# Flow and pattern formation in a binary mixture of rotating granular materials

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In a horizontally rotating cylinder, size segregation, pattern formation, and its time development are studied for a binary mixture of rodlike and disklike materials at various rotational frequencies. The rodlike particles formed a network that influenced their mobility and the shape of the avalanching surface. Windows installed on the cylinder enabled us to examine and control the distribution of the components of the mixture throughout the bulk. This has allowed us to study the evolution of naturally occurring and artificially created patterns. All observed patterns had a degree of asymmetry and were unstable. The stability of a band pattern is shown to depend on its symmetry. Qualitatively, the time for the transition from one set of bands to another was inversely related to the degree of asymmetry of the pattern. In addition, we propose that the parameter $D/d$ (diameter of the cylinder over the diameter of the grains) plays a significant role in the functional dependence of the avalanching surface current on the dynamical angle of repose, and in the segregation process itself.

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## I. INTRODUCTION

In addition to having numerous industrial and geological applications, the unusual and counterintuitive properties of granular material have captured the interest of scientists and engineers alike. While granular materials, under some circumstances, exhibit fluid or solidlike behavior, they are distinctly different from ordinary solids and liquids. For example, as with fluids, the collective shape of a static pile of granular material is defined in part by the shape of the boundaries. On the other hand, equally important influences are friction, gravity, and recent dynamic history. An example that is relevant to the present work is the behavior of a sand pile. A sand pile at rest with a surface slope less than the angle of repose, $\theta_r$, behaves in some respects like a solid in that it keeps its shape. As material is added to the pile, no spontaneous movement takes place until the surface slope is increased above the maximum angle of stability, $\theta_c$, whereupon grains begin to flow and an avalanche of particles occurs in a relatively thin boundary layer near the surface. Between $\theta_r$ and $\theta_c$ is a region of complex, bistable behavior where the surface can either be moving or stationary depending on its dynamic history. For angles larger than $\theta_c$ the flow can be either smooth or accompanied by avalanche waves. This aspect of granular dynamics has been used to model systems that are marginally stable. Other aspects of granular dynamics that depend on the size and surface properties of the grains, such as convection, size segregation, and pattern formation, are unique and the subject of many investigations [1–4].

Size segregation, unique to granular materials, has been observed and studied in many different experiments where a mixture composed of different grains is shaken or placed under shear. When granular materials are shaken, particles of different size tend to separate. This process is believed to be driven by convection currents generated within the medium caused by the shaking process. These convection rolls, for a given composition of grains, can be changed depending on the shape of the container and the wall friction [5]. Size segregation has also been observed in Couette-like flow where the grains of different sizes occupy the space between two coaxial cylinders while the inner cylinder is rotated. Here the larger particles rise to the top and remain on the top surface. The segregation can be explained in terms of a combination of sifting and convective flow generated by the shear [6]. A third kind of segregation, accompanied by pattern formation, occurs when a cylinder partially filled with a mixture of granular materials that may differ in size, roughness, density, etc., is rotated about a horizontal axis that is coincident with the cylinder's axis of symmetry.

Under certain circumstances, for example, a two component mixture segregates into an alternating pattern of bands arranged along the cylinder's axis (axial segregation). The pattern is unstable and changes with time through band shrinking and merging until steady state is reached, as first reported by Oyama [7]. Donald and Roseman did an extensive study of the mixing and segregation in small aspect ratio (length-to-diameter) cylindrical tubes for many different binary mixtures. They concluded that, in addition to the difference of the static angle of repose of the components, the effect of the ends of the cylinder is fundamental in starting the axial segregation [8]. Their work was further developed by Das Gupta *et al.*, who investigated the role of rotation speed on the segregation and the axial pattern formation in small aspect ratio tubes. They concluded that the parameter that drives the segregation process is the difference between the dynamic angle of repose of the components [9]. The dynamic angle of repose, the angle that the moving surface of rotating granular material makes with the horizontal, is rotational frequency dependent, and the functional dependence can be different for different components. Therefore a system can show segregation and instability in some ranges of frequencies and not others.

A model was proposed by de Gennes and Savage to explain the physical mechanism for the segregation based on the difference of the dynamic angle of repose of the two components [10]. Hill and Kakalios reported that in the case of some binary mixtures, using glass beads of different sizes, the difference between the dynamic angle of repose for the mixture and the more mobile component approached zero as

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the rotation frequency was lowered. As a result they could achieve homogeneous mixing by lowering the rotation speed after the band had formed at some higher frequency [11]. Zik *et al.* performed experiments with mixtures of sand and glass beads, and with mixtures of glass beads of various sizes, in large aspect ratio tubes. For a mixture of sand and glass beads, they observed that the nucleation of bands occurred roughly simultaneously in various places along the tube [12]. This is in contrast with the earlier findings that the segregation begins at the end walls and propagates through the tube length [8]. They also found that for a mixture of glass beads of various sizes and identical surface properties no segregation was observed, in contrast to the results previously reported [11]. Frette and Stavans have studied the long-term evolution of the band formation in a mixture of sand and glass beads in large aspect ratio tubes. After the initial establishment of the bands they noticed that the two types of band (glass and sand) evolved differently, indicating a different mechanism was at work in addition to the diffusion. They noticed that the glass beads flowed smoothly while the motion of the sand was punctuated by avalanches that propagated axially. These avalanche ''waves'' were responsible for the exchange of material between the glass bands through wide sand-rich bands resulting in the change of the initial band structure [13].

The present work focuses on the pattern formation, at various frequencies, for a mixture of rodlike (rice) and disklike (split pea) particles with comparable surface properties. In addition to exploring the effect of these shapes on the patterns, we examined the long-term development of band formation by monitoring both naturally occurring patterns and patterns artificially set up by us. We redesigned the cylinder so that we were able to observe the surface development as well as probe into the interior. In addition we looked for evidence of avalanche waves and their role in pattern dynamics.

## II. EXPERIMENTAL SETUP

All experiments reported here were performed in a cylinder rotated about a horizontal axis arranged to coincide with the axis of cylindrical symmetry. The cylinder is made of clear acrylic with an inner diameter of 5.75 in. and a length of 25 in. Our basic experimental setup was similar to that used in Refs. [14] and [11] with some modification discussed below. During the course of our experiments we used two sets of end caps, one constructed out of acrylic (the same as the cylinder) and the other from rubber with a different frictional property than acrylic.

Four windows of size $76\times 51$ mm were cut along the length of the cylinder. The windows and the caps were carefully machined so that they did not alter the shape or the frictional property of the tube. A series of identical experiments were performed in tubes with and without windows, and the results showed that the presence of windows had no effect on the outcome of the experiments reported in this paper. The windows were designed for the purpose of testing various aspects of the flow dynamics such as introducing bands or material at any position along the cylinder and achieving a good homogeneous starting mix that we did not find possible using other techniques. The granular mixture we used consisted of long grain rice and yellow split peas. The rice grains are nearly rodlike with diameters of $1.8\pm 0.2$ mm and lengths of $6.5\pm 0.2$ mm; the pea grains, nearly hemispherical in shape, have diameters of $5.8\pm 0.2$ mm and thicknesses of approximately one half the diameter $2.8\pm 0.2$ mm. The surface texture of split peas is very similar to that of rice, as concluded from static friction measurements. However, rice grains can roll and slide along an avalanching surface while the peas can only slide, occasionally flipping over.

In all experiments, the cylinder was half filled with either one of the components or a mixture of equal parts by volume of rice and split peas. The range of rotation frequencies used was 2–25 rpm. The various measurements of the angles of repose and the evolution of the bands were made by using a charged-coupled-device camera in conjunction with a video recorder and an image processing system. An isolation collet was used in conjunction with the motor for greater stability and noise reduction. A sieve was used to sort the two components for determining the changes of the concentration of a particular point along the tube after a period of rotation.

## III. RESULTS

Our experimental results are divided into three parts: measurements of the angle of repose, band formation, and stability of the bands.

### A. Measurements of the angle of repose

Figure 1 shows the measurement of the dynamic angle of repose $\theta$ versus rotation speed $\Omega$ for each component and for the mixture. The average and the standard deviation were based on data taken from videotaped images. The angle of repose for the rice was higher than that for the peas due to the difference in shape of the two components. The needlelike structure of the rice helped the formation of clumps and contributed to a larger effective friction and a larger angle of repose. The values of the standard deviation for the pea at various frequencies are approximately constant and lower than the corresponding values for the rice. This difference can be attributed to the surface flow properties of the two components. Our observations show that the peas moved down the avalanching plane in smooth continuous fashion while this was not the case for the rice. The flow of rice down the avalanching surface was continuous but accompanied by random clumps resulting in a large variation in the measurements of the angle of repose, which in turn is reflected in the size of the error bars. The dynamic angle of repose versus rotational frequency for split peas shows an approximately linear functional dependence. In comparison, the plot for the rice shows a higher rate of increase at low rotation speeds and a lower rate, approximately the same as the peas show for the higher rotation speeds. The lower rate may be caused by the decrease of effective friction at higher rotation speeds that is, in turn, due to dilation of the medium. The angles of repose for the mixture and the peas alone both decrease as the rotation speed is decreased. Still, the two remain distinctly different, thus eliminating the possibility of mixing at very low rotation speeds [11].

An important feature of the avalanching surface is its profile. At low rotational frequencies the surface is flat. As the

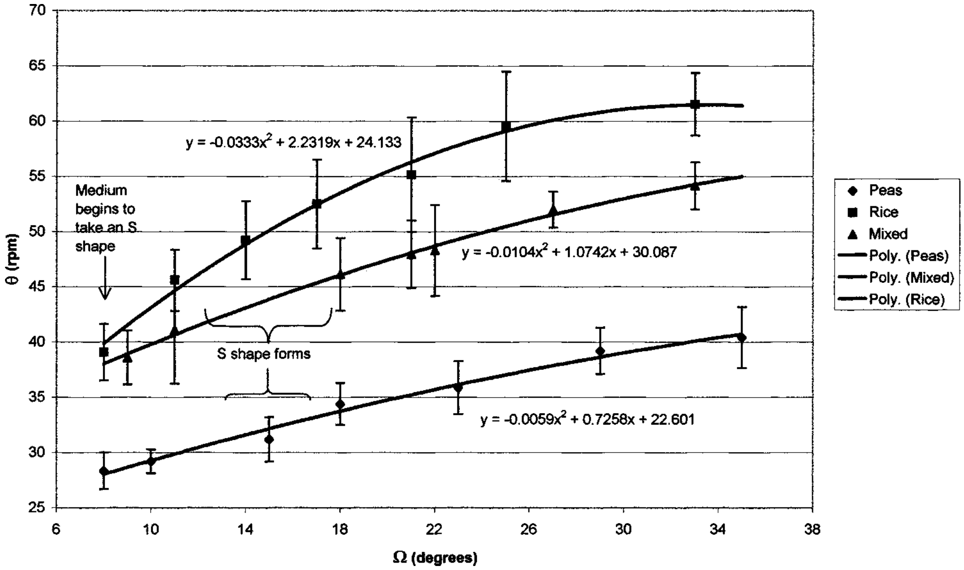


FIG. 1. Dynamic angle of repose ($\theta$) as a function of rotation frequency ($\Omega$) for rice, peas, and the mixture of the two.

rotation frequency increases so does the speed of the avalanching material, and the surface gradually acquires an *s*-shaped contour [11,15]. The change of contour from flat to S shaped occurs at 14 rpm for the peas and at 6 rpm for the rice. Since the two surface profiles do not change at the same frequency, there are in a mixture three possible different combinations of surface profiles over the range of frequencies used in our experiments (2–30 rpm).

Rajchenbach investigated the relationship between the surface current $J$ and the deviation of the dynamic angle of repose from the critical angle, $\theta-\theta_c$, for a range of frequencies where the avalanching surface was flat and the flow was continuous [15]. It has been suggested by de Gennes and Bak and Tang [16] that the current follows the following critical law:

$$J \sim (\theta-\theta_c)^m.$$

For a half-filled cylinder it can be shown that

$$J=\tfrac{1}{2}L\Omega R^2,$$

where $L$, $R$, and $\Omega$ are the length, the radius, and the rotational frequency of the cylinder, respectively. As a result of the two relations above, one can see that

$$\Omega \sim (\theta-\theta_0)^m,$$

and $m$ can be found experimentally by plotting the log of $\Omega$ the log of $(\theta-\theta_c)$. For spherical glass particles Rajchenbach found $m=0.5\pm0.1$, and it differs from the numerical prediction ($m=0.7$) of Tang and Bak appropriate for surface flows and independent of the container's geometry. Our experimental results are shown in Fig. 2 with the corresponding values of the exponent $m$. These values, 1.9 and 2.6, are radically different from the predicted values. The main sources contributing to the discrepancy are the nonspherical shapes of the grains and the small ratio of the cylinder diameter to the grain size ($D/d=22$) used by us compared to the ratio ($D/d=63$) used by Rajchenbach. The larger exponent for the peas is consistent with the slower rate of change of the angle of repose for the peas compared to that for the rice, as seen in Fig. 1.

### B. Segregation and pattern formation

The evolution of band formation was studied at frequencies 5, 10, 15, 20, and 25 rpm in a one-to-one mixture by volume of rice and split peas. Radial segregation has been observed and reported in previous experiments [8,11]. In our experiments, the needlelike rice particles sieved through the holes generated due to the velocity gradient in the avalanching top layers and formed a rice core enveloped by a few layers of peas as shown in Fig. 3. Using the removable windows along the cylinder, we were able to check and verify the simultaneous development and formation of the rice core along the cylinder's axis. The core formation completes within a couple of minutes after the rotation begins and normally before the appearance of any obvious visual signs of axial segregation. The speed at which the radial segregation occurs depends on the rotation frequency of the cylinder. We observed radial segregation in our experimental frequency range and noticed its absence at higher frequencies. For example, at 45 rpm radial segregation did not take place and most of the cylinder stayed in a fairly mixed state except for two narrow rice bands (with some pea content) near the end walls.

This leads us to believe that the radial segregation is a precursor to the axial segregation. The radially segregated

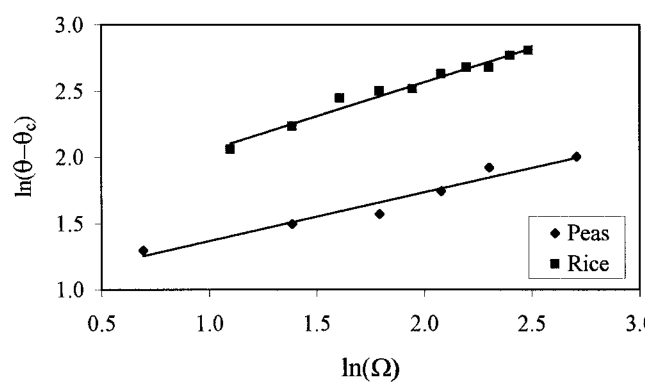


FIG. 2. Log of the deviation of the dynamic angle of repose from critical angle ($\theta-\theta_c$), as a function of the log of the rotation frequency ($\Omega$). The data are for the range of $\Omega$ where the avalanching surfaces are flat.

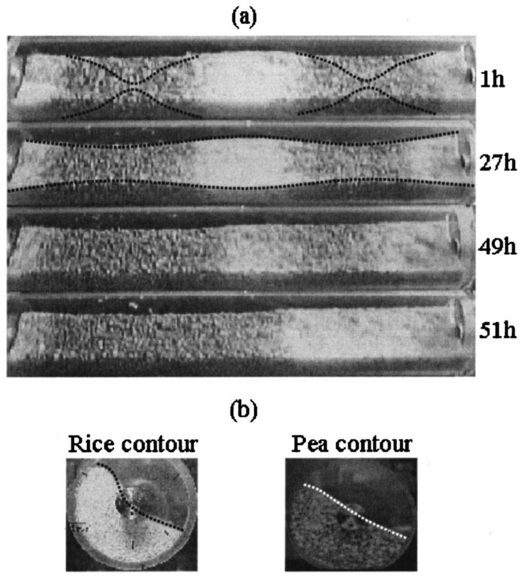


FIG. 3. (a) The evolution of a band pattern at $\Omega = 20$ rpm. The black dotted lines on the top picture show the rice profile under the pea bands and the lines on the second from the top picture show the top and bottom profile of the band pattern. (b) Rice and pea surface contours at 20 rpm.

rice core moves solidly with the cylinder and is in contact with the end walls at both ends while the surface flow is purely made up of the more mobile component (peas). Within 5–10 min the initial signs of the axial segregation appear at the end walls of the cylinder. The higher friction between the end wall and the rice layer compared to that between the rice layers increases the dynamic angle of repose of the radially segregated mixture at the end walls. This fact is revealed by the height profile of the highest point on the free surface, which is representative of the variation of the dynamic angle of repose along the tube. The larger angle of repose at the end wall causes the surface material to avalanche down and away from the end wall, leaving an area with higher concentration of rice next to the walls and creating an area with higher concentration of peas next to it. Since the two components in a mixture have different dynamic angles of repose, then any concentration fluctuation will be unstable and result in segregation [10]. In principle, a concentration fluctuation can exist anywhere along the cylinder. However, in this case the largest concentration fluctuation is generated at the end wall. The segregation process, initiated at the end walls, propagates in the axial direction toward the center of the cylinder, resulting in a pattern of segregation bands. This process overrode any other ripples of segregation that might have begun elsewhere in the medium. One could liken the process to two strong traveling waves moving in opposite directions through the material with their interference resulting in a standing wave pattern. This observation is in agreement with some previous experiments [11]. Zik *et al.* [12] found the nucleation of bands to be homogeneous, initiated by fluctuations and not initiated at the ends of the tube unless the material at the end had very different frictional properties from the rest of the tube.

In our experiment, after the formation of the core and before the formation of bands, the rice core will be in contact only with the end walls and the surrounding mixture. If the frictional force at the end wall is small then the bands that nucleate due to fluctuations away from the wall get a chance to grow before any strong process begins at the end walls. In our experiment, the large fluctuations at the end walls speeded up the propagation of the segregation and overrode the small local nucleations. When we artificially introduced a stronger fluctuation at a random point along the axis, provided it was large enough, it did initiate band formation and influenced the overall structure. To check the effect of the end wall we changed one of the caps to rubber, introducing a larger coefficient of friction at one end. Segregation first began at the rubber cap and moved through the medium—the lack of symmetry in the friction resulted in the lack of symmetry of the pattern.

Zik *et al.* did not observe any segregation with a mixture of glass beads of different sizes. We ran one experiment at 10 rpm with a one-to-one mixture, by volume, of 2 and 3 mm transparent glass beads (we did not color them in order to avoid possibly altering the surface properties). The mixture segregated in bands, which was confirmed by using the windows and examining the contents at various points along the tube. When comparing our results with Zik *et al.*, a parameter that may be of significance is the ratio of the tube's diameter to the average particle size, $D/d$, which was typically much smaller in our experiments (20 for us compared to their 100). The combination of low friction at their end walls and their larger value of $D/d$ could underlie the differences in experimental outcomes cited above. As a result, we believe that the parameter $D/d$ may play a crucial role in the segregation process.

### C. Number of bands and their stability

Starting with a homogeneous mixture and identically machined end caps, we followed band formation and evolution over a frequency range of 5–25 rpm at five specific frequencies. The summary of the results is found in Table I. The second column of the table reveals that the lower the rotation frequency, the larger the number of bands in the initial band structure. Our experiments show that the symmetry of the band structure is greatly influenced by any nonuniformity of the initial mixture and the levelness of the tube. Since we were able to get a fairly homogeneous initial mix, all of our patterns were fairly symmetric and the experiments were repeatable. In all cases the patterns contained an odd number of bands. All the initial patterns remained seemingly unchanged for an extended period of time before making a transition to a new metastable state with fewer bands. All subsequent band structures made transitions to new metastable states until they reached a final and seemingly stable state. The final state is asymmetric and composed of two rice bands at the two ends separated by the more mobile split pea band. As is seen from Table I, the pattern evolution time varies a great deal and some transitions took several days. Examining the transition time data for the 5 rpm run in Table I, we observe that the wider the bands of a pattern the longer it takes for that pattern to make a transition to one of fewer bands. Also, the comparison of the time interval for a five-band pattern to change to a three-band pattern, at various frequencies, suggests a minimum in the vicinity of 10 rpm. We noted that no transition leading to an increase in the

TABLE I. A summary of the number of bands and the time for the evolution of each band pattern at various frequencies. $t_1$, time to establish initial pattern; $t_{7\rightarrow5}$, time for transition from seven bands to five bands; $t_{5\rightarrow3}$, time for transition from five bands to three bands; $t_{tot}$, total experimental run time.

| $\Omega$ (rpm) | Surface profile | No. of initial bands | $t_1$ (minutes) | $t_{7\rightarrow5}$ (hours) | $t_{5\rightarrow3}$ (hours) | $t_{tot}$ (hours) |
|---|---|---|---|---|---|---|
| 5 | Flat profiles for both | 7 | 120 | 45 | >100 | 100 |
| 10 | Peas flat; rice S shaped | 5 | 90 | | 14 | 30 |
| 15 | Peas intermittent; rice S shaped | 5[a] | 30 | | 23 | 25 |
| 20 | Both S shaped | 5 | 22 | | 57 | 113 |
| 25 | Both S shaped | 5 | 17 | | [b] | 2 |

[a]Occasionally a seven-band pattern was observed that was short lived or partially developed.
[b]No stable pattern apeared within the time allotted.

number of bands was observed after the system was started with a homogeneous mix and an initial pattern had been established. On the other hand, when we manually set up an asymmetric three-band pattern (with two pea bands separated by a rice band), the number of bands first increased by two and then changed to the final asymmetric three-band pattern.

To isolate the cause of the general pattern of shrinkage of the number of bands, we examined the videotape of the experiments where this effect took place. We found that whenever two split pea bands of unequal width are separated by a rice band, the content of the narrower split pea band seems to be transported over the rice band to the wider split pea band. This process increases the width of one band and shrinks the other until it produces a pattern with two fewer bands. The speed of the process is proportional to the difference of the two split pea bandwidths and, thus, it speeds up as it goes. All patterns resulting from various experiments beginning with a homogeneous mixture have some degree of asymmetry and, therefore, are all unstable. The more asymmetric a pattern, the faster its transition to a new band structure. Figure 3(a) shows a typical band pattern at 25 rpm and its subsequent development. We used the windows along the cylinder to probe into the interior of the bands and discovered that the pea bands are not made of pure peas and that the rice bands extend beyond the visible boundaries under the pea bands. In a narrow enough pea band the rice forms a core that tends to raise the pea surface. Therefore the narrower a pea band is, the larger the rice core, and the higher the angle of repose of the pea band at its center. The dotted lines in Fig. 3(a) schematically represent the rice profile within the pea band. The shape of the avalanching surface when segregated is complex. Radial profiles of the two surfaces show that both are *s* shaped, with the rice band having a deeper curvature as shown by an inset in Fig. 3(b). Axially, at the top of the avalanching surface, the rice band, with a larger angle of repose, curves down on either side of the band to match the lower angle of the adjacent pea bands. This creates a hump in the rice profile and a valley in the pea profile. At the bottom of the avalanching surface, the situation is reversed since the rice band curves up to match the adjacent pea bands. Thus a valley is created in the bottom of the rice band and a hump at the bottom of the pea band. The axial cross section of the surface is also S shaped, and as one moves to the middle of the surface the S-shaped cross section flattens out. Below the central line the cross sections are the mirror images of the ones above the axis of the cylinder. The dotted lines in Fig. 3(a) demonstrate this fact. As a result of the geometry, the peas at the lower half of the avalanching surface can flow to the rice band where with the rest of the flow they are brought to the top surface. Once on the top surface they can move down and sideways (on the top half) due to the curvature of the surface. However, there is a higher probability of sliding to the wider pea band since the asymmetry of the two pea bands has caused an asymmetry in the curvature of the rice band (steeper on the side of the wider band).

To confirm the aforementioned observation, we used the windows along the cylinder to directly access the rice and split peas, and we created five-band patterns, with and without symmetry, to study their evolution. Figure 4 shows two such sets of initial conditions and their development. The symmetric band structure lasted for over a day (we did not continue the experiment) but the asymmetric structure very quickly showed signs of change and within a few hours made a transition to a new structure with fewer bands. Since even the symmetric structure has some small degree of unintended asymmetry, we expect it to change to a three-band pattern after a very long time (of the order of a week).

## IV. CONCLUSION

Our study showed that the critical exponent *m*, relating surface current and the dynamic angle of repose for our binary mixture of rodlike rice particles and disklike split pea particles, is quite different from the *m* obtained for spherical particles. Also, the shape of the particles affected the local

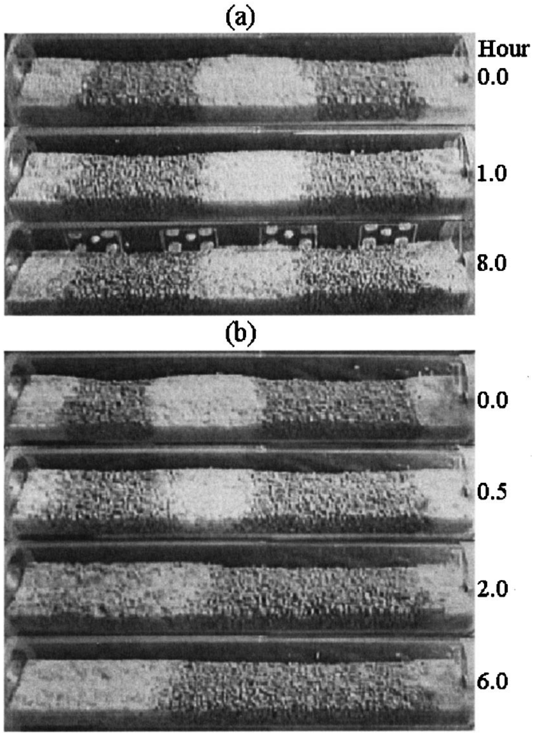


FIG. 4. The evolution of two sets of artificial band patterns. (a) A symmetric band pattern. (b) An asymmetric band pattern.

dynamics of the flow along the avalanching surface in that the disk shaped peas flowed smoothly while the flow of rod-like rice particles was accompanied by avalanche waves. We have investigated the global behavior; that is, the formation of patterns and their evolution, of the mixture in a cylinder specially designed to allow us to move past surface observation alone.

Installing windows on the cylinder enabled us to examine and control the distribution of the components of the mixture throughout the bulk. First, we have been able to examine the distribution of the components beneath the surface as they evolve away from a homogeneously mixed state. Second, we have set up our own starting distributions of the two components and studied the natural evolution of these artificial initial states. By monitoring the evolution of the mixture below the surface we observed segregation within any given cross section of the cylinder (radial segregation) as well as along its axis (axial segregation). In general, we noted that radial segregation preceded axial segregation and we found it possible to relate the subsurface behavior to surface dynamics with a resulting detailed explanation of the surface shapes of the avalanching materials.

We followed the formation of bands in our binary mixture, starting from a homogeneous state, for a number of rotational frequencies and found that the number of initial bands increased with decreasing frequency. By following the time for transitions from the first metastable pattern of bands to the next metastable pattern, with fewer bands, we noted that the transition time first fell and then steadily rose as the rotation rate was increased from 5 to 25 rpm. The most rapid transition rate was in the neighborhood of 10 rpm.

Examining the interior of the bands we found, for example, that there were no entirely pure pea bands although, in the middle of a wide pea band (about the diameter of the cylinder) the center of the band was almost pure. The rate of change of, or instability of, the naturally occurring bands appears to increase with the asymmetry of the surface band structure. By hand creating artificially asymmetric bands, we were able to study their evolution and verify this behavior. We identified the end-cap friction and the parameter $D/d$ (diameter of the cylinder over the diameter of the grains) as playing a significant role in the functional form of current along the avalanching surface and the segregation process as a whole. Our future plans include looking into the effect of $D/d$, the role of the avalanche waves in our system, and studying the transition time for $n$ to $n$-2 bands as a function of frequency, starting with manually prepared, fixed initial conditions.

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