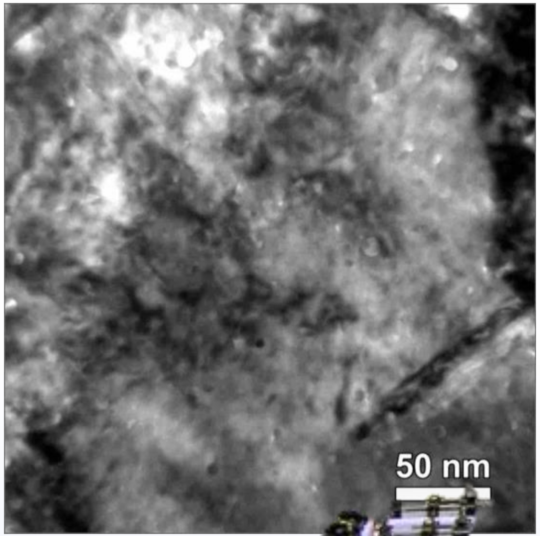
Deconvoluting Reactor Environmental Effects with Nanometer Resolution

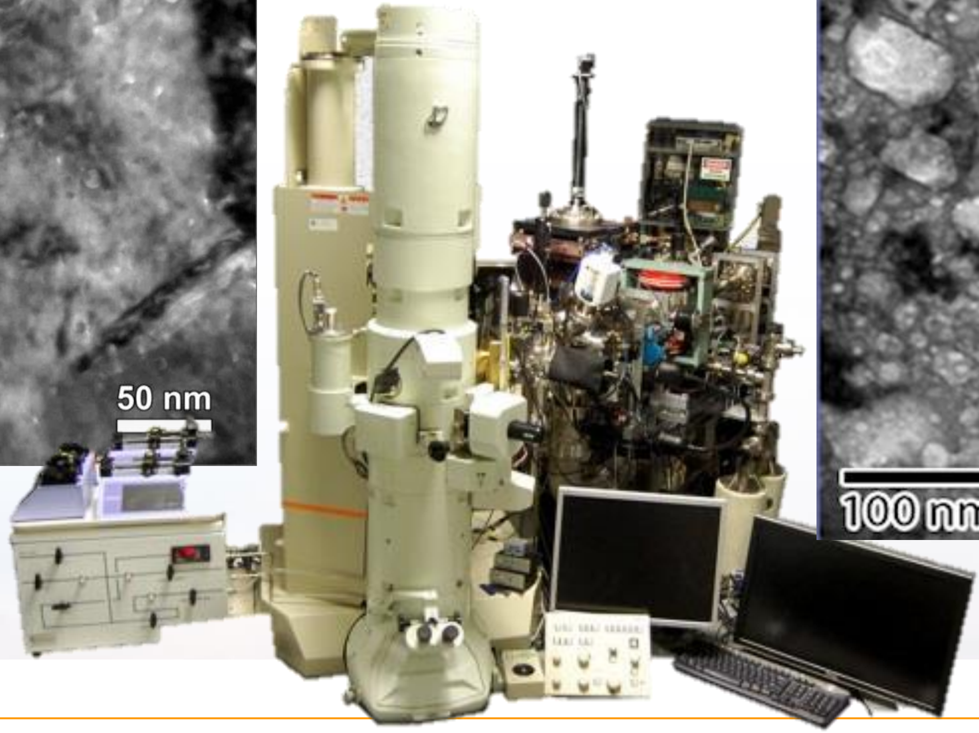
SAND2018-0715PE

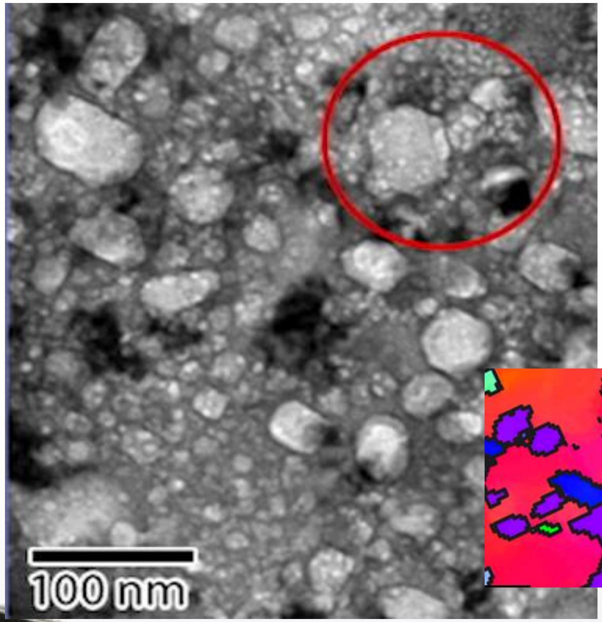
K. Hattar

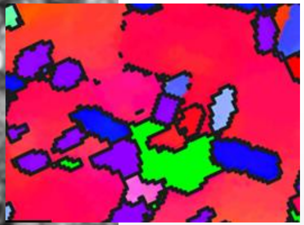
Ion Beam Lab at Sandia National Laboratories

January 22nd, 2018









Collaborators:

- IBL: C.Taylor, C.M. Barr, S.A. Briggs, D.C. Bufford, D. Buller, C. Chisholm, B.G. Clark, M.T. Marshall, B. Muntifering, S.H. Pratt, & P. Price
- Sandia: M. Abere, B. Boyce, T.J. Boyle, R. Dingreville, R.F. Hess, A.C. Kilgo, B.E. Klamm, W.M. Mook, J.D. Puskar, J.A. Scott, & J.A. Sharon
- External: A. Aitkaliyeva, H. Bei, P.J. Ferreira, K.J. Ganesh, E.P. George, D. Gross, P. Hosemann, J. Kacher, S. Maloy, A. Minor, J. Qu, S. Rajesekhara, I.M. Robertson, D. Stauffer, & Hysitron Inc.

This work was partially funded by the Division of Materials Science and Engineering, Office of Basic Energy Sciences, U.S. Department of Energy. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

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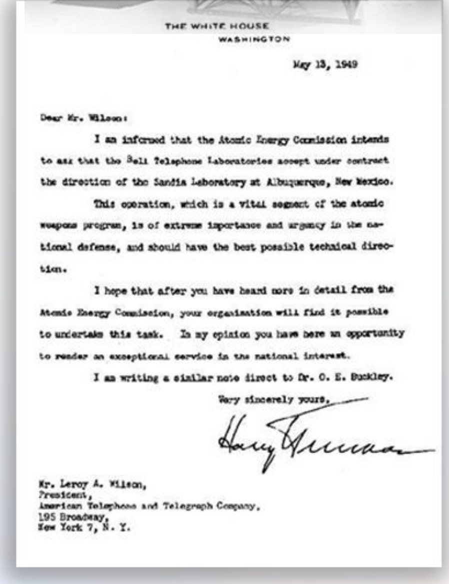
"Exceptional service in the national interest"



THE WHITE HOUSE
WASHINGTON

May 13, 1949

Dear Mr. Wilson:

I am informed that the Atomic Energy Commission intends to ask that the Bell Telephone Laboratories accept under contract the direction of the Sandia Laboratory at Albuquerque, New Mexico.

This operation, which is a vital segment of the atomic weapons program, is of extreme importance and urgency in the national defense, and should have the best possible technical direction.

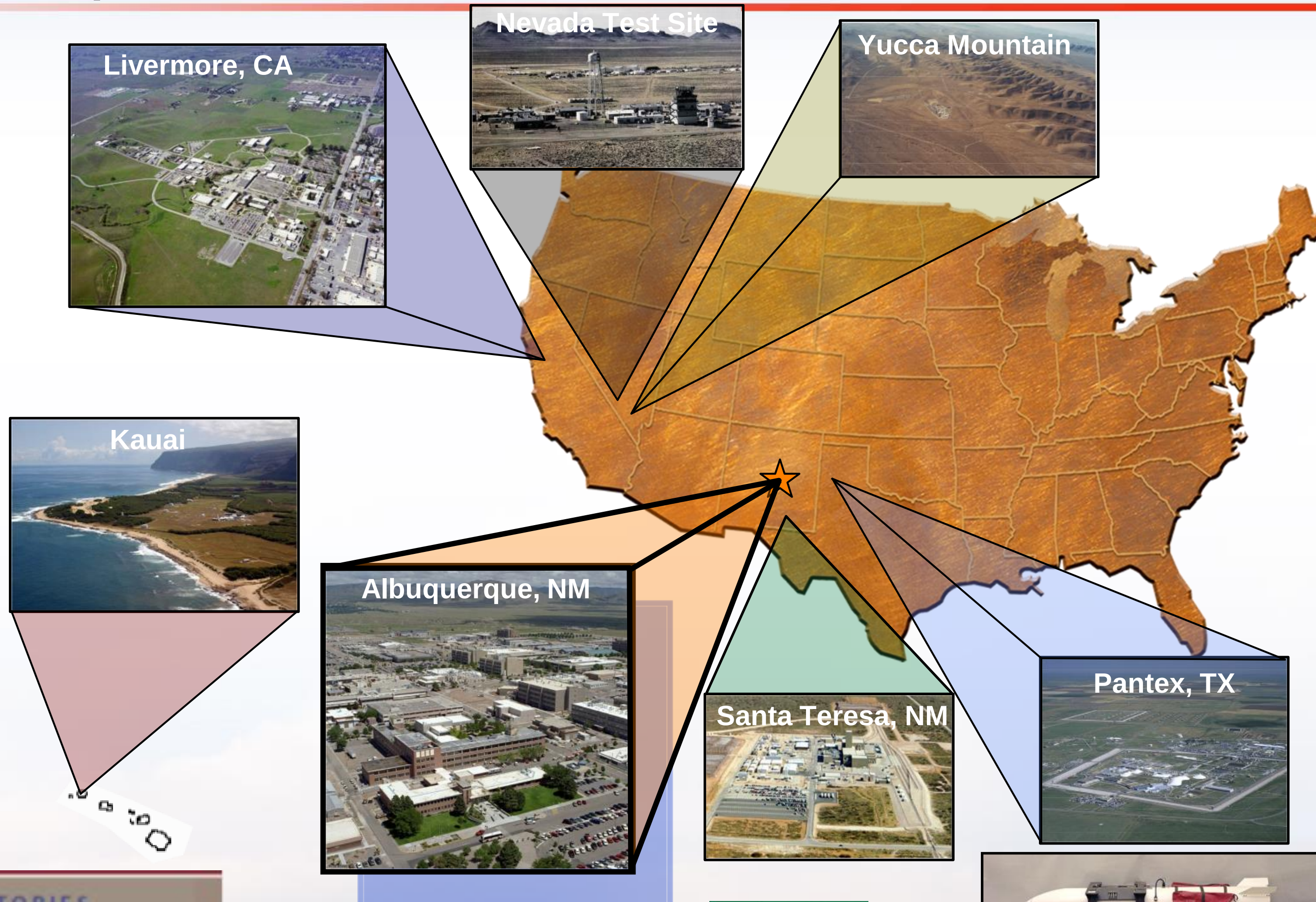
I hope that after you have heard more in detail from the Atomic Energy Commission, your organization will find it possible to undertake this task. In my opinion you have here an opportunity to render an exceptional service in the national interest.

I am writing a similar note direct to Dr. O. E. Buckley.

Very sincerely yours,

Harry Truman

Mr. Leroy A. Wilson,
President,
American Telephone and Telegraph Company,
195 Broadway,
New York 7, N. Y.



"Sandia develops advanced technologies to ensure global peace." – S. Younger

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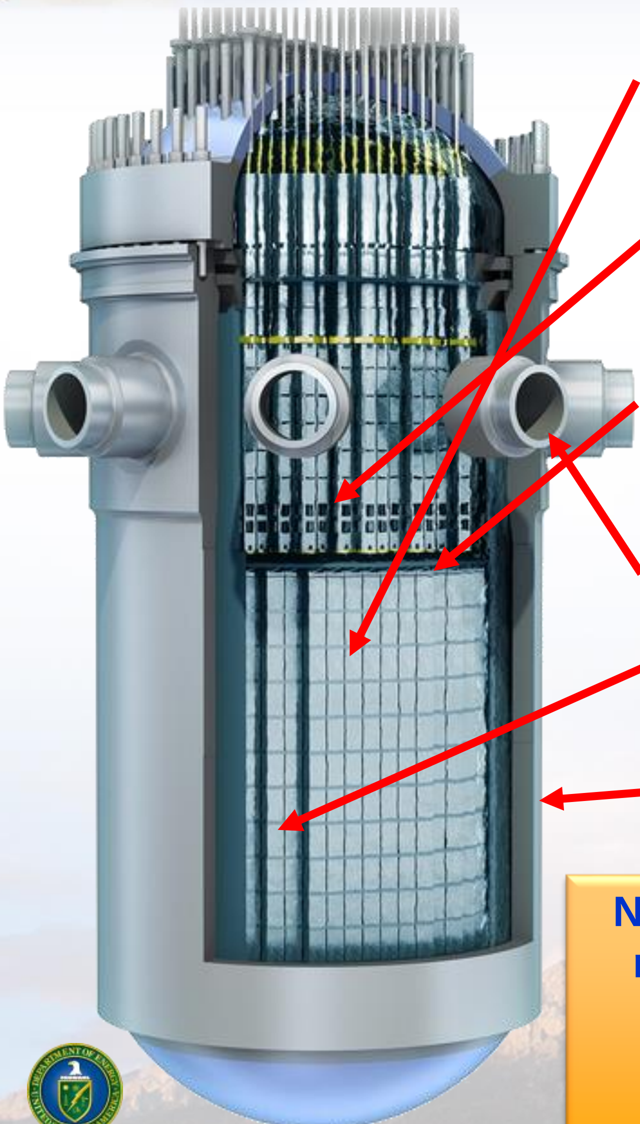
Reactor Materials Challenges



Nuclear Fuels

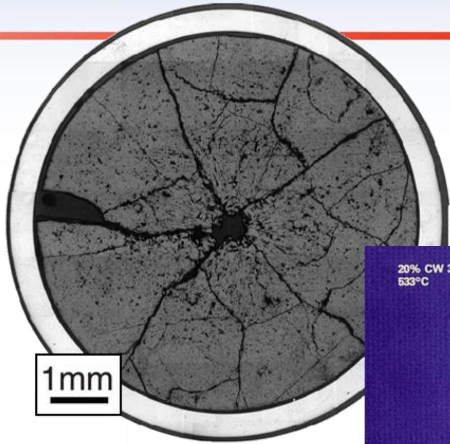
Displacement Damage Effects

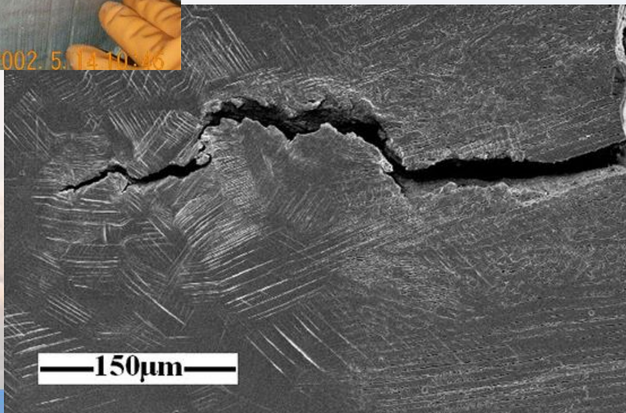
Transmutation Effects

Corrosion

Creep

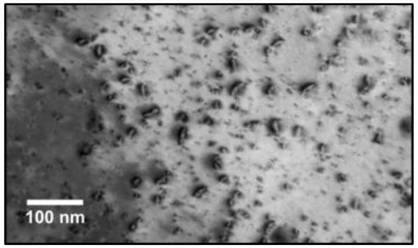
Fatigue



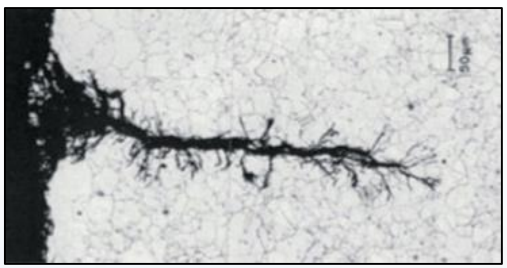


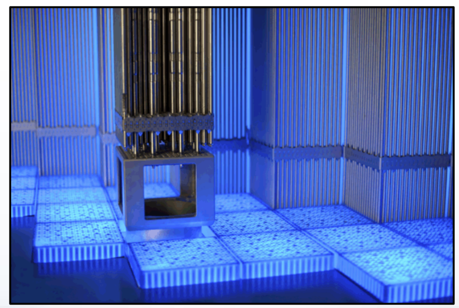
Nuclear reactors pose unique materials challenges due to degradation effects from combined environmental stressors

Investigating the nm Scale to Understand the km Scale



1 nm | 1 μm | 1 mm | 1 m | 1 km | 10^7 m





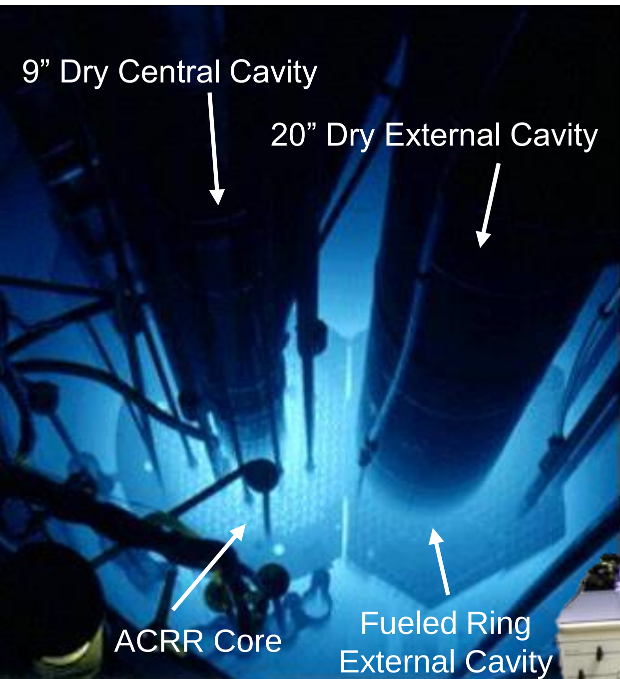
In situ Ion Irradiation TEM (I^3TEM)

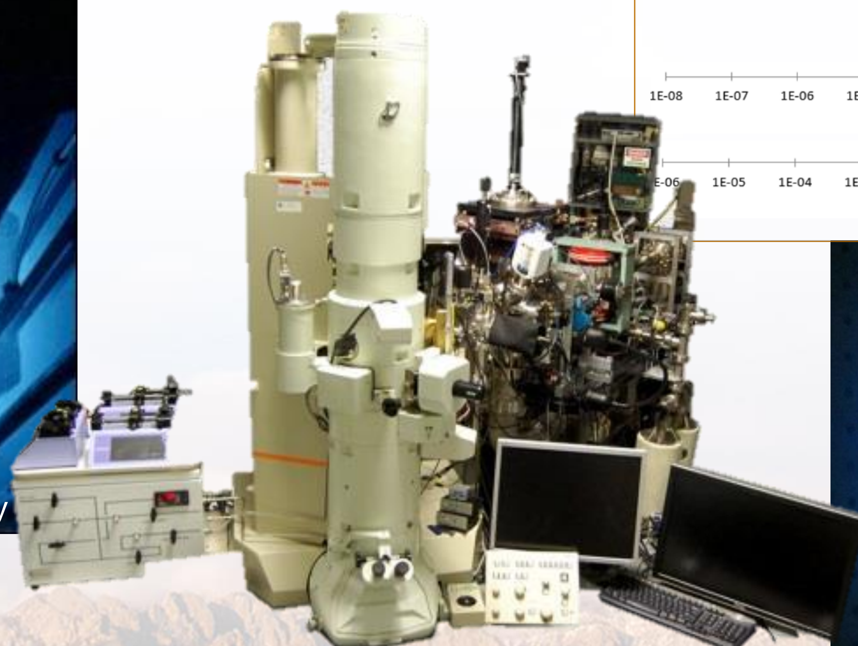
Ion Beam Lab (IBL)

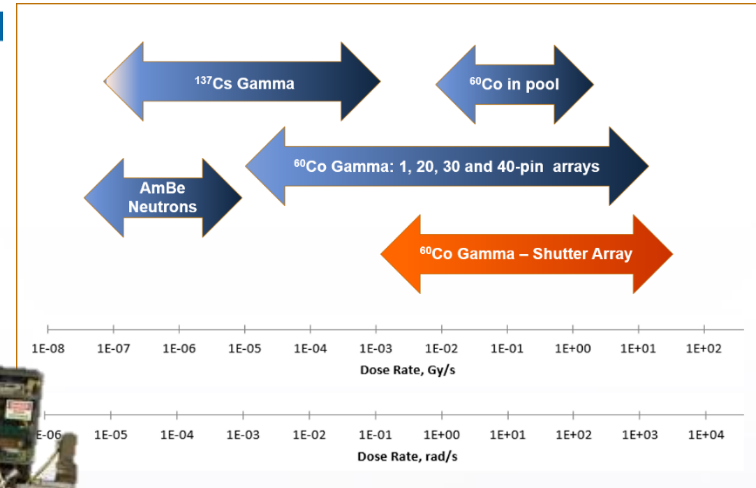
Sandia's NSUF Capabilities

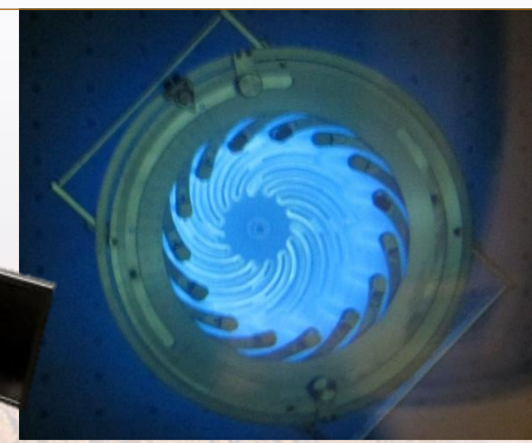
K. Hattar, D. Hanson, W. Martin, M. Wasiolek

NSUF
Nuclear Science
User Facilities





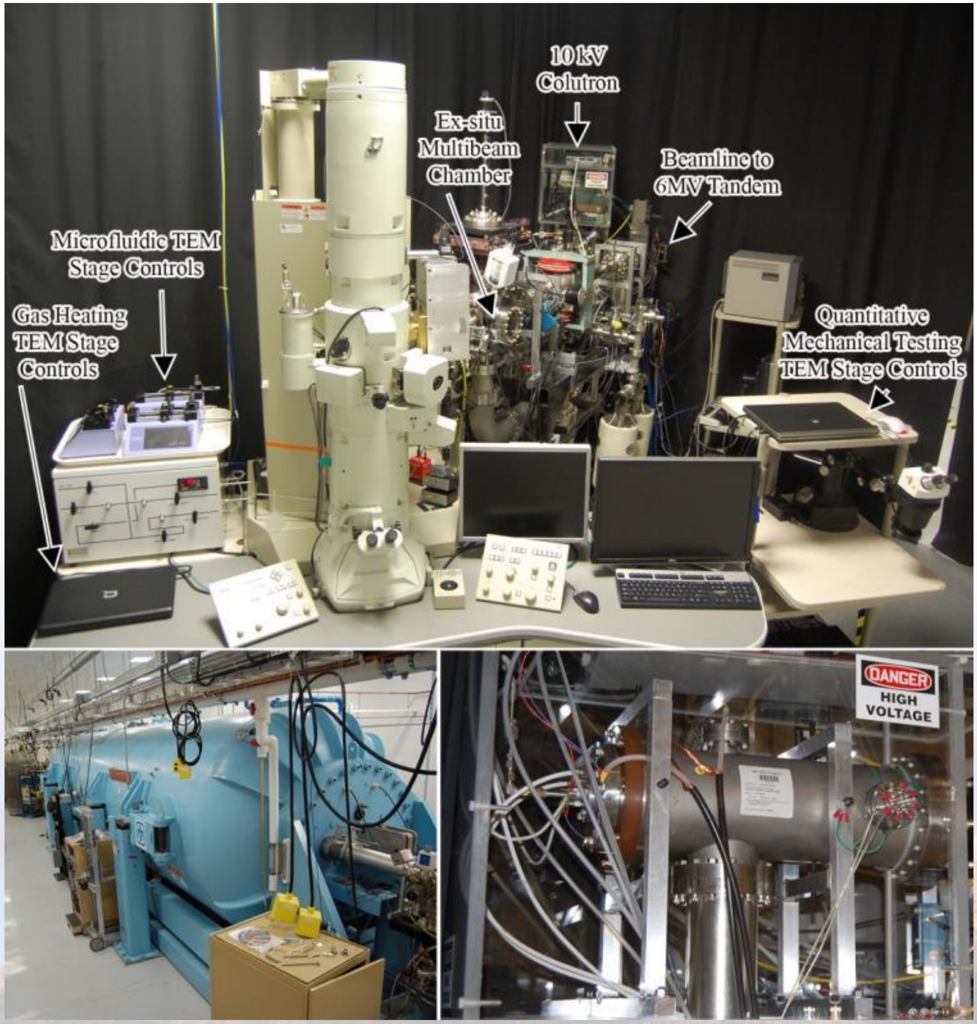




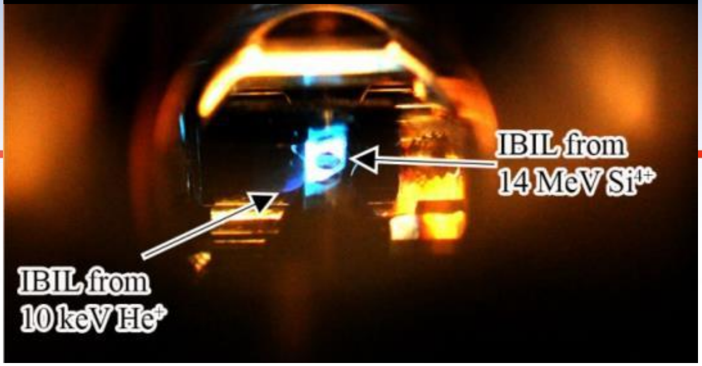
Sandia's Concurrent *In situ* Ion Irradiation TEM Facility

Collaborator: D.L. Buller

10 kV Colutron - 200 kV TEM - 6 MV Tandem

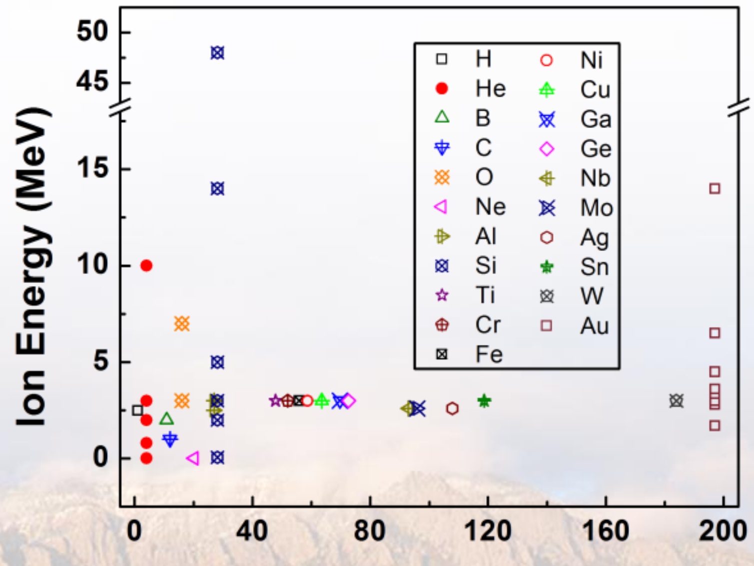


IBIL from a quartz stage inside the TEM



Direct real time observation of ion irradiation, ion implantation, or both with nanometer resolution

Ion species & energy introduced into the TEM



Atomic Mass

Sandia National Laboratories

Benefits & Limitations of *in situ* TEM

Benefits

1. **Real-time nanoscale resolution observations of microstructural dynamics**

Limitations

1. **Predominantly limited to microstructural characterization**
   - Some work in thermal, optical, and mechanical properties
2. **Limited to electron transparent films**
   - Can often prefer surface mechanisms to bulk mechanisms
   - Local stresses state in the sample is difficult to predict
3. **Electron beam effects**
   - Radiolysis and Knock-on Damage
4. **Vacuum conditions**
   - 10^{-7} Torr limits gas and liquid experiments feasibility
5. **Local probing**
   - Portions of the world study is small

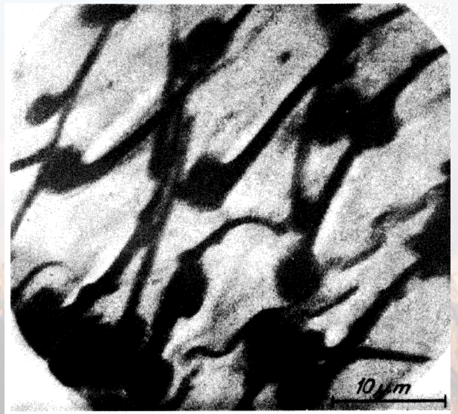


Fig. 6: Wing surface of the house fly.
(First internal photography, U = 60 kV, M_{el} = 2200)
(Driest, E., and Müller, H.O.: Z. Wiss. Mikroskopie 52, 53-57 (1935)

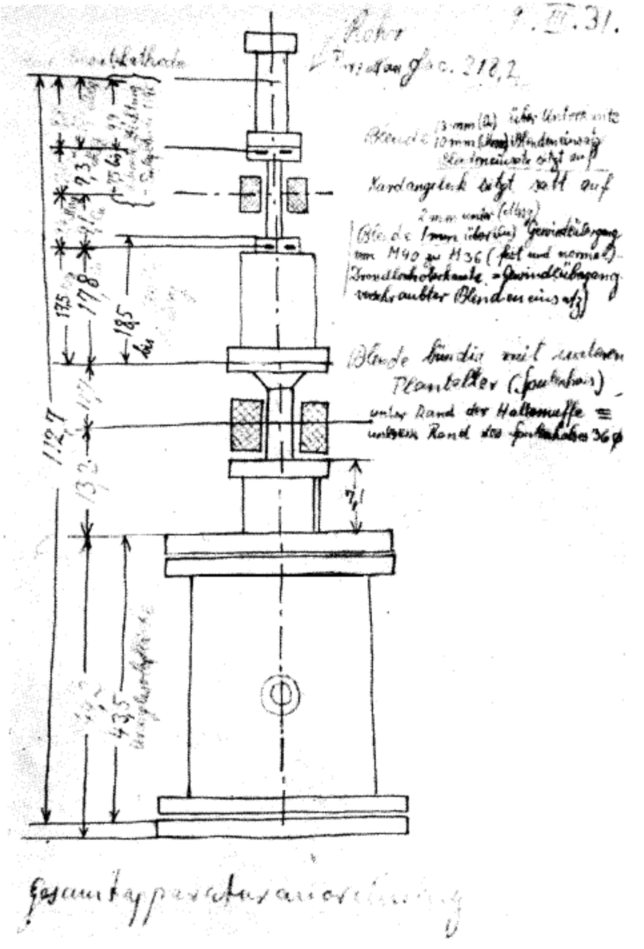
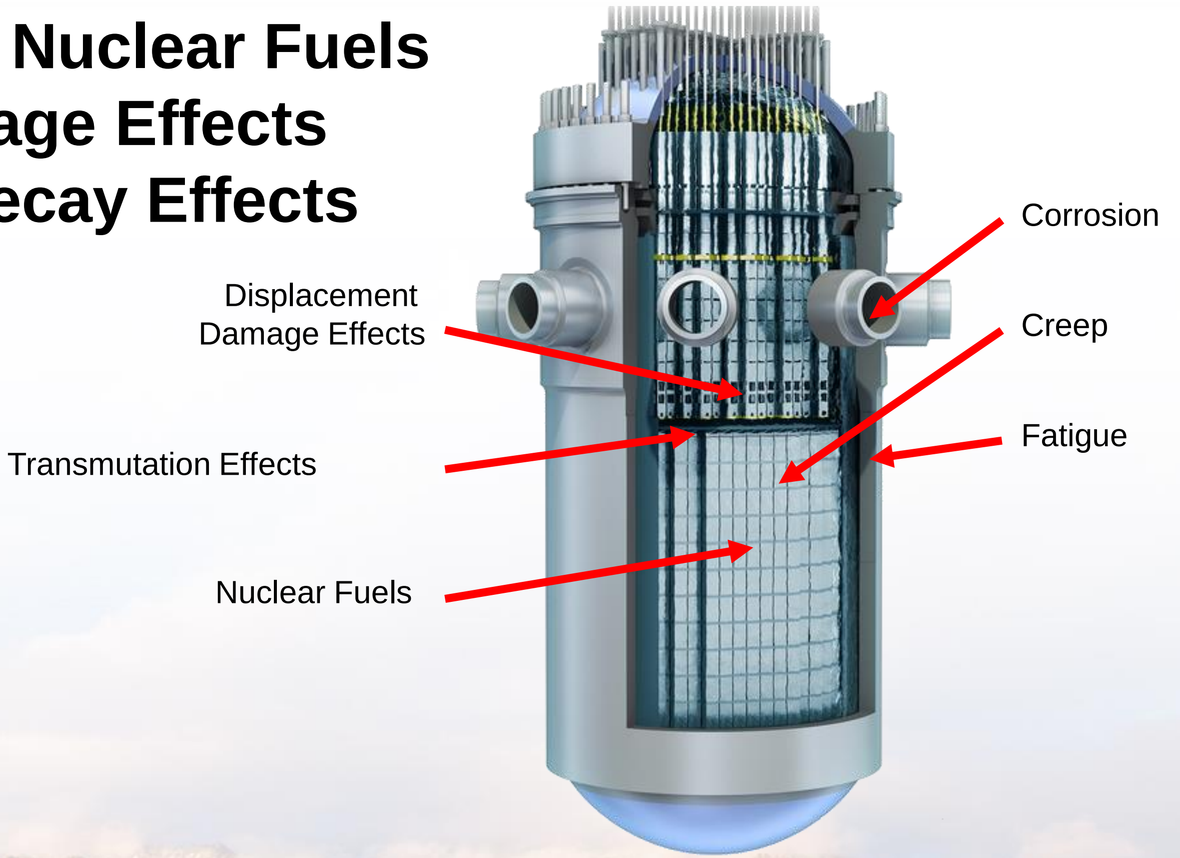


Fig. 2: Sketch by the author (9 March 1931) of the cathode ray tube for testing one-stage and two-stage electron-optical imaging by means of two magnetic electron lenses (electron microscope) [8].

E. Ruska Nobel lecture (1986)

Outline

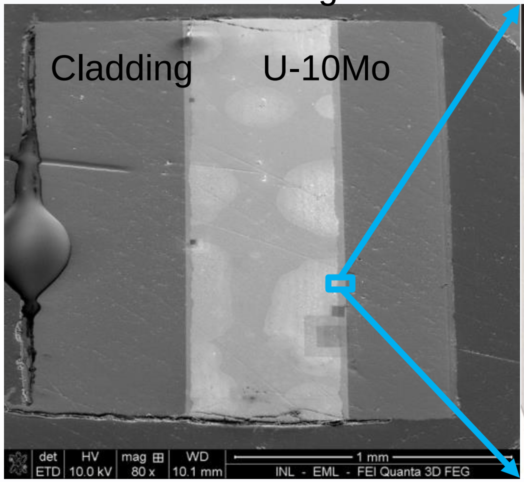
1. Characterization of Nuclear Fuels
2. Displacement Damage Effects
3. Transmutation & Decay Effects
4. Corrosion
5. Creep
6. Fatigue



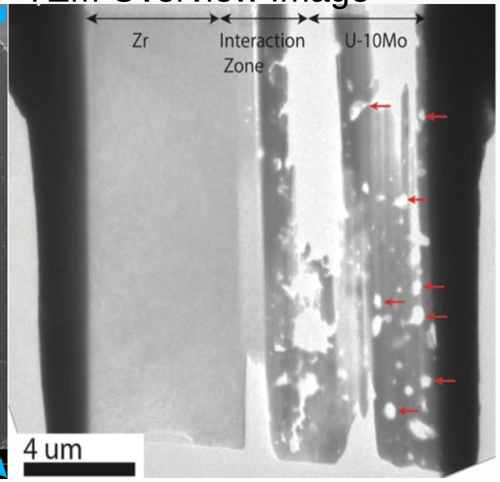
Post Irradiation Characterization of U-10Mo/Zr monolithic interface

Collaborators: C. Barrr, A. Aitkaliyeva

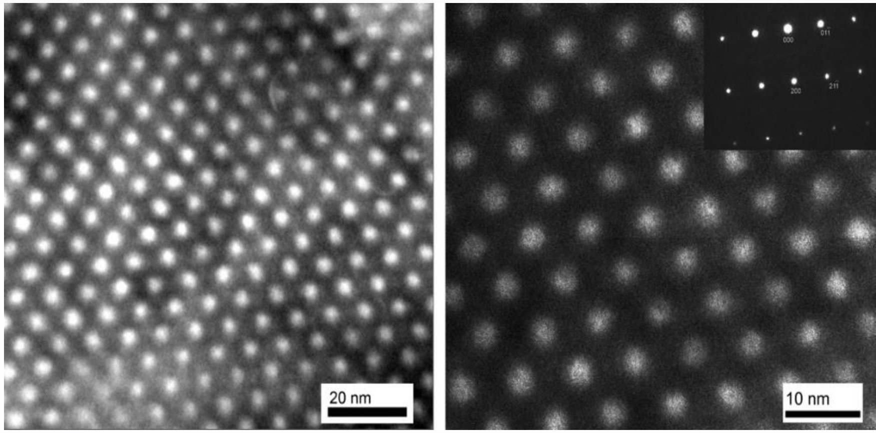
SEM Overview Image



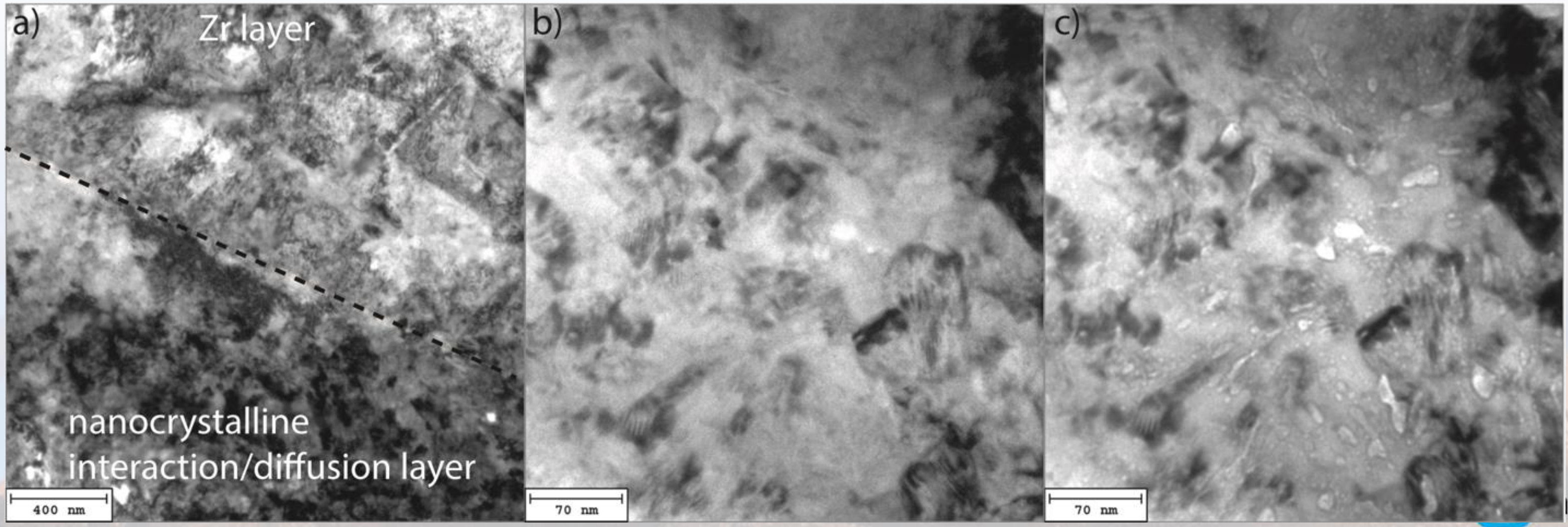
TEM Overview Image



Gas superlattice bubbles ZA [110] in U-10Mo.

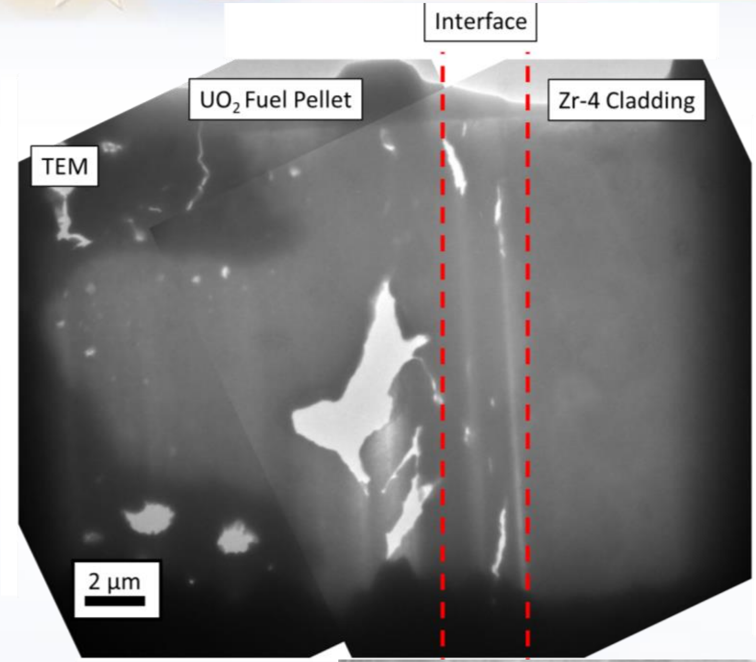


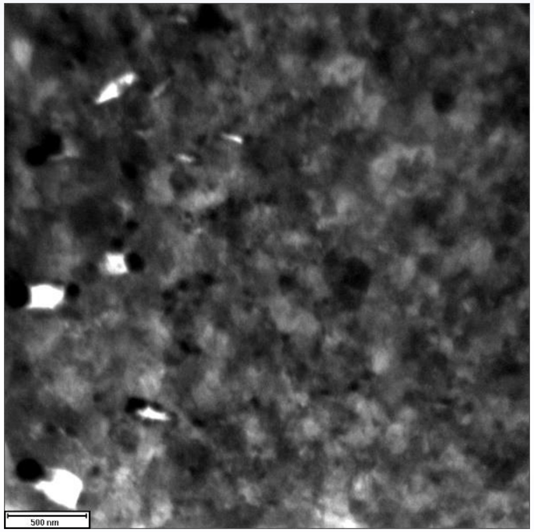
- U-10Mo Monolithic fuel with fission density (fiss/cm^3) = 4.4 x 10^{21}
- Average gas superlattice bubble diameter distribution is 3.5 ± 0.25 nm diameter.



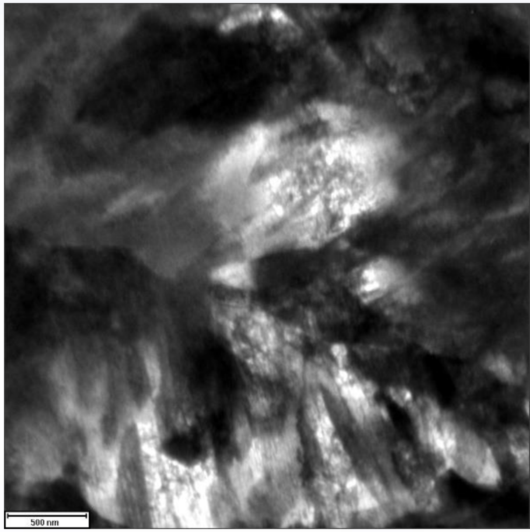
Zr layer and interaction layers where in-focus image of interaction layer (center) and de-focus (-2um) image of interaction layer with high level of porosity apparent

Characterization of HB Robinson



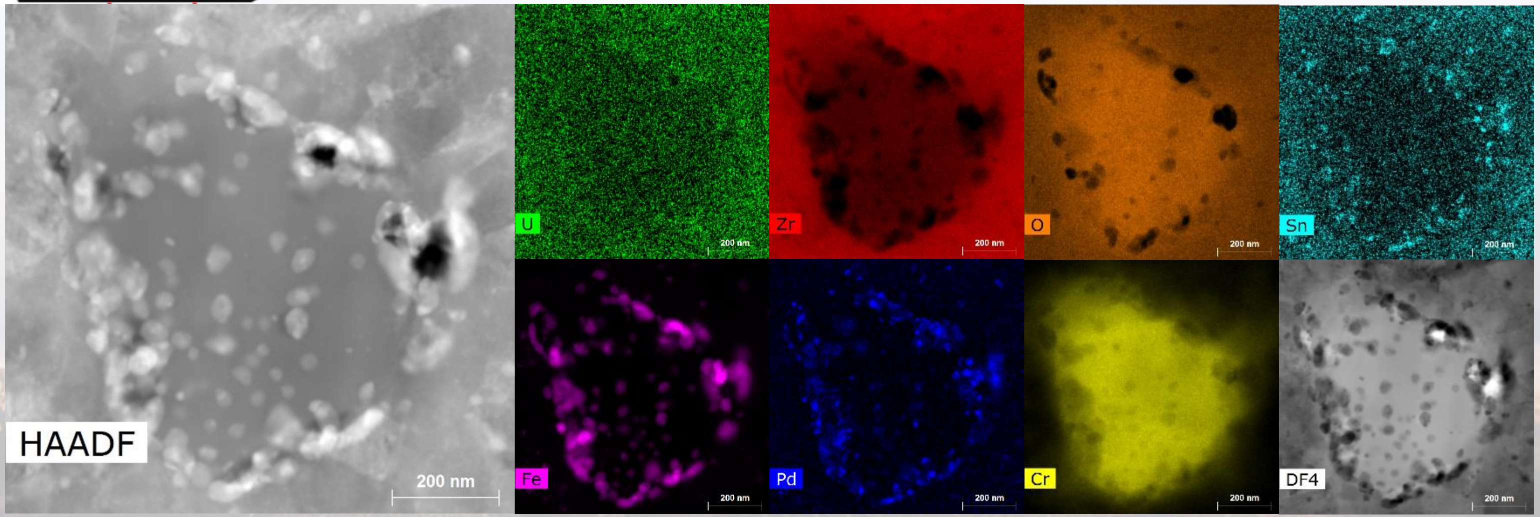


UO_2 Fuel Pellet



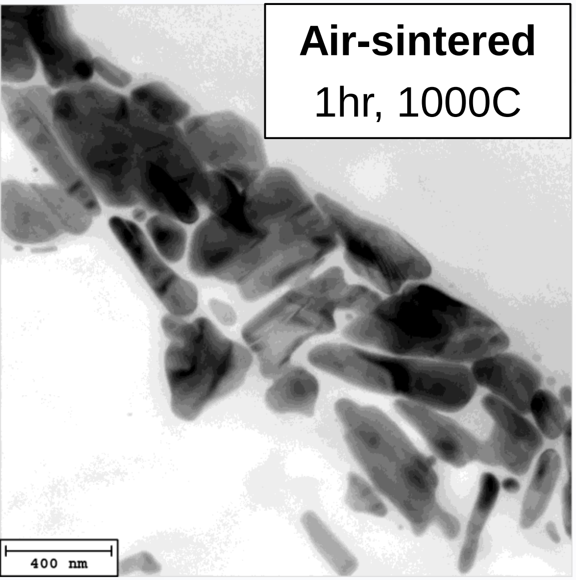
Zr-4 Cladding

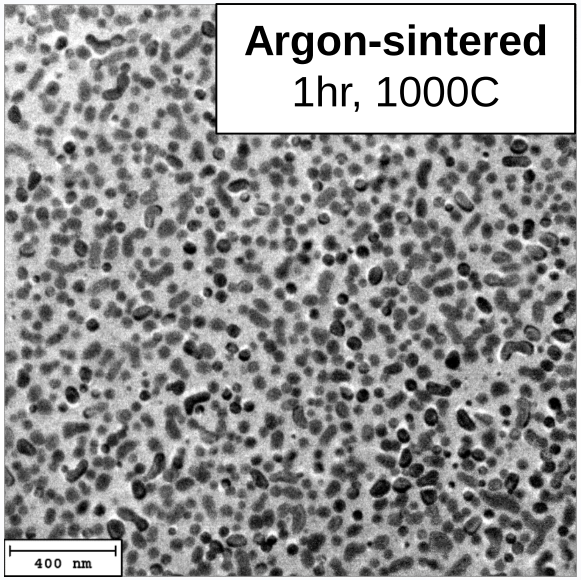
Complex Nanostructured Material Resulting from Typical Nuclear Reactor Operation

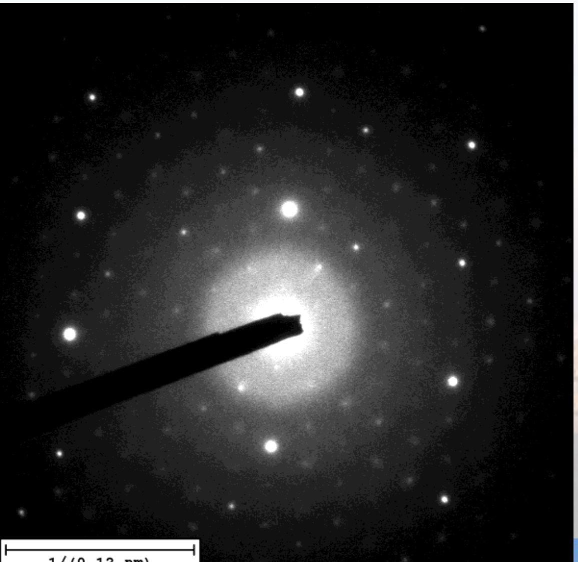


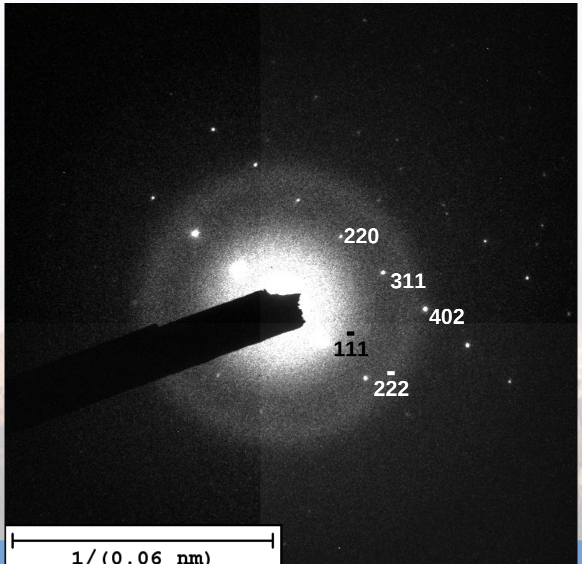
Uranium Oxide Formation in Different Sintering Environments

Collaborators: S.A. Briggs, R. Hess, and B. Klamm









Nanoparticles prepared from solution of $UO_2(NO_3)_2$, PEI, and EDTA

Ar-sintered specimen phase diffraction patterns map to fluorite/FCC structure characteristic of UO_2

- Larger lattice parameter suggests hypostoichiometric uranium-dioxide phase (UO_{2-x})

Air-sintered specimens do not map to fluorite/FCC

- Likely a hyperstoichiometric uranium-oxide phase (U_3O_8, U_4O_9)

Characterization of ion-irradiation response is work-in-progress

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Outline

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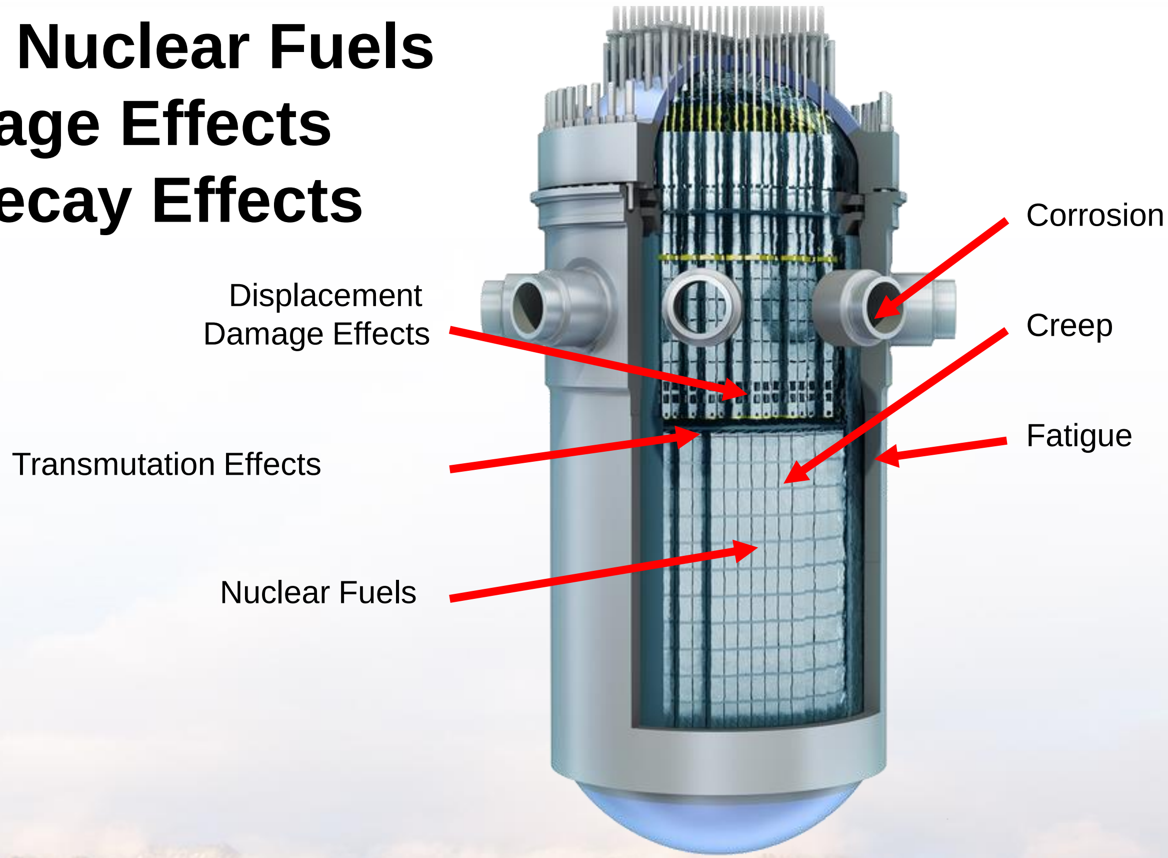


Cumulative Effects of Ion Irradiation as a Function of Ion Energy and Au Particle Size

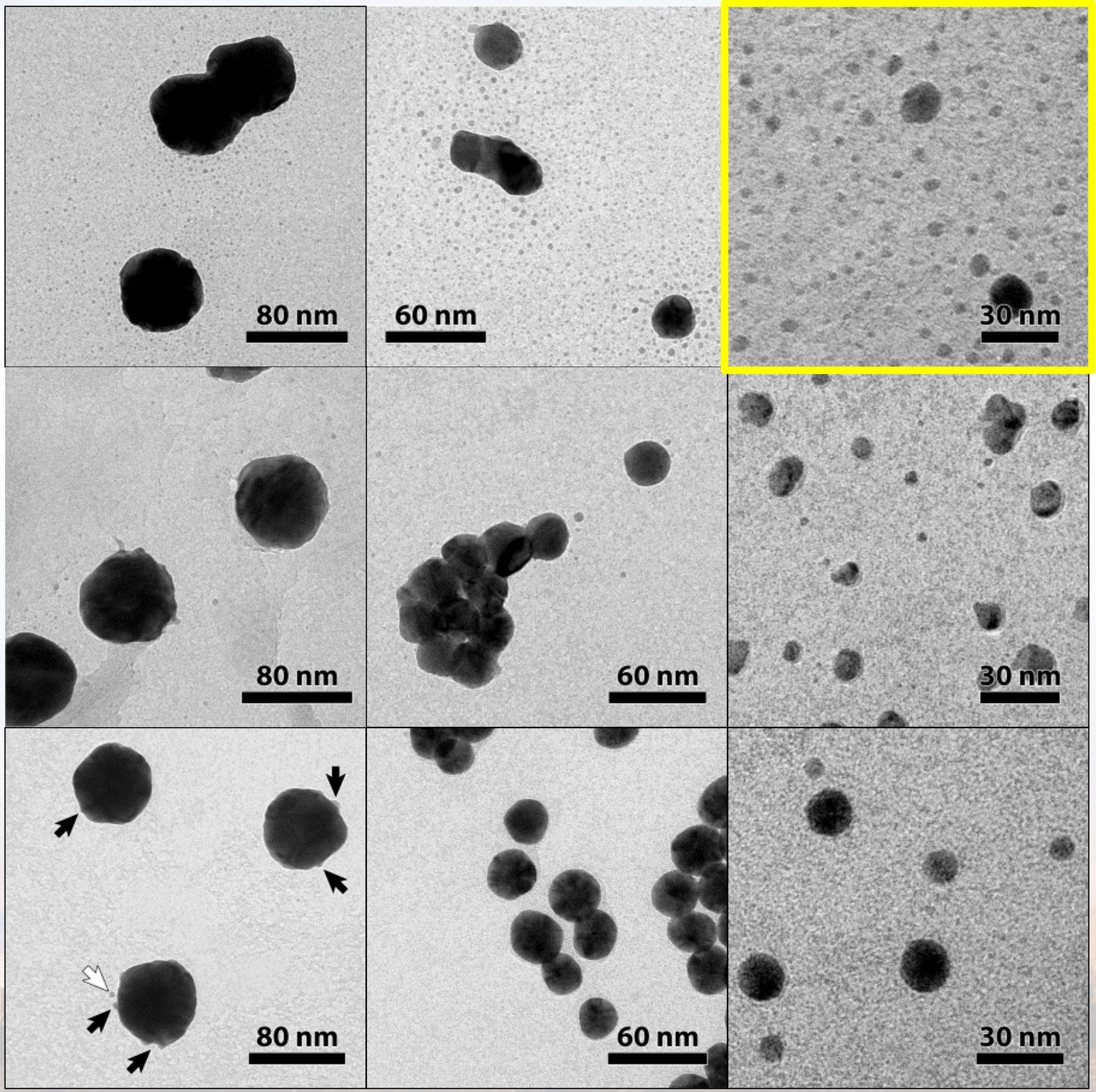
Collaborator: D.C. Bufford

60 nm | **20 nm** | **5 nm**

46 keV Au^{1-}
3.4 × 10^{14} /cm^2

2.8 MeV Au^{4+}
4 × 10^{13} /cm^2

10 MeV Au^{8+}
1.3 × 10^{12} /cm^2



Particle and ion energy dictate the ratio of sputtering, particle motion, particle agglomeration, and other active mechanisms

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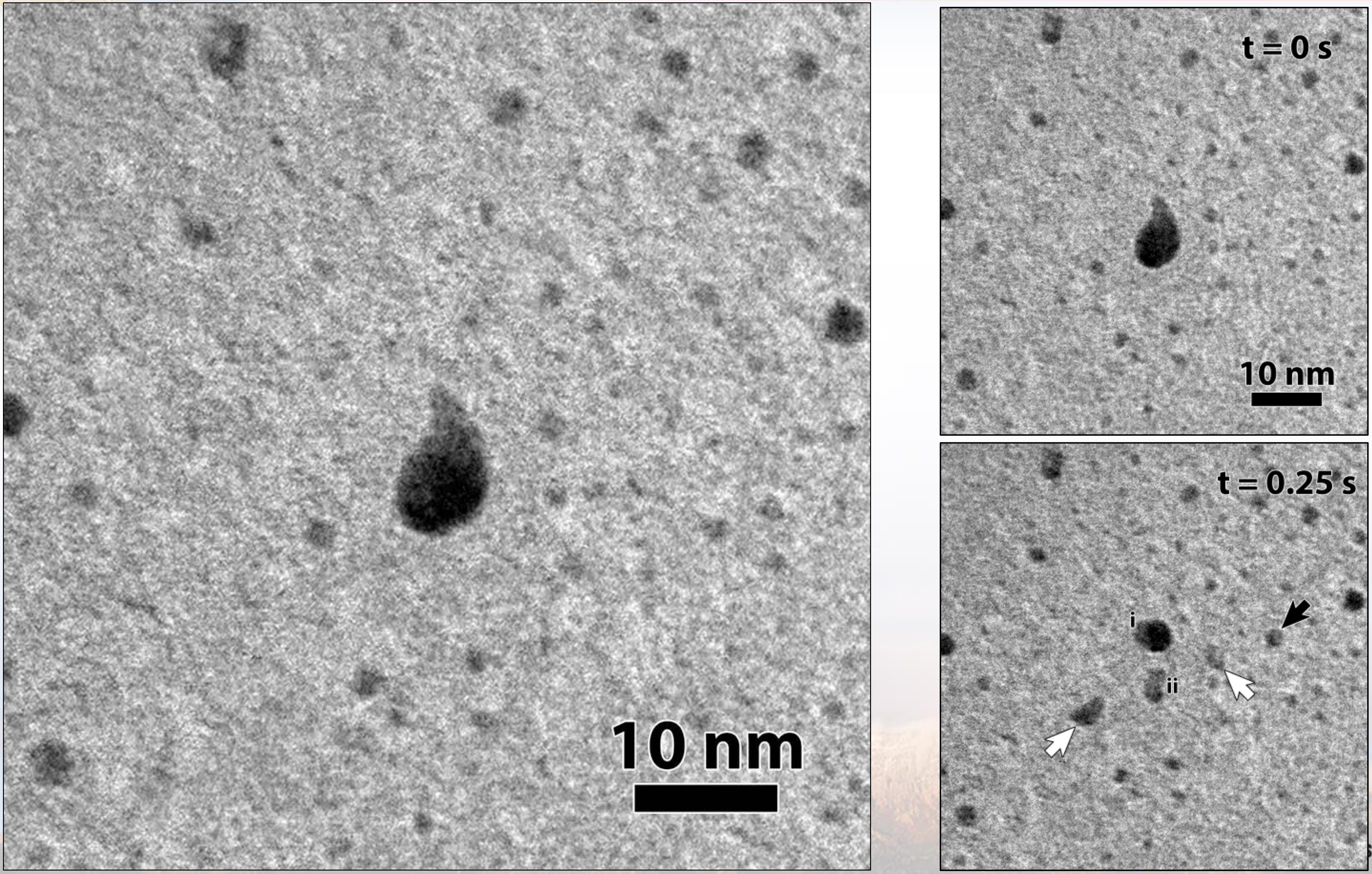
Single Ion Effects with 46 keV Au^{1-} ions: 5 nm

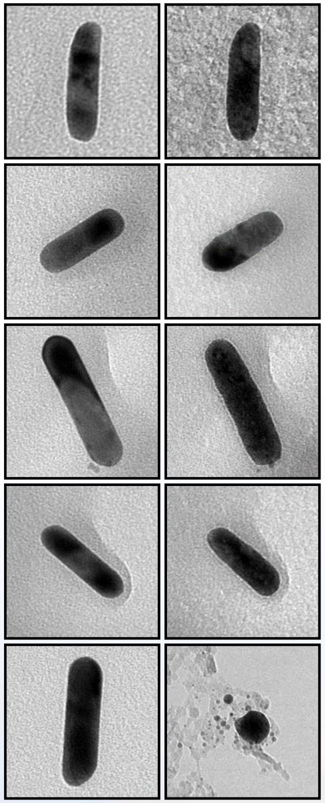
Collaborator: D.C. Bufford

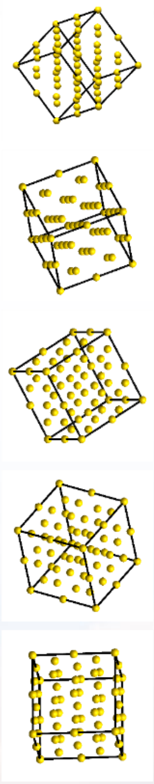


Exploring Radiation effects in Au Nanorods

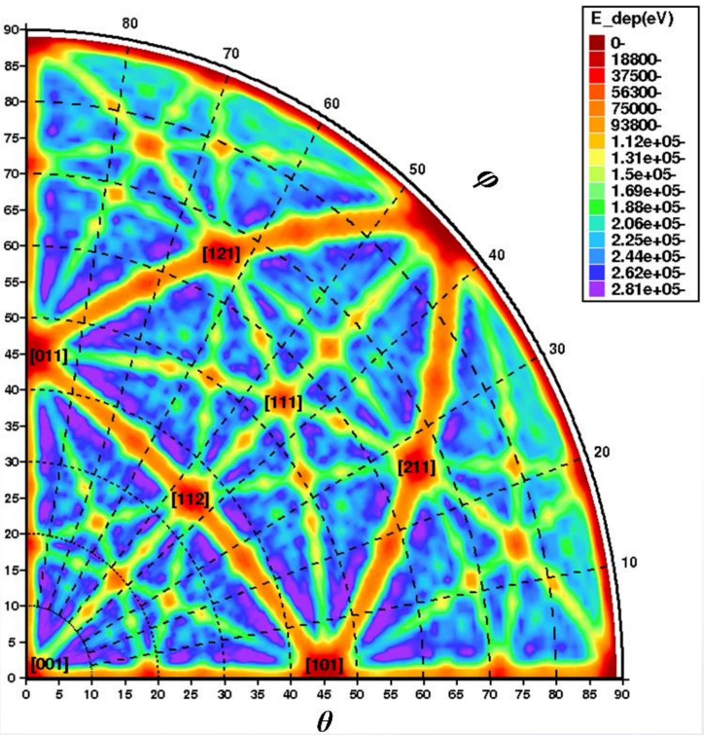
Collaborators: J. A. Hinks, F. Hibberd, A. Ilinov, D. C. Bufford, F. Djurabekova, G. Greaves, A. Kuronen, S. E. Donnelly & K. Nordlund

50 nm



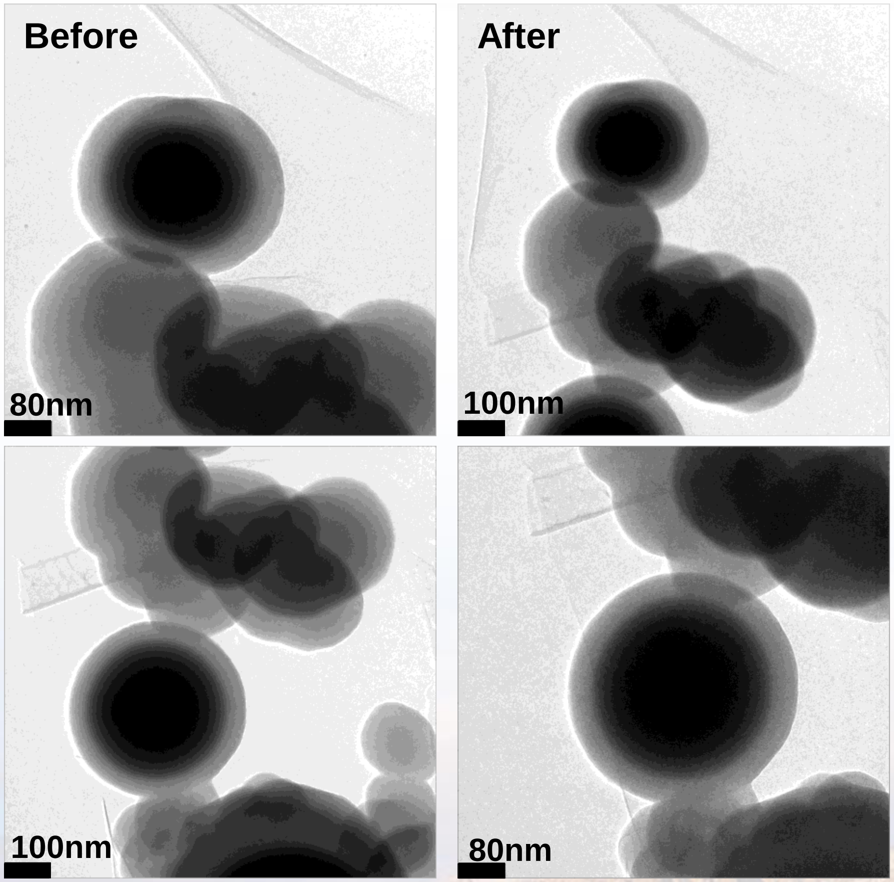


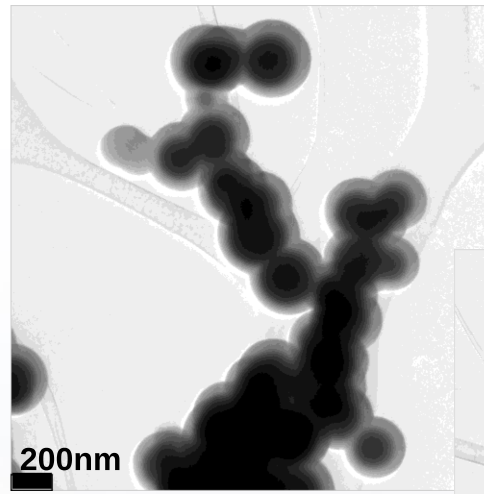
	Fluence (ions.cm^{-2}) 0	Fluence (ions.cm^{-2}) 3.6×10^{15}	Sputtering ΔV (%)	Sputtering Atoms.ion^{-1}	Irradiation orientation: Model view	θ	φ
AuNR-1			+8.1%	–2.1		39.1°	21.9°
AuNR-2			+3.5%	–1.0		42.8°	13.7°
AuNR-3			+1.5%	–0.3		25.6°	30.1°
AuNR-4			–1.2%	0.3		59.6°	46.0°
AuNR-5			–89.7%	30.1		18.5°	75.4°



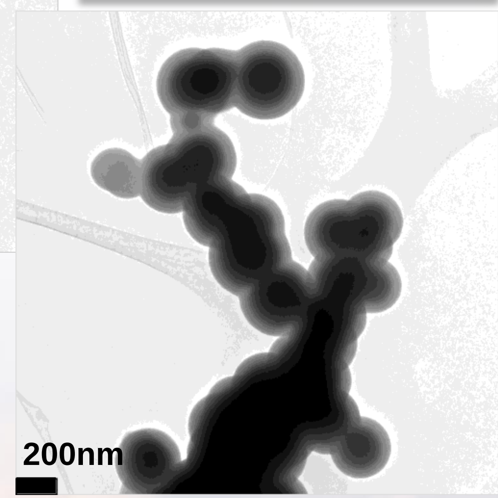
Crystal Orientation Matters!

Irradiation of Amorphous Hf Oxide Nanoparticles with 10 nA of 3 MeV Cu Resulted in No Obvious Changes





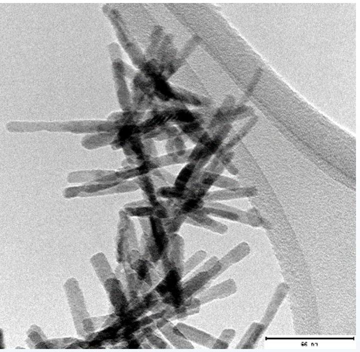
Sample was placed in tomography holder, tilted to 80°, and irradiated for an additional 2.5 h. No obvious changes.



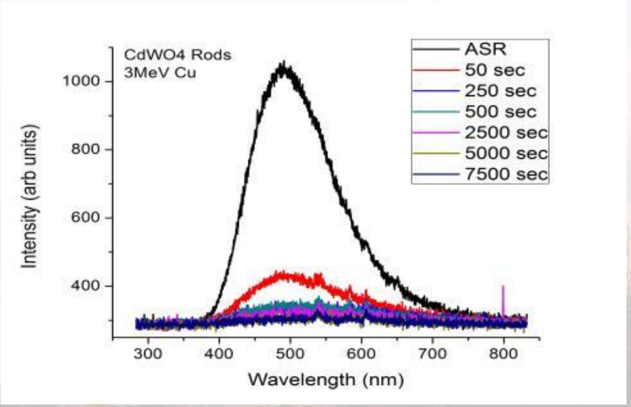
3 hour irradiation: no obvious changes.

Radiation Tolerance is Needed in Advanced Scintillators for Non-proliferation Applications

Contributors: S.M. Hoppe, B.A. Hernandez-Sanchez, T. Boyle

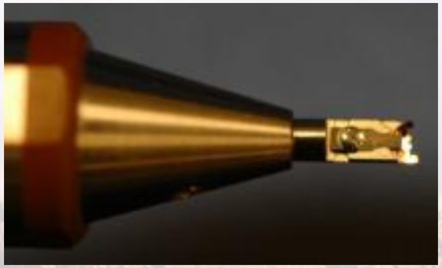


High-Z nanoparticles ($CdWO_4$) are promising, but are radiation sensitive



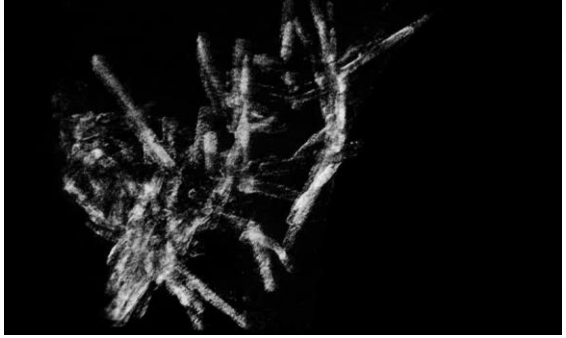
***In situ* Ion Irradiation TEM (I^3TEM)**

Hummingbird tomography stage

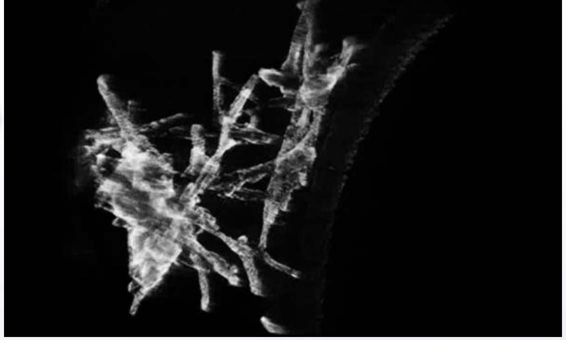


Tomography of Irradiated $CdWO_4$:
3 MeV Cu^{3+} at ~30 nA

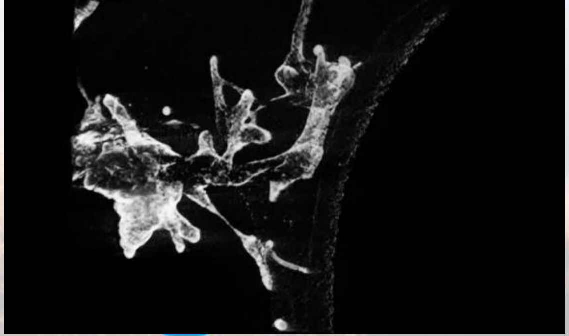
Un-irradiated



5 minutes



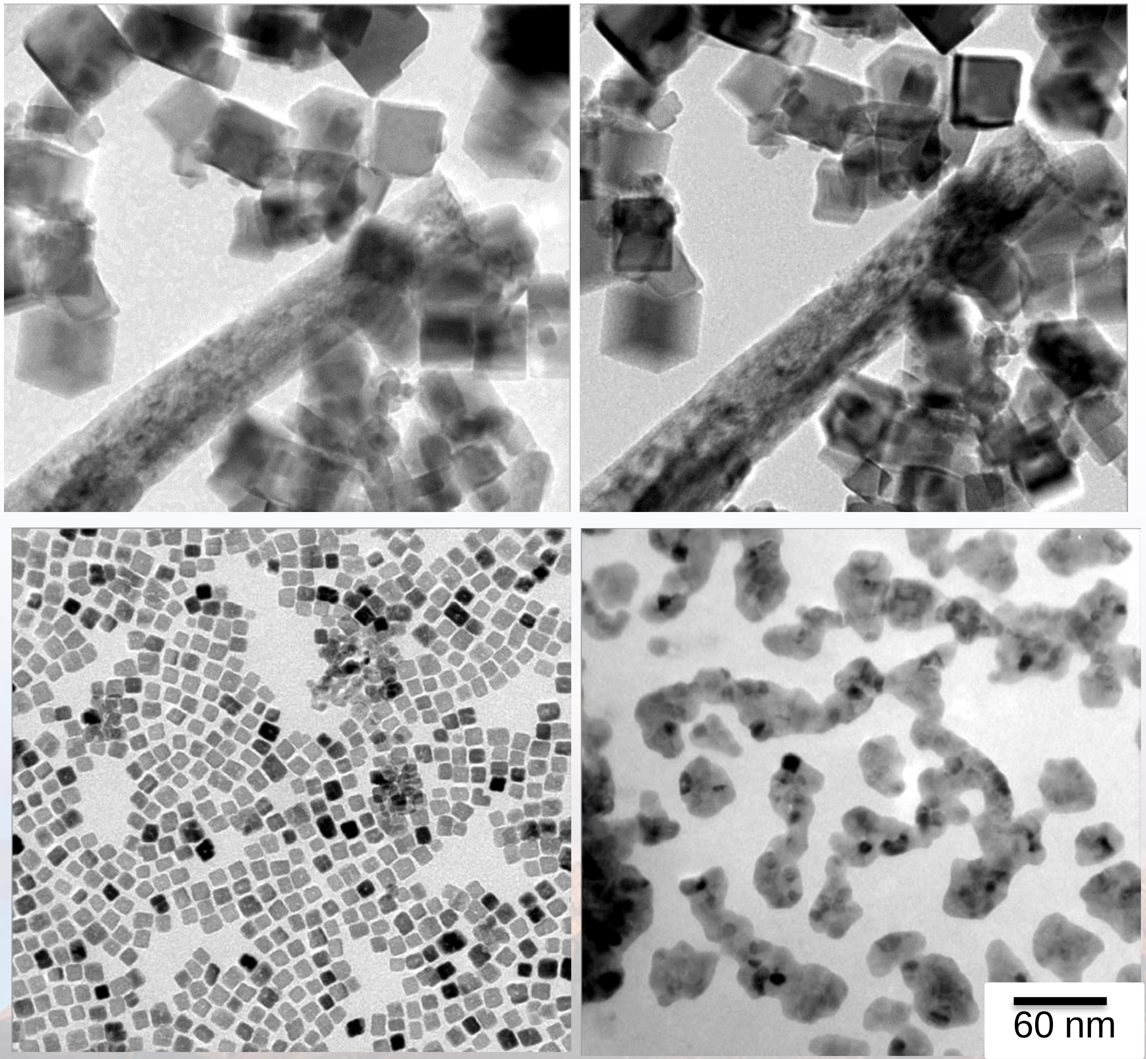
30 minutes



Details of Radiation Environment is Important!

Collaborators: S.A. Briggs, C. Taylor, J. Kolar, T. Boyle

Radiation Stability of CeO_x Nanoparticles



In situ

- 2.7 MeV Si^{+}, RT, 8.83E11 ions/cm^2s, 7.96E-5 dpa/s
- $CeCl_3$ irradiated for 6 mins, 2.87E-2 dpa
- $CeNO_3$ irradiated for 18 minutes 8.60E-2 dpa

Ex situ

- 2.7 MeV Si^{+}, RT, 2.41E11 ions/cm^2s, 2.2E-5 dpa/s
- Both irradiated for 66 mins to 8.7E-2 dpa

Difference between a broad DC beam vs. rastered beam is significant!

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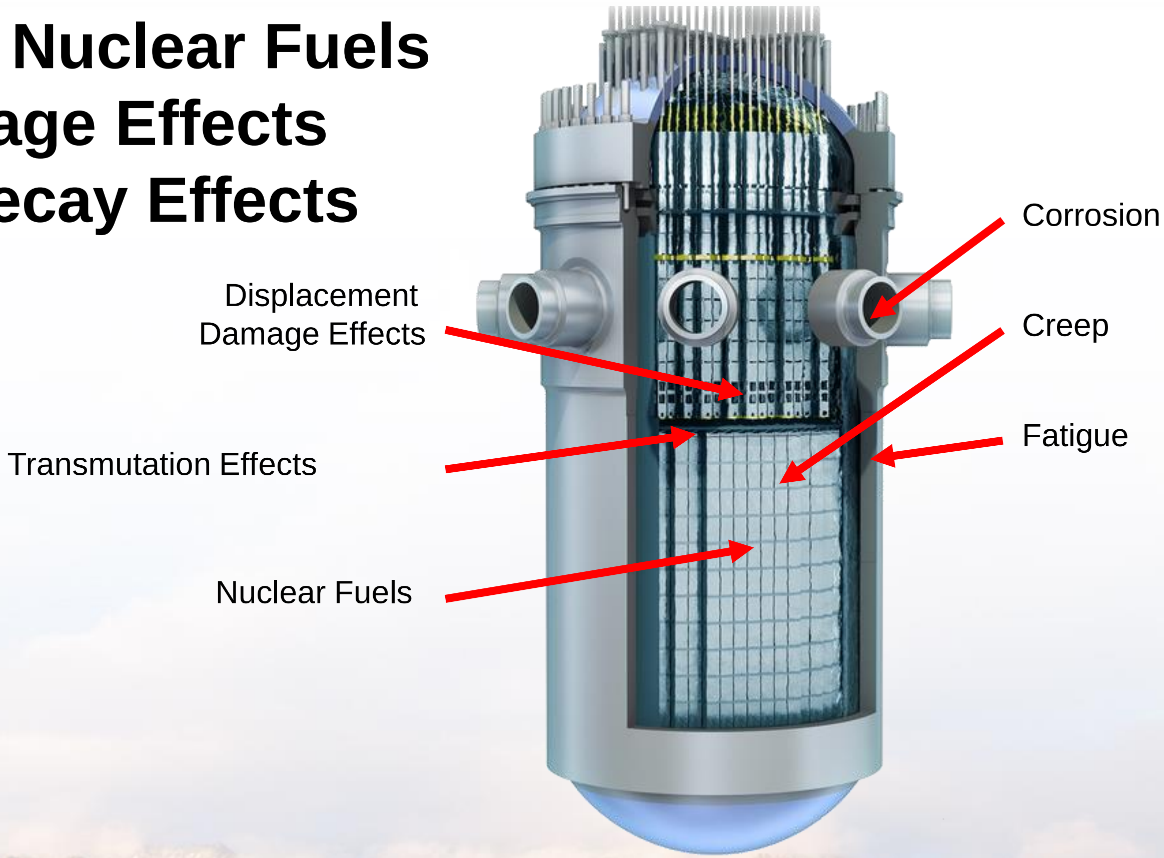

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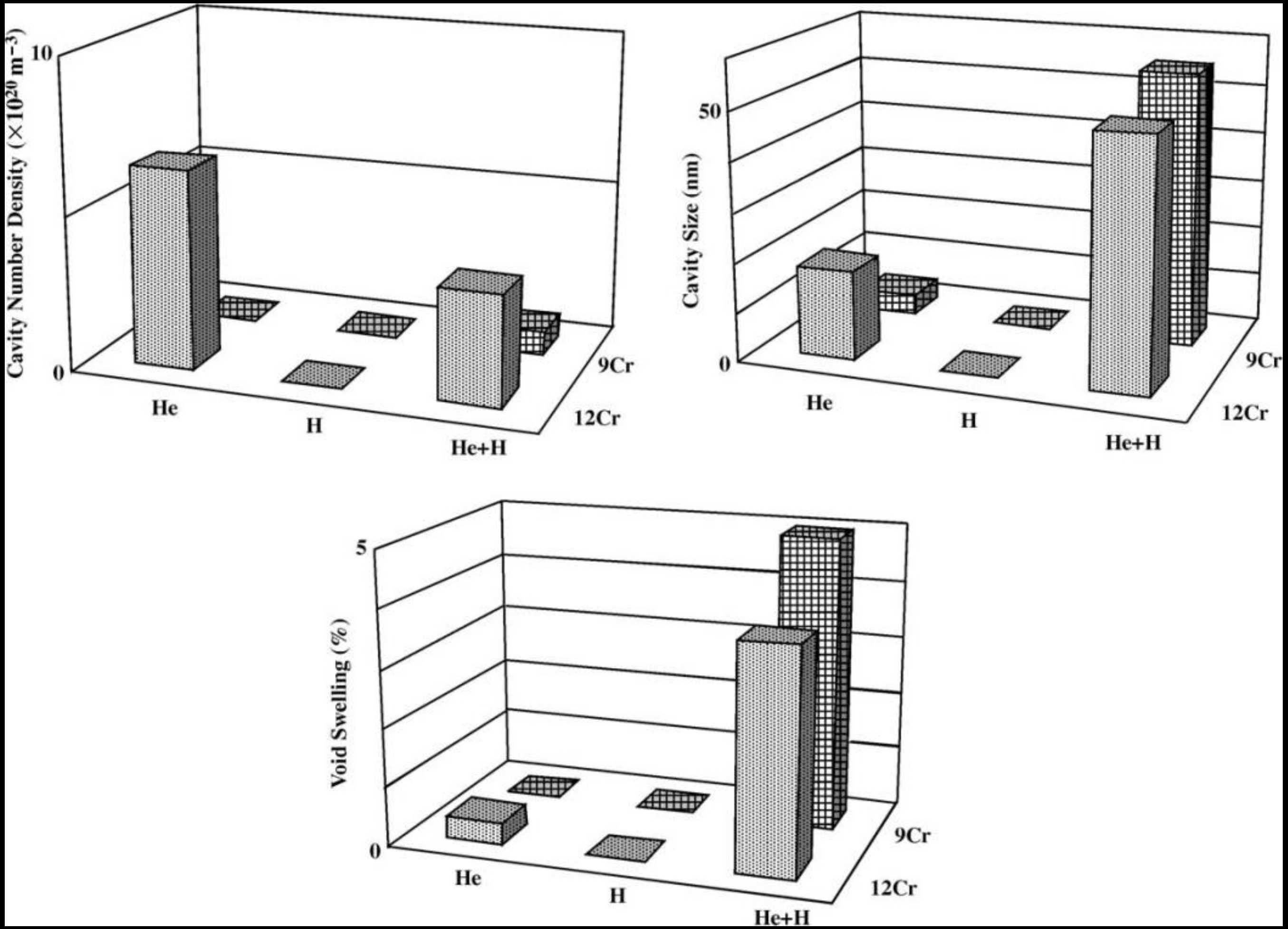
H, He, and Displacement Damage Synergy



T. Tanaka et al. “Synergistic effect of helium and hydrogen for deffect evolution under milt-ion irradiation of Fe-Cr ferritic alloys”

Coupling Effect

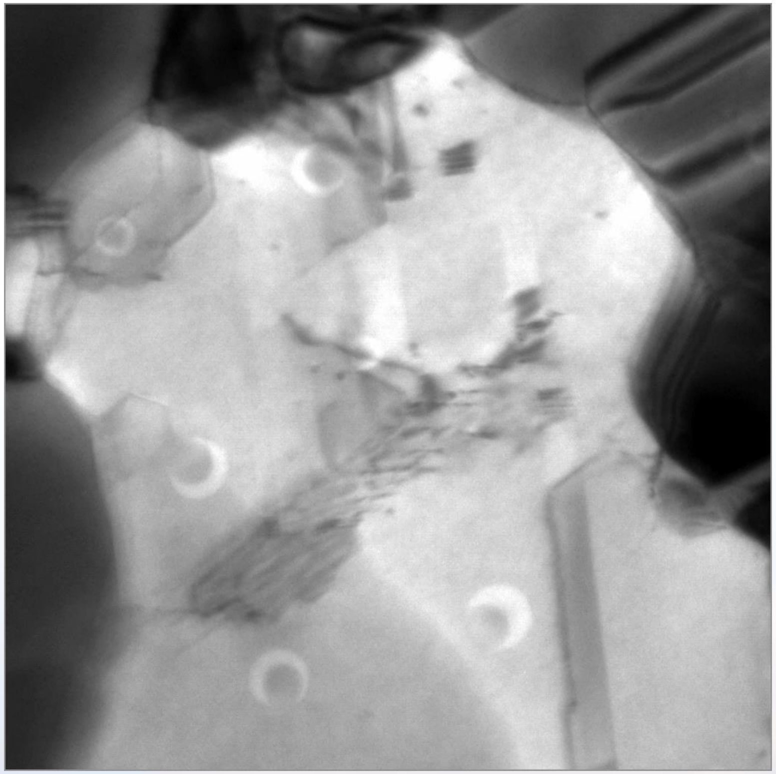
- H and He are produced as decay products
- The relationship between the point defects present, the interstitial hydrogen, and the He bubbles in the system that results in the increased void swelling has only been theorized.
- The mechanisms which governs the increased void swelling under the presence of He and H have never been experimental determined

No capability currently exist for triple beam irradiation in the U.S. and No capability for tripple beam TEM ion irradiation exists in the world

Dose Rate Effects in Nanocrystalline Metals

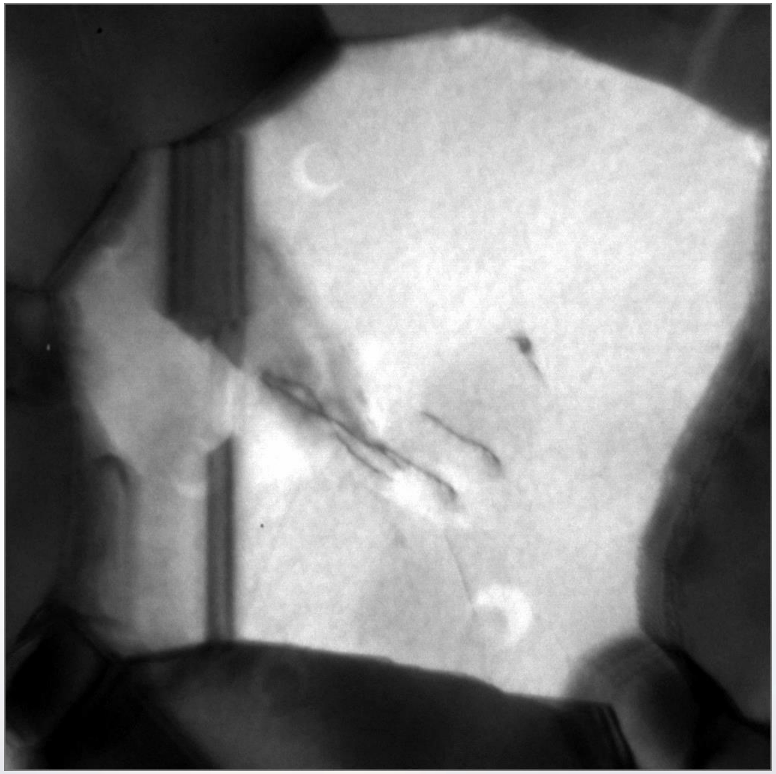
Collaborators: C. Chisholm , P. Hosemann, & A. Minor

7.9 x 10^9 ions/cm^2/s



VS

6.7 x 10^7 ions/cm^2/s

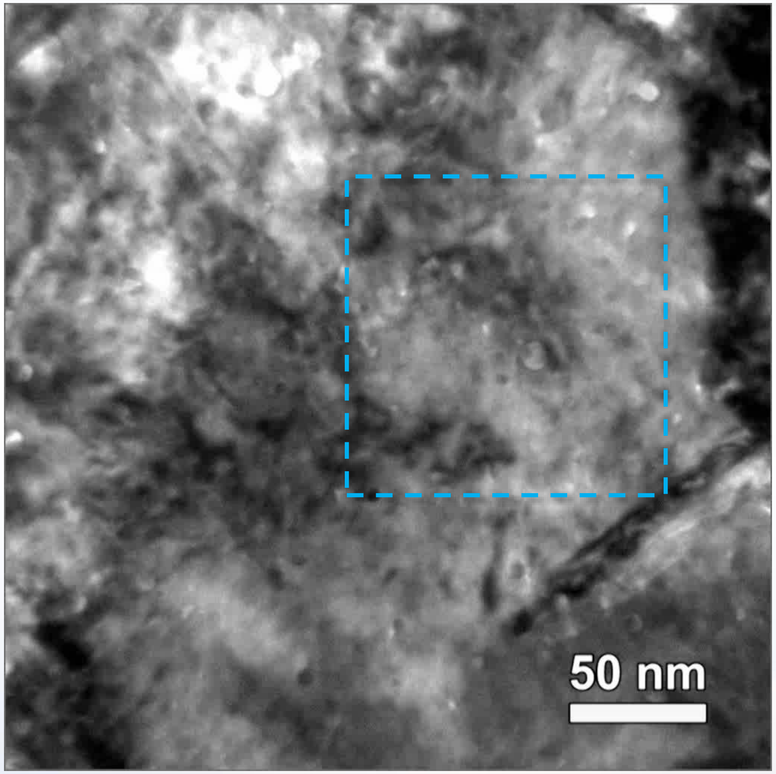


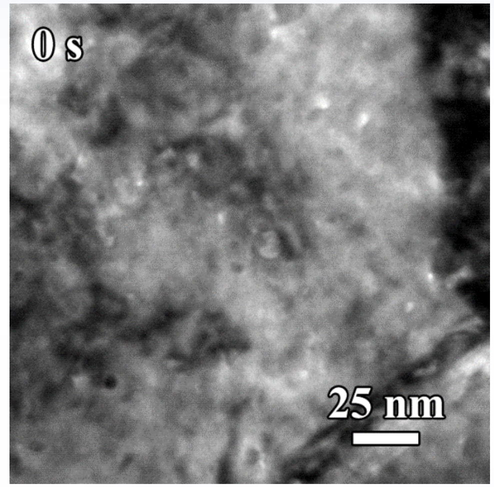
Improved vibrational and ion beam stability permits us to work at 120kx or higher permitting imaging of single cascade events

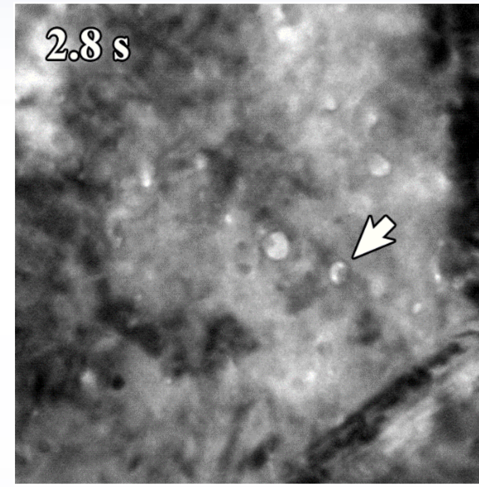
Simultaneous *In situ* TEM Triple Beam: 2.8 MeV Au^{4+} + 10 keV He^{+}/D_2^{+}

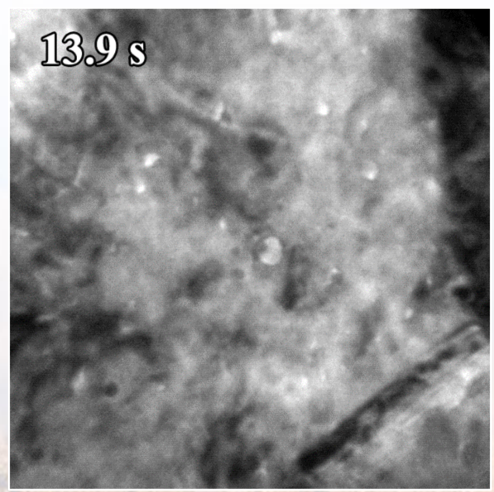
Collaborator: D.C. Bufford

Video playback speed x1.5.







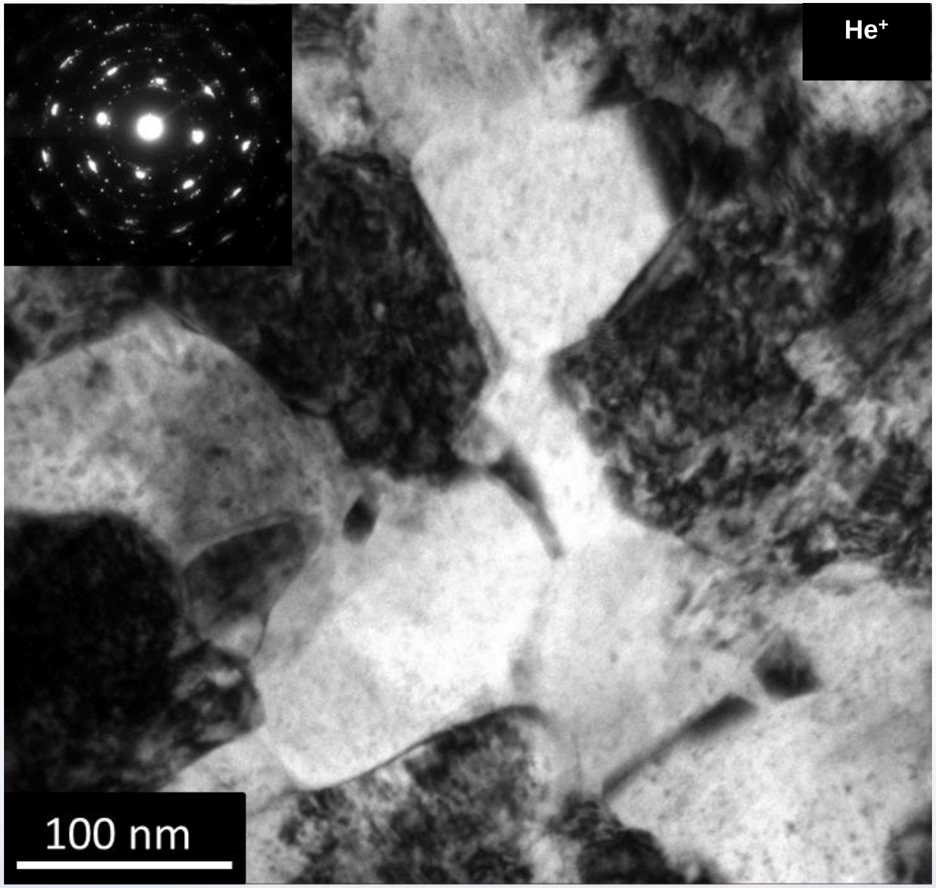


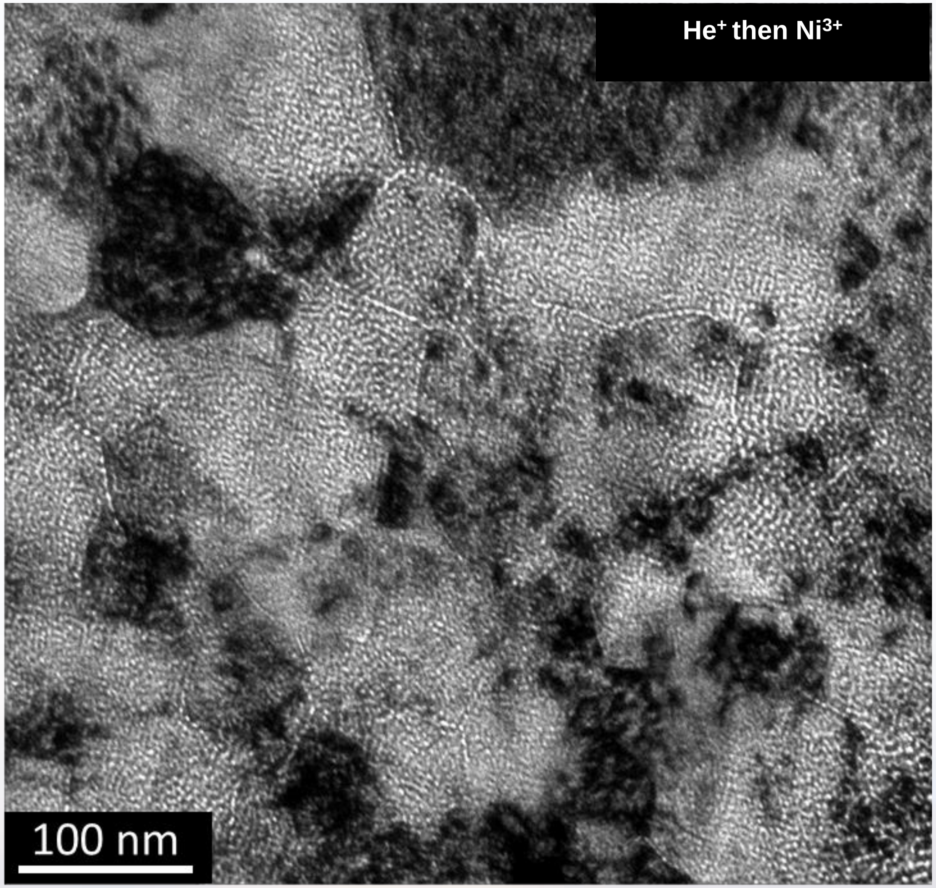
- **Approximate fluence:**
  - Au 1.2 $\times 10^{13}$ ions/cm^2
  - He 1.3 $\times 10^{15}$ ions/cm^2
  - D 2.2 $\times 10^{15}$ ions/cm^2
- **Cavity nucleation and disappearance**

In-situ triple beam He, D_2, and Au beam irradiation has been demonstrated on Sandia's I^3TEM! Intensive work is still needed to understand the defect structure evolution that has been observed.

Heterogeneous Bubble Formation under Some Radiation Environments

Collaborator: B. Muntifering & J. Qu





10^{17} He^+/cm^2

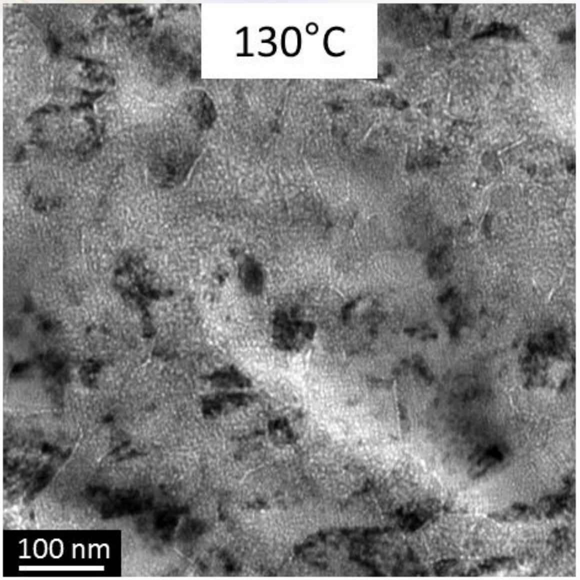
Visible damage to the sample

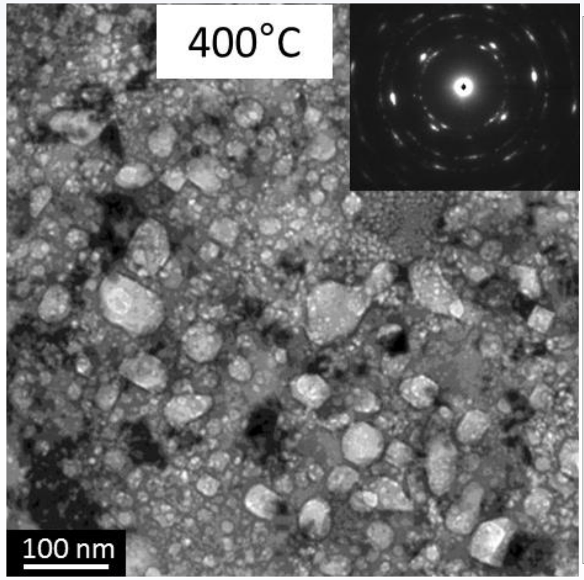
0.7 dpa Ni^{3+} irradiation

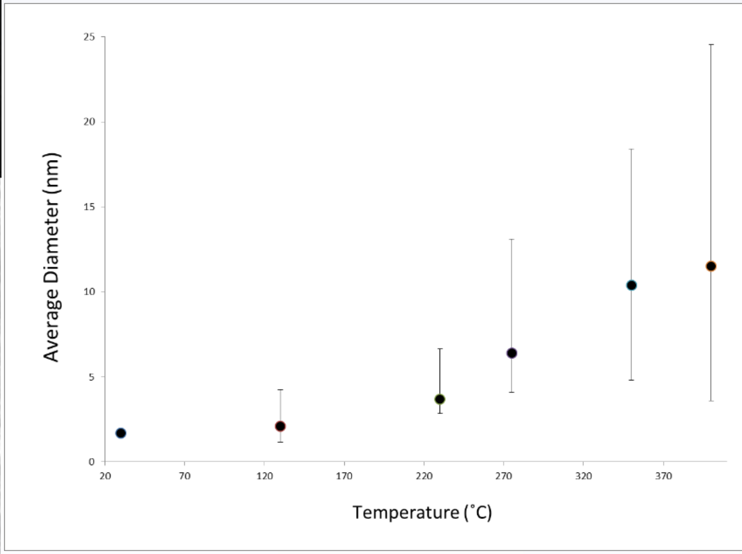
High concentration of cavities along grain boundaries

Sandia National Laboratories

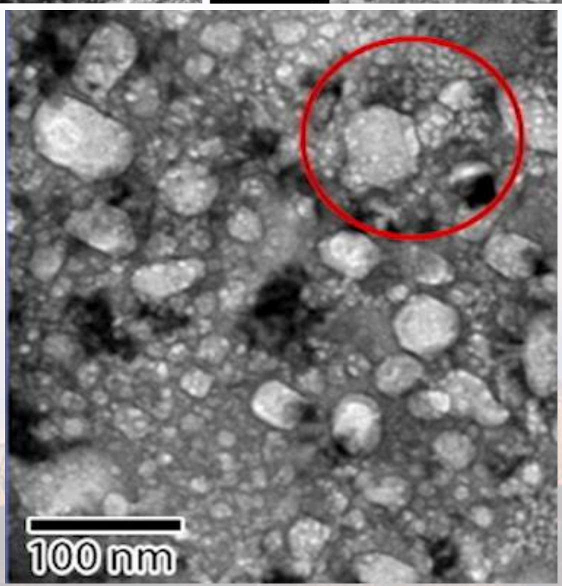
Cavity Growth during In-situ Annealing of 10 keV He^{+} Implanted and then 3 MeV Irradiated Ni^{3+}

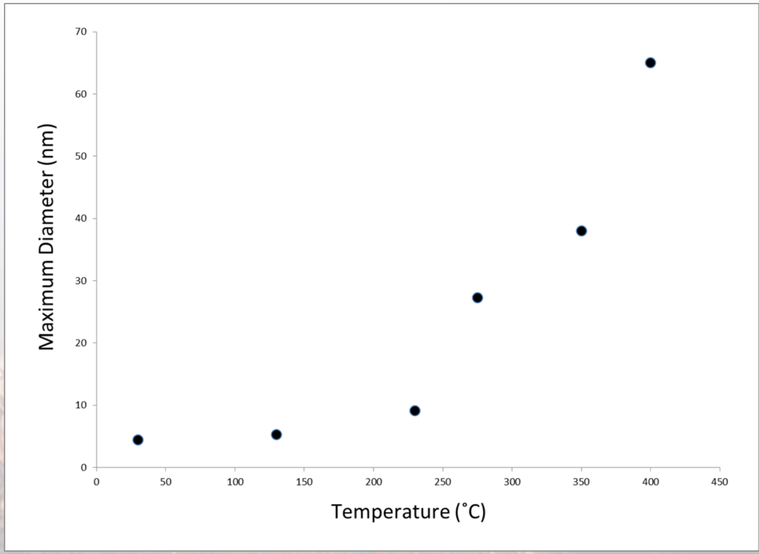






Bubble to cavity transition and cavity evolution can be directly studied

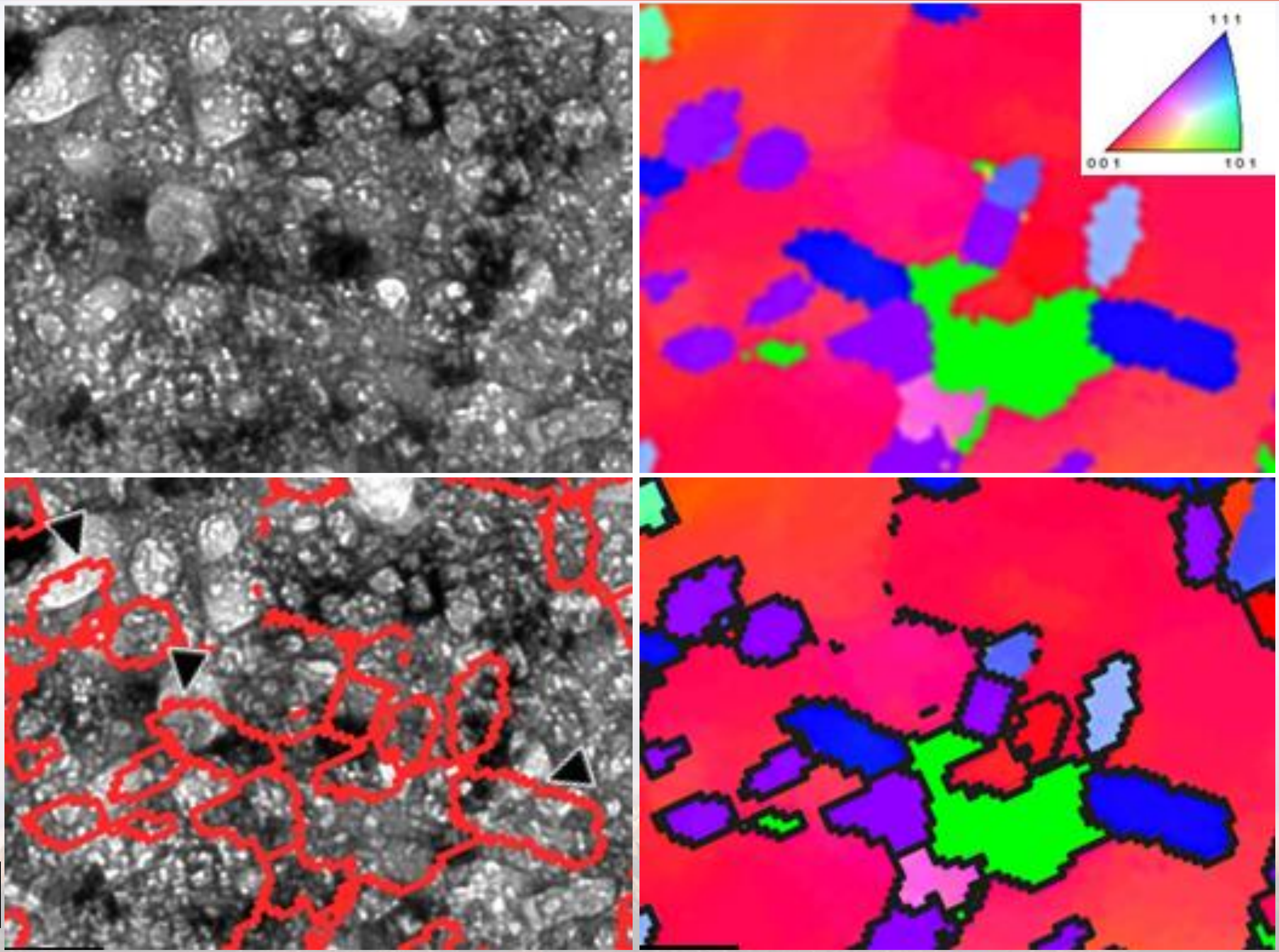




Precession Electron Diffraction Reveals Hidden Grain Structure

Cavities in helium implanted, self-ion irradiated,

nc nickel film annealed to 400 ˚C

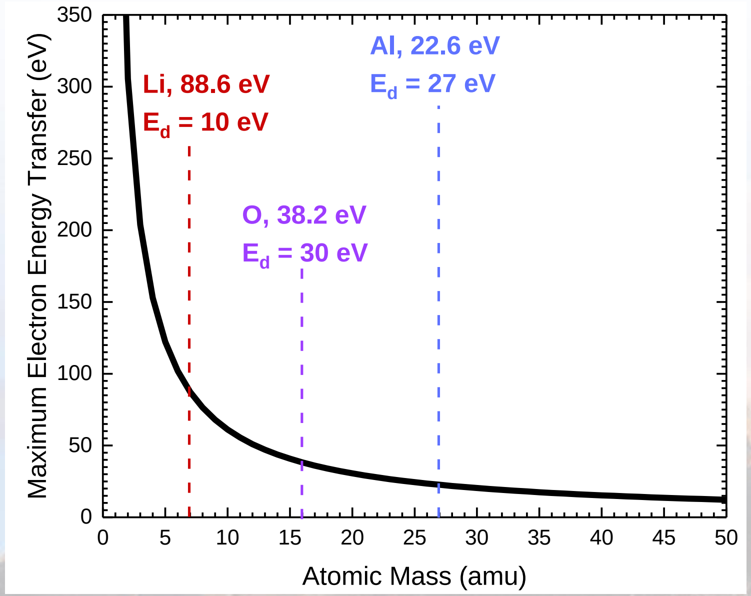


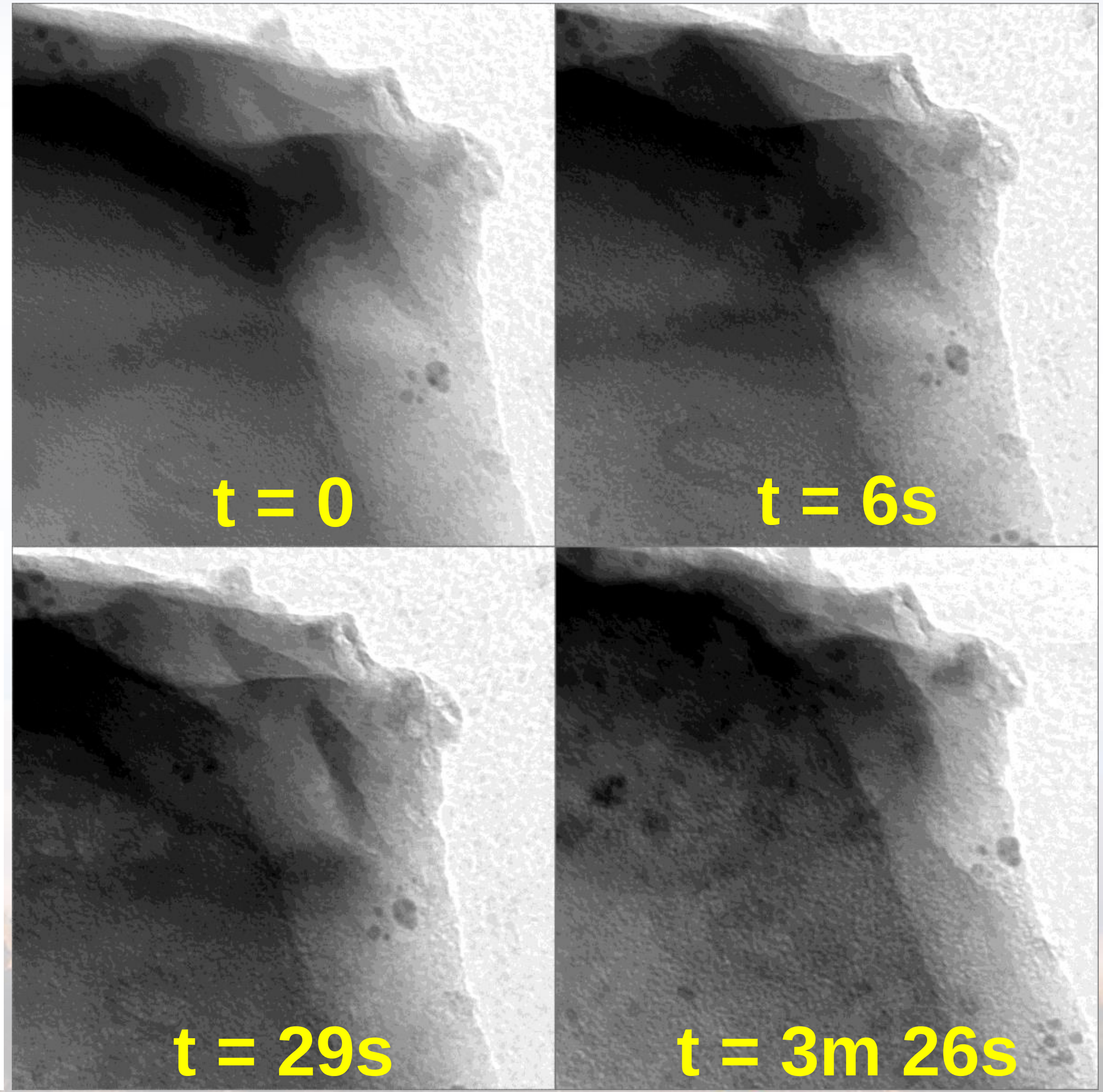
Cavities span multiple grains at identified grain boundaries

100 nm

Electron beam induced void growth

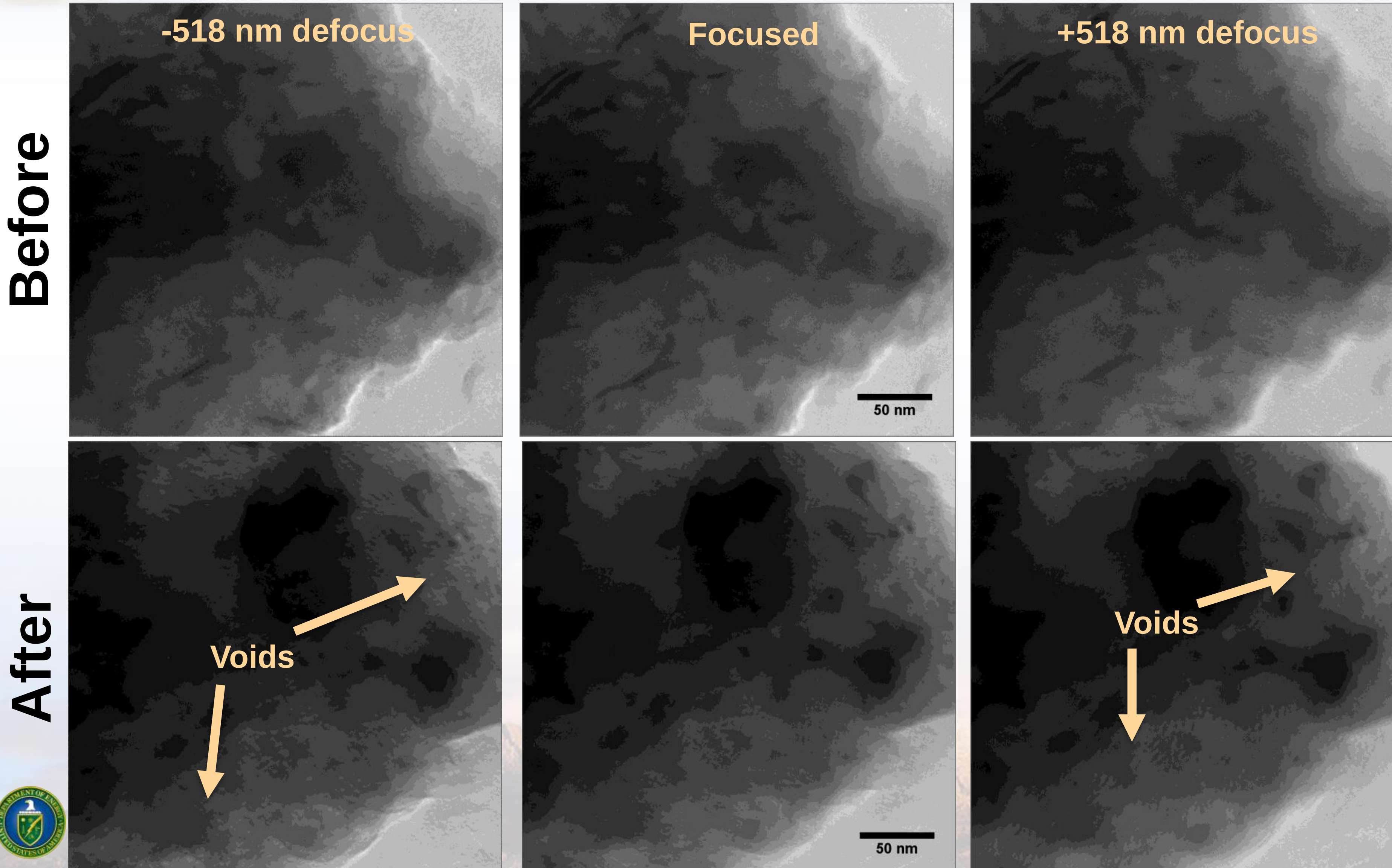
- Voids were observed to form under the electron beam in several particles
- Rate of void formation is not consistent between particles
- Possibly due to electron beam displacing Li and O atoms





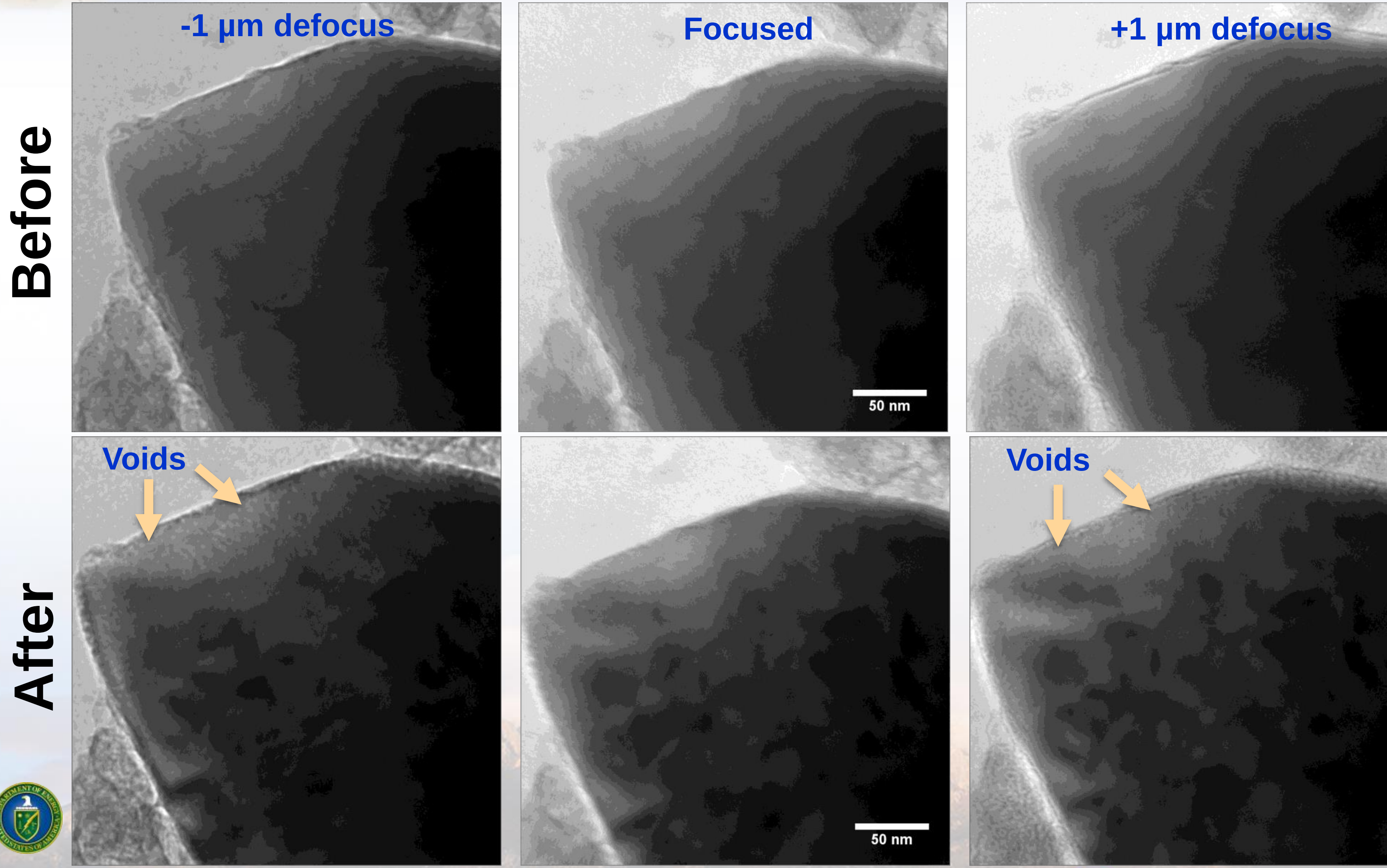
In-situ He implantation @ 310°C

Bubbles formed after ~13 min (1.5×10^{17} He/cm^2)



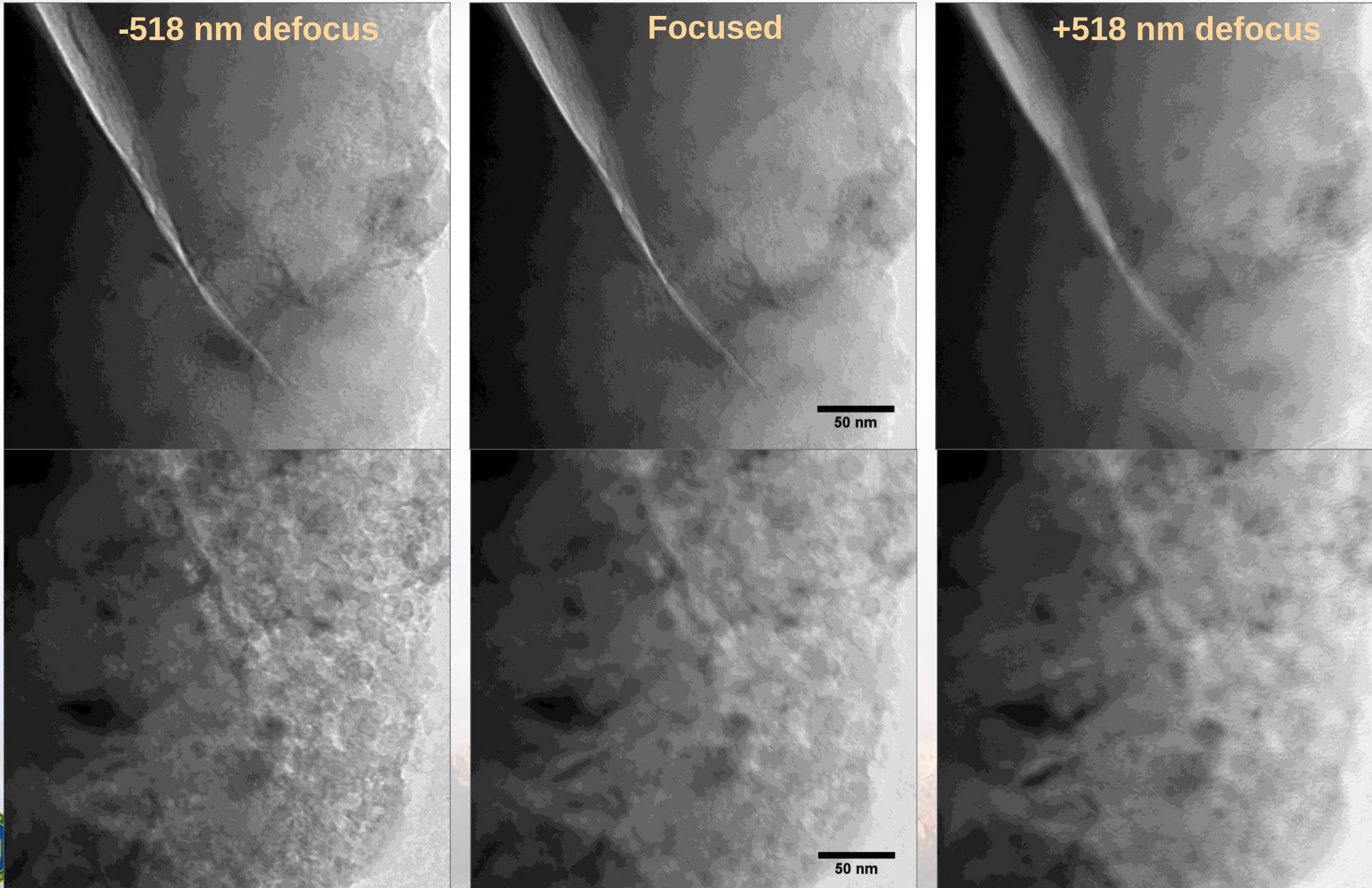
In-situ He + D irradiation @ 310°C

Bubbles formed after ~60 min (1.7×10^{17} He/cm^2, 3.4×10^{17} D/cm^2)



In-situ He + D + Au @ 310°C

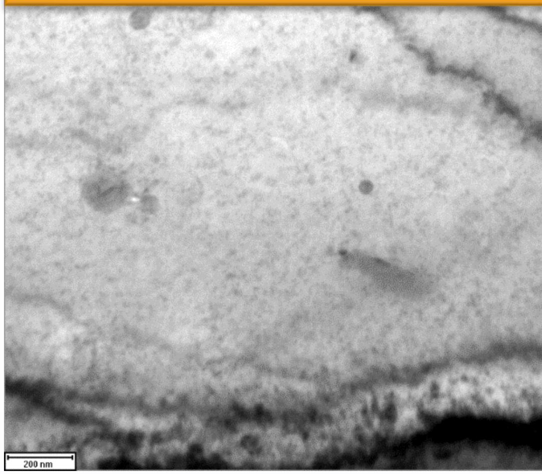
No single eucentric: Drastic increase in surface roughness!

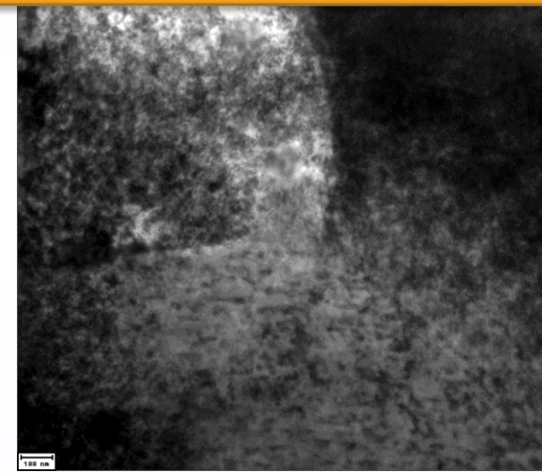


Comparison between $LiAlO_2$ and Zr-4

Zr Alloys

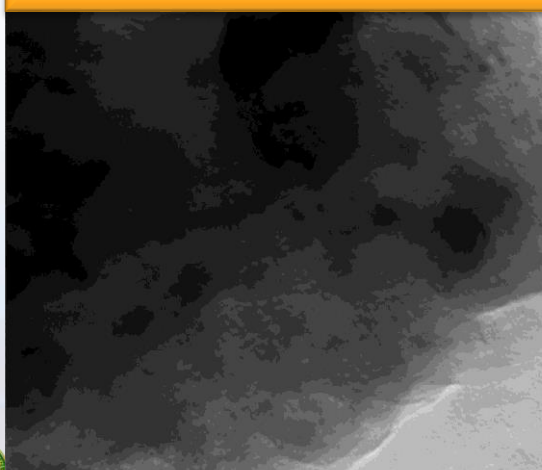
He accumulation → Damage + He → Damage + He + ^{3}H

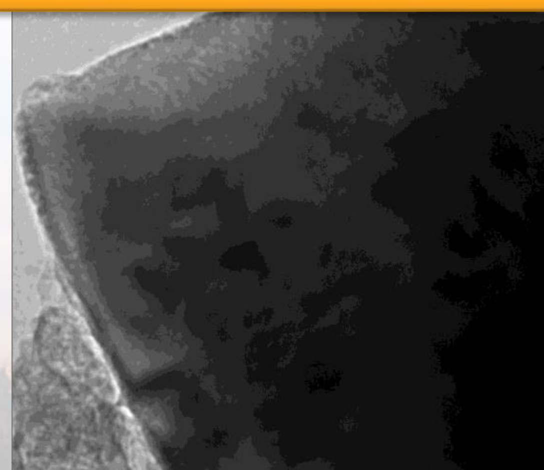


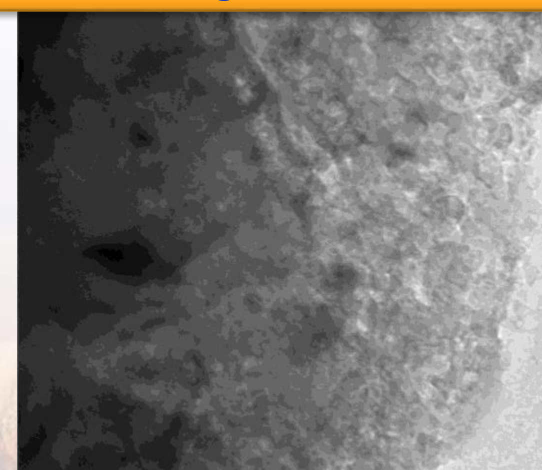


$LiAlO_2$

He accumulation → ^{3}H + He → Damage + He + ^{3}H







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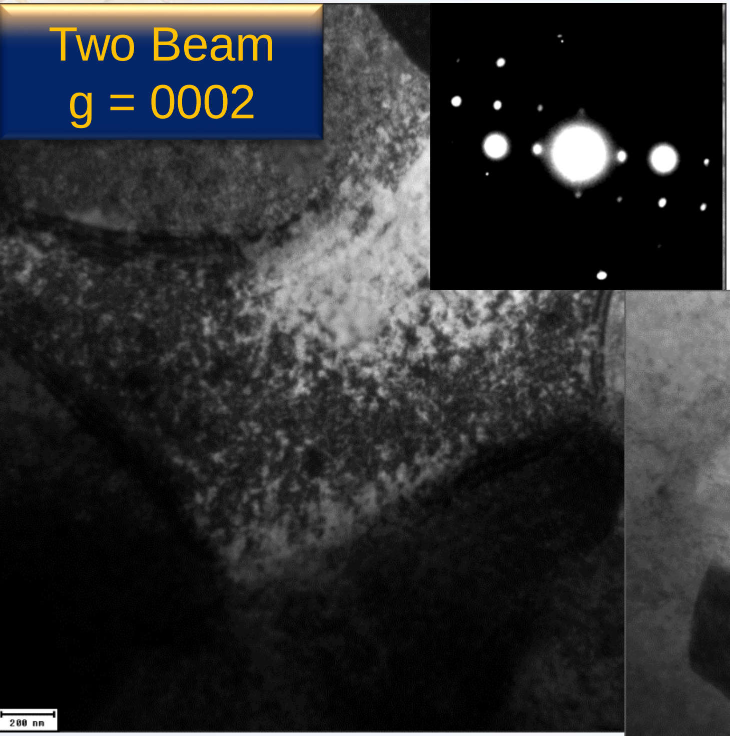
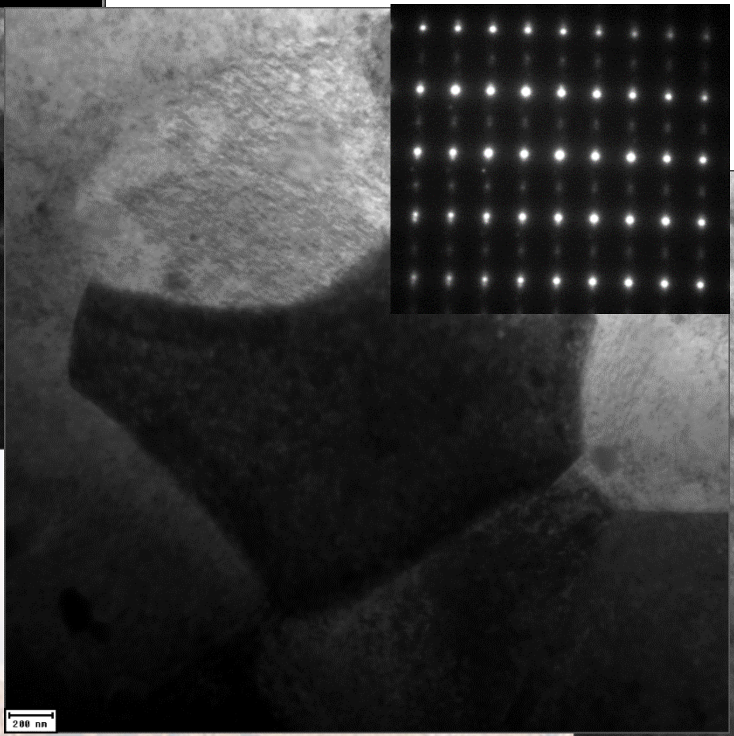
10 keV He^{+} Implantation
Then 3 MeV Self Ion Irradiation into Zr-4 at 310 C

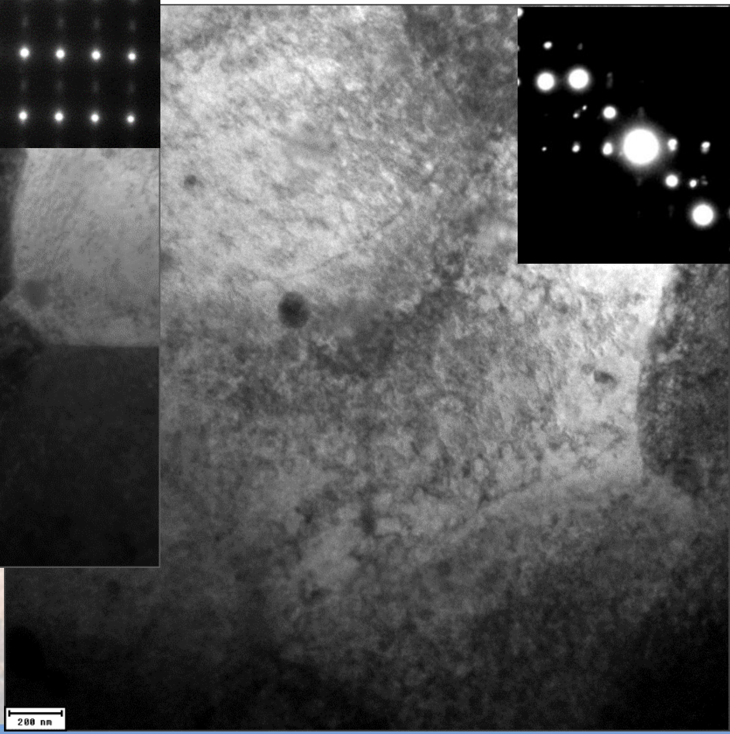
Two Beam
g = 0002



On Axis

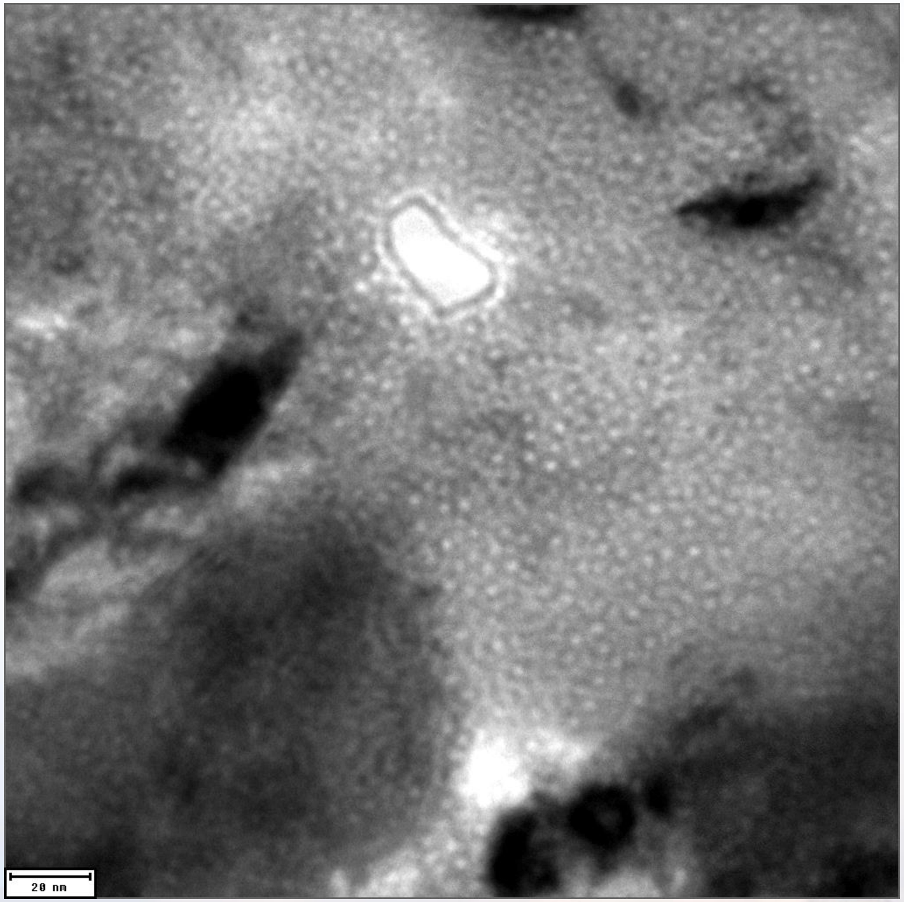


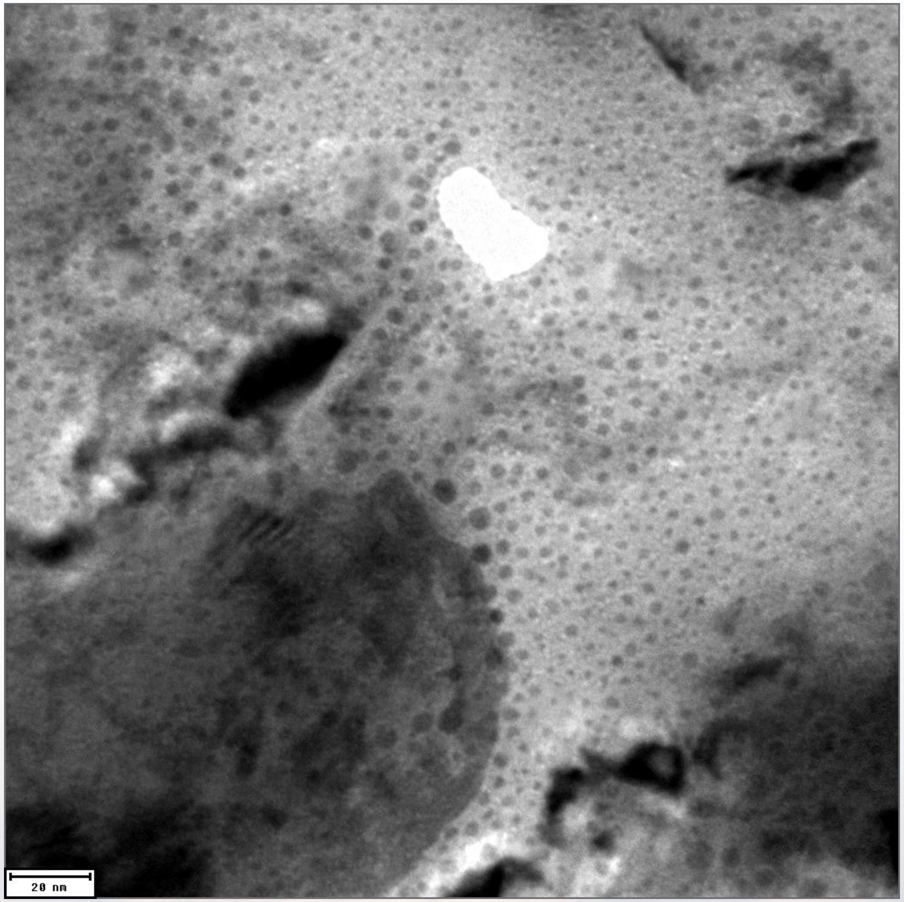
Two Beam
g = 01-12



Sequence maters, as well as imaging condition!

PIE: Through Focus Images
30 Days After the *In situ* Experiment





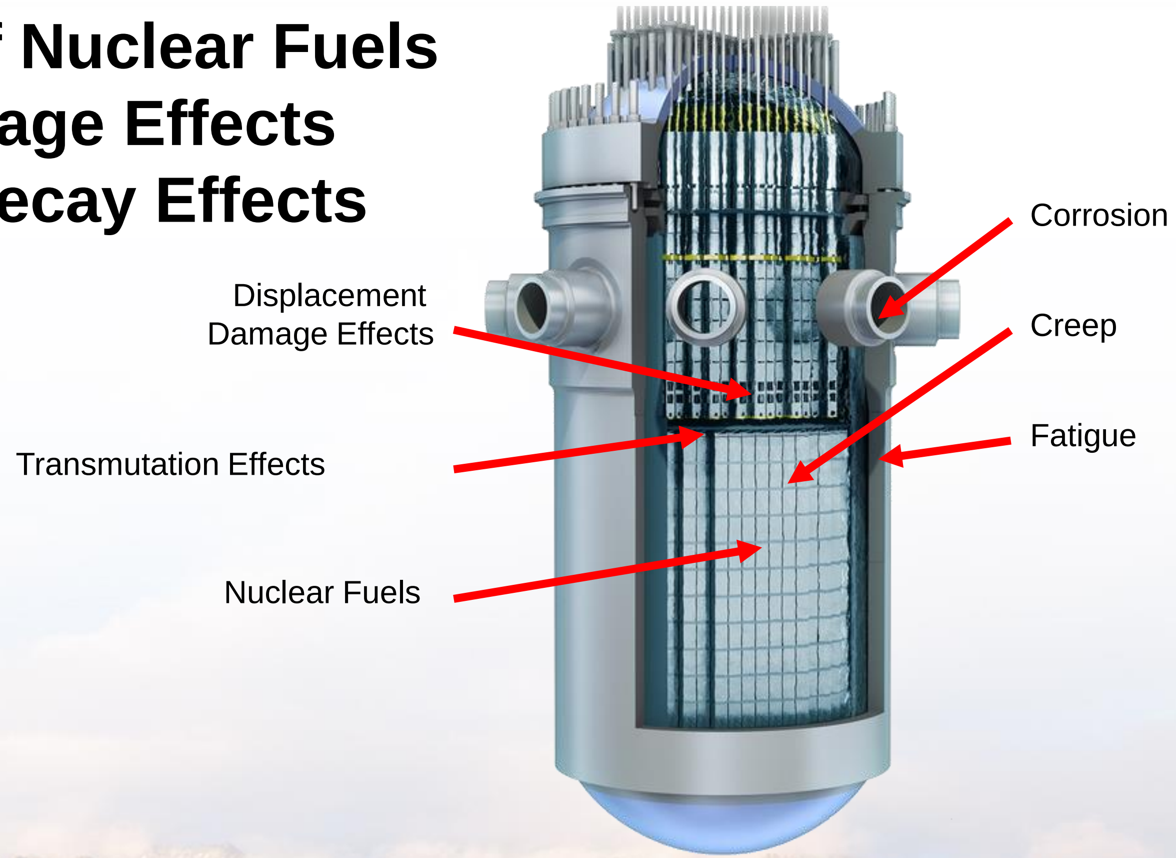
Under Focus

Over Focus

The end of accelerated aging experiment is not the end of the materials aging

Outline

1. Characterization of Nuclear Fuels
2. Displacement Damage Effects
3. Transmutation & Decay Effects
4. Corrosion
5. Creep
6. Fatigue

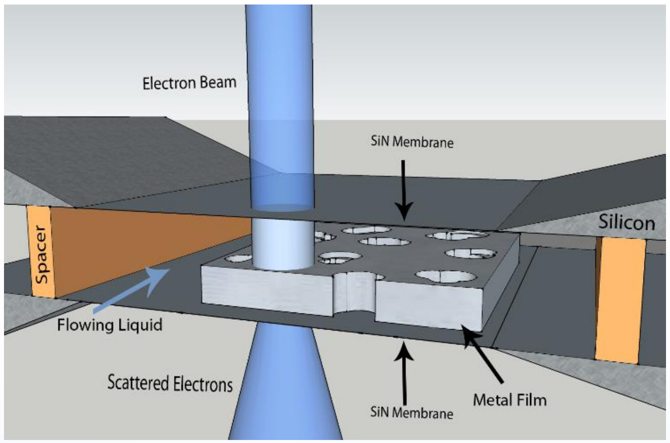


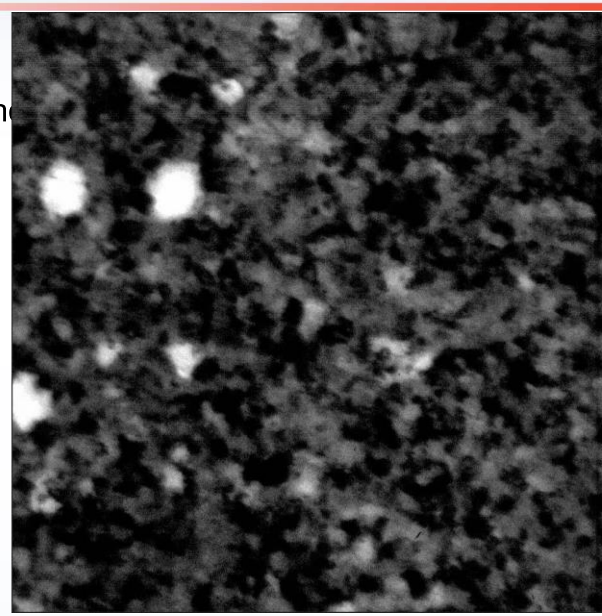
Can We Gain Insight into the Corrosion Process through *In situ* TEM?

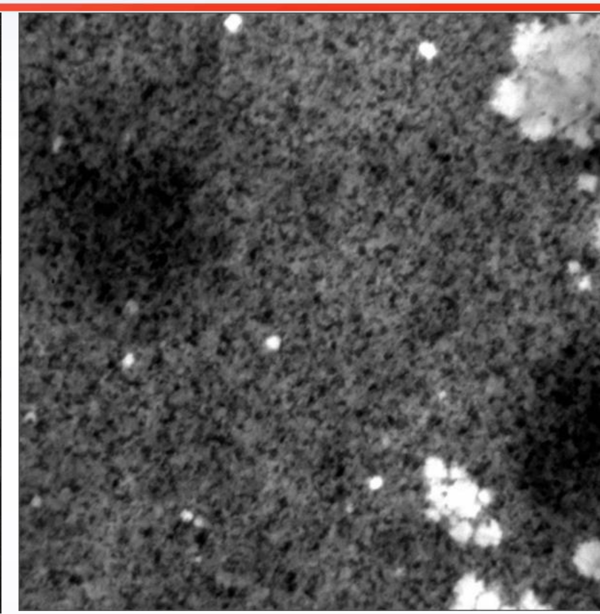
Contributors: D. Gross, J. Kacher, I.M. Robertson & Protochips, Inc.

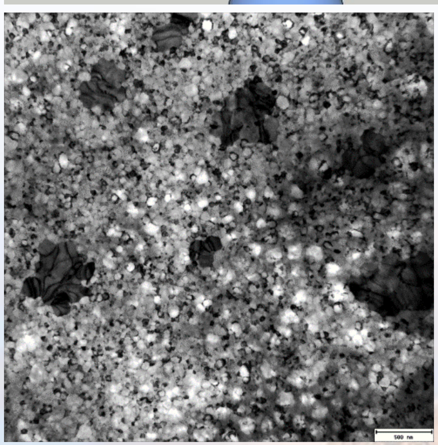
Microfluidic Stage

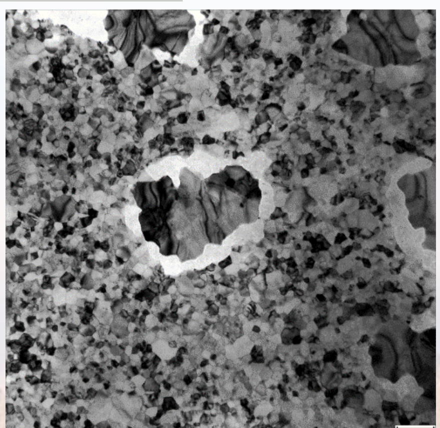
- Mixing of two or more channels
- Continuous observation of the reaction chann

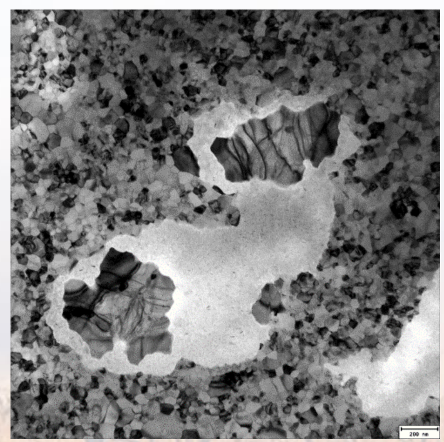


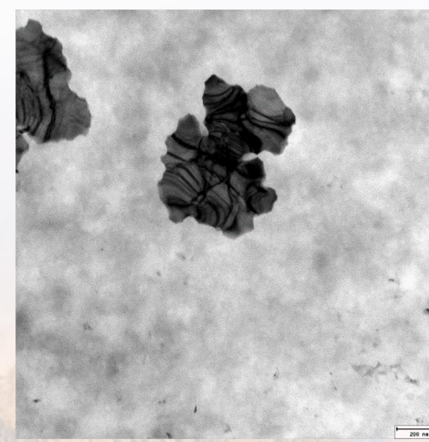












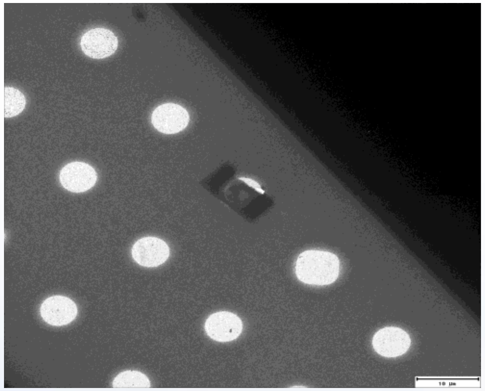
Pitting mechanisms during dilute flow of acetic acid over 99.95% nc-PLD Fe involves many grains.
Large grains resulting from annealing appear more corrosion tolerant

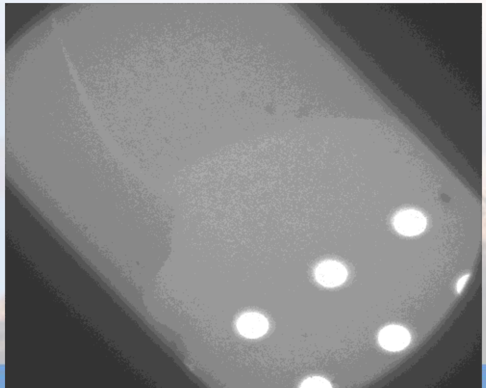
Feasibility of Studying Zircaloy 2 at Nominally 1 atm

Collaborators: S. Rajasekhara and B.G. Clark

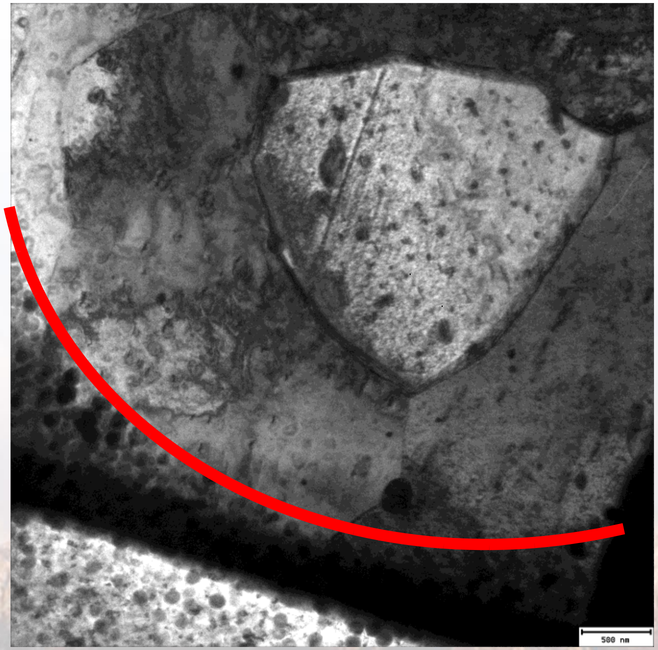
Vapor-Phase Heating TEM Stage

- Compatible with a range of gases
- *In situ* resistive heating
- Continuous observation of the reaction channel
- Chamber dimensions are controllable
- Compatible with MS and other analytical tools

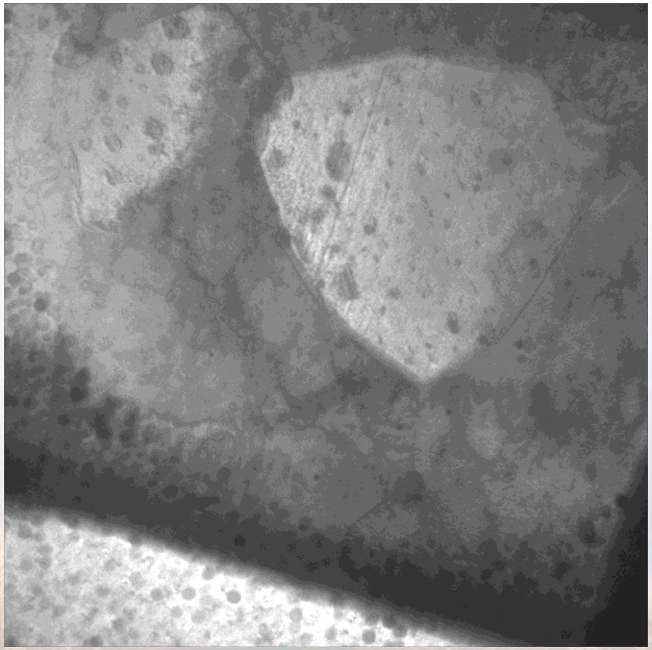




Vacuum & Single Window



Nominally 1 atm H_2 & Two Windows



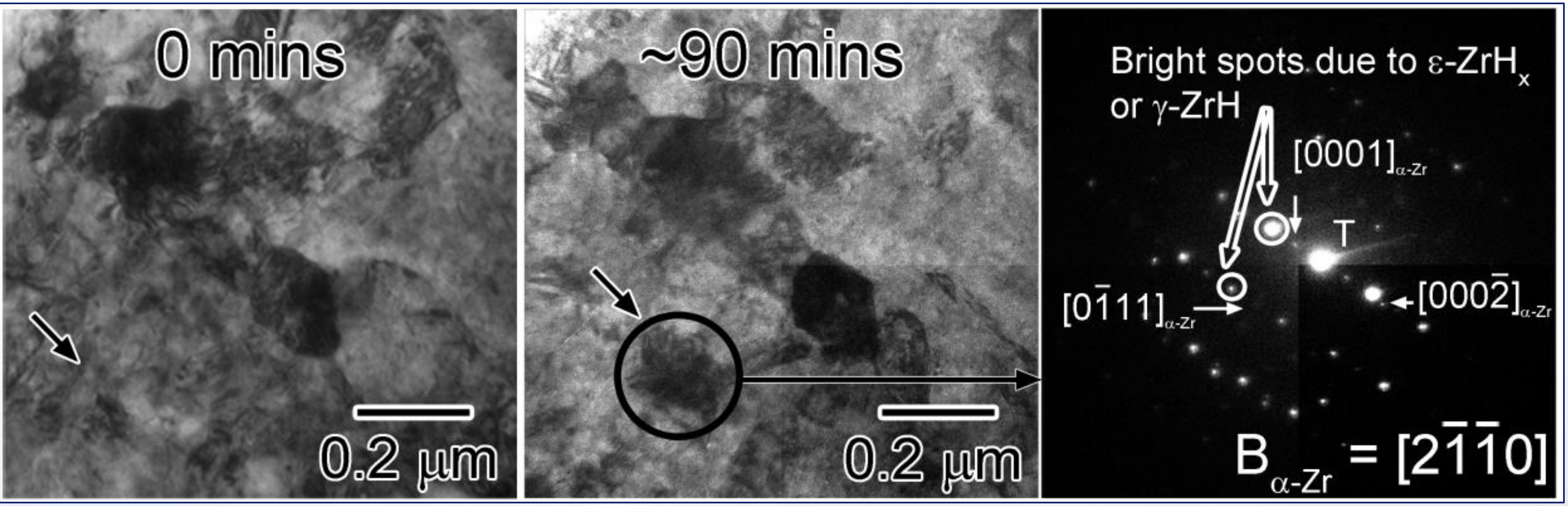
Most features are observed in both despite the decreased resolution resulting from the additional SiN window and 5 μm of air

In situ Observation of Hydride Formation in Zirlo

Collaborators: S. Rajasekhara and B.G. Clark

Absolute hydrogen pressure: 327 torr (~ 0.5 atm),

Ramp rate: 1 ^{o}C/s, Final temperature: ~ 400 C, Dwell time: ~ 90 mins



Hydride formation shown, for the first time by use of a novel TEM gas-cell stage, at elevated temperature and hydrogen pressure

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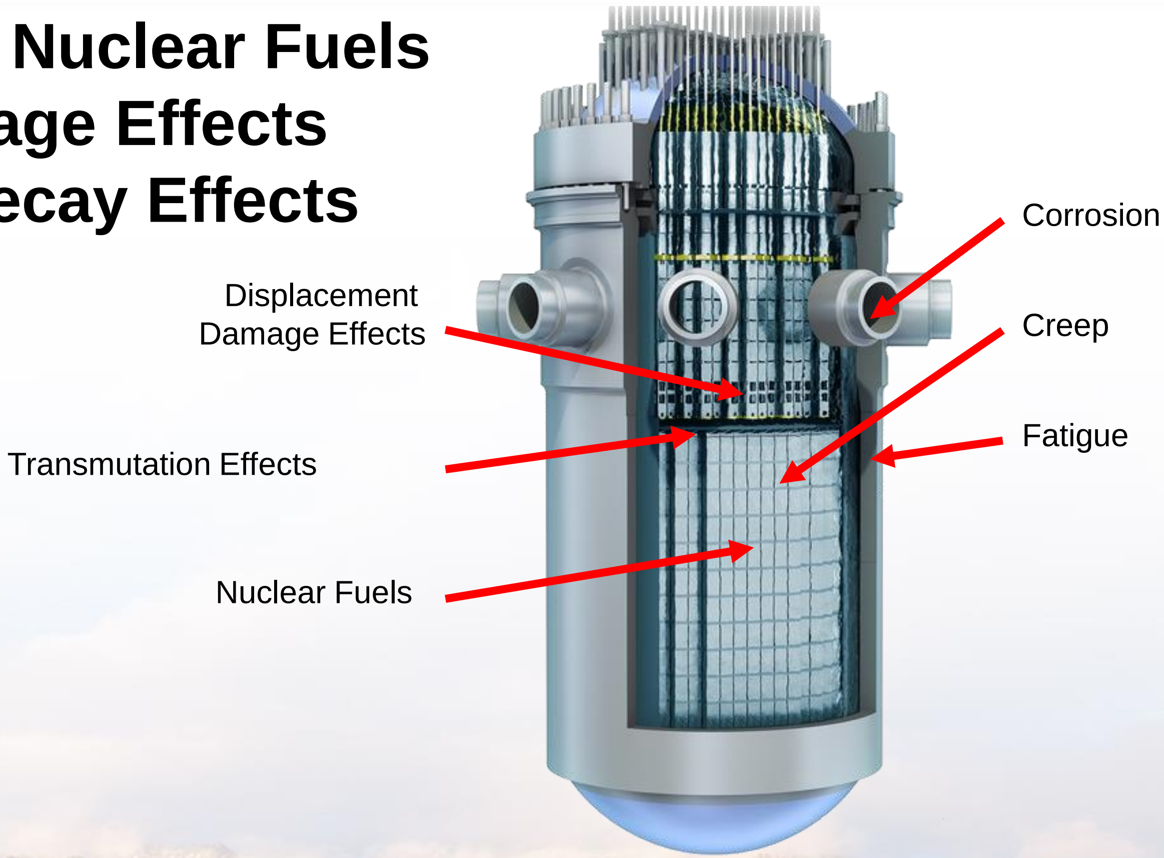


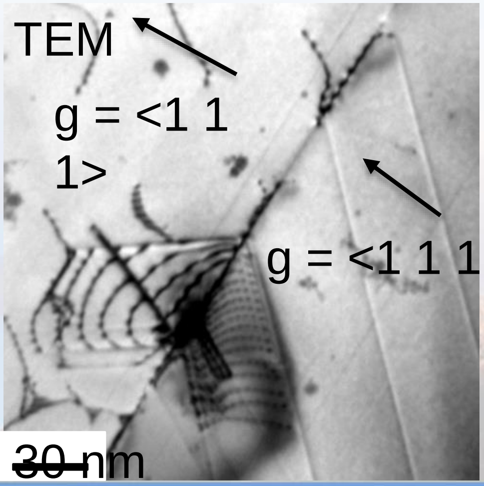
In situ Mechanical Testing

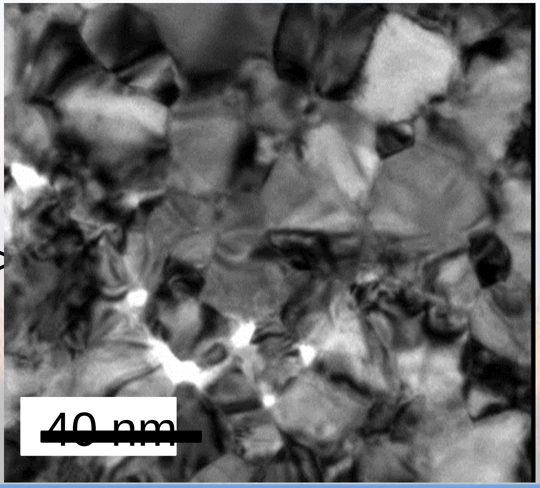
Qualitative "Bulk" Mechanical Testing

Minimal control over displacement and no "out-of-box" force information

- Successful in studies in observing dislocation-GB interactions/mechanisms
- Ideally both grains have kinematic BF 2-beam conditions: challenging in ST holder

Traditional Gatan Heating and Straining Holder





Quantitative Mechanical Testing

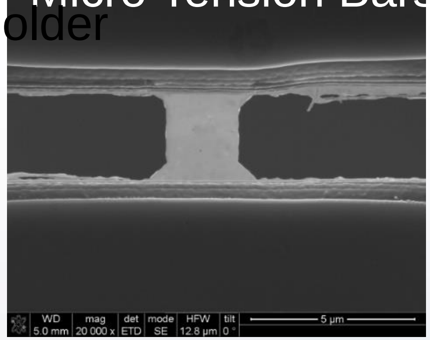
Minimal control over displacement and no "out-of-box" force information

- Sub nanometer displacement resolution
- Quantitative force information with µN resolution

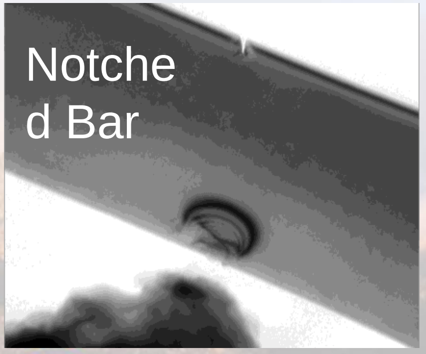
Hysitron PI-95 Holder

1) Indentation
2) Tension
3) Fatigue
4) Creep
5) Compression
6) Bend

Micro Tension Bars

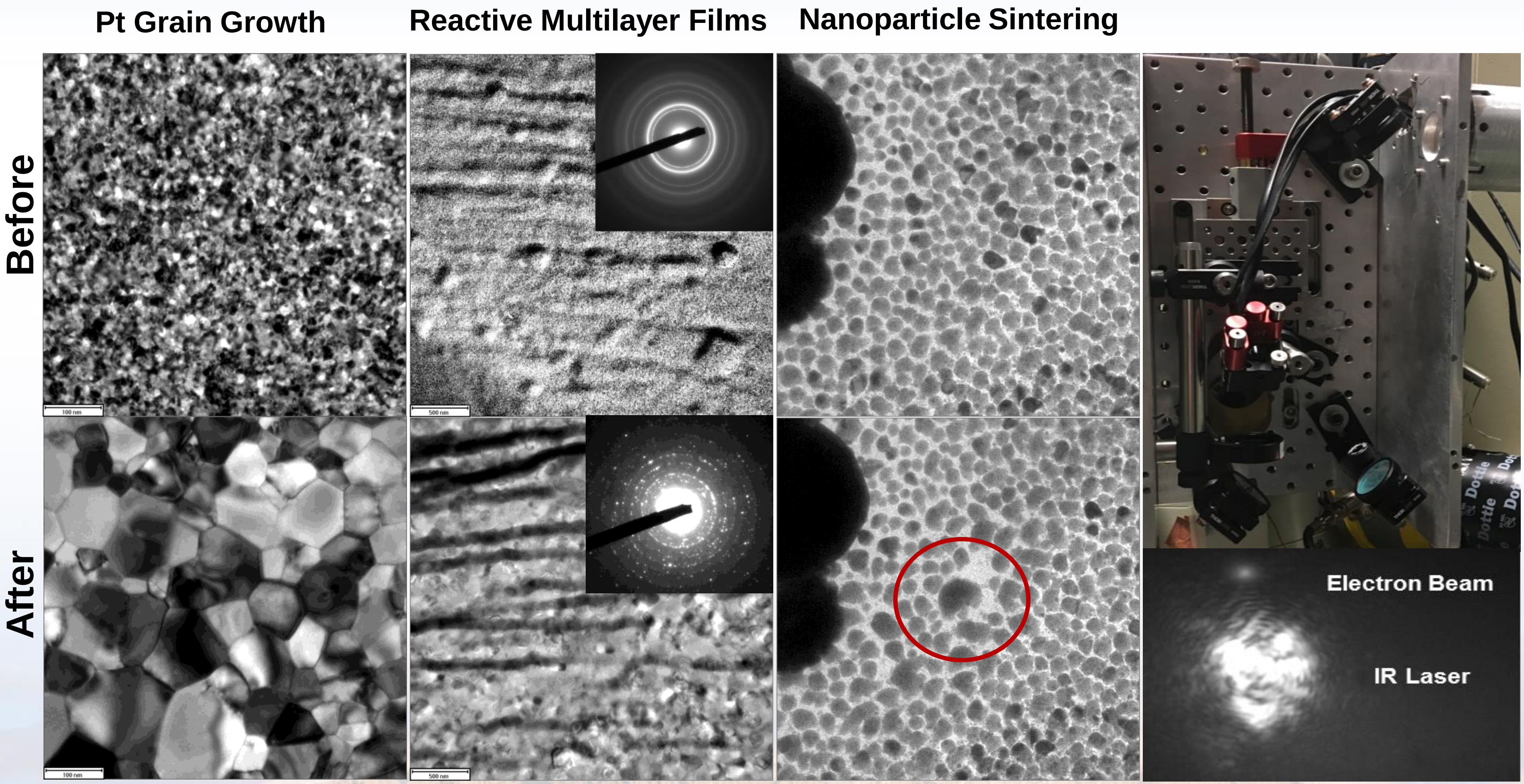


Notched Bar



Laser Initiation of Multilayer Reactive Foils

Collaborator: P. Price, C.M. Barr, D. Adams, M. Abere

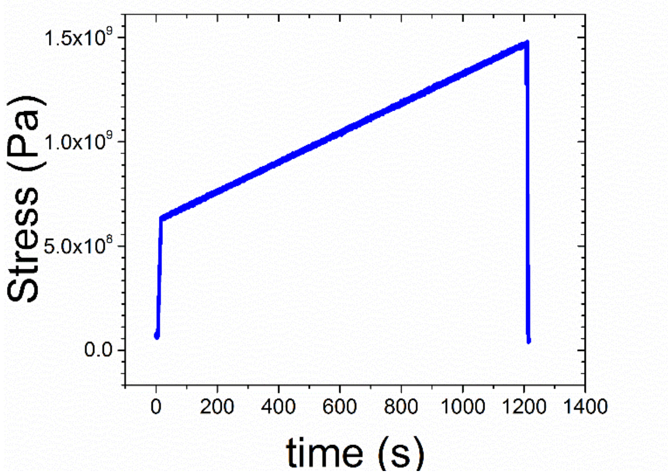


Understanding materials in extreme thermal environments as well as additive manufacturing processes

Irradiation Creep (4 MeV Cu^{3+} 10^{-2} DPA/s)

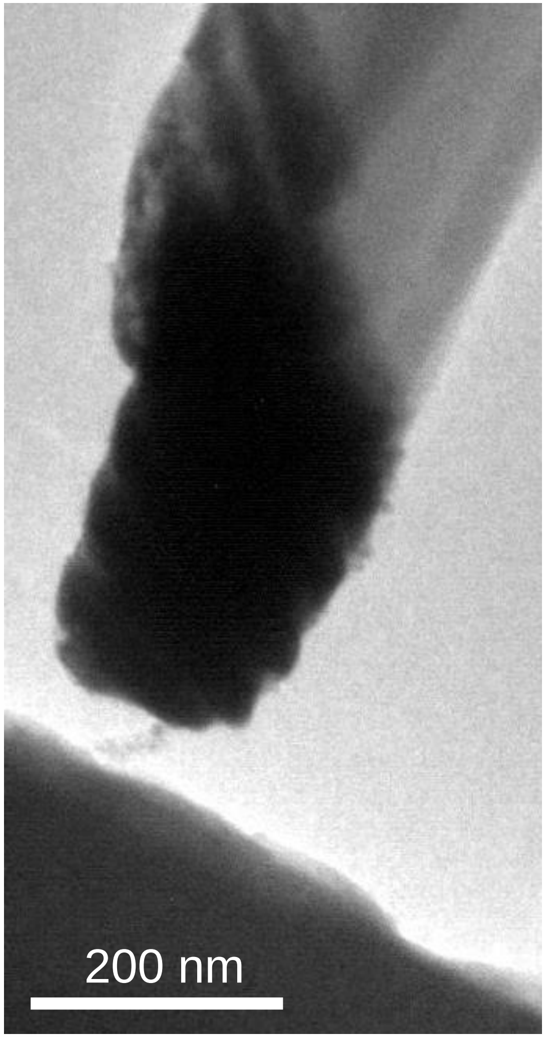
Contributors: S. Dillon & R.S. Averback

Controlled Loading Rate Experiments

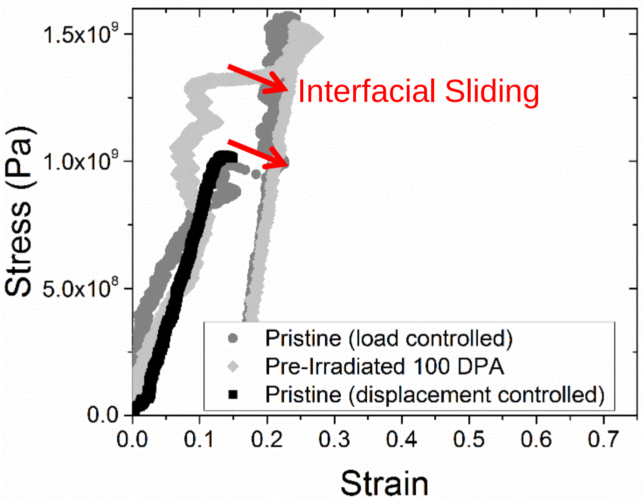


In-situ TEM radiation creep is feasible!

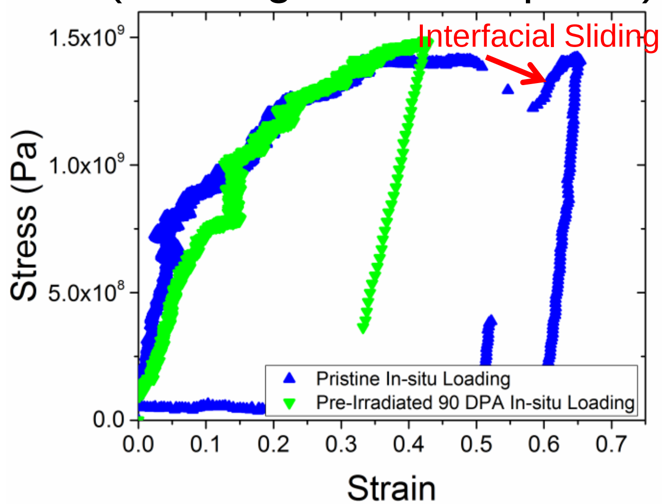
50 nm Cu-W multilayer
20 Min



No Irradiation
(Loading rate 0.6 Mpa s^{-1})

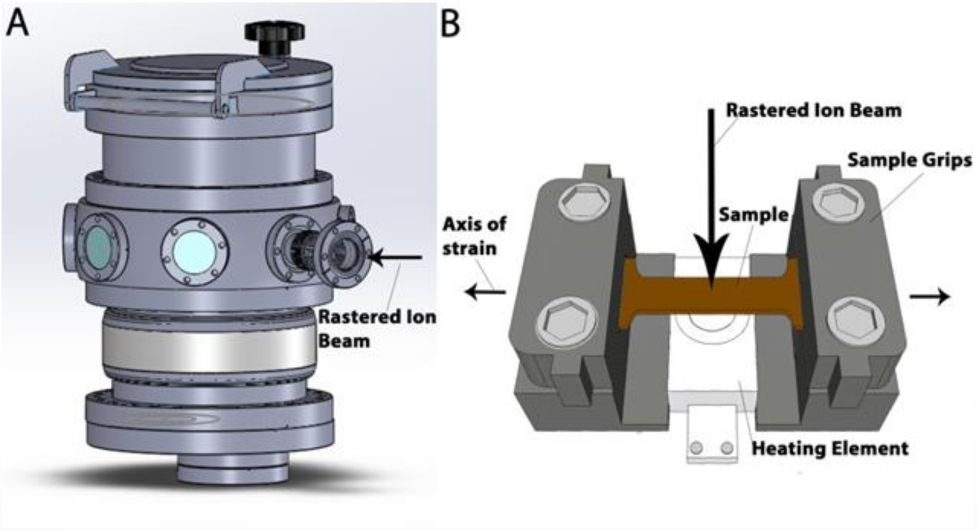


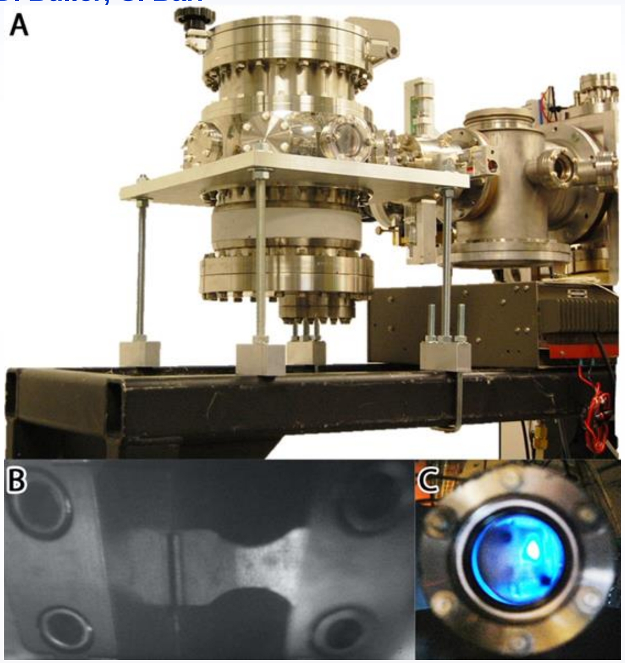
Irradiation Creep
(Loading rate 0.6 Mpa s^{-1})



Mechanical Testing End Station

Collaborator: M. Steckbeck, B. Boyce, T. Furnish, D. Bufford, D. Buller, C. Barr

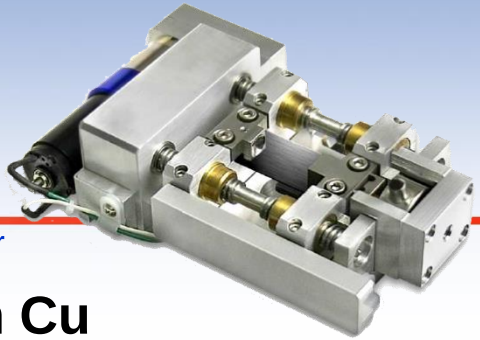




End-station developed consists of a two possible micro-mechanical test frames situated to receive a variety of ion species at energies up to 88 MeV from the 6 MV Van de Graff Tandem accelerator:

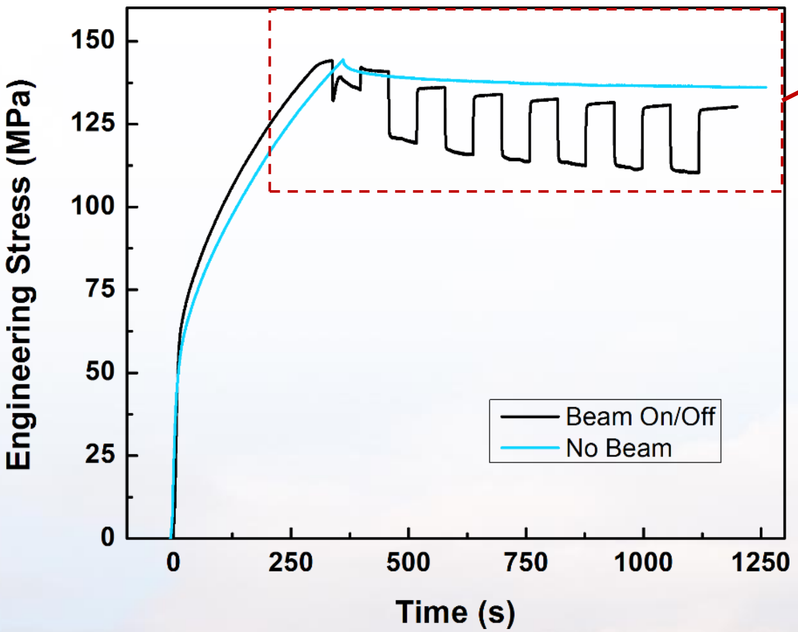
1. Commercial MTI/Fullham Multi-use Mechanical Stage (4000 N max): tensile, three point bend, creep, stress relaxation, others
2. Fatigue (custom design) for thin (5 to 10 micron samples)

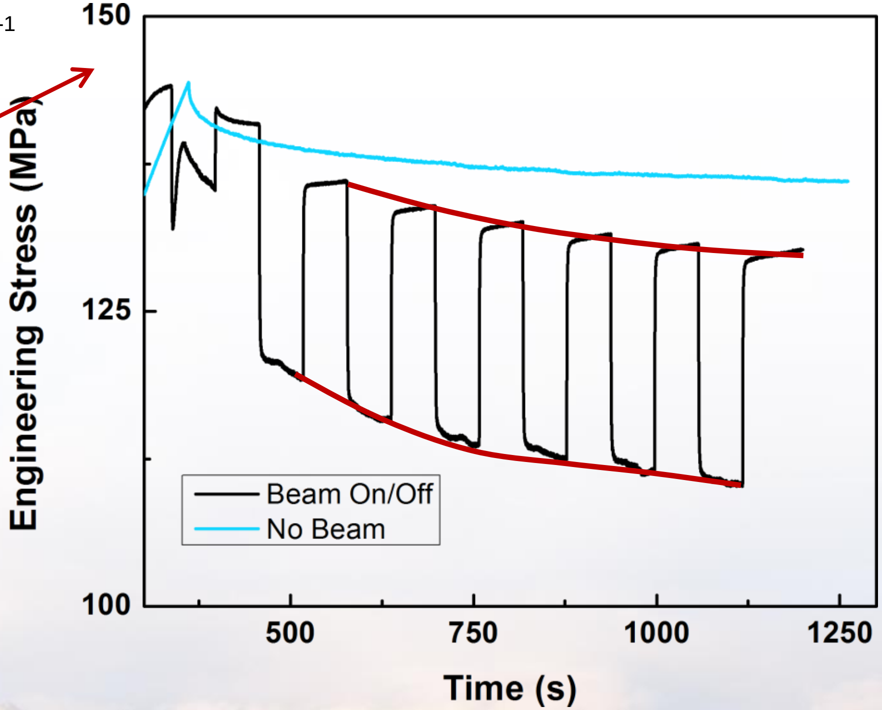
Irradiation and Stress Relaxation


MTI/Fullham SEMTester

Collaborator: M. Steckbeck, B. Boyce, T. Furnish, D. Bufford, D. Buller, C. Barr

- **0.25mm/min elongation rate to 22.5 N load in 50 um Cu**
  - Approximately 75% of typical ultimate tensile load
  - 900 s hold at constant position
  - Beam cycled on/off at 60 s intervals
  - Beam conditions: 4.5 MeV H^+: 2.1 × 10^{11} ions $cm^{-2}s^{-1}$





- Offset likely an artifact
- Different relaxation rates with beam on and off

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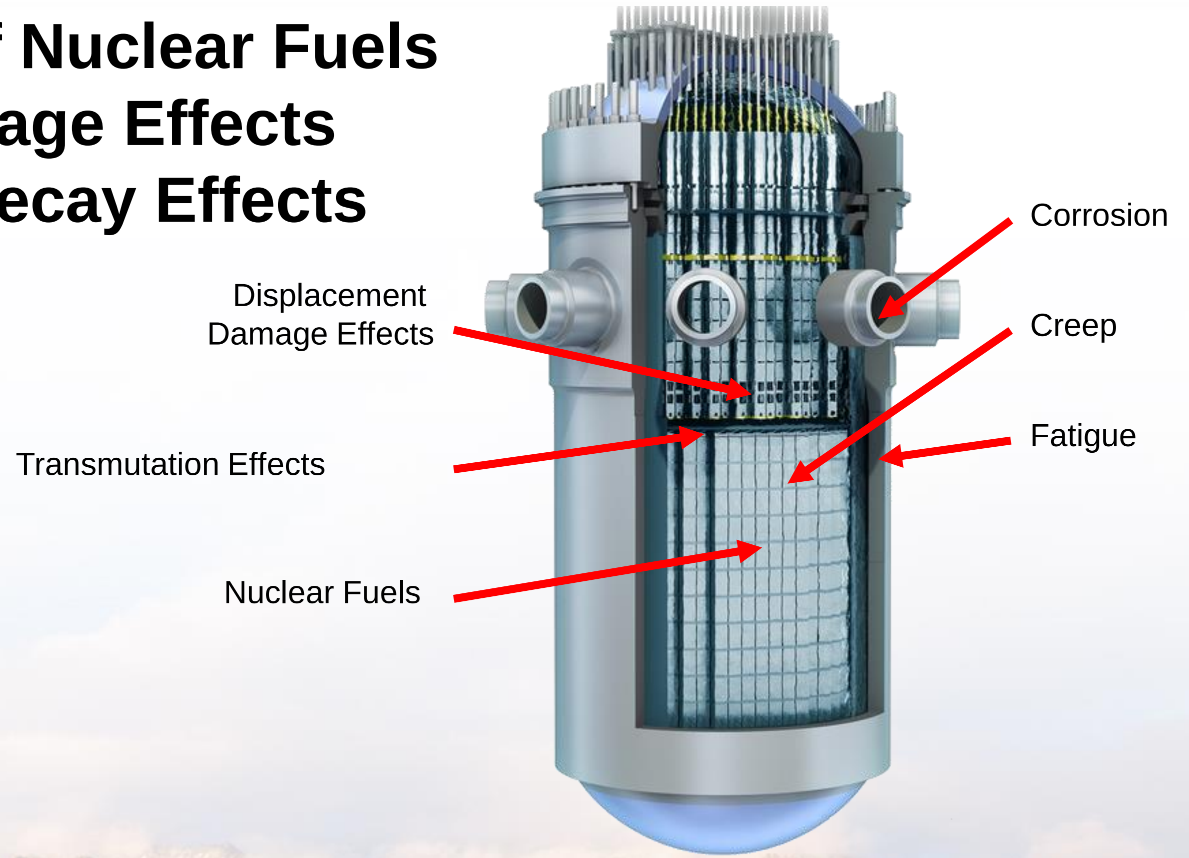
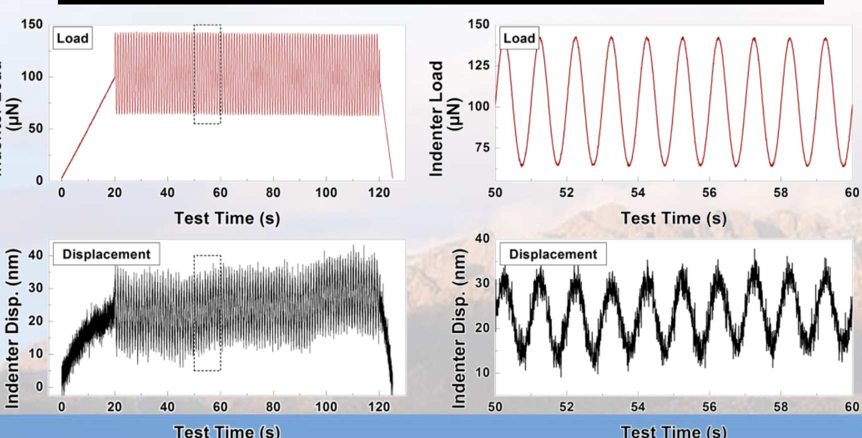


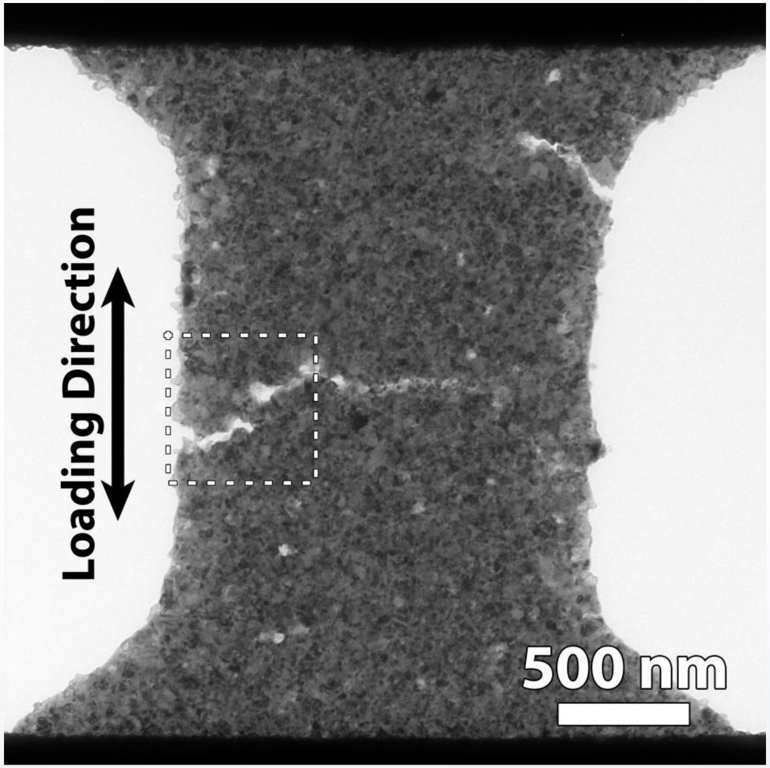
In situ TEM Quantitative Fatigue Testing

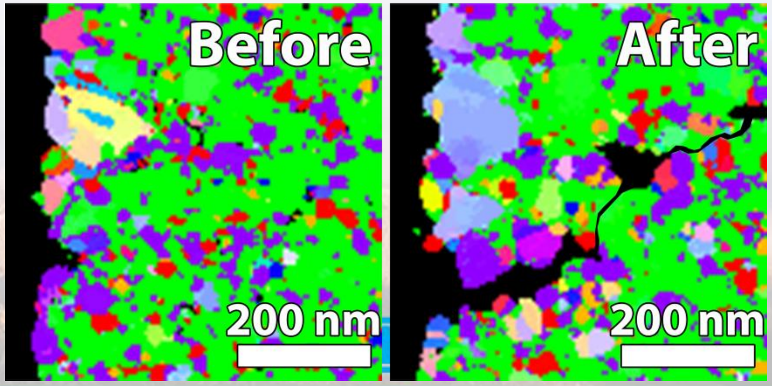
Contributors: D.C. Bufford, D. Stauffer, W. Mook

3× playback speed



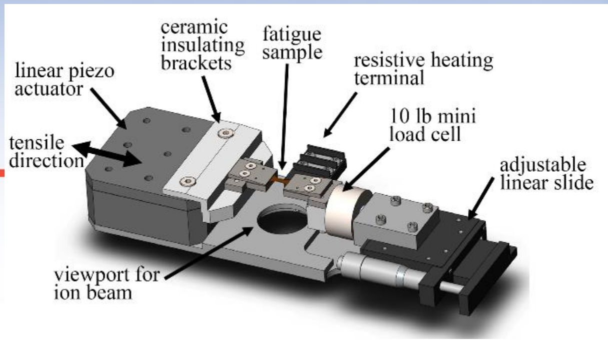
High cycle fatigue in real time with nanometer resolution

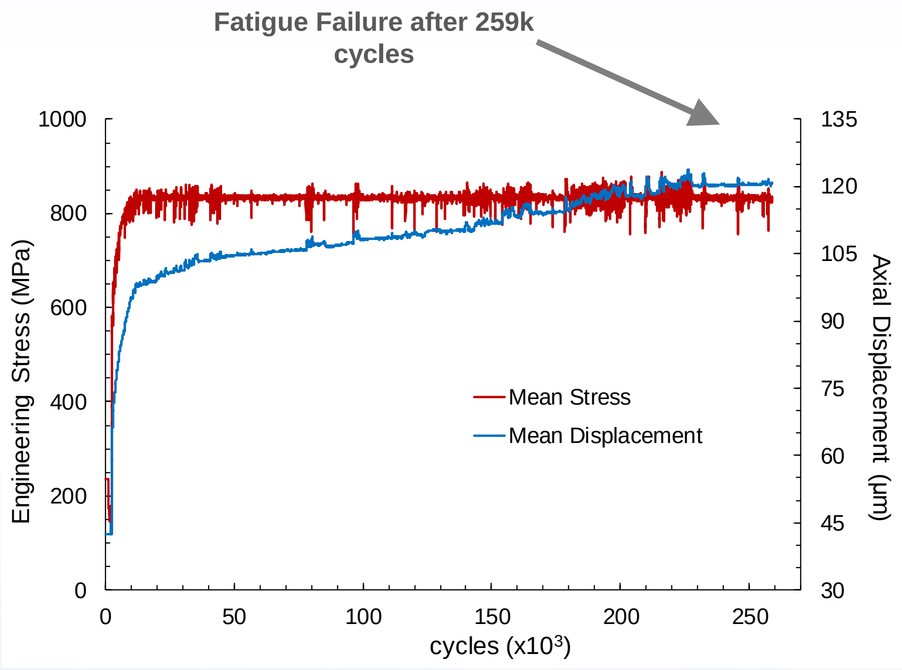


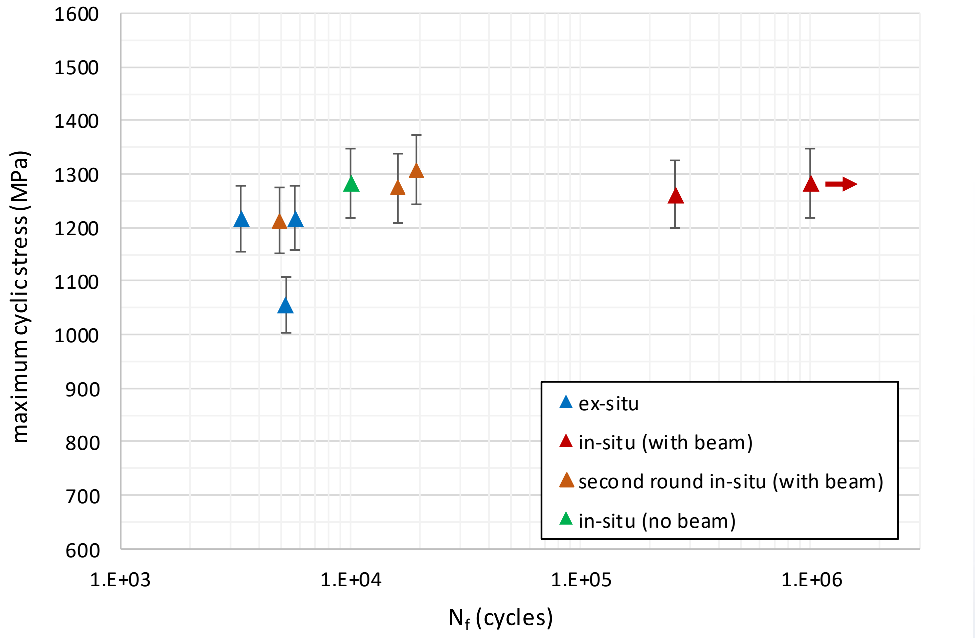


Irradiation and Fatigue

Collaborator: B. Boyce, T. Furnish, D. Buller







- Preliminary fatigue results in Ni-Fe alloy with both irradiation beam on sample (10 MeV He^{+}) and beam off sample.
- Custom mechanical stage designed for thin samples (5 to10 um thickness: suitable for allowing entire thickness to be irradiated with raster or defocused beam)

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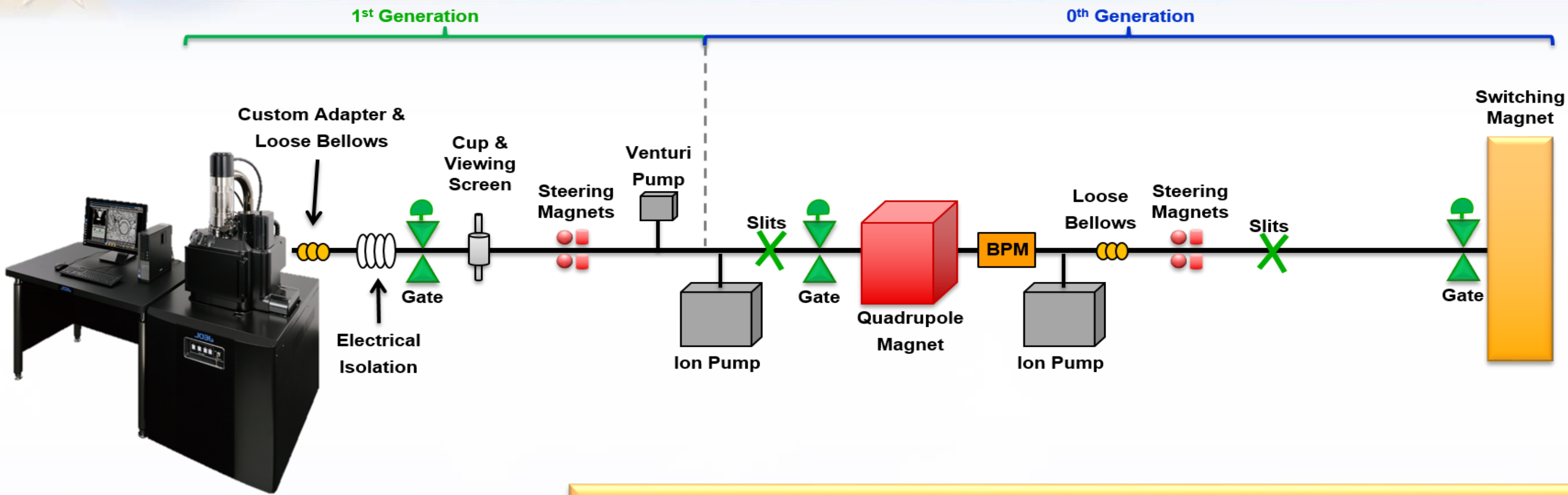
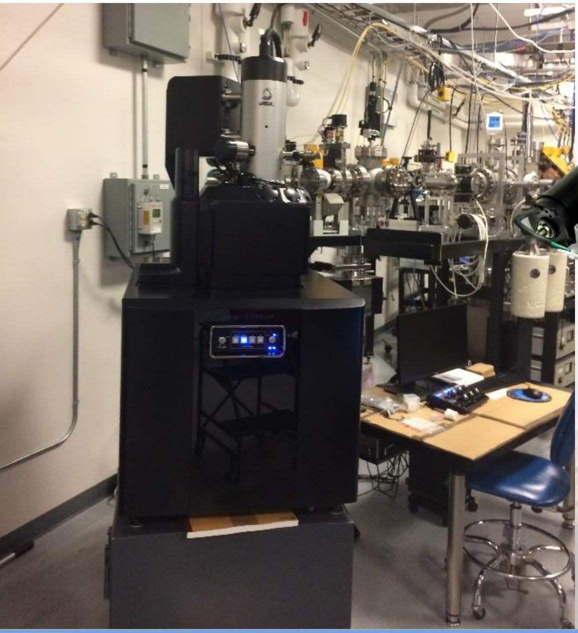
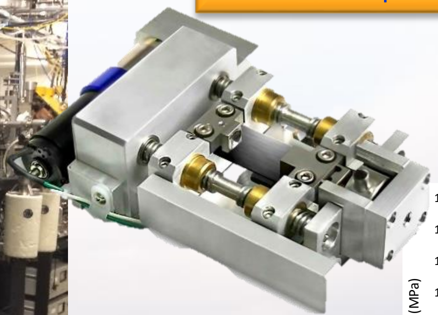
Schematic of the *In situ* SEM Beamline

Collaborators: D.L. Buller & S. Briggs

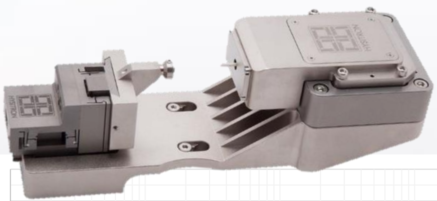




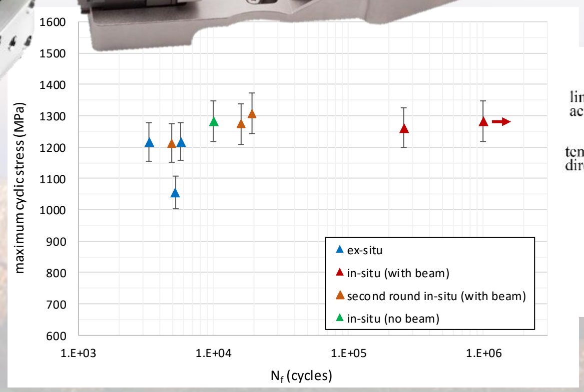
Beam Line planned for the *in situ* SEM will be developed in phases. Ultimate plan is for multiple accelerators being attached for dual or triple beam experiments.



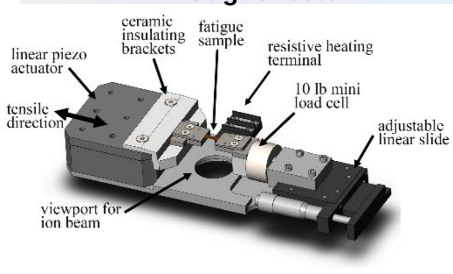
MTI Fullam Straining Heating



Hysitron PI85 Nanoindenter

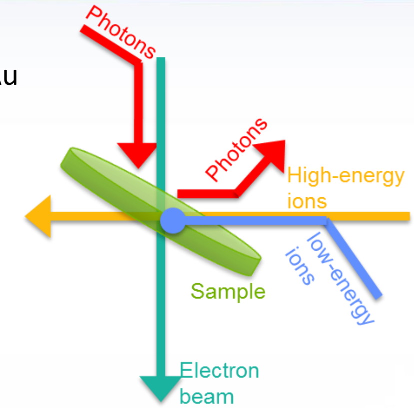


Custom-built Piezo Fatigue tester



Summary & Still Father-out Future Directions

- Sandia's I^{3}TEM capabilities:
  - *In situ* high energy ion irradiation from H to Au
  - *In situ* gas implantation
  - Heating up to 1,000 ˚C
  - Quantitative and bulk straining
  - Two-port microfluidic cell
  - Gas flow/heating stage
  - Electron tomography
  - Precession Electron Diffraction



Currently applying the current I^{3}TEM capabilities to various material systems in sequential or combined harsh environmental conditions

- Sandia's I^{3}TEM future capabilities being developed:
  - In situ ion irradiation TEM in liquid or gas (currently capable)
  - DTEM: Nanosecond resolution (laser optics being developed)
  - Beamline: Add 1 MV NEC Tandem & convert 90˚ magnet to bend beams 45˚

<u>Collaborators:</u>

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