The atomistic phenomena at the plasma-surface interfaces

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Layout:

• General remarks on PMI
• How about tungsten 😊?
• Concluding notes
How does PMI see a flux of $10^{25}$ particles/m$^2$s mean (ITER)?

The flux is 0.01 particle/nm$^2$ns, i.e. 1 particle each 10 ns at 10 nm$^2$

A typical evolution of deuterium impact at 100 eV even with chemical sputtering in carbon takes no more than 50 ps, and penetration no more than 2 nm; in tungsten events even faster

Each particle will functionalize the material, change the surface for the subsequent impact!

Processes essentially discrete  Atomistic Happening at nanoscale in both time and space, scales determined by impact plasma particle energy

The traditional trial-and-error approach to PMI for future fusion devices by successively refitting the walls of toroidal plasma devices with different materials and component designs is becoming prohibitively slow and costly

Need bottom-up approach arising from the fundamental atomistic and nano science
Materials exposed to plasma are modified, resulting in a “dynamical” surface.

Study the dynamically prepared surface.
Challenges at the Plasma-Material INTERFACE

This is not the material science!
Science of the interface has many fundamental processes & synergies

- Elastic reflection
- Implantation
- Re-emission & sputtering & chemistry
- Trapping/detrapping retention
- Diffusion, permeation
- First few nanometers play decisive role

Drivers:
- Multi-T, -n, -species, plasma irradiation, neutrons
- Sheath acceleration

Give rise to synergistic effects

Damage Effects:
- Vacancies, bubbles, blisters, dislocations, voids, neutrons?
- Re-deposition
- Co-deposition
- Erosion
- Ablation
- Melting (metals)
Plasma induced synergy in the evolution of an irradiated surface

He suppresses H retention in W
- He penetrates deeper than H
- Strong dependence on energy
- He bubbles: barrier to H diffusion?

How to do it? At initial conditions!

T=200,000 K
Maxwell-Boltzmann

~sqrt(E) exp(-E/kT)

T=200,000 K
All VZ>0 inverted sign
Classical MD is only as good as the interatomic potential model used:

**Significant element of the epi**

**Most advanced: hydro-carbon potential developed for chemistry**
- Brenner, 1990, 2002: REBO, short range, 0.2 nm
- more sophisticated AIREBO (Stuart, 2000, 2004, 1.1 nm)
- > 400 semi-empirical parameters, “bond order”, chemistry

Adaptive Intermolecular Reactive Bond Order (AIREBO) potential: torsion, dispersion, Van der Waals,

**EX: MD calc. of reflection coeff.**
- Significant sensitivity to changes in potential model for some processes
- Experimental validation essential to establish credible MD simulation.
- Interatomic potentials for W and Be are less mature than for carbon and require more experimental validation.

Improvements to CH potentials done (Kent et al, 2010)
New Li-C-H-O potentials being developed (Dadras et al, 2010)

Even for hydrocarbons problems visible

Remarkable agreement of theory & beam exp’t when simulation prepare the sample at the level of nanoscale (fluence) to mimic exp’t. No fitting parameters!

Chemical sputtering of carbon


Carbon

Lithium

From in-situ experiments labs, and number of tokamak machines with graphite: nm’s thin lithium coatings have a "significant" effect on plasma behavior, in particular on reducing hydrogen recycling, improved energy confinement time, a reduction of ELMs:

If there is a SIGNIFICANT amount of oxygen on surface with lithium present in the graphite matrix, OXYGEN becomes the main player in retention-erosion chemistry; NOT LITHIUM!!!

Experiments validated(Taylor, 2013) presence of O

Main challenges and opportunities in the new phenomenologies lay in the lithium based divertor!!!
Electronegativity is a chemical property of an element defining its tendency to attract electrons: Li has it exceptionally low in comparison to H, C, O, Mo, W.

Consequence: Bonding between Li and other atoms is covalent and polar; Long-range nonbonding: Coulomb: $1/R$, Lennard-Jones: $1/R^6$, $1/R^{12}$

Lithium wall conditioning improves confinement! How? Answer is in quantum mechanics i.e. in change of multibody electron cloud with change of coordinates. Classical mechanics cannot treat this by definition!

Quantum-Classical MD based on Self-Consistent-Charge Density-Functional Tight-Binding (SCC-DFTB) method (developed by Bremen Center for Computational Mat. Science, Germany) a possible answer for qualitative phenomenology is our choice. Collaboration with Stephan Irle & K. Morokuma.
Can we afford quantum mechanics for the PMI very-many body problems?

Answer depends on the question!

If the question is chemistry: YES

Also experiments check: No problem with impact energy!!!

Chemistry evolves at thermal energy anyway!!!
HOW ABOUT TUNGSTEN?

- Why tungsten?
- Response to the self-atom ("neutron") damage
- Response to D impacts, retention
- Nano-grain structure or...

- The dynamics of the system with D impact determined by the details of the potential, W-W and W-H.
- We use in what follows the BOP potential of Juslin et al (2005)
- A few thousands cores at TITAN, impact surfaces up to 300 nm²

More details on the potentials in Harrison’s talk.
Challenges with tungsten in fusion

Neutron inflicted defects

- 14 MeV Neutrons? Energetic particles?

High Temperatures!

For realistic energy conversion (DEMO)
need a hot surfaces > 600C

Carnot thermodynamical process
of high efficiency is needed

On the other hand:
ITER will work at low T’s (400K)
Most of the experiments done at room temperature (300K)
Neutron-caused defects simulated by effects of W self-atom (ion in exp.) damage

One way to study impact of 14 MeV neutrons and other light energetic particles

\[
\frac{4M_1M_2}{(M_1 + M_2)^2} E_{k0}
\]

• Virgin W has very low density of intrinsic defect sites at which to trap He
• 30 keV W ion exposure creates extrinsic near-surface defects that should facilitate He trapping during subsequent He ion exposures, perhaps even amorphize surface, and/or accelerate fuzz growth, but effect is small

Exp::

Why so small effects?

Strong recombination of Frenkel pairs

Evolution of Defects in a Tungsten Surface by Cumulative Bombardment with Self-Atoms: Classical MD is here a good tool !!! We choose LAMMPS and BOP
Molecular dynamics results of damage accumulation for consecutive W impacts:
• Unlike TRIM, accumulated damage is not linear with dose (# of impacts)
• In fact, accumulated damage saturates at much smaller doses than applied in expt.
• Evidence for spontaneous recombination of Frenkel pairs inside critical distance

Recent experiments on deuterium retention in pre-damaged W by self-ions show saturation about 1 dpa!!!
The calculations of self-damage in tungsten done in range 250 eV -10 keV (collaboration with Yong Wu, IAPCM, Fu Yong Zhao (2014))

- For impact energies < 2 keV implantation dominates over sputtering:
  - Interstitials dominates vacancies
- At higher energies the trend inverts
  - At large fluences:
    - The dominant defect linearly Increases with increase of the dominant one of the Implanted and sputtered;
    - Sub-dominant defects saturates into a constant
<table>
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<tr>
<th>E(keV)</th>
<th>Vacancies</th>
<th>Interstitials</th>
<th>Sputt</th>
<th>Impl</th>
<th>Impl2</th>
<th>Refl</th>
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<td>A</td>
<td>B</td>
<td>C</td>
<td>D</td>
<td>E</td>
<td>F</td>
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<td>0.25</td>
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<td>1.01±3.05</td>
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<td>10</td>
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<td>2.90±11.05</td>
<td>110.38±0.96</td>
<td>0</td>
<td>3.00±0.004</td>
</tr>
</tbody>
</table>

E≤1 keV,
Vacancies: A*k/(B+k)+C*k , Interstitials use the same A and B as in Vacancies, but notice C(negative) : A*k/(B+k)+C*k
Sputtering: D*k; Implantation: E*k; Impl2 (inty2): F*k; Refl: G*k

E>1 keV,
Interstitials: A*k/(B+k)+C*k , Vacancies use the same A and B as in Interstitials, but notice C(positive): A*k/(B+k)+C*k
Sputtering: D*k; Implantation: E*k; Impl2 (inty2): F*k; Refl: G*k

How to calculate fluence:
E≤1 keV, Fluence=k*1.733126x10^{12}
E=2 keV, E=4 keV: Fluence=k*6.23925x10^{11}
E=6 keV, E=8 keV, E=10 keV: Fluence=k*3.1832923x10^{11}
**Incidence angle dependence and dual exposure**

**He impact on tungsten**

- **The fuzz growth remembers the incidence angle**
  - So not totally random

- **Reduced tendril density after dual exposure**
  - Due to enhanced sputtering?

Mass loss would have significant implications for W-based magnetic fusion devices (e.g. during disruptions, when incidence angle and energy can change)

F.W. Meyer, 2013

Incidence angle exp studies only possible using ion beam approach (currently)!
Islands of deuterium retention known in various materials

Examples:

Protective layer of D formed in tungsten by impact of 25 eV D (Krstic 2014) has inherently the width

Seen in DFFER experiment recently at low 5 eV


“Island” forms at the end of the collision cascade, upon thermalization
The life of the layer will depend on the competition of the impact flux and outgoing diffusion (at longer time scale).

Can we control it?

Only layer of cumulated D’s shown in the figure

J. Wells
Simulations of Single Grain Boundaries In tungsten

Boundary orthogonal to a D impact

Boundary parallel to the D impact
Formation and variation of a layer with increase of fluence
Pay attention to the short, ns times, and huge impact fluxes

More fluence...leads to saturation, amorphization, swelling and de-gasing...

1000 D
2000 D
3000 D
1000 K
This dynamics at ns time scales depends strongly on temperature.
Different direction of the grain boundary leads to different D accumulation, still consistent with previous conclusions.

Note: Formation of D2 molecules evident.
Boundaries do accumulate D

- Cumulative # of retained D after 2000 impacts
- Cumulative # of retained D after 1000 impacts

- Cumulative # of retained D vs. midpoints of y bins in nm for TW=1000K and TW=300K
- Slope: -16% for TW=1000K
- Total slope: ~21% for TW=300K

- Cumulative # of D impacts vs. cumulative # of D impacts for TW=1000K and TW=300K
- Mid 1 nm range
Applications: Nanograineds tungsten or...

Polycrystalline metal

May be worth thinking on the multi-film tungsten as alternative technology
CONCLUDING REMARKS
What have we learned from studies of surfaces, i.e. interfaces of plasma and materials?

- PMI extremely difficult interfacial problem (Material mixing create SURFACE entity; its scale depends on impact energy: For sub-100eV => nm-ns scales)
- PMI science can be built from bottom-up recognizing its multiscale character and building from shortest time/spatial scales (fs/Angstrom) up
- Theory&modeling of PMI must be validated by experiment (and v.v.), the qualitative understanding on phenomenology rewarding
- Irradiation create dynamical surface, changing interface, cumulative bombardment is the key for agreement with experiment
- Surface responds to synergy in plasma irradiation (angles, energies, particles), NOT following linear superposition principle; Plasma irradiation modeling and experiments with beam experiments.
- Chemistry&dynamics of lithiated and oxygenated surfaces must be treated by QM -> QCMD
- Self-healing feature of tungsten defects upon cumulative bombardment of ions and “neutrons”; clustering; nanograining.
Looking forward

• The plasma-material interface has a big effect on the plasma performance, and we don't understand why! The answers can be found in the plasma-PMI integration science.

• The main weight in the science of integration of fusion plasma and its interfacial surface boundaries is carried by PMI because 1) the basic PMI phenomenology evolves much faster than the plasma time scale, and 2) it evolves through wider range of scales, which partially overlap with the scale of plasmas. The PMI has to be understood and parameterized at nanoscale before integrating it with plasma at the “same footing” at micro-scale.

• Bringing together the various scales of PMI and plasma is the fundamental multidisciplinary question, covering plasma science, surface science, atomic physics, computer science and applied mathematics.

• The team of physicists, computer scientists and mathematicians is needed to perform the multiscale integration task. Need to do from low Z to high Z, from liquid metals to polycrystals, chemical and physical processes. Computer resources, computer codes, knowledge “how-to” are available. Funding the PMI-plasma integration science would avoid trail-and-error loses and save millions of dollars.

• UQ and Quality validation of the simulations is the key for the “right track”. Mimicking the experiments by simulation is the key for the successful validation. High quality experiments, well suited for the purpose do exist.
Strategic objectives: Integrated plasma & material modeling system

Plasma codes resolve events at the scale of µs

At shorter than µs time: Study phenomenology, provide parameters for MC approaches at longer time scale!!!

Atomistic PMI codes (Computational chemistry NWChem, Approximate DFT: SCC-DFTB, Quantum-Classical Molecular Dynamics, Classical Molecular Dynamics, LAMMPS)

Mesoscopic PMI codes (DEM: LIGGGHTS and KMC:SPPARKS, referenced in the text, and Lattice-Boltzmann codes [PALABOS] and [SAILFISH]),

Plasma codes (XGC family and DEGAS2)

Study PMI separately, with plasma drivers

D. Gersappe

Integration of PMI and plasma at the “same footing”, with nano PMI drivers
Many thanks to PMI close collaborators:

Theory:

- Fred Meyer (PD, ORNL)
- Steve Stuart (Clemson U.)
- Paul Kent (ORNL)
- Alain Allouche (CNRS, Fr)
- K. Morokuma, Kyoto U.
- J. Jakowski, NICS
- S. Irle, Nagoya U

Experiment:

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Many thanks!