Dynamics of electrostatically driven granular media: Effects of humidity

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(Received 31 January 2001; published 19 April 2001)

We performed experimental studies of the effect of humidity on the dynamics of electrostatically driven granular materials. Both conducting and dielectric particles undergo a phase transition from an immobile state (granular solid) to a fluidized state (granular gas) with increasing applied field. Spontaneous precipitation of solid clusters from the gas phase occurs as the external driving is decreased. The clustering dynamics in conducting particles is primarily controlled by screening of the electric field but is aided by cohesion due to humidity. It is shown that humidity effects dominate the clustering process with dielectric particles.

Driven granular materials exhibit complex behavior that resembles some aspects of conventional solids, liquids, and gases, yet also demonstrate some considerable differences [1–4]. A key distinction that separates granular phases from their conventional counterparts is the presence of dissipation of energy through inelasticity of collisions and friction. For the gaslike phase, dissipation may be responsible for non-Maxwellian velocity statistics [5], and may cause the fluidized state to collapse locally, forming clusters of particles at rest [6]. There have been some studies of this spatial inhomogeneity where energy is added by mechanically vibrating the system [7–9]. Environmental effects, such as humidity and surface coating, are additional factors making the physics of granular systems especially complex.

Recent studies have explored humidity effects in static granular materials consisting of large particles (about 1 mm) [11–16], but little has been done to examine the effect on dynamic systems. This wetting effect intervenes in the dynamics causing agglomeration, charging, etc.

Dynamics of large ensembles of small (below 30 μm) particles is even more challenging. There is a trend in technology to operate with finer particles [17]. Fascinating collective behavior appears when small particles acquire an electric charge and respond to competing long-range electromagnetic and short range contact forces, e.g., due to wetting. Unfortunately, traditional methods to probe the dynamics of granular materials through mechanical excitation become inefficient for very small particles. Fine particles are more sensitive to electrostatic forces which arise through particle friction or space charges in the particle environment. Their large surface to volume ratio also amplifies the effect of water or other surfactants. While dry grains interact only through repulsive contact forces, wet grains attract each other through adhesion of liquid bridges between grains.

Recently, we studied the dynamics of granular materials consisting of small conducting particles through the technique of electrostatic excitation [10]. Electrostatic driving makes use of these *bulk* forces, and allows control by long-range electric forces. This technique enables one to deal with extremely fine powders. In Ref. [10] we reported on the discovery of phase transitions in electrostatically driven granular material consisting of less than a single layer of conducting particles.

In this Rapid Communication we focus on the effects of controlled humidity on the dynamics of electrostatically driven granular materials. Studies were performed on both conducting and dielectric particles. We show that humidity does not qualitatively change the phenomenology of conducting particles. In contrast, the behavior of dielectric particles changes drastically. In particular, the dielectric medium shows dendritic clusters at high humidity, whereas the conducting medium exhibits cluster coarsening irrespective of humidity.

Apparatus. Our experimental setup is similar to that in Ref. [10], although important modifications were made to control the moisture level inside the experimental cell (see Fig. 1). Particles are placed between the plates of a large capacitor which is energized by a constant or alternating electric field. To provide optical access to the cell, the upper plate is made of glass with a clear conducting coating. We used 11×11 cm capacitor plates with a spacing of 1.5 mm. The particles consisted of 165 ± 15 μm and 35 ± 5 μm conducting copper spheres and 165 ± 15 μm and 85 ± 5 μm
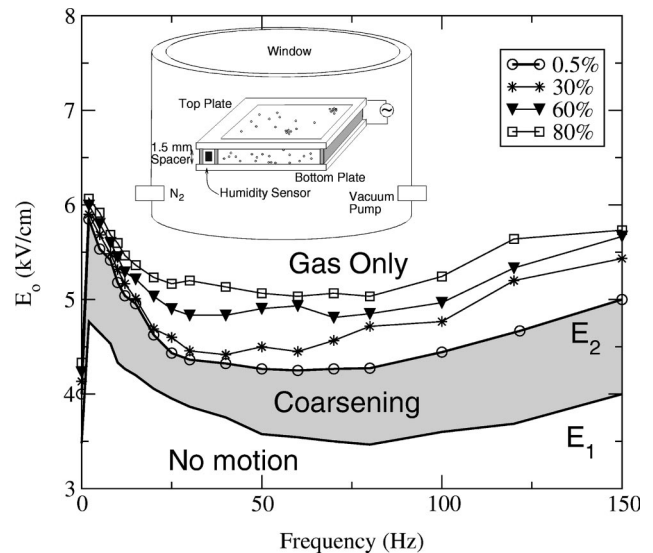

FIG. 1. Phase diagram in the (f,E_0) plane for 165 μm spherical copper particles. Lines with symbols show E_2 for different humidity levels. The solid line indicates E_1 for 0.5% humidity. In the *gas only* part clusters do not form, and in the *coarsening* part clusters and gas coexist. Inset: scheme of the experimental setup.

glass spheres. The field amplitude varied from 0 to 10 kV/cm and the frequencies on the interval of 0 to 150 Hz. The number of particles in the cell varied between 10^5 and 10^6.

To control the humidity of the system, the amount of water vapor was held constant by placing the apparatus in a sealed chamber. Before starting the experiments, the chamber was evacuated to approximately 10^{-6} torr. To maintain low humidity at atmospheric pressure, we backfilled the chamber with dry nitrogen. Higher humidity levels were set by feeding water vapor into the chamber, and allowing the system to equilibrate for ~ 24 hrs. We measured relative humidity with a capacitive sensor whose dielectric layer equilibrates with the surrounding gas [18]. The sensor has a linear response from 0–90 % humidity, and when calibrated with the local temperature, the accuracy of this measurement is $\sim 2\%$. Since humidity between the plates of the experimental cell can be somewhat different from the surrounding chamber, the sensor must be placed within the apparatus to acquire an accurate measure.

Conducting particles acquire a surface charge when they are in contact with the capacitor plate. As the magnitude of the electric field in the capacitor exceeds the critical value E_1 the resulting (upward) electric force overcomes gravity mg (m is the mass of the particle, g is the acceleration due to gravity) and pushes the charged particles upward. When the grains hit the upper plate, they deposit their charge and fall back. By applying an alternating electric field $E = E_0 \sin(2\pi ft)$, and adjusting its frequency f, one can control the vertical excursion of particles by effectively turning them back before they collide with the upper plate.

The phase diagram for 165 μm copper particles is shown in Fig. 1. At amplitudes of the electric field above a *second* threshold value, $E_2 > E_1$, the granular medium forms a uniform gaslike phase (granular gas). This second field E_2 is 50–70 % larger than E_1 in nearly the whole range of the parameters used. This effect is also seen in the finer 35 μm copper particles (see Ref. [10] and Fig. 2). In the field interval $E_1 < E < E_2$, a phenomenon analogous to coalescence dynamics in systems exhibiting first order phase transitions [19,20] is observed. Upon decreasing the field below E_2, the gas phase loses its stability and small clusters of immobile particles surrounded by the granular gas form. These clusters then evolve via coarsening dynamics: small clusters disappear and large clusters grow. Eventually the clusters assume almost perfect circular form. [A close-up image of one of the clusters is shown in Fig. 4(a).] After a very large time ($t \approx 30\,000$ sec) a single cluster containing about 10^5 grains survives. At the final stage, a dynamic equilibrium between the granular solid and the surrounding gas persists; not all the particles join the last cluster.

We observed a very sharp peak in $E_{1,2}$ for the frequency range from $0 < f < 2$ Hz. In this range, the typical time for cluster formation becomes smaller than the applied field frequency. In this frequency range coarsening is not well defined because the system follows the instantaneous value of the electric field and samples through both phases each cycle. Similar behavior was observed for small (35 μm particles; see Ref. [10]), with the difference that the peak oc-
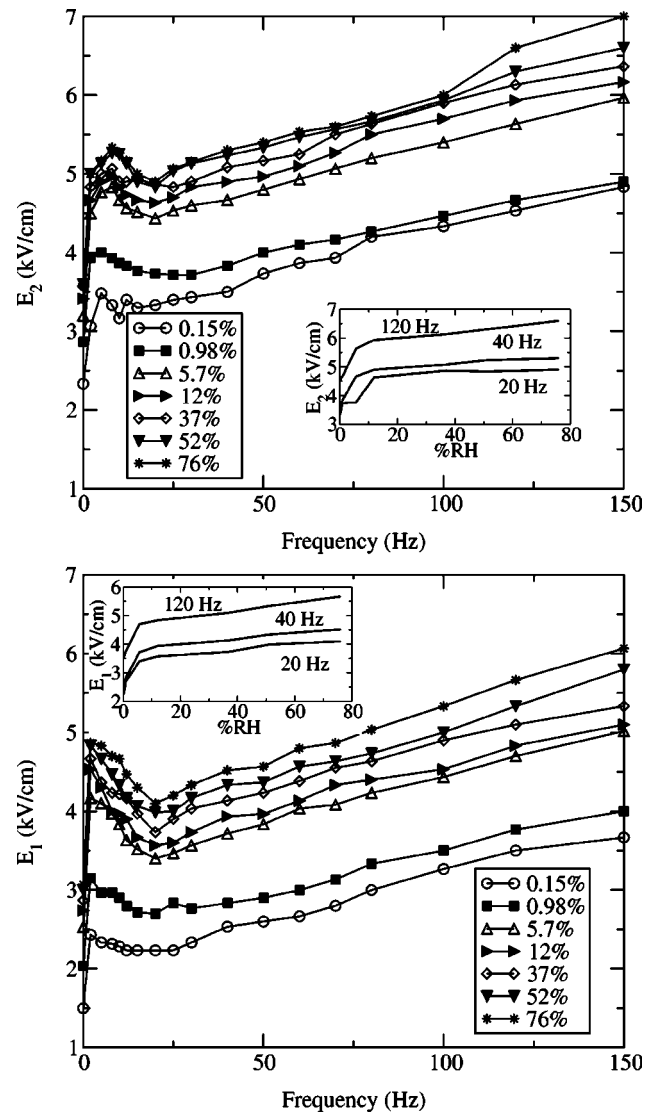

FIG. 2. Top: E_2 vs f plane for several different humidities. Bottom: E_1 vs f plane for several different humidities. Particles are spherical copper with 35 μm diameter. Inset 2: $E_{1,2}$ vs humidity for different values of the frequency f.

curred at $f \sim 12$ Hz. This can be attributed to higher mobility of small particles, which results in a smaller time scale for cluster formation.

The mechanism that allows cluster formation involves screening of the electric field by groups of particles [10]. If spheres are in contact with each other, the surface charge will redistribute: each sphere in the cluster acquires smaller charge than that of an individual sphere due to a screening of the field by its neighbors. Details about the necessary amplitude of the applied electric field and the coarsening dynamics for 35 μm particles are discussed elsewhere [10]. Data taken using 165 μm particles was consistent with these previous calculations.

We are interested in how humidity affects dynamics in the system. Figures 1 and 2 show the $E_{1,2}$ line response for different humidity levels and for different particle diameters. The difference between particles in vacuum and those at at-

mospheric pressure with very low humidities was negligible. As the amount of water vapor increases, the clustering starts for higher values of E_0 independent of the frequency. The same is true for the E_1 line. Our measurements indicate an almost linear increase of both critical fields $E_{1,2}$ for larger particles (165 μm) and a very abrupt increase followed by a linear regime for smaller particles (35 μm); see the insets to Fig. 2.

The most natural reason for this increase is the addition of a cohesive force due to humidity. Water clings to the particles and the plates forming a liquid layer on the plates and around each grain. When these layers interact they create liquid bridges, which increase both the particle-plate and the particle-particle cohesion. This extra force prevents particles from moving at lower fields. Since smaller particles have larger surface-volume ratios, one expects the effects of surfactants to be more pronounced with small particles.

Dielectric particles are by definition nonconducting, and therefore should not be affected by uniform electric fields. However, materials that we consider to be electric insulators in practice are never perfect, and therefore, have some small conductivity. The resistivity of silica glass, for example, is not infinite but $\sim 10^{18}$ higher than that of copper. The electric force F_e necessary to move a dielectric particle is the same as that to move a conducting particle of the same weight; however, the time it takes for a glass particle to gain enough charge for this force balance is determined by the time constant $\tau \sim RC$, where R and C are the effective resistance and capacitance of the sphere. Following the method used in Ref. [10], the dependence of the electric force between a conducting sphere and a plate is $F_e = c a^2 E^2$, where a is the radius of the sphere and E is the field in the capacitor. The constant $c = \zeta(3) + 1/6 \approx 1.36$ comes from summing of infinite series derived in Ref. [21]. If we take into account the charging process of the sphere, the electric force assumes the form (compare to the charging of a capacitor):

$$F_e = c a^2 E^2 (1 - e^{-t/\tau}). \quad (1)$$

The electric force F_e has to counterbalance the gravitational force $G = 4/3 \pi \rho g a^3$, where ρ is the density of the material. Solving for τ one derives

$$\tau = -t_0 / \log\left(1 - \frac{\tilde{E}^2}{E^2}\right), \quad (2)$$

where t_0 is the time for first jump. Here $\tilde{E} \approx \sqrt{4\pi \rho g a/3c}$ is in fact the first critical field for the conducting sphere. For 85 μm silica glass particles ($\rho = 2.55$ g/cm^3) one has an estimate $\tilde{E} = 2.1$ kV/cm. In general, we expect that the value of $\tilde{E}$ will be slightly larger due to additional cohesion forces. We find t_0 at low humidity and applied field of 6 kV/cm to be $\sim 10^3$ sec. The value of τ for different values of humidity is given in Fig. 3.

Figure 3(b) indicates that the dependence of t_0 versus the humidity level h is in fact exponential. Indeed, all the measurements of t_0 versus E_0 for different values of h collapse on one line by scaling $t_0^* = t_0 / \exp(\lambda h)$ with $\lambda = 19$. This ex-
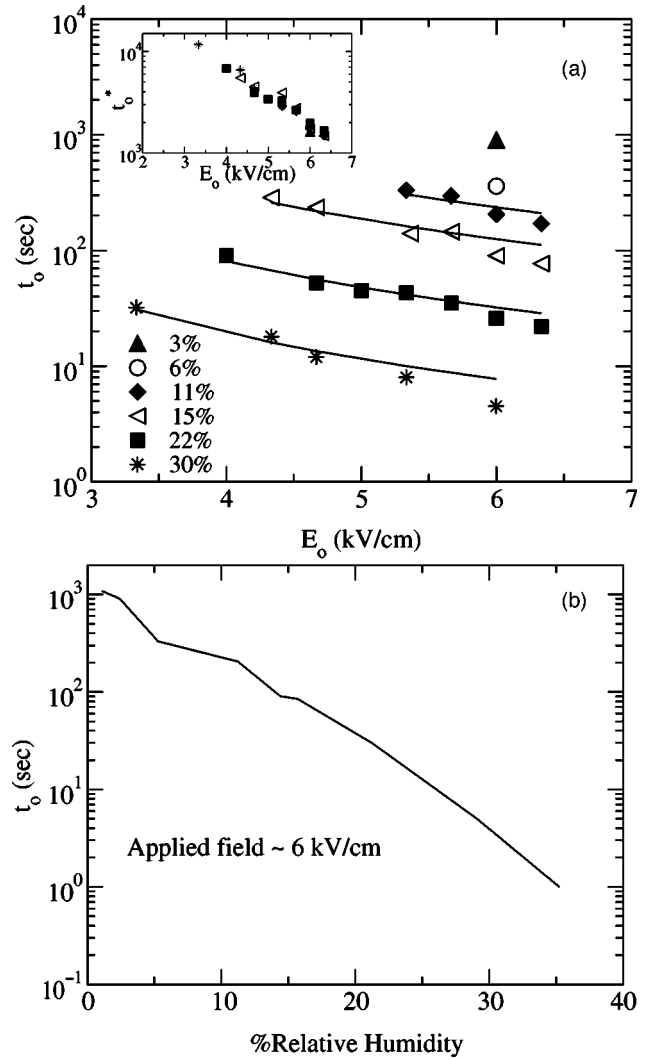

FIG. 3. Time before first movement t_0 for 85 μm glass beads. (a) t_0 vs applied dc electric field Solid lines show fits according to Eq. (2) with $\tilde{E} = 2.2$ kV/cm and $\tau = 52, 215, 838,$ and 1579 for the humidity levels $h = 30\%, 22\%, 15\%,$ and 11% correspondingly. Inset: t_0 vs E_0 scaled by the factor $\exp[\lambda h]$, where $\lambda = 19$. (b) t_0 vs % relative humidity for applied dc electric field of 6 kV/cm.

ponential (as opposed to linear) dependence of the charging time versus humidity is a strong indication of a probabilistic nature of the charging process. It may resemble features of the "asperity" regime of Refs. [11,12] of cohesion between two grains. According to Refs. [11,12] the cohesion process is dominated by random surface roughness of the grains if the thickness of the fluid layer is smaller than the typical roughness scale. Since we are working at extremely small liquid fractions, we expect that the fluid does not cover the bead uniformly. In contrast, only a very small fraction of the grain surface contributes to the charging, and this rare event is described by an exponential distribution.

As in the case of conducting particles, a surface layer of water causes glass particles to become more cohesive, but also allows faster charging of the bead surface. At low humidities, the beads charge slowly, moving with a time near τ for a dc charge. Because of the slow buildup and release of
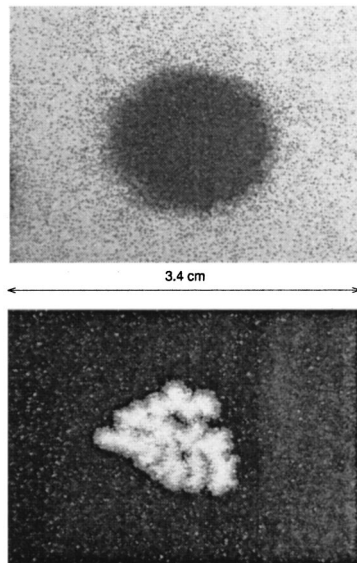
FIG. 4. Comparison of clusters formed from copper and glass 165 μm particles.

charge, subsequent ac driving moves the particles rapidly allowing some discharge every time the plate is touched. This transient ac-driven movement depends on charging time and the driving frequency. As the humidity is increased, a layer of water surrounds the particles, allowing the particles to gain and lose charge more efficiently. Figure 3(a) shows time before the first movement t_0 versus the applied field for different humidity levels. Figure 3(b) shows the decrease in this time for a specific field amplitude (6 kV/cm).

Since the time between jump events in a dielectric at low humidity is of order 10^3 sec, the formation of clusters due to field screening takes a prohibitively long time to observe. As the humidity increases, the time between events decreases,

allowing an observable gas phase, but the subsequent clustering is dominated by a different mechanism than in the conducting case. Added water vapor increases the probability that collisions between particles will result in cohesion. This leads to clusters of a different type than those observed with conducting grains. Figure 4 is a comparison between a typical screening charge-dominated metal cluster and a cohesion-dominated glass cluster, both consisting of 165 μm particles. The metal produces smooth circular shapes (similar to surface tension), while the glass produces a towerlike structure with many observable branches. This branch formation indicates that the ''surface tension'' for the glass particles is small compared to the cohesion forces, and that the intergrain cohesion forces are large compared to the electric force that supports grain motion. We believe that due to large charging time and strong cohesion, the dynamics of dielectric particles is similar to diffusion-limited aggregation [22]. In contrast, fast charge redistribution enhances coarsening of clusters for conducting particles.

In conclusion, we have shown that humidity has a noticeable effect on dynamics of electrostatically driven granular media. In highly conducting materials, such as copper spheres, the dynamics is manifested by coarsening, which is due to screening of the electric field in dense cluster. In this case humidity aids the process by creating cohesive liquid bridges between the particles and with the plate. In dielectric materials, both the dynamics and the coarsening are dominated by humidity effects. A water layer allows the particles to gain and lose charge more efficiently leading to movement, but also creates a cohesive force that creates clusters which behave quite differently from the conducting case.

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