Supplementary Information for:

A mechanistic view of drying suspension droplets

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Information about Supplementary Movies

Movie 1) Light transmission through a drying droplet of 5 mol% crosslinked particles
Full time series of Figure 4a in the main text, 10× accelerated. A packing front propagates inwards, followed by a wrinkling front at t = 8 s. Just before the last bulk water has evaporated, at t = 13 s, a second packing front appears which propagates from the centre outwards. The two packing fronts merge at the transition between the edge and centre of the droplet. At t = 14 s, air invasion fronts advance from the contact line and centre towards the top of the edge, followed by capillary deformation fronts.

Movie 2) Light transmission through a drying droplet of 35 mol% crosslinked particles
Last 12 seconds of the time series of Figure 4b in the main text, 2× decelerated. The inward propagating packing front is accompanied by an outward propagating packing front, similar to Movie 1. When these fronts meet, the droplet shrinks slightly, followed rapidly by the emergence of cracks. Simultaneously, air invades the droplet as percolating fronts. The decrease in intensity upon air invasion is partially compensated by the subsequent capillary deformation, which manifests as fronts advancing from the contact line, centre, and cracks towards the top of the edge.

Movie 3) Light transmission through a drying droplet of 20 mol% crosslinked particles
8 seconds of cracking and crack widening corresponding to Figure 9 in the main text, 3× decelerated. After merging of the inward and outward propagating packing fronts, cracks nucleate, propagate and expand to several times their initial width. Simultaneously, air invades from the contact line and centre inwards, and the cracked fragments partially delaminate from the substrate. Because of the transparency of this droplet, the delamination is visible as propagating optical interference fringes around the still adhering regions, originating from the air cavity between the detached film and the substrate.[1] The crack expansion, air invasion and delamination stop simultaneously.

[1] Giorgiutti-Dauphiné and L. Pauchard, *Soft Matter*, 2015, **11**, 1397–1402

Changes in intensity upon air invasion and capillary deformation in the droplet centre

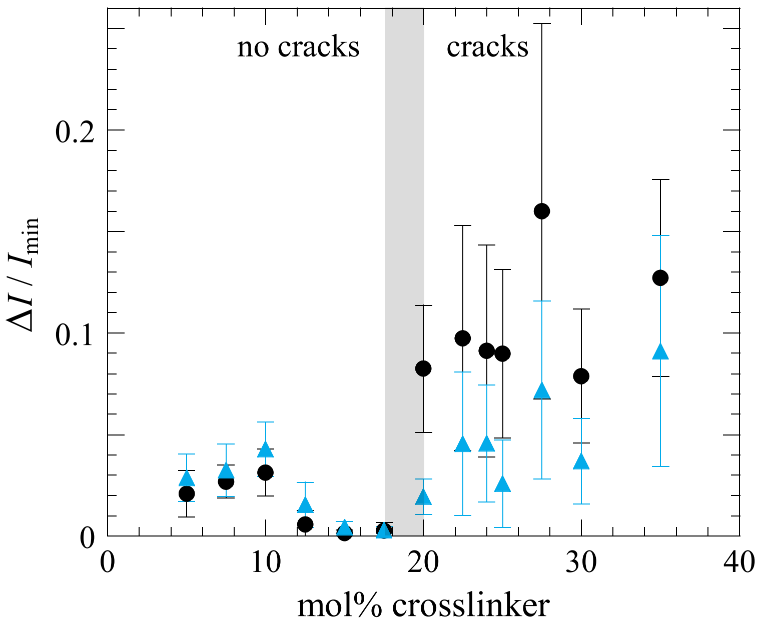

Figure S1: Normalized decrease in transmitted light intensity due to air invasion ($\Delta I_{\mathrm{ai}}/I_{\mathrm{min}}$, ●) and subsequent increase in intensity due to capillary deformation ($\Delta I_{\mathrm{cd}}/I_{\mathrm{min}}$, ▲) in the centre of droplets versus mol% crosslinker, and averaged over three measurements. The error bars represent the standard deviations. The grey vertical bar indicates the transition region between absence and presence of cracks.

Scanning electron micrographs of particle deformation in the centre of dried droplets

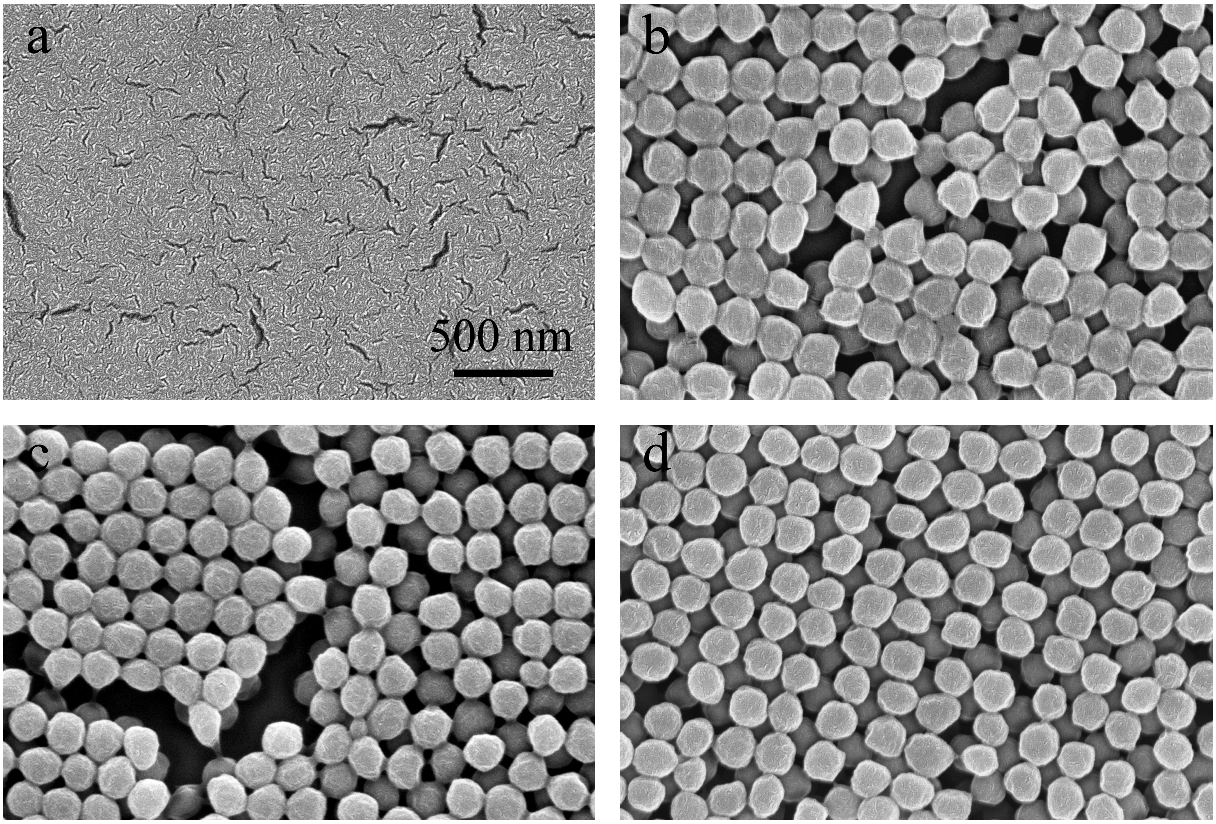

Figure S2: SEM images showing particle deformation in the centre of dried droplets with (a) 5, (b) 20, (c) 22.5, and (d) 30 mol% crosslinker. In (b–d), two layers of particles are visible with the substrate underneath. All images have the same scale. The tiny cracks are artefacts caused by the sputter-coating.