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The shear modulus of wet granular matter

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Abstract – The strength of different wet granular materials is investigated as a function of the liquid volume fraction by measuring the elastic shear modulus, G' . We show that the optimum strength is achieved at a very low liquid volume fraction of 1–3%. Surprisingly we find that the macroscopic strength of different wet granular materials depends with a power of $2/3$ on the microscopic elastic modulus of the individual grains, with a power of $-1/3$ on the radius, and with a power of $1/3$ on the surface tension. This can be explained by assuming that the attractive capillary force between two grains deforms the grains elastically, yielding a “spring constant” for further deformation. Averaging over many grain-grain orientations allows us to predict the macroscopic shear modulus in excellent agreement with our experiments.

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Physicists have for quite some time been fascinated by the far-from-equilibrium properties of granular systems and the phenomena they give rise to. Tremendous activity within the field of granular research gives proof of this [1–3]. Furthermore the properties of granular materials are of huge importance to engineers and it is estimated that about 10% of all energy consumption on Earth is spent on the handling of granular materials [4]. In spite of the huge interest of both scientists and engineers the properties of granular systems are still imperfectly understood [5].

One of the most spectacular and fascinating properties of granular materials is how the addition of a small amount of fluid dramatically changes the macroscopic properties of the material. Just a bit of water turns a boring pile of dry sand into a spectacular sandcastle [6–8] while too much water will destabilize the material, as observed in landslides [9]. A favorite tool for studying the strength of such moist granular systems has been the rotating drum experiment, where the material is put into a slowly rotating drum where the angles of repose and avalanche are recorded as a measure of material strength [8]. This type of experiment by its nature limits one to examining relatively weak, non compacted materials, and does not allow for studying materials of a strength suitable for building sandcastles for instance. In a recent paper Fournier *et al.* imposed oscillatory shear deformation to wet granular materials in a pressure-driven shear cell [10]. They recorded the hysteresis pressure as a measure of the material yield stress as a function of liquid volume

fraction. Because of considerable scatter, the best one can say about the liquid volume fraction at which maximum strength occurs is that it is between 0.1% and 5% and the issue of what volume fraction of water gives the highest elastic strength of the granular assembly remains largely unresolved.

In this letter we measure the elastic modulus as an expression of material strength and give an answer to the question of what the optimum liquid volume fraction is, and provide a quantitative model that is able to account not only for the optimum, but also for the value of the elastic modulus. We use a conventional rheometer (Rheologica, Stresstech) to measure the elastic shear modulus, G' , of different partially saturated granular materials as a function of the liquid volume fraction. G' is related to the macroscopic Young's modulus, E_{mac} , by $E_{mac} = 2(1 + \nu)G' \approx 3G'$, ν being Poisson's ratio¹. The grains used in the experiments are; sand (from Fontainebleau, sieved to have a radius between 90 and 110 μm), spherical glass beads of 25 and 100 μm radius, polystyrene beads of 100 μm radius (Dynoseeds, from Microbeads), and PMMA spheres of a 3 μm radius (Calibre, from Microbeads). All beads are thoroughly rinsed and dried before use. The fluids used are ultra pure water and silicone oil (Rhodorsil from CRC Industries, France). The surface tensions of the fluids in the presence

¹In this paper we assume $\nu = 0.5$ which corresponds to that of an incompressible medium. If we choose $\nu = 0.3$ which corresponds to steel, the numerical values of our calculations change by about 4%, so that we are not very sensitive to the exact value of ν .

of beads are determined using the Du Nuoy ring method. For water it is found to be 72 mN/m and 20 mN/m for silicone oil. After preparing a mixture of beads and fluid, a small quantity of this mixture is put in a vane-in-cup geometry in the rheometer. For the almost completely dry and almost completely wet mixtures the yield stress is quite low and the grains reach something resembling a close packing when poured into the rheometer geometry, but for the intermediate liquid volume fractions the yield stress is quite high and the material does not compact under its own weight giving much lower grain filling fractions. In order to compare the elastic moduli at different liquid volume fractions and to get results not depending strongly on how the material is loaded into the cup, the material is after loading compacted by dropping a small thumper (12.5 mm in radius, 40 mm in height and with a 30 g mass) from a small height (about 10 mm) at least 100 times. We found that this ensures a reproducible compaction (a filling fraction of about 0.63 ± 0.01) for all liquid volume fractions and for each loading of the sample. This observation and the fact that the yield stresses of completely dry and wet materials were too low for us to measure their elastic moduli demonstrates that the compaction of the materials does not squeeze the beads together in the cup. Since the capillary rise ($h = 2\gamma \cos \theta / \rho g r$ [11]) in our samples is at least 15 cm and thus much higher than our filling height, we expect that surface tension along with mechanical mixing prior to the experiment prevents drainage and other inhomogeneities of the distribution of liquid among the grains.

The modulus G' is subsequently measured at a frequency of 1 Hz. We use 1 Hz since this allows for rapid measurements; we verified that G' does not depend on frequency for frequencies between 10^{-2} and 10 Hz, the frequencies readily accessible on the rheometer (see fig. 1). To ensure that the measurements probe the linear, elastic response, the shear strain is continuously increased during the course of one experiment and the value of G' is averaged over the linear regime (see fig. 1). At least ten such measurements are performed for each material composition and for each volume fraction. The vane-in-cup geometry used for most of the experiments consists of four vanes of 10 mm in height and 5 mm in width attached to a rod 4 mm in radius, thus giving a total inner radius of 9 mm and a cup outer radius of 13 mm with a 10 mm strip of sandpaper glued on at the same height as the vanes². This gives a 4 mm gap corresponding to at least 20 bead diameters. To ensure that this gap size is sufficient to avoid confinement effects we checked that a vane-in-cup geometry with a much larger outer radius —22.5 mm (corresponding to a gap of 13.5 mm)— gave results identical to the smaller one

²One needs the above values for the inner and outer radii, r_i and r_o , and height, h , in order to convert the torque, τ , and angular deformation, $\Delta\theta$, measured by the rheometer into a value for G' : $r_i \Delta\theta = \int_{r_i}^{r_o} \gamma dr = \int_{r_i}^{r_o} \sigma / G' dr = \int_{r_i}^{r_o} \tau / 2\pi h r^2 G' dr \Rightarrow G' = (\tau / \Delta\theta) \cdot (r_i^{-1} - r_o^{-1}) / 2\pi h r_i$.

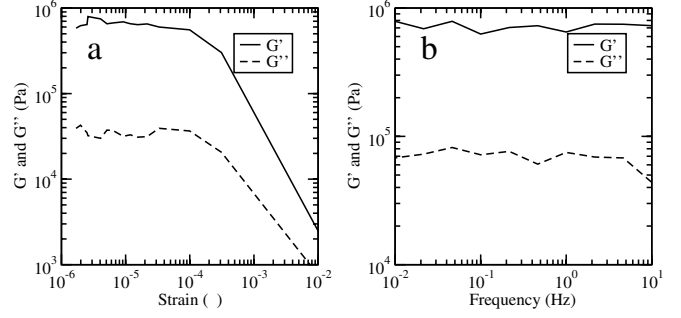


Fig. 1: A typical measurement of the elastic storage and loss moduli G' and G'' as a function of strain at a frequency of 1 Hz (a), and as function of frequency at a fixed strain of $3 \cdot 10^{-6}$ (b). Since G' and G'' are seen to be independent of frequency between 0.01 and 10 Hz, all experiments were done at 1 Hz. Since G' and G'' are seen to be roughly constant for strains between 10^{-6} and 10^{-4} , the average value of G' in this regime is taken for each experiment. The fact that G'' is more than an order of magnitude smaller than G' shows that this regime is one of elastic deformation and without flow. Using a value of Poisson's ratio, G' can be converted into Young's modulus and the bulk modulus so that the exact material deformation to any type of load can be computed. Materials normally have some finite deformation they can sustain before they fail, and knowing both G' and that critical deformation allows one to predict if the material will fail under a given load or not. In (a) the value of G' is reduced by one order of magnitude when the strain is about 10^{-3} , which is a reasonable value to choose for the critical strain. This material can thus sustain a tangential load of about 1 kPa before fracturing, which compares well with the fact that weights on the order of grams are needed to deform a cube centimeter of sand.

(fig. 2). In all cases the distance to the bottom of the cup (which was smooth to allow as much slip as possible) was 10 mm and the total filling height about 30 mm.

Figure 2 depicts the results for a variety of bead materials and fluids. We find that wet sand has a shear modulus similar to that of spherical glass beads, while that of spherical polystyrene beads is much smaller: the latter has a G' that is more than one order of magnitude lower. Apart from this difference in absolute strength the curves show a similar dependence on the liquid volume fraction.

To understand this, we need to look in detail at what happens when one adds a small volume of fluid to a stack of spherical grains. The surface tension of the fluid pulls it into small bridges connecting individual beads (fig. 3). The attractive capillary force between two spheres is $F_{cap} = -\pi r_1^2 \Delta p + 2\pi r_1 \gamma = -\pi r_1^2 C \gamma + 2\pi r_1 \gamma$, where γ is the surface tension and C and r_1 are the curvature and the radius of the bridge, respectively (fig. 3(a)) [12,13]. The curvature is a function of R , r_1 and the separation between beads, d , but for $d = 0$ and $r_1 \ll R$, one can approximate $C \approx -1/r_2 \approx -2R/r_1^2$ so that the force is $F_{cap} \approx 2\pi R \gamma$.

In practice it is well known that surface roughness and finite sphere separations mean that the actual force will

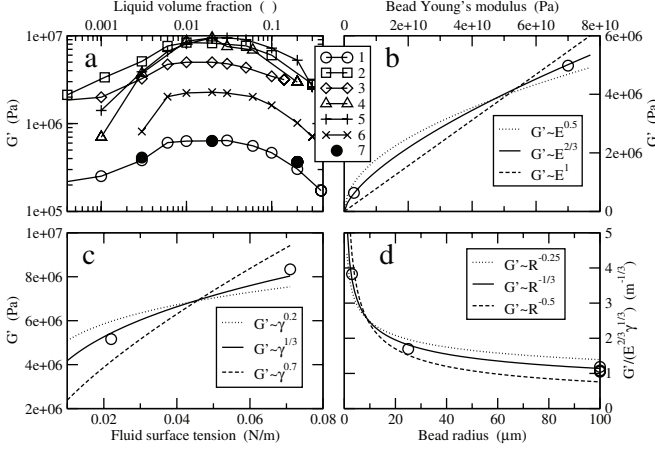


Fig. 2: (a) Measured elastic shear modulus G' as function of liquid volume fraction for different granular materials; 1: 100 μm polystyrene beads and silicone oil, 2: 100 μm glass beads and water, 3: 100 μm glass beads and silicone oil, 4: 100 μm sand and water, 5: 25 μm glass beads and silicone oil, 6: 3 μm PMMA beads and silicone oil, 7: 100 μm polystyrene beads and silicone oil in a larger-gap vane-in-cup geometry. Apart from the absolute value of G' , which differs more than an order of magnitude, the curves for different materials are seen to have similar shapes. Most measurements were performed in a geometry with a 4 mm gap, but measurements with a gap of 13.5 mm give identical results as seen in series 7. Note that none of the materials are completely wet or dry. Indeed, for completely wet materials and materials with too low a volume fraction, the elastic strength is too low to be measured by the rheometer. (b), (c): The dependence of G' at a 1% liquid volume fraction on Young's modulus of the bead material and the surface tension of the liquid, respectively. (d) Rescaling G' by the relations found in sub figures (b) and (c), we plot $G'/(E^{2/3}\gamma^{1/3})$ as a function of the bead radius.

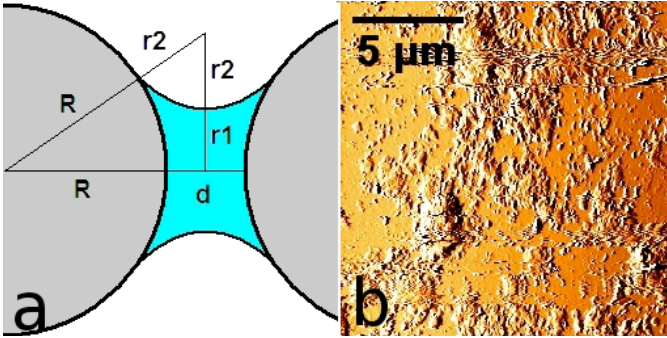


Fig. 3: (a) Schematic of a liquid bridge. The distance between beads has been exaggerated for illustrative purposes. (b) An Atomic Force Microscopy (AFM) picture of the surface of a 100 μm glass bead. The height of the surface roughness is on the order of 1 μm .

not be independent of the bridge volume [14]. While the above formula is a very good approximation at intermediate volume fractions, it overestimates the strength at low ones where the fluid is trapped in the surface roughness and at high ones where single bridges merge into larger

aggregates and loose strength. To allow for this dependence, we multiply the simplified bridge force equation with a factor between 0 and 1, depending on the liquid volume fraction: $F_{cap} = 2\pi R\gamma f(V_f)$. From AFM pictures of the bead surfaces (fig. 3(b)) and fluorescence pictures of the liquid bridges (fig. 5) this factor can be computed as described below.

The bridge force will suck the beads together and deform them elastically until balanced by the elastic response of the beads. The relation between force F , compression δ , of two identical spheres of radius R , and Young's modulus E is given by Hertzian response theory [15]: $F_H = -\frac{4\sqrt{2}}{9}R^{1/2}E\delta^{3/2}$. Linearizing the total force around the new equilibrium position where $F_{cap} + F_H = 0$, gives a spring constant against further displacement of the beads:

$$F_{tot} = -\frac{4\sqrt{2}}{9}R^{1/2}E((\delta_{eq} + \Delta\delta)^{3/2} - \delta_{eq}^{3/2}) \quad (1)$$

$$\approx -\frac{2\sqrt{2}}{3}R^{1/2}E\delta_{eq}^{1/2}\Delta\delta \quad (2)$$

$$\approx -\left(\frac{\pi}{24}\right)^{1/3}R^{2/3}E^{2/3}\gamma^{1/3}f(V_f)^{1/3}\Delta\delta \equiv -k\Delta\delta. \quad (3)$$

To compute how this spring constant results in G' , one needs to average over all sphere-sphere bond orientations with respect to the shearing plane and to the shearing direction. To compute the macroscopic Young's modulus (E_{mac}) however, one needs only average over bond orientations with respect to the stress plane, which is much simpler. G' can subsequently be found from E_{mac} via $E_{mac} = 3G'$. Let Δl be the extension of the material normal to the stress plane and F_e the extensional stress. Then $E_{mac} \equiv \frac{F_e/A}{\Delta l/l}$, where A is the area where the stress is applied and $\Delta l/l$ is the relative elongation. Consider a sphere-sphere bond of length $2R$ oriented with polar angle θ to the stress plane, and azimuthal angle ϕ . Then $\Delta\delta = \Delta l \sin \theta$, $l = 2R \sin \theta$ and $F_e = F \sin \theta$. If one now takes the area where the force is applied to be $(2R)^2$, one gets

$$E_{mac} = \frac{F_e/A}{\Delta l/l} = \frac{F \sin^3 \theta}{2R \Delta\delta} = \frac{k}{2R} \sin^3 \theta. \quad (4)$$

And averaging over all angles, θ and ϕ ,

$$\langle E_{mac} \rangle = \frac{k}{\pi R} \int_0^{\pi/2} \sin^3 \theta \cos \theta d\theta = \frac{k}{4\pi R} \quad (5)$$

leading to

$$\langle G' \rangle = \frac{k}{12\pi R} \approx 0.054 R^{-1/3} E^{2/3} \gamma^{1/3} f(V_f)^{1/3}. \quad (6)$$

This simple formula directly explains why moist polystyrene beads have a much smaller G' than moist sand or glass beads. This is because Young's modulus of sand is like that of glass, namely 68 GPa, while that of polystyrene and PMMA is only 3 GPa [16]. Furthermore the surface tension of water is 72 mN/m, while that of oil

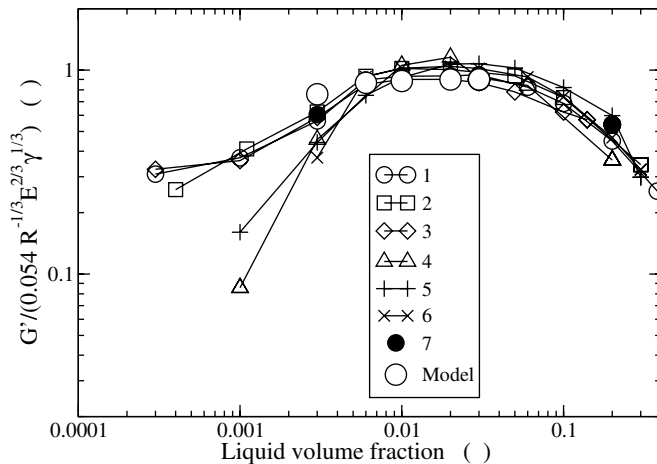


Fig. 4: Master curve of G' for several materials (see the caption of fig. 2 for the legends). The curves for the different materials have been rescaled using: $G' \rightarrow G'/(0.054 R^{-1/3} E^{2/3} \gamma^{1/3})$, where R is the bead radius, E Young's modulus of the beads material and γ the surface tension of the fluid. The theoretical prediction has been computed directly from the known values of these parameters and from the measurements of the liquid bridge diameter for each volume fraction. It contains no fitting parameters.

is 20 mN/m; the difference in the measured value of G' between sand and water on the one side and polystyrene beads and silicone oil on the other is indeed on the order of $(68/3)^{2/3}(72/20)^{1/3} \approx 12$. This therefore gives a quantitative explanation for the one order of magnitude difference between glass and polystyrene beads observed in fig. 2. Systematically and independently varying the grain size (a factor of 30), fluid surface tension (a factor of 4), and the Young's modulus of the grain material (a factor of 20), we find that the measured elastic moduli collapse onto a single master curve when rescaled in the above way (fig. 4).

The strength of all the grain/liquid systems is thus seen to depend strongly on the liquid volume fraction, with a maximum strength at about 2% of liquid, independently of the system details (fig. 4). Because of the relative low scatter in the data of this figure the liquid volume fraction for maximum strength can be limited to the range of 1–3% which is a significant improvement compared to the previous range of 0.1–5% [10]. Furthermore we demonstrate that the behavior of rough sand is like that of spherical glass beads which was an open question. It is perhaps surprising that our experiments and those of [10] give similar curves for strength as a function of volume fraction. Strength is maximised when the liquid bridge attraction peaks however, which should not depend on the measurement method.

What happens when the bridge volumes grow is well understood [14]. At very low volume fractions, most of the fluid is trapped in the surface roughness, and the bridge force is dominated not by the curvature of the sphere, but by the local roughness. At higher volume fractions,

a significant fraction of the fluid is still caught in the surface roughness but the bridge force is dominated by the curvature of the spheres. At even higher volume fractions, the bridges start merging and loose strength. From fig. 4 it is seen that some materials loose their strength faster as the volume fraction is decreased below about 1%. This is very reasonable since the fraction of fluid caught in the surface roughness increases as the liquid volume fraction is lowered and these materials (4 : 100 μm sand and water, 5 : 25 μm glass beads and silicone oil, and 6 : 3 μm PMMA beads and silicone oil) have higher surface areas per unit volume of material.

Since the elastic moduli of the materials in fig. 4 are rescaled by $G' \rightarrow G'/(0.054 R^{-1/3} E^{2/3} \gamma^{1/3})$, both the experimental data and the prediction are actually showing the shape of $f(V_f)^{1/3} = (F_{cap}/2\pi R\gamma)^{1/3}$. To predict not only how the modulus of a material depends on the bead material, the bead size, and the fluid surface tension, but also on the liquid volume fraction one needs to predict how $f(V_f)$ varies. To get an accurate value of the bridge force one needs to integrate the Laplace equation in cylindrical symmetry: $\Delta p = \gamma(1/r + 1/r')$, where r and r' are the principal radii of curvature [11]. As boundary conditions for this equation, one needs to know the bridge radius, the sphere radius, and the sphere separation³. The sphere radii are known from standard optical microscopy, the sphere separation is taken to be equal to the height of the surface roughness which was found with an AFM (fig. 3), and the bridge radii as a function of volume fraction was found with fluorescence microscopy (fig. 5). Using these values as boundary conditions, the Laplace equation is numerically integrated to give the model prediction in fig. 4. As this calculation assumes that bridges do not merge and needs the bridge radii as input values, predictions can only be made of course in the regime where bridge radii are optically measurable and bridges have not yet merged. Unfortunately single bridges can be observed only between volume fractions of 0.3% and 3% (fig. 5), which limits the theoretical prediction to this range. As seen in fig. 4, the prediction is very good between volume fractions of 0.6% and 3%, while it is too big at 0.3%.

A second effect of the separation between beads is that bridges cannot exist below a critical volume. The number of bridges has been demonstrated to increase abruptly with volume fraction (from about one per sphere to about six) at a volume fraction of about 0.2% [10]. This change in bridge number is not included in our model, and this may be the reason why for the lowest volume fraction the model slightly overestimates the modulus.

In sum, we investigated the impact of the amount of fluid and composite material on the strength of a partially saturated granular material and found that the optimum is

³In principle one also needs to know the contact angle, but since the contact angles in our mixtures are always very low and since the resulting force depends very weakly on the contact angles, they are all assumed to be zero.

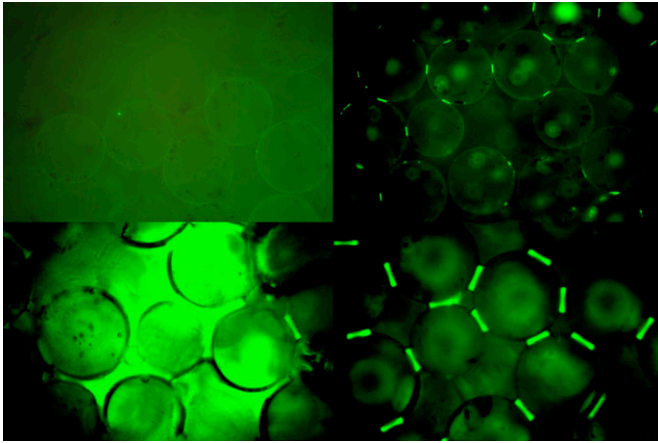


Fig. 5: The liquid bridges between beads as seen in fluorescence microscopy. From the upper left corner and clockwise, volume fractions of 0.1%, 0.3%, 3% and 6%, respectively. At a volume fraction of 0.1% the fluorescence of the fluid caught in the surface roughness is seen to dominate that of the fluid in the bridges. At a volume fraction of 6% a significant number of the bridges are merged into large aggregates. The bridges consist of water with added fluoresceine and the interstitial air has been replaced by an oil with an index of refraction matching that of the glass beads.

achieved at a liquid volume fraction as small as 1–3%. We present a very simple model that successfully describes the elastic modulus of wet granular matter. This is of practical interest for civil engineering and soil mechanics, as well as of fundamental interest to come to a better understanding of partially saturated granular materials.

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