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Wet granular matter under vertical agitation

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Abstract

We have studied the effect of small amounts of liquid on the dynamic behaviour of a granular system consisting of spherical glass beads under vertical agitation. The critical acceleration Γ_{crit} at which fluidization sets in is found to increase strongly when liquid is added. It furthermore depends upon sample parameters such as the bead size and the shaking frequency. A simple model is proposed which accounts quantitatively for our results and suggests that fluidization by vertical agitation is mainly a surface phenomenon.

More than half of the world production of raw materials for the chemical industry comes as granulates. When granulates are handled and processed, the strongly nonlinear mechanical properties of these materials lead to clogging, avalanching, and other hard to control effects, which pose serious problems at all levels of engineering. A widely used approach for circumventing at least some of these problems is to fluidize the granulate by some external agitation. It then behaves rather like a liquid, such that material transport, e.g. through pipe systems, becomes well controllable.

One classical scheme, on which we want to concentrate here, is to oscillate the container vertically. At sufficiently high amplitude and frequency, the grains start to move in an erratic fashion, very much reminiscent of the molecular motion in a liquid, while the density of the granulate remains close to dense packing. It is easy to derive conditions for this type of fluidization to occur. If the vertical position of the container is given by $A \cos(2\pi vt)$, its maximum acceleration is $4\pi^2 A v^2$. If this is less than the acceleration of gravity, it is clear that the granulate will remain at rest with respect to the container, and no fluidization takes place. If we define $\Gamma := 4\pi^2 A v^2/g$, we can say that $\Gamma > 1$ is a necessary condition for fluidization. Experimentally, one finds that fluidization of dry granulates occurs at $\Gamma_{crit} \approx 1.2$ [1, 2]. It manifests itself in the onset of an irregular, relative motion of the grains, usually accompanied by some convective flow within the container.

As some liquid is added to the granulate, the mechanical properties of the latter dramatically change, due to the internal cohesion mediated by the capillary forces arising from liquid bridges between the grains [3–8]. While recent years have seen considerable progress in understanding the dynamics of dry granular materials [1, 2, 9-11], the physical mechanisms



Figure 1. The dependence of the critical acceleration for fluidization on the water content, at three different frequencies for $R = 138 \ \mu m$. Note the logarithmic scale.

underlying the properties of wet systems still pose many unanswered questions. We have thus investigated the dependence of Γ_{crit} upon the liquid content of a granular system under vertical agitation. To the best of our knowledge, the only systematic studies of fluidization of wet granulates concern fluidization by blowing air though the sample [12]; these are not immediately comparable to the experiments presented in the present paper.

A cylindrical glass container (inner diameter: 2.6 cm; height: 4 cm) was mounted onto an inductive shaker which could apply amplitudes between 1 μ m and 1 mm, at frequencies between 20 and 333 Hz. The container was filled to a height of about 3 cm with glass beads, with average radii ranging from 138 to 500 μ m. In order to prevent crystallization, the beads in each sample were chosen to have a few per cent of polydispersity. Controlled amounts of liquid were added and the lid closed in order to prevent any loss of liquid by evaporation. For the results presented here, we used water as the liquid, but similar results were obtained using nonane. We can therefore exclude the possibility that leaching of ions from the glass, and other side effects due to the specific properties of water [13], play a significant role. Before the container was mounted on the shaker, the sample was agitated by hand for some minutes in order to obtain a homogeneous distribution of liquid. At fixed agitation frequency, the amplitude *A* was increased until a relative motion of the particles was visible through the glass wall. This transition turned out to be rather well defined; small variations (<5%) in amplitude were decisive for fluidization to occur.

As one expects, we found the critical acceleration for fluidization, Γ_{crit} , to depend strongly on the water content W, which is defined as the volume of liquid added, divided by the total sample volume. Complete saturation corresponds to $W \approx 0.36$, if the packing fraction of the beads is 64%. Figure 1 shows this for three different frequencies, thereby demonstrating that there is also a dependence on frequency. We have found that all of our data can be well fitted with

$$\Gamma_{\rm crit} = \Gamma_0 + f(W) p(R) q(v) \tag{1}$$

where *R* is the radius of the glass beads, $\Gamma_0 = 1.2$ is the critical acceleration for the dry case, and f(0) = 0. In other words, we find that the dependences on moisture, bead size, and frequency factorize, at least within the accuracy of our experiments. In what follows, we will discuss the functions f(W), p(R), and q(v).

Let us first turn to the dependence of Γ_{crit} on the liquid content. As figure 1 shows, Γ_{crit} first increases strongly from its 'dry' value (\approx 1.2), but then stays roughly constant over about two orders of magnitude in liquid content. This is reminiscent of the behaviour of the capillary force, $F_{\rm B}$, exerted by a liquid bridge between adjacent beads. For ideally spherical beads, $F_{\rm B}$ is independent of the liquid volume in the bridge [14]. More precisely, it is equal to $2\pi R\gamma \cos\theta$, where γ is the surface tension of the liquid (72 mN m⁻¹ for water), and θ is the contact angle that the liquid makes with the bead surface.

For beads with a certain roughness, there is a transition from quasi-dry behaviour at small liquid content to fully developed liquid bridges, when there is enough liquid to fill the crevices and troughs on the bead surfaces [6, 15]. The attractive force due to the liquid bridge is then described by $F_{\rm B} = 2\pi R\gamma b(w) \cos \theta$, with b(0) = 0, and $b(w) \rightarrow 1$ for $w \ge w_0$. Depending on the type of roughness, *b* may acquire various forms. In an earlier paper [16], we have heuristically (but successfully) used

$$b(w) = \frac{w}{w + w_0} \tag{2}$$

with $w_0 := R\delta^2$ characterizing the roughness amplitude, δ [16], but this is not very sensitive to the precise form of b(W). It would be equally justified to use $b(w) = 1 - e^{w/w_0}$, or $b(w) = e^{-w_0/w}$. Each describes a transition from dry to wet around a certain bridge volume w_0 , corresponding to the amplitude and structure of the roughness. Clearly, w_0 may be converted into a global liquid content, W_0 . For glass beads as used in the present study, we have found $W_0 \approx 5 \times 10^{-4}$ [16].

The similarity to our results in figure 1 is striking. We observe a transition from the dry case ($\Gamma_{\rm crit} \approx 1.2$) to larger $\Gamma_{\rm crit}$ between $W_0 \approx 10^{-4}$ and 10^{-3} , followed by a wide regime of rather constant $\Gamma_{\rm crit}$. All of our data could be well fitted with

$$\Gamma_{\rm crit} = \Gamma_0 + \Gamma_1 e^{-W_0/W}.\tag{3}$$

This suggests that the extra acceleration required for fluidizing a wet bed of spherical beads seems to scale with the *force* of the capillary bridges between the beads. Although this might appear rather natural, it is in fact anything but trivial, since for a different fluidization scheme we have found that it is the *binding energy* (or the energy required to break a bridge) which rules the fluidization parameters [16]. This is not constant, but scales as $W^{1/3}$, in clear contrast to our data for Γ_{crit} . One may like to argue that since vertical agitation is a scheme based on acceleration, forces should be the paradigm quantities. These are linked to accelerations just by mass as a prefactor. We will have to discuss further, below, which mass is the relevant one in our system.

It turned out that when Γ_0 was fitted anew for every sample, the fit was not significantly better than with simply setting $\Gamma_0 = 1.2$ for all data sets. We thus kept this fixed value for all of the analysis. In this way we could extract, for all frequencies and bead radii investigated, the quantity $\Gamma_1 = p(R)q(\nu)$, which corresponds to the extra acceleration required for fluidizing a bed of ideally spherical beads, without roughness.

Let us now turn to the frequency dependence of Γ_{crit} . According to equation (1), this is contained in Γ_1 , which is plotted in figure 2. For large frequencies, Γ_1 acquires a constant value, while it approaches zero as $\nu \to 0$. For all samples investigated, our data could be well fitted with $\Gamma_1 = p(1 - e^{-\nu/\nu_0})$, which is shown as the solid curve in the figure. As a result, we obtain the high-frequency zero-roughness critical acceleration, p(R), which we will discuss further, below.

Quite remarkably, the parameter v_0 was about 62 Hz for all samples investigated, independent of bead radius. In searching for an explanation of this finding, it is instructive to contemplate the type of motion occurring when $\Gamma > 1$. In this case, the acceleration of the



Figure 2. A typical frequency dependence of the critical acceleration for fluidization. The fitting function (solid curve) is described in the text.

container periodically exceeds -g, such that the mass in the container (or at least parts of it) may temporarily detach from the bottom. If the vertical position of the container is $A\cos(2\pi vt)$, it is clear that for small frequencies, the amplitude A must be much larger in order to achieve the same acceleration Γ . Since the motion of the detached load is governed only by the acceleration of gravity, the consequence of increasing the amplitude is that the relative velocity, v_i , at which the load impacts again on the bottom of the container, increases drastically. A detailed calculation shows that it scales as $v_i \propto \Gamma g/\nu$. This impact is transferred as kinetic energy to the beads, and rapidly randomizes within the pile, thus increasing its granular temperature. Inserting numbers, one finds that below about 100 Hz, the impact velocity exceeds a few centimetres per second. In an earlier study [16], we have found that this is a typical velocity range (or granular temperature) at which fluidization by breaking liquid bridges within the pile takes place. In other words, the kinetic energy transferred into the sample is sufficient to reverse the liquid-induced condensation of the granulate. If one assumes that a certain velocity v_0 , corresponding to a certain granular temperature, is necessary for fluidization, then the above scaling law yields $\Gamma_1 \propto v_0 \nu/g$, in accordance with the linear regime observed for small ν . We are thus led to assigning this mechanism to the regime below about 100 Hz, marked by the decrease of Γ_1 in figure 2.

At larger frequencies, $\nu > \nu_0$, the energy transferred into the pile upon impact with the bottom of the container becomes negligible. In order to explain the constant regime found for frequencies well above ν_0 , we thus have to invoke a different mechanism. We thus consider now the case of large frequency and ideal spheres, for which we have simply

$$\Gamma_{\rm crit} = \Gamma_0 + p(R). \tag{4}$$

In contrast to what happens at the bottom of the container, the relative motion of the container side walls with respect to the glass beads represents a shear. It is tempting to consider this motion as a candidate for being the fluidization mechanism.

According to what has been suggested above, we estimate the forces involved in the interaction of the container walls with the glass beads, and the acceleration forces acting on the sample. The latter is given by

$$F_{\rm acc} = (\Gamma - \Gamma_0)\rho_{\rm s} V g \tag{5}$$



Figure 3. The dependence of the critical acceleration upon the radius of the glass beads. The solid curve represents the theoretical model $(p(R) \propto 1/R)$.

where $\rho_s = 1.60 \times 10^3 \text{ kg m}^{-3}$ is the weight density of the sample, and V is the sample volume considered. The capillary forces acting between the container walls and the glass beads are assumed to be similar to the force acting between two beads. The total capillary force is then

$$F_{\rm cap} = 2\pi R\gamma \cos\theta\rho_{\rm 2D}O\tag{6}$$

where ρ_{2D} is the two-dimensional density of the bridges formed between the container wall and the glass beads, and *O* is the total surface considered. For hexagonal close packing, $\rho_{2D} = (2\sqrt{3}R^2)^{-1} \approx 0.29/R^2$. For our random packing, ρ_{2D} is certainly smaller than that, but of the same order of magnitude. Relative motion between the glass pile and the container walls will take place if $F_{acc} \ge F_{cap}$. The condition for the onset of fluidization is then

$$\Gamma_{\rm crit} \approx \Gamma_0 + 0.29 \cos \theta \, \frac{2\pi\gamma}{\rho_{\rm s}g} \frac{O}{VR}.\tag{7}$$

This predicts the extra acceleration due to the addition of moisture, for ideal spheres and high frequency, to be inversely proportional to the bead radius. Figure 3 shows that this is well fulfilled for almost all of our samples. It is apparent that the smallest beads, in this respect, stand out from the data set. We have no explanation for why this sample of beads was apparently much more easily fluidized than the slightly larger beads, against the common trend observed otherwise. But it should be noted that they were purchased from a different supplier, who provided less thoroughly characterized specifications. If we leave this sample aside, the predicted behaviour is well fulfilled; the solid curve represents $p(R) = 570 \ \mu m/R$. For a cylindrical sample with radius r, V/O = r/2. In our case, we thus have $O/V = 154 \text{ m}^{-1}$. Furthermore, for water as the liquid, we have $\frac{2\pi\gamma}{\rho_{sg}} = 2.9 \times 10^{-5} \text{ m}^2$. Together with equation (7), this yields, for a closely packed surface layer, $0.29 \cos \theta = 0.128$, corresponding to a contact angle of about 60°. As mentioned above, the density of bridges at the container surface may be smaller than with close packing (i.e., the prefactor of the cosine may be smaller than 0.29), yielding smaller angles. In any case, the numbers that we obtain appear very reasonable, corroborating our model. It should finally be noted that the offset data point in figure 3 might be due to a larger contact angle, possibly due to some hydrophobic contamination. This can be resolved by independent contact angle measurements which are to be done in the future.

In conclusion, we have shown that fluidization of wet granular matter, at least for our model system, can be quite well understood in terms of the interaction of the pile with the walls of the

container. *A posteriori*, this appears quite natural, since in this scheme, the relative motion of the container with respect to the sample does not necessarily imply a deformation of the pile, in contrast to other fluidization schemes [16]. As a consequence, fluidization must start from the sample–container interface and must propagate inside. The bottom of the container exerts only a compressive motion and, since the sample itself is incompressible, is thus not capable of inducing any deformation at all. But it can 'radiate' kinetic energy into the sample if it hits it at sufficiently high speed. This is expected to occur at low agitation frequencies, as is in fact observed. The cylindrical walls of the container cannot transfer kinetic energy, but can exert a shear force by means of the liquid bridges between the wall and the sample. This is expected to lead to fluidization at just the critical accelerations observed in our experiment.

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