Electronic Supplementary Material (ESI) for Soft Matter.

Electronic Supplementary Information

Harmonic Analysis of Surface Instability Patterns on Colloidal Particles

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Table S1 Numerical values ($\pm$ 95 % confidence interval) of particle parameters: root-mean-square surface roughness σ, initial aerosol droplet radius R_I, buckling transition radius R_B, final mean spherical radius $\langle R_\mathrm{F}\rangle$, spherical harmonic mean sphere radius R_S, final surface area A_F and final volume V_F.

Parameter	C1	C2	W3
σ (nm)	26.1	36.0	29.7
R_I (nm)	498	668	1181
R_B (nm)	140	196	283
$\langle R_\mathrm{F}\rangle$ (nm)	103.7	138.8	235.9
R_S (nm)	92.3	126.2	233.8
A_F (nm^2)	2.456×10^5	4.842×10^5	1.007×10^6
V_F (nm^3)	3.979×10^6	9.624×10^6	5.303×10^7

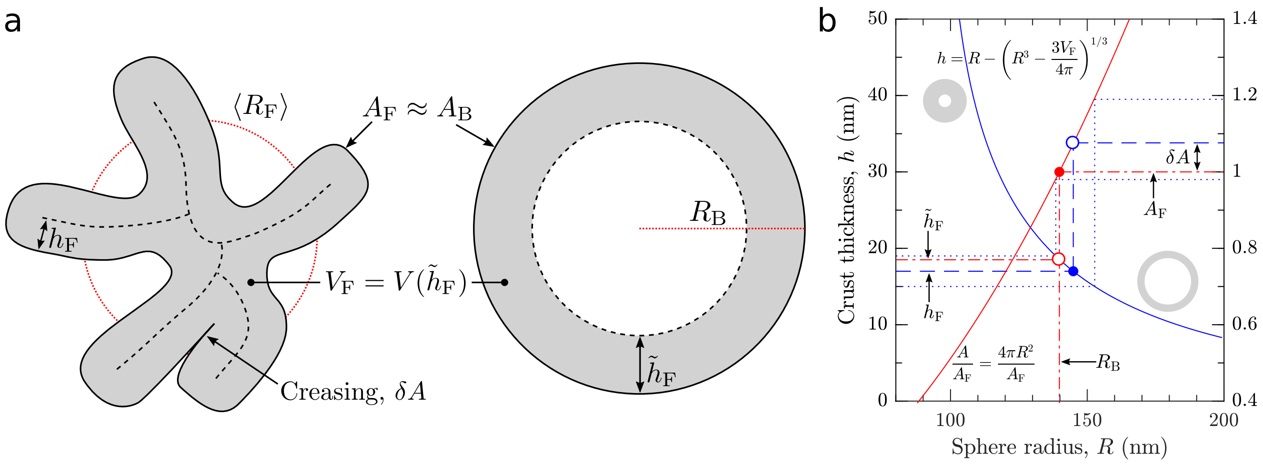

Fig. S1 (a) A schematic illustration of the particle at time $t = t_\mathrm{F}$ (left) and at its pre-buckled state accompanied with different variables used in this study: radius R, mean spherical radius $\langle R_\mathrm{F}\rangle$, crust thickness h, volume V and surface area A. (b) Estimation of the hidden surface area fraction due to creasing (self-contact) for C1. In the left vertical axis, crust thickness as a function of sphere radius needed to reproduce the final particle volume V_F. The two annuli represent schematically the shape transition as R increases, whereby a hollow cavity of larger volume is needed to keep the solid volume constant. Following the blue dashed line, an estimate for $\delta A = 0.07^{+0.12}_{-0.09}$ can be calculated from the measured crust thickness $h_\mathrm{F} = 17$ nm (for C2, $\delta A = 0.01^{+0.09}_{-0.07}$). The blue dotted lines represent the 95 % confidence bounds for the crust thickness measurement ($\pm$ 2 nm). In the right vertical axis, sphere surface area normalized with respect to the final surface area A_F determined from the tomogram. The red dash-dot line shows the buckling transition radius R_B as well as the calculated crust thickness $\tilde{h}_\mathrm{F} = 18.6$ nm when the volume of the crust equals V_F.

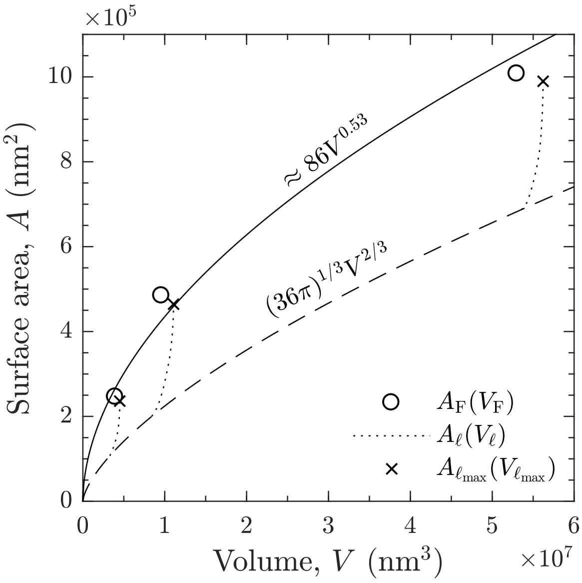

Fig. S2 Surface area A as a function of volume V. The dashed line represents $A(V)$ of a sphere whereas the dotted lines represent the $A(V)$ evolution of the spherical harmonic models as the maximum harmonic degree $\ell_{\max}$ used in the reconstructions is increased from zero to its final value 64 (marked by $\times$). Hollow circles show the final surface area A_{F} as a function of the final volume V_{F} determined from the tomograms, with the solid line showing their power law fit.

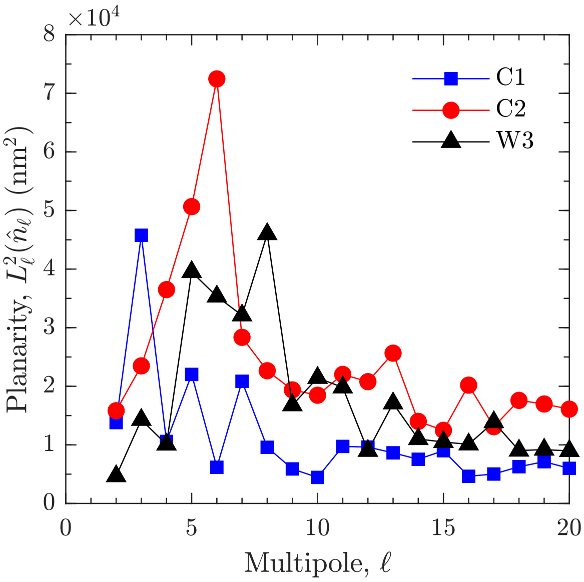

Fig. S3 Value of multipole $r_\ell(\theta, \phi)$ planarity measure $L_\ell^2(\hat{n}_\ell)$ when the multipole is aligned with its preferred axis $\hat{n}_\ell$.