Defining random loose packing for nonspherical grains

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The concept of "random loose packing" (RLP) has evolved through extensive study of loose packings of spheres, which has resulted in an accepted definition as the loosest packing that can be obtained by pouring grains. We extend this consideration to packings of nonspherical grains (ellipsoids) formed by slow settling in a viscous liquid, and perform a detailed analysis of the structural properties of the resulting packings. We find that as in the case of spheres the loosest ellipsoid packings are generated for grains with high interparticle friction. However, unlike spheres, these packings cannot be considered random as they have a significant degree of orientational ordering that increases with the grain's aspect ratio. This demonstrates that applying sedimentation or pouring techniques that have become part of the commonly held definition of RLP, will not generate random packings of ellipsoids. The consequences for the accepted definition of RLP and its applicability to nonspherical grains is discussed.

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When a set of spherical grains is allowed to settle into a packed state, a random packing will generally be formed, with the density of the final assembly lying between the so-called "random loose packing limit" (RLP) and "random close packing limit" (RCP) [1–5]. Though these two limits have been extensively studied, establishing their exact definitions has been a difficult challenge [6]. The concept of the random close packing limit has been synonymous with the idea of a densest random packing and is found to lie at around 64% for spheres. An exact definition is made complicated by the competition between order and density, that allows one to generate increasingly dense packings at the expense of increasing the order within the system. The maximally random jammed state (MRJ) has been proposed as a means of resolving these two competing effects, and has a precise definition for frictionless hard particles as the jammed packing that minimizes a chosen order parameter [6].

The topic of random loose packing has previously been examined in terms of packings of spherical grains, with several very different definitions of RLP proposed. Bernal and Scott simply considered RLP to be the random packing with the minimum density and employed various methods in attempting to achieve this, including tipping a cylindrical vessel on its side and rotating it before slowly returning it to a horizontal position [7,8]. It was found that friction played a key role in achieving low-density packings and that a density of 0.57 could be achieved for nylon spheres with a rough surface [9]. Onoda also considered RLP in these terms and more precisely stated the definition as the "loosest possible random packing that is mechanically stable" [3], finding a value of $\Phi_{\mathrm{RLP}} = 0.555 \pm 0.005$, obtained when glass spheres were sedimented in a gravity matched liquid (here mechanically stable simply refers to the grains being at rest in mechanical equilibrium, with the total net force on each grain being zero). These definitions inherently assumed that the spheres being considered were cohesionless. Dong *et al.* proposed a definition of RLP that included systems of cohesive grains and thus argued that due to the stability provided by the cohesion forces, Φ_{RLP} can range from 0 to 0.64 [4]. While more recent work instead focused on a definition related to the actual preparation method, with Φ_{RLP} being defined as the "loosest possible random packing that is mechanically stable that one can achieve by pouring grains" [10] (this again inherently assumes that the grains are cohesionless). Jerkins *et al.* also consider the problem in terms of pouring or sedimenting grains and found a value of $\Phi_{\mathrm{RLP}} = 0.550 \pm 0.001$ for glass spheres sedimented via a fluidized bed technique [5]. This definition which is based on preparation method has lead to a proposed definition of a "random very loose packing" (Φ_{rvlp}), as the loosest mechanically stable random packing generated by any preparation method [10].

In this paper we will extend this consideration to packings of nonspherical grains (ellipsoids) and investigate the applicability of these definitions. Based on previous definitions of RLP, a clear first consideration is: does pouring (or sedimenting) nonspherical grains result in a random packing? This is an important question, as if in general it does not, then any definition of RLP with such a basis may be of no use beyond the idealized case of spheres. To be exact, we will refer in this article to a sedimented loose packing density Φ_{SLP} as the lowest packing density generated by sedimenting (or pouring) a noncohesive infinitely frictional set of grains. Some previous studies utilizing spherical grains [5,10] have considered that $\Phi_{\mathrm{SLP}} = \Phi_{\mathrm{RLP}}$. We will examine how Φ_{SLP} for ellipsoids depends on the grain's aspect ratio and consider the structural properties of the generated packings.

The properties of dense random packings of frictionless elliptical grains have been studied both experimentally and computationally [11–13]. Loose packings have however received little attention. We employ the discrete element method (DEM) to simulate the interactions between a set of three-dimensional ellipsoids [14–16]. The normal force between two contacting particles is given by

$$F_n = -k_n \xi_n + C_n v_n, \tag{1}$$

where k_n is a spring constant determining the stiffness of the particles, ξ_n is the linear overlap of the particles, v_n is the relative normal velocity and C_n is a constant related to the coefficient of restitution. The tangential force is given by

$$F_t = \min\left\{\mu F_n, k_t \int v_t dt + C_t v_t\right\}, \quad (2)$$

where the force vector F_t and velocity v_t are defined in the plane tangent to the surface at the contact point [16,17]. The total tangential force, F_t, is limited by the maximum Coulomb friction μF_n, at which point the surface contact shears and the particles begin to slide over one another. The grains are sedimented in a viscous fluid, where the fluid is modeled using the Stokes equations. The fluid drag force on each particle, $\mathbf{F}_D$, is given by

$$\mathbf{F}_D = -V\nabla p + \tfrac{1}{2}\rho|\mathbf{u}-\mathbf{v}|^2 C_D A_\perp(\mathbf{u}-\mathbf{v}), \quad (3)$$

where V is the particle volume, p the fluid pressure, ρ the fluid density, $\mathbf{u}$ the fluid velocity, C_D the fluid drag coefficient, and $A_\perp$ the particle cross sectional area perpendicular to the flow, which is numerically calculated [18]. We use a drag coefficient given by Hölzer and Sommerfeld for nonspherical particles [19]. A rotational drag term is also imposed, $\tau_D = 8\pi\eta r^3[\frac{1}{2}(\nabla\times\mathbf{u}) - \boldsymbol{\omega}]$, where $\boldsymbol{\omega}$ is the angular velocity of the particle.

Our simulations are performed in a tall rectangular box, with a fixed base and periodic boundary conditions at the sides. Ellipsoids are placed at random locations and with random orientations within the box (Fig. 1). The box has a height $h = 0.8$ m and a width and depth of $w = 0.1$ m. Packings were generated for prolate ellipsoids with aspect ratios α ranging from 0.3 to 1.0, where for an ellipsoid defined by the equation $x^2/a^2 + y^2/a^2 + z^2/b^2 = 1$, the aspect ratio α is given as $a = \alpha b$. The ellipsoids in each setup have the same volume, which is chosen to be equal to the volume of a 2 mm radius sphere. The base of the simulation box has a layer of ellipsoids fixed in position at random orientations, so as to ensure that the grains are settling onto a disordered surface. This ensures that no ordering effects are caused by particles simply aligning on a flat base.

The parameters chosen for the simulation are $k_n = 10^5$ N/m, a restitution coefficient of $e = 0.5$, a particle density $D = 2.7\times10^3$ kg/m^3, and values of interparticle friction $\mu = 0$ and $\mu = 1000$. Values of viscosity ranging from $\eta = 0$ Pa s to $\eta = 1$ Pa s are examined. The fluid has a density $\rho = 10^3$ kg/m^3, giving an effective gravity for the particles in the fluid of 0.63g. Slow settling of particles can be simulated using a nearly gravity matched fluid and/or a high viscosity value. As a gravity matched fluid mainly affects the vertical motion of the particles, we employ a high viscosity fluid which reduces both the vertical settling speed of the grains (their terminal velocity) and also any lateral motions of the particles as they settle. This ensures the minimal grain motion during the settling process and leads to the loosest packing arrangements.

The packing fraction for the final static system is determined in a $\delta = 1$ cm thick region centered in the vertical direction. The final packing is around 5 cm high, so our sample region is sufficiently distant from both the top and bottom of the packing to be unaffected by any density variations at these limits.

The final packing fractions plotted against aspect ratio for a range of viscosities are shown in Fig. 2. The curves for both the zero-friction and the high-friction case are shown. The final packing fractions for zero-friction grains are independent of the value of viscosity, even though the settling rate is greatly reduced for higher viscosities. This is due to the ability of the grains to freely slide against one another until they achieve the required number of contacts necessary for mechanical stability.

In the high-friction limit, the additional structural support provided to the packing as a result of frictional contacts results

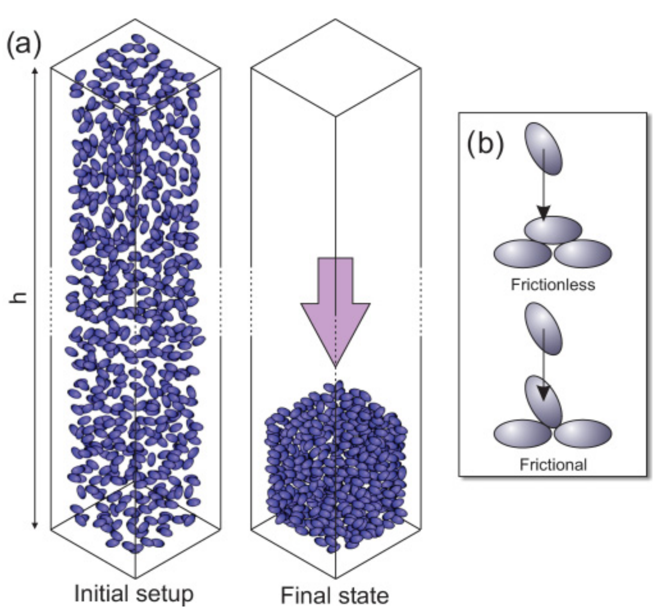

FIG. 1. (Color online) (a) Schematic of initial setup and final state of simulation. (b) Illustration of a frictionless elliptical grain (top) and a frictional grain (bottom) settling onto a surface composed of elliptical grains. The frictionless grain slides into the position where its center of mass is lowest, causing its axis to align in the plane normal to gravity. The frictional grain is held in place at a random angle due to frictional forces.

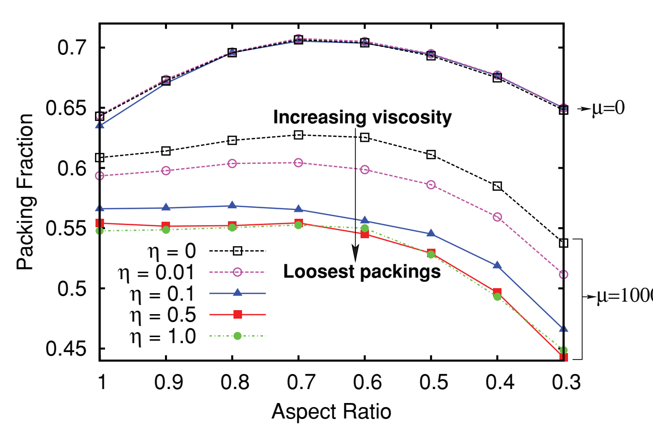

FIG. 2. (Color online) Packing fraction variation with the ellipsoid aspect ratio for high-friction and zero-friction cases. The zero-friction curves collapse onto a single line, with a maximum packing fraction at an aspect ratio of around 0.7. For the high-friction case, higher viscosities lead to looser packings across all aspect ratios. The sedimented loose packing limit (Φ_{SLP}) is reached at high viscosity and high friction.

in a reduction in packing fraction for all aspect ratios. As the viscosity is increased, the packing fraction progressively declines until the curves converge to a sedimented loose packing limit Φ_{SLP}. It can be seen that the curves for $\eta = 1.0$ Pa s and $\eta = 0.5$ Pa s are almost coincident, suggesting that this is a sufficiently slow sedimentation rate to generate packings that are representative of the loosest obtainable via a sedimentation technique. Equivalent density packings are also obtained for $\mu = 10\,000$, indicating that this is also representative of the infinite friction limit.

The average number of grains in contact is one of the key properties of granular packings that can be used to understand both their geometrical properties and their structural stability [20,21]. The early study of this quantity was due to Maxwell [22], with more recent work by Bennet highlighting its importance in understanding sphere packings [23]. The standard isocounting conjecture, based on a simple application of constraint theory, asserts that to constrain a system an average of two contacts per degree of freedom d_f are required ($Z = 2d_f$) [12]. This would suggest a discontinuous jump from $Z = 6$ to $Z = 10$ as we go from a sphere to a frictionless uniaxial ellipsoids with an infinitesimal aspect ratio. However, this has been shown to not be the case [24]. In Fig. 3, we see that isocounting performs well for spheres with $Z = 6$, but there is a smooth variation in Z with ellipticity from $Z = 6$ (spheres) up to $Z = 10$ (large aspect ratio ellipsoids). It has however been demonstrated that such underconstrained packings with $Z < 2d_f$ can in fact still be jammed [25].

When we introduce friction, again simple arguments based on constraints suggest that for spheres a minimum number of contacts $Z = d_f + 1$ (where $d_f = 3$ is the number of degrees of freedom of a sphere) are required to constrain the system. In Fig. 3 we see that this minimum number of contacts is approached in the limit of high friction and high viscosity, with our sphere packings having a value of $Z \simeq 4.2$ for $\eta = 1$ and $\mu = 1000$. When we introduce ellipticity, the same number of

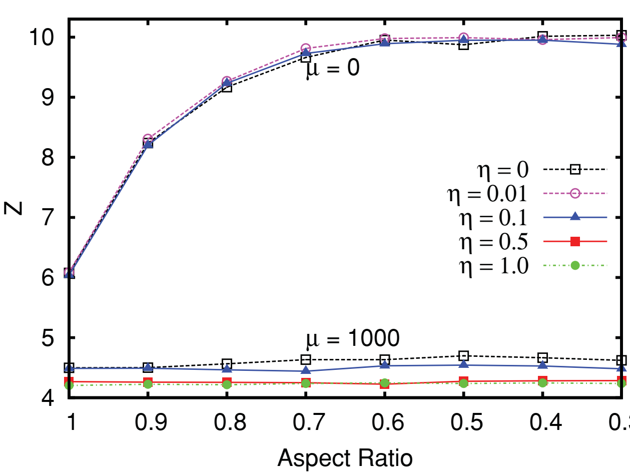

FIG. 3. (Color online) Variation of the average contact number Z with aspect ratio for the high friction-and zero-friction cases. The average contact number for the zero-friction case varies smoothly from $Z = 6$ to $Z = 10$ as the aspect ratio is varied, independent of the viscosity used in the simulation. For the high-friction case ($\mu = 1000$), Z decreases with increasing viscosity (η), tending toward a value of $Z \simeq 4.2$.

frictional contacts between grains are sufficient to constrain any movement of the grains despite their additional degrees of freedom. We find a minimum value of $Z \simeq 4.2$ for ellipsoids with aspect ratios over the full range from $\alpha = 1$ to $\alpha = 0.3$. This is due to a frictional contact's ability to constrain both the rotational and translational degrees of freedom of a grain at the contact point, providing additional mechanical stability that allows stable packings to form at low densities.

We will now consider the ordering present in our packings by quantifying the degree of orientational alignment of the grains. The lowest energy position for a prolate ellipsoid placed on a surface under the influence of gravity is for its center of mass to be at the lowest possible height and this will correspond to its semimajor axis lying flat in the plane normal to the vertical direction. Monte Carlo packings of frictionless ellipsoids that incorporate a gravitational energy have been found to have a considerable degree of such ordering [26]. Here we examine the degree to which gravity induces orientational alignment of the grains and the roles that friction and liquid viscosity play. We quantify the degree of orientational alignment using an order parameter

$$\chi_\theta = \frac{3}{2}\left\{\frac{1}{N}\sum_{i=1}^{N}\cos 2\left(\theta_i - \frac{\pi}{2}\right) - \frac{1}{3}\right\}, \tag{4}$$

where θ_i is the angle between the semimajor axis of the ith particle and the vertical axis [26]. If all grains have random orientations, $\chi_\theta = 0$, while if the grains all lie flat $\chi_\theta = 1$. We have also determined the more commonly used S_2 order parameter [27], quantifying the degree of orientational ordering of the major axes of the grains about their average nematic director, and have found that this parameter follows the same trends observed for χ_θ.

Figure 4(a) shows the variation in χ_θ for packings of ellipsoids with $\mu = 0$. For the zero friction case, all packings have a significant degree of orientational ordering, with $\chi_\theta > 0.1$ over the full range of ellipticities considered. Increasing viscosity causes more orientationally ordered packings to be formed for a given aspect ratio ellipsoid. This is due to the larger degree of grain motion at low viscosities, which leads to numerous collisions between grains before they have dissipated their energy and come to rest. As the viscosity is increased, the deposition of the grains becomes closer to sequential deposition onto an already packed bed and thus each grain is relatively unencumbered by interactions with other grains that are depositing onto the packing surface. Each grain can then slide into its equilibrium position corresponding to the best available orientation nearest to pointing in the plane normal to the vertical (see illustration in Fig. 1(b)). Interestingly, although these packings have very high degrees of orientational ordering, they have equivalent densities to those obtained for MRJ packings of ellipsoids [27]. This shows that when real granular systems are found to have densities near to those in simulations of random packings, they may still have a high degree of internal orientational ordering, which can have a significant effect on their structural and mechanical properties.

We see in Fig. 4(b) that for the high-friction grains the opposite is true, with larger viscosities causing a decrease in

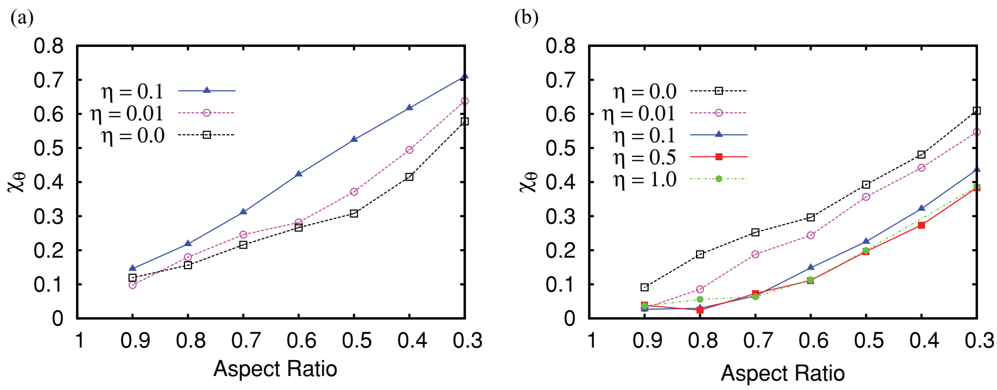

FIG. 4. (Color online) Variation in the orientational order parameter χ_θ for packings of ellipsoids with $\mu = 0$ (a) and $\mu = 1000$ (b).

the degree of orientational alignment. Higher viscosities cause the grains to settle more gently onto the packing surface, making it more likely that they obtain a statically stable configuration where frictional contacts between grains support a given grain at an orientation far from lying flat in the plane normal to gravity. However, while friction does reduce the degree of orientational alignment, a very significant amount still remains. When we reach the sedimented loose packing limit (Φ_{SLP}) at high viscosities and high friction, the degree of orientational ordering is small only for grains with low aspect ratios, with $\chi_\theta < 0.1$ for $1.0 \geqslant \alpha \geqslant 0.7$. As the ellipticity is increased, a rapid increase in χ_θ is observed with values of up to $\chi_\theta = 0.4$ found for the most elliptical grain considered ($\alpha = 0.3$). The high degree of ordering present in these packings clearly shows that $\Phi_{\mathrm{SLP}} \neq \Phi_{\mathrm{RLP}}$ and thus definitions of RLP based on a preparation method of slow pouring or sedimenting are not applicable to systems of nonspherical grains.

We have demonstrated that slowly settling a set of ellipsoidal grains with high intergrain friction in a viscous liquid produces a loose granular packing wherein the grains have a high degree of orientational ordering. This settling technique has been widely used both experimentally and computationally to generate loose random packings of spheres and has become part of the actual definition of RLP. These results demonstrate that such a definition of RLP that is based on the preparation method is not appropriate, as it at best has applicability only to systems of perfectly spherical grains. We would thus propose that a new definition of RLP is required that includes a reliable measure of the degree of ordering present in the system and in contrast to current definitions, is independent of the preparation method used.

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