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Sandia National Laboratories

3D Mapping of Helium Nanobubbles in a Pd Alloy by Electron Tomography

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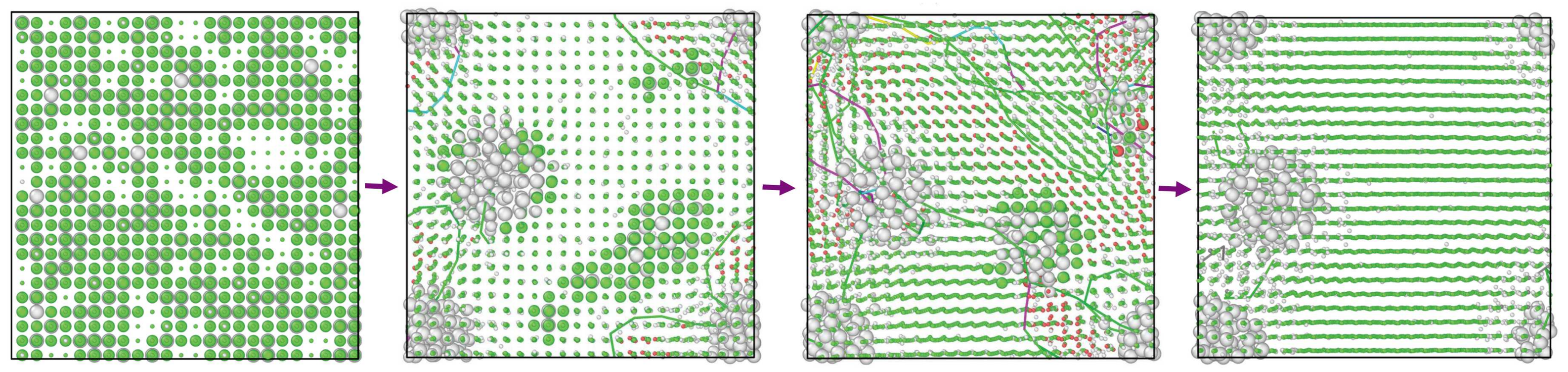
Background & Motivation

I

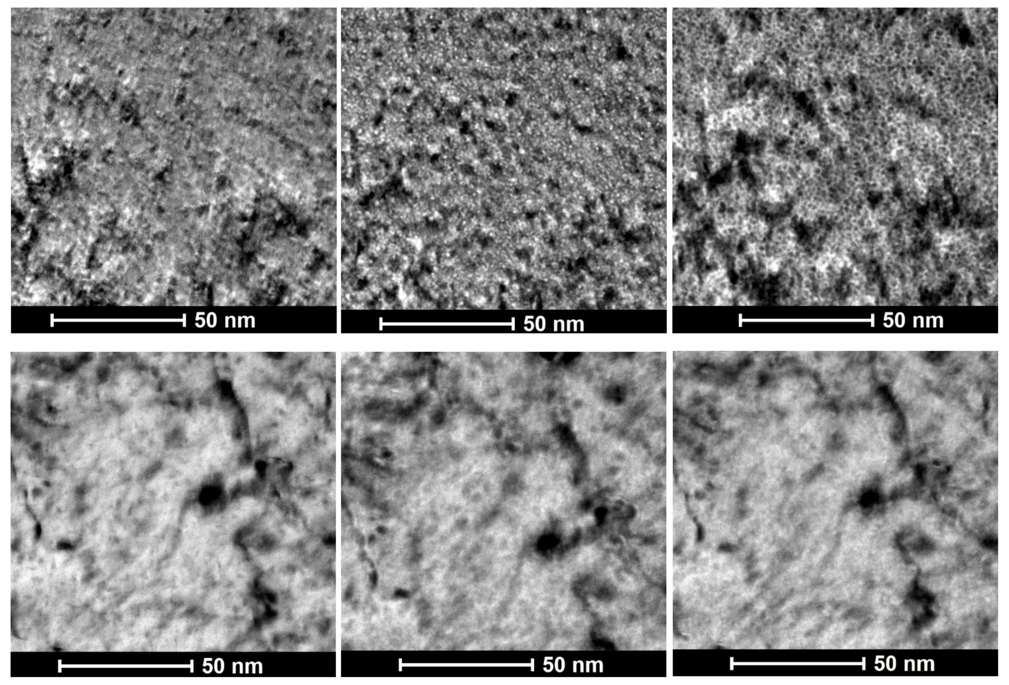
Helium nanobubbles are a common form of radiation damage that degrades mechanical and electrical properties of metals in semiconductors, nuclear reactors, tritium storage materials, and nuclear waste containers. Knowledge of the nucleation and growth mechanisms of helium bubbles is critical to predicting 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 4 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 a pressure range of a few GPa. We then calculated the total helium content trapped in the metal lattice as nanobubbles. These results inform development of a more accurate model of nanobubble nucleation and growth.



Molecular dynamics simulations of He bubble growth

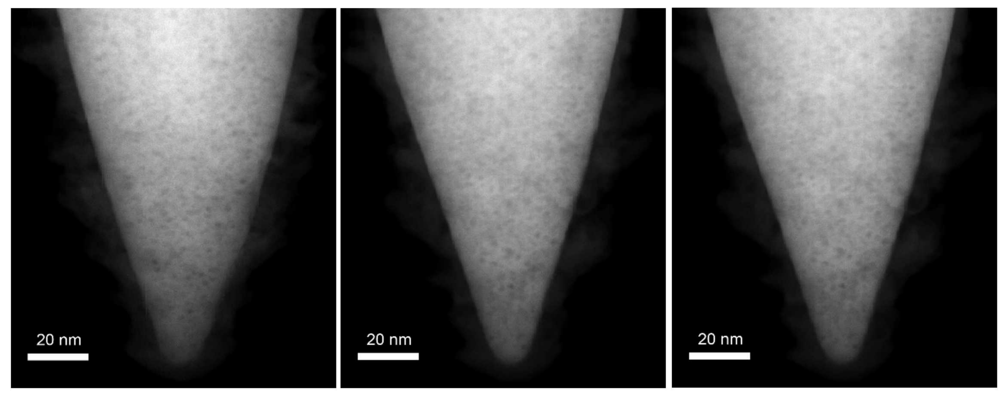
TEM Imaging

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.

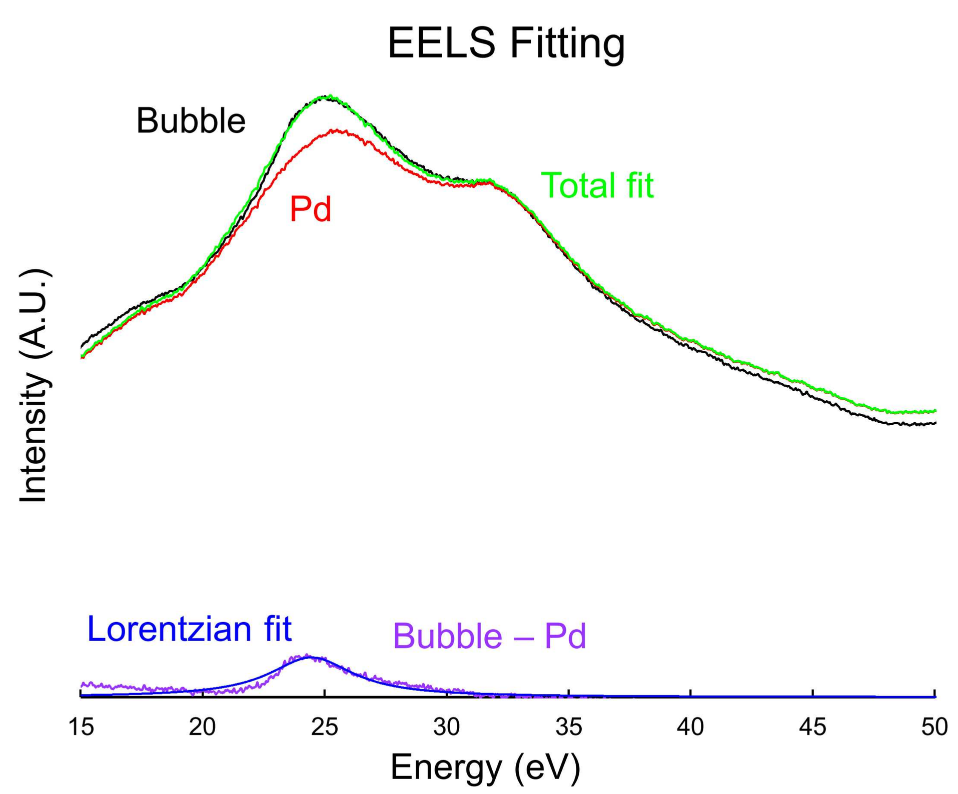
Round features and contrast inversion are absent from a non-tritiated $Pd_{95}Ni_5$ control sample.



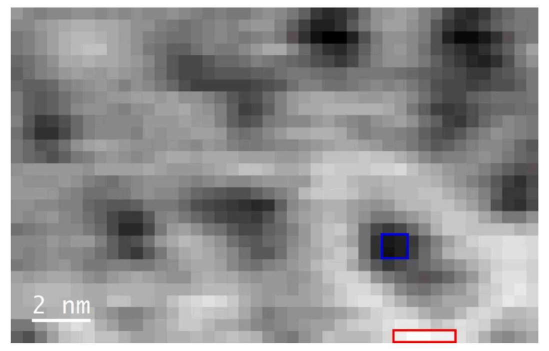
The thinned $Pd_{95}Ni_5$ tip was rotated through 140° with an image collected every 1° to generate a set of projections for 3D tomographic reconstruction. Bubbles appear dark in HAADF images.

Electron Energy Loss Spectroscopy

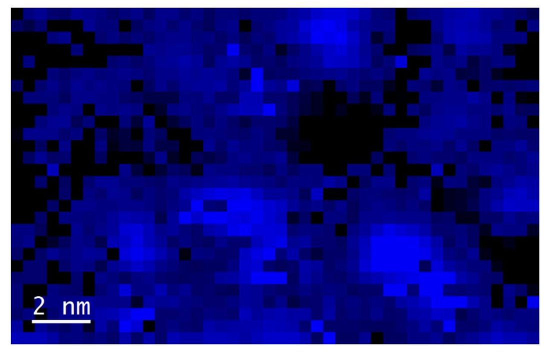
V



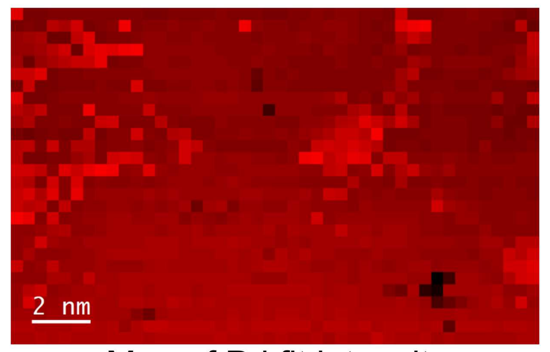
An EELS spectrum image of the $Pd_{95}Ni_5$ alloy was fit with the background spectrum of the metal. Mapping the residual signal reveals the location of bubbles.



ADF image



Map of Lorentzian fit intensity

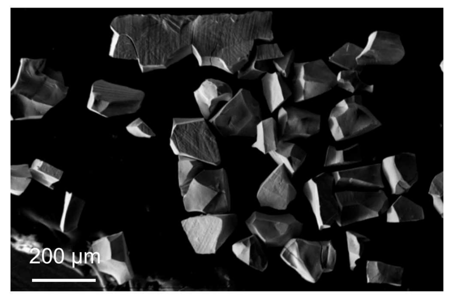


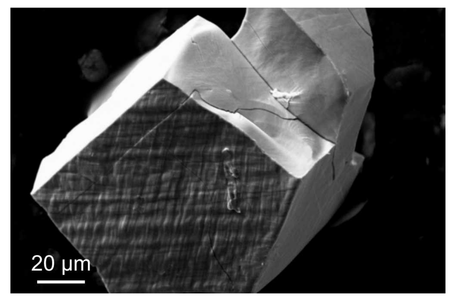
Map of Pd fit intensity

Preparation & Properties of Tritiated PdNi

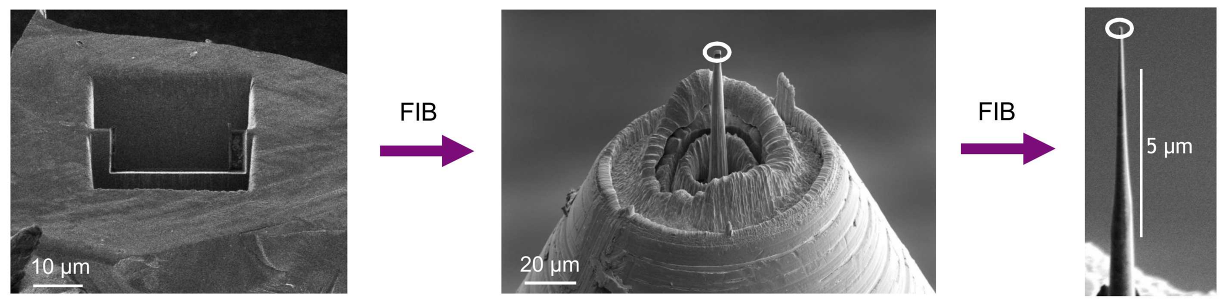
II

- $Pd_{95}Ni_5$ alloy was obtained as a foil
- EDS indicates Ni is distributed homogenously as a solid solution
- $Pd_{95}Ni_5$ aged as a tritide for 3.8 years with a tritium to metal ratio of 0.6
- Tritium was removed by cycles of exposure to D_2 and vacuum at RT
- Embrittlement, presumably due to He bubbles, caused fracture into particles





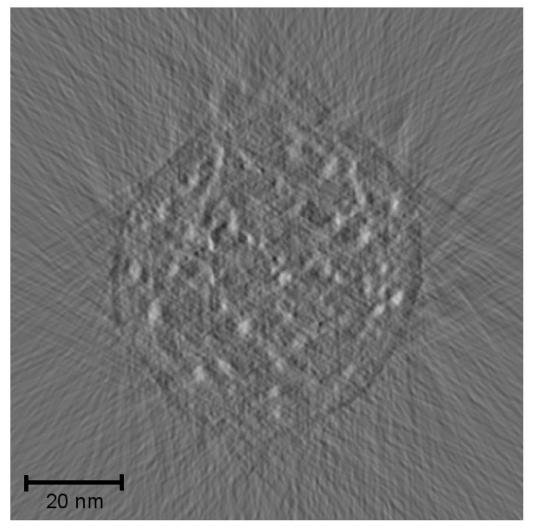
- Section of particle was lifted out, welded to post, and thinned with a FIB/SEM to produce a tip that could be imaged by TEM from a sequence of angles



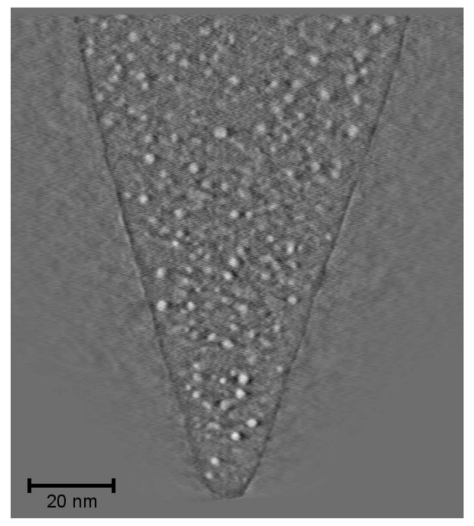
3D Reconstruction & Voronoi Tessellation

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 brighter than the surrounding Pd matrix.

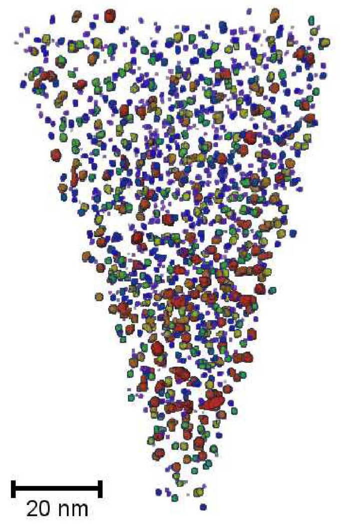


Snapshot of reconstruction perpendicular to tip axis



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.



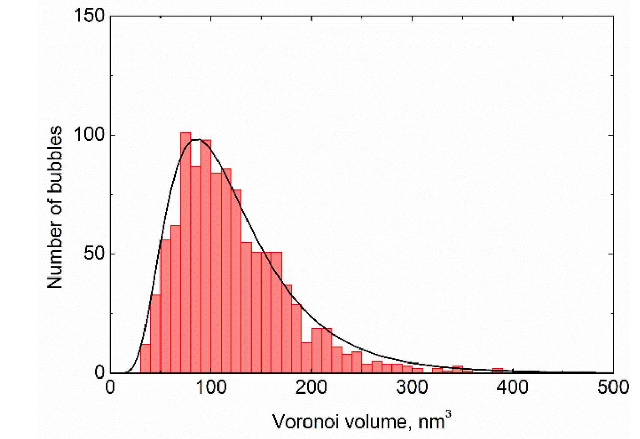
He bubbles colored by size

Bubble capture volumes

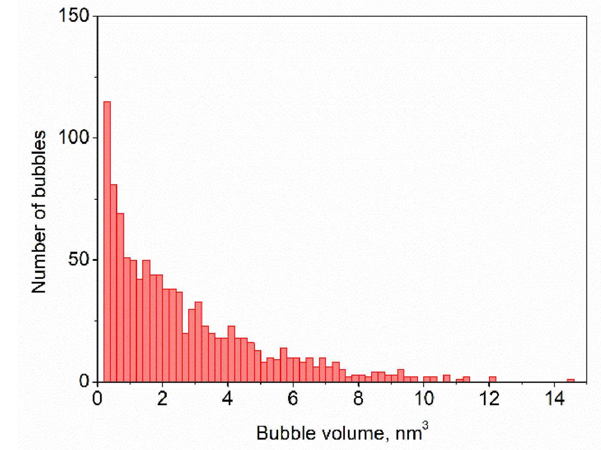
Data Analysis & Conclusions

VI

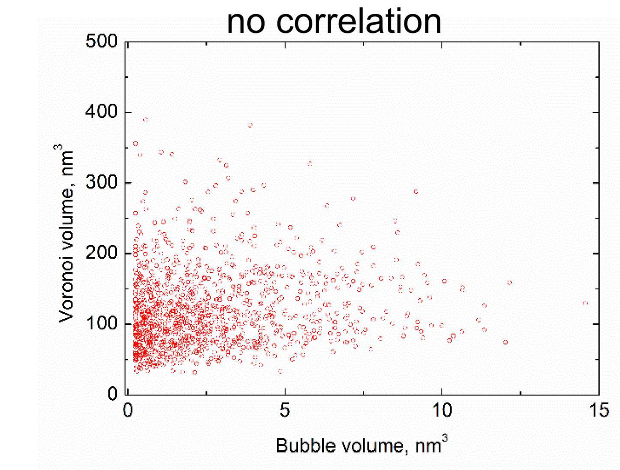
Capture volume distribution: log-normal



Bubble volume distribution: not log-normal



Capture volume & bubble volume: no correlation



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

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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