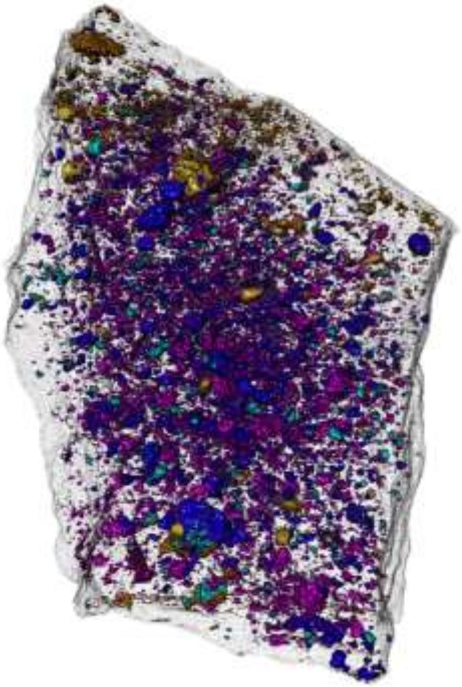
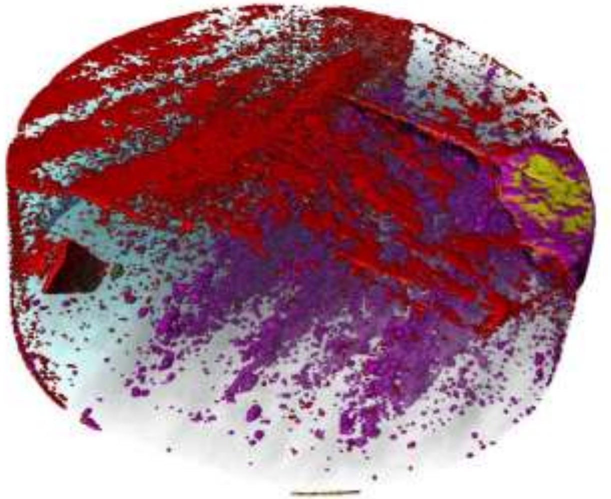
Emerging Neutron Imaging Methods

(focused mostly on geosciences, mostly…)





Jacob M. LaManna

Cyrus Daugherty, Youngju Kim, Victoria DiStefano, Dan Hussey, Eli Baltic, David Jacobson

Physical Measurement Laboratory, National Institute of Standards and Technology

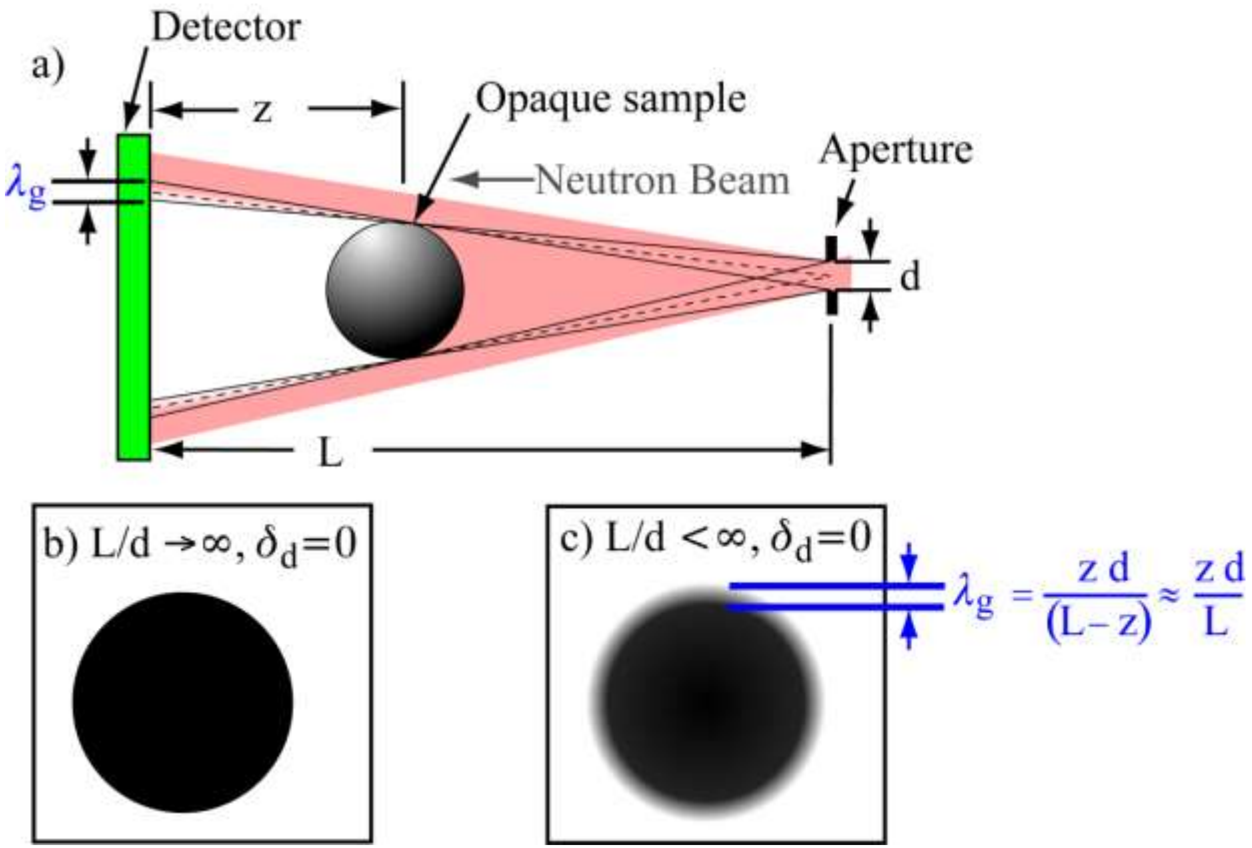
Overview

- How do we more efficiently use neutrons?
  - Wolter Optics
- How do we see smaller features?
  - Grating Interferometry
- How do we identify minerals?
  - Bragg Edge Imaging
- How do we enhance contrast in samples?
  - NeXT
- How do we go faster with current beamlines?
  - Improved reconstruction algorithms

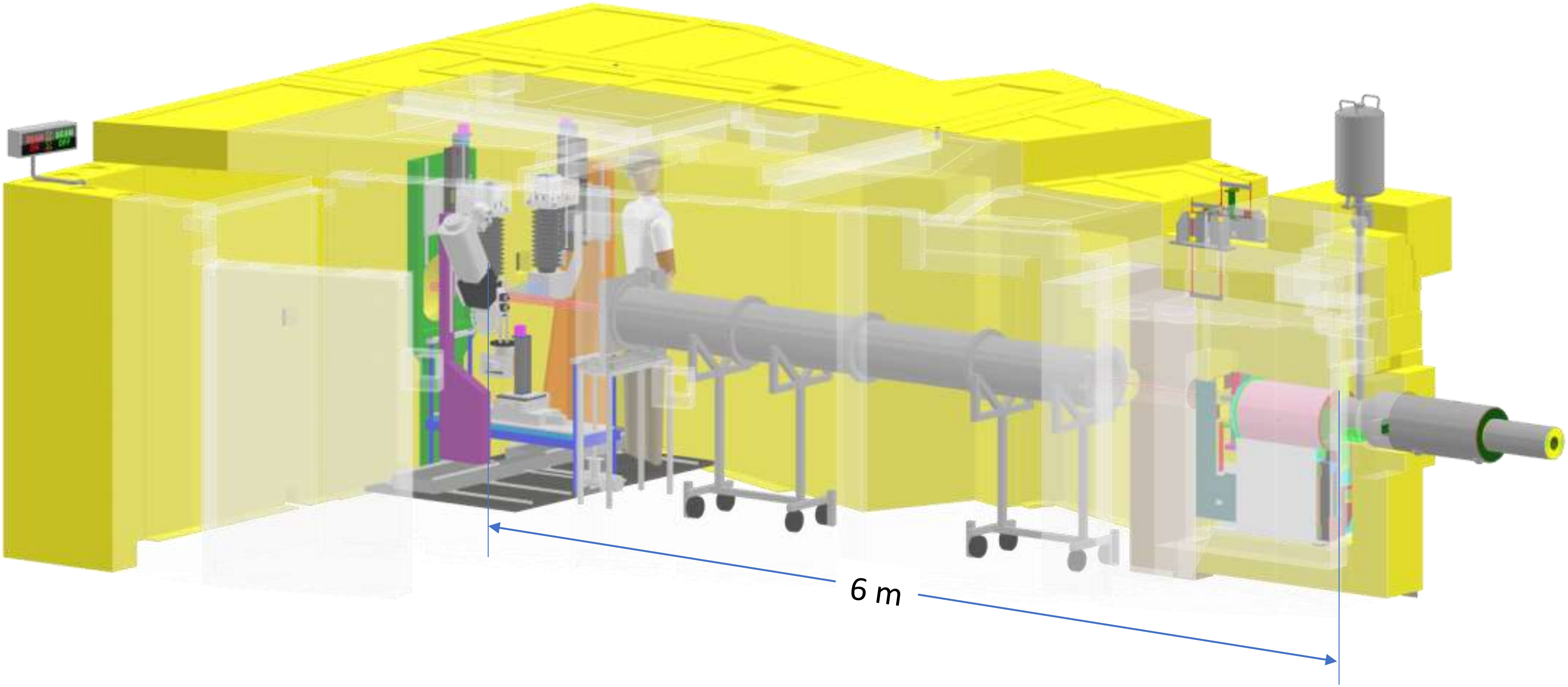
The Limits of Pinhole Optics and Conventional Neutron Imaging

- Poke hole in reactor wall, form image of core at detector
- Best resolution when object ***contacts*** detector due to ~cm sized apertures
  - No geometric magnification
- Resolution derived from collimation, producing geometric blur:

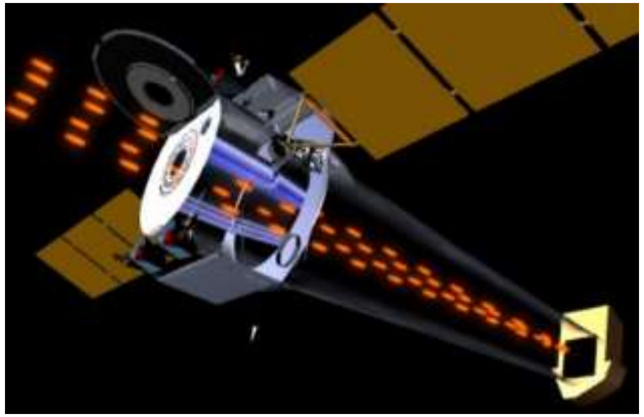
  $\lambda_g \approx z\, d\, /\, L$
- Flux goes as $(d/L)^2$, Small d and/or large L → small Flux → ☹
- Even with better detectors, in a 1 µm pixel with flux 10^6 $\mathrm{cm}^{-2}\ \mathrm{s}^{-1}$, there's only 1 neutron every 100 s. ☹☹



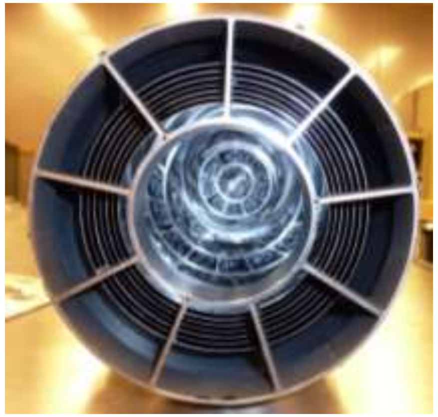
Finite instrument length necessitates variable apertures



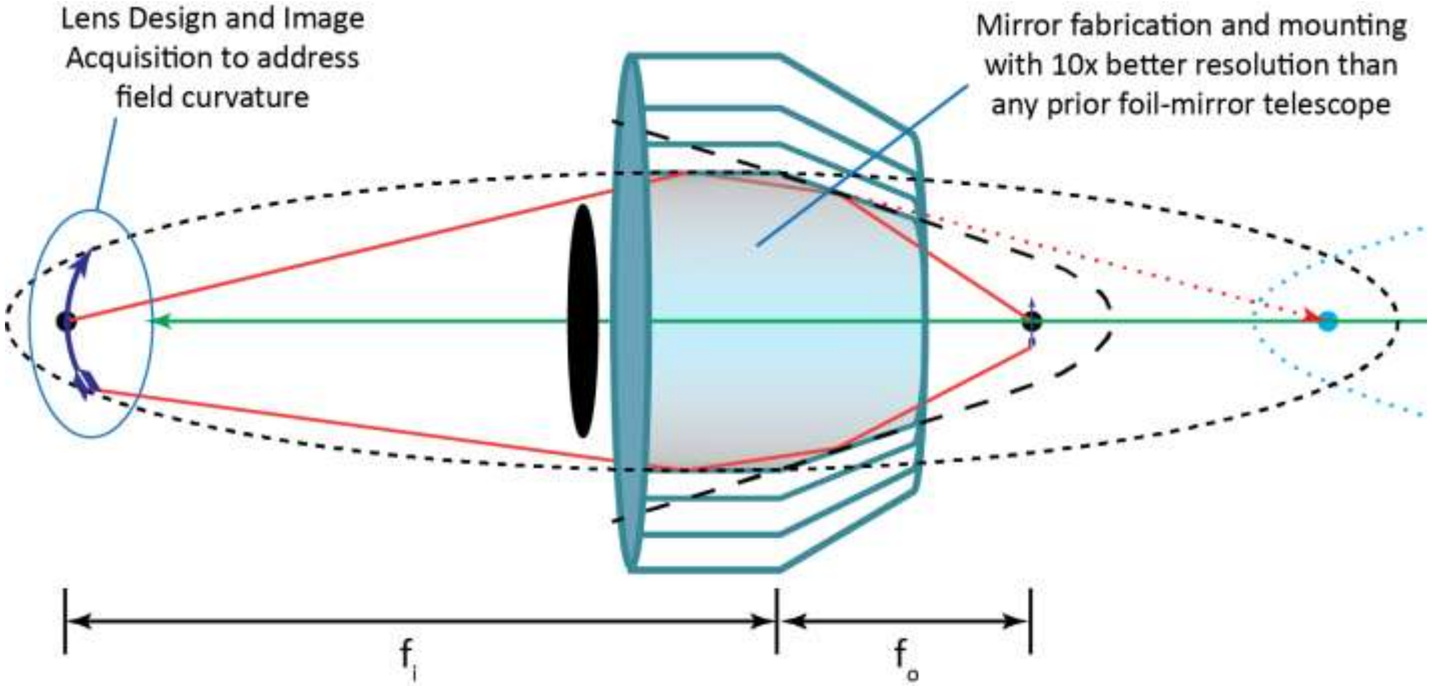
Transforming x-ray telescopes into neutron microscopes



Wolter Optics power CHANDRA



NiCo-foil Focused
X-ray Solar Imager



Lens Design and Image Acquisition to address field curvature

Mirror fabrication and mounting with 10x better resolution than any prior foil-mirror telescope

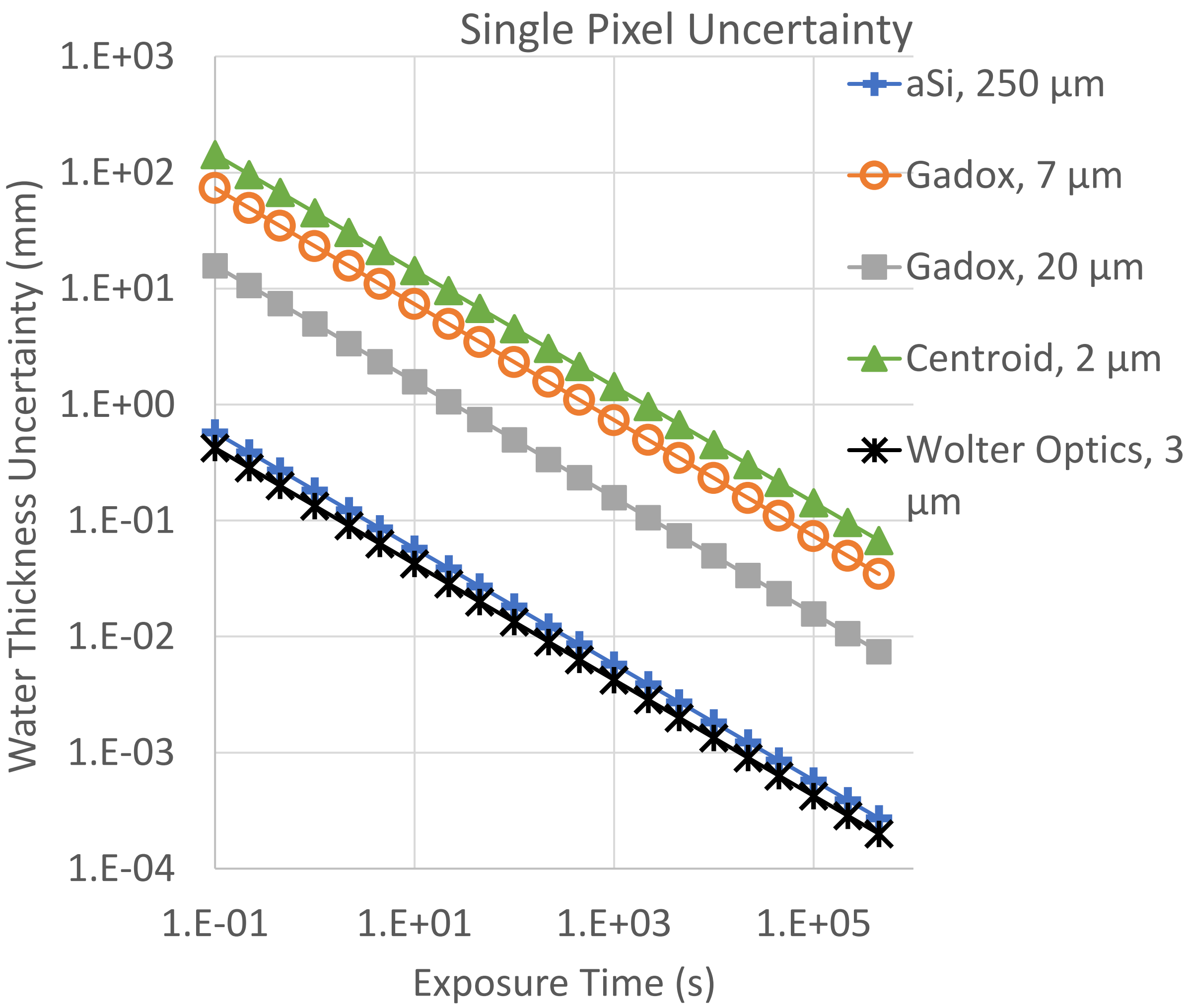
The neutron lens is based on mirror foil Wolter Optics:

- Need to realize 1 arcsec angular resolution
- ***x1000 flux***
- Image magnification for ***spatial resolution of 3 μm***
- Achromatic lens
- ~1 m separation between lens, object, and detector

*Win-win over pinhole cameras: boosts intensity **and** resolution*

D. Liu et al, Appl. Phys. Lett. 102, 183508 (2013); doi: 10.1063/1.4804178

Liquid water uncertainty for various methods/detectors



- Time and Spatial Resolution would approach a conventional synchrotron imaging beamline
- Sample environments like Furnaces, Griggs Rigs, Pressurized Fluid Flow Cells, Magnets, Cryostats,... will be straightforward to incorporate on the beamline
- Can improve quantitative analysis using a velocity selector to coarsely define the wavelength band Δλ/λ ~ 10-15%

DS Hussey et al NIMA (2021) 987, 164813, doi: 10.1016/j.nima.2020.164813

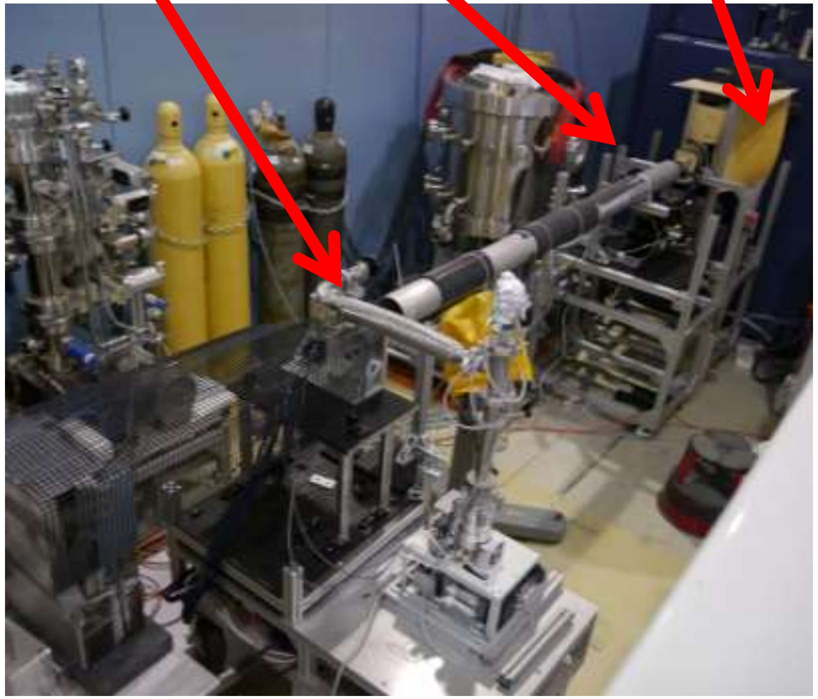
Pinhole Optics vs. Wolter Optics layout

Load Frame and Pressure Vessel

Detector

Lens

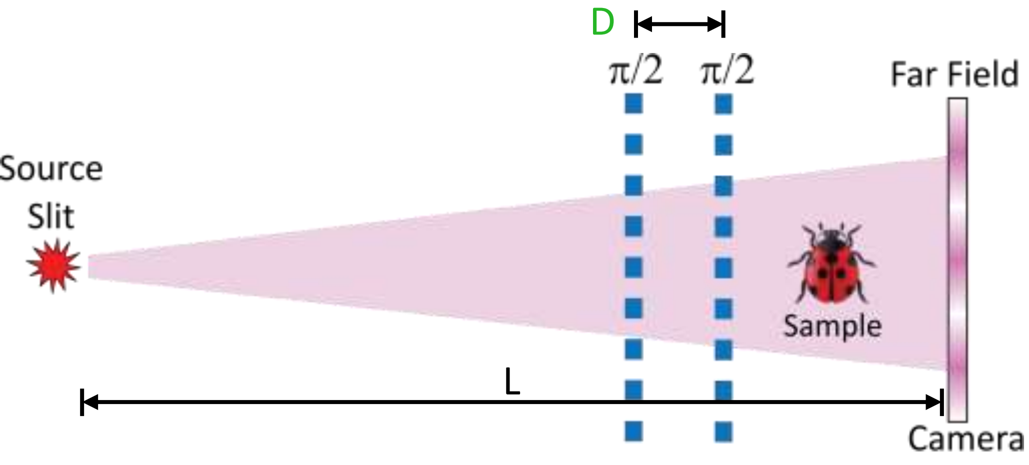
~~Fuel Cell~~



Wolter Optics Setup: 60 cm between sample and lens, 2.5 m between lens and detector

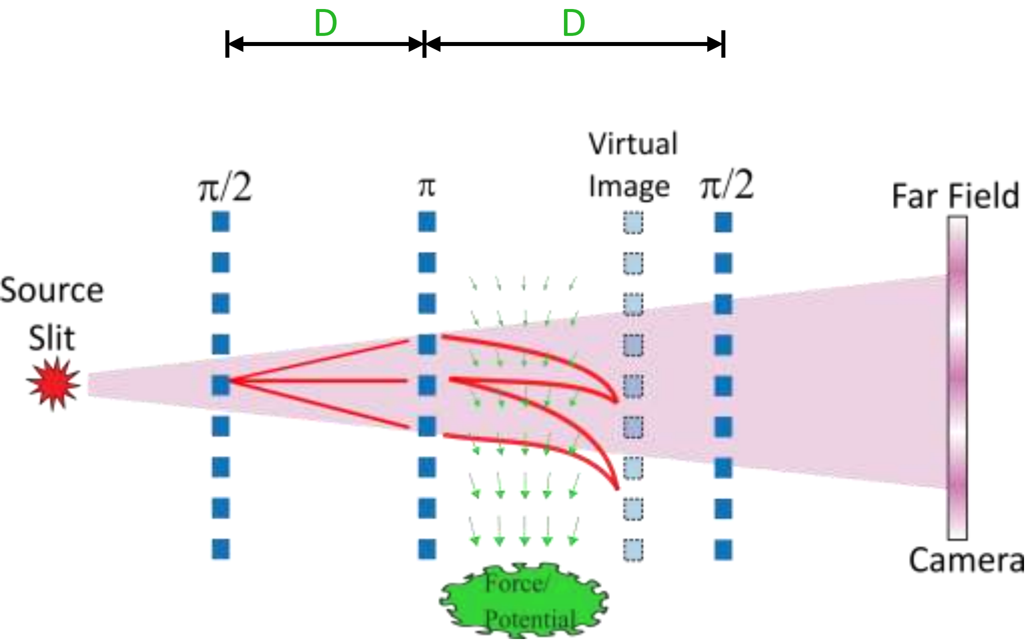
D. Liu et al, Appl. Phys. Lett. 102, 183508 (2013); doi: 10.1063/1.4804178

Novel Neutron Imaging Far Field Interferometer



2-Grating Geometry

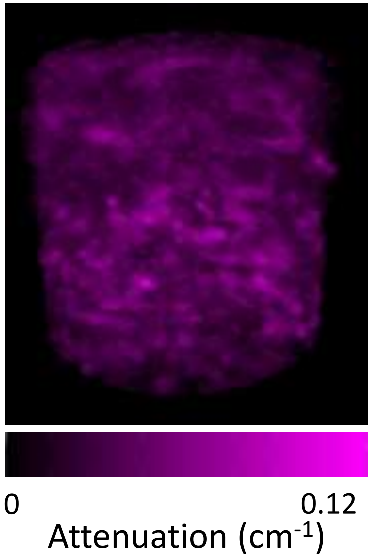
- D = [0.05 to 5 cm]
- $P_D = P_G L / D$
- Sample microstructure reduces visibility
- ***Tunable period probes $1\text{-}10^4$ nm***



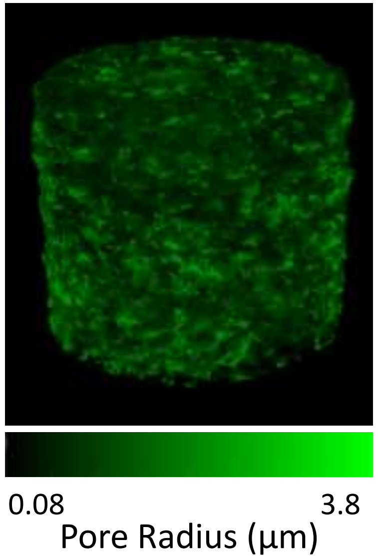
3-Grating Geometry

- First practicable meters long neutron interferometer
  - ➢ Enables measure of Big “G”

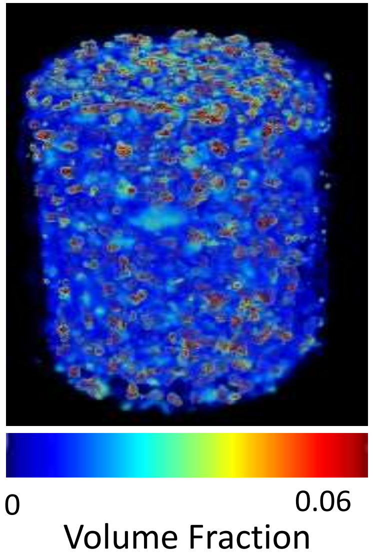
1 cm diameter core sample



Attenuation (cm^{-1})



Pore Radius (μm)

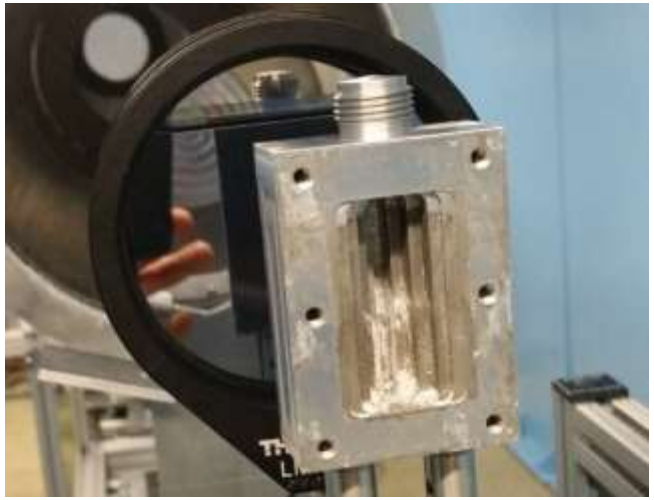


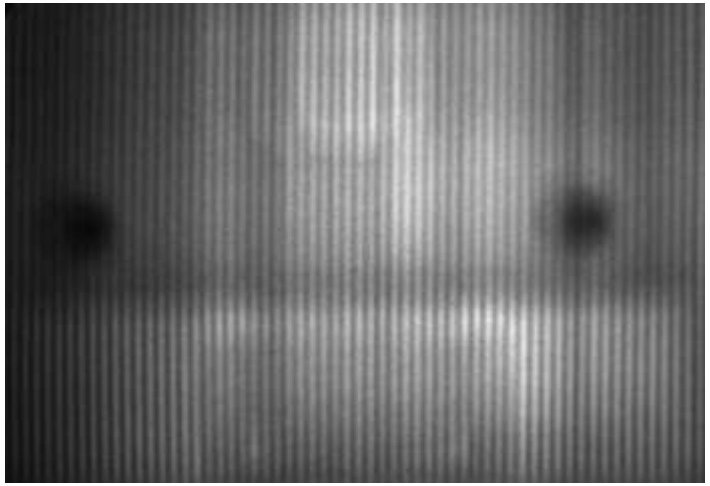
Volume Fraction

IMS-funded "INFER project" to create NCNR user instrument for “SANS-tomography” and measure G

*1st neutron demonstration performed at CNII: PRA **95**, 043637 (2017), PRL **120**, 113201 (2018)*

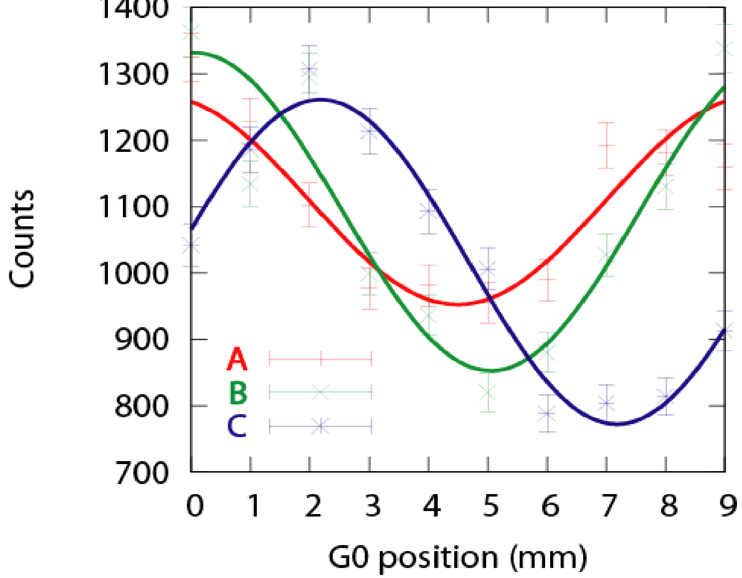
Grating Interferometry Images

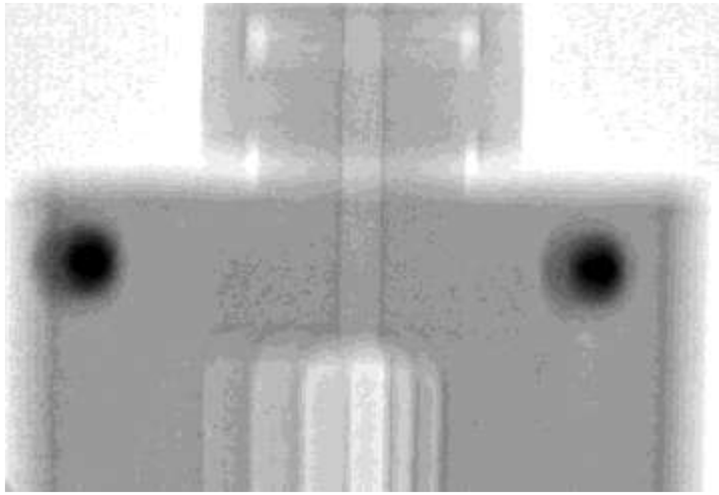




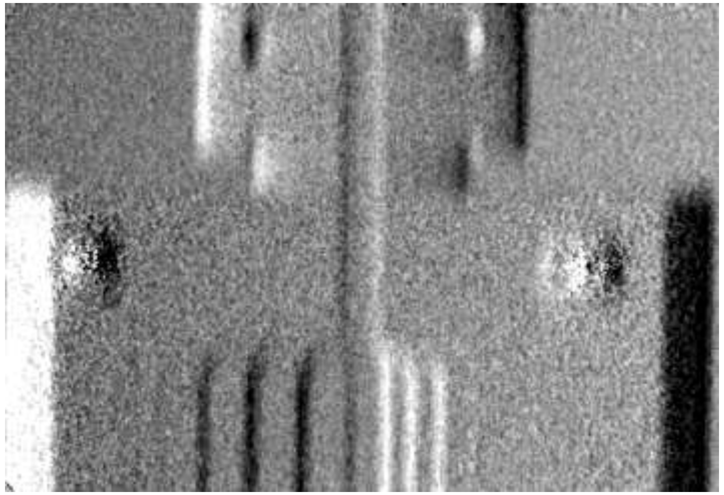
Raw Phase Step Images

Intensity Variation during a phase step scan

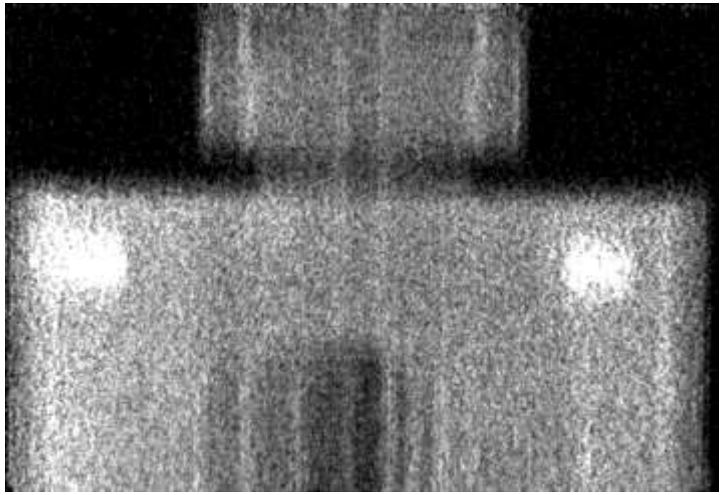




Mean, or H0



Phase Gradient



(loss of) Visibility H1
Shown is –ln(H1s/H1o)

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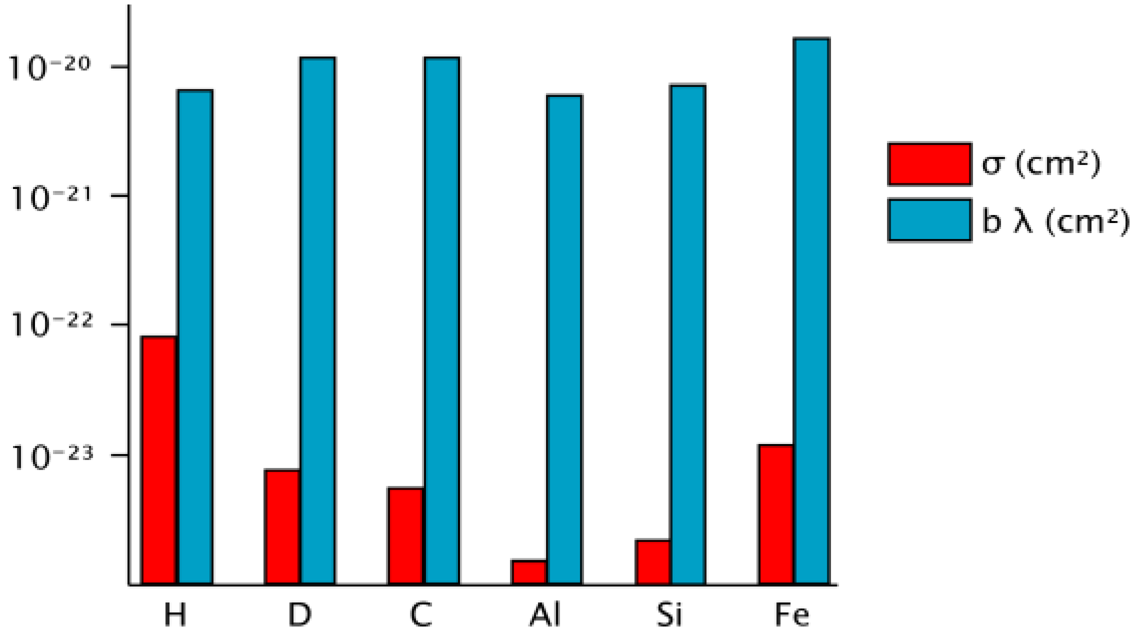
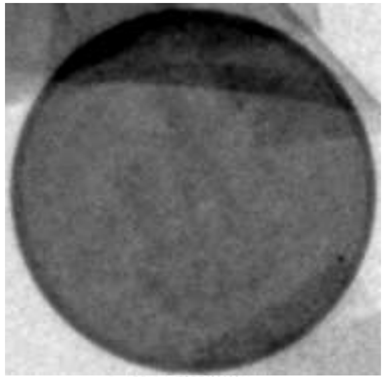
D. Pushin et al, PRA 95, 043637 (2017), DOI: 10.1103/PhysRevA.95.043637

Neutron Phase Imaging



- Attenuation radiography measures: **σ {N t}**
- Phase radiography one measures: **$b_c\ \lambda$ {N t}**
- For equal incident neutron intensities, phase imaging is ~10^3 more sensitive to changes in the number density, N, or material thickness t



Attenuation Image

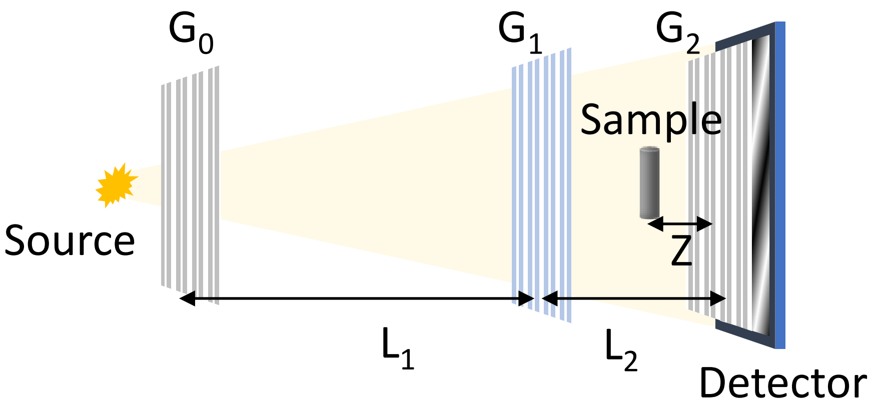
U.S. Quarter

Phase Gradient Image

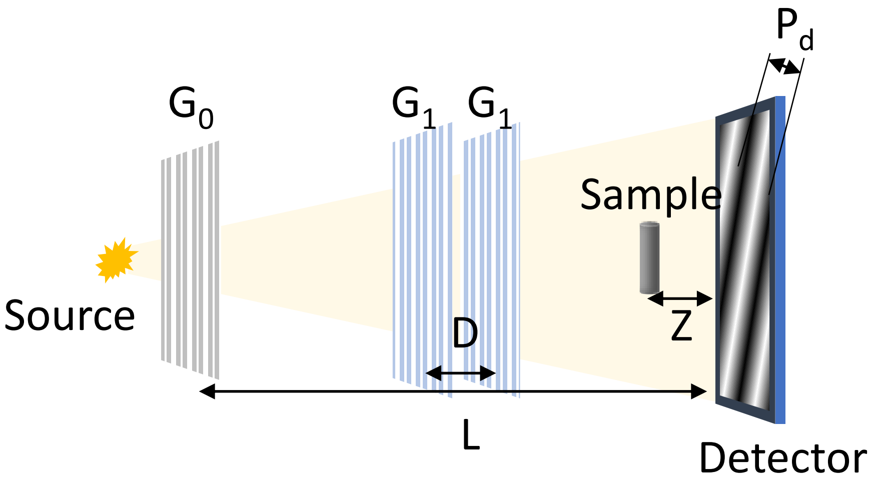
*Seung Wook Lee et al, NIMA **605** (2009)*

Grating Interferometry: Talbot-Lau and Far-field

Talbot-Lau grating Interferometer (TLI)



Far-Field Interferometer (FFI)



Autocorrelation length (ξ)

- *System parameter in grating interferometer*
- *Measurable resolution (structure size) in dark-field imaging*

$$\xi = \frac{\lambda Z}{p_d}$$

λ: wavelength
Z: sample-to-detector distance
p_d: fringe period

Fringe period: $p_d = p_2$ *(@TLI)* $\quad p_d = \frac{Lp_1}{D}$ *(@FFI)*

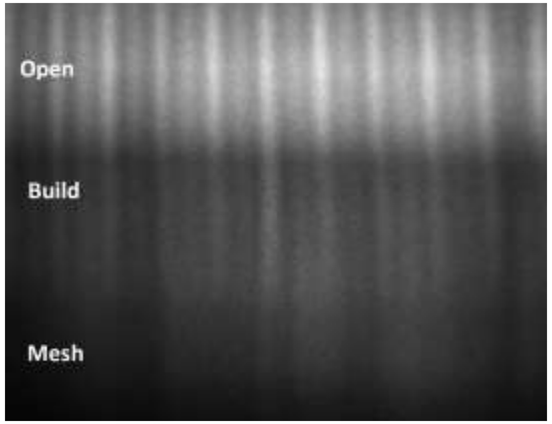
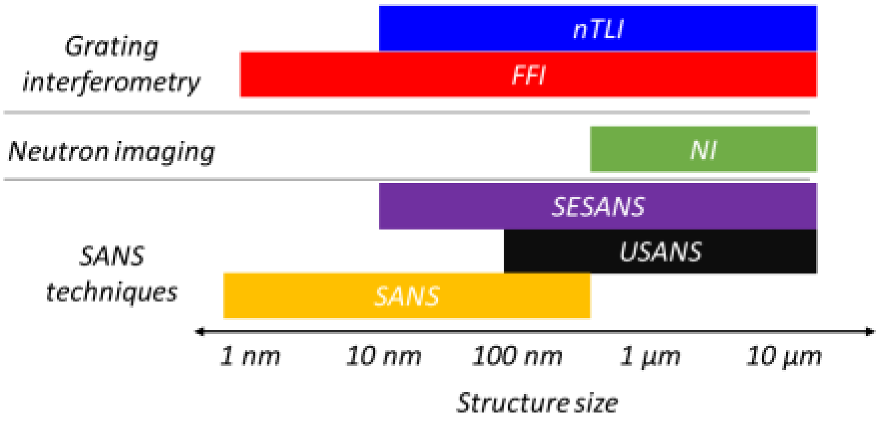
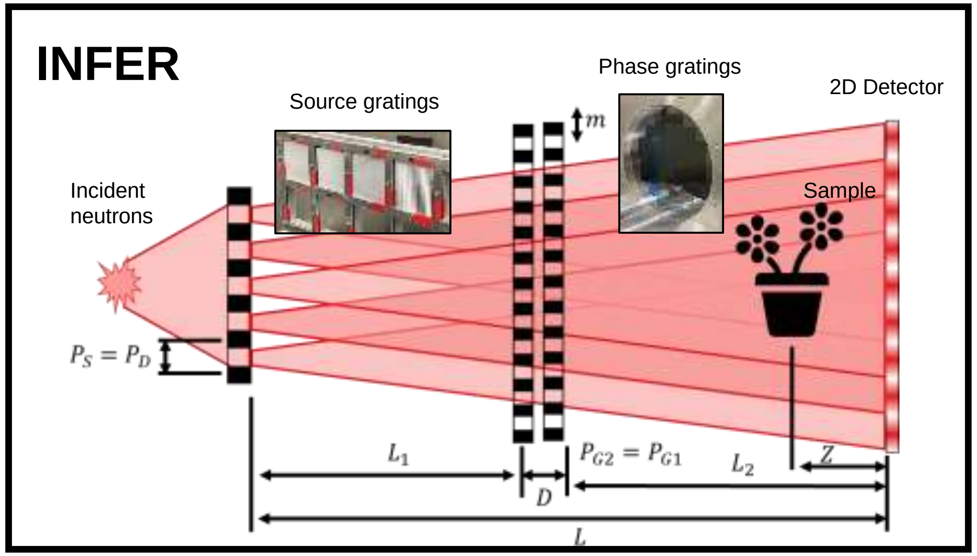


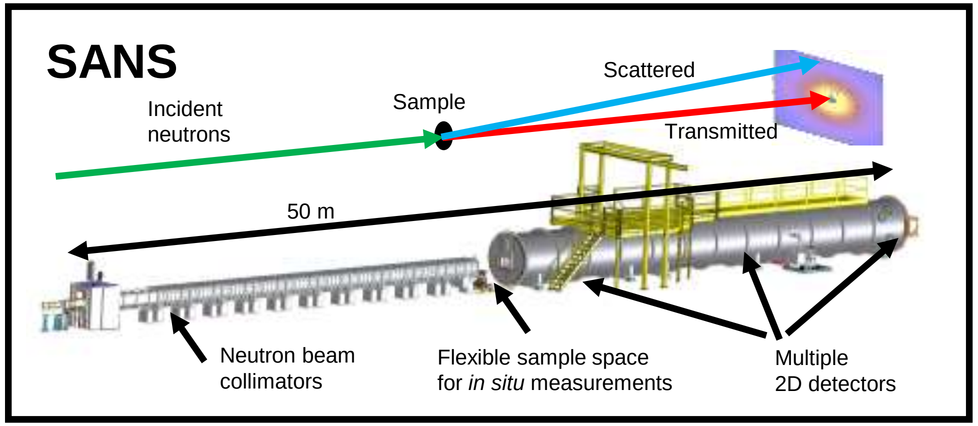
Image of fringe period varying D (@FFI)



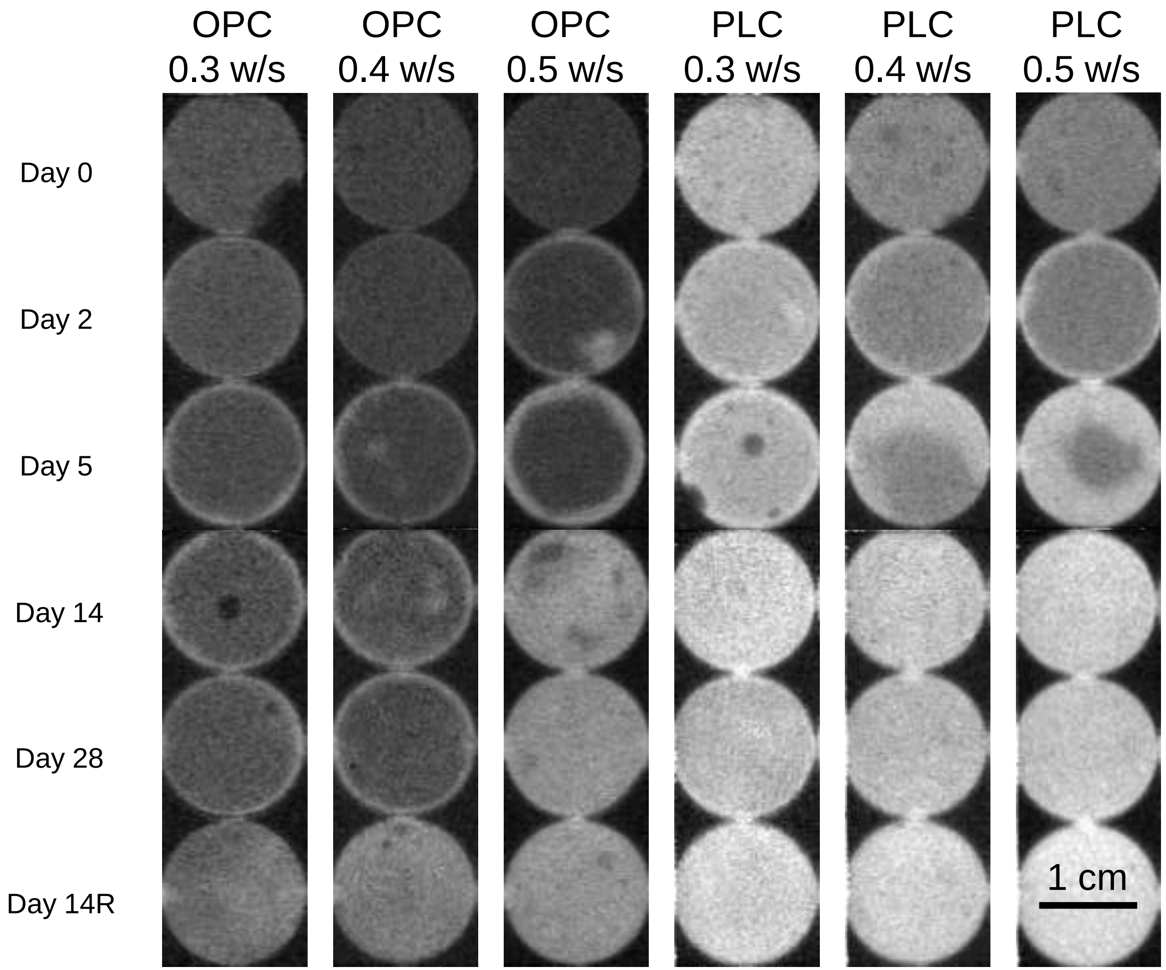
Carbonation Fronts in Cement

Carbonated cement





- Ordinary portland cements (OPC) used in concretes contain $Ca(OH)_2$ that can react with CO_2 to form solid $CaCO_3$ (carbonation)
- Carbonation kinetics and transport depend on the cement formulation, structure, porosity, pore connectivity, and environment conditions
- Measuring carbonation-induced changes in the hierarchical structure of cement requires methods that can probe length scales from nanometers up to centimeters
- Neutron scattering and imaging methods provide good scattering contrast and high penetration depths (1 mm to 1 cm)
- Small angle neutron scattering (SANS) provides ensemble averaged structure from 1 nm to 100 nm (up to 10 µm with USANS)
- Neutron interferometric imaging (INFER) is an ongoing NIST IMS project that aims to measure heterogeneous and hierarchical structures that range from 1 nm up to 5 cm



Neutron dark field image (83 nm)

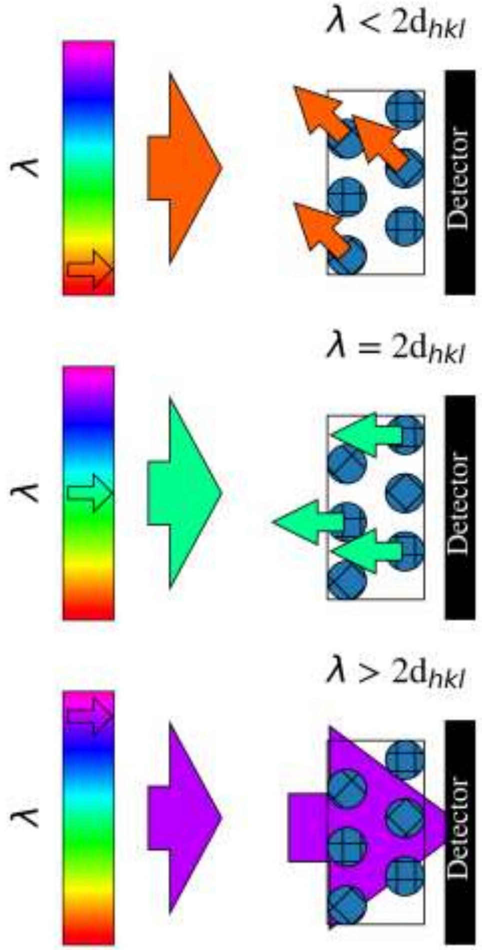
- Observed nm-to-cm heterogeneities from the carbonation (brighter regions) of various cements. These heterogeneities would be averaged or missed with SANS using volume-averaging near the sample center region
- Analyzing and modeling large data set of structures from INFER measurements, comparing with select SANS measurements, and aiming to extract important reactive transport properties of different cement formulations
- Comparing structural measurements with other carbonation quantification (TGA, mass loss after ignition)
- Planning sample preparation for INFER tomography at the NCNR, carbonation of mortar/concrete with aggregate, and carbonation with steel reinforcement bars (pending reactor status)
- Publication in preparation for 2023

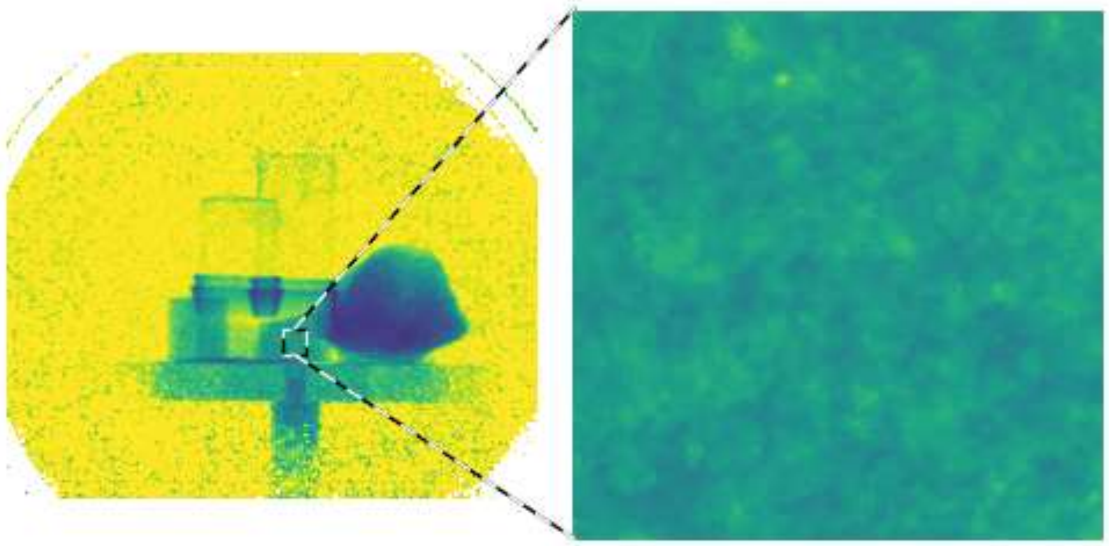
INFER Collaborators and Alum

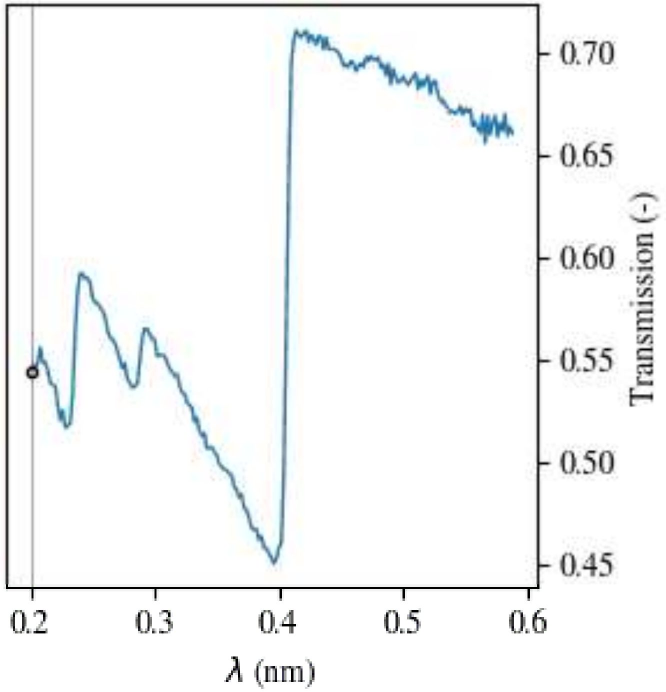
Active Collaborator	Affiliation
Hunter	LSU, Physics (Graduate student)
Joyoni Dey	LSU, Physics
Peter Bajcsy	NIST, ITL
Pushkar Sathe	NIST, ITL
Katie M. Weigandt	NIST, NCNR
Paul A. Kienzle	NIST, NCNR
Ryan P. Murphy	NIST, NCNR
Caitlyn Wolf	NIST, NCNR (NRC Post Doc Fellow)
Hubert King	NIST, NCNR
Daniel S. Hussey	NIST, PML
David L. Jacobson	NIST, PML
Jacob M. LaManna	NIST, PML
M. Cyrus Daugherty	NIST, PML (UMD CREB Postdoc)
Michael G. Huber	NIST, PML
Nikolai N. Klimov	NIST, PML
Sarah Robinson	NIST, PML
Youngju Kim	UMD PREP Post Doc
Dimitry Pushin	University of Waterloo
Dusan Sarenac	University of Waterloo
Connor Kapahi	University of Waterloo (Graduate student)
Atishay Jain	Brown University
Ritambhara Singh	Brown University

Alum	NIST Affiliation / New Post if Known
Yin Huang	NIST, ITL (UMD PREP Post Doc)
Victoria H. DiStefano	NIST, PML – Now AAAS Fellow with DOE office of Science
Ben J. Heacock	NIST, PML (NRC Postdoc Fellow) Now Bechtel
Chris Haddock	NIST, PML (NRC Postdoc Fellow) Now Hedgefog Research
Ivan J Hidrovo Giler	LSU, Physics (Masters) Now Tulane Health Physicist

Bragg Edge tomography of Samples







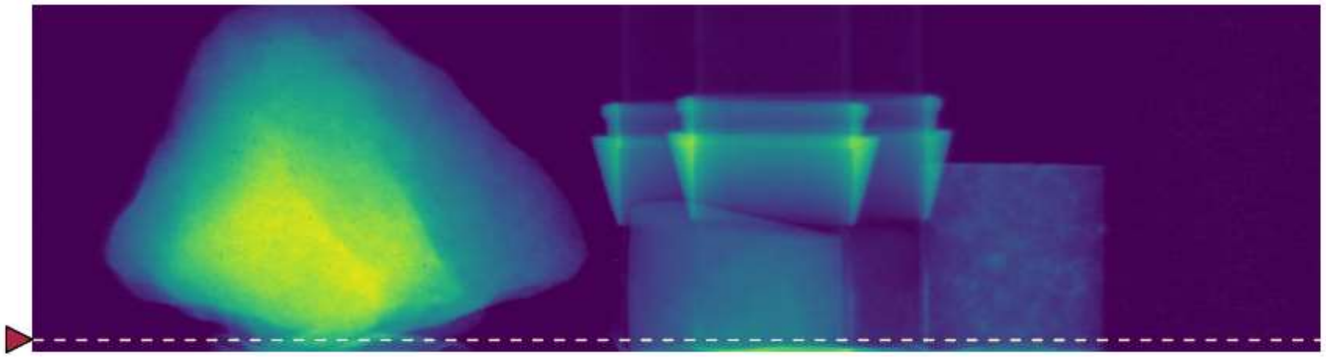
M.C. Daugherty et al, accepted SN Computer Science, SNCS-D-22-02635R2 (June, 2023)

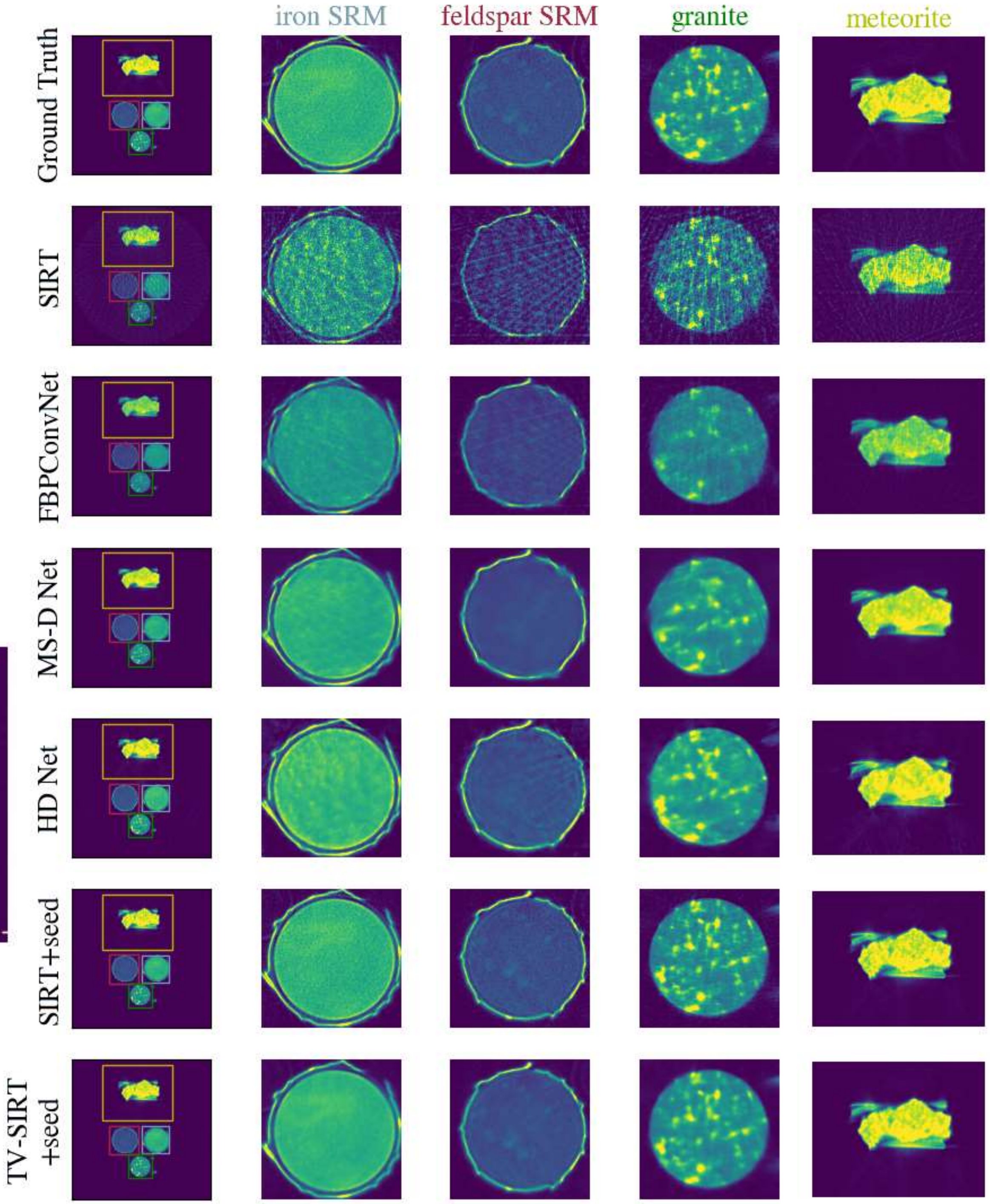
Experimental Methods

- Samples:
  - Meteorite
  - Granite Core
  - SRM: iron powder
  - SRM: potassium feldspar
- Polychromatic Scan
  - 2400 Projections
  - P43 scintillator (Gd_2O_2S:Tb)
  - Seeding iterative methods
  - Training neural networks
- Dose-Reduced Monochromatic
  - Double Crystal Monochromator
    - $\frac{\Delta\lambda}{\lambda} = \sim 1\%$
  - Spectrum Scan:
    - 80 Projections
    - 195 Wavelengths (0.2 nm – 0.58 nm; with 0.002 nm intervals)
  - Reference Scan: 720 Projections; λ=0.37 nm
  - Zinc sulfide/lithium fluoride scintillator

M.C. Daugherty et al, accepted SN Computer Science, SNCS-D-22-02635R2 (June, 2023)

Polychromatic Results





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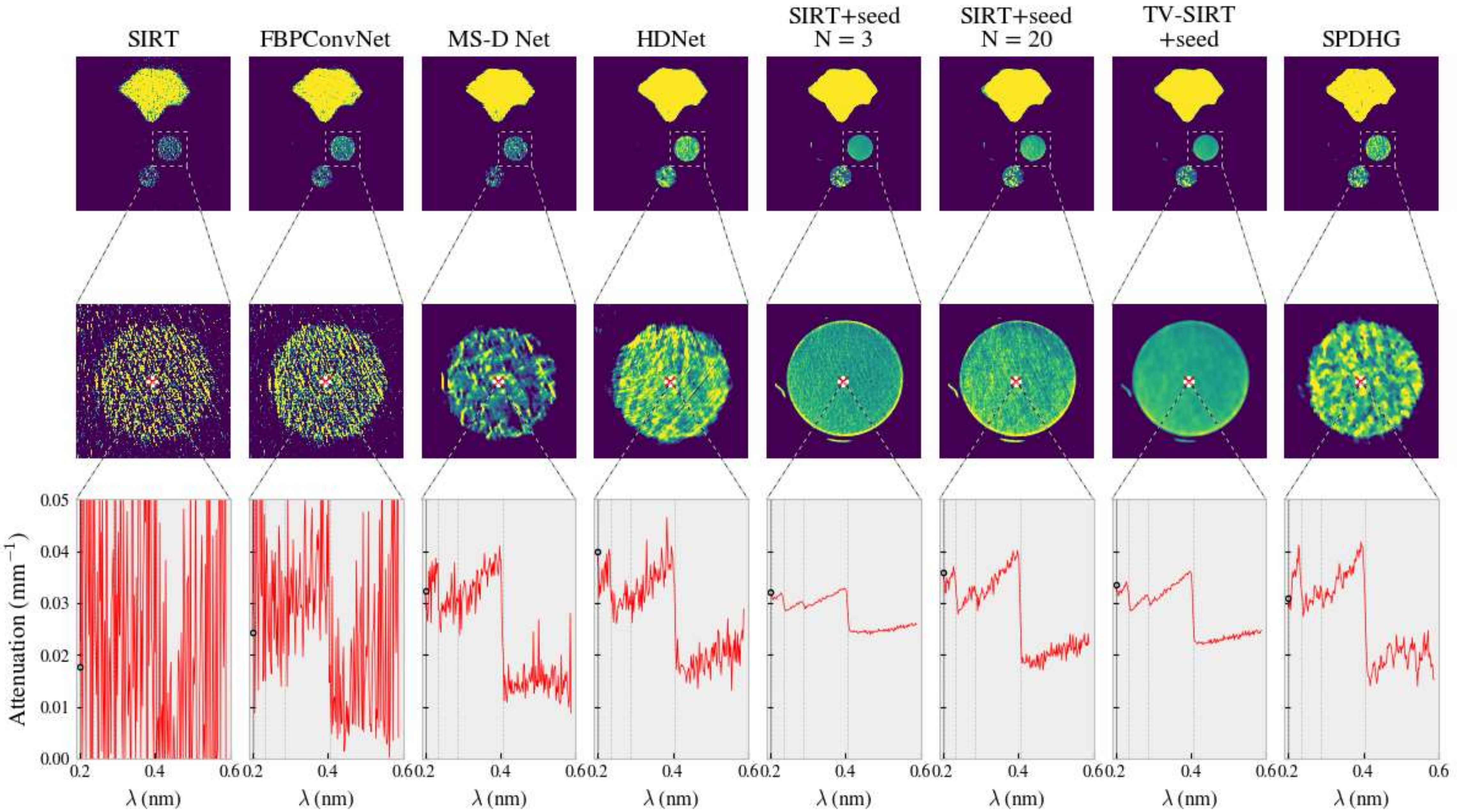
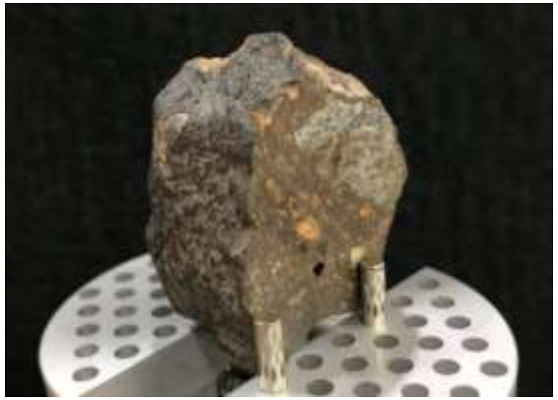
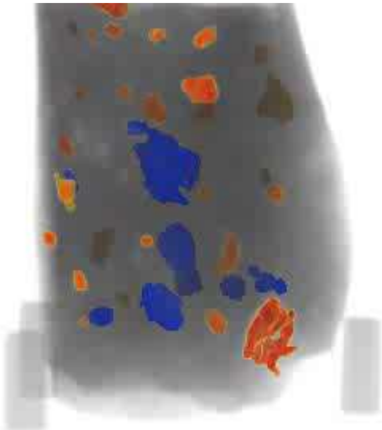
Monochromatic Results: Single Voxel



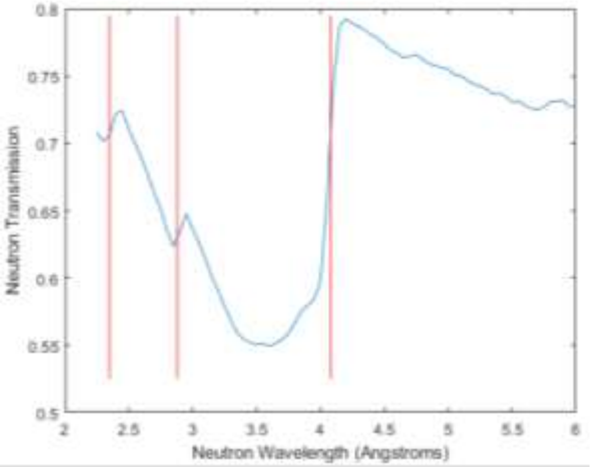
M.C. Daugherty et al, accepted SN Computer Science, SNCS-D-22-02635R2 (June, 2023)

Mineralogy of Meteors via energy selective tomography??

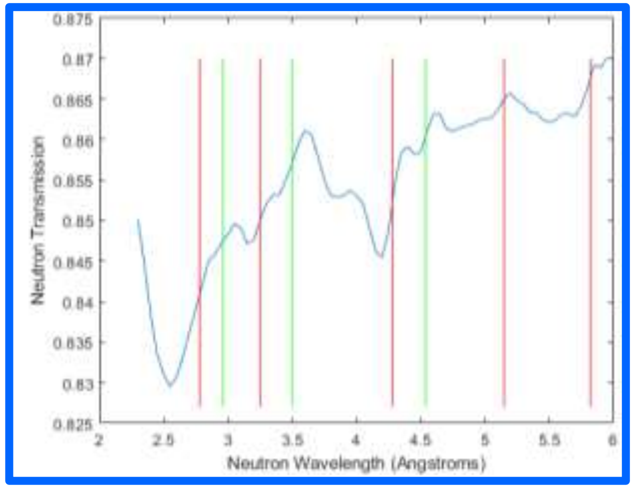




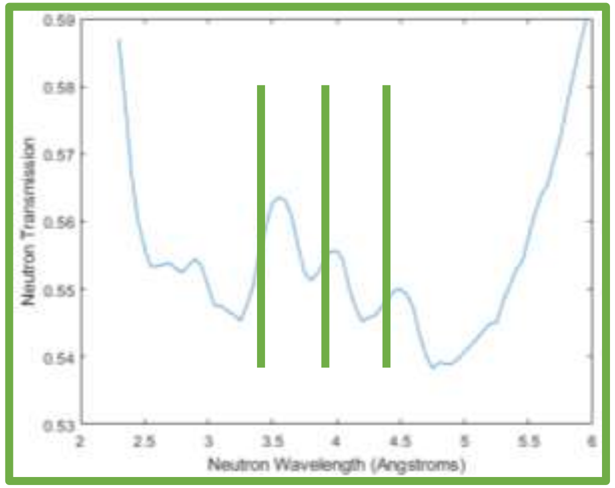
Can non-destructively identify ~1 mm grains



Martensite
Posts

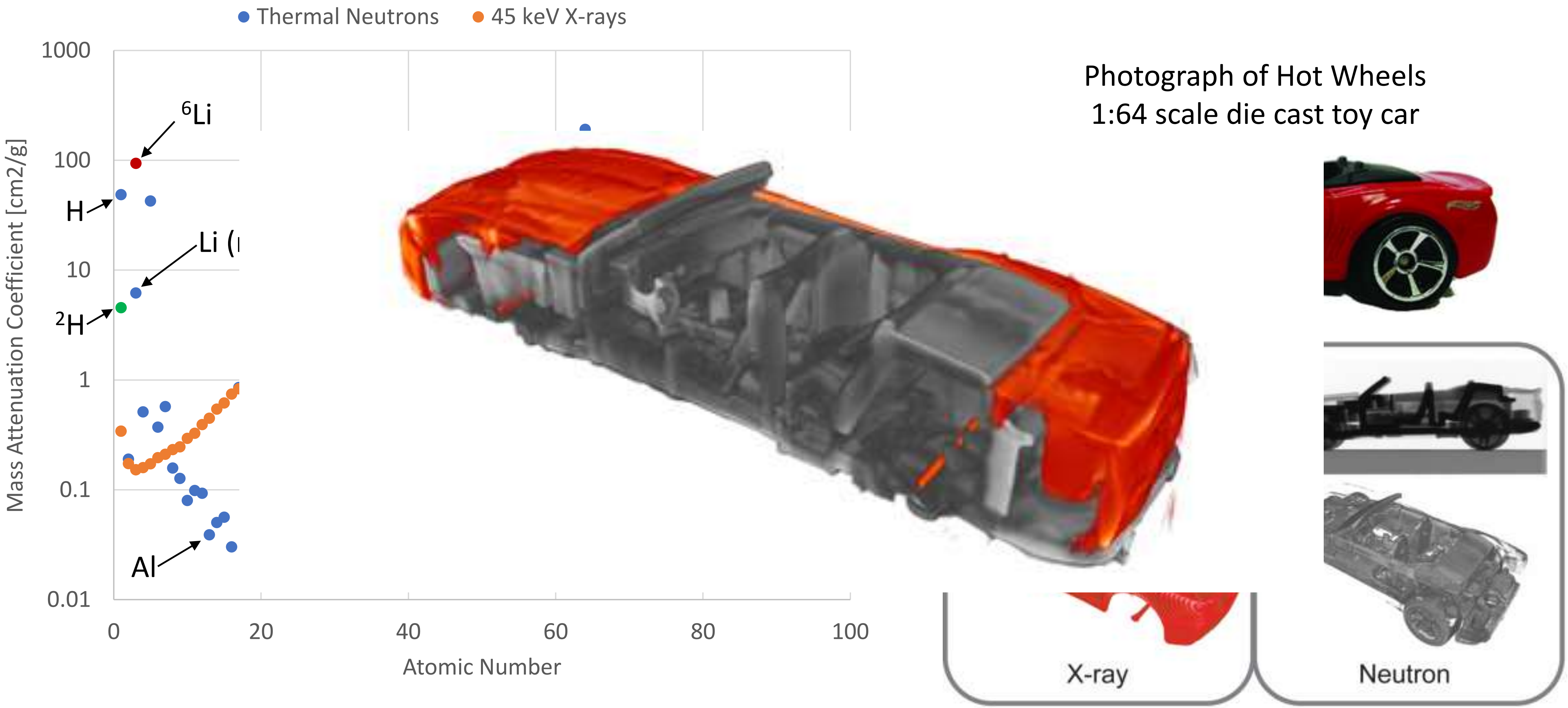


Pigeonite
Olivine



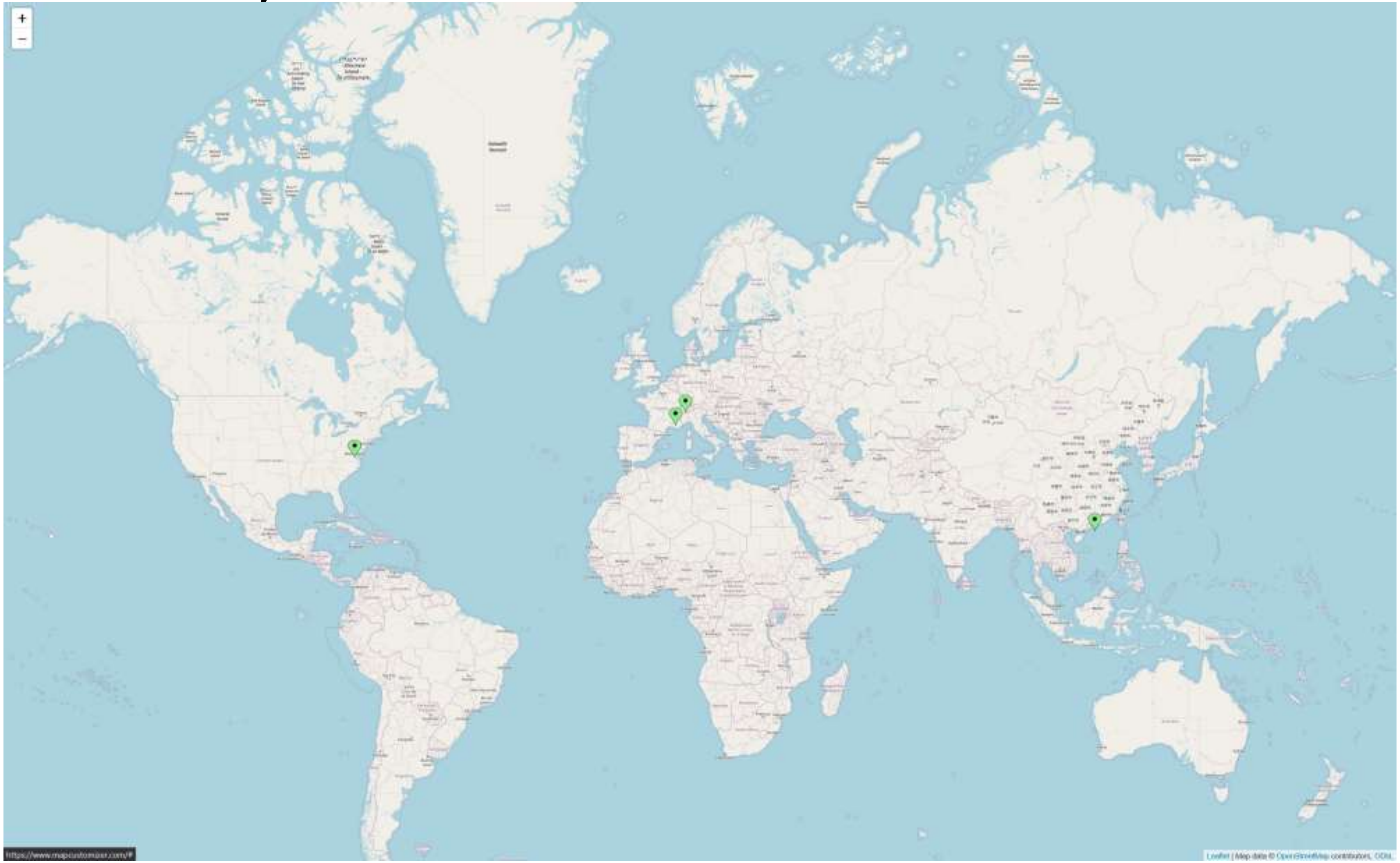
Not Identified

Why combine neutrons and X-rays? Awesome complementarity!

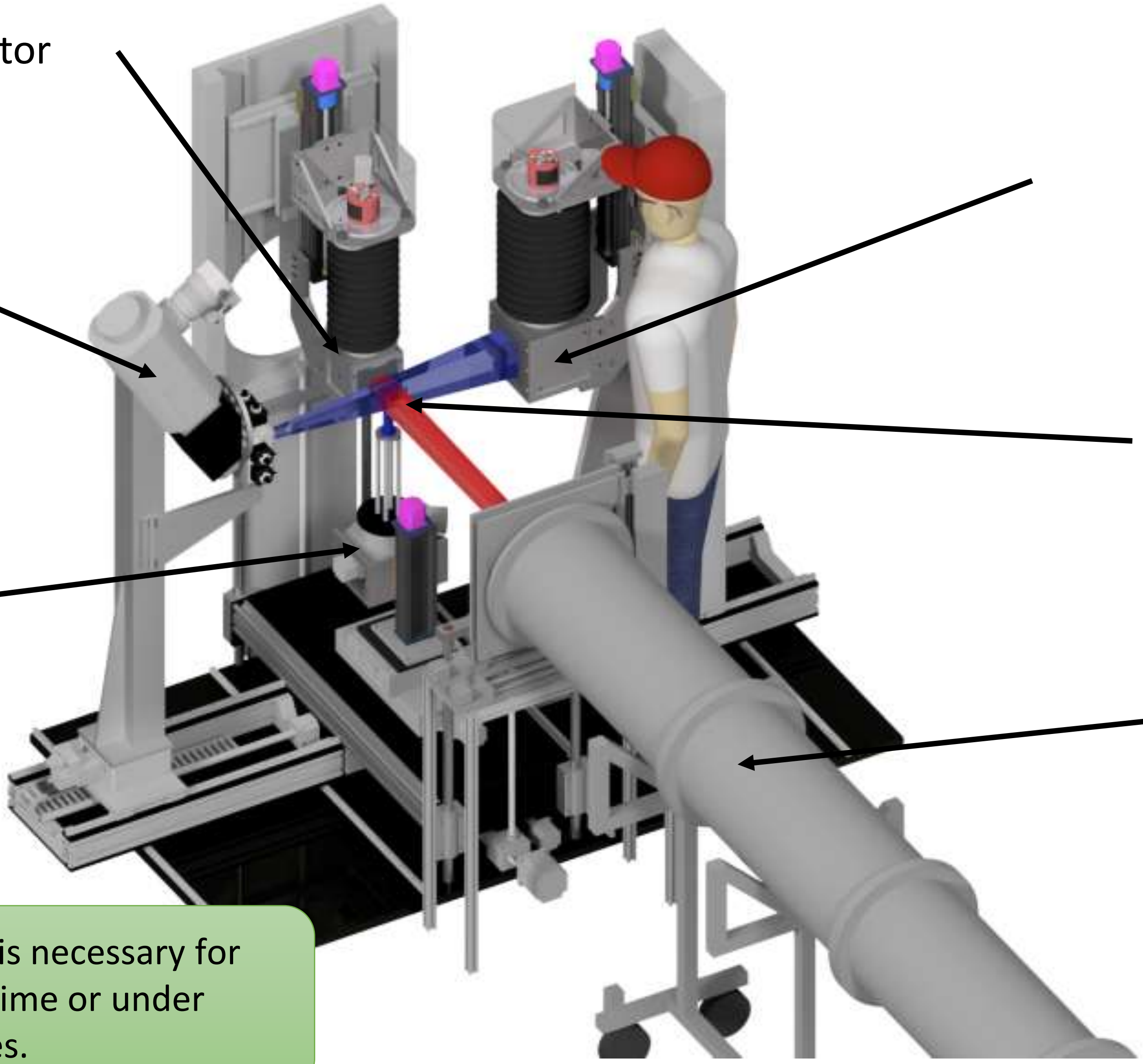


Photograph of Hot Wheels
1:64 scale die cast toy car

Where are NeXT systems available



The NIST **Simultaneous** Neutron and X-ray Tomography System



Neutron detector

225 keV X-ray
tube

detector

Sample position

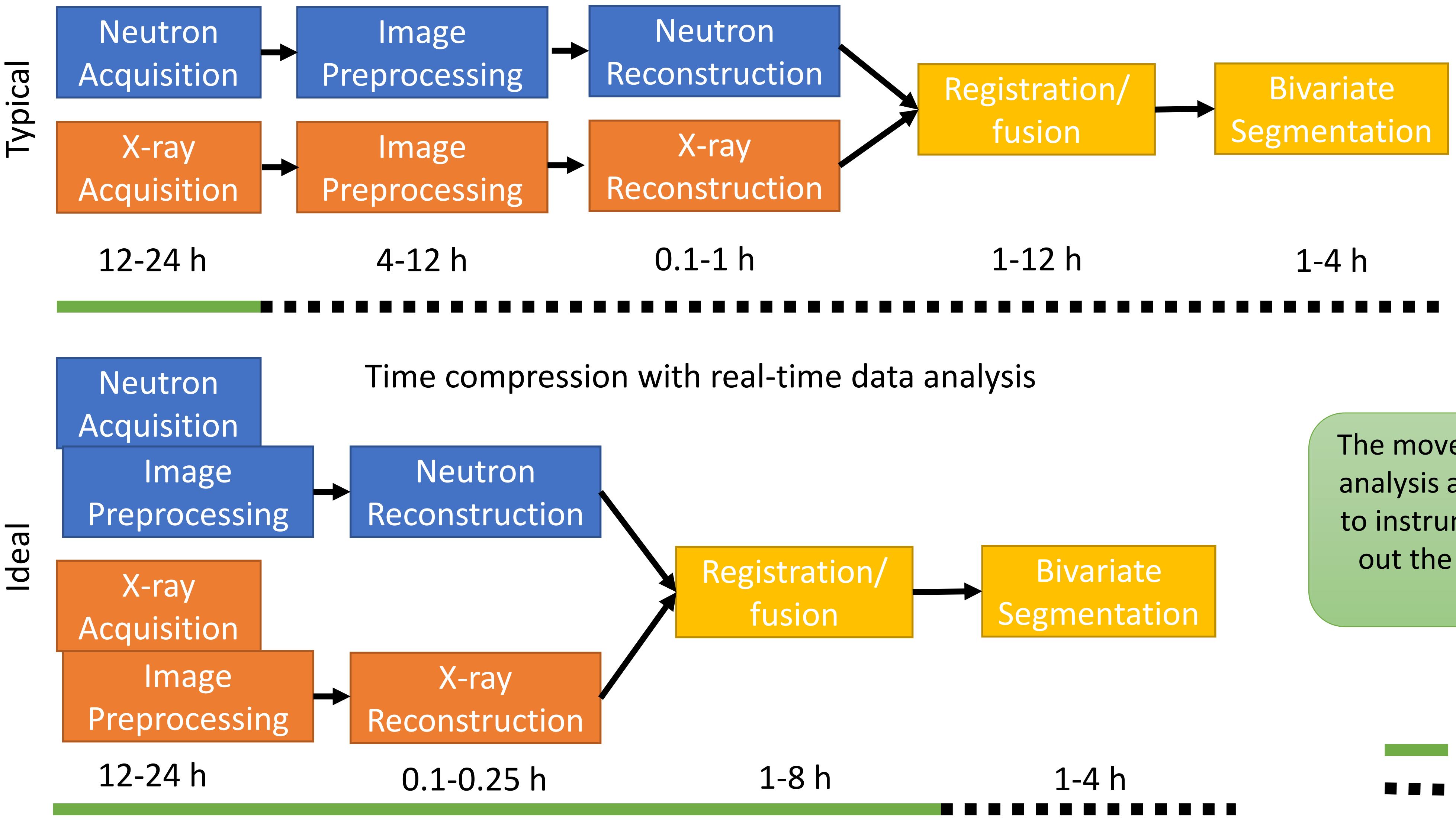
Sample
manipulation stages

Neutron flightpath

Truly simultaneous tomography is necessary for samples slowly evolving with time or under stochastic processes.

J.M. LaManna et al., “Neutron and X-ray Tomography (NeXT) System for Simultaneous, Dual Modality Tomography”, Rev. Sci. Inst., 88, 113702 (2017).

Tomography Data Acquisition and Analysis Workflow and Timeline



Typical

Neutron Acquisition → Image Preprocessing → Neutron Reconstruction

X-ray Acquisition → Image Preprocessing → X-ray Reconstruction

→ Registration/ fusion → Bivariate Segmentation

12-24 h | 4-12 h | 0.1-1 h | 1-12 h | 1-4 h

Ideal

Time compression with real-time data analysis

Neutron Acquisition / Image Preprocessing → Neutron Reconstruction

X-ray Acquisition / Image Preprocessing → X-ray Reconstruction

→ Registration/ fusion → Bivariate Segmentation

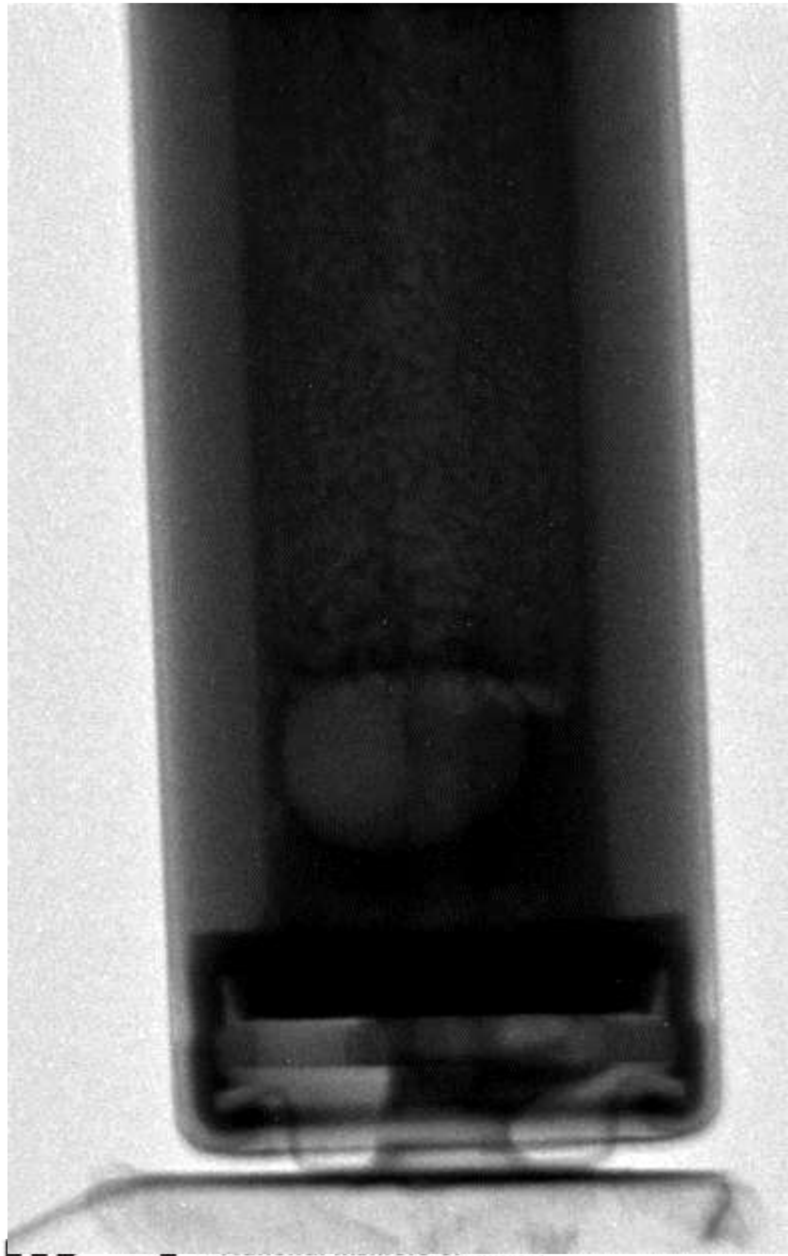
12-24 h | 0.1-0.25 h | 1-8 h | 1-4 h

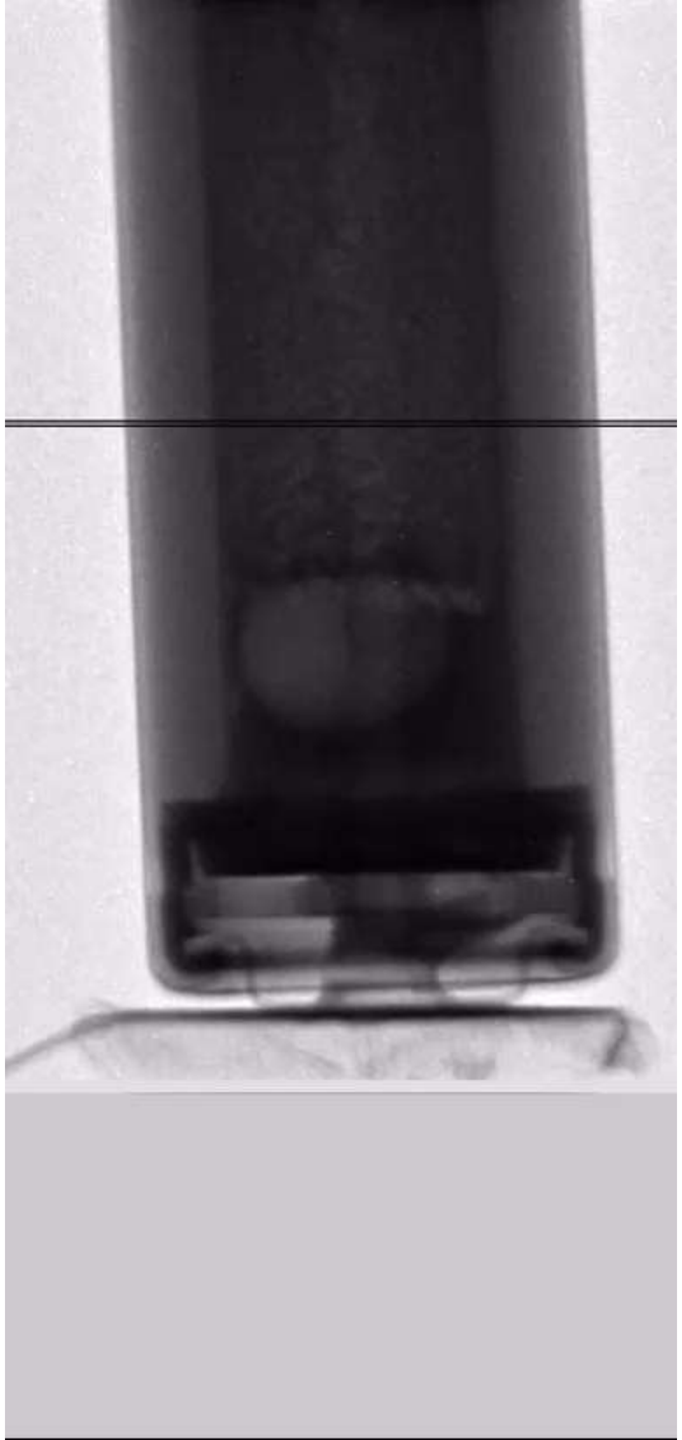
The move to real-time streaming data analysis and upcoming improvements to instrument will allow users to walk out the door with ready to analyze data!

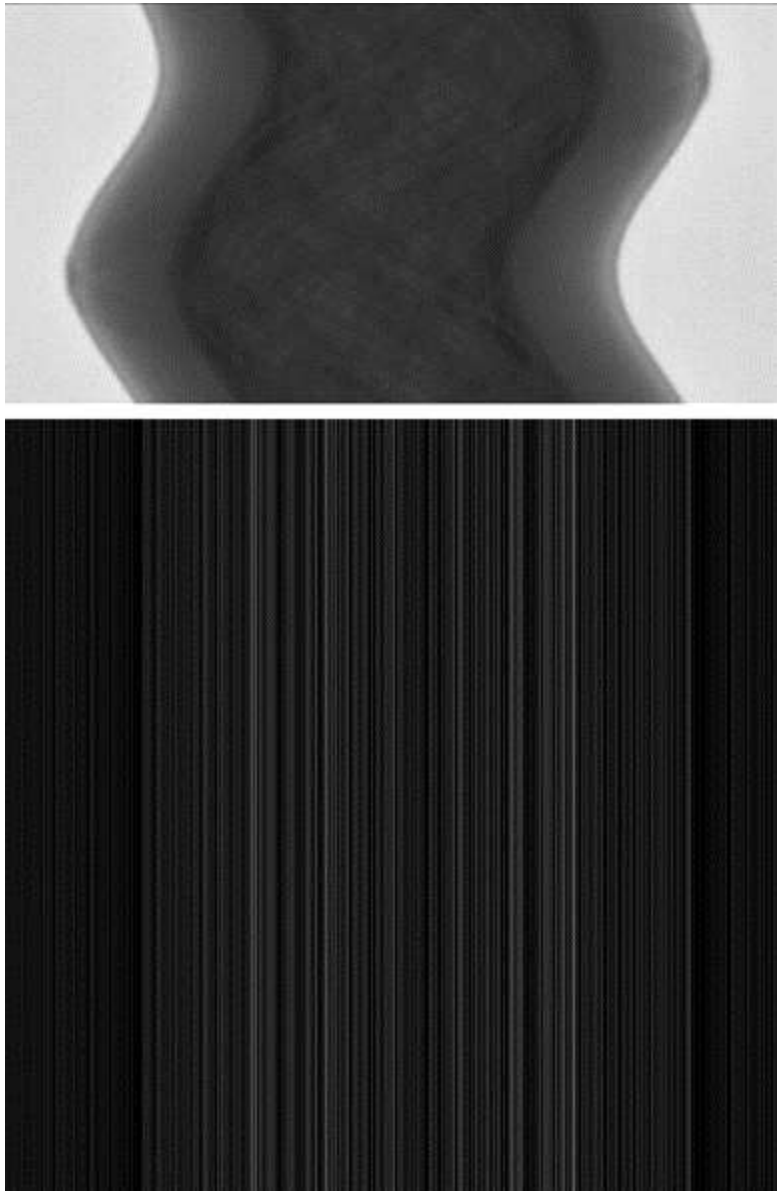
━ Users present at NIST

▪▪▪ Users return to home

Tomography intro





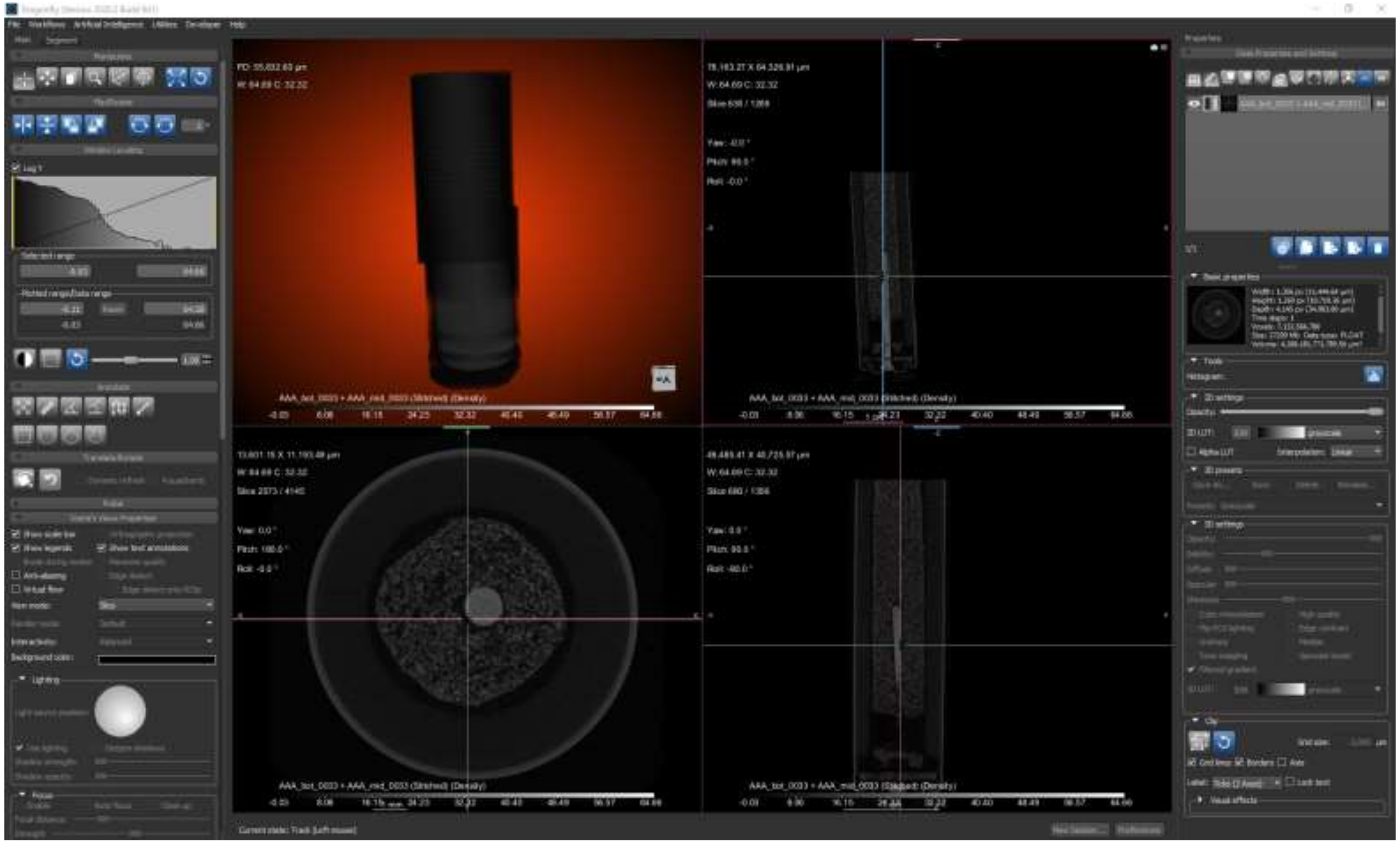


Volume stitching for long samples in Dragonfly

- Our Matlab registration program does no currently support stitching
- Dragonfly works well for this task
- Biggest challenge is the RAM requirements to load in multiple volumes
    - ~20-30 GB per volume
    - Need space for original volumes and new stitched volume

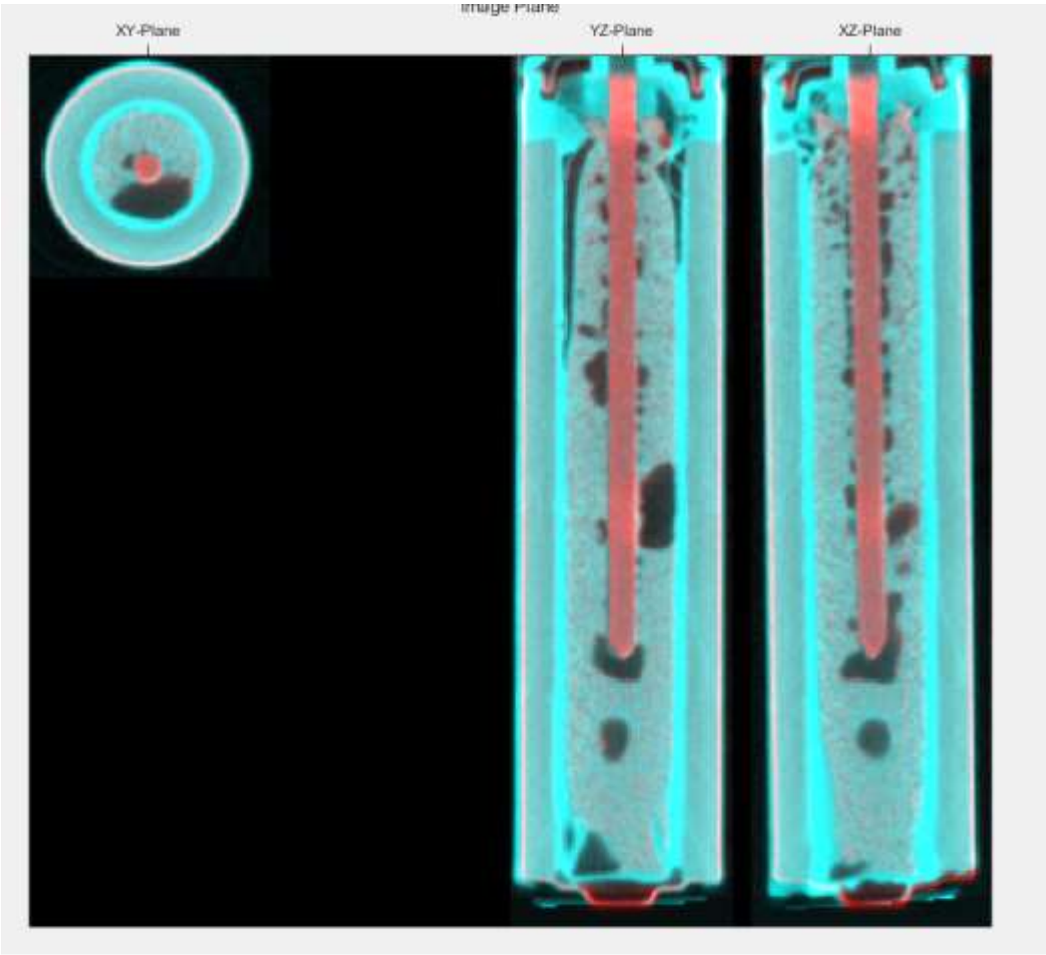
- Biggest volume I've currently produced: 3 scans resulting in a 1400x1400x5000 volume

(will get worse with larger format cameras)



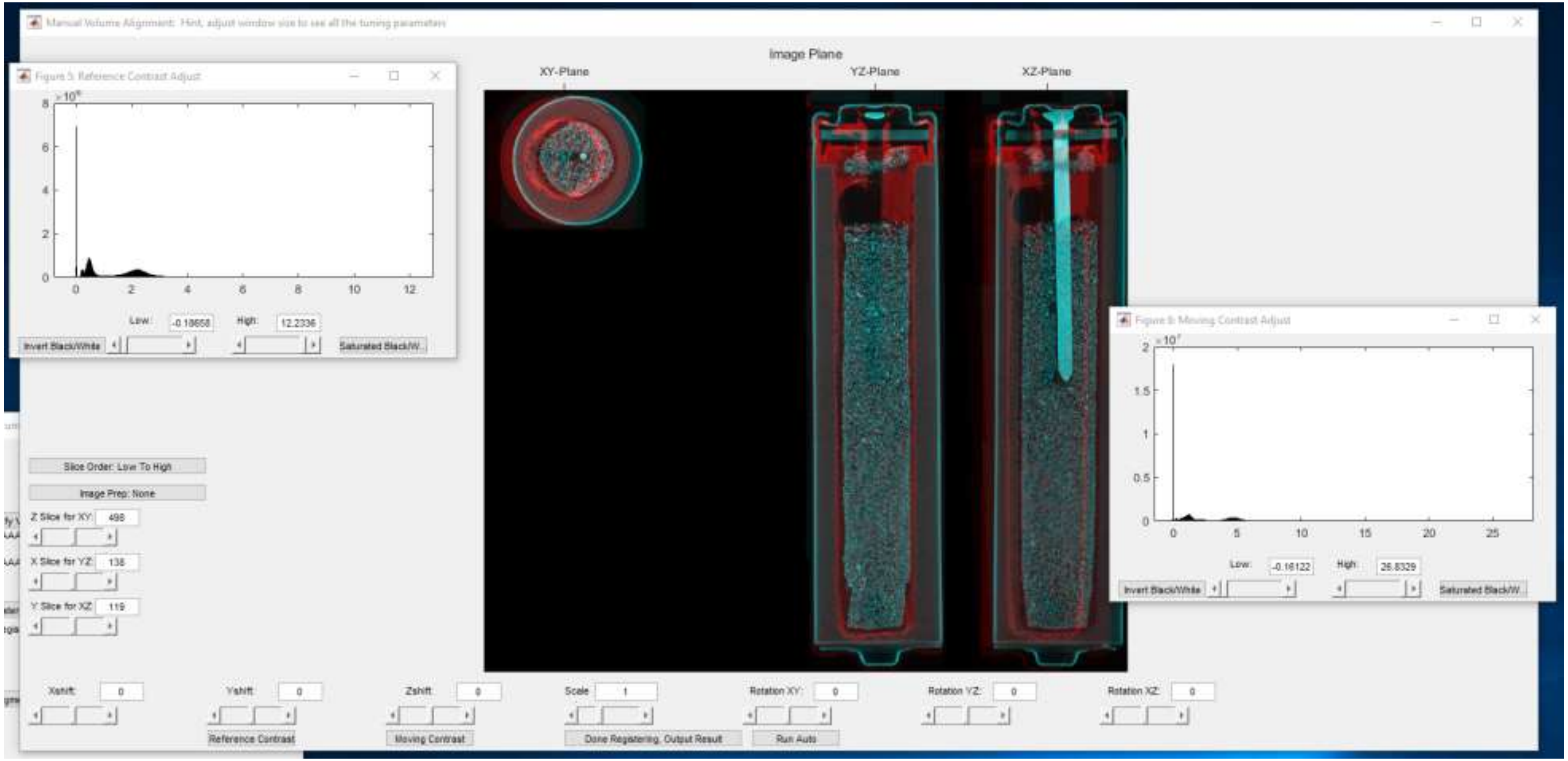
Start fusing through registration

- Binning
- Moving volume manipulation
  - Rotation, flip, etc.
- Applying previous transformation to a new dataset
- Single- and Multi-modal registration
  - Mean squares
  - Mattes Mutual Information
- Contrast adjustment
- Manual positioning
- Output volume cropping



J. M. LaManna et al, Proc. SPIE 11494 (2020); doi: 10.1117/12.2569666

Overview of the program



Low: -0.18658
High: 12.2336

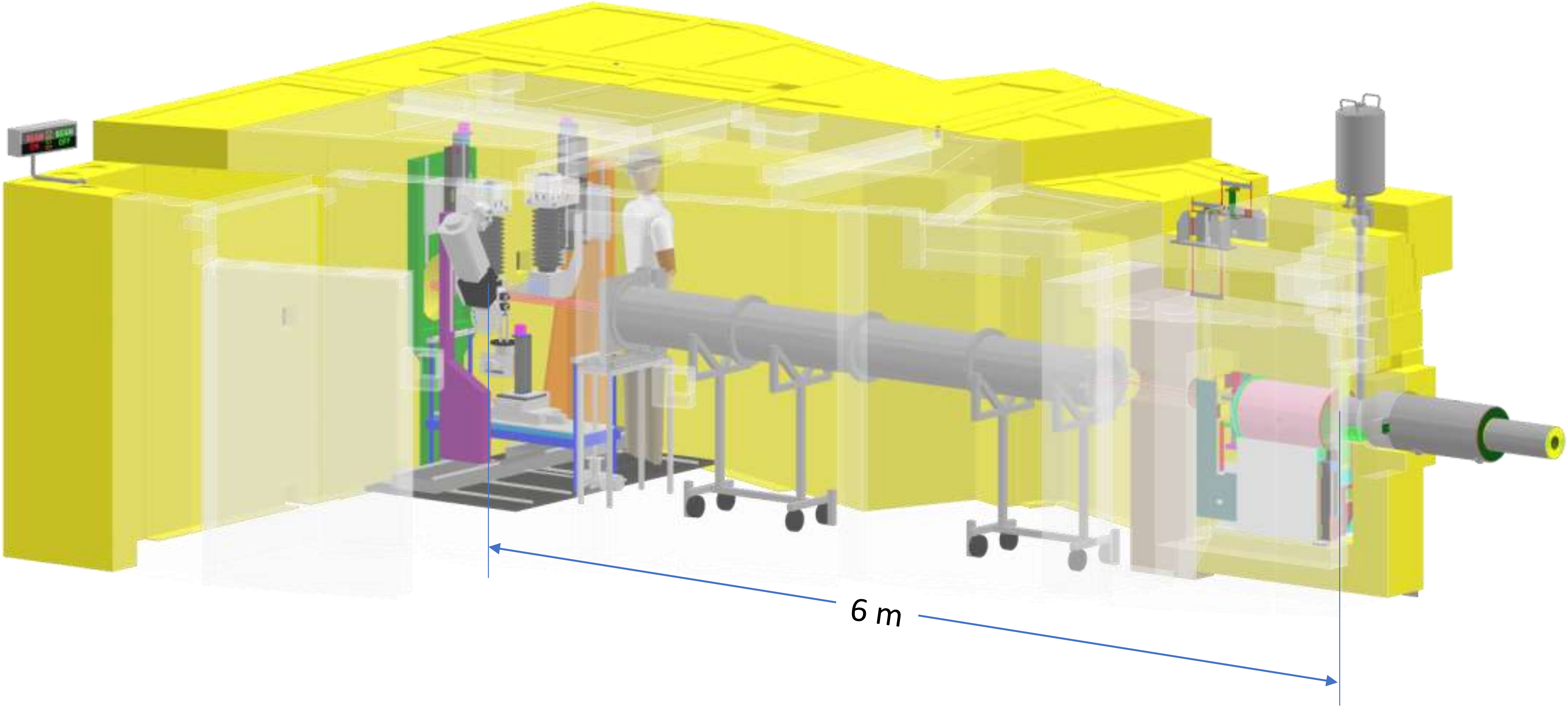
Low: -0.16122
High: 26.8329

Z Slice for XY: 498
X Slice for YZ: 138
Y Slice for XZ: 119

Xshift: 0
Yshift: 0
Zshift: 0
Scale: 1
Rotation XY: 0
Rotation YZ: 0
Rotation XZ: 0

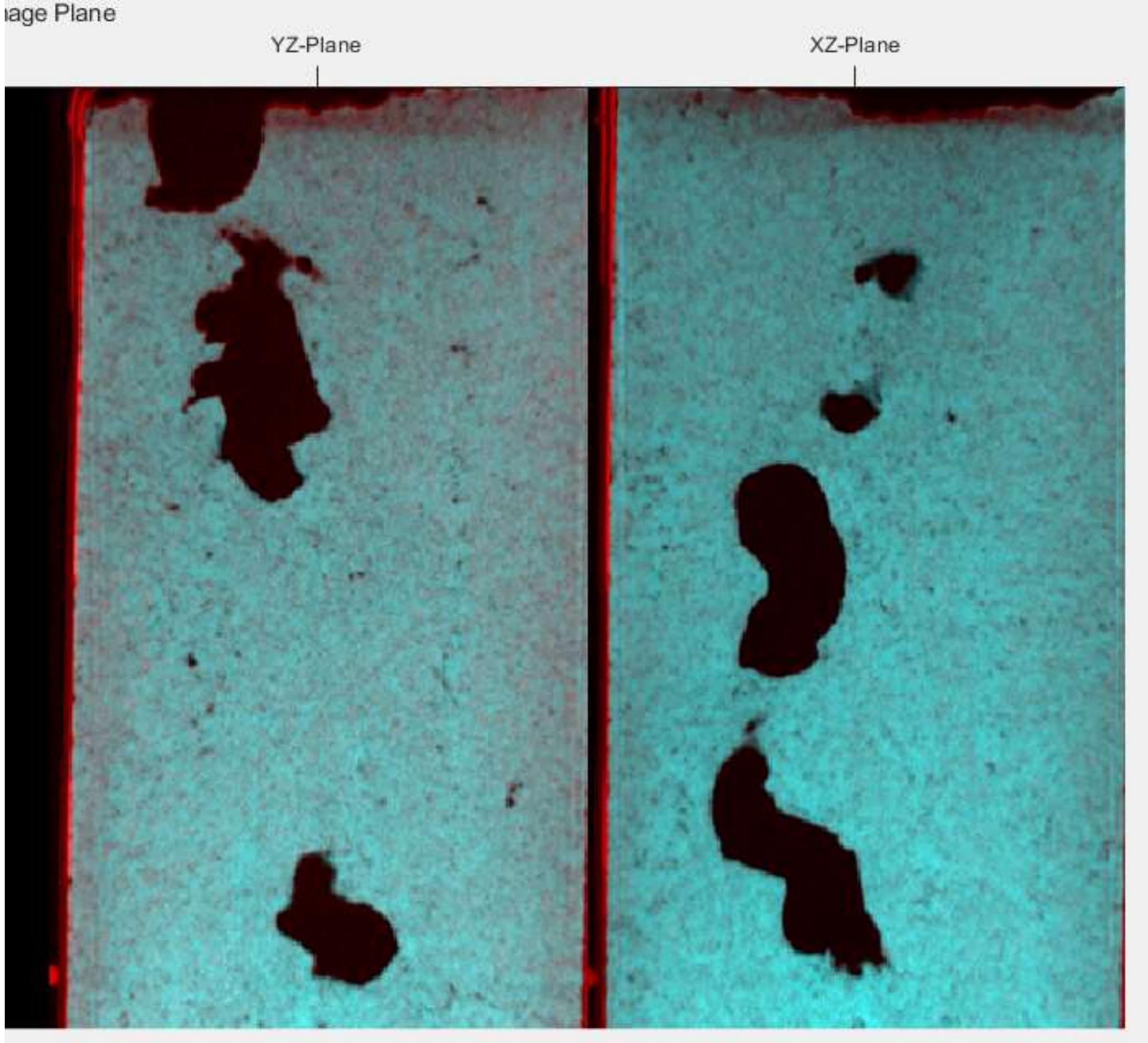
J. M. LaManna et al, Proc. SPIE 11494 (2020); doi: 10.1117/12.2569666

Instrument configuration: Parallel vs Cone

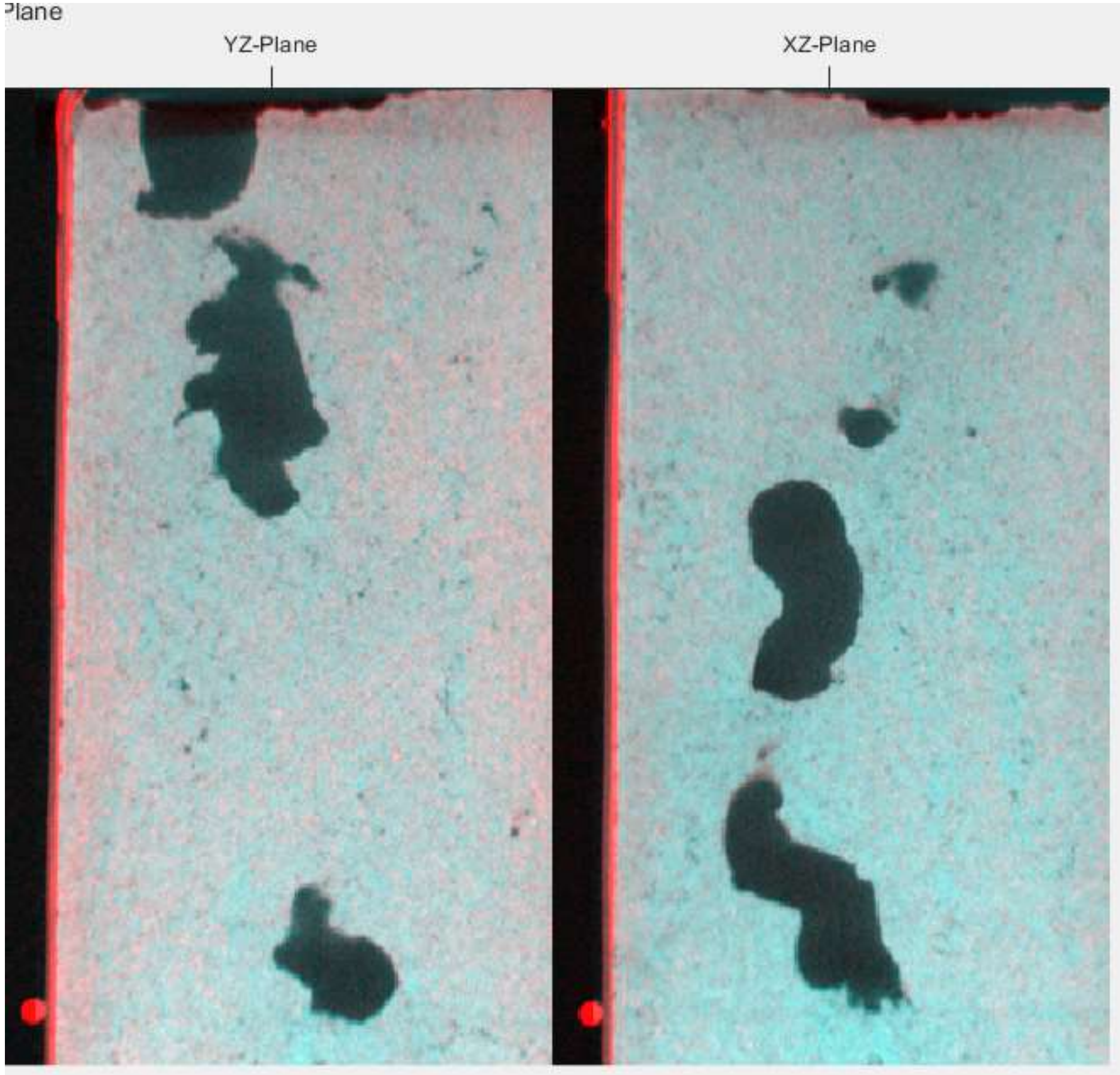


J. M. LaManna et al, Proc. SPIE 11494 (2020); doi: 10.1117/12.2569666

Neutron Parallel beam



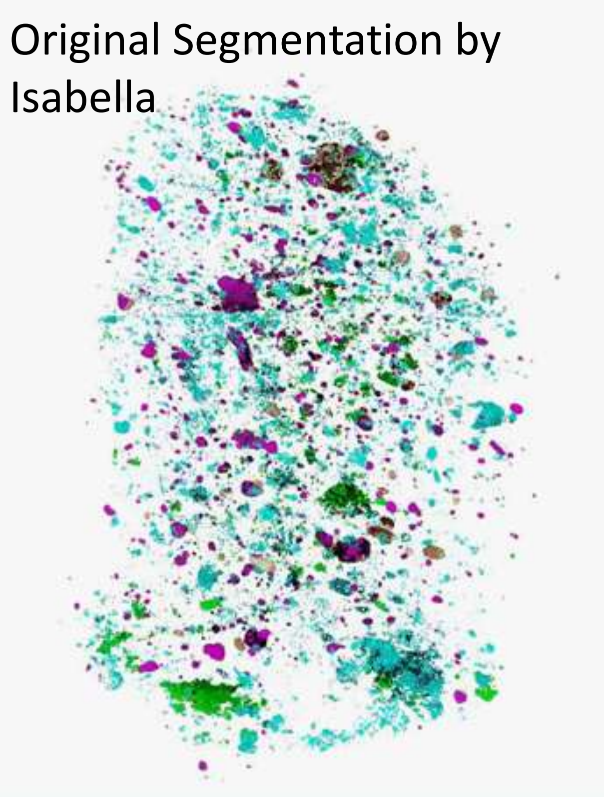
Neutron Cone Beam



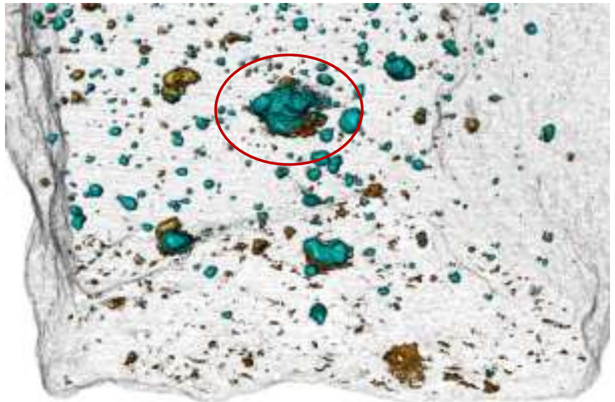
J. M. LaManna et al, Proc. SPIE 11494 (2020); doi: 10.1117/12.2569666

Improvements to Meteorite segmentation

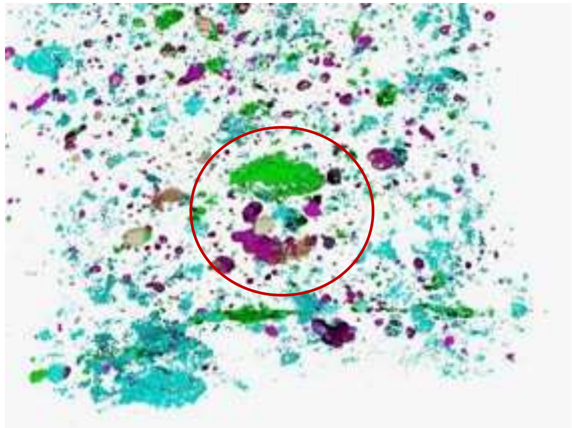
Original Segmentation by Isabella

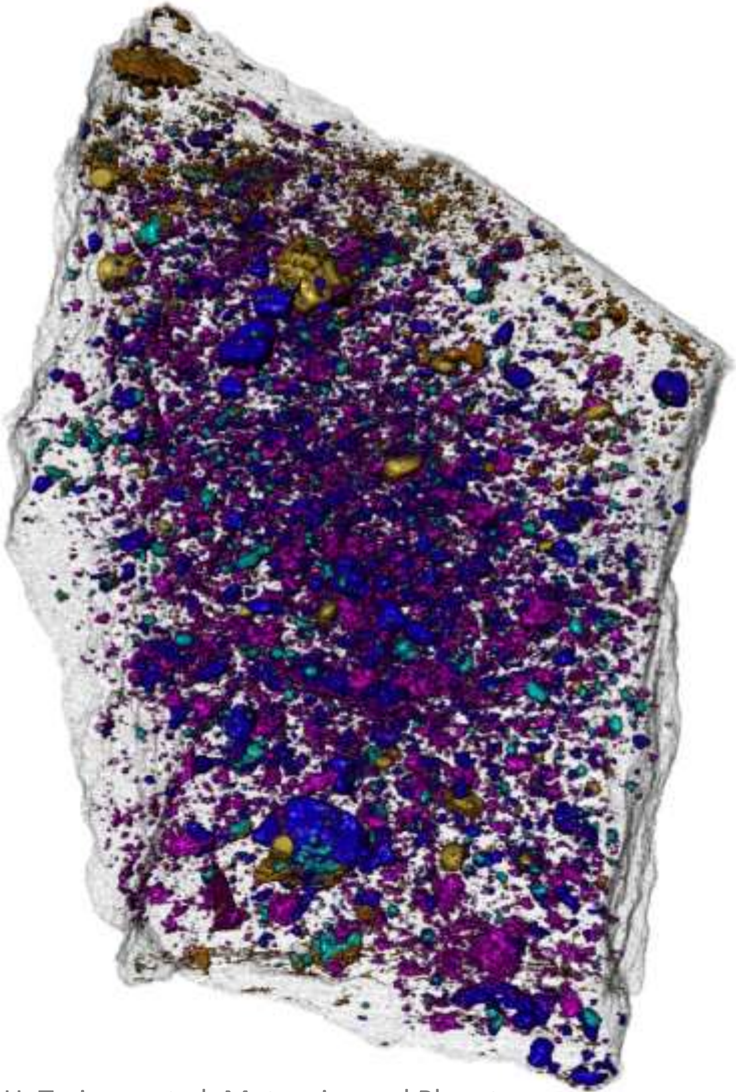


- H-Bearing Minerals
- Diogenite Pyroxene
- Impact Melt Clasts
- Eucrite Pyroxene and Plagioclase



Improvements to the understanding of reconstruction parameters and volume registration has greatly improved the segmentation quality. Isabella did the best she could with the poor original data I provided her.





A.H. Treiman et al, Meteorics and Planetary Science, 2022, doi: 10.1111/maps.13904

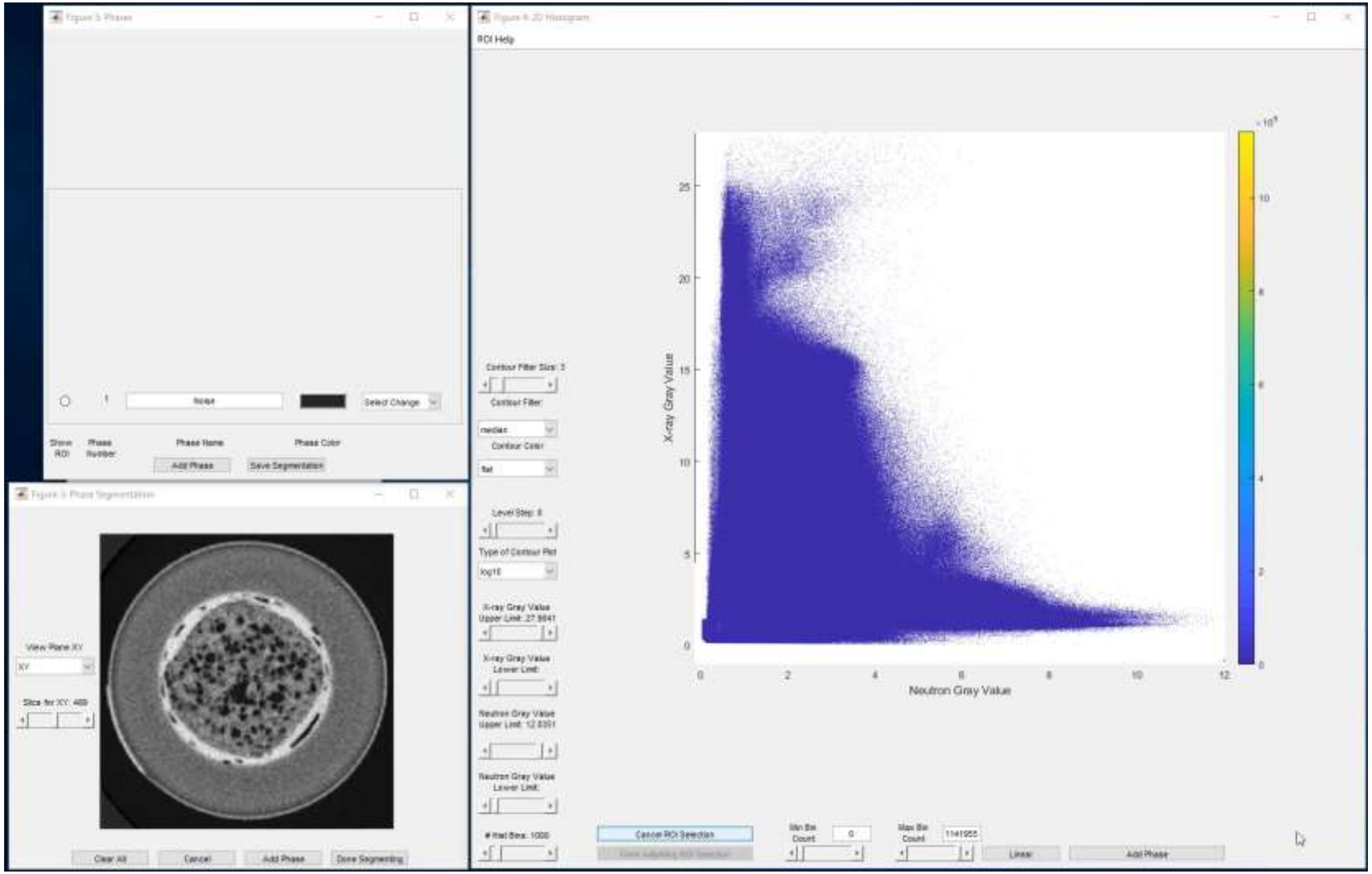
- Hydrogenous Materials
- Moderately Hydrogenous Materials
- Metallic Materials
- Moderately Metallic Materials
- Neutron Dark Spots (hidden in other phases)
- Low N and X attenuating Materials

The Bivariate Histogram Segmentation Program

List of selected regions

Output: 1 binary volume for each ROI

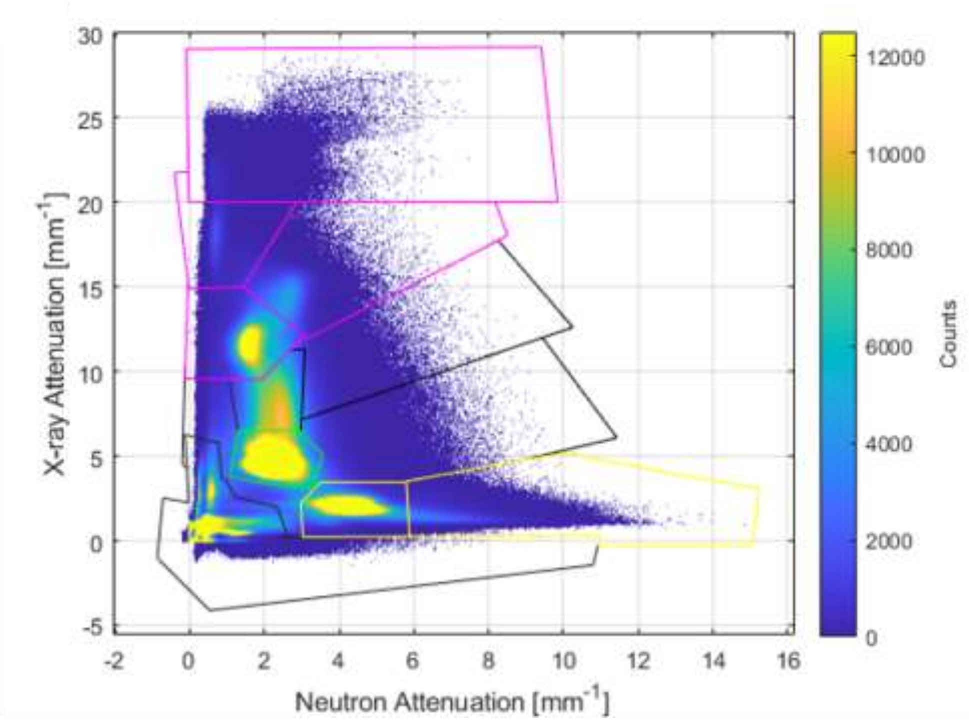
Selectable view direction showing progress of segmentation



Bivariate histogram

J. M. LaManna et al, Proc. SPIE 11494 (2020); doi: 10.1117/12.2569666

Segmentation based on bi-variate histogram of attenuation values

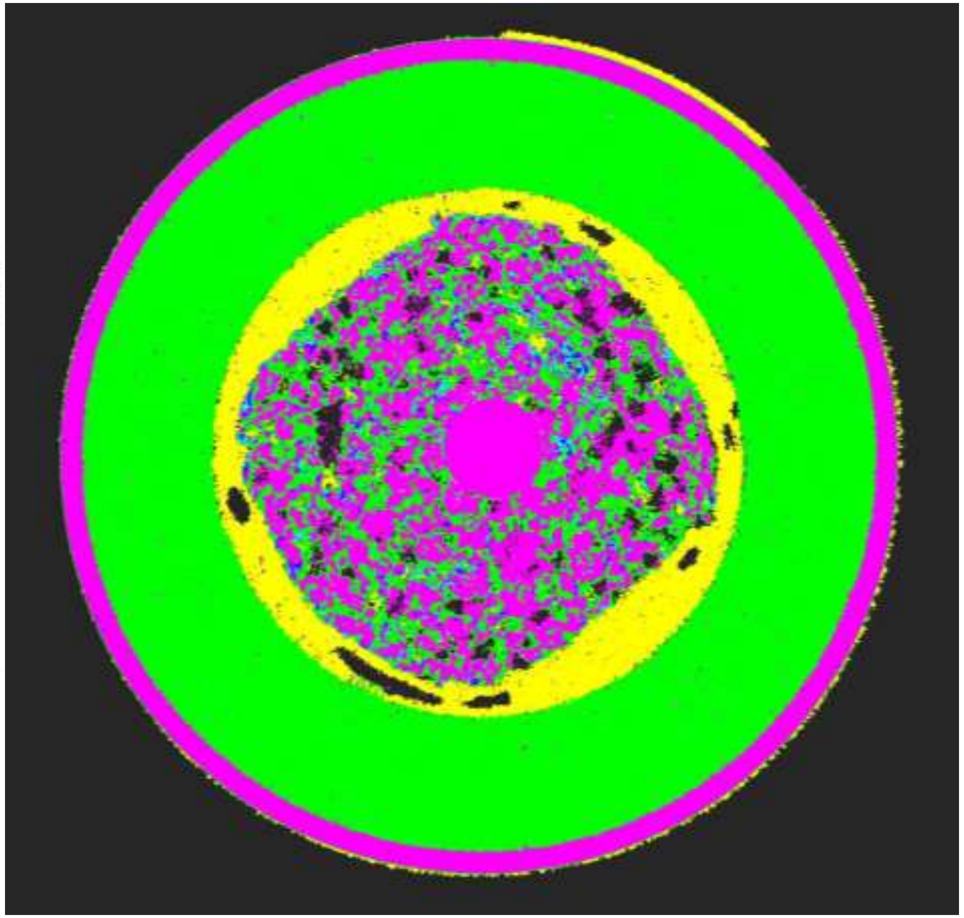


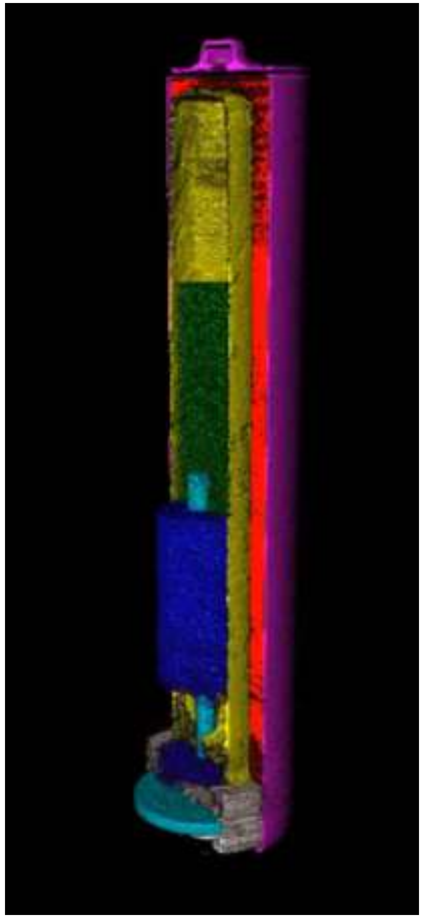
Cathode and electrolyte 🟩

Polymer 🟨

Can and zinc anode 🟪

Void ⬛





Volume is 1400 x 1400 x 5000

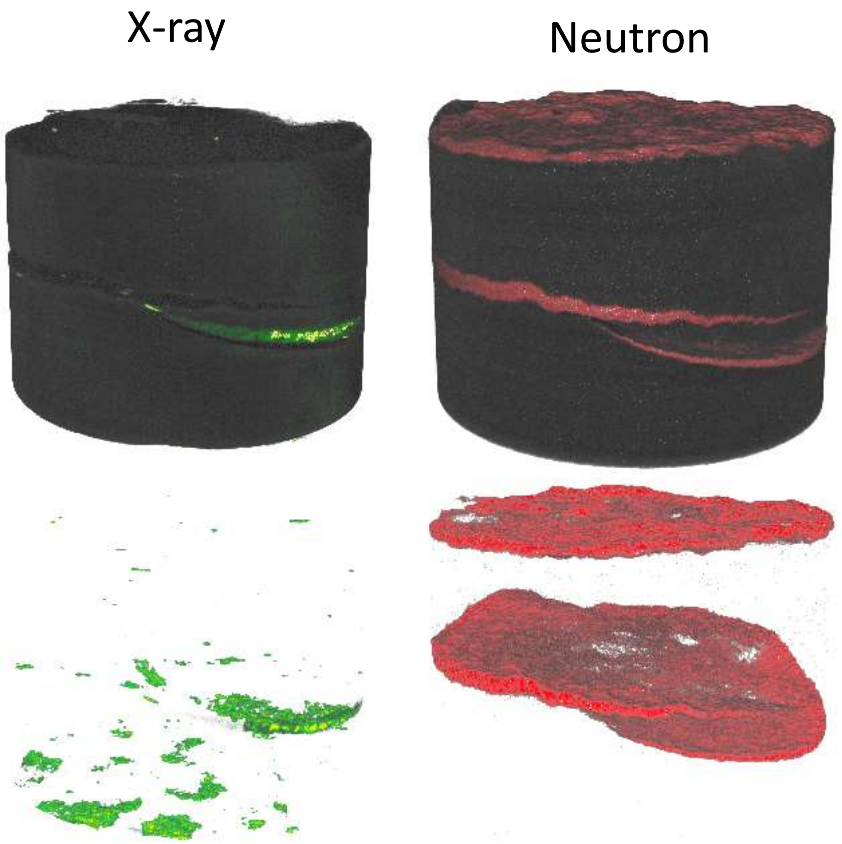
National Institute of Standards and Technology
U.S. Department of Commerce

J. M. LaManna et al, Proc. SPIE 11494 (2020); doi: 10.1117/12.2569666

Improvements by using iterative registration and phase segmentation

Segmented ϕ25mm shale presented in 2017

Independent 1D histogram segmentation



15 µm pixel, 18 hr scan time

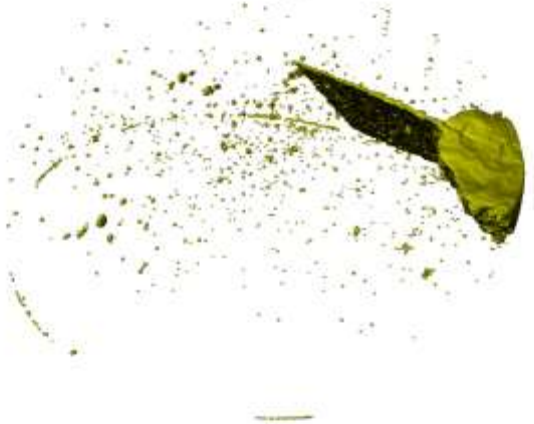
Bivariate histogram segmentation provides better fidelity on strong organic layers and captures additional mineral content (magenta)

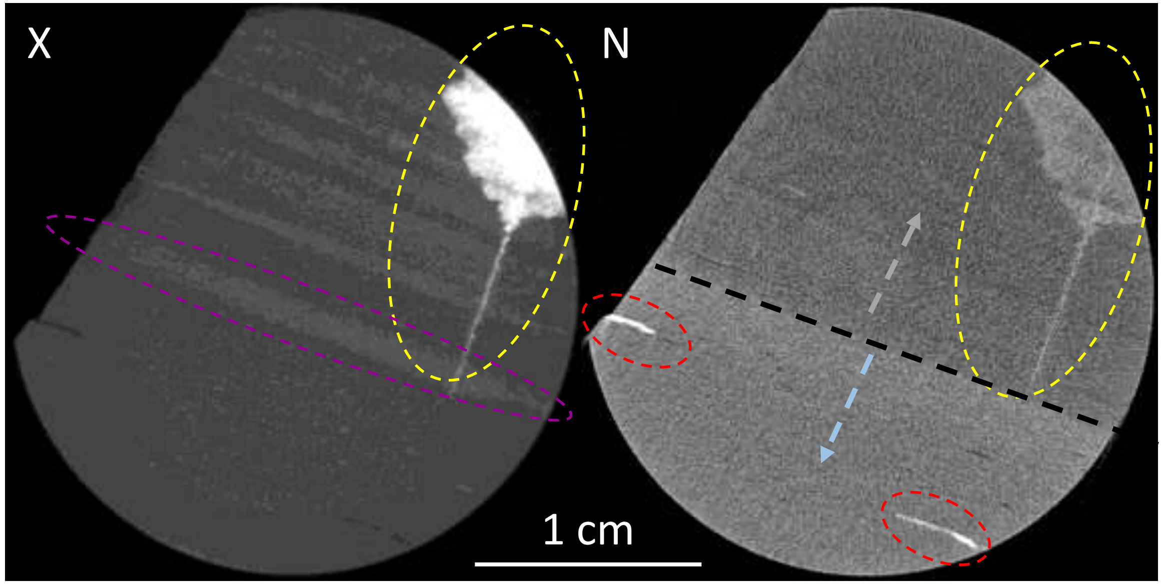
W. Chiang, et al, Petrophysics, 59(02), 2018, 153-161. https://doi.org/10.30632/PJV59N2-2018a3.
J.M. LaManna, et al, Microscopy & Microanalysis, Vol 26, 2020, 3220-3221. https://doi.org/10.1017/S1431927620024204.

Organic Matter and Mineral ID in Shale [in collaboration with Aramco]

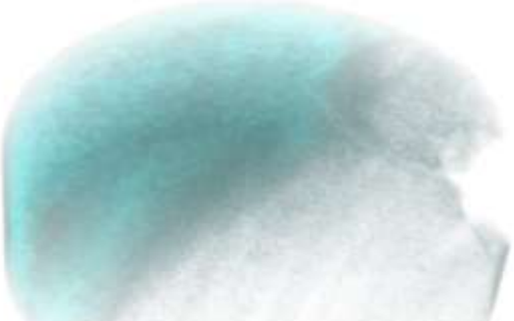
Unidentified Mineral (1.7 %)

Pyrite (3.3 %)





Bivariate histogram segmentation improves selectivity and identification of constituent components



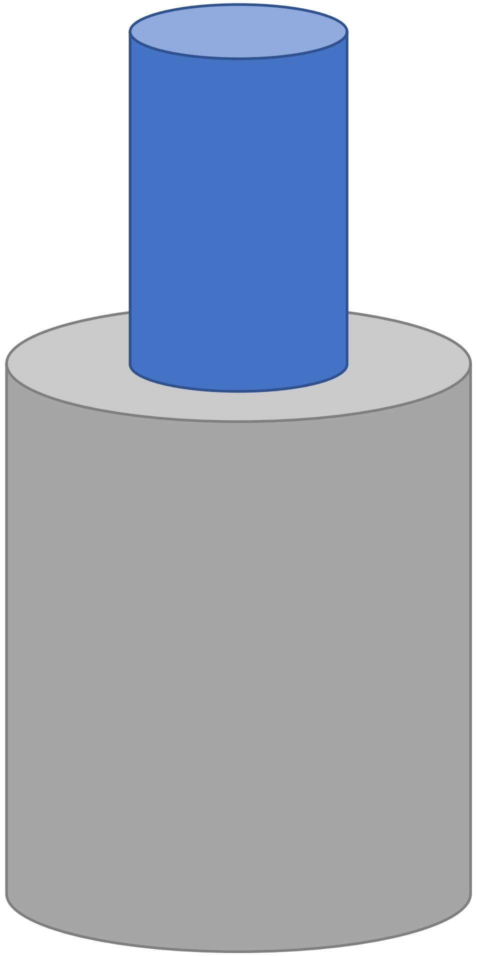
Higher Contrast Matrix (50.9 %)

Lower Contrast Matrix (43.5 %)

Organic Matter (0.5 %)

W. Chiang, et al, Petrophysics, 59(02), 2018, 153-161. https://doi.org/10.30632/PJV59N2-2018a3.
J.M. LaManna, et al, Microscopy & Microanalysis, Vol 26, 2020, 3220-3221. https://doi.org/10.1017/S1431927620024204.

Water Infiltration into Concrete [Collab: NCSU]

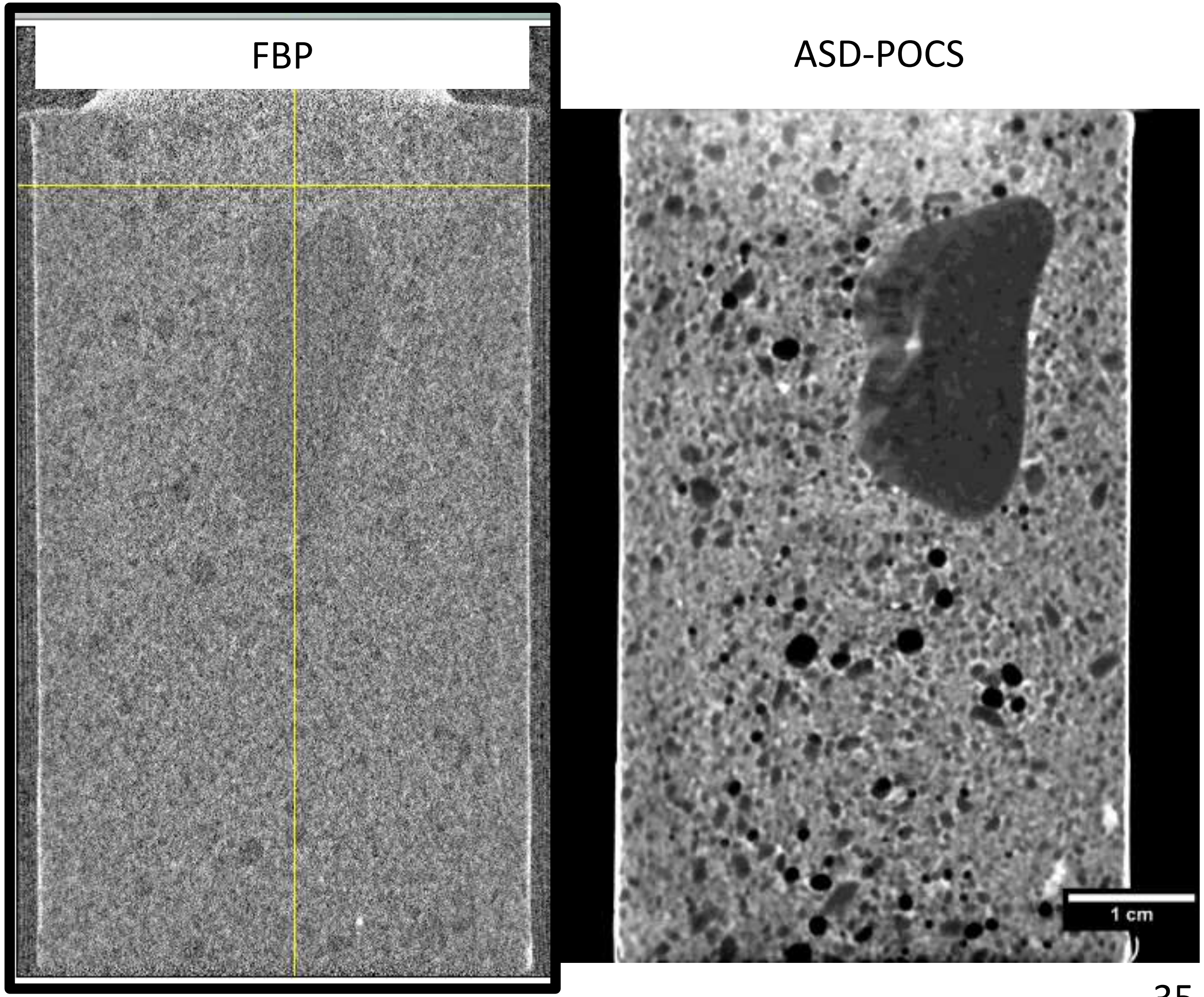


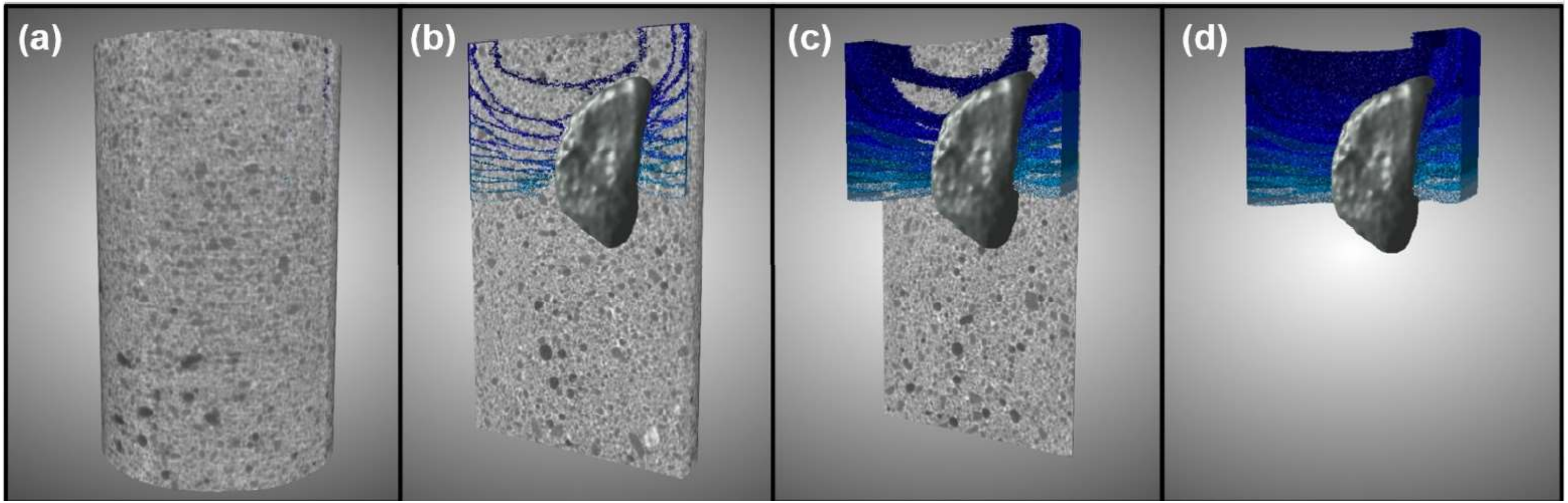
1.5” diameter core

Core made from cement with fine aggregates, one large aggregate placed near center

Column of water placed on top of core to provide reservoir for infiltration experiment

1 h tomography scans acquired with only 60 projections





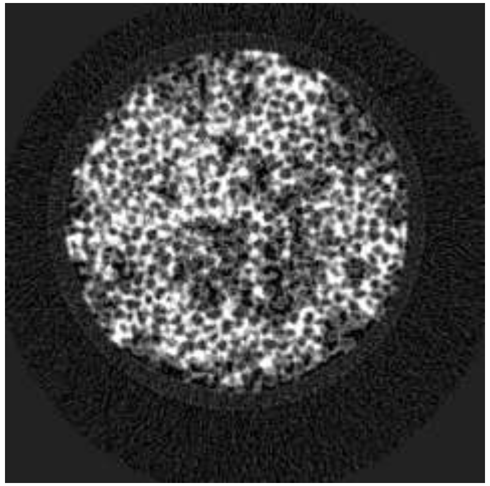
Legend: (a) 38 mm diameter concrete cylinder, (b) cut-away view revealing large aggregate and contours of progressive water infiltration, (c) 3D water contours, (d) aggregate and water contours only

Water placed on top of 38 mm diameter concrete cylinder made with mostly fine aggregates except for one large aggregate. Sample scanned with NeXT every hour for 8 hours to track water infiltration with time to understand interfacial effects along cement/large aggregate interface.

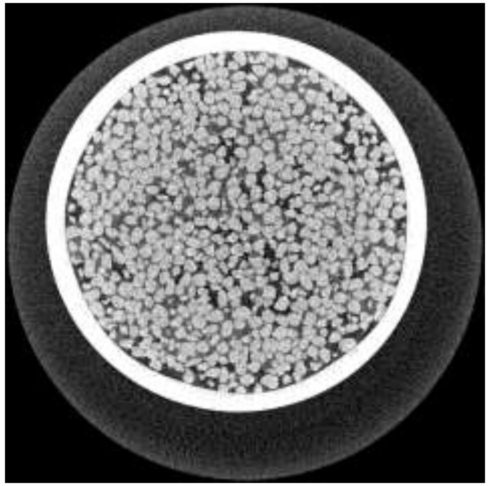
Simultaneous tomography critical to capture the water infiltration and changes to the concrete. Cement will swell with increases in hydration which can cause deformation in the material. The swelling effect is the primary driving force in the slowing of the infiltration with time.

Current work in collaboration with Prof. Mohammad Pour-Ghaz and PhD student Laura Dalton in the Civil Engineering Department at the North Carolina State University

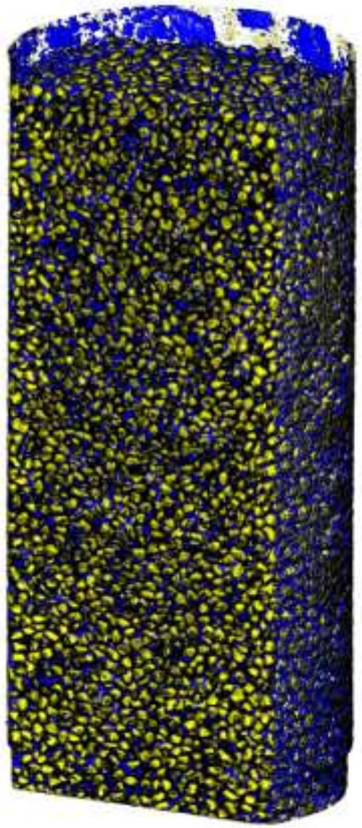
Bimodal Segmentation of water evaporation in sand columns [Univ. of Delaware]



Neutron



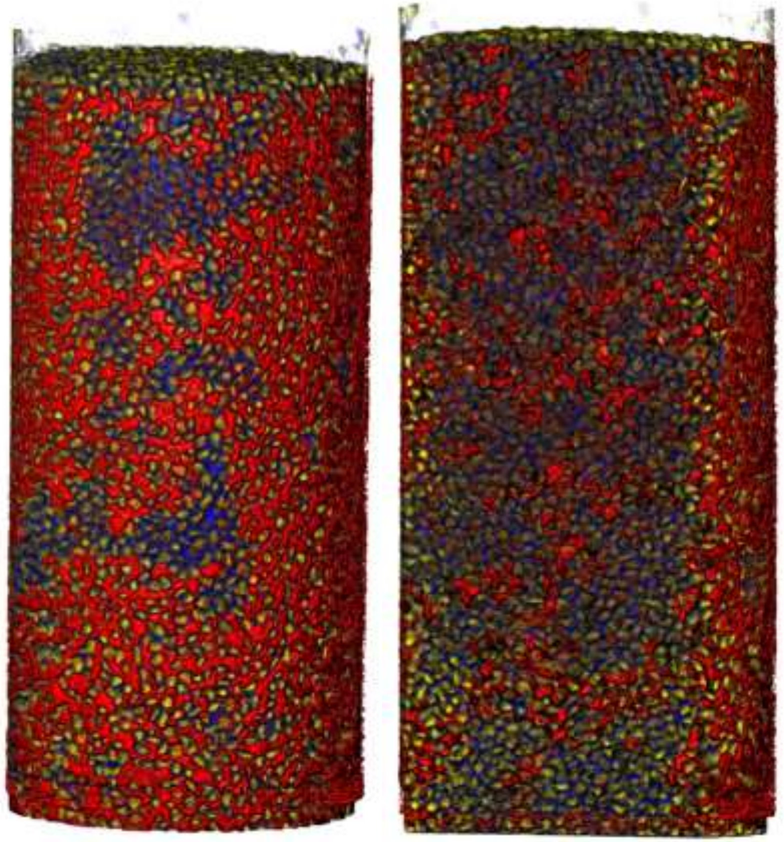
X-ray

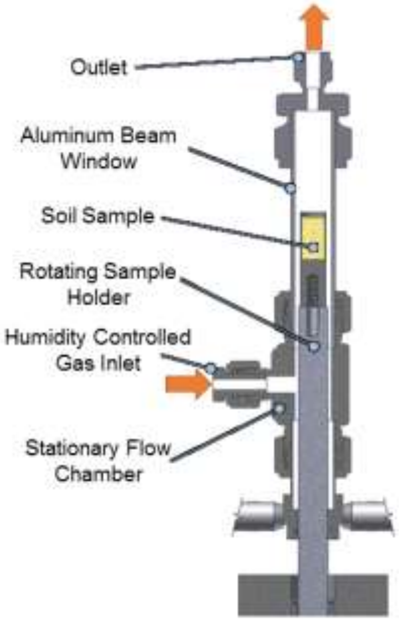


Water

Sand

Air

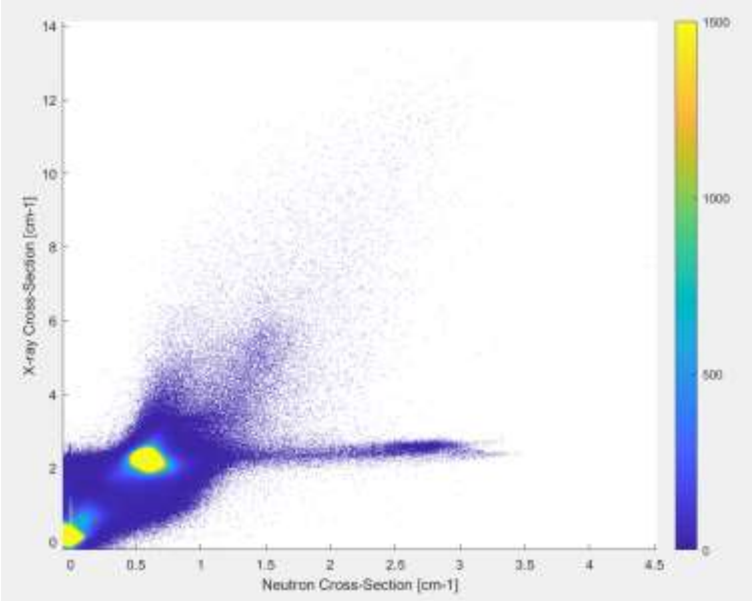


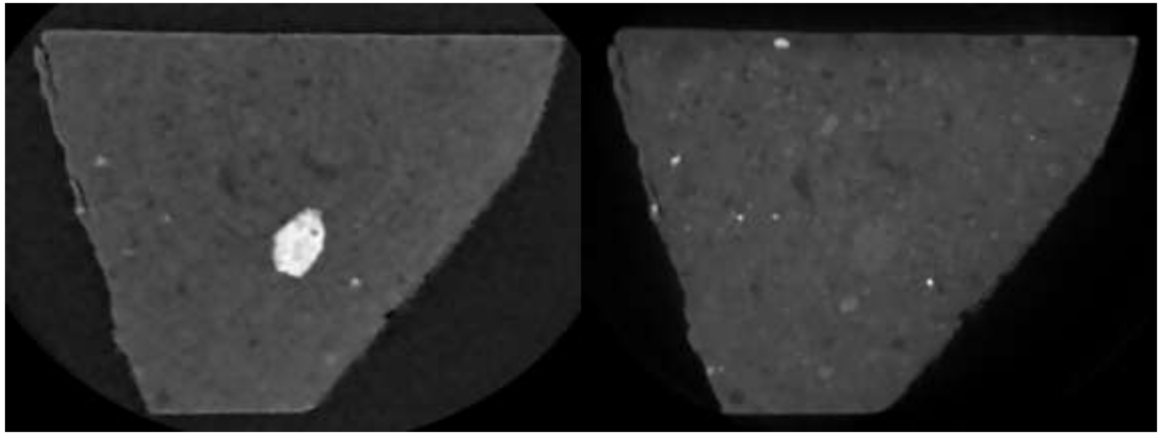


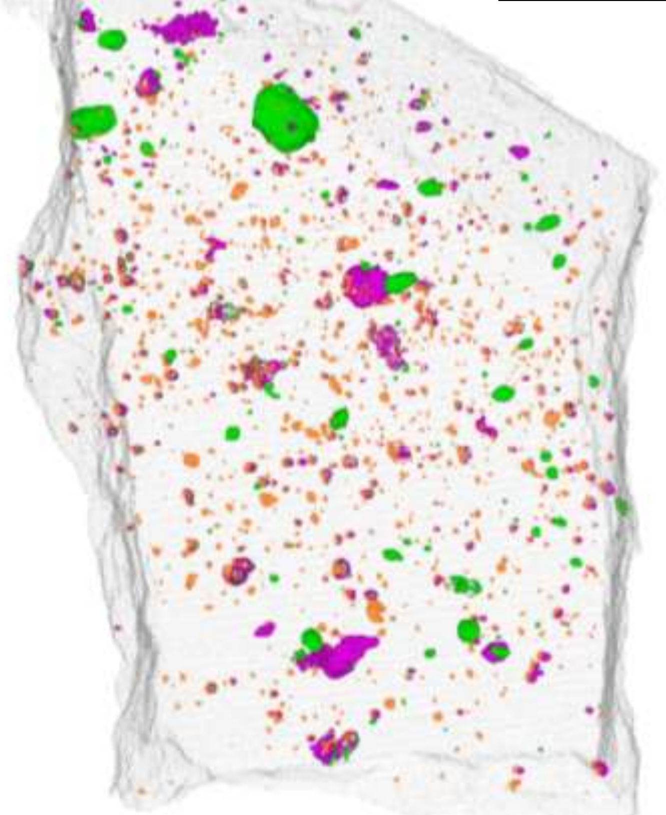
Bimodal segmentation improves water identification, especially water films on sand particles.

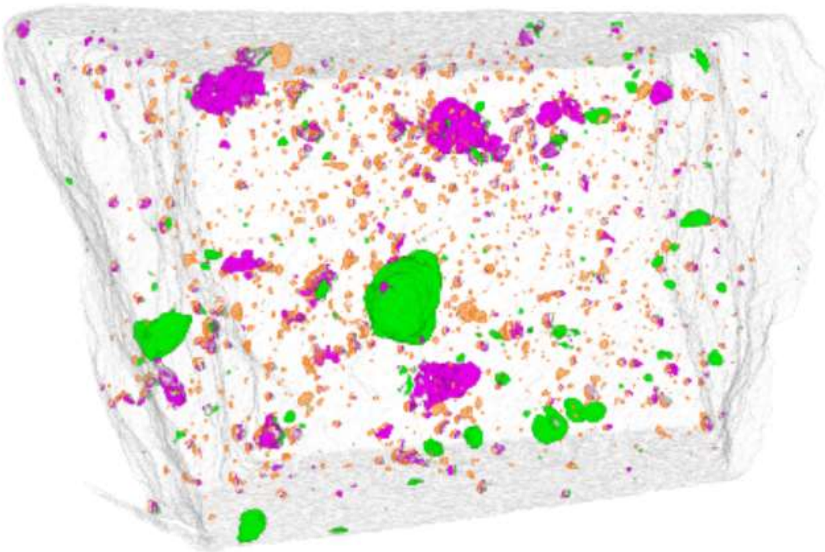
Calculation of water volume and comparison to weighed value pending

Meteorite from asteroid 4 Vesta



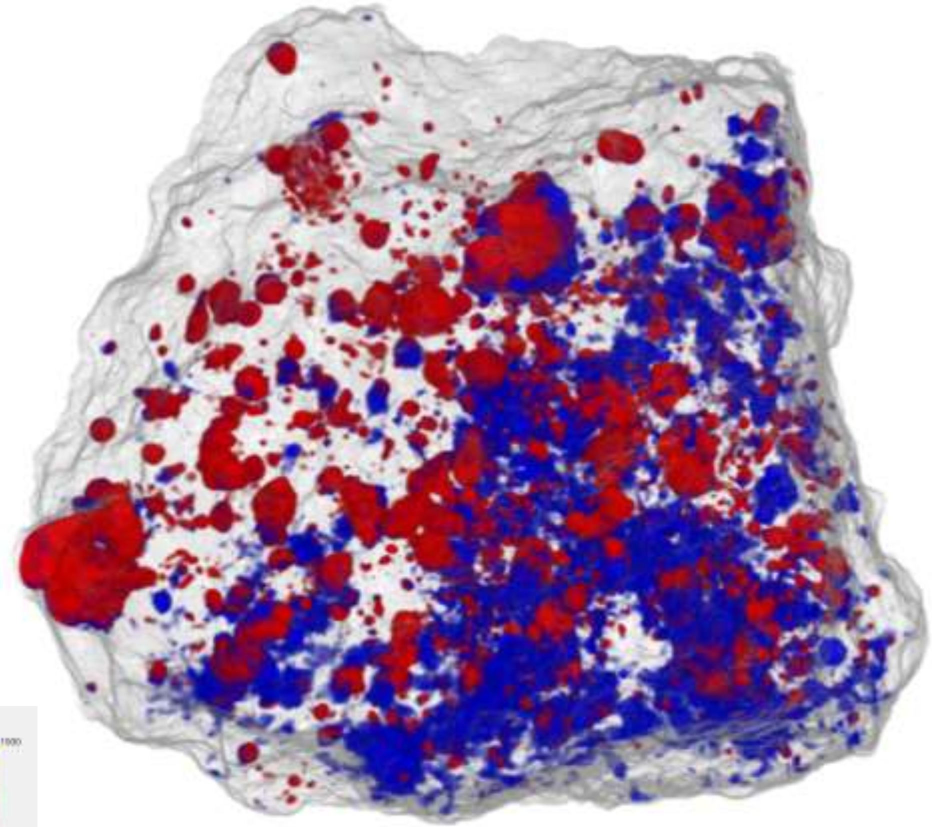


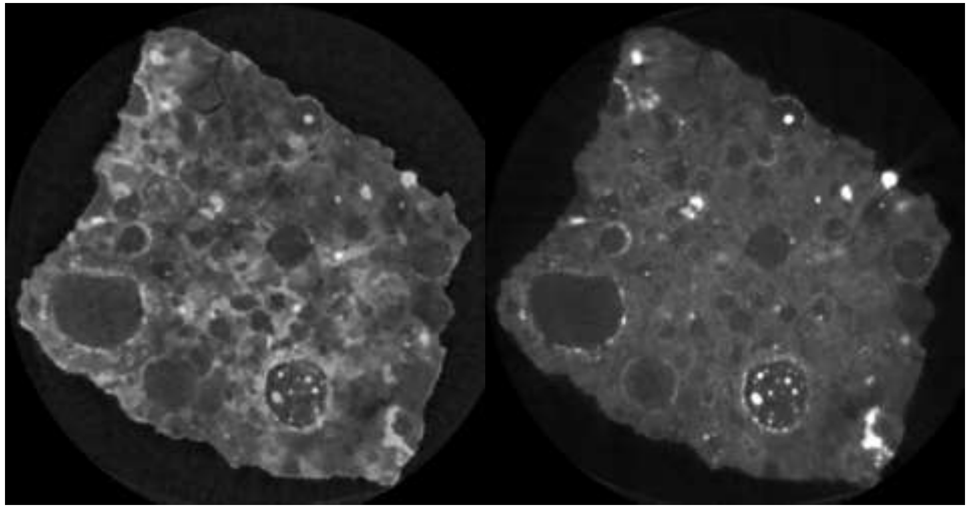


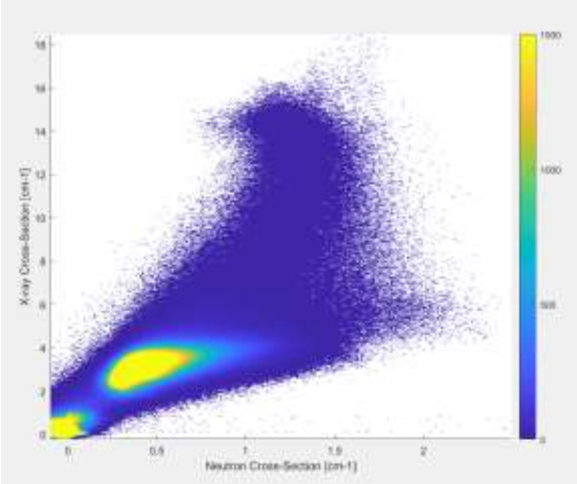


A.H. Treiman et al, Meteorics and Planetary Science, 2022, doi: 10.1111/maps.13904

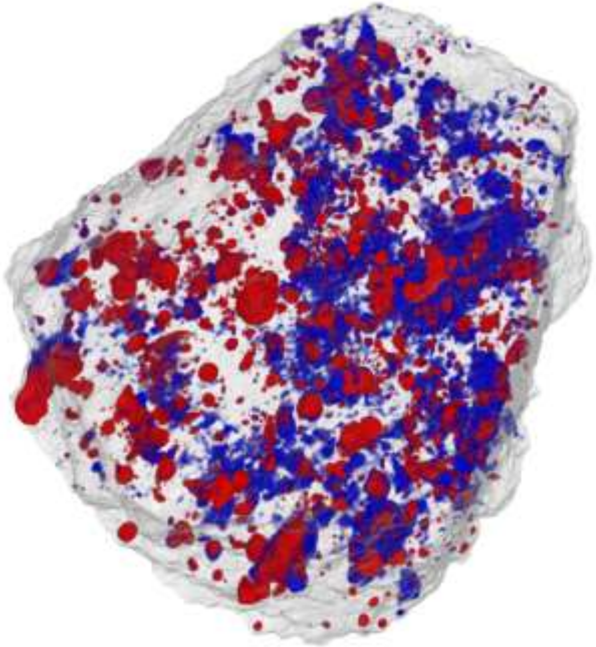
CR2 Chondrite meteorite from early solar system





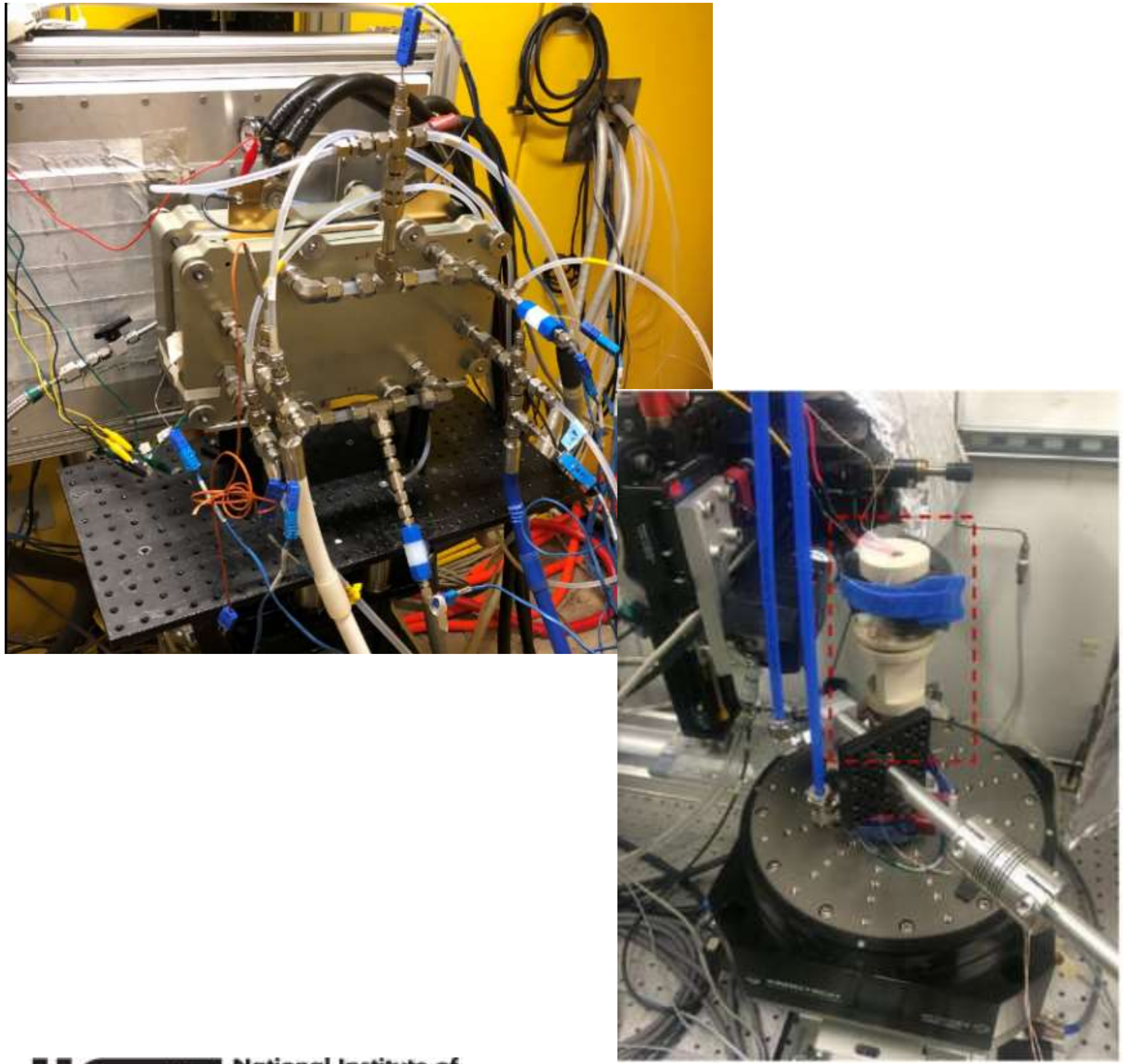


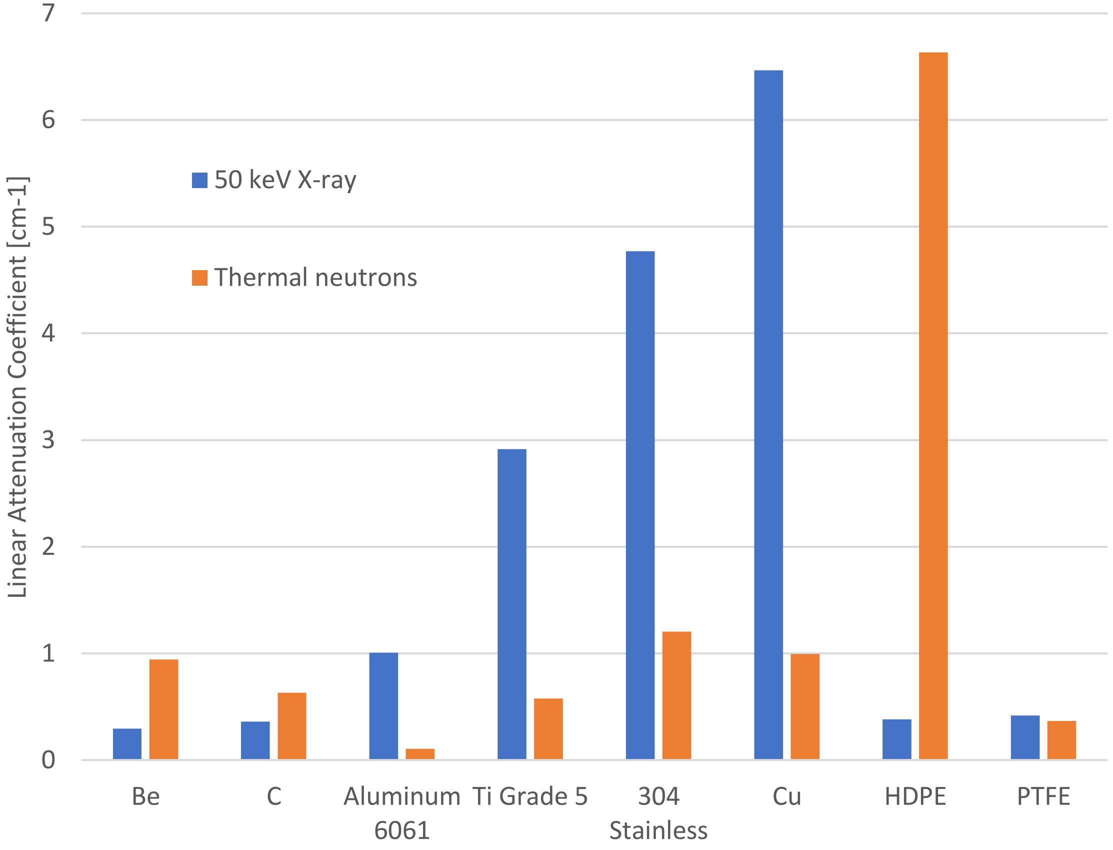
NeXT tracks the extent of aqueous modification within the meteorite. The combination of X-rays with neutrons are necessary to fully separate aqueously modified material from the iron-nickel chondrules and shells.



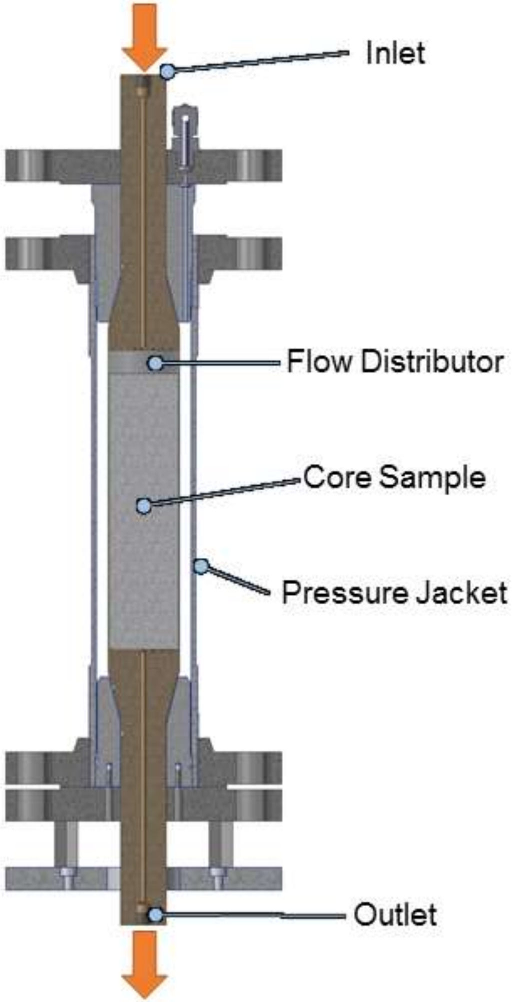
A.H. Treiman et al, Meteorics and Planetary Science, 2022, doi: 10.1111/maps.13904

How to design samples and environments for NeXT





Sample environment available for geological samples

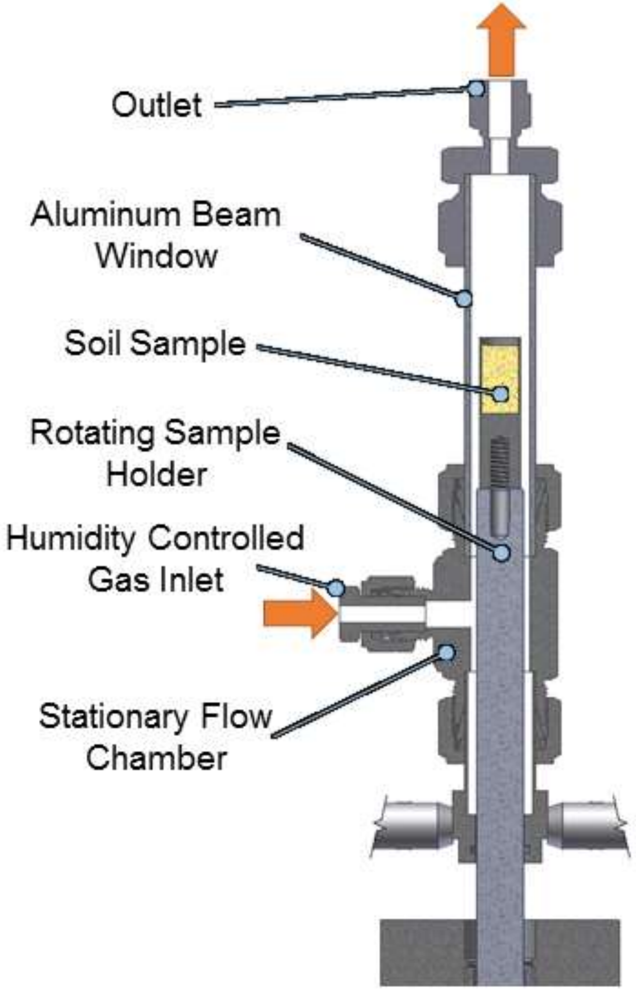


High Pressure Axial Flow Cell

- Confining pressure to simulate subsurface conditions
- Aluminum walls allow X-ray tomography through vessel
- Current cell for large 38 mm diameter by 152 mm long cores
- Non-metallic, chemical resistant wetted components

Evaporation Control Chamber

- Humidity controlled gas flows through cell to prevent or facilitate evaporation
- Gas flow rate, humidity, temperature, and pressure controlled by fuel cell test stand
- Thin aluminum walls for transparency

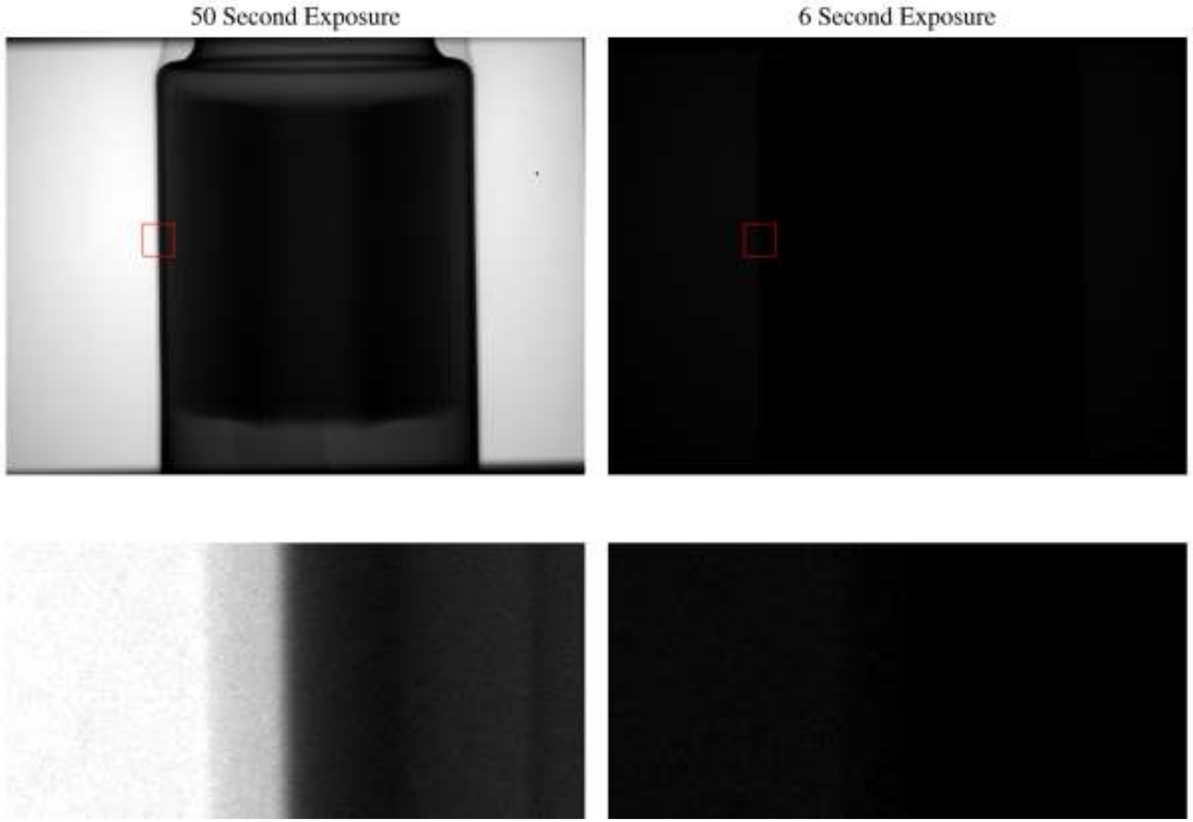


Neutrons and X-rays can penetrate through sample environment that provide confining pressure and/or environmental controls for temperature, humidity, etc.

Technique Developments for Dynamic Systems

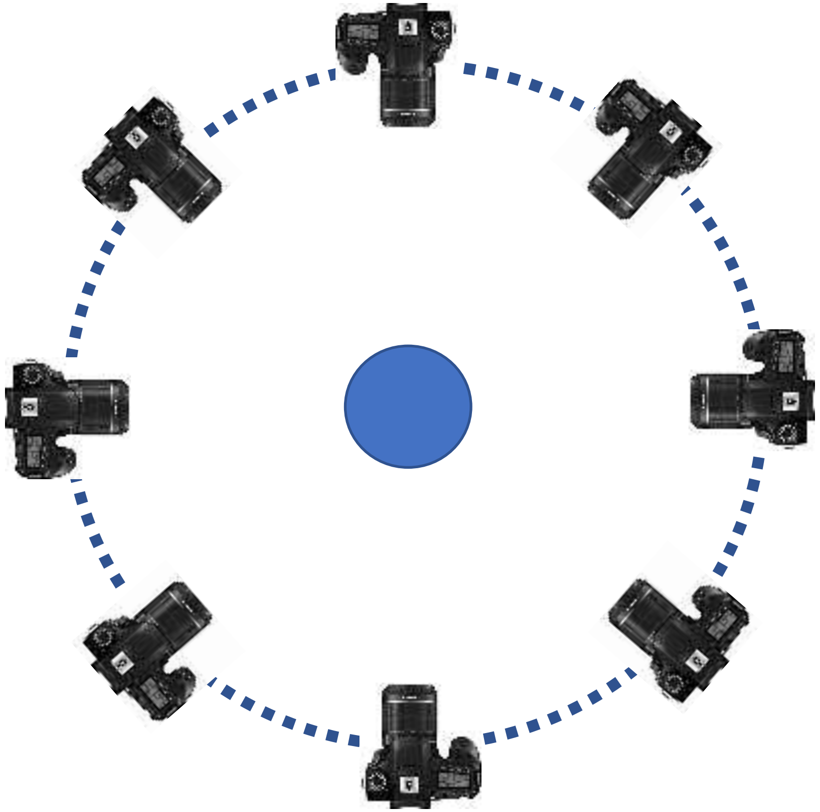
Options to reduce tomography acquisition time

Reduce exposure time



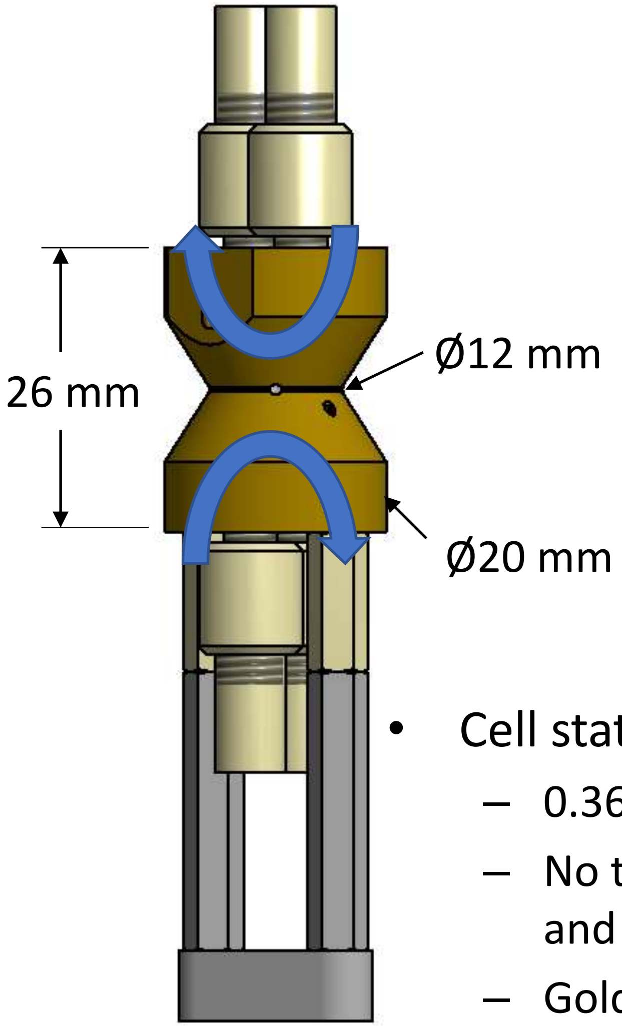
Cons: reduced SNR

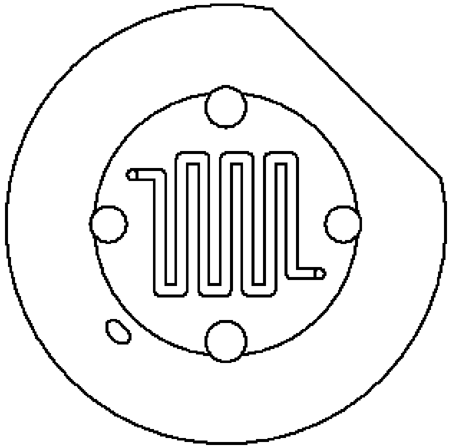
Reduce number of projections



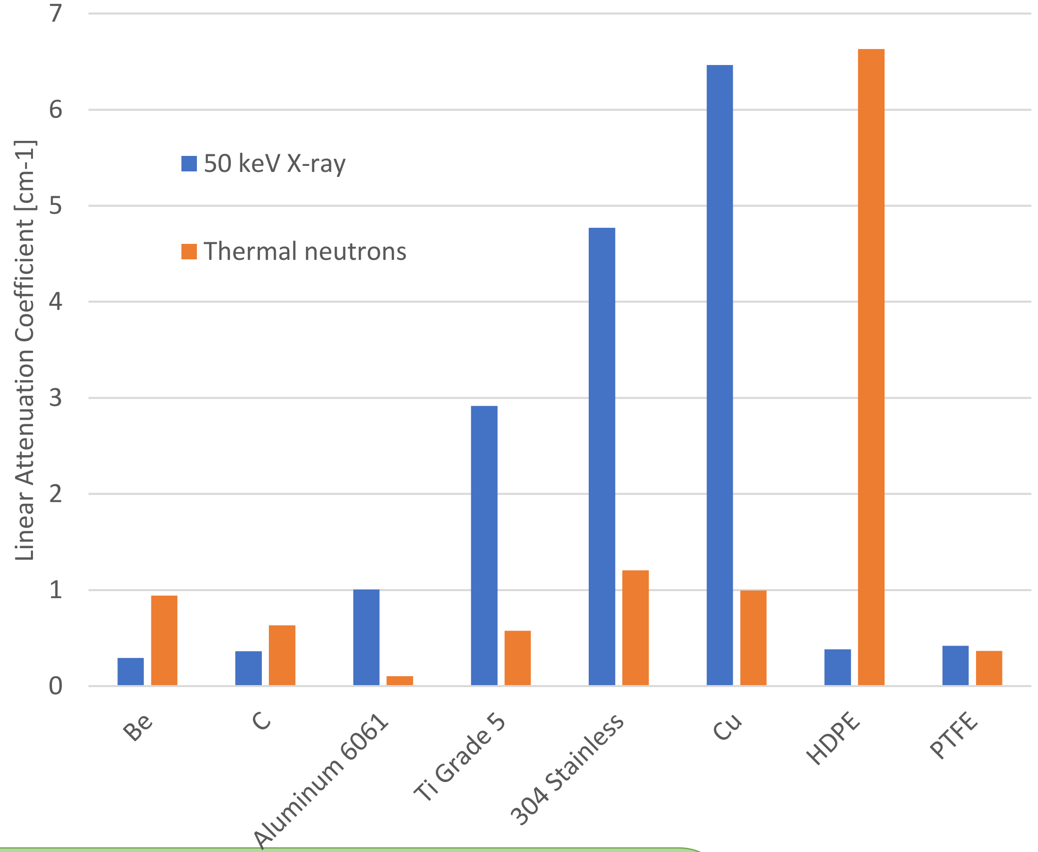
Cons: potential loss of reconstructed resolution

Material selection for NeXT compatible fuel cell





Channels 0.5 mm (w) x 0.4 mm (d), 0.5 mm (w) lands

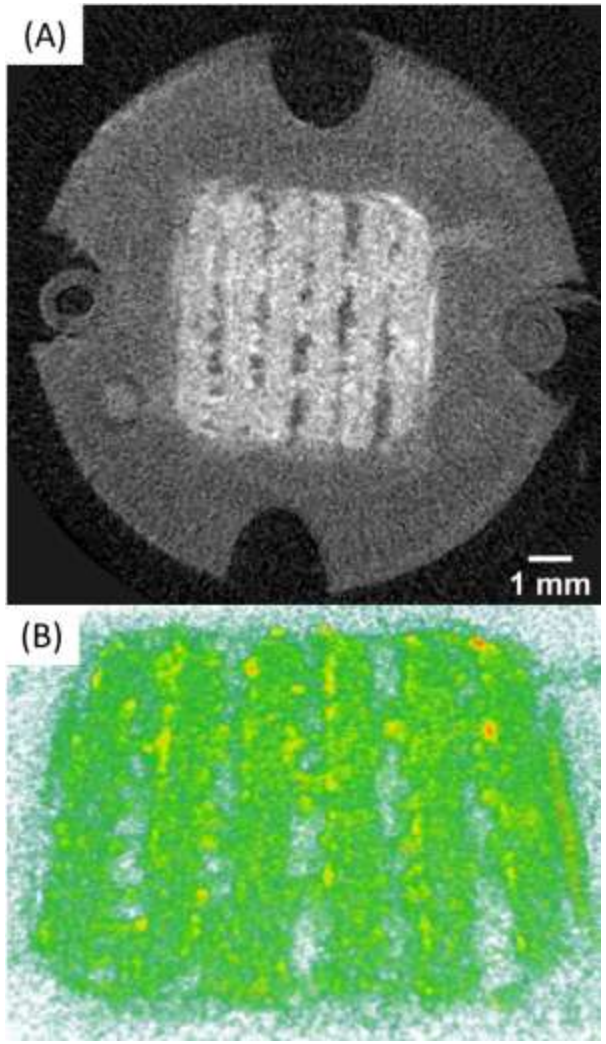


- Cell stats:
  - 0.36 cm^2 active area
  - No temperature control due to size and time constraints
  - Gold coated aluminum construction

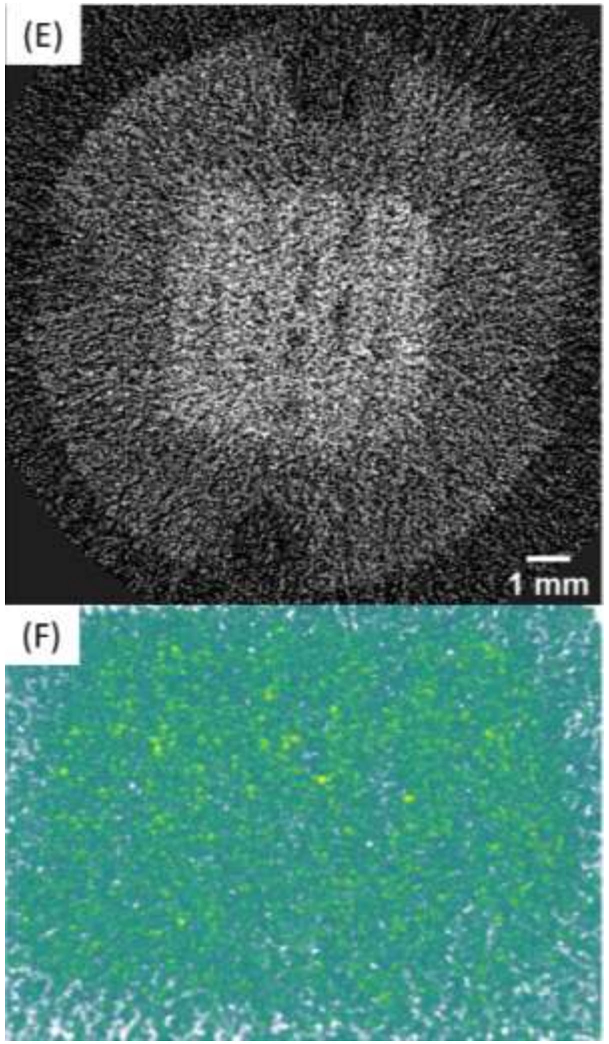
Use of aluminum for hardware allows highest possible neutron transmission while being acceptable for X-ray. Horizontal orientation allows the use of light water temperature control.

What Happens When You Reduce Projection Numbers

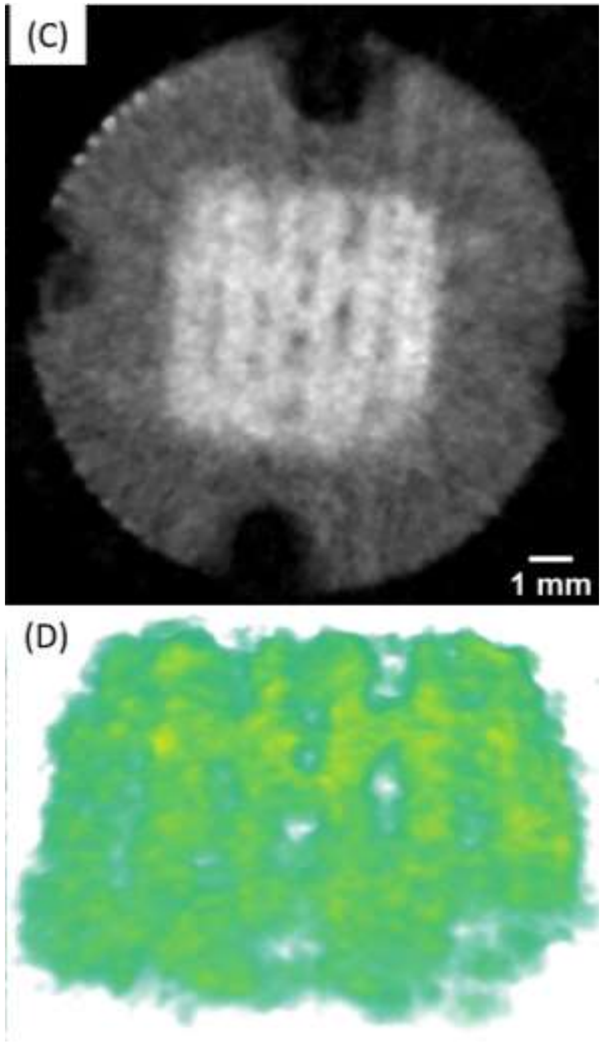
High Res FBP



Simulated 30 minute scan
FBP



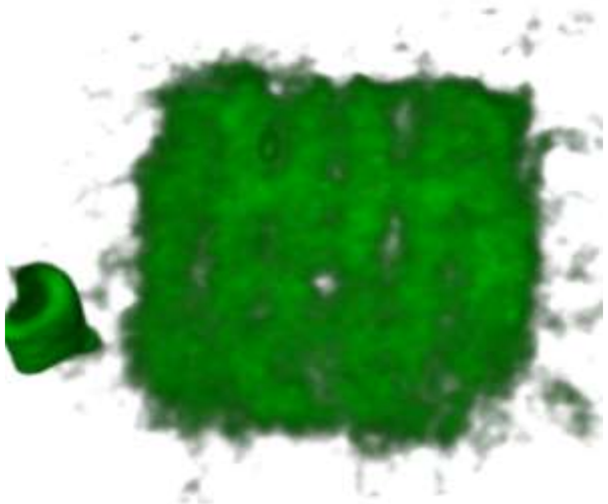
Simulated 30 minute scan
ASD-POCS w/ seed



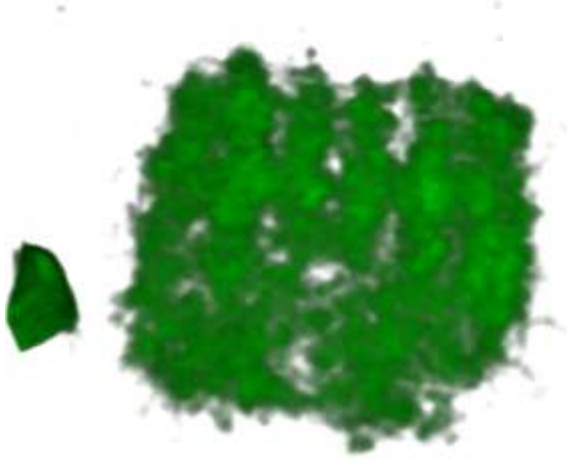
How Fast Can We Image in 3D?

Ground Truth
851 Projections
8 hours scan time

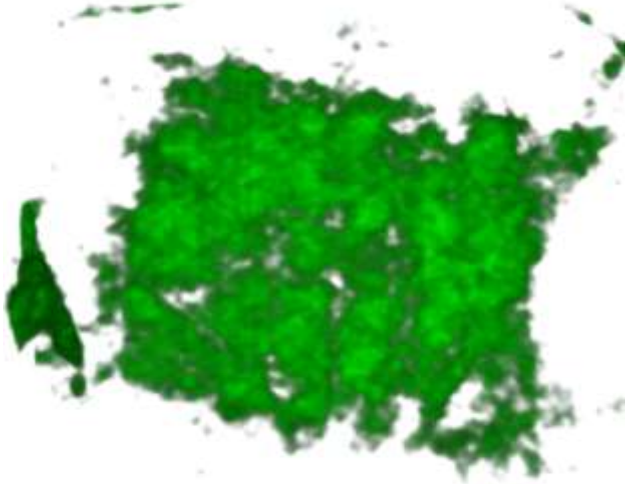
Iterative Reconstruction 1
54 Projections
10 minute scan time



Iterative Reconstruction 2
27 Projections
5 minute scan time



Iterative Reconstruction 3
14 Projections
2.5 minute scan time



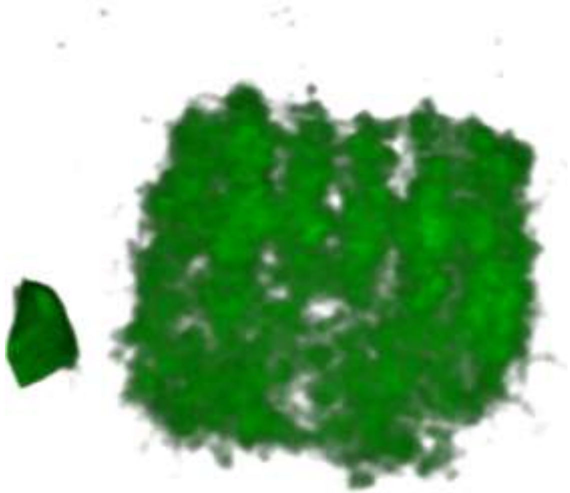
- Switched from SIRT to ASD-POCS algorithm
- Only used single frames from each projection
- Each fast scan is simulated by selecting subset of projections from ground truth
- Dry high resolution reconstruction used as seed

Dry seeded ASD-POCS shows superior quality to seeded SIRT. Structure shown in 27 projection recon shows similar structure to ground truth. This shows it is possible to acquire fast neutron tomography scans!

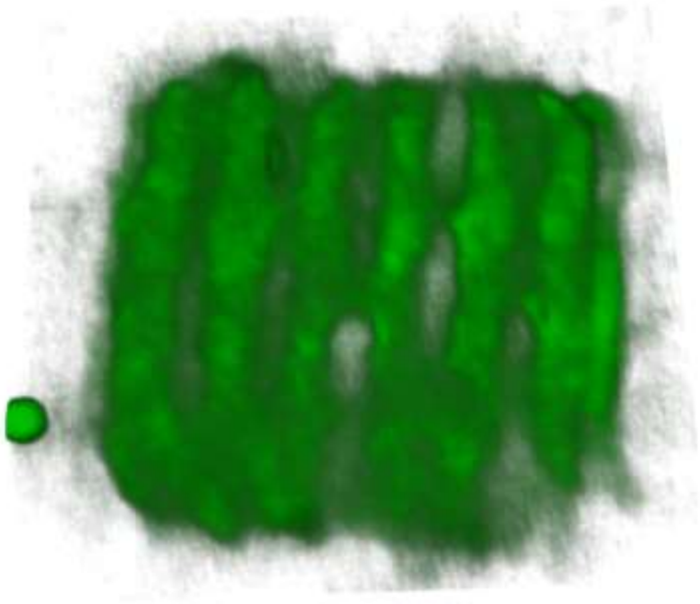
Machine learning algorithms show promise to further reduce scan time

Ground Truth
851 Projections
8 hours scan time

Iterative Reconstruction
27 Projections
5 minute scan time



FBPConvNet Reconstruction
27 Projections
5 minute scan time



- Investigating machine learning algorithms to improve reconstruction quality of sparse scans
- Currently lacking on training data
- Have looked at one algorithm, beginning to look at others

FBPConvNet machine learning denoising algorithm shows great promise to improve reconstruction quality. Need improved training data.

Conclusions and Outlook

- Wolter optics will unlock neutron imaging performance not seen before opening a new class of experiments
- Neutron grating interferometry can potentially provide robust 3D resolved SANS data for geological samples
- Bragg-edge imaging can map and classify multiple minerals in geological samples, potentially providing identification of the minerals
- The NIST-NeXT system is a powerful tool for geology and planetary research
- Neutrons and X-rays strongly complement each other for material identification
- We are looking to improve the method through upgrades to the instrument and increasing robustness of analysis tools
- We are looking for collaborators and postdocs interested in working with multimodal, multidimensional data

Thank you!

For more information:
Email jacob.lamanna@nist.gov