Is there a lower bound for solid volume fraction in random loose packing of noncohesive rigid spheres?

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Abstract

The packing obtained by pouring bearing balls in a container, and then by shaking it in order to get the maximal densification (once eliminated wall effects), has a solid packing fraction of 0.6366 ± 0.0004 [1]. This state has been named random close packing (RCP). Theoretical ideas as well as numerical simulations give a packing fraction near the value of 0.64. Much less well defined is what is named as random loose packing (RLP) that is obtained in experiments by gently pouring steel balls in a container without trying to maximize density. In this way experimenters [1] have found packing fractions as low as $\phi_{RLP} = 0.60$. This was an accepted value for RLP for noncohesive spheres until Onoga and Liniger [2] found a lower value by sedimentation of spheres in liquids with almost matched densities. They attained a lower limit given by $\phi_{RLP} = 0.55 \pm 0.006$. However, a criticism to this work is that in the limit of zero buoyancy we can not avoid the effect of attractive interparticle forces. Recently Farrel et al. [3] avoided the pitfall of the influence of interparticle attractive forces by sedimentation of frictional balls, with finite buoyancy forces. They adjusted the physical parameters of particles and liquids in order to ensure that the colliding velocity at contact is almost zero. For a coefficient of friction $\mu_s = 0.96 \pm 0.03$ they found a packing fraction below 0.54. Extrapolating their results they predict a packing fraction of 0.53 for a coefficient of friction $\mu_s = 1.2$. This value is below the theoretical minimum for frictional spheres given by $\phi_{RLP} = 4/(4 + 2\sqrt{3}) = 0.536$. We show in this work that unless we include rolling friction this is not possible. A simple estimation, based on isostaticity, show that for a disproportionate large value of rolling friction we could achieve densities much lower than 0.536, with a lower bound quite near to 0.15.

1 Introduction

A packing is a static set of particles in a containing space which do not overlap among themselves or with the space boundary. The packing obtained by pouring bearing balls in a container, and then by shaking it in order to get the maximal densification (once eliminated wall effects), has a solid packing fraction of 0.6366 ± 0.0004 [1]. This state has been named random close packing (RCP). Berryman [4] defined random close packing of geometrically perfect spheres as the one which has minimum packing fraction for which the median nearest-neighbor radius equals the diameter of the spheres, and he obtained a value of $\phi_{RCP} = 0.64 \pm 0.02$. Numerous computational schemes of random packing of frictionless spheres, though they do not give the same packing fractions, they all pack near the value of 0.64. The word random, implying maximum disorder, is an elusive concept difficult to define. Nevertheless, the random close packing of spheres in containers, when taking precautions to avoid cristallization zones and wall effects, have astonishing robust properties. The discrepancy between the experiment and the theory and simulations is ascribed to the non ideality (finite elasticity, friction, viscoplasticity, plasticity) of real spheres in the experiments.

Much less well defined is the random loose packing (RLP) obtained in experiments by gently pouring steel balls in a container without trying to maximize density. In this way experimenters [1] have found packing fractions as low as $\phi_{RLP} = 0.60$. This was an accepted value for RLP until Onoga and Liniger [2] found a lower value by sedimentation of spheres in liquids with almost matched densities. They attained a lower limit given by $\phi_{RLP} = 0.55 \pm 0.006$. However, a criticism to this work, is that in the limit of zero buoyancy we can not avoid the effect of attractive interparticle forces. In fact, the granular cohesive Bond number, defined as the ratio of interpaticle force to particle weight, could be greater than one (if exactly effective gravity equals zero, the Bond number will be infinite, because Van der Waals forces are always present). Song et al. [5] gave a phase diagram for jammed frictional spheres in the space of "coordination number-packing fraction". They found the diagram based on a statistical formulation of granular materials due to Edward and Oakeshot [6] and on detailed numerical simulations. In this diagram all disordered packings lie within a triangle demarcated by the RCP line, RLP line and a line called as granular line characterized by a coordination number of 4. In this work the theoretical minimum packing fraction of RLP is given by $\phi_{RLP} = 4/(4 + 2\sqrt{3}) = 0.536$.

Recently Farrel et al. [3] avoided the pitfall of the influence of interparticle attractive forces by sedimentation of frictional balls, with finite buoyancy forces. They adjusted the physical parameters of particles and liquids in order to ensure that the colliding velocity at contact is zero. They argue that they have measured the real coefficient of friction μ_s between two spheres in the liquid, and they attain a packing fraction slightly below 0.54, which is very close the theoretical minimum given by Song et al. From his work it is implied that for a coefficient of friction of $\mu_s = 1.2$ the extrapolated packing fraction would be 0.53 below the theoretical limit.

We suggest that including rolling friction we could attain lower values of the packing fraction. For the majority of the materials this effect would be negligible as the coefficient of rolling friction is very small. Theoretically, however, it is easy to see that if both the coefficient of rolling and dry friction approach infinity the falling particle will stick to the touching particle in the sediment and the packing fraction should approach the packing fraction of random ballistic deposition without restructuring which is quite near to 0.15, [7], [8]. This value is impossible to achieve in real experiments as there are no particles with neither an infinite value of dry friction, nor of the rolling coefficient. However, a general conclusion is that loose random packing of real spheres can not be defined only in terms of geometry because it is a dynamical problem dependent on the physical assembling procedure of the packing. We can not exclude that the packing fraction of the random loose packing in experiments will continue to decrease in the future, but we can not predict a well defined experimental lower limit for these RLP packings.

2 Coordination number in packing of spheres

The network of contacts inside the packing determine their mechanical properties. It is well known that the number of neighbours in contact with a given grain has a statistical distribution. Its average, Z, is called the coordination number of the packing. This is the simplest key parameter characterizing the network of contacts. Experimentally, Z for spheres was determined by Bernal and Mason [9], by coating a system of ball bearings with paint, draining the paint, letting it dry, and counting the number of paint spots per particle when the system was disassembled. They obtained a coordination number close to 8.5. This technique was used by Donev et al [10] for measuring the contacts between candies as model of ellipsoids claiming an excellent agreements between simulations and its unique experimental point. However this technique is very time consuming and prone to errors as it is very difficult to distinguish between true and close (but not touching) contacts. Recently Brujić et al [11] used as a model of granular packing fluorescently labeled silicone oil droplets with Nile red dye, suspended in a solution of 1:1:05 water to glycerol volume ratio to ensure refractive index matching. By means of confocal microscopic they were able to measure the contacts between droplets, and obtained a coordination number of $Z = 6.08$ for the RCP of the droplets, despite the polydispersity of the packing.

Compared with the scarcity of experimental measurements there are a plethora of numerical simulations of the packing of granular media in which we can found the statistics of contacts between grains as a function of shape, hardness, friction, interparticle forces and external load. There is an interesting work by Silbert et al [12] which simulates the packing of frictionless and frictional spheres. In this work it is shown that the coordination number decreases as we increase dry friction from $Z = 6$ for frictionless spheres to $Z = 4$ for spheres with an infinite coefficient of Coulmb friction. In the next section we discuss the relevance of these numerical findings.

3 Isostatic packings

In the area of Structural Rigidity it is well known that in a system of rotatable rigid rods in equilibrium the number of equations is exactly equal to the number of unknown internal stresses. This property of the rigidity between rotatable rigid rods is exploited by Moukarzel [13] to introduce the concept of isostaticity in granular media (a detailed discussion of this concept may be seen in [14]). Consequently a packing is said to be isostatic when the number of interparticle contacts is exactly the minimum needed for the packing to be stable under small loads. If the particles are themselves rigid the number of independent equations for the forces and torques must be equal to the number of contacts in order to determine univocally the unknown interparticle forces at each contact. For a packing of N rigid smooth spheres we have $3N$ equations for each grain to be at equilibrium, and an unknown normal interparticle force at each of the $NZ/2$ contacts. Therefore the coordination number for the packing to be isostatic must be $Z = 6$. For fairly enough hard particles we have the coordination number very close to 6 as have shown in the previous section, both by experiments and numerical simulations. However, for real smooth particles due to elastic deformation we may close contacts otherwise open, and the coordination number can be larger than six.

In the case of infinite coefficient of dry friction, the spheres will never slip and at each contact the friction force is unknown. Therefore the number of unknown forces would the number of contacts multiplied by 3 (in components: one normal force, plus two tangential forces in the plane perpendicular to the normal). The number of equations would be 3N for the forces and 3N for the torques. Thus for isostaticity we must have $3 \times NZ/2 = 6N$ which gives $Z = 4$ for the coordination number. For frictional spheres with a finite coefficient of friction there will be a fraction of the set of contacts that have yielded under Coulomb dry friction law (fully mobilized friction contacts). For these contacts the tangential forces are known. Therefore the total number of unknown tangential forces will depend on the coefficient of dry friction, and consequently the coordination number would vary from 6 to 4 as the coefficient of friction increases from zero to infinity. This is in agreement with the simulations [12] (see also [15]).

4 The role of rolling friction

If we include a finite coefficient of rolling friction, the packing density as well as the coordination number should both decrease. However, as the rolling friction, is much smaller than the coefficient of dry friction, the number of fully mobilized contacts under rolling friction would be the majority of contacts. The number of unknown rolling torques would therefore be small and as a consequence the coordination number would be quite near 4.

From a purely theoretical point of view we may introduce at hoc an infinite value for the coefficient of rolling friction. Under this assumption the number of unknown components of the rolling torque torque increases by two at each contact, since this torque lies in the plane perpendicular to the normal force. Therefore we should have now

$$5 \times \frac{NZ}{2} = 6N \tag{1}$$

which gives for the coordination number $Z = 2.4$. Such a low value of the coordination number is consistent with the coordination number of spheres deposited by ballistic aggregation with no restructuring (hit and stick). This same packing is obtained under the assumption of infinite attractive interparticle forces, since the spheres also hit and stick at the point of contact. In both cases we obtain the theoretical minimum density slightly below 0.15 as have been verified numerically [7] and experimentally [8].

5 Conclusion

The arguments presented in this paper are based on the isostaticity of packings. Real packings are made of material particles, and the forces and torques are always well defined when taking into account the deformations of the particles under the forces. Real packings do not require isostaticity for being marginally rigid. The deformations and forces will depend on the elastic constants, or viscoplasticity and/or plasticity at contacts. Assuming that the deformations are negligibly small we may arrive at isostaticity by quite different physical deformation processes. In particular for hard particles under elastic deformations we will be nearer to the isostatic packing the harder is the particle. The arguments presented here, though theoretical in nature, opens the door to packings with density below the theoretical limit of frictional particles.

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References

[1] GD Scott and DM Kilgour. The density of random close packing of spheres. *Journal of Physics D: Applied Physics*, 2:863, 1969.

[2] S. Torquato, T. M. Truskett, and P. G. Debenedetti. Is random close packing of spheres well defined? *Phys. Rev. Lett.*, 84:2064–2067, Mar 2000.

[3] G.R. Farrell, K.M. Martini, and N. Menon. Loose packings of frictional spheres. *Soft Matter*, 6(13):2925–2930, 2010.

[4] J.G. Berryman. Random close packing of hard spheres and disks. *Physical Review A*, 27(2):1053, 1983.

[5] C. Song, P. Wang, and H.A. Makse. A phase diagram for jammed matter. *Nature*, 453(7195):629–632, 2008.

[6] S.F. Edwards and RBS Oakeshott. Theory of powders. *Physica A: Statistical and Theoretical Physics*, 157(3):1080–1090, 1989.

[7] P.K. Watson, H. Mizes, A. Castellanos, and A. Pérez. The packing of fine, cohesive powders. In *Powders & Grains 97: Proceedings of the Third International Conference on Powders & Grains held in Durham, NC 18-23 May 1997*, pages 109–12, 1997.

[8] J. Blum and R. Schräpler. Structure and mechanical properties of high-porosity macroscopic agglomerates formed by random ballistic deposition. *Physical review letters*, 93(11):115503, 2004.

[9] JD Bernal and J. Mason. Packing of spheres: co-ordination of randomly packed spheres. *Nature*, 188(4754):910–911, 1960.

[10] A. Donev, I. Cisse, D. Sachs, E.A. Variano, F.H. Stillinger, R. Connelly, S. Torquato, and PM Chaikin. Improving the density of jammed disordered packings using ellipsoids. *Science*, 303(5660):990–993, 2004.

[11] J. Brujić, C. Song, P. Wang, C. Briscoe, G. Marty, and H.A. Makse. Measuring the coordination number and entropy of a 3d jammed emulsion packing by confocal microscopy. *Physical review letters*, 98(24):248001, 2007.

[12] L.E. Silbert, D. Ertaş, G.S. Grest, T.C. Halsey, and D. Levine. Geometry of frictionless and frictional sphere packings. *Physical Review E*, 65(3):031304, 2002.

[13] C.F. Moukarzel. Isostatic phase transition and instability in stiff granular materials. *Physical review letters*, 81(8):1634–1637, 1998.

[14] J.N. Roux. Geometric origin of mechanical properties of granular materials. *Physical Review E*, 61(6):6802, 2000.

[15] K. Shundyak, M. Van Hecke, and W. Van Saarloos. Force mobilization and generalized isostaticity in jammed packings of frictional grains. *Physical Review E*, 75(1):010301, 2007.

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