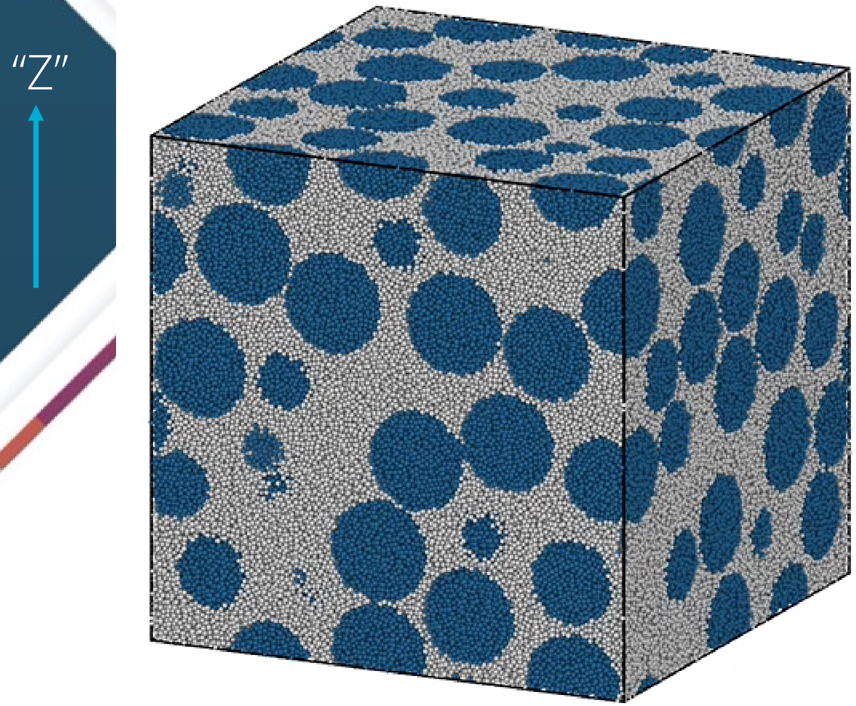
SAND2023-07151C

Sandia National Laboratories

Exceptional service in the national interest

MESOMECHANICS OF HIGHLY FILLED PARTICLE REINFORCED COMPOSITES

Using A Bonded Particle Method



Judith A. Brown[1], Joel T. Clemmer[1], **Kevin N. Long[1,*]**

[1]Engineering Sciences Center, Sandia National Laboratories, Albuquerque, NM 87185

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly ...artment of Energy's National Nuclear Security Administration under contract DE-NA0003525.

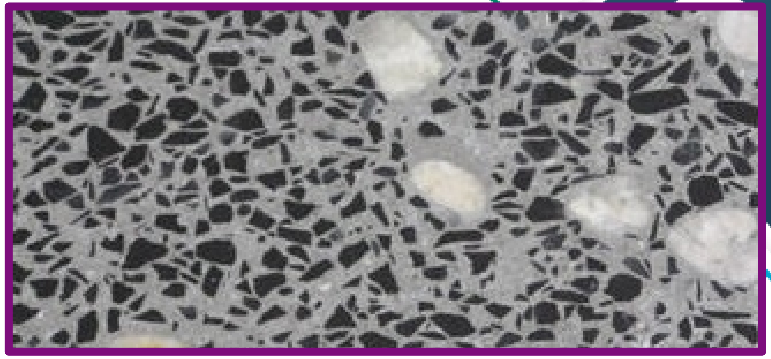
POLYMER BOUND GRANULAR COMPOSITES

Many instances Across industries:

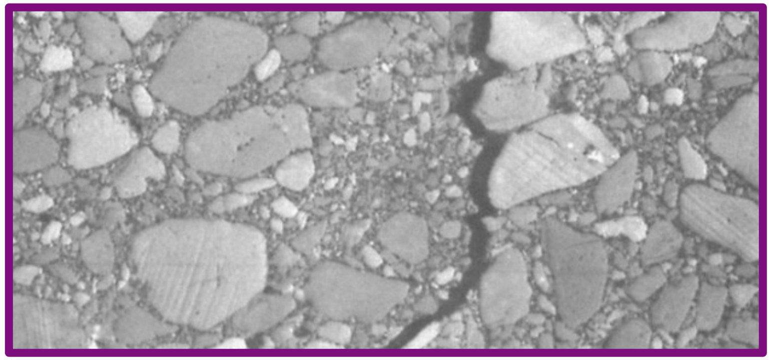
- Concrete, asphalt
- Polymer-bonded explosives
- Electronics Packaging (Electrically conductive adhesives and CTE matched overpotting materials)
- 3D binder jet additive manufacturing

Complex mechanics:

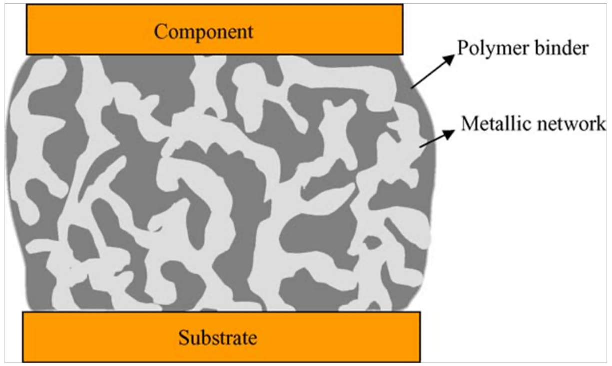
- Highly heterogeneous, granular vs. polymer
- Phases often have markedly different properties
- Length scale of each can be very different, ~100:1



Polished concrete (Wikipedia)



PBX microstructure ~ 1 mm
(PJ Rae *Proc. R. Soc. Lond. A.* 2002)



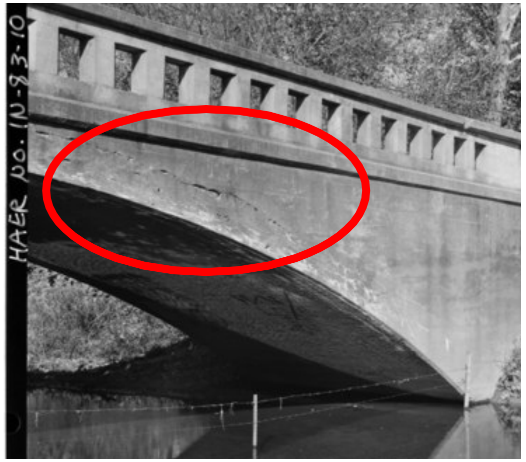
Conductive Adhesive
(Li and Wong, 2006)

THEIR MECHANICS AFFECTS SYSTEMS RELIABILITY

Transportation

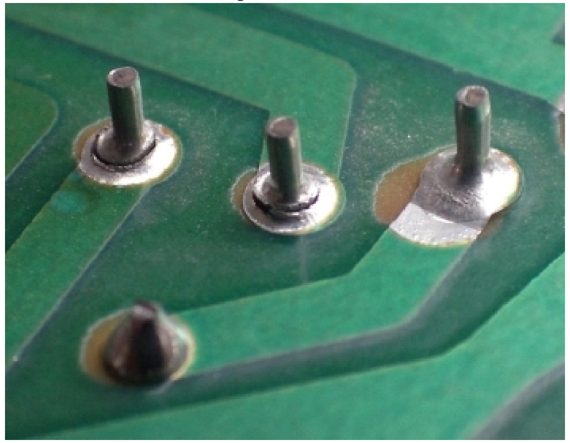
commons.wikimedia.org/wiki/File:Pothole_Big.jpg

Construction



www.loc.gov/pictures/item/in0447.photos.374550p/

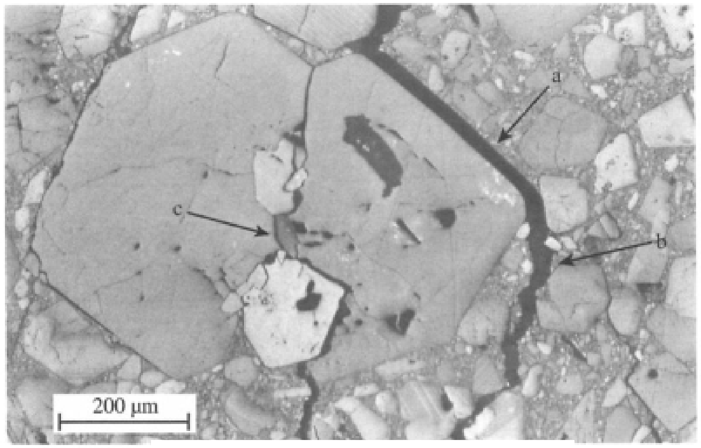
Reliability of electronics



commons.wikimedia.org/wiki/File:Gebrochene_loetstellen.jpg

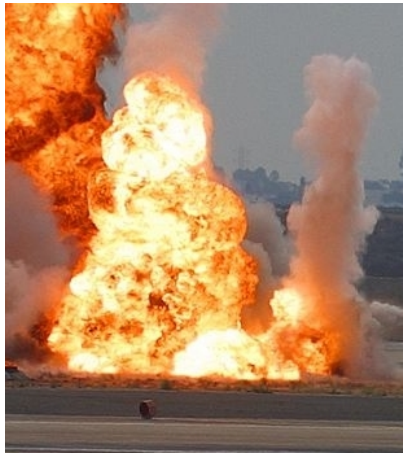
Food industry - tactile response

commons.wikimedia.org/wiki/File:Nestle-crunch-broken.jpg



PJ Rae Proc. R. Soc. Lond. A. 2002

Safety in damaged components



commons.wikimedia.org/wiki/File:
Explosions_at_Miramar_Airshow.jpg

KNOWLEDGE GAP: HOW DO WE CONNECT MATERIAL INPUTS TO COMPLEX MECHANICAL BEHAVIORS?

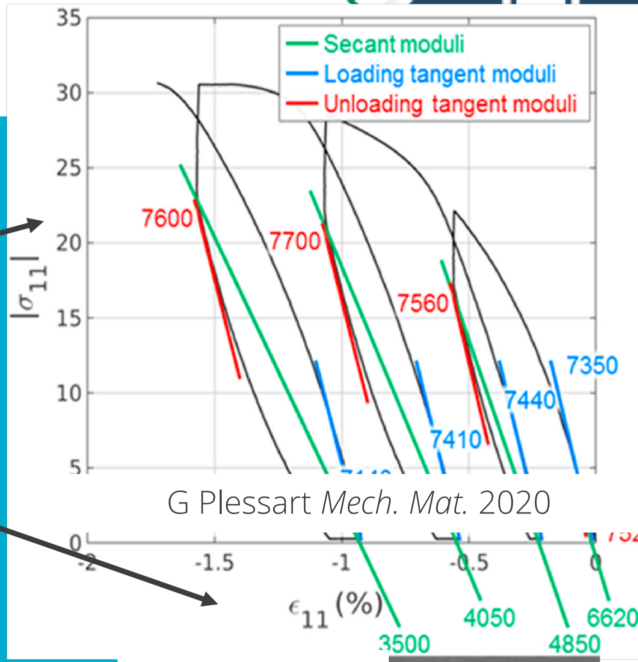
Material inputs

- Filler materials, structure, and size distribution
- Polymer binder characteristics
- Filler volume fraction
- Filler packing
- Processing

?

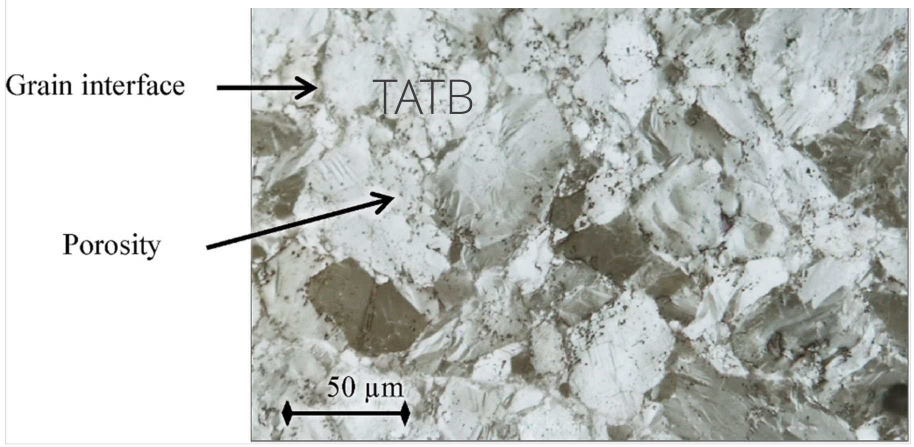
Mechanical Behavior

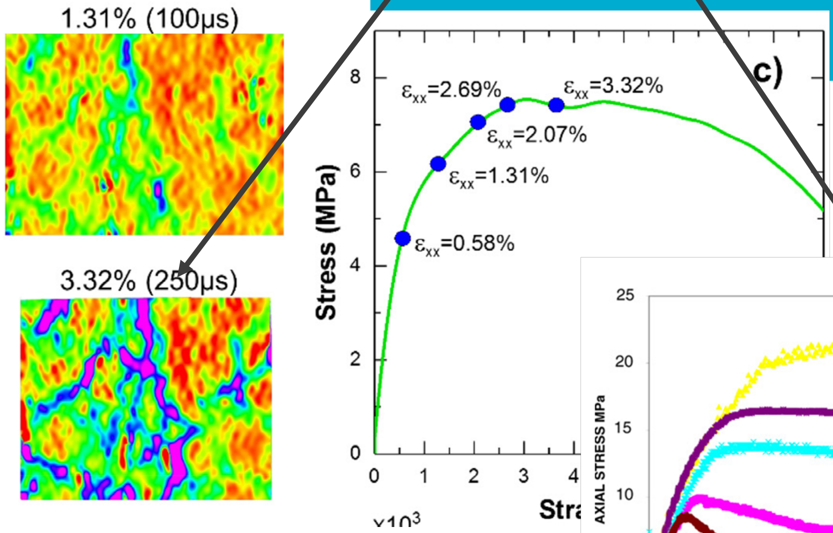
- Viscoelasticity
- Failure
- Plasticity
- Damage



G Plessart *Mech. Mat.* 2020

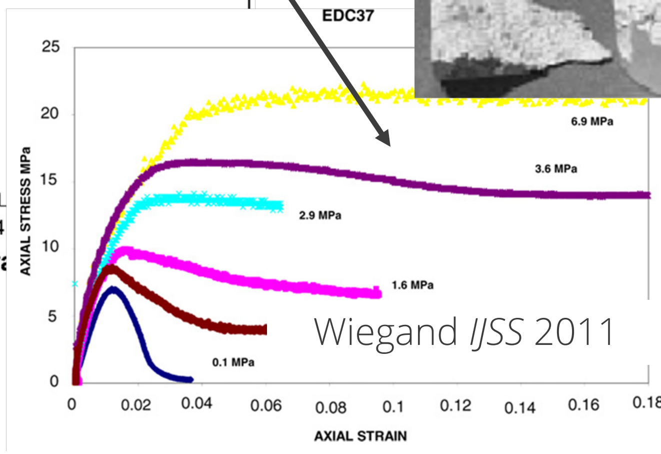
G Plessart *Mech. Mat.* 2020





Ravindran *Mech. Mat.* 2017

Peterson *Prop., Exp. Pyro.* 2005



Wiegand *IJSS* 2011

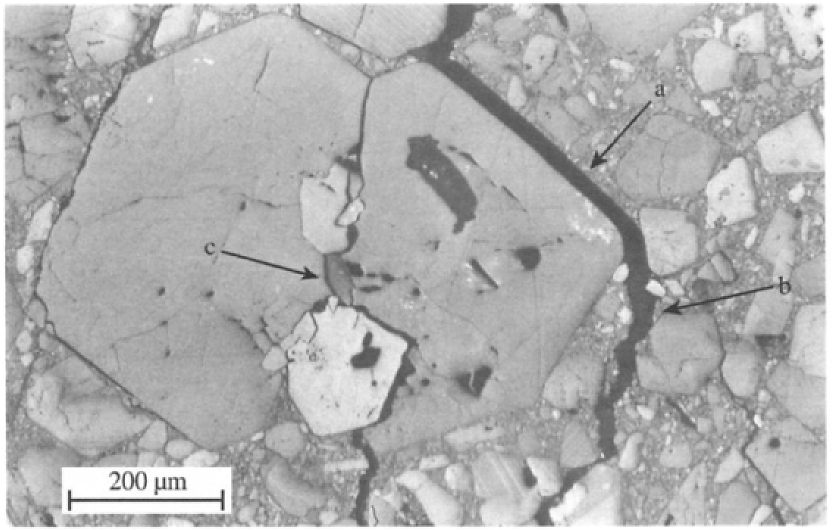
CHALLENGES IN MODELING MECHANICS AND FAILURE

Many physical mechanisms:
- Plasticity/viscoelasticity in binder (or grains)
- Rotation/delamination/friction in grains
- Crack nucleation, growth, coalescence

Variety of length scales
- Failure often dominated by binder phase:
  Can be much smaller/thinner than grains
- Macro response depends on 100s-1000s of grains
  Need to simulate an representative sample

Constituent phase mechanics may be unknown

Oodles of discontinuities

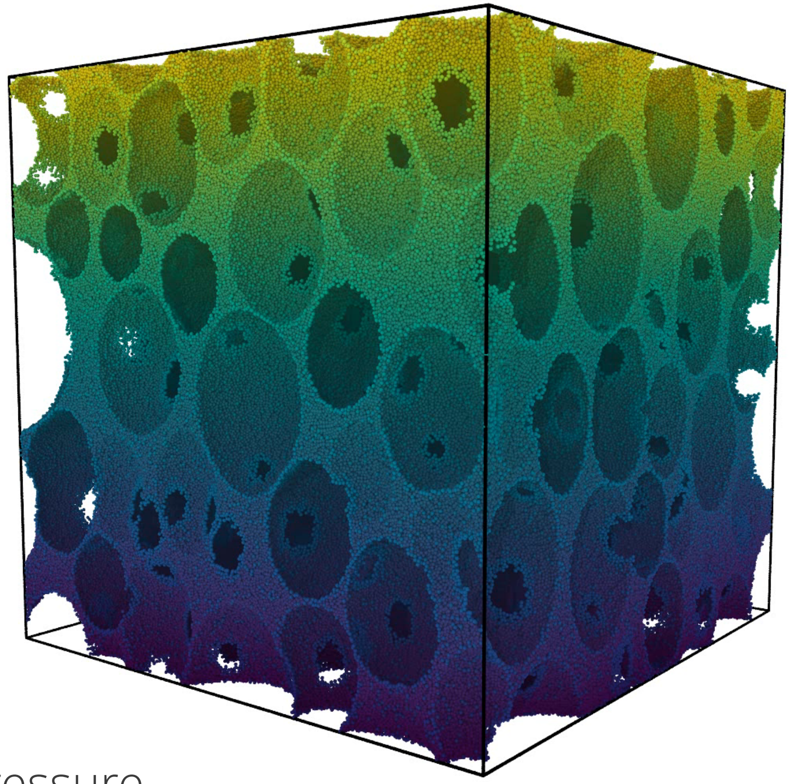


PJ Rae Proc. R. Soc. Lond. A. 2002

We need a flexible, inexpensive, and robust modeling approach to connect microstructure inputs to emergent macroscale responses

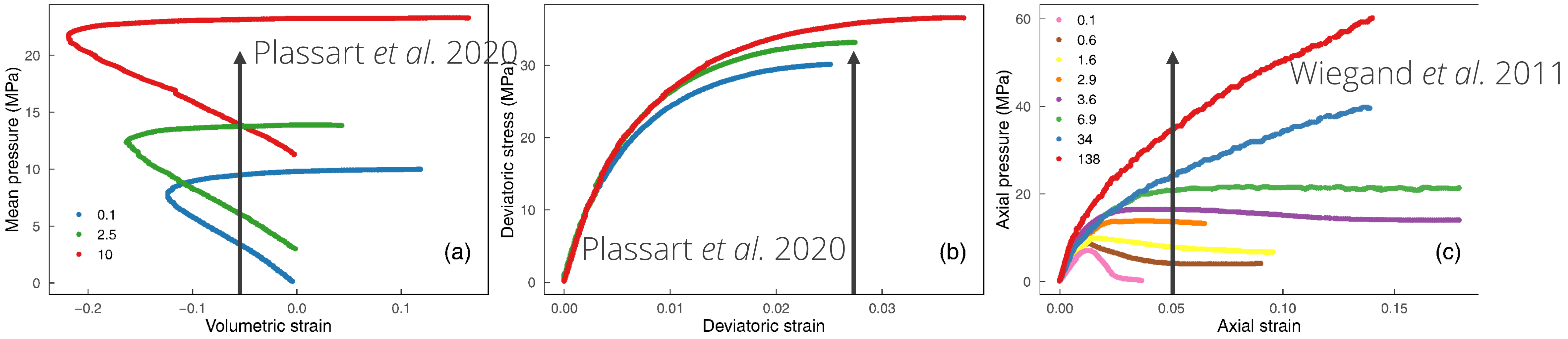
OBJECTIVES

1. Develop and deploy a robust computational micromechanics capability to simulate emergent macroscale phenomena from known microstructural and constituent material inputs



2. Investigate a relatively simple problem:
   - the origin of pressure dependence in limiting forms of highly filled particle re-enforced composites

Increasing Confining Pressure



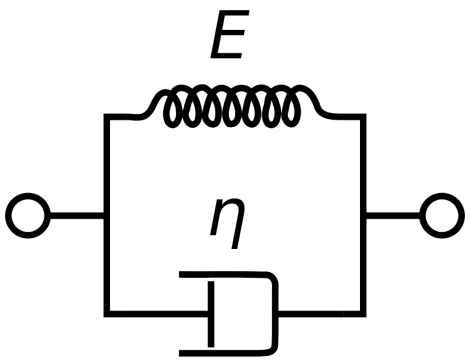
OUR MODELING APPROACH: A BONDED PARTICLE MODEL (BPM)

- Lagrangian material points connected by a breakable (spring) network of bonds
- Unbonded particles repel with various contact forces

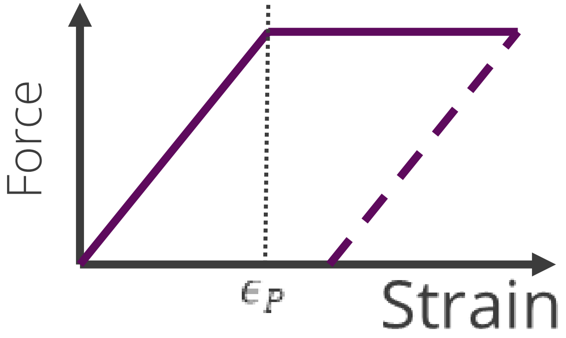
- Solve Newtonian equations while obeying symmetries
⇒ Physics-based approach, focus on emerging behavior

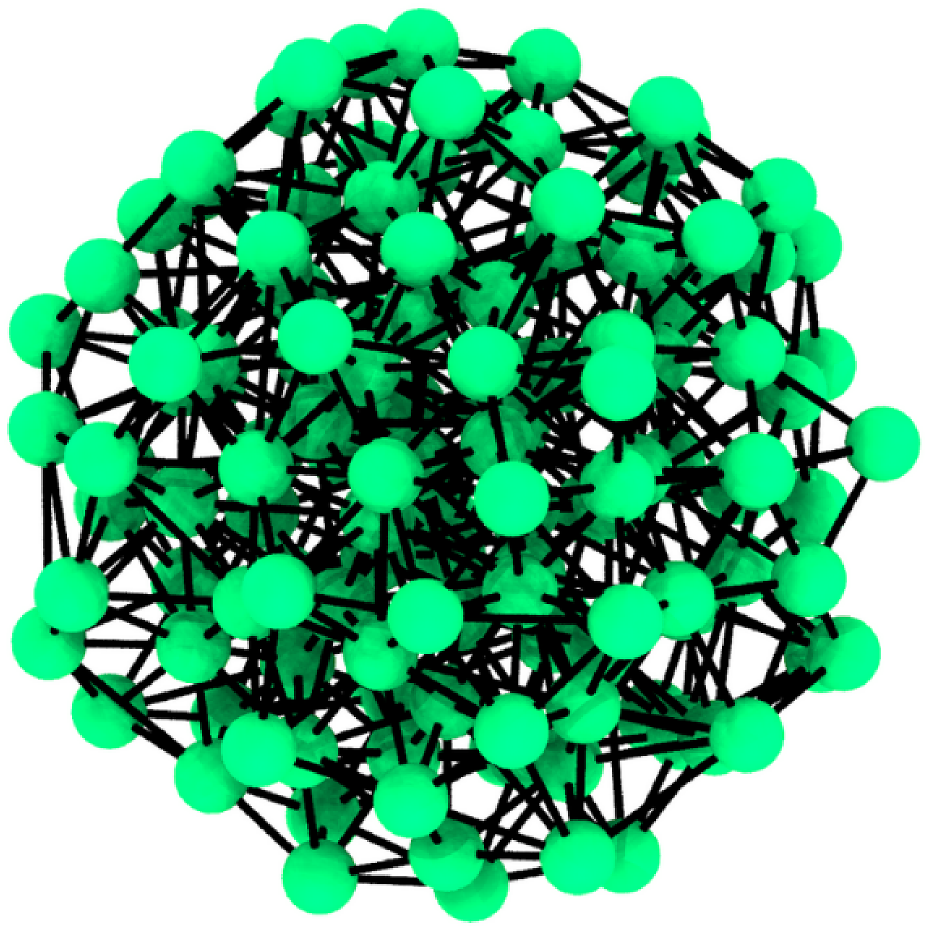
- Vary details of bond forces to capture different physics

Viscoelastic bond



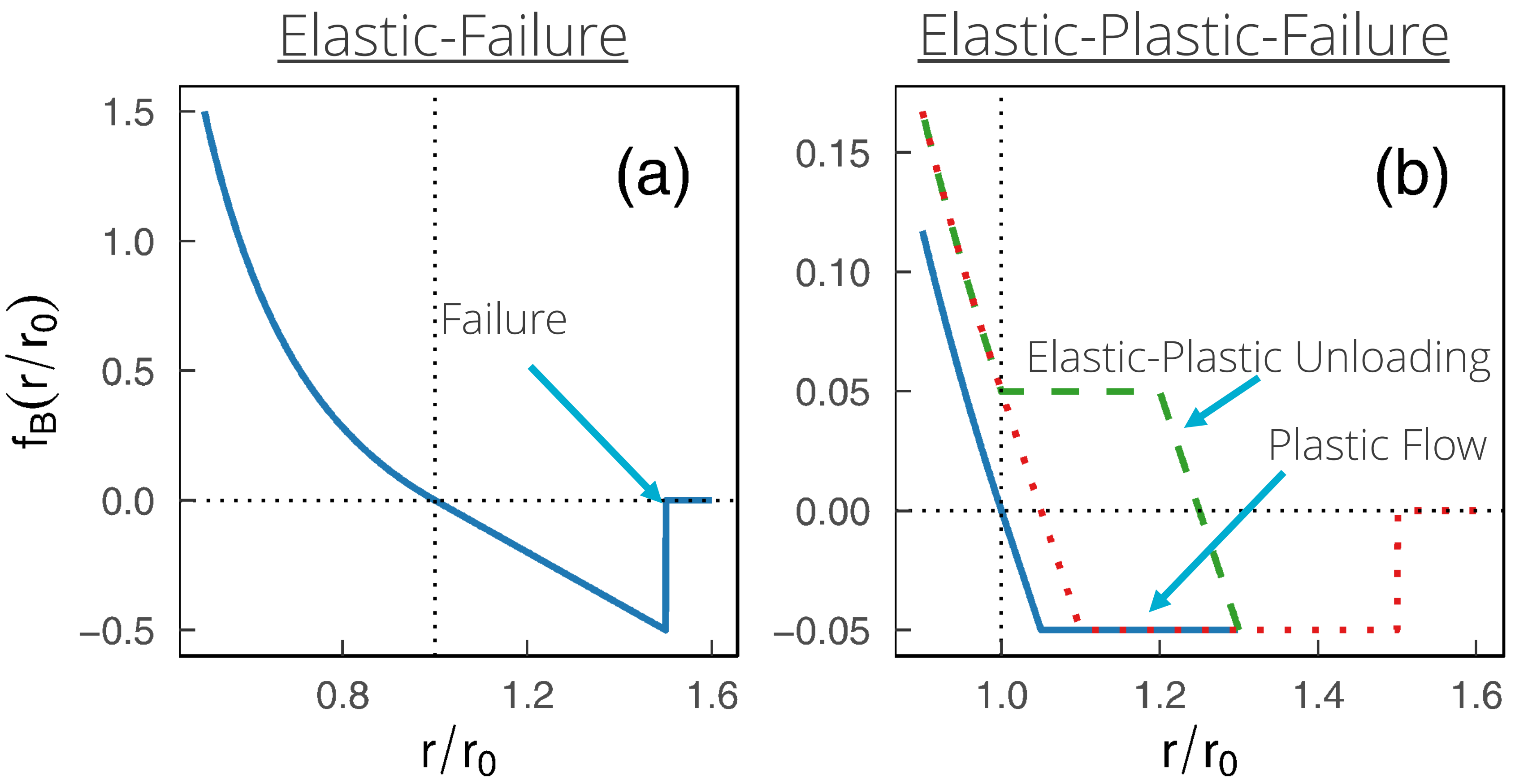
Elastic-Plastic bond





BOND MODELS USED IN THIS STUDY

$$\vec{f}_B(\vec{r}, r_{eq}) = \hat{r} \begin{cases} 0.5 k_b r_{eq} \left([r_{eq}/r]^2 - 1 \right) & \epsilon \leq 0 \\ -k_b r_{eq} \epsilon & 0 < \epsilon \leq \epsilon_p \\ -k_b r_{eq} \epsilon_p & \epsilon_p < \epsilon \leq \epsilon_{max} \\ 0 & \epsilon > \epsilon_{max} \end{cases}$$

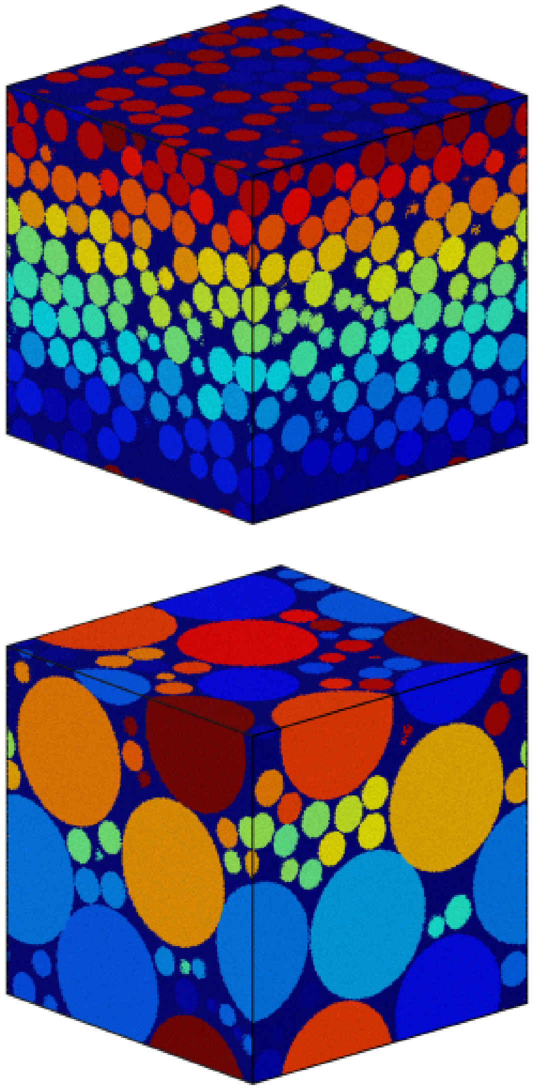


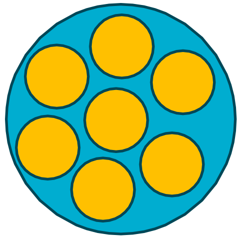
By tuning the initial bond stiffness, plasticity, and strain to failure, the response of different phases and their adhesive interactions can be qualitatively modeled

MESOMECHANICS APPROACH
REPRESENTATIVE VOLUME GENERATION

1. **PACK**: We use Hertzian contact and pack a specific size distributions of filler particles into a representative volume using the open source code “Large-scale Atomic/Molecular Massively Parallel Simulator” (LAMMPS, www.lammps.org)
   1. 1024 Monodisperse, 64% Volume Fraction will be studied over 5 statistically equivalent RVEs
2. **INSERT**: A smaller set of (fundamental) particles, mass *m* and diameter *d* are inserted into an equally sized RVE from step 1.
3. **CONNECT**: All smaller particles within 1.5*d* of their centers are connected with a spring
4. **ASSIGN**: Each spring behavior is assigned based on which material each particle resides from 1. include a specific bonds across material change interfaces

Different FILLER diameter distributions





MESOMECHANICS APPROACH: MACROSCOPIC QUANTITIES OF INTEREST

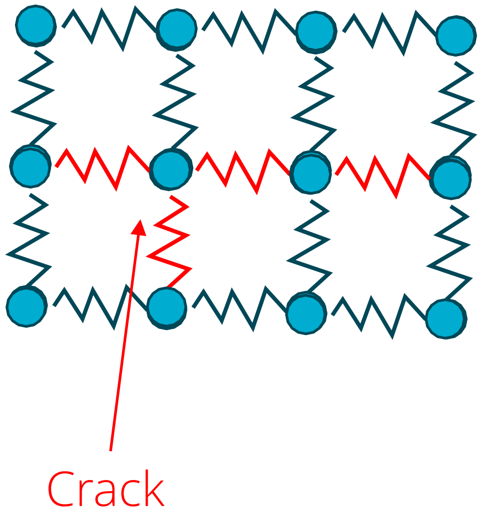
Homogenized logarithmic strain from the time history of the box dimensions and 3 isotropic invariants:

$$L_\alpha(t+\Delta t) = L_\alpha(D(t) + \Delta t \dot{\epsilon}_\alpha)$$

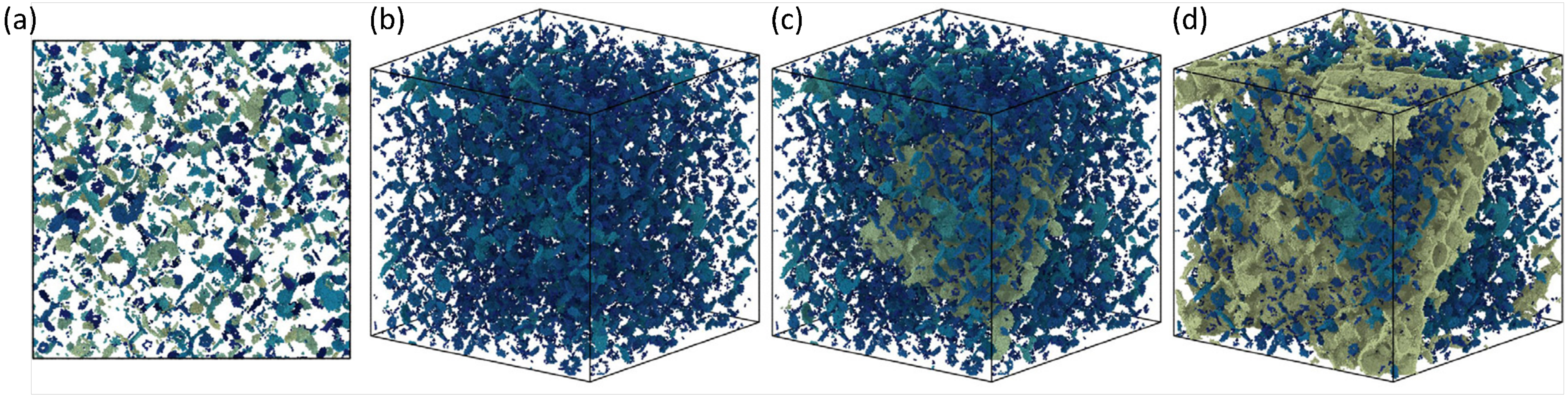
Homogenized Cauchy Stress from the Virial theorem (Subramanyian, *IJSS*, 2008) and associated 3 Isotropic Invariants:

$$P = -\mathrm{Tr}(\sigma)/3 \qquad s_{ij} = \sigma_{ij} - P\delta_{ij}$$

$$\sigma_{ij}^V = \frac{1}{V}\sum_\alpha \left[\frac{1}{2}\sum_{\beta=1}^{N}(R_i^\beta - R_i^\alpha)F_j^{\alpha\beta} - n_d k_B T^\alpha\right] \qquad J_2 = \frac{1}{2}\left(s_1^2 + s_2^2 + s_3^2\right) \quad J_3 = \frac{1}{3}\left(s_1^3 + s_2^3 + s_3^3\right)$$



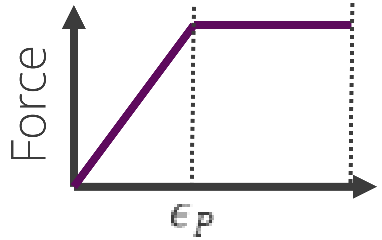
Identification of distinct cracks as clusters of broken bonds with more than one nearest neighbor



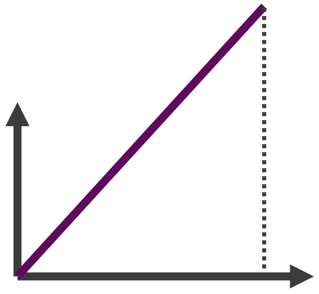
IDEALIZED MATERIAL MODELS

Consider 2 axes:

1. T_{glass} of binder relative to operating temp T
   I.e. adjust plasticity of binder bonds



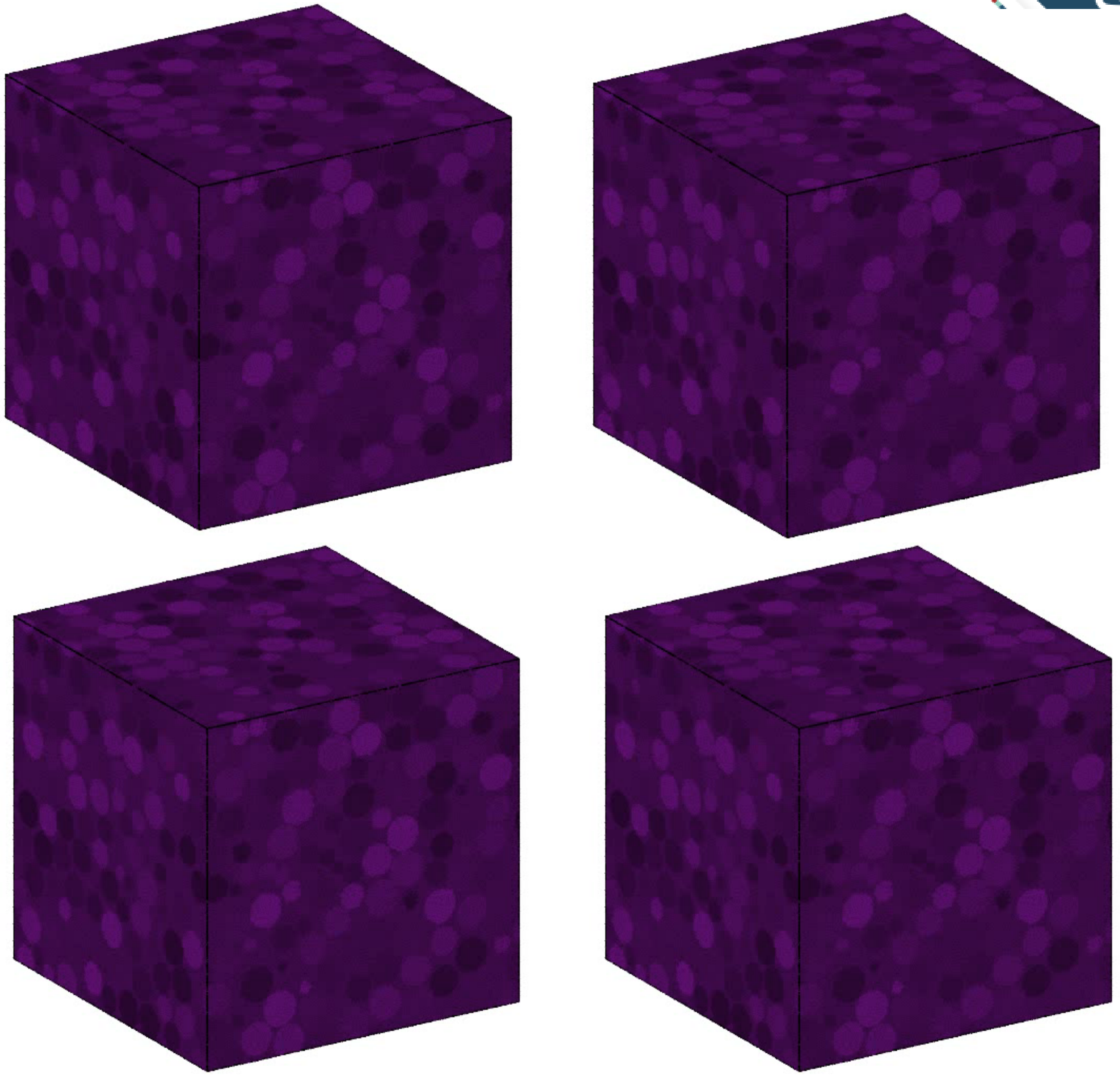
Vs.



1. Interfacial adhesion strength
   I.e. adjust strength of binder-grain bonds

Elastic, $T_{glass} < T$

Elastic-Plastic, $T_{glass} > T$



Perfectly adhered

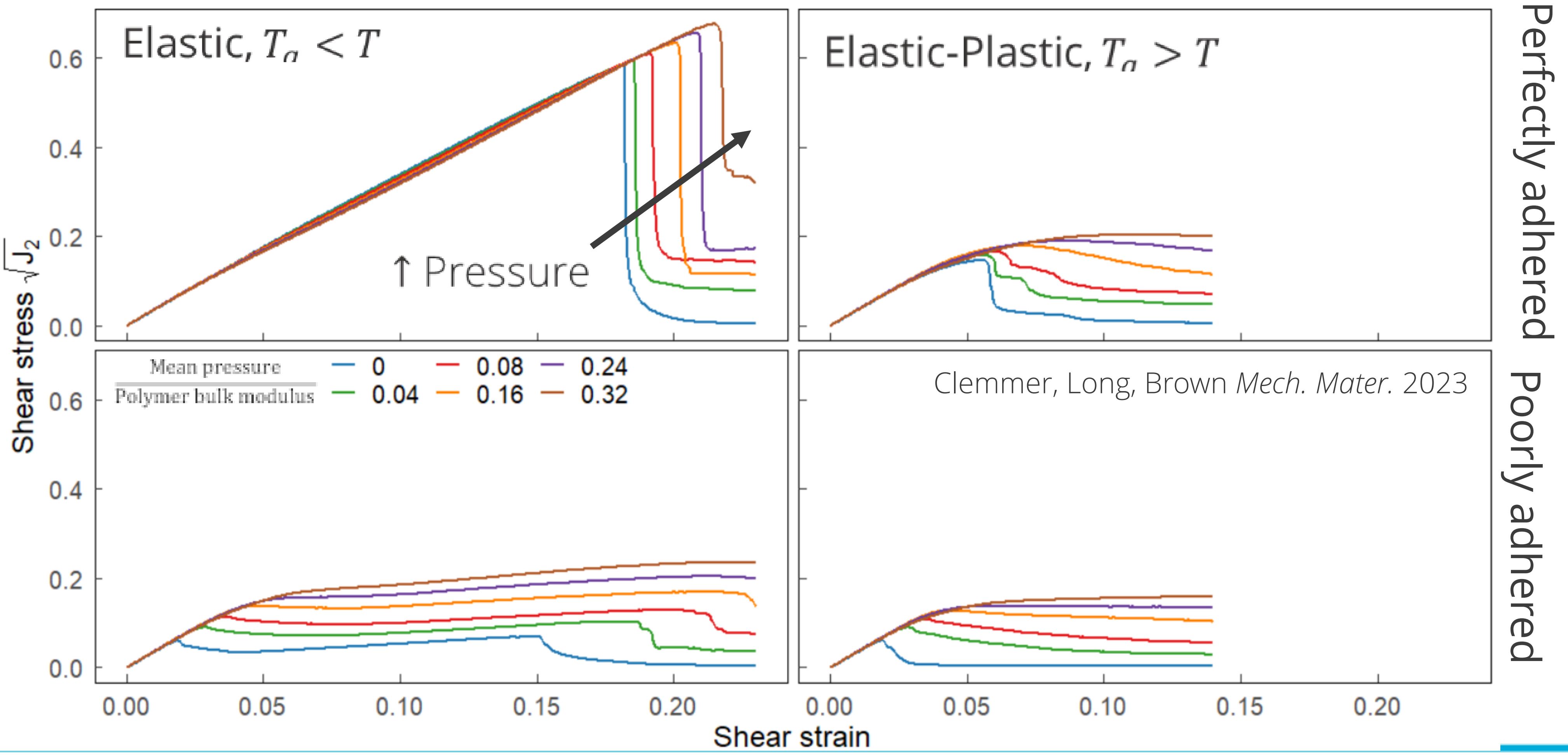
Poorly adhered

Color indicates # of broken bonds, ~Damage

PRESSURE DEPENDENT MECHANICAL RESPONSE

Spherical grain systems, failure under triaxial compression at constant mean pressure

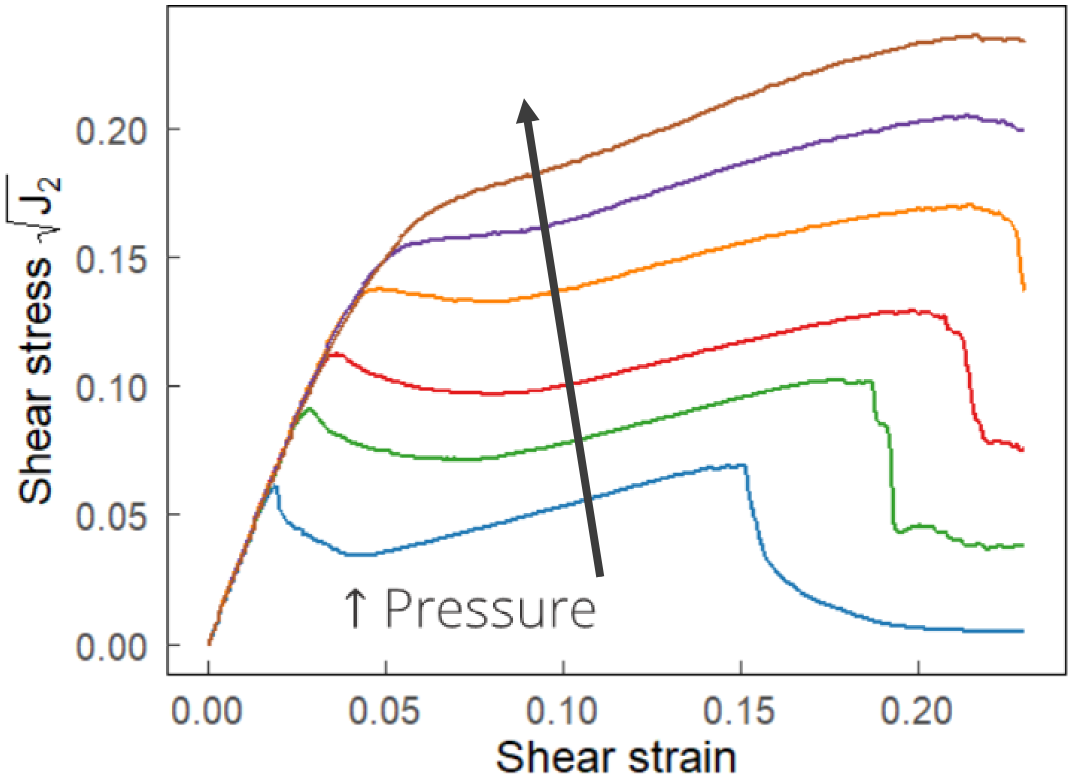
How do we explain/model how this complex behavior is governed by material inputs?



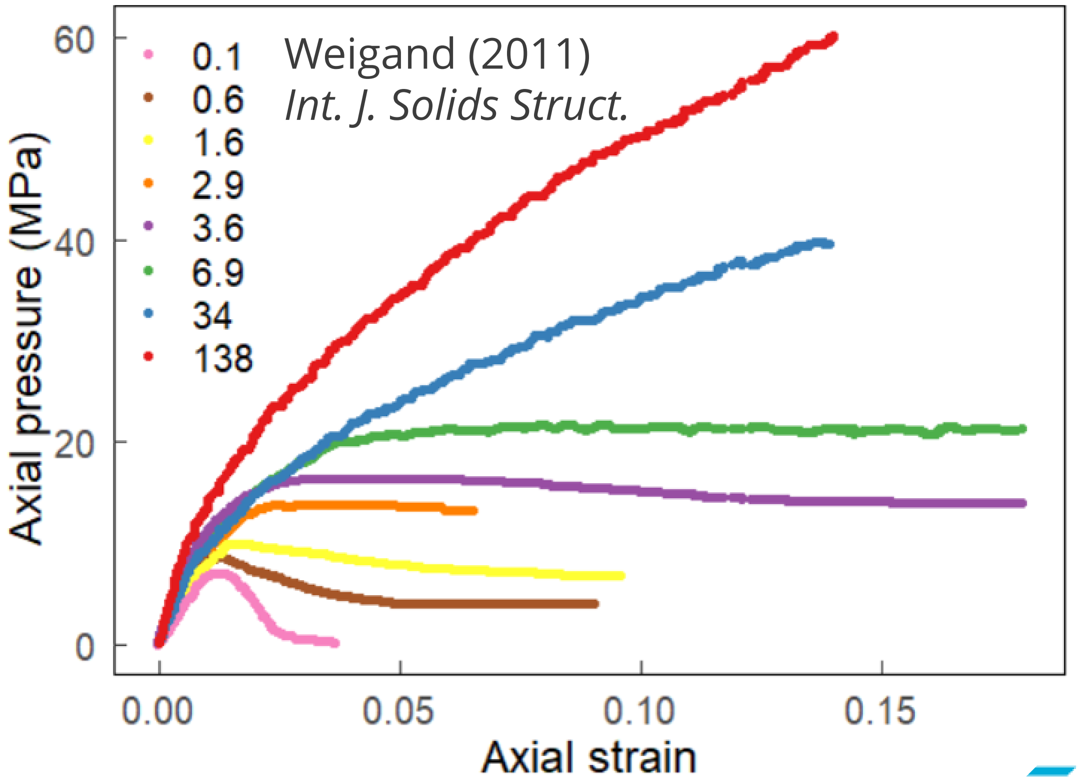
EXPERIMENTALLY CONSISTENT RESULTS

In low$-T_{glass}$ limit, capture disappearance of strain-softening regime with increasing pressure

Elastic, $T_g < T$, poorly adhered

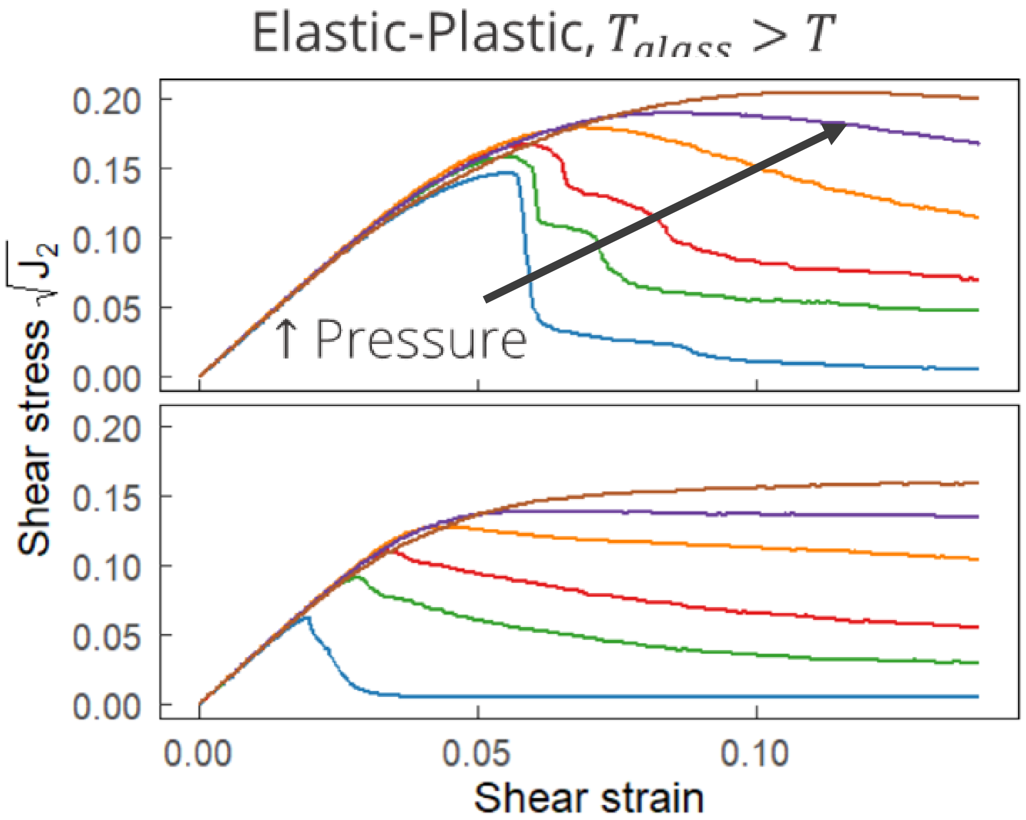


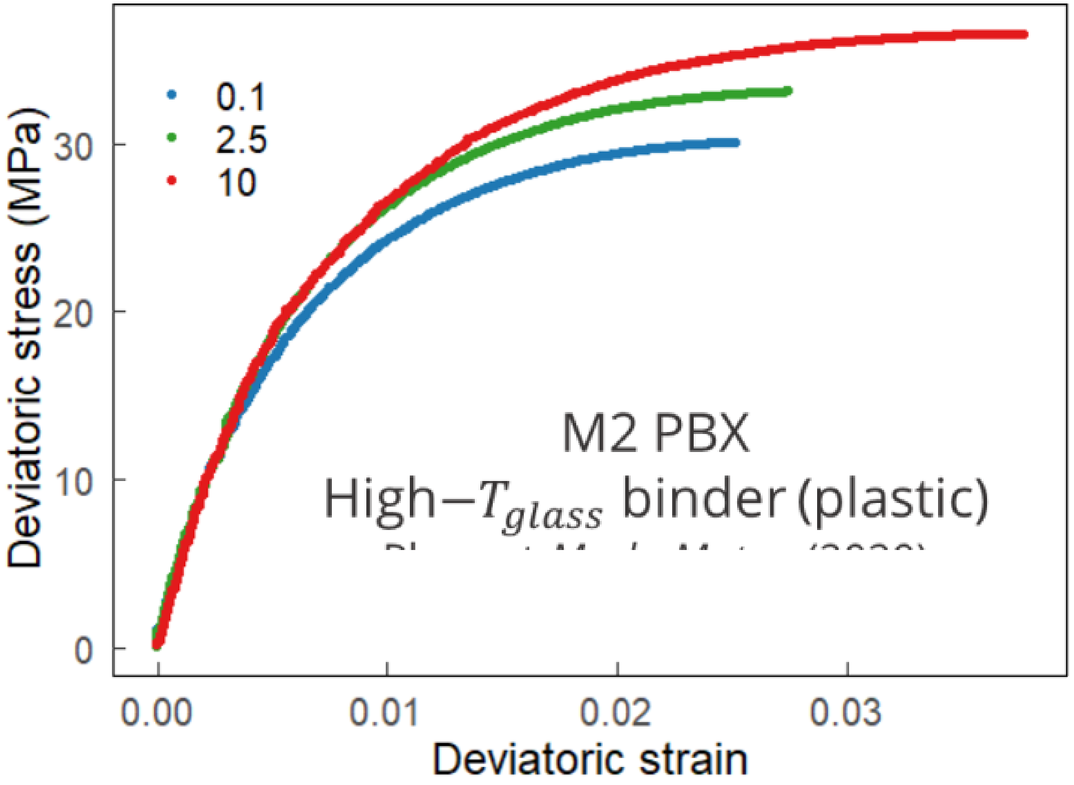
EDC PBX, low$-T_{glass}$ binder (elastic)



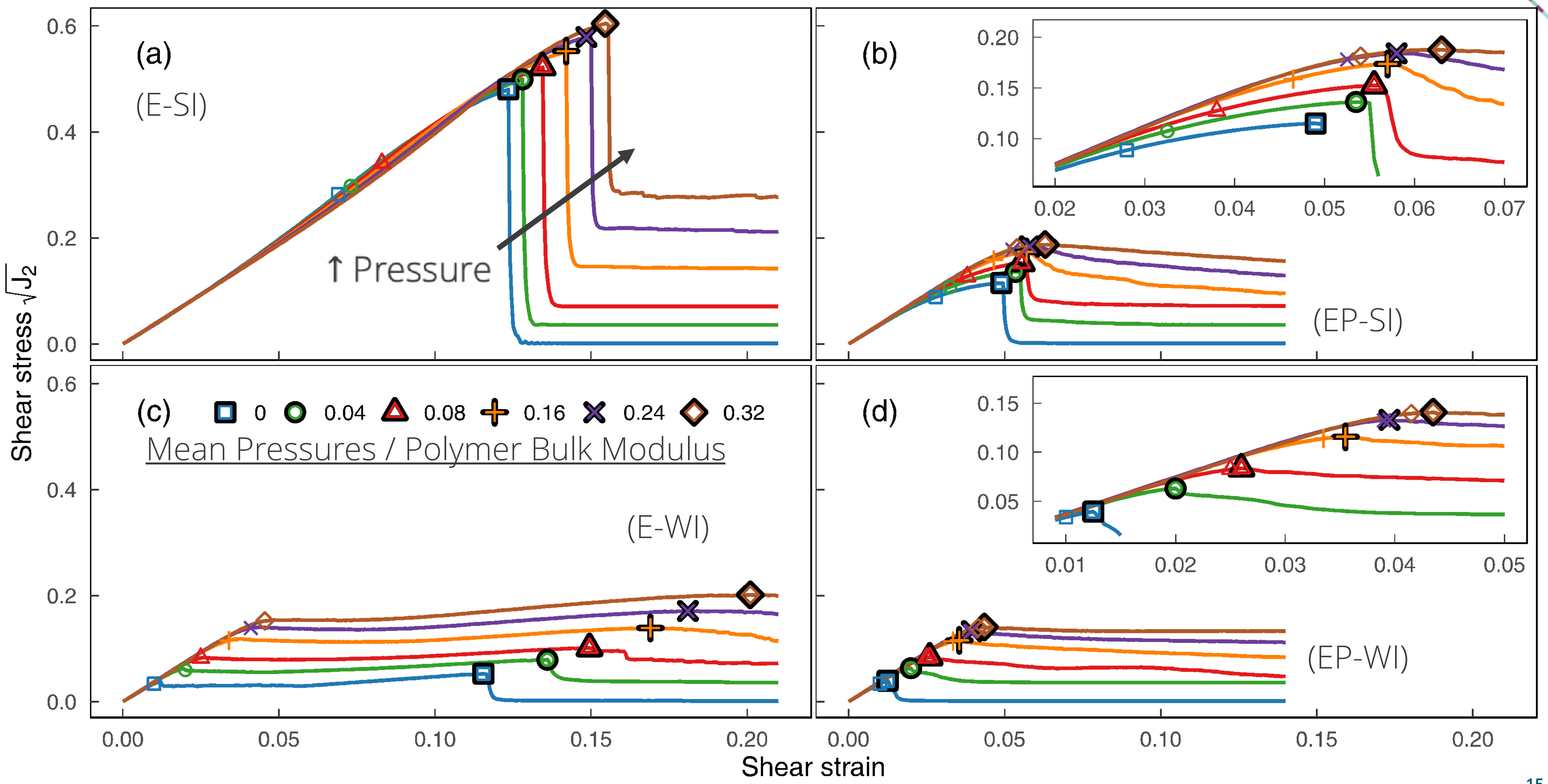
EXPERIMENTALLY CONSISTENT RESULTS

In high$-T_{glass}$ limit, identify similar shift in yield/failure with increasing pressure, also capture transition from small-strain compressive volume strain to dilation post yield (not shown)



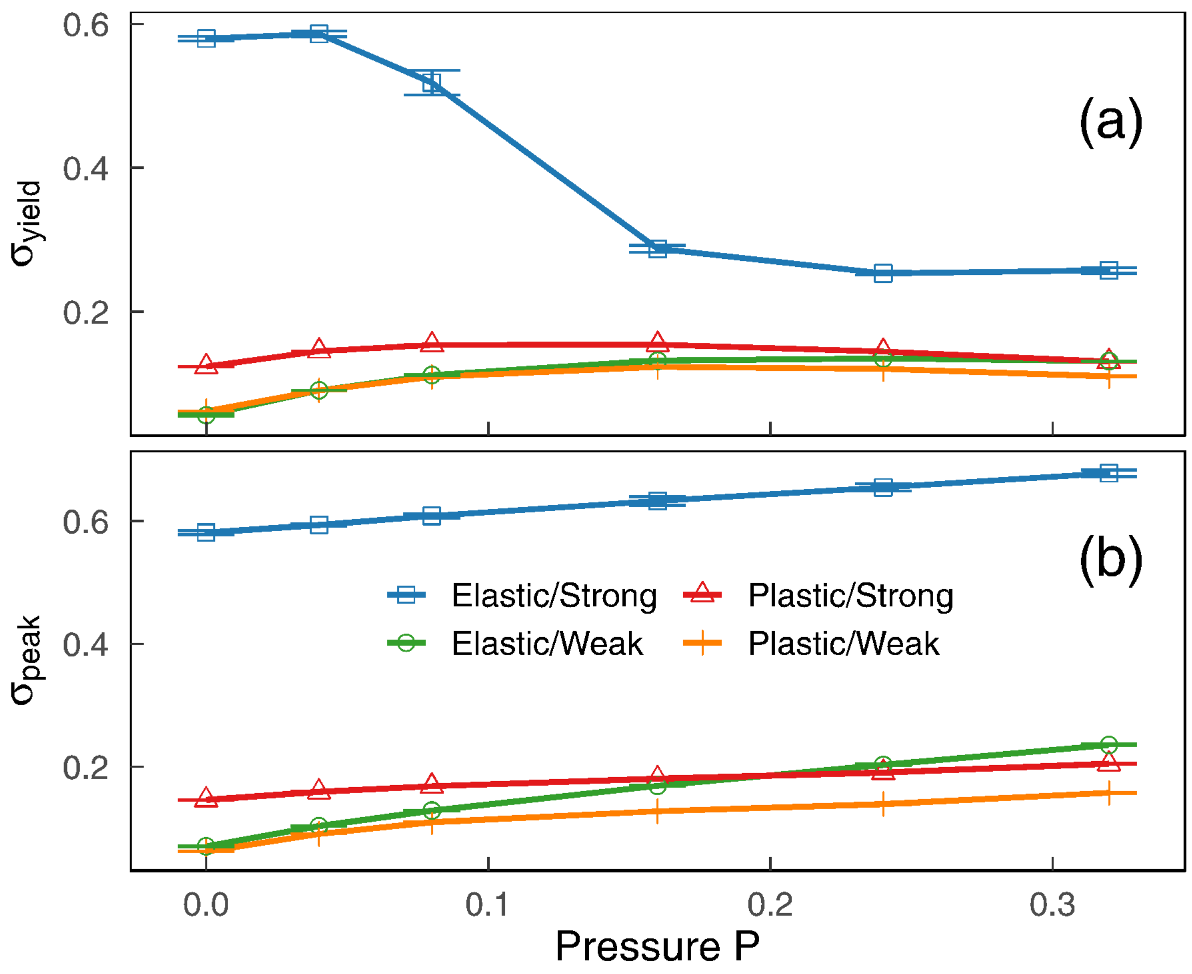


RESULTS: TRIAXIAL EXTENSION AXIAL STRESS FOR THE 4 CASES

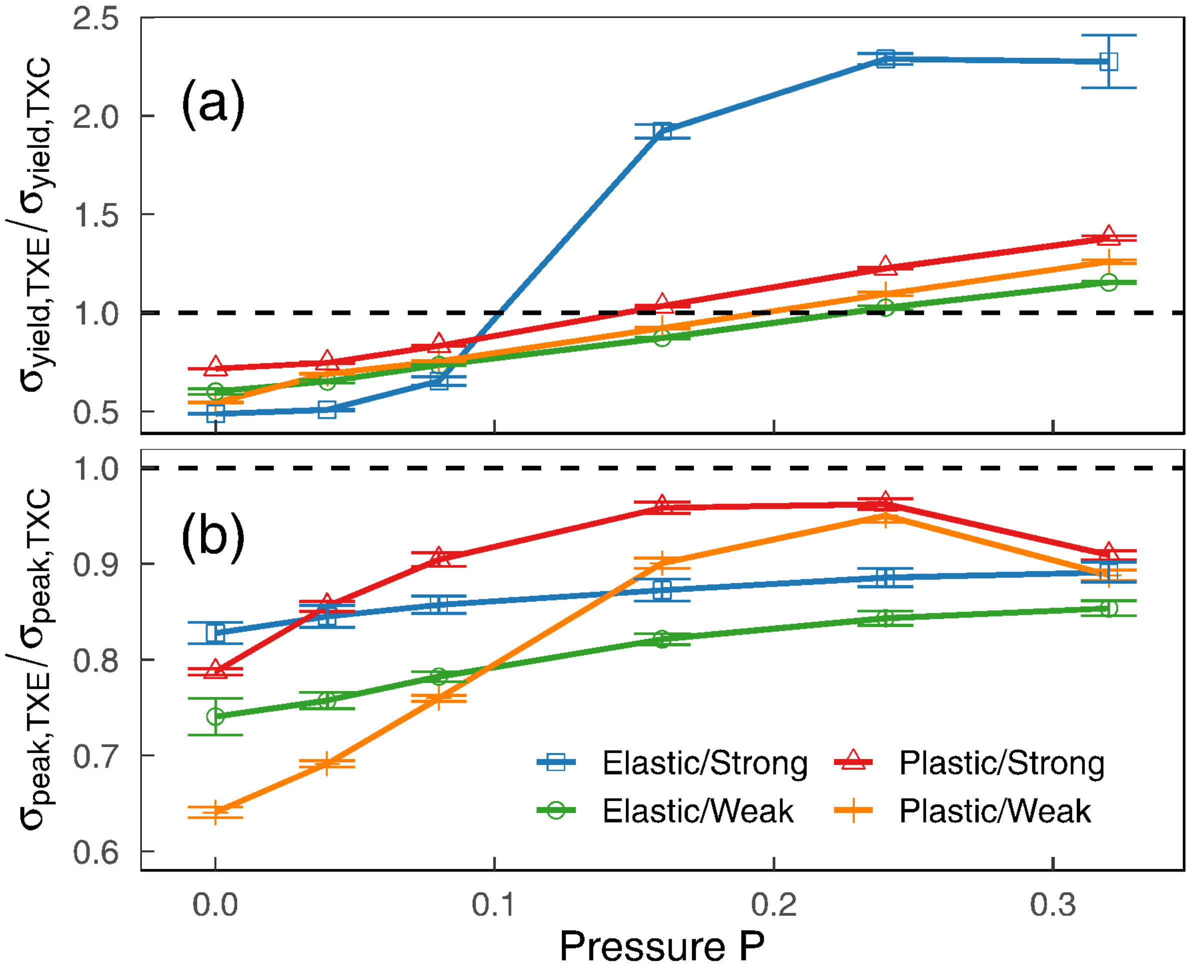


OBSERVATIONS

Triaxial Compression (TXC)



Triaxial Extension (TXE) Normalized By Triaxial Compression

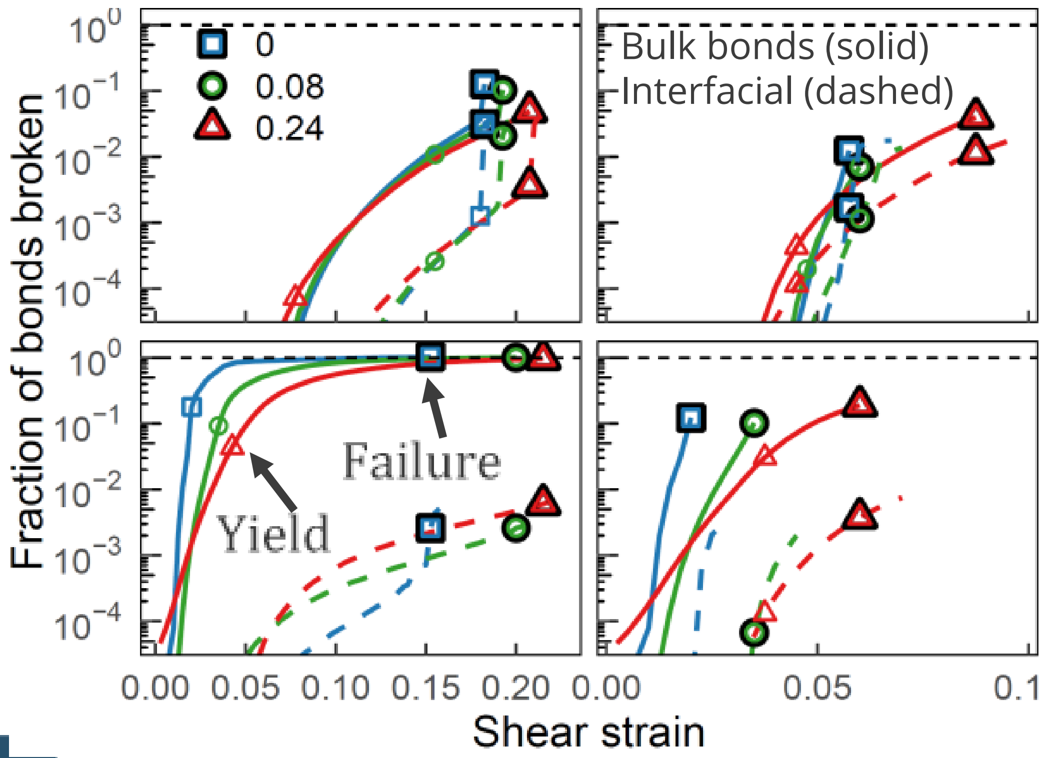


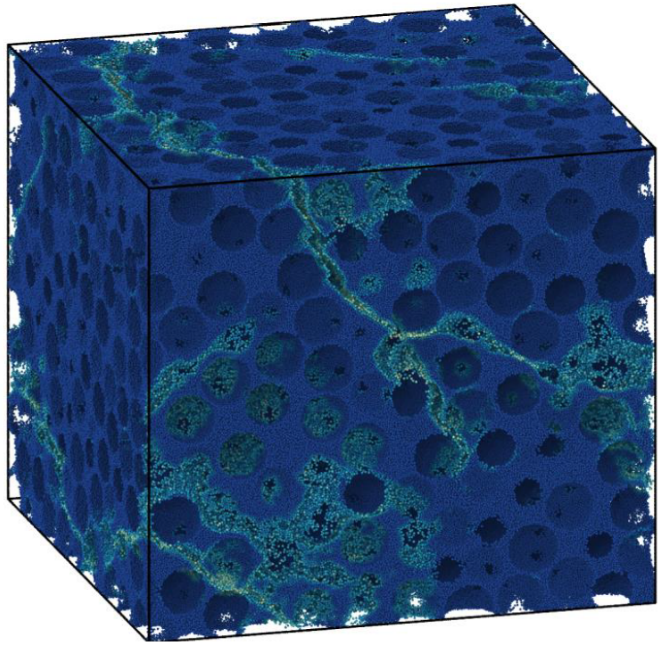
TRACKING MICROSCOPIC ACTIVATION OF INELASTIC MECHANISMS

Demonstrated method reproduces key experimental findings

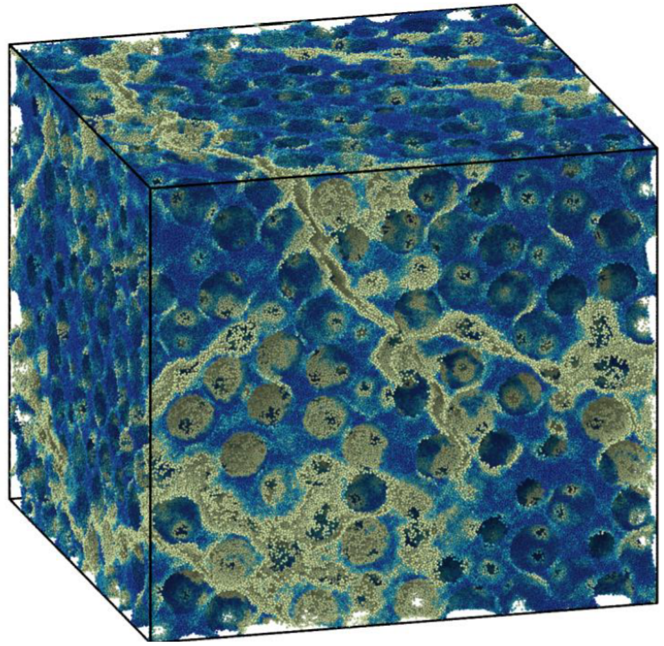
Use full access of microscopic dynamics to identify inelastic origin of key features/transitions on macroscopic stress strain curves

e.g. delamination of binder-grain interfaces leads to strain softening regime at low pressures





Broken bonds



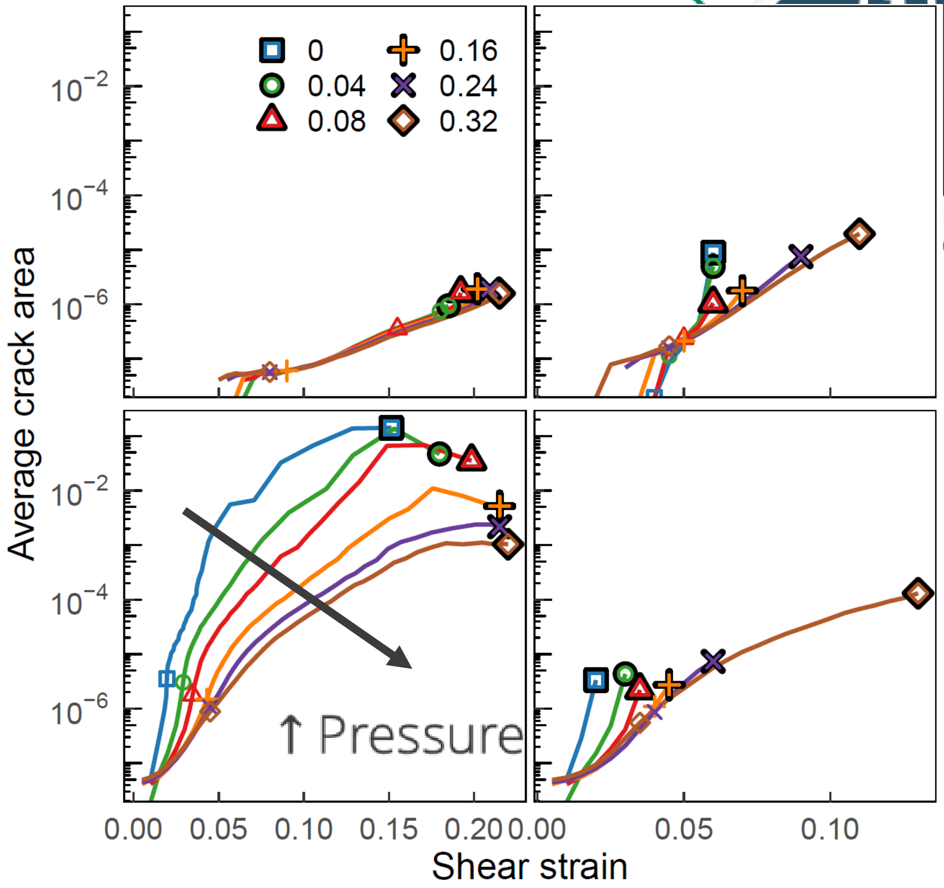
Plastically activation

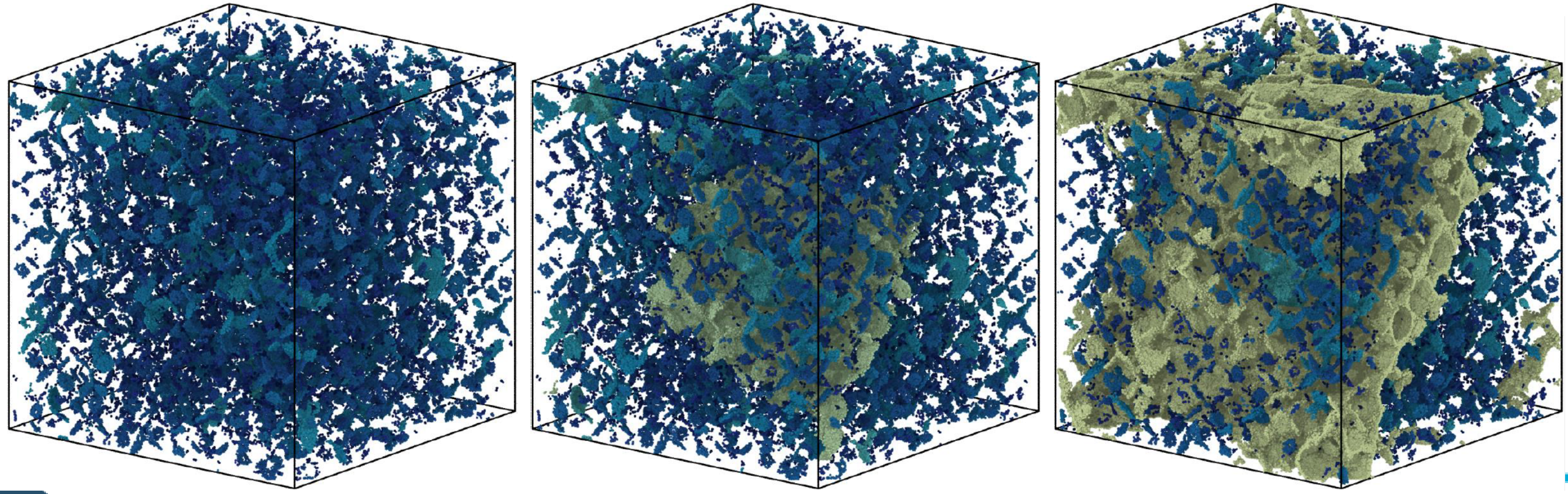
EXPLORING MESOSCOPIC CRACK DYNAMICS

Continuum modeling of PBXs utilizes damage metrics to capture degradation in mechanical performance with deformation history – e.g. cracking

Long term goal to evaluate accuracy of such metrics by tracking evolution of crack-size distribution

Find standard results, pressure suppresses crack growth





To identify distinct cracks, spatially cluster locations of broken bonds

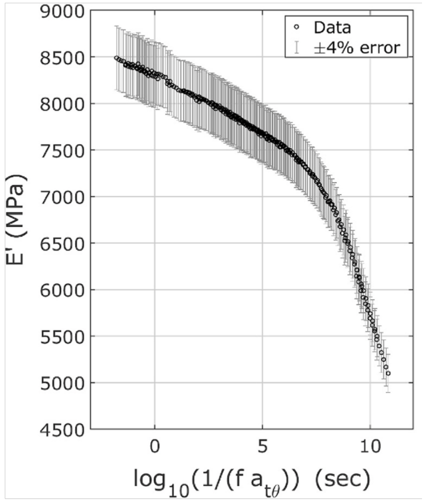
Perform at regular strain intervals

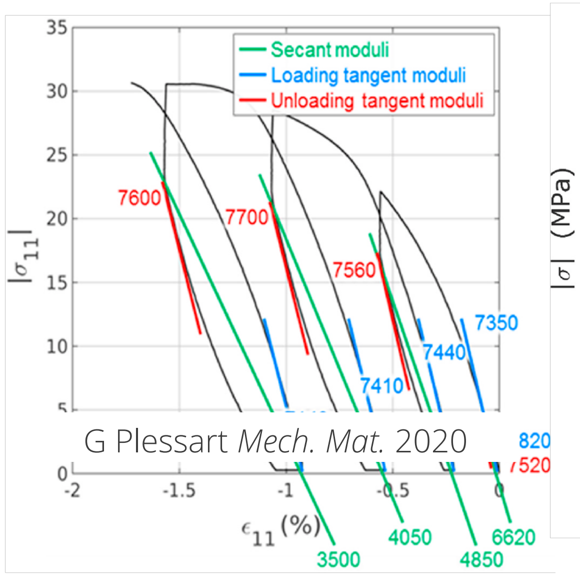
OBSERVATIONS AND TAKEAWAYS

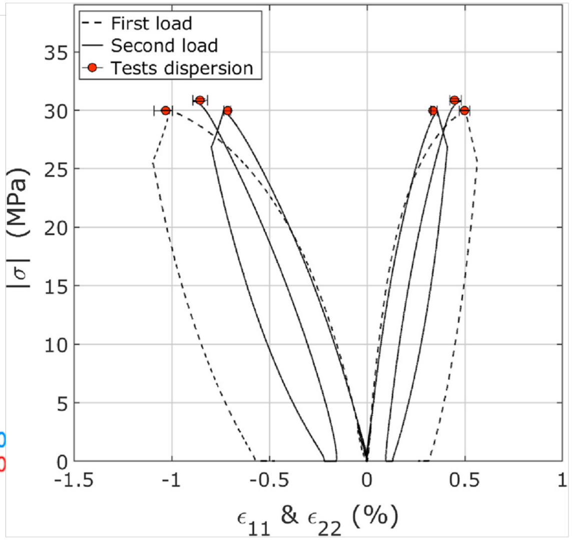
- Using a bonded particle model gave flexibility to easily study abstract changes in material properties or loading conditions through simple changes in the bond force model.
- Simulations were robust and able to run (in displacement control) through localization
- Two types of bond models were used to characterize elastic and elastic-plastic interactions, which allowed 4 types of limiting cases to be considered:
- Increasing confining pressure increases strength in all cases
- A significant lode angle dependence is observed vs. pressure by comparing TXE and TXC
  - Drucker-Prager does not describe the data
  - Cracking is not isotropic and indicates anisotropic damage is occurring

FUTURE WORK

- Investigate the role of cracking and developed damage anisotropy
- Explore anisotropic filler packings
- Reduce data to analytic constitutive forms
- Develop a micromechanics informed constitutive model

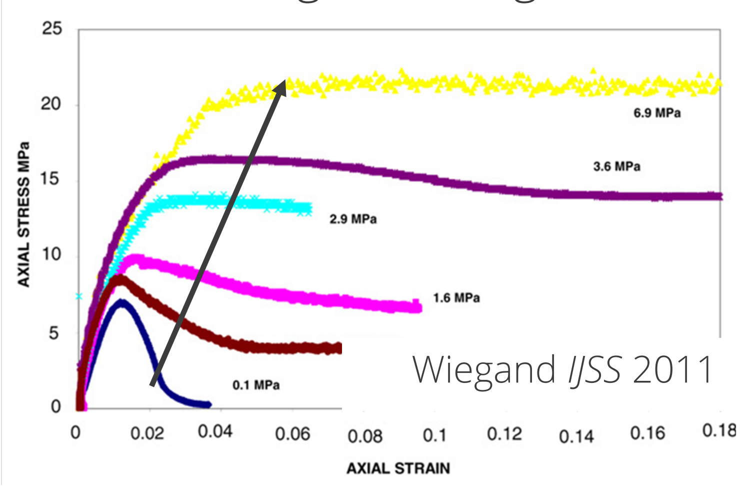






G Plessart *Mech. Mat.* 2020

Increasing Confining Pressure



Wiegand *IJSS* 2011

7/28/2023

References:

QUESTIONS

EXTRAS

MESOMECHANICS APPROACH: TRIAXIAL COMPRESSION BOUNDARY VALUE PROBLEM

- Kinematically homogenous boundary conditions applied
- Linear mean pressure ramp
  - A spherical motion is applied to maintain achieve the target mean pressure with time
- Linear deviatoric strain ramp
- Triaxial compression and extension are performed under different mean pressure loadings

Increment in spherical strain to achieve the target pressure

$$D(t) = \left(1 + c_g[P(t) - P_T]\Delta t\right)^{1/3}$$

Incremental change in length of the "α" direction (spherical + deviatoric)

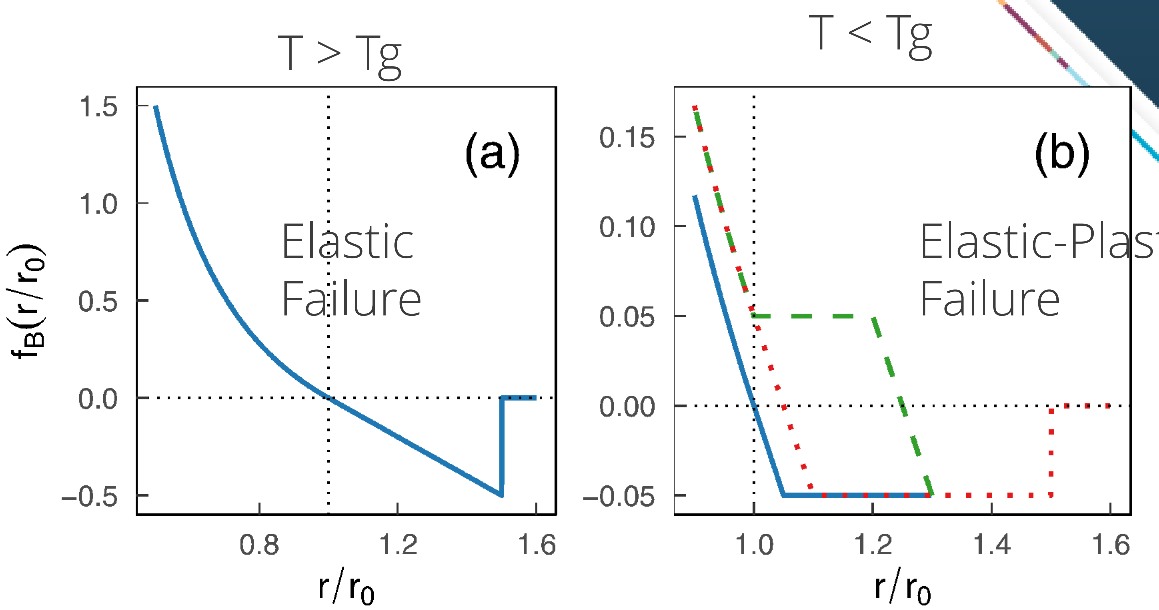
$$L_\alpha(t + \Delta t) = L_\alpha(D(t) + \Delta t\dot{\epsilon}_\alpha)$$

Length of the "x, y, or z" cube edge

MESOMECHANICS APPROACH: MECHANICS OF A BREAKABLE SPRING NETWORK WITH TUNABLE CONSTITUTIVE RESPONSES FOR EACH TYPE OF INTERACTION

DEFINE SPRING CONSTANTS

- Stiff, elastic response for the filler
- Elastic-failure response for the polymer above Tg
- Elastic-perfectly plastic failure response for a polymer below Tg
- Interfacial bonds are given an elastic-failure response different from the polymer-polymer interactions



PARAMETRIZED TRIAXIAL COMPRESSION PROTOCOL

- We use the “Bonded Particle Method”, a package in LAMMPS ([1-2]) to explicitly time integrate the system’s momentum balance
- Periodic boundary conditions are applied, and the system is subjected to a linear mean pressure ramp
- An additional deviatoric strain rate is prescribed differentiating the “z” from the “x” and “y” directions

Increment in spherical strain to achieve the target pressure

$$D(t) = \left(1 + c_g[P(t) - P_T]\Delta t\right)^{1/3}$$

Incremental change in length of the “α” direction (spherical + deviatoric)

$$L_\alpha(t + \Delta t) = L_\alpha(D(t) + \Delta t\dot{\epsilon}_\alpha)$$

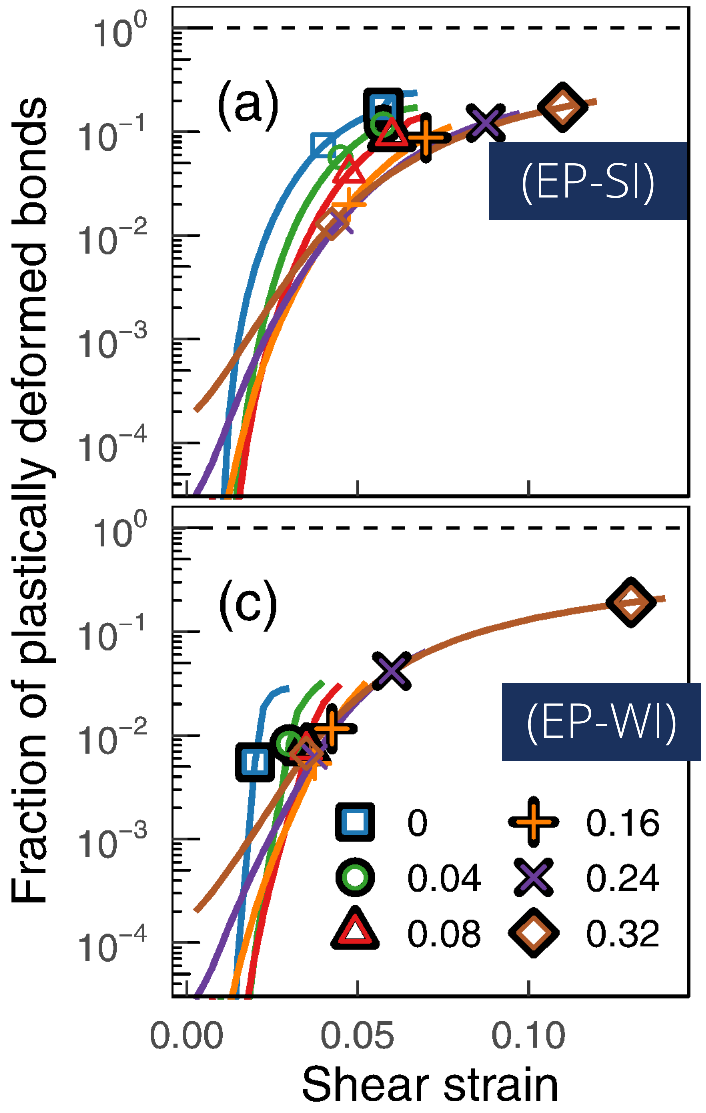
4 LIMITING CASES USING 64% VOLUME FILL BI-DISPERSE RVES

	T > Tg	T < Tg
Strong Adhesion	Elastic Binder, Strong Interface (E-SI)	Elastic-Plastic Binder, Strong Interface (EP-SI)
Poor Adhesion	Elastic Binder, Weak Interface (E-WI)	Elastic-Plastic Binder, Weak Interface (EP-WI)

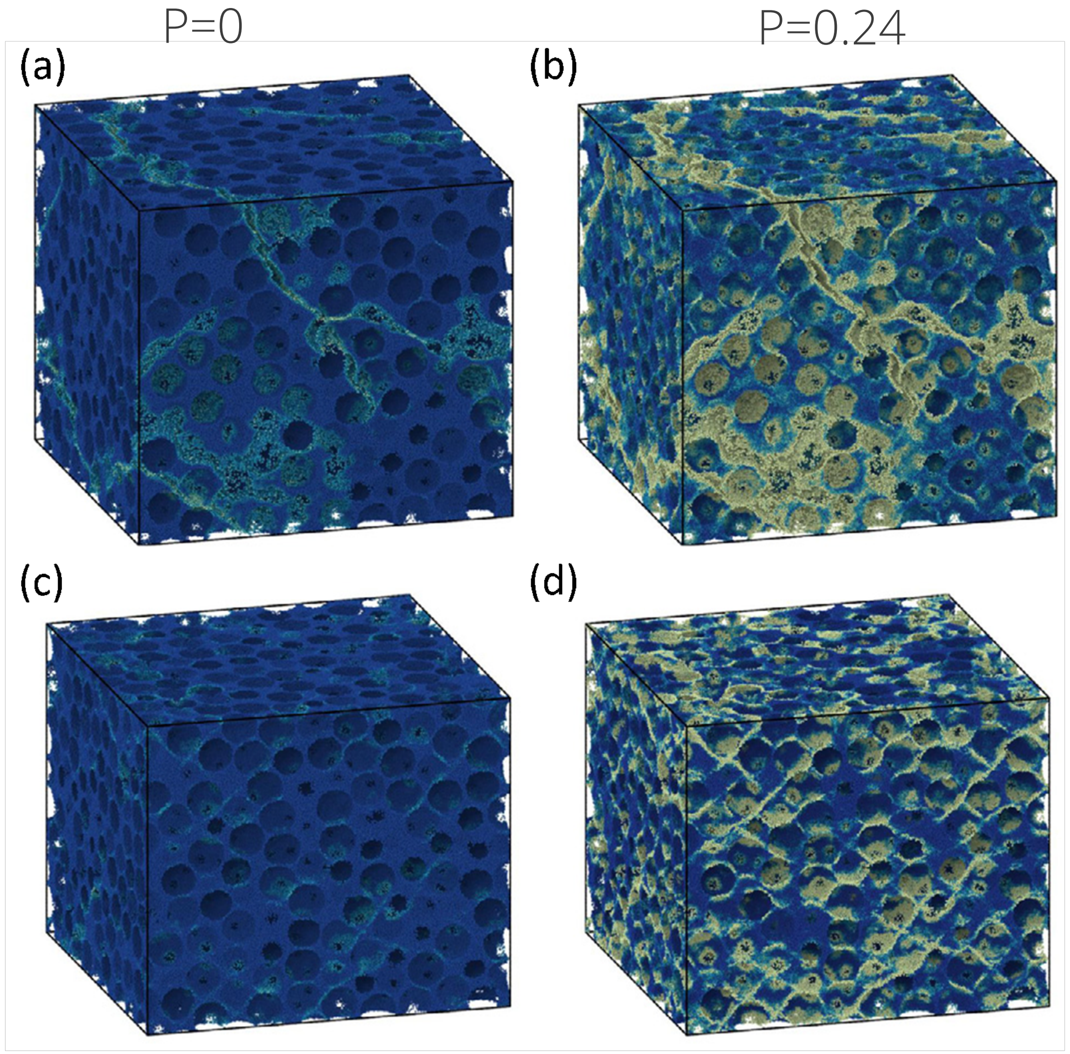
BOND INTERACTION PARAMETERS FOR THE FOUR LIMITING CASES

Models \ Bond Parameter	Filler-Filler Stiffness	Filler-Filler Strain to Failure	Polymer-Polymer Stiffness	Polymer-Polymer Strain to Failure	Polymer-Polymer Yield Force	Filler-Polymer Stiffness	Filler-Polymer Strain to Failure
Elastic Polymer--Strong Adhesion	10	100	1	0.5	0.5	1	0.5
Elastic Polymer--Weak Adhesion	10	100	1	0.5	0.5	1	0.05
Elastic Plastic Polymer--Strong Adhesion	10	100	1	0.5	0.05	1	0.5
Elastic Plastic Polymer--Weak Adhesion	10	1000	1	0.5	0.05	1	0.05

RESULTS: FOCUS ON THE HIGH TG ELASTIC-PLASTIC BINDER



Elastic-Plastic Binder with a strong interface. Bonds colored by how many neighbors are broken



Panels just after failure at global 0.5% shear strain

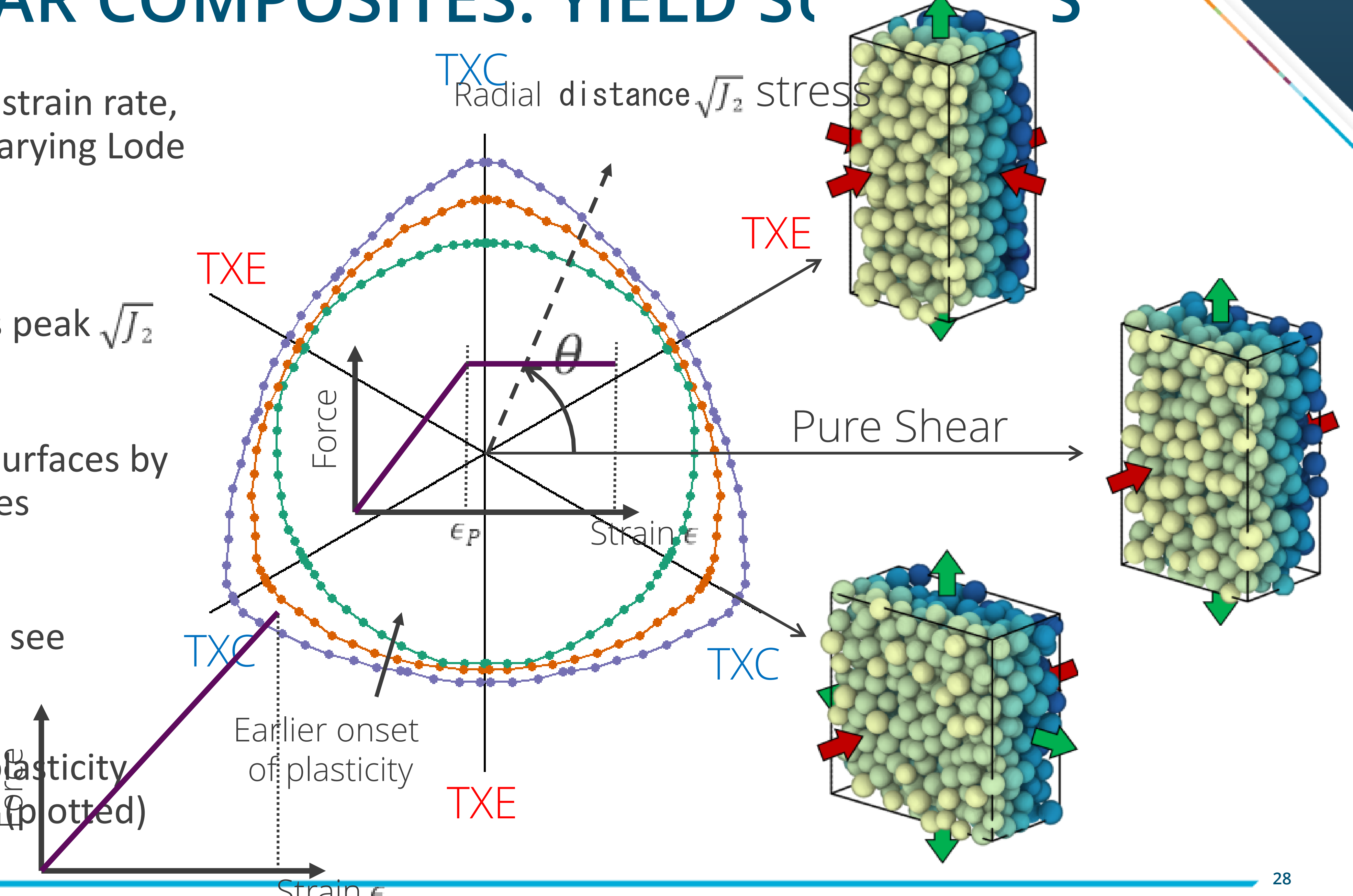
GRANULAR COMPOSITES: YIELD SURFACES

Deform system at fixed strain rate, constant volume, and varying Lode angle θ

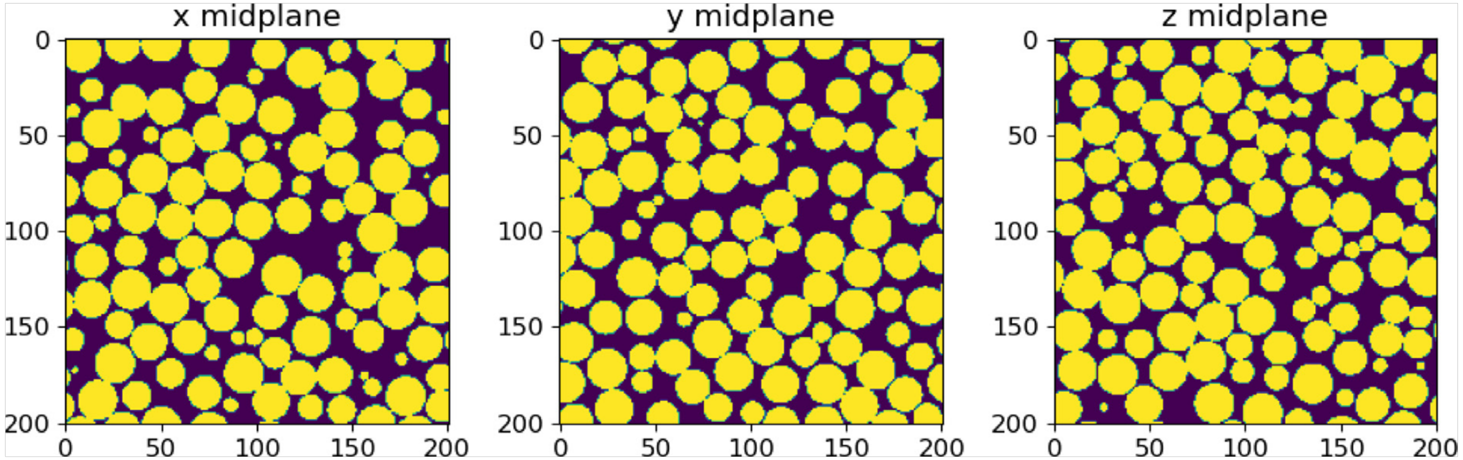
Identify failure stress as peak $\sqrt{J_2}$

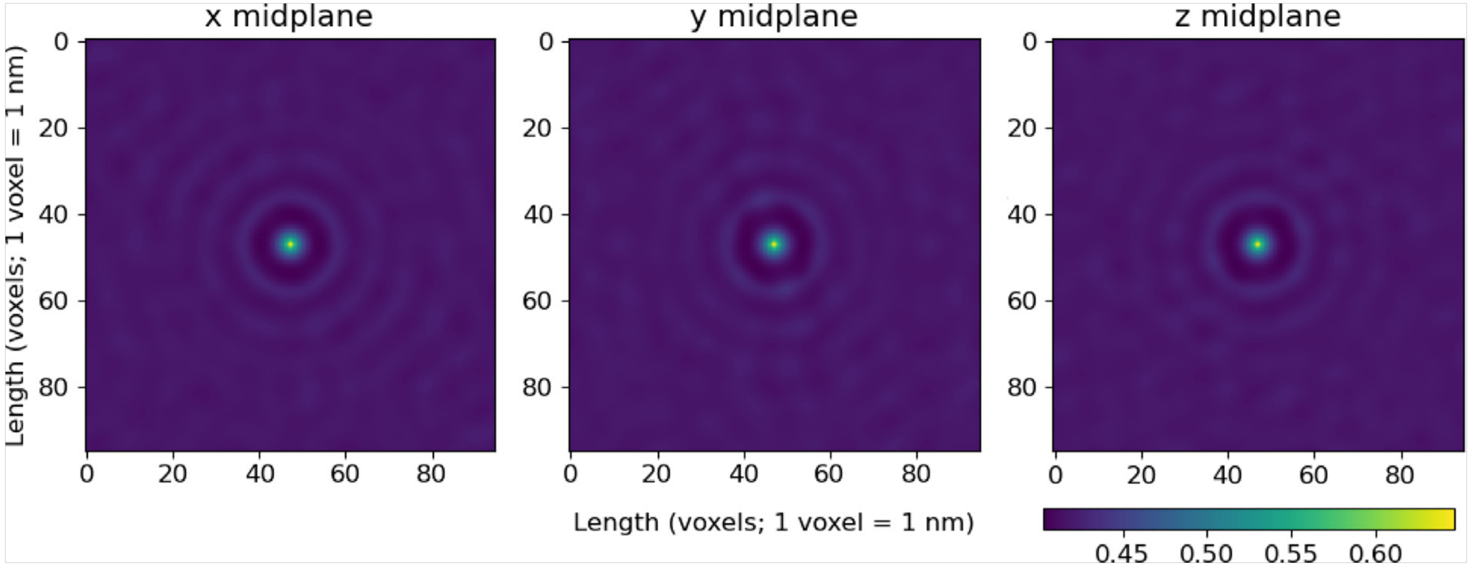
Study trends in failure surfaces by varying binder properties

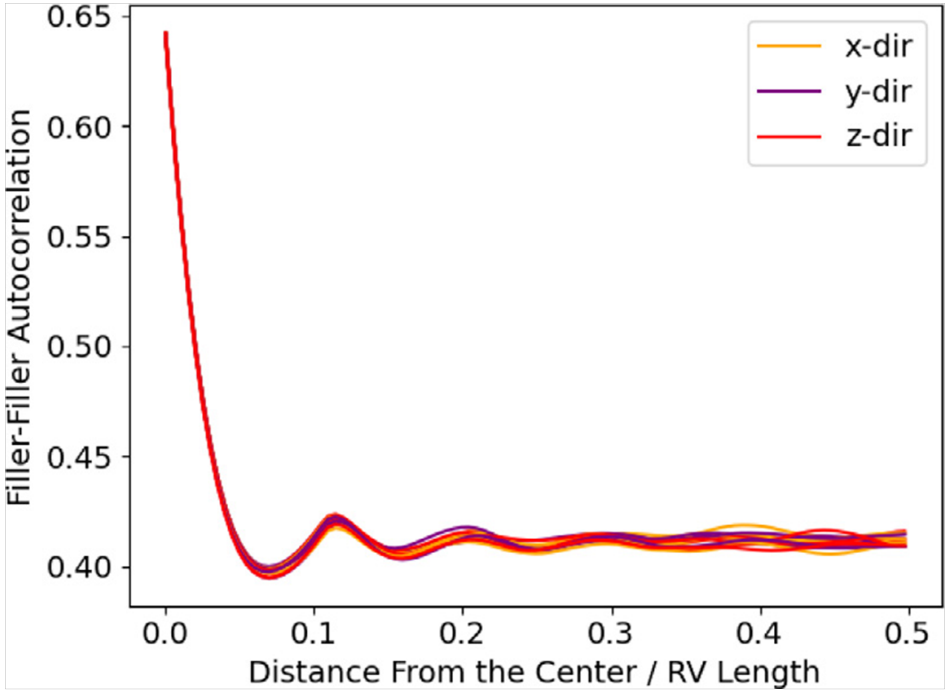
Preliminary simulations see behavior goes from Duncan-Lade-like to Drucker-Prager-like as plasticity occurs at a lower strain (plotted)



DUE DILIGENCE THAT THE MICROSTRUCTURES ARE ISOTROPIC

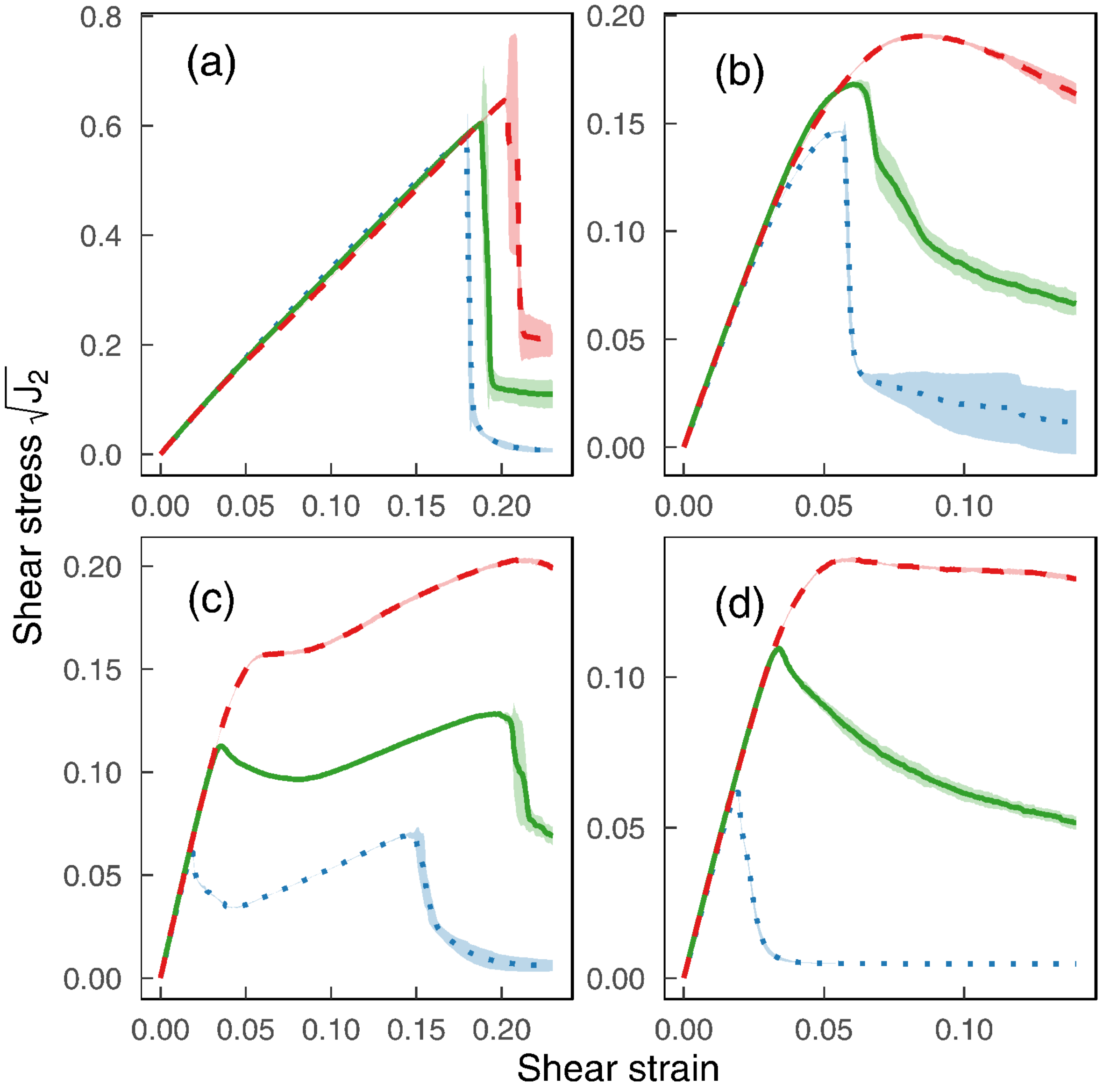






DUE DILIGENCE THAT THE RVES ARE SIZE CONVERGED

- Five statistically equivalent microstructures were generated
- The four limiting cases were considered at normalized pressures of 0, 0.08, and 0.24
- Variability is only observed in the Elastic-Weak Interface case



ABSTRACT—TALK 936, MESOMECHANICS OF HIGHLY FILLED PARTICLE REINFORCED COMPOSITES USING A BONDED PARTICLE METHOD

The mechanical behaviour of highly filled polymer composites is complex, exhibiting traits of both polymeric and granular materials with additional inelastic mechanisms introduced by interactions between the constituents. How these mechanisms interact for a given mechanical loading scenario is dependent on material constituent properties, filler volume fraction, and aspects of the mechanical loading (strain rate, ambient temperature, confinement, and sample history). From a practical standpoint, validated constitutive models are needed to predict the homogenized behaviour of highly filled particle composites in complex thermal and mechanical loading scenarios. However, development, parameterization, and validation of such constitutive models for engineering applications is impeded both by the complexity in describing the underlying and interacting inelastic mechanisms and frequently by a paucity of experimental discovery and characterization data for materials.

Fortunately, highly filled polymer composites do share many features across different material systems. The polymer binder phase is typically operating either well above or well below its glass transition, and the binder and particles phases can delaminate at the mesoscale. We seek to take advantage of these known underlying behaviours at the mesoscale to better understand the characteristic behaviours of highly filled polymer composites.

We present a multi-scale study that explores mechanical behaviour under a variety of different loading states and material design space parameters on representative volumes (RV) of highly filled particle composites. We use a minimalist bonded particle model (BPM) which represents solids as a collection of point particles connected by pairwise bonds. Different material properties such as stiffness, plasticity, and failure are captured by adjusting the functional form of bond forces. Our mesoscale modelling links material design parameters to emergent inelastic deformation mechanisms that drive the macroscale mechanical behaviour up to and including localization (deformation cracking that spans the representative volumes). We explore a variety of material design parameters, such as particulate to binder volume ratio, binder glass transition temperature, and binder-particulate interfacial failure and extract homogenized RV responses and microstructure evolution metrics. In particular, we note that the relation between the analysis of crack evolution of the RVs and bulk constitutive behaviour remains an open area of research. We summarize insights gained from these studies and their implications for macroscale constitutive modelling are discussed, including yield surface shapes, pressure dependence, damage evolution mechanisms and material failure criteria.

This work was supported by the Laboratory Directed Research and Development program at Sandia National Laboratories. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the US Department of Energy's National Nuclear Security Administration under contract DENA0003525. This abstract describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the abstract do not necessarily represent the views of the US Department of Energy or the United States Government.

20 minute time slot (15 min + 5 min Q&A)