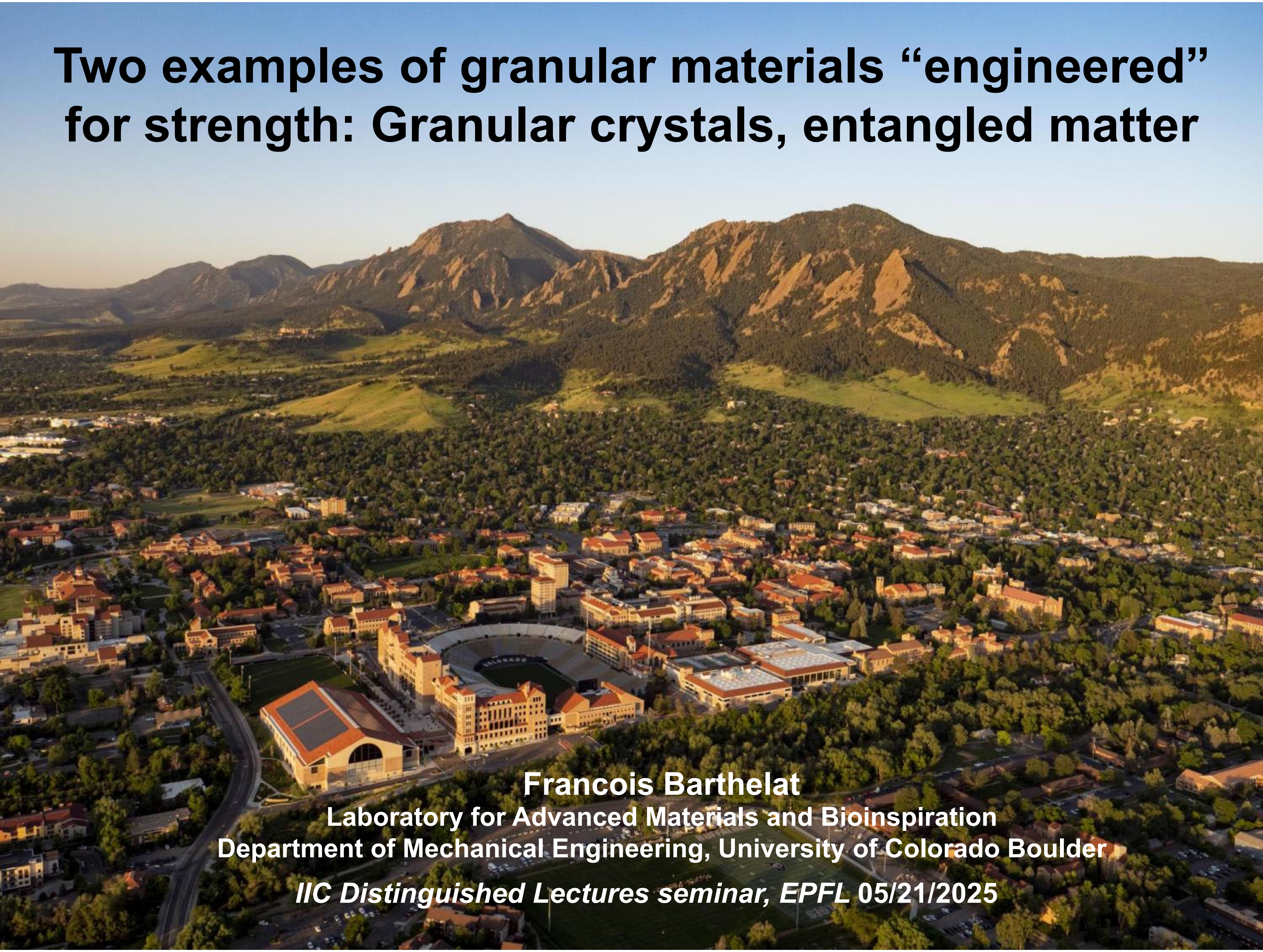


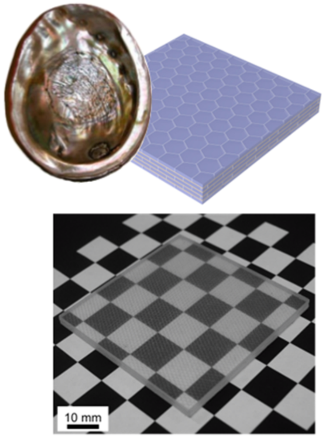
Two examples of granular materials “engineered” for strength: Granular crystals, entangled matter

Francois Barthelat
Laboratory for Advanced Materials and Bioinspiration
Department of Mechanical Engineering, University of Colorado Boulder

IIC Distinguished Lectures seminar, EPFL 05/21/2025

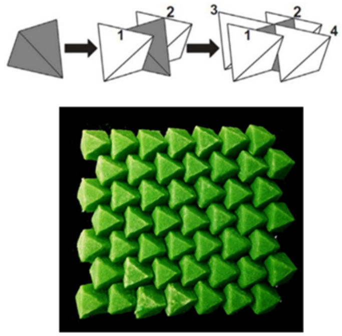
Our approach to materials design…

A nacre-like glass



Yin, Barthelat, *Science* (2019)

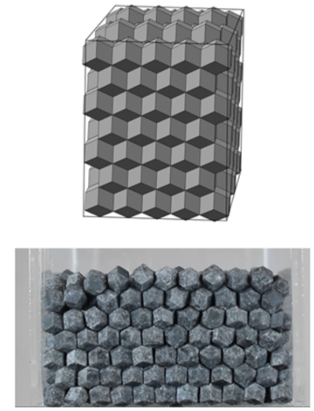
Topologically interlocked panels



30 mm

Mirkhalaf, Barthelat, *PNAS* (2018)

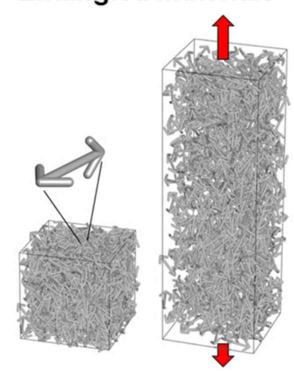
Granular crystals



25 mm

Karuriya, Barthelat, *PNAS* (2023)

Entangled materials



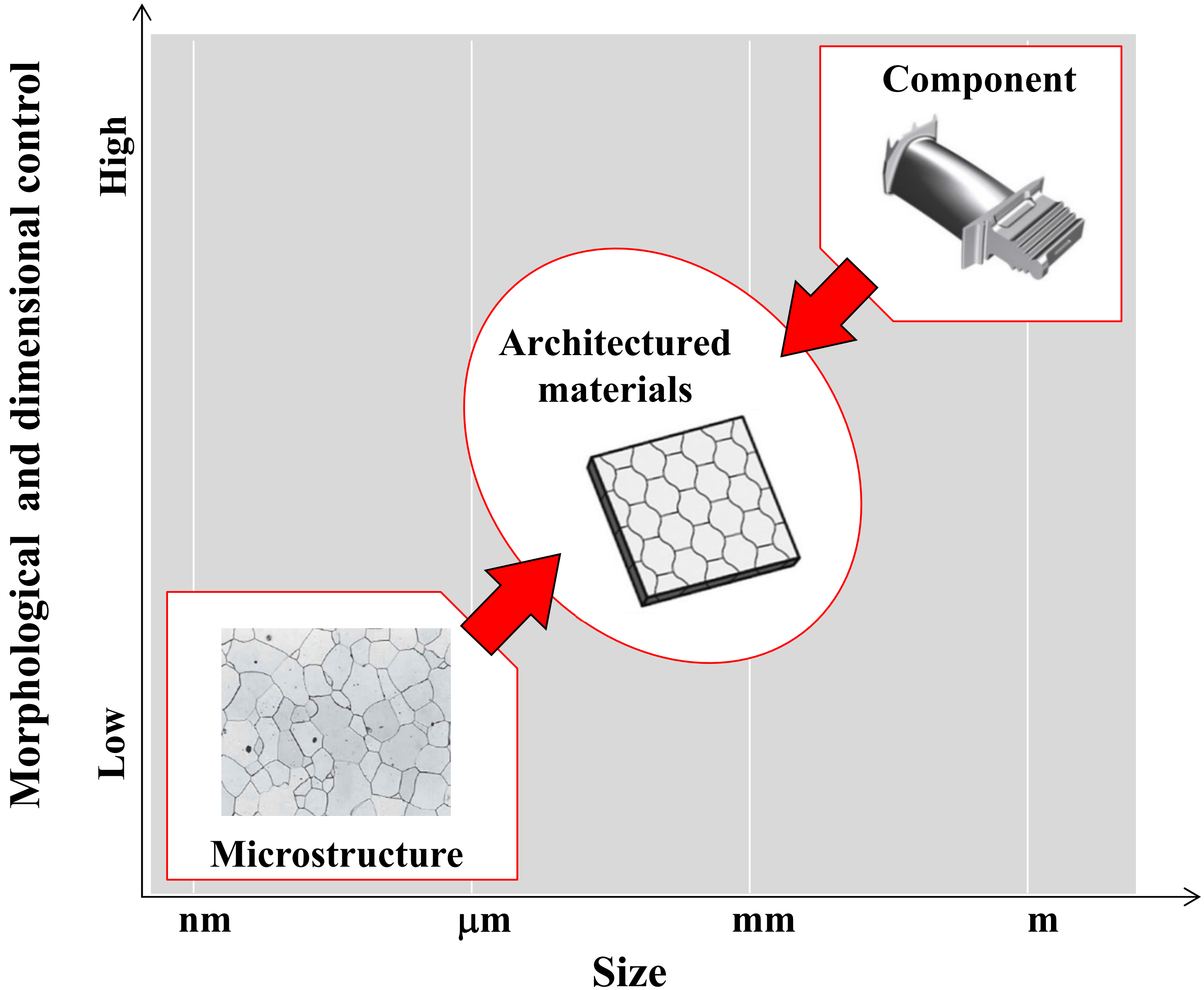
50 mm

Pezeshki, Barthelat *JMPS* (2025)
Sohn, Barthelat *Granular Matter* (2025)

General design recipe:

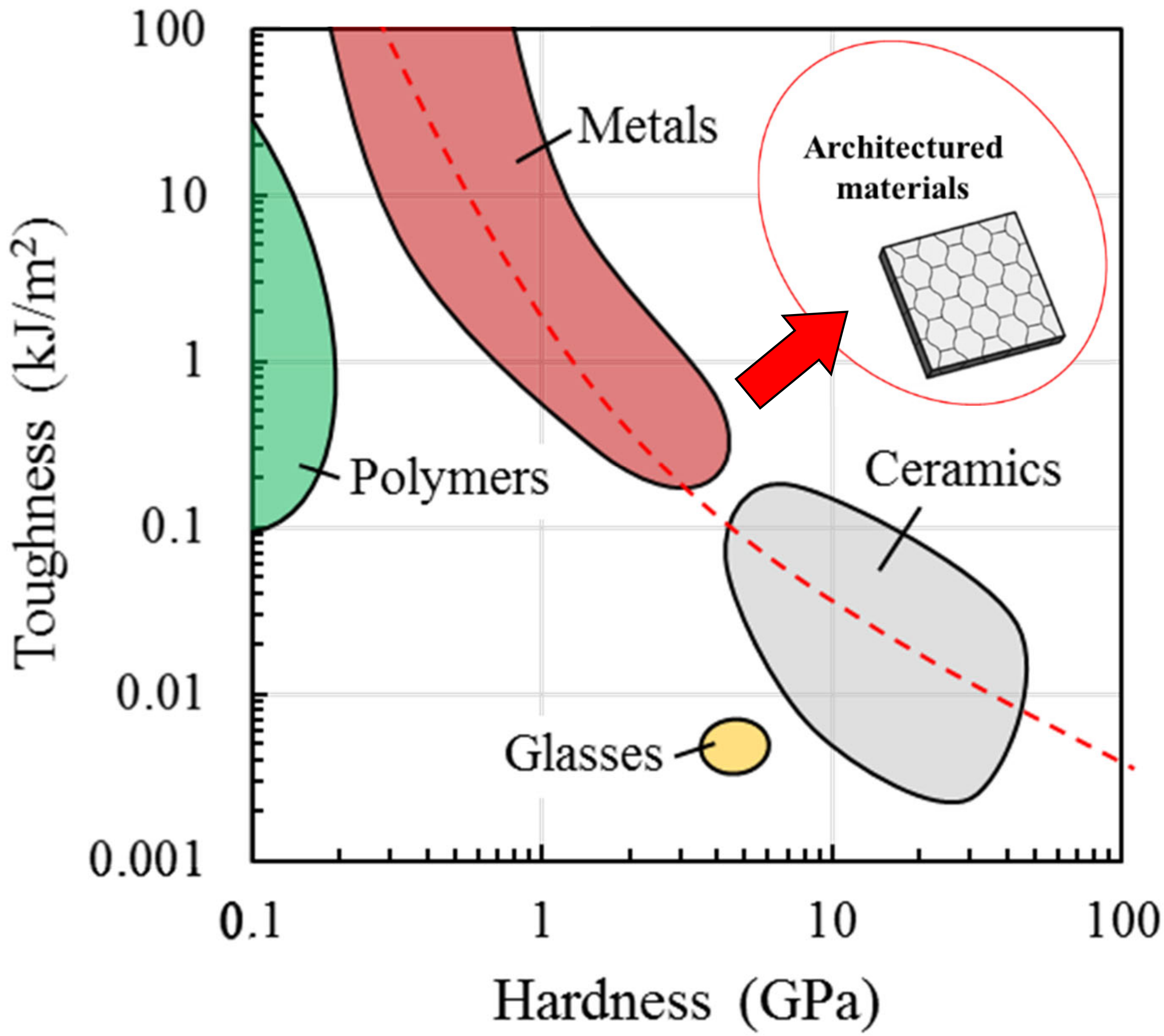
- Stiff, millimeter size, identical "building blocks"
- Material architecture
- Weaker interfaces
- Geometric hardening, interlocking

Material architecture

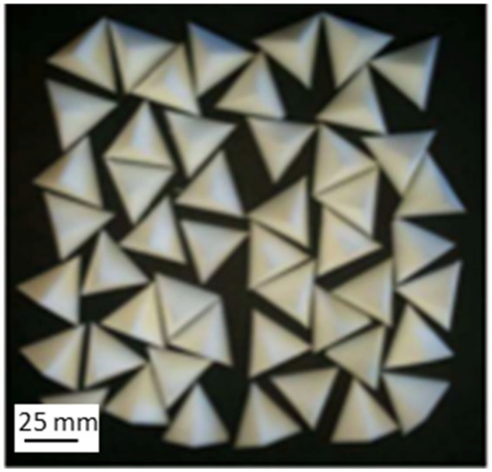


Adapted from Brechet & Embury Scripta Materialia 68 (2013)
F. Barthelat, International Materials Reviews 60(8) (2015)

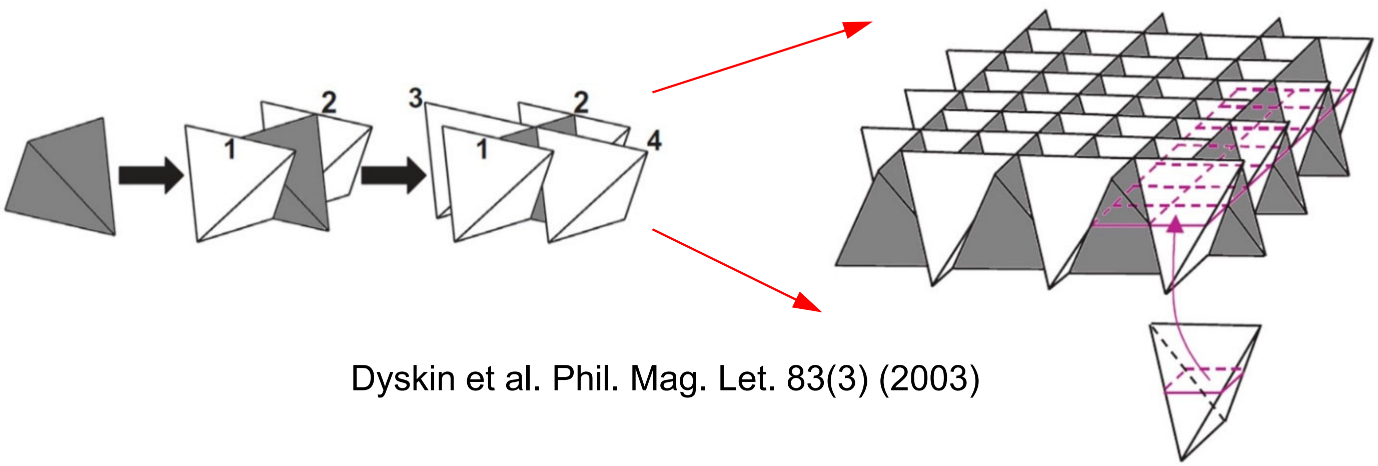
Architectured materials for strength and toughness



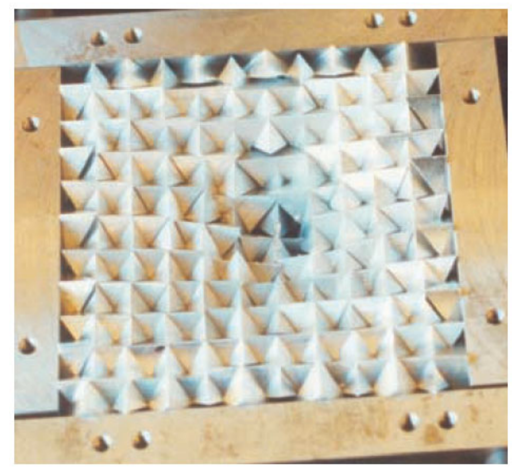
Topologically interlocked materials (TIMs)



- Interlocked tetrahedral blocks
- No physical joining
- Individual blocks remain elastic, but can slide on one another: large inelastic deformations are possible at the macroscale
- Toughness is gained, but generally at the expense of strength

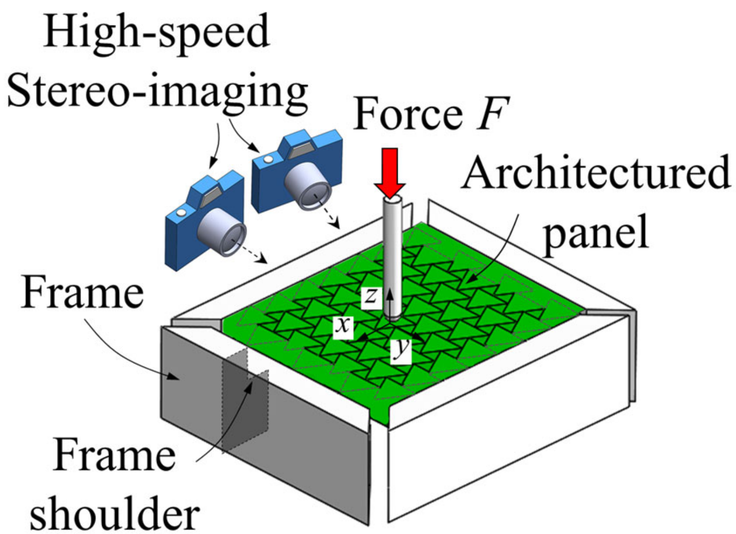


Dyskin et al. Phil. Mag. Let. 83(3) (2003)

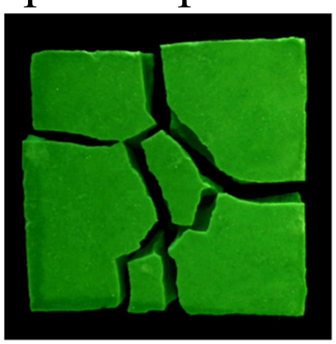


S. Khandelwal and T. Siegmund
Int. J. Solids Struct. 49 (2012)

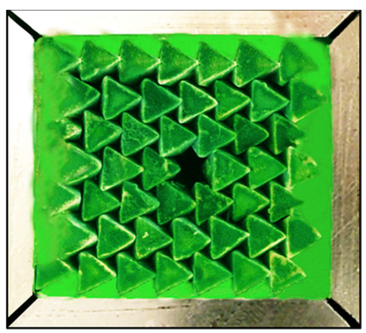
Experiments

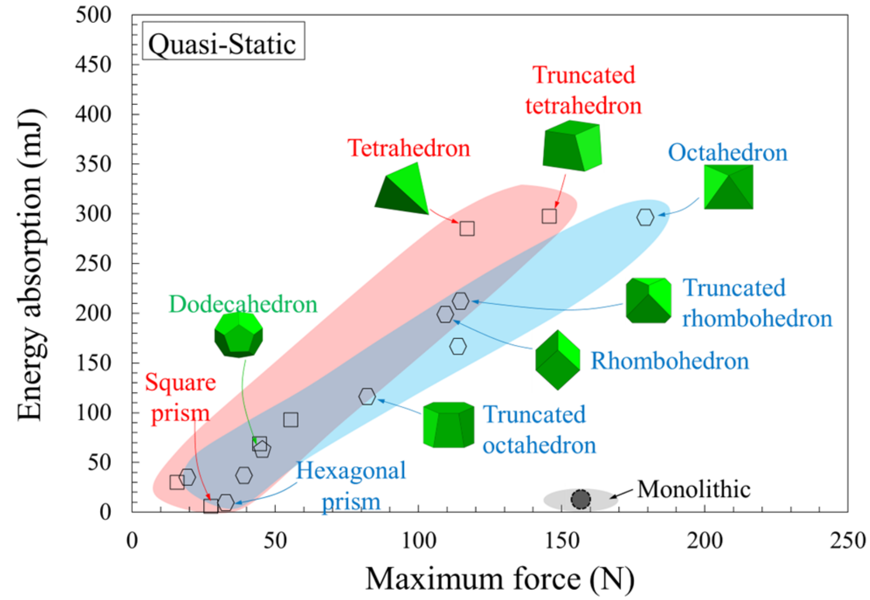


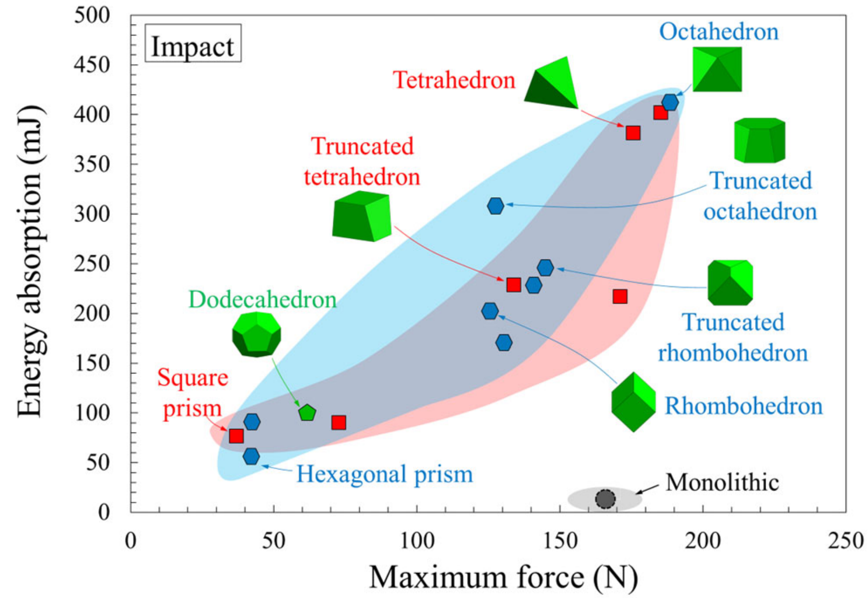
Monolithic plaster panel



Architectured plaster panel

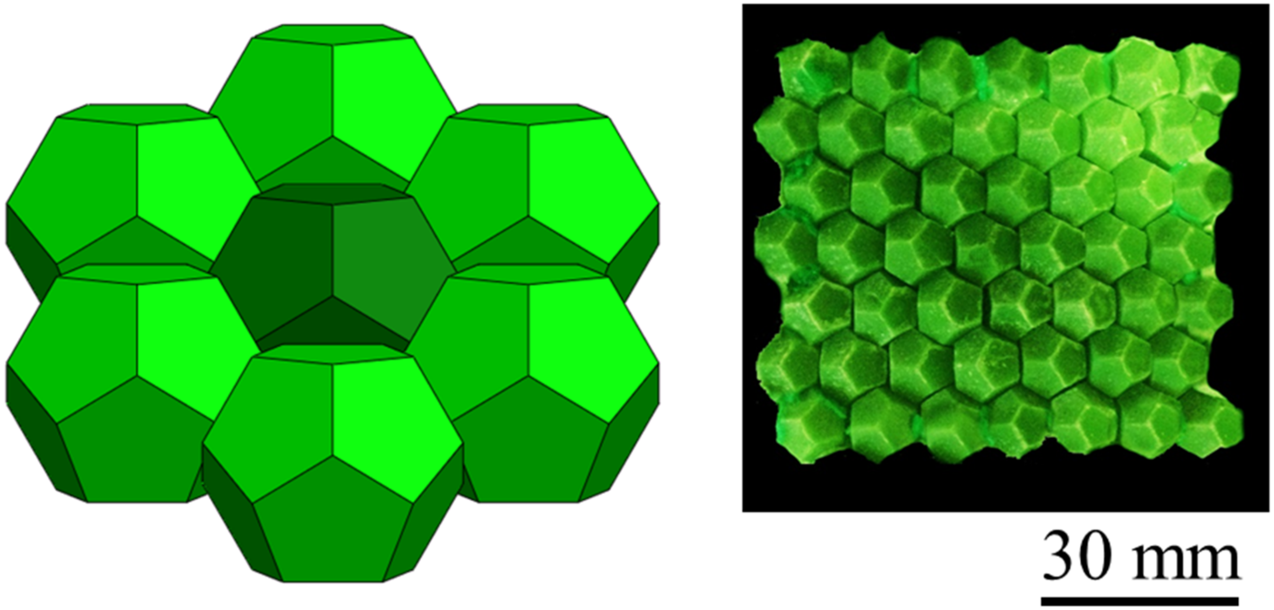




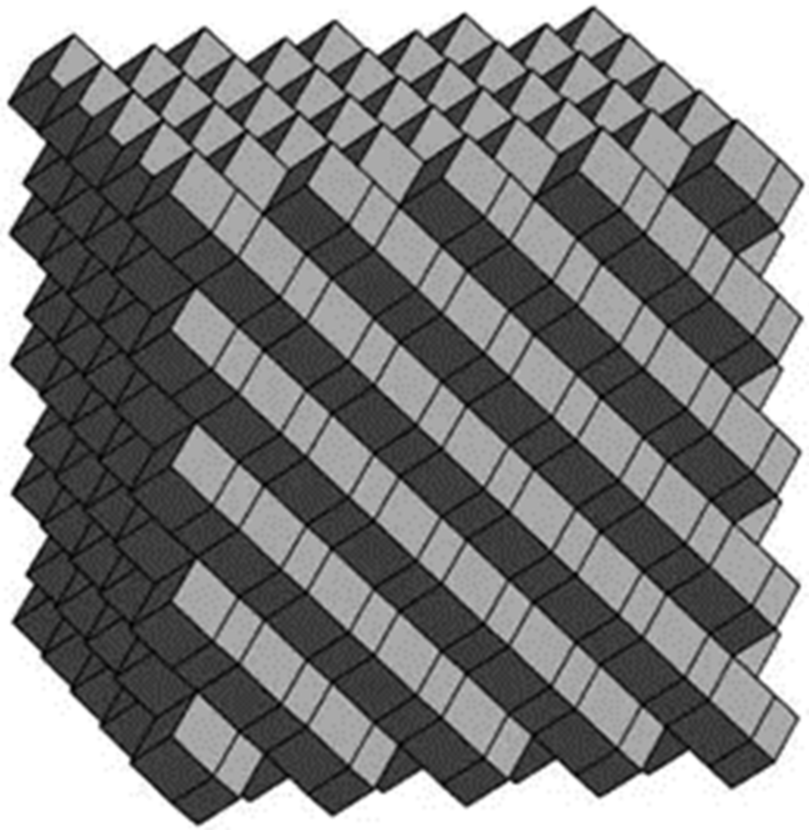


Octahedral blocks provide the highest performance: 50 times improvement in impact resistance, 20% improvement in strength

Are topologically interlocked panels *2D granular crystals?*



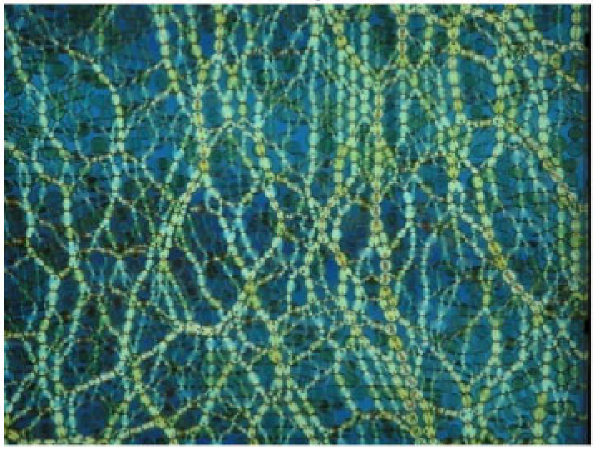
Expand these concepts to 3D materials?



Mechanics of sand and other typical granular materials

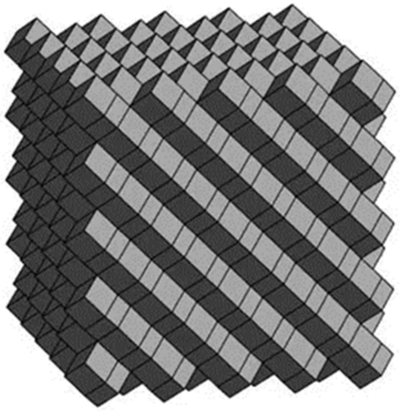
- Sand – and granular materials in general – are fascinating materials that can behave like solid or liquids
- Traditional granular materials make poor structural materials: poor packing, suboptimal load transfer and randomness limit design possibilities, performance and advanced functionalities

Load transfer in typical granular materials is localized along thin force lines

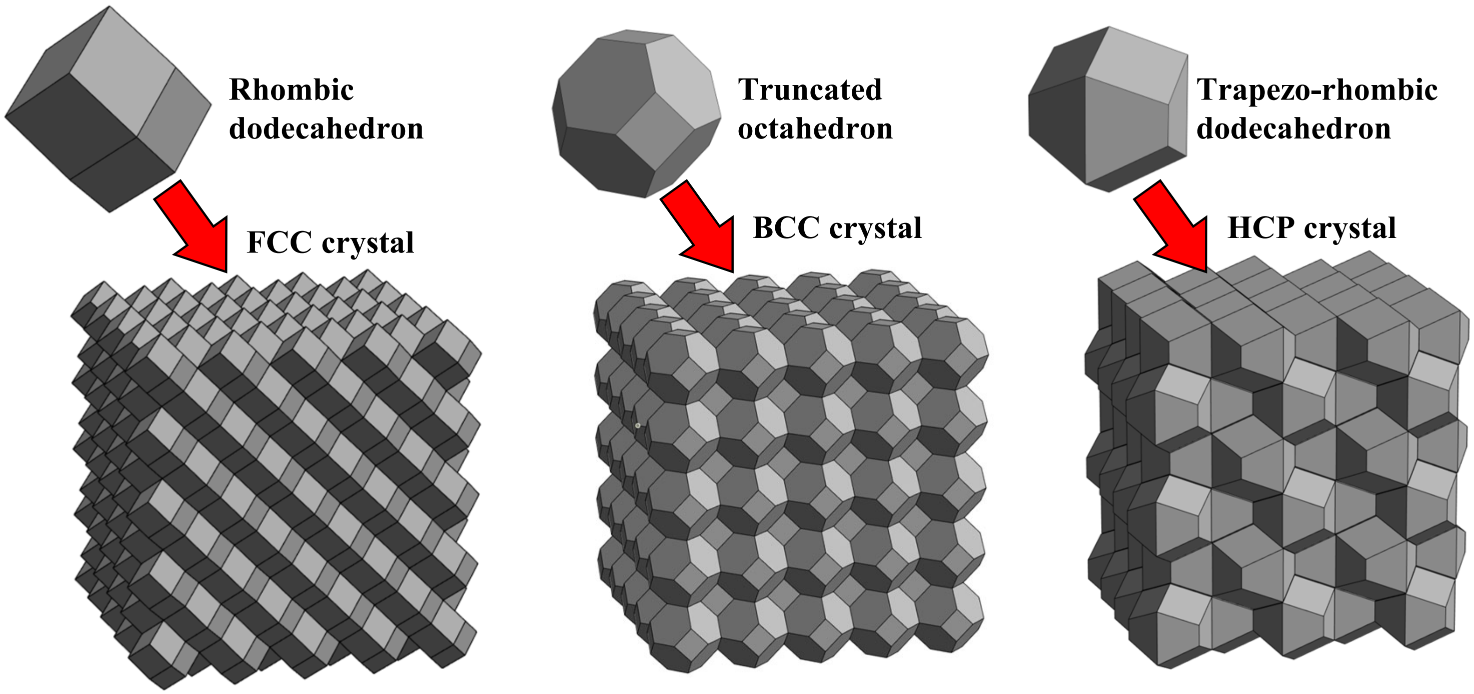


Behringer, Rep. Prog. Phys. 82, 012601 (2018)

Can we design the shape of the grains, their arrangement and their interfaces to improve mechanical performance?



Fully dense granular crystals?



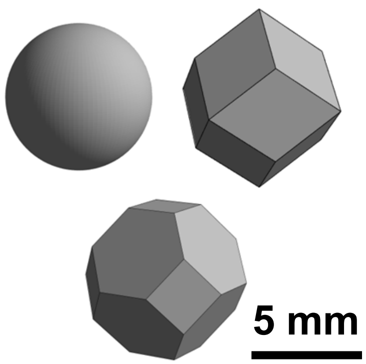
Key Advantages

- **Optimized flat-on-flat contact mechanics**
- **Dense network of ordered force lines**
- **High stiffness and strength**
- **Mechanics and properties can be manipulated over multiple length scales**

Navdeep Karuriya

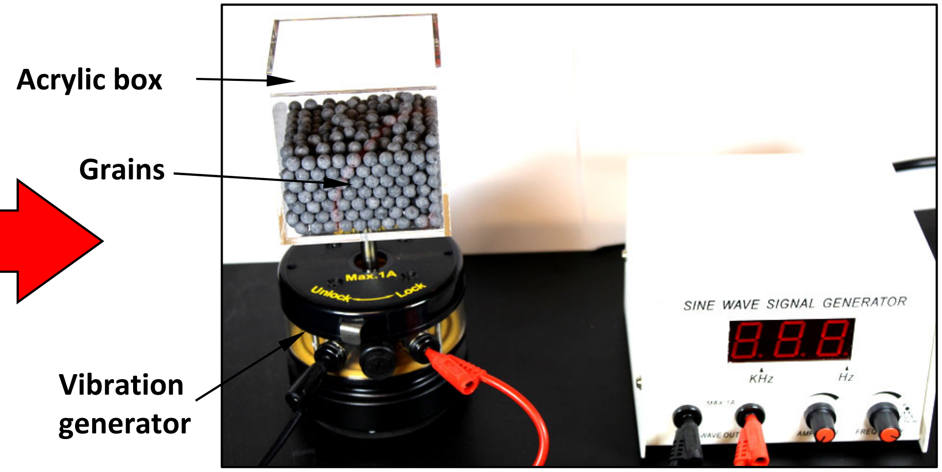
Fabrication and assembly

Solid modeling



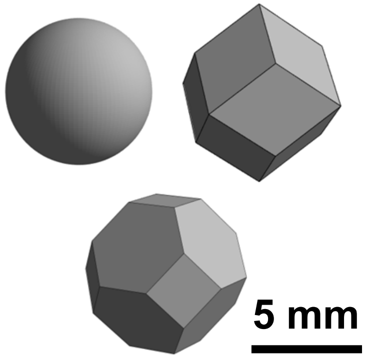
High-res 3D printing (digital light processing)

Vibration platform



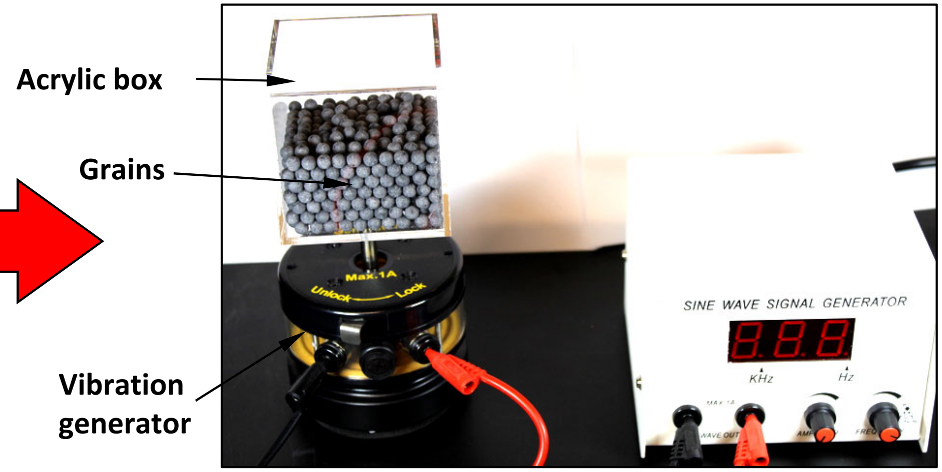
Fabrication and assembly

Solid modeling

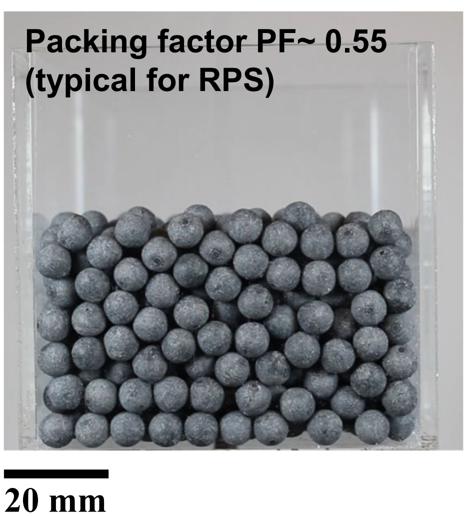


High-res 3D printing
(digital light processing)

Vibration platform

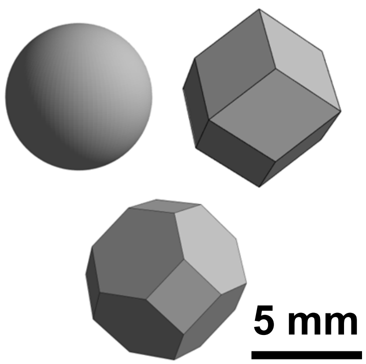


Crystallization of spherical grains
(25 Hz, 2mm and 5mm amplitudes)



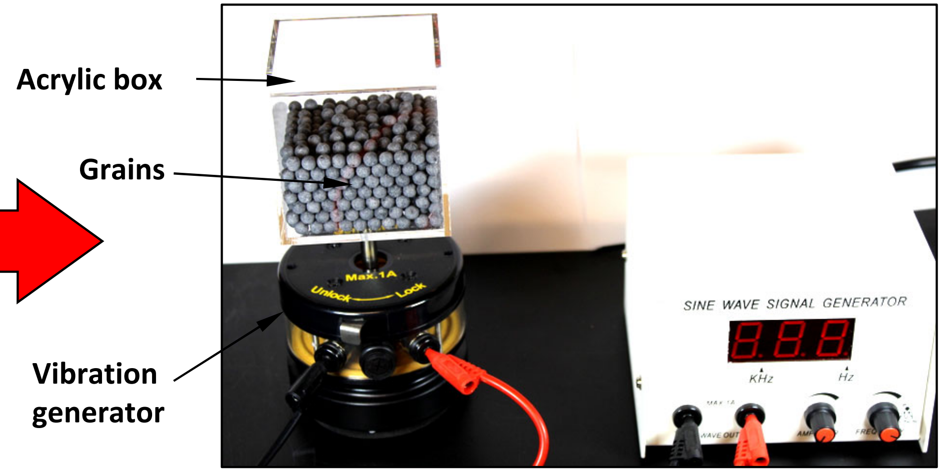
Fabrication and assembly

Solid modeling

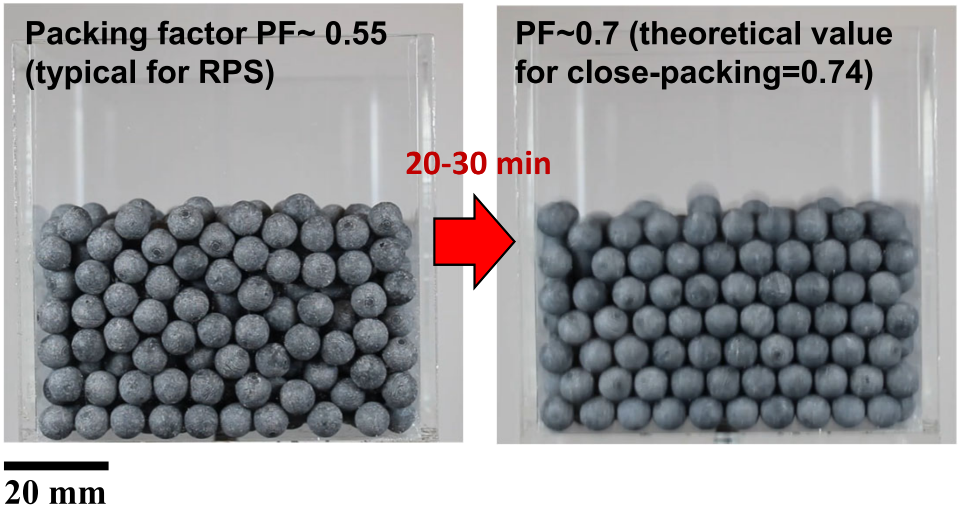


High-res 3D printing (digital light processing)

Vibration platform

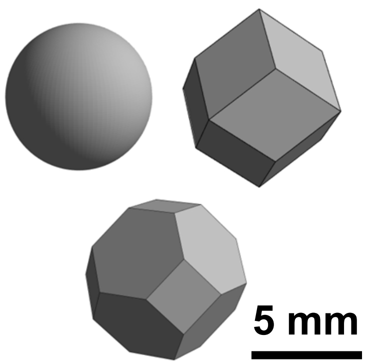


Crystallization of spherical grains (25 Hz, 2mm and 5mm amplitudes)



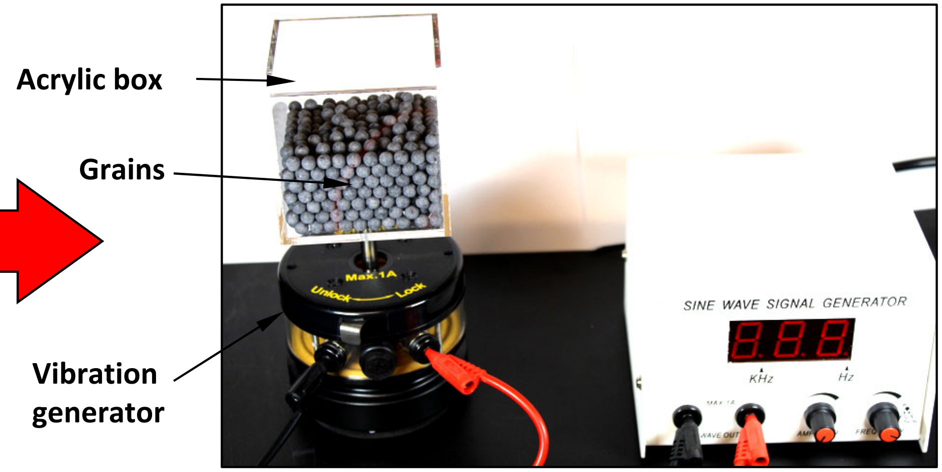
Fabrication and assembly

Solid modeling

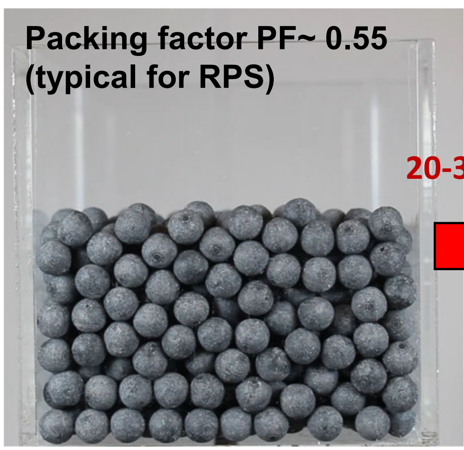


High-res 3D printing
(digital light processing)

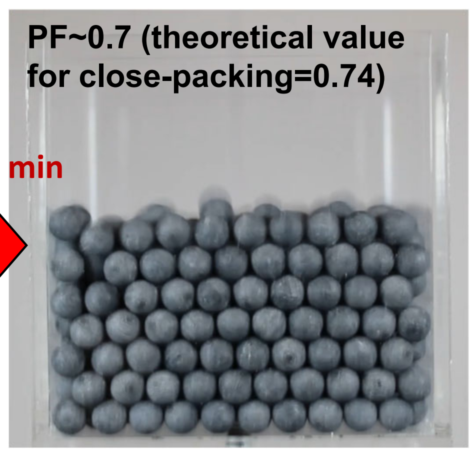
Vibration platform



Crystallization of spherical grains
(25 Hz, 2mm and 5mm amplitudes)



20-30 min



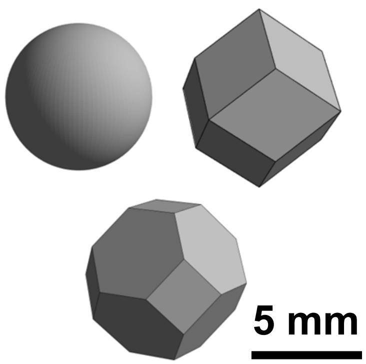
20 mm

Crystallization of dodecahedral grains
(25 Hz, 2mm and 5mm amplitudes)

20 mm

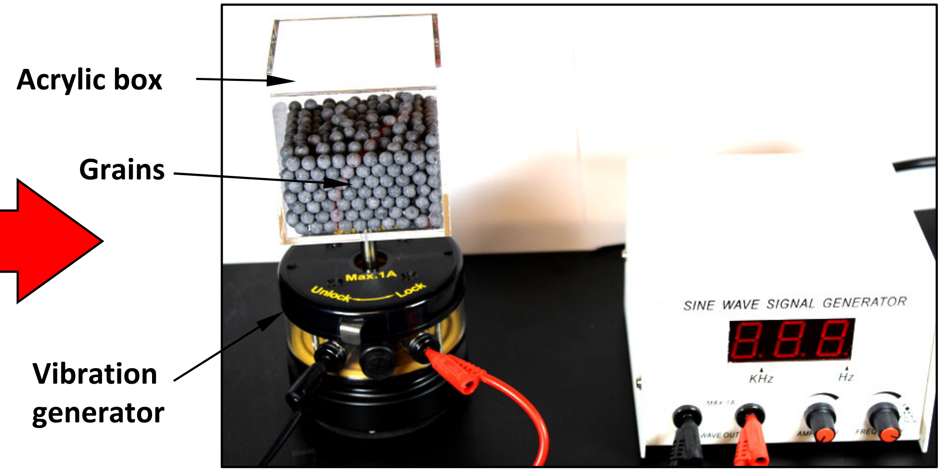
Fabrication and assembly

Solid modeling

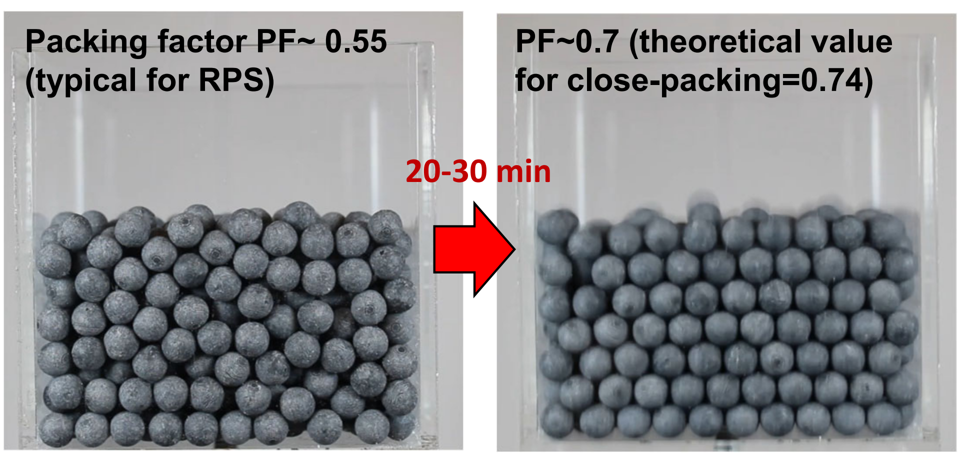


High-res 3D printing (digital light processing)

Vibration platform

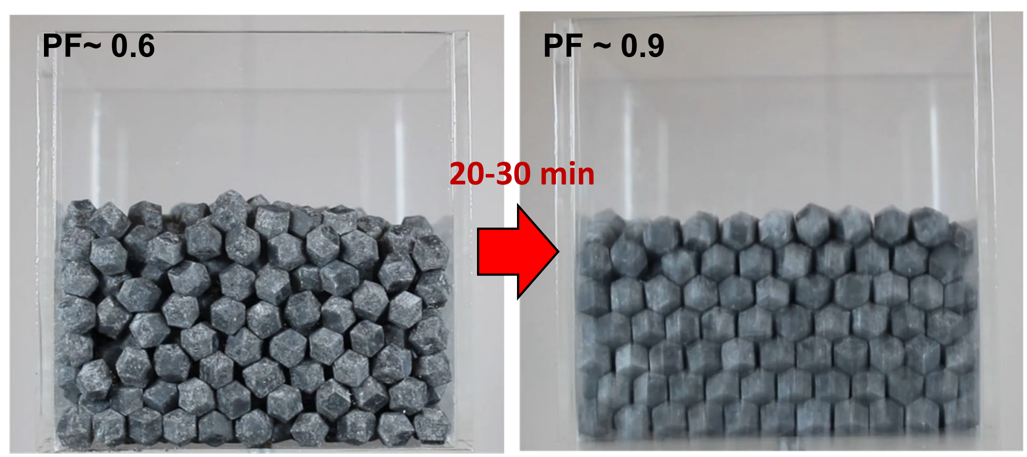


Crystallization of spherical grains (25 Hz, 2mm and 5mm amplitudes)



20 mm

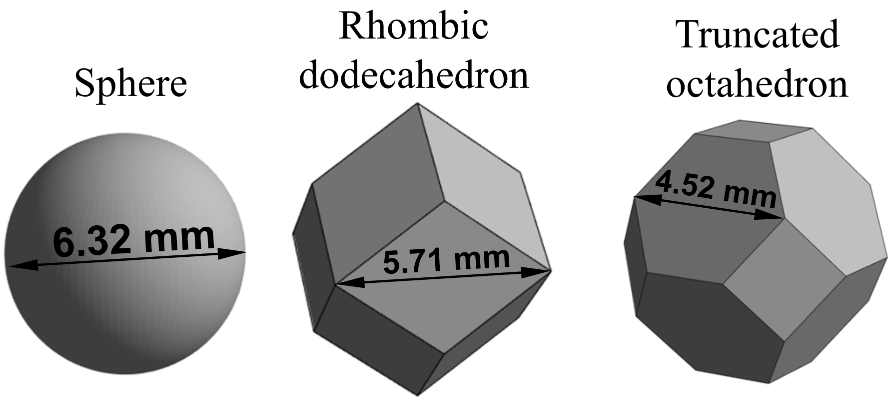
Crystallization of dodecahedral grains (25 Hz, 2mm and 5mm amplitudes)



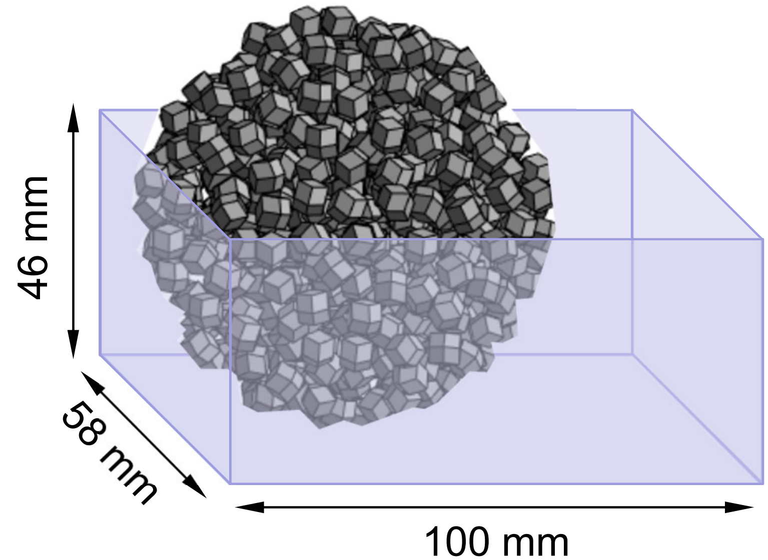
20 mm

Fine tuning the experimental protocol

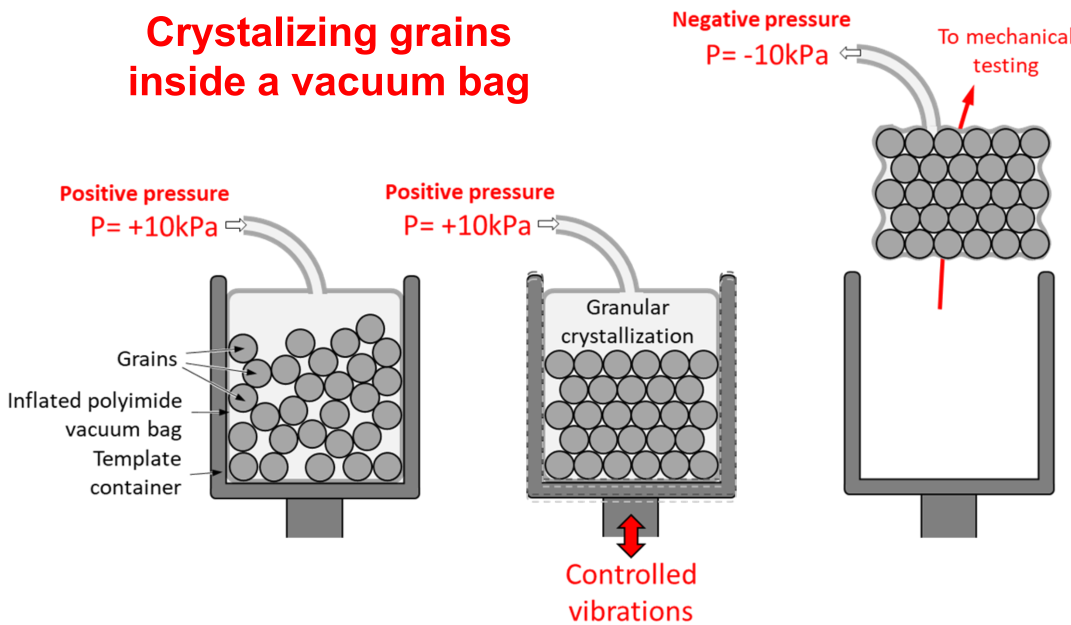
Volume of individual grains kept constant across all geometries (132 mm³)



Number of grains (N=800) and sample geometry identical for all samples



Crystalizing grains inside a vacuum bag



N. Karuriya and F. Barthelat "Granular crystals as strong and fully dense architectured materials" *PNAS* 120(1) (2023)

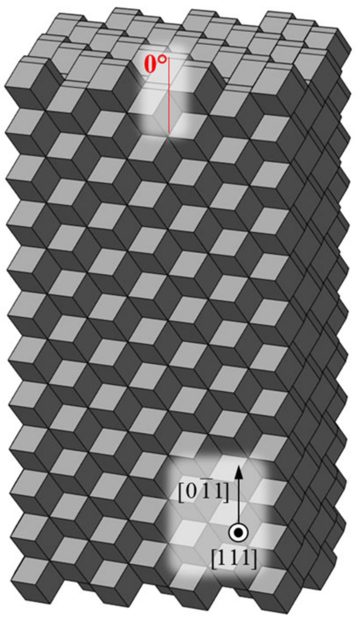
Crystallographic orientations

Vibration box as template for crystallization:

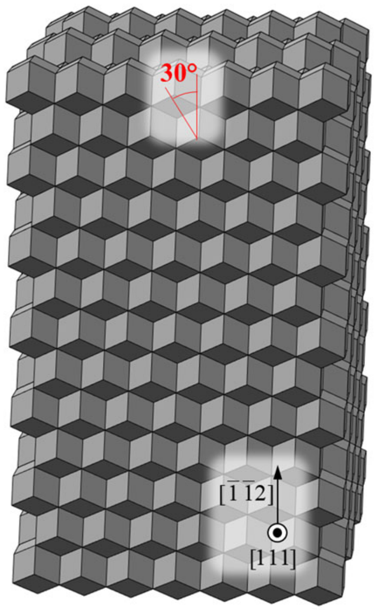
- **Planes of densest packing are templated by the flat floor of the box**
- **Rows of densest packing are templated by long walls of the box (on-axis samples), or by the short walls (in off-axis samples).**

FCC Crystal of Rhombic Dodecahedra (CRD)

On-axis (0°)

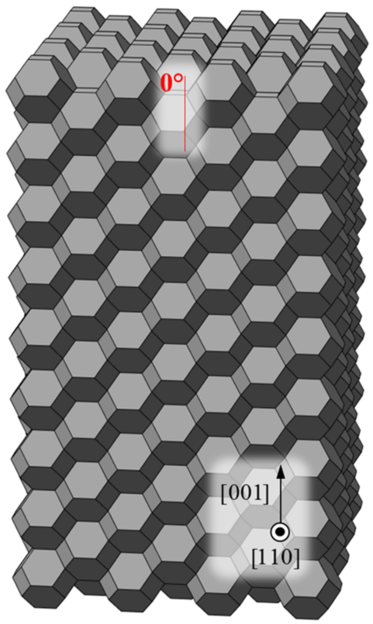


Off-axis (30°)

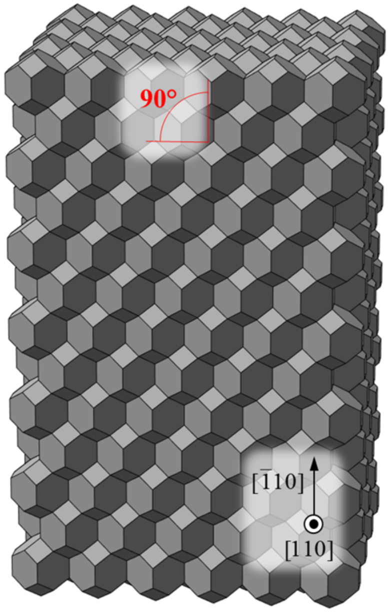


BCC Crystal of Truncated Octahedra (CTO)

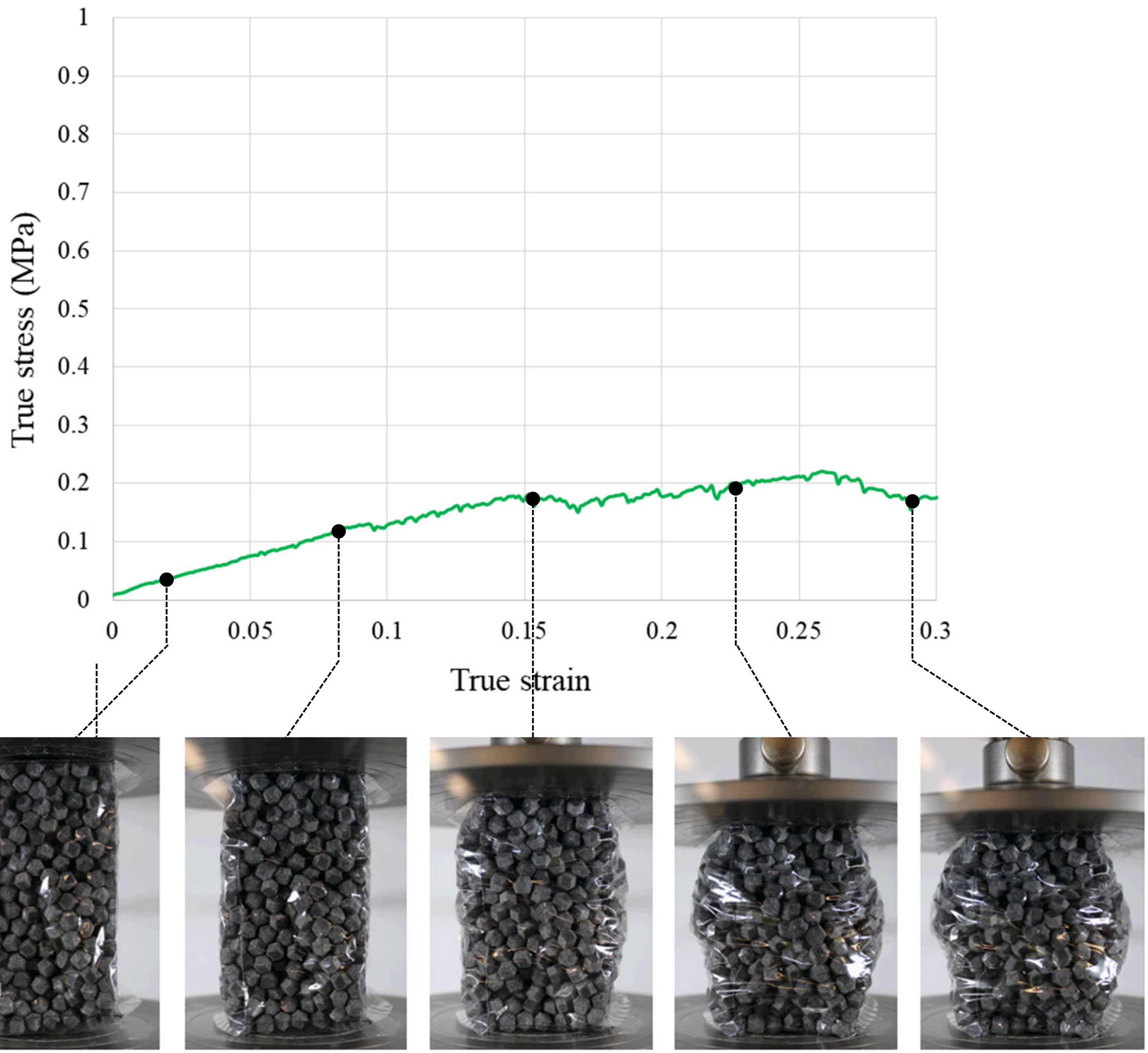
On-axis (0°)



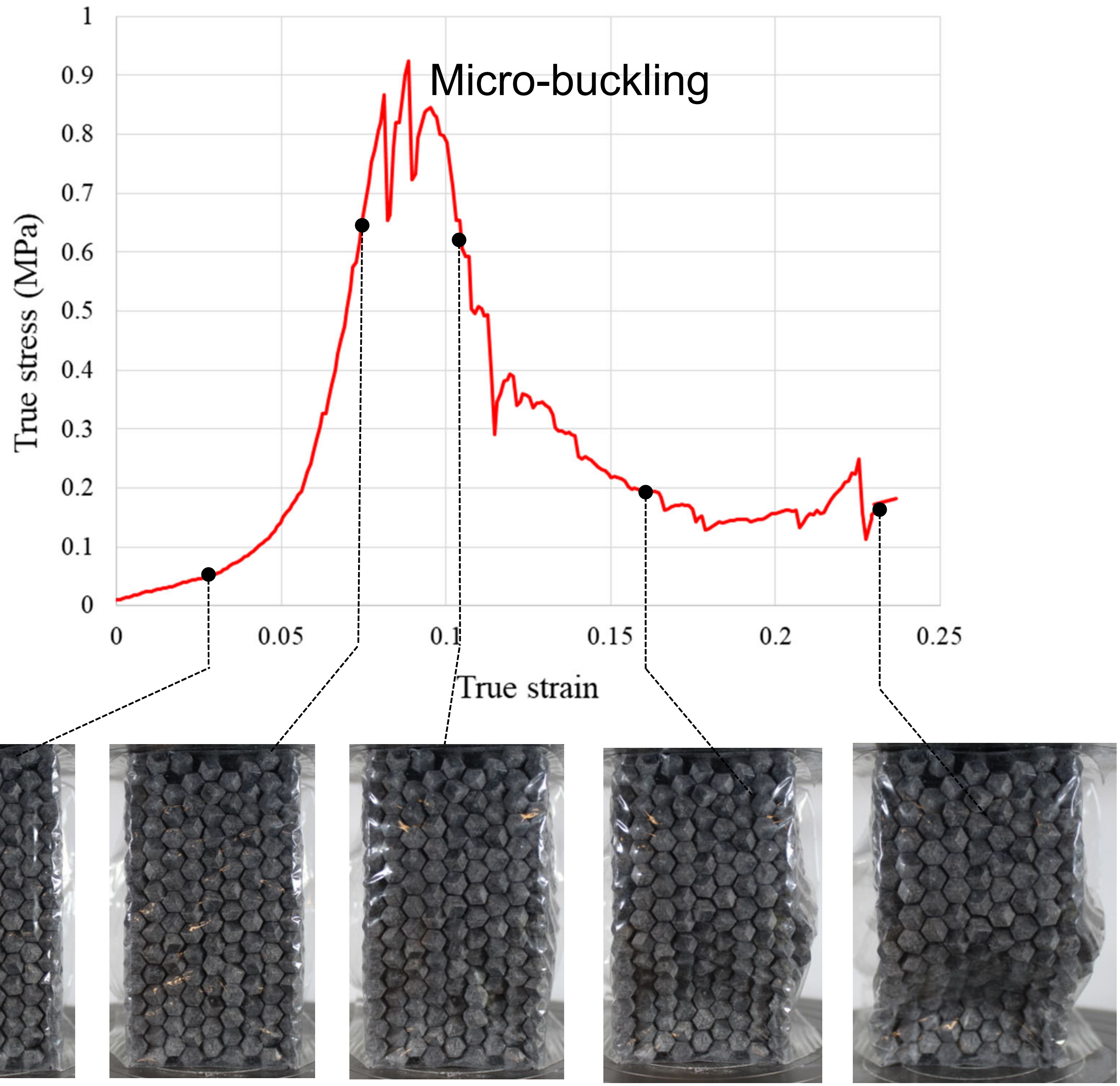
Off-axis (90°)



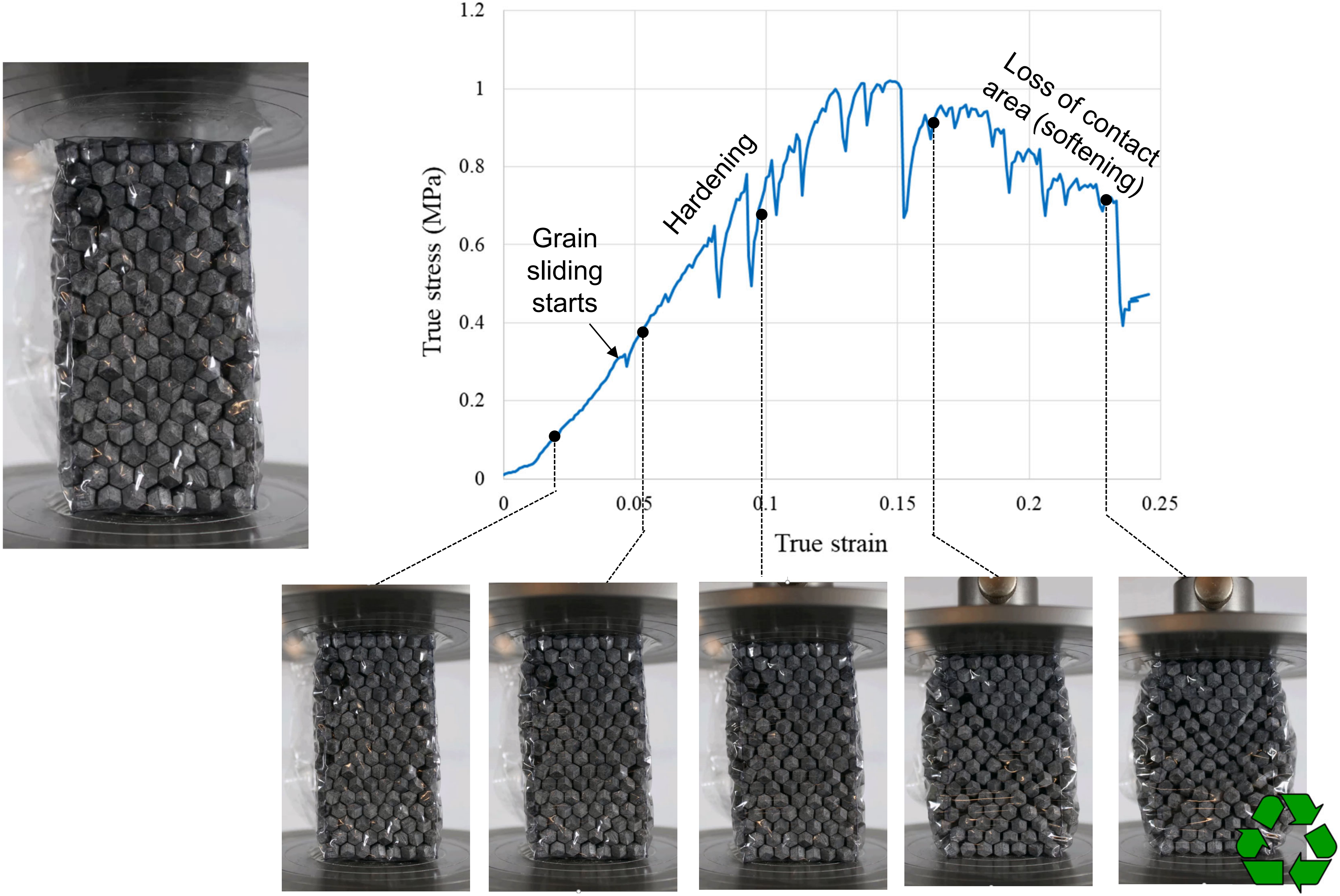
Randomly packed grains

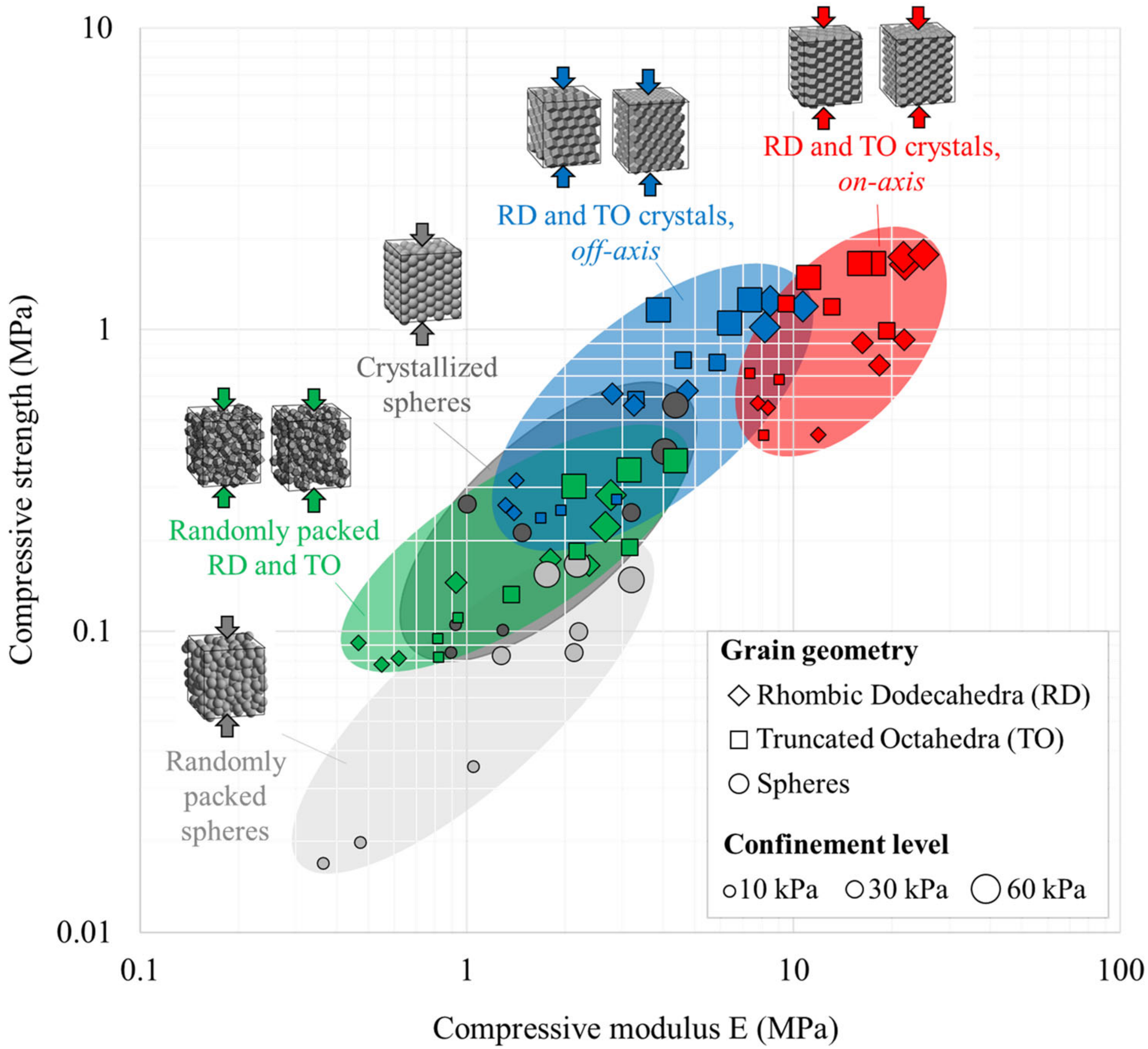


On-axis FCC crystals



Off-axis FCC crystals



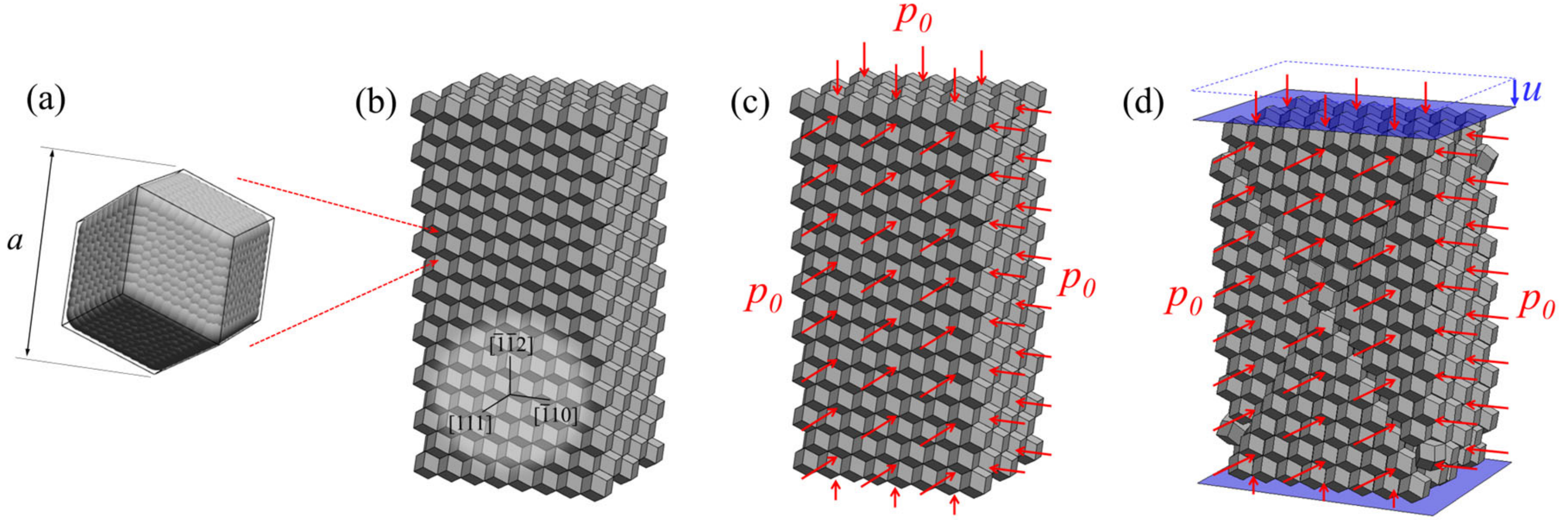


- **Strong correlation between strength and modulus**
- **Crystallized spheres are 2 to 4 times stronger than randomly packed spheres (same stiffness)**
- **RD and TO based granular crystals are 3 to 7 times stronger than their randomly packed counterparts**
- **Dense granular crystals are up to 25 times stronger than regular granular materials**

N. Karuriya and F. Barthelat "Granular crystals as strong and fully dense architectured materials" *PNAS* 120(1) (2023)

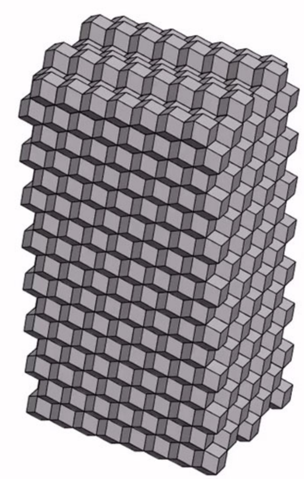
Discrete Element Models (DEM)

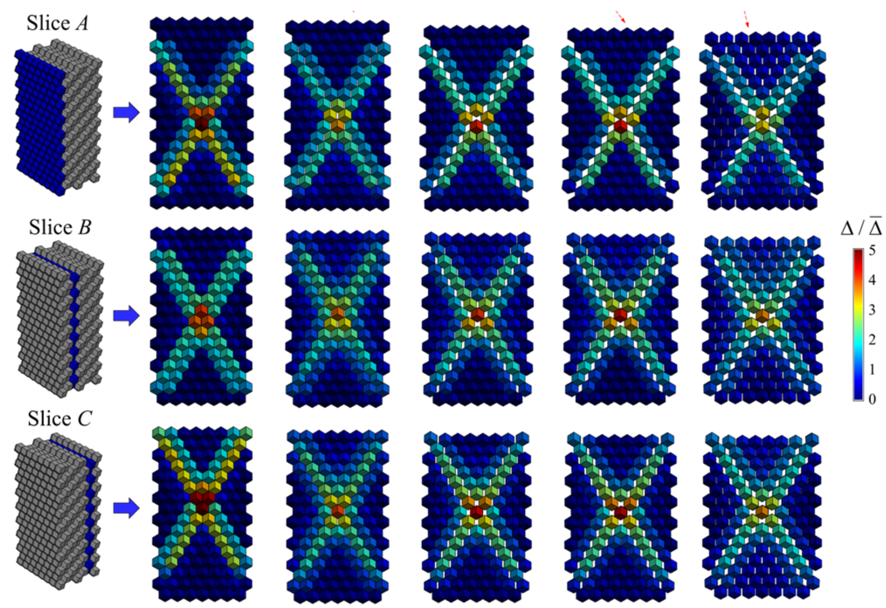
Overlapping spheres DEM model with LAMMPS



Experiment

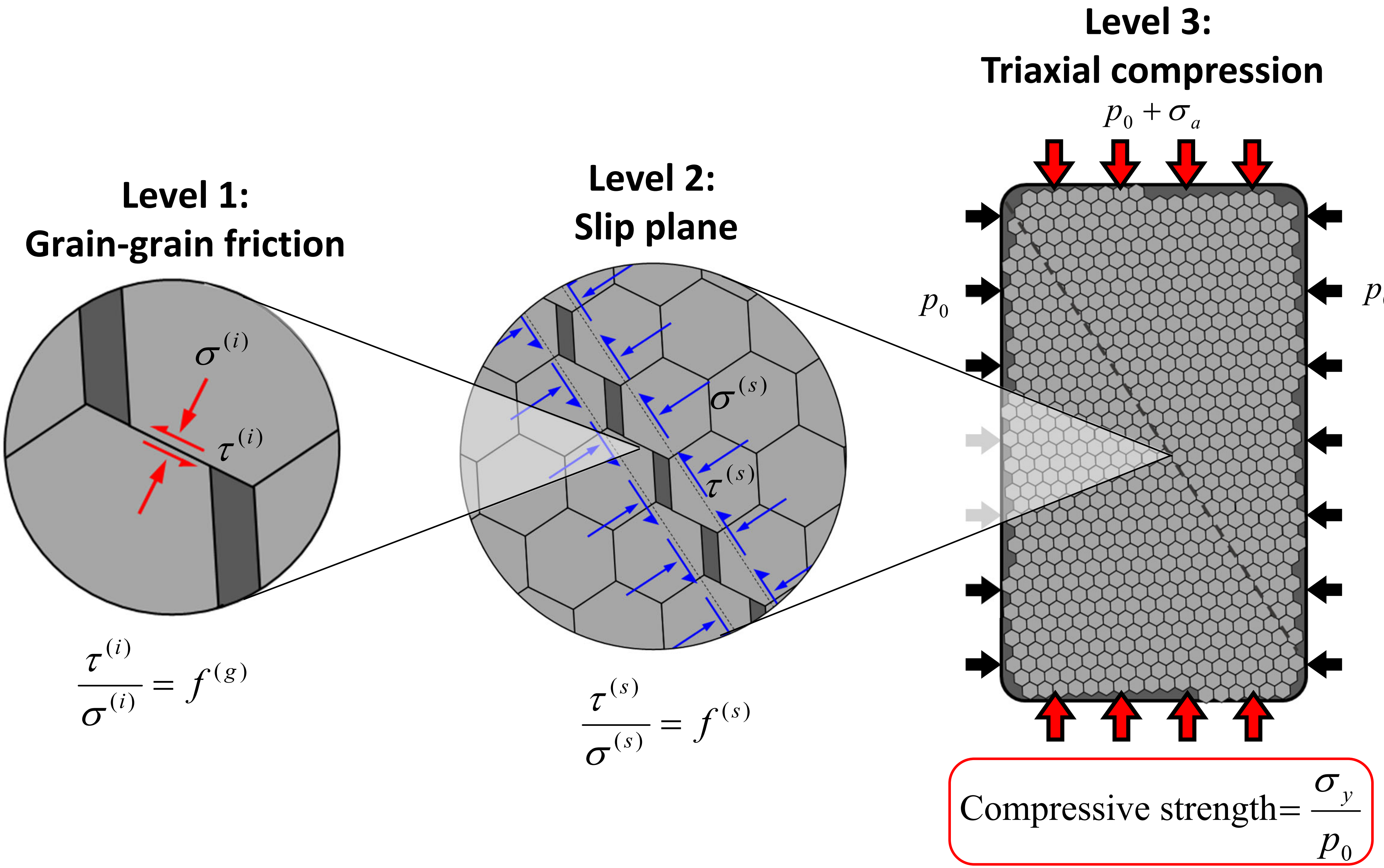
DEM model





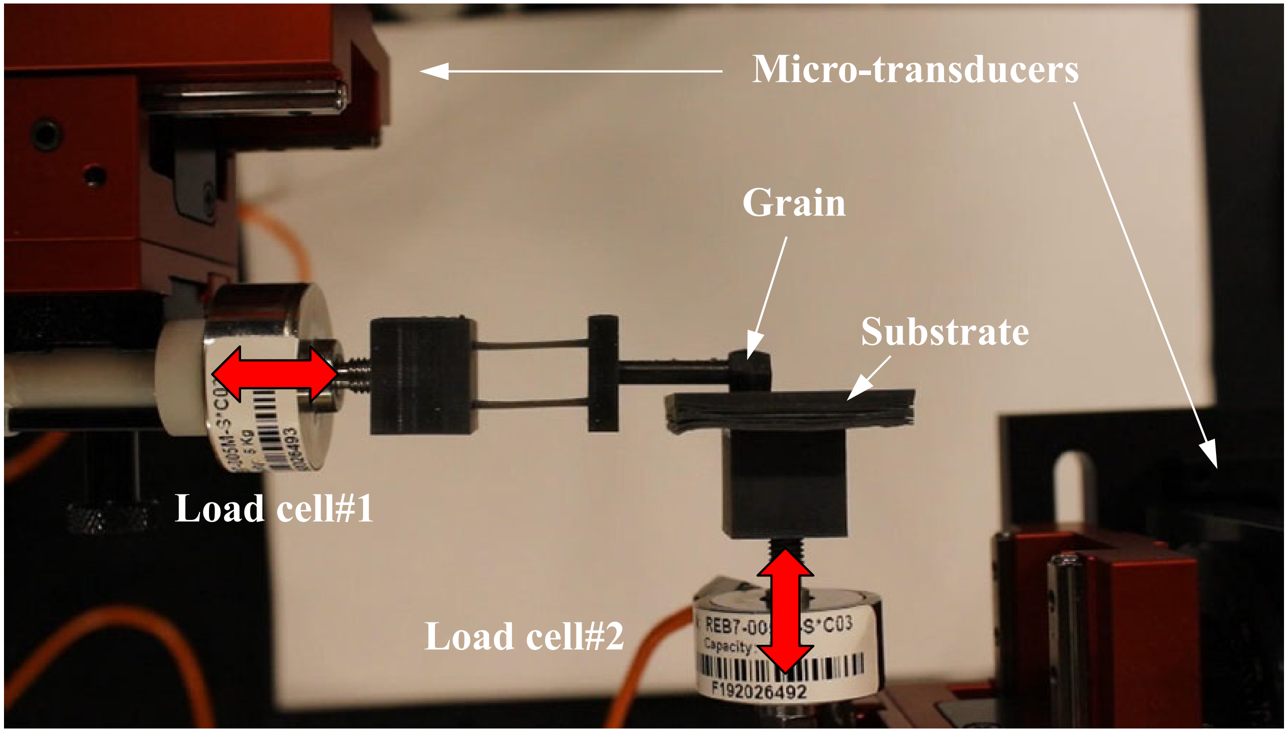
A.N.Karuriya and F. Barthelat, “Plastic deformations and strain hardening in fully dense granular crystals” (JMPS 186, 105597, 2024)

A "Granular crystal plasticity" model



N. Karuriya and F. Barthelat "Granular crystals as strong and fully dense architectured materials" *PNAS* 120(1) (2023)

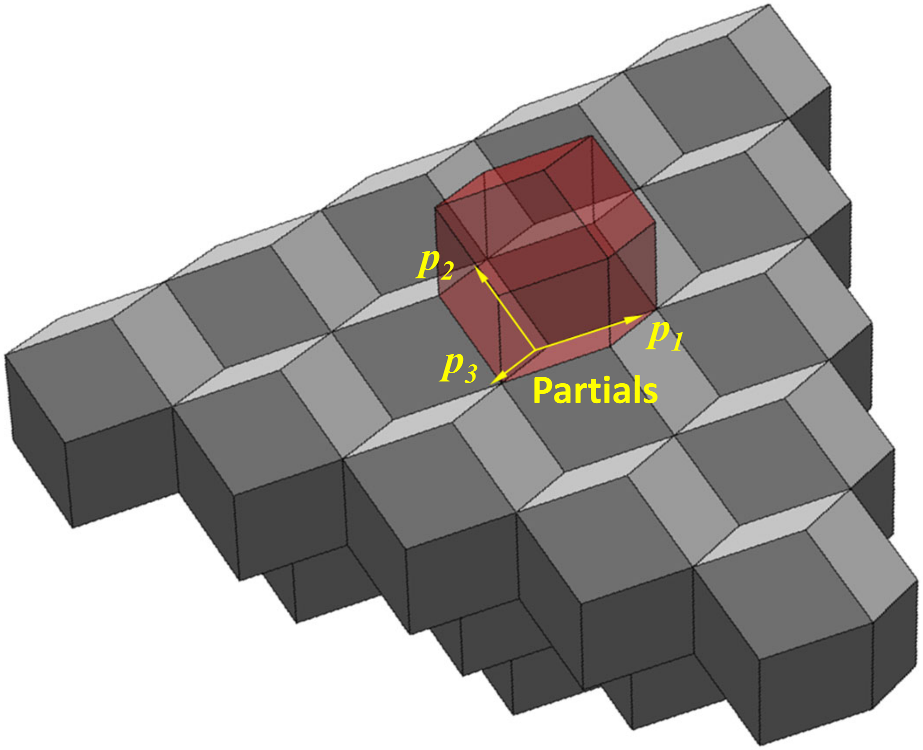
Level 1: Grain-on-grain friction



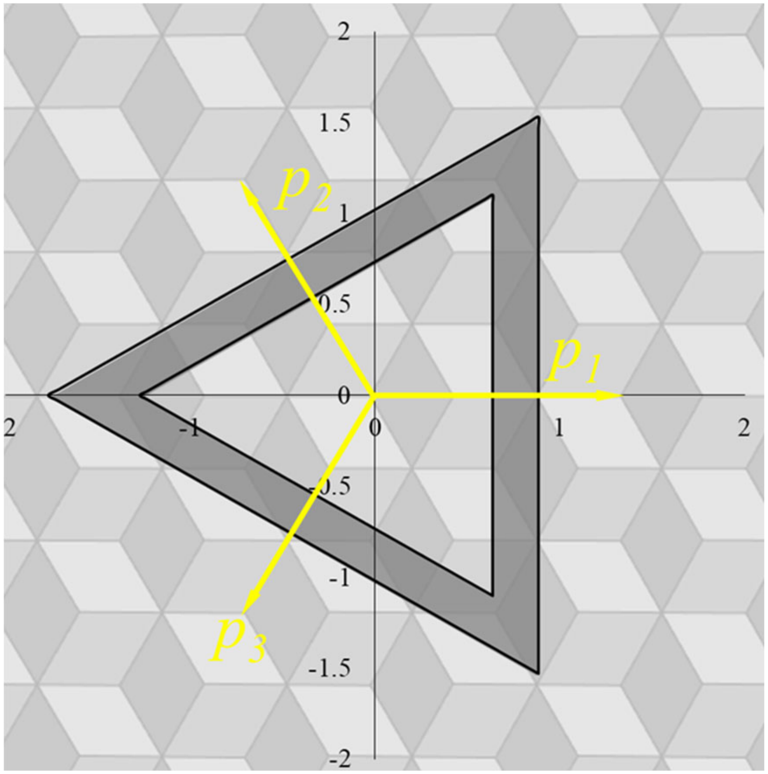
Measured coefficient of friction: $f^{(g)} = 0.30 \pm 0.05$

Level 2: Slip plane friction

Frictional grain motion over a (111) slip plane in FCC crystal

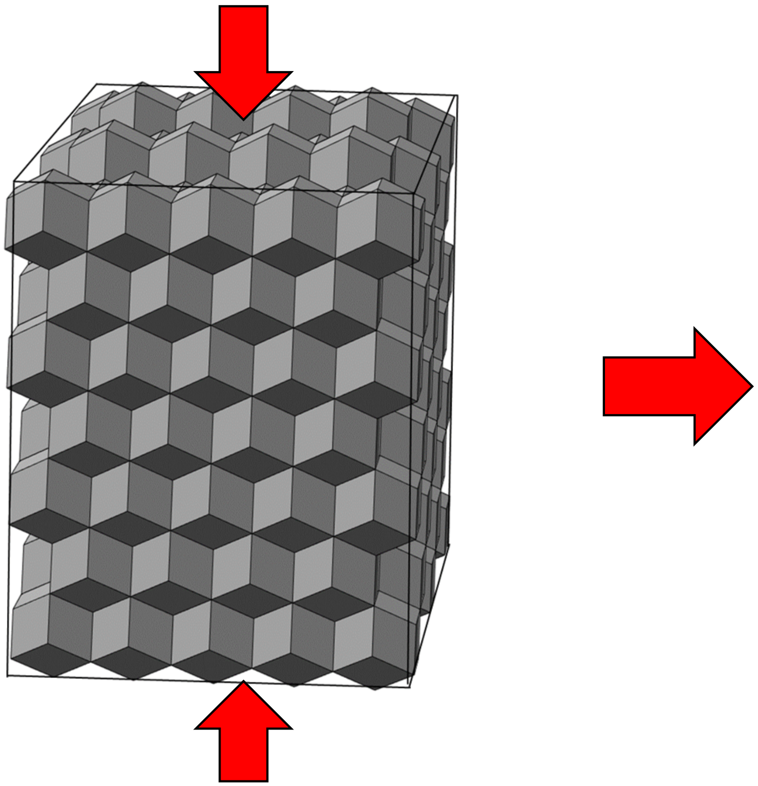


Range for friction coefficient of (111) slip plane with grain-to-grain friction 0.25< $f^{(g)}$ <0.35

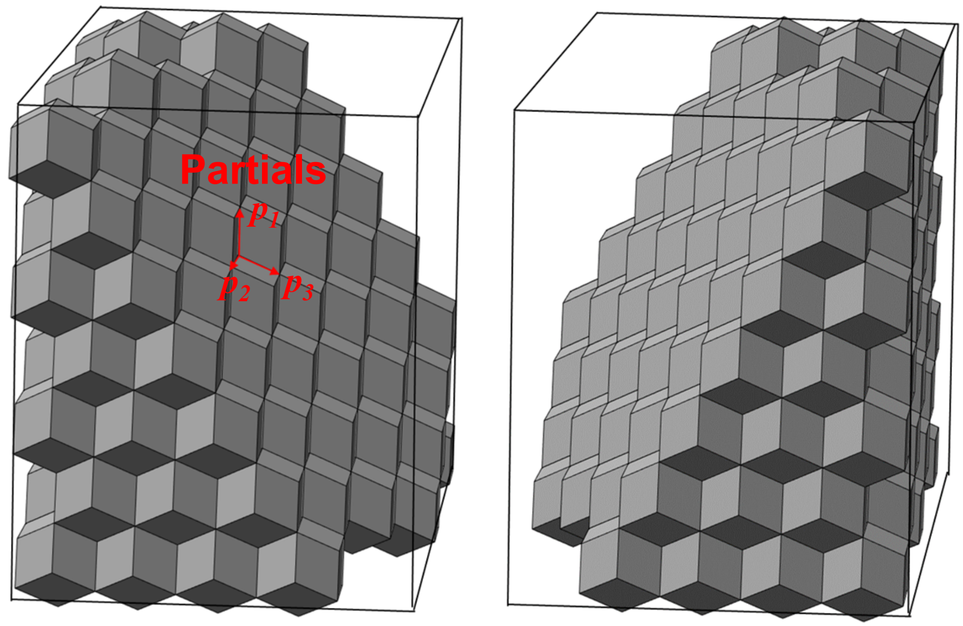


Level 3: Granular crystal plasticity

Off-axis loading direction

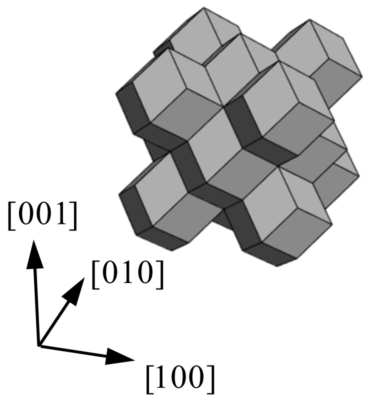


Example showing an activated (111) plane

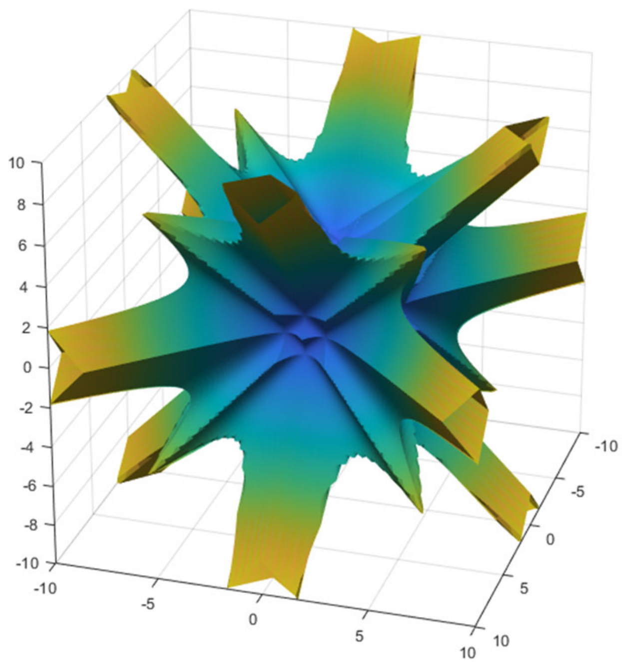


Plasticity in FCC granular crystal

FCC crystal of dodecahedra

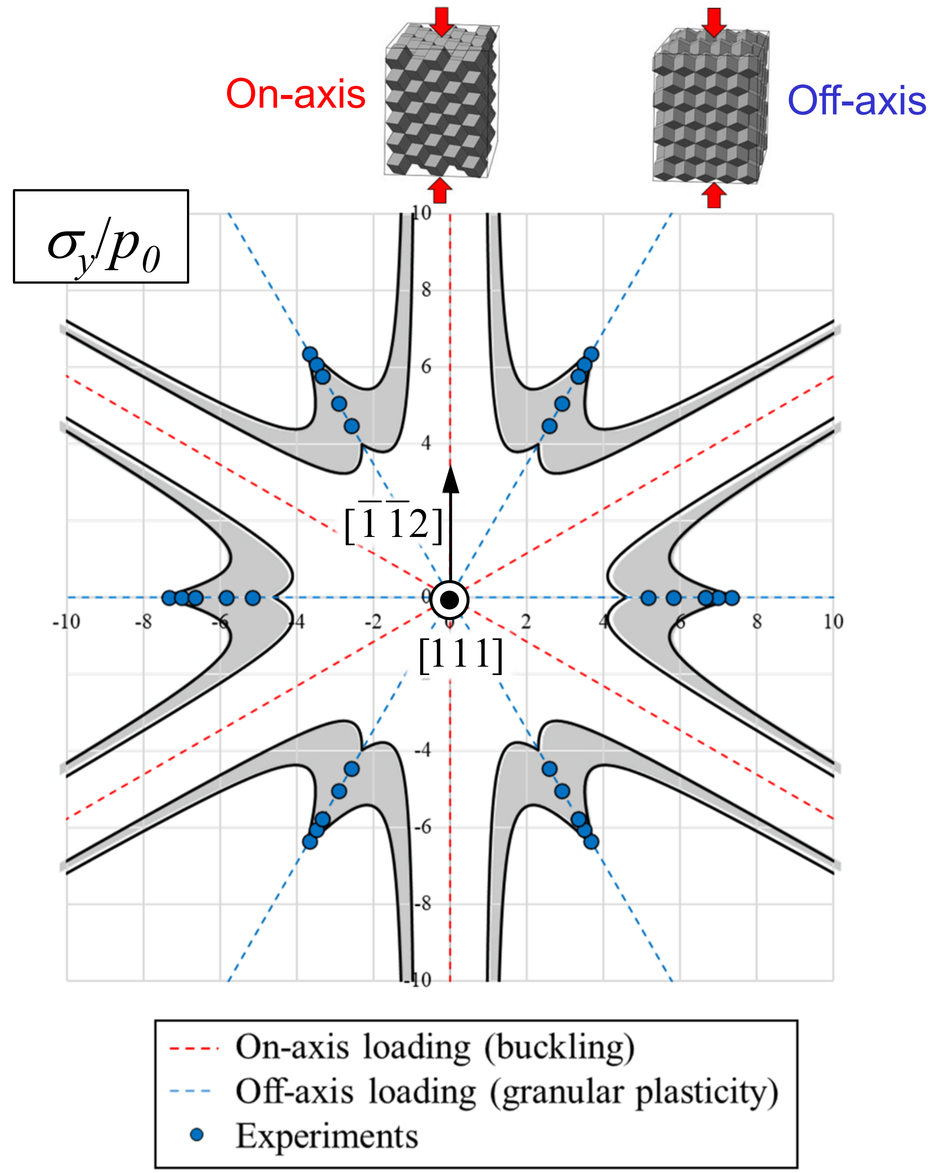


Anisotropic yield strength σ_y/p_0



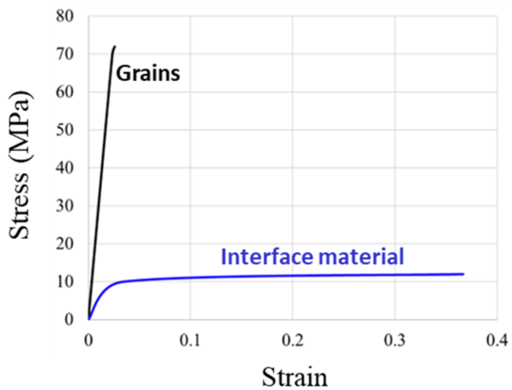
Intersection with (111) plane

Model range $0.25 < f^{(g)} < 0.35$

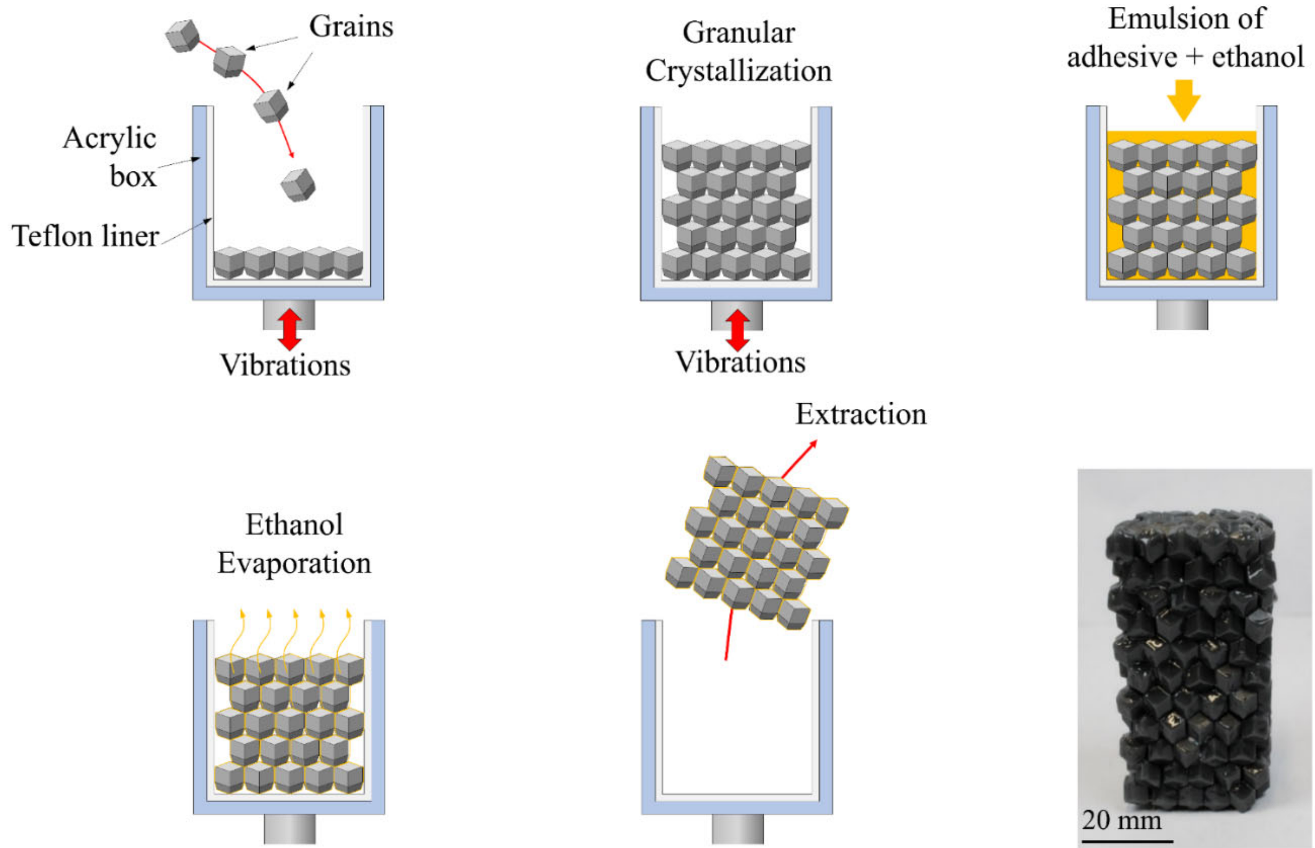


Free-standing cohesive granular crystals

Desired interface behavior:



Selection: a methacrylate-based pressure-sensitive adhesive (PSA)



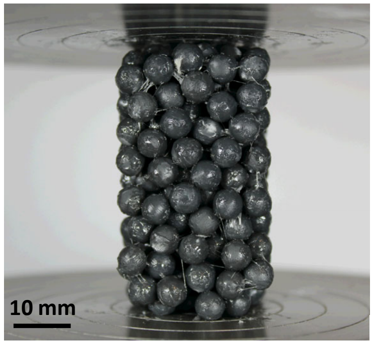
Navdeep Karuriya

Jeremy Simoes

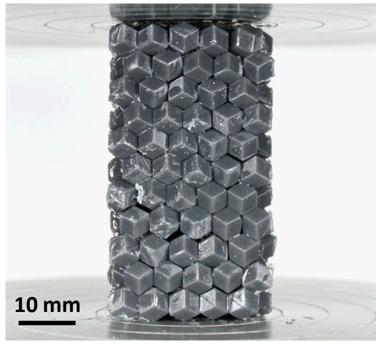
A.N.Karuriya, J. Simoes and F. Barthelat, "Fully dense and cohesive FCC granular crystals" Extreme Mechanics Letters 71, 102208 (2024)

Free-standing cohesive granular crystals

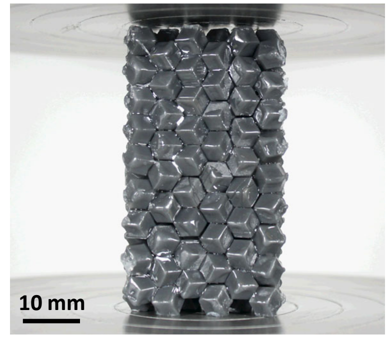
Randomly distributed spheres (RDS)



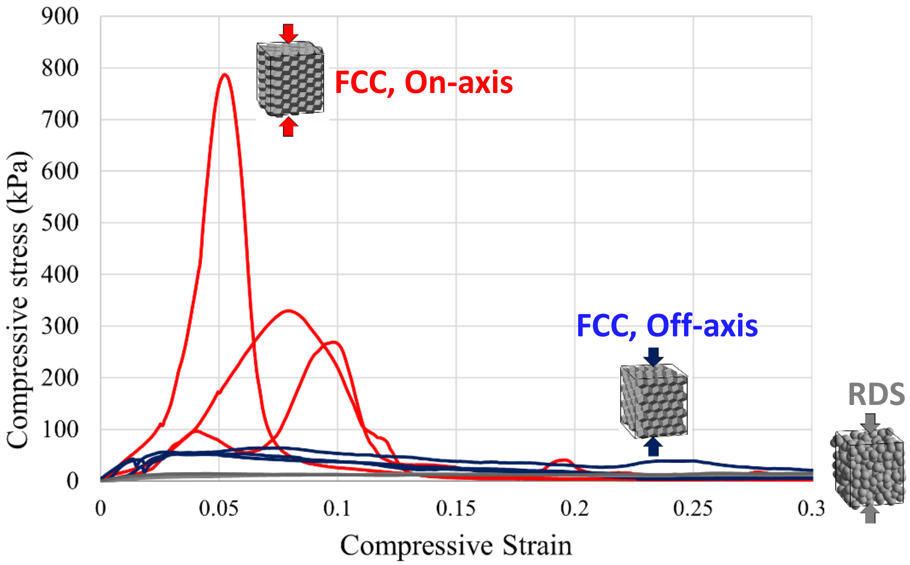
Off-axis FCC

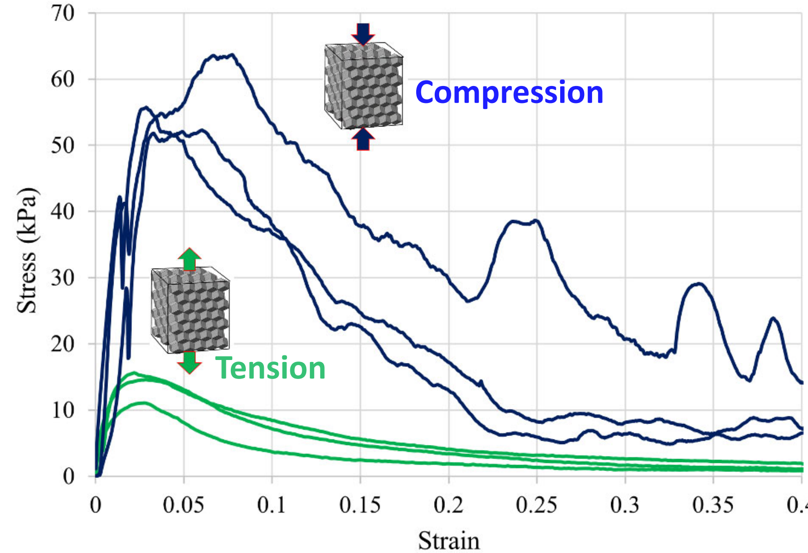


On-axis FCC



Off-axis FCC cohesive granular crystal



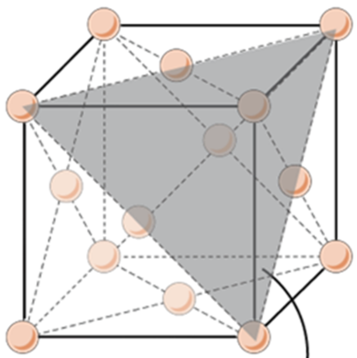


- **FCC crystal in compression: Slip along {111} or micro-buckling**
- **FCC crystal in compression: up to 60x stronger than randomly distributed spheres**

A.N.Karuriya, J. Simoes and F. Barthelat, "Fully dense and cohesive FCC granular crystals" Extreme Mechanics Letters 71, 102208 (2024)

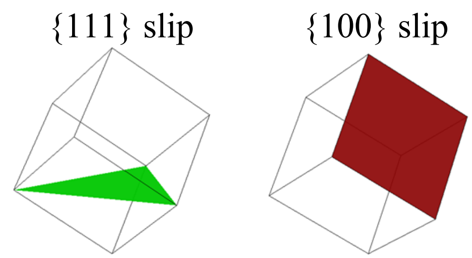
FCC vs. HCP in classical metallurgy

Face-centered cubic
(FCC) lattice

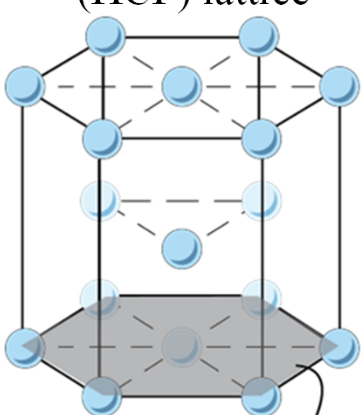


Close packing
plane: {111}

Classical metallurgy for FCC: Slip on {111} systems is the prominent deformation mechanism

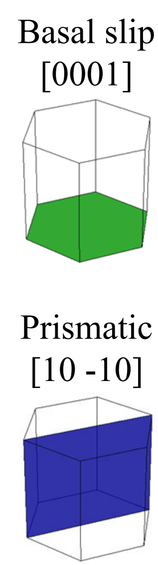


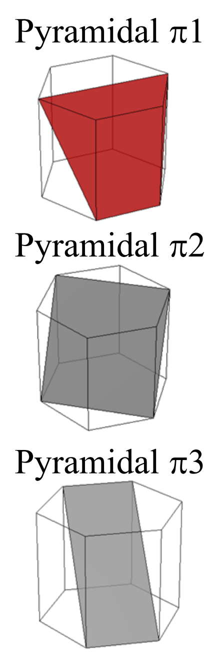
Hexagonal Close packed
(HCP) lattice



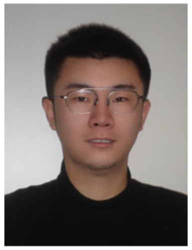
Close packing
plane:{0001}

Classical metallurgy for HCP: Less symmetries, more families of slip planes but fewer total available slip systems: higher yield strength.



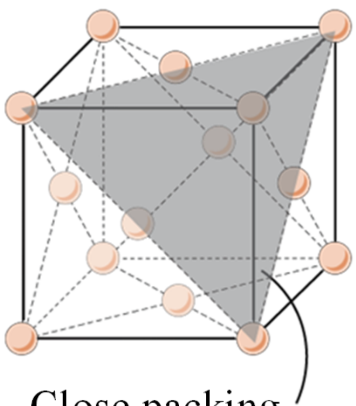


FCC vs. HCP granular crystals


Tian Gao

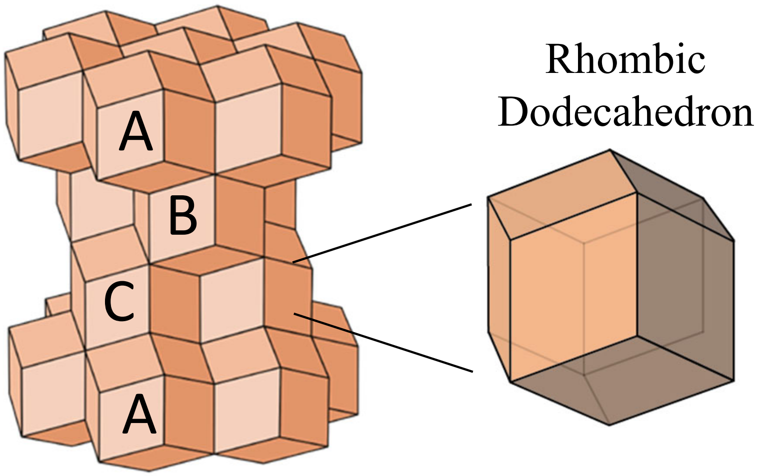

Navdeep Karuriya

FCC lattice



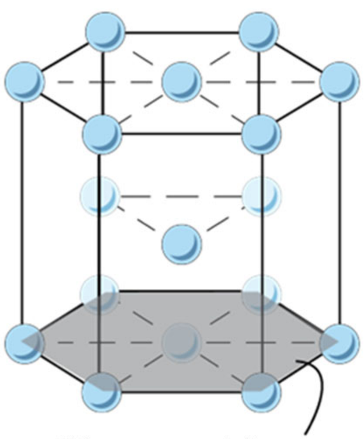
Close packing plane: {111}

Close packing oreintation <111>



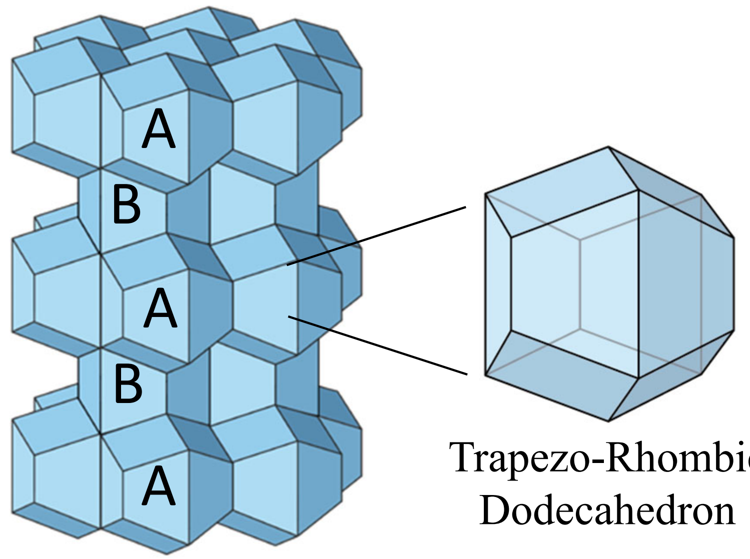
Rhombic Dodecahedron

HCP lattice



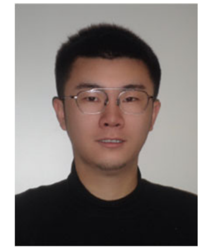
Close packing plane: {0001}

Close packing oreintation <0001>

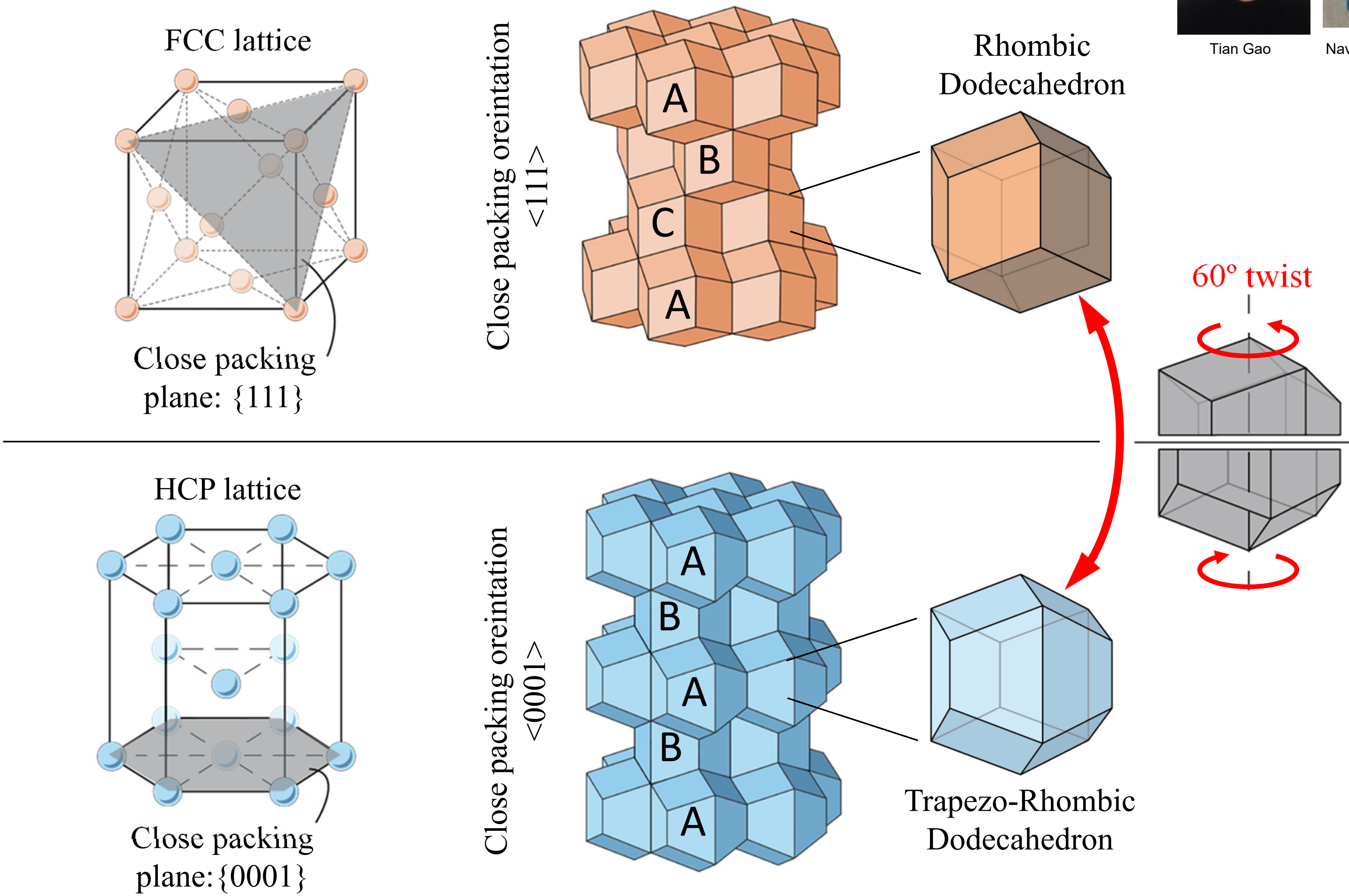


Trapezo-Rhombic Dodecahedron

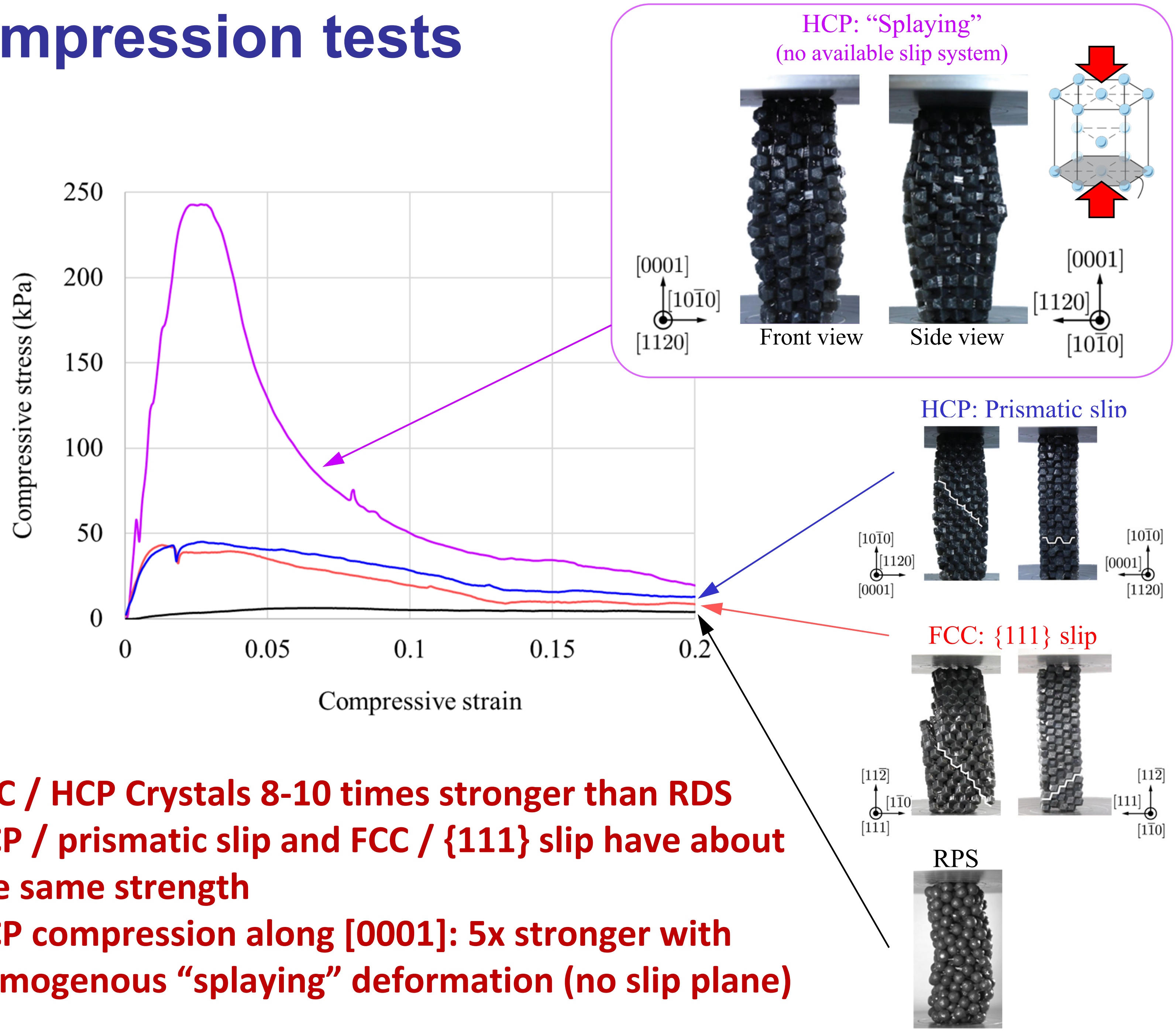
FCC vs. HCP granular crystals


Tian Gao


Navdeep Karuriya

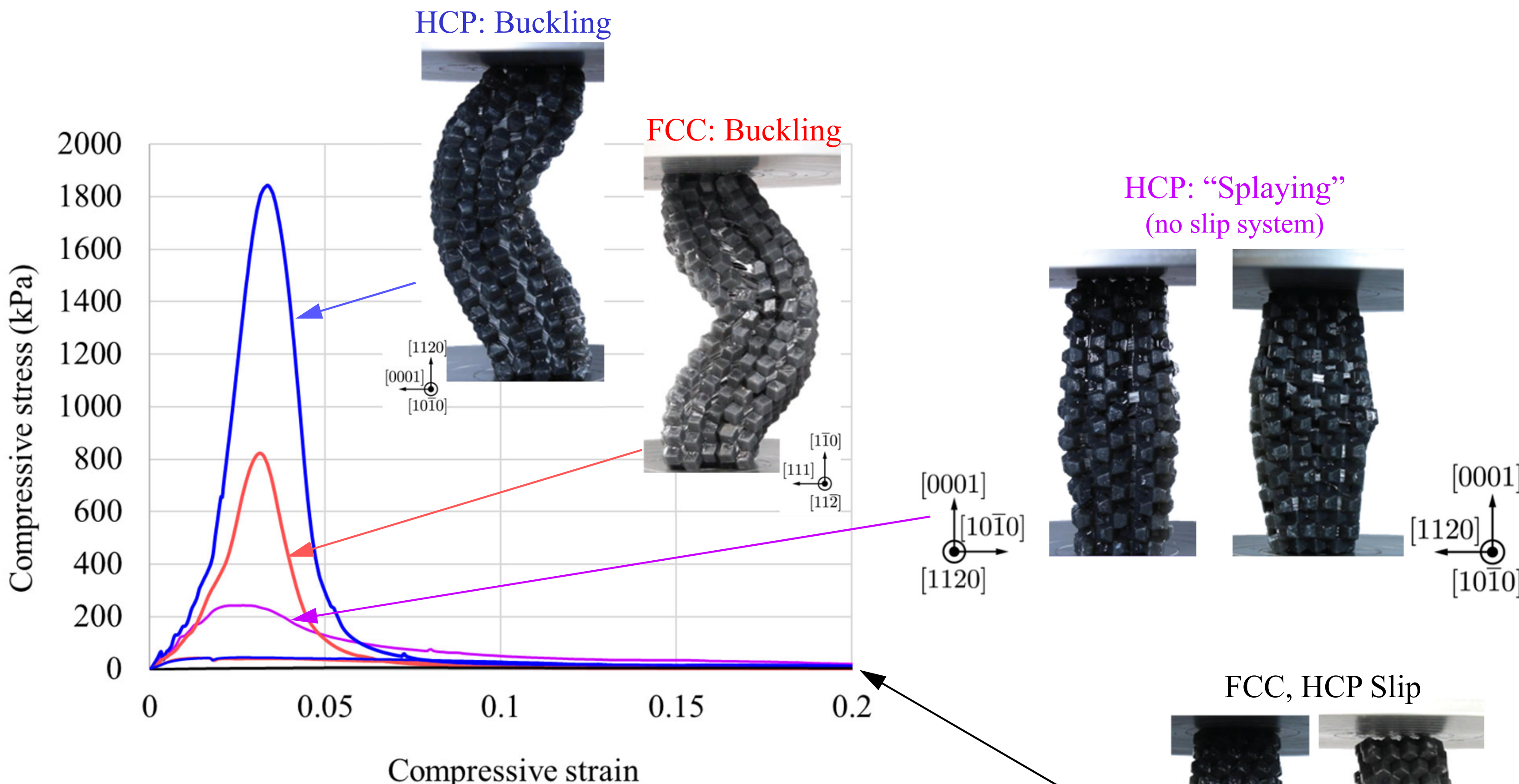


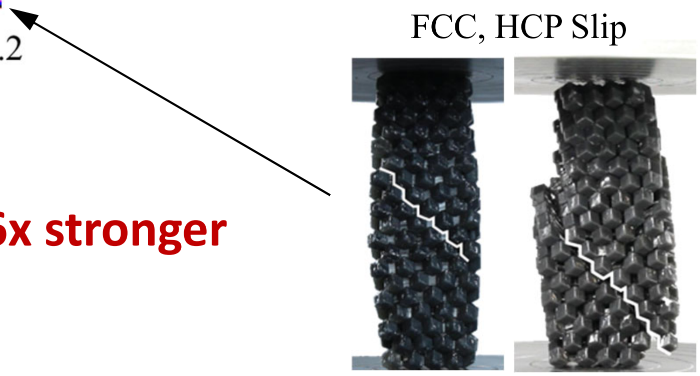
Compression tests



- **FCC / HCP Crystals 8-10 times stronger than RDS**
- **HCP / prismatic slip and FCC / {111} slip have about the same strength**
- **HCP compression along [0001]: 5x stronger with homogenous "splaying" deformation (no slip plane)**

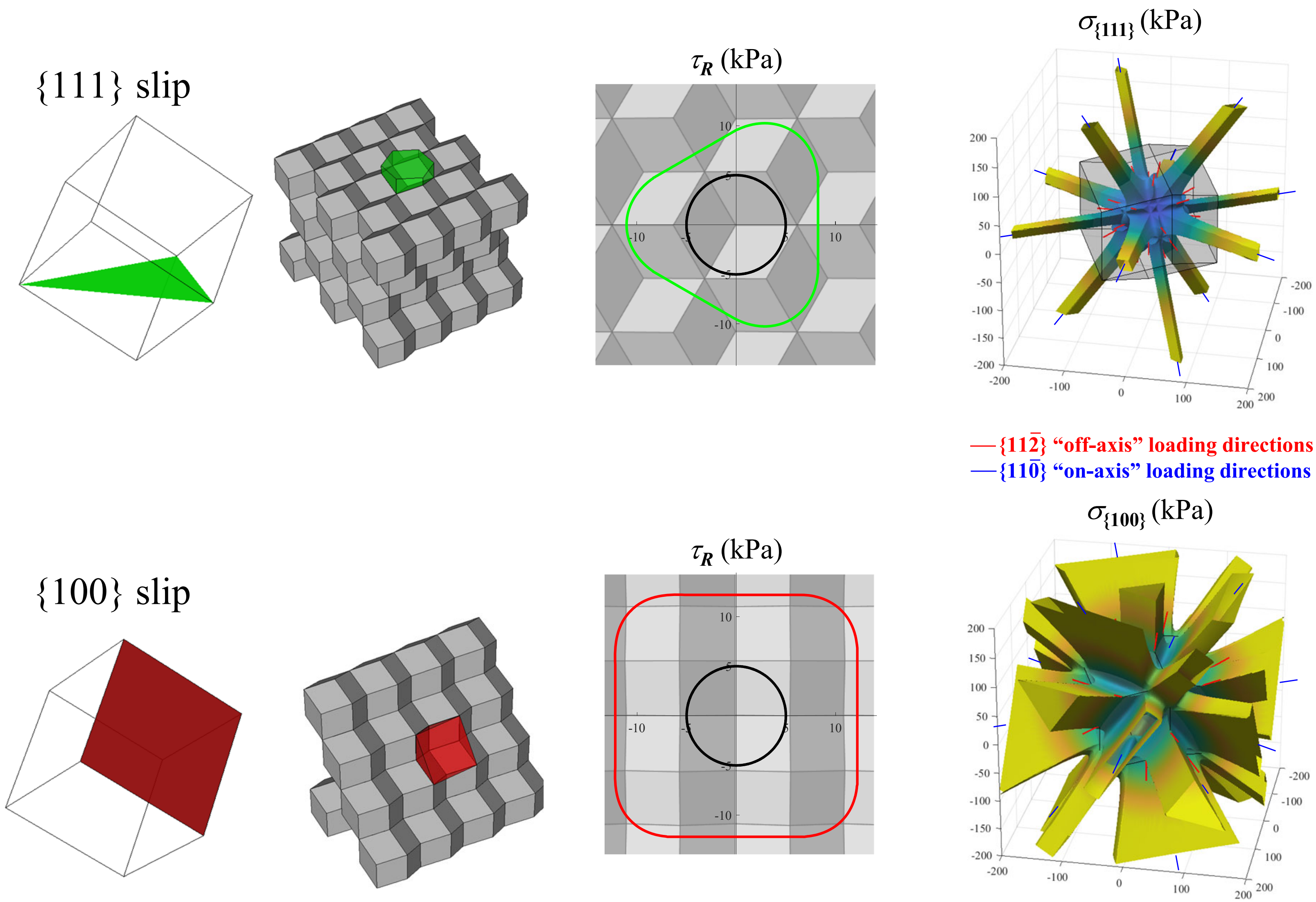
Compression tests





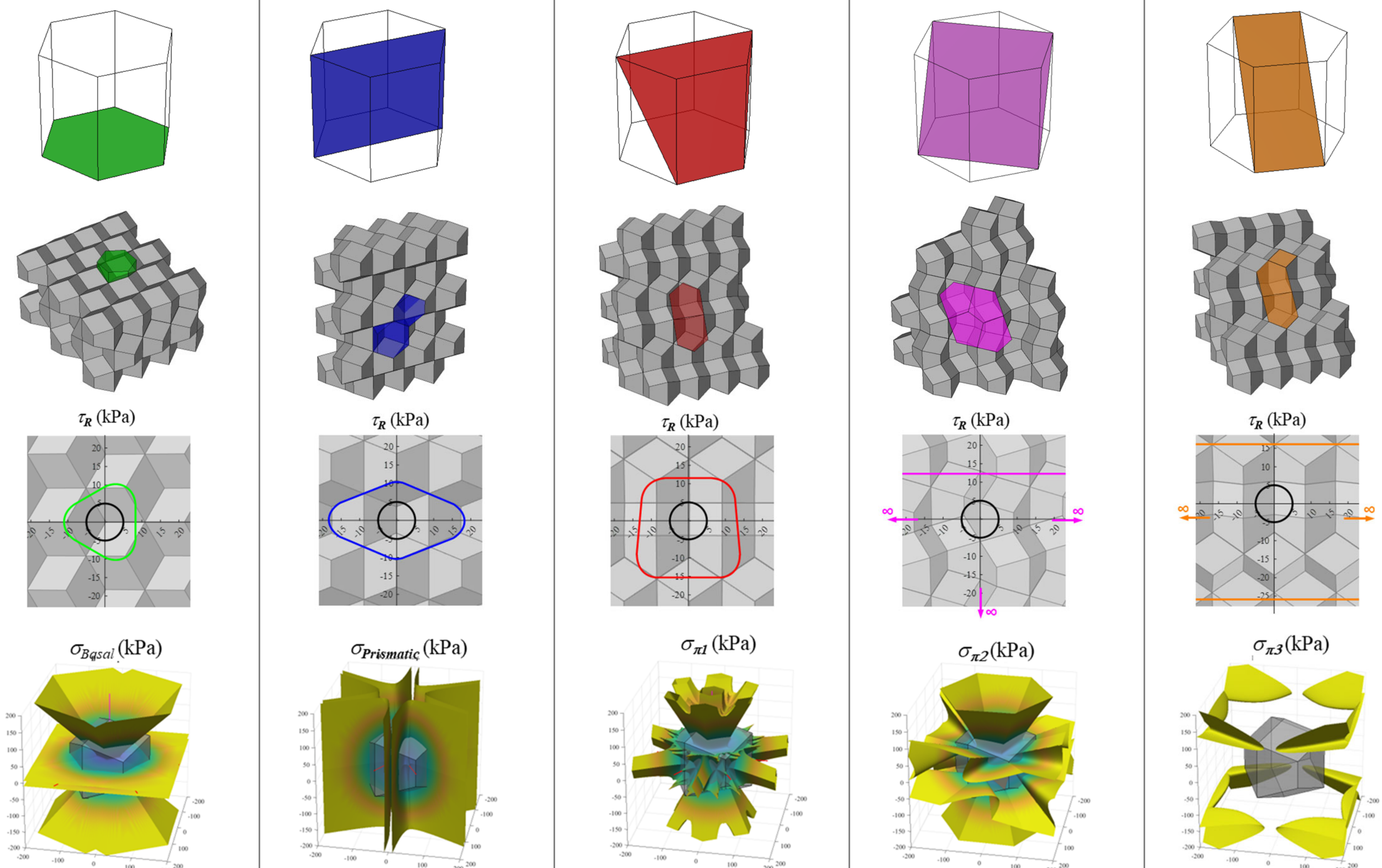
On-axis directions: FCC is 3x stronger, HCP is 6x stronger

FCC granular slip planes



HCP granular slip planes

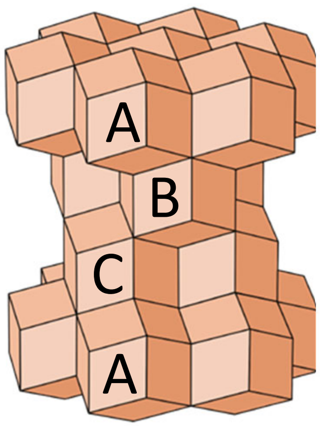
Basal slip [0001] | Prismatic $[10\bar{1}0]$ | Pyramidal $\pi 1$ | Pyramidal $\pi 2$ | Pyramidal $\pi 3$

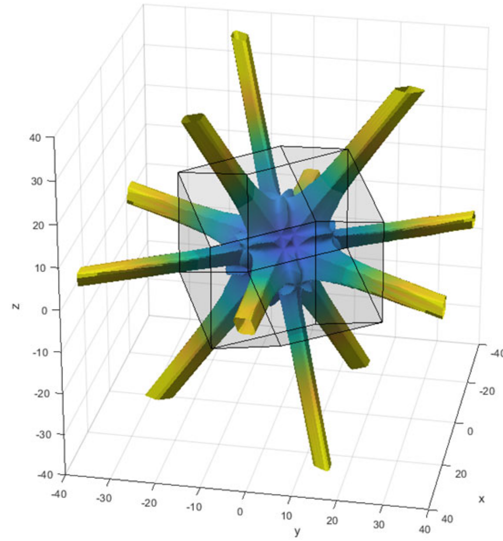


Basal slip [0001] | Prismatic $[10\bar{1}0]$ | Pyramidal $\pi 1$ | Pyramidal $\pi 2$ | Pyramidal $\pi 3$

Comparison FCC-HCP

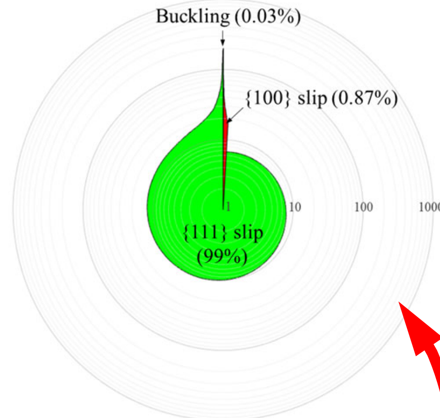
FCC

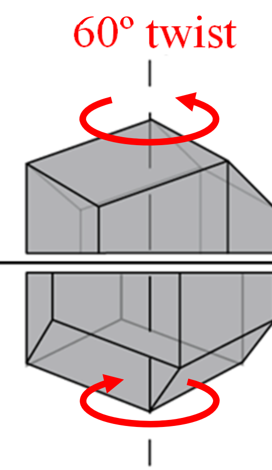




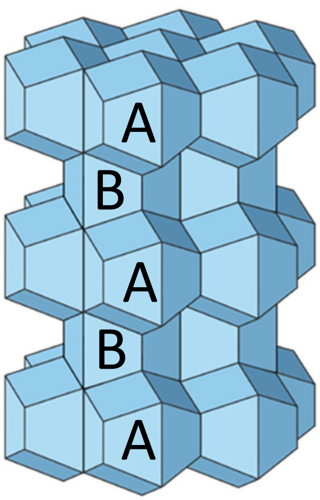
Average strength

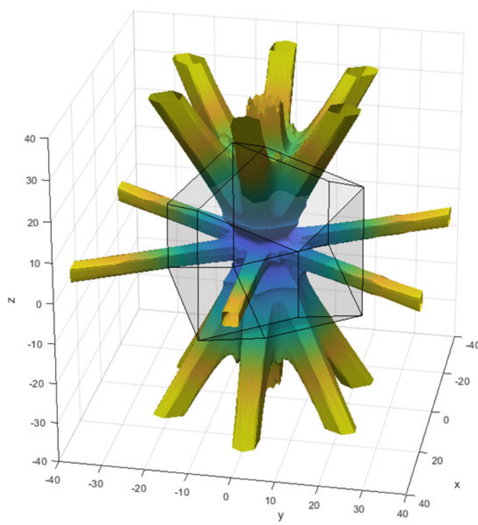
$$\langle \sigma_y \rangle / \tau_0 = 11.4$$





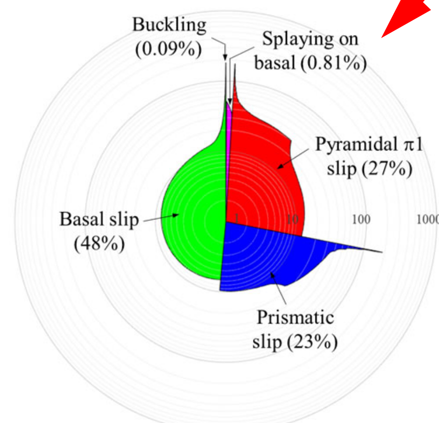
HCP





Average strength

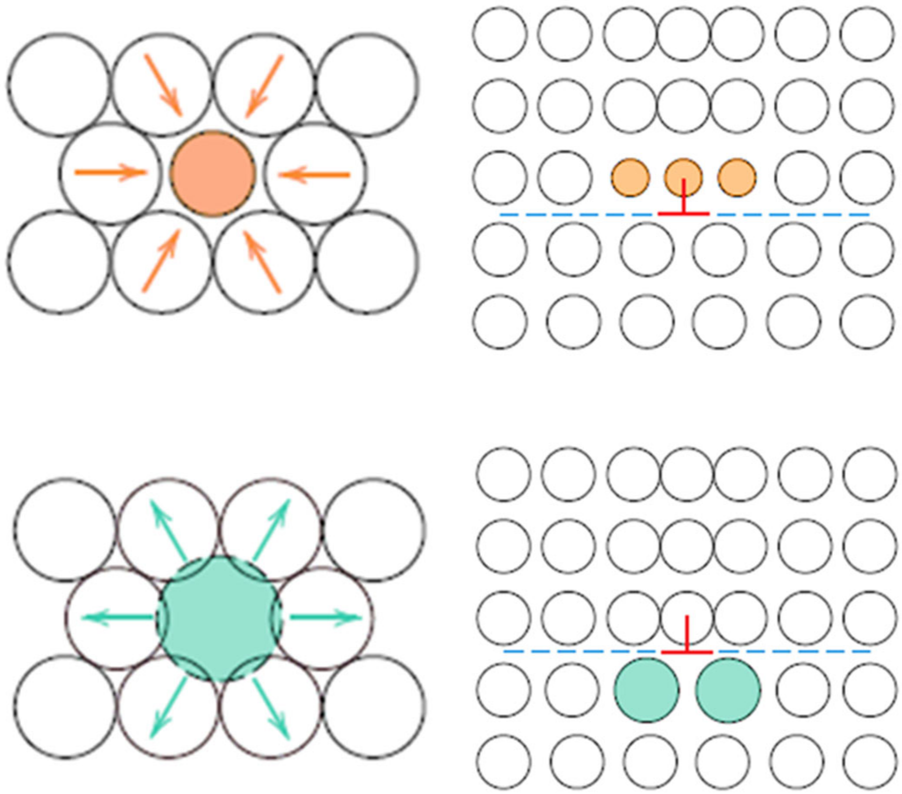
$$\langle \sigma_y \rangle / \tau_0 = 16.3$$



HCP = On average 50% stronger than FCC, broader variety of failure modes

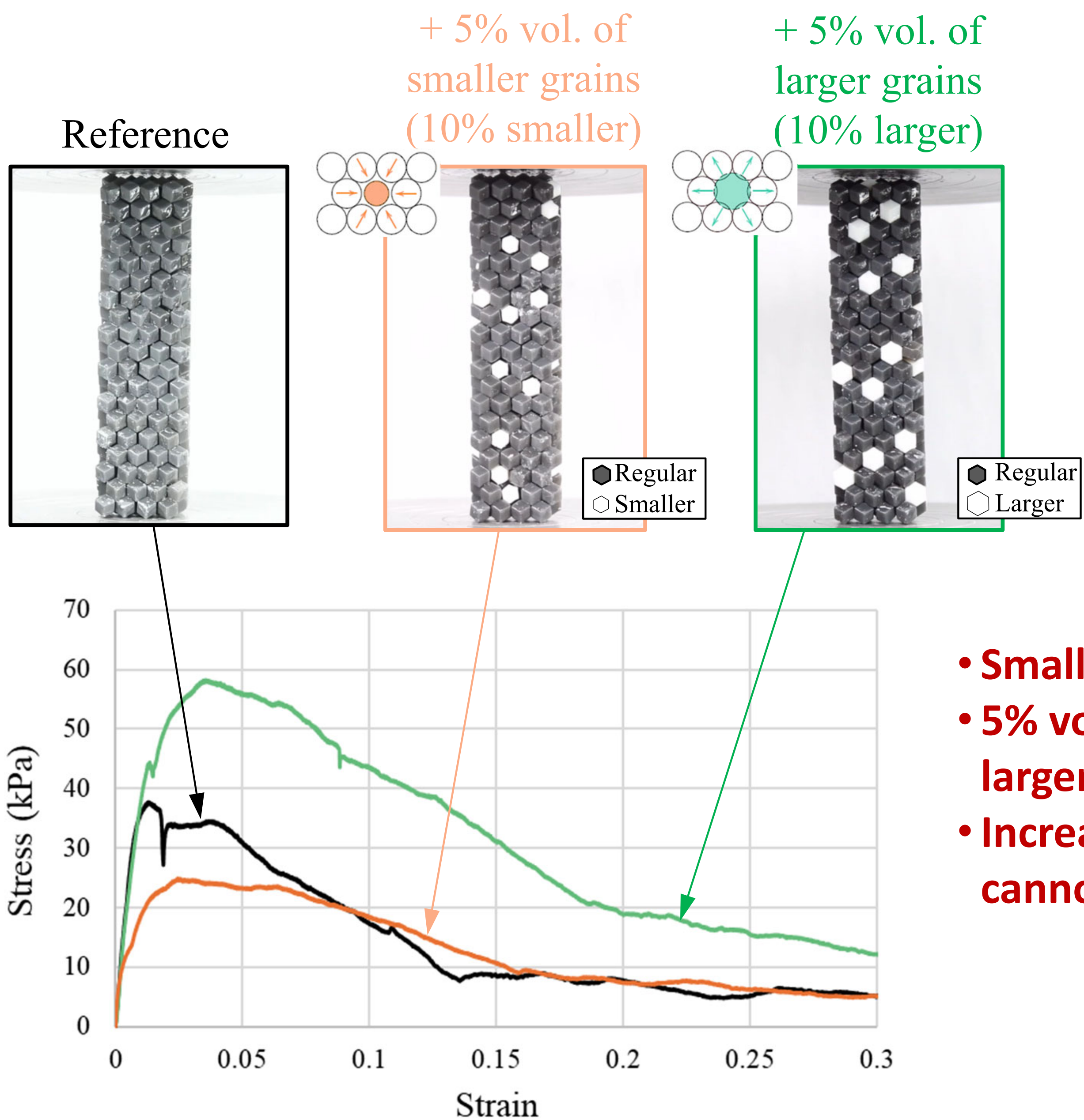
Solid solution strengthening in classical metallurgy

Substitution atoms create tensile or compressive lattice strains that pin dislocations

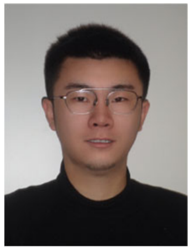


Callister, Materials Science and Engineering

Granular solid solution strengthening?

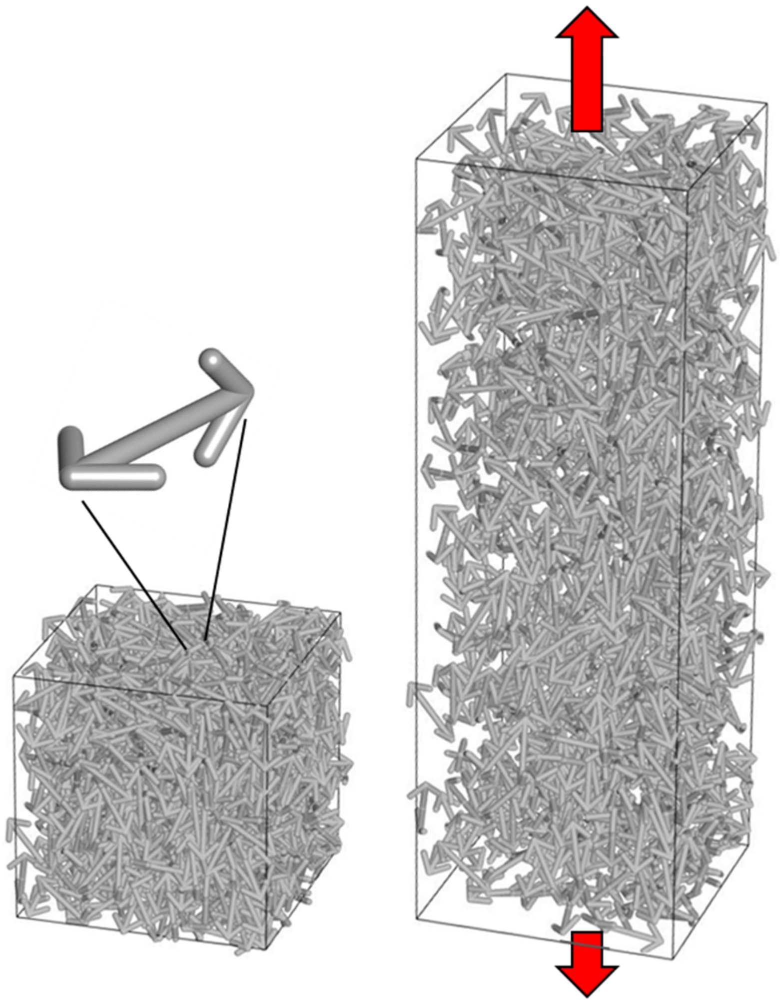



Navdeep Karuriya

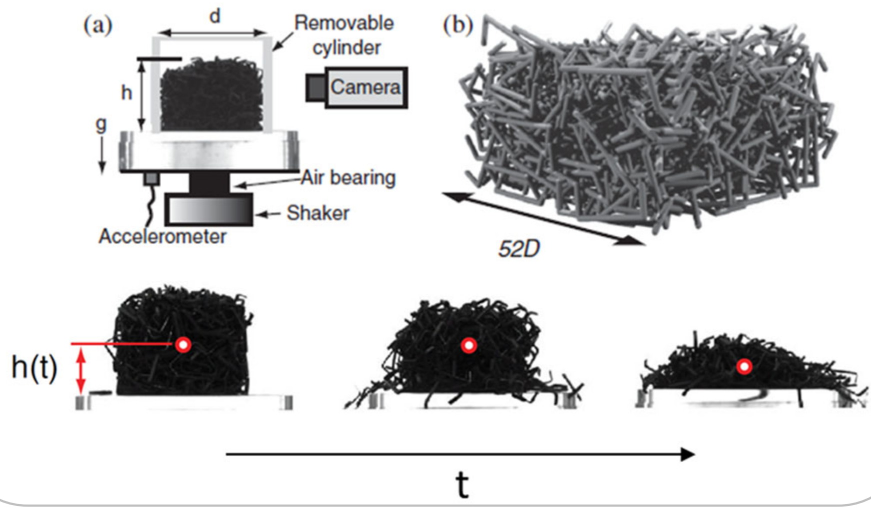

Tian Gao

- **Smaller grains: strength decreases**
- **5% vol. of larger grains (10% larger): strength increases by 50%**
- **Increase the size of larger grains: cannot assemble crystal**

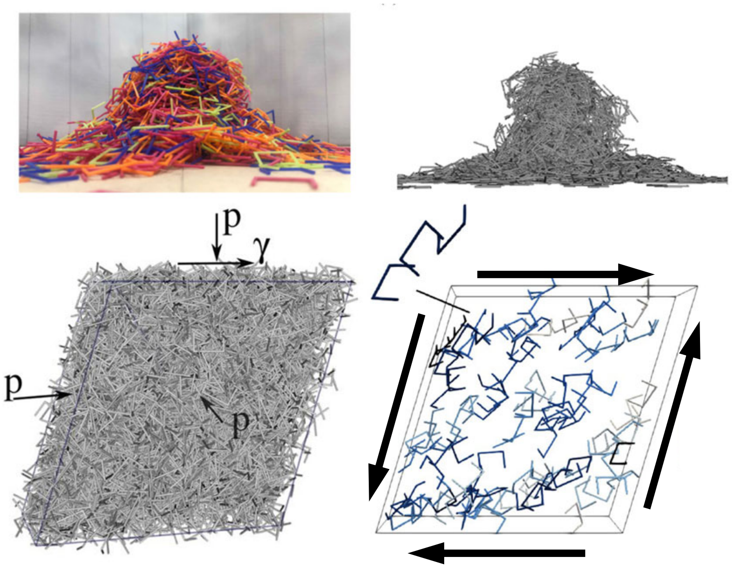
Entangled granular materials with tensile strength



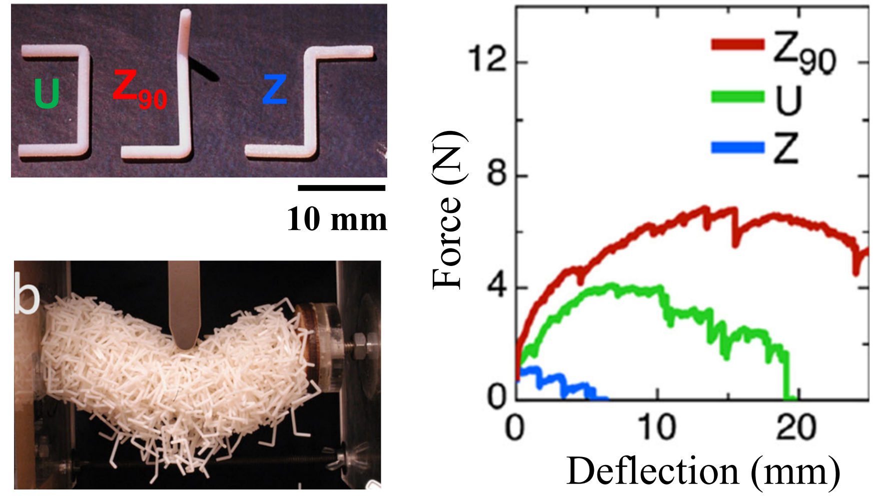
Free standing columns of entangled staples
(Gravish, Franklin, Hu and Goldman *PRL* 2012)



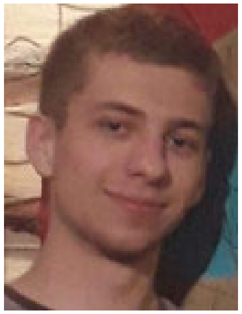
Stress transmission in entangled granular structures
(Karapiperis, Monfared, Buarque de Macedo, Richardson and Andrade, *Granular Matter* 2022)



Strong bundles of "Z shape" particles
(Murphy, Jaeger, *Granular Matter* 2016, Murphy, Roth, Peterman, Jaeger, Architectural Design 2017)

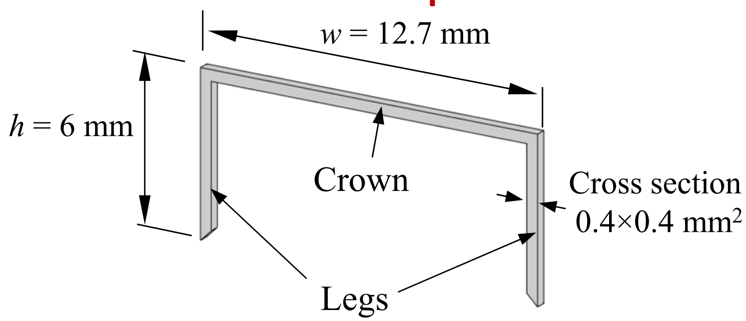


Our first model material: Staples

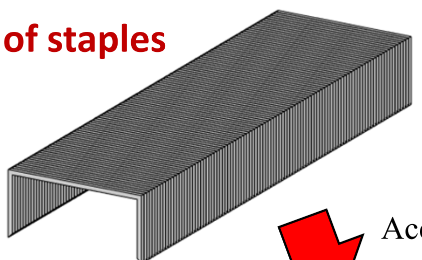


Vivien Fouquet

Standard steel office staple

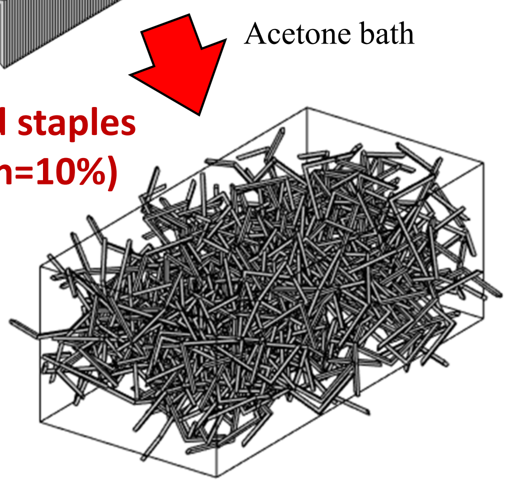


A stick of staples

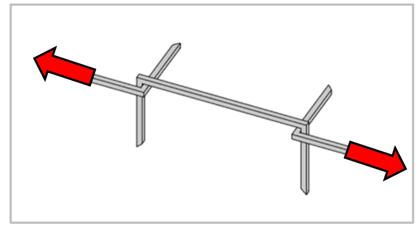


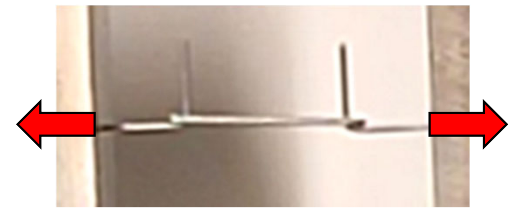
Acetone bath

Randomly distributed staples (typical solid fraction=10%)

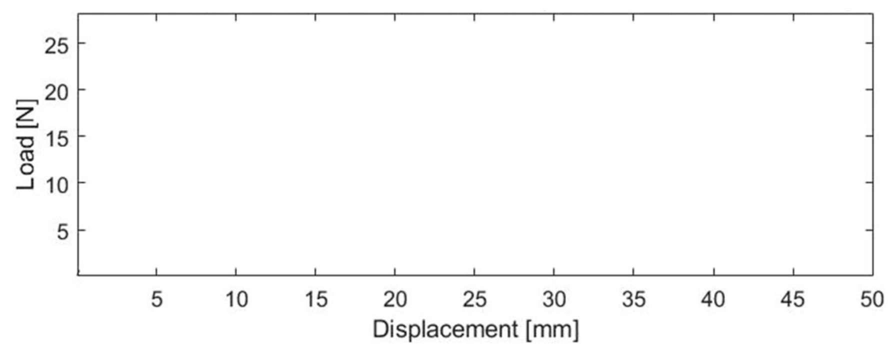


Staple pair strength test:Strength = 9±4 N
Failure occurs by elastic deformation + slipping or rotation and then unstable snap

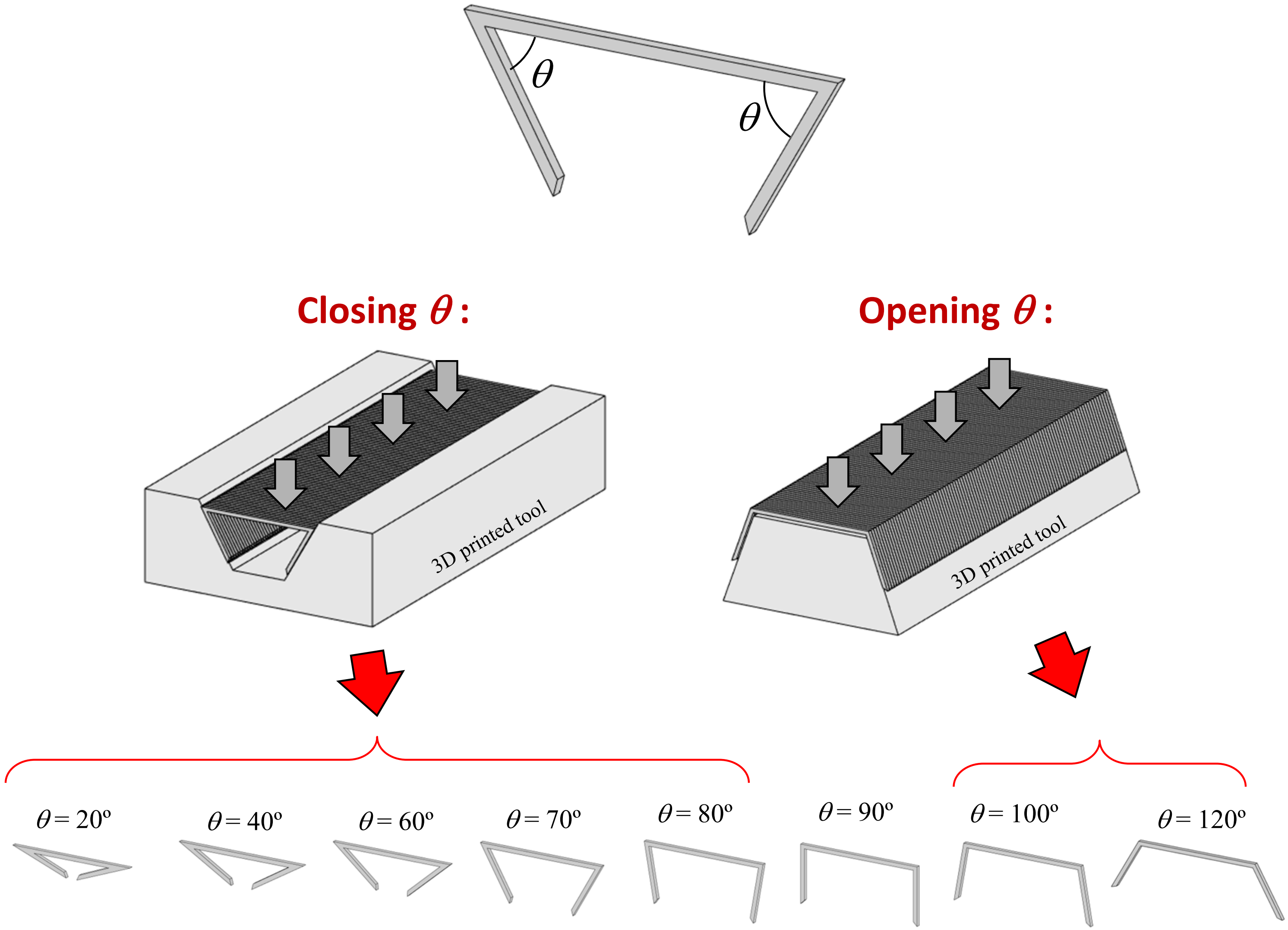




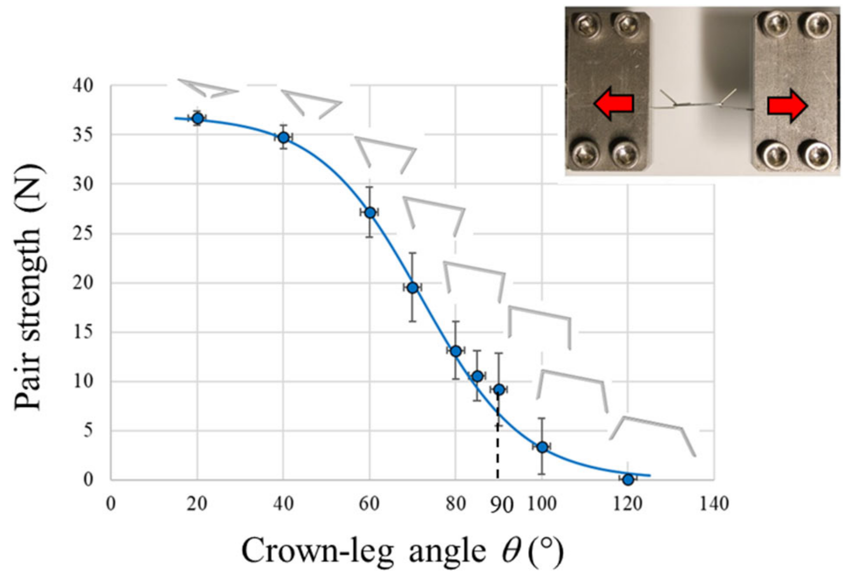
Tensile test on a bundle of 2000 staples: Strength = 28±14 N



Manipulating the crown-leg angle θ

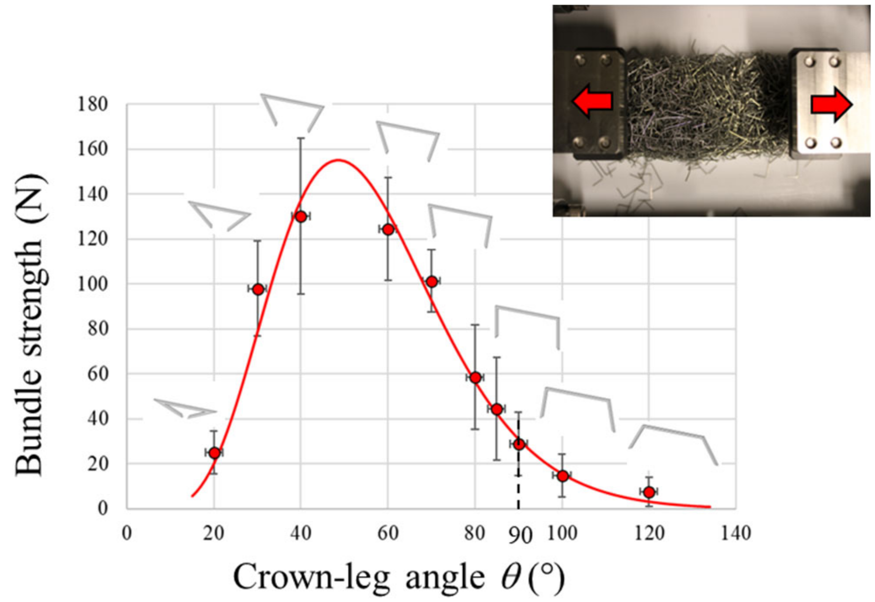


Staple pair strength



- Strength of a pair of 90° staples ~ 9N
- Closing the angle amplifies strength up to 4x
- Opening the angle rapidly leads to vanishing strength

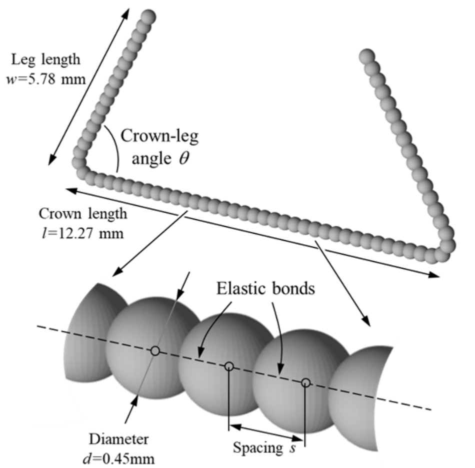
Staple bundle strength



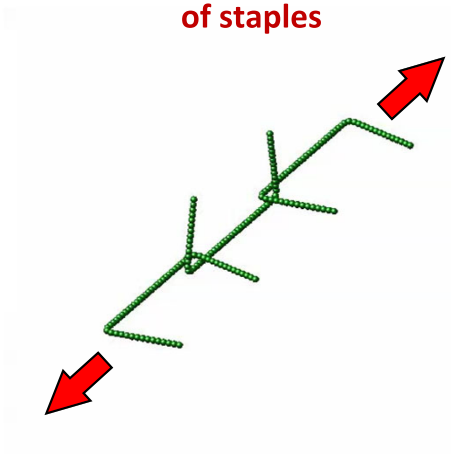
- Strength of a bundle of 90° staples ~ 28 N
- Closing the angle amplifies bundle strength up to 4.5x
- Maximum strength at angle $\theta \approx$ 40-60°

Discrete Element Method (DEM)

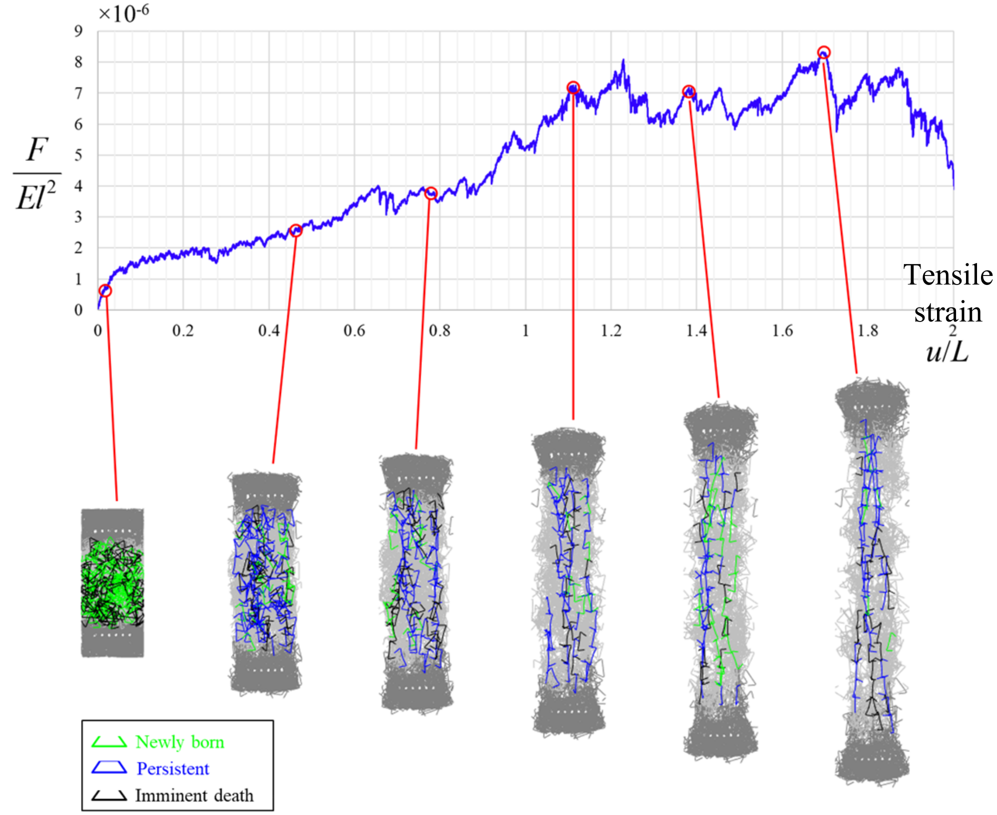
Overlapping sphere model with elastic bonds (LAMMPS)



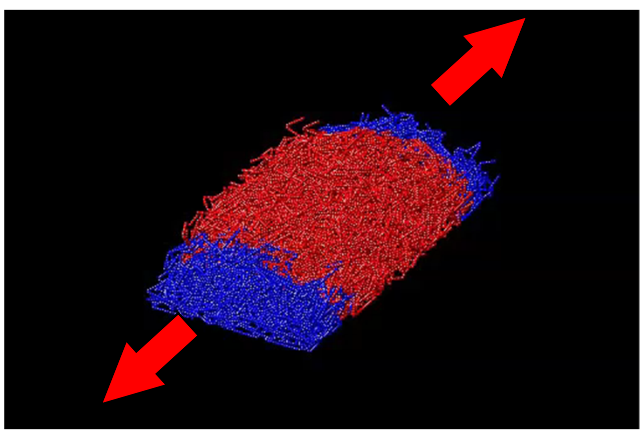
Tensile DEM on a chain of staples



Force extension curve of bundle



Tensile DEM of a bundle of 2000 staples

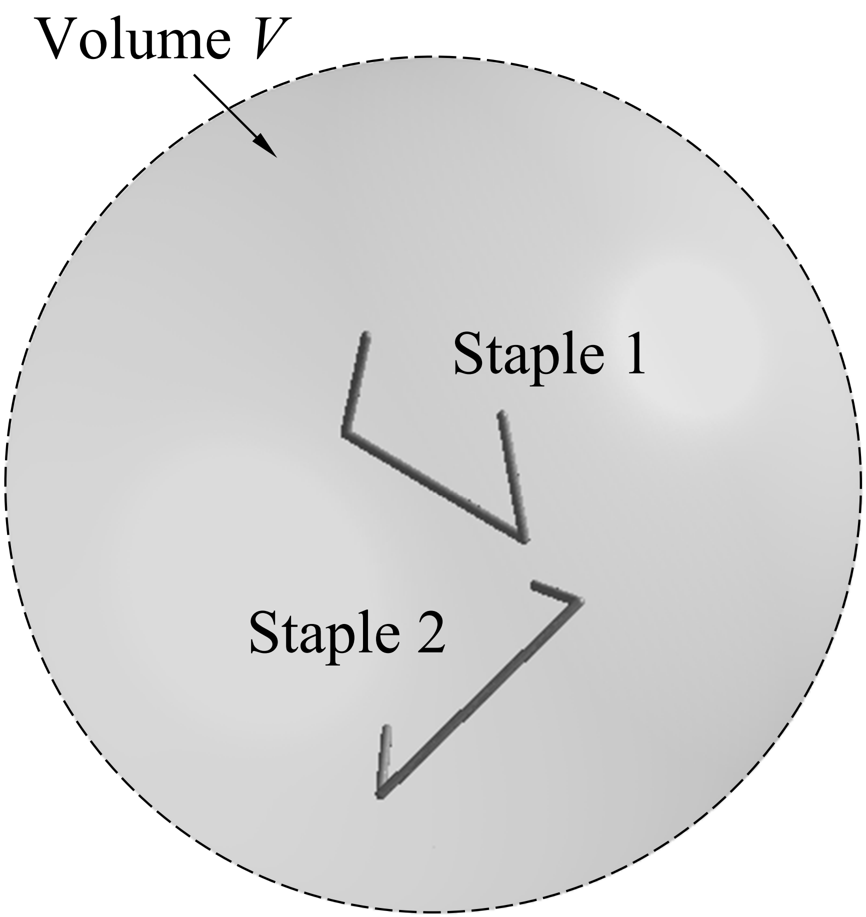


Saeed Pezeshki

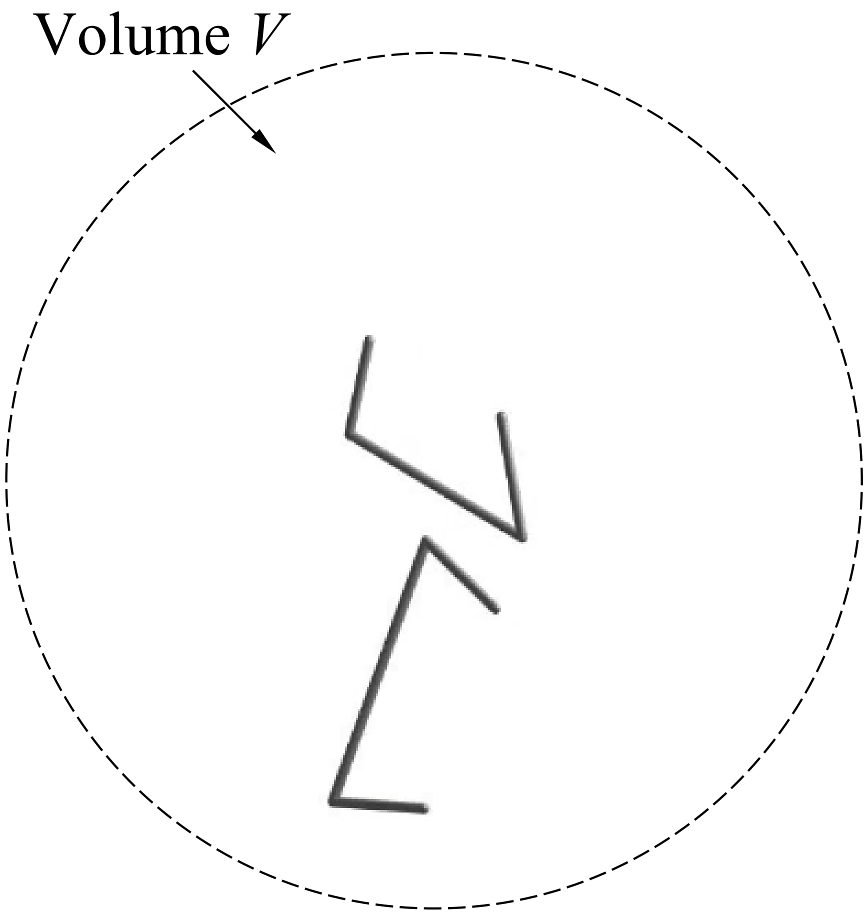
- **DEM model captures staple-staple interactions, large deformations in the bundle**
- **Tensile force lines are highly dynamic: As deformation increases they appear, break, others force lines appear and take over**

"Tunable entanglement and strength with engineered staple-like particles: Experiments and discrete element models" S. Pezeshki, Y. Sohn, V. Fouquet and F. Barthelat, JMPS 2025

What can we learn from two staples?



What can we learn from two staples?



Staple 1: fixed
Staple 2: Random position & orientation within volume V

Monte Carlo (N=10^6):

Excluded volume

Probability of collision

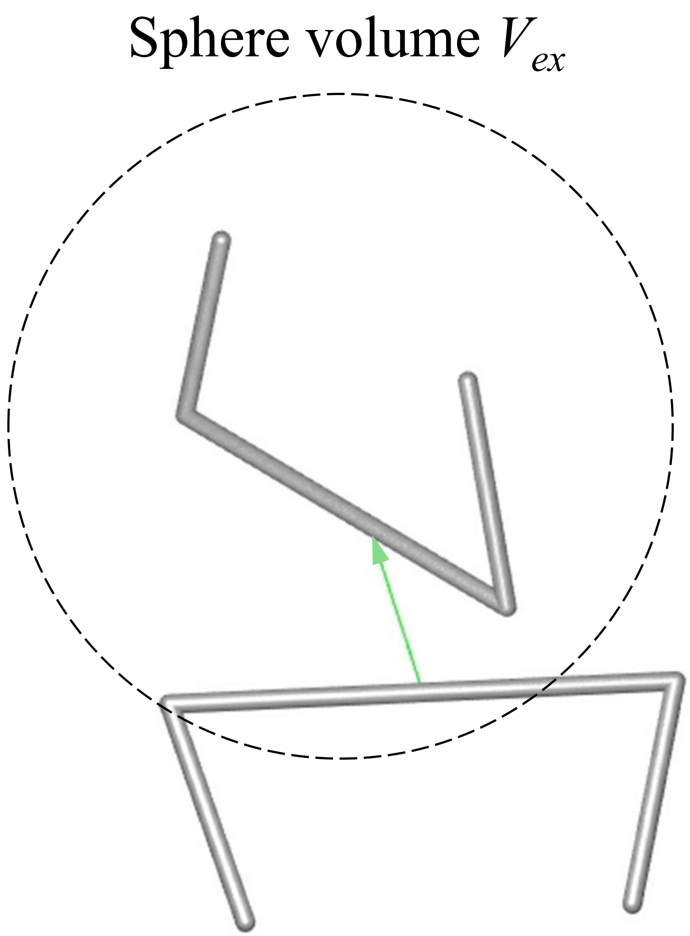
$$V_{ex} = p_c V$$

Volume fraction of staples

$$\phi \approx \frac{10}{V_{ex}}$$

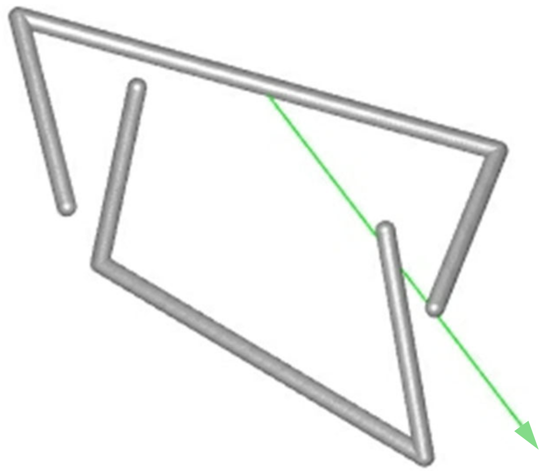
Philipse *Langmuir* 1996
Gravish, Franklin, Hu and Goldman *PRL* 2012

What (more) can we learn from two staples?

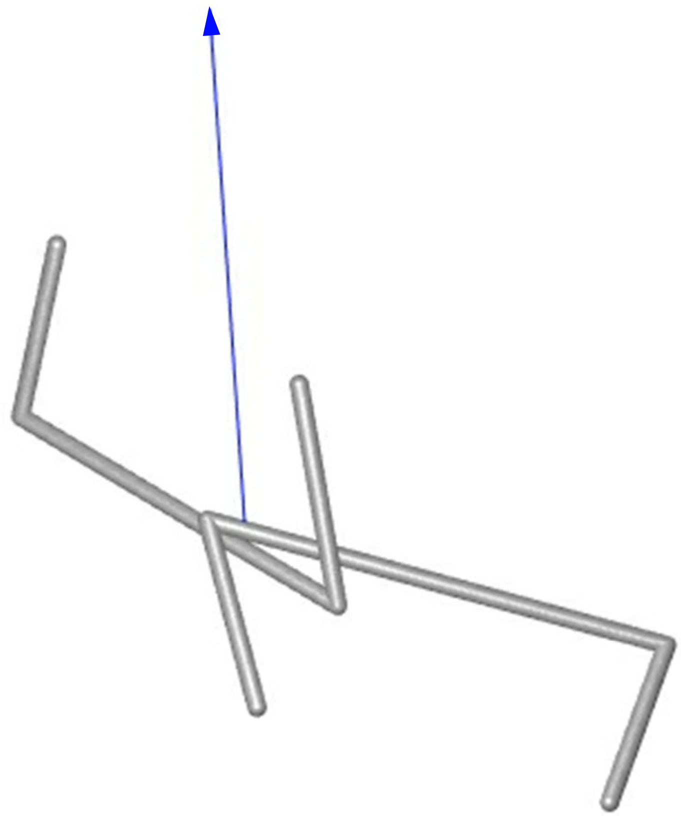


(here staple 2 does not entangle with staple1)

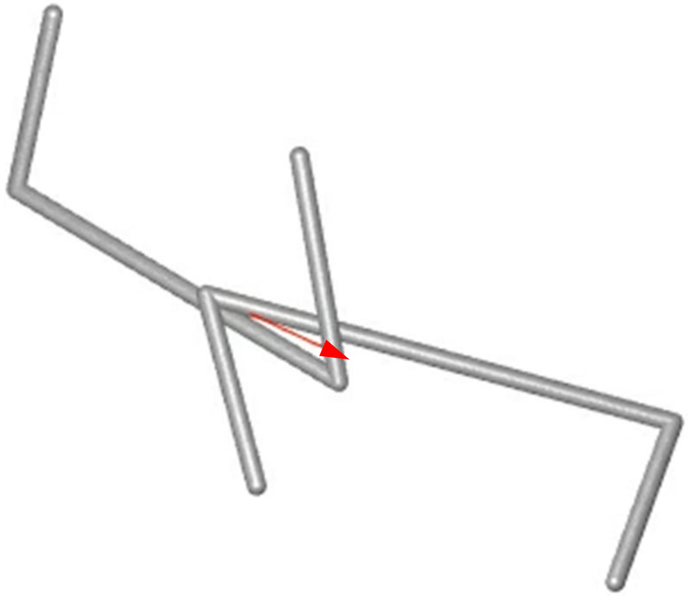
Staple "visibility":
probability p_v of colliding with another staple



If visible, staple 2 may “engage” with staple 1



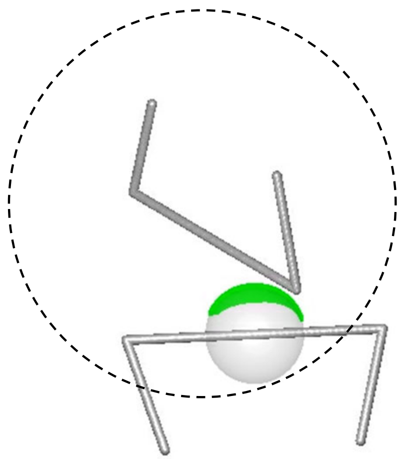
After a "rebound", staple 2 may disengage, or…



Staple 2 may entangle deeper with staple 1

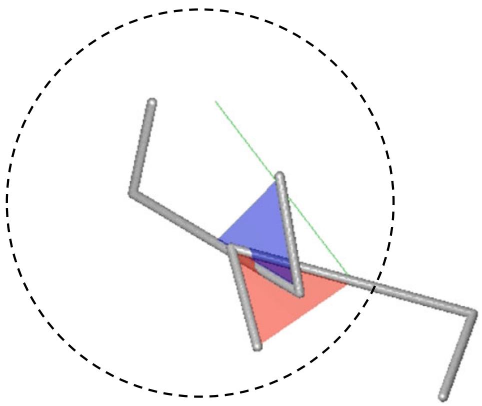
Entanglement Density

Sphere volume V_{ex}



Staple "visibility": probability p_v of colliding with another staple

Sphere volume V_{ex}



Staple entanglement: probability p_t of two stable to entangle

Monte Carlo (N=10^6):

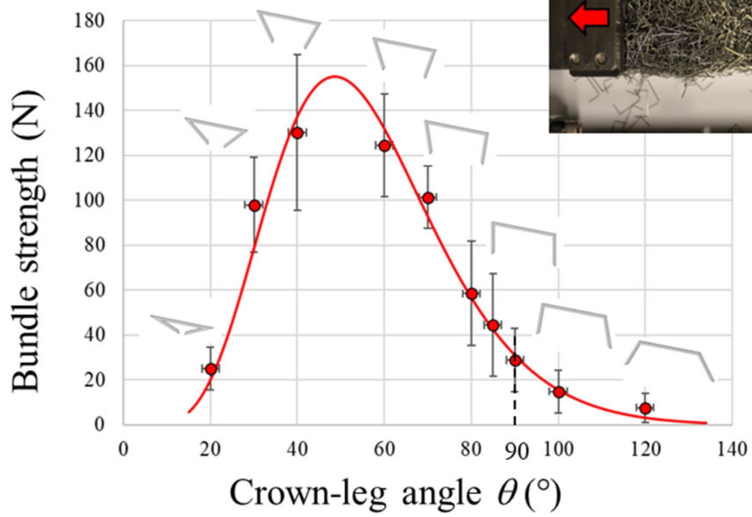
Average entanglement density: $\phi_t = p_v p_t \phi$

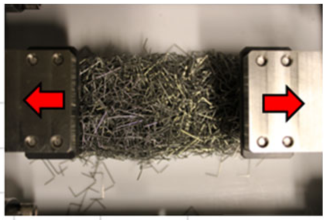
Average number of entangled staples in a $l \times l \times l$ volume (l=size of the staple)

Sohn, Pezeshki and Barthelat, to appear in *Granular Matter*

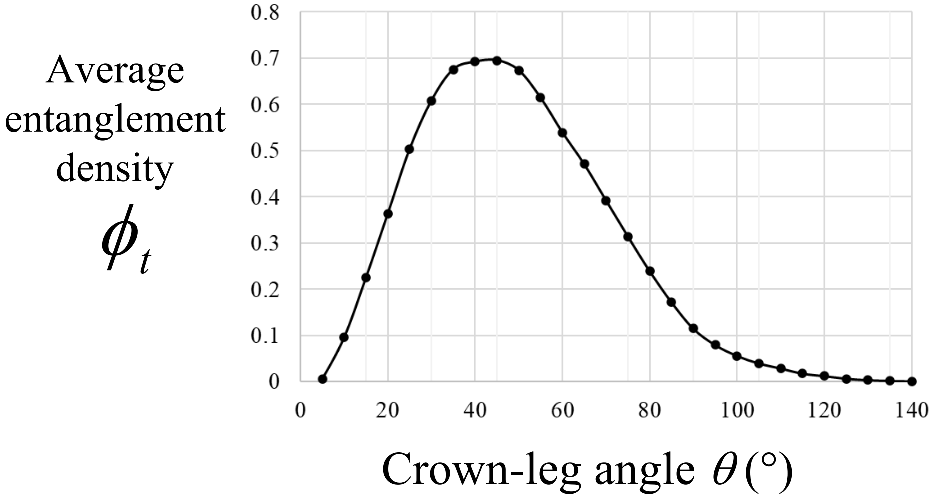
Effect of crown-leg angle

Experiments:



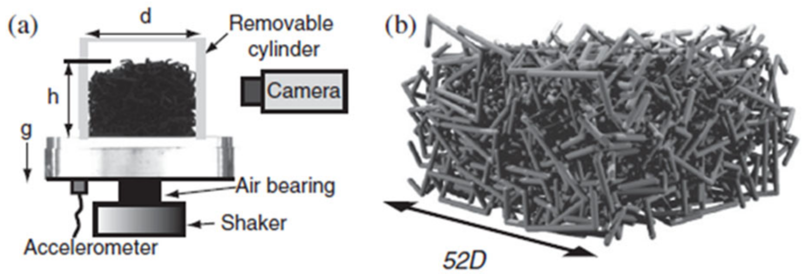


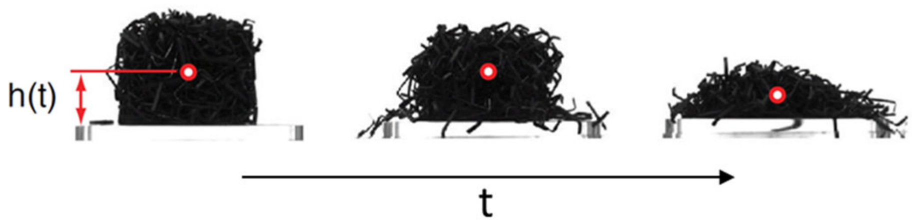
Two-staple Monte Carlo:

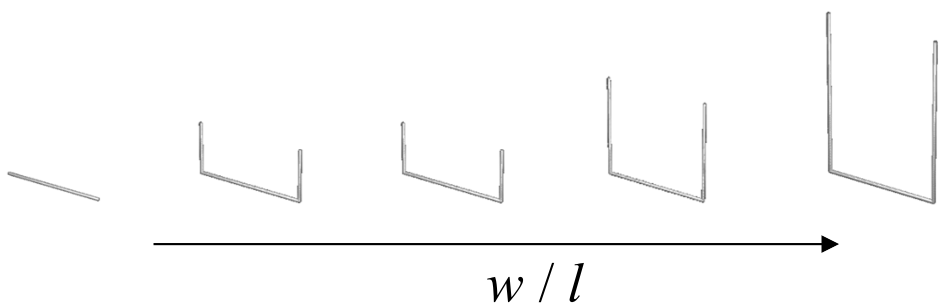


Effect of leg length

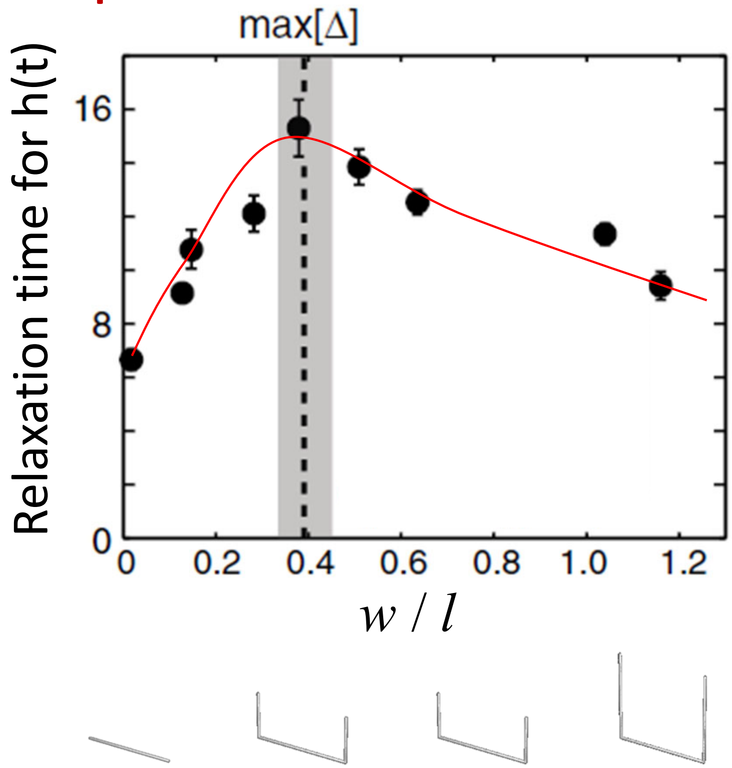
Gravish, Franklin, Hu and Goldman:
"Entangled Granular Media" *PRL* (2012)



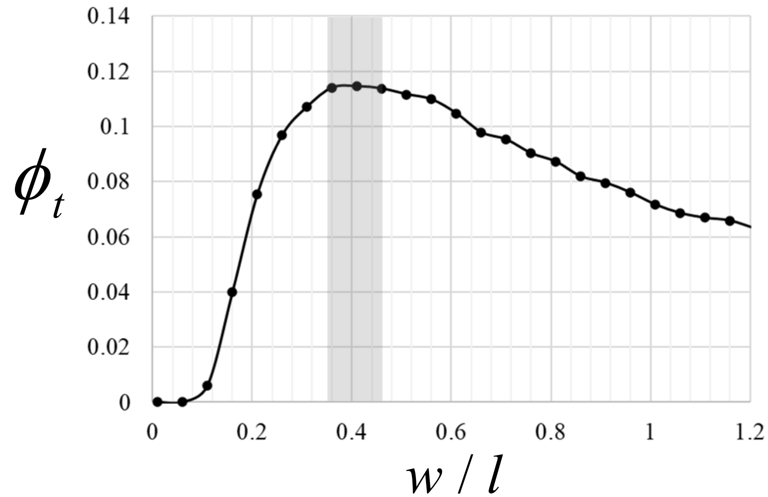




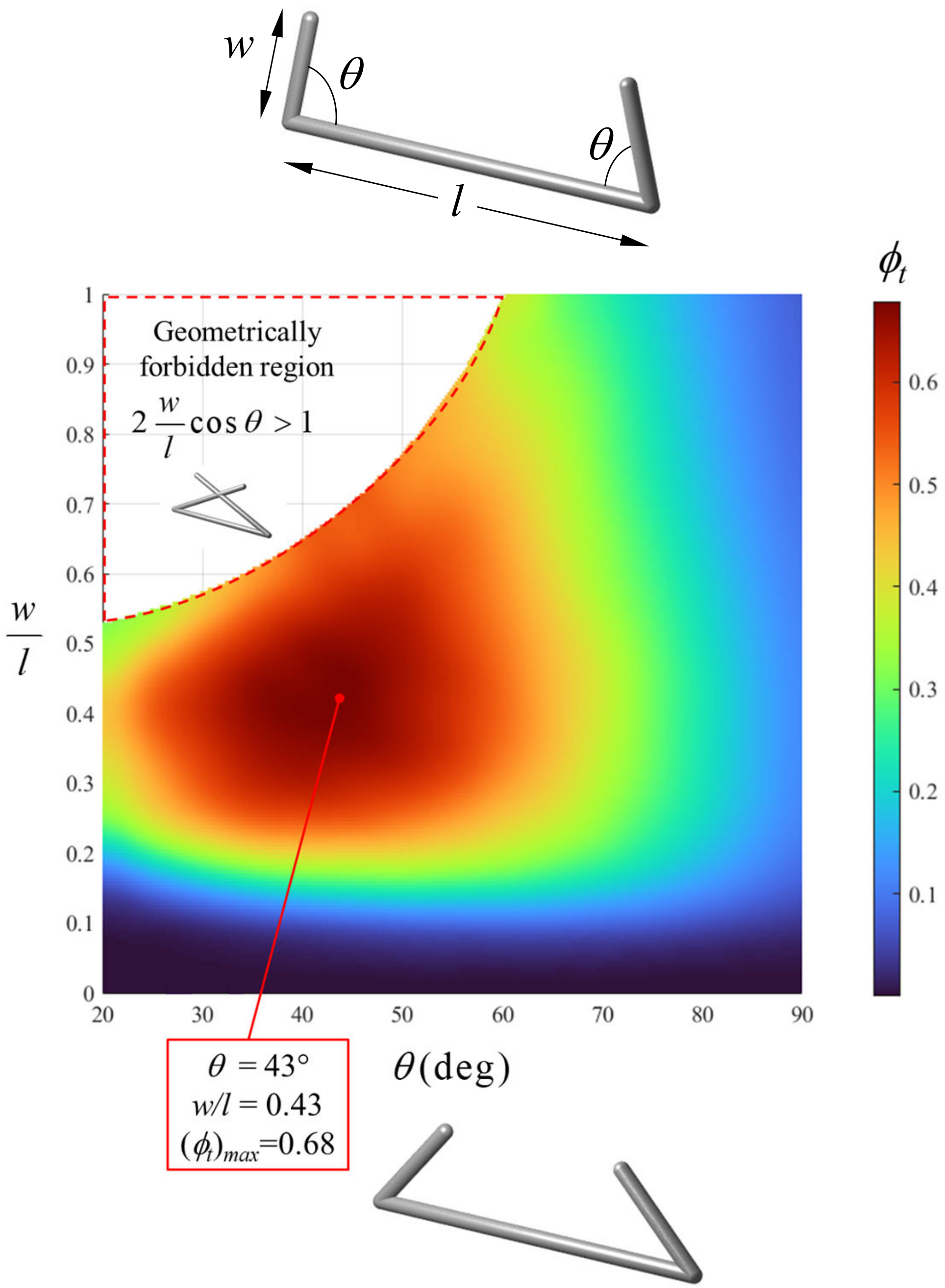
Experiments:



Two-staple MC:



Designing / optimizing particle shape



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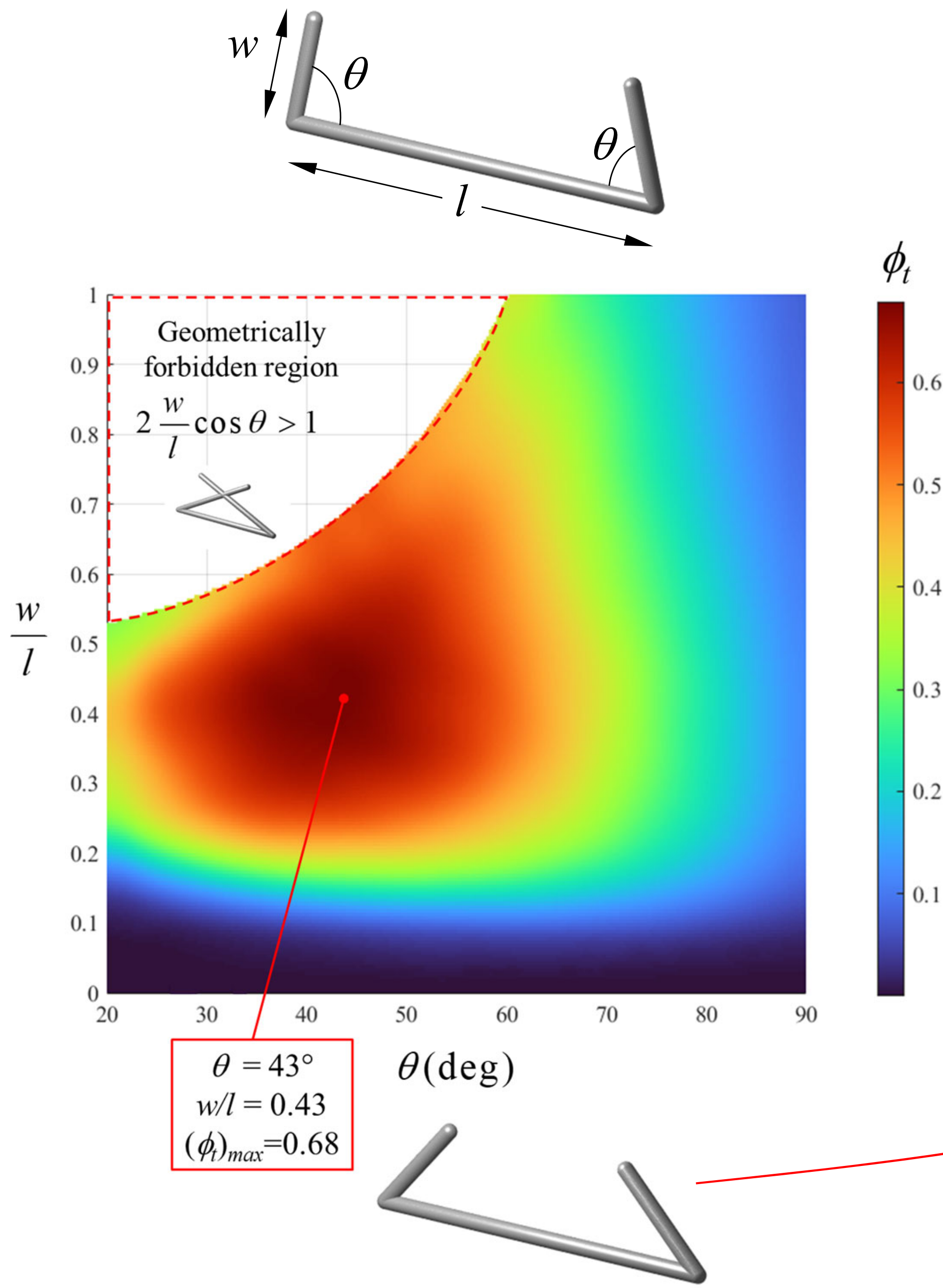
Designing / optimizing particle shape



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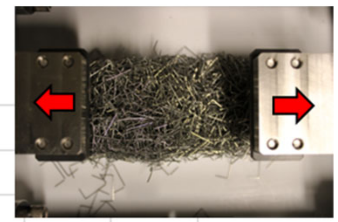
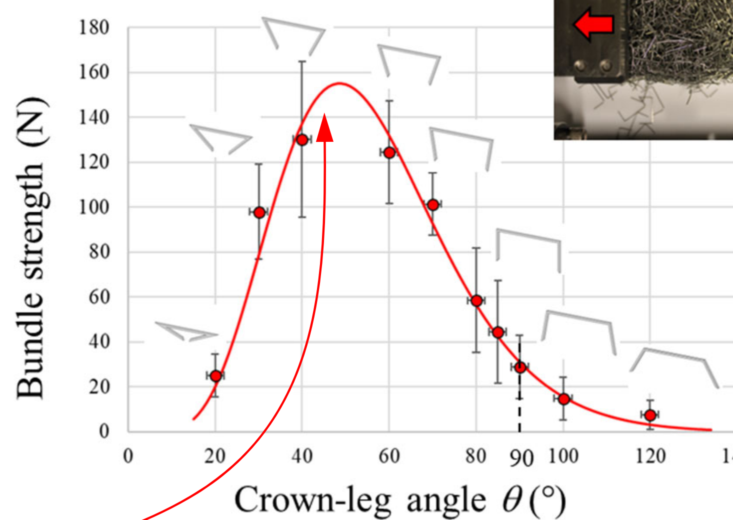
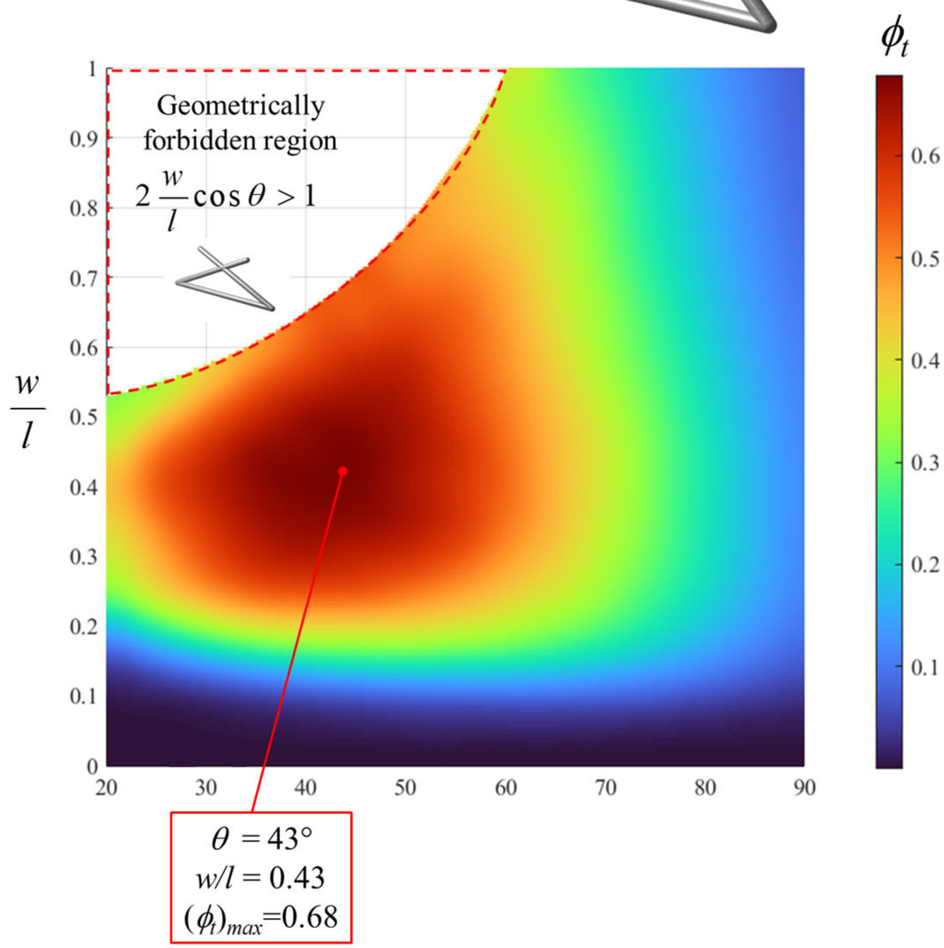
Experiments:



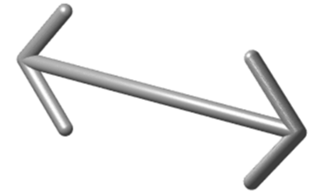


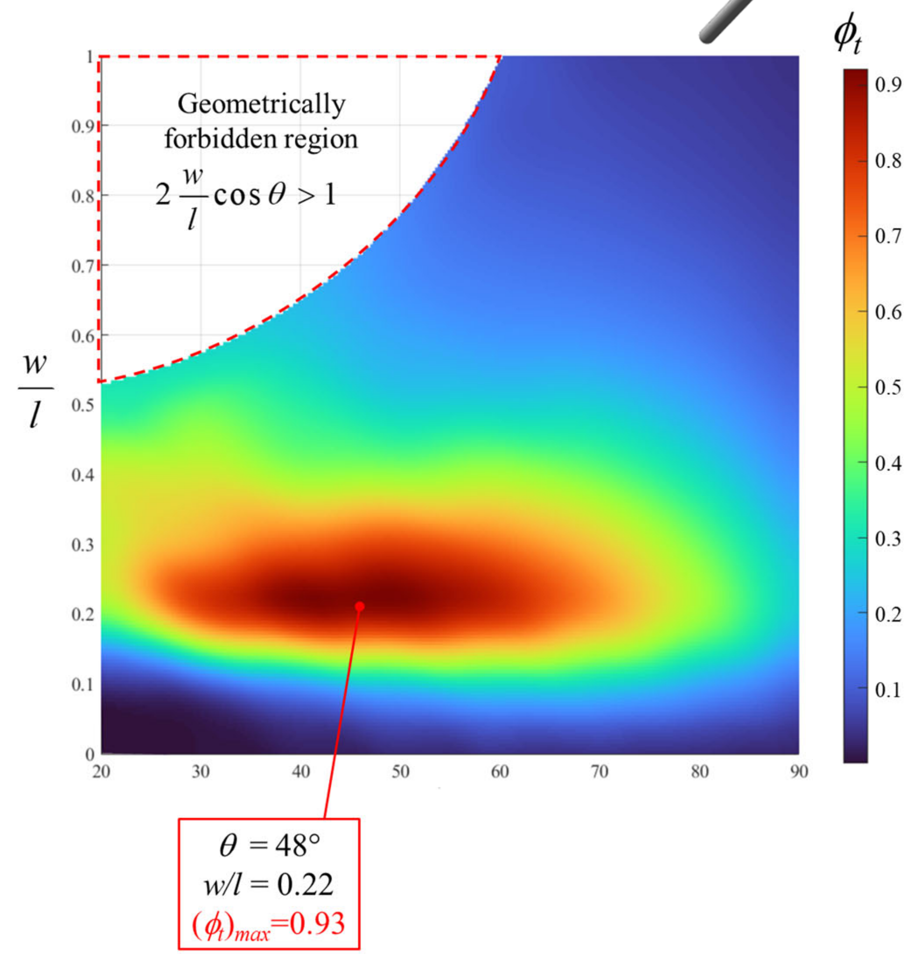
Designing / optimizing particle shape

C-shape staple:

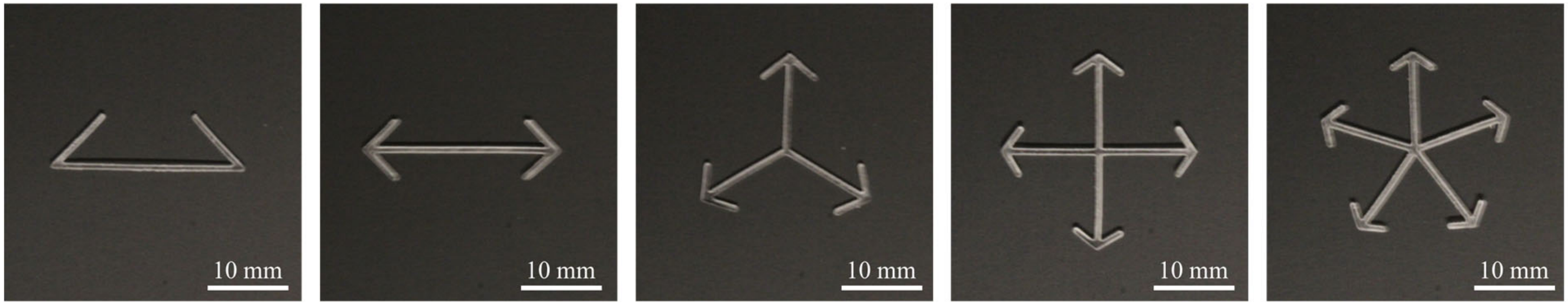


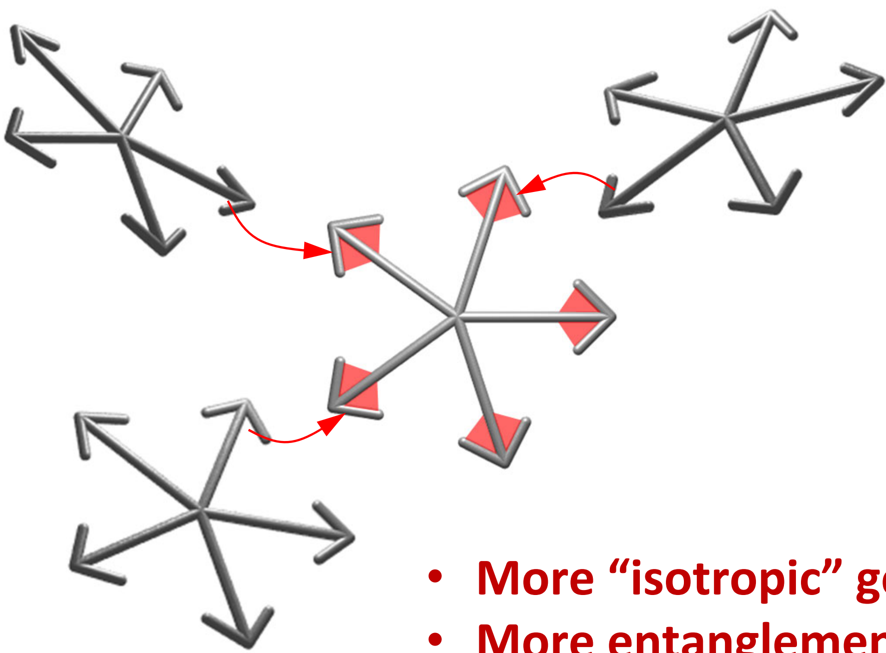
Barbed particle:





Enriching particle geometry

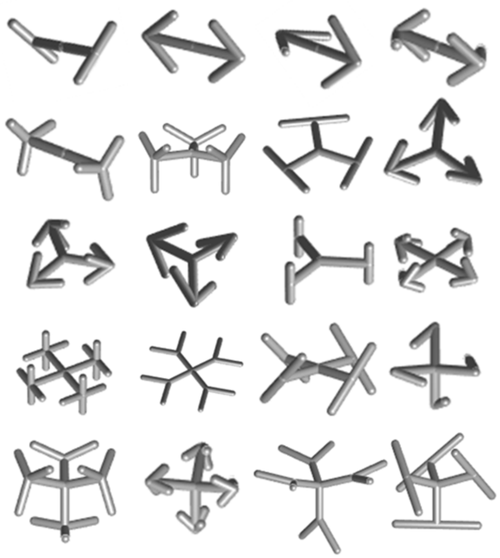




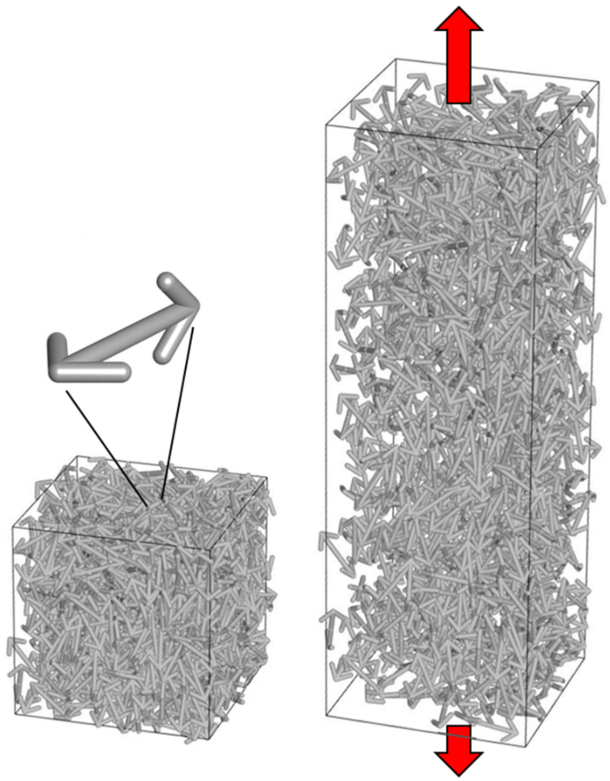
- More "isotropic" geometries
- More entanglement sites = higher entanglement densities

(Many) more particle geometries

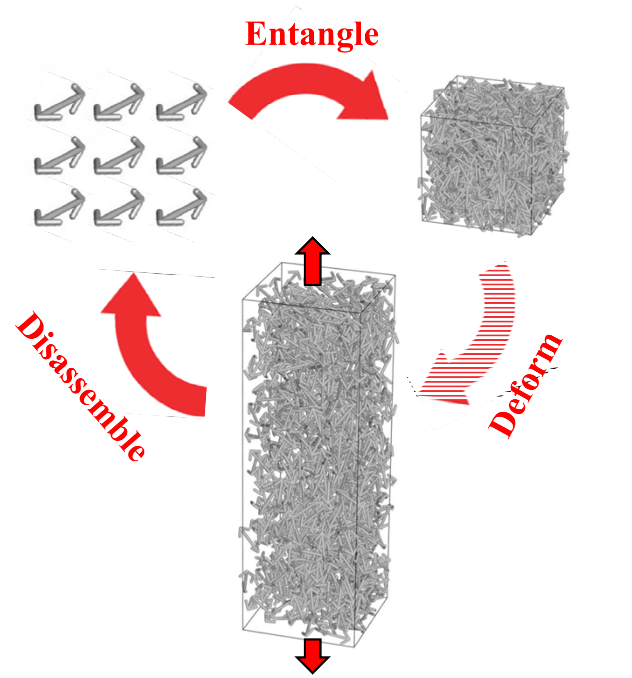
Individual particle design: branches, hooks and barbs



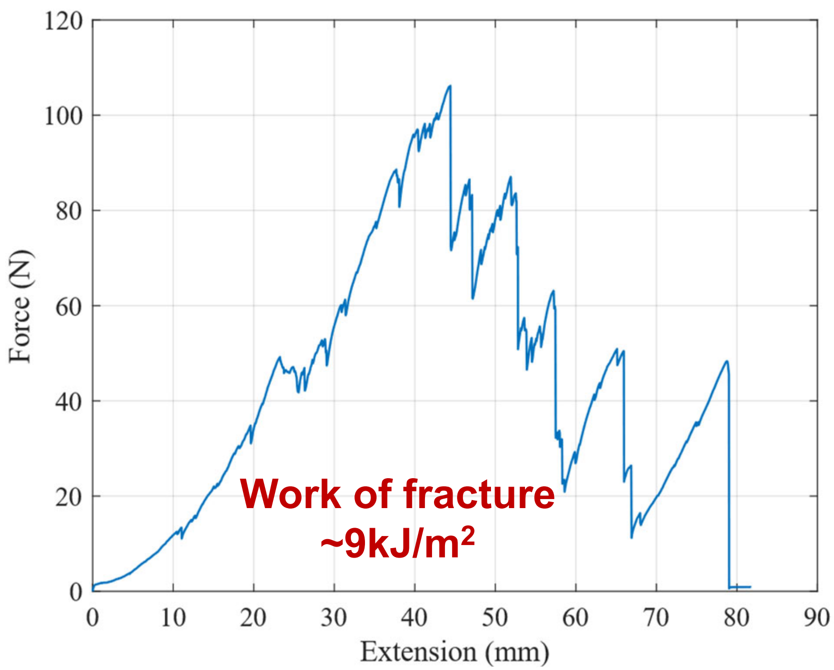
Tensile strength

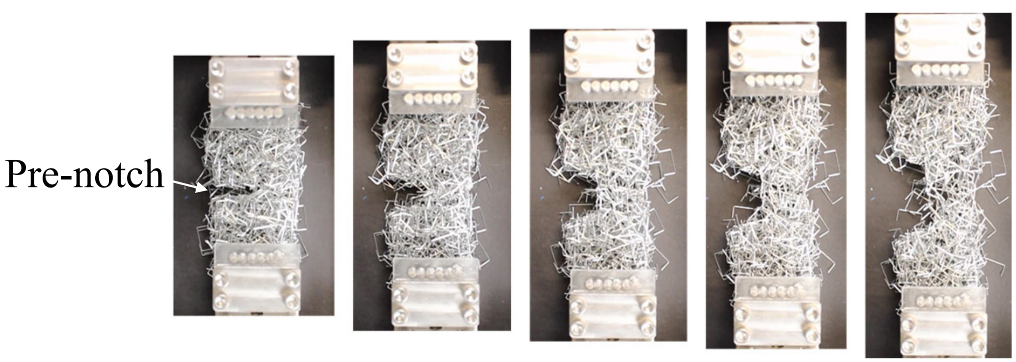


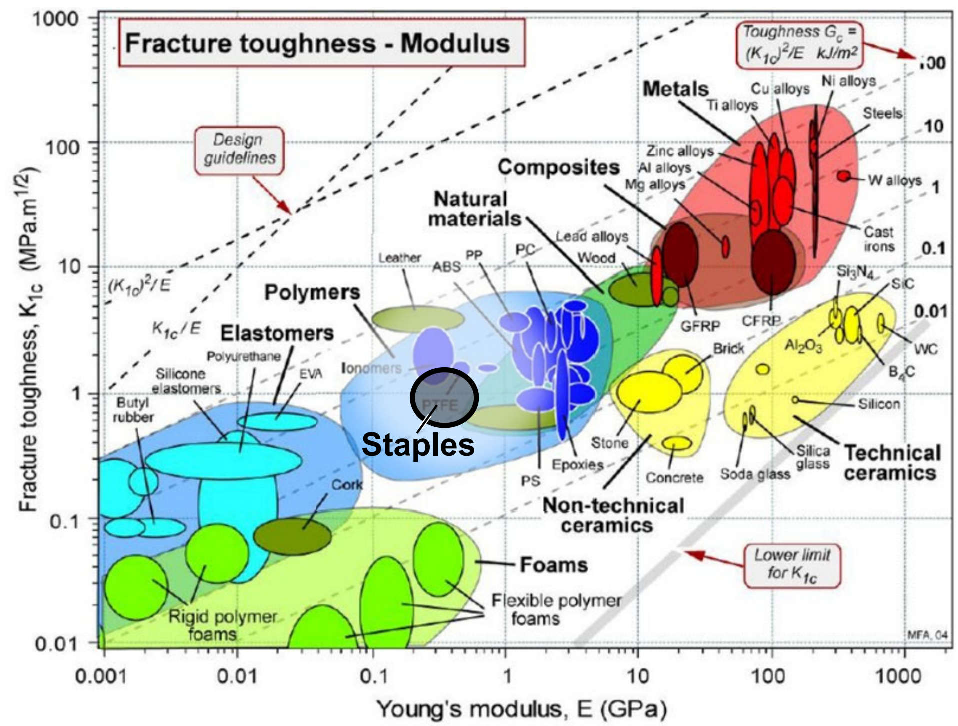
Recyclability / healing



Fracture toughness of entangled particles



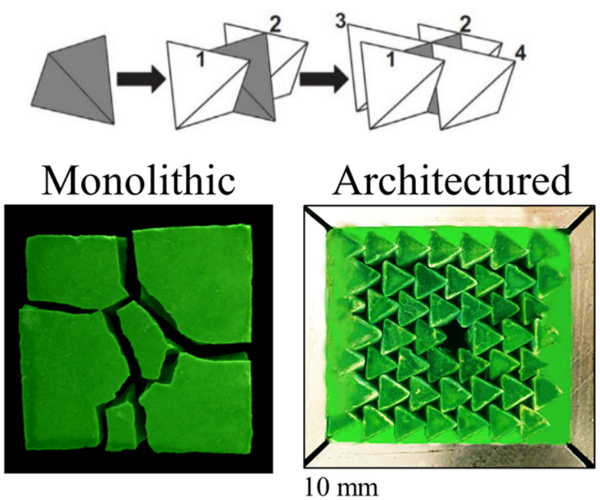




Summary

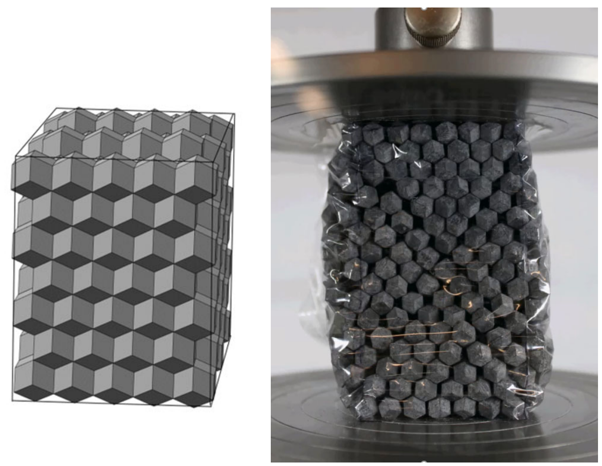
Rigid blocks + weak and deformable interfaces+ architecture
=
Rich set of tunable micromechanics and high performance

Topologically interlocked panels are as strong as the monolithic but 30-50 times tougher



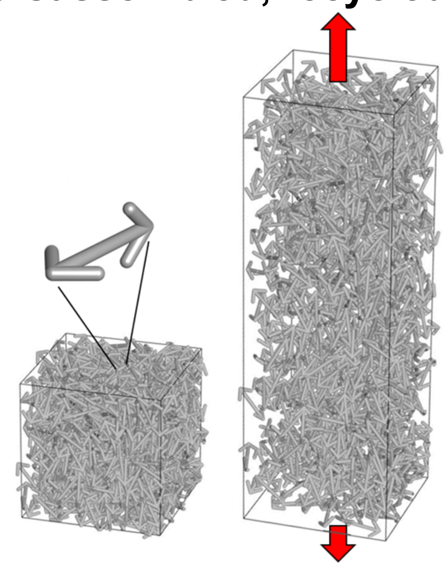
Mirkhalaf, Zhou, Barthelat
PNAS (2018)

Granular crystals are 10-30 times stiffer and stronger than randomly packed spheres



Karuriya, Barthelat *PNAS* (2023)
Karuriya, Barthelat *JMPS* (2024)
Karuriya, Barthelat *EML* (2024)

Entangled materials generate tensile strength, can be assembled, disassembled, recycled.



Pezeshki, Barthelat JMPS (2025)
Sohn, Barthelat, to appear in *Granular Matter*

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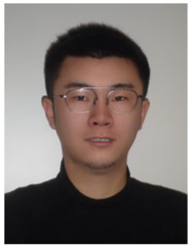
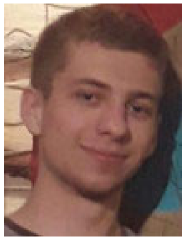


Tian Gao

Youhan Sohn

Saeed Pezeshki



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Mechanics of Materials
CMMI-2033991