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Pattern formation during mixing and segregation of flowing granular materials

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Abstract

Powder mixing plays an important role in a number of industries ranging from pharmaceuticals and food to ceramics and mining. Avalanches provide a mechanism for the stretching and folding needed to mix granular solids. However, unlike fluids, when particles dissimilar in size, density, or shape flow, they can spontaneously demix or segregate. Using magnetic resonance imaging, we track the transport of granular solids in a slowly rotating tube both with and without segregation effects. Compared with experiments in a 2-dimensional rotating disk partially filled with colored particles, the mixing kinematics and the granular pattern formation in a tube are changed by an axial flow instability. From simple physical principles we argue how size and density segregation mechanisms can be made to cancel, allowing good mixing of dissimilar particles, and we show experiments verifying this. Further experiments isolate the axial transport in the slowly rotating tube. Axial transport can appear faster with segregation than without.

1. Introduction

Granular solids flow and mix in novel unexpected ways, but do so unlike liquids. For instance, nothing as settled as the Navier–Stokes equations exists for granular flow, and a continuum description may not even be appropriate [1]. Our ignorance notwithstanding, the transport and mixing of solids plays an important role in many industries, ranging from pharmaceuticals and food to ceramics and minerals. In proportion to its technological importance [2, 3] however, almost nothing is known about the basic physics of flowing granular materials or how they mix. To suggest why this is so, consider that from a theoretical viewpoint, mixing problems usually appear complex and unwieldy, while from an applied viewpoint, it is often considered

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expedient to focus on the details of particular cases without ever searching for underlying structure. As a step towards uncovering basic principles, we believe it is desirable to identify granular mixing problems with the potential for focused experimentation and theoretical tractability. Recent work has considerably advanced the basic understanding of granular flow [1, 4, 5] and granular mixing [6].

In this paper we report Magnetic Resonance Imaging (MRI) experiments of 3-dimensional granular mixing in a slowly rotating tube. MRI techniques are particularly well-suited for the study of granular flows: they are non-invasive; they see through opaque solids; and, since granular solids "freeze" when external forcing is removed, imaging time-resolution is not an important consideration for many issues of interest. Normally solids cannot be imaged with MRI, but certain seeds contain natural oils which give a good signal [7]. Ours is not the first use of MRI on granular solids, but previous uses [7, 8] could be characterized, to make a fluids analogy, as Eulerian in spirit whereas ours is distinctly Lagrangian. We note other non-invasive imaging techniques (e.g. positron and gamma-ray tomography [9, 10]) are being developed and experiments do seem on the verge of providing a wealth of new information on 3-dimensional granular flows.

In the rest of the paper we first briefly describe the experiments then present data for radial and axial mixing both with and without segregation effects from grain size and density differences.

2. Experiment

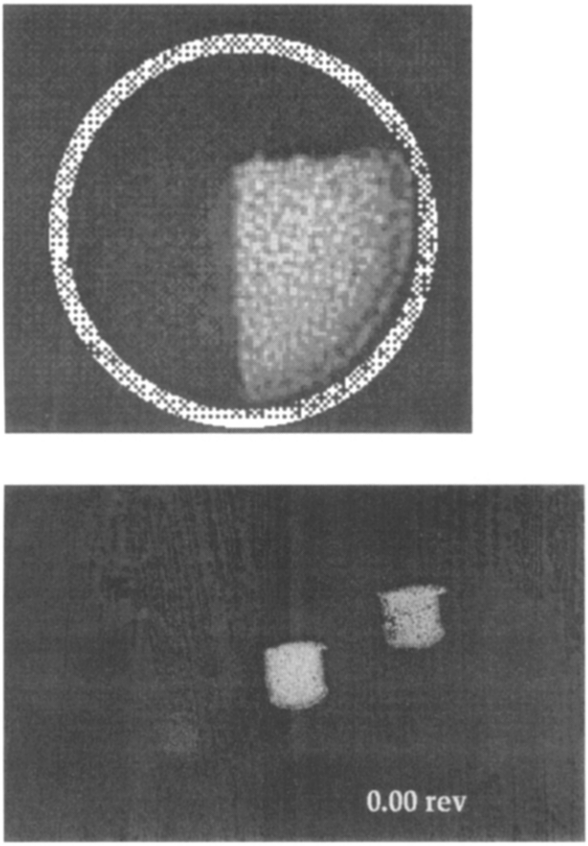
The granular materials we use are brown or yellow mustard seeds, glass beads, or sugar balls. The glass and sugar are smooth spheres; the mustard seeds are approximately smooth and spherical. Table 1 lists the particle sizes and densities. These materials allow several size and density ratios with which to systematically explore segregation effects.

The tube, which is plexiglass, 5 cm in diameter and 15 cm long, fits in the bore of a 2 T magnet. Based on a 1.5 mm particle, the tube dimensions are 33×100 particle diameters. Using a 3 d radial pulse sequence [11] modified for larger arrays, data resolution in the plane perpendicular to the tube axis in 128^2 with 128 data planes spaced uniformly along the tube axis. Mustard seeds give a good MRI signal – seeds are white in the images – while glass and sugar give no signal.

Experiments begin with segregated volumes of seeds and other grains. Fig. 1 shows two types of initial conditions. Fig. 1(a) shows the initial condition in the tube for radial mixing experiments. Shown is the middle slice perpendicular to the long axis. Equal volumes of mustard seeds and sugar balls fill $\frac{3}{4}$ of the tube volume. Seeds show as white on the right. Sugar balls fill the corresponding locations on the left but do not appear in the MR image. The initial condition is the same throughout the length of the tube. MR does not image the tube: the white line showing the tube is drawn afterwards. The oddities near the tube boundary are due to susceptibility differences.

Table 1
The average and standard deviation of the distribution of particle diameters and densities

	Brown mustard	Yellow mustard	Small glass	Large glass	Sugar balls
size (mm)	1.4 ± 0.2	1.9 ± 0.2	1.6 ± 0.1	2.0 ± 0.1	1.6 ± 0.1
density (g/cm^3)	1.7 ± 0.2	1.8 ± 0.3	2.6 ± 0.1	2.6 ± 0.1	1.4 ± 0.3



(a)
(b)

Fig. 1. (a) Initial condition in the tube for radial mixing experiments. (b) Initial condition in the tube for axial mixing experiments. Seeds show as white. Other grains do not appear in the MR image.

Fig. 1(b) shows the initial condition in the tube for axial mixing experiments. Shown are five slices perpendicular to the long axis. Several slices in the middle of the tube have seeds; other grains fill out the rest of the tube volume to $\frac{3}{4}$ full. Here we do not draw in the tube.

During experiments, the tube rotates counterclockwise about its long axis $\frac{1}{4}$ revolution, stops during image acquisition, then resumes rotation. During image acquisition, grains are not moving. As the tube rotates, the flat material surface rotates too until reaching a point where the static equilibrium of the particles fails and an avalanche occurs. After the avalanche the surface returns to its angle of repose. The surface avalanche motion accomplishes the mixing. The rotation speed is slow enough so that an avalanche ceases completely before the next avalanche begins. This separation of the rotation timescale from the avalanche timescale defines slow rotation. Note that the motion of the surface, which causes mixing, is iterative [6].

3. Mixing and segregation

When the properties of a collection of grains are dissimilar or distributed across a range of values, the grains will segregate as they move instead of mixing. Some questions of interest are: How closely matched must particle properties be for segregation to dominate? Do all segregation mechanisms produce similar effects? How do mixing patterns change as segregation effects are increased? Here we consider only size ratio s and density ratio d differences. Other effects, such as from shape and cohesion, are left for future consideration.

3.1. Radial transport

Fig. 2 shows radial transport experiments with different combinations of s and d. The figure shows five slices, displayed diagonally, the central slice in the middle, from 128 total slices perpendicular to the tube axis. Fig. 2(a) shows mixing with no segregation[1] in a tube $\frac{3}{4}$ full of equal volumes of sugar/brown mustard: $s = 1.1 \pm 0.2$, $d = 0.8 \pm 0.2$. After 8 revolutions, grains are well-mixed throughout the tube. Segregation forces arise from size and density differences (among others) of the individual grains [13]. For similar densities and different sizes, it is well-known that larger particles segregate to the outside of a tumbling container [3]. Fig. 2(b) shows density segregation with similar sizes and different densities using smaller glass beads/brown mustard: $s = 1.1 \pm 0.2$, $d = 1.4 \pm 0.2$. The denser glass beads go to the center of the tumbling container. Fig. 2(c) uses larger glass beads/brown mustard: $s = 1.4 \pm 0.2$, $d = 1.4 \pm 0.2$. It shows a surprising outcome when both size *and* density segregation forces operate at the same time, which is the more typical case both in nature and in industry. The glass is both denser and larger. Density segregation tends to move the glass to the inside of the tumbling container; size segregation tends to

[1] Axial banding, the most discussed form of segregation in a tube [12], does not appear in the slowly rotating tube.

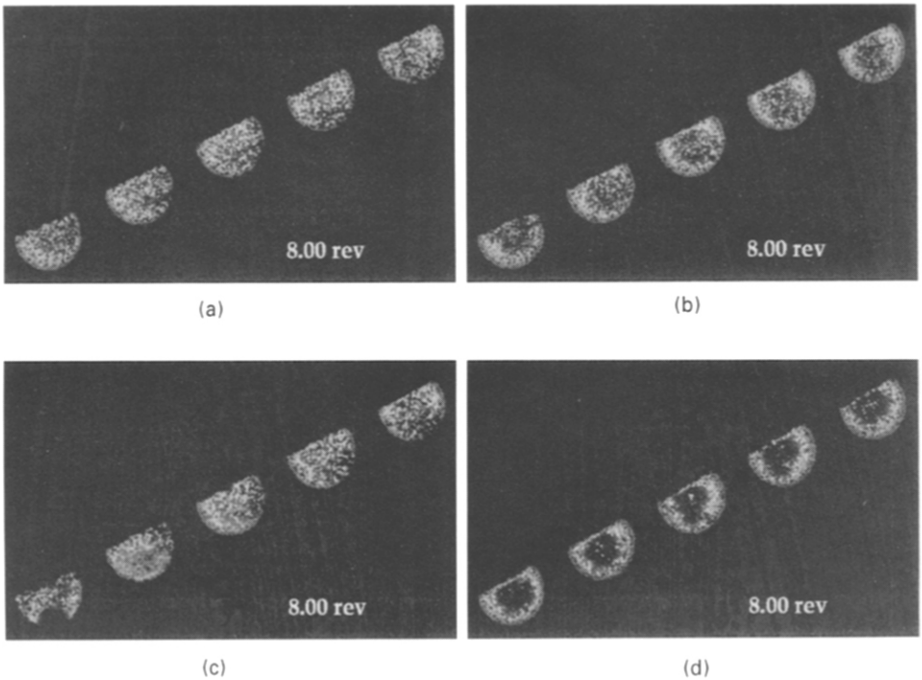


Fig. 2. Mixing with and without segregation effects. Five slices from experiments initialized as in Fig. 1, all after 8 revolutions of the tube. (a) Particles of same size and density mix. (b) Particles of similar size and different density segregate. (c) Particles of different size and different density can in some cases mix due to cancellation of the segregation forces. (d) Reverse the sense of one of the segregation mechanisms in (c) to get the strongest segregation.

move the glass to the outside. The end result is that the segregation forces approximately cancel, giving good mixing where it would not normally be expected. This suggests there is a line in the $d-s$ plane where mixing is always good, and where segregation becomes ever stronger the farther you move from this line. In contrast to Fig. 2(c), Fig. 2(d) shows the same density ratio but with the larger yellow mustard and smaller glass beads: $s = 0.8 \pm 0.1$. Now, size segregation moves the larger mustard to the outside and density segregation moves the denser glass to the inside, with the result that by manipulating the sence of the various segregation mechanisms, we have the most strongly segregating mixture of all.

In light of 2-dimensional granular mixing experiments, it is striking how little difference there is in Fig. 2 from slice to slice along the tube's axis. In a 2-dimensional mixer, a stable non-mixing core region forms regardless of material differences whenever the container is more than half full [6]. Fig. 3 shows a situation in an 2-dimensional experiment mixing 1.8 mm sugar balls and 0.6 mm salt cubes in a slowly rotating disk after 4 revolutions of the disk. Radial size segregation is quickly completed, though the central invariant core remains a prominent feature. In

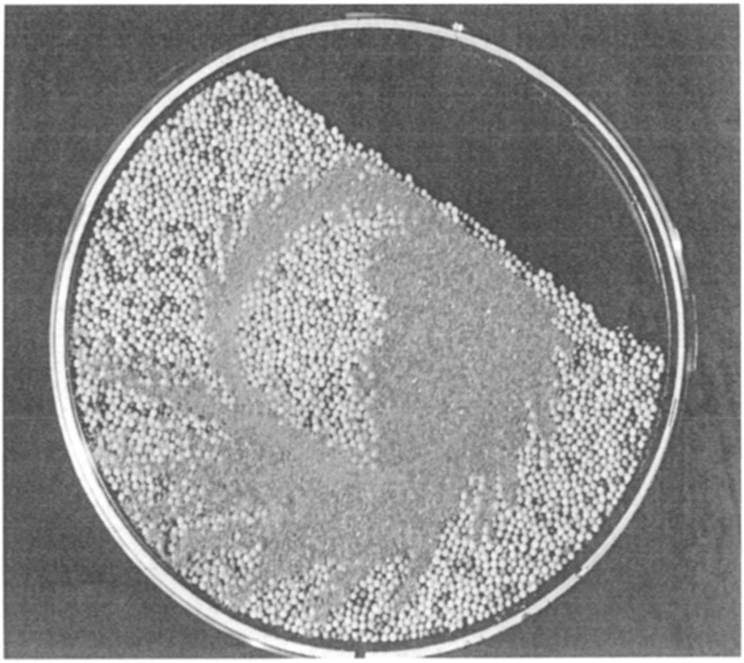


Fig. 3. Photograph of a 2-dimensional experiment mixing 1.8 mm sugar balls and 0.6 mm salt cubes in a slowly rotating disk after 4 revolutions of the disk. The patterns do not change with further disk rotation, but the layering at the outer segregation boundary precesses.

addition, the segregation creates a pattern of alternating layers on the outer (i.e. outside the core) segregation boundary. This 2-dimensional pattern is stable and does not change with further rotation of the disk, though the layered pattern does precess in the direction of rotation. In contrast in 3d we observe from videos of the motion that a core starts to form but subsequently breaks up after 3 revolutions. The elimination of the stationary 2-dimensional segregation patterns from 3-dimensional flows seems to be due to the axial motions described below.

3.2. Axial transport

Because of their industrial use, many researchers have modeled axial transport in 3d tube mixers, usually as a diffusion process and averaging over the radial directions [13, and references therein]. Fig. 4(a) shows the progression of an axial transport experiment with brown mustard and sugar balls, i.e. no segregation. The seeds begin confined to a thin ($\approx 20\%$ of grain volume) slab in the middle of the tube. As the tube rotates, the seeds spread out. Several observations may be made: (1) Axial transport is much slower than radial transport. It takes more than 200 revolutions to disperse the seeds. (2) Seeds disperse axially along the tube boundary and then mix radially. This is

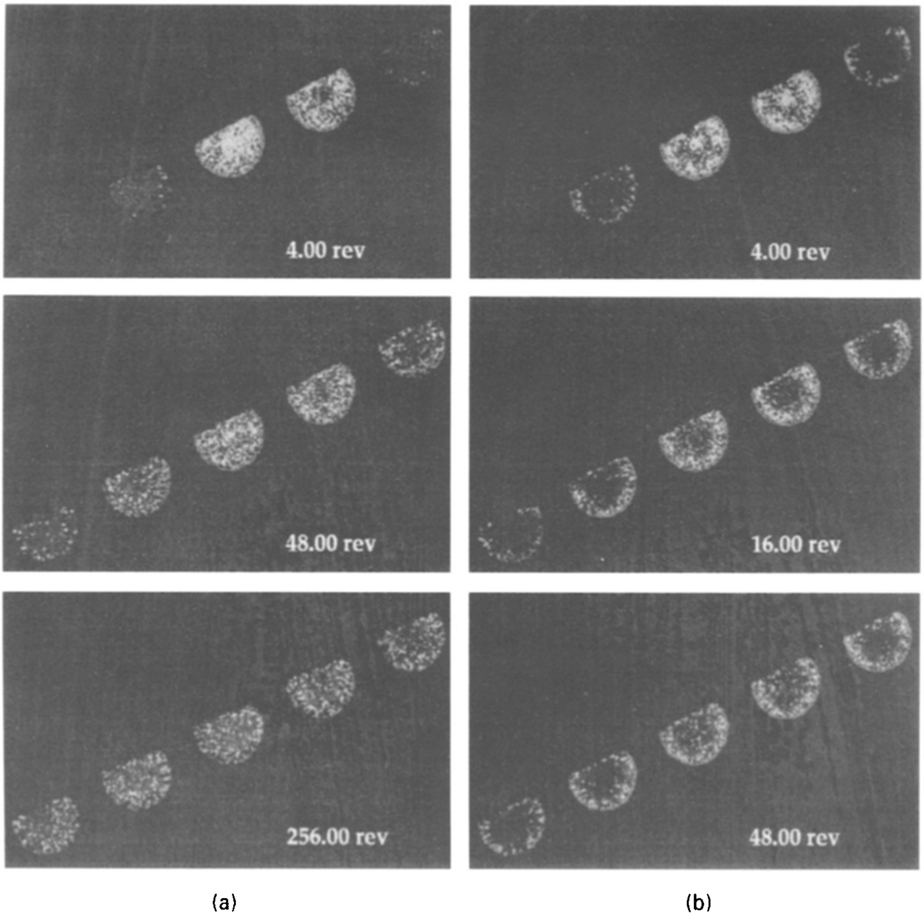


Fig. 4. (a) Axial transport with no segregation. (b) Axial transport with segregation. Five slices from experiments initialized as in Fig. 1(b). Frames are labelled by the number of tube revolutions.

physically plausible given that the only motions mixing the material are the avalanches across the free surface.

Fig. 4(b) shows the same experimental setup but with the brown mustard and smaller glass beads. With segregation added, the central axial region in the initial slab in quickly depleted of seeds. The seeds disperse along the container boundary but cannot mix radially to the interior due to segregation forces. The stable distribution is reached in about 40 revolutions instead of the 200 revolutions of Fig. 4(a). The rate of change of the widths of the distributions became too small after respectively 40 and 200 revolutions to continue to track. Presumably, based on the data of Fig. 2, if we rotate long enough the final distribution of particles will be uniform.

Note the implication segregation has on a mixing measure: if grain concentration were measured in a coarse-grained way by averaging over slices (as we have done in

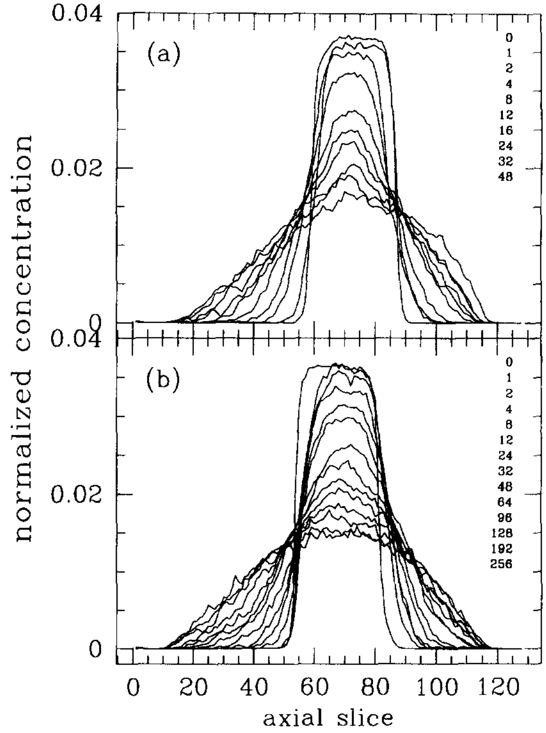


Fig. 5. Normalized concentration summed radially over slices versus the distance along the axis. The initial profile of mustard seeds spreads as the tube rotates, reaching a stable profile much faster with segregation (a) than without (b). The numbers of revolutions on the right timestamp the individual profiles.

Fig. 5), such measurements would show that segregation enhances transport, an incorrect result illustrating the importance of identifying the mixing patterns along with any coarse-grained measures of mixing rate.

4. Future directions

Recent studies of granular flow and mixing raise a host of questions with combined technological and scientific import, e.g. what are the detailed segregation mechanisms in a device for given particle characteristics; what are strategies for suppressing *or* enhancing segregation; are there granular manifolds and mixing templates analogous to those so important in fluid mixing; what are the mixing patterns and rates in commercial mixers? A goal of these and future experiments is to measure granular transport systems in enough detail to answer some of these questions, leading hopefully to greater predictive facility and a deeper level of physical understanding of the flow of granular materials.

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