Overview of Radiation Materials Science and Radiation Damage Modeling

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PNNL-SA-191704

Outline

- Radiation damage
  - Point defects
  - Displacement cascade
  - Defect annihilation, clustering and mobility
- Radiation damage microstructures
  - Radiation induced segregation
  - Dislocations
  - Voids and bubbles
  - Phase stability
- Macroscopic effects of radiation damage
  - Irradiation hardening and embrittlement
  - Radiation growth and swelling
  - Irradiation creep
  - Irradiation assisted stress corrosion cracking
  - Transport property degradation
- Radiation damage modeling
  - Atomic Level Simulations
  - Microstructural Modeling
  - Machine Learning to Integrate Experiments and Modeling

Radiation Damage

Radiation Damage: Energetic Damage Processes

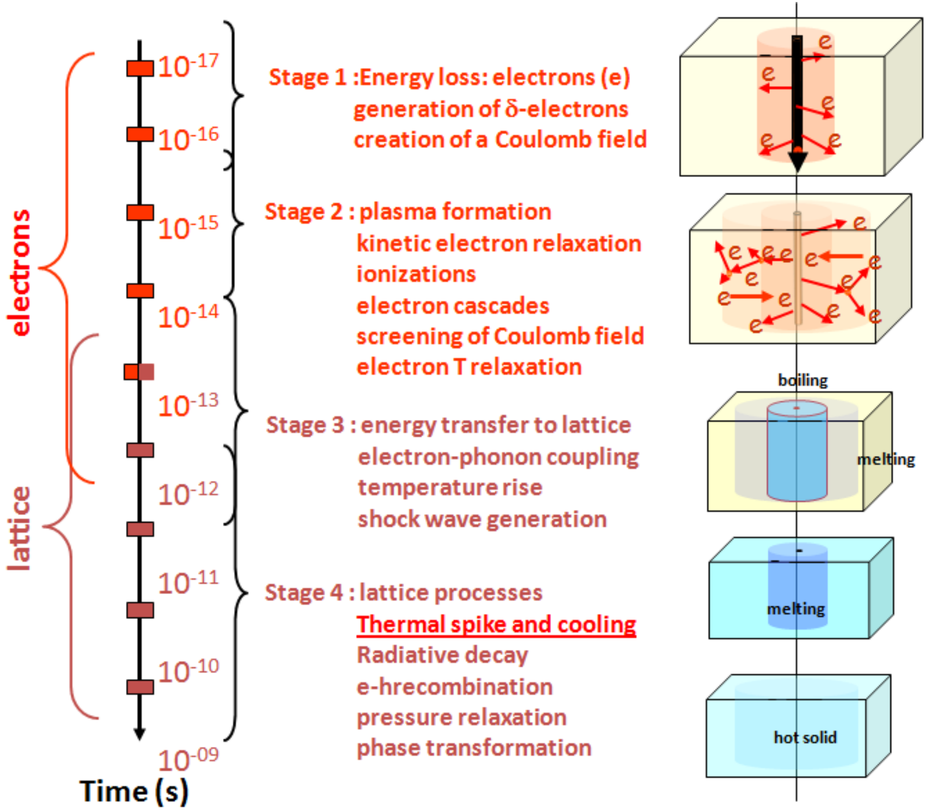
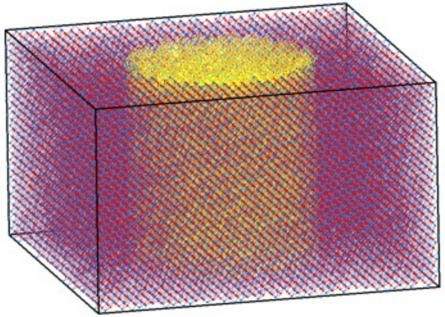


Image courtesy of Marcel Toulemonde, GANIL

Relevant problems:
Fission and fusion reactor materials
Swift heavy ions (accelerators)

Possibilities:
- Synthesize new phases
- Study systems far from equilibrium
- Develop radiation tolerant materials



Computational model of a thermal spike

Point Defects

- Fundamentally, all radiation damage effects are caused by creation of point defects
  - Interstitial defects (displaced, implanted, or transmuted atom)
  - Substitutional defects (an atom occupying a lattice position of a different type of atom)
  - Vacancies (missing atom)
  - An interstitial-vacancy pair are called a “Frenkel pair”
- Point defects diffuse to form larger radiation damage microstructures
  - Frenkel pair annihilation
  - Interstitial clusters
  - Dislocations and dislocation loops
  - Voids and bubbles
- The degree of radiation damage in a material is typically expressed in “displacements per atom” or dpa
  - dpa is not a physical phenomenon that can be measured, but is intended to be a relative basis for comparison of different radiation types and different materials
  - Multiple approaches exist to calculate dpa – most use some variant of the NRT method

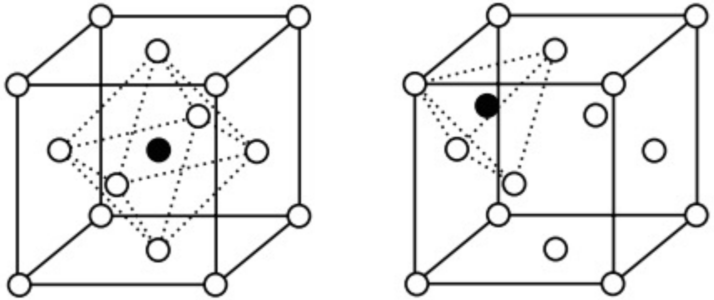


Fig. 4.2. Interstitial positions in the fcc unit cell

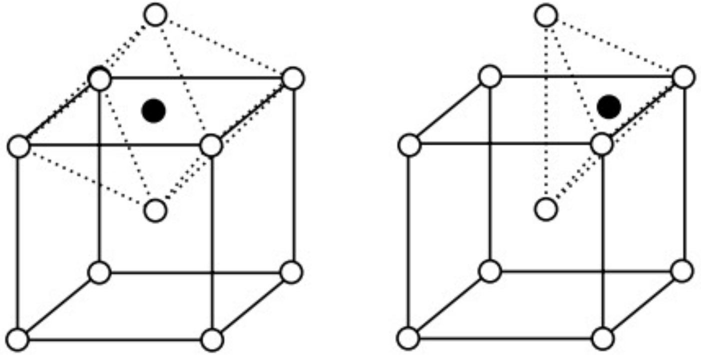


Fig. 4.4. Interstitial positions in the bcc unit cell

Was. 2007. *FRMS*, Springer

Displacement Cascades

- Point defects are created when an incident particle or displaced atom interacts with an atom on a lattice site
  - The chain of atomic displacement events is called a displacement cascade
  - The nature of the displacement cascade is a function of incident particle type and energy

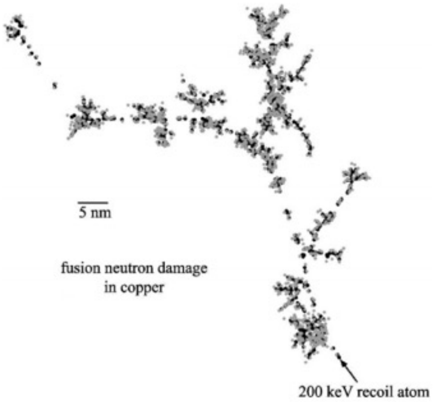


Fig. 3.8. Collisional stage of a 200keV cascade in copper simulated using the binary collision approximation in MARLOWE (after [8])

Heinisch. 1996. *JOM*, 48:38

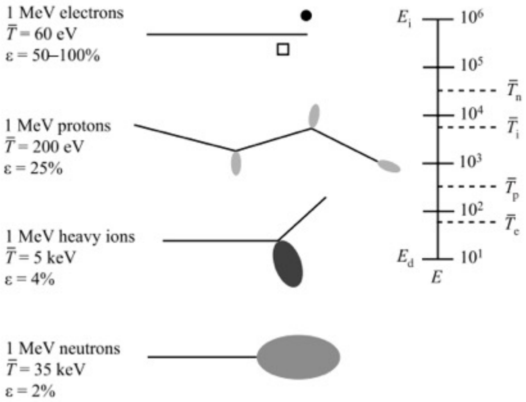


Fig. 3.7. Difference in damage morphology, displacement efficiency and average recoil energy for 1 MeV particles of different type incident on nickel (after [6])

Was and Allen. 1994. *Mater. Char.*, 32:239

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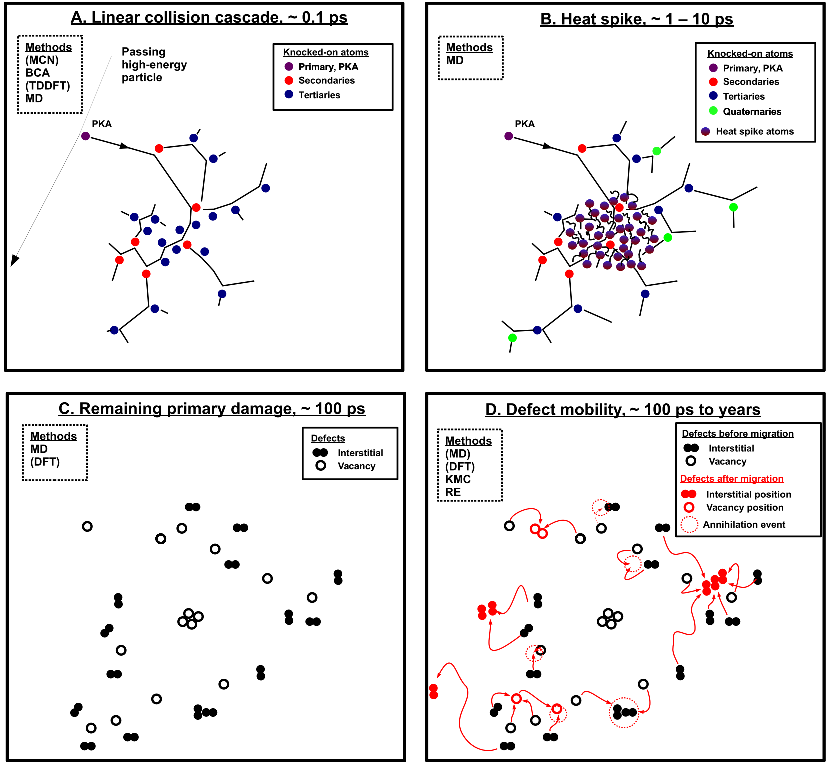
Defect Annihilation, Clustering and Mobility

- Most point defects created during the displacement cascade will annihilate quickly (picoseconds)
- The remaining defects will migrate and potentially interact over longer time periods (nanoseconds to years)
- Defect mobility is a function of temperature, dpa, and crystal lattice characteristics



Nordlund. 2019. *J. Nucl. Mater.*, 520:273

Radiation Damage Microstructures

Radiation Induced Segregation

- The mixing caused by atomic displacements coupled with energy from temperature (thermal diffusion) and radiation damage (irradiation-enhanced diffusion) can cause elemental segregation
- Preferential diffusion will lead to selective concentration or depletion at defect sinks like surfaces, grain boundaries, dislocations, and voids

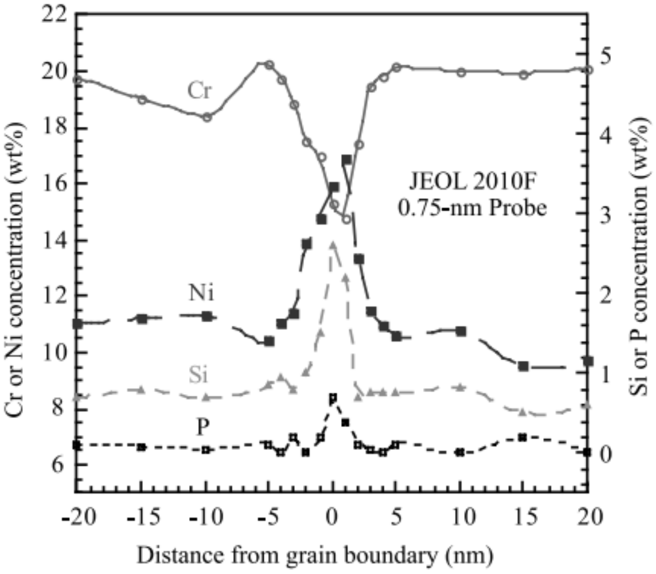


Fig. 6.1. Radiation-induced segregation of Cr, Ni, Si and P at the grain boundary of a 300 series stainless steel irradiated in a light water reactor core to several dpa at ~300 °C (after [1])

Bruemmer et al. 1999. *J. Nucl. Mater.*, 274:299

Dislocations

- Displaced atoms can diffuse to form “extra” or “distorted” planes in a crystalline lattice
  - Small clusters (<2 nm) of atoms will appear in TEM images as black spots
  - Larger clusters will appear in TEM images as loops

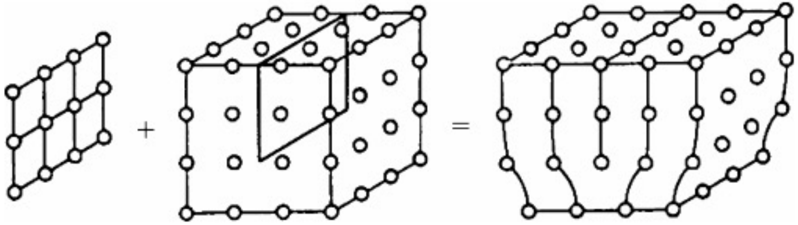
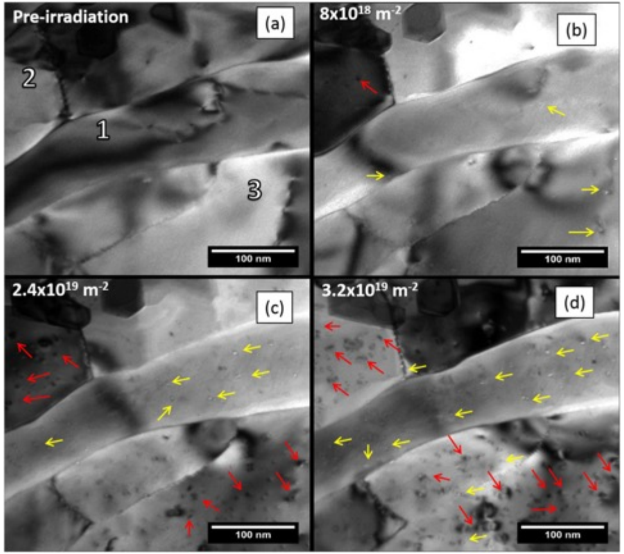


Fig. 7.2. An edge dislocation described as an extra half plane of atoms above the slip plane

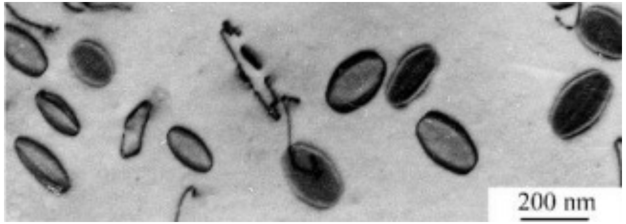
Was. 2007. *FRMS*, Springer

W irradiated at 950ºC



El-Atwani et al. 2014. *Scientific Reports*, 4:4716

Ni irradiated at 500ºC

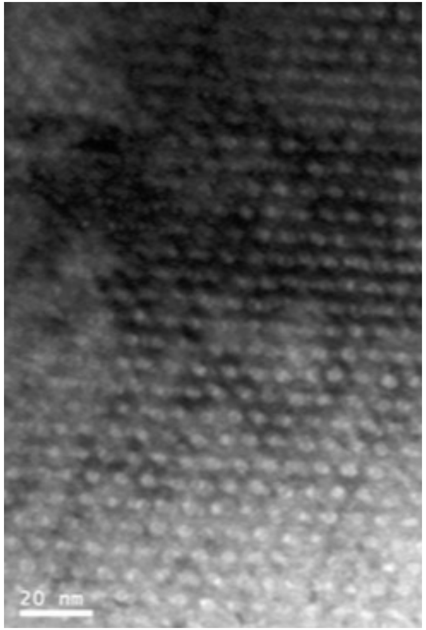


Kiritani. 1994. *J. Nucl. Mater.*, 216:200

Voids and Bubbles

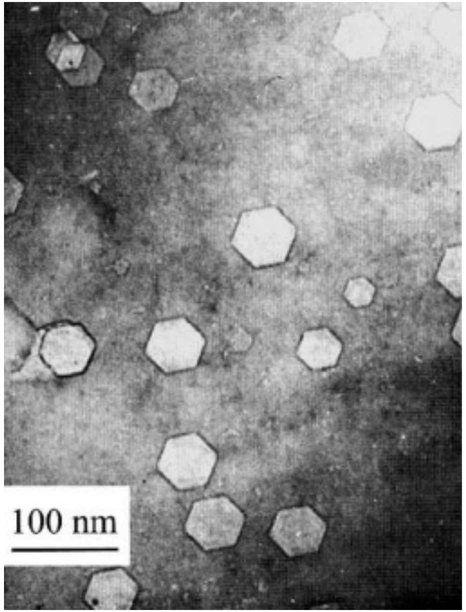
- Vacancies typically become mobile at higher temperatures than interstitials
  - A cluster of vacancies creates a void
- Bubbles are clusters of atoms in the gas phase
  - Transmutation during irradiation (e.g., He in stainless steel)
  - Implantation during ion irradiation (e.g., H in proton accelerator)
  - Fission products in nuclear fuel (e.g., Xe, Kr)

Fission gas bubble superlattice in U-7Mo



Gan et al. 2009. *RRFM*

Voids in irradiated Al



Jenkins and Kirk. 2001. *Characterization of Radiation Damage by TEM*, IOP

Phase Stability

- A consequence of elemental concentration or depletion at defect sinks is the precipitation of distinct phases
- The energy associated with irradiation can cause phase changes that would not be expected based on temperature
- Below a threshold temperature and dose (dpa), irradiation will cause crystalline materials to become amorphous

Rehn. 1982. *Metastable Materials Formation by Ion Implantation*, Elsevier

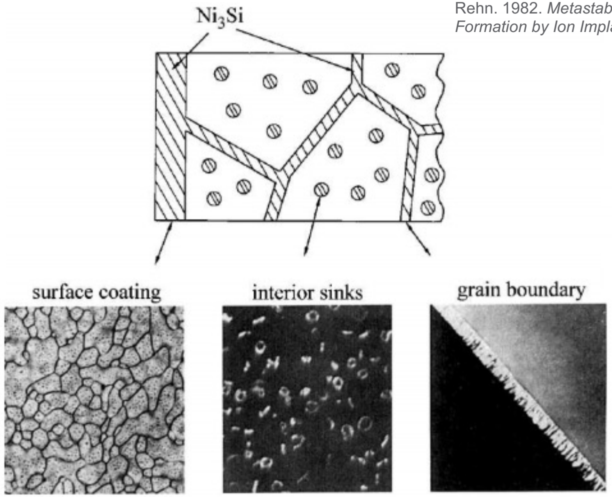
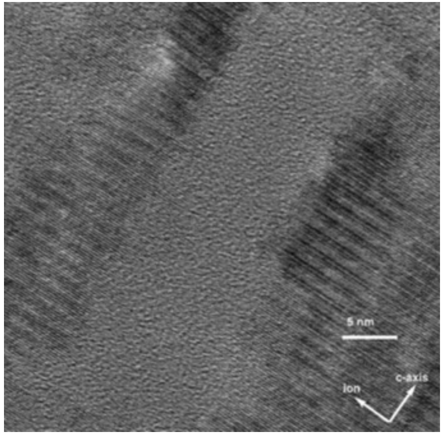


Fig. 9.2. Formation of γ'-Ni_3Si on defect sinks in a solid solution of Ni–6at% Si alloy showing (a) a surface coating of Ni_3Si, (b) toroidal γ' precipitates at dislocation loops and (c) a grain boundary coated with Ni_3Si (from [1])

Si ion-irradiated SiC showing onset of amorphization

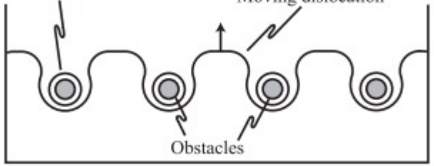


Snead et al. 1998. *Nucl. Instr. Meth. Phys. Res. B*, 141:123

Macroscopic Effects of Radiation Damage

Irradiation Hardening and Embrittlement

- Dislocation movement allows materials to deform under stress
- Impurities and irradiation-induced defects decrease dislocation mobility
  - Increased strength
  - Decreased ductility
- In the extreme (brittle fracture) the same mechanisms decrease fracture toughness and increase ductile-to-brittle transition temperature (DBTT)



Was. 2007. *FRMS*, Springer

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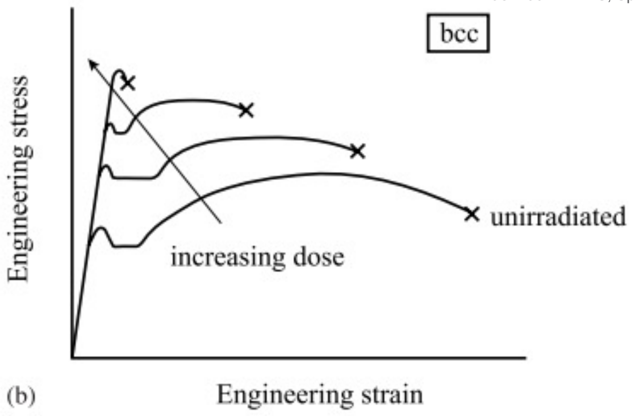


Steele. 1975. *Neutron Irradiation Embrittlement of RPV Steels*, IAEA

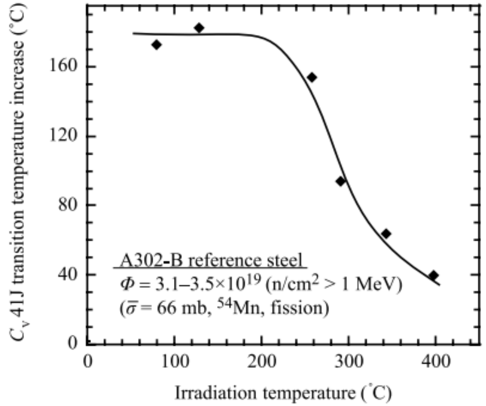
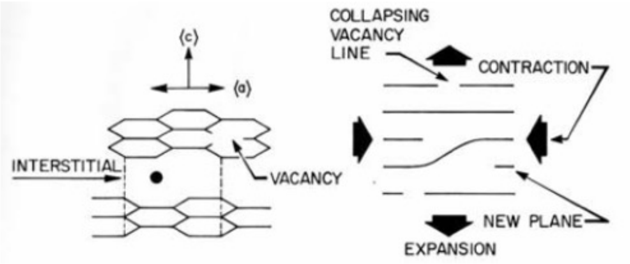


Fig. 13.25. Effect of irradiation temperature on transition temperature increase for an A302-B steel (from [19])

Irradiation Growth and Swelling

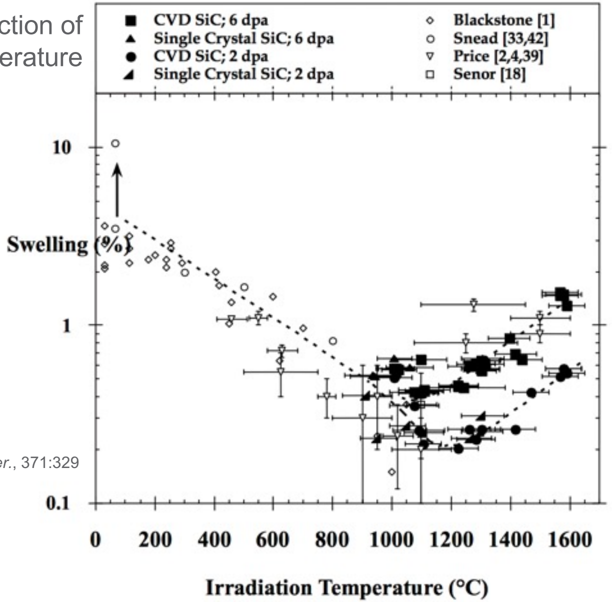
- Irradiation-induced growth and swelling can be caused by several mechanisms
  - In highly oriented lattices (e.g., HCP) Insertion of "extra" interstitial planes causes growth in one direction and shrinkage in the other (irradiation growth)
  - In more isotropic lattices, interstitial defects cause uniform distortion – the equilibrium concentration of defects varies with irradiation temperature
  - At high temperatures – vacancy diffusion and clustering causes void swelling
  - At high dpa or after H/He irradiation – gas atom diffusion and clustering causes bubble swelling
  - In nuclear fuel at high burnup – gaseous fission products cause bubble swelling

Irradiation Growth in Graphite



Burchell. 1999. *Carbon Materials for Advanced Technologies*

Swelling in SiC as a Function of Irradiation Temperature

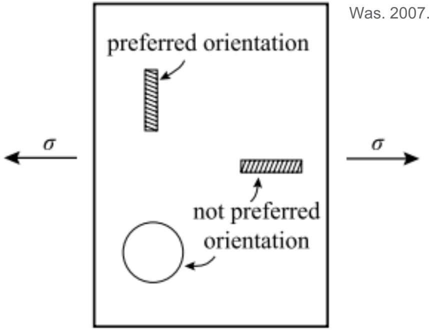


Snead et al. 2007. *J. Nucl. Mater.*, 371:329

Irradiation Creep

- Irradiation-induced defects increase creep rate relative to thermal creep at the same temperature
  - Mechanisms similar to those responsible for irradiation growth
  - Applied stress causes dislocation loops to form and grow perpendicular to the stress, resulting in an elongation in the direction of the stress
  - Absorption of irradiation-induced defects also enhances the ability of dislocation loops to release from obstacles by climb
  - Irradiation does not enhance diffusional creep because equilibrium concentration of defects is constant at a given temperature

Was. 2007. *FRMS*, Springer



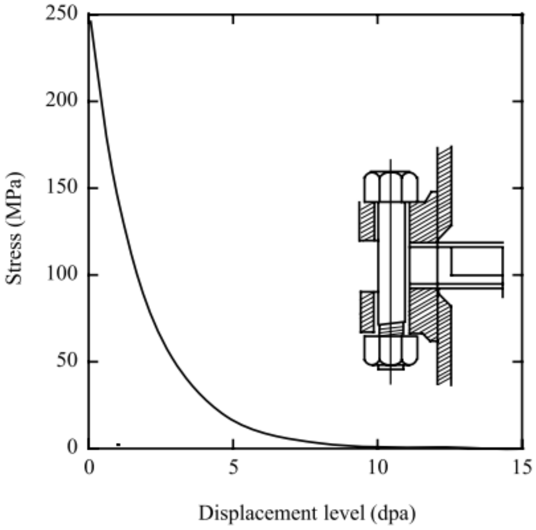


Fig. 15.41. Irradiation-induced creep causing stress relief in a bolt that was originally torqued to 250MPa and then exposed to a neutron flux (from [47])

Grossbeck and Mansur. 1991. *J. Nucl. Mater.*, 179-181:130

Irradiation Assisted Stress Corrosion Cracking

- Components exposed to an electrolyte can have increased sensitivity to stress corrosion cracking in an irradiation environment - caused by several factors
  - Radiolysis of the electrolyte (e.g., H^+ and OH^- in a LWR)
  - Irradiation hardening and embrittlement
  - Radiation induced segregation (e.g., to grain boundaries) of susceptible elements or phases

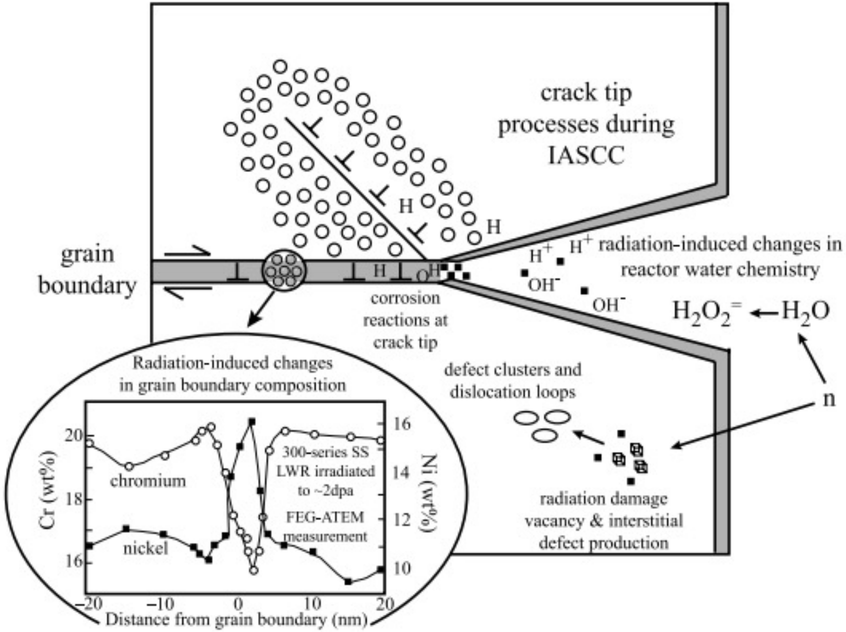


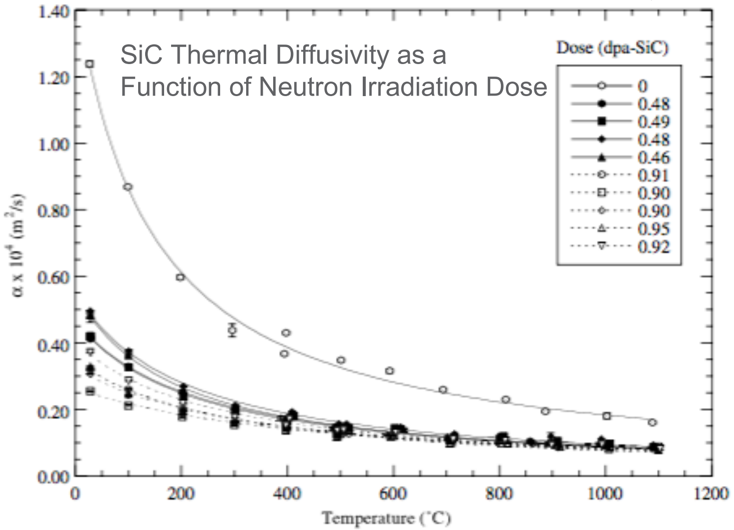
Fig. 15.3. Schematic illustration of mechanistic issues believed to influence crack advance during IASCC of austenitic stainless steels in LWRs (from [6])

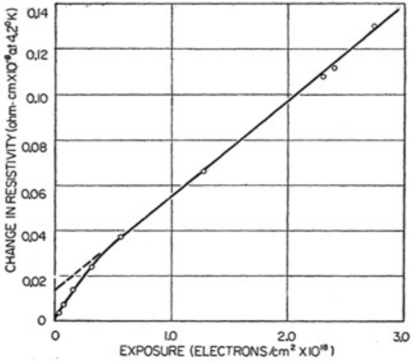
Bruemmer et al. 1999. *J. Nucl. Mater.*, 274:299

Transport Property Degradation

- Irradiation damage degrades thermal diffusivity and conductivity
  - In ceramics and other phonon conductors this is due to isolated point defects (especially vacancies)
  - In metals and other electronic conductors this is due to creation of impurities (transmutation) and voids at high temperatures
- Electrical resistivity is degraded by the same mechanisms

Senor et al. 2003. *J. Nucl. Mater.*, 317:145





Meechan and Brinkman. 1956. *Phys. Rev.*, 103(5):1194

FIG. 2. Electrical resistivity changes in copper during a 1.25-Mev electron irradiation at 80°K.

Radiation Damage Modeling

Radiation Damage is Modeled at Different Scales

Long term degradation of properties (mechanical, thermal, electrical …)

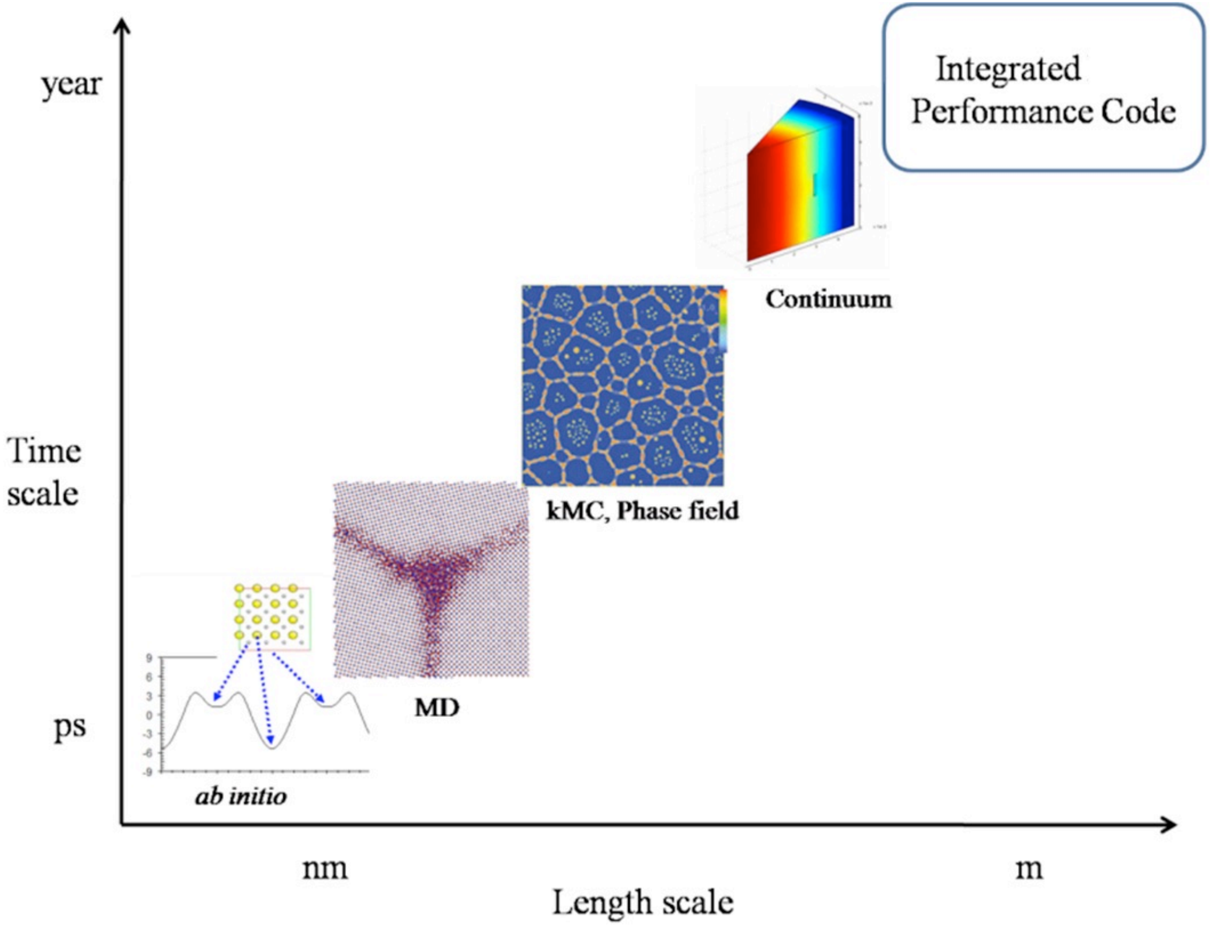
Microstructure changes, phase transformation, precipitates, gas bubbles

Radiation enhanced diffusion, segregation, defect-gas clusters

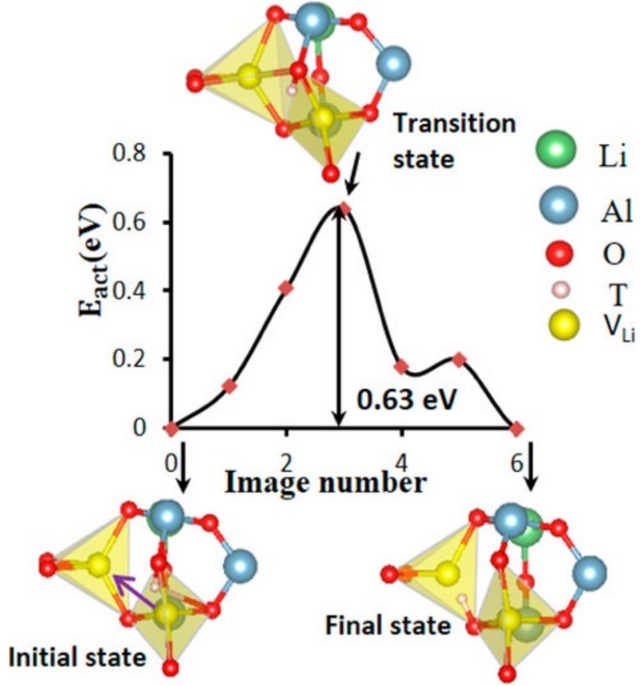
Dynamic annealing & solute redistribution

Displacement cascades and thermal spikes

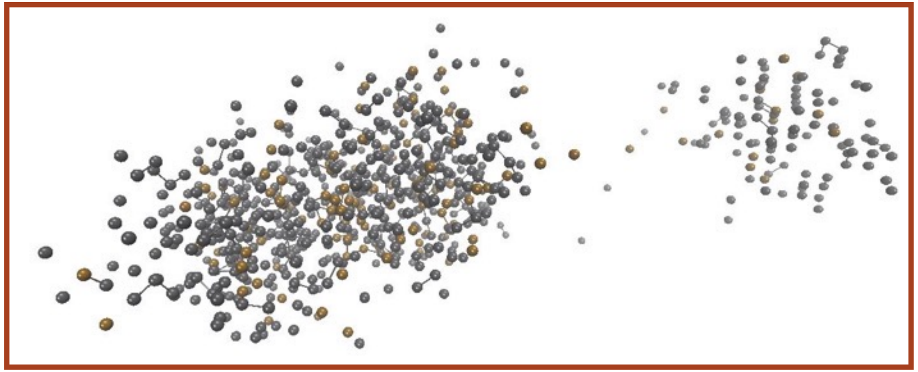
Electronic excitation



Atomic Level Simulations (DFT and MD)



DFT study of tritium migration in γ-$LiAlO_2$
Paudel et al, J. Phys. Chem. C 2018, 122, 18, 9755–9765

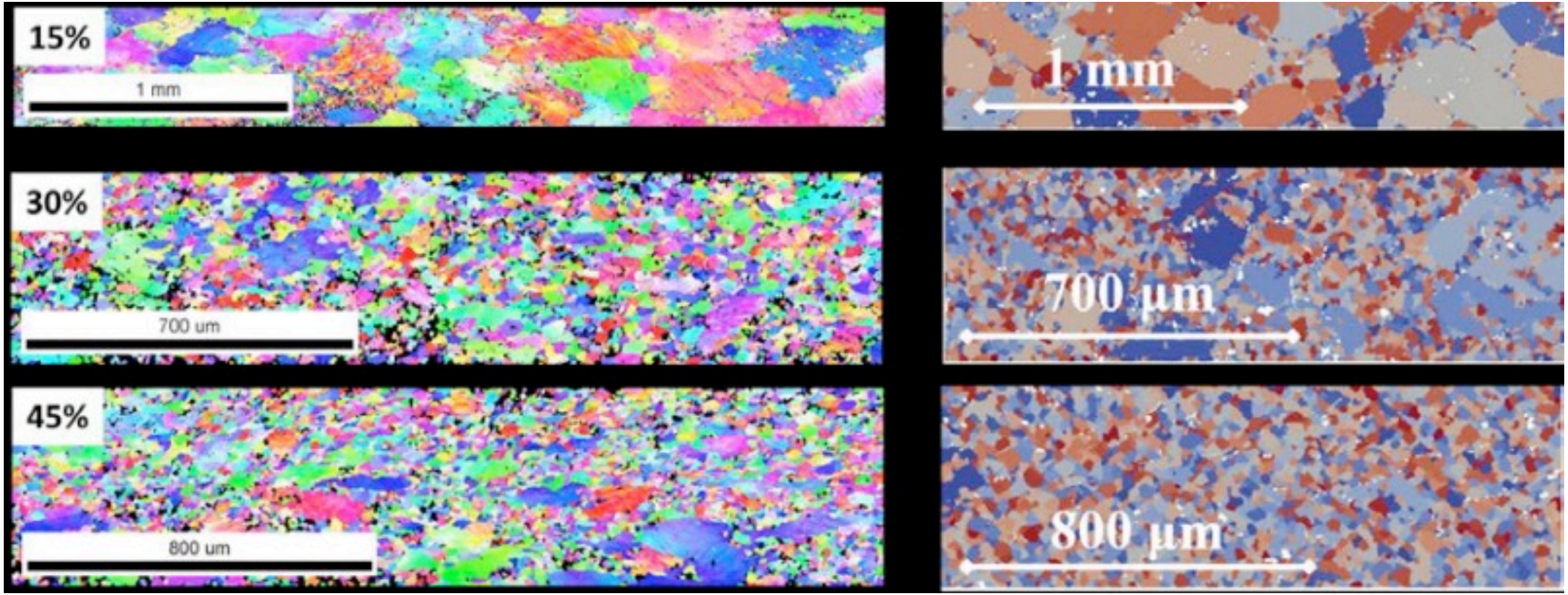


MD simulation of atoms displaced > 0.5 nm by a 10 keV Fe recoil in Fe-10 wt% Cr.
Box size shown is 10 nm by 4 nm.

Simulations shed light on primary damage state and relative proportion of elements displaced.

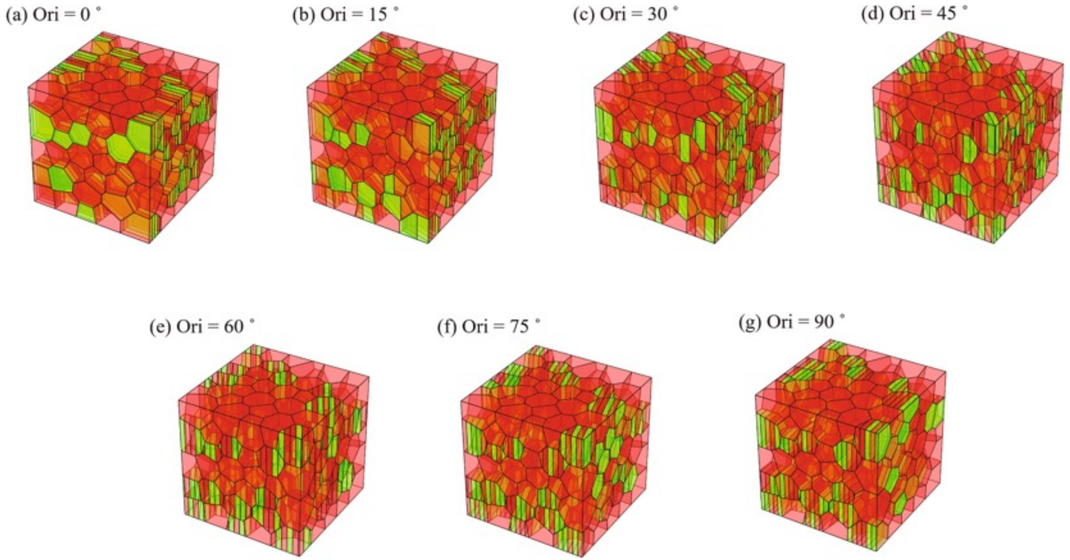
(PNNL unpublished results)

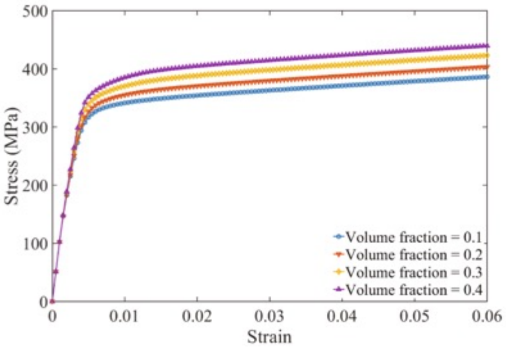
Microstructural Modeling



Comparison of experimental electron back scatter diffraction image (left) with Potts KMC model microstructure (right) of an alloy under different processing conditions (top to bottom).
Image courtesy: William Frazier (PNNL)

Microstructure-informed Crystal Plasticity

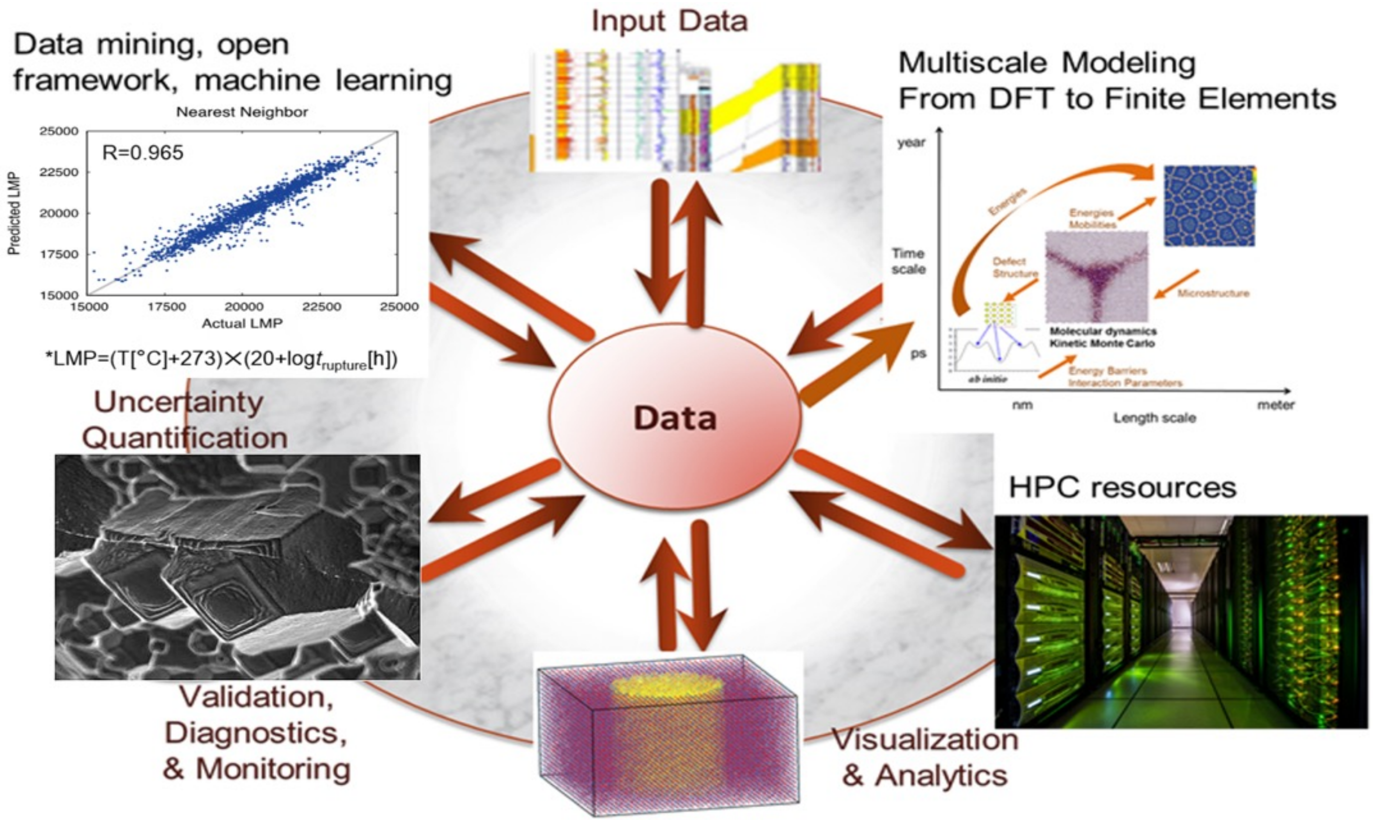




One can vary extended defects—say different thicknesses, orientations, and volume fractions of hydrides in zircaloy and calculate the effect on mechanical properties.

Kulkarni et al, Computational materials science (2021)

ML to Integrate Experiments and Modeling



Selected References