Abstract. – Radial and axial segregation is investigated experimentally in polydisperse mixtures of granular materials rotated in a long, partly filled, horizontal cylinder. Radial segregation by size is observed in all polydisperse mixtures. Axial segregation, with smaller size particles forming bands within bands of larger size particles, is observed for mixtures of 3 sizes. In addition, bands in ternary mixtures oscillate axially under some conditions. It is observed that the surface flow speeds depend on the particle size, both in the mixed and segregated case. A simple microscopic model based solely on different frictional properties of three particles yields qualitatively similar results.

Band-in-band segregation of multidisperse granular mixtures

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A striking property of forced granular matter is the tendency of mixtures of different types of materials to unmix (e.g., during avalanching [1] or under vertical vibration [2]). Partially filled cylindrical rotating drums, with horizontally oriented axes, are often used in industry for various processes, such as mixing and grinding. When mixtures of two different particle types are rotated in a drum they tend to segregate in two ways: first, the smaller particles tend to segregate radially forming a radial core near the radial center of the drum; after that, the particles separate into bands along the axis of the drum.

Segregation in a rotating drum was first observed by Oyama in 1939 [3] for an equal volume mixture of glass spheres of two different sizes and has been subsequently studied [4–11, 13] both experimentally and theoretically. Nevertheless, full understanding is lacking, and the assumptions and fundamental equations of proposed theoretical models differ considerably [7–9]. Also, the case of ternary mixture segregation has been briefly discussed [10]. We expand upon these previous works by studying the segregation phenomena for mixtures of three and more particle sizes. We find new patterns and dynamical instabilities relevant to the mechanism of segregation of multiple species granular mixtures in a rotating drum.

Direct imaging of the interior of rotating drum flows was obtained by Hill *et al.* using Magnetic Resonance Imaging (MRI) techniques [11]. They found the radial segregation to persist, even after seemingly complete axial segregation. They also saw subsurface band formation preceding banding visible at the cylinder surface.

In our experiments on mixtures of three to six different particle sizes, we measured the shape of the surface, the surface flow speeds, the dynamical evolution of bands and the radial core, and the steady state banding patterns. We found very rich dynamics, including new dynamical instabilities not seen with binary mixtures. We were able to reproduce most of the observed phenomena with a discrete Monte-Carlo simulation similar to one designed by Yanagita [13].

We rotate the particles in a 10 cm diameter, 67 cm long, transparent perspex cylinder with perspex endplates. For all measurements, the cylinder was half filled with a mixture of glass beads of varying sizes (Jaygo, Inc.). The bead diameters varied for each experiment, but were all between 0.2 mm to 8 mm, and were similarly spherical in shape. We imaged the system with a high resolution color CCD camera from the long side to study axial segregation. In addition, we investigated radial segregation through the transparent endplates.

The richest patterns and dynamical instabilities were obtained from mixtures of three particle types. We used three sizes of colored glass beads, 0.6-0.8 mm (blue), 1-1.25 mm (gold), and 2 mm (red). The drum was rotated at 30 rpm for periods of several hours.

Fig.1 shows radial segregation and the axial band formation process. Similar to binary mixtures, we found radial segregation after only a few rotations with the small particles in the center surrounded by the medium particles, with the large particles on the outside. This radial segregation can be seen at the endwalls as shown in the inset to Fig. 1. Using transparent large and/or medium size beads we were also able to visually confirm that this core extends throughout the entire tube at this point for both small and medium particles. The radially segregated core persisted after axial segregation was observed. Since the drum is half filled, all material flows at some stage during rotation, and the core region extends into the thin surface flow region.

After radial segregation, but before axial segregation, the largest (red) particles predominate on the externally visible cylindrical surface of the tube. After about one minute of rotation axial bands begin to form, starting with bands of the medium sized particles. Within another minute, the smallest particles form bands inside the bands of medium sized particles. Bands within bands like this form across the entire tube until, after 5 minutes, there is a roughly evenly spaced array of bands within bands.

As seen in previous experiments on binary mixtures, the axial pattern coarsens with time (i.e., bands merge to form wider bands), sometimes on very long timescales of hours and days. In some cases, we found this to lead to a final state with a single band within band pattern.

When we mixed four particle types (with 4 mm particles added in equal volume to the mixture) we found them to also segregate both radially and axially. The axial segregation pattern again showed band within band formation starting with the largest particle sizes and going to the smallest. The banding is not as distinct as for ternary mixtures and the order of band formation is also not as easily observable.

Mixtures of five and six different particle sizes also quickly segregate radially, but do not segregate axially for the large range of rotation rates we have investigated (from intermittent avalanching to centrifugally dominated regimes). To determine whether axial segregation is too slow to be observed, we seeded mixtures with an initial band of the smallest particles (one sixth of the total mixture) and observed the evolution of that band. The band gradually disappeared after roughly 200 rotations for a large range of rotation speeds (between 10 and 110 rpm), indicating that axial segregation is suppressed for mixtures of 5 or more particle types in our system.

For mixtures of four or fewer particle sizes, the axial segregation pattern and the sequence in which the bands appear is consistent with describing the axial banding proceeding as shown schematically in a cross-section parallel to the free surface in Fig. 2(a)-(d). After a few

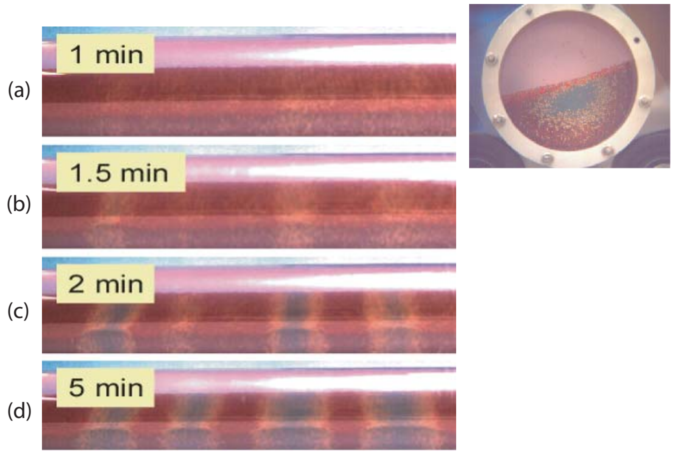

Fig. 1 – Image sequence of ternary mixture of .6 mm (blue), 1 mm (gold), and 2 mm (red) particles. The medium sized particles first form surface bands within the largest particles (1.5 min) and the smallest particles then form bands within the medium particle bands (2 min). These form an array of 'bands within bands' across the length of the tube. On Right: A picture of the radial core through the endwall of the cylinder after 1 minute of rotation. The smallest particles (blue) form a core in the center with the medium (gold) next out and the large farthest out (red).

rotations, radial segregation extends throughout the entire tube (Fig. 2a). An instability of this axially uniform state begins to expand outward (Fig. 2b) until the outermost edge of the core, made up of medium sized particles, reaches the cylindrical wall of the tube (Fig. 2c). As viewed from the outside of the tube, this would be a band of medium sized particles (Fig. 1b). The inner core continues to expand until the smallest particles reach the surface (Fig. 2d) thus forming the band within band structure visible at the surface (Fig. 1c,d). We can see that the qualitative shape of the core is consistent with this schematic as seen through back illumination of a partially tranparent sample (Fig. 1e). (A technique developed by Steve Morris) [12].

For mixtures of three sizes of particles, we visualize this core by mixing opaque small particles with transparent medium and large particles and illuminating from behind. The shape of the cohesive inner core of small particles becomes clearly visible (see Fig. 2e).

When we increased the concentration of small particles in a mixture of three particle types, we observed oscillations in some of the band positions and widths over time. Figure 3 shows a space-time plot of a mixture with excess small particles (40% small particles, 33% medium, and 27% large particles). The inset to Fig. 3 shows that the band widths for large particles (dashed line) is unchanged with time while the width of small particle bands (solid line) oscillates with time. The observed change in width of bands of small particles requires influx and outflux of particles, which is consistent with our observation that an inner core of small and medium sized particles remains within bands of larger particles (Fig. 2). Transport of material between bands of large particles, on the other hand, is inhibited.

In addition to such macroscopic observations, we also studied the flow of individual particles. We took a high speed, closeup image sequence of the banding pattern with small particles in left third, medium sized particles in middle third, and large particles in right third of the field of view (Fig. 4). We then tracked the individual particle positions and extracted downhill

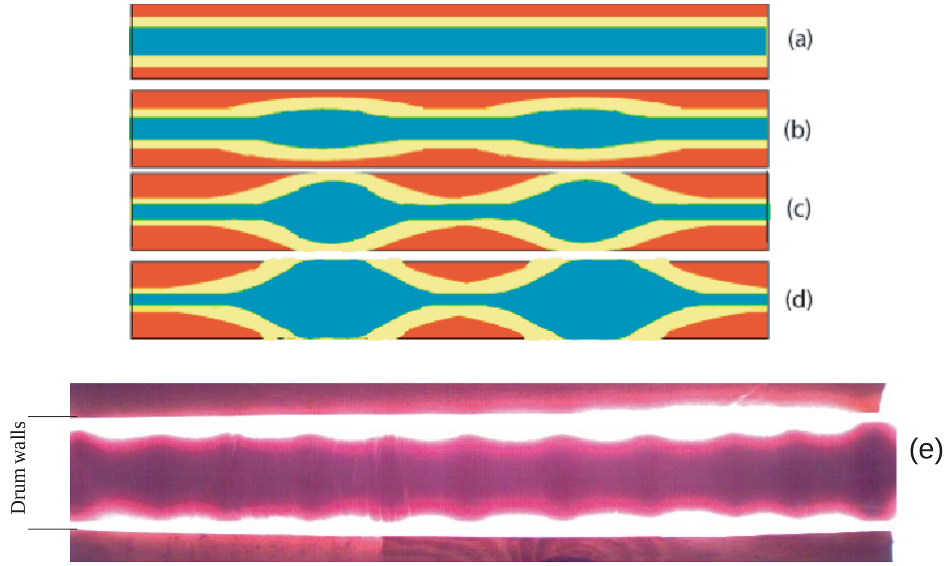

Fig. 2 – (a)-(d) Schematic of the band formation process shown schematically in a cross-section parallel to the free surface. (e) An actual image of the inner core of small particles, the medium and large particles are transparent, and the mixture is illuminated from behind. Similar images can be obtained for medium opaque particles.

flow velocities averaged over time and downhill position at evenly spaced axial slices. We find that small particles travel $\sim 20\%$ faster than large particles. We note, however, that the small particles are likely to have a narrower surface flow layer than the large particles and thus the larger surface flow speed does not neccessarily imply a larger surface flux. We investigated

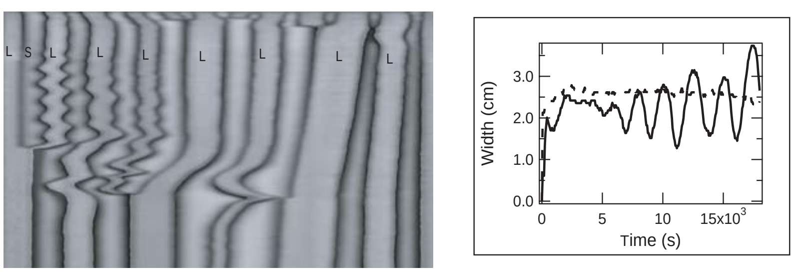

Fig. 3 – On Left: Spacetime plot of oscillations in a ternary mixture with excess small particles (40% small, 33% medium, and 27% large particles). Each line on the space time plot represents a vertical average of one image of a sequence of images taken every 60 seconds. In the image, the Large and Small particle bands are light grey, with the large particle bands marked with an L and alternating with the small particle bands across the image. The medium particles are the dark grey/black bands in between the small and large particles. Note the oscillations for closely spaced bands on the left side of the cylinder. Eventually, bands merge and the oscillations stop. On the Right: The width of one band of small particles (solid line) oscillates, while the width of a large particle band (dashed line) remains constant.

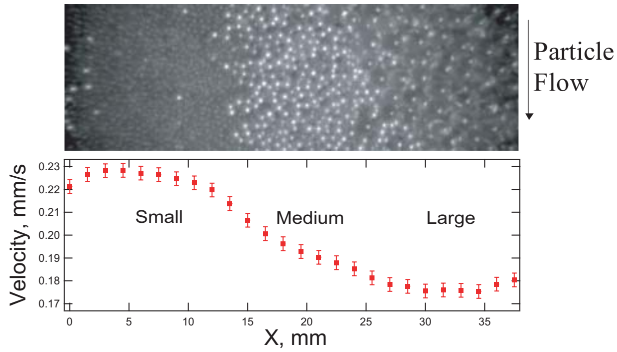

Fig. 4 – On top, one frame of a high resolution close-up image sequence of small (left) medium (center) and large particle (right) bands near the center of the drum. Below, velocities of surface particles averaged over time and downhill position at evenly spaced axial slices, extracted through particle tracking for this sequence. The small particles have a 20% greater flow speed than the largest particles. We used the variance in the data across the small and large particle bands to calculate the error bars.

particle motion immediately after the rotation of a mixture was started, prior to radial and axial segregation. We observe flow speed differences of roughly 3%, above the noise level, with the smaller particles seen overtaking the large particles. Differences in flow speed may thus drive segregation, both axially and radially. This effect will be investigated in more detail in a future publication.

To investigate whether the observed segregation phenomena can be explained solely on the basis of differing surface flow properties of the different types of particles, we use a discrete Monte Carlo simulation to model a rotating drum filled with three species of particles. This microscopic model generalizes a binary mixture model by Yanagita [13] to include three particle types. Unlike the experiment, where the particle types differed in size, the model uses particles with different frictional properties to simulate the differences in surface flows. We employ a three dimensional square lattice with dimensions of 200 lattice sites in the axial direction, 80 lattice sites in the vertical direction, and 10 lattice sites in the horizontal direction (1.6×10^5 total lattice sites). We impose periodic boundary conditions in the axial direction, and load the lattice with a mixture of three particle types (8×10^4 total particles, corresponding to a half-filled cylinder). As mentioned above, the three particle types (A, B, and C) differ in their frictional properties, with A particles having the least friction, B particles having intermediate friction, and C particles having the greatest friction. We use six friction parameters, $F_{ij} = F_{ji}$, to specify the friction strength between an i particle and a j particle. Frictional properties between dissimilar particles, F_{ij}, are chosen to be between F_{ii} and F_{jj}. The specific parameter values used in our simulation were: $F_{AA} = 0.4$, $F_{AB} = 0.44$, $F_{BB} = 1.0$, $F_{AC} = 0.9$, $F_{BC} = 1.5$, and $F_{CC} = 2.4$. Similar behavior is observed for other parameter sets, e.g., within a range of at least plus or minus 12% of the chosen value.

The simulations carry out a large ($> 10^5$) number of discrete time steps. Each time step involves two ingredients to mimic (a) rotation of the cylinder and (b) flow of the top granular layer:

(a) First the particles in the back half of the lattice are all moved upward by one lattice site. Then particles in the lowest layer of the front half of the lattice are moved to the now-vacant lowest layer of the back half of the lattice by reflection of their positions about the back-front midpoint. Finally, the remaining particles in the front half of the lattice are all moved downward by one lattice site.

(b) Next, a particle is chosen at random from the free surface. A destination for the particle is randomly chosen from one of the three empty lattice sites above the three surface particles directly in front of it, directly in front of it and one step to the right, and directly in front of it and one step to the left. The particle's local friction is calculated [13] by adding up the F_{ij}'s between the particle and its four touching neighbors located at the lattice sites immediately to the left and right of the particle, as well as those immediately below and behind it. This number is compared to the difference between the height of the original particle and the height of the destination of the particle. If this height difference is larger than the sum of the four F_{ij}'s, the particle is moved downhill to the new position. If not, the move is not made.

Step (b) is repeated 2.5×10^4 times before the lattice is discretely rotated again (step (a)). Since for our grid there are $200 \times 10 = 2000$ surface particles, surface particles on average experience 12 moves on step (b).

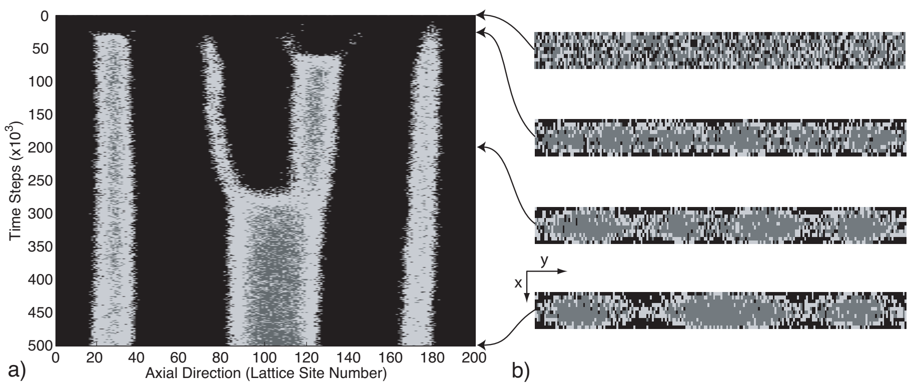

Fig. 5 – Spacetime plot of the discrete Monte Carlo simulation for a total of 5×10^5 timesteps. In each time step the rear slice of the lattice is averaged vertically such that the most abundant particle type is chosen for each axial position. The black, light gray, and dark gray regions correspond to particles A, B, and C respectively.

Our simulations reproduce a wide range of the experimentally observed phenomena. As was the case in the experiment, the mixture quickly segregates radially (within 100 simulation time steps, corresponding to roughly 1 or 2 rotations), with the low friction particles A forming the outer layer, followed by particles B, and then particles C forming the innermost core. Next, axial bands develop (as in Fig. 2(a-d)). Fig. 5 is a space-time plot of a simulation run with 25% A particles, 30% B particles and 45% C particles (cf. Fig. 3). In each time step the rear slice of the lattice, which most closely resembles the laboratory view of the particle mixture surface in a physical cylinder, was averaged vertically and for each axial position the dominant particle type was chosen. Here the black areas correspond to A particles, the light gray areas to B particles, and the dark gray areas to C particles. The initial formation of light gray bands followed by the appearance of dark gray bands exclusively within the light gray regions is clearly observed. One can also see the coarsening of the bands, another feature observed in the experiment. Also, the core structure illustrated in Fig. 2 was seen to persist in the banded state of the simulation as well.

The bulk motion step (a), treats all particles the same, and differing frictional properties of the model particles only matter in the surface flow step (b). Thus, in the model, all segregation, both radial or axial, is due to differing surface flow properties of the three particle types, suggesting that differences in the surface flow properties of the particles in the physical system, may play an important role in the experimentally observed segregation.

In conclusion, we find that mixtures of multiple particle sizes segregate radially by size, with the smallest particles in the center. Mixtures of three and four particle sizes also subsequently segregate axially into bands of fast surface speed small particles within bands of successively larger and slower surface speed particles. In mixtures of more than four particle sizes radial segregation occurred, but we did not observe axial segregation. Mixtures of three and four particle sizes allow us to observe that following radial segregation, bands of the second largest sized particles appear first, and that bands of smaller particles subsequently appear within them. This indicates that the observed axial segregation is a secondary instability of the axially uniform radially segregated state as shown in Fig. 2. The simulation indicates that even a thin flowing layer of particles can exhibit such radial segregation and subsequent axial segregation, and that differences in frictional properties suffice to qualitatively model the segregation pattern and the sequence in which it appears. That such a simple simulation, paramaterized only by friction, can accurately reproduce the experimental results shows the basic robustness of the physical phenomena. Our results show that the number of particle sizes in a mixture is a useful variable in studiying of flow segregation and may be valuable to test the predictive power of segregation models. Additional microscopic measurements of particle motion are underway and will be reported elsewhere.

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