Modeling Granular Materials: A Test Bed for Framing and Analysis

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Neal Amundson's seminal contributions in chemical engineering stem from the mathematically framed elegance of his research and the consequences of rigorous analysis. However, many significant problems in chemical engineering have resisted couching in elegant formalisms, the flow of granular materials being a particularly important example. There are valuable lessons that come sharply into focus by examining the nature of the difficulties. Here, we focus on questions of framing and analysis—determining a theoretical approach, choosing an appropriate level of description, and selecting from multiple quantitative tools—using examples spanning vibration, mixing, surface flow, and segregation and pattern formation. © 2013 American Institute of Chemical Engineers *AIChE J*, 59: 3237-3246, 2013
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Introduction

This article appears in an issue honoring the memory of Neal Amundson, someone whose influence in chemical engineering is profound and impacts directly or indirectly almost every area of the discipline. But the area we will describe is one where Amundson did not work.

Amundson was a pioneer and an intellectual leader who set the tone for a significant territory of chemical engineering research and education. Part of his influence can be traced back to the air of finality of his work. Finality is associated with timelessness and timelessness with elegance, and Amundson's mathematically framed papers have an unquestionable aura of elegance. It is the elegance aspect that concerns us here, for this has had two inter-related consequences in chemical engineering. The first was people gravitating to elegant subjects, problems that could be framed in mathematical terms. The second was that, everything being equal, people neglected inelegant problems: Cleanliness and elegance were good, messiness was not. Reaction engineering was elegant and so were fluid dynamics and transport, all are topics where a mathematical framework provided a clear and strong foundation. The focus of this brief article is on granular matter, one of the topics that has been left out of the mainstream of chemical engineering, despite its importance in many applications, simply because it was perceived as inelegant.

The objective here is to bring to the forefront questions of framing and analysis: decisions regarding the level of description and caveats constraining the range of solutions. We will present several examples from our own research and that of others to illustrate these two interrelated issues. The first issue is the challenge that lies in picking the appropriate level of description to address a specific question; the choice often being one from among a huge hierarchy of length scales and a range of approaches: continuum, discrete, purely geometrical, with much in between. The second issue, a characteristic of many if not most problems in granular matter, is that solving a problem comes with conditions, often associated with various time and length scales. When one digs deeper one finds more questions.

Granular Materials and the Mainstream of Chemical Engineering

One of us, in a article honoring John Bridgwater's career at Cambridge, commented on the issue that the study of granular materials seemed to have been left out of the

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mainstream of chemical engineering, at least in the United States.[1] The question is why? Parts of the reason were alluded to above: The problems are hard, the complications are many, and a general mathematical framework that spans large swaths of the granular landscape is lacking. In a direct sense, the closest Amundson seems to have come to this subject, at least in terms of tools, is in a paper titled "Analysis of Breakage in Dispersed Phase Systems," with Valentas and Bilous, published in 1966.[2] One of us also made an incursion into this territory—breakage, and its opposite, aggregation—in a series of papers with Shawn Hansen.[3–5] Work in fragmentation theory by Redner[6] also fits in this mold; the math in some of these descriptions is unquestionably elegant.

But the main reason that the study of granular materials has remained on the sidelines—as hinted at above—is that framing dominates analysis. Clearly, one cannot do analysis without framing. But questions in granular materials can almost invariably be framed in more than one way, with multiple viewpoints in terms of scales and tools. There is usually no single approach, so choosing an approach is often the most challenging part of the problem. On the other hand, this richness of possibility gives us the ability to explore viewpoints, a characteristic for which the educational upside is high.

Framing Granular Materials

Granular materials have been studied[7] for two principal reasons: practicality and paradigm. Mechanical engineers, civil engineers, and geologists have sought to understand granular materials for at least two centuries and have uncovered several empirical laws describing their behavior for practical use across a range of industries and applications—geotechnics, ceramics, grains, fertilizers, cosmetics, food products, resins, electronics, polymers, construction materials. Physicists joined in more recently and typically see granular matter as a chameleon-like state of matter that bridges gas, fluid, and solid states, and thus represents an important paradigm for the study of nonequilibrium systems.

Several challenges are immediate. The dissipative nature of the interactions of the constitutive particles of granular matter makes the application of equilibrium thermodynamics highly nontrivial, a key difference being that grains irreversibly lose energy in each collision to unrecoverable "thermal" modes but atoms and molecules do not. Even if the inelasticity of granular collisions is small, it can produce significant effects, such as granular clustering. If a granular system is excited to keep it in motion, for example by shaking, its dynamics can resemble that of gases. But the resemblance is misleading. Theory and experiments show that granular gases do not become homogeneous with time, but instead form dense clusters of nearly stationary particles surrounded by lower density regions of more energetic particles: a particle entering a region of slightly higher density, has more collisions, loses more energy, and is, therefore, less likely to leave the region—see McNamara and Young.[8] The effect is autocatalytic for it increases the local density which in turn increases the likelihood of capturing more particles.

Levels of Description

Describing flowing granular matter presents formidable problems across a number of density regimes, though the dilute regime—where granular matter resembles a gas—seems within reach. At the other extreme, dense flowing granular matter presents myriad open questions. But it is worth noting that even nonflowing granular matter poses serious challenges and the flowing/nonflowing transition even more.

For example, in its solid-like state, granular matter displays traits of self-organization—crystallizing into regular arrays or forming intricate force networks in response to an applied load, which often rearrange dramatically when the load is perturbed. We can imagine several scales of description driven by the underlying questions, and we commented on this in an earlier publication.[9] Granular solids display memory (the structure of a granular network gives clues to how the system was formed) and a stress response that resists traditional categorization (they are neither entirely elastic, nor entirely plastic). At the smallest scale, there are the statistics of single elements—contacts and particles. The central question here is how particles contact one another; this is where network analysis can find fertile ground.[10] At slightly larger scales, the viewpoint can shift to structural motifs, typical arrangements comprising clustered elements. The central question here is how organization among small groups of particles affects granular stability and flow properties. Then, at scales much larger than the grain size, there are questions about network structure and organization, not as the sum or average of its individual parts, but as a connected whole. This includes percolation, a common paradigm for describing jammed material. For example, Silbert and colleagues showed[11] that length scales of dynamical correlations in a granular material near the jamming transition obey percolation-like scaling—a result that links the granular jamming transition with the glass transition in thermodynamic systems.

Tools

Granular materials admit multiple levels of analysis which can be based on discrete and/or continuum descriptions. Discrete approaches include particle dynamics (PD), also known as the discrete element method, Monte Carlo methods (MC), and cellular automata calculations (CA). MC simulations are often too idealized to mimic specific materials; CA computations can yield considerable insight but at the cost of sacrificing specificity. PD methods come close to the ideal of a first-principles approach. The technique is based on the methodology of molecular dynamics developed for the study of liquids and gases, but with some important differences. In the simplest case, when the particles are large (say 100 μm or more), interactions are primarily mechanical. The motion of each particle is governed by Newton's laws; the purpose of PD simulations is to compute the evolution of position and orientation of every particle by using appropriate, though typically quite simple, contact force models. Thus, as opposed to conventional molecular dynamics, torques are transmitted and dissipation plays a crucial role. However, predictive calculations for specific materials with complex shapes are, in general, nearly impossible as "exact" PD simulations require precise physical properties (Young's moduli, restitution coefficients, Poisson ratios, etc.) and complex methods of determining interactions between nonspherical particles. And notwithstanding questions about the nature of the contact force models, many important questions remain even in the case of spherical particles.

First principles-based continuum models have been devised using analogues of the kinetic theory of hard spheres for nearly elastic spheres. The main obstacle in continuum descriptions is the importance of intermediate scales (mesoscales). As Liu and coworkers have shown, manifestations occur in stress chains[12] and jamming.[13] Nevertheless, there is emerging evidence that simplified continuum-based descriptions, with constitutive relations supported by particle dynamics simulations, may form the basis of a general expandable and coherent framework for the description of a variety of flow and segregation processes of granular materials. In general, continuum models work remarkably well, even when we expect that they should not, bringing to mind Eugene Wigner's observation of the "unreasonable effectiveness of mathematics" or, at the very least, what one may term "the unreasonable effectiveness of the continuum hypothesis." For example, lens-like shaped flowing granular layers in laboratory tumbler experiments are often only 5–10 particles deep at their thickest, tapering to 1–2 particles at their thinnest, but continuum ideas work surprisingly well most of the time. A significant advantage of continuum descriptions is that they provide a direct connection with studies based on nonlinear and complex dynamics.

Framing Problems

What are the "basic" properties of granular materials in contrast to fluids? In fluids a few continuum properties like density, viscosity, conductivity, heat capacity, and compressibility, tell us basically all that is needed to analytically describe a wide range of flows without having to know anything about the microscopic shapes or interaction details of the constituent particles. These continuum properties typically come about from/through constitutive relations, which can be phenomenological or derived from first principles. Though this is a bit of an over simplification (let us leave turbulence out of the picture), once a problem has been formulated—the continuum description being well established in terms of Navier-Stokes equations and the like—what remains is analysis, either mathematical or computational. There are open problems in fluids, but certainly not in the same class as those appearing routinely in granular materials. A characteristic of many problems in granular matter is the surprisingly broad range of tools, descriptions, and approaches at our disposal. Framing problems is, therefore, highly nontrivial.

What follows are examples that highlight all the above issues. In one case (vibration) simple physics explains broad aspects of what is a surprisingly rich phase diagram. In another (avalanching) elementary geometric ideas capture 90% of the behavior, if not more, and in a third case (cutting and shuffling mechanisms), geometry captures behavior that bring us in contact with new math. Two examples involving segregation, one radial (in two-dimensions), the other, axial (in what superficially can be envisioned as segregation in one-dimension), raise questions of time scales. There are rough theory-based models to interpret radial (two-dimensional [2-D]) segregation processes, but no widely accepted model to describe axial segregation. Models for both segregation processes fail to capture long time coarsening effects.

Examples

Vibration induced granular wave patterns

What on first viewing appears as remarkable granular self-organizing behavior can be partially explained by elementary physics.

A spherical particle dropped on a horizontal plate bounces predictably until it stops. However, this predictability vanishes when the surface is vertically vibrated, see, for example, the work of Mehta and Luck.[14] Although vibration parameters exist for which the particle bounces periodically, most trajectories are instead chaotic, that is, nonperiodic and sensitive to initial conditions. Adding a second particle makes predicting the positions effectively impossible. However, by adding many more particles, a remarkable transition occurs which causes temporal and spatial fluctuations of positions and velocities to become correlated. Goldman and coworkers found[15] that for sufficient density, collisions of particles in a given region occur at roughly the same time, and periodic variations in the local particle density increase. Above a critical number, the vertical motion of all particles becomes locked and collisions with the container occur nearly simultaneously, in sharp contrast to the motion of a small number of particles. Figure 1 shows that for sufficient peak shaking acceleration, Γ, highly organized horizontal particle motions occur in the form of ordered standing waves of squares, stripes, hexagons, and more exotic patterns.[16]

The dependence of pattern transitions on Γ is surprisingly simple if one focuses on the collective behavior. Like a sand bag or a hacky sack, grains do not bounce when they hit the plate but instead rapidly match the plate velocity, a system for which the behavior is easy to calculate. At low Γ, grains collide with the plate once per cycle. If they rest on the plate for too long before being thrown off, dissipation dominates and no patterns occur. However, if grains collide briefly before being thrown, patterns of squares occur at lower frequencies while pattern of stripes occur at higher frequencies. As Γ is increased the vertical motion of the grains undergoes a period doubling at which both patterns of squares and stripes become hexagons. Further bifurcations in the vertical motion of the grains leads to even more patterns.

Predicting which pattern occurs for a given underlying vertical motion remains an open question, although insight has been provided by Bizon et al. in PD simulations,[17] Shinbrot using CA[18] and Eggers and Riecke as well as Bougie et al. via continuum models.[19,20] In all cases, however, it is clear that the cooperative behavior induced by dissipation induced collisions is vital.

Gravity driven flow

Simple descriptions capture flow in shear layers….but there is more behavior in store if one looks at long time scales.

Granular matter supports shear stresses without flowing due to frictional and steric interactions between particles and, consequently, it flows only when inclined above a maximum "angle of stability." When this angle is exceeded, flow typically occurs in a thin surface layer (5–10 particle diameters deep in laboratory scale devices) whose velocity decreases approximately linearly with depth.[21] Two industrially important geometries with granular surface flows are the rotating tumbler and the heap.

Tumblers typically consist of horizontal cylinders partially filled with particles. For slow rotation, intermittent avalanches occur, while for faster rotation, flow is steady. In both cases, flow is maintained by the recirculation of particles transported to the flowing layer by the rotation of the tumbler. When particles identical except for color are placed in opposite halves of the tumbler and the tumbler rotated,

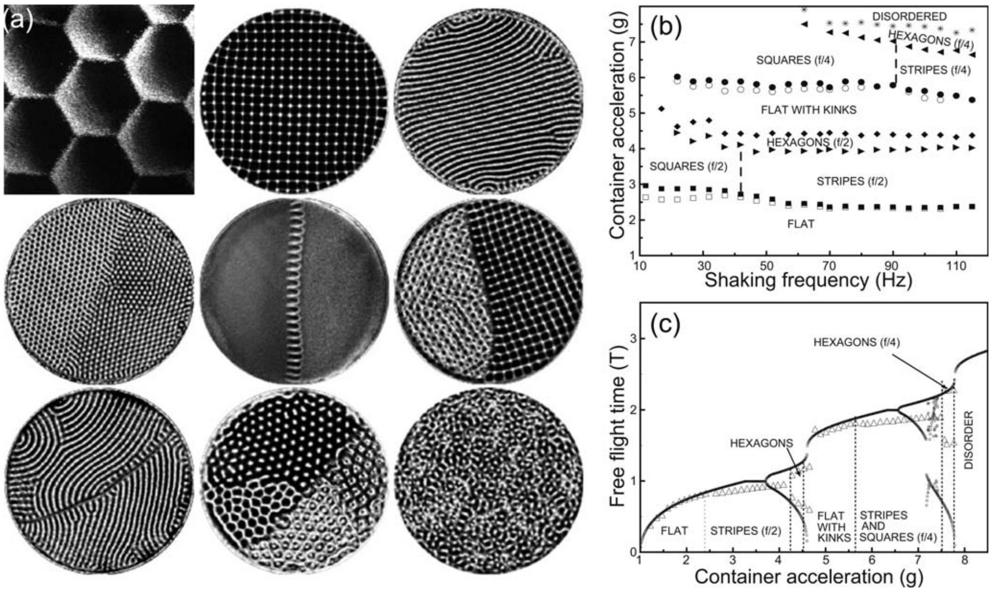

Figure 1. Standing wave patterns in vertically oscillated granular matter.

(a) Patterns in a 7 particle deep layer of 165 μm bronze spheres in an evacuated, 13 cm diameter, rigid container. Upper left image is a close up of a hexagonal pattern while the remaining images show patterns for various shaking frequencies (30–90 Hz) and accelerations (3–9 g's). (b) Pattern bifurcations depend primarily on container acceleration. (c) Layer free flight times from experiment (open symbols) reveal that all pattern transitions (except between squares and stripes) are driven by period doubling bifurcations in the center of mass motion as predicted by a simple model that treats the entire collection of grains as a single completely inelastic particle (filled symbols). Reproduced from Ref 16, with permission from The American Physical Society.

mixing is observed.[22] The character of the mixing depends on the fill level: if it is less than half-full, uniform and rapid mixing occurs within a few rotations, but when it is greater than half-full an unmixed core appears whose size increases with fill level, see Figures 2a,b. The core is a direct consequence of the localized surface flow since particles in the core never enter the flowing layer and thus do not mix. Closer inspection reveals unexpected dynamics and very slow mixing. Rotating the tumbler hundreds to thousands of times reveals that the core is not static but rotates slightly faster than the tumbler and decreases in size,[23] providing evidence of a slow creeping region below the flowing layer.

Heap flow is produced by pouring particles in a pile and is often studied in a restricted quasi-2-D geometry by forming the heap between parallel vertical walls with a finite base. With open end walls, the heap size remains constant and flow is similar to that in a tumbler, flowing intermittently at low fill rates and continuously at higher fill rates. Figure 2c shows that the flowing layer is thin when captured in a 1 second exposure. However, as Komatsu et al. discovered,[24] longer exposures (Figure 2d) reveal that deeper particles move as well, in agreement with the core precession and erosion observed in tumbler mixing. In this creeping layer, velocity decreases exponentially with depth with a characteristic length scale of a grain diameter and grain motion is discontinuous with particles moving in fits and starts whose probability decreases with depth.[24] These intermittent localized motions are likely related to nonlocal perturbations (see for example, Kamrin and Koval[25] and Reddy et al.[26]) from collisions in the flowing layer transmitted via networks of force chains (Liu et al.[12]).

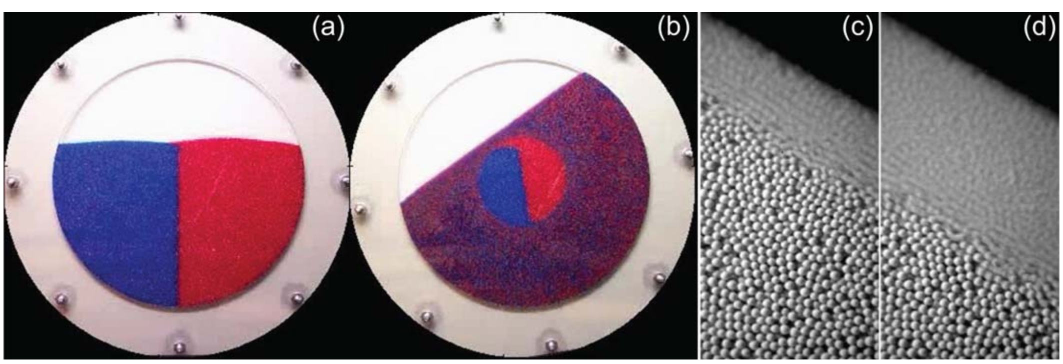

Figure 2. Visualizing the creeping region of the granular flowing layer.

(a,b) Grains identical except for color are mixed in a tumbler. The unmixed core of particles in (b) slowly rotates counterclockwise in the tumbler frame and erodes via diffusion at the boundary with the mixed region. Reproduced from Ref 23, with permission from The American Physical Society. (c,d) Continuous grain flow down a chute with (c) 1 s exposure and (d) 1 h exposure from Komatsu et al.[24] Grains that appear stationary in the 1 s exposure move downslope in the longer exposure. Reproduced from Ref 24, with permission from The American Physical Society. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com].

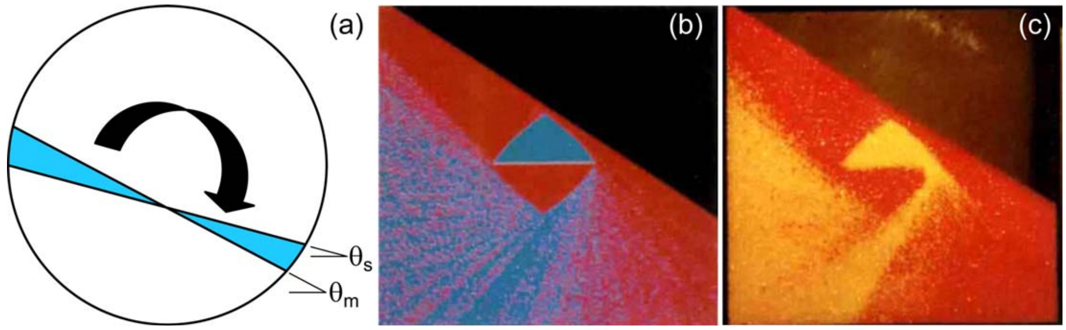

Figure 3. Wedge model of avalanche mixing.

(a) An avalanche can be modeled as particles in the upper wedge moving to the lower wedge where they rearrange randomly. Reproduced from Ref 27, with permission from Wiley Blackwell. (b) Application of wedge model to monodisperse particles starting as red and blue on each side of a partially filled square 2-D tumbler. (c) Equivalent experiments showing the effectiveness of the model. Reproduced from Ref 22, with permission from the Nature Publishing Group. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com].

Cutting and shuffling

In some granular flows, like avalanches, the framework of a geometrical model captures the most important short time behavior. Extending the geometrical model leads to continuum models which capture several aspects of mixing patterns. Further development of the ideas bring us full circle to where mixing can be imagined to occur by what, at first glance, can be viewed as a simpler and, again, geometrical picture: cutting and shuffling.

A remarkably simple model for the study of mixing particles in a slowly rotating cylindrical drum is based on mapping avalanche wedges,[22,27] as shown in Figure 3. While this model totally neglects the details of particle motion, mixing rates based on the wedge model match experiments for monodisperse particles, and side-by-side comparison of the mixing patterns show nearly identical results.

For continuously flowing granular systems, simple geometric models can be used because the flow can be divided into two distinct regions as discussed above: a thin region at the surface (the "flowing layer") in which the particles flow downhill and a larger region below it (the "bulk" or "fixed bed") in which the particles are nominally stationary with respect to the container. In a quasi-2-D tumbler, this simple framework opens a surprisingly rich range of approaches to study mixing, shown in Figure 4. The top row of images shows the initial configuration and the pattern that forms for bidisperse particles (red/black) and monodisperse particles (white/black). This similarity in patterns for monodisperse and bidisperse systems by itself is intriguing given that segregation is bound to play an important role in the second case but is nonexistent in the first (see "Segregation Patterns" below).

The lower rows in Figure 4 shows results for different tools used to model the flow of monodisperse particles: (middle row from left to right) a purely geometric model, the finite-time Lyapunov exponent field, and a Poincaré section (stroboscopic mapping of points advected according to a very simple kinematic continuum model) are Lagrangian and assume a vanishingly thin flowing layer; (bottom row, left) an Eulerian eigenmode analysis including diffusion.[28,29] All four models capture the lobe pattern and the unmixed core at the center of the square tumbler. While the Lagrangian approaches clearly show mixing and even chaos (particularly the Poincaré section), streamline crossing, a necessary condition for chaos, is absent because of the vanishingly thin flowing layer assumption. Instead, what causes mixing (and the apparent chaotic nature of the flow) is a framework known colloquially as "cutting and shuffling,"[30] depicted in the last image of the bottom row. Cutting and shuffling occurs when the tumbler is not half-full. A particle initially on the solid blue streamline when the tumbler is in position A jumps onto the dashed red streamline upon reaching the flowing layer when the labeled corner of the tumbler reaches orientation B. This "streamline jumping" has the same net result as "streamline crossing" in a chaotic system giving rise to a "chaotic sea" punctuated by

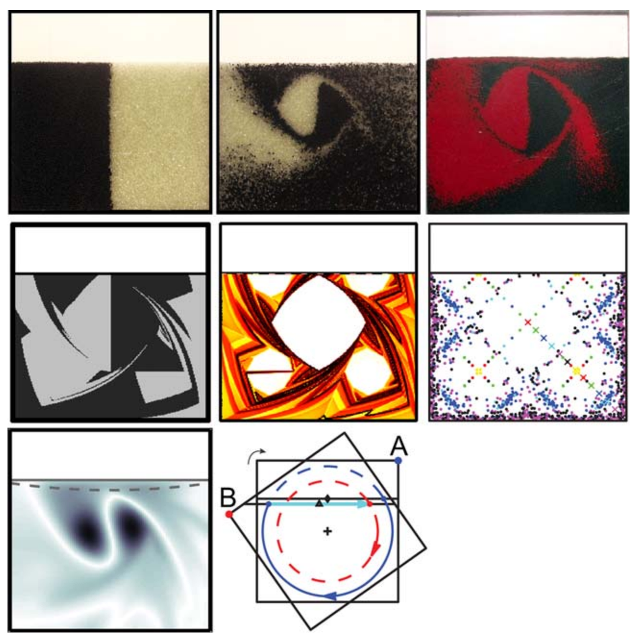

Figure 4. Analytic approaches to granular mixing.

Top row: Initial condition before rotating the 75% full square tumbler as well as the results for experiments with monodisperse particles (white/black) and bidisperse particles (red/black) after 8 quarter-rotations. Bottom two rows: A variety of analytic approaches based on simple geometric or kinematic approaches to studying mixing. Reproduced from Refs 28 and 30, with permission from the American Institute of Physics. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com].

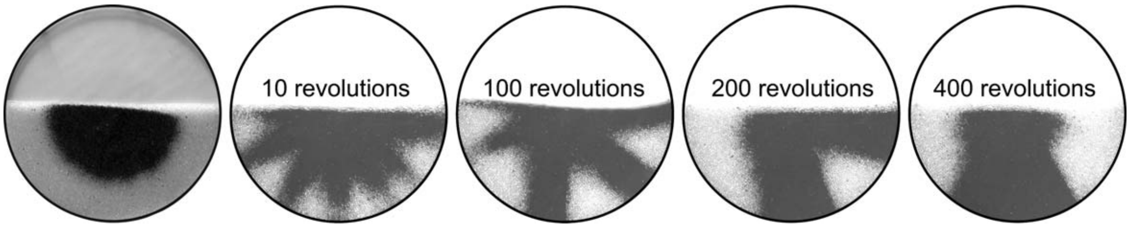

Figure 5. Radial core and streaking segregation.

Left: Radial core segregation for large (1.1 mm) clear and small (0.35 mm) black particles in a nearly half-full quasi-2-D (6 mm thick, 200 mm diameter) circular tumbler. Right four images: Radial streaking segregation and subsequent coarsening for identical particles in a 58%-full tumbler. Reproduced from Refs 42, with permission from Nature Publishing Group.

islands whose locations coincide with symmetries of the tumbler evident in the Poincaré section.

Streamline jumping[31] can be framed in terms of cutting and shuffling dynamics,[32] not unlike mixing a deck of cards. "Shuffling" occurs by mapping each initial position on the flowing layer interface to a new location on the flowing layer at a later time. "Cutting" occurs by reflecting points across the flowing layer. Considerable insight into the cutting and shuffling process can be gained by considering an even simpler system—a line segment. Cut the line segment at several locations, shuffle (reorder) the subsegments, and repeat. For some rearrangement protocols the line segment becomes well mixed; for others, the subsegments initially mix but later reassemble.[33] One can even add diffusion.[34,35]

Segregation patterns

There are ways to rationalize segregating patterns in two dimensions, but the picture gets complicated if one looks at long-term behavior.

One of the practical challenges in handling granular materials is preventing the segregation of different (size, shape, density, etc.) particles subject to excitation (vibration, flow, temperature cycling). At the simplest level of description, size segregation comes about from the percolation of small particles through interstices between large particles and density segregation from buoyancy differences between light and heavy particles. For instance, as small and large particles cascade down the thin flowing layer at the surface in a half-filled quasi-2-D tumbler, smaller particles drift to the bottom of the flowing layer and come to rest in the bed of particles below the flowing layer, while larger particles rise to the surface of the flowing layer so that they travel further downstream before leaving the flowing layer. The result, after only a half rotation or so, is a semicircular pattern of smaller particles surrounded by larger particles, shown in the first panel of Figure 5.

But this segregation mechanism is more complicated than it seems. Fill the tumbler to slightly more than half full and instead of a semicircle, radial streaks of small particles occur within a few rotations, as shown in Figure 5 (see Hill et al.[36–39]; Meier et al.[40–42]). The reason that a particular combination of parameters results in streak formation rather than radial segregation is not completely clear, although it appears to be a result of different streamwise velocities in the flowing layer for the different particle sizes. What is more surprising is that leaving the tumbler rotating overnight (as accidentally happened in our lab) results in coarsening of the radial streaks into a single streak. Again, the exact mechanism is unclear. Coarsening also occurs for particles of different densities (instead of different sizes) and for tumblers of different shapes (square for instance).

The segregation patterns in noncircular tumblers reveal lobes at the interface between the segregated particles, as shown in Figure 6 for a square tumbler. We can frame the problem from a dynamical systems standpoint. Using Poincaré sections, it becomes evident that the character of the stroboscopic mapping matches the segregation pattern, regardless of the fraction of small black particles. What is remarkable is that the Poincaré sections match at all the segregation patterns, given that the kinematic continuum model used to create the Poincaré sections has nothing in it that is even remotely connected to segregation. Thus, by framing the system in terms of a dynamical system, it is clear that the dynamics is dominated by geometrical aspects rather than particle level interactions. Further exploration using the dynamical systems tool of visualizing the unstable manifolds, traced in the right image of the figure, indicates that the segregation pattern is related to the dynamics resulting from the flow geometry, specifically, the directions of stretching around a hyperbolic point in the flow. Since the manifold is an integrated view of the effect of the chaotic regions, it reveals how the pattern changes (how the lobes curve) as the small particle concentration is increased, resulting in the final segregation pattern.

Segregation can even occur in the absence of any obvious flowing layer as Rietz and Stannarius have shown[43] using a quasi-2-D mm square box packed (98% full) with two different size particles and rotated about a horizontal axis through the thin dimension of the tumbler. After O(100,000) rotations, single vortex rolls are observed in the cell plane which result in nonuniform segregation patterns that interact with the rolls, see Figure 7. How such circulation and segregation can even occur in tightly packed systems is unknown.

Axial band formation

Segregation in long tubes leading to a succession of bands of segregated material still eludes a clean explanation, but the picture gets even more complex if one looks at long term behavior. Examining what at first seems to be a simpler case, a system producing only two bands, does not help matters as the bands can alternate positions depending on experimental conditions.

Many years ago in an initially unnoticed paper by Oyama,[44] a curious phenomenon was reported—when particles of two different sizes are tumbled in a half-filled, long, cylindrical tumbler rotating about a horizontal axis, the particles first segregate radially, as expected, but then, curiously, form axial bands formed of particles of different

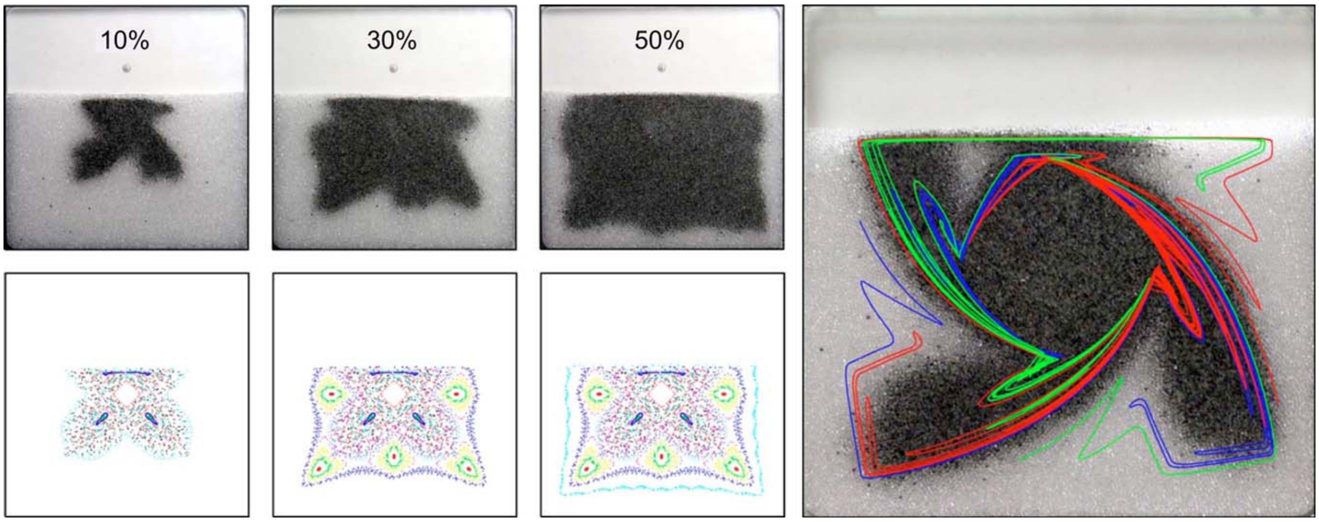

Figure 6. Poincaré sections and unstable manifolds predict radial segregation.

Comparison of segregation experiments (1.21 mm clear glass beads 0.29 mm black glass beads) to Poincaré sections for a 63%-full square tumbler (157 mm on a side and 6 mm thick). Top row: experiments varying small particle concentration in weight percent. Bottom row: Poincaré sections. Right: Unstable manifold analysis for a 75%-full square tumbler overlaid on an experimental image of a mixture with 30% small particles. Reproduced from Ref 40, with permission from The American Physical Society. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com].

sizes. Although this phenomenon of axial segregation occurs in practical engineering processes such as rotary kilns (in fact, none other than Thomas Edison in 1904[45] invented a device to destroy klinker rings resulting from this phenomenon), the physics was not addressed for many decades. Axial segregation occurs much more slowly than radial segregation, typically, in O(100) rotations of the tumbler instead of O(1) rotation. As shown in Figure 8a, the internal structure of the bands consists of a core of small particles that extends all the way to the surface of the flow at fairly regular intervals, though the individual bands of small particles may be separated from one another under certain conditions.[46] At the highest level of description, the bands tend to have an initial characteristic length of about one tumbler diameter,[47] but it is not clear why this should be so. Once

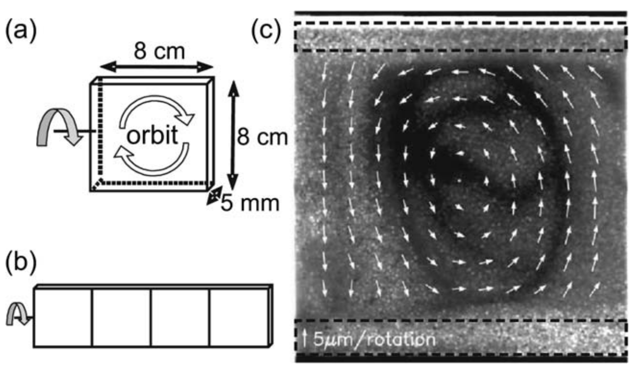

Figure 7. Slow orbiting granular segregation in a 98% full tumbler.

(a) Sketch of the experimental setup of Rietz and Stannarius[43] showing one of two possible directions of the granular circulation. (b) An array of several quasi-2-D containers for simultaneous experiments. (c) Segregation pattern after 107,000 rotations for a 98% full tumbler with a mixture of 0.3 and 0.9 mm particles. Reproduced from Ref 43, with permission from The American Physical Society.

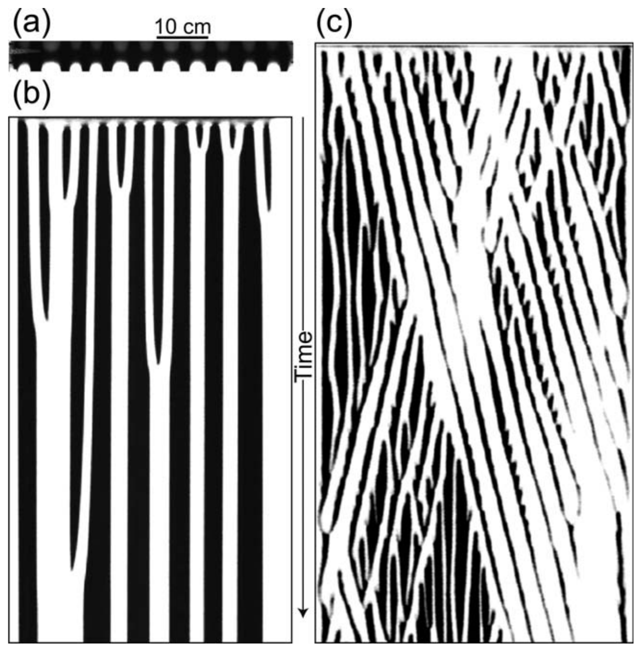

Figure 8. Axial size segregation in long cylinders.

(a) Image of an experimental run cropped to coincide with the top view of the granular bed containing a 2:1 mixture of 0.88 mm clear glass particles and 0.27 mm black glass particles in water. The acrylic tumbler has a diameter of 63.5 mm and a length of 750 mm. Small particles (dark) form a radial core running along the axis with axial bands between bands of larger particles (white and gray). (b) Space–time plot for 2000 tumbler revolutions showing band dynamics including coarsening. Reproduced from Ref 50, with permission from The American Physical Society. (c) Phenomena such as travelling waves can also be observed in space–time plots, in this case a dry granular system with particles of two sizes. Reproduced from Ref 46, with permission from The American Physical Society.

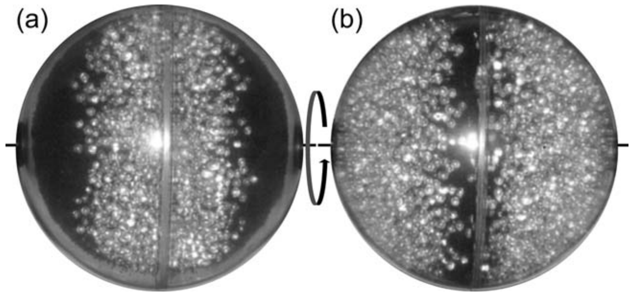

Figure 9. Axial segregation in a spherical tumbler.

Top view of the two surface segregation patterns that occur for 1 mm black and 4 mm clear glass beads in a 14 cm spherical tumbler rotated at 20 rpm. (a) 30% full, (b) 60% full. The narrow vertical stripe on the tumbler is the seam between the two halves of the clear plastic tumbler. Reproduced from Ref 55, with permission from The American Physical Society.

the bands form, they tend to merge after long times (hundreds of rotations), as shown in the space time plot of Figure 8b. Again, why this happens is unclear, though it occurs in both dry and submersed granular systems and depends on the relative fractions of small and large particles as well as the rotational speed.[48] Under some operating conditions, bands may form and then disintegrate or move along the axis of the cylinder as travelling waves (see Hill et al.[36]; Choo et al.[49]; Fiedor and Ottino[50]), as shown in Figure 8c.

The mechanism for the formation of axial bands is puzzling—most explanations are based on differences in the angles of repose of the two different particle types or of phases of mixed or pure particle types (Hill and Kakalios[36]). However, delving into a more fundamental level of description, it has become clear that the bands near the endwalls are directly influenced by friction at the endwalls. Small and large particles flow away from the endwall in the upstream portion of the flowing layer near the endwall due to friction slowing particles down combined with mass conservation, but more large particles tend to flow back toward the endwall than small particles in the downstream portion, because percolation has carried small particles deeper into the flowing layer. The end result is a band of larger particles near the endwalls with an adjacent band of smaller particles[51,52] as is evident in the space-time plot of Figure 8. Of course, this does not explain the appearance of bands far from the endwalls. In fact, bands will form in simulations of tumblers with no endwalls (periodic boundary conditions), though they do not form as quickly or easily.[52] The mechanism is unknown. Systems of same-size heavy and light particles segregate in the flowing layer in a similar way to particles of different sizes (with heavy/small particles falling to the bottom of the flowing layer and light/large particles rising to the top). Based on this, one would expect that, analogous to a size system, bands of light particles would accumulate at the endwalls, which occurs experimentally (Sanfratello and Fukushima[53]; Pereira et al.[54]). However, surprisingly, bands never form away from tumbler endwalls as occurs with particles of different sizes.

More unusual is the situation when the container is a partially filled spherical tumbler. In this geometry, three bands always form in the tumbler: a band at the equator with particles of one size and bands at each pole of the other particle size. The odd thing is that small particles are at the poles when the tumbler is less than half full, but large particles are at the poles when the tumbler is more than half full, as shown in Figure 9.[55] An explanation based on competition between sidewall friction, surface sliding, and segregation has been proposed.[56]

Conclusions

It is easy to get lost in the complexity of granular matter. The 125th anniversary issue of *Science* identified the 125 hardest unsolved questions in science. Granular matter was not in the top 25, but the question "Can we develop a general theory of the dynamics of turbulent flows and the motion of granular materials?" made it to the list of the next 100, with the comment that "So far, such 'nonequilibrium systems', defy the tool kit of statistical mechanics, and the failure leaves a gaping hole in physics." Gaping hole indeed. Questions in the set of 100 included "Is ours the only universe?" "When and how did the first stars and galaxies form?" "What is the nature of black holes?" and "What is the nature of gravity?" Pierre de Gennes, Nobel Prize of Physics in 1991 and a master outliner of problems, in an article titled, "Granular Matter: A Tentative View,"[57] challenged readers with several open questions, many of them having to do with problems that to the uninitiated, like calculating the stress on the bottom of sandpile, would appear solved long ago. In fact, open questions abound. Even problems that are regarded as partially solved, like granular gases, are littered with questions, as noted by Goldhirsch and colleagues.[58] There are also the broad conceptual aspects brought up by a granular view. As Richard Feynman pointed out,[59] the most important fact humanity has learned about the natural world is that, at its heart, all matter is composed of particles. "If, in some cataclysm, all of scientific knowledge were to be destroyed, and only one sentence passed on to the next generation of creatures, what statement would contain the most information in the fewest words?" His answer?: "…particles that move around in perpetual motion, attracting each other when they are a little distance apart, but repelling upon being squeezed into one another. In that one sentence…there is an enormous amount of information about the world, if just a little imagination and thinking are applied." And over a century ago Osborne Reynolds, known now in chemical engineering circles mostly by the Reynolds number, proposed a purely granular theory of the ether,[60] a work that started with one of the biggest opening sentences of any scientific work: "By this research it is shown that there is one, and only one, conceivable purely mechanical system capable of accounting for all the physical evidence, as we know it, in the Universe."

Much good has been written on granular matter asking and addressing questions spanning the arch from the deeply scientific to the eminently practical with the overarching theme being that the number of questions greatly outnumber the solutions. Pick any theme in physics, macroscopic irreversibility, the validity of the continuum hypothesis, and granular matter often offers a paradigm, a clear educational example, but also, frequently more questions.

Here, we have taken a somewhat pedagogical view to the complexity inherent in describing granular matter, bringing to the forefront the kinds of questions framed by every modeler: decisions regarding the level of description and caveats constraining the range of the solutions. The first issue is the challenge that lies in picking the appropriate level of description to address a specific question; the choice is often one from among a huge hierarchy of length scales and a range of approaches: continuum, discrete, purely geometrical, with much in between.

The second issue, a characteristic of many if not most problems in granular matter, is that solving a problem comes with conditions, often associated with various time and length scales. When one digs deeper one finds more questions.

Granular matter offers one of the best organizing pedagogical frameworks to bring into focus a wide range of issues. A course organized around this theme could address a wide range of topics with very significant educational value.

Neal Amundson's influence in chemical engineering stems from papers, but also from teaching. His teaching—one third of his math sequence is condensed in a book[61]—equipped people with tools and a viewpoint to see the world. It is tempting to conclude—even though it is hard to transfer lessons across times—that granular materials would benefit from an Amundson-like approach. People still gravitate to elegant problems, and there is now plenty of accumulated knowledge about granular materials that—to answer the rhetorical question hinted at in the introduction—can be distilled into an elegant framework. Admittedly, the framework will be incomplete, but even an incomplete framework is preferable to no framework. An elegant framework will have the power to attract teachers and form students in what could be an integral part of the chemical engineering curriculum with valuable lessons on framing and analysis along the way.

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