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Axial Segregation of Powders in a Horizontal Rotating Tube

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Powder mixtures partially filling horizontal, slowly rotating tubes segregate under certain conditions into bands of different composition along the tube axis. The one-dimensional patterns of bands evolve in time through coarsening. Recent work on the origin and dynamics of axial segregation of powder mixtures is reviewed.

KEY WORDS: Powders; avalanches; segregation.

One of the most intriguing and ubiquitous properties of powders is their tendency to segregate according to size when sheared or jostled.[(1)] This phenomenon is apparent for example after landslides and avalanches, where large rocks are found at the bottom of montains, whereas smaller ones remain higher up. Segregation is also important in many applications where granular materials are handled, such as in the pharmaceutical and metallurgical industries: one would like to hope that a batch of pills manufactured from a mixture of powders have all the same composition!

A beautiful demonstration of segregation is afforded by a binary mixture of two powders partially filling a horizontal rotating tube. After a few minutes of rotation, a nearly homogeneous mixture may segregate into a one-dimensional pattern of bands of different composition as illustrated in Fig. 1. The present short review focuses on this phenomenon, called *axial segregation*. In contrast to other ways of effecting segregation, e.g., vertical vibration of powders, axial segregation in a rotating tube occurs in the absence of convective instabilities, and therefore the segregation process

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This work is dedicated to Leo Kadanoff on the occasion of his 60th birthday. Cheers Leo!

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can be studied cleanly. Axial segregation was first reported by Oyama nearly sixty years ago.[2] It is different from the *radial segregation* observed in transverse cross sections of the granular medium, a phenomenon which has also come under scrutiny lately.[3] More than twenty years after Oyama's work, Donald and Roseman studied segregation in small aspect ratio (length/diameter) tubes in an attempt to find the conditions which determine whether bands will form or not.[4] They claimed that banding occurs when the static angles of repose of the small and large particles, (θ_s and θ_l respectively) obey the inequality:

$$\theta_s > \theta_l \tag{1}$$

and that violation of this condition leads instead to a state of radial segregation, in which a core of small particles runs along the tube axis surrounded by a shell of large particles near the tube wall. Static angles of repose are determined by powder characteristics such as the geometrical shape of the grains, and the roughness of their surfaces. Donald and Roseman further observed that axial segregation started at the tube ends, a fact which motivated suggestions that axial segregation is due to end effects: friction at the end walls results in an axial gradient in slope.[5]

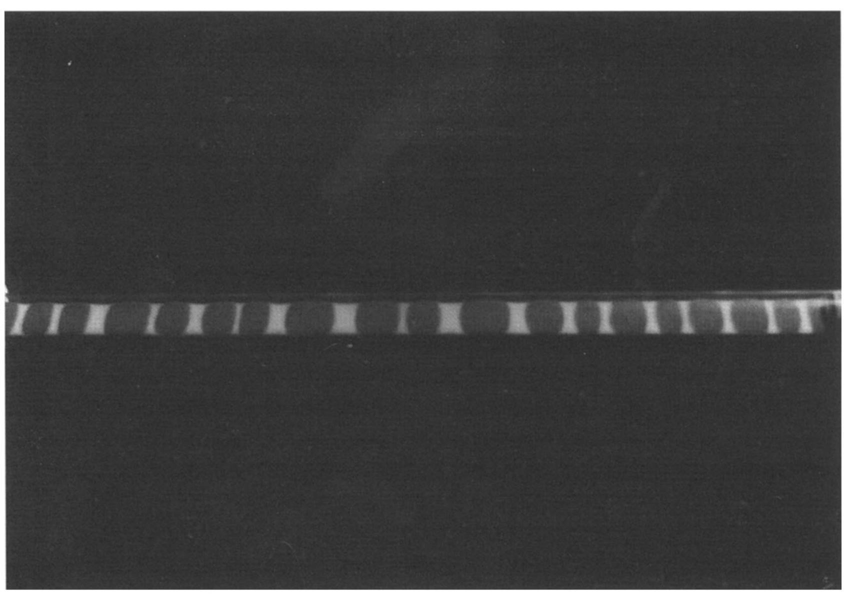

Fig. 1. Typical band pattern formed in a sand-glass bead mixture (black and white respectively) in a tube 60 cm. long after 340 revolutions at 10 rpm. Sand sizes ranged between 0.104 and 0.147 mm; glass beads were 0.208 mm in diameter.

A heuristic picture of why inequality (1) is a good rule of thumb goes as follows: upon rotation, an initially nearly-homogeneous mixture develops local concentration fluctuations. Assuming the validity of the inequality, regions with a higher concentration of small particles are characterized by a steeper slope than adjacent regions in which the concentration of small particles is smaller. The net effect is the creation of an axial slope gradient. Grains at the top of the slope will then not only roll down, but follow axial gradients as well. Since large particles have a larger mobility (they do not get trapped in the roughness of the slope as much as small grains do) they are driven out of steeper sloped regions more readily. This provides a positive feedback mechanism which amplifies existing concentrations fluctuations and promotes the banding instability.

While condition (1) on the static angles of repose provides a guide as to what one may expect to see in an experiment, it does not capture the full complexity of the experimental observations. The angle of the thin layer of flowing grains induced by the rotation is frequency-dependent, as experiments in mixtures of sand grains of different size show.[6] Moreover, the way this *dynamical* angle of repose changes with frequency varies from powder to powder. One could therefore envision a situation in which for some range of frequencies bands would form, whereas starting from a banded configuration and changing the rotation frequency outside that range would lead instead to remixing. This effect has actually been observed by Hill and Kakalios.[7]

More recently Zik *et al.* made experiments in large aspect ratio tubes, tested whether banding occurs only as a result of a size difference, and studied the effects of axial modulations of the tube radius.[8] Their large aspect ratio experiments with sand-glass beads mixtures clearly showed that bands form homogeneously along the tube, ruling out boundary effects as the source of the instability. To study the effects of a size difference alone, they made experiments with internally-colored spherical glass beads of two different sizes. The independence of packing properties of spheres on radius, together with the fact that both constituents had the same surface characteristics led them to expect $\theta_s \sim \theta_l$ and thus to observe no banding. This expectation was borne out by their results (Fig. 2a). Nonetheless, a curious feature was consistently observed: there was some degree of segregation near the curved end of the test tube used in the experiment. This local feature disappeared when the curved end was replaced with a flat wall, hinting at the importance of axial modulations of the tube radius in triggering the formation of bands. Accordingly, the same mixture rotated in a tube with an axial modulation of the radius displayed bands locked with the modulation periodicity (Fig. 2b). The large beads were found in the bellies of the tube. Complete powder segregation was observed when the

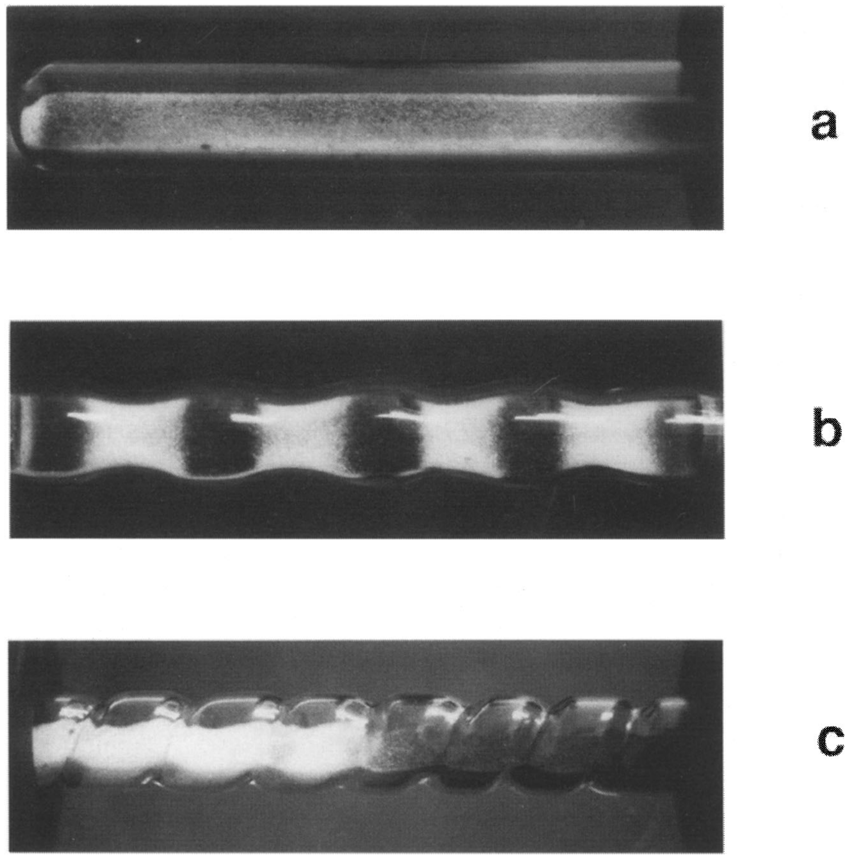

Fig. 2. Segregation in glass beads of two different sizes, 88 μm (black) and 125 μm (white) in (a) a non modulated test tube, (b) a tube with periodic modulations and (c) a tube with a helical modulation. All tubes had a mean diameter of 0.7 cm. (from ref. 8).

mixture was rotated in a tube with a helical modulation (Fig. 2c). The powders exchanged sides upon a change in helicity or rotation sense. Zik *et al.* stressed the importance of the non-symmetric s-shape of the flowing layer in accounting for these observations, and for axial segregation in general: had the profile been straight, segregation at the top of the slope would be compensated by remixing at the bottom. Based on this they developed a model for the initial stages of the segregation process. In the linear regime the model reduces to a diffusion equation with a diffusion coefficient which can become negative. The picture that emerged then is that the banding instability can be regarded formally as a spinodal decomposition process. The putative negative diffusion coefficient is a result of axial gradients in slope which bias the random kicks experienced by grains as they fall down the slope. This latter process, alone, has a tendency to remix rather that to promote segregation.

More recently, two experimental works have challenged this picture. Choo *et al.* studied the initial phases of band formation in large aspect ratio tubes, for different values of the relative volumetric composition ϕ (before mixing) of sand-salt mixtures.[9] The rotation frequency in their experiments was chosen so that both components rolled down smoothly. They noticed that the process of band formation depends on ϕ: whereas for $\phi = 0.55$ bands develop readily after a few hundred rotations, traveling waves of composition are observed for larger values of ϕ, as illustrated in Fig. 3. These waves can propagate in the two directions, and pass through

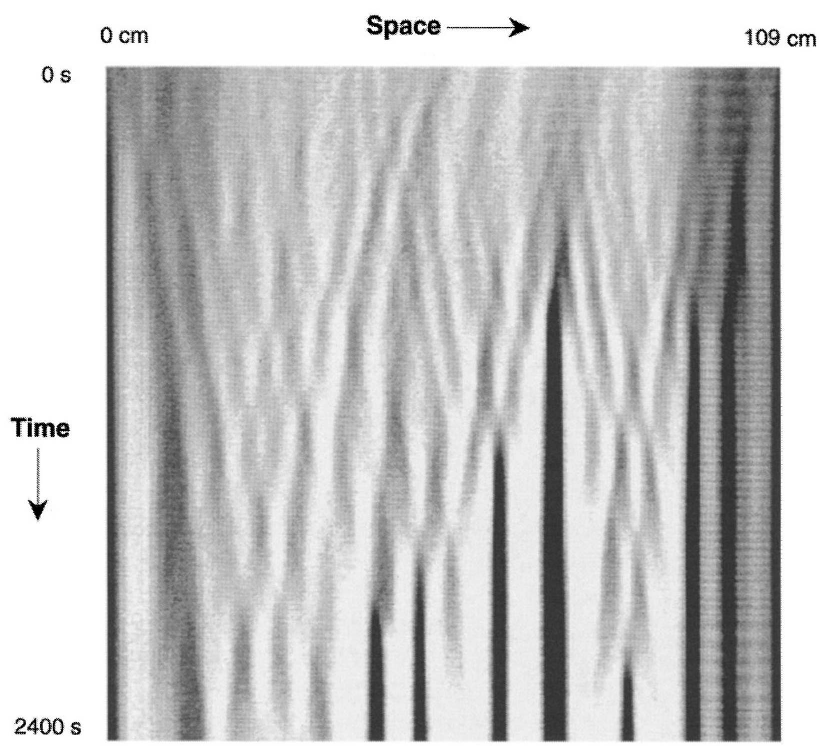

Fig. 3. Spacetime plot of the evolution of a sand-salt mixture $\phi = 0.67$ starting from a well-mixed initial condition. Time runs downward and the frame shows the full length of the tube (from ref. 9).

one another producing regions of standing waves and spatiotemporal disorder. The formation of bands in this regime is much slower, and may take more than a few thousand revolutions.

Frette and Stavans focused on the characterization ot the long term evolution of band patterns.[10] In contrast to the experiments of Choo *et al.*, they chose sand-glass bead mixtures, and rotation frequencies for which sand flow was punctuated by avalanches propagating axially, in contrast to the smoothly rolling down glass beads. After their formation some bands merged, while others grew at the expense of shrinking ones, resulting in a very slow, nearly logarithmic coarsening of the band pattern. A typical space time diagram of the evolution is shown in Fig. 4. Interestingly, the rare occurrence of band formation at very long times hinted that the coarsening process does not lead to complete segregation, and that the system finally settles down into a steady state regime in which the number of bands fluctuates. While this regime was not reached in the experiments, runs commencing from a completely segregated state evolved into configurations in which the smoothly running beads penetrated into the sand half of the tube, eventually forming there new glass bands. The order and position in which glass bands were formed was not consistent with what one would expect from concentration gradients induced by diffusion. Moreover, no sand bands were formed within the glass half of the tube. These facts suggested that the difference in axial transport may be due to the different flow behavior of the glass and sand grains. An analysis of the propagating avalanches in the sand bands indeed showed that the direction of transport in the sand is opposite to the direction of propagation of avalanches.

The noisy behavior of avalanches in this experiment lead Levitan recently to extend the approach of Zik *et al.* to model the long term evolution of bands.[11] He obtained the following equation for ψ, the deviation of the local relative concentration of glass beads from the average value characterizing the non-segregated state:

$$\frac{\partial \psi}{\partial t} = -\frac{\partial J}{\partial z} \tag{2}$$

where the current J is given by:

$$J = k(1-\psi^2)\frac{\partial \psi}{\partial z} + q(1-\psi^2)\frac{\partial \xi}{\partial z} \tag{3}$$

Here z is a coordinate in the axial direction, k and q are constants, and ξ is normalized white noise. The only difference between these equations and

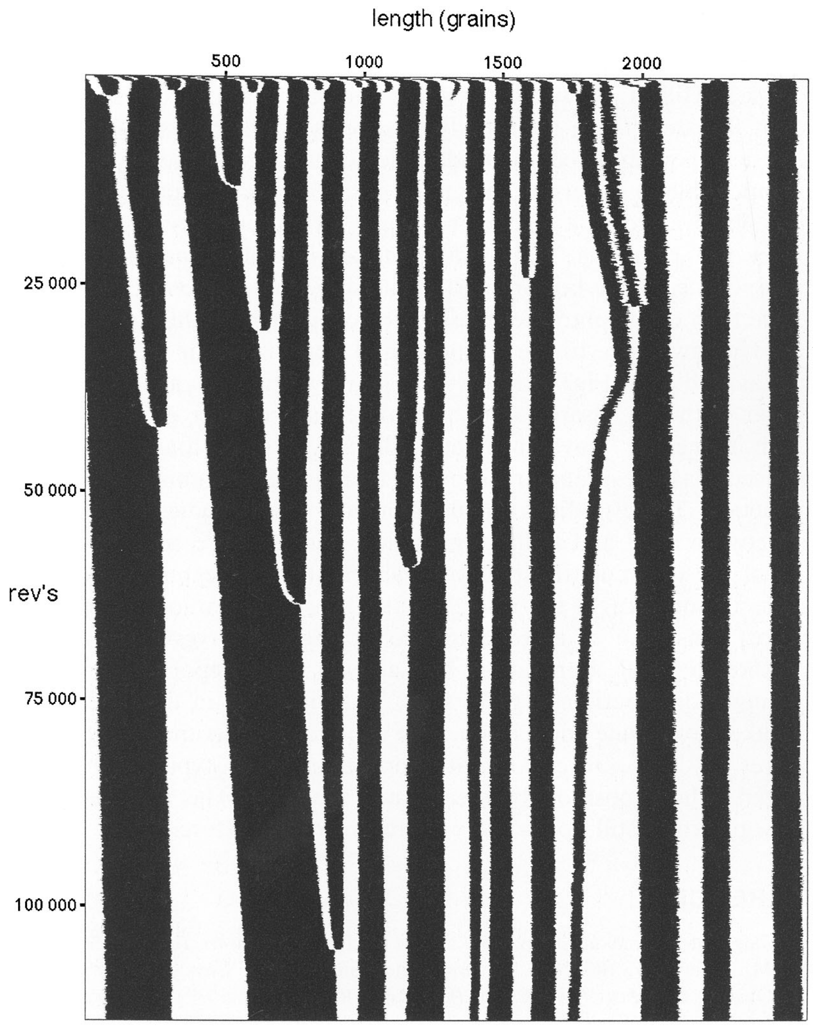

Fig. 4. Spacetime diagram of band pattern evolution in a sand (black) glass bead (white) mixture. Notice the large number of revolution in this run, which lasted for 123 hours (from ref. 10).

that of Zik *et al.* is the presence of the noise term. Levitan solved Eqs. (2) and (3) numerically, and obtained a logarithmic decrease of bands followed by saturation at very long times, in qualitative agreement with the behavior observed in the experiments.

In the works reviewed so far, it has been implicitely assumed that all the segregation process takes place in the thin fluidized layer of grains induced by the rotation, and that the composition in a band cross section is homogeneous. This assumption has been challenged by the recent findings of Hill *et al.*, who have made magnetic resonance imaging of the interior of bands.[12] In their study of spheres of different sizes and surface properties, they observed a considerable degree of radial segregation which was impossible to discern from surface observations. In fact, radial segregation is observed to be a precursor of band formation in many experiments. After a few rotations, large grains in an initially nearly-homogeneous mixture cluster at the bottom part of the slope due to their larger mobility, and are then drawn into the interior, to reappear later during the rotation cycle. This gives rise to the striated band appearance in the right part of Fig. 3 as well as in Fig. 10 of ref. 10. The magnetic resonance experiments, together with the disappearance of axial composition gradients at low enough frequency[7] have prompted Hill *et al.* to claim that radial segregation occurs as the granular assembly attempts to minimize its degree of compaction. These findings should motivate future studies of the connection between axial and radial segregation. Furthermore, a more thorough study of the different transport mechanisms behind segregation and their relative importance is in order. Finally, the problem of segregation in mixtures consisting of more than two powders deserves more attention, even though axial segregation has already been reported in mixtures involving a distribution of grain sizes.[13] A first step in this direction has been taken by Levine and Lipson, who conducted experiments with ternary mixtures.[14] These revealed that more than three types of bands are observed, whose positional order is not arbitrary. The rich behavior of axial segregation still holds many more surprises in store.

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