Introduction to Radiation Damage in Metals

Journey from incoming particles

Most of the slides are borrowed from

22.14 Materials in Nuclear Engineering

22.74 Radiation Damage and Effects in Nuclear Materials

Was. means from Prof. Was’ text book:

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22.01: Intro to Nuclear Engineering and Ionizing Radiation
Radiation Damage

Radiation Damage Event (Monday)
the transfer of energy from an incident projectile to the solid and the resulting distribution of target atoms after completion of the event

Radiation Damage Effects (Wednesday)
Subsequent events involving the migration of the point defects and defect clusters and additional clustering or dissolution of the clusters
From Radiation Damage Event to Radiation Damage Effects

Unit Processes, Clustering

Short-Range Interactions

Long-Range Interactions

Transport

Component Response

Reactor components
New periodic structures
Defect superstructures
Defect pair interactions
Atomic level defects

Radiation Damage Event

MSS


Materials Intro.

- Crystalline vs. Amorphous: The difference is *long-range* order
Materials Intro.

Crystalline materials lattices

- Body-Centered Cubic (BCC) Structure
- Face Centered Cubic (FCC) Structure
- Hexagonal Close Packed (HCP) Structure

These images showing BCC structure, FCC structure, and HCP structure are reprinted/reused by permission from, © Iowa State University Center for Nondestructive Evaluation (CNDE).
Form Follows Structure

Pyrite (FeS₂), simple cubic (SC)

Gold (Au), face centered cubic (FCC)

Gypsum, monoclinic

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Point Defects (0D) – Vacancies

Vacancy in FCC lattice

Vacancy in BCC lattice
Point Defects (0D) – Interstitials

- Extra atoms shoved into the crystal lattice

Octahedron

Tetrahedron

Octahedral interstitial in BCC lattice

Tetrahedral interstitial in BCC lattice
Point Defects (0D) – Frenkel Pair

Frenkel Pair: an atom is displaced from its lattice position to an interstitial site, creating a vacancy at the original site and an interstitial defect at the new location.

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Dislocations (1D)

- Two types: Edge & Screw

- **Edge dislocation**: Extra half-plane of atoms shoved into the lattice

![Diagram of edge dislocation]

**Fig. 7.2.** An edge dislocation described as an extra half plane of atoms above the slip plane
Dislocations (1D)

- Two types: Edge & Screw

- Screw dislocation:

Fig. 7.3. (a) A screw dislocation formed by a cut and a shift of atoms in a direction parallel to the cut line. (b) A schematic showing the “parking ramp” nature of a screw dislocation in which atom planes spiral about the dislocation line.
Grain Boundaries (2D)

- Regions of different orientation

Tilt grain boundary in Al

Courtesy of Sandia National Lab.

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http://www.tf.uni-kiel.de/matwis/amat/def_en/kap_7/backbone/r7_2_1.html
Inclusions (3D)

From Prof. Short’s own image collection

• Other phases trapped within base material

• Examples:
  • Secondary particle precipitates in Zircaloys
  • Carbides in steels
  • Y$_2$O$_3$ particles in Oxide Dispersion Strengthened (ODS) steels

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Summary of Material Intro.

- Crystalline Solids
- 0D Defects
  - Vacancies & Interstitials
- 1D Defects (Dislocations)
- 2D & 3D Defects

We are interested in how many defects (Frenkel pairs & defect clusters) are produced by incoming particles.
The result of a radiation damage event is the creation of a collection of point defects (vacancies and interstitials) and clusters of these defects in the crystal lattice. ($10^{-11}$ s)

This can be described by how many displacements of atoms (DPAs) are created by incoming particles
• Define a rate of atomic displacements using flux:

\[
R = \int_{0}^{E_{\text{max}}} N \times \Phi(E_i) \times \sigma_D(E_i) \, dE_i
\]

- Maximum energy available
- Energy dependent flux distribution
- Reaction rate \( \frac{\text{displ.}}{m^3 \text{-sec}} \)
- Material number density \( \frac{\text{atoms}}{m^3} \)
- Displacement cross section
Define a rate of atomic displacements using flux:

\[ R = \int_0^{E_{max}} N \times \Phi(E_i) \times \sigma_D(E_i) \, dE_i \]

\[ \frac{R}{N} = \frac{DPA}{sec} = \int_0^{E_{max}} \Phi(E_i) \times \sigma_D(E_i) \, dE_i \]
Displacement Theory

• Define a rate of atomic displacements using flux:

\[
\frac{DPA}{sec} = \int_0^{E_{\text{max}}} \Phi(E_i) \ast \sigma_D(E_i) dE_i
\]

Probability that an atom displaced by a particle with energy \(E_i\) leaves with recoil energy \(T\) (differential energy transfer cross section)

\[
\sigma_D(E_i) = \int_{T_{\min}}^{T_{\text{max}}} \sigma(E_i, T) \nu(T) dT
\]

• \(T\) is the PKA (displaced atom) recoil energy

Number of atomic displacements from a PKA with energy \(T\)
Displacement Theory

• Define a rate of atomic displacements using flux:

\[
\frac{DPA}{sec} = \int_0^{E_{max}} \int_{T_{min}}^{T_{max}} \Phi(E_i) \ast \sigma(E_i, T) \nu(T) dT dE_i
\]

Probability that an atom displaced by a particle with energy \(E_i\) leaves with recoil energy \(T\) (differential energy transfer cross section)

Number of atomic displacements from a PKA with energy \(T\)
Journey of an incoming ion

\[ (E_i) \]

Total path length \( R \) and projected range \( R_p \) for an ion incident on a target
What happened until it stops

Explanation on board

\((\sigma_n, \sigma_e, T_n, T_e)\)
Journey of multiple incoming ions

\((f_{E_i})\) \rightarrow \text{Continuous description of a large quantity discrete events}

\downarrow \text{Stopping power}
Coulombic/Nuclear Stopping Power

• *Stopping Power* is defined as differential energy loss as a function of energy:

\[ N \ast S(E) = -\frac{\partial E}{\partial x} \]

• Separable components due to nuclear (screened nucleus Coulombic), electronic, and radiative terms:

\[ N \ast S(E) = -\left(\frac{\partial E}{\partial x}\right)_{\text{nucl.}} - \left(\frac{\partial E}{\partial x}\right)_{\text{elec.}} - \left(\frac{\partial E}{\partial x}\right)_{\text{rad.}} \]
Relative Stopping Powers
Range

- Integrate inverse of stopping power over the energy range of the particle: \( \text{Range} = \int_0^{E_{\text{max}}} \frac{1}{S(E)} \, dE \)
- Not all particles have identical range, \textit{straggling} describes this variation
Range of the incoming ions

(a) High-energy ion

(b) Low-energy ion

The total path length $R$ is:

$$R = \sum (\lambda_1 + \lambda_2 + \lambda_3 + \ldots etc.)$$
journey continues by PKAs

PKAs → How many atoms will be displaced by a PKA of energy $T$, $v(T)$

Kinchin and Pease Model (K-P Model)
Kinchin-Pease Model

- Final formulation:

\[ \nu(T) = \begin{cases} 
0 & \text{for } T < E_d \\
1 & \text{for } E_d < T < 2E_d \\
\frac{T}{2E_d} & \text{for } 2E_d < T < E_c \\
\frac{E_c}{2E_d} & \text{for } T \geq E_c 
\end{cases} \]
Assumptions made by K-P model

1. The cascade is created by a sequence of two-body elastic collisions between atoms.
2. The displacement probability is 1 for $T \geq E_d$
3. No energy passes to the lattice during the collision process
4. For all energies less than cutoff energy $E_c$, electronic stopping is ignored
5. The energy transfer cross section is given by the hard sphere model
6. Effects due to crystal structure are neglected
### Table 2.4. Modifications to the displacement cross section

<table>
<thead>
<tr>
<th>Assumption</th>
<th>Correction to $\nu(T)$</th>
<th>Equation in text</th>
</tr>
</thead>
<tbody>
<tr>
<td>3: Loss of $E_d$</td>
<td>$0.56 \left(1 + \frac{T}{2E_d}\right)$</td>
<td>Eq. (2.31)</td>
</tr>
<tr>
<td>4: Electronic energy loss cut-off</td>
<td>$\xi(T) \left(\frac{T}{2E_d}\right)$</td>
<td>Eq. (2.50)</td>
</tr>
<tr>
<td>5: Realistic energy transfer cross section</td>
<td>$C \frac{T}{2E_d}$, $0.52 &lt; C \leq 1.22$</td>
<td>Eqs. (2.33), (2.39)</td>
</tr>
<tr>
<td>6: Crystallinity</td>
<td>$\frac{1-P}{1-2P} \left(\frac{T}{2E_d}\right)^{(1-2P)} - \frac{P}{1-2P}$</td>
<td>Eq. (2.104)</td>
</tr>
<tr>
<td></td>
<td>$\sim \left(\frac{T}{2E_d}\right)^{(1-2P)}$</td>
<td>Eq. (2.105)</td>
</tr>
</tbody>
</table>
The Real $\sigma_D$ Is Ugly!

$$\sigma_D = \frac{\sigma_s(E_i)}{\gamma E_i} \int_{E_d}^{\gamma E_i} \left[ \frac{1 - P}{1 - 2P} \left( C' \xi(T) \frac{T}{2E_d} \right)^{(1-2P)} - \frac{P}{1 - 2P} \right]$$

$$\times \left[ 1 + a_1(E_i) \left( 1 - \frac{2T}{\gamma E_i} \right) \right] dT$$

$$+ \sum_j \frac{\sigma_{sj}(E_i, Q_j)}{\gamma E_i} \left[ 1 + \frac{Q_j}{E_i} \left( \frac{1+A}{A} \right) \right]^{-1/2}$$

$$\int_{T_j}^{\hat{T}_j} \left[ \frac{1 - P}{1 - 2P} \left( C' \xi(T) \frac{T}{2E_d} \right)^{(1-2P)} - \frac{P}{1 - 2P} \right] dT$$

$$+ \int_{E_i-E_m}^{E_i} \sigma_{(n,2n)}(E_i, T) \left[ \frac{1 - P}{1 - 2P} \left( C' \xi(T) \frac{T}{2E_d} \right)^{(1-2P)} - \frac{P}{1 - 2P} \right] dT$$

$$+ \sigma_\gamma \left[ \frac{1 - P}{1 - 2P} \left( C' \xi(T) \frac{E_\gamma^2}{8E_d(A+1)c^2} \right)^{1-2P} - \frac{P}{1 - 2P} \right]. \quad (2.117)$$
**Example $\sigma_D$**

Fig. 2.18 The displacement cross section for stainless steel based on a Lindhard model and ENDF/B scattering cross sections (after [21])
Displacement Theory

• Define a rate of atomic displacements using flux:

\[
\frac{DPA}{sec} = \text{Does the number of displacements per atom per second we calculated this way equal with the stable defects (Frenkel pairs & defect clusters) introduced by incoming particles?}
\]
Cascade Stages – Ballistics

- PKA initiates a cascade of displacive collisions
- Ends until no atom contains enough energy to create further displacements

Cascade Stages – Thermal Spike

• Collisional energy of the displaced atoms is shared among their neighboring atoms

• Temperature rises very locally for a very short time

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Cascade Stages – Quench

- Heat is conducted away EXTREMELY quickly
- Stable lattice defects form either as point defects or defect clusters

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Radiation Damage Event

• Radiation damage event completes after the quench stage

• We define the displacement efficiency $\xi$, as the fraction of the “ballistically” produced displacements that survive the cascade quench

$$\xi \ast \frac{DPA}{sec} = \xi \ast \int_0^{E_{max}} \int_{T_{min}}^{T_{max}} \Phi(E_i) \ast \sigma(E_i, T)\nu(T)dTdE_i$$
3.1 Molecular Dynamics (MD) simulation of the development of a cascade from a 200 keV recoil in iron at 100K.

Note the extension of the cascade to the lower right hand corner of the screen. This lobe likely results from channeling of a knock-on atom. Also note the striking difference in defect density between the peak of the ballistic regime (~0.7 ps) and that at the end of the quench (~21.6 ps). (courtesy, R. Stoller, Oak Ridge National Laboratory)
• Different radiation produces different cascades

Mass & Charge

Increasing mass, same charge

Moderate mass, no charge

Stopping Mechanism

Almost All electronic

Mostly electronic

Nuclear and electronic

Entirely nuclear

Fig. 3.7 Difference in damage morphology, displacement efficiency, and average recoil energy for 1 MeV particles of different types incident on nickel (after [6])
Radiation damage effects

Physical effects
- RIS (Radiation-induced segregation)
- Nucleation and growth of dislocation loops and voids
- Phase Stability

Mechanical effects: when stress is applied
- Irradiation hardening and deformation
- Fracture and Embrittlement
- Irradiation Creep and Growth
- IRSCC (irradiation-assisted stress corrosion cracking)
Building Up to Radiation Effects

We have introduced


Wednesday

Unit Processes, Clustering

Short-Range Interactions

Transport

Long-Range Interactions

Component Response

We have introduced

MSS

Reactors

New periodic structures

Defect superstructures

Defect pair interactions

Atomic level defects


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Radiation Damage Event: from incoming particles to point defects and defect clusters

\[ \xi \int_{E_{\text{max}}} E_i \int_{T_{\text{min}}}^{T_{\text{max}}} \Phi(E_i) \sigma(E_i, T) \nu(T) dT dE_i \]

Incoming particle \( \Phi(E_i) \) \hspace{1cm} PKA with energy \( T \) \( \sigma(E_i, T) \)

200 keV cascade in Fe, 100K

\[ \nu(T) \cdot \xi \]

 Courtesy of Oak Ridge National Laboratory, U.S. Dept. of Energy.
Damage After the Cascade

What happens to damage after the cascade?

- Production
- Recombination (One interstitial finds one vacancy and they annihilate)
- Absorption at sinks (migrate to the sink and get trapped)
- Migration (keep moving)
Point Defect Balance

Change = Gain – Loss (where have we used this format before?)

- What are the possible gain terms?
  - Displacement production

- What are the possible loss terms?
  - Recombination
  - Loss to sinks
  - Diffusion
Point Defect Balance

Change = Gain − Loss

• What are the possible sinks?
  • Grain boundaries
  • Dislocations
  • Impurities
  • Free surfaces
  • Incoherent precipitates

For point defects, sinks are higher dimensional defects
Cluster Dynamics

• Equations that govern the growth and shrinkage of clusters via defect emission and absorption

• Same way to use equation to describe it:

\[
\frac{\partial \text{Cluster}}{\partial t} = \text{Gains} - \text{Losses}
\]
Cluster Dynamics (for Vacancies)

\[ \frac{\partial n_j}{\partial t} = Gains - Losses \]

- What are the gain terms?
  - Direct production
  - Absorption of same type defects & clusters from smaller clusters
  - Emission of same type defects & clusters from larger clusters
  - Absorption of different types of defects & clusters from larger clusters
Cluster Dynamics (for Vacancies)

\[ \frac{\partial n_j}{\partial t} = Gains - Losses \]

- What are the loss terms?
  - Direct destruction (loop collapse, etc.)
  - Absorption of same type defects & clusters from size \( j \)
  - Emission of same type defects & clusters from size \( j \)
  - Absorption of different types of defects & clusters from size \( j \)
Bridging radiation damage event to radiation damage effects

http://www-personal.umich.edu/~gsw/movies.html

point defect kinetics

Change = Gain – Loss

defect cluster dynamics

Courtesy of Oak Ridge National Laboratory, U.S. Dept. of Energy.

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Bridging radiation damage event to radiation damage effects

point defect kinetics

\[ \text{Change} = \text{Gain} - \text{Loss} \]

defect cluster dynamics

Population and distribution of point defects and defect clusters
**Radiation damage effects**

Population and distribution of point defects and defect clusters

**Physical effects**
- RIS (Radiation-induced segregation)
- Nucleation and growth of dislocation loops and voids
- Phase Stability

**Mechanical effects: when stress is applied**
- Irradiation hardening and deformation
- Fracture and Embrittlement
- Irradiation Creep and Growth
- IRSCC (irradiation-assisted stress corrosion cracking)
RIS (Radiation induced segregation)

**RIS**: the spatial redistribution of solute and impurity elements in the metal at elevated temperature under irradiation

**Influence**: changes in the local properties of the solid, which may induce susceptibility to a host of processes that can degrade the integrity of the component.

RIS was discovered around 1970s

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*Fig. 6.1*  Radiation-induced segregation of Cr, Ni, Si, and P at the grain boundary of a 300 series stainless steel irradiated in a light water reactor core to several dpa at ~300 °C (after [1])

*Was, p. 255*
Mechanism of RIS

Excess point defects produced from radiation

Defect fluxes to defect sinks (GB)

Different atoms couple differently to the defect fluxes

Was, p. 256
RIS Effect on Material Properties

- Corrosion
  - Cr depletion
- Embrittlement, Fracture Toughness Reduction
  - P, S segregation to microstructural sinks
- Hardening, Strengthening
  - Precipitate formation by smaller vacancy solutes (Mo, Cu, Si) towards sinks
  - Precipitate formation by interstitial solutes (C, N) towards sinks
Nucleation and growth of dislocation loops and voids

Defect cluster dynamics (size or number of defects)

Nucleation

Dislocation loops and voids
(Configuration of defect clusters)

Point defect

Defect clusters

Population and distribution

Growth
Example dislocation loops

**Fig. 7.66** Images of large Frank loops in (a) a 300 series stainless steel irradiated at 500 °C to a dose of 10 dpa (from [42]), and (b)–(e) in irradiated aluminum, copper, nickel, and iron, respectively (after [18])

Was, p. 370
Example voids

Fig. 8.1 Micrographs of irradiation-induced voids in (a) stainless steel, (b) aluminum, (c) and (d) magnesium [2, 3]

Was, p. 380
Voids lead to Swelling (dimensional change)

Figure 4.1: Photograph of 20% cold-worked 316 stainless steel rods before (left) and after (right) irradiation at 533°C to a fluence of 1.5×10^{23} neutrons m^{-2} in the EBR-11 reactor (Mansur, 1994).

Swelling is a big issue

- Spiral distortion of 316-clad fuel pins induced by swelling and irradiation creep in an FFTF fuel assembly where the wire wrap swells less than the cladding.

- Reproduced from Makenas, B. J.; Chastain, S. A.; Gneiting, B. C. In Proceedings of LMR: A Decade of LMR Progress and Promise; ANS: La Grange Park, IL, 1990; pp 176–183; (middle) Swelling-induced changes in length of fuel pins of the same assembly in response to gradients in dose rate, temperature, and production lot variations as observed at the top of the fuel pin bundle. Reproduced from Makenas, B. J.; Chastain, S. A.; Gneiting, B. C. In Proceedings of LMR: A Decade of LMR Progress and Promise; ANS: La Grange Park, IL, 1990; pp 176–183; (bottom) swelling-induced distortion of a BN-600 fuel assembly and an individual pin where the wire swells more than the cladding. Reproduced from Astashov, S. E.; Kozmanov, E. A.; Ogorodov, A. N.; Roslyakov, V. F.; Chuev, V. V.; Sheinkman, A. G. In Studies of the Structural Materials in the Core Components of Fast Sodium Reactors; Russian Academy of Science: Urals Branch, Ekaterinburg, 1984; pp 48–84, in Russian.
Phase instability

Local enrichment or depletion of solute atoms

Formation or dissolution of phases

Extreme case

Amorphization
Radiation damage effects

Population and distribution of point defects and defect clusters

Physical effects
- RIS (Radiation-induced segregation)
- Nucleation and growth of dislocation loops and voids
- Phase Stability

Mechanical effects: when stress is applied
- Irradiation hardening and deformation
- Fracture and Embrittlement
- Irradiation Creep and Growth
- IRSCC (irradiation-assisted stress corrosion cracking)
Irradiation Assisted Stress Corrosion Cracking (IASCC)

- Stress corrosion cracking requires:
  - Tensile stress
  - Susceptible material
  - Aggressive environment

- Radiation can:
  - Increase susceptibility
  - Generate stresses
  - Induce hydrolysis, free radicals, more corrosion

Schematic illustrating mechanistic issues believed to influence crack advance during IASCC of austenitic stainless steels

Two big problems here: (1) The bolts can break, loosening the baffle. (2) The bolt heads get swept up by the coolant, becoming foreign material.
General challenges

- Quantify radiation damage
  - Problems using DPA
    - DPA is calculated, not measurable
    - Too many steps from DPA to the effects
    - Difficult to compare different conditions
  - What is a good unit?
    - Not that general (not too far away from effects)
    - Not that specific (should not only for one specific effect)
    - Can be calculated
    - Can be measured
General challenges

- Simulation limitations

Fig. 3.13 Time and length scale for radiation damage evolution and the corresponding simulation methodologies (courtesy F. Gao)

Was, p. 149
General challenges

- Experimental constrains
  - Limitations for experiments
    - Time
    - Cost
    - Safety
    - Sample examination
  - What is a good experimental design
    - Not that simplified (not too different with application conditions)
    - Not that complex (not involving too many variables)