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Granular matter as a window into collective systems far from equilibrium, complexity, and scientific prematurity☆

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Dedicated to Prof. John Bridgwater, for many years a guiding voice in the granular landscape

Abstract

Granular matter serves as a prototype of collective systems far from equilibrium and can be used to exemplify many concepts now associated with nonlinear dynamics and complex systems: Self-organization, invariance and symmetry breaking, various forms of pattern formation—ranging from waves to chaos to coarsening—networks, and hysteresis. The foundational concepts apply across a wide range of scales—from fine particles to ice floes—and across a wide range of technological fields. It serves also as a test-ground and illustration of the benefits and drawbacks of discrete and continuum viewpoints. However, in spite of being an integral part of the origins of chemical engineering and a topic of unquestionable practical importance, granular matter as a sub-discipline was not a central part of the basic tool-kit that launched the modern version of chemical engineering back in the 1960s. It should have been. One can only speculate how the course of ChE would have been altered if this had happened.

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1. Introduction: the two questions

John Bridgewater's name is intimately associated with granular matter and chemical engineering, and it is an honor to be able to offer a few personal remarks in this symposium. The granular matter domain is practically important and scientifically rich. The "practically important" aspect is old; the "scientifically rich" aspect is new. Granular matter is an example of a system where interactions among elementary building blocks—granules, in this case—does not give a glimpse of the behavior of the global system itself. Forced granular matter has become a paradigm of collective systems far from equilibrium and of complex systems, systems consisting of a large number of nonlinearly interacting parts (Ottino, 2003). Complex systems can be identified by what they do—display organization without a central organizing principle (emergence)—and also by how they may or may not be analyzed—decomposing the system and analyzing subparts does not necessarily give a clue as to the behavior of the whole.

Granular matter serves also to exemplify concepts now associated with nonlinear dynamics and complex systems and, in particular, *dissipative dynamical systems* (Jaeger et al., 1996a, b); it is also one of the most obvious examples for the healthy interaction between continuum and discrete viewpoints (Gollub, 2003a, b).

Granular matter offers also useful metaphors. Sandpile avalanches are the center of Self-Organized Criticality (Bak et al., 1987; Bak, 1996), a concept that, within limits, applies to a wide spectrum of systems spanning from microscopic to astrophysical scales. And excited granular matter serves as an analog for the *slow relaxation* found in glasses, spin glasses, and the like. All this is relatively recent.

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On the other hand the industrial relevance of granular matter is old, as old as engineering itself. And flow of granular materials is so pervasive in industry and natural processes that one would think that a theoretical framework would have been developed long ago. This, however, is not the case. A suitable knowledge-base would be useful across a wide range of scales—from fine particles to ice floes, and across a range of technological fields and disciplines. Consider for example mixing: The literature on granular mixing is scattered among various branches of engineering—chemical, civil, mechanical, as well as geophysics, pharmacy, materials science, powder metallurgy, and in the last ten years, physics. However, in contrast to mixing of liquids, flow and mixing of granular materials have received disproportionally little attention in chemical engineering.

Two questions come to mind: (1) What if granular materials research had been embraced by the academic chemical engineering community much earlier, say in the 1960s? And (2) Why is it that this did not happen?

2. Granular matter, difficulties

In spite of early efforts (e.g. Brown and Richards, 1970) granular matter research did not take root in chemical engineering, at least not in the US. The question is; why not? Part of the reason is that the problem is hard and, on first viewing, inelegant and messy. And that the complications are many. For example a significant difficulty in describing granular flow is the unavoidable tendency for materials to segregate or demix as a result of differences in particle properties; flow processes involving mixtures of large/small (S-systems) or dense/less-dense particles (D-systems) often lead to baffling results (Ottino and Khakhar, 2000; see Fig. 1). Then there is the issue of aspects associated with the specific nature of the materials in question, as in cohesion (Li and McCarthy, 2003).

Another differentiating aspect, as opposed to classical fluid mechanics, is the impossibility of having a fit-all approach. *Continuum* field descriptions and *Discrete* approaches—particle dynamics (PD), lattice Boltzman, Monte Carlo (MC), cellular automata calculations (CA)—all have a role. And they all have drawbacks as well (Gollub, 2003b). The main obstacle of continuum descriptions is the critical role of intermediate scales (mesoscales); manifestations occur in jamming and stress chains. On the other hand MC simulations are often too idealized to mimic specific materials, and CA computations often yield considerable insight but at the cost of sacrificing specificity. PD methods come close to the ideal of a first-principles approach. The motion of the particles is governed by Newton's laws and interactions via appropriate contact force models. But PD simulations require precise physical properties (Young's modulus, restitution coefficients, Poisson ratios, etc.) and predictive calculations for specific materials with complex multi-dispersed shapes are difficult, if not impossible.

But the most profound complication has to do with the role of thermodynamics and how it should be applied (thermodynamics, of course, does apply but nothing useful comes about by straightforward application) and there is no agreement as to what is the best route to develop a suitable theory. Consider the simplest case, when grains are large, say 100–300 μm or more, and the grain–grain interactions are purely mechanical. The kinetic energy, $(1/2)mv^2$, and potential energy, mgd, of a typical grain with mass m, diameter $d = 100\,\mu\text{m}$, and speed $v = 1\,\text{cm/s}$, are about 10^{-12} J. The necessary temperature to achieve a comparable value of $k_B T$ (k_B being the Boltzman constant) is 10^{11} K (Duran, 2000). Thermal energy, the cornerstone of the thermodynamic formalism, plays no role. No energy in, and the system freezes (jams) in a metastable state. There is still no agreement as to how formalize thermodynamics to apply to granular matter and open questions abound on many fronts (de Gennes, 1999).

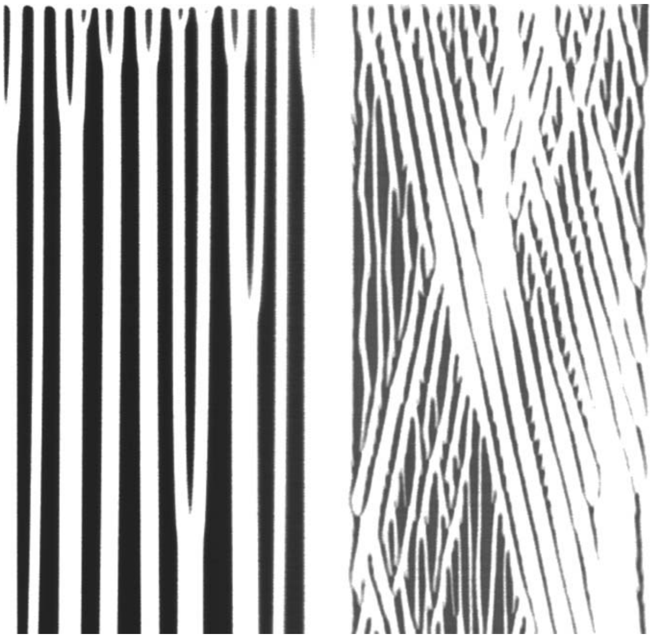


Fig. 1. Axial segregation of bidisperse S-systems in dry and slurry states. Left: Axial segregation in slurries; the rotation rate is 3 rpm; Right: Axial segregation and traveling waves in a dry system at a fast rotation rate (25 rpm) (from Fiedor and Ottino, 2003).

3. Two examples of pattern formation

Consider two examples taken from our work. For many other examples see Shinbrot and Muzzio (2000). Rotated high aspect ratio cylindrical tumblers filled with binary mixtures lead to the formation of axial alternating bands of large/small (S-systems) or dense/less- dense materials (D-systems). A few facts are well established for half full containers; upon rotation particles first separate radially in the plane perpendicular to the axis of rotation, forming a classical "radial segregation" pattern in the circular cross section. This process occurs very quickly, within a single rotation. Within the next O(10^1–10^2) rotations, the particles separate further into bands of seemingly monodisperse regions. After that a complex dynamics of bands merging may ensue (Fiedor and Ottino, 2003). Under some conditions (for example Fig. 1, left), these bands combine over many

Fig. 2. Radial segregation of bidisperse granular materials (S-systems in dry and slurry states) in periodically forced tumblers. The system is half full and has a forcing frequency of 6 cycles/revolution. Left image is the dry system, middle image is the same system in a slurry state, and the right image is the corresponding Poincaré section made using a continuum model (Fiedor and Ottino, 2005).

revolutions of the tumbler, a process known as coarsening. In other cases, (Fig. 1, right), bands may form and split and collapse and traveling waves appear. There is currently no theoretical explanation of these results.

Another example of self-organization is provided by the competition between mixing, which often creates a chaotic environment, and segregation and segregation which tries to unmix the materials (Ottino and Khakhar, 2000). The simplest case occurs in quasi-2D systems with non-circular cross sections or systems, possibly circular, rotated with a time periodic angular speed (Hill et al., 1999). Both cases lead to chaotic advection in the sense that any two particles initially close separate exponentially fast. Nevertheless, the chaos is not total: chaotic systems display regular (non-chaotic) and chaotic regions. Counteracting the tendency towards disorganization is the tendency of the material towards segregation. Experiments reveal that one class of particles moves towards the regular regions, the other towards the chaotic regions (Fig. 2). The 3D case is considered by Gilchrist and Ottino (2003).

4. The appeal of granular matter

Granular materials are now in vogue. Why now? What changed that made them fashionable? Were the difficulties mentioned earlier insurmountable in 1960 and less daunting in 1990? Probably not. As mentioned above there are still plenty of open questions. It would be too easy to attribute the change in attitude to prevalence of computers and/or improved experimentation. But this would also be wrong. One may also conjecture that appetite for metaphors and linkages—seeing granular as a window into complexity—is a modern phenomenon and that this was the key to acceptability. But this view is naïve.

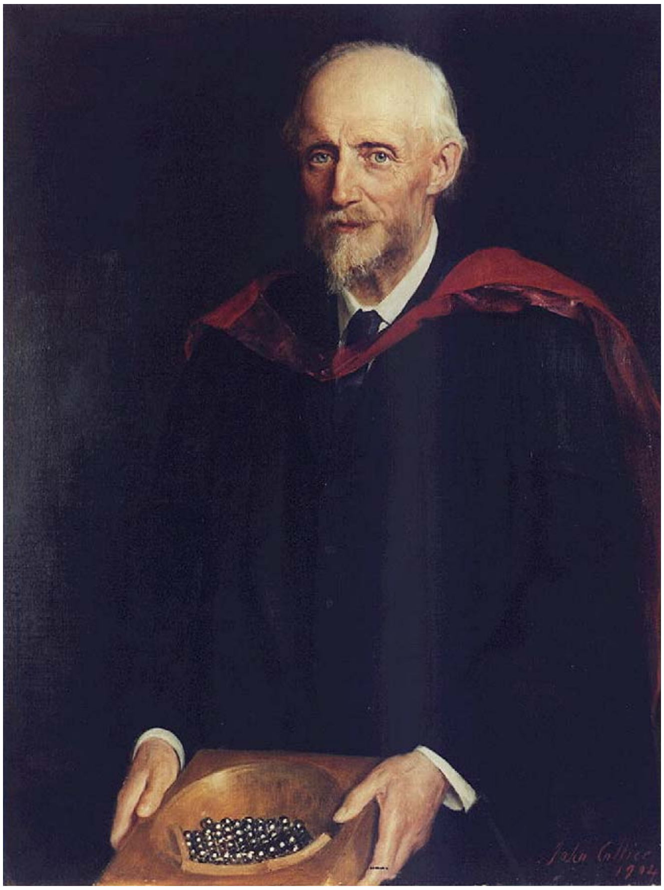


Fig. 3. Osborne Reynolds ("*I have in my hand the first experimental model universe, a soft India rubber bag filled with small shot*"). Portrait by John Collier (1904).

A century ago Osborne Reynolds wrote a short paper, one that caused quite a stir in its day. ('*On the dilatancy of media composed of rigid particles in contact. With experimental illustrations*', which was published in the *Philosophical Magazine*, in December 1885; see Reynolds, 1885, 1901). Reynolds saw in dilatancy—his discovery that granular media has to expand before it can flow—a window into the inner workings of the universe. In his 1902 Rode Lecture titled "*On an inversion of ideas as to the structure of the universe*", he makes his view clear. He said "*I have in my hand the first experimental model universe, a soft India rubber bag filled with small shot*" (Kargon, 1975; see Fig. 3). Reynolds elaborated this issue in detail in a long paper—actually, at 251 pages, more of a monograph (Reynolds, 1903). The opening sentence says it all: "*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.*" [Parenthetically, an excellent resource on Osborne Reynolds is the website of Prof. J.D. Jackson from the Manchester School of Engineering, http://www.eng.man.ac.uk/historic/reynolds/oreyna.htm).]

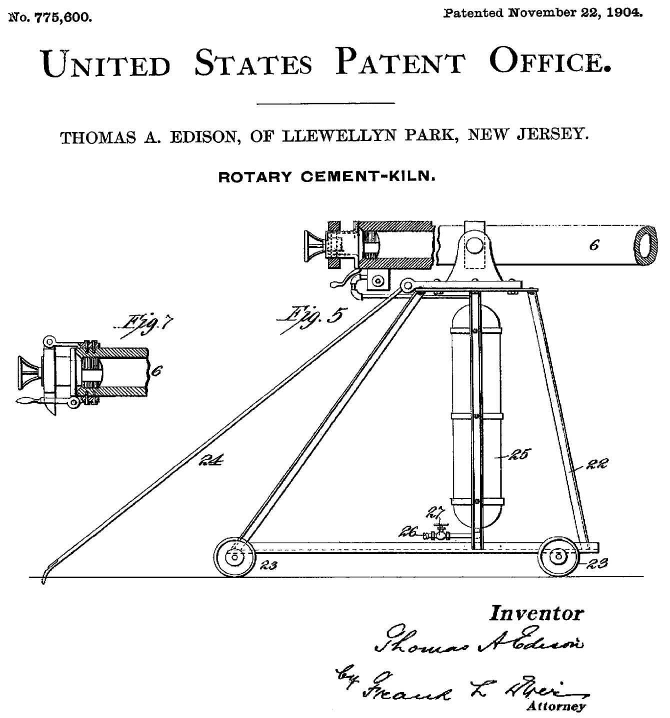
Reynolds believed that the universe was filled with rigid grains and went so far as to calculate the size of these particles: 5.534×10^{-18} cm, with a mean free path of 8.612×10^{-28} cm. This may be the smallest scale to emanate from a continuum picture. As reference consider the Planck length, the 'quantum of length', the scale at which classical ideas about gravity and space-time cease to be valid, and quantum effects dominate, the smallest measurement of length with any meaning; this is 1.6×10^{-37} cm or about 10^{-20} times the size of a proton. All this may sound far fetched now, but Reynolds was a teacher of J.J. Thomson (who received a Nobel Prize for his discovery of the electron), so one may argue that he inspired the right ideas. So much for the modernity of far-reaching analogies... .

The possibility of seeing analogies in granular matter is clearly part of the appeal. But there are other reasons for the popularity. I speculated earlier—in a book review for the *Journal of Fluid Mechanics* (Ottino, 2000) and in a perspective piece for *Powder Technology* (Ottino and Khakhar, 2001)—that the reason for the popularity may be traced to several interconnected factors. The first is that there is new physics and that open theoretical questions abound. The second is that experimentation, even though it often requires far more sophistication than may appear at first glance, is still accessible and creativity still plays an important role. The third element is that intuition—often built on fluids—often does not work. And the final reason for the appeal—one that undoubtedly should resonate with the readers of *Chemical Engineering Science*—is the clear interplay between science (understanding and explaining) and technology (making and building).

5. Sleeping beauties and prematurity

In spite of all the above, through the 1850–1970s, fluid mechanics developed while granular matter languished. And in the mid 1990s the granular field took off. Could some of the recent work have been done earlier? The answer to this question is unquestionably yes. There is no reason that the work on vibrated layers (Umbanhowar et al., 1996), stratification (Makse et al., 1997), and our own work on avalanche mixing (Metcalfe et al., 1995) could have been done even decades earlier. The issue, it appears, is not one of available techniques and machinery but more of framing and perspective. Once one sees patterns, for example, one looks for them. The growth in these areas and even sub-areas has been explosive. Consider for example the review of size segregation in vibrated granular materials written by Kudrolli (2004).

An interesting and not all too distant focal point of interest, one that I believe could have signaled the beginning of granular matter as an analog for other phenomena, is a paper by a Japanese researcher, Yositisi Oyama. In 1939 Oyama wrote a paper (Oyama, 1938) which clearly identified axial segregation. [Oyama's work is usually listed as I.P.C.R. 18, 600 (1939), in Japanese. Attribution to this work as "in Japanese" in virtually all papers is baffling as the paper is translated, though far from flawlessly, into English.] (as usual one can find earlier precedents. The formation of rings was a well-know problem in the cement industry. None other than Edison (1904) invented a device to destroy molten clinker rings that form in rotary cement kilns; see Fig. 4).


No. 775,600. Patented November 22, 1904.

UNITED STATES PATENT OFFICE.

THOMAS A. EDISON, OF LLEWELLYN PARK, NEW JERSEY.

ROTARY CEMENT-KILN.

Inventor
Thomas A Edison
by Frank L Dyer
Attorney

Fig. 4. Thomas Edison's incursion in axial segregation. US patent 775,600, 1904: Device to destroy molten clinker rings found in rotary cement kilns.

A citation search in ISI for Oyama, 1939 lists 49 hits, 42 of them from 1991 or later. The first of the "recent" citing papers is Weidenbaum (1958), then working at Corning Glass Works after a PhD in 1953 at Columbia. Widenbaum is very precise and he even fills in missing words in the translated title of Oyama's most famous paper (he refers to several other papers, those in Japanese). *But not much is made out of banding* by either Weidenbaum or by Oyama himself who in fact saw degree of packing and how it is related to composition as at least an equally important part of his work (the pictures of axial segregation show views along the axis of cylinder).

Is Oyama's (1939) a "Sleeping Beauty Paper"? In the words of van Raan (2004) a "sleeping beauty" in science is a publication that goes unnoticed ('sleeps'), gathering less than one citation a year for many years, and then, almost suddenly, attracts a lot of attention (the paper is awakened by a "prince"). Tracking of citations in ISI starts in 1955, but it is likely that the paper was not widely cited before then. It had only 7 citations in nearly 50 years (period 1955–1991).

Bridgwater (1976) cites the paper and attributes the reference to Weidenbaum; the next influential reference is Dasgupta et al. (1991), and after that the paper awoke (all Oyama's papers are listed in a literature survey by Bridgwater and collaborators, Cooke et al., 1976).

Indebtedness is not always acknowledged; in fact no one may know that a paper exists. An example I am familiar with is another paper by Osborne Reynolds, "*The Method of Coloured*

Bands" (Reynolds, 1893, 1894). [Parenthetically a second appearance by Reynolds it is suitable in the context of this symposium since, as his father before him, Osborne Reynolds passed through Cambridge. As an undergraduate Reynolds attended some of the same classes as Rayleigh, who was one year ahead of him.] "*The Method of Coloured Bands*" paper by Reynolds enters in the category of overlooked papers (it has 16 citations in the period 1955–2004). This work, actually the transcript of a lecture/demonstration given at the Royal Institution in London, puts forward the idea that the essence of mixing is stretching and folding. These ideas were developed, by me and others, in complete ignorance of this paper. I started with a lamellar model with stretching being the central idea (Ottino et al., 1979), but folding was missing. I made the connection with folding when I learned of Smale horseshoes and the surrounding math. I became aware of Reynolds's paper sometime in the 1990s and found a way to weave in Osborne Reynolds contributions in a symposium in Asilomar honoring the late Bill Reynolds from Stanford (Ottino et al., 1994). Had I known about Osborne Reynolds's paper in 1979 I could have saved at least 10 years. This is a different kind of sleeping beauty. The idea is recognized retrospectively, long after a new strand has been created.

The "*The Method of Coloured Bands*" paper by Reynolds is closer to what has been called *prematurity in science*, an idea developed by Stent (1972). Stent used Oswald Avery's discovery that DNA is genetic material and Watson and Crick's discovery of the DNA double helix structure to investigate the question: What does it mean to say that a discovery is premature? He points out that Avery's identification of DNA as genetic material in 1944 had virtually no effect on the field of genetics—the discovery was premature. And premature, Stent argued, is when: "[the] *implications cannot be connected by a series of simple logical steps to canonical or generally accepted knowledge*." Oyama's paper, I would argue, falls in this category.

6. The role of the innovator

The innovator has to prepare the ground. This was said best by Wordsworth in the context of literature: "*Never forget what I believe was observed to you by Coleridge*, *that every great and original writer*, *in proportion as he is great and original*, *must himself create the taste by which he is relished*" (William Wordsworth (English poet, 1770–1850) in Letter to Lady Beaumont, 21 May 1807; in E. de Selincourt (ed.) *Letters of William and Dorothy Wordsworth* vol. 2; revised by M. Moorman, 1969).

The ground was not ready for Oyama's paper. But the ground in ChE was ready for fluids, transport and reaction engineering. The prevalent problems reinforced the prevalent tools, and *vice versa*: fluid mechanics, transport phenomena and reaction engineering were firmly grounded in math, and math contributed to a deep understanding of these subjects (Ramkrishna and Amundson, 2004). Foundations were clear and elegant. Not much need for resorting to approximate scaling concepts and no need to question deep assumptions. Thus the work by Haff (1983), for example, may have been perceived as too heuristic by the chemical engineering fluid mechanics community.

One could argue that many of John Bridgewater's papers were also a bit premature. Consider some titles in the period 1969–1979: "Particle mixing by segregation" (Bridgwater et al., 1969); "Rate of spontaneous inter-particle percolation" (Bridgwater and Ingram, 1971); "Mixing of dry solids by percolation" (Campbell and Bridgwater, 1973); "Interparticle percolation—Fundamental solids mixing mechanism" (Scott and Bridgwater, 1975); "Self-diffusion of spherical particles in a simple shear apparatus" (Scott and Bridgwater, 1976); 'Interparticle percolation—statistical mechanical interpretation" (Cooke and Bridgwater, 1979).

All these could be perfectly respectable titles for papers written today.

7. Bifurcations, roads not taken

What if granular materials and Oyama's paper had been in the first issue of *Chemical Engineering Science*? What metaphors could have been part of the picture? How could the ChE curriculum have developed? What metaphors could have been developed in the context of ChE? What tools could have been part of the ChE culture? One cannot fail to think that Oyama's paper, or a version of it, could have been a bifurcation point. And that what could have emerged from it could have been be part of the modern version of ChE emerged that in the 1960s. Pattern formation as observed in vibrated layers of particles and many kinds of unmixing processes, ranging from waves to coarsening, could have been even more prevalent than it is now, where most examples occur in the context of diffusion-reaction problems (Kiss and Hudson, 2003). There could also have been more exposure to various discrete models such as Cellular Automata models (Makse et al., 1997); rheology could have evolved smoothly to include pastes (another of the areas pioneered by John Bridgwater), and polymer physics and granular matter could have found common ground under jamming. And quite possibly ChE would have had more exposure to solid mechanics.

Why did granular materials research not catch on from the 1960s to the 1980s? Clearly there were serious efforts but they were scattered and never percolated. One strand of the work in the 1960s–1980s was being done by engineers who were trying to solve practical problems in what was essentially a case-by-case approach. Some work that went systematically into phenomena such as axial segregation was done by Donald and Roseman (1962); Roseman and Donald (1962), Rogers and Clements (1971) and others. Other efforts connected mostly with solid mechanics (e.g. Nedderman, 1992). Fluidization was more glamorous and this sub-field was much more developed than granular materials as a whole. Most of the seminal work done in the 1960s (single bubble hydrodynamics, correlations for heat and mass transfer, etc.) could have been used/extended for granular materials as a whole. Even more recently there was notable work on fundamentals of granular materials. Here we could cite Jackson, Savage, Jenkins, and Campbell, but their work remained a sort of niche/specialized effort. In fact, until recently, most of this body work was largely ignored or undiscovered by physicists and hence few people flocked into the subject and the foundations never developed in a coherent way. In the late 1980s and early 1990s physicists acknowledged that granular material physics was largely unexplored (Jaeger, 1996a, b) and engineers (Ennis et al., 1994) argued emphatically that better understanding would have a tremendous benefit to industry. These two camps were completely independent of each other. The field got respectability with many high profile researchers jumping in (Pierre de Gennes in France, Sam Edwards in England) and it very quickly became a hot area.

Research is disorganized; there is always overshooting; it may well be that there is too much curiosity-driven research on the physics side of granular matter and that work is needed to bring to the center the nature of the materials themselves (chemistry and physical chemistry) into the picture. But this is precisely what chemical engineers can do (Li and McCarthy, 2003). And on the technological front I would argue that many of the necessary building blocks for understanding of industrial systems are already developed (Ottino and Khakhar, 2002). The bridge between academic research and industrial practice is far from insurmountable. Scouting can yield significant results for those who are persistent enough and have the right training (or work at acquiring it).

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