

NONEQUILIBRIUM PATTERNS IN GRANULAR MIXING AND SEGREGATION

For over 5000 years, granular mixing has been a topic of acutely practical concern. Paleolithic cave painters mixed their colors from blends of ochre and animal products; ancient Chinese and Egyptians blended inks and cosmetics from pork soot, crushed pearls, and compounds of lead; Aztec priests prepared drugs from concoctions of herbs and roots; and Michelangelo pigmented the Sistine chapel frescoes with blends including chalk, charcoal, and lead.¹

In the modern world, blending of granular materials remains of great importance. It is an unavoidable step in most technical endeavors, from the construction of houses and roads to the manufacture of pharmaceuticals, foods, ceramics, glass, explosives, metals, and most other mass-produced consumer goods. The magnitude of applications involving granular processes is huge: US production presently accounts for over a trillion kilograms each year of granular pharmaceuticals, foods, and bulk chemicals.² Despite its importance, however, our understanding of granular processes is limited. As a result, factories that rely on powder handling require much longer start-up times than are typical of plants involving only fluids, and oftentimes facilities intended for powder production are abandoned after the expense of many millions of dollars for want of an effective means of inducing powder flow or blending.³

Over the past decade, granular systems have attracted significant attention from both physicists and engineers, working in a variety of “sandboxes” (see the article by Heinrich Jaeger, Sidney Nagel, and Robert Behringer in *PHYSICS TODAY*, April 1996, page 32). Quantitative studies of mixing and segregation patterns in practical tumblers have only recently begun, and have already revealed rich and surprisingly varied dynamics.⁴ However, most analytic work is highly simplified—with some important exceptions, the sandbox is two-dimensional, and the sand is spherical and of uniform size. These simplifications are a useful step toward establishing a beachhead into the field, but we still have far to go before we

Granular flows that mix different species exhibit a surprisingly diverse repertoire of striking and beautiful behaviors. Better understanding of the mixing process should help in predicting whether a given flow will mix—or segregate—its constituents.

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arching, among others, but there is currently no general theory that shows how these many mechanisms are related or under which conditions one or another mechanism will dominate (see box 1). Even the way in which powders are loaded into blenders of common design can alter the time needed to homogenize them by as much as two orders of magnitude.⁶ And, arguably of the greatest practical concern, even if a certain blender delivers acceptable performance in the laboratory, we have no consistent procedure to scale the process up and achieve the same performance in blenders of industrial size.

This article highlights a small subset of the striking and beautiful behaviors that are observed in granular mixing flows of direct practical relevance, and speculates briefly on the mechanisms believed to control them.

Mixing of similar particles

Let us begin by considering the simplest problem, the mixing of similar grains. In a typical application, several granular species are sequentially loaded into a device where they are caused to flow, with the aim of generating a blend that is “homogeneous” for sampling scales larger than a specified minimum volume (such as the size of a pharmaceutical tablet). Such devices are usually of one of two types: convective blenders, where the vessel is fixed and flow is induced by moving an internal agitator, and tumblers, where the entire vessel is rotated and the powder flow is driven by gravity. Here we will focus on tumblers, and we begin by considering relatively large particles ($\geq 300 \mu\text{m}$).

Mixing in tumbling blenders consists of a very fast convective stage, driven by the mean velocity of many particles, followed by a much slower diffusive (or dispersive) stage, driven by the velocity fluctuations of individual particles.⁷ For a given blending process, the relative importance of each mechanism is determined by where each species is initially placed in the mixer.

Convection is by far the faster and more efficient mixing mechanism in grains (as in fluids), but convective mix-

can begin to address most problems of practical importance.

Thus, as this article is written, we do not know how to predict *a priori* whether two powders will mix or segregate when stirred together in a given blender. Numerous mechanisms for segregation of dissimilar grains have been cataloged,⁵ including percolation, convection, inertia, ordered settling, and

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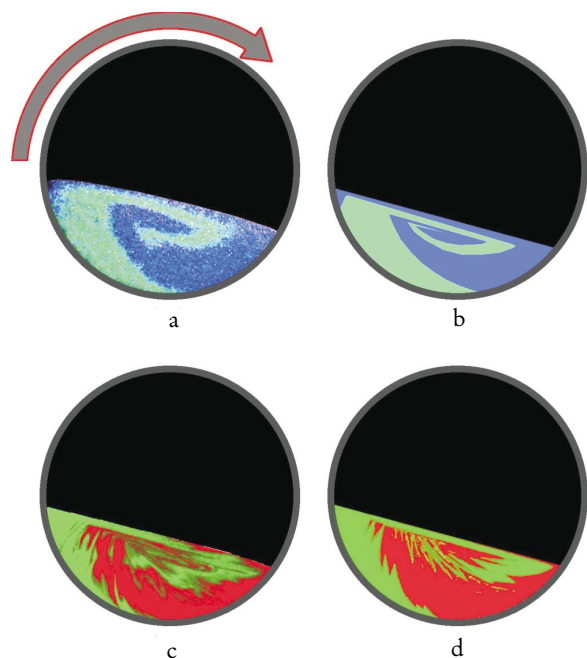


FIGURE 1. PATTERNS FORMED BY MIXING IDENTICAL GRAINS in a cylindrical tumbler can be seen in snapshots of the axial cross sections of the tumbler. On the left are experimental photographs, and on the right are results of theoretical models. For the experiment, equal amounts of differently colored but otherwise identical grains were placed on opposite sides of a plane passing through the drum axis and the drum was turned through one complete revolution. The granular bed was then infiltrated with a polymeric solution, the solution was allowed to set, and the drum was sliced open at the axial midpoint and photographed. For coarse grains (initially separated with blue on the left and blue-green on the right), the mixing pattern is seen in (a) an experimental snapshot and (b) the results of a continuum model. The model assumes that above a thin parabolic interface near the free surface, grains flow downhill with uniform velocity; below the interface, grains rotate uniformly with the drum; and at the interface, velocities change discontinuously but mass is conserved. For fine grains (initially separated with red on the left and green on the right), the mixing pattern is seen in (c) an experimental snapshot and (d) the results of a continuum model similar to (b), but where the parabolic interface was periodically distorted to mimic slow downhill stick cycles interleaved with rapid uphill slip events.

ing suffers from the same limitation with grains as with fluids: the development of barriers to mixing, such as islands that do not interact with surrounding material. Although little is currently known about the phase space of flows in realistic blenders, two pathologies are readily observed: Overfilled mixers develop elliptic, nonchaotic islands (termed “cores”), that rotate as a unit in the center of the granular bed, and symmetric blenders (seen in most standard designs) exhibit separatrices that divide the flow into noninteracting sectors. Industrial practice typically attempts to solve the problems of convective mixing by attacking the symptoms. Segregated regions are avoided by decreasing the blender fill level (which also reduces operational efficiency), and the effects of separatrices are alleviated by carefully loading all components into each sector in equal parts. Unfortunately, these ad hoc “solutions” do not address the underlying cause of the problem, which can only be found by understanding the way the grains mix in a given blender. The result is often a mixing process vulnerable to small changes in material properties or operating conditions.

Diffusion is much slower than convection, but it occurs in all directions and lacks the barriers seen with convection. Thus, in the absence of segregational tendencies between dissimilar particles (discussed below), diffusion will eventually lead to a completely homogeneous mixture. When diffusion dominates, the mixing problem is reduced to that of finding geometric and operational parameters that minimize the blending time. Currently, however, it is impossible to perform even such a conceptually simple task as optimizing blender design from first principles.

Mixing of particles smaller than about $300\ \mu\text{m}$ is complicated by the tendency of the particles to stick together, a property loosely referred to as “cohesion.” For such small particles, the intrinsic interparticle cohesive forces (believed to be a combination of van der Waals forces and surface tension of adsorbed water layers) become comparable with the particle weights, and particles can stick to one another in relatively rigid aggregates. This phenome-

non is unlike anything seen in fluids. Unless the aggregates are destroyed, the system will behave as if it had an effective particle size much larger than the primary particle size. To homogenize such materials at the most intimate scale, it is often necessary to introduce shear (strong velocity gradients) to break the aggregates apart—for example, by using rapidly spinning impellers. Cohesion has an enormous importance for both mixing and segregation, yet at the present time an effective technique for quantifying this property in a meaningful way is not available.

Flow regimes

A surprisingly diverse repertoire of behaviors has been observed in granular mixers. The surface granular flow can be steady, forced-periodic, spontaneously periodic, or aperiodic, depending on such factors as the vessel geometry, size, and speed, as well as the properties of the grains.^{4,8–10} One example of a mixer is the horizontal drum, the simplest and one of the most common of all tumblers. Several distinct flow regimes have been observed in the horizontal drum, including, in order of increasing rotation speed,⁷ avalanching, rolling, cataracting, and centrifuging. Most mixing applications operate in the rolling regime, which for large particles is characterized by a smooth, steady flow with a nearly flat inclined surface. Granular flow measurements obtained using magnetic resonance imaging (MRI) by Eiichi Fukushima and others¹¹ indicate that, in the rolling regime, flow in a radial-azimuthal slice through the drum occurs in two distinct regions. In a thin layer near the top of the bed, particles follow nearly parallel downhill trajectories. Beneath this “cascading” region, particles are largely interlocked and experience solid body rotation with the tumbler. The two regions are separated by a thin shear band.¹²

Cross-sectional mixing patterns formed by identical grains tumbled in a horizontal drum are shown in figure 1, where observations are compared with predictions from a computer simulation. For coarse grains, the pattern after a single vessel rotation (figure 1a) compares favor-

ably with results from a simulation (figure 1b) that assumes the shear band to be parabolic in shape and the flow in the surface layer to be uniform with depth. (The simulation is available on line at <http://sol.rutgers.edu/~shinbrot/Marwan/RollingApplet.html>.) The simulation, based on an approach originally developed by Devang Khakhar at Indian Institute of Technology, Mumbai (formerly Bombay), uses one of the few examples of an analytic solution for granular flow, in this case in the rolling flow regime. The analytic solution relies on the continuum hypothesis, which assumes that the grains are vanishingly small. A similar continuum model for granular flow developed by Stuart Savage (McGill University) and Kolumban Hutter (University of Darmstadt) has recently been extended in elegant computations by Nico Gray and collaborators⁸ to generate solutions for a variety of flow regimes in the horizontal tumbler.

As the vessel rotates, the two groups of particles, identical except for color, wrap around one another in a spiral pattern. Mixing is slow and regular, and the length of the interface between the two colored particle regions grows no faster than linearly with time. In the axial direction, only diffusive mixing takes place, and hundreds of revolutions are required to blur any initial axial gradients, even for small vessels.

The situation is dramatically different when the particles are smaller and cohesive forces play an important role. Because cohesive forces cause particles to aggregate, we might expect that mixing would be slower for small grains. Actually, however, mixing is enormously faster with small particles, as long as the cohesive forces are not too strong. Furthermore, a much more complex mixing pattern is observed for small particles (figure 1c), involving nested striations that are characteristic of chaotic mixing processes. This evidence of chaos is curious, since two-dimensional steady flows cannot be chaotic: In confined, area-preserving flows such as we consider, it has been known for over a century that particles must follow precisely closed orbits, with completely predictable trajectories. The paradox is resolved by the observation that, for small particles, cohesive forces cause stick-slip oscillations in the flow (recently studied in delicate experiments by Satoru Nasuno, Arshad Kudrolli, and Jerry Gollub¹³) that allow particles to deviate from closed orbits. Thus, the flow is not steady, but oscillatory. A simple model incorporating these oscillations (and taking the shape of the shear layer to be time-dependent, in agreement with experimental observations) leads to verifiably chaotic

mixing⁴ (figure 1d). The stick-slip model predicts exponentially rapid mixing throughout the drum cross section, resulting in mixing times many orders of magnitude shorter than those predicted by a coarse-grain model. Drums with noncircular cross section have also been constructed that show reductions in mixing time compared to circular drums. In this case, the drum asymmetry causes a periodic modulation of the geometry of the flowing layer that drives a time dependence in the flow.¹⁰

For both coarse and fine grains, mixing along the drum axis remains diffusive and slow. This problem can be mitigated by imparting an axial rocking motion to the drum, thus generating axial convective flow.⁹ In our laboratory, this stratagem has increased the axial mixing rate by orders of magnitude. However, such improvements depend strongly and nonmonotonically on the rocking frequency, with the greatest improvements being achieved when the rocking and rotating frequencies are incommensurate. In industrial practice, vessels are often too large to be easily rocked, and equipment manufacturers have resorted to other, intuitive means of achieving three-dimensional convection, such as baffles or asymmetric blender geometry.

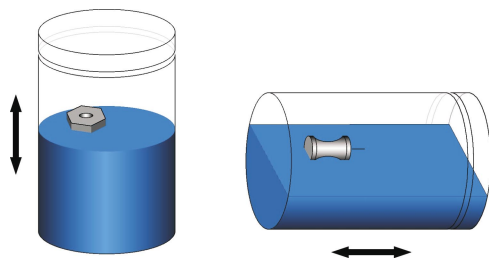
Segregation of dissimilar particles

So far, we have only discussed mixing of similar particles. In the real world, however, common applications involve processing of particles with nonuniform sizes, densities, shapes, and surface properties. Such systems should be assumed to segregate whenever they are shaken, tumbled, stirred, poured, or conveyed. We are not referring to subtle effects here: In many cases, particles with nonuniform properties separate almost entirely in a matter of seconds. As a household demonstration of this process (another example can be found in box 1), we invite the reader to tap a can of bread crumbs repeatedly on the kitchen counter. Larger crumbs will be seen to rapidly rise to the surface. Furthermore, if the crumbs are carefully poured out into a heap, the finer particles will accumulate near the center of the heap. Such a phenomenon can have serious practical implications: If the particles of different sizes also have different chemical composition—a very common industrial situation—the rapid segregation can compromise the quality of the ultimate industrial product.

Segregation processes have been studied primarily in three situations: vibrated beds, filling and emptying of vessels, and rotating cylindrical drums. To date, there is no unified framework for understanding segregation, and

Box 1: The Hex Nut Effect

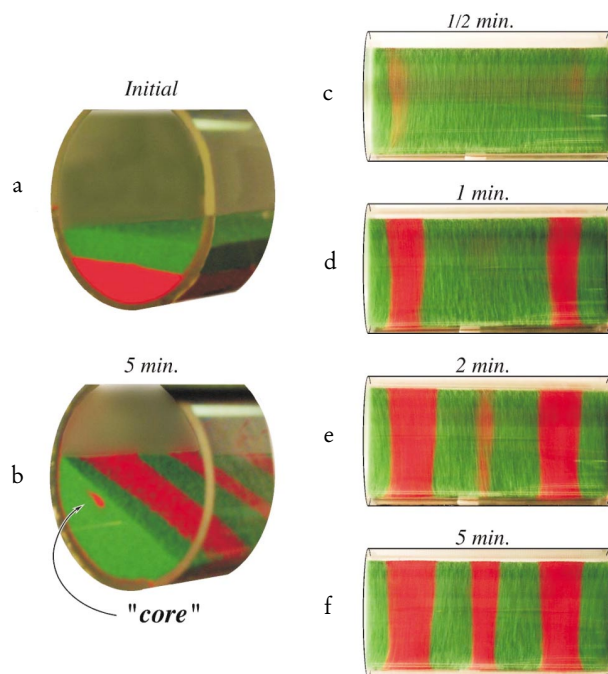
The prototypical example of granular segregation, termed the “Brazil nut effect,” can be easily re-created—and challenged—in any household. The figure illustrates the effect: Take a small spice jar and partially fill it with salt, then add a large heavy intruder (such as a steel hex nut) and an equally large but light intruder (such as a pushpin). If the jar is shaken vertically (left), the nut will rapidly rise to the surface, while the pin will sink; but if the jar is shaken horizontally (right), the pin will rise and the nut will sink.



sinking of the pushpin and of the reversal of the intruders' fates in a horizontal bed remain conjectural at best.

A simpler experiment could scarcely be devised, yet the paradoxical outcome reveals the complexity that one can find in even the simplest of granular problems—and that can conspire to make the more complex industrial granular systems appear to be utterly intractable. The cause of the buoyancy of large intruders such as the hex nut in vertically vibrated granular systems has been described by a number of competing models; the causes of the

FIGURE 2. BANDING SEGREGATION IN A ROTATING CYLINDRICAL DRUM partially filled with 1.4 mm green and 0.8 mm red glass beads. (a) Side view showing initial loading pattern, with large beads above small; (b) side view after 5 minutes of tumbling at 20 rpm. Notice the core of fine, red grains at the end wall; this core extends as an unbroken channel across the length of the tumbler (see text). (c–f) Time sequence of top views after approximately 1/2, 1, 2, and 5 minutes of continuous tumbling, showing emergence of axial segregation “bands.”



the topic consequently remains controversial in all of these environments. Attempts have been made to predict segregation from a thermodynamic viewpoint,¹⁴ although granular assemblies are quenched systems very far from equilibrium, and the granular “temperature” (customarily defined in terms of the rms fluctuational velocity of grains) is typically close to zero. Computational approaches have been pursued by numerous researchers—for example, particle dynamics simulations have been used (see box 2) to decipher the roles of the various processes in practical mixers. A variety of segregation mechanisms have been investigated. For example, in vibrated beds, three dynamical mechanisms have been proposed: size percolation (in which fine grains infiltrate beneath larger ones), convection/size exclusion (in which large grains are excluded from narrow downwelling convective channels), and cascading segregation (in which larger grains roll more rapidly downhill than smaller ones). Usually, all three—and probably several more—are simultaneously present, the prominence of each contribution depending on the specific situation.

In drum tumblers, size-based segregation is readily produced by loading the drums with both large and small grains. Figure 2 shows the development of segregation patterns in a 7.5 cm diameter glass drum initially loaded with equal volumes of larger green glass beads (above) and smaller red glass beads (below). Segregation typically occurs in two stages. First, large grains rapidly segregate radially, producing a central core of fine grains surrounded by larger grains; next, grains in the core migrate along the axis of tumbling until the core extends axially for the length of the tumbler. Core formation is seen in both quasi-two-dimensional and fully three-dimensional blenders of various geometries. In a long tumbling horizontal drum loaded at one end with fine grains and at the other with larger ones, the fine grains will infiltrate along the axis until they emerge as a core at the far end of the drum from within the bed of larger grains (figure 2b).

Axial transport in the core has been studied using MRI,¹⁵ revealing that the axial core bulges outward (in a manner reminiscent of the Rayleigh capillary instability seen in immiscible fluids), and that bands of fine particles progressively emerge at the surface, first near the side walls (figure 2b), and later at the center of the drum (figure 2c through 2f). As time progresses, the bands become larger and better defined, until finally the two components reach a stable pattern, typically with a single band of fine grains. In this final state, two pure phases of material are formed, divided by sharp boundaries with very little intermixing—suggesting a natural analogy with the phenomenon of spinodal decomposition, where a mixture of immiscible fluids separates, in a well-characterized manner, by forming pure droplets that coalesce and grow.

The mechanism for the initial stage of radial segrega-

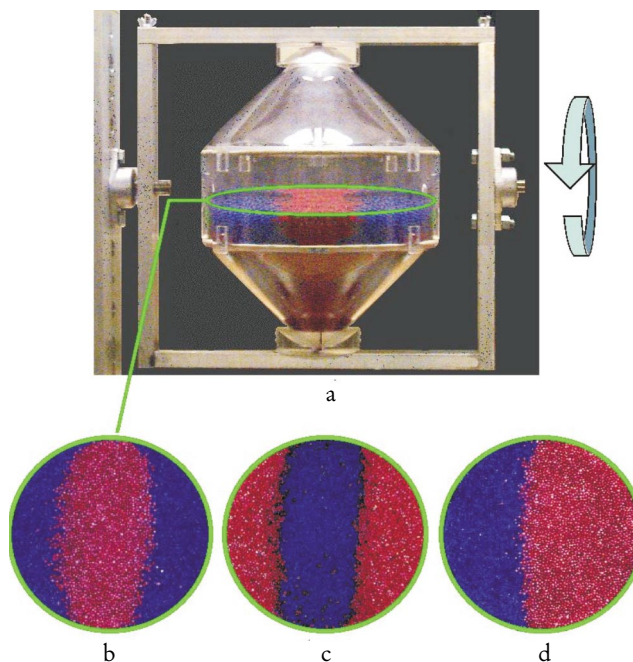
tion was proposed as early as 1976 to be percolation (described above), although more recent work by Souma-va Das Gupta, Khakhar, and Suresh K. Bhatia at the Indian Institute of Technology and by Kimberly Hill and James Kakalios at the University of Minnesota indicates that the greater tendency of larger grains to cascade down the free surface may be at least equally important. Still more recently, observations of traveling concentration waves in certain granular mixtures by Kiam Choo, Tim Molteno, and Stephen Morris⁴ have made manifest the remarkable complexity of segregation dynamics. Clearly, establishing the details of the segregation process in tumbling drums remains an open issue.

Three-dimensional tumblers

Having described flows in simple, axially symmetric drums, let us turn to a fully three-dimensional rotating blender of direct practical relevance, the so-called double-cone blender, depicted in figure 3. In this blender, as in the drum blender, fine particles exhibit stick-slip motion near the free surface, resulting in intricate mixing patterns (figure 4) similar to those displayed in figure 1c and 1d. Although fully three-dimensional, the double-cone blender exhibits fast convective mixing in the radial-azimuthal plane and slow diffusive mixing along the axis of rotation, like the two-dimensional tumblers discussed earlier. Moreover, the system has a plane of reflection symmetry perpendicular to the rotation axis, through which convective flow is exactly null, so that mixing between the left and right halves of the mixer is strictly diffusive. In this design, engineers’ love of symmetry has served us poorly: The flow separatrix results in a time scale for global homogenization that can be 1000 revolutions or longer, depending on vessel size.

For segregating particles, the double-cone blender exhibits several unique—and largely unexplained—behaviors. For fill levels above 25% of tumbler volume, the particles segregate rapidly, typically with large particles populating the center of the vessel and small particles accumulating on both sides (figure 3a and 3b). This pattern is exceedingly robust, and is easily reproduced for

FIGURE 3. SEGREGATION IN THE DOUBLE-CONE BLENDER can be total, and depends sensitively on the mixing parameters. (a) Geometry of the blender, consisting of two cones separated by a cylindrical midsection, and filled here with 4 mm red and 1.4 mm blue glass beads. (b–d) Top views of three segregation patterns that are observed with the beads. As described in the text, the patterns change from one state to another abruptly and reproducibly with only slight changes in the fill level or rotation speed.



other initial placements of components. However, if the fill level is decreased by as little as one percent (or if the rotation speed is changed by an equivalent amount), the pattern inverts: Small particles accumulate in the center, and large particles migrate outwards (figure 3c). In both cases, segregation is complete; essentially 100% separation effectiveness is produced by this “mixer.”

The boundary in parameter space between the two patterns is extremely sharp and reproducible: When the transition is induced by changing the rotation speed, the system can be repeatedly driven from one pattern to the other by introducing at most a few percent change in rotation speed. Intriguingly, the system also displays some of the classic symptoms of a phase transition. For example, at parameter values near the transition between patterns, critical slowing down is observed; that is, the time needed to cross over from one pattern to the other increases sharply.

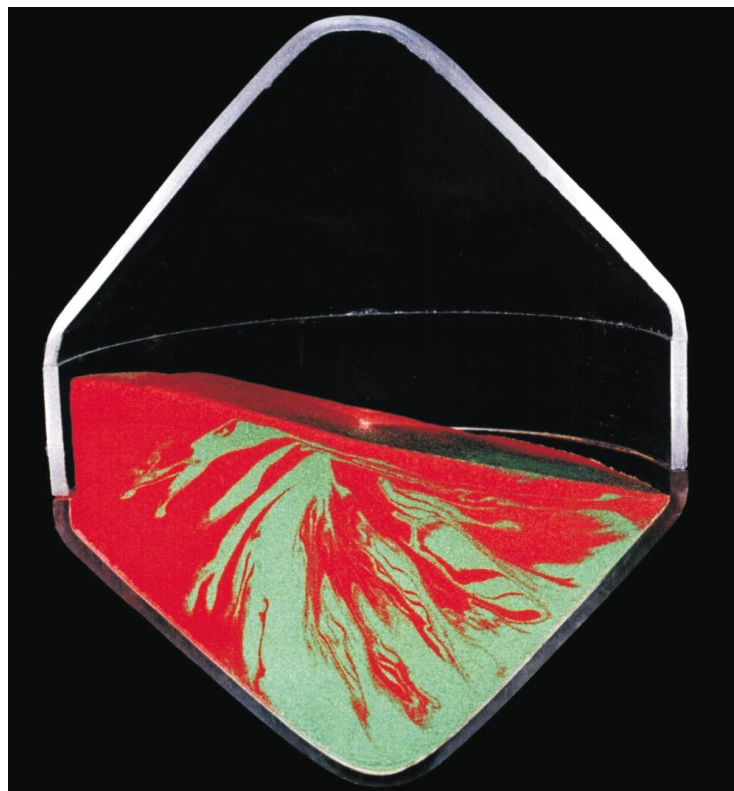
In a small region in parameter space near the transition between the two patterned states, an even more striking behavior is observed: The system displays spontaneous symmetry breaking. All large particles migrate to one half of the blender (chosen seemingly at random), and essentially all small particles migrate to the other half. This behavior, shown in figure 3d, has been reproduced in other blender designs and persists despite meticulous efforts to ensure that the blender is not tilted. The asymmetric state is strongly hysteretic: Once adopted, particles do not revert to the centrally symmetric pattern without dramatic parameter variations.

The few cases discussed here are the tip of a very large iceberg. A great many novel segregation behaviors are easily generated when we consider particles varying not only in size, but also in density and shape. The different particle properties can have coupled effects, so that a given ensemble

of particles can be well mixed for certain flow parameters and completely separated for other flow parameters.

Developing models to predict the behavior of granular flows is essential if we are to gain control of mixing and segregation and thus avoid many of the problems that plague modern industry. Predictive models could also contribute to the development of new technologies, such as blending of nano-composites—now an entirely unexplored field. Even for large, simple grains, much work remains to be done before we understand the relationship between a

FIGURE 4. CHAOTIC MIXING PATTERNS IN THREE DIMENSIONS result from blending otherwise identical red and green fine grains in a double-cone tumbler. With fine grains (smaller than about $300\ \mu\text{m}$), cohesive interparticle forces cause a stick-slip flow that leads to chaotic mixing. Initially, all the red grains were located in the upstream (left) half of the blender and all the green grains in the downstream (right) half. The tumbler was then rotated clockwise through 1 revolution at 5 rpm, and the blended grains were infiltrated with a polymer solution. When the polymer had set, the tumbler was sliced open at the symmetry plane perpendicular to the rotation axis, revealing the intricate mixing patterns shown.

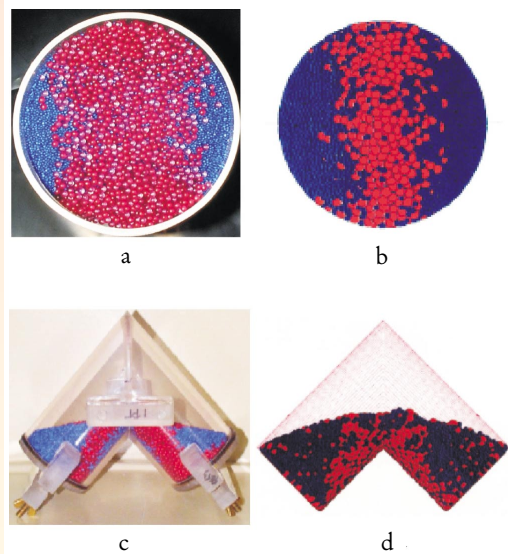


Box 2: Particle Dynamics Simulations

Analytic models for fluid flows are possible partly because the response of simple fluids to stress can be spatially uniform and history independent. Granular responses to stress, on the other hand, are both strongly nonuniform and history dependent, except in special cases, like uniform rapid flows. (For an excellent review, see Leo P. Kadanoff in reference 12.) For example, grains in a slowly revolving tumbler will rotate with the tumbler until a critical stress is exceeded, after which a surface layer of grains will avalanche downhill. The behavior of the avalanching grains is necessarily very different from the solid bed beneath, and the situation is complicated still further by the existence of a glassy shear layer between them,¹² so obtaining differential equations generally applicable to all granular regimes seems impossible.

Lacking a general set of differential equations for granular flows, researchers resort to particle dynamics simulations. These simulations, in close analogy to molecular dynamics simulations, model the forces and reactions between multiple individual particles, typically using nonlinear and history-dependent equations to account for complex interactions like those giving rise to stick-slip motion. As the figure shows, the simulations have been reasonably successful in modeling complex flows. Shown are comparisons between experiment and simulation for two common industrial tumbler geometries: the double-cone

blender (top view), from (a) experiment and (b) simulation; and the "V-blender" (front view), from (c) experiment and (d) simulation.



Although respectable agreement with experiment is attainable, simulations of this kind have several limitations. The number of particles that can be simulated is no more than about 10^4 , while practical mixing devices commonly use well over 10^6 grains. Moreover, although simulations can generate enormous amounts of data, making sense of the data and developing useful, predictive models is at best a long-term goal. Questions also remain about the effects of complex particle properties like particle shape,¹⁶ and there are concerns about numerical reliability.¹⁷

Several teams are currently exploring particle dynamics simulations of granular mixing¹⁸ and flow, and many of these have Web sites. Contributions from many research groups are summarized in <http://www.granular.com>. Some good Web

sites with simulations by these groups include:

<http://chaos.ph.utexas.edu/research/granular/granular.html>
<http://mrsec.uchicago.edu/granular/>
http://sol.rutgers.edu/~shinbrot/Group_Index.html
<http://summa.physik.hu-berlin.de/~kies/>
<http://www.haverford.edu/physics-astro/Gollub/lab.html>
<http://www.ica1.uni-stuttgart.de/>
<http://www.phy.duke.edu/~bob/>.

particle's size, density, shape, and velocity in a given granular flow, and the resulting mixing behavior and segregation patterns. These questions, and many others, deserve further examination.

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