Microstructural Characterization: Application to Granular Cohesion

IFPRI Cohesion Workshop, January 2020

Ryan C. Hurley[1]
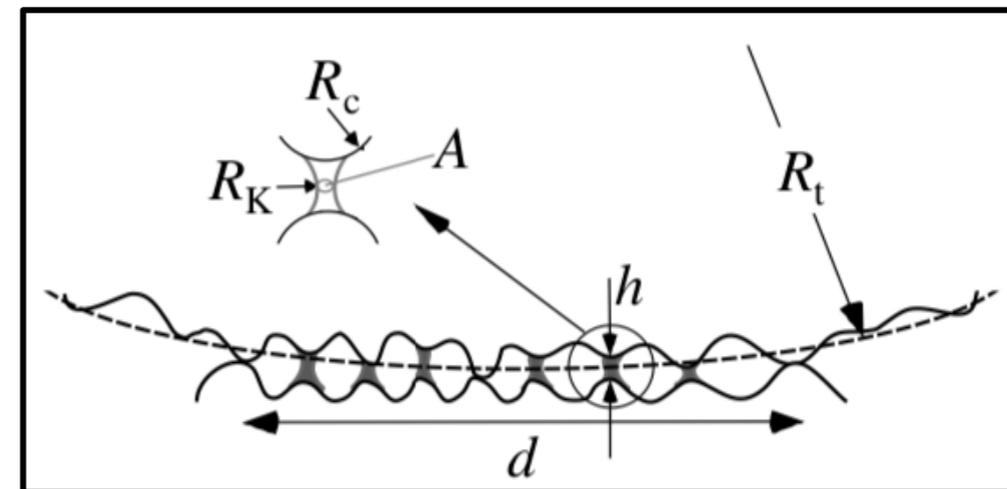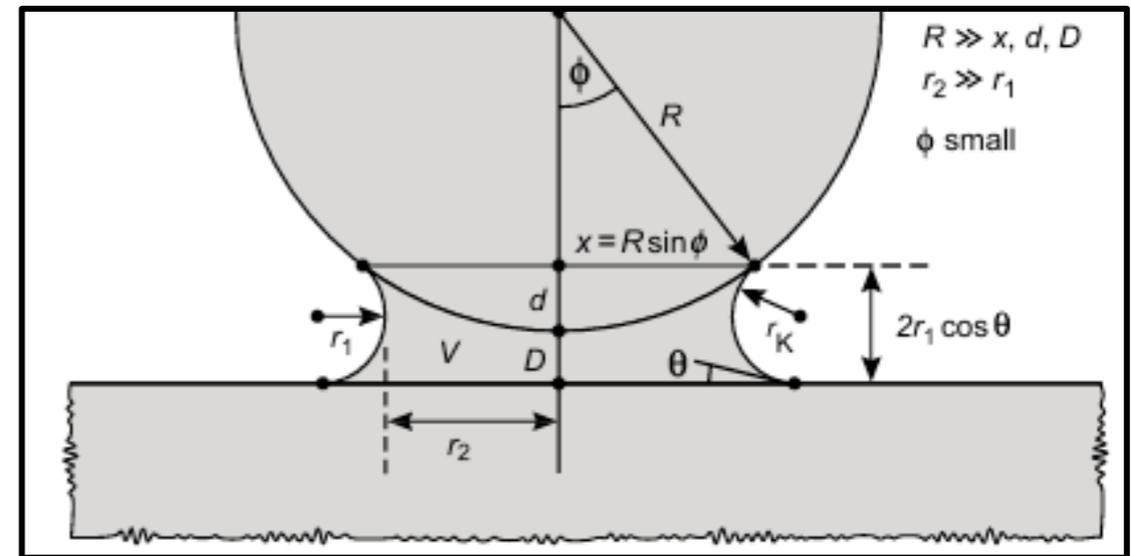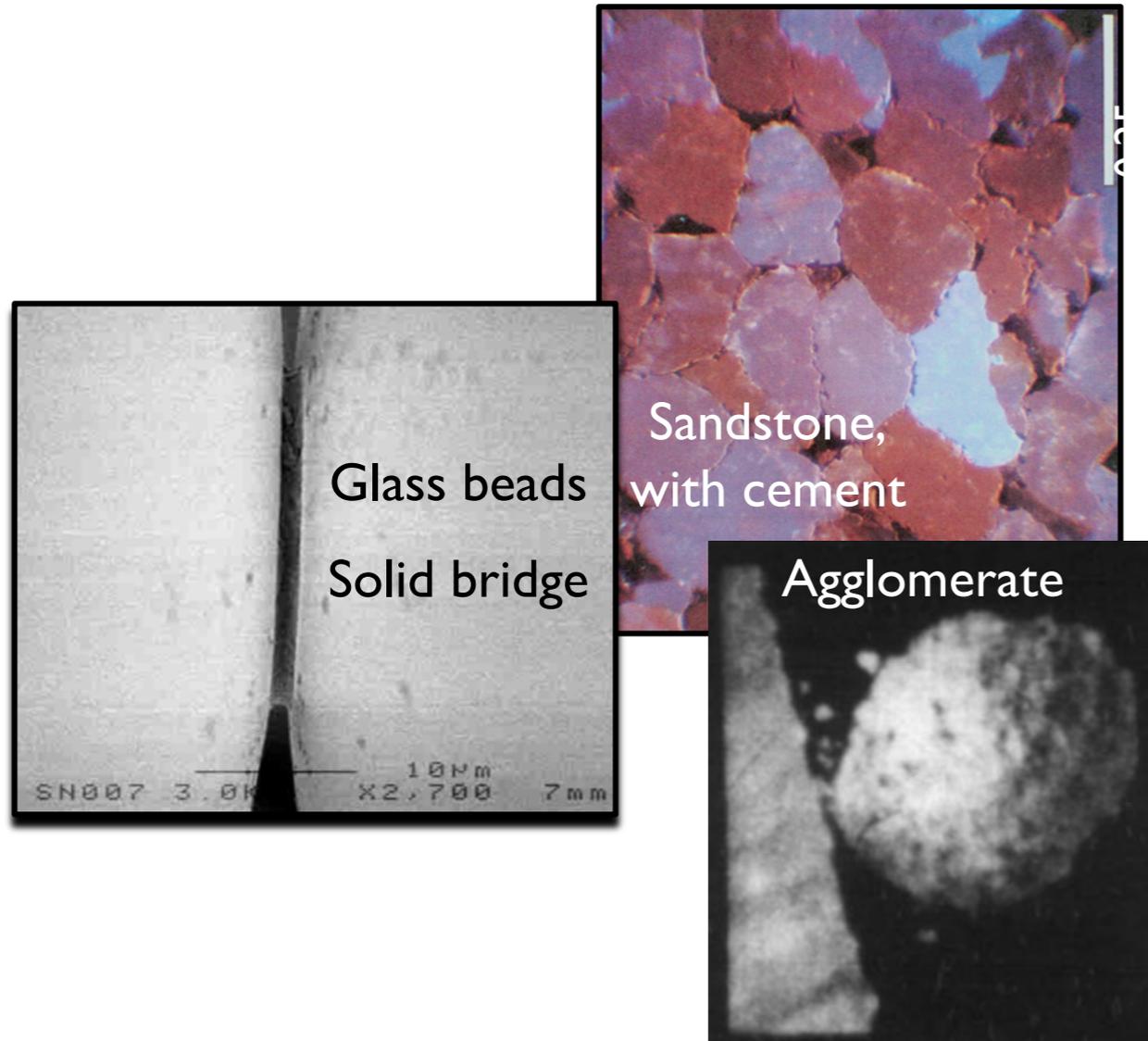
[1]Mechanical Engineering, Hopkins Extreme Materials Institute, Johns Hopkins University

(1) Cohesion mechanisms of interest? (2) What are our relevant microscale characterization techniques at bulk, particle levels?

Sandstone, with cement

Glass beads

Solid bridge

Agglomerate

Cementation: geology (diagnesis), engineering (bio-cementing), tableting
[Houseknecht, *AAPG*, 1987.; Olivi-Tran, *et al., Eur. Phys. J. B*, 2002.; Mullier, *et al., Pow. Tech.,* 1991.]
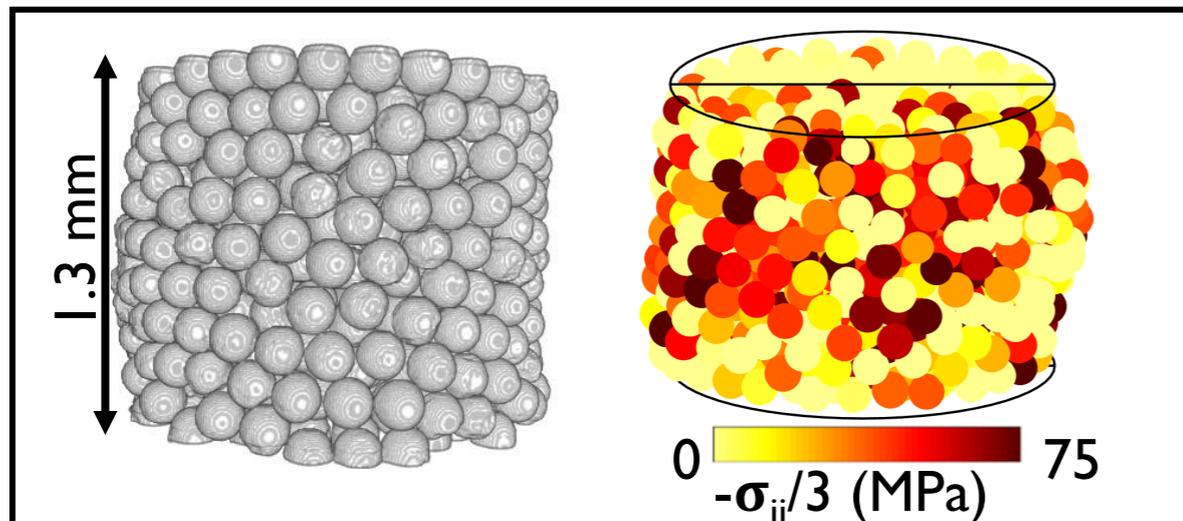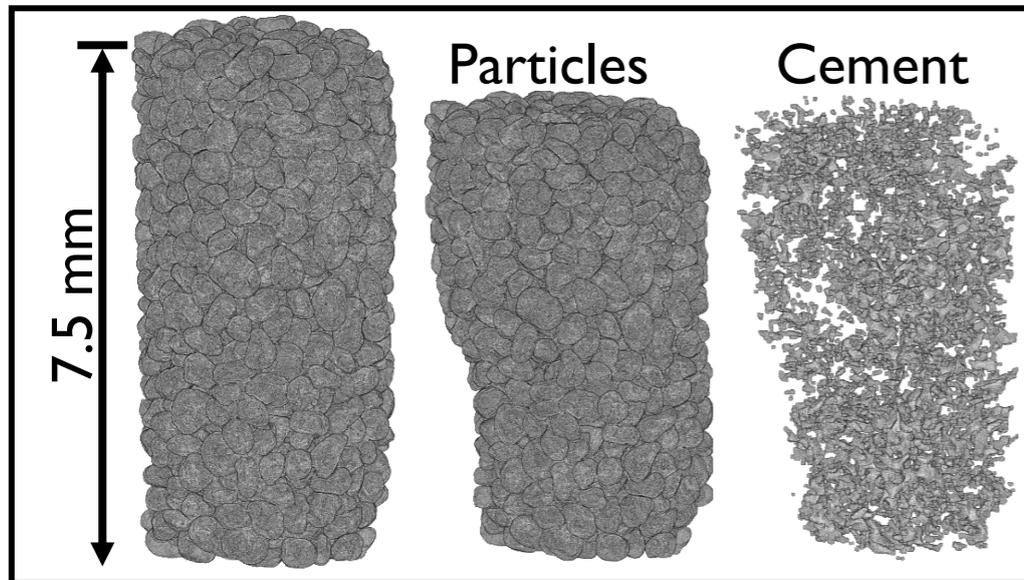
Capillary condensation: aging/adhesion of any rough surfaces in humid environments
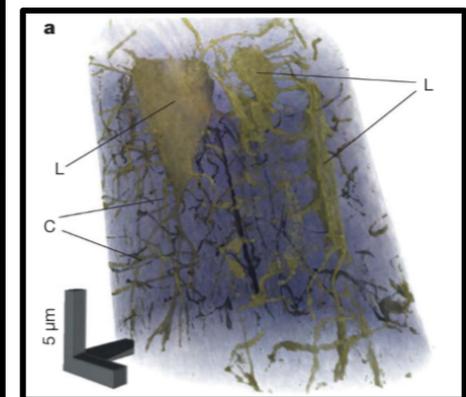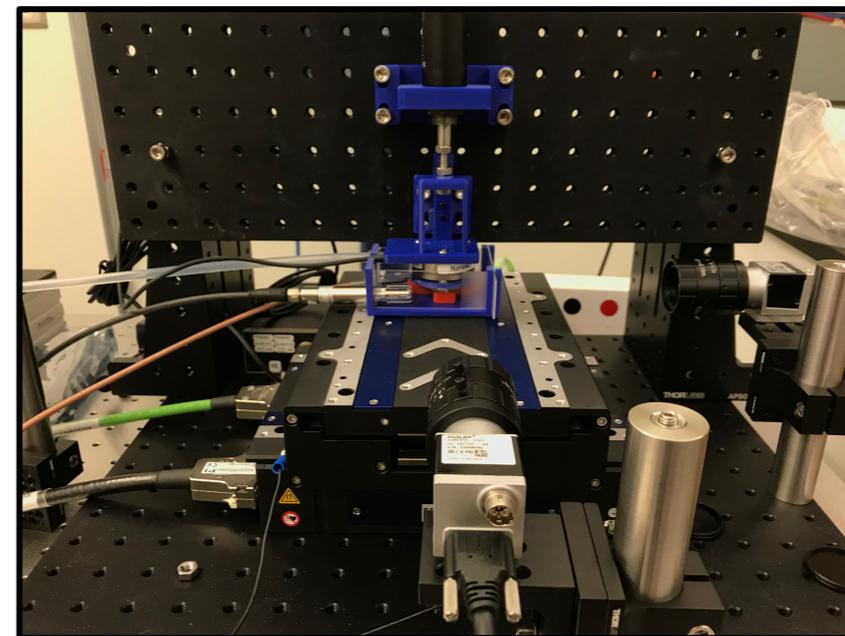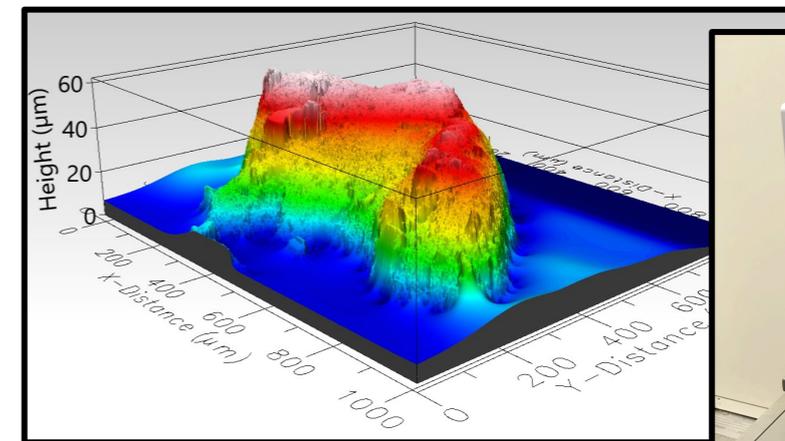[Israelichveli, 2011. Bocquet, *et al, Nature,* 1998.]

Motivation

(1) Cohesion mechanisms of interest? (2) What are our relevant microscale characterization techniques at bulk, particle levels?
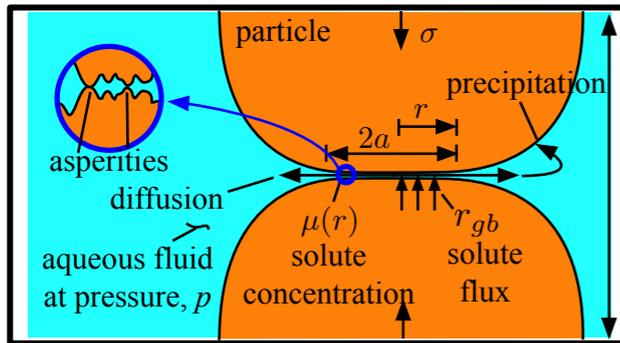

Micro, bulk: *in-situ* laboratory X-ray tomography, *in-situ* synchrotron 3D X-ray tomography, diffraction

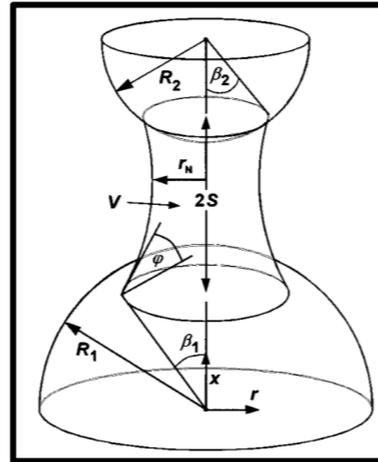[Zhai, *et al., JOM*, 2020. Zhai & Hurley, *unpublished*.]

Micro, particle: surface profilometry, *in-situ* contact mechanics testing, *in-situ* 3D X-ray nanotomography
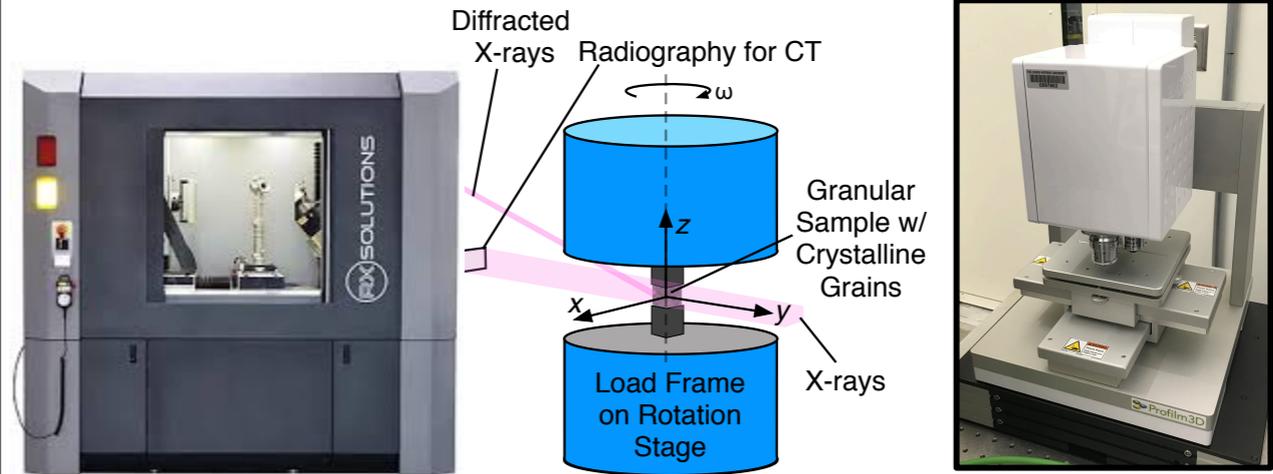
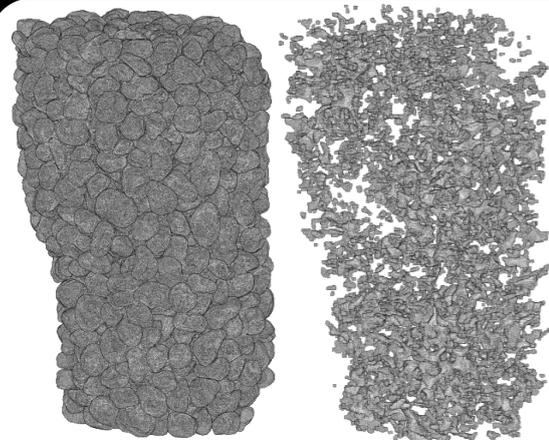[Hurley lab at JHU.; Dierolf, *et al. Nature*, 2010.]
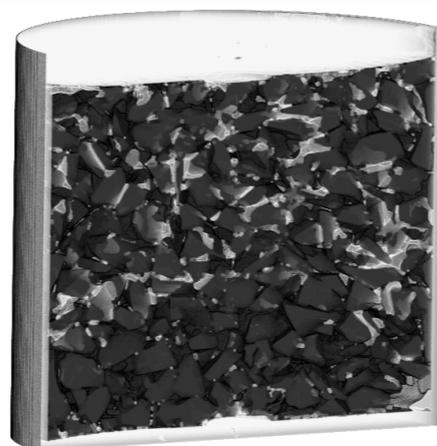
Outline

Inter-particle cementation

Capillary condensation

1. Mechanisms, Challenges

Diffracted X-rays — Radiography for CT

Granular Sample w/ Crystalline Grains
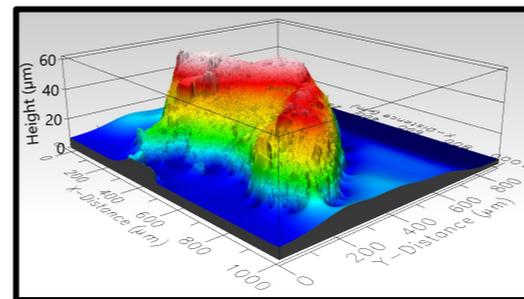
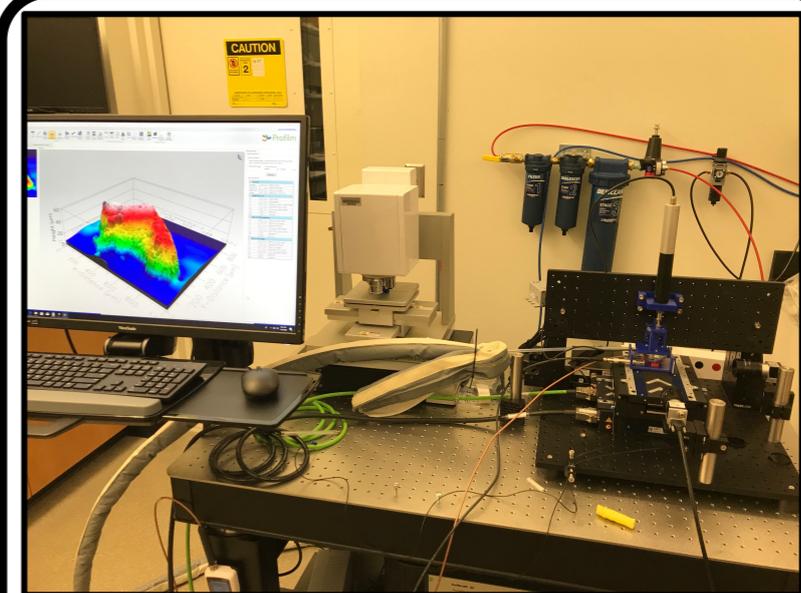Load Frame on Rotation Stage

X-rays

2. Characterization Tools

(1) XRCT: cemented cohesive mechanics

(2) XRCT + 3DXRD: particle-level stresses

(3) Roughness, %RH effects on particle-scale interactions

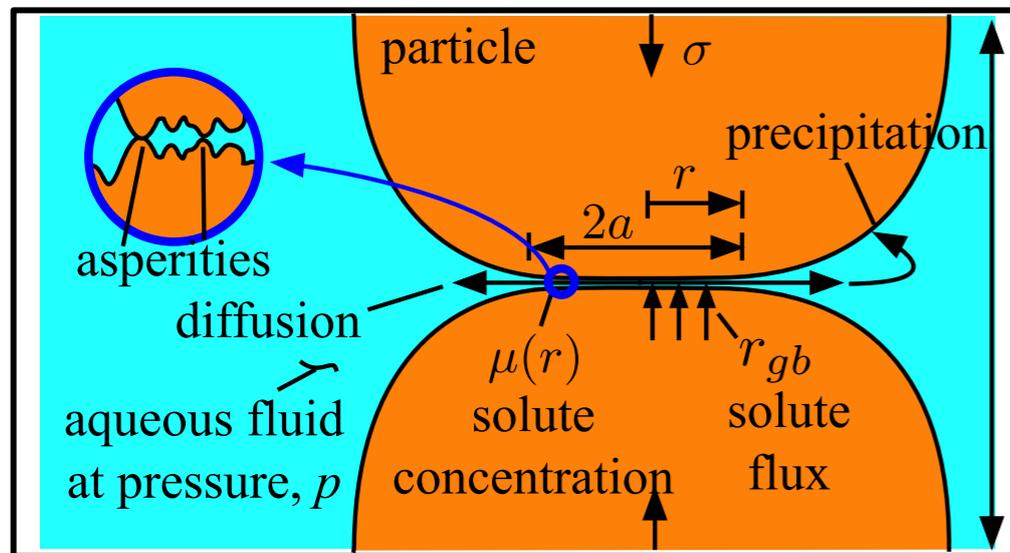## 3. Past & Future Applications

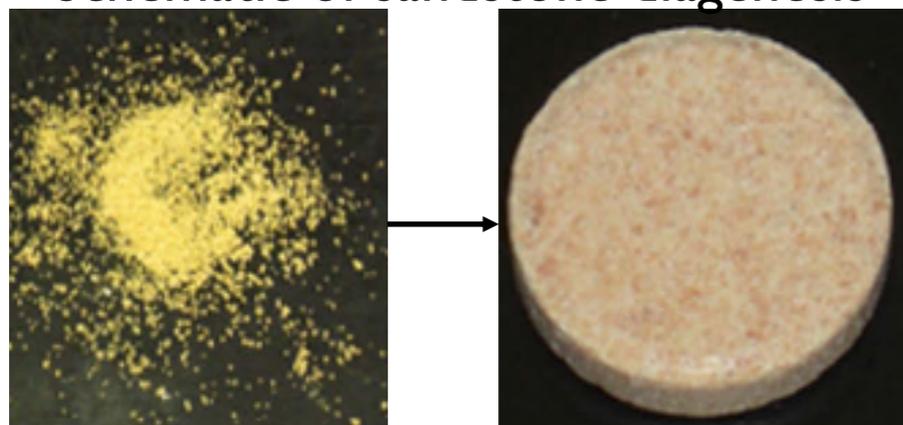## 4. Summary, Goals

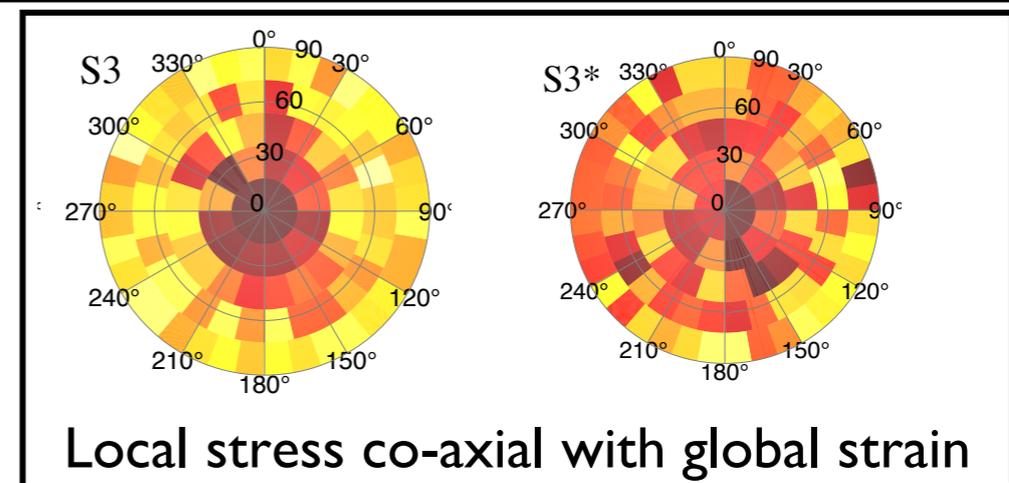1. <u>Cementation</u>: chemical or physical sintering of inter-particle contacts, found in geology and industry

Schematic of sandstone diagenesis

Tablet formation

(a) Cemented ϵ_{zz} (b) Non-cemented ϵ_{zz}

Local stress co-axial with global strain

<u>Causes</u>: Natural & synthetic reactions, polymer binders for agglomeration, tableting
[Hurley, *unpublished*.; Lehner & Leroy, *Int. Geophys. Ser.*, 2004. Alanazi, *S. Pharm. J.* 2010.]

<u>Effects</u>: Porosity (bulk), kinematics (particle), strength & coaxiality increase (both), fracture changes (both)
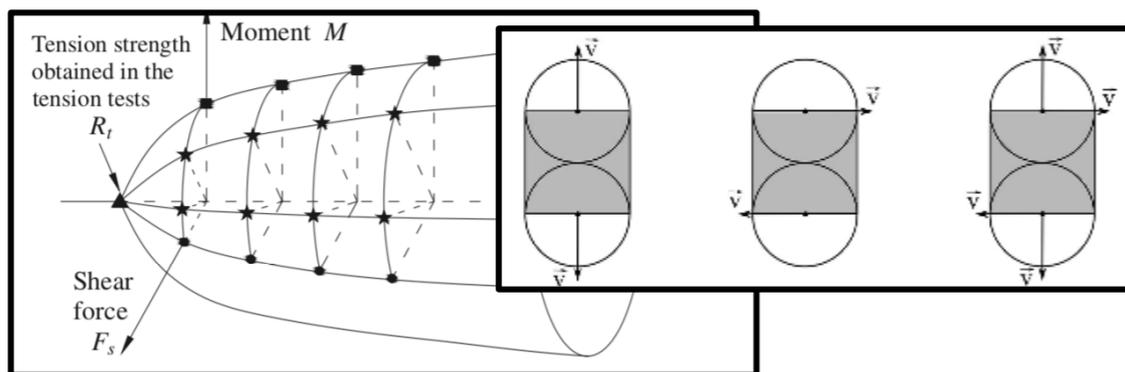[Zhai, *et al.*, *JOM 2020 & unpublished*.;.]

<u>1. Cementation, challenges</u>: (1) Link particle/binder props. and bulk elastic (*E, v*) and fracture (*K, G*) properties? (2) Particle/contact-scale fracture/fragmentation modes? (3) Non-uniform strain/stress fields?
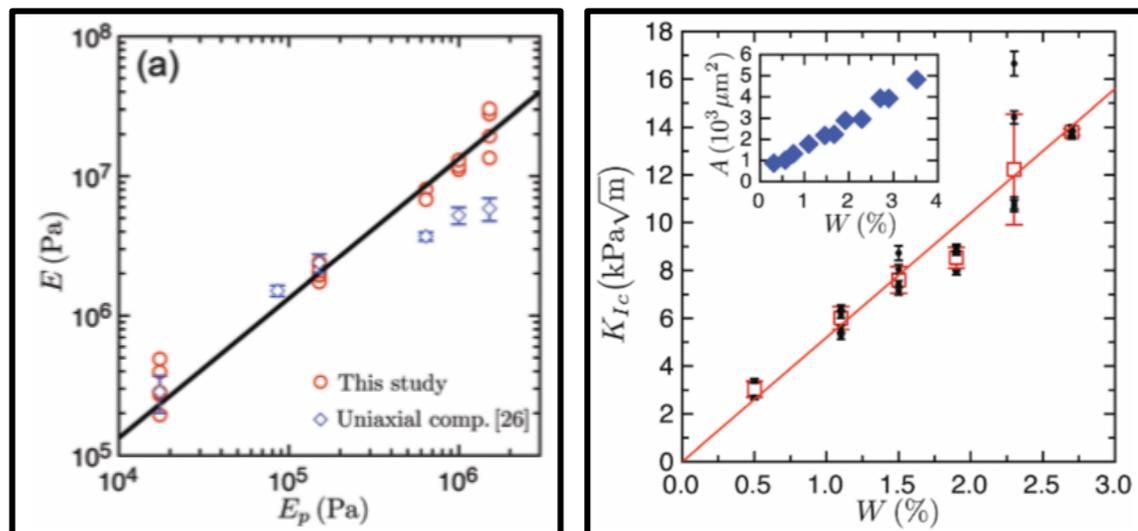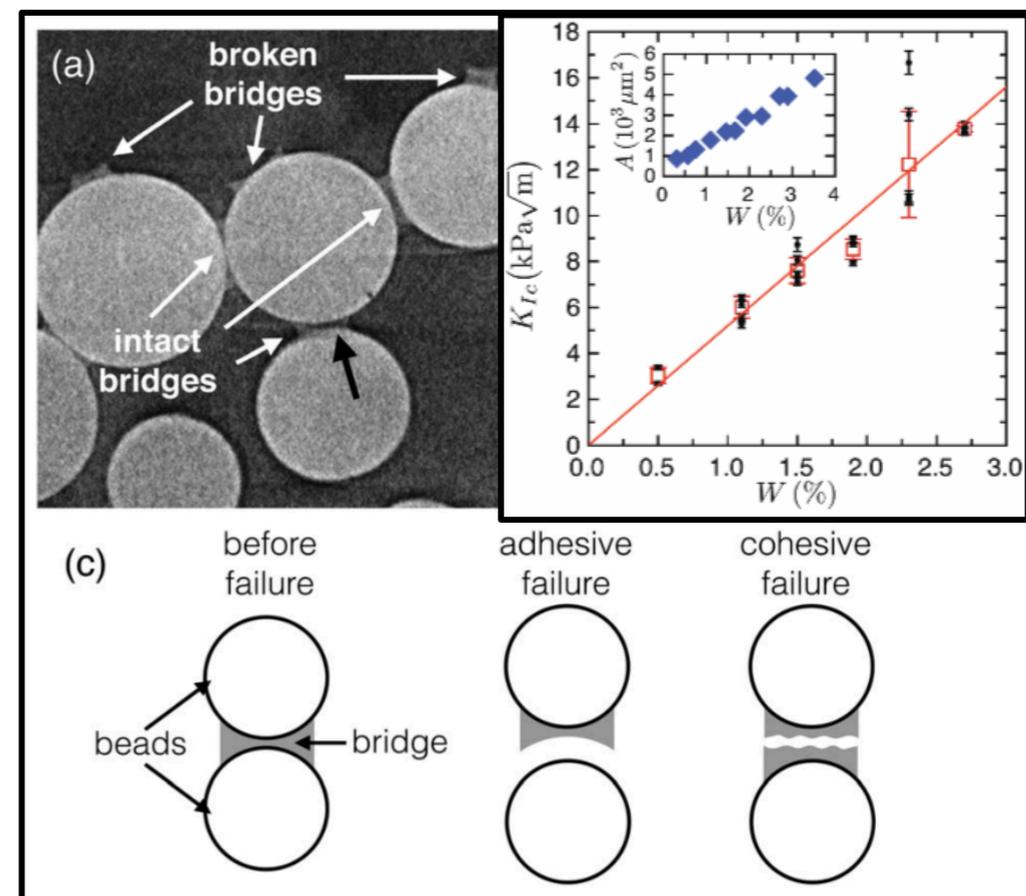

Elasticity scaling: $E \propto A, E \propto E_p$
[Hemmerle, *et al., Sci. Rep.* 2016.]

Theoretical/2D exps. for fracture modes [Nguyen, *et al. IJP,* 2017.; Jiang, *et al., MoM,* 2012.]

Fracture scaling: $K_{IC} \propto \phi_p, G \propto A$
$$K = \sqrt{(GE)} \rightarrow K \propto A$$

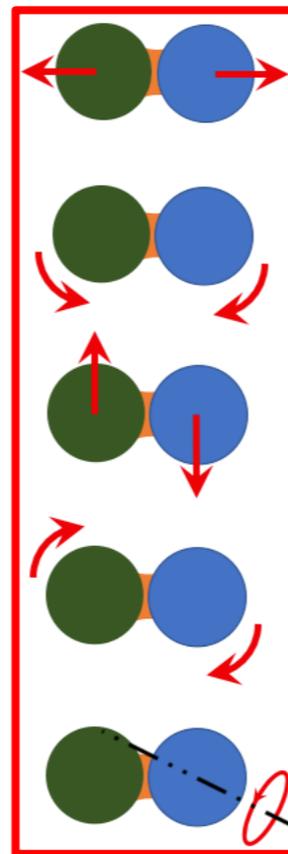Fracture modes: bridge detachment, some plasticity [Schmeink, *et al. Soft Matter,* 2017.]
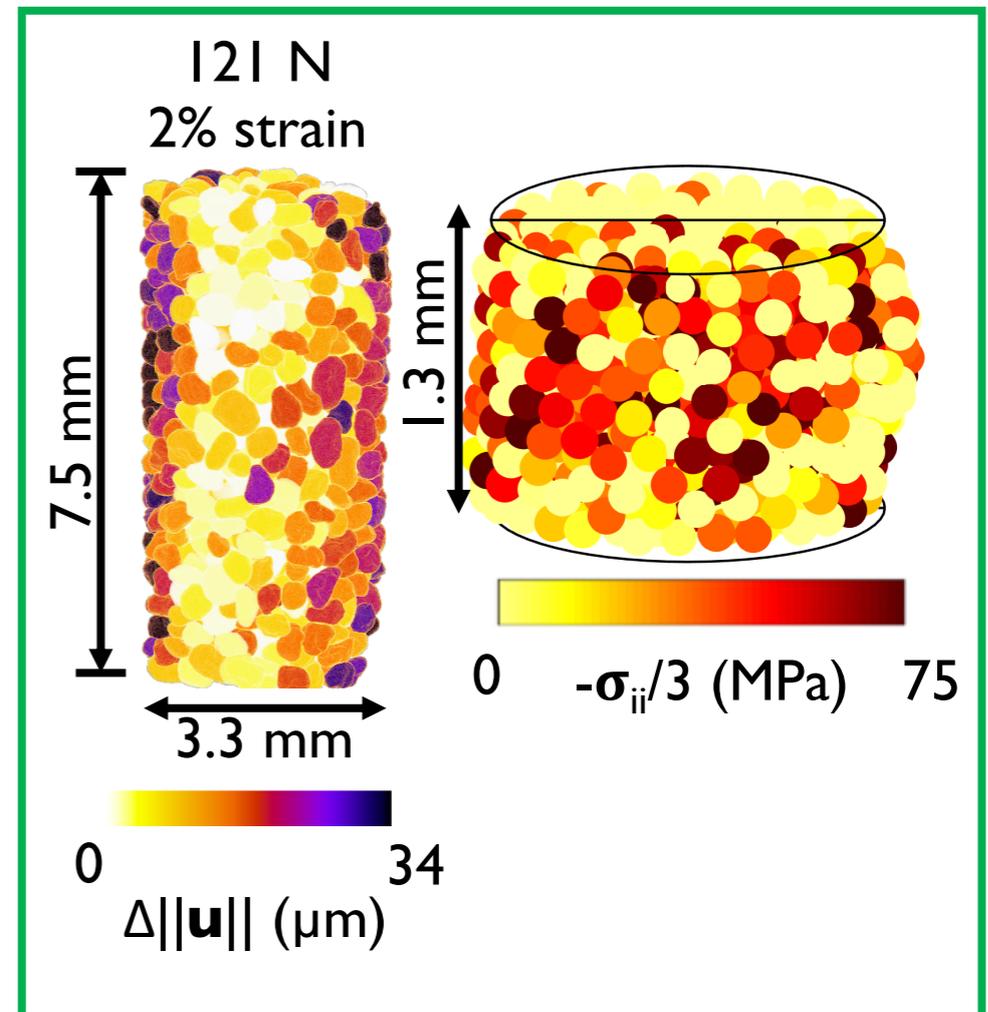
1. Cementation, challenges: (1) Link particle/binder props. and bulk elastic (*E, v*) and fracture (*K, G*) properties? (2) Particle/contact-scale fracture/fragmentation modes? (3) Non-uniform strain/stress fields?
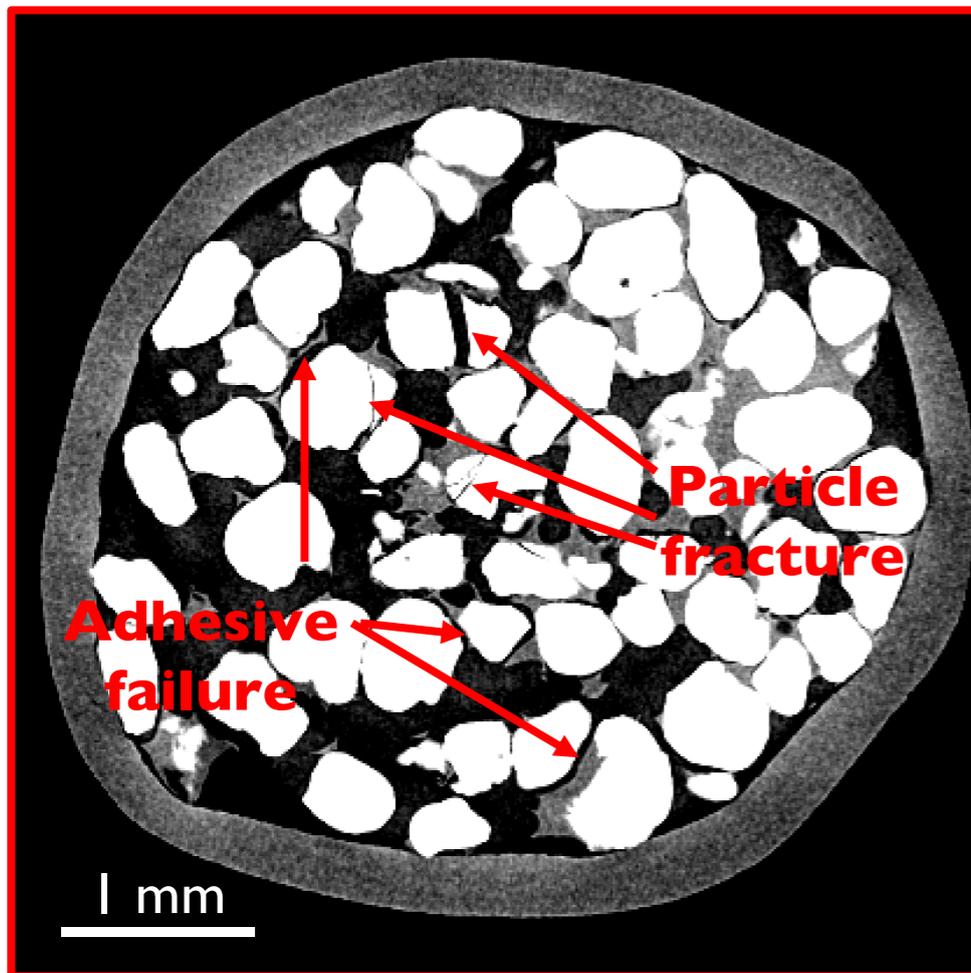
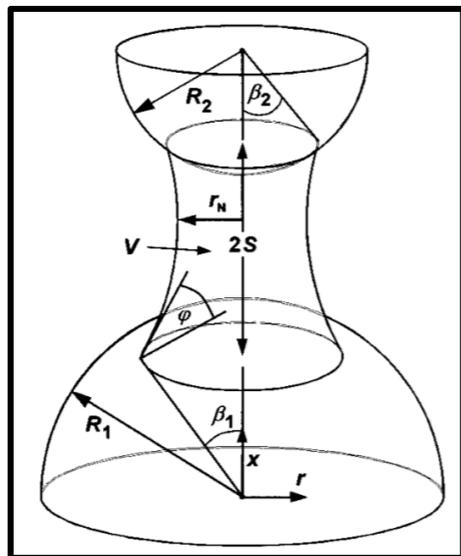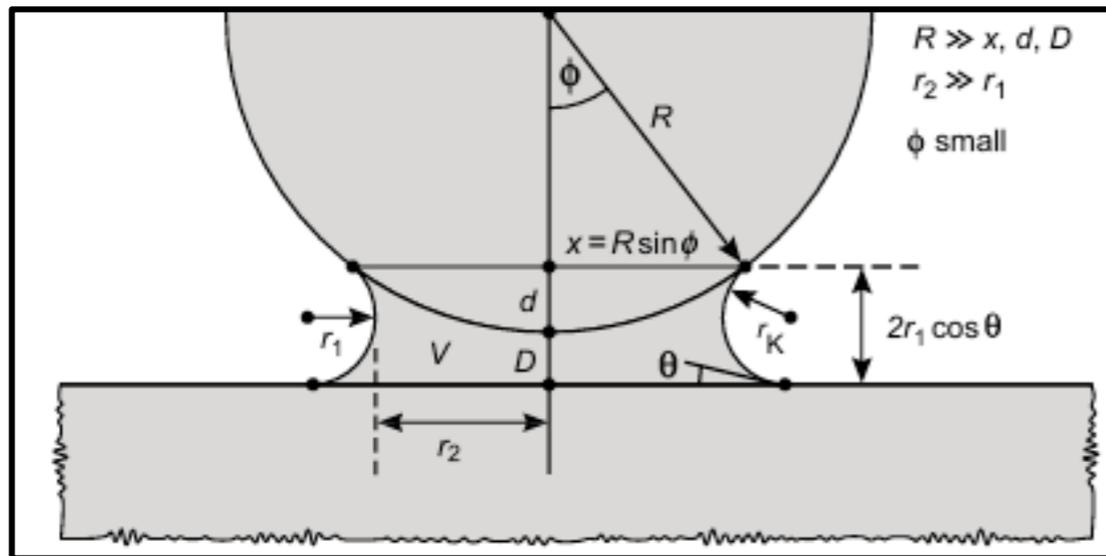Ottawa sand

Is adhesion-failure always strength-limiting?

121 N
2% strain

7.5 mm
3.3 mm
1.3 mm

0 — $-\sigma_{ii}/3$ (MPa) — 75

0 — $\Delta\|\mathbf{u}\|$ (μm) — 34

Most recent work focuses on the case when particle-binder adhesion is limiting. Other fracture modes?

Most experiments assume uniform strain fields. Other mechanical loading, non-uniform strain and stress fields, *in-situ* monitoring?
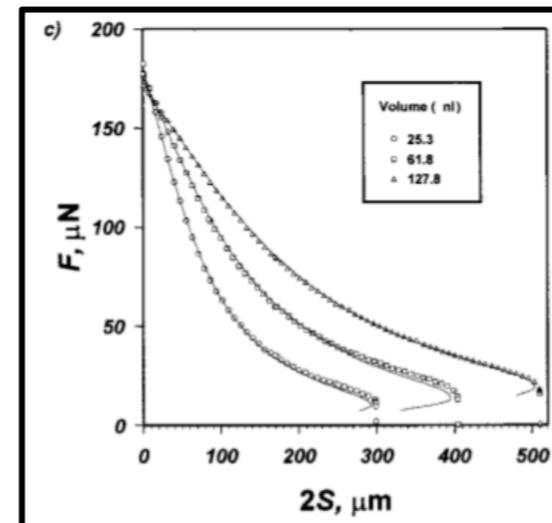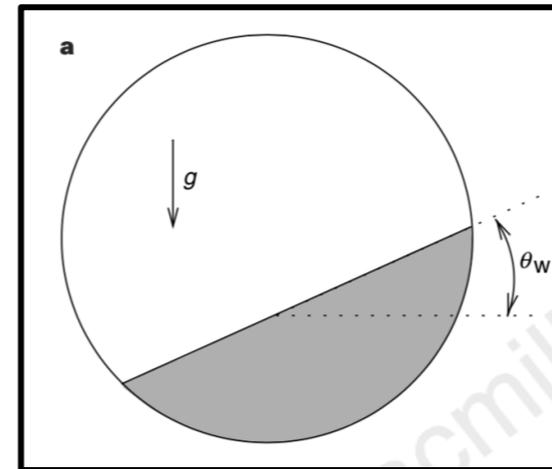
2. <u>Condensation/humidity mechanism</u>: capillary bridges in pores and contact throats due to reduction in vapor pressure

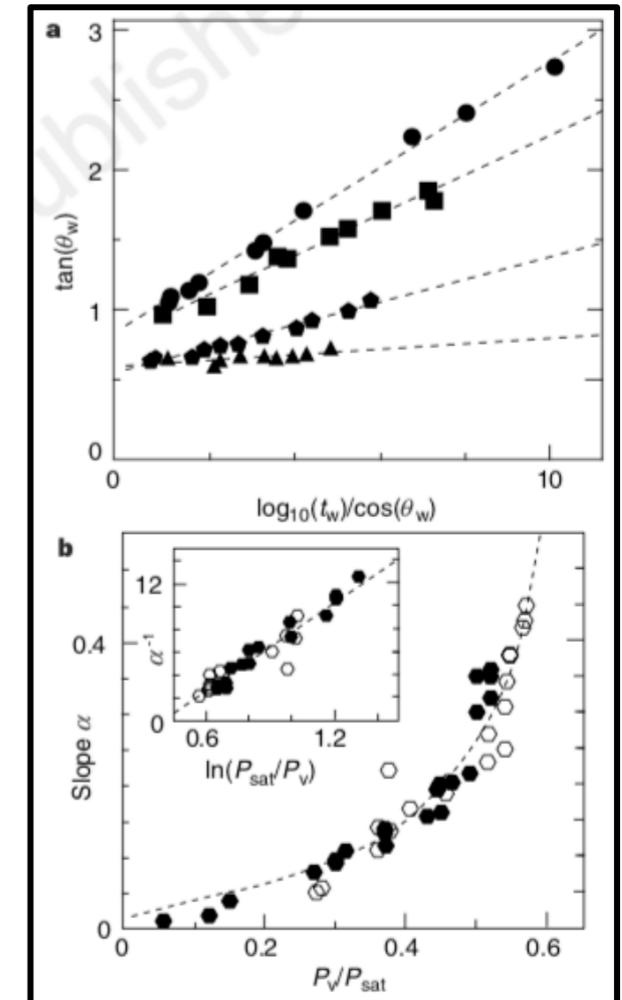$$F_{ad} = 2\pi r \gamma + \pi r^2 \Delta P$$

Adhesion as a function of bridge volume

Logarithmic friction aging, friction vs. humidity

<u>Causes</u>: Condensation in narrow spaces, favorable due to reduction in vapor pressure

[Israelichveli, 2011. Willet, *et al. Langmuir*, 2000.]

<u>Effects</u>: Adhesion (particle-scale), enhanced friction & viscosity (bulk scale), logarithmic aging (both scales)

[Israelichveli, 2011. Willet, *et al. Langmuir*, 2000. Thornton, *et al., J. Phys. D*, 1996. Bocquet, *et al., Nature*, 1998.]

2. Condensation/humidity challenges: (1) Adhesion and friction forces as a function of humidity & roughness at both contact- and bulk scales?
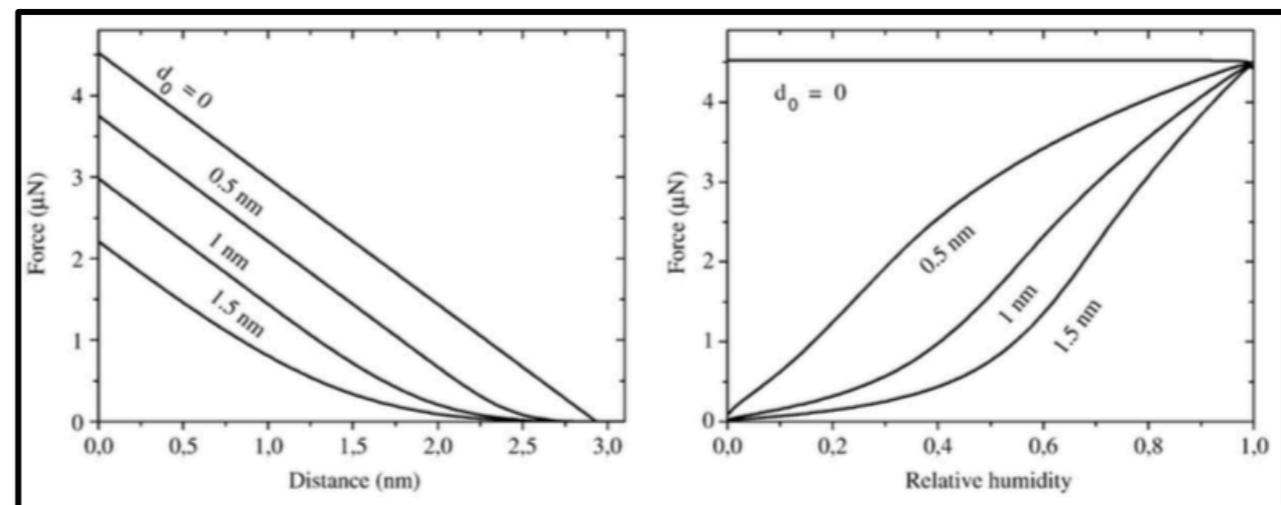
$$F_{ad} = \frac{AR}{6H_0^2}\left[\frac{1}{1+\left(\frac{32Rk_1\text{rms}}{\lambda^2}\right)} + \frac{1}{\left(1+\frac{k_1\text{rms}}{H_0}\right)^2}\right]$$
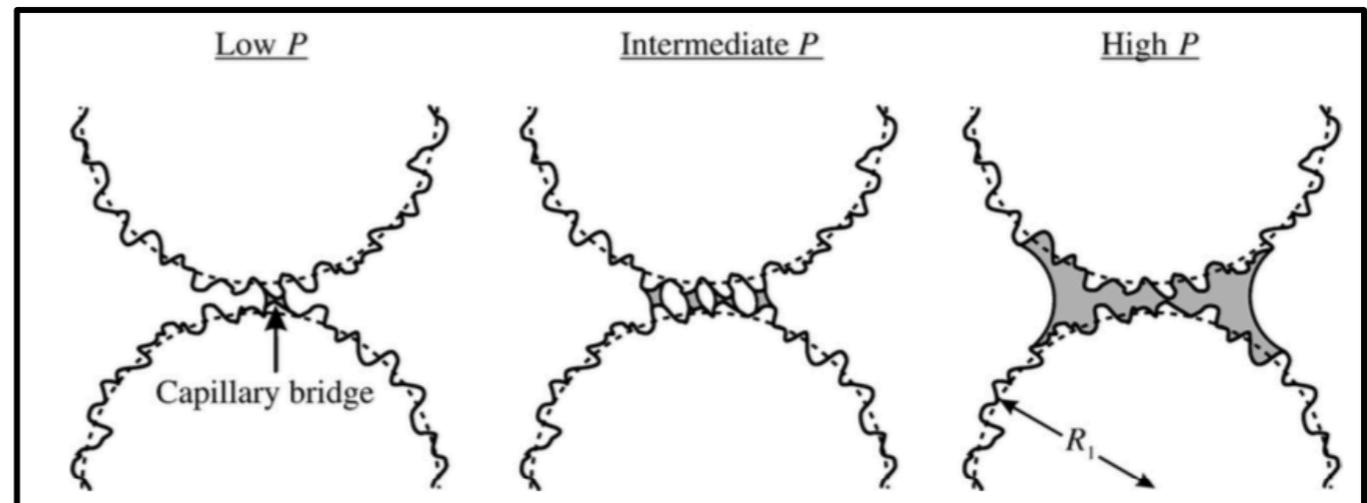
$$f_A \sim \frac{\Gamma V_1}{l_R^2}\left(\frac{V}{V_1}\right)^{(2-\chi)/(2+\chi)}, \qquad \text{for } V < V_1$$

$$f_A \sim \frac{\Gamma V}{l_R^2}, \qquad \text{for } V_1 < V < V_2$$
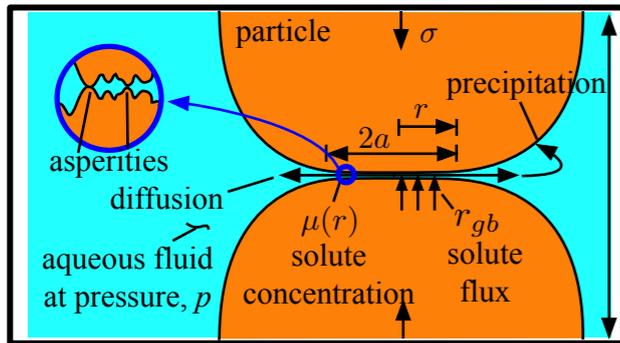
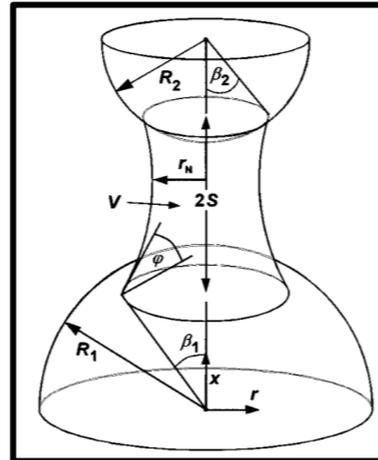$$f_A = 2\pi\Gamma R, \qquad \text{for } V > V_2$$

Roughness can reduce adhesion forces & separation distances/strains for rupture:
[Rabinovitch, *et al., J. Coll. & Int. Sci.* 2000.]

Roughness leads to three regimes of capillary adhesion, changes force vs humidity profile
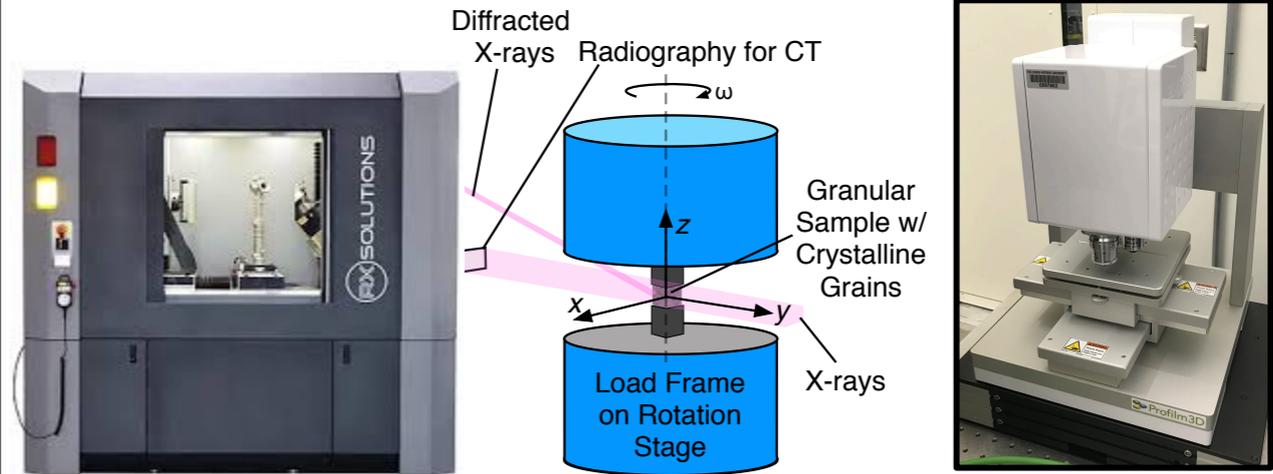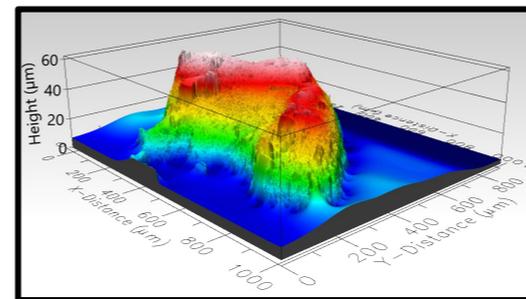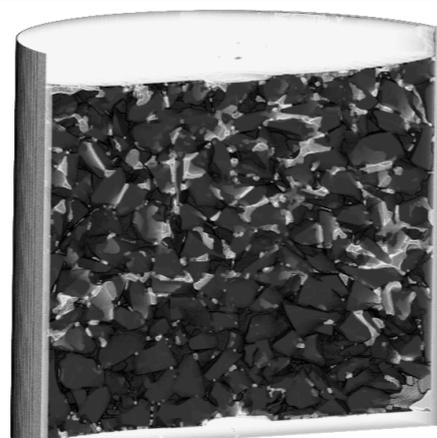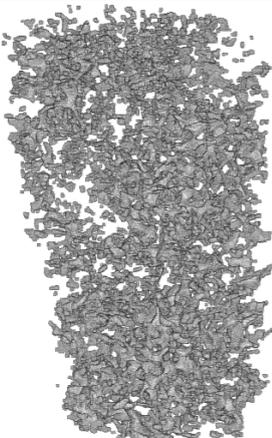[Butt and Kappl, *Adv. Coll. & Int. Sci.,* 2009.]

Outline

Inter-particle cementation
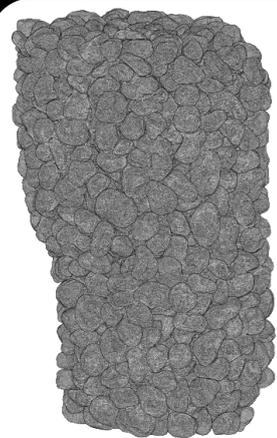
Capillary condensation

1. Mechanisms, Challenges

Diffracted X-rays
Radiography for CT
Granular Sample w/ Crystalline Grains
Load Frame on Rotation Stage
X-rays

2. Characterization Tools

(1) XRCT: cemented cohesive mechanics

(2) XRCT + 3DXRD: particle-level stresses

(3) Roughness, %RH effects on particle-scale interactions

3. Past & Future Applications
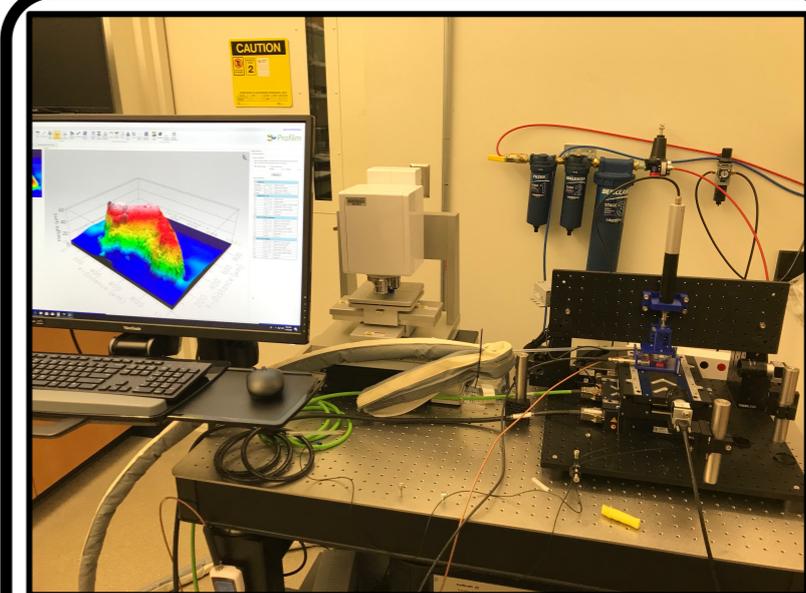
4. Summary, Goals
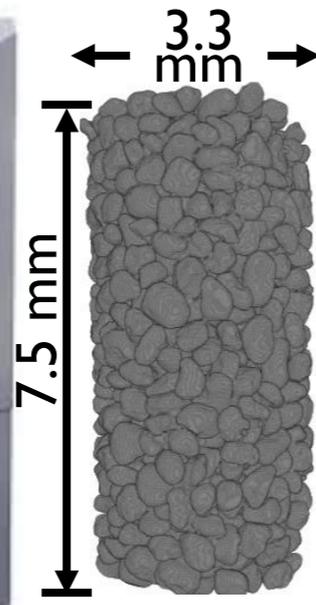
1. *In-situ* laboratory X-ray computed tomography (XRCT).
2. *In-situ* synchrotron XRCT and 3D X-ray diffraction (3DXRD).
3. Surface profilometry & friction/contact/adhesion testing.

3.3 mm

7.5 mm

10.9% strain

Particles

Epoxy

RX Solutions Easytom 160

Sample

X-ray radiographs

3D volume correlation (DVC) – Example later

Can do this with water

1 mm

[Hurley, *unpublished*. Scheel, *et al., Nature,* 2008.]
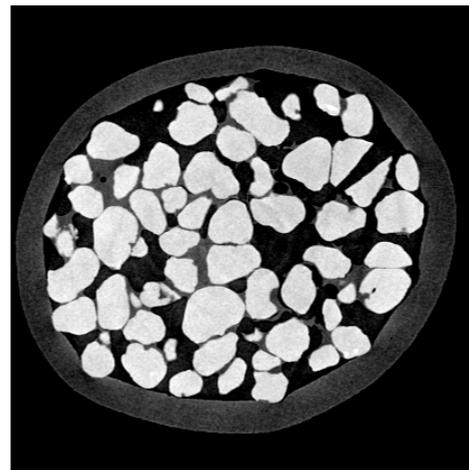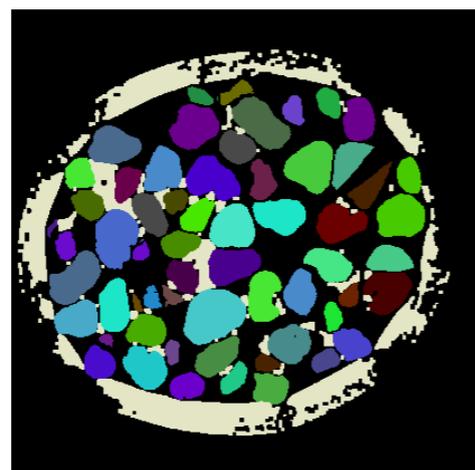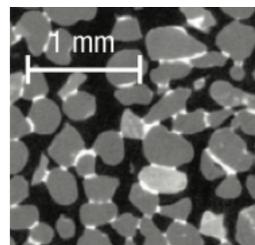
Multi-phase segmentation

In-situ XRCT images, resolutions to 0.3 μm

1. *In-situ* laboratory X-ray computed tomography (XRCT).
2. *In-situ* synchrotron XRCT and 3D X-ray diffraction (3DXRD).
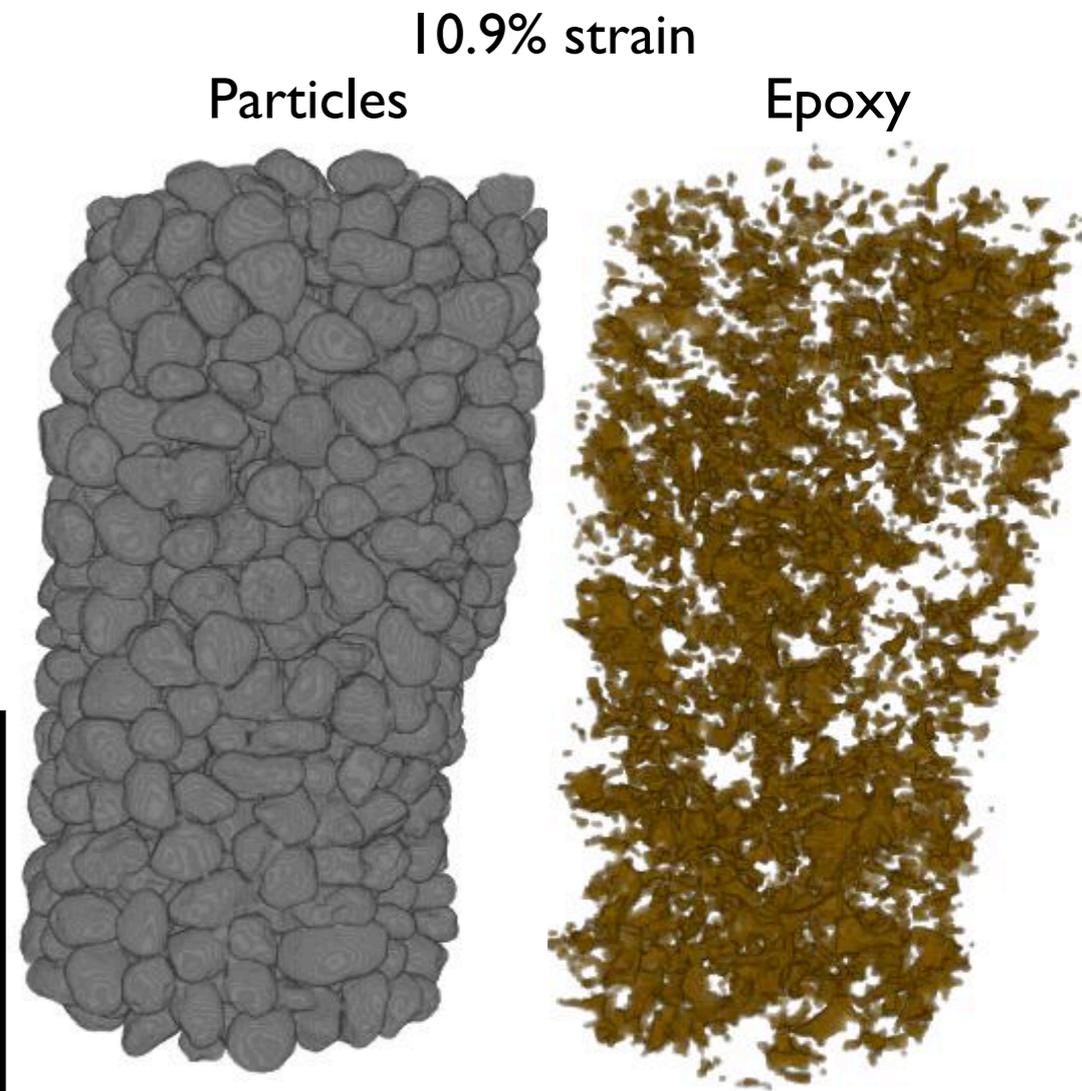3. Surface profilometry & friction/contact/adhesion testing.

Advanced Photon Source (APS)

Experiment Schematic

Single crystal particles

Bragg peaks with 2θ, ω, azimuth

Quantitative inter-particle force vectors at all contacts
[Hurley, *et al., PRL,* 2016.]

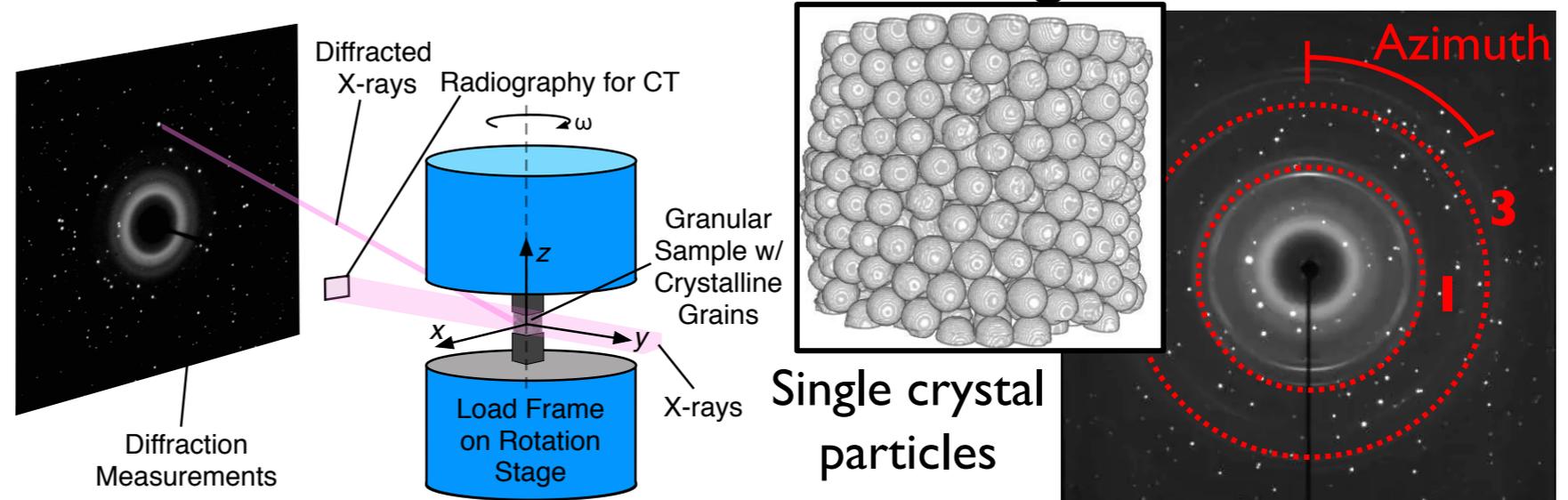Peaks processed for strain *tensors*, orientations, positions

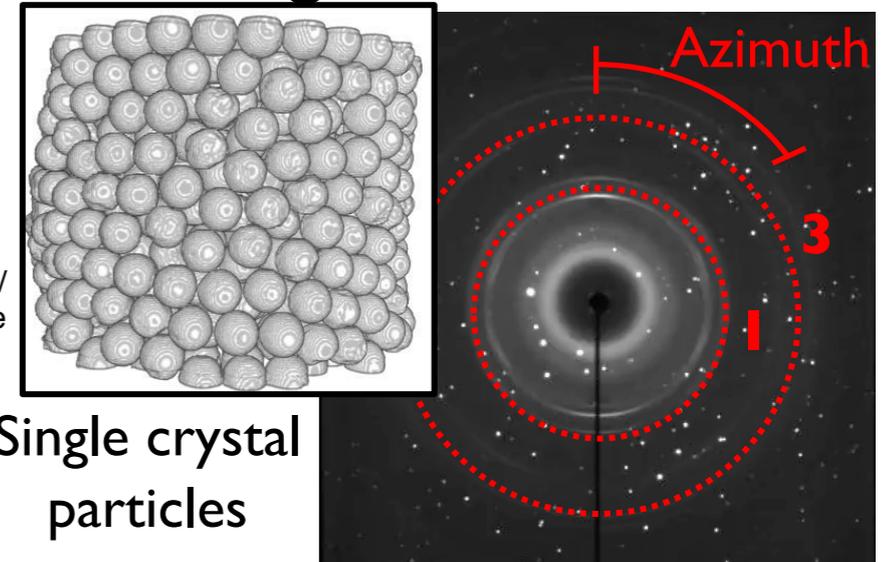0°   Δθ   6°   0   -σ$_{ii}$/3 (MPa)   75

1. *In-situ* laboratory X-ray computed tomography (XRCT).
2. *In-situ* synchrotron XRCT and 3D X-ray diffraction (3DXRD).
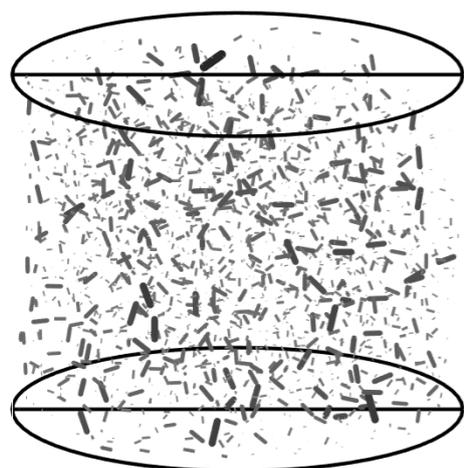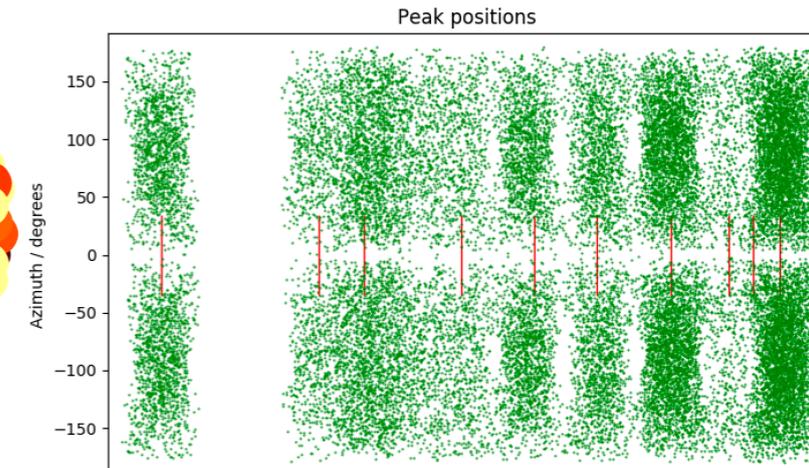3. <u>Surface profilometry & friction/contact/adhesion testing.</u>
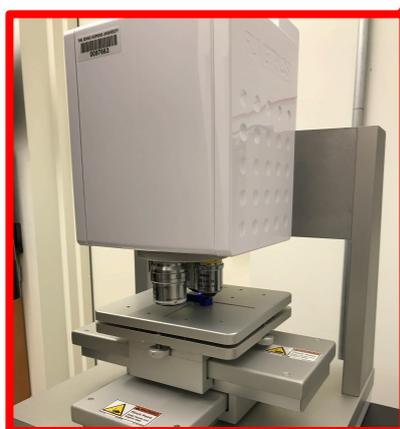
Profilm3D Profilometer,
~ 1 nm surface feature height

2 mm

XYZ contact/shear tests. 1 nm
XY, 50 nm Z, 6-axis load cell,
forces down to 4 μN
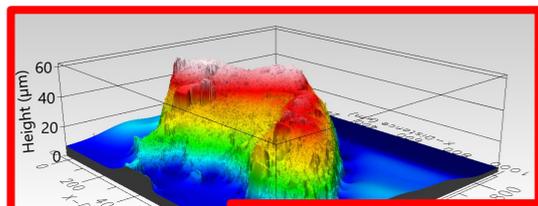
Ex: Ottawa sand
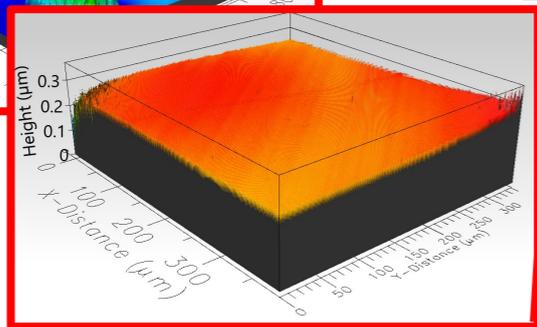
Ex: Quartz wafer
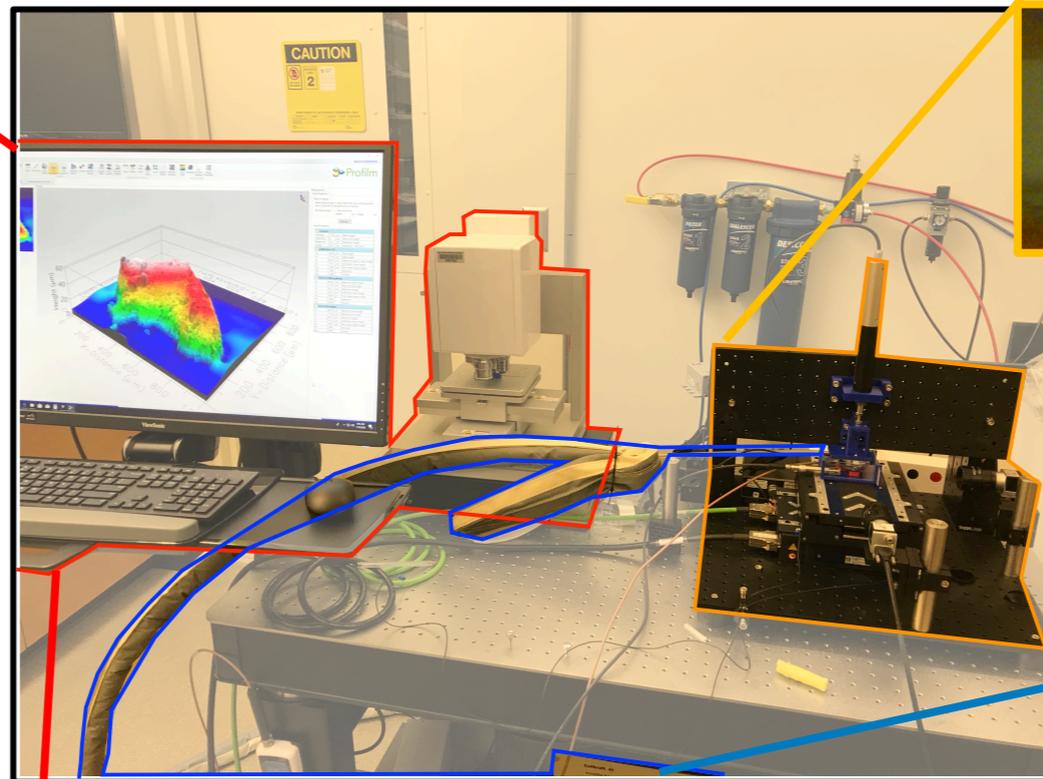
Surface and Contact Micromechanics
Device at JHU

(Miniaturized version
for X-ray nano-
tomography, 3DXRD)

Humidifier: 0-100%, 0-50°C

Outline
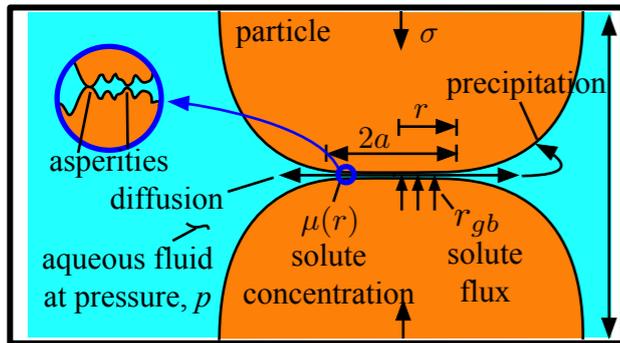
Inter-particle cementation
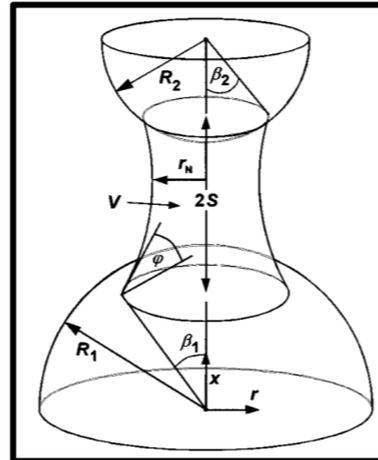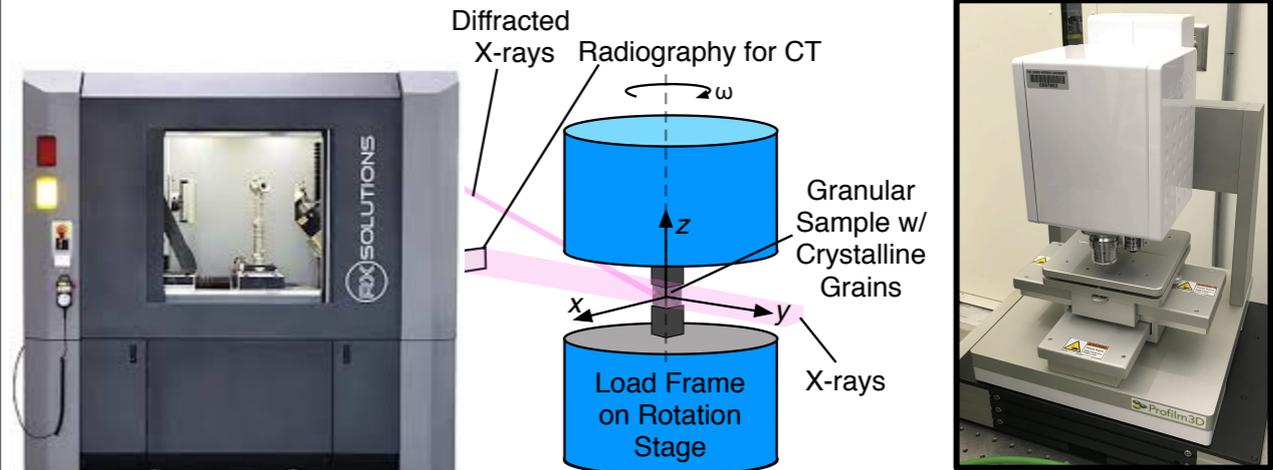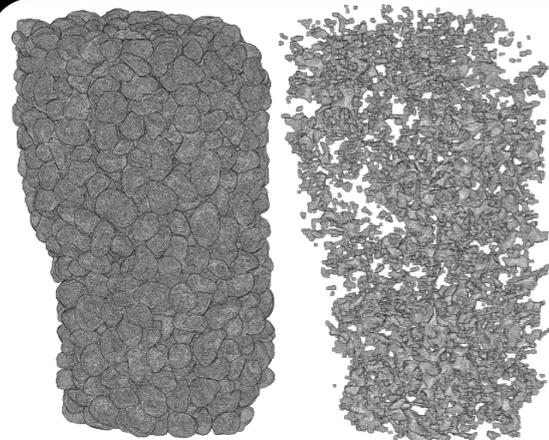
Capillary condensation

1. Mechanisms, Challenges

Diffracted X-rays — Radiography for CT
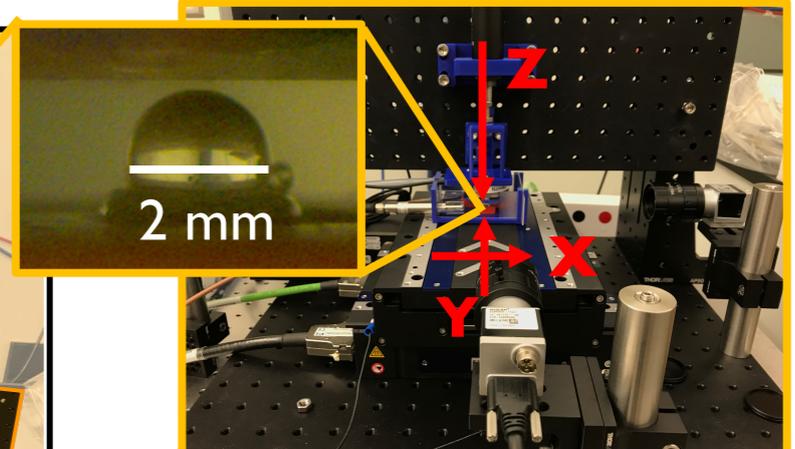
Granular Sample w/ Crystalline Grains

Load Frame on Rotation Stage — X-rays

2. Characterization Tools

(1) XRCT: cemented cohesive mechanics
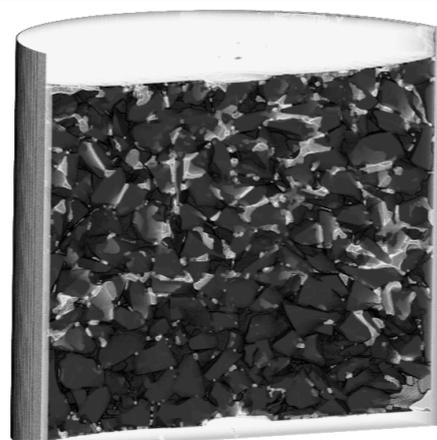
(2) XRCT + 3DXRD: particle-level stresses
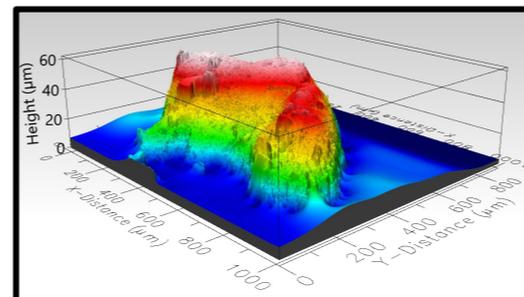
(3) Roughness, %RH effects on particle-scale interactions
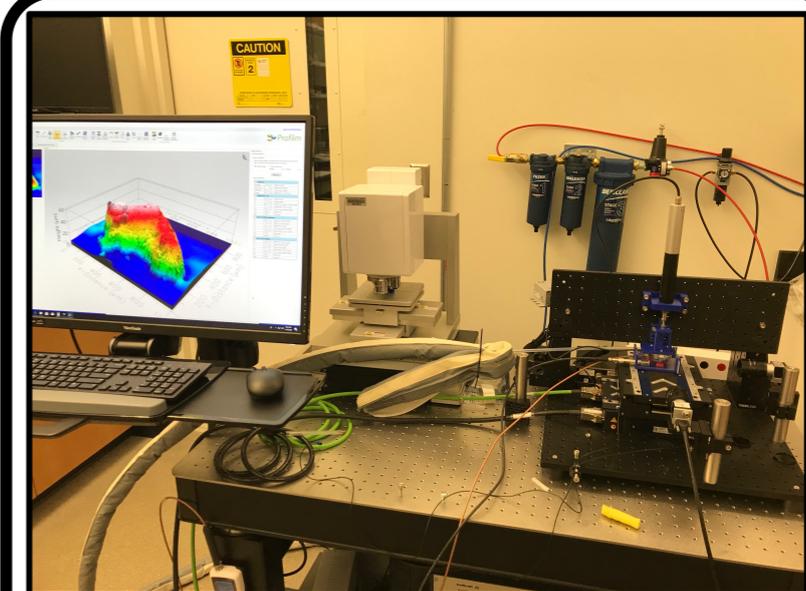
3. Past & Future Applications

4. Summary, Goals

Addressing cementation challenges: (1) Link particle/binder props. and bulk elastic (*E*, *v*) and fracture (*K*, *G*) properties? (2) Particle/contact-scale fracture/fragmentation modes? (3) Non-uniform strain/stress fields?
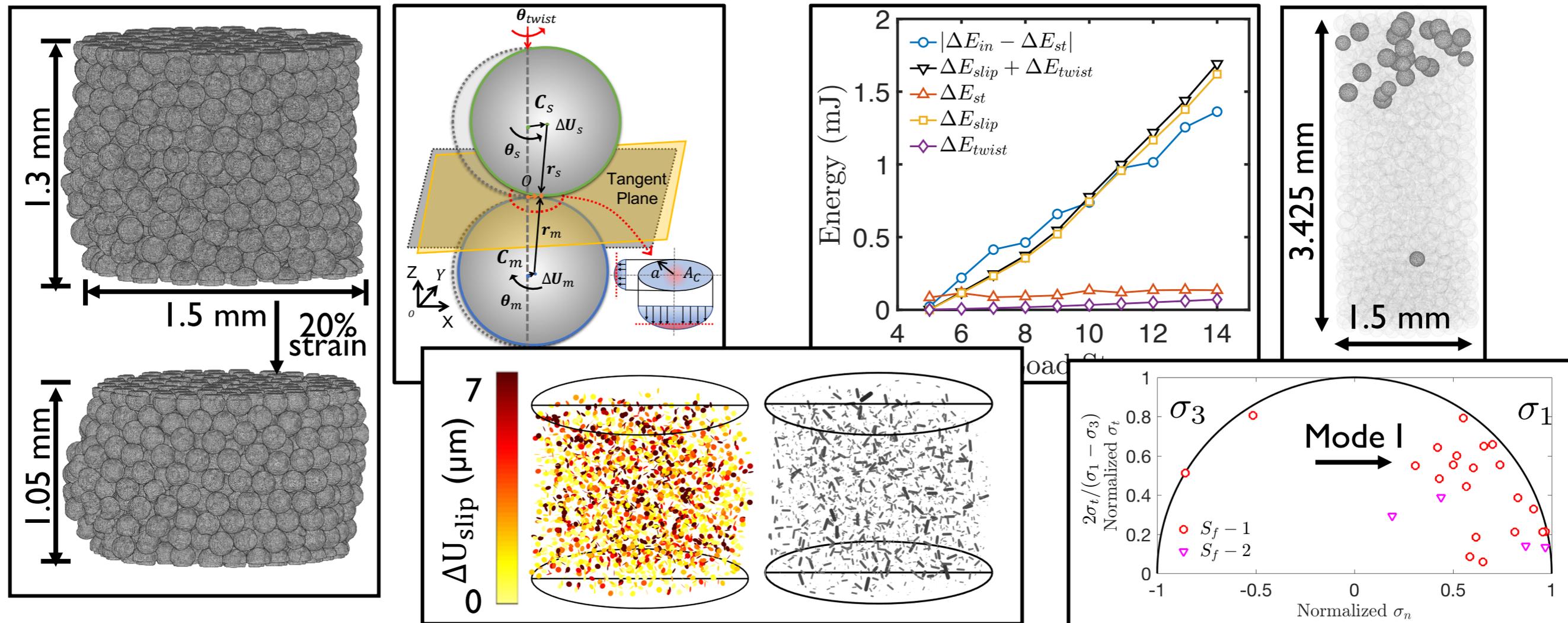
We have developed tools for XRCT data that evaluate local kinematics & dissipation (and local particle stress fields using 3DXRD). These are being used to address fracture modes in cemented materials.

[Zhai, *et al. JMPS*, 2019. Hurley, *et al., PRL*, 2016. Hurley, *et al., J. Mech. Phys. Solids*, 2018.]

Cohesive cementation: fracture (*K, G*) properties? (2) Particle/contact-scale fracture/fragmentation modes? (3) Non-uniform strain/stress fields?

Particles: 1,000 Ottawa (98% SiO_2) sand particles, passing 300 µm sieve

Sample: Sand mixed with 5% (by vol.) Extec epoxy, cured 24 hours

Loading: Uniaxial compression with 316 µm-thick membrane
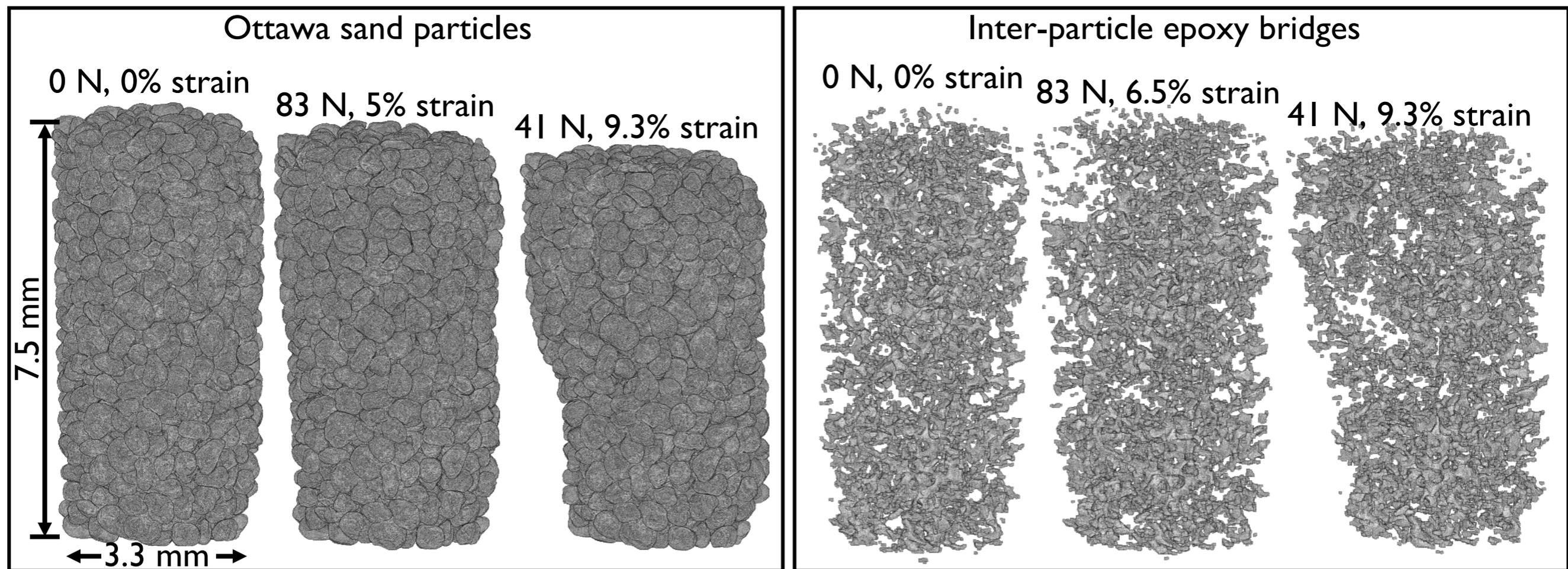
Ottawa sand particles

0 N, 0% strain
83 N, 5% strain
41 N, 9.3% strain

7.5 mm
3.3 mm

Inter-particle epoxy bridges

0 N, 0% strain
83 N, 6.5% strain
41 N, 9.3% strain

[Zhai & Hurley, *unpublished,* 2020.]

Cohesive cementation: mechanisms of fragmentation – *in-situ* 3D DVC for particle kinematics, image analysis for particle/cement fracture
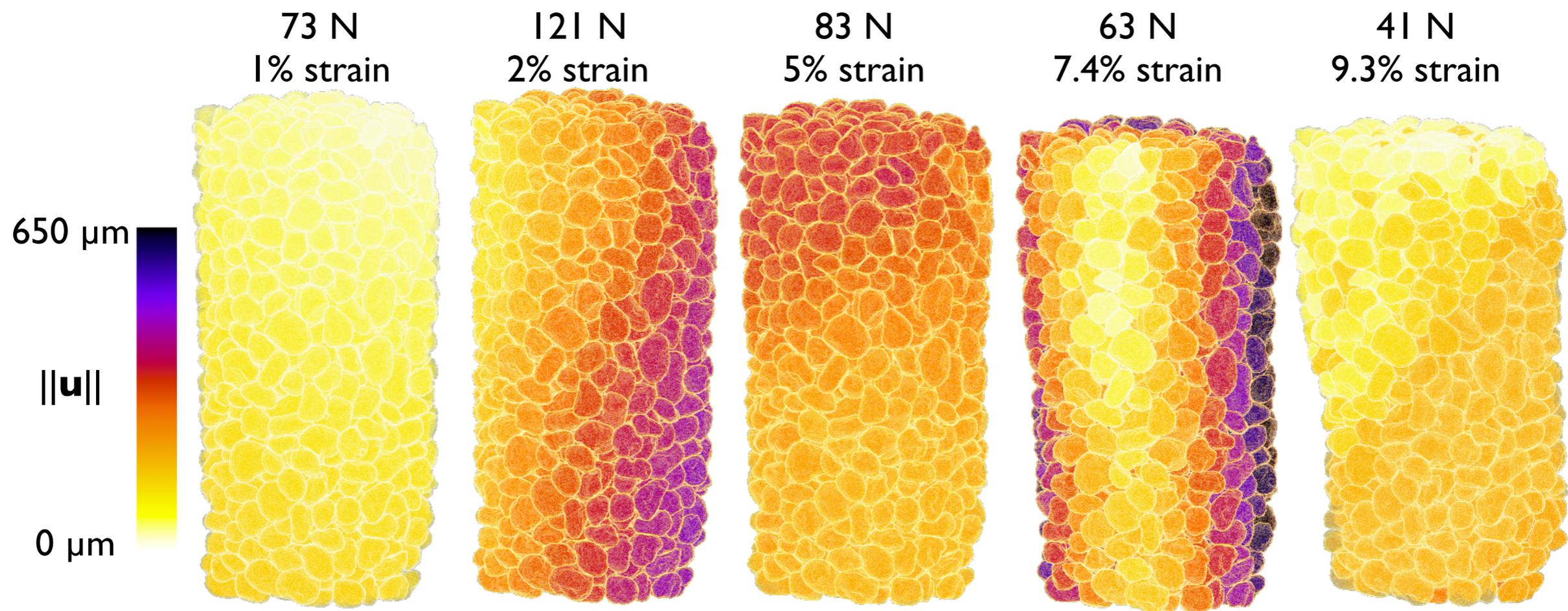
Total displacement of particles from 3D Discrete Digital Volume Correlation (3D-DVC). 3D-DVC code is SPAM, partially developed with Lab 3SR, Grenoble.

[Zhai & Hurley, *unpublished,* 2020. DVC code: Ando, *et al., unpublished*]

Cohesive cementation: mechanisms of fragmentation – *in-situ* 3D DVC for particle kinematics, image analysis for particle/cement fracture

73 N
1% strain

121 N
2% strain

83 N
5% strain

63 N
7.4% strain

41 N
9.3% strain

34 µm

$\Delta\|\mathbf{u}\|$

0 µm

Relative Displacements of particles from their 15 nearest neighbors from 3D Discrete Digital Volume Correlation (3D-DVC)
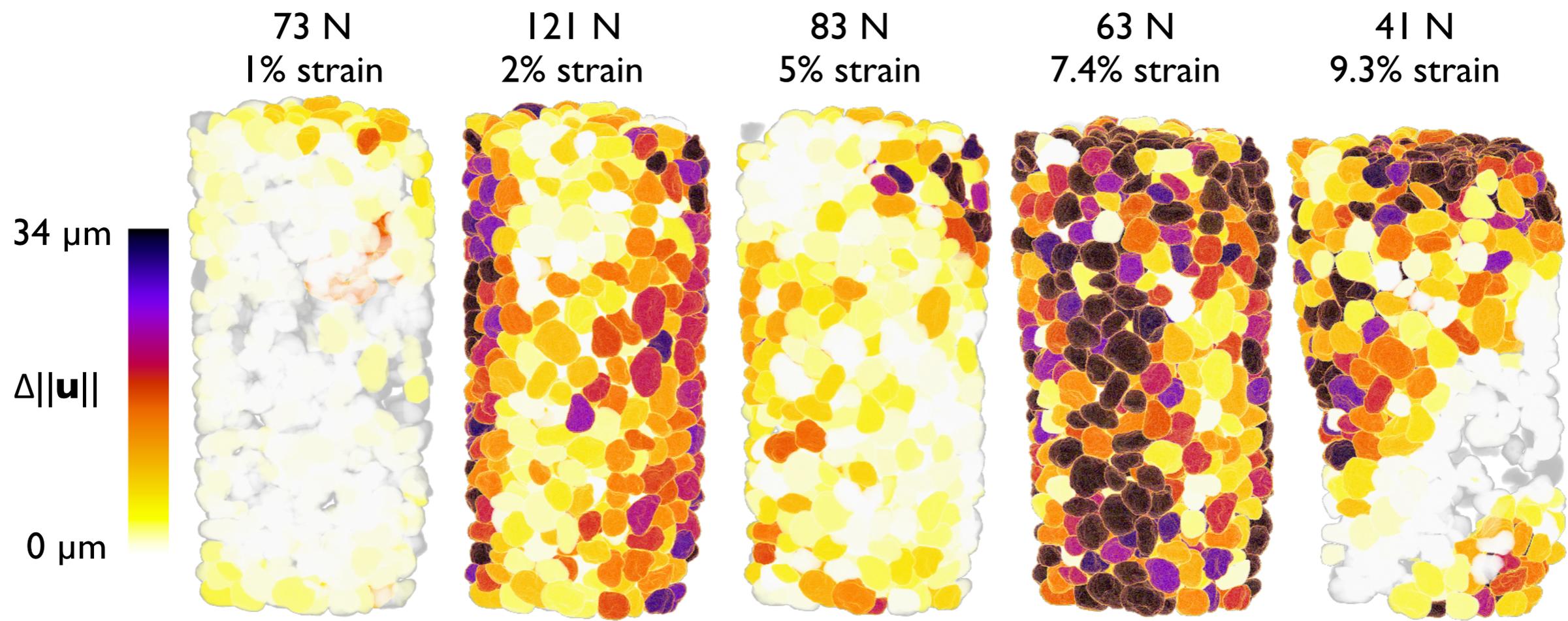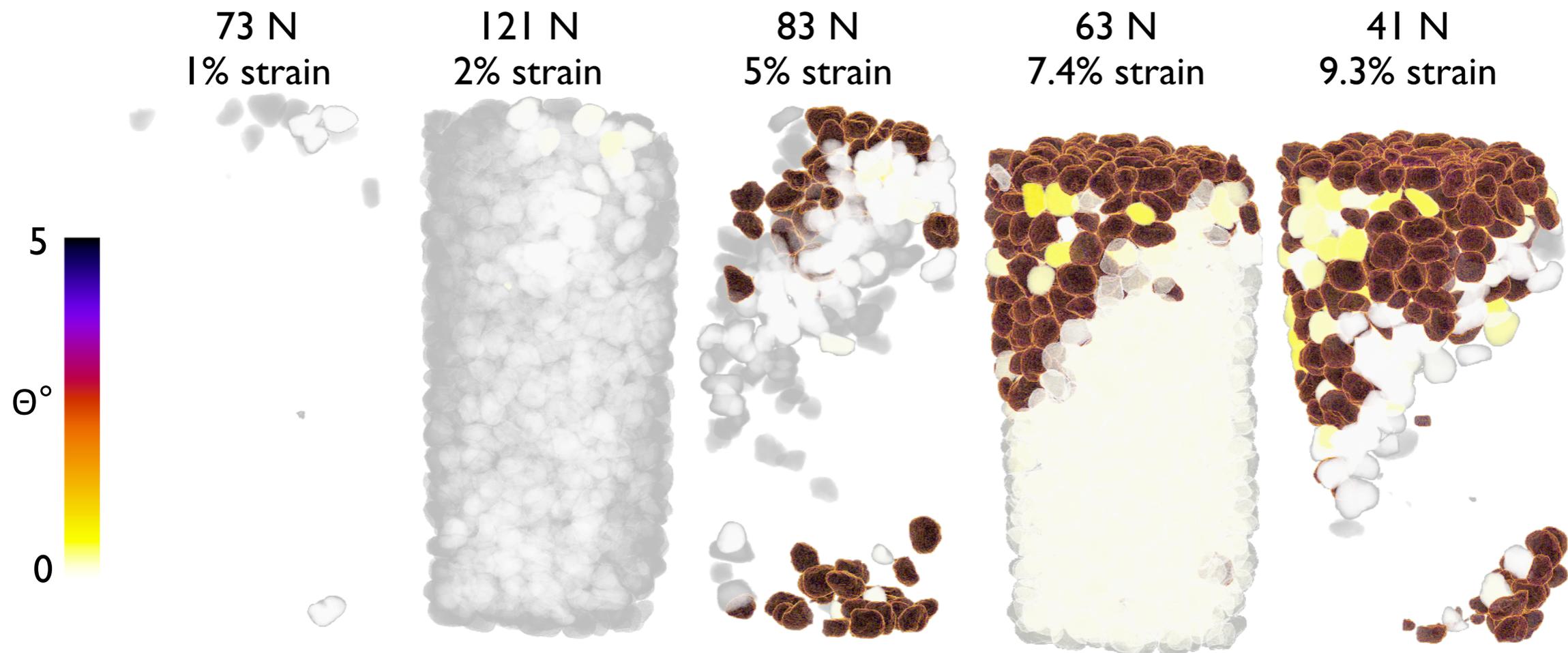
[Zhai & Hurley, *unpublished,* 2020. DVC code: Ando, *et al., unpublished*]

Cohesive cementation: mechanisms of fragmentation – *in-situ* 3D DVC for particle kinematics, image analysis for particle/cement fracture


Rotations of individual particles from 3D Discrete Digital Volume Correlation (3D-DVC) [Note: fractured particles will appear to rotate more]

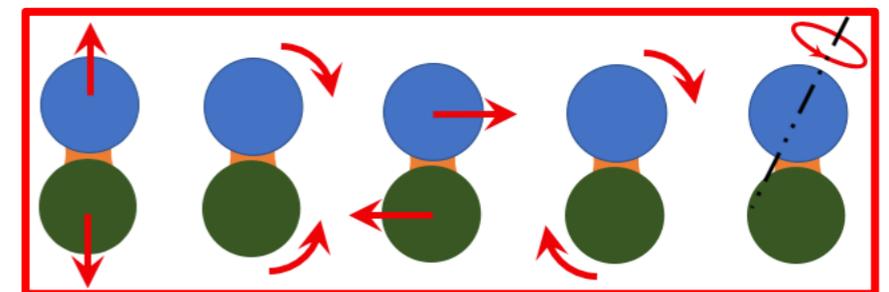[Zhai & Hurley, *unpublished,* 2020. DVC code: Ando, *et al., unpublished*]

Cohesive cementation: mechanisms of fragmentation – *in-situ* 3D DVC for particle kinematics, image analysis for particle/cement fracture

0 N
1% strain

121 N
5% strain

63 N
9.3% strain

650 μm

||**u**||

0 μm
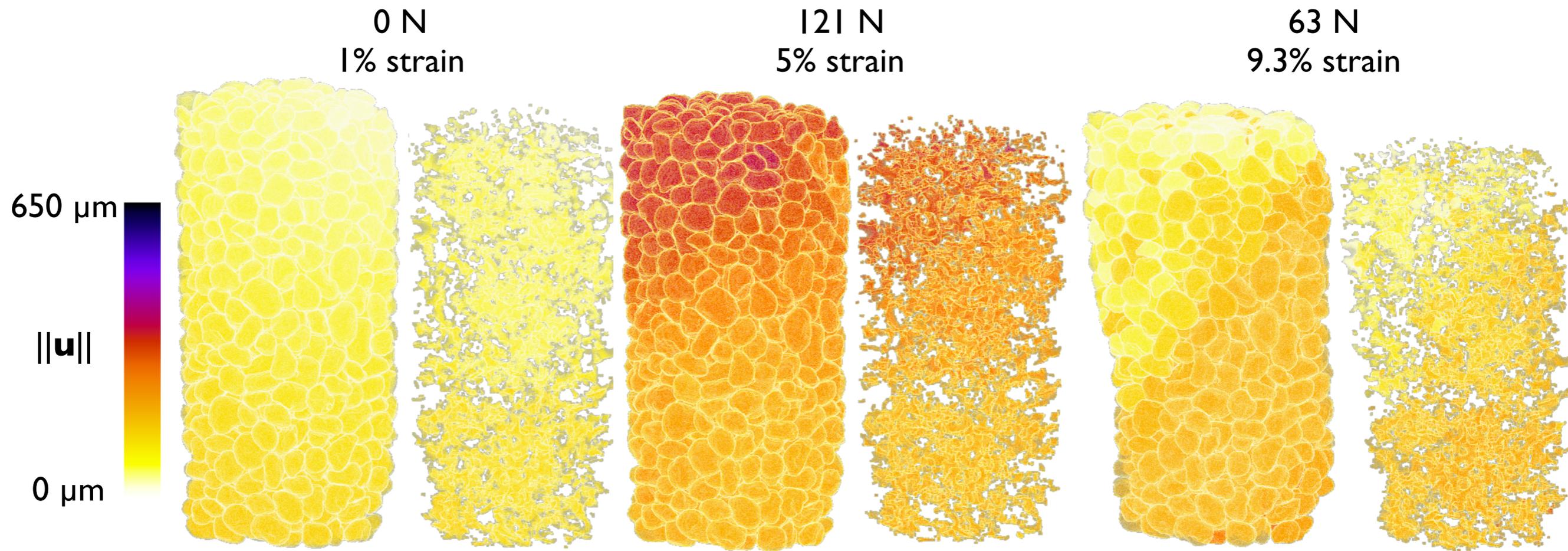
Total displacement of *both* particles and cement bridges can be used to quantify relative contribution of adhesive/cohesive/fracture modes to overall elastic and fracture properties of bulk.

Can evaluate at each contact

[Zhai & Hurley, *unpublished*, 2020. DVC code: Ando, *et al., unpublished*]

Cohesive cementation: fracture (*K, G*) properties? (2) Particle/contact-scale fracture/fragmentation modes? (3) Non-uniform strain/stress fields?

Particles: single-crystal quartz, ball milled, sieved between 150-277 μm
Sample 1: ~2,000 particles, 3.72 mm tall, 1.7 mm diam., strained to > 20%
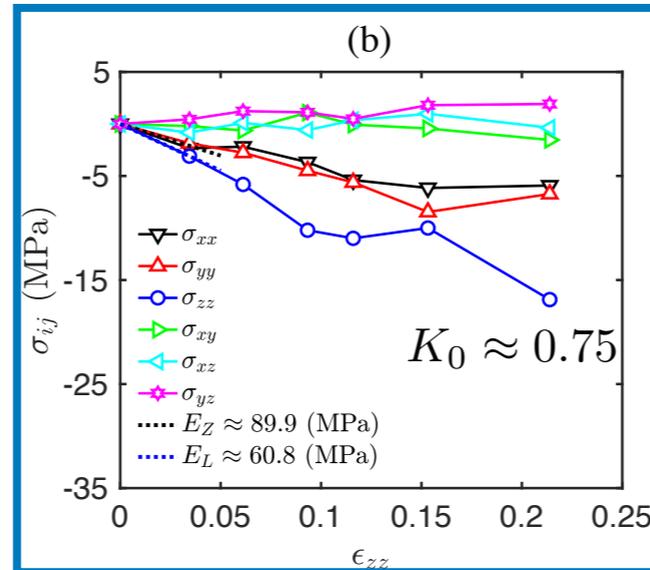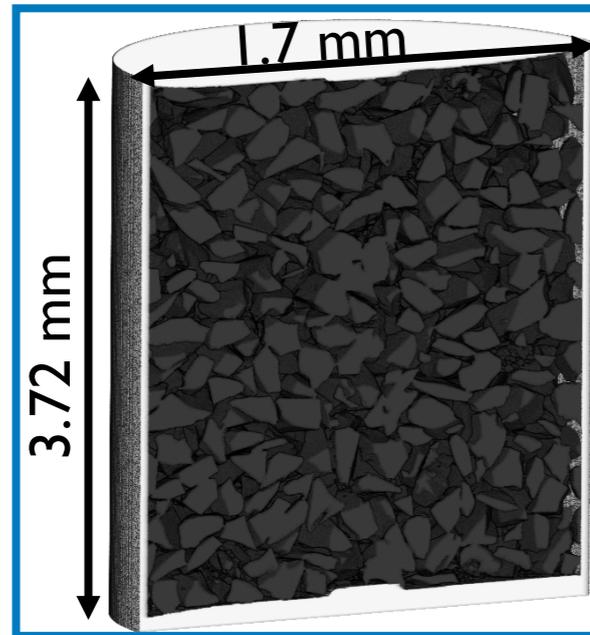Sample 2: ~1,700 particles, 2.98 mm tall, 1.7 mm diam., strained to > 4%

Uncemented sample of quartz

Epoxy-cemented (10%) sample of quartz
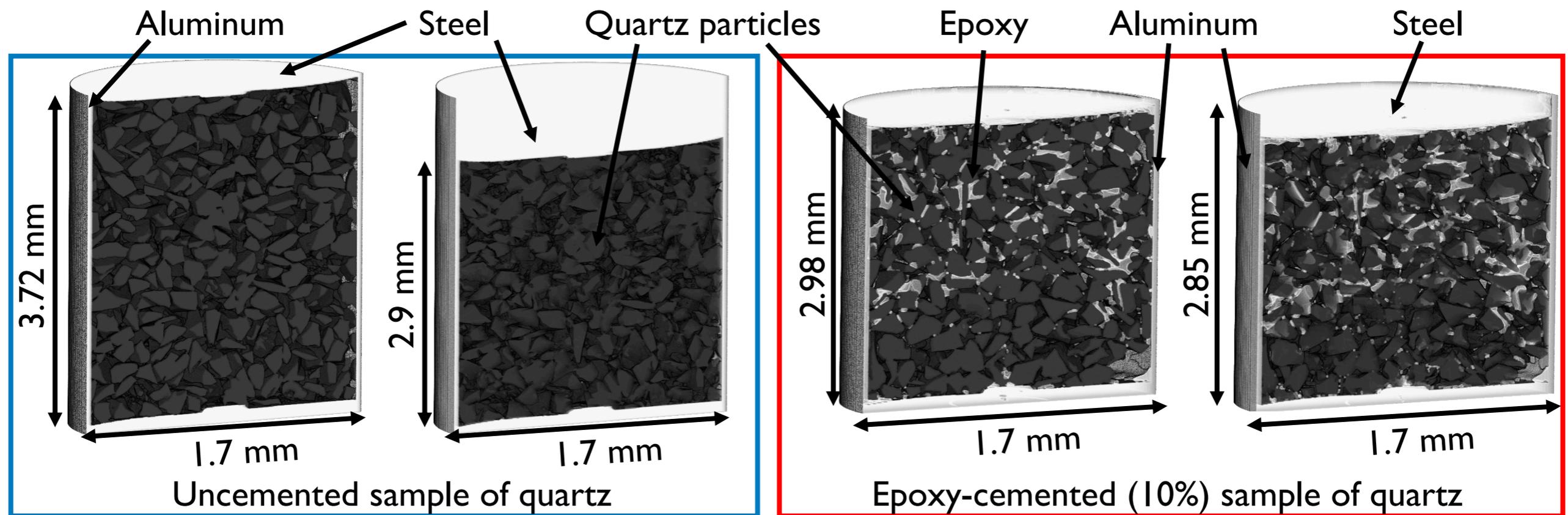
[Zhai, *et al., JOM*, 2020.]

Comparison of bulk behavior: epoxy-cemented sample is stiffer, has a lower K_0 (proportionality) value (ratio of lateral to axial stress)
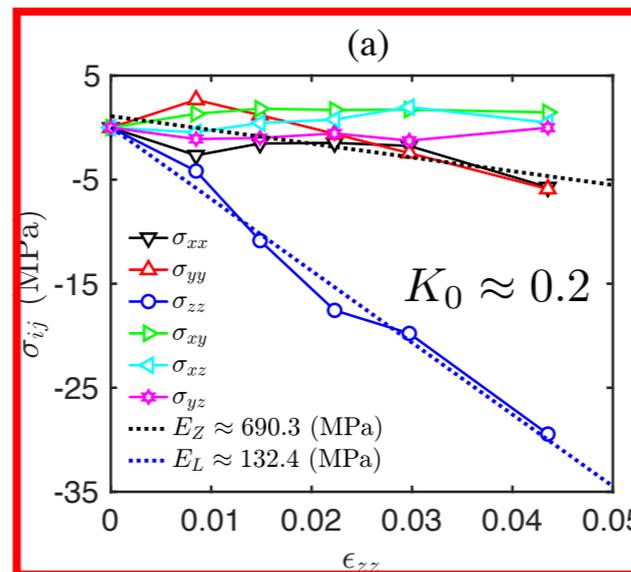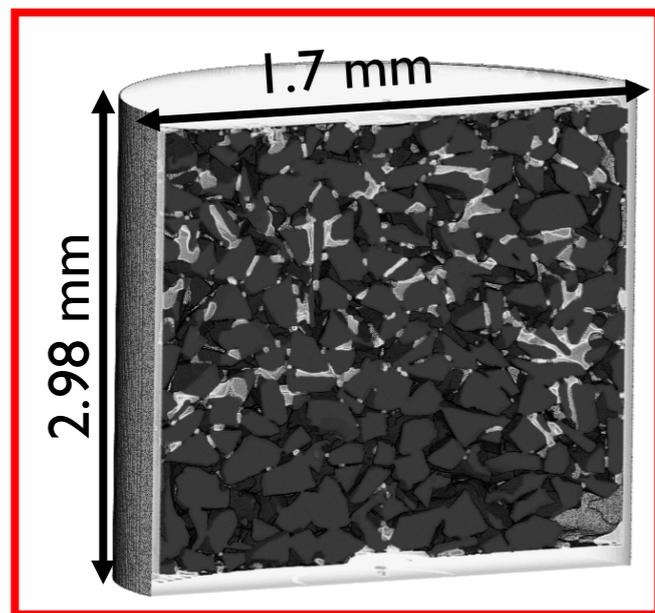

Uncemented sample

$E_Z = 89.9$ MPa

$E_L = 60.8$ MPa

$K_0 = 0.75$

Dramatic shift in elastic properties with 10% binder

Epoxy-cemented sample

$E_Z = 690.3$ MPa

$E_L = 132.4$ MPa
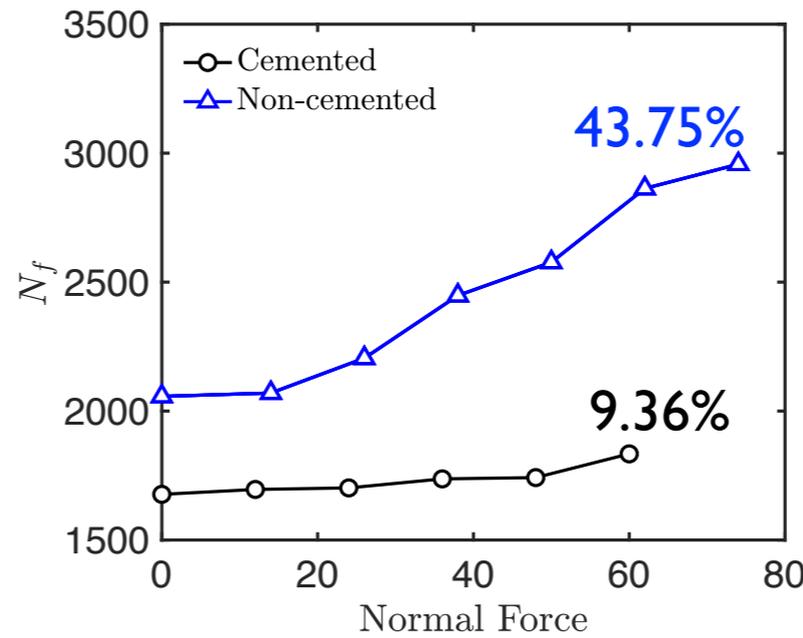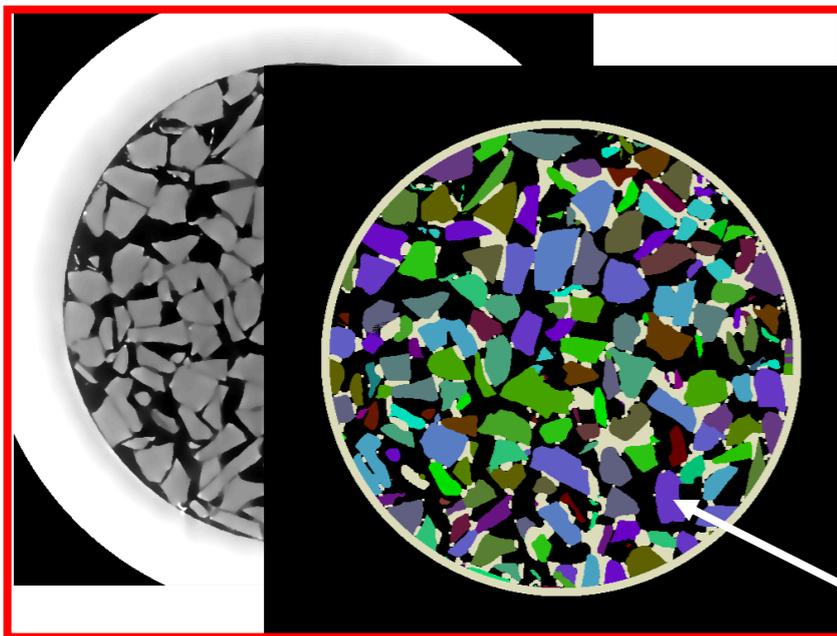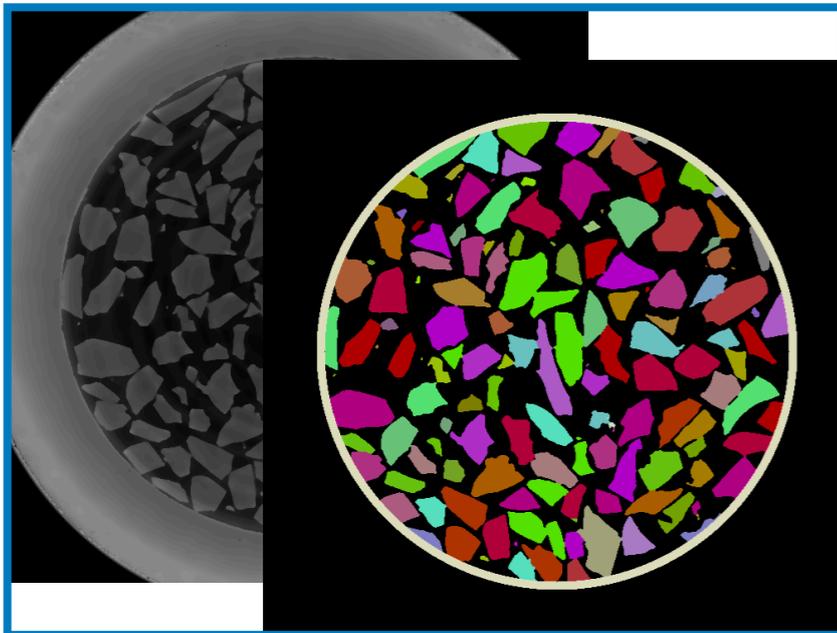
$K_0 = 0.2$

[Zhai, *et al., JOM,* 2020.]

Microstructural characterization: XRCT allows particle, cement-bridge segmentation (via MATLAB or SPAM), analysis of fragmentation, kinematics

Non-cemented: Significant more particle & relative-particle motion

Significantly more fragmentation in non-cemented sample

Each with unique label
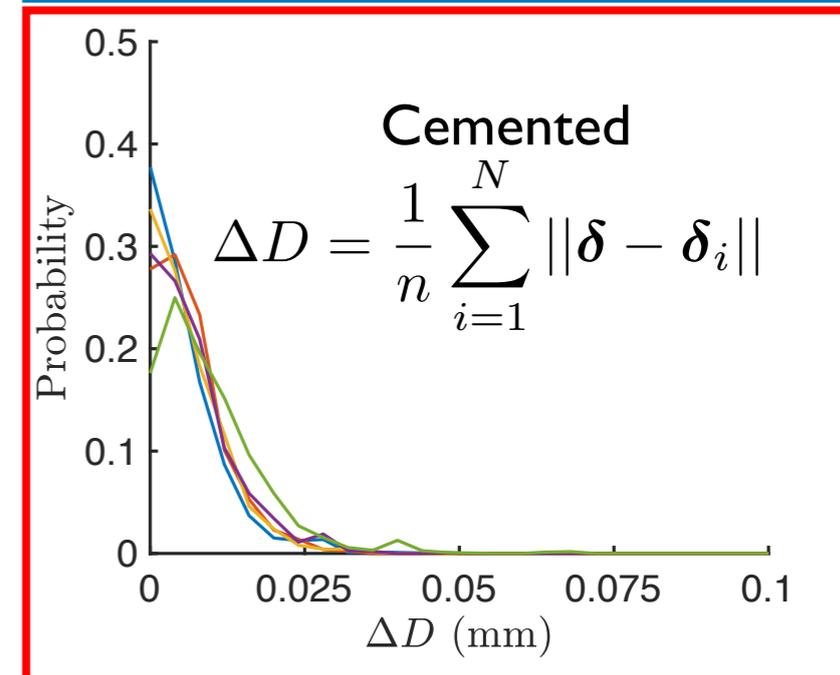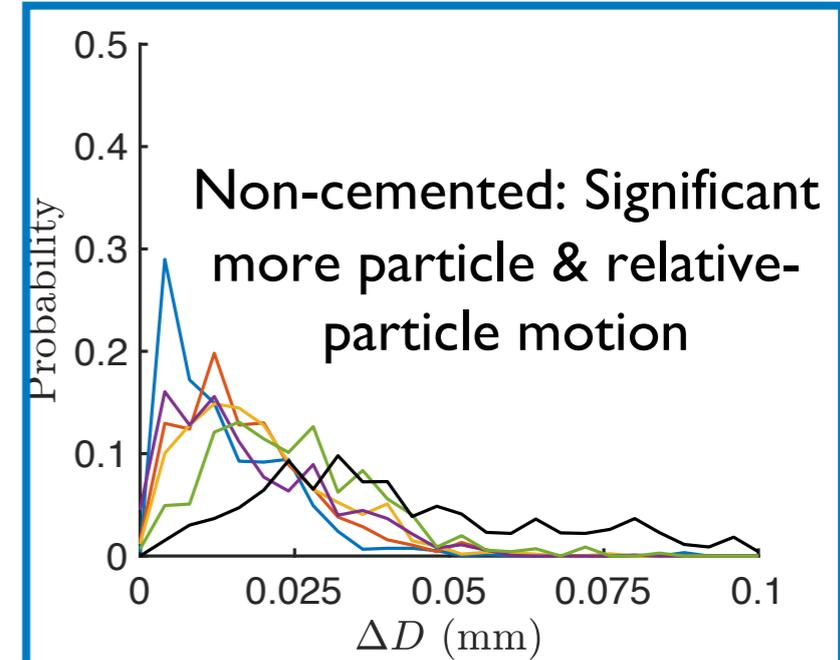
Cemented

$$\Delta D = \frac{1}{n} \sum_{i=1}^{N} ||\boldsymbol{\delta} - \boldsymbol{\delta}_i||$$

43.75%

9.36%

[Zhai, *et al., JOM*, 2020.]

Microstructural characterization: 3DXRD allows calculation of intra-particle stress tensors and assessing local stresses vs bridges, co-axiality, etc.

15% strain: particles' stress *not* co-axial sample strain

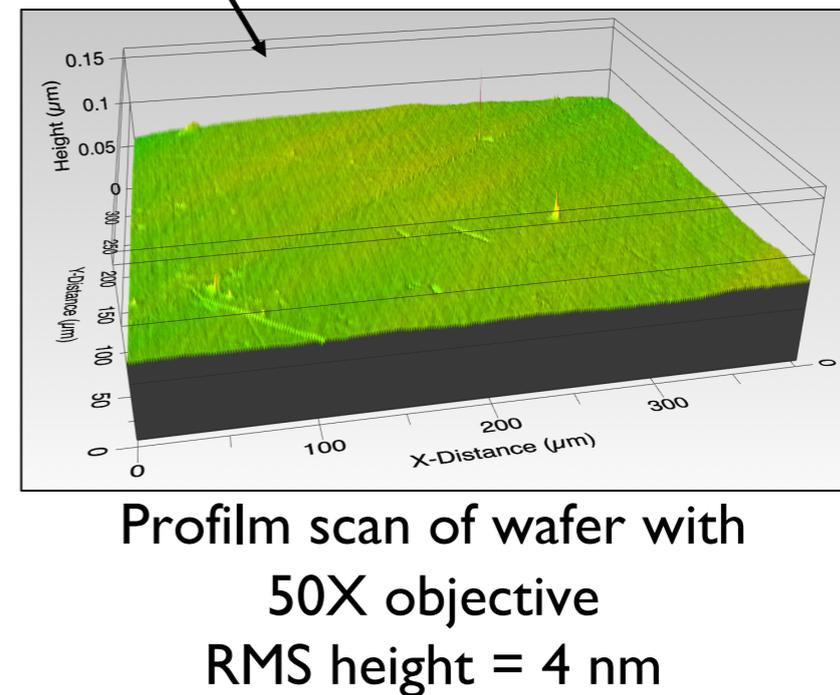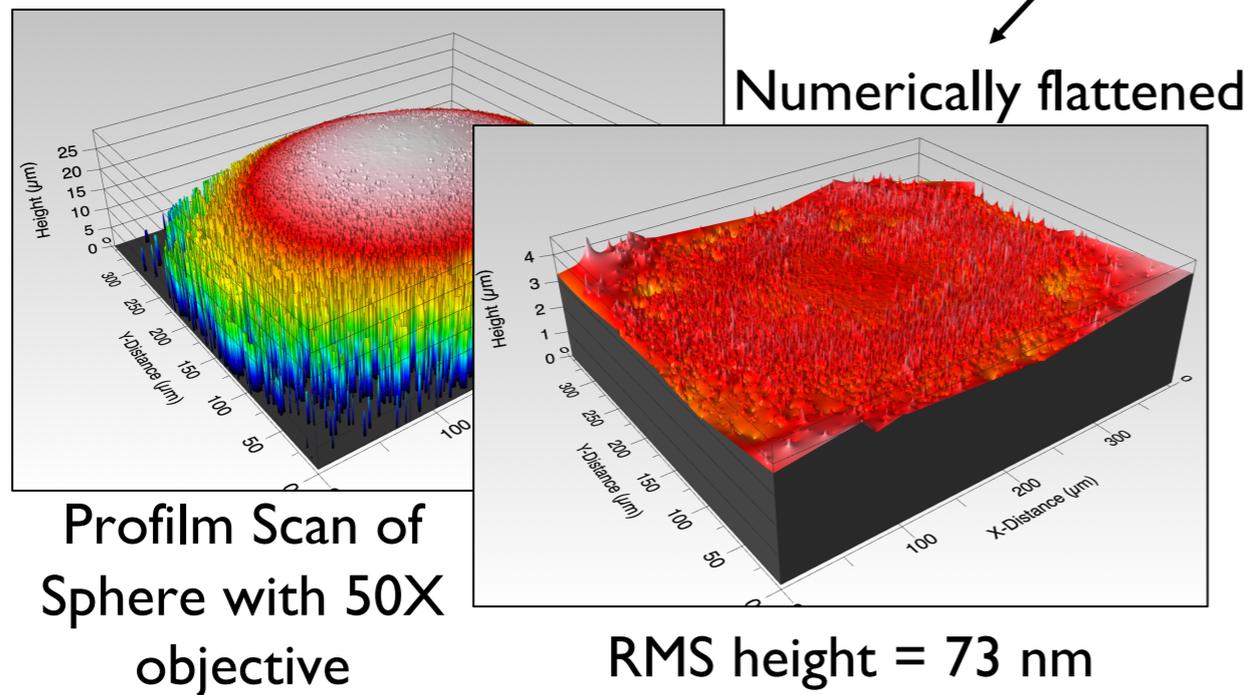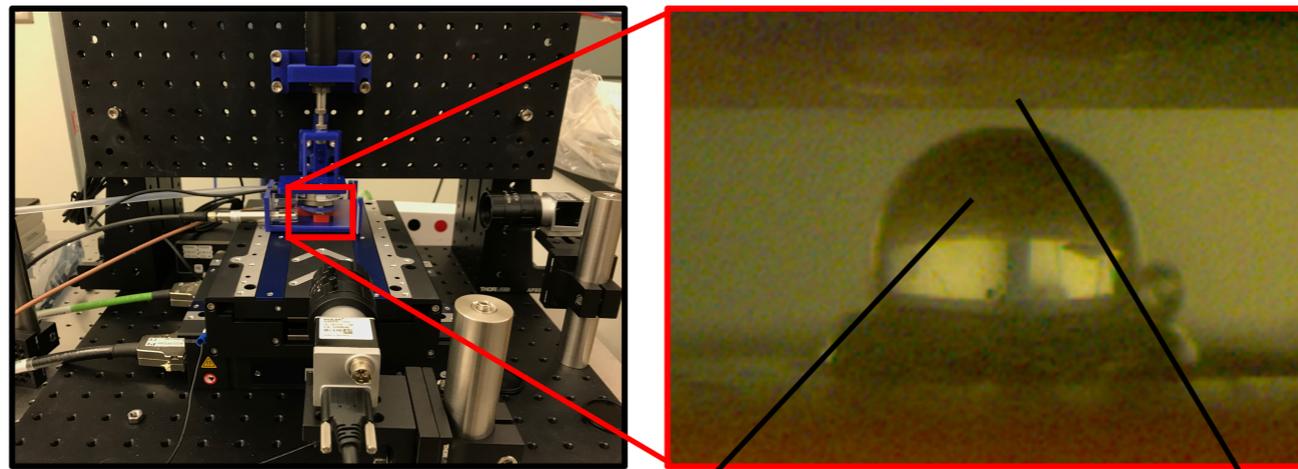<1% strain: particles' stress co-axial w/ sample strain
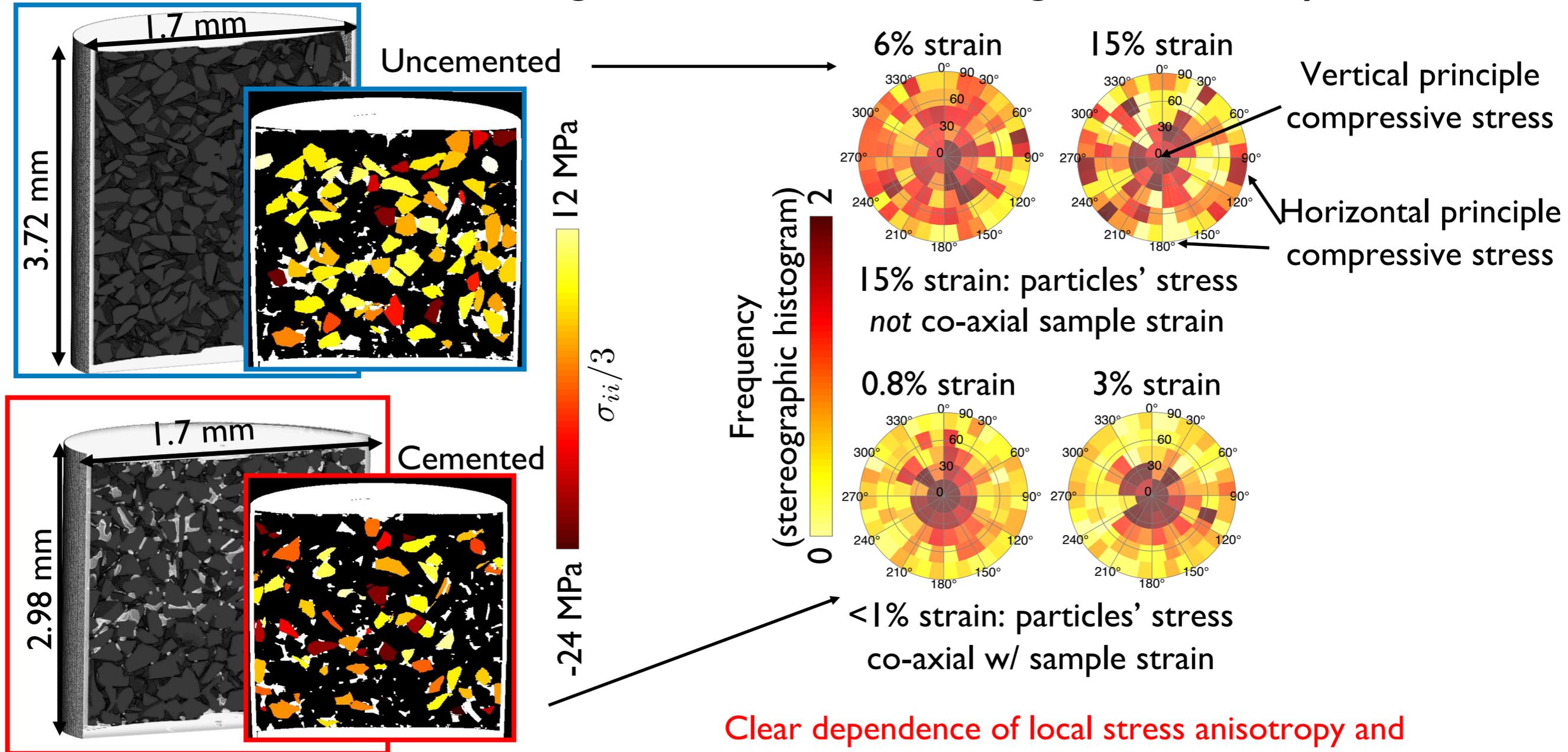
Clear dependence of local stress anisotropy and homogeneity with cementation. Can also look at force chains as a function of cementation.

[Zhai, *et al., JOM,* 2020.]

<u>Addressing humidity/roughness:</u> (1) Adhesion and friction forces as a function of humidity & roughness at both contact- and bulk scales?

<u>Test 1:</u> compression and sliding of 2 mm borosilicate sphere on a quartz wafer at 35%, 55%, 75% relative humidity

Numerically flattened

Profilm Scan of Sphere with 50X objective

RMS height = 73 nm

Profilm scan of wafer with 50X objective
RMS height = 4 nm

Addressing humidity/roughness: (1) Adhesion and friction forces as a function of humidity & roughness at both contact- and bulk scales?

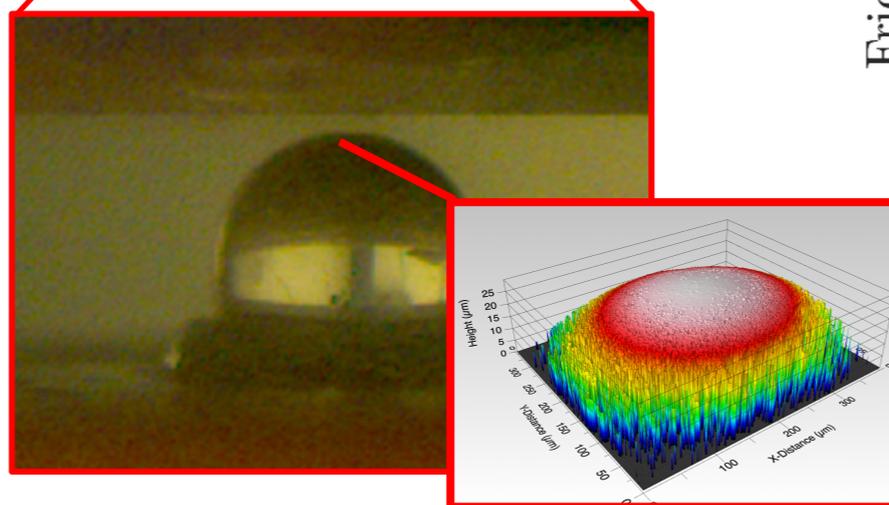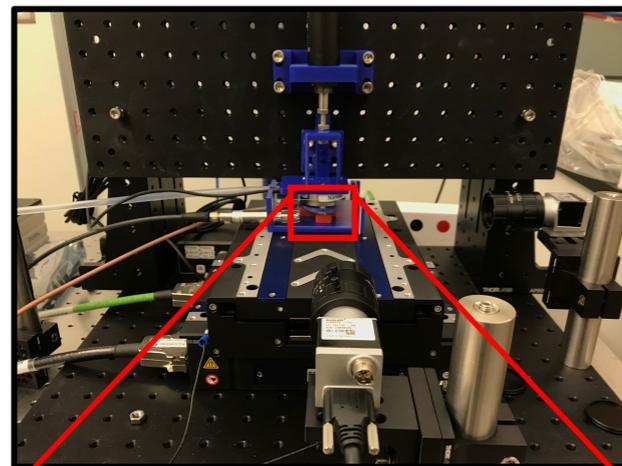Test 1: compression and sliding of 2 mm borosilicate sphere on a quartz wafer at 35%, 55%, 75% relative humidity
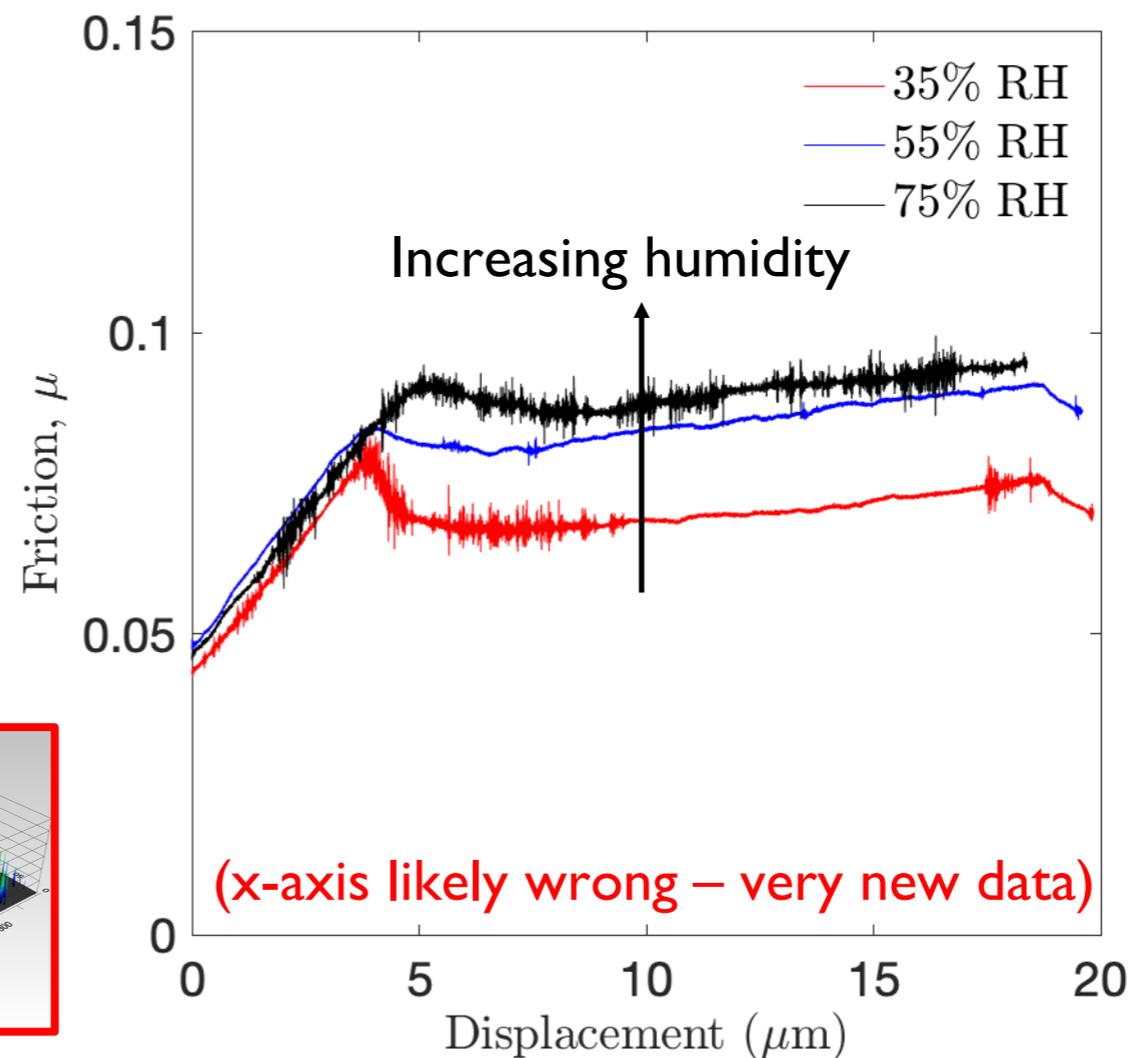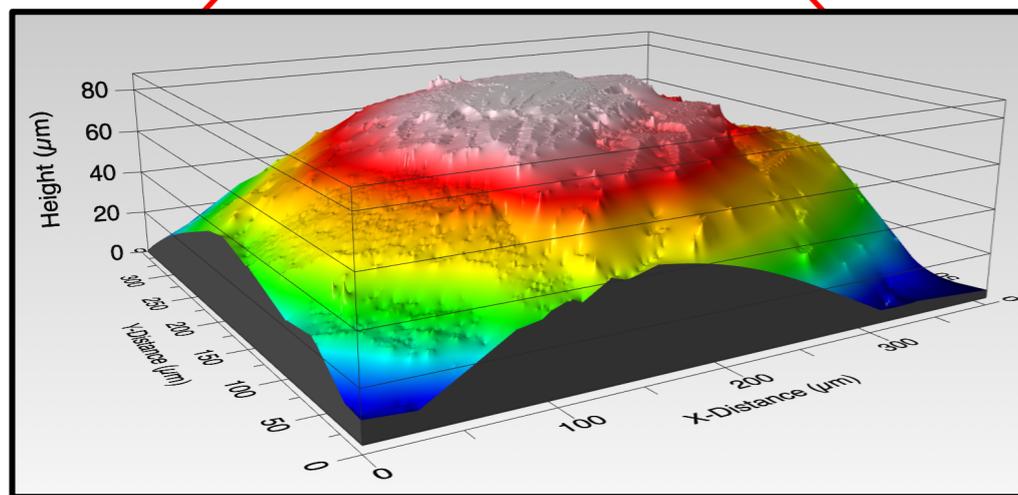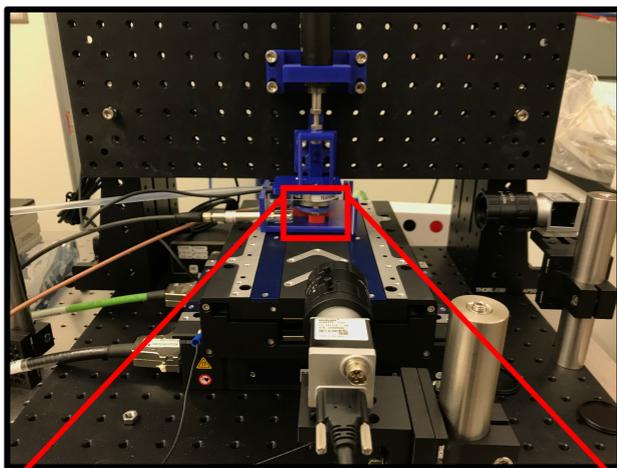

RMS height = 73 nm

Increasing humidity

(x-axis likely wrong – very new data)

Compression followed by shear shows minor but clear humidity dependence at 4 N compressive

<u>Addressing humidity/roughness:</u> (1) Adhesion and friction forces as a function of humidity & roughness at both contact- and bulk scales?

<u>Test 2:</u> compression and sliding of 0.8 mm Ottawa sand particle on a quartz wafer at 35%, 55%, 75% relative humidity
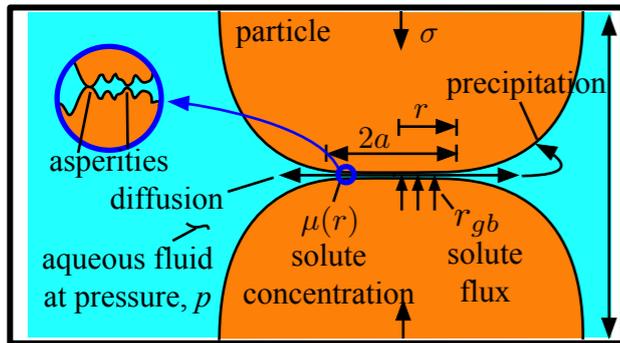

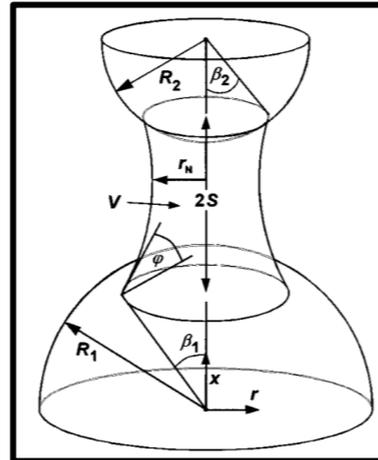RMS height on contact: 1.067 µm
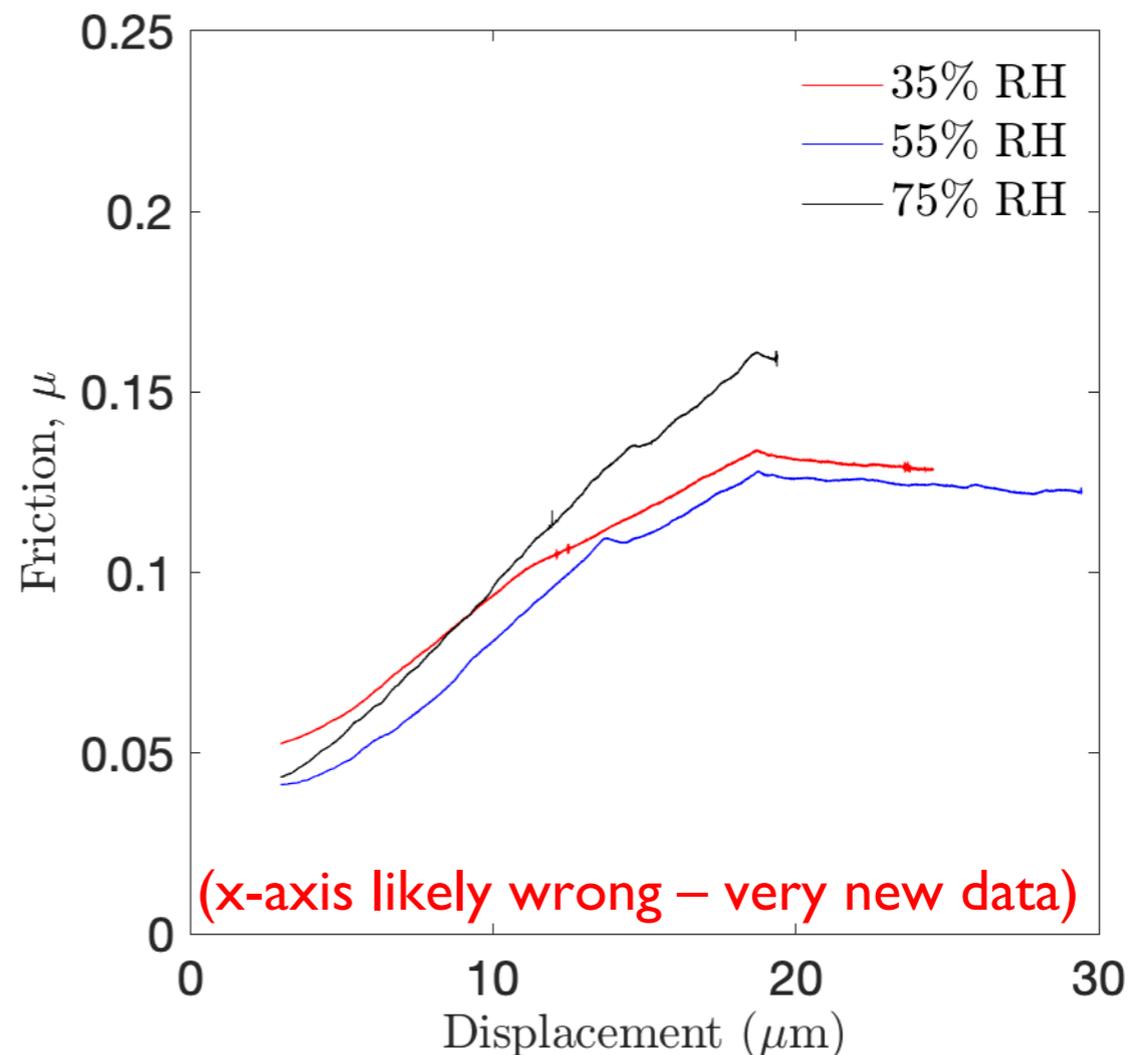
Compression followed by shear shows minimal and mixed humidity dependence at 4 N compressive
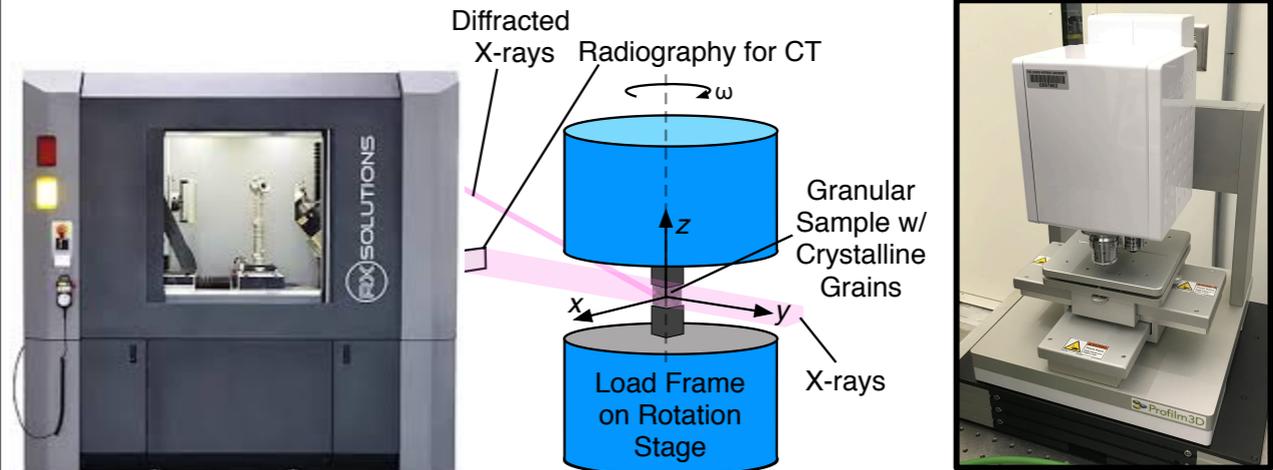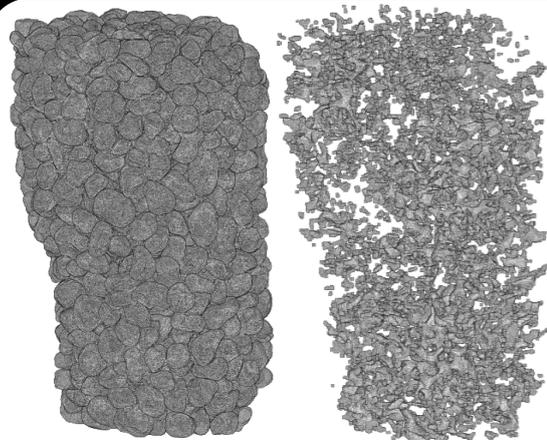
Outline

Inter-particle cementation

Capillary condensation

1. Mechanisms, Challenges

Diffracted X-rays   Radiography for CT

Granular Sample w/ Crystalline Grains
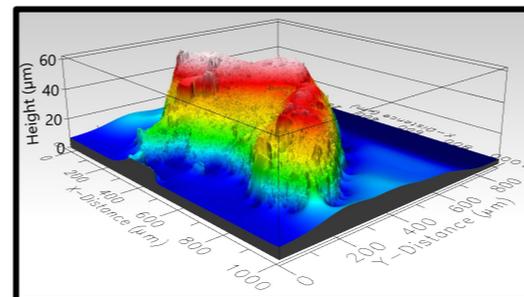
Load Frame on Rotation Stage   X-rays

2. Characterization Tools

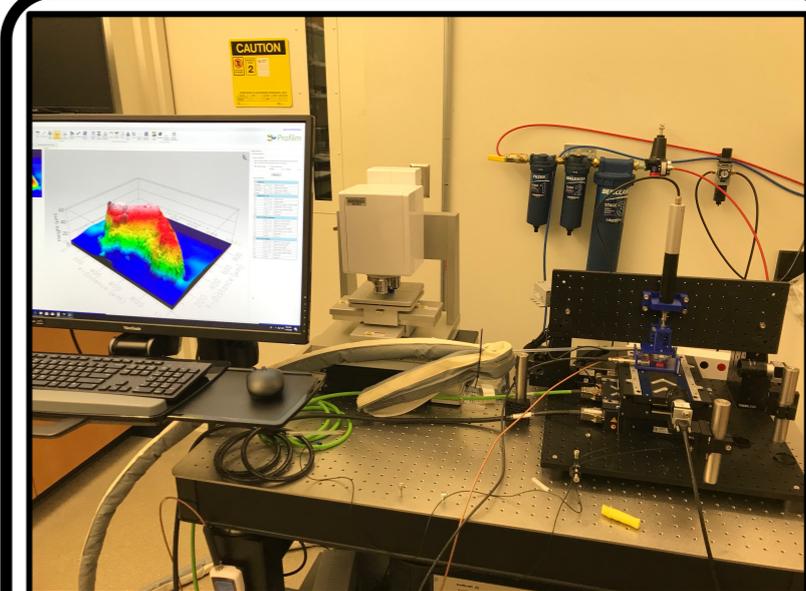(1) XRCT: cemented cohesive mechanics

(2) XRCT + 3DXRD: particle-level stresses

(3) Roughness, %RH effects on particle-scale interactions

3. Past & Future Applications
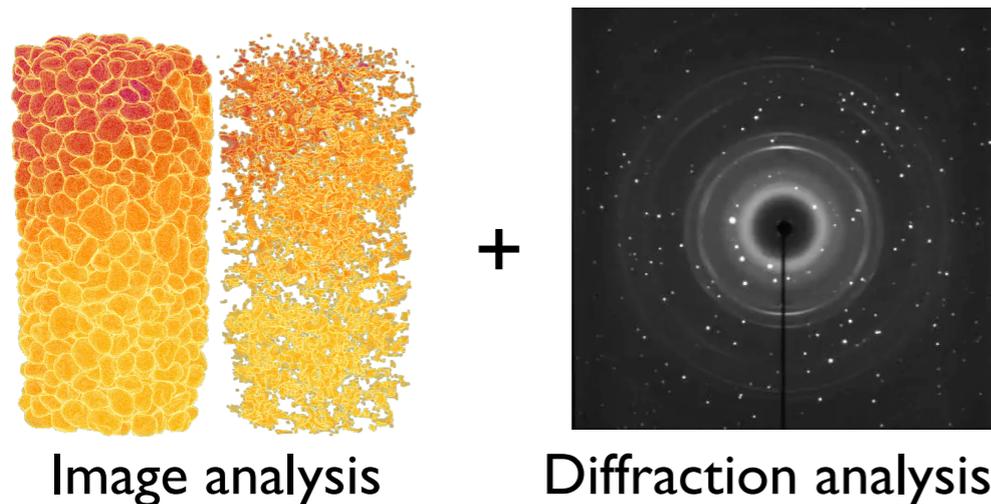

4. Summary, Goals

Where are we going next?

Cementation:

- Using XRCT & 3DXRD, we are developing tools to quantify microscale contributions to elastic & fracture properties of cemented granular media.


Image analysis + Diffraction analysis

- Upcoming synchrotron experiments (Feb 2020): XRCT & 3DXRD to quantify microscopic contributions to failure in cemented materials.
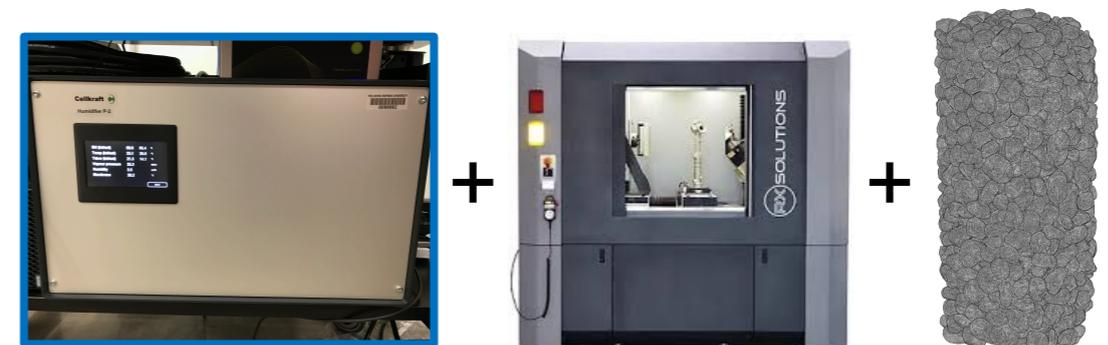

0 — $-\sigma_{ii}/3$ (MPa) — 75
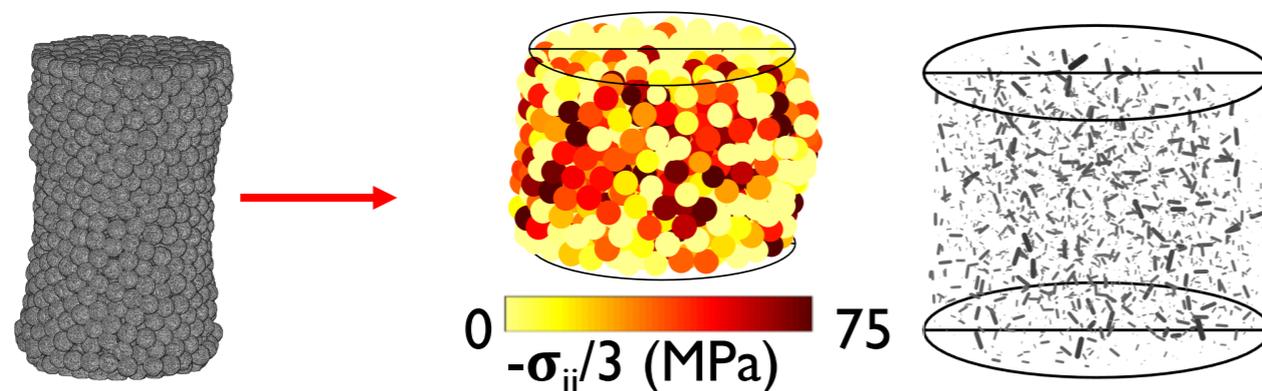
Humidity/roughness:

- A new capability for surface structure analysis (profilometry) and in-situ contact (friction/adhesion) testing at any humidity will elucidate cohesion mechanisms across scales.
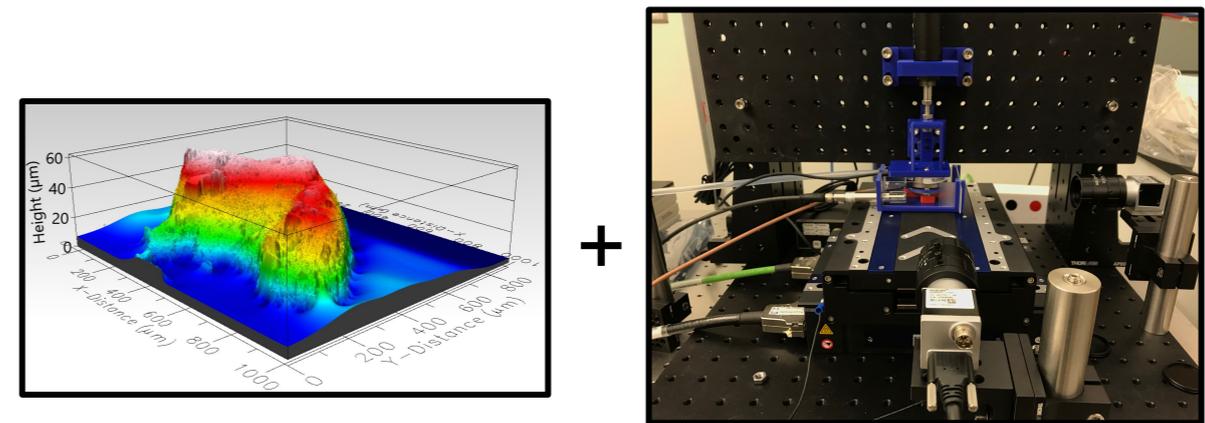
- Future work may combine contact-scale with bulk-scale 3D XRCT studies of humid powders.

Thank you!

ARO DURIP Award

CHESS

Johns Hopkins
University

Ryan C. Hurley[1]
[1]Mechanical Engineering, Hopkins Extreme Materials Institute, Johns Hopkins University

Thanks to: Chongpu Zhai, Postdoc
Hopkins Extreme Materials Institute, Johns Hopkins University