Nucleation and Growth of Helium Nanobubbles in Palladium Alloy Tritides

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Helium nanobubbles were found in a PdNi tritide

Thinned section of a PdNi alloy previously exposed to tritium

Bubbles appear dark

Bubbles are a few nm in diameter and spaced a few nm apart
Tritium decays to insoluble helium-3 within metal lattices

$^3$H decays to $^3$He without damage to lattice

$^3$He occupies interstices

Interstitial $^3$He diffuses rapidly

$^3$He becomes trapped at point defects

- Pd
- Pd with $^3$He in $O_y$ hole
- $^3$He at lattice site
- $^1$H
Helium-3 forms nanobubbles that cause defects in metal

$^3$He forms clusters

Clusters displace metal atoms and cause dislocations

Clusters diffuse within lattice

Clusters form bubbles that grow in volume

- Pd
- Pd with $^3$He in $O_y$ hole
- $^3$He at lattice site
- $^1$H
Open questions of helium bubble nucleation and growth

Questions
- What determines helium bubble size and spatial distribution?
- Do all helium bubbles nucleate within a narrow time range?
- What mediates bubble nucleation?

Goals
- To develop a model for helium bubble formation and evolution
- To validate model with experimental observations

Approach
- Generate three-dimensional maps of helium bubbles trapped in metals by electron tomography
- Detect helium and measure pressure by electron energy loss spectroscopy (EELS)

Overarching aim is to develop a well-validated, comprehensive model for the nucleation, growth, and release of helium bubbles in metals

PdNi foil was exposed to tritium, aged, and decontaminated

Sample characteristics

5 atom % Ni solid solution in Pd
Aged 3.8 years under tritium
He/Pd ratio: 0.12
Tritium removed for analysis by cycling D₂ and vacuum near room temperature
40 μCi / g by dissolution followed by liquid scintillation counting

Fresnel contrast seen in tritium-exposed sample but not in control

Under-focused images

Over-focused images

Change in contrast with focus indicates round features (bubbles) are regions of lower density. No contrast reversal occurs in the PdNi control sample that was never exposed to tritium.
Electron energy loss spectroscopy detects He in He$^+$ implanted Pd

Pd implanted with 1 x 10$^{17}$ helium ions / cm$^2$
Annealed at 600°C for 2 hours
Large, sparse, low-pressure bubbles
He signal maps on to bubble locations

Electron energy loss spectroscopy detects He in tritium-exposed PdNi

PdNi was thinned to a tip for collection of tomography data set

Measurement of 3D bubble volumes and locations are needed for validation of He transport models

Specimen from PdNi particle was extracted and thinned with a focused ion beam to acquire a series of images at various angles by tilting sample
Set of images for tomographic reconstruction

First set of images:
- 2D high-angle annular dark-field STEM images
- Raw images from microscope
- Helium bubbles appear dark

Second set of images:
- Filtered, contrast-inverted images
- Low-frequency background removed
- Basis for Simultaneous Iterative Reconstruction Technique (SIRT) algorithm

Series of images in 1° increments through 140°
Visualization of helium bubble-impregnated PdNi tip parallel to tip axis
Visualization of helium bubble-impregnated PdNi tip along tip axis
Thresholding of reconstruction yields discrete bubbles of differing sizes

1248 bubbles
Colored from largest (red) to smallest (blue)

Capture volume is described by Voronoi tessellation
Larger capture volume → larger bubble?

Spacing of bubbles yields capture volumes of various sizes

Outlines of Voronoi polyhedra formed by bubbles

Voronoi polyhedra expanded for visualization

PdNi tip was divided into polyhedra based on distribution of bubbles

Each bubble has a corresponding polyhedron representing its capture volume
Experimental results differ from predictions of capture volume theory

Capture volume and bubble volume: no correlation

Voronoi volume distribution: log-normal

Bubble volume distribution: not log-normal

Theory suggests:
- Capture volume and bubble volume adhere to log-normal distributions
- Capture volume determines bubble volume
Conclusions

What determines helium bubble size distribution?
No correlation between capture volume and bubble volume → Bubble size is not primarily determined by proximity to other bubbles

What determines helium bubble spatial distribution?
Capture volume distribution is log-normal → Nucleation is spatially random and homogeneous

Do all helium bubbles nucleate within a narrow time range?
Bubble size distribution is not log-normal → Raises possibility of late nucleation of bubbles
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