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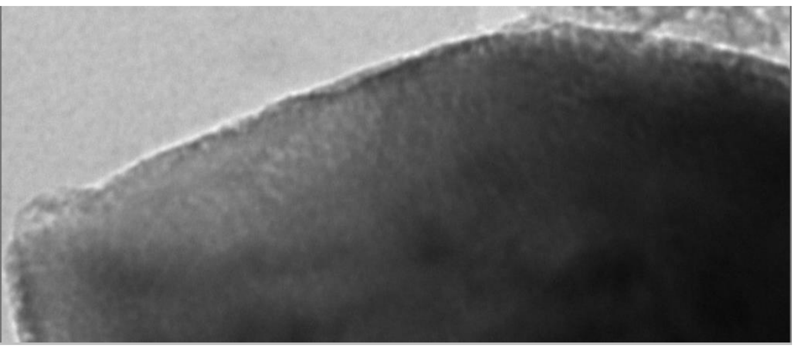
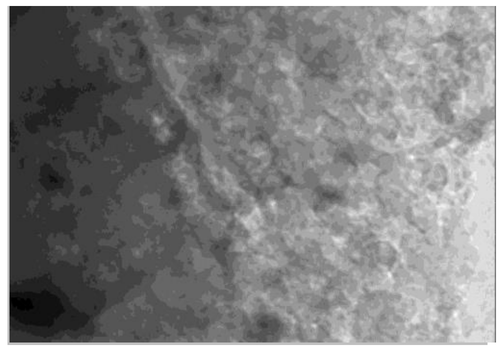
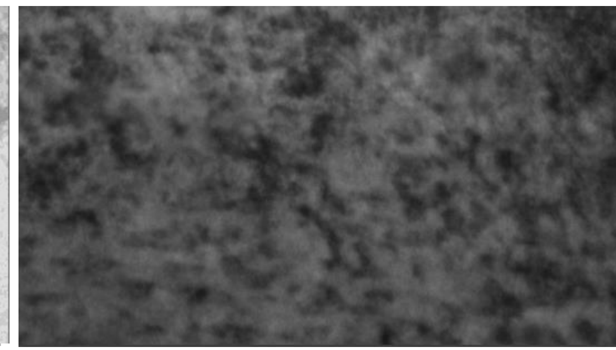

Simulations of Damage and Gas Accumulation in TPBAR Materials with In-situ Triple Ion Beam Irradiation TEM

Caitlin A. Taylor, Brittany Muntifering, David Senor, Clark Snow, and Khalid Hattar

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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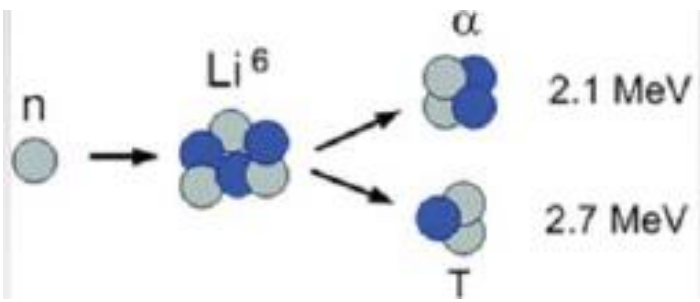
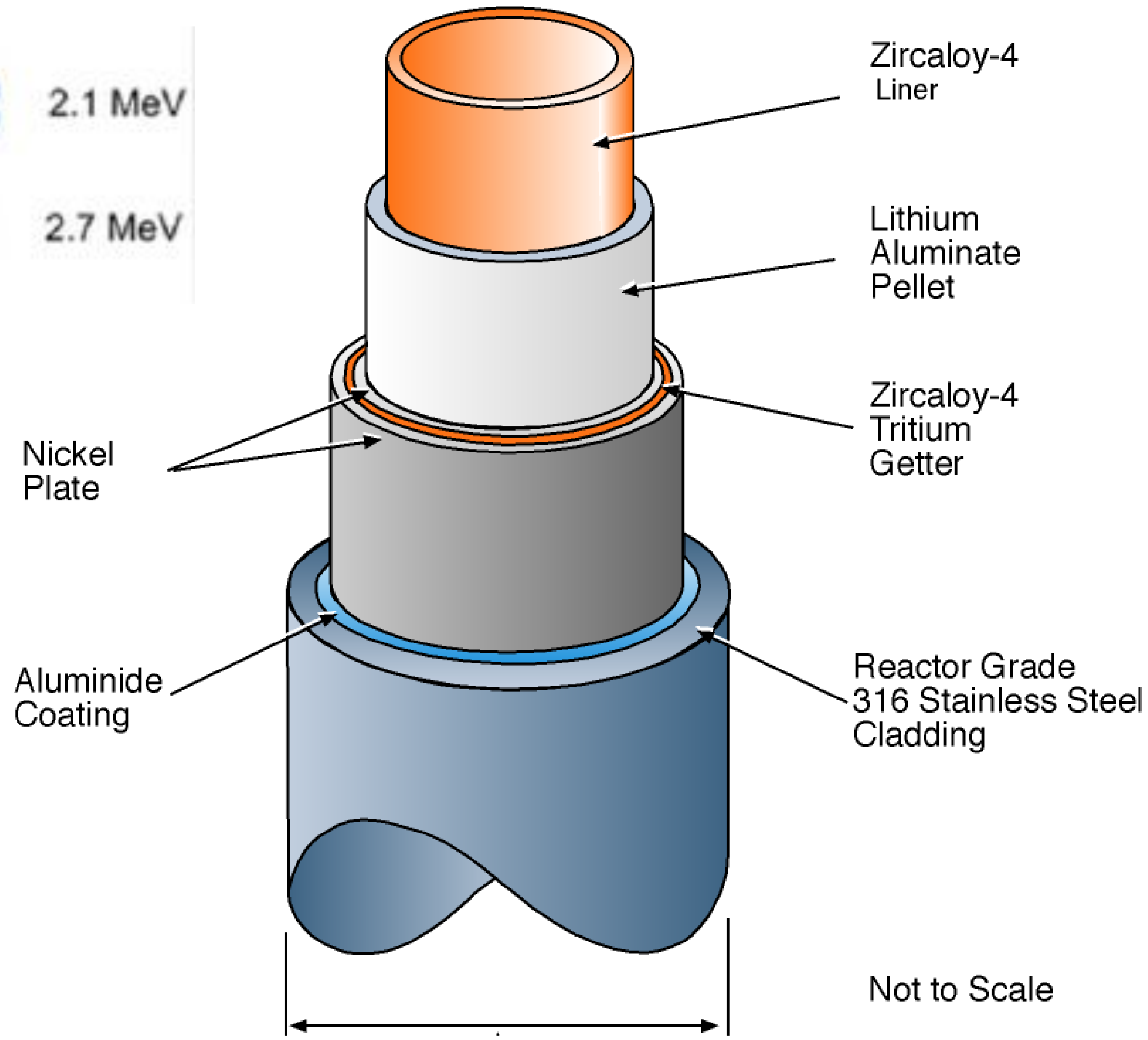
Tritium Producing Burnable Absorber Rod

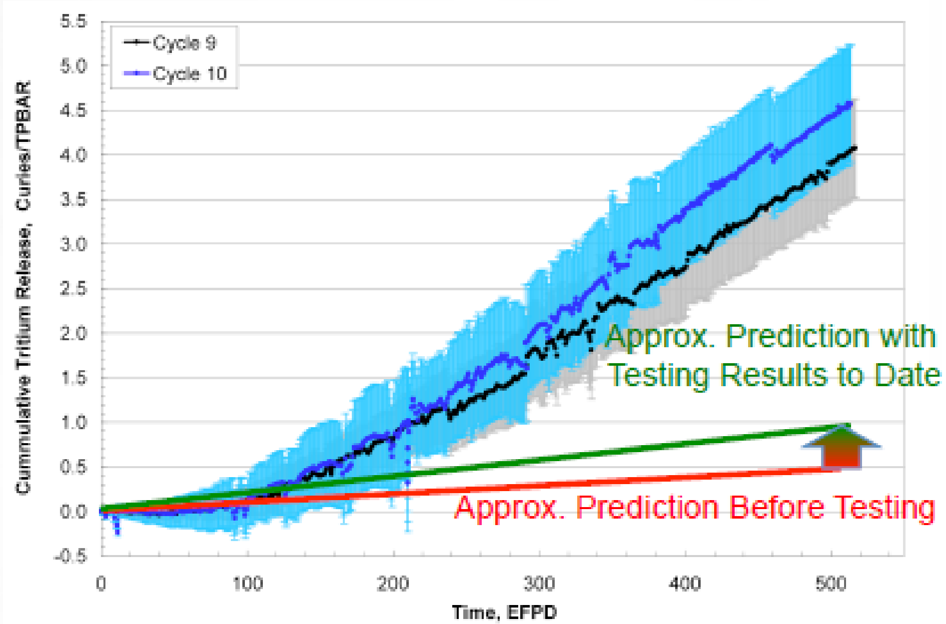


- Displacement Damage
- Helium Implantation
- Tritium Implantation
- Elevated Temperatures

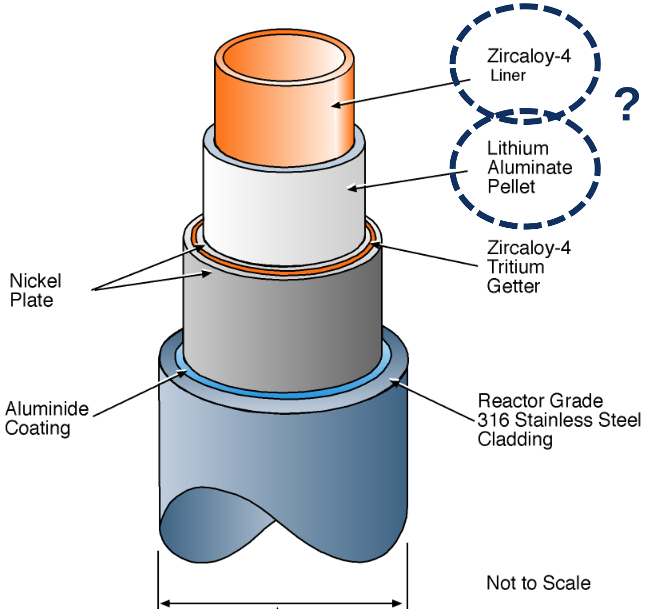


Understanding Tritium Permeation in TPBAR

- 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-He-defect interactions

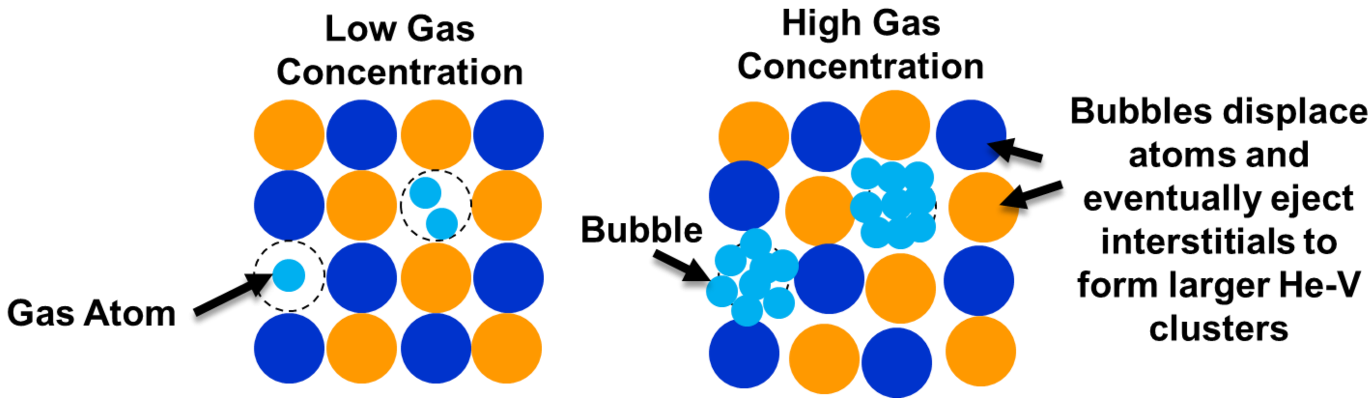


Burkes, Senor, Longoni and Johns, TFG Meeting 2016, Rochester, NY



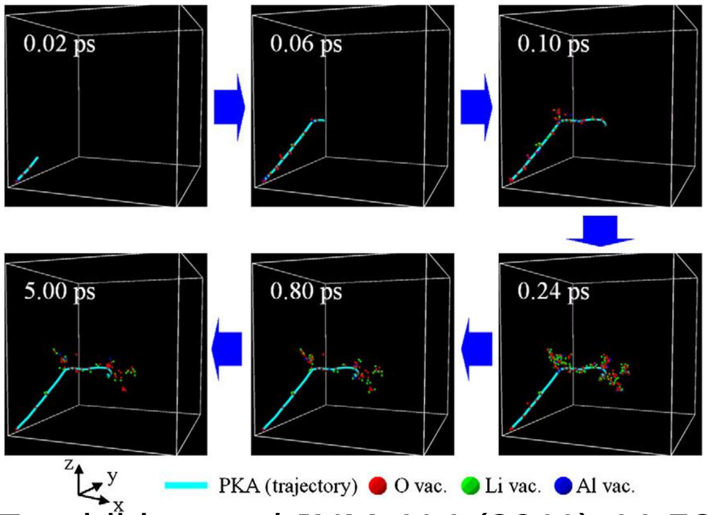
Bubbles May Affect ^{3}H Release

- Bubbles form due to He trapping in lattice defects



- Neutron irradiation produces displacement cascades, providing complex defect structures for He or ^{3}H to trap in.

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

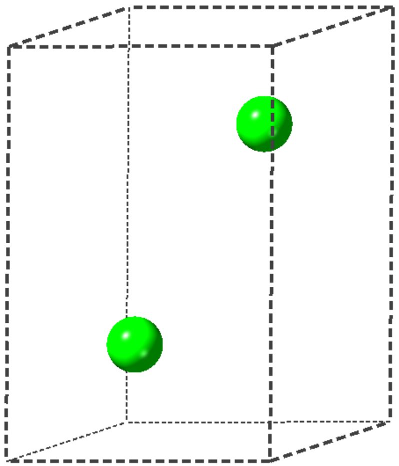


Tsuchihira *et al* JNM 414 (2011) 44-52

Zircaloy Background

What is Zircaloy?

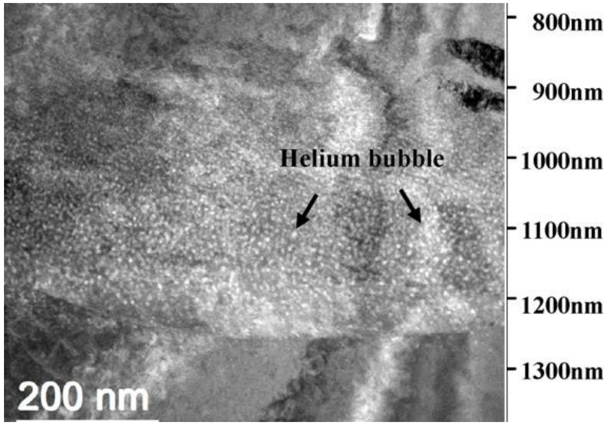
- Zircaloy-2: predominantly used as fuel cladding for BWRs
  - α-Zr, 1.5% Sn, 0.15% Fe, 0.1% Cr, 0.05% Ni
- Zircaloy-4: Removed the Ni and increased Fe content for less H uptake in certain reactor conditions
  - α-Zr, 1.5% Sn, 0.2% Fe, 0.1% Cr
- Zr-Nb alloys (e.g. Zirlo) are also common
- α-Zr has a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



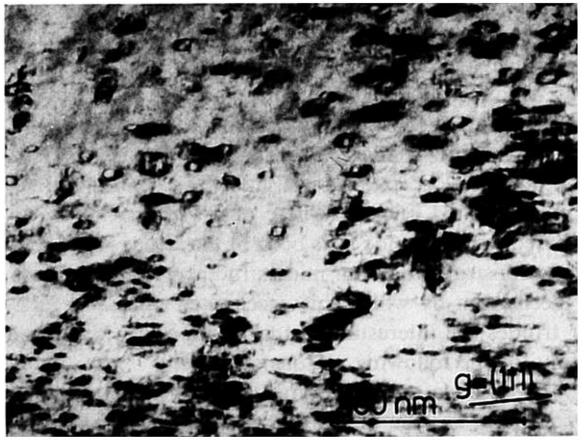
Crystal Structure of α-Zr (HCP)

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

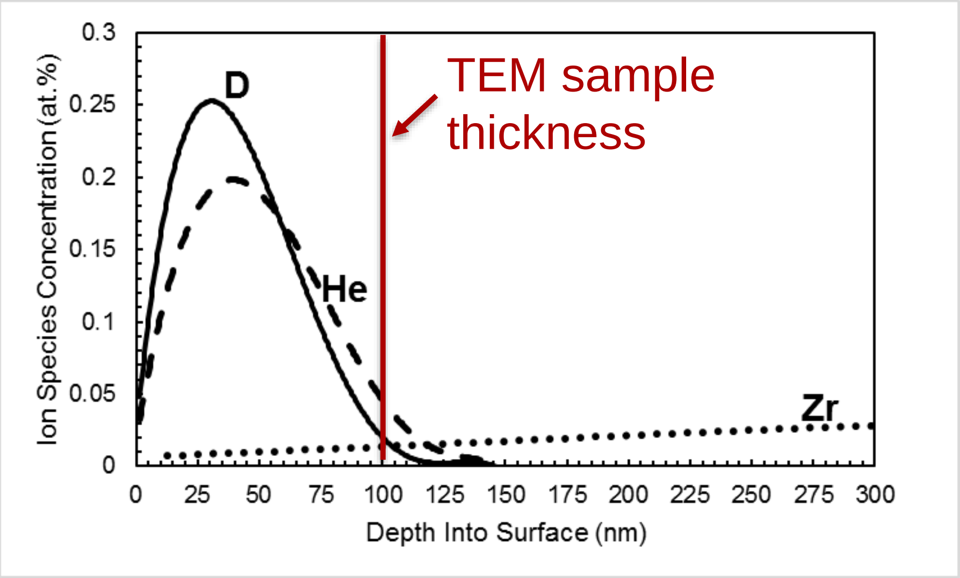


TEM of Zr tritide after 325d
Schober et al JNM 141-143 (1986) 453-457

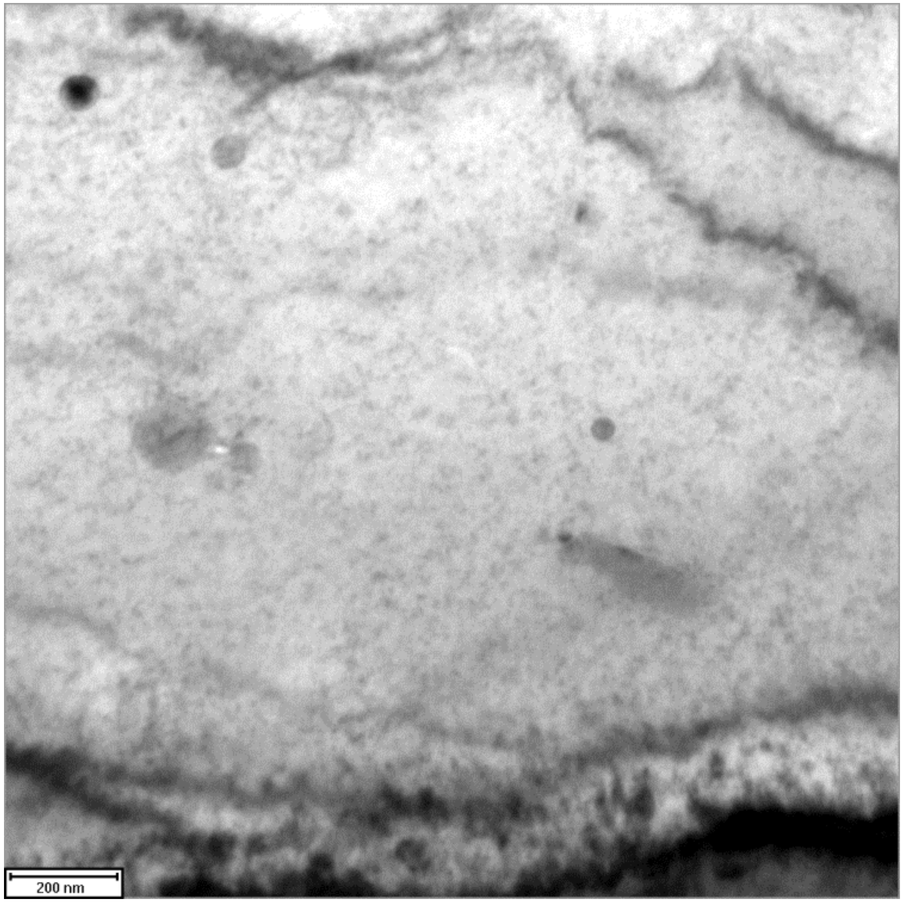
Zr alloy in-situ ion irradiation parameters

- Samples were prepared by electropolishing zirconium alloy samples (mostly ZIRLO)
- Several sets of irradiations done at 310°C, including:
  - 10 keV He → simulates He accumulation from ^{6}Li transmutation and ^{3}H decay
  - 10 keV He + 5 keV D + **3 MeV Zr**→ simulates gas build-up + displacement cascades
- SRIM, a Monte Carlo based program for simulating the number of displacements produced by an ion, was used to predict damage dose and concentration profiles.
- These preliminary experiments were run overnight and the exact gas concentrations/damage doses are not all known

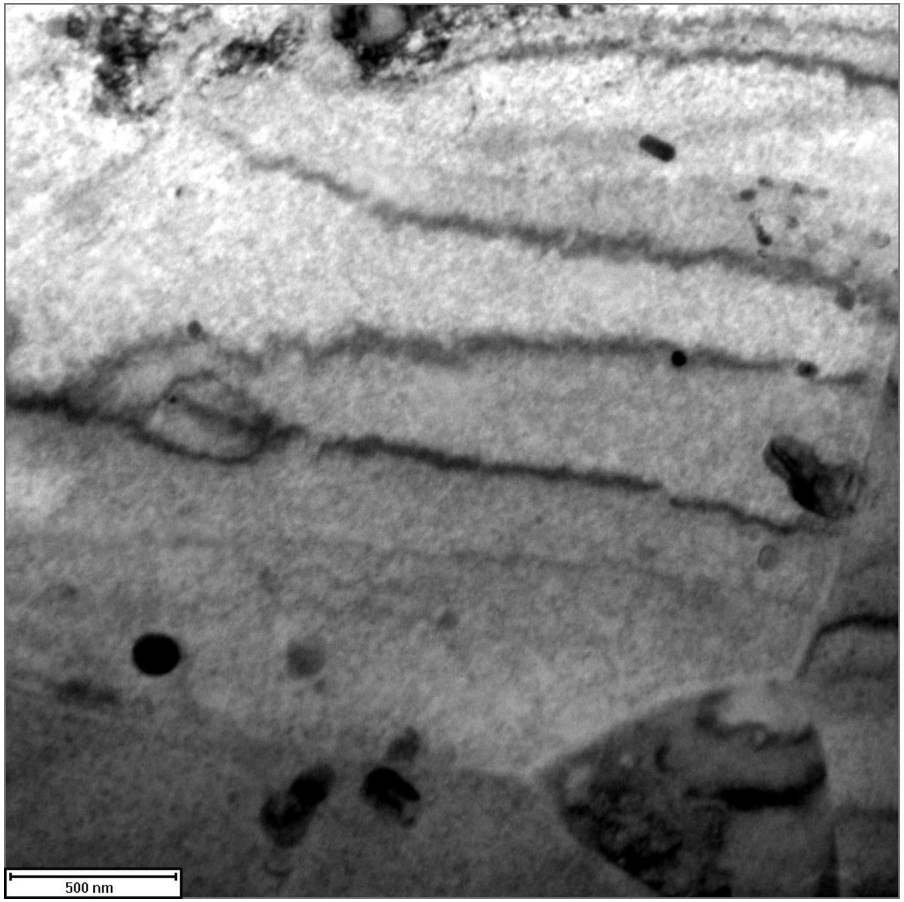
He and D profiles are implanted within the TEM sample, while most Zr passes through the sample, leaving only cascade damage.



10 keV He^{+} Implantation at 310°C



After Implantation.
Damage, No Cavities.

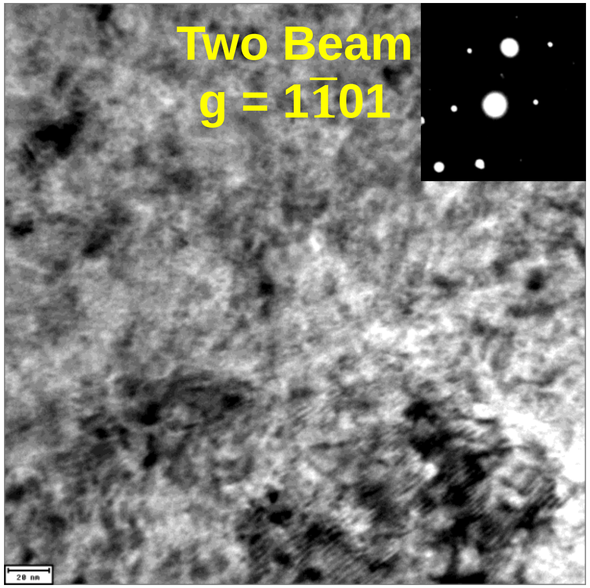


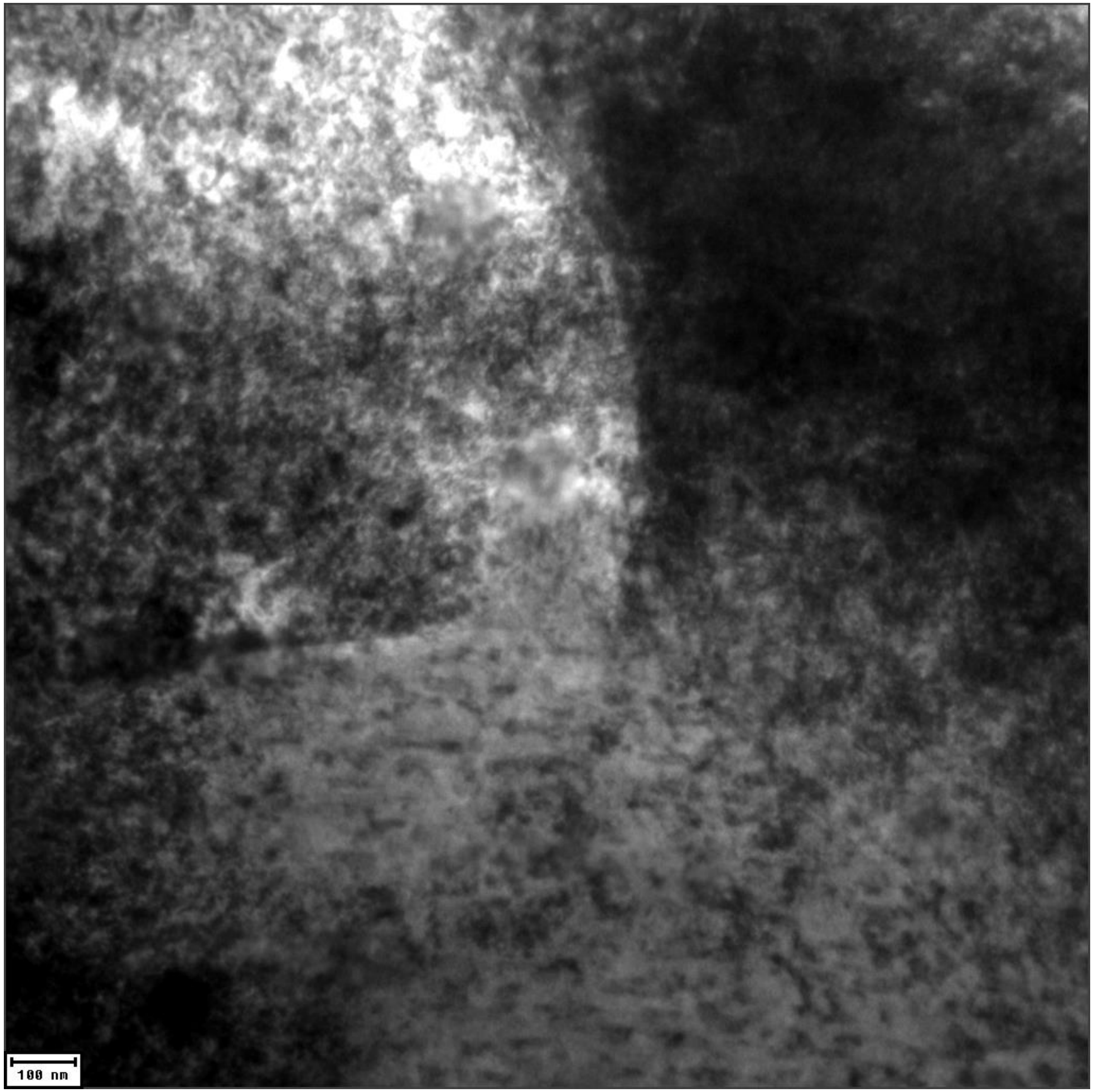
Still no cavities after subsequent irradiation with 3 MeV Zr.

Concurrent D & He Implantation & Zr Irradiation

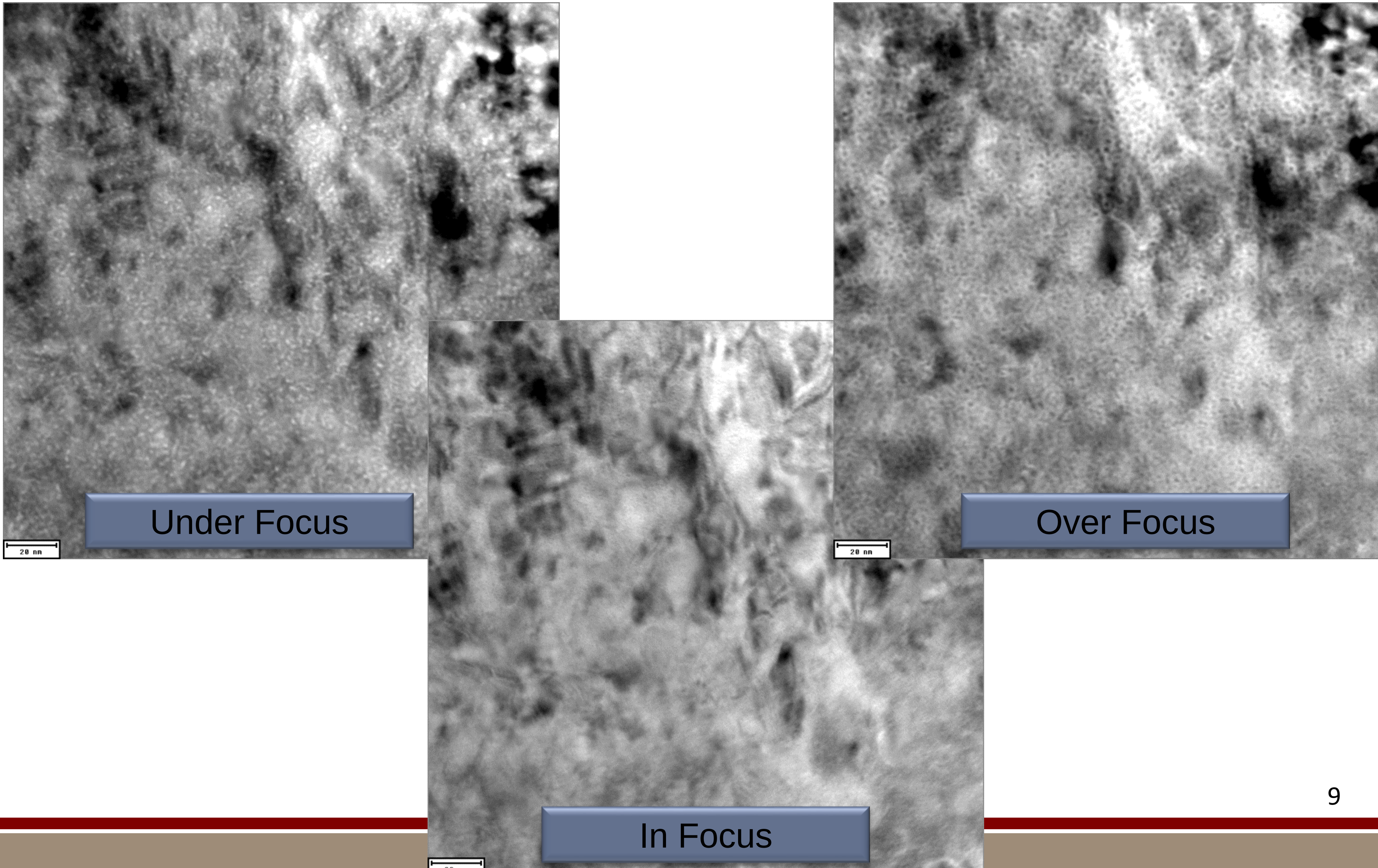
<u>After triple beam irradiation</u>

- Very dense, complex defect structure
- No visible cavities
- Fuzzy defects difficult to characterize

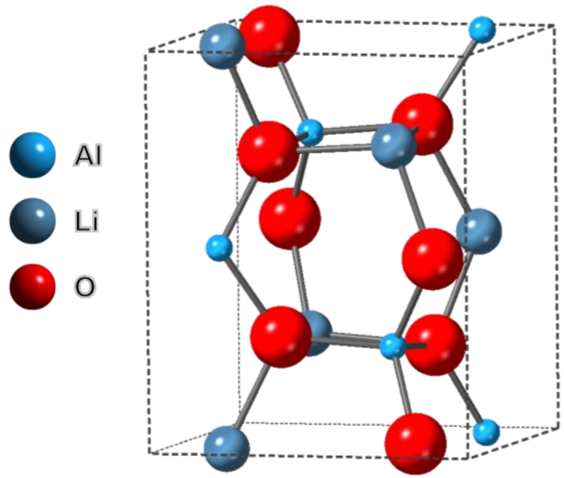




Cavities were observed in He implanted Zr alloy samples 30 days after irradiation



$LiAlO_2$ Background

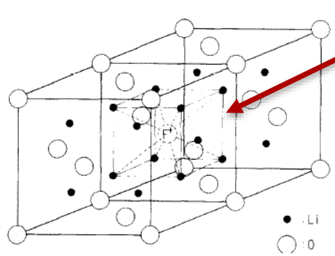


γ–$LiAlO_2$ is tetragonal
(space group: P 41 21 2)

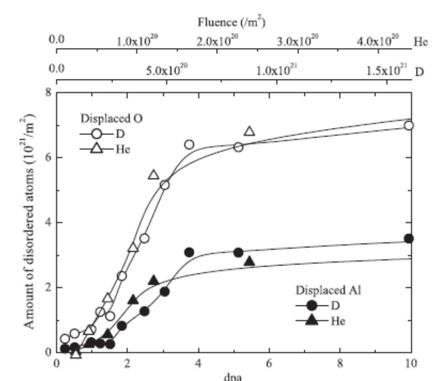
Previous Work

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

- H isotopes are thought to trap in oxygen vacancies
  - ^{2}H release occurs at the same temperature as defect annealing in implanted $LiAlO_2$



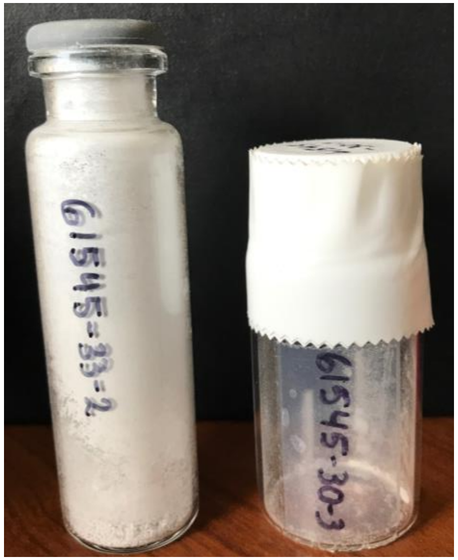
Noda JNM 179-181 (1991) 37-41

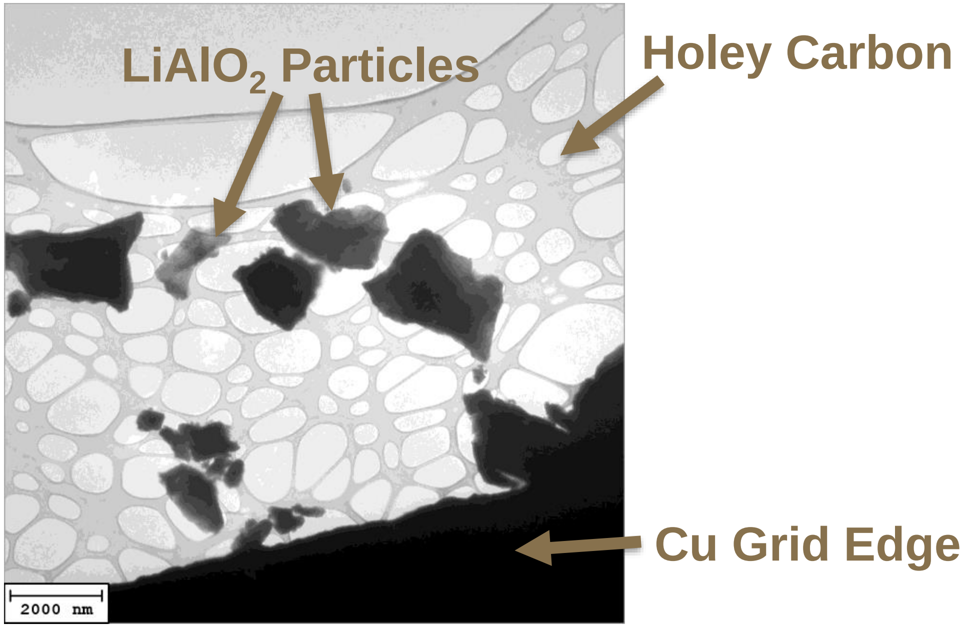


Katsui *et al.* NIMB 268 (2010) 2735-2739

$LiAlO_2$ in-situ ion irradiation parameters

- Powders were drop-cast onto TEM grids





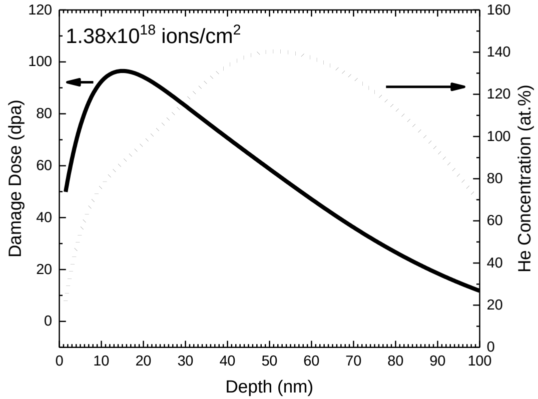
- Samples were heated to **310°C** using Hummingbird HT stage
- Three sets of irradiations:
  - 10 keV He → simulates He accumulation from ^{6}Li transmutation and ^{3}H decay
  - 10 keV He + 5 keV D → simulates He and ^{3}H interaction
  - 10 keV He + 5 keV D + **1.7 MeV Au**→ simulates gas build-up + displacement cascades

$LiAlO_2$ in-situ ion irradiation parameters

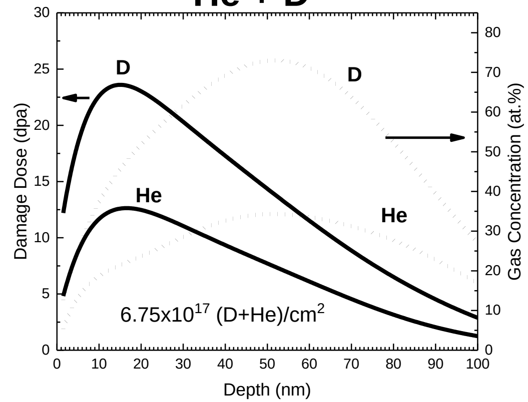
Most He/D diffuses out of thin film immediately

- SRIM Calculations

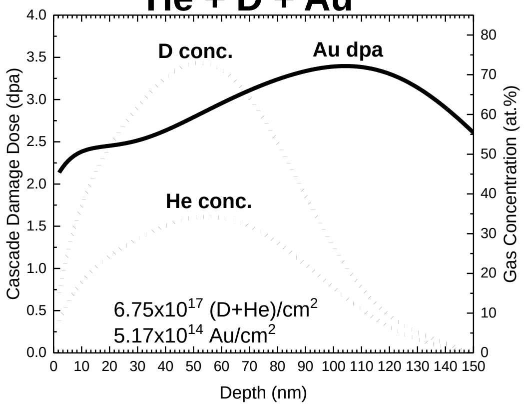
10 keV He



He + D



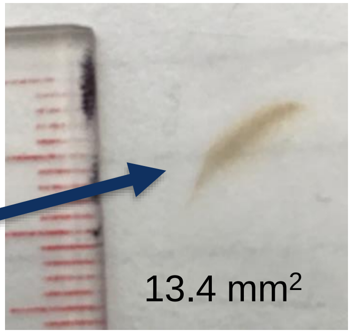
He + D + Au



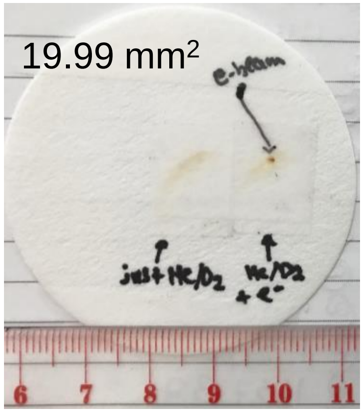
- Burn spots were used (1) to confirm that electron beam spot overlaps ion beam spot and (2) to determine the ion beam irradiation area.

He & D beams are shaped like a comet due to TEM objective lens

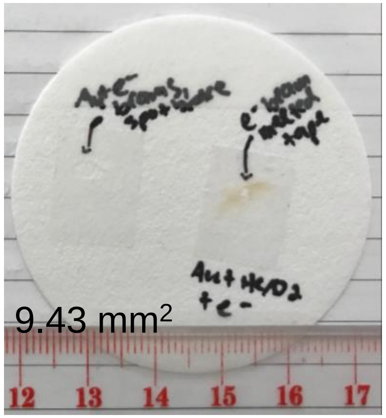
He



He + D

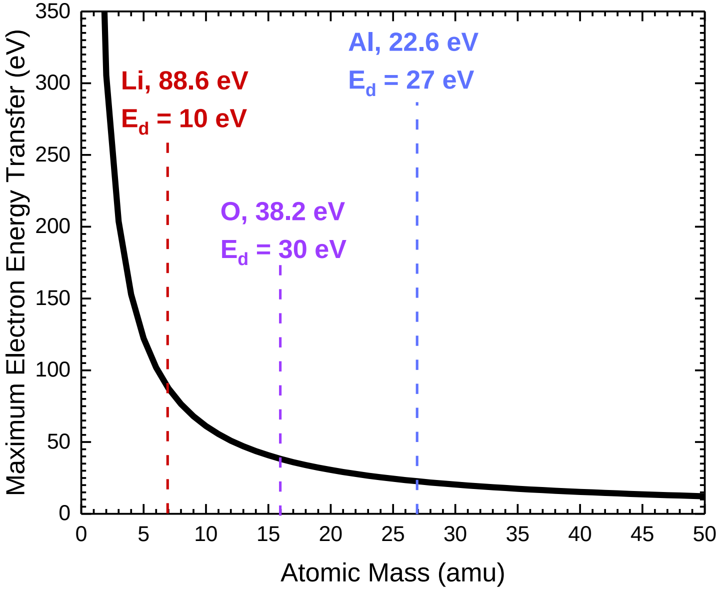


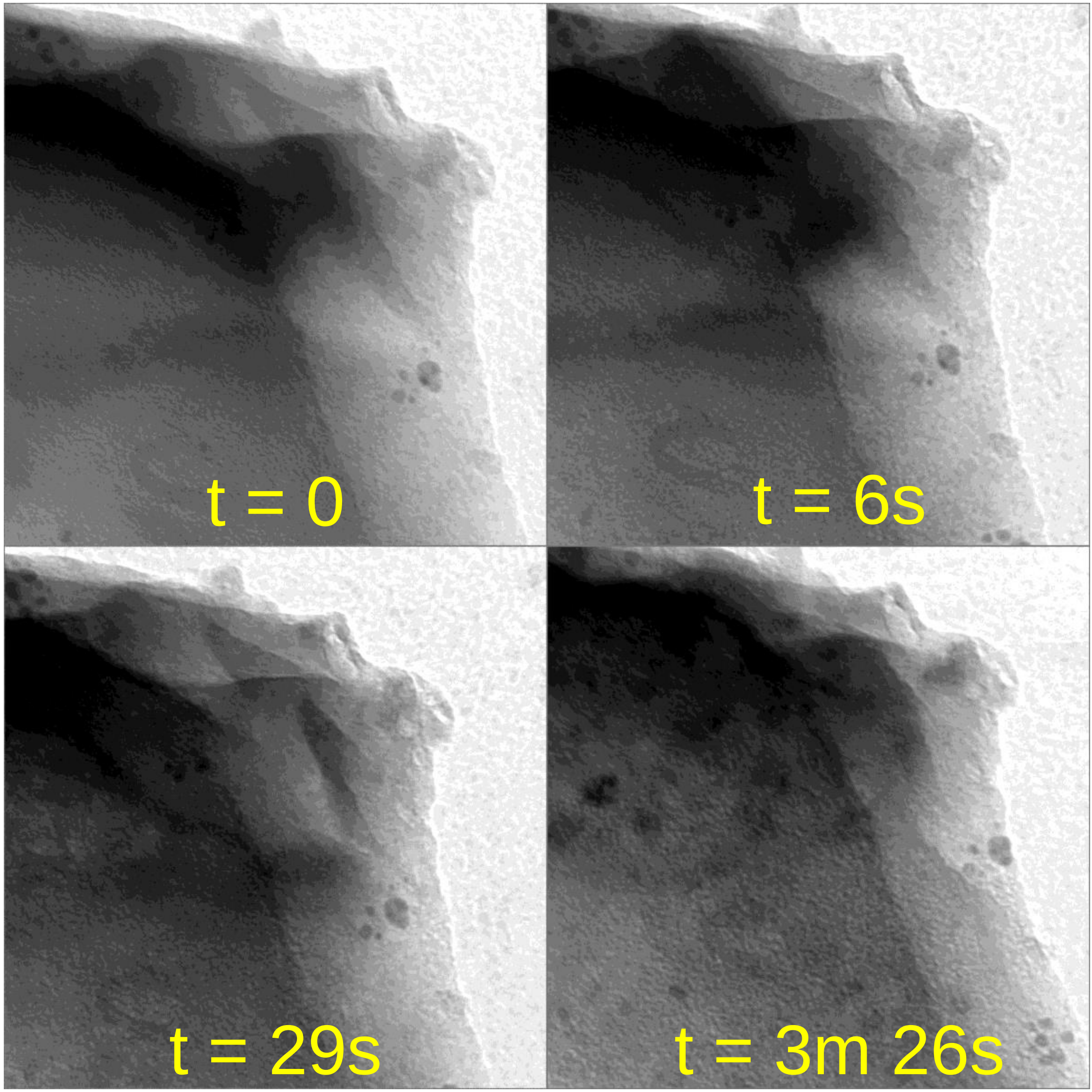
He + D + Au



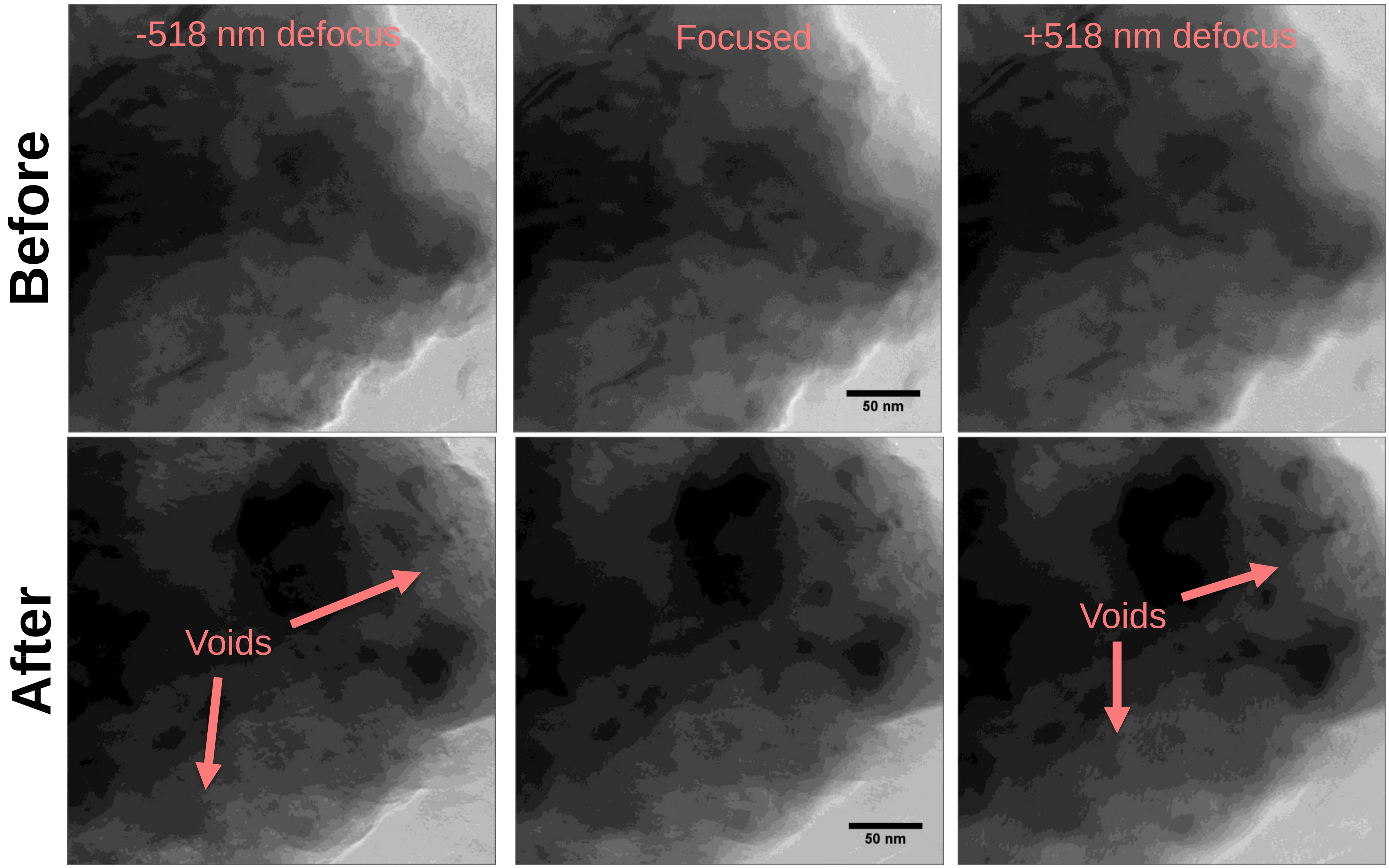
Electron beam induced void growth

- 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





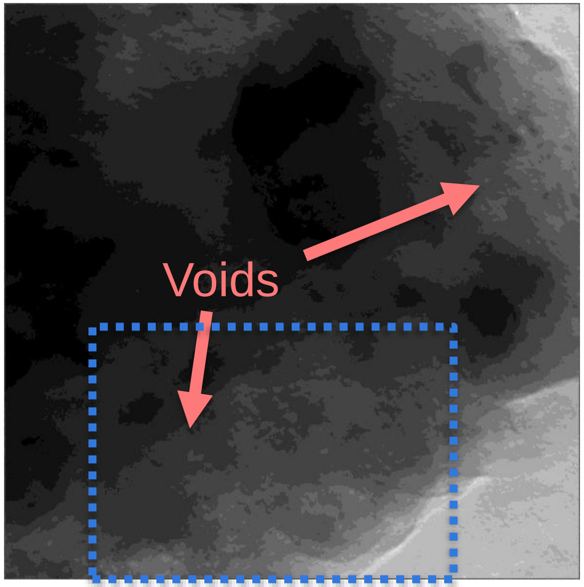
In-situ He implantation @ 310°C



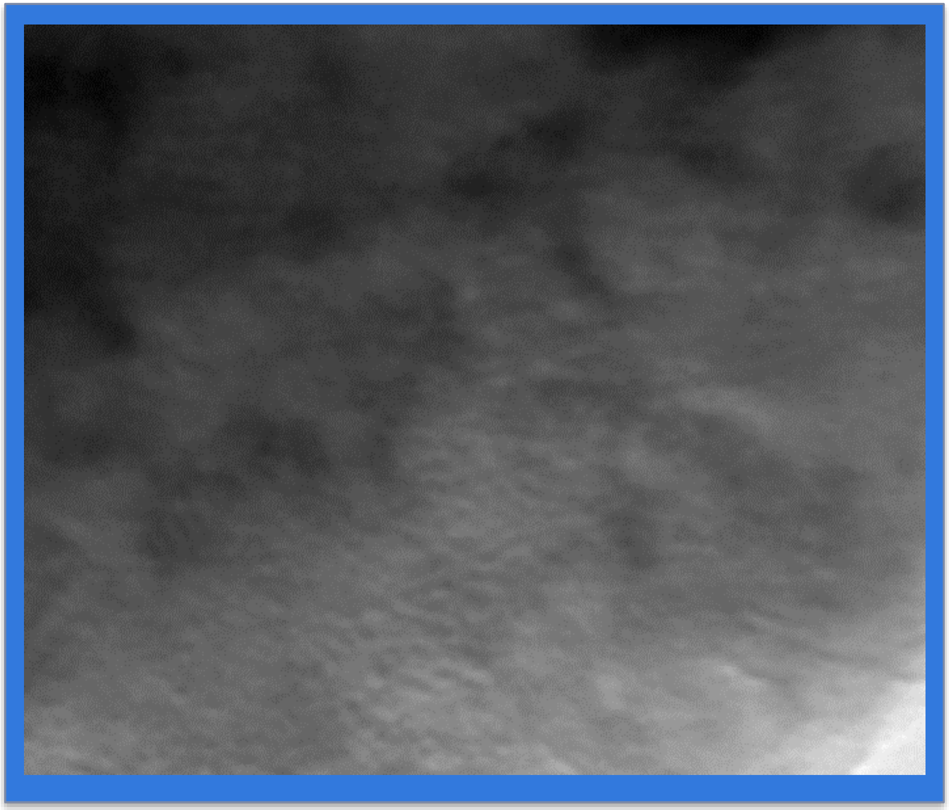
In-situ He implantation @ 310°C

- Each frame = 1 min of irradiation
- Because the voids are difficult to see in powders, I paused the video at a few points to show overfocus images
- Electron beam on for most of experiment
- Bubbles formed after ~13 min (1.5×10^{17} He/cm^2)

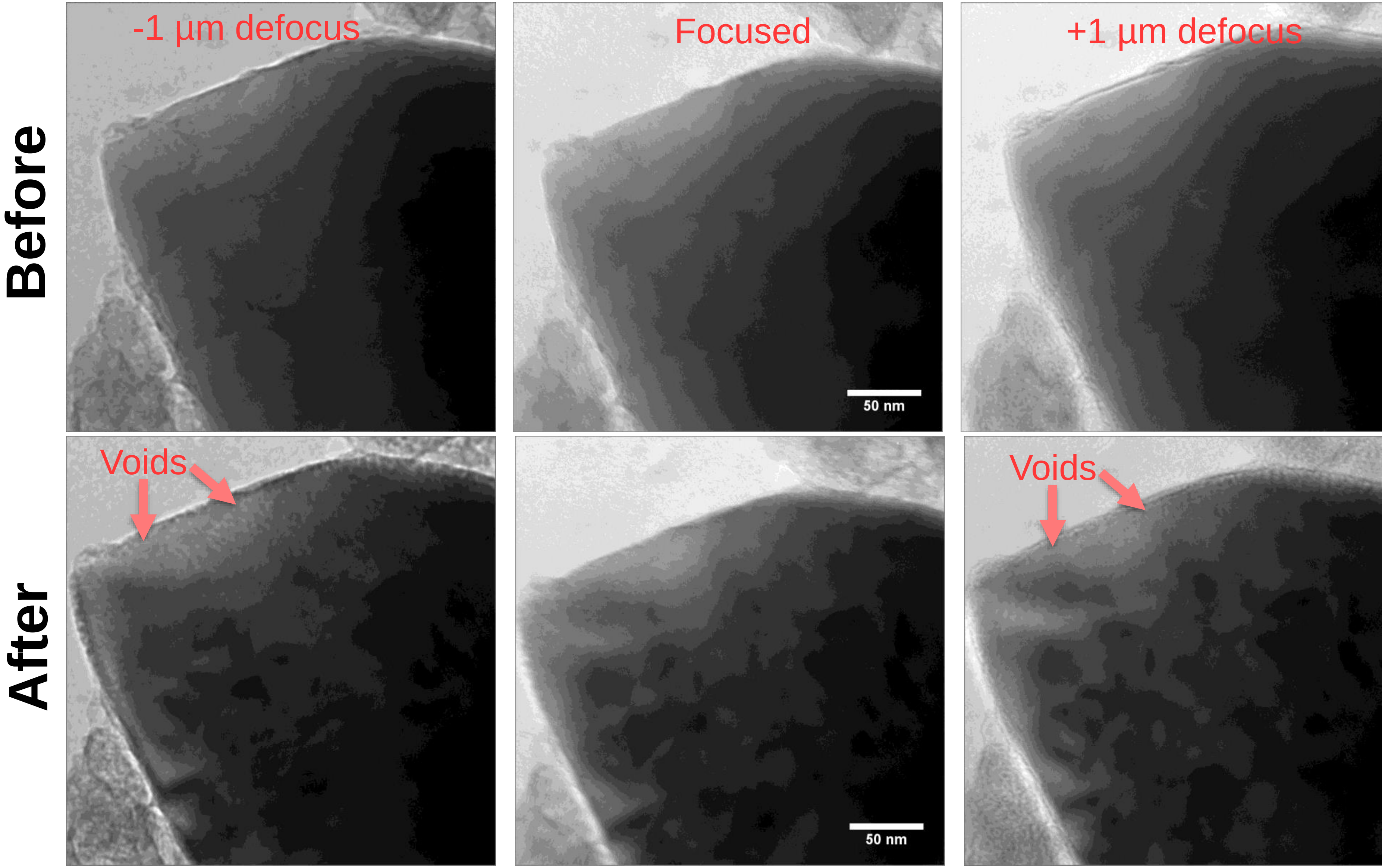
After Irradiation



In-situ Video



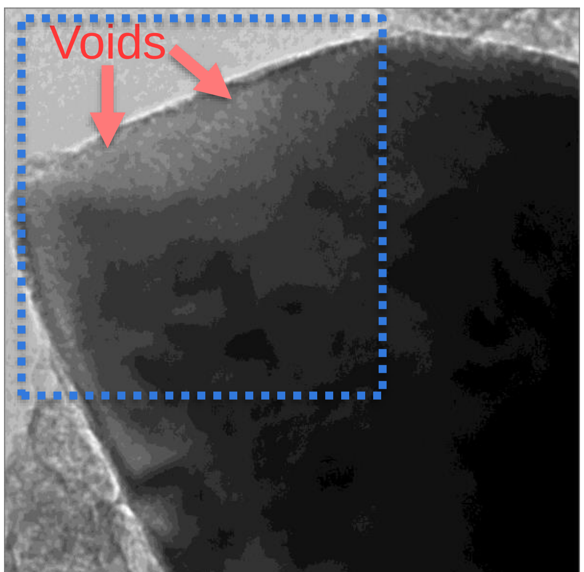
In-situ He + D irradiation @ 310°C



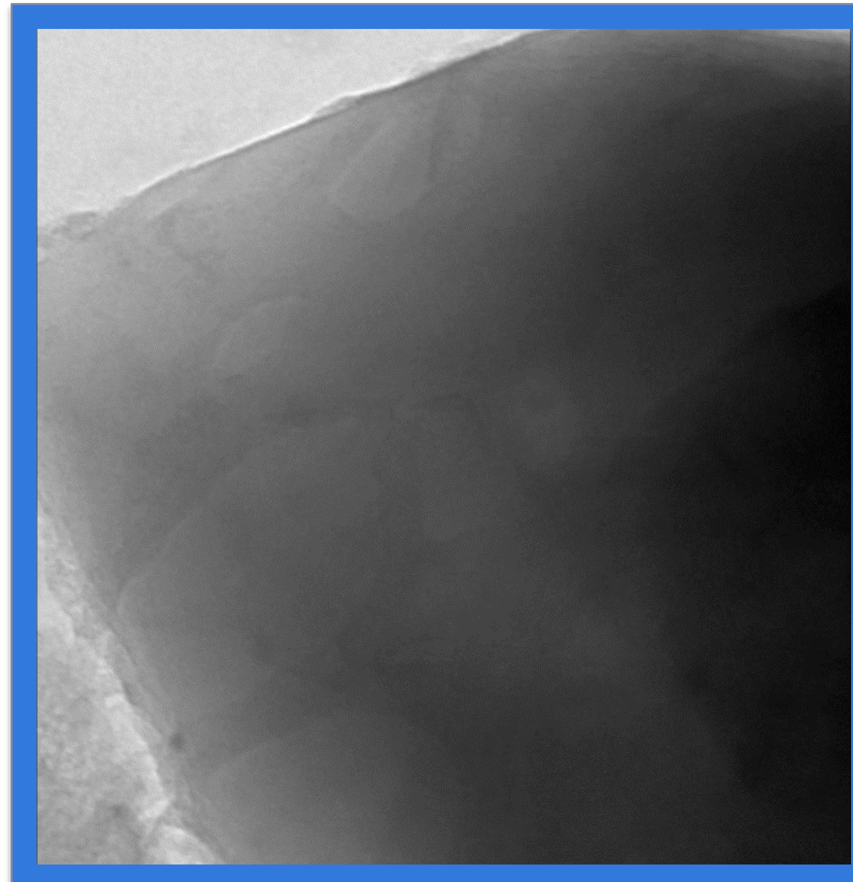
In-situ He + D irradiation @ 310°C

- Each frame = 5 min of irradiation
- All underfocus images
- Electron beam was off except for imaging
- Bubbles formed after ~60 min (1.7×10^{17} He/cm^2, 3.4×10^{17} D/cm^2)

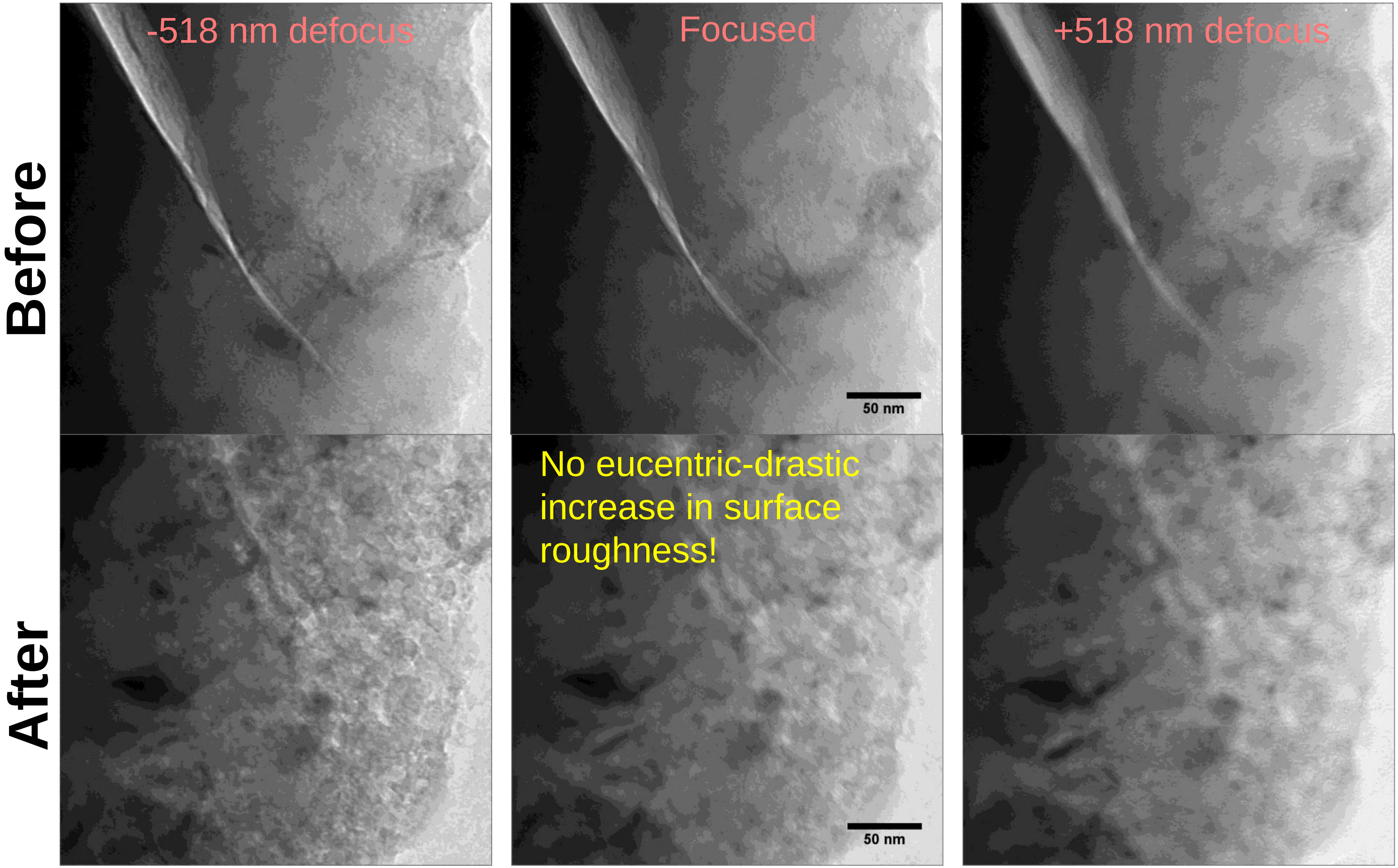
After Irradiation



In-situ Video



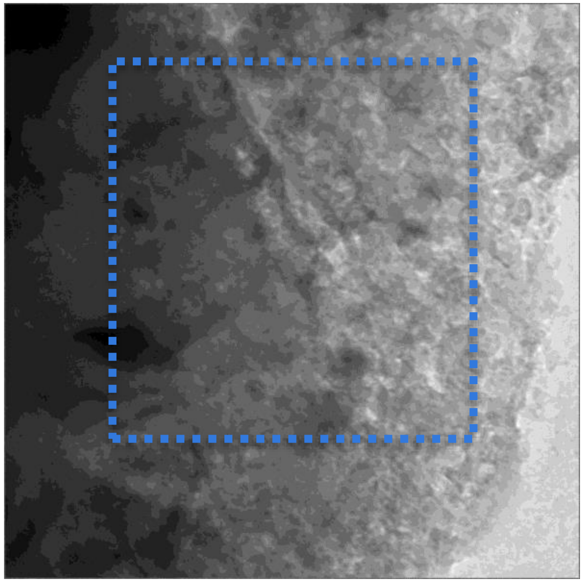
In-situ He + D + Au @ 310°C



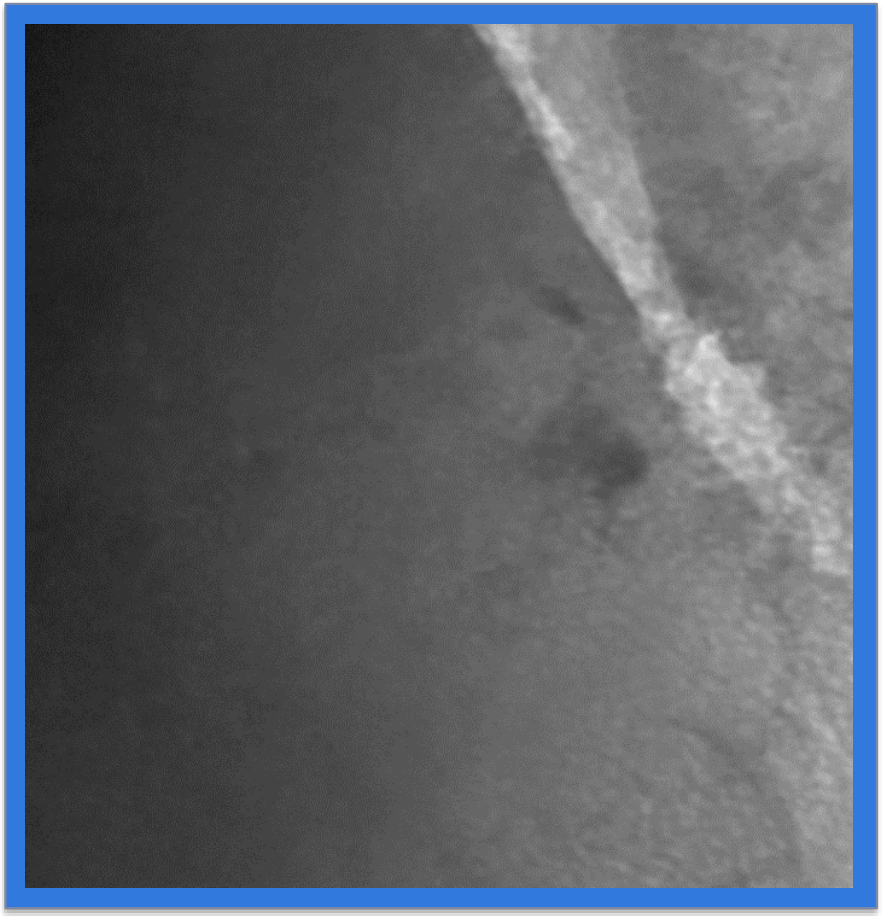
In-situ He + D + Au @ 310°C

- Each frame = 5 min of irradiation
- Pre-existing voids could have an effect on this final microstructure
- Electron beam was on for most of the experiment

After Irradiation



In-situ Video



Summary

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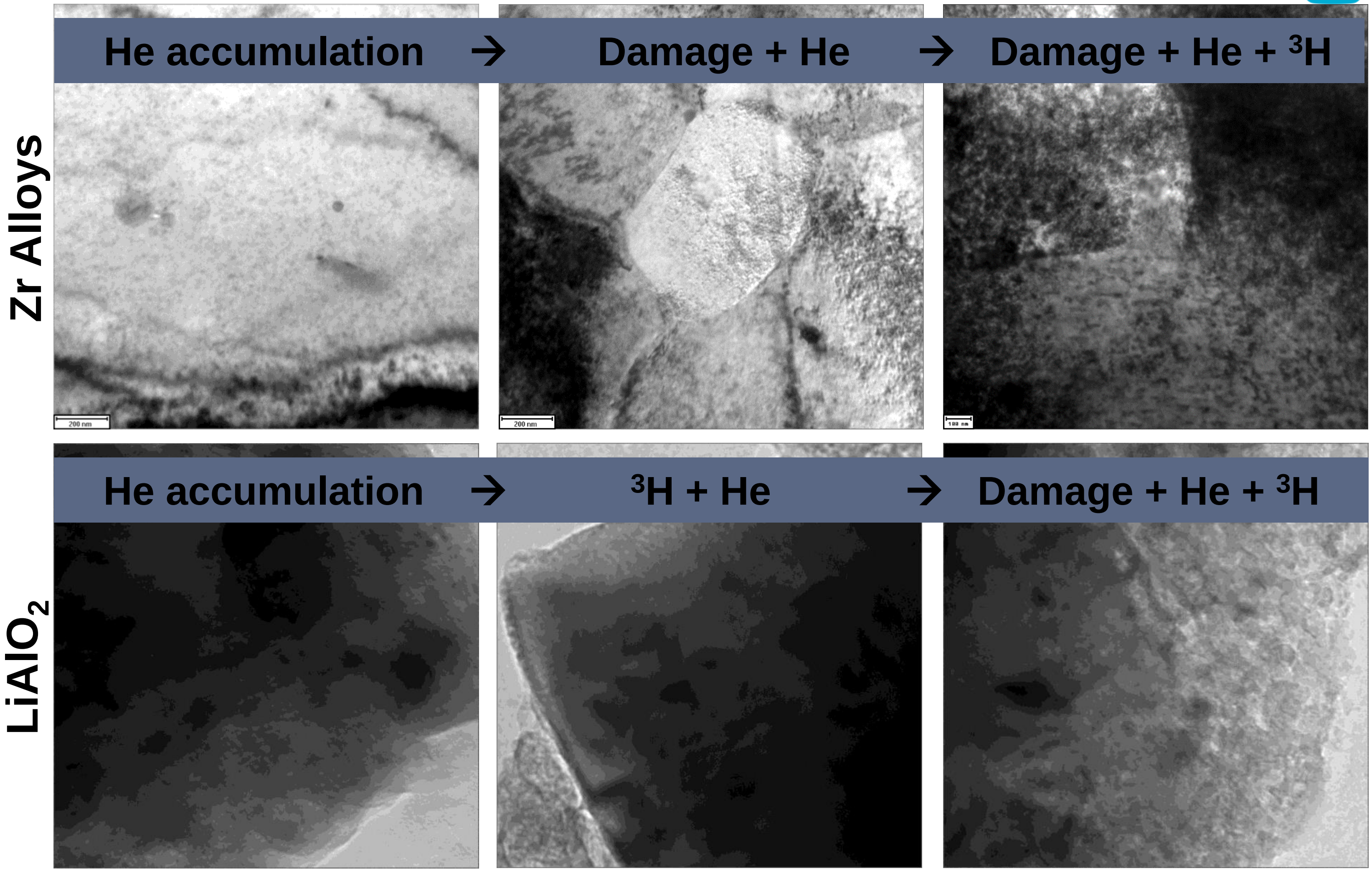

- Synergistic effects between He and ^{3}H accumulation and neutron irradiation induced displacement cascades were simulated in-situ at reactor temperature.
- Microstructure resulting from triple ion irradiation is complex in both materials studied, and will require additional studies to understand defect gas interactions in this complex environment.