

## Traveling Wave of Segregation in a Highly Filled Rotating Drum

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The dynamics of a segregation pattern for a granular mixture in a highly filled rotating drum were studied. A spontaneously segregated band pattern traveled laterally and was accompanied by the repeated creation of new bands near the center of the drum and annihilation at both of its ends. The presence of nearly stationary convection plays an essential role in causing this traveling wave. Based on direct observations of both the interior and the exterior of the segregation pattern, this spatiotemporal pattern is interpreted in terms of a one-dimensional Cahn-Hilliard equation by including the effect of stationary convection.

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In dry granular materials composed of heterogeneous particles, the individual species often segregate under mechanical agitation, such as flowing, shaking, or rotating, although one might naively expect mixing of the particles based on a consideration of entropy [1,2]. Unlike mixtures of usual liquids, dissipative interaction between the particles gives rise to unconventional macroscopic behavior. When a horizontal drum that is partially filled with a mixture of two different types of grains is rotated, the binary mixture segregates into alternating bands of relatively pure single concentrations along the axis of rotation. The axial pattern is then accompanied by a monotonic decrease in the number of domains [3].

Despite the absence of a general theory of granular media, many experimental [4–8] and theoretical [9–11] studies have been carried out to elucidate the physics of this counterintuitive phenomenon. The axial segregation process has also been reproduced numerically ([12,13], and references therein). In these previous studies, it has been widely believed that segregation is driven by successive avalanche on the free surface [8,9]. Since the dynamic angle of repose is the quantity which characterizes the angle of avalanche, it has been considered as a control parameter. However, observations of interior segregation by magnetic resonance imaging (MRI) have revealed that the bands are not necessarily composed of pure single species, but rather, the smaller species seemed to exist along the core of the overall axis [14]. This indicates that the banding dynamics do not evolve exclusively due to an avalanche, but also arise due to internal flow along the core in an axial direction [11]. In highly filled cases, it would be suitable to see the effect of the global convection with minimizing a diffusive effect driven by avalanche. Nevertheless, there has been only one previous study on a nearly filled case in a cylinder [15], which observed axial segregation but not a banding dynamics. There are also a couple of studies on nearly filled cases of quasi-2D cell systems [16,17], although they might have restricted axial effects by the geometry.

In this study, we considered a rotating drum and started with a completely filled state. We focused on the axial motion of grains by minimizing the effect of surface flow by setting the filling fraction as high as possible. We also show a direct evidence of the interior segregation structure by simply opening the drum horizontally. The experiments were performed using two species of grains in a transparent acrylic drum with an inner radius  $R$  of 4 cm and a length  $2L$  of 32 cm [Fig. 1(a)]. To observe interior segregation in the bulk, we first cut the drum along the axis into two semi-circular pieces, which we then bolted firmly together so that we could separate them later. The granular media was composed of garnet sand (red, mean diameter  $d \sim 1$  mm) and silica sand (white,  $d \sim 0.15$  mm) with irregular shapes, and all of the pictures presented here are monochromatic. Thus, garnet sands are referred to as black particles hereafter. Into a vertical drum that had one end open, we first poured garnet sand up to the midpoint, and then completely filled the drum with silica sand. The drum was placed horizontally [Fig. 1(a)], then it was rotated at a constant angular frequency  $\omega$  of 30 rotations per minute (rpm). Over time, slight compaction was noticed, and the final filling fraction  $\phi$  was approximately 0.96. Figure 1(b) shows the exterior segregation patterns that were photographed every 8 min from  $t = 1216$  to 1248 (min). The black line at the center is a marker to indicate the middle of the drum. The bands are created at the middle of the drum and then travel outward. In the image at 1248 min, the creation of new bands is observed.

Figure 1(c) shows a space-time diagram of the segregation pattern at the surface of the drum during rotation for 0–2000 min. The diagram is generated by stacking one-pixel-wide horizontal lines along the axis every 1 min. After approximately 100 rotations, bands of larger black grains start to appear one after another near the middle of the drum and move toward the ends. The segregation dynamics does not reflect monotonic coarsening, as is seen in partially filled cases. The creation and annihilation of the traveling bands sustains over several days,

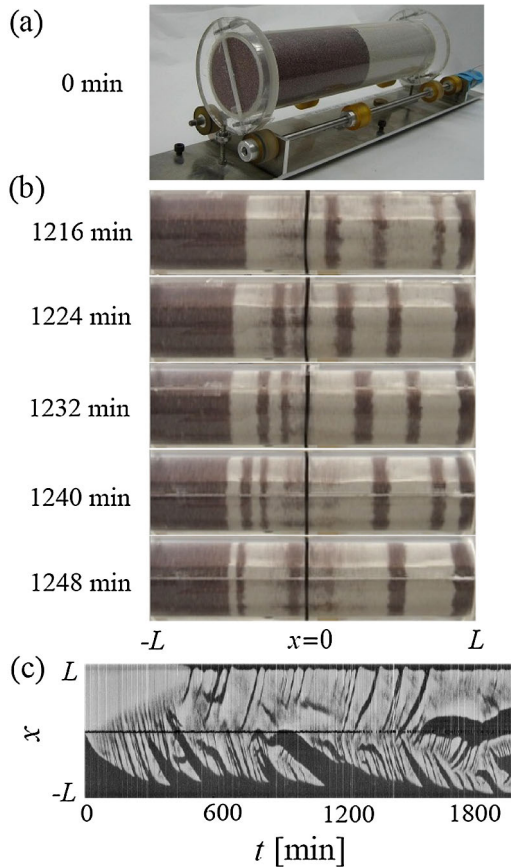


FIG. 1 (color online). (a) Experimental setup. (b) Traveling waves of axial bands. (c) Spatiotemporal diagram of the evolution of the segregation pattern at the surface.

suggesting that such a spatiotemporal pattern is the representation of a nonequilibrium structure under dissipative condition. In addition to the regular traveling pattern, we note an appearance of intermittent type irregularity.

To obtain insight into the interior dynamics of axial segregation, we studied the segregation pattern in the interior after 8555 min of rotation. The upper image in Fig. 2(a) shows the exterior at that time. The drum was then opened and the upper half of the drum with its sand was removed with meticulous care. The lower image shows the interior segregation. The interior of the drum can be classified into three regions: the outer, middle, and core parts (regions I, II, and III, respectively), as shown in Fig. 2(b). In region I the media is well segregated into black and white phases. In region III, the core domain along the axis consists of a homogeneous mixture. Region II, between regions I and III, consists of a pure white phase. Bands tend to appear near the middle of the drum and travel toward the ends, as shown in Fig. 1(c), and a tentative sketch of particle flow inside the drum is drawn in Fig. 2(c) based on a consideration of the internal structure. The black domain in region I is surrounded by the pure white phase of regions I and II. Inspections on the interior pattern indicated that the pure white phase in region II appears at

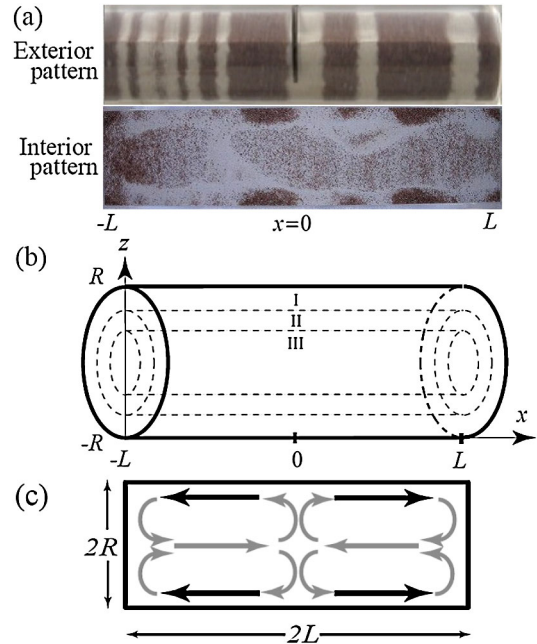


FIG. 2 (color online). (a) The exterior and interior segregation patterns after 8555 min of rotation. (b) Schematic diagram of the domains; region I: two-phase separation; region II: pure white phase; region III: mixed phase. (c) Tentative sketch of particle flow. Outward flow (black arrow) occurs in region I and inward flow (gray arrow) occurs in region III.

essentially the same depth at different experimental runs regardless the rotation period of 1–110 h. This suggests that there is almost no mixing effect between regions I and III. Thus, we conjecture that the white grains in region II behave as an intervening layer. The well-segregated grains in region I flow outward as denoted by black arrows. When the grains reach the ends of the drum, the grains form a mixture. This mixed phase returns to region III and flows inward (gray arrows). At the middle of the drum, the mixture in region III again enters region I with phase separation. Although the boundary of region I exhibits time-dependent fluctuation, here we would like to focus our interest on the manner of the global convection.

Inspired by the direct observation of the interior and exterior segregation patterns, we propose a simple model to describe the band dynamics of axial segregation based on a one-variable 1D Cahn-Hilliard equation. First, we focus on the two-phase separation in region I and the traveling bands at the surface of the drum. We define the fraction of black grains at  $x$  as  $c(x) = S_{bl}(x)/[S_{bl}(x) + S_{wh}(x)]$ , where  $S_{bl}$  and  $S_{wh}$  are local partial volumic concentrations at  $x$  in region I of black and white grains, respectively. The variable  $x$  is the spatial coordinate parallel to the axis of the drum and its origin represents the middle of the drum. We define an order parameter  $\eta$  as  $\eta(x) = [c(x) - c_{min}]/(c_{max} - c_{min})$ , where  $c_{max}$  and  $c_{min}$  are the maximum and minimum concentrations of the phase. We conjecture that at  $x = 0$  the mixed

phase is a source of grains. As soon as the grains enter region I, they flow toward the end of the drum at the advection velocity  $u$ . As seen in Fig. 1(c), relatively narrow stripes travel almost parallel until they meet infrequent wide bands. We assume that the flow velocity in region I is constant. Since there is no exchange of black grains between regions I and III, we consider that the system is conservative in region I. Thus, we can write the equation for the concentration of the black phase  $\eta$  as

$$\frac{\partial \eta}{\partial t} + u \nabla \eta = \lambda \nabla^2 \left\{ -\epsilon^2 \nabla^2 \eta - \frac{df}{d\eta} \right\} + a^+ \eta|_{x=0} + a^- \eta|_{x=l}, \quad (1)$$

where  $df/d\eta = -\eta(\eta - 1/2)(\eta - 1)$ ,  $\epsilon$  corresponds to the thickness of the boundary layer associated with the diffusion coefficient,  $a^+ \eta|_{x=0}$  is the amount of grains at  $x \approx 0$  incoming from region III, and  $a^- \eta|_{x=l}$  is the amount of grains at  $x \approx l$  returning to region III. Since the grains arriving from region III consist of a homogeneous mixture of black and white grains,  $a^+ \eta|_{x=0}$  is set at 0.5. While it can be determined experimentally, in this study we chose a value of 0.5 for simplicity. Since there is no restriction on  $\eta(x=l)$ , the Neumann boundary condition is chosen. Equation (1) is discretized with a space interval  $dx = 2.5 \times 10^{-3}l$  and a time interval  $dt = 1.0 \times 10^{-6}$ . The parameter values are set at  $\lambda = 0.45$ ,  $\epsilon = 4.5 \times 10^{-3}$ , and  $u = 8.0$ . As shown in Fig. 2(b), the incoming source position shows small fluctuation around  $x = 0$ . However, we adapt the model by ignoring such fluctuation for simplicity. The initial state is chosen randomly between 0 and 1. It then changes to two-phase separation, through a transient coarsening process.

Figure 3(a1) shows a numerical solution obtained by Eq. (1). It shows good agreement with the experiment ( $\phi = 0.96$ ) shown in Fig. 3(a2), which is part of the experiment (Fig. 1). Since there is convection effect near the end of the walls, this domain is not plotted. With turning off the advection term ( $u = 0.0$ ), Eq. (1) is numerically solved as shown in Figure 3(b1). Since there is no source of grains, we set the range from  $x = -l$  to  $x = l$  and the boundary conditions at both ends to the Neumann boundary condition. This is similar to past studies with partially filled drums [5,6,9,10]. For comparison with the numerical results, we also performed an experiment with a partially filled drum ( $\phi = 0.50$ ), as shown in Fig. 3(b2). The filling fraction was changed and the initial condition was a premixed state. All other parameters were the same as in the case of  $\phi = 0.96$ . The space-time diagram coincides very well with the numerical solution shown in Fig. 3(b1). Figure 3(c) shows the exterior and interior segregation patterns of Fig. 3(b2) after rotation for 1100 sec. We obtained the interior image by inserting a transparent plate between the semicircular halves before separating them. This direct observation agrees very well with the results obtained by MRI [14].

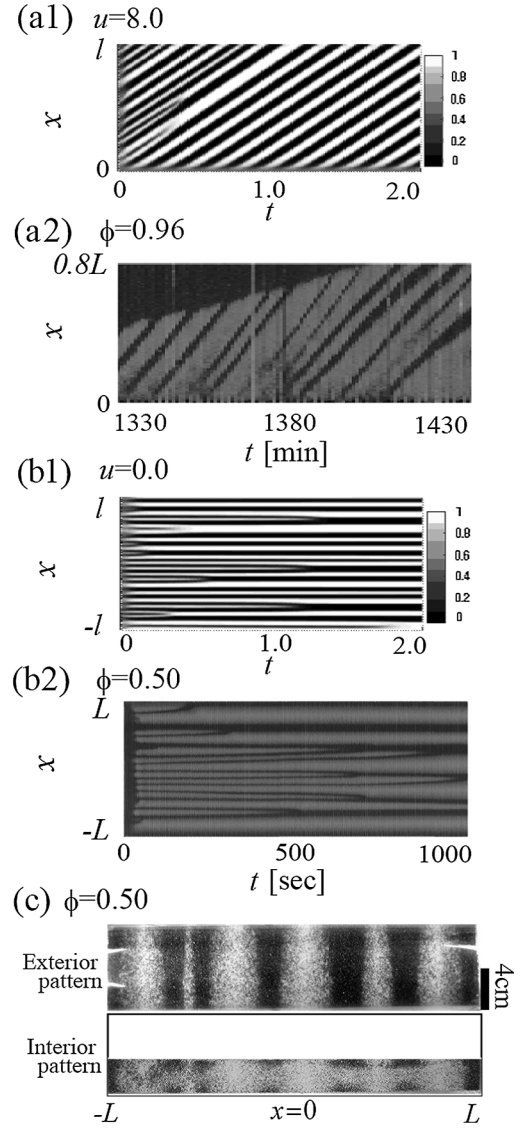


FIG. 3. Space-time diagrams of the numerical results obtained from Eq. (1) in the case that (a1)  $u = 8.0$  and (b1)  $u = 0.0$ , and the experimental results in the case that (a2)  $\phi = 0.96$  and (b2)  $\phi = 0.50$ . (c) The exterior (upper) and interior (lower) segregation patterns in the half filled drum (b2) after rotation for 1100 sec.

In summary, we performed experiments with a nearly filled rotating drum and observed the segregation pattern from both the interior and the exterior. Although it has long been believed that axial segregation is driven by surface flow, we observed segregation even with minimizing the effect of surface flow at a high filling fraction. The evolution of a segregated pattern has been actively studied, yet past reports on the time development have been limited to monotonic type toward a certain stationary pattern. On the contrary, in a nearly filled case, we found an emergence of a new type of dissipative spatiotemporal pattern accompanied by time-successive creation or annihilation of traveling waves. Furthermore, it is noted that the axial motion is



generated in a remarkable manner with suppressing surface flow, being even more significantly than that in partially filled cases. This observation suggests an existence of a further important origin, in addition to the currently believed mechanism of segregation originated in surface flow.

Concerning a further possible origin of axial segregation, we suppose that the grains in region I rotate at the same angular velocity with rotation dragged by the shear force from the interior wall of the cylinder. On the other hand, because of the decay of shear force with depth, the grains in region III cannot follow the rotation and rotate at the lower angular velocity than that in region I. Thus, region II exists between the two regions which flow with different angular velocity. Since region I shows clear segregation and region III shows mixture phase, the shear force from the interior wall of the cylinder can be another possible origin to cause the segregation. To verify this, it would be interesting to study dependence of the segregation dynamics on the roughness of the interior wall, besides the effects of the dynamic angle of repose. In the present study, we assumed that region II is secondary. For the full account of the observed phenomena, further study on the formation and maintenance of the intervening layer would be necessary. Recently, an appearance of segregation-pattern drift has been reported in an experiment on rimming flow in a cylinder consisting of partially filled fluid with granular additives [18]. This study, together with the result of the present study, suggests that a similar mechanism works for the dynamic phase segregation not only for dry granules but also for a liquidlike system.

Based on our observations of the interior and exterior of the segregation, we successfully reproduced traveling bands with a one-variable 1D Cahn-Hilliard equation with advection. We presume that if the angular frequency is decreased, advection will be weakened, and we may observe smaller advection velocity and narrower bands because of the smaller diffusion coefficient  $\epsilon^2$ . The undulating phenomenon at the boundary of region I seems to correlate with the intermittent appearance of the wide bands as shown in Fig. 1(c). We expect that such irregularity would be attributable to a kind of low dimensional chaos, although further careful study is needed to make clear the full scenario.

In a coarsening process, the grains whose bands disappear from the surface tend to be the smaller species in both highly and partially filled cases, as observed in Fig. 1(c), Fig. 3(b2), and in the previous studies [5]. It is also known that before axial segregation occurs, the smaller particles form a radial core, which is retained even during axial segregation, as observed in Fig. 3(c) and MRI experiments [14]. The reason why smaller species are always the ones which disappear can be interpreted as a result of retracting grains of smaller species into the core

domain with approaching two adjacent bands of larger species. This is a novel understanding of segregation dynamics in partially filled cases based on our direct observation of the interior as well as the exterior dynamics.

Generally, our system can be considered as a phase separation that originates in a first-order phase transition under the criteria of Landau. In simple conservative systems, we only observe relaxation processes accompanied by coarsening. However, the strongly dissipative nature of our system can realize convection driven through rotation of the drum. In many cases, the spatiotemporal patterns of phase separation exhibit traveling waves because of hydrodynamic effects. For example, our model is applicable to other conservative reaction-diffusion systems which have incoming and outgoing flux [19]. Our findings are expected to be common to many of these systems. Further studies along these lines may be promising.

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