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Spontaneous Air-Driven Separation in Vertically Vibrated Fine Granular Mixtures

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We report the observation of the spontaneous separation of vertically vibrated mixtures of fine bronze and glass spheres of similar diameters. At low frequencies and at sufficient vibrational amplitudes, a sharp boundary forms between a lower region of glass and an upper region of the heavier bronze. The boundary undergoes various oscillations, including periodic tilting motion, but remains extremely sharp. At higher frequencies, the bronze separates as a mid-height layer between upper and lower glass regions, and the oscillations are largely absent. The mechanism responsible for the separation can be traced to the effect of air on the granular motion.

Granular materials occur widely in nature, and the ability to handle grains and powders is central to numerous industrial processes (1, 2). The dynamics of large grains is controlled by the inelastic collisions between grains and with constraining surfaces (3); under vibration the grains may exhibit flow (4), convection (5, 6), arching (7, 8), and the formation of striped, square, or hexagonal patterns (9, 10). The "Brazil nut effect," in which a larger grain moves to the top of a collection of smaller grains (11), is also well known.

Since the time of Faraday, air has been known to strongly influence the motion of fine particulates (12), leading to the spontaneous formation of heaps in vertically vibrated granular layers (12, 13) and to the spontaneous tilting of collections of fine grains vibrated vertically within a box (14). Recently it has been reported that air also has an influence on the Brazil nut effect (15). However, to date, there is no general consensus on the basic mechanism responsible for these air-driven effects (15-18). Similarly, there is now a substantial body of knowledge on segregation and separation in granular composites (19), but a clear understanding of many of the physical processes involved is still lacking (20-24). Furthermore, much of the attention has been focused on large particulate systems where air effects do not play a major role.

We report investigations of the behavior of mixtures of fine particles when subjected to vertical vibration in the presence of air, carried out to explore the interplay between granular separation and the influence of air on the motion. We restrict ourselves to describing the surprising separation behavior exhibited by a fine binary mixture having components with very similar properties save for their densities. The effect represents a novel ordering mechanism in vibrated granular materials in the presence of a surrounding fluid. This phenomenon, being distinct from size segregation, clearly has important implications for the mineral extraction, pharmaceutical, and powder processing industries, where the separation of fine particulates of equal sizes has been notoriously difficult.

Initially we consider a mixture consisting of bronze spheres of density 8900 kg/m3 and of diameters spanning the range 90 to 120 µm, mixed with glass spheres of density 2500 kg/m3 and of the same 90 to 120 µm size range, in the proportion 25%:75% by volume. The dynamic angles of repose are $23.4^{\circ} \pm 0.5^{\circ}$ and $23.9^{\circ} \pm$ 0.5° for the glass and bronze spheres, respectively, and measurements in vacuum indicate that, at low impact velocities, the coefficients of normal restitution are both very close to unity. The mixture, of mean depth 20 mm, is contained in a glass box 50 mm high and of internal dimensions 40 mm by 10 mm in the horizontal plane. The box is attached to an electromechanical transducer that is used to induce vertical sinusoidal motion, with the axis of the trans35. Supported by NASA grant 5-7012, an MIT C. E. Reed award, and a NATO collaborative linkage grant. We thank P. F. A. M. van Putten for providing Fig. 1. M.v.P. thanks R. Weiss and P. L. Fortini for stimulating discussions.

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ducer and the sides of the box aligned to the vertical to within 1°. The resulting motion is monitored by accelerometers attached to the box. The vertical vibration may conveniently be characterized by a parameter $\Gamma = A\omega^2/g$, where *A* is the amplitude of the oscillation, $\omega = 2\pi f$ is the angular frequency, and Γ is the ratio of the maximum acceleration of the box to the acceleration due to gravity, *g*. At higher values of Γ , some static attraction between the glass spheres and the walls of the box occurs. A trace of antistatic agent is added as an aid to photography; this is observed not to affect the separation phenomena that we now report.

We have studied the behavior between $\Gamma = 1$ and $\Gamma = 20$ and over the frequency range from f = 10 Hz to f = 170 Hz. Figure 1 indicates the regions of the f- Γ plane where the various effects are observed. Once Γ slightly exceeds unity, relative motion of the grains occurs. For all frequencies, there is a low- Γ



Fig. 1. Schematic diagram showing the various phenomena found at different conditions of Γ and f. Region A: region of convection and weak separation. Region B: the lower limit indicates appearance of sharp separation boundaries; the upper limit indicates almost complete separation into bronze-rich and glass-rich phases. Regions C and D: regions of oscillation of the upper surface and separation boundaries. Region D: region of simple tilt oscillations. Region E: region of intense throwing of the upper grains. Region F: region of bronze sandwiched between upper and lower glass domains. Region G: high-frequency region where inversion occurs and the bronze layer moves to a lower level.

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region (region A in Fig. 1) where any weak attempts at separation are thwarted by convection. However, as Γ is increased, very sharp separation boundaries appear between bronzerich domains and glass-rich domains at a critical value of Γ , Γ_c , which is a function of frequency. $\Gamma_c(f)$ is shown as the lower limit of region B in Fig. 1.

The separation behavior is qualitatively different in a lower frequency regime, below about 65 Hz, and in a higher frequency regime above 65 Hz. In the lower frequency regime, $\Gamma_{c}(f)$ varies approximately linearly with frequency. Typical separation behavior is shown in Fig. 2. Initially the grains are well mixed. Within a few vibratory cycles, separation into domains with increasingly sharp boundaries occurs, and the bronze domains appear preferentially near the upper surface. Meanwhile, the upper surface heaps and then breaks symmetry by tilting. Soon the system has separated into a lower phase that consists almost entirely of glass spheres, and an upper phase that has a very high proportion of bronze spheres. The separation boundary is now extremely sharp, being only about one grain diameter wide.

At these lower frequencies, neither the form of the upper surface nor the separation boundary is static; rather, they undergo oscillatory motion. This occurs throughout regions C and D in Fig. 1. At frequencies of 10 to 25 Hz, only periodic oscillations are found for values of Γ just exceeding the critical value, the region D. These oscillations consist of periodic swings between the two alternative symmetry-breaking tilts (Fig. 3). The motion takes place entirely within the larger height and width dimensions of the box. The smaller 10-mm depth dimension is not appreciably involved. Above 25 Hz, higher values of Γ are required to obtain swings between the two tilts, and at these values of Γ a range of aperiodic oscillations are also observed, involving both horizontal planes. At yet higher values of Γ , corresponding to region E in Fig. 1, the upper bronze grains are thrown forcefully into the air during the vibratory cycle and the separation phenomenon becomes less effective.

In the higher frequency regime there is also a low- Γ region (region A in Fig. 1) for which convection and only weak separation are observed. Above this lies a critical value $\Gamma_c(f)$ at which sharp separation boundaries first appear. $\Gamma_{c}(f)$ is indicated by the lower limit of region B. In this higher frequency regime, $\Gamma_{\rm c}$ is not strongly dependent on frequency (Fig. 1). By the upper limits of region B, separation into bronze-rich and glass-rich domains is largely complete. However, the bronze-rich phase is not at the top. Rather, it occurs as a horizontal or slightly slanting layer, sandwiched between a lower glass and an upper glass domain. As Γ is increased through region F, the bronze-rich layer moves closer to the upper surface. When Γ reaches region G, the granular system undergoes an abrupt inversion and the bronze moves to form a layer close to the bottom of the box. For region G, Γ is again linearly dependent on frequency.

The separation and inversion behavior is shown in Fig. 4 by a series of "snapshots" of our granular system in the steady state for increasing values of Γ . After separation, the boundary between bronze-rich and glass-rich domains is extremely sharp. It is also noteworthy that, once the bronze is within the body of the glass, the upper surface becomes horizontal. The kinetic energy of the glass grains is then greatly reduced, whereas the bronze grains are very active. The movement of the upper glass grains is very small indeed relative to the throwing observed at comparable amplitudes of vibration in the low-frequency regime.

Observation of the upper glass surface in region G and the upper parts of region F (Fig. 1) reveals patterning, usually of a hexagonal form. After an inversion we often observe small but

very active "puddles" of bronze spheres, trapped on the upper surface of the glass. Within a puddle, the individual bronze grains exhibit violent motion, in great contrast to the relative tranquility of the neighboring glass grains. Nearby puddles have a tendency to merge. However, if a sufficient mass of bronze collects together in such a process, the granular glass surface appears to be unable to support it, and some or all of the bronze mass forms a small globule, which then passes down through the glass to join the main bronze layer.

In both the low-frequency and high-frequency regimes, the separation boundaries appear to form by local diffusion-like processes, followed by coarsening. Once an upper or intermediate bronze-rich layer has become established, convection may be observed within the individual glass-rich and bronze-rich regions, the motion generally being more rapid in the latter. However, no convective flow mixes these regions.

When these experiments are repeated in a



Fig. 2. A time sequence of photographs showing the separation of a homogeneous bronze-glass mixture into separate upper bronze-rich and lower glass-rich phases for f = 40 Hz and $\Gamma = 6.7$. The vibration was stopped and photographs were taken after (**A**) 0, (**B**) 8, (**C**) 13, (**D**) 33, (**E**) 63, and (**F**) 750 sine cycles.



Fig. 3. A time sequence of photographs for 15 Hz and $\Gamma = 2.3$, illustrating a half cycle of simple tilt oscillations. Ten complete sine cycles of vibration were applied between each of the sequential images (**A** to **F**). The configurations of the bronze upper surface and the separation boundary periodically swing together back and forth between the two alternative configurations where most of the surfaces are tilted at an angle close to the dynamical angle of repose.

box with horizontal dimensions of 10 mm by 10 mm, similar strong tendencies for the bronze and glass to separate are again observed, and low- and high-frequency regimes can similarly be identified. However, the simple tilt oscillations are suppressed, and once a bronze layer has formed, it cannot move to a new position by an inversion process after a change of Γ . Rather, it moves by the diffusive passage of glass grains through the bronze layer. This is often a very slow process indeed.

If the air is slowly evacuated from the box, the separation is eventually lost. In the lowfrequency regime, downward convection of the bronze at the front and back faces of the box becomes increasingly evident as the pressure falls. By 10 to 15 mbar at 20 Hz and by 20 to 25 mbar at 40 Hz, rapid global convection thwarts any tendency to separate. At somewhat lower pressures, the surface tilting disappears (15). In the high-frequency regime, the bronze-rich region becomes increasingly diluted with glass and expands as the pressure is lowered. By 60 mbar at 80 Hz, and by 90 mbar at 140 Hz, the bronze-rich region has expanded to occupy the whole volume, which now contains well-mixed grains carried on rapid convection currents. We also note that, in air, the sharpness of the separation boundaries weakens as the grain sizes are increased; at grain diameters of 400 to 500 µm, the tendency to separate has largely disappeared.

Our experimental findings show that there is a strong tendency for fine glass and bronze particles to separate under vibration in the presence of air. The fact that separation is avoided only if the air pressure is substantially reduced, or if the size of the particles is greatly increased, suggests that air effects are important. By treating the granular bed as a porous medium, the local variation in air pressure, P', can be modeled by

$$\phi \, \frac{\partial P'}{\partial t} \, = \, P_0 \nabla \, \cdot \, \left(\phi \, \frac{\kappa}{\mu} \, \nabla P' \right) \, - P_0 \nabla \cdot \, \mathbf{u} \tag{1}$$

where ϕ is local porosity, κ is the permeability of the granular bed to air, μ is air viscosity, **u** is granular velocity, ∇ is the gradient operator, and P_0 is ambient air pressure (25). At high ambient pressures and low vibrational frequencies, the time derivative in Eq. 1 can be neglected. In this limit, the resultant force on the grains acts as viscous damping in the reference frame of the container and is independent of P_0 . An order-of-magnitude analysis of Eq. 1 suggests that viscous damping will dominate for pressures greater than $10^{-6} \omega L^2/a^2$ mbar, where a is the radius of the grains and L is the depth of the bed. We observe good separation only for pressures exceeding this value, hence separation occurs in the damping-dominated regime.

The bronze and glass spheres, being of similar size, will experience the same viscous damping force when moving at the same velocity. However, as bronze is more dense than glass, this force will retard the motion of the glass spheres more strongly. Thus, under comparable conditions, the bronze spheres are more mobile, as we observe.

Under vibration, an initially well-mixed bed will dilate as a result of particle motion. Because the bronze spheres are more kinetically active than the glass spheres, regions that are slightly bronze rich will dilate more than glass-rich regions. Any inhomogeneity in the composition of the mixture will thus result in spatial variations in particle density. Over many cycles of the vibration, there will be a tendency for particles to diffuse from compact to dilated regions down the particle density gradient. However, as bronze is more mobile, this will result in a net transfer of bronze from compact, glass-rich regions to dilated, bronze-rich regions, thus enhancing the effect further. Hence, there is a dynamical instability in the presence of air that causes the bronze and glass to separate. Once separated, it will be extremely difficult for



Fig. 4. A sequence of photographs showing the behavior at 150 Hz for increasing amplitude of vibration. The sequence corresponds to Γ values of (A) 1, (B) 6, (C) 10, (D) 16, (E) 17, and (F) 18. The photographs were taken in the steady state at each value of Γ . Between (E) and (F) the bronze collected into a cylindrical roll, passed to the bottom of the box, and unrolled into the layer shown in (F).

bronze spheres to re-enter the compact glass-rich regions, whereas the occasional glass sphere will be able to enter the more dilated bronze-rich phase, as is also observed.

This qualitative explanation is consistent with our experimental findings, but it cannot explain why, once separated, the bronze-rich region either forms on the top of the sample or is sandwiched between glass-rich layers. Two further effects seem to be in competition. The heavier bronze spheres will be thrown higher than the glass spheres because of the differential effect of air damping, and bronze-rich regions once formed must be supported by collisions from below. At lower frequencies, and consequently greater amplitude of vibration, the former effect appears to dominate. However, at higher frequencies, a more detailed description of the dynamics is required to predict the distribution of particles in the separated state.

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