Combined size and density segregation and mixing in noncircular tumblers

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Flowing granular materials segregate due to differences in particle size (driven by percolation) and density (driven by buoyancy). For noncircular tumblers the additional interaction between the segregation mechanisms and chaotic advection complicates the physics. Experiments are performed using a bidisperse mixture of equal volumes of different sizes of steel and glass beads in a quasi-two-dimensional square tumbler. Mixing is observed instead of segregation when the denser beads are larger than the lighter beads so that the ratio of particle sizes is greater than the ratio of particle densities. This can be expressed in terms of the particle diameters and masses as $d_{\mathrm{heavy}}/d_{\mathrm{light}} > (m_{\mathrm{heavy}}/m_{\mathrm{light}})^{1/4}$. Segregation patterns vary from a semicircular core when the fill level is below 50% to more complicated patterns including lobes and streaks for fill levels above 50%. Temporal evolution of segregated patterns is quantified in terms of a "segregation index" (based on the area of the segregated pattern) to capture both the rate and extent of segregation at different particle properties. The circular and noncircular tumblers have no significant difference in the segregation index, even though the segregation patterns differ significantly.

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I. INTRODUCTION

The current understanding of dry granular mixing is not sophisticated. Unlike fluids, the flow in a "mixer" does not necessarily lead to mixing. When flowing, granular materials often segregate due to differences in particle properties—size and density differences often being the key drivers. Examples of granular segregation exist in nature (avalanches, riverbeds, etc.) as well as industrial applications such as blenders and rotating kilns. Attempts to understand the flow, handling, and characterization of granular materials date at least half a century back [1–3]. Considerably more is known about granular flow thanks to efforts in both the physics and engineering communities over the last decade [4–7]. Despite this research, there are many practical questions that remain unanswered. (i) Can segregation be reduced or eliminated by controlling the particle properties? (ii) How does the shape of the tumbler affect granular segregation? (iii) How do the mechanisms for size segregation and density segregation interact with each other? (iv) How does chaotic advection due to a noncircular tumbler geometry interact with size and density segregation?

The focus of this work is to understand granular segregation for dry particles having different sizes and densities in a quasi-two-dimensional (quasi-2D) noncircular rotating tumbler that is partially filled with granular material and is rotated about its horizontal axis to produce a circulating flow of the granular material. The bulk of the material rotates as a solid body. However, the granular material flows along the sloped surface in a narrow surface layer of thickness δ due to gravity, as shown schematically in Fig. 1 for a square tumbler. The process of segregation takes place in this narrow surface layer due to size or density differences. For particles that differ in size only (called S systems), smaller particles snake through the interstices between the larger particles and deposit near the top of the fixed bed as a semicircular segregated core. This mechanism is called "percolation." A similar process occurs for particles that differ in density only (called D systems), except that in this case lighter particles rise to the surface of the flowing layer while heavier particles sink down and accumulate at the top of the fixed bed. Thus, "buoyancy" is the driving mechanism behind density segregation. A number of studies have been performed to understand size or density segregation alone in a rotating tumbler [8–13]. Thomas [14] considered how the relative concentration of beads of different sizes but the same density affects segregation. At a lower size ratio, geometrical effects (per-

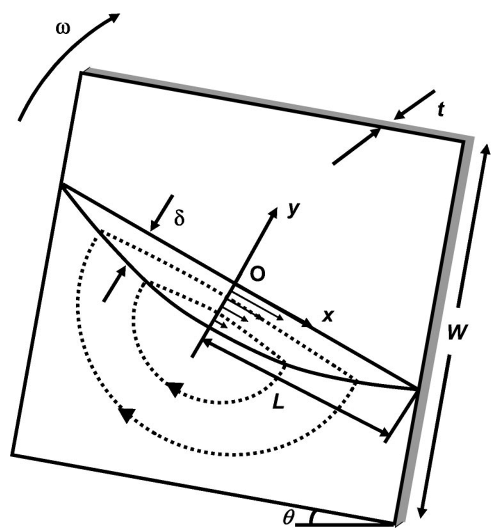

FIG. 1. Schematic of the square rotating tumbler geometry with the coordinate system and other parameters. Streamlines are sketched as dotted curves.

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colation) force the larger beads to the free surface. As a result, these beads are located close to the periphery of the tumbler when entrapped in the fixed bed. As the size ratio is increased, the mass of the larger beads becomes important and opposes these geometrical effects. At size ratio greater than 5, a segregation reversal takes place with larger beads sinking into the core while the smaller beads are located at the periphery of the tumbler.

A complicated situation occurs when the particles differ in both size and density (SD systems). A relatively large body of literature exists related to the study of the "Brazil nut effect," which is essentially percolation due to vertical vibration, named for the property of larger Brazil nuts to rise to the top of a container of mixed nuts upon vibration. Most Brazil nut studies were carried out with a single large particle in a sea of small particles, which is not pertinent here. However, a handful of studies considered bidisperse mixtures of particles [15,16]. Based on the competition between periodic percolation and condensation, Hong *et al.* [16] predicted that percolation (the Brazil nut effect) will occur when the ratio of large to small particle diameters exceeds the inverse of the ratio of large to small particle densities. In the opposite case, the "reverse Brazil nut effect" results in larger particles sinking to the bottom of a vibrated container.

Here we study the segregation of particles of different sizes and diameters in the flowing layer of a rotating tumbler. We use the Brazil nut effect as a comparison point, as it is the only possible point of reference. However, the physics of the Brazil nut case is very different from our case of rotating tumbler. In a rotating tumbler, the segregation occurs continuously due to constant dilation of a thin gravity-driven granular flowing layer rather than due to periodic dilation and condensation of an entire bed of particles with no net flow taking place for the case of the Brazil nut effect. In a SD system in a tumbler, one possibility is that smaller particles are also heavier than the larger particles. In this case percolation and buoyancy act together causing the smaller and heavier beads to drift to the bottom of the flowing layer to form a core region at the center of the fixed bed while the larger and lighter particles remain in the flowing layer to end up at the periphery of the fixed bed. However, our experimental work on circular tumblers showed that in some cases this leads to the formation of radial streaks instead of a semicircular core due to the greater "fluidity" of the smaller, heavier particles [17]. The streak formation was very sensitive to particle size and did not occur if the difference in particle size was either too large or too small.

The other possibility when granular materials differ in both size and density is that smaller particles are also lighter than the larger particles. In this situation, percolation and buoyancy oppose each other. Buoyancy drives the larger and *heavier* beads toward the core and smaller and *lighter* beads toward the periphery. Conversely, percolation drives the *smaller* and lighter beads toward the core and *larger* and heavier beads toward the periphery. This interplay between percolation and buoyancy can lead to mixing instead of segregation in a circular tumbler under certain conditions [17]. The segregation for steel and glass beads in a circular tumbler is reduced when the diameter of steel beads (d_s) is more than twice the diameter of glass beads (d_g). Mixing occurs with no segregation at all when the ratio of the heavy to light particle sizes is between 10 and 20. At particle size ratios of 1.5 and above, percolation dominates the granular segregation, while for particle size ratios less than 1.5, segregation is dominated by buoyancy. For particle size ratios between 1.5 and 2, percolation dominates buoyancy, but the segregation pattern is complicated. A band of glass beads forms near the periphery of the tumbler in addition to glass beads forming a relatively small core, with a band of steel beads between the core and the periphery of the tumbler.

Prior to our work, Drahun and Bridgwater [18], Alonso *et al.* [19], and Metcalfe and Shattuck [20] studied granular segregation involving particles of different sizes and densities in a rotating tumbler. The parameter space considered in our previous study was much broader than the studies of Drahun and Bridgwater, Alonso *et al.*, and Metcalfe and Shattuck. The size ratio ($d_{\text{heavy}}/d_{\text{light}}$) and mass ratio ($m_{\text{heavy}}/m_{\text{light}}$) of heavier to lighter beads was varied up to 20 and 24 000, respectively, in our study compared to 4 and 190, respectively, in the other three studies [21]. Furthermore, these studies focused primarily on finding the combination of particle size and density at which segregation is eliminated. They did not investigate the variety of segregation patterns that can appear including streaks and bands of heavier beads between lighter beads [17]. In spite of the differences in types of granular flow in these studies (avalanching vs rolling regime, free surface flow vs flow in a tumbler) and differences in methods to measure segregation (invasive sampling, magnetic resonance imaging, digital imaging), there is one surprising commonality. It appears that segregation is reduced when the size ratio of heavier to lighter beads exceeds the density ratio of heavier to lighter beads [17].

Nearly all of the studies on granular segregation in tumblers have been done using tumblers with a circular cross section. More recently, there has been both experimental and computational work that deals with two-dimensional and three-dimensional tumblers of noncircular cross sections using particles that differed either in size but not density or in density but not size [22–24]. The segregation mechanism in such a tumbler is quite complex, since the underlying flow is chaotic. For both circular and noncircular tumblers streamlines are concentric semicircles in the fixed bed (because it rotates as a solid body) and nearly straight lines parallel to the free surface in the flowing layer as sketched in Fig. 1. If streamline plots obtained at different time instants are overlaid, the streamlines superimpose for a circular tumbler. However, for a noncircular tumbler, the flowing layer changes thickness and length depending on the orientation of the tumbler. This results in streamline crossing in the flowing layer [24]. It has been shown that streamline crossing leads to chaotic advection [25]. Hill *et al.* [23] described the interplay between chaotic advection and segregation for the case of granular materials in circular and noncircular geometries using particles that differed only in either density or size. They observed that in a noncircular tumbler geometry (square or ellipse), the segregation patterns differ substantially from that in a circular geometry [23]. In addition, the segregation patterns also change dramatically with the fill level for noncircular tumblers. For fill levels below 50%, the segregation pattern resembles the "classical" semicircular core pattern (observed for circular tumblers). However, for fill levels of 50% and above, the segregation pattern differs significantly from the "classical" semicircular core, having radial streaks or lobes.

It is important to place the granular system and the underlying flow in the tumbler in the context of recent studies. Studies on either *S* or *D* systems have been conducted for steady flows [10] and in time-periodic flows: periodic forcing caused either by geometry of the tumbler [23] or by the angular velocity of the tumbler [26]. All of these studies show that the granular systems segregate in a way that clearly mimics the underlying structure of the flow. Studies on SD systems have only been conducted in circular tumblers where the flow is steady. The central objective of the current work is to explore how time-periodic flows (chaotic advection) in a noncircular geometry interact with mixing and segregation of particles that differ in both size and density and compare the outcome with our previous results on a circular tumbler. Prior work on *S* and *D* systems suggests that in a noncircular tumbler the competition between chaos and segregation leads to a wide variety of segregated patterns [23]. For SD systems, there is an additional competition between the mechanisms driving segregation (percolation vs buoyancy) and the interaction between chaos and segregation.

II. EXPERIMENTAL PROCEDURE

The square tumbler used in the quasi-2D experiments is shown schematically in Fig. 1. Although the square tumbler offers complexity in terms of chaotic advection (the key motivation for this study), it is also a simple geometry. A square tumbler is a rotationally symmetric mixer and therefore, has the simplification that for a half-filled tumbler the center of the flowing layer coincides with the center of the tumbler. As a result of this simplification, there is no vertical motion of the flowing layer for different orientations of the tumbler, and the shape of the flowing layer is symmetric with respect to its midpoint at low rotational speeds, as shown in Fig. 1.

The flowing layer is key to the flow and segregation of granular materials [27]. In order to have an accurate comparison between the segregation results for a circular tumbler [17] and a square tumbler, it is important that the length of flowing layer in the circular tumbler be equal to the time-average length of the flowing layer in the square tumbler. Based on geometry, the average length of the flowing layer ($2L$) for a square tumbler of side (W) assuming a 50% fill level is

$$2L = \frac{W\int_0^{\pi/4} \sec\theta\, d\theta}{\pi/4} = 1.12W \qquad (1)$$

where θ is the angle that the side of square tumbler makes with the horizontal. In our previous work, we used a 28 cm diameter circular tumbler [17]. In order to match the condition in Eq. (1), the length of the square tumbler (W) is 25 cm. The front faceplate of the tumbler is clear acrylic to permit optical access. The back surface of the tumbler is also made of clear acrylic to ensure similar boundary conditions on both sides of the tumbler. A stepper motor and micro series driver combination (SLO-SYN®) are used to rotate the tumbler at a rotational speed (ω) of 0.21 rad/s (2 rpm), corresponding to a Froude number $\mathrm{Fr}=\omega^2 L/g$ of about 6.3×10^{-4}, where L is the average half length of the flowing layer. A limited number of experiments are performed at rotational speeds of 0.105 rad/s (1 rpm), 0.42 rad/s (4 rpm), 0.84 rad/s (8 rpm), and 1.68 rad/s (16 rpm) to determine the sensitivity of granular segregation to rotational speed.

Similar to our previous work [17], spherical chrome steel beads of density 7.5 g/cm^3 with diameters (d_s) ranging from 1 to 4 mm and glass beads of density 2.5 g/cm^3 with diameters (d_g) ranging from 0.2 to 4 mm are used for experiments. The ratio of the length of the tumbler side (W) to bead diameter ranges from 62.5 to 1250. In all experiments, the dimensionless axial length of the tumbler, t, is set to 3.2 times the diameter of the larger beads in order to maintain similarity with respect to the particle diameter. Choosing the appropriate tumbler thickness when studying granular segregation is a bit of a balancing act. A thickness that is very small can lead to end wall effects, while too large of a thickness can lead to axial segregation [22,28].

Experiments are conducted using a 50-50 volume fraction of steel and glass beads. Beads are initially well mixed. Images are obtained with a TSI® charge-coupled device camera for the initial condition and every 1/8 rotation thereafter to allow quantitative image analysis. Twenty different experiments are performed using various combinations of steel and glass beads. In four cases, the sizes of the steel and glass beads are identical so segregation is driven by density alone. In six cases, the steel beads are smaller than the glass beads ($0.25 \leqslant d_s/d_g \leqslant 0.75$), so that percolation based on size (S) and buoyancy based on density (D) act in the same direction, denoted "$S+D$." For the remaining ten cases, the steel beads are larger than the glass beads ($1.33 \leqslant d_s/d_g \leqslant 20$), denoted "$S-D$," since the percolation and buoyancy oppose one another. After each experiment steel and glass beads are separated using a magnet. During this process, steel beads acquire a small residual magnetism and tend to be attracted to other steel beads. Therefore, after separating the beads, the steel beads are demagnetized.

III. RESULTS

First, consider the results for the D system when the steel beads (darker areas) and glass beads are of the same size, shown in Fig. 2. Granular segregation is fast and similar in both a circular tumbler and a square tumbler—heavier steel beads accumulate in the center of the bed, while the lighter glass beads segregate toward the periphery. However, time-dependent changes in the flowing layer length and thickness result in streamline crossing, which in turn causes the segregation patterns in a square tumbler to differ significantly from those of a circular tumbler, consistent with previous results in a square tumbler for S and D systems [23]. At a fill level of 50%, the segregation pattern in the square tumbler has "lobes" with two high concentration regions of steel

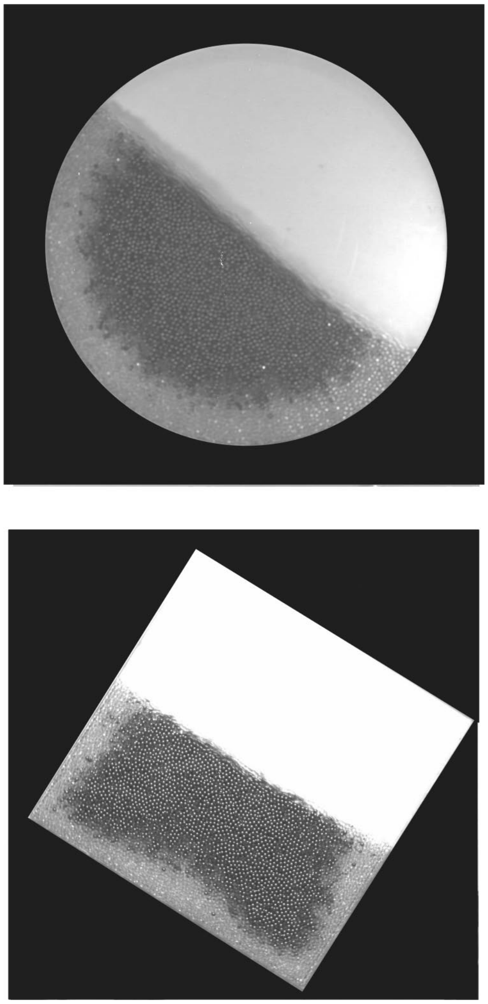

FIG. 2. Granular segregation in half-filled circular and square tumblers for 3 mm glass and steel beads (D system) resulting in "classical" core formation for the circular tumbler and lobes for the square tumbler. Experiments are performed at 2 rpm and images are taken after 20 rotations.

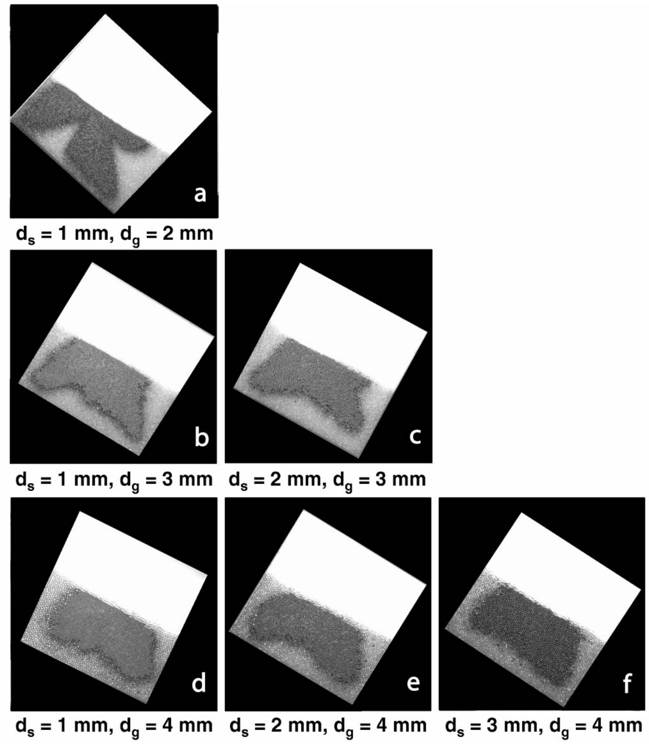

FIG. 3. Segregation patterns when buoyancy and percolation reinforce each other ($S+D$) in a half-filled square tumbler. (a)–(c) for 1 mm steel beads and 2, 3, and 4 mm glass beads, respectively (d_s/d_g=0.5, 0.33, 0.25); (d), (e) for 2 mm steel beads and 3 and 4 mm glass beads, respectively (d_s/d_g=0.66, 0.5); and (f) 3 mm steel beads and 4 mm glass beads (d_s/d_g=0.75). Experiment is performed at 2 rpm and images are taken after 20 rotations.

beads nearly reaching the corners of the square. The lobed structure is a bit difficult to see in Fig. 2. However, note that the band of glass beads at the periphery is thicker at the center of the base of the particle bed than near the corners. Likewise the glass bead layer is thicker at the top of the sides of the bed. These are characteristics typical of the lobed structure often seen in square tumblers [23]. Hill *et al.* [29] suggested that the lobed structure is related to the location of regular regions (Kolmogorov-Arnold-Moser islands or elliptical points). For a 50% full square tumbler, these regular regions are located midway between the center and the corners of the tumbler. Heavier particles that *sink* or smaller particles that *percolate* to the regular regions are likely to be mapped back to the same position after each rotation. These regions continue to grow with time into lobe-shaped patterns because both the original particles in these regions are mapped to the same position and the new particles from the flowing layer sink or percolate into the regular regions.

Next, consider the case when both the size and the density of particles are different with the steel beads smaller than the glass beads, an $S+D$ system. Figures 3(a)–3(c) show the segregation pattern after 20 revolutions for 1 mm steel beads and 2, 3, and 4 mm glass beads at a fill level of 50%. Once again, granular segregation is fast with both buoyancy and percolation forcing the steel beads toward the center of the tumbler forming a lobe pattern. The lobes are narrower and more distinct for the case of smaller glass beads ($d_s/d_g=1/2$ or $1/3$) than for the larger glass beads ($d_s/d_g=1/4$). This is consistent with previous results for $S+D$ systems in circular tumblers where thin radial streaks occur for 1 mm steel beads with 2 and 3 mm glass beads ($d_s/d_g=1/2$ or $1/3$) [17]. These streaks formed as a consequence of the higher fluidity of the smaller steel beads compared to the larger glass beads. For the case of 1 mm steel and 2 mm glass beads, the steel beads flowed at a speed more than twice that of the glass beads [17]. As the size of glass beads was increased to 4 mm, the segregation pattern reverted back to the classical core because of the strong percolation flux counteracting the fluidity. Similar reasoning can explain the results for square tumbler shown in Figs. 3(a)–3(c). For smaller sized glass beads [Figs. 3(a) and 3(b)], the fluidity of the steel beads in the flowing layer allows most of them to reach the downstream side of the tumbler before percolating through the interstices between the glass beads resulting in narrow lobes. For 4 mm glass beads [Fig. 3(c)], the steel beads percolate down through the flowing layer more quickly, counteracting the effect of the fluidity of the steel

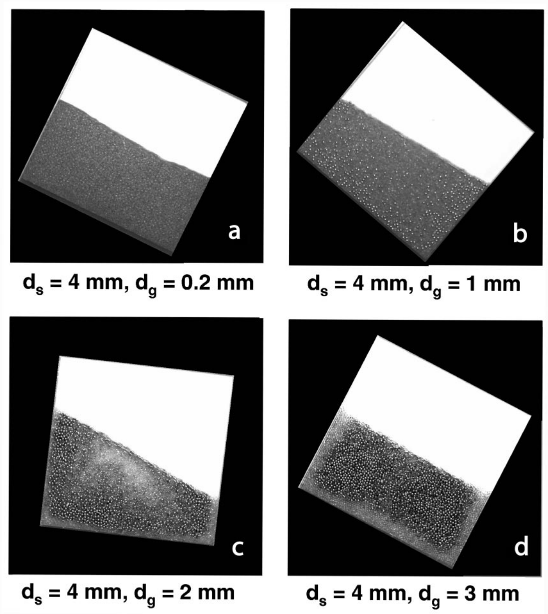

FIG. 4. Segregation patterns when buoyancy and percolation oppose each other ($S-D$) in a half-filled square tumbler. 4 mm steel beads tumbled with (a) 0.2 mm glass beads ($d_s/d_g=20$)—no segregation; (b) 1 mm glass beads ($d_s/d_g=4$)—band of high concentration steel beads at the tumbler periphery; (c) 2 mm glass beads ($d_s/d_g=2$)—small lobes of glass beads, a band of steel beads, and glass beads at the periphery; and (d) 3 mm glass beads ($d_s/d_g=1.33$)—core of steel beads with a slight tendency toward lobes. Experiments are performed at 2 rpm and images are taken after 20 rotations.

beads, thereby reducing the sharpness of the lobes.

Similar segregation patterns also occur for steel beads greater than 1 mm in diameter tumbled with glass beads of a larger size. Figures 3(d) and 3(e) show the segregation pattern after 20 rotations for 2 mm steel with 3 and 4 mm glass beads. Figure 3(f) shows the segregation pattern for 3 mm steel beads and 4 mm glass beads. Given the results in Fig. 2 for a D system, it seems that the primary reason behind lobe formation is the geometry of the tumbler and the resulting advection in that geometry as described by Hill *et al.* [29]. This advection results in formation of regular and chaotic regions regardless of any segregation mechanisms. However, the results in Fig. 3 indicate that particle properties and the resulting segregation mechanisms play a role in lobe formation as they interact with the chaotic advection and regular regions to change the segregated structure. For cases, where fluidity dominates percolation, smaller steel beads outrun the glass beads in the flowing layer and are therefore located further away from the center of the tumbler. This causes the lobes to be long and thin. However, when percolation dominates over fluidity, the steel beads in the flowing layer sink to the bed before reaching the periphery of the tumbler causing the lobes to be short and thick.

Finally, consider the case when the steel beads are larger than the glass beads ($S-D$ systems), so that percolation and buoyancy oppose each other. Figure 4 shows different segregation patterns after 20 rotations for the case of 4 mm steel beads tumbled with 0.2, 1, 2, and 3 mm glass beads. No segregation is evident in Fig. 4(a) for 0.2 mm glass beads and 4 mm steel beads ($d_s/d_g=20$) suggesting that percolation and buoyancy offset each other for this particle size combination, similar to the results for a circular tumbler [17]. Increasing the size of the glass beads to 1 mm introduces a tendency toward segregation into the system, as shown in Fig. 4(b). For this situation, percolation is more powerful than buoyancy resulting in a higher concentration of larger (steel) beads close to the periphery of the tumbler.

Increasing the size of glass beads to 2 mm results in three distinct segregation regions, shown in Fig. 4(c): a central area with small glass beads tending toward a lobe pattern, small glass beads near the walls of the tumbler, and a middle band composed primarily of large steel beads between the glass beads in the lobed core and the glass beads at the walls. Once again percolation dominates over buoyancy, evident in the small glass beads present near the center of the bed. However, the percolation mechanism is weaker relative to the buoyancy mechanism in this case than for the smaller glass beads, shown in Fig. 4(b). As a result, fewer glass beads segregate in the central portion of the bed. Buoyancy results in some of the less dense glass beads staying near the walls of the tumbler, so that the steel beads are pushed inward from the walls.

Further increasing the size of glass beads to 3 mm in diameter changes the dominant segregation mechanism from percolation to buoyancy. The central portion of the bed is mostly steel beads due to density segregation, while the glass beads are located at the periphery of the tumbler, as shown in Fig. 4(d). Although, the central, slightly lobed region is predominantly composed of steel beads, some glass beads are mixed in as a result of the relatively weak effect of percolation. Thus, the segregation is not as complete as for density segregation for equal sized glass and steel beads (Fig. 2).

The results for other sizes of steel beads where buoyancy and percolation oppose each other are similar to results shown in Fig. 4. No segregation is observed for 0.2 mm glass beads tumbled with 2 or 3 mm steel beads ($d_s/d_g=10,15$), similar to Fig. 4(a). A segregation pattern similar to that shown in Fig. 4(b) is observed for 0.2 mm glass beads with 1 mm steel beads and 1 mm glass beads with 3 mm steel beads ($d_s/d_g=3,5$). Finally, for 1 mm glass beads with 2 mm steel beads and 2 mm glass beads with 3 mm steel beads, the segregation pattern resembles Fig. 4(c) with glass beads close to the center and at the periphery of the tumbler, while a band of steel beads is present between the two regions of glass beads ($d_s/d_g=3/2,2$). Similar results occur in a circular tumbler except that the shape of the segregated core is semicircular instead of having lobes [17].

The segregation patterns in a square tumbler when percolation and buoyancy oppose each other can be categorized based on the size ratio of steel beads and glass beads. No segregation is observed when $10 \leqslant d_s/d_g \leqslant 20$. Reducing the particle size ratio to between 3 and 5 introduces segregation in the system with a dilute band of steel beads near the periphery of the tumbler. For even smaller differences in particle size ($3/2 \leqslant d_s/d_g \leqslant 2$), glass beads are present near the center of the tumbler as well as close to the periphery with a band of steel beads in between. At even smaller particle size ratios ($1 < d_s/d_g \leqslant 4/3$), buoyancy becomes the dominant segregation mechanism so that a core of steel beads forms. Except for the differences in the shapes of segregation patterns, categorizing the segregation based on d_s/d_g for $S-D$ systems is very similar to previous results for a circular tumbler [17]. Even the particle size ratios at which these transitions occur are the same for the circular tumbler and the square tumbler. This indicates that while the shape of the tumbler influences the shape of the segregation pattern, particle properties play the key role in controlling the extent of segregation.

We now return briefly to the prediction for the Brazil nut problem that percolation dominates when the ratio of large to small particle diameters exceeds the inverse of the ratio of larger to small particle densities. Applying this rule, which is based on vertical vibration, to a SD system in a rotating drum correctly predicts the nature of the segregation in only five out of the 16 cases for which it applies (in four cases it cannot be used, since the bead diameters are identical). All but one of the cases for which it works are situations in which percolation and buoyancy act together, although the rule even fails here in one case. The prediction does not work well for situations where percolation and buoyancy oppose one another. Clearly, this is a consequence of the Brazil nut rule being based on periodic dilation and condensation during vibration, whereas segregation in a tumbler results from the continuously dilated flowing layer.

Previous results have shown that granular segregation for both S systems and D systems in a square tumbler is sensitive to the fill level [23]. Figure 5 shows the effect of the fill level on the segregation patterns for two cases—1 mm steel and 2 mm glass beads ($S+D$ system) and 3 mm steel and 2 mm glass beads ($S-D$ system). At low fill levels, the $S+D$ system shows a tendency toward streak formation. These streaks of steel beads related to the higher fluidity of the steel beads [17] are incomplete compared to greater fill levels and do not reach the periphery of the tumbler. On the other hand, the $S-D$ system has a core of glass beads with glass beads also at the periphery of the tumbler, while the steel beads are between the core and the periphery.

The segregation pattern in a square tumbler have also been shown to be extremely sensitive to the fill level around a fill level of 50% for S and D systems [23]. At a fill level of 50%, both the $S+D$ and the $S-D$ systems have a lobed core, although it is somewhat difficult to see for the $S-D$ system in Fig. 5. As the fill level of the $S+D$ system is increased from 50% to 52%, the segregation pattern changes from the lobe pattern to radial streaks. Radial streaks also were observed for the same particle size combination in a circular tumbler at fill levels ranging from 40% to 60% [17]. A similar increase in the fill level from 50% to 52% does not result in radial streaks for the $S-D$ system. For the $S+D$ system, radial streaks continue to occur at a fill level of 60% although they are thicker than at a fill level of 52%. The $S-D$ system shows a stronger lobe pattern of the core at a fill level of 60% with two additional and symmetrical lobes above the center of the tumbler and pointing toward the other corners of the square. These additional lobes are difficult to discern in Fig. 5, but are readily evident as the tumbler operates. Like the lower fill levels, glass beads are present in the center of the tumbler as well as near the periphery, while

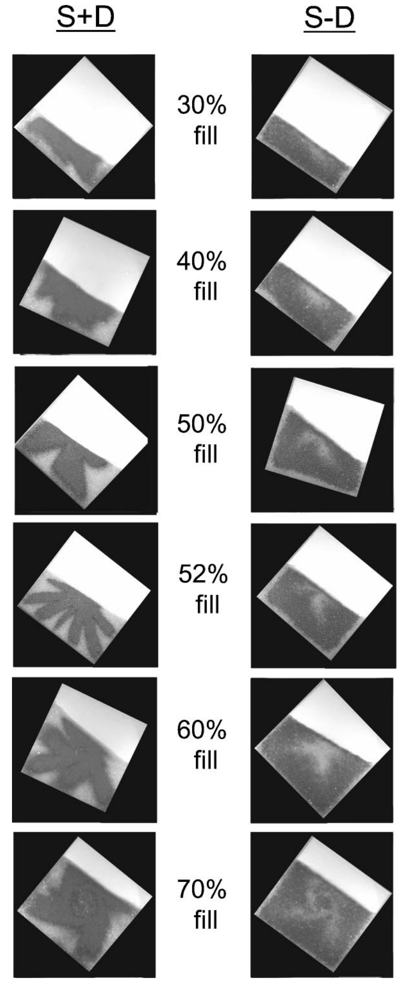

FIG. 5. Effect of fill level on granular segregation. Segregation patterns in the left column are for 1 mm steel and 2 mm glass beads ($S+D$ system, $d_s/d_g=0.5$) while segregation patterns in the right column are for 3 mm steel and 2 mm glass beads ($S-D$ system, $d_s/d_g=1.5$). Experiments are performed at 2 rpm and images are taken after 20 rotations.

the steel beads are present in between these two regions. As the fill level is increased even further to 70%, both the $S+D$ and the $S-D$ systems show a similar shape of the core pattern, although it is not as distinct for the $S-D$ system. Radial streaks disappear for the $S+D$ system, and in both the cases the segregation pattern has four arms pointing to the four corners of the granular bed. The $S-D$ system continues to have glass beads in the core as well as near the periphery, while $S+D$ system has steel beads in the core and glass beads towards the periphery. Note that the unmixed core region at the center of rotation for both systems is a rounded square, as would be expected in a monodisperse system [30]. This region never enters the flowing layer and, hence, remains unmixed.

All of the experiments presented thus far are performed at an angular velocity of 2 rpm. However, previous research for S systems indicates that the rate of segregation depends on the rotational speed of the tumbler [11]. This suggests that the angular velocity of the rotating tumbler may alter the nature of the segregation in the SD systems considered here.

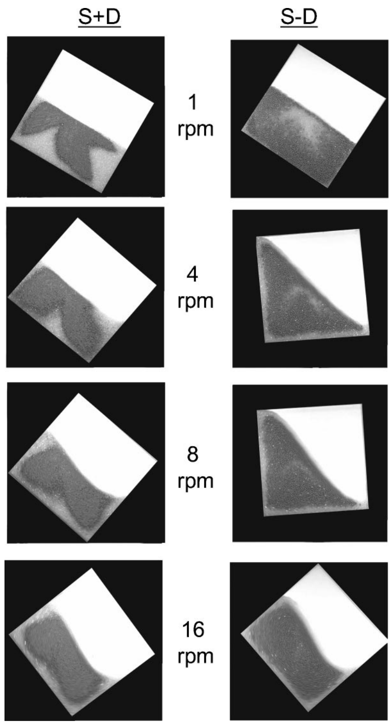

FIG. 6. Effect of angular velocity on granular segregation. Segregation patterns in the left column are for 1 mm steel and 2 mm glass beads ($S+D$ system, $d_s/d_g=0.5$) while segregation patterns in the right column are for 3 mm steel and 2 mm glass beads ($S-D$ system, $d_s/d_g=1.5$). Experiments are performed for half-filled tumbler and images are taken after 20 rotations. The angle of the tumbler with respect to horizontal varies somewhat to best display the segregation pattern.

To test this, experiments are performed using 1 mm steel and 2 mm glass beads ($S+D$ system) and 3 mm steel and 2 mm glass beads ($S-D$ system) at angular velocities ranging from 1 to 16 rpm ($1.58\times10^{-4}\leqslant \mathrm{Fr}\leqslant 4.03\times10^{-2}$). The results are shown in Fig. 6. For the $S+D$ system, distinct lobes are observed for 1 rpm (Fig. 6) and 2 rpm (Fig. 3). As the angular velocity is increased from 2 to 8 rpm, the lobes become wider and increase in area. At an angular velocity of 16 rpm, the steel beads are mixed with glass beads in a large core with a thin band of glass beads near the periphery of the tumbler. The dependence of the lobed structure on the angular velocity is likely related to the "fluidity" of the smaller sized steel beads [17] as well as the length of flowing layer, which changes with tumbler orientation. At lower angular velocities, the smaller beads (steel for the $S+D$ system) outrun the larger glass beads to reach the downstream portion of the flowing layer. Of course, this process is more noticeable when the flowing layer is longest (flowing layer extends between the corners of the tumbler), thus resulting in distinct lobes. At higher angular velocities, the velocity of all beads in the flowing layer is higher, so the steel beads are less likely to outrun the glass beads, thus reducing the tendency toward a lobed structure and forming a mixed core of steel and glass beads. The segregation pattern at 1 rpm in the $S-D$ system consists of small lobes of glass beads surrounded by steel beads, with glass beads at the corners of the tumbler. As the angular velocity increases from 1 to 4 rpm, the lobes of glass beads become less distinct in the center, and a band of glass beads forms at the periphery of the tumbler with steel beads between the lobed core and the periphery. As the angular velocity is increased to 4 and 8 rpm, the area occupied by glass beads in the center decreases while no changes are evident in the area occupied by the glass beads by the periphery of the tumbler. At an angular velocity of 16 rpm, the lobed core disappears and the glass beads are mixed with the steel beads except at the periphery, resulting in a situation that is similar to that for the $S+D$ system at 16 rpm. In this case, the dependence of the segregation pattern on angular velocity is likely related to the square tumbler and the velocity of the flowing layer. We speculate that at low angular velocities, the small glass beads have enough time to percolate through the flowing layer to form a small core of glass beads. The tendency toward a lobed structure is related to the shape of the tumbler [23]. At high angular velocities, all of the beads are moving faster in the flowing layer, so the small glass beads are less likely to reach the bottom of the flowing layer to form a core, and some glass beads even float to the top of the flowing layer to form the band of glass beads at the periphery.

To quantify the time development of the segregation patterns, digital images of the quasi-2D tumbler are obtained after every 1/8 rotation for each of the SD systems. As is evident in the previous images, darker areas correspond to regions where steel beads are plentiful, and lighter areas correspond to regions where glass beads are plentiful. The degree of segregation measured as a function of area occupied by the segregated pattern can be quantified as [17]

$$\sigma=\frac{[A(W^2/2-A)]^{1/2}}{W^2/2} \qquad (2)$$

for a half-full tumbler, where A is the area occupied by one bead type and $W^2/2-A$ is the remaining area occupied by the other bead type. Thus, the segregation index is the geometric mean of the area occupied by the steel and glass beads normalized by the total area for a 50% fill level. For perfect segregation, the core area *(A)* should be one-quarter of the area of the tumbler resulting in a segregation index (σ) of 0.5. Conversely, a core area that is too small or too large compared to theoretical area for maximum segregation would suggest better mixing and poorer segregation. As an example, a core that occupies 5% or 95% of the total filled area will have a segregation index of $\sigma=0.22$. For no segregation when core occupies 0% or 100% of the total filled area, $\sigma=0$.

Figure 7(a) shows the dependence of the segregation index on the number of rotations for 1 mm steel beads and all sizes of glass beads (D systems, 1 mm; $S+D$ systems, 2, 3, and 4 mm). The segregation is complete within the first few rotations and remains constant after the initial transient. Segregation is faster and reaches a slightly higher level for the combination of 1 mm steel beads and 2 mm glass beads where differences in particle fluidity causes the lobes to be thin and distinct. The small oscillatory variation of the segregation index comes about as the individual lobes enter and

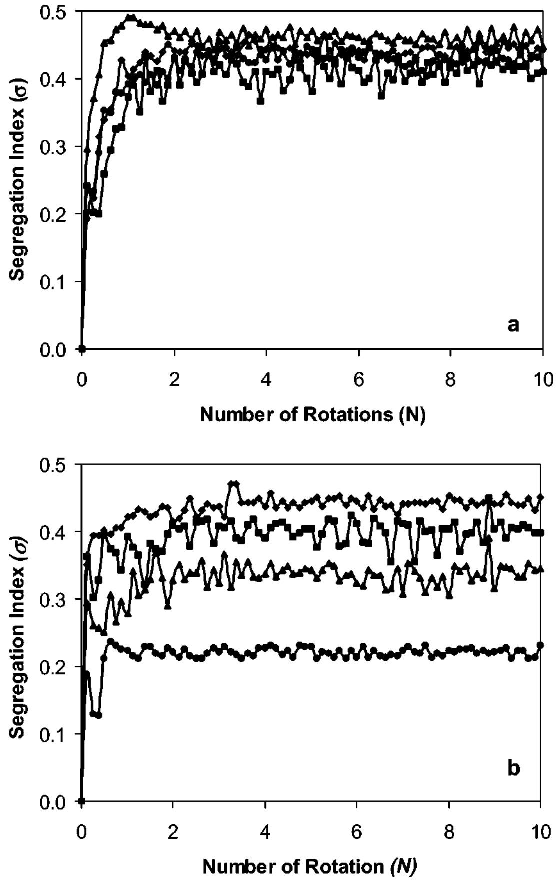

FIG. 7. Segregation index (σ) as a function of the number of rotations (N). (a) 1 mm steel beads tumbled with 1, 2, 3, and 4 mm glass beads (d_s/d_g=1.0, 0.5, 0.33, 0.25); (b) 4 mm steel beads tumbled with 1, 2, 3, and 4 mm glass beads (d_s/d_g=4, 2, 1.33, 1.0). d_g=●1, ▲ 2, ■ 3, ◆ 4 mm.

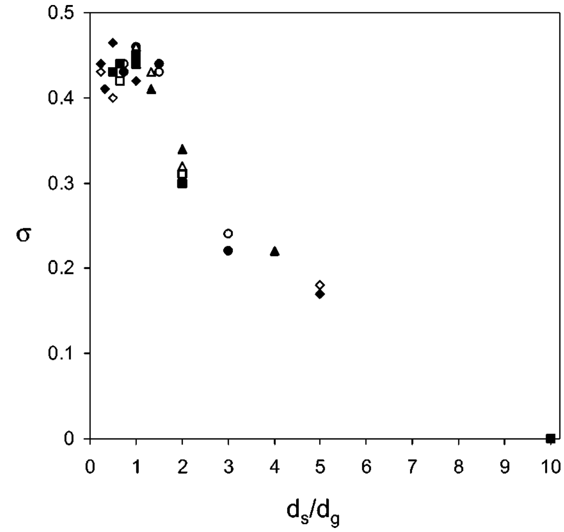

FIG. 8. Steady state segregation index as a function of the ratio of steel beads to glass beads (d_s/d_g) for both circular and square tumblers. d_s= ◆ 1, ■ 2, ● 3, ▲ 4 mm. Open symbols, circular tumbler; black symbols, square tumbler.

leave the flowing layer. Segregation is nearly as strong for other sizes of glass beads, as indicated by the values of σ near the maximum of 0.5 for perfect segregation. The segregation is much more dependent on bead size for $S-D$ systems. Figure 7(b) shows the dependence of the segregation index (σ) on the number of rotations (N) for 4 mm steel beads and all sizes of glass beads ($S-D$ systems, 1, 2, and 3 mm; D systems, 4 mm). Again, segregation occurs quite quickly and remains nearly constant after the first few rotations. The segregation index is highest for 4 mm glass beads (D system) and decreases as the size of glass beads decreases due to the increased counteraction of buoyancy and percolation. For the case of 4 mm steel beads and 0.2 mm glass beads [not shown in Fig. 7(b)], σ=0 indicating completely mixed beads.

The steady state segregation index depends on the size ratio d_s/d_g as shown in Fig. 8 for all particle combinations and results for both square and circular tumblers [17]. Independent of the shape of the tumbler, segregation is strong when $d_s/d_g \leqslant 1.5$. As the size of steel beads is increased to twice the size of glass beads or larger ($d_s/d_g \geqslant 2$), the segregation index decreases to a level of $\sigma \approx 0$ at $d_s/d_g \geqslant 10$, indicating complete mixing. Since the steady state segregation index is similar for circular and square tumblers, one might argue that the *interplay between buoyancy and percolation on segregation is largely unaffected by the complexity introduced by the noncircular tumbler*. This result is surprising in light of measurements comparing the mixing efficiency of square and circular tumblers using particles of two different colors but same size and density [24]. In that case, the initial condition corresponded to perfect separation with either the left half of the tumbler consisting of one particle type and right half of the tumbler consisting of other particle type or with the two particle types layered on top of each other. For both of these initial conditions, mixing of monodisperse particles was faster in a square tumbler than a circular tumbler. However, the difference in speed of mixing for square and circular tumblers half filled with monodisperse particles was a consequence of chaotic advection in the square tumbler. The results for SD systems, on the other hand, also involve differences in particle properties. Thus, it appears that while chaotic advection modifies the shape of segregation pattern between square and circular tumblers, the *extent of segregation* is predominantly controlled by particle properties. Furthermore, the rate of segregation is essentially independent of the shape of the tumbler, as is evident by comparing the transient in Fig. 7 to analogous results for a circular tumbler [17]. Regardless of the shape of the tumbler, steady state segregation is achieved within 2–3 rotations. It is possible that there is a small difference in the *rate of segregation* between the circular and square tumblers that cannot be detected. Within just a few rotations it is difficult to ascertain if the mixing or segregation is faster in a square tumbler than a circular tumbler, or vice versa. Furthermore, comparing the mixing efficiency of several uniformly-convex 2D geometries for avalanching flow, square tumblers are only marginally more efficient than circular tumblers to achieve a mixed state [30]. While the physics for avalanching flow is different from that for the continuously flowing layer studied here, their reported small differences in mixing efficiency could have been related only to the difference in the time-averaged

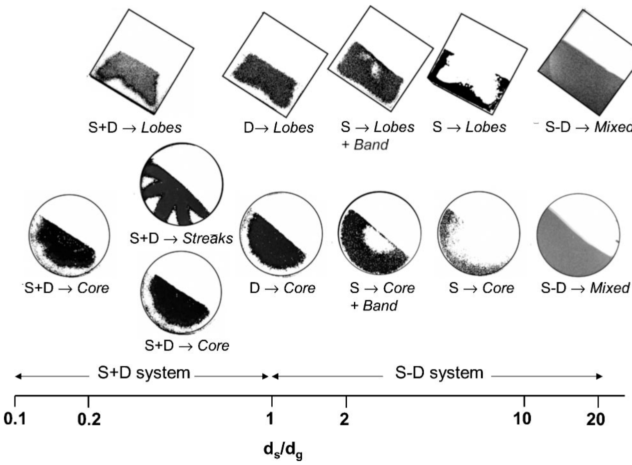

FIG. 9. Summary of segregation regimes for different tumbler shape and different size ratios (d_s/d_g). Patterns are for half-filled tumblers and dark areas represent the regions where the concentration of steel beads is significantly higher than concentration of glass beads. Symbol below each segregation pattern represents the dominant segregation mechanism, S (percolation or size); D (buoyancy or density); $S+D$ or $S-D$ (percolation and buoyancy); and the final pattern shape (in italics), no segregation *(Mixed)*, a core or lobe of glass or steel beads *(Core/Lobe)*, streaks of steel beads *(Streaks)*, and a core or lobe of glass beads with a band of glass beads at the periphery *(Core/Lobe+Band)*.

flowing layer length in tumblers having different shapes but the same area, not the complexity of the system.

IV. CONCLUSIONS

This work extends our previous study of SD systems in circular tumblers and provides insight into mixing and segregation of granular particles when buoyancy, percolation, fluidity, and chaotic advection interact with each other. The results show that segregation patterns change dramatically with changes in the particle properties. Figure 9 shows the rich variety of patterns for different particle sizes and tumbler shapes. When percolation and buoyancy support each other $(d_s/d_g<1)$, the segregation pattern in a circular tumbler is usually a semicircular core, although for some particle combinations, the classical core is replaced by radial streaks. For half-full square tumblers, the characteristic core shape is lobed instead of a semicircular core. For certain particle size combinations, radial streaks are observed in a square tumbler but unlike circular tumbler these streaks are only formed at fill levels greater than 50%. Segregation is reduced when percolation and buoyancy oppose each other $(d_s/d_g>1)$. While the shape of the segregated pattern is different for circular and square tumblers, a key similarity between these two tumblers is the preferential location of steel or glass beads at various particle size ratios. For $1\leq d_s/d_g\leq 1.33$, steel beads are located in the center for both the tumblers. For $1.5\leq d_s/d_g\leq 2$, steel beads are located in between glass beads at the center and periphery of the tumbler. As the size ratio is increased to greater than 2, steel beads are preferentially located near the periphery of tumbler. Beyond a size ratio of 5, steel beads and glass beads mix throughout the tumbler. Thus, the change in the shape of the tumbler does not shift the key threshold points at which the nature of the segregation changes.

Granular segregation is of immense practical importance and is encountered in a variety of industries including cement, pharmaceutical, metallurgical, polymer, and agrochemical industries [31]. Despite this practical importance, it has not been clear how to reduce or eliminate segregation. As shown visually in Fig. 9 and quantified in Fig. 8, segregation diminishes when the size ratio between steel beads and glass beads (d_s/d_g) is greater than 2. Although our current work considers only one particle density ratio, these results together with the work of Drahun and Bridgwater [18], Alonso *et al.* [19], and Metcalfe and Shattuck [20] for a variety of types of granular flow, methods to quantify segregation, and particle properties indicate that *segregation is reduced when the heavier beads are larger than the lighter beads so that the size ratio exceeds the density ratio* $(d_{\text{heavy}}/d_{\text{light}} > \rho_{\text{heavy}}/\rho_{\text{light}})$ [17]. Since the particle mass is directly related to the particle diameter and density, this relation can be recast in terms of the mass of the particles, m, so that mixing dominates when

$$d_{\text{heavy}}/d_{\text{light}} > (m_{\text{heavy}}/m_{\text{light}})^{1/4}. \tag{3}$$

The results presented here for a square tumbler reinforce this statement, even in the presence of a complex flow resulting from a noncircular tumbler.

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