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# Humidity effects on the stability of a sandpile

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**Abstract.** Humidity is well-known to significantly affect the mechanical properties, static as well as dynamic, of granular materials. We present the method of humidification of granular media from an undersaturated vapor that we designed in order to experimentally quantify such moisture-induced effects under accurately-controlled humidity conditions. We report the quantitative measurements of the maximum angle of stability of a pile made of small glass beads, as a function of the relative vapor pressure, up to close to saturation. The results obtained with liquids differing in their wetting properties on glass, namely water and heptane, are presented. It is shown that the wetting properties of the liquid on the grains have a strong influence on the cohesion of the non-saturated granular medium.

**PACS.** 83.70.Fn Granular solids – 68.45.Gd Wetting – 81.05.Rm Porous materials; granular materials

## 1 Introduction

Granular materials are ubiquitous in nature, in the industrial world and in everyday life. These materials most of the time behave in very specific ways, differing in so from the other states of the matter (solids, liquids and gases). As such, dry granular media have been the subject of a renewal of interest for the physicists in the last ten years or so [1,2]. However, in actual practice, grains are usually not dry: they are stored and handled in the open air, and atmospheric humidity can have very important consequences in many industrial processes as well as in laboratory experiments. The nature of contacts between grains plays a major role on the mechanical properties, either static or dynamic, of granular media. Whereas solid friction governs the dry case, capillary forces and viscous forces have to be taken into account when dealing with non-saturated humid granular materials. At the global scale, cohesion and possibly lubrication effects may result from these local interactions between grains. Note that the consequences of these effects in most of the cases are negative (*e.g.* blocked flows from hoppers or silos) but might be positive as it is conceivable that humidity effects oppose segregation for instance.

Despite their importance, for applications and also from a purely fundamental point of view, non-saturated humid granular media have been the matter of very few studies. Most of these studies have been carried out in the field of soil mechanics; they deal with non saturated granular soils and rather high liquid contents. To a less extent, food industry and pharmaceutics have motivated studies which often deal with chemically reactive materials. Only recently physicists have tackled the question, looking at model systems [3–7].

In this paper we present some new experiments performed under accurately controlled humidity conditions. We first introduce our original method for humidification, based on thermodynamic equilibrium of the three-phase system grains-liquid-vapor contained in a close cell. Then we report quantitative results on the stability of a pile as a function of its degree of humidity. For the first time the influence of the wetting properties of the fluid on the solid is investigated, providing an insight into the importance of the local spatial liquid distribution on the macroscopic mechanical behavior.

## 2 Humidification of the granular medium

### 2.1 Principle and implementation of the method

The study of non-saturated humid granular media requires an extreme care during the preparation of the sample. In addition to the usual parameters that should be controlled in experiments on dry granular media, as the packing density for example, or less palpable ones as the way the packing has been built, one should make sure that humidity is uniformly distributed throughout the sample and is constant in time. This very last point is in favor of the use of non-volatile liquids in order to prevent evaporation [3], but, in our opinion, it is difficult to accurately control other parameters in this case. We chose to work with volatile liquids, which incidentally means that water can be studied. It is the fluid of interest in most

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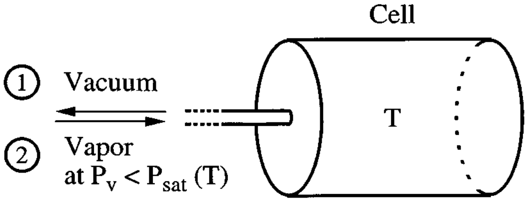

**Fig. 1.** Schematic illustration of the two main steps in the experimental protocol for humidification. (1) The cell, which contains the oven-dried granular medium, is put under vacuum. (2) Undersaturated vapor at pressure $P_{\mathrm{v}}$ is injected into the cell thermalized at temperature $T$. Adsorption and capillary condensation occur in the granular sample according to thermodynamic equilibrium between the liquid, vapor and solid phases.

of practical applications. Basically we reproduce the natural phenomenon of humidification from the water vapor present in the ambient atmosphere.

The experimental protocol is schematically summarized in Figure 1. The granular sample is prepared into the desired cell when dry. The cell is connected through an airtight seal to a set-up which allows us first to make vacuum, and then to inject vapor into the cell. From then on, the cell remains at all times in communication with a reservoir of liquid the temperature of which sets the value of the undersaturated vapor pressure. The pressure in the cell is monitored: a constant value is reached after a few hours typically, indicating that the equilibrium has been reached. Experiments on the humid granular sample can then be performed.

The relevant parameter to quantify humidity is the relative vapor pressure $P_{\mathrm{v}}/P_{\mathrm{sat}}(T)$, where $P_{\mathrm{v}}$ is the experimentally measured vapor pressure and $P_{\mathrm{sat}}(T)$ is the saturated vapor pressure at the temperature $T$ of the cell. This parameter sets the thermodynamic equilibrium of the three-phase system[1]. In order to compute the relative vapor pressure, the saturated vapor pressure has to be known. For usual liquids, it is tabulated as a function of the temperature, but its variation is rapid and these data can be used only if the absolute cell temperature is known to a very good accuracy. In practice, relative variations of the cell temperature can be controlled to a much better accuracy. Our cell is contained in a thermalized box; we checked that the temperature in this box was uniform and stable over several days to within 0.05 °C. This allows us to keep $P_{\mathrm{sat}}$ constant to a good accuracy throughout a whole set of experiments performed at different degrees of humidity, *i.e.* at different vapor pressures $P_{\mathrm{v}}$. Finally we directly measure the pressure at saturation $P_{\mathrm{sat}}$ by increasing $P_{\mathrm{v}}$ up to the point where condensation of the liquid occurs.

Our experimental method of humidification from a vapor offers three main advantages:

(i) First of all, it is a very versatile method which can be implemented with any volatile liquid, on any kind of grains, in very varied experimental geometries.

(ii) The second noticeable advantage of our method of humidification is that it does not require mixing in order to get a uniform distribution of liquid throughout the granular sample. This is particularly crucial when studying very cohesive media [8]. Here the medium is totally and uniformly invaded by the vapor and local thermodynamic equilibrium is reached everywhere, with no perturbation of the initial packing built with dry grains (unless rearrangements occur as a result of capillary forces).

(iii) Thirdly, putting the cell under vacuum prior to injection of vapor leads to much shorter equilibration times than working in air or in any inert atmosphere: mechanical pressures equilibrate in the cell and throughout the granular medium almost instantaneously, whereas transport in air is governed by molecular diffusion, which is a very slow process. Moreover, measuring pressures directly is much easier and usually much more accurate than measuring partial vapor pressure indirectly (using hygrometers for instance if working with water).

### 2.2 Model materials

In this study we worked on non-colloidal monodisperse (diameter 200−250 $\mu$m) glass beads wet by two liquids having different wetting properties on glass:

a) $n$-heptane, a light saturated alkane. Molecular interactions with glass are simple Van der Waals interactions [9] leading to complete wetting (zero contact angle). They are weakly sensitive to local inhomogeneities in the surface properties of the glass substrate. Practically speaking, this is favorable to a homogeneous humidification of the medium. Also, from the fundamental point of view, this system constitutes a well-known model system which should allow a full quantitative interpretation of the experimental results.

b) Water, which is obviously of great practical interest. However, rather complex molecular interactions with glass are involved [10]. In particular, this might result in wetting properties very sensitive to local inhomogeneities in the surface properties of the glass beads, and humidification of the medium might not be totally homogeneous.

## 3 Influence of humidity on the stability of a pile

### 3.1 Characterization of the stability of a pile

We measured the stability of a pile of glass beads using the so-called rotating drum method [11]. The cell here is a glass cylinder (length 10 cm, diameter 10 cm, axis horizontal) which can be slowly rotated around its axis while connected to the set-up as described in Section 2.1.

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[1] Note that $P_{\mathrm{v}}/P_{\mathrm{sat}}$ expressed in percent is strictly equivalent to the percentage of relative humidity that is used to characterize the atmospheric degree of humidity, commonly noted %RH.

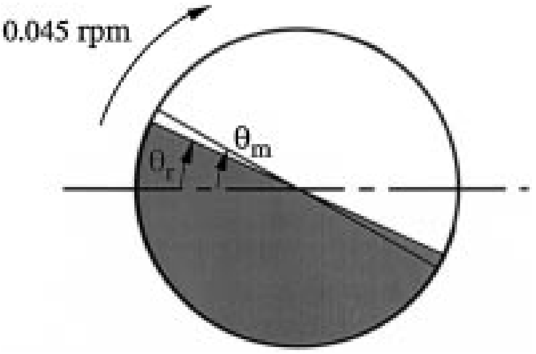


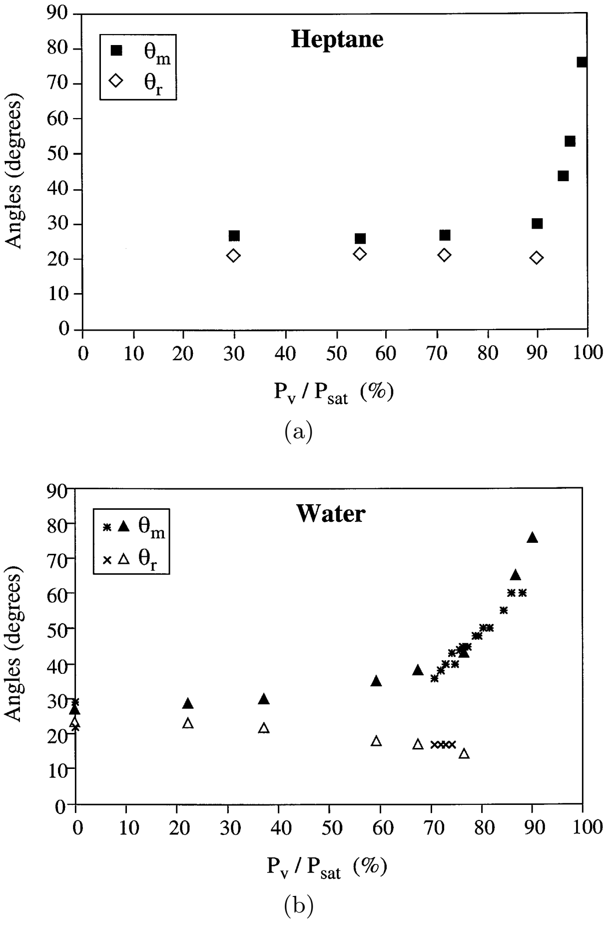
**Fig. 2.** View through the glass end-face of the rotating drum (rotation speed 0.045 rpm) half-filled with glass beads. $\theta_m$ is the maximum angle of stability and $\theta_r$ is the angle of repose, as defined in the text.

The glass beads fill 50% volume of the cylinder; this is the ratio which minimizes wall effects, in depth as well as in the direction parallel to the surface of the pile, for a given size of the cylinder and for a given diameter of the beads. Figure 2 shows a view through the glass end-face of the cell.

It is well-known for dry granular media that, at small rotation speed of the rotating drum, the pile remains unaffected until the angle $\theta$ of inclination of its free surface with the horizontal reaches the "maximum angle of stability", $\theta_m$. The pile cannot be raised beyond this value and, as the rotation of the cylinder continues, an avalanche occurs, which causes the angle $\theta$ to relax to a smaller value that we will call "angle of repose", $\theta_r$.

Qualitatively, we have observed that small degrees of humidity do not change the general scheme described above, although modifying the values of the angles. For large degrees of humidity resulting in rather strong cohesion, the angle $\theta_m$ is still well-defined experimentally since the pile has been prepared with oven-dried beads and has an initial free surface well-defined and plane; however, the angle $\theta_r$ becomes more and more difficult, and even impossible to measure as the free surface of the pile becomes more and more uneven after collapse.

One *a priori* expects complex capillary dynamics in such systems, and the time to reach equilibrium might be an important parameter of our study. We checked that the angles were constant to the experimental accuracy when varying the resting time of the pile (at constant vapor pressure) in the following experimental conditions: for granular samples humidified with heptane, the range of waiting time investigated was 40 s to 75 min, at relative vapor pressures of 0.72 and 0.90; for granular samples humidified with water (see Fig. 3b and its caption), waiting time for crosses in Figure 3b was approximately 1 min whereas it was approximately 5 h for triangles, for relative humidity ranging from 0.70 to 0.88. No significant variations were found, for waiting times differing by more than two orders of magnitude. We conclude from this study that our experimental conditions correspond to a steady-state for the angles.



(a)

(b)

**Fig. 3.** The maximum angle of stability $\theta_m$ (filled symbols) and the angle of repose $\theta_r$ (open symbols) as a function of the relative vapor pressure for (a) *n*-heptane and (b) water. Crosses in Figure 3b correspond to a first series of experiments performed without drying the beads between two consecutive measurements. Because the surface of the pile becomes more and more uneven as the degree of humidity increases (*cf.* Sect. 3.1), uncertainty on those points is larger than on the others, which we have obtained later following the experimental procedure detailed in the text. Error bars on $\theta_m$ in the usual experimental conditions are 1°. Note that error bars are larger for perfectly dry glass beads because of the existence of electrostatic forces due to friction: the dispersion in the values of $\theta_m$ can exceed 5°.

This result seems to be in contradiction with aging of the maximum angle of stability recently reported by Bocquet *et al.* [6]: they observed a logarithmic dependence of $\theta_m$ on waiting time, for waiting times ranging from 5 s to 2 h, for small glass beads in the presence of water vapor; moreover they showed that this aging effect strongly increases with the atmospheric degree of humidity. However, it is very likely that this discrepancy is related to the fact that the experimental procedures differ on a point which has major consequence on the characteristic time-scales of the problem: experiments in [6] are performed under atmospheric pressure, in the presence of an inert gas (air) in addition to water vapor, whereas our experiments are performed at low pressure, with water vapor only. Because transport of matter is the limiting process in these systems, we estimate the kinetics to be two orders of magnitude faster in our experimental conditions. Practically speaking, it means that our work corresponds to longer waiting times (expressed in units of the proper characteristic time-scale) than the ones investigated in the work by Bocquet *et al.*, preventing direct comparison of the results. However, this does not allow us to conclude whether aging effects simply do not exist in our experiments for an unknown reason, or whether the logarithmic regime observed by Bocquet *et al.* is followed by a saturation regime at long time. Further work needs to be done to clarify this important point.

### 3.2 Experimental results and discussion

Figure 3 shows the maximum angle of stability $\theta_\mathrm{m}$ and the angle of repose $\theta_\mathrm{r}$ (when measurable – see above) as a function of the relative vapor pressure expressed in percent, for $n$-heptane (Fig. 3a) and water (Fig. 3b). Humidifying the glass beads with heptane does not significantly change the angles up to a value of about 0.9 for $P_\mathrm{v}/P_\mathrm{sat}$ *i.e.* until very close to saturation; at relative vapor pressures larger than 0.90, a sudden and strong increase in the maximum angle of stability is observed while the angle after collapse of the pile is no longer meaningful. On the contrary, humidification with water leads to a gradual increase in $\theta_\mathrm{m}$ with the percentage of relative humidity, as well as a gradual (but less pronounced) decrease in $\theta_\mathrm{r}$, or equivalently a gradual increase in the so-called "relaxation angle" ($\theta_\mathrm{m} - \theta_\mathrm{r}$).

The slight decrease in the angle of repose observed for water might be the result of indirect humidity effects, related to wall effects the magnitude of which would be humidity-dependent. Furthermore, the angle of repose is the result of a dynamic process the analysis of which is very complex, even for dry granular media, and beyond the scope of this article. We will thus focus the discussion on the maximum angle of stability, which characterizes a critical state and thus the stability of the pile.

The $\theta_\mathrm{m}$ curves in Figure 3 first show the influence of humidity (in the broad sense) on the stability of a granular pile, and quantify it. Secondly they make clear the importance of the wetting properties of the fluid on the grains as different wetting properties lead to strongly different behaviors.

These observations prove that the quantitative change in the stability of the pile induced by humidity is intimately related, as expected, to the global liquid content on the one hand, and to the local spatial distribution of the liquid in the medium on the other hand. These quantities result from adsorption and capillary condensation of the vapor in the porous medium constituted by the packing of glass beads. Regarding the spatial distribution of the liquid at equilibrium, we can distinguish between two contributions: a thin wetting film adsorbed on the solid surface, and liquid bridges formed at the contact zones between grains. The liquid bridges generate capillary forces, and thus increase the stability of the pile through cohesive effects. The adsorbed film is not expected to alter static properties of the pile.

In order to fully interpret the quantitative variations that we experimentally observed for $\theta_\mathrm{m}$, either for heptane or water, we have to compute the capillary forces which result from the liquid bridges, before carrying out an analysis of the stability of the pile. Such an analysis has been done for instance by Halsey and Levine [5] by applying a failure criterion in the framework of continuum mechanics, and by Albert *et al.* [4] by applying a stability criterion based on geometric arguments.

In order to be able to compute the corresponding capillary forces, it is essential to have information on the actual size of the microscopic liquid bridges and also on the exact geometry of the solid surfaces in contact. In experiments on non-saturated humid granular media, the volume of the microscopic liquid bridges is not directly accessible experimentally as one controls a global parameter, which can be either the relative vapor pressure [6, this work], or the total amount of liquid added to the granular sample [3,7]. However, because we work with volatile liquids, we can use the Kelvin equation [9] to theoretically determine the radius of curvature $r$ of the liquid menisci at thermodynamic equilibrium, as a function of $P_\mathrm{v}/P_\mathrm{sat}$:

$$RT\,\mathrm{Log}\frac{P_\mathrm{v}}{P_\mathrm{sat}} = \frac{\gamma_\mathrm{lv} V_\mathrm{m}}{r}$$

where $\gamma_\mathrm{lv}$ is the surface tension between the liquid and the vapor, and $V_\mathrm{m}$ is the molar volume of the liquid (note that capillary condensation corresponds to menisci which are concave and thus have negative mean radii of curvature). Calculation of the capillary force between two surfaces can then be carried out by solving the Young and Laplace equation, at least numerically [12,13]. The results of direct measurements of the capillary force on Surface Force Apparati are also available for some volatile liquids, especially for heptane, and some particular geometries of the solid surfaces [14,15].

Yet, application to our particular problem is complicated by the fact that radii of curvature found in practice for menisci of liquids like water and heptane at room temperature are of nanoscopic orders of magnitude. Molar volume and surface tension of liquid heptane at 20 °C are respectively equal to 146.5 $\mathrm{cm}^3$ per mole and 19.9 $\mathrm{mN\ m}^{-1}$; inserting these values into the Kelvin equation, we find $r \approx -1.7$ nm at $P_\mathrm{v}/P_\mathrm{sat} = 0.5$, $r \approx -3$ nm at $P_\mathrm{v}/P_\mathrm{sat} = 0.7$, $r \approx -11$ nm at $P_\mathrm{v}/P_\mathrm{sat} = 0.9$, $r \approx -120$ nm at $P_\mathrm{v}/P_\mathrm{sat} = 0.99$. Water has a smaller molar volume (18.0 $\mathrm{cm}^3$ per mole) and a larger surface tension (72.75 $\mathrm{mN\ m}^{-1}$ for ultra-pure water) than heptane, which yields radii of curvature slightly smaller, but of the same order of magnitude. Obviously we can no longer ignore the microscopic roughness of the beads. This implies that we have to compute capillary forces between microscopic asperities having a complex geometry, and not simply between two spheres. A better knowledge of the beads surface is clearly needed. Experiments on macroscopically spherical grains having different microscopic surface roughness could also be useful.

In lack of precise information at the microscopic scale, assumptions on the geometry of the microscopic asperities in contact can be made. Interestingly, using a statistical model for the grains roughness, Halsey and Levine [5] theoretically predict three distinct regimes for the capillary force, and thus for the stability of the pile, as a function of the volume of liquid per contact. Unfortunately, the link between the parameters they introduce and the parameters we have access to experimentally is not straightforward, and the whole picture remains quite unclear at the time being[2]. In order to understand this point, it would be essential to have informations on the local spatial distribution of liquid in the medium. The relevance of this quantity is confirmed by the fact that different behaviors are observed for water and heptane: the macroscopic wetting angle for water on clean glass surface is very small, if not zero as for heptane, but molecular interactions with glass are significantly different for these two liquids as pointed out in Section 2.2, which leads to different wetting properties at the microscopic scale, in other words to different local spatial distribution of liquid in the medium.

## 4 Conclusion

In the first part of this paper we have presented the experimental method we designed in order to achieve an accurate control of the degree of humidity of a granular sample. The present method is based on the phenomena of adsorption and capillary condensation of a vapor in a porous medium; the thermodynamic equilibrium of the corresponding three-phase system is determined by one parameter, which is the relative vapor pressure. We have also underlined the main features of our method, in particular its versatility regarding its implementation on various systems.

In the second part of this paper, we have reported experimental results obtained on the maximum angle of stability and the angle of repose of a pile of glass beads as a function of its degree of humidity, for two different liquids, water and heptane. A strong influence of the wetting properties of the liquid on glass was observed on the maximum angle of stability. This is assuredly related to the local spatial distribution of liquid at equilibrium and it would be highly desirable to get information on the liquid distribution. In connection to this point, we have shown that humidity conditions in our experiments correspond to rather small liquid contents for which the microscopic surface roughness of the macroscopically spherical grains plays a major role. Experimental investigations on characterization of the surface roughness of the beads are currently under way to quantitatively understand its influence on the moisture-induced cohesion of the medium. Experiments on macroscopically spherical grains having different microscopic surface roughness are also under consideration.

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[2] Except in the simple regime, called "spherical regime" in [5], which corresponds to very large liquid bridges (larger than the ones we have in our experiments), for which the surface roughness no longer plays a significant role. This is the regime of interest in soil mechanics and it is well-known that the cohesiveness of a granular soil decreases when the liquid content increases [16]. This somewhat counter-intuitive result is confirmed by a rigorous computation of the capillary force as a function of the volume of the liquid bridges [12].