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Coupled Axial–Radial Segregation in Rotating Drums with High Fill Levels

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Axial segregation and radial segregation within the granular bed in a rotating drum with a fill level > 50% were experimentally investigated. The axial segregated patterns and the radial segregated patterns were coupled within the bed, resulting in observation of new segregated patterns at the plane along the rotational shaft. The segregated patterns are different from the previous axial band and radial core patterns and show coupled new patterns, including the "cross" patterns, the "band-core-in-core" patterns, and other geometric patterns. The new segregation patterns are strong functions of the fill level and the rotational speed of the drum. The formation of the axial–radial coupled patterns was attributed to the fill level, the free flowing surface of the bed. A virtual drum hypothesis is proposed to explain the formation of the complex segregated patterns. © 2006 American Institute of Chemical Engineers *AIChE J,* 52: 2422–2427, 2006

Introduction

Particulate materials exist widely in nature and industrial products. The mixing of particles with different properties is frequently encountered in nature and is an important process in many industries, such as in the manufacture of pharmaceuticals, polymers, food, and consumer products. In a mixing process, particles are usually placed in a vessel and the vessel is subjected to rotation, vibration, fluidization, mechanical stirring, or other operations to mix the particles. However, Williams and Khan[1] reported that particles of different sizes, densities, shapes, roughness, and elasticity performed segregation when they are mixed. When mixing particles of two different sizes, the particulate mixture segregates according to particle sizes, such as when pouring to form a heap, spontaneous stratification occurs[2,3]; when vibrating in a box, segregated layers (or, the so-called Brazil nut effect) are observed[4,5]; when rotating in a long cylindrical drum, axial bands[6,7] and radial cores[8] are observed. The latter case has been extensively studied because of the popularity of rotating drum mixers in a number of industries. Therefore, we focus on segregation in rotating drums in current work.

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When particles of two different sizes are mixed in a rotating drum and the ratio of the large particle diameter to the small particle diameter is >1.2, the granular mixture segregates.[1] The smaller particles move toward the rotational axis, resulting a radial segregation core. The core is formed rather quickly, usually within the first few revolutions[9] and is usually not visible from the bed surface. Rapid radial segregation is followed by slower axial segregation. The individual species segregates into alternating bands of relatively pure single concentrations along the rotational axis.[9] The band array is dynamic and exhibits a reasonably well defined and quasi-stable emergent wavelength λ.[10]

Axial segregated bands were first reported by Oyama[11] and other later studies.[12-14] Visual observation reported that formation of the bands took place in the vicinity of the end of the drum. The larger particles tend to remain in the end region, whereas a band of smaller ones moves away from it, which is similar to the diffusion process. Some previous studies considered the diffusion-like process to be attributed to the coupling of the concentration to the static angle of repose,[12] the Kolmogorov diffusion,[13] or the dynamic angle of repose.[14] These researchers developed band-formation models based on the diffusion-like process and ignored the radially segregated core. Recently, there are reports indicating that (1) the formation of the segregated bands and cores should not be considered separately and (2) the dynamics of the radially segregated core is

Table 1. Experimental Conditions

Case	Drum	Initial condition	Experimental time (h)	Fill level (%)	Rotational speed (rpm)
A	Long	Fine on top of coarse	1	60, 70, 80, 90	10,20,30
B	Long	Coarse on top of fine	1	60, 70, 80, 90	10,20,30
C	Short	Fine on top of coarse	1	60, 70, 80, 90	10,20,30
D	Long	Fine on top of coarse	2	60, 70, 80, 90	10,20,30

important in the axial segregation process.[9,15,16] The axial bands are seen to arise from core instability, leading to a periodic thickening of the core, and then when the thickened regions break the surface, bands are observed.[16]

Apparently, the study of drum segregation should *simultaneously* consider three-dimensional axial segregation and radial segregation during the course of the segregation process. However, because of the fact that the core is buried in the bed, most of the previous studies on radial segregation were carried out at the two ends of the drum and some continuum models were also developed based on the end-drum core studies.[17-20] Three-dimensional radially segregated core experimental studies have been carried out by the MRI technique[21] and the bulk visualization technique.[16] The MRI experiments showed that details within the bulk could not be inferred from surface observations. When the rotation speed was reduced and the homogeneous mixed state appears to be restored on the surface, the radially segregated state persisted. The axial segregation effect may result from variations in the radially segregated core within the bulk, which extended to the free surface.[21] The bulk visualization technique studied the projected shadows from the mixture of the translucent coarse salt grains and the opaque fine sands.[16] They suggested that the surface concentration, surface shape, and the thickness of the radially segregated core are slaved together throughout the segregation motion and the oscillatory behavior of the segregation process emerges from the complete three-dimensional configuration of the local concentration alone. According to these recent three-dimensional studies on drum segregation, the thickened core seems to be the origin of the axial segregated bands. However, except from very limited studies,[22,23] most of the previous drum segregation studies were conducted with fill levels $< 50\%$ and the cores were relatively small. In this work, experiments were conducted to investigate the coupled axial segregation and radial segregation within the bed in a rotating drum with a fill level $> 50\%$.

Experimental

The segregation studies were performed in two partially filled drums horizontally placed on a pair of rollers, in a similar way to a ball mill. The rotational speed of the drum was directly controlled by the rollers and ranged from 10 to 30 rpm. Two drums were used in the experiments to study the effect of the drum geometry on segregation patterns. The long drum was 199 mm in length and 98 mm in internal diameter; the short drum was 100 mm in length and 98 mm in diameter. The cylindrical column of the drum was made of stainless steel and the two end walls were made of glass to facilitate end-view observation. The cylindrical column of the drum was cut into two shells through the plane along the rotational axis. The two shells could be joined together by two outer rings. A hole was drilled on one of the shells to allow injection of the "frozen" agar liquid (see later discussion) and the hole was covered by a lid. The shape of the inside of the lid had the same curvature as the inside of the shell. All the gaps between the parts were <0.10 mm. Spherical glass particles with density 2521 kg/m^3 were sieved to two size ranges: the white fine beads of sizes between 0.25 and 0.42 mm (mean size 0.35 mm) and the red coarse beads of sizes between 0.59 and 0.84 mm (mean size 0.70 mm) and thus no particles could potentially be trapped by the gaps.

In a typical run, an equal-apparent volume mixture of fine and coarse beads was filled in the drum. The apparent volume occupied by all particles in the drum was set to match the specified experimental filling condition. Two initial conditions were applied: fine particles on top of the coarse particles and coarse particles on top of the fine particles. Each experiment was run for 1 or 2 h. The experimental environment was controlled at 60–70% relative humidity. At the end of each run, transparent agar liquid was carefully poured into the drum through the injection hole without damaging the bed structure. The agar liquid was gelled at room temperature and bed structure was "frozen." A similar solidification method was applied in previous work. For example, a series of experiments was conducted by Muzzio's group at Rutgers University, who used low-viscosity epoxy resin to solidify the granular bed.[24,25] After removing the outer rings and shells, the bed was cut through the plane along the rotational axis. The segregation patterns on the plane along the rotational axis were recorded by a CCD camera. At each experimental condition, at least three times of experiments were performed to verify the reproducibility. Table 1 shows the experimental conditions. The results of cases A and B examine the initial condition effect; the results of cases A and C evaluate the drum geometry effect; and the results of cases A and D show the operating time effect.

Results and Discussion

The segregation patterns at the plane of observation for cases A, B, C, and D are shown in Figures 1, 2, 3, and 4, respectively. In all experimental cases at 60% fill, segregation is observed in which perpendicular radial and axial segregation patterns appear, which we term a *cross-segregated pattern*. One/two band(s) in the axial direction and a core in the radial direction form a cross/double-cross at the plane of observation. When the fill is $<50\%$, the core usually means a tube of overturning and constantly deforming (smaller) grains, whereas for the fill $> 50\%$, the static and unmoving tubelike core exists only at an initial transient. After the initial transient, the tube shape is gradually destroyed and a more complicated structure core is formed. The plane views of the complicated structure core at the plane of observation are shown in Figures 1, 2, 3, and 4. Therefore, we emphasize here that, although the "core" term is applied herein, the terminology of tubelike core is not applicable to fills $> 50\%$. In the radial direction, the size of the core reduces as the rotational speed increases in the long drum in cases A, B, and D and remains approximately constant in the short drum in case C. In the axial

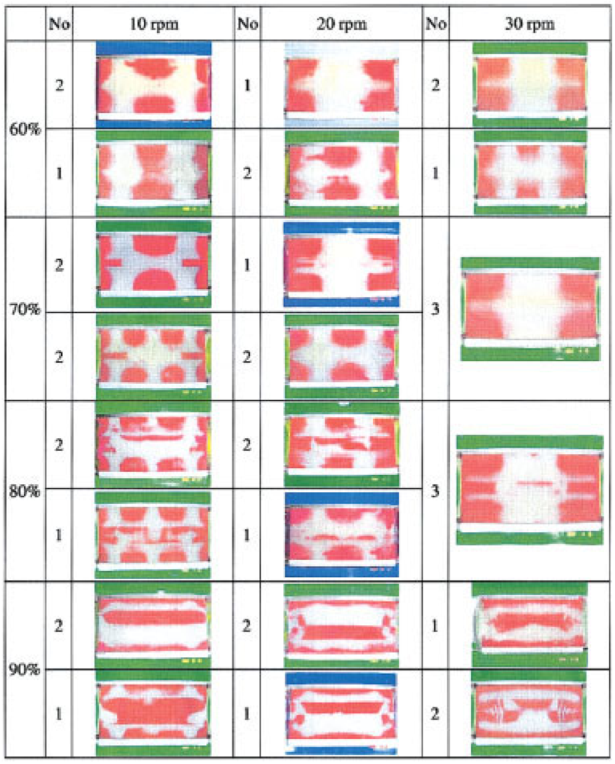

Figure 1. Segregation patterns observed at the plane of observation of different operating conditions in the long drum after a 1-h run for case A.

The numbers in the "No" column represent the observation times in three repeat runs. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com]

direction, the number of the bands decreases as the rotational speed increases in the long drum. At 60% fill, the segregation pattern at the plane of observation is simply the addition of the axial band(s) and the radial core. The formation of such a "cross" pattern is the result of the *independent* axial segregation and radial segregation. The major difference between current observations and previous reports at 60% fill is the size of the core. In previous reports,[26] the core threads the entire tube.

At 70% fills, a red core is developed within the white core (that is, core-in-core), causing a band-core-in-core segregation pattern at the plane of observation in all cases with rotational speeds of 10 and 20 rpm, although the red cores are different in different cases. At 30 rpm, the red core within the white core is not observed. Observations suggest that at 30 rpm, the red coarse particles experience greater centrifugal forces compared to those at 10 and 20 rpm. Therefore, the red particles move toward the cylindrical wall and the red core disappears. In the long drum, the red core threads the entire drum within the white core in case B, whereas in cases A and D, the red core within the white core is blocked by white fine particles and is not able to thread the entire drum. In the experiment, the red coarse particles were wrapped within the white fine particles in the first few revolutions in case B and the core-in-core structure was formed rather rapidly. The initial packing condition (or, the direction of rotation) causes the different red core sizes within the white cores in cases A, B, and D. We illustrate the formation of the core-in-core pattern in more detail below. The observation of the core-in-core structure implies that the previous core study[16] by the bulk visualization technique might not faithfully present the correct core structure.

Previous experiments using radioactive tracers confirmed that granular flow in the transverse plane consists of a thin surface-flow layer and a solid-body motion under the surface-flow layer.[27] Initially, the red coarse particles are placed under the white fine particles (Figure 5a). As the clockwise rotation of the drum begins, the red coarse particles avalanche down the surface-flow layer and the white fine particles move as a solid body (Figure 5b). Because the particles are more active in the surface-flow layer they thus roll faster down the surface-flow layer, causing the slow moving white particles to be wrapped by the fast-moving red particles. When the white particles reach the top of the free surface of the bed, the red particles are wrapped by the white particles (Figure 5c). The wrapping process repeats (Figures 5d and 5e) and a portion of the red particles is completely wrapped by the white particles (Figure 5f). The red particles wrapped by the white particles represents the core-in-core pattern at the plane of observation. The core-in-core structure gradually diminishes (Figures 5g and 5h). The diminution of the core-in-core segregation at the end walls is caused by shearing at the end walls. In a previous report,[8] the formation of the core is caused by the higher likelihood of the smallest particles in a faster flowing circulation layer to become trapped within the gaps of the more slowly flowing layer beneath it and thus to move into that circulation path close to the center of rotation. The mechanism of the formation of the

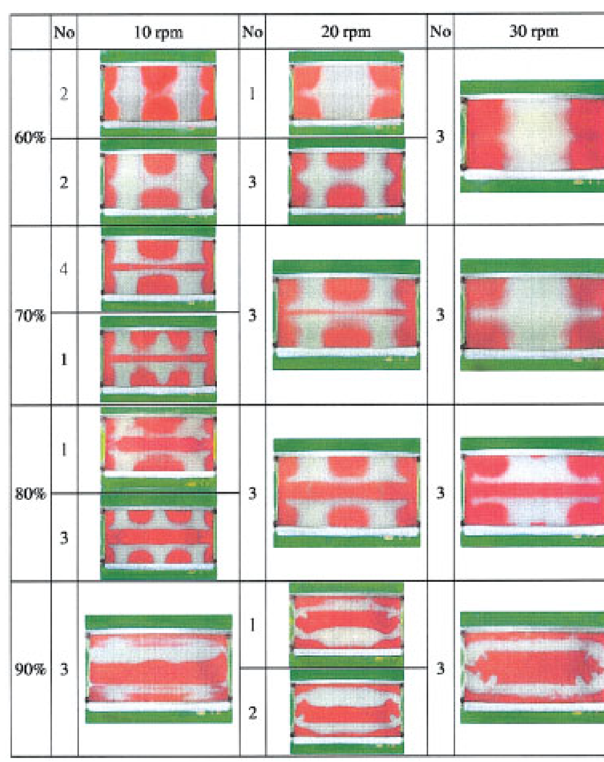

Figure 2. Segregation patterns observed at the plane of observation of different operating conditions in the long drum after a 1-h run for case B.

The numbers in the "No" column represent the observation times in three repeat runs. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com] core-in-core pattern is different from the simple core formation mechanism: the core-in-core pattern is caused by the repeating wrapping mechanism.

Data indicate that the high level of fill stops the observation of this core-in-core structure earlier because most of the early studies focused on fill levels < 50%. For the drum used in current study, the fill level must be >70% fill to ensure that the numbers of the white particle are adequate to exist both at the surface-flow layer and at the solid-body rotating region to wrap the red particles (Figure 5c). Thus, the high fill level and the existing surface-flow layer of the bed are the two important factors causing the formation of the core-in-core structure.

At 80% fill, the core-in-core segregation patterns at the plane of observation are observed in all cases, although the red cores are different in different cases. In case B, the red core threads the entire drum within the white core, whereas in other cases, the red core within the white core is blocked by white fine particles in a more complex fashion. Interestingly, the distribution of the white particles within the red core is approximately symmetrical about the central plane. A comparison of the results of cases A and D shows that the segregated patterns are similar, indicating that the pattern formation process has reached a steady state after a 1-h operation. Although the underlying physics causing the complicated segregated patterns is not well understood, our observation implies that radial segregation and axial segregation are closely linked. Axial and radial segregation should be considered together, at least at high fill level conditions. The theory of the formation of the axial band attributed to core instability and thickening[16,28-30] cannot satisfactorily explain the current observations.

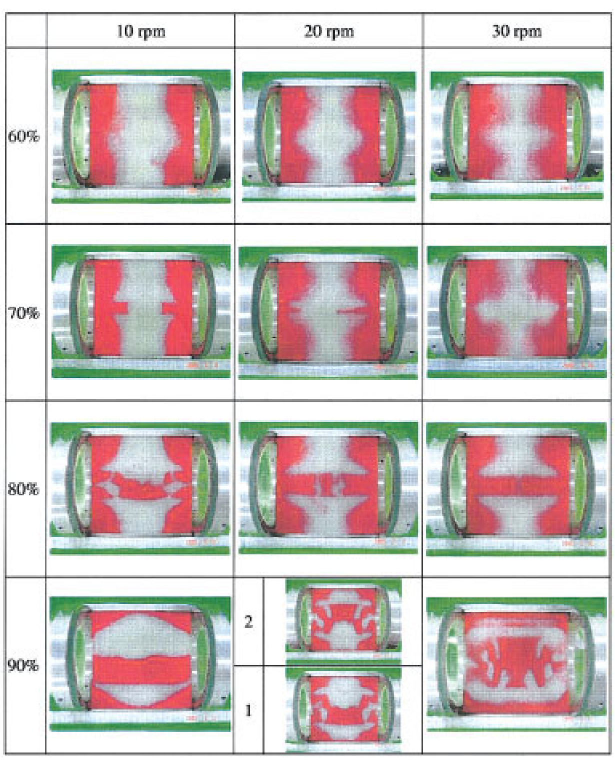

Figure 3. Segregation patterns observed at the plane of observation of different operating conditions in the short drum after a 1-h run for case C.

The numbers in the "No" column represent the observation times in three repeat runs. If there is no "No" column, all three times obtain the same segregated pattern. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com]

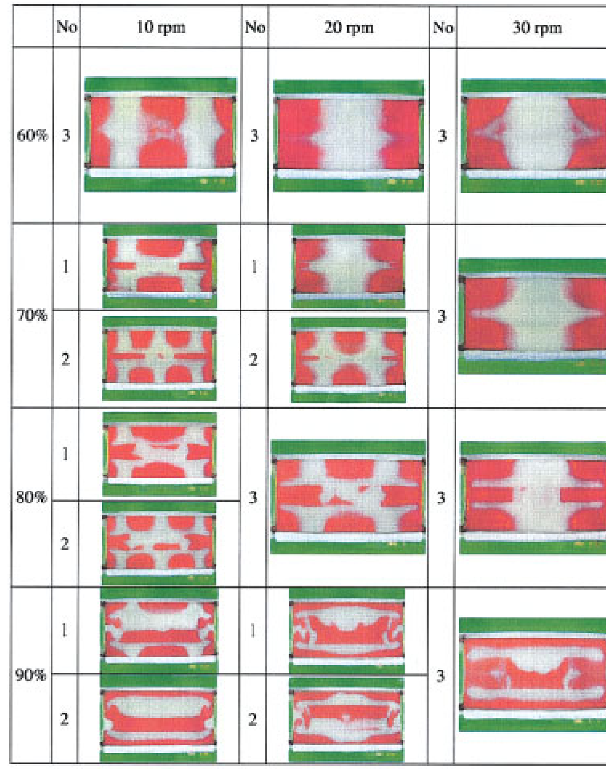

Figure 4. Segregation patterns observed at the plane of observation of different operating conditions in the long drum after a 2-h run for case D.

The numbers in the "No" column represent the observation times in three repeat runs. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com]

At 90% fill, either no or only very faint bands are formed. Previous studies show that the origin of the axial segregation is from the surface-flow layer.[6] At 90% fill, the ratio of the size of the surface-flow layer to the solid-body rotation region decreases[27] and thus the size of the surface-flow layer is restricted and the formation of the bands is suppressed. The core-in-core pattern shows more complicated geometry and the red core within the white core is symmetrical about the central plane and is not symmetrical about the rotation axis. According to the view of Barker and Mehta,[31] granular configurations are not subject to the constant jostling of Brownian motion, and therefore clusters of grains (that is, segregated region), once formed, are frozen in. The competition between the motion of these clusters and the motion of the independent grains is crucial for the segregation patterns. Therefore, the complicated geometric patterns observed at 90% fill suggest the competitions between the motion of these clusters (including the core, the core-in-core, and the bands) and independent particles.

In cases A, B, and D, the segregation patterns at 20 rpm and 90% fill show two different kinds. One pattern is exactly upside-down to the other pattern, implying a solid-body rotation of the red core within the white core. Currently, there is no theory that can satisfactorily explain such phenomena. We

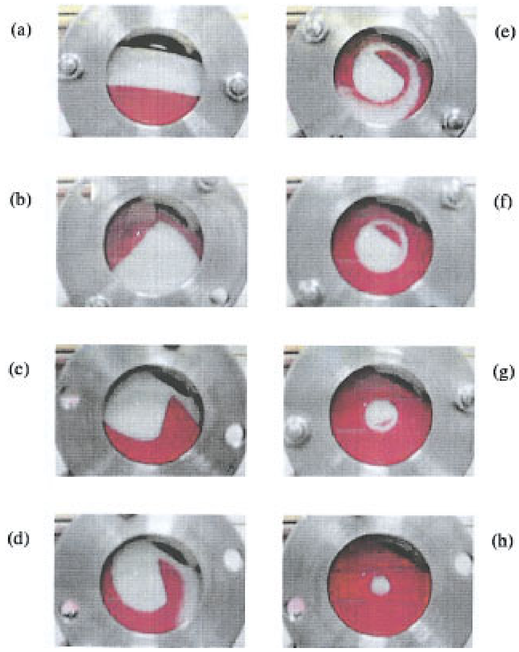

Figure 5. Formation of the core-in-core structure by the repeating wrapping process.

(a) Initial condition; (b) the red coarse beads avalanche down the surface-flow layer and wrap the white fine beads; (c) the white fine beads avalanche down the surface-flow layer and wrap the red coarse beads; (d and e) the repeating wrapping processes continue; (f) part of the red coarse beads are completely wrapped by the white fine beads, resulting in the core-in-core structure at the plane of observation; (g) the shrinking of the core-in-core structure; (h) the core-in-core structure gradually diminishing at the end view. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com]

propose a hypothesis: the solid-body rotating white core can be regarded as a virtual drum rotating within the granular bed. When the bands are formed within the virtual white drum, the red core within the white drum is blocked by the white bands at 70 and 80% fill in cases A and D, respectively, and segregation patterns observed at 80% fill, 20 rpm in case C. In the virtual drum, the two end bands are coarse particle rich red bands, consistent with previous reports.[6,7]

Conclusions

The segregation of the granular mixtures in a rotating drum has long been studied and many previous models have been proposed to analyze such a system. However, these models have been based on the segregated bands and cores observed at the bed surface and most of them discuss axial and radial segregation independently. Here we show the evidence that the simple band–core segregation structure (that is, the "cross" pattern) exists only at 60% fill levels. When the level of fill is equal to or greater than 70%, the segregation patterns within the bed are much more complicated, including the "cross" patterns, the "band-core-in-core" patterns, and other geometric patterns. The present work demonstrates conclusively that there are convective currents within the core that strongly affect segregation, even at large fills (whereas current dogma says that the core, once formed, is static) and even after a long transient period. The new segregation patterns are strong functions of the fill level and the rotational speed of the drum. The complicated patterns are related to, at least, the fill levels and the surface-flow layer. We qualitatively explain the formation of the core-in-core pattern and propose the virtual drum hypothesis. The challenge for the future will be to provide a fundamental understanding of the connection between the micro- and macrogranular dynamics to obtain improvements in granular processing.

Acknowledgments

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Literature Cited

1. Williams JC, Khan MI. The mixing and segregation of particulate solids of different particle size. *Chem Eng.* 1973;Jan:19-25.
2. Makse HA, Havlin S, King PR, Stanley HE. Spontaneous stratification in granular mixtures. *Nature.* 1997;386:379-382.
3. Baxter J, Tüzün U, Heyes D, Hayati I, Fredlund P. Stratification in poured granular heaps. *Nature.* 1998;391:136-136.
4. Burtally N, King PJ, Swift MR. Spontaneous air-driven separation in vertically vibrated fine granular mixtures. *Science.* 2002;295:1877-1879.
5. Rosato AD, Blackmore DL, Zhang N, Lan Y. A perspective on vibration-induced size segregation of granular materials. *Chem Eng Sci.* 2002;57:265-275.
6. Gupta SD, Khakhar DV, Bhatia SK. Axial segregation of particles in a horizontal rotating cylinder. *Chem Eng Sci.* 1991;46:1513-1525.
7. Kuo HP, Hsu RC, Hsiao YC. Investigation of axial segregation in a rotating drum. *Powder Technol.* 2005;153:196-203.
8. Zik O, Levine D, Lipson SG, Shtrikman S, Stavans J. Rotationally induced segregation of granular materials. *Phys Rev Lett.* 1994;73:644-647.
9. Hill KM, Caprihan A, Kakalios J. Axial segregation of granular media rotated in a drum mixer: Pattern evolution. *Phys Rev E.* 1997;56:4386-4393.
10. Choo K, Molteno TCA, Morris SW. Traveling granular segregation patterns in a long drum mixer. *Phys Rev E.* 1998;58:6115-6123.
11. Oyama Y. Report 5. *Bull Inst Phys Chem Res* (*Tokyo*). 1939;18:600. [Oyama's work was reported by Weidenbaum SS. In: Drew TB, Hoopes JW, eds. *Advances in Chemical Engineering*. Volume 2. New York, NY: Academic Press; 1958:211.]
12. Donald MB, Roseman B. Mixing and de-mixing of solid particles: Part I. Mechanisms in a horizontal drum mixer. *Br Chem Eng.* 1962;7:749-753.
13. Fan LT, Shin SH. Stochastic diffusion model of non-ideal mixing in a horizontal drum mixer. *Chem Eng Sci.* 1979;34:811-820.
14. Hill KM, Kakalios J. Reversible axial segregation of rotating granular media. *Phys Rev E.* 1995;52:4393-4400.
15. Ristow GH, Nakagawa M. Shape dynamics of interfacial front in rotating cylinders. *Phys Rev E.* 1999;59:2044-2048.
16. Khan ZH, Tokaruk WA, Morris SW. Oscillatory granular segregation in a long drum mixer. *Europhys Lett.* 2004;66:212-219.
17. Dury CM, Ristow GH. Radial segregation in a two-dimensional rotating drum. *J Phys I France.* 1997;7:737-745.
18. Khakhar DV, McCarthy JJ, Ottino JM. Radial segregation of granular materials in rotating cylinders. *Phys Fluids.* 1997;9:3600-3614.
19. Prigozhin L, Kalman H. Radial mixing and segregation of a binary mixture in a rotating drum: Model and experiment. *Phys Rev E.* 1998;57:2073-2080.
20. Khakhar DV, Orpe AV, Hajra SK. Segregation of granular materials in rotating cylinders. *Physica A.* 2003;318:129-136.
21. Hill KM, Caprihan A, Kakalios J. Bulk segregation in rotated granular material measured by magnetic resonance imaging. *Phys Rev Lett.* 1997;78:50-53.
22. Awazu A. Size segregation and convection of granular mixtures almost completely packed in a thin rotating box. *Phys Rev Lett.* 2000; 84:4585-4588.
23. Turner JL, Nakagawa M. Particle mixing in a nearly filled horizontal cylinder through phase inversion. *Powder Technol.* 2000; 113:119-123.
24. Wightman C, Mort PR, Muzzio FJ, Riman RE, Gleason EK. The structure of mixtures of particles generated by time-dependent flows. *Powder Technol.* 1995;84:231-240.

25. Shinbrot T, Zeggio M, Muzzio FJ. Computational approaches to granular segregation in tumbling blenders. *Powder Technol.* 2001;116:224-231.
26. Charles CRJ, Khan ZS, Morris SW. Pattern scaling in the axial segregation of granular materials in a rotating tube. *Phys Rev E.* (http://www.physics.utoronto.ca/~nonlin/papers_sand.html)
27. Parker DJ, Dijkstra AE, Martin TW, Seville JPK. Positron emission particle tracking studies of spherical particle motion in rotating drums. *Chem Eng Sci.* 1997;52:2011-2022.
28. Hill KM, Caprihan A, Kakalios J. Axial segregation of granular media rotated in a drum mixer: Pattern evolution. *Phys Rev E.* 1997;56:4386-4393.
29. Alexander A, Muzzio FJ, Shinbrot T. Effects of scale and inertia on granular banding segregation. *Gran Matt.* 2004;5:171-175.
30. Nakagawa M, Altobelli SA, Caprihan A, Fukushima E. NMRI study: axial migration of radially segregated core of granular mixtures in a horizontal rotating cylinder. *Chem Eng Sci.* 1997;52:4423-4428.
31. Barker GC, Mehta A. Size segregation mechanisms. *Nature.* 1993; 364:486-487.

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