Bulk Segregation in Rotated Granular Material Measured by Magnetic Resonance Imaging

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Many mixtures of granular materials tend to segregate when tumbled in a rotating horizontal cylinder, with the different components separating into bands of relatively pure single concentration along the rotational axis. The use of magnetic resonance imaging (MRI) to study concentration variations within a mixture of different sized spheres segregated in this manner indicates a more complicated structure within the bulk than can be observed or even inferred from surface measurements. Moreover, when the rotation speed is reduced and the homogeneous mixed state appears to be restored on the surface, the MRI data reveals that the radially segregated state persists. [S0031-9007(96)02069-8]

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An intriguing property of granular materials such as sand or powders is the tendency for mixtures to segregate, even in situations such as shaking or rotation where one would expect mixing. Rotating cylinders or drum mixers are used for mixing granular materials as well as for other industrial processes including calcinating and drying granular materials. (See, for example, Refs. [1] and [2], and references therein.) In some cases, however, horizontal drum mixers will also segregate the individual species. In the case of axial segregation, the components separate into bands of relatively pure single concentrations along the rotational axis of the cylinder (see, for example, Refs. [3–10]). In certain cases the axial bands remix when the rotation speed is reduced; the segregated state is restored when the rotation speed is increased again [9,10]. Although it has been thought that the segregating and remixing are driven primarily by surface effects [9,10], we report measurements of a more complicated segregation structure beneath the surface than can be seen from outside the cylinder, which may partially influence the axial segregation.

All granular mixtures that axially segregate in horizontally rotated cylinders also display radial segregation [4–6,9–11]. For a mixture of different sized particles, the smaller particles form a radial core before axial segregation occurs. Rajchenbach and co-workers [12] and Cantelaube and Bideau [13] have studied radial segregation of different sized particles in a two-dimensional cylinder, that is, a cylinder that is only one particle long. However, there has been no published analysis of radial segregation in a three-dimensional system, nor any theory relating the development of the radial segregation within the bulk to the axial segregation observed from outside the cylinder.

Magnetic resonance imaging (MRI) can be used to examine the development of the axial and radial segregations inside the bulk of granular media. Altobelli and co-workers first showed that liquid state MRI techniques can noninvasively image arbitrary planes within the bulk of fluid-filled granular materials [14]. These techniques have been used to study granular flow in rotating drums [11,14–16] and vertically shaken containers [17,18]. Ehrichs and co-workers used MRI to study convection of granular material in a vertically shaken cylinder [17,18]. Metcalfe and Shattuck used MRI to investigate the effect of segregation on the measurement of axial transport of granular solids in a slowly rotating drum [16]. They also discussed how varying more than one particle property in the mixture affected the segregation process. Nakagawa and co-workers investigated the dynamics of granular flow in a partially filled rotating drum by measuring the velocity and concentration profiles in planes perpendicular to the cylinder axis for a system of single-sized mustard seeds [11,14]. This group also studied the migration of the segregated radial core for a mixture of different sized liquid-filled spheres by imaging planes perpendicular to the cylinder axis while the cylinder was rotated within the magnet [15]. We report additional MRI studies on the segregation of granular materials in a rotating drum in which we attempt to develop a more complete description of axial and radial segregation within the bulk of the material as it relates to surface observations. Towards this end, measurements were taken of the component concentrations in a slice parallel to the cylinder axis, in addition to transverse slices as measured in Refs. [11,14,15]. Measurements of the segregated bands were also taken outside the cylinder and compared with those taken within the bulk using MRI.

When the MRI images are compared to the banding pattern observed from outside the cylinder, it is apparent that the structure within the bulk is often more complicated than the surface state. While these studies confirm a direct relationship between the segregated bands of small beads seen from the surface and undulations in the core of radially segregated small beads, they also reveal that surface observations are often insufficient to interpret the bulk segregation. For example, there are sometimes more axially segregated regions within the bulk than can be seen from the surface which over the course of continued rotation never develop into bands that reach the surface. Furthermore, when the rotation speed is reduced and the mixture appears to have returned to a homogeneous state on the surface, the MRI images show that the radial core of segregated small beads remains.

The experimental setup used is similar to that employed by Nakagawa, *et al.* [11,14], although we use a mixture of one MRI-sensitive component and one that is not MRI active, so that a concentration image will directly show the distribution of the MRI active species. The MRI-sensitive beads are 1 mm diameter pharmaceutical spherical pills of density 1.1 g/cm^3 while the inert beads are 3 mm plastic spheres of density 1.6 g/cm^3. The spheres were rotated in two different acrylic cylinders, one 45 cm long and the other 24 cm long, both $\sim$7.5 cm in diameter. Pharmaceutical spheres were taped around the outside of the cylinders at 5 cm axial intervals to provide reference points to indicate where the cylinder walls are in the images. An NMR imager/spectrometer (Nalorac Cryogenics Corp.) was used at The Lovelace Institutes with a 1.9 Tesla Oxford superconducting magnet. The 31 cm magnet bore is horizontal, so a cylinder could be placed into the birdcage coil of the magnet without disturbing its contents. Image analysis was performed on a Power Macintosh 7100/80 computer using the public domain NIH image program.

The cylinder used was approximately one-half filled (by volume) with a binary 50-50 mixture of plastic spheres and pharmaceutical pills. The partially filled cylinder was rotated outside the magnet where measurements of the surface segregation were taken. Rotation was then stopped, and the cylinder was imaged in the bore of the magnet with spin-warp static NMR imaging. The cylinder was removed from the magnet and this procedure was repeated. In this way the evolution of the radial and axial segregation could be serially monitored.

We imaged a 5 mm thick slice either perpendicularly to the axis at several points along the length of the cylinder or parallel to and containing the axis of the cylinder as schematically illustrated in Figs. 1(a) and 1(b), respectively. The intensity of the image is directly proportional to the local concentration of the smaller MRI-sensitive spheres; the maximum intensity light regions contain only the MRI-sensitive, i.e., small spheres.

Images taken parallel and perpendicular to the long axis of each cylinder prior to rotation confirmed that our technique of shaking and rocking the particles inside the tube achieves a relatively uniform mixture. Figure 2(a) schematically illustrates the system as seen from outside the cylinder after the 24 cm long cylinder had been rotated for approximately 5.5 min at 30 rpm. The system is axially segregated into three bands of small spheres and four bands of large spheres.

The images perpendicular to the axis [Fig. 2(b)] at axial locations indicated in Fig. 2(a) show that the pattern is roughly cylindrically symmetric around the axis at any given location, and thus the images containing the cylinder axis [as in Fig. 1(b)] are sufficient for a representation of the state of the mixture below the surface. Figures 2(b) and 2(c) reveal that the small beads have receded from the endplates, but the core of small spheres near the center of the rotational axis exists along the entire length of the cylinder, even within the bands of the larger 3 mm spheres. Figure 2(c) clearly shows that bands of small beads visible on the surface are merely those parts of the radial core that are wide enough to extend through to the surface. Thus, at least for this case, the bands of large beads are isolated rings that encircle the core of small beads at certain axial locations. When the rotation speed was reduced to 4 rpm, the axial bands disappeared from the surface. After 45 min of this slow rotation, there were no vestiges of axial segregation, but the radial segregation remained, as indicated in Fig. 2(d).

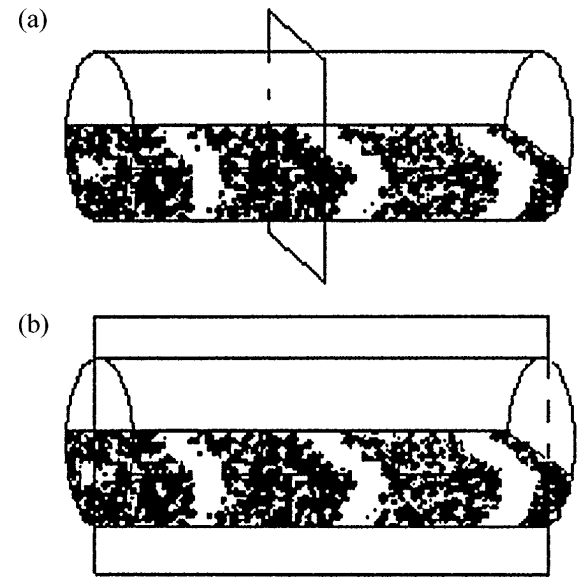

FIG. 1. Sketch of the axial segregation effect in a horizontal cylinder, indicating the orientations of the MRI images used. Both were vertical; one was perpendicular to the axis of the cylinder (a), while the other was parallel to and contained the axis of the cylinder (b). The former was taken at various positions along the length of the tube.

Figure 3 clearly demonstrates that surface observations do not reveal the full complexity of the segregation within the bulk. This MRI data shows additional segregated structure within the bulk that has no corresponding concentration changes visible from the top surface. Figure 3(a) is an image taken parallel to the cylinder axis for the system after the spheres had been rotated for approximately 9 min at 30 rpm in the 45 cm long cylinder. As in the previous example, the mixture appeared from the outside of the cylinder to have segregated both radially and axially. Figure 3(b) is a concentration profile of the top surface indicating, also as before, three sharp bands of small spheres and four bands of large spheres over the outside surface as was observed from outside the cylinder. However, from the MRI image, it is apparent that five areas of a higher

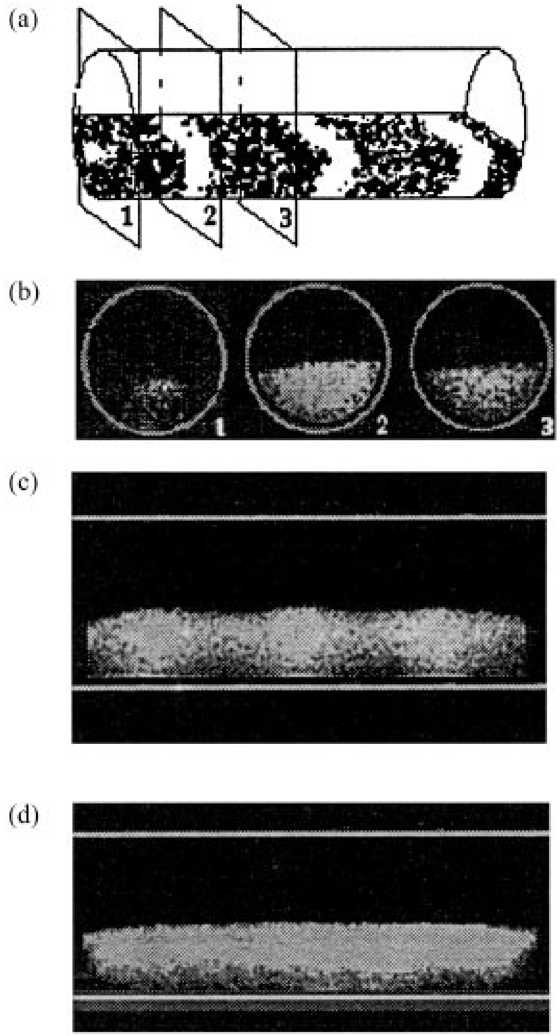

FIG. 2. MRI data for a mixture of small pharmaceutical spheres (MRI active) and plastic spheres rotated in a 24 cm long cylinder. (a) schematically illustrates the state of the system as viewed from outside after rotation for 5.5 min at 30 rpm. The mixture appears to have segregated radially and axially. (a) shows the approximate locations of the planes from where the images of (b) were taken: near the end of the cylinder (1), within a small sphere band (2), and within the large sphere band (3). (c) is the image along the plane parallel to the axis of the cylinder. The brighter regions represent higher concentrations of 1 mm spheres. (d) is the image along the same plane after the cylinder was rotated at 4 rpm for approximately 45 min indicating that the radial segregation remained even though the axial bands had disappeared.

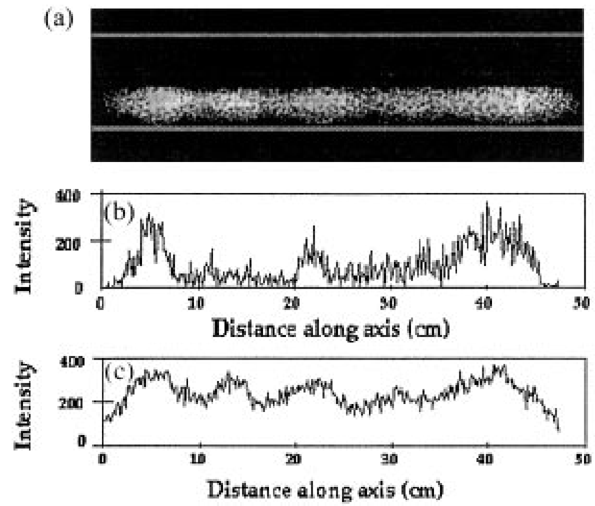

FIG. 3. Data from MRI image parallel to the axis of the cylinder (a) of a mixture of 1 mm pharmaceutical and 3 mm plastic spheres after 9 min of rotation in a 45 cm tube. (b) MRI intensity along the top surface of the system, verifying three axial bands of small spheres as seen from outside the cylinder. (c) Average intensity as a function of position along the axis, from the image shown in (a), indicating five regions of a high concentration of small spheres.

concentration of the smaller spheres are present along the axis of rotation within the bulk of the material, as demonstrated by the average concentration along the axis presented in Fig. 3(c). Two of these regions never extended to the surface, but instead, with further rotation, merged into neighboring bands.

Magnetic resonance measurement of concentration profiles is an invaluable technique for studying the radial and axial segregation in the bulk of rotated granular media. These experiments show details within the bulk that cannot be inferred from surface observations. First, the remixing that occurs when the rotation speed is reduced appears limited to axial remixing, while the radial segregation persists. This supports the conjecture that the radial segregation occurs as the system attempts to minimize its compactivity, as stated in Refs. [9] and [10]. Second, the observation that some axially segregated regions may exist in the bulk without ever extending to the surface implies that axial segregation may not be driven exclusively by a surface phenomenon such as differences in angles of repose as suggested in Refs. [9] and [10]. The segregation effect may result from variations in the radially segregated small bead concentration within the bulk, which then extends to the free surface. The dynamic angle of repose may therefore reflect these variations in the bulk concentration rather than cause the segregation process. Future studies will include correlating changes in the radial core with the development and evolution of the axial segregation bands. Similar studies as a function of the percentage of smaller beads in the mixture may reveal the relative importance of the axial and radial segregations. There may be a minimum fraction of the smaller spheres below which there will not be enough small beads to support both axial and radial segregations.

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