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Modeling of Hydrogen Interactions with Helium Bubbles in Tungsten

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Outline

- Motivation
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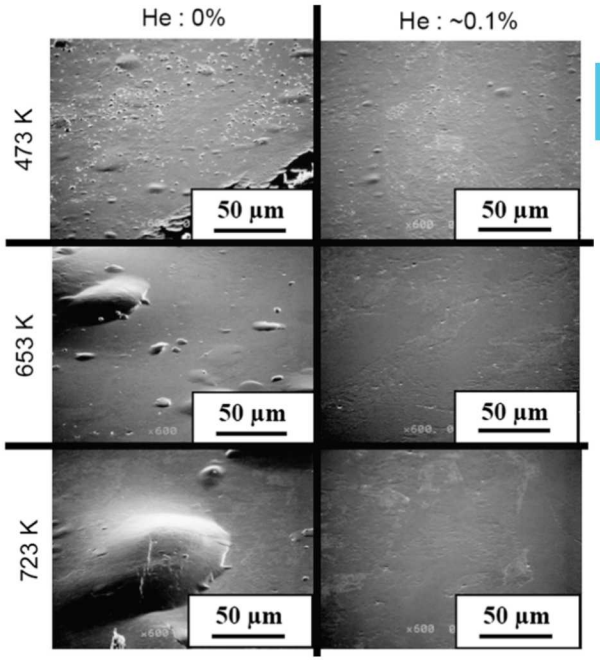
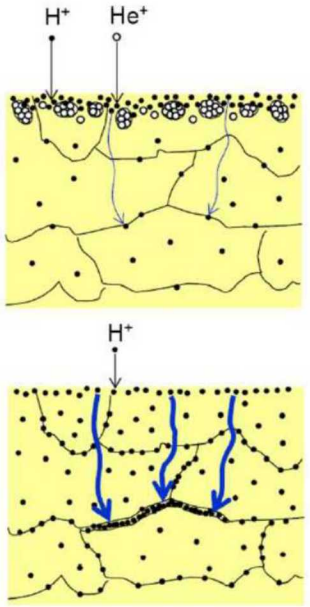
  - Methods
  - Results
- Large-scale W-H-He Simulations
  - Methods
  - W-H-He Results
- Experimental Results
- Conclusions

Motivation

- Tungsten is a candidate material for the divertor of ITER and other future fusion reactors
- Divertor will be subject to high temperatures and particle fluxes of both hydrogen and helium
- Under hydrogen implantation, micron sized blisters on the surface have been observed
- The presence of helium in the plasma, under simultaneous or subsequent helium-hydrogen irradiation, suppresses blisters
- It is hypothesized that the helium bubble layer prevents hydrogen from diffusing deeper into the material to form blisters, but could more strongly trap hydrogen
- Here, we describe MD simulations to investigate He-H synergies in W



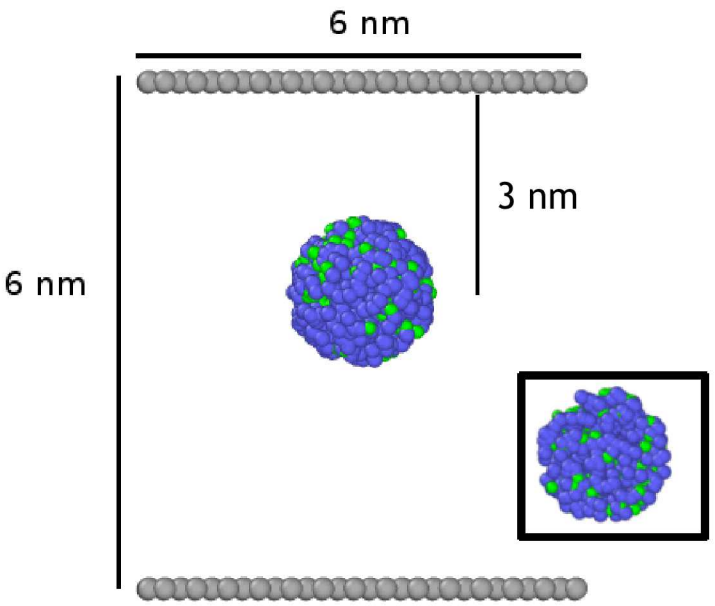


Y. Ueda, et al. *Journal of Nuclear Materials* 386-388, 725 (2009)

Subsurface Mixed H-He Bubble Methods

- 6 nm x 6 nm x 6 nm simulation cell
- Cavity first created with a 1 nm radius and located 3 nm below the surface
- Concentrations of 3,3.5, or 4 He/V and 0.5 or 1 H/V are randomly distributed within the bubble
- Simulation evolved for 100 ps and diffusion of hydrogen is assessed
- Temperatures of 1200 K, 1500 K, 1800 K, and 2000 K
- (110) and (111) surface orientations
- Tersoff bond order potential for W-H interactions developed by Juslin et al. [1] and modified for H_2 desorption by Guterl et al. [2]

Blue: Helium
Green: Hydrogen
Grey: Tungsten

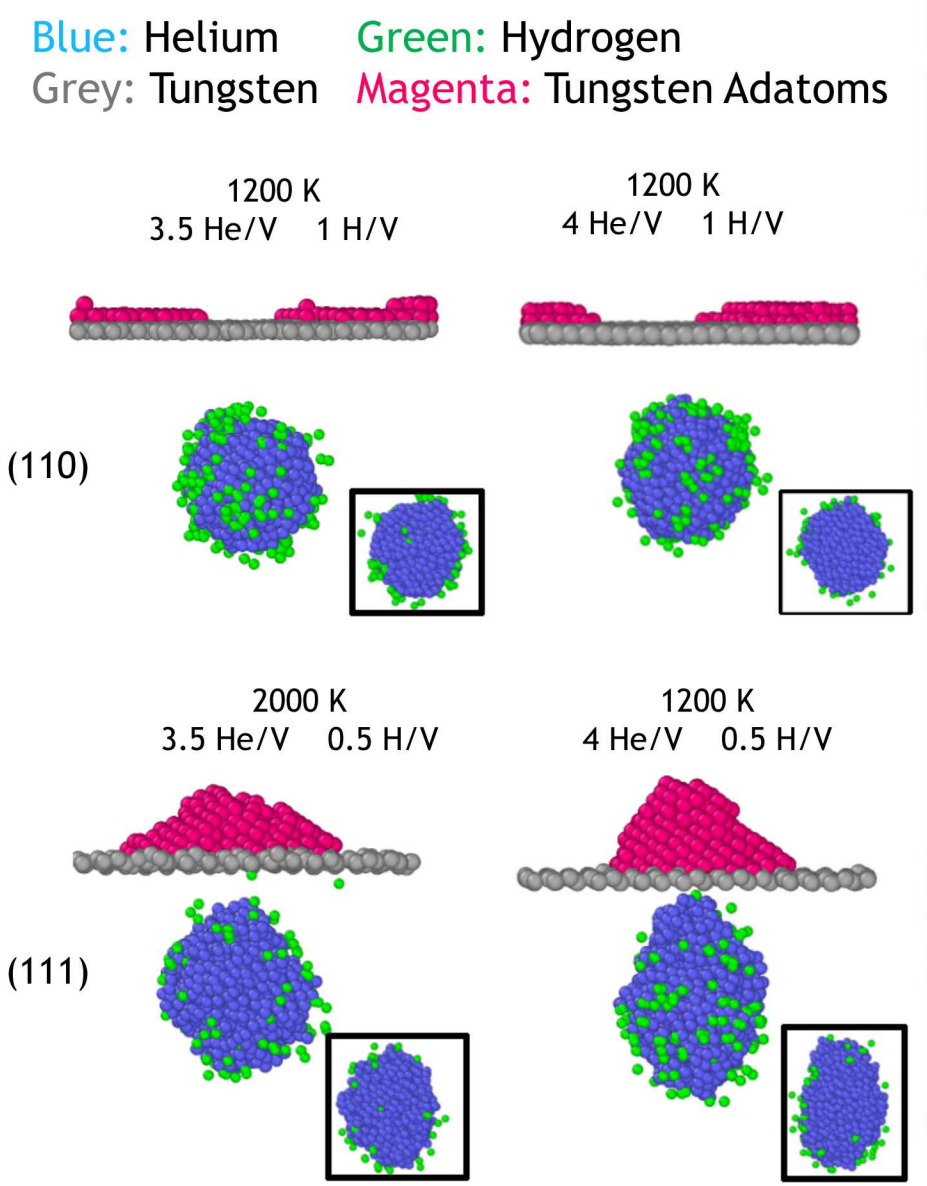


Initial simulation setup with inset image of a slice through the bubble

[1] N. Juslin, et al., J. Applied Physics. 98, 123520 (2005)
[2] J. Guterl, et al., J. Nucl. Mater. 463, 263-267 (2015)

Atomistic Snapshots after 100 ps

- Initially over-pressurized bubble leads to expansion
- Bubble expansion and loop punching produces adatom formations consistent with previous MD work of helium bubbles
- Expansion occurs within first few ps
- More surface deformation and faster loop punching at higher temperatures and helium concentrations
- Bubble evolution independent of hydrogen
- Adatom/surface patterning depends on surface orientation
- Significant diffusion of hydrogen to helium bubble periphery
- Hydrogen segregation to the bubble edges is observed for all simulations regardless of initial parameters

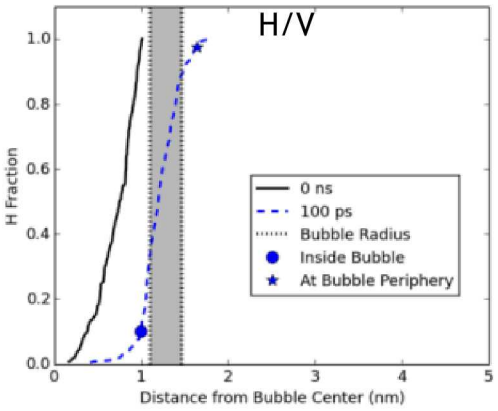


Hydrogen Cumulative Radial Distributions

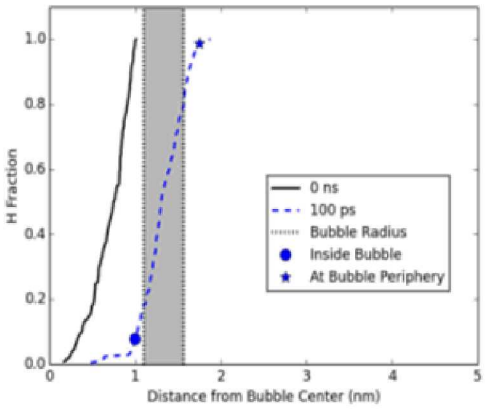
- Plots of H distribution from bubble center
- Black line is at 0 ps and blue is at 100 ps
- Gray bar represents range of bubble radius due to non-isotropic expansion
- Circle and star represent hydrogen in the bubble and at the periphery respectively
- Large portion of hydrogen is located at the bubble periphery, ~75-95% (17-22 H/nm^3)
- Typically less than 20% remains in the bubble interior and is mostly H_2
- Standoff distance between helium bubble and tungsten matrix creates excess volume that could be a low energy site for hydrogen, which prefers vacancy-like defects

(110)

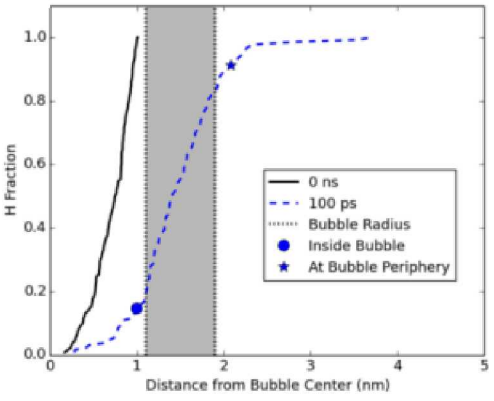
1200 K
3.5 He/V 1 H/V



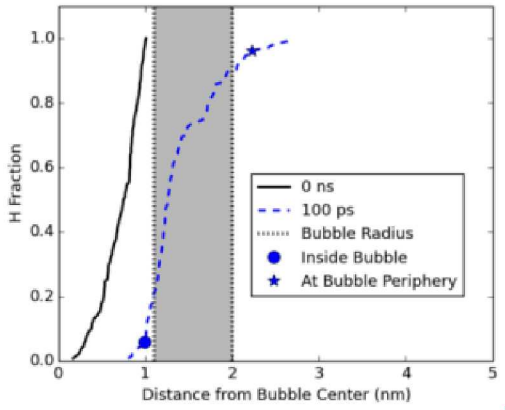
1200 K
4 He/V 1 H/V



(111)



2000 K
3.5 He/V 0.5 H/V



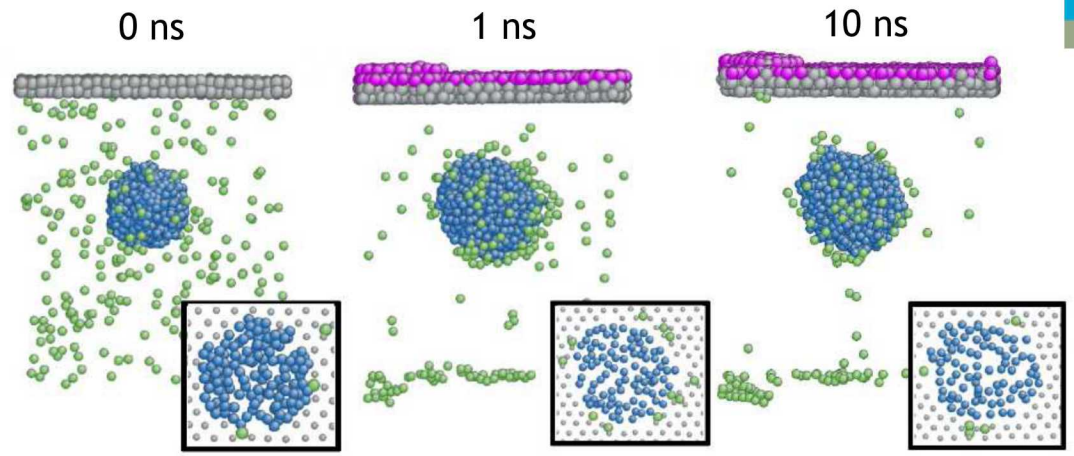
1200 K
4 He/V 0.5 H/V

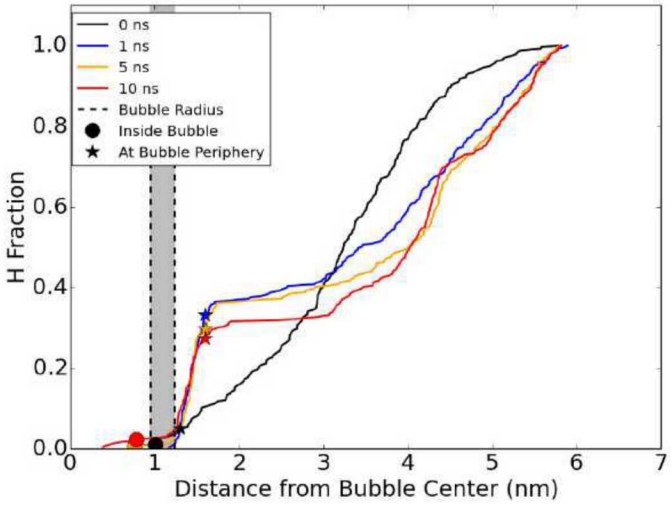
Placing the H Initially Throughout the Box

- Initially place hydrogen randomly throughout the simulation cell
- Simulation run for 10 ns to allow enough time for hydrogen to diffuse
- Hydrogen still diffuses to bubble periphery, ~25-40% of total inventory depending on initial temperature
- Hydrogen very rapidly accumulates at the bubble periphery within first nanosecond and remains there for rest of simulation
- Additional hydrogen at either top or bottom surface
- Hydrogen concentration at bubble periphery higher than at the surface, 4.5 H/nm^3 vs. 1.5 H/nm^3

Blue: Helium Green: Hydrogen
Grey: Tungsten Magenta: Tungsten Adatoms

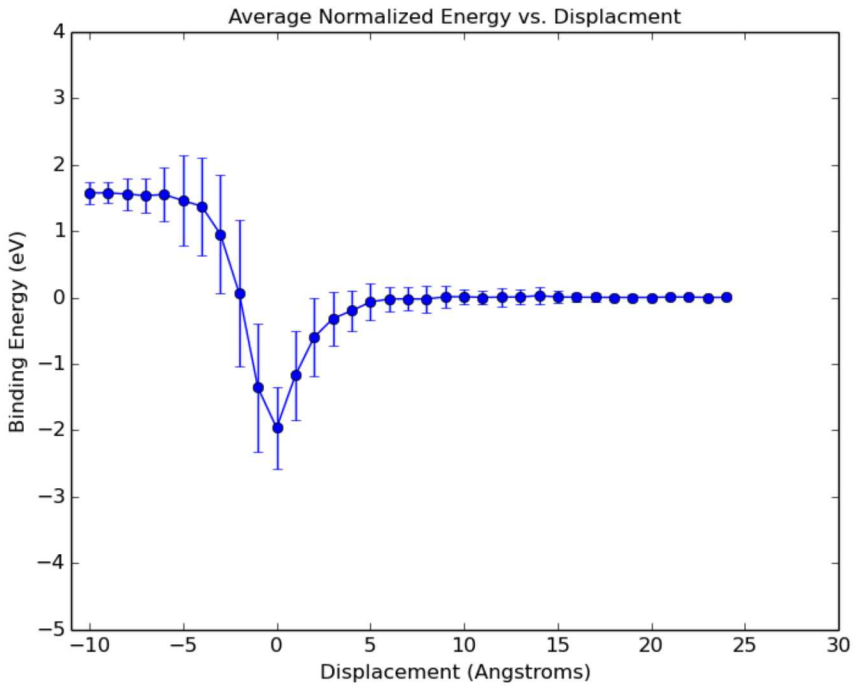
(110) Surface at 1800 K and 3 He/V and 1 H/V





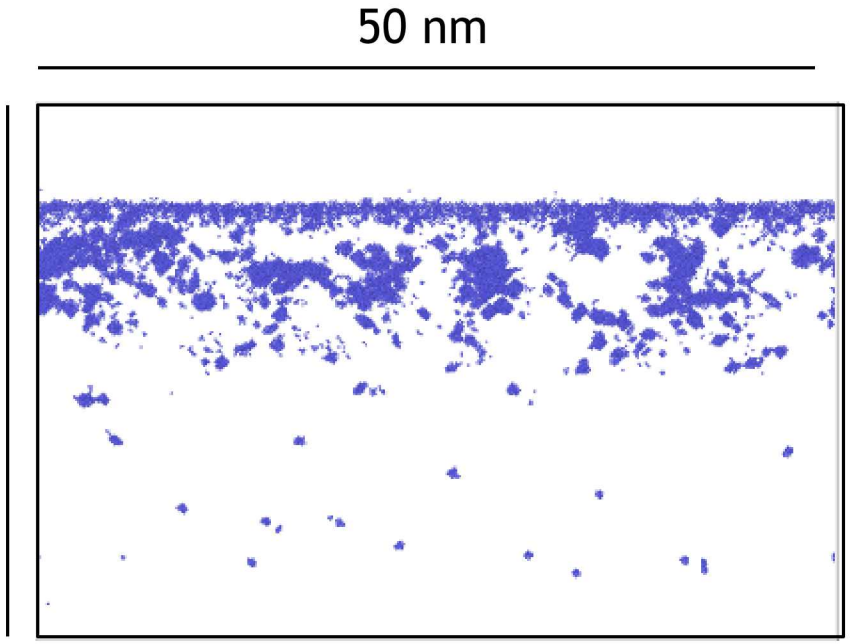
Binding/Trapping Energy Calculations

- Simulations quenched and minimized after 100 ps
- Hydrogen atoms around the bubble are then moved in 1 Å steps, both towards and away from the bubble, and the energy is minimized and recorded
- The energy at each step is normalized to the energy in the bulk and plotted for ~100 hydrogen atoms from 20 different simulations
- There is a clear trapping location at the bubble periphery with a binding energy of 1.5-2.5 eV The high energy within the helium bubble itself will drive the hydrogen out of the bubble
- Moving hydrogen into the bubble is repulsive
- A binding energy of ~2 eV and an activation energy of ~0.3 eV (migration energy of interstitial H), implies desorption temperatures > 1000 K -- the energy needed to overcome this potential well could be as high as ~2.3 eV



Large Scale Hydrogen and Hydrogen-Helium Modeling

- Large scale 50 nm x 50 nm x 25 nm box with a free surface in the z direction
- ~4 million tungsten atoms with initial helium implantation to a fluence of $\sim 10^{19}$ m^{-2}
- Hydrogen implanted at thermal energy based on 60 eV depth distribution from SRIM at an effective flux of $\sim 10^{25}$ $m^{-2}s^{-1}$
- Compare hydrogen implantation in pure tungsten and helium irradiated tungsten
- 933 K
- Two different hydrogen potentials compared
  - Tersoff bond order potential for W-H interactions developed by Juslin et al. [1] and modified for H_2 desorption by Guterl et al. [2]
  - EAM W-H potential developed by Wang, et al [3]



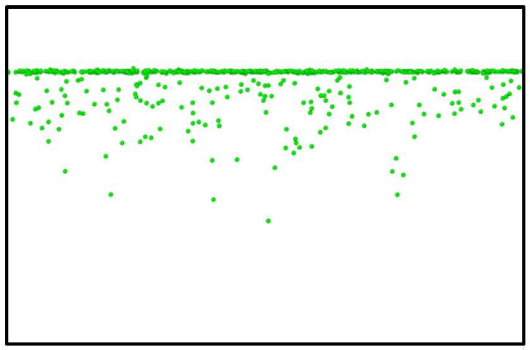
Initial He distribution for W-H-He simulations

[1] N. Juslin, et al., J. Applied Physics. 98, 123520 (2005)
[2] J. Guterl, et al., J. Nucl. Mater. 463, 263-267 (2015)
[3] XXXX

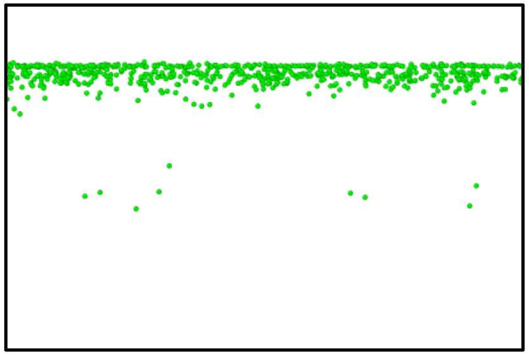
W-H and W-H-He at a fluence of 2.8 x 10^{17} m^{-2}

Pure Tungsten

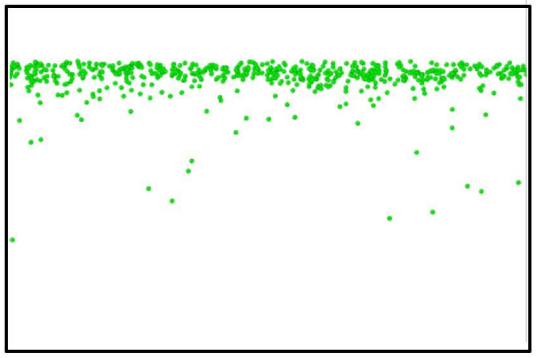
He Irradiated



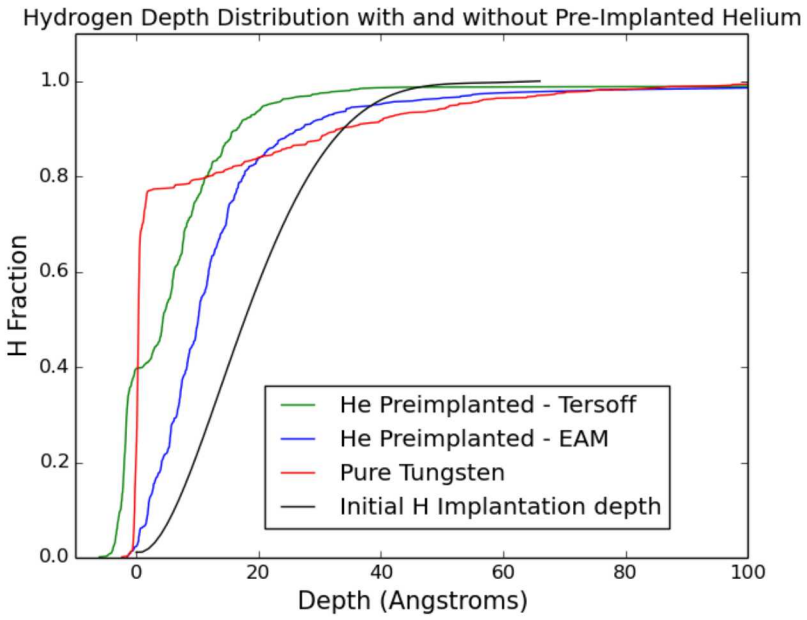
Tersoff



Tersoff



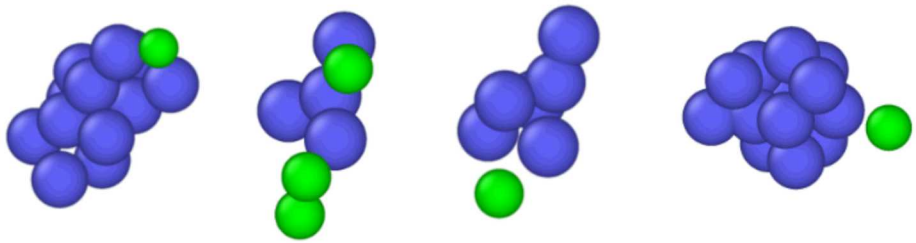
EAM



- Hydrogen depth distributions modified in presence of helium
- In pure tungsten case, ~80% of hydrogen is located directly at the surface and some diffuses deeper into the bulk
- In the helium pre-implanted case, the hydrogen is preferentially located in the helium bubble layer 5-10 Angstroms below the surface, about 60% and 80% for the Tersoff and EAM potential respectively
- More hydrogen has diffused deeper in the material for the pure tungsten case. Very few, about ~10 hydrogen atoms, have diffused passed the bubble layer in the pre-implanted cases

H Binding to He Clusters (Observed Clusters from MD)

Snapshots of H – He cluster interactions in MD simulations with pre-implanted He



Blue: Helium
Green: Hydrogen

Hydrogen Binding Energies to Small Clusters		
Cluster Identified Using Tersoff		
Configuration	Tersoff Binding Energy (eV)	EAM Binding Energy (eV)
$He_{40}H_2V_{23}$	0.26	0.25
$He_{16}H_1V_3$	1.71	0.32
$He_{11}H_1V_1$	1.11	0.82
$He_4H_1V_1$	1.48	0.79
Clusters Identified Using EAM		
Configuration	Tersoff Binding Energy (eV)	EAM Binding Energy (eV)
$He_{145}H_1V_{23}$	0.31	0.86
$He_{20}H_1V_4$	1.12	0.93
$He_{19}H_1V_3$	0.63	0.94
$He_9H_1V_1$	0.67	0.28

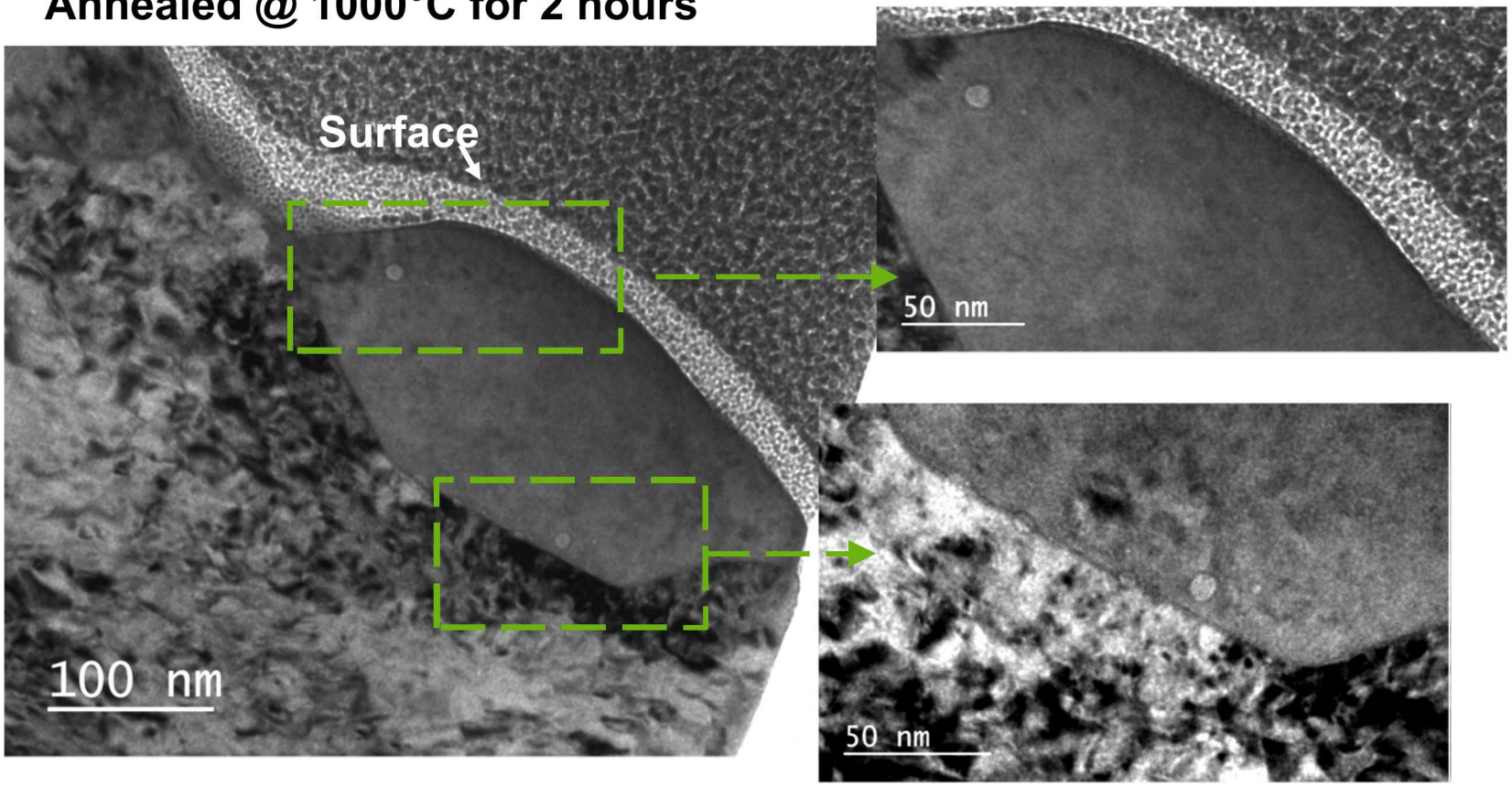
- 37.5% and 60% of the H located in the He bubble layer is located near a helium cluster/bubble for the Tersoff and EAM potential respectively (using a search algorithm & interaction cutoff distance of 0.32 nm)
- Remainder of H is mostly atomic with a few H_2 molecules
- Binding energy tends to be slightly lower with EAM

Planned Experimental Work

- Perform experiments to validate observations from modeling work, namely if helium bubbles will trap hydrogen in tungsten
- Initially perform helium implantations to get the desired microstructure and bubble density
- Subsequently expose the helium implantated tungsten samples to hydrogen using a hydrogen permeation setup being built at ORNL
- Bulk helium implantation and subsequent annealing has been performed at SNL and TEM is currently being utilized to observe bubble distribution.
- In-situ helium implantation and subsequent in-situ annealing was done at SNL to observe bubble growth at different temperatures

Cross-section TEM of the initial bulk helium implanted samples resulted in unexpected results

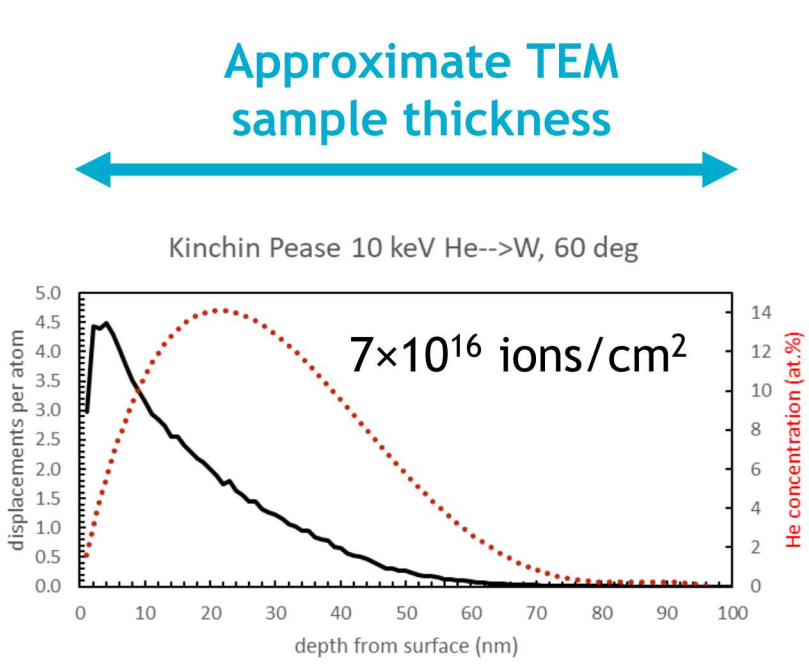
700keV He, $5x10^{16}/cm^2$, RT implantation
Annealed @ 1000°C for 2 hours

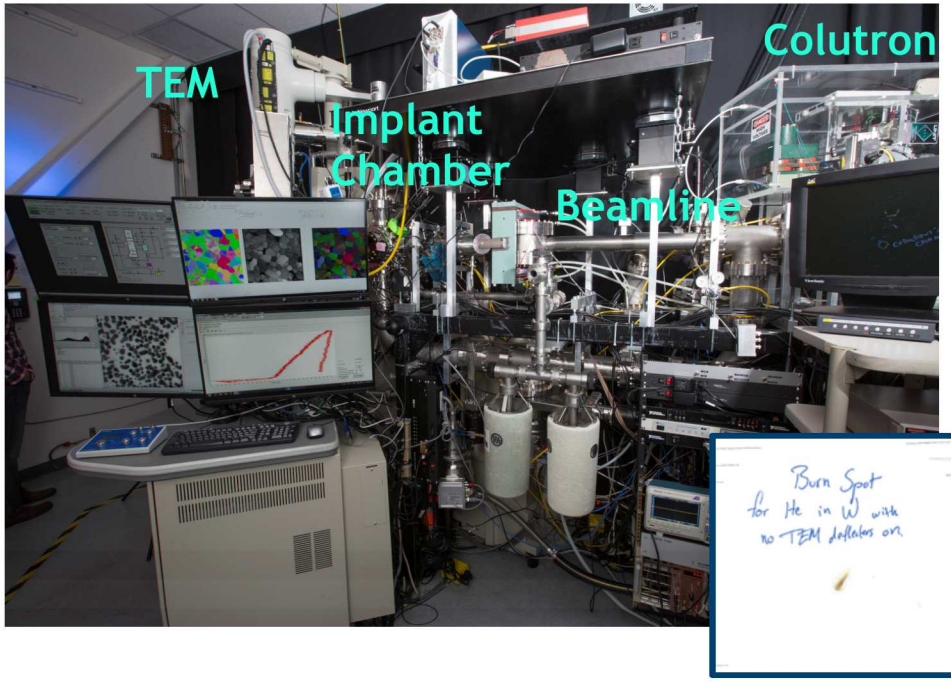


- According to the SRIM simulation, He bubbles should be observed at a depth of 300 nm - 1.4 μm. They were instead only observed at the surface.
- Samples were annealed after implantation, but bubbles were expected to grow in the same region as the initial implantation.

In-situ helium implantation and annealing were used to observe bubble growth and migration kinetics

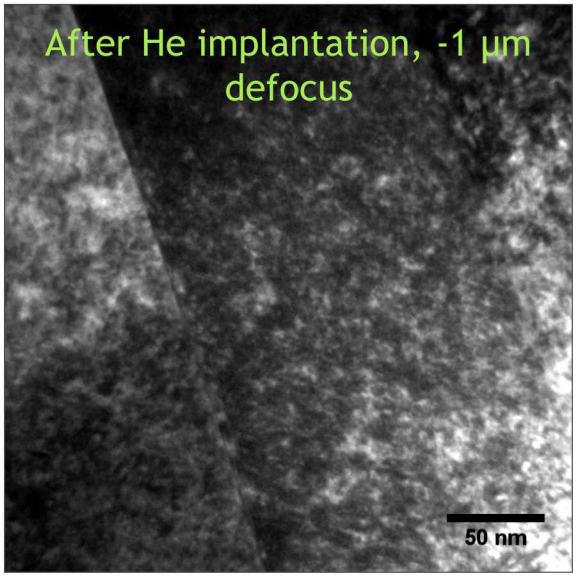
- FIB lift-out of W plate was used for TEM experiments
- He ion flux ranged from 1.29×10^{13}-7.81×10^{13} ions/cm^2/s
- JEOL single tilt stage was used for He implantation and Gatan double tilt heating stage was used for annealing
- During in-situ annealing, due to the high melting temperature of W (3,422°C), the temperature was ramped in 100°C increments. The temperature was held for 5 min at each step until 900°C, when bubble growth was first observed.

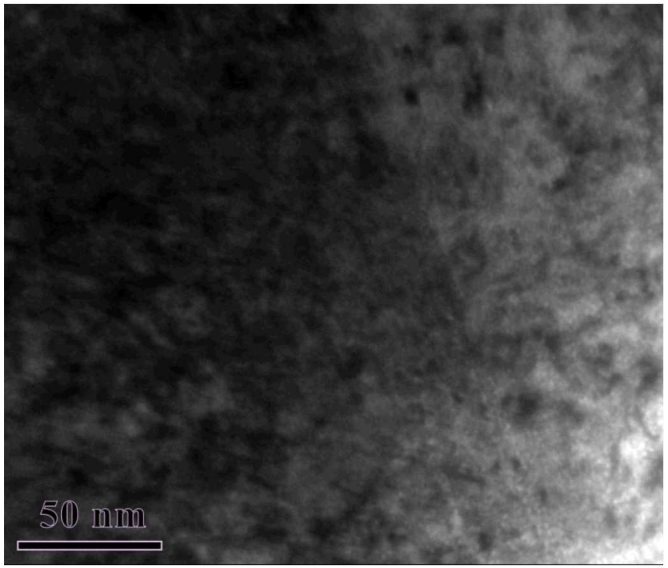




Bubbles were not observed until annealing at 900°C (T_h = 0.32)

- Due to significant FIB & He implantation damage, small bubbles could not be observed during in-situ He implantation
- Small bubbles were eventually observed during annealing at a temperature of 900°C.
  - At this temperature, the FIB damage started to anneal away.
  - Bubbles began to grow, making them easier to observe.
- In some cases, cavities appear to be diffusing through the W and coalescing.
  - Surprising observation at this low of a homologous temperature, but maybe explains bulk results





Conclusions

Subsurface Mixed H-He Bubbles

- Bubble (growth/loop punching) evolution is similar to previous MD results and depends on helium concentration and temperature
- For all cases, the hydrogen diffuses to the bubble periphery, with about 75-95% of the hydrogen located here after 100 ps
- A significant amount of hydrogen diffuses to the bubble periphery, ~25-40%, even when the hydrogen is initially distributed throughout the simulation cell
- The hydrogen at the bubble periphery may be strongly bound, with a binding energy of ~2-2.5 eV
- These results indicate that helium bubbles may be a significant trapping site for hydrogen, and therefore tritium – although validity of interatomic potentials and short-time MD simulations necessitate further study

Large-scale W-H-He

- When helium and helium bubbles are present, the H depth distribution is modified such that the hydrogen is located within the subsurface helium bubble layer as opposed to directly at the surface
- Helium appears to limit deeper hydrogen diffusion into the material
- For the hydrogen that is in the bubble layer, ~66% is clustered with helium which is consistent with the results from the mixed bubble simulations
- Longer simulation times are needed to further assess these results

Experimental Results

- Experiments planned to investigate H-He interactions in tungsten
- Initial He implantation in tungsten performed and bubbles were observed using TEM after annealing to 900 K
- Subsequent exposure to hydrogen will be performed

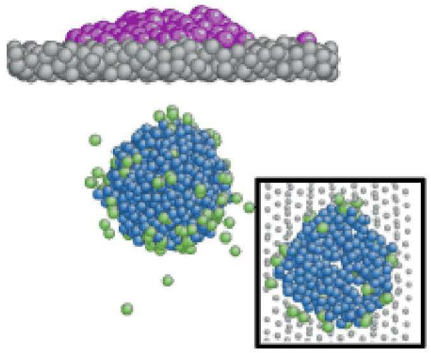
Backup Slides

Sensitivity of Simulation Parameters

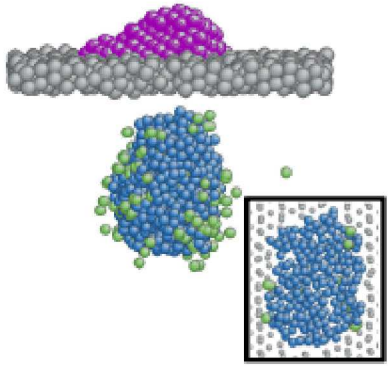
- Extended time
  - Select simulations were run up to 1 ns
  - Some hydrogen starts to diffuse further from the bubble but the majority still remains near the bubble periphery
- Double the tungsten width from 6 nm to 12 nm
  - Changes the areal density of bubbles due to periodic boundary conditions
  - Affects the initial bubble expansion
  - Leads to more destructive surface deformation especially for the (110) surface and higher gas atom concentrations
- Double the depth of the box from 6 nm to 12 nm and the bubble depth from 3 nm to 6 nm
  - Virtually no surface deformation
  - Bubble expands more laterally
  - Does not affect hydrogen partitioning to the bubble periphery

(111) Surface at 1800 K and 3 He/V and 1 H/V

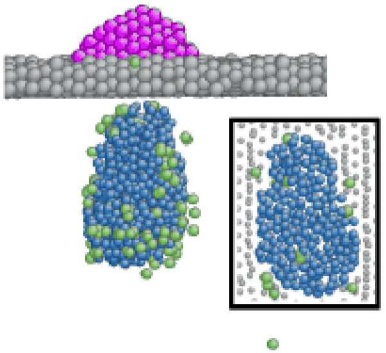
6 nm x 6 nm x 6 nm
100 ps



6 nm x 6 nm x 6 nm
1 ns



12 nm x 12 nm x 6 nm
1 ns



6 nm x 6 nm x 12 nm
1 ns