Low Energy Ion Irradiation and Its Applicability to Mimic Materials Irradiation Damage from High Energy Protons

Weilin Jiang, David Senor

Pacific Northwest National Laboratory

HPT R&D Roadmap Workshop
May 31 - June 1, 2017, Fermilab
1. Nuclear energy deposition:
   Elastic collision, damage cascades

2. Electronic energy deposition:
   Electron excitation, ionization

3. Electron-phonon coupling:
   Heat production, temperature increase
Emulation of Microstructural Features Using MeV Ion Irradiation and Thermal Annealing

Benefits:

😊 Accurate dose for emulation of material age
😊 Accurate temperature for emulation of the location inside the material with a temperature gradient
😊 Minimum or no radiological activation for immediate release and characterization of irradiated materials.
😊 Implantation of impurity species into a pre-existing structure without thermal constraints.
😊 Fast emulation of structural features within hours to days
😊 Low cost

Limitations:

😊 High dose rate
😊 Possible temperature shift
Irradiation damage in HPT materials starts from production of point defects, followed by their accumulation and interactions, leading to formation of defect clusters up to full amorphization.

Point defects are produced mainly by irradiation of spallation neutrons and ions, especially at low energies, which may be emulated by low energy ion irradiation.

The effects of temperature and its possible gradient in HPT materials may be emulated through post-irradiation thermal annealing at high temperatures, which may lead to formation of fractures and cracks.

Gas bubbles and solid state precipitates in HPT materials may be emulated by implanting the species.

Each contributor may be emulated separately or in a combined way to some extent.
To simplify data interpretation, start with highly oriented pyrolytic graphite (HOPG), pure light metals or model alloys without grain boundaries, pores or high-level impurities, followed by polycrystalline materials with increasing levels of material complexities.

Perform in-situ damage accumulation study of HOPG irradiated, for example, with H\textsuperscript{+} ions and self-ions (C\textsuperscript{+}) as a function of dose and temperature.

Perform in-situ and ex-situ thermal annealing study of defect recovery and clustering.

Perform in-situ HIM irradiation study of microstructural evolution in polycrystalline graphite.

Perform microscopy study of HOPG and polycrystalline graphite implanted with H, He and non-gaseous spallation/transmutation species (e.g., Li) and annealed at high temperatures to emulate microstructures for study of various features, including polycrystallization, amorphization, shrinking/swelling, creep, Mrozowski cracks, gas bubbles, and precipitates.

Measure physical properties, including thermal conductivity, electrical conductivity, and mechanical strength.

Compare the emulated microstructures and properties with those of high energy proton irradiated graphite and develop a fundamental understanding of the structure-property relationships, which may help assess and predict material performance.
Fundamental Processes of Ion-Solid Interactions in the MeV Energy Range

- Rutherford Backscattering (RBS)
- Product of Nuclear Reaction (NRA)
- X-Ray (PIXE)
- γ-Ray (PIGE)
- Recoil Target Atom (ERDA)
- Damage Peak
Ion Channeling and RBS/C

From L. C. Feldman, et al., “Materials Analysis by Ion Channeling”
Disorder Accumulation in $\gamma$-LiAlO$_2$ at 573 K

- Disorder on the Al sublattice saturates at levels of 0.3 and 0.5.
- No full amorphization occurs at the highest applied dose of 1 dpa.
Effect of Irradiation Temperature on Disordering Rate

Disordering rate decreases with increasing irradiation temperature due to simultaneous recovery.
Thermal Recovery of Defects on Both Si and C Sublattices in Irradiated SiC

20-min Isochronal Anneals

Similar recovery stages (I, II, III) on both Si and C sublattices
Li and H Out-diffusion in H\(^+\) Irradiated \(\gamma\)-LiAlO\(_2\)

- Material decomposition, Li diffusion and loss during irradiation
- H diffusion and release during thermal annealing
Amorphization and Precipitate Formation

\[
\gamma\text{-LiAlO}_2 \text{ implanted to } 10^{17} \text{ H}^+ / \text{cm}^2 \text{ at } 773 \text{ K}
\]

- The precipitate in rectangular shape is identified as cubic LiAl$_5$O$_8$ with zone axis [211] that is parallel to $\gamma$-LiAlO$_2$ [100].
- Precipitates also show in triangular shape, which has a zone axis [111].
- Amorphization and gas bubbles near the surface are observed.
STEM-EELS Mapping of Precipitates in 3C-SiC

3C-SiC implanted to $9.6 \times 10^{16}$ $^{25}$Mg$^+$/cm$^2$ at 673 K and annealed at 1573 K for 12 h

Formation of cubic Mg$_2$Si and tetragonal MgC$_2$ tetrahedra in Mg$^+$ implanted 3C-SiC.
As an advanced instrument, HIM was developed and commercialized in 2007, providing cutting-edge imaging and chemical analysis with a sub-nanometer probe. One of the unique capabilities is the in-situ study of microstructural evolution in bulk material at a microscopic site of choice under He⁺ ion irradiation.
He Bubble Formation in $\gamma$-LiAlO$_2$

$\gamma$-LiAlO$_2$ irradiated with 25 keV He$^+$ at RT under HIM
(He$^+$ ion projected range: 236 nm; max. 62.3 at.% He)

5.6 dpa
62.3 He%
He Bubble Formation in a $\gamma$-LiAlO$_2$ Grain under HIM

1.0 $\mu$m $\times$ 1.0 $\mu$m; 2.1 at.% He
Microstructural Evolution of Amorphous SiO₂ Nanoparticles and LiAlO₂ at a Void under HIM

0.785 μm × 0.785 μm; 0.19 dpa
Mg\textsuperscript{+} and H\textsuperscript{+} Irradiated HOPG

Graphite

A: Mg\textsuperscript{+} and H\textsuperscript{+} irradiated
B: H\textsuperscript{+} irradiated
C: Mg\textsuperscript{+} irradiated
D: Non-irradiated

G: Graphite peak at 1580 cm\textsuperscript{-1}
D: Disorder peak at 1360 cm\textsuperscript{-1}
DLC: Broad diamond like carbon peak ranging from 1100 to 1700 cm\textsuperscript{-1}

keV Mg\textsuperscript{+}/cm\textsuperscript{2}
70  7.1 \times 10\textsuperscript{16}
120 1.0 \times 10\textsuperscript{17}
200 2.0 \times 10\textsuperscript{17}

0.78 MeV H\textsuperscript{+}
10 \mu m Al foil
1.05 \times 10\textsuperscript{17} H\textsuperscript{+}/cm\textsuperscript{2}