New Journal of Physics
The open-access journal for physics

Tunable random packings

G Lumay and N Vandewalle

Group for Research and Applications in Statistical Physics (GRASP),
Institut de Physique B5a, Université de Liège, B-4000 Liège, Belgium
E-mail: geoffroy.lumay@ulg.ac.be

New Journal of Physics **9** (2007) 406
Received 28 June 2007
Published 8 November 2007
Online at http://www.njp.org/
doi:10.1088/1367-2630/9/11/406

Abstract. We present an experimental protocol that allows one to tune the packing fraction η of a random pile of ferromagnetic spheres from a value close to the lower limit of random loose packing $\eta_{\mathrm{RLP}} \simeq 0.56$ to the upper limit of random close packing $\eta_{\mathrm{RCP}} \simeq 0.64$. This broad range of packing fraction values is obtained under normal gravity in air, by adjusting a magnetic cohesion between the grains during the formation of the pile. Attractive and repulsive magnetic interactions are found to affect stongly the internal structure and the stability of sphere packing. After the formation of the pile, the induced cohesion is decreased continuously along a linear decreasing ramp. The controlled collapse of the pile is found to generate various and reproducible values of the random packing fraction η.

Contents

1. Introduction

How a large number of spherical objects can fill a volume is one of the most puzzling problems in mathematics and science [1]–[4]. Indeed, this question concerns a broad range of systems: structures of living cells, granular media and amorphous solids. The relevant parameter that characterizes a pile of spheres is the dimensionless packing fraction η, defined as the volume of all particles divided by the apparent volume of the assembly. This packing fraction has a maximum value $\eta_{\rm fcc} = \pi/3\sqrt{2} \simeq 0.74$ for spheres, corresponding to the face-centered cubic (fcc) lattice. When marbles are however randomly packed [5], a so-called random close packing (RCP) is created. Repeated experiments lead to a broad range of values for the packing fraction: $0.60 < \eta_{\rm RCP} < 0.64$, far below $\eta_{\rm fcc}$. The value of the packing fraction depends strongly on the history of the pile, i.e. on the way particles are sequentially placed in the assembly. When a dense fluid provides a strong buoyancy, settling particles form a so-called random loose packing (RLP) with values $0.56 < \eta_{\rm RLP} < 0.60$, depending on the fluid density [6]. In all experimental situations, mechanically stable random configurations cover a broad spectrum of η values.

Random assemblies have been extensively studied through experiments and numerical simulations. Experimentally, three-dimensional (3D) structures of large packings have been analyzed by x-ray tomography [7, 8]. These studies emphasize the random character of a 3D monodisperse spheres packing. Numerical models permit a wide variety of pile initialization methods to be explored and easy variation of the physical parameters. Different procedures for pile creation give different values of η in the range defined by $\eta_{\rm RLP}$ and $\eta_{\rm RCP}$. Moreover, the physical parameters used in the simulations select a range of η values. In some cases, the values are found in a narrow interval. The packing fraction η is also affected by the particle shape [9, 10], particle size distribution [11] and forces defined between particles [12].

Ordered assemblies have also been the subject of numerous studies. One can consider some ordered mechanically stable packing with very low packing fraction values. The lowest known packing fraction for a stable assembly is $\eta \simeq 0.0555$ [13]. Regarding the upper limit of the packing fraction, it was hypothesized by Kepler in 1611 that fcc packing is the densest possible with $\eta \simeq 0.7404$. In 1900, Hilbert included this conjecture in his list of unsolved mathematical problems.

When the size of the grains becomes smaller than about 50 μm, the interparticle cohesive forces begin to play an important role. Indeed, cohesive forces are known to affect strongly the structure and the flow properties of fine powders [14]–[16], since they induce the formation of large aggregates. Also, the effects of grain bridges due to humidity and arches in the macroscopic properties of granular packings have been demonstrated [17, 18].

In order to obtain tunable forces between particles, a magnetic field can be used with ferromagnetic grains [19, 20]. Some experimental studies have also been performed with permanently magnetized grains [21]. If the force is attractive for two grains situated along the direction of the magnetic field, the force becomes repulsive along the perpendicular direction. Therefore, the controlled magnetic interactions between particles are expected to change deeply the internal structure of the packing and to create large voids in the structure [22], thus decreasing the packing fraction. Indeed, the magnetic interactions between the grains may create a strong force network inside the packing. This decrease of the packing fraction as a function of the magnetically induced attraction between two grains has been measured in [20].

In this paper, we investigate the lower limit of the packing fraction of spheres without the use of a dense fluid. The basic idea of our work is to use a tunable cohesive force between the

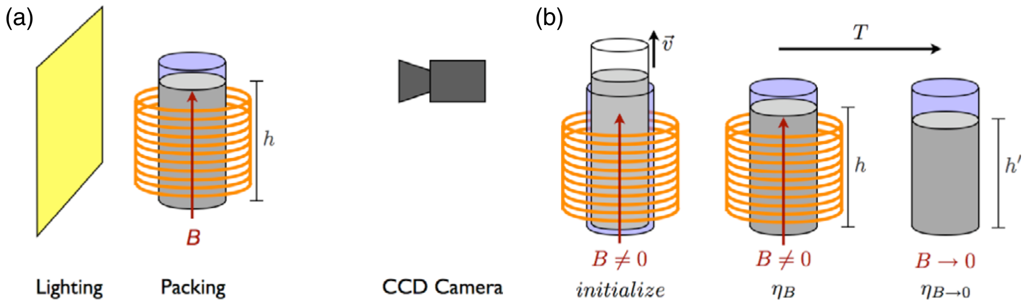

Figure 1. (a) Sketch of our experimental set-up. A high resolution camera (2048 × 2048 pixels) records the top of the packing placed in a glass tube. The pile is back-illuminated by a homogeneous lighting system. (b) Illustration of the pile creation and measurement protocol. Left: a smaller bottomless tube is inserted into the main glass cylinder. Then, a magnetic B field is applied through the packing. Afterward, this small tube is filled with spherical particles (in gray). The small tube is removed at constant speed v. Center: the position h of the upper grains allows the determination of the packing fraction η_B. Right: the magnetic field starts to decrease linearly to zero (reached after T seconds), the packing collapses partially and the new position h' of the upper grains gives an estimate of $\eta_{B\to 0}$.

grains during the pile preparation in order to control the packing fraction η of the pile. For this purpose, we consider ferromagnetic spherical beads which are submitted to an external magnetic field. After the formation of the pile, the magnetic field is decreased continuously along a linear ramp. This method to control experimentally the packing fraction opens new perspectives in the field of granular media and in the mathematical study of sphere packings.

2. Experimental set-up

A sketch of our experimental set-up is illustrated in figure 1(a). A glass tube of internal diameter $D = 21$ mm is placed into a vertical magnetic coil where a constant current could be injected. The magnetic field is thus parallel to the gravity field. The strength of the magnetic field can be fixed between 0 and 100 G. We checked that the variation of the magnetic field along the vertical axis of the coil does not exceed 10%.

The beads are monodisperse (Fe–Cr–C alloy) spheres of size $d = 2 \pm 0.001$ mm. We checked that, within the magnetic values we considered ($B < 100$ G), the spheres are ferromagnetic entities characterized by a nearly zero remanence (see the magnetization curve of a single bead in figure 2). Therefore, after switching off the magnetic field, the cohesion inside the packing is expected to vanish.

We measure the packing fraction of the pile by image analysis from a CCD high resolution camera (2048×2048 pixels). The average position h of the upper interface of the packing is measured and gives the packing fraction η by using the known value for the volumic mass $\rho = 7850\,\mathrm{kg\,m^{-3}}$ of the beads and the total mass m of grains. One has

$$\eta = \frac{4m}{\rho h D^2 \pi}. \qquad (1)$$

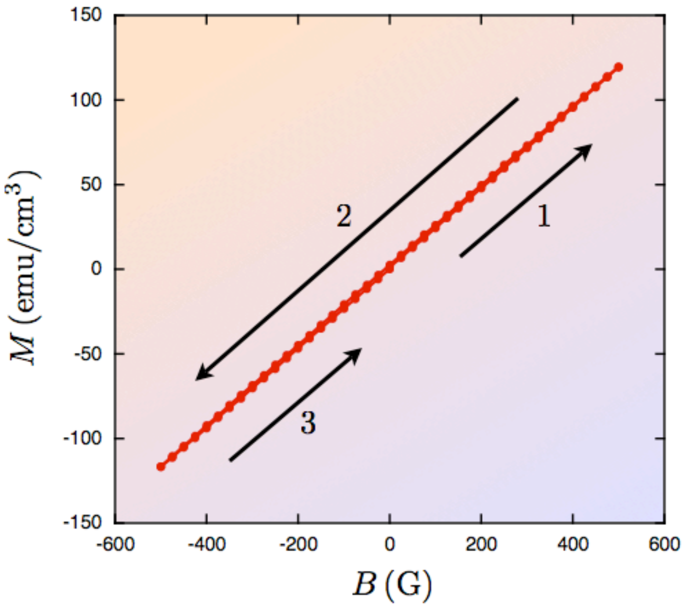

Figure 2. Magnetization M of a single grain as a function of the magnetic field B. The magnetic field increases first from 0 to 500 G (arrow 1). Then, B decreases from 500 to -500 G (arrow 2). Afterward, the loop is closed by increasing B to 0 (arrow 3). The absence of hysteresis loop means that the remanent magnetization is almost zero.

The part of the pile situated above the coil is typically less than 10% of the total height of the pile.

The figure 1(b) presents a sketch of the measurement protocol. In order to obtain a reproducible and spatially homogeneous initial packing fraction, we use an initialization protocol. A second smaller bottomless glass tube (external diameter below 21 mm) is inserted into the previous one. The magnetic field B is set at this step of the protocol. The small tube is filled with a defined number (thousands) of beads. The small tube is then removed upward at a low and constant velocity $v = 1\,\mathrm{mm\,s^{-1}}$, leaving the grains to rearrange themselves into the larger tube. At the end of this initialization process, we measure optically the packing fraction η_B. Then, the magnetic field B is left to vanish linearly during a time T. Afterward, the packing fraction $\eta_{B\to 0}$ is measured.

3. Packing fraction

Figure 3 presents both packing fractions η_B and $\eta_{B\to 0}$ as a function of the magnetic field B. In this figure, the decreasing time of the magnetic field is fixed to $T = 0.12$ s. Between two successive measurements, a pile without magnetic field was created. Then, we checked that η_0 does not present any deviation due to a remanence of the grain magnetization. When $B = 0$ (without the coils), one recovers a value $\eta_0 \simeq 0.61$. This η_0 value is reproducible for the fixed value ($v = 1\,\mathrm{mm\,s^{-1}}$) of the rising velocity during the initialization protocol. A much slower removal would lead to smaller η_0 values. From a naked eye observation of the grains

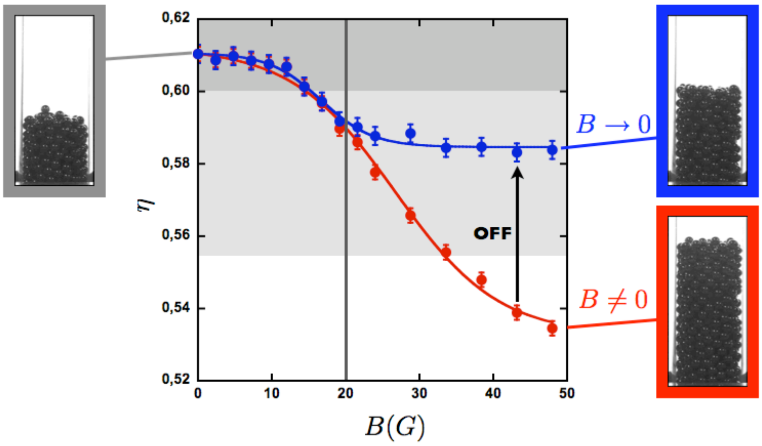

Figure 3. (in red) Packing fraction η_B as a function of the applied magnetic field B (expressed in gauss). (in blue) Packing fraction $\eta_{B\to 0}$ after switching off the magnetic field. Error bars are indicated. Hyperbolic tangent curves, fitting the data, are guides for the eye. The dark gray region denotes the usual RCP values, while the light gray region represents settling (RLP) experiments [6]. The vertical line at $B = 20$ G corresponds to the situation for which the magnetic force between two contacting grains along the magnetic field is equal to the weight of a grain. Snapshots of the upper grains, just above the coils illustrate the changes of the packing arrangements.

situated near the wall, we can conclude that the packing is completely disordered. From that pile preparation, we checked that large RCP values up to 0.64 can be approached after tapping the glass tube several times. After a large number of taps, the packing fraction slightly exceeds 0.64 and a crystallization of the grains is observed near the walls. This compaction-like process has been extensively studied in recent years [4, 23, 24]. When a magnetic field is applied during the preparation of the pile, the packing fraction could reach low values such as $\eta_B = 0.54$, i.e. significantly below the values obtained in settling experiments! One should note that, when the magnetic field is applied after the pile preparation at $B = 0$, no modification is seen. From these observations, one can conclude that global gravity forces (due to the weight of the packing) are still larger than magnetic cohesive ones. However, the presence of B creates mechanically metastable contacts inside the packing, as we will see below. Indeed, when cohesive forces between grains become stronger, the formation of arches is favored, thus increasing the pile stability for low packing densities. More interestingly, when the magnetic field is switched off, some contacts deep inside the packing are lost and the pile collapses partially (see the blue curve in figure 3). Internal rearrangements lead to an increase of η to a value $\eta_{B\to 0} = 0.585$. This reproducible value is significantly lower than $\eta_0 = 0.61$, expressing the metastable nature of magnetic arches.

The cohesion in the packing is due to dipole–dipole interactions. We consider that each grain receives a magnetic moment $\vec{\mu}$ parallel to the magnetic field $\vec{B}$. The magnetic moment of a grain is proportional to its volume; one has $\mu \approx \frac{(1+\chi)Bd^3}{\mu_0}$. Attractive interactions take place along the field direction while repulsive interactions are seen perpendicular to the magnetic

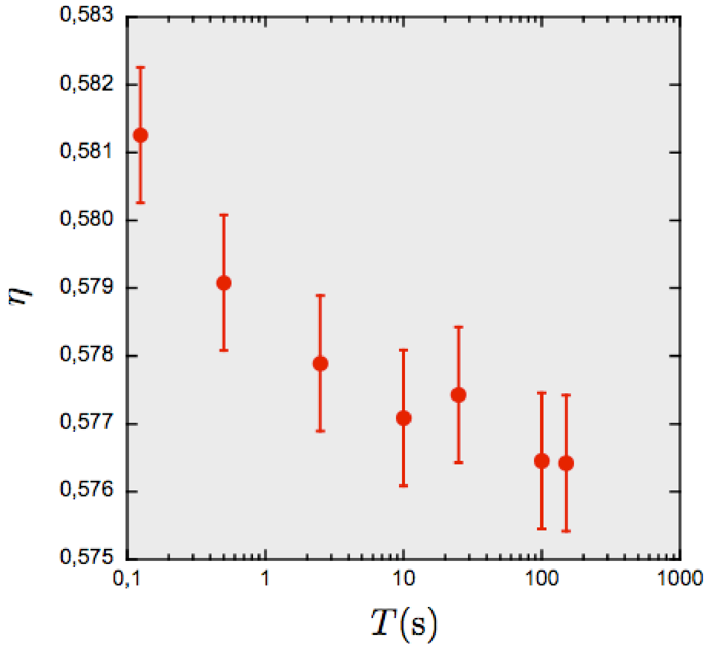

Figure 4. Semi-log plot of the packing fraction η when the magnetic field is removed along a ramp during T seconds. The initial magnetic field is $B = 45\,\mathrm{G}$. A slow collapse (long T) involves a loose and cohesiveless packing fraction below $\eta = 0.58$.

field. Chains of particles are expected to form in the packing. We will see below that the magnetic cohesion has indeed changed the packing density. Since the attractive cohesion due to the magnetic forces is roughly given by $U \approx \frac{\mu_0 \mu^2}{r^3}$, we can estimate the magnetic force between two contacting grains (in the field direction) to be $F_\mathrm{m} \approx \frac{\mu_0 \mu^2}{d^4}$. By comparing the magnetic force F_m and the weight of a grain $F_\mathrm{g} \approx \rho g d^3$, one has to consider the dimensionless ratio

$$\frac{F_\mathrm{m}}{F_\mathrm{g}} = \frac{(1+\chi)^2 B^2}{\mu_0 \rho g d}. \tag{2}$$

This ratio tells us that the gravity forces could dominate the magnetic cohesion in our experimental system. We checked indeed that two contacting beads detach from each other for $B < B_\mathrm{c} \approx 20\,\mathrm{G}$ due to gravity. Our calculation above gives the right order of magnitude which is within the range of our measurements. In our figure 3, the critical value B_c is denoted by a vertical line. This value corresponds roughly to the inflexion points of blue and red curves. This is the signature of a threshold by the contact disorder of neighboring beads. One should remark that the physical arguments given above (equation (2)) could be applied to other systems.

Figure 4 presents the final value of $\eta_{B\to 0}$ after a decay of the magnetic field from $B = 45\,\mathrm{G}$ (denoted by the vertical arrow in figure 3) to $B = 0$. The duration T of the current decay in the coils is controlled such that a long time T implies a slow collapse of the granular assembly. In that case, the final packing fraction remains below $\eta_{B\to 0} = 0.58$. This irreversible way to prepare the collapsed packing preserves some metastable arches in the system, thus explaining low values for η. In order to put those magnetic arches into evidence, we have observed the

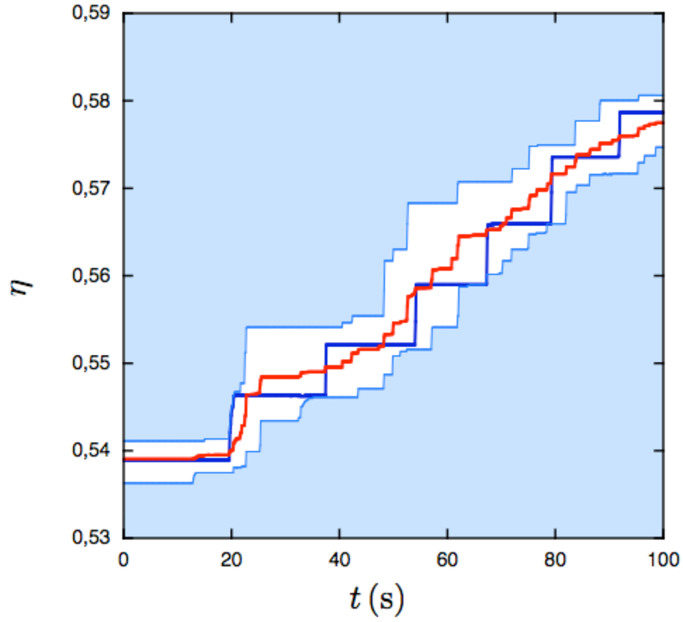

Figure 5. The thick blue curve presents a typical temporal staircase evolution of the packing fraction η during the 100 s collapse. The red curve corresponds to an average over several experiments. Both the maximal and minimal values of the packing fraction η observed during the series of experiments are shown by the thin blue curves.

evolution of η during the continuous and linear B decay for a long duration $T = 100$ s. The thick blue curve in figure 5 shows such a typical collapse. The packing fraction η is seen to increase by successive jumps, revealing the successive breaking of metastable magnetic arches. Indeed, an arch breaking induces internal rearrangements leading to a jump of the packing fraction. This staircase evolution is similar to what was recently observed when slowly changing the effective gravity in granular systems [18, 25]. These previous studies have proven that they are directly linked to the presence of arches in the system. The latter being controlled by friction between grains. In the present case, friction plays a role but could be supplemented by cohesive forces. When the magnetic field is decreased, the gravity forces begin to play a more important role. Moreover, these previous experiments have shown that the average length and number of stairs depends strongly on the container size. The temporal evolution of the packing fraction during the magnetic field vanishing has been measured several times. From these measurements, the average (red curve in figure 5), the maximum and the minimum (thin curves) values of the packing fraction η have been calculated for each time t. The difference between maximal and minimal curves is small (around 0.005) with respect to the overall excursion (around 0.04) of the packing fraction, emphasizing that the size of the fluctuations taking place in the assembly is controlled by the magnetic field. A plateau is always observed at the beginning of the curve for $0 < t < 15$ s. This short plateau is followed by an increase of the packing fraction η along a roughly linear trend. The absence of a robust plateau when the magnetic field is vanishing, suggests that the system is still in a metastable state even for $B = 0$. Indeed, we checked that a small perturbation of the system at the end of the process induces an additional densification of the pile. However, in the absence of any perturbation, the pile keeps a low and metastable packing fraction value.

4. Conclusion

Our experimental results emphasize that the packing fraction of random particle assemblies depends strongly on the preparation history. More importantly, we demonstrated that magnetic particles can form loose packing fractions in a reproducible way, due to metastable magnetic arches. The latter are revealed by some discontinuous collapse of the packing when the magnetic field is slowly removed. By combining our experimental procedure and the tapping (compaction) method, the packing fraction η could be controlled in a reproducible way from 0.58 to 0.64. By maintaining the presence of a magnetic field, low packing fractions such as 0.53 have been obtained. Our study proves the important role played by cohesive forces in granular systems and opens new perspectives in the research into sphere packings. Future work concerns the study of the internal structure of the packing by using x-ray tomography techniques. Numerical simulations will also provide useful information on the local structure of the sphere packing such as coordination numbers.

Acknowledgments

This work has been supported by the contract ARC 02/07-293. We thank J F Fagnard for magnetization measurements, C Becco, S Dorbolo, H Caps and F Ludewig for valuable discussions.

References

[1] Aste T and Weaire D 2000 *The Pursuit of Perfect Packing* (Bristol: Institute of Physics Publishing)
[2] Jaeger H M and Nagel S R 1992 *Science* **255** 1524
[3] Weitz D A 2004 *Science* **303** 968
[4] Nowak E R, Knight J B, Ben-Naim E, Jaeger H M and Nagel S R 1998 *Phys. Rev.* E **57** 1971
[5] Bernal J D 1961 *Nature* **188** 910
[6] Onoda G Y and Liniger E G 1990 *Phys. Rev. Lett.* **64** 2727
[7] Richard P, Philippe P, Barbe F, Bourlès S, Thibault X and Bideau D 2003 *Phys. Rev.* E **68** 020301
[8] Aste T, Saadatfar M and Senden T J 2005 *Phys. Rev.* E **71** 061302
[9] Donev A, Cisse I, Sachs D, Variano E A, Stillinger F H, Connelly R, Torquato S and Chaikin P M 2004 *Science* **303** 990
[10] Lumay G and Vandewalle N 2004 *Phys. Rev.* E **70** 051314
[11] Brouwers H J H 2006 *Phys. Rev.* E **74** 031309
[12] Dong K J, Yang R Y, Zou R P and Yu A B 2006 *Phys. Rev. Lett.* **96** 145505
[13] Gardner M 1966 *Packing spheres* (New York: Simon and Schuster)
[14] Umbanhowar P B and Goldman D I 2006 *Am. J. Phys.* **74** 720
[15] Castellanos A, Valverde J M, Prez A T, Ramos A, Keith P and Watson P K 1999 *Phys. Rev. Lett.* **82** 1156
[16] Valverde J M and Castellanos A 2006 *Europhys. Lett.* **75** 985–91
[17] Horváth V K, Jánosi I M and Vella P J 1996 *Phys. Rev.* E **54** 2005
[18] Dorbolo S 2005 *Eur. Phys. J.* E **17** 77
[19] Peters F and Lemaire E 2004 *Phys. Rev.* E **69** 061302

[20] Forsyth A J, Hutton S R, Osborne C F and Rhodes M J 2001 *Phys. Rev. Lett.* **87** 244301
[21] Blair D L and Kudrolli A 2003 *Phys. Rev.* E **67** 021302
[22] Vandewalle N and Ausloos M 1995 *Phys. Rev.* E **51** 597
[23] Richard P, Nicodemi M, Delannay R, Ribière P and Bideau D 2005 *Nat. Mater.* **4** 121
[24] Lumay G and Vandewalle N 2005 *Phys. Rev. Lett.* **95** 028002
[25] Scheller T, Huss C, Lumay G, Vandewalle N and Dorbolo S 2006 *Phys. Rev.* E **74** 031311