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Effect of interparticle force on mixing and segregation of dry granular materials

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In this paper, the effects of interparticle force on mixing, segregation, and stratification in dry granular materials are investigated. Avalanche segregation, stratification and also segregation in rotating drums are examined. A series of binary mixtures of granular materials is prepared which consists of spherical iron particles and a nonmagnetic material. By placing each mixture in a magnetic field, the induced magnetic interparticle force could be altered and the effects on particle segregation observed. Using this technique, the effects of altering interparticle force on both avalanche and radial segregation are examined. It is found that altering interparticle force could induce mixed materials to segregate and also induce segregating granular materials to mix. We also report a complete reversal of segregation and stratification as interparticle force was increased. These results have important implications for the mixing of cohesive powders.

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I. INTRODUCTION

Binary mixtures of particles with different physical characteristics such as size or shape are known to segregate when allowed to flow [1]. The mixing and segregation of granular materials are of interest both from the viewpoint of basic research and for their importance to industrial processes. An important question is how the interparticle force F_{ip} affects the mixing and segregation of powders and other granular materials where cohesive forces are present. This has implications in, for example, the pharmaceutical industry, where the mixing of powders often needs to be well controlled.

Recent experiments have shown that altering the cohesion can alter mixing and segregation in wet granular media [2]. Here we generalize this result to include dry granular materials. Studying the effects of altering F_{ip} in these systems can be problematic since it is difficult to alter these forces independently of other factors. Here a technique recently developed by the authors [3,4] for studying the effect of altering F_{ip} in granular materials circumvents these difficulties.

A. Avalanche segregation

A Hele-Shaw cell consists of two vertical transparent plates held a small distance apart by spacers at the edges and sealed at the bottom and sides. When a binary mixture of granular material is poured into the space between the plates, material begins to pile. It then undergoes segregation as material avalanches down the free surface. In the absence of significant interparticle forces, binary mixtures of large and small grains of otherwise identical material will segregate. Under these circumstances, small grains will segregate to the top and the larger grains to the bottom.

B. Avalanche stratification

If, in addition to the size difference, the materials have differing angles of repose, then the mixture can undergo stratification [5]. The material will take on a layered appearance as material spilling down segregates, forming bands of material roughly parallel to the free surface. Makse *et al.* [6] performed experiments on a mixture of sand and glass beads and found that the grains formed alternating layers of sand and glass. This behavior has implications for flows of granular materials in silos and hoppers [7] as well as for understanding several geological processes [8,9].

It has been suggested that the process of segregation is due to the ability of the larger grains to travel faster than the smaller grains across the avalanching surface [10]. Since they have lower inertia, the smaller grains will be more susceptible to being stopped by small bumps and gaps in the underlying grain profile. Stratification, however, appears to rely upon differences in the angles of repose of the two materials in the mixture. If the larger particles have the smaller angle of repose, they segregate out at the bottom of the slope and form a kink in the grain profile. As the avalanching grains hit the kink, they segregate and add more material to it. As material is added, the kink propagates up the grain profile, forming a stripe of segregated material as it grows. Once the kink reaches the top of the pile, material spills down to the bottom of the profile again and the process repeats.

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C. Radial segregation

Another example of segregation occurs when binary granular mixtures are placed in a cylindrical drum and rotated [11]. For complete mixing the drum must be less than half full. Consider, for example, a drum filled to under half full with a binary mixture of different sized but otherwise identical grains. As the drum rotates the smaller grains would be found to segregate to the center of the drum. Recent computer models by Shinbrot *et al.* [12] have shown that binary mixtures of granular materials can undergo chaotic mixing. In these simulations altering interparticle forces was found to alter the mixing behavior from smooth to chaotic. This chaotic mixing is associated with the growth of a "fractal" interface between originally segregated particles. These findings have been confirmed by subsequent experiments by Nasuno *et al.* [13].

II. INTERPARTICLE FORCES

For many granular systems, such as powders, segregation is complicated by the appearance of interparticle forces. There are several methods currently used by researchers to increase F_{ip} and study its effect on granular behavior. Several groups [14–16] have added liquids of varying surface tensions and viscosities to dry powders to observe the increase in F_{ip}. The limitation of this method is that the addition of more and more liquid increases F_{ip} in a stepwise manner. It is difficult to remove liquid in a controllable way and hence hysteretic effects cannot be examined. Altering the interstitial fluid may have other effects in addition to altering F_{ip}. It is known that segregation in other contexts can be heavily influenced by interparticle forces [17]. Adding a liquid to dry powders not only increases the interparticle interaction, but also the interaction between the powder and wall, further complicating the analysis.

To avoid these complications, the behavior of iron spheres within a magnetic field was examined. Varying the strength of the field allowed the resulting interparticle magnetic force to be continuously varied. As the walls of the vessel are nonmagnetic Perspex, the particle-wall interaction is unchanged. If it is assumed that the particles are magnetically linear, it follows that the interparticle force between two particles, F_{ip}, will vary in proportion to the square of the applied magnetic field ($F_{ip} \propto B^2$). The ratio of interparticle force to buoyant weight, F_{ip}/mg, was measured directly, as described below.

III. EXPERIMENTAL APPARATUS

A pair of Helmholtz coils supplied the magnetic field for this experiment. These coils have an inner diameter of 456 mm, an outer diameter of 568 mm, and a thickness of 76 mm. The former are constructed from nylon and were each wound with 1.1 km of 2.24-mm-diam insulated copper wire. The coils are powered by a 16-A, 16-V stabilized dc power supply, and have a total resistance of 2.2 Ω. The magnetic field axis was in the vertical direction. The magnetic field was measured as a function of current by a Bell 2400 series Gauss meter. The field was found to vary less than 10% throughout the volume between the coils and by less than 5% within a sphere of diameter 140 mm centered on the axis of symmetry, midway between the coils.

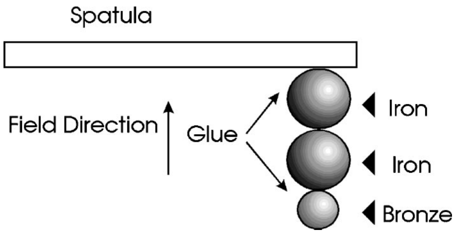

FIG. 1. Method for determining interparticle force to bouyant weight ratio.

To directly measure F_{ip} between two particles as a function of field strength, the following method was used. An iron sphere of the appropriate size was glued to a plastic spatula and positioned on the axis of symmetry at the point midway between the coils. With the field at full strength (approximately 6400 A/m) a second particle of the same size was brought into contact with the fixed sphere. Because of the strong magnetic force between the two particles, the second particle became suspended from the first. The current in the coils—and hence B—was slowly decreased until the suspended particle fell from the fixed particle. At this point, F_{ip} was equal to (or marginally less than) the weight of the particle. This measurement and all subsequent measurements were repeated several times for accuracy. The weight of the particle was measured with a sensitive laboratory balance. To obtain further points for the graph, small (0.5 mm) bronze spheres were carefully glued to the "bottom" of the particle to be suspended (see Fig. 1). This served to increase the weight of the particle without significantly altering F_{ip}. This new "particle" was weighed and the measurements repeated, making sure that none of the bronze particles became dislodged when the suspended particle fell. The resulting calibration was expressed in terms of a multiple of particle weight.

Binary mixtures were prepared using iron particles and a variety of materials with varying static angles of repose and particle diameters. In all experiments the binary mixtures were made of 50%, by volume, iron particles. To investigate both avalanche segregation and stratification for each mixture, a narrow rectangular box of width 4 mm, length 300 mm, and height 150 mm was filled with the binary mixture from a funnel with a stopcock attached. The stopcock was used to control the flow rate. To investigate radial segregation, a narrow Perspex drum of diameter 150 mm and thickness 10 mm was placed horizontally on a set of aluminum roller supports. These were driven by an electric motor. Due to the presence of the magnetic field, only nonmagnetic materials were used in the construction of the roller supports which were connected to the electric motor via a 1-m-long drive belt. The drum was just under half-filled with the binary mixtures.

A. Avalanche segregation

Experiments were then carried out on segregation for increasing values of F_{ip} for each binary mixture. In these ex-

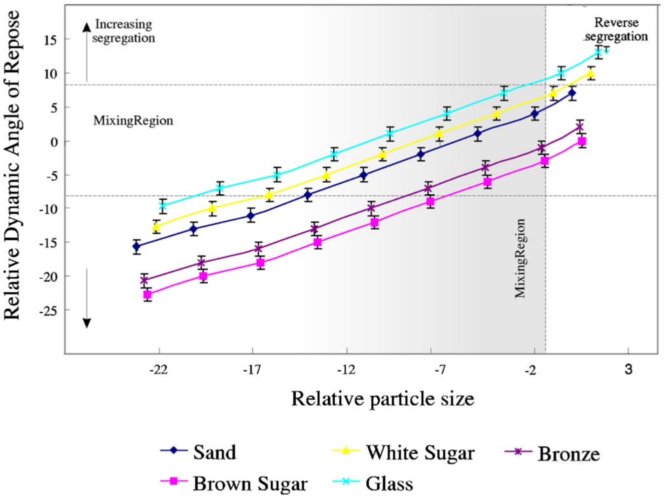

FIG. 2. Phase diagram for mixing and segregation in rotating drums. Relative angle of repose is obtained by subtracting the angle of repose of the nonmagnetic species from that of the effective angle of repose of the iron particles in the field.

periments it was noted that the strength of segregation is unaffected by flow rate until a critical flow rate f_c is reached at which point stratification abruptly disappears. Above f_c the flow is too fast for kinks in the grain profile to form and propagate up the slope, and so continuous flow across the free surface is observed. In order to investigate the effect of F_{ip} on segregation, experiments were carried out with flow rates above f_c.

B. Avalanche stratification

A second series of experiments was carried out to investigate the stratification process. Experiments were again carried out for increasing values of F_{ip} for each binary mixture. However, the flow rate in these experiments was kept constant and below .

C. Radial segregation

Similar segregation experiments were also carried out in which the binary mixture was placed in the narrow Perspex drum. The drum was rotated at approximately 20 revolutions/min. Experiments were conducted for different rotation speeds; however, the segregation behavior was not observed to depend upon rotation rate. The results of these experiments are summarized in Fig. 2.

IV. RESULTS AND DISCUSSION

A. Avalanche segregation

Segregation was observed to occur in mixtures of sand and iron particles. As the magnetic field was increased, the segregation effect was eliminated and then reversed. The experiments were repeated with other materials and similar results were found. The results of these experiments are summarized in the phase diagram (Fig. 3). The effect of applying a magnetic field to the mixture was to reverse the segregation. At low fields, the smaller iron shot was found mostly at the top of the slope. For high fields, however, the iron was found to segregate to the bottom of the slope. For very high fields, the avalanche flow is slowed sufficiently for some stratification to occur.

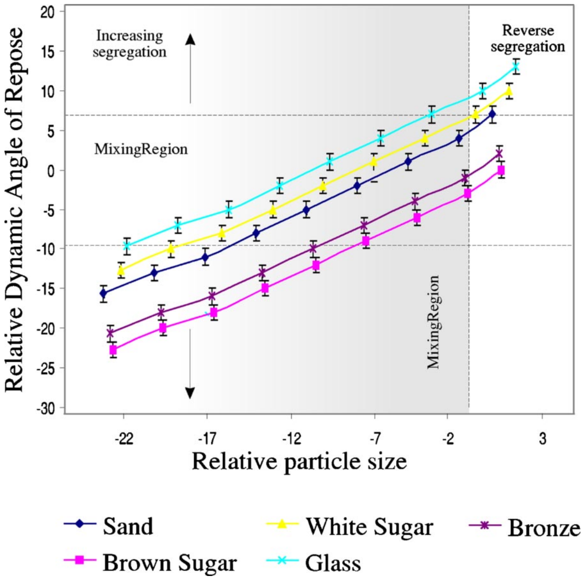

FIG. 3. Phase diagrams for avalanche segregation and stratification. Relative angle of repose is obtained by subtracting the angle of repose of the nonmagnetic species from that of the effective angle of repose of the iron particles in the field.

B. Avalanche stratification

Without an applied field, the sand has a larger angle of repose ($\theta = 41°$) than the iron particles ($\theta = 32°$) and so stratification occurs spontaneously during avalanching. As the field was increased the width of the stratified layers decreased until the material appeared well mixed. As the field increased further the width of the layers increased again (Fig. 4). However, at low field the layers consisted of sand over

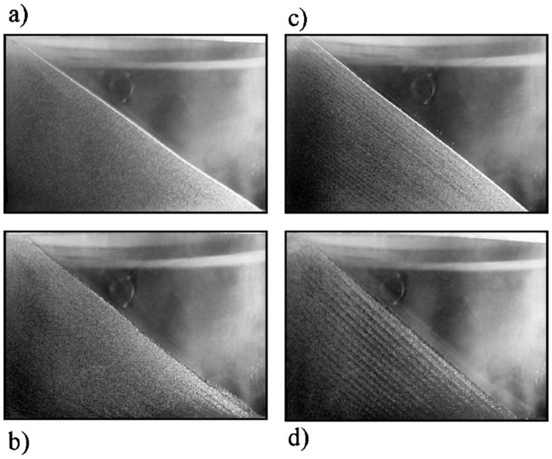

FIG. 4. Reversal of segregation with increasing F_{ip}/mg: (a) low field, (b) high field. Reversal of stratification with increasing F_{ip}/mg: (c) low field, (d) high field.

iron shot and at high field the iron layers formed above the sand layers. The direction of stratification was therefore reversed by increasing F_{ip}. The experiments were repeated for a range of different applied fields and materials. The results are summarized in the phase diagram (see Fig. 3).

Only the sand and iron shot mixture was observed to undergo a full transition between the stratified, mixed, and reversed stratified phases. It was noted that at high field the granular profile became more irregular as long chains of particles formed. This irregularity translated into irregularities within the layers of grains. As material avalanched down the slope, at high field, the flow became more intermittent. Groups of particles will often stick together impeding the flow of material from above. Eventually enough material accumulates above the jammed material for it to flow over the top. The result is that at high field the stripes become increasingly irregular.

C. Radial segregation

Without an applied field, radial segregation was observed in a mixture of sand and iron particles as the drum was rotated. The iron particles segregated to the center of the drum. As the applied field was increased the slip stick behavior was observed to occur at the surface of the material. Initially segregated layers were observed to begin mixing. Our results show that as F_{ip} increased, this mixing reaches a maximum and then decreases as segregation reverses. As F_{ip} increased, the iron particles were observed to begin to stick to one another. Simulations carried out by Shinbrot and Muzzio [1], and experiments carried out by Nasuno *et al.* [13] have shown that increasing F_{ip} can increase mixing by increasing slip stick shearing in the free surface. Slip-stick surface avalanches were observed in these experiments for low rotation velocities. The results of the segregation experiments suggest that as F_{ip}/mg is increased the particles act as if they have a much larger effective particle size.

D. Phase diagrams

In order to investigate the effect of F_{ip} on segregation, an effective particle size can be defined as follows. Imagine the iron particle is suspended in the field. Imagine also that for a given F_{ip} a second particle of equal size and mass is suspended from the first. These two particles can be thought of as constituting a new particle which is twice the length of the original. Similarly by gluing another particle to the first and altering the interparticle force so that they are both just suspended from the first a new "particle" is created which is three times the length of the first and so on. Dividing by the size of the nonmagnetic particle in the binary mixture gives a dimensionless number which can be used to compare binary mixtures. This number, the relative effective particle size, we denote by r_{eff}. Using this definition a phase diagram relating r_{eff} to the angle of repose can be plotted for avalanche segregation and stratification. A similar plot for the dynamic angle of repose and r_{eff} was also obtained. Both the r_{eff} and the angle of repose were found to increase linearly with F_{ip}.

When $F_{ip} < mg$, where m is the mass of the nonmagnetic particle, then particles will have insufficient force to stick together. However, when $F_{ip} > mg$, the effective particle size begins to increase. When r_{eff} is equal to 1 mixing occurs. When r_{eff} becomes much greater than 1 segregation is reversed. The reversal of stratification appears to occur due to changes in the angle of repose of the iron particles relative to the other materials. For low interparticle force, the iron has both a lower angle of repose and low r_{eff} and so segregates to the bottom. As F_{ip} increases, the angle of repose increases. For some binary mixtures the angle of repose increased to the point where it was greater than the other material and stratification was observed to reverse. One might assume that these results hold only for magnetic particles. To test this assumption, experiments were performed using flour and salt. On the basis of particle size alone the relatively large salt grains should strongly segregate and stratify to the top of the very fine grains of flour. Interparticle force strongly alters the segregation and stratification behavior. When lightly stirred mixtures of salt and flour were poured through a funnel and allowed to avalanche they stratified so that the flour was at the top and salt at the bottom of the layers. The flour particles were observed to stick together and clump together into large lumps which acted like larger particles. Vigorous stirring can break these lumps up and produce more homogeneous mixing.

In all experiments the binary mixtures were made of 50% iron by volume. This proportion was chosen to enhance the visibility of the stratification process. It would be interesting to see how stratification changes as the proportion of particles in the mixture is changed. Since the width of the stratified layers depends upon the size of the Hele-Shaw cell, a larger apparatus than the one considered here would allow any such effects to be more easily observed.

It is interesting to note the correspondence between the reversal of segregation and the onset of slip-stick friction in the rotating drum experiments. In models of mixing by Shinbrot and Muzzio, the slip-stick behavior is linked to time variations in the surface profile. Exploring potential connections between slip-stick behavior in radial segregation and avalanche stratification would be an interesting direction for further research.

V. CONCLUSION

A novel means of studying the effect of interparticle force on segregation and mixing has been demonstrated. These experiments show that interparticle forces can cause significant changes in the segregation and stratification behavior of granular mixtures. It was found that increasing F_{ip} in one species of the material increases its ability to mix. For high values of F_{ip} the avalanche segregation, avalanche stratification, and radial segregation were all observed to reverse.

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