Fluidization of wet granular matter

Dimitrios Geromichalos, Mario Schedl, and Stephan Herminghaus

Applied Physics Lab., Ulm University, Ulm, D-89069, Germany

(Dated: November 10, 2018)

We have studied the effect of small amounts of added liquid on the dynamic behavior of a granular system consisting of spherical glass beads. The critical acceleration $\Gamma$ for fluidization is found to increase strongly when liquid is added. It furthermore depends on the bead radius, the height of the sample, and the shaking frequency. As our main result, we find that these dependencies can be presented in a factorized form. We present a simple model which accounts for most of our findings.

PACS numbers: 05.65.+b, 45.70.-n, 45.70.Mg

As it is generally known from everyday experience, the mechanical properties of a granular material change dramatically if some liquid is added. The main reason is the internal cohesion due to capillary forces arising from liquid bridges between the grains. While recent years have seen considerable progress in understanding the dynamics of dry granular materials, the physical mechanisms underlying the properties of wet systems still pose many unanswered questions.

In this article we consider granular matter in a container vibrated vertically with a certain frequency $\nu$ and an amplitude $A$. If this agitation is weak, the particles of the granulate remain fixed and do not move against each other. When the strength of the agitation is beyond the so-called fluidization threshold by enhancing $A$ or $\nu$, the granulate becomes fluidized. In this state the cohesiveness of the granulate is overcome. This enables a liquid-like flow of the granular matter, e.g. through pipe systems. Furthermore there starts in general a convective flow resembling to that of a fluid. The form of the vibration used in most experiments was a sinusoidal horizontal or, in particular, a vertical vibration.

In our experiments, we have studied the effect of capillary forces, which are characteristic for wet granular matter, on the fluidization behavior of the granulate. We mounted a small cylindrical glass container (diameter: 25 cm; height: 4 cm) filled at about three quarters with several samples composed of particles with radii from 138 $\mu$m to 500 $\mu$m, respectively. In order to prevent crystallization, the beads in each sample were chosen to be slightly polydisperse: the spread in bead size ranged from 10 to 20 %. We added controlled amounts of liquid and shook the sample by hand for some minutes in order to obtain a homogeneous distribution of liquid.

For the results presented here, we used water as the liquid, but similar results were obtained using nonane. We can therefore exclude that leaching of ions from the glass, and other side effects due to the specific properties of water, play a significant role.

We measured the fluidization threshold as follows: The shaking frequency $\nu$ was fixed at a certain value. The shaking amplitude $A$ was increased until a relative motion of the particles was visible through the glass wall. We used a rapidly increasing but inductive shaker. The container was shaken vertically with the granular matter consisting of glass beads on an inductive shaker. The form of the dependencies is described below.

**Dependence on water content:** The fluidization amplitude $\Gamma$ was found to depend strongly on the water content $W$, but also on the bead radius $R$, the sample height $H$ and the shaking frequency $\nu$ as follows:

$$\Gamma(W, H, R, \nu) = \Gamma_0(1 + G(W, H, R, \nu))$$

with $\Gamma_0 = 1.2$, being the value for the dry case. As it turned out, the function $G(W, H, R, \nu)$ can be factorized and written as

$$G(W, H, R, \nu) = f(W)h(H)p(R)q(\nu)$$

i.e. the contribution of each single parameter can be treated separately. This is the main result of this paper. The form of the dependencies is described below.
We can provide an explanation for this behavior in terms of the capillary forces which have a hysteretic character [12, 16, 17, 18].

For this we look at first at the capillary forces between two glass beads with a radius $R$. As it can be shown analytically [15], the capillary force between two perfectly smooth beads depends for sufficient small liquid contents only on the radius $R$, but not on $W$:

$$F_{\text{cap}} = 2\pi R \gamma$$

with the surface tension $\gamma$.

On the other hand it was found experimentally [19] that for very small water contents and beads with a rough surface, the capillary forces are not constant but decrease monotonically and smoothly to zero for $W \to 0$:

$$F_{\text{cap}} = 2\pi R \gamma f(w)$$

with the volume of a single capillary bridge $w$ and $f$ being a function which depends on the roughness of the sphere surfaces [1, 17].

The function $f(w)$ is for perfect spheres and complete wetting equal to one but for a rough surface, it tends to zero as $w \to 0$. The shape of $f(w)$ has been discussed in detail before [1]. Based on these considerations, it may be approximated by

$$f(w) = \left[ \frac{w}{w + w_0} \right]^\mu$$

with $w_0 := R\delta^2$ characterizing the roughness amplitude, $\delta$. For the exponent, we have $\mu = (2 - \chi)/(2 + \chi)$, where $\chi$ is the roughness exponent of the bead surface. For (typical) scratch and dig roughness as for our beads, $\chi$ is close to zero, and $\mu \approx 1$. This corresponds to the regime found by Hornbaker et al. [20].

At equilibrium, we assume that all of the liquid is in the bridges, $w = \frac{2\pi R^2 W}{\rho_{\text{cp}}}$, where $c$ is the coordination number of the network of bridges ($c \approx 6$) [20] and $\rho_p$ is the packing density of the spheres $\rho_p \approx 0.64$. Under dynamic conditions the typical bridge volume would be expected to be somewhat smaller than the equilibrium value. Our own measurements [21] suggest that $w = \alpha R^4 W$, where $\alpha \approx 0.25$ [22].

We assume that the starting condition for fluidization due to vertical shaking is that the force $F_0$ exerted from the vibration is big enough to break the capillary bridges in at least one horizontal plane in the pile. Here the vibrational force is given directly by Newton’s law:

$$F_0 = (\Gamma - \Gamma_0)Mg$$

with the mass of the granular sample $M$ and $\Gamma_0 := \Gamma(W = 0) \approx 1.2$ [7]. The mass of the (cylindrical) pile is given by

$$M = \rho_g \rho_{\text{cp}} HB$$

with the density of the bead material (glass) $\rho_g$, the random close packing density for beads $\rho_{\text{cp}} \approx 0.64$ [23], the height of the cylindrical sample $H$ and its base area $B$.

The expression for the force exerted from the capillaries in the relevant horizontal plane is

$$F_{\text{pl}} = \frac{B}{R^2} \rho_{\text{cp}} 2\pi R \gamma f(w)$$

with the two-dimensional 'density' $\rho_{\text{cp}}$ of the bridges which have to break.

From equating (3) and (1), one gets

$$\Gamma(W) = \Gamma_0 \left( 1 + \tilde{c} f(w) \frac{w}{HR} \right)$$

with $\tilde{c} = \frac{2\gamma \rho_{\text{cp}}}{\Gamma_0 \rho_g \rho_{\text{cp}} g}$. As it can be seen in Fig. 1 this can be used quite well to describe the $W$-dependence of $\Gamma$. $\tilde{c}$ was a fitting parameter which was also a function of the bead radius $R$ and the shaking frequency $\nu$ (see below).

The function $f(w)$ connects the bridge formation directly to the roughness. From $W_0 \approx 0.05\%$ (see Fig. 11) we get $\delta \approx 500\mu m$, which is similar to the peak-to-peak roughness obtained from the inspection of the beads by atomic force microscopy [28]. As it can be seen in Fig. 11, the dependence of the fluidization acceleration on the liquid content is described quite well by this theory.

A further prediction is the dependence of $\Gamma$ on $H$, which we could confirm at least qualitatively in additional control measurements at $0.25 mm \leq H \leq 35 mm$ with $R = 138\mu m$ and $W = 0.13\%$ as is shown in Fig. 4. We could fit the values plotted in Fig. 4 quite well with

$$\Gamma(W, H) = \Gamma_0 \left( 1 + \tilde{c} f(w) \frac{1}{H} \left( \frac{1}{H} + \frac{1}{H_\infty} \right) \right)$$

FIG. 1: Dependence of the fluidization acceleration for different frequencies for $R = 0.138 mm$ on the water content. The fitting functions are described in the text.
The finite limiting value $H_c$ may be understood as follows. One ruptured horizontal plane may be sufficient to fluidize the surrounding medium only to a certain vertical distance. Hence in samples with large $H$, the initial fluidization occurs in horizontal layers with a thickness $H_c < H$ such that $\Gamma$ becomes independent of $H$. We were able observe this behavior by direct optical monitoring.

Dependence of the fluidization acceleration on the height $H$ of the granular pile. The fitting function is described in the text ($H_c \approx 4\text{mm}$).

The deviation of the measured values from the prediction according to Eq. 5 gets larger for smaller and smaller bead radii. Hence we set $R_0 = 0.5\text{mm}$ as the value where 5 and 6 yield the same value for $\Gamma$. Therewith we obtained a $\rho_{2D}$ ranging between 3 and 4, which is a reasonable result.

A possible explanation for $\beta \neq 1$ is that the particles of a wet granulate tend more and more to form (statistical and temporary) clusters the smaller they are. Since these clusters may behave like single bigger particles \cite{24, 25}, $\Gamma(W,R)$ should be especially for small $R$ smaller than the fluidization acceleration expected from Eq. 5. This is consistent with $\beta < 1$, as observed.

Frequency dependence:
The fluidization amplitude turned out to depend also on the shaking frequency $\nu$. In general we observed that $\Gamma$ was a concave and monotonous rising function of $\nu$. This frequency dependence was most distinctive for small beads and at high liquid contents, although it did not depend very strongly on $W$ for the bigger $R$. Some sample curves of this dependence are shown in Fig. 4.

The frequency dependence could be fitted with the function

$$
\Gamma(\nu) = \Gamma_0 \left(1 + \frac{C_1 \bar{c} f(w)}{R^{\beta} R_0^{1-\beta}} \left(\frac{1}{H} + \frac{1}{H_c}\right)\right)
$$

with

$$
\zeta := \frac{C_1 \bar{c} f(w)}{R^{\beta} R_0^{1-\beta}} \left(\frac{1}{H} + \frac{1}{H_c}\right)
$$

We were able to describe the data with

$$
\Gamma(W,R) = \Gamma_0 \left(1 + \frac{\bar{c} f(w)}{R^{\beta} R_0^{1-\beta}} \left(\frac{1}{H} + \frac{1}{H_c}\right)\right)
$$

FIG. 2: Dependence of the fluidization acceleration on the height $H$ of the granular pile. The fitting function is described in the text ($H_c \approx 4\text{mm}$).

FIG. 3: Bead radius dependence of $\Delta \Gamma := \Gamma(W,R) - \Gamma_0$ for different frequencies (circles and squares). The asterisks represent the radius dependence of the slope $\zeta$ (described in the text; see Fig. 4). The solid line is proportional to $R^{-\beta}$ with $\beta = 0.5$, the dotted line is proportional to $R^{-1}$.

FIG. 4: Frequency dependence of the fluidization acceleration for different bead radii for $W = 0.3\%$ where $\Gamma(W)$ has already saturated. The fitting functions are described in the text.
(see Eq. 1) for the applied large frequencies with the fitting parameter \( \zeta \) (and accordingly \( C_1 \)), as is shown in Fig. 1. The value of \( \nu_0 \) turned out to be of the order of 1 Hz. The frequency dependence was more pronounced for the small beads, i.e., \( \zeta \) was a monotonous falling function of \( \nu \). It emerged that \( \zeta(R) \) was roughly proportional to \( R^{-\beta} \) (see Fig. 3).

We will now provide a qualitative explanation for this behavior. The amplitudes \( A \) corresponding to the frequencies \( \nu \) were only about 1 \( \mu m \) for the higher \( \nu \) which is much smaller than the radii of the glass beads. This means that a translational motion of the glass beads is more and more difficult for higher frequencies and occurs only through many-particle collisions. Thus the rotational degree of freedom becomes more and more important for the higher frequencies which leads to a concave form of \( \Gamma(\nu) \), since rotational motion is not impeded significantly by small \( A \) if the particles are perfect spheres 30. We have already mentioned the occurrence of particle clusters in the wet granulate. Since these clusters are irregular, they cannot easily rotate. Hence one expects a more pronounced frequency dependence, i.e., a bigger \( \zeta \), for the smaller beads where the assumed clusters are more pronounced (see above). This is in qualitative agreement with our measurements.

The rotational motion of non-spherical particles in a granulate consisting of them is much more difficult (as for the case of clusters described above) and depends clearly on \( A \). Hence we conducted also some experiments with cubes 31 and other particles like cylinders, river sand and foraminiferae in order to check our assumption. For the case of cubic particles 32 we observed indeed a very different behavior: The increase of \( \Gamma(\nu) \) was nearly linear and quite strong 33. The \( \nu \)-dependence of \( \Gamma \) was for the other non-spherical particles also linear but not as pronounced as for the cubical case. The dependence \( \zeta_1 := \frac{d\Gamma(\nu)}{d\nu} \) in the case of cylinders was very close to that for the glass beads, while the cases of river sand and foraminiferae lied in-between the observed extremes given by the spheres and the cubes. Concluding it can be said that \( \zeta_1 \) increased with the ‘roughness’ (or ‘edginess’) of the particles. Furthermore \( \zeta_1 \) was a monotonous rising function of \( W \) which is also consistent with our explanation of the \( \nu \)-dependence 34.

It is interesting to note that for the beads, with a roughness exponent \( \chi \approx 0 \), we get \( \Gamma - \Gamma_0 \propto \ln(\nu) \), while for edgy grains, with \( \chi \approx 1 \) we find \( \Gamma - \Gamma_0 \propto \nu^1 \). Whether this finding represents a coincidence or bears some fundamental physics remains to be investigated.

Inspiring discussions with M. Schulz are gratefully acknowledged. The authors thank the German Science Foundation for financial support within the Priority Program ‘Wetting and Structure Formation at Interfaces’.

[1] T. Mikami, H. Kamiya, and M. Horio, Chem. Eng. Sci. 53, 1927 (1998).
[2] J. N. Israelevich, Intermolecular and Surface Forces (Academic, London, GB, 1991), 2nd ed.
[3] D. J. Hornbaker, R. Albert, I. Albert, A.-L. Barabasi, and P. Schiffer, Nature 387, 765 (1997).
[4] T. C. Halsey and A. J. Levine, Phys. Rev. Lett. 80, 3141 (1998).
[5] L. Bocquet, E. Charlaix, S. Ciliberto, and J. Crassous, Nature 396, 735 (1998).
[6] N. Fraysse, H. Thomé, and L. Petit, Eur. Phys. J. B 11, 615 (1999).
[7] G. H. Ristow, Pattern Formation in Granular Materials (Springer, Berlin, Germany, 2000).
[8] H. M. Jaeger, S. R. Nagel, and R. P. Behringer, Rev. Mod. Phys. 68, 1259 (1996).
[9] L. P. Kadanoff, Rev. Mod. Phys. 71, 435 (1999).
[10] P. K. Dixon and D. J. Durian, Phys. Rev. Lett. 90, 184302 (2003).
[11] J. Duran, Sands, Powders, and Grains (Springer Verlag, New York, 2000).
[12] G. H. Ristow, Europhys. Lett. 40, 625 (1997).
[13] G. Metcalfe, S. G. K. Tennakoon, L. Kondic, D. G. Schaeffer, and R. P. Behringer, Phys. Rev. Lett. 65, 031302 (2002).
[14] N. Olivi-Tran, N. Fraysse, P. Girard, M. Ramonda, and D. Chatain, Eur. Phys. J. B 23, 217 (2002).
[15] C. D. Willett, M. J. Adams, S. A. Johnson, and J. P. K. Seville, Langmuir 16, 9396 (2000).
[16] J. Crassous, E. Charlaix, and J.-L. Loubet, Europhys. Lett. 28, 37 (1994).
[17] L. Zitzler, S. Herminghaus, and F. Mugele, Phys. Rev. B 66, 155436 (2002).
[18] M. Schulz, B. M. Schulz, and S. Herminghaus, submitted to Phys. Rev. E (2003).
[19] H. Rumpf, Agglomeration p. 379 (Interscience Publishers, 1962).
[20] D. Geromichalos, M. M. Kohonen, M. Scheel, and S. Herminghaus, to be submitted (2003).
[21] M. M. Kohonen, D. Geromichalos, and S. Herminghaus, manuscript in preparation (2003).
[22] D. Geromichalos, M. M. Kohonen, F. Mugele, and S. Herminghaus, Phys. Rev. Lett. 90, 168702 (2003).
[23] D. Scott, Nature 188, 908 (1960).
[24] A. Samadani and A. Kudrolli, Phys. Rev. Lett. 85, 5102 (2000).
[25] A. Samadani and A. Kudrolli, Phys. Rev. E 64, 051301 (2001).
[26] S. J. R. Simons, J. P. K. Seville, and M. J. Adams, Sixth International Symposium on Agglomeration, Nagoya, Japan p. 117 (1993).
[27] The only fluidization experiments which are mentioned in literature and concern wet granular matter were done by blowing air through the granulate 22 and are therefore not immediately comparable to the experiments presented in this paper.
[28] Furthermore we should add that the capillary forces depend strongly on the particle shape as we saw when we did similar experiments with NaCl-cubes and nonane as wetting liquid and found no saturation.
[29] For $R < R_0$ the measured fluidization acceleration according is smaller than the value obtained from $\beta = 0.5 < 1$. Assuming that particles with $R \geq R_0$ do not form clusters (explained in the text), one would expect $\beta = 1$ for that case.

[30] One important parameter for this dependence is the bead roughness.

[31] We used NaCl with a size of 300$\mu$m; as wetting liquid we used nonane. (M. Scheel, D. Geromichalos, S. Herminghaus; manuscript in preparation)

[32] $W = 0.3\%$; $50\,Hz \leq \nu \leq 333\,Hz$

[33] We observed a linear increase from $\Gamma \approx 2$ at $50\,Hz$ to $\Gamma \approx 8$ at $333\,Hz$.

[34] The liquid leads to a formation of clusters. This causes an additional obstruction of rotational motions and consequently an increase of the fluidization acceleration.