Overview of SciDAC – PSI: A multiscale – multiphysics approach to simulating tungsten plasma surface interactions from the boundary plasma to the bulk substrate

This work was supported by the U.S. Department of Energy, Office of Fusion Energy Sciences and and Advanced Scientific Computing Research (ASCR) through the SciDAC-3 program.

Rick Kurtz, on behalf of:

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

Presented at the IAEA CRP meeting on Plasma Material Interactions
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This work was supported by the U.S. Department of Energy, Office of Fusion Energy Sciences and and Advanced Scientific Computing Research (ASCR) through the SciDAC-3 program.
Application of leadership class computing and computational materials science are key tools to accelerate fusion materials development. However, as governing phenomenon span decades in length and time scale; this challenge involves numerous grand challenges.
Objective is to develop PSI simulation capability across three coupled spatial regions:

- Edge/scrape-off-layer region of the plasma, with sheath effects
- Near surface material response to plasma exhaust, with neutron damage and influenced/coupled to plasma sheath
- Structural materials response to intense, 14 MeV-peaked neutron spectrum

Material surface - erosion (impurity & dust), T retention, surface evolution

Material bulk – neutron damage & transmutation

SIA clusters form in subcascades interconnect region
Goal: Discovery science to identify mechanisms of W nano-scale fuzz formation and synergies between He & H exposure that impact D/T permeation & retention – and surface mass loss (dust)

Mechanisms of interest: sputtering, surface adatom formation, diffusion, He bubble formation, expansion & rupture

Focus on MD & kinetic modeling approaches, leading to a large-scale continuum-level reaction-diffusion code for plasma materials interactions

Biggest long-term scientific challenge is understanding the kinetics of coupled defect–impurity evolution with a disparate range of kinetic rates --- this requires algorithmic improvements on both the physics and computing side

Key MD observations of early stage He bubble evolution

- Helium insoluble but highly mobile and can self-trap (at high implantation rates) due to strong He-W repulsion to form highly mobile, strongly bound helium clusters – *implantation rate effects are very important*

- Significant surface evolution through tungsten adatom formation, driven by trap mutation and loop-punching as tungsten interstitials rapidly diffuse to surface

- As bubbles continue to grow at very high pressure, eventually rupture
Thermodynamics & kinetics of small He clusters*

- Atomistic simulations (AMD, MD, statics) used to identify unit transport/reaction mechanisms
  - Challenges relate to multitude of pathways with increasing cluster size

4-He cluster migration:

Thermodynamics (binding) & kinetics (migration):

Larger clusters undergo ‘trap’ mutation which decreases mobility

Defect evolution by trap mutation reaction:

Monotonic increase in binding energy, Complex, size-dependent kinetics

First simulation of He bubble growth at He-irradiation flux appropriate for fusion first-wall in ITER. The simulations find a qualitatively different growth mode when rates approach experimental values. They reveal rate effects on bubble size, shape, pressure, and surface damage.

**Research**
Parallel Replica Dynamics simulations of bubble growth with He injection rate ranging from $10^{12}$ s$^{-1}$ to $2 \times 10^6$ s$^{-1}$. EFFICIENT TO PETASCALE: UTILIZED 160,000 CORES (OVER HALF OF TITAN) AT ORNL AT 77% EFFICIENCY.

Slower growth leads to smaller, more anisotropic bubble that grows in a directed way towards surface, producing fewer adatoms during growth and creating less surface damage upon bursting.

Collaboration with BES program
Accelerated Molecular Dynamics (Voter) at LANL.

Our approach to large-scale MD simulations

- Develop a database of implanted He range

- Bombard a “clean” surface with 100 eV helium 20,000 times

- Calculate, for each crystallographic surface,
  - Fraction of ions that reflected
  - Fraction that embedded but escaped in < 5 ps
  - Depth distributions

- Advantages
  - Constant (large) time step
  - Don’t simulate ions scattering off the surface
  - > 90% reduction in simulation time

Caveat: ignores electronic stopping


- Periodic conditions in the x, y directions and Free Surface in z (typical simulations are 50x50 nm cross section and 25-50 nm deep (O(20 Million atoms))

- Every 10 ps a He atom is added according to the He depth distribution from MD (ignores electronic stopping) and reflection, but effective implantation flux of \(\sim 4 \times 10^{25} \text{ m}^{-2}\text{s}^{-1}\)
Tungsten surface response to low-energy He exposure

(110) surface, 1.25 $\mu$s

$\Gamma = 5.3 \times 10^{26}$ m$^{-2}$ s$^{-1}$; $\Phi \in [0, 6.8 \times 10^{20}$ m$^{-2}$]
Large-scale MD results (100 eV He -> W)

- Raised portions (red/yellow/white) are “pushed up” by helium bubbles
- Bubbles typically **vent non-destructively** and may “heal”
- Still too low a fluence to see significant bursting events or tendrils larger than $\approx 2$ nm

(111) surface, $1.5 \ \mu$s, $\Gamma = 4 \times 10^{25} \ \text{m}^{-2}\text{s}^{-1}$ ($\Phi = 6 \times 10^{19} \ \text{m}^{-2}$)
Tungsten surface response to low-energy He exposure

• MD* of 100 eV He implanted into W reveals formation and growth of over-pressurized, sub-surface He bubbles thru self-trapping, trap mutation, loop punching and bubble bursting that evolve tungsten surface (hillocks & craters)
  → Qualitatively consistent with experiments** of W surface evolution following 60 eV He on tungsten
  → Quantitative comparison requires evaluation of rate & scale effects ($\Gamma$: MD $10^{26}$ vs expt $10^{19}$; $\Phi$: $10^{20}$ vs $10^{24}$)

* Hammond & Wirth, UTK/ORNL

** Donovan, Buchenauer, Kolasinski et al., SNL
Xolotl (continuum) calculations to reach experimental scale

- Xolotl (SHO-lottle) is the Aztec god of lightning and death & is our continuum cluster dynamics code for modeling plasma surface interactions, focused on sub-surface bubble dynamics & surface evolution (to date), will include erosion in near future
- Developed from ‘scratch’ for the SciDAC project, designed for HPC (current and emerging architectures – multicore, multicore+accelerator)

**Atomistic picture**
- H/He
- sputtering
- saturated layer
- co-deposition
- erosion
- vacancy
- trapped H/He
- interstitials

**Continuum picture**
- Surface
- \( C(\text{He}_{16} V_{4}) \)
- \( C(V_{1}) \)
- \( C(\text{He}_{1} V_{1}) \)
- \( C(l_{1}) \)

**MD simulations of He retention**
- He flux = \( 4 \times 10^{25} \text{ m}^{-2}\text{s}^{-1} \)

![Graph showing He retention over time and nominal fluence](image)
Xolotl (continuum) calculations to reach experimental scale

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Using MD to ‘train’ continuum scale (Xolotl)

**MD simulations** – $\Gamma \sim 5 \times 10^{27} \text{ m}^{-2}\text{s}^{-1}$

**Xolotl simulations**

![MD simulations graph](image1)

![Xolotl simulations graph](image2)
Using MD to ‘train’ continuum scale (Xolotl)

MD simulations – $\Gamma \sim 4E25 \text{ m}^2\text{s}^{-1}$

Xolotl simulations

Cumulative Fraction of Retained Helium vs. Depth (nm)

- (001) surface, $\Phi_{\text{nominal}} = 3.17 \times 10^{18} \text{ m}^{-2}$
- (011) surface, $\Phi_{\text{nominal}} = 2.97 \times 10^{18} \text{ m}^{-2}$
- (111) surface, $\Phi_{\text{nominal}} = 5.22 \times 10^{18} \text{ m}^{-2}$
- (211) surface, $\Phi_{\text{nominal}} = 4.21 \times 10^{18} \text{ m}^{-2}$

Cumulative Fraction of Helium vs. Depth (nm)

- (001), initial depth
- (011), initial depth
- (111), initial depth
- (001)
- (011)
- (111)
Xolotl now calculates experimental timescales

Flux = $4 \cdot 10^{22}$, W100.
MD database demonstrates drift and segregation of He

- Large-space-scale molecular-dynamics (MD) simulations ($T = 933$ K) of evolution of implanted He in model polycrystalline tungsten
- Simulations reveal helium aggregate formation and growth, as well as mobile He$_n$ cluster diffusion, drift, and segregation on W surfaces and GBs
- Example: He distribution near GB intersections with W\{110\} and W\{111\} surfaces. Helium distribution determined by sink segregation strength, as well as trap mutation reactions that are activated as the clusters approach the sinks

Significantly reduced He mobility on W grain boundaries

- Atomistic simulations across representative grain boundaries indicate strong trapping of He at grain boundaries, with significantly reduced He diffusivity along the grain boundary compared to the bulk.

Helium “gettered” by grain boundaries; fewer bubbles in the bulk, more on the grain boundary.

But, *He is not completely trapped by W grain boundaries*

- Grain boundaries "getter" helium
- We’ve done several **perpendicular** to the surface plane
- This one is **parallel**; helium isn’t stopped!
Additional atomistic/Accelerated MD in progress

Deep bubble growth

He bubble initially located in a spherical void of 277 vacancies. \(~10^5\) atoms at 1000 K.

As in the shallow bubble case, slower growth rates favor transitions with lower He content.

• At the over-driven rates simulated with MD, the tungsten matrix responds differently than at the slower rates representative of experiments.

Bubble growth near \(<111>\) screw dislocation

He bubble growth process strongly influenced by dislocations, which act as traps. For example, a He bubble nucleated in the at a screw dislocation (right) grows along the core and reaches the surface faster, as compared with the perfect crystal case (left).

Bubble-bubble coalescence

Simulations of bubbles growing in close proximity show a strong directionality of the growth process for the smaller bubble. The coalescence is characterized by the frequent nucleation and growth of connecting dislocations, eventually released from the bubbles as dislocation loops.

Sandoval, Uberuaga, Perez (LANL) unpublished results, in collaboration with BES funding (Voter)
Further Xolotl code development

• Verification of Xolotl 1D through cross code comparison against LAMMPS, Paraspace, and KSOME, as well as multiscale integration & benchmarking to large-scale MD

• Performance profiling performed against Paraspace by P. Roth (SUPER)

• Generalization of the system of equations in 2D and 3D, working closely with B. Smith and S. Aithal (FASTMath).

• Significant improvement of the memory usage and performance run-time through strong interactions with B. Smith (FASTMath):
  - 4th Order implicit Runge-Kutta ODE integrators with adaptive time steps allows much larger time steps while preserving accuracy
  - Composite pre-conditions for linear systems with direct (1d) or multigrid (2 or 3d) solves for the diffusion terms with point-block Gauss-Siedel for reaction solves appears to be optimal and scalable solver for larger problems
KMC simulation of He clustering below W surfaces

T=973K, Flux (Γ) of 100 eV He at 4E25 He m\(^{-2}\)s\(^{-1}\)

Kinetic Monte Carlo (KMC) simulations incorporating atomistic gas diffusion, clustering mechanisms used to extrapolate from ultra-fast MD implantation fluxes to experimentally relevant rates but limited to relatively short times O(seconds).

Indicate mechanism boundary of gas bubble nucleation mechanism \(f(Γ, T)\)

He-H defect interactions in W

- Interatomic potential(s) derived to describe W-He* and W-He-H** interactions

Ab-initio data of H binding to He-H-V in W

Validated potentials used to evaluate H partitioning to sub-surface He bubbles
- He is uniform, but H partitions to the bubble surface
- evaluating H storage capacity as function of bubble size & He pressure

Atomistic result from potentials – Validating comparison

**H – W potential validation & modification**

- Benchmarking H adsorption energies on W surfaces

\[ E_{ads} = 2.4\,eV\ (B), 2.3\,eV\ (T), 2.1\,eV\ (O), 2\,eV\ (D) \]

- But MD doesn’t indicate H\(_2\) formation and desorption at 2500 K, so 3-body W-H term modified (now called modified Juslin W-H potential), resulting in H\(_2\) desorption

*J. Guterl et al., J. Nucl. Mater. 463 (2015) 263-267*
Atomistic modeling of He-H synergies

- MD simulations performed for 2 nm diameter bubble containing high pressure He (3 He/vac) and random distribution of H (0.5 H/vac) at 1800K

- H is observed to rapidly migrate to bubble periphery and remains ‘trapped’ at the bubble interface

- Raises question about potential for tritium trapping/inventory
  - artifact of interatomic potential
  - short time MD simulations

Experimental evidence of D/T trapping/retention*

Ogorodnikova et al., JAP 109 (2011) 013309.

“D not retained in He/D exposed plasma (NRA measurements) following 1000° C”

Doerner et al., ICFRM19 & PSI2016, personal communication
**Binding energy of H to the He bubbles**

- Simulations quenched to 0 K then hydrogen atoms moved along the [100] direction towards and away from the bubble in 0.1 nm steps with energy minimization.
- Plots are normalized such that the energy of hydrogen far from the bubble is 0 on the y axis and the edge of the bubble is 0 on the x axis.
- A well of approximately 1.5-2.5 eV that is roughly 1-2 lattice units thick exists near the bubble periphery while there is a high potential energy within the bubble.
- 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.

H partitioning to He bubbles

- H/He implantation below W surface at 1200 K ($\Gamma \sim 4 \times 10^{27} \text{ m}^{-2} \text{s}^{-1}$) --- dramatically different H behavior and He/H clustering depending on W-H potential (900H/100He implantation shown below)

Juslin BOP (~85% H & He retained)

Li BOP (~50% H & He retained)

atomic H at surface hollow sites

Green = H
Blue = He
Purple = W adatoms

H partitioning to He bubbles

- Introduced H into pre-He implanted Tungsten (MD simulations)
**H binding to He clusters (observed clusters from MD)**

Snapshots of H – He cluster interactions in MD simulations with pre-implanted He (green is H, blue is He)

- 60% of H is located in near-surface helium cluster saturated layer
- 37.5% of this H is located near a helium cluster/bubble (using a search algorithm & interaction cutoff distance of 0.32 nm)
- Remainder of H is mostly atomic with a few H\(_2\) molecules

<table>
<thead>
<tr>
<th>Cluster Size</th>
<th>Binding Energy (eV)</th>
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<tr>
<td>He(_{11})H(_1)V(_2)</td>
<td>2.41</td>
</tr>
<tr>
<td>He(_{11})H(_3)V(_3)</td>
<td>0.05</td>
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<tr>
<td>He(_3)H(_1)V(_1)</td>
<td>1.63</td>
</tr>
<tr>
<td>He(_8)H(_1)V(_3)</td>
<td>2.50</td>
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</table>
• Spectrum of W PKAs due to 14-MeV neutrons shows a significant number of PKAs up to 280 keV of recoil energy or 196 keV of damage energy ($E_{MD}$)

• Previously, primary defect damage database includes $E_{MD}$ up to 100 keV

• New displacement damage data generated at 150 and 200 keV for 300, 1025, and 2050 K

• Data at 150 and 200 keV follow the trend of defect production curve ($N_F$) for $E_{MD} > 30$ keV

• KMC simulations of irradiation damage accumulation due to 14 MeV neutrons are currently underway

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KSOME: Kinetic Simulations of Microstructural Evolution

- KSOME is an object kinetic Monte Carlo code to simulate the evolution in time and space the distribution of lattice defects in crystalline materials accumulated during irradiation.
- KSOME is designed to simulate the diffusion, emission, transformation and reaction events of vacancies, interstitials, impurities and their complexes, including any number of combinations of point defects, for bcc and fcc lattices at a given temperature.
- Cascade insertion is random, based on a specified cascade production rate and additionally allows for creation of any type of defect based on their production probability.
- Interaction of mobile defects (simple absorption) with sinks such as dislocations, grain boundaries and free surfaces.
- Defect parameters like type, size, orientation, etc. are used to identify defect diffusion, emission, reaction and transformation events between various types of defects.
- Example events:
  - Rotation or change of direction of 1D-diffusing SIA clusters
  - Vacancy loop transforming into a spherical cluster or void
  - Emission can also be associated with a loop punching or trap mutation event
  - Allows emission of multiple (size or type) defects from a single source
- Simulation of radiation damage according to HFIR PKA spectrum and migration parameters.
Dose Dependence of Vacancy Cluster Densities and Sizes

- With increasing dose rate:
  - Number density of vacancies increases
  - Vacancy cluster density decreases
  - Average vacancy cluster size decreases

- Fraction of visible clusters:
  - $10^{-8}$ dpa/s - saturates at 95% of the vacancy population
  - $10^{-4}$ dpa/s – reaches 55% of the vacancy population at 1 dpa
  - Visible clusters - 2 nm diameter or about 300 vacancies

- Vacancy cluster sizes at $10^{-8}$ dpa/s:
  - Grow larger than at higher dose rates due to the greater time between cascade insertions permitting more defect diffusion
  - Di-vacancies are not stable, which suppresses nucleation of new clusters

- No formation of SIA clusters
  - SIAs quickly diffuse to grain boundaries
  - SIAs are more likely to recombine with the increasing population of vacancy clusters
Summary & Future Work

- Fuzz formation mechanism remains to be conclusively determined; but significant hints that helium gas bubble formation, agglomeration/coalescence and bursting phenomena play a key role in driving initial W surface roughening.
  - MD reveals initial surface roughening by adatom formation (trap mutation) followed by adatom island (bubble expansion by loop punching) and subsequent bubble bursting & indicates H/D/T trapping at He bubbles.

- Kinetic models (Xolotl-PSI) now benchmarked to predict He bubble R, N & P as a function of He exposure conditions & models for W defect/loop/surface adatom diffusion to model both bubble formation, evolution & topology changes – framework for coupling to plasma edge is emerging.

- Key uncertainties: He implantation rate effects; influence of temperature/stress gradients (TRANSIENTS).
  - What mechanisms transform W surface instability into tendril filaments of 10-100 nm diameter?
  - T storage & retention in He bubbles & W-He-H(+C, Be, impurity) materials.