

Defining Temperatures of Granular Powders Analogously with Thermodynamics to Understand the Jamming Phenomena

Tian Hao

15905 Tanberry Dr., Chino Hills, CA 91705

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Abstract

For the purpose of applying laws or principles extracted from thermal systems to granular powders, we may need to define temperature properly in granular powders. The conventional environmental temperature in thermal systems is too weak to drive particles in granular powders move around and cannot function as a thermal energy indicator. Several common scenarios in granular powder systems are discussed in this article and the corresponding analogous temperatures are defined in a similar way that the temperature of granular powders can have a same functionality as in thermal systems. For differentiating those two temperatures, the temperature in granular powders is named granulotemperature and expressed as T_{gp} rather than T_g in order to avoid confusion with the glass transition temperature expression. The jamming transition temperature is defined analogously in a uniformed manner, too. The particle volume fractions at jamming points are thus obtained by assuming that the ratio of the granulotemperature to the jamming temperature equals to one. The predictions from the equations of the jamming volume fractions at several cases like granular powders under a shear or a vibration are in line with experimental observations and empirical solutions in powder handlings. The goal of this article is to lay a foundation for establishing similar concepts in granular powders and then the granular powders can be described with common laws or principles we are familiar with in thermal systems. Our intention is to build up a bridge between thermal systems and granular powders for accommodating many similarities already found between those two systems.

1. Introduction

As everybody already knows, thermal energy can drive an atom or a molecule move around in gases, liquids and solids. In colloidal suspensions where small particles are dispersed in a liquid medium, the thermal energy can drive particles move around too, which is called the Brownian motion if the particle size is smaller than 1 micron. For granular powder systems where the particles are large and dispersed in air, thermal energy is too weak to move the particles and is negligible to make any contributions to particle movements. This is the big difference between conventional thermal systems and granular powder systems. However, there are many articles both experimentally and theoretically showing that granular materials behave like molecular thermal systems [1] [2] [3] [4] [5] [6] [7]. In the article titled “theory of powders”, Edwards [2] formulated a theory of granular powders based on analogies with the statistical mechanics and transport theories of regular thermal systems, and introduced the “compactivity” that is analogous to the temperature in thermodynamics. This approach was further extended to powder mixtures where the statistical mechanics was applied for mapping out phase separations [1], phase diagrams [8] [9], jamming transition and mixing separation [8] [9]. In Edwards’ analogous statistical mechanical approach, the role of energy traditionally played in thermal systems was replaced by the free volume per particle, which was found to be capable of predicting phase diagrams of jammed granular matter [9] and agreeing with the experimental results very well [10]. The force fluctuations in packed beads was experimentally found to obey a simple exponential law [11] and can be elegantly predicted with similar Edwards’ approaches [12] [13]. The extended stress ensemble mirroring the equilibrium statistical mechanics was well applied to the deformable grains [14] [15] for addressing particle packings and jamming transitions, with the experimental confirmation [16]. Not only the stress but also the force-tile area were argued to play an important role in addressing the stress distribution [17] [18], though an angularly anisotropic orientation correlation was experimentally found to be critical, too [19]. Clearly, both the experimental and theoretical evidences suggest that granular powders can be analogously treated with the methods or laws collected from thermal systems, though the traditional temperature concept should be modified accordingly for granular powders.

The “temperature” in granular powders, termed as “granulotemperature” in this article, has been addressed immediately at the beginning of utilizing thermodynamic principles. In thermodynamics, the temperature may be expressed as:

$$T = \frac{\partial E}{\partial S} \quad (1)$$

where E is the internal energy, and S is the entropy. In Edwards’ theory, the energy was replaced by the volume actually taken by the powder, V, thus Edwards’ granulotemperature was defined as:

$$T = X = \frac{\partial V}{\partial S} \quad (2)$$

Since $X = 0$ indicates that the volume of powder is not going to change with the entropy, the most compacted case, while $X = \infty$ represents the least, Edwards called this parameter as the compactivity of powders. Nonetheless, Edwards’ granulotemperature is not easy to be estimated

due to the difficulty of obtaining the entropy dependence information; In addition, the temperature defined with Eq. (2) will acquire a different unit than the traditional temperature, not very intuitive to analogously utilize the thermodynamic laws. By constructing analogous entropy and internal virial functions in granular powders equivalent to entropy and energy in thermal systems, a granulotemperature was defined very similarly to that in thermal systems as [14]:

$$T = \frac{1}{\alpha} = \frac{\partial \Gamma}{\partial S} \quad (3)$$

where S is the entropy, Γ represents the internal virial equivalent to energy, and α denotes the pre-factor in front of Γ in Boltzmann distribution. This kind of temperature definition is frequently used in nonequilibrium thermodynamic processes [20] [21]. Although the temperature definitions shown in both Eq. (2) and (3) is in line with the traditional thermodynamic temperature definition shown in Eq. (1), it is still difficult to maintain the original meaning of temperature, as kinetic energy term is missing in granular powders, in contrast with that in thermal systems where the kinetic energy is always clearly associated with temperature. Experimental and numerical results have verified that this kind of temperature definition for granular systems typically involving with slowly moving particles are actually working [9,22,23,24].

For granular systems of fast moving particles, the granulotemperatures are usually defined in consistent with the ideal gas case using the kinetic energy connection, $\frac{3}{2} k_B T = \frac{1}{2} m v^2$, where k_B is the Boltzmann constant, m is the mass of the particle, and v is the velocity of particles [25,26,27,28]. This kind of granulotemperature definitions can be easily connected back to the conventional temperature concept in thermal systems, thus the Boltzmann equation can be applied to such fast moving particle systems. Nonetheless, the distribution function is found to be reproducible, but often not Gaussian [29]. Undoubtedly, no matter which approaches are taken, the definition of temperature is always focused on and properly addressing this issue will definitely create a bridge easily connecting the traditional thermodynamic principles to nonequilibrium even athermal systems like granular powders.

The purpose of this article is to define the temperature in granular materials using kinetic energy approach via borrowing the ideas from traditional thermodynamics in very similar and consistent manners. There are two reasons of taking this approach: First, defining granulotemperature only in this way may allow us to apply the fundamental Boltzmann distribution equation to granular powders; Second, this may be the simplistic route without introducing mystery parameters like entropy and internal energy, most time unknown to a system. For avoiding possible confusions and distinguishing granular powders from traditional thermodynamics systems that are governed by thermal energy and often have the Brownian motion when the entities are smaller than 1 micron, we may term the “temperature” in granular powders as the “granulotemperature”; The corresponding dynamics built on the “granulotemperature” concept and may be called “granulodynamics”, which describes the physicochemical properties of granular powders, in analogy with “thermodynamics” that describes the energy exchange of molecular thermal systems with the surrounding environments. The granulotemperature will be expressed as T_{gp} rather than T_g , as the latter is frequently referred to the glass transition temperature in polymeric and ceramic materials fields. The side by side

comparison with the laws of thermodynamics will be used to generate the laws constituting the granulodynamics. The ultimate goal is to define the jamming temperature at which the granular systems start to jam in a uniform manner and thus the jamming phenomena can be further explored under these granulotemperature definitions.

The article is arranged as follows: We first will examine if the four thermodynamic laws can still hold for granular powders; We then consider several common cases of granular matter and define granulotemperatures using the kinetic energy approach across all cases; The granulodynamics is extended on the basis of granulotemperature, and the temperatures at jamming points are defined in a similar manner, too. The jamming volume fraction equations are thus obtained by assuming that the ratio of the granulotemperature to the jamming temperature equals to one; The predictions from the jamming volume fraction equations are focused and compared with the experimental results in literature; The future attempts based on the newly defined granulotemperatures will be discussed and the final summary and conclusions will be presented in the end.

2. Theory

In thermodynamics, there are four laws generally applied to any thermal systems [30] [31]. The zeroth law of thermodynamics states that if a thermal system A is in thermal equilibrium with a thermal system B and the thermal system B is in thermal equilibrium with a thermal system C, then thermal system A will be in thermal equilibrium with the thermal system C. The underlying implication is that if we want to know two thermal systems are at the same temperature, it is unnecessary to bring those two systems together in contact to wait for equilibration and it can be told by a third temperature medium—the thermometer that can measure the temperature. Back to granular systems, we should be able to tell if two granular systems are in equilibrium state via a granulotemperature parameter defined in such a way that the granulotemperature has a same functionality as the temperature in thermal systems. The first law of thermodynamics is about the conservation of energy: the change of internal energy of a close system is equal to the change of the heat that the system adsorbed or gave off plus the work that is done on the system or by the system. In other words, the energy cannot be created without the expense of other forms of energy or destroyed without the creation of other forms of energy. This should be true for granular systems, too, though many granular systems have a dissipative nature due to the interparticle frictional forces and inelastic collisions [7]. The second law of thermodynamics is about entropy that scales the degree of disorder or a randomness of a system. The entropy should increase over time in an isolated thermal system, approaching to a maximum value. In granular systems under a vibration or a shear field, the entropy should increase with time, too, as more particles would participate the movements due to interparticle interactions and continuous application of an external excitation. The third law of thermodynamics states that the absolute zero temperature is unattainable, as thermal motions never can stop. Unlike an ideal gas system, the particles in a granular powder cannot move freely without any external mechanical perturbation, if they are not aerated or cannot flow by themselves due to the gravity. As we know in ideal gas systems, the gas molecules can fly around due to the thermal energy, as the weights of molecules are negligible. However, in granular systems the driving force expelling particles to move is the external mechanical force or the gravitational force from particles themselves. The driving force is zero if there is no such an external mechanical force or the particles sit

quiescently, due to the cancellation of the gravitational force of particles resulted from the supporting particles that hold the particles unmovable. This by no means indicates that there is no pressure on the wall of the container and the granulotemperature is zero.

Consider a granular powder sitting inside a cylinder shown in Figure 1. As indicated by Janssen's equation [32] [33], the pressure on vertical direction, P_v , may be expressed as:

$$P_v = \frac{\rho g D}{4\mu K} \left[1 - \exp\left(-\frac{4\mu K z}{D}\right) \right] \quad (4)$$

where ρ is the density of the particle material, g is the gravity constant, D is the diameter of the cylinder, μ is the frictional coefficient between the particles and the wall of the cylinder, z is the depth where the pressure is considered, and K is the ratio of the horizontal pressure to the vertical pressure with the relationship:

$$K = \frac{P_h}{P_v} \quad (5)$$

The pressure on the bottom of the cylinder should be:

$$P_{vb} = \frac{\rho g D}{4\mu K} \left[1 - \exp\left(-\frac{4\mu K h}{D}\right) \right] \quad (6)$$

where h is the height of the powder bed. Note that the horizontal pressure at the top is equal to zero and at the bottom can be simply estimated with Eq. (5) and (6). Since the horizontal pressure is dependent on the powder depth, the average pressure may be approximately expressed as

$$P_{ha} = \frac{\rho g D}{8\mu} \left[1 - \exp\left(-\frac{4\mu K h}{D}\right) \right] \quad (7)$$

by simply adding the horizontal pressures at the top and at the bottom and then divided by two. The average pressure on the cylinder surfaces may be written:

$$P_{av} = \frac{P_{ha} + P_{vb}}{2} = \frac{\rho g D(2+K)}{16\mu K} \left[1 - \exp\left(-\frac{4\mu K h}{D}\right) \right] \quad (8)$$

There are extensive publications on utilizing kinetic gas theory to treat granular powders and the theoretical treatments are in very good agreement with experimental results [4] [7] [34] [35] [36] [37] [38] [39] [40] [41] [42] [43] [44], implying that we may be able to define granulotemperature analogously with kinetic gas theory. According to the kinetic theory of gases [45], the pressure of a gas may be expressed as:

$$P = \frac{nmv_{rms}^2}{3} \quad (9)$$

where n is the number density of molecules, $n = N/V$, with N is the number of the molecules, V is the volume, m is the mass of a molecule, v_{rms} is the root-mean-square velocity. In addition, the kinetic energy of a molecule may be expressed as:

$$\frac{1}{2}mv_{rms}^2 = \frac{3}{2}k_B T \quad (10)$$

where k_B is the Boltzmann constant. Combing Eq. (9) and (10), one may obtain the relationship between the pressure and the temperature as:

$$P = nk_B T \quad (11)$$

Eq. (11) is the ideal gas law. If one considers the pressure expressed in Eq.(8) in granular systems is caused by the imaginary particle movement, then the granulotemperature may be defined similarly as:

$$T_{gp} = \frac{\rho g D(2+K)}{16n\mu K k_B} \left[1 - \exp\left(-\frac{4\mu K h}{D}\right) \right] \quad (12)$$

Since n is a very large number, T_{gp} is expected to be very small, close to zero, which seems to be reasonable, as at such conditions there is almost no particle movement in the system. Under this temperature definition, one may claim that the absolute zero granulotemperature is unattainable, even when a whole granular system is in stationary state, which is very similar to the third law of thermodynamics. In summary, the four laws of thermodynamics may be analogically applied to granular systems with apparently different but essentially same definition of temperatures. A comparison between thermal systems and granular powders is given in Table 1.

Table 1, Four laws of thermodynamics in thermal systems and granular powders

	Thermal systems	Granular powders
The zeroth law	If $T_A = T_B, T_B = T_C$, then $T_A = T_C$	Same $T_{gp}^A = T_{gp}^C$
The first law	Conservation of energy, $\Delta E^{tot} = Q + W$, where Q is heat and W is work.	Same $\Delta E^{tot} = Q + W$
The second law	Entropy tends to increase, $\Delta S \geq 0$	Same $\Delta S \geq 0$
The third law	Absolute zero temperature is unattainable, $T \neq 0$	Same $T_{gp} \neq 0$

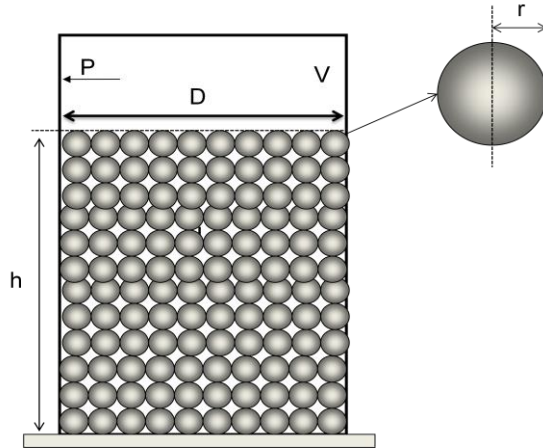


Figure 1 A granular powder sits inside a cylinder without any movement.

Now one may turn attention to define the granulotemperatures of other common cases, which are related to powder flows and tapping processes. They are shown in Figure 2: a) a powder under a simple shear; b) a powder rolling on a slope; and c) a box of a powder under a vibration. First, let's consider a very simple granular system—a box of the volume V with many spheres sitting inside shown in Figure 2 (c). Since the spheres have nonnegligible weights, they will generate a pressure on the bottom of the box and the sides of box, too. As shown earlier in Eq. (4), the pressures on the sides should differ from the total weight of all spheres. The whole box is fixed on a plate that can move horizontally back and forth with a vibration expressed as $L = L_0 \exp(i\omega t)$, where L_0 is the amplitude of vibration, ω is the angular frequency, and t is the

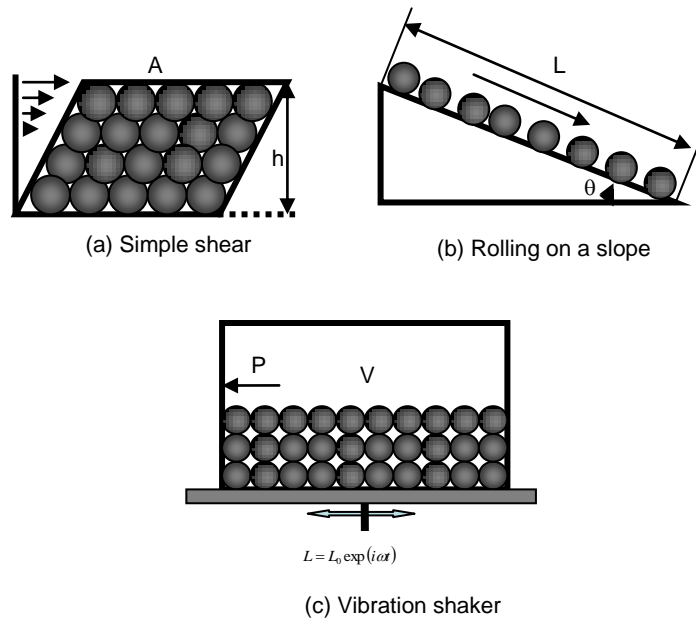


Figure 2 Granular systems under (a) a simple shear; (b) rolling by themselves; (c) under horizontal vibration $L = L_0 \exp(i\omega t)$.

time. When an external vibration is not applied to the granular system, all particles are stationary and at this moment the granulotemperature is very close to zero, as indicated in Eq. (12). The entropy of the whole system should be very small too. When an oscillatory vibration is applied as $L = L_0 \exp(i\omega t)$, the energy flow rate to the granular system may be calculated as [46]:

$$\dot{E} = F(t) \cdot v(t) \quad (13)$$

where $F(t)$ and $v(t)$ are the force and velocity at the interface, respectively. One may assume that $F(t) = Mg$, i.e., the force is equal to the weight of whole spheres inside the box. The $v(t)$ may be expressed as the amplitude divided by the time within a cycle

$$v(t) = \frac{L_0}{1/(\omega/2\pi)} = \frac{L_0\omega}{2\pi} \quad (14)$$

The energy flow rate from the vibration shaker to the granular system is thus expressed as:

$$\dot{E} = \frac{MgL_0\omega}{2\pi} \quad (15)$$

If the number of vibration is assumed to be n_v , then the total time spent in vibration may be expressed below:

$$t = n_v \times \left(\frac{1}{\omega/2\pi} \right) = \frac{2\pi n_v}{\omega} \quad (16)$$

The total energy flowing into the powder system may be expressed as:

$$E = \dot{E} t = MgL_0 n_v \quad (17)$$

Eq. (17) may indicate that the total energy transferred into the powder system is independent of the frequency of vibration, and only dependent of the amplitude of vibration. According to the kinetic theory of gases [45], the kinetic energy of a molecule may be expressed as Eq. (10). If the number of molecules is N , then the total kinetic energy is

$$E = \frac{1}{2} m v_{rms}^2 N = \frac{3}{2} N k_B T \quad (18)$$

As indicated earlier, there are a large number of theoretical treatments of granular flows using the analogy of molecular fluids via standard statistical mechanics and kinetic theory, which are generally in a good agreement with the experimental results [4] [7] [34] [35] [36] [37] [38] [39] [40] [41] [42] [43] [44] [47] [48] [49] [50] [51]. We thus continue to utilize the kinetic theory to analogously define the granulotemperatures. Assume that the energy flowed to the granular

system contributes to the movement of particles inside the box. Replacing the temperature in Eq. (18) with the granulotemperature, one may easily reach

$$E = MgL_0n_v = \frac{3}{2} Nk_B T_{gp} \quad (19)$$

If the particles have the true density of ρ and radius r , then

$$M = N \frac{4}{3} \pi r^3 \rho \quad (20)$$

Substituting Eq. (20) into Eq. (19) and re-arranging may lead to the granulotemperature of particles under a vibration:

$$T_{gp} = \frac{8}{9} \frac{\pi r^3 \rho g L_0 n_v}{k_B} \quad (21)$$

The granulotemperature defined in Eq. (21) has a unit of Kelvin, same as the regular temperature for thermal systems. For one micrometer sized particles of density 1g/cm^3 under a vibration, $L_0 = 1\text{ cm}$, $n_v = 1000$, the granulotemperature expressed in Eq. (21) is equal to 1.98×10^{10} K, a very high temperature in comparison with the temperature in thermal systems, However, in thermal systems the molecules or particles usually travel in sub-micrometer scale, while in vibrated granular powders particles may travel in a full distance of vibration amplitude, a centimeter scale. The traveling distance difference between those two movements is approximately in the order of $10^5 \sim 10^7$, which makes the granulotemperature is relatively on a par with the conventional thermal temperature.

Note that the temperature defined above is only appropriate for granular systems with an external vibration excitation. If a granular system is under a simple shear as shown in Figure 2(a), the granulotemperature should be defined differently, as the energy flowing into the granular system is different. Supposing that the shear stress is σ and the shear rate is $\dot{\gamma}$, then the force F and the velocity v may be expressed as:

$$F = \sigma A, \quad v = \dot{\gamma} h \quad (22)$$

where A is the area of the sample and h is the thickness of the sample. On the basis of Eq. (13), the injected energy flowing rate from a simple shear field may be expressed as:

$$\dot{E} = Fv = \sigma \dot{\gamma} Ah = \sigma \dot{\gamma} V \quad (23)$$

where V is the volume of the granular system, $V = M / \rho_b = \frac{4}{3} \pi r^3 N \rho / \rho_b$, and ρ and ρ_b are the true and bulk density of the granular powder, respectively. Using Eq. (18), the granulotemperature of a sheared powder after a time period of t may be expressed as:

$$T_{sp} = \frac{8 \pi \sigma r^3 \dot{\gamma} \rho t}{9 k_B \rho_b} \quad (24)$$

Again, assuming one micrometer sized particles of true density 1g/cm^3 , bulk density 0.3g/cm^3 , under a shear field, $\sigma = 1\text{Pa}$, $\dot{\gamma} = 1\text{Hz}$, and shearing for 5 min., $t = 300\text{s}$, the granulotemperature expressed in Eq. (24) is equal to $2.02 \times 10^8\text{K}$, still a very high temperature.

If granular spheres flow over a slope as shown in Figure 2 (b), the granulotemperature should be defined differently, too. The force that drives spheres to move downward should be $mg \sin \theta$, where m is the mass of a sphere, θ is the angle of slope. If the friction coefficient between the particles and the slope surface is μ , the frictional force should be $\mu mg \cos \theta$. The net force on a particle may be expressed as:

$$F = mg \sin \theta - \mu mg \cos \theta \quad (25)$$

According to Newton's second law, $F = ma$, where a is acceleration, one may find

$$a = g(\sin \theta - \mu \cos \theta) \quad (26)$$

The initial velocity of a sphere at the top of the slope is zero and at the time t the velocity is assumed to be v , thus

$$\frac{v}{t} = a \quad (27)$$

which is the definition of acceleration. Us the energy defined in Eq. (17), i.e., the energy is the energy rate multiplied by the time:

$$E_p = Fvt = mg^2(\sin \theta - \mu \cos \theta)^2 t^2 \quad (28)$$

Eq. (28) gives the energy of one single particle. For a granular powder containing N particles, the total energy may be expressed as:

$$E = E_p N = Nmg^2(\sin \theta - \mu \cos \theta)^2 t^2 \quad (29)$$

Using Eq. (18) again, one may obtain granulotemperature for spheres on a slope

$$T_{gp} = \frac{2}{3k_B} mg^2(\sin \theta - \mu \cos \theta)^2 t^2 \quad (30)$$

For particles of radius r , $m = \frac{4}{3}\pi r^3 \rho$, so Eq. (30) may be further written as:

$$T_{gp} = \frac{8\pi r^3 \rho}{9k_B} g^2 (\sin\theta - \mu \cos\theta)^2 t^2 \quad (31)$$

Assuming one micrometer sized particles of true density 1 g/cm^3 , $\theta = 45^\circ$, $t = 10 \text{ s}$, $\mu = 0.3$, then the granulotemperature defined in Eq. (31) is equal to $4.76 \times 10^{11} \text{ K}$, even much higher temperature than the one defined in vibration conditions. Note that this definition is suitable for very idealized conditions where there is no interparticle collisions and the particle can move freely under the gravitational force, which leads to a very high granulotemperature. The actual net force and acceleration should be much smaller than that indicated in Eq. (25) and (26). If there is 10% reduction in both force and acceleration due to the resistance from other particles, the obtained granulotemperature would be 100 times smaller, which sets the granulotemperature is on par with that defined in vibration and shear cases.

3. Particle jamming and associated temperatures

Jamming is a very common phenomenon in granular powders, where particles suddenly stop moving due to the strong connectivity or interaction between particles in a constrained space [9] [52] [53] [54] [55]. It is very similar to the frozen phase transition in thermal systems, where a liquid state is transited to a solid state due to the temperature drops, and the whole system changes from a free flow state to a solidified stationary state [55,28,56]. It would be interesting to evaluate the granulotemperatures at jamming points based on the definitions proposed earlier. Since the granulotemperature attains a very similar functionality as the conventional temperature, we thus may analogously assume that the “thermal” energy from the granulotemperature is the source of particle motions and thus associated with the particle jamming, too. The jamming will be defined as a phenomenon when particles are unable to travel the allowed free distance on the basis of the free volume in a granular system. The interparticle spacing (IPS) of a granular powder may be expressed as [57,58]:

$$\text{IPS} = 2(\sqrt[3]{\phi_m/\phi} - 1)r \quad (32)$$

where ϕ_m is the maximum packing fraction, ϕ is the particle volume fraction, r is the particle radius. At a free flowing unjammed state, particles are supposed to have the energy capable of travelling the full distance shown in Eq. (32). However, at jammed states, particles don't have such a sufficient energy and are assumed to be capable of “vibrating” within the half of the distance expressed above. Note that the IPS equation above is derived on the basis of Kuwabara's cell model [59] and the half the IPS distance means that there is a great extent of virtual cell overlap between two particles and basically these two particles touch each other. Under such a definition of jamming state, the energy required for N particles to move a half the IPS distance may be expressed as:

$$E = Nm g (\sqrt[3]{\phi_m/\phi_j} - 1)r \quad (33)$$

where ϕ_j is the particle volume fraction when particles are jammed. According to Eq. (18), the energy shown in Eq. (33) should be equal to the kinetic energy for particles, which has been used in this article many times for defining the granulotemperatures. Therefore, the granulotemperature at jamming points may be expressed as:

$$T_J = \frac{2mg\left(\sqrt[3]{\phi_m/\phi_j - 1}\right)r}{3k_B} \quad (34)$$

where T_J is the granulotemperature at a jamming point. Since $m = \frac{4}{3}\pi r^3\rho$ by the definition, Eq. (34) may be further written as:

$$T_J = \frac{8\pi\rho g\left(\sqrt[3]{\phi_m/\phi_j - 1}\right)r^4}{9k_B} \quad (35)$$

Since ϕ_m is only related to the packing structure for monodispersed particle systems [52,60], one may infer that the jamming transition temperature is dependent on the density of the particulate materials, the radius of the particles, and the particle volume fractions at jamming points. For obtaining an intuitive idea how the jamming transition temperatures change with the particle volume fractions at jamming points, we schematically plot Eq. (35) at several different particle radii in Figure 3, under assumption that $\rho=1 \text{ g/cm}^3$, $\phi_m = 0.72$. The jamming temperature generally decreases with the increase of particle volume fractions at jamming points and are strongly dependent on the particle sizes. There are several orders of magnitudes difference among the jamming temperatures when the particle sizes only increase 10 times. This is probably due to the fact that the jamming temperatures are directly proportional to the 4th powers of the particle radius as indicated in Eq. (35). When the particle volume fractions at jamming points approach to the maximum volume fraction, the jamming temperatures quickly drop to a very low temperature, no matter that the particle sizes are large or small. In addition, Fig.3 shows small particles tend to jam at very low granulotemperatures, which is consistent with practical observations: large granules usually flow much better than small particles. When particles become smaller and smaller, interparticle forces become more important and particles tend to aggregate or bridge very easily, resulting in a very poor flowability. As one may tell, for particles of radius about 0.1 micron, the jamming transition temperature is little bit below one

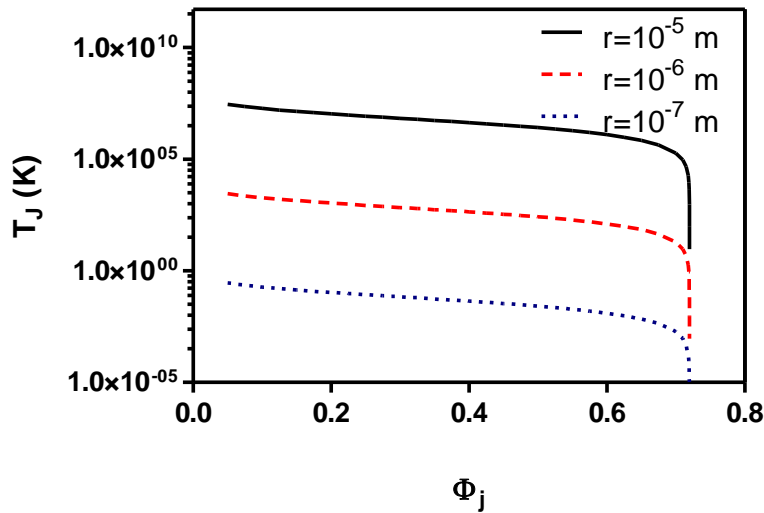


Figure 3, Jamming transition temperature vs. the jamming volume fractions under several particle radii based on Eq. (35), under assumption that $\rho=1 \text{ g/cm}^3$, $\phi_m = 0.72$.

Kelvin, an extremely low temperature. While in comparison with 10 micron sized particles the jamming happens at a very high temperature, about 10^7 K . This is mainly resulted from the fact that the jamming temperature is defined if the external energy that drives particles to move can overcome the weight of particles over a certain distance in granular systems. This prediction seems to qualitatively agree with the experimental observation on the superheating phenomena of monodispersed metal beads of diameter 3.15 mm reported in literature [6]: Under a vigorous vertical shaking, a hexagonal closed packed crystal structure was observed and eventually melted away (or called evaporate in the literature) after a period of time. As stated earlier, a granular powder under a vibration may have a very high granulotemperature. Based on Eq. (21), the granulotemperature of such a metal beads system is in the order of 10^{20} K , and also, it is time dependent. Longer time vibrations will create higher granulotemperatures, which could be the reason that the crystal structure was finally evaporated after a relatively long vibration. For particles of size about 0.1 micron, jamming should happen at much lower granulotemperatures about one Kelvin based on Fig. 3. Such a low granulotemperature may correspond to a quiescent state where no apparent motions are obviously detected. In reality, submicron or nanometer sized powders of low densities typically tend to have a very poor flowability and easily form arching structures [61,62]. The newly defined granulotemperatures seem to agree well with the empirical observations.

It would be very valuable to explore at what conditions that the jamming could happen by simply using the granulotemperatures defined earlier at several common cases divided by the jamming granulotemperature defined in Eq. (35). The ratio equal to one gives the jamming conditions for particular granular systems. For a simple shear case:

$$\frac{T_{gp}}{T_j} = \frac{\sigma \dot{\gamma} t}{(\sqrt[3]{\phi_m/\phi_j - 1})gr\rho_b} = 1 \quad (36)$$

Thus one may easily get:

$$\phi_j = \frac{(gr\rho_b)^3 \phi_m}{(gr\rho_b + \sigma \dot{\gamma} t)^3} = \frac{\phi_m}{[1 + \sigma \dot{\gamma} t / (gr\rho_b)]^3} \quad (37)$$

Eq. (37) defines the conditions that the jamming happens at a simple shear case. It clearly tell that the particle volume fractions at the jamming points are dependent on the shear stress, shear rate, and surprisingly the radius of the particles. For illustrative purpose, ϕ_j is plotted against $\dot{\gamma}$ over a wide range and shown in Figure 4, under the assumption that $\phi_m = 0.72$, $\rho_b = 0.3 \text{ g/cm}^3$, $t=60 \text{ s}$, $\sigma = 1 \text{ Pa}$, $r=10^{-3}$, 10^{-4} , and 10^{-5} m . Note that there are two regions where ϕ_j is insensitive to shear rates, very high shear rates above $10^2/\text{s}$ and very low shear rates below $10^{-5}/\text{s}$. In the shear rate ranges between 10^{-5} - $10^2/\text{s}$, ϕ_j dramatically increases with the decrease of shear rates. In other word, the jamming may happen at lower particle volume fractions when the shear rate increases, which is consistent with experimental observations [53,63]. In addition, apparently there seems to be two regions existing in the system: when the shear rate is smaller

than 10^{-5} 1/s, the system is fully jammed; when the shear rate is larger than 10^{-5} 1/s, the volume fractions at jammed points become lower and lower under higher and higher shear rates. The system starts to jam when the shear rate is about 10^{-2} 1/s. The big jump between 10^{-5} - 10^{-2} 1/s may indicate that a shear induced structure change happens in this area. These two regions are very similar to the “fragile states” and “shear-jammed states” observed experimentally [53,63], where the “fragile states” correspond to a strong network structure percolated in one direction and the “shear-jammed states” correspond to a strong network percolated in all directions. Back to Figure 4, the “fragile states” is somewhat similar to the big jump region between 10^{-5} - 10^{-2} 1/s, while “shear-jammed states” is the region where the shear rate is below 10^{-5} 1/s. The qualitative agreement with the experimental observation may imply that the granulo-temperature defined in a consistent manner with the conventional temperature in thermal systems is actually working.

The particle radius has a clear impact on the jammed volume fraction, too, based on Eq. (37) and shown in Figure 4. Smaller particles can only jam at lower shear rates and melt at higher shear rates; Larger particles wouldn't jam at lower shear rates, unless the particle volume fractions are close to the maximum packing fraction. For clearly demonstrating the influence of particle size on the jamming volume fraction, the particle volume fraction at the jamming points, ϕ_j , plotted in Figure 5 against the particle radius, r , under several different shear rates, $\dot{\gamma}$, from 10^{-2} to 10^{-5} 1/s, under the assumption that $\phi_m = 0.72$, $\rho_b = 0.3$ g/cm³, $t=60$ s, and $\sigma = 1$ Pa.

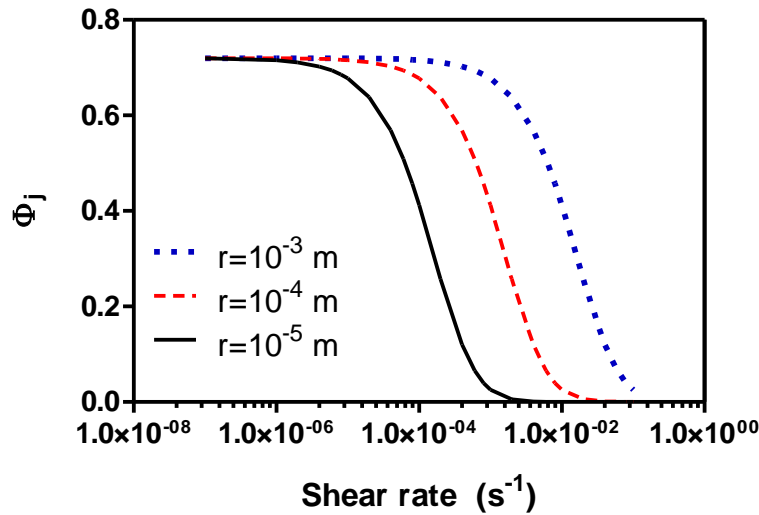


Figure 4, The particle volume fraction at the jamming points, ϕ_j , is plotted against the shear rate, $\dot{\gamma}$, from Eq. (37) under the assumption that $\phi_m = 0.72$, $\rho_b = 0.3$ g/cm³, $t=60$ s, $\sigma = 1$ Pa, $r = 10^{-3}$, 10^{-4} , and 10^{-5} m.

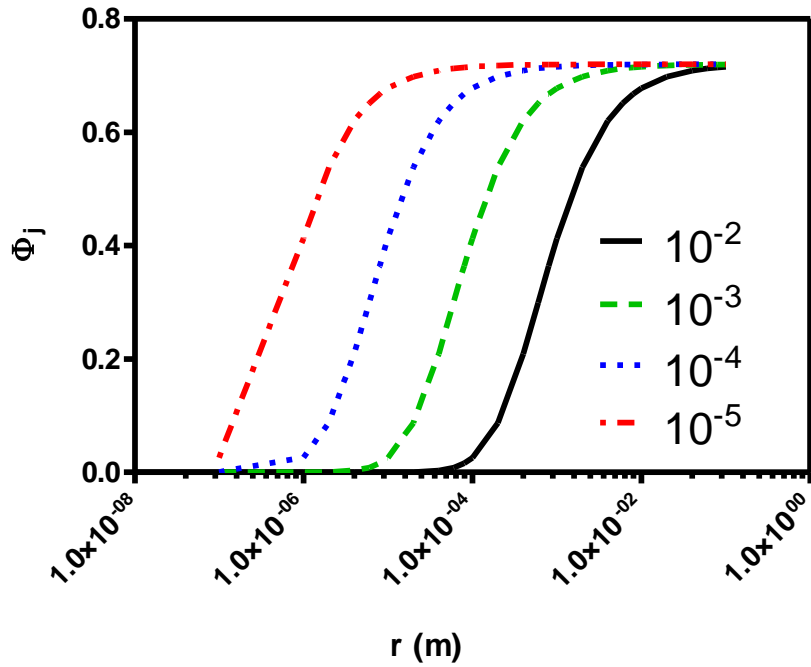


Figure 5, The particle volume fraction at the jamming points, Φ_j , is plotted against the particle radius, r , under several different shear rates, $\dot{\gamma}$, from 10^{-2} to 10^{-5} 1/s, obtained from Eq. (37) under the assumption that $\Phi_m = 0.72$, $\rho_b = 0.3$ g/cm³, $t=60$ s, and $\sigma = 1$ Pa.

When the particles have a size in micron scale, the jamming can only happen at low shear rates. At high shear rates only particles that have a large particle size may jam. In other words, high shear rates would break jams easily and smaller particles don't have a chance to jam. When particle volume fractions are close to the maximum packing fraction, particles may always jam no matter that they have a large or small particle size. Under a shear field, the jamming is a phenomenon dependent on both the particle sizes and the shear rates, which are two major parameters determining the granulotemperature as shown in Eq. (24).

Similarly, one may find the jamming volume fraction conditions for granular powders under a vibration. Using Eq. (21) divided by Eq. (35) and assuming that it equals to 1 leads:

$$\frac{T_{gp}}{T_j} = \frac{L_o \omega t}{2\pi r (\sqrt[3]{\Phi_m/\Phi_j} - 1)} = 1 \quad (38)$$

Thus the particle volume fraction at jamming points may be expressed as:

$$\Phi_j = \frac{\Phi_m}{[1 + L_o \omega t / (2\pi r)]^3} \quad (39)$$

Eq. (39) indicates that the particle jamming volume fractions are dependent on the amplitude and frequency of the vibration, the time, and the particle radius. For clearly illustrating the relationship between Φ_j and the particle radius, these two parameters are plotted in Figure 6 and

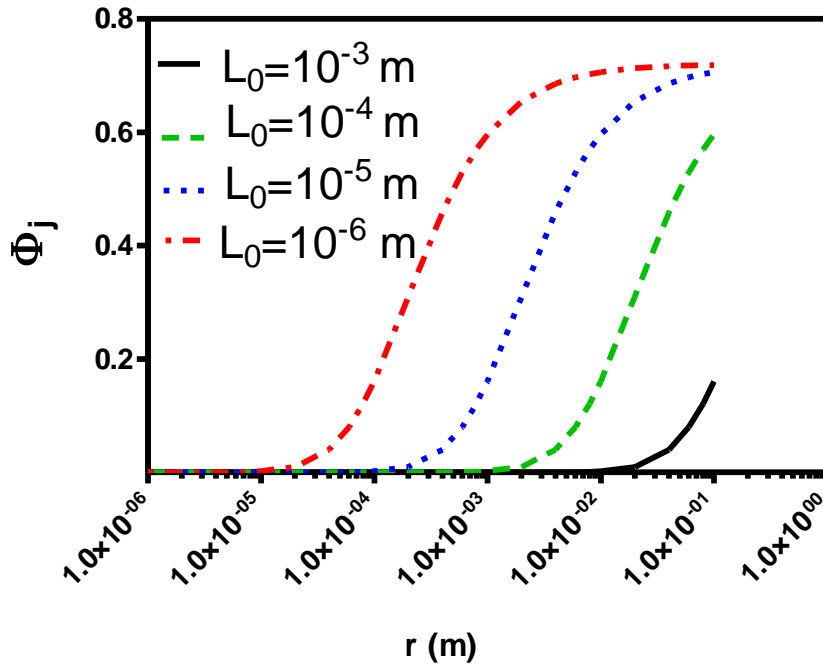


Figure 6, The particle volume fraction at the jamming points, Φ_j , is plotted against the particle radius, r , under several different vibration amplitudes from 10^{-3} to 10^{-6} m, obtained from Eq. (39) under the assumption that $\Phi_m = 0.72$, $\omega = 6.8$ rad/s, $t=60$ s.

computed with Eq. (39). With the increase of particle sizes, particles tend to only jam at higher vibration amplitudes. In other words, particles of smaller particle sizes tend to only jam at relative small vibration amplitudes. Both the vibration amplitude and the particle radius play significant roles in determining the critical jamming points. Again, only at very high particle volume fractions close to the maximum particle volume fraction, the jamming happens independent of the particle radii, which is similar to what is demonstrated for granular powders under a shear. Figure 7 shows the particle volume fraction at the jamming points, Φ_j , plotted against the vibration amplitude under several different particle radii from 10^{-4} to 10^{-1} m, obtained from Eq. (39) under the assumption that $\Phi_m = 0.72$, $\omega = 6.8$ rad/s, $t=60$ s. No matter what the particle sizes are, particles tend to jam at very high particle volume fractions close to the maximum volume fraction. However, when the vibration amplitude increases, jammed particles tend to melt and then jam at lower particle volume fractions. This “melt-jam” meta-stable region is very narrow, implying that the granular powders may quickly melt with the increase of the vibration amplitudes. Since the vibration frequency locates at the identical position as the vibration amplitude in Eq. (39), a very similar dependency of the jamming particle volume fractions on the vibration frequency is expected, i.e., high vibration frequencies may melt the jammed particles, too. In industries, vibration conveyors with controllable amplitudes and frequencies are frequently employed to transport granular powder materials. The results expressed in Eq. (39) and demonstrated in Figures 6 and 7 seem to be consistent with the empirical practical solutions that have been used for a long time.

It would be interesting to see how the vibration time would impact on the jamming particle volume fractions, as longer time means more energy flowing into the systems indicated in Eq. (21). Figure 8 shows the particle volume fractions at the jamming points, Φ_j , plotted against the vibration time under several different

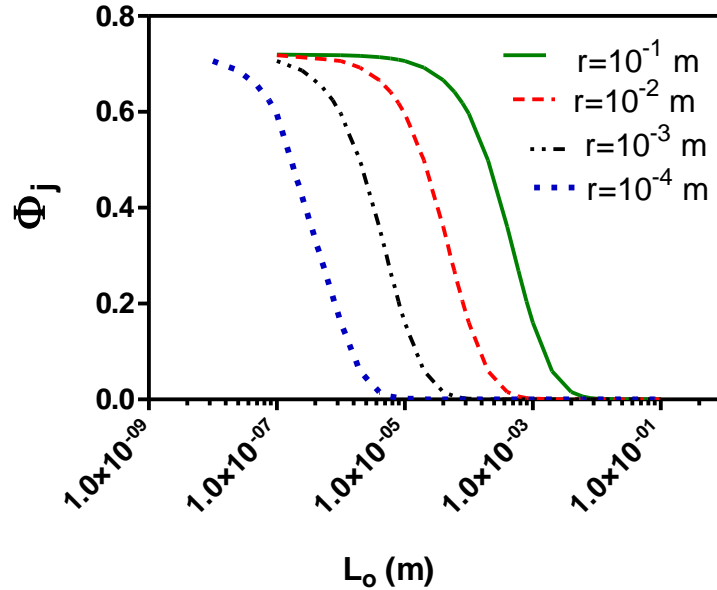


Figure 7, The particle volume fraction at the jamming points, Φ_j , is plotted against the vibration amplitude under several different particle radii from 10^{-4} to 10^{-1} m, obtained from Eq. (39) under the assumption that $\Phi_m = 0.72$, $\omega = 6.8$ rad/s, $t=60$ s.

vibration amplitudes from 10^{-3} to 10^{-6} m, obtained from Eq. (39) under the assumption that $\Phi_m = 0.72$, $\omega = 6.8$ rad/s, $r=0.001$ m. Again, the particles may jam at the very beginning of the vibration when particle volume fractions are high enough, and quickly enter into “melt-jam” regions. At lower vibration amplitudes, the “melt-jam” cycles a relative long time, while at high vibration amplitudes, jammed particles may rapidly melt in much faster paths. Again, particle sizes should play a role in determining where the jamming points are. Figure 9 shows particle volume fractions at the jamming points, Φ_j , plotted against the vibration time under several different particle radii from 10^{-3} to 10^{-1} m, obtained from Eq. (39) under the assumption that $\Phi_m = 0.72$, $\omega = 6.8$ rad/s, $L_0=0.0001$ m. At such a small vibration of amplitude 10^{-4} m, particle sizes are critical: small particles can easily melt and larger particles may go into the “melt-jam” cycles for a long time, before they completely melt. Particles of size below 10^{-3} m may start to jam at low particle volume fractions, but can melt easily if a small vibration amplitude is continuously provided. Again, those speculations are in consistent with experimental observations reported in literature [6], where the crystalline structure was observed at the very beginning of vibration and quickly melt later with a continuous vibration.

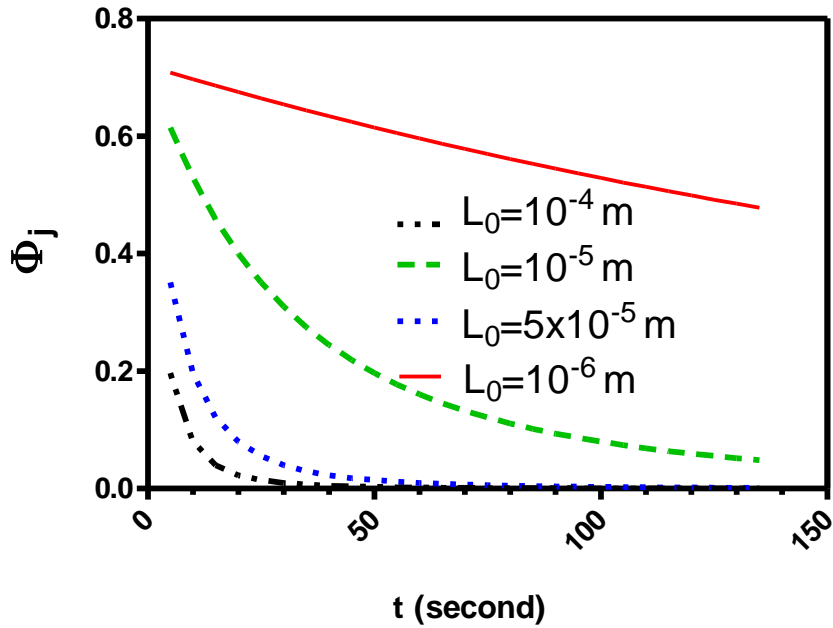


Figure 8, The particle volume fraction at the jamming points, Φ_j , is plotted against the vibration time under several different vibration amplitudes from 10^{-3} to 10^{-6} m, obtained from Eq. (39) under the assumption that $\phi_m = 0.72$, $\omega = 6.8$ rad/s, $r=0.001$ m.

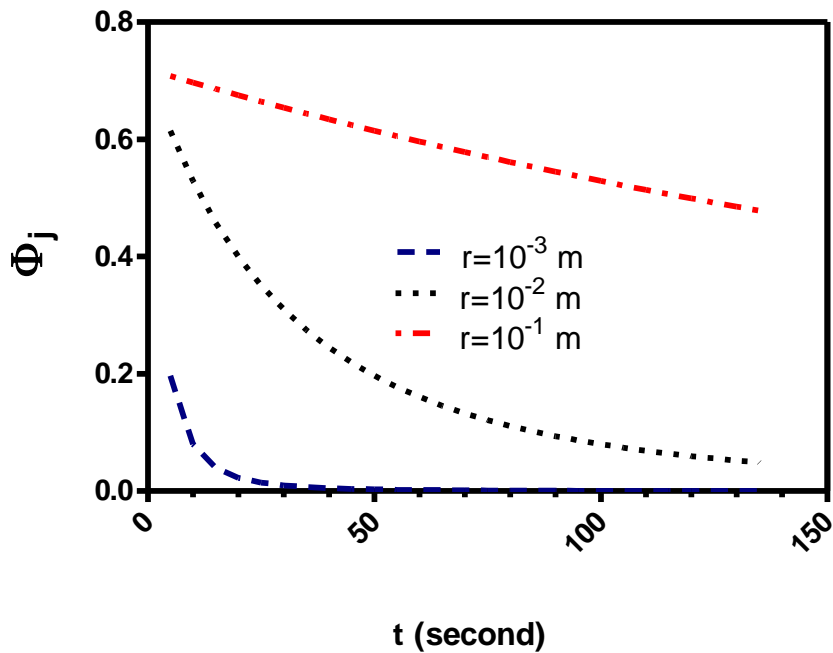


Figure 9, The particle volume fraction at the jamming points, Φ_j , is plotted against the vibration time under several different particle radii from 10^{-3} to 10^{-1} m, obtained from Eq. (39) under the assumption that $\phi_m = 0.72$, $\omega = 6.8$ rad/s, $L_0=0.0001$ m.

Following the same procedures described previously, one may easily obtain the particle volume fraction at jamming points for particles rolling on a slope:

$$\phi_j = \frac{\phi_m}{\left[1 + \frac{gt^2}{r}(\sin\theta - \mu\cos\theta)\right]^3} \quad (40)$$

and the particle volume fraction at jamming points for particles sitting inside a cylinder without any movement:

$$\phi_j = \frac{\phi_m}{\left[1 + \frac{9D(2+K)}{96\mu Kr\phi_A} \left[1 - \exp\left(-\frac{4\mu Kh}{D}\right)\right]\right]^3} \quad (41)$$

where $\phi_A = \frac{4\pi r^3 N}{3V}$, is the real particle volume fraction excluding all interstitial empty spaces in the cylinder, $\phi_A \geq \phi_m$. Readers are encouraged to explore the relationships among the particle volume fractions at jamming points and other related parameters under those two cases.

4. Discussion

Although granular powders are athermal systems, utilization of thermodynamics and statistical mechanical theories extracted from conventional thermal systems to treat granular powders are found to be in good agreement with experimental results [12,28,58,64]. We thus examine the applicability of the four laws of thermodynamics on granular powder systems, and define granulotemperatures for several granular powder systems in an analogous manner. The key point is to define the temperatures in granular powders through the kinetic energy connection with temperature, as shown in ideal gases. The main goal is to establish an approach that can facilitate the easy applications of thermodynamic principles to granular powder systems. Such attempts have been made before for addressing both wet particle systems like colloidal suspensions and dry particle systems like granular powders. For examples, Hao [57] has successfully used the Eyring's rate theory [65] and the free volume concept for obtaining the viscosity equations of colloidal suspensions and polymeric systems with substantial modifications; A very similar theoretical approach is successfully employed to derive the two popular empirical tap density equations, the logarithmic and stretched exponential equations [58,64]. All these successes evidenced in literature imply that both thermal and athermal systems can be well described with common thermodynamic principles. What we need is a bridge that can build up a uniform connection between those two systems. This article represents the first attempt in this direction and further refinements are expected. Future attempts will be to utilize the Eyring's rate process theory and free volume concept to treat the granular systems for the purpose of deriving viscosity equations of granular systems under various conditions. Similar methods and approaches shown in our previous publications will be employed again to treat granular powder systems in a much natural manner once the granulotemperatures are properly defined.

5. Summary and conclusions

In summary, the thermodynamics originated from thermal systems is utilized to define the granulotemperatures in granular systems in an analogous manner. The key point is to connect the kinetic energy to the temperature, and thus the temperature can be defined in a uniformed manner across the conventional thermal systems like colloidal suspensions to athermal systems like granular powders. This is a necessary step, as in granular systems thermal energy is too small to drive the granular particles move around; New temperature definitions are needed for properly applying the thermodynamic principles established in thermal systems to granular systems. Several common granular systems are analyzed and the defined granulotemperatures are summarized in Table 2. The obtained granulotemperatures seem to be very high in comparison with the temperatures in thermal systems. However, please keep in mind that in conventional thermal systems, the molecule movements are very mild in much smaller distance scales; Lower temperatures seem to be adequate for thermal systems. On the other hand, the particle movements in granular powders are typically very intensive and wild, and higher granulotemperatures seem to be adequate.

Once the granulotemperatures are defined, the jamming temperature is analogously defined, too. The jamming particle volume fractions are thus obtained by assuming that the ratio of the granulotemperatures to the jamming temperature equals to one. Therefore, the jamming points can be predicted and the obtained results agree qualitatively very well with experimental observations and empirical solutions in powder handlings. The particle volume fractions at jamming points obtained at several common cases are listed at Table 2.

Table 2 Proposed granulotemperatures and the particle volume fractions at jamming points predicted in several granular systems.

Granular systems	Granulotemperatures	Typical values (K)	Conditions	Particle volume fractions at jamming points
Powders in stationary bins	$T_{gp} = \frac{\rho g D (2 + K)}{16 n \mu K k_B} \left[1 - \exp\left(-\frac{4 \mu K h}{D}\right) \right]$	Very close to zero	The particle number density n is a very large number	$\phi_j = \frac{\phi_m}{\left[1 + \frac{9D(2+K)}{96\mu K r \phi_A} \left[1 - \exp\left(-\frac{4\mu K h}{D}\right) \right] \right]^3}$
Powders in vibrations	$T_{gp} = \frac{8 \pi r^3 \rho g L_0 n_v}{9 k_B}$	1.98×10^{10}	$r = 1 \mu\text{m}$, $\rho = 1 \text{g/cm}^3$, $L_0 = 1 \text{cm}$, $n_v = 1000$	$\phi_j = \frac{\phi_m}{[1 + L_0 \omega t / (2\pi r)]^3}$
Powders under a shear	$T_{gp} = \frac{8 \pi \sigma r^3 \dot{\gamma} \rho t}{9 k_B \rho_b}$	2.02×10^8	$r = 1 \mu\text{m}$, $\rho = 1 \text{g/cm}^3$, $\rho_b = 0.3 \text{g/cm}^3$, $\sigma = 1 \text{Pa}$,	$\phi_j = \frac{\phi_m}{[1 + \sigma \dot{\gamma} t / (g r \rho_b)]^3}$

			$\dot{\gamma} = 1 \text{ Hz}$ $t = 300 \text{ s}$	
Particles rolling on a slope	$T_{gp} = \frac{8\pi r^3 \rho}{9k_B} g^2 (\sin\theta - \mu \cos\theta)^2 t^2$	4.76×10^{11}	$r = 1 \mu\text{m}$, $\rho = 1 \text{ g/cm}^3$, $\theta = 45^\circ$, $t = 10 \text{ s}$ $\mu = 0.3$	$\phi_j = \frac{\phi_m}{\left[1 + \frac{gt^2}{r} (\sin\theta - \mu \cos\theta)\right]^3}$

The work in this article may lay a foundation for building up the “granulodynamics” on the basis of the granulotemperatures defined analogously with that in thermodynamics. The four laws of thermodynamics are applicable to the granular powders with such definitions. Since the most important jamming phenomena in granular powders under a shear and a vibration are intensively examined, the results presented in this article may provide further insights on how to efficiently control the jamming process that has vast and important applications in industries like soft robotics and architecture [52].

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