Development of Hydrogen Transport Models for High Temperature Metal Hydride Moderators

March 4-5, 2025

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Project Motivation

- Minimizing core size is an important consideration in many (all?) microreactor concepts
- The incorporation of a moderator into a reactor core can reduce the amount of fissile material required for criticality
  - This can lead to reduced size or reduced enrichment
- The high temperatures expected in most microreactor designs limit the use of water (the most common moderating material)
- Metal hydrides can be nearly as effective as water at elevated temperatures
- Understanding the fabrication, incorporation, and performance of high temperature metal hydride moderators is an enabling technology for the development of future microreactors
- This project seeks to develop validated computational methods to predict the short- and long-term reactor performance impacts from hydrogen transport in zirconium- and yttrium-hydrides

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Project Objectives

- Develop neutron radiography techniques to measure time-dependent hydrogen concentrations in metal hydride moderators
- Derive updated hydrogen diffusion coefficients for metal hydride moderator materials
- Demonstrate and validate multiphysics-based reactor performance models incorporating improved models for the transport of hydrogen in metal hydride moderators

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Project Plan

- Year 1
  - Develop neutron imaging techniques to measure hydrogen content in metal hydrides

- Year 2
  - Collect data on the diffusion of hydrogen in yttrium and zirconium hydride in response to chemical, stress, and thermal gradients
  - Derive appropriate transport models for the diffusion of hydrogen in hydride moderators

- Year 3
  - Update reactor simulation codes (BISON and GRIFFIN) with the new diffusion models
  - Demonstrate the impact short- and long-term hydrogen mobility on the performance of hydride moderated microreactors

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Driving Forces for Hydrogen Migration

- Hydrogen in zirconium is driven by three gradients in the material.[1]
  - Concentration,
  - Temperature,
  - And stress.
- The generalized equation for hydrogen migration flux is:[1]

$$J_H = -D_H * \left(\nabla \cdot C_{ss} + \frac{Q^* C_{ss}}{RT^2} * \nabla \cdot T + \frac{V^* C_{ss}}{RT} * \nabla \cdot \sigma_h \right)$$

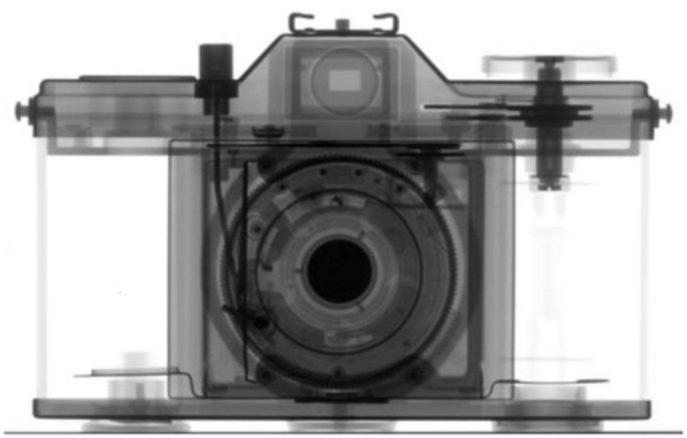
Fickian — Soret Effect — Stress Cross-Effect

- Interstitial solutes will diffuse towards areas of:[1]
  - Low solid solution concentration.
  - Low temperature.
  - High tensile hydrostatic stress.
- Accurate measurements of D_H, Q^*, V^* are required for accurate modeling.

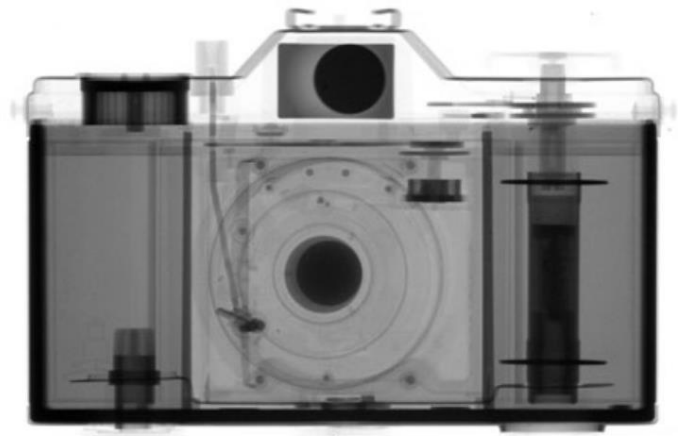
[1] B. F. Kammenzind, B. M. Berquist, R. Bajaj, P. H. Kreyns, and D. G. Franklin, The long range migration of hydrogen through Zircaloy in response to tensile and compressive stress gradients, Bettis Atomic Power Laboratory, Pittsburgh, PA, rep., 1998.

Neutron Radiography

- Neutron radiography can, theoretically, be a valuable tool for studying the presence and migration of hydrogen in metal hydrides
- Neutron beams are strongly attenuated by the presence of hydrogen and only weakly attenuated by zirconium and yttrium
- The key challenge lies in extracting high-precision quantitative information from the resulting images
  - This project is synergistic with a parallel project sponsored by Naval Reactors focuses on quantifying hydrogen behavior in zirconium cladding and structural materials using NR techniques



X-Ray Radiograph



Neutron Radiograph

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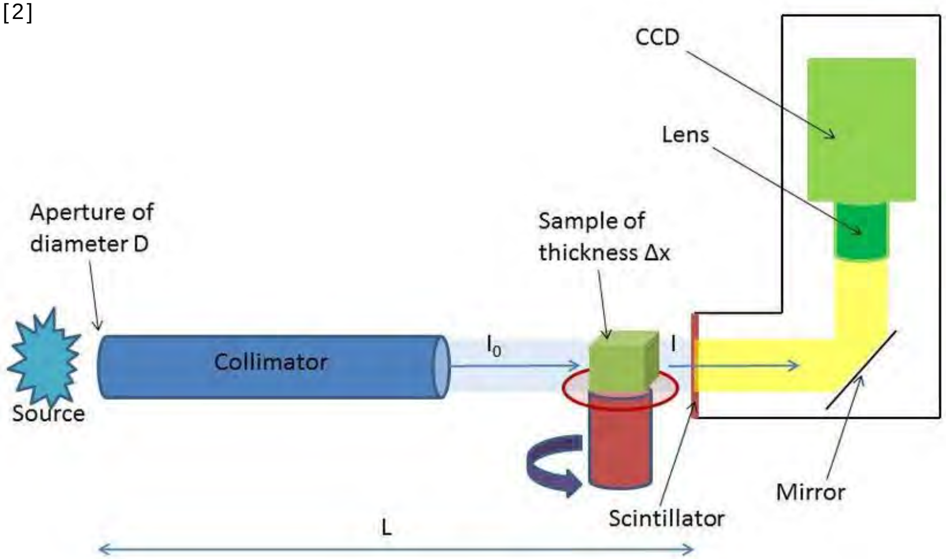
Neutron Radiography Setup

- Neutron radiography relies on the strong interaction between hydrogen and neutrons to create image contrast.[1]

[2]



Schematic of a neutron radiography set-up

- Neutrons pass through samples, are absorbed by a scintillator, emitting light, which is collected by film or a CCD camera.[1]
  - Image brightness is spatially resolved, based on the neutron beam.
  - The more neutrons that reach the scintillator, the brighter the image.

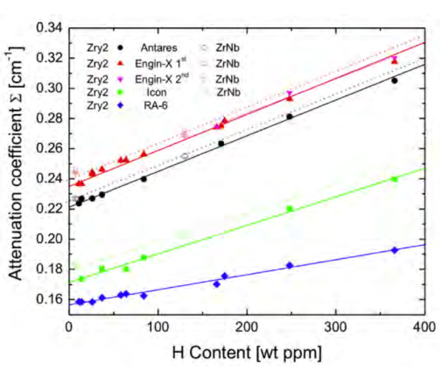
[1] N. L. Buitrago, J. R. Santisteban, A. Tartaglione, J. Marín, L. Barrow, M. R. Daymond, M. Schulz, M. Grosse, A. Tremsin, E. Lehmann, A. Kaestner, J. Kelleher, and S. Kabra, "Determination of very low concentrations of hydrogen in zirconium alloys by neutron imaging," Journal of Nuclear Materials, vol. 503, pp. 98–109, 2018.
[2] K. Ryzewski, S. Herringer, H. Bilheux, L. Walker, B. Sheldon, S. Voisin, J.-C. Bilheux, and V. Finocchiaro, "Neutron imaging of archaeological bronzes at The Oak Ridge National Laboratory," *Physics Procedia*, vol. 43, pp. 343–351, 2013.
[3] A.M. Long, J.R. Torres, D.T. Carver, C.G. Cardona, E.P. Luther, A.P. Shivprasad, C.A. Taylor, H.R. Trellue, and S.C. Vogel, In-Situ Spatial Mapping of Hydrogen in Yttrium Hydrides at LANSCE, LA-UR-22-29025, Los Alamos National Laboratory, Los Alamos, NM., rep., 2022.

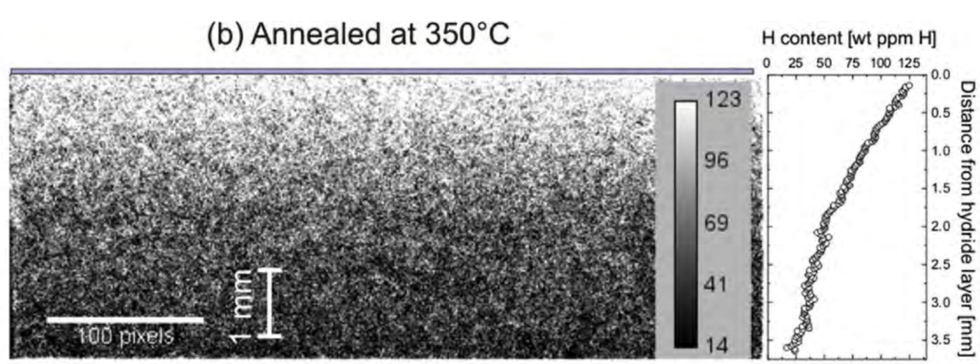
Neutron Radiography-Based Hydrogen Characterization

- The intensity of the incident neutron beam is used to characterize the transmission of the neutron beam through the sample.[1]
  - The transmission is used determine the total interaction cross section.

$$T(x,y) = \frac{I_s(x,y) - B(x,y)}{I_0(x,y) - B(x,y)} = e^{-\Sigma_t(x,y,E)z(x,y)}$$

- Hydrogen characterization requires three different scans.[1]
  1. A background scan, with no incident neutron beam, $(B(x,y))$
  2. A reference scan of the unobstructed neutron beam, $(I_0(x,y))$
  3. An obstructed scan with the sample in the neutron beam path, $(I_s(x,y))$
- The measured interaction cross section is then related to hydrogen concentration through calibration.
  - Measured interaction cross section will depend specific reactor.





[1] N. L. Buitrago, J. R. Santisteban, A. Tartaglione, J. Marín, L. Barrow, M. R. Daymond, M. Schulz, M. Grosse, A. Tremsin, E. Lehmann, A. Kaestner, J. Kelleher, and S. Kabra, "Determination of very low concentrations of hydrogen in zirconium alloys by neutron imaging," Journal of Nuclear Materials, vol. 503, pp. 98–109, 2018.

Theory Versus Reality

- While the basic physics and concept of using NR as a quantitative technique as clear, several things make that challenging
  - Neutron beams are uncommon
    - Unfortunate events can eliminate availability
      - Neither the GSTR nor LANSCE have been available to support this project
    - Pricing of replacements is inconsistent
  - Many of the available facilities are primarily qualitative
    - Is the wire connected?
    - Can we see the priming charge?
  - Imaging standards and practices to look at “shapes” is not what you would expect to look at grey scales
    - Extracting data from grey scales requires consistency and accounting for a large number of variables
    - Very few neutron sources are dedicated to a single imaging purpose

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Current Challenges

- After a few false starts, we have moved the imaging work to the McClellan Nuclear Research Center
- Beam time is essentially free (bills are paid by Defense-related QA work)
  - But, many challenges in extracting useful quantitative data
    - Significantly non-flat background
    - Likely caused by the camera setup

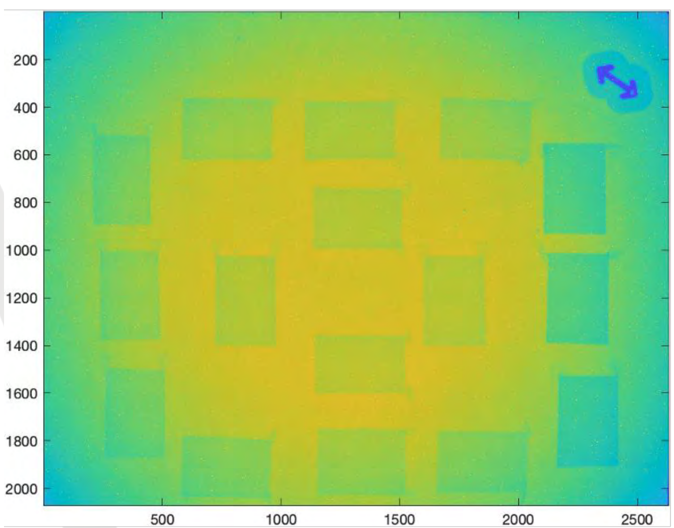

Plate A
“Radial”

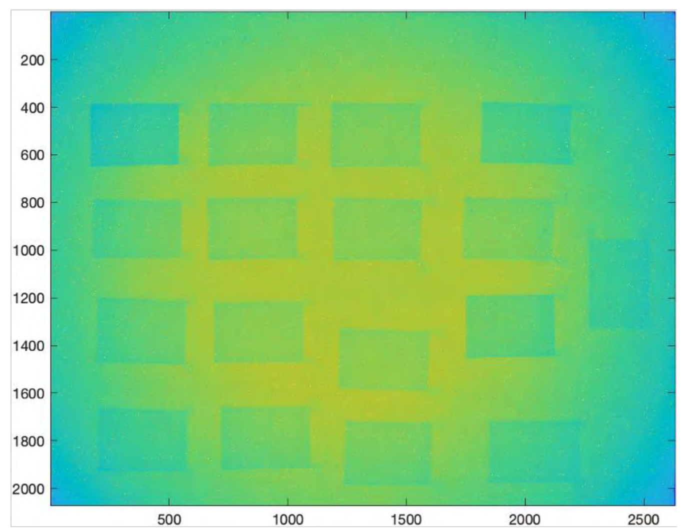

Plate B
“Block”

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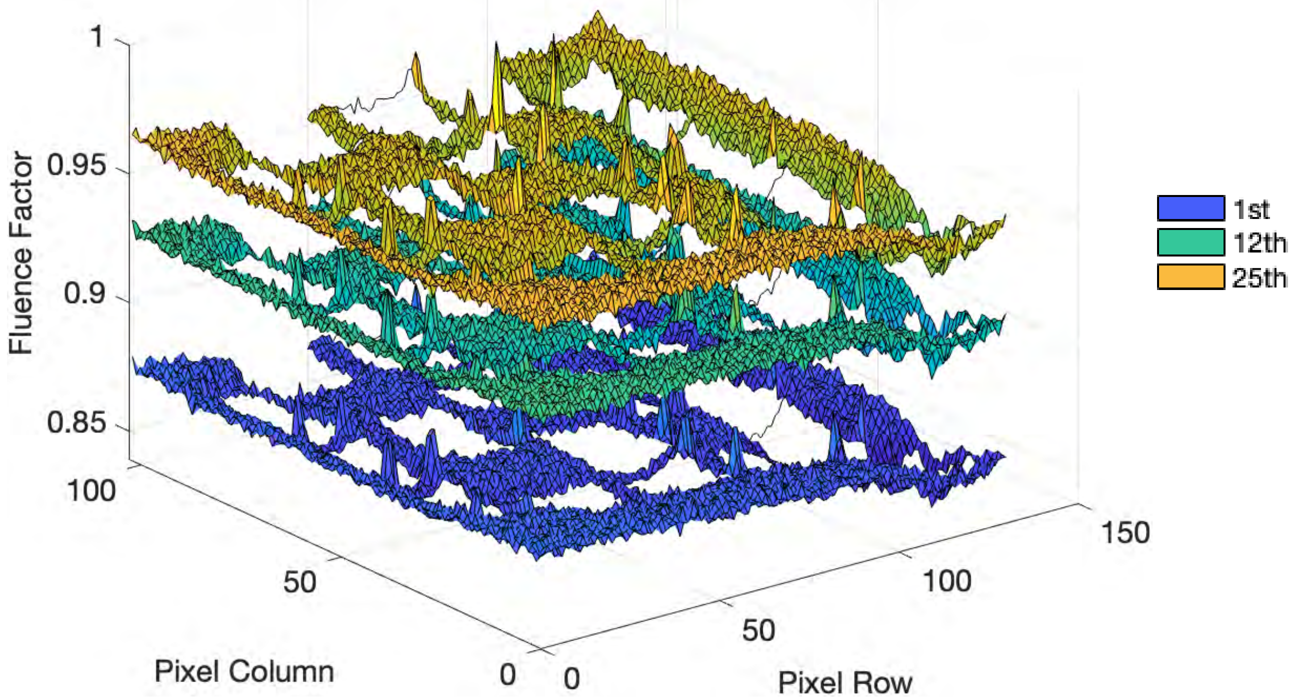
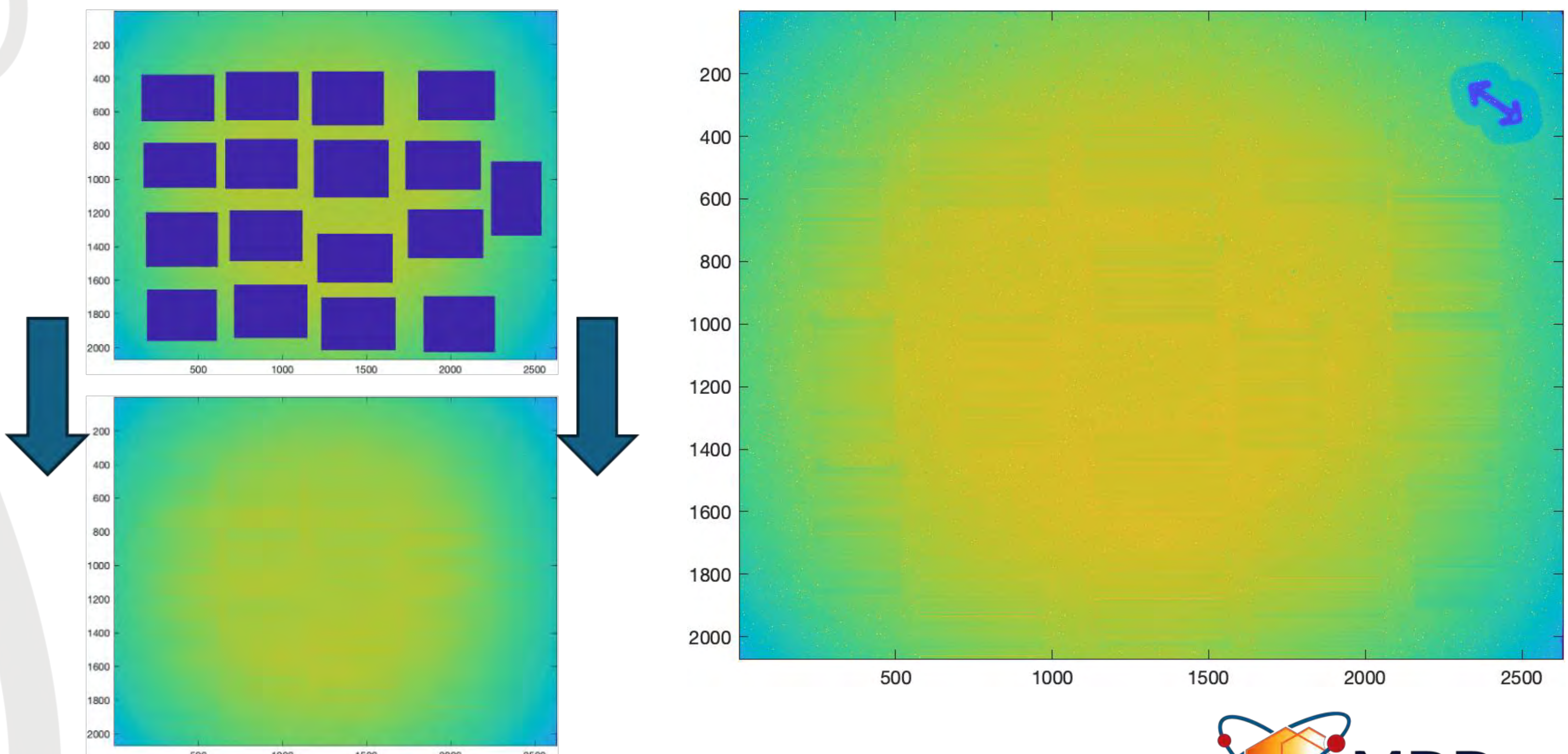
Current Challenges, cont.

- Flux also varies over time (likely a function of reflector temperature)
- This makes it very difficult get usable background / reference images



Synthetic Reference Images

- One solution may be to develop a technique to produce "synthetic" reference images
- Basically, interpolate the background gray scale values based on the non-sample areas
  - Determining the exact interpolation function is not easy



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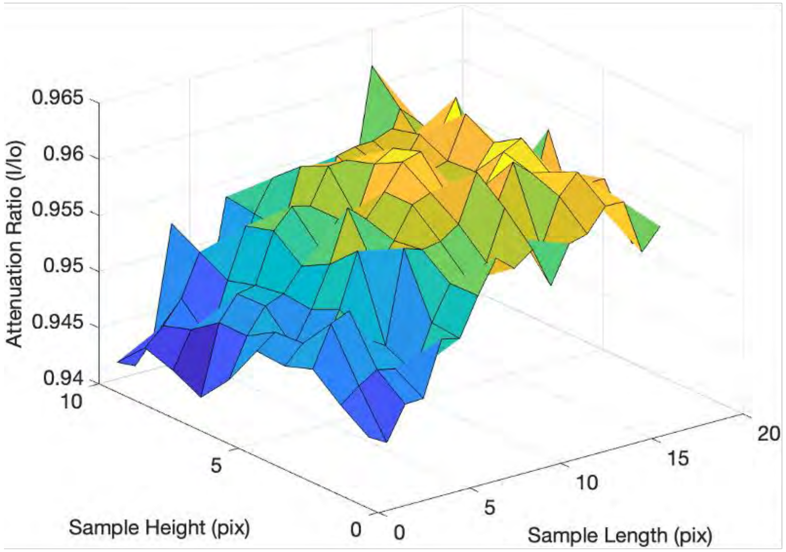
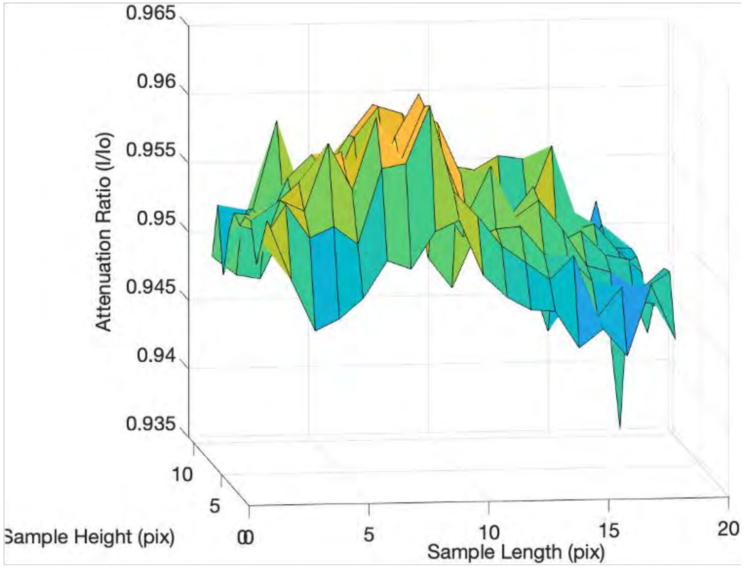
Initial Results

- The synthetic image approach seems promising
  - We can determine variable attenuation fields across samples

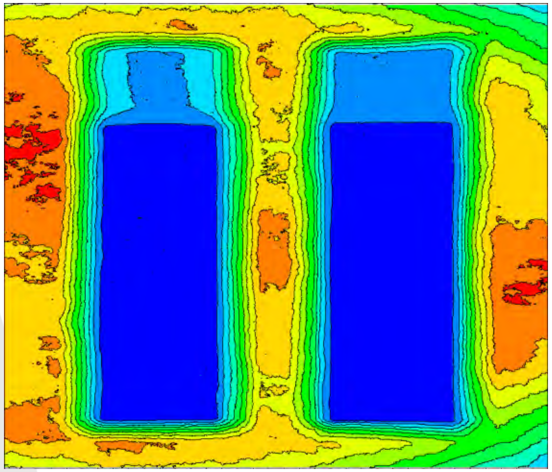




These are not the same samples!

Neutron Radiograph Simulation

- Accurate and benchmarked image simulation will improve data extraction from neutron radiographs
- This has several pre-requisites
  - Good understanding of the experimental facility
    - Neutron energy, angular distribution, etc
  - A model that is an accurate representation of the physical beamline
    - MCNP does some of this well
    - Accurate beam line models can be challenging
    - Can become very computationally intensive
  - Post-processing that accurately represents the imager response



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Proof of concept

- Object with 8 different zirconium hydride ratios
  - Tested as computational solid geometry as well as unstructured mesh

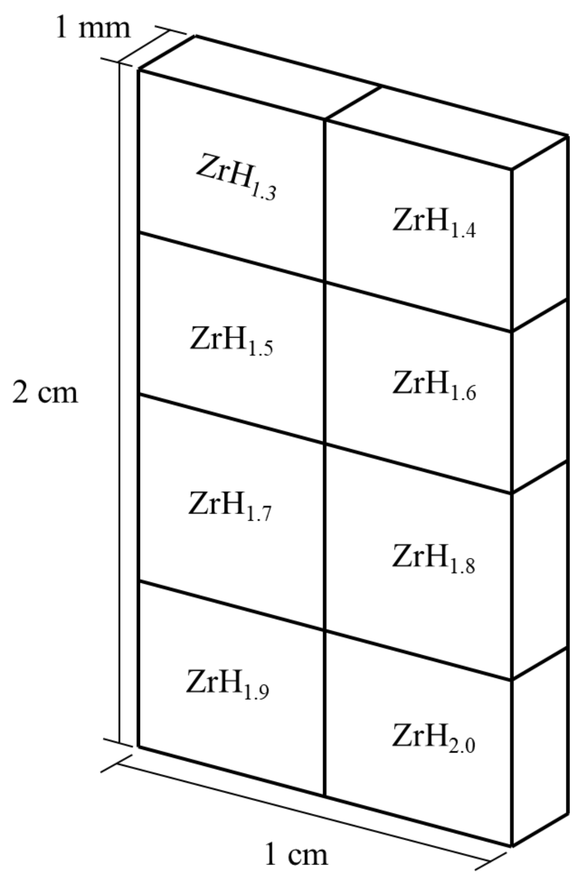

Figure 3. Zirconium hydride coupon

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Proof of Concept Results

- Using a monoenergetic, monodirectional, radially uniform source
- Using a FMESH tally array to measure neutron flux behind the object
- Convert neutron flux to grayscale
- Demonstrate grayscale differences corresponding to areas behind different hydride ratios
- Very long run times to get meaningful results

Figure 4. 16-bit grayscale representation of neutron field created by object in figure 3

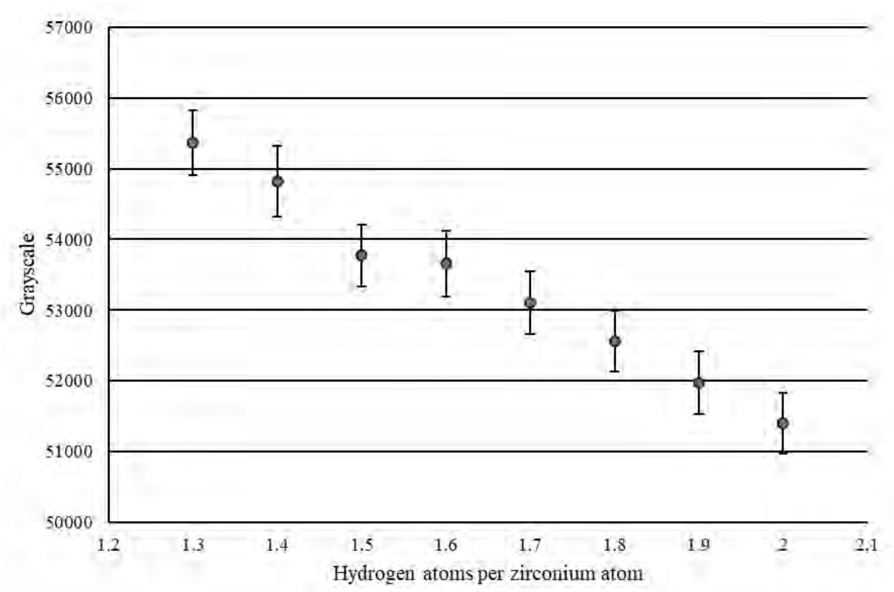

Figure 5. Plot of grayscale values in figure 4 as function of the represented hydrogen stoichiometry

Improved Neutron Source Term

- Replace the approximated neutron source term in the radiography model with an accurate representation of the neutrons exiting the beamline
  - Use a ptrac file to record the position, direction and energy of the neutrons as they cross the beamline exit surface in the GSTR model
  - Process these tracks to build distributions of the position, direction and energy that the radiography model can sample from

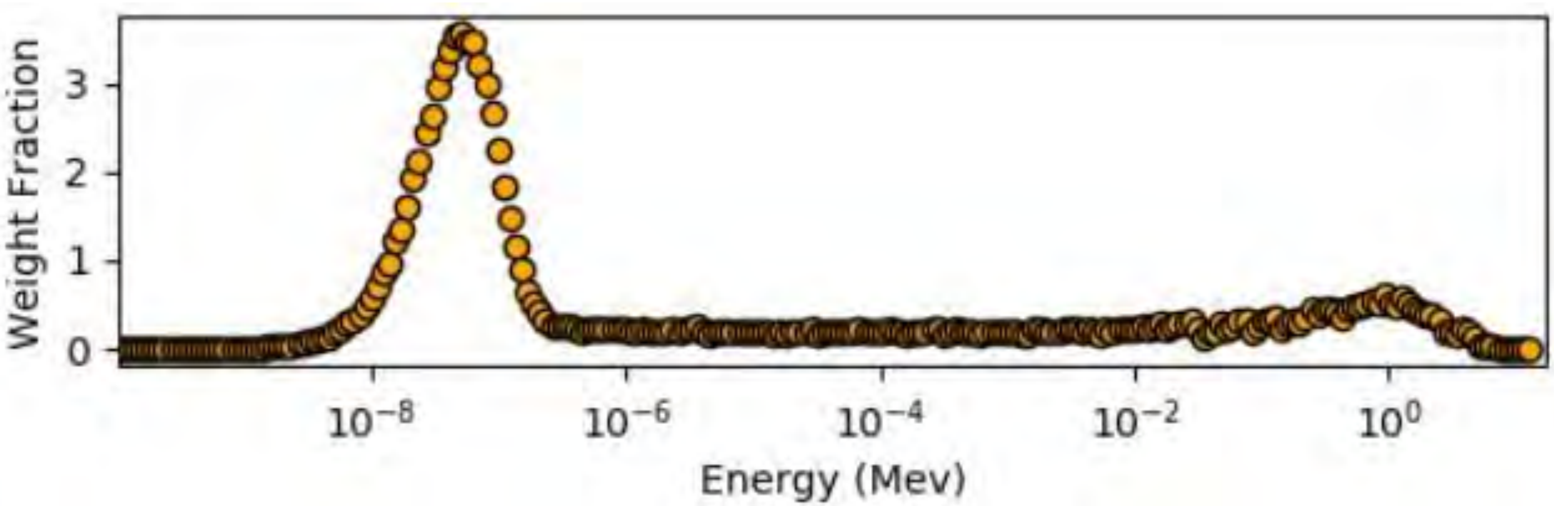

Figure 6. Particle weight fraction of neutron energy bins at the beamline exit of MINER.

Unstructured Mesh Example

- 1.5 x 1 x 0.2 cm zirconium coupon
- 1D diffusion of Hydrogen along the 1.5 cm axis (x direction)
    - 445 wppm at highest down to 85 wppm
    - 10 bins of hydrogen content created
- Implemented into the radiography MCNP model as a mesh
    - The x-coordinate of the nodes assign the hydrogen content to each cell in the mesh
    - Simulates a hydrogen content gradient

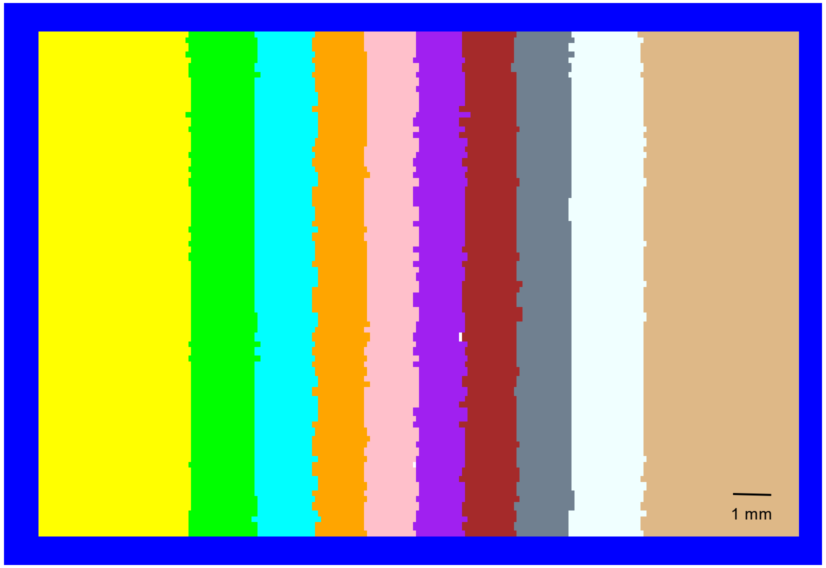

Figure 7. Unstructured mesh representation of a 1D hydrogen gradient in a zirconium coupon.

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Creating an "image"

- Instead of using a FMESH tally to measure neutron flux behind the sample, insert geometry representing a neutron scintillator
  - Scintillator captures neutrons using lithium-6 and converts the resulting alpha particle into photons that are captured by a camera
  - Use the ptrac file again to record the locations of the neutron capture events in the scintillator
    - Assuming the photon creation radius is small relative to the pixel size and the lens is ideal, the capture location corresponds to where the photons reach the camera senser
    - Bin the scintillator into "pixels" and count the number of neutron captures occurring in each pixel

Figure 8. Grayscale representation of the number of neutron captures in a scintillator behind the object in Figure 7.

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Analyzing the "image"

- Using Beer's law, and an open beam image, each pixel can be turned into a measure of the attenuation coefficient
- Because the sample had 1D diffusion, each column of pixels corresponds to a specific hydrogen content in the sample
  - oCombine all the columns corresponding a specific hydrogen content and plot the attenuation coefficient as a function of the hydrogen content

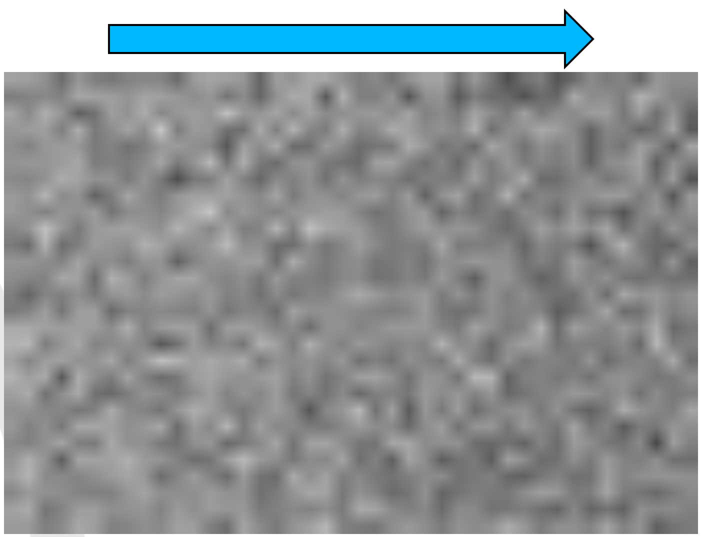

Figure 9. Grayscale representation of the neutron attenuation coefficient of the object in Figure 7, the arrow represents increasing hydrogen concentration.

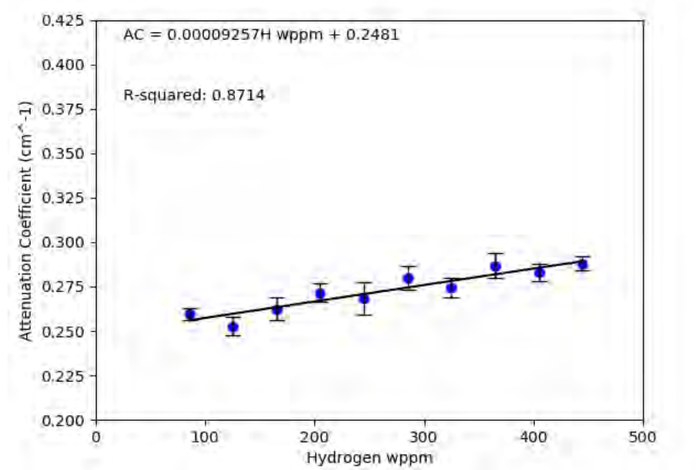

Figure 10. Plot of the attenuation coefficient as a function of the hydrogen content of the object in Figure 7

Questions?

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