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Title: NEUTRON RADIOGRAPHY AT LANSCE: Interrogation and Characterization of Materials for Next Generation Nuclear Reactor Designs

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NEUTRON RADIOGRAPHY AT LANSCE: Interrogation and Characterization of Materials for Next Generation Nuclear Reactor Designs

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Twelfth International Conference on Methods and Applications of Radioanalytical Chemistry
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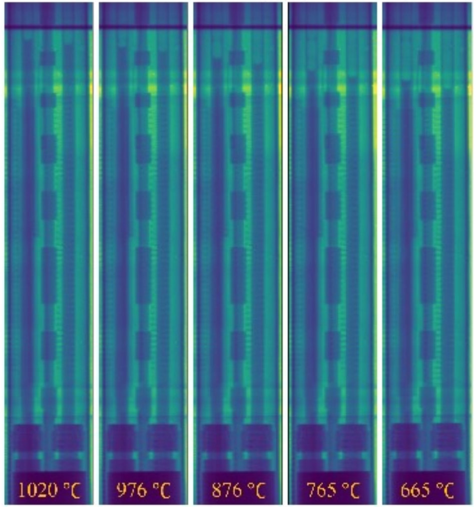
Thermophysical measurements of molten salts

Measuring densities in chloride based molten salts to evaluate MSR designs and performance



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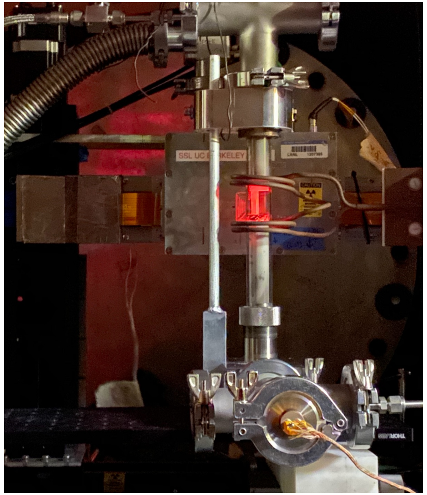
Mapping hydrogen in YH_{2-x} for potential use as high density moderators in microreactors



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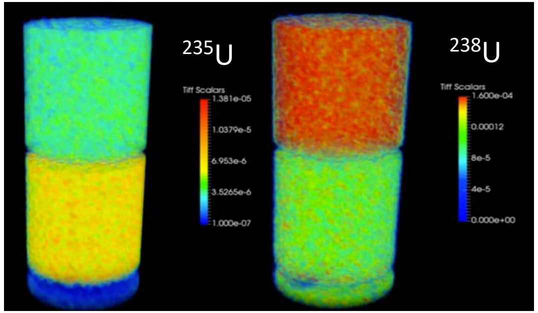
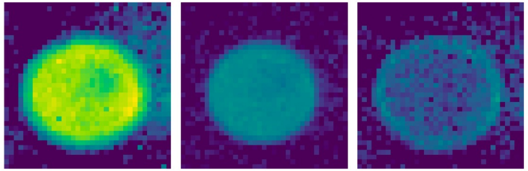
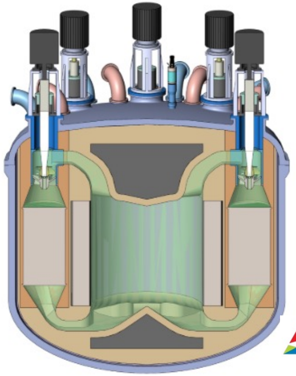




Traditional — Imaging Techniques — Advanced

Remote Density Measurements of Molten Salts

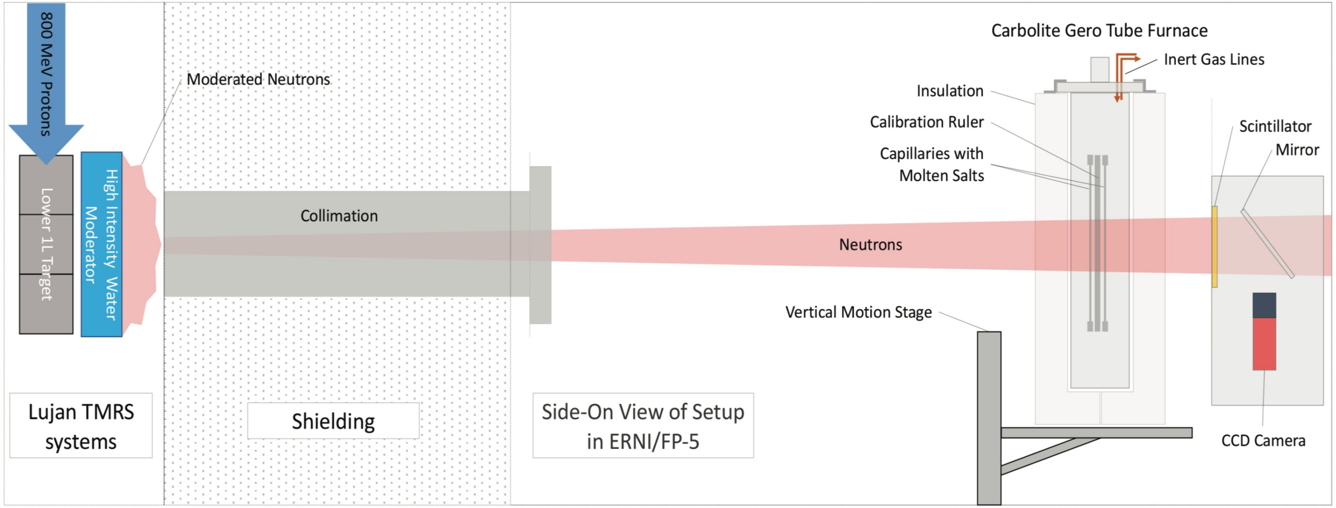


Next Gen. molten chloride fast reactors

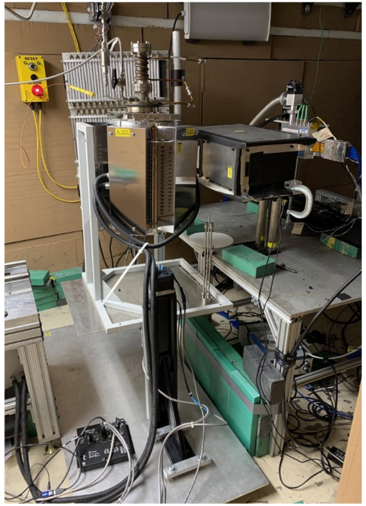
TerraPower
Southern Company

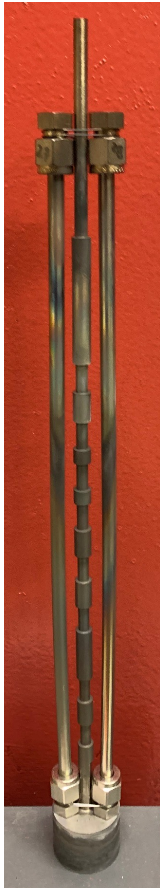
Many chloride-based salts currently lack much needed thermophysical data at the relevant temperatures to confidently assess and evaluate new reactor designs.

Measuring density as a function of temperature and chemical composition is critical to predicting salt behavior and performance.

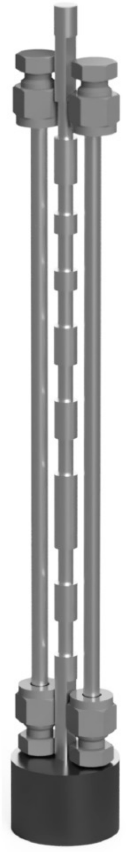


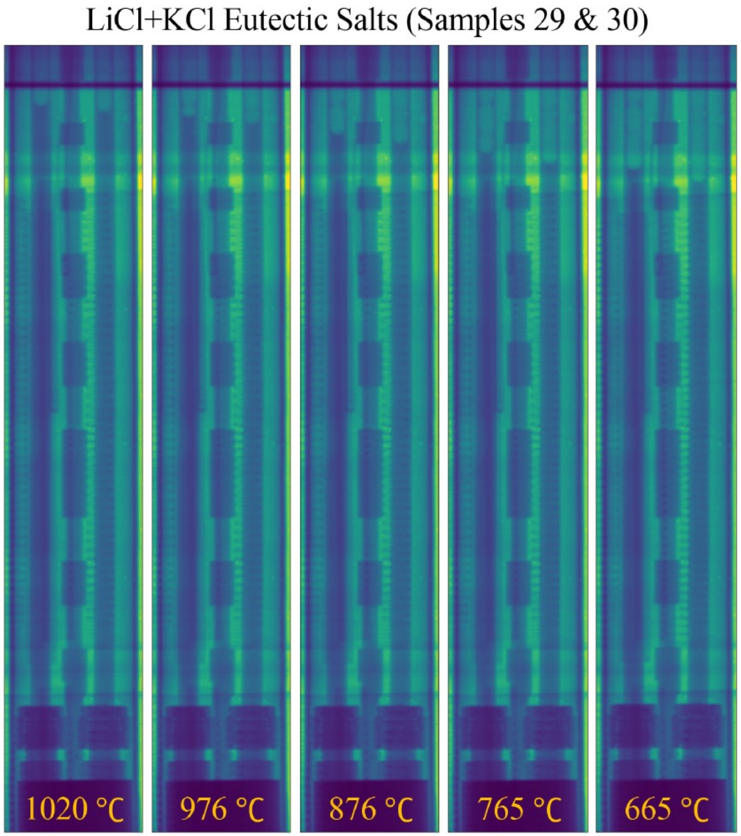
Density measurements via neutron radiography on FP5 (Oct 2020)

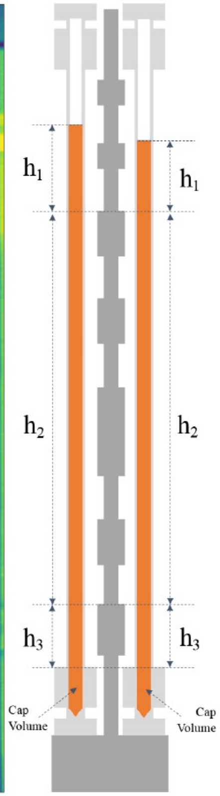




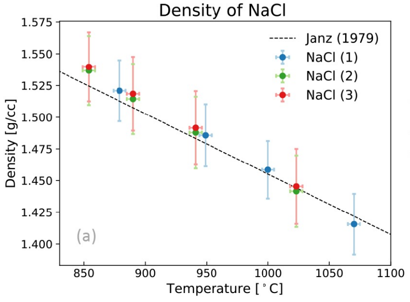
Some Density Measurement Results with Molten Salts

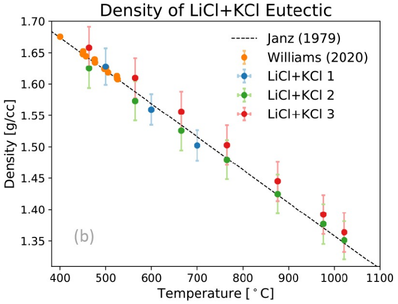


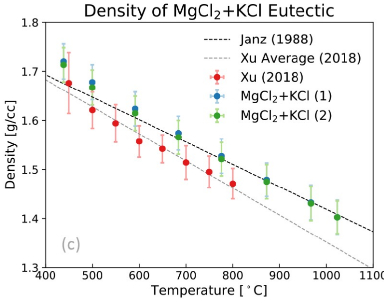


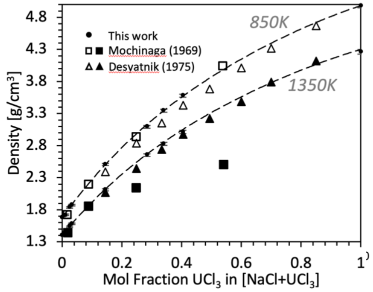


Final uncertainties in density ~ 1-2%









Advantages over other methods:

- Compact designs = very little sample material
- Measure multiple samples at a time.
- Able to see the material through out the measurement.

"Remote Density Measurements of Molten Salts via Neutron Radiography" **A. M. Long et. al.**, *J. Imaging* **2021**

"Thermophysical Properties of Liquid Chlorides from 600 – 1600K' **S. S. Parler et. al.**, *J. Molecular Liquids* **2021**

Looking Forward with Neutron Imaging of Molten Salts

Part of NE-GAIN with TerraPower to measure $PuCl_3$- based salts using neutron radiography at LANSCE during 2022 RC.

GAIN Gateway for Accelerated Innovation in Nuclear

TerraPower™ A Nuclear Innovation Company

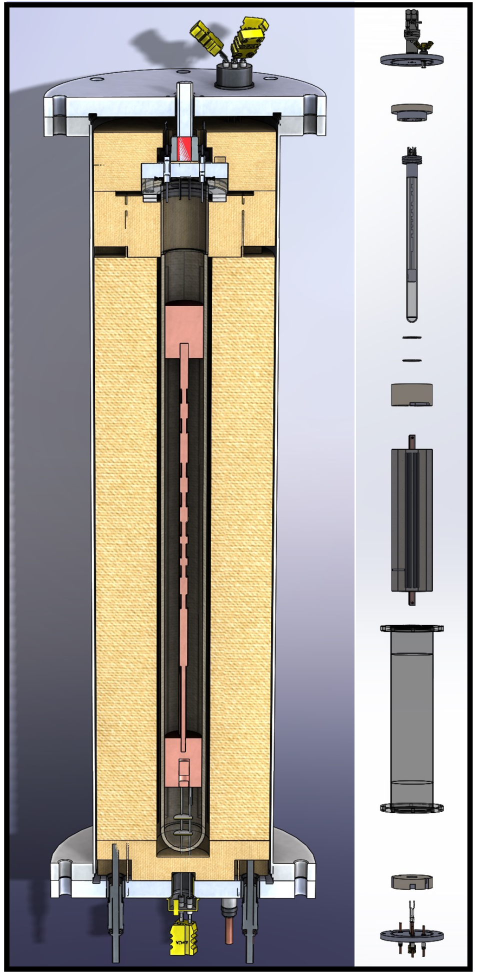
Building a custom compact tube furnace…

- Improved overall accuracy with better imaging geometry.
- Allow for sample rotation for nCT of meniscus shape.
- House high hazardous samples with multiple levels of containment.

Parts have arrived and testing will be underway soon!

Additional planned neutron imaging measurements on molten salts:

- Exploring falling sphere measurements with neutron and x-ray radiography to characterize viscosity (with compact furnace)
- Preform Energy Resolved Neutron Imaging (ERNI) measurements on salts to map out actinide distributions at temperatures.



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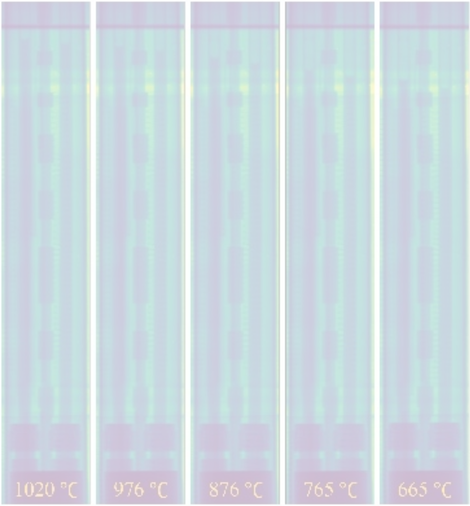

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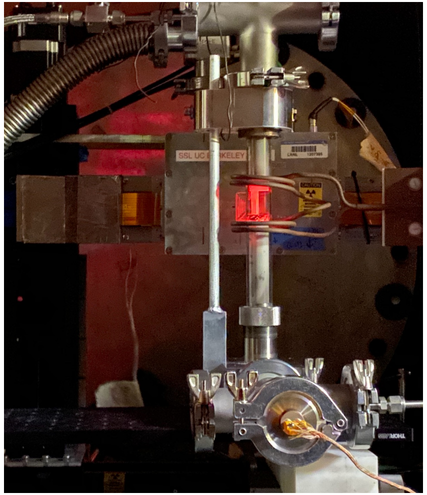
Mapping hydrogen in YH_{2-x} for potential use as high density moderators in microreactors



Energy resolved neutron imaging on nuclear fuels

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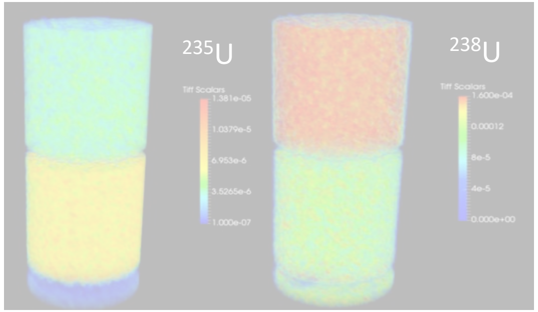
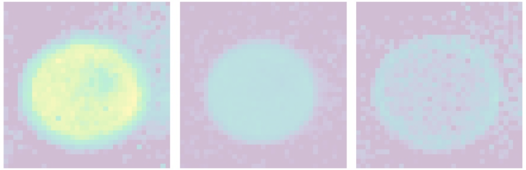
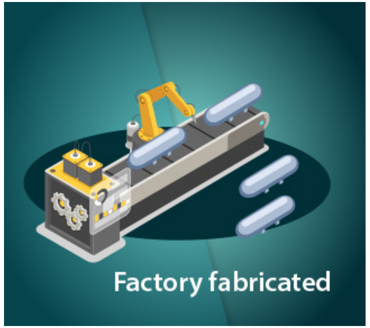


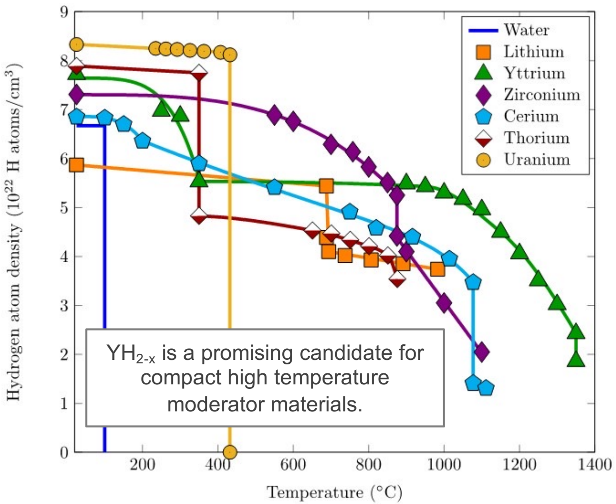


Traditional ← Imaging Techniques → Advanced

Why are We Interested in Measuring H-diffusion in YH?

Class of very small modular reactors targeted for non-conventional nuclear markets (remote locations), that can be factory fabricated, readily deployable, and relatively safe and reliable with small operations footprint.

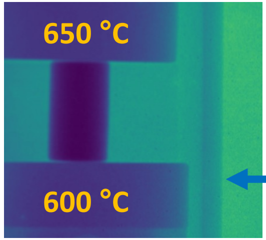




GOAL: Measure H-Diffusion coefficients at various conditions (stoichiometry, temperatures, phase fractions) in YH samples.

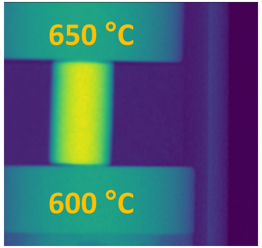
- Build up similar neutron radiography capabilities to quantify and map out H-concentrations.
- Build a sample environment capable of inducing high temperature gradients.

YH_{2-x} samples fabricated @ LANL



Measure / Determine / Calibrate/Model

$$T(x,y,E_n) = exp(-z_H(x,y)\ \Sigma(x,y,E_n))$$



Use models to extract diffusion flux

$$J_{net} = J_{\nabla C} + J_{\nabla T} + J_{\nabla \sigma}$$

$J_{\nabla C}$ Fickian (Concentration)

$J_{\nabla T}$ Soret (Temperature)

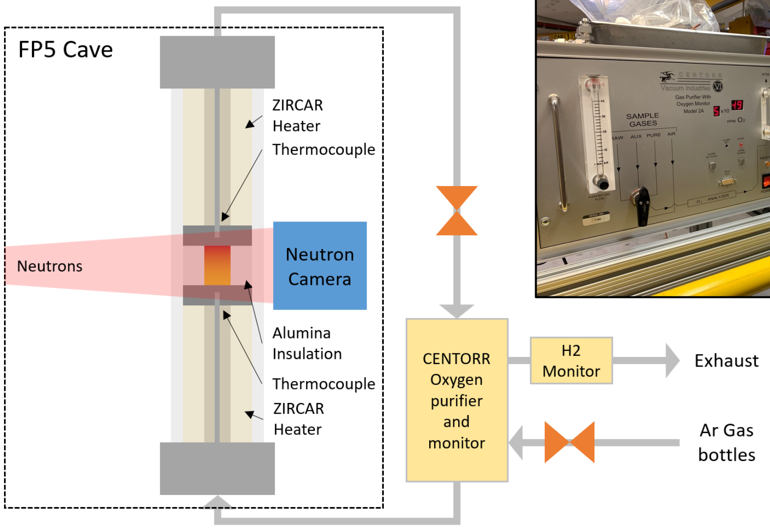
$J_{\nabla \sigma}$ Stress (Temperature)

Most Recent H-Diffusion Measurements on FP5 (2021 LANSCE RC)

- Single heating measurement took about 24 hours.
- Selected sample $YH_{1.9}$ "178-1"
- Used Compact Dual-Zone (CDZ) furnace (developed in house)
- Use ATIK 490ex CCD camera coupled with 200um thick ZnS screen
- Selected temperature profiles that would increase from 300°C to 900°C in 50°C increments.
- Included extra auxiliary diagnostics
  - Oxygen analyzer on furnace outlet
  - H2 analyzer on exhaust

Compact Dual-Zone Furnace

- Using two independent heating elements to induce temperature gradients.
- Max temperatures of ~1100°C
- Sample, insulation, and heater all fit within a 1" diameter tube (reducing geometric image blur).
- Using purified Ar gas flow from bottom to top.
- During testing, we were able to induce 200°C (700-900) gradient across a steel surrogate sample.



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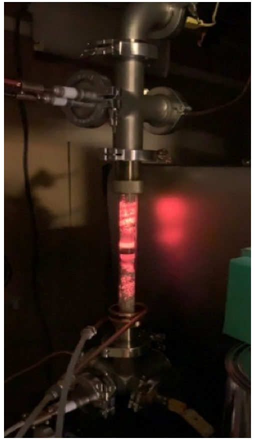
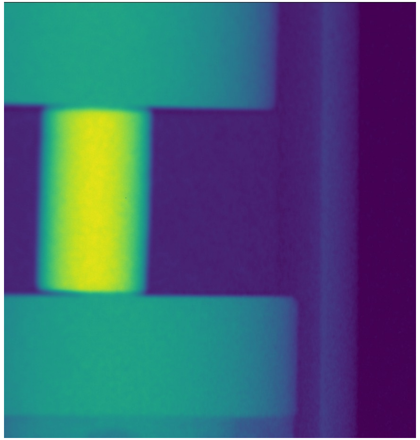
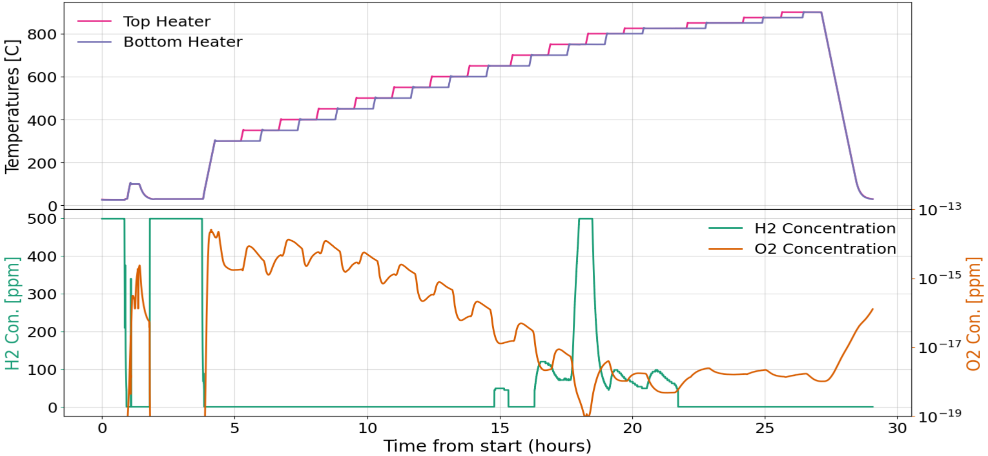
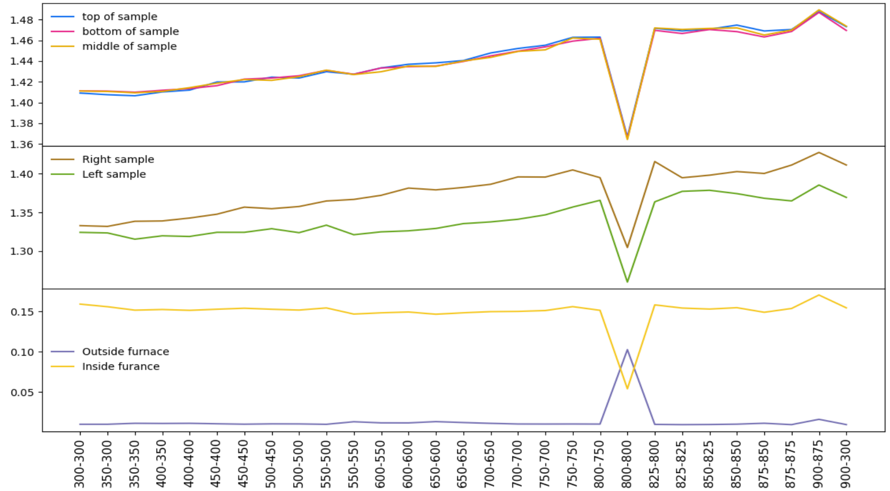








Initial observations and thoughts…

- Attenuation appear flat across sample and increasing as a function of temperature. This might be due to oxidation of sample during heating.
- Oxygen monitor shows clear decrease in O_2 concentration as temps increase.
- The H2 monitor observes H leaving system starting at 650°C steps and ending around 800°C.
- Possibly look at attenuation ratio images, but difficult as sample moves due to thermal expansion of platens.

Pursuing a more Sophisticated Analysis

Working with the Initiative for Scientific Imaging (ISI) group at LANL to build better analysis techniques to overcome deficits in measurements and build confidence in results.

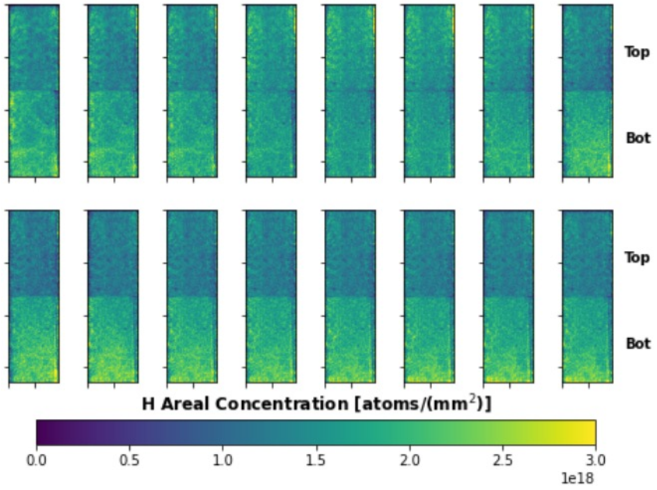
Sometimes there maybe no calibration samples that can be used or they are less than ideal…

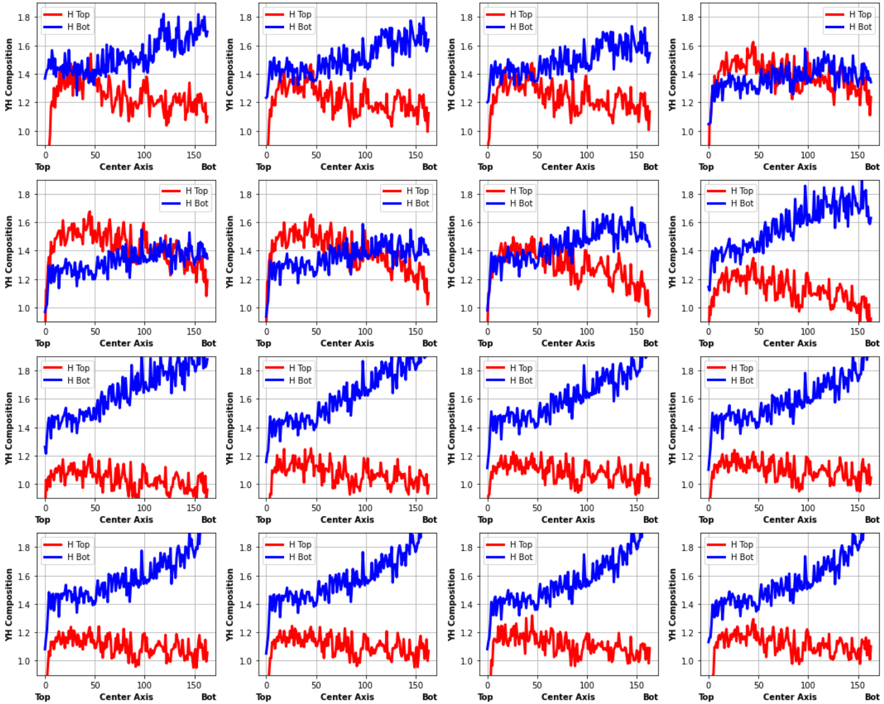
Using solvers along with constraints, several unknowns can be determined.

$$h_{ij} H_{cs} + y Y_{cs} = p_{ij}$$

$$\frac{\sum_{ij} h_{ij}}{yN} = C$$

- h_{ij}: Hydrogen areal concentration (spatially heterogeneous) [atoms/(mm^2)]
- y: Yttrium areal concentration (spatially constant) [atoms / (mm^2)]
- H_{cs}: Hydrogen cross-section for neutrons (known constant) [82.3 barns $\equiv$ 82.3×10^{-22} mm^2 / atom]
- Y_{cs}: Yttrium cross-section for neutrons (known constant) [9 barns $\equiv$ 9×10^{-22} mm^2 / atom]
- p_{ij}: Pixel intensity (known measurement)
- C: Composition (known and constant)
- N: number of pixels per temperature measurement (known)
- M: number of temperature measurements (known)





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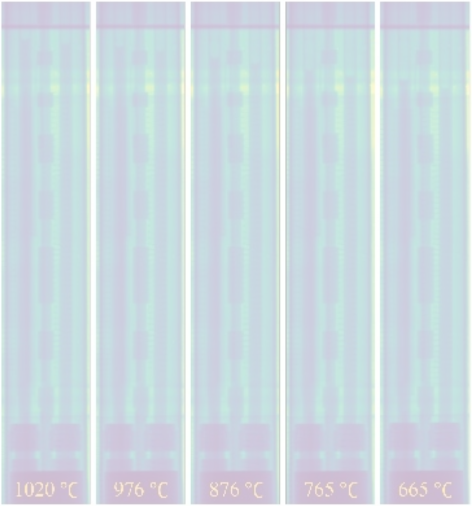

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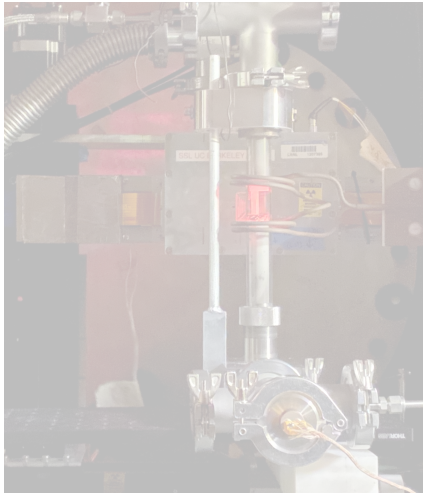

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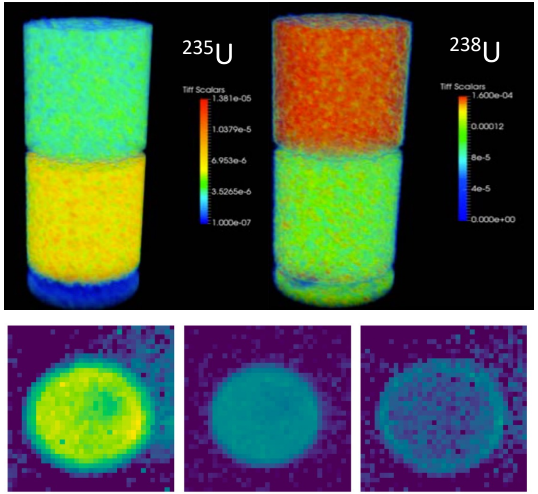

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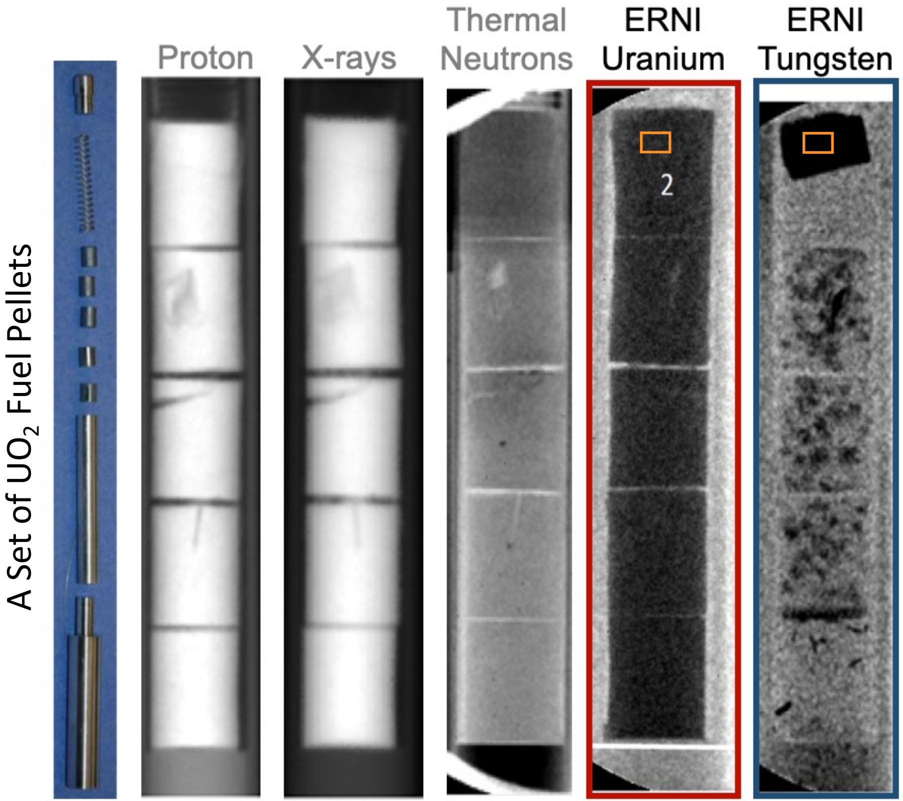
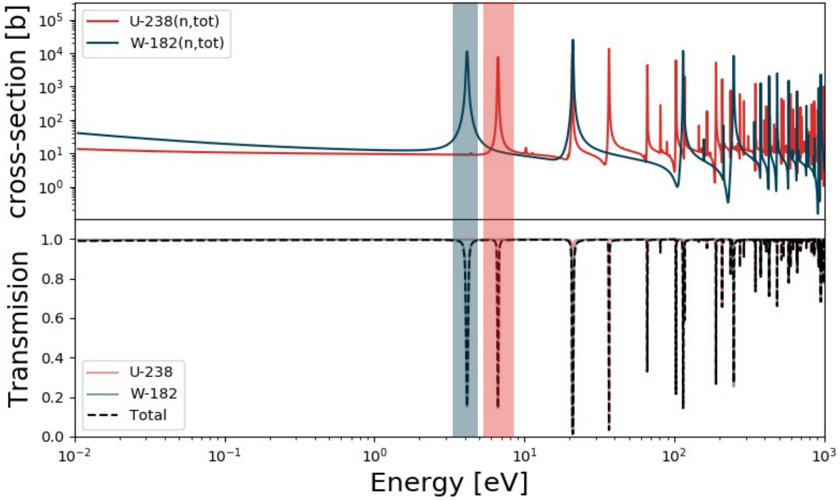
Energy Resolved Neutron Imaging (ERNI)



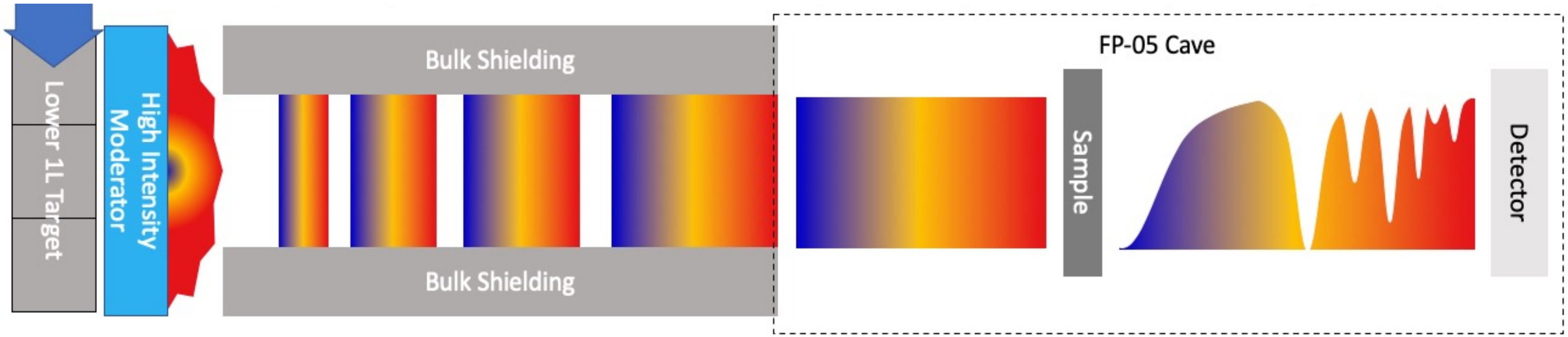
- Advanced neutron radiography technique
- Neutrons have complex cross-sections
- If neutron energy is known, transmissions can be further resolved based on incoming neutron time of flight

Each pixel has additional energy information in the form of a transmission spectra

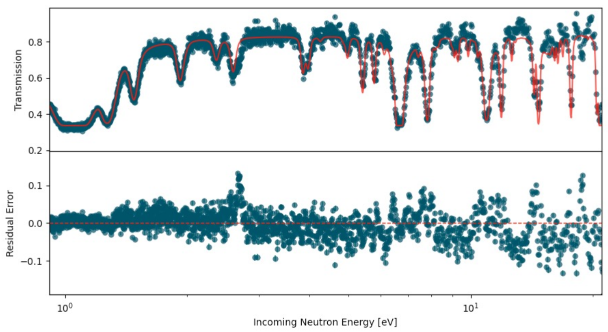


Morris *et. al.* Review of Scientific Instruments 84 (2013)
Tremsin *et. al.* Neutron News 24 (2013)

ERNI on FP5 @ LANSCE

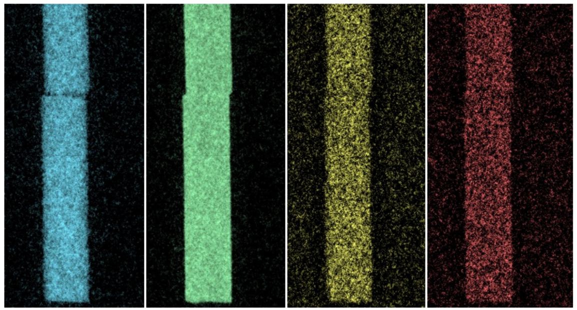


Fitting transmission spectra from each pixel

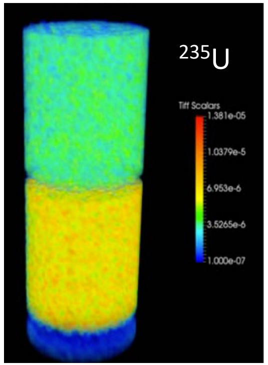


Obtain 2D heat maps of each isotope

Isotopic areal densities in g/cm^2



Perform CT reconstruction on each isotope

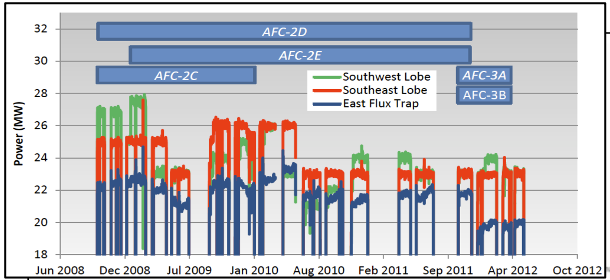


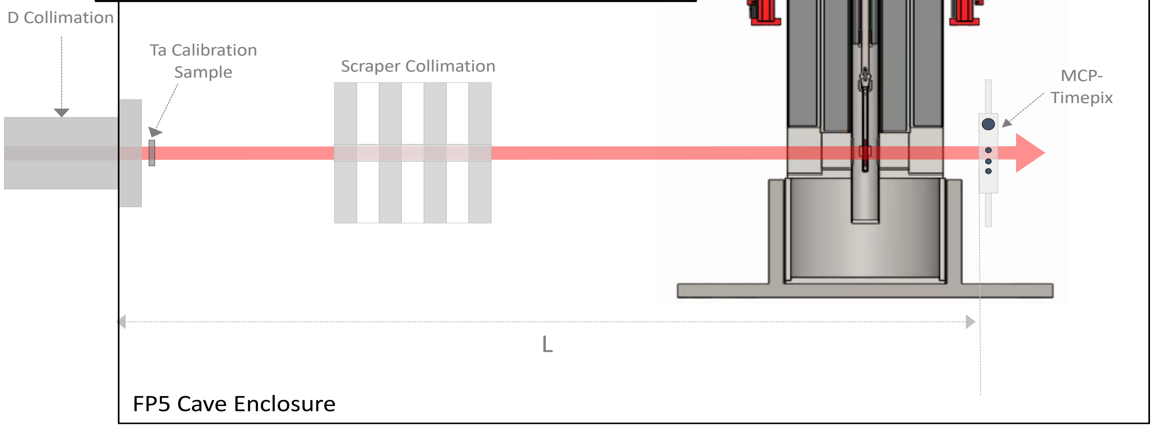
ERNI-PIE on irradiated fuels at FP5

Post irradiation examination is critical to understanding fuel performance.

ERNI-PIE can be extremely useful:

- **Remote** ➞ Able to measure highly radioactive samples.
- **Non-destructive** ➞ Can guide further, more direct, PIE measurements.
- **Probe Internal structure** ➞ Help identify any "off-normal" conditions due to irradiation condition.
- **Spatial isotopic info** ➞ Spatially measurements of fuel and fission product distributions can help solve certain questions surrounding FCCI, fuel redistribution, and uniformity of burnup.

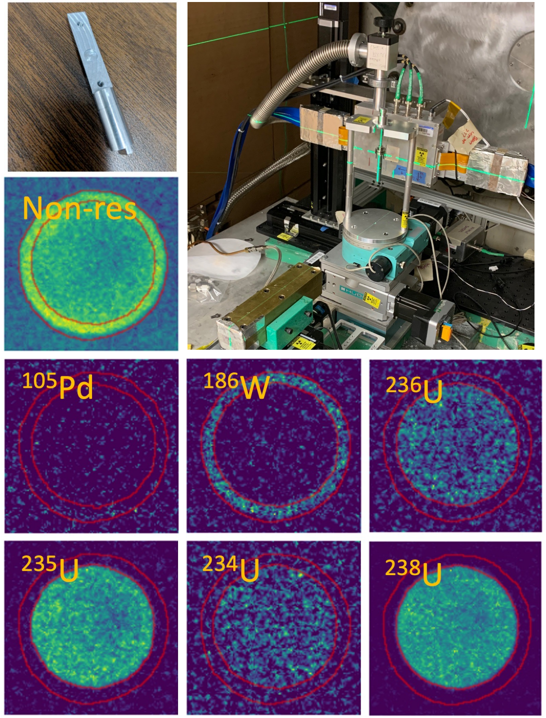




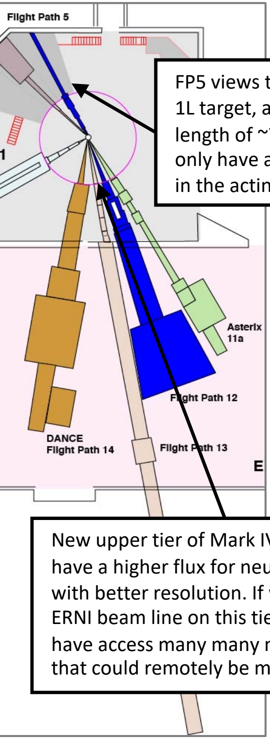
Preliminary ERNI measurements on U-1Pd-10Zr sample irradiated at the ATR @ INL

~1.5 mm thick disk prepared from AFC-3A-R5A

Dose rate on contact: ~3R/hr

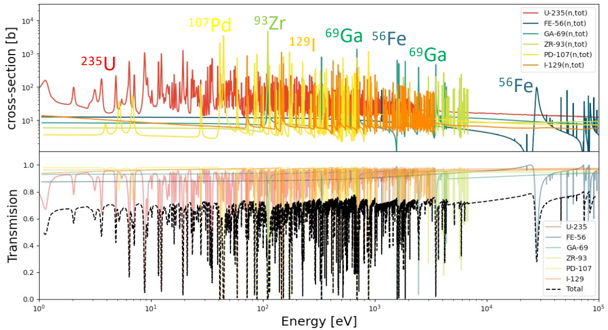


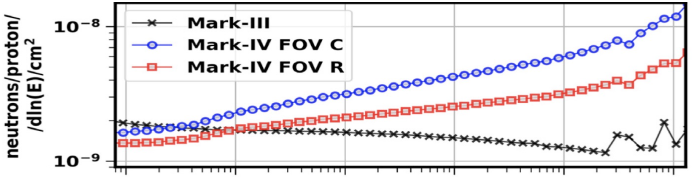
Potential Futures at LANSCE: ERNI on FP12

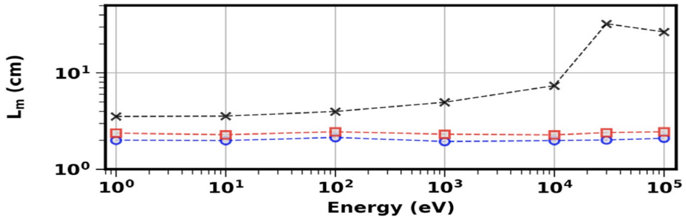


FP5 views the lower tier of the 1L target, and with a flight path length of ~7-11 m we effectively only have access to resonances in the actinide regime.

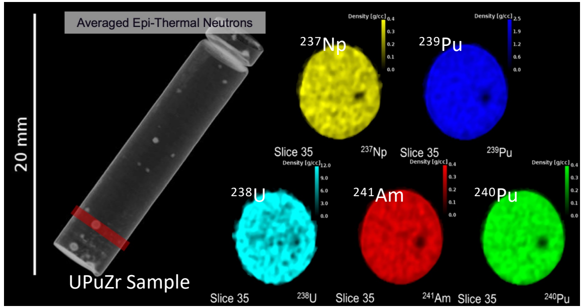
New upper tier of Mark IV target will have a higher flux for neutrons > 10 eV with better resolution. If we invest in a ERNI beam line on this tier we would have access many many more isotopes that could remotely be mapped out!





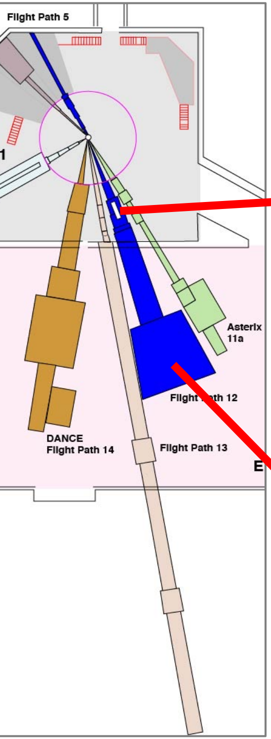


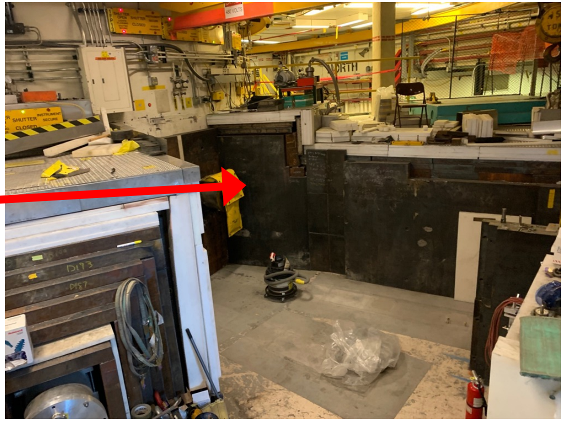
New beam line on upper tier target could map out fission products and isotopes down to the Fe group. For example, fresh and irradiated fuels, along with materials like U-Nb and Pu-Ga, can be non-destructively characterized.



Additionally, FP12 has a much bigger footprint and will allow for more exotic sample environments (most importantly the SHERMAN tank and irradiated fuels).

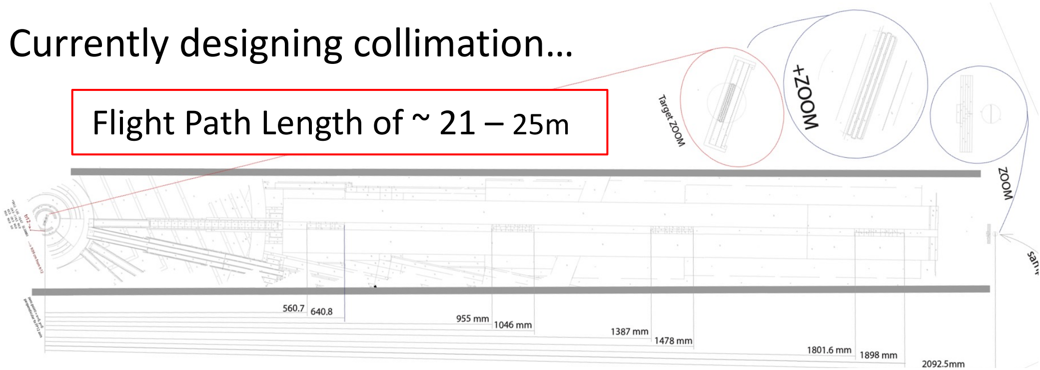
Progress of ERNI on FP12

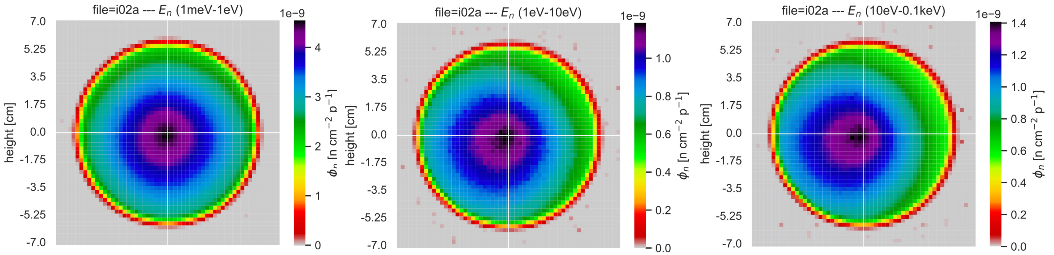


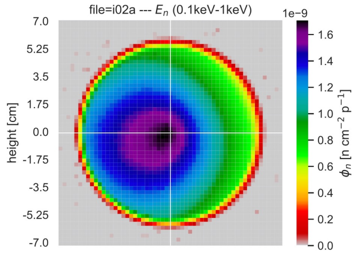


Currently designing collimation…

Flight Path Length of ~ 21 – 25m







- 10cm diameter beam spot creates extremely high backgrounds at beam dump (~ 85 mrem/hr)!
- Until more shielding is added we will run with ~2.5cm diameter beam spot.

Will hopefully design and install new FP12 collimation and be able to run test late in the 2022 LANSCE run cycle.

LDRD
LABORATORY DIRECTED RESEARCH & DEVELOPMENT

GAIN Gateway for Accelerated Innovation in Nuclear

MRP Microreactor Program

AFC Advanced Fuels Campaign

TRIAD National Security, LLC

Thank you!

Density measurements of molten salts:

Team: A.M. Long, S.S. Parker, M. Monreal, M. Jackson, D.T. Carver, and S.C. Vogel.

Funding: LANL LDRD Office (20210113DR & 20190650DI)
DOE-NE GAIN (2021 NE Voucher)

Hydrogen measurements in YH_{2-x}:

Team: A.M. Long, H. Trellue, A. Shivprasad, E. Luther, D.T. Carver, and S.C. Vogel.

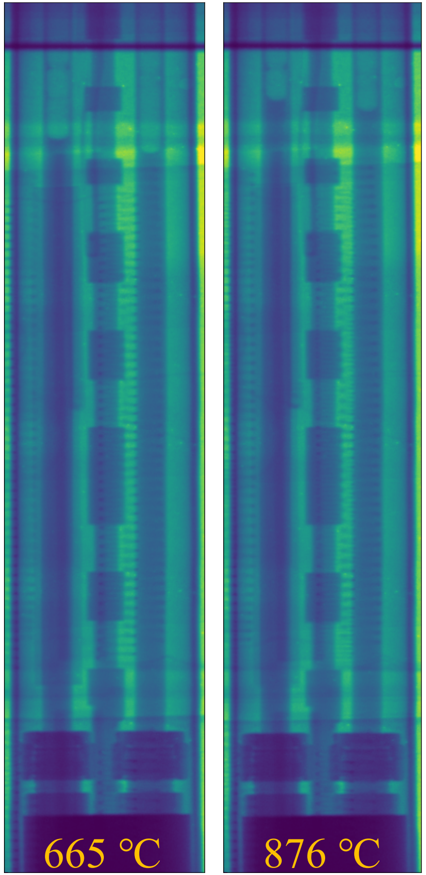
Funding: LANL LDRD Office (20190649DI)
DOE Microreactor Program

ERNI with fresh and irradiated fuels:

Team: S.C. Vogel, A.M. Long, K.J. McClellan, J.R. Angell, T. Balke, L. Capriotti, A.E. Craft, J. Harp, P. Hosemann, J. Lin, E.J. Larson, D.C. Schaper, B. Wolhberg

Funding: LANL LDRD Office (20200061DR)
DOE-NE Advanced Fuels Campaign
Nuclear Science User Facilities

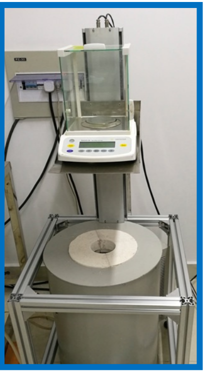
Why Neutron Radiography over other applications



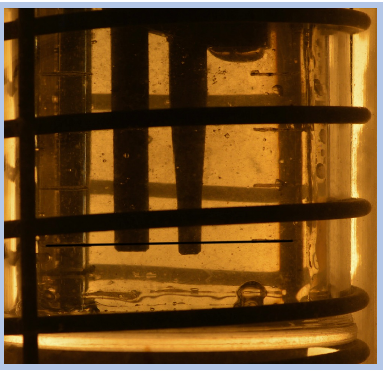
Developing measurements with neutron radiography adds an additional technique to compare to already established methods and opens up the ability to measure otherwise difficult samples

Pycnometer[1]

Archimedes method[2]



Bubbler System[3]



Neutron radiography can be used to measure heights (2D) and volumes (3D) of well know quantities of salts at relative temperatures to determine densities.

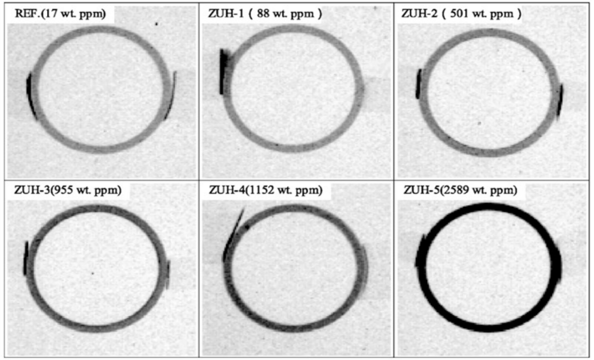
Some Advantages…

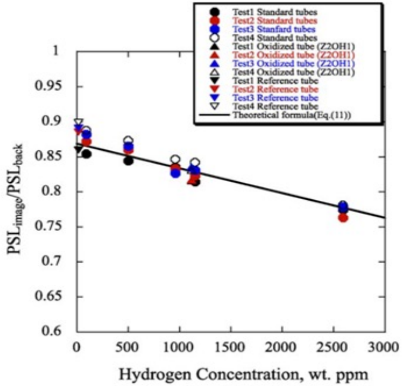
- You have eyes on the sample the whole time (watch out for bubbles!).
- Can have compact design that allows for less sample materials.
- Modular setup: multiple samples can be measured simultaneously and samples can be swapped quickly.
- Measurement times depend on furnace.
- Can measure same samples multiple times.
- Suitable hazardous materials ($PuCl_3$).
- Potential to extract a lot of additional information with more advanced neutron imaging techniques. Temps and actinide density can be measured in-situ with neutron resonances.

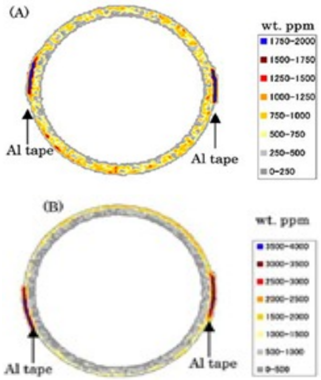
Previous Works looking at Hydrogen in Metal Hydrides

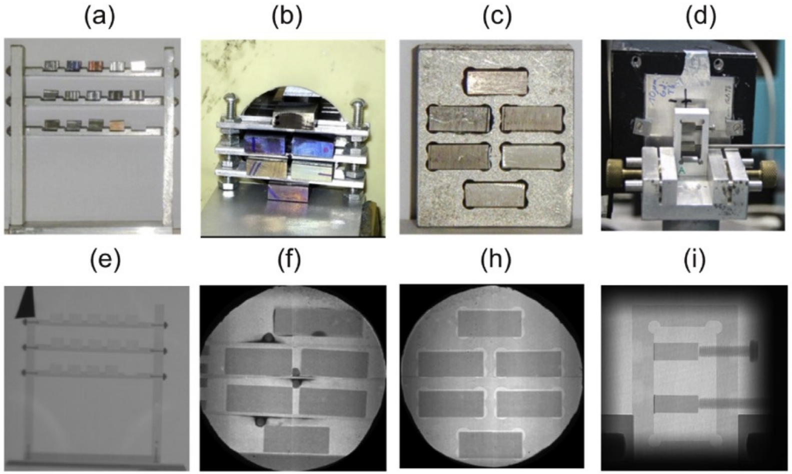
Measure — Determine — Calibrate/Model

$$T(x,y,E_n) = exp(-z_H(x,y)\ \Sigma(x,y,E_n))$$

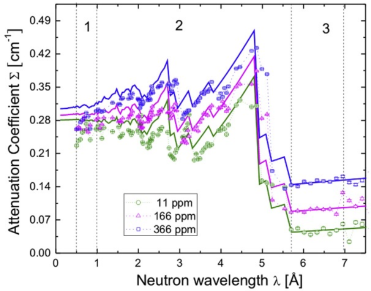


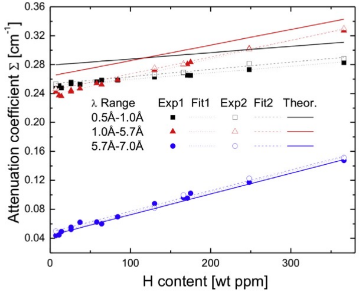






If neutron source is pulsed, then **Energy Resolved Neutron Imaging** (ERNI) measurements can be performed. ERNI measurements have been shown not need calibration sample and to resolve H-concentrations down to 5ppm %wt.

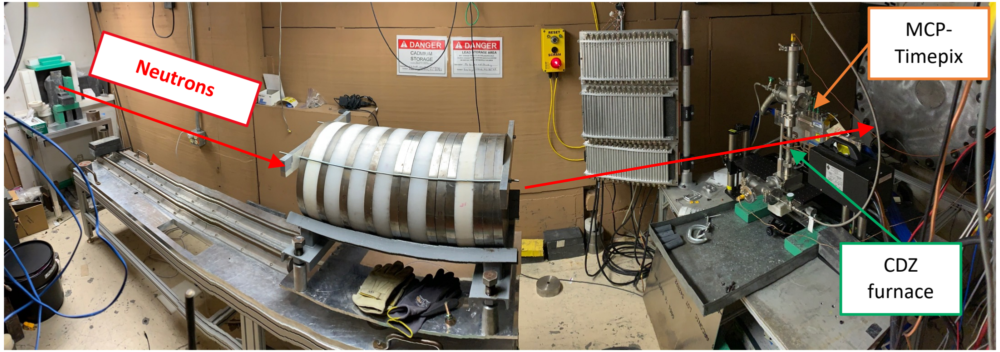


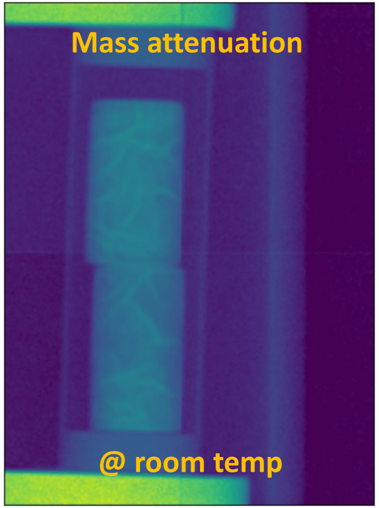


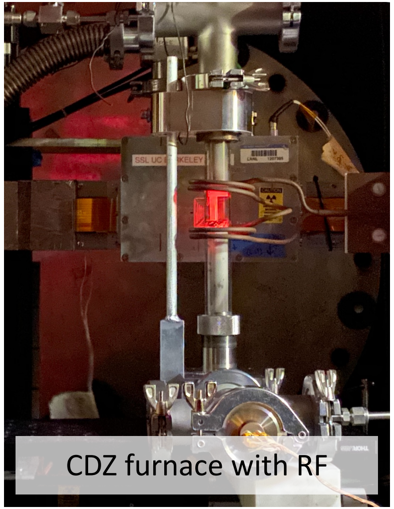
Hydrogen Mapping in YH_{2-x} on FP5 (2020 Measurements)

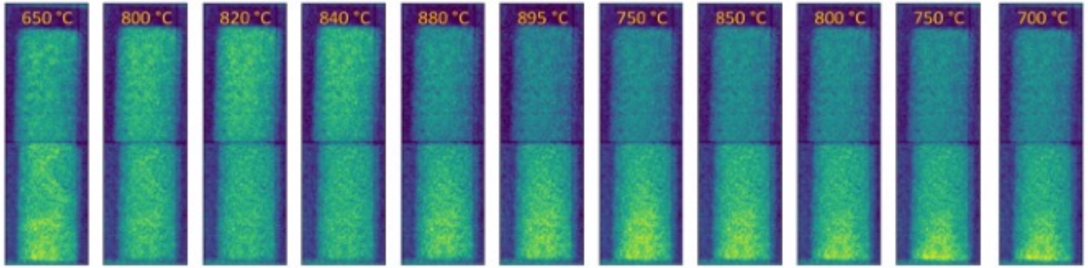
Main Goal: To observe H-distribution response to elevated temperatures and temperature gradients in YH_{2-x} samples.

- Commissioned new compact dual zone (CDZ) furnace.
- Performed mid Dec. 2020
- YH pellets in TZM cans ($YH_{1.15}$ - $YH_{1.9}$)
- Dec 2020 measurements used RF
- Also took nCT scans of several "as manufactured" YH and ZrH samples on FP5 and FP11
- Used MCP-TIMEPIX detector





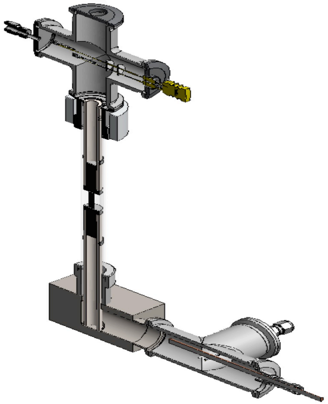


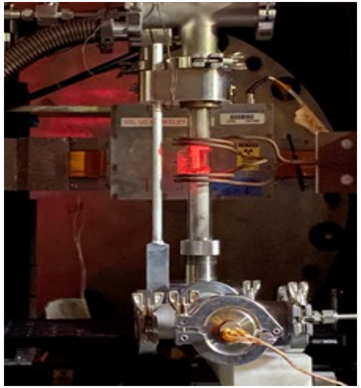


Clearly observed movement of Hydrogen, but ultimately did not have a good understanding of temperature profile within the samples…

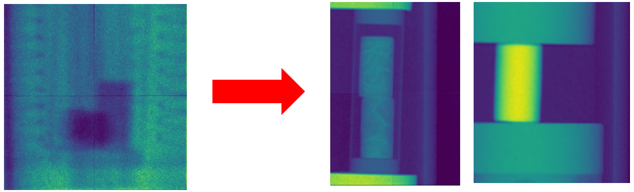
"Effects of Hydrogen Redistribution at High Temperatures in Yttrium Hydride Moderator Material", Trellue et. al., JOM Vol. 73, **2021**

1st Generation Compact Dual-Zone Furnace



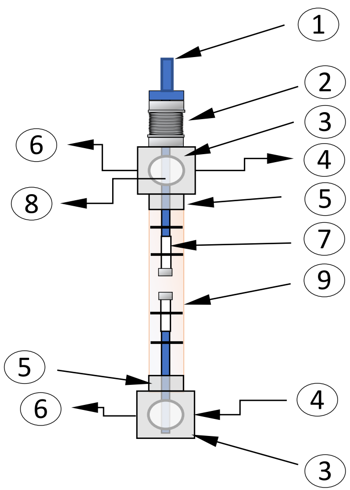


- Operated on FP5 in ‘20 and ‘21 run cycles.
- Suffered from delayed parts.
- Allowed for higher resolution imaging with screen to sample distances of ~2-3 inches.



- Could induce temperature gradients up to 100C across samples.
- Rubber O-rings on quick connectors seemed to allow some amount of oxygen into the Ar environment around sample during last run.
- Sample loading/unloading was difficult

2nd Generation Compact Dual-Zone Furnace

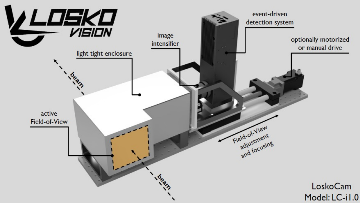


1. Sample stick
2. Linear motion device (bellows)
3. 6-way cube (UHV)
4. Flow gas
5. Glass-to-metal seal
6. Thermometry & electrical feedthrough
7. Resistive heating
8. Vacuum line
9. Glass tube

- Sample stick design for easy mounting.
- Linear motion device for variable sample lengths.
- Oxygen-tight seals.
- Modular feedthrough system.
- Baffles for sample and stick centering.

Work done by James Torres and Travis Carver.
Expected to be up and running for 22 run cycle.

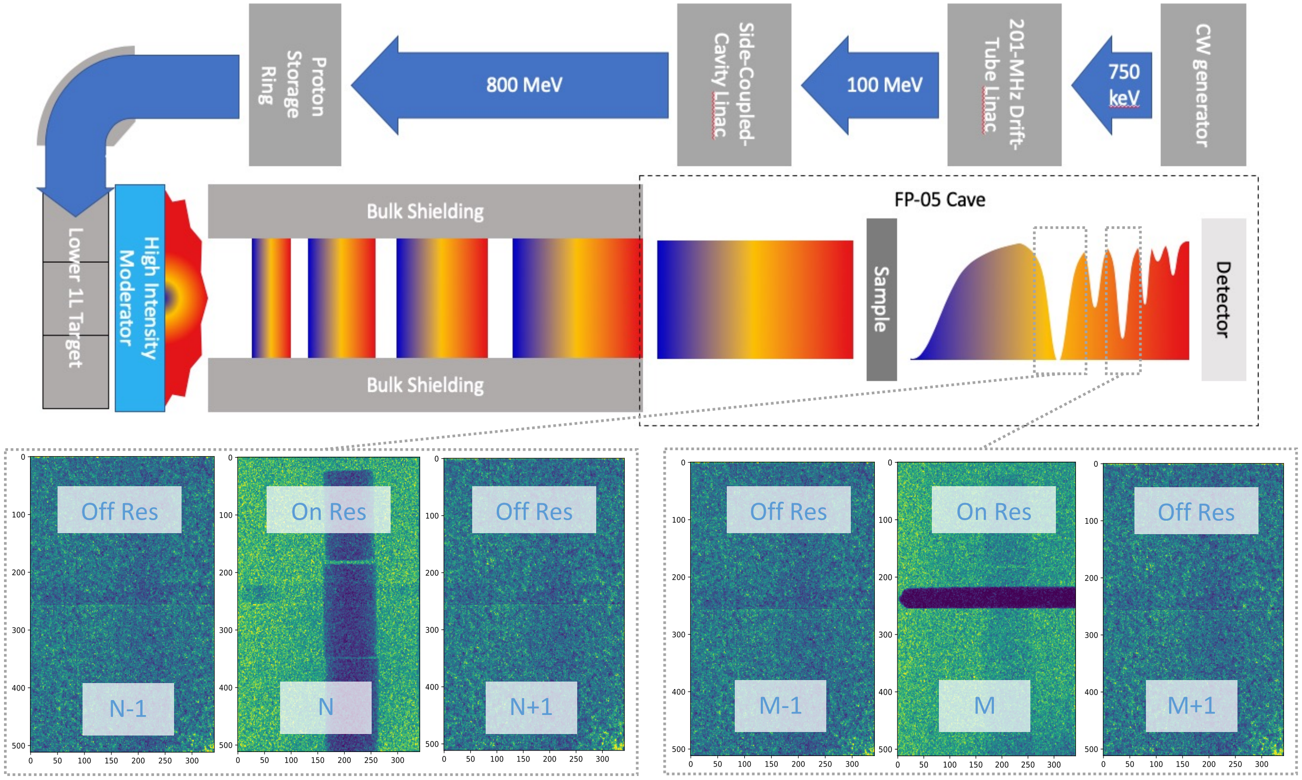
Better camera setups (BONUS)

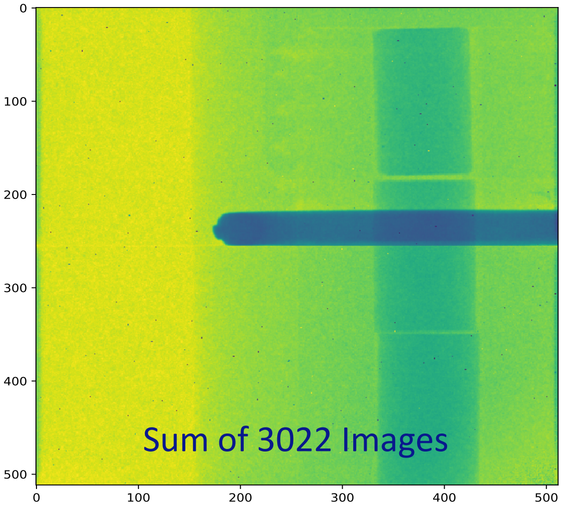


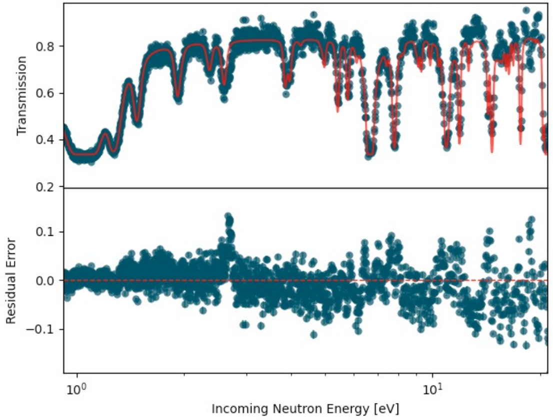
- Using hybrid camera setup, event-mode imaging with thick scintillator.
- Allow us to keep faster acquisitions and energy-resolved capabilities.
- From ASI and LoskoVision.

Energy Resolved Neutron Imaging on FP5

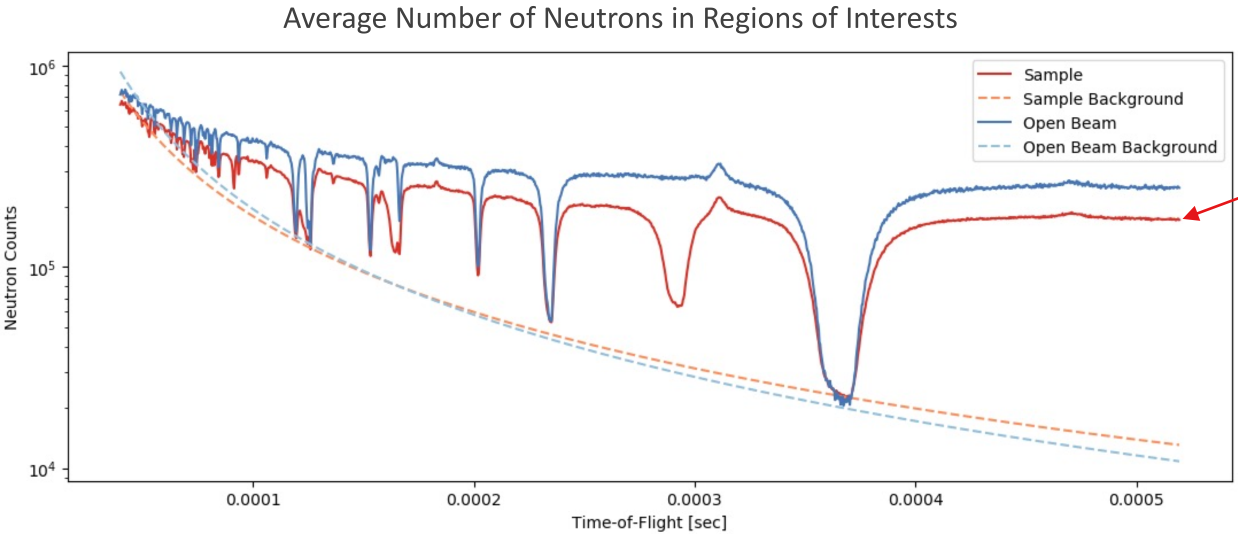
Using epi-thermal neutrons and absorption resonances to create contrast

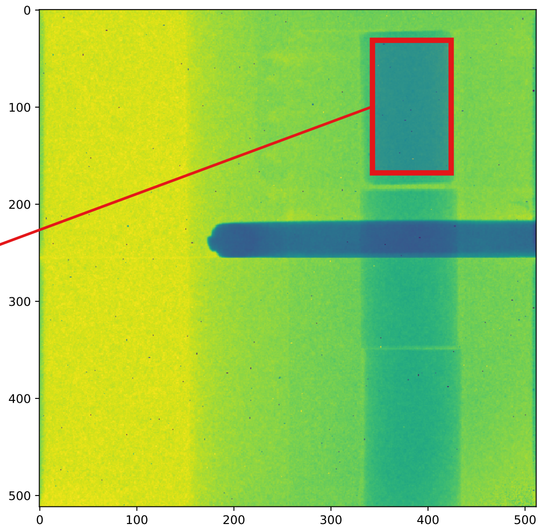




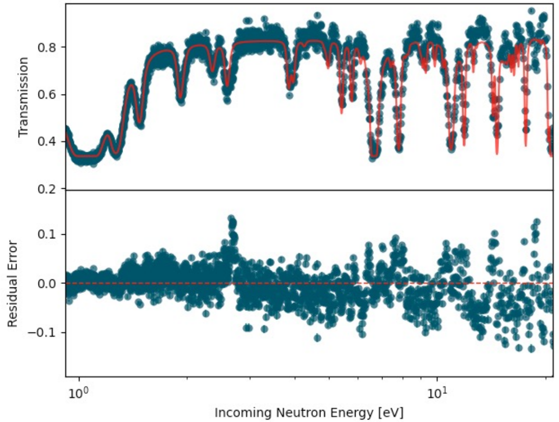


Extracting Isotopic Densities with ERNI





$$T(E_n) = N_b \frac{I_{obj}(E_n) - B_{obj}(E_n)}{I_{blk}(E_n) - B_{blk}(E_n)} = e^{-\sum_k n_k \sigma_{tot,k}(E_n)}$$

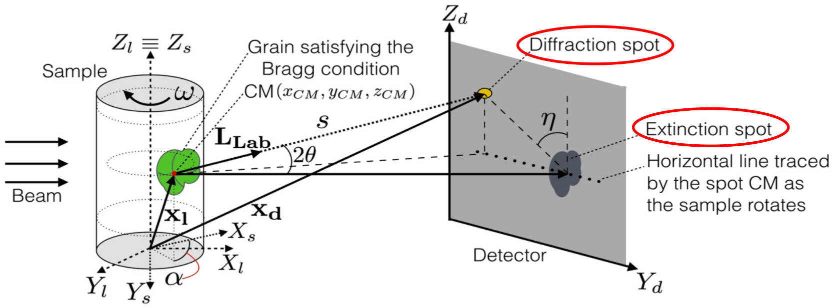


With a transmission spectrum, we can now use R-matrix codes like SAMMY, *or new material decomposition codes**, to extract areal densities (n_k) of specific isotopes.

** See talk by Thilo Balke*

Potential Futures of NI: Three-Dimensional Mapping of Crystal Grains

ToF 3D Neutron Diffraction (w/ Transmission)

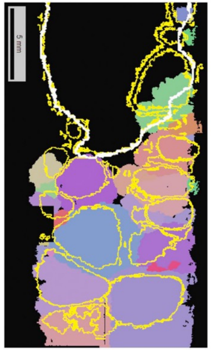


Highlights:

- Able to reconstruct the position and shape of **108 grains** in iron sample (verified with EBSD)
- Found the limit of resolution to be about **200 um**.
- Measurement in total took about **2 days**.(1 hour/exposure), good for static measurements.
- Uses MCP-TIMEPIX imaging detector (same as FP5)
- Difficult to ID grain orientation.

3D reconstruction of 108 grains

Validation of method with EBSD



Better spatial resolution and ability to identify grains, but slower and harder to get gain orientation.

→

Could possible be used to characterize and map grains larger (>100 um) in uranium materials!

First ToF-3DND measurements were performed on FP5 in December 2021

- Performed ToF3DND measurements on several depleted Uranium samples.
- proof-of-principle measurements to better optimize future attempts.
- Analysis is underway with advice from Sean Agnew at UVa.

Cereser et. al. SCIENTIFIC REPORTS | 7: 9561 | DOI:10.1038/s41598-017-09717-w