Building up Capabilities to Characterize Hydrogen in YH_{2-x} with Neutron Radiography at LANL

Alexander Long, Travis Carver, Erik Luther, Vedant Mehta, Caitlin Taylor, James Torres, Aditya Shivprasad, Holly Trellue, and Sven Vogel

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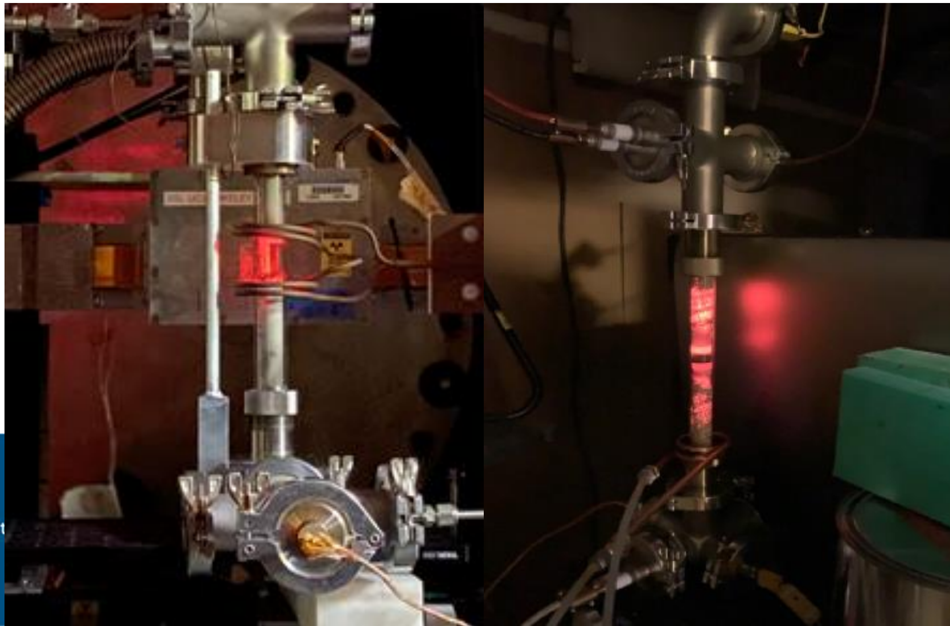


INL Idaho National Laboratory

OAK RIDGE National Laboratory

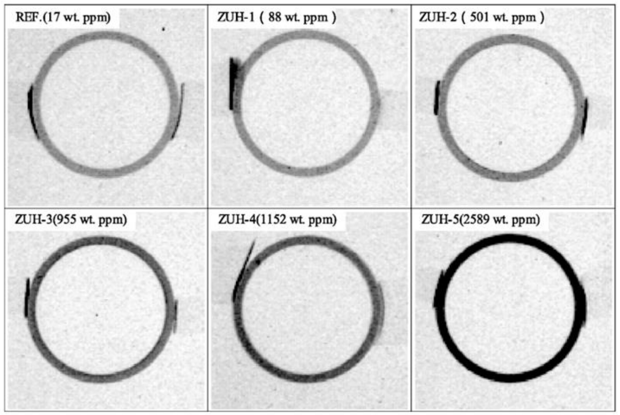
MRP Microreact Program

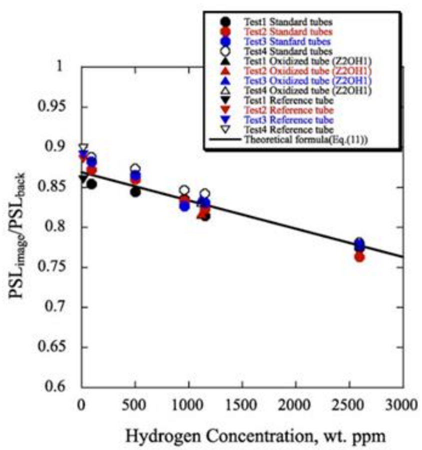
Previous Works of Neutron Imaging to Probe H-concentrations

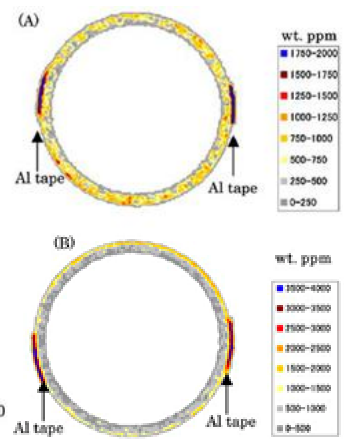
Most Neutron imaging measurements to date have looked at H-concentrations in Zirconium alloys.

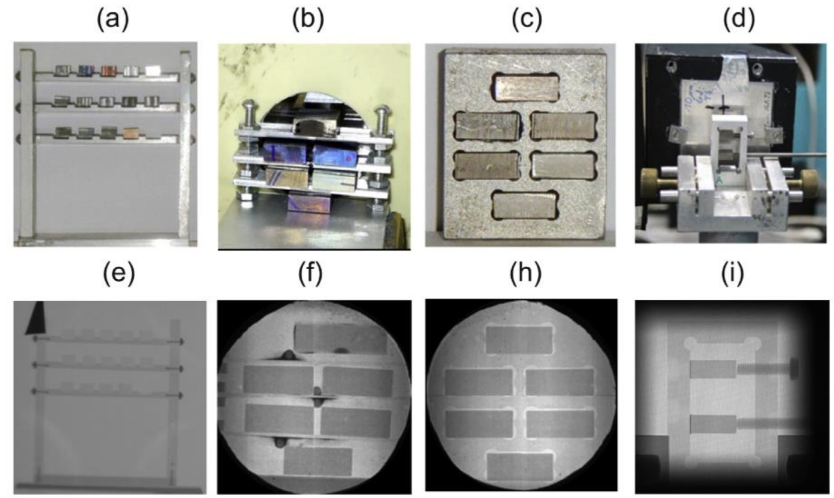
Measure — Determine — Calibrate/Model

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

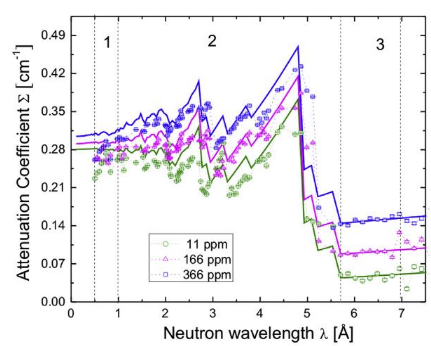


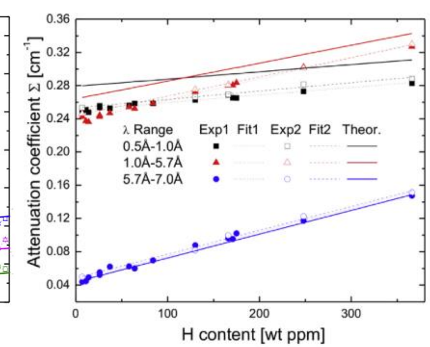






If neutron source is pulsed, then **Energy Resolved Neutron Imaging** (ERNI) measurements can be performed. ERNI measurements have been shown not need calibration sample and to resolve H-concentrations down to 5ppm %wt.





Our work aims to set up similar capabilities at LANSCE to measure H-concentrations in YH metals, both at ambient and extreme (applied temperature gradient) conditions.

R. Yasuda et. al. Journal of Nuclear Materials 320 (2003) 223-230
N.L Buitrago et. al. Journal of Nuclear Materials 503 (2018) 98-109

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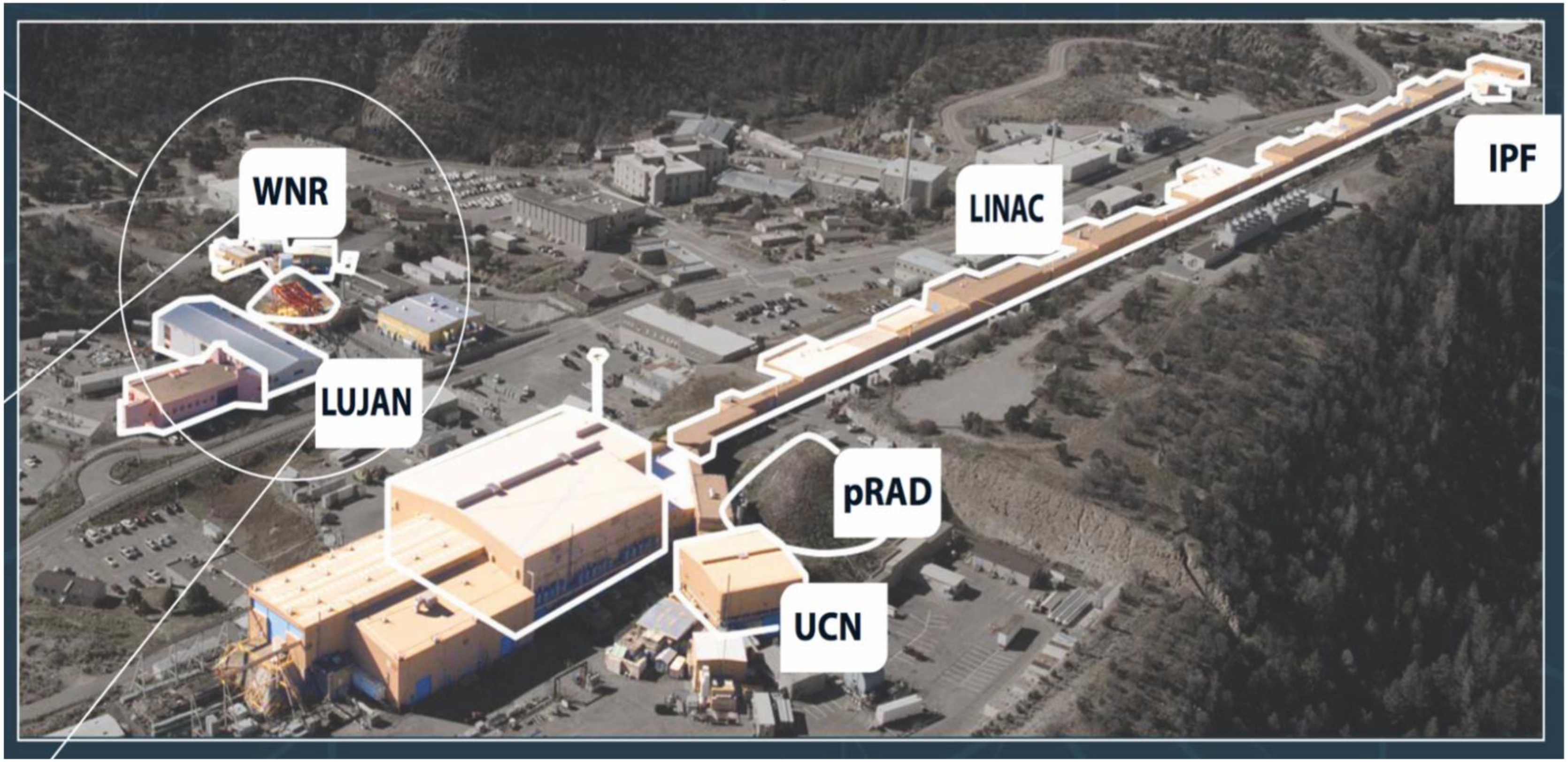
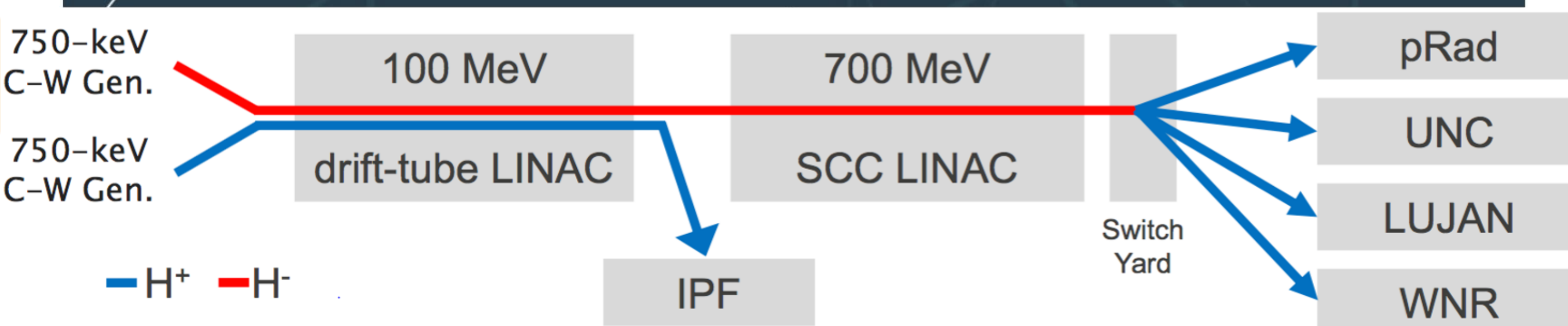
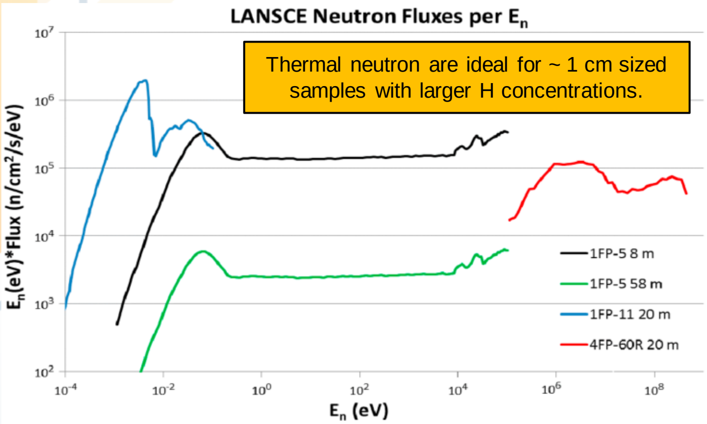
The Los Alamos Neutron Science Center (LANSCE)





Measuring H-Concentrations on Flight Path 5 at LANSCE

FP5 views pulsed thermal neutrons from the 1L target

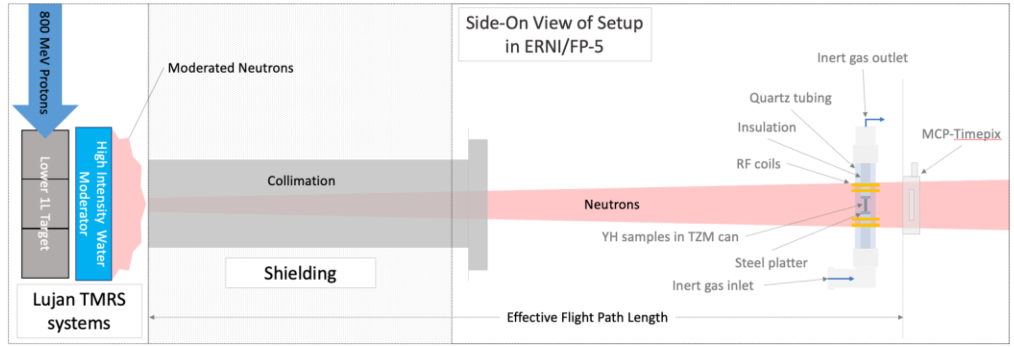


Goals and Approach

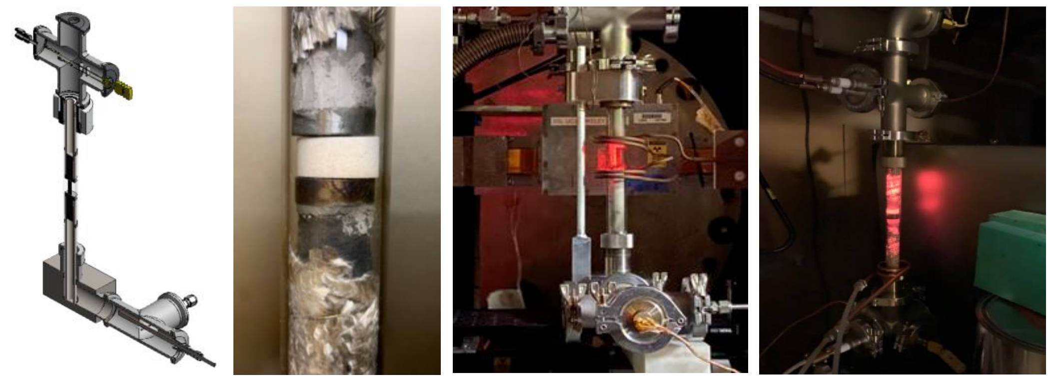
Measure H-Diffusion coefficients at various conditions (stoichiometry, temperatures, phase fractions) in YH samples.

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

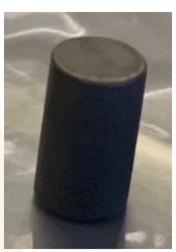
General neutron imaging setup of FP5

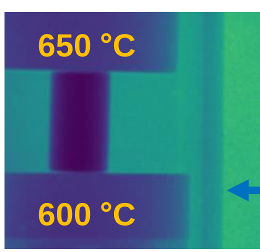


Compact dual zone furnace for in-situ heating



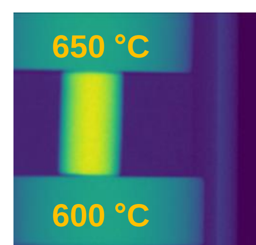
YH$_{2\text{-}x}$ samples fabricated @ LANL





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)

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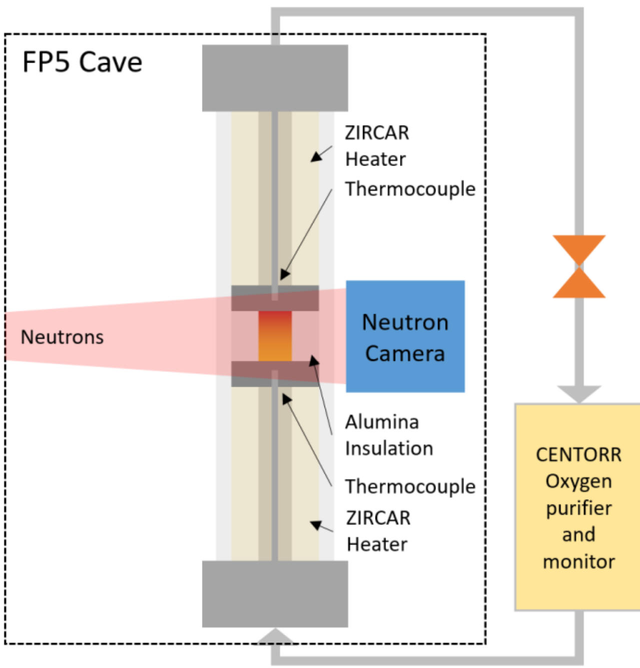
2021 in-Situ YH Measurements with Temp Gradient

Neutron Imaging measurements were performed on FP5 during Nov. 2021

- 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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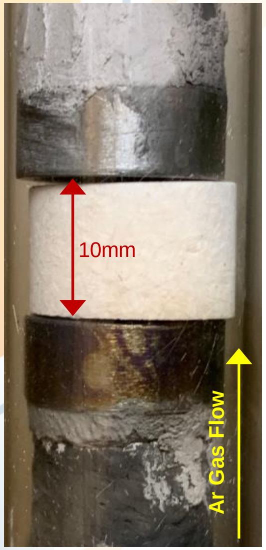
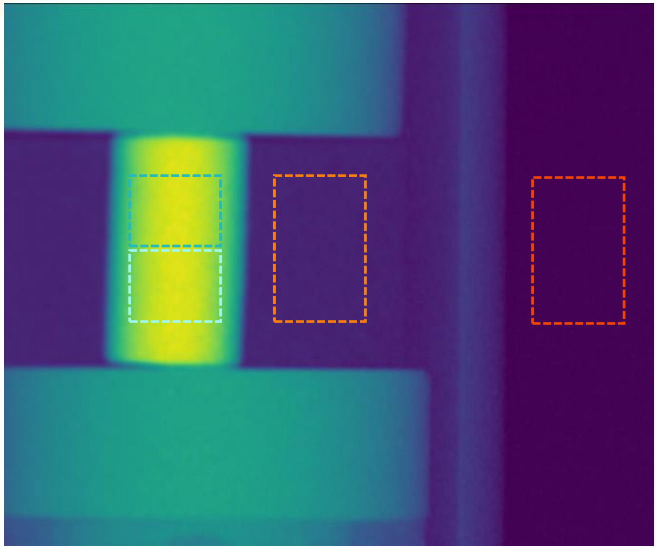
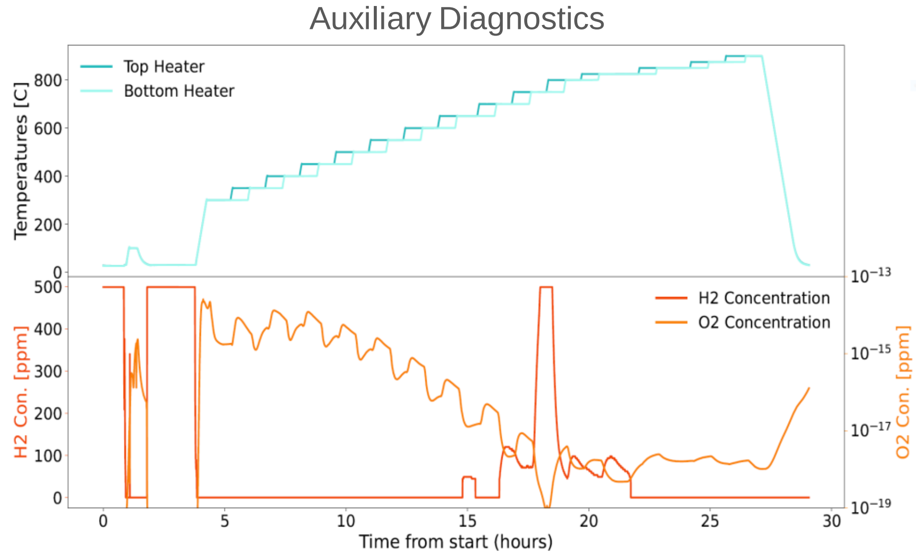
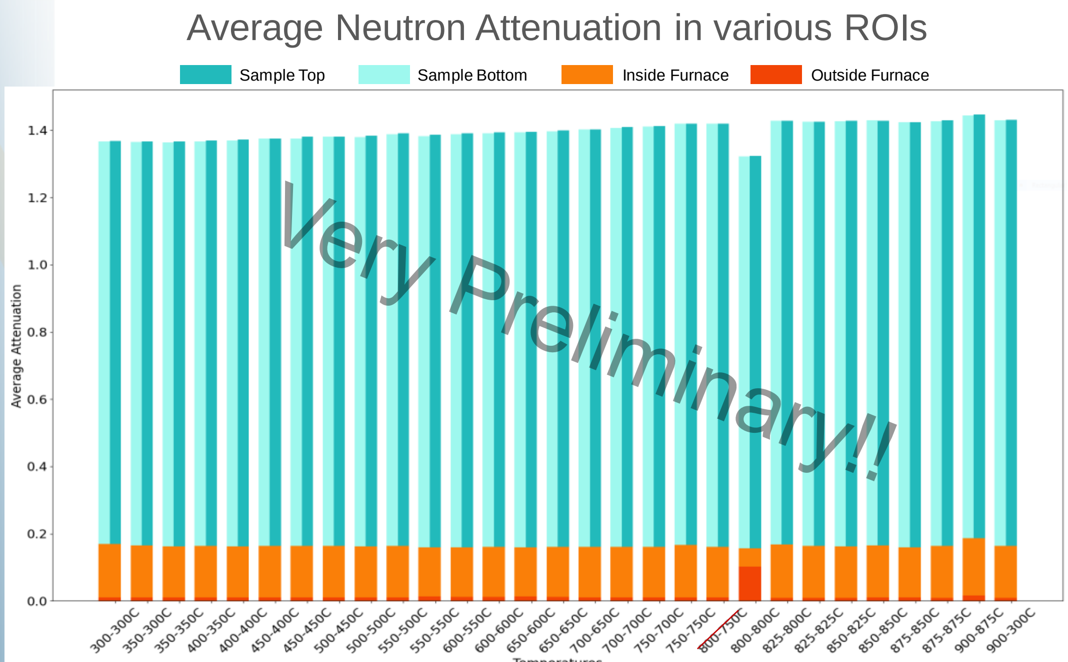
2021 in-Situ YH Measurements with Temp Gradient









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.

A more thorough analysis needs to be performed to investigate and interoperate results…

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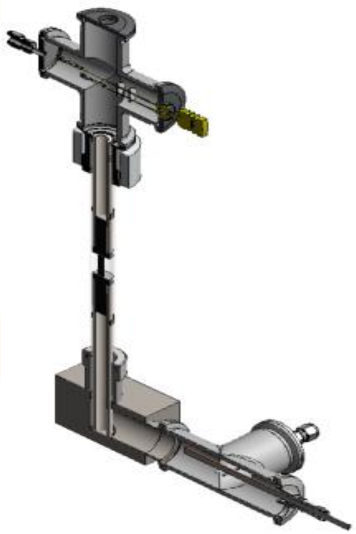
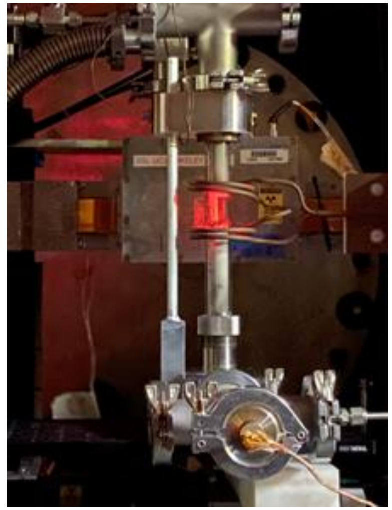
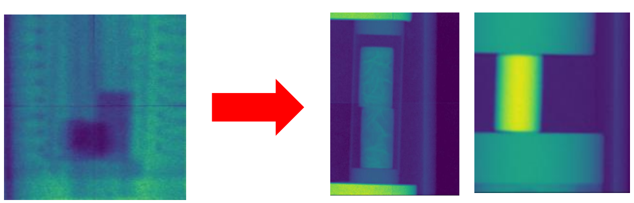
Looking forward…

1st Generation Compact Dual-Zone Furnace



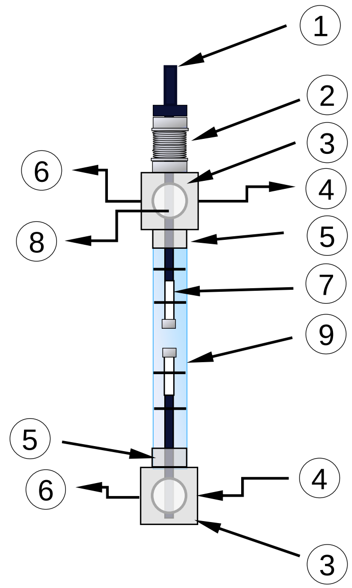


- Operated on FP5 in ‘20 and ‘21 run cycles.
- Suffered from delayed parts.
- Allowed for higher resolution imaging with screen to sample distances of ~2-3 inches.



- 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

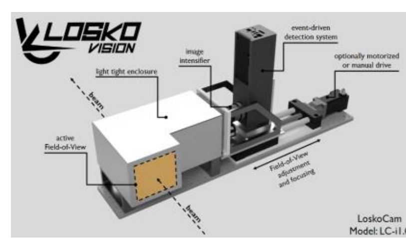


1. Sample stick
2. Linear motion device (bellows)
3. 6-way cube (UHV)
4. Flow gas
5. Glass-to-metal seal
6. Thermometry & electrical feedthrough
7. Resistive heating
8. Vacuum line
9. Glass tube

- 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.

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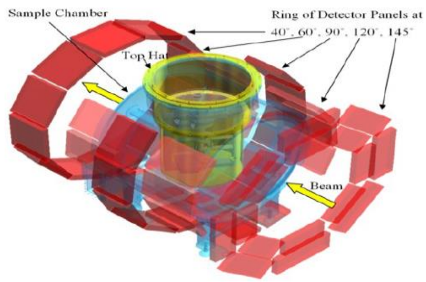
Multi-modal Pre- and Post-Hydride Characterization

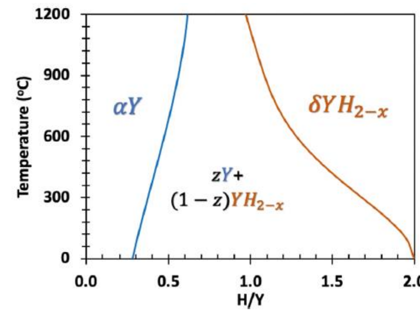
Neutron Diffraction on FP4/HIPPO

Neutron CT on FP5/ERNI

Samples fabricated in house @ LANL
- Direct Hydriding
- Powder Met.
- Initial bulk stoichiometry measured.

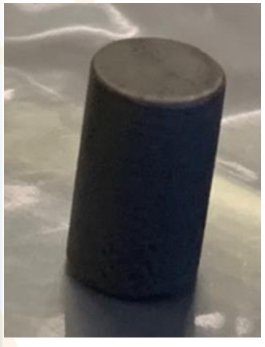
Phase fraction and microstructure characterized

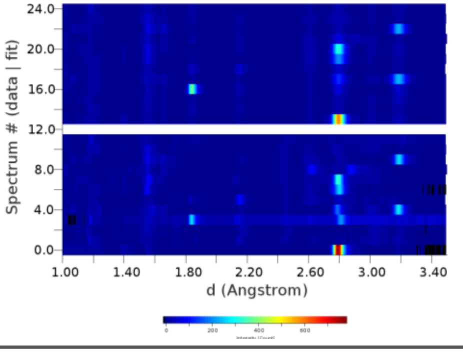




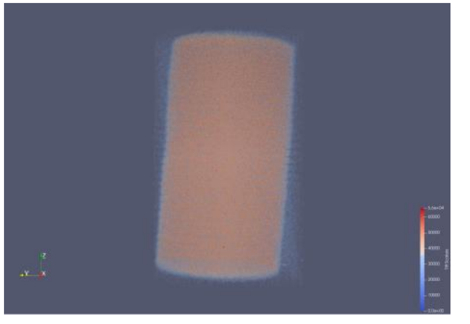
H-distribution characterized

Sample 157-1

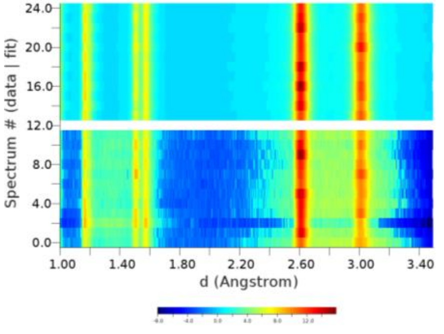




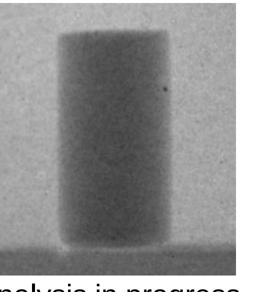
Diffraction Results:
- H/Y < 0.5
- mostly α-Y
- α-Y almost one single crystal
- Only a few grain boundaries.



Sample 176-1



Diffraction Results:
- H/Y > 1.9
- No detectable preferred orientation in YH_2 phase
- Hydride process may have randomized texture



Analysis in progress

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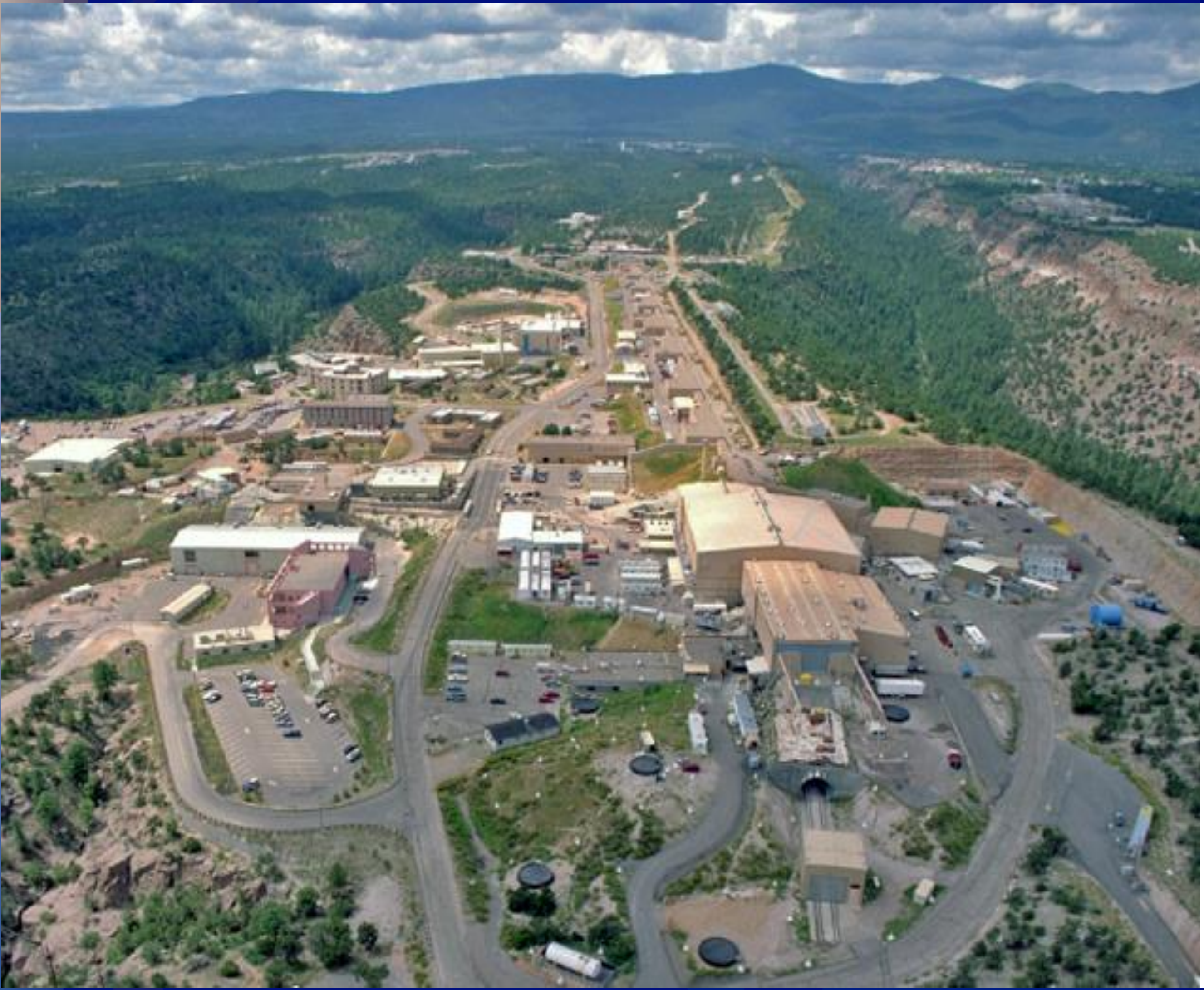

Thank you!

LANL Collaborators

Holly Trellue (NEN-5)
Travis Carver (MST-8)
Alexander Long (MST-8)
Erik Luther (SIGMA-2)
Vedant Mehta (NEN-5)
Caitlin Taylor (MST-8)
James Torres (MST-8)
Aditya Shivprasad (MST-8)
and Sven Vogel (MST-8)

Please feel free to contact us if you have any additional questions!

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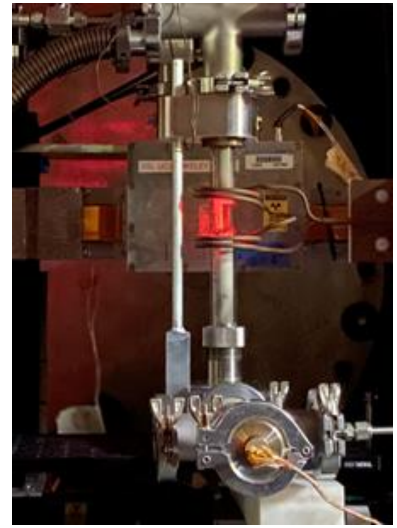
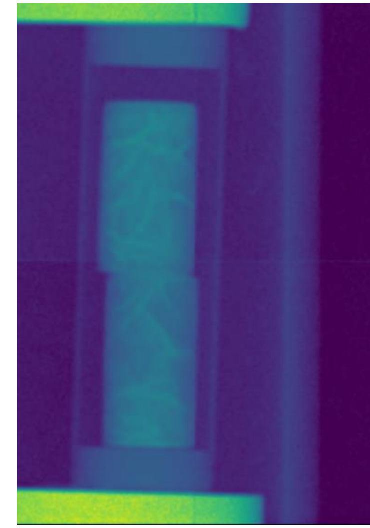
Backup: First in-situ YH Measurements at Elevated Temps

First measurements with new Compact Dual Zone (CDZ) furnace performed on FP5 in Dec 2020.

Measuring $YH_{1.2}$ and $YH_{1.7}$ @ Elevated Temps

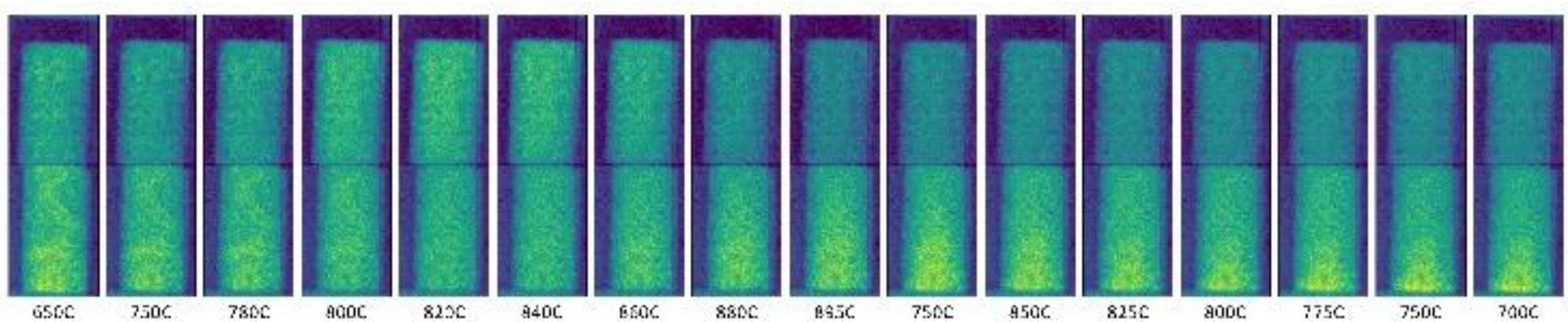
- Samples placement at ~ 1.5”
- Original designed with two heating elements:
  - Each element can go up to 1000C
  - Capable of inducing 100 C temperature gradient.
  - Additional RF configuration.
- Uses flowing Ar cover gas.
- Thermocouples were attached to each steel platen (top and bottom)
- Heating elements failed. Used RF config.
- Two YH samples in a TZM can
- No temp gradients with RF
- Each temperature was held for ~3 hours during which roughly six 30 min exposures were taken.
- All radiographs were integrated over thermal neutron energies





$YH_{1.19}$*

$YH_{1.70}$*



Demonstrated the capability to observe hydrogen redistribute in yttrium hydride over cm length scales in-situ at high temperatures.
“Effects of Hydrogen Redistribution at High Temperatures in Yttrium Hydride Moderator Material”, H. Trellue et. al. JOM (2021)

Lower sample appears to accumulate H during cooling, though because of the TZM can and Ar gas flow temperature profiles were not spatially characterized.

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Backups: 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.

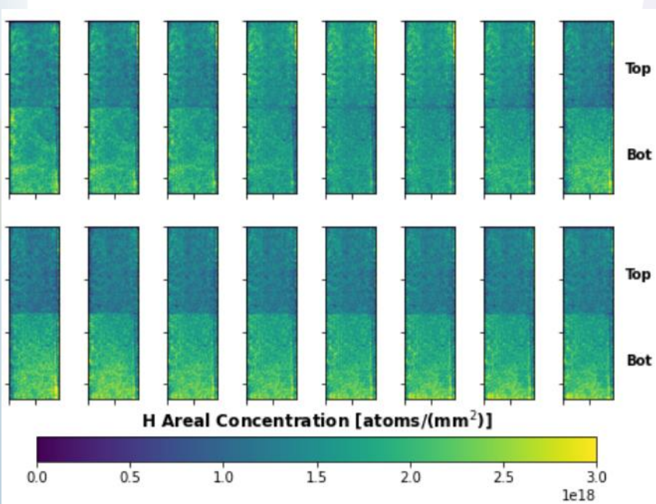
Sometimes there maybe no calibration samples that can be used or they are less than ideal…

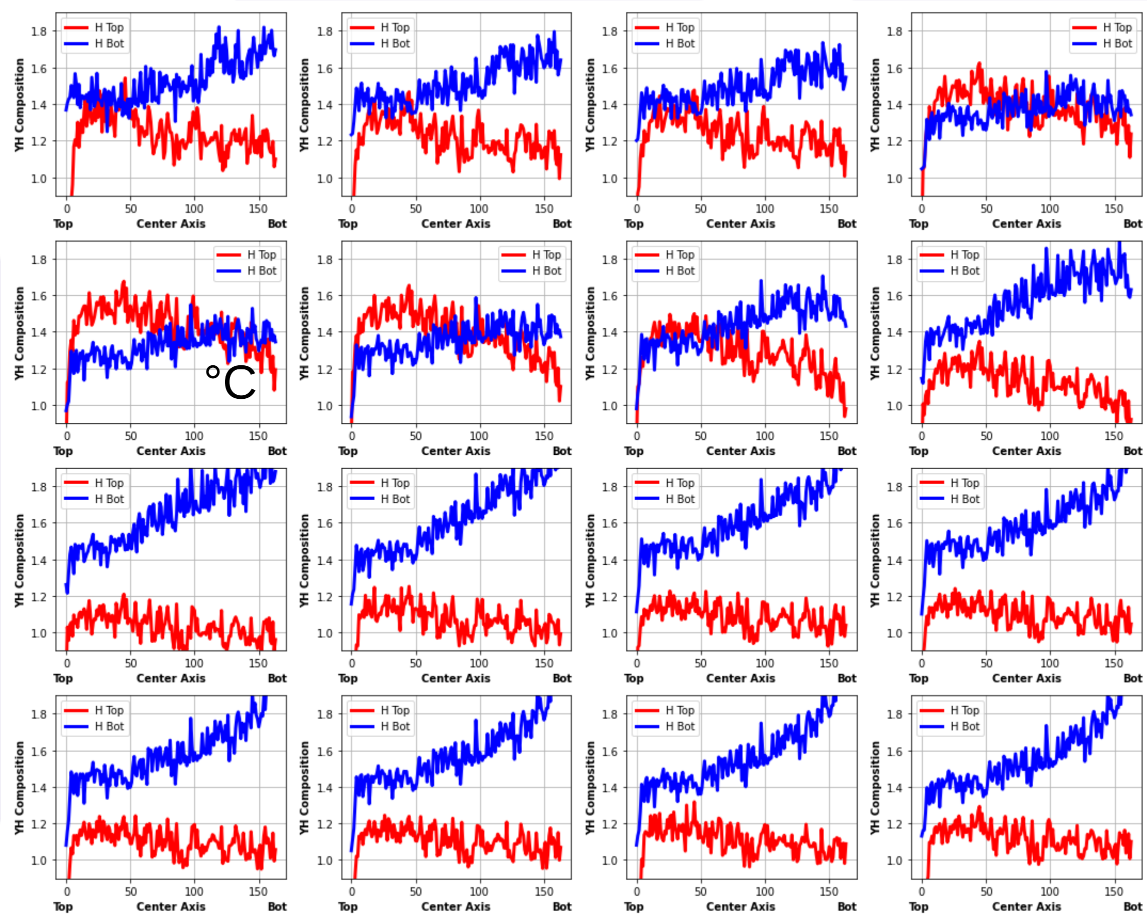
Using solvers along with constraints, several unknowns can be determined.

$$h_{ij}H_{cs} + yY_{cs} = p_{ij}$$

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

- 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 $\equiv 82.3 \times 10^{-22}$ mm^2 / atom]
- Y_{cs}: Yttrium cross-section for neutrons (known constant) [9 barns $\equiv 9 \times 10^{-22}$ 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)

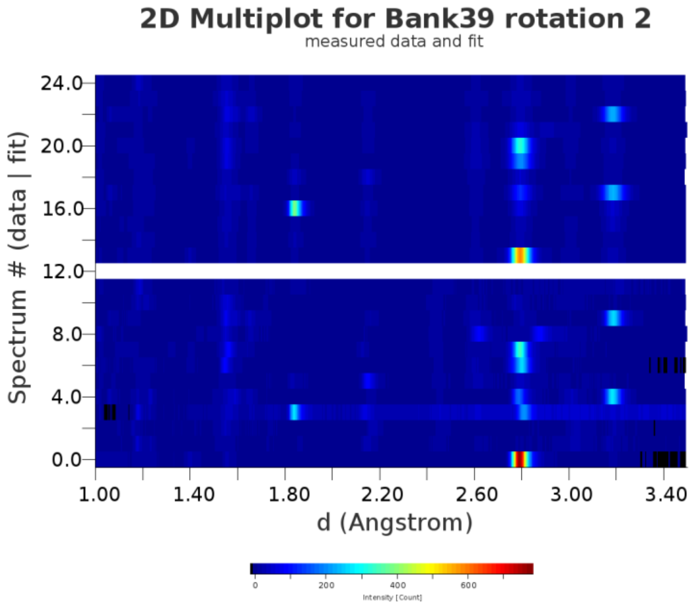




*Will be verifying analysis with water plate samples.

Backups: Diffraction results on YH samples in High-Pressure Preferred Orientation (HIPPO instrument)

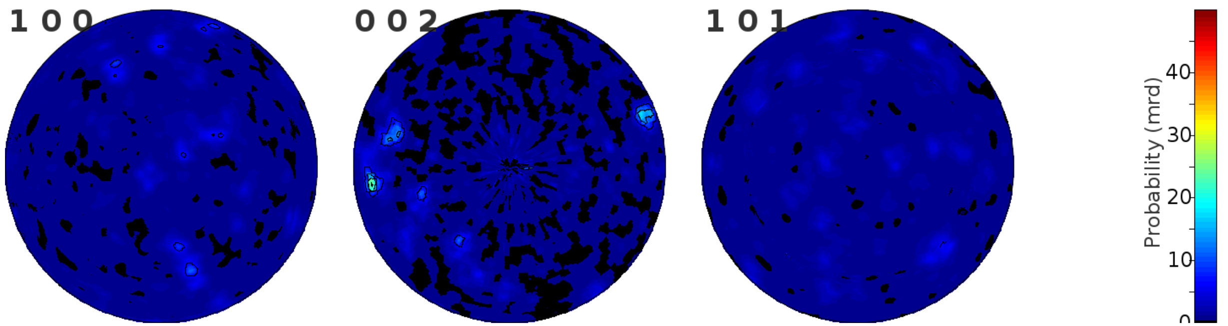
- YH was loaded in 6mm vanadium cans
  - 0, 67.5, 90 degree rotations on
  - HIPPO@20min/rot
  - E-WIMV representation of ODF, 7.5 degree resolution expect for α-Y in 157-1 which was 2.5 degree
- Very strong texture visible in raw data, peaks from both α-Y and δ-YH2



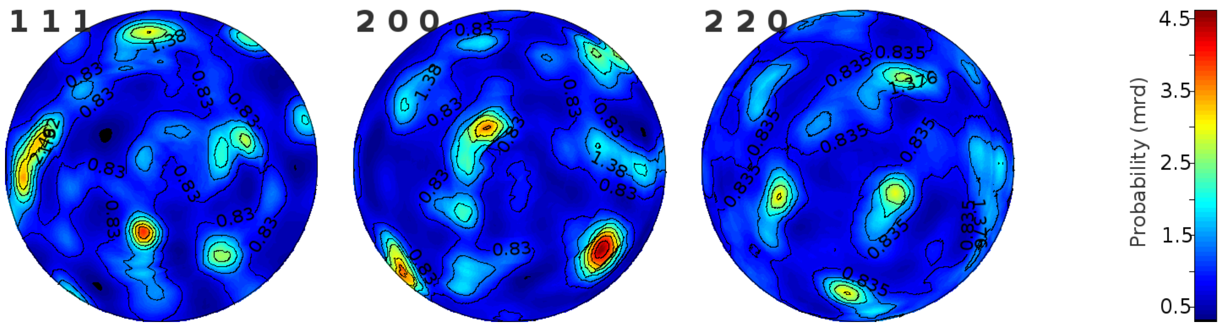
Backups: Diffraction results on YH samples in High-Pressure Preferred Orientation (HIPPO instrument)

157-1

- Alpha-Y
  94.1 wt%
  almost
  sxtal



- YH2
  5.9 wt.%



- Note different mrd scales