Examining Synergistic Effects Between Damage and Gas Accumulation with In-situ TEM

SAND2017-5099C

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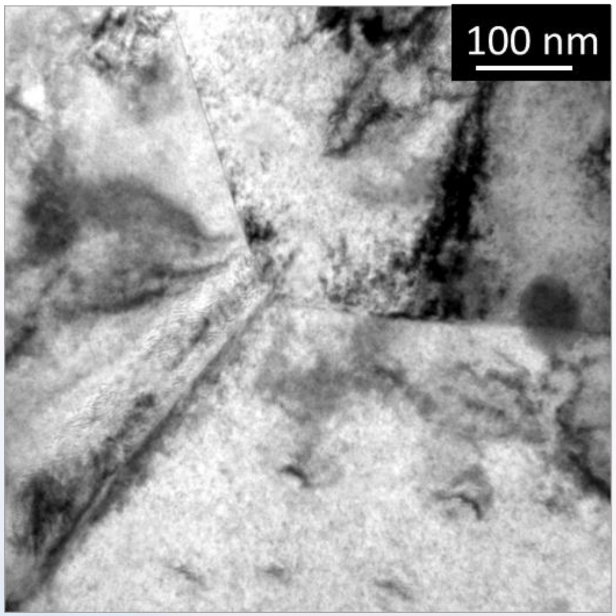
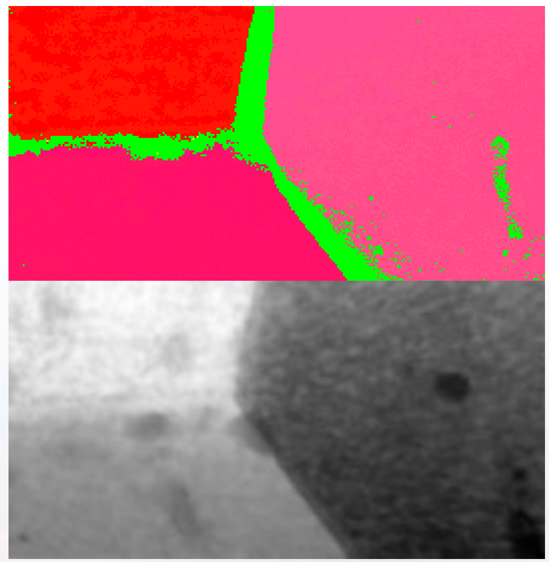
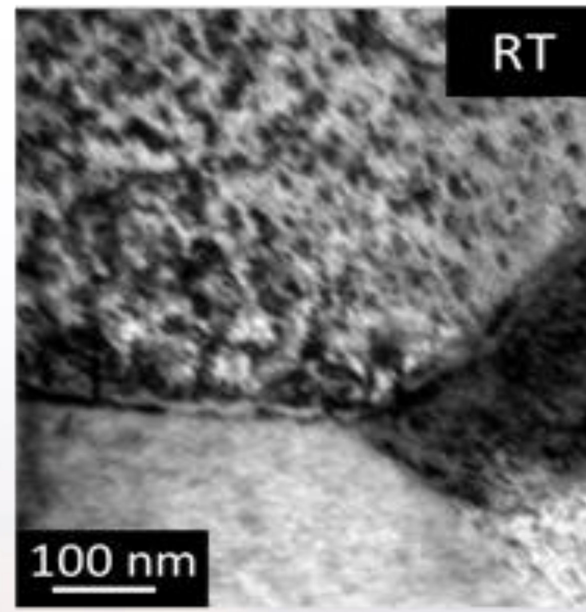
May 9th, 2017







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Outline

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2. Accelerated Aging of Neutron Damage and Gas Accumulation using Ion Beams
3. Overview of in-situ Transmission Electron Microscopy Irradiation Facility at SNL
4. **Zircaloy**
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   B. In-situ TEM Results
      i. Self Ion (Zr) irradiation
      ii. 10 keV He implantation
      iii. 10 keV He followed by Zr irradiation
      iv. Concurrent He + Zr irradiation
      v. Concurrent He + D + Zr irradiation
5. **$LiAlO_2$**
   A. Background and Planned Experiments
   B. Sample Description and Images
6. Summary

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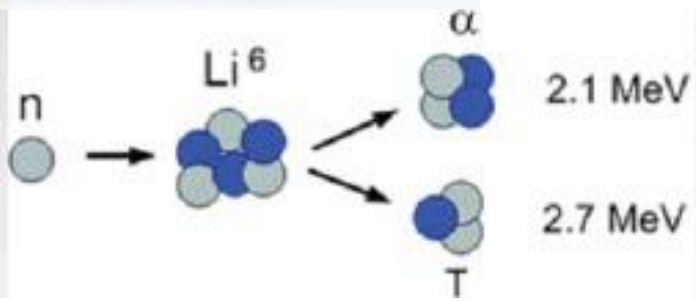
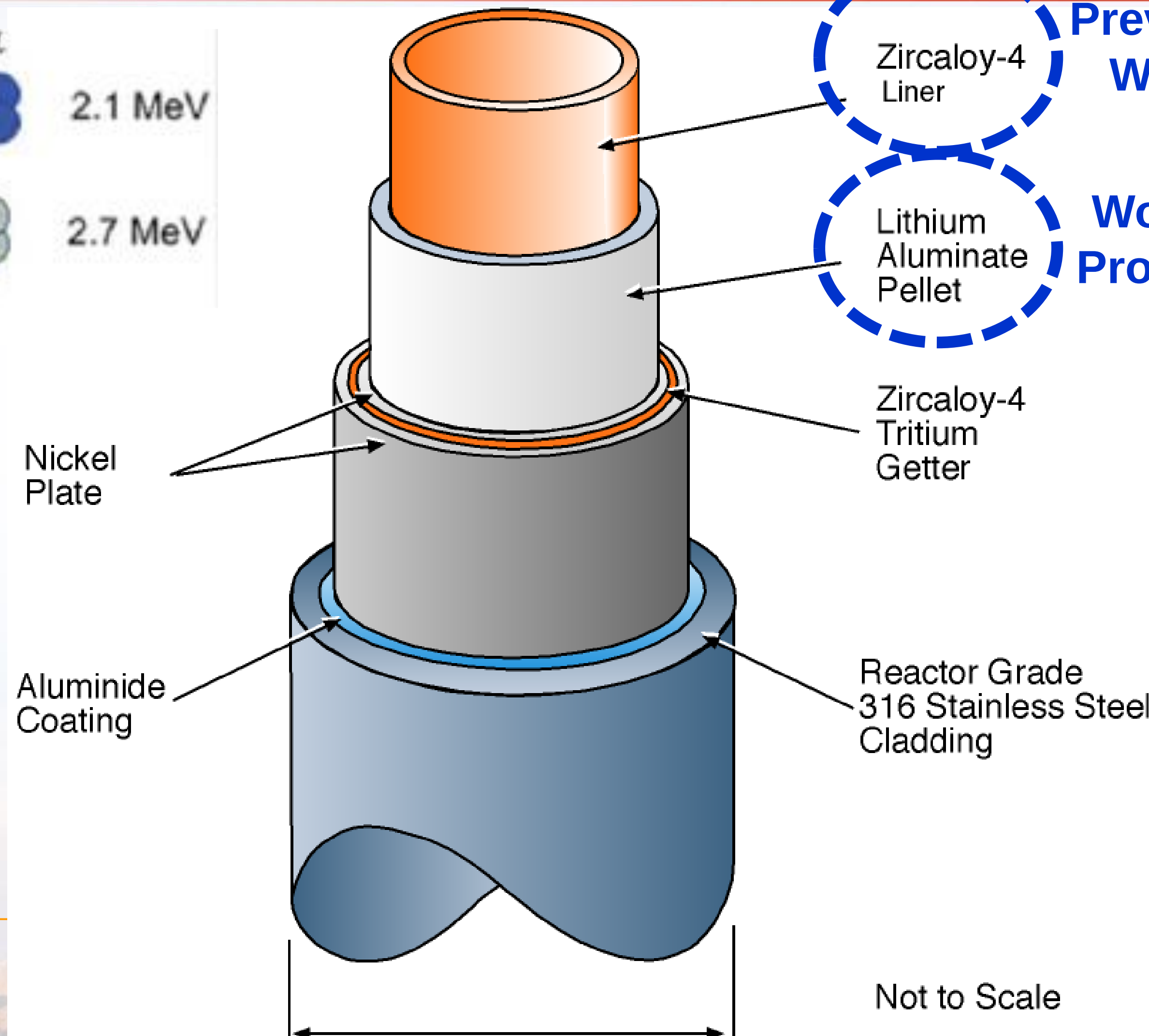
Tritium Producing Burnable Absorber Rod (TPBAR)





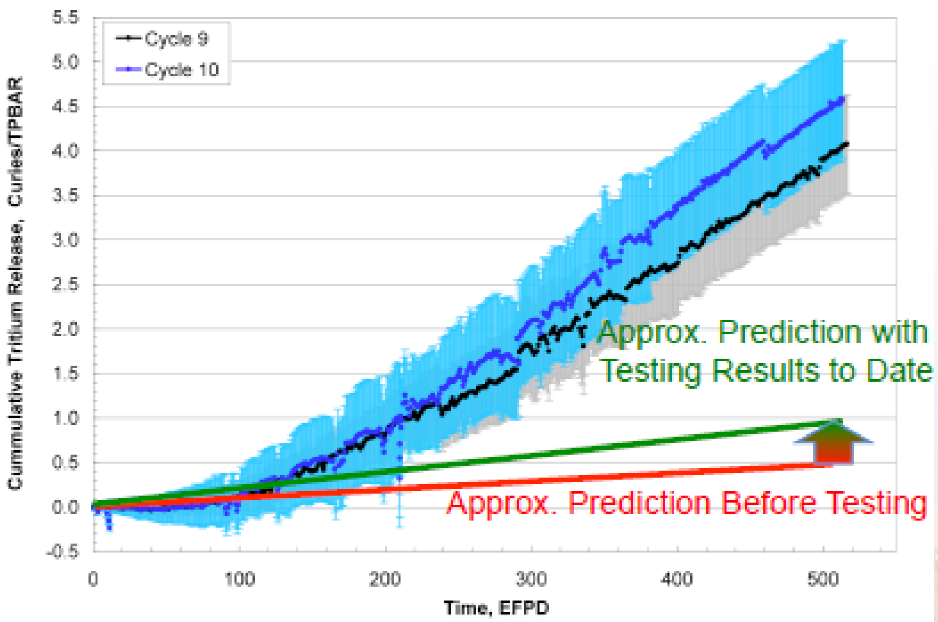
Previous Work

Work in Progress

- Displacement Damage
- Helium Implantation
- Tritium Implantation
- Elevated Temperatures

Understanding Tritium Permeation in TPBAR

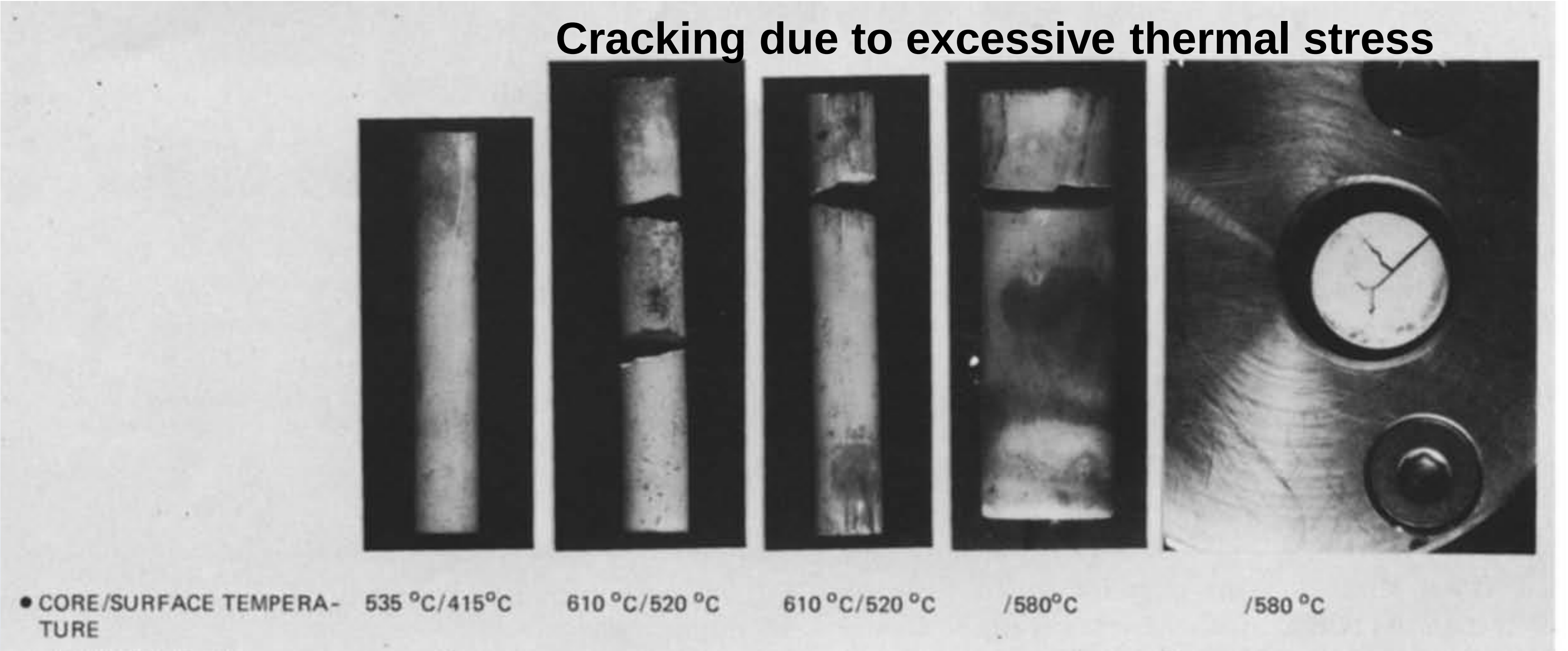
- TPBAR ^3H permeation is higher than predictive performance models
  - In 2004, during Cycle 6, the predicted levels were ~0.5 Ci/TPBAR/cycle and actual levels were ~4 Ci/TPBAR/cycle (0.04% of total ^3H produced)
- Mechanisms responsible for differences between predictions and observations are not well understood
- Currently building an understanding of fundamental ^3H-defect interactions



Burkes, Senor, Longoni and Johns, TFG Meeting 2016, Rochester, NY

Traditional Experiments use Fast Reactor Irradiations

- In-reactor irradiations of bulk $LiAlO_2$ at high flux test reactors
- Typically quantify **macroscopic** (e.g. porosity, volume, cracking) and **mechanical** property changes



Botter *et al* JNM 160 (1988) 48-57

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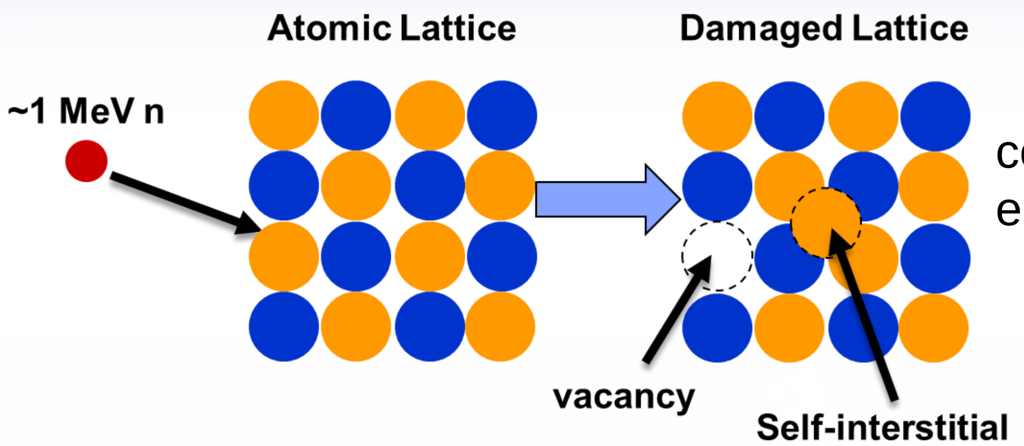
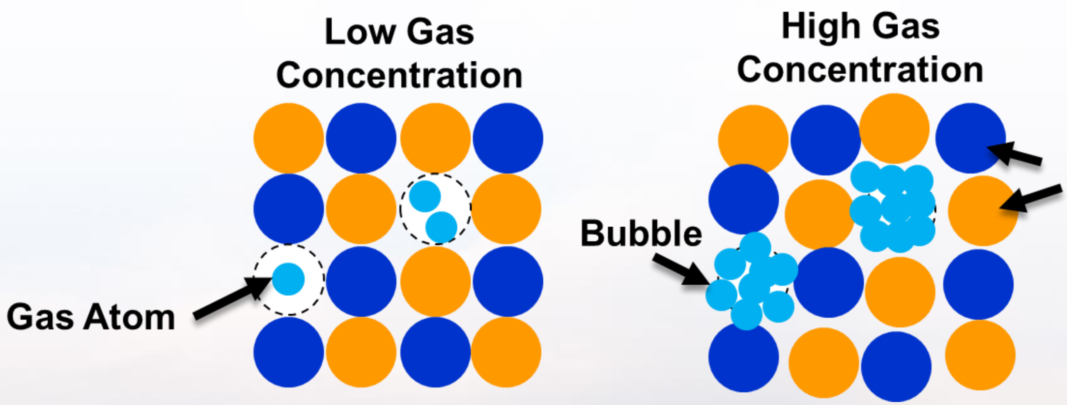
Fundamental Interaction of Neutron Irradiation Damage and Gas Accumulation May Play a Role



Over time more complicated defects, e.g. dislocations and voids, will form

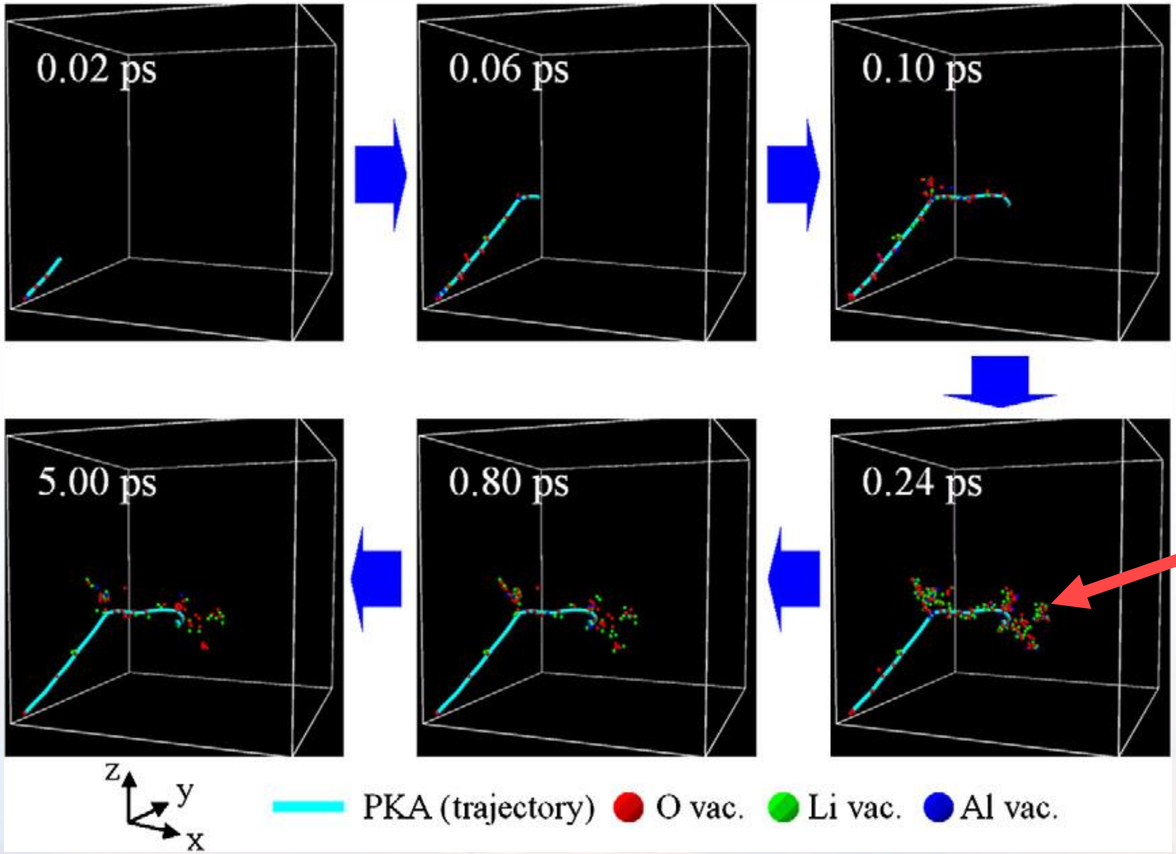


Bubbles displace atoms and eventually eject interstitials to form larger He-V clusters

The constantly changing damage state changes the way gas atoms accumulate in the material

High Energy Ions Can Produce Multiple Defects in Displacement Cascade

MD simulation of displacement cascade in $LiAlO_2$ (PKA = 5 keV)



Many defects form due to impact of ONE ion (most eventually recombine)

Tsuchihira *et al* JNM 414 (2011) 44-52

Accelerated Aging by Ion Beam Irradiation

Benefits

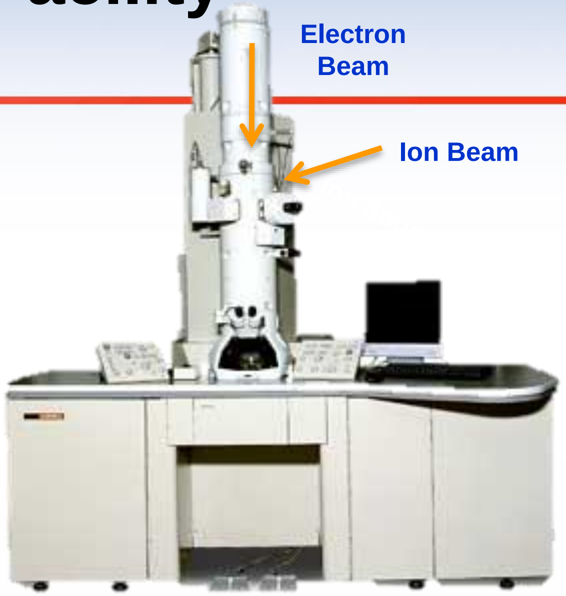
- Predict material behavior in radiation environment from a fundamental point of view
- Isolate specific variables (e.g. ion, damage, gas, temperature).
- Damage that would normally occur over several months or years in a reactor can be simulated in a matter of minutes or hours with an ion accelerator, **without activation**

Ion irradiation is used to understand fundamental mechanisms occurring due to radiation damage at the atomic scale.

Limitations

- Higher ion flux than reality
- Difficult to predict dose rate effects
- Injected ions can influence the damage properties or chemistry of material
- Only irradiates surface layers

In situ Ion Irradiation TEM Facility



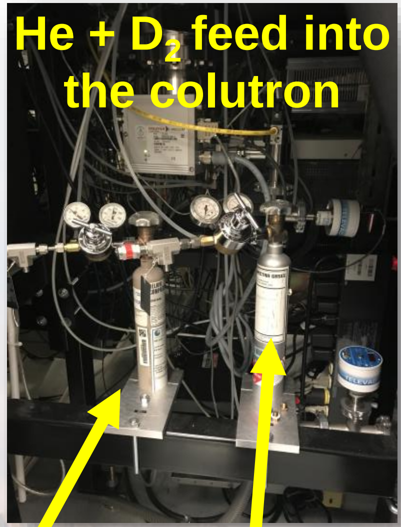
Capabilities

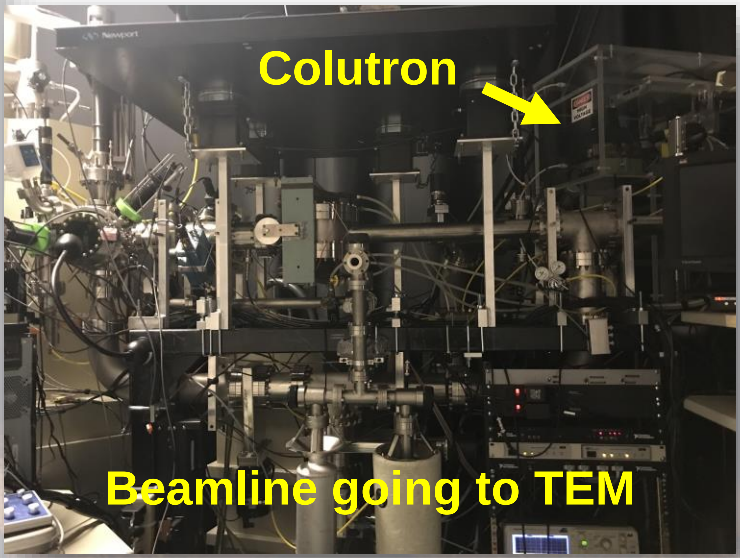
- 200 kV LaB_6 TEM
- Ion beams considered:
  - Range of Sputtered Ions
  - 10 keV D_2^{2+}
  - 10 keV He^+
- All beams hit same location

We have produced 430 eV He with the Colutron –below knock-on energy!

- Nanosecond time resolution (DTEM)
- Precession scanning (EBSD in TEM)
- *In situ* PL, CL, and IBIL

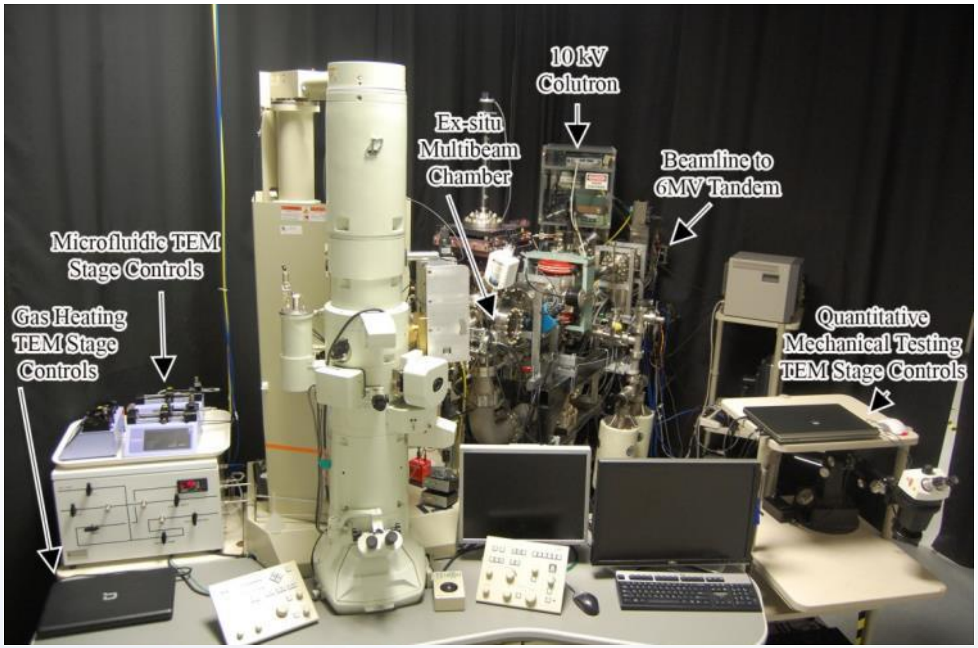
- *In situ* vapor phase stage
- *In situ* liquid mixing stage
- *In situ* heating
- Tomography stage (2x)
- *In situ* cooling stage
- *In situ* straining stage



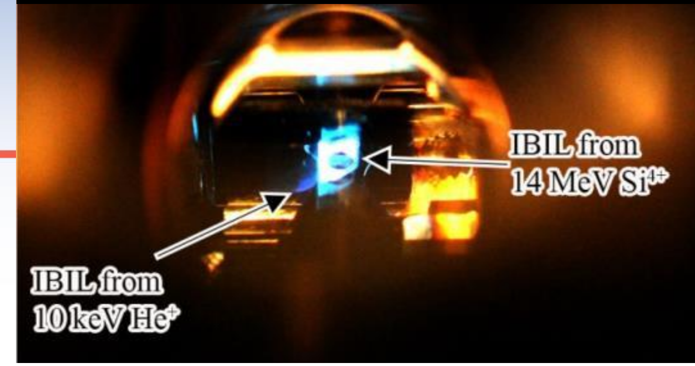


Sandia's Concurrent *In situ* Ion Irradiation TEM Facility

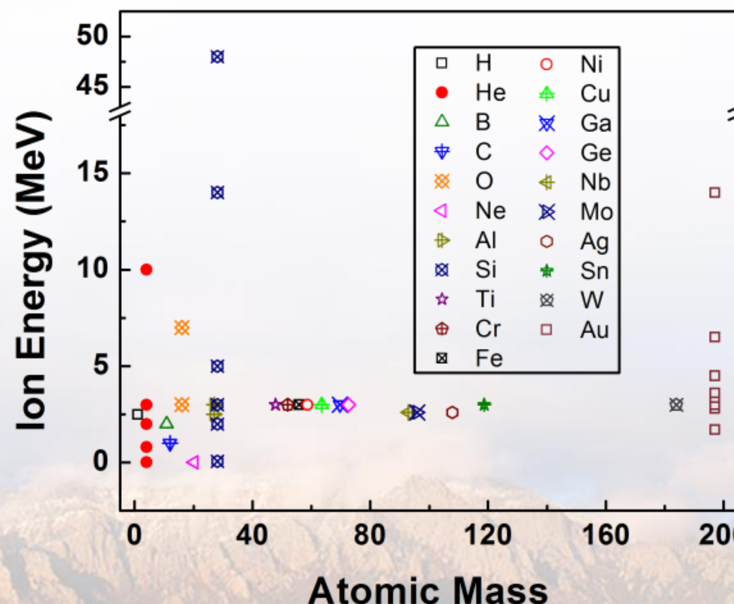
10 kV Colutron - 200 kV TEM - 6 MV Tandem



IBIL from a quartz stage inside the TEM



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



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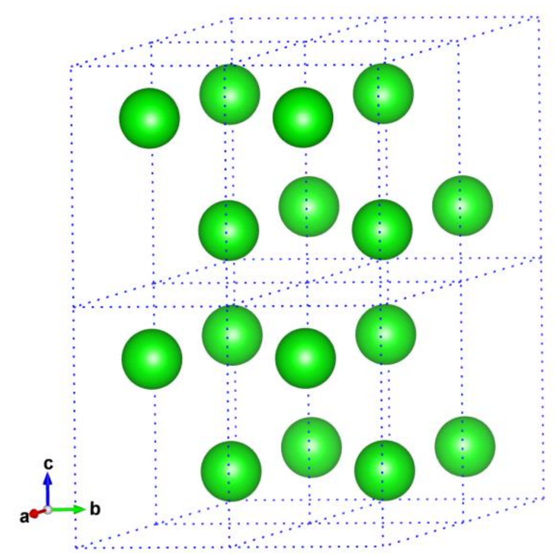
Zircaloy Background



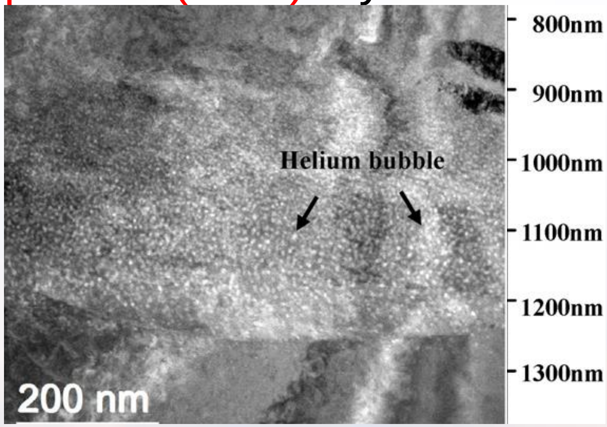
Crystal Structure of α-Zr (HCP)

What is Zircaloy?

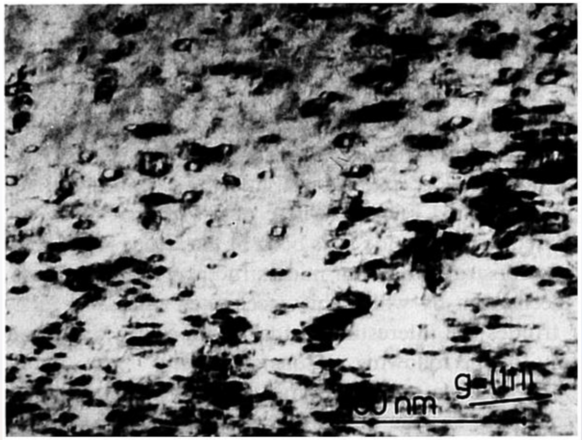
- Zircaloy-2: predominantly used as fuel cladding for BWRs
  - α-Zr, 1.5% Sn, 0.15% Fe, 0.1% Cr, 0.05% Ni
- Zircaloy-4: Removed the Ni and increased Fe content for less H uptake in certain reactor conditions
  - α-Zr, 1.5% Sn, 0.2% Fe, 0.1% Cr
- Zr-Nb alloys (e.g. Zirlo) are also common
- α-Zr has a hexagonal close-packed (HCP) crystal structure up to 810ºC

Gas and defect behavior in Zr/Zr alloys

- ^{3}H, H, and He diffusion and release
- Bubble formation
- Irradiation induced metallic precipitate formation



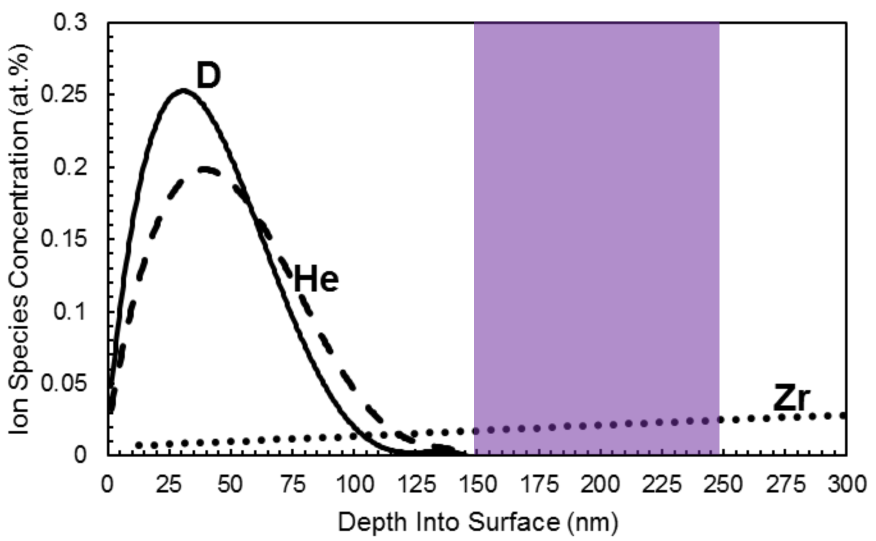
He bubbles in Zr-Nb alloy
Shen et al Mat Char 107 (2015) 309-316

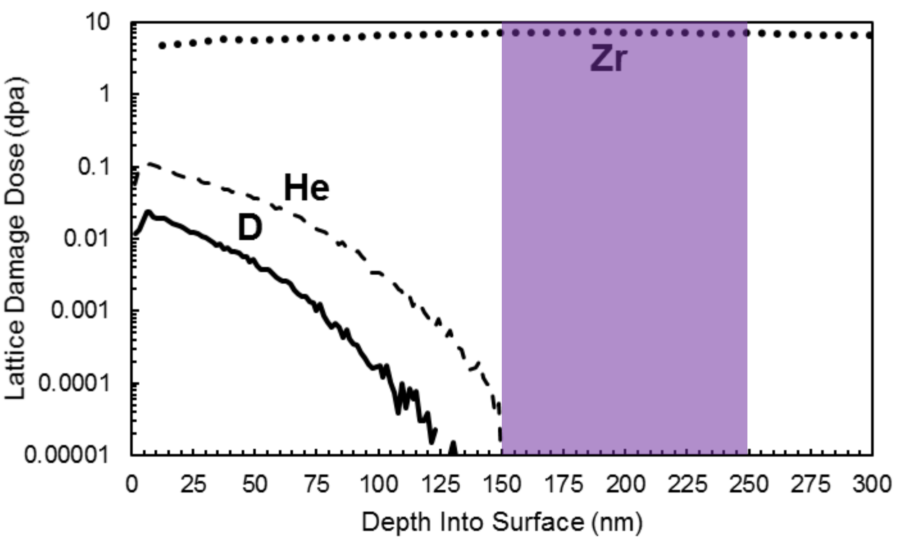


TEM of Zr tritide after 325d
Schober et al JNM 141-143 (1986) 453-457

Relative Damage and Gas Distributions in Zr

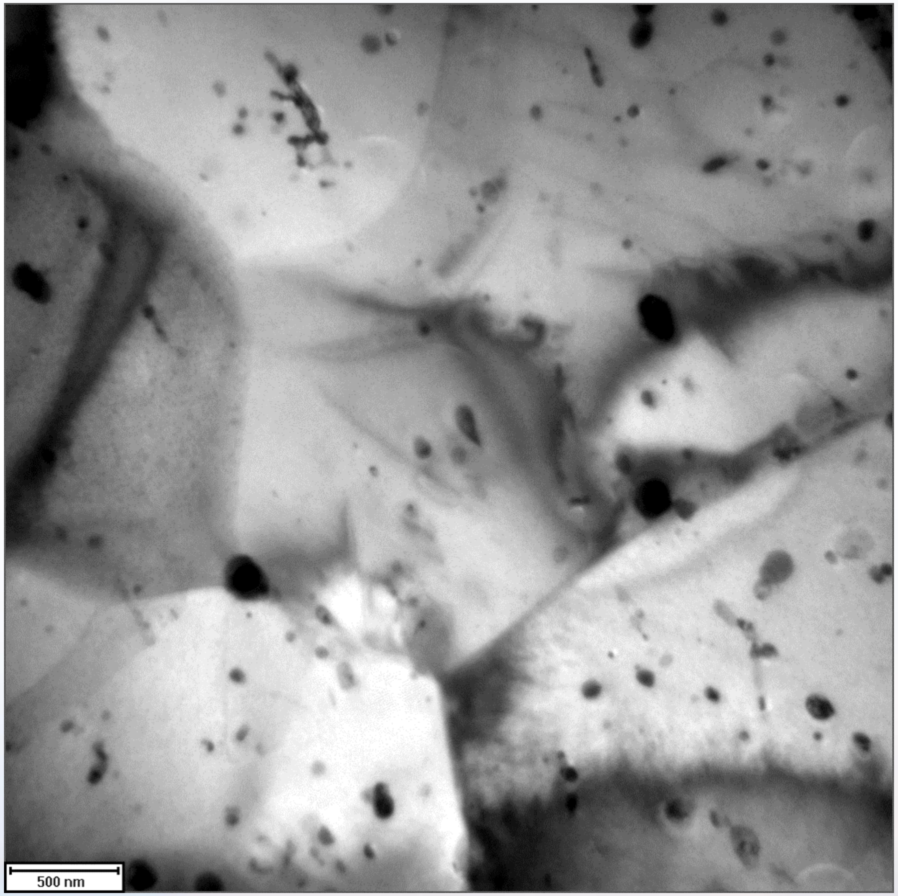
- Ion concentration and damage are scaled based on the irradiation time
- Most Zr travels through entire TEM foil
- Zr produces two orders of magnitude higher damage than He
- These experiments were aimed at observing kinetic effects in-situ, so experiments were run overnight and the exact gas concentrations/damage doses are not all known

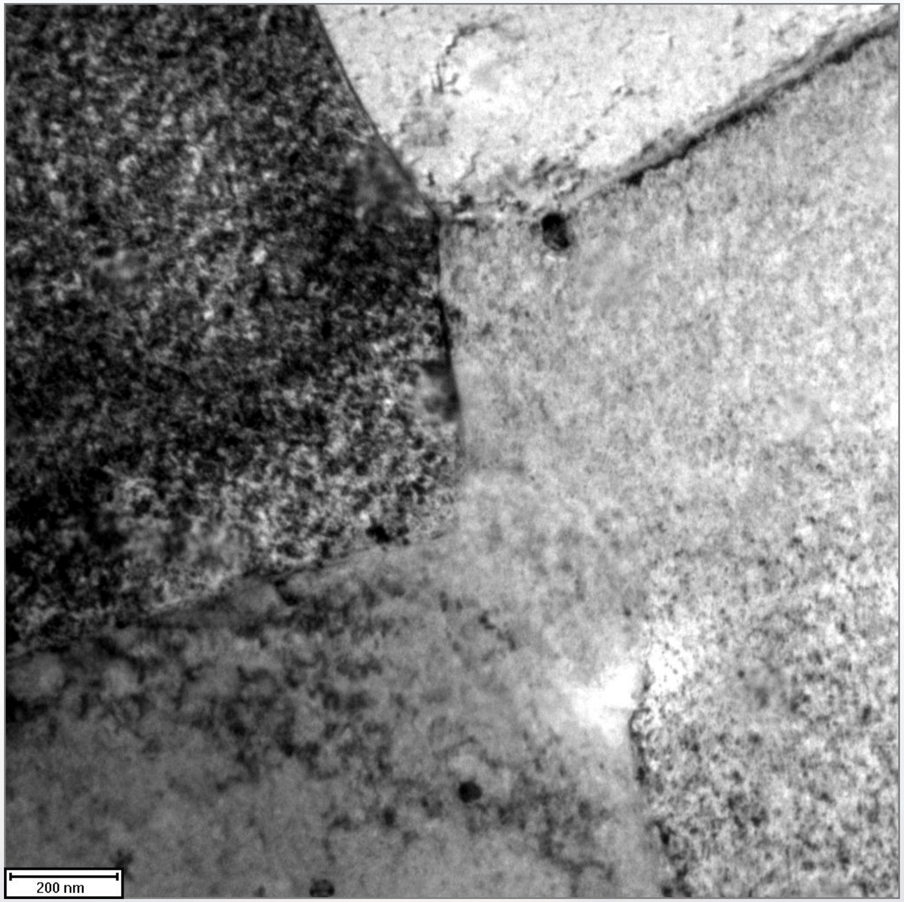




SRIM calculations of 10 keV He, 5 keV D, 3 MeV Zr implantation depth and damage

3 MeV Self Ion Irradiation at 310 C

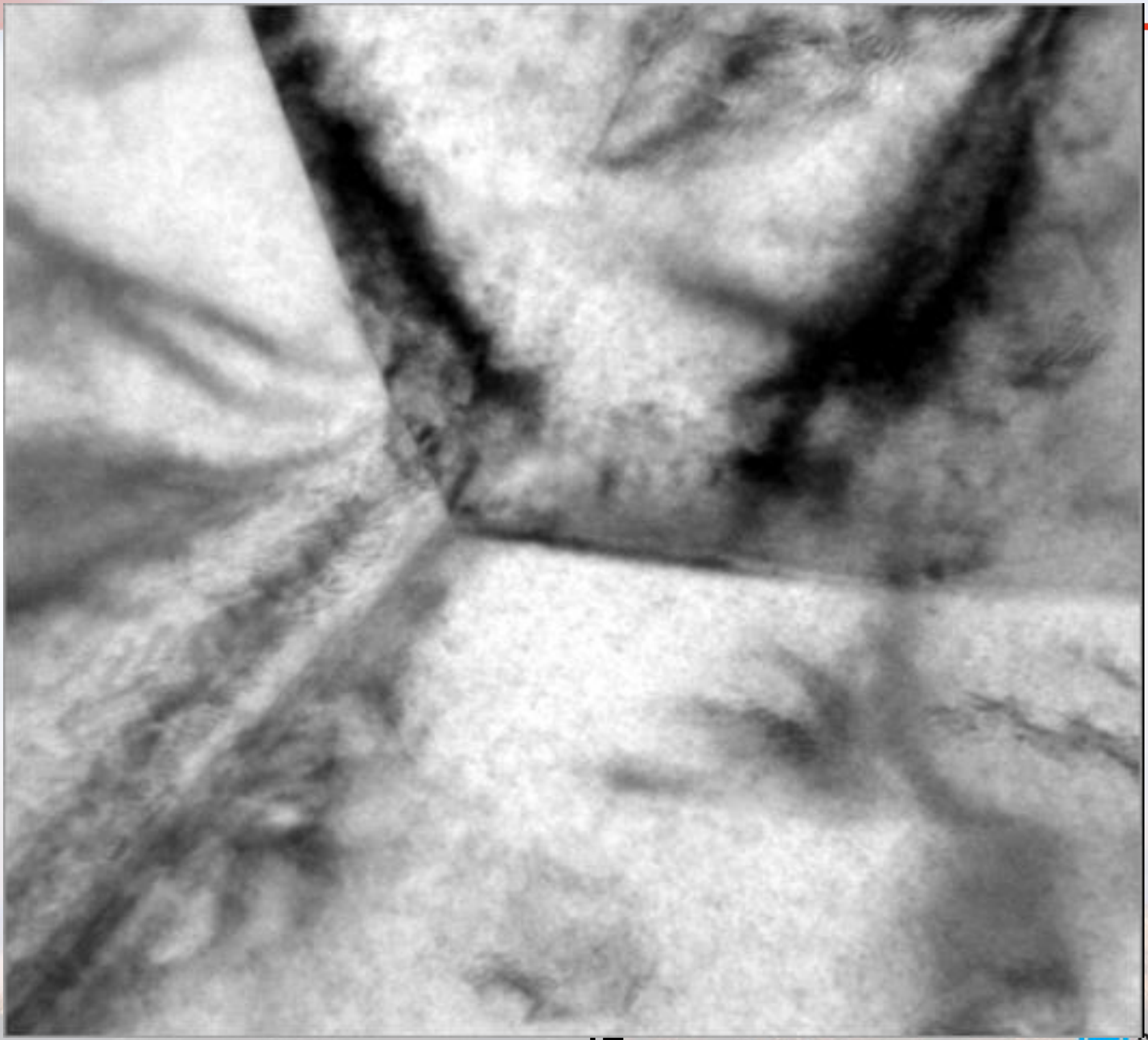




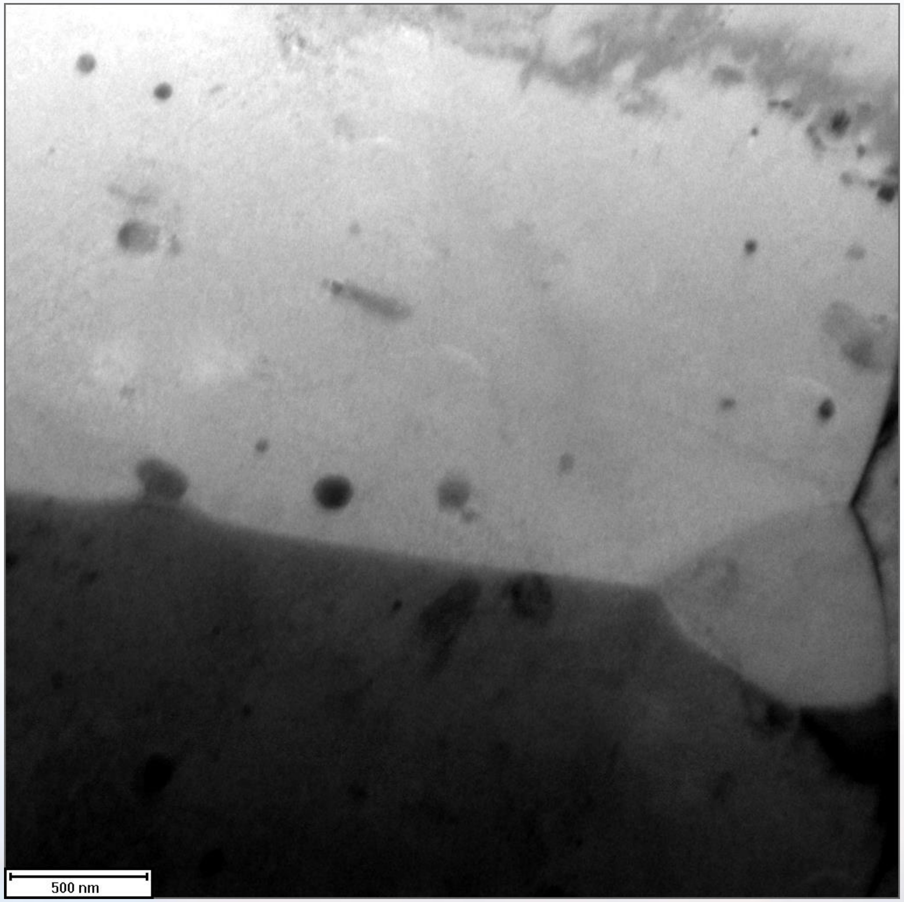
Before Irradiation

After Irradiation ≈ 7 DPA

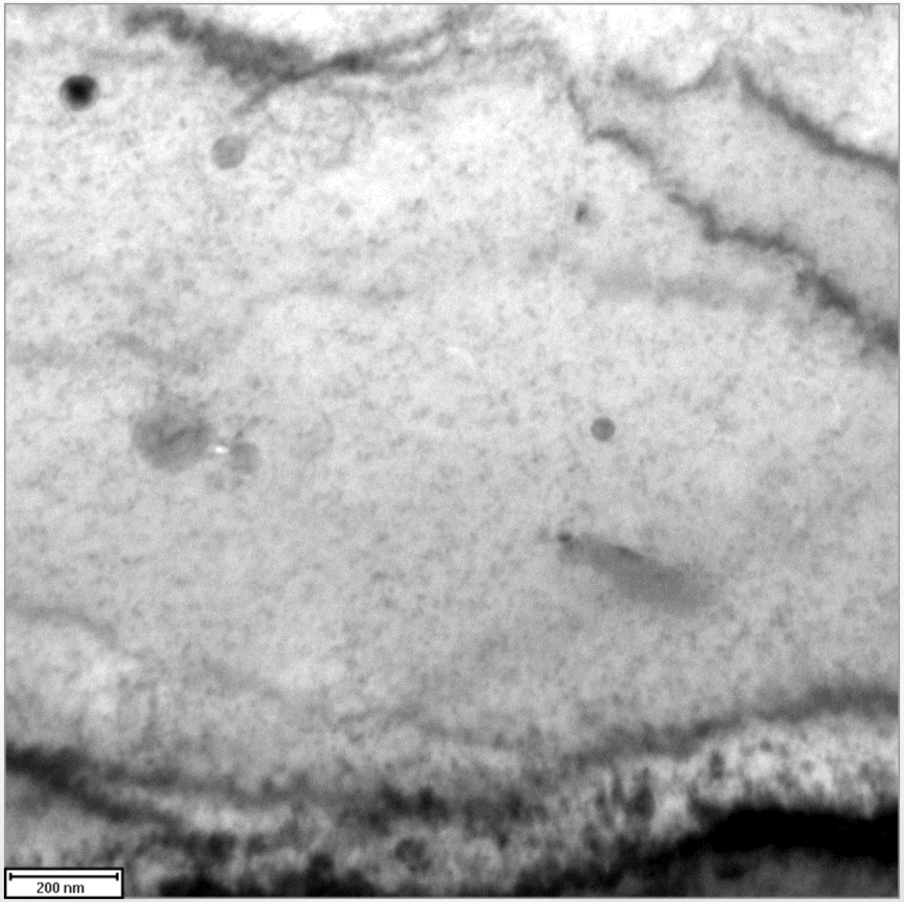
3 MeV Self Ion Irradiation at 310 C



10 keV He^+ Implantation at 310 C



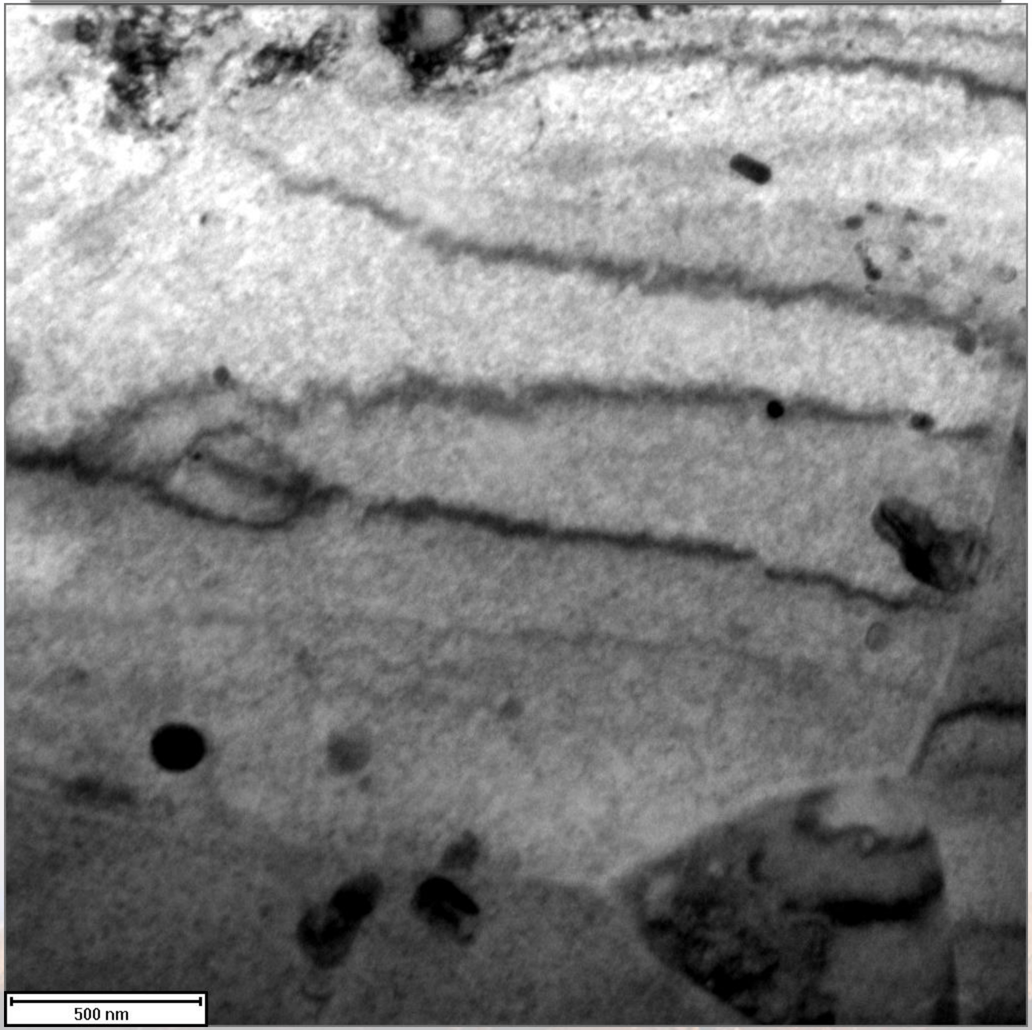
Before Implantation



After Implantation Damage, No Cavities

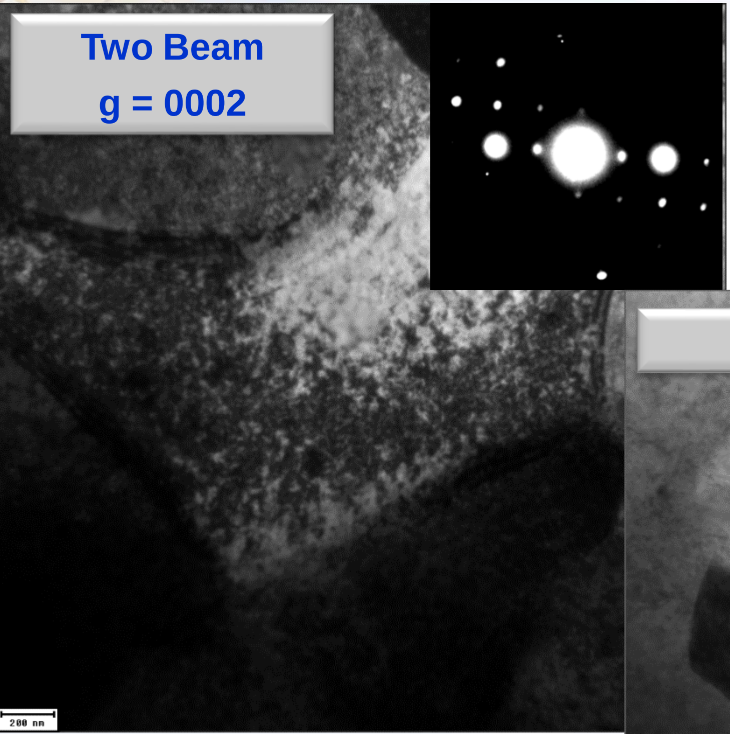
3 MeV Self Ion Irradiation after He^+ Implantation

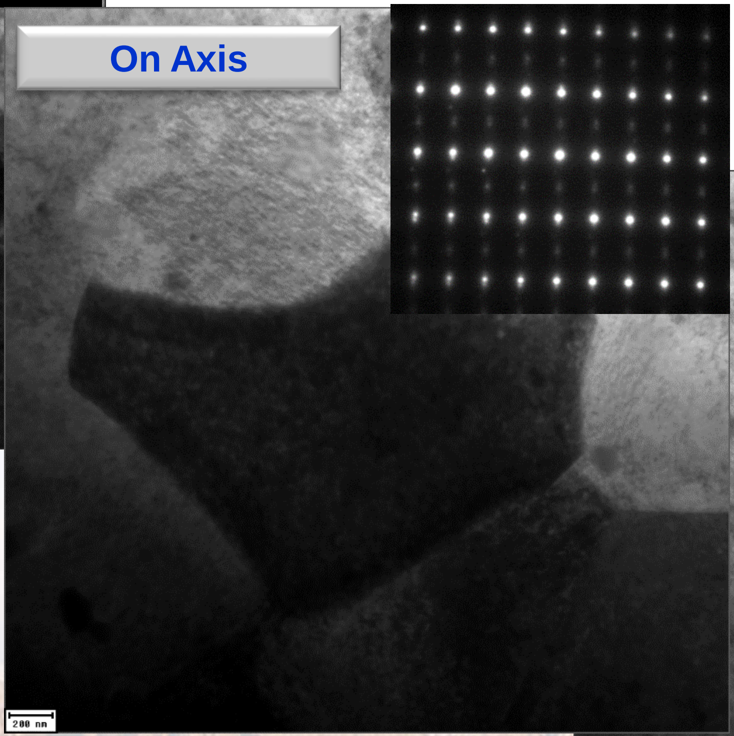
High Density of Defects but No Cavities

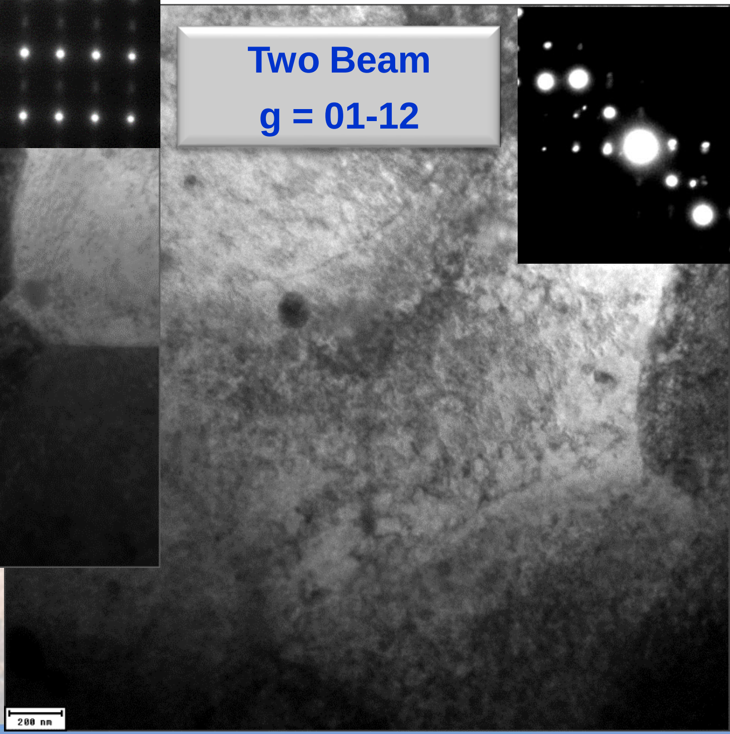


3 MeV Self Ion Irradiation after He^+ Implantation

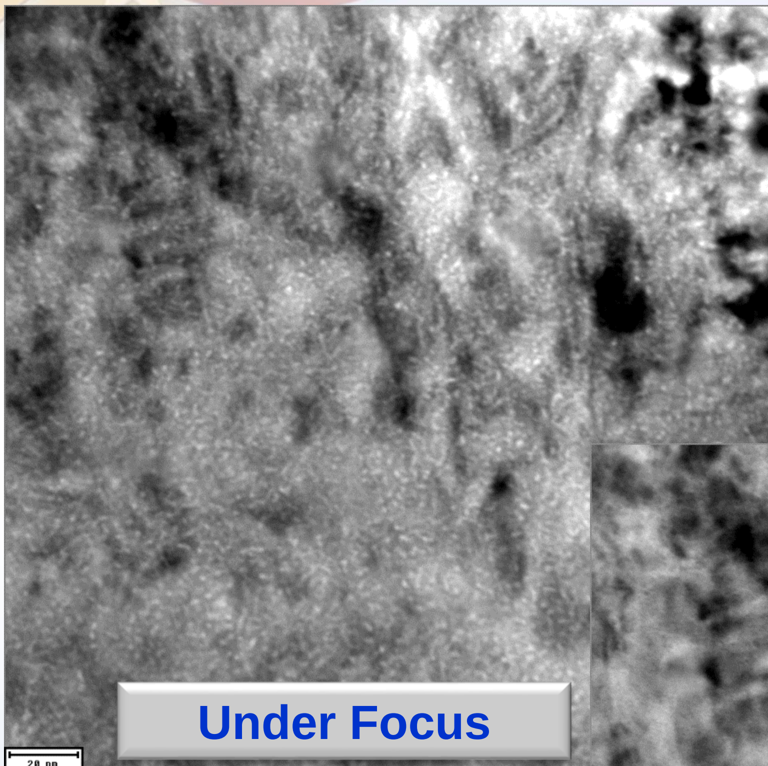
No distinct quantifiable defect structures were observed

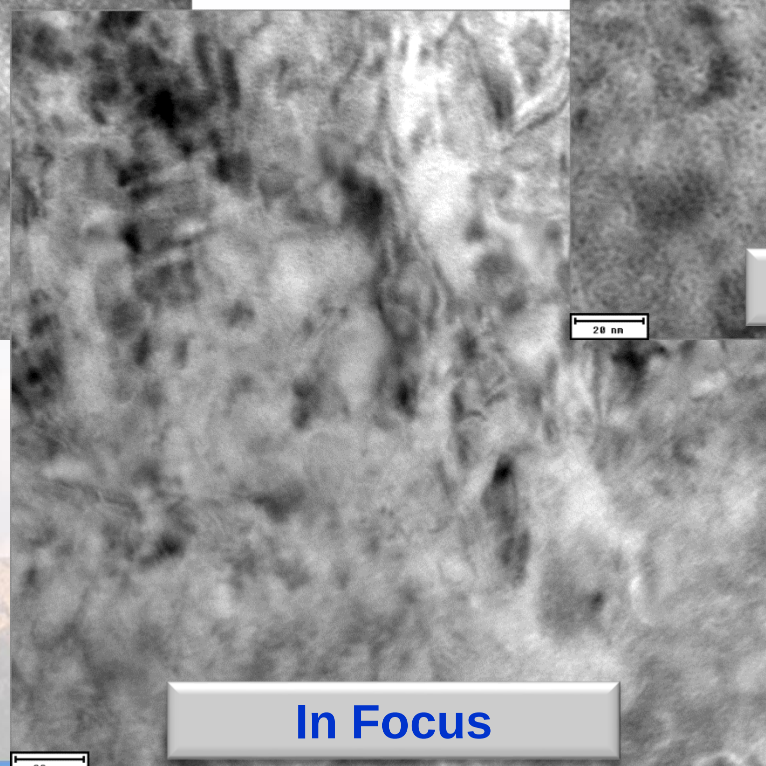


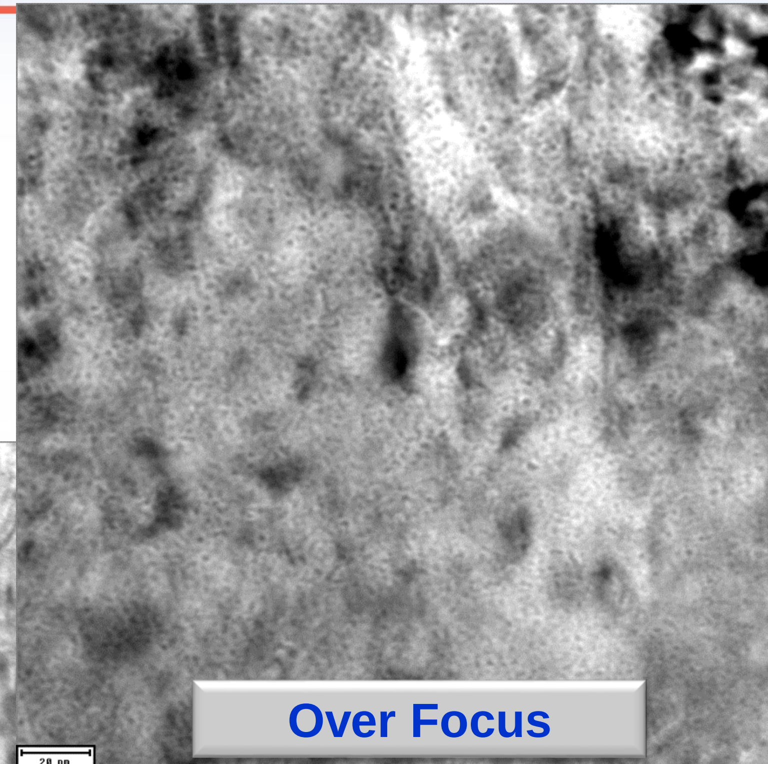




Through Focus Imaging of Cavities: 30 Days Later

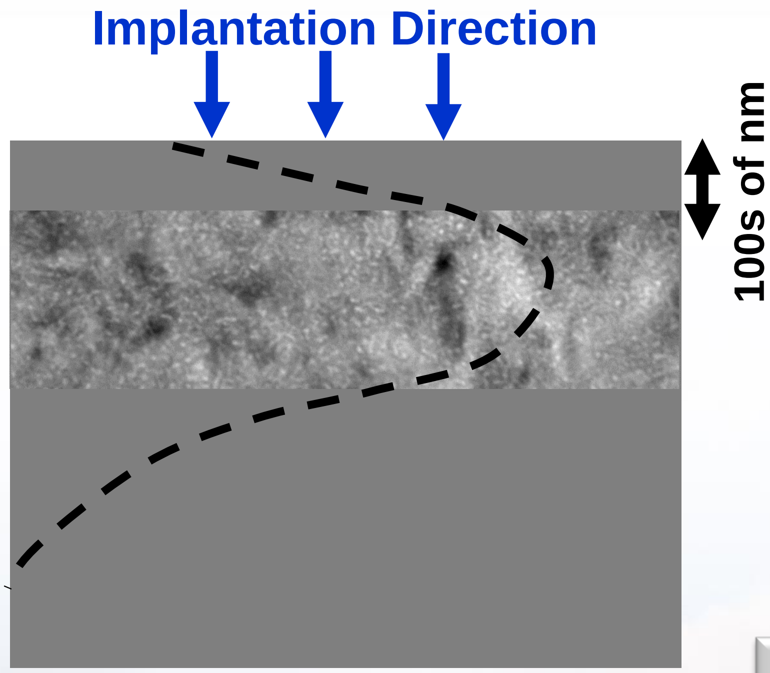




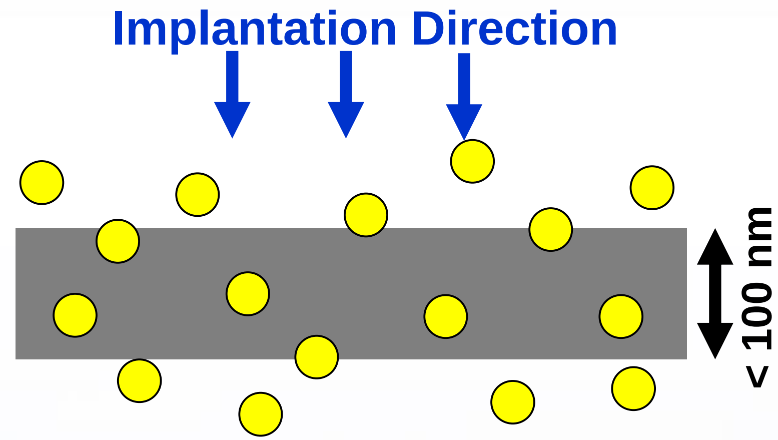


Why is finding bubbles after 30 days interesting?

- In-situ ion irradiation produces a different set of issues due to surface effects



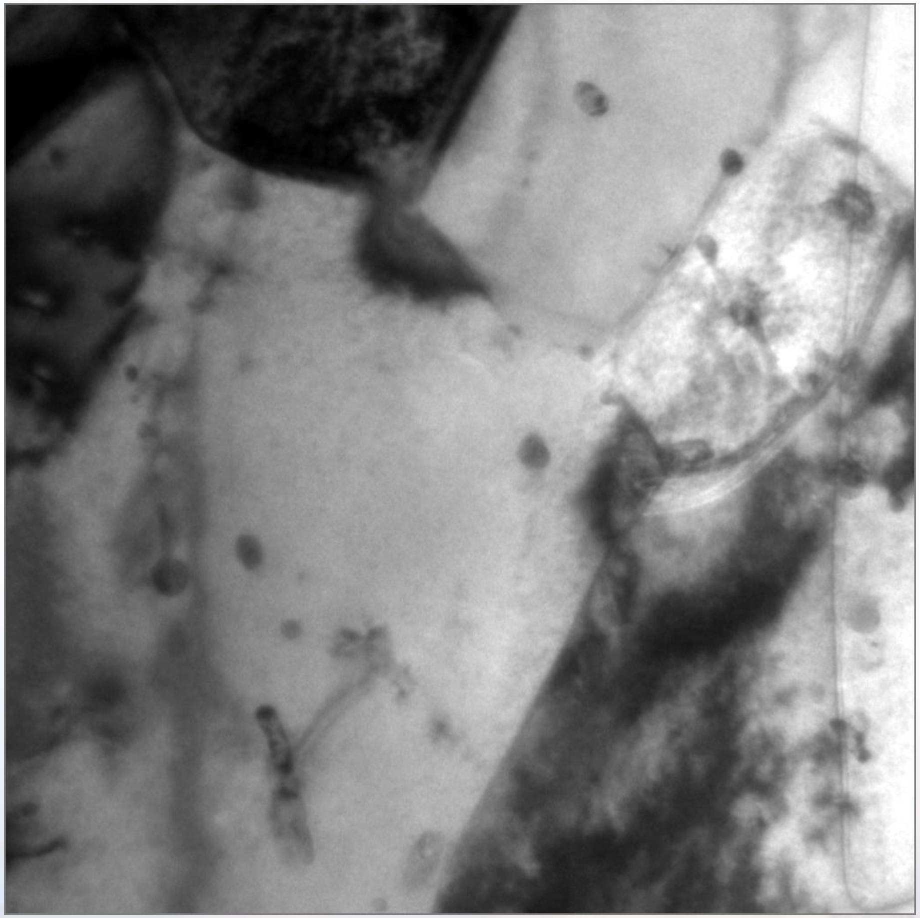
Bulk Irradiation: He/D diffuses at the same rate, but becomes trapped by defects before reaching surface.



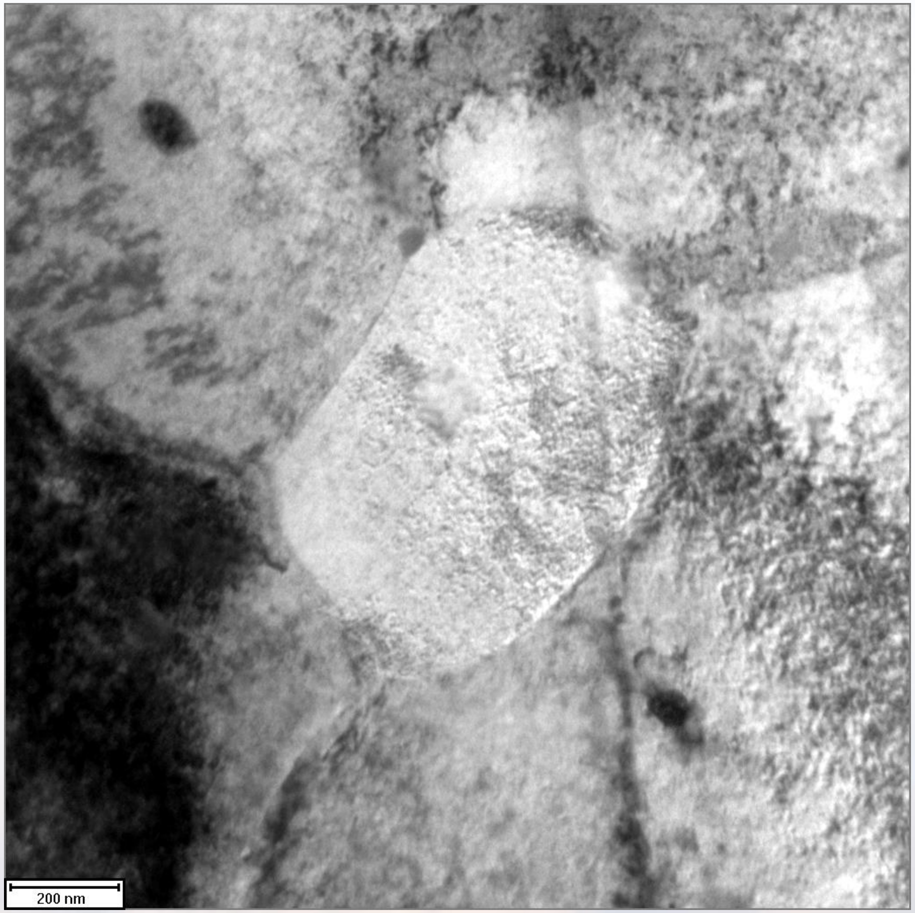
Thin-Film Irradiation: Much of the He/D diffuses to the surface before being trapped by a defect

Bubbles form in bulk at a much lower fluence that in thin-films. If there was not enough He/D to form bubbles in-situ, why did they form after 30 d? Some other mechanism is occurring.

Concurrent He^+ Implantation and Self Ion Irradiation



Before Implantation/Irradiation

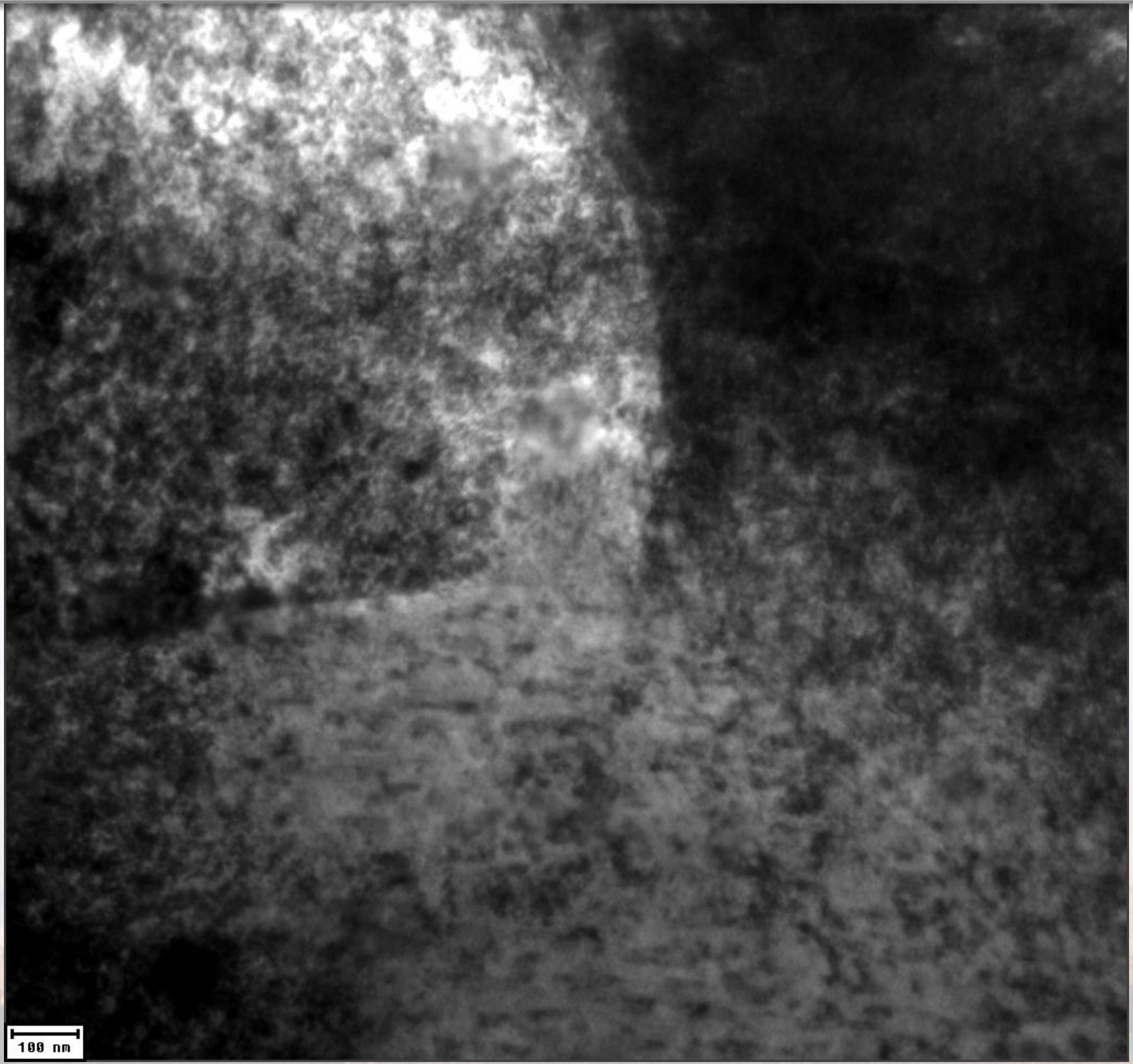


After Implantation/Irradiation

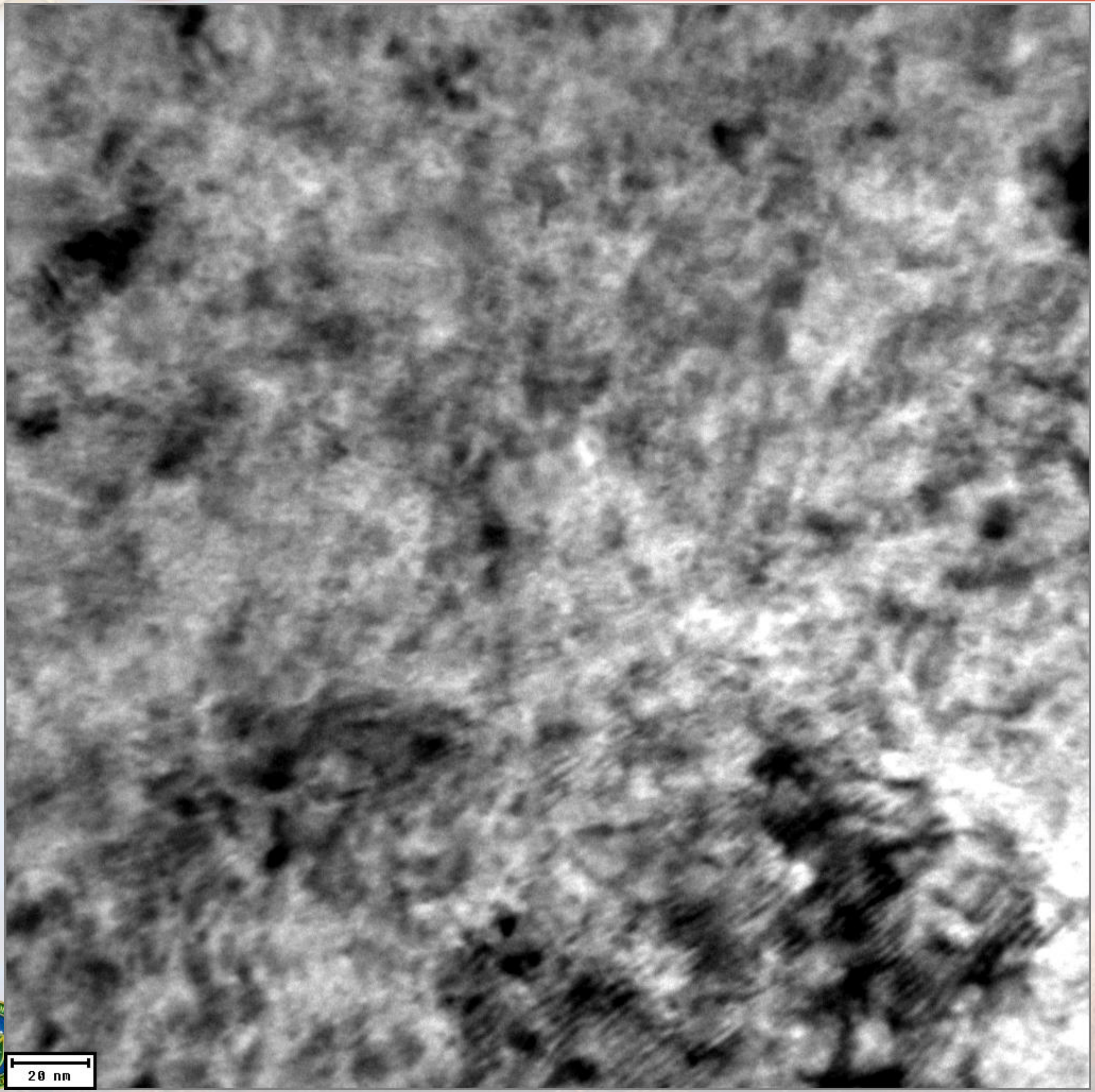
Damage, No Cavities

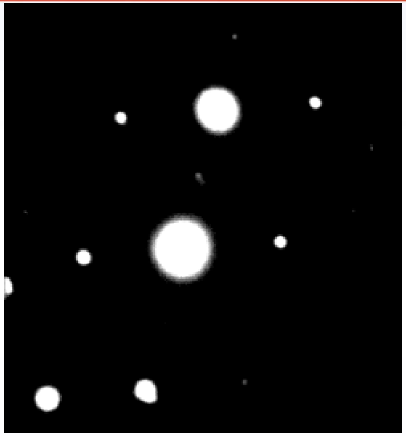
Concurrent D & He Implantation & Zr Irradiation

After Implantation/Irradiation Damage, No Cavities



Concurrent D & He Implantation & Zr Irradiation

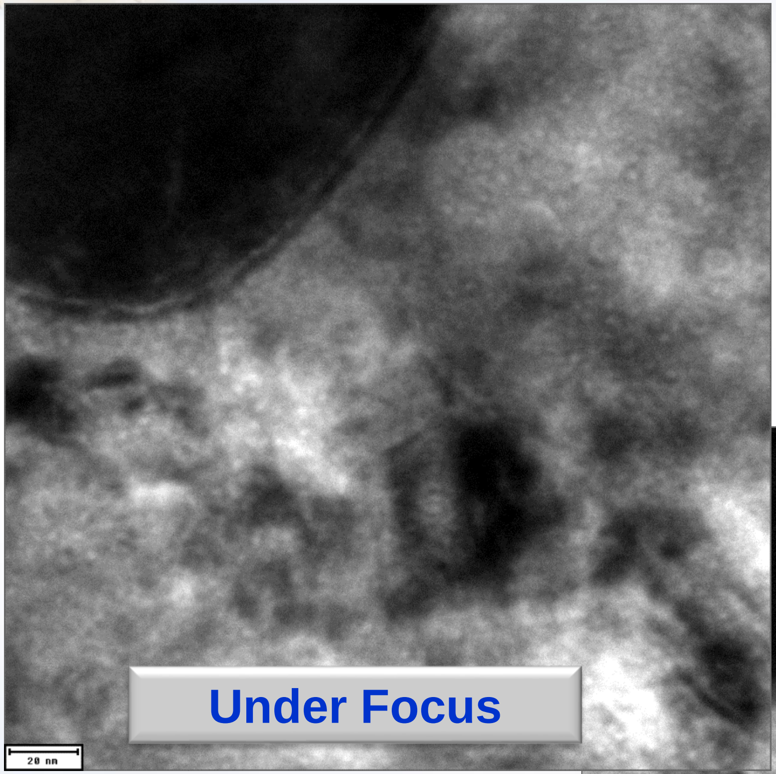


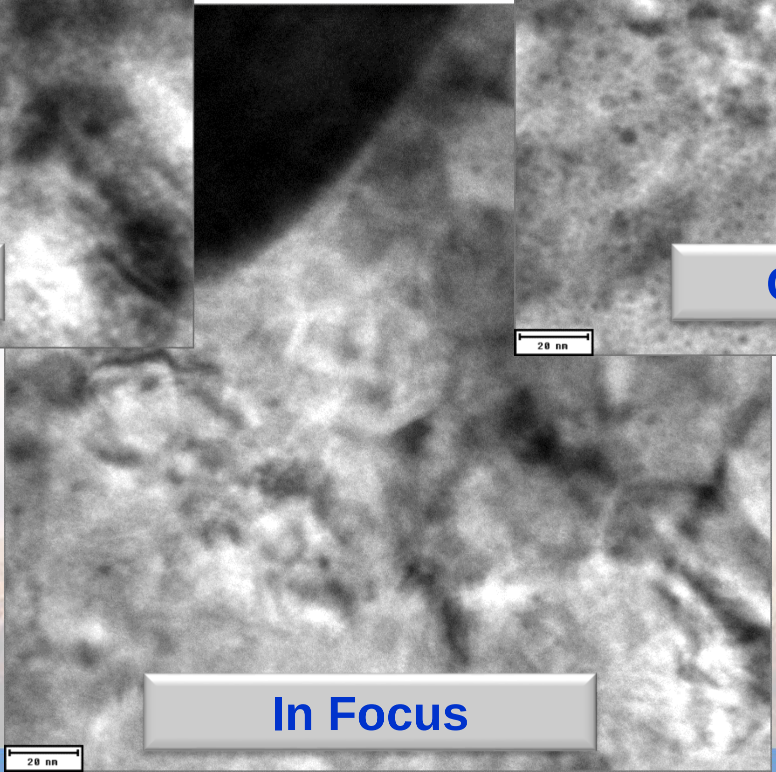


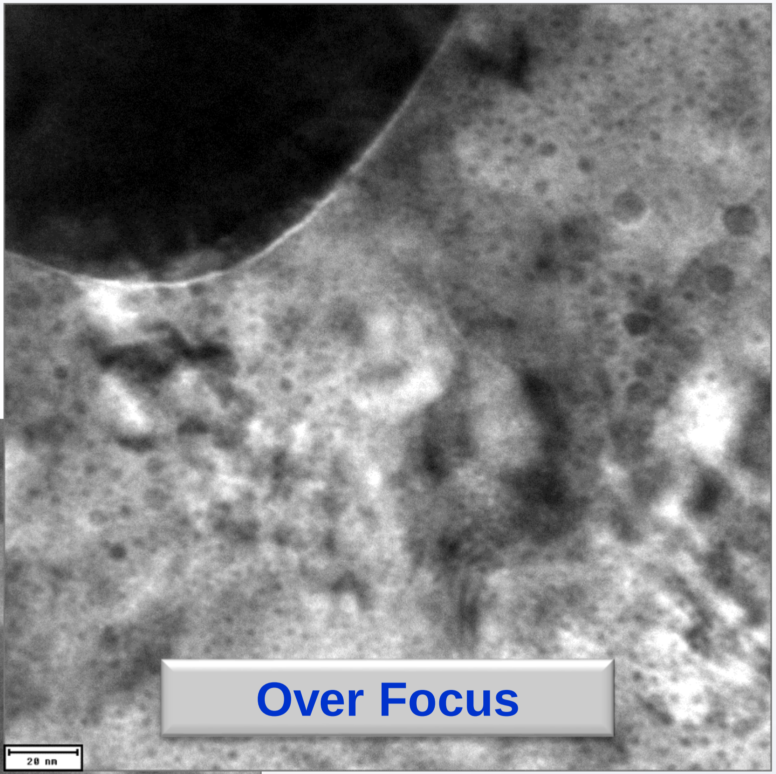
Two Beam

g = $1\bar{1}01$

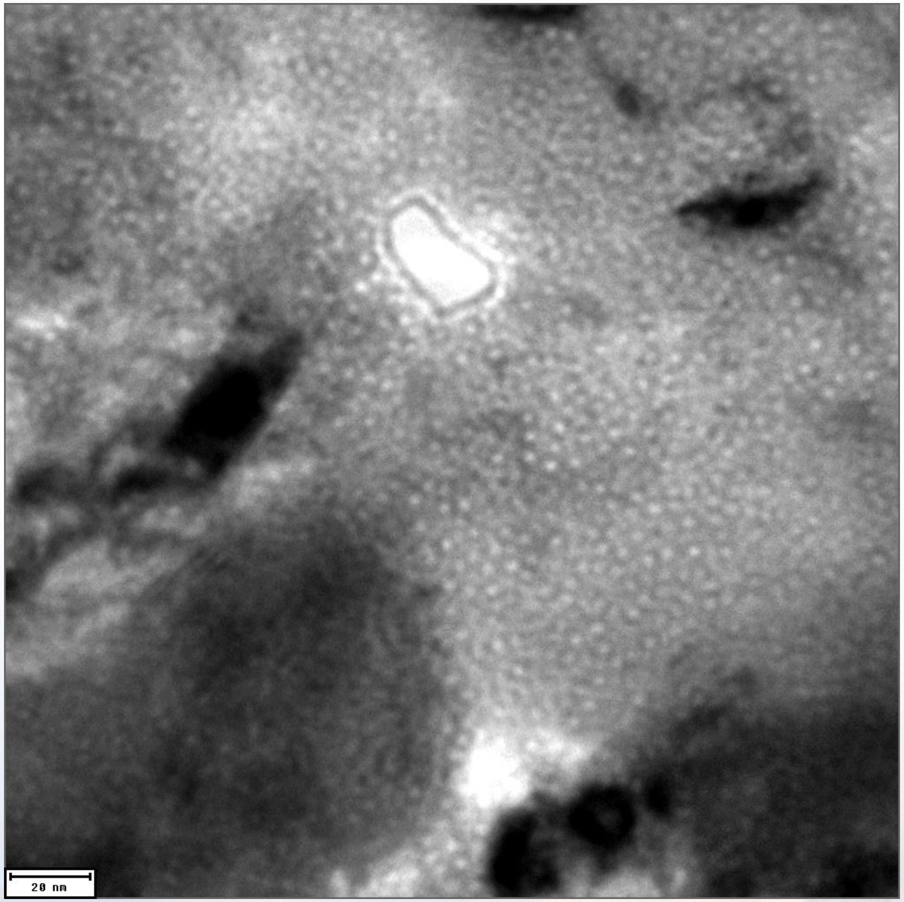
Through Focus Images: 30 Days Later



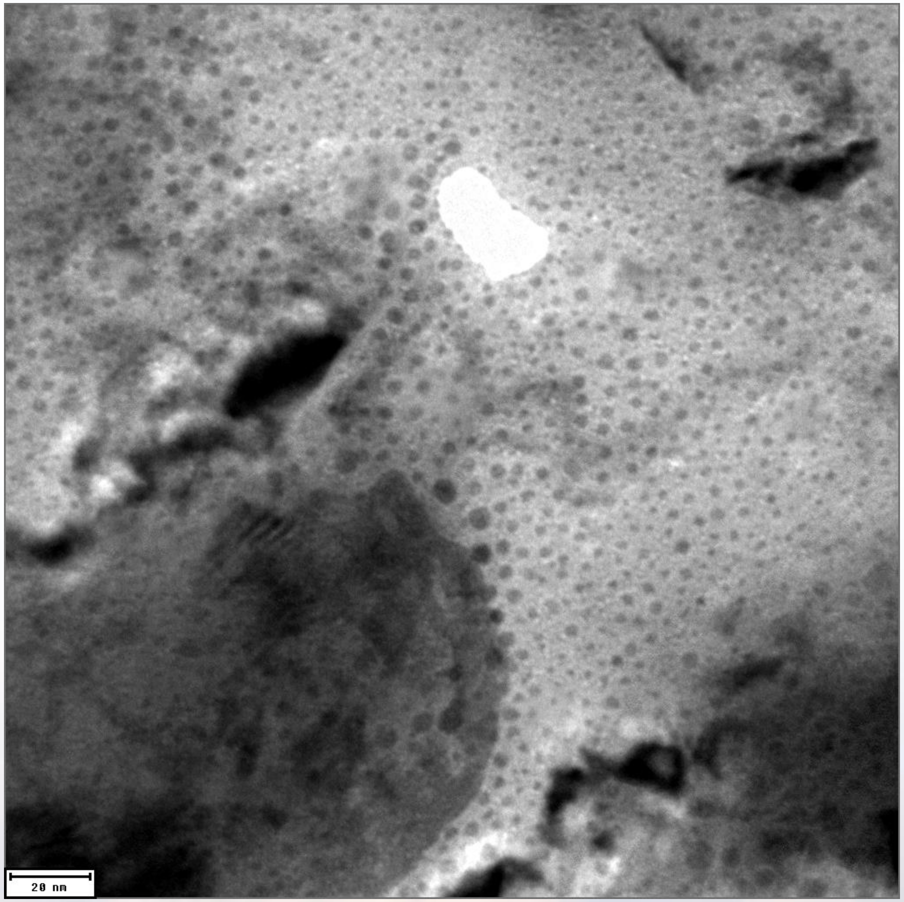




Through Focus Images: 30 Days Later

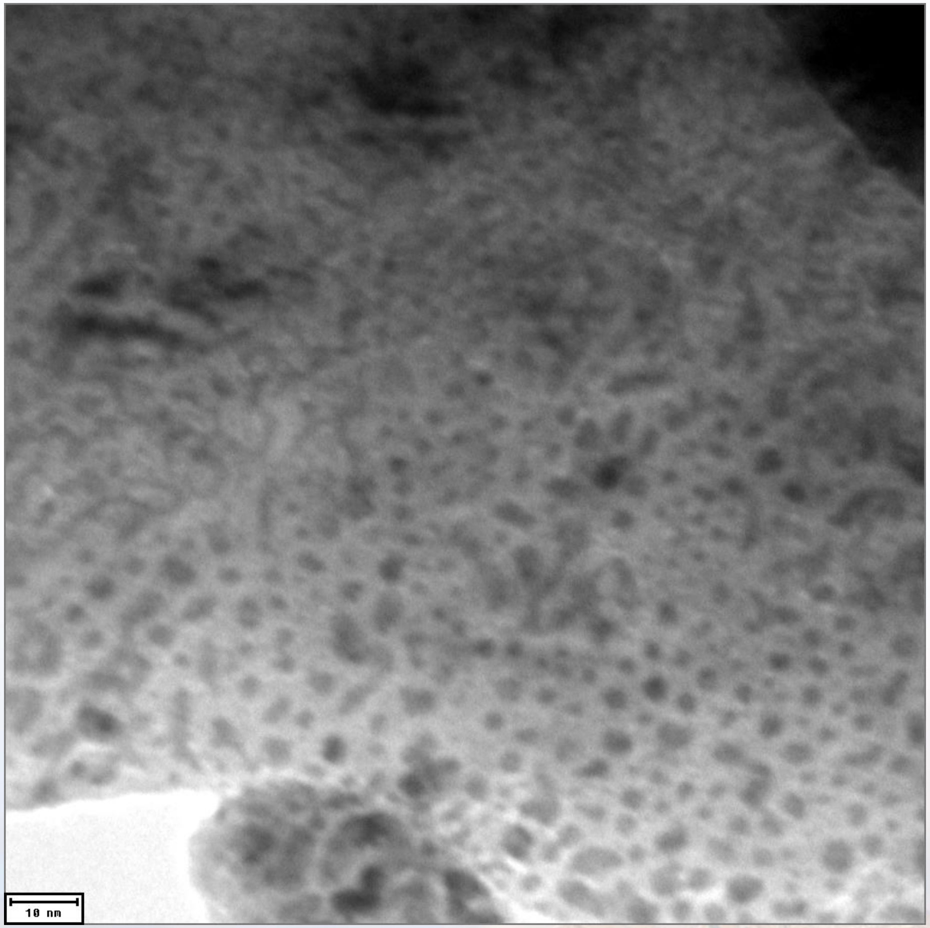


Under Focus

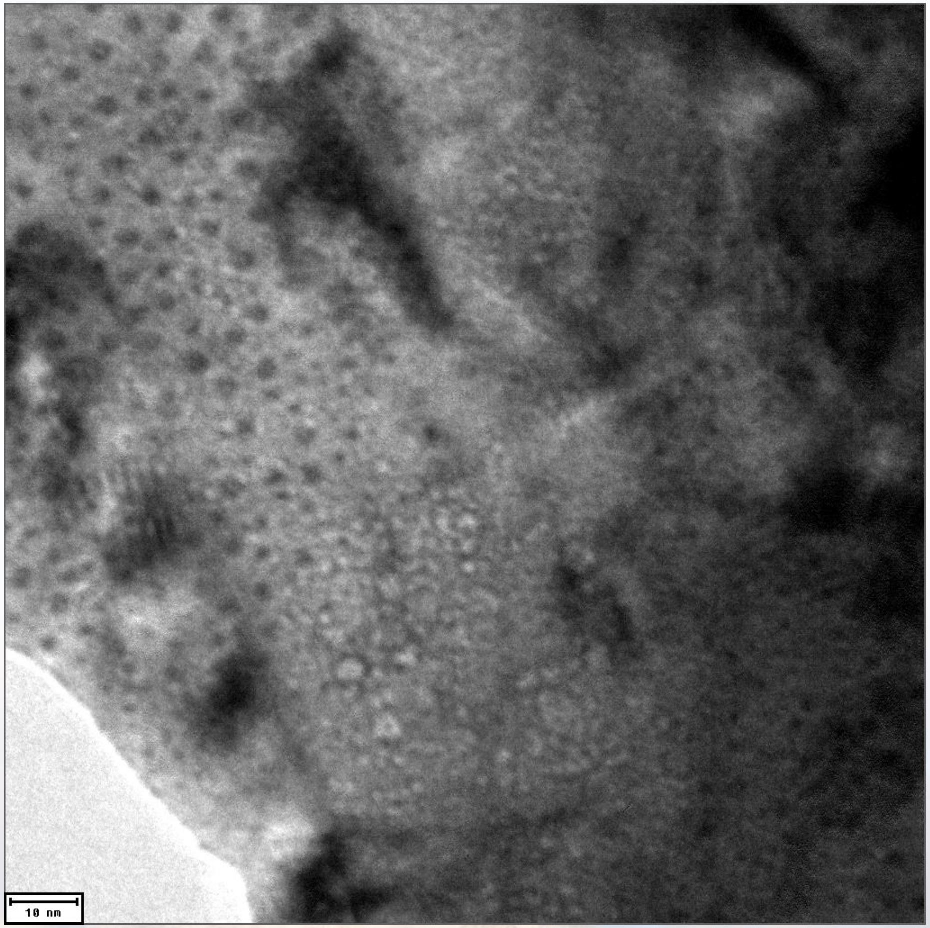


Over Focus

Surface Effects?

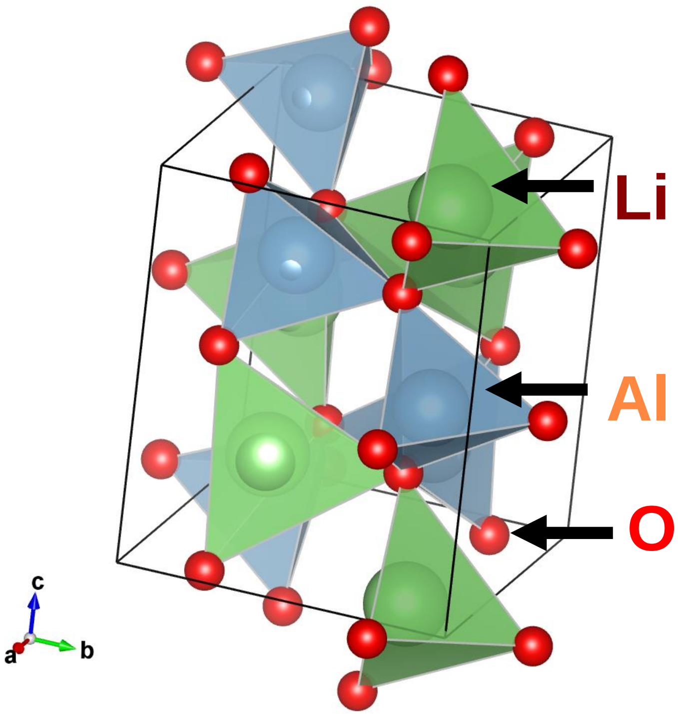


Under Focus



Under Focus

$LiAlO_2$ Background



γ–$LiAlO_2$ is tetragonal (space group: P 41 21 2)

$LiAlO_2$ transforms to /precipitates out $LiAl_5O_8$ (cubic spinel) under electron irradiation and some ion irradiation conditions.

How is $LiAlO_2$ used in the TPBAR?

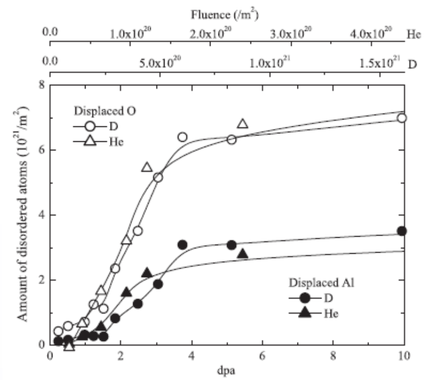
- $^6Li(n,\alpha)^3H$, emitting **^3H** (~2.75 MeV) and **^4He** (~2.05 MeV)
- ^3H β-decays to **^3He**
- Experiences displacive damage and gas accumulation at high temperature in reactor
- In addition to TPBAR, $LiAlO_2$ has been considered as a candidate for ^3H production in fusion reactors

Previous Work

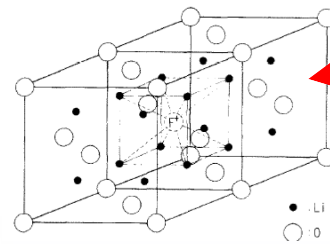
- Structural defects
  - Luo *et al* JNM 372 (2008) 53-58
- Volume swelling
  - Noda JNM 179-181 (1991) 37-41
- ^3H detrapping
  - Oyaidzu *et al* JNM 375 (2008) 1-7
- Gas diffusion and release
  - Raffray *et al* JNM 210 (1994) 143-160

Gas Behavior $LiAlO_2$ Background

- H isotopes are thought to trap in oxygen vacancies
  - ^2H release occurs at the same temperature as defect annealing in implanted LiAlO



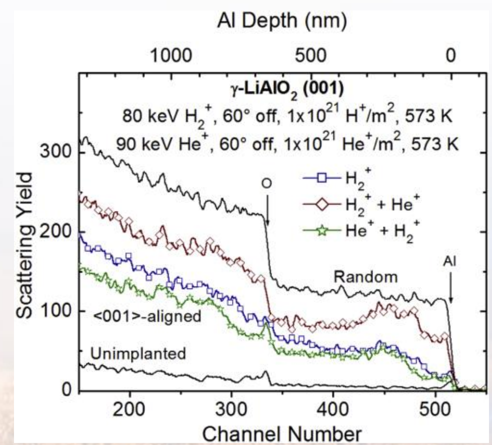
Katsui *et al.* NIMB 268 (2010) 2735-2739



Noda JNM 179-181 (1991) 37-41

Can be determined with luminescence

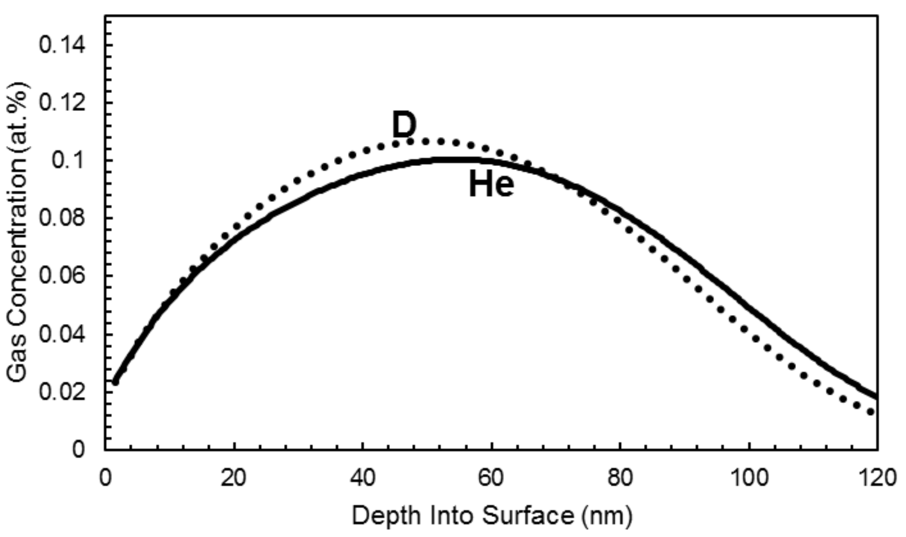
- Subsequent H + He implantation results in a higher ion channeling yield than subsequent He + H implantation
  - Synergistic effects between H isotopes and He are not well understood
  - Defect trapping of H isotopes in $LiAlO_2$ is not well understood

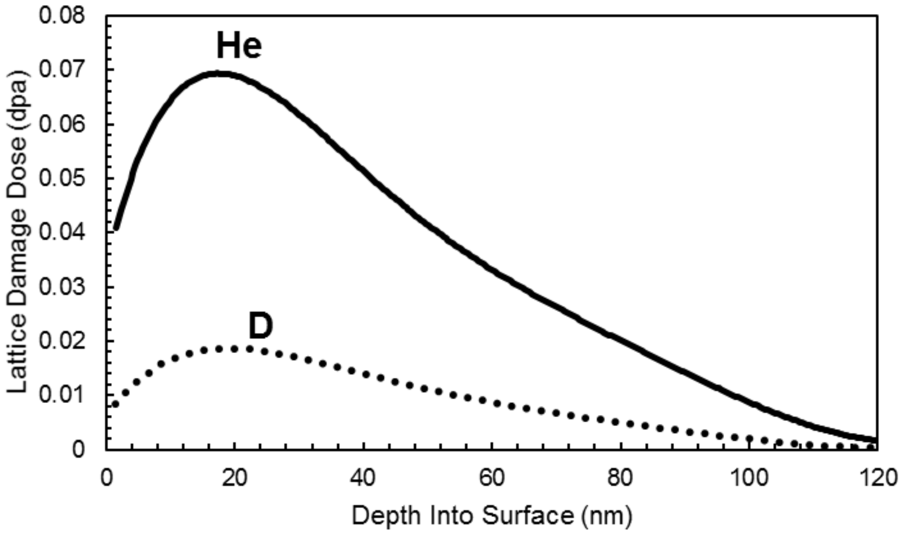


Jiang *et al* JNM 484 (2017) 374-381

Understanding Synergy Between Damage and Gas Bubble Formation

- Helium is known to form bubbles in materials, especially when defect traps are present
- May be a synergy between He and ^3H behavior, so we are planning dual beam implantations using ^2H to simulate ^3H
  - He bubbles may form and affect ^2H diffusion or trapping
- Bubble nucleation will be observed *in-situ* with the TEM

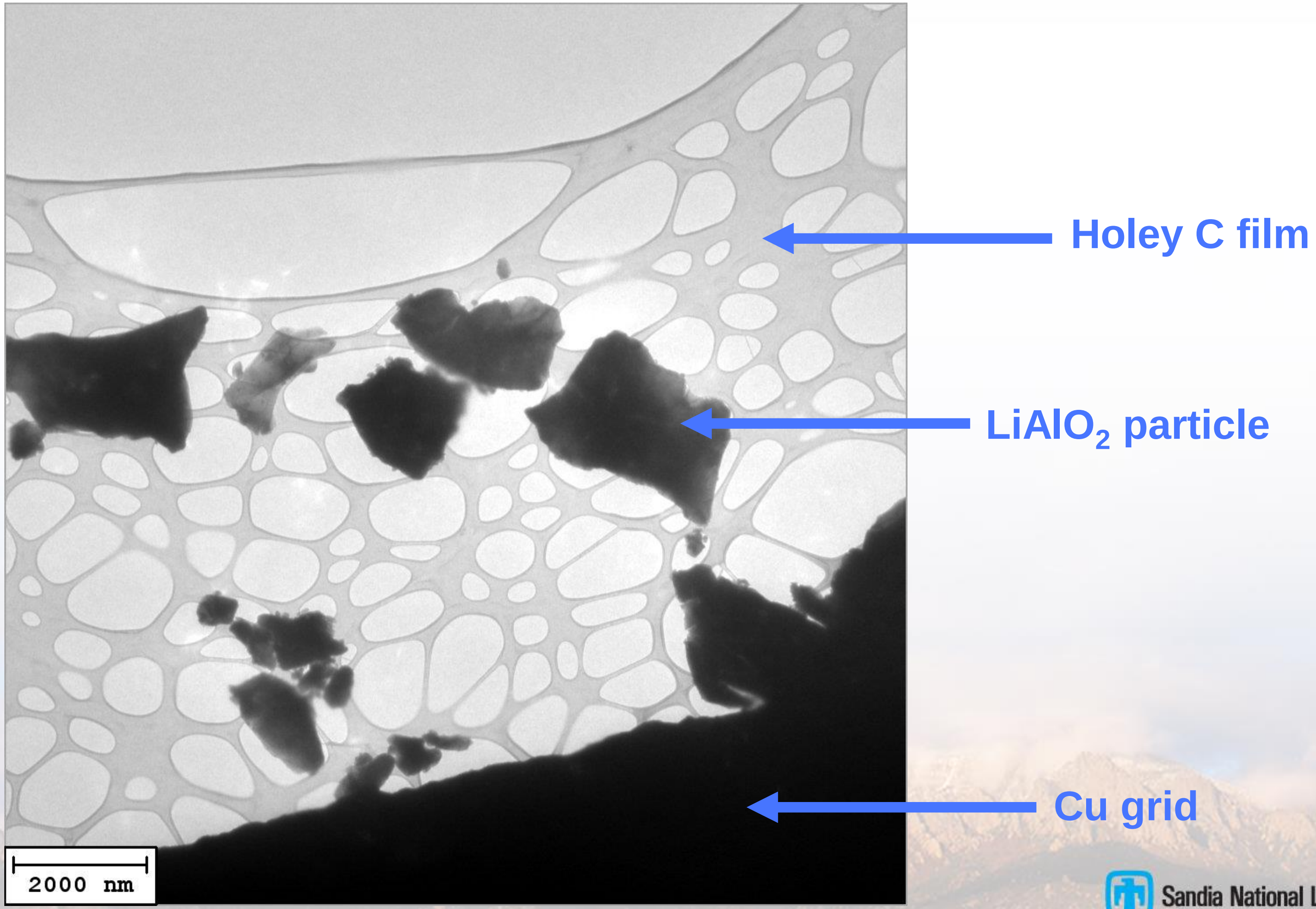




SRIM calculations of 10 keV He and 5 keV D implantation depth and damage

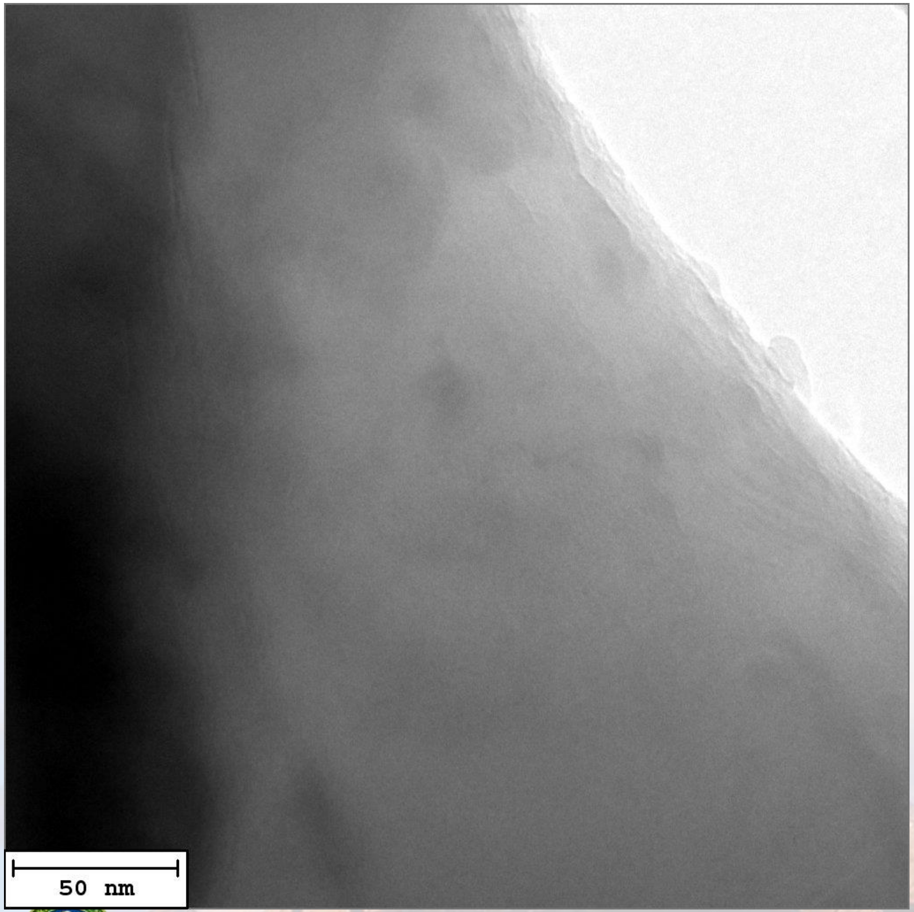
In-Progress TPBAR Work: $LiAlO_2$ Pellet

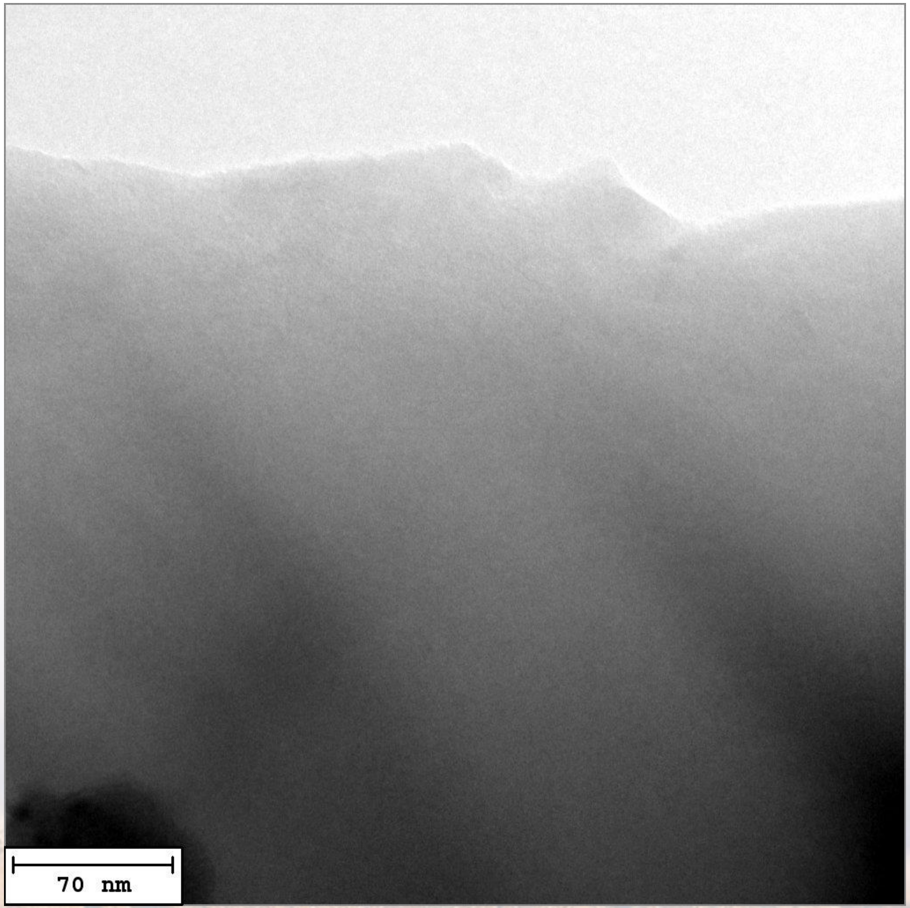
$LiAlO_2$ Powder Deposited on a TEM Grid



Current In-Progress TPBAR Work: $LiAlO_2$ Pellet

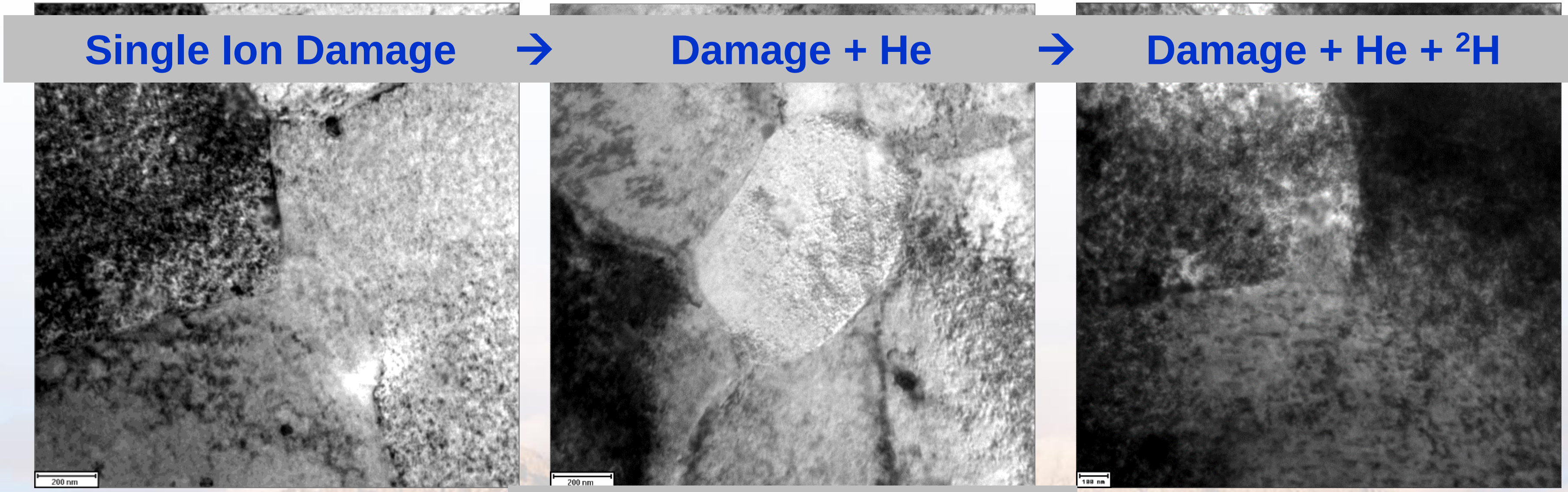
Some particles contain regions thin enough for TEM imaging of bubbles





Summary

- Synergistic effects between damage and gas accumulation are being simulated in TPBAR materials, in-situ, at the SNL I^3TEM facility, using heavy ion irradiation and D_2 + He implantation
- Aimed at understanding fundamental defect interactions that affect ^3H retention
- In-situ triple beam irradiations can be coupled with **HT TEM stage** for more accurate simulation of reactor conditions
- In Zr alloys, various irradiation/implantation conditions resulted in no bubble formation in-situ, but bubbles were observed in irradiated thin foils 30 days later



All at reactor temperature