Electronic Supplementary Material (ESI) for Soft Matter.

Supporting Information for

Universality in Buckling Behavior of Drying Suspension Drops.

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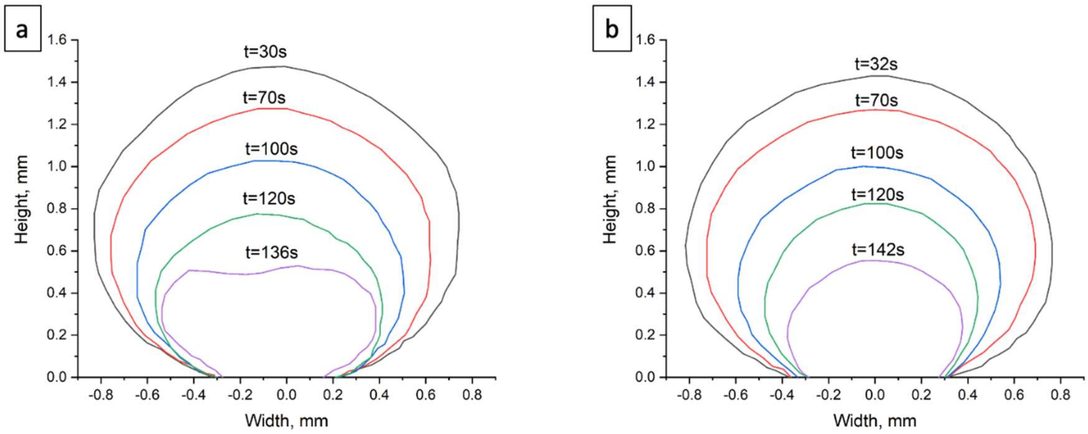

Fig. S1. The figures show the shape evolution of a fast drying sessile drop over a superhydrophobic surface of initial volume (a) 3 µL containing 5 µm PS particles, which buckles, and (b) 3 µL containing 15 µm PS particles, which does not buckle.

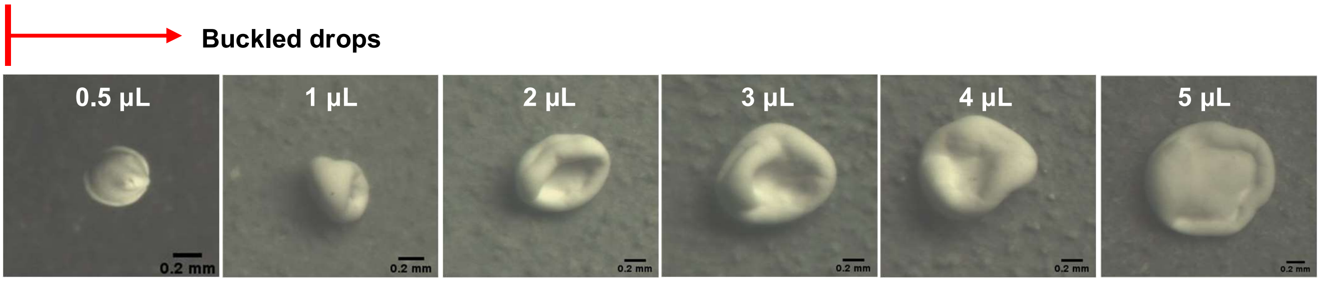

Fig. S2. Dried granules formed from drops with 1 μm silica particles at 5 % (wt/wt) concentration (top view). All drops buckled during drying.

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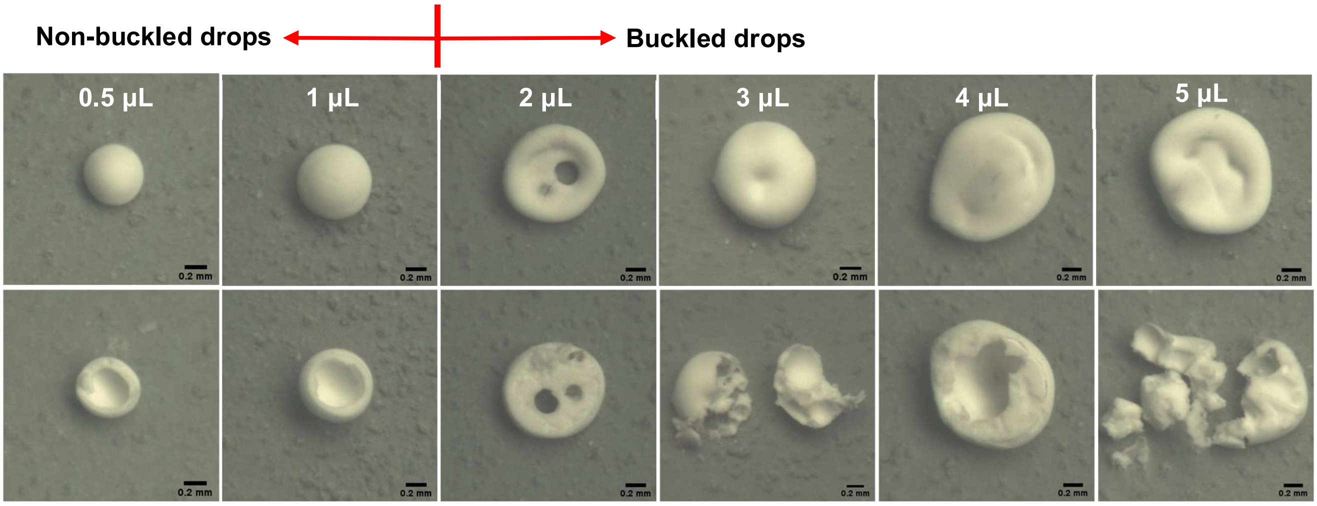

Fig. S3. Dried granules formed from drops with 3 µm silica particles at 5 % (wt/wt) concentration. The first row shows the top view of the dried drop while the bottom row confirms the formation of shell. Drops with initial volumes of 0.5 µL and 1 µL did not buckle while 2 µL to 5 µL drops buckled during drying.

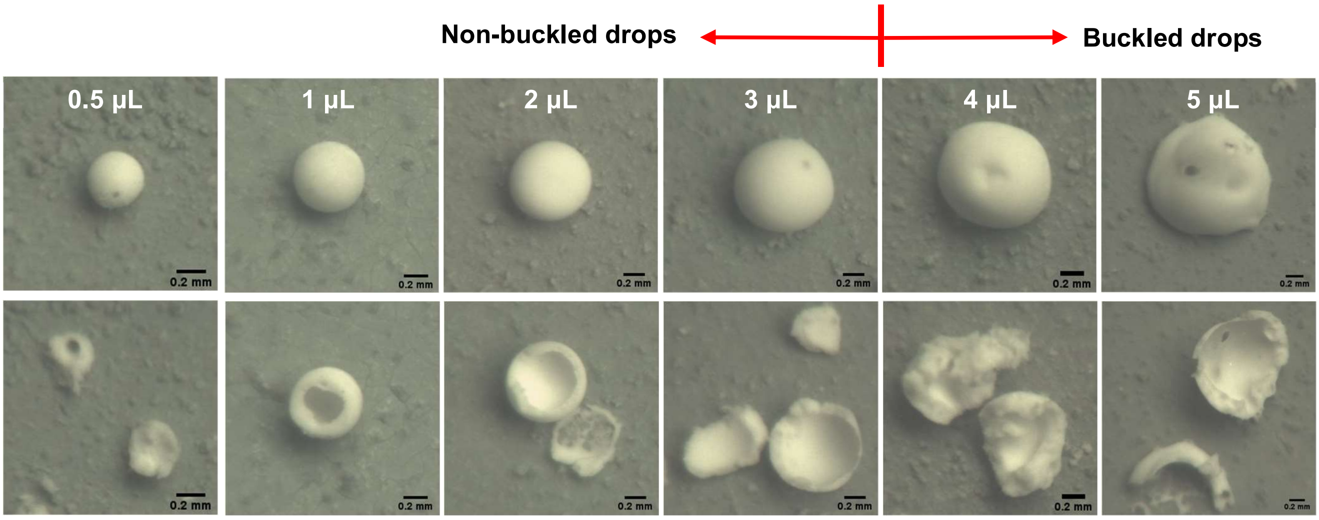

Fig. S4. Dried granules formed from drops with 5 μm silica particles at 5 % (wt/wt) concentration. The first row shows the top view of the dried drop while the bottom row confirms the formation of shell. Drops with initial volume of 0.5 μL to 3 μL did not buckle while the 4 μL and 5 μL drops buckled during drying.

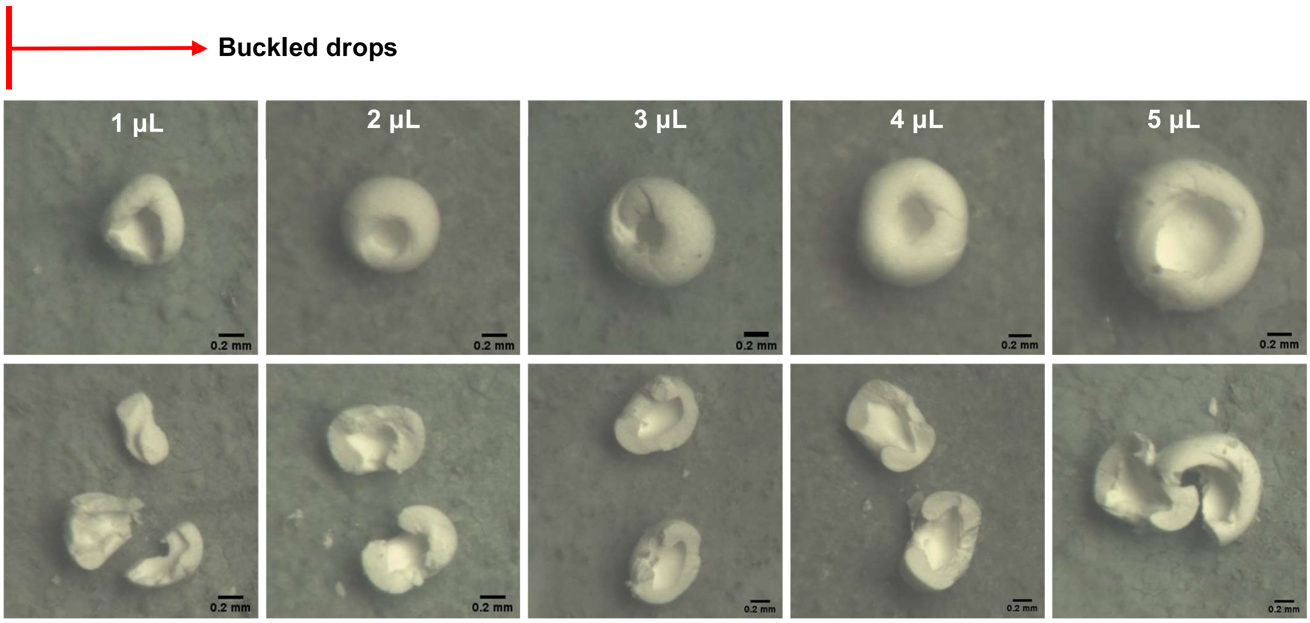

Fig. S5. Dried granules formed from drops with 1 µm PS particles at 5 % (wt/wt) concentration. The first row shows the top view of the dried drop while the bottom row confirms the formation of shell. All drops buckled during drying.

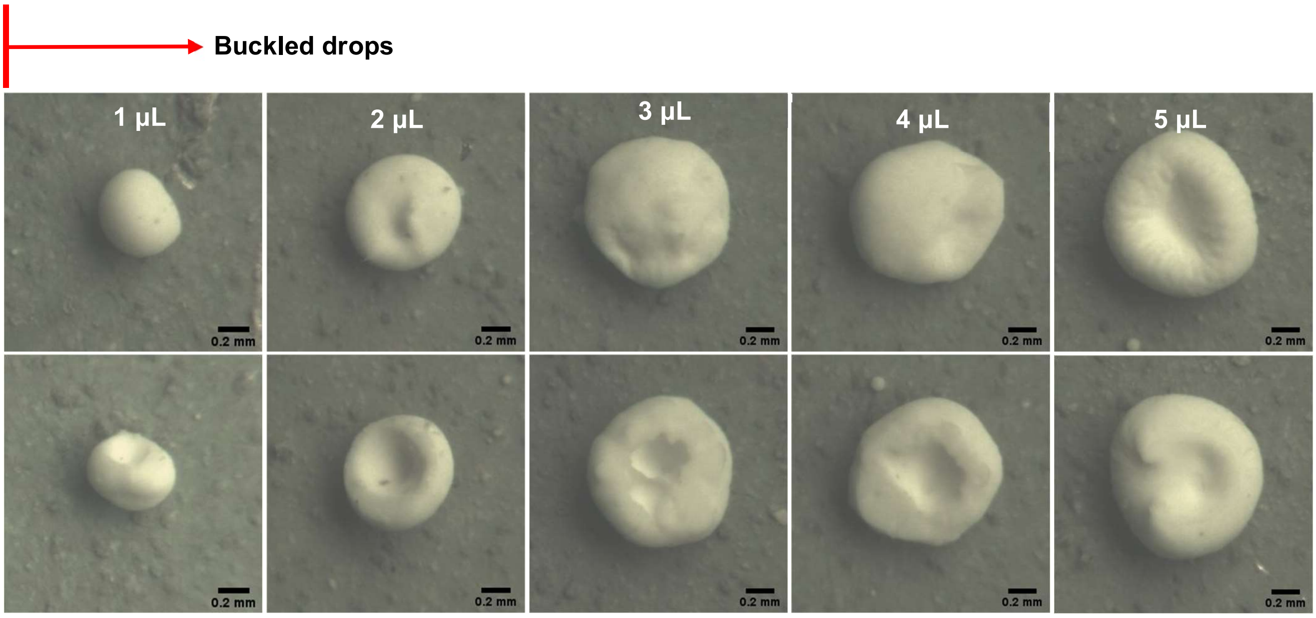

Fig. S6. Dried granules formed from drops with 5 µm PS particles at 5 % (wt/wt) concentration. The first row shows the top view of the dried drop while the bottom row confirms the formation of shell. All drops buckled during drying.

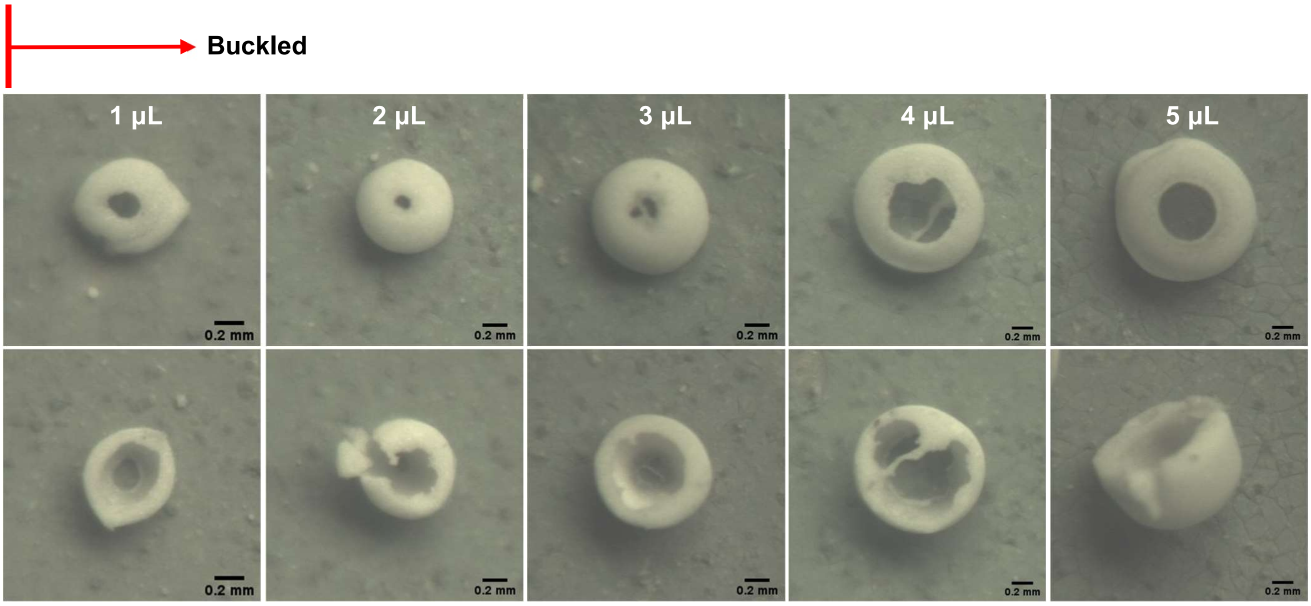

Fig. S7. Dried granules formed from drops with 10 µm PS particles at 5 % (wt/wt) concentration. The first row shows the top view of the dried drop while the bottom row confirms the formation of shell. All drops buckled during drying.

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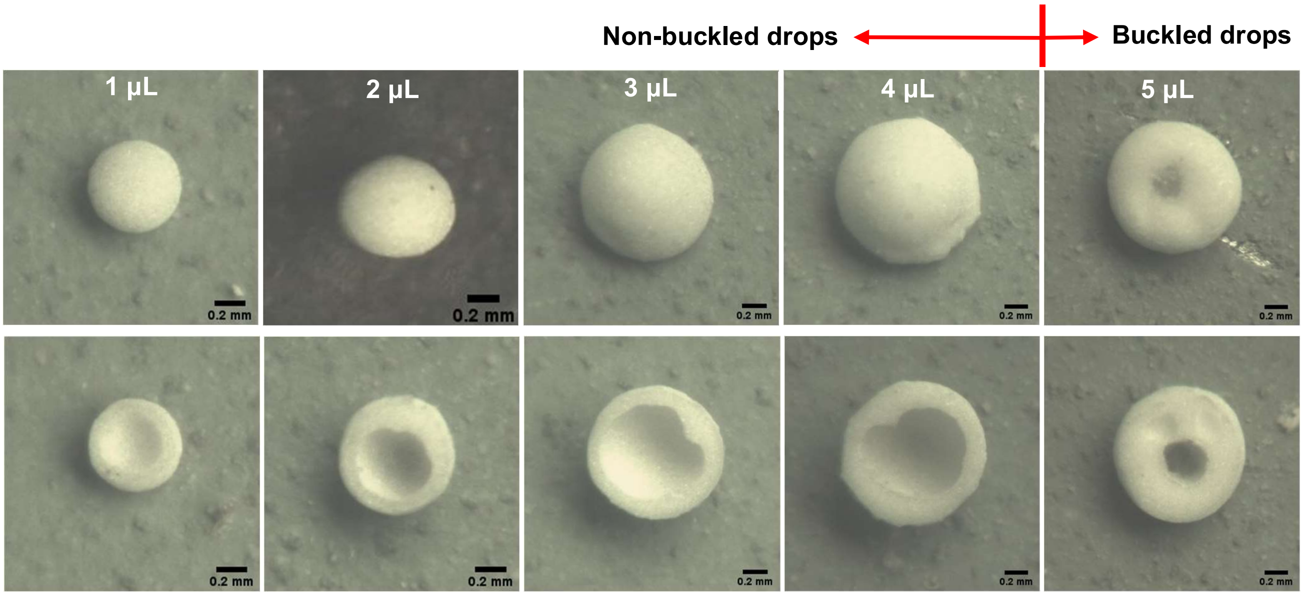


Fig. S8. Dried granules formed from drops with 15 μm PS particles at 5 % (wt/wt) concentration. The first row shows the top view of the dried drop while the bottom row confirms the formation of shell. Drops with initial volume of 1 μL to 4 μL did not buckle while the 5 μL drops buckled during drying.

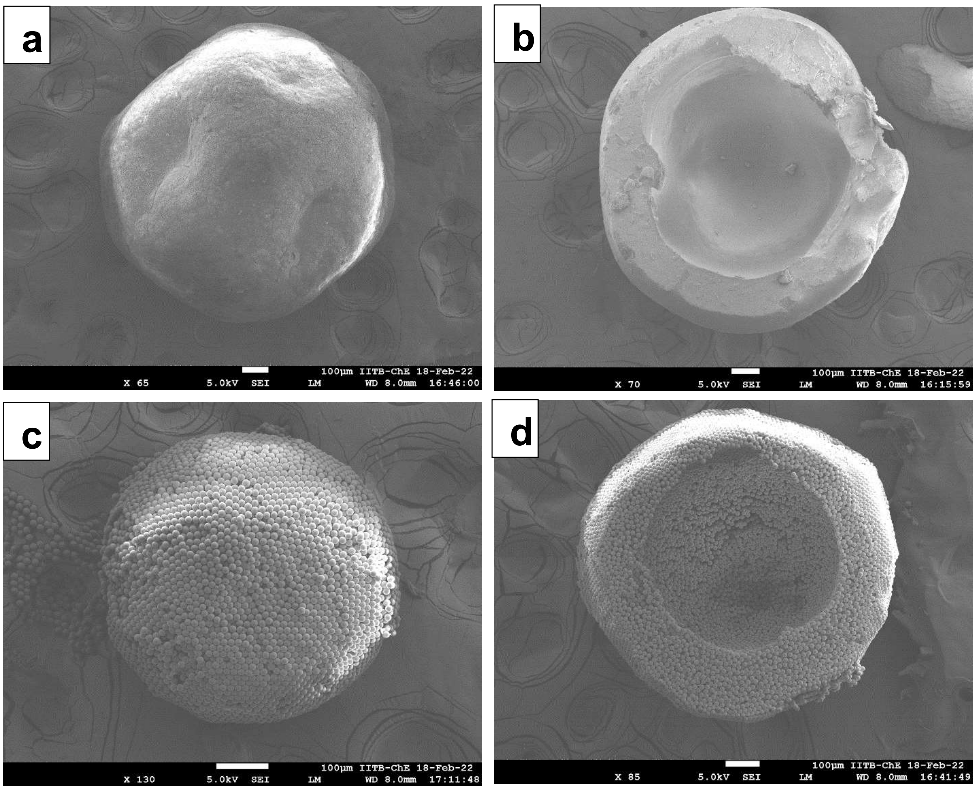


Fig. S9. SEM images of buckled and non-buckled dried granules. (a) Top view of a 1 µm PS buckled shell, (b) Bottom view of a 1 µm PS buckled shell, (c) Top view of a 15 µm PS particles non-buckled shell, (d) Bottom view of a 15 µm PS particles non-buckled shell.

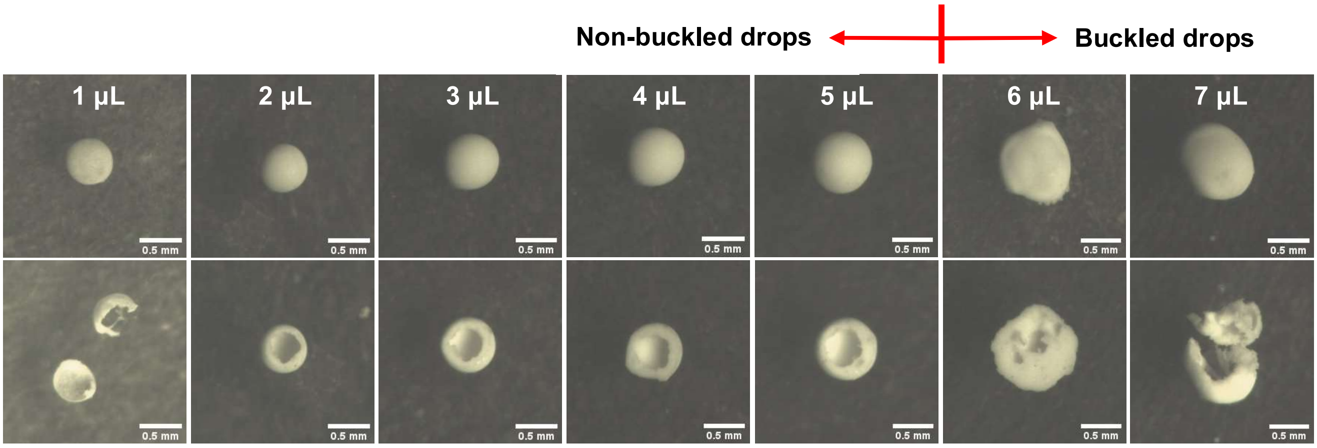


Fig. S10. Dried granules formed from drops with 5 μm silica particles at 2 % (wt/wt) concentration. The first row shows the top view of the dried drop while the bottom row shows the hollow shell. Drops with initial volume of 1 μL to 5 μL did not buckle while the 6 μL and 7 μL drops buckled during drying. Compare this with the 5 % (wt/wt) suspension drop that buckled at 4 μL and 5 μL (Figure S4). For both cases, the critical shell radius (*R*) lies between 0.4-0.5 mm suggesting that the initial concentration of particles does not influence the critical shell radius at buckling.

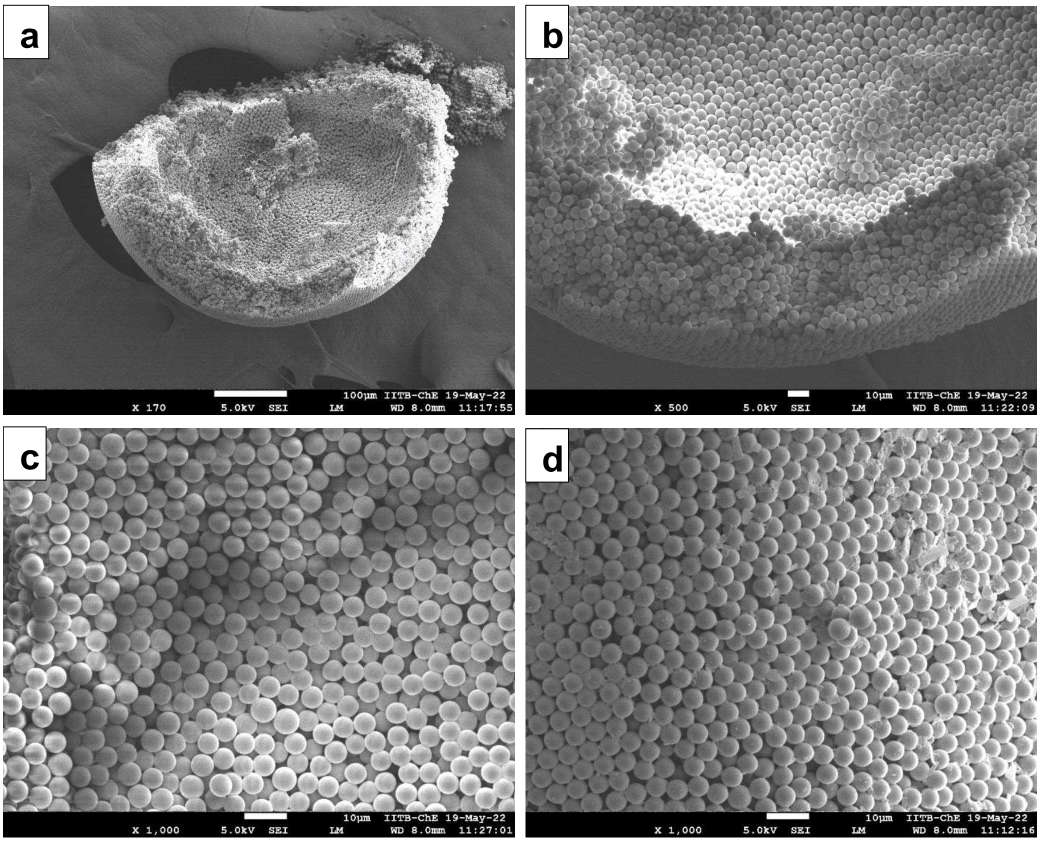


Fig. S11. SEM images of dried granule formed from a mixture of 5 µm silica particles and 5 µm PS particles (ratio of number density of particles in the mixture, silica:PS = 1.6:1). (a) and (b) Images of a section of the shell. (c) and (d) A close-up view of the particles on the outer and inner surface of the shell, respectively. These images show that the particles contained in the droplet did not undergo permanent deformation during drying thereby confirming the assumption of elastic deformation.

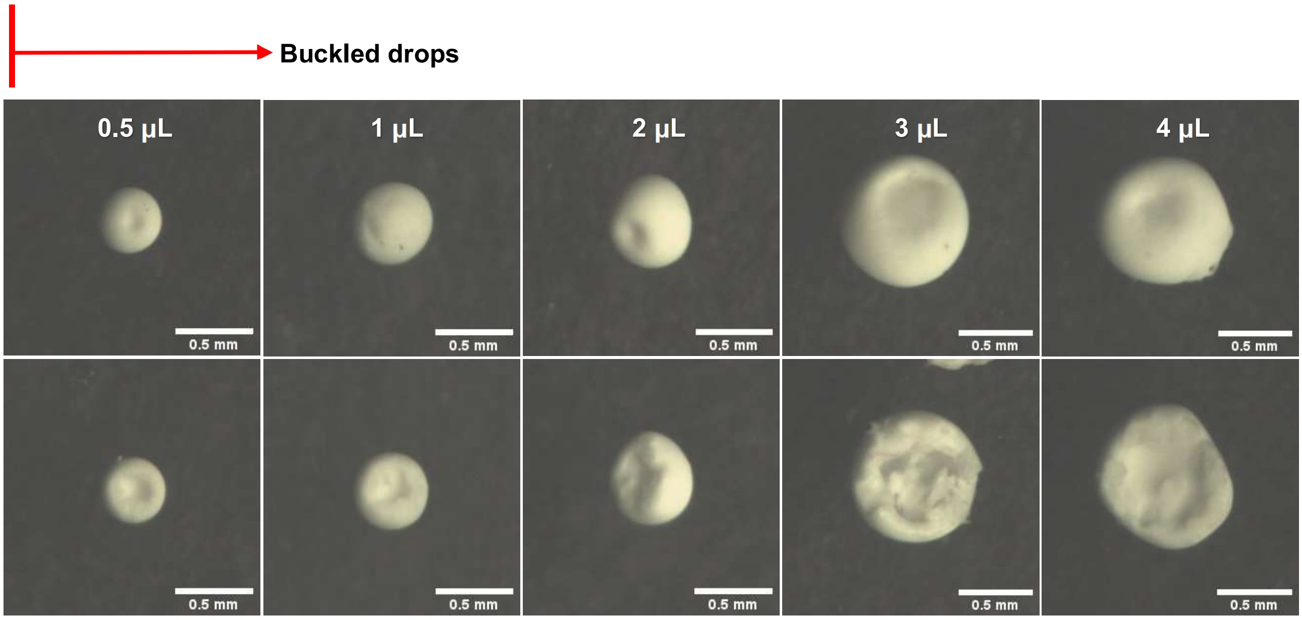


Fig. S12. Dried granules formed from a mixture of 5 µm and 1 µm silica particles. The mixture was prepared by mixing 10 parts of 5 µm suspension with 1 part of 1 µm suspension, each at 5 % (wt/wt) concentration. The corresponding ratio of number density is, 5 µm : 1 µm=1:12.5. The first row shows the top view of the dried drop while the bottom row confirms the formation of shell..

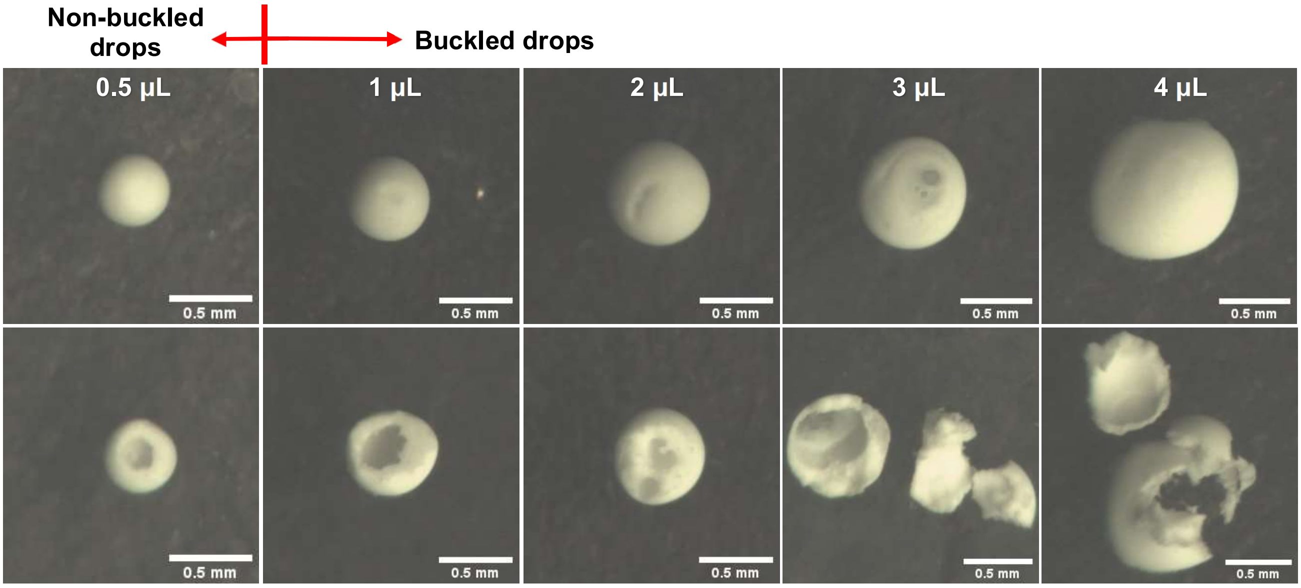


Fig. S13. Dried granules formed from a mixture of 5 μm and 1 μm silica particles. The mixture was prepared by mixing 20 parts of 5 μm suspension with 1 part of 1 μm suspension, each at 5 % (wt/wt) concentration. The corresponding ratio of number density is, 5 μm : 1 μm=1:6.25. The first row shows the top view of the dried drop while the bottom row confirms the formation of shell.

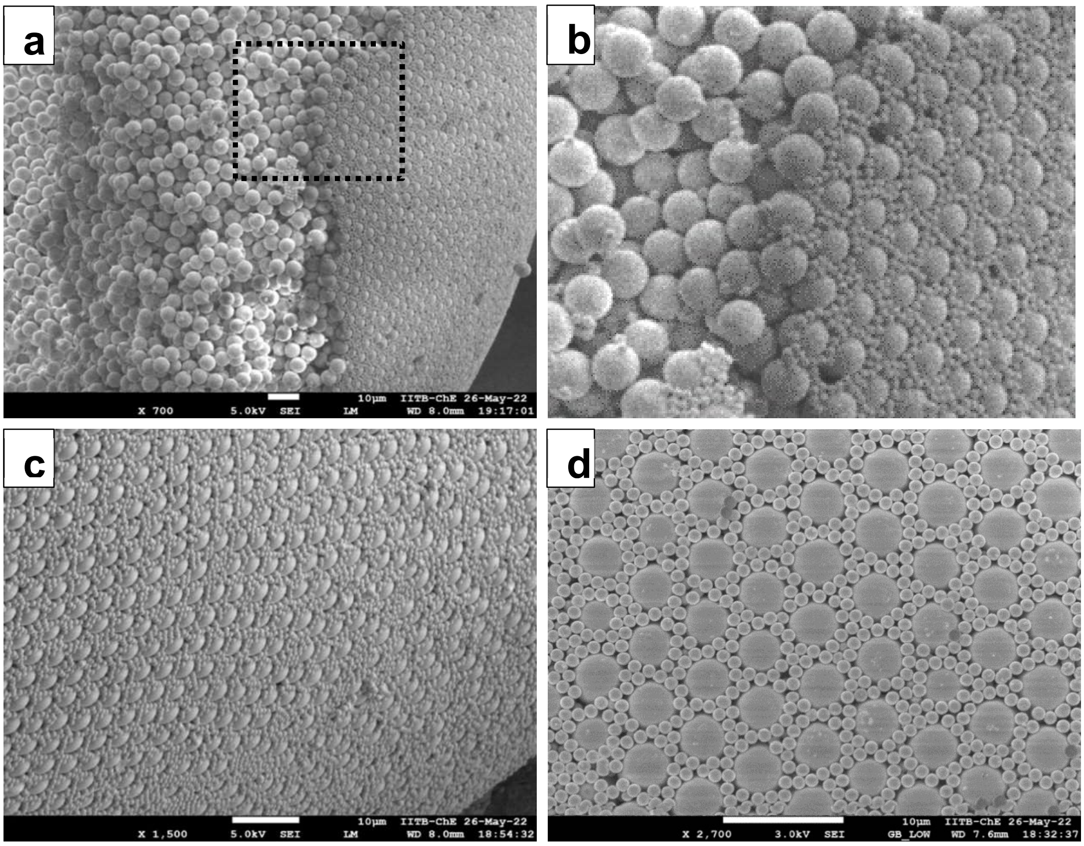
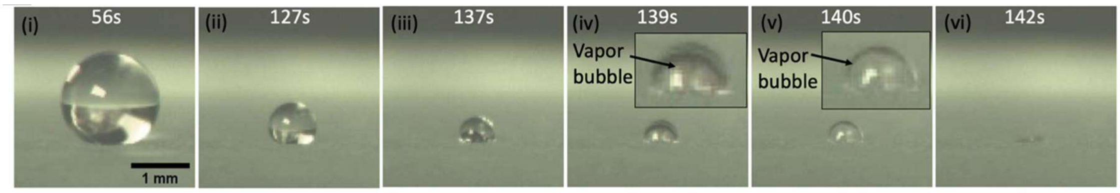


Fig. S14. SEM images of dried granule formed from a mixture of 5 μm and 1 μm silica particles (ratio of number density, 5 μm : 1 μm=1:12.5). (a) Image shows the shell thickness along with the arrangement of particles on the surface and through the thickness. It is clear that all small particles are close to the top surface with negligible numbers present in the interior. (b) A close-up view of the box section of image (a). (c) Ordered packing of large particles on the shell surface interspersed with smaller particles. (d) A close-up view of the shell surface.

(a) 5 µL drop dried at 160 ºC and atmospheric pressure



(b) 3 µL drop dried at 80 ºC and 760 mmHg vacuum

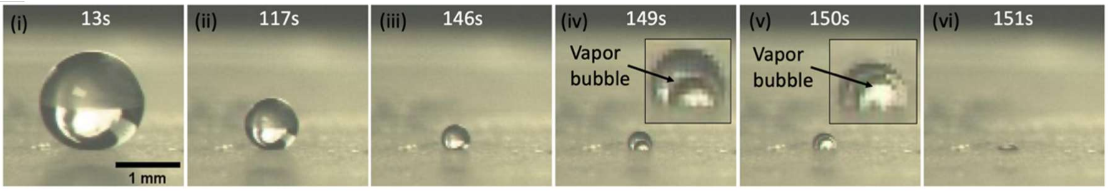


Fig. S15. Images of drying of pure water drop. (a) 5 µL drop dried at 160 ºC and atmospheric pressure. (b) 3 µL drop dried at 80 ºC and 760 mmHg vacuum. For both cases, a vapor bubble nucleates and expands to fill the drop, see the inset in (iv) and (v). The bubble completely evaporates within a few seconds.

Table S1. Physical properties of particles and solvent.

Particle	Supplier	T_g (°C)	Density (g/cc)	Shear modulus (GPa)	Poisson ratio
Silica[1]	Sigma- Aldrich	1200	2.65	31	0.17
PS[2]	Sigma- Aldrich, Cospheric	100	1.05	1	0.34
DI Water	Milli-Q (Millipore)	-	1.0	-	-

Table S2. Product details of all particles used in experiments.

S. No.	Product name	Particle size	Supplier	Product number	Batch number	Product brand	Conc.
1	Micro particles based on silicon dioxide	1 µm	Sigma-Aldrich	56798	BCBV2906	Sigma	5 % WT.
2	Micro particles based on silicon dioxide	3 µm	Sigma-Aldrich	66373	BCBN8195V	Fluka	5 % WT.
3	Micro particles based on silicon dioxide	5 µm	Sigma-Aldrich	44054	BCBT0088	Sigma	5 % WT.
4	Micro particles based on polystyrene	1 µm	Sigma-Aldrich	89904	BCBR6474V	Sigma	10 % WT.
5	Micro particles based on polystyrene	5 µm	Sigma-Aldrich	79633	BCCF6420	Sigma-Aldrich	10 % WT.
6	Micro particles based on polystyrene	10 µm	Sigma-Aldrich	72986	BCBW4523	Sigma-Aldrich	10 % WT.
7	Micro particles based on polystyrene	15 µm	Sigma-Aldrich	74964	BCCG3435	Sigma-Aldrich	10 % WT.
8	Polystyrene microsphere	14-20 µm	Cospheric	PSMS-1.07 14-20um - 500mg	NA	Cospheric	Powder

Table S3. Instruments and manufacturer.

Instrument	**Manufacturer**
Vacuum oven	Pooja Lab, Mumbai, INDIA
Micropipette	Eppendorf Research
USB Microscope camera	Lumenera Infinity-1
Contact angle measurement Goniometer	GBX Digidrop, Software: Windrop
FESEM	JSM 7600F, JEOL Inc.

SI References

1. https://www.azom.com/properties.aspx?ArticleID=1114. (Accessed on Oct 27, 2022)
2. Mott, P.H., Dorgan, J.R. and Roland, C.M., 2008. The bulk modulus and Poisson's ratio of "incompressible" materials. Journal of Sound and Vibration, 312(4-5), pp.572-575.