Development of Hydrogen Transport Models for High Temperature Metal Hydride Moderators

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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 will develop validated computational methods to predict the short- and long-term reactor performance impacts from thermally-driven hydrogen transport in zirconium- and yttrium-hydrides

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

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

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]

$$J_H = -D_H * \left(\underset{\text{Fickian}}{\boxed{\nabla \cdot C_{ss}}} + \underset{\text{Soret Effect}}{\boxed{\frac{Q^* C_{ss}}{RT^2} * \nabla \cdot T}} + \underset{\text{Stress Cross-Effect}}{\boxed{\frac{V^* C_{ss}}{RT} * \nabla \cdot \sigma_h}} \right)$$

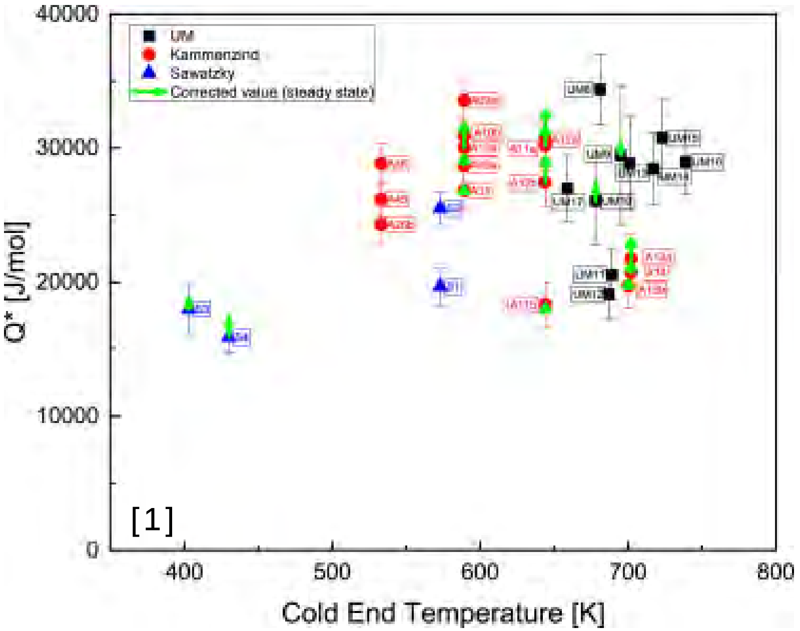
- Interstitial solutes will diffuse towards areas of:[1]
  - Low solid solution concentration.
  - Low temperature.
  - High tensile hydrostatic stress.
- Accurate measurements of D_H, Q^*, V^* are required for accurate modeling.

[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.

Heat of Transport

- The heat of transport, Q^*, is typically considered a constant.[1-3]
- Measurements exhibit scatter.
- Lack of agreement in measurement may indicate variable dependence.
  - Q^* can be reasonably hypothesized to have concentration, temperature dependence.[4]
- Heat of transport can be measured by creating a Soret gradient.[4]
  - Imposed temperature gradient creates a concentration gradient, given by:

$$\frac{\partial \ln(C_H)}{\partial x} = -\frac{Q^*}{RT^2}\frac{\partial T}{\partial x}$$



Comparison of measurements of hydrogen's heat of transport

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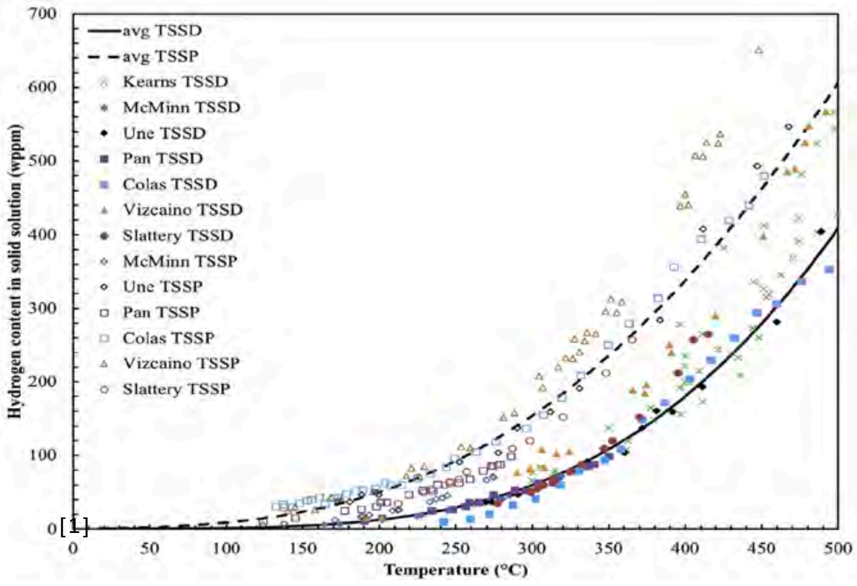
[1] S. Kang, P.-H. Huang, V. Petrov, A. Manera, T. Ahn, B. Kammenzind, and A. T. Motta, "Determination of the hydrogen heat of transport in zircaloy-4," *Journal of Nuclear Materials*, vol. 573, p. 154122, 2023.
[2] B. F. Kammenzind, D. G. Franklin, H. R. Peters, and W. J. Duffin, "Hydrogen Pickup and redistribution in alpha-annealed zircaloy-4," *Zirconium in the Nuclear Industry: Eleventh International Symposium*, pp. 338–370, 1996.
[3] J.T. Merlino, Master of Engineering Paper in Nuclear Engineering, Experiments in Hydrogen Distribution in Thermal Gradients Calculated Using Bison, The Pennsylvania State University, 2019.
[4] J. R. Manning, *Diffusion Kinetics for atoms in Crystals*, 2nd ed. Ann Arbor, MI: UMI, Books on demand, 1994.

Solubility of Hydrogen

- Hydrogen in solid solution exhibits a solid solubility hysteresis.[2]
  - The Terminal Solid Solubility (TSS) of hydrogen in zirconium is path dependent.
    - The two TSSs are referred to as TSSp and TSSd, respectively.
  - Both TSSp and TSSd follow Arrhenius-type dependencies.[3]



Hydrogen solubility in zirconium alloys

- The current theory for the hysteresis mechanism is plastic deformation.[3]
  - γ and δ-hydrides occupy 12.3% and 17.2% more volume than α-zirconium.
- The free energy of formation of a δ-phase hydride is given as:[3]

$$\Delta G_{\alpha-\delta} = \Delta G_{\alpha-\delta}^{\mathrm{chem}} + \Delta G_{\alpha-\delta}^{\mathrm{elastic}} + \Delta G_{\alpha-\delta}^{\mathrm{plastic}} + \Delta G_{\alpha-\delta}^{\mathrm{surface}}$$

- The sum elastic and plastic strain terms is termed the accommodation energy.[3]
  - Differences in accommodation energy on precipitation and dissolution cause hysteresis.[3]

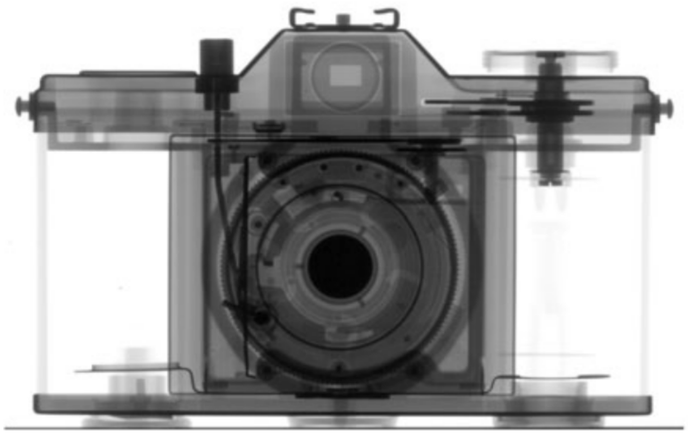
[1] A. T. Motta, L. Capolungo, L.-Q. Chen, M. N. Cinbiz, M. R. Daymond, D. A. Koss, E. Lacroix, G. Pastore, P.-C. A. Simon, M. R. Tonks, B. D. Wirth, and M. A. Zikry, "Hydrogen in zirconium alloys: A Review," *Journal of Nuclear Materials*, vol. 518, pp. 440–460, 2019.

[2] A. McMinn, E.C., Darby, and J.S., Schofield, "The terminal solid solubility of hydrogen in zirconium alloys." *ASTM special technical publication*, vol. 1354, pp. 173-195, 2000.

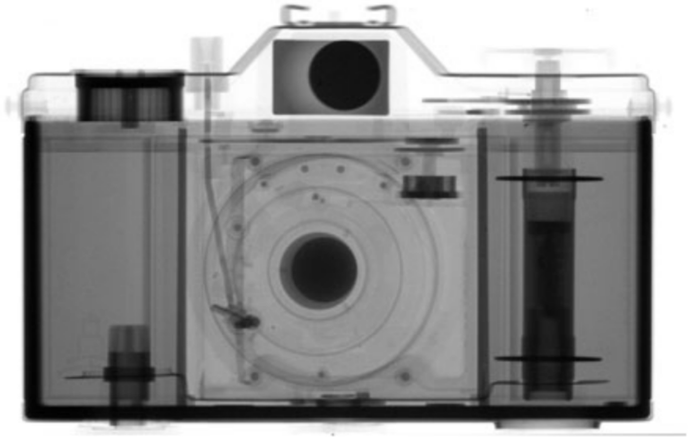
[3] B. W. Leitch and S.-Q. Shi, "Accommodation energy of formation and dissolution for a misfitting precipitate in an elastic - plastic matrix," *Modelling and Simulation in Materials Science and Engineering*, vol. 4, no. 3, pp. 281–292, 1996

Neutron Radiography

- Neutron radiography can 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 us NR techniques



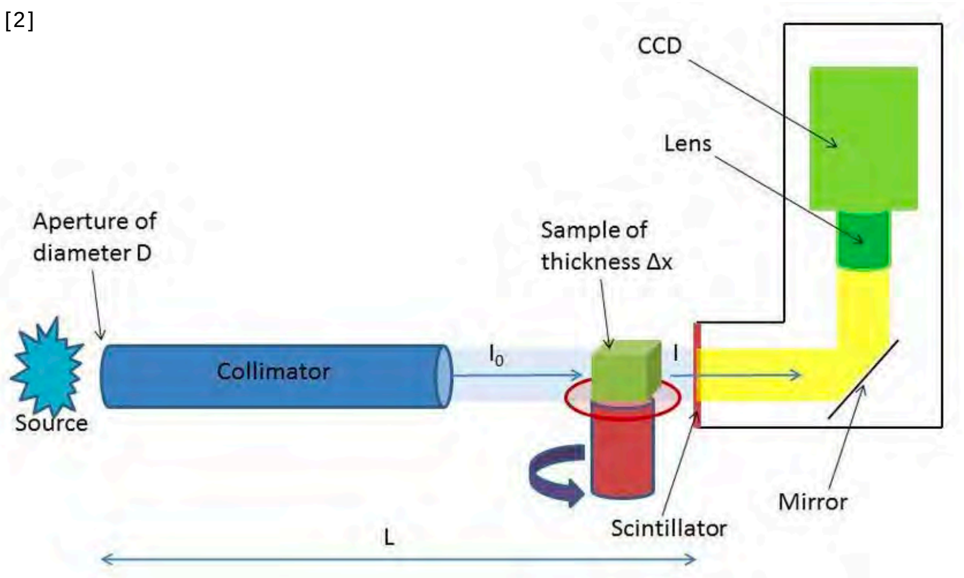
X-Ray Radiograph



Neutron Radiograph

Neutron Radiography Setup

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



[2]

Schematic of 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.

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[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

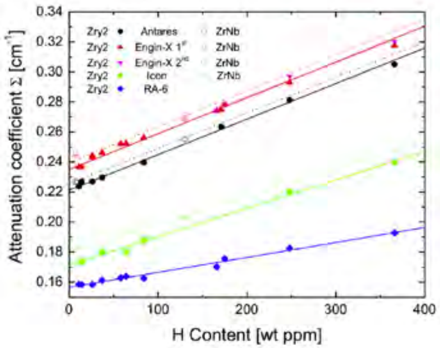
- The intensity of the incident neutron beam is used to characterize the transmission of the neutron beam through the sample.[1]
  - The transmission is used determine the total interaction cross section.

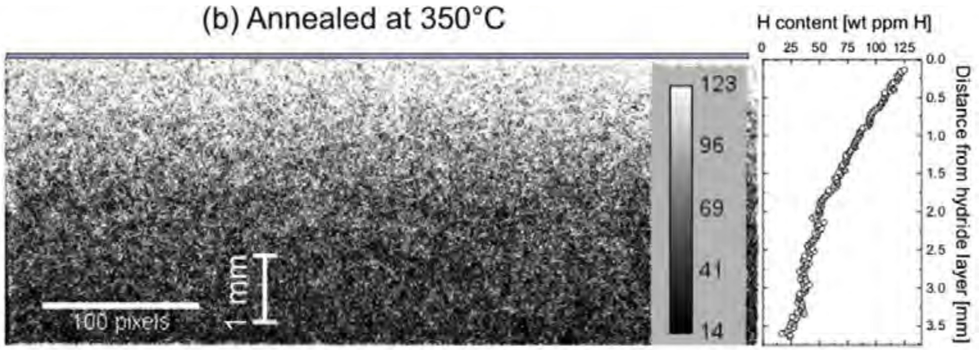
$$T(x,y) = \frac{I_s(x,y) - B(x,y)}{I_0(x,y) - B(x,y)} = e^{-\Sigma_t(x,y,E)z(x,y)}$$

- 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.





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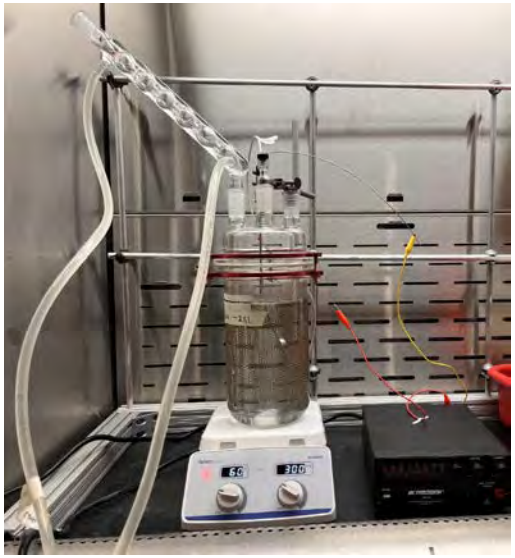
[1] N. L. Buitrago, J. R. Santisteban, A. Tartaglione, J. Marín, L. Barrow, M. R. Daymond, M. Schulz, M. Grosse, A. Tremsin, E. Lehmann, A. Kaestner, J. Kelleher, and S. Kabra, “Determination of very low concentrations of hydrogen in zirconium alloys by neutron imaging,” Journal of Nuclear Materials,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

Work in Progress – Specimen Preparation

- Developing the capabilities to produce hydrided imaging specimens at Mines, with parallel elemental analysis
- Electrolytic process
  - Primarily supports NR project (with funding from that source)
  - Calibration is ongoing
- Thermal (Sievert's) process
  - Existing vacuum tube furnace retrofit with Ar-5H atmosphere
  - To be completed over the summer



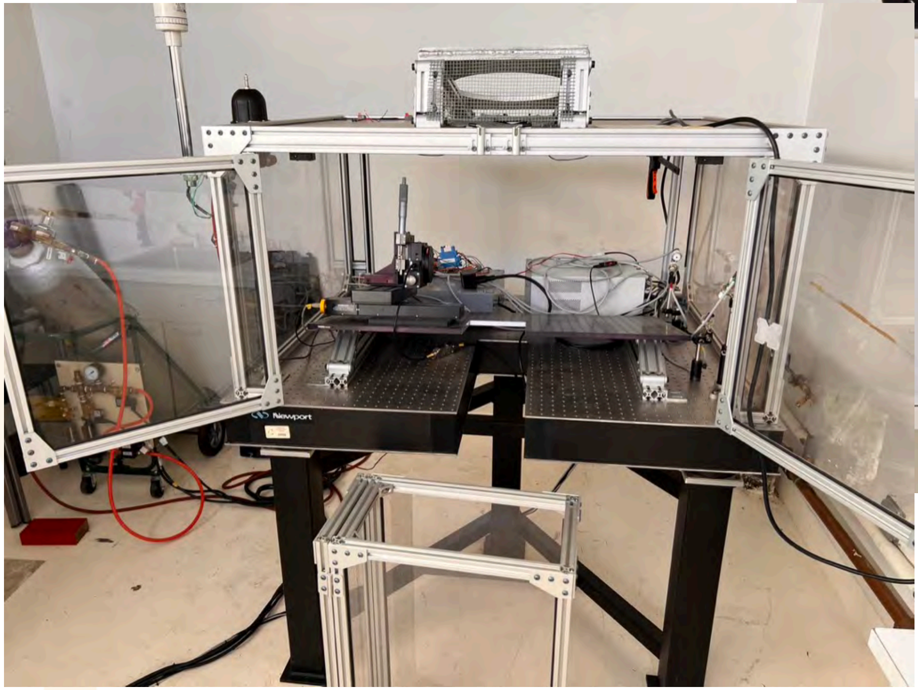
Electrolytic Hydriding Cell

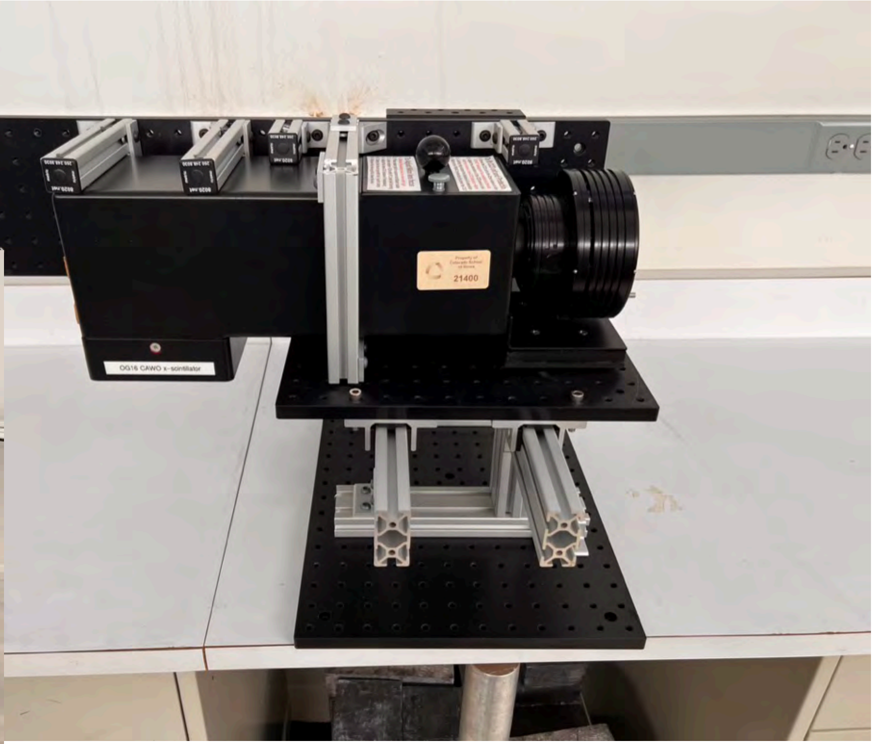
Cleaned (L) and Hydrided (R) Samples

Work in Progress - Radiography

- Reinstalling neutron beamline capabilities at the GSTR
  - Post-COVID recovery
  - Foil and film (transfer) radiography
  - Digital radiography

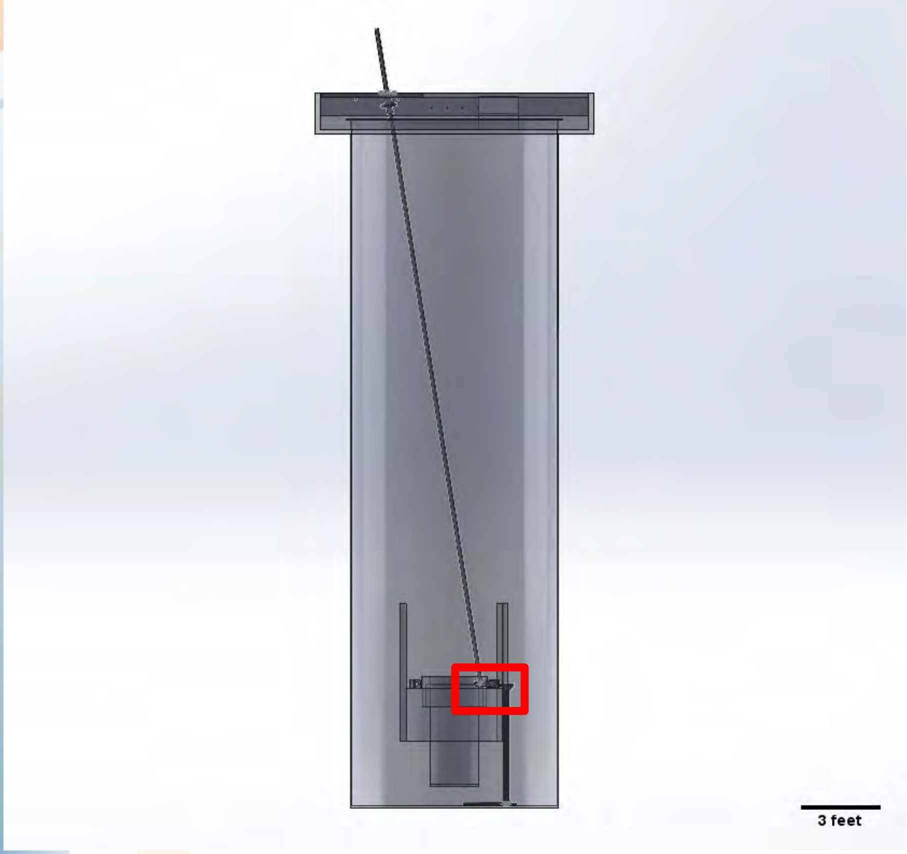
Experimental Station

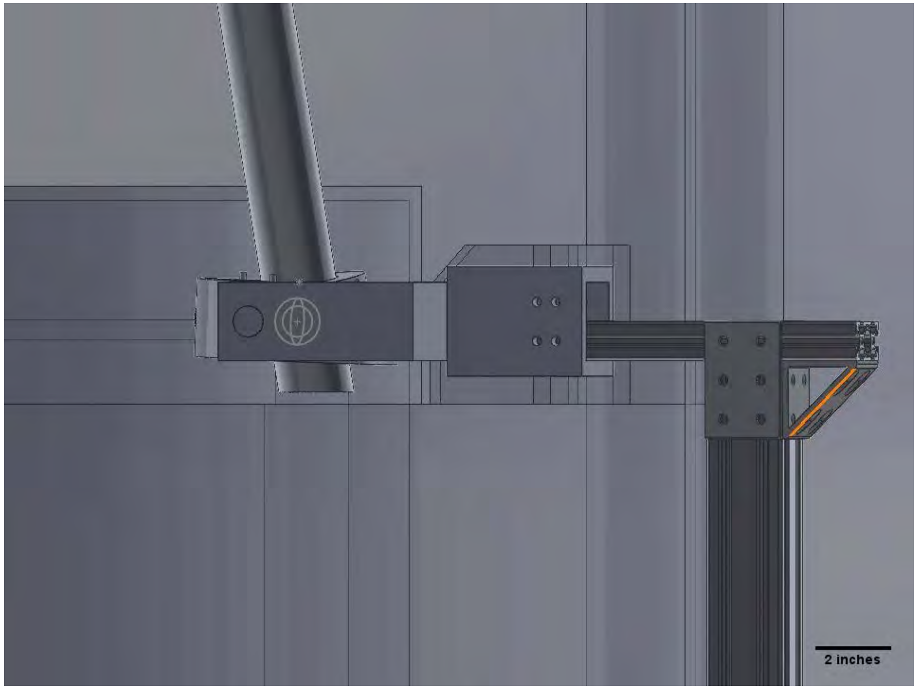




Scintillator-based Digital Imager

MiNeR Beamline



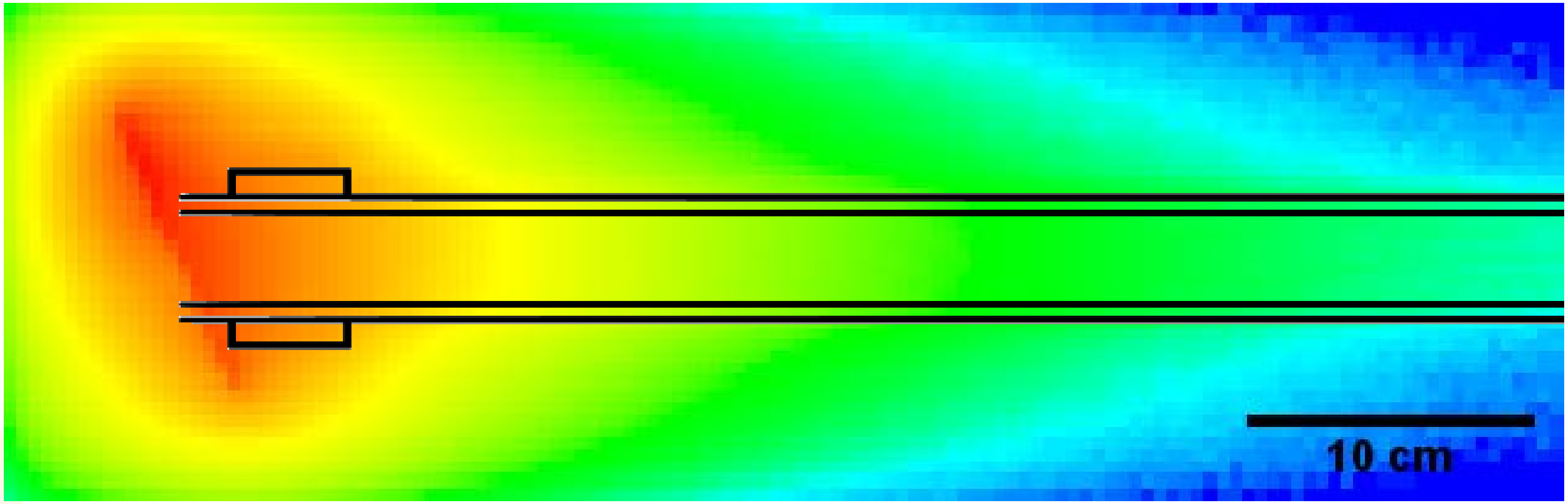


Neutron Radiograph Simulation

- 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

GSTR Neutron Source

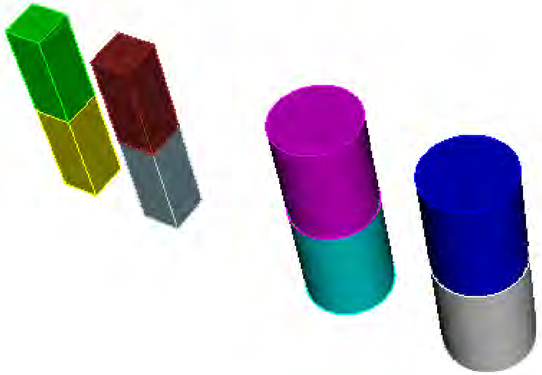
- The current MiNeR beamline is angled
- Using an existing model of the GSTR core, we can create a representative neutron source
  - Can estimate angular and energy distributions



Incorporating Unstructured Mesh Geometry

- Represent objects for simulated radiographs as unstructured meshes, and import the mesh into the imaging model
    - Will also allow better incorporation of gradients into the model
    - Can predict images based on FEA results

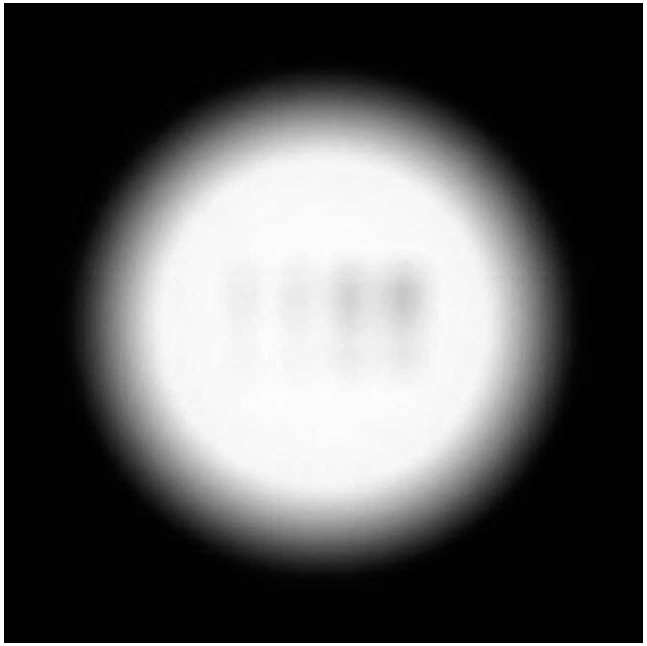
- First test, shown to the right, to begin testing imaging simulation capabilities
    - Two rectangular prisms
        - Zirconium
            - Metal (bottom)
            - 40 at% Hydride
        - Yttrium
            - Metal (bottom)
            - 40 at% Hydride
    - Two cylinders
        - Zirconium
            - Metal (bottom)
            - 40 at% Hydride
        - Yttrium
            - Metal (bottom)
            - 40 at% Hydride



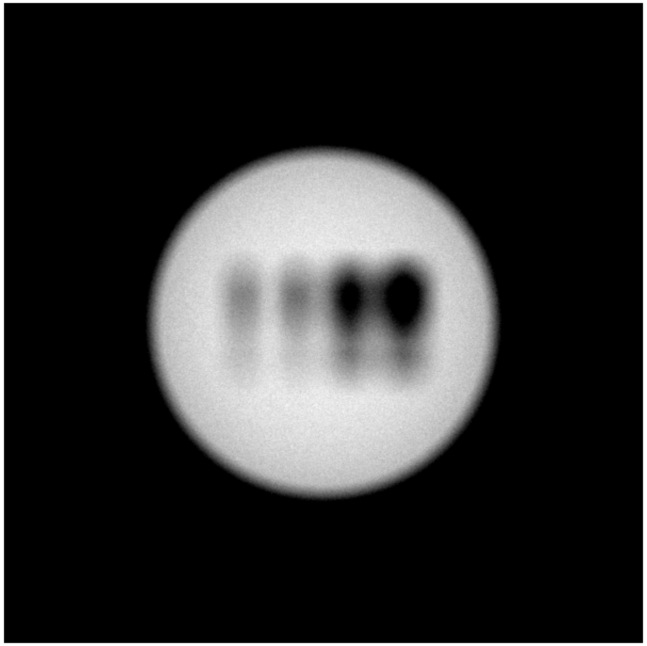
2 mm

Simulated Radiographs

- Simulated Radiograph of the 4 objects shown before
- Array of 600x600 pixels



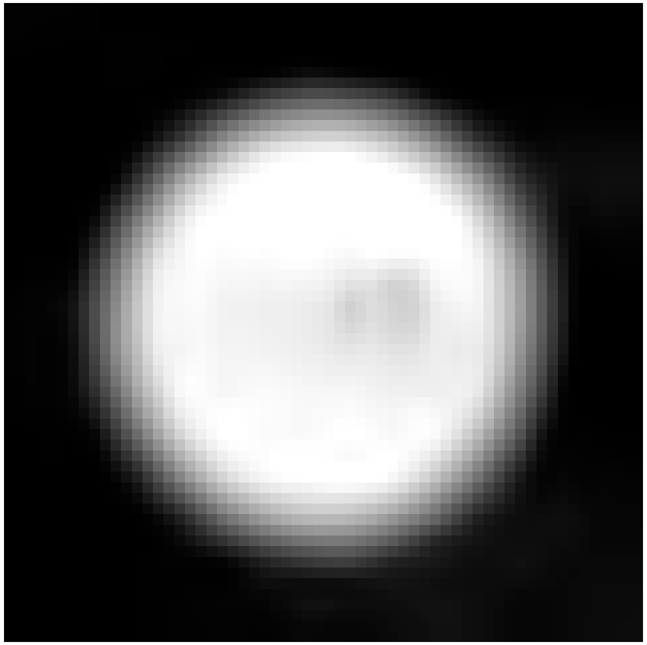
All neutrons reaching imaging plane displayed as a 16-bit image



Showing a smaller range of gray values reveals additional details

Benefit of Simulated Radiography

- Simulated Radiograph of the 4 objects shown before
- 60x60 “pixel” grid, to examine scattering



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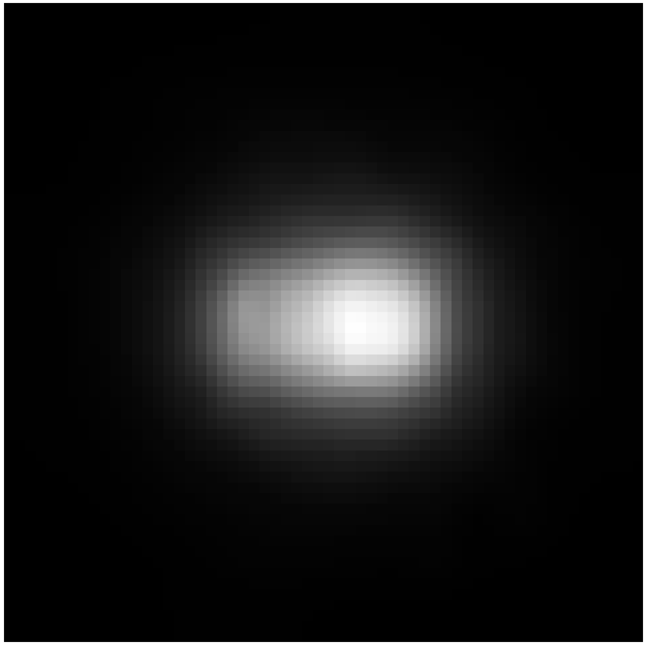
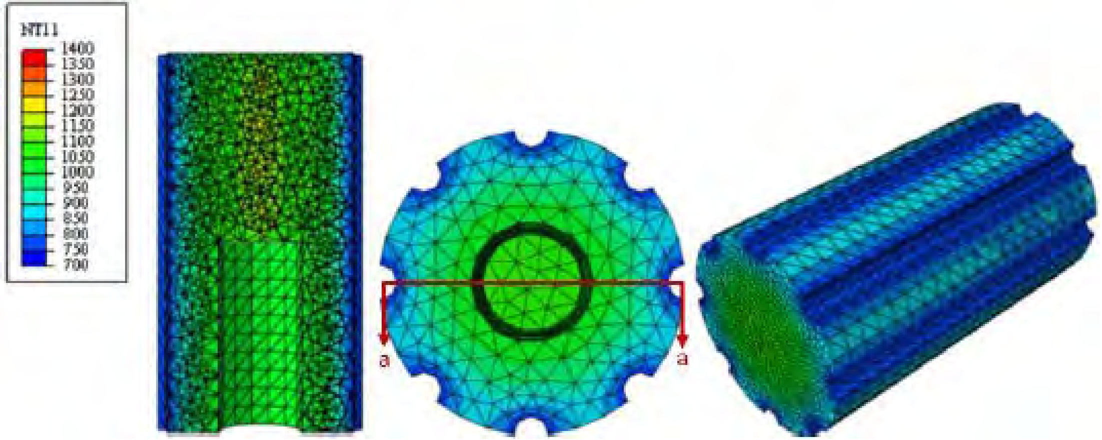
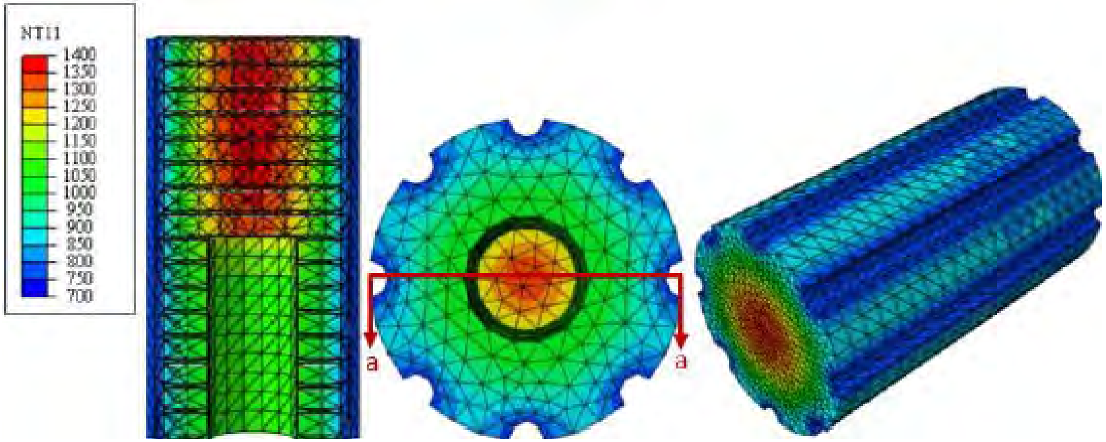


Neutrons that entered one of the 4 objects and had at least 1 collision

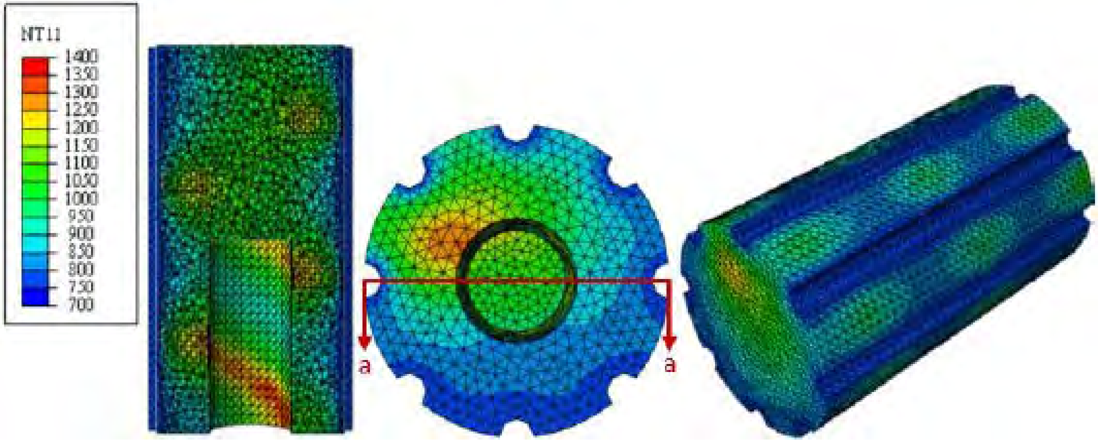
ABAQUS Models for HALEU Fueled Kilopower Reactors



ABAQUS Results for Spherical Spherical at a Total Power of 15 kW_{th}



ABAQUS Results for Disc Fuel at a Total Power of 15 kW_{th}



ABAQUS Results for Helical Fuel at a Total Power of 15 kW_{th}

Questions?