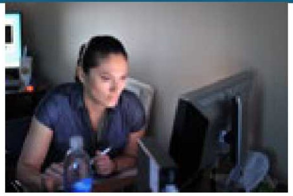
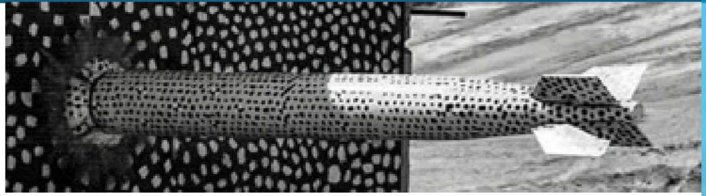
Sandia National Laboratories

SAND2019-10882C

Evolution of He Nanobubbles and Nanocavities in Palladium





PRESENTED BY

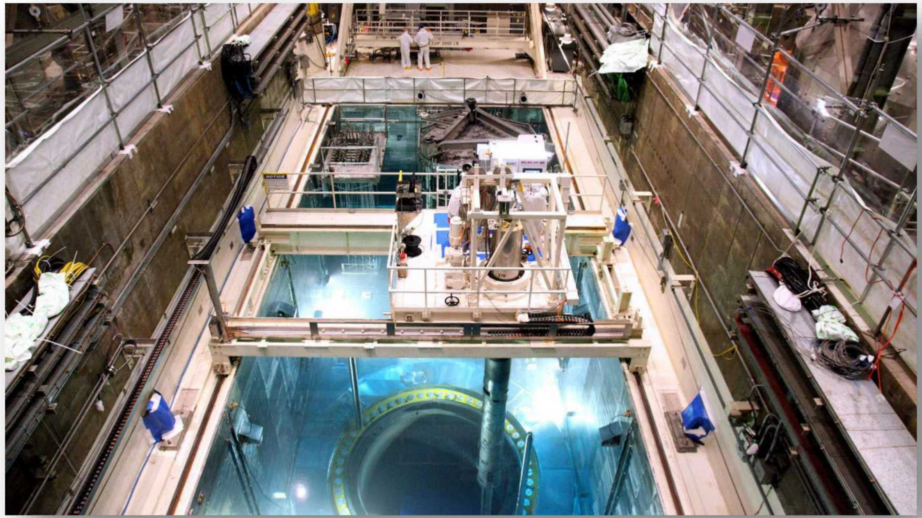
Trevor Clark, Sandia National Laboratories, Albuquerque, NM

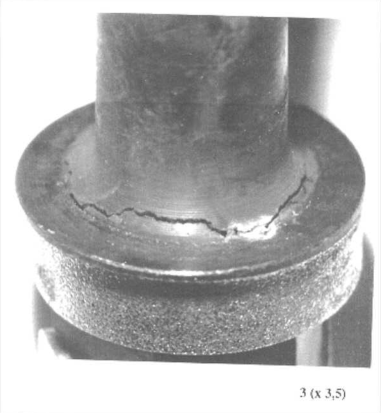
Caitlin Taylor, Khalid Hattar (SNL/NM)
Joshua Sugar, David Robinson (SNL/CA)

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & 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.

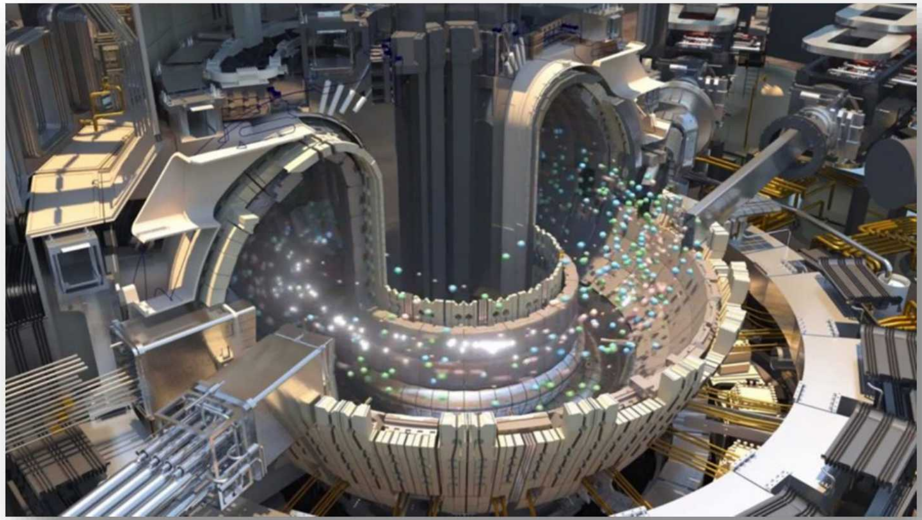
Helium Interactions with Materials

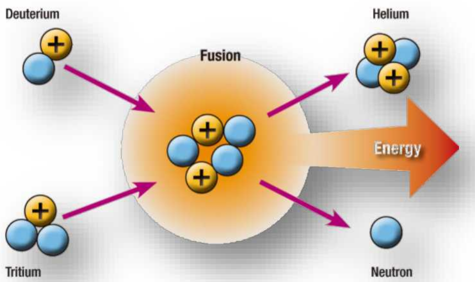
Reactor Materials

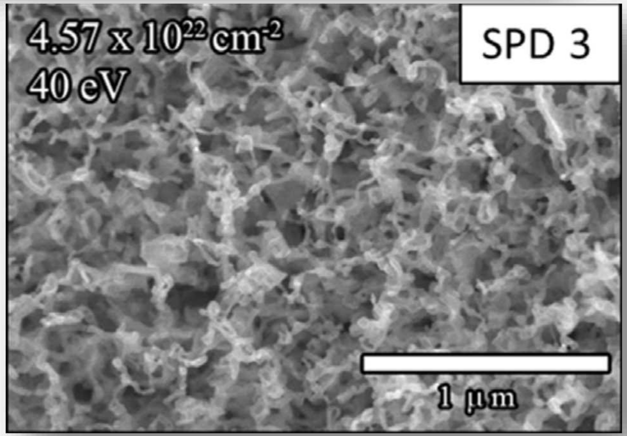




Fusion Energy





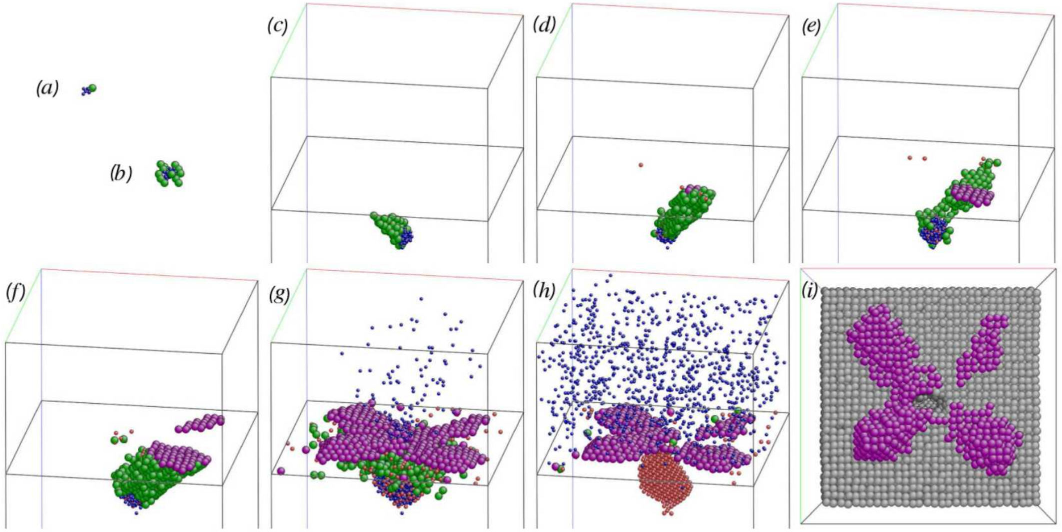


Sources: Iter, AZPM news, J.T. Busby Nanonuclear Workshop (2012), El-Atwani Nuclear Fusion (2014) 54, Fundación General CSIC, Thielmann, AG Materials, physics.Utah.edu.

Bubbles and Cavities: Palladium as a model FCC system

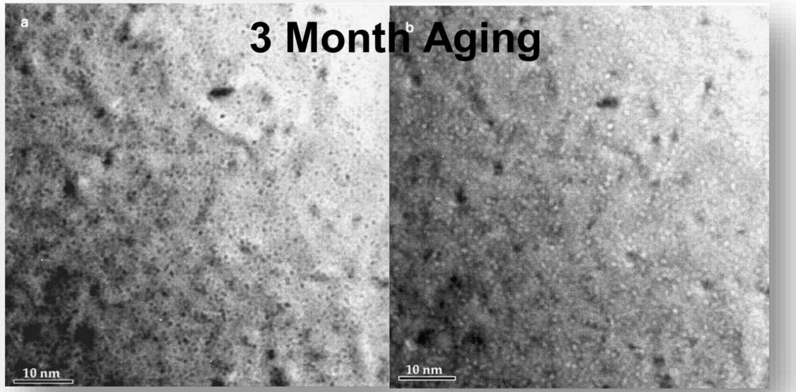
1. Nanobubbles and Nanocavities
   - **β**-decay
   - Implantation

2. Bubble evolution mechanisms
   - Nucleation
   - Bubble growth
   - Migration and coalescence
   - Cavity formation



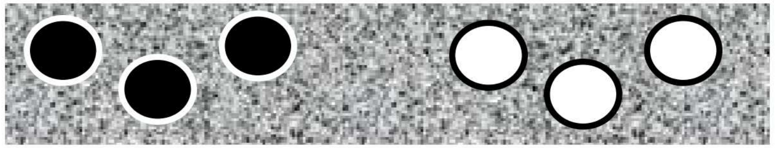
Sefta et al, Nuc. Fus. 53 (2013)

3. Tritium **β**-decay causes He bubbles to form in PdT



Fabre et al, JNM 342 (2005) 101-107

Fresnel contrast in BF TEM



Above eucentric (overfocus)

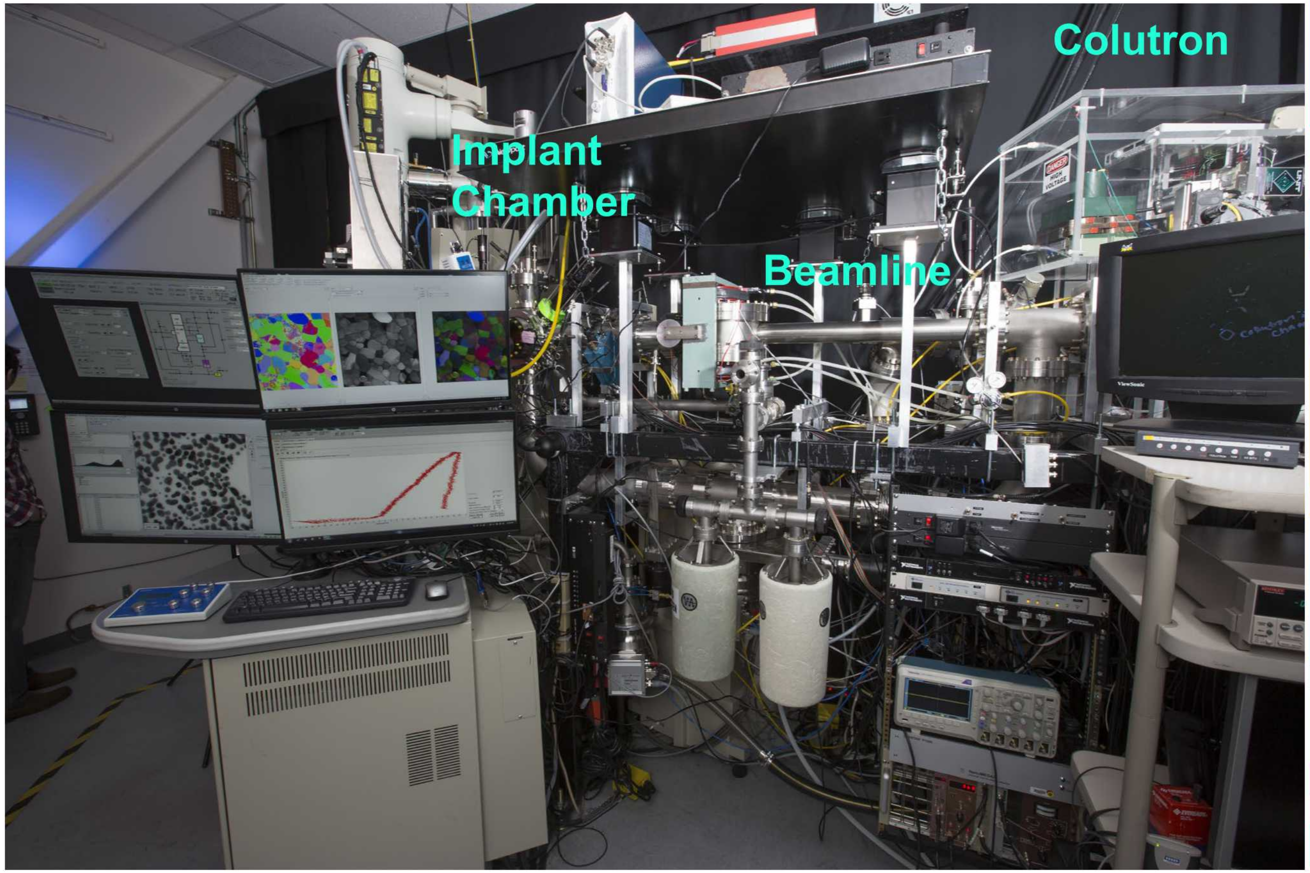
Below eucentric (underfocus)

4. Implantation experiments were done using pure Pd. Minimal oxide concerns.

Multiple Advanced In-situ TEM Techniques were Combined to Increase Our Understanding of He Behavior in Palladium

1. **In-situ Helium Implantation**
   - Accelerated aging: reach high He concentration without making radioactive samples
   - Bubble nucleation kinetics, nucleation sites, and growth
   - How He behavior changes at room, elevated, and cryogenic temperatures

2. **In-situ Annealing in Vacuum**
   - How bubbles evolve under extreme temperatures
   - Provides microstructural information important for interpreting thermal desorption data
   - Accelerated aging method for understanding long-term bubble growth

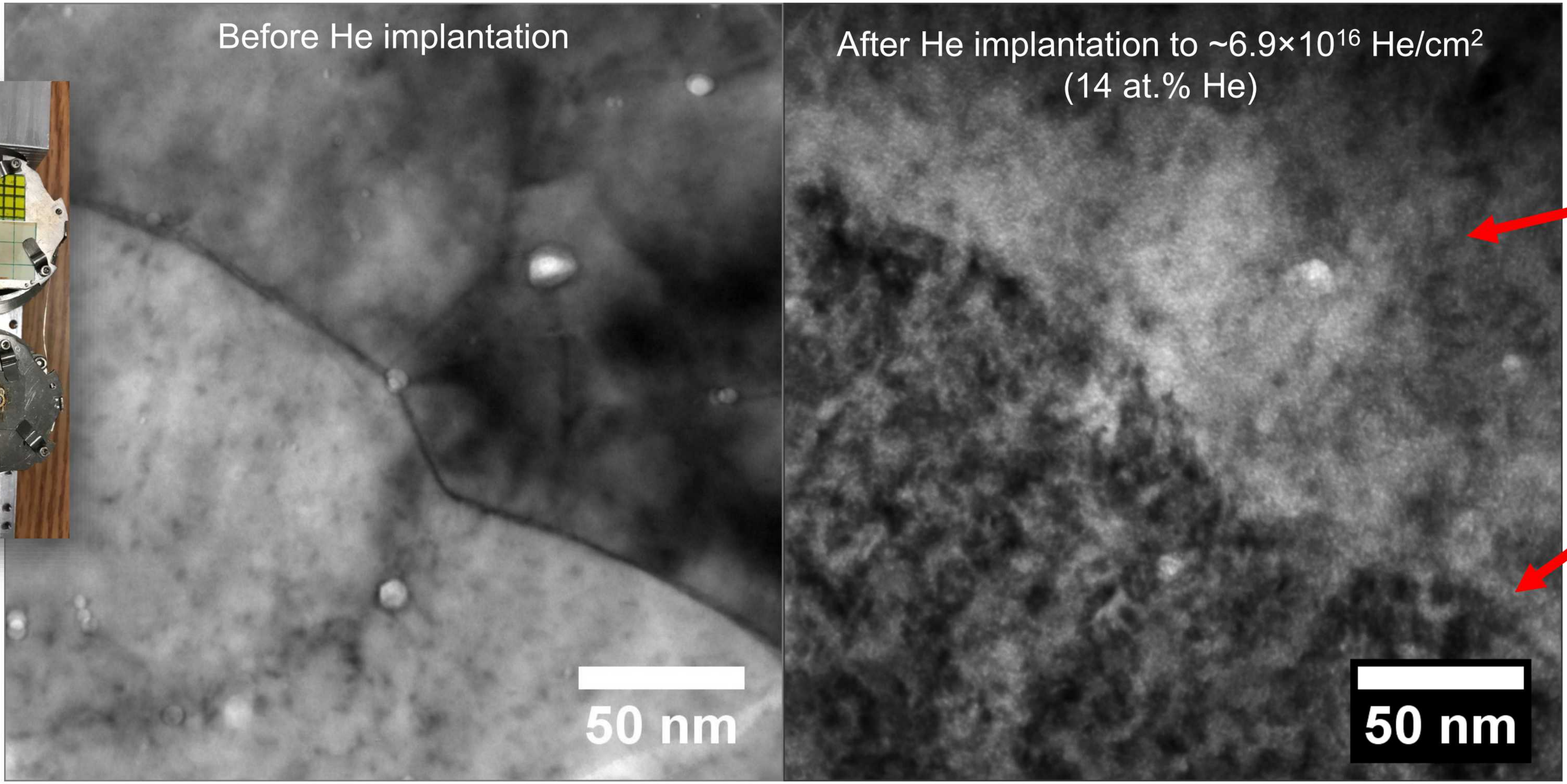


In-situ Helium Implantation

Sandia National Laboratories

In-situ He Implantation: Bubble Nucleation as a Function of Temperature

Pseudo-In-situ Room Temperature Implantation, T/T_M = 0.16

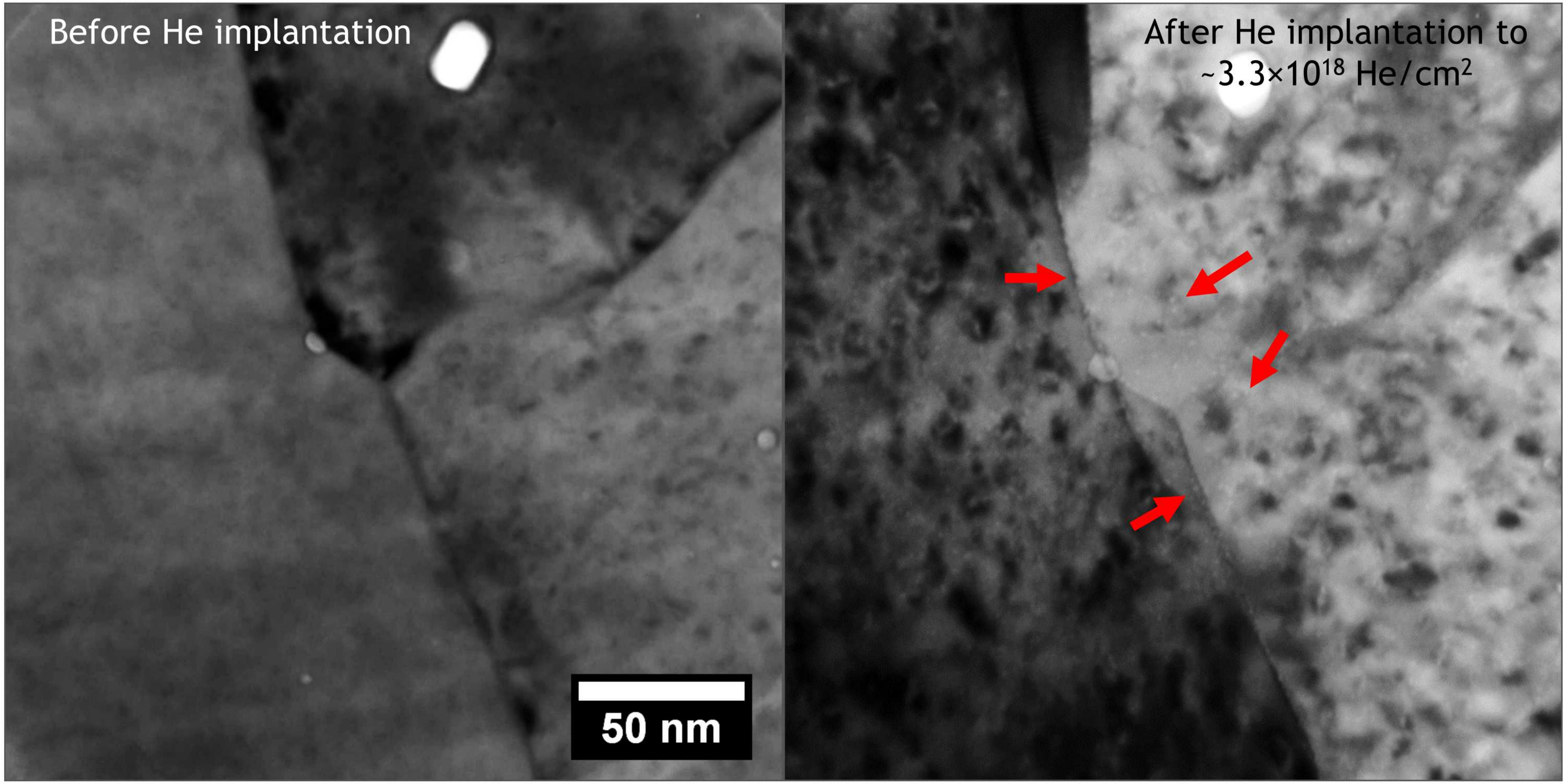


Homogenous distribution of helium bubbles

No preferential nucleation at GBs. Indicates very little diffusion before trapping + nucleation.

In-situ He Implantation: Bubble Nucleation as a Function of Temperature

During in-situ implantation at 250°C (below, T/T_M = 0.29) and 400°C (T/T_M = 0.37), He bubbles nucleated at boundaries and inside the grains



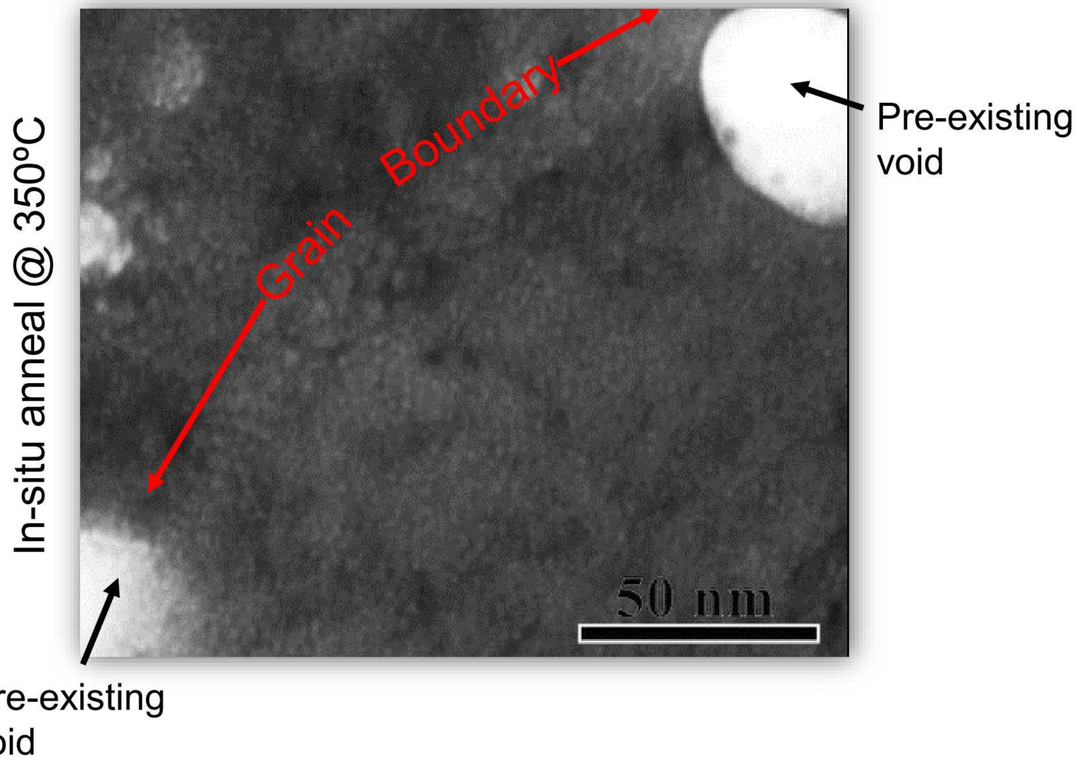
*Note that fluence measurement is an overestimate during in-situ implantation

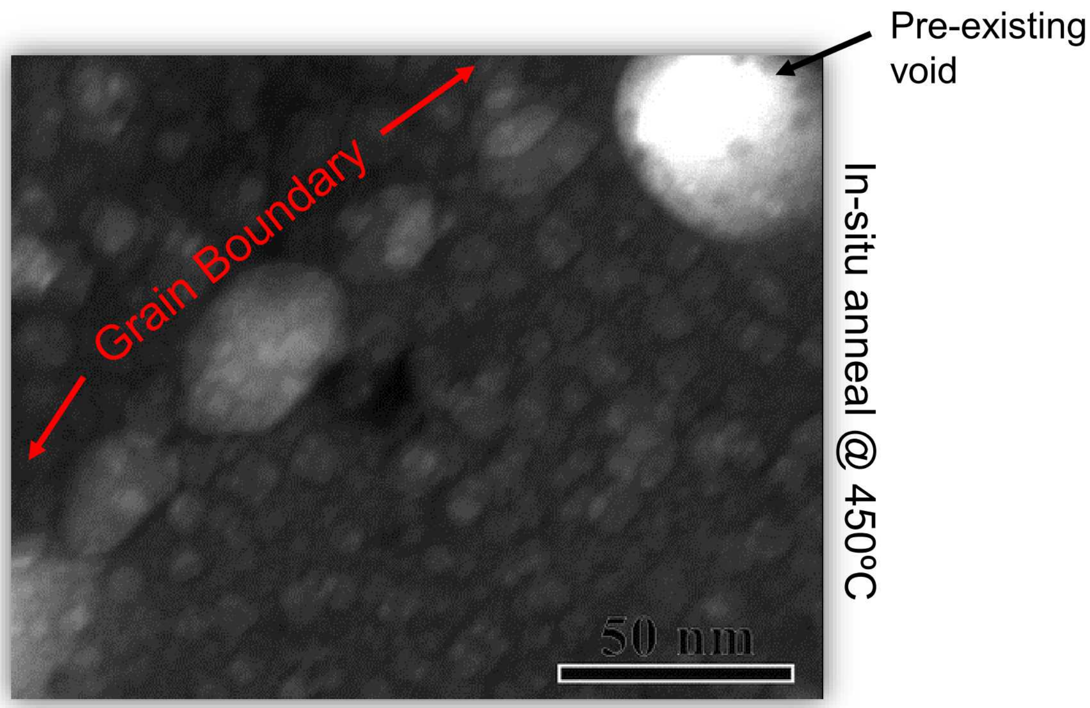
Sandia National Laboratories

In-situ Annealing in Vacuum

In-situ Annealing: Bubble Diffusion and Growth at Elevated Temperatures

Sample implanted with helium at room temperature was annealed in-situ. This sample appeared to have a much higher helium content.



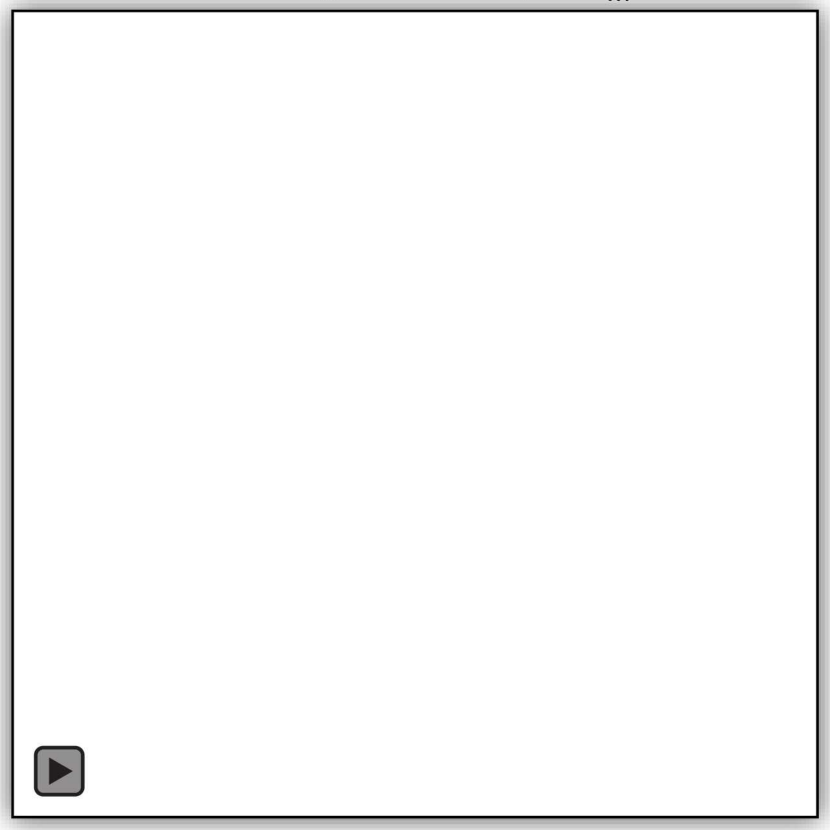


- Blisters form at boundaries by absorbing nearby cavities
- Large faceted cavities form inside the grains by absorbing smaller bubbles and possibly He from the matrix
- Blisters eventually burst, leaving behind a denuded zone at the boundary. Material remains in-tact.

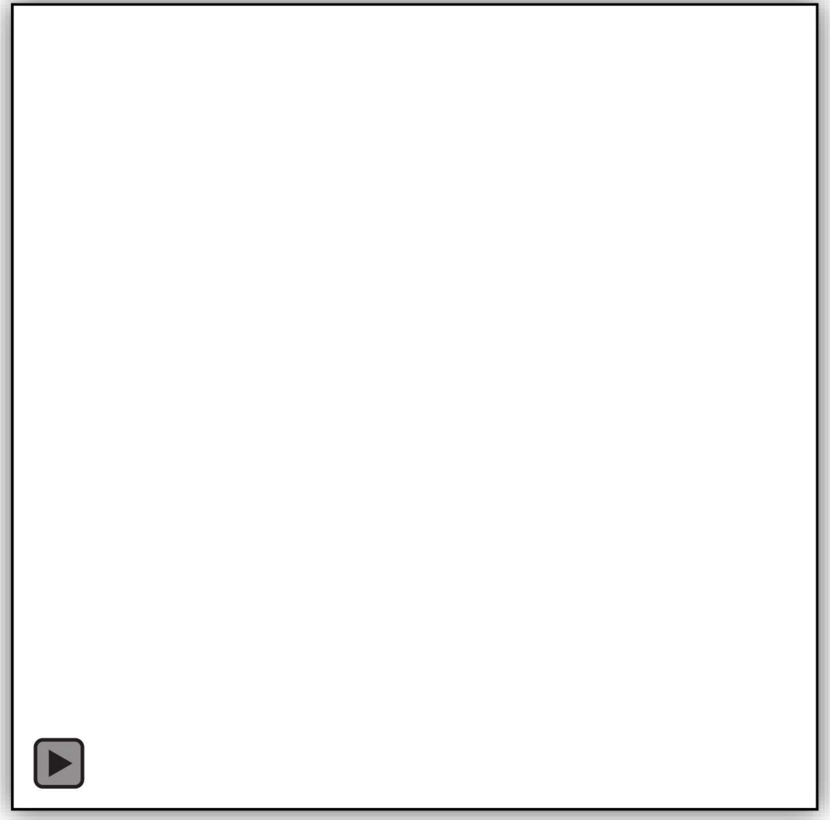
In-situ Annealing: Bubble Diffusion and Growth at Elevated Temperatures

Samples implanted with helium at elevated temperatures were annealed in-situ. These samples appeared to have a much lower helium content. Same behavior was observed in both samples, sample implanted at 400ºC shown below.

In-situ annealing at 550ºC, $T/T_M = 0.45$



In-situ annealing at 700ºC, $T/T_M = 0.53$

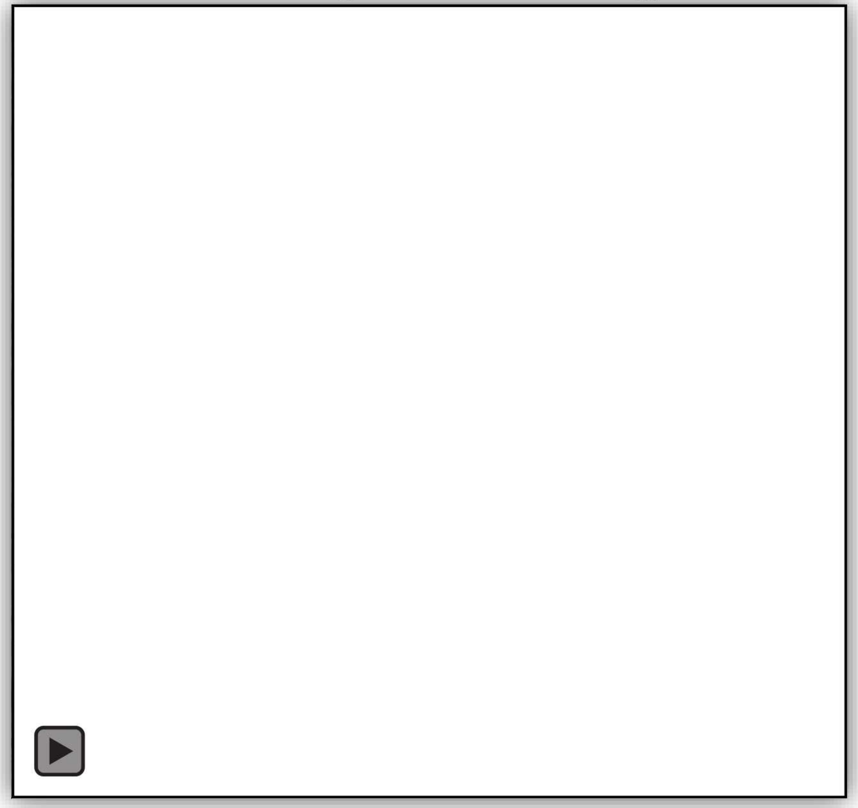


- Bubble growth by bubble migration and coalescence
- Bubbles remain tied to boundaries during grain growth and appear to pin boundaries in some cases
- Some bubbles appear to be strongly trapped inside grains (e.g. at defects)

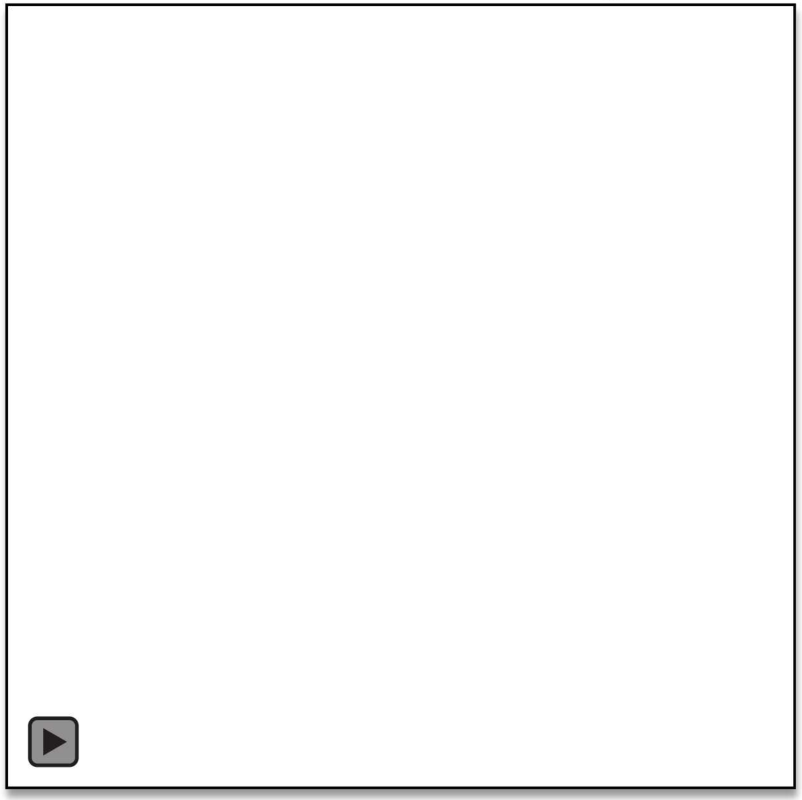
In-situ Annealing: Bubble Diffusion and Growth at Elevated Temperatures

In-situ anneal was also performed on tritium aged Pd- 5% Ni alloy containing ~12 at.% He (similar to the sample implanted with 14 at.% He at room temperature and annealed).

In-situ annealing at 400ºC , $T/T_M = 0.37$



In-situ annealing at 900ºC , $T/T_M = 0.64$



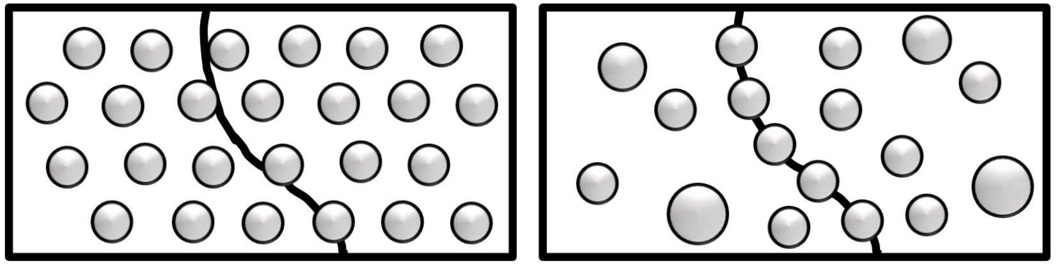
- Facets indicated that cavities are near equilibrium by $T/T_M = 0.37$
- At high temperature, cavities coalesced or reached the surface, leaving behind denuded zones

Sandia National Laboratories

Summary

Helium Implantation

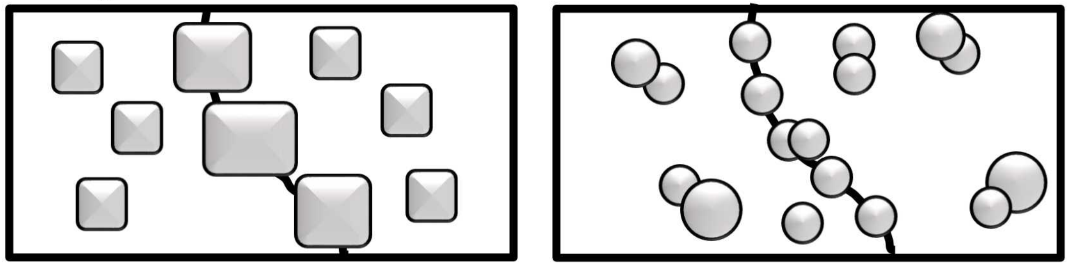
Room Temperature | High Temperature



He becomes trapped immediately at room temperature, not very mobile. Diffuses before trapping at high temperature.

Annealing in Vacuum

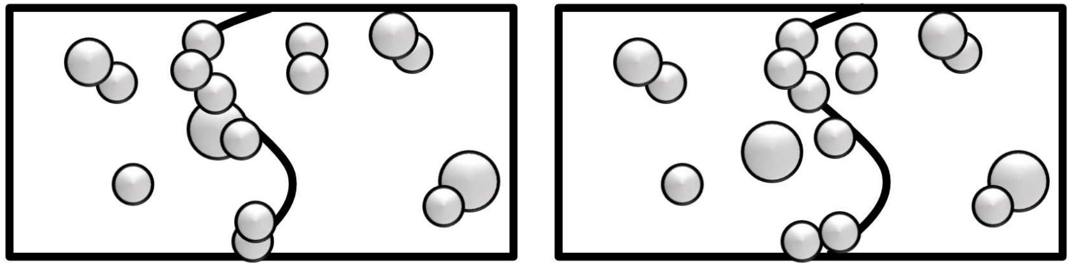
High He Concentration | Low He Concentration



Growth through absorption of nearby cavities with high He concentration, facet formation at $\sim T/T_M = 0.4$ as thermal vacancy concentration increases. Growth through bubble diffusion and coalescence with low concentration.

Grain Boundary Interaction

Bubbles Pin Boundary | Release from Boundary



Growth through absorption of nearby cavities with high He concentration, facet formation at $\sim T/T_M = 0.4$ as thermal vacancy concentration increases. Growth through bubble diffusion and coalescence with low concentration.

Sandia National Laboratories

Questions

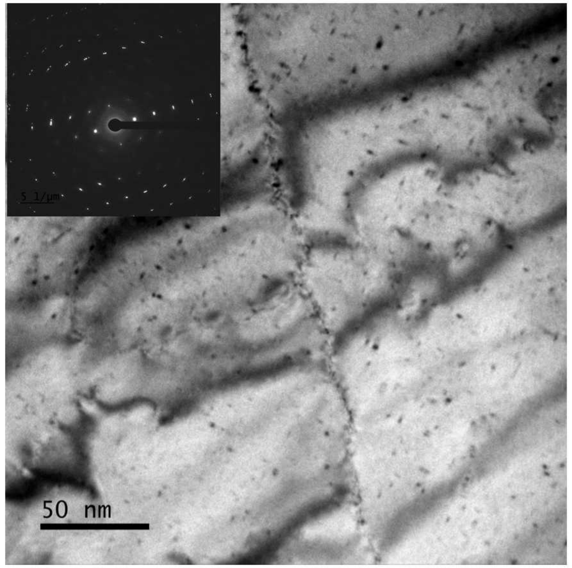
Thank you

Extra slides

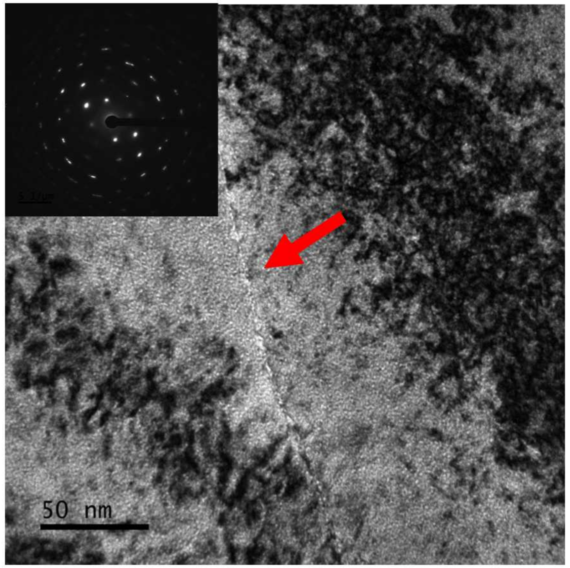
In-situ He Implantation: Bubble Nucleation as a Function of Temperature

Collaborators: Jonathan Hinks, Steve Donnelly, Robin Grimes, Emily Aradi

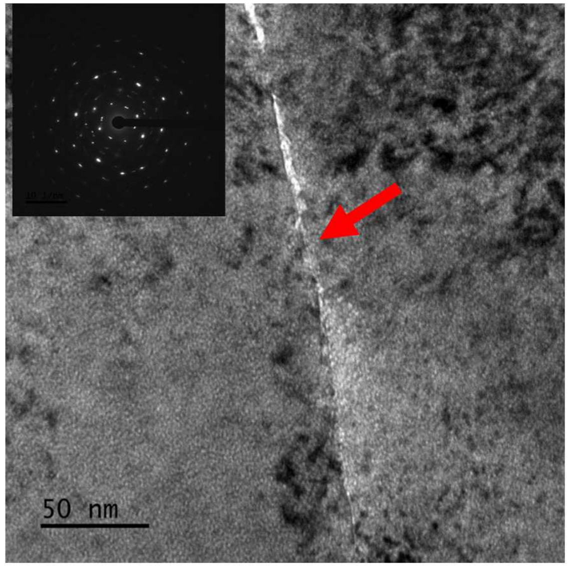
In-situ cryogenic helium implantation in a H_2 gas environment was performed on one sample using the in-situ helium implantation environmental TEM at Huddersfield in the UK.



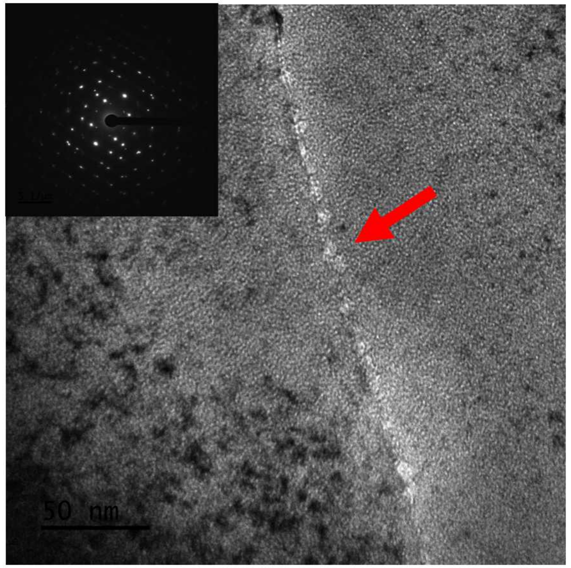
H_2 gas flow at -100°C



H_2 gas flow at -100°C, after He implantation to ~ 1.0 x 10^{17} ions/cm^2



H_2 gas flow at -60°C, after He implantation



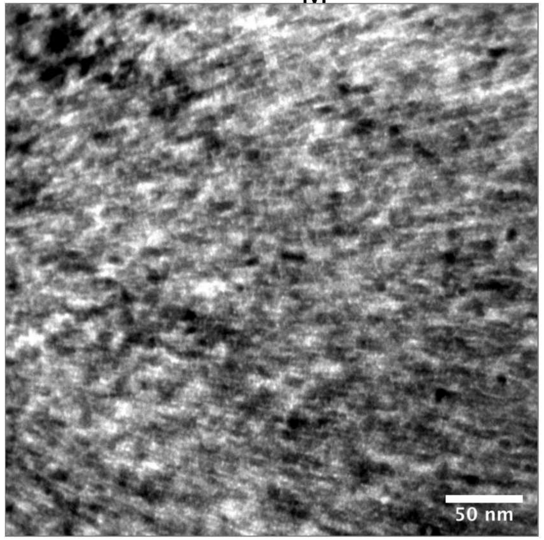
H_2 gas flow at 0°C, after He implantation

- E-TEM gas pressure was in the mTorr range, so cryogenic temperatures were required for hydriding. At 1 mTorr, the concentrated hydride phase should eventually form at temperatures <-95°C, however, hydrogen diffusion and hydride reaction kinetics are unknown at cryogenic temperatures.

In-situ Annealing of Tritium Aged Pd-5% Ni in **Vacuum Protochips**

All Images at 100 KX Magnification

25ºC, T/T_M=0.16



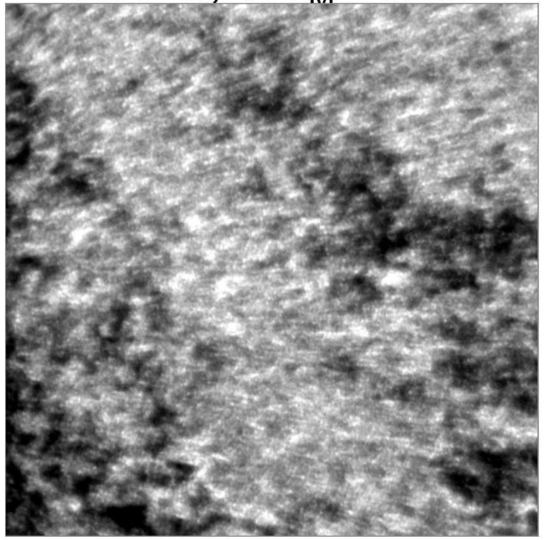
100ºC, T/T_M= 0.20

No data

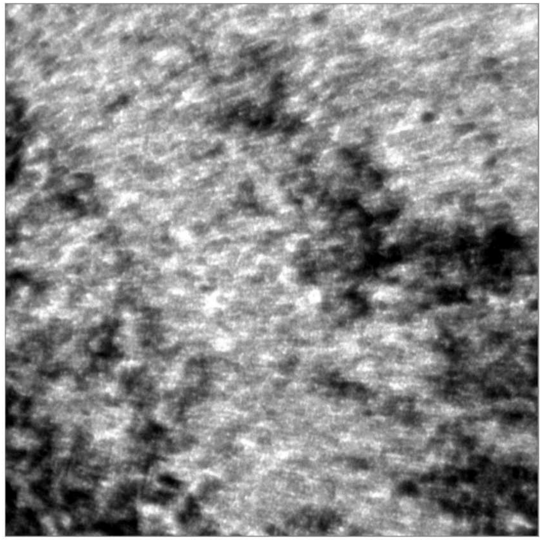
150ºC, T/T_M= 0.23

No data

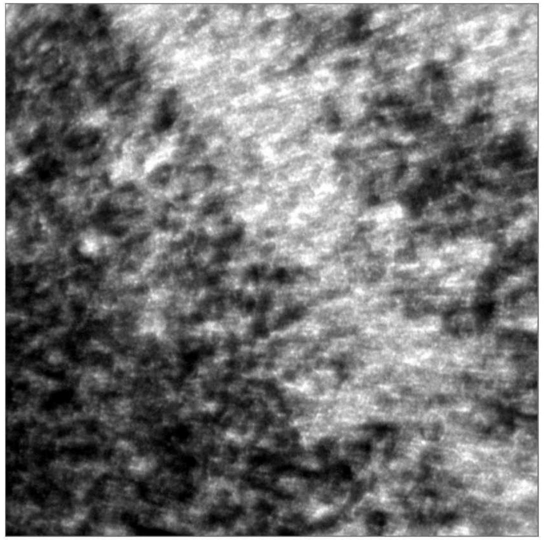
200ºC, T/T_M= 0.26



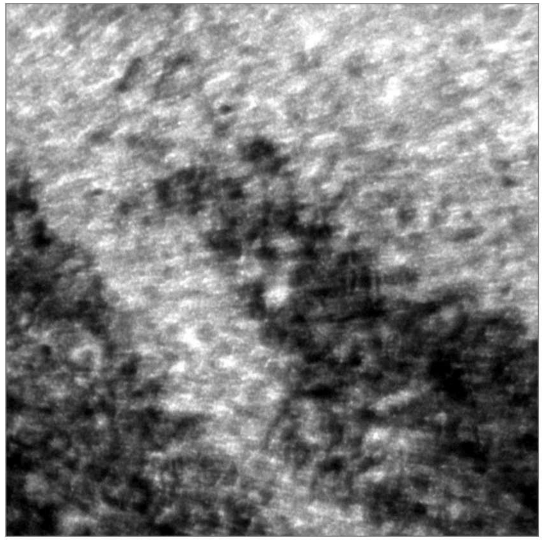
250ºC, T/T_M= 0.29



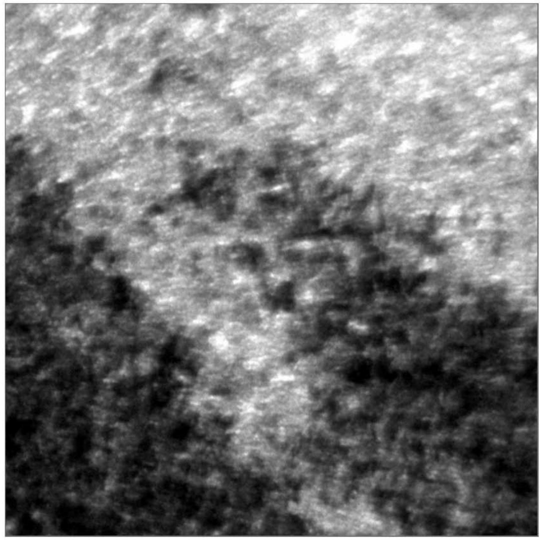
300ºC, T/T_M= 0.31



350ºC, 0.34



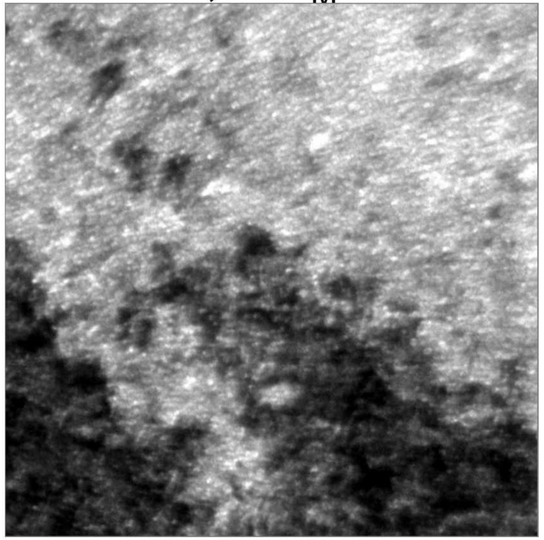
400ºC, T/T_M= 0.37



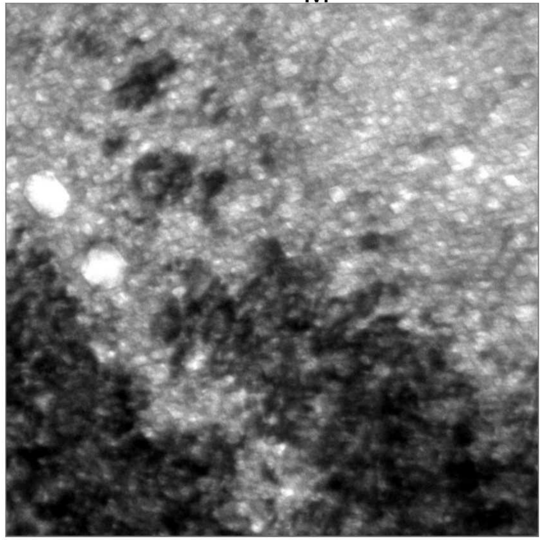
In-situ Annealing of Tritium Aged Pd-5% Ni in **Vacuum Protochips**

All Images at 100 KX Magnification

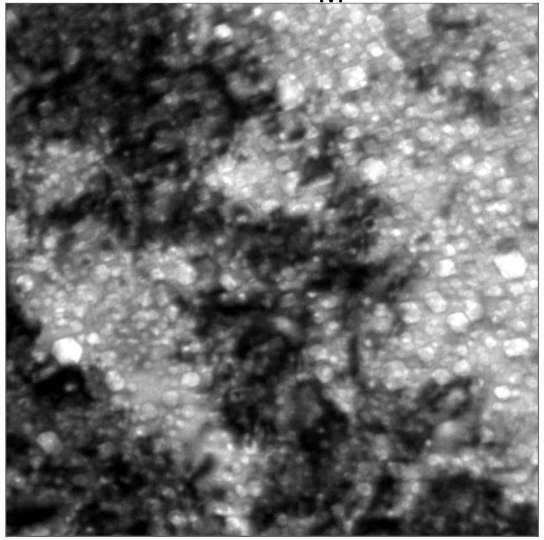
450ºC, T/T_M= 0.40



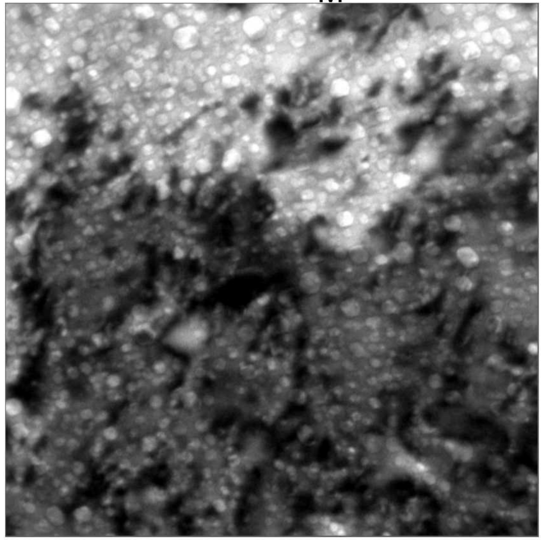
500ºC, T/T_M= 0.42



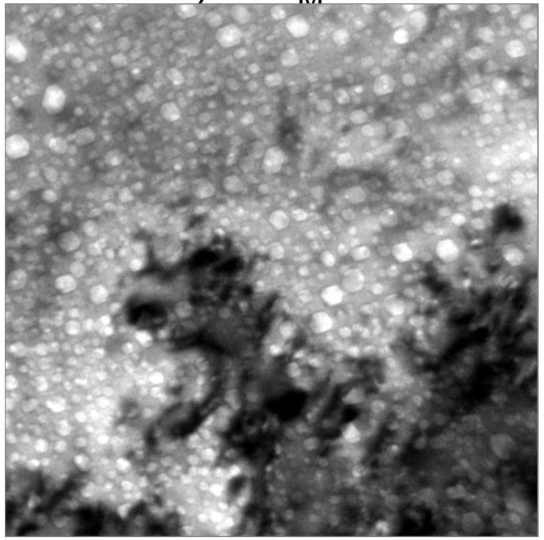
550ºC, T/T_M= 0.45



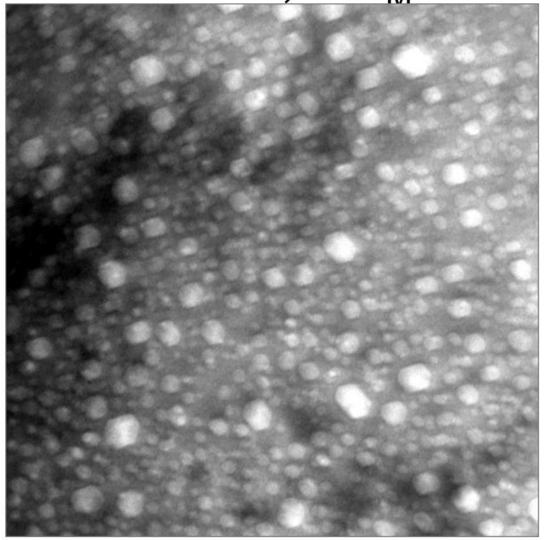
600ºC, T/T_M= 0.48



650ºC, T/T_M= 0.50



700ºC – 6 min, T/T_M= 0.53

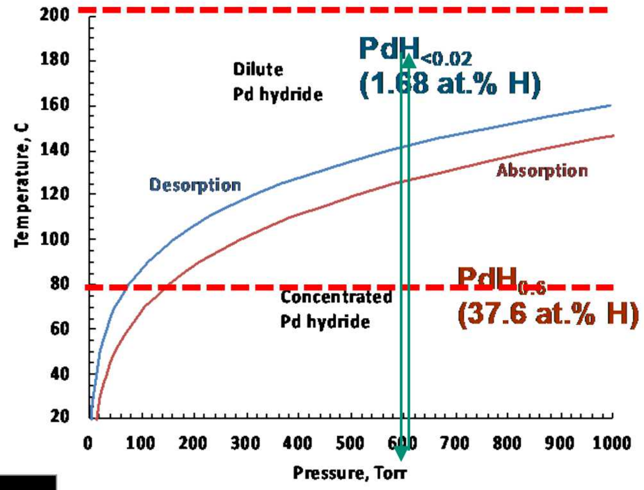


No data

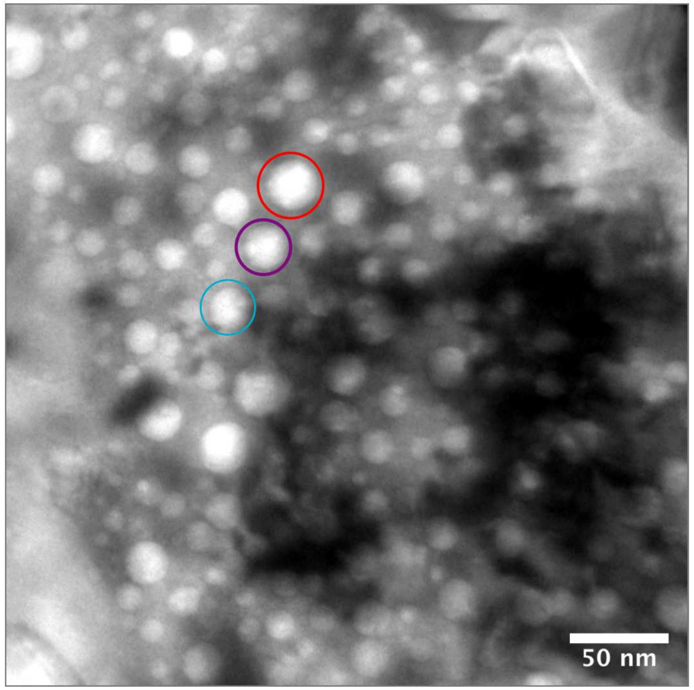
No data

In-situ Hydriding Post-Annealing

- Cooled the annealed sample to 200ºC, filled the stage with 1 atm H_2 gas, and performed 5 hydride cycles between the dilute and concentrated phases by switching between 200ºC and 80ºC.
- Could clearly observe movement into the two phase region and then into the concentrated region through movement of bend contours, as well as sample drift and z-height changes.
- No obvious changes in bubble size.



5th hydride cycle, 200ºC



5th hydride cycle, 80ºC