448 nm-5% and -10% particles are not shown. Scale bar: 2µm d) Relative standard deviation of nearest neighbor distances for different particles.

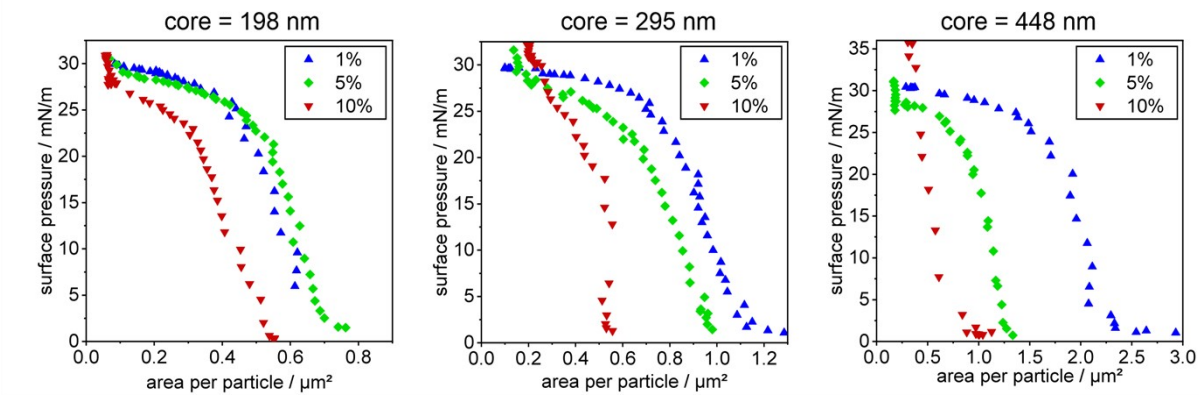

Figure S3: Surface pressure-area per particle isotherms for three different core diameters and crosslinking densities. The particle areas were extracted from SEM images and correlated to the respective surface pressures.

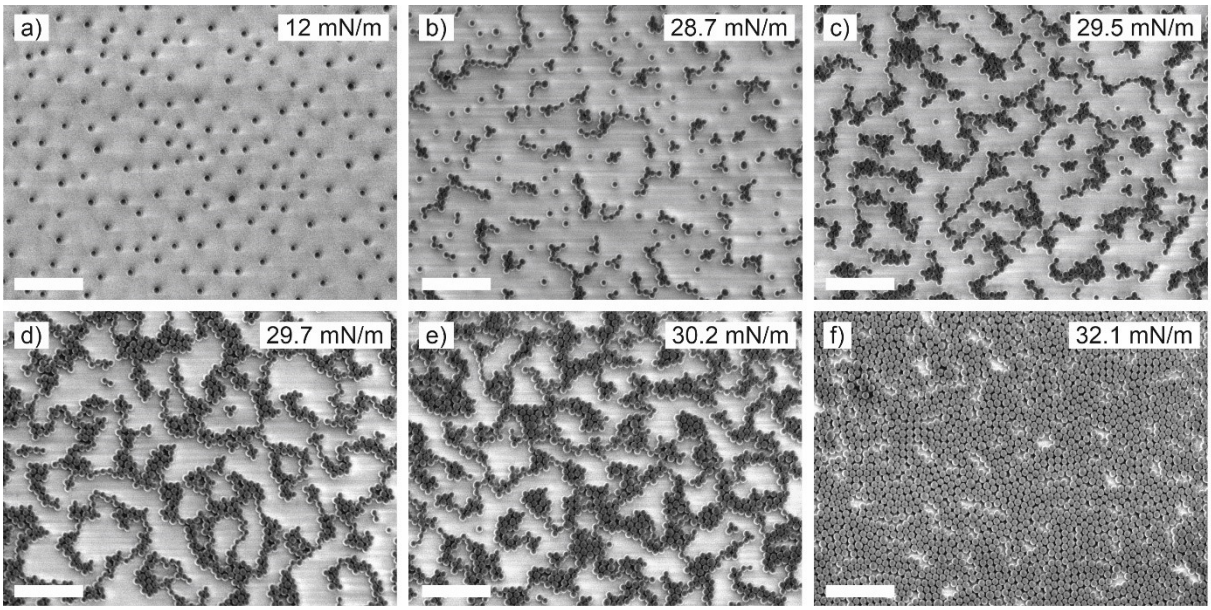

Figure S4: SEM images of 198 nm-1 mol% core-shell particles deposited from an air/water interface at different surface pressures. They undergo a phase transition from a hexagonally non-close packed arrangement (a) over chain-like networks (b-e) to a hexagonal close-packed phase (f) under increasing pressure. *Scale bar: 2 µm*

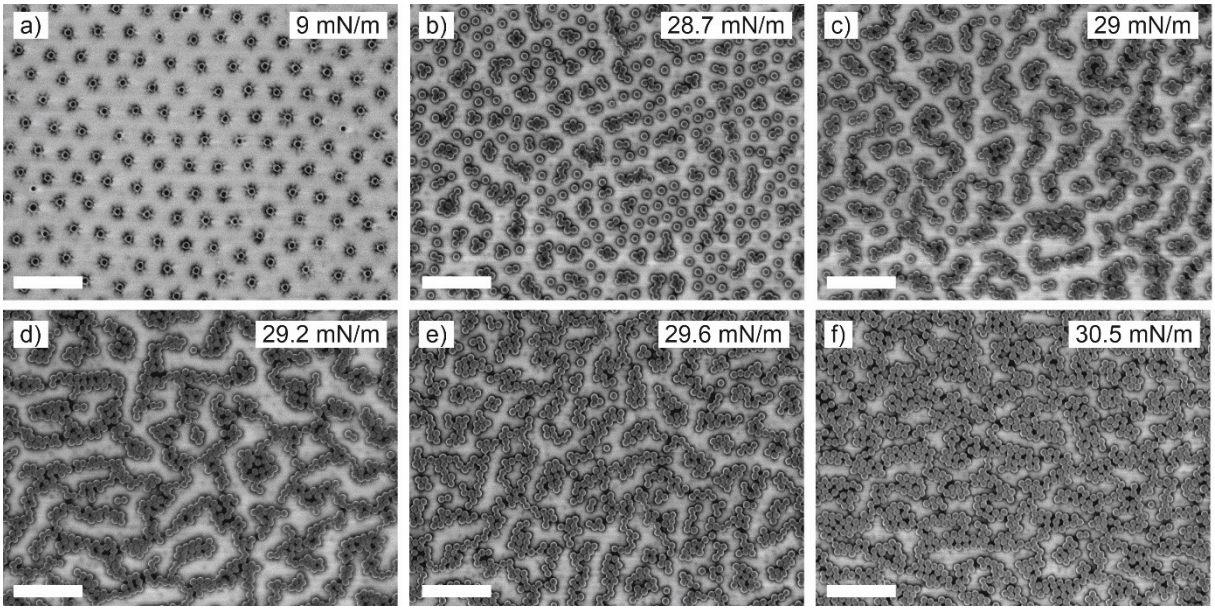

Figure S5: SEM images of 198 nm-5 mol% core-shell particles deposited from an air/water interface at different surface pressures. They undergo a phase transition from a hexagonally non-close packed arrangement (a) to chain-like networks (b-f), becoming increasingly dense with increasing external pressure. *Scale bar: 2 µm*

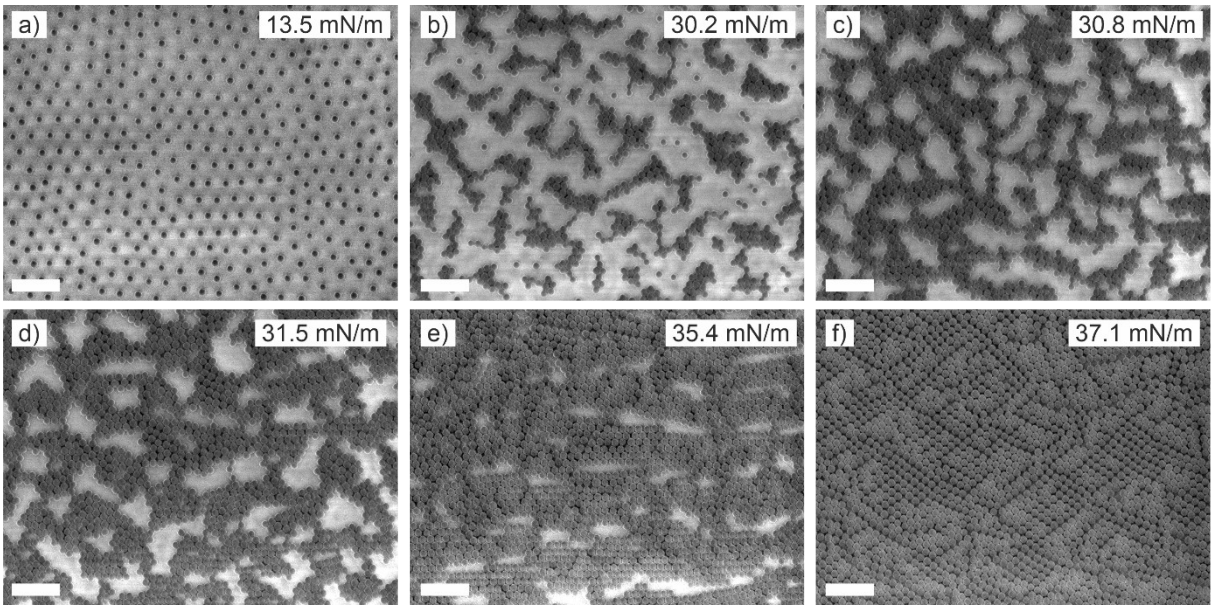

Figure S6: SEM images of 295 nm-1 mol% core-shell particles deposited from an air/water interface at different surface pressures. The particles transit from a hexagonally non-close packed arrangement (a) over a network of connected hexagonal islands (b-e) to a hexagonal close-packed phase coexisting with areas of *rhombic* packing (f). *Scale bar: 2 µm*

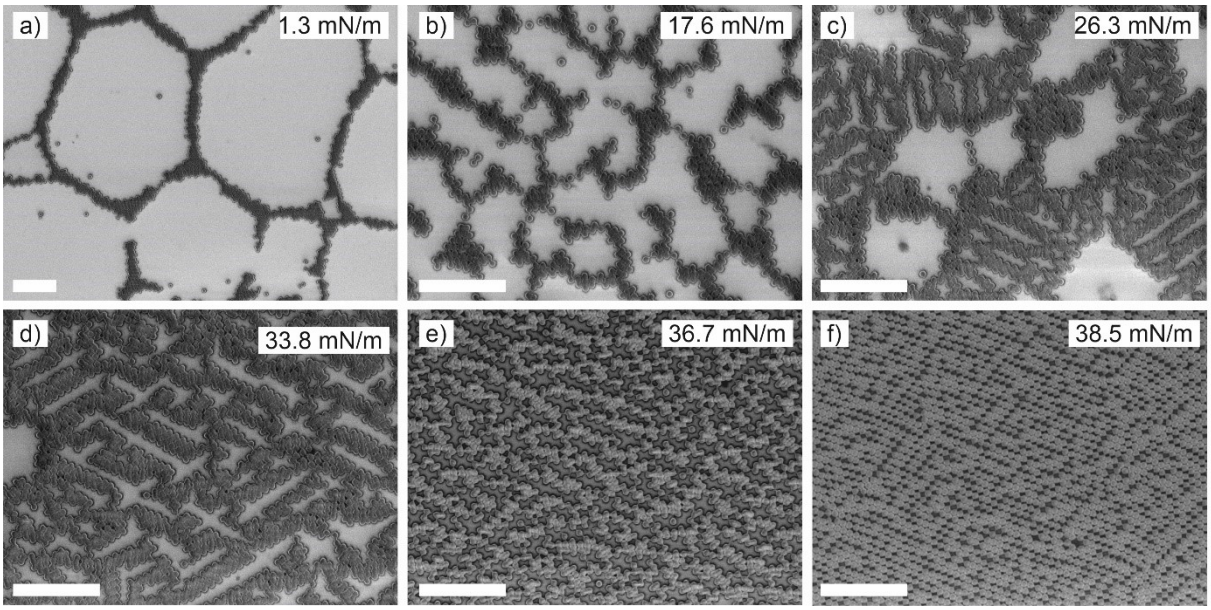

Figure S7: SEM images of 295 nm-10 mol% core-shell particles deposited from an air/water interface at different surface pressures. The particles agglomerate when attached to the interface (a,b) and transition to zig zag like structures under compression (c-e). They arrange in a rhomboid lattice at high surface pressures (f). *Scale bar: 5 µm*

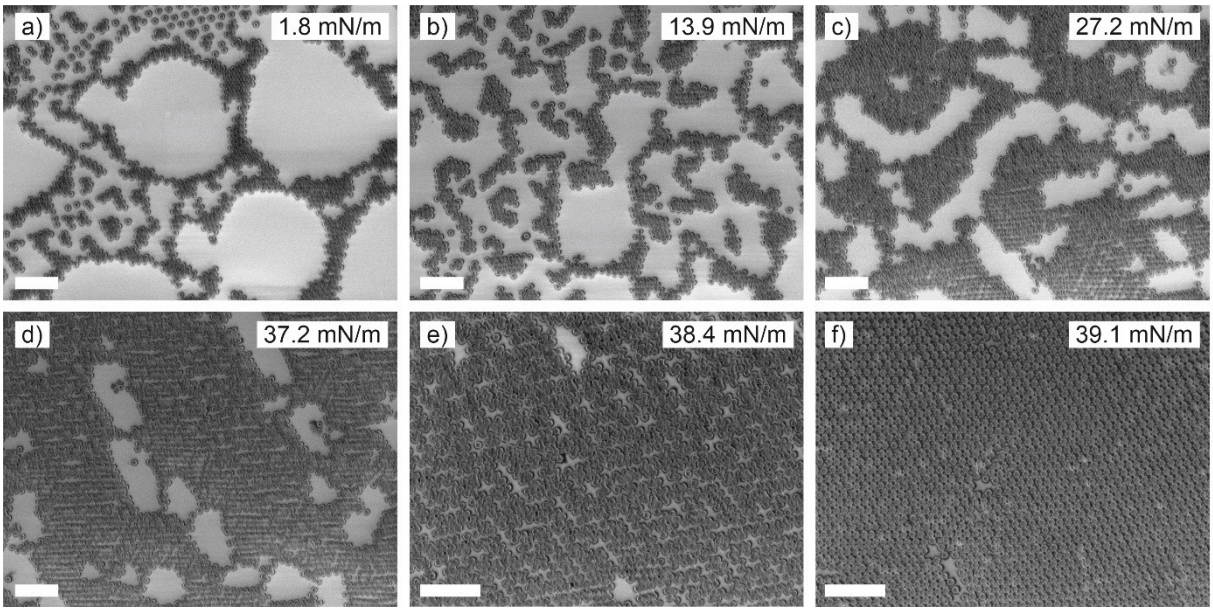

Figure S8: SEM images of 448 nm-5 mol% core-shell particles deposited from an air/water interface at different surface pressures. The particles agglomerate when attached to the interface (a). During the compression, the islands become more dense (b-e) and then form a hexagonal close packed phase (f). *Scale bar: 5 µm*

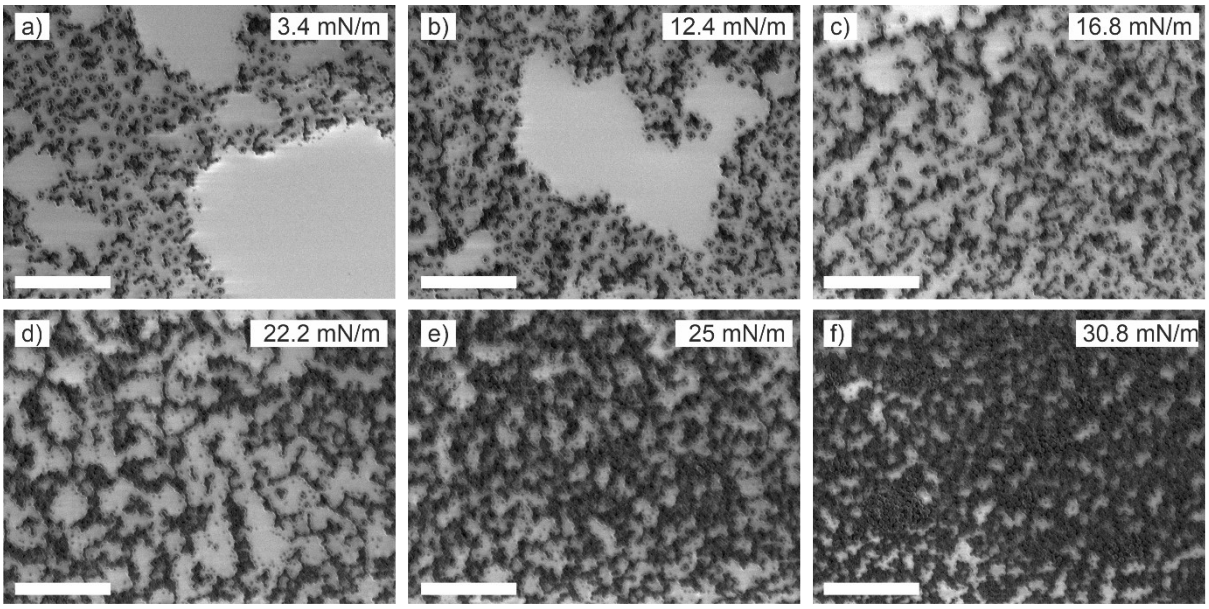

Figure S9: SEM images of 448 nm-10 mol% core-shell particles deposited from an air/water interface at different surface pressures. The particles were not able to form a stable monolayer at any external pressure and aggregated into ill-defined structures. *Scale bar: 5 µm*

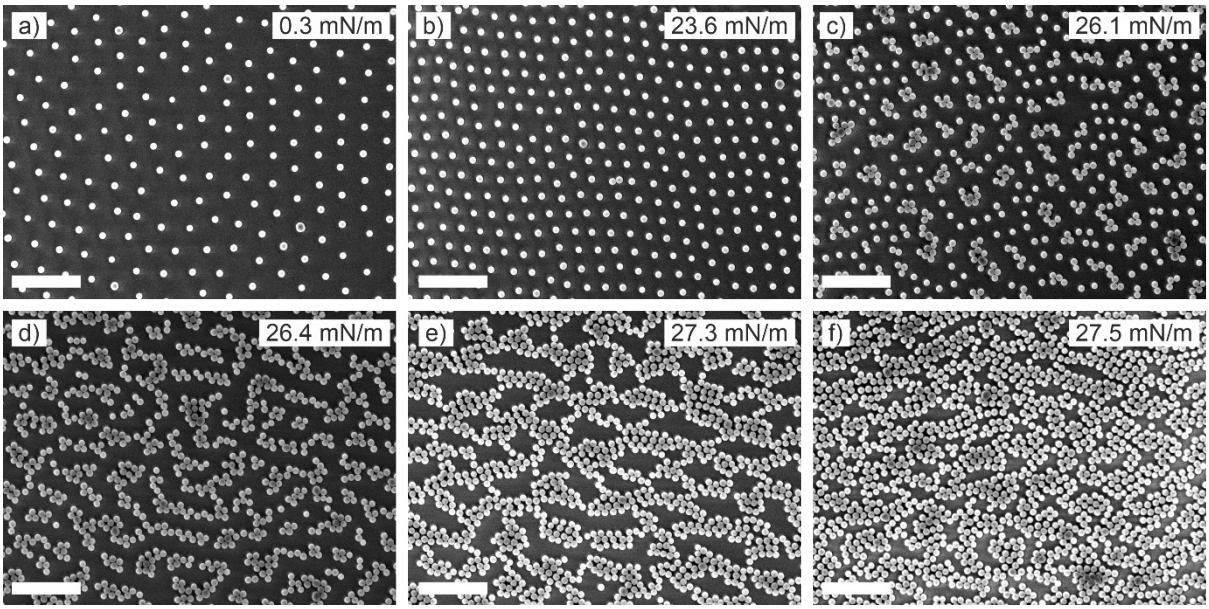

Figure S10: SEM images of the deposited 198 nm-10 mol% core-shell particles after the shells were removed, used for statistical image analysis of Fig. 3. Scale bar: 2 µm.

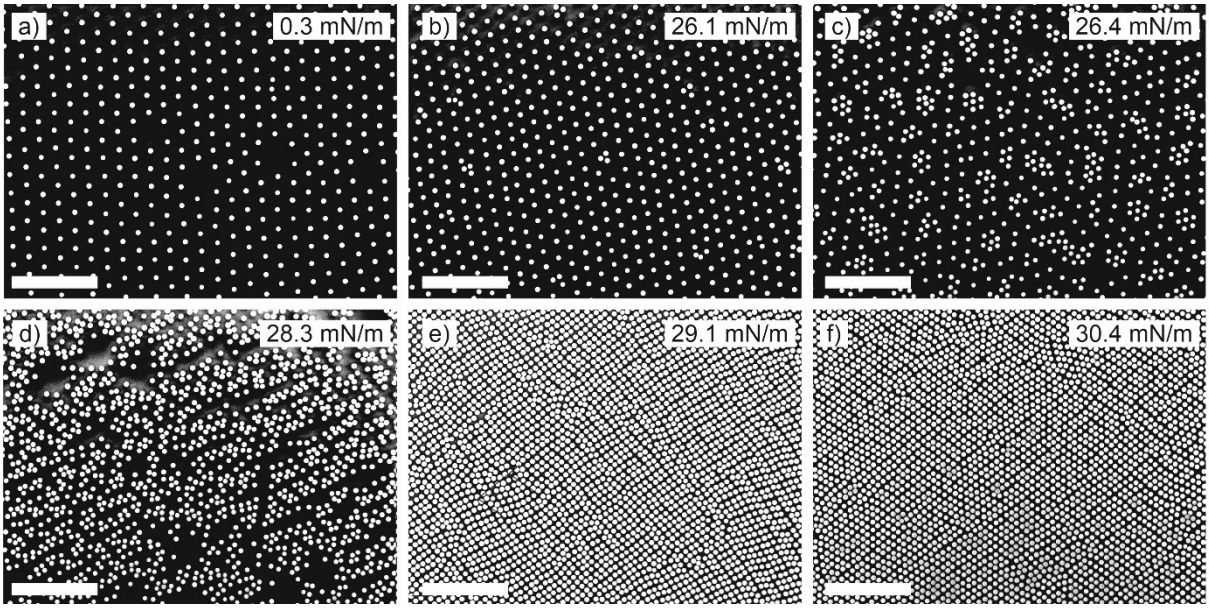

Figure S11: SEM images of the deposited 295 nm-5 mol% core-shell particles after the shells were removed, used for statistical image analysis of Fig. 4. Scale bar: 5 µm.

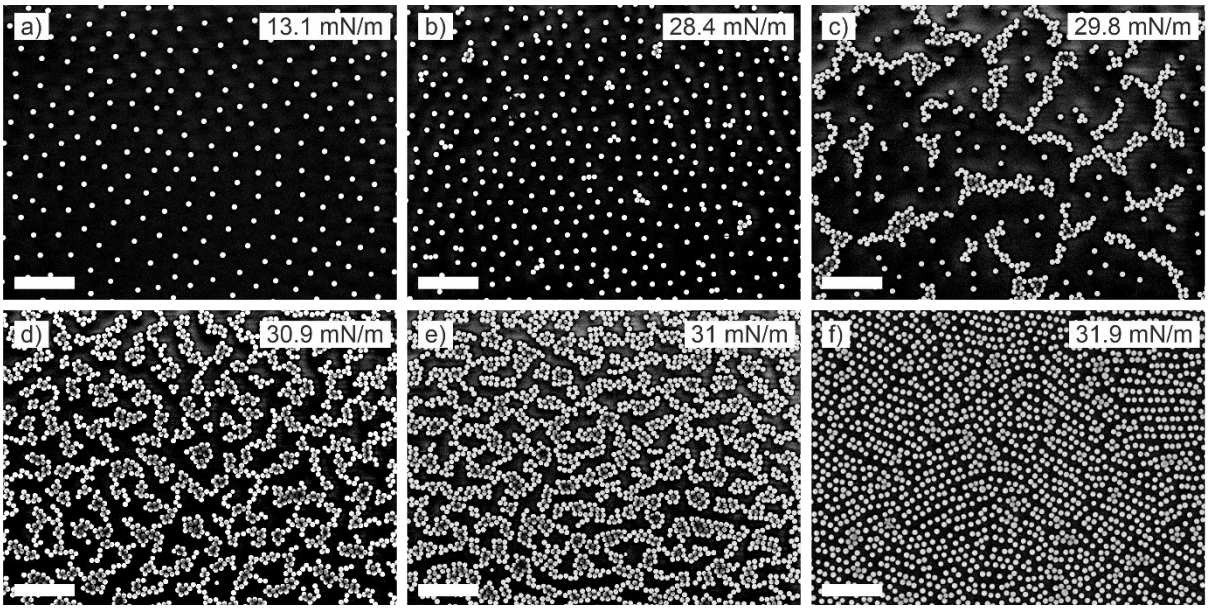

Figure S12: SEM images of the deposited 448 nm-1 mol% core-shell particles after the shells were removed, used for statistical image analysis of Fig. 5. Scale bar: 5 µm

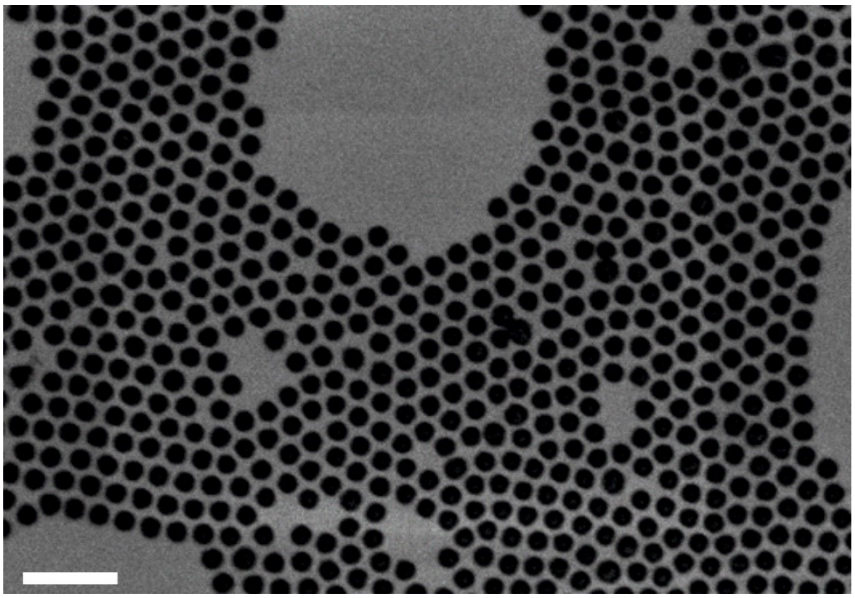

Figure S13: SEM image of 198 nm - 5 mol% core-shell particles deposited from an air/water interface at a surface pressure of 0.5 mN/m. Scale bar: 4 µm.