INSTABILITY REVEALS CLUSTERING IN COHESIVE GRANULAR MATTER

LA INESTABILIDAD REVELA "CLUSTERING" EN MATERIA GRANULAR COHESIVA

J.-C. Géminard

Laboratoire de Physique de l'Ecole Normale Supérieure de Lyon, CNRS, Université de Lyon, Lyon, France,
jean-christophe.geminard@ens-lyon.fr

We lately reported on a periodic pattern which spontaneously forms at the surface of a thin layer of a cohesive granular material submitted to in-plane stretching [1] or to bending [2]. The mechanism responsible for the instability is the *strain softening* exhibited by humid granular materials above a typical strain. The present short article gives the opportunity to review the potential physical origin of the cohesion in granular materials and, then, to discuss one experimental example which points out the fundamental role played by the clustering in the mechanical response of the cohesive materials.

Recientemente, hemos reportado la formación espontánea de un patrón periódico en la superficie de una lámina fina de material granular cohesivo sometido a estiramiento en el plano [1] o a pandeo [2]. El mecanismo responsable de dicha inestabilidad es el reblandecimiento que exhiben los materiales granulares húmedos por encima de cierta deformación típica. Este artículo corto nos brinda la oportunidad de revisar el posible origen físico de la cohesión en los medios granulares, y de discutir un ejemplo experimental que sugiere el papel fundamental que juega el "clustering" en la respuesta mecánica de los materiales cohesivos.

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INTRODUCTION

Adding even small minute amounts of liquid can change dramatically the mechanical properties of sand. During the building of sand castles, one observes a transformation from a fluidlike to a sticky and deformable material with increasing water content. Indeed, at very low water content, the formation of partially developed capillary bridges leads to a fast increase of tensile strength whereas, for large enough fluid content, tensile strength is nearly constant [1]. Cluster formation was identified as the main mechanism responsible of such a behavior [2].

POTENTIAL SOURCES OF COHESION

The granular matter is cohesive when attractive forces between the grains are at play. The aim of the present section is to briefly review the potential sources of interaction between the grains.

To begin with a source of cohesion which is one of the most difficult to account for [3], we mention that, due to friction, the grains, if made of a dielectric material, can acquire a static electric charge when flowing. In a dry atmosphere, if the container is not grounded, the electrostatic force can be large enough to lift grains of millimetric size [4]. The charge is difficult to estimate. So are the resulting interaction forces. However, we note that, for solid grains made of a dielectric material, the charge of all the grains has the same sign, which leads to repulsive forces and, thus, not to cohesion [5].

The electroclamping of the particles is a second, but peculiar, source of cohesion which is worth mentioning here [6]. Some authors reported the possibility to induce and, thus, to tune the cohesion of granular materials by applying an external electric field [7].

Finally, even when the grains are electrically neutral, they are subjected to an attractive electrostatic force arising from the polarisability of the material. In order to evaluate the intensity of the associated effect, one can compare the magnitude of the Van der Waals force between two spheres of radius R separated by the minimum distance δ, $F_{VdW} = AR/12\delta^2$, where A is the Hamaker constant [8], with the weight of one grain, $W = \frac{4}{3}\pi r^3 \rho g$, where g is the acceleration due to the gravity and ρ the density of the material the grains are made of. The force F_{VdW} diverges if the grains are smooth and in contact but, in practice, the distance δ is limited by the roughness of the grain surface. For a typical value of the Hamaker constant of about 10^{-19} to 10^{-21} J and a typical size $\delta \sim 100$ nm associated with the roughness, one estimates that the Van der Waals force is significant for grains having a radius of about a few micrometers or less, thus for fine powders only.

Like electrostatic interactions, magnetic interactions can lead to cohesion. Even if this case is again peculiar, particles having an induced or permanent magnetic moment can attract each other. Experiments [9] and, accordingly, numerical

simulations [10] show that, because of long-range interactions, the effects of the magnetic interactions on the angle of repose remains weak even if the forces associated with the bead-bead interactions can be significantly larger than the weight of the beads.

Together with the Van der Waals interaction, the most frequent source of cohesion is the capillary force associated with the liquid bridges that spontaneously form at the contact points between the grains in contact with a humid atmosphere. This latter case will be thoroughly discussed in the next section but, before ending this list of the main potential cohesion sources, we mention that, in some cases, the liquid at the contact points between the grains mediates chemical reactions that result in solding the grains to each other. Such solid bridges form, for instance, for glass beads in contact with a humid atmosphere [11] or immersed in water [12]. Again, the magnitude of the associated cohesion is difficult to estimate because the strength of the solid contacts is largely system dependent. The same conclusion holds true for sticky grains.

The next section focusses on the special case of the cohesion due to the liquid bridges formed at the contact between the grains.

HUMIDITY RELATED COHESION

For grains having a typical size from tens to hundreds of micrometers, the main source of cohesion is the formation of capillary bridges between the grains [13, 14, 15, 16]. In order to approach the phenomenon, one can estimate that, for a bridge at equilibrium between two smooth grains of diameter *d*, the typical value of the force is γd, where γ denotes the surface tension of the liquid-vapor interface [8, 13]. Note that the result is independent of the bridge size and that, in such conditions, the capillary forces would be important for any grains smaller than the capillary length (constructed with the density of the beads material), $\sqrt{\gamma / \rho g}$ which is of the order of 1 *mm*. The conclusion that, independent of the water content, grains smaller than 1 *mm* would naturally remain stuck to each other is obviously contradicted by everyday experience.

In practice, the roughness of the grain surface makes the situation slightly more complicate. Indeed, when the grains are not smooth, depending on the amount of water at a contact between two grains, one can identify three different regimes, namely the *asperity*-, the *roughness*- and the *sphere*-regimes [14]. For grains in contact with a humid, but reasonably dry atmosphere (relative humidity smaller than typically 70%), one must thus consider that two neighbouring grains are connected by a series of small bridges at the scale of the roughness and not by a single bridge.

Another experimental feature is that the water is not homogeneously distributed in the bulk of the granular material [17, 18]. First, due to the heterogeneity of the packing at the grain scale, the nucleation is easier at some contacts. We mention, even if the phenomenon is out of the scope of the present article, that the thermally activated nucleation of the liquid bridges in the material leads to an ageing of its mechanical properties, for instance of the angle of avalanche [12, 19, 20]. Second, even at equilibrium, the geometry of the bridge is entirely imposed by the thermodynamic equilibrium with the vapor and by the boundary conditions. Thus, an increase of the water content mainly leads to an increase in the number of bridges and not to a change in the volume of the existing bridges [21]. Due to the history of the bridges formation, the material consists of an assembly of wet clusters. As a consequence, for instance, the discharge of a wet granular material through an orifice involves the flow of clusters instead of individual particles [22], leading to a larger effective size of the flowing particles [23].

The mechanical properties of the cohesive granular matter are the subject of many experimental and theoretical studies [24, 25, 26, 27]. The authors mainly focussed on the assessment of the tensile stress and shear modulus as a function of the water content. Here, we understand by tensile stress the maximum stress necessary to open a cut plane in the bulk of the granular material. However, it is well-known that the force due to a liquid bridge decreases when the grains are pulled apart [28]. This feature at the contact level leads, at the level of the granular material, to a *stretch-thinning* behavior that leads to the mechanical instability of the material under tensile forces [1] and to the formation of fractures whose origin qualitatively differs from that of the fractures more classically observed in compression [29].

The *stretch-thinning* properties of the cohesive granular matter have been recently studied by H. Alarcón and F. Melo [30, 31]. For the purpose of the present article, let us very briefly summarize the conclusions. First, the tensile stress indeed decreases when the material is stretched. The maximum tensile stress, σ_s, increases when the humidity content, R_H, is increased and the grain size, *d*, decreased. The associated shear modulus is proportional to the tensile stress: $G \propto d\sigma_s$. The decrease of the tensile stress, when two blocks of the cohesive granular are pulled apart, is characterized by a distance $\delta \sim \sigma_s d^2 / \gamma_f$, where γ_f has the dimension, and the order of magnitude, of a surface energy.

We shall see in the next section, that such properties explain the dependence of the typical size associated with the fracturing of a thin, cohesive, granular layer on the layer thickness *h*, grain size *d* and relative humidity R_H.

INSTABILITY AND FRACTURING OF A THIN COHESIVE GRANULAR LAYER

Spreading a thin layer (typically 1 *mm*) of cohesive granular matter (flour, for instance) onto an elastic membrane and then stretching it by pulling two opposite ends reveals a very intriguing and beautiful pattern (Fig. 1). In Ref. [1], we provided an explanation for the well-defined angle the

fractures make with the stretching direction and reported the dependence of the wavelength on the experimental parameters.

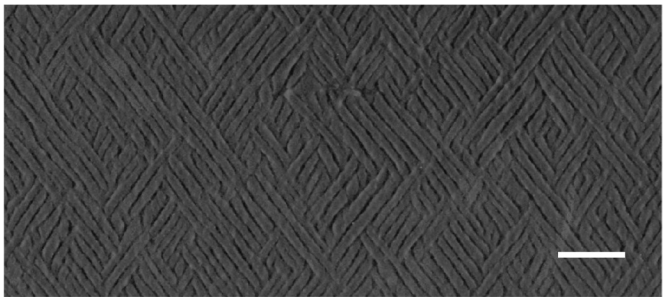

Figure 1: Fracture pattern that spontaneously forms when a cohesive granular layer is stretched on top of an elastic membrane. The layer is stretched along the left-to-right axis and the scale bar is worth 1 *cm*. The granular material consists of spherical glass-beads (USF Matrasur, sodosilicate glass, bead diameter d = 0 - 45 μm). The free surface of the layer (thickness, h = 1.5 *mm* is imaged from above by means of a digital camera (Konica-Minolta, A200). Two linear arrays of LEDs, placed at two opposite sides, about 20 *cm* away from the sample, a few centimeters above the table plane, provide grazing light and, thus, a good contrast when the upper surface of the material is deformed (see [1] for details).

The angle that the fractures make with the stretching direction is independent, for spherical glass beads, of the size d of the particles, of the layer thickness h and of the relative tive humidity, R_H. As a matter of fact, the elongation of the membrane in a given direction produces a contraction in the perpendicular direction. As a result, in the perpendicular direction, the grains are pushed against each other. The tensile stress σ_s is thus involved in the stretching direction whereas the much larger elastic modulus E of the material the grains are made of is involved in the perpendicular direction. The stress is negligible in the stretching direction and one can account for the experimental angle by considering the Mohr criterion for a pure compression in the perpendicular direction [32]. The stripes make the angle $\pm\alpha$ with the stretching direction, with $\alpha = \frac{4}{\pi} + \frac{\Phi}{2}$ and $\tan\Phi = \mu$, the internal friction coefficient.

The typical spacing between the fractures is governed by the *stretch-thinning* properties of the material as proven by results obtained for a pure uniaxial stretching [1] or for a pure bending [2]. Indeed, due to the strain softening, in response to the overall stretch, the system spontaneously tends to modulate the deformation: Regions of large deformation are associated with a smaller modulus and regions of large modulus are associated with a smaller deformation, which results in an overall decrease of the energetic cost. In turn, the modulation induces a shear deformation in the thickness h of the sample, which is associated with an energetic cost. Thus, the wavelength is governed by the balance of the gain associated with the modulation of the horizontal strain and of the loss associated with the induced shear. The main result of the theoretical analysis is that the wavelength is proportional to the layer thickness and that, in the limit of small liquid bridges (small cohesion),

$$\lambda = 4\sqrt{\frac{2}{G}\frac{\partial\sigma}{\partial\theta}}\,h, \qquad (1)$$

the slope $\frac{\partial\sigma}{\partial\theta}$ accounting for the decrease of the tensile stress σ when the uniaxial strain θ is increased [1].

In the light of the recent assessment of the mechanical properties of the cohesive granular matter [30, 31], one can understand where the dependence of the wavelength on the grain size and humidity comes from. Indeed, one can write $\frac{\partial\sigma}{\partial\theta} = \frac{\sigma_s}{\theta_m}$ and, then, $\lambda = 4\sqrt{2\sigma_s / G\theta_m}\,h$, where θ_m is the characteristic uniaxial strain associated with the rupture of the material. From the latter relation and from $G \propto d\sigma_s$, one leads to $\lambda \propto h/\sqrt{d\theta_m}$. Denoting nd the typical size of a cluster and considering that the maximum distance over which the grains can be pulled apart before the rupture is governed by the roughness of the surface, one obtains $\theta_m = \delta/nd$ and, then, $\lambda \propto h\sqrt{n/\delta}$, where δ is a length which characterizes the roughness (δ is found independent of the grain size d for our samples). We find that the whole set of experimental data is correctly described taking $n = 1 + \sigma_s d^2/\gamma^*\delta$. The only adjustable parameter γ^* is found to be of the order of 10^{-3} J/m^2 for glass beads in equilibrium with a humid atmosphere.

The conclusion is that the phenomenon is drastically dependent of the properties of the grain surface, in particular to the roughness, as expected at small relative humidity R_H, in the asperity or capillary regimes [14]. The phenomenon also points out the importance of the heterogeneity of the water concentration in the system. Indeed, the dependence of λ on R_H is only due to the dependence of the cluster size n on the humidity content.

CONCLUSIONS

The present article was the opportunity to briefly review the physical mechanisms at play in the cohesion of the granular matter and to illustrate, in a simple experiment, that the properties at the contact scale and the heterogeneity of the water distribution at the scale of a few grains play a crucial role in the mechanical response of the material subjected to a tensile deformation. The *stretch-thinning* feature of the adhesion force leads to the formation of a pattern whose characteristics have been related with microscopic parameters. Note finally that the mechanical instability should be observed in any material exhibiting a negative sensitivity to the strain increase, might prove to be more general and, thus, deserves further investigation.

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