Modes of granular segregation in a noncircular rotating cylinder

K. M. Hill,* Nitin Jain, and J. M. Ottino
Department of Chemical Engineering, Northwestern University, Evanston, Illinois 60208
(Received 31 July 2000; published 14 June 2001)

Axial segregation is a well-known example of segregation of granular materials. However, at present, there is no conclusive explanation as to why it occurs. Most studies of axial segregation to date are based on cylinders with circular cross sections, and models focus on the character of the surface flow without accounting explicitly for the influence of any subsurface detail. The present experiments demonstrate that the cross section of the mixer has a significant influence on axial segregation and that subsurface dynamics are, in fact, important. Unlike circular mixers, in square mixers the subsurface segregation patterns change with filling level, as does the time dependence of axial segregation. Furthermore, when radial segregation patterns in noncircular mixers most closely resemble that observed for circular cylinders, the time dependence for axial band formation deviates the most. These results challenge segregation theories of axial segregation that ignore subsurface effects.

DOI: 10.1103/PhysRevE.64.011302 PACS number(s): 81.05.Rm, 05.45.Gg, 47.54.+r, 64.75.+g

I. INTRODUCTION

Axial segregation in a rotating drum mixer is one of the most striking examples of segregation of granular materials. Segregation is an important problem for industries ranging from pharmaceuticals to paper processing to agriculture. While the scientific community is developing an ever-improving understanding of granular materials, their tendency to segregate in an often nonintuitive manner serves as an important test bed for newer models.

When a circular drum mixer is partially filled with binary mixtures of granular materials differing only slightly in size, density, or shape, the particles segregate into seemingly pure bands along the axis of rotation. This effect—termed axial segregation—is well known and was first reported by Oyama in 1939 [1]. There is no complete theory as to why this occurs. A few experimental observations have emerged though. For many systems, the particles first separate radially in the r,θ plane perpendicular to the axis of rotation. This occurs within a single rotation [2]. Let us denote the mass fraction of one of the components at time t as $c_A = c_A(r,\theta,t)$. We define *standard* radial segregation, normally referred to as radial segregation in the literature, as the case when $c_A = c_A(r)$. This effect is seen in circular quasi-two-dimensional (quasi-2D) containers and is apparent within a single rotation [2–4]. The situation in a long cylinder is more complex. In many systems, in order 10^2 rotations, the particles separate further into bands along the axis of rotation z, leading to relatively pure monodisperse regions, $c_A = c_A(r,z)$. Thus, in both radial and axial segregation, after initial concentration fluctuations die out, the long-term stable patterns are largely independent of θ. This picture has been verified experimentally using nuclear magnetic resonance imaging (MRI) for subsurface studies [5,6].

One explanation for axial segregation holds that standard radial segregation is a prerequisite for axial segregation. (See, for example, Ref. [7] and references therein.) Evidence from MRI supports this view [5,6,8]. After this, concentration fluctuations along the axis of rotation work as seeds for axial bands. Fickian diffusion, which would normally remix these fluctuations, is foiled when there is a significant difference between the dynamic angle of repose of the mixture and that of one of the components. This can work as a positive feedback mechanism that tends to strengthen any initial concentration fluctuation. The argument treats the θ dependence of the radial segregation only cursorily, as long as the radial segregation leaves the larger or lighter material dominating the top of the flowing layer. Implicitly, all models developed to date have been developed for circular drums where the geometry is independent of θ.

Recent mixing-segregation experiments in noncircular quasi-2D mixers show that the physics is considerably more complex than in the circular case. The (averaged or continuum) flow field induced by time periodic boundaries is, in general, chaotic [9]. Chaotic advection interacts in nontrivial ways with density and size segregation in granular materials and gives rise to self-organized structures. These segregation patterns are critically dependent on the degree of filling, particularly near the half-full level [9,10].

The second column from the left of Fig. 1 shows computational results in terms of Poincaré sections (based on a model reported in [9,10]) and experimental observations (third column from left) for different fill levels of 2D square and circular mixers. The mixers have cross-sectional areas of 150 cm^2 and thickness 9.5 mm. The experiments are performed using spherical glass beads of size 0.8 mm and 3.0 mm diameter. For all fill levels the initially well-mixed system segregates radially within 1 rotation at ≈ 1 rpm. For most experiments in the square mixer (and for the circular mixer), this is the ''final'' segregation pattern. For the experiments in the square system where the mixer is filled $\sim 50\%$, it evolves into a petal pattern which becomes the final stable segregation structure. (Note the similarity between this pattern and the Poincaré section for this fill level.) For the 55% full mixer the structure develops into an unstable pattern that has some resemblance to the petal pattern

*Present address: Department of Theoretical and Applied Mechanics, University of Illinois, 104 S. Wright St., Urbana, IL 61801.

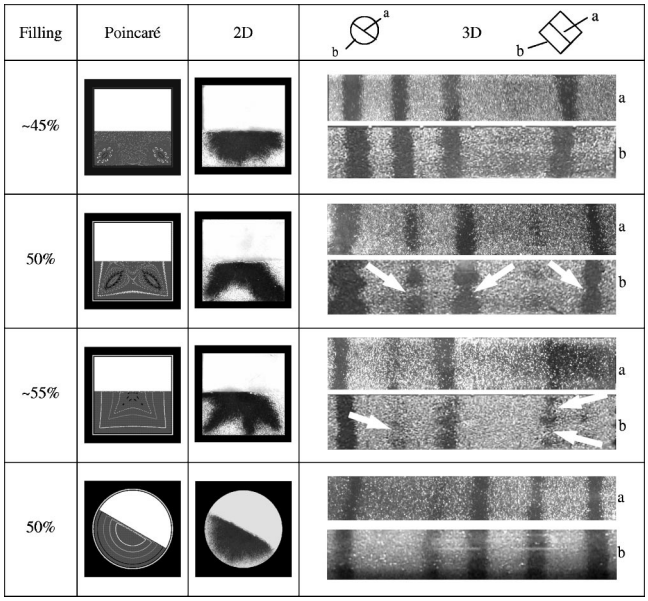

FIG. 1. The Poincaré sections show particle positions from every quarter rotation for the square mixers and every half rotation for the circle. In the square the chaotic-regular structure (location of elliptic points and hyperbolic points) is sensitive to the filling level. The 2D and 3D experiments used 50/50 mixtures of glass spheres, 0.8 mm and 3.0 mm in diameter. The mixer sizes are comparable as described in the text. Radial segregation is observed in 2D circular mixers and most fill levels in square mixers. Near 50% filling, radial segregation gives way to petal formation and at still higher filling ($\sim$55%) a more complicated striping pattern is observed. (Arrows indicate the breaks in the axial bands.) The right column shows axial segregation from the top and bottom of the 3D mixers. The bottom patterns should be compared with the bottom of the patterns of the middle column.

observed in the 50% mixer but is unstable and changes with time. (The segregated pattern for this fill level least resembles the Poincaré section, probably due to complications in the flowing layer as noted in Ref. [10].) We consider now the companion axial segregation experiments.

II. EXPERIMENT

Experiments are conducted in 3D circular and square drum mixers using binary 50/50 mixtures of noncohesive spherical glass beads (Epworth Mfg.©) with sizes ranging from 0.8 to 3 mm in diameter and a density of 2.5 g/cm^3. The square and circular drum mixers are made of Plexiglas; both are 3/4 m long and have approximately the same cross-sectional area, 150 cm^2. The small beads are dyed black for efficient digital image analysis.

To study the equilibrium structures that arise in these systems, the components are initially well mixed. A computer-controlled stepper motor (Compumotor©) rotates the mixers at approximately 8 rpm so that a steady flow with a flat surface is developed. The Froude number $F=\omega^2 L/g$ (where ω is the angular velocity of the tumbler and L is the length scale of the system), is about 0.003 for the circle. Images are obtained at regular intervals throughout each experiment

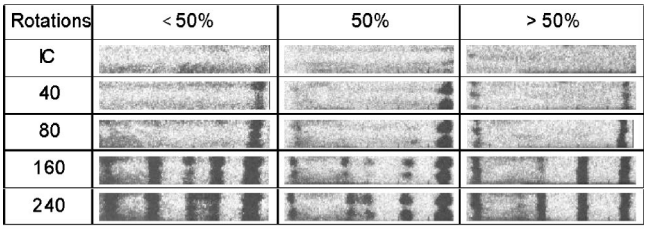

FIG. 2. Time evolution of the pattern formation in 3D square mixers. For 45% filling bands of small beads start forming from the corners of the mixer but these nonuniform regions quickly merge to give homogeneous bands. For 50% filling the bands of small beads are never uniform and appear from the back not as a solid band but as two circular dots (petals in 2D). With additional rotations these regions grow bigger but do not merge, and the bands remain nonuniform. For 55% filling more than two regions of nonuniformity are evident in any band of smaller bead (analogous to striping in 2D). They merge very quickly, however, most likely due to spatial nonperiodicity of stripes [10].

with a Kodak© charge-coupled device camera for quantitative image analysis.

For the circular mixer, ''standard'' axial bands form at all filling levels examined. (See Fig. 1.) The bands look uniform from the *front* (free surface) and the *back* (areas in contact with walls) of the mixer and take approximately the same time to form. However, for the square mixer, the results obtained are more complicated and depend on the fill level.

The 3D segregation patterns in the square mixer appear analogous to the experimental results obtained in the quasi-2D mixers. For all systems, the standard axial banding pattern is observed from the front. In the 40% full square mixer (not shown) and for all filling levels in the circular mixer, the bands appear solid from the front and back, indicating that the bands are independent of θ, as in the traditionally studied radial and axial segregation. In the 45% full mixer there is some evidence of nonuniformity. This may be related to the presence of small islands in the corresponding Poincaré section that quickly connect with the radially segregated core to give standard banding. For the 50% full square mixer, the bands of smaller beads appear even less uniform, with a higher concentration of small beads near the corners of the mixer, noticeable from the back of the mixer. These are strongly suggestive of the patterns in the Poincaré sections and experiments for 2D mixers. While, again, axial bands form in the 55% full mixer and look fairly uniform, there is evidence of additional complicated (nonradial segregation) patterns when viewed from the back. Irregular regions of small beads, indicative of coexistence of the nonstandard radial segregation patterns and axial segregation, occur for this fill level as well. The time dependence of these 3D patterns observed from the back of the mixer is shown in Fig. 2. For the 50% filling fraction the nonuniformity of the axial bands subsists throughout the experimental run; however, this is not the case with 45% or 55% filling.

The correspondence of the 2D information to the 3D case should be handled with care. The 2D experiment is not a slice of the 3D case as the confining walls most likely affect the dynamics. Moreover, we have conducted extensive ex-

periments and we have noted that the 2D results are significantly affected by angular speed, and to a much lesser extent by the size of the system. In fact, in the experiments of Fig. 1, the sizes of the 2D and 3D systems are the same but the angular speed of the 2D system is 1 rpm. Nearly identical results are obtained if the area of the 2D system is increased by a factor of 4. In both 2D systems, when the speed is increased to 8 rpm, the details are less distinct.

III. ANALYSIS

Two different methods of digital image analysis are used to quantify the degree of segregation. For both, digital images of the free flowing surface are obtained every two rotations for each of the filling levels, where the light intensity is representative of the concentration of the lighter of the two components (here, the larger beads). From these, we obtain the average light intensity along the axis of rotation—this produces a graph with a series of peaks and troughs representative of the bands of two sets of beads. Fourier analysis is performed on this series of numbers to switch from the time domain to the frequency domain. As the bands form, one wavelength, representative of the average distance between like-bead bands, becomes dominant. This value is used to represent the time dependence of the band formation. The second method of analysis uses the standard deviation of the mean concentration. Here, again, average light intensity along the axis of rotation is used as representative of the concentration c_i of the large beads at point x_i along the axis. Using $\overline{c}$ as the average along the entire axis, we obtain a measure of mixing using the standard deviation of the mean concentration along the axis of rotation.

Both methods of analysis give similar results. Figure 3 shows the results obtained from standard deviation analysis. In the circular mixer [Fig. 3(a)], for all fill levels examined (40%–60%), the segregation time is roughly equivalent—approximately 130 revolutions. The banding appears to vary slightly with increasing filling level—the lowest fill level takes slightly less time to segregate, while the highest takes slightly longer. Otherwise, the differences are within the errors of the system.

The results from the square mixer [Fig. 3(b)] show that the time dependence of axial segregation depends strongly on the filling level. Counterintuitively, it most closely resembles the results for the circular mixer at fill levels *where the radial core least resembles the segregation pattern for the circular mixer*. At 50% and 55% full, where the transverse segregation pattern appears completely replaced in the square mixer by nonstandard radial segregation patterns, the rate of segregation and the final degree of mixing resemble those of the circular mixer near halfway full. For the 40% full square mixer, the highest filling level where traditional radial segregation occurs, the results are unexpectedly slower than for the circular mixer and appear never to reach the full degree of axial segregation. Similar effects are seen at 60% full when the radial segregation pattern returns with the addition of a small unmixed core.

The difference in the dynamics of segregation of the two cases is shown in Fig. 4. The results from the Fourier analysis for the time dependence of segregation are fitted to the exponential $s=S_0(1-e^{-kt})$ (the initial part of the curve, as

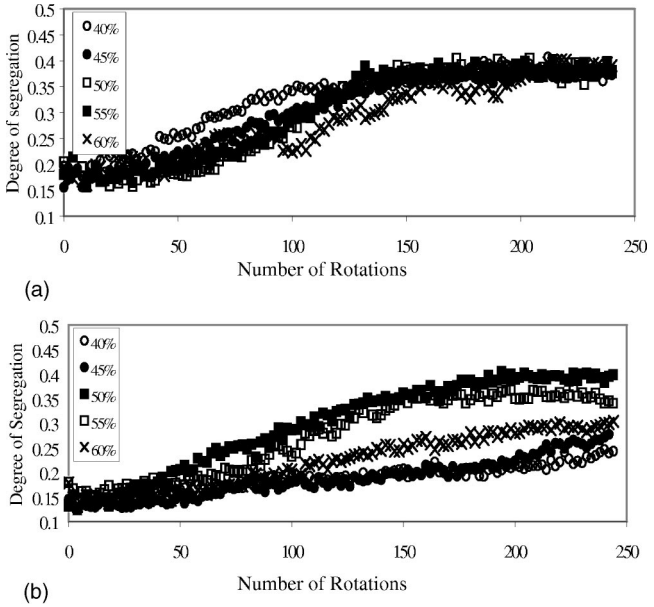

FIG. 3. Results from segregation measurements using the standard deviation about the mean concentration (a measure of the intensity of segregation) for the circle (a) and the square (b). The data are normalized so that a value of 0.5 would mean complete segregation of the two particles. Plots indicate that the segregation dynamics are virtually independent of filling fraction for the circular tumbler and sensitive to fill level for the square mixer.

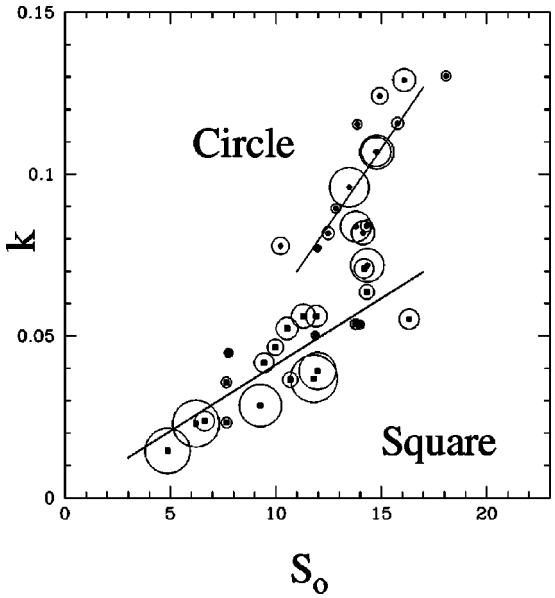

FIG. 4. Analysis of the axial segregation time dependence in the square and circular mixers. The curves from Fourier analysis are fitted to an exponential curve of the form $s=S_0(1-e^{-kt})$, where k is in $(\mathrm{rot})^{-1}$. The diameter of the circles represents the quality of the fitting given by the value of the correlation coefficient; small circles represent good fit. The range of correlation coefficients for these data is from 0.91 to 0.99.

in Fig. 3, appears to be affected by initial conditions and is disregarded in the fitting). The inverse time of segregation k is plotted against the final segregation $s=S_0$. The results show that, while square mixers can reach the same degree of segregation, in general they take much longer to do it.

IV. SUMMARY AND CONCLUSIONS

While traditional radial segregation patterns disappear in noncircular mixers at certain filling fractions, axial segregation still occurs. Furthermore, although changes in radial segregation have a strong influence on the time dependence of axial segregation, these changes are exactly opposite what one might expect. When the subsurface segregation in square mixers deviates the most from standard radial segregation, the time dependence of axial segregation is the most similar. On the other hand, a square mixer with standard radial segregation takes much longer to axially segregate.

Standard axial segregation is observed in both the square mixer and the circular mixer at all the filling fractions examined between 40% and 60%. Experiments using quasi-2D square mixers and external observations in 3D mixers show that the radial segregation pattern is replaced by more complicated segregation patterns. This indicates that axial segregation occurs in the absence of standard radial segregation. Also, while the time dependence for banding in the circular cylinder is approximately the same for all filling levels, it varies significantly with fill level for noncircular mixers, particularly where the nonradial segregation pattern occurs. These observations challenge our current understanding of the interactions of radial and axial segregation and provide a strong argument that the current physical picture of axial segregation needs to be refined.

Conventional thinking suggests that the axial flux and segregation depend on the surface slope (dynamic angle of repose), which is in turn dependent on the layer composition. For circular systems, the layer composition is roughly constant within the time scale of a single rotation. In the square mixers, depending on the fill level, the segregated petals or stripes move in and out of the flowing layer, thus changing the layer composition within a single rotation. Along with this, a periodic change in the flowing layer length also results in a surface slope that is continually changing in time. From a purely geometric standpoint, a square has the additional complexity of transverse chaos that is absent in a circular mixer [9,11]. The coexistence of all these forcing functions in a seemingly simple geometry apparently gives rise to nontrivial effects for axial banding. Any complete theory of axial segregation has to be able to account for these effects, and cannot apply solely to circular cylinders. A more complete experimental study would benefit from companion MRI experimentation to investigate the internal structure of the segregated patterns [5,6,12].

ACKNOWLEDGMENTS

This work was supported by the Engineering Research Program of the Office of Basic Energy Sciences of the Department of Energy, the National Science Foundation, Fluid Particle and Hydraulic System, and the Petroleum Research Fund, administered by the American Chemical Society.

[1] Y. Oyama, Sci. Pap. Inst. Phys. Chem. Res. (Jpn.) **6**, 600 (1939).
[2] N. Nityanand, B. Manley, and H. Henein, Metall. Trans. B **17**, 247 (1986).
[3] E. Clément, J. Rajchenbach, and J. Duran, Europhys. Lett. **30**, 7 (1995).
[4] F. Cantalaube and D. Bideau, Europhys. Lett. **30**, 133 (1995).
[5] M. Nakagawa, Chem. Eng. Sci. **49**, 2540 (1994).
[6] K.M. Hill, A. Caprihan, and J. Kakalios, Phys. Rev. Lett. **78**, 50 (1997).
[7] K.M. Hill and J. Kakalios, Phys. Rev. E **49**, R3610 (1994).
[8] G. Metcalfe and M. Shattuck, Physica A **233**, 709 (1996).
[9] D.V. Khakhar, J.J. McCarthy, J.F. Gilchrist, and J.M. Ottino, Chaos **9**, 195 (1999).
[10] K.M. Hill, D.V. Khakhar, J.F. Gilchrist, J.J. McCarthy, and J. Ottino, Proc. Natl. Acad. Sci. U.S.A. **96**, 11 701 (1999).
[11] J.M. Ottino and D.V. Khakhar, Annu. Rev. Fluid Mech. **32**, 55 (2000).
[12] G. Metcalfe, L. Graham, J. Zhou, and K. Liffman, Chaos **9**, 581 (1999).