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Los Alamos NATIONAL LABORATORY

Neutron Imaging at LANSCE: Characterizing Materials for the Next Generation of Nuclear Reactor Designs

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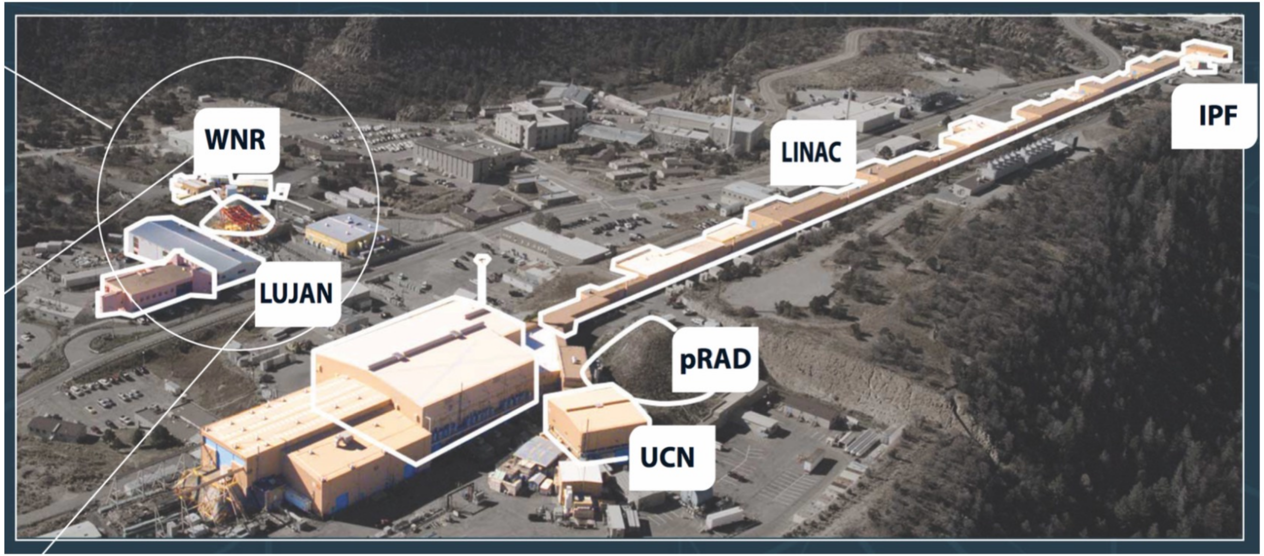
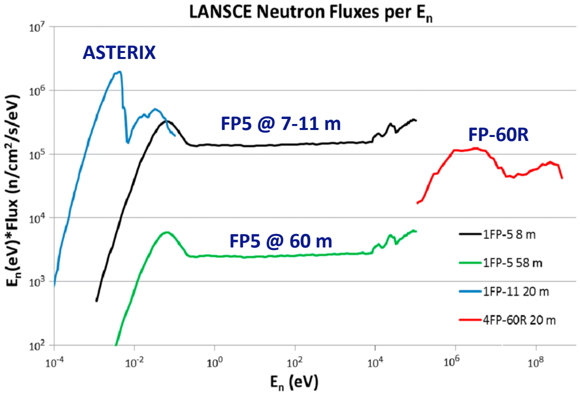
Computational Imaging XXI

electronic IMAGING

JANUARY 2023

Quick introduction to the Los Alamos Neutron Science Center (LANSCE)





Neutron Imaging Flight Paths

***Flight Path 5**:*

- *Thermal and epi-thermal neutron radiography and energy resolved neutron imaging.*

Flight Path 11 (ASTERIX):

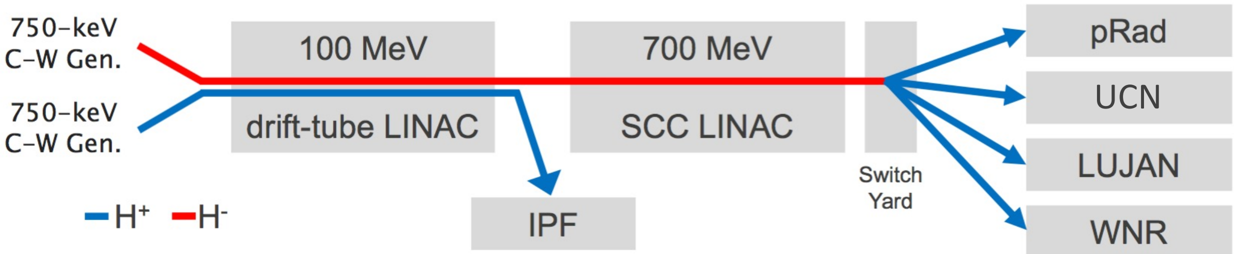
- Cold neutron imaging and neutron grating interferometry.

(coming soon)* *Flight Path 12:

- *Epi-thermal to MeV energy resolved neutron imaging for irradiated nuclear fuels.*

WNR 60R:

- MeV neutron imaging and scintillator characterization.

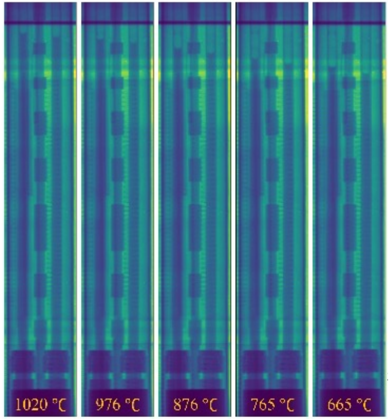


Some neutron imaging projects utilizing epi-thermal and thermal neutrons

Recently, we have been developing new neutron imaging capabilities @ LANSCE
with the main goal of characterizing materials for nuclear energy.

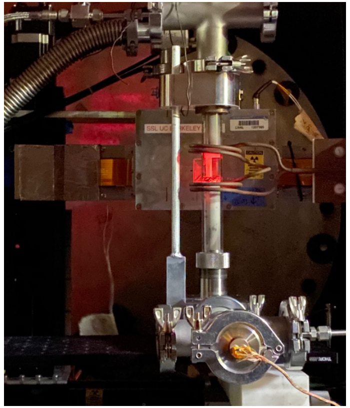
Thermophysical measurements of molten salts

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



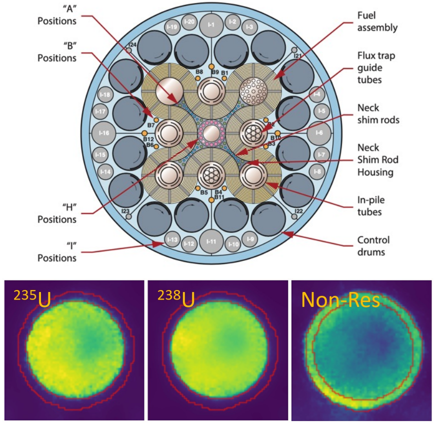
Quantifying hydrogen concentrations in YH_{2-x}

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



Neutron imaging as a PIE techniques with irradiated fuels

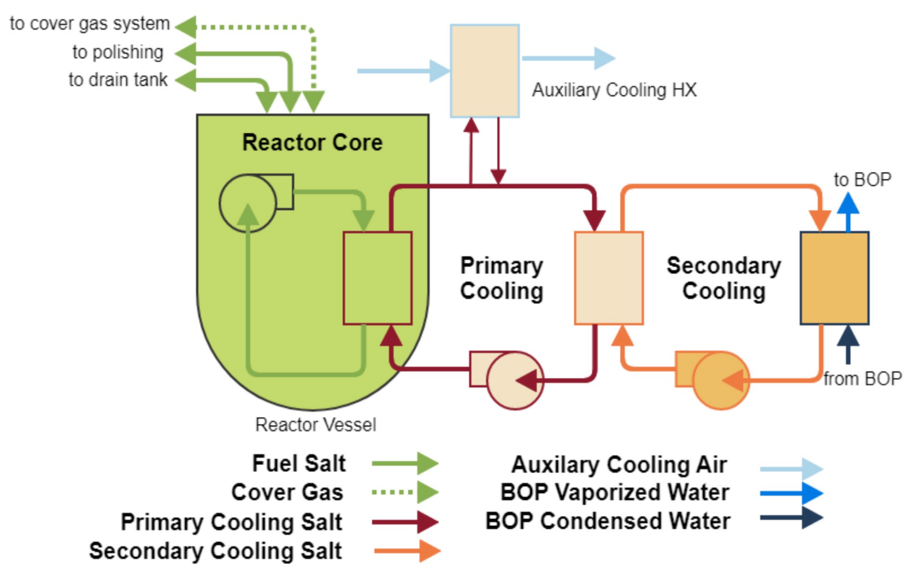
Utilizing ERNI techniques to map out isotopic distributions in **irradiated** fuels



Traditional — Imaging Techniques — Advanced

Current thermophysical needs for molten salts in nuclear energy

Where can salts be used in a reactor



Property	Application
Density (ρ)	**Thermal Hydraulics, Neutronics**
Heat Capacity (Cp)	Heat and Energy Transfer
Viscosity (μ, ν)	Thermal Hydraulics
Phase Stability, Solubility	Operating Parameters, Salt Composition
Enthalpy of Mixing (ΔH_m)	Thermochemistry
Thermal Diffusivity (α)	Heat and Energy Transfer
*Vapor Pressure (P_v)	Mechanistic Source Term Calculations

Properties

ρ	μ
κ	C_p

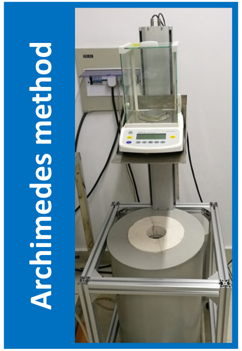
- Single salt components
- Binary, Ternary, and Quaternary
- Wide Temperature Ranges
- Corrosion and Fission Products

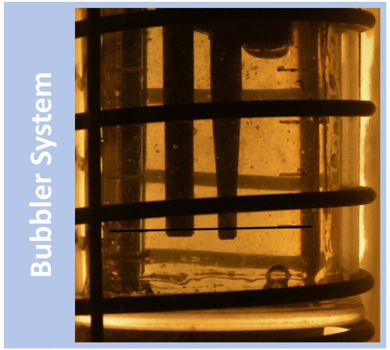
Increasing number of measurements

"**Thermal properties of molten salts**, both with and without fissile and fertile material, **are fundamental to modeling and simulation** that supports design, construction, operation, and accident progression evaluation of molten salt reactors"[1]

NEAMS
Nuclear Energy Advanced Modeling and Simulation

Why did we develop a new technique for measuring densities?





Challenges encountered using existing methods:

- Often need significant amount of sample material (10-100 g)
- May suffer from voids of bubbles
- Can not see the material during the measurement
- Surface Tension needs to be well understood
- May need calibration sample runs

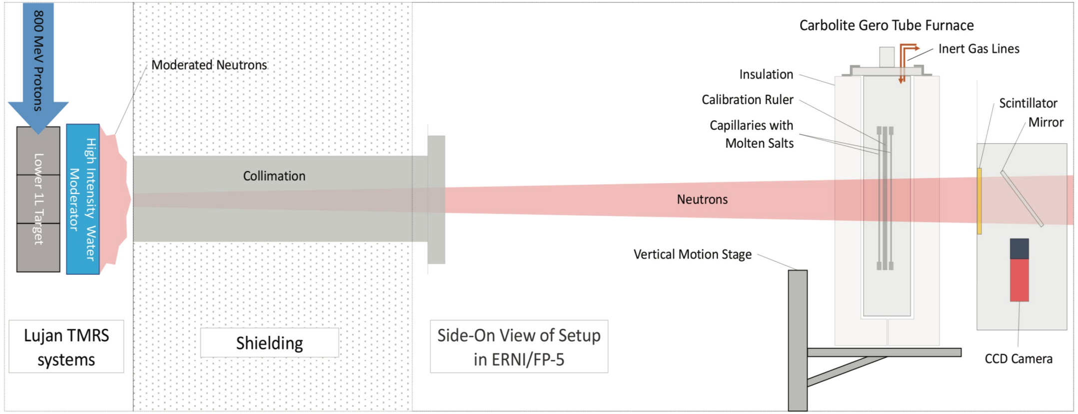
Large Discrepancies often observed in reported results!

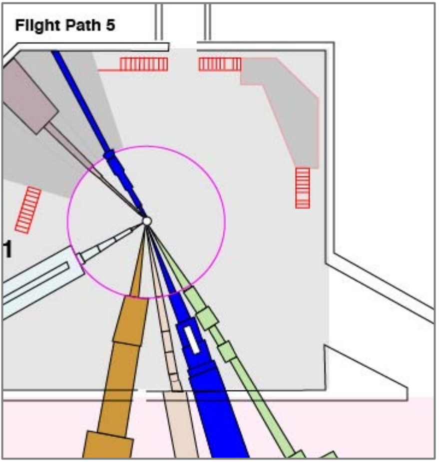
Measure densities of molten salts using neutron radiography!
Similar to a dilatometer, where you measure volume changes as a function of temperature. Or think mercury thermometer…

Density via Neutron Radiography

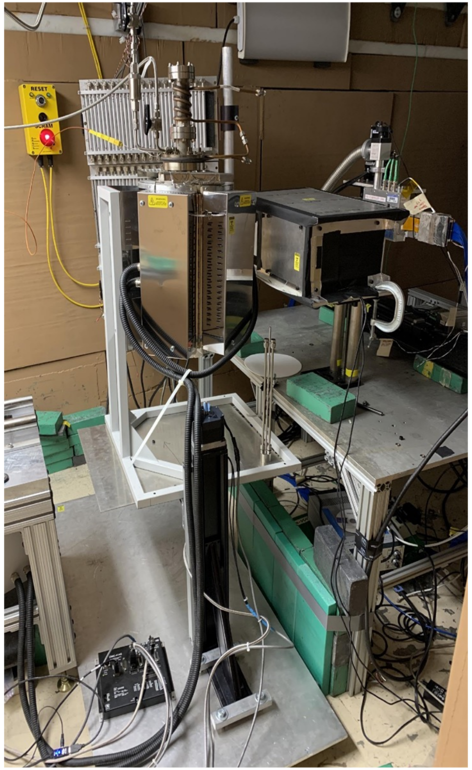
You have eyes on the sample the whole time (watch out for bubbles!).
Can have compact design that allows for less sample materials.
Modular setup: multiple samples can be measured simultaneously, and samples can be swapped quickly. Measurement times depend on furnace.
Can measure same samples multiple times.
Suitable for 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. Material compositions can be measured with diffraction!

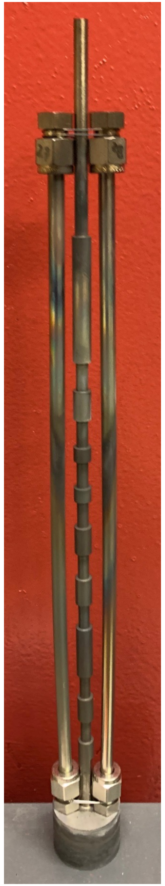
What does this technique look like in practice

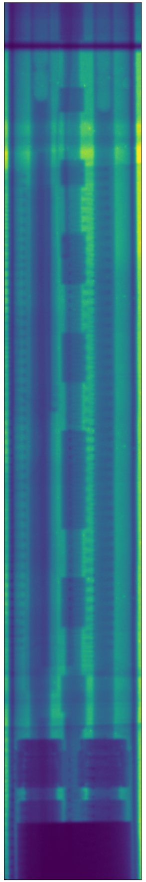




Measurements on FP5 October 2020







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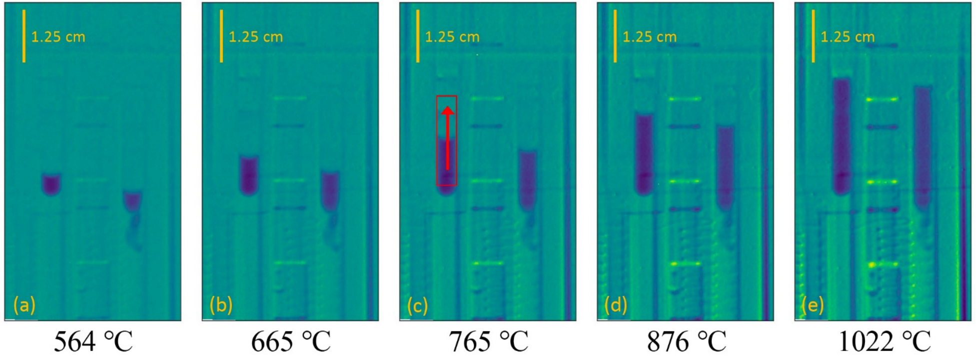
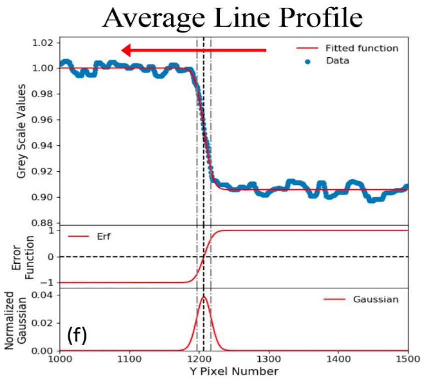
Measuring heights of fluids and determining volumes

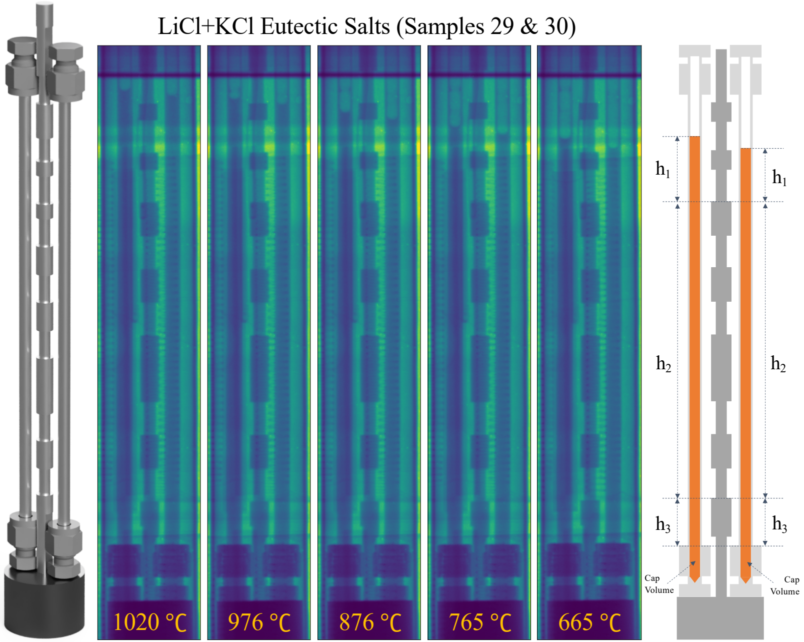
Images ratios at different temperatures were used to isolate changes in meniscus heights.





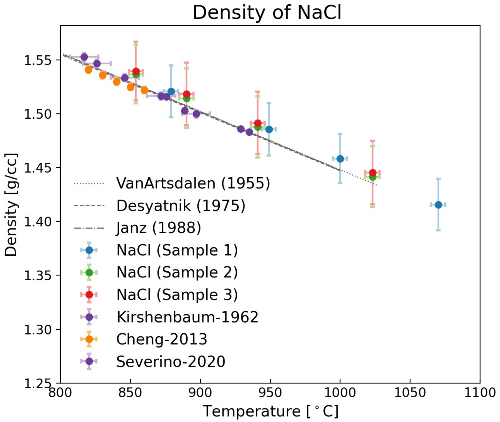
$$G(y) = A\, erf\left(\frac{y - y_0}{\sqrt{2}\sigma}\right)$$

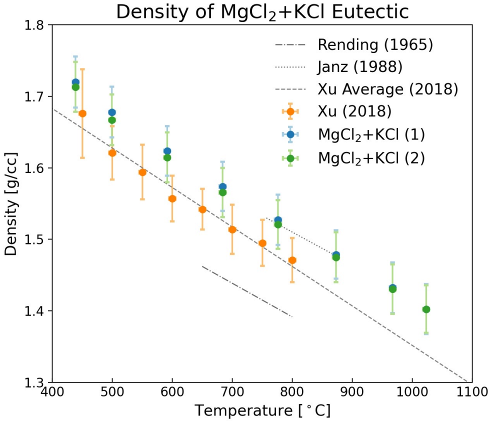
meniscus **positions (y_o)** and **uncertainties (σ)** were found by fitting line profiles with error functions.

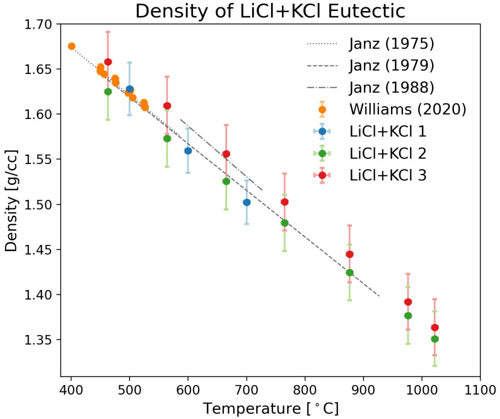


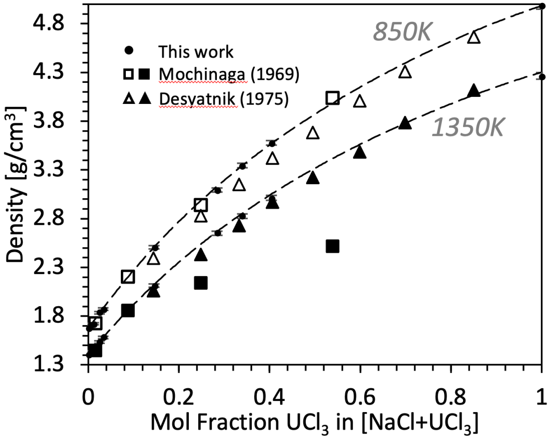
- CTE for 304 stainless steel was measured and used to adjust volumes at any given temperature.
- Assuming radial symmetry of ¼" Swagelok tubing, volumes were determined for three different regions.
- Volume of bottom cap was determined separately.
- With masses measured before hand, densities could be determined at each temperature.

Some density measurement results with molten salts









After measuring well known salt samples, various single- and multi-component chlorides were examined.

Single Comp: NaCl, LiCl, KCl, $CaCl_2$, and $MgCl_2$

Binary Eutectics: LiCl + KCl, $MgCl_2$+NaCl, NaCl+KCl, $MgCl_2$+KCl, NaCl+UCl_3, and KCl+UCl_3

Ternaries: NaCl + KCl + UCl_3 and LiCl +KCl + UCl_3

In addition, the NaCl + UCl_3 system was examined with varying molar concentrations of uranium.

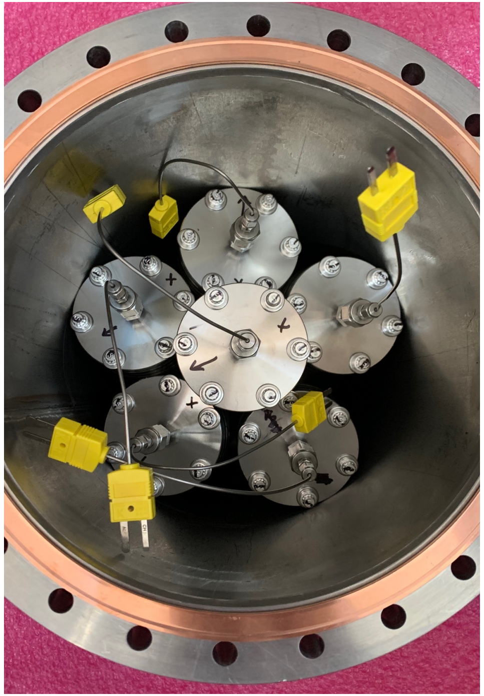
"Remote Density Measurements of Molten Salts via Neutron Radiography" **A. Long et. al.**, *J. Imaging* **2021**
"Thermophysical Properties of Liquid Chlorides from 600 – 1600K' **S. Parker et. al.**, *J. Molecular Liquids* **2021**

We were able to get our hands on some $PuCl_3$-based salts!

Measurements of $PuCl_x$- based salts as part of an LDRD-DR, TerraPower GAIN award, and the MSR National Campaign

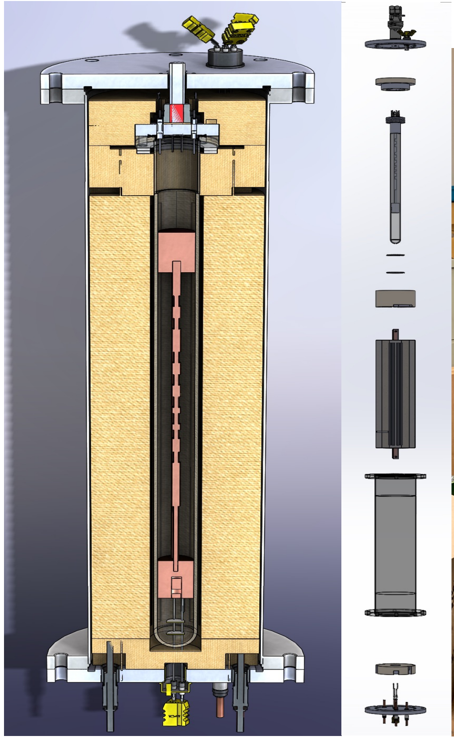
$PuCl_3$ material was secured by LDRD-DR team from INL

Composition	Ratios (mol%)
NaCl-$MgCl_2$- $PuCl_3$	50.6 - 42.4 - 7.0
NaCl-$MgCl_2$- $PuCl_3$	51.5 - 45.0 - 3.5
NaCl-UCl_3- $PuCl_3$	67.0 - 28.7 - 4.3
NaCl-UCl_3 -$PuCl_3$	67.0 - 30.8 - 2.2
*NaCl- $PuCl_3$	98.0 - 02.0
*NaCl- $PuCl_3$	64.0 - 36.0



This was a monumental 2-YEAR effort by Matt Jackson, Marisa Monreal, and Scott Parker!

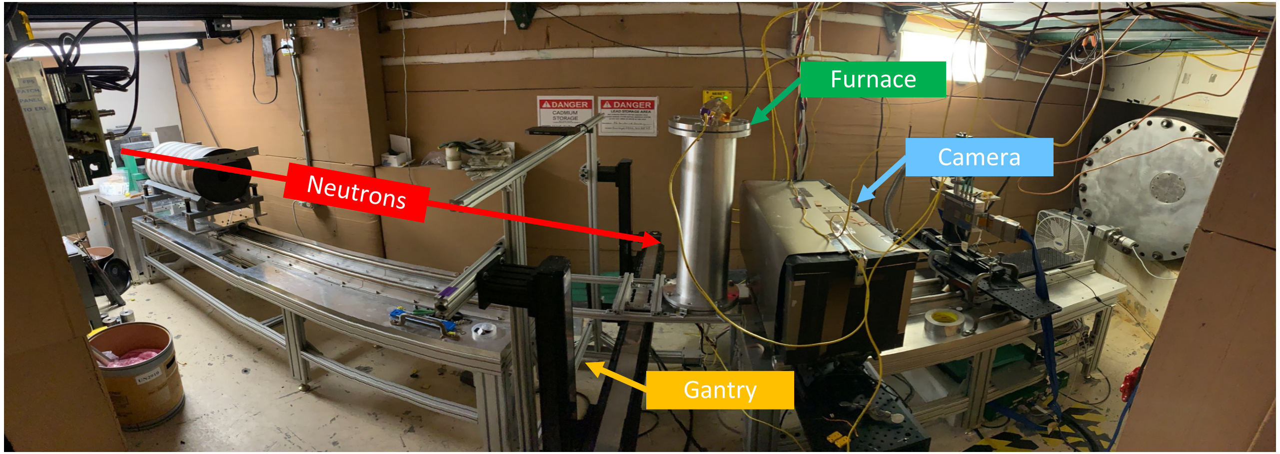
Required a New State-of-the-art Furnace

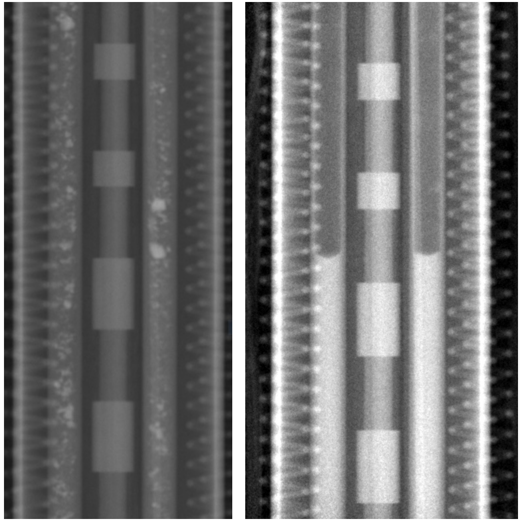


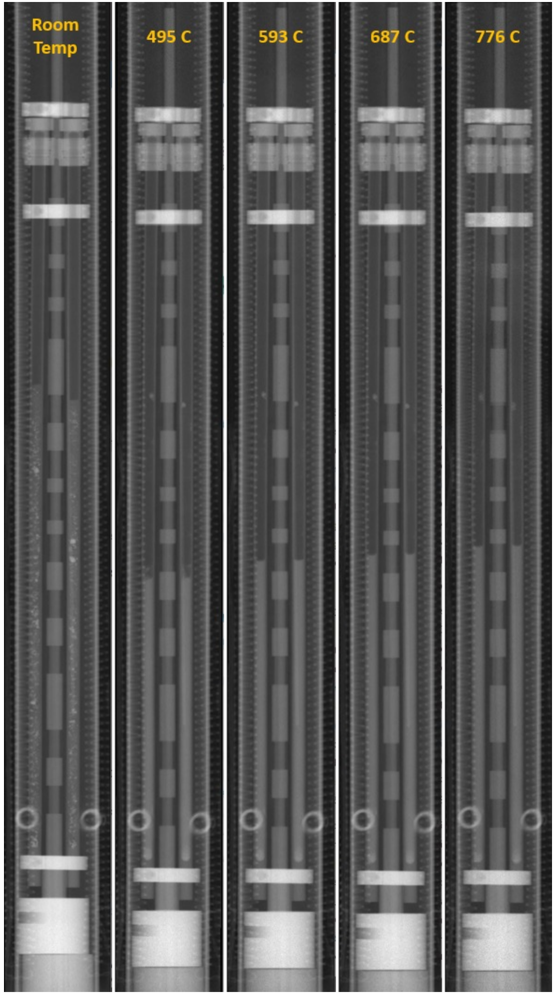
Furnace Design by Travis Carver

Los Alamos NATIONAL LABORATORY GAIN Gateway for Accelerated Innovation in Nuclear TerraPower A Nuclear Innovation Company

Most Recent Measurements on FP5: $PuCl_3$-based Salt







Some Experimental Information:

- Beam Time on FP5: Dec 10th – 20th
- Maximum Temperatures: ~850 C
- Exposure Times: 1 min
- ~12- to 15-hour measurements per sample pair
- Each pair was measured once*
- Used gantry to move samples through FOV
- 4-5 full sample scans were performed at elevated temperatures for each sample-pair

Analysis is currently underway so stay tuned!

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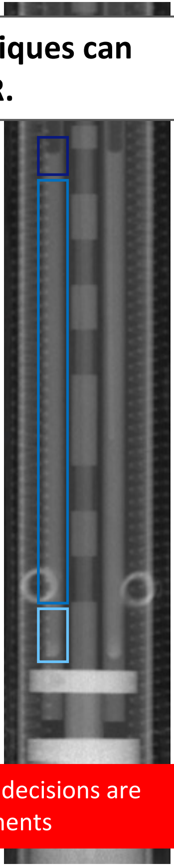
Utilizing advanced NI techniques to extract more…

We need to be able to extract as much information as possible, especially if real world decisions are being made based on NI results.

How advanced neutron imaging techniques can help build confidence in DvNR.



Flight paths at pulsed neutron facilities have the potential for delivering high-quality and information-rich data sets.

Achieving higher resolution can help determine contact angles (wetting vs. non wetting) and relate that to surface tension.

Resonance Imaging (transmission) can be used to measure actinide areal densities and temperature profiles within the melts.

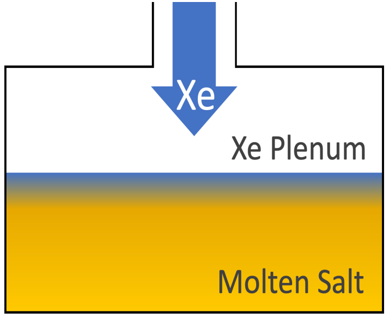
Neutron diffraction and scattering can be used to measure chemical compositions and potential contamination.

All of this information comes together to boost confidence in **WHAT** is being measured and **HOW** it is being measured.

Confidence is **CRITICAL** if real world reactor design decisions are being made from these types of measurements

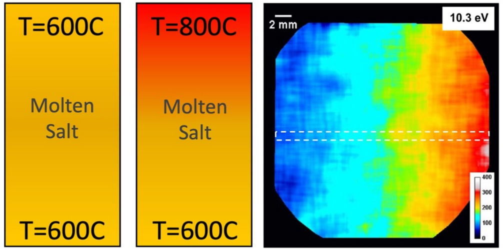
Additional applications of Resonances Imaging (Venus & FP5)

Diffusion of fission gases



Measuring Xe areal densities as a function of position and time.

Thermal Diffusivity



Changes in resonance broadening due to changes in temps as a function of position and time.

Underlying techniques have already been demonstrated by Tremsin *et. al.*

These measurements will not only be difficult to design but also require advance imaging analysis (think T. Balke code for resonance imaging). We need to work with computational imaging community to develop analysis tools for these advanced techniques.

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)
Molten Salt Reactor Program

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

Back up slides…

and other things related to neutron imaging for NE programs.

Why Do We Need Post Irradiation Examination?

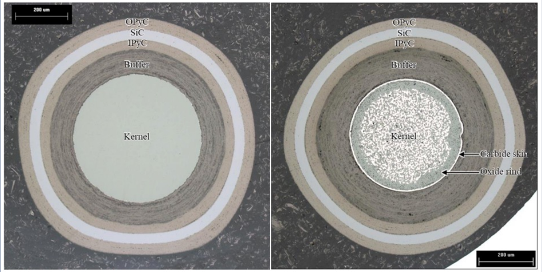
Post Irradiation Examination **(PIE)** plays a critical role in understanding and predicting nuclear material response and performance under normal, transient, and accidental conditions

Need to test drive (or crash test) nuclear fuels and materials in conditions that they might encounter in their lifetime of deployment.

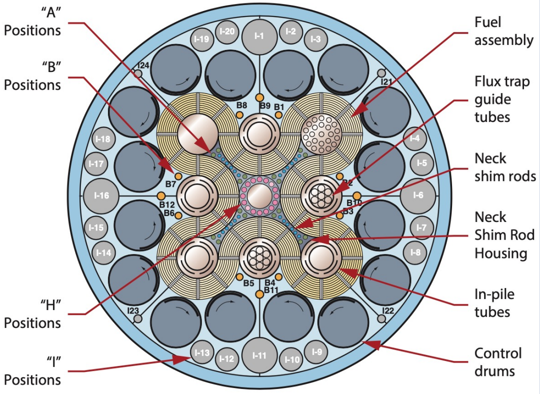
As-Built Characterization

Obtain baseline characteristics of fresh fuel before irradiation:

- Microstructure, geometries, isotopic and chemical compositions.



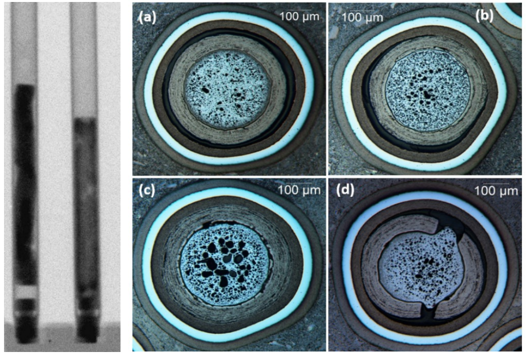
Loading and Irradiation!



Irradiate for months to years!

Post Irradiation Examination

Non-destructive & destructive examination @ NSUF facilities

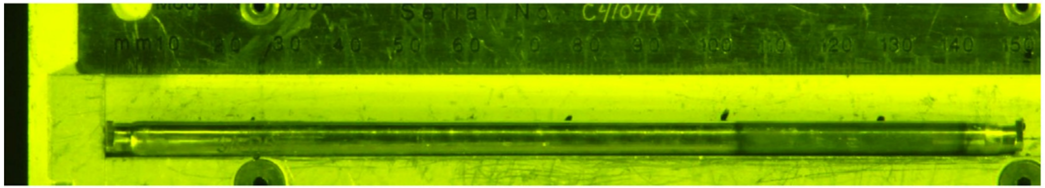


Comparisons to determine changes due to irradiation

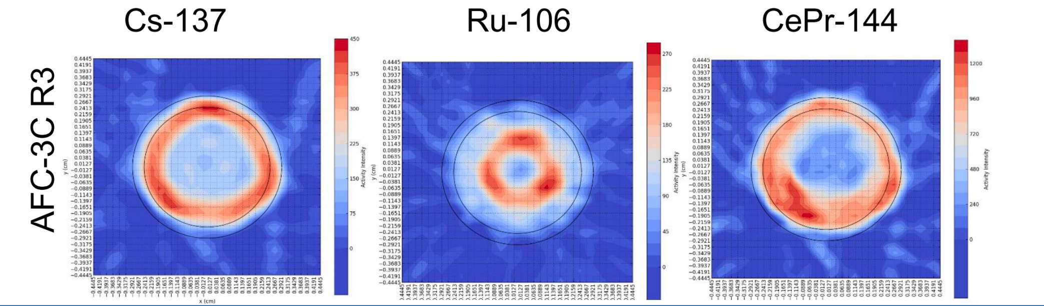
Stempien J.D, *et. al. AGR-2 TRISO Fuel Post-Irradiation Examination Final Report*. 2021
Campbell J.L., Advanced Test Reactor User Guide, 2021

Current Status of PIE Capabilities…

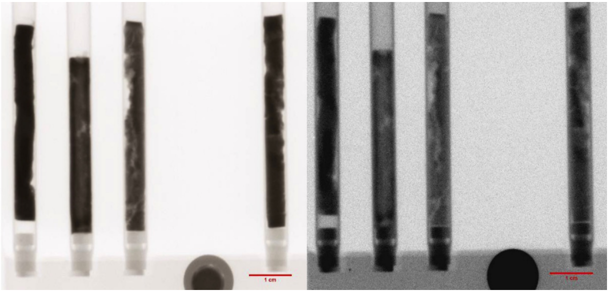
Visual Examination



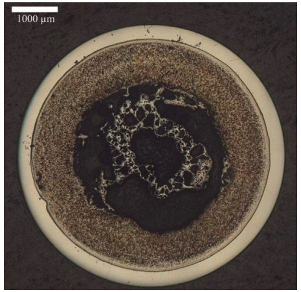
Gamma Emissions Tomography



Neutron Radiography



Optical Microscopy



…and many more!

Challenges facing PIE of Fuels

- **PIE can be extremely inefficient!!** You are either extracting less-detailed global info or highly-detailed local info.
- Samples often too hot to handle in glove box

Economic budgeting of samples and test!

Key Questions to address:

- Which regions of the sample are "normal", which regions are "unusual"?
- Which regions provide the best return of investment when prepared in destructive testing?

How Pulsed Neutrons Can Help

- Characterize the entire irradiated sample volume of a few cm^3 (**isotopics**, **geometry**, **phase fractions**, and **microstructure**)
- Non-destructive and remote measurements can guide further destructive measurements.

HIPPO to probe bulk microstructure!

ERNI to map out isotopic distributions!

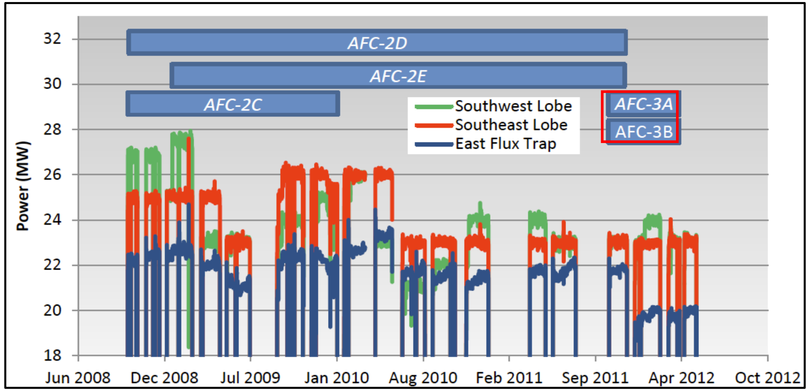
Harp et al. "Postirradiation examination results of several metallic fuel alloys and forms from low burnup AFC irradiations" Journal of Nuc. Mat. 2018

Proof-of-principle with Irradiated Samples from AFC-3C

Irradiation campaign for Nuclear Technology Research and Development (NTRD) Advanced Fuel Campaign (AFC) program.

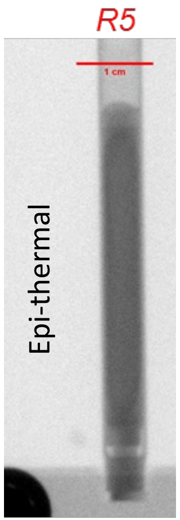
Irradiations at the ATR in INL exploring alternate alloys and forms for fast reactor and transmutation fuels (U-Mo & UPdZr)

- Irradiated at the ATR from Sep. 2011 to Apr 2012.
- Pd additive to mitigate fuel-cladding chemical interaction (FCCI)
- Low burn up -> Fuel cladding mechanical interaction due to swelling.



3A and 3B fuel sample information

Rodlet ID	Alloy	Fuel Form	Bond Material	Nominal Smear Density
3A-R1	U-10Mo	Solid	Sodium	75%
3A-R2	U-10Mo	Annular	Helium	55%
3A-R4	U-10Zr	Annular	Helium	55%
3A-R5A	U-1Pd-10Zr	Solid	Sodium	75%
3A-R5B	U-2Pd-10Zr	Solid	Sodium	75%
3B-R1	U-4Pd-10Zr	Solid	Sodium	55%
3B-R2	U-4Pd-10Zr	Annular	Helium	55%
3B-R4	U-10Mo	Solid	Sodium	55%
3B-R5	U-10Mo	Solid	Sodium	55%



Microstructure with HIPPO

- Measure phase fractions of metallic samples (α-U + δ-UZr_2)

Isotopic mapping with ERNI

- Observation of chemical redistribution (U, Zr, Pd)
- Look for Xe in sample for fission gas representation.
- Characterize U235 and U238 enrichment levels for burn-up.

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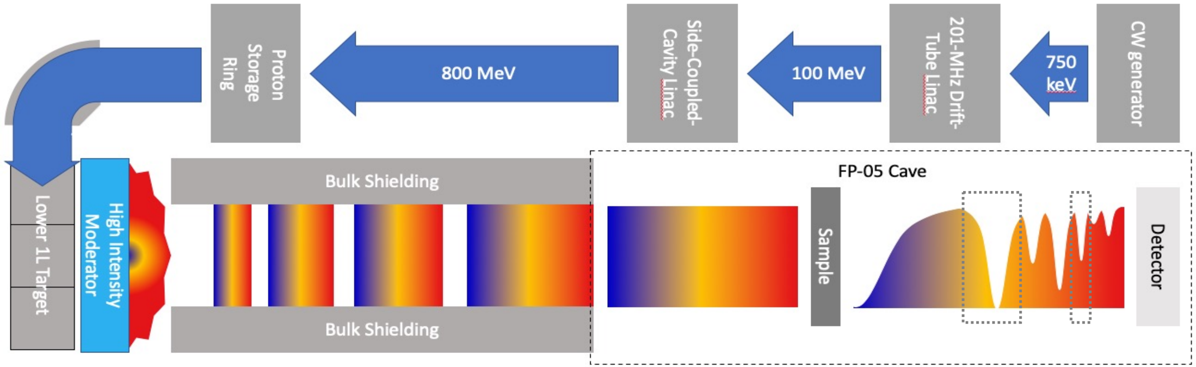
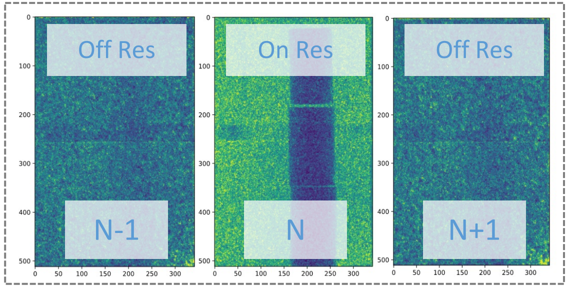
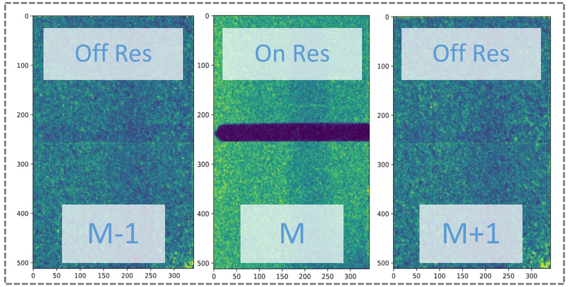
Harp et al. "Postirradiation examination results of several metallic fuel alloys and forms from low burnup AFC irradiations" Journal of Nuc. Mat. 2018

Energy Resolved Neutron Imaging on FP5

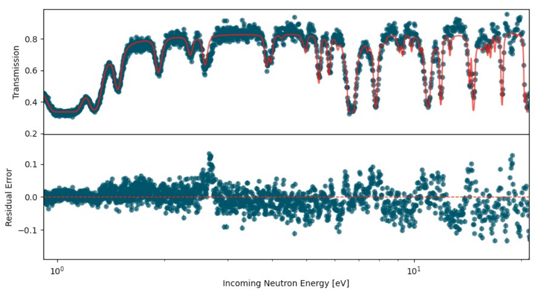
Using epi-thermal neutrons and absorption resonances to create contrast





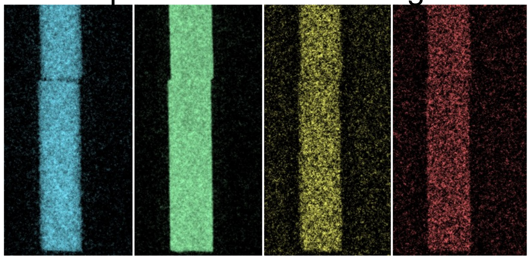


Fitting transmission spectra from single pixel

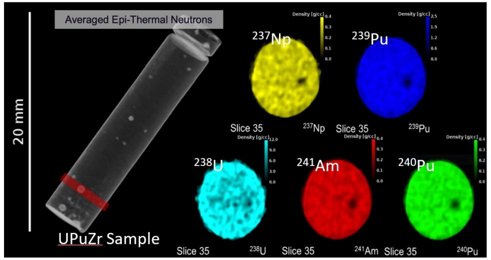


Obtain 2D heat maps of each isotope

Isotopic areal densities in g/cm^2



CT reconstruction for each isotope



3D Isotopic reconstruction

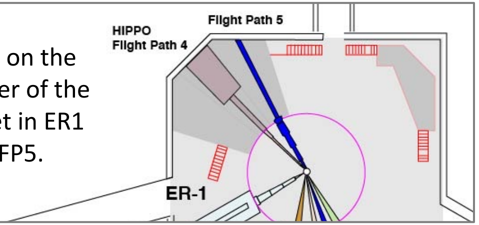
Los Alamos NATIONAL LABORATORY

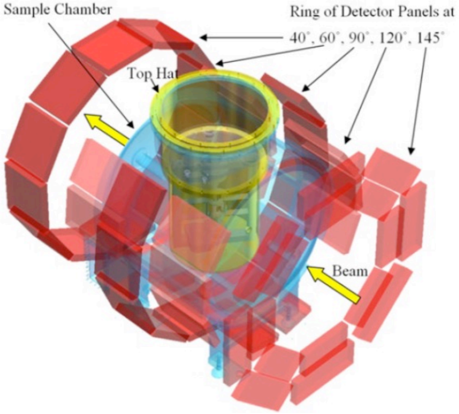
"3D isotope density measurements by energy-resolved neutron imaging", Losko *et. al.* Scientific Reports **volume 12**, Article number: 6648 (2022)

Diffraction Measurements of AFC-3A-R5A on HIPPO

High-Pressure-Preferred Orientation time-of-flight powder diffractometer

Located on the lower tier of the 1L target in ER1 next to FP5.

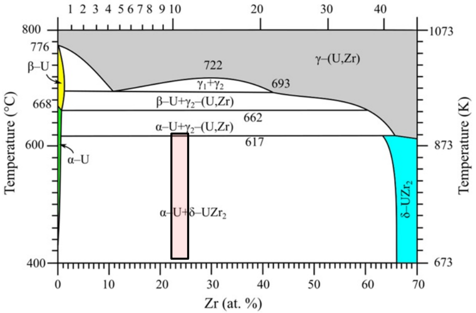




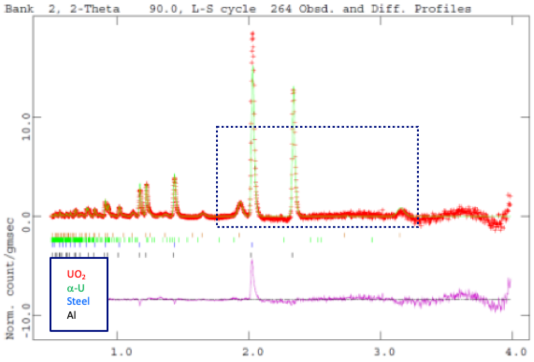
High intensity and large solid angle coverage allows for efficient texture (grain orientation distributions) and phase fraction measurements.

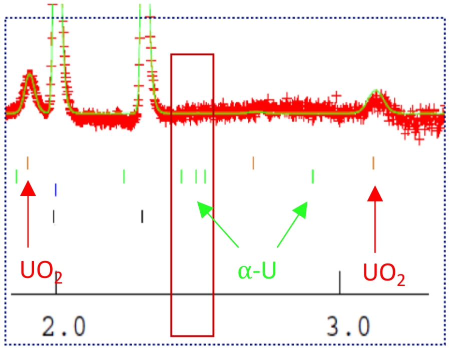
Expectations: cast α-U + δ-UZr_2

- Per phase diagram: U-10wt%Zr (U-22.5 at% Zr) should consist of α-U and δ-UZr_2
- HIPPO measurement on fresh UZr samples results in ~85 wt% α-U in a U-10wt%Zr sample
- Possible phase changes due to irradiation and heating
  - Yao *et al.* observed both ω- and γ-phases in similar irradiated U-10Zr samples (AFC-3A R4)



HIPPO Observation: Lots of UO_2 peaks and No α-U peaks!?





Results from HIPPO measurements show that the sample fully oxidized while in capsule.
Need to work out capsule sealing to make sure this does not happen with future samples.

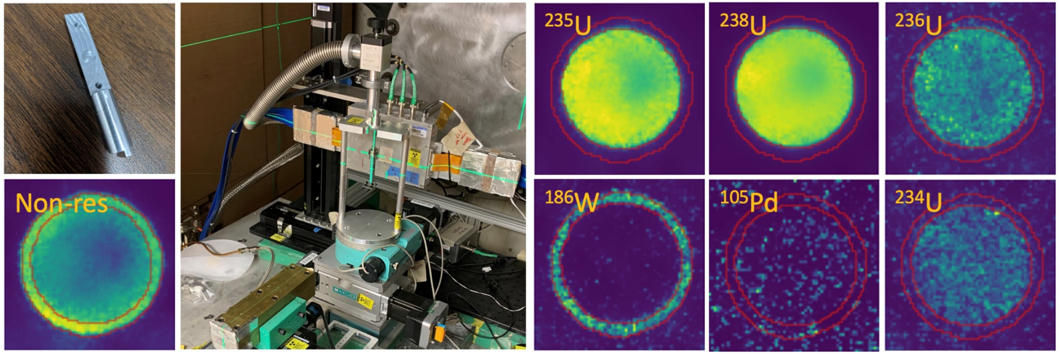
ERNI Measurements of AFC-3A-R5A on FP5

PIE Sample sent to LANSCE

- 6 mm ∅, ~1.5 mm thick disk prepared from AFC-3A-R5A
- Burnup of 2.5 % fissions per initial metal atom (FIMA)
- Solid metalic U(1Pd)10Zr sample encapsuled in aluminum shell.
- Received at LANSCE November 2019 with NSUF grant

- Dose rate on contact: ~3R/hr (DOE allowable dose for public is 0.1R)
- Neutron diffraction on HIPPO to study phase fractions and microstructure.
- Energy Resolved Neutron Imaging on FP5 to measure actinide distributions.
- Acted as dry run for more active samples.

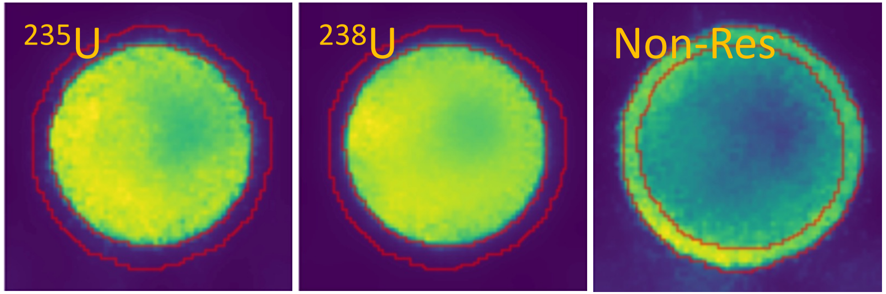
ERNI measurements on U-1Pd-10Zr sample on FP5 during 2021-RC



Using new in-house isotopic decomposition code "MAD-Neutron" by Thilo Balke *et. al.*

Some results from ERNI Measurements of AFC-3A-R5A on FP5

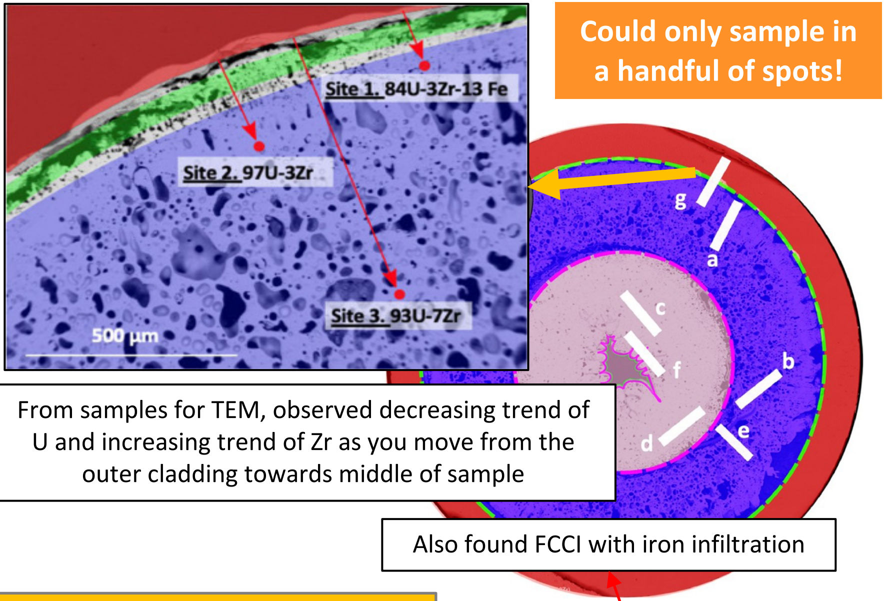
ERNI Results of Uranium Distribution in AFC-3A-R5A



- **Observed decrease in Uranium (^{235}U and ^{238}U) as you move towards the center (with slight offset) of the sample.** This agrees generally with other PIE measurements by Yao et al. on U-10Zr samples (AFC-3A-R4) by Transmission Electron Microscopy (TEM)
- **Observed increase in Uranium enrichment as you move outward!!** This can be explained by the use of a cadmium shroud absorbing thermal neutrons (En<0.4 eV) during irradiation.

PIE-TEM of AFC-3A-R4 by Yao *et al.*

Could only sample in a handful of spots!



From samples for TEM, observed decreasing trend of U and increasing trend of Zr as you move from the outer cladding towards middle of sample

Also found FCCI with iron infiltration

More on this later!!

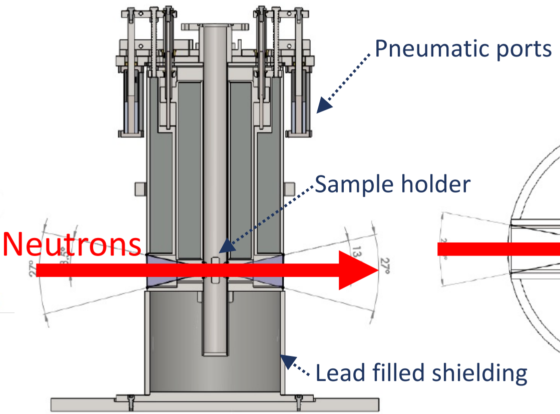
ERNI Results and Takeaway

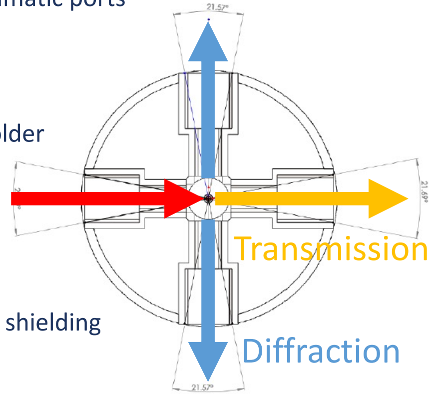
- ERNI PIE measurements on the AFC-3A-R5A sample are promising!
- ^{235}U and ^{238}U show significant variations within the sample with a reduction of U towards the middle.
- Would be nice if we could confirm corresponding increase in Zr.

Future of PIE at LANSCE: The SHERMAN Cask

Moving forward we plan to characterize more highly irradiated nuclear fuels!

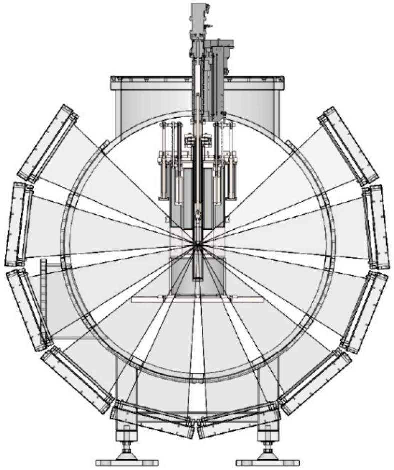
- End goal is characterization of irradiated actinide mixed oxide (MA-MOX) fuels with estimated dose rates at **~900 R/hr** on contact.
- Higher activity samples require shielding cask for transportation from INL to LANSCE and sample handling while at Lujan Center.





SHERMAN on HIPPO

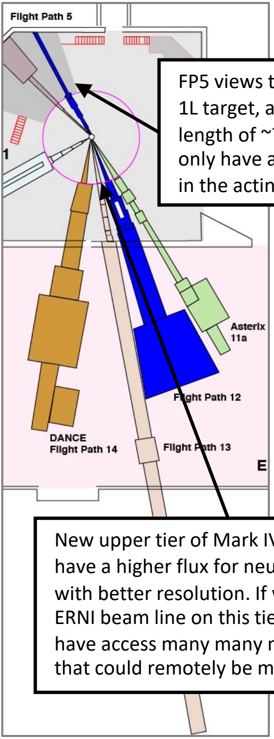
Once certified, first measurements of irradiated fuels will be performed on HIPPO (Diffraction and Imaging)



- First proto-type of SHERMAN was built in 2020-2021
- Working on authorization and safety basis for INL & LANL operation (drop test and fire simulations along with detailed shielding calculations)
- Commissioning with dry runs (without lead filler) to test sample motion control, shutter operation, all while using surrogate samples in beam on HIPPO.

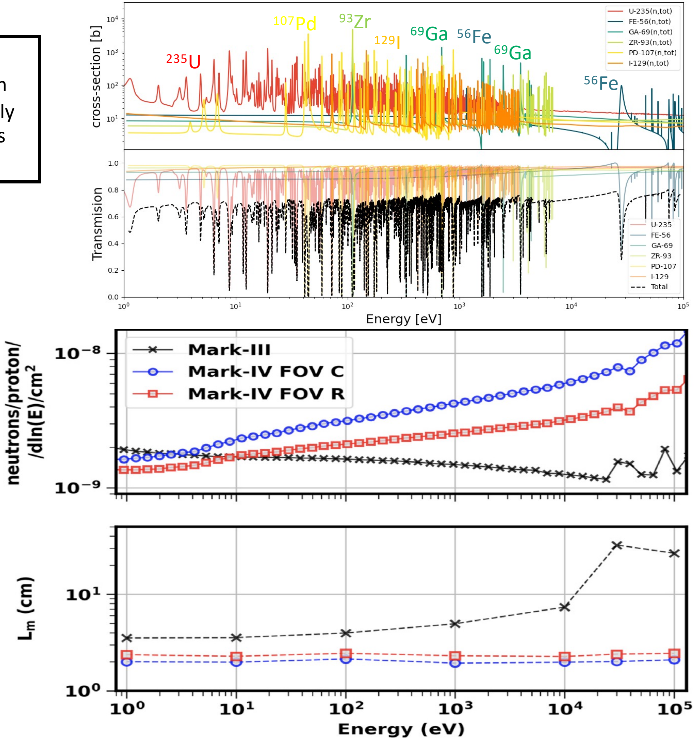
Future of PIE at LANSCE: ERNI on FP12

Moving forward we plan to probe isotopes with higher energy neutron absorption resonances!

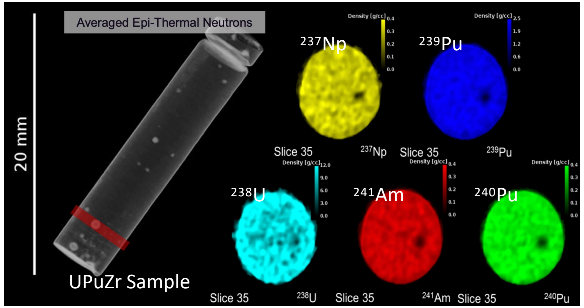


FP5 views the lower tier of the 1L target, and with a flight path length of ~7-11 m we effectively only have access to resonances in the actinide regime.

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!

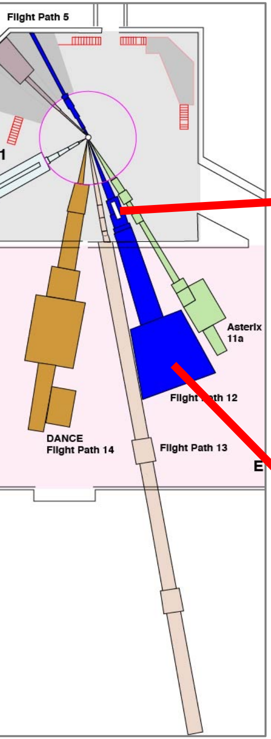


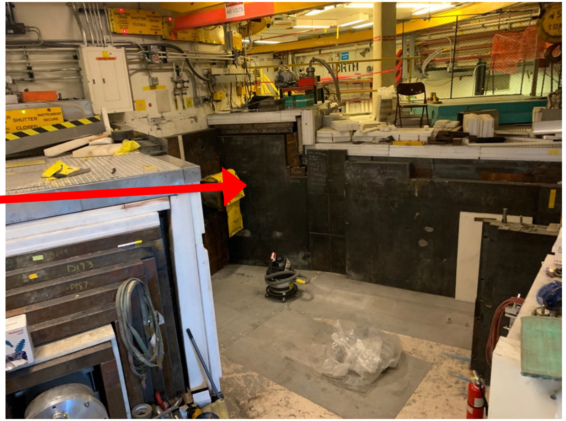
New beam line on upper tier target could map out fission products and isotopes down to the Fe group. For example, fresh and irradiated fuels, along with materials like U-Nb and Pu-Ga, can be non-destructively characterized.



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

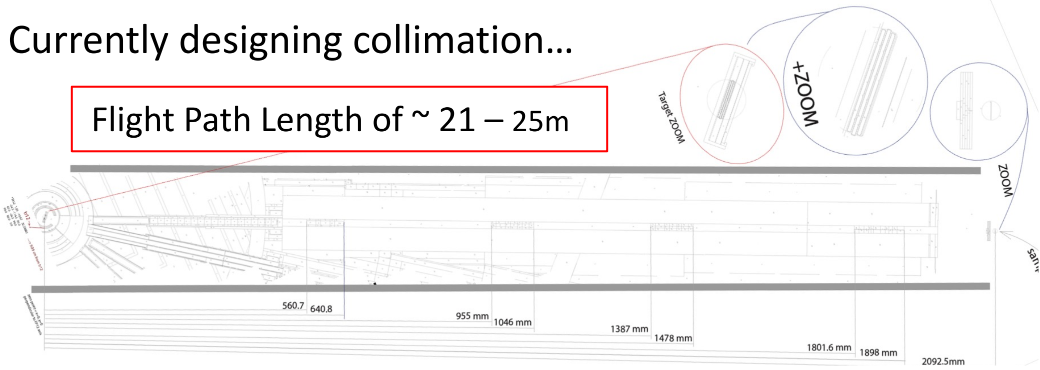
Progress of ERNI on FP12

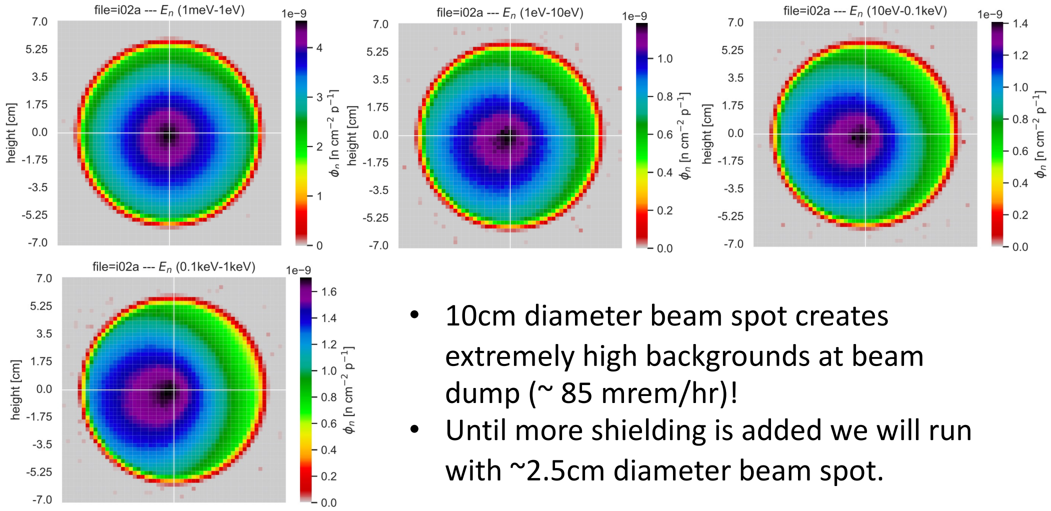




Currently designing collimation…

Flight Path Length of ~ 21 – 25m

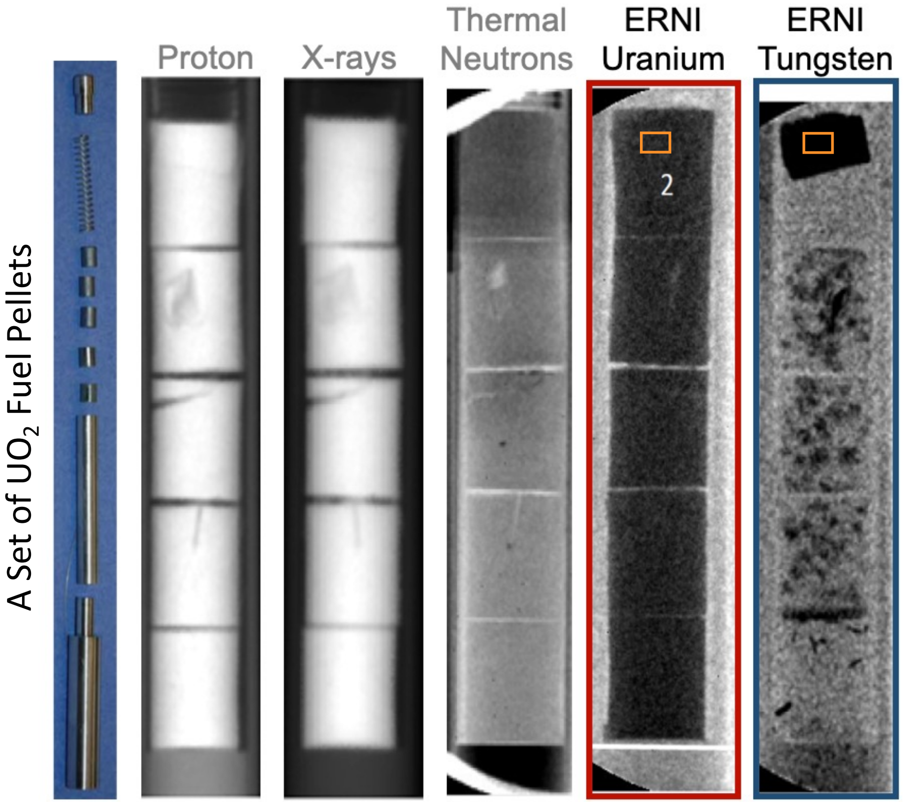




- 10cm diameter beam spot creates extremely high backgrounds at beam dump (~ 85 mrem/hr)!
- Until more shielding is added we will run with ~2.5cm diameter beam spot.

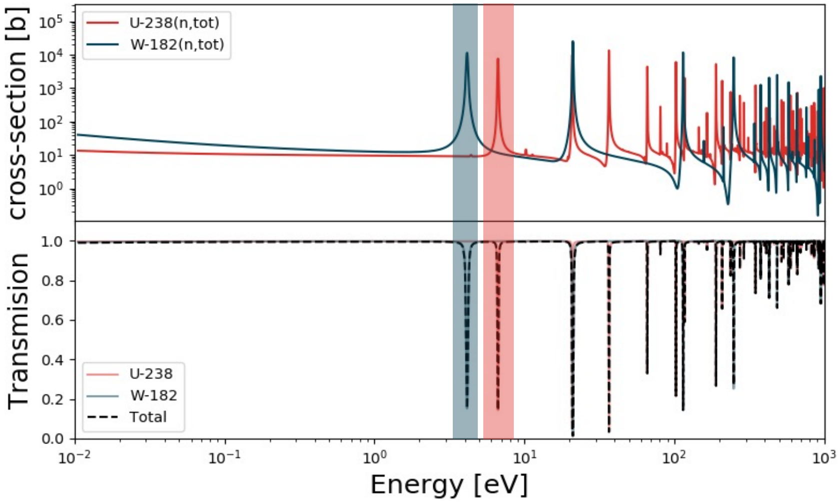
Will hopefully design and install new FP12 collimation and be able to run test during the 2023 LANSCE run cycle.

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

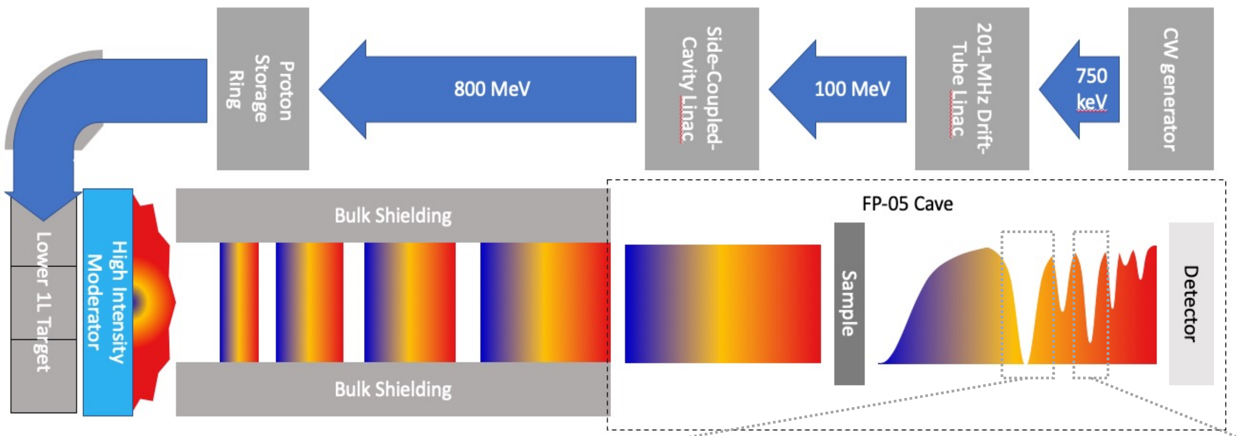
Each pixel has additional energy information in the form of a transmission spectra

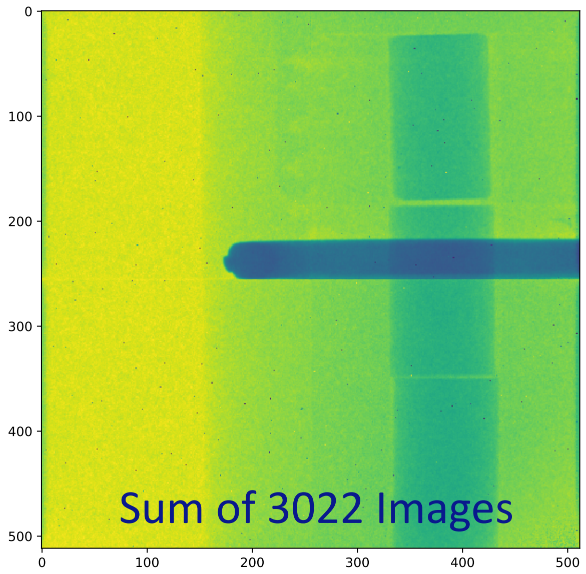


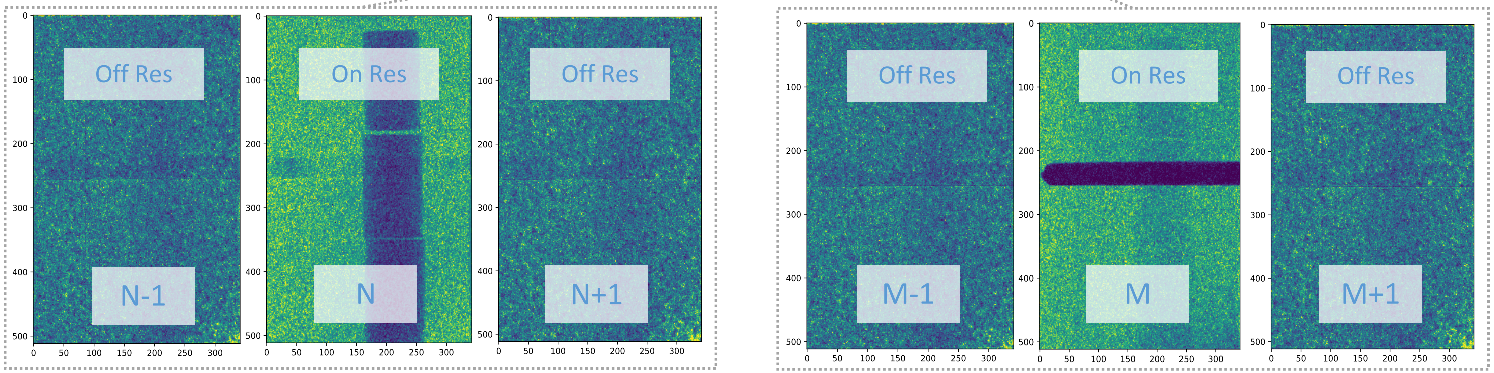
Morris *et. al.* Review of Scientific Instruments 84 (2013)
Tremsin *et. al.* Neutron News 24 (2013)

Energy Resolved Neutron Imaging on FP5

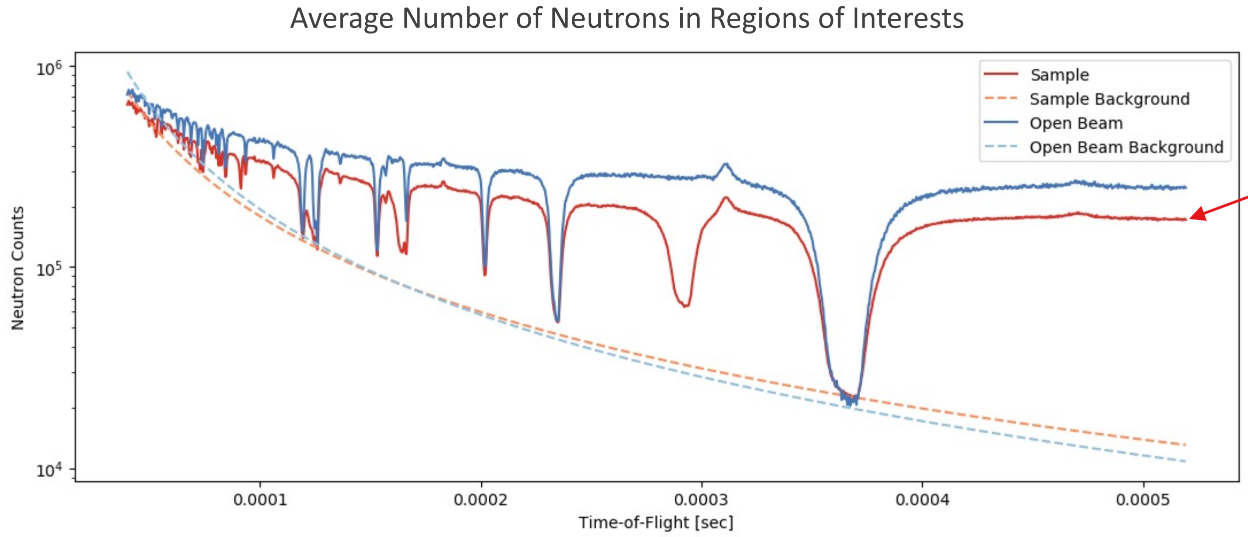
Using epi-thermal neutrons and absorption resonances to create contrast

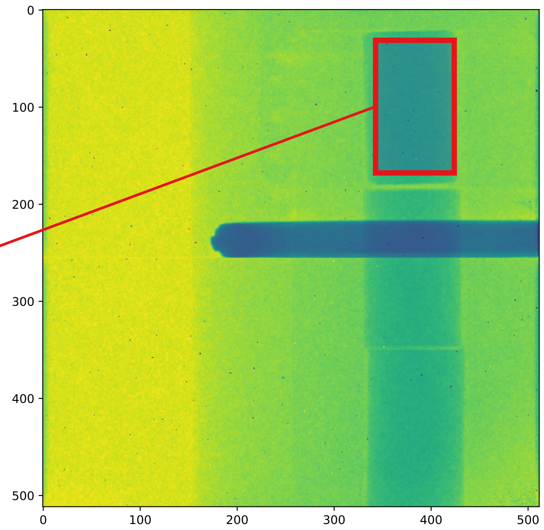






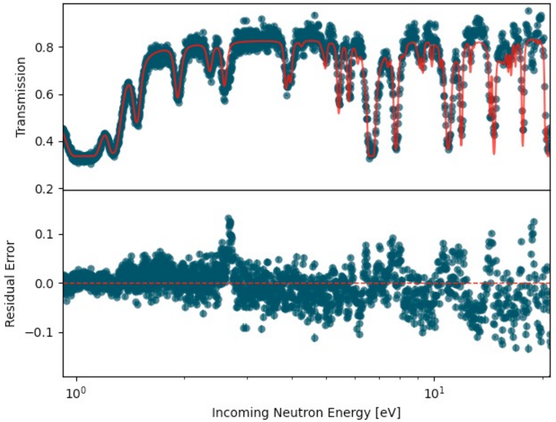
Extracting Isotopic Densities with ERNI





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

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*