Overview of Radiation Damage Effects

Steve Roberts
U. Oxford
May 19, 2015

2nd RaDIATE Collaboration Meeting
RAL
Radiation damage in Materials

The interaction of radiation damage processes.
Transmutation
# Tungsten Transmutations in Fusion Reactor

**Time: 0.00 seconds**

<table>
<thead>
<tr>
<th>79</th>
<th>Au 185</th>
<th>Au 186</th>
<th>Au 187</th>
<th>Au 188</th>
<th>Au 189</th>
<th>Au 190</th>
<th>Au 191</th>
<th>Au 192</th>
<th>Au 193</th>
<th>Au 194</th>
<th>Au 195</th>
<th>Au 196</th>
</tr>
</thead>
<tbody>
<tr>
<td>78</td>
<td>Pt 183</td>
<td>Pt 184</td>
<td>Pt 185</td>
<td>Pt 186</td>
<td>Pt 187</td>
<td>Pt 188</td>
<td>Pt 189</td>
<td>Pt 190</td>
<td>Pt 191</td>
<td>Pt 192</td>
<td>Pt 193</td>
<td>Pt 194</td>
</tr>
<tr>
<td>77</td>
<td>Ir 182</td>
<td>Ir 183</td>
<td>Ir 184</td>
<td>Ir 185</td>
<td>Ir 186</td>
<td>Ir 187</td>
<td>Ir 188</td>
<td>Ir 189</td>
<td>Ir 190</td>
<td>Ir 191</td>
<td>Ir 192</td>
<td>Ir 193</td>
</tr>
<tr>
<td>76</td>
<td>Os 181</td>
<td>Os 182</td>
<td>Os 183</td>
<td>Os 184</td>
<td>Os 185</td>
<td>Os 186</td>
<td>Os 187</td>
<td>Os 188</td>
<td>Os 189</td>
<td>Os 190</td>
<td>Os 191</td>
<td>Os 192</td>
</tr>
<tr>
<td>75</td>
<td>Re 180</td>
<td>Re 181</td>
<td>Re 182</td>
<td>Re 183</td>
<td>Re 184</td>
<td>Re 185</td>
<td>Re 186</td>
<td>Re 187</td>
<td>Re 188</td>
<td>Re 189</td>
<td>Re 190</td>
<td>Re 191</td>
</tr>
<tr>
<td>74</td>
<td>W 179</td>
<td>W 180</td>
<td>W 181</td>
<td>W 182</td>
<td>W 183</td>
<td>W 184</td>
<td>W 185</td>
<td>W 186</td>
<td>W 187</td>
<td>W 188</td>
<td>W 189</td>
<td>W 190</td>
</tr>
<tr>
<td>73</td>
<td>Ta 178</td>
<td>Ta 179</td>
<td>Ta 180</td>
<td>Ta 181</td>
<td>Ta 182</td>
<td>Ta 183</td>
<td>Ta 184</td>
<td>Ta 185</td>
<td>Ta 186</td>
<td>Ta 187</td>
<td>Ta 188</td>
<td>Ta 189</td>
</tr>
<tr>
<td>72</td>
<td>Hf 177</td>
<td>Hf 178</td>
<td>Hf 179</td>
<td>Hf 180</td>
<td>Hf 181</td>
<td>Hf 182</td>
<td>Hf 183</td>
<td>Hf 184</td>
<td>Hf 185</td>
<td>Hf 186</td>
<td>Hf 187</td>
<td></td>
</tr>
<tr>
<td>71</td>
<td>Lu 176</td>
<td>Lu 177</td>
<td>Lu 178</td>
<td>Lu 179</td>
<td>Lu 180</td>
<td>Lu 181</td>
<td>Lu 182</td>
<td>Lu 183</td>
<td>Lu 184</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Z**  
105 106 107 108 109 110 111 112 113 114 115 116 117

**N**  
1 2 3 4

Pure W irradiated in a DEMO divertor armour spectrum  
Total flux: $5.49 \times 10^{14}$ n cm$^{-2}$ s$^{-1}$  
m - concentration dominated by metastable nuclide(s)
### Tungsten Transmutations - activity

**Time:** 0.00 seconds  
**Total Dose Rate (Sv/hr):** 0.000E+00

<table>
<thead>
<tr>
<th>79</th>
<th>Au 185</th>
<th>Au 186</th>
<th>Au 187</th>
<th>Au 188</th>
<th>Au 189</th>
<th>Au 190</th>
<th>Au 191</th>
<th>Au 192</th>
<th>Au 193</th>
<th>Au 194</th>
<th>Au 195</th>
<th>Au 196</th>
</tr>
</thead>
<tbody>
<tr>
<td>78</td>
<td>Pt 183</td>
<td>Pt 184</td>
<td>Pt 185</td>
<td>Pt 186</td>
<td>Pt 187</td>
<td>Pt 188</td>
<td>Pt 189</td>
<td>Pt 190</td>
<td>Pt 191</td>
<td>Pt 192</td>
<td>Pt 193</td>
<td>Pt 194</td>
</tr>
<tr>
<td>77</td>
<td>Ir 182</td>
<td>Ir 183</td>
<td>Ir 184</td>
<td>Ir 185</td>
<td>Ir 186</td>
<td>Ir 187</td>
<td>Ir 188</td>
<td>Ir 189</td>
<td>Ir 190</td>
<td>Ir 191</td>
<td>Ir 192</td>
<td>Ir 193</td>
</tr>
<tr>
<td>76</td>
<td>Os 181</td>
<td>Os 182</td>
<td>Os 183</td>
<td>Os 184</td>
<td>Os 185</td>
<td>Os 186</td>
<td>Os 187</td>
<td>Os 188</td>
<td>Os 189</td>
<td>Os 190</td>
<td>Os 191</td>
<td>Os 192</td>
</tr>
<tr>
<td>75</td>
<td>Re 180</td>
<td>Re 181</td>
<td>Re 182</td>
<td>Re 183</td>
<td>Re 184</td>
<td>Re 185</td>
<td>Re 186</td>
<td>Re 187</td>
<td>Re 188</td>
<td>Re 189</td>
<td>Re 190</td>
<td>Re 191</td>
</tr>
<tr>
<td>74</td>
<td>W 179</td>
<td>W 180</td>
<td>W 181</td>
<td>W 182</td>
<td>W 183</td>
<td>W 184</td>
<td>W 185</td>
<td>W 186</td>
<td>W 187</td>
<td>W 188</td>
<td>W 189</td>
<td>W 190</td>
</tr>
<tr>
<td>73</td>
<td>Ta 178</td>
<td>Ta 179</td>
<td>Ta 180</td>
<td>Ta 181</td>
<td>Ta 182</td>
<td>Ta 183</td>
<td>Ta 184</td>
<td>Ta 185</td>
<td>Ta 186</td>
<td>Ta 187</td>
<td>Ta 188</td>
<td>Ta 189</td>
</tr>
<tr>
<td>72</td>
<td>Hf 177</td>
<td>Hf 178</td>
<td>Hf 179</td>
<td>Hf 180</td>
<td>Hf 181</td>
<td>Hf 182</td>
<td>Hf 183</td>
<td>Hf 184</td>
<td>Hf 185</td>
<td>Hf 186</td>
<td>Hf 187</td>
<td></td>
</tr>
<tr>
<td>71</td>
<td>Lu 176</td>
<td>Lu 177</td>
<td>Lu 178</td>
<td>Lu 179</td>
<td>Lu 180</td>
<td>Lu 181</td>
<td>Lu 182</td>
<td>Lu 183</td>
<td>Lu 184</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**pure W irradiated in a DEMO FW armour spectrum**  
**Total flux:** $5.49 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$  

$\text{m} - \text{Dose Rate dominated by metastable nuclide(s)}$

---

Tungsten transmutation in fusion power reactor

- Pure W under outboard equatorial FW armour flux for 5 fpy
Tungsten transmutation in fusion power reactor

- Pure W under outboard equatorial FW armour flux for 5 fpy

Graph showing the concentration of various elements over irradiation time and percentage of W.
Tungsten transmutation in fusion power reactor

- Ion irradiation can’t mimic transmutation effects

- But we can MODEL what transmutations we expect

- And can ion-irradiate pre-made W alloys

- To see what effect displacement damage -

  - (and if we like, He, H )

- have on microstructures and properties.
Radiation-induced clustering in alloys

W- 5%Re should be a stable solid solution
… but it’s not when it is irradiated

Under Fusion Power Reactor conditions, pure W transmutes to give 5%Re in ~ 7 years.

Ion irradiation to $2.64 \times 10^{15} \text{ W}^+/\text{cm}^2$ (33dpa)
Dose rate: $3.57 \times 10^{-4} \text{ dpa} / \text{s}$
Temperature: $300^\circ\text{C}$

A. Xu (Oxford) unpublished 2013
Clustering and Hardening in “Transmuted” W

Preliminary analysis indicates that the clusters are very weak obstacles:

\[ \Phi_c \approx 85^\circ \]

But there are lots of them, very closely spaced - especially in W-Re-Os

Same would apply to neutron irradiation ???
Transmutation gases
Transmutations producing He in fusion power reactor
Effects of Implanted He on T91 steel

"Uniform implantation of 23 MeV α-particles up to 5000 appm at 250°C carried out at the compact cyclotron of Forschungszentrum Jülich (FZJ)"
Tungsten – Hardening by Displacement Damage and Helium

Polycrystalline W 99.99% pure, sequentially ion-implanted with W⁺ and He⁺ at 300°C. W implantation depth 0 - 200nm; He implantation depth range 0 - 2500 nm. Hardness data at 100 nm indenter depth.

Tungsten – Hardening by Displacement Damage and Helium

Polycrystalline W 99.99% pure, sequentially ion-implanted with W\(^+\) and He\(^+\) at 300°C
W implantation depth 0 - 200nm; He implantation depth range 0 - 2500 nm
Hardness data at 100 nm indenter depth.

**Tungsten – Embrittlement by Displacement Damage and Helium**

<table>
<thead>
<tr>
<th>Irradiation</th>
<th>Fracture Data</th>
</tr>
</thead>
<tbody>
<tr>
<td>none</td>
<td>0 / 7</td>
</tr>
<tr>
<td>1.7 dpa W^{12+}</td>
<td>1 / 16</td>
</tr>
<tr>
<td>600 appm He</td>
<td>0 / 13</td>
</tr>
<tr>
<td>1.7 dpa W^{12+} + 600 appm He</td>
<td>7 / 10</td>
</tr>
</tbody>
</table>

Polycrystalline W 99.99% pure, ion-implanted with W^{12+} (36MeV) and He^+ at 800°C

W + He simultaneously implanted

W implantation depth ~2.5 µm; He depth range ~2.8 µm

Fracture data from microbeams 3 µm deep.

J. Gibson & D. Armstrong (Oxford) *unpublished* 2013
“Damage”
Elastic scattering of neutrons

.... What about the “target” atoms?

<table>
<thead>
<tr>
<th></th>
<th>$E_d$</th>
<th></th>
<th>$E_d$</th>
<th></th>
<th>$E_d$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>~32eV</td>
<td>Fe</td>
<td>~24eV</td>
<td>Si</td>
<td>~13eV</td>
</tr>
<tr>
<td>Cu</td>
<td>~25eV</td>
<td>W</td>
<td>~40eV</td>
<td>C</td>
<td>~25-60eV</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(graphite)</td>
</tr>
</tbody>
</table>

Fusion neutrons are ~14MeV!
Elastic scattering of neutrons

\[ v_1^2 = 2(1 - \cos \phi) \frac{u_1^2 M_1^2}{(M_1 + M_2)^2} \]

\[ E_2 = \Lambda E_1 \sin^2(\phi/2) \]

\[ \bar{E}_2 = \Lambda E_1 / 2 \]

For 14 MeV neutrons into iron, \( M_1 = 1 \), \( M_2 = 56 \), so \( \Lambda = 0.069 \)

So \( \bar{E}_2 = 482 \text{ keV} \) .... about 34 collisions per neutron
Elastic scattering of neutrons

\[ I = I_0 \exp(-N\sigma x) \]

For \( \sigma = 3.2 \) barns, 
\( N = 8 \times 10^{28} \text{ m}^{-3} \)

Neutrons penetrate in Fe to 100 – 200 mm

If \~30\ collisions over this depth, and “random walk”,

Spacing between neutron collisions is \~10 - 20\ mm
The collision cascade

Energy loss from C ions in graphite

At high energies, ions lose energy mostly by electronic excitation
At low energies, ions lose energy mostly by collisions

\[ E_{ex} (\text{keV}) \sim M^2 \text{ (a.m.u.)} \]
The collision cascade

Tracks of ten 482keV Fe ions in Fe target.

“Consequent events” show in red and green.

Note long gaps between sub-cascades, and randomness of process.
Ion irradiation: dpa

Displaced target atoms vs depth
Unit: Number / (Å – ion) (!)

\[ dpa = \frac{D \phi}{\rho} \]

- \( D \): displacements / (m – ion)
- \( \phi \): irradiating particles / m^2
- \( \rho \): atoms / m^3 in target

Here: Fe ions into Fe

- \( D \): ~ 4.5 at peak
- \( \rho \): \( 8 \times 10^{28} \) m^-3

\( \phi \): typically \( 10^{18} \) m^-2 (\( 10^{14} \) cm^-2)

\[ = 0.56 \text{ dpa} \]
The collision cascade: evolution after initiation

After $i^{th}$ stage, we have $2^i$ displaced atoms:

 Stops when: $\frac{E_k}{2^i} < 2E_d$

So $N_{\text{displacements}} = 2^i = \frac{E_k}{2E_d}$
The collision cascade: evolution after initiation

"Molecular dynamics" simulation: up to 30 ps.

Only the out-of-place atoms are shown – vacancies in green; self interstitials in red.

Started by a low-energy collision (10keV ion): end of cascade branch.

10 keV < $E_{ex}$, so $N_{\text{displacement}} \approx 10000/25 = 400$

D. Bacon, Y. Osetsky, U. Liverpool
Radiation ➔ dislocation loops
Radiation → dislocation loops

Iron, ion-irradiated to 2.5 dpa at 300°C
Dislocation loops in Fe irradiated at 500°C

Dose: $3 \times 10^{18}$ ions m$^{-2}$,  
Dose: $1 \times 10^{19}$ ions m$^{-2}$

UHP-Fe irradiated at 500°C with 150 keV Fe$^+$ ions
- loops are of $<100>$ type.
- loops are of *interstitial* type
Radiation Damage as it happens…

IVEM – Argonne NL

MIAMI – U. Huddersfield
Damage accumulation; Fe, 300°C, high dose
Loop density vs dose: W-5%Re

$g=200$

0  0.38

Unit: $10^{13}W^+/cm^2$
Loop density vs dose: W-5%Re

Unit: $10^{13}W^+/cm^2$
Loop density vs dose: W-5%Re

Unit: $10^{13}W^+/cm^2$
Loop density vs dose: W-5%Re

$g = 200$

0 0.38 1.0 1.75 3.25

Unit: $10^{13}W^+/cm^2$

~10 nm 100 nm
Loop density vs dose: W-5%Re

Unit: $10^{13}$ W$^+/\text{cm}^2$

$g=200$

$0 \quad 0.38 \quad 1.0 \quad 1.75 \quad 3.25 \quad 6.25$

$\sim 10^{15}$ loops/m$^2$
Loop density vs dose: W-5%Re

Unit: $10^{13} W^+/cm^2$

$g=200$

$\sim 10^{15}$ loops/m$^2$
Self-ion irradiations of pure materials and FeCr alloys - 100keV room temperature irradiations

Iron-based metals (Body centred cubic metals in general?) are more resistant to radiation damage.

- Fewer defects
- Threshold dose
Effects of radiation damage on materials

1. **Sputtering**
2. **Stored energy**
   a) “Wigner energy” release
3. **Point defect accumulation**
   a) Swelling, dislocation loops, nano-voids
   b) Changes in thermal & electrical conductivity
4. **Enhanced diffusion**
   a) Radiation-enhanced creep
   b) Faster kinetics of phase transitions
   c) Radiation-enhanced grain-boundary segregation
5. **Gas from transmutation**
   a) Bubbles, swelling (with 2)
6. **Soluble and insoluble transmutation products**
   a) Hardening, precipitation
7. **Knock-on randomisation of structure**
8. **Non-equilibrium microstructures?** (related to 3. 6)
9. **Hardening** (from 2, 4, 5)
10. **Embrittlement** (from 7, 3c, 4)
11. **Radiation-enhanced corrosion** (related to 3a, maybe 5)
Radiation-induced swelling

Type 316 stainless steel:
Fe 16-18%Cr, 10-14%Ni, 2%Mo, <0.08%C

Straalsund, 1982, and F. Garner
Radiation-induced swelling

- Increased Sinks
- 304 SS
- 316 SS
- CW 316 SS
- D9 (CW, ppt, alloying additions)
- HT9

Dose vs. Swelling (%)

- Increased Ni
- Cold work
- ODS (v. high sinks)

G. Was, Michigan
Radiation-induced grain boundary segregation

Type 304L stainless steel: Fe 18-20%Cr, 8-12%Ni, <0.03%C

- Depletion of Cr and enrichment of Ni are consistent among all the irradiations.
- Self-ion irradiations produced wider and shallower segregation profiles compared to those by neutron irradiations

Michigan Engineering
G. Was, Michigan
Radiation-induced dislocation loops: effects on mechanical properties

Copper

3.5 x 10^2 dpa
γ = 15%

"Strain localisation"

τ [MPa]

yield point

unirradiated
1.1 x 10^{-4} dpa
7.9 x 10^{-4} dpa
6.6 x 10^{-3} dpa
3.9 x 10^{-2} dpa

γ [%]

yield region
Stress-strain curves: Iron alloys

T91 steel

Eurofer 97

Fe Cr alloys

- n-irradiated up to 1.5 dpa
- $T_{\text{irr}} = 300^\circ\text{C}$
- $T_{\text{TEST}} = 27^\circ\text{C}$

**T91**:
- Fe - 8.3%Cr – 0.4%Mn - 1%Mo - 0.2%V - 0.08% Nb - 0.1%C

**Eurofer 97**:
- Fe - 8.9%Cr – 1%W - 0.2%V - 0.14%Ta - 0.12%C
Stress-strain curves: Iron alloys

Plastic instability after yield

Eurofer 97

n-irrad up to 1.5 dpa
$T_{\text{irr}} = 300^\circ\text{C}$,
$T_{\text{TEST}} = 27^\circ\text{C}$
# Dose Rates

<table>
<thead>
<tr>
<th>Radiation Source</th>
<th>Dose Rate (dpa / s) in Iron-based Alloys</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fission Reactor Pressure Vessels (RPV)</td>
<td>~ $10^{-12} - 10^{-11}$</td>
</tr>
<tr>
<td>Rotating Target Neutron Source (RNTS-II)</td>
<td>~ $10^{-10}$</td>
</tr>
<tr>
<td>Fast Flux Test Facility (FFTF)/DEMO Fusion Reactor</td>
<td>~ $10^{-8} - 10^{-6}$</td>
</tr>
<tr>
<td>Ion Implantation – Low Dose Rate</td>
<td>3 x $10^{-5}$</td>
</tr>
<tr>
<td>Ion Implantation – High Dose Rate</td>
<td>6 x $10^{-4}$</td>
</tr>
<tr>
<td>HVEM Irradiation</td>
<td>~ $10^{-3}$</td>
</tr>
</tbody>
</table>
Nanoindentation hardness: Fe 5%Cr 0.6dpa

Hardness (GPa)

Fe 5%Cr
Hardness vs Displacement Into Surface

Low Dose Rate
High Dose Rate
Un-irradiated

Nominal dose rate
0.5MeV - 1.0MeV - 2.0MeV

Displacement Into Surface (nm)
Nanoindentation hardness: Fe 5%Cr 0.6dpa

Fe 5%Cr
Hardness vs Displacement Into Surface

Hardness (GPa)

Low Dose Rate
High Dose Rate
Un-irradiated

Displacement Into Surface (nm)
TEM of ion irradiation damage Fe 5%Cr 300°C

Low dose rate

High dose rate
APT data for the Fe 5%Cr alloy irradiated at 400°C with the low dose rate (a) and high dose rate (b). The figure includes an atom map showing Fe atoms and a 0.5 at.% CrN isoconcentration surface (i) and proximity histograms showing the variation in composition from the centre of enrichment outwards into the matrix (ii). Centre of enrichment is defined as the region of highest CrN concentration.
Micro-mechanical Testing

Fe-12%Cr
Ion – irradiated
Fe+ 0.5MeV & 2MeV
320°C

0 dpa
6.8 dpa

20μm
2μm
Micro-mechanical Testing

Fe-12%Cr
Ion – irradiated
Fe⁺
0.5MeV & 2MeV
320°C

Fe-6%Cr
neutron – irradiated
288°C
1.7 dpa

Micromechanical testing Fe-6%Cr – Size effects

Yield Stress (GPa)

Beam depth (μm)

Ion-irradiated
Neutron-irradiated
Unirradiated

0.1mm
Micromechanical testing Fe-6%Cr – Size effects

Yield Stress (GPa) vs. Beam depth (μm)

- Ion-irradiated
- Neutron-irradiated
- Unirradiated

Beam depth (μm)
Micromechanical testing Fe-6%Cr – Size effects
Micromechanical testing Fe-6%Cr – Size effects
Protons ?
SRIM: 2 MeV protons into Fe
Proton irradiation for (micro) mechanics?

SRIM: 2 MeV protons into Fe

Could use specimens 10 – 12 µm thick – well beyond the size-dependent mechanics regime
Proton irradiation for (micro) mechanics?

SRIM: 2 MeV protons into Fe

For 2MeV H⁺ into Fe:

- Total damage: \( \sim 1 \times 10^{-4} \) events / (Å - ion)
- \( \Rightarrow \sim 10^{21} \) ions cm\(^{-2}\) for 1 dpa
- Depth \( \sim 15 \) μm
Proton irradiation for (micro) mechanics?

SRIM: 2 MeV protons into Fe

For 2 MeV H\(^+\) into Fe:
- Total damage: \(~1 \times 10^{-4}\) events / (Å - ion)
- \(\Rightarrow \sim 10^{21}\) ions cm\(^{-2}\) for 1 dpa
- Depth \(\sim 15\) \(\mu\)m

For 2 MeV Fe\(^+\) into Fe:
- Total damage: \(~4\) events / (Å - ion)
- \(\Rightarrow \sim 2 \times 10^{16}\) ions cm\(^{-2}\) for 1 dpa
- Depth \(\sim 1\) \(\mu\)m
Radiation damage in Materials

The interaction of radiation damage processes.