Overview on Irradiation Effects in Fusion Materials

Anton Möslang
High Performance Materials for Energy

First Reactors
- 1-3 dpa

Current Reactors
- < 200 dpa

Advanced Reactors
- < 200 dpa

Future Systems

Strategic Missions:
- Electricity, Hydrogen, Heat
- Contribute to lower greenhouse gas emission

Specific challenges for fusion:
- Short development path
- More demanding loading conditions
- ~30 times more Helium/dpa in steels

ITER
- 1-3 dpa

IFMIF

DEMO, Fusion Reactor
- < 150 dpa

20-40/year
Comparison: Fusion, Fission, Spallation
- Similarities and differences
- Requirements and performance goals for structural materials

Interaction of energetic particles (ions and neutrons) with condensed matter and related damage mechanisms
- Calculation of displacement damage („dpa“) in metals

Irradiation damage effects in fusion materials
- Mostly Steels and Oxide dispersion strengthened variants
- Present status, critical issues and prospects

Summary
FUSION: Requirements for “in vessel components”

**Blanket:** $\leq 30 \text{ dpa/yr, } 2.5 \text{ MW/m}^2$
- Plasma Facing Materials
- Reduced Activation Structural Materials:
  - RAFM Steels, EUROFER 250-550 °C
  - EUROFER-ODS 250-650 °C
- Functional Materials
  - Neutron multiplier (Be)
  - Tritium Breeder (Li$_4$SiO$_4$, …)

**Divertor:** $\leq 10 \text{ dpa/yr, } 10-15 \text{ MW/m}^2$
- Refractory alloys (e.g. W-ODS)
  - 850-1200 °C $\rightarrow$ 600 - 1300 °C
- Nano-scaled RAF-ODS Steels
  - 300-650 °C $\rightarrow$ 250 - 750 °C

Power: 1.30 GW$_e$
Plant Efficiency: 37-45%

A. Möslang, ISFNT-8, Keynote Heidelberg, 2007
### Requirements for “in vessel” structural materials

**Fission – Fusion – Spallation: Three different irradiation loadings**

<table>
<thead>
<tr>
<th></th>
<th>Fission (Gen. I)</th>
<th>Fission (Gen. IV)</th>
<th>Fusion (DEMO/PROTO)</th>
<th>Spallation (ADS)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Structural alloy $T_{\text{max}}$</td>
<td>&lt;300°C</td>
<td>500-1000°C</td>
<td>550-1000°C</td>
<td>400-600°C</td>
</tr>
<tr>
<td>Max dose for core internal structures</td>
<td>~1 dpa</td>
<td>~30-100 dpa</td>
<td>~150 dpa</td>
<td>≤60 dpa/fpy</td>
</tr>
<tr>
<td>Max transmutation helium concentration</td>
<td>~0.1 appm</td>
<td>~3-10 appm</td>
<td>~1500 appm (~10000 appm for SiC)</td>
<td>~2000 appm/fpy</td>
</tr>
<tr>
<td>Particle Energy $E_{\text{max}}$</td>
<td>&lt;1-2 MeV</td>
<td>&lt;1-3 MeV</td>
<td>&lt;14 MeV</td>
<td>several hundred MeV</td>
</tr>
</tbody>
</table>

Materials R&D towards:
- improved irradiation resistance
- enhanced temperature window
- convincing compatibility with coolants
Typical Particle energies:

Particle energies are usually measured in eV. 1 eV is the energy needed to accelerate one electron in an electric field of 1 Volt. \( 1 \text{eV} = 1.6022 \times 10^{-19} \text{J} \)

<table>
<thead>
<tr>
<th>Particle Type</th>
<th>Description</th>
<th>Energy Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \gamma )-irradiation</td>
<td>- from nuclear decay:</td>
<td>( \leq \sim 3 \text{ MeV} )</td>
</tr>
<tr>
<td></td>
<td>- fission technology:</td>
<td>( \leq \sim 5 \text{ MeV} )</td>
</tr>
<tr>
<td>X-rays</td>
<td>- Tomography</td>
<td>( \leq \sim 450 \text{ keV} )</td>
</tr>
<tr>
<td>Neutrons</td>
<td>- fission reactors</td>
<td>( \leq \sim 5 \text{ MeV} )</td>
</tr>
<tr>
<td></td>
<td>- fusion reactors</td>
<td>( \leq \sim 14 \text{ MeV} )</td>
</tr>
<tr>
<td></td>
<td>- spallation sources</td>
<td>( \leq \sim 600 \text{ MeV} )</td>
</tr>
<tr>
<td>Charges particles</td>
<td>- ( \alpha )-Particles</td>
<td>( \leq \sim 1-5 \text{ MeV} )</td>
</tr>
<tr>
<td></td>
<td>- atoms</td>
<td>displacement damage</td>
</tr>
<tr>
<td></td>
<td>- ( \beta ) decay</td>
<td></td>
</tr>
</tbody>
</table>
Irradiation effects depend seriously on neutron spectrum

Relevant for damage and transmutations in steels

neutron flux density \([\text{cm}^{-2} \text{s}^{-1} \text{MeV}^{-1}]\)

neutron energy \([\text{MeV}]\)

MTS sample can #3
MTS sample can #7
DEMO first wall
IFMIF back plate
IFMIF HFTM

A. Fischer, A. Möslang, P. Vladimirov, 2011
Ions and Neutrons: Interaction with matter - energy loss by scattering -

Primary damage:
Each projectile (e.g. Neutron, Electron, Ion) is scattered elastically, usually frequently at atoms in the solid state and creates thereby „PKA‘s“ (primary knock-on atoms) in in different orientations and with different Energies T (PKA—energy spectrum)

Secondary damage:
PKA's deposit their energy T as follows:
- inelastic collisions with electrons (T_{elektr})
- elastic nuclear reactions to create
  - Frenkel pairs (vacancy + interstitial atom)
  - cascade formation

Threshold energy $E_d$:
Min. energy to create a vacancy (V). $E_d$ depends on the lattice orientation
Crystal structure and material:
Fe: $<E_d> = 20-40$ eV
W: $<E_d> = 193$ eV
If the energy $T$ transferred to the lattice atom is very big, this energetic recoil atom (or primary knocked on atom PKA) will very effectively replace the surrounding atoms (equal masses, like in a billiard game):

- High point defect density
- Chemical disorder
- Topologic disorder

Displacement cascade in Ni$_3$Al, $T = 8$ keV

Vacancies, interstitial atoms, displaced atoms

(C. Lemaignan, M. Guttmann)
Interaction with matter: determination of the displacement damage

The displacement damage is usually measured in "Displacements per atom (dpa)". 1 dpa means, that during irradiation each atom is displaced in average one time.

Scattering events – like the scattering of ions on target atoms – is usually described via a **cross section**:

\[
\sigma = \frac{\text{number of "reactions" per scattering center/s}}{\text{current density of the incoming projectile}}
\]

With the unity \([s^{-1}/(s^{-1} cm^{-2}) = cm^2]\)

For better handling, often the unity "Barn" \([b]\) is used:

\[
1b = 10^{-28} m^2
\]

Light ion irradiation: In the energy range <30 MeV elastic nuclear reactions are dominating (that is, the sum of the kinetic energies = constant).
To determine the displacement damage, it is necessary, to handle the primary damage and the secondary damage differently:

**A. Primry damage**

For lighter ions with kinetic energies $E$ from ~5 keV to ~30 MeV the differential Rutherford’ cross section $d\sigma(E, T)$ is valid. It describes the „probability“, that an ion (projectile) of mass $M_1$, der atomic number $Z_1$ und the energy $E$ is scattered at a target atom of mass $M_2$ and the atomic number $Z_2$, and thereby transfers the energy $T+dT$:

\[
d\sigma(E, T) = 4\pi a_0^2 Z_1^2 Z_2^2 \frac{M_1 E_R^2}{M_2 E T^2} dT
\]

„Bohr‘ radius“ $a_0 = 0.053 \text{ nm}; E_R = e^2/a_0 = 13.6 \text{ eV} \)
A. Primary damage

In the center of mass system
The following equation is valid
\( (\theta \leq 180^\circ) \):

\[
T = \frac{4 M_1 M_2}{(M_1 M_2)^2} E \sin^2 \left( \Theta / 2 \right) = T_{\text{max}} \sin^2 \left( \Theta / 2 \right)
\]

(2)

From eq Gl. (2) follows: Even at \( E = \text{const.} \) the PKAs have an energy spectrum
From eqs (1) u. (2) follows: With increasing \( E \), the cross sections decline with \( 1/E^3 \)
B. Secondary damage

The PKA deposit the Energy $T$ (often $T \gg E_d$) along a displacement cascade on the surrounding atoms, until it becomes thermalized. The energy $T$ splits into

- displacement energy $T_{\text{dam}}$ (formation of FPs & sub cascades)
- Electronic excitation energy $T_{\text{electr}}$

The number of displaced atoms $\nu(T)$ is then (internat. recommendation):

$$\nu(T) = 0 \quad \text{for} \quad T_{\text{dam}} < E_d$$
$$\nu(T) = 1 \quad \text{for} \quad E_d \leq T_{\text{dam}} \leq 2E_d$$
$$\nu(T) = \frac{k \cdot T_{\text{dam}}}{2E_d} \quad \text{for} \quad T_{\text{dam}} \geq 2E_d$$

(3)

The displacement efficiency $k$ is expected to be constant (NRT): $k = 0.8$; in Fe: $E_d = 40$ eV

More detailed analysis by Molecular Dynamics calculations
B. Secondary damage

- The damage energy $T_{\text{dam}}$ has a maximum. PKA-Energies above $10^5$ eV are, however, in praxis very seldom.

- With increasing PKA-Energy $T_{\text{elektr}}$ increases strongly and dominates above $3 \times 10^5$ eV completely (heat production).

**Example:** How much displacement damage we have in Fe for $T_{\text{dam}} = 2 \times 10^5$ eV?

From Eq. (3) follows:

$$\nu(T) = 2000$$ displacements!
Ions: Interaction with matter: 
- visualizing the secondary damage -

The PKA-atom we have spoken about....

MARLOWE binary collision code

PKA atom
**Ions:** Interaction with matter: determination of the displacement damage

### C. Combination of primary and secondary damage

To combine the primary damage (probability to create a PKA) with the secondary damage (number of displaced atoms per PKA), Eq. (1) is combined with eq. (3) and integrated over the entire energy range $T$. Consequently the **total displacement cross section** for ions of energy $E$ is:

$$\sigma_d(E) = \int_{E_d}^{T_{\text{max}}} \left[ \frac{d\sigma(E,T)}{dT} \right] \cdot \nu(T) dT$$  \hspace{1cm} (4)

That is, $\sigma_d(E)$ describes the average number of displacements per lattice atom and per ion/cm$^2$.

The non-analytical Integral in Eq. (7) can be solved numerically (A. Möslang KfK Report 03.02.02P94B (1990))
C. Combination of primary and secondary damage

Die total displacement cross section follows then simply by multiplication with the ion current \( \Phi \) and the total irradiation time \( t \):

\[
\text{Displacement damage [dpa]} = \sigma_d(E) \cdot \Phi \cdot t \quad (5)
\]

Units:

\[
\frac{\text{displ./Atom}}{\text{Ion/cm}^2} \cdot \frac{\text{Ions}}{\text{cm}^2 \cdot \text{s}} \cdot [\text{s}]
\]

In this how for ion irradiations the displacement damage can be calculated.
Neutrons: Interactions with matter:
Determination of displacement damage

- As neutrons do not have an electric charge, they cannot transfer any energy to electrons. Neutrons can only “communicate” via the so-called „Strong Interaction“ with Protons and Neutrons and consequently can transfer also energy via this interaction.

- The range of neutrons in matter is therefore substantially longer as the range of ions, electrons or γ-radiation.

- That is, neutrons can only scatter at the nuclei via:
  - elastic interaction (sum of kinetic energy = const)
  - inelastic interaction (e.g. emission of γ-radiation)
  - non-elastic interaction (nuclear transformation via particle emission (n, p, α-particles,…)

  Creation of long living radioactive isotopes

- In the vast majority, neutrons are not mono-energetic but have a broad energy spectrum.
Therefore, to calculate the (average) **displacement cross-section for neutrons**, we have to integrate not only over all PKA-energies $T$, but in addition also over all neutron energies $E$.

From this it follows for neutrons from Eq. (7):

$$
\sigma_d = \int_0^{E_{\text{max}}} \left( \frac{d\Phi}{dE} \right) dE \int_{E_d}^{T_{\text{max}}} \frac{d\sigma(E,T)}{dT} \cdot \nu(T) dT
$$

$d\Phi/dE$ shows, how much neutrons of a given energy $E'$ we have in the intervall $(E'+dE)$.
In analogy to Eq. (4), the total displacement cross section is then obviously given by multiplication of the integral neutron flux density $\Phi_{tot}$ and the total irradiation time $t$:

$$
\text{Displacement damage [dpa]} = \sigma_d \cdot \Phi_{tot} \cdot t
$$

(7)
Development of cascades in time

(C. Lemaignan, M. Guttmann, EDF DER 1994)

Only very few defects survive!
Peak damage state in iron cascades at 100K

- DT fusion neutrons produce atomic recoils at much higher energies than fission neutrons
- Use of large-scale atomic simulations demonstrates that subcascade formation minimizes differences in defect production
- Average defect production per unit cascade energy is essentially the same for fission and fusion
- However, differences between fusion and fission irradiation are expected at high dose due to fusion’s higher transmutant He and H production

Courtesy of R. Stoller, ORNL
Criteria for adequate materials irradiation: The «correct» PKA-Spectrum

<table>
<thead>
<tr>
<th>Particle type ((E_{\text{kin}} = 1) MeV)</th>
<th>Typical recoil (or PKA) feature</th>
<th>Typical recoil energy (T)</th>
<th>Dominant defect type</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electron</td>
<td><img src="#" alt="PKA" /></td>
<td>25 eV</td>
<td>Frenkelpairs (FP: Vacancy-Interstitial pair)</td>
</tr>
<tr>
<td>Proton</td>
<td><img src="#" alt="PKA" /></td>
<td>500 eV</td>
<td></td>
</tr>
<tr>
<td>Fe-ion</td>
<td><img src="#" alt="PKA" /></td>
<td>24 000 eV</td>
<td>Cascades &amp; sub-cascades</td>
</tr>
<tr>
<td>Neutron</td>
<td><img src="#" alt="PKA" /></td>
<td>45 000 eV</td>
<td></td>
</tr>
</tbody>
</table>

Typical impact on materials properties:
- FPs as “freely migrating defects”: Alloy dissolution, segregation, irradiation creep
- Cascades & sub-cascades: Irradiation hardening, ductility reduction
MD Simulation und comparison with TEM
- Interaction of energetic projectiles with condensed matter -

20 keV cascade in iron at 100K

T$_{irr}$ = 250 °C, 2.5 dpa
IA dominated

50 nm

E. Materna-Morris, IMF I

20 keV cascade in iron at 100K

T$_{irr}$ = 450 °C, 2.5 dpa
Vac. dominated

50 nm

- R. Stoller -
Ferritic/Martensitic Steels vs. Austenitic Steels

Irradiation-induced swelling

CW 316 steel

unirradiated

Irradiated
533 °C
~15 dpa

R. Klueh, 2007

Phénix :
Fuel Element Cladding irradiation

Swelling (%)

Displacement damage (dpa)

316L, Ti stabilized

FM steels (1.4914, EM10, EM12, F17)
Steels after Neutron irradiation

Austenitic steel 304

Y. De Carlan et al, ASTM STP, 2004

Ferritic-Martensitic steels

A. Alamo et al, EFDA-report 2006

Neutron irradiation below ~400°C: Increase strength, decrease ductility
Tensile properties after neutron irradiation (~15 dpa)

Ferritic/martensitic steels:
- Significant strength increase (irradiation hardening) for $T_{\text{irrad}} < 400 \, ^\circ\text{C}$
- This irrad. hardening comes mostly from interstitial type loops and $\alpha'$-precipitates.
  These loops and precipitates do not form above about 400-420 °C

**ODS EUROFER after Neutron irradiation:**

**Substantial Improvement of tensile properties**

![Graph showing tensile properties of Eurofer and ODS EUROFER](image)

- **RAFM Steel**
  - Eurofer 97 (FZK)
  - \( T_{\text{test}} = T_{\text{irr}} = 250°C \)
  - Irradiated, 15 dpa
  - Unirradiated

- **RAFM-ODS Steel**
  - Eurofer-ODS (FZK)
  - 0.5% Y\(_2\)O\(_3\)
  - \( T_{\text{test}} = T_{\text{irr}} = 250°C \)
  - Irradiated, 15 dpa
  - Unirradiated

- **Comments**
  - 😁 Substantial irradiation hardening
  - 🙂 Early strain localization due to dislocation channeling \( \rightarrow A_u \sim 0.3\% \)
  - 😝 Somewhat less irrad. hardening
  - 😏 Still work hardening \( \rightarrow \) almost no loss of uniform elongation \( A_u \sim 7\% \)
ODS EUROFER after Neutron irradiation:
Substantial Improvement of tensile properties in a wide temperature window

RAFM Steel

RAFM-ODS Steel

Eurofer 97
15 dpa, $T_{test} = T_{irr}$

ODS-Eurofer HIP
15 dpa, $T_{test} = T_{irr}$

R. Lindau, et al, KIT
Coherency properties of nano-dispersoids in steel matrix

- \((111)\text{Y}_2\text{O}_3 \parallel (110)\text{FeCr}\) - orientation of atomic planes, misfit only 0.5 %
- Coherence despite of the high melting temperature (\(\sim 2500\) °C) of \(\text{Y}_2\text{O}_3\)
TEM Bright field

EFTEM with HAADF Detector

+ Usually (not in EUROFER-ODS) Ar gas is used during mechanical alloying
+ Nano-dispersoids (Y$_2$O$_3$, Y$_2$Ti$_2$O$_7$) are very effective trapping centers for noble gases (→ suppression of He embrittlement)
Analytical TEM: nanoscaled iron based ODS-alloys

"Interface-Engineering": Nanoscaled ODS particles like $Y_2O_3$ or $Y_2Ti_2O_7$ are indispensable "Marketplaces" for defect recombination & trapping of diffusing alloy elements.
Neutron irradiation, “no” helium:
Irradiation induced yield strength seems to saturate

\[ \Delta R_{p0.2} / \text{(MPa)} \]

- \( T_{\text{irr}} = 300-335^\circ\text{C} \)
- \( T_{\text{test}} = 300-350^\circ\text{C} \)

<table>
<thead>
<tr>
<th>Material</th>
<th>Symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>EUROFER97</td>
<td>□</td>
</tr>
<tr>
<td>F82H</td>
<td>▲</td>
</tr>
</tbody>
</table>

“Orowan-type” fit (solid line):

\[
\Delta \sigma_{\text{irr}} = M \alpha \mu b \sqrt{Nd} \approx \Delta \sigma_{\text{sat}} \sqrt{1 - \exp\left(-\Theta / \Theta_0\right)}
\]
Is it possible to anneal irradiation embrittlement?

- How often can this recovery be repeated?
- What happens if large concentrations of He are present?
Conventional 12%CrNbMo steel

RAFM steels

\[ T_{irr} = 300-330°C \]
16 dpa Neutron irradiation, Helium effects after B-doping

Heat 806 - 82 ppm $^{\text{nat}}$B ($\sim$80 appm He)

Heat 826 - 83 ppm $^{10}$B ($\sim$415 appm He)

Un-irradiated Tests

Test Temperatures

$\leq$415 appm: He shows strength increase but no reduction of rupture strain

E. Materna-Morris, et al.
JNM 386(2009)422
Irradiation induced DBTT after 16 dpa HFR irradiation: Effect of irradiation temperature and He concentration

EUROFER97 HT, <10 appm He

- $T_{irr} = 250 \, ^\circ C$: $\Delta DBTT = 149 \, ^\circ C$

EUROFER-type, ~415 appm He

- $T_{irr} = 250 \, ^\circ C$: $\Delta DBTT = 370 \, ^\circ C$

E. Materna-Morris et al, SOFE-2009
Microstructure after neutron irradiation (~15 dpa)
Example: 9CrWTa steel, effect of dpa and 415 appm He

\[ T_{\text{test}} = T_{\text{irr}} = 300 \, ^\circ\text{C} \]

- Small loops & precipitates (dark dots)
- Small He bubbles (white dots)

\[ T_{\text{test}} = T_{\text{irr}} = 450 \, ^\circ\text{C} \]

- Small loops & precipitates not formed
- He bubbles along dislocation lines

E. Materna-Morris et al, 2008
Application window of RAF(M)-ODS-Steels - schematic -

- Irradiation enhanced creep without fatigue; prediction
- Irradiation embrittlement (~10 appmHe/dpa); constructed from experimental data (T_{irr}=250-300°C)
- Prediction for advanced alloys

RAFM (EUROFER)
RAFM steel (EUROFER)
RAF(M)-ODS steel

A. Möslang, unpublished
V alloys

Reference: V-4Cr-4Ti

Design Window

+ Irradiation embrittlement below 400-420 °C
+ Creep rupture strength comparable to state-of-the-art RAFM ODS steels
+ Coatings (like CaO, AlN) to prevent from MHD and embrittlement not yet available

S. Zinkle et al, ORNL
SiC\textsubscript{f}/SiC Composites for Irradiation Studies

S. Zinkle, L. Snead et al.

Reference in US/JAPAN collaboration: Chemical Vapor Infiltrated (CVI) SiC composites

Bend strengths of irradiated “3rd generation” composites show no degradation up to 10 dpa

Future work:
Advanced matrix infiltration R&D; joining; hermetic coatings; SiC/graphite composites
Summary
Application window of today’s materials (~10 dpa)

- W alloys
  - Mo (TZM)
  - Ta-8W-2Hf
  - Nb-1Zr-.1C
  - V-4Cr-4Ti

- ODS-RAF-steels
- RAFM-steel
  - 316 SS
  - Inconel 718
  - CuNiBe

- SiC_f/SiC

Temperature (°C)