Random Loose Packings of Uniform Spheres and the Dilatancy Onset

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The random-loose-packing fraction of uniform spheres at the limit of zero gravitational force is determined to be 0.555 ± 0.005. This structure corresponds to a sphere packing at its rigidity-percolation threshold. The onset of dilatancy was also measured to be at approximately the same packing fraction. Preliminary evidence also indicates that shear thickening of suspensions with macroscopic spheres and a negative pore liquid pressure develops at the rigidity threshold. These provide further evidence that a hard-sphere glass transition could exist in the packing fraction range of 0.555 to 0.645.

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Random packings of uniform spheres have long been of interest as models for the arrangement of atoms in simple liquids[1] and glasses.[2] Stable random arrangements were first studied in detail by Scott[3] with spherical balls poured into rigid containers. He found that there existed a range of random-packing densities η (the fraction of space occupied by the balls), with an upper limit of 0.63 and a lower limit of 0.60. He concluded that "it seems unlikely that there are other stable random-packing arrangements for equal spheres in space which have packing densities outside these limits." The upper limit is often referred to as random close packing (RCP), and the lower limit as random loose packing (RLP).

The random-close-packing structure, since the work of Scott, has been studied extensively[4] in relation to its density, radial distribution function, number of neighbors, pore-size distributions, etc. It is now generally agreed that the RCP represents the densest packing of uniform spheres having a random structure and that it has a density of 0.635 ± 0.005.

The random-loose-packing limit has received much less attention[4] than the RCP limit. No clear definition exists for the RLP, except for the implication that it represents the loosest possible, random packing that is mechanically stable. The value of $\eta_{\mathrm{RLP}}=0.60$ was obtained[3] by rolling spheres gently into a container, without shaking. Computer simulations of the RLP suggest that the density strongly depends upon the method of packing.[5] Values obtained by different methods fall in a range of $0.56 < \eta < 0.60$, suggesting that stable structures may be possible below the limit given by Scott.

Important for the concept of an RLP is the exclusion of cases where attractive forces exist between spheres, as with colloids. If spheres are strongly bonded, very loose, stable structures are possible, due to linear chains and diffusion-limited-like aggregates,[6] and other arrangements that would not be stable if the bonding forces were not present.

The term "mechanically stable" means that the packing of spheres is in static equilibrium under an existing set of externally applied forces. In the case of macroscopic, spherical balls poured in a container, the external forces are due to gravity and the reaction forces of the container walls. It is also possible to increase the density of loose packings by applying hydrostatic pressure, as in a rubber bag. Thus, it seems clear that the loosest possible packing of spheres in a container would depend on the magnitude of the gravitational forces, which depends on the value of g, the acceleration due to gravity, and on the density of the spheres. If we wish to define the RLP as the loosest packing that can support an external load, the appropriate RLP limit would be the lowest density possible at the limit of $g \rightarrow 0$. In this paper we determine the limit experimentally.

The RLP($g \rightarrow 0$) we seek must consist of a continuous network of touching spheres. Its structure must also be more interconnected than one resulting from simple percolation, since it must also be able to support infinitesimal, external forces. The RLP($g \rightarrow 0$) should in fact be a packing of spheres at its rigidity-percolation threshold,[7] where the infinite percolation clusters can support a load.

An important property of granular materials, known as dilatancy, should also be affected by percolation networks. Dilatancy[8] is the property where the overall volume of packed particles expands when the packing is deformed by shear stresses. If expansion is constrained, a hydrostatic stress develops on the constraining walls as the particles are sheared. This phenomenon has been attributed to crowded packing conditions, where particles must spread apart to allow for their relative displacements. We would expect that, as packings become less dense, dilatancy would diminish in magnitude, until a low-enough density is reached where no expansion is promoted by shear. The density at which the dilatancy effect disappears will be called the "dilatancy onset." Although the structure at the dilatancy onset is dynamic, transmitting internal stresses to the boundary still requires, at least intermittently, a rigid continuous network. It could be expected, therefore, that the dilatancy onset and the RLP($g \rightarrow 0$) would closely correspond.

The RLP and the dilatancy onset both relate to minimum packings where hydrostatic and shear stresses

are coupled. As packings increase in density above the RLP, hydrostatic compaction forces are supported without internal shear displacements. As packings increase in density above the dilatancy onset, shear displacements cause an outward acting, hydrostatic stress on confining boundaries. A coupling between hydrostatic and shear stresses is fundamental to the critical-state theory in soil mechanics, where the resistance to shear is proportional to the hydrostatic stress and the proportionality constant increases with increasing density.[9] Assuming this relationship, we expect that the onset for dilatancy would correspond to the soil density where the proportionality constant diminishes to zero. This limit has been of little interest in soil mechanics since most soils of interest are under the influence of the Earth's gravity. But this limit is of interest in physics because it represents an important transition in how hard spheres interact, and as such may provide additional insights on the factors that determine the hard-sphere liquid-solid and glass transitions. In this paper, we also show in fact that the RLP limit and the dilatancy onset occur at the same packing fraction.

The approach taken in the experiments described below is to minimize the effects of gravity by studying the behavior of glass spheres in a liquid where the density can be adjusted to include neutrally bouyant conditions. The glass spheres[10] had a diameter of 250 ± 20 μm and a specific gravity of 2.46; these spheres were coarse enough for interparticle forces (e.g., van der Waals force) to be negligible compared with inertial forces so that aggregation effects are not present. A mixture of 64% di-iodomethane and 36% toluene with the same specific gravity as the glass beads provided neutral bouyancy. The density of the liquid is adjusted to lower densities by increasing the concentration of toluene, so that Δg, the effective gravitational force on the spheres, could be varied. Isopropopyl alcohol and water were also used for greater Δg.

The η_{RLP} as $g \rightarrow 0$ was determined for loose packings by allowing the spheres to settle gently in a 500-ml-graduated cylinder while immersed in liquids of varying Δg. The ratio of the diameter of the cylinder to the sphere diameter was 200 to 1. The variations in η with Δg are shown in Fig. 1, curve A. Extrapolating these data to $\Delta g = 0$ gives $\eta_{RLP}(g \rightarrow 0) = 0.555 \pm 0.005$.

Dilatancy was measured directly by shearing material in the gap between two concentric, cylindrical surfaces, with the inner surface rotating about its vertically aligned axis and with the outer surface stationary. The volumetric expansion resulting from shear is measured directly by observing the rise of the material in the cell. The inner cylindrical surface consists of the surface of an aluminum rod with diameter of 3.5 cm. The surface of the rod is roughened to reduce slippage of the beads during rotation. The outer cylinder was clear acrylic, with an inside diameter of 4.0 cm, so that the gap between it and the rod is 0.25 cm. This narrow gap width was necessary to insure adequate shear strains in the material. The ratio of the gap width to sphere diameter is 10 to 1, which is large enough to avoid large errors in the mean packing density due to wall effects, but errors on the order of 1% on the low side can be expected.[4] The length of the cell is 20 cm, and the height of material in the cell is of order 10 cm. The bottom of the cell is sealed to allow liquid to be contained in the gap with the spheres. The inner cell is coupled to an electric motor so that its rotational speed could be varied from 0 to 300 rpm. Dry material was evaluated by placing it in the gap and tapping the outer cylinder to obtain different packing densities. Spheres in liquids were studied by filling the gap with liquid and then allowing the spheres to settle in the liquid, with tapping. The inner rod was then rotated and the expansion of the material was observed. The quantity of liquid present was always sufficient to insure the presence of a supernatant layer above the packing after expansion. It was found that the volume increased in the 0–150-rpm range and remained constant at higher rpm. This limiting volume was used for the calculating η.

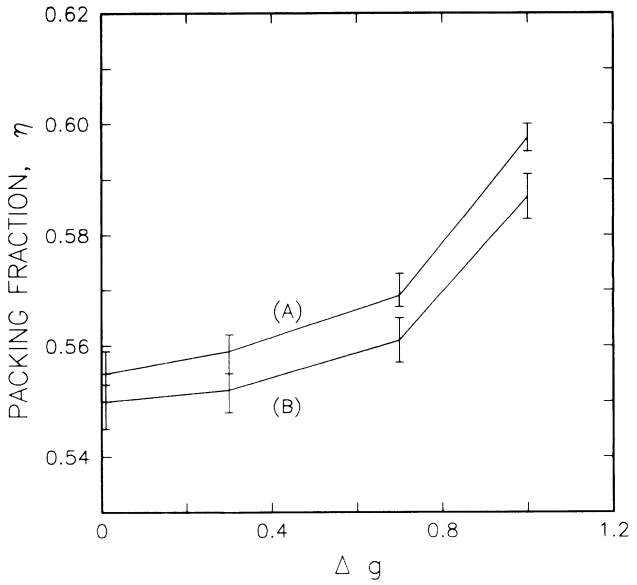

FIG. 1. Variation in packing fractions of spheres with the effective gravitational acceleration. Curve A is for the random loose packings obtained in the settling experiments, while curve B is for the expanded packings obtained in the shear-cell experiments.

Dry, glass beads with initial packing fractions between 0.61 and 0.63 all expanded to a packing fraction of 0.59. When the beads were immersed in a liquid, the expansion packing fraction varied with Δg as shown in Fig. 1. The value of η at $\Delta g \rightarrow 0$ is 0.550 ± 0.006.

The η of curve B are about 1% lower than the corresponding η values of curve A. However, it is likely that

curve B is lower only because of the wall effects. It is well known[4] that bead packings near a container wall are of lower density than in the bulk. This wall effect is negligible when the ratio R of container width to bead size is 200, but contributes around a 1% error on the low side when $R=10$. To correct for this effect, curve B would be shifted upward about 1%, which would make it closely correspond to curve A. As a result, $\eta(\Delta g \rightarrow 0) = 0.555 \pm 0.006$ becomes the best estimate for the expansion experiments.

If dilatancy does involve a rigid, continuous network, it should be possible to confirm this by measuring the pore liquid pressures. The load carried by a percolation network of touching particles is equivalent to an effective stress in soil mechanics. If the boundary walls are applying a hydrostatic load on the liquid-saturated packings, some of the load is distributed as pressure in the liquid and the rest is supported by the percolation network of particles. If a suspension of spheres in a liquid is concentrated enough for dilatancy to act, a shear deformation tends to expand the packing of spheres. As expansion occurs, the free liquid at the boundary recedes into the pore spaces between particles at the boundary. With wetting liquids, this creates a negative gauge pressure in the liquid due to capillarity that is equal and opposite to the mean effective stresses in the particle network. Thus, the presence of a load bearing network is signaled by a negative gauge pressure in the liquid (also known as the negative pore liquid pressure). We carried out a few experiments using a pore-liquid-pressure probe[11] with glass beads in a neutrally bouyant liquid. The pore-liquid probe measures negative pressures from 1 to 1000 mbar. When a suspension is stirred by a mechanical stirring rod in a 500-cm^3 beaker, it was found that negative pore pressures are sensed at $\eta \geq 0.56$, but not at lower values. While we are not able to show that negative pore liquid pressures below the detection limit for the probe do not occur for $\eta < 0.56$, we have shown that dilatancy is accompanied by the presence of a negative pore liquid pressure. This provides additional evidence that dilatancy involves a rigid, continuous network.

Our work also has implications to the rheology of concentrated suspensions. Shear thickening is observed in such systems, where the apparent viscosity increases with the shear strain rate.[12] In ordered colloidal systems, shear thickening is associated with the transformation of the structure caused by shear.[13] However, experiments with macroscopic spheres in neutrally bouyant liquids, where spheres do not order, also exhibit shear thickening.[14] For these cases, shear thickening might be associated with dilatancy, as has been often suggested but not proven. To study this possibility, rheological measurements using a cone and plate rheometer were made on glass beads in the neutrally bouyant liquid. It was found that the shear thickening behavior began at $\eta \simeq 0.56 \pm 0.01$, which essentially confirms previous findings.[13] These results support the hypothesis that the shear thickening behavior in these systems is caused by dilatancy.

The correspondence of the RLP($g \rightarrow 0$) and the onset of dilatancy at $\eta = 0.555$ for uniform spheres also provides additional insights for atomic, hard-sphere models. From molecular-dynamics and Monte Carlo simulations,[15] it is well known that a hard-sphere liquid-solid transition occurs, with the coexistence range $0.50 < \eta < 0.55$. It has also been suggested[16] that a glass transition exists in a hard-sphere system, somewhere in the range $0.55 < \eta < 0.61$. The dilatancy effects that we have observed indicate that a glass transition could be possible at $\eta > 0.555$, since the presence of a rigid percolation network could retard the rearrangements required for ordering. The particles become more difficult to rearrange as η increases until particles become immobile at the RCP limit.

Our conclusion is that the RLP($g \rightarrow 0$) for spheres occurs at $\eta = 0.555 \pm 0.005$ and that this corresponds to a rigidity percolation threshold for packings of spheres. This percolation network also gives rise to dilatancy and shear thickening in suspensions with macroscopic spheres. As such, the RLP($g \rightarrow 0$) is the dividing η between two fields, rheology and soil mechanics. In rheology, all theories assume no coupling between hydrostatic effective stresses and shear stresses, whereas in soil mechanics, this coupling is fundamental. Our evidence also indicates that a hard-sphere transition, if it exists, would occur in the range $0.555 > \eta > 0.635$.

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