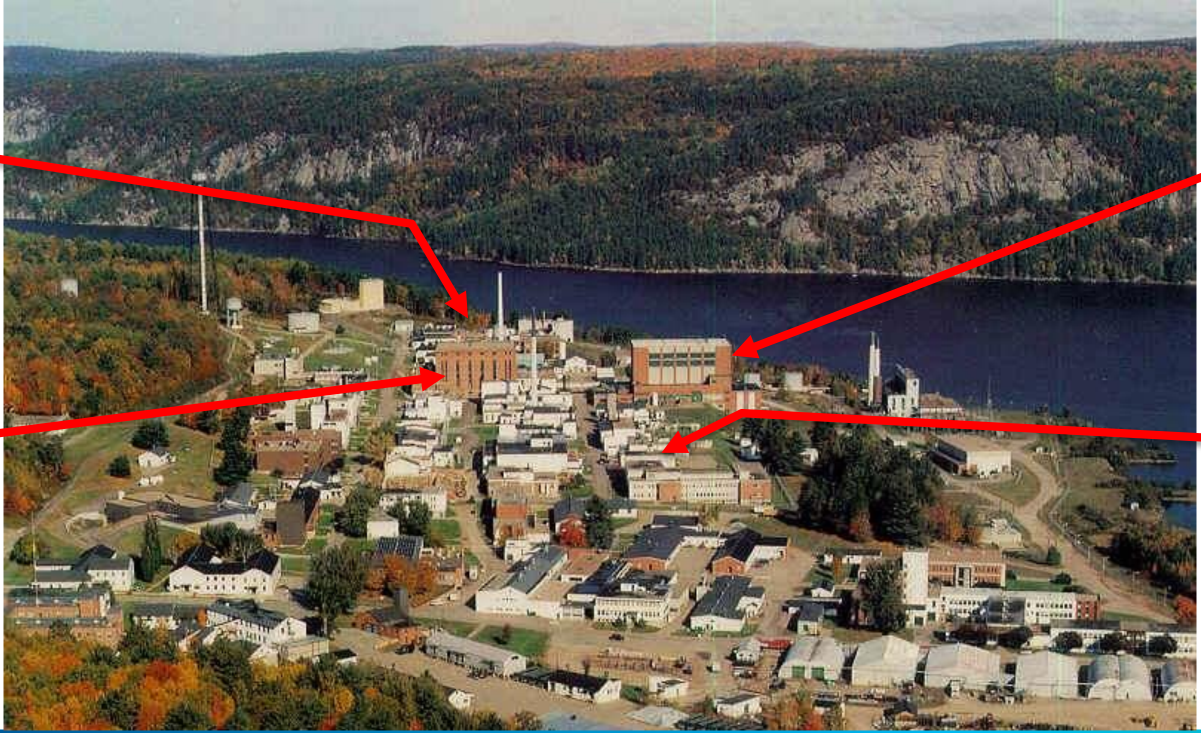


Neutron Beams for Reactor and Related Materials Research

R.B. Rogge

Joint ICTP/IAEA Workshop, 2017 November 6-10

Introduction

Chalk River is the nucleation point of the Canadian nuclear industry—natural outcome was that much of the neutron beam work was on nuclear materials

- Reactors and nuclear R&D require a wide range of materials research capabilities
- Why use neutrons?
- Present several studies:
  - Phase analysis of irradiated fuel
  - Hydrogen in zirconium
  - Identifying found material

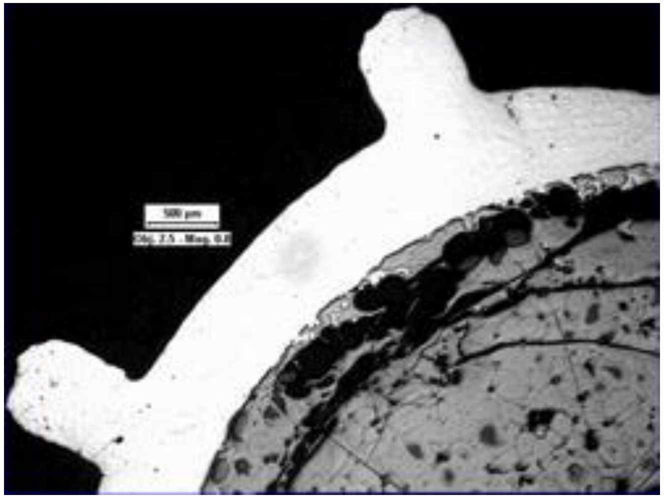
The Neutron's Unique Properties

- Wavelength, λ, comparable to interatomic spacing
- Energies comparable to *excitations*
- Magnetic moment
- Scatters coherently and incoherently, or is absorbed
- Cross sections are of same order of magnitude for all nuclei (*e.g.* can 'see' Mg about as well as Fe or Au)
- Cross sections vary irregularly across periodic table (*e.g.* Fe about 10x Co)
- Highly penetrating (no charge, interacts with nucleus)

Bottom Line:
Neutrons and X-Rays do many of the same things. Neutrons are sometimes flux-limited in comparison, but some things only neutrons can do.

Powder Diffraction of Active Samples

Irradiated Nuclear Fuels



Sears et. al RRFM 2006 - Cdn pin type fuel

- There are many Research and Test Reactors in the world that require the development of new very high U-density fuels (U density > 8 grams U/cc) based on LEU (< 20 at.% ^{235}U)
- Canada, USA, France, Argentina, Russia have all tested UMo-Al dispersion fuel and reported a failure
- Some postulated that the fuel, initially crystalline, becomes amorphous or partly amorphous due to irradiation
- How does this compare with NRU fuel which has been reliable for decades?

Scattering Lengths

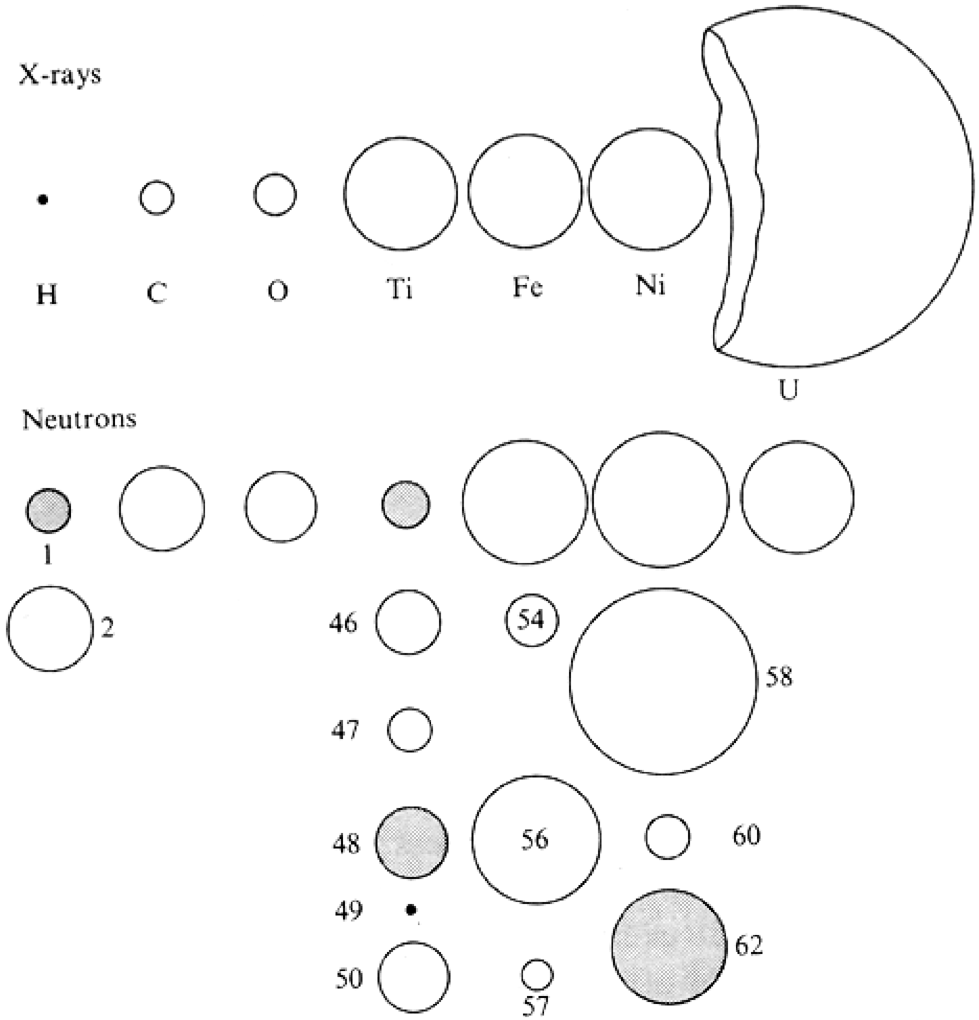
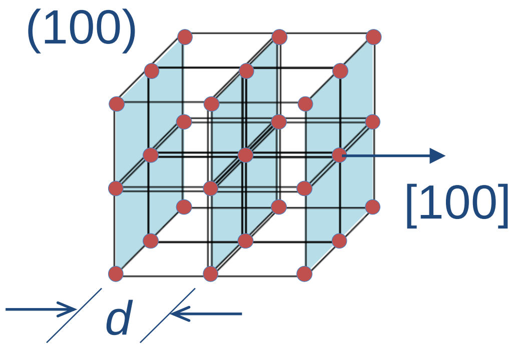
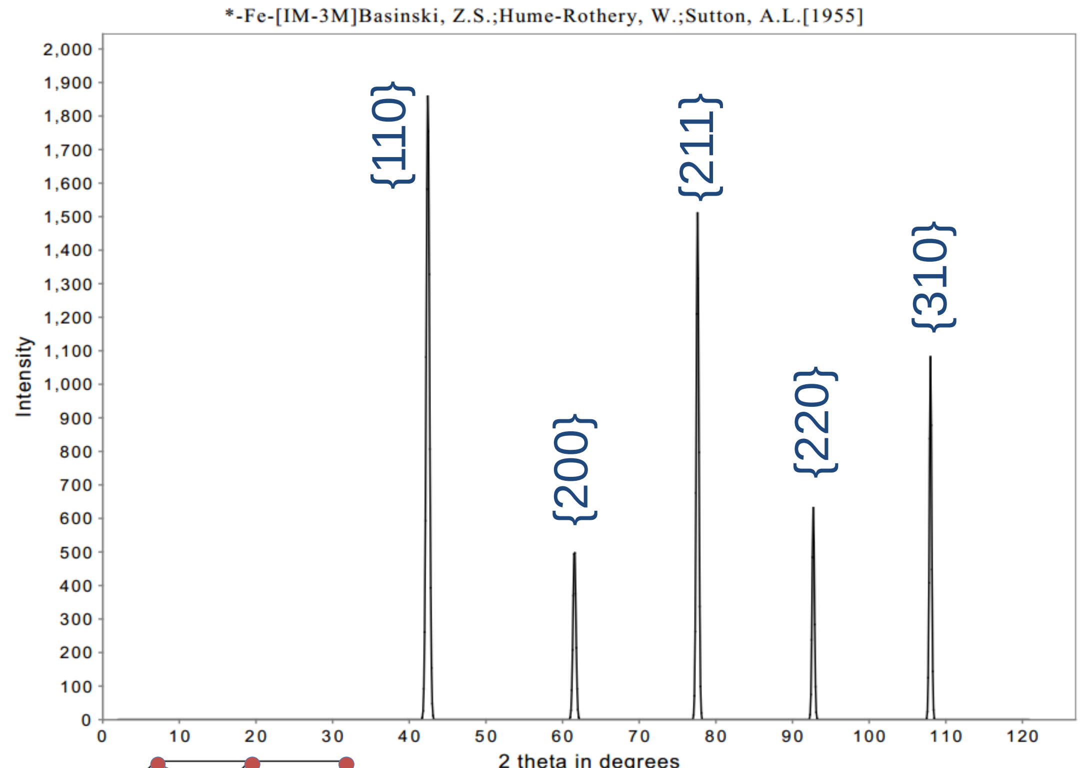


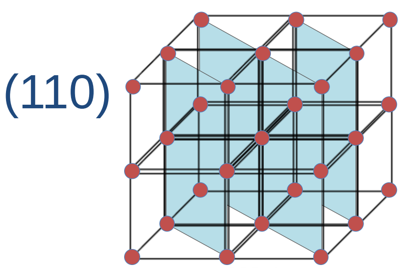
FIG. 25. The visibilities of some atoms and isotopes for X-rays and neutrons. The radii of the circles are proportional to the scattering amplitude *b*. Negative values of *b* are indicated by the cross-hatched shading.

G.E. Bacon

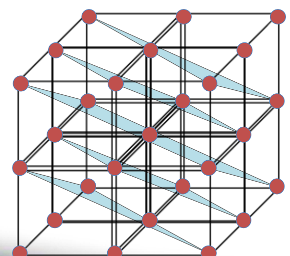
Crystal Structure and Diffraction Patterns







(112)



Calculated pattern for ferrite

$$n\lambda=2d_{hkl}\sin(\theta_{hkl})$$

Neutron Powder Diffractometer

C2 at NRU

- Variable incident wavelength, though typically select 1.33 Å and 2.37 Å (46.2 meV and 14.6 meV)
- Detector after the sample spans 80˚ with 800 elements

Powder Diffraction of Active Samples

Phase Analysis of Irradiated Fuel

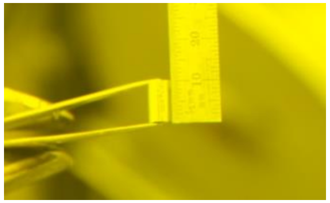
Near contact, gross γ-fields

Sample:

750 mSv/hr (75 Rem/hour).

Shielded Cell:

0.3 μSv/hr (30 mRem/hour)



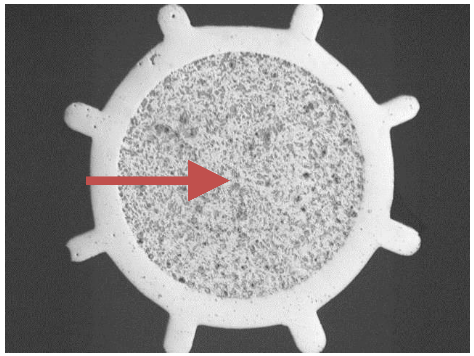
- 300 kg shielded cell
- 80 deg. exit window
- sample is *captive*

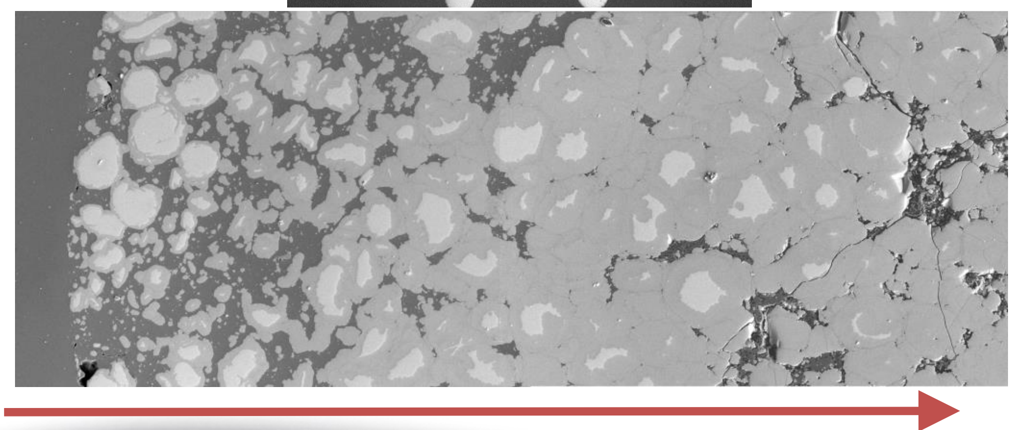
AECL EACL

INL Idaho National Laboratory

U-Mo Test Irradiation in NRU

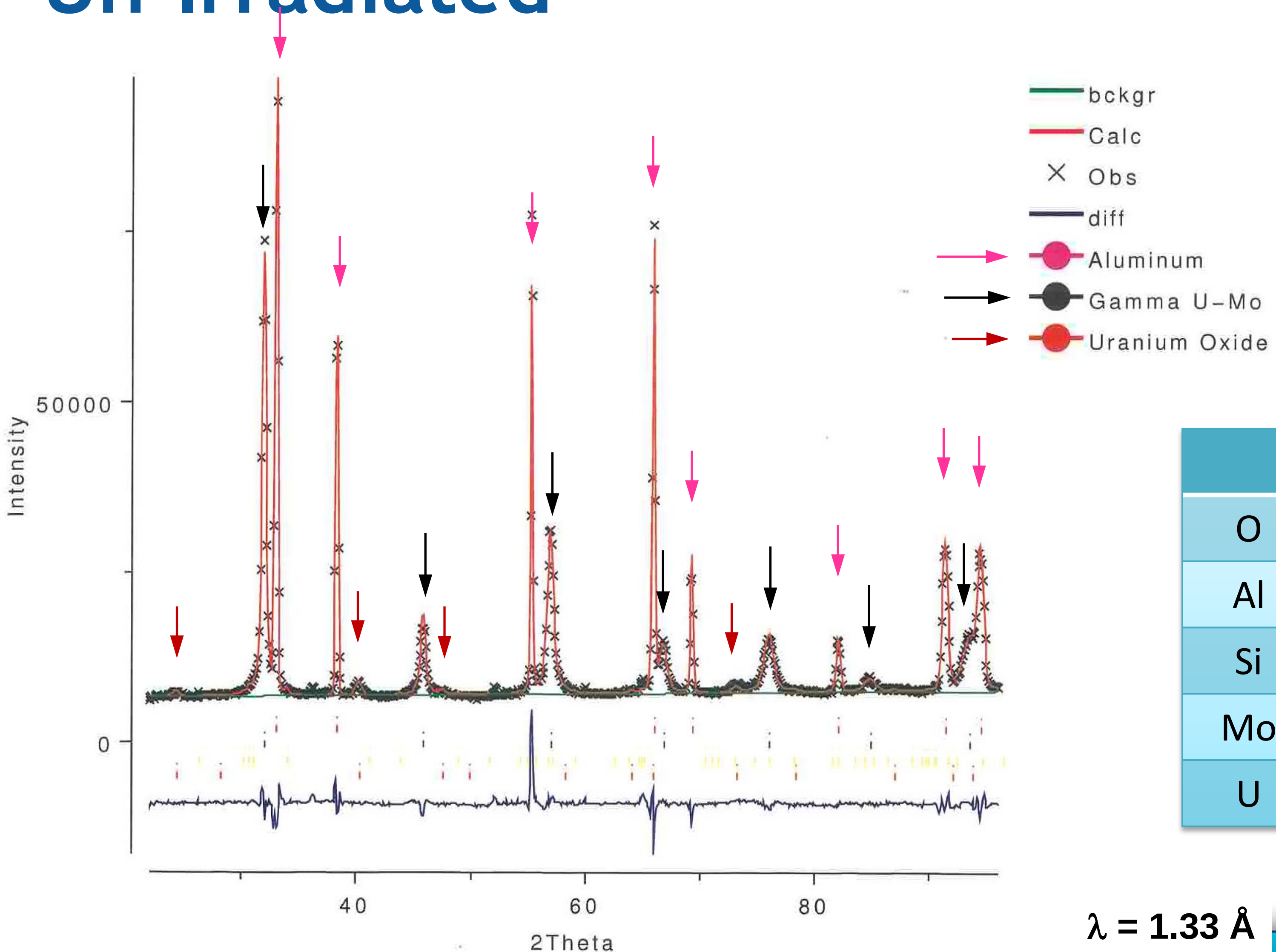
- Visually external appearance was fine
- U-Mo and Al matrix reacted inside: 6% volume swelling at 20% Burnup





60 at% BU Visual Exam, Both U-10Mo and U-7Mo fuel types intact

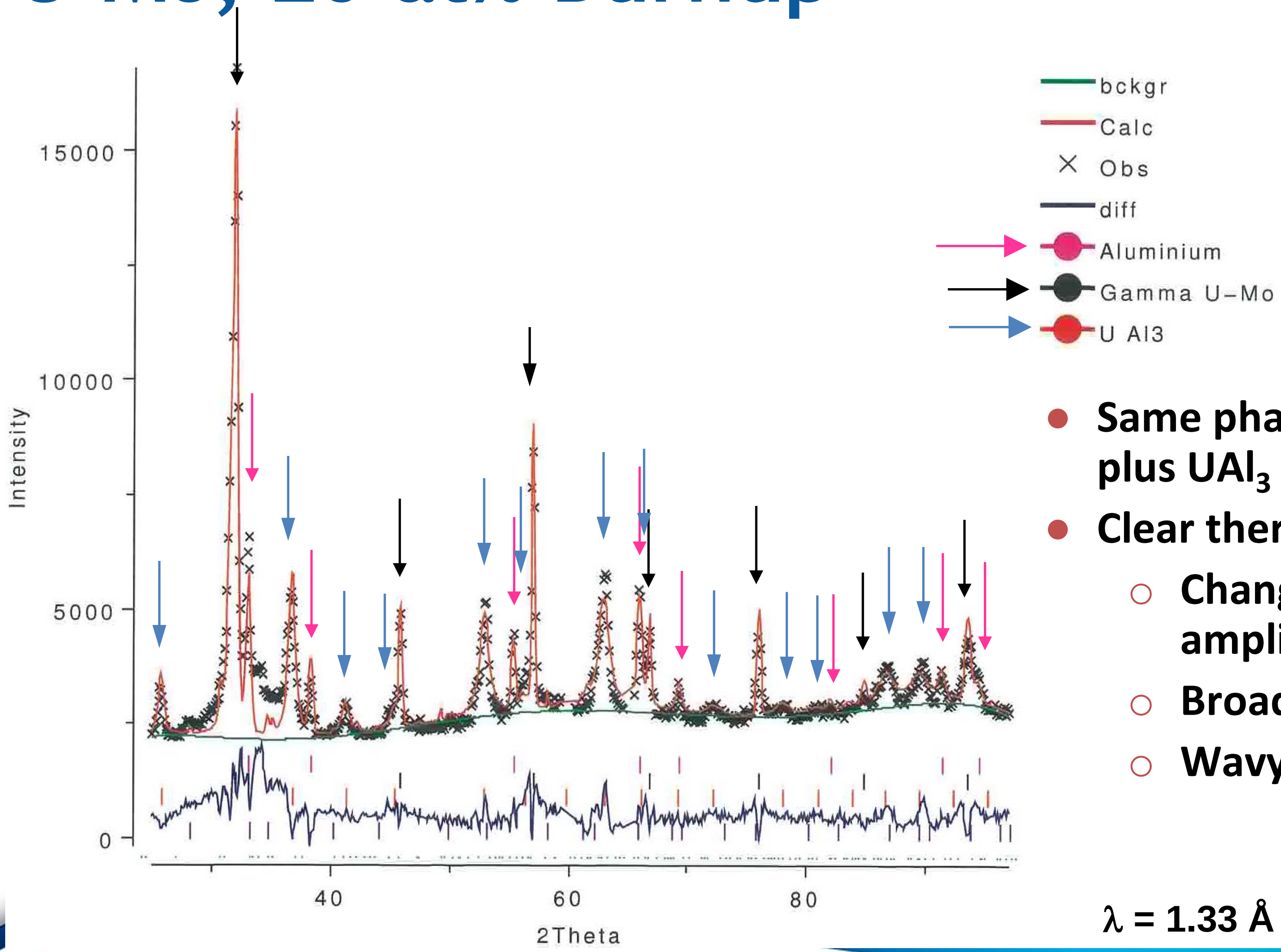
Un-Irradiated



	σ (barns)
O	4.2
Al	1.5
Si	2.2
Mo	5.7
U	8.9

λ = 1.33 Å

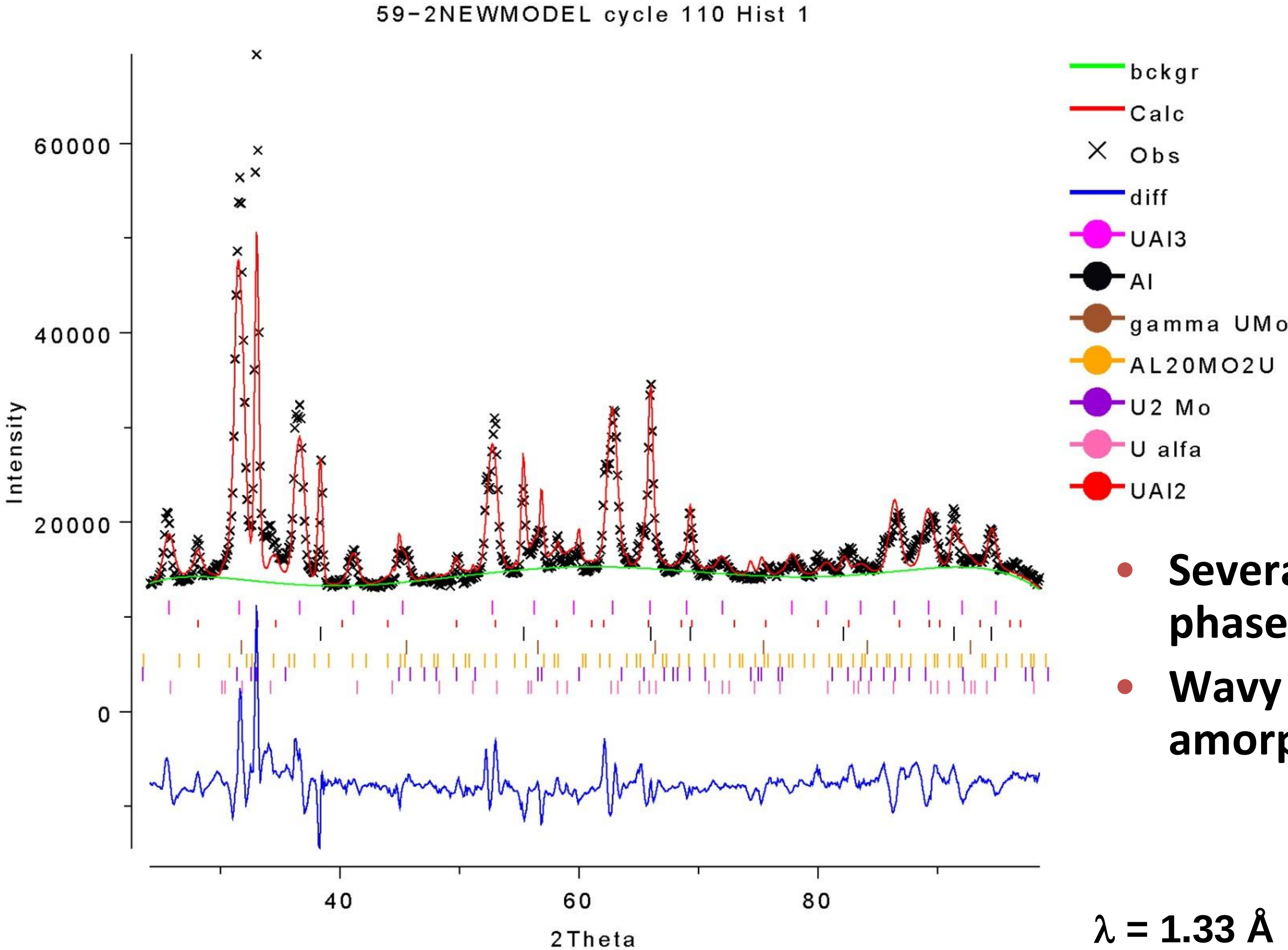
U-Mo, 20 at% Burnup



- **Same phases present plus UAl_3**
- **Clear there are changes**
  - **Changes in amplitudes**
  - **Broad features**
  - **Wavy background**

λ = 1.33 Å

U-Mo, 60 at% Burnup



- **Several identifiable phases**
- **Wavy background; amorphous?**

λ = 1.33 Å

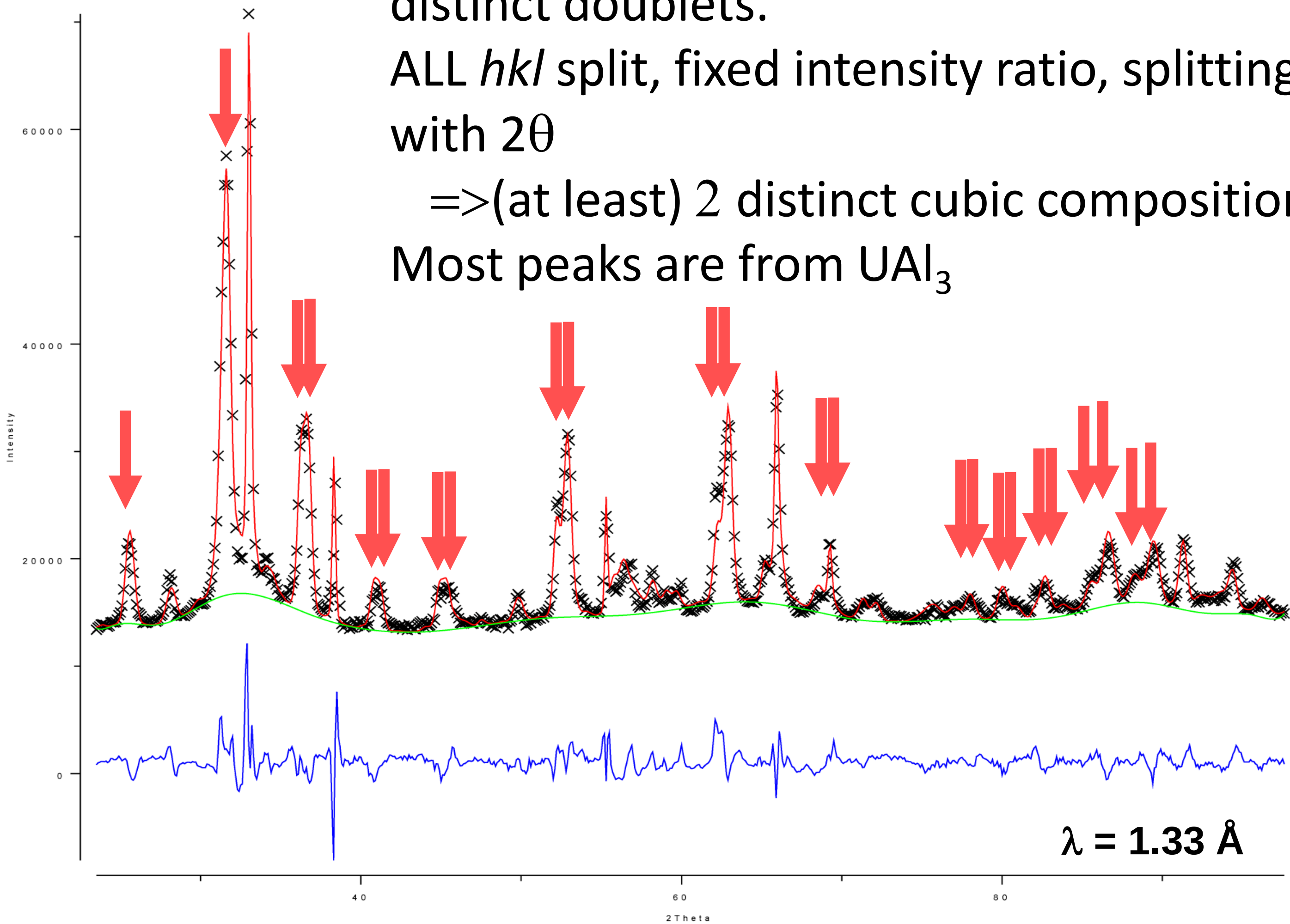
U-Mo, 60 at% Burnup

UAl_3 is cubic, and broad and in some samples has distinct doublets.

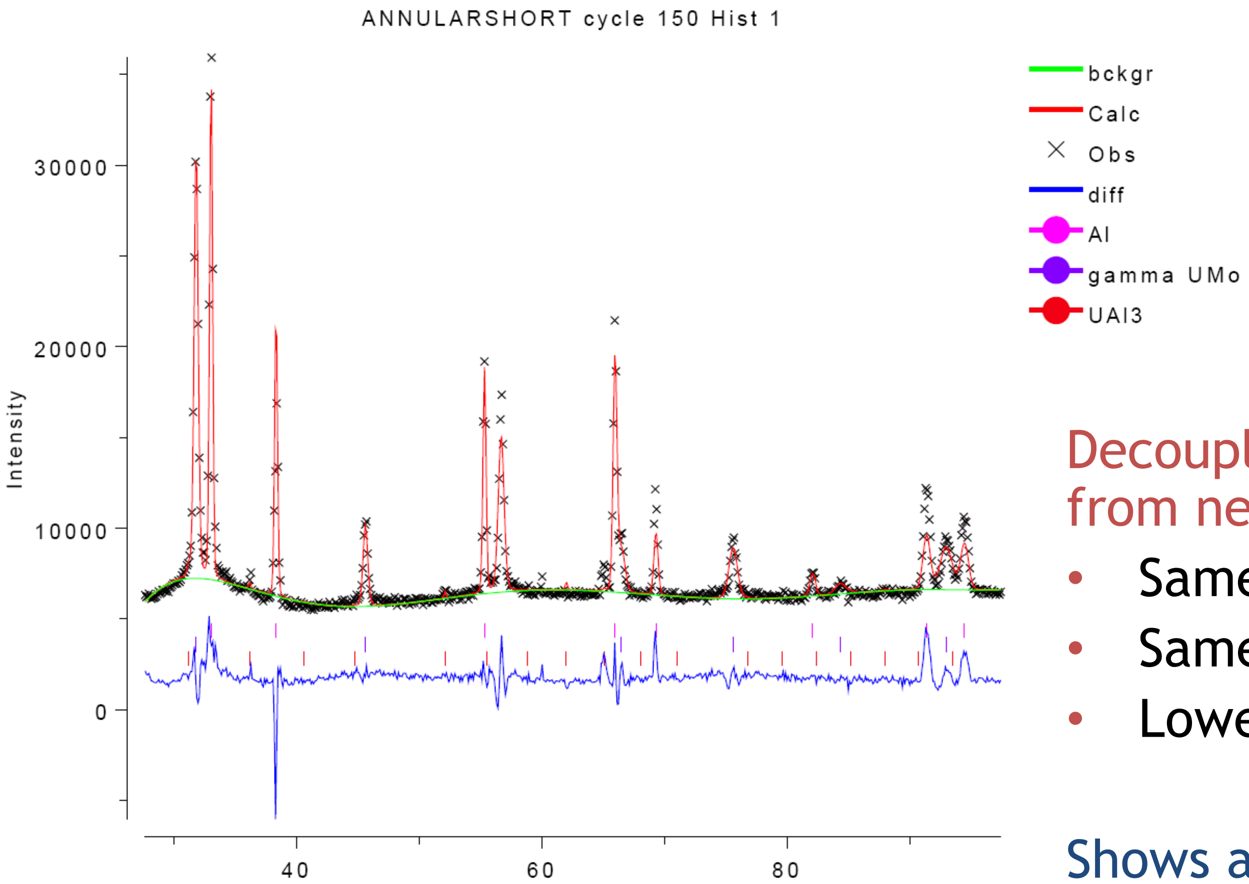
ALL *hkl* split, fixed intensity ratio, splitting increasing with 2θ

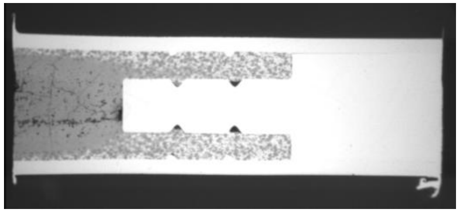
=>(at least) 2 distinct cubic compositions

Most peaks are from UAl_3



End-plug: 60% burnup at lower *T*, same *t*



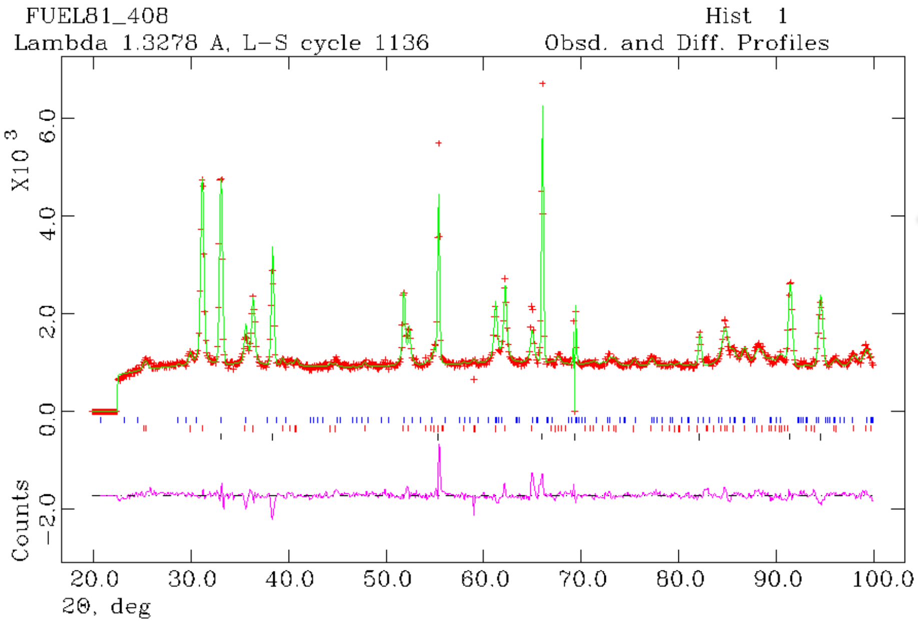


Decoupling temperature and time from neutron flux

- Same burnup
- Same time
- Lower temperature end

Shows assemblage close to that of 20% burnup

NDA Results From Al-U_3Si Fuel

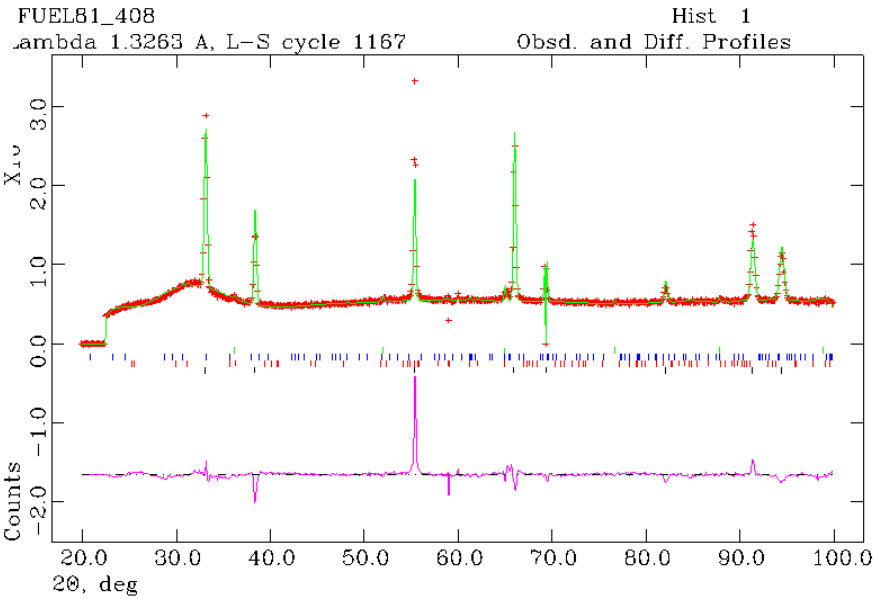


Unirradiated

See peaks that correspond to Al matrix, and U_3Si fuel

Irradiated to 90 at% Burnup

Peaks *only* from Al matrix

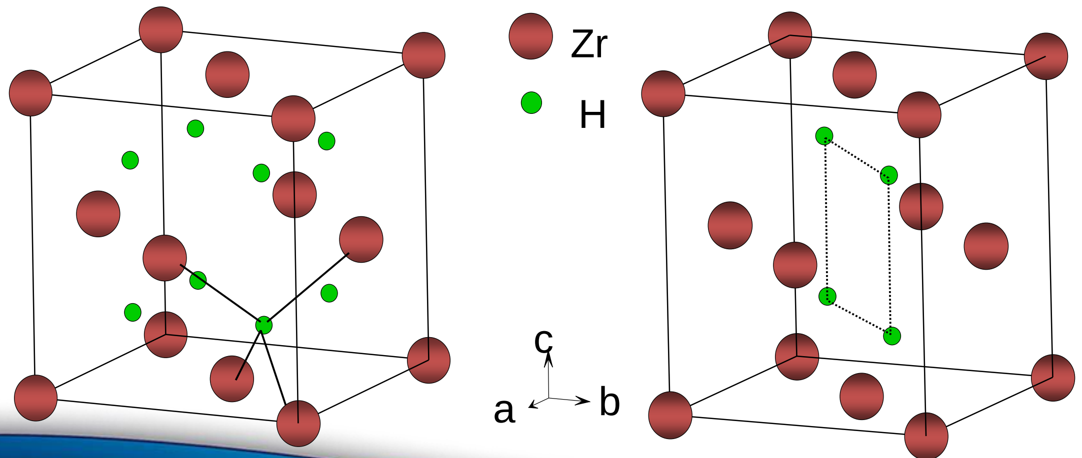


Hydrogen in Zr Alloys

Zr-2.5Nb

- δ-$ZrH_{1.6}$, a cubic cell of Zr with tetrahedral sites, randomly occupied by H
- γ-ZrH, a tetragonal cell of Zr with ordered, fully occupied H sites
- α–Zr, a hexagonal close-packed cell
- β–ZrNb, a body-centered cubic cell

δ phase hydride



γ phase hydride

Sidhu et al, Adv. Chem Ser. ***39*** *(1963) 87.*

Scattering Lengths

H incoherent

H coherent

Deuterium

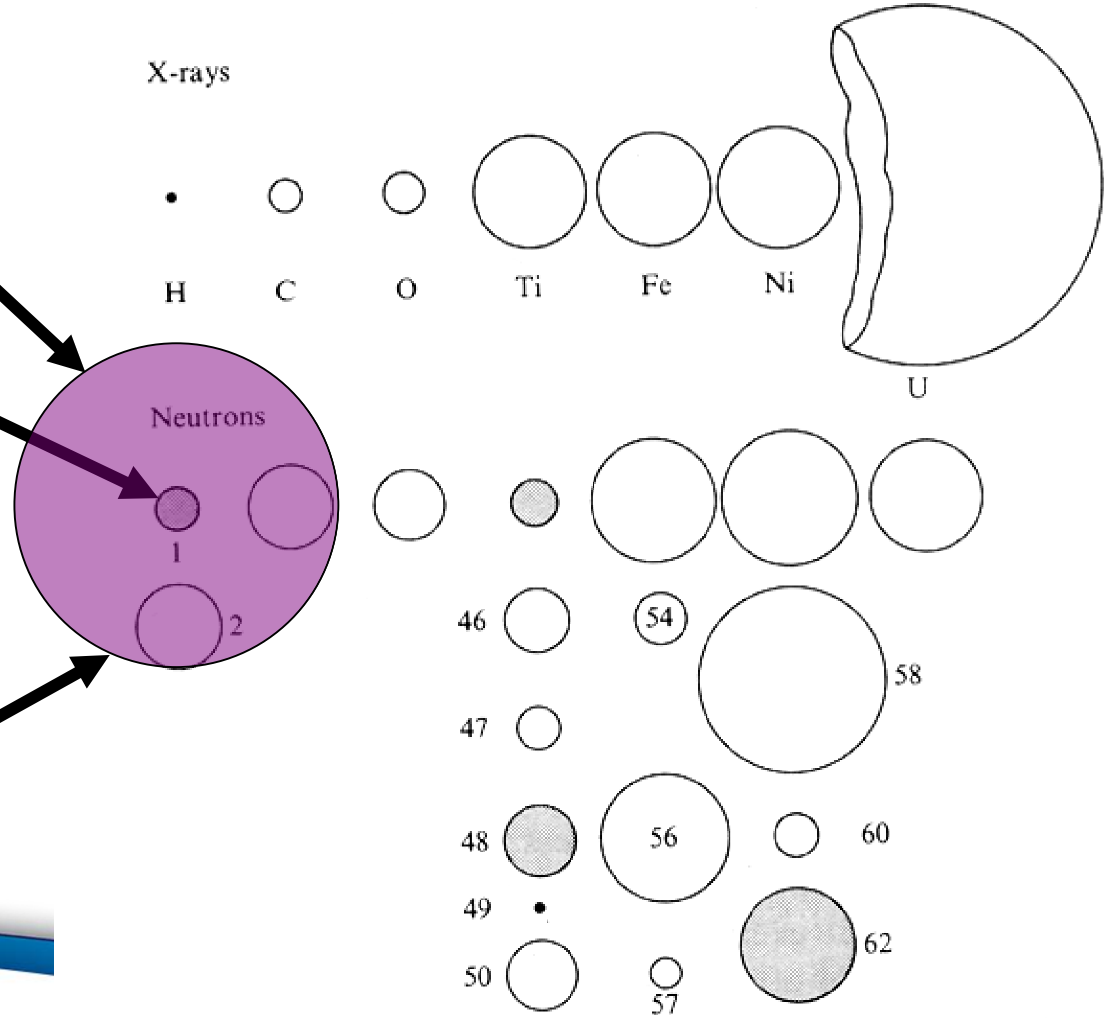


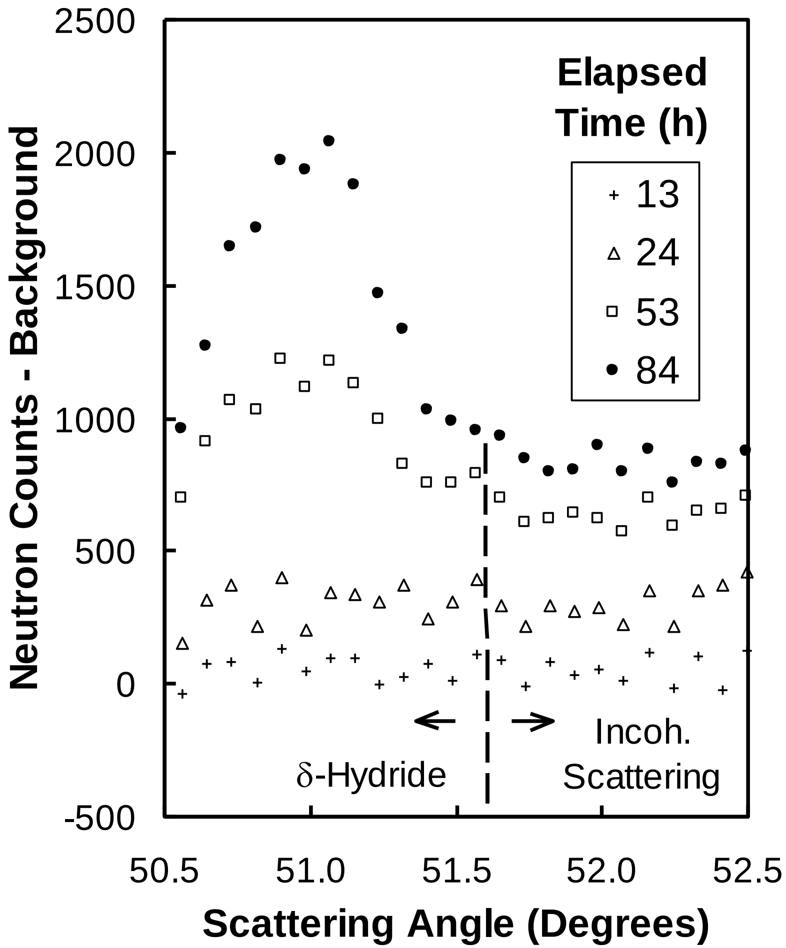
FIG. 25. The visibilities of some atoms and isotopes for X-rays and neutrons. The radii of the circles are proportional to the scattering amplitude b. Negative values of b are indicated by the cross-hatched shading.

Possible to mix + and – isotopes to vary “contrast”

G.E. Bacon

Hydrogen Ingress

T = 300 °C, Autoclaved Zr-2.5Nb

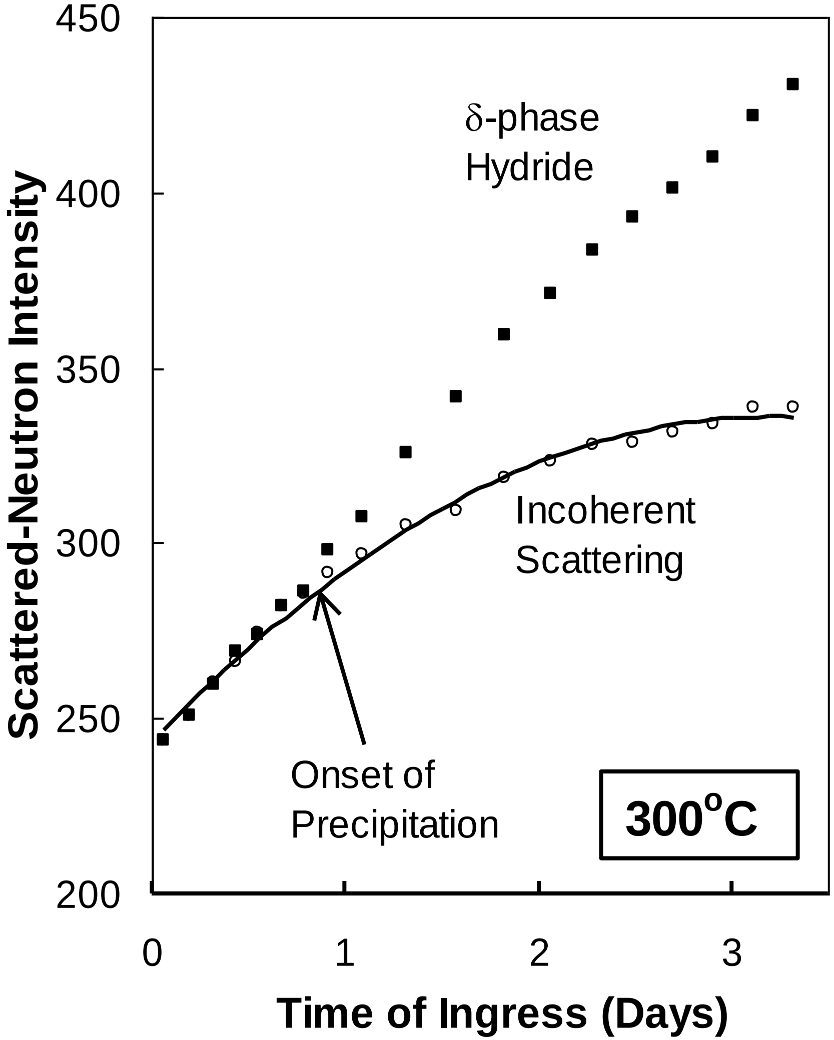


- Light hydrogen is absorbed by the Zr-2.5Nb specimen.
- The background increases with time, as incoherent scattering is proportional to the total amount of hydrogen in the specimen.
- When the solubility limit is exceeded, a diffraction peak indicates the presence of δ-phase $ZrH_{1.6}$ precipitates.

Hydrogen Ingress

T = 300 °C, Autoclaved Zr-2.5Nb

- During the lifetime of zirconium alloy pressure tubes, there is gradual accumulation of hydrogen
- Exceeding the solubility threshold, zirconium hydrides appear and can deteriorate material performance

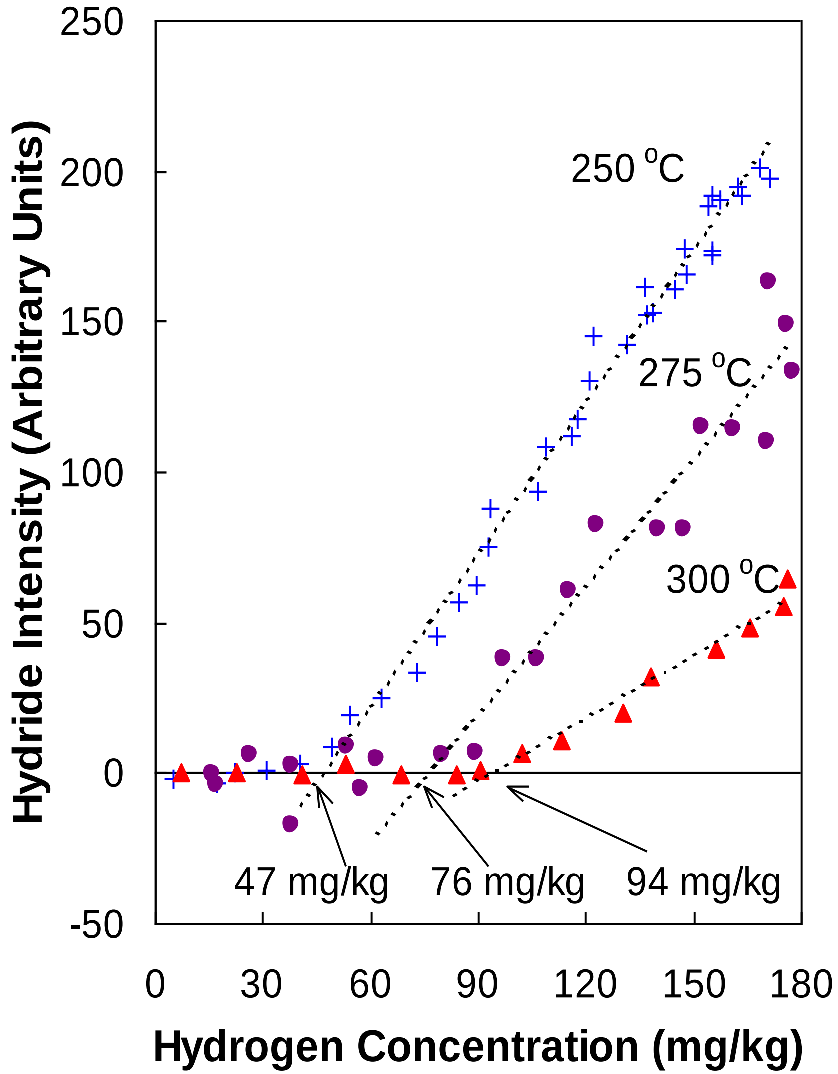


Physica B ***241-243*** *(1998) 1181.*

Hydrogen Ingress

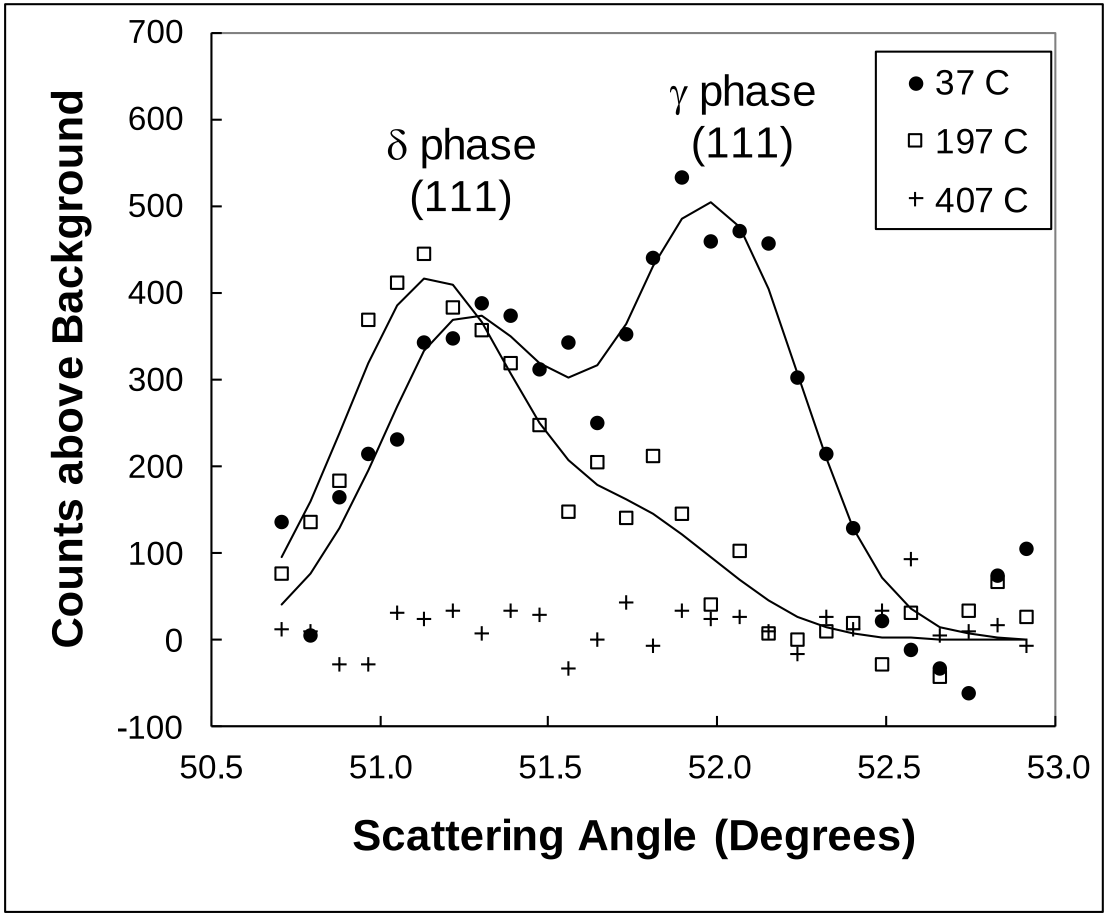
T = 300 °C, Autoclaved Zr-2.5Nb

- Definitive measure of onset of precipitation vs concentration at given temperature.
- Fundamental knowledge could be applied to refine fitness-for-service guidelines.



Hydride Dissolution

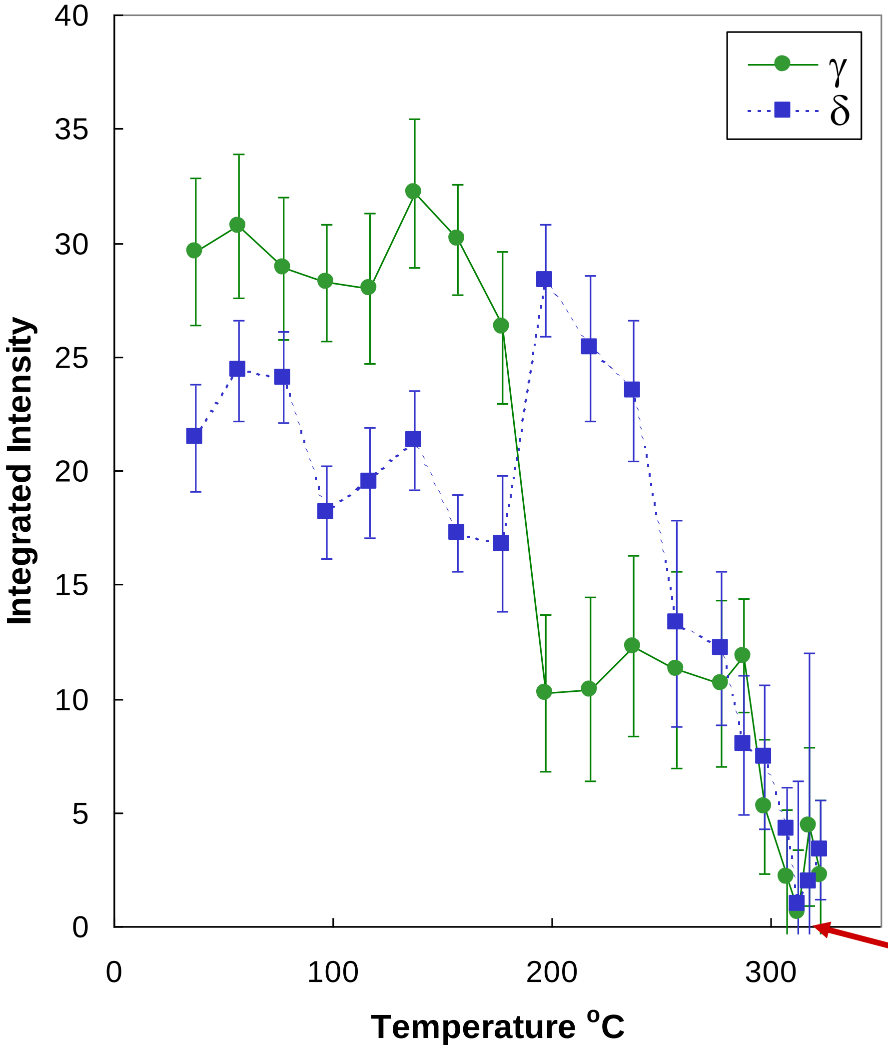
Heating Experiments



As temperature increases, first the γ phase dissolves, then the δ phase dissolves.

Phase Transformation

Seems to be about 180±10 C



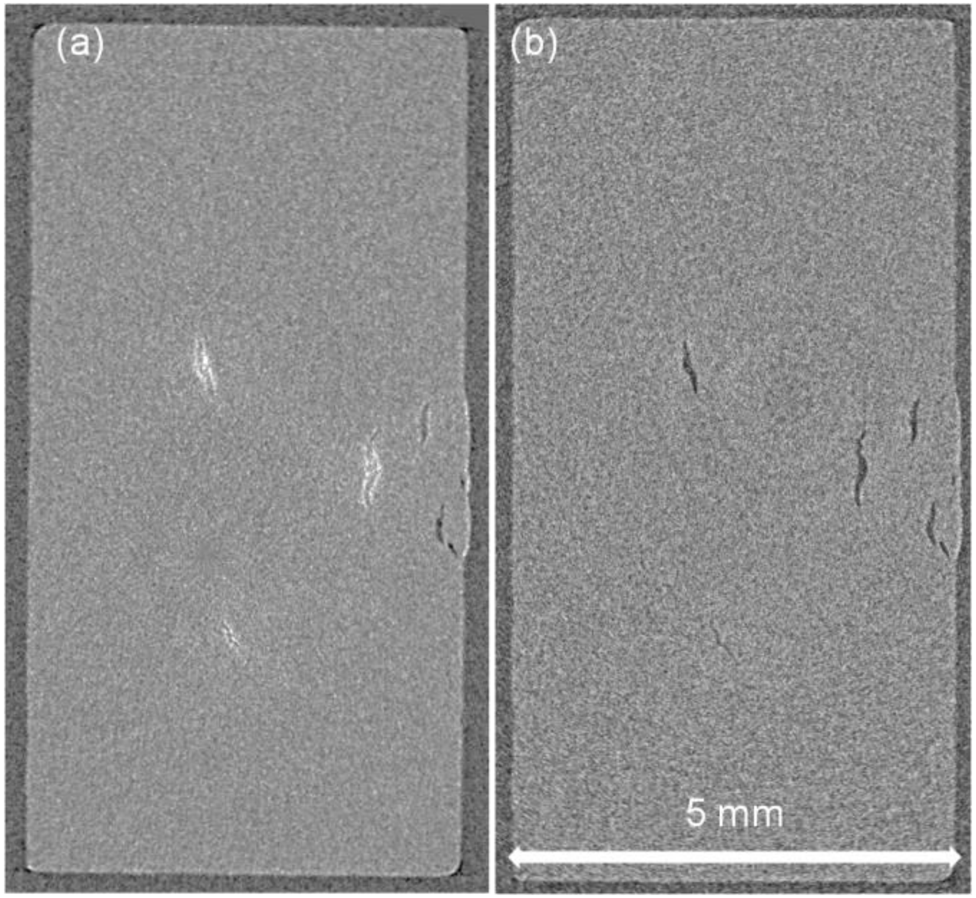
Zr-2.5Nb alloy matrix was in the typical reactor conditions:
α–phase Zr grains in a continuous network of Nb-rich β–phase metal with 10x solubility of hydrogen.

Both hydride phases were present at the start of this heating experiment.

TSSD can be determined ±3 °C

J.Nucl.Mat. ***232*** *(1996), 75-85.*

Imaging Hydrogen Distribution in Zr-Alloy



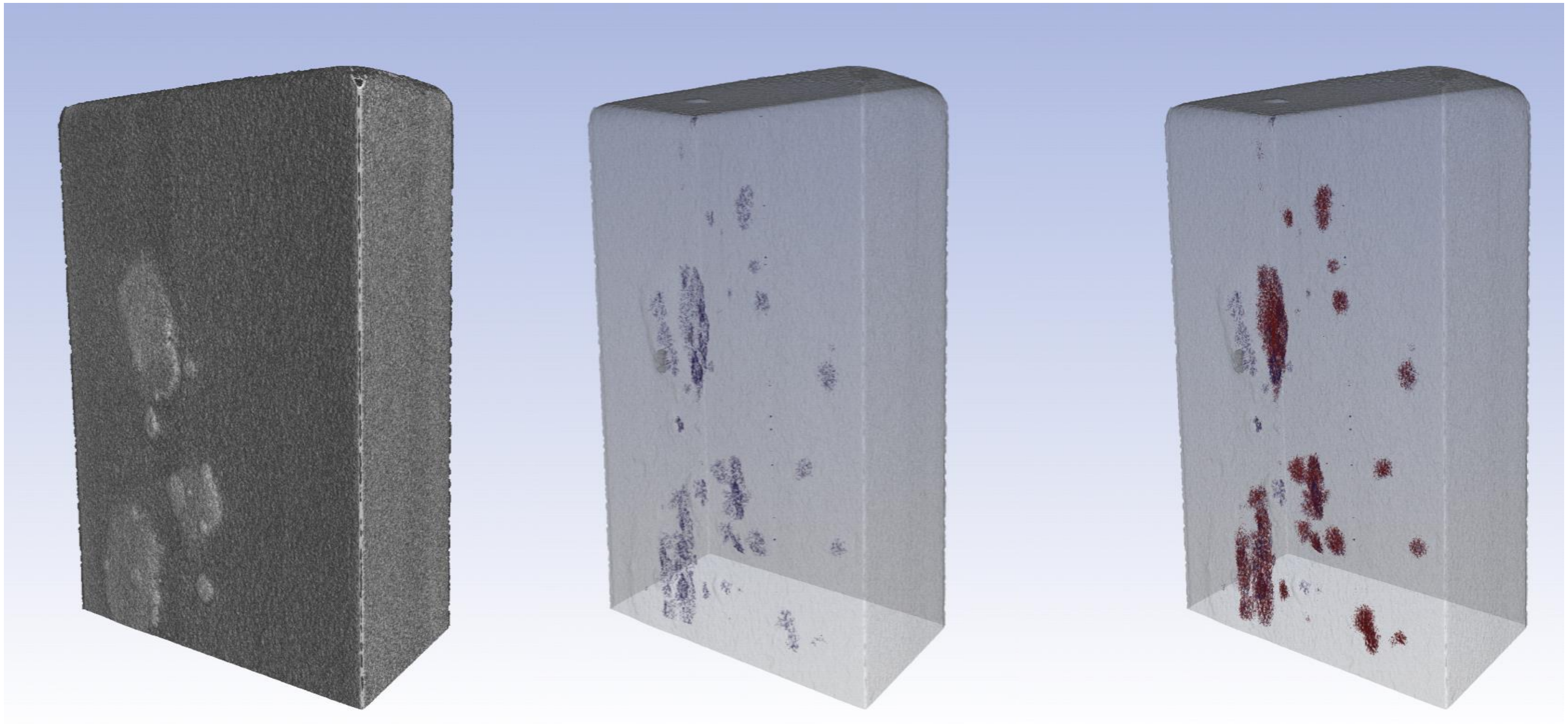
- H clustered near cracks before (a) annealing
- After (b) annealing, H is dispersed

(b): 48 h @ 50 °C (ISO 3690)

BAM Federal Institute for Materials Research and Testing

Courtesy of Axel Griesche, BAM

Neutron Tomography



surface

cracks

cracks + H

Courtesy of Axel Griesche, BAM

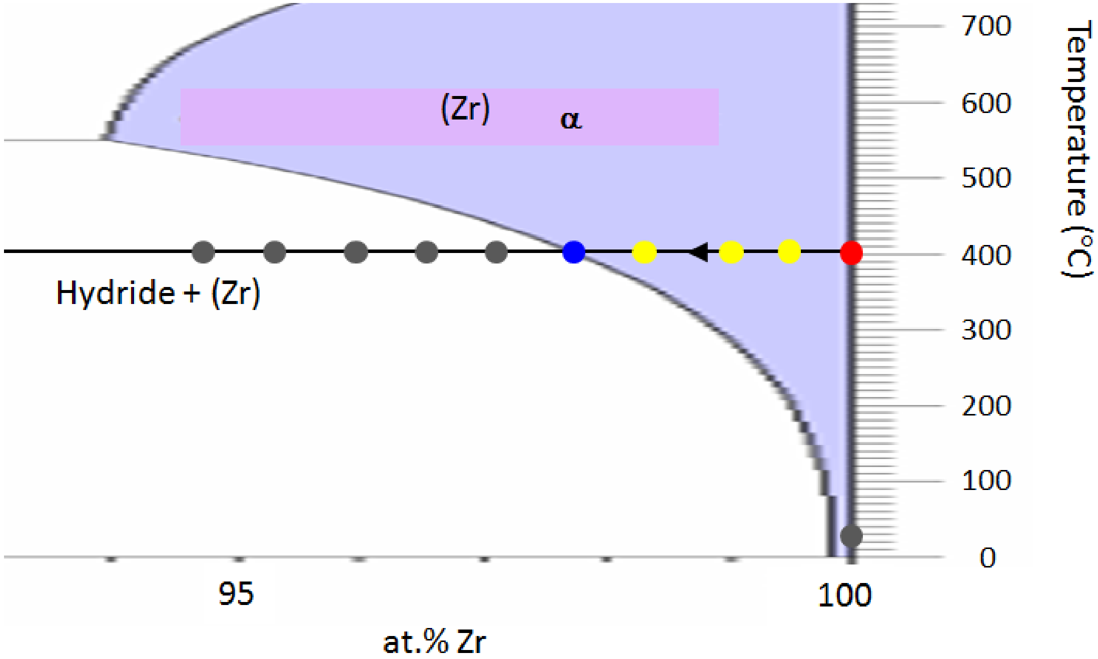
BAM Federal Institute for Materials Research and Testing

Canadian Nuclear Laboratories | Laboratoires Nucléaires Canadiens

Hydrogen in Zr and Effect of Stress

Objective:

Investigate the effect of stress and thermal cycling on hydride dissolution, precipitation, orientation, and morphology.

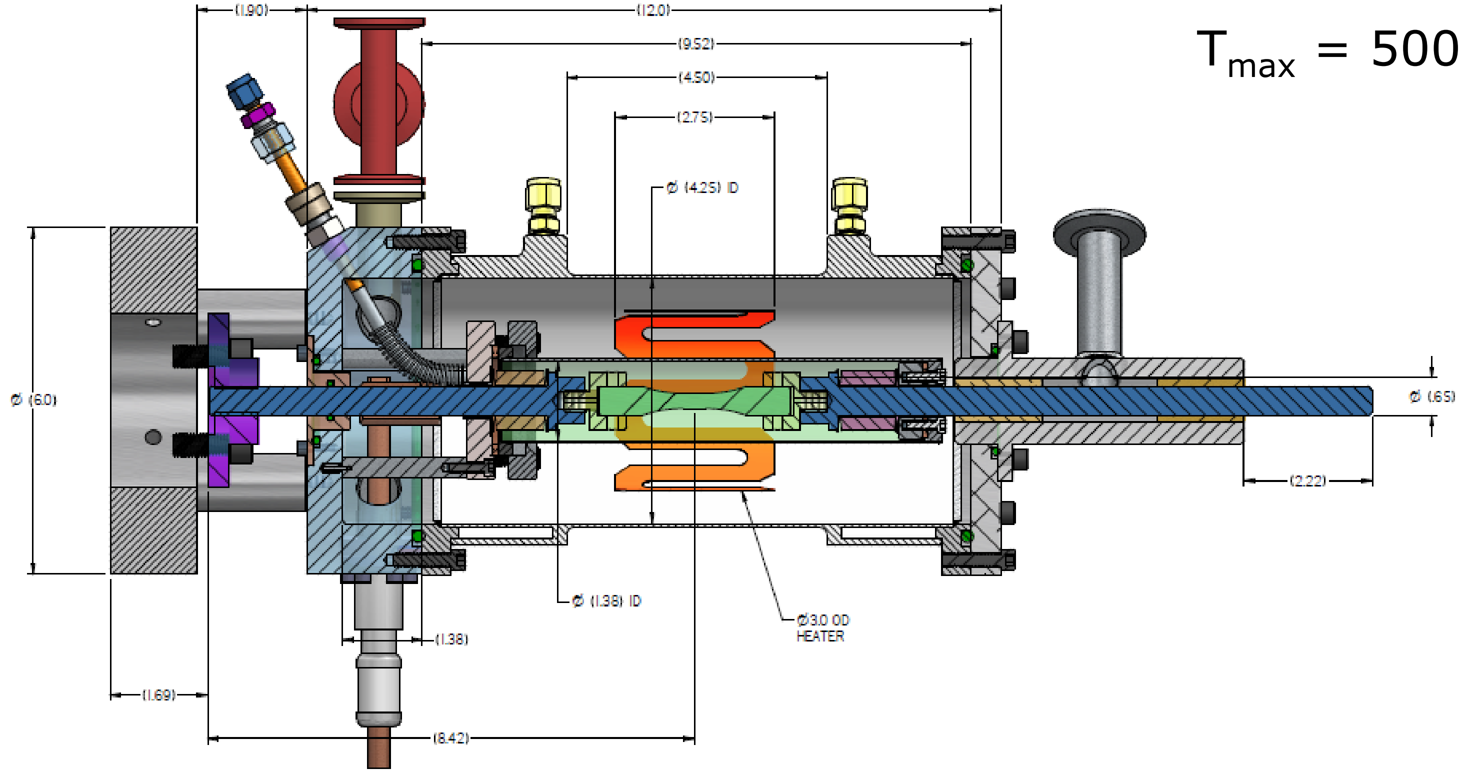


Output:

Hydrogen solubility curves for Zr-2.5Nb for a range of thermo-mechanical cycles.

Hydrogen in Zr and Effect of Stress

A unique controlled-atmosphere furnace has been fabricated, commissioned and interfaced with the neutron spectrometer control system. Furnace is not available commercially.



T_{max} = 500°C

Mystery Flask

Found unidentified legacy lead flask with unknown radioactive materials

- In-field inspection, only ^{137}Cs γ-rays
- Preliminary examination at the Universal Hot Cells:
  - Flask contained four capsules in a chandelier holder
  - Capsules had radiation fields up to 63 Rem/h (0.63 Sv/h) near contact
  - Each capsule ~ 2.5 cm x 14 cm
  - Weight: 180 g – 300 g
  - *Active length* ~ 5 cm

Identifying the Origin & Composition

- Preliminary examination of capsules (mass, appearance, & radioactivity) has not been able to identify the contents
- A search of available records has not resulted in any significant clue as to the contents of the capsules
- Fissile Content and physical configuration required for safe and secure disposition by Waste Management

But…

Catch 22!

- Fissile Content and physical configuration required to reduce risk when opening capsules in Hot Cells

⇒Non-traditional non-destructive nuclear forensics techniques

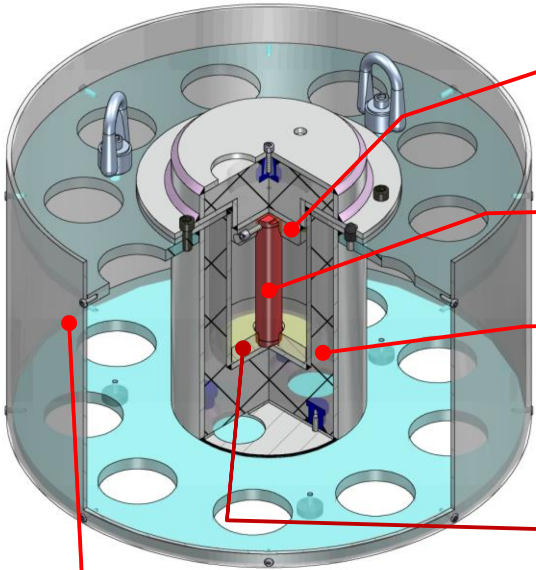
X-RAYS & NEUTRONS TO THE RESCUE

What We Did and How

- Solved a practical problem for the disposition of unknown nuclear (fissile) material
- Demonstrated *non-destructive* examination techniques for proposed use in Nuclear Forensics
    - X-Ray Fluorescence
    - Neutron diffraction
    - Neutron imaging
    - Delayed neutron activation analysis

Measurement on Radioactive Samples

On-Site Flask

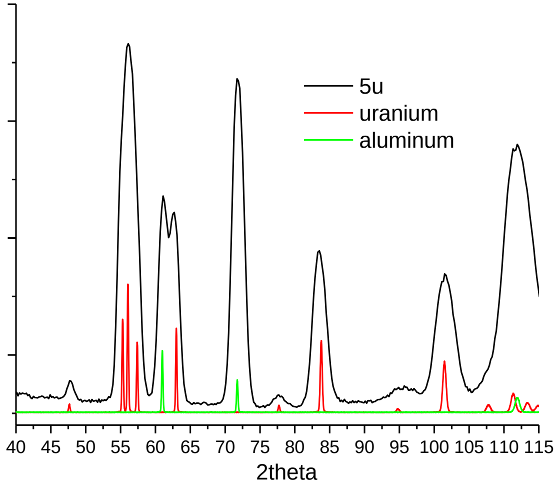


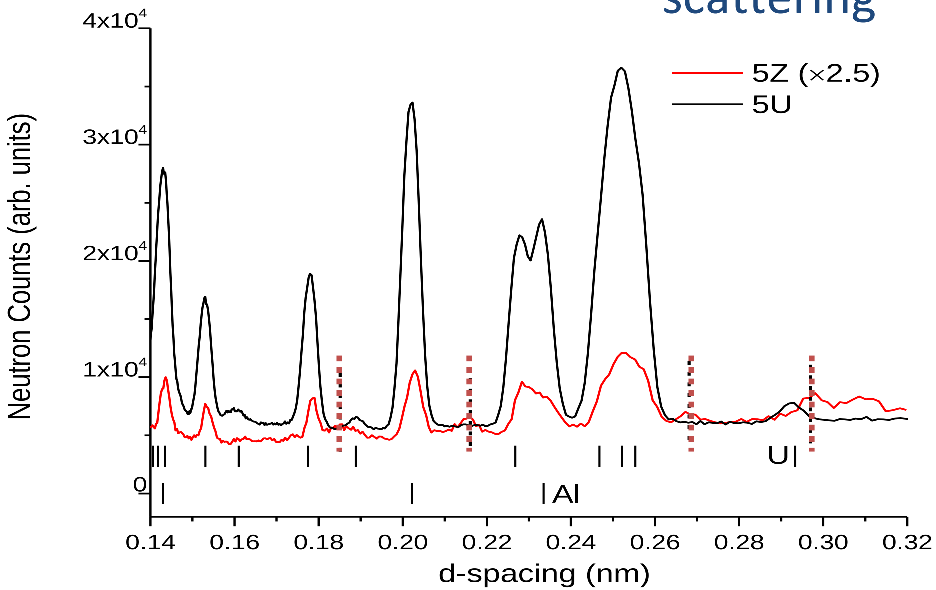
Depth (mm) to reduce beam intensity to 10%

Element	Neutrons	X-Rays (0.154 nm)
Be	24.9	9.372
C	37.3	1.886
Al	220.4	0.175
Fe	19.1	0.009
Cd	0.2	0.011
Pb	61.9	0.008

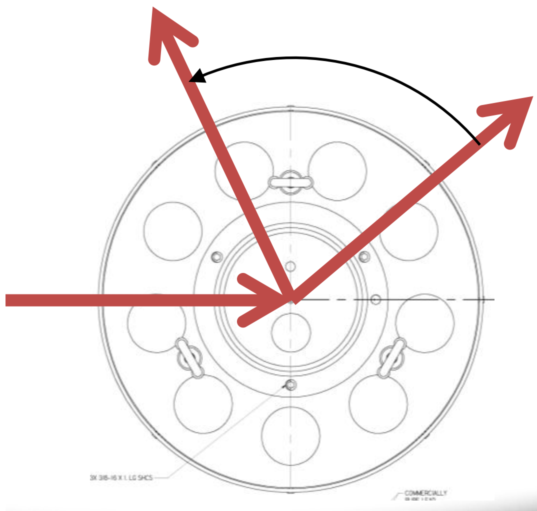
Neutron energies ~meV
X-ray energies ~keV

All Peaks Identified

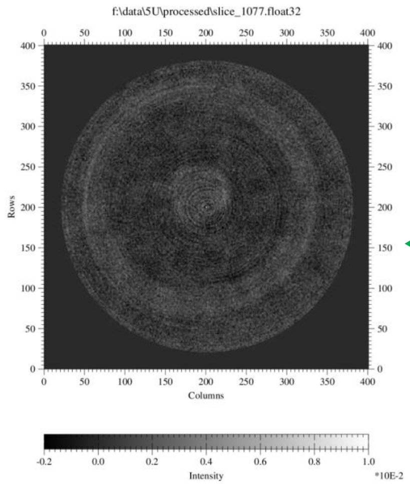


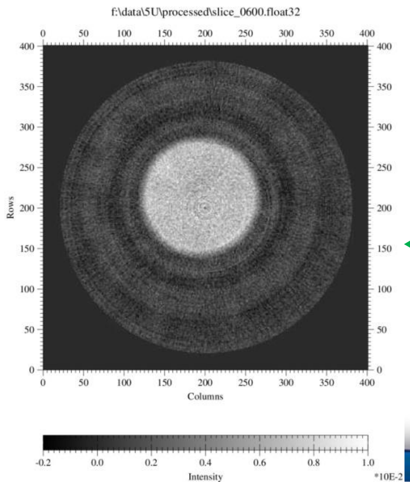


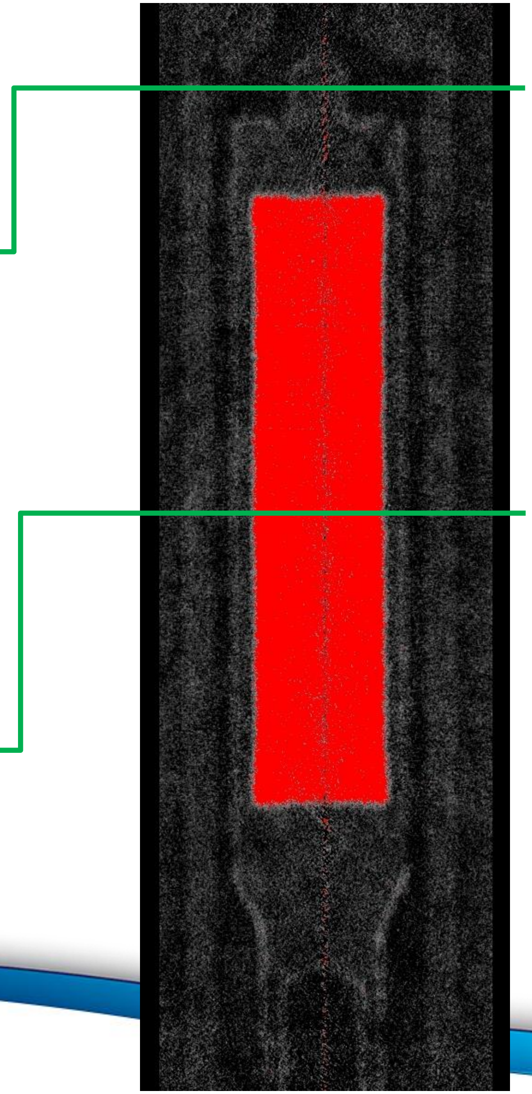
- Peaks are all indexed as λ, λ/2 & λ/3 peaks of:
  - U and Al for capsules 5U and 5Z.
  - Th and Al for capsules 8X and 8W.
- Al textured, U & Th random
- No clear evidence of liquid or amorphous scattering

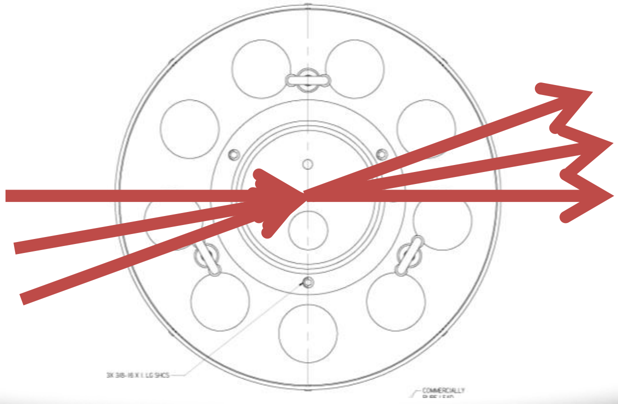


5U Tomographic Slices

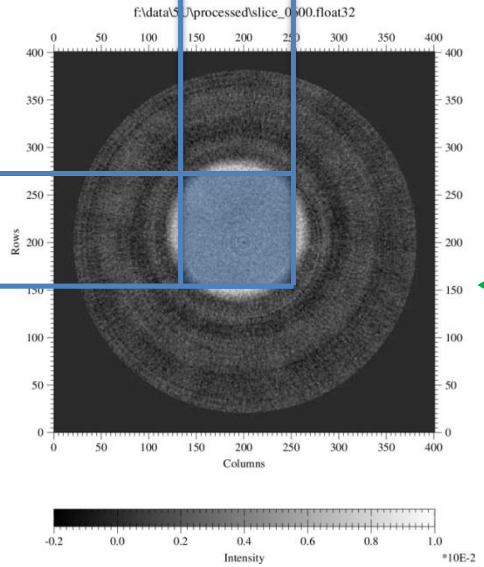


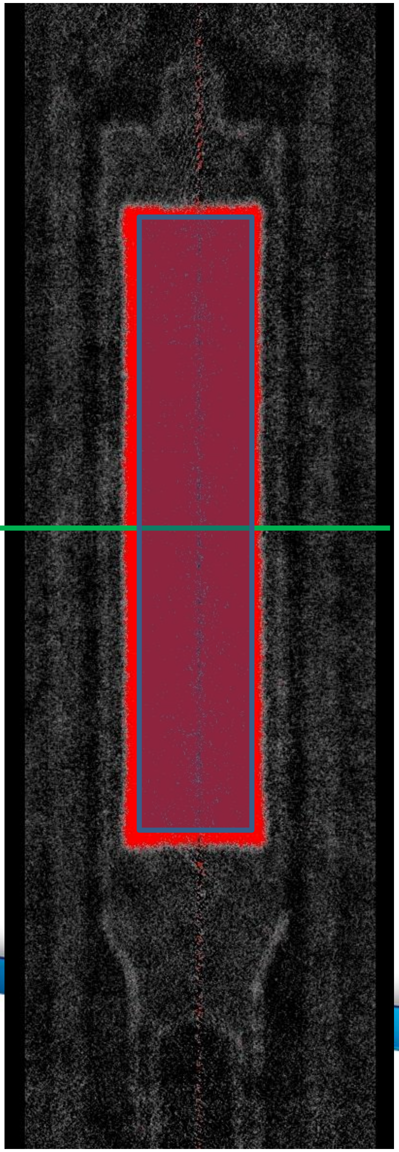


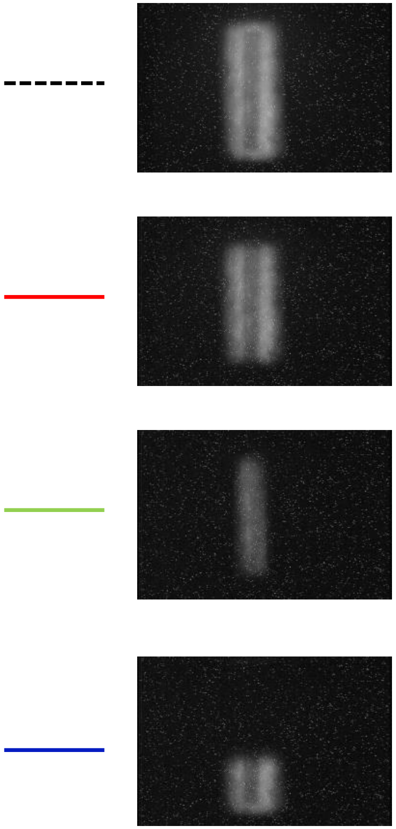




Spatially Refining Diffraction Data



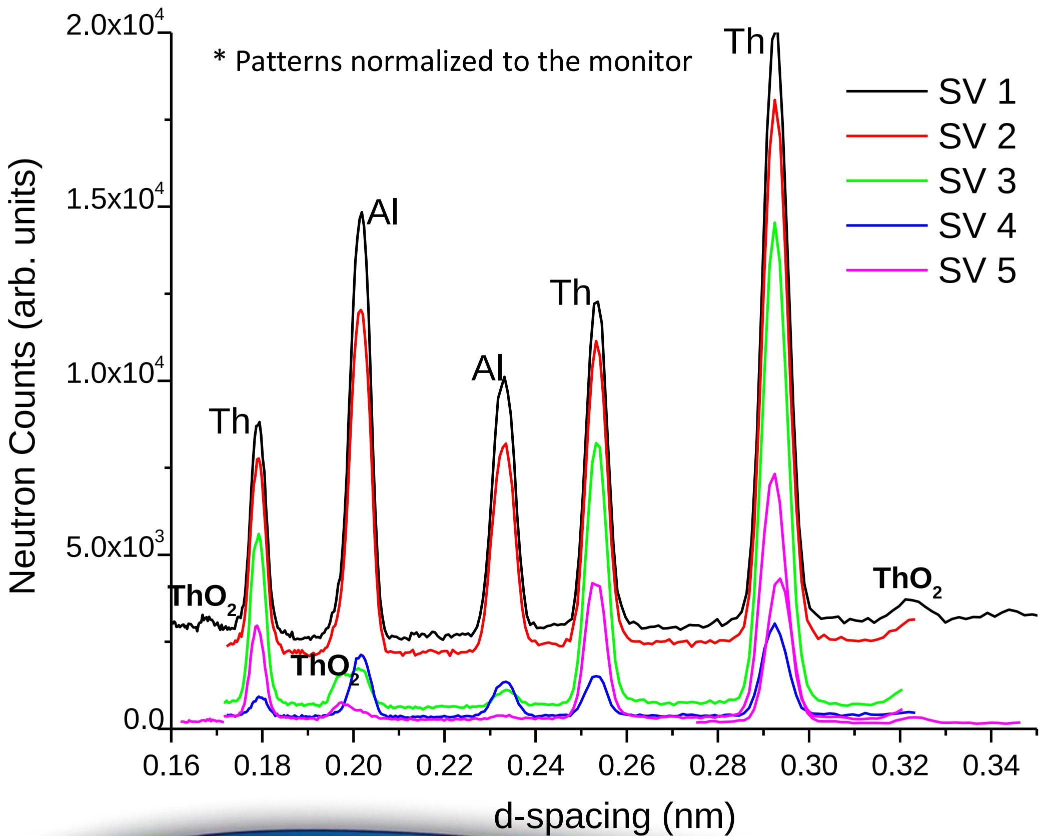


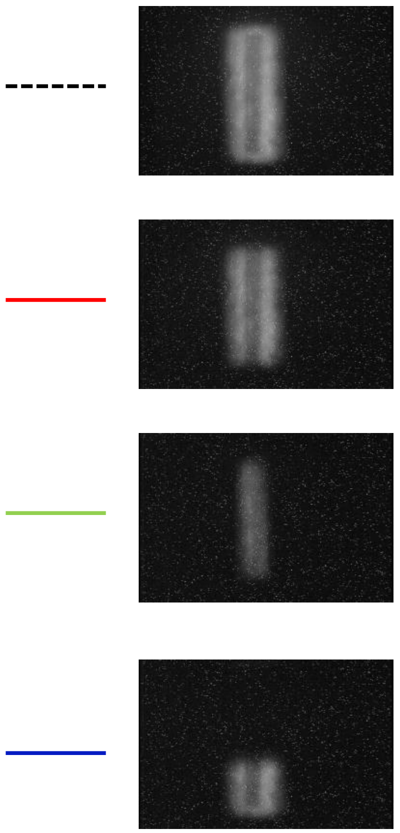


inner part of the core

Spatial Differentiation (8W)

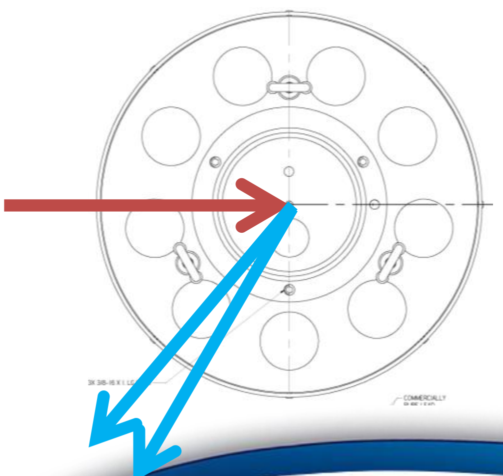
No Al in Core

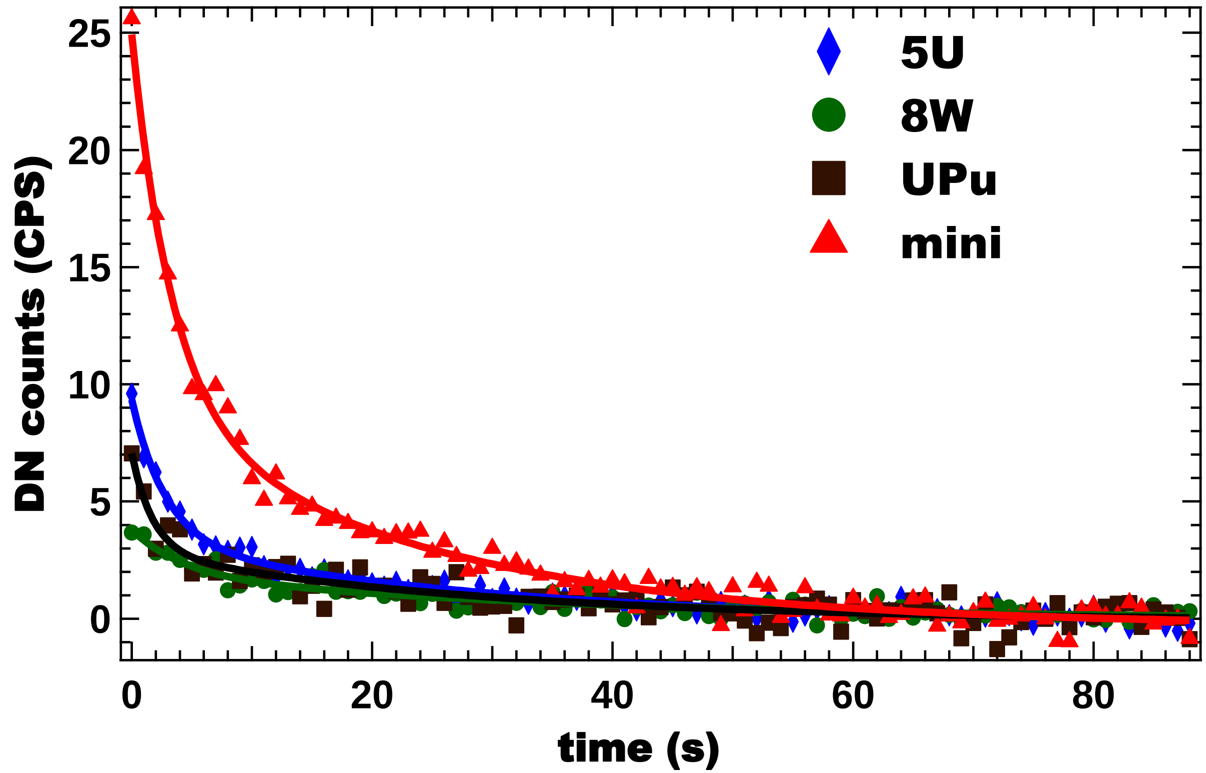




Delayed Neutron

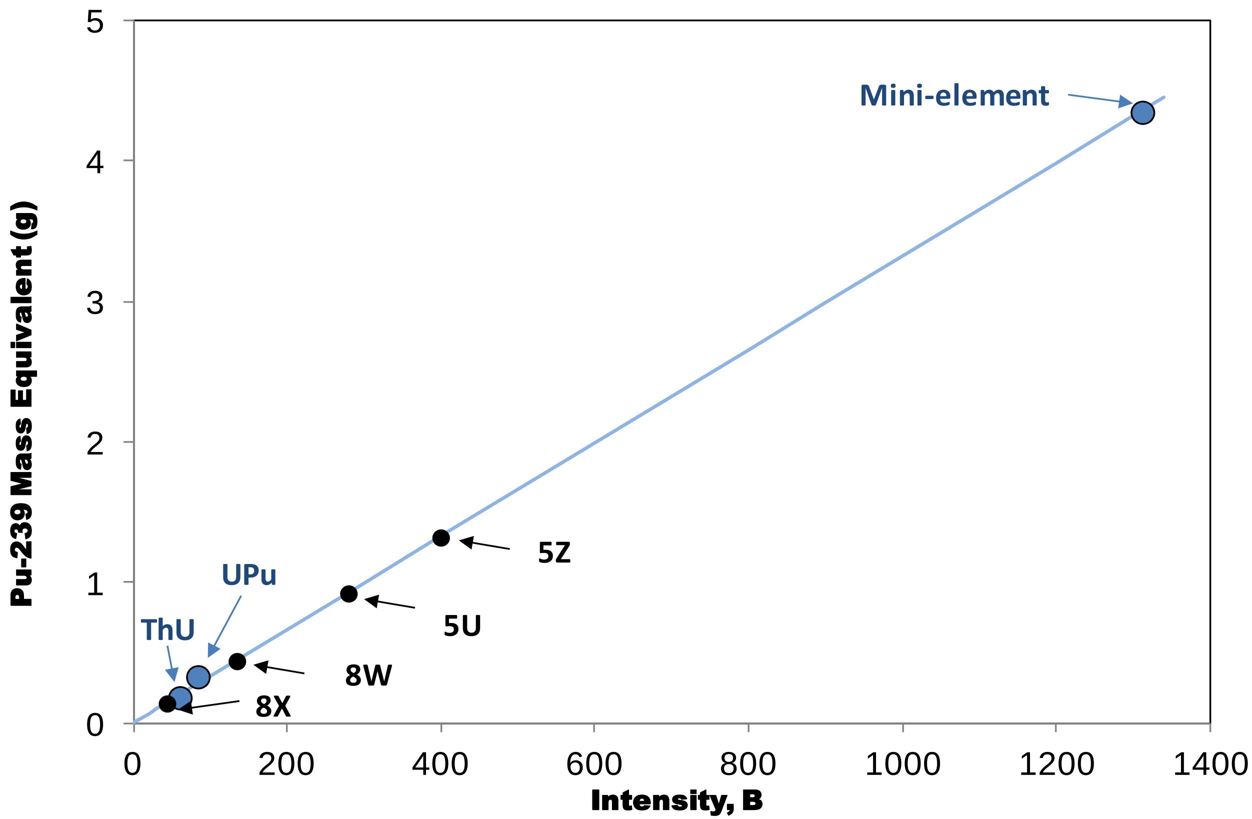
- Counts per unit time
- Corrected for Pb attenuation
- Corrected for volume
- Clearly less fissile material than U_3Si fuel (mini-element)



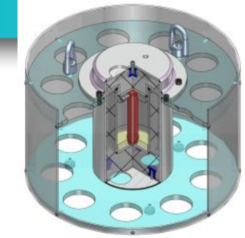


Cannot clearly identify different time constants in order to distinguish elements yet alone isotopes

Can Estimate Fissile Mass

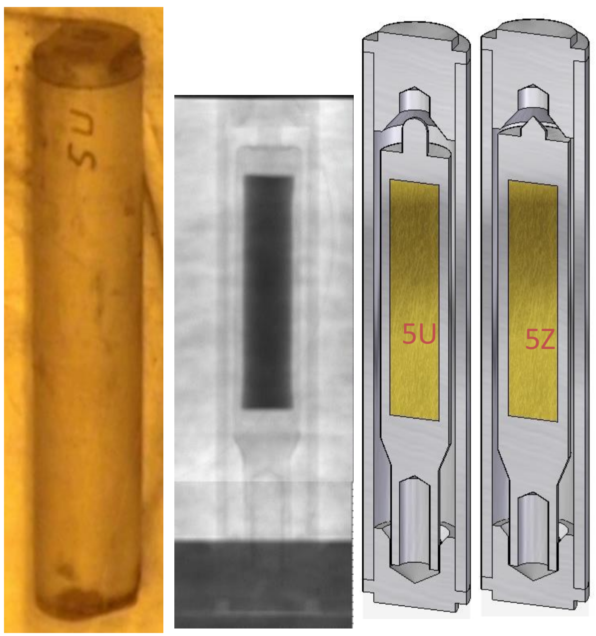


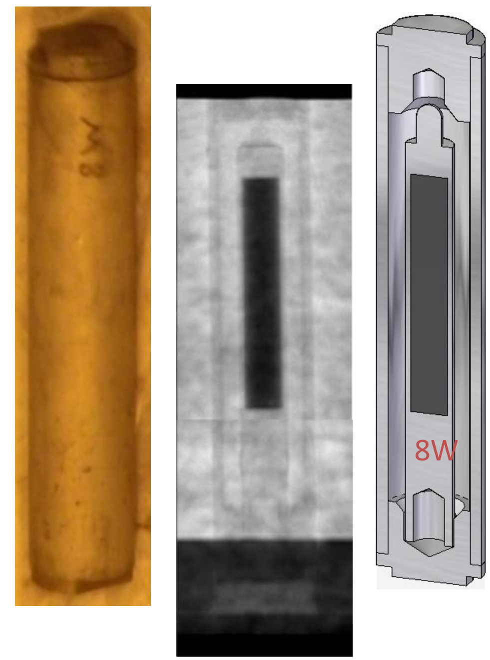
Use known sample results as calibration and determine effective ^{239}Pu mass of unknown sample from the calibration line



Mystery Solved

(A Little Reverse Engineering)





5U & 5Z		
Measured	Calculated	U
296 g 300 g	298±26 g 305±26 g	146±12 g <1.8 g ^{239}Pu

8X & 8W		
Measured	Calculated	Th
180 g 180 g	180±15 g 180±15 g	47±4 g <0.9 g ^{233}U

Rogge, R.B. *et al.,* Proc. of Pacific Basin Nuclear Conference (2014), paper 310.

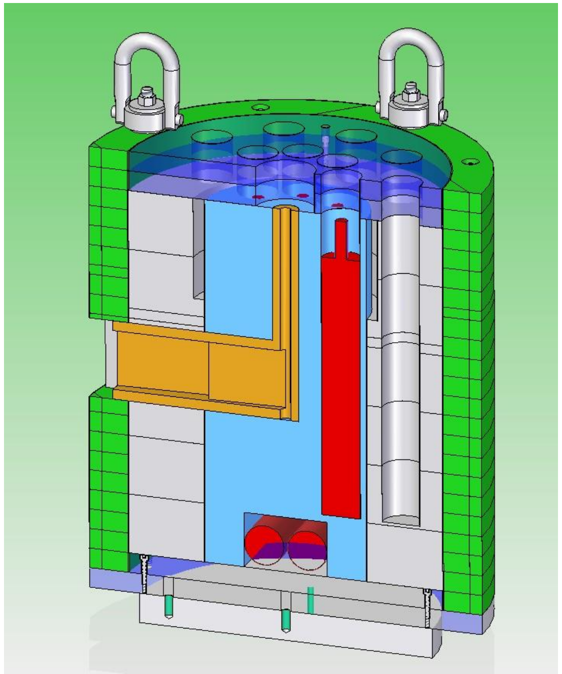
In-Beam Delayed Neutron Analysis

One-time demonstration?

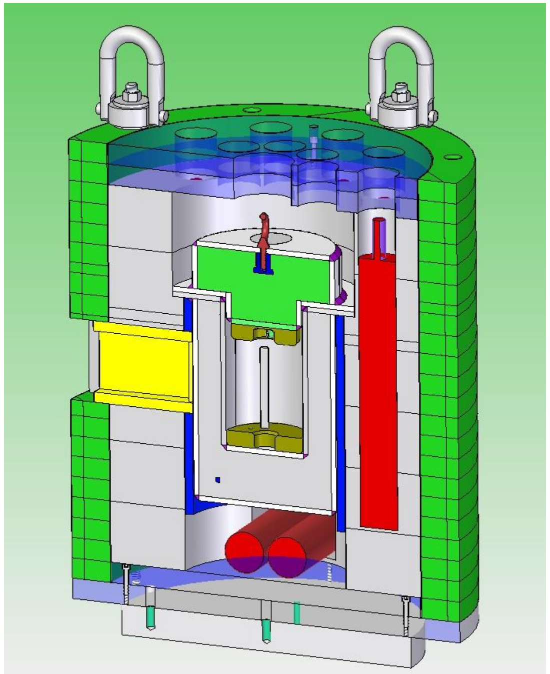
- Demonstrated that in-beam delayed neutron (DN) is possible and met minimum objective—fissile material present and upper limit on mass
- In-beam advantages:
  - Large samples
  - Early time data
  - Unknown samples
  - Avoid ^{238}U correction
- Traditional DN analysis has much higher fidelity due to high-flux, in-core irradiation <u>and</u> detector design
  - Tough to increase flux by many orders of magnitude, but can easily improve detector

New In-Beam DN Detector Design

Dual Purpose, Optimized Using MCNP Simulations



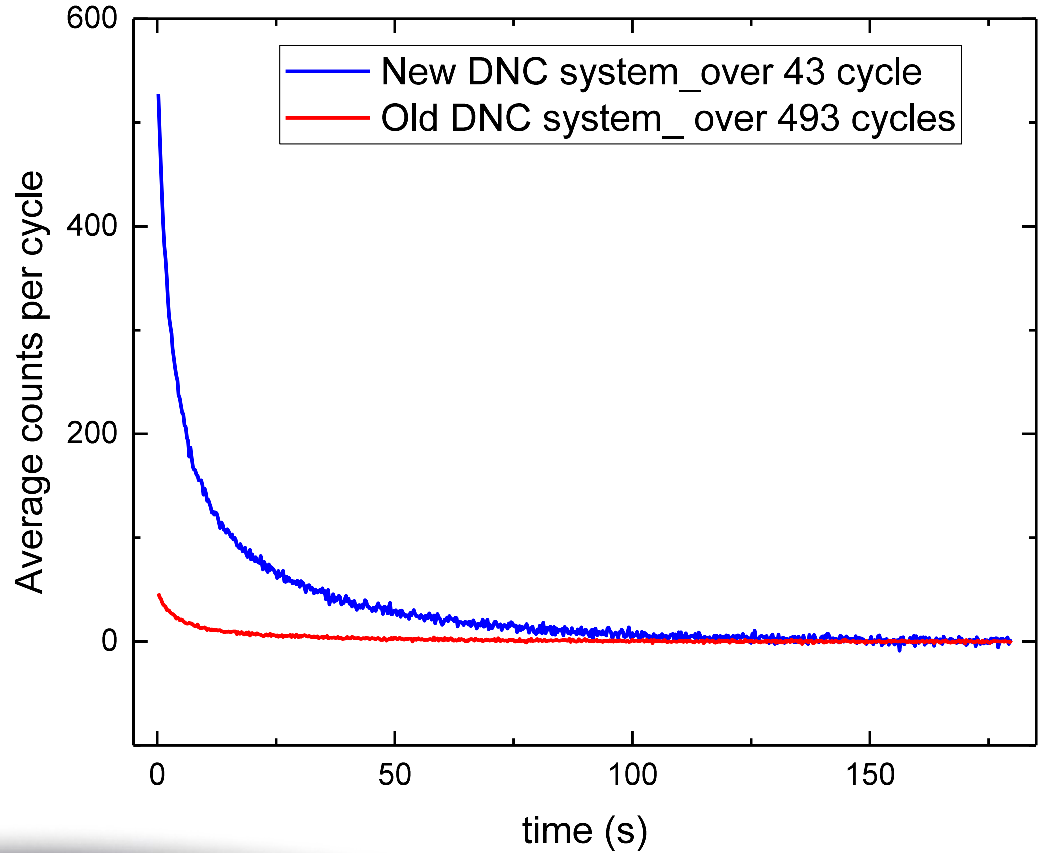
Small sample,
high detection efficiency (33%)



Large or shielded sample,
lower detection efficiency (15%)

γ detector option not shown

Performance Comparison



Summary

- The unique properties of neutrons often make neutron scattering the technique of choice
  - Phase analysis of fuels sometimes better suited to neutrons
  - Neutron PD of irradiated materials
  - High transmission through lead permits NDE while protecting personnel
  - In-beam delayed neutron analysis can be competitive and provides some advantages
- The Chalk River environment, personnel and facilities, has provided opportunity to employ neutron scattering to develop capabilities for studies of reactor and other nuclear materials

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