Heterogeneous structure of granular aggregates with capillary interactions
Supplementary Information

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In this supplement, we provide measurements giving evidence that the system of floating particles is homogeneously compressed in Section 1, discuss the shape heterogeneity of Voronoi cells using a shape factor in Section 2, and then present further pores size analysis in Section 3.

1 Homogeneous compression

Density of particles is increased because of the uniformly convergent flow generated inside the conical funnel shaped container as the level of the liquid inside is reduced. From the measured positions of the particles, we obtain the mean number of the particles per unit area ρ_n as a function of radial distance from the center axis of the funnel. Figure 1 shows ρ_n during three stages of compression for $N = 688$ and $N = 1139$. The plots show that the mean density remains flat as the system is compressed in both examples. In particular, there is no preferential increase of density at the periphery which would result if compression was applied only at the boundaries. Further, the length scale over which ρ_n decreases to zero is more or less constant and occurs because of the repulsive boundary conditions.

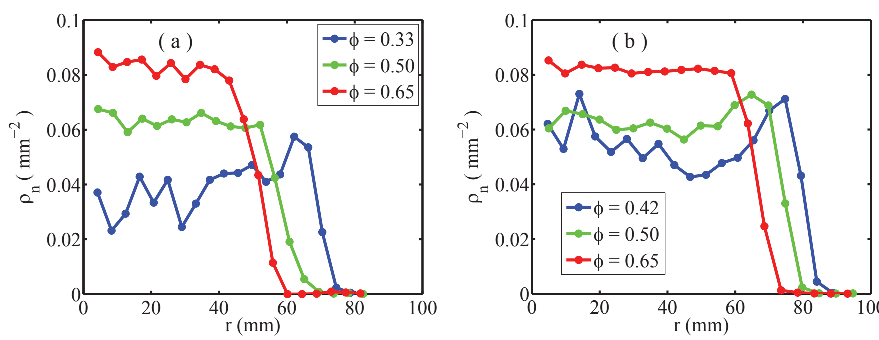

Figure 1: The mean number of particles per unit area as a function of distance from the axis of the conical container at three stages of the compression. Number of particles $N = 688$ (a) and $N = 1139$ (b).

2 Distribution of the shape factor of the Voronoi cells

A measure to characterize heterogeneity of two dimensional system of particles is given by the shape factor of the Voronoi cells [1]:

$$\xi = \frac{\mathcal{C}^2}{4\pi A_v},$$

where, $\mathcal{C}$ is the cell perimeter and A_v is the cell area. This parameter has its minimum value $\xi_c = 1$ for circles, increases slightly for regular polygons ($\xi_s = 1.273$ for squares and $\xi_h = 1.103$ for regular hexagons), and diverges for a needle-like cell. We first plot the distribution of ξ observed for various ϕ in the Fig. 2. As ϕ increases, the initial broad distribution narrows with ϕ, with values approaching that for a regular hexagon. Therefore, to obtain a measure of deviations from hexagonal order, we plot $(\langle\xi\rangle - \xi_h)/\xi_h$ as a function of ϕ in the Inset to Fig. 2. A decrease is observed indicating that cells become more regular shaped during compression and the heterogeneity decreases, but the value does not reach zero showing that a crystalline state is not reached as the system reaches the maximum density before buckling.

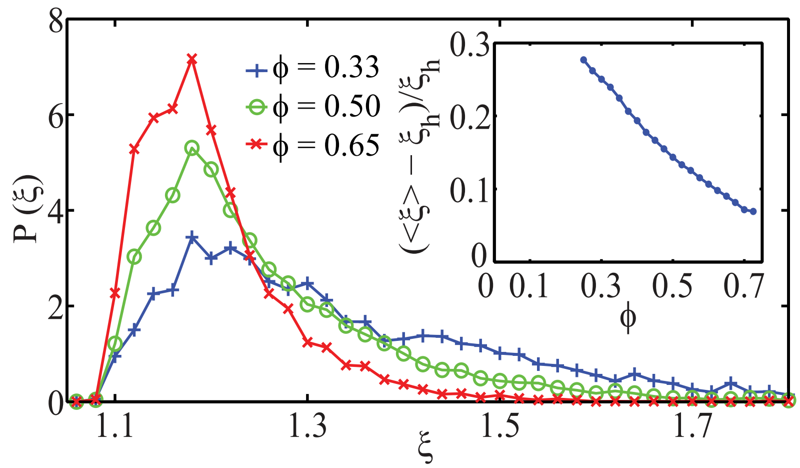

Figure 2: PDF of the shape factor ξ for $\phi = 0.33, 0.50$ and 0.65. Inset: Evolution of the mean shape factor rescaled by the value for a hexagonal cell ξ_h versus ϕ.

3 Pore size distribution

A direct measure of distribution of voids spaces or pores present in a heterogeneous system is given by the *pore-size probability density function* $P(\delta)$, where δ is the distance of a random point in the pore from the nearest point on the pore-solid interface [2]. $P(\delta)$ plotted in Fig. 3(a) shows a wide distribution of δ which becomes narrower as ϕ is increased reflecting the changes in the Voronoi free area PDF (Fig. 2 in the article) because of the decrease of void spaces. The corresponding average value of $\langle\delta\rangle$ computed from the first moment of $P(\delta)$ is shown in Fig. 3(b). For regular pores, this size represents the average radius of the largest circle which can be inserted inside a pore. The average pore size decreases smoothly with ϕ, but remains above the value for a hexagonal lattice ($\delta_c \sim 0.023d$). Finally, although the property of the pores captured by the Voronoi areas and $P(\delta)$ are complementary, it remains difficult to establish a quantitative connection due to the irregularity and anisotropy of void spaces shapes as illustrated in Section 2.

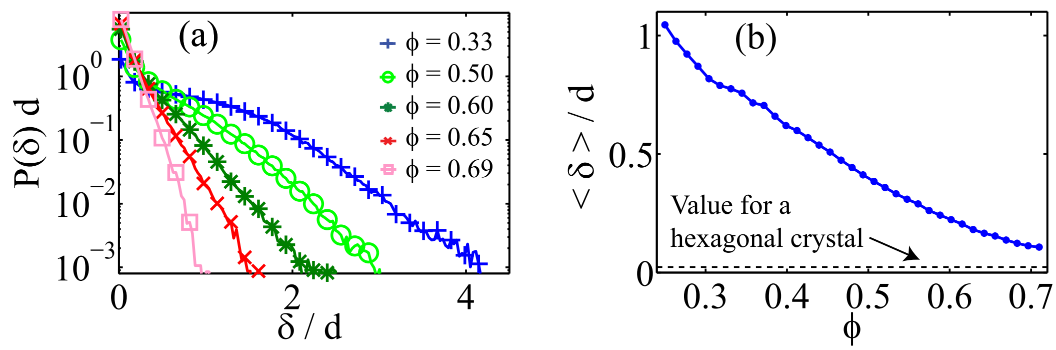

Figure 3: (a) Pore-size probability density function over a range of ϕ. The distributions narrow as large pores are filled in upon compression. (b) The average pore size $\langle\delta\rangle$ decreases as a function of ϕ.

References

[1] F. Moučka and I. Nezbeda, Phys. Rev. Lett.**94**, 040601 (2005).

[2] S. Torquato, in *Random Heterogeneous Material* (Springer-Verlag New York, 2002) pp. 48-57.