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Irradiation Effects in Materials for Nuclear Applications

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Irradiation Effects in Materials for Nuclear Applications

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There has been a resurgence of international interest in nuclear energy as a clean energy source, and the continued success of existing nuclear reactors, the promise of advanced reactor designs, and the acceptance of used nuclear fuel disposition options are dependent on the performance of materials in extreme nuclear environments. The effects of irradiation on materials properties and performance are critical to safe and reliable reactor operations, efficient use of uranium resources, reduced production of nuclear waste, and acceptable used nuclear fuel disposition [1]. This lecture will provide an introduction to irradiation effects in materials for nuclear applications. The fundamental irradiation damage mechanisms will be described, and the effects of different irradiation environments for nuclear fuels (nuclear fission damage), structural components (neutron damage), and used nuclear fuel and nuclear waste forms (alpha decay damage) will be discussed.

Irradiation damage in materials for nuclear applications primarily results from the production of energetic particles in fission, nuclear reaction and radioactive decay events. The interaction of these energetic particles (fission products, fast neutrons, protons, alphas, and recoil nuclei) with materials results in the production of atomic-scale defects from ballistic collisions and introduction of new chemical elements. In the case of high-energy fission products, the intense ionization along the fission product path can also introduce defects or damage [2]. These irradiation damage processes control the long-term evolution of the materials response to the production and diffusion of defects, fission products and nuclear reaction products. With the exception of fast neutrons, the energetic particles produced in nuclear environments are charged ions that can be also be produced using ion accelerator facilities, such as the JANNuS facility in France [3] or facilities in the USA [4, 5]. Even in the case of a fast neutron, 50 to 100 keV of kinetic energy is transferred on average to a primary knock-on atom (PKA) [6], which is essentially an energetic ion that produces a cascade of displaced atoms through screened-Coulomb collisions, as illustrated in Fig. 1 for silicon carbide [7].

Ion-beam irradiation techniques can be effective in simulating neutron irradiation effects [6, 8], fission damage [9] and alpha-decay damage [10] on short laboratory time scales over a large range of experimental conditions in order to develop more detailed scientific understanding and predictive models of the complex evolution microstructure and phase changes under irradiation. The unique effects of ion irradiation and the use of ions to study fast neutron irradiation effects is a critical topic, since development and qualification of new structural materials requires neutron doses that are too high to be obtained in existing nuclear test reactors or spallation neutron sources.

The advantages and disadvantages of using ions to simulate nuclear radiation environments will be reviewed, with specific examples provided. The integration of computer simulations into these irradiation studies have advanced the interpretation of experimental results

Fig. 1: Defects created by a 50 keV Si PKA cascade in silicon carbide (only defects are shown) [7].

[2, 7] and provided a more comprehensive atomic-level understanding and predictive models of irradiation damage processes.

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Nuclear Radiation Environments of Interest

Nuclear (Fission) Power

Fusion Reactor

Nuclear Waste

Radiation Damage in Nuclear Materials

- Fundamental Radiation Damage Processes
- Nuclear Fuel (in reactor)
 - Fission Damage
 - Accumulation of Fission & Transmutation Products
- Structural Components
 - Fast Neutron Damage
 - Helium & Hydrogen Production
 - Other Transmutation Products
- Nuclear Waste Forms and Used Nuclear Fuel
 - Fission Damage (negligible)
 - Beta Decay Damage (important in short-term – <600 years)
 - Alpha Decay Damage (important over long-term – million years)
- Use of Ion Beams to Study Radiation Damage Processes

Defect Production from Ballistic Processes

- Energetic neutrons, ions and electrons displace atoms
- Energetic Primary Knock-on Atoms (PKAs) create a cascade of ballistic collisions between atoms

Radiation Damage in Nuclear Materials Primarily Caused by Energetic Ions created by Fission, Fast Neutron Collisions, or Radioactive Decay

**Using Ion Beams to Study Irradiation
Effects in Nuclear Materials has become a
Widely-Practiced Approach**
(particularly with lack of fast neutron test facilities)

- Important for separate effects studies
- Use for development & validation of predictive models
- Offers wider range & more control of irradiation conditions

Some Experimental Research Facilities

Fission Damage

Nuclear Fission

- Fission products (**high-energy ions**) lose energy creating linear tracks of defects or structural changes in materials
- Ballistic collision cascade near end of range
- Fission tracks: nano radii (~ 5 nm) and macro lengths (~ 10 microns)
- Accumulation of fission products & gases affects microstructure
- In ~ 0.2 to 0.4% of fission events, helium is produced (ternary fission)

Fission Product Energy Loss

Electronic Energy Loss:

- 1) ~20,000 defects along fission track (MD Simulations)
- 2) Dislocation loops along fission track

Nuclear Energy Loss:

- 1) ~ 65,000 displaced atoms in fission product cascade
- 2) Defect clusters in cascade

Radiation Damage in Nuclear Fuel

Radiation Damage in ^{244}Cm -doped $\text{Ca}_2\text{Nd}_8(\text{SiO}_4)_6\text{O}_2$

- Fission Tracks from Spontaneous Fission
- Alpha-Recoil Tracks (95 keV ^{240}Pu) from Alpha Decay

Studying Fission Damage with Swift-Heavy Ions

➤ Nanoscale, Linear and Parallel Structures

apatite

mica

Leo A Kim *et al.*, Neuron 41, 513 (2004)
Weixing Li *et al.*, EPSL 302, 227 (2011)

Simulating Fission Tracks with Swift-Heavy Ions

1.43 GeV Xe⁺ Tracks in Gd₂Ti_xZr_{2-x}O₇

Bright-Field TEM Images

Gd₂Ti₂O₇

Gd₂TiZrO₇

Gd₂Ti_{0.5}Zr_{1.5}O₇

Swift Heavy Ions Tracks are similar to Fission Tracks

1.43 GeV Xe Track in $\text{Gd}_2\text{Ti}_2\text{O}_7$

$dE/dx = 28.5 \text{ keV/nm}$

HRTEM Image
(M. Lang et al. *Phys. Rev. B* **79**,
224105 (2009))

$dE/dx = 12 \text{ keV/nm}$
(No electron-phonon coupling efficiency)

MD Thermal Spike Simulation
(J. Zhang et al., *J. Mater. Res.* **25**, 1344 (2010))

Neutron Damage in Nuclear Reactors

Core Components

Irradiation-Induced Growth

Irradiation-Induced Cracking

Neutron Spectra vs Monoenergetic Ion Beam

**Fast Neutron
Spectra**

Fig. 11.1. Energy spectra of incident ions in a monoenergetic ion beam, and neutrons from a variety of reactor types (from [7])

Thermal & Fast Neutron Irradiation

Other Considerations

- Production of Helium and Hydrogen from (n, α) and (n,p) nuclear reactions
 - Promotes void and bubble formation
- Accumulation of Transmutation and Decay Products (changes in chemistry of materials)
- Accumulation of Helium, Hydrogen, and Transmutation/Decay Products can be simulated to some extent with Multiple Ion Beams

Protons vs Neutrons: GB Segregation

Protons can provide reasonable simulation of effects from mixed (thermal/fast) neutron spectra

Fig. 11.18. Comparison of grain boundary segregation of Cr, Ni and Si in commercial purity 316 stainless steel following irradiation with either protons or neutrons to similar doses (from [21])

Protons vs Neutrons: Loop Formation

Note Temperature Difference

Fig. 11.19. Comparison of (a) loop size distributions and (b) loop diameter and loop number density for commercial purity 304 and 316 stainless steels irradiated with neutrons or protons to similar doses (from [21])

Fast Neutron Damage

Radiation Damage from Fast Neutrons

- Fission PKAs: 50 to 300 keV Ions
- Fusion PKAs: up to several MeV Ions

Defect Microstructures in Irradiated Materials

Point defect accumulation

Dislocation loop formation

Voids, precipitates, solute segregation

Helium bubbles (grain boundaries)

0.2

0.3

0.4

0.5

0.6

Irradiation Temperature (T/T_M)

Swelling Changes due to Temperature & Dose (Neutron Irradiation)

Fig. 8.19. Swelling in a cold-worked 316 SS baffle bolt in a PWR as a function of position along the bolt length. The bolt head was closest to the core and the temperature distribution is caused by a combination of gamma heating and whether the bolt was exposed to the coolant (courtesy S.M. Bruemmer and Garner FA, PNNL)

Temperature Shift of Void Growth Rate with Dose Rate

Fig. 8.24. Variation of the function $F(\eta)$ in Eq. (8.124) with temperature, illustrating the shift of the peak with dose rate, K_0 . Parameters used to construct the curves are as follows: $\rho_d = 10^9 \text{ cm}^{-2}$, $E_f^v = 1.6 \text{ eV}$, $z_i - z_v = 0.01$, $z_v = 1$, $4\pi R\rho_v = 10^{-11} \text{ cm}^{-2}$, $D_v = \exp(-1.4 \text{ eV}/kT) \text{ cm}^2 \text{ s}^{-1}$ (after [15])

Void Free Regions & Void Size/Density Changes near Grain Boundaries in neutron-irradiated Cu

Zinkle & Farrell, J. Nucl. Mater. 168 (1989) 262

Effect of Grain Boundaries in Vanadium Ion Irradiated at 500°C to 1 dpa

Larger Voids Near
Grain Boundaries

Void and Helium Bubble Lattices

Fig. 8.39. Void lattice in (a) Nb irradiated with 8.5 MeV Ta^+ at 800 °C to 300 dpa, and helium bubble lattice in (b) Mo irradiated with 2 MeV N^+ ions to a dose of 100 dpa at 870 °C (after [39])

3-D Void Lattice in Nb

3-D Bubble Lattice in Mo

G.S. Was, *Fundamentals of Radiation Materials Science* (Springer, 2007)

Weighted Recoil Spectra for Ions in Ni

$W(T)$ = Fraction of defects produced by recoils of energy less than T

Weighted Recoil Spectra for 1 MeV Particles in Cu

**Fast
Neutrons
produce
different
recoil spectra
than Protons**

Fig. 3.6. Weighted recoil spectra for 1 MeV particles in copper, (after [2]). Curves representing protons and neutrons are calculated using Eqs. (3.14) and (3.15), respectively. $W(T)$ for other particles were calculated using Lindhard cross sections and include electronic excitation

Comparison of Damage from 1 MeV Particles in Ni

Alpha-Decay Damage

Alpha-Decay Damage in Materials

Alpha-Recoil Nucleus

- 70 - 100 keV **ions**
- 30 - 40 nm Range
- Creates More Damage (~2000 Displaced Atoms)

Alpha-Particle

- 4.5 - 5.8 MeV **ions**
- 16 - 22 μm Range
- Creates Less Damage (~350 Displaced Atoms)

- Atomic Collision Damage from Recoil Nucleus and Alpha Particle (**both are energetic ions**)
- Helium accumulation

Self-Radiation Damage in ^{244}Cm -doped $\text{Gd}_2\text{Ti}_2\text{O}_7$

$0.6 \times 10^{18} \alpha\text{-decays/g}$

**Fission Tracks
Alpha-Recoil Tracks**

$3.4 \times 10^{18} \alpha\text{-decays/g}$

Amorphous

From Weber, Wald & Matzke, *J. Nuclear Materials* (1986)

Dose Dependence: Large Time Scale Range

Ion Irradiation (minutes) & alpha decay (years) results in excellent agreement

Gd₂Ti₂O₇: Temperature Dependence

Such data can be used to predict long-term behavior of actinide waste forms

Model Predictions Based on Experiment Data

Temperature Dependence

Dose/Time Dependence

Summary on Use of Ion Beams to Study Radiation Effects in Nuclear Materials

- Be aware of dose rate effects (temperature shifts)
- Be aware of mass difference effects (recoil spectra)
- Ion irradiations should complement in-reactor testing or other bulk irradiation (short-lived actinides)
- Ion irradiation studies can be used to guide in-reactor or bulk irradiation testing for model validation
- Ion beams can be used to more precisely control irradiation conditions and parameters to study separate effects and develop/validate models
- Ion beams can be used to achieve high doses currently unachievable (structural materials and waste forms)