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## **Report Title**

### **Final Report: Jamming / Unjamming Dynamics in Granular Materials**

#### **ABSTRACT**

Granular materials can exhibit both solid-like jammed and liquid-like unjammed behavior. The project investigates the role of local, particle-scale properties in affecting the overall response to applied stresses as the jamming transition is crossed. It extends the current understanding of the jamming transition by focusing on aspects that allow for control of the dynamic interactions among particles, in particular particle shape and interstitial fluids. This is of fundamental interest for developing accurate models for the onset or demise of mechanical rigidity, and it also provides a path toward engineering new classes of particulate materials with properties by design. Scientific accomplishments include (i) the investigation of dynamic jamming fronts that drive the impact-induced jamming of dense suspensions, (ii) the development of a new computer-aided method for identifying optimized particle shapes via evolutionary algorithms, and (iii) the development of a high-speed ultrasound imaging system for non-invasive three-dimensional imaging of dense suspensions.

**Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:**

**(a) Papers published in peer-reviewed journals (N/A for none)**

| <u>Received</u> | <u>Paper</u>                                                                                                                                                                                                                                                                                 |
|-----------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 07/31/2015      | 15 Heinrich Jaeger. Toward Jamming by Design, Soft Matter, (01 2015): 12. doi:                                                                                                                                                                                                               |
| 07/31/2015      | 16 Sayantan Majumdar, Qin Xu, Eric Brown, Heinrich M. Jaeger. Shear thickening in highly viscous granular suspensions, EPL (Europhysics Letters), (09 2014): 68004. doi: 10.1209/0295-5075/107/68004                                                                                         |
| 08/26/2013      | 1 Marc Z. Miskin, Heinrich M. Jaeger. Adapting granular materials through artificial evolution, Nature Materials, (01 2013): 326. doi: 10.1038/nmat3543                                                                                                                                      |
| 08/26/2013      | 5 H. M. Jaeger, S. R. Waitukaitis. In situ granular charge measurement by free-fall videography, Review of Scientific Instruments, (02 2013): 25104. doi: 10.1063/1.4789496                                                                                                                  |
| 08/26/2013      | 4 Ivo R. Peters, Qin Xu, Heinrich M. Jaeger. Splashing Onset in Dense Suspension Droplets, Physical Review Letters, (07 2013): 28301. doi: 10.1103/PhysRevLett.111.028301                                                                                                                    |
| 08/26/2013      | 2 Heinrich M. Jaeger, Scott R. Waitukaitis. Impact-activated solidification of dense suspensions via dynamic jamming fronts, Nature, (07 2012): 205. doi: 10.1038/nature11187                                                                                                                |
| 08/30/2014      | 8 Ivo R. Peters, Heinrich M. Jaeger. Quasi-2D dynamic jamming in cornstarch suspensions: visualization and force measurements, Soft Matter, (07 2014): 0. doi: 10.1039/C4SM00864B                                                                                                            |
| 08/30/2014      | 9 Marc Z. Miskin, Heinrich M. Jaeger. Evolving design rules for the inverse granular packing problem, Soft Matter, ( 2014): 0. doi: 10.1039/c4sm00539b                                                                                                                                       |
| 08/30/2014      | 10 Athanasios G. Athanassiadis, Marc Z. Miskin, Paul Kaplan, Nicholas Rodenberg, Seung Hwan Lee, Jason Merritt, Eric Brown, John Amend, Hod Lipson, Heinrich M. Jaeger. Particle shape effects on the stress response of granular packings, Soft Matter, ( 2014): 0. doi: 10.1039/c3sm52047a |
| 08/30/2014      | 11 Eric Brown, Heinrich M Jaeger. Shear thickening in concentrated suspensions: phenomenology, mechanisms and relations to jamming, Reports on Progress in Physics, (04 2014): 0. doi: 10.1088/0034-4885/77/4/046602                                                                         |
| 08/30/2014      | 12 Athanasios G. Athanassiadis, Patrick J. La Rivière, Emil Sidky, Charles Pelizzari, Xiaochuan Pan, Heinrich M. Jaeger. X-ray tomography system to investigate granular materials during mechanical loading, Review of Scientific Instruments, (08 2014): 0. doi: 10.1063/1.4893555         |
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Number of Papers published in peer-reviewed journals:

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(b) Papers published in non-peer-reviewed journals (N/A for none)

Received              Paper

**TOTAL:              2**

Number of Papers published in non peer-reviewed journals:

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(c) Presentations

- 10/30/2014 Soft|MetaMatter Meeting, University of Chicago
- 01/19/2015 Master Class on Granular Matter, Annual Mtg Dutch Physical Society, Veldhoven
- 01/20/2015 Granular Matter by Design, invited talk, Annual Mtg Dutch Physical Society, Veldhoven
- 02/20/2015 Granular Streams, Northwestern University Colloquium
- 04/25/2015 Jammable Granular Matter, MIT Active Matter Summit
- 05/08/2015 Dynamic Jamming in Dense Suspensions, UIUC seminar

**Number of Presentations: 6.00**

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Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received              Paper

**TOTAL:**

**Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):**

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**Peer-Reviewed Conference Proceeding publications (other than abstracts):**

Received

Paper

08/26/2013 6.00 Heinrich M. Jaeger, Marc Z. Miskin, Scott R. Waitukaitis. From nanoscale cohesion to macroscale entanglement: Opportunities for designing granular aggregate behavior by tailoring grain shape and interactions, POWDERS AND GRAINS 2013: Proceedings of the 7th International Conference on Micromechanics of Granular Media. 08-AUG-13, Sydney, Australia. : ,

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**(d) Manuscripts**

Received

Paper

**TOTAL:**

**Number of Manuscripts:**

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**Books**

Received

Book

08/30/2014 13.00 Scott Waitukaitis. Impact-Activated Solidification of Cornstarch and Water Suspensions, Switzerland: Springer International, (01 2015)

**TOTAL: 1**

Received

Book Chapter

**TOTAL:**

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**Patents Submitted**

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**Patents Awarded**

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**Awards**

n/a

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**Graduate Students**

| <u>NAME</u>            | <u>PERCENT SUPPORTED</u> | Discipline |
|------------------------|--------------------------|------------|
| Kieran Murphy          | 0.16                     |            |
| Endao Han              | 1.00                     |            |
| Qin Xu                 | 0.08                     |            |
| <b>FTE Equivalent:</b> | <b>1.24</b>              |            |
| <b>Total Number:</b>   | <b>3</b>                 |            |

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**Names of Post Doctorates**

| <u>NAME</u>            | <u>PERCENT SUPPORTED</u> |
|------------------------|--------------------------|
| Ivo Peters             | 0.75                     |
| Sean McBride           | 0.08                     |
| <b>FTE Equivalent:</b> | <b>0.83</b>              |
| <b>Total Number:</b>   | <b>2</b>                 |

---

**Names of Faculty Supported**

| <u>NAME</u>            | <u>PERCENT SUPPORTED</u> | National Academy Member |
|------------------------|--------------------------|-------------------------|
| Heinrich Jaeger        | 0.08                     |                         |
| <b>FTE Equivalent:</b> | <b>0.08</b>              |                         |
| <b>Total Number:</b>   | <b>1</b>                 |                         |

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### Names of Under Graduate students supported

| <u>NAME</u>            | <u>PERCENT SUPPORTED</u> | Discipline |
|------------------------|--------------------------|------------|
| Clare Singer           | 0.08                     | physics    |
| Nikolaj Reiser         | 0.08                     | physics    |
| Darius Choksy          | 0.16                     | physics    |
| <b>FTE Equivalent:</b> | <b>0.32</b>              |            |
| <b>Total Number:</b>   | <b>3</b>                 |            |

### Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: ..... 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 3.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 3.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense ..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:..... 0.00

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### Names of Personnel receiving masters degrees

|                      |
|----------------------|
| <u>NAME</u>          |
| <b>Total Number:</b> |

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### Names of personnel receiving PHDs

|                      |
|----------------------|
| <u>NAME</u>          |
| Qin Xu               |
| <b>Total Number:</b> |

---

### Names of other research staff

|                        |                          |
|------------------------|--------------------------|
| <u>NAME</u>            | <u>PERCENT SUPPORTED</u> |
| <b>FTE Equivalent:</b> |                          |
| <b>Total Number:</b>   |                          |

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### Inventions (DD882)

### Scientific Progress

See Attachment

## Technology Transfer

none to report

**Proposal Number: 61714-MS, Agreement Number: W911NF-12-1-0182**

## *Jamming/Unjamming Dynamics in Granular Materials*

PI: Heinrich Jaeger, University of Chicago

### **Final Report**

May 01, 2012, to April 30, 2015

## **1 Summary**

Granular materials are random aggregates of a large number of individually solid particles that interact primarily via direct contacts among neighboring grains. They encompass a wide range of particulate media from sand, gravel, soil or mud to pharmaceutical powders to much of the non-liquid raw ingredients used for processing in industry. Despite their seeming simplicity, these materials exhibit astoundingly complex dynamics in which the aggregate can transition between solid-like “jammed” and liquid-like “unjammed” behavior. The detailed understanding of the dynamics of granular materials affects the plastic deformation of the particle arrangement in response to local stresses during any type of impact, where the granular aggregate deforms and rearranges before again turning rigid and jammed in a new packing configuration. From a basic scientific perspective granular matter, and jamming in particular, provides a paradigm for emergent cooperative behavior in a nonlinear and dissipative system far from equilibrium.

The overarching goal for the project has been to extend the current knowledge about the jamming/unjamming transition by exploring the fundamental mechanisms that drive unjamming under stress loading, and to map out the nonlinear dynamic system responses in the unjammed region. This was achieved through an approach that combined unique experimental capabilities with computer simulations on model systems. The project focused on fully three-dimensional systems and concentrated on two aspects, particle shape and interstitial fluids, since they directly affect the interactions among particles and therefore are critical for the qualitative as well as quantitative delineation of different dynamic regimes. This enabled the development of a new level of predictive capability that encompasses the full range from micro- to macro-scale phenomena while crossing the jamming phase boundary under applied stress.

Key achievements include the systematic investigation of the role of particle shape on the aggregate behavior of granular materials under stress loading, using 14 convex and non-convex shapes; the development of a new approach, based on artificial evolution, for creating granular materials with aggregate properties by design; the discovery of dynamic jamming fronts in dense suspensions during impact; and the development of high-speed ultrasound imaging techniques to track these fronts in three-dimensional systems.

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### 3 Statement of Problem Studied

Granular materials can exhibit both solid-like jammed and liquid-like unjammed behavior. The project investigated the role of local, particle-scale properties in affecting the overall response to applied stresses as the jamming transition is crossed. It extended the current understanding of the jamming transition by focusing on aspects that allow for control of the dynamic interactions among particles, in particular particle shape and interstitial fluids. This is of fundamental interest for developing accurate models for the onset or demise of mechanical rigidity, and it also provides a path toward engineering new classes of particulate materials with properties by design.

The project was divided into two main lines of investigation that complement each other, exploring the jammed and unjammed regions on either side of the transition. For the purpose of this project we define a particulate aggregate to be jammed until strains become large enough to induce global, catastrophic yielding. The aggregate is unjammed beyond this point. In macroscopic particulate systems or suspensions of sufficiently large particles, where thermal fluctuations play no role in driving relaxation and creep is minimized, this operational definition can serve as a practically useful demarcation between the (quasi-) static jammed and flowing unjammed regimes.

**Jammed Regime.** This effort investigated the response of granular packings to applied stresses up to the point of catastrophic failure. The *specific objectives* were to establish a) how the shape of the jamming phase boundary depends on particle-scale properties and b) how the feedback between local packing structure and applied stress can be exploited to control the stress-strain characteristics. This allowed us to address questions such as how particle shape affects dilation or compaction and thereby determines the degree of plastic deformation at the point of failure.

**Unjammed Regime.** This effort focused on the regime where the material has yielded and is flowing. Both steady-state and transient responses to the applied loading were investigated. The *specific objective* was to establish how different types of dynamic response in the unjammed regime depend on local packing geometry (controlled by particle shape) and interparticle contacts (in addition controlled by interstitial fluids). Of particular interest was the question to what extent an interstitial liquid, and the dynamic response associated with it, can be used to tune the role played by particle shape. Thus, a main focus of this effort was directed at investigating the dynamic response of dense suspensions, i.e., concentrated dispersions of solid particles in a liquid.

### 4 Scientific Accomplishments

#### 4.1 Overview

Below we describe key results from efforts investigating both the jammed and unjammed regimes. Our approach used a combination of systematic experiments and numerical simulations.

On the experimental side, our lab made extensive use of a unique suite of capabilities for non-invasive imaging during stress loading. Combining an Instron materials tester and a surgical x-ray system, we developed a triaxial test system for automated, fully 3D tomographic reconstruction of particle configurations under varying loading conditions. A new capability was developed for fast ultrasound imaging at frame rates up to 10,000 frames per second, which allowed us to extract flow fields non-invasively from the interior of dense suspensions near the jamming transition. We also set up 3D printing capabilities for particle fabrication with spatial resolution better than 40 microns and the ability to digitally mix different print materials in order to vary the elastic modulus of the printed object. In addition, a new experimental method to determine charges on individual grains of granular powders was developed.

On the numerical side, we developed a new approach for optimizing the structural and/or dynamic behavior of granular materials. This approach is based on the combination of DEM simulations with evolutionary algorithms. A dedicated mini-cluster of computers was set up to run these algorithms and to model the behavior of jammed, granular aggregates.

*Overall, the research resulted in 16 papers. One paper each was published in Nature [1], Nature Materials [2], Physical Review Letters [3], Europhysics Letters [4], and Reviews of Progress in Physics [5]. Two papers were published in Review of Scientific Instruments [6, 7], and four papers in Soft Matter [8-11]. One further paper was published as an AIP conference proceedings [12]. Four additional papers based on this research have just been submitted to peer-reviewed journals. Three Ph.D.s were awarded, and one Ph.D. thesis was published as book by Springer Int'l [13].*

Highlights from the research are reported in the following Sections.

## 4.2 Jammed Regime

In this project component we focused on two aspects: non-invasive x-ray imaging of three-dimensional granular aggregates during stress loading, and evolutionary strategies to predict optimal particle shapes for a given performance goal of the jammed aggregate.

In order to explore the role of particle shape, we implemented a two-pronged approach: (i) We fabricated by 3D-printing a set of shapes that have been proposed in the literature as particularly suitable for producing strongly jammed configurations. (ii) We demonstrated successfully an evolutionary strategy to predict optimal particle shapes for a given performance goal of the aggregate material. Specifics and key outcomes are as follows.

### 4.2.1. Role of Shape in Setting the Stress Response

We performed measurements of the stress response of random packings formed from a wide range of particle shapes (14 shapes in total). Besides spheres these include convex shapes such as the Platonic solids, truncated tetrahedra, and triangular bipyramids, as well as more complex, non-convex geometries such as hexapods with various arm lengths, dolos, and tetrahedral frames. All particles were 3D-printed. Well-defined initial packing states were established through pre-conditioning by cyclic loading under given confinement pressure. Starting from such initial states, stress-strain relationships for axial compression were obtained at four different confining pressures for each particle type. While confining pressure has the largest overall effect on the mechanical response, we found that particle shape controls the details of the stress-strain curves and can be used to tune packing stiffness and yielding. By correlating the experimentally measured values for the effective Young's modulus under compression, yield stress and energy loss during cyclic loading, we identified trends among the various shapes that allow for designing a packing's aggregate behavior.

This part of the project completes an effort that was originally started under grant W911NF-08-1-0140 (Darpa, *Programmable Matter*) in collaboration with Hod Lipson's group at Cornell. That initial part of the effort produced some of the Platonic solids and first, exploratory measurements of the stress-strain

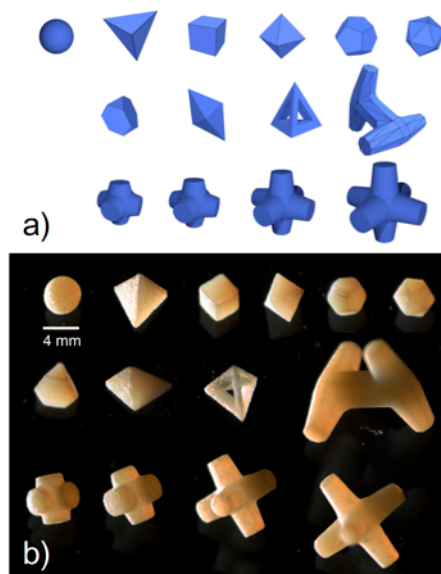


**Fig. 1.** Particle types used in our study. a) Rendered models. b) 3d printed.

curves. Under the current grant additional shapes were added, shapes were re-printed, and more detailed measurements were performed.

By measuring the stress-strain relationship of the particle aggregate under quasistatic compression, we focused on the overall, macroscopic response. For each particle type, we performed three to five triaxial tests at four different confining pressures for a total of 190 independent experiments. From our data we extracted parameters characterizing the aggregate performance for each shape, such as an effective Young's modulus, a yield stress, and the amount of energy loss during cyclic compression.

The data from these experiments provide *a first extensive overview of the trends that emerge when shape is varied*. Further, we are able to identify correlations among the aggregate parameters and how they vary with geometric characteristics of the particle shape. These experimental data provide both a benchmark for comparison with simulations and a reference guide for picking appropriate shapes for applications.

**Fig. 1a** shows renderings of the 14 geometric models used to 3D-print the particles. We designed all eight convex shapes to have equal volume  $V = 22.5 \text{ mm}^3$ , corresponding to a side length of 2.8 mm for the cubes. The tetrahedral frames have the same outer dimensions as the solid tetrahedron, with beams along the edges thick enough to withstand the stresses in the experiments without breaking (1.4 mm). The four jacks are formed by a central sphere with six truncated, conical arms at right angles; the only parameter we varied was the arm length (0.92 mm, 1.3 mm, 2.6 mm, 3.6 mm). The dolos (twisted 'H') were printed at a larger size (10.7 mm arm length). We printed the particles (**Fig. 1b**) in sets of 5500 on our Objet Connex 350 3D-printer (**Fig. 5**), using 50 $\mu\text{m}$  print resolution and a UV-cured resin ("Vero White Plus", Objet Geometries Inc.). To characterize the resin material itself, we compressed individual cubes to determine a compressive modulus  $E_{mat} = 1260 \pm 120 \text{ MPa}$ .

For the mechanical tests, we measured the stress response of the granular packings in a high-precision triaxial test. We prepared random packings by slowly pouring particles through a funnel into a cylindrical latex membrane (Durham Geo-Enterprises, 0.30 mm thickness) of inner diameter  $d=50.8 \text{ mm}$ , filling it to a height  $h=102 \text{ mm}$ . In this configuration, each sample contained 5000-5500 particles and measured 15-20 particles across the membrane diameter (except dolos and large jacks, which had closer to 10 across). We maintained this  $d:h = 1:2$  initial sample aspect ratio for all triaxial tests, as is standard in soil mechanics. Once the sample was loaded, we capped the open top of the membrane with an aluminum disc and rigidly connected the sample to the testing apparatus, an Instron 5869 materials tester. The bottom end cap of the packing was covered by a porous sintered disc and was connected to a vacuum pump. This pump allowed us to apply confining pressures between 0.001 MPa and 0.080 MPa to the packing during the triaxial test.

To guarantee reproducibility in triaxial compression tests with frictional particles, care must be taken ensure a uniform, isotropic confining stress prior to any additional axial compression. We achieved this with the following protocol. We first applied the desired radial confining stress  $\sigma_{con}$  to the sample

| Shape                                                                                             | $\phi_0$        | $\phi_{cyc}$    |
|---------------------------------------------------------------------------------------------------|-----------------|-----------------|
|  Cubes         | $0.59 \pm 0.04$ | $0.58 \pm 0.04$ |
|  Tetrahedra    | $0.57 \pm 0.04$ | $0.57 \pm 0.04$ |
|  Octahedra     | $0.57 \pm 0.04$ | —               |
|  Spheres       | $0.56 \pm 0.04$ | $0.55 \pm 0.04$ |
|  Dodecahedra   | $0.56 \pm 0.04$ | —               |
|  Trunc. Tetra. | $0.56 \pm 0.04$ | —               |
|  Icosahedra    | $0.55 \pm 0.04$ | —               |
|  0.9mm Jacks   | $0.54 \pm 0.04$ | —               |
|  1.3mm Jacks   | $0.52 \pm 0.03$ | —               |
|  Tri. Bipyr.   | $0.48 \pm 0.03$ | —               |
|  Dolos         | $0.46 \pm 0.03$ | —               |
|  2.6mm Jacks   | $0.46 \pm 0.03$ | —               |
|  3.6mm Jacks   | $0.39 \pm 0.03$ | —               |
|  Tet. Frames   | $0.25 \pm 0.02$ | $0.25 \pm 0.02$ |

**Table 1.** Packing fractions of the amorphous configurations of various particle types as poured ( $\phi_0$ ) and, for a subset of the shapes, measured after pre-conditioning of the packing by cyclic loading ( $\phi_{cyc}$ ).

using vacuum, while the top cap was rigidly held in place by the material tester. Monitoring the axial stress  $\sigma_a$  with the Instron's load cell, we then lowered the top cap until it matched the radial confining stress. This stress-balanced state defined our initial state with deviatoric stress  $q = \sigma_a - \sigma_{con} = 0$ . Next, to reduce effects from run-to-run variations associated with sample preparation, we axially compressed each sample by 3% (vertical displacement of 3mm for sample height 102 mm) and then returned to  $q=0$ . In our experiments, we found that conditioning the packing with  $N = 10$  cycles resulted in a state that was largely independent of the initial pouring and yielded stress-strain curves that were highly reproducible from run to run. We therefore used the state with  $q = 0$  after  $N = 10$  cycles as our reference state. To map out the mechanical response, packings were further compressed up to a strain  $\varepsilon = 0.02$ . **Table 1** lists the packing fractions of the samples as poured and after pre-conditioning by cyclic loading.

**Figure 2a** shows the ensemble averaged traces for each of the five Platonic solids and spheres at a confining pressure of 0.080 MPa. The equivalent plots for the other shapes tested are shown in **Fig. 2b,c**. Inspection of these plots reveals a number of trends. Compared to spheres (bottom trace in **Fig. 2a**) the introduction of facets in the Platonic solids increases the initial slope, i.e., the packing's stiffness, as well as the stress level at which large-scale plastic deformation sets in. Relatively compact, sphere-like shapes (including highly faceted particles as well as jacks with very short arms) exhibit nearly perfectly plastic failure beyond yielding, i.e., a nearly horizontal  $q(\varepsilon)$ . Shapes with

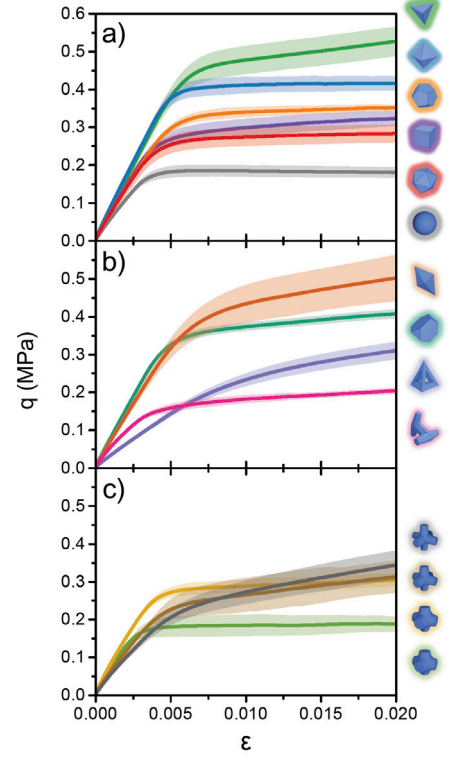


**Fig. 2.** Ensemble-averaged stress-strain curves.

| Shape         | $\theta_m$ (deg) | $\sigma_{con} = 0.001$ MPa |                  |                                  | $\sigma_{con} = 0.080$ MPa |                  |                                  |
|---------------|------------------|----------------------------|------------------|----------------------------------|----------------------------|------------------|----------------------------------|
|               |                  | $E$ (MPa)                  | $\sigma_y$ (kPa) | $\varepsilon_y (\times 10^{-3})$ | $E$ (MPa)                  | $\sigma_y$ (kPa) | $\varepsilon_y (\times 10^{-3})$ |
| 3.6mm Jacks   | 57±3             | 6.63±0.15                  | 9.51±0.11        | 1.43±0.04                        | 59.5±0.2                   | 167±1            | 3.86±0.02                        |
| Dolos         | 41±3             | 9.67±0.33                  | 5.25±0.08        | 0.93±0.03                        | 63.2±0.3                   | 109±1            | 2.54±0.01                        |
| 2.6mm Jacks   | 39±3             | 7.62±0.25                  | 5.51±0.13        | 0.97±0.04                        | 68.6±0.3                   | 162±1            | 3.15±0.01                        |
| Tri. Bipy.    | 39±3             | 4.14±0.20                  | 12.65±0.03       | 3.41±0.18                        | 71.2±0.3                   | 324±1            | 5.53±0.03                        |
| Trunc. Tetra. | 39±3             | 3.15±0.10                  | 8.19±0.07        | 2.59±0.09                        | 77.9±0.3                   | 292±1            | 4.41±0.02                        |
| Tetrahedra    | 37±3             | 5.60±0.16                  | 7.83±0.16        | 1.72±0.05                        | 82.0±0.3                   | 380±1            | 5.38±0.02                        |
| Octahedra     | 37±3             | 3.16±0.29                  | 8.84±0.27        | 3.15±0.30                        | 90.0±0.3                   | 340±1            | 4.47±0.02                        |
| Tet. Frames   | 37±3             | 2.47±0.14                  | 8.75±0.04        | 3.68±0.24                        | 33.3±0.2                   | 171±1            | 6.45±0.05                        |
| 1.3mm Jacks   | 35±3             | 6.05±0.17                  | 4.96±0.06        | 1.11±0.03                        | 91.3±0.3                   | 203±1            | 2.99±0.01                        |
| Cubes         | 35±3             | 3.04±0.10                  | 5.58±0.14        | 2.46±0.08                        | 77.4±0.3                   | 221±1            | 3.54±0.02                        |
| 0.9mm Jacks   | 34±3             | 4.27±0.16                  | 3.45±0.17        | 1.22±0.05                        | 90.2±0.3                   | 119±1            | 1.91±0.01                        |
| Icosahedra    | 34±3             | 2.80±0.14                  | 4.49±0.09        | 2.45±0.13                        | 74.7±0.4                   | 210±1            | 3.45±0.02                        |
| Dodecahedra   | 34±3             | 2.74±0.10                  | 8.30±0.06        | 3.61±0.14                        | 73.5±0.3                   | 268±1            | 4.43±0.02                        |
| Spheres       | 26±3             | 3.24±0.19                  | 1.84±0.12        | 0.92±0.06                        | 60.2±0.2                   | 147±1            | 3.04±0.01                        |

**Table 2.** Parameters characterizing the stress response of packings of various shapes: maximum angle of stability ( $\phi_m$ ) from avalanche experiments, and effective modulus ( $E$ ), yield stress ( $\sigma_y$ ) and yield strain ( $\varepsilon_y$ ) from triaxial compression as in Fig. 2 at two different confinement pressures ( $\sigma_{con}$ ).

sharp points (tetrahedra) or significant protrusions (long-armed jacks), on the other hand, even after yielding produce significant further increases in stress.

From these data, several aspects emerge: For given confining pressure, particle shape can indeed be used to tune and optimize the aggregate response within factors of two or three as far as properties like compressive modulus or yield stress are concerned. However, there is no obvious reason for why certain shapes perform better than others, and simple shape variations do not produce dramatic changes in aggregate properties. This holds for convex shapes such as Platonic and Archimedean solids, which attracted much recent research concerning their packing properties, as well as non-convex shapes such as ‘jacks’ (hexapods with arms of various length) and ‘dolos’, two particle types used in huge sizes for breakwaters (**Fig. 1**). For many shapes, furthermore, changes in confining pressure also induce changes in performance vis-à-vis other shapes. This becomes apparent by comparing the ordering of the shapes in **Table 2**: The maximum angle of stability is a proxy for the yield stress at the lowest confining pressure (basically the self-weight of the particles) and as the confining pressure is increased the ordering clearly changes, implying that certain shapes perform best at specific pressure ranges. This highlights the complexity of the problem and the importance of accounting for preparation and boundary conditions when attempting to optimize.

A convenient way to visualize the relationship between effective modulus and yield stress is to present the data as an Ashby-type plot (**Fig. 3**). To the extent that the particle shapes used to compile **Fig. 3** and **Table 2** represent a sufficiently wide range of different geometries, the data coverage defines a region of typical material response to applied compressive stress and is useful for selecting suitable shapes.

*This work was published in Soft Matter (A. G. Athanassiadis, M. Z. Miskin, P. Kaplan, N. Rodenberg, S. H. Lee, J. Merritt, E. Brown, J. Amend, H. Lipson, and H. M. Jaeger, Particle shape effects on the stress response of granular packings, Soft Matter 10, 48–59 (2014). doi: 10.1039/c3sm52047a) and selected for the cover (Fig. 4).*

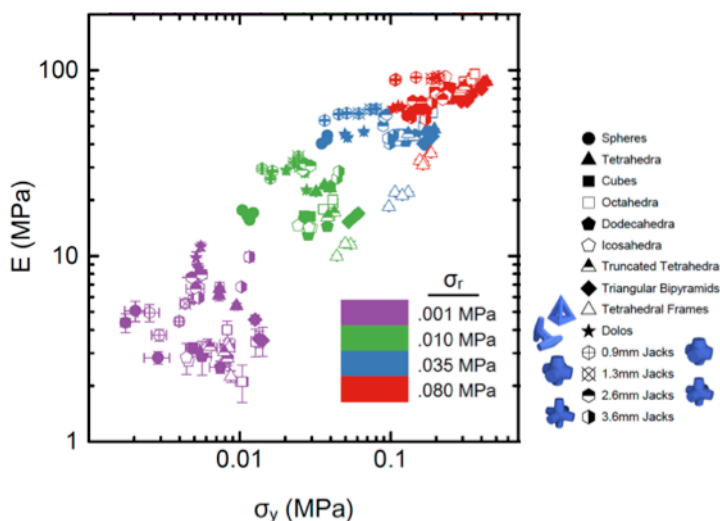


**Fig. 3.** Compressive modulus vs. yield stress from curves as in Fig. 2.



**Fig. 4.**

#### 4.2.2 X-ray imaging of jammed granular aggregates during mechanical loading

The results described in the previous section detailed the global response of the jammed aggregate, but they did not reveal specific microstructural (re-) configurations during loading. To track this internal microstructural evolution we developed a versatile x-ray tomography system that can be combined with the triaxial load tests.

What has been lacking in granular mechanics research is a versatile x-ray system that can be combined with existing materials testing equipment *and* can image the large samples used in such equipment

(typically 50–75 mm in diameter and 100–150 mm tall). We developed such a combination, based on a small, mobile, medical x-ray system called a mini C-arm (**Fig. 5**). In a C-arm, the x-ray source and detector are located at the two ends of a C-shaped metal beam that can be positioned as needed in 3D. These machines typically are used in image-guided medical procedures, where the open side of the “C” can easily be positioned to fit around the body part to be imaged. With large C-arms, computer controlled rotation of the “C” around the patient allows for 3D tomography; with mini C-arms, the beam position must be adjusted manually and tomography is not performed in a medical setting. Instead, mini C-arms are typically used to perform quick diagnostics or monitor the positioning of catheters during surgical procedures. Recently, medical C-arms have been used outside of hospitals as well; Dan Goldman and co-workers at Georgia Tech used projection images from a mini C-arm to track the movement of small lizards (“sandfish”) through granular media [14].

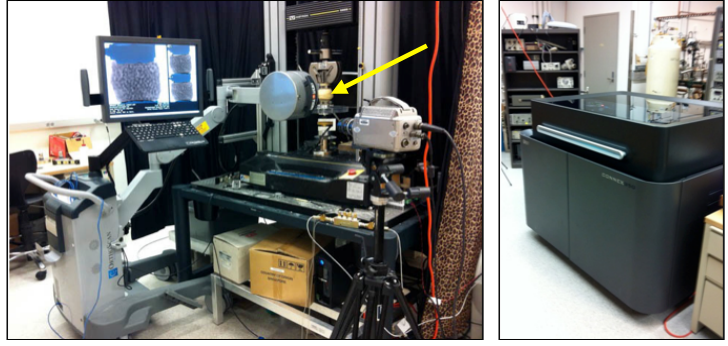


**Fig. 5.** Left: 2-column Instron materials tester (center) with C-arm x-ray system imaging a granular packing (arrow) as it is compressed. High-speed video camera (at right) records deformations visible on the outside. Right: Objet Connex 350 3D-printer in the PI’s lab.

While these examples demonstrate how a fixed mini C-arm can be used for 2D-projection data, tomography of a static sample is also possible if the sample is rotated in front of the beam. Our x-ray tomography system integrates an Orthoscan mini C-arm with a custom sample rotation stage. It is combined with an Instron 5800 series materials tester that we use to strain the sample while measuring stress. This setup, shown schematically in **Fig. 6**, allows us to perform tomographic analysis of the local internal structure of samples at any stage of a mechanical loading process. Advantageously, this combination provides a large field of view— $(6\text{ cm})^3$ —reconstructed to resolve  $<520\text{ }\mu\text{m}$  features, together with the ability to measure forces up to 50 kN and displacements as small as  $10\text{ }\mu\text{m}$ . With this setup, we demonstrated the ability to track microstructural changes in relation to the exact stresses exerted on the sample.

The x-ray tomography system was built and extensively calibrated. Particular attention was paid to the fact that the x-ray beam is cone shaped, and this needs to be considered in the image reconstruction. First experimental runs with the completed system were performed to ascertain the resolution of reconstructed images, and here specifically the ability to cleanly detect ages of arbitrarily shaped particles.

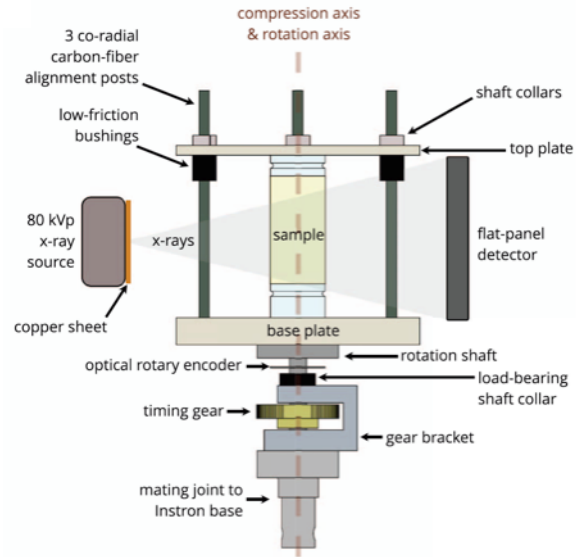


**Fig. 6.** Schematic of the rotation stage built for the C-arm x-ray system.

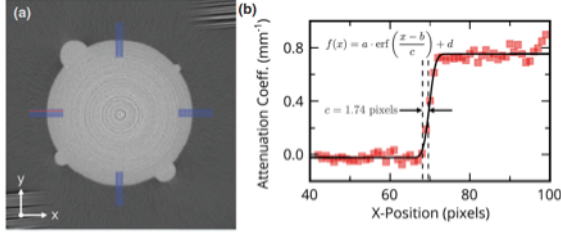


**Fig. 7.** Spatial resolution of the C-arm based x-ray tomography system. a) Tomographic reconstruction of the phantom. b) Edge profile and fit to error function.

**Figure 7** provides details about the imaging resolution. To measure our system's spatial resolution, we reconstructed a 3D-printed phantom shown in (a), and extracted edge profiles from different points around the perimeter (highlighted in blue and red). Fitting each profile to an error function, we extracted the width

parameter,  $c$ , which give the resolution of our system with respect to the ability to clearly identify edges. In (b), the red profile from (a) is plotted. Red squares represent the measured intensity values along the profile, and the black line denotes an error function fit. **Figure 8** shows an example of a reconstructed jammed packing of 3D-printed particles strained to 4.5%.

*A paper describing this new, versatile x-ray system was published in Reviews of Scientific Instruments (A. G. Athanassiadis, P. J. La Rivière, E. Sidky, C. Pelizzari, X. Pan, and H. M. Jaeger, X-ray tomography system to investigate granular materials during mechanical loading, Review of Scientific Instruments 85, 083708 (2014). doi: 10.1063/1.4893555).*

#### 4.2.3 Adapting Granular Materials Through Artificial Evolution

Since particle shape determines both the packing arrangement and the network of contact interactions in a granular material, selecting a particular particle shape can select a particular mechanical response. However, there currently are no general rules to predict the behavior of a packing as particle shape is modified. Simulations can start to address this issue, but in trying to explore, and exploit, the role of particle shape, the problem faced is the overwhelming size of the search space. The key task, therefore, is to find a way to parse the vast space of possible shapes efficiently, taking account of both the desired goal and the given boundary conditions.

This research effort grew out of earlier work in which we asked whether the phenomenon of granular jamming could be utilized for unique applications and, in particular, as a new enabling technology for soft robotics. The prototype systems in these first, proof-of-principle applications relied on the jamming of generic granular material, such as ground coffee or glass beads. Thus, a naturally emerging

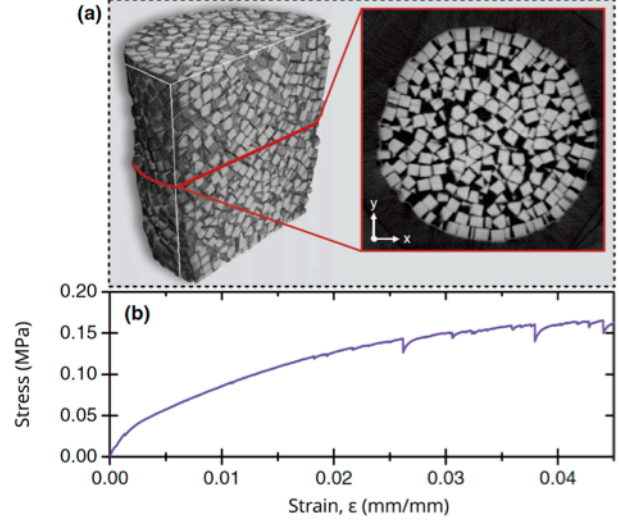


**Fig. 8.** a) Tomographic reconstruction of a jammed packing of 3d-printed cubes. b) Stress-strain curve.

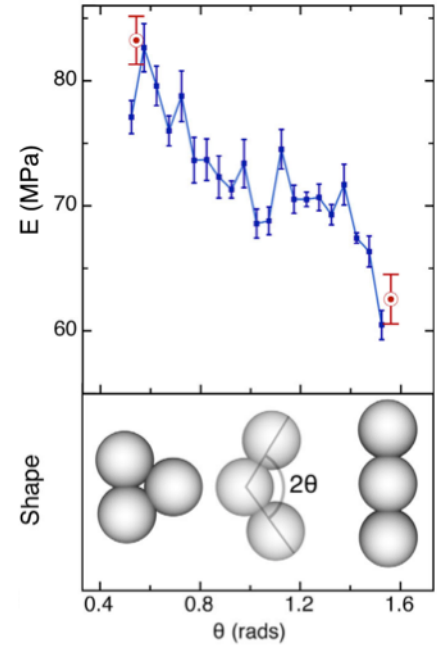


**Fig. 9.** Effective modulus under triaxial compression at 80kPa confining pressure for compound particles made from 3 adjoined spheres. For such trimers the full range of shapes can be parameterized by the opening angle  $\theta$ , as indicated.

question was: What kind of particle should be used to optimize the jamming behavior? This question directly fed back into basic research and, under this grant, led to a new approach toward finding optimized particle shapes.

The key difficulty in expressing the role of particle geometry in driving certain behaviors of granular material lies in the fact that shape is an inexhaustible parameter. Therefore, trying to isolate particular shapes that generate a given desired response might appear infeasible. **Figure 9** shows how already for simple shapes, here a compound particle or ‘granular molecule’ composed of three rigidly bonded solid spheres (‘trimers’), the effective modulus  $E$  of the aggregate material exhibits a very complicated shape dependence. Even though in this particular case shape is controlled by only one free parameter, the opening angle  $\theta$ , the detailed relationship  $E(\theta)$  is highly non-intuitive.

For more complicated shapes, the number of parameters very quickly becomes so large and the associated optimization space so high-dimensional and also ragged, that the task of identifying a shape that would produce the particular, desired value for  $E$  becomes highly non-trivial. However, we found that optimizers based on evolutionary strategies vastly outperform standard search methods (such as simulated annealing) and make it possible to find optimal or at least near-optimal particle shapes.

To apply this technique, two components are essential. The first is a genome, or means of encoding solutions, so that mutations can be freely applied. The second is a fitness metric, i.e., a means of determining which potential solutions are better than others. Once these two components have been established, an optimization algorithm can be constructed to efficiently search the solutions space. Specifically, we chose the Covariance Matrix Adaptation Evolution Strategy (CMA-ES), developed by Hansen et al. Like most evolution strategies, this algorithm attempts to improve the quality of solutions by examining random perturbations around a given mean solution. The CMA-ES draws these guesses using a multivariate Gaussian distribution. The key feature of this algorithm is that it uses information from prior iterations of the search to deterministically update the mean and covariance matrix. Specifically, the mean of the distribution is updated so that the likelihood of drawing a previously found good candidate is maximized, whereas the covariance matrix is updated so as to increase the probability of a successful next step.



**Fig. 10.** ‘Granular Molecules’, each 3D-printed as one piece from hard plastic. Each sphere is  $\sim 3\text{mm}$  in diameter.

For simplicity, we represented particles as granular molecules, i.e., sets of equal-sized spheres that overlap a fixed distance. The main reason was that this limited the type of particle-particle contacts to sphere-sphere contacts that can be handled straightforwardly in standard DEM simulations, as shown previously in work using such ‘bonded-sphere’ compound particles as a means to explore non-sphericity and aspects of (geometric) friction. Different particle shapes were represented by  $N$  spheres placed in configurations specified by a construction protocol we call a blueprint. **Figure 10** shows examples of physical realizations of specific granular molecules with  $N = 3, 4$ , and  $5$ . Note that these configurations do not have to be planar, and for  $N > 3$  can extend in all three dimensions. For example, for  $N = 5$  a blueprint can specify configurations such as the asymmetric, wishbone-like particle shown on the right.

*Implementation.* In comparing our evolutionary algorithm to other optimization techniques, we found that the CMA-ES outperforms alternatives by requiring significantly fewer function calls to identify a solution. A good

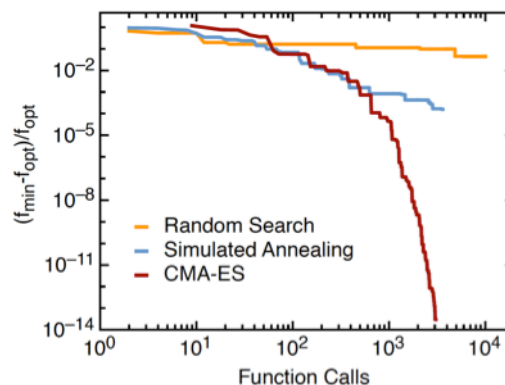


**Fig. 11.** Benchmarking: Distance to global minimum vs. number of function calls.

benchmark test is to find, starting from random sphere configurations, the arrangement that minimizes the sum of the distances to the center of mass. For objects made with  $N < 13$  spheres, the correct solutions are polyhedra with triangular faces. This problem, previously studied by Sloane *et al.*, is remarkably challenging since the number of local minima increases rapidly with  $N$ . Having tested our algorithm up to (so far)  $N = 8$ , it always found the global optimum. **Figure 11** compares the performance of CMA-ES, simulated annealing, and random search in identifying the correct sphere configuration, here for tetrahedra. We plot the relative difference between the best solution found and the global optimum against the number of function calls required. Already after about 500 calls, CMA-ES begins to dramatically outperform simulated annealing. Since each generation typically uses about 10 function calls (one for each member of the population), this corresponds to just over 50 generations. The population size required in CMA-ES for reliable performance scales as  $\log(N)$  and thus only grows slowly with increasing complexity of the simulated object. Furthermore, since function calls can be sent out in parallel, larger populations do not necessarily slow down the optimization.

When used to optimize particles in this project, the function calls at each generation will start DEM simulations to run ‘virtual experiments’. Each of these virtual experiments is a standard ‘forward’ simulation with input parameters provided by CMA-ES, and based on the particles’ performance the evolutionary algorithm will iterate the process with changed parameters. For this work we used PFC3d from Itasca (Minneapolis, MN) under an educational license agreement, but other DEM codes are viable alternatives (we currently use LAMMPS). Typical simulations so far use up to 2,000-3,000 compound particles per packing. PFC3d has built-in functionality for arbitrarily shaped compound particles composed of spheres as building blocks. It also allows us to straightforwardly simulate various initial and boundary conditions as well as experimental protocols (pouring under gravity, tapping, cyclic compression, triaxial test, etc.). With a fast, multi-core Mac desktop running PFC3d on Windows, finding optimized compound particles takes from 1 day to ~1 week, mainly depending on the complexity of the ‘forward’ DEM simulation.

**Key results.** We used blueprints as our genotypic expression for particle shape, and evolved populations by mutating the sphere configurations. Taking the performance in DEM simulations of materials tests, such as triaxial tests, as a fitness metric, we applied this evolutionary approach to

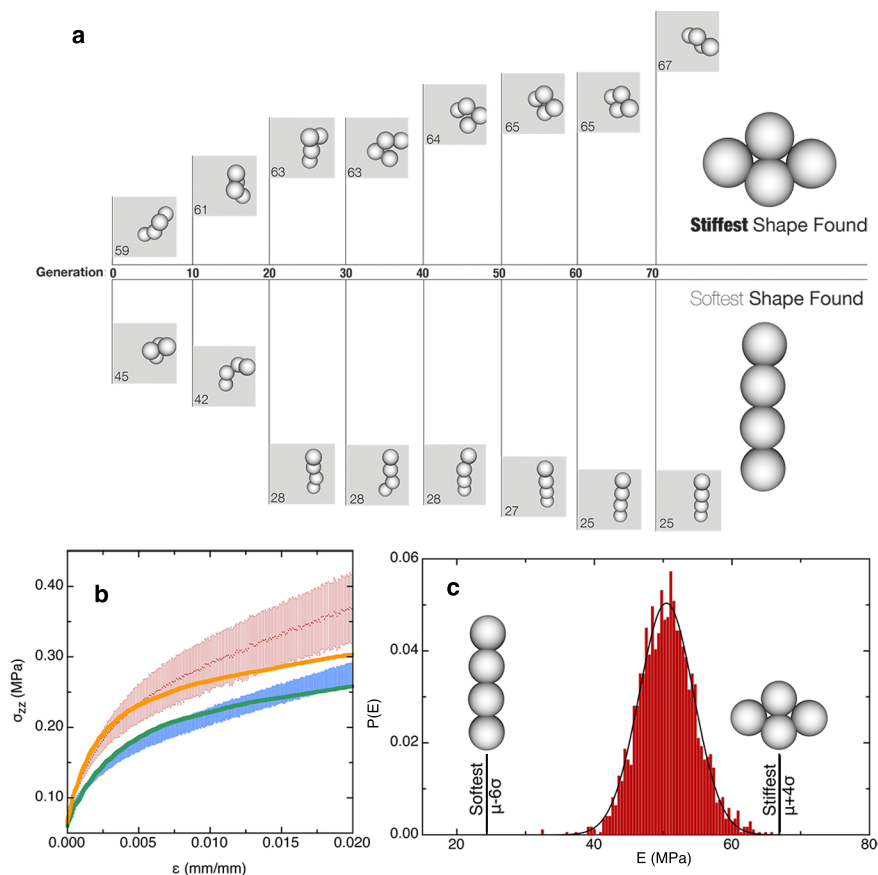


**Fig. 12.** Optimizing the stiffest and softest packings for particles made from 4 spheres. (a) Parent shapes and their fitnesses leading up to the optimal design. (b) Measured and simulated (solid lines) stress strain curves. (c) Distribution of modulus  $E$  resulting from random blueprints (mean 51MPa, standard deviation ~4MPa), with the discovered extrema shown for comparison.

jammed particle packings. We show here results from two initial tests: to find the particle shapes that form packings that, when confined by a given pressure, produce the stiffest response and the softest response to compressive loading. That is, shapes which produce packings with the largest (smallest) elastic modulus  $E$  near zero strain. For these tests, we fixed  $N = 4$ .

**Figure 12a** shows the parent shapes that ultimately lead to the best shapes found. After convergence, we used 3D-printing to fabricate particles with those best shapes, constructed physical packings, and used the experimental data from triaxial compression tests to validate the simulated stress strain curves. The significant differences between the shapes producing the stiffest and softest response shows that the evolutionary algorithm makes good use of the available configuration space. **Figure 12c** emphasizes this point with a histogram of moduli for simulated packings of  $N = 4$  particles composed of *randomly* configured spheres. The shapes with the stiffest and softest response have moduli 4 and 6 standard deviations from the mean, respectively, demonstrating that the evolutionary approach to granular design can be both accurate and efficient.

The link between contact locking and stress response invites more exotic design goals. For example, in packings near the jamming point, prior work has shown that the potential mobility of a given contact can depend strongly on the geometry of the particle [15]. Now, suppose a material was initially in a state characterized by weak particle contacts, but as these contacts were broken, new contacts formed that were stronger. Such material would stiffen the further it was compressed, in contrast to the strain softening common for granular materials (note that in triaxial tests the lateral confinement is not via a hard, immobile wall, but via a surface held at constant pressure). We used our algorithm to search for appropriate shapes, i.e. shapes producing the largest second derivative of stress with respect to strain. The evolutionary strategy in this case led to the discovery of a unique, asymmetric particle that, with just 5 constituent spheres, produces a concave-upward dependence for  $\sigma(\epsilon)$  (**Fig. 13**). This strain-stiffening behavior differs qualitatively from the stress-strain relationship found in ordinary granular materials under the same confinement conditions.

Strain stiffening was recently found also in packings of granular polymers (linear chains of flexibly linked beads) if the polymers were long enough. However, the wishbone-like granular molecule in **Fig. 13** is not only rigid but also smaller, i.e., consists of fewer spheres, than granular polymers that strain stiffen. This relatively small molecule achieves remarkable self-confinement under load: it can sustain stress levels 10 times larger than the confining pressure (**Fig. 13**). We also note that these packings exhibit a low modulus, yet do not fail up to 10% strain, an order of magnitude larger than simple spheres under the same conditions.

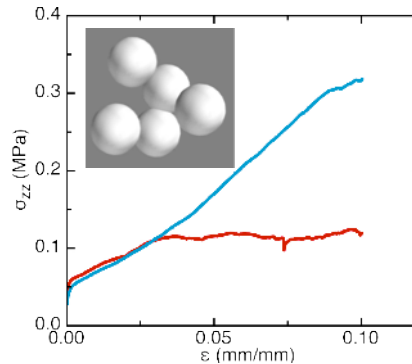


**Fig.13.** Measured stress-strain curve for aggregate of 3d-printed  $N = 5$  particles designed to strain stiffen (inset: rendered single particle; cf. image in Fig. 10 on the right). For comparison, data for  $N = 2$  ‘dimer’ particles are shown in red.

These results are a first glimpse of the power of an inverse method based on evolutionary strategies. The key point is that this approach very effectively works in a parameter space traditionally deemed too complex to find solutions. *This opens up a whole new way of approaching the design of structures composed of granular materials: while the previous state-of-the-art relied on trying out various shapes in an Edisonian fashion, we can now start with the desired goal, namely a specified response for the granular material as a whole, and find the particle shape that will enable this response* [11].

*This work was published in Nature Materials* (M. Z. Miskin and H. M. Jaeger, Adapting granular materials through artificial evolution, *Nature Materials* **12**, 326-331 (2013). doi: 10.1038/NMAT3543).

#### 4.2.4 Designing the Packing Density of Granular Materials Through Artificial Evolution

As a next step, graduate student Marc Miskin used the evolutionary algorithm for finding optimized solutions for a different class of problems, namely the packing of particles. It is easy to appreciate the fact that granular aggregates can have different physical properties if the particle shapes are different. For example, it is known that the mechanical strength and stiffness of a granular packing are both influenced by particle shape. This follows since particle geometry directly determines both how grains arrange themselves when packed and how stresses propagate throughout the aggregate. Yet at present, there is no straightforward way to exploit this relationship and find shapes that produce aggregates with specific, desired target stiffnesses and/or strengths.

The same problem manifests itself when examining the transport properties of granular packings. Here, microscopic features like the structures of interstices and the number of contacts per particle exert a strong influence on macroscopic properties like fluid permeability and thermal/electrical conductivities. Again, it is clear that these bulk properties are intimately connected with particle shape, while it remains unclear how to leverage this knowledge to select shapes that tailor aggregates to have preferred behaviors.

The heart of this issue can be put into stark contrast with two, deceptively similar, questions. First, “Are there particle shapes that pack randomly to different densities?” Clearly the answer is yes. Yet change the question to “which particle shapes pack randomly to a density of  $X$ ?” and a general answer is unknown.

In short, there is an apparent disparity between the number of relationships linking particle shape to aggregate behaviors, and the capacity to transform these same relationships into design rules. In light of this, answering a question like the latter one above is tantamount to inventing a method that generically solves the “inverse problem” of associating shape to response. For problems like this, an aggregate’s behavior is pre-specified and the goal is to find a particle geometry that produces this behavior.

**Key results.** We demonstrated a solution to this inverse-packing problem by framing it in the context of artificial evolution. By representing shapes as bonded spheres, we showed how shapes can be mutated, simulated, and selected to produce particularly dense or loose packing aggregates, both with and without friction. Moreover, we showed how motifs emerge linking these shapes together. The result is a set of design rules that function as an effective solution to the inverse packing problem for given packing procedures and boundary conditions. Finally, we showed that these results are verified by experiments on 3D-printed prototypes used to make packings in the real world.

For a given granular inverse problem, we believe a true solution should come in the form of “design rules.” By this, we mean a set of instructions that can be used in and of themselves to easily associate particle shapes to any aggregate behavior that falls within an achievable range. We contrast this with seeking a single geometry that solves a single inverse problem. We seek rules that can be followed to generate solutions to all the inverse problems that can be posed within a specific class.

We introduced a new method that discovers such design rules. This method is built on a framework we introduced earlier that combines artificial evolution, computer simulation, and 3D printing to explore the role of shape in granular matter (see Section 4.2.3). In this framework, we generate different shapes by rigidly bonding spheres of different diameters into molecule-like compound particles. Given a “granular molecule”, we use discrete element method (DEM) simulations to calculate the properties of its resulting aggregate, and we use these calculated properties to categorize and compare shapes. This allows us to determine how close a given “molecule” comes to achieving a user-specified goal. By selecting and mutating the shapes that come the closest to achieving said goal, we can efficiently and automatically explore the role of particle shape.

In prior work we used this method to identify particles that produce minimum and maximum responses (e.g. particles that produce the stiffest/softest packings). Here we extended this process to build design rules by generating a path of shapes that leads through response space from the minimum response to the maximum response. This path effectively operates as a “design rule solution,” at least to the extent that

the evolutionary process produces a close approximation to the range of possible behaviors. The power to achieve this goal comes from our shape representation. Bonded spheres with variable sphere radii are a design space in which any one shape can be morphed into another. By extension, any response can be morphed from one to another, provided the connection between shape and response is continuous.

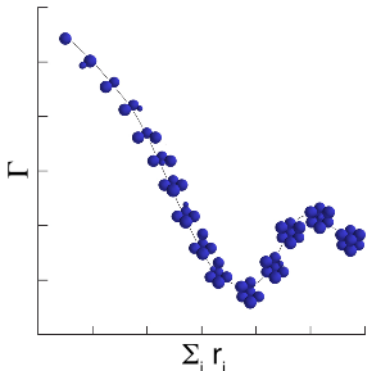


**Fig. 14:** Schematic illustrating how a series of deformations in shape can lead to a path of different aggregate responses  $\Gamma$ . In this case, a cube-shaped granular molecule is being transformed into a sphere by systematically sending the constituent sphere radii,  $r_i$ , to zero, until a single sphere remains. Given an order for removing the spheres, each shape in the path is uniquely specified by the sum of all radii in the granular molecule. Thus, if desired, this graph could be used to solve an inverse problem for the aggregate property  $\Gamma$ : simply draw a horizontal line from the y-axis to the intercept with the response space graph to locate the appropriate shape.

This process is illustrated schematically in **Fig. 14**. Suppose, for example, that a cubic granular molecule needed to be morphed into a sphere. While there are infinitely many ways to accomplish this goal, a direct and convenient approach is to send the radius of each sphere in the cube to zero, one at a time, until a single sphere remains. In other words, one labels each of the spheres in cube arbitrarily, uses the labels to specify an order, and then shrinks the spheres one at a time until a single sphere remains. The advantage of this procedure is that once the order of elimination is specified, a single parameter, i.e. the sum of all sphere radii in the molecule, specifies each intermediate shape uniquely. By making a plot comparing this “shape index” to an aggregate property, designated in Fig. 5 by  $\Gamma$ , a graphical map emerges that links any response in its range to at least one shape. Because the shape indices effectively encode the particle geometries, this map is a design rule that functions as an independent inverse problem solution.

With this in mind, the best design rule is the one that extends the range of responses to be as large as possible. This rephrases the inverse problem as an optimization problem that can be solved via artificial evolution: find the shapes that produce the minimum and maximum responses. The design rule then emerges from considering the path, as in **Fig. 14**, that continuously transforms these two extreme shapes into one another. As a concrete example, we showed how to find particle shapes that pack randomly to a specified target density. Since packing fraction is known to depend on both shape and on the role of friction, we considered both packings that are poured in place and packings that are tapped until reaching an asymptotic density. In simulations, we approximated these loading protocols by pouring particles into a box under gravity with frictional and frictionless contact force models.

As the first step to answering this “granular inverse packing problem”, we used our evolution framework to identify shapes that generate particularly dense and particularly loose packings when poured under gravity. We allowed our optimizer to make granular molecules from a maximum of 10 constituent spheres of variable radii. **Figure 15** shows the median fitness for the simulated populations, under our two loading procedures at each generation. After roughly 50 generations, our algorithm converged when the simulations included friction (**Fig 15a**) and roughly 100 generations when the simulations were frictionless (**Fig 15b**).

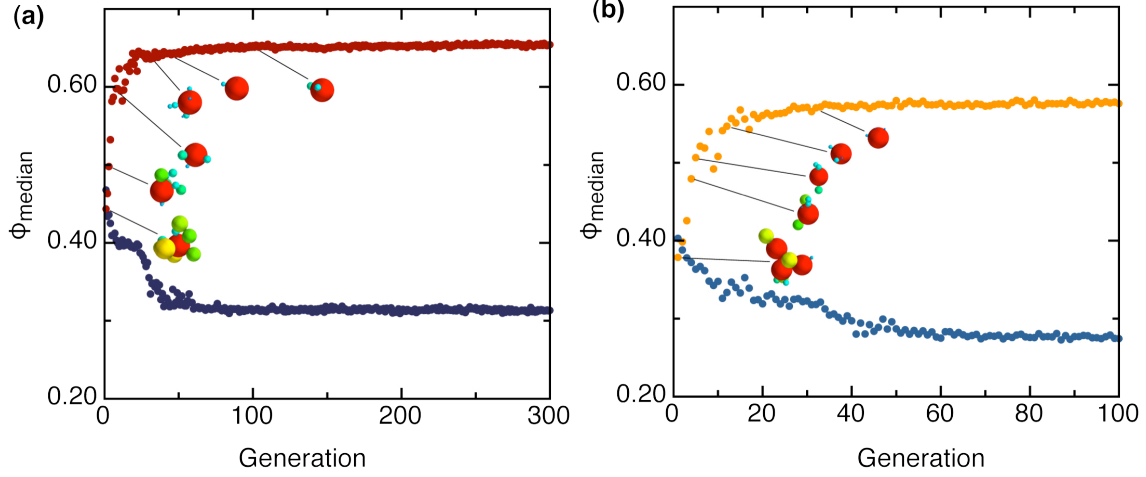


**Fig. 15:** Evolving the densest and loosest packing shapes. Starting from random configurations, our evolutionary algorithm adapted particles to pack densely and loosely when poured under gravity without friction (a) and with friction (b). The median packing densities are plotted against generation number. For comparison, we also include call outs to some of the shapes that generated these median values. Note how our evolution process essentially filters out complicated, but suboptimal shapes to discover surprisingly simple shapes as extreme packers.

After starting from the random, un-adapted configurations (**Fig. 15**), our algorithm identified three distinct geometries (**Fig. 16**): a sphere, a trimer, and 10mer rod. The trimer shape is symmetric, made from two secondary spheres with radii 0.3 the size of the main sphere, and a bond angle of approximately  $70^\circ$ . These shapes have been adapted to pack densely with friction, densely without friction and loosely, respectively.

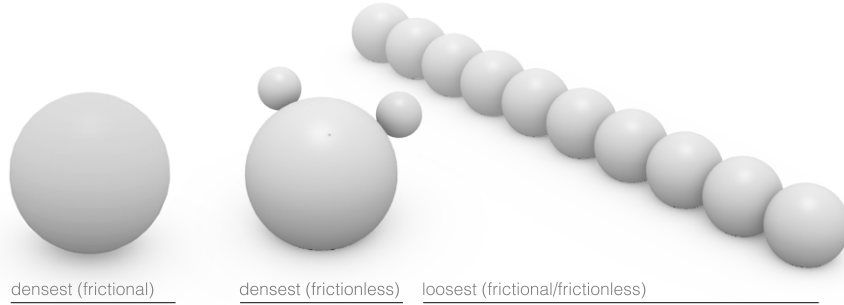


**Fig. 16:** Three extreme packing shapes. The optimizer converged to these three shapes as the particle shapes that generated the most extreme packing densities, given our loading protocol and material parameters. While the rod and trimer exhibit several symmetries, so as to appear relatively simple, these are emergent features. They were not programmed as design goals, but rather came about through generations of random mutation and selection.

To verify the results of the optimizer, we 3D-printed particles and measured their packing fractions under the appropriate loading protocols. For rods, our simulation predicted packing fractions of  $0.271 \pm 0.009$  and  $0.313 \pm 0.002$  with and without friction. In experiments, we measured packing fractions of  $0.26 \pm 0.03$  and  $0.31 \pm 0.01$  for the as poured packing and the tapped packing, respectively. For spheres, our frictional simulations predicted a packing fraction of  $0.578 \pm 0.002$ . Using the “as poured” experimental procedure we measured a volume fraction of  $0.59 \pm 0.02$ , again within error. Finally, we measured a density of

$0.67 \pm 0.03$  for tapped packings of our trimer particle, within error of the simulation prediction,  $0.658 \pm 0.003$ .

For the particular optimization goal of finding shapes that minimize or maximize packing density when poured under gravity, the evolution process reduced much of the shape complexity that would have been possible using all 10 spheres. Perhaps surprisingly, all three shapes are relatively simple granular molecules. Others have identified two of the shapes, rods and spheres, as extreme packing shapes. When considering loose packings prepared by fluidization, Baker and Kudrolli demonstrated that spheres pack denser than any of the Platonic solids [16]. For rods, it has been reported that the volume fraction for an aggregate can be made arbitrarily low by increasing the number of monomers. The fact that the optimizer used all 10 spheres in constructing the rods suggests that here too the packing fraction could have been made lower had we allowed the optimizer more constituent elements.

Perhaps the most surprising shape identified was the trimer particle. We anticipated cubic or tetrahedral granular molecules to be the densest packing shapes without friction. Yet by directly simulating these shapes with our loading procedure and material parameters, we found them to be suboptimal: cubes and tetrahedra made from equal sized spheres packed to 0.465 and 0.641, respectively.

However, we also note that the trimer shape is not the densest packing particle out of all shapes: ellipsoids pack randomly to a density up to 0.74. We speculate that this discrepancy comes from the fact our optimizer was constrained to build particles out of bonded spheres with no overlap. Such shapes always have surface concavities, which may preclude crucial mechanisms, like sliding, that promote dense packing for smooth shapes like ellipsoids. Schreck and O’Hern demonstrated that, in 2D, a pronounced difference in packing fraction between dimers and ellipsoids results when the aspect ratio exceeds roughly 1.2 [17]. It would be straightforward to extend the approach outlined here to evolve smoother geometries by including particle overlap as a mutable parameter. Yet here, the optimizer had to design a particle that packs densely by only exploiting the typical rearrangement mechanisms available to particles made from bonded spheres.

The optimizer’s choice became clear after we examined the void space left in a packing of frictionless spheres. Specifically, we studied points that were equidistant to each particle in a set of nearest neighbors (see sketch in **Fig. 17a**). If a sphere were placed at one of these points and expanded until just touching all the neighbors, it would be as large as possible without requiring the packing to restructure, making each of these sites local maximum for the size of a spherical void. Therefore, designing granular molecules that fill these voids can potentially be a good strategy to maximize packing density, provided the particles are free to explore different local configurations.

To test this hypothesis we took a packing of spheres of radius  $a$  and calculated all such local-maximum void sizes and simulated appending spheres to fill them. We skip any void that overlaps with a void that has been previously added. By making a histogram of the appended sphere radii,  $d$ , we found that the most probable void size is  $0.3 a$  (**Fig. 17a**). This is the same size ratio as in the trimer particle. What’s more, we found a surprising structure in the pair correlation function between these voids. Specifically, we found a local maximum in the probability density corresponding to two voids distributed about  $1.5 a$  apart (**Fig. 17b**). Assuming a void size of  $0.3 a$  and that these two voids both touch the same sphere, this would correspond to a bond angle of  $70^\circ$ . This is the same bond angle as in our trimer. While there is also a second peak at  $2.5 a$ , two  $0.3 a$  appendages can at most be spaced  $2.3 a$  apart, making it impossible to exploit this correlation. Evidently, our trimer was designed to exploit features lurking in an ordinary, random packing of spheres.

Combined, the experimental validation and the theoretical rationalization of the shapes found by the optimizer provide a reasonable basis to accept them as good approximations to the true densest and loosest packing shapes. As the next step, we considered how to deform one into the other, and how to use this deformation as a set of design rules.

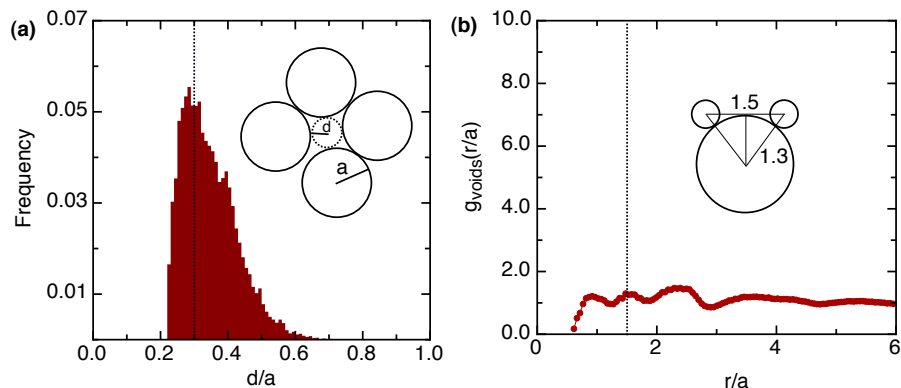


**Fig. 17:** Filling the voids of a sphere packing. To rationalize the geometry selected by the optimizer as the densest packing shape, we considered the size (a) and pair correlation (b) of voids in a random packing of spheres of radius  $a$ . Voids have a most probable size of  $0.3 a$  and are distributed roughly  $1.5 a$  apart. The optimizer picked granular molecules consisting of a central sphere with two nubs that nicely fit into these voids (inset in b).

First, we deformed our rod shapes into spheres by sending the sphere radius at the end of the rod to zero, one at a time. We then took the trimer particle and send the secondary spheres to zero radius, again one at a time. These two sets of deformations overlap once the mutated shapes become dimers and are transformed into spheres. Note that, while there are infinitely many ways to perform this deformation, this choice provides a convenient way to index shapes. By systematically sending the furthest spheres to zero, a single number, the sum of all radii in a particle, marks each shape uniquely within the deformation path. For example, if the sum of all radii is 4.2, we know that the shape in question is a linear configuration of four  $a=1$  spheres plus one sphere with  $a=0.2$  sitting at the end.

For each of these mutated shapes, we ran a corresponding DEM simulation, and in **Figs. 18a** and **18b** we plot the resulting packing fractions against our “shape index,” the sum of the radii in the particle. In **Fig. 18a**, the rods form a lower branch spanning all the packing fractions from 0.65 to 0.3 while the trimers yield a second branch, spanning packing fractions from 0.64 to 0.66. For the frictional simulations in **Fig. 18b**, rods alone span every packing fraction up to 0.59. Thus, every packing fraction between the minimum and maximum discovered by the optimizer can now be mapped to at least one shape. Furthermore, the shape’s geometry can be simply read off the graph, given our deformation procedure.

As an illustration of this inverse method in action, we picked an arbitrary packing density, in this case 0.445. Suppose that one wants to find a particle that produces this packing fraction when poured in place. As shown in **Fig. 18b**, the intercept between 0.445 and our response curve occurs for a shape with a shape index of about 3.5. Because the response curve was constructed by sending each sphere to zero sequentially, there is only one shape with this value: a linear particle with three identically sized spheres plus a fourth at the end with one half the radius. We 3D-printed this particle and measured its as poured packing density. We found, as desired, a packing density of  $0.45 \pm 0.01$ .

One can note that our inversion method is not one-to-one. For example, the shape index vs. packing fraction graph for frictionless packings exhibits non-monotonic behavior. Thus, a designer trying to find particles that pack to densities between 0.64 and 0.65 may choose from either an asymmetric dimer or trimer geometry. From a practical point of view, this degeneracy is unimportant. Both geometries answer the same granular packing problem and so a designer may feel free to choose either one to pack to the desired density.

We contrast simple design rules and the simple shapes they lead to against the other shapes considered by the optimizer while perusing the extrema. In **Figs. 18c** and **18d** we plot all the points it explored in

finding the minimum and maximum response shapes with call outs to some of the curious geometries that generated them.

It is clear that the optimizer parsed through a tremendous number of exotic and complicated geometries. This large degree of diversity is indicative of how effectively artificial evolution, as a process, filters a broad number of possibilities into an outstanding subset. The optimizer had sufficient evidence to select the shapes in **Fig. 16** as a basis for design rules only after parsing this wide range of potential configurations.

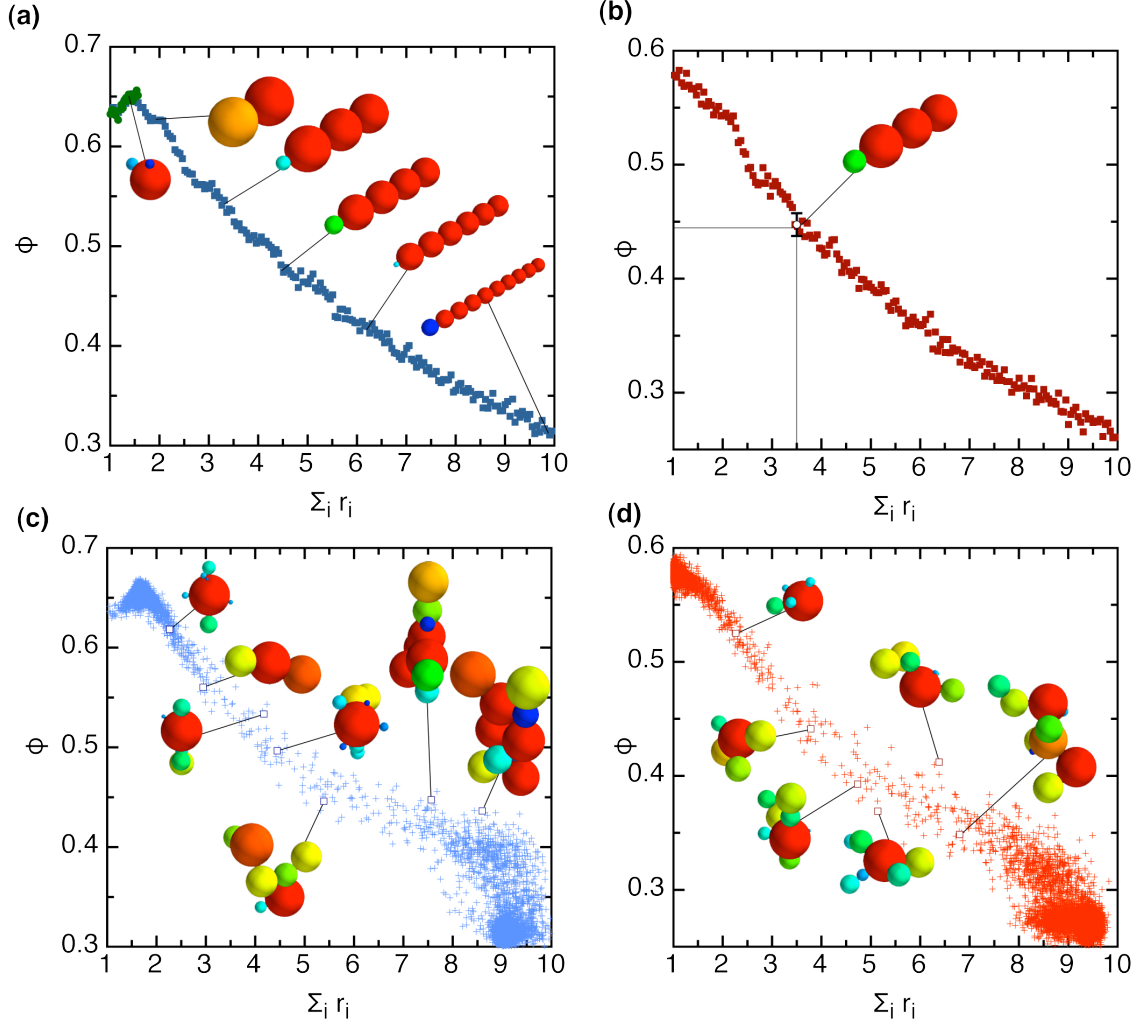


**Fig. 18:** Inverting a packing problem. Because the optimizer identified rods, spheres and a trimer as extreme packing shapes, we used these as a basis to build an inverse problem solution. Specifically, we deformed rods (blue in a and red in b) and trimers (green in a), by sending each sphere radius to zero sequentially and simulating the resulting shapes both without (a) and with (b) friction. This procedure generates the plotted paths that can be used to solve inverse problems graphically (b). While this procedure is extremely simple, it is founded on the immense search procedure performed by the optimizer (c,d). For both simulations with and without friction, the optimizer explored a tremendous number of complex and exotic geometries before ultimately converging on the shapes used as a basis for (a) and (b). Any one of these exotic behaviors could, a priori, have been optimal, illustrating the powerful simplifications created by our evolutionary method.

Had we considered packings formed by a protocol different from simple pouring, for example isotropic compression, we may have found different optimal shapes and different maximum/minimum volume fractions. As a case in point, the inclusion of friction produced different shapes as densest packers. Yet the optimizer operates independently from the simulation procedure at hand, and so these cases would be straightforward generalizations.

Furthermore, our inversion method, based on deforming shapes, can always be used to find a simple graphical map, provided the shape to response mapping is continuous. This is a powerful advantage: it condenses a complicated design problem into a simple set of instructions. Indeed, as shown here, Fig. 10, can be used by itself as a solution to the inverse granular packing problem, given material and loading procedures similar to ours.

*This set of results was published in Soft Matter* (M. Z. Miskin and H. M. Jaeger, Evolving design rules for the inverse granular packing problem, *Soft Matter* 10, 3708–3715 (2014). doi: 10.1039/c4sm00539b).

### 4.3 Unjammed Regime

New physics can be expected once the rate of forcing approaches the characteristic time scales associated with the nonlinear structural rearrangements in the unjammed system. This happens most typically in systems where the granular particles are suspended in a liquid and the liquid's viscosity introduces a characteristic time scale for flow. In particular, we expected that the nonlinearities quite generally can give rise to propagating density fronts: under an applied stress particles starting in a low-density configuration will collide and agglomerate locally into a configuration that is dynamically jammed (but might unjam once the stress is removed).

#### 4.3.1 Propagating Jamming Fronts

This part of the research effort resulted in a better understanding of the limitations of current models for shear thickening in dense suspensions and in the discovery of a new mechanism for the extraordinary normal stresses generated by such suspensions under normal impact. *The initial findings of this work were published in Nature* (Scott Waitukaitis and Heinrich M. Jaeger, "Impact-activated solidification of dense suspensions via dynamic jamming fronts", *Nature* **487**, 205-209 (2012). Doi: 10.1038/nature11187). A more detailed account was published as Scott Waitukaitis' Ph.D. thesis (S. Waitukaitis, *Impact-Activated Solidification of Cornstarch and Water Suspensions*, Springer International Publishing Switzerland, 2015. ISBN-13: 978-3-319-09182-2).

To produce significant shear or normal stresses, most explanations for the hardening observed in driven suspensions all require some confinement, via the presence of walls or boundaries. In models based on liquid-mediated formation of particle clusters these need to percolate between the shearing walls, and in models treating dense suspensions as granular, frictional fluids dilation must be counteracted by confinement, similar to "shear-jamming" in dry granular systems. The upper limit of (normal or shear) stresses under steady state shearing is then set by the stiffness of the particles or the boundary, whichever is more compliant. In prototypical suspensions of hard particles such as cornstarch in water, compliance of the interface with the surrounding air limits stresses to no more than 5-10kPa under shear or extension. The requirement for stiff boundaries is also problematic in light of strong shear thickening observable in large containers where poking (or stepping on) the suspension solidifies it locally but leaves most of its volume in a liquid-like state. Experiments by groups at the Courant Institute at NYU [18] and the University of Twente in the Netherlands [19] have suggested that jammed regions transmit stresses to the container bottom, but did not address how these regions form or how they evolve during impact. Using a large suspension volume (25 l) to track the growth of such regions with precision we demonstrated that, in fact, stresses onto the impactor in the MPa range can be generated dynamically ('dynamic jamming') and without requiring confinement or proximity to hard surfaces.

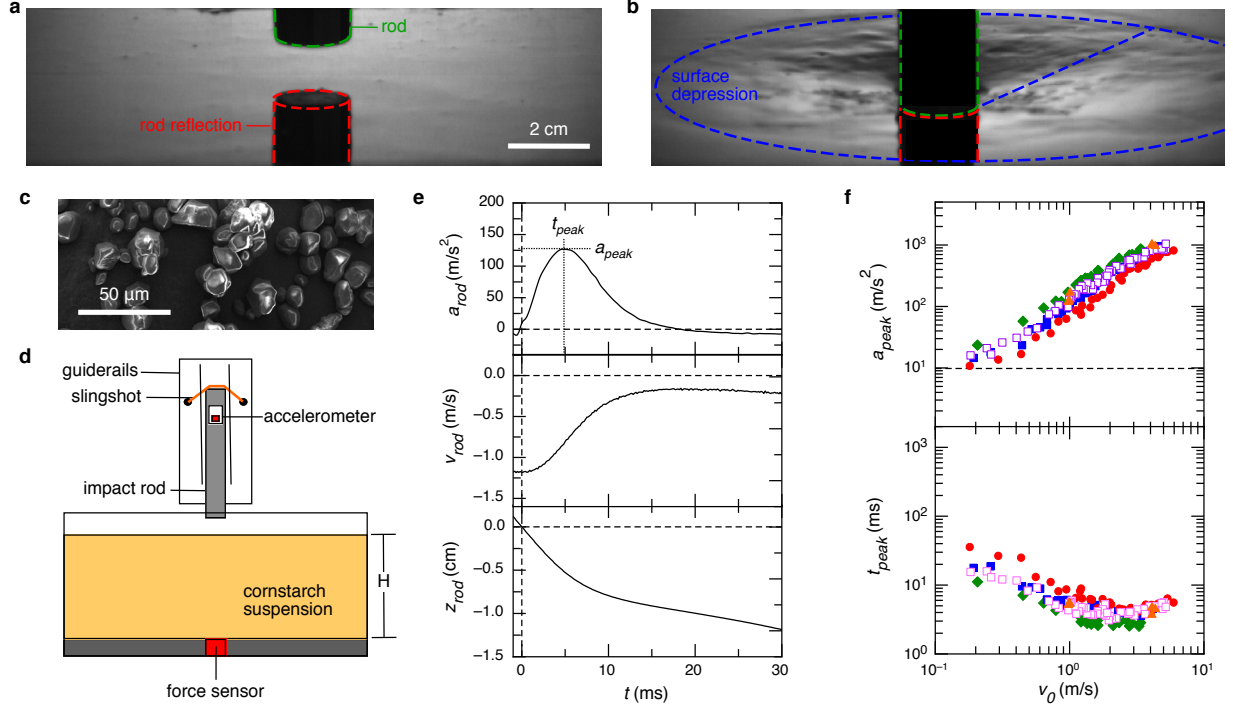


**Fig. 19.** Impact-induced solidification of a dense suspension of hard particles. A) Still from high-speed video of striker rod just before impact. B) Lateral spreading of solidification front after impact. C) Optical micrograph of cornstarch particles. D) Schematic of set-up. E) Data recorded by force sensor attached to impacting rod. F) Characteristic acceleration and time scales for different impact velocities.

**Figure 19** shows images before and after an aluminum rod strikes a dense suspension of cornstarch and water. After impact, the initially smooth and flat suspension surface develops a rapidly growing depression. This immediately sets the medium apart from water or grains alone, where impact is generally accompanied by splashing and where the impactor penetrates significantly. We plot the rod's instantaneous acceleration ( $a_{rod}$ ), velocity ( $v_{rod}$ ), and position ( $z_{rod}$ ) vs. time  $t$  in **Fig. 19e**. The acceleration is characterized by a pronounced peak  $a_{peak}$  occurring at time  $t_{peak}$ . The scale of this peak at a given impact velocity is typically  $\sim 100$  times larger than for water alone, with decelerations as high as 100 g and pressures on the rod face exceeding 1 MPa. At times much larger than  $t_{peak}$ , the impacting object, now nearly stationary, begins to slowly sink. In the following we focus on the early-time behaviour before this sinking and on the velocity regime  $0.2 < v_0 < 2$  m/s, beyond which this material fails and the rod begins to penetrate the surface. We used cornstarch/water mixtures as a model suspension because of its pronounced impact hardening and the ready availability of large quantities, but expect similar behaviour with other types of dense suspensions with particle concentration close to, but below, the value required for jamming.

The existence of peaks in  $a_{rod}$  vs.  $t$  suggests that the force exerted on the rod by the suspension is a competition between contributions that grow with time and others that diminish. One of these is related to  $v_{rod}$  (**Figs. 19f**) as increasing  $v_0$  causes  $a_{peak}$  to grow from  $\sim 10$ -100 g and  $t_{peak}$  to shrink. Raising the packing fraction  $\phi$  (the ratio of total particle volume to system volume) by even a small amount leads to a considerably stronger impact response (factor  $\sim 2$  for  $\phi = 0.46$  to 0.52), but preserves the shape of the curves shown in **Fig. 19**. While the response depends strongly on the grain packing fraction, it is surprisingly insensitive to the fluid parameters. By adding a  $\sim 1$  cm layer of water to the suspension surface and thereby effectively setting the surface tension  $\gamma$  to zero, we see that the response is not a consequence of particles dilating into the liquid-air interface, as is observed in steady-state shear experiments<sup>2</sup>. Mixing the water with glycerol and increasing its viscosity  $\mu$  by more than a factor of 10

changes neither the shape of the acceleration curves nor the height and time of the peaks, even though it strongly affects the slow, steady-state sinking behavior after impact.

To understand the role of boundaries, we changed the suspension depth  $H$  (**Fig. 20a**). For the deepest suspension,  $a_{peak}$  occurs at  $t_{peak} \sim 10$  ms, but a second, weaker peak is just visible near  $t \sim 75$  ms. Lowering  $H$  causes this peak to intensify and move to earlier times. For the smallest  $H$  a third peak emerges, resulting from a second impact of the rod after a subtle bounce. This second peak does not arise from transmitted and reflected waves, but is the signature of solidification suspected by Liu *et al.* (NYU) and von Kann *et al.* (Twente) [19]. To verify this, we simultaneously measure the force  $F_b$  transmitted to the bottom boundary with an immersed force sensor (**Fig. 20a**). While virtually no force is measured at  $t_{peak}$ ,  $F_b$  strongly correlates with peaks two and three. This has many important implications: first, the primary response is *not* the result of stress transmission to the boundary; second, the solidification process requires a finite amount of time to propagate through the suspension; third, once solidification reaches the bottom boundary, forces propagate with no detectable delay through a now jammed, solid-like region back toward the impactor; and fourth, this jammed solid can bear stress and store energy, allowing, for example, the bounce of the impactor. We can thus interpret the second peak in  $a_{rod}$  as occurring at the

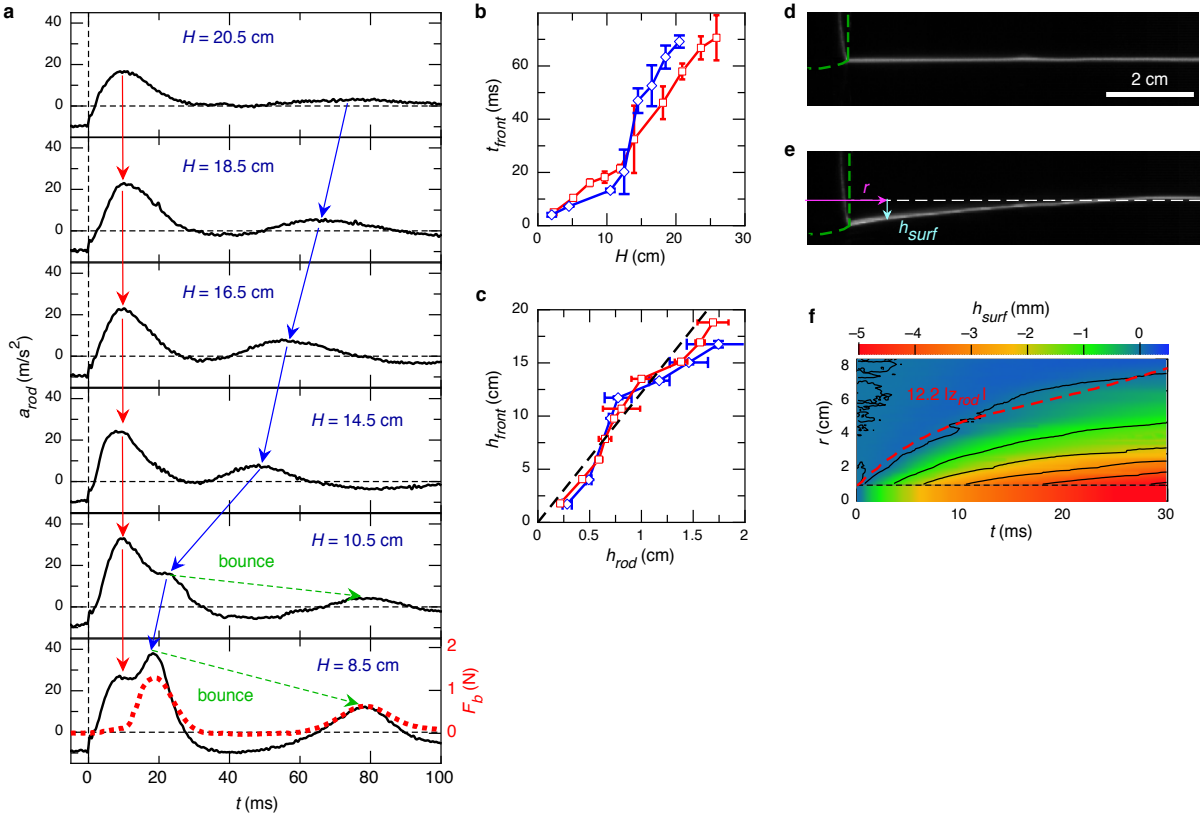


**Fig. 20.** A) Evolution of the impact response with decreasing depth  $H$  of the suspension (top to bottom). B) Front arrival time as function of  $H$ . C) Front propagation distance is proportional to vertical rod movement. D&E) Laser scan of top surface depression. F) Spatial reconstruction of surface depression, showing that lateral and vertical front growth are similar (red dotted line).

time  $t_{front}$  required for the front of a developing solid to reach bottom. **Figure 20b** shows  $t_{front}(H)$  for two different impact velocities  $v_0$ . Using these data to compute the travel distance of the front  $h_{front} = H - h_{rod}$  and plotting this against the travel distance of the rod ( $h_{rod} = |z_{rod}|$  at  $t_{front}$ ), we find nearly linear collapse, i.e.  $h_{front} = k h_{rod}$  with  $k = 12.2 \pm 0.1$ . This establishes that the solid growth is driven by the rod's motion, with the solidification front moving ahead of the rod at velocity  $v_{front} = k v_{rod}$ .

This behavior suggests similarities with granular shocks in the jammed regime or solidification fronts in supercooled glass-forming liquids. With granular shocks, however, the front propagates through an already-jammed medium and its speed is governed by elastic energy stored in grains. While supercooled liquids, like the system here, are initially unjammed, their solidification fronts propagate at a constant, thermodynamically favored speed. Here, solidification is more reminiscent of a snowplow, as illustrated in **Fig. 20d**. In this picture, hard, highly dissipative grains of diameter  $d$  have initial spacing  $\delta$ . Pushing this arrangement with speed  $v_{rod}$  creates a solidification front moving (relative to the rod) with velocity  $v_{front}=v_{rod}d/\delta$ , which after integration leads to  $h_{front}=kh_{rod}$ , with  $k=d/\delta$ . While dry cornstarch grains themselves are not perfectly inelastic, their interactions in suspension are mediated by lubrication forces. These depend strongly on the relative velocity  $v_{rel}$  of the grains ( $F=3\pi\mu d^2 v_{rel}/8\delta$  for head-on collisions), leading to significant dissipation even before contact. Despite the viscosity dependence, grains aligned as in **Fig. 20d** but surrounded by fluid produce front speeds simply set by geometry because no energy storage is present. From the experimental value for  $k$  in **Fig. 20c** this scenario suggests an interparticle gap  $\delta \sim d/12.2$  or  $\sim 1\mu\text{m}$ , a realistic value for the dense suspensions used.

To see this better, we looked inside the optically opaque suspension with x-ray videography, loading it with high-contrast tracer particles and using particle imaging velocimetry to calculate the displacement field resultant from impact. The displacement field (**Fig. 21a**) shows a large region below the rod that is pushed straight downward, the qualitative signature of the jammed solid. Just outside of this, the field begins to curl outward and upward to conserve volume globally. The overall size of the region that is moving downward provides visual evidence of where the rod's momentum has been spent.

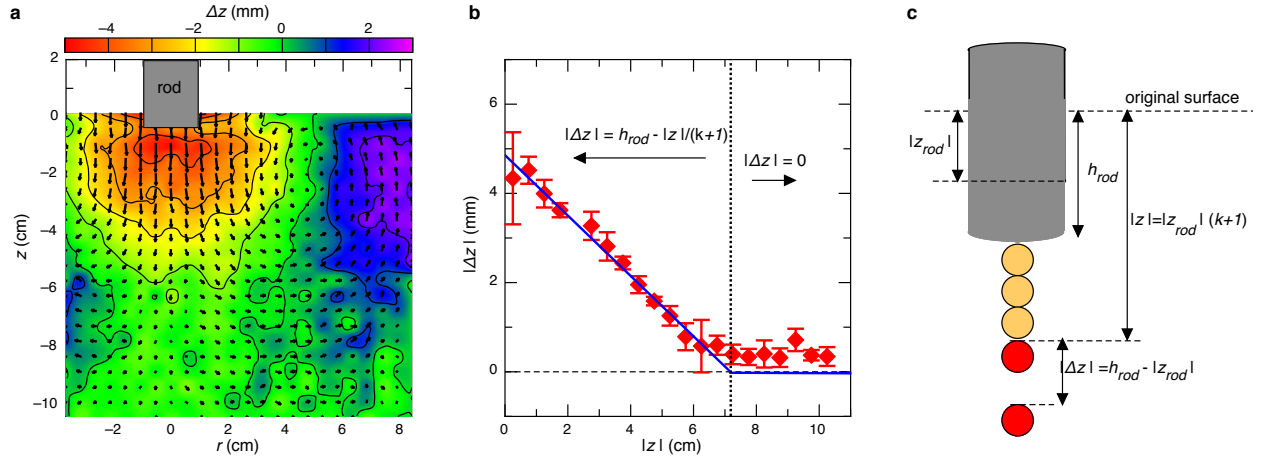


**Fig. 20.** A) Mapping of the displacement field obtained from x-ray measurements. B) Vertical displacement as a function of depth directly underneath the impactor. C) Sketch of 1D ‘snow-plow’ model with relevant distances indicated.

For the jammed region below the rod ( $z < 0$ ,  $r < r_{rod}$ ) we can use the simplified 1D model to predict the magnitude of the vertical displacements  $|\Delta z|$ . A grain at  $z$  (**Fig. 21c**) is assumed motionless until the front reaches it. Since the front and rod positions are related by  $z_{front}=k|z_{rod}|$  ( $z_{front}$  measured relative to the rod) and  $|z|=|z_{rod}|+z_{front}$ , this happens when  $|z_{rod}|=|z|/(k+1)$ . Afterwards, the grain moves with the rod. Thus, if the rod moves a total distance  $h_{rod}$  between the two x-ray images, the grain moves  $|\Delta z|=h_{rod}-|z_{rod}|=h_{rod}-|z|/(k+1)$ , while beyond  $|z|=h_{rod}/(k+1)$  we expect  $|\Delta z|=0$ . Fitting this form to the data for  $|\Delta z|$  obtained from the x-rays (**Fig. 21b**) yields  $k=13.1 \pm 0.9$ , close to the value  $k=12.2 \pm 0.1$  obtained from the acceleration curves (**Fig. 20c**).

These results indicate that the origin of the response is the dynamic growth of the jammed solid below the impact site. As this solid grows and is forced to move with the rod, it causes flow in the surrounding

fluid. The interplay between this growing region of moving suspension and the slowing of the rod is the competition mechanism responsible for the observed peak in the rod deceleration. We can capture the essence of this behaviour using the concept of added mass, as is frequently done for surface impact in regular liquids. The key idea is to treat the impact as an inelastic collision with a growing mass,  $m_a$ . The rod dynamics are then captured by force balance,

$$(m_{rod} + m_a)a_{rod} = -(dm_a/dt)v_{rod} + F_{ext} \quad \text{Eq. 1}$$

where  $F_{ext}$  in our case accounts for gravity ( $F_g = -m_{rod}g$ ) and the buoyant force from the displaced fluid in the depression (from **Fig. 22**,  $F_b \sim 1/3\pi\rho g(r_{rod} + k|z_{rod}|)^2|z_{rod}|$ ,  $F_b \approx 1/3\pi\rho g(r_0 + k|z_{rod}|)^2|z_{rod}|$ ). With regular liquids,  $m_a$  is typically limited by the density of the liquid and the size of the impactor, e.g.

$m_a < C4/3\pi\rho r_{rod}^3$ ,  $m_a \leq C4/3\pi\rho r_{rod}^3$  for disk impact. The factor  $C$  is the "added mass coefficient" and accounts for the fact that the fluid does not actually move like a solid object (consequently  $C$  is typically  $< 1$ ). The suspension is capable of responding so dramatically because the solidification below the rod leads to rapid, effectively unlimited growth of  $m_a$ . We can estimate its size from **Figs. 20&21**, which show that the impact creates substantial flow in a region that extends  $k|z_{rod}|$  below and radially away from the rod. Approximating these points as bounding a cone-like region gives  $m_a$  the form

$$m_a = (1/3)C\pi\rho(r_{rod} + k|z_{rod}|)^2k|z_{rod}|. \quad \text{Eq. 2}$$

Using this in Eq. 1 with the initial conditions  $v_{rod}(0) = v_0$  and  $z_{rod}(0) = 0$  allows us solve numerically for the rod dynamics. Using the average measured value  $k = 12.5$  and leaving the coefficient  $C$  as the only adjustable parameter, this minimal model reproduces both magnitude and timing of the impact response extremely well over the whole range of initial velocities tested (**Fig. 22**). We find good agreement for  $C \sim 0.37$ , similar to what is encountered for disk impact into regular liquids. We can also extract  $m_a$  directly from our data and confirm the scaling with  $z_{rod}$  as given by Eq. 2.

While large impact resistance has in the past been taken as prototypical example of shear-induced thickening, our findings point to a different mechanism more akin to crossing the jamming transition by compression along the packing density ( $\phi$ ) axis. Surface impact precipitates a sudden, local compression of the particle matrix, forcing it across the jamming transition and leading to a rapidly growing solid mass whose motion drives flow in the surrounding suspension. The impact-jammed solid is transient, but prior to "melting" exhibits a yield stress and elastic properties, in contrast to shear thickened states that only exist beyond yielding. Our findings provided direct evidence that this solid can transmit stress between the moving object and a boundary, and that this depends on both the size and

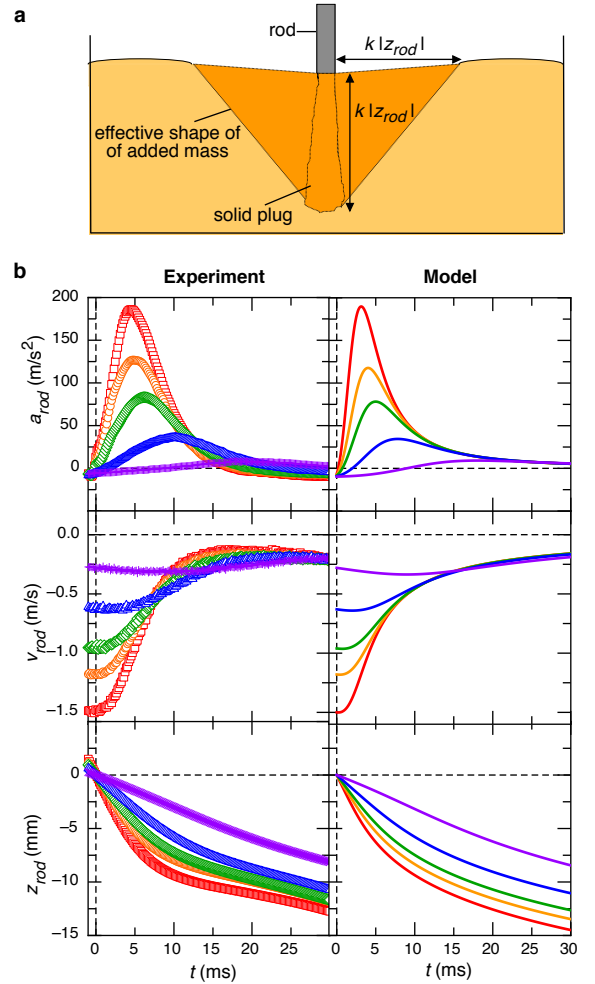


**Fig. 22.** A) Sketch of the jamming front propagation initiated by impact. Note that axial and lateral growth are occurring at the same rate (same parameter  $k$ ). B) Comparison of the measured experimental data (left) with predictions from the model encapsulated by Eqs. 1 and 2.

the speed of the object. Importantly, the critical element in creating large normal stresses during impact is not the presence of a boundary, but instead the momentum transferred as the impact-activated jamming transition triggers the formation and growth of a solid underneath the impactor.

#### 4.3.2. Propagating Jamming Fronts in 2D

In order to image the dynamic evolution of the fronts more directly we performed quasi-2D experiments using a suspension of  $\sim 20\mu\text{m}$  diameter cornstarch particles in a simple liquid (water and glycerin). In order to prevent settling of the particles during the experiments, we used CsCl to density-match the suspending liquid to the cornstarch. The amount of glycerin used to adjust the viscosity of the solution varied from 1 cSt to  $\sim 50$  cSt. The suspension was floated on a heavy hydrophobic liquid (FC-3283), which has a low viscosity to provide an approximately stress-free boundary condition at the bottom surface of the suspension. The upper surface is in contact with air, resulting in a quasi-2D system where we can neglect any variations in the vertical direction. The fluid and the suspension sat in a rectangular acrylic container, where the suspension was restricted to one side of the container by a thin silicone rubber sheet. We impacted the suspension from the side using a linear actuator at a constant speed, controlled between 0.05 to 0.45 m/s. Using tracer particles and high speed imaging (Phantom V12 at 5000 frames/s) we performed time-resolved Particle Image Velocimetry (PIV) to visualize the growth of the jamming front and measure its properties.

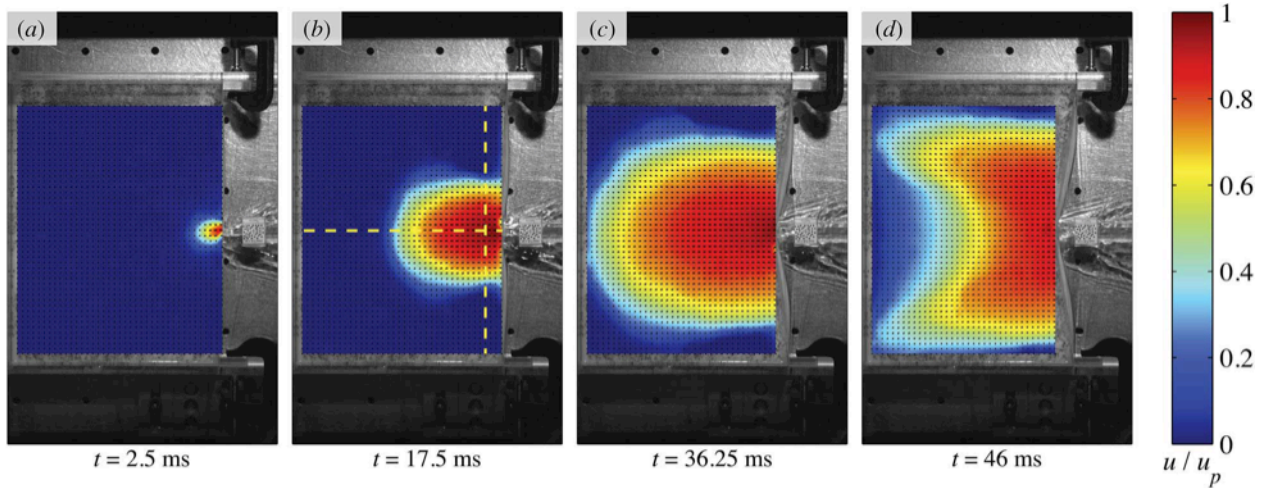


**Fig. 23** – Evolution of dynamic jamming front in a quasi-2D system, where a layer of suspension floating on a heavy liquid is horizontally impacted from the right. The images show snapshots from a high-speed video with PIV data overlaid. The colors indicate the local velocity normalized by the impactor velocity. At time 36.25ms the front has reached the system boundary and the jammed region, which was moving effectively as a plug, starts to compress, resulting in a front that appears to move backward.

**Figure 23** provides three snapshots from the front evolution. In these images, the PIV analysis is overlaid on stills taken from the high-speed video. We define the front position as the points at which the velocity has decayed to 50% of the impactor speed. From the analysis we found that the front propagates at a constant speed in both axial and transverse directions (**Fig. 24**), resulting in a self-similar shape (**Fig. 25**).

The constant axial and transverse speeds result in a speed ratio around 2 that is independent of how much faster the front propagates than the impactor. The latter is given by the ratio  $k$  and is a measure of how close the system is to the jamming point ( $k$  diverges at jamming).

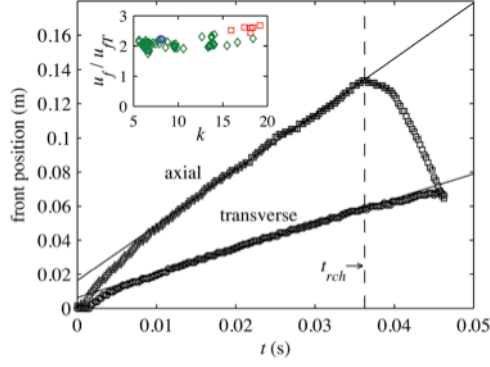


**Fig. 24.** Front propagation in both axis (longitudinal) and transverse directions. There is an initial transient, followed by a constant propagation velocity. At time  $t_{rch}$  the boundary is reached. Inset: Axial to transverse speed ratio as a function of distance to jamming point, parameterized by the ratio of axial front speed to impactor speed,  $k$ .

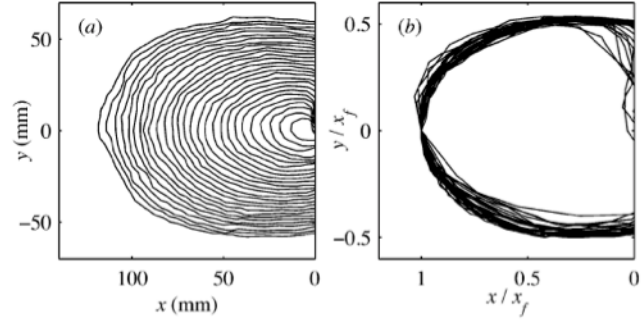


**Fig. 25.** (a) 2D front shape at successive time intervals. (b) Front shape normalized by  $x$ -coordinate of front at given times. Data collapse indicates a self-similar front profile.

One key result is that we can identify three regions from the axial velocity profile, going from (i) unjammed to (ii) jammed, and (iii) further compression of a jammed packing. A second key result is (**Fig. 26**) that the force response on the impactor can be *quantitatively* accounted for by the added mass, as we originally proposed in our Nature paper from 2012 (see Section 4.3.1). This holds until the jamming front reaches the system boundaries (**Fig. 23c**). The vertical dotted lines in **Figs. 24** and **26** shows that this speed is reduced once there is interaction with the boundaries at  $t_{rch}$ . Third, we observed that after the moment of first interaction with the boundaries, there is a delay in the response of the force sensor. This delay corresponds to the time it takes for the jammed region to become homogeneous across the full system, at  $t_{span}$ , beyond which point force can be transmitted directly between impactor and boundary, and thus the measured force shoots up dramatically.

*Results from this work were published in Soft Matter: I. R. Peters and H. M. Jaeger, Quasi-2D dynamic jamming in cornstarch suspensions: visualization and force measurements, Soft Matter* **10**, 6564-6570 (2014). doi: 10.1039/C4SM00864B.

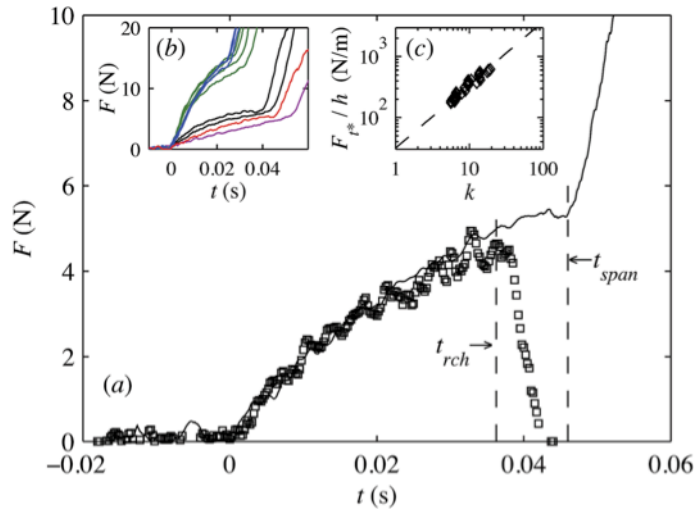


**Fig. 26.** (a) Direct comparison, without adjustable parameters, of the force calculated from momentum conservation (squares) and the force measured with the force sensor (solid line). Only after interaction with the boundary do the values deviate. (b) Representative set of measured forces as a function of time for different experimental conditions ( $h$  = thickness of suspension layer;  $k$  = ratio of front speed to impactor speed;  $v$  = solvent viscosity). Black:  $h = 16.5$  mm,  $k = 8.0$ ,  $v = 6.9$  cSt; green:  $h = 30.0$  mm,  $k = 12.1$ ,  $v = 7.2$  cSt; purple:  $h = 13.0$  mm,  $k = 6.0$ ,  $v = 43$  cSt; blue:  $h = 22.0$  mm,  $k = 17.9$ ,  $v = 7.8$  cSt; red:  $h = 15.0$  mm,  $k = 7.5$ ,  $v = 6.9$  cSt. (c) Force per unit suspension depth at time  $t^* = 0.02$ s as a function of  $k$ . The dashed line is a fit with slope 1.0.

### 4.3.3 Direct observation of impact activated fronts with fast ultrasound imaging

3D granular systems, including dense suspensions, are optically opaque and our x-ray system (Section 4.3.1.) only achieves imaging speeds up to 30fps. In order to track the front with more precision and also to probe the properties of the emerging jammed solid, we set up a fast, 3D ultrasound imaging system, capable of imaging up to 10,000 frames per second. This system, funded through a DURIP grant (ARO Proposal No. 63464-MS-RIP), is based on a Verasonics ultrasound transceiver (Verasonics Vantage 128 Research Ultrasound Platform) and the hardware was specifically geared at imaging dense suspensions.

With this system, we achieved the first direct tracking of the evolution of an impact activated jamming front inside a concentrated, 3D suspension. We confirmed both qualitative and quantitative properties of the jamming front observed in the quasi-2D experiment described in Section 4.3.2. Based on the observations we propose a new model that highlights the importance of shear in the formation and propagation of the jamming front. It is based on the quasi-static shear jamming scenario introduced by Bi et al. [20], but slightly modified to allow for some compaction of the granular subphase at a high impact velocity. The ratio  $k$  between the longitudinal and transverse front speeds, which was previously discovered to be constant with a value  $\sim 2$ , can be explained successfully with this new model. The model can also explain two other key experimental observations: (1) that the critical velocity to jam the system varies with the geometry of the system, while the critical shear rate stays constant, and (2) that there is a lower boundary in packing density  $\phi$  with no jamming below this boundary.

*Results from this effort, described below, have been submitted for publication* (Endao Han, Ivo R. Peters and Heinrich M. Jaeger, “Direct observation of impact activated fronts in dense suspensions” (2015)).

As schematically illustrated in **Fig. 27**, the experimental setup consisted of a cylindrical container filled with suspension, a vertical impactor, and the ultrasound system. The container was made of a UV-cured resin (“Vero White Plus”, Objet Geometries Inc.) whose acoustic impedance matched the suspensions we studied. The inner diameter of the container was 10.0 cm and the typical fill depth  $H$  of the suspension was 2.5 - 3 cm. The impactor was a cylinder connected to a computer-controlled linear actuator (SCN5, Dyadic Systems). A sensor (DLC101, Omega) measured the force on the impactor applied by the suspension. The ultrasound transducer (Philips L7-4) contacted the bottom of the container through a thin layer of ultrasound gel. It has 128 independent elements with a total width of 3.8 cm and scans a vertical thin sheet indicated by the shaded area. For the data described below we define a cylindrical coordinate system whose  $z$ -axis aligns with the central line of the impactor.

The suspension was a mixture of cornstarch particles and CsCl-glycerol-water solution. Both the particles and the liquid have a density of  $1.63 \pm 0.01 \text{ g/cm}^3$  and the viscosity of the liquid is adjusted by the mass ratio between glycerol and water. The mixed suspension is degassed before experiments to remove any air bubbles that would scatter the ultrasound signals. Once the packing fraction dependent speed of sound  $c$  was measured after degassing, a certain amount of air bubbles were put back into the suspension to act as tracer particles for PIV analysis of the ultrasound images.

In each impact experiment, the impactor moved down vertically with a velocity  $v_p$  and stopped at a distance  $d_g$  above the bottom of the container. The ultrasound system was triggered as the impactor approached the top suspension surface, imaging at 100 to 10,000 frames per second (adjusted according to  $v_p$ ). During each acquisition (one frame) the transducer sends a sinusoidal wave at 5MHz frequency

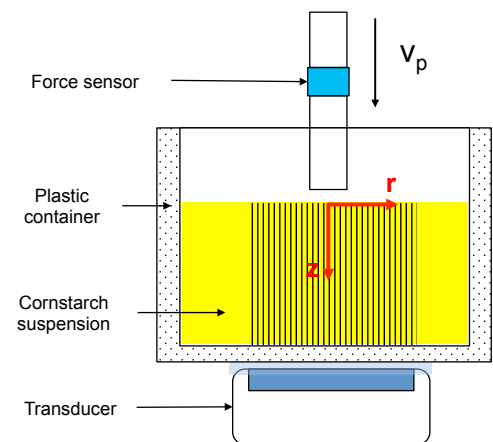


**Fig. 27.** Schematic of the setup. The shaded yellow area outlines the field of view of the ultrasound transducer (see Fig. 28).

modulated by a Gaussian profile and each of its 128 elements receives a reflected time series. With the previously measured speed of sound  $c$  we can then reconstruct B-mode images from the time series. By analyzing the B-mode images with a particle imaging velocimetry (PIV) algorithm, we thus obtain two-dimensional flow field slices from inside the bulk of a 3D suspension (**Fig. 28a**).

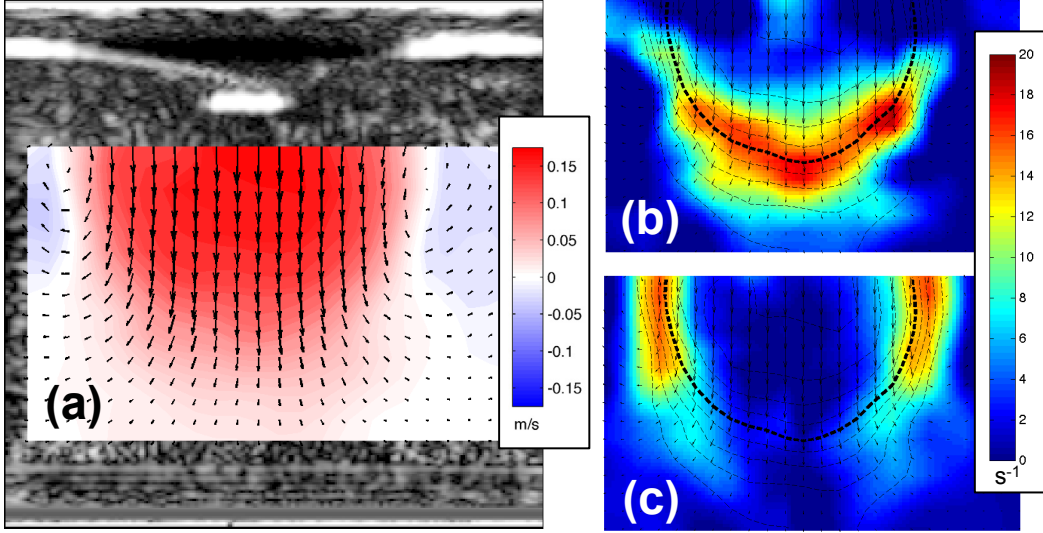


**Figure 28.** Visualization of the front. (a) Velocity field of a propagating front. Color represents the magnitude and direction of the vertical component of the local velocities: red means moving down and blue up. Initial suspension depth  $H = 30$  mm, impactor diameter  $D = 6.0$  mm, and  $v_p = 175$  mm/s. (b) and (c) plot two components of the shear rate tensor at the same moment in time as the velocity field shown in (a). (b) shows  $-\dot{\epsilon}_{zz}$  and (c) shows  $\dot{\epsilon}_{rz}$ . The thin dashed curves represent velocity contour lines; the thick dashed line indicates the front position, defined by  $v_z = v_p/2$ .

A typical flow field in the suspension before the front reaches the bottom is shown in **Fig. 28a**. The vertical and horizontal axes correspond to the vertical ( $z$ ) and radial ( $r$ ) directions in cylindrical coordinates, respectively. The flow field clearly shows a jammed region in the middle, where all points move vertically with uniform  $v_p$  (red). Underneath the jammed region the suspension is forced to flow to the side, and around it the suspension is dragged down. Different from the quasi-2D experiment in Section 4.3.2, vortices are observed on the side of the propagating front. This is due to volume conservation.

We calculated the local shear rate from two-dimensional velocity fields ( $u_r, u_z$ ) as shown in **Fig. (a)**. Assuming that the system is rotational symmetric so that  $u_\theta$  and its derivatives are all zero, the shear rate tensor is given by

$$\dot{\epsilon} = \begin{pmatrix} \frac{\partial u_r}{\partial r} & 0 & \frac{1}{2} \left( \frac{\partial u_r}{\partial r} + \frac{\partial u_r}{\partial r} \right) \\ 0 & \frac{u_r}{r} & 0 \\ \frac{1}{2} \left( \frac{\partial u_r}{\partial r} + \frac{\partial u_r}{\partial r} \right) & 0 & \frac{\partial u_z}{\partial z} \end{pmatrix}.$$

**Figures 28b&c** show  $-\dot{\epsilon}_{zz} = -\frac{\partial u_z}{\partial z}$  and  $-\dot{\epsilon}_{rz} = \frac{1}{2} \left( \frac{\partial u_r}{\partial r} + \frac{\partial u_r}{\partial r} \right)$ , respectively. The data show that  $\dot{\epsilon}_{zz}$  dominates just below the front, which corresponds to compression on the  $z$  direction and to a radial expansion, ie., in this region we have pure shear. However, at the edge of the front in the transverse direction,  $-\dot{\epsilon}_{rz}$  dominates, and most contributions arise from the term  $\frac{\partial u_z}{\partial r}$ . Here the velocity changes along the direction perpendicular to the impact direction, which is analogous to simple shear. Though the

shear types are different, the regions of high shear rate always coincide with the edge of the front, while inside and away from the jamming front the velocity changes are very gradual. Thus the system can be divided into three parts: a solid core moving as a plug with a velocity close to  $v_p$ , the shear zone at the edge of the moving front, and the un-perturbed fluid far away.

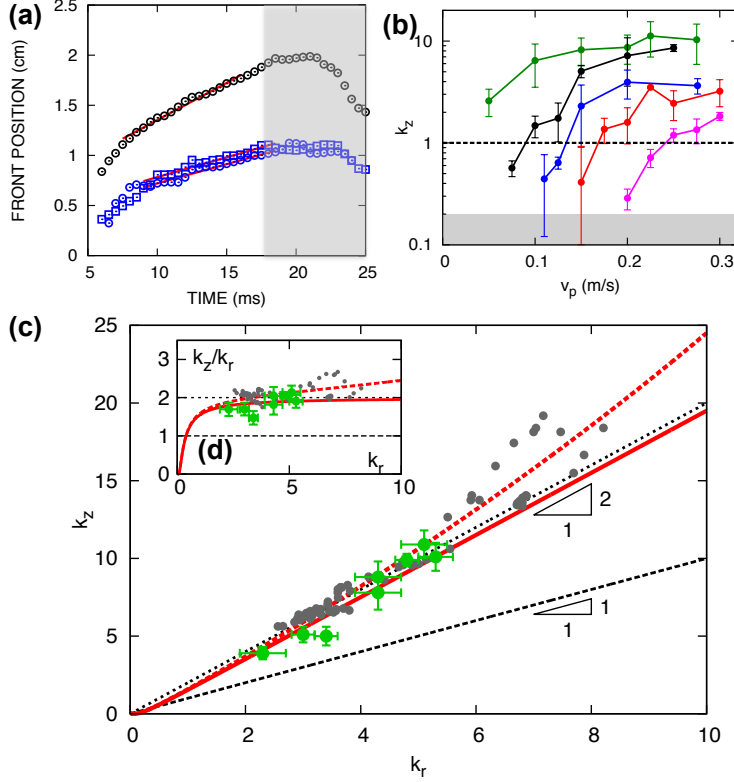


**Figure 29.** Propagation of the jamming front. (a) Positions of the front on the longitudinal (black) and transverse (blue) directions as functions of time. The blue circles and squares represent the right and left boundaries respectively. The impactor reaches the surface of the suspension (0 cm) at time  $t = 0$  ms. The left boundary of the gray region labels the time when the longitudinal front reaches the bottom of the container. (b)  $k_z$  as functions of  $v_p$  at the same liquid viscosity, but different  $\phi$ , which increases upwards from pink to green. The gray region labels the level of noise. (c) Comparison of  $k_z$  and  $k_r$ . The red solid line is the prediction with the shear jamming model and the dashed red line shows the prediction of the modified version of the model.

We define the front position as the line of points in the  $z$ - $r$  plane where the vertical component of the velocity is  $v_p/2$ . As illustrated in **Fig. 29a**, after an initial transient stage, the fronts propagate linearly as function of time in both axial and transverse directions. The linear growth slows down when the front reaches a boundary and subsequent compression causes a reversed propagation of contours as described in the quasi-2D experiments in Section 4.3.2. Here we focus on the spatial regime with the linear propagation, where the front velocities  $v_{fr}$  and  $v_{fz}$  are simply the slopes of the data traces. To compare how fast a front propagates compared to the impactor's (pushing) velocity  $v_p$ , we define two dimensionless front propagation factors  $k$  as

$$k_r = \frac{v_{fr}}{v_p}, \quad k_z = \frac{v_{fz}}{v_p} - 1.$$

For an ordinary, simple liquid, both  $k_r$  and  $k_z$  are effectively zero in the steady state. In a suspension, the relation between  $k$  and  $v_p$  depends on the suspending liquid's viscosity and on the packing fraction of the solid subphase. In the liquid-like unjammed state,  $k$  is zero or very small while  $v_p$  is below a critical value  $v^*$ . Then there is a sharp transition after which  $k$  increases with  $v_p$  until it reaches a plateau at high  $v_p$ . A higher viscosity decreases  $v^*$ , but it does not change the asymptotic value as  $v_p$  approaches  $\infty$ . When the packing fraction was adjusted while keeping the suspending liquid viscosity constant, as illustrated in **Fig. 29b**, the onset where  $k_z > 0$  shifts to higher  $v_p$  as  $\phi$  decreases. However, different from the previous case the plateau value decreases with  $\phi$ . Lastly, we compare  $k_z$  with  $k_r$ . As plotted in **Fig. 29c** we find a constant ratio  $k_z/k_r = 2$  when  $k$  is sufficiently large. This agrees with the quasi-2D dimensional experiments [10] in

the high  $v_p$  regime, and it therefore emerges as a robust result regardless of the coordinate system (Cartesian in [10] and cylindrical here).

Based on the observations above, we propose a new mechanism for impact-activated solidification in dense suspensions. We argue that frictional interactions between particles and a structural change due to shear play a much more significant role than the change of local packing fraction in the solidification process. The solidification happens only when the stress and the strain in the suspension both exceed critical values. In the low  $v_p$  regime the stress is small, thus the suspension is in a lubrication dominated state and behaves like a Newtonian fluid. Therefore, with increasing  $v_p$  the stress will increase until it reaches the critical stress for the onset of shear jamming in a system of frictional particles [20]. Since the effective viscosity of the suspension increases with packing fraction  $\phi$  or liquid viscosity  $\eta$ , by increasing either of these system parameters the threshold value for shear jamming can be achieved at a lower  $v_p$ . This explains the shift of the critical velocity in **Fig. 29 b&c**. In the high  $v_p$  regime, when the stress in the shear zone is much larger than the critical onset value, front propagation speed requires a minimum strain  $\epsilon_c$ . The physical meaning of  $\epsilon_c$  is the strain needed for the particles to move out of the initial uniform distribution and form connected force chains. In this regime the propagation factor  $k$  is simply  $k = q/\epsilon_c$ , where  $q = 1$  for pure shear and  $q = 1/2$  for simple shear. Here  $k_z$  has the same expression as in the ‘snow plough’ model [1], though  $k_z \neq \phi/(\phi_J - \phi)$  in this case. The model easily explains the ratio  $k_z/k_r = 2$  by assuming that  $\epsilon_c$  is the same in  $z$  and  $r$  directions. As the data in **Fig. 29c** implies,  $\epsilon_c$  decreases as  $\phi$  increases.

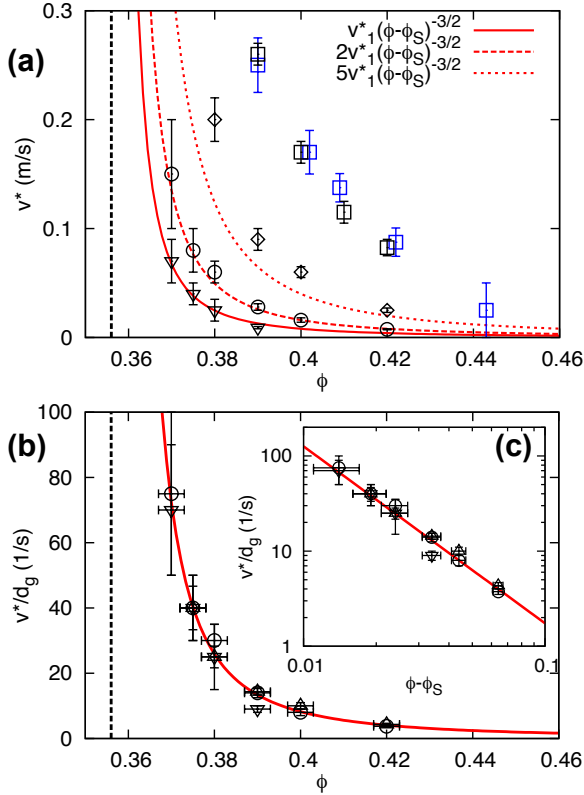


**Figure 30.** (a) The critical velocities  $v^*$  as function of packing fraction  $\phi$ . The blue squares are obtained from the ultrasound data and the black data are from the force measurements. Different symbols represent different  $d_g$ . Squares:  $d_g = H/2$  ( $k_z=1$  when the front just reaches the bottom); diamonds:  $d_g = 5$  mm; circles:  $d_g = 2$  mm; down triangles:  $d_g = 1$  mm. (b)  $v^*$  rescaled with  $d_g$ , which is the characteristic shear rate as a function of  $\phi$  for  $d_g = 1$  mm (down triangles), 2 mm (circles) and 3 mm (up triangles). The dashed line labels  $\phi$ . (c) The same data in (b) plotted in a log-log scale. In (b) and (c) the solid red curve is a power law fitting with  $\dot{\epsilon}_c(\phi - \phi_S)^\alpha$ . In (a)  $v_1^* = \dot{\epsilon}_c \cdot 1$  mm.

The model makes two interesting predictions that were tested directly in our experiments. Firstly, when the front approaches a boundary, the shear rate of the fluid in the gap will increase dramatically because the characteristic length scale that determines the shear rate eventually crosses over from the front width to the (smaller) gap size  $d_g$ . Thus we expect the suspension to jam at a lower pushing velocity when  $d_g$  becomes small. Secondly, according to the quasi-static shear jamming for dry granular systems [20] there

exists a lower boundary in packing fraction  $\phi_s$  below which shear jamming ceases. We verified the existence of this lower boundary by measuring the force response of the suspension at low packing fractions.

As  $v_p$  increases past a critical velocity  $v^*$  the force increases before the impactor reaches  $d_g$ . Compared with the quasi-static dry granular shear jamming [20], this corresponds to the boundary between the unjammed phase and the fragile phase. In the fragile phase force chains percolate along the longitudinal direction in pure shear, which causes a measurable force response. The critical velocities as functions of  $\phi$  are plotted in **Fig. 30a**. Firstly, we set  $d_g = H/2$ , thus  $v^*$  represents the  $v_p$  at which  $k_z = 1$  if we follow the previous definition and relate a non-zero force response with the front reaching the bottom. In **Fig. 30a**, we compared these  $v^*$  with the  $v_p$  at which  $k_z = 1$  as obtained from the ultrasound visualization method shown in **Fig. 28c**, and they match well at different  $\phi$ . This corresponds to the limit  $d_g$  approaches  $\infty$  where the characteristic length scale is the front width. Then we changed  $d_g$  from 1 mm to 5 mm and plotted  $v^*$  in the same figure. It shows that  $v^*$  decreases with  $d_g$ , which implies a change of length scale. To show that  $d_g$  is the dominate length scale when it is small, we collapse the data for small  $d_g$  by calculating the characteristic shear rate  $\dot{\gamma}^*/d_g$  in the system as shown in **Fig. 30b**. Comparing this curve multiplied by  $d_g$  with the experimental data in **Fig. 30a**, we find that the rescaling fails at  $d_g = 5$  mm or above. From **Fig. 30b** we also observe a dramatic increase in  $v^*$  as  $\phi$  decreases. It was fit by a power law as shown in **Fig. 30c**. This shows that a lower limit in  $\phi$  does exist and below this limit the suspension behaves like an unjammed fluid no matter how fast it is impacted or sheared. In our case this limit is reached at  $\phi_s = 0.36$ .

#### 4.3.4 Jamming front propagation under pulling in dense particulate suspensions

It requires a lot of force to quickly pull out an object immersed in a bath of dense granular suspension like cornstarch mixed in water. To understand such striking *tensile* force response, we experimentally measured the normal force required for pulling out a rod vertically from the suspension at a controlled pulling velocity. We observed that for slow pulling velocities the force response is similar to that of highly viscous fluids, but that above a threshold velocity the force shows a diverging behavior soon after an initial viscous-like response. We used non-invasive methods (X-ray and fast ultrasound imaging) to map out the local velocity profiles inside the optically opaque suspension during pulling. These measurements clearly showed that under rapid pulling, there is a growing jammed region inside the suspension. This region interacts with the rigid boundaries of the container, giving rise to the observed force divergence. Our findings suggest that under rapid pulling the force response in dense granular suspensions is very similar to impact activated solidification as discussed in the preceding sections, even though the concept of jamming under pulling is highly non-intuitive.

The notion of a rapidly growing jammed region is qualitatively similar to what we observed for impact activated solidification. Jamming and fracture in dense suspension of Brownian particles under extensional shear flow has been studied before [21, 22], but the dynamical route to jamming with the emergence of a dynamic jamming front under extensional flow is still unexplored both for dense Brownian as well as non-Brownian suspensions. This was the main motivation of this effort.

Cornstarch suspensions were prepared by adding cornstarch powder (Agro) at volume fractions ranging from  $\rho = 0.44$  to 0.48 slowly in a density matched brine solution of water cesium chloride (CsCl) and glycerol mixture. By varying the glycerol content, we could tune the solvent viscosity. The solvent viscosity used ranged from 10 mPas to 50 mPas. Higher solvent viscosity delays the melting of the jammed region and thus can be probed at slower velocities. To study the force response under pulling, we used a MCR 301 rheometer (Anton Paar, Germany) in the normal force measurement mode. Unless otherwise specified, we used a circular plate of diameter 25mm connected to the force transducer through a cylindrical rod as shown in **Fig. 31a**. With the transducer, the force on the plate could be precisely

determined down to values of  $\sim 0.01$  N, while the built-in optical position sensor measured the vertical velocity of the plate. By a feedback mechanism, the pulling velocity could be set over a wide range ( $1 \mu\text{m/s}$  -  $8 \text{ mm/s}$ ). For a typical measurement the plate was first placed on the surface of the suspension, which was kept in a container of much larger diameter than the plate, and the normal force ( $F_N$ ) was measured on the plate as it was pulled upward at constant velocity  $v$  (**Fig. 31b**). The variation of  $F_N$  as a function of time is shown for two different pulling velocities,  $1 \text{ mm/s}$  and  $8 \text{ mm/s}$ , in **Fig. 31c**. We see a strong force divergence close to time  $t = 2 \text{ s}$  for a pulling velocity of  $8 \text{ mm/s}$  whereas for  $1 \text{ mm/s}$ , the force on the plate remains close to zero (on a log scale the peak force for  $1 \text{ mm/s}$  pulling velocity remained almost two orders of magnitude lower compared to that for  $8 \text{ mm/s}$  pulling velocity). We also imaged the suspension under pulling with a high-speed video camera (Phantom V12, Vision Research).

The typical images of the suspension is shown in the inset of the **Fig. 31c** at different points in the force response curve for both  $8 \text{ mm/s}$  and  $1 \text{ mm/s}$  pulling velocities as indicated in the figure. We have observed that for higher pulling velocities ( $> 4 \text{ mm/s}$ ), the contact line of the suspension with the plate remains pinned before it gets detached after the force divergence and there is a formation of truncated conical (frustum) shaped jammed region (indicated in **Fig. 31b**, by the region enclosed by black dashed-line) that grows as a function of time and maintains its shape under gravity. When this growing frustum shaped region interact with the nearest rigid walls (in this case it is the side walls) of the container force shoots up by an order of magnitude before breaking away from the plate. This is a clear signature of solid-like jammed behavior, because no pure liquid can maintain this frustum-like shape under gravity. For lower pulling velocities ( $3 < \text{mm/s}$ ) the system behaves more like a highly viscous liquid and the growing, frustum-like shape is not observed. Instead, under pulling the contact line retracts towards the center of the plate and the suspension flows down into the container before breaking away from the plate.

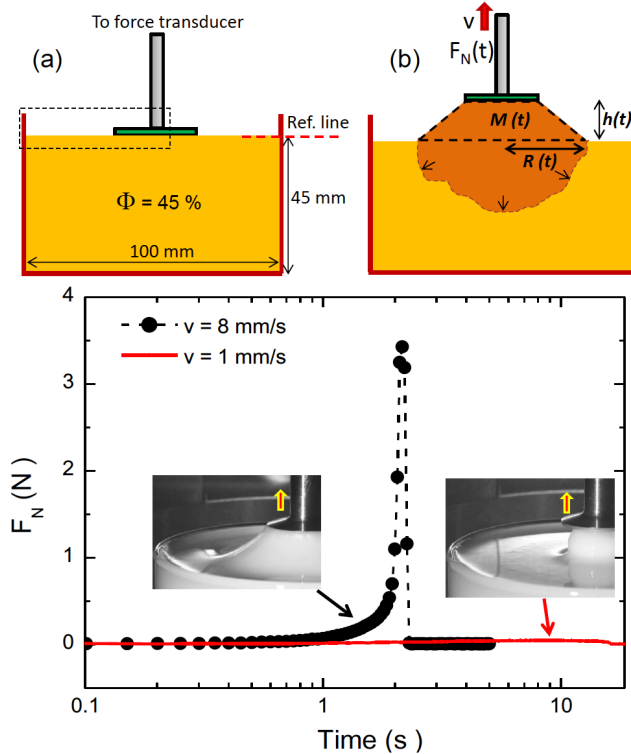
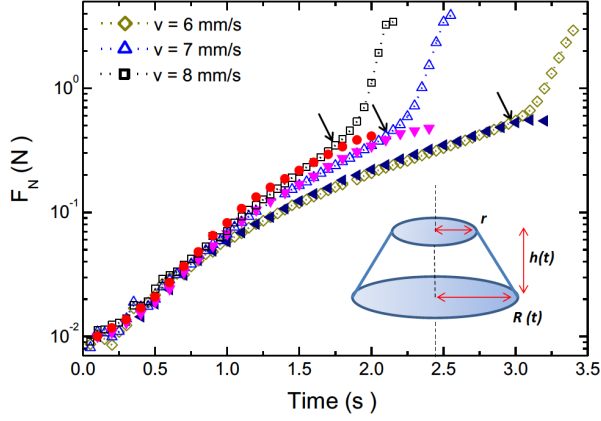


**Figure 31.** (a) Schematic of the experimental set-up to study the jamming in dense cornstarch suspension under pulling. The plate is coupled to a force transducer (rheometer head). (b) Formation of growing jamming front (brown colored region) under pulling the plate at a constant velocity. (c) Force as a function of time for two pulling velocities shown in the key. The images of the suspension are shown along at particular points in the force response curves for two velocities indicated in the figure. Note how the system behaves liquid-like at small pulling speed (red) and solid-like, seen directly in the sloped cone angle, at larger speed (black).

To understand this striking force response under higher pulling velocities, we note that the jammed suspension forms a temporally growing truncated frustum that is pulled out under gravity. We want to calculate the instantaneous gravitational pull on this jammed region having mass  $m(t)$  and moving upward



with a constant velocity  $v$ . The instantaneous normal force on the rod is given from Newton's law:

$$F_N(t) = d[m(t)v]/dt = v d[m(t)]/dt + m(t)g.$$

The instantaneous mass  $m(t) = \rho V(t)$ , where  $V(t)$  is the volume of the jammed region pulled out of the bulk and  $\rho$  is the density of the suspension.  $V(t)$  can be estimated from geometry (inset to **Fig. 32**), where  $r$  and  $R(t)$  are the respective top and bottom radii of the frustum and  $h(t)$  is the instantaneous height of the plate from the initial surface position of the suspension. It is observed that for the range of pulling velocity we used, the term  $m(t)g \gg v d[m(t)]/dt$ . Thus,  $F_N(t) \sim m(t)g$ .

We compared this estimated force response with direct measurements by the force transducer of the rheometer for three pulling velocities. This is plotted in **Fig. 32**. We find an excellent agreement between the calculated and directly measured values for early times, but the comparison breaks down completely close to the force divergence regime, where the calculated values of  $F_N(t)$  fail to capture the divergence. It is remarkable that, despite complex hydrodynamic and frictional interactions responsible for the jamming front, once the front is formed the initial force evolution is entirely dominated by the growing volume of the jammed region outside the suspension.

As mentioned earlier, our impact experiments in 3-D and quasi 2-D systems have shown that the force on the transducer diverges when the jamming front interacts with the container walls (see for example **Fig. 26**). In order to explain the rather similar divergence seen in **Figs. 31c** and **32**, we conjecture the jamming front again grows in all directions and that the force divergence takes place when the jamming front hits the wall.

**Figure 32.** Force response as a function of time (open symbols) for different pulling velocities indicated in the figure. The arrows indicate the onset of force divergence. Solid symbols indicate the comparison with the force due to the instantaneous weight of the jammed region (indicated by the dashed line in Fig.31b) pulled out from the bulk suspension estimated from the frustum shape shown in the inset.

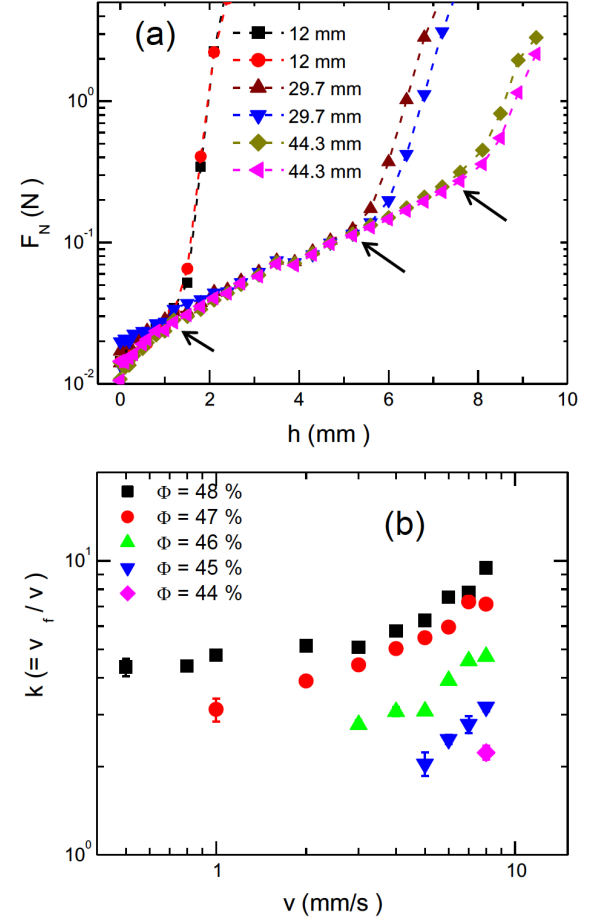


**Fig. 33.** (a) Force response curves as a function of plate displacement measured with respect to its initial position for different initial depths of the suspension as indicated. For each depth, two consecutive measurements were recorded. The arrows indicate the force divergence onset. (b) The ratio of jamming front and plate velocities ( $k$ ) as a function of pulling velocity for different volume fractions indicated in the key. The initial depth of the suspension was 27.5 mm.

To test these assumptions, we changed the initial depth of the suspension from 12 mm to 44.3 mm, to see if the position of the force divergence gets shifted. In **Fig. 33a** we plot the force response under pulling with a velocity 8 mm/s for different initial depths of the suspension as a function of the displacement ( $h$ ) of the pulling plate. The force divergence points indeed shifts to higher values of  $h$  as we increase the initial depth  $d$  of the suspension. We can again define a dimensionless front propagation speed via  $k = v_f/v$ , the ratio of average jamming front speed to plate pulling velocity. This can be expressed as  $k = 1 + d/h_c$  where  $h_c$  is the critical plate displacement measured from the initial position of the free surface of the suspension. **Fig. 33b** shows the dependence of  $k$  as a function of pulling velocity for different volume fractions  $\phi$ . We see that the values of  $k$  increase both for increasing  $\phi$  as well as with increasing pulling velocity, a feature also observed for jamming fronts under impact. Furthermore, the values of  $k$  at a particular velocity observed in the impact experiments were found to be similar to those obtained in the pulling experiments.

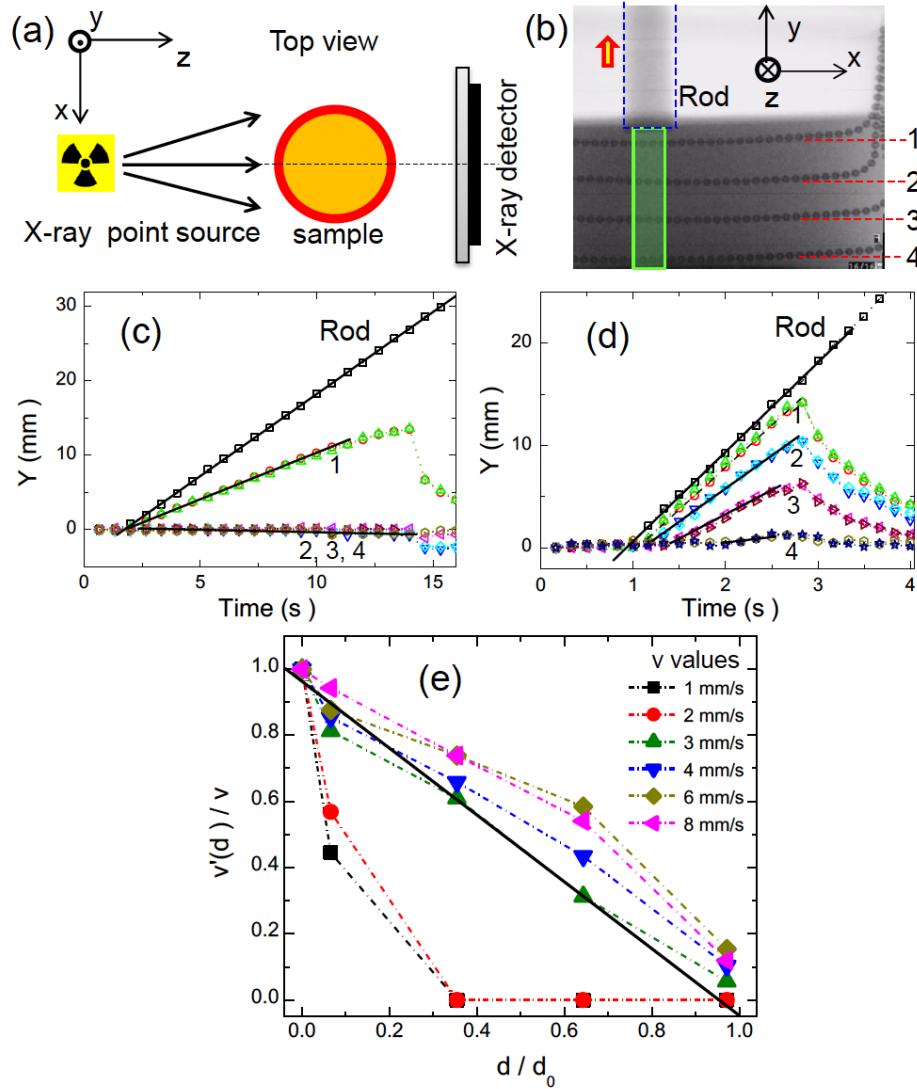


**Figure 34.** (a) Schematics of set-up to study in-situ x-ray imaging. (b) Typical image of the suspension containing the hanging pull chains made of brass and contained in the same vertical plane as the rod. The layers of the chains are numbered. The shaded region contains the bids of the chain used for tracking. Vertical displacement of the pulling rods and bids under pulling for (c) 2 mm/s and (d) 8 mm/s, solid lines indicate the linear fits to the data. (e) Normalized velocity profiles as a function of pulling velocities indicated in the keys.

We already pointed out that due to the opaque nature of the cornstarch suspensions it is not possible to observe flow profiles inside the suspension by optical methods. In this project we used in-situ X-ray imaging (**Fig. 34a**). We used metallic pull chains of brass beads (size = 1.2 mm) as tracer particles. The reason to use pull chains instead of isolated beads is that the particles needed to be confined in the same vertical plane perpendicular to the direction of x-ray propagation (imaging frame rate 30 fps).

A snapshot of an X-ray image is shown in **Fig. 34b**. Here, the tip of the pulling rod made from aluminum was placed on the surface of the cornstarch suspension. The layers of pull chains are clearly visible and different layers are sequentially marked by numbers. **Figures 34c&d** show the positions of two beads close to the long axis of the pulling rod in each layer as a function of time for 2 mm/s and 8 mm/s pulling velocities, respectively. For 2 mm/s no force divergence is observed, and only the first layer gets deformed while the other layers remain static. This shows that even for large vertical displacements of the free surface by the pulling rod, the velocity field inside the suspension is confined to the surface. By contrast, for 8 mm/s, all the chains are deformed under pulling, indicating that the velocity field extends all the way down to the bottom of the container, resulting in a force divergence.

We mapped out the axial velocity profile (normalized by the velocity  $v$  of the pulling rod) inside the suspension for different pulling velocities. This is plotted in **Fig. 34e** as a function of depth (normalized by the initial depth  $d_0$  of the suspension). We see a very clear transition in the overall shape of the velocity profile above a 2 mm/s, exactly at the velocity above which the force divergence takes place. The linear velocity profile observed at higher pulling velocities is similar to that of a homogeneous solid under stretching at uniform rate, indicating that the suspension forms a solid like jammed region extending from the pulling rod to the bottom wall of the container.

All of this demonstrated that under rapid pulling a granular suspension can undergo a jamming transition very similar to that of impact activated solidification, in both cases with a propagating solid-like jamming front that can give rise to force divergence by interacting with the rigid walls of the container.

To understand this solidification under tensile stress, we again invoke the concept of shear-jamming in dense granular suspensions. Dense assemblies of frictional granular particles can form dynamically jammed configurations under shear at packing fractions well below the random close packing density and without requiring the packing fraction to change [20]. More recent high-speed ultrasound measurements we performed in complete analogy with the impact experiments (**Fig. 28**) indicate that the 3D velocity fields are remarkably similar in both cases. This is expected based on time reversal arguments. Therefore, the shear flow patterns we found for impact have a direct analog for pulling, and the same considerations apply as outlined in Section 4.3.3, above, including the transition to squeeze flow once the jamming front gets close to the bottom boundary. Conversely, the fact that we observe jamming fronts during pulling rules out jamming by compaction (compare for the original ‘snow plow’ model). Instead, it lends strong credibility to the shear jamming scenario, precisely because shear jamming does not require densification.

*Results from this effort have been submitted for publication (Sayantan Majumdar, Ivo R. Peters, Endao Han, and Heinrich M. Jaeger, “Jamming front propagation under pulling in dense particulate suspension” (2015)).*

#### 4.3.5 Dynamic shear jamming and general phase diagram

The findings discussed in the two preceding Sections suggested a strong connection to a shear jamming transition. Shear jamming is, however, well established only for dry granular materials (as an extension of the jamming phase diagram for frictionless systems). In this part of the project we demonstrated the first time direct observation of shear jammed states in dense suspensions and the connection to shear thickening, resulting in a unifying phase diagram. A unique property, different from shear jamming in dry granular materials, is the rate-dependence, which gives rise to transient shear-jamming fronts that share

the properties of fronts that have been observed in impact experiments. Our unifying picture suggests a re-interpretation of existing observations of shear thickening and dynamic jamming fronts. Down the road we expect that it also can serve as a guide to create suspensions with new dynamic properties by design.

Frictional contact between particles is the key property that allows for shear jammed states to exist. While frictionless systems, where particles interact purely via hydrodynamic interactions, show continuous shear thickening, discontinuous shear thickening (DST) is only observed when frictional contacts are taken into account [5]. A number of studies have suggested a connection between DST and shear jamming, but there are no direct observations of shear jamming in suspensions. There has also been no clear way to link the observed steady state shear thickening behavior and transient (impact, pulling) responses. Furthermore, a phase diagram compatible with the one suggested by Bi et al. [20] for dry granular systems is still missing.

In this project component, we showed that using a single experimental system, a large Couette cell, all states of a dense suspension can be observed. Specifically, we could track both the steady state and transient response as well as that shear jammed state with precision. We resolved the apparent contradiction where the onset stress for DST is mostly independent of packing fraction, while the onset stress for shear jamming is strictly decreasing with increasing packing fraction. As a consequence, we showed that DST can be seen as a precursor state to shear jamming rather than representing the shear jammed state itself. Our findings suggested instead that DST corresponds to a fragile state, which shows the behavior of marginal material that is neither freely flowing nor fully jammed.

Experiments were performed with an Anton Paar MCR301 rheometer. We used a measuring system consisting of two concentric cylinders. For the first set of experiments we used a large gap between the cylinders, where the outer cylinder (cup) was a glass container with an inner diameter of 98 mm and a height of 42 mm. This cylinder was filled approximately to the rim with the suspension. The inner cylinder (bob) had a diameter of 26.7 mm and was partially submerged in the suspension. We verified that the measurements are not influenced by the bottom of the cup by performing experiments at different depths of the bob. For a second set of experiments we used both the geometry just described, as well as a standard small-gap measurement cup (inner diameter 28.8 mm). The large-gap geometry was used to observe transient effects at high packing fractions, while we used the small-gap geometry for steady-state experiments at lower packing fractions.

For the experiments investigating the transient response, i.e., the propagation of jamming fronts, the rheometer was set to rotate at a constant speed. The time to accelerate the rheometer tool was always much smaller than the time during which we observed the transient response of the system. The experiments were recorded using a high-speed camera (Phantom V9), recording the top free surface of the suspensions at frame rates up to 4500 frames/sec, depending on the driving speed. Tracer particles on the surface of the cornstarch suspension allowed us to perform Particle Image Velocimetry (PIV) and obtain time-resolved velocity fields (**Fig. 35**). In addition, we measured the torque applied by the rheometer during the experiments. The steady-state experiments with the small-gap geometry were performed by setting a constant torque and measuring the rotation speed as a function of time. We repeated this for a range of torques to obtain viscosity-stress curves for our samples. For a number of samples we performed the same procedure using the large-gap cell to ensure consistency between the different measurement geometries.

The suspensions consisted of a density-matched solution of water, glycerol, and CsCl, mixed with cornstarch. The cornstarch was stored at a controlled temperature (22<sup>0</sup> C) and relative humidity (50 +/- 5 %). For each suspension, we measured the exact density and viscosity. In order to calculate the correct packing fraction of the suspensions, we took into account the moisture content the cornstarch absorbed from the environment.

To illustrate the effect of rate dependence on the transient response we show in **Fig. 35** the evolution of

the velocity field for two different driving speeds. We normalized the measured velocities by the prescribed velocity at the outer wall of the inner (driving) cylinder. We observed strikingly different behavior between slow and fast driving of the inner cylinder. For a slow driving speed (**Fig. 35e-h**), we observed a diffuse steady velocity profile, reminiscent of that of a viscous liquid. At high driving speeds (**Fig. 35a-d**), the response was markedly different: a sharp front developed, which rapidly traveled radially outward. Behind the front the velocity of the suspension was fairly uniform, while ahead of the front the suspension was still at rest. After the front reached the outer wall (**Fig. 35d**), the system fully jammed while the inner cylinder continued rotating. In the following, we first analyze the properties of this front for different driving speeds, packing fraction and solvent viscosity, and then use the data to develop a phase diagram.

At driving speeds within the regime where we observed the formation of distinct, sharp jamming fronts, the front speed was found to be proportional to the driving speed. The front position as a function of time for three different driving speeds is shown in **Fig. 36a**, showing an increase of front speed with increasing driving speed. Rescaling the x-axis by the driving speed collapses these curves, as is shown in **Fig. 36b**. The collapse demonstrates that the ratio between the driving speed and the front propagation speed is independent of the driving speed. This suggests that the front is resulting from a (local) critical strain, which locally jams the system. Once the critical strain is reached and the system is jammed locally, it will be able to strain the still unjammed suspension near it. This process will continue as long as the system

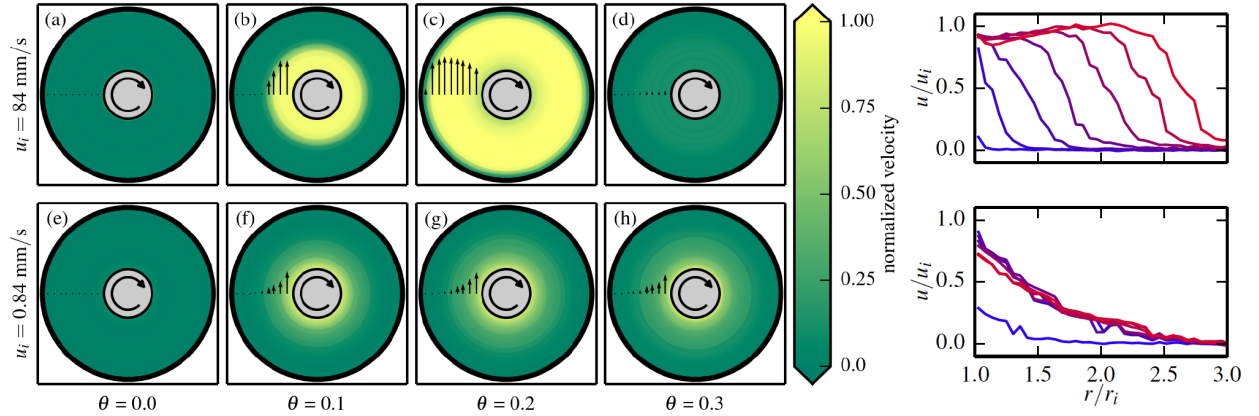


**Figure 35.** Evolution of the velocity profile for different driving speeds. Left hand side images (a-h): Driving speeds 84 mm/s (a-d) and 8.4 mm/s (e-h). Images for both driving speeds are given for the same amount of strain, velocities are normalized by the driving speed. Color and arrows give local velocity as obtained from PIV analysis. Fast driving shows the formation of a jamming front and eventually a shear-jammed state (d), whereas the slow driving results in a velocity profile that gradually diffuses outwards. Right hand side plots: Radial velocity profiles for fast (top) and slow (bottom) driving, corresponding to the images on the left.

is actively driven and no boundary has been reached. The closer the packing density is to the jamming point for the suspension at rest, the smaller a critical strain will be required to reach a shear-jammed state [20]. In terms of the propagation of the jamming front, this means that its speed will increase with packing fraction. This is also shown in **Fig. 36b**, where the different symbols denote different packing fractions.

We can take the connection between front propagation in suspensions and shear jamming of dry granular systems one step further by accounting for an important qualitative feature of the shear jamming phase diagram by Bi et al. [20], namely that critical stress and critical strain both approach zero as the packing fraction approaches the isotropic, frictionless jamming point  $\phi_j$ . A vanishing critical strain would, according to our reasoning above, result in a diverging front speed, since an infinitesimal perturbation would immediately propagate through our complete system. The functional form of how the critical strain approaches zero, and consequently how the speed diverges as  $\phi_j$  is approached is *a priori* unknown. We

adopted a generalized form of the analytical expression for front speeds due to the compression of granular materials close to the jamming point, which predicts a ratio of front speed to driving speed given by  $k = \phi/(\phi_J - \phi)$  [10, 23]. Even though our experiment does initiated jamming by Couette shear rather than compression, we found that the same expression for  $k$  still fit the data well (**Fig. 36c**). We emphasize, however, that the mechanisms are different and the only shared property is an expected divergence as point  $\phi_J$  characterizing frictionless jamming is approached. As can be observed in **Fig. 36d**, which gives the torque response for the different driving speeds, there is a critical driving speed where the torque response jumps by orders of magnitude, much like in the DST transition.

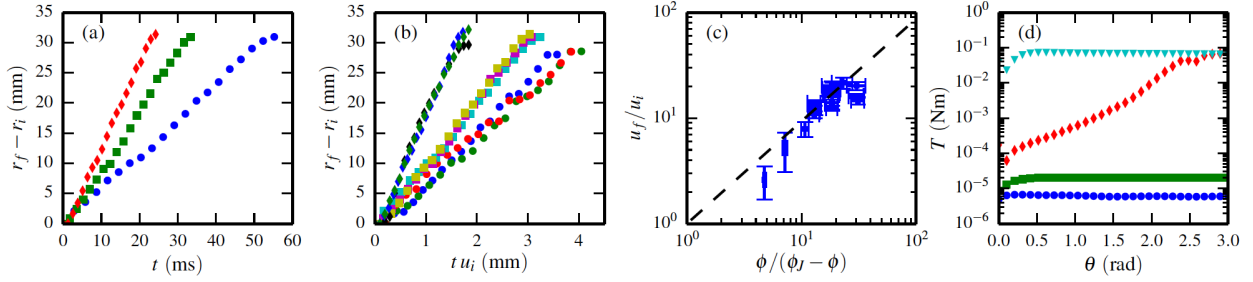


**Figure 36.** Influence of driving speed and packing fraction. (a) Position of the front for different driving speeds (blue circles: 59 mm/s, green squares: 92 mm/s, red diamonds: 126 mm/s);  $\phi = 0.52$ . (b) Front position normalized by driving speed for different packing fractions (circles:  $\phi = 0.51$ , squares:  $\phi = 0.52$ , diamonds:  $\phi = 0.53$ ). (c) Front and driving speed ratio  $k$  vs packing fraction (blue data points), dashed red line:  $\phi_J$ . (d) Measured torque for different driving speeds spanning 3 orders of magnitude (blue circles: 84  $\mu$ m/s, green squares: 0.84 mm/s, red diamonds: 8.4 mm/s, cyan triangles: 84 mm/s). Note the crossover in behavior around 8.4 mm/s (red).

The analysis so far has been on the transient properties above the shear rate at which we dynamically jam our system. A remaining question is how this is related to the steady state properties of dense suspensions. In previous works, the transient and steady state have been investigated independently in different systems suitable for only one of the two branches. Our investigation was the first to study both in a single system.

For each sample, which is defined by its packing fraction and the solvent viscosity, we identified the following behaviors: Newtonian (N), Shear Thinning (ST), Continuous Shear Thickening (CST), Discontinuous Shear Thickening (DST), and Shear Jammed (SJ). All the measurements that contribute to the phase diagram were performed under stress controlled conditions, which ensured that the DST regime can be resolved accurately. **Figure 37a** shows an example of such a measurement. The DST regime was identified by a quadratic or stronger increase of the viscosity with the applied stress [5, 24, 25]. We determined the onset of DST by a linear fit to the DST regime on a  $\log \eta$  vs.  $\log \tau$  plot, and found the stress value at which it intersects with a constant viscosity from the Newtonian regime. The region where the viscosity increases with increasing stress, but which is below the onset of DST we defined as CST. Similarly, we labeled as CST the continuous increase (slope less than 1 on a  $\log \eta$  vs.  $\log \tau$  plot) which we observe at lower packing fractions after the DST regime (note that this definition is simply operational; we are not implying similar mechanisms; in fact, this second regime labeled CST is likely simply a crossover due to internal softening of the suspension or due to softening of the boundaries).

At higher packing fractions, the CST regimes became much smaller or vanished completely, which resulted in a direct transition from Newtonian into DST, directly followed by shear jamming (SJ). We identified the SJ state by impacting the suspension with steel spheres (diameter 5.0 mm) and tracking their position after they touch the surface of the suspension (**Fig. 37b**). As we applied more stress to our system, the trajectory of the sphere changed from slowly sinking in to rebounding and remaining on the surface for as long as stress was applied to the suspension by rotating the inner (driving) cylinder.

By performing experiments across a large range of packing fractions we constructed the phase diagram shown in **Fig. 37c**.

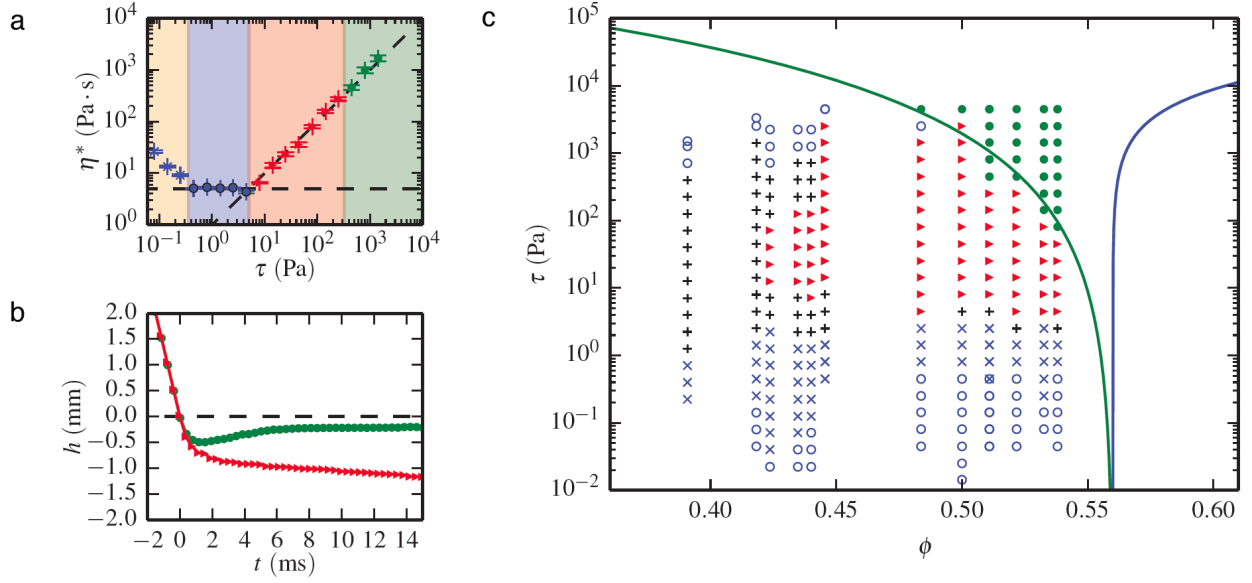


**Figure 37.** Transitions between the different dynamic states of a dense suspension. a, Viscosity-stress curve for a single packing fraction, with different states indicated by different colors: shear thinning (orange), Newtonian (blue), discontinuous shear thickening (red), and shear jammed (green). In this example, the continuous shear-thickening phase is negligible. b, Trajectories of spheres impacting a suspension subjected to different magnitudes of shear stress, showing a slowly sinking sphere in the DST regime (red curve) and rebound in the shear jammed regime (green curve). c, Phase diagram delineating the different dynamic states of a suspension as a function of two control parameters, packing fraction  $\phi$  and shear stress  $\tau$ : shear thinning (blue circles), Newtonian (blue crosses), continuous shear thickening (black crosses), discontinuous shear thickening (red triangles) and shear jammed (green dots).

We point out an important difference with previous studies, which have suggested DST itself as the shear jamming transition [26-29]. These studies have shown that the shear rate at which DST is observed approaches zero towards the jamming point, which is only in apparent agreement with the shear jamming phase diagram, because the phase diagram is based on shear stress [20]. Other studies have shown that the onset stress for DST, however, is *independent* of packing fraction, and even increases on the approach to the isotropic jamming point, which is due to a yield stress at high packing fractions [5, 25, 30]. The onset stress for DST therefore seems in direct contradiction to the predictions of the shear jamming phase diagram. *In this project we resolved this contradiction by identifying the shear jamming transition as the upper bound for the DST regime.*

Our observations are in agreement with all the previous studies, showing both the decrease in shear rate and the constant onset stress for DST. Focusing on the upper boundary where we enter the shear jammed states, however, we found new behavior. Close to the isotropic jamming point at  $\phi_j = 0.56$ , there is a clear decreasing trend in the shear jamming transition, which is consistent with the picture of Bi et al. and gives the first clear hint of a phase diagram that unifies DST with shear jamming. As we move towards lower packing fractions, we see an increased appearance of CST, which partially replaces DST, until eventually DST has completely disappeared. This provides a first estimate for the packing fraction boundary below which SJ states cannot be reached anymore, corresponding to  $\phi_s$  in Ref. 20. We expect the actual boundary for  $\phi_s$  to be at a somewhat higher packing fraction, where there is no CST anymore at higher stresses. We were however not able to reach this regime, due to limitations in the stress that we can apply with our current experimental system.

*These finding have been submitted for publication (Ivo R. Peters, Sayantan Majumdar, and Heinrich M. Jaeger, “Dynamic shear jamming” (2015)).*

#### 4.4 Other projects

With support from this grant, two further projects were carried out. They are highlighted in this Section.

##### 4.4.1 Onset of Splashing During Impact of Suspension Droplets

In the experiments described so far, a solid object impacted a dense suspension at rest. The opposite situation exists when a suspension is slowly extruded from a nozzle and the resulting droplets, after freefall, impact a flat solid surface. For simple liquids the impact energy can produce splashing, and the determination of the parameters that predict the splashing onset has long been of much interest both scientifically and for applications such as coating or printing. The question we addressed was whether adding small particles to a pure liquid would enhance or reduce the propensity for splashing.

There has been a growing interest in what happens to the spreading and splashing if particles are added to the liquid. On micron scales,  $\text{ZrO}_2$  suspensions have been used in studies aiming to optimize ink-jet printing applications, and on truly macroscopic scales there has been the development of 3D printers that dispense cement slurry. In all of these situations, an important concern is to prevent splashing, and particles from escaping, when droplets hit a surface. However, the question of when and why particles are ejected has remained unsettled, and existing experimental studies mostly focus on dilute suspensions.

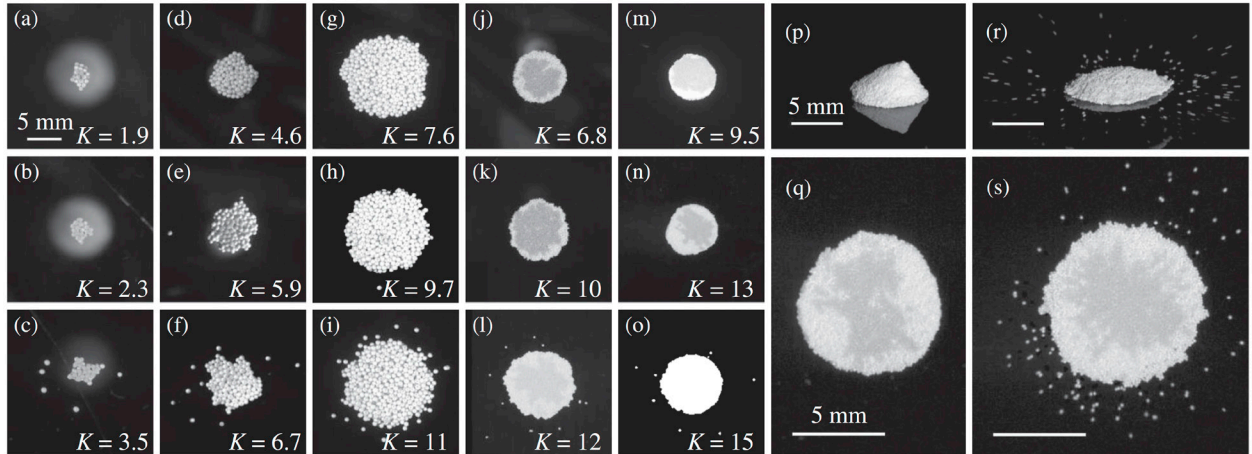


**Fig. 38.** The images above are stills from high-speed videos. They show the splats right after impact and are produced by dense suspensions of  $\text{ZrO}_2$  particles in water that are dropped onto a smooth glass plate. Outcomes are shown for different droplet radii  $r_d$  (see text above the images) and particle radii: (a-i)  $r_p = 362 \mu\text{m}$ , (j-l)  $r_p = 138 \mu\text{m}$ , (m-s)  $r_p = 78 \mu\text{m}$ . Images (a-o) are organized in vertical columns, with drop impact speed and impact parameter  $K$  increasing from top to bottom in order to bracket the onset of splashing, defined here as the ejection of individual particles. For the  $K$  values listed in the top/middle/bottom rows we never/sometimes/always observed particle ejection. The blurred background in (a) and (b) is the out-of-focus image of the syringe. Note that (o) has been thresholded and dilated in order visualize the ejected particles that would otherwise be invisible due to their small size. (p,q) side and bottom view of a droplet that does not splash ( $We_p = 12$ ). (r,s) side and bottom view of a splashing droplet ( $We_p = 26$ ). The scale bar in the images is 5 mm, images (a-o) all have the same scale.

Current models for suspension drop impact associate the onset of splashing with the condition that the impact parameter  $K = We^{1/2} Re^{1/4}$  exceeds a critical value  $K_0$ . This has been the same as traditional criterion for pure liquid splashing on dry surfaces at atmospheric pressure in a regime independent of surface roughness (but fails, for example, at lower pressures). Here the Weber and Reynolds numbers are

defined as  $We = \rho r_d U^2 / \gamma$  and  $Re = \rho r_d U / \eta$  where  $r_d$  the droplet radius,  $U$  the droplet impact velocity, and  $\rho$  the suspending liquid's density,  $\gamma$  its surface tension, and  $\eta$  its dynamic viscosity. In these models, the addition of particles has been captured by replacing  $\eta$  with an effective viscosity  $\eta_{eff}$  that increases with the packing fraction.

We demonstrated that this onset criterion fails dramatically for dense suspensions (see **Fig. 38**) and that the underlying global hydrodynamic balance, which considers stresses at the level of the droplet as a whole, is unable to predict the splash onset correctly. We propose to replace it by an energy balance at the level of the particles in the suspension. We experimentally verified that the resulting, particle-based Weber number  $We_p = \rho_p r_p U^2 / \gamma$  gives a reliable, particle size  $r_p$  and density  $\rho_p$  dependent splash onset criterion, with a transition to splashing occurring when  $We_p > 14$  for the case of inviscid suspending liquids. We further showed that the same argument can explain why, in bimodal systems, smaller particles are more likely to escape than larger ones.

*These findings were published in Physical Review Letters (I. R. Peters, Q. Xu, and H. M. Jaeger, Splashing Onset in Dense Suspension Droplets, Phys. Rev. Lett. **111**, 028301 (2013)).*

#### 4.4.2 Single Particle Charge Measured by Free-Fall Videography

In a separate line of work, focusing on dry granular materials, we developed a new experimental technique for measuring individual particle charges in large ensembles of macroscopic grains. The measurement principle is qualitatively similar to that used in determining the elementary charge by Millikan in that it follows individual particle trajectories. However, by taking advantage of new technology we were able to work with macroscopic grains and achieve several orders of magnitude better resolution in charge-to-mass ratios. By observing freely falling grains accelerated in a horizontal electric field with a co-falling, high-speed video camera, we dramatically increased particle tracking time and measurement precision.

**Figure 39** shows a sketch of the apparatus. Particles are kept in a container (hopper) inside a vacuum system. As they leave the hopper through a small orifice at the bottom (nozzle) they enter a freefall region. Individual particles are deflected into the x-direction (into or out of the plane of the figure) by a horizontal electric field applied between two large copper plates. Keeping the granular medium under vacuum, we eliminate air drag, leaving the electrostatic force as the primary source of particle accelerations in the co-moving frame. The camera moves on two low-friction rails and, because it accelerates to (nearly) freefall, it can track particle trajectories over long times. Because the technique is based on direct imaging, we can distinguish between different particle types during the experiment, opening up the possibility of studying charge transfer processes between different particle species.

For the  $\sim 300 \mu\text{m}$  diameter grains used in the test experiments we performed, we achieved an average acceleration resolution of  $\sim 0.008 \text{ m/s}^2$ , a force resolution of  $\sim 500 \text{ pN}$ , and a median charge resolution  $\sim 6 \times 10^4$  elementary charges per grain (corresponding to surface charge densities  $\sim 1$  elementary charge per  $\mu\text{m}^2$ ). This high degree of resolution and the ability to visually identify particles of different species or sizes with direct imaging make this a powerful new tool to characterize charging processes in granular media.

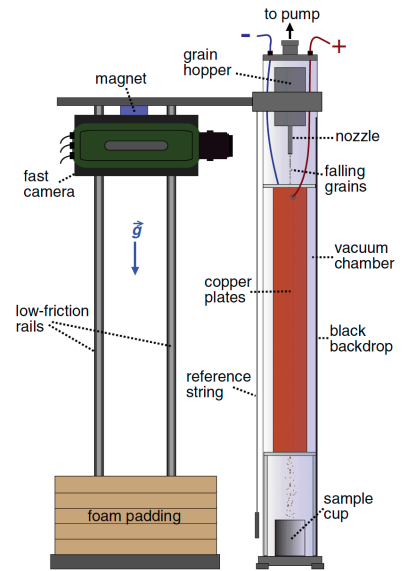


**Fig.39.** Sketch of experimental apparatus for free-fall videography of granular streams. The total height of the cylindrical chamber on the right is 3m.

*This work was published in Review of Scientific Instruments* (Scott Waitukaitis and Heinrich M. Jaeger, “In situ granular charge measurement by free-fall videography”, *Rev. Sci. Instr.* **84**, 025104 (2013)).

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