UNIVERSITY OF ILLINOIS
AT URBANA-CHAMPAIGN

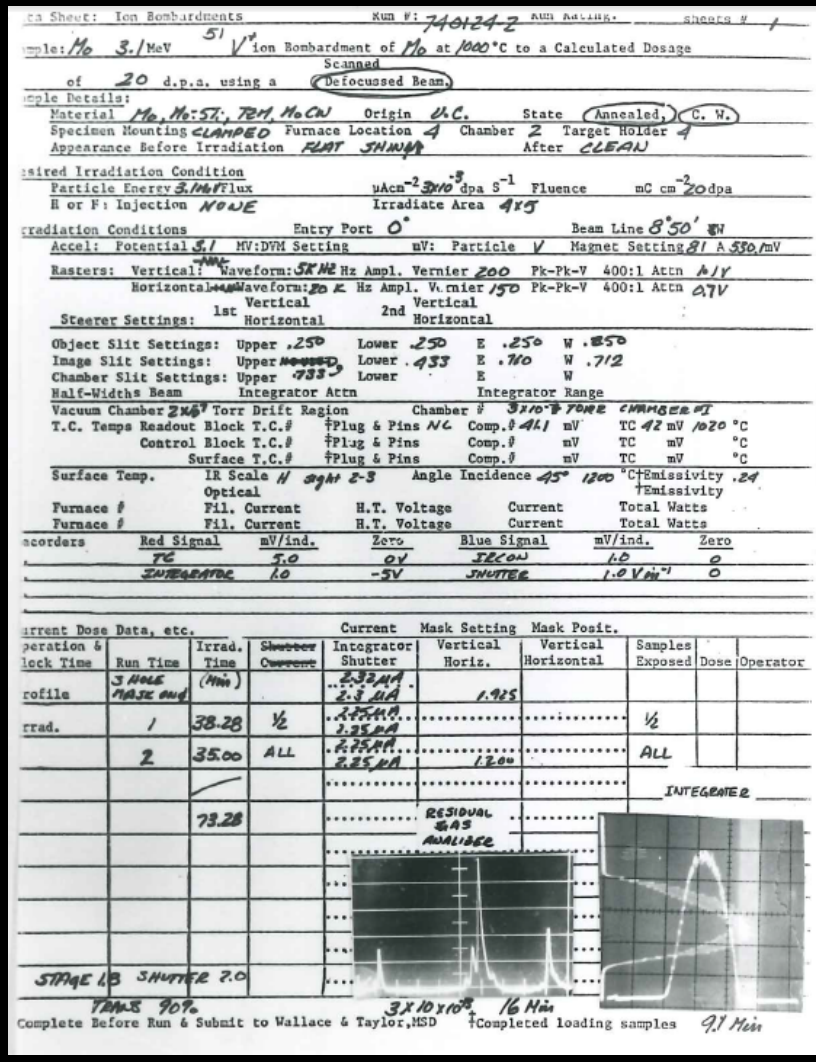
A University Perspective on the Value of NSUF: Nuclear Fuels, Structural Materials and Advanced Analysis

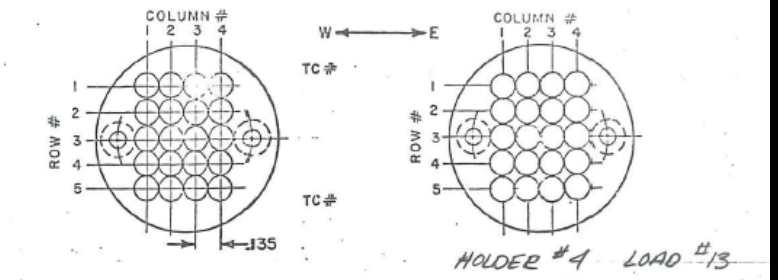
Students, Collaborators, Facilities and Technical Personnel

James F. Stubbins

Department of Nuclear, Plasma, and Radiological Engineering, University of Illinois at Urbana-Champaign

Thanks





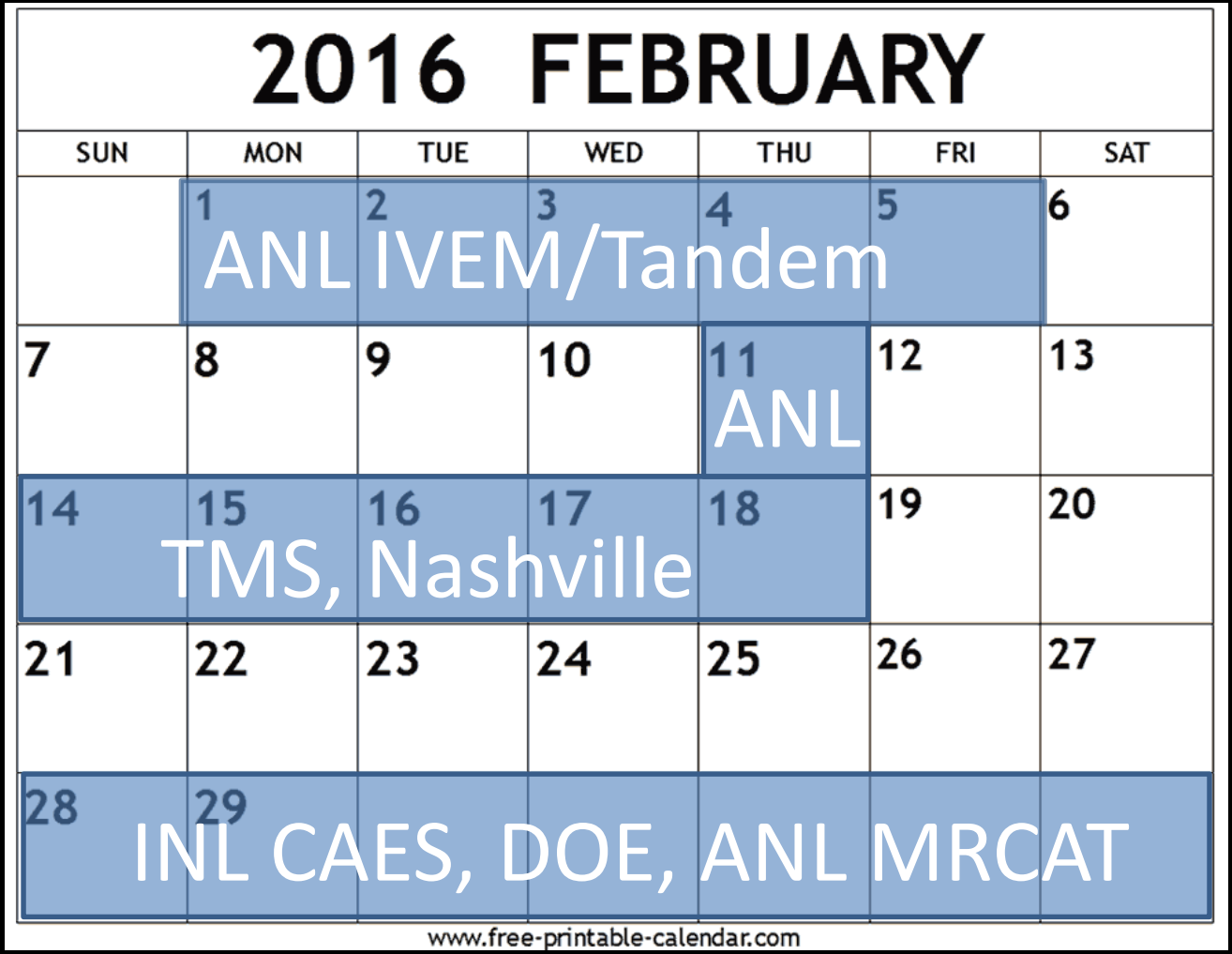
Location Col.	Location Row	Composit.	Foil Section Depth Å	Dose/Dose Rate Tot. Time Secs.	Dose/Dose Rate μA/cm²	Computations mc/cm²	Computations dpa/sec (×10⁻³) dpa	Comment
1	1	Mo	5750	4397	.521	2.29	2.7 / 11.8	
1	2	Mo	5500	4397	.607	2.67	3.2 / 13.4	
1	3	Mo	6500	4397	.691	3.04	3.4 / 15.1	
1	4	TZM	4750	4397	.748	3.29	3.5 / 14.5	
1	5	TZM	3000	4397	.628	2.76	1.8 / 8.0	
2	1	Mo:5Ti	6000	4397	.823	3.62	4.3 / 18.7	
2	2	Mo:5Ti	4250	4397	1.094	4.81	4.0 / 17.4	
2	3	Mo:5Ti	4500	4397	1.387	6.10	5.1 / 22.6	
2	4	CW		4397	1.474	6.48		
2	5	CW		4397	1.105	4.86		
3	1	TZM	5250	2297	.836	1.96	4.1 / 9.3	
3	2	Mo	1750	2297	1.180	2.71	2.6 / 5.9	
3	3	Mo	5500	2297	1.480	3.40	7.3 / 16.7	
3	4	TZM	6750	2297	1.567	3.60	7.6 / 17.4	
3	5	TZM	5500	2297	1.180	2.71	8.5 / 19.5	
4	1	CW		2297	.644	1.48		
4	2	Mo:5Ti	5500	2297	.718	1.65	3.5 / 8.1	
4	3	Mo:5Ti	3500	2297	1.158	2.66	3.4 / 7.9	
4	4	CW		2297	1.232	2.83		
4	5	CW		2297	.836	1.92		

RUN 740124-2

Perspective

User Facility	Location	Project	Agency
Dynamatron	ANL	Mo, TZM, Mo-.5Ti	DOE-BES Irradiation Effects
Accelerator	Darmstadt, Germany	Vanadium Alloys	GDR
HVEM	Harwell Labs, UK	Fe-Cr-Ni Alloys	UK
Microscopy Suite	Oxford, UK	Fe-Cr-Ni Alloys	UK
Center for Microstructure Matl	MRL Illlinois		DOE-BES, NSF
HVEM	ANL	Fe-Cr-Ni Alloys	DOE-BES Irradiation Effects
IVEM-Tandem	ANL		DOE-BES Irradiation Effects
High Temperature Materials Lab	ORNL		DOE
Advanced Photon Source	ANL	Mulitple	DOE
Ion Beam Lab	U Michigan	Fe Alloys, Zr Coating	Multiple DOE-NE NEUP
Positron Facility	Washingtion State U	Fe Alloys	DOE OS
Microscopy Center	ANL		DOE-BES Irradiation Effects
Advanced Test Reactor	INL	Neutron Irr Fe-Cr Alloys	DOE-NE
Atom Probe, TEM, SEM, etc.	ORNL-SHaRE	ATR, A709 & G92, A617	DOE-NE NEUP
Atom Probe, Hot Microscopy, Micr	CAES	F/M - ATR Materials	DOE-NE NEUP
Nano Hardness/Pillar	UC-Berkeley	A709 & G92	DOE-NE NEUP

Perspective

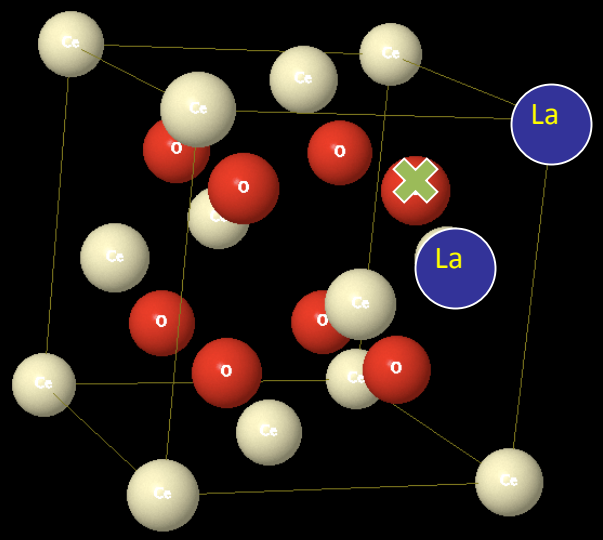


Overview

- Research Highlights
  - 1. Nuclear Fuels
  - 2. ATR Results
  - 3. APS Results
- ATR Experiments
  - Experiment Formulation
  - History
  - Issues
- Future
  - Dynamic Experiments vs Modeling
  - 3D,
- Summary – Important Considerations

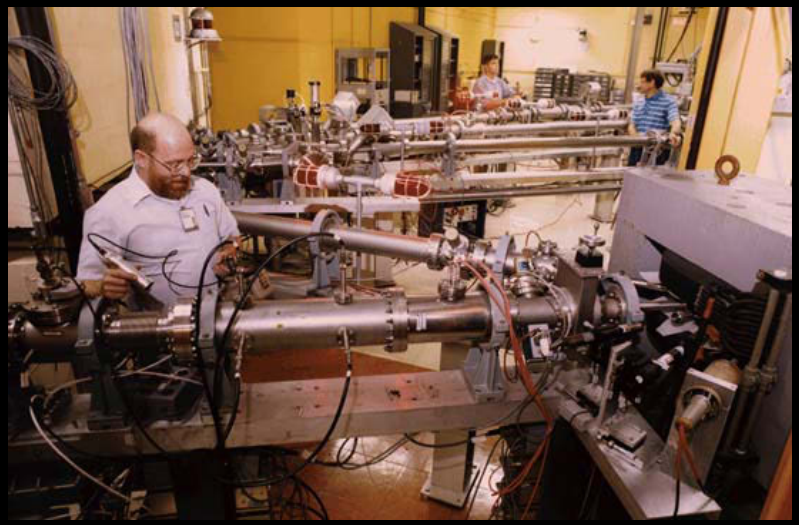
1. Fuels: CeO_2 +La: a UO_2 Surrogate

- CeO_2 is used for UO_2 surrogate due to their similar crystal structure (fluorite structure) and physical property
- Lanthanum (La) is added to CeO_2 to simulate $(Pu,U)O_2$ and impurity effect
- La(3+) substitutes Ce(4+) position, creating 0.5 oxygen vacancy to maintain neutrality



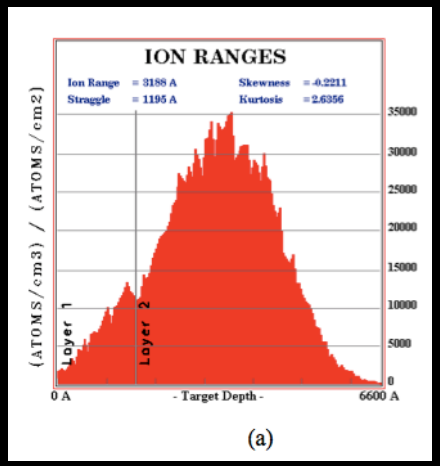
IVEM-TANDEM Facility in ANL

- Operating voltage: 40 – 500 keV (single charge)
- Ion source: noble gases Xe, Kr, H, Fe, Ni, Cu, Au
- Typical flux: 1×10^{12} cm^{-2} s^{-1}



Irradiation Condition

- Source: 1 MeV Kr
- Temperature: 800 °C
- Final dose: 8×10^{14} cm^{-2}
- In-Situ

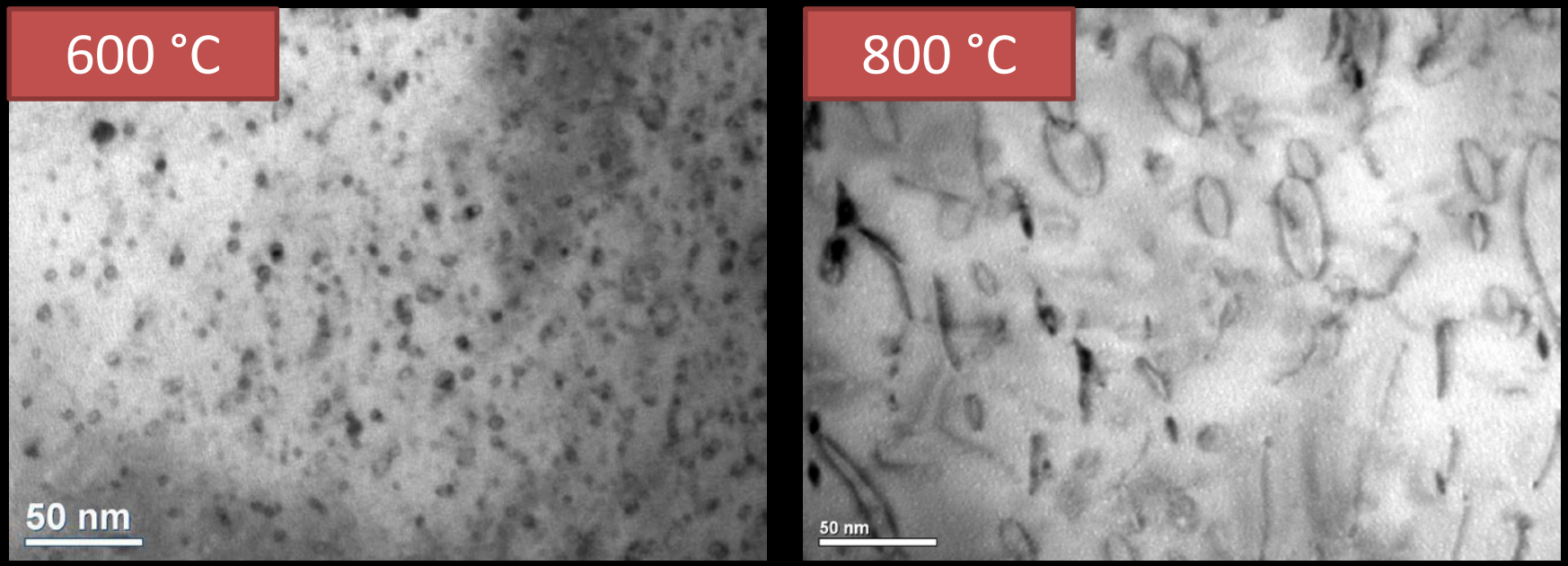


(a)

- Source: 700 keV Xe
- Temperature: 800 °C
- Final dose: 1×10^{16} cm^{-2}
- Ex-Situ

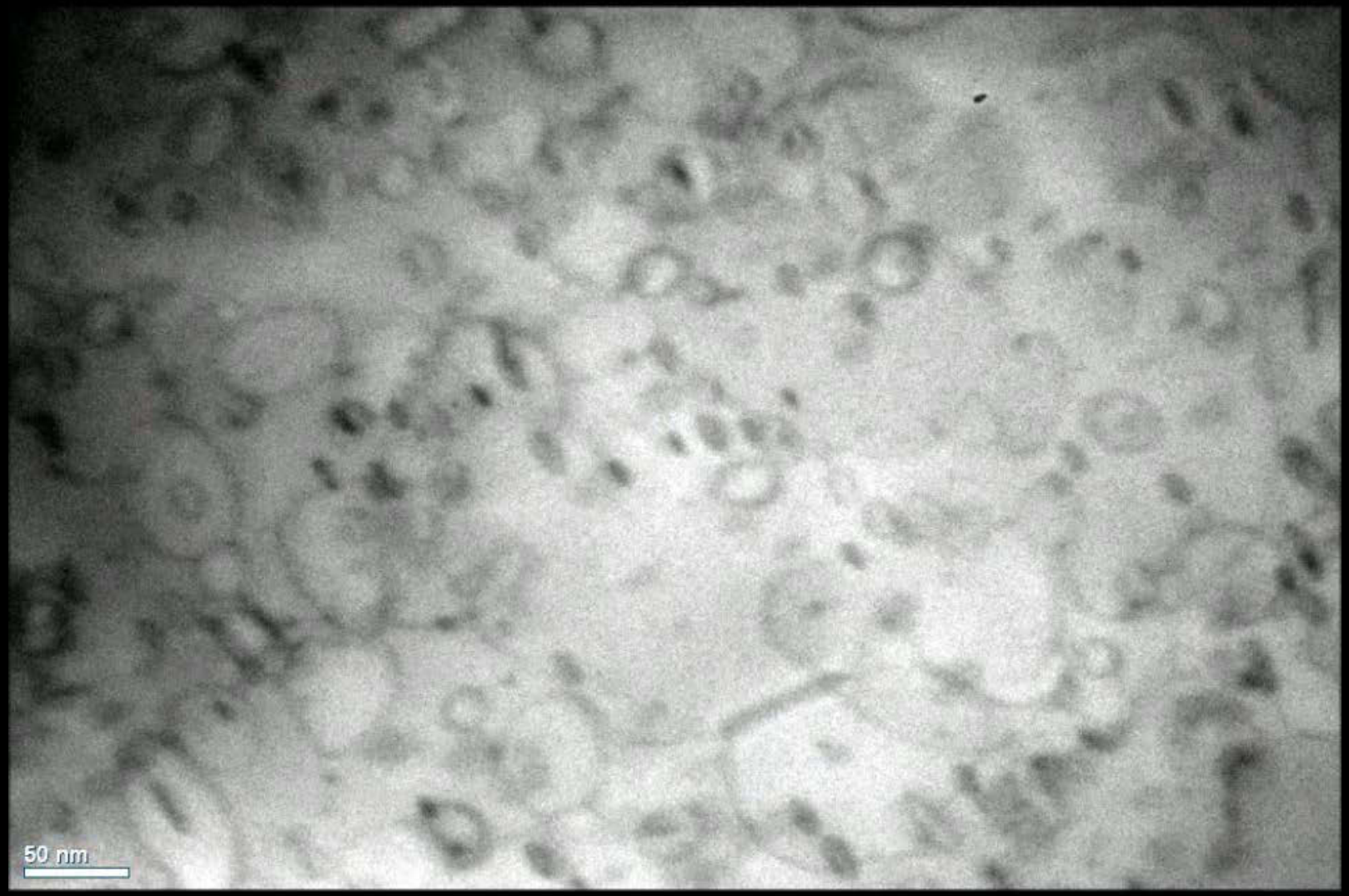
Temperature Effects

- Dramatic change in kinetics for dislocation loops was found between 600 °C and 800 °C
- Temperature of 800 °C was thus selected in this study



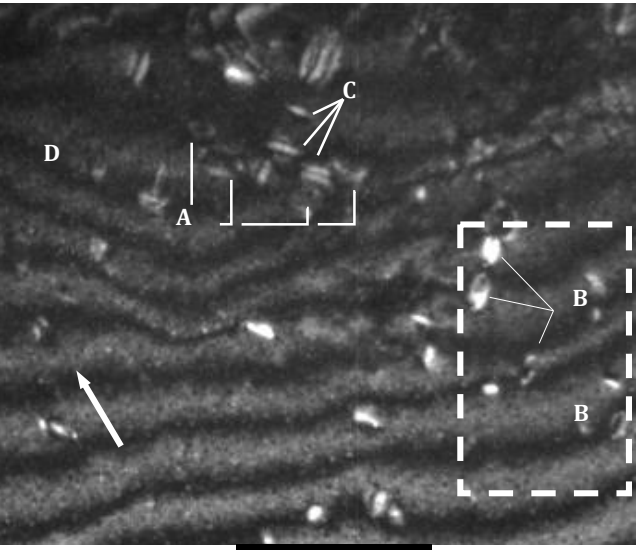
Dislocation loop structure in CeO2 irradiated by 1 MeV Kr^{+}.
Dose = 1×10^{15} cm^{-2} (Ye at el. 2011)

25% La Doped CeO_2
1 MeV Kr Irradiation at 800 C
Dose ~ 1.2 ions/cm^2

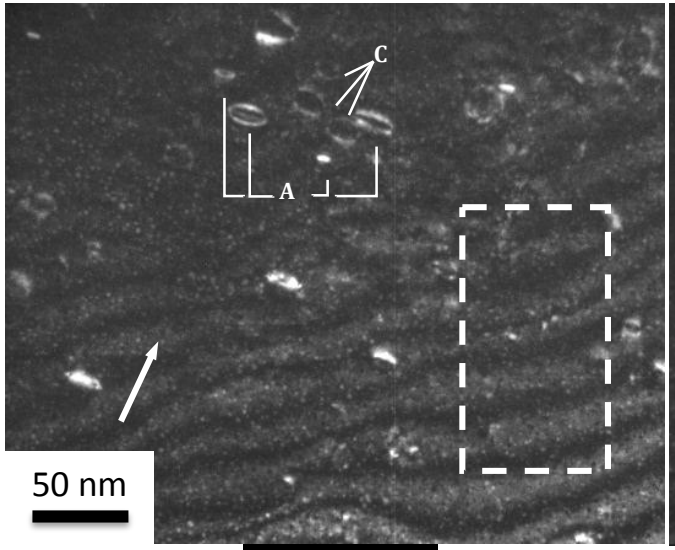


Burgers' Vector

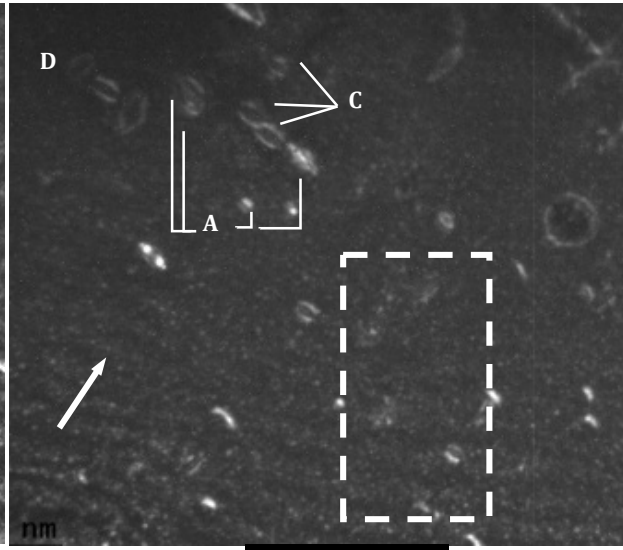
- Reflections g = (020), (220) and (42-2) were used for b • g analysis
- Results => b // <111> (perpendicular to habit planes)



g = (020)



g = (220)



g = (42-2)

Atomistic Structure of Dislocation Loops: MD

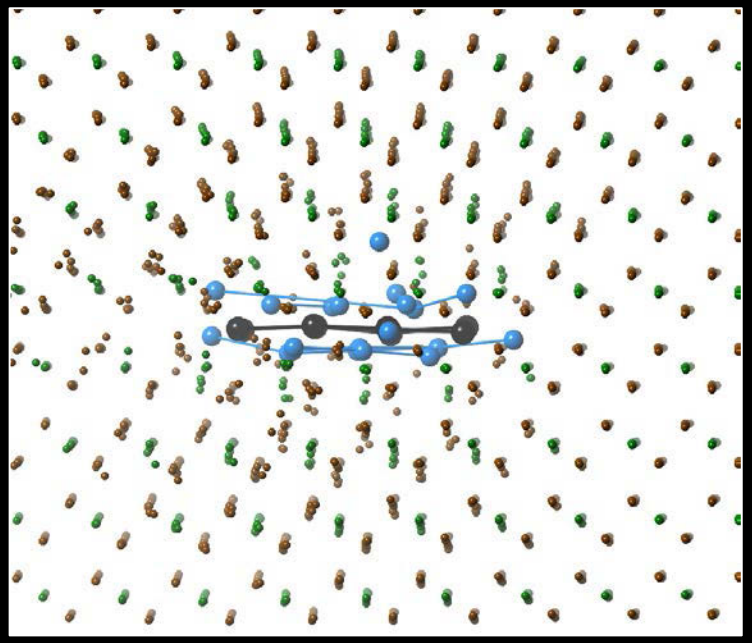
- "O - Ce - O" sandwich structure is suggested instead of a single layer of oxygen ions
  - both cerium and oxygen sublattices were displaced
  - Stoichiometric structure are more energetically favorable

O interstitial

Ce interstitial

<111>

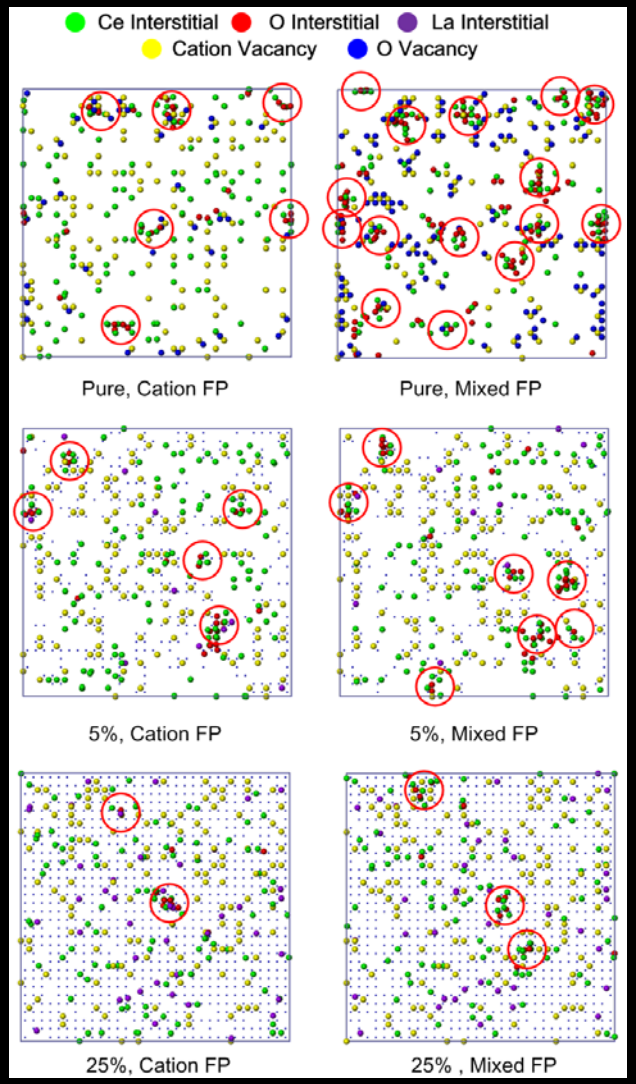
<110>

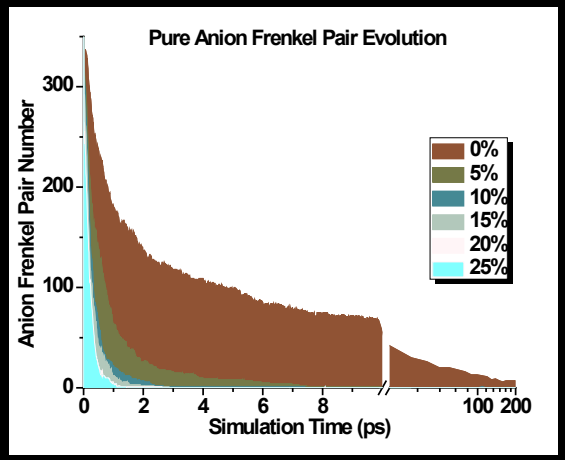


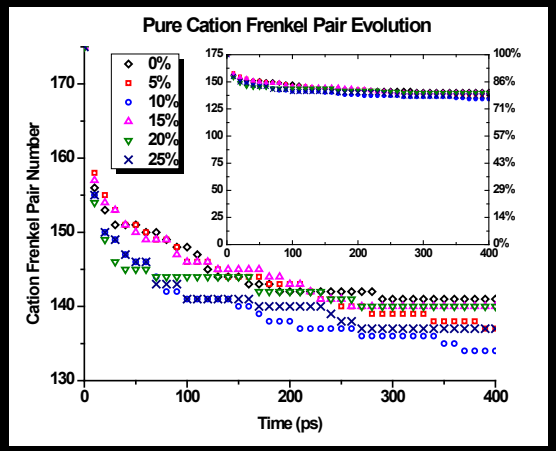
D.S. Aidhy et al. *Scripta. Mater.*(2011)

La Effects on Point Defects: MD & kMC

- Properties of Point Defects
  - Pure oxygen point defects recombine quickly
  - Cation point defects have long lifetime and dominate the loop evolution
  - Oxygen point defects enhance loop evolution
  - La-induced-O-vacancies shorten the lifetime of oxygen Frenkel pairs
  - Fast loop evolution ⇔ Fast cation defect recombination (cation interstitial mobility)
  - La lowers cation interstitial mobility







2. ATR: Fe-Cr Alloys

PIE summary

Alloys (Irradiation in ATR)	Target Temperature (°C)	Target Dose (dpa)
Fe Poly Crystals	**300, 450** (550)	**0.01, 0.1, 1** (5, 10)
Fe - 10Cr Poly Crystals		
Fe - 14Cr Single Crystals		

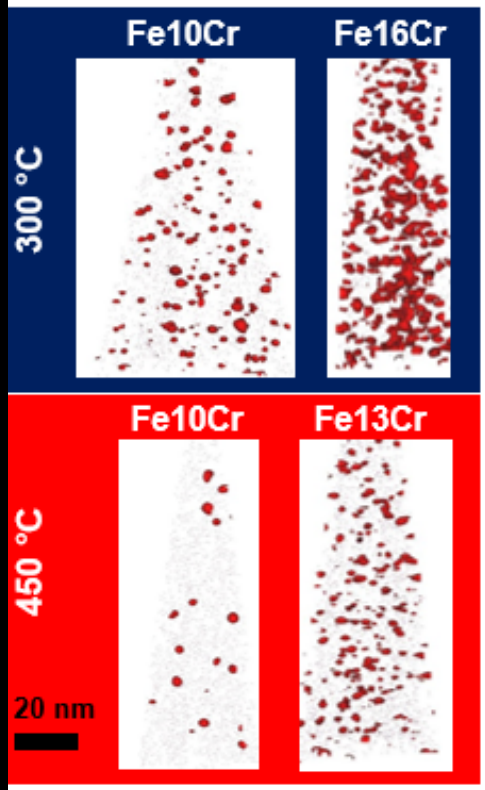
Microstructure	
TEM	• Dislocation structure • Voids • Composition
APT	• α′ precipitation • Composition
SEM/EBSD	• Grain size

Mechanical Property	
Hardness	• Micro-hardness • Nano-hardness

The evolution of **microstructure** and **mechanical properties** as a function of **Cr concentration** and **irradiation temperatures**

ATR: CAES Atom Probe – α’

Atom Probe Tomography

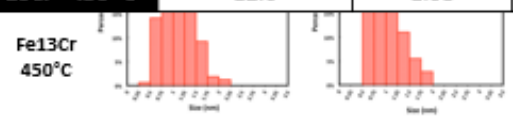


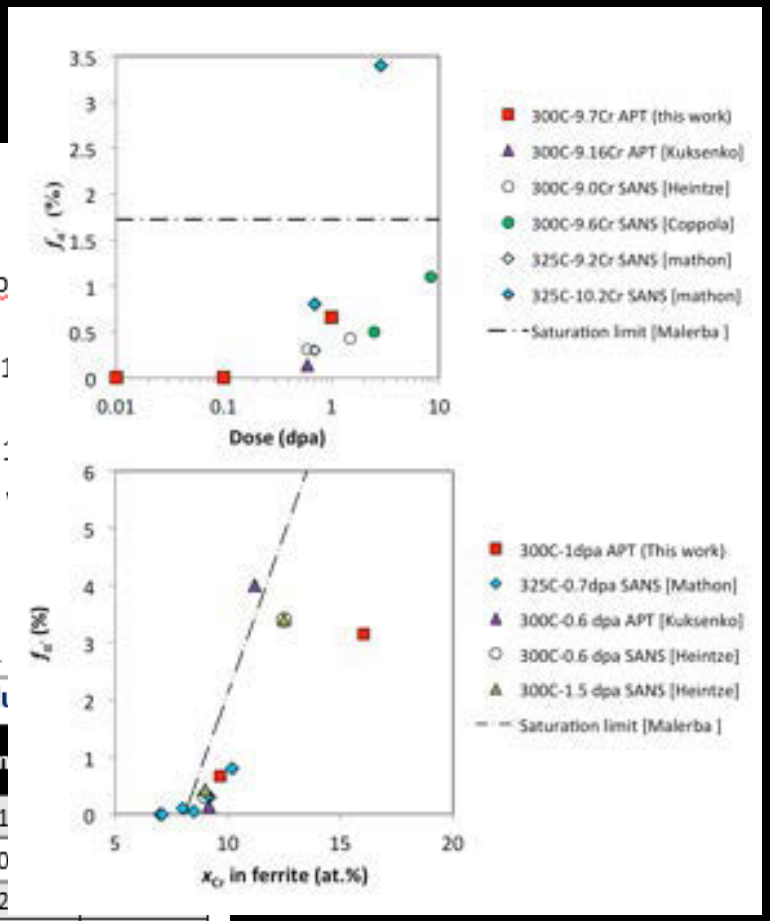
- α′ precipitates defined by 20%Cr iso surfaces
- Evident α′ precipitates observed in 1 and not in lower doses
- Density varies significantly ~ 10^{23} to 1
- The volume fraction $f_{\alpha'}$ is consistent of phase diagram prediction
- Radius ~ 1 nm

Size distribution of α′ precipitates in 1

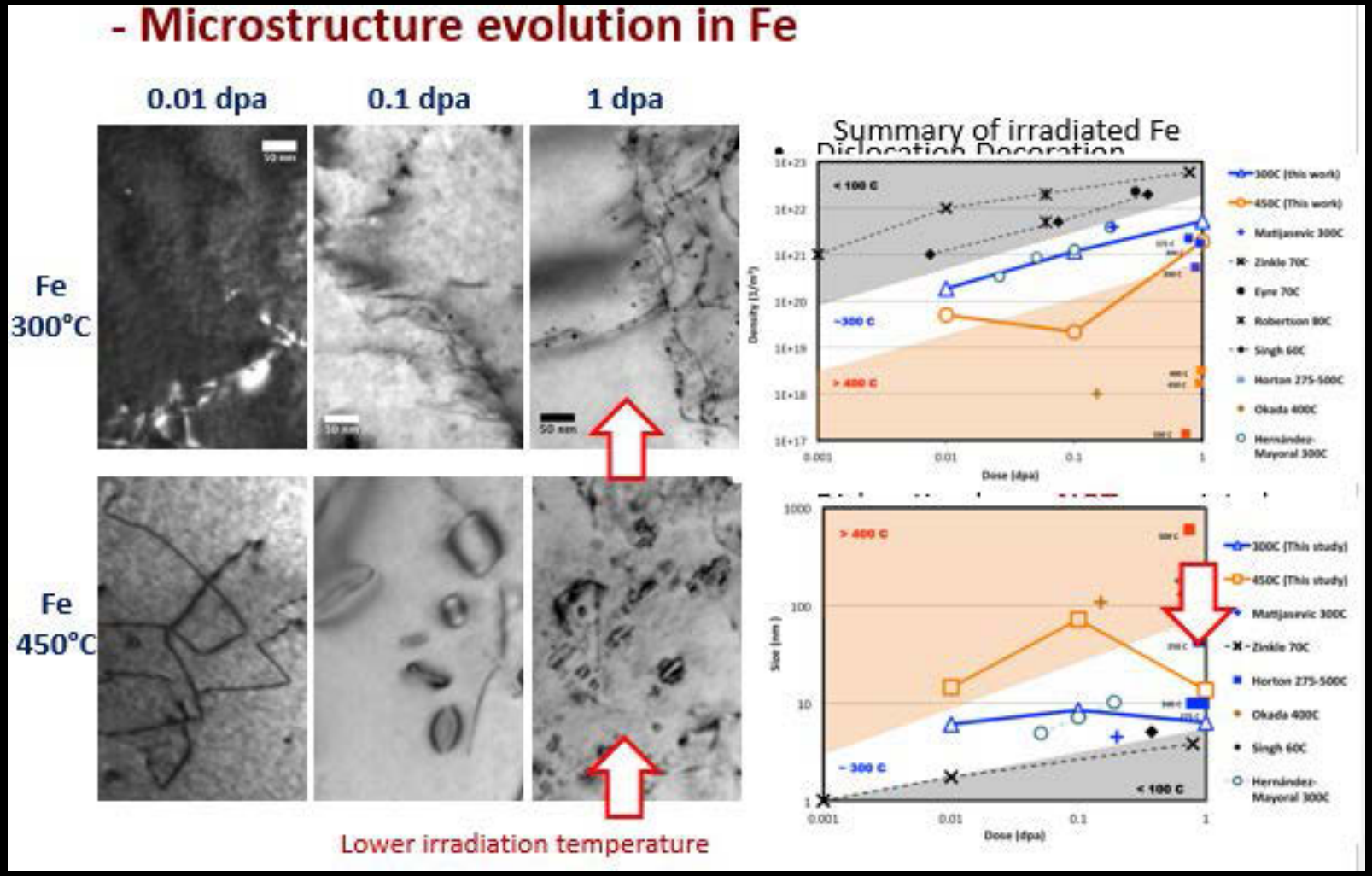
Summary of size, density and volu

Specimen	$N_{\alpha'}$ (x10^{23} 1/m³)	$r_{\alpha'}$ (n	
Fe-10Cr - 300 °C	7.66	1.1	
Fe-10Cr - 450 °C	1.08	1.0	
Fe-16Cr - 300 °C	27.1	1.2	
Fe-13Cr - 450 °C	12.6	1.08	0.85





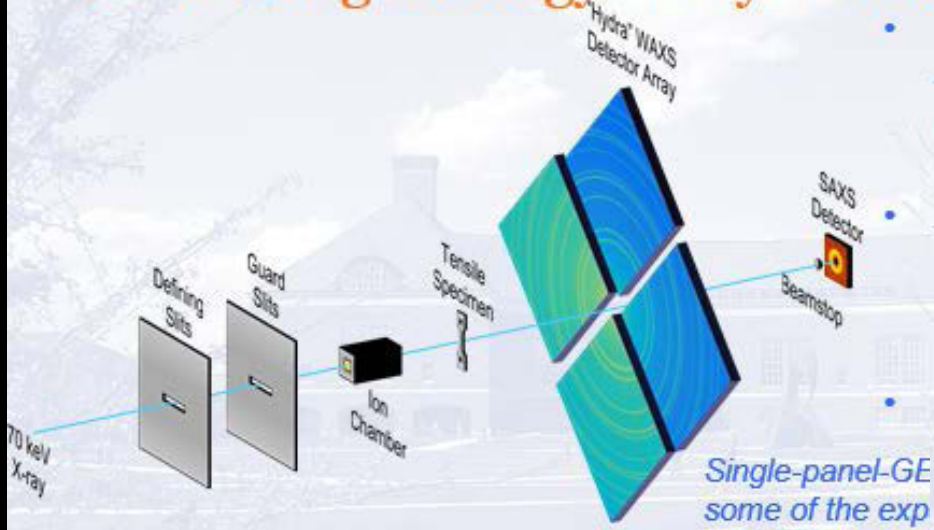
ATR: Dose & Temperature Effects



3. ANL APS: Deformation Dynamics

In-situ High-energy X-ray Diffraction

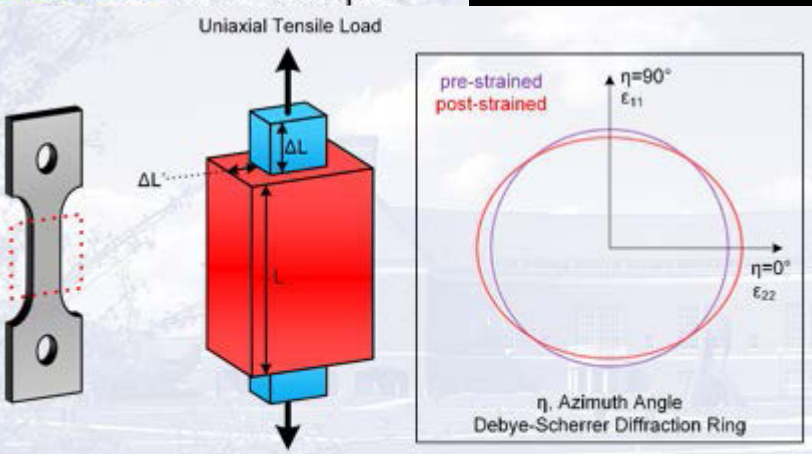
4



- *Sector 1-ID beamline at the Advanced Photon Source (APS)*
- *X-ray energy:* 70 keV (λ=0.0177nm) or 86 keV (λ=0.0144nm)
- *Beam size:* 300×300 μm²

Single-panel-GE
some of the exp

Experimental setup for in-situ synchrotron X-ray scatte

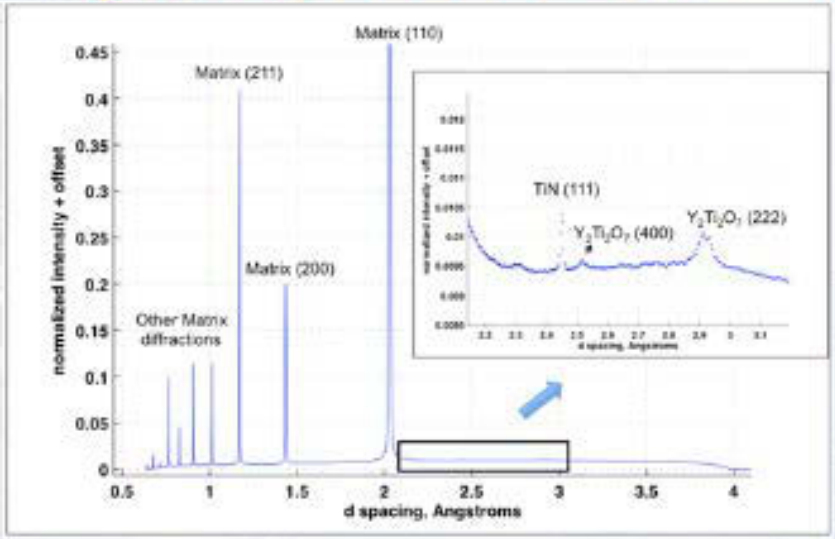


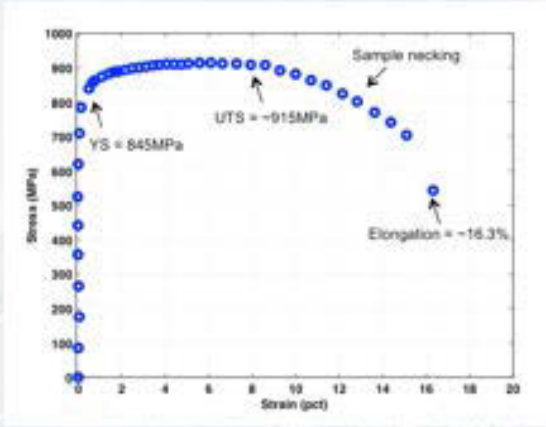
$$\varepsilon_{11} = \Delta L / L = (d_{t,90^\circ} - d_0) / d_0$$

$$\varepsilon_{22} = \Delta L' / L = (d_{t,0^\circ} - d_0) / d_0$$

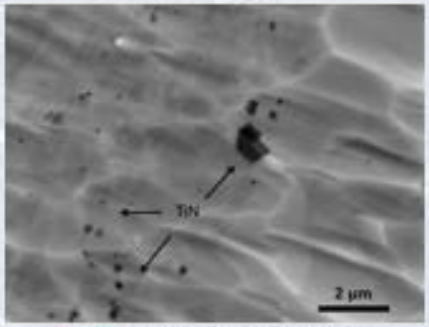
Results

9Cr ODS: Phases and Tensile Properties

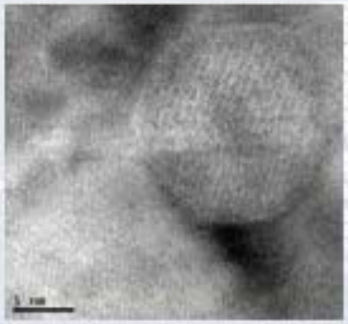




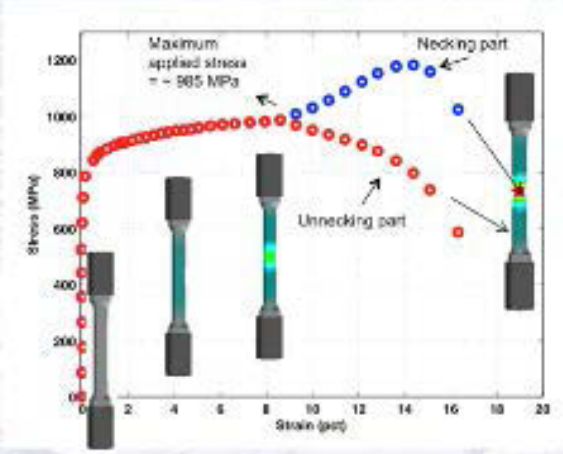
Engineering stress vs. strain



TiN Particles (50~2000 nm)



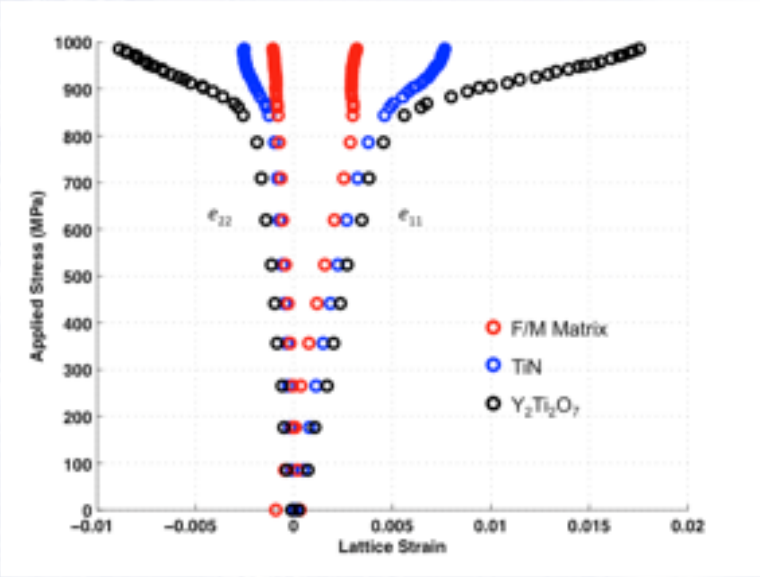
A Y-Ti-O nanoparticle showing its (222) d-spacing



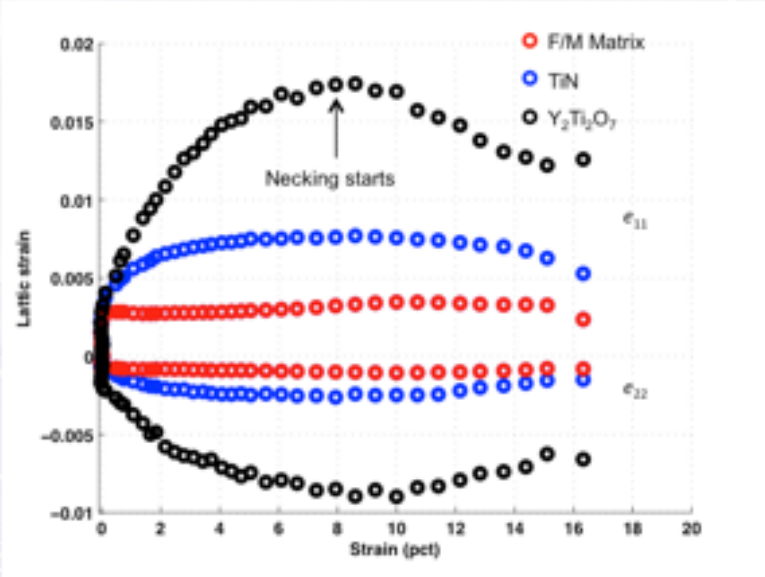
True stress vs. strain

Results

9Cr ODS: Load Partitioning



Applied stress vs. lattice strain for F/M matrix (321) reflection $Y_2Ti_2O_7$ (222) reflection, and TiN (111) reflection in the 9Cr ODS steel.



Lattice stress vs. macroscopic strain for F/M matrix (321) reflection $Y_2Ti_2O_7$ (222) reflection, and TiN (111) reflection in the 9Cr ODS steel.

- Load partition during plastic deformation
- Significant load placed on the nanoscale $Y_2Ti_2O_7$ particles (size effect)
- F/M matrix is slightly relaxed after sample yielding

Illinois ATR Experiments

Coordinated Choice of Alloys to Align with Previous Studies and Other Experiments

Table 1. Test Matrix (12 materials, 3 irradiation temperatures, 6 doses)

Alloy	Temperature (°C)	Dose (dpa)	Specimen Types
Model Alloy: Fe, Fe-9Cr, Fe-9Cr-0.1C, Fe-9Cr-0.5C, Fe-12Cr, Fe-12Cr-0.2C, Fe-12Cr-0.5C, Fe-14Cr*, Fe-19Cr*	300, 450, 550	0.01, 0.1, 0.5, 1.0, 5.0, 10	TEM, Miniature tensile
Commercial Alloys: T91, HT-9	300, 450, 550	0.01, 0.1, 0.5, 1.0, 5.0,	TEM, Miniature tensile
Developmental Alloys: MA-957	300, 450, 550	0.01, 0.1, 0.5, 1.0, 5.0,	TEM, Miniature tensile

* Single crystal materials, no miniature tensile specimens

Illinois ATR Experiments Timeline

- 2008 Experiment Accepted - $40K for specimens
- 2009 – PIE Funds Limited to at most 10 specimens, the other specimens will go into the Library
- Low dose requires pneumatic tube system

- 2012-2015 on third try, awarded NEUP to support science costs at Illinois, however, limited access to specimens and facilities

- NEUP should end 9/2015
- Access Only Proposal Process starts – Awarded two Access Only contracts to look at more samples: CAES & MRCAT
- RTE Options for limited samples

Illinois ATR Irradiations Issues

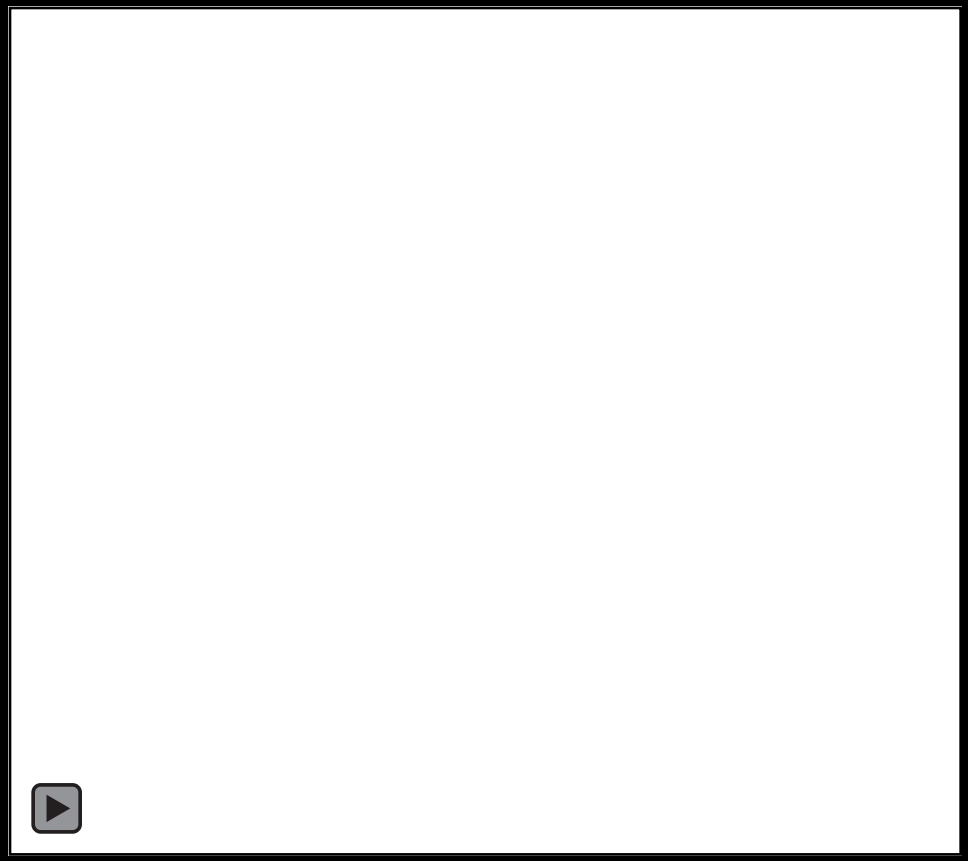
- Library
  - Access and Collaborations
- Dose & Temperature Histories
- General Access
  - Specimen Type and Preparation
  - Shipping and Interlab Coordination
    - ORNL, ANL-APS, LANL, …

The FUTURE

- Dynamic Experiments
  - In Situ Experiments
    - IVEM/Tandem, ANL-APS
  - Modeling
- 3D Tomography
  - Defect Structures and Interactions (TEM)
  - Atom Probe
  - Modeling

Loop Transformation

- A two-step transition was observed
- Dislocation loop become mobile and eventually migrate to the surface



Summary

- User Facilities are a tremendous resource
- Other DOE Programs (NEUP) have a very heavy reliance on these facilities to meet program goals
- For Universities, NSUF is an important training ground for students – the future of DOE Science
- Dynamic Experimentation the necessary link with modeling
- Access is still the limiting issue