Two-phase densification of cohesive granular aggregates*

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Abstract. When poured into a container, cohesive granular materials form low-density, open granular aggregates. If pressed upon with a ram, these aggregates densify by particle rearrangement. In this paper we introduce experimental evidence to the effect that particle rearrangement is a spatially heterogeneous phenomenon that occurs in the form of a phase transformation between two configurational phases of the granular aggregate. We then show that the energy landscape associated with particle rearrangement is consistent with our interpretation of the experimental results. Our conclusions are relevant to many engineering processes and natural phenomena.

Cohesive granular materials have been the focus of only a small fraction of recent research into the granular state [1]. Yet cohesive granular materials will surely draw increasing attention from scientists and engineers, if only because they are used in numerous applications. Conspicuous examples are the forming of ceramic parts, pulvimetallurgic components and pharmaceutical tablets by compaction of fine powders [2]. The cohesiveness of powders stems from the large surface/volume ratio of their constitutive particles, which enhances the effect of attractive van der Walls forces. In other applications, e.g. the stabilization of soils, the cohesiveness is due to the presence of liquid menisci among the particles. Our interest in the densification of cohesive granular materials was prompted by the recent compaction study of Kong and Lannutti [3]. These authors used X-ray tomography to document the evolution of density during the static compaction of alumina powders (particle diameter $\sim 60\,\mu$m). By *static* compaction we mean that compaction was effected by applying pressure, without shaking. (There exists an important body of work on compaction by shaking, usually of noncohesive granular materials [4].) Kong and Lannutti reached the tantalizing conclusion that densification "seems to proceed as a wave initiated at the advancing ram" [3]. Our aim here is to eluci-

*arXiv:cond-mat/0105020, 2 May 2001

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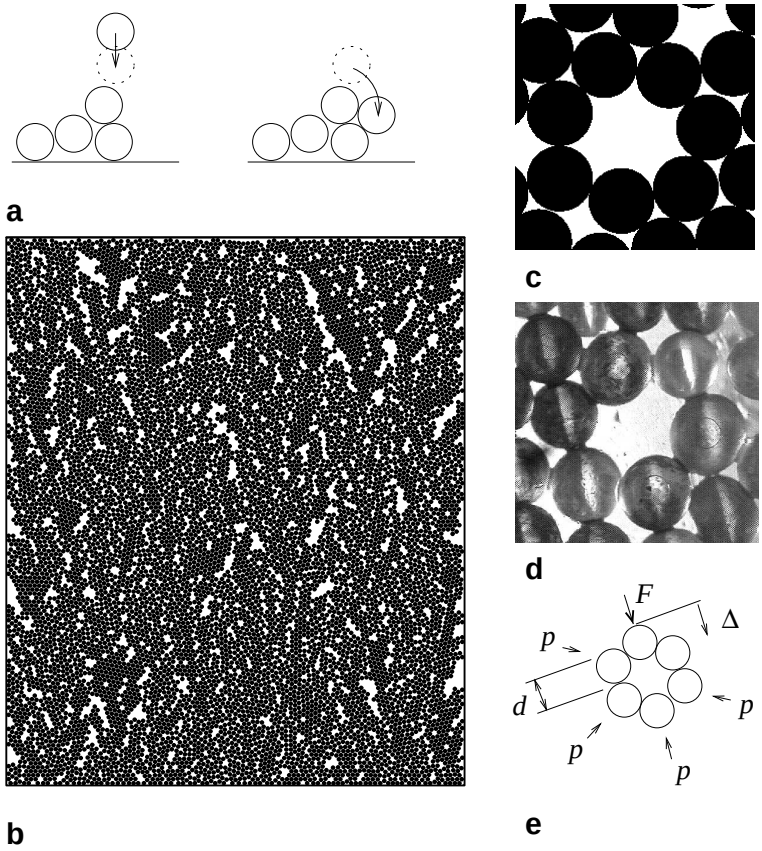

Figure 1: **a)** Computer simulation of container filling by a ballistic aggregation method [10]. The particles are sequentially deposited along random vertical paths, and then allowed to roll down until they make contact with any two points (at least) on the surface of the growing aggregate. The magnitude of the cohesive forces is assumed strong enough to stabilize a particle both at the time of aggregation and afterwards, but it remains otherwise unspecified. **b)** Obtained particle aggregate. **c)** More or less regular rings of particles are a pervasive feature of the simulated aggregate, and also of **d)** aggregates obtained in quasi two-dimensional container-filling experiments. Note the water menisci among the particles. **e)** Collapse of a ring of particles by snap-through buckling: one of the particles jumps to the center of the ring. The driving force F is a relatively large contact force localized on the jumping particle; both photo-elastic and numerical studies indicate that such forces exist with magnitudes several times larger than the average contact force [6,11], which is represented by the hydrostatic pressure p [6].

date the nature of this 'wave,' and to relate its behavior to the micromechanics of densification in cohesive granular aggregates.

When, preceding compaction, a cohesive granular material is poured into a container, the mobility of the particles reaching the bottom of the container is hindered by the cohesive forces, Fig. 1a. As a result, a low-density, open aggregate of particles obtains inside the container, Fig. 1b. Open aggregates densify by particle rearrangement at relatively low pressure [5]. It has been proposed [6] that particle rearrangement occurs when the *rings of particles* of the open aggregate collapse by snap-through buckling, see Fig. 1c-e. To investigate this phenomenon we prepared a quasi two-dimensional open aggregate [7] by filling a narrow Plexiglas container (thickness $\sim 1.9\,\mathrm{mm}$) with monosized glass beads (diameter $\sim 1.7\,\mathrm{mm}$). Before pouring them into the container, we wetted the beads with water in order for menisci to form among the beads, Fig. 1d. These menisci provided the required cohesion. We then compacted the aggregate using a ram. Fig. 2a shows three stages during the experiment. A high-density region (the phase H, wherein rearrangement has taken place already) and a low-density region (the phase L, wherein the open aggregate remains unchanged) are clearly discernible. Densification proceeds by growth of the volume fraction of H at the expense of the volume fraction of L. Visual inspection revealed that no rearrangement occurs within the high- or low-density regions. In a narrow vicinity of the H-L interface or *rearrangement front*, on the other hand, we could clearly observe the collapse by snap-through buckling of successive layers of rings of particles, see Fig. 2b. The rearrangement front, which we identify with the densification wave of Kong and Lannutti, advances in the form of broad, very shallow tongues (visible in Fig. 2a) darting forward in coincidence with the collapse of ring of particles. These results suggest that densification occurs in the form of a phase transformation L$\rightarrow$H, Fig. 3a-b.

To substantiate our interpretation of the experimental evidence we turn to the micromechanics of particle rearrangement. Consider a ring of particles undergoing snap-through buckling, Fig. 1e. For an increasing displacement Δ, Fig. 3c, the force F vanishes as the particle snaps into the void, and again when the particle reaches the center of the void. Fig. 3d shows the attendant evolution of internal energy. The energy associated with the initial (tangent) response of the ring, W_t, is represented by a dashed curve; the relaxation effected by buckling, W_b, is represented by an arrow. W_b causes the function $W(\Delta)$ to be nonconvex. Nonconvex energy functions are characteristic of systems which undergo phase transformations [8].

To move on to the macroscopic scale, we consider an open granular aggregate contained in a *frictionless* container of volume V and constant cross-sectional area. We study the energetics of densification in the space of the local stretch, λ. (The local stretch is defined by $\lambda = \rho_0/\rho$, where ρ is the local density and ρ_0 is the

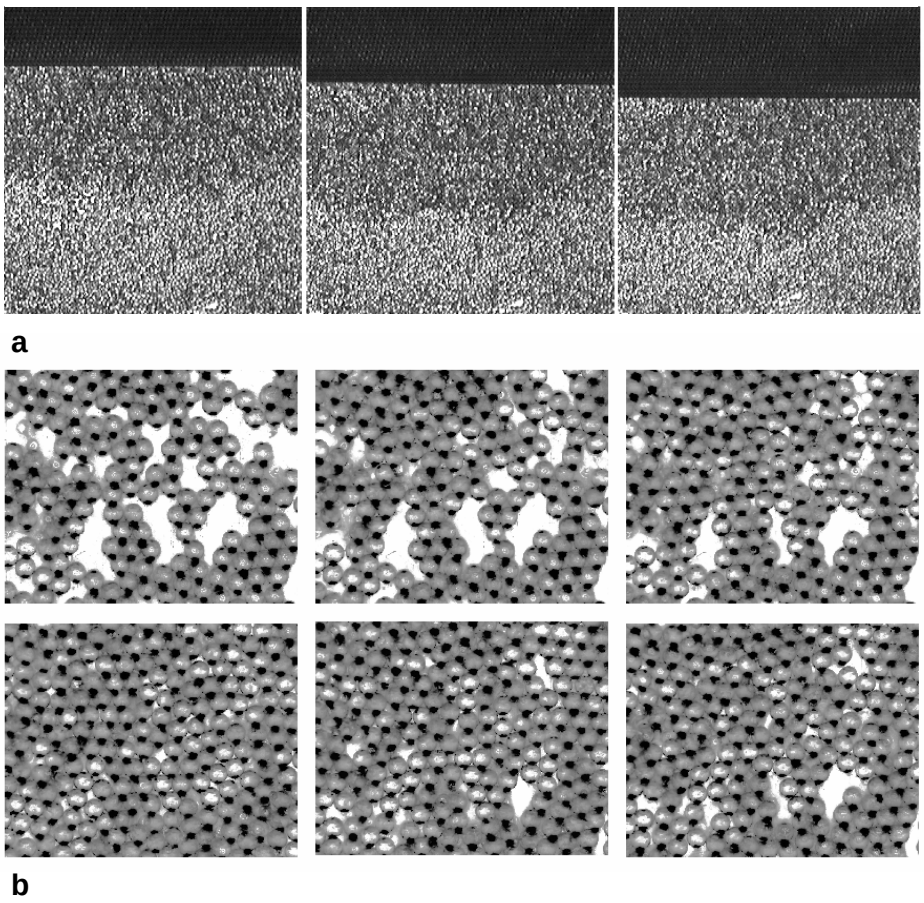

Figure 2: **a)** Three successive times during densification. The ram is black, and moves from the top down. The high- and low-density regions are dark and light gray, respectively; they are separated by the rearrangement front. The area shown is $\sim 25 \times 25\,\mathrm{cm}^2$. **b)** Clockwise starting from the top left: successive collapse of rings of particles in a vicinity of the rearrangement front. The particle diameter is $\sim 1.7\,\mathrm{mm}$. The thickness of the container is $\sim 1.9\,\mathrm{mm}$ (perpendicular to the page).

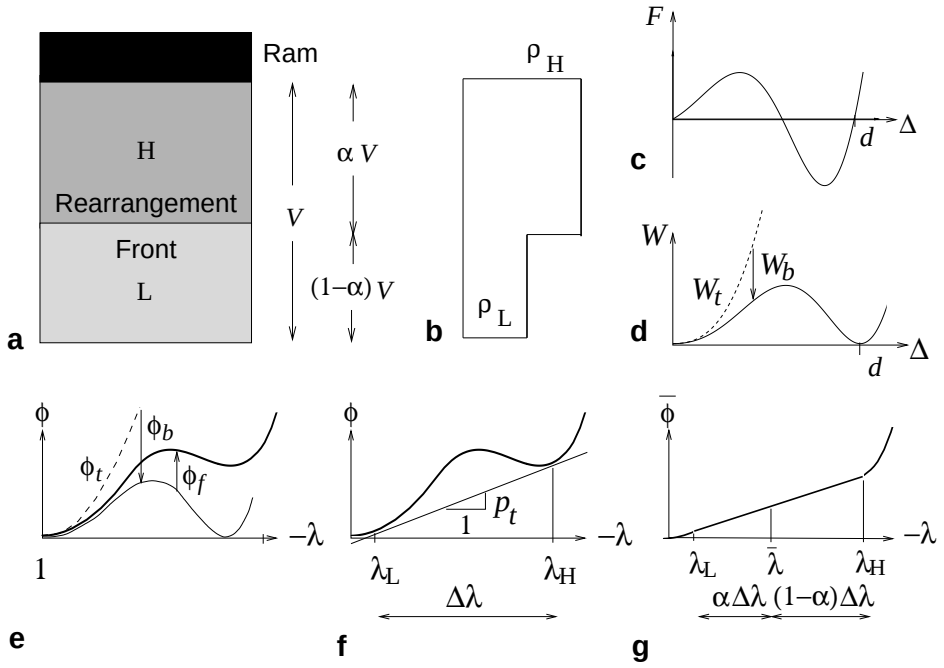

Figure 3: **a)** Interpretation of the experimental results: V is the total volume of the aggregate; the phases H and L are separated by the rearrangement front; α is the volume fraction of H. As α increases from 0 to 1, the rearrangement front sweeps through the aggregate from ram to bottom. Cf. Fig. 2a. **b)** The density jumps by $\Delta\rho = \rho_{\mathrm{H}} - \rho_{\mathrm{L}}$ across the rearrangement front. **c)** Mechanical response of the ring of particles of Fig. 1e: force F vs. displacement Δ (schematic). d is the particle diameter. **d)** Internal energy W vs. Δ. **e)** We write the energy per unit volume of aggregate as $\phi = \phi_t + \phi_b + \phi_f$, where ϕ_t and ϕ_b correspond to the terms W_t and W_b of (d) averaged over a statistically representative volume of aggregate, and ϕ_f is the energy per unit volume dissipated by the inter-particle frictional forces. **f)** Geometrical interpretation of the equilibrium equations, Eq. 1. **g)** The average energy per unit volume, $\bar{\phi}(\bar{\lambda})$.

density of the initial open aggregate.) The energy per unit volume is $\phi(\lambda)$, Fig. 3e; it is nonconvex by inheritance from $W(\Delta)$, Fig. 3d. The local pressure is given by $p = -d\phi(\lambda)/d\lambda$. We now set the average stretch to a given value, $\bar{\lambda} = \frac{1}{V}\int_V \lambda dV$, and minimize the total energy of the aggregate, $\int_V \phi(\lambda) dV$, using conventional tools of nonconvex analysis [8]. This leads to the following equilibrium equations

$$p_t = -\frac{\phi(\lambda_{\rm L}) - \phi(\lambda_{\rm H})}{\lambda_{\rm L} - \lambda_{\rm H}} = -\frac{d\phi}{d\lambda}(\lambda_{\rm L}) = -\frac{d\phi}{d\lambda}(\lambda_{\rm H}), \tag{1}$$

which allow for the computation of the *characteristic stretches*, $\lambda_{\rm L}$ and $\lambda_{\rm H}$, and the *transformation pressure*, p_t, Fig. 3f. It is apparent from Eq. 1 that $\lambda_{\rm L}$, $\lambda_{\rm H}$ and p_t are independent of $\bar{\lambda}$, and can be construed as properties of the initial open aggregate. The characteristic stretches define two configurational phases, L and H, of density $\rho_{\rm L} = \rho_0/\lambda_{\rm L}$ and $\rho_{\rm H} = \rho_0/\lambda_{\rm H}$, respectively. When $\lambda_{\rm L} > \bar{\lambda} > \lambda_{\rm H}$, the phase H occupies a volume αV, and the phase L a volume $(1-\alpha)V$, where $\alpha = (\lambda_{\rm L} - \bar{\lambda})/(\lambda_{\rm L} - \lambda_{\rm H})$, Fig. 3a. As α increases from 0 to 1 during densification, $\bar{\lambda}$ decreases from $\lambda_{\rm L}$ to $\lambda_{\rm H}$, the average density increases from $\rho_{\rm L}$ to $\rho_{\rm H}$, and the pressure remains spatially uniform and equal to p_t. We note that $p_t \neq 0$ on account of the dissipative effects of inter-particle friction, Fig. 3e-f. The average energy per unit volume is given by the convexified form $\bar{\phi} = \alpha\phi(\lambda_{\rm H}) + (1-\alpha)\phi(\lambda_{\rm L})$, which for $\lambda_{\rm L} > \bar{\lambda} > \lambda_{\rm H}$ yields the correct value $p = -d\bar{\phi}(\bar{\lambda})/d\bar{\lambda} = p_t$.

The spatial distribution of phases is not given by our analysis. Nuclei of H could form in many places throughout the aggregate, leading to stratified mixtures of H and L [9]. However, there are no such mixtures in Fig. 2a. We ascribe this fact to the regularizing effect of container-wall roughness. Because of wall roughness, the pressure is no longer spatially uniform; instead, it decreases monotonically away from the ram. The applied pressure may reach values much larger than p_t (it remains scaled by p_t, however). Due to the high pressure, rearrangement and particle deformations are simultaneously active, with a marked effect on the mechanical response of the granular aggregate. Under these conditions, mucleation must invariably occur at the ram, leading to the establishment of a single H-L interface—the rearrangement front. We shall further explore this and other effects of wall roughness in a separate publication.

We conclude that if, as indicated by our experimental observations, snap-through buckling is the dominant mechanism of particle rearrangement, then densification must occur in the form of a phase transformation L→H. Container-wall roughness accounts for the presence of a single H-L interface. It follows that our interpretation of the experimental evidence is consistent with the micromechanics of particle rearrangement.

Supported by a grant from the International Fine Particle Research Institute. We thank Mr. Tomás Uribe for his help with the experimental work.

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