An Experimental Study of Random Loose Packing of Uniform Spheres

By

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Introduction:

Granular materials surround us. Sand, rice, beans, pebbles, and dirt are just a few examples of this class of materials. As this list of examples shows granular materials are large collections of “grains,” each of which is itself large in size compared to molecular length scales. The properties of granular media and their handling are also important for many of our industries including mining, food and pharmaceuticals. Additionally, they serve as an important model for phenomena ranging from traffic jams and the structure of comets or asteroids on the large scale to the packing of atoms in glasses on the microscopic scale[1]. Uniform spheres are a simple model of these granular materials.

A simple question one might ask about a collection of spheres is how they pack. If carefully arranged, as in a stacking of fruit or cannonballs, they can be made to form a dense, crystalline array. The fraction of the overall volume of the structure that is occupied by spheres is known as the packing fraction, ϕ. ($\phi \equiv V_s/V_c$ where V_s is the volume occupied by the spheres, and V_c is the volume they are contained in.) For the best crystalline packing, $\phi = 0.74$. If the spheres are just poured into a container, they form a random, disordered structure. As one experiences when filling a can with coffee or flour, such a packing can be made denser, merely by shaking the can and allowing the particles to settle into the lowest unoccupied spaces they can find. One major research focus of the structure of granular materials was the random close packing (RCP) limit[2]. Numerous experiments have shown that the densest, random packing that one can obtain for a collection of spheres is $\phi_{RCP} = 0.63$. Very few people have specifically studied random loose packing (RLP) the low density end of gravitationally stable random structures of spheres. Onoda and Liniger[3] studied random loose packing by gently allowing glass spheres to settle in a liquid whose density could be adjusted to minimize the effects of gravity. They quantify the effective weight of the spheres by the ratio

$\Delta g = (\rho_{ball} - \rho_{liq}) / \rho_{ball}$ where ρ_{ball} and ρ_{liq} are the mass densities of the ball and liquid. Extrapolating to the neutral buoyancy limit, $\Delta g \rightarrow 0$ they got $\phi_{RLP} = 0.555 \pm 0.005$.

It was the goal of my project to explore this limit in greater detail and to try to generate an even looser packing of spheres than Onoda and Liniger. The general strategy was inspired by an observation regarding the relationship between the frictional properties of particles and the so-called coordination number Z of a packing. The coordination number refers to the number of particles in direct contact with any given particle. For example, in a crystalline cannonball packing, the coordination number is Z = 12. Computer simulations[4] and general arguments using conditions for mechanical stability[5] have shown that as friction increases the coordination number for a random packing of balls should decrease from six (no friction) to four in three dimensions. These arguments are based on the observation that for balls with friction, in addition to the fact that the net force on any given particle is zero, there is the requirement that the net torque has to be zero. The larger number of constraints for mechanical equilibrium reduces the coordination number. Physically it appears reasonable that friction provides the means for stabilizing a larger number of delicate structures like arches and voids, than just geometric interference alone. The logical prediction that follows is that the packing fraction will decrease as the coordination number decreases; but no one has examined the systematic effect of friction on the random packing of spheres experimentally.

Experimental Strategy

This research investigates experimentally the influence of surface roughness and friction on the packing fraction of spheres. The random structures of spheres were assembled by dropping acrylic spheres into a container, then measuring the resulting packing fraction. The roughness of the spheres was varied in a controlled manner by etching the balls in acetone for different lengths of time. Methods employed to reach the lowest possible packing fractions included decreasing the apparent weight of the random structure of spheres and decreasing the impact velocity of the aggregating spheres.

A loose, fluffy packing of spheres is very fragile and will collapse and form a denser packing if it is mechanically disturbed. It is therefore important that the spheres be deposited very gently into the container. In order to reduce the influence of inertial and gravitational forces on the loose random structures, the spheres were dropped into liquids of different densities and viscosities. The balls were nearly neutrally buoyant and were further slowed down by the viscous drag of the fluid. They thus hit the structure with smaller terminal velocities and loaded them less with inertial forces than would be the case for deposition in air. The buoyant force of the liquid made the apparent weight of the structure adjustable.

The packing fraction was measured by determining the ratio between the volume of the spheres and the volume of the loose structure made by the spheres. The volume of the spheres V_s was determined from their total mass m_s and density ρ_s ($V_s = m_s / \rho_s$). Since the range of packing fractions to be explored is very small, it was crucial to measure the packing fraction with good accuracy and high precision.

Experimental Procedures

To be able to perform the main experiments of measuring the packing fraction I first had to characterize the spheres used in it. The balls were made of acrylic and purchased from McMaster-Carr.

The mass of 509 smooth balls was measured to be 10.014 ± .001g giving an average mass per ball of 0.01967 ± .00001g. (see Picture 1)

Volume and Density:

To determine the average volume of the balls, I measured 10 balls using a micrometer. I measured each ball in four different directions, giving an average diameter of 0.3195 cm with a standard deviation of ± 0.0005 cm. Figure 1 shows the range of diameters for smooth balls. Care was taken not to deform the spheres by barely touching them with the micrometer. This was accomplished by measuring the diameter of a sphere when it started to drop from the micrometer. The average volume computes to be 0.01708 ± 0.00008 cm^3 and the density of the acrylic spheres to be 1.152 ± 0.001 g/cm^3.

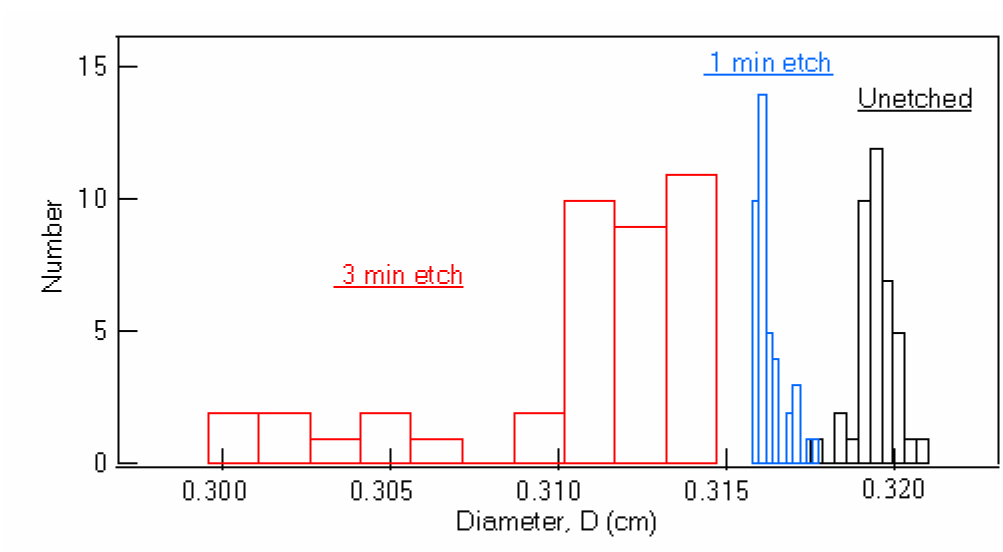

Figure 1: Distribution of the diameters of the sets of etched acrylic spheres

I used the displacement method to double check the density. I measured the volume of the spheres by measuring the volume of fluid displaced by a known mass of spheres. The density of the unetched acrylic balls was 1.16 ± 0.02 g/cm^3, confirming the results from direct measurements of mass and diameter. This method for determining density is less precise partly because the volume of 500 spheres is very small making it hard to measure precisely.

The balls did not absorb any water when soaked in it. The balls did not swell as confirmed by mass and diameter measurements respectively within the same accuracy as the above results.

Properties of Rough Balls

In order to change the roughness of the balls I etched them in acetone as it is a solvent for acrylic. I placed the balls into a beaker containing acetone, put the beaker in an ultrasonic shaker, and stirred the balls to ensure that the contacts between balls did not become permanent. I dried off the balls with a Kim wipe immediately and compared their before and after etch masses. I also measured the ball diameters to see how they changed. I observed with the microscope that for the shortest etches the surface of the balls became pockmarked with many small, irregular-shaped dimples. The longer the balls were in the acetone, the bigger and deeper the indents became. They also tended to look more and more like miniature golf balls compared to the smooth clear surface of the unetched spheres (see Picture 2). As the table below and Figure 2 show, I observed a linear decrease in mass and diameter of the balls with time in the acetone. The mass loss rate was about 3% per minute and the diameter of the balls shrank by about 1% per minute etch time. This is a confirmation of a uniform etching of the balls as this ratio of 3 between mass and diameter loss rate is expected as the mass is proportional to the third power of the diameter of the balls.

Time in acetone	Diameter of ball ±0.0005 cm	% Diameter loss ±0.1	% mass loss ± 0.1
30 sec	0.3190 cm	0.25%	2.6%
1 min	0.3167 cm	1%	3.6%
3 min	0.3106 cm	2.9%	9.2%
5 min	0.3038 cm	5%	15.7%
7 min	0.2982 cm	6.8%	18.3%

For further experimentation, I produced two large batches of etched balls. A three minute etch produced balls that had lost 9.7% of their mass. I measured with a micrometer the diameter of 20 etched balls for their new diameters. The average new diameter was 0.3109 cm showing that the balls had lost 2.8% of their diameter. A one minute etch created roughened balls that had a new diameter of 0.3162 cm or lost 1.0% of their original diameter and about 3.1% of their original mass (see Figure 1 for their statistical distribution). The diameters that I measured are an upper limit since the micrometer only measures the largest asperities ignoring the dimples in the balls.

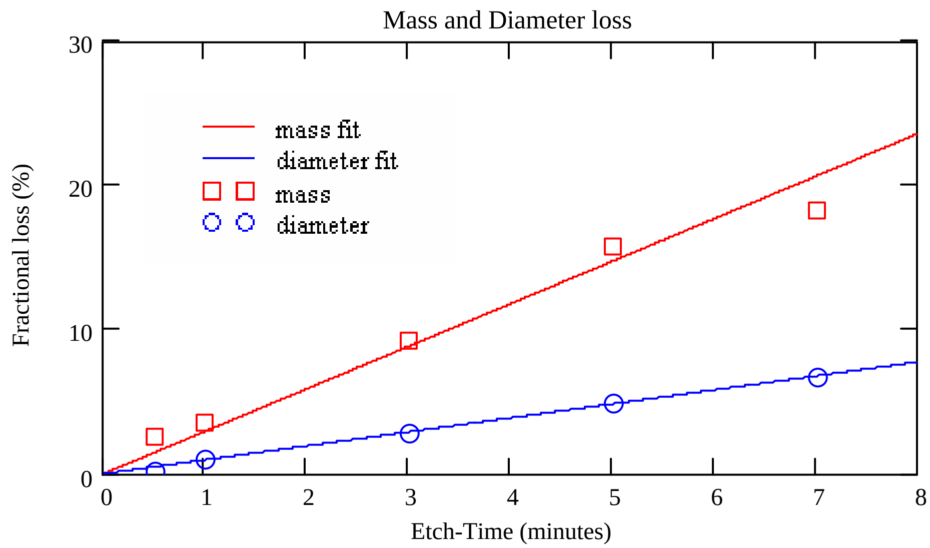

Figure 2: Fractional mass and diameter loss of etched acrylic spheres versus etch time

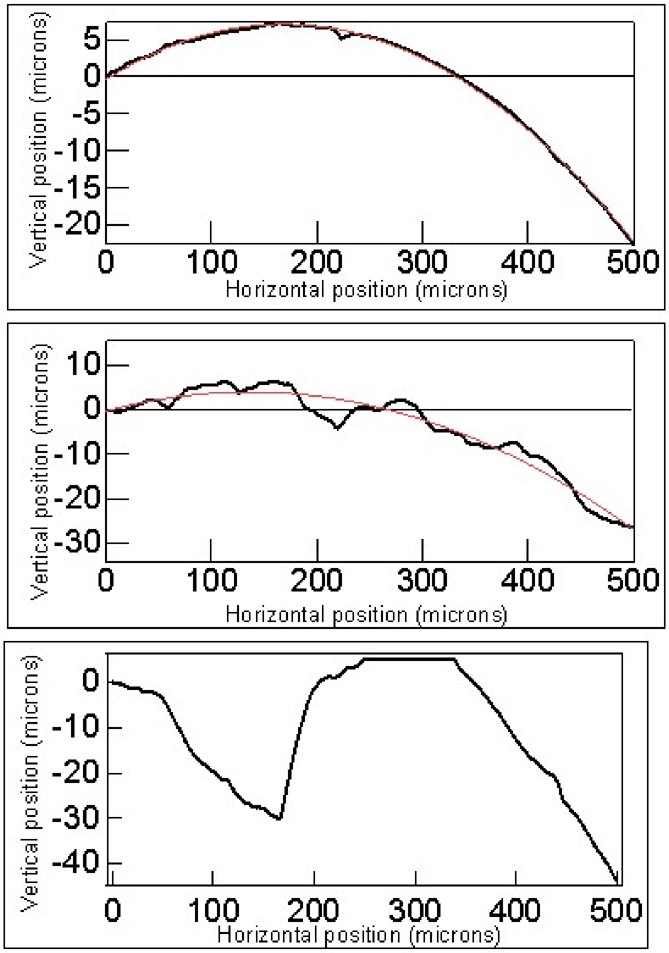

Figure 3: profilometry of unetched, 1min etch, and 3min etch acrylic spheres

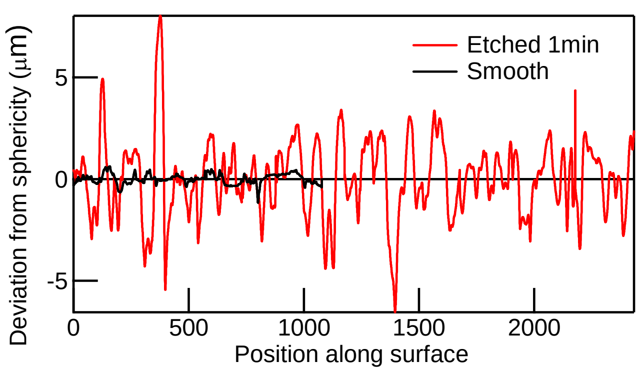

Figure 4: Residual roughness of unetched and 1min etch acrylic spheres

Roughness

To measure the roughness of the balls I performed a surface profilometry on different spheres. Profilometry uses a machine called a profilometer that drags a tip over a surface and reconstructs a profile of the surface from this information. I conducted several 500 micrometer long runs (see Figure 3). I analyzed the data by subtracting a circle from the profile to get the residue. The resulting residue shows how rough the surface of any given sphere is (see Figure 4). The unetched spheres were very smooth with an average surface roughness of ±0.25 µm. The one minute spheres had multiple dimples with an average surface roughness of ± 1.9 µm, but the largest asperities and holes were already more than ±7µm deep. The three minute spheres had dimples and asperities larger then ±30 µm but at a

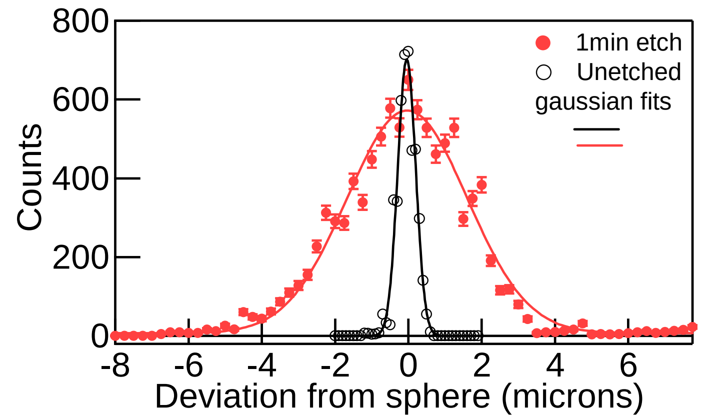

Figure 5: Statistics of roughness of unetched and 1min etch acrylic spheres

less frequent rate. A statistical analysis of the surface roughness is shown in Figure 5.

Sphericity and Size dispersion

I confirmed that the balls were spherical even after being soaked in acetone. The sphericity was defined as the standard deviation of four measurements along different diameters on a given sphere. This number was averaged over ten spheres for each sample. The sphericity of the smooth and one minute balls borders on the resolution of the measurement that I took with the micrometer. As a percentage of average diameter, the sphericity was ± 0.13% for the smooth spheres, ± 0.12% for the one minute etched balls and ± 0.36% for the balls of the three minute etch. The variation of the size of the three minute etched spheres was the greatest (about ±1%). The size variation for the smooth and 1min etched balls was much less (about ± 0.2%). This is a much smaller size dispersion than that of Onoda and Liniger who had balls of diameter 250 ± 20 μm. I also checked to make sure that the three sets of balls had the same density using the displacement method. The results are summarized in the following table.

Type of ball	Average Diameter	Sphericity (%)	Density
Unetched ball	0.3195 ± .0005 cm	± 0.13	1.16 ± 0.02 g/cm^3
1 minute ball	0.3162 ± .0005 cm	± 0.12	1.17 ± 0.02 g/cm^3
3 minute ball	0.3109 ± .0043 cm	± 0.36	1.16 ± 0.02 g/cm^3

Since all these measurements imply that the solvent only changes the surface geometry of the balls and does not affect the bulk density, I proceeded to use the more precise value of mass density that I had obtained by a direct measure of the density i.e. 1.152 ± .001 g/cm^3.

Angle of Repose, Angle of Stability and Friction

I tried several methods for measuring the effect of the etching on the coefficient of friction. These methods are described below; however, consistent trends were not obtained. Consequently, the surface roughness was used as a descriptor of the effects of etching.

I measured the angle of repose and the angle of stability of the balls in air to determining the differences in friction between the unetched and etched spheres. The angle of stability is the steepest slope that the pile can have before collapsing. The angle of repose of a pile is the angle of the slope that the pile will collapse to. I poured the balls into a Petri dish until it was about half full. Then I closed the top and taped it to the bottom. I flipped the dish on its side and slowly rotated it. The maximum observed angle before the slope collapsed was recorded as the angle of stability and the resulting angle after the collapse was the measured angle of repose. I repeated this experiment ten times and got the average angle of repose to be $25 \pm 3°$ and the angle of stability to be $30 \pm 3°$ for the unetched balls. The coefficient of static friction is $\mu = \tan(\theta)$, where θ is the angle of repose. I measured $\mu \cong 0.5$ (acrylic on acrylic) with no discernible difference between smooth and etched balls (see table below).

Another method employed to measure the coefficient of friction was the critical angle measurement. I glued three balls from a given set onto the bottom of a glass slide and placed this assembly onto another clean glass surface. I slowly increased the angle until the assembly moved. I was getting angles of about 40° with a large scatter in the data ($\mu \cong 0.8$ acrylic on glass, normal force about 0.05N). Then I glued a 118g mass on top of the assembly and repeated the experiments and got much lower angles. The difference in the results between unetched and etched balls were within error bars, giving now a coefficient of friction $\mu = \tan(\theta)$ of $\mu \cong 0.32$ (acrylic on glass) where θ is the critical angle (normal force about 1.2N) (see table below).

Type of ball	Angle of repose	Angle of stability	Average critical Angle
normal ball	25 ± 4°	31 ± 3°	17 ± 1°
1 minute ball	28 ± 4°	31 ± 3°	18 ± 1°
3 minute ball	26 ± 4°	33 ± 3°	19 ± 1°

I finally used a Pasco force sensor to measure the friction force directly while I slowly dragged the different assemblies on a surface of paper. You can notice the transitions from static to sliding friction in the stick-slip behavior shown in Figure 6. These measurements were made under a fairly large normal force load. I found again that all of the balls had roughly the same coefficient of friction ($\mu = F_{Friction} / F_{Normal}$) of $\mu = 0.62 \pm .02$ (acrylic on paper), where $F_{Friction}$ is the maximal force before sliding occurred. There was very little difference between smooth and etched balls again.

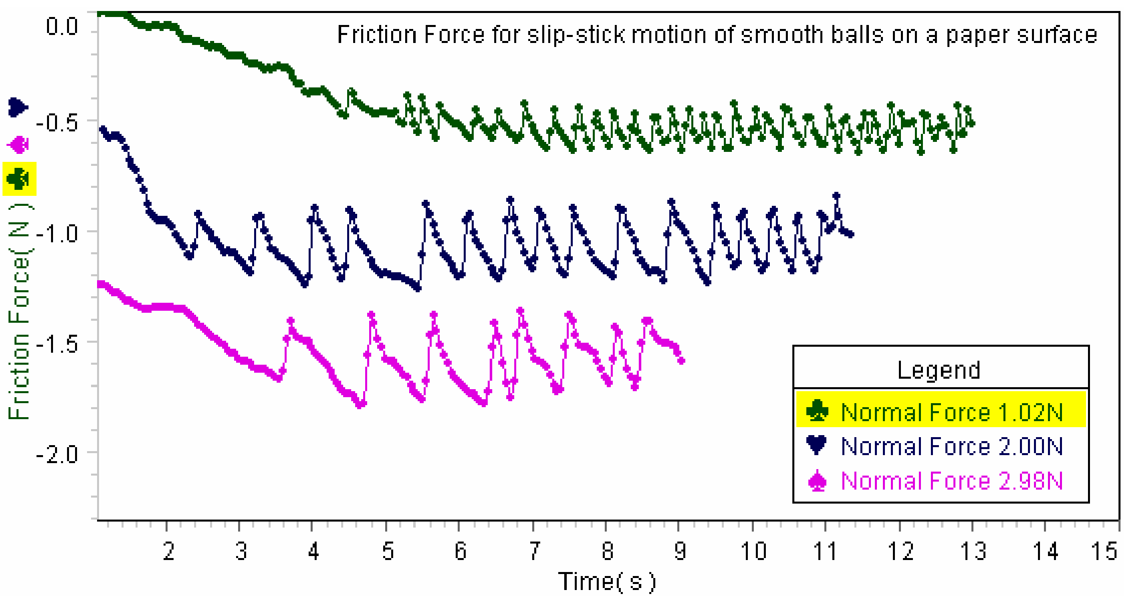

Figure 6: Friction Force to pull assembly of balls over a paper surface

The problem with all the friction determinations was that they measured the coefficient of friction under extremely large normal loads compared with the situation that arises in random loose packing. In random packing I only expect maximum forces of 10^{-2}N. Prior research [6,7] indicates that there is the possibility that the coefficient of friction will increase dramatically when the normal loading is decreasing to small values and that it will not follow the standard Coulomb law of friction at all. A hint of an increase of μ as the normal force decreased was seen in the critical angle measurements.

Measuring the Packing Fraction

To measure the packing fraction as accurately as possible I decided to use a volumetric flask for most of my experiments. To avoid having large terminal velocities in the accumulating spheres, the flask was completely filled with liquid giving me at least 10 cm of fluid above the aggregating structures at all times. This procedure also gave me the most consistent results. I always performed at least five independent runs for each of the three batches of balls. I used a digital camera for taking a picture of the top of the pile of spheres in order to correctly determine the volume of the pile, taking into account the irregular packing at the free surface of the pile. I used a mirror to avoid a parallax in these measurements and to ensure I was always viewing the surface of the structure at right angles (see Picture 3). I analyzed the images using the software package ImageJ comparing projections of known volumes with the top surface of the pile, allowing me to calculate the volume of the different random structures with an accuracy of $\pm 0.02 cm^3$. The volume of the spheres themselves was determined with a mass measurement and density calculation. In order to reduce any problems arising from the curved meniscus of the liquid I reduced the surface tension by adding a drop of soap.

To vary the gravitational load on the balls in the structure I varied the density of the liquid. I performed experiments with pure water and with solutions mixed by volume of 25% Glycerol 75% Water and 50% Glycerol 50% Water (with respective liquid densities of 1, 1.065, and 1.131 g/cm^3). The last of these gave almost neutral buoyancy conditions. These mixtures exerted an effective gravitational force as determined from the ratio of the relevant densities of $\Delta g = (\rho_{ball} - \rho_{liq}) / \rho_{ball}$. The increase in the proportion of glycerol in the mixture also increased the viscosity of the mixture resulting in slower terminal impact velocities. I repeated the experiments using a graduated cylinder to check on the influence of the container shape and wall friction on the packing fraction.

Results

Test of RCP: Smooth Balls in Air

First, I repeated some of the prior experiments. For smooth balls dropped in air I measured $\phi = 0.60 \pm 0.01$. I tried to attain random close packing by shaking the container. The balls tended to compact faster to a RCP with the application of small short vertical shakes. Long slow shakes sometimes even made the structure of the balls have a looser structure. When I shook the cylinder in a side to side motion only the top few layers of balls were affected. For the densest packing that I was able to create $\phi = 0.63 \pm 0.01$ confirming previous results[2].

RLP with Unetched Balls

The balls sank very slowly in the 50/50 glycerol / water mixture taking half an hour for all of the balls to settle. At this point any slight vibration or knock against the container could cause this fragile structure to collapse. Using smooth balls, the average packing fraction was $\phi = 0.558 \pm 0.010$ again confirming previous results [3]. Using water I got packing fractions of $\phi = 0.57 \pm 0.001$ and using the 25/75 glycerol / water mixture I got $\phi = 0.568 \pm 0.003$ (see Figures 7a and 7b).

Even Looser Packing: Rough Balls in Liquid

I also conducted the same experiments with etched balls. All results are displayed in Figures 7a and 7b showing the average packing fraction versus effective gravitational force. The unetched balls always had the highest packing fractions, the three minute etch the lowest packing fractions, and the one minute balls had packing fractions between the other two at all effective gravitational values investigated. This shows that the amount of roughness does make a difference on the RLP for any density of the liquid and in air as seen in the Figures 7a and 7b. The other clear trend is the dramatic reduction in packing fraction with a lowering of the gravitational forcing as already seen previously[3].

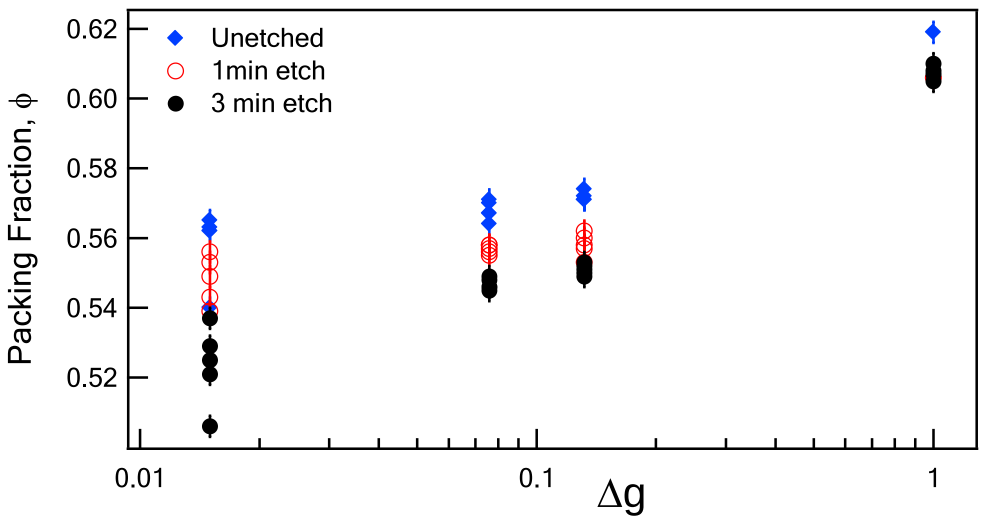

Figure 7a: Packing fractions of unetched and etched balls in a volumetric flask

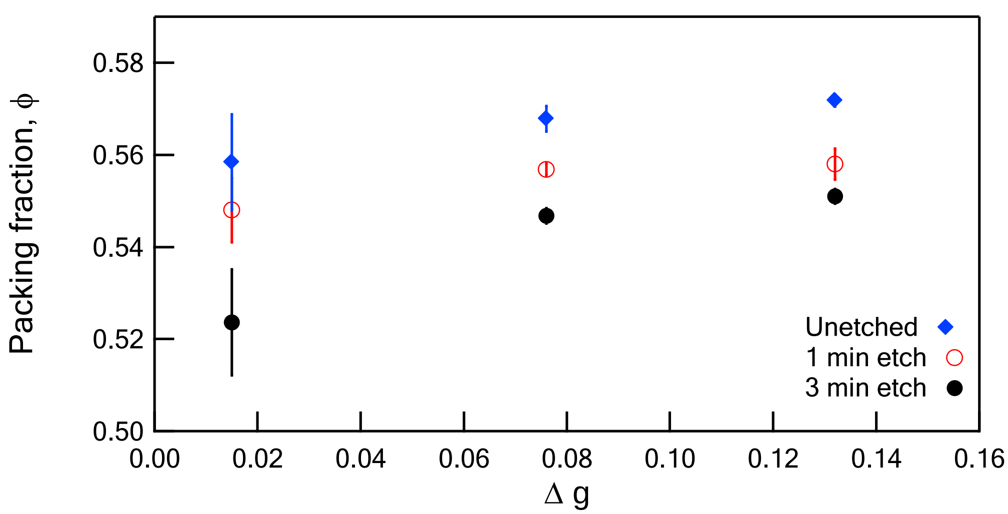

Figure 7b: Averaged packing fractions of unetched and etched balls in a volumetric flask

With the roughest balls (three minute etch) I observed an average packing fraction of 0.524 ± 0.011 in the 50/50 solution with the lowest packing fraction measured at 0.506± 0.005. To my knowledge, this is the lowest density random loose packing that has ever been reported in the literature.

To confirm that the measured packing fractions were not skewed by the container shape, I repeated the experiments in a graduated cylinder. I got similar results though the results from the graduated cylinder were less accurate and tended to be systematically slightly lower than the results from the volumetric flask (see Figure 8). This points to the influence of the wall friction on the structure of the random packing as the cylinder diameter was only 10 times the diameter of the spheres. Following Onoda and Liniger[3,2] this effect will reduce the expected packing fraction by at least 1%.

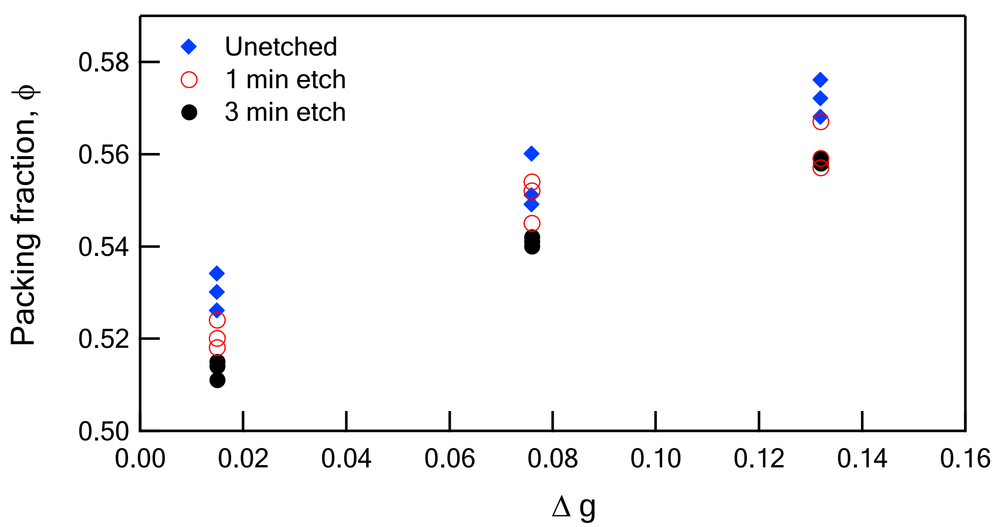

Figure 8: Packing fractions of unetched and etched balls in a graduated cylinder

Discussion

My data clearly confirm the assumption that the roughness of the balls affects the random loose packing fraction of uniform spheres. The balls with greater roughness have the lowest average packing faction of 0.524 ± 0.011, well below the previous record. It seems to follow that even though I was unable to directly measure the friction of the balls, the coefficient of friction did increase with the surface roughness of the spheres. The observed lowering of the packing fraction from 0.60 in air to 0.52 in the 50/50 mixture could either be attributed to an increase in friction as observed by Etsion[6] or a decrease in the gravitational load on the spheres. Further experiments are required to determine which one of these possibilities is the dominant factor in lowering the packing fraction. My experiments confirm the expected lower packing fraction for loose random structures of balls with friction that were predicted on general theoretical grounds[4,5]. My results are also independent of the geometry of the container, but the friction with the wall of the container had an observable effect of lowering the RLP by 2% compared to the volumetric flask.

Conclusion

My research has shown that roughness makes a difference on the packing fraction of granular materials leading to an average RLP of 0.52 and a record lowest RLP of 0.506 ever observed in uniform rough spheres. My research opens many doors to further studies on loose packing of spheres and leaves many questions unanswered. What is the coordination number of these structures? Is there a unique relationship between coordination number and packing fraction? How sturdy are these structures? How do they collapse? What is the relationship between roughness and packing ratios? What is the relation between roughness and friction under the conditions of these experiments (i.e. extremely low normal force loading). Are we close to the ultimate limit for random loose packing? All these questions could be the topics of future research in this field.

Acknowledgements

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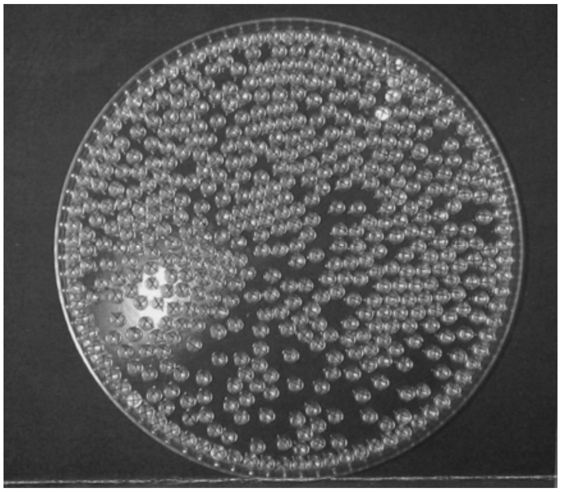
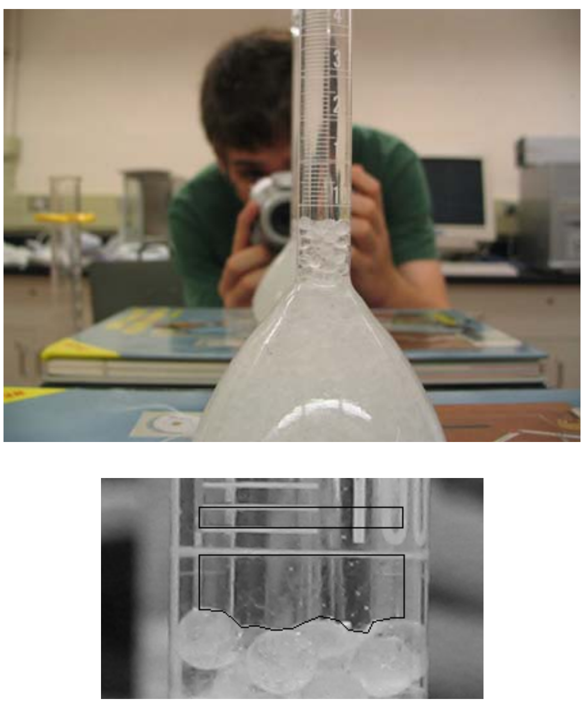
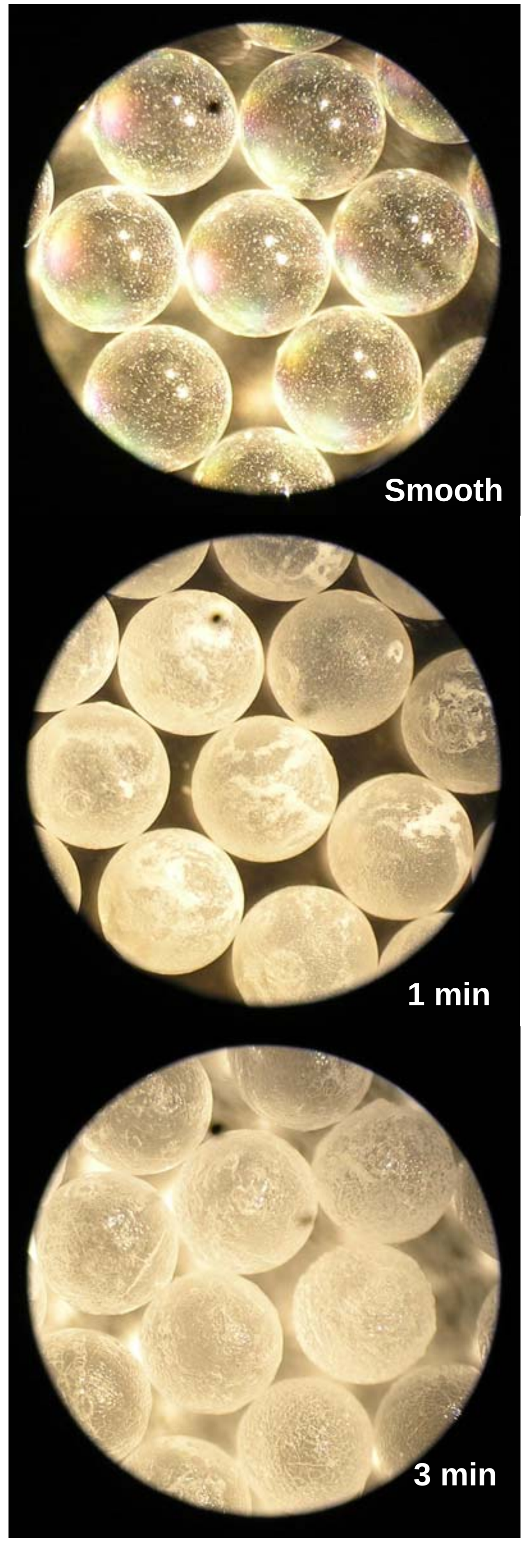


Picture 1: 509 balls in Petri dish



Picture 3: Original picture and picture edited with ImageJ



Picture 2: Picture of spheres under microscope