# Plasma-Surface Interactions (PSI): Atomistic Insights into He Transport and Agglomeration in Plasma-Exposed Tungsten


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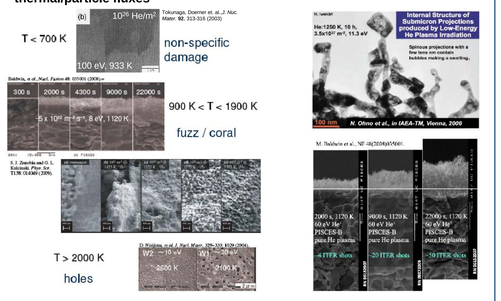
Project web site: https://collab.mcs.anl.gov/display/PSIscidac/

## Introduction

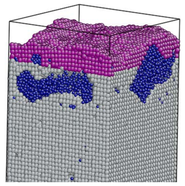
- **Experimentally observed tungsten (W) surface dynamics under combined thermal/particle fluxes**





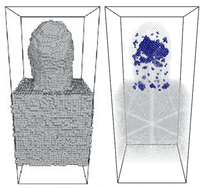



- **Small-scale MD simulations reveal continual bubble formation, bursting, and W surface modification**



Initially perfect crystal (atomistically smooth surface)

- Observed near-surface structure: He cluster/bubble distribution (blue spheres), W adatoms (purple spheres), and sub-surface W atoms (grey spheres).
- Configuration shown after ~2x10$^{20}$ He/m$^2$, but with a very high implantation rate of 10$^{27}$ He/(m$^2$s), i.e., about 5 orders of magnitude too large!
- Repeated formation and subsequent bursting/rupture of over-pressurized gas bubbles observed.
- How does surface morphology and implantation rate impact bubble formation?



Initial tendril – cylindrical morphology (atomistically smooth surface)

### Solid Surface Modeling Roadmap

- Low-temperature (< ~1000 K) regime of low energy (~100 eV).
- He (later mixed He-H) plasma exposure to tungsten: focus on bubble formation, growth, and over-pressurization leading to tungsten surface morphology changes.

**Key Physics Questions:**

- Rate effects (explored by AMD, MD and KMC simulations) versus continuum reaction-diffusion model predictions and experimental measurements
- Validity of dilute-limit approximation in concentrated He bubble populations
- Introduction of drift (driven diffusion) into transport formalism due to interaction of clusters with sinks such as surface and grain boundaries
- Multiscale integration

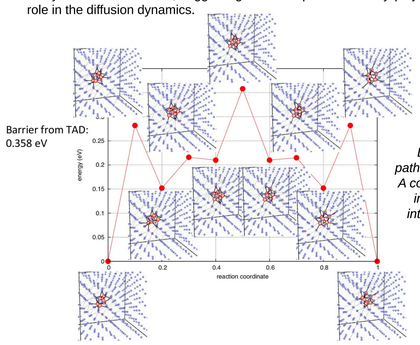



## Bulk diffusion of Small He Clusters

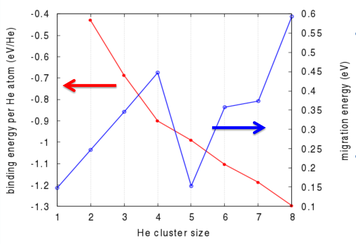
- To parameterize macroscopic reaction-diffusion models, we rely on atomistic simulations to identify unit transport/reaction mechanisms.
- Using a combination of MD and Accelerated MD (AMD), we have investigated the bulk diffusion of small He clusters, with sizes ranging between 1 and 8.
  - With AMD: Find and analyze the dominant diffusion pathways at low temperatures
  - With MD: Compute the diffusivity of the clusters between 250 and 1400 K. Assess whether the low-T kinetics hold at operation temperatures

### Diffusion Pathways and Energetics

- Pathways become increasingly complex with cluster size. They typically contain many intermediate states, suggesting that entropic effects may play an important role in the diffusion dynamics.



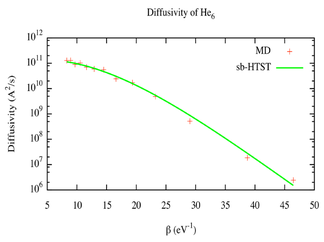

*Diffusion pathway for $He_6$. A complete hop involves 4 intermediate states.*




- The cluster migration energy is highly non-monotonic vs. cluster size: larger barriers at sizes 4 and 6, but a small barrier at size 5.
- In contrast, the increase in binding energy is monotonic.

### Kinetics

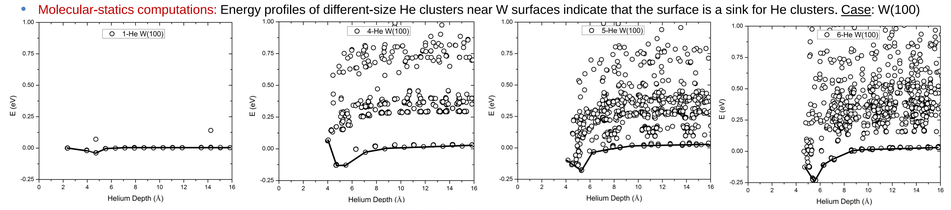
- At higher temperatures, relevant for reactor conditions, the entropic effects of the multiple minima exert themselves.
- For all cluster sizes, the cluster mobility can be accounted for by the pathway identified with AMD, over the entire temperature range.
- The non-Arrhenius temperature dependence can be captured by a generalized transition-state theory that accounts for intermediate states (green curve on the right).



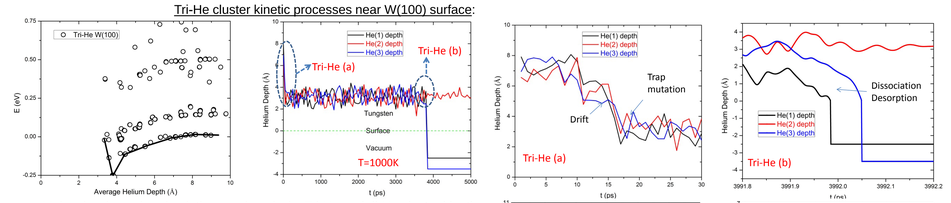

- Larger clusters (≥7) quickly experience *trap mutation*, i.e., the formation of a bubble nucleus by the creation of a W vacancy-interstitial (Frenkel) pair. Once bound to a vacancy, He clusters are essentially immobile.
- Our combined MD/AMD approach provides a comprehensive picture of the unit mechanisms of the diffusion of small He clusters in bulk W:
  - Relevant diffusion pathways have been identified
  - Diffusivity has been characterized over a wide range of temperatures

## Dynamics of Mobile He Clusters near W Surfaces: Drift due to Surface Segregation and Near-surface Kinetic Processes

- Motivation for this study: The diffusional transport of mobile He clusters mediates the evolution of surface morphology and the structural evolution of the near-surface regions of the plasma-exposed material.
- Molecular-statics computations: Energy profiles of different-size He clusters near W surfaces indicate that the surface is a sink for He clusters. Case: W(100)

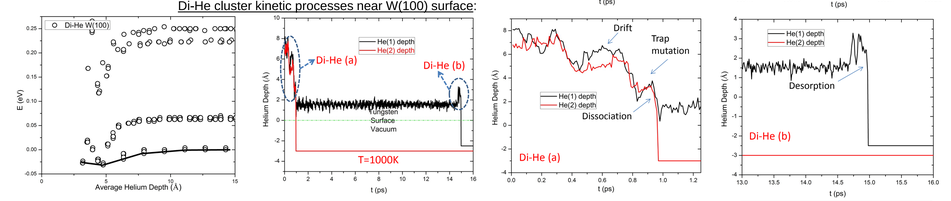



Tri-He cluster kinetic processes near W(100) surface:




- Molecular-dynamics simulations and analysis: Tri-He and di-He cluster kinetic processes shown near W(100) surface
- Mobile He clusters migrate to the near-surface region under the action of the thermodynamic driving force (drift) for surface segregation
- Multiple kinetic processes occur at the surface/near-surface region, including **trap mutation**, **cluster dissociation**, and **He desorption** from the surface
- Identification and characterization of kinetic processes for parameterization of coarse-grained cluster dynamics models

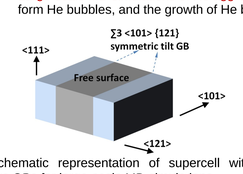
Di-He cluster kinetic processes near W(100) surface:



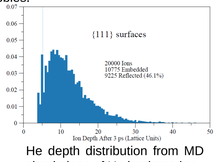

- This analysis sets the stage for bridging atomistic and continuum modeling and simulation toward a rigorous and quantitative description of drift and diffusional transport of such He clusters in near-surface W regions
- Further analysis provides the required closure relations for the drift and diffusive fluxes and for the cluster reaction rates in continuum cluster mass transport (drift-diffusion-reaction) models through predictions of: diffusion coefficients, segregation potentials as a function of distance from the surface, and reaction rate constants for the surface/near-surface kinetic processes.

### He Agglomeration in Near-Surface Regions of Polycrystalline Tungsten: Surface and Grain-Boundary (GB) Segregation

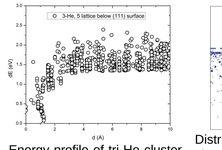
- Large-scale MD simulations of He aggregation in a model of polycrystalline tungsten reveal transport of He atoms (drift-diffusion), their aggregation to form He bubbles, and the growth of He bubbles.



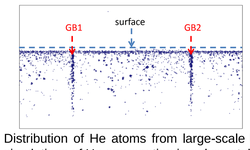

Schematic representation of supercell with two GBs for large-scale MD simulations




He depth distribution from MD simulations of He implantation




Energy profile of tri-He cluster near GB




Distribution of He atoms from large-scale MD simulations of He aggregation in polycrystalline tungsten. (W atoms not shown)

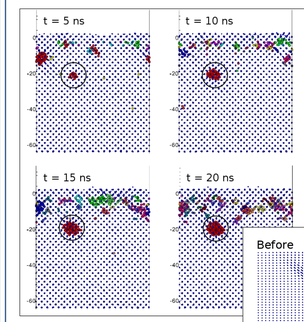
*Setup of the large-scale MD simulations*:

- GB type: ∑3 <101> {121} symmetric tilt GB with a W{111} free surface
- Simulation cell size: 50 nm x 50 nm x 18 nm
- Number of W atoms in simulation cell: 2.4 million
- Distance between GBs: 25 nm
- Rate of He atom insertion: 1 every 10 ps
- Implantation depth distributions for the insertion of He atoms into the W matrix derived based on MD simulations of He-atom impingement onto W surfaces

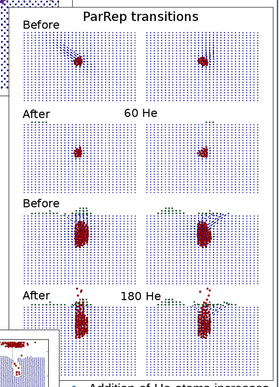
- MD results show that GBs and W surface serve as sinks for He atoms
- Calculations of He small-cluster energetics as a function of cluster distance from grain boundaries in W and from the W surface aid in providing thermodynamic and kinetic interpretations of the He bubble formation processes observed in the MD simulations

## Helium Bubble Growth

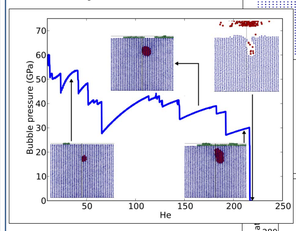
- In addition to the migration behavior of small He clusters, the behavior of large He bubbles is critical for parameterizing reaction-diffusion models and understanding the morphological evolution of the surface.



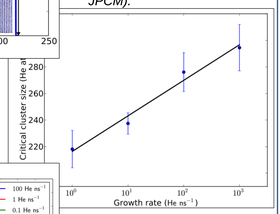

- As shown in the MD simulation on the left, once clusters undergo trap mutation, they act as bubble nuclei. Subsequently, these bubbles grow by absorbing mobile He clusters.
- To understand the effect of the He absorption rate on the bubble evolution, we perform AMD simulations of the growth of single bubbles at carefully controlled rates.

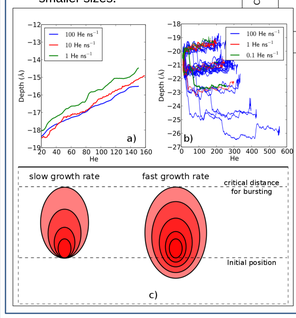



- We use parallel replica dynamics (ParRep). ParRep allows for the temporal parallelization of the discrete state-to-state dynamics and, therefore, greatly increases the time scale amenable to direct simulation.
- For example, by using 12000 cores, we were able to reach simulation times of 7.25 μs, which allows the simulation of realistic growth rates.




- Addition of He atoms increases the pressure in the bubble, causing the nucleation of <111> interstitials loops. The process continues until the bubble bursts (F. Sefta, N. Juslin, and B. D. Wirth, submitted to *JPCM)*.
- Our results indicate that the average bubble size right before bursting is very sensitive to the growth rate: decreasing the growth rate leads to bursting at smaller sizes.




- To explain this behavior, we tracked (a) the motion of the bubble's center of mass and (b) the maximum depth reached by He atoms as a function of the number of He atoms.
- Slow growing bubbles grow preferentially towards the surface, while faster growing bubbles grow more isotropically (c).
- In the latter case, bursting is delayed, as more He atoms are required for the bubbles to reach the surface and burst.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