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Title:	Advanced Sample Environments for in situ Neutron Diffraction Studies of Nuclear Materials
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Intended for:	Lujan Presentation, 2012-07-17 (Los Alamos, New Mexico, United States)

ADVANCED SAMPLE ENVIRONMENTS FOR *IN SITU* NEUTRON DIFFRACTION STUDIES OF NUCLEAR MATERIALS

Dr. Matt Reiche

July 17th, 2012

LA-UR 11-06821 - Unclassified

In a Nutshell

- Challenging Material Conditions in Gen. IV Nuclear Reactors
- Characterize, Understand Materials to Refine Predictive Models
- Access to Atomic Structure during Manufacturing, Operating and Accident Conditions *In Situ*
- Sample Environments (high T, σ) for Neutron Diffraction

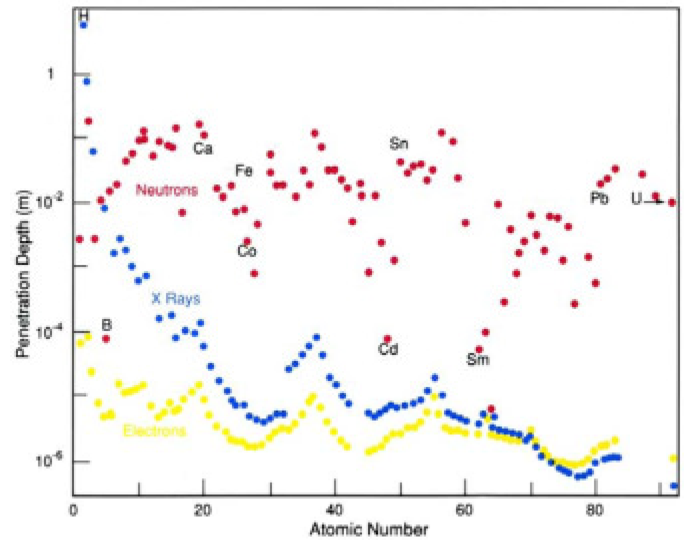
- Zirconium Alloy (Pressure Tubes)
- Uranium Carbide/UO_2 (Nuclear Fuel)

Outline

1. Theoretical Foundation and Approach
2. Experimental Techniques
3. Sample Environments & Application Examples
    A. Sample Changer
    B. Creep Furnace for Zr-2.5Nb Pressure Tubes
    C. High-Temperature Furnace for UO_2 & UC Fuels
4. Summary

Neutron Diffraction

- Deep Penetration Depth
  - ➢ Bulk Material Properties
  - ➢ Sample Environments
- Erratic Variation with Z+N
  - ➢ Distinction of Isotopes
  - ➢ H,C,O,… visible near Pb,U,etc
- Low Flux (ND ~10^7,Synchr. ~10^{22})
  - ➢ Large Samples
  - ➢ Longer Count Times
- Large Scale Facility
  - ➢ Expensive



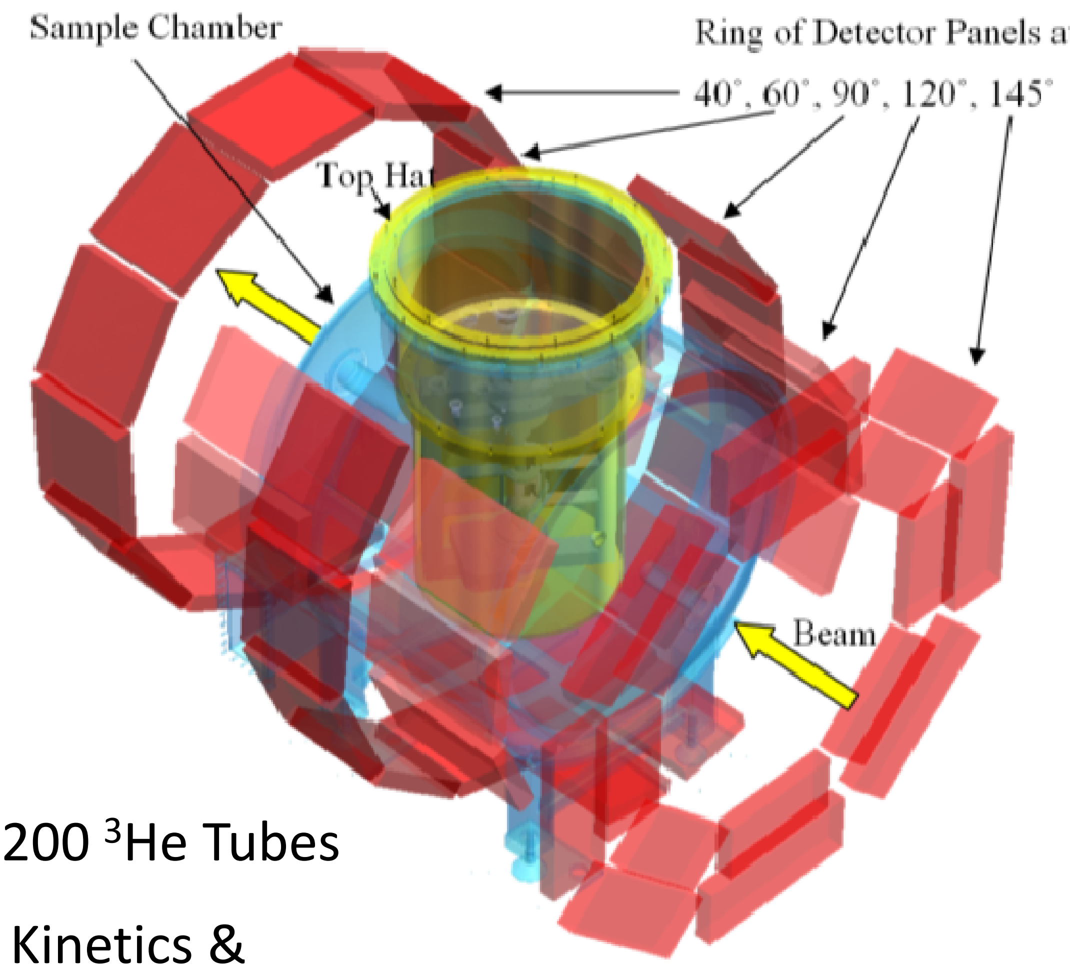
Vogel &

Outline

1. Theoretical Foundation and Approach
2. Experimental Techniques
3. Sample Environments & Application Examples
4. Neutron Diffraction Studies on Zr-2.5Nb
5. Neutron Diffraction Studies on UC
6. Summary

Neutron Diffraction using HIPPO

IS: Sven Vogel



- HIPPO: High Pressure – Preferred Orientation
- 8.83 m from Moderator
- Beam spot ~ 10-14 mm^2
- ~$7 \cdot 10^{14}$ p/s → ~10^7 n/s
- 4.9 m^2 Detector Coverage
- 51 Detector Panels with 1200 ^{3}He Tubes

→ Good Statistics to Study Kinetics &

→ Reliable Texture Measurement with Rotation Around Single Axis Only

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HIPPO Sample Changer: Motivation

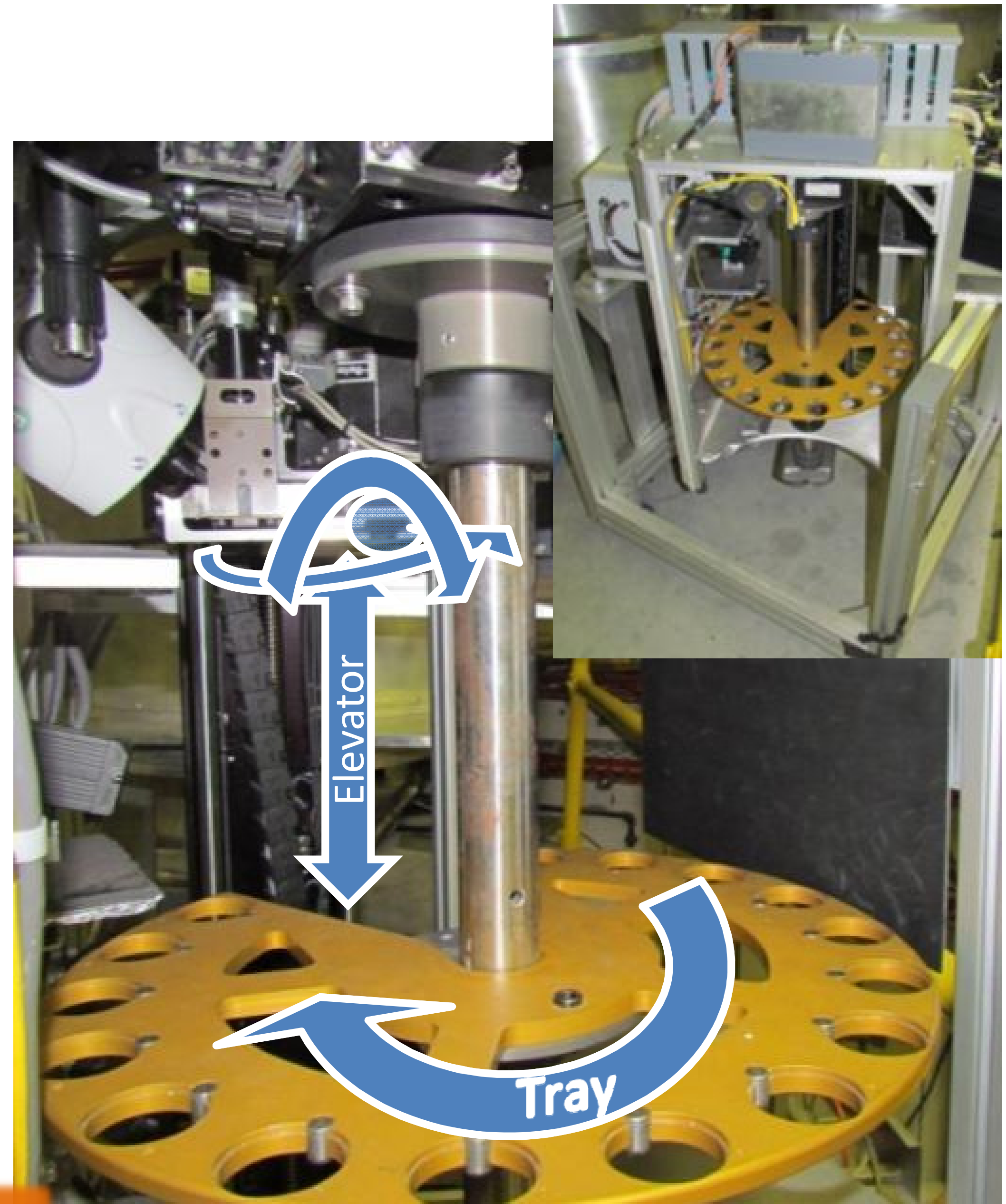
- Short measurements: ~2 min/orientation; 6 min/sample
- Operation: 24/7
- No commercial available sample changer for neutron diffraction experiment. Custom build versions:
  - Shah, 1991, Physica B, →ISIS
  - Rix el al., 2007, Rev. Sci. Instrum, → SNS
  - Hoshikawa et al, 2008, Nucl. Instrum. Meth. A, → J-PARC

- No Sample Changer available for preferred orientation / texture
- Space limitations of Diffractometer

→ **Custom build, Instrument specific solution necessary!**

Sample Changer

- Four motion axes (Parker):
  - 820mm travel elevator
  - Sample tray
  - ±20° Goniometer
  - 245° Gripper
- El. actuated Sample Gripper
- Solid samples: 0.1 – 10 cm^3 or powder samples in V cans



Zygo, Middlefield, CT &
Square One Design, Jackson, WY

Sample Changer

My Improvements

- Sample handling issues:
  - Unreliable sample gripping
  - Crushing of vanadium cans
- Discrepancies between script and actual sample; incl. orientation
- Time consuming repairs: subroutines stored in peripheral devices
- Intermittent operation without consistent notification lead to substantial beam time loss (commercial value ~$10k/day)

- Improved motion control:
  - Optical limit switch for precise reference point and repeatability
  - Video feedback with image analysis
- Image of sample incl. orientation archived
- Motion subroutines copied in startup routine
- Resilient software with self-diagnostic, error mitigation + instant text messaging

➔ Less downtime

➔ Reiche and Vogel, Rev. Sci. Instrum., 2010

➔ Science Highlights Dec,2010

We can still do better…and offer more versatility…

CAD: Mark Taylor

Project Leader:

- Matt Reiche

Software Developer:

- Adrian Losko

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Creep Furnace: Motivation

- Deconvolute impact of thermo-mechanical processes occurring during manufacturing, operation, accident on material behavior.
- Material behavior is based on atomic structure, not accessible.

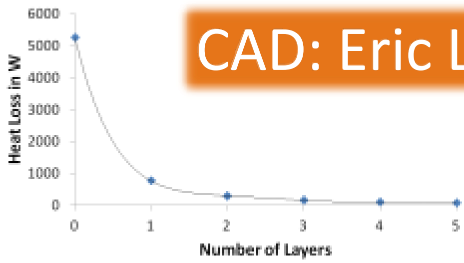
- Simulate conditions during manufacturing etc. in an environment that permits access to atomic structure.

➔ Neutron diffractometer allowing study of texture ➔ HIPPO

➔ High T, stress and sample rotation; not available ➔ Creep Furnace

- Study of microstructural evolution, such as texture, strain evolution
- Phase transition kinetics
- Variant selection
- Active deformation mechanisms during creep

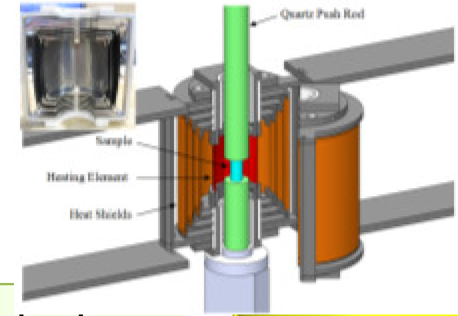
➔ Understand and predict material behavior at T and σ through mechanical models (e.g. visco-plastic self-consistent models)



CAD: Eric Larson

Creep Furnace

Design Criteria



1. Safe operation (esp. radioact. mat.)
2. Temperature control: Highly T-dep.
3. Stress control: no overshoot, wind-up, deformation vs. misalignment
4. Sample & heating element oxidation
   ➔ Vacuum ~~or inert gas~~
5. Conduction: $\dot{Q} = k \cdot A \frac{T - T_{Sur}}{x}$

   Radiation: $\dot{Q} = \varepsilon \cdot \sigma \cdot A(T^4 - T_{Sur}^4)$
6. Low neutron attenuation
7. Compression: avoid buckling and thermal gradient in sample

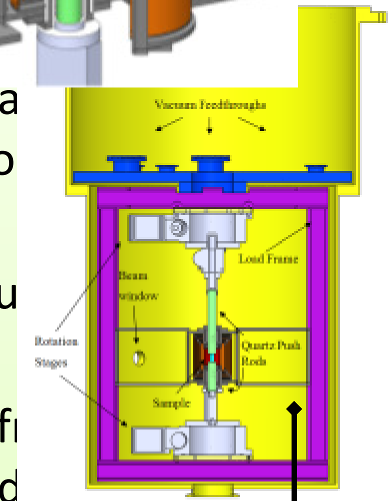
- T_{max} = 1000°C
- θ_{max} = 80° (vertical rotation)
- σ_{max} = 2.7 kN, 100 MPa (rotation stage)

Mark Daymond
Paula Mosbrucker

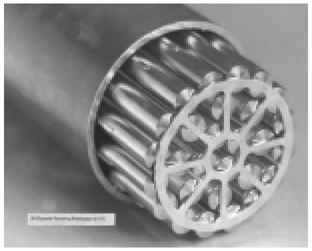
1. HW & SW interlocks: va water flow, 4 thermoco LabVIEW heart beat
2. SW PID with temperatu rate limiter, ramp rate.
3. SW Fuzzy controller, ε fı microsteps, σ from load
4. Heat shields made out of vanadium
5. Heating element made out of V; Al chamber; SS load frame with cooling water in its shadow
6. SiO_2 push rods: comp. strength 1.1 GPa, therm. cond. 2 W/ (m*K)

➔ Reiche et al., Rev. Sci. Instrum, 2012

➔ LANL Science Highlights June,2012



1m

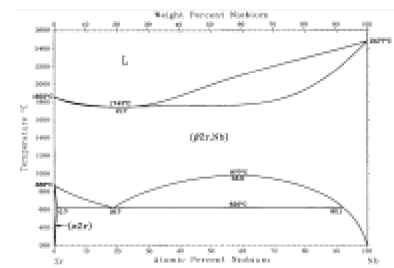


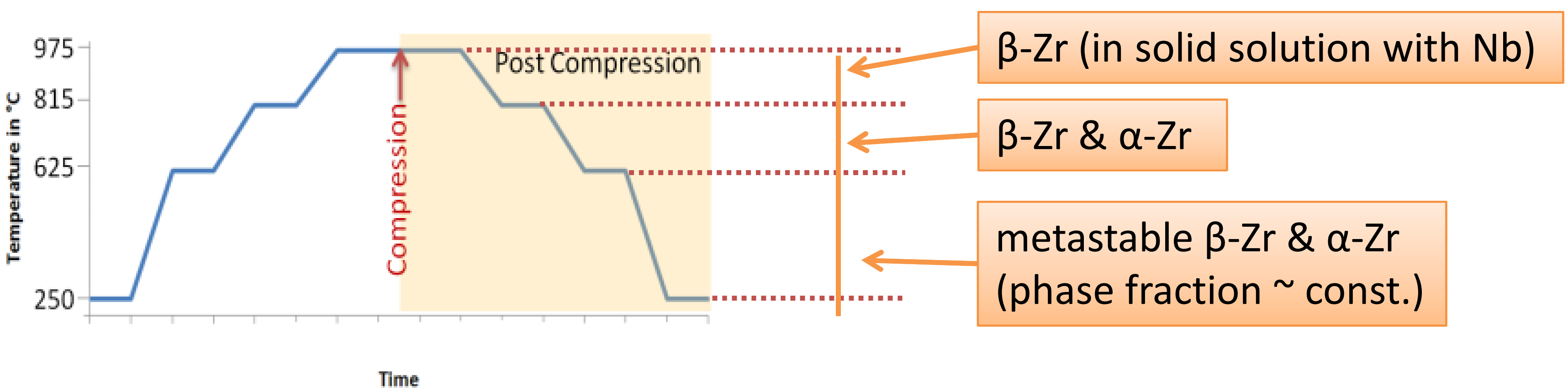
Application: Zr2.5wt%Nb

CANDU Reactor Fuel Assembly:

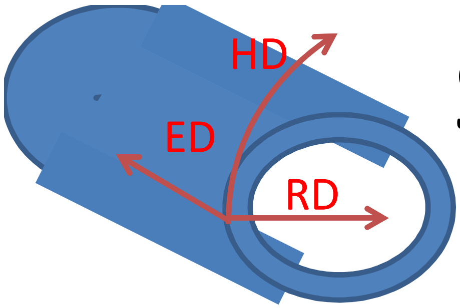
- What is the impact of individual manufacturing steps on the properties of Zr-2.5Nb?
- This material is used for pressure tubes in present CANDU reactors and proposed for use in future supercritical-water-cooled reactors (374˚C, 22.1 MPa).
- Extrusion temperature during manufacturing ~ 815°C
- Phase transition: α (hcp) →β (bcc) > ~815°C

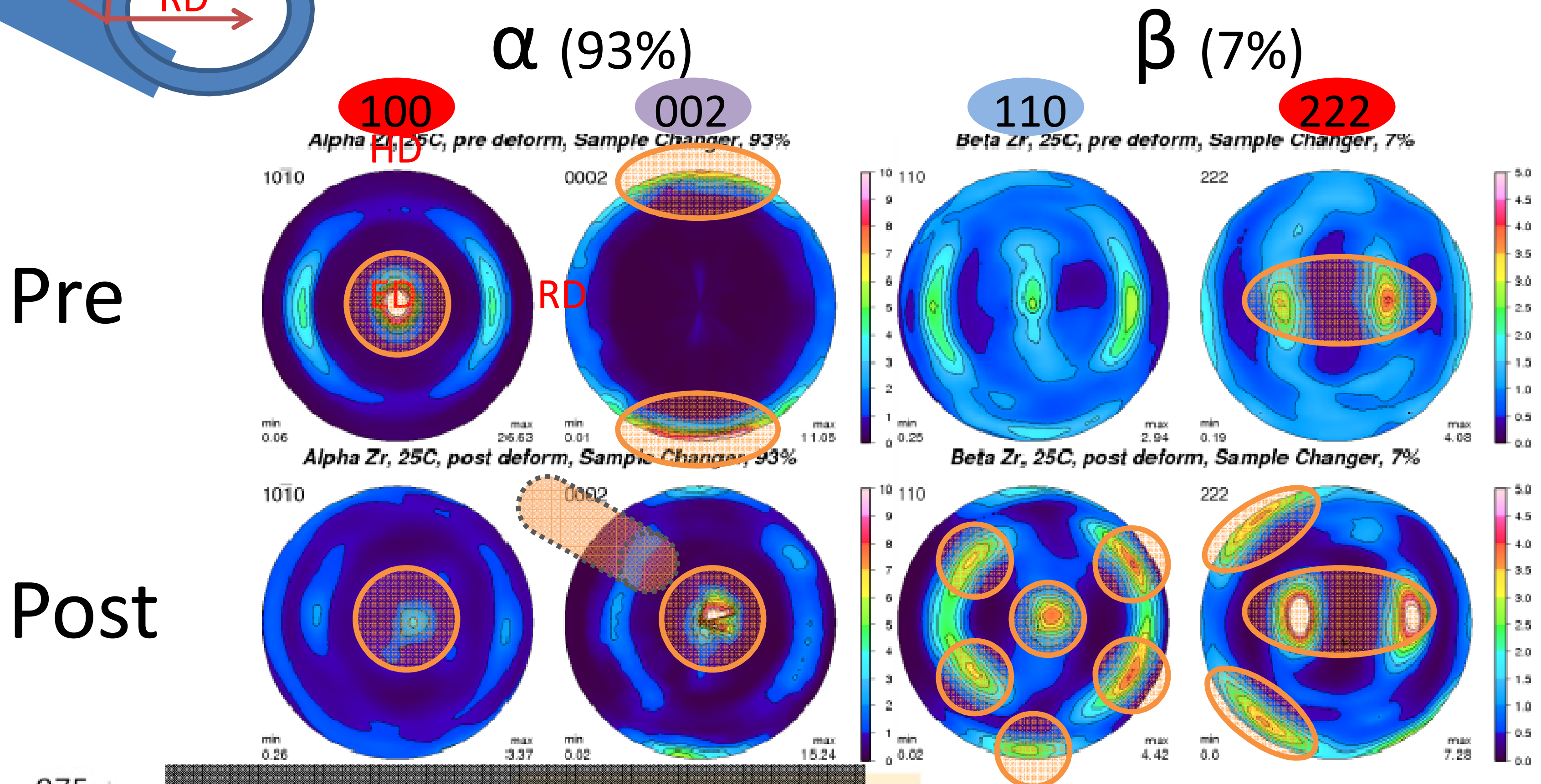
2477°C

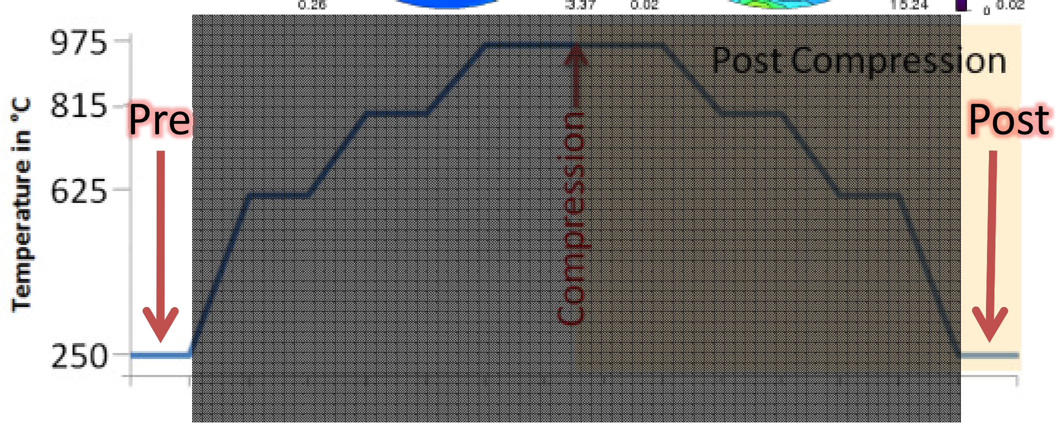




Sample Changer Data







Influence of
Heating, Phase Transition,
Compression, or Cooling?

Creep Furnace Data

- ➔ C…
- ➔ C… during α➔β transition
- ➔ <111> slip in β-phase
- ➔ Variant selection during β➔α transition
- ➔ Quantify texture evol…
- ➔ This apparatus provi… constraints necessary… benchmark mech. m…
- ➔ Collaboration with M… who devise predictive models

Compare Sample Changer with Creep Furnace

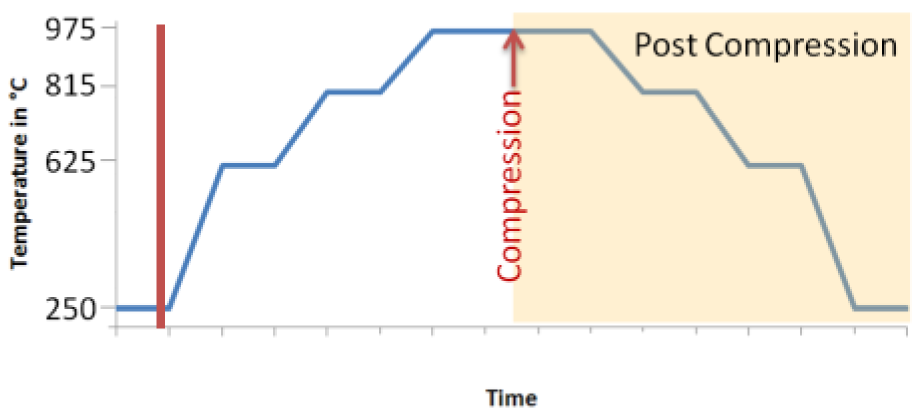
(111) align with CD; <111> slip in bcc

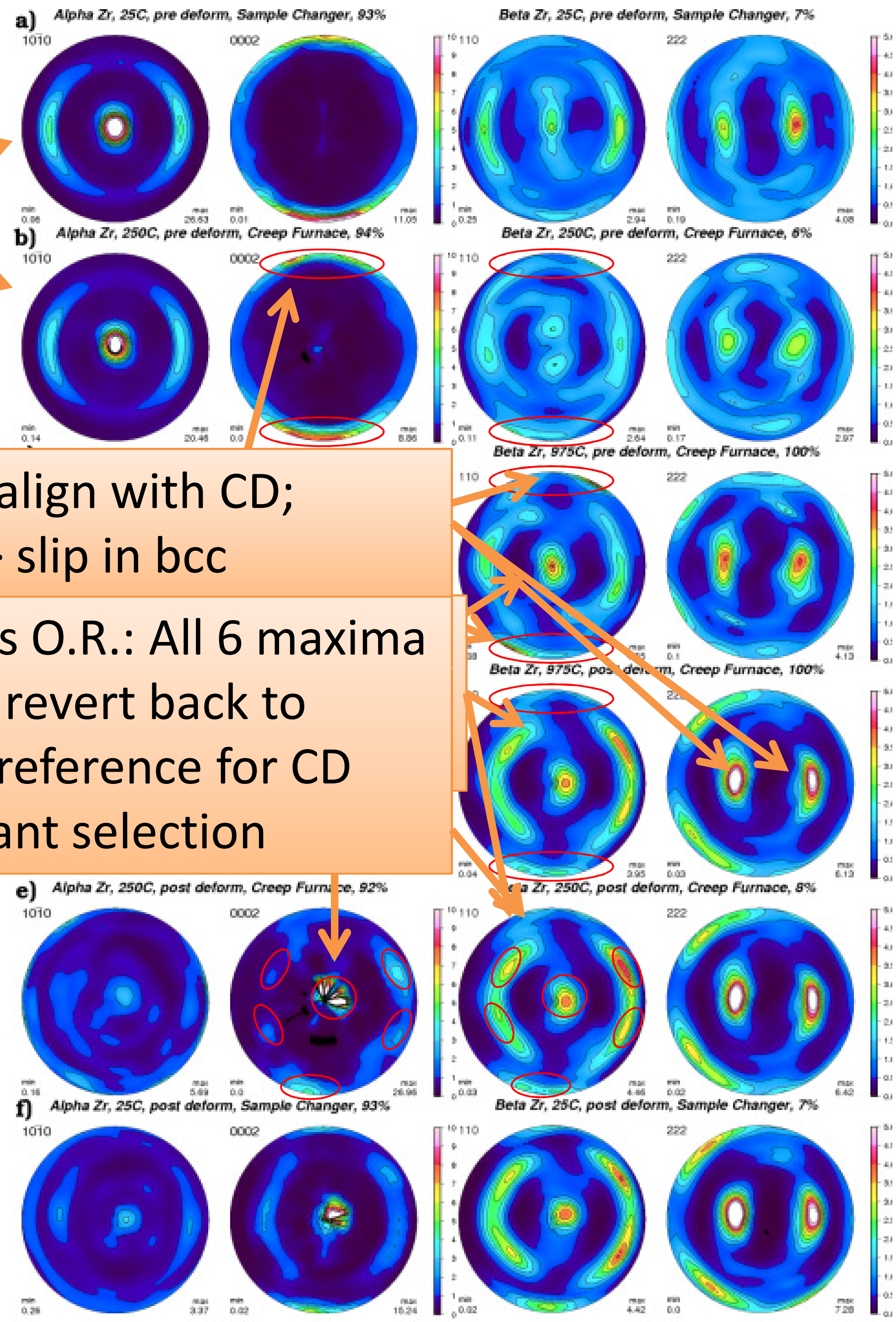
Burger'…

Vari… pha… anis… of α-Z…

β … ➔ … p…

Burger's O.R.: All 6 maxima in 110_{β} revert back to 002_{α}, preference for CD ➔ Variant selection





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High-Temperature Furnace: Motivation

- Nuclear fuels are generally actinides (Th, U, Pu) with a light element (O,C,…) with T_m > 2500°C → ~~XRD, Synchrotron~~
- Phase transition kinetics require fast acquisition time
- Some models predict a UO_2 fuel centerline of T > 1850°C for SCWR
- Many materials have non-quenchable high temperature phases that are poorly known (e.g. cubic UC_2)

- Neutron Diffraction
- HIPPO: Large detector coverage and proximity to source allows for study of kinetics

➔ Build custom hi

Facility	Beamline	High Temperature
ILL, France	D20	ILL: 1150°C (1500°C Nb setup)
LANSCE, NM	HIPPO	ILL: 1000°C
HFIR	HB-2A	ILL: 800°C (1340°C Nb setup)
SNS, TN, USA	---	<under development>
ISIS, UK	GEM	1100°C
J-PARC, Japan	iMATERIA	<under development>

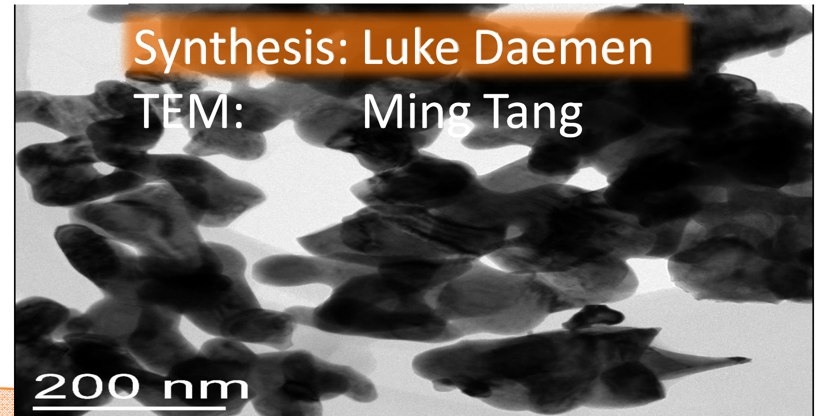
High Temperature Furnace Design

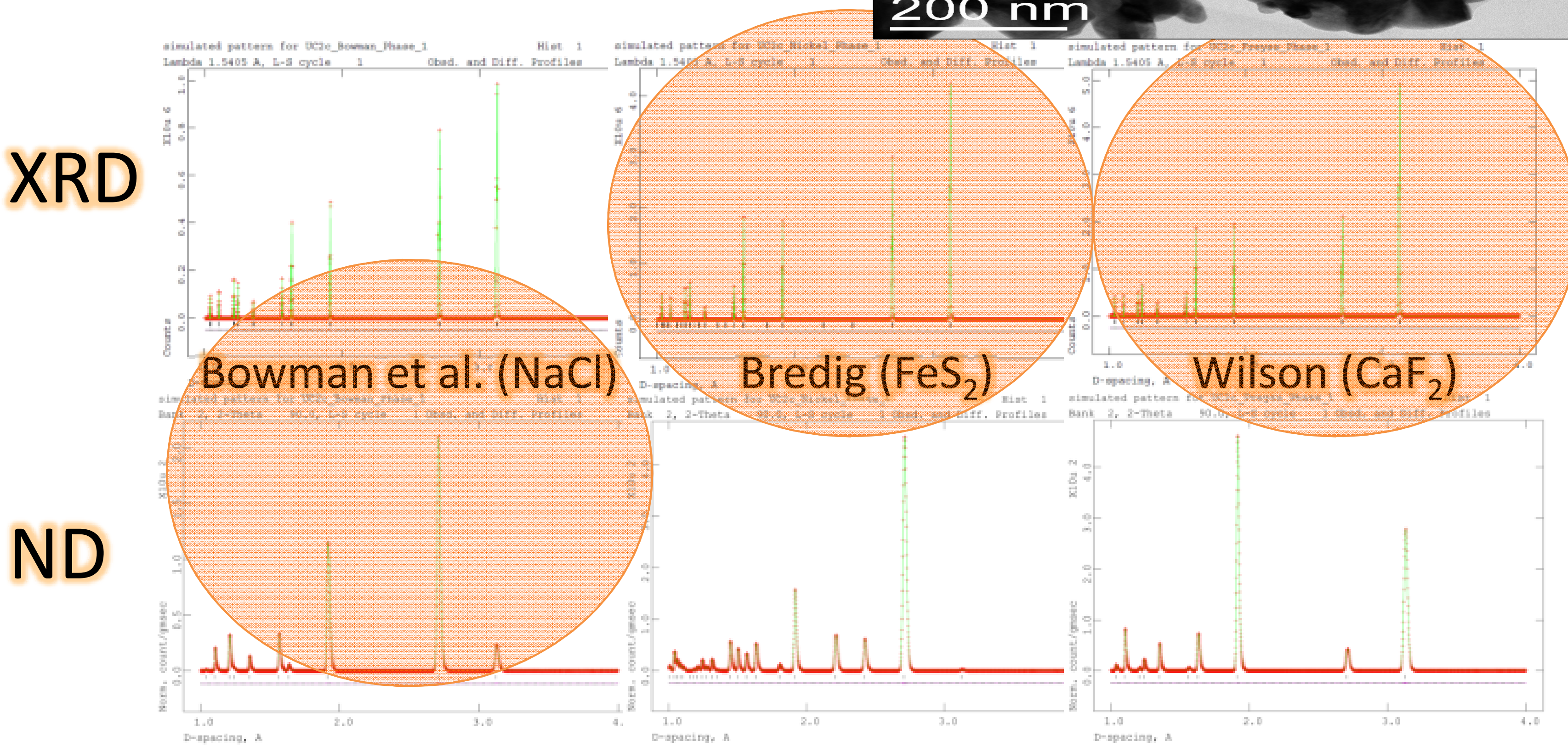
THE UNIVERSITY of TENNESSEE
Joe Wall

- T_{Max} > 2200°C
- Max Rate=100°C/min
- 360° Sample Rotation
- 50 mm Sample Height Adjustment
- Heat shield and element made of graphite
- SCR controlled Transformer: 10V@2000A
- Safety Interlock system (HW & SW)
  - Water flow
  - Vacuum
  - Temperature at Selected Locations
  - LabVIEW Heart beat
- LabVIEW: PID, Error Handling and Notification

Application: UO_2 & UC Nuclear Fuels

A.L. Bowman, G.P. Arnold, W.G. Wittman, **T.C. Wallace**, and N.G. Nereson, "Crystal Structure of UC_2," *Acta. Crystallogr.*, vol. 21, pp. 670-671, **1966**.



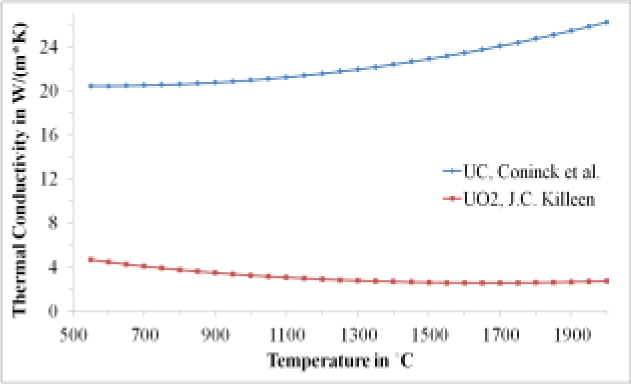


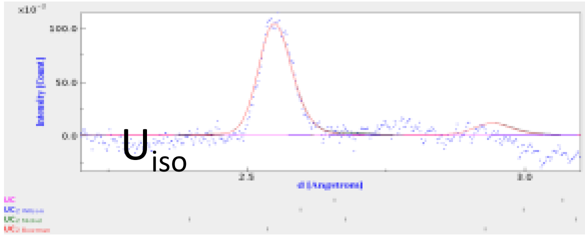
Application: UO_2 & UC Nuclear Fuels

1. UC: higher thermal conductivity and higher fissile density than UO_2
2. ICSD (*Inorganic Crystal Structure Database*) lists 15 entries for UC_2;
   - 12 describe tetragonal phase
   - 3 non-quenchable cubic phase, stable 1769 - 2560˚C; conflicting
3. Uranium powder is pyrophoric

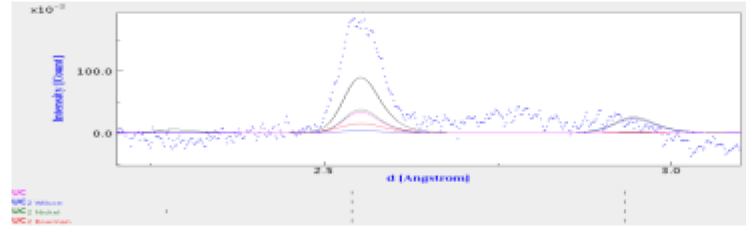
1. Study uranium carbides
2. Study UC_2 in situ at T > 1769˚C using neutron diffraction
3. $UO_2 + 2C \rightarrow UC + CO_2$

A.L. Bowman, G.P. Arnold, W.G. Wittman, **T.C. Wallace**, and N.G. Nereson, "Crystal Structure of UC_2," *Acta. Crystallogr.*, vol. 21, pp. 670-671, **1966**.

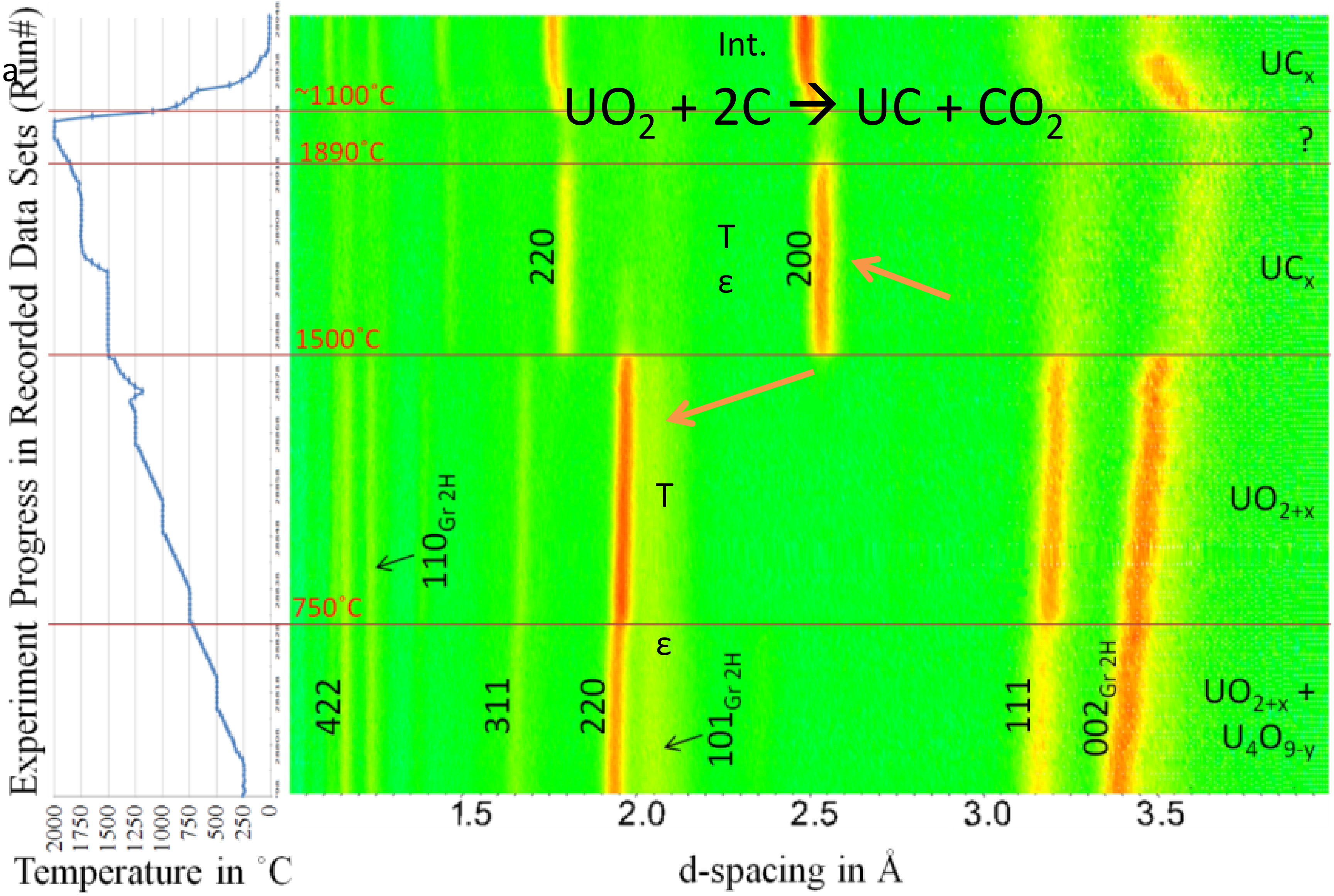




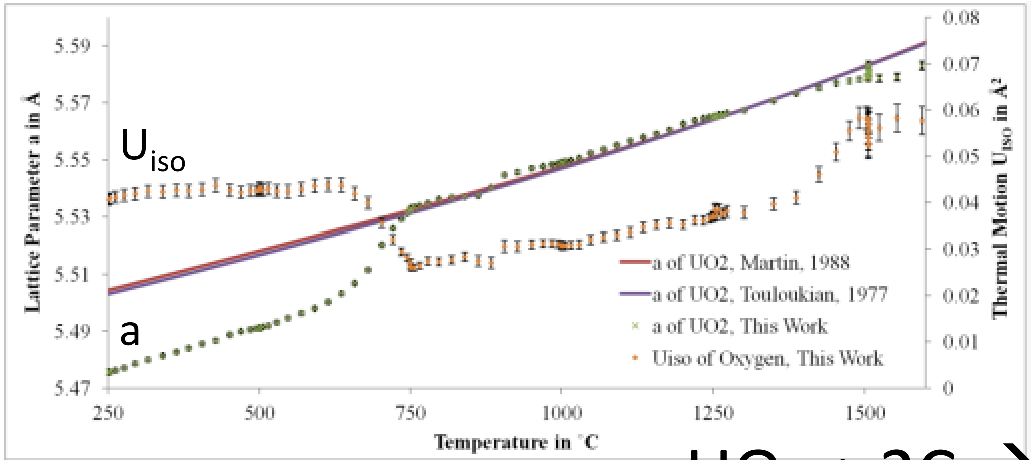
100.0
50.0
0.0
U_{iso}
2.5
d [Angstrom]
3.0

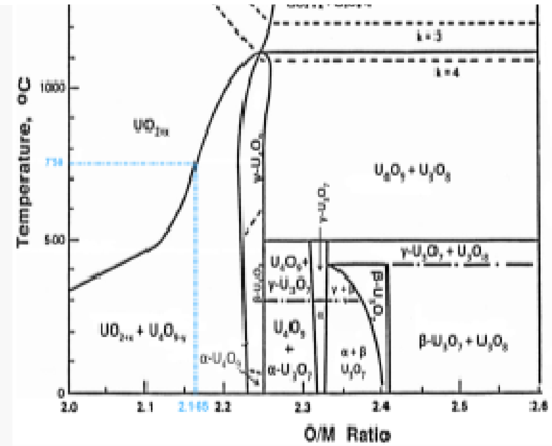


100.0
0.0
2.5
d [Angstrom]
3.0



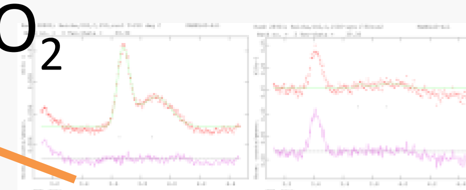
a
Experiment Progress in Recorded Data Sets (Run#)
Int.
~1100˚C
$UO_2 + 2C \rightarrow UC + CO_2$
1890˚C
1500˚C
750˚C
UC_x
?
UC_x
UO_{2+x}
UO_{2+x} + U_4O_{9-y}
220
T
ε
200
T
ε
$110_{Gr\ 2H}$
422
311
220
$101_{Gr\ 2H}$
111
$002_{Gr\ 2H}$
2000
1750
1500
1250
1000
750
500
250
0
Temperature in ˚C
1.5
2.0
2.5
3.0
3.5
d-spacing in Å



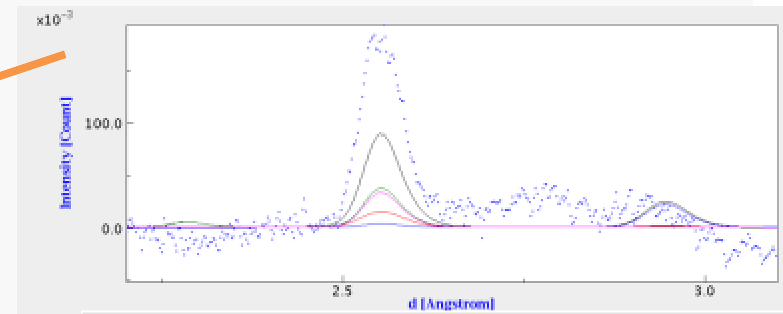


$$UO_2 + 2C \rightarrow UC + CO_2$$

Not predicted by theoretical work from Basak, 2007!



1. Sample at correct temperature
2. Thermometry correct



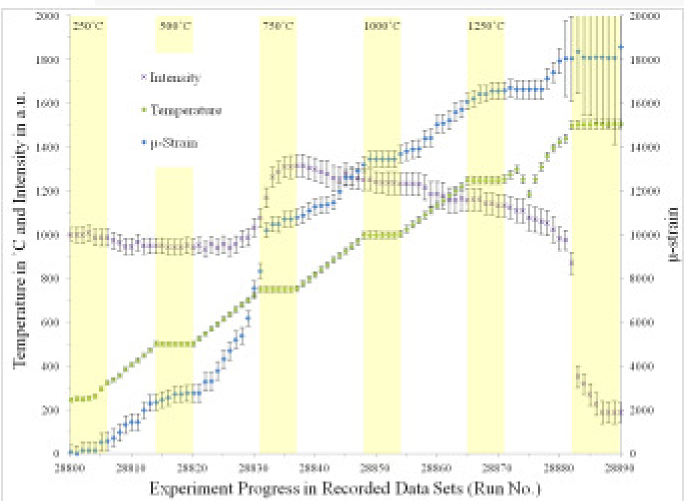
T

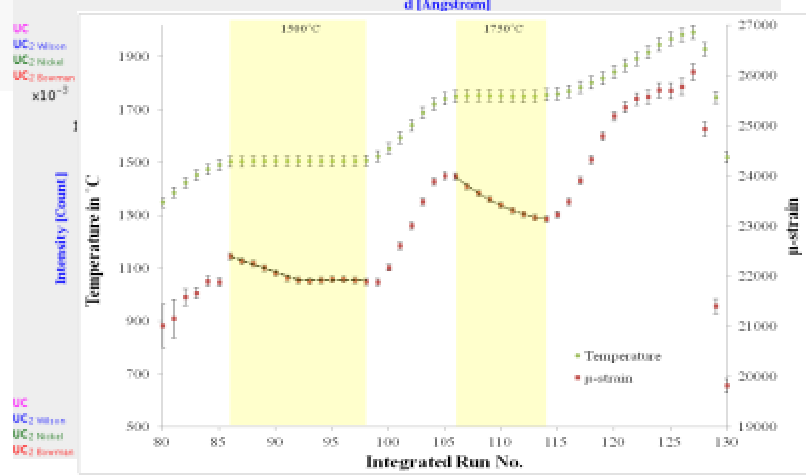
ε

Int.

T

ε





Order-Disorder Transition

Why loss of intensity?

1. Melt?
2. Bredig Transition (-> Superionic)?
3. Motion of C_2 Molecule

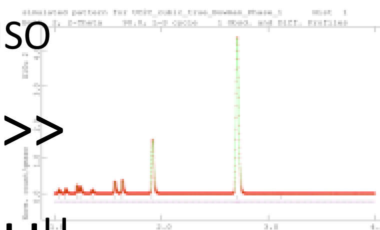
A. ‘Free Rotation Model’ by Bowman et al., 1966, ND.

B. ‘Oscillating’ Model by Wen et al. , submitted, simulation.

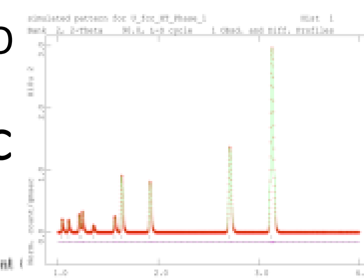
➔ Collaboration with T-1, LANL; We will provide experimental data to validate their HSE model

➔ Molecular Oscillation Cannot Be Readily Modeled with Rietveld software ➔ Maximum Entropy Method

$UC^{C}_{ISO} >> U^{U}_{IS}$



fc o c



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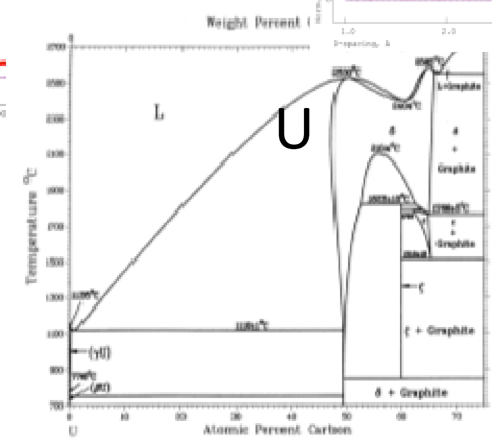
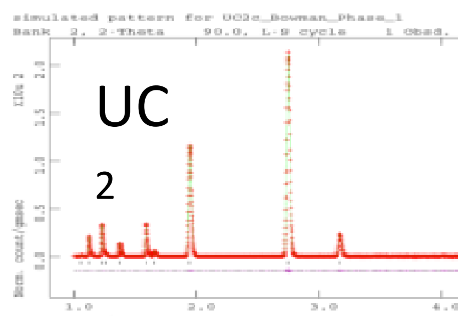
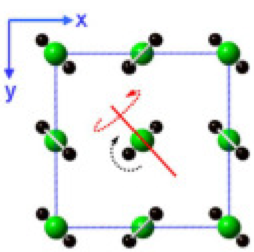
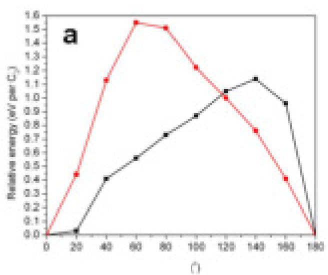
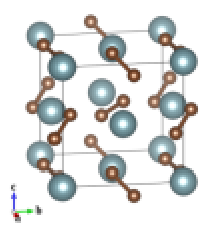


UC 2

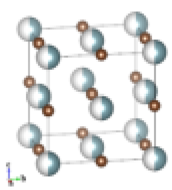




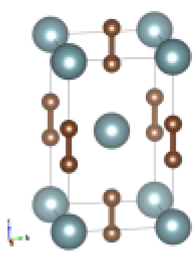




FeS$_2$



NaCl



UC_2 tet.

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Summary

- I successfully developed and commissioned three sample environments; as well as applied them to study scientific problems of nuclear materials.
- Sample Changer: Reliable, unattended operation; Reiche & Vogel, 2010.
- Creep Furnace: 1000˚C, 2.7kN, texture; Reiche et al., 2012.
- Zr-2.5Nb Pressure Tubes: Deconvolute the contributions of heating, phase transformation, deformation at temperature in the β-field, and phase transformation during cooling.
- Our data will inform currently developed mechanical models predicting texture evolution during phase transformation and high temperature deformation (MST-8, LANL).
- High-Temperature Furnace: >2200˚C, texture; paper in progress.
- UC: Bowman's NaCl-type structure correct for cubic UC_2. Recent reactor safety calculations by Chevalier & Fischer (2001) and independent work by Freyss (2010) incorrect, as fluorite structure is assumed.
- UC: Order-disorder transition starting at 1800˚C discovered which will improve current models, as these do not predict this transition (e.g. Basak, 2007), or confirm the prediction (Wen et al., 2012, T-1, LANL).

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Thank YOU!

Special Thanks to:

- Our Creator

- Erica & Shoshanah
- Dr. Sven Vogel
- Prof Dr. Steve Stochaj
- Prof. Dr. Heinz Nakotte

Publications

1. H.M. Reiche, S.C. Vogel, Mosbrucker P., Larson E.J., and M.R. Daymond, "A furnace with rotating load frame for in situ high temperature deformation and creep experiments in a neutron diffraction beam line," Rev. Sci. Instrum., vol. 83, p. 053901, 2012.
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