

**PHYSIQUE DE LA MATIÈRE EN GRAINS**  
**PHYSICS OF GRANULAR MEDIA**

**Physics of humid granular media**

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**Abstract**

A very small amount of liquid added or condensed from a vapor in a granular heap can induce dramatic changes of its static properties. In this paper we review recent advances in humid granular media. We discuss the first approaches for describing the cohesion forces acting between spherical rough beads, and their effect on the maximum avalanche angle of a granular heap. We also discuss the time dependency of these cohesive forces leading to ageing effects in the properties of the medium. *To cite this article: L. Bocquet et al., C. R. Physique 3 (2002) 207–215.* © 2002 Académie des sciences/Éditions scientifiques et médicales Elsevier SAS

**granular media / humidity / ageing / adhesion / capillary condensation**

**Physique des matériaux granulaires humides**

**Résumé**

De très petites quantités de liquide dans un milieu granulaire peuvent changer de façon importante ses propriétés statiques. Cet article présente la compréhension actuelle des propriétés des milieux granulaires humides. Nous discutons les premières approches décrivant les forces de cohésion entre grains rugueux et leur prise en compte à l'échelle macroscopique, ainsi que la dépendance temporelle de ces forces cohésive induisant le vieillissement des propriétés du milieu granulaire. *Pour citer cet article : L. Bocquet et al., C. R. Physique 3 (2002) 207–215.* © 2002 Académie des sciences/Éditions scientifiques et médicales Elsevier SAS

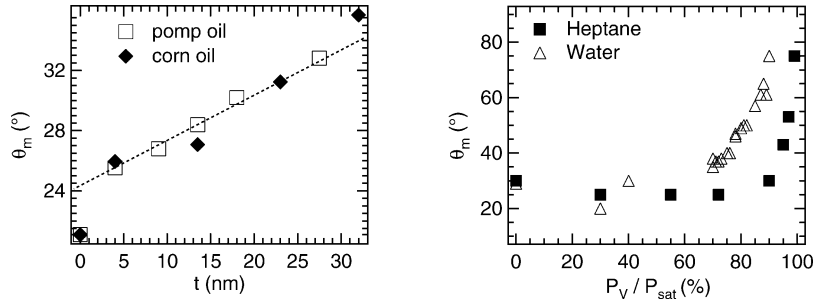
**granulaires / humidité / vieillissement / adhésion / condensation capillaire**

**1. Introduction**

It is an everyday experience that a small amount of liquid can change drastically the physical properties of granular matter. Such effects are important not only on the beach for building sandcastles, but also in a number of industries as diverse as pharmaceutical, construction and agriculture, and in a number of geophysical applications.

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**Figure 1.** Left (after Hornbaker et al. [2]): variation of the angle of repose  $\theta_r$  of 0.8 mm diameter polystyrene spheres with the amount of oil in the sample. The volume of liquid has been converted to an equivalent thickness  $t$  on the spheres. Right (after Fraysse et al. [4]): variation of the maximum angle of stability  $\theta_m$  of 200  $\mu\text{m}$  glass beads with the vapor content of the surrounding atmosphere for heptane and water vapor.

The changes induced by a small amount of liquid in granular materials are primarily caused by adhesive forces associated with interstitial liquid bridges between grains [1]. The role of those capillary forces has been known for long, however their consequences on the properties of granular matter are still poorly understood. In this paper, we briefly review the actual understanding of the specific problems raised by humid granular media, such as it emerges from the studies undertaken recently and independently by several research groups.

## 2. Some experiments in humid granular materials

Two recent experiments illustrate more specifically the effect of a small amount of liquid on the static properties of granular matter Fig. 1. The first one, done by the group of Hornbaker et al. in Notre-Dame [2,3], shows the effect of small quantities of oil added to spherical polystyrene beads on their angle of repose. The measurement—the draining-crater method—is based on the formation of a crater in a thick horizontal layer of beads by opening a small aperture in the bottom plane supporting them. Hornbaker et al. find a large increase of the angle of repose, which grows linearly with the the amount of liquid added to the beads.

The other experiment, done by Fraysse et al. in Nice [4], studies the effect of a condensable vapor in the atmosphere on the avalanche angle of glass beads. The measurements are made in a horizontal rotating drum. They show an enormous increase of the avalanche angle of the glass beads when the humidity content of the atmosphere is larger than some value—70% for water vapor, 90% for heptane vapor—the humidity  $H$  being defined as the ratio  $P_V / P_{sat}$  of the vapor partial pressure to its saturating pressure at the temperature of the experiment.

The common point of these experiments is the extremely small quantity of liquid needed to obtain large effects. In Hornbaker’s experiment, the average thickness of the oil-layer on the beads is less than 50 nm, which corresponds to a volume of liquid at most  $10^{-4}$  of the solid volume. In Fraysse’s experiment, the estimated amount of liquid condensed in the beads is even lower.

A first attempt to understand such large effects is to calculate the adhesion force between grains. Adhesion of particles has sustained the interest of colloid scientists for the major part of the last century, and the cohesion forces between smooth surfaces are now well understood. The force exerted by a liquid bridge connecting two spherical grains can be calculated in the frame of classical capillarity down to very small radii of curvature  $r$  of the liquid meniscus ( $r \sim 4\text{--}5$  nm, [5]). This force is:

$$F_{cap} = 2\pi\gamma \cos\theta_Y R(1 - D/2r), \quad (1)$$

with  $R$  the radius of the beads,  $D$  their distance,  $\gamma$  the liquid surface tension and  $\theta_Y$  its contact angle on the solid. This expression holds for a small liquid bridge filling an angle up to  $5^\circ$  from the center of the sphere [6,7]. It shows the major feature of the capillary force: its range is given by the height of the meniscus, and its maximum value  $F_{\text{cap}} = 2\pi\gamma \cos\theta_Y R$  when the spheres are in contact, does not depend on the amount of liquid. Furthermore the strength is very large: in the case of water, with  $\cos\theta_Y = 1$ , the adhesive force between glass spheres of radius  $R = 3$  mm equals their weight. Thus even a very small meniscus could be able to stick completely a particule to another against gravity if  $R < 3$  mm.

An even more disturbing property appears when one takes into account the adhesive forces due to solid-solid interactions. The theory of adhesive contact between elastic spheres has been treated in detail by Derjaguin–Muller–Toporov and Johnson–Kendall–Roberts, and later by Maugis et al. [8–10]. For elastic spheres of high Young’s modulus, the adhesion force is  $F_{\text{SS}} = 2\pi\gamma_{\text{Sx}}R$ , where  $\gamma_{\text{Sx}}$  is the interfacial tension of the solid and the fluid phase (liquid or gas) between the spheres. Taking into account Young’s equation of wetting:  $\gamma_{\text{SL}} + \gamma \cos\theta_Y = \gamma_{\text{SG}}$ , it appears that the total adhesion force in the presence of a liquid bridge, including the capillary force, has exactly the same value as in the dry case. This result is limited to the case of partial wetting, i.e.  $\theta_Y > 0$ . In the case of total wetting, adsorption and wetting liquid films have to be taken into account [11]. The adhesion force is then lower in the presence of a liquid or a condensable vapor atmosphere than in the vacuum, a result well known in the surface science community [5].

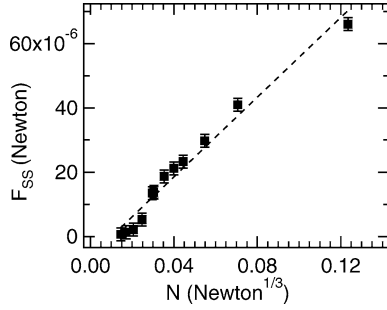
All this physics of smooth surfaces clearly disagrees with experimental observations of cohesive effects in granular media. The reason is qualitatively understood since the work of Tabor [12], and is due to the roughness of real surfaces. A first effect of roughness is to screen all interactions between surfaces. The shorter the interaction range, the more efficient is the screening. Since solid-solid interactions have a molecular range they are very efficiently screened by a roughness of nanometric height. The range of capillary forces however increases with the size of the liquid patches connecting the surfaces, which explains that cohesive behavior in a granular medium is found in the presence of liquid and increases with the amount of liquid. Another effect of roughness is that normal stresses concentrate at the tip of asperities in contact and lead to their plastic deformations, which in turn modifies the real area of contact and its adhesion properties. In the next paragraph we summarize the actual understanding of the cohesion forces between rough grains.

### 3. Cohesion forces between grains: a problem of surface roughness

On a quantitative point of view very little is known up to now on the interactions between rough grains—or rough surfaces. One reason is that the relevant lengths scales for cohesive forces are usually in the nanometer range—in Hornbaker’s experiment the largest film thickness studied is 50 nm—and it is only lately that powerful experimental methods at that scale have become easily available.

The range of solid–solid interactions is at the molecular scale, and the specific methods required at that scale are the Surface Forces Apparatus (SFA) or the atomic force microscopy. In our group, we have made first studies of the adhesion force between surfaces of nanometric roughness, by using float glass immersed in dodecane [13]. Typical experiments with a SFA involve approaching a sphere and a plane up to contact, then separating them to measure the maximum adhesion force (‘pull-off’ force). The amplitude of this adhesion force is much lower than expected in the case of smooth surfaces. However, the most important result of these first experiments is the dependency of the adhesion force on the maximum normal load  $N$  that has been applied on the surfaces before separating them: we find a power law dependance  $F_{\text{adh}} \propto N^{1/3}$  (see Fig. 2).

This feature is in total disagreement with the DMT or JKR theory for smooth surfaces, and is even not compatible with a purely elastic deformation of the solids in contact. This effect can however been understood if one take into account the plastic deformation of asperities [14]. When the normal load  $N$  acting on the contact is such that the elastic limit of the samples is not exceeded, the bulk deforms elastically and the macroscopic area of contact satisfies Hertz’s law:  $A_{\text{H}} = \pi(NR/E^*)^{2/3}$ , with  $E^*$  the



**Figure 2.** The maximum adhesion force  $F_{SS}$  between a sphere and a plane of float glass immersed in liquid dodecane, measured with a Surface Force Apparatus. The surfaces are rough at the nanometer scale. The radius of the sphere is  $R = 1.8$  mm. The  $x$ -axis is the power  $1/3$  of the maximum load  $N$  that has been applied on the surfaces before pulling them apart.

reduced Young’s modulus of the solids. However molecular contact is not achieved everywhere inside this region, because of small asperities. Several mechanisms suggest that the real (molecular) area of contact  $A_r$  is proportionnal to the normal load  $N$  [12,15]. This is obtained if the asperities flatten plastically, or if the interstitial medium exhibits a plastic behaviour induced by the confinement. Neglecting the interaction between surfaces which are not in molecular contact, the effective surface energy created when the surfaces are separated is  $\gamma_{\text{eff}} = \gamma_{\text{SL}}A_r/A_H$ . The DMT theory of the adhesion force then predicts that:

$$F_{SS} = 2\pi R\gamma_{\text{eff}} = 2\gamma_{\text{SL}}E^{*2/3}(RN)^{1/3}/H, \tag{2}$$

where  $H$  is the parameter relating the real area of contact to the normal load:  $A_r = N/H$ . Taking for  $H$  the hardness of glass, the comparison with experimental results gives a value of  $\gamma_{\text{SL}}$  of  $0.43 \text{ J/m}^2$ , which is the order of magnitude expected for atomically smooth glass surfaces interacting with a liquid alkane.

These first results show that adhesion forces between rough surfaces can be very different than for smooth surfaces: not only the amplitude is different, but a new parameter arises, the external force applied to the contact. On a macroscopic level, this means that in modelling cohesive granular media one should eventually take into account a dependance of the cohesion forces on the externally applied stress. We discuss this aspect in the next paragraph.

The capillary force between rough surfaces is also expected to be very different from the one given by Eq. (1). In a first approximation this force is  $F_{\text{cap}} = \gamma A/r$  where  $A$  is the area of the solid surface immersed in liquid bridge(s) having all the same radius of curvature  $r$ . For a given roughness the determination of  $A$  and  $r$  is a geometrical problem. Two different physical situations must be considered. If a non-volatile liquid is added to the grains as in Hornbaker’s experiment, the constraint is the fixed value of the liquid volume  $V$ . Halsey and Levine [16] have made a first calculation of the capillary force in this case. They describe the roughness by its typical height  $l_R$ , its correlation length  $d$ , and an exponent  $\aleph$  describing its shape at small scales. The gap  $\delta(x)$  between the two opposite surfaces at a distance  $x$  from two asperities in contact is:  $\delta(x) = l_R(x/d)^\aleph$  when  $x < d$  and  $\delta(x) \sim l_R$  when  $x \gg d$ . Halsey et al. find three different regimes of variation of the capillary force with the volume  $V$  of liquid added.

- For very small values of  $V$  some liquid bridges grow around asperities in contact: this is the ‘asperity regime’. The volume of such bridges is roughly  $V \sim Ar$ , where  $r$  increases as  $A^{\aleph/2}$ . Therefore the capillary force in the asperity regime scales as  $F_{\text{cap}} \propto V^{(2-\aleph)/(2+\aleph)}$ .
- When the lateral extension of the bridges becomes larger than the correlation length  $d$ , their height saturates at the value  $l_R$ , and liquid fills the contact between two beads while the radius of curvature of the meniscus remains constant. This is the ‘roughness regime’, in which the capillary force grows linearly with the volume  $V$  of liquid.
- Finally when the lateral extension of the liquid patch between the two beads becomes larger than  $\sqrt{l_R R}$ , the macroscopic curvature  $R^{-1}$  of the surfaces overcomes the roughness, and the capillary force reaches its value for smooth surfaces:  $F_{\text{cap}} = 2\pi\gamma R$ .

These three regimes have not been explicitly observed experimentally, however Halsey and Levine consider that the ‘roughness regime’ describes the dependency observed in Hornbaker’s experiments.

A different physical situation is when the granular medium is in an atmosphere of condensable vapor. The equilibrium radius of curvature  $r$  is then determined by the value of the chemical potential. For instance, in an atmosphere of humidity  $H = P_V/P_{\text{sat}}$   $r$  is given by Kelvin’s law:

$$\gamma/r_K = \rho_L k_B T \ln P_{\text{sat}}/P_V \quad (3)$$

with  $\rho_L$  the liquid molecular density. In the framework of the Halsey–Levine analysis, the ‘roughness regime’ should be reduced to a narrow range of humidity values such that  $r_K \sim l_R$ , in which the capillary force should jump from a weak value to the constant value  $2\pi\gamma R$ . The experiment of Fraysse et al. actually shows a sharp increase of the avalanche angle as a function of  $H$ . However a numerical estimation shows that the humidity value needed for reaching radii of curvature of the order of a typical roughness—0.1  $\mu\text{m}$  to 1  $\mu\text{m}$ —is extremely close to 100%. At a humidity of 75%, for which a large increase of the avalanche angle is observed in Fraysse’s experiment, the equilibrium radius of curvature of a water meniscus is about  $r_K = 5$  nm. This value is very small; it is then likely that capillary condensation occurs at the level of the tip of asperities in contact. As discussed before, the area of such contacts depends on the normal load, and the latter should be taken into account to evaluate the capillary force. Another important point is that capillary condensation is a first order transition, associated to metastability effects. We come back to this problem in the last paragraph.

#### 4. Cohesion and avalanche angle

In the previous section we have discussed the nature and behavior of cohesion forces at the scale of one grain. In order to discuss the stability of a heap, a macroscopic criterion for failure is required. This is provided by the Coulomb yield criterion. For a noncohesive material, the latter states that on the slip plane the tangential and normal stresses, respectively  $\tau$  and  $\sigma$ , are linearly related:  $\tau = \mu\sigma$ . In this equation,  $\mu = \tan\phi$  is a friction coefficient. This condition fixes the upper limit for the shear stress on any plane. A Mohr–Coulomb analysis then shows that the maximum angle of stability of a granular heap is simply  $\theta = \phi$  [17]. This analysis can be extended to the cohesive case. The adhesive stress  $c$  is included phenomenologically in the Coulomb yield criterion for failure as  $\tau = \mu(\sigma + c)$ . This simply amounts to add the effects of adhesion and normal ‘compressive’ forces. On this basis it is possible to obtain the maximum angle of stability of a cohesive heap by performing a common Mohr–Coulomb analysis, as done, e.g., by Halsey and Levine [16]. This analysis predicts that the failure occurs at the bottom of the granular heap, when the angle exceeds a critical angle  $\theta_m$  verifying the implicit equation:

$$\tan\theta_m = \tan\phi \left( 1 + \frac{c}{\rho gh \cos\theta_m} \right) \quad (4)$$

with  $h$  the height of the granular heap,  $\rho$  the mass density of the granular material and  $g$  gravity. The cohesive stress  $c$  is eventually related to the (average) adhesion force  $f_{\text{adh}}$  between two grains as  $c = \alpha f_{\text{adh}}/R^2$ , with  $R$  the radius of one grain. The numerical prefactor  $\alpha$  only depends on the volume fraction of the granular material and is thus expected to be of order unity. As expected cohesion increases the maximum angle of stability  $\theta_m$  as compared to the dry case. An angle exceeding  $\pi/2$  can even be reached for sufficiently strong adhesion,  $\mu c > \rho gh$ .

A few remarks have to be made on this analysis. An important feature of the result (4) is that a specific size effect is predicted: when the depth of the material  $h$  reaches infinity, cohesion has no effect on the stability of the granular material and the maximum angle of stability is  $\theta = \phi$ . This result could be however questioned since it is intimately related to the assumption of a spatially uniform cohesive stress: for a given cohesive stress, the ratio between weight and cohesion is maximum at the bottom and the material will fail at

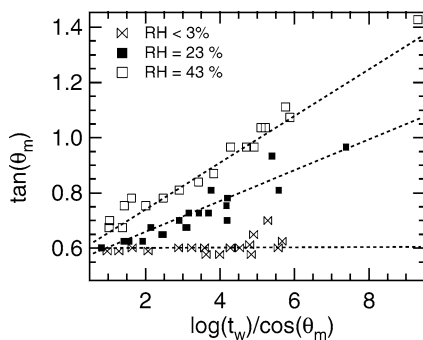
depth. But as discussed in the previous section, the adhesion force between two grains is likely to depend—at least in some regimes—on the normal force exerted on the grains. Consequently, the cohesive stress would be a function of the normal stress  $c = c(\sigma)$  and the Mohr–Coulomb analysis has to be reconsidered accordingly. For example, if  $c$  is linear in  $\sigma$ , the size dependence of  $\theta_m$  disappears. Such a situation would occur in a regime where the adhesion force is proportional to the real contact surface between two grains: according to the classical argument of Tabor [12] this area is proportional to the normal force  $N$  acting on the grains, so that  $f_{adh} \propto N$  and  $c \propto \sigma$ . In general, however, the specific dependence of the cohesive stress on  $\sigma$  has to be known to discuss the size dependence in avalanche processes. More experimental work is anyway needed to explore these size effects in cohesive materials.

### 5. Humidity induced ageing in granular media

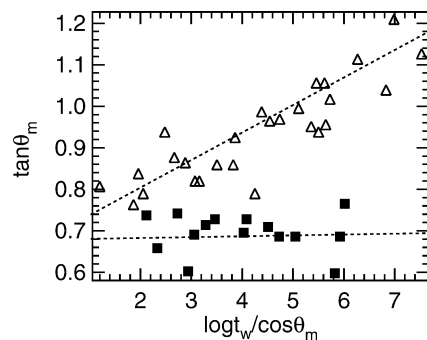
The experiments described in Section 1 do not explicitly incorporate time as a parameter for cohesion effects in granular media. The cohesion parameters are seen as intrinsic properties of the material. It is well known from the study of solid friction, however, that the properties of a contact are time-dependent, the so-called ‘ageing’ effect [18–20]. The static friction coefficient, in particular, exhibits a logarithmic dependence on the time elapsed after the contact is formed (the ‘waiting time’). As friction effects are an essential ingredient in the static properties of granular media, it seems appropriate to investigate the occurrence of similar ageing effects in a granular system. For this purpose, we have developed an experimental setup allowing a controlled study of the avalanche angle as a function of the waiting time—in this case the time elapsed after the granular heap has been formed [21,13].

The experiments are performed in a controlled environment—temperature and humidity. A rotating drum geometry allows us to vary easily the waiting time  $t_w$  during which a granular heap is kept at rest before undergoing an avalanche. We emphasize that this waiting time is by no means the time during which the granular medium has been exposed to humidity: the beads are incubated at least 48 hours in the controlled environment before any experimental run.

Fig. 3 shows the evolution of the maximum stability angle as a function of the waiting time for glass beads of diameter comprised between 200 and 250  $\mu\text{m}$  in different humidities. The plot is done along the analysis leading to Eq. (4), i.e.  $\tan\theta_m$  is plotted against  $\log t_w / \cos\theta_m$ . The data clearly show a temporal evolution of the cohesion force, compatible with a logarithmic dependence in time over time scales ranging from 10 min to 24 hours. No ageing is observed in a dry atmosphere, which is consistent with the absence



**Figure 3.** Time evolution of the maximum stability angle  $\theta_m$  of a 200  $\mu\text{m}$  glass beads heap for various water content  $H$  of the atmosphere. The experiments are made in a rotating drum of 10 cm in diameter and length. An experiment consists in stopping a quick rotation of the drum, wait a time  $t_w$ , and rotate slowly the drum until an avalanche occurs.



**Figure 4.** Time evolution of the maximum stability angle of the heap at an humidity  $H = 43\%$ , for different positions during the resting time. Open symbols: the free surface of the heap rests at an angle  $\theta_{ref} = 28^\circ$  with respect to an horizontal. Filled symbols:  $\theta_{ref} = 0^\circ$  the heap rests with an horizontal free surface.

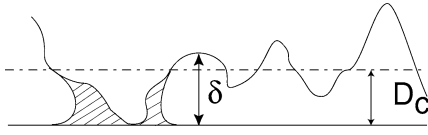
of liquid-induced cohesion in this case. It also shows that the ageing behavior observed here has nothing to do with the compaction induced ageing reported in the literature [22]. In the following we measure the amplitude of this humidity-induced ageing by the slope  $\alpha$  of the best linear fit in the plot of Fig. 3.

The amplitude of this ageing behavior depends on a number of experimental parameters, aside humidity, such as the size, the shape and the state of wear of the grains, the angle of the free surface of the heap during the resting time, the nature of the condensable vapor, etc. [13]. Before discussing in more detail the role of humidity, we underline the important effect of the resting angle  $\theta_{\text{ref}}$ , i.e. the angle at which the heap is kept at rest. Fig. 4 compares the temporal evolution of  $\tan \theta_m$  for a sample of glass beads which has aged at  $\theta_{\text{ref}} = 0^\circ$  and a sample of glass beads which has aged at a reference angle closer to the avalanche angle measured at short time:  $\theta_{\text{ref}} = 28^\circ$ . The amplitude of ‘horizontal’ ageing is much smaller than when the pile ages at an angle close to the avalanche angle. This behavior can be related to results reported in solid and granular friction. In granular friction, Nasuno et al. [23] report some very slow displacements (creep) which precedes the rapid events in a stick–slip experiment on granular media. The same precursors have been also reported in solid friction by Baumberger et al. [24]. Those precursors change dramatically the history of the adhesive contacts by breaking old ones and reforming new young ones. To induce such changes, their amplitude should overcome the range of the cohesive forces, which is much smaller than the size of the grains, as seen previously. Such precursors are much more numerous and important when the heap is tilted from an horizontal position up to the avalanche angle, which explains the weak ageing effect obtained in this case.

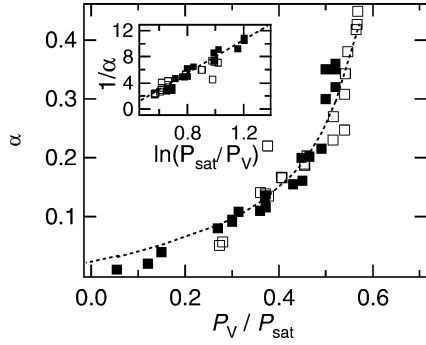
As shown in Fig. 3, humidity is the main parameter acting on ageing. By comparison with the experiments of Fraysse et al., one sees that the values of the vapor pressure which induce a temporal behavior are significantly lower than the one needed to modify the short time avalanche angle. Indeed, for  $H$  larger than about 60%, ageing become so large that after some time the avalanche angle may exceed  $90^\circ$ , and the plot of Eq. (4) cannot be used anymore. This strong influence of humidity, and the fact that ageing disappears in dry atmosphere, suggests a temporal evolution of the liquid-induced cohesion forces in the medium. In solid friction, ageing properties have been studied on various materials, and have been related to the slow viscoplastic increase of the area of contact between asperities induced by the high values of the stress in the contact region [25,26]. However the importance of humidity has been reported by geophysicists in rock onto rock solid friction [27]. In the presence of a vapor atmosphere, the condensation of a liquid phase appears as an important factor. This is due to the large metastability associated with the capillary condensation processes of a wetting phase between two surfaces, which lead to slow dynamics processes. Such effects are currently encountered in porous media, where they are responsible for hysteresis of the adsorption isotherms, and have also been studied on a single contact in SFA experiments [11].

The origin of this metastability in capillary condensation is the following: consider two parallel plates separated by a distance  $D$ , in contact with a vapor. The free energy per unit area is  $2\gamma_{\text{SV}}$ . When the liquid condenses it becomes  $2\gamma_{\text{SL}} + \Delta\mu\rho_{\text{L}}D$ , where  $\Delta\mu = \mu_{\text{sat}} - \mu_{\text{v}}$  is the vapor undersaturation. If the liquid wets even partially the surfaces,  $\gamma_{\text{SL}} < \gamma_{\text{SV}}$ , and an unfavorable liquid phase ( $\mu_{\text{v}} < \mu_{\text{sat}}$ ) can be stable in the confined geometry. This occurs if  $D < 2r_{\text{K}}/\cos\theta_{\text{Y}}$ , with  $r_{\text{K}}$  the Kelvin’s radius (Eq. (3)) and  $\theta_{\text{Y}}$  the contact angle. However the free energy favoring the liquid phase is gained only when the space between the walls is fully saturated, and the nucleation of a liquid phase is associated with an energy barrier. The height of this barrier has been evaluated in the ideal case of smooth parallel walls [28]. For a humidity of 50% and a value of  $D \sim 2 \text{ \AA}$ , the energy barrier is much larger than  $100 k_{\text{B}}T$  at ambient temperature, which explains the very long time scale in capillary condensation phenomena.

Based on these facts we can propose a model for the slow dynamics of the cohesion forces induced by humidity between rough surfaces, using the same ingredients than those used to explain the subexponential relaxation reported in glassy systems such as spin glasses [29], complex fluids [30], etc. We consider two surfaces rough at the nanometric scale, facing each other (Fig. 5). Without loss of generality one may assume that one of these surfaces is flat. Preferred sites for the nucleation of a liquid bridge are local minima of the gap  $\delta(x)$  between the surfaces. Those points are localized on the tip of asperities whose radii



**Figure 5.** Schematic view of a contact at nanometric scale.



**Figure 6.** Variation of the amplitude  $\alpha$  of ageing of the maximum stability angle with the relative humidity  $P_V/P_{\text{sat}}$ . Open symbols: glass beads of diameter between 140 and 260  $\mu\text{m}$ . Filled symbols: glass beads of diameter between 200 and 250  $\mu\text{m}$  (after Bocquet et al. [21]).

of curvature  $\lambda$  is assumed independent of the value of the gap. A liquid bridge can be stable if the gap  $\delta$  is lower than the critical distance  $D_c = 2r_K / \cos\theta_Y$  for capillary condensation. Dimensionally, the energy barrier for nucleating such a liquid bridge is  $\Delta\Omega = v_d \rho_L \Delta\mu$  with  $v_d$  a nucleation volume. We assume that (i)  $v_d$  depends only on the gap and not on the local radius of curvature  $\lambda$ , and (ii)  $v_d$  is uniformly distributed: the probability that a site has a nucleation volume  $v_d$  is  $\phi(v_d) dv_d = dv_d / V_0$  with  $V_0$  some roughness-dependant parameter. This is justified since the scales involved for capillary condensation are orders of magnitude smaller than the height of a typical surface roughness. The probability that condensation occurs at an asperity before time  $t_w$  is thus:

$$\Pi(t_w) = 1 - \exp(-t_w/\tau) \quad \text{with} \quad \tau = \tau_0 \exp(\Delta\Omega/k_B T). \quad (5)$$

Each liquid bridge contributes to the capillary force with an average value  $f_{\text{cap}} = 2\pi\gamma\lambda$ . Therefore at time  $t_w$  the capillary force is

$$F_{\text{cap}} = F_0 + 2\pi\gamma\lambda N_{\text{tot}} \int \Pi(t_w) \frac{dv_d}{V_0} \sim F_0 + 2\pi\gamma\lambda N_{\text{tot}} \frac{k_B T}{V_0 \rho_L \Delta\mu} \ln \frac{t_w}{\tau_0} \quad (6)$$

with  $F_0$  the initial capillary force and  $N_{\text{tot}}$  the number of nucleation sites. One obtains a logarithmic evolution of the cohesion force, with a prefactor increasing with humidity and diverging at saturation. This humidity dependency of ageing is in good agreement with experimental results (see Fig. 6).

## 6. Conclusion

In this paper, we have reviewed the current experimental situation concerning humid granular media. Adding a small quantity of liquid to a granular medium induces drastic changes compared to the dry case: increase of the avalanche angles, ageing... This subject, however, has up to now received little attention from physicists. Its study requires an understanding of the system at very different scales, from the nanometer scale at which capillary condensation takes place, to the macroscopic one for describing the equilibrium state of a pile. Many domains of physical sciences are involved: continuum mechanics, friction, surface science, wetting and phase transition in confined systems.

Recent work in this area was focused on cohesion effects induced by humidity, and how they manifest themselves at the level of macroscopic repose angles. At the ‘microscopic’ (grain) level, it is now well understood that roughness plays an important role. First attempts have been made to describe the cohesion



forces between rough surfaces, but many aspects remain largely unexplored: effect of plasticity, role of normal loads, ageing of adhesion forces. Understanding these phenomena is essential to achieve a better description of the cohesion forces at the macroscopic level.

At the macroscopic scale, the simplest analysis of cohesion forces leads to the prediction of a size effect for the macroscopic properties of a pile. This analysis is based on constant cohesion forces, i.e. the dependence on local stresses is not accounted for. These size effects, if present, have not yet been observed experimentally.

Finally, other effects that are specific to humid granular media, such as the formation of clusters and fractures, are observed when cohesion forces become important. These effects, as well as the dynamical (rheological) properties, are important for practical applications, but have received much less attention than the similar problems in dry granular systems.

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