Supplementary Material

FOTS-coating

For the Fluoroctatrichlorosilane (FOTS) coating of our substrates, we follow a vapour deposition protocol:

- clean substrate for 30min in Nitric Acid
- rinse with copious amounts of (milliQ) water, to remove traces of Nitric Acid
- place substrate for 30min in Plasma Cleaner
- place substrate in desiccator, next to (liquid) FOTS reservoir (roughly 30 uL for 4 cm^2)
- put under vaccum for 24h
- clean substrate for 30min in Chloroform
- clean substrate for 30min in IPA
- heat substrate to 60°C for 3h

Note, that our samples are sensitive to mechanical abrasion and we therefore refrained from using ultrasonic cleaning in this protocol. Especially the cleaning steps after the vapour deposition can also be done in an ultrasonic bath, if the sample allows for it.

Pinning

One of the signs for contact line pinning of an evaporating droplet is a sudden spike in the contact angle of the droplet. For an unpinned contact line, the contact angle stays constant as the contact line moves across the substrate due to decreasing droplet volume. As the contact line pins, the contact angle will start to decrease, because of the continued droplet volume loss. Once the contact line de-pins, the contact angle jumps back up to its original receding value. In order to show that we do not observe significant pinning events on our substrates throughout the evaporation of the droplets, we plot the change in contact angle as a function of time in Figure 1.

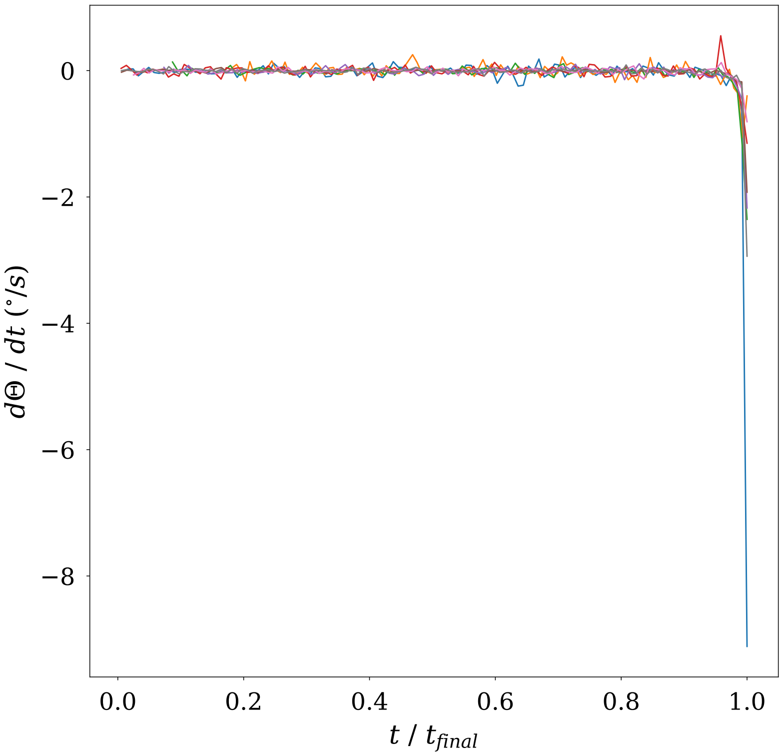

Figure 1: For experiments of pure water droplets or droplets with very low particle concentrations, we show the change in contact angle $\frac{d\Theta}{dt}$ as a function of $\frac{t}{t_{final}}$. The contact angle stays constant throughout the majority of the droplet's lifetime (i.e. $\frac{d\Theta}{dt} \approx 0$), only showing a sudden and steep decline of $\frac{d\Theta}{dt}$ at the final stage of evaporation.

For droplets with very low particle concentrations we observe flattened clusters after the evaporation, probably due to some pinning of the contact line in the end stage of evaporation. To investigate the precise moment of pinning, and whether or not it is related to a critical droplet radius, we plot the change of contact angle as a function of effective radius in Figure 2. The effective radius was calculated based on the measured droplet volume.

Furthermore, we measured the base diameter of the droplet at the pinning moment, for different initial particle packing fractions (see Figure 3). If the particles played any role in the pinning of the droplet, we would expect a trend within the pinning base diameter with in increasing initial particle packing fraction. However, no clear trend was found and we therefore conclude that the particles do not influence the pinning.

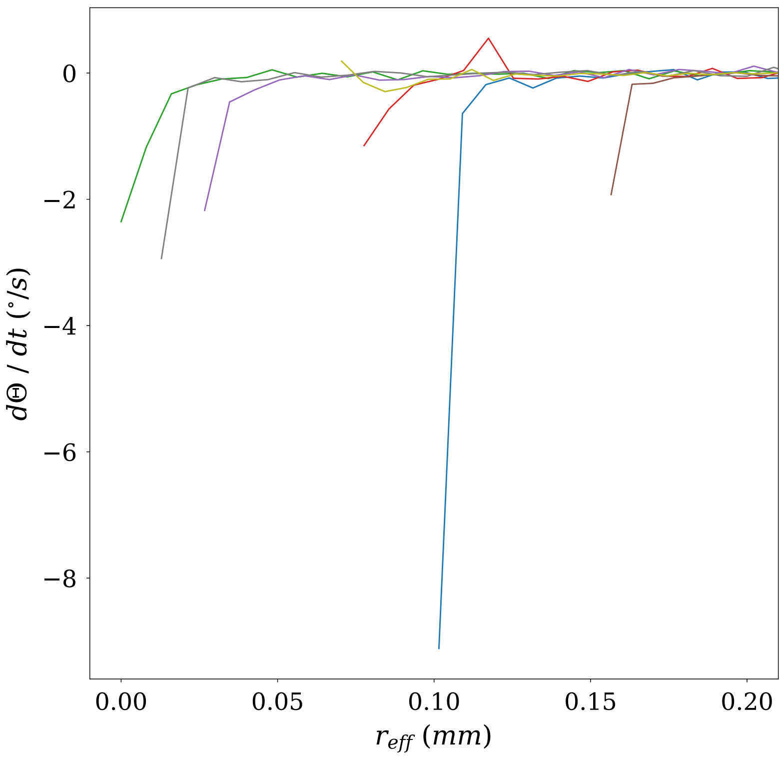

Figure 2: All experiments show a decline in $\frac{d\Theta}{dt}$ at different effective radii. Therefore, we conclude that the pinning, indicated by $\frac{d\Theta}{dt} < 0$, happens at random times and is unrelated to droplet volumes.

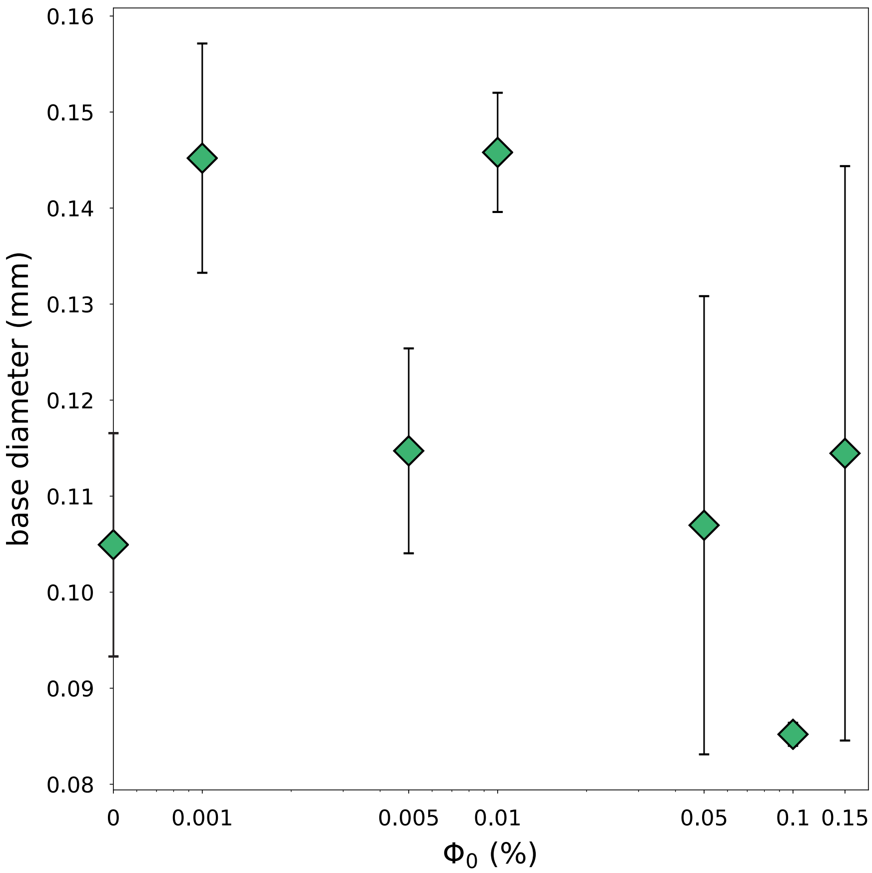

Figure 3: Pinning Base diameter of droplets with different particle loads. Note the logarithmic scaling of the x-axis, with the data point for pure water droplets (0% packing fraction) plotted on top of the y-axis. No clear trend can be distinguished. The markers show the mean base diameter for all experiments conducted at one initial packing fraction and the errorbars represent the standard deviation of the sampled values.

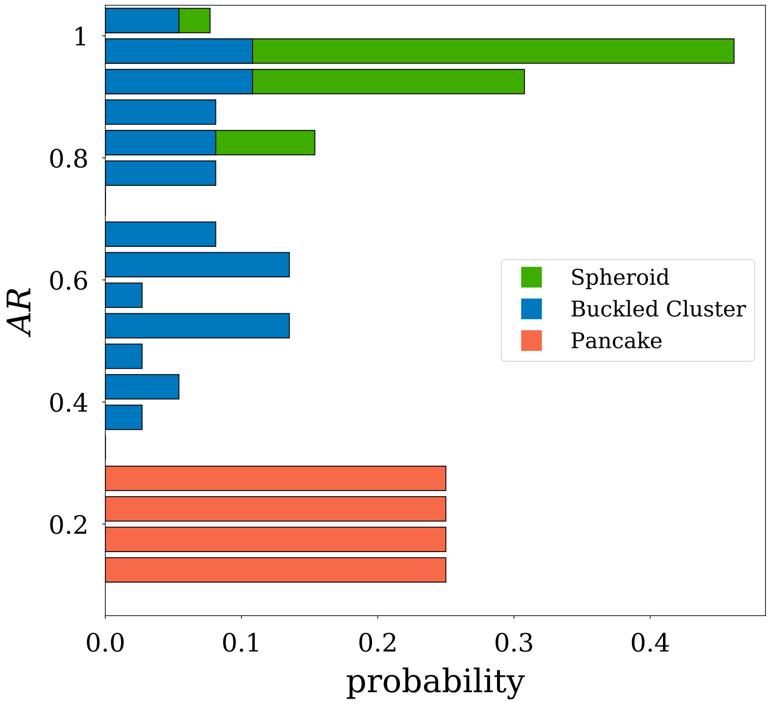

Figure 4: Probability of finding a certain aspect ratio within each category. All probabilities of the three individual distributions add up to one. *Spheroids* are most likely found with high aspect ratios above 0.95, *buckled clusters* with aspect ratios around 0.6, while *pancakes* generally present with low AR.

Aspect Ratio Histogram

In order to give a better statistical overview of the aspect ratio of clusters, we plot histograms of the probability of aspect ratios, for each category, see Figure 4. The three probability distributions are plotted on top of each other. All probabilities of the three individual distributions add up to one. We see a clear trend for *spheroids* to present with high aspect ratios, while *pancakes* feature only low aspect ratios, since they are flattened deposits. The probability distribution of *buckled cluster* is spread the widest, with clusters presenting a wide range of aspect ratios, from below 0.4 up to and even above 1.0. This can be explained with the very different shapes a cluster can buckle to, as well as the calculation of the aspect ratio being based on a side view image alone. The highest probability is found for aspect ratios around 0.6, which is much lower than the most likely aspect ratio for *spheroids*, close to 1.0.