Mesoscale mean-field modeling of neutron irradiation damage accumulation in tungsten

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2014 Joint ICTP-IAEA Conference on Models and Data for Plasma-Material Interaction in Fusion Devices
Motivation

- W is a very attractive candidate as a first wall/divertor material in MFE devices.
- Significant efforts mounted to study W as a PFC material, both experimental and modeling.
- Significantly less effort devoted to study W as a structural material—for several technical reasons—, with a gap of several decades in experimental work and sporadic modeling work in the last decade or so.
- In light of the lack of suitable neutron sources, what about modeling of neutron damage in W?
The place of W as a structural material in fusion technology

Zinkle and Ghoniem (2000)

- W
- Mo (TZM)
- Ta-8W-2Hf
- Nb-1Zr-.1C
- V-4Cr-4Ti
- ODS ferritic st.
- F/M steel
- 316 SS
- CuNiBe
- SiC/SiC

Temperature (C)

uncertainties in minimum temperature limit
uncertainties in maximum temperature limit
Swelling behavior of W and W-Re alloys

- Irradiations in EBR-II up to ~9 dpa:
  - Herschitz and Seidman [Acta Metall. (1984); NIMPR-B (1985)] irradiated W-10%Re and observed no voids, formation of (coherent) disc-shaped precipitates.

- $\sigma$ and $\chi$ phases can form after sustained irradiation leading to severe embrittlement [Cottrell JNM (2004); He et al, JNM (2008)].
Hasegawa and co-workers have performed fast-neutron irradiations of well annealed W specimens at 400~800°C. 
- Voids and dislocation loops are observed above 0.1 dpa.
- Void lattices formed at higher doses (≈1 dpa).
- At higher doses, >5 dpa, transmutation into Re cannot be neglected → Re needle precipitates.
- Re precipitates suppress damage accumulation, increase hardening.

[Tanno et al, JNM (2009); Tanno et al, Mat Trans. (2011); Hasegawa et al, JNM (2011)]
Void lattice formation in irradiated arc-melted W

<table>
<thead>
<tr>
<th>Temperature</th>
<th>DPA</th>
</tr>
</thead>
<tbody>
<tr>
<td>538°C/0.96dpa</td>
<td></td>
</tr>
<tr>
<td>750°C/1.54dpa</td>
<td></td>
</tr>
</tbody>
</table>

[Hasegawa et al, Mat Trans (2013)]
Loops and precipitates in irradiated W-Re and W-Re-Os alloys

[Hasegawa et al, Mat Trans (2013)]
Radiation induced (enhanced?) precipitation results in significant hardening.

- Subsaturated precipitation of Re in W!
- Why such huge hardening?

[Tano et al, JNM (2009)]
What about neutron damage accumulation in W?

- Calculation of damage accumulation into the dpa range requires a special compromise of accuracy vs efficiency.
- Discuss mean-field approximation vs spatial correlations.
- Make connection with microscopic scale by parameterization of physical coefficients.
- Study damage accumulation in W under fast and fusion neutron irradiation conditions.
Reduced fundamental master equation for irradiation damage accumulation
Reduced fundamental master equation for irradiation damage accumulation

\[ \dot{C}_i = g_i - s_i C_i + \sum_j \left( \sum_k k_{jk} C_j C_k - k_{ij} C_i C_j \right) + \sum_j \sum_k \alpha_{ijk} C_i C_j C_k \ldots \]
Reduced fundamental master equation for irradiation damage accumulation

\[
\dot{C}_i = g_i - s_i C_i + \sum_j \left( \sum_k k_{jk} C_j C_k - k_{ij} C_i C_j \right) + \\
+ \sum_j \sum_k \alpha_{ijk} C_i C_j C_k \ldots
\]

\{g_i\}  \quad 0^{th} \text{ order term (source term)}

\{s_i\}  \quad 1^{st} \text{ order reactions (thermal dissolution, annihilation at sinks)}

\{k_{ij}\}  \quad 2^{nd} \text{ order reactions (clustering, recombination)}
Reduced fundamental master equation for irradiation damage accumulation

\[ \dot{C}_i = g_i - s_i C_i + \sum_j \left( \sum_k k_{jk} C_j C_k - k_{ij} C_i C_j \right) + \sum_j \sum_k \alpha_{ijk} C_i C_j C_k \ldots \]

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1^{st} order reactions (thermal dissolution, annihilation at sinks)

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All the physics (mechanisms, rate constants, etc) is included via these coefficients
Reduced fundamental master equation for irradiation damage accumulation

\[ \dot{C}_i = g_i - s_i C_i + \sum_j \left( \sum_k k_{jk} C_j C_k - k_{ij} C_i C_j \right) + \sum_j \sum_k \alpha_{ijk} C_i C_j C_k \ldots \]

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All the physics (mechanisms, rate constants, etc) is included via these coefficients

• Highly non-linear (\{k_{ij}\} not sparse)
• Highly-spatially correlated (\{s_i\} and \{k_{ij}\} depend on \(C\))
• Highly-dimensional (\(i\) potentially very large)
How do we solve this chemical master equation?

**MFRT** provides deterministic time-dependent solutions:
- *Workhorse* method for irradiation damage calculations.
- Numerically efficient for large systems of equations.
- Disregards spatial correlations (based on *mean-field* approximation).
- Requires knowledge of coefficient matrices a priori.
- Prone to combinatorial explosion.

**kMC** provides stochastic integration of evolution equations:
- Capable of resolving spatial effects by solving spatially-dependent diffusion equation.
- Numerically inefficient (but accurate) in inhomogeneous domains and wasteful in homogeneous cases.
- Lots of recent work to study mechanisms but low doses achievable.
Physical coefficients in rate theory

- Coefficients can be obtained by solving the diffusion equation under mean-field conditions in idealized geometries:

\[
s_i = Z_i \left( \rho_d + 4\pi r_p \rho_p + \frac{\sqrt{\rho_d + 4\pi r_p \rho_p}}{d_{GB}} \right) D_i
\]

\[
k_{ij} = 4\pi (r_i + r_j) (D_i + D_j)
\]

- Need for more kMC and MD calculations to improve the accuracy and meaningfulness of these physical constants \( \rightarrow \) W-Re defect energetics.
Finite-volume stochastic cluster dynamics simulations of neutron damage in W

• Solve the chemical master equation using kinetic Monte Carlo [Marian and Bulatov, *JNM* (2011)].

• Allow for finite volume fluctuations, trivial treatment of multispecies irradiations.

Recast ODE system:

\[
\frac{dC_\nu}{dt} = g_\nu - \sum_\nu s(\nu \to \mu)C_\nu + \sum_\mu s(\mu \to \nu)C_\mu + \\
- \sum_{\lambda\mu} k(\nu + \mu \to \lambda)C_\nu C_\mu + \sum_{\lambda\mu} k(\lambda + \mu \to \nu)C_\nu C_\mu
\]

into stochastic framework:

\[
\frac{dN_\nu}{dt} = \tilde{g}_\nu - \sum_\nu \tilde{s}(\nu \to \mu)N_\nu + \sum_\mu \tilde{s}(\mu \to \nu)N_\mu + \\
- \sum_{\lambda\mu} \tilde{k}(\nu + \mu \to \lambda)N_\nu N_\mu + \sum_{\lambda\mu} \tilde{k}(\lambda + \mu \to \nu)N_\nu N_\mu
\]

• \(N\): finite integer-valued population of clusters in a finite volume \(\Omega\).

• One defect reaction is executed per Monte Carlo cycle with correct rates.

• Monte Carlo rates are taken directly from the conventional RT coefficients:

\[
\tilde{g} = g\Omega \\
\tilde{s} = s \\
\tilde{k} = k/\Omega
\]
Stochastic cluster dynamics can be very efficient
Obtaining the damage source term in neutron irradiation of W:

- SPECTER calculations of PKA spectra using MCNP-calculated neutron fluxes in JOYO and ITER.
Calculations of damage source term using SPECTER

- Damage rate is function of damage cross section and damage model:

\[
\dot{k} = \rho \int \sigma_d(E) \phi(E) dE
\]

\[
\sigma_d(E) = \int_{E_{th}}^{E_l} \sigma_n(E, T) \nu(T) dT
\]

- Kinchin-Pease model:

\[
\nu(T) = \begin{cases} 
0, & T < E_{th} \\
1, & E_{th} < T < 2E_{th} \\
0.8T / 2E_{th}, & T > 2E_{th}
\end{cases}
\]
Calculations of damage source term using SPECTER

- Dose rates:
  - ITER: $5.7 \times 10^{-8}$ dpa s$^{-1}$ (ITER: 1.1 He appm/dpa)
  - JOYO: $3.5 \times 10^{-7}$ dpa s$^{-1}$

[Marian and Toang, JNM (2012)]

[Greenwood, JNM (1994)]
SCD has spatial resolution capabilities:

- From Becquart and Domain [JNM (2009, 2010)]

**Single Vacancies (nm^-3)**

- 0.04
- 0.03
- 0.02
- 0.01
- 0
Calculations of neutron transmutation of W in ITER

Very little transmuted He – High Os and Re transmutation rate – Decrease in thermal conductivity

[Gilbert and Sublet, *Nucl. Fusion* (2010)]
Parameterization of SCD calculations

- Cascade data from Troev et al [NIMPR-B (2011)]
- Clustering fractions from Fikar and Schäublin [JNM (2009)]

### 10 keV – 10 K

<table>
<thead>
<tr>
<th>Potential</th>
<th>vac</th>
<th>$V_{vac}$ (Å$^3$)</th>
<th>$V_{int}$ (Å$^3$)</th>
<th>vac in cl. (%)</th>
<th>int in cl. (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>F II</td>
<td>11.6 ± 0.8</td>
<td>3.8 ± 1.1</td>
<td>15.1 ± 4.2</td>
<td>33 ± 4</td>
<td>31 ± 8</td>
</tr>
<tr>
<td>A II</td>
<td>9.8 ± 0.8</td>
<td>4.1 ± 1.1</td>
<td>13.2 ± 3.8</td>
<td>27 ± 7</td>
<td>25 ± 9</td>
</tr>
<tr>
<td>D I</td>
<td>9.8 ± 0.6</td>
<td>4.9 ± 0.6</td>
<td>10.8 ± 2.2</td>
<td>19 ± 4</td>
<td>52 ± 5</td>
</tr>
<tr>
<td>D II</td>
<td>9.5 ± 0.4</td>
<td>4.2 ± 0.4</td>
<td>8.7 ± 1.9</td>
<td>22 ± 3</td>
<td>53 ± 4</td>
</tr>
</tbody>
</table>

### 10 keV – 523 K

<table>
<thead>
<tr>
<th>Potential</th>
<th>vac</th>
<th>$V_{vac}$ (Å$^3$)</th>
<th>$V_{int}$ (Å$^3$)</th>
<th>vac in cl. (%)</th>
<th>int in cl. (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>F II</td>
<td>9.7 ± 0.7</td>
<td>4.6 ± 0.8</td>
<td>48.5 ± 8.2</td>
<td>32 ± 7</td>
<td>19 ± 6</td>
</tr>
<tr>
<td>D I</td>
<td>6.8 ± 1.6</td>
<td>4.5 ± 1.3</td>
<td>11.3 ± 7.0</td>
<td>17 ± 12</td>
<td>58 ± 8</td>
</tr>
</tbody>
</table>

Vacancy clustering ratio: 30%
SIA clustering ratio: 25%

300K: $N_{fp}=1.5E^{0.82}$
Defect Energetics

- Energetics from Becquart and Domain [JNM (2009, 2010)]

\[
\begin{align*}
E^{\text{SIA}}_m &= 0.013 \text{ eV} \\
E^V_m &= 1.66 \text{ eV} \\
E^{V-V}_b &= -0.1 \text{ eV}
\end{align*}
\]
## Defect reactions

### From Becquart and Domain [JNM (2009, 2010)]

<table>
<thead>
<tr>
<th>Reaction</th>
<th>He binding energy (eV)</th>
<th>Reaction</th>
<th>Vacancy binding energy (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>He + v → He · v</td>
<td>4.57</td>
<td>v + He → He · v</td>
<td>4.57</td>
</tr>
<tr>
<td>He + He · v → 2He · v</td>
<td>3.11</td>
<td>v + He · v → He · 2v</td>
<td>0.07</td>
</tr>
<tr>
<td>He + 2He · v → 3He · v</td>
<td>3.28</td>
<td>v + He · 2v → He · 3v</td>
<td>0.70</td>
</tr>
<tr>
<td>He + 3He · v → 4He · v</td>
<td>2.61</td>
<td>v + He · 3v → He · 4v</td>
<td>1.01</td>
</tr>
<tr>
<td>He + 4He · v → 5He · v</td>
<td>1.44</td>
<td>v + 2He → 2He · v</td>
<td>6.65</td>
</tr>
<tr>
<td>He + 5He · v → 6He · v</td>
<td>2.08</td>
<td>v + 2He · v → 2He · 2v</td>
<td>1.82</td>
</tr>
<tr>
<td>He + 2v → He · 2v</td>
<td>4.69</td>
<td>v + 2He · 2v → 2He · 3v</td>
<td>0.68</td>
</tr>
<tr>
<td>He + He · 2v → 2He · 2v</td>
<td>4.85</td>
<td>v + 2He · 3v → 2He · 4v</td>
<td>1.29</td>
</tr>
<tr>
<td>He + 2He · 3v → 3He · 3v</td>
<td>3.97</td>
<td>v + 3He → 3He · v</td>
<td>8.57</td>
</tr>
<tr>
<td>He + 3v → He · 3v</td>
<td>5.35</td>
<td>v + 3He · v → 3He · 2v</td>
<td>2.65</td>
</tr>
<tr>
<td>He + He · 3v → 2He · 3v</td>
<td>4.83</td>
<td>v + 3He · 2v → 3He · 3v</td>
<td>1.83</td>
</tr>
<tr>
<td>He + 2He · 3v → 3He · 3v</td>
<td>5.08</td>
<td>v + 3He · 3v → 3He · 4v</td>
<td>1.49</td>
</tr>
<tr>
<td>He + 4v → He · 4v</td>
<td>5.74</td>
<td>v + 4He → 4He · v</td>
<td>10.22</td>
</tr>
<tr>
<td>He + He · 4v → 2He · 4v</td>
<td>5.11</td>
<td></td>
<td></td>
</tr>
<tr>
<td>He + 2He · 4v → 3He · 4v</td>
<td>5.29</td>
<td></td>
<td></td>
</tr>
<tr>
<td>He + 3He · 4v → 4He · 4v</td>
<td>5.17</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Results
Modeling the temperature dependence of defect accumulation in ITER (reflector)

[Figure: Graph showing the number density (SIA Loops and He bubbles) as a function of dpa (displacements per atom) for different temperatures (400°C, 500°C, 550°C). The graph includes multiple curves for different conditions and temperatures, illustrating the variation in defect accumulation with increasing dpa.

[Marian and Hoang, J. Nucl. Mat (2012)]]
Modeling the temperature dependence of swelling in W:
Void size histogram at maximum doses:

![Histogram of void sizes at different temperatures for JOYO and ITER.](image-url)

- JOYO:
  - 400°C: 0.17
  - 538°C: 0.40
  - 583°C: 0.96

- ITER:
  - 400°C: 0.10
  - 500°C: 0.44
  - 550°C: 1.54

Number density $[\text{m}^{-3}]$ vs. Size [nm]
SIA cluster size histogram at maximum doses:

![Histogram of SIA cluster sizes at 400°C, 500°C, and 550°C](chart)

- **400°C**: Number density [m^{-3}] = 10^{22} (0.10), 10^{21} (0.44), 10^{20} (1.54)
- **500°C**: Number density [m^{-3}] = 10^{22} (0.10), 10^{21} (0.44), 10^{20} (1.54)
- **550°C**: Number density [m^{-3}] = 10^{22} (0.10), 10^{21} (0.44), 10^{20} (1.54)

**Size [nm]**

- 1 nm
- 2 nm
- 3 nm
- 4 nm
- 5 nm
- 6 nm
Calculation of hardening from defect concentrations

\[ \Delta \sigma = M \alpha \frac{\mu b}{4\pi L} \]

Total hardening due to obstacles separates a distance \( L \)

\[ L = (\bar{D} \rho_l)^{-1/2} \]

\( L \) is function of average diameter and total loop density

\[ \bar{D} \rho_l = \int_{\Omega} D(n) \rho(n) \, dn \]

\( D(n) \) and \( \rho(n) \) taken from size histograms
Modeling the temperature dependence of defect accumulation in ITER (reflector)

<table>
<thead>
<tr>
<th></th>
<th>JOYO irradiations</th>
<th>ITER irradiations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dose rate (dpa s(^{-1}))</td>
<td>(3.5 \times 10^{-7})</td>
<td>(5.8 \times 10^{-8})</td>
</tr>
<tr>
<td>He production (appm dpa(^{-1}))</td>
<td>(2.5 \times 10^{-11})</td>
<td>1.1</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>400</td>
<td>500</td>
</tr>
<tr>
<td>Dose (dpa)</td>
<td>0.17</td>
<td>0.44</td>
</tr>
<tr>
<td>Hardening (MPa)</td>
<td>68.5(\sim)136.9</td>
<td>23.1(\sim)46.2</td>
</tr>
</tbody>
</table>
Final thoughts

- Mesoscale model of neutron damage accumulation in W under fusion conditions.
- Still much work remains to be done to include effect of Re/Os/Pt and H in damage models for irradiated W.
- Mesoscale models need to be informed by accurate defect energetics and kinetic parameters obtained in more physical conditions.