SAND2019-3689C

Electron Tomography and Energy Loss Spectroscopy of Helium Nanobubbles Formed in a Palladium Tritide

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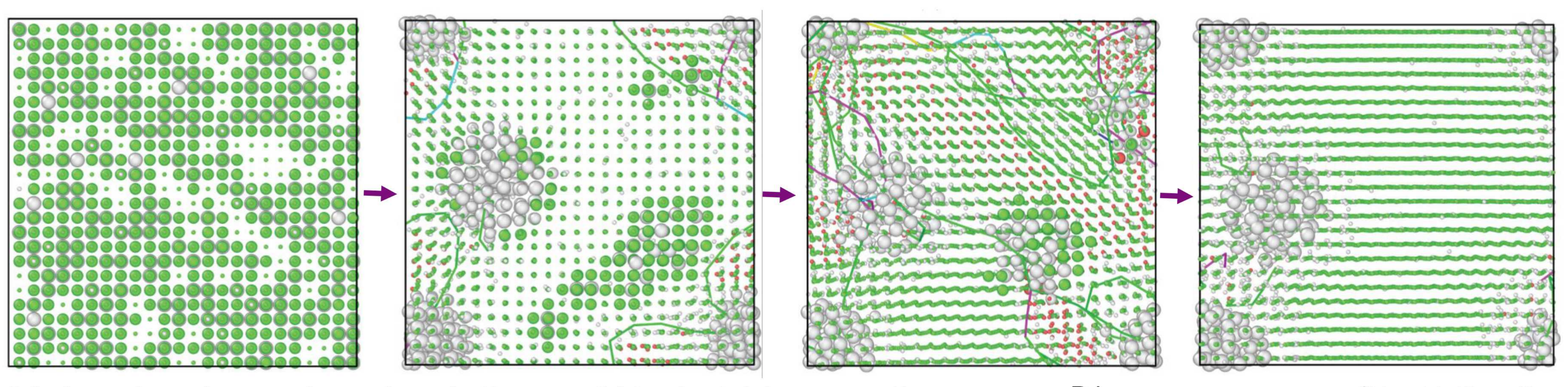
Background & Motivation

I

Helium nanobubbles are a common form of radiation damage that degrades mechanical and electrical properties of metals in tritium storage materials, nuclear fusion and fission reactors, and nuclear waste containers. Knowledge of the nucleation and growth mechanisms of helium bubbles is critical to predicting the lifetime of these materials. To better understand the process of bubble formation, we harnessed high resolution transmission electron microscopy to characterize the 3D configuration of helium bubbles in a palladium alloy aged as a tritide for 3.8 years. We generated a three-dimensional image of the sample by tomographic reconstruction and determined the number, volume, and spatial distribution of helium nanobubbles in the metal. By measuring the electron energy loss spectra of an ensemble of individual bubbles, we established the presence of helium in bubbles. These results inform development of a more accurate model of nanobubble nucleation and growth.

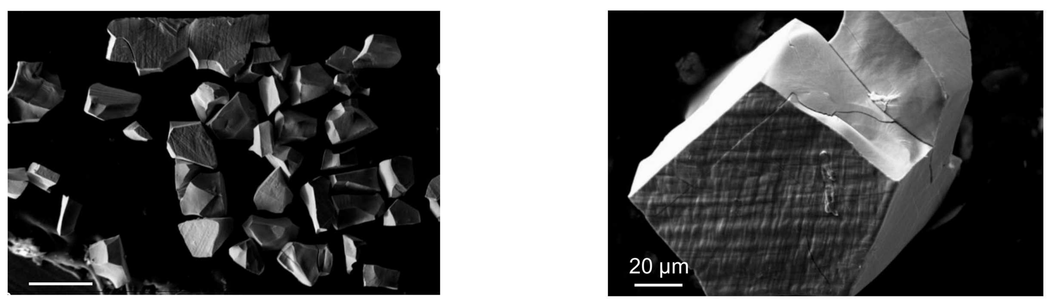


Molecular dynamics simulations of He bubble growth
Lines indicate dislocations

Preparation & Properties of Tritiated PdNi

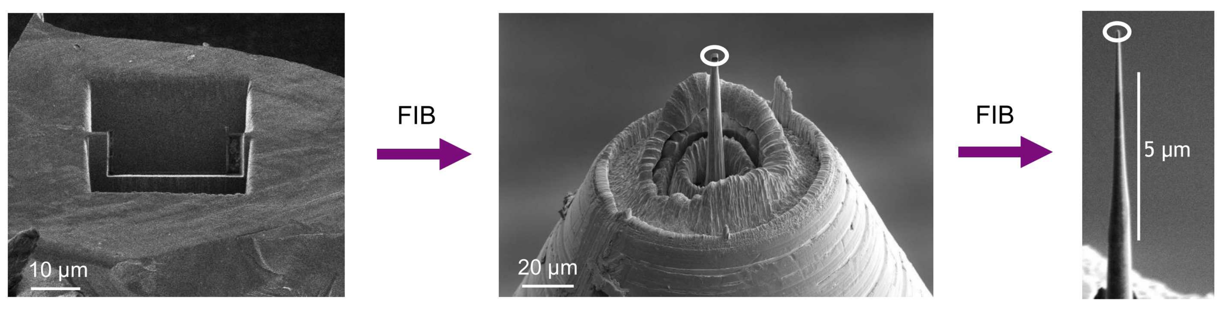
II

- $Pd_{95}Ni_5$ alloy was obtained as a foil.
- STEM-EDS indicates Ni is distributed homogeneously as a solid solution.
- $Pd_{95}Ni_5$ was aged as a tritide for 3.8 years with a tritium to metal ratio of 0.6.
- Tritium was removed by cycling in D_2 and vacuum near room temperature.
- Embrittlement, presumably due to He bubbles, caused fracture into particles.



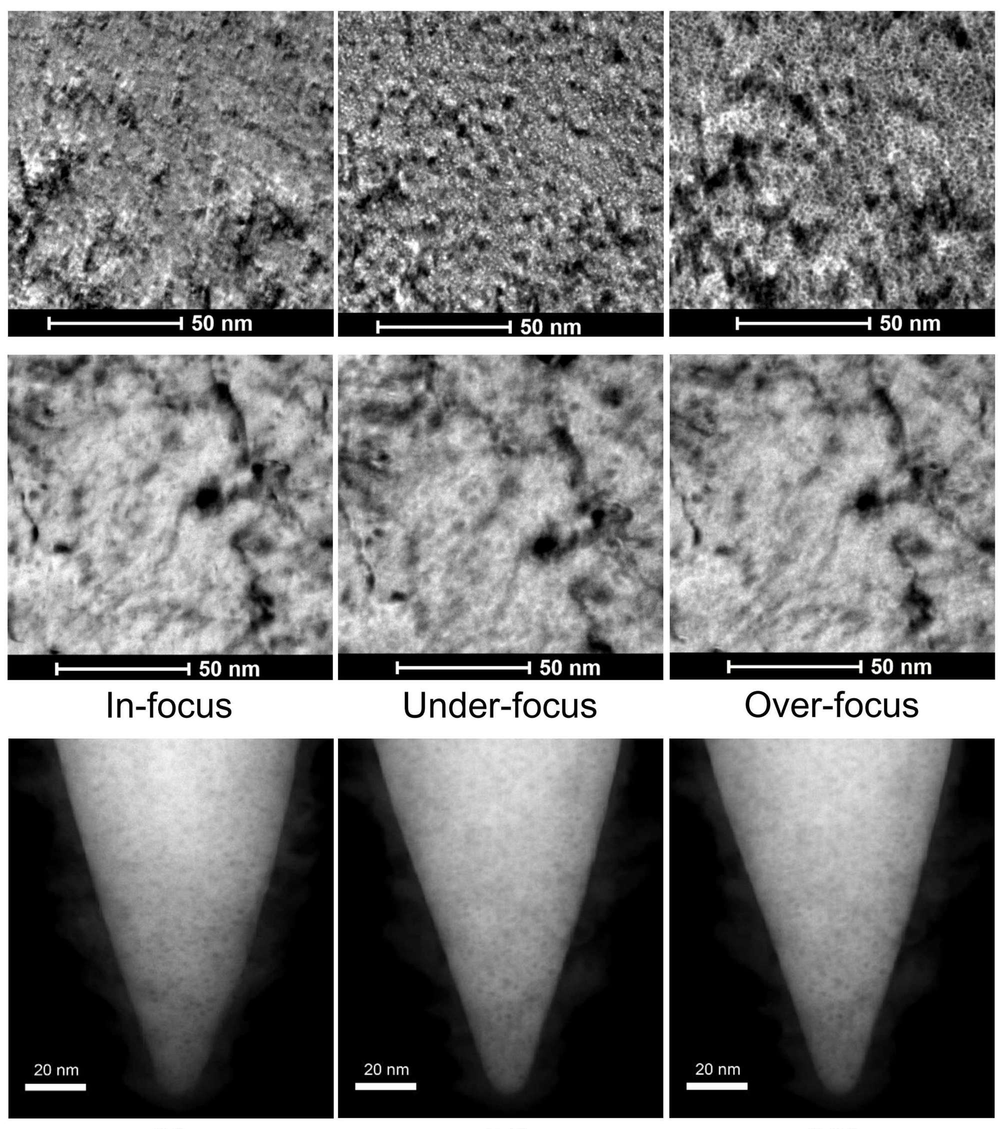
K.L. Shanahan, Fusion Sci. Tech. 71 555 (2017)

- Section of particle was lifted out, welded to post, and thinned with a FIB/SEM to produce a tip that could be imaged by TEM from a sequence of angles.



TEM Imaging

III



Round features 2 nm in diameter are He bubbles. Contrast inversion from under- to over-focus indicates these features have lower density than the Pd.

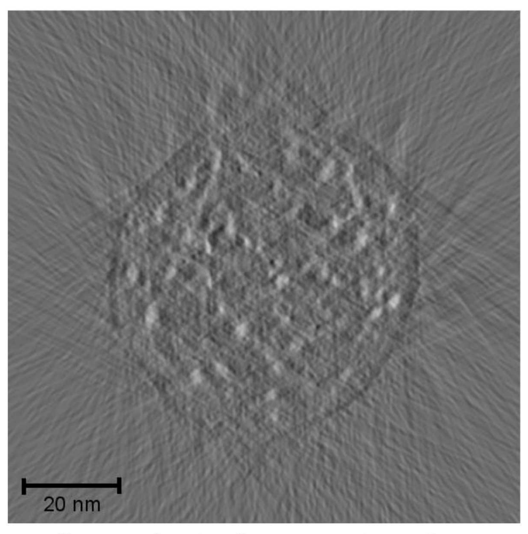
Round features and contrast inversion are absent from a non-tritiated $Pd_{95}Ni_5$ control sample.

The thinned $Pd_{95}Ni_5$ tip was rotated through 140° with an image collected every 1° to generate a set of projections for 3D tomographic reconstruction. Bubbles appear dark in HAADF STEM images.

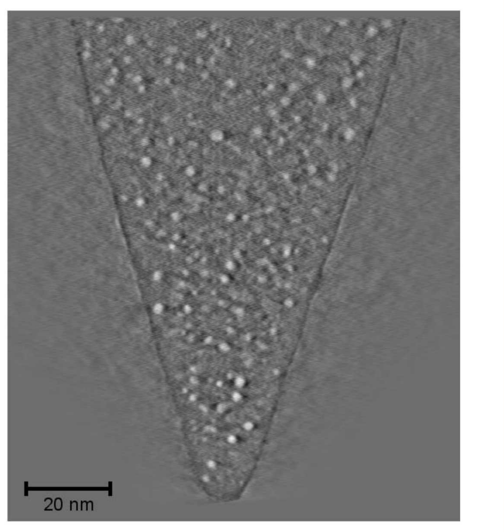
3D Reconstruction & Voronoi Tessellation

IV

A 3D image of the Pd tip was reconstructed from the tilt series of 2D micrographs by a weighted back-projection algorithm. The He bubbles appear bright after image processing.

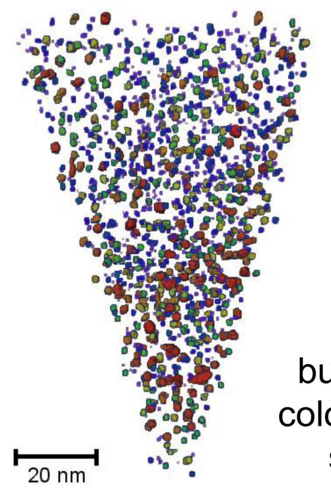


Snapshot of reconstruction perpendicular to tip axis

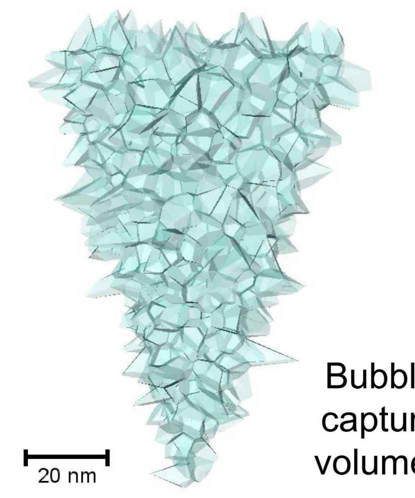


Snapshot of reconstruction parallel to tip axis

Thresholding the 3D image yielded ~1500 discrete bubbles. Voronoi tessellation of the Pd sample based on bubble positions generated a set of polyhedra representing the capture volumes of the bubbles.



He bubbles colored by size



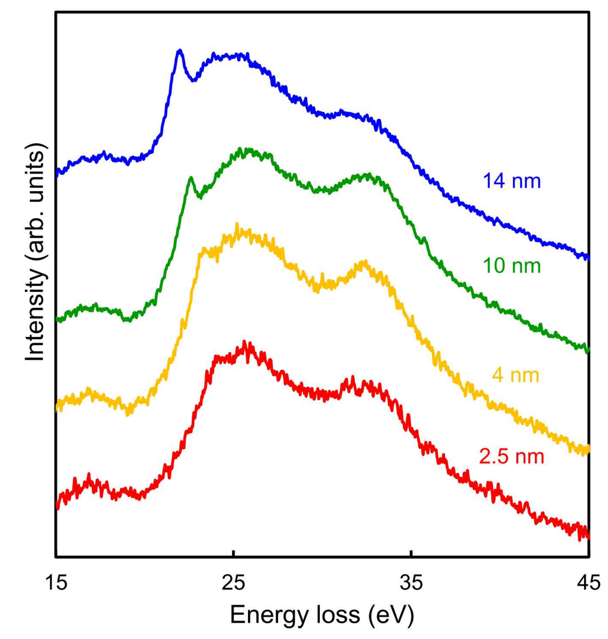
Bubble capture volumes

Electron Energy Loss Spectroscopy

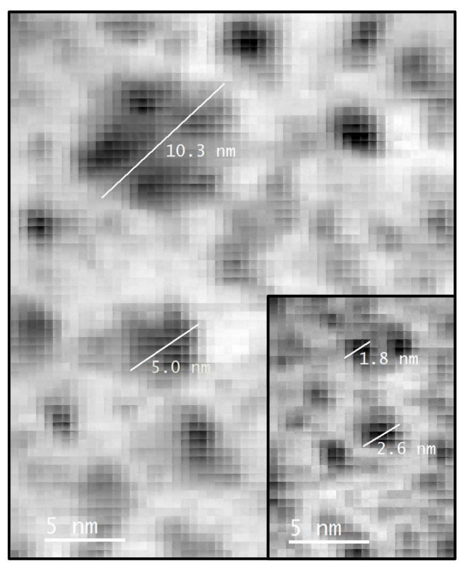
V

The He 1s → 2p transition absorbs energy from the electron beam to produce an "energy loss" peak. The excited state is less stable at high He density. He excitation occurs at higher eV with increasing He bubble pressure. The He peak of small, high pressure bubbles is obscured by metal plasmon peaks.

EELS of helium bubbles in $Pd_{95}Ni_5$

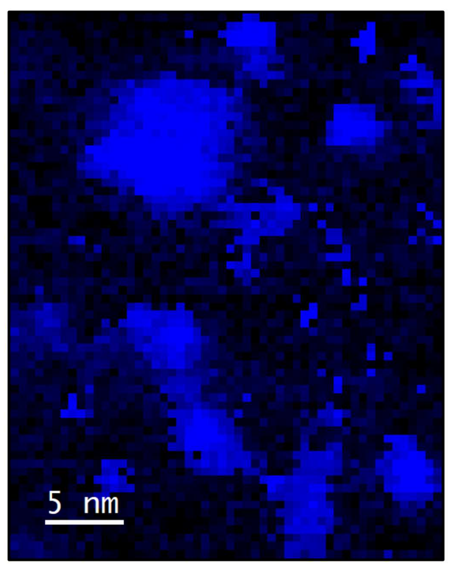


Dark field image



After heating   Before heating

Amplitude of He peak fit



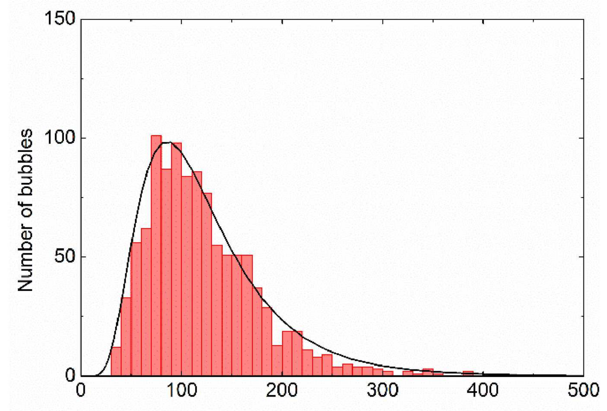
After heating

We heated the sample at 400 °C to coarsen 2 nm bubbles to a range of sizes. The He peak becomes clearer in larger bubbles. The amplitude of a Gaussian fit to the He peak at each pixel shows that high amplitudes correspond to the voids in the metal detected by dark field imaging, confirming that they are He bubbles.

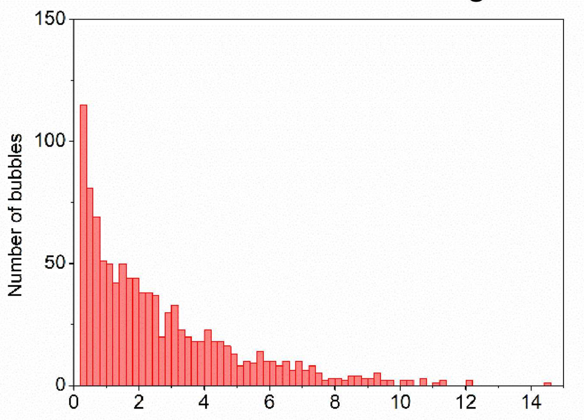
Data Analysis & Conclusions

VI

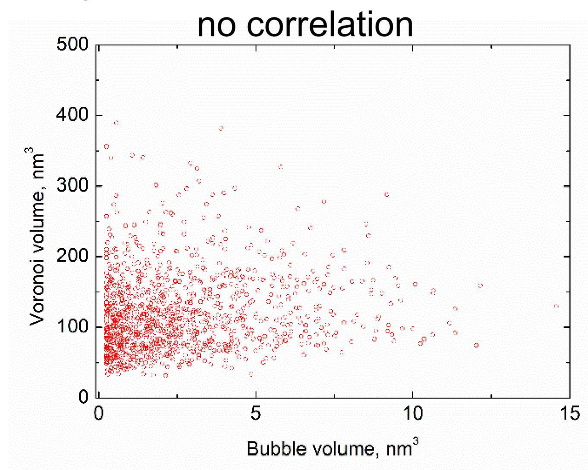
Capture volume distribution: log-normal



Bubble volume distribution: not log-normal



Capture volume & bubble volume: no correlation



- *Capture volume distribution is log-normal:* Nucleation is spatially random and homogeneous within the metal
- *Bubble volume distribution is not log-normal:* Raises possibilities of late stage and ongoing nucleation of bubbles.
- *No correlation between capture volume and bubble volume*: Bubble size is not determined by proximity to other bubbles.

Bubble evolution is more complex than predicted by a diffusion-limited growth model assuming early-stage nucleation within a narrow time window.

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