Size segregation in vibrated granular systems: A reversible process

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The interior of a vibrated bed of mixed-size particles is examined experimentally, revealing segregation patterns that are considerably different than the ''layered cake'' structures published in previous literature. The frequency of vibration has a strong effect on such patterns, which are destroyed when the vibration frequency is increased past ≈ 20 Hz. The segregation process is reversible; the granular system can be driven back and forth between segregated and homogeneous states by decreasing or increasing the vibration frequency. [S1063-651X(97)13006-9]

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A thorough understanding of granular flows is highly desirable. Naturally occurring systems such as soil, sand, and gravel have flow properties that are the result of interactions of large numbers of macroscopic particles. The quality of many industrial products (pharmaceuticals, food, cement, glass, polymers, etc.) is controlled by the flow properties and mixing behavior of granular materials. This paper deals with a type of granular systems, particle beds subject to vertical vibration, that have recently attracted considerable interest [1–7]. Several studies have shown that macroscopic convective motion develops in such systems [1–5]. The most commonly observed flow patterns exhibit circulation rolls in which upward motion takes place at the center of the container, and downward motion occurs near the walls, although the direction of the convection can also be reversed by modifying the frictional characteristics of the container walls. In many instances, formation of a heap at the free surface of the granular material is also observed. Such phenomena have been reproduced by computer simulations that represented the granular system as a collection of discrete particles with friction and elasticity [4] and also as a Newtonian continuum with negative slip at the boundaries [5].

Another effect is observed in systems with particles of mixed sizes. As vibration is applied, particles of different sizes segregate into separate regions, increasing the heterogeneity (and decreasing the entropy) of the system (size segregation occurs also in sheared, fluidized, and gravity-driven flows [8]). For vibration-driven segregation, it has often been reported that large particles tend to rise to the top of the granular system (regardless of their density) [9]. Recent studies have used computational models to explain this phenomenon. Rosato *et al.* [6] performed two-dimensional Monte Carlo simulations to determine the trajectory of large and small particles within a bed as they collectively fell from a certain height. After several raising-falling cycles, large particles migrated to the top of the container and remained there for all particle size ratios $\Phi \geqslant 1.5$ (where Φ is defined as the diameter of large particles divided by that of the small particles). In these simulations, large particles moved upwards obeying a local geometric mechanism. As particles dropped, small particles fell into gaps under large particles. A large particle could move down only if several small particles simultaneously moved out from under the large particle; such an event had a low probability and did not occur very often. Hence, this mechanism resulted in a net upward motion of the large particles. In a related simulation by Jullien, Meakins, and Parlovitch [7], particles fell one at a time until reaching a local minimum. Once again, the final result showed the large particles populating a horizontal layer on top of the small particles. Both computations used periodic boundary conditions in the horizontal direction (neglecting frictional interactions between particles and container walls), and, hence, were devoid of convective flow.

Knight, Jaeger, and Nagel [2] recently examined experimentally the motion of a single large particle in a bed of small particles with a large upward convection zone at the center of a cylindrical container and a narrow downward convective zone near the wall of the container. The large particle rose with a speed equal to the velocity of the upward convective zone. Since its diameter was larger than the width of the downward convection zone, once the particle reached the top of the bed, it stayed there. Knight, Jaeger, and Nagel suggested that this mechanisms was responsible for segregation in vibrated granular materials, generating a long-time segregation structure displaying large particles on top of small particles.

However, as shown below, other patterns are also possible. Two previously unreported experimental observations are communicated in this letter: (i) The *interior* of a vibrated bed of mixed-size particles is revealed to show that segregation patterns can be considerably different than the ''layered cake'' structures described in previous studies [2,6,7,9]. (ii) It is also shown that the frequency of vibration

TABLE I. Properties of particles used in the experiments reported in this paper.

Nominal size (μm)	dp, 10% (μm)	dp, 50% (μm)	dp, 90% (μm)	density (g/cm^3)
66	30	66	103	2.46–2.49
180	116	180	280	2.46–2.49
600	490	600	700	2.55
1500	1400	1500	1600	2.95
5000	4500	5000	5500	2.55

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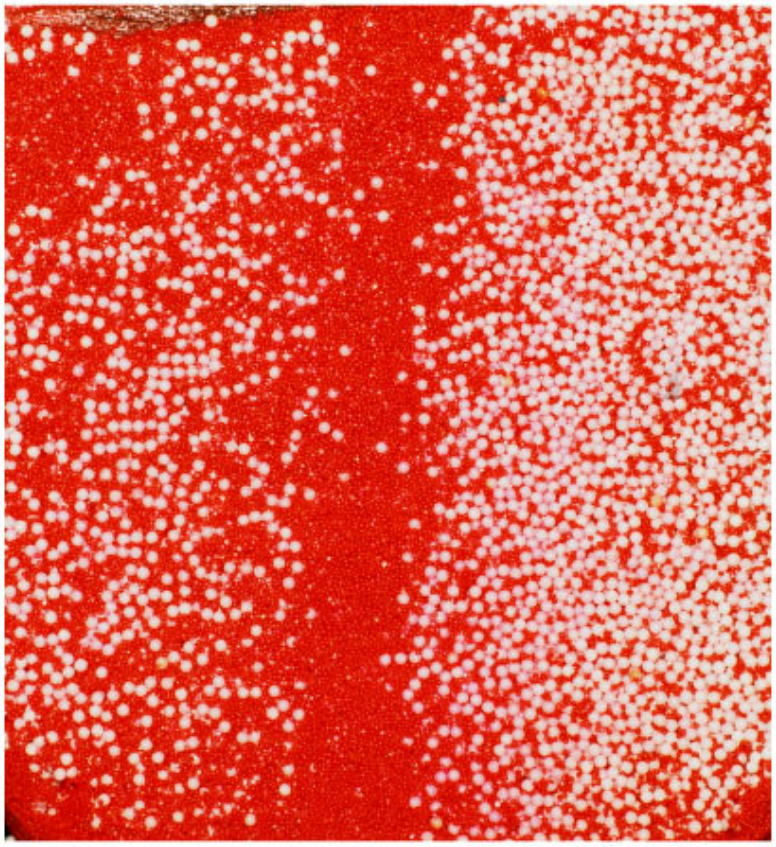

FIG. 1. (Color) A mixture of 1500 μm (white) and 600 μm (red) glass beads in a 102 mm diam by 114 mm high bed, vibrated at 14 Hz for 20 min.

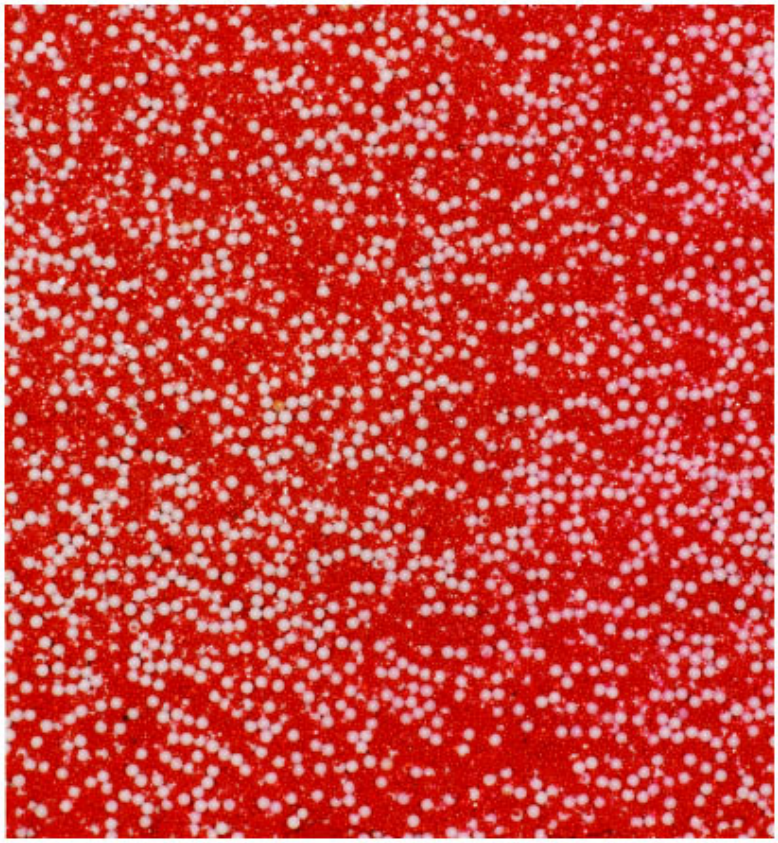

FIG. 2. (Color) A mixture of 1500 μm (white) and 600 μm (red) glass beads in a 102 mm diam by 114 mm high bed, vibrated at 22 Hz for 20 min.

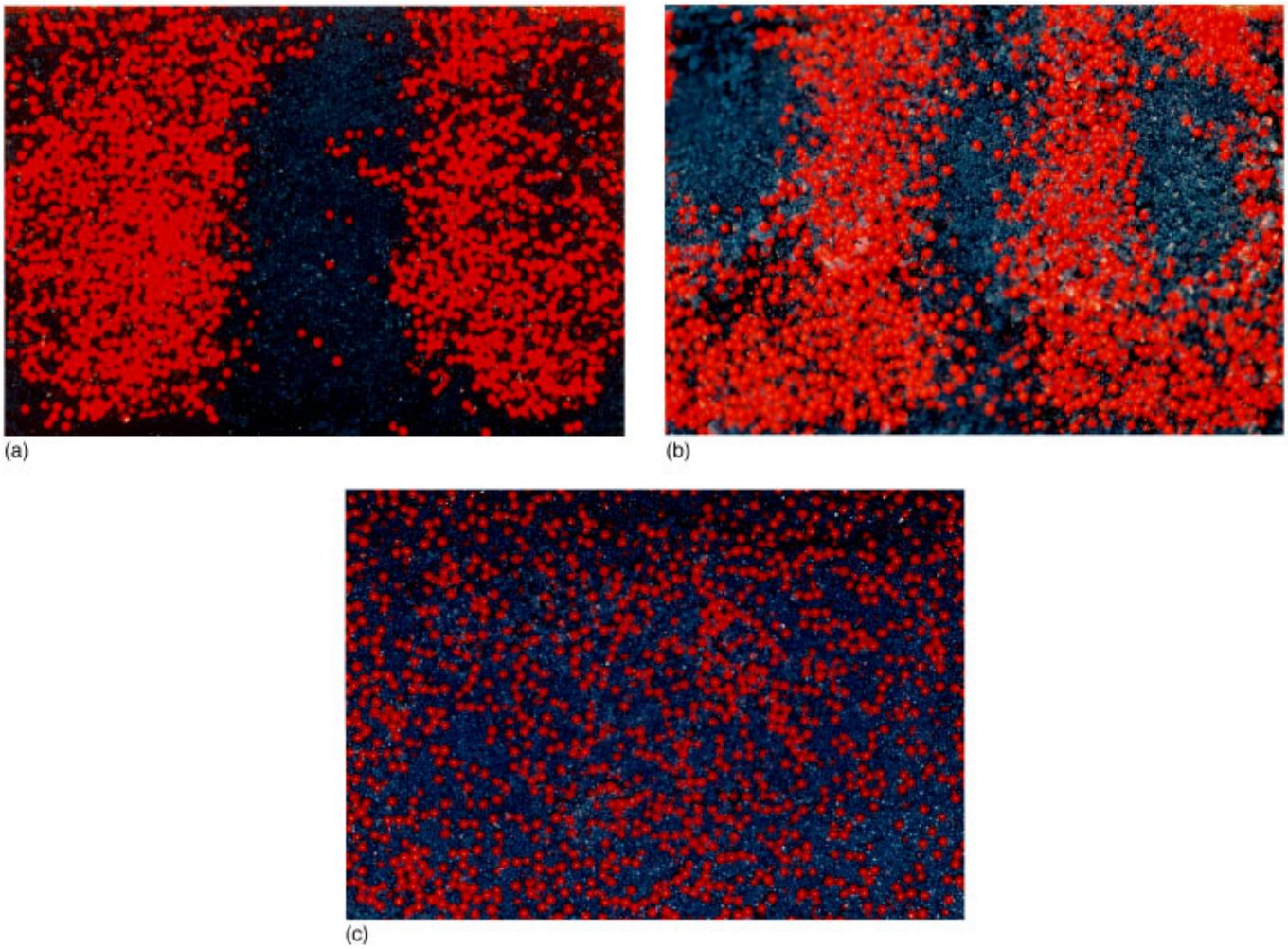

FIG. 3. (Color) Evolution of an initially layered system of 1500 μm (red) over 600 μm (blue) glass beads in a 102 mm diam by 75 mm high bed, vibrated at 22 Hz for (a) 5 min, (b) 10 min, and (c) 20 min.

has a strong effect on segregation patterns. Segregated systems become well mixed if the vibration frequency is increased past a certain threshold. Moreover, the process is reversible; the granular system can be repeatedly driven between segregated and homogeneous states by modifying the vibration frequency.

In the experiments reported in this paper, granular beds consisting of glass beads of two different sizes were vertically vibrated in cylindrical containers. Particles used in such experiments include 5000 μm, 1500 μm, and 600 μm beads from Jaygo Inc. (Union, NJ); and 200 μm and 60 μm beads from Potters Industries Inc. (Parsippany, NJ). Properties of these particles are summarized in Table I. The beds were driven using a mechanical vibration table (All American Co., Chicago IL). The bed depth was 10–15 cm for 10 cm diameter containers and up to 20 cm in 15 cm diameter containers. The table was operated with a fixed amplitude of oscillation $A = 1.56$ mm, and a frequency range f from 10–50 Hz, corresponding to an acceleration $a = A(2\pi f)^2$ between 6.16 m/s^2 and 153.97 m/s^2.

Experiments begun by placing equal volumes of beads of two different sizes in two horizontal layers of equal depths. The container was then vibrated at a given frequency for a specified amount of time. Subsequently, the resulting mixtures were ''frozen'' by infiltrating them with a binder. Gelatin was used as a binder for systems where the beads where 600 μm or larger, and a commercially available mixture of SD alcohol 40, water, octylacrylamide, acrylates and butylaminomethacrylate copolymer (Rave, Chesebrough Ponds, USA Co., Greenwich, CT) was used for all other systems. Repeated experiments demonstrated that the infiltration process did not disturb the interior structure of the bed [11]. Solidified structures were subsequently sliced using a saw to reveal the internal structure of the granular mixtures. All results displayed in this paper (photographs as well as measurements) correspond to vertical slices (i.e., parallel to the axis of symmetry) through the center of the mixture.

The first experiment described here was prepared by placing a 5 cm layer of 600 μm beads on top of a 5 cm layer of 1500 μm beads. The system was then vibrated for 20 min at 14 Hz ($a = 12.1$ m/s^2). Under such conditions, vertical vibration of the powder bed led to the formation of a well-defined heap at the center of the bed's upper surface. Convective flow was immediately apparent at the exposed surface, with particles surging vertically through the center of the heap, from where they cascaded down the heap towards the outer walls of the container. A segregation process could be directly observed on the exposed surface of this cascading flow. Once reaching the exposed surface, large beads rapidly cascaded all the way to the bottom of the heap, always en-

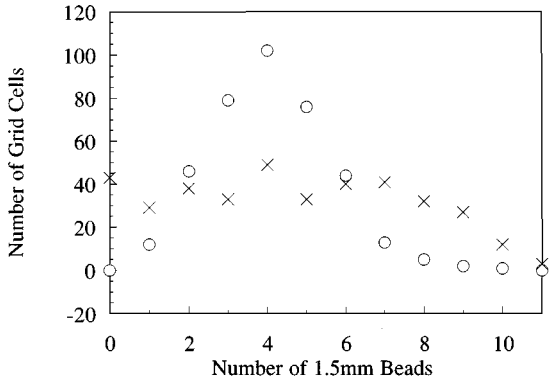

FIG. 4. Composition distributions for 1500 μm–600 μm systems vibrated at 14 Hz ($\times$) and 22 Hz (O) for 20 min.

tering the bed near the lateral walls of the container. Small beads, however, moved much more slowly and entered the particle bed at all locations along the downward convection zone. This process generated a radial segregation pattern that was qualitatively different than the ''layered cake'' structure reported in previous literature. As shown in Fig. 1, small particles concentrated in a central core, surrounded by an annulus of large particles that extended all the way to the side walls. Similar radial segregation patterns, accompanied by the formation of a coherent heap at the center of the bed's upper surface, were observed in numerous experiments at frequencies between 14 and 20 Hz. Repeated experiments for long vibration times showed that these patterns were not a transitional condition but rather a fully developed state.

Completely different behavior was observed at slightly higher vibration frequencies. Figure 2 shows a system of 600 μm over 1500 μm beads after it was vibrated at 22 Hz for 20 min. Convective flow was apparent by direct observation. However, no heap developed under these conditions; the surface of the bed remained mostly flat, displaying occasional surface waves. As a result, large particles no longer cascaded to the side walls of the container. They entered the downward convection zone at all locations. As shown in Fig. 2, under such conditions the process yielded a homogenous mixture. This result was independent of the initial condition, as is shown in Fig. 3 for a system in which the large beads were initially placed on top. The figure shows the time evolution of the structure as the system is vibrated at 22 Hz. Convection rolls are revealed for short times [Figs. 3(a) and 3(b)]. After 20 min [Fig. 3(c)], a homogeneous mixture is obtained once again. It seems clear from these experiments that segregation in vibrated beds is intimately linked to heap formation. Material segregates as it cascades down the heap. If the heap is stable and motion is coherent, well-defined segregation patterns occur. At high frequencies, the heap is unstable (or nonexistent), no coherent segregation process takes place, and the end result is a homogeneous system.

The differences in homogeneity between Figs. 1 and 2 were quantified by enlarging the photographs to 22.5 cm$\times$25 cm, placing a grid over the photographs, and counting the number of large beads within each grid cell. The resulting distributions are shown in Fig. 4. The 14 Hz mixture shows a wide distribution (x) with a relative standard deviation (σ/μ) of 30%. A much narrower distribution (o), with a relative standard deviation of 7.6%, is obtained

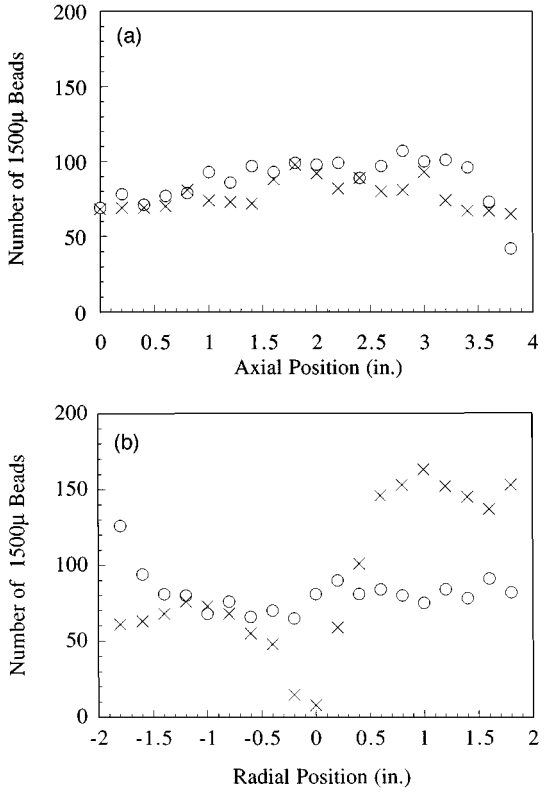

FIG. 5. Composition profiles for 1500 μm–600 μm systems vibrated at 14 Hz ($\times$) and 22 Hz (O) for 20 min as a function of (a) vertical position, measured in 0.5 cm wide bands from the bottom edge of the container, and (b) radial position, measured in 0.5 cm wide bands from the center of the container.

for the considerably more homogeneous 22 Hz mixture. Segregation was examined by computing composition profiles in the vertical and horizontal directions. As shown in Fig. 5(a), the vertical composition profiles for the segregated (14 Hz) and well mixed (22 Hz) systems differ only slightly. However, as shown in Fig. 5(b), large quantitative differences between both systems are revealed by the horizontal composition profiles, indicating that radial segregation (and not ''layered cake'' segregation) is the dominant feature in the system.

The fact that long-time segregation patterns depend on frequency, but not on the initial distribution of material in space, means that vibration-driven segregation is a reversible phenomenon. An initially layered 1500 μm–600 μm system vibrated at 22 Hz becomes homogeneous in about 20 min (Fig. 2). If the system is subsequently vibrated at 14 Hz for 20 additional minutes, a heap forms, cascading motion ensues atop the particle bed, and well-defined radial segregation patterns are developed. If the system is then vibrated at 22 Hz for an additional 20 min, the heap is destroyed, and the combination of vibration and convection render the system homogeneous once again. In some experiments, the system was repeatedly driven back and forth between segregated and homogeneous states by increasing and decreasing the frequency of vibration. All of these observations were confirmed for smaller particles with nearly the same Φ (600 μm–200 μm and 200 μm–60 μm). Heap formation and vertical segregation patterns occurred at low frequencies ($\omega \leqslant 19$ Hz), and homogeneous mixtures were obtained at

higher frequencies ($\omega \geqslant 22$ Hz). Segregation patterns could be repeatedly destroyed and recreated by manipulating the vibration frequency. The only quantitative difference was that for small particles the segregation patterns took longer to develop. This reversibility is potentially important because in principle it could be used both to mix and to separate particles in a controlled manner.

Knight, Jaeger, and Nagel [2] found that for $\Phi \geqslant 3$, the single large bead considered in their experiments remained at the top of the bed. In our experiments, both for $\Phi \approx 3$ (200 μm–60 μm) and for $\Phi \approx 8.3$ (5000 μm–600 μm), large particles consistently displayed radial segregation patterns and traveled along the full convection cycle, regardless of whether there were one or several large particles in the system. We observed ''layered cake'' segregation only for particle beds with $\Phi \approx 83$ (5000 μm–60 μm) vibrated at 24 Hz in 3 in. diameter acrylic containers. In experiments using a single large bead, the bead rose to the top of the particle bed, migrated to the edge of the can, and stayed there. However, in experiments using equal amounts of small and large beads vibrated at 24 Hz, while only large particles reached the top of the bed, such particles also followed convection loops that sampled much of the system's volume. An explanation for the difference between our results and those reported in the literature [2] is that in previous studies the particle bed was not truly vibrated but rather it was exposed to a sequence of discrete ''tapping'' events during which the container was lifted a given distance and then lowered with $a > g$; the particle bed was allowed to relax between events. Tapping generates a narrow downward convection zone that is less than three small particle diameters in thickness. Continuous vibration generates a much wider downward convection region, as revealed both by our experiments and those of Ratkai [1], who also used continuous vibration between 10 and 80 Hz. Differences with computational studies [6,7] can be attributed to the fact that such studies neglected convection caused by particle-wall interactions.

In summary, vibration-driven segregation can generate both ''layered cake'' and radial segregation patterns. When vibration is continuous, segregation is reversible, which could have important implications for both mixing and separation processes. Admittedly, the results presented here leave many open questions. A more extensive survey of segregation mechanisms and patterns is needed in order to develop a thorough understanding of the segregation process. While it is clear that vibration frequency is an important parameter affecting many aspects of the problem, it is currently unclear whether the transition between the segregated and the well-mixed states is gradual or sharp. If a sharp frequency threshold exists between these states, it would be important to characterize the dependence of the threshold on frequency, amplitude, and particle size ratio. If the transition is gradual, it would be desirable to know the extent of homogeneity that is achievable as a function of such parameters. Since heap formation is clearly an important factor in the development of segregation patterns, the connection between heap formation, convective flow, and segregation also needs to be studied in more detail.

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