Supporting Information

Roughening up polymer microspheres and their diffusion in a liquid.

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Figure S1. Scanning Electron Microscopy (SEM) micrographs of rough PS particles deformed in the presence of cigar-shaped calcium carbonate. The PS particles were exposed to heat for (A): 10 min, (B): 30 min, (C): 1hr, (D): 2 hr. Scale bars = 1.5 μm.



Figure S2. Scanning Electron Microscopy (SEM) micrographs of rough PS particles deformed in the presence of rod-shaped calcium carbonate. The PS particles were exposed to heat for (A): 10 min, (B): 30 min, (C): 1hr, (D): 2 hr. Scale bars = $1.5 \mu m$.



Figure S3. Low Magnification Scanning Electron Microscopy (SEM) micrograph of rodshaped PCC calcium carbonate particles with "crisscross" stacking. Scale bar = $20 \mu m$.





Figure S4. Example phase contrast light microscope images of rough particles in the wet state a) and b). Scale bar = 25 μ m. Distribution of rough particles in aggregation states according to the number of particles, N_{Ag} c). A total of 1079 particles were analysed in their wet state by imaging with light microscopy. The projected particle areas were measured and normalized against the average particle diameter for a single rough particle (determined from SEM images see Figure S7), giving N_{Ag}. The majority of particles remain well dispersed, *i.e.* N_{Ag} = 1, after deformation.



Figure S5. Sample phase contrast light microscope images a) and b) of rough particles after drying onto glass cover slips. Particles come together into higher agglomerates than in the wet state driven by capillary forces between particles adsorbed at the air-water interface.¹ Scale bar = $25 \mu m$.



Figure S5. Scanning Electron Microscopy (SEM) micrographs of rough PS particles deformed in the presence of nano-sized ZnO. The PS particles were exposed to heat for (A): 10 min, (B): 30 min, (C): 1hr, (D): 2 hr. Scale bars = $1.5 \mu m$.



Figure S6. A snapshot from the processed time series of a rough PS particle diffusing in bulk H_2O/D_2O with an overlay of the result from the Laplace-of-Gaussian detection (purple circle) and linear assignment protocol for linking displaying a typical Brownian trajectory.



Figure S7. Displacement of a typical Brownian rough poly(styrene) particle tracked in the bulk for 50 s at a time interval of 0.02 s a) and the normalized velocity autocorrelation function for all 25 rough particle tracks b).



Figure S8. Typical SEM images of smooth and rough particles utilized for size analysis a) and c) respectively. The result of the detect circles/Hough circles algorithm applied in imagej b) and the result of particle size analysis on the SEM image of the rough particles utilizing a thresholding technique d).

References

1 P. A. Kralchevsky, N. D. Denkov and K. D. Danov, *Langmuir*, 2001, **17**, 7694–7705.