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Three-Dimensional Tomography of Helium Bubbles in a Palladium Alloy Tritide

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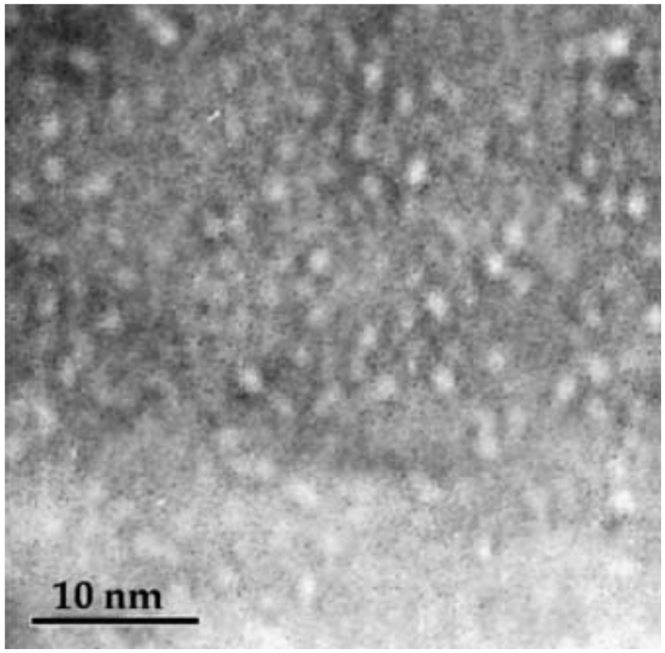
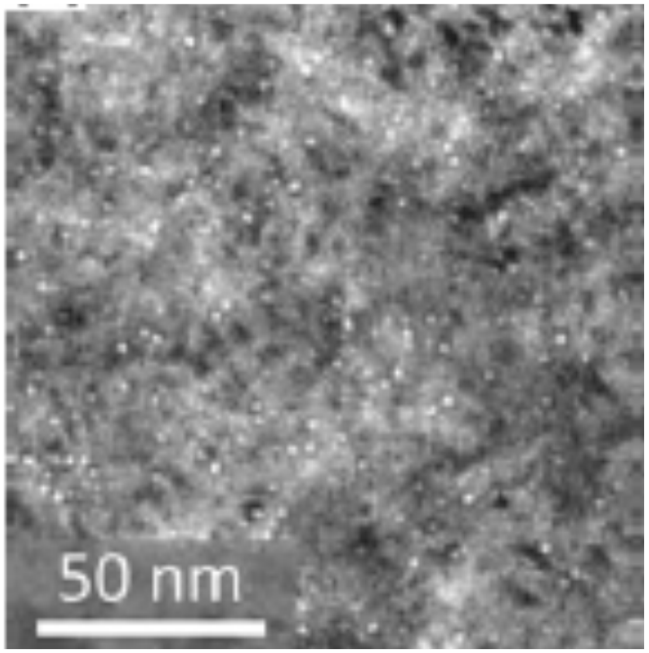
Helium Bubbles in Metals



A. Fabre et al., 8-month Pd tritide
J. Nuc. Mater. 342 101 (2005)



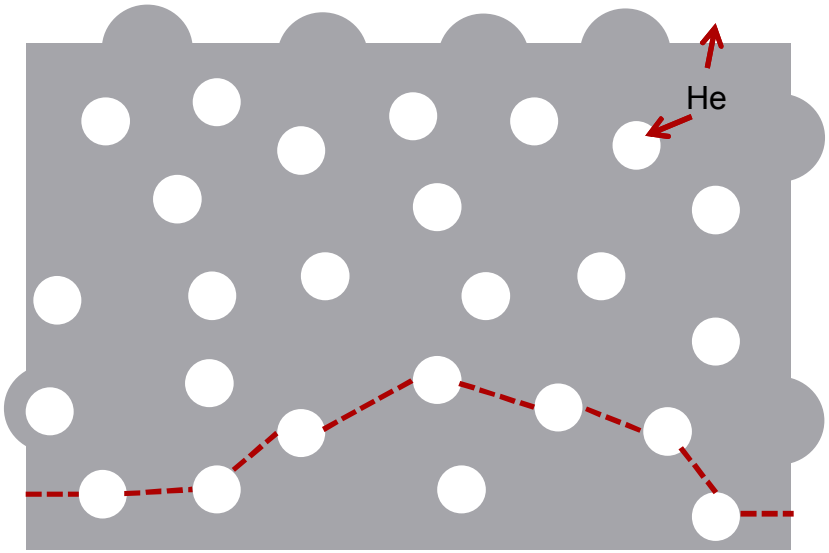
J. R. Jeffries et al., Pu-Ga alloy
J. Nuc. Mater. 410 84 (2011)

- Caused by ^{3}H β decay, α decay, neutron (n, α) reactions
- He and/or radiation-induced vacancies form bubbles
- Bubbles are a few nm diameter, a few nm apart

2D TEM Images

- No information on bubble spacing in z direction
- Large bubbles may obscure small bubbles, skewing distribution
- Smallest bubbles may be difficult to observe

^{3}He causes bubbles in metal tritides



- ^{3}H decays to insoluble ^{3}He
- ^{3}He clusters push metal atoms, forming bubbles
- Bubbles create fracture paths, swelling/deformation
- ^{3}He may escape at surfaces (and fracture surfaces), through grain boundary paths
- Properties are believed to depend on size and spacing distribution of bubbles

Early nucleation process is conventionally thought to define distributions, property evolution

D.F. Cowgill, Fusion Sci. Tech. 28 539 (2005)
J.H. Evans, J. Nuc. Mater. 68 129 (1977)
F. Montheillet et al., Mat. Sci. Eng. A 494 407 (2008)

Questions and Solutions

- How do helium bubbles nucleate (early nucleation vs. continuous nucleation) and grow in Pd?
- What determines bubble density?

Without direct 3D information, 2D TEM images of bubbles cannot answer these questions

We aim to generate three dimensional images of bubble configurations and use them to deduce the history of their evolution

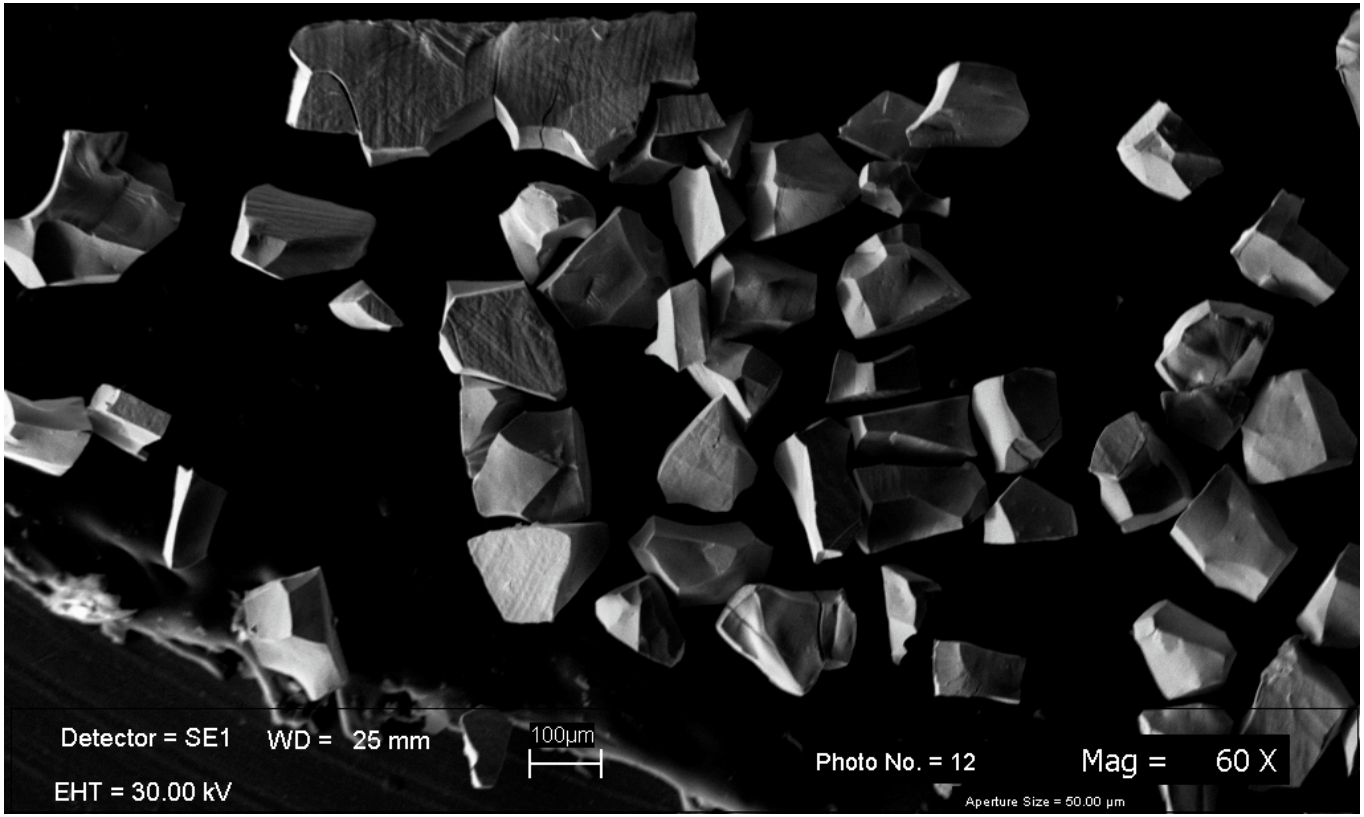
Significance of Knowledge of 3D Distribution of ^{3}He Bubble size and Spacing

- Validate models of early nucleation process as balance between ^{3}He self trapping, ^{3}He transport, and lattice defect interactions
- Assess extent of late nucleation, interbubble interactions during growth phase
- Allow models to better predict macroscopic effects: swelling, fracture, ^{3}He release

Experimental Challenges … need to obtain aged samples!

SRNL Pd-5 at.% Ni ribbon

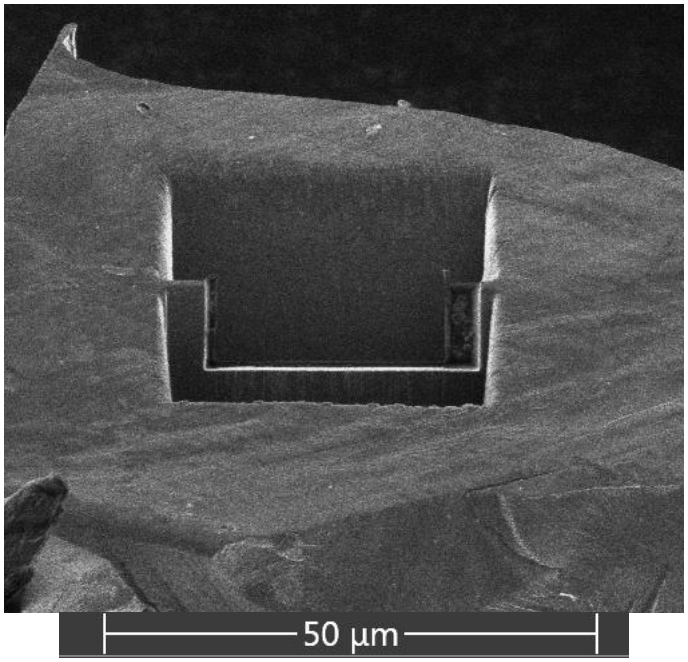
Sample crumbled during its lifecycle

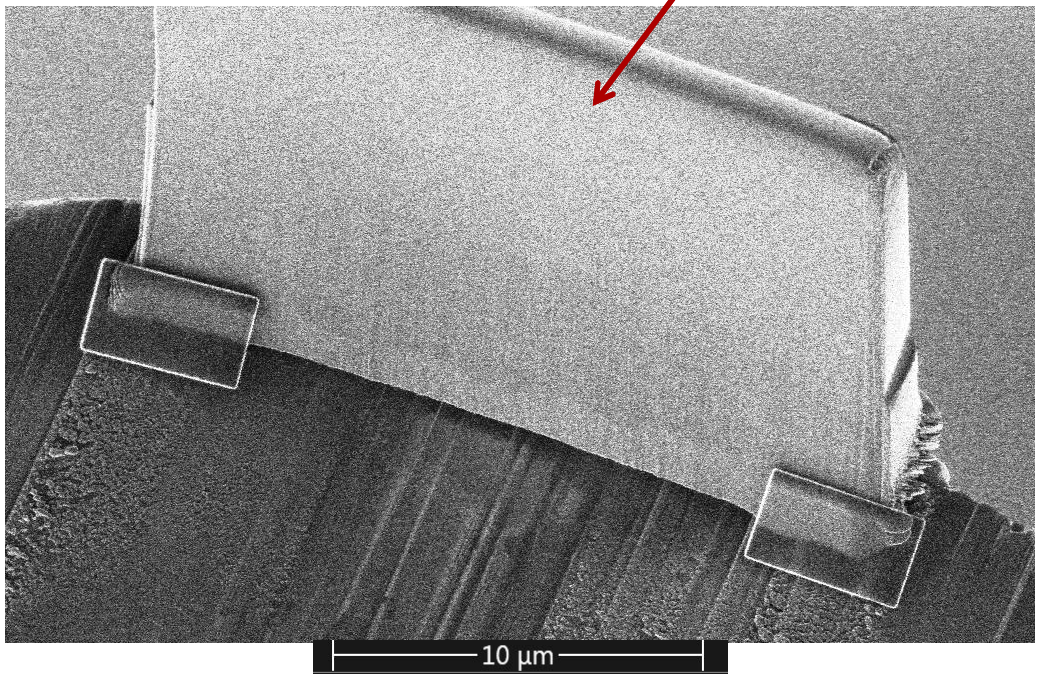


- Tritided for 3.8 years
- Estimated He/Pd = 0.12
- De-tritided by D_2-vacuum cycles

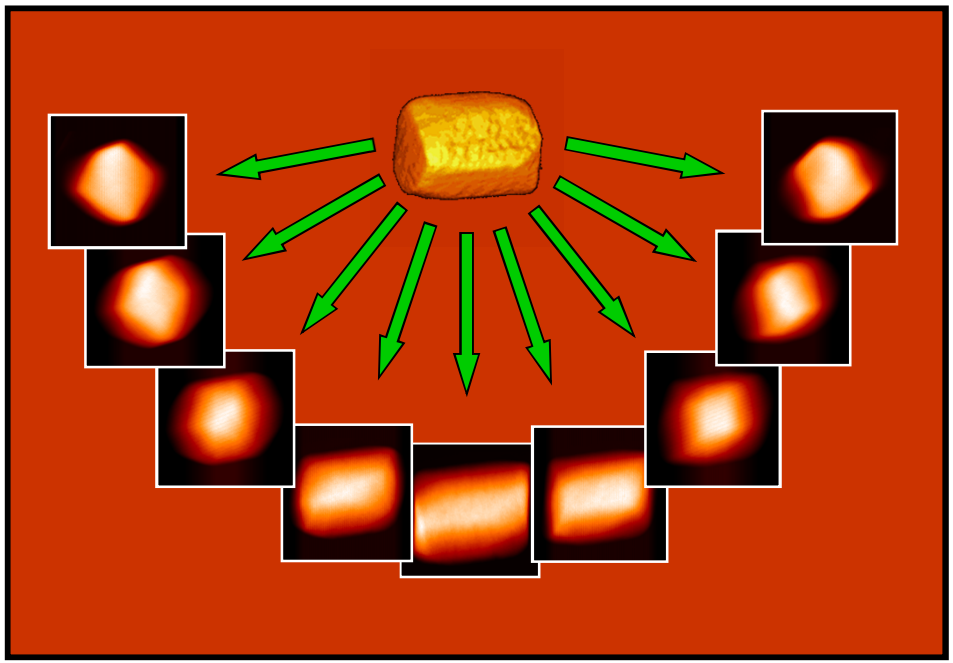
FIB-SEM Sectioning

- Avoids epoxy binder, excess cuttings of microtome
- ~ 10 picoliters vaporized into FIB-SEM, 0.4 pL goes into TEM
- Some Ga ion implantation is expected





Electron Tomography



Sample images are recorded in a transmission electron microscope at many angles.

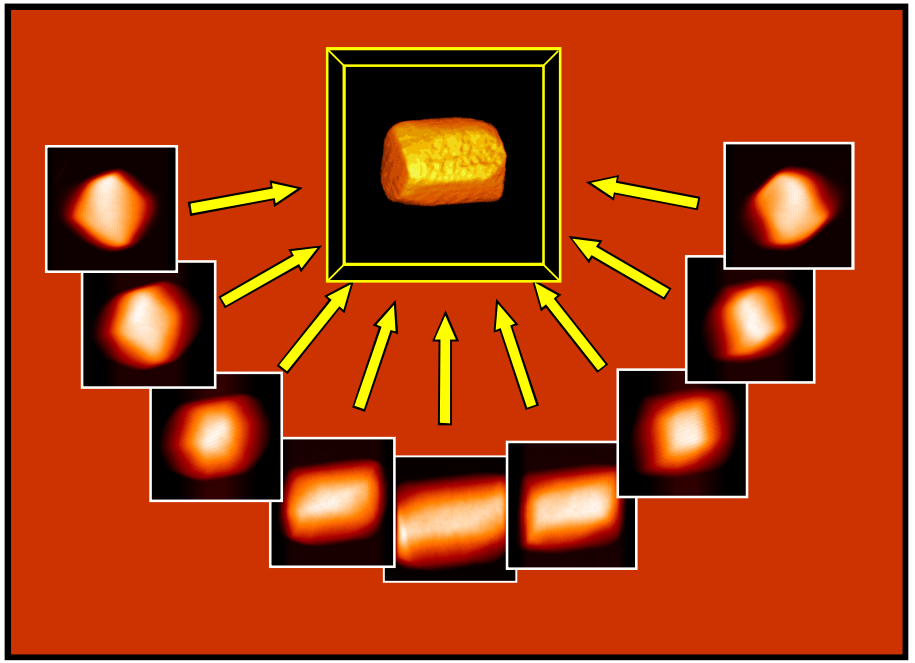


Image data are used to reconstruct a 3D representation of the sample.

Figure prepared by Ilke Arslan

FIB-SEM sample for tomography

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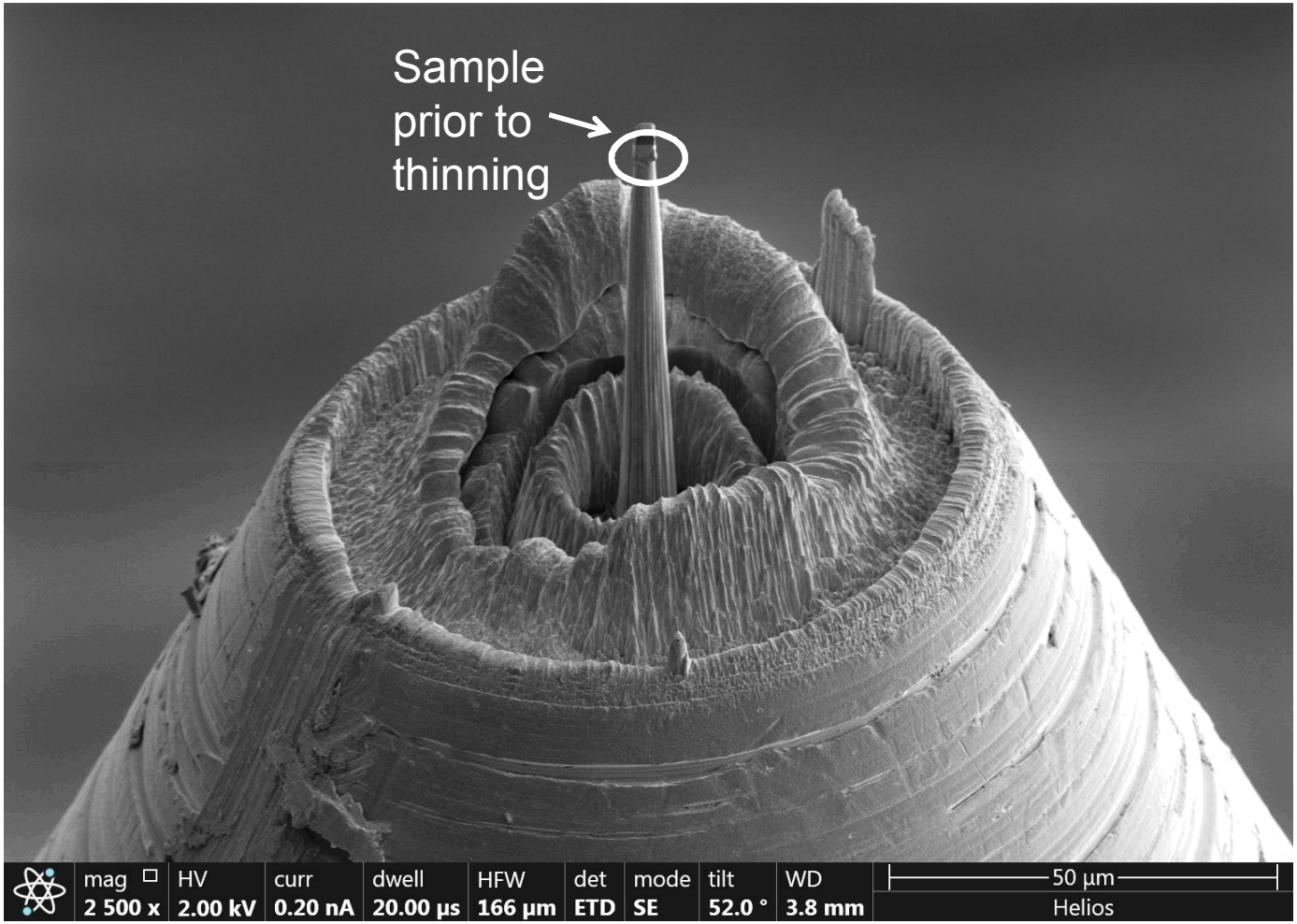
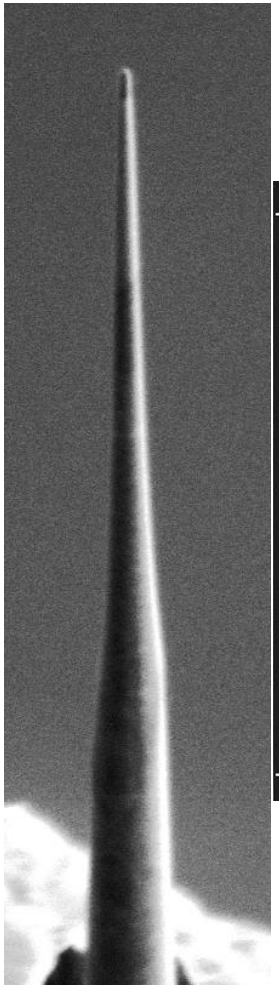
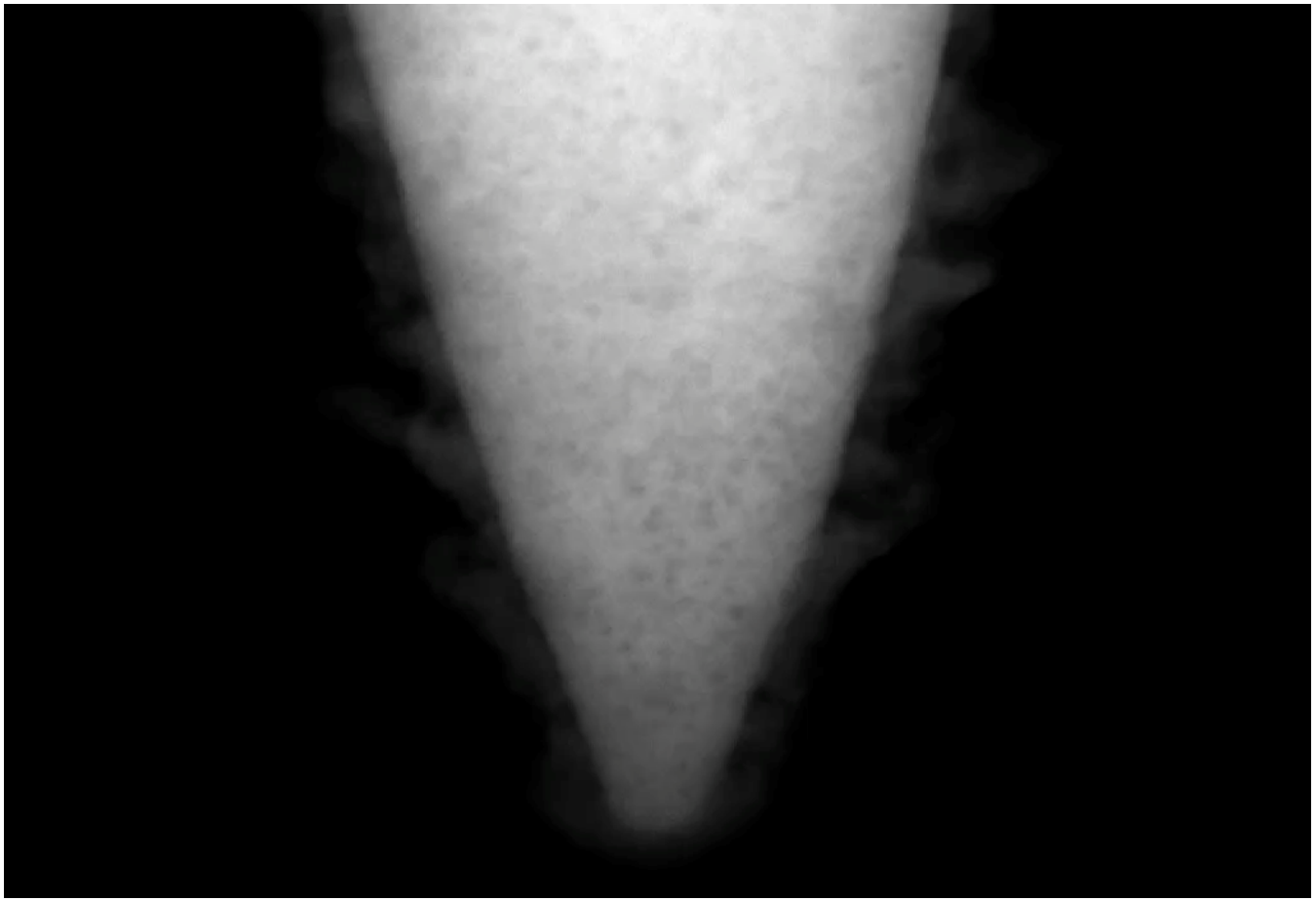
Tip geometry allows imaging at many angles





Nearly finished sample

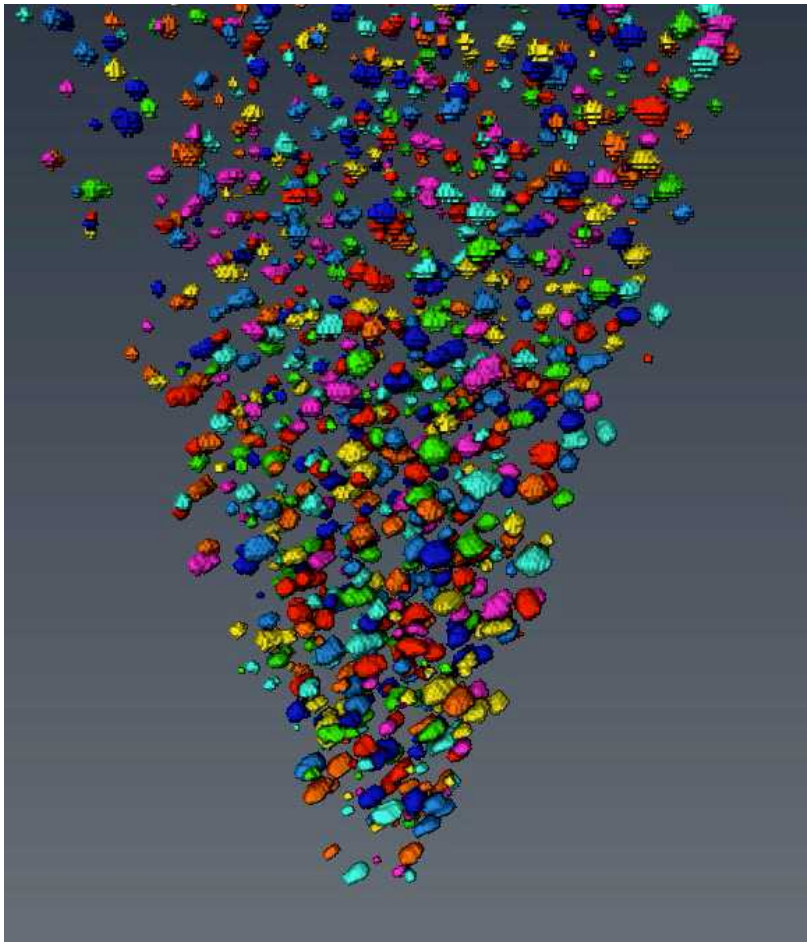
Dark field (HAADF) TEM Images



180 nm

- Tritided for 3.8 years (from a SRNL Pd-5 at.% Ni ribbon)
- Single crystal
- Bubbles are dark, ~2 nm diameters
- Images taken from -70^{o} to 70^{o} in 1^{o} increments

Reconstruction of the 3D Bubbles

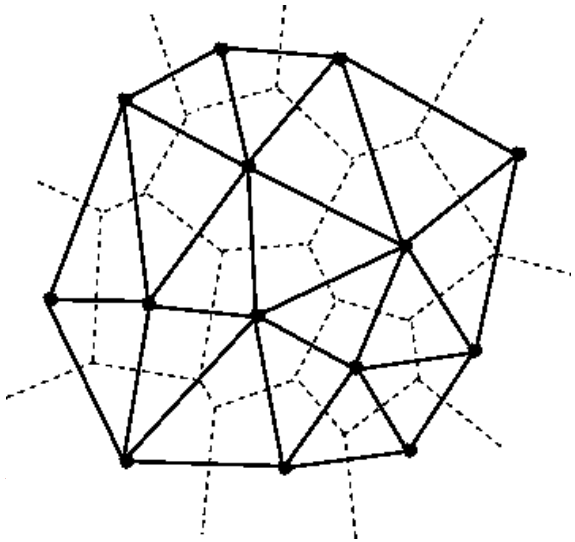


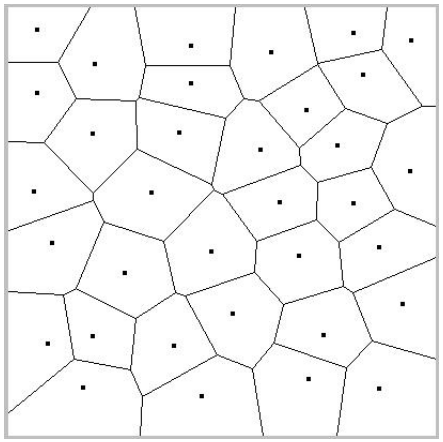
- Each bubble has own color
- The 3D bubbles are iteratively matched to 2D experimental images using the "*Simultaneous Iterative Reconstruction Technique.*"
- ~1000 bubbles with average diameter 2 nm
- Bubbles can be elongated due to reconstruction artifacts

Capture Volume Analysis

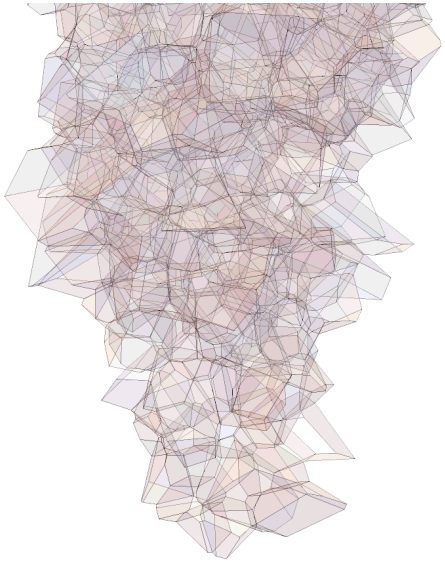
- If all bubbles nucleate at a same early time and their growth is diffusion limited, then they should contain the He generated in a capture region geometrically nearest each bubble
- Capture volume is represented by Voronoi tessellation

2D voronoi tessellation





3D voronoi polyhedra of a sample



small capture area $\Rightarrow$ small bubble?

Histograms of particle size and capture volume

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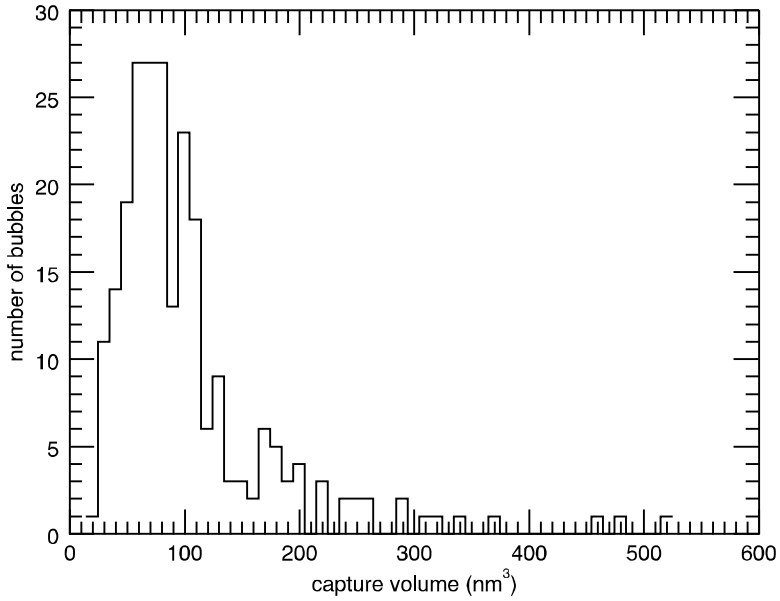
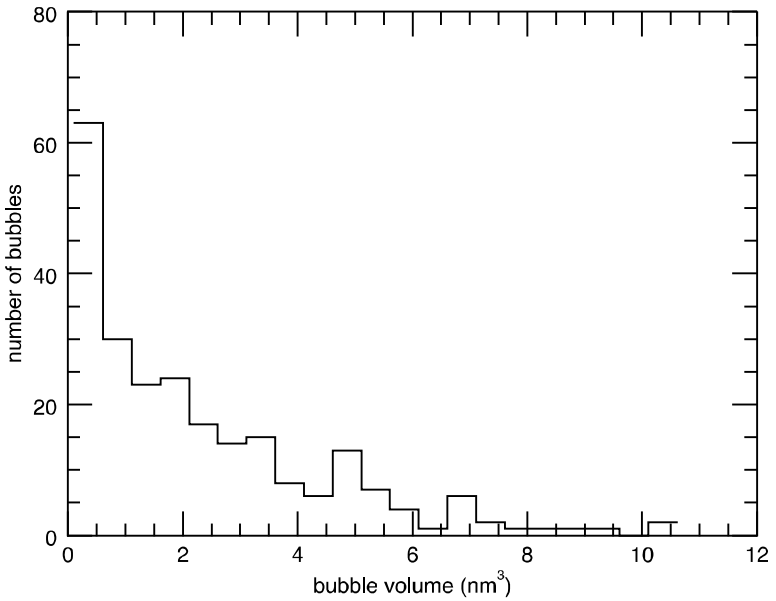
Capture Volume Distribution



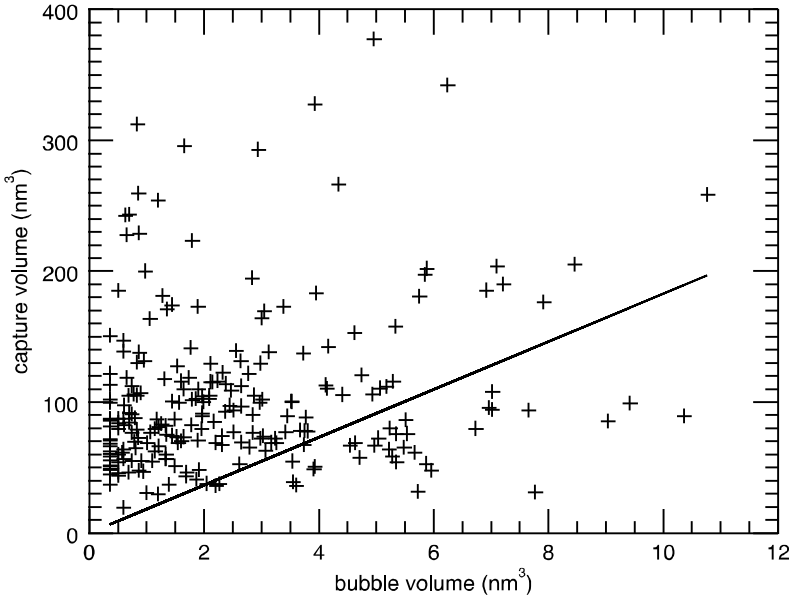
Bubble Volume Distribution



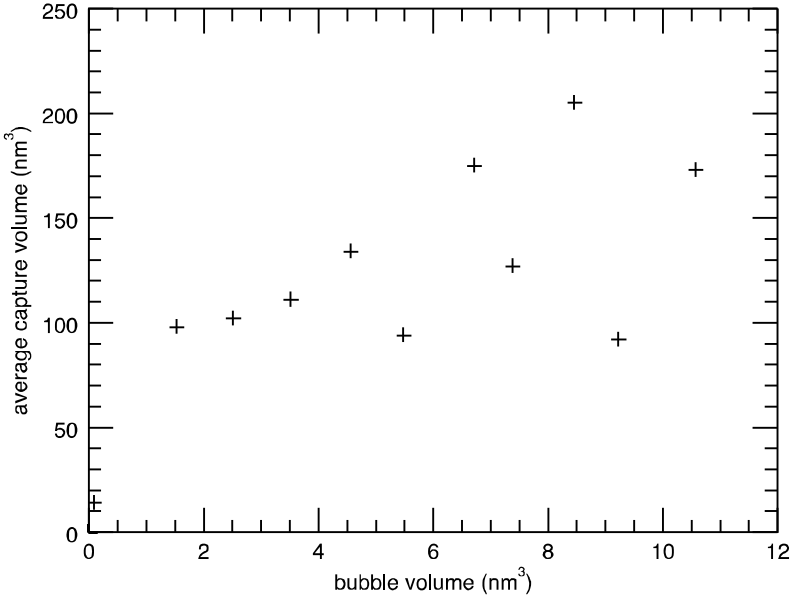
- Capture volume distribution is consistent with log normal distribution, but bubble volume distribution is not
- Large number of small bubbles $\Rightarrow$ Late nucleation?

Bubble and Capture Volume Correlation

Individual Capture Volume



Averaged Capture Volume



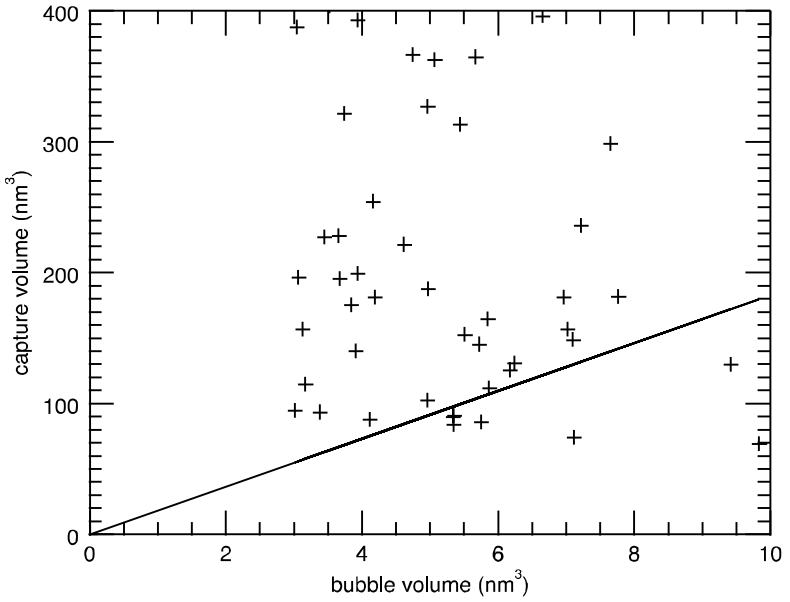
Solid line: expected for 3.8 year of tritium decay (He/Pd = 1.2) if bubbles have 5GPa pressure based on the loop punching growth

No bubble and capture volume correlation is found!

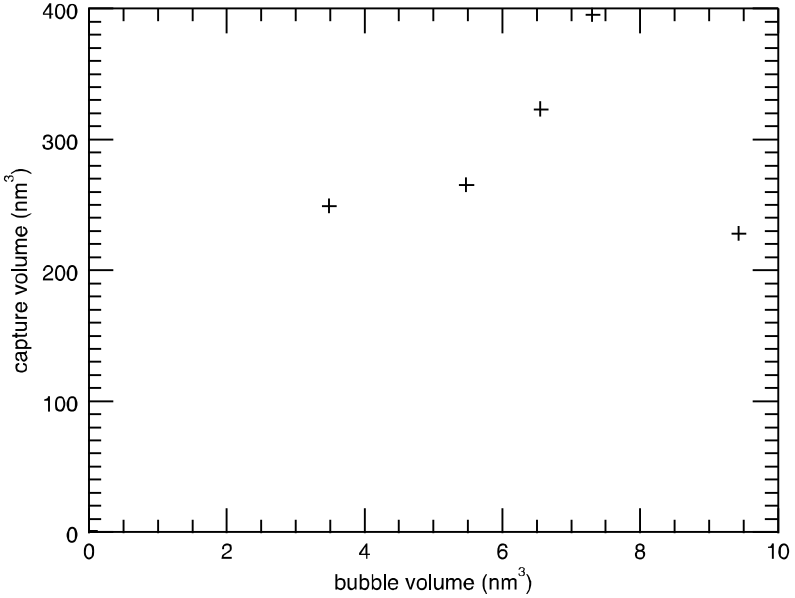
What If Small Bubbles Are Excluded?

Perhaps small bubbles nucleated late or reconstruction algorithm was inaccurate for small bubbles ⇒ excluding from the analysis all bubbles smaller than 3 nm^3

Individual Capture Volume



Averaged Capture Volume



Still no correlation!

Tentative Conclusions

- Early nucleation + diffusion limited growth of He bubbles do not explain all observations
- Lack of environmental dependence of growth rate suggests that there exists a barrier for He incorporation into bubbles
- Growth of small bubbles requires fast generation of Pd interstitials – a rate limiting process

Acknowledgement

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000. Work at Savannah River National Laboratory was performed under contract number DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Under and Overfocus

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Fresnel diffraction pattern of bubbles is bright when underfocused (left) and dark when overfocused (right), confirming presence of void spaces, and not other types of crystal defects