

Supporting Information

Triple-line dynamics of a soft colloid-laden drop on a hydrophobic surface

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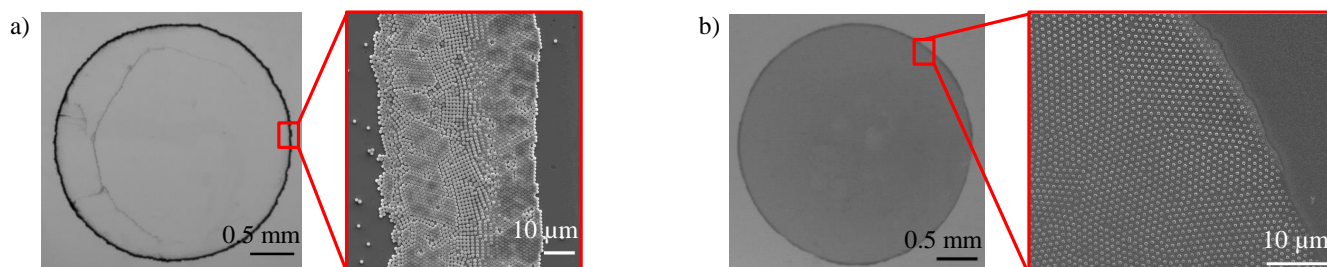


Figure S1: a) Coffee-ring like deposit formed by PS particles on the hydrophilic substrate and the multi-layer thick ring imaged using SEM b) Uniform deposit formed by p-NIPAM microgel particles on the hydrophilic substrate and the monolayer particle assembly imaged using SEM.

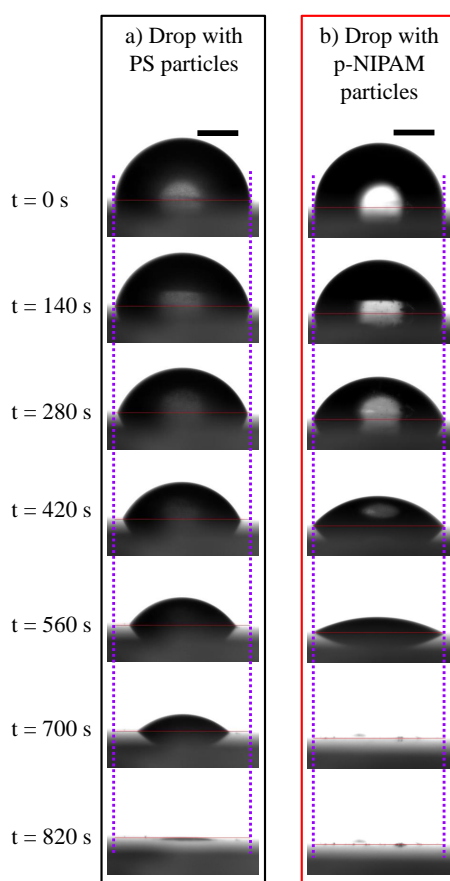


Figure S2: Side view images of drops containing a) PS particles and b) p-NIPAM microgel particles on hydrophobic substrate at different times during the evaporation. Purple dotted lines in the images are used to keep track of the drop diameter as the evaporation progresses. Scale bar: 0.5 mm.

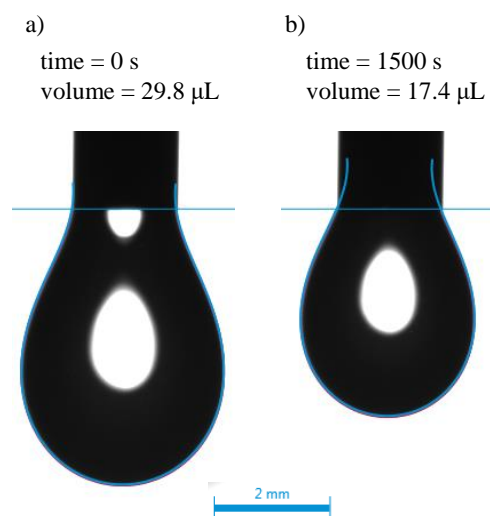


Figure S3: Images of the microgel-laden pendant drop at a) time = 0 s and b) time = 1500 s during the pendant drop tensiometry measurement. The baseline (horizontal line through the drop contact with the needle) and the best fit to the detected drop contour (obtained by the drop shape analysis software) are also shown in the images in blue lines.

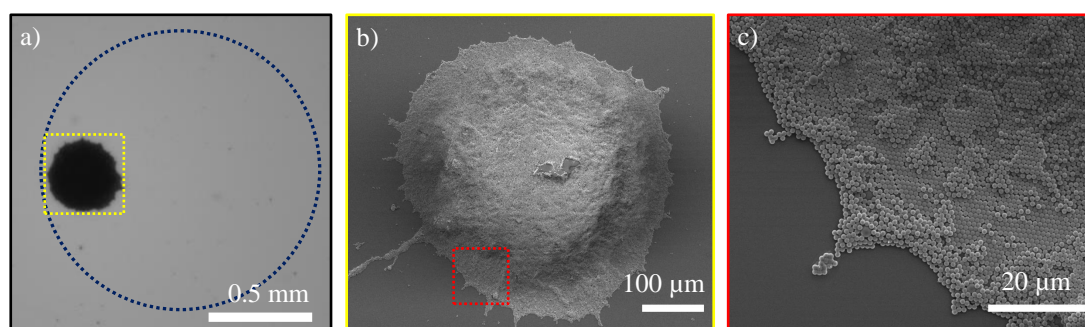


Figure S4: a) Inner deposit formed by the PS particles on the hydrophobic substrate. Blue dotted circle corresponds to the initial pinned contact line b) Magnified image of the deposit (within yellow dotted lines in (a)), obtained using SEM c) Magnified image of (b) within the red dotted lines, obtained using SEM. A mount-like deposit with multi-layer of particles can be seen. Scale bars: 100 μm (low magnification image) and 20 μm (high magnification image).



Figure S5: SEM images of the particulate deposit formed by the p-NIPAM microgel particles on the hydrophobic substrate at different regions marked a), b), c) and d) in the central low magnification image. Microgels at different levels of compression can be seen at different regions of the deposit. The yellow dashed line in (d) is drawn to indicate the exterior of the deposit. Near the periphery, the dark regions in the image (a and d) correspond to particles that are highly compressed and probably forming multi-particle layer. Scale bars: 500 μm (central low magnification image) and 10 μm (high magnification images a, b, c and d).

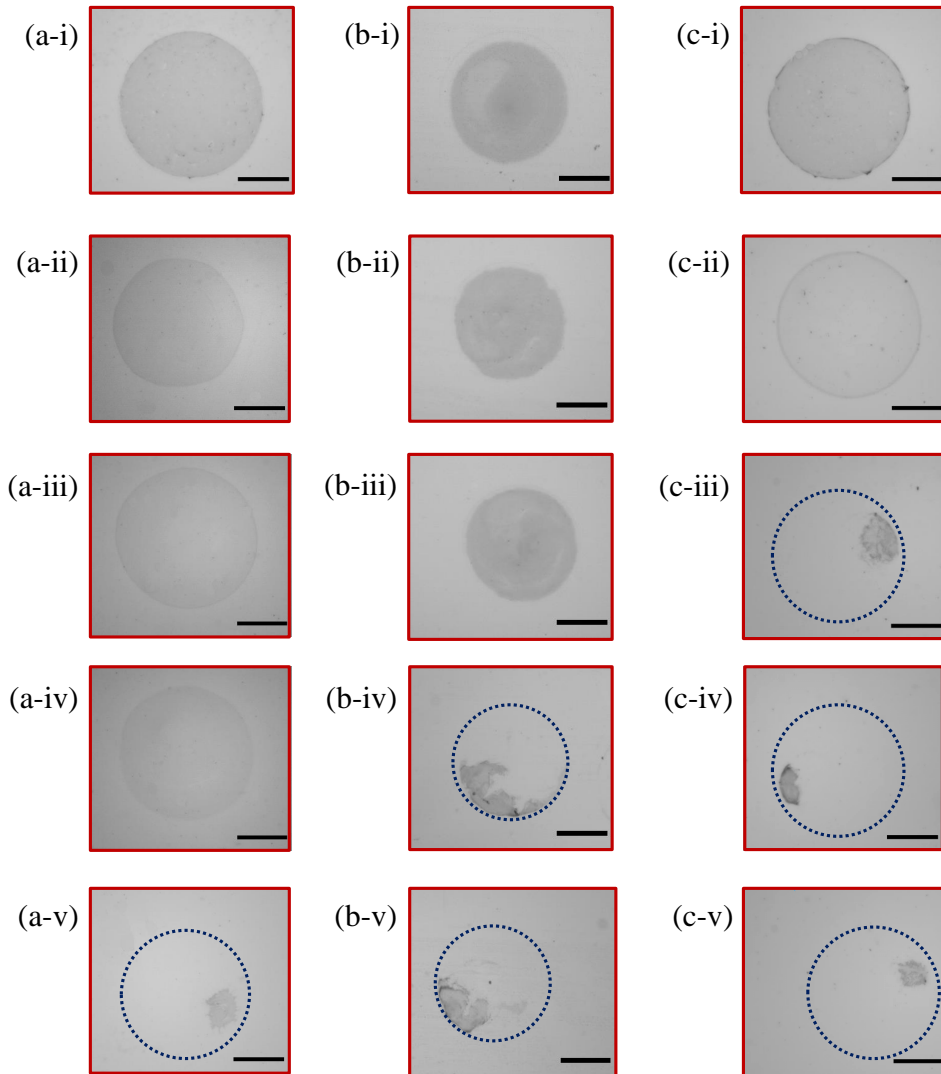


Figure S6: Final deposits from droplets dispersed with microgel particles of different crosslinking densities at various concentrations. (a-i) to (a-v) corresponds to CL2.5 microgel particles, (b-i) to (b-v) corresponds to CL4 microgel particles and (c-i) to (c-v) corresponds to CL7 microgel particles. Particles concentrations are 0.02, 0.015, 0.01, 0.007 and 0.004 wt% from top to bottom. Scale bars: 0.5 mm. Blue dotted circles for some images correspond to the initial pinned contact line.

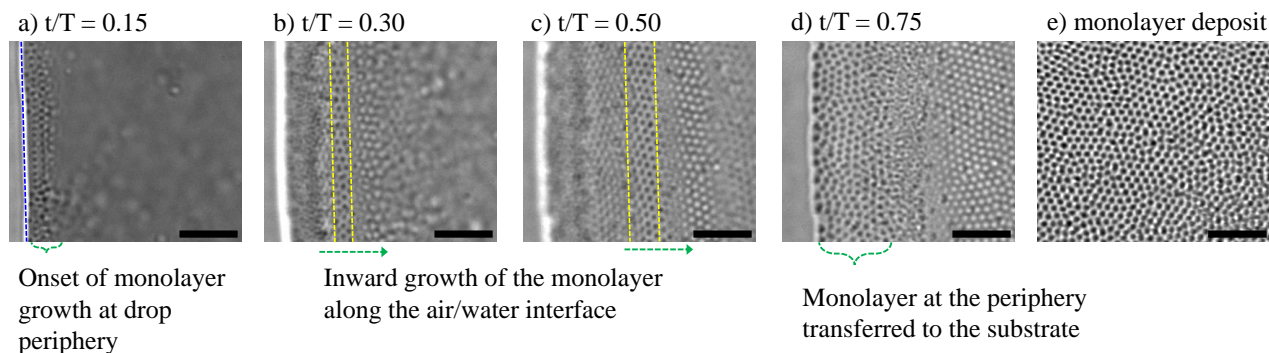


Figure S7: Optical microscopy images recorded near the periphery at different times during the evaporation of a microgel-laden drop on hydrophilic glass substrate. a) $t/T = 0.15$ b) $t/T = 0.30$ c) $t/T = 0.50$ and d) $t/T = 0.75$. The three-phase contact line is shown in blue dashed lines in (a). The capillary flow within the evaporating drop transports the dispersed particles to the periphery. The microgel particles owing to their inherent surface activity, adsorb onto the air/water interface near the periphery and this starts the growth of a particulate monolayer along the interface as shown in (a). With time, more and more particles reach the periphery and the monolayer grows along the air/water interface. As the interface is curved, only a thin strip of the interfacial monolayer of particles can be brought to focus as shown by the yellow dashed lines in (b) and (c). The particulate monolayer along the air/water interface of the drop descends with solvent loss from the drop owing to evaporation. Towards the end of the evaporation, the interfacial layer gets transferred to the substrate, resulting in the monolayer particulate deposit as shown in (d) and (e), which emphasises the fact that there are negligibly few particles on the substrate or within the bulk and majority of the particles assemble along the air/water interface during the evaporation. Scale bars: $10 \mu\text{m}$.

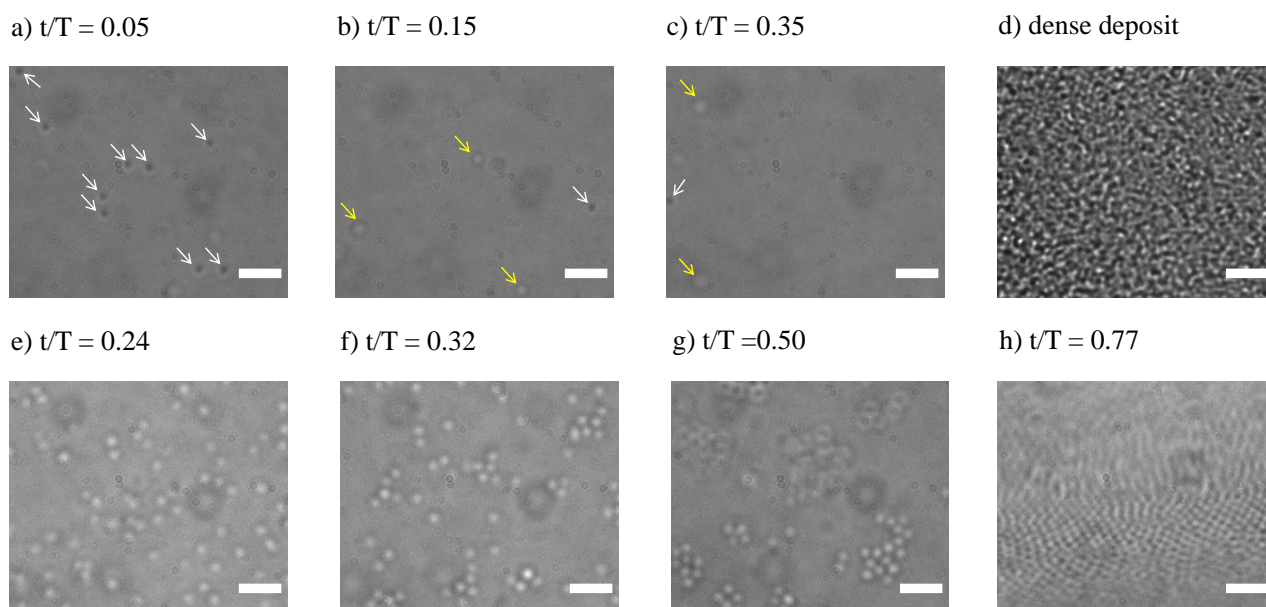


Figure S8: Optical microscopy images recorded during the evaporation of a drop dispersed with microgel particles, placed on a hydrophobic substrate. The top row (a-d) shows the solid/liquid interface in focus, at different times during the evaporation, a) $t/T = 0.05$ b) $t/T = 0.15$ and c) $t/T = 0.35$. In a)-c), the white arrows show particles on the substrate, whereas the yellow arrows show particles in the bulk (out of focus). The dense deposit at the end of evaporation is shown in (d). The bottom row (e-h) shows the liquid/air interface in focus, at different times during the evaporation e) $t/T = 0.24$ f) $t/T = 0.32$ g) $t/T = 0.50$ and h) $t/T = 0.77$. The number density of particles along the interface grows with time and the interfacial microstructure transforms from particle clusters (a) to a network spanning the entire interfacial area (d). From the images, we clearly see preferential adsorption of the microgel particles to the air/water interface of the evaporating drop in line with the expectation for amphiphilic microgel particles. Scale bars: $10 \mu\text{m}$.