LA-UR-20-21710
Approved for public release; distribution is unlimited.

Title: High temperature moderator material for Microreactors

Author(s): Shivprasad, Aditya Prahlad; Luther, Erik Paul; Long, Alexander Makenzie; Cutler, Theresa Elizabeth; Grove, Travis Justin; Mehta, Vedant Kiritkumar; Cooper, Michael William Donald; White, Joshua Taylor; Saleh, Tarik A.; Wermer, Joseph Raymond; Trellue, Holly Renee

Intended for: internal presentation

Issued: 2020-03-03 (rev.1)

Disclaimer:
Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by Triad National Security, LLC for the National Nuclear Security Administration of U.S. Department of Energy under contract 89233218CNA000001. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

High temperature moderator material

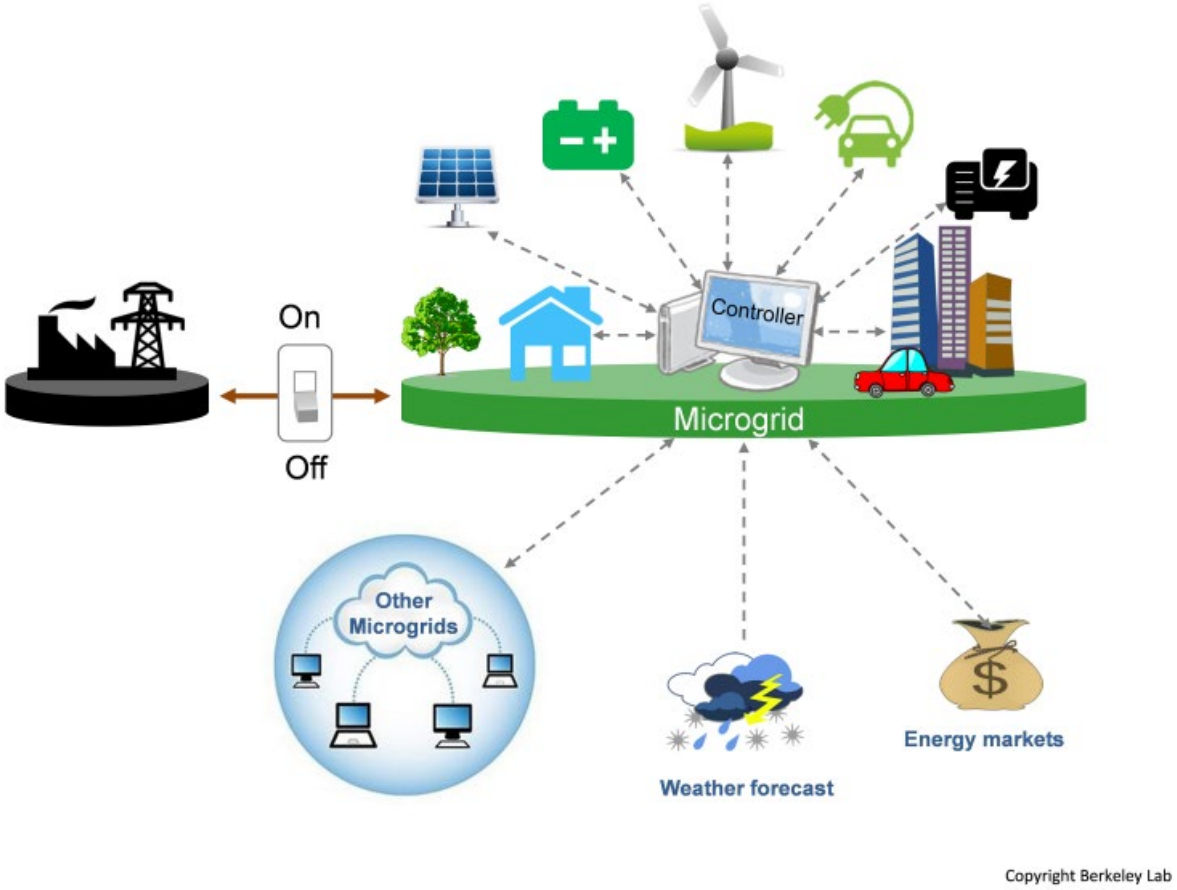
A.P. Shivprasad, E.P. Luther, A.M Long, T.E. Cutler, T.J. Grove, V.K. Mehta, M.W.D. Cooper, J.T. White, T.A. Saleh, J.R. Wermer, H.R. Trellue

LA-UR-20-21710

02/25/2020

Managed by Triad National Security, LLC for the U.S. Department of Energy's NNSA

Microreactors could be used to address power needs of microgrids



Sendai microgrid - Japan

Huatacondo village - Chile

building-microgrid.lbl.gov

nedo.go.jp

Yttrium dihydride (YH_{2-x}) is a promising candidate for moderator applications

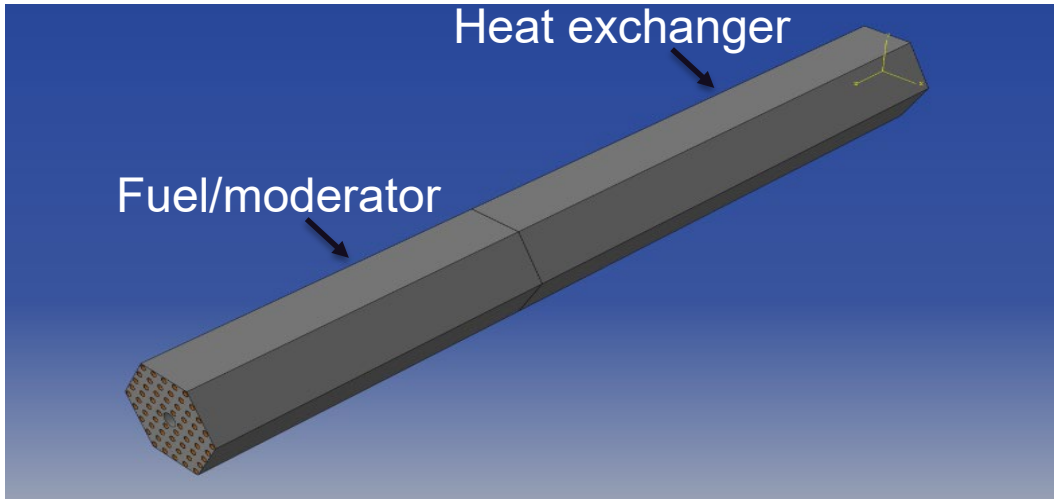
- Why use a moderator?
  - Moderator improves economics of the reactor
  - Used in lieu of large amounts of high-assay LEU

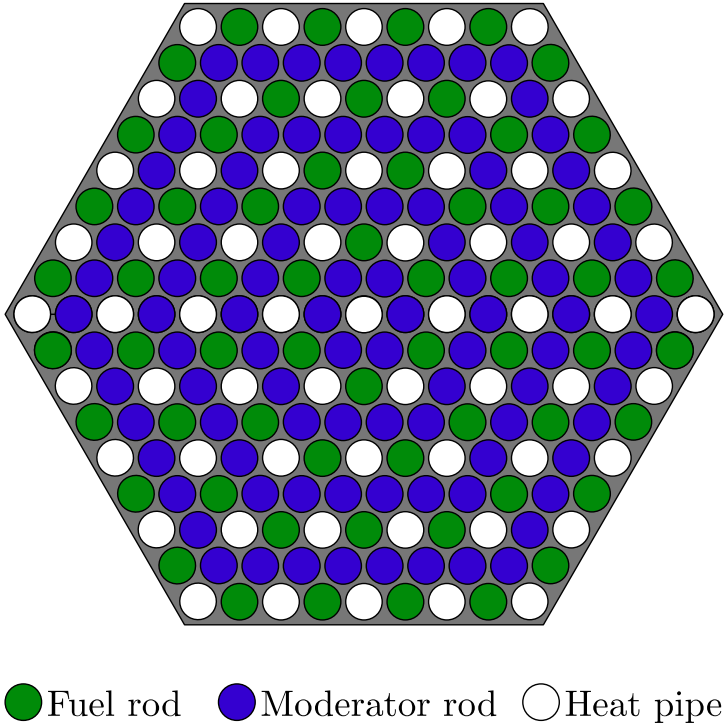
- Why YH_{2-x} over traditionally-used moderators?
  - High thermal stability compared to other metal hydrides
  - Relatively low thermal neutron absorption cross section
  - Good elastic properties for a metal hydride

- Challenges associated with YH_{2-x} fabrication
  - Near net-shape parts are difficult to achieve for complex geometries
  - Massive hydriding may result in structural degradation

Heat pipes enable simple reactor core designs

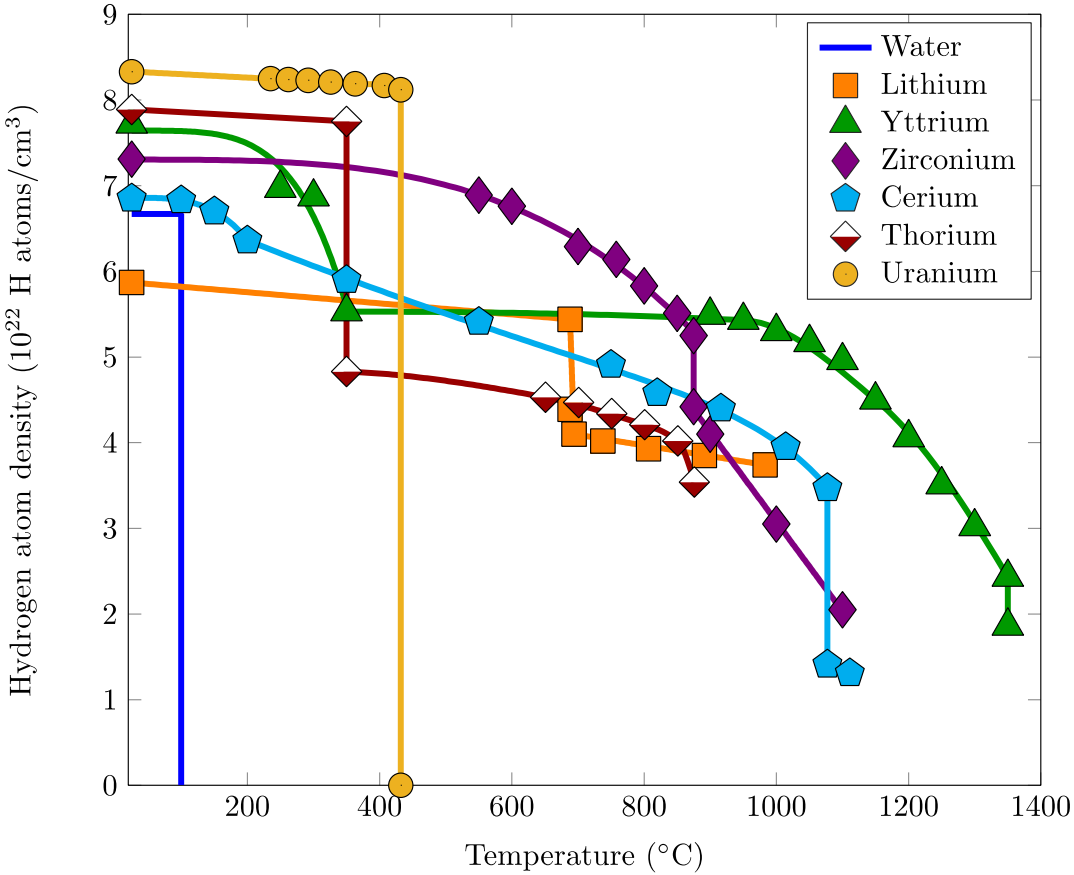
- Systems of 0.1-20 MWt using heatpipe technology offer potential.
- Heat pipes, fuel rods, and/or moderator are intermixed in an assembly.
- Fuel and/or moderator would exist in the bottom half and heat exchangers in the top.





Yttrium dihydride is a promising candidate hydrogenous moderator

Metal hydrides and water in equilibrium with 1 atm of 100% H_2

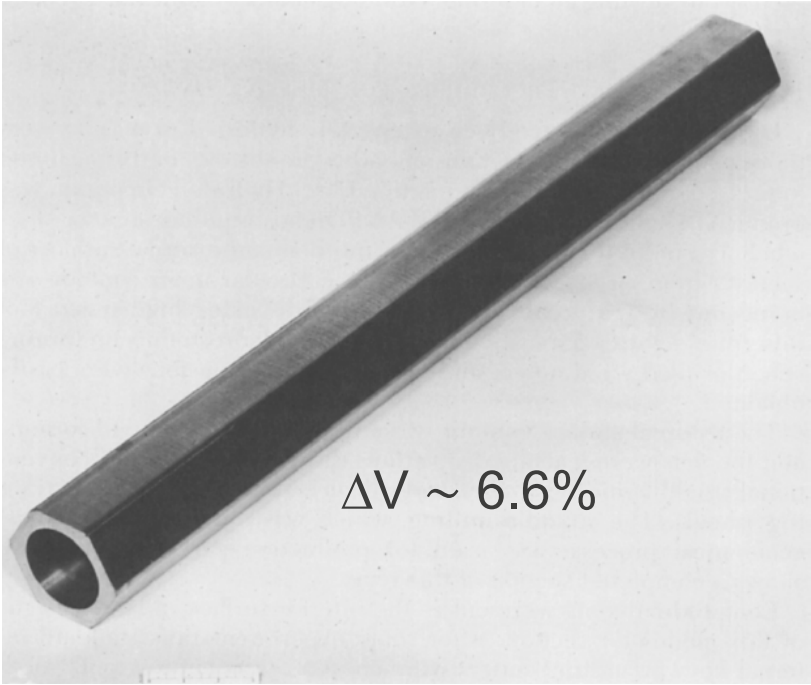


Element -	Thermal (n,γ) σ (b)
Zr-90	0.010
Y-89	1.278
Li-7	0.045
Ce-140	0.577

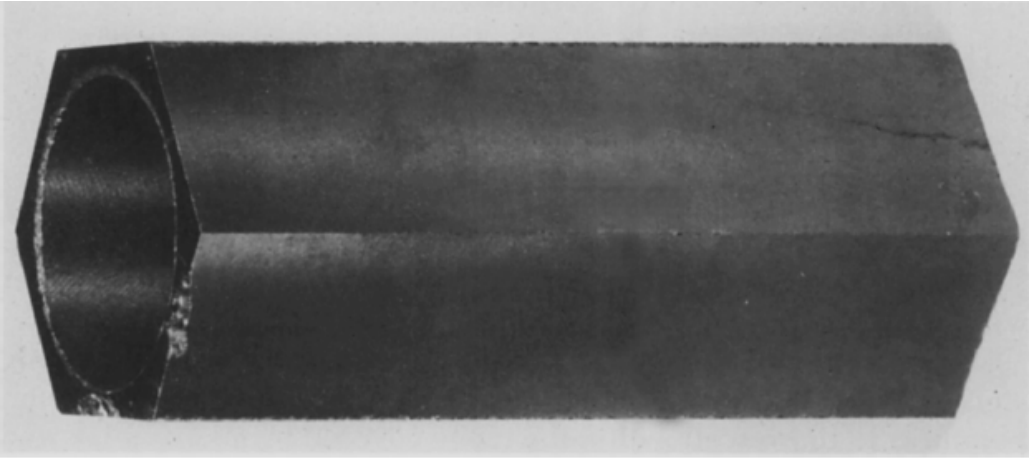
Shivprasad, et al. 2020
atom.kaeri.kr

Metal hydrides for moderator applications is not a new concept

Machined section of ZrH_{1-x}



Machined section of YH_{2-x}



$\Delta V \sim 4.5\%$

Mueller, 1968

Not all yttrium hydride is created equal

YH_{2-x}

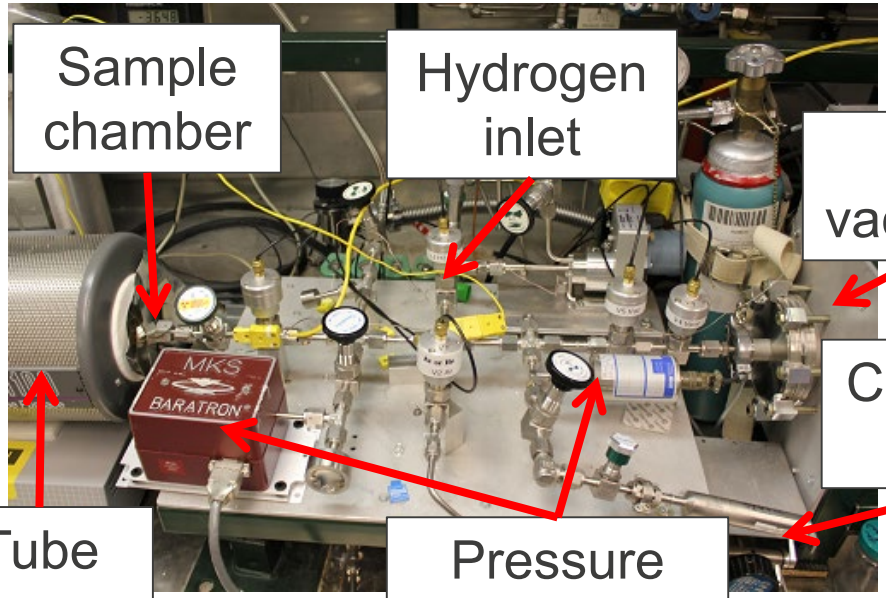
YH_{3-x}

YH_{2-x} is able to retain its shape, while YH_{3-x} causes rapid pulverization on formation

Luther, 2017

The traditional method of making yttrium dihydride is using the direct-hydriding technique

Slow addition of hydrogen is done at temperature by using many small aliquots over approximately 1 week in a Sievert's apparatus



2" diameter YH_{2-x} cylinders

Shivprasad, et al. 2018

Luther, 2017

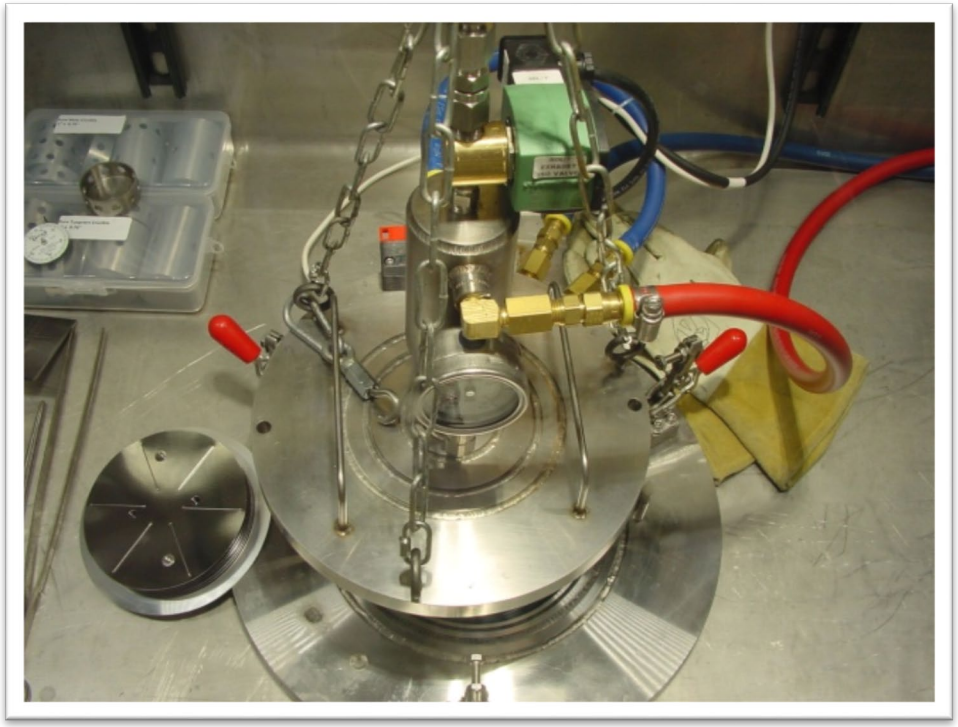
Massive hydrides were crushed and pressed into compacts for sintering

YH_{2-x}

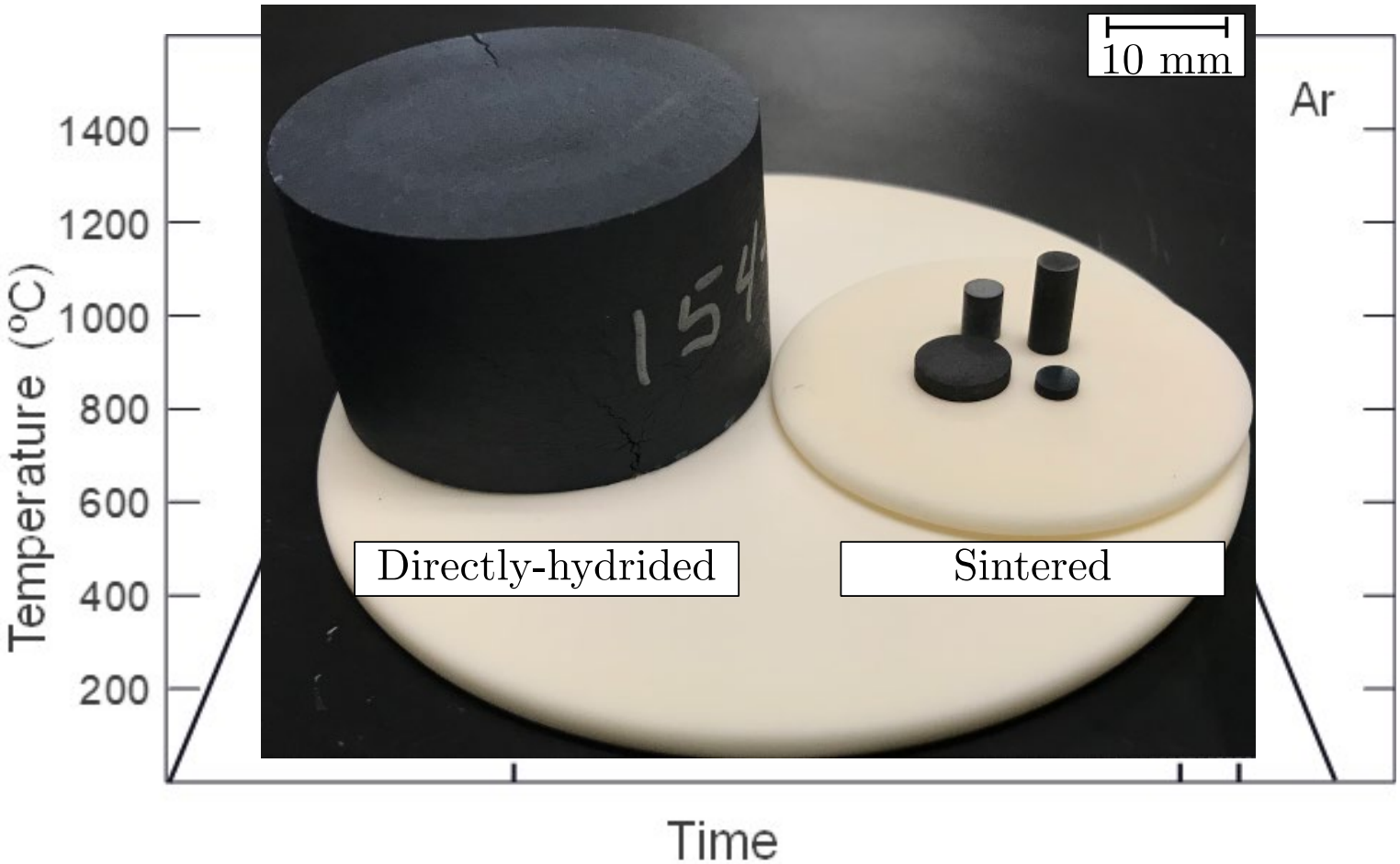
YH_{2-x} powder ball-milled

Green pellets of YH_{2-x}

Sintered in glovebox metal furnace

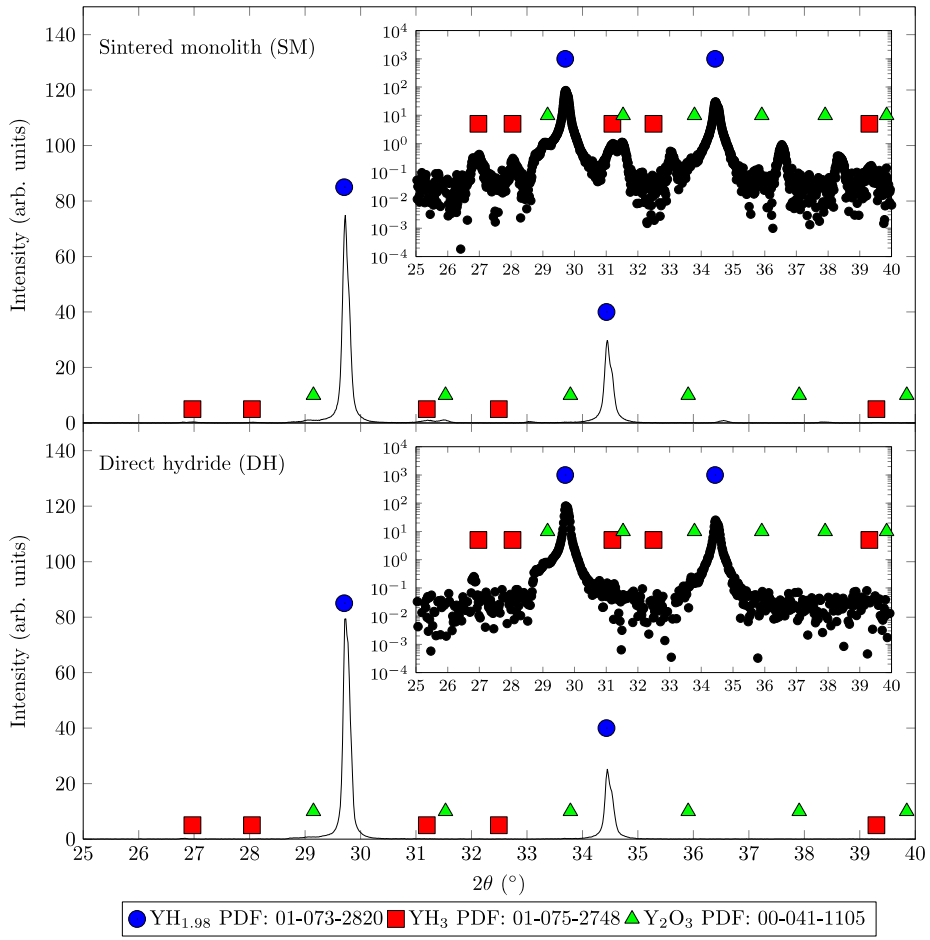


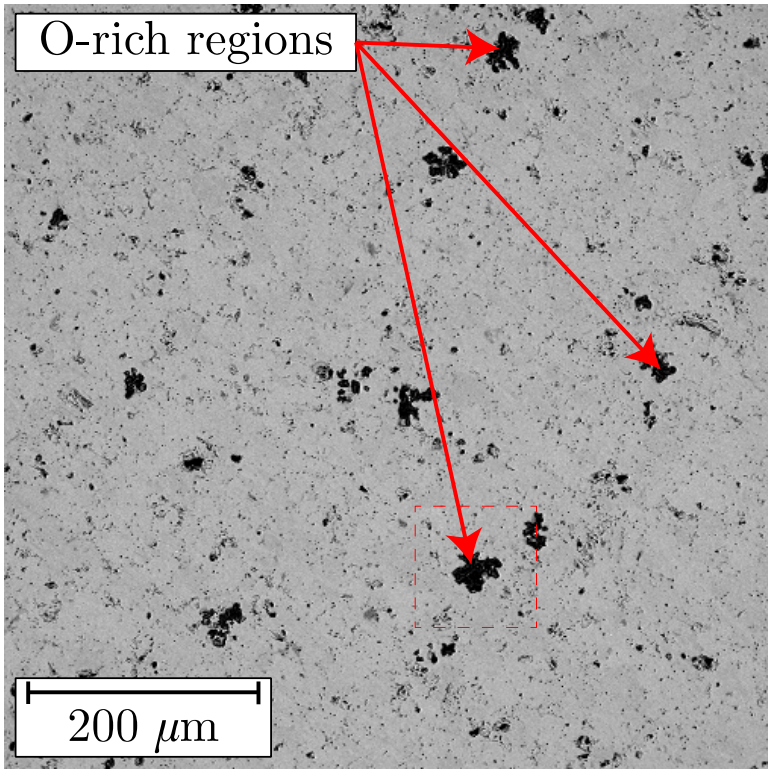
Powder metallurgy process for yttrium dihydride invented at LANL shows promising results



Shivprasad, et al. 2020

Secondary phases may have formed during sample preparation

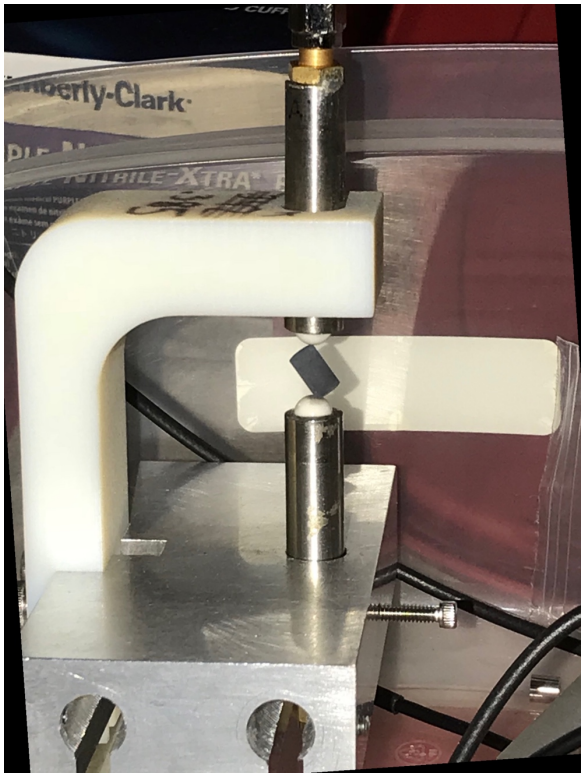




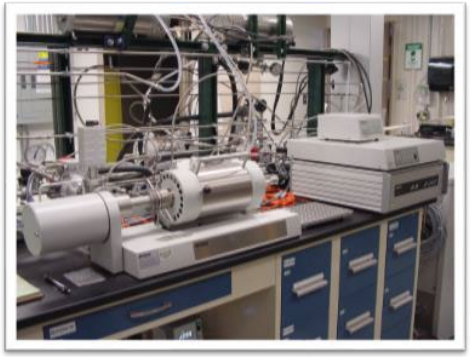
Shivprasad, et al. 2020

Elastic and thermophysical properties were measured for sintered pellets

Elastic moduli -
Resonant Ultrasound Spectroscopy

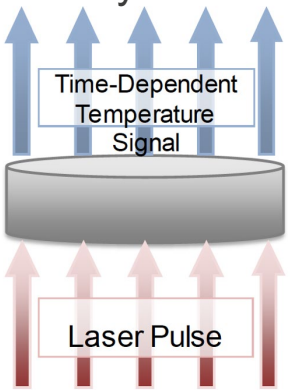


Coefficient of Thermal Expansion –
Dilatometry



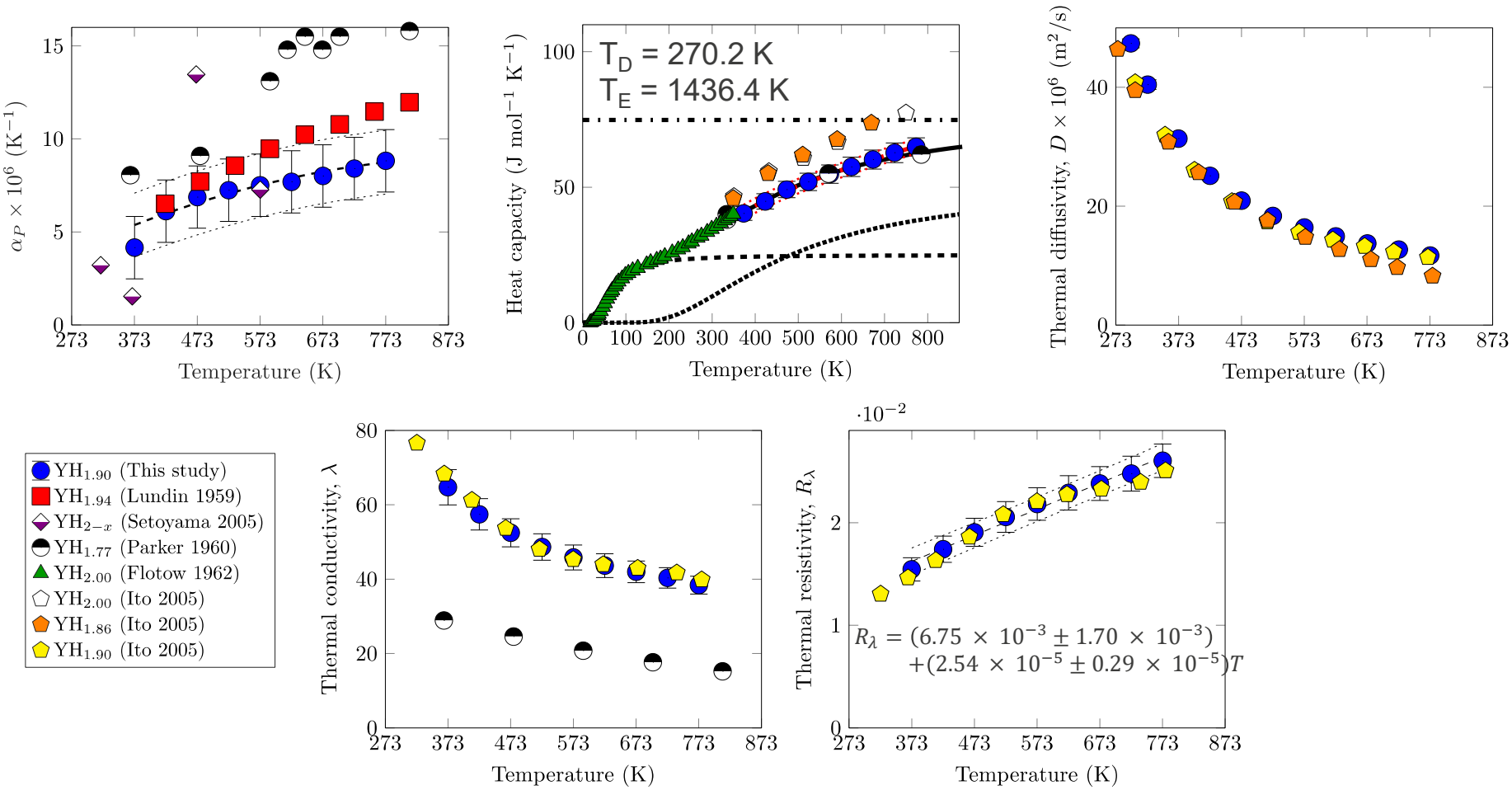
Heat capacity –
Differential Scanning Calorimetry

Thermal diffusivity -
Laser Flash Analysis



Shivprasad, et al. 2019

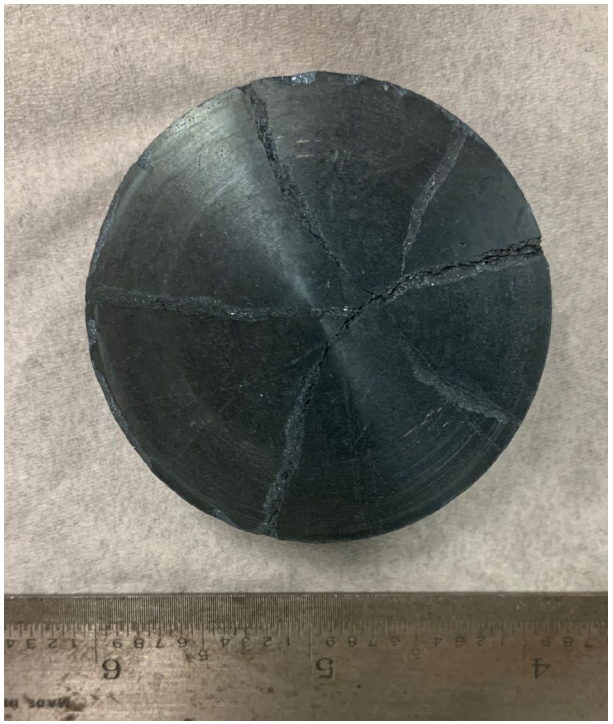
Thermophysical properties were also consistent with literature

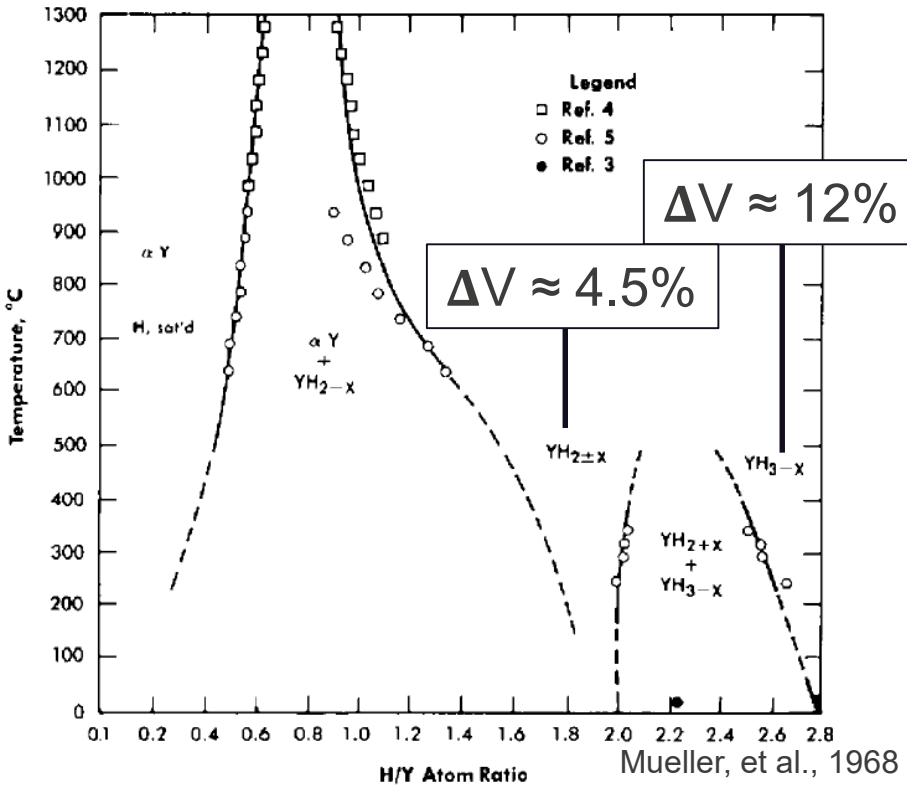


Shivprasad, et al. 2019

Fabrication of massive YH_{2-x} monoliths has technical challenges

YH_{2-x} with severe cracking



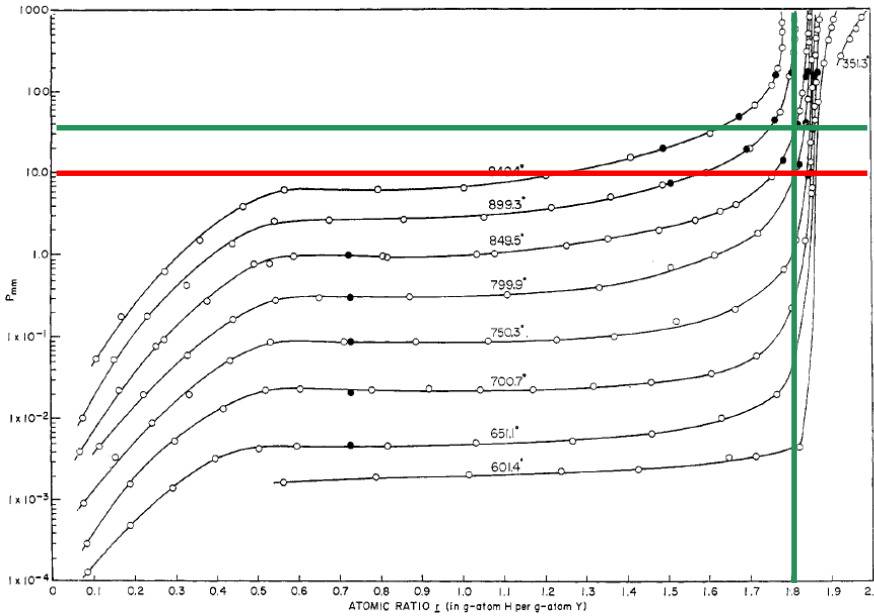


Mueller, et al., 1968

Challenges in volume expansion and tight chemistry control promote interest in alternate fabrication pathways

YH_2 Monolith Preparation

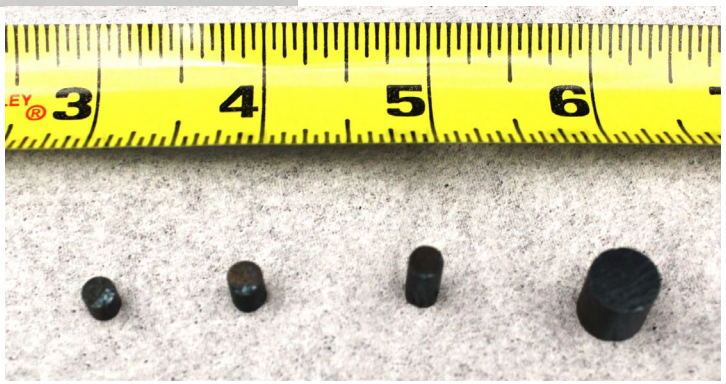
- Yttrium hydride ($YH_{1.9}$, 21.1 g hydrogen/kg $YH_{1.9}$) prepared by direct reaction of yttrium metal with hydrogen gas at 820°C +/-5˚C
  - Slow addition of hydrogen using many small aliquots over time
    - <15 torr
    - Small sample e.g. 0.5” cylinder 1 day
    - Large sample e.g. 2” disk 6-8 days (60-80 aliquots)
  - Exothermic
  - Molybdenum foils or boats were used to prevent contact of yttrium metal with quartz



Yttrium hydride by direct hydriding

2” diameter YH_2 cylinders

1” diameter cylinder machined dry



Cylinders machined by EDM

- Machining generally successful conventionally wet/dry or by EDM
  - Residual stresses limit size to > 0.10”

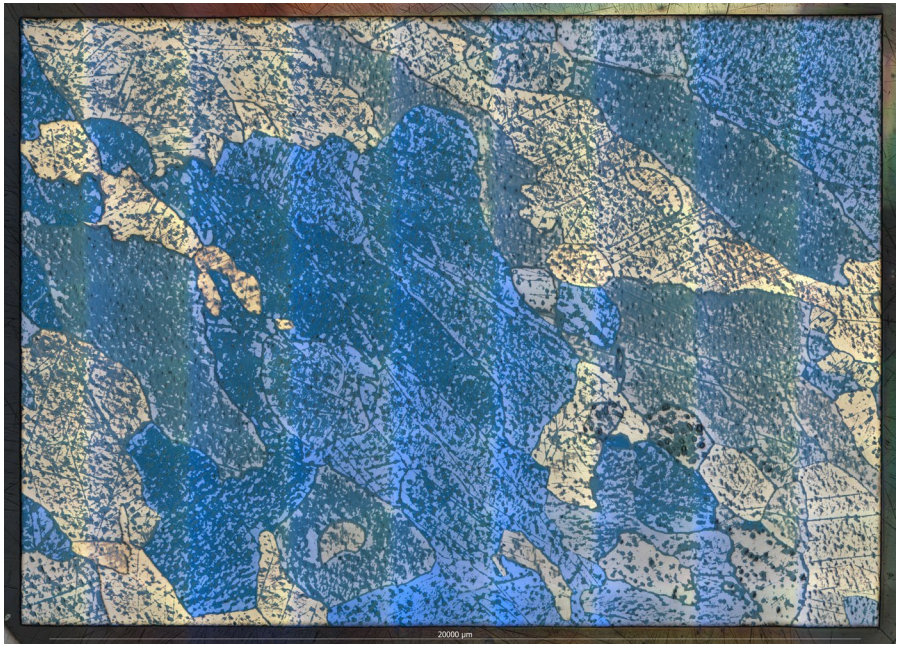
YH_2 Monolith Preparation

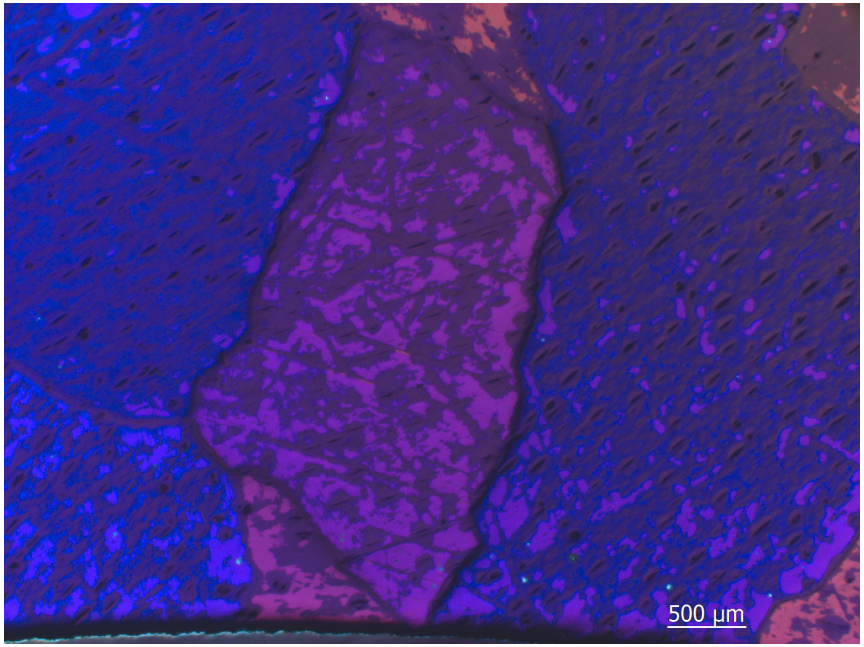
- Yttrium from legacy stock (~1960s)
  - High purity stock
  - Large as-cast grains result in large residual stresses
- Kinetics depend on source material
  - Studies in progress
  - 99.5% vs 99.99% purities

Element (-)	Composition (wt. ppm)	Standard deviation (wt. ppm)
Zr	4837	4612
O	2900	1380
Ta	807	566
N + C	275	64
Fe	213	35
Transition metals (excl. Fe)	197	53
Alkaline earth metals	147	86
Refractory metals (excl. Zr and Ta)	120	46
Alkali Metals	100	46
Lanthanides	90	48
Non-metals (excl. O, C, N)	50	0
Other metals	33	25

Microstructure

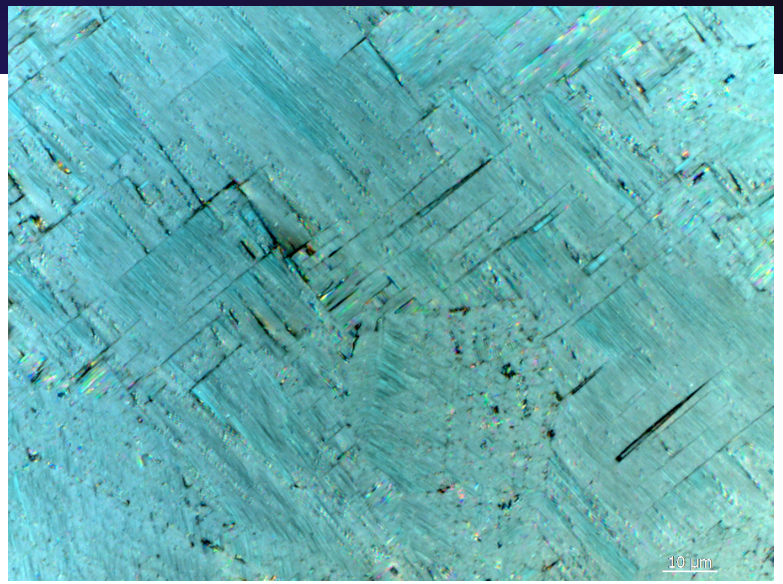
- Y ingot
  - HCP
  - Millimeter size, aniostropic grains
  - Elongated due to solidification front

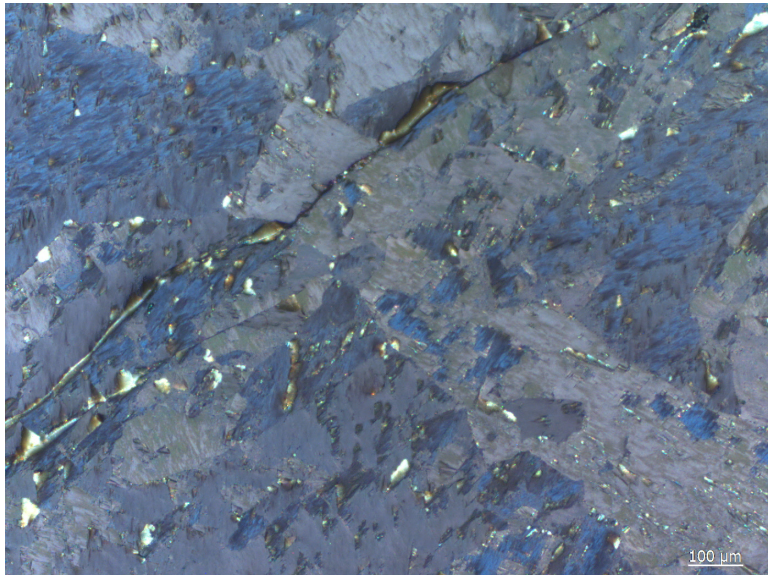




Microstructure

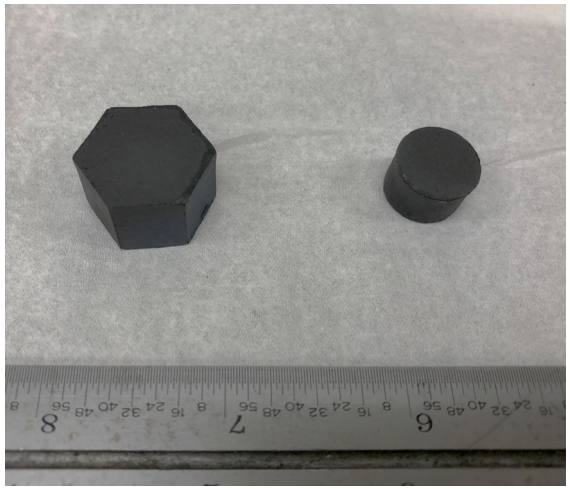
- YH_2
  - FCC (~4.5% expansion)
  - “Herringbone” pattern
  - Significant cracking
    - Large and micro cracking evident

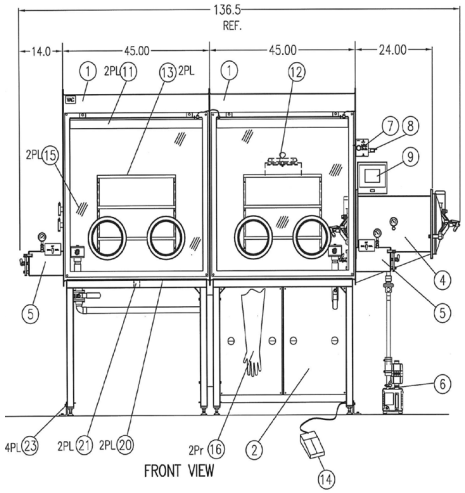




YH_2 Fabrication Capability/Research

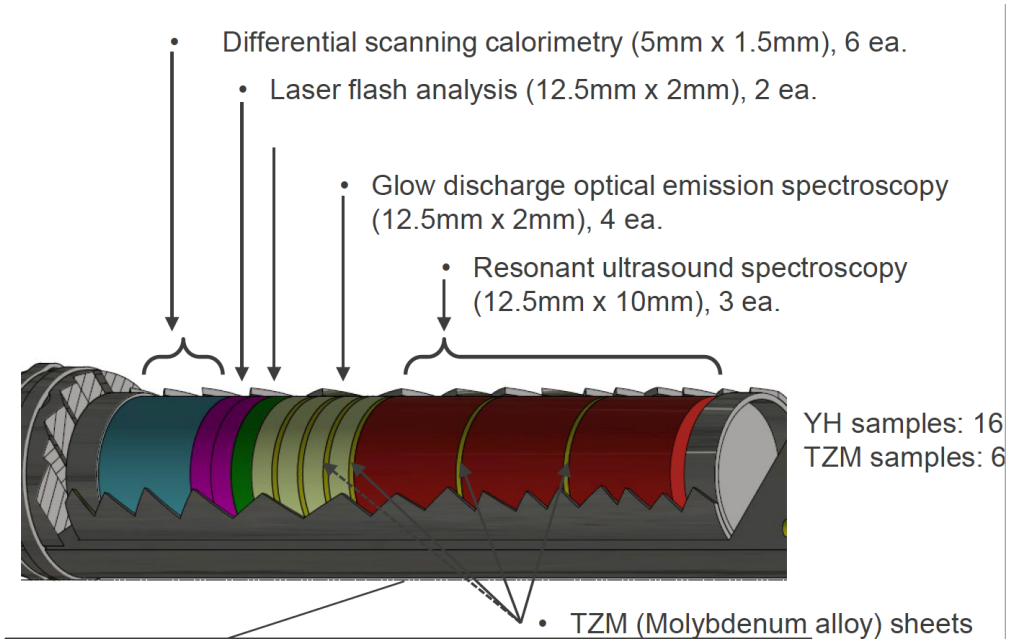
- Establish YH_2 fabrication capability
- Kgs of YH_2
- Relevant sizes
- Support experimental research for LANSCE, ATR, and NCERC measurements





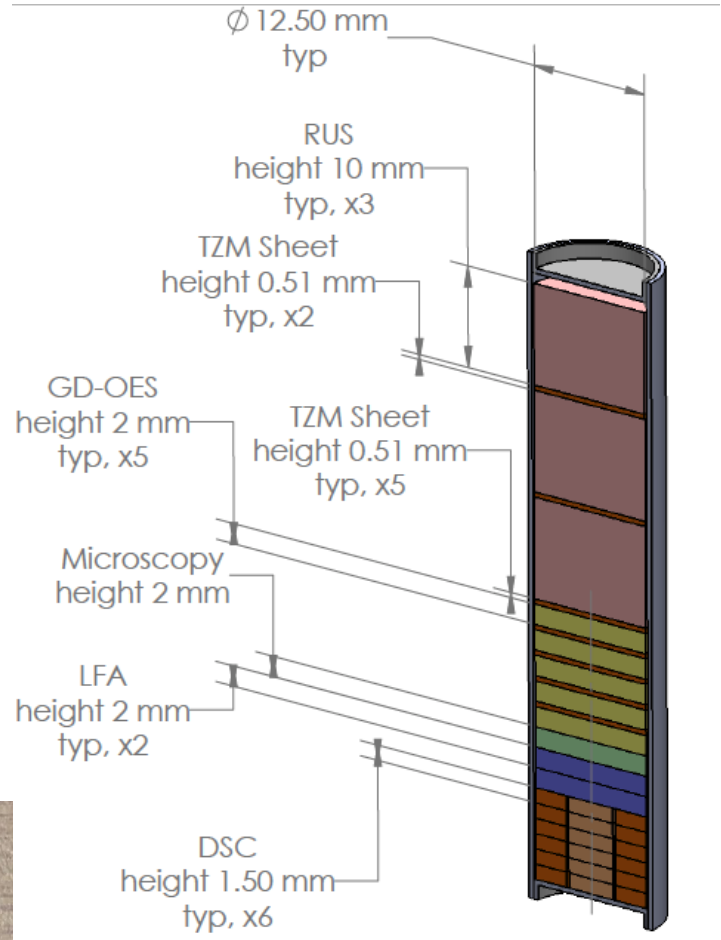
Irradiating YH_2 in ATR

- Plan/perform YH_2 irradiation
- ATR irradiation will test suitability in nuclear environment
  - Some historical results but almost no quantitative data
- Test Plan
  - Gas head space
  - Dimensional stability
  - Microscopy
  - Resonant ultrasonic spectroscopy
  - GD-OES
  - Laser Flash Analysis
  - Differential Scanning Calorimetry



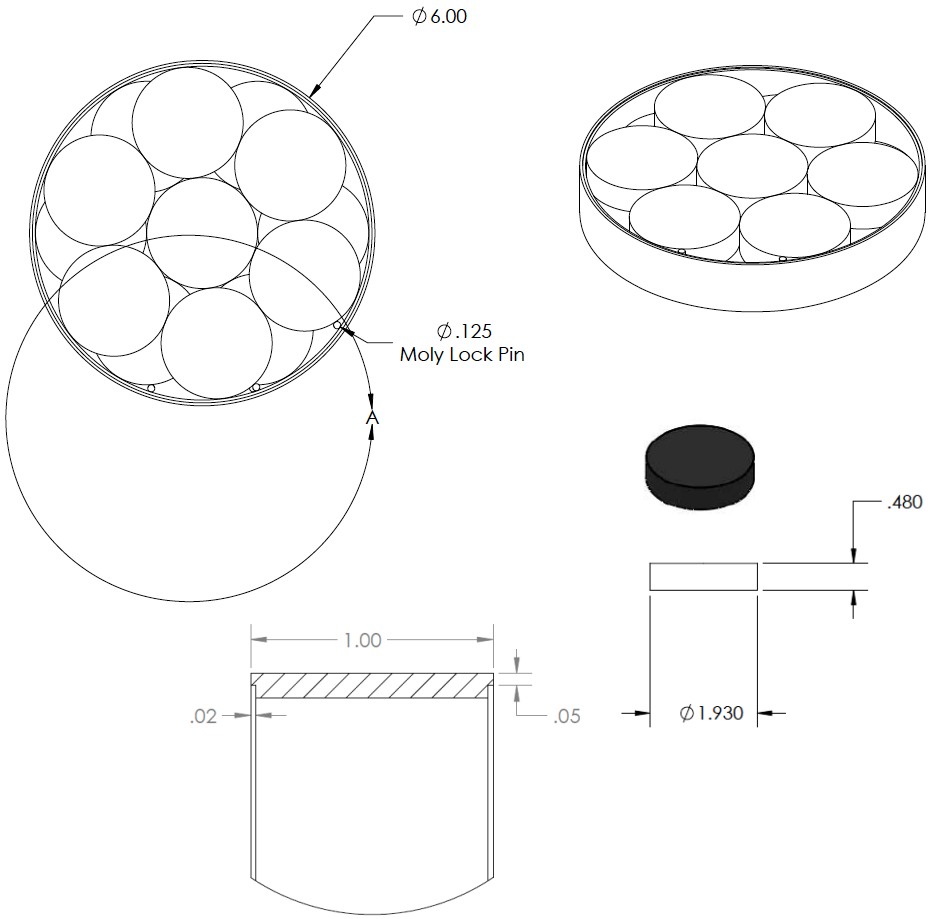
Fuel Fabrication for ATR Irradiation

- Fabrication of samples
- Sample geometry based on desired post-irradiation examination (PIE)
  - Direct hydriding and powder metallurgy
    - Y samples machined to size
      - Legacy and high purity materials
    - Powder prepped from hydrided legacy material
      - Dies received
    - TZM cladding, shims and rings machined



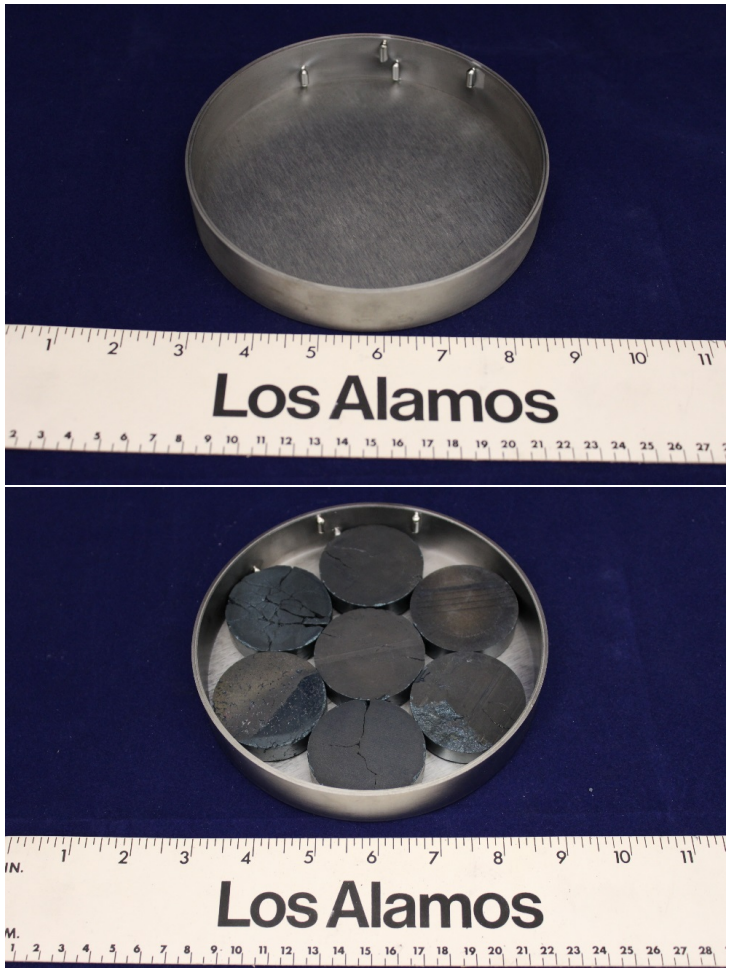
Plan/Perform Integral Critical Experiment

- Planning for August insertion at NCERC
- Sample fabrication
  - 6” molybdenum plates with 2” YH_2 disks
  - 2 fabricated in 2017
  - 2 additional plates in process
  - Molybdenum cans fabricated
  - Y disks machined



Directly Hydrided YH_2 Clad in Molybdenum

- Weld in retaining pins
- Weld bottom lid
- First layer of YH_2
- Second layer of YH_2
- Weld top lid



More on Integral Critical Experiment

NCERC Overview

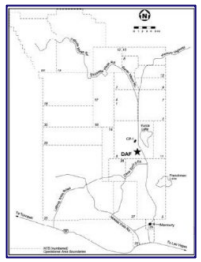
NCERC: National Criticality Experiments Research Center

Location: Device Assembly Facility (DAF) at the Nevada National Security Site (NNSS)

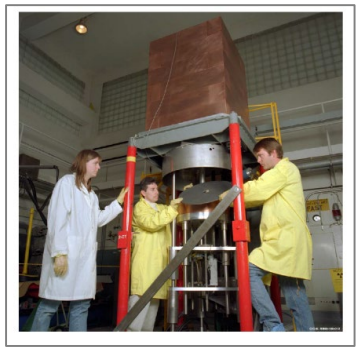
Operated by: Los Alamos National Laboratory

NCERC Mission Statement:

- The mission of the National Criticality Experiments Research Center (NCERC) is to conduct experiments and training with critical assemblies and fissionable material at or near criticality in order to explore reactivity phenomena, and to operate the assemblies in the regions from subcritical through delayed critical. One critical assembly, Godiva-IV, is designed to operate above prompt critical.



NCERC Assemblies

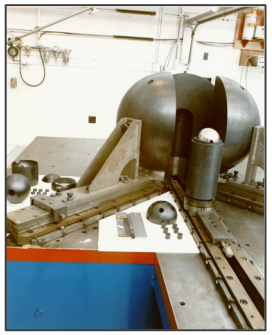


Comet

- JAEA U-Lead and Pu-Lead
- NASA KRUSTY
- NTNF irradiations
- CURIE

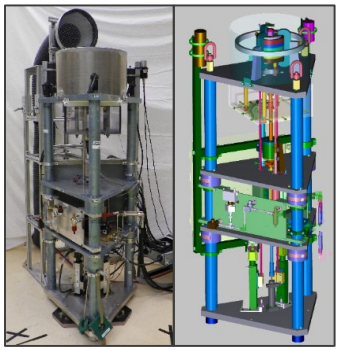
Planet

- NCSP training
- NTNF irradiations
- Alternative nuclear materials
- TEX



Flat-Top

- NCSP training
- NTNF irradiations
- NASA DUFF
- ICSBEP (NCSP projects)



Godiva IV

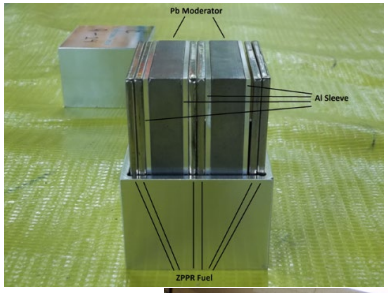
- NCSP training
- NTNF irradiations
- NCSP projects
- ICSBEP (NCSP projects)

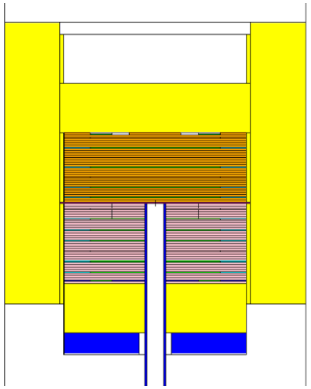
Subcritical

- NCSP Training
- NDSE
- DTRA, NA-84, NA-22
- University Consortia
- IRSN and AWE measurements

Comet/Vertical Lift Assembly

- Vertical lift assembly
- Capable of any situation allowing for separation distance to achieve critical
  - Kilopower
  - JAEA Lead series
  - CURIE URR experiment
  - Hypatia

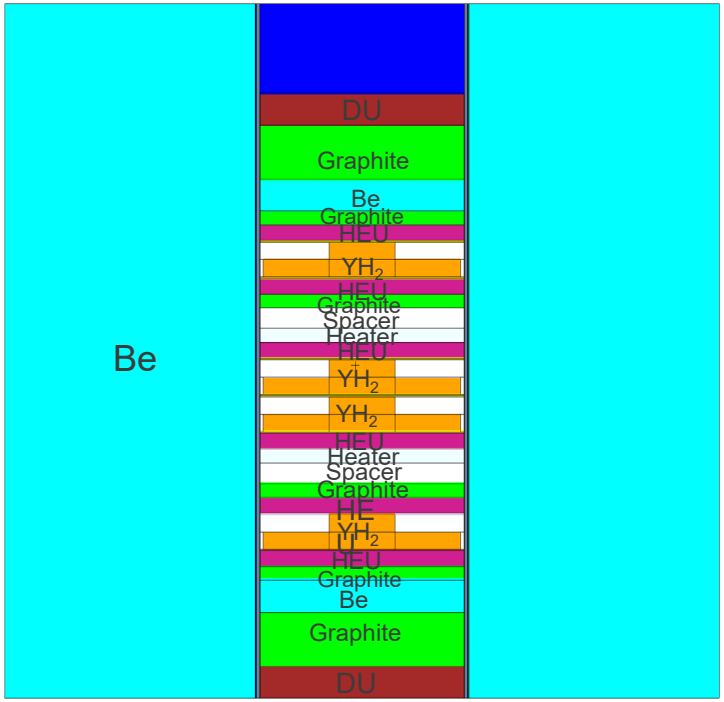




Motivation

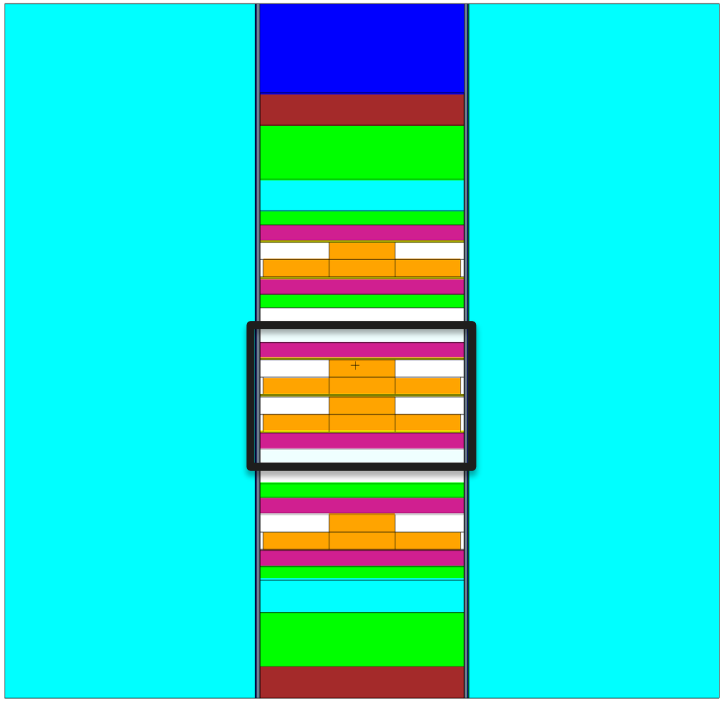
- Yttrium dihydride YH_2 is a promising candidate for moderator applications
  - High thermal stability compared to other metal hydrides
  - Significantly reduces critical mass

- Lots of material property measurements
- New S(a,B) in ENDF/B-VIII.0 for YH_2
- Many differential measurements across DOE
- No integral experiments



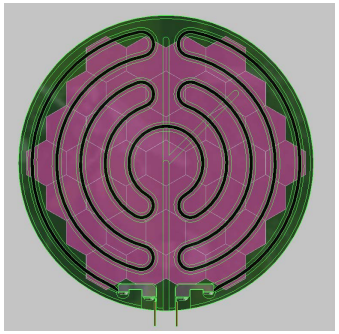
Experimental Objectives

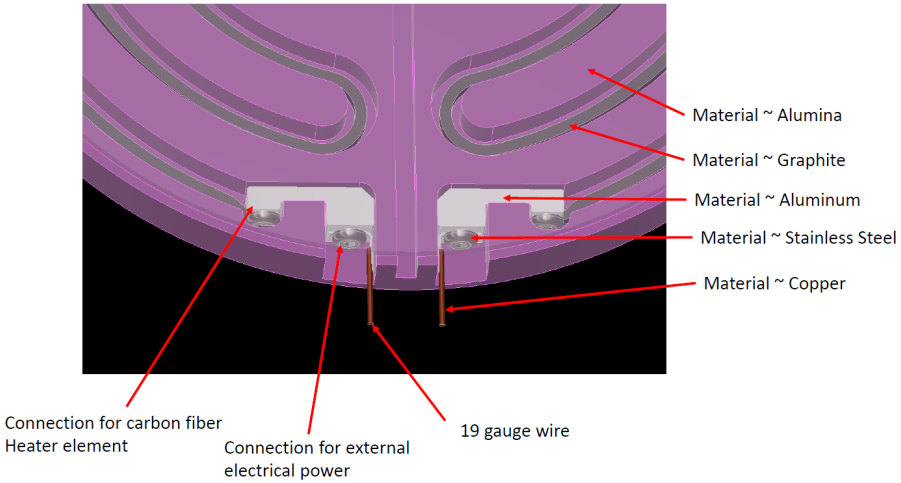
- Measure reactivity with central region heated
  - Various points from 20 C to 330 C
  - Compare the change in reactivity at these points
  - Repeat without YH_2 in central region
- Why not heat all?
  - Decouple many competing effects
  - Reduce heating elements needed
  - Reduce time to heat the region
- Why limit to 330 C?
  - HEU metal used is not alloyed
  - Staying away from HEU phase transition



Heaters

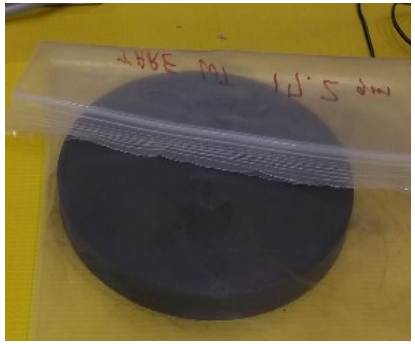
- Manufactured by NASA
  - Alumina outer surfaces
  - Graphite inner heating element
- Dimensions
  - OD: 5.85 inches
  - Height: 0.406 inches
- Heat Control
  - Waiting for direction from NASA
- Temperature Readouts
  - Waiting for direction from NASA
- Status
  - Start temperature testing by Ap





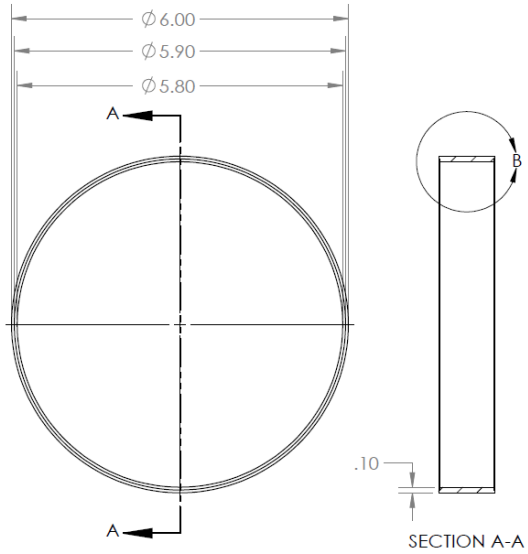
HEU Fuel

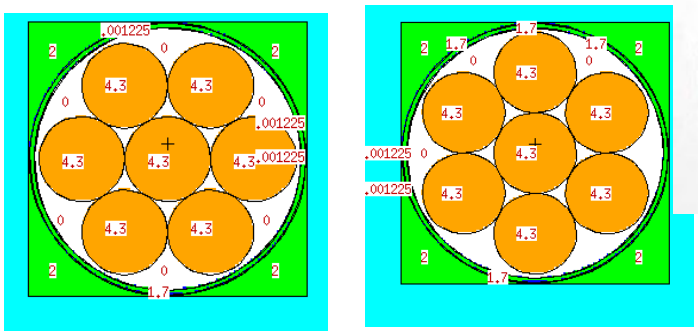
- General information
  - Commonly referred to as “C-discs”
  - 6 total
  - Bare/ unclad
- Dimensions
  - Outer Diameter: 5.94 inch
  - Height: 0.465 inch
- Isotopics
  - U-235: 93.1 %
  - U-238: 6.9 %
- Mass
  - 3920 g



Yttrium Hydride

- 14 discs per can
- 2 layers of discs per can
- Canning
  - Molybdenum
  - E-beam welded
  - vaccum
- Can Dimensions
  - OD: 6.00 inch
  - ID: 5.80 inch
  - Height: 1.00 inch
  - Top and Bottom Height: 0.2 inch
- YH_2 Disc Dimensions
  - OD: 1.930 inch
  - Height: 0.480 inch

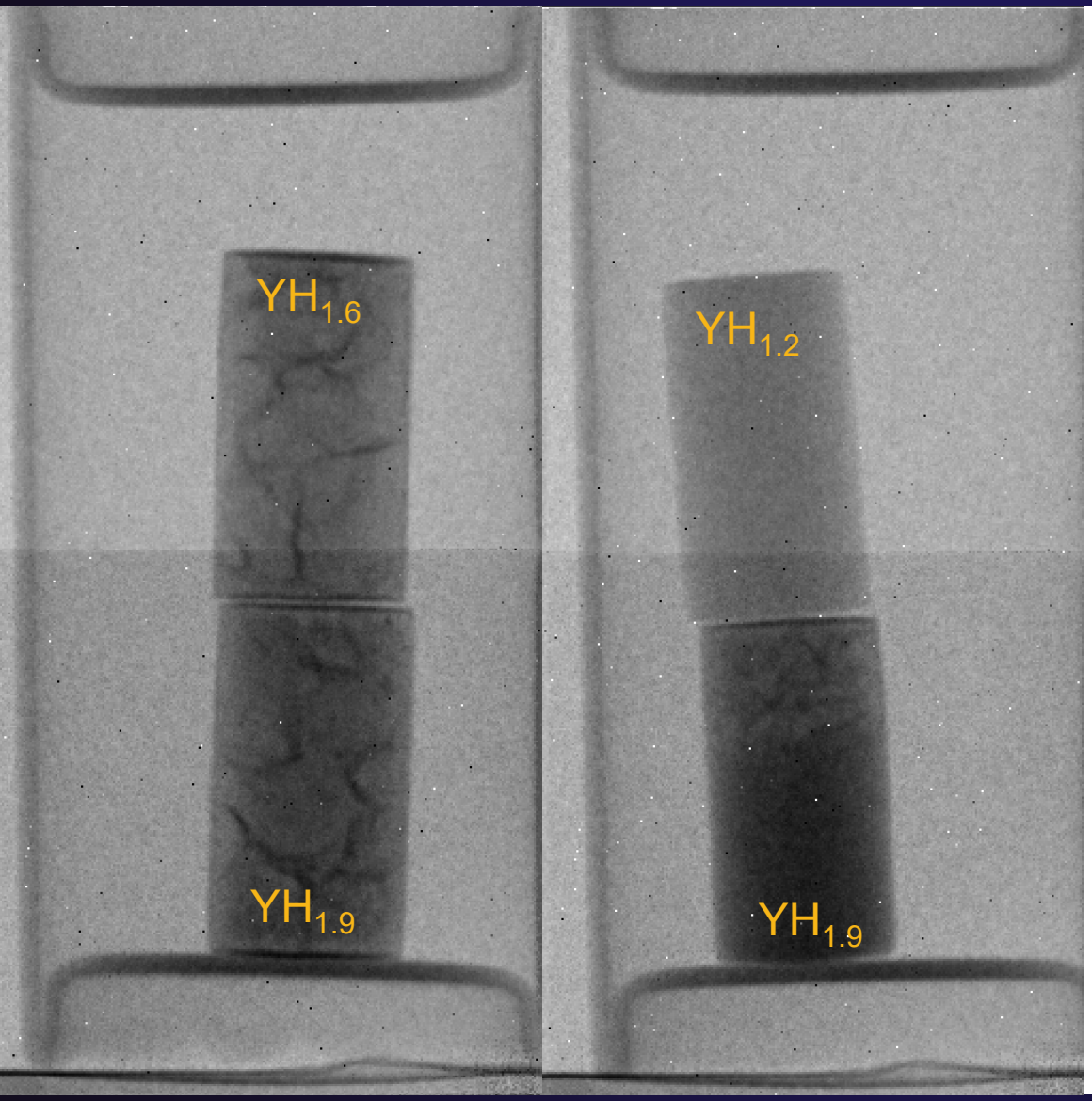




$YH_{1.9}$

YH_2 plates in MCNP®

How Neutron Radiography @ LANSCE can be used to Quantify H-concentrations in YH Samples



LDRD-Director's Initiative Team:

Alex Long[2]

Sven Vogel[2]

Holly Trellue[1]

Adi Shivprasad[2]

Erik Luther[3]

Vedant Mehta[1]

Mike Cooper[2]

Jerawan Armstrong[4]

Mikaela Blood[1]

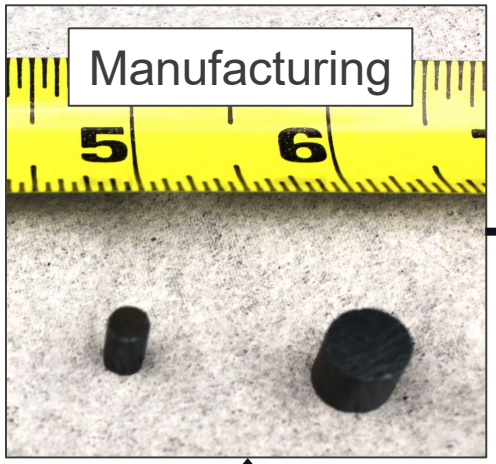
[1]NEN-5, [2]MST-8, [3]Sigma, and [4]XCP-3

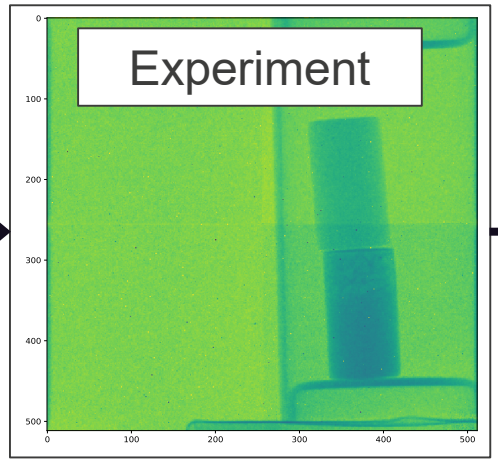
Modeling and Simulation

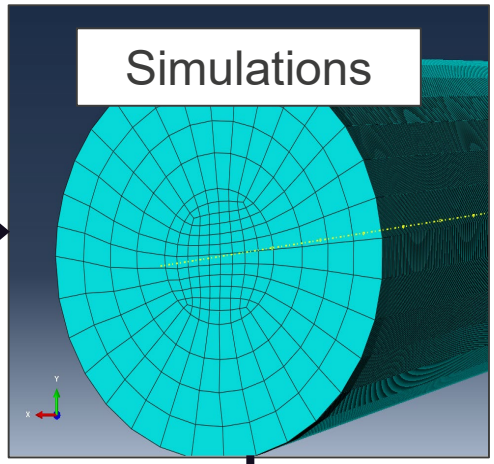
Experimental

02/25/2020

Combining Manufacturing, Experiments, and Simulations to Improve Understanding of Moderator Performance







Direct Hydriding
Powder Met.

@ SIGMA

$H(x, y, T)$
Stoichiometry (YH_x)
H-Diffusion
Microstructures
Thermophysical Props.

@ LANSCE, UT-Austin, and NCERC

DFT

ABAQUS

NJOY

MCNP

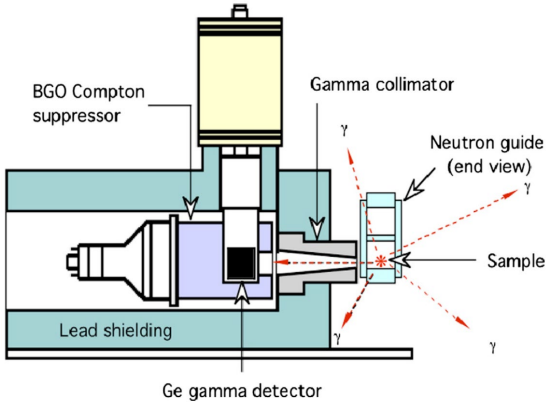
Goal:
Moderator Performance

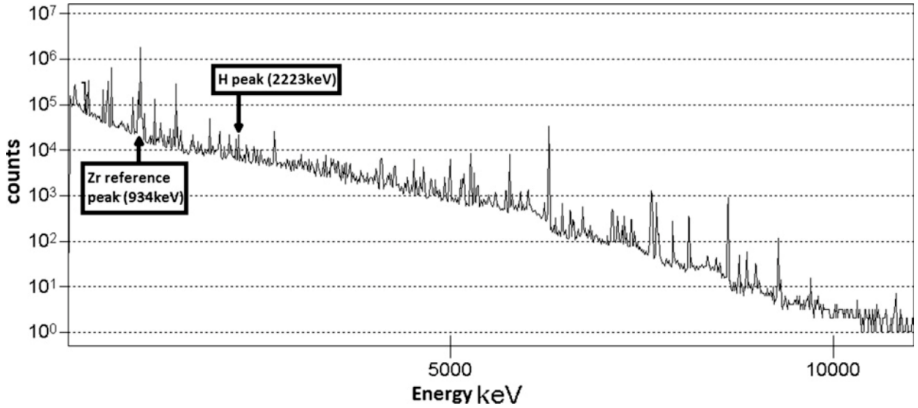
Our Overall Objective: To initiate advancements in materials development through manufacturing, experimental, and modeling capabilities with the aim of accelerating the discovery process for optimal moderator materials.

Non-Destructively Quantifying Hydrogen Concentrations with Neutrons

Prompt Gamma Activation Analysis

$$n + H \longrightarrow d + \gamma$$

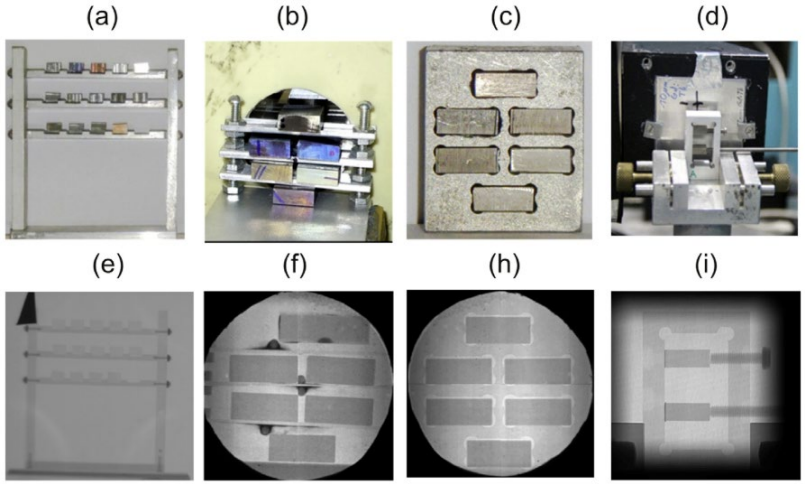




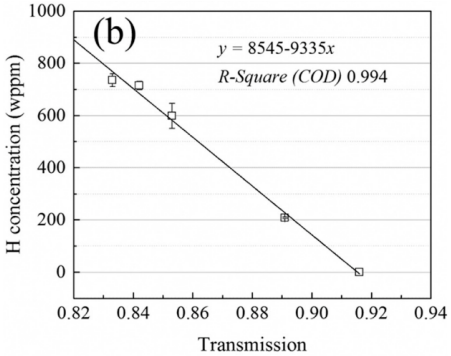
Highly accurate bulk measurement

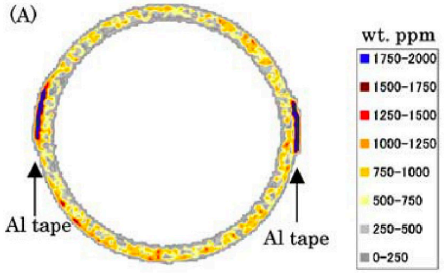
YH_2 Measurement performed @ UT-Austin

Attenuation via Neutron Radiography



Accurate spatially resolved measurement

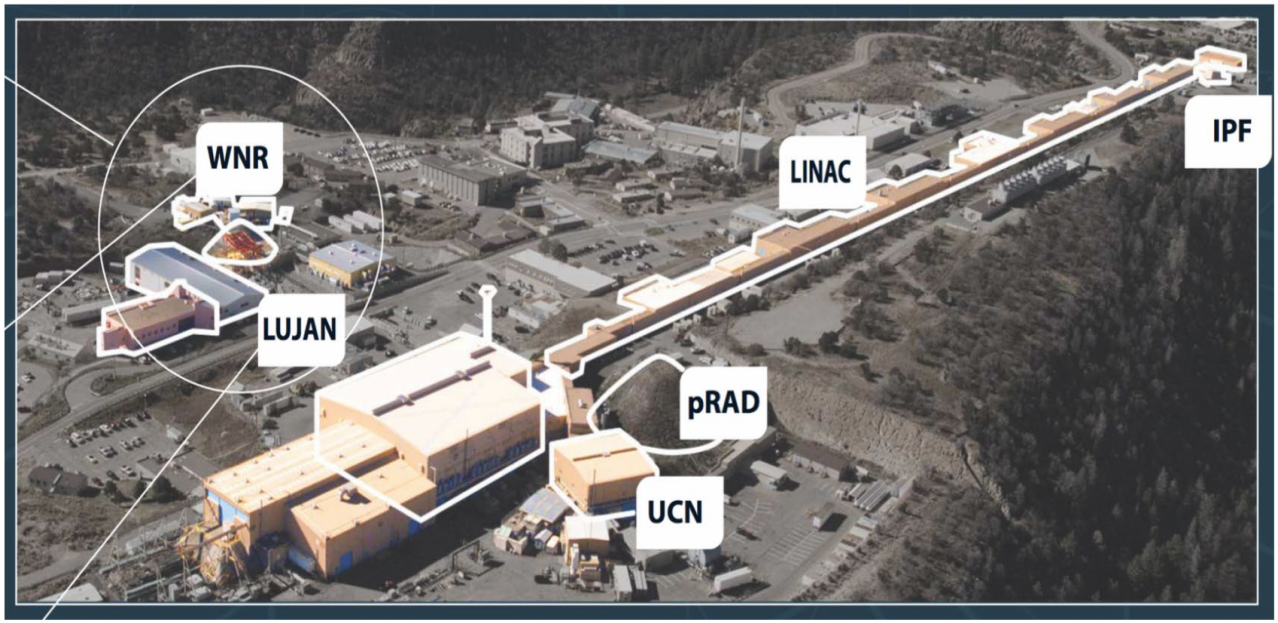


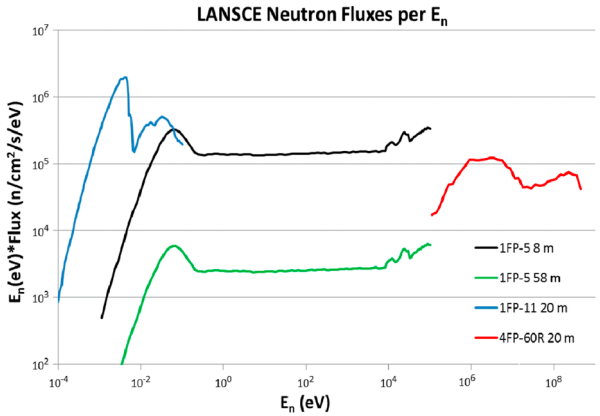


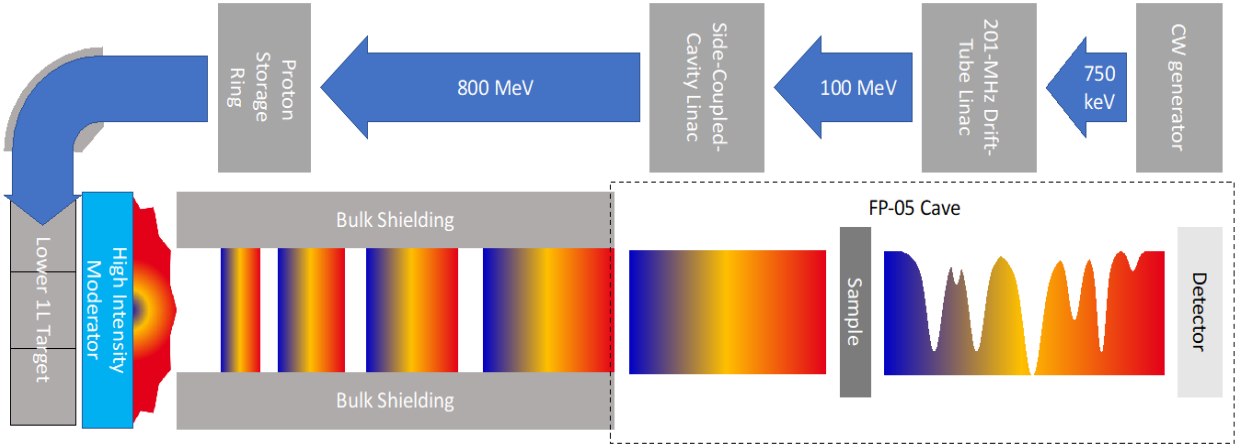
YH_2 Measurement performed @ LANSCE

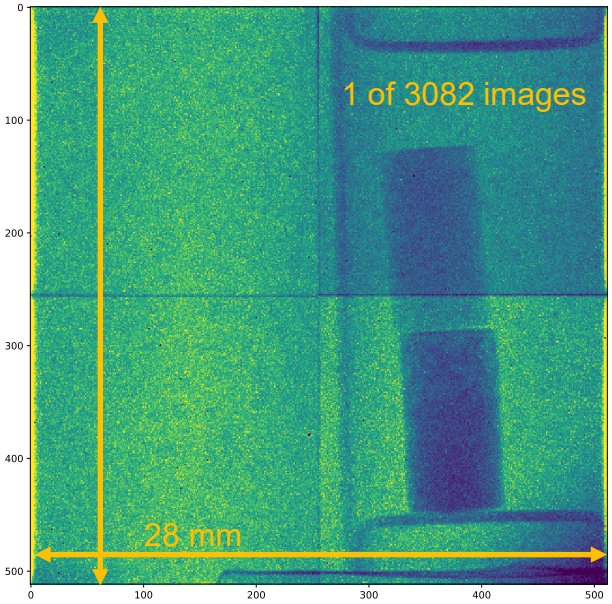
A. Couet et al. /Journal of Nuclear Materials 425 (2012) 211–217
N.L. Buitrago et al. / Journal of Nuclear Materials 503 (2018) 98-109
W. Gong et al. / Journal of Nuclear Materials 508 (2018) 459-464

The Energy-Resolved Neutron Imaging (ERNI) Flight Path 05





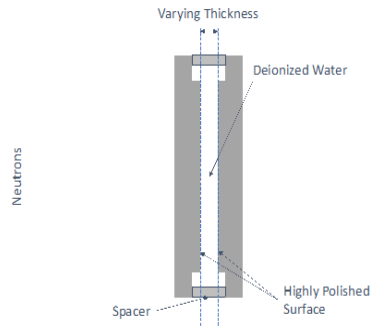




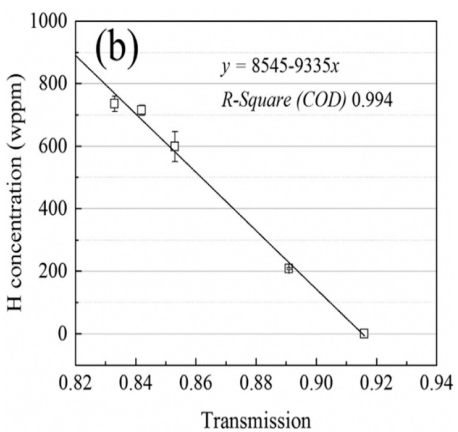
Measurements Performed on FP05 in the 2019 LANSCE Run Cycle

Quantifying H-concentrations spatially @ ambient condition

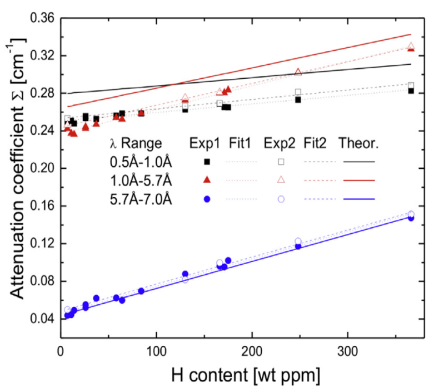
Use samples with well-known Hydrogen concentrations per pixel to calibrate Transmission or H-attenuations coefficients.



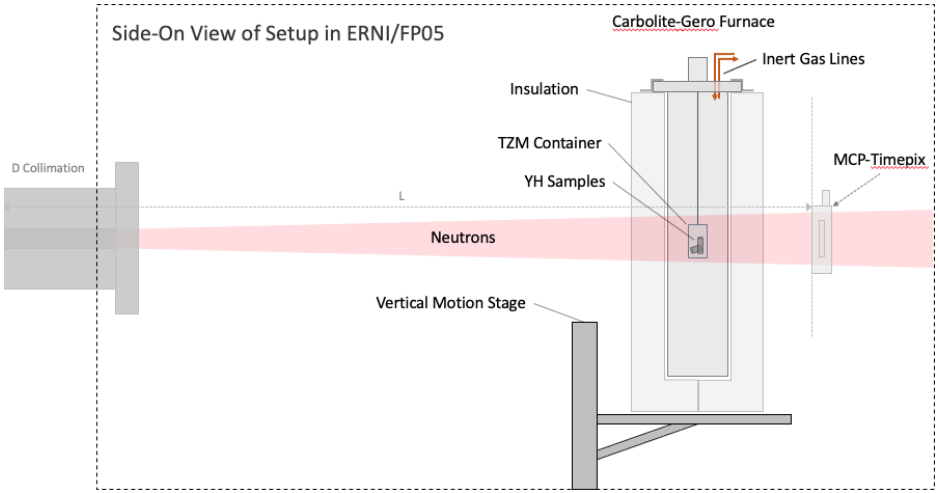
Measure transmission or H-attenuations coefficients in unknown sample and use calibration from water samples to determine H-concentrations



Calibration lines for wavelength-resolved attenuation coefficients of Zircaloy-2 calibration specimens.

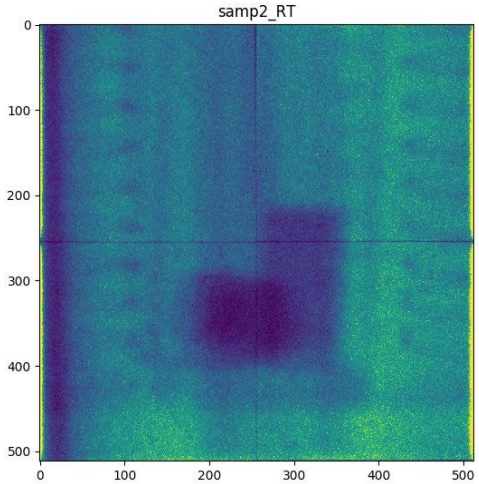


Investigating possible H-diffusion @ elevated temperatures



Used a tube furnace to heat two YH samples while performing neutron radiography measurements.

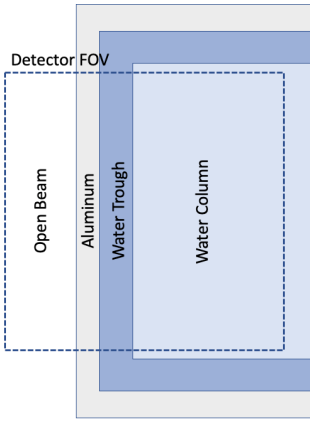
Wanted to see if there was any relative change in the H-concentration as temperatures increase.

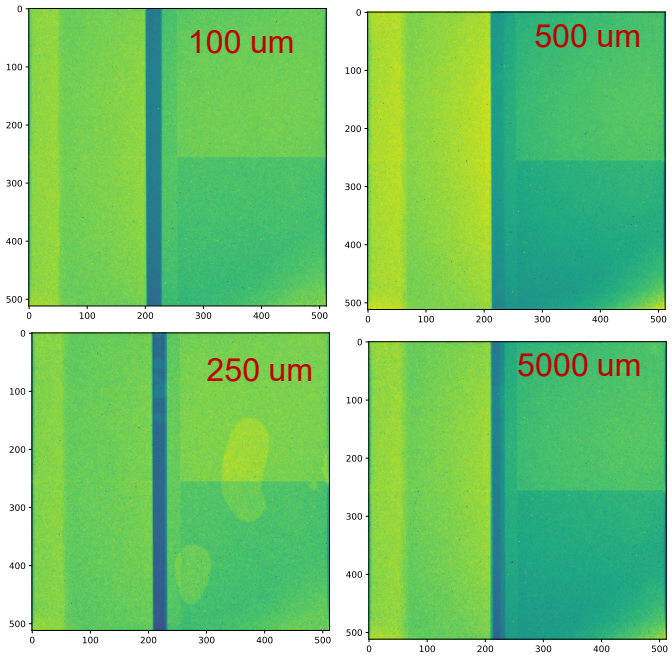


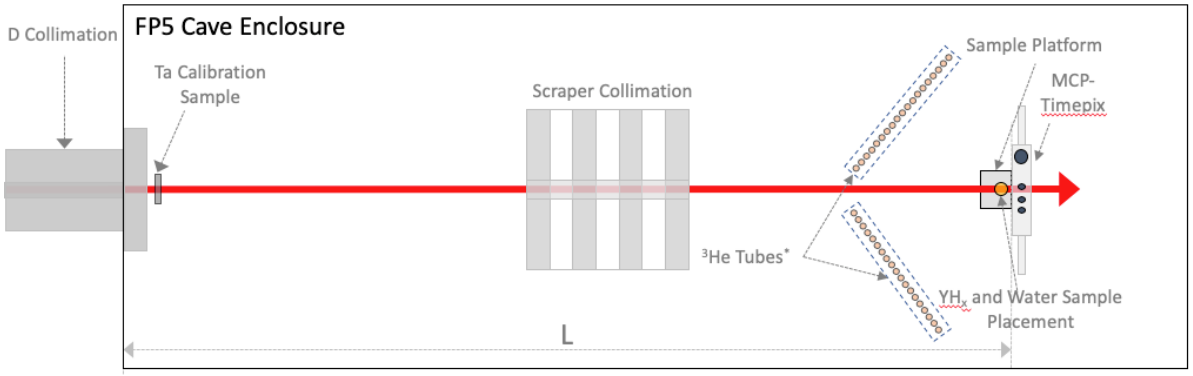
W. Gong et al. / Journal of Nuclear Materials 508 (2018) 459-464
N.L. Buitrago et al. / Journal of Nuclear Materials 503 (2018) 98-109

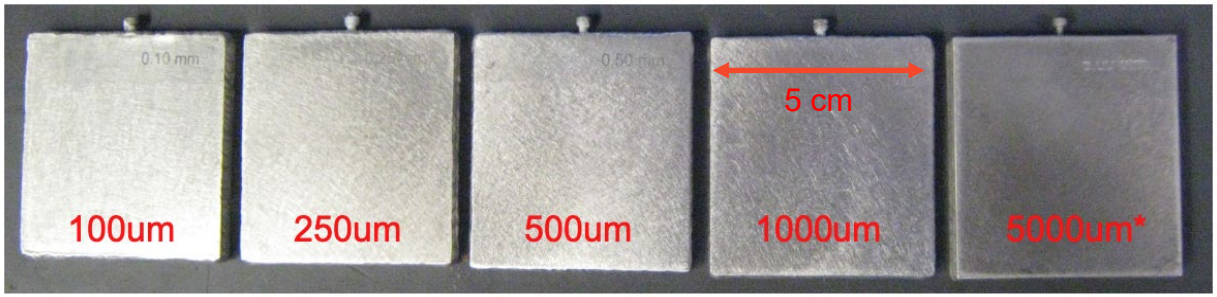
Quantifying H-concentrations Spatially at Ambient Conditions: Experimental Setup

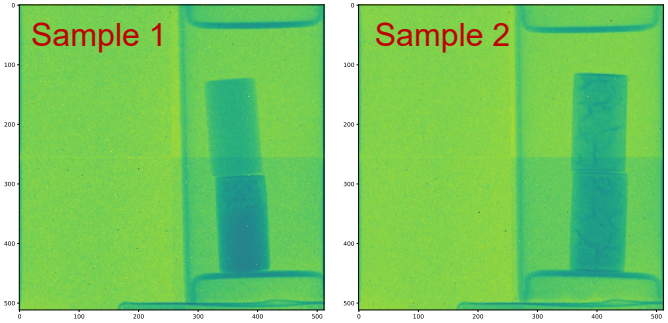
- Measurements were performed from **Dec 3rd - Dec 10th**
- Calibration Samples: Highly polished Al containers with DI water.
- Used ERNI camera (MCP-Timepix)
  - Used Ta foil for calibration of TOF
- YH_x Samples:
  - Container #1: $YH_{1.2}$ and $YH_{1.9}$
  - Container #2: $YH_{1.6}$ and $YH_{1.9}$







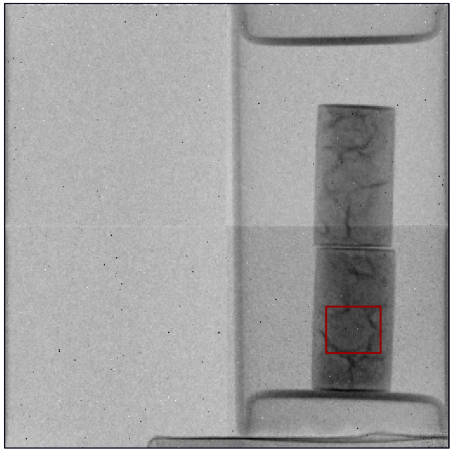


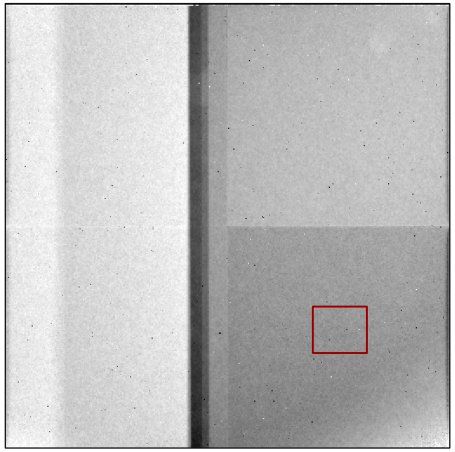


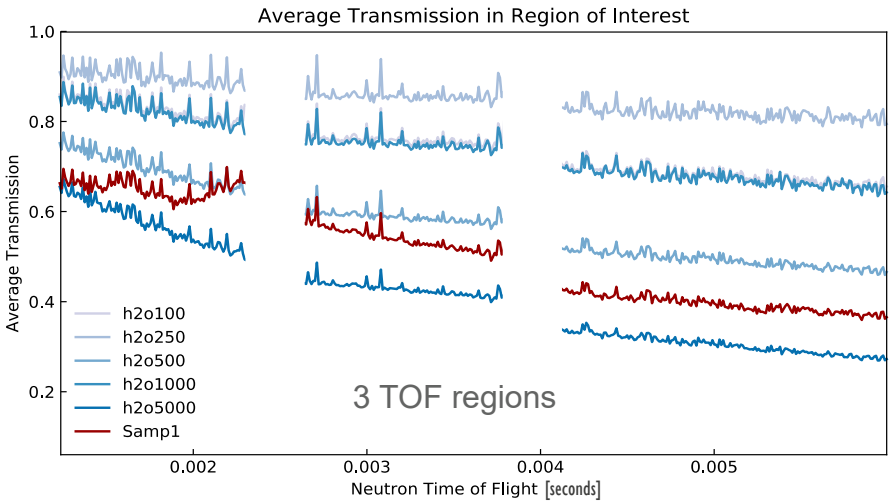
Quantifying H-concentrations Spatially at Ambient Conditions: Current Status of Analysis

$$T(x,y,E_n) = \frac{I_s(x,y,E_n) - B(x,y,E_n)}{I_o(x,y,E_n) - B(x,y,E_n)}$$

$$\Sigma(x,y,E_n) = \frac{ln(T(x,y,E_n))}{z}$$



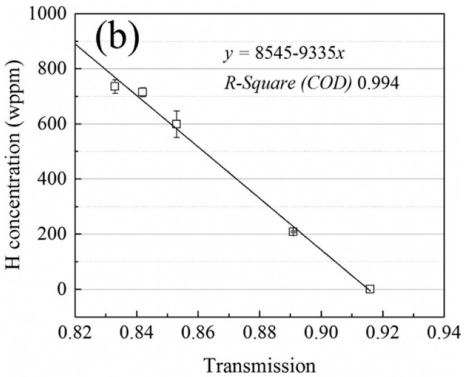


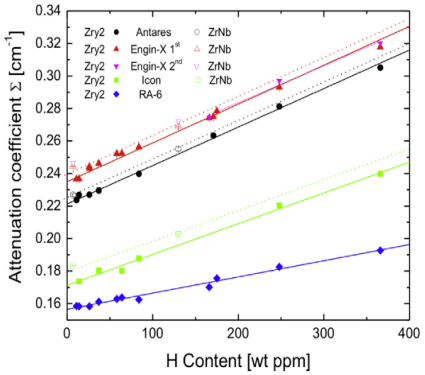


Plot H-Concentrations as a function of Transmission for each pixel using water samples.

Fit points with simple linear function:
H(wt%ppm) = A*Trans + B

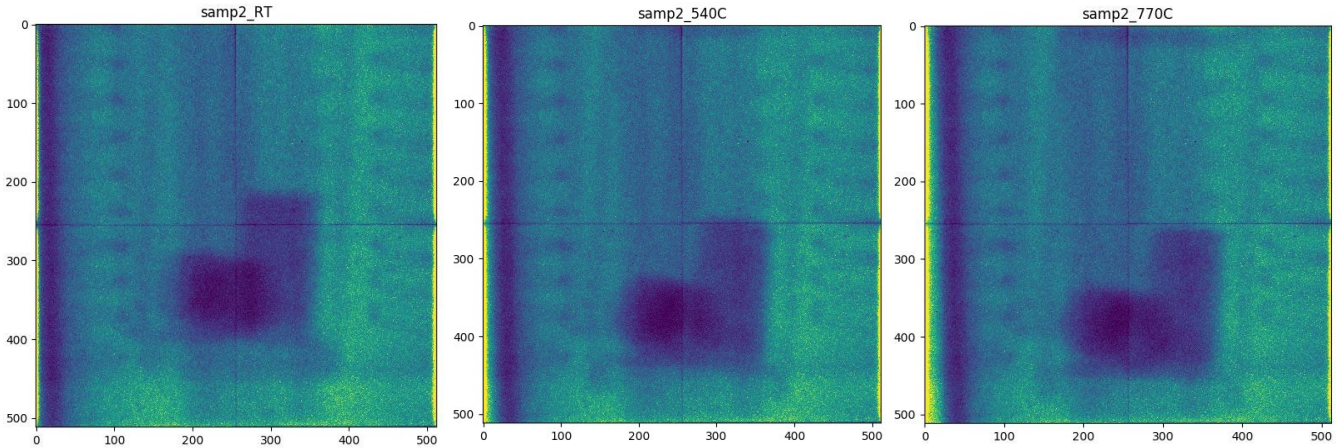
Determine H-Concentrations per pixel in YH sample images using simple calibration function.



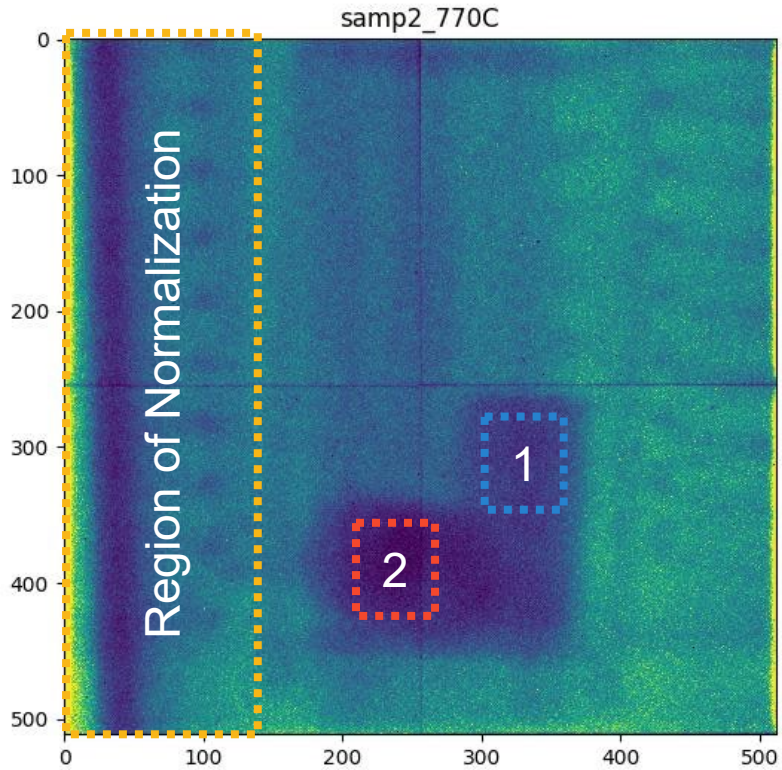


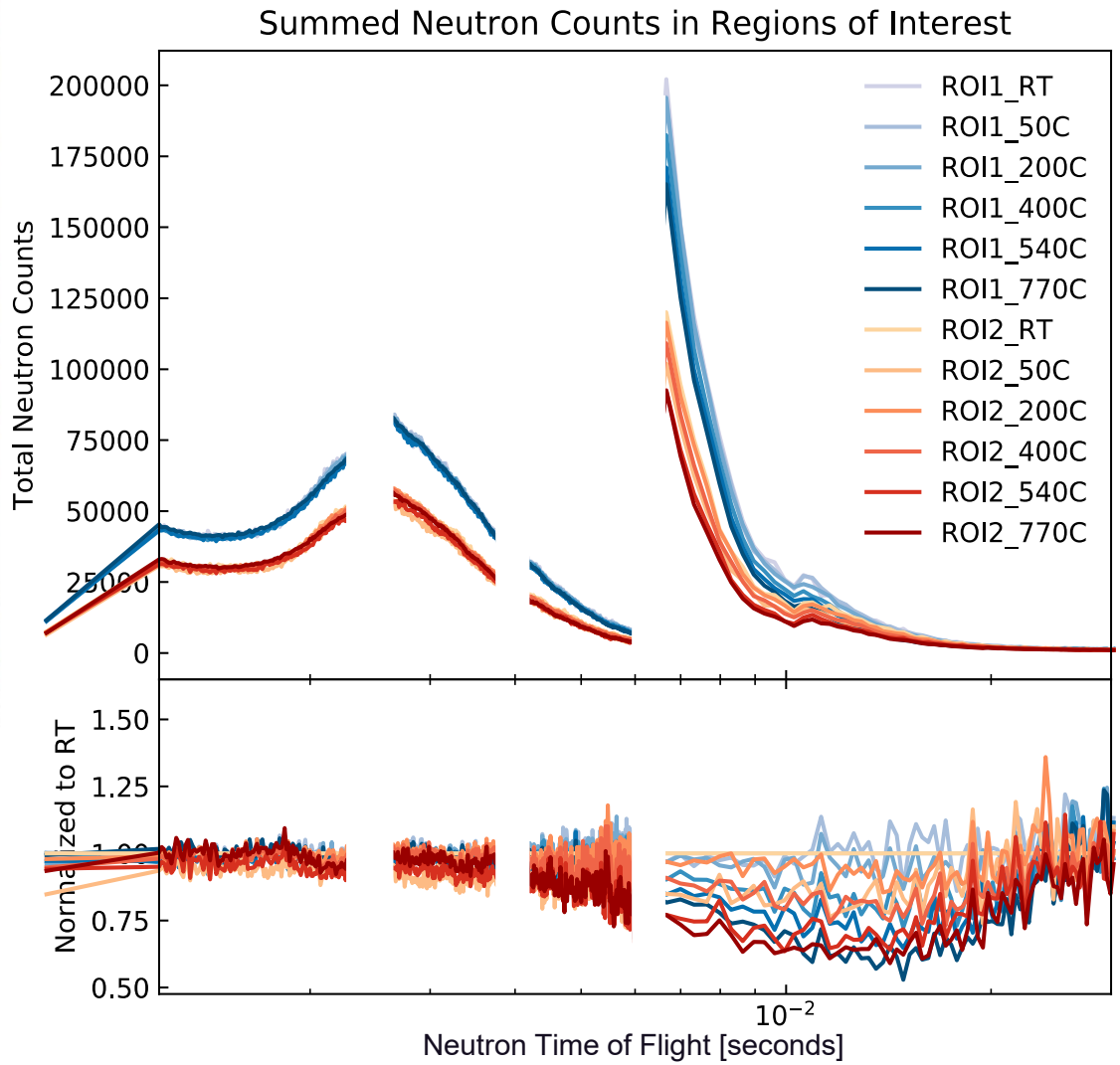
Investigating Possible H-diffusion at Elevated Temperatures: Experimental Setup

Temperature	Time (hours)
30 ℃	~ 5
200 ℃	~ 3
400 ℃	~ 3
540 ℃	~ 3
770 ℃	~ 12
50 ℃	~ 5



Investigating Possible H-diffusion at Elevated Temperatures: Current Status of Analysis





Looked at two regions:

1:$YH_{1.6}$ and 2:$YH_{1.9}$

Still sorting through and processing results…

Road Map for FY2020

Finish up Analysis on 2019 Run Cycle Measurements

- Sum up all runs for each measurement
- Denoise and interpolate out and zero (non-working) pixels.
- Normalize all radiographs to open beam area
- Calculate Transmission Images
- Verify Calibration Samples
- Break up Transmission into neutron energy groups
- Determine H-Concentrations
  - H(wt % ppm) = a*(Trans) + b
  - Calculate attenuation coeff's
- Compare determined H-concentrations to quoted stoichiometries
- Determine uncertainties
- Investigate distributions and homogeneity of H through-out samples.

New and Improved measurements Proposed on ERNI/FP05 for 2020 Run Cycle

- Elevated Temperature Measurements.
  - Building new High-temp Compact Furnace
  - Would like to take both non-temp and temp gradient images to look at H-diffusion through YH_2 samples
- Improved ambient condition measurements
  - Possibly improve upon water plates.
  - Look at more YH2 samples.

N.L. Buitrago et al. / Journal of Nuclear Materials 503 (2018) 98-109

Conclusions

- LANL has led the development of yttrium hydride as a high temperature moderator material, which is beneficial in microreactors.
- We are fabricating YH_x samples by numerous techniques for different geometries/purposes:
  - LANSCE measurements
  - ATR irradiation experiment
  - NCERC integral critical experiment
- An upcoming talk will discuss diffraction measurements at LANSCE for YH_2 and calculations of data for cross sections/reactor performance.

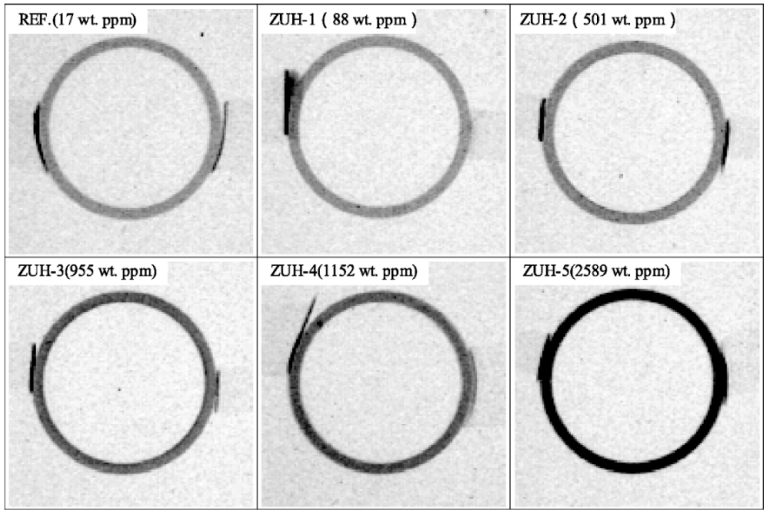
Thank you for your attention

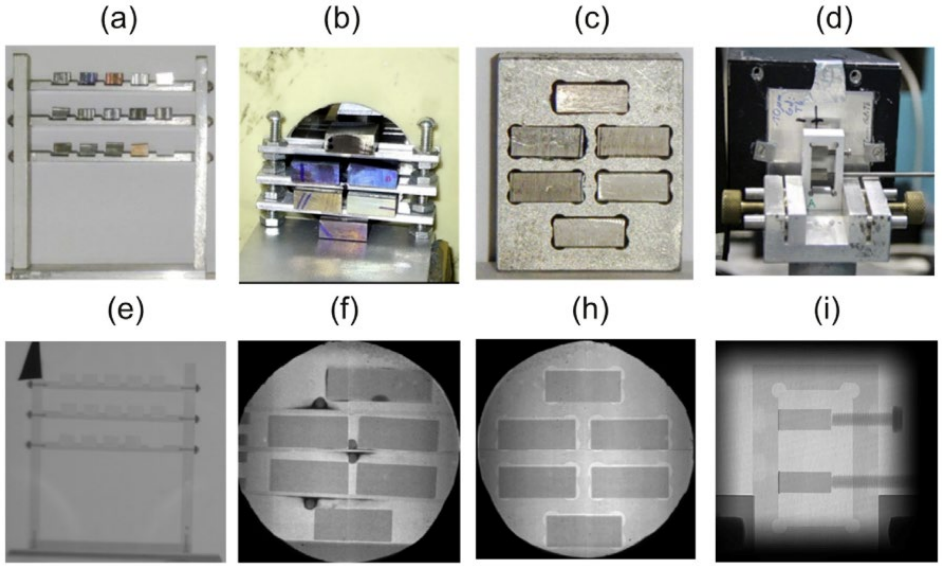
Los Alamos
NATIONAL LABORATORY
EST. 1943

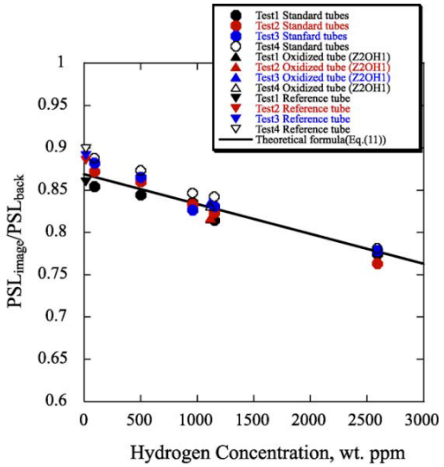
Delivering science and technology
to protect our nation
and promote world stability

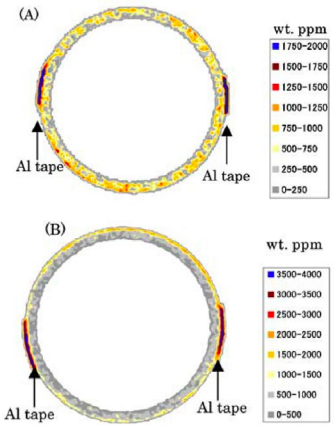
Backup Slides

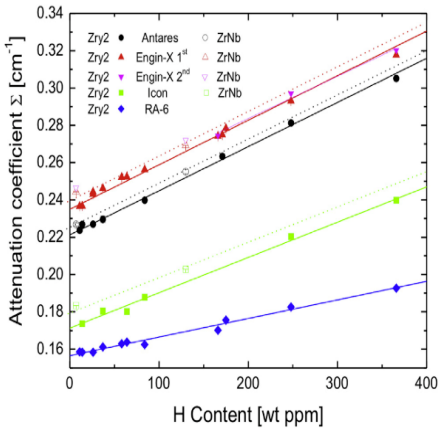
Previous Work Measuring H-Concentrations in Zircaloy



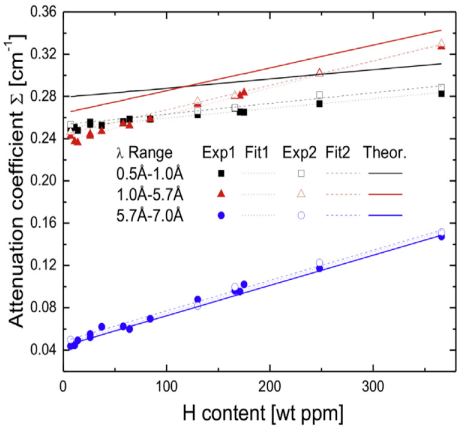








Calibration lines for wavelength-resolved attenuation coefficients of Zircaloy-2 calibration specimens.

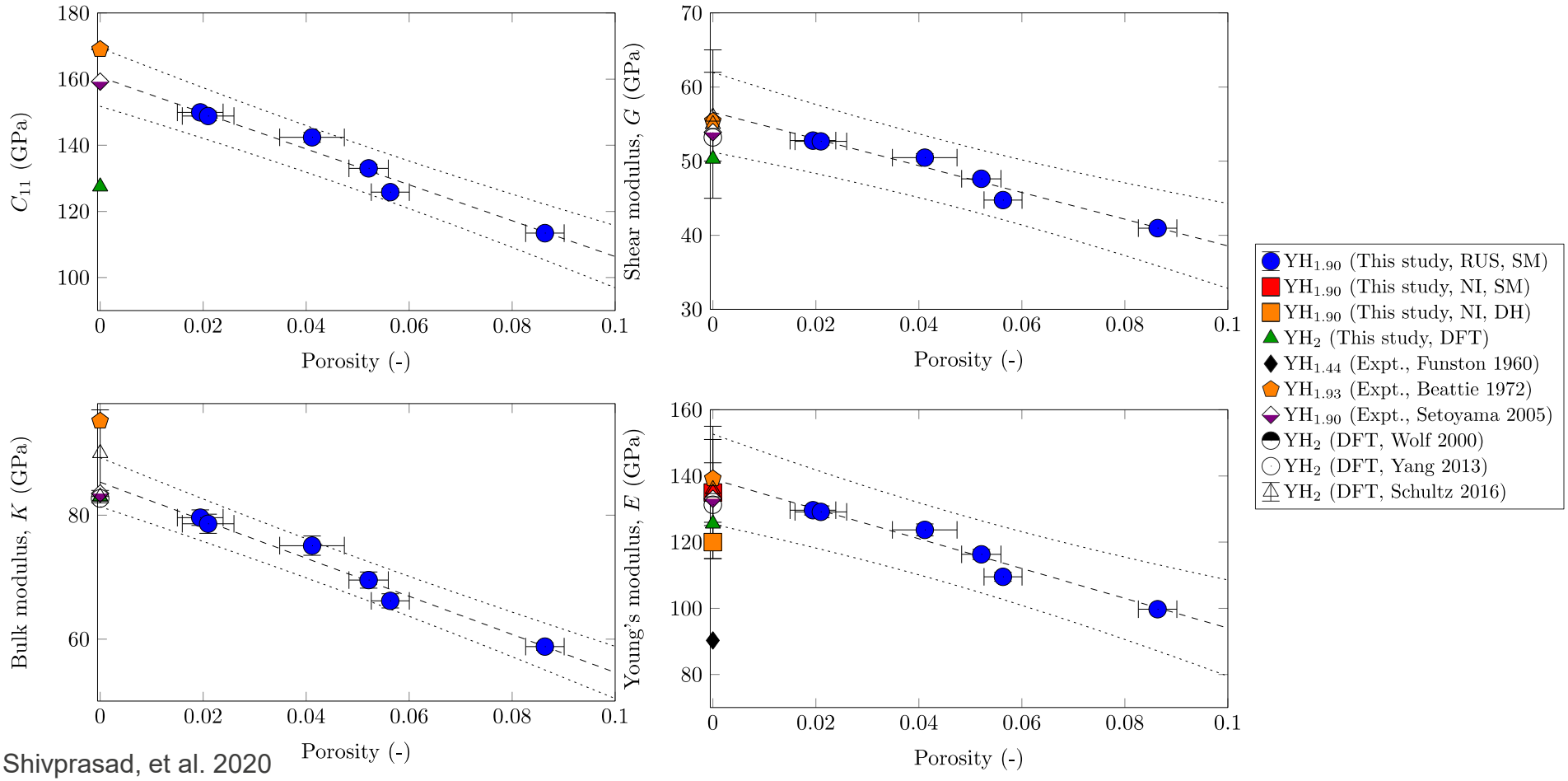


A. Couet et al. /Journal of Nuclear Materials 425 (2012) 211–217
N.L. Buitrago et al. / Journal of Nuclear Materials 503 (2018) 98-109

Future work will focus on scaling up fabrication and evaluating degradation during reactor operation

- Quantify H-content of pellets using techniques other than mass gain and IGF
- Shift to production of larger pellets and more complex geometries in-line with reactor designs
- Evaluate cladding candidates for hydrogen diffusion barrier at temperature
  - Mo alloys
- Examine other metal hydride moderators
  - Y/Cr and Y/Ce
- Advanced Moderator Material Handbook

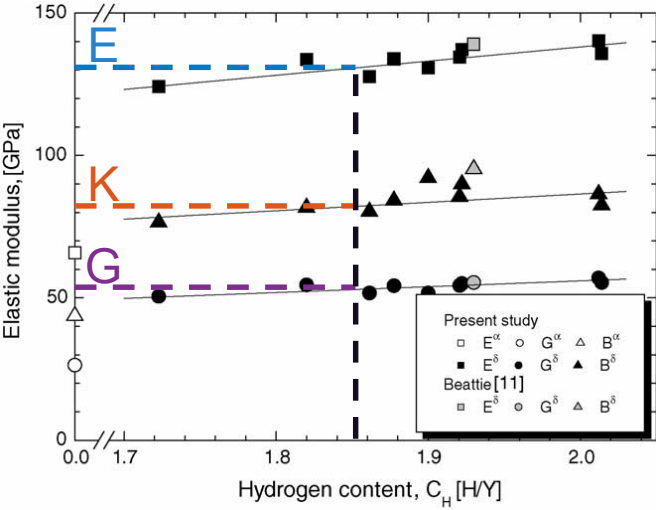
Elastic modulus values were consistent with literature values for directly-hydrided yttrium



Shivprasad, et al. 2020

Elastic moduli are consistent with literature values

Interpolated values from literature



Values for 100%-dense YH_2

	This work	Literature
Young's modulus, E	139.0 ± 8.1 GPa	130.7 GPa
Bulk modulus, K	85.1 ± 8.3 GPa	82.0 GPa
Shear modulus, G	56.6 ± 1.8 GPa	52.9 GPa
Poisson's ratio	0.230 ± 0.079	0.239
Debye temperature	365 ± 21 K	356 K

Reasonable overlap in measured values with literature values indicates we're on the right track in fabricating high-quality YH_{2-x} pellets