Moving from Dpa to Changes in Materials Properties

Meimei Li, Argonne National Laboratory

Radiation Effects in Superconducting Magnet Materials (RESMM12)
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Outline

- Irradiation sources
- Fundamentals of radiation damage
  - Displacement damage
  - Transmutation effects
- Irradiation-induced microstructural changes
- Irradiation-induced property changes
- Damage correlation
- Radiation damage modeling and benchmarking experiments
- Summary

Irradiation Sources

- Radiation damage has been studied using various irradiation sources, such as fission neutrons in nuclear reactors (e.g. fast fission reactors, mixed-spectrum fission reactors), fusion neutrons in a D-T fusion neutron source, spallation neutron sources, ion irradiation with accelerators, and high-energy electron beams, etc.

- Simulation irradiation techniques have been used when there is lack of prototypic irradiation facilities. For instance, material development for fusion reactors has been made primarily in fission reactors.

| Irradiation source	Facility	Particles	Displacement dose rate (dpa/s)	He (appm)/dpa	H (appm)/dpa
Mixed spectrum fission reactor	HFIR	Neutron	1.1×10^{-7}	3.4	62
Mixed spectrum fission reactor	BR2	Neutron	4×10^{-7}	40	
Mixed spectrum fission reactor	HFR	Neutron	1.6×10^{-7}	20	
Fast fission reactor	EBR-II	Neutron	1.2×10^{-6}	0.15	2.3
Fast fission reactor	JOYO	Neutron	3×10^{-6}	0.17	
Fast fission reactor	PHENIX	Neutron	1.8×10^{-6}	0.3	
Accelerator/Cyclotron	PIREX	590 MeV proton	5×10^{-7}	100	800
Accelerator/Cyclotron	Single beam, HMI, Germany	M^+, 10-20 MeV proton, and α-particle	10^{-4}		100

(Klueh and Harries 2001)

Displacement Damage

- Radiation damage is produced by energetic particles (neutrons, ions, protons, electrons, etc.) interacting with a crystalline solid.
- An incident particle transfers recoil energy to a lattice atom, forming a primary knock-on atom (PKA)
- A PKA displaces neighbouring atoms, resulting in an atomic displacement cascade, leading to formation of point defects and defect clusters of vacancies and interstitial atoms.
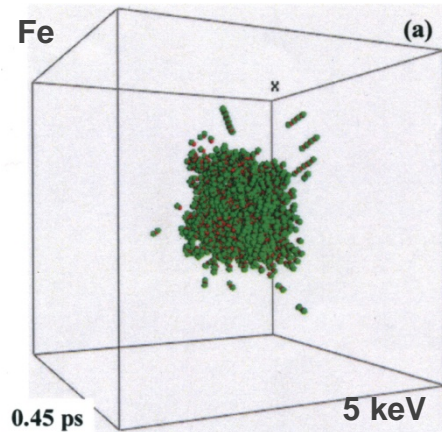
t = 0.45 ps

(Stoller 1997)
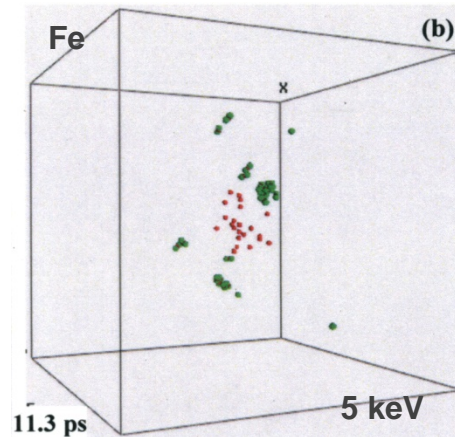
t = 11.3 ps

Molecular dynamic simulation of displacement cascade: Peak damage < 1 ps; stable configuration ~10 ps

(green: interstitials, Red: vacancies)

Displacements Per Atom (DPA)

- To evaluate radiation damage, a fundamental parameter that characterizes lattice displacement events is required.

- Dpa has been used to compare radiation damage by different radiation sources. It is a damage-based exposure unit and represents the number of atoms displaced from their normal lattice sites as a result of energetic particle bombardment.

- Calculations of dpa values

$$N_d = \begin{cases} \dfrac{\kappa(T - E_e)}{2E_d} = \dfrac{\kappa T_{dam}}{2E_d}, & T_{dam} > 2E_d \\ 1, & E_d < T_{dam} < 2E_d \\ 0, & 0 < T_{dam} < E_d \end{cases}$$

- N_d is the number of displaced atoms produced by a PKA

- T is the recoil energy of a PKA; E_e is the total energy lost by electron excitation; k is the damage efficiency; T_{dam} is the damage energy available for elastic collisions; and E_d is the threshold displacement energy. $\sigma(E)$ is the displacement cross section for an incident particle at an energy E.

$$dpa = \Phi\sigma = \Phi \int_{E_d}^{T_{max}} \Phi \frac{d\sigma(E,T)}{dT} N_d$$

- Irradiation-induced changes of material properties are measured as a function of dpa

Transmutation Effects

- Nuclear transmutation reactions occur, producing solid transmutation products and He and H gas atoms

- He and H gas atoms can have pronounced effect on materials performance even at low concentrations

- The production rates of He, H (He/dpa, H/dpa) can be quite different in different irradiation environments.
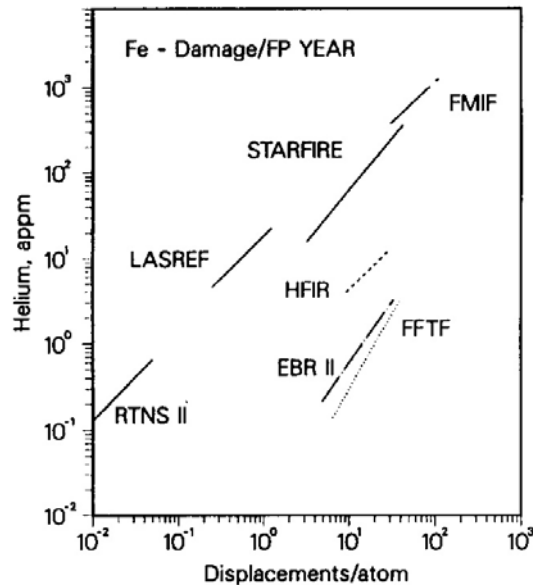

Comparison of displacement and He production in Fe in different irradiation facilities (Greenwood 1994)
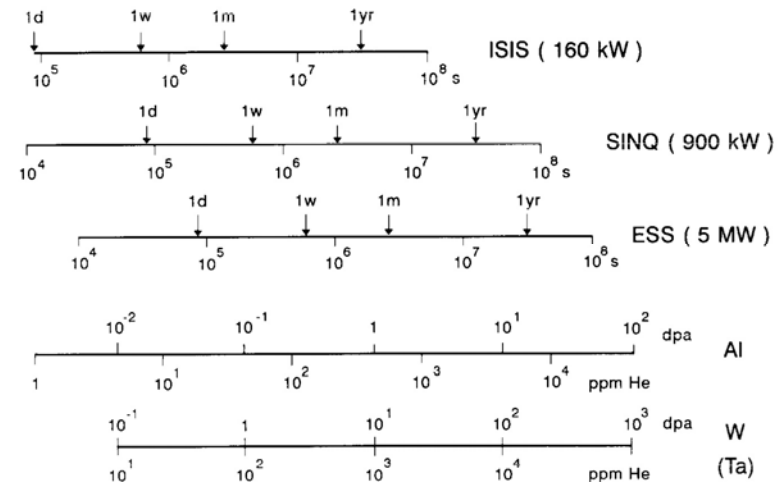
Displacement and helium production in Al and W, respectively as a function of service time for targets exposed to the proton beams of ISIS, SINQ and ESS (Ullmaier and Carsughi 1995)
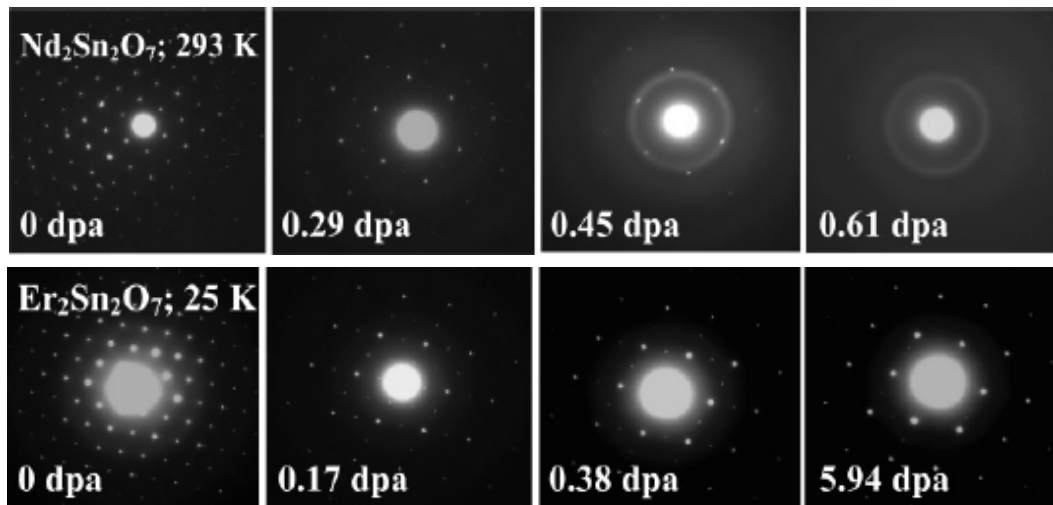
Radiation-induced Microstructural Changes

- Atomic displacements by high energy particles induce the formation of point defects and defect clusters within picoseconds. With time, normal diffusion processes take place and irradiation-induced defects recombine or cluster to form more stable damage structure.

- A large population, different type of defect structure forms during irradiation, e.g. dislocation loops, dislocation network, stacking fault tetrahedra, voids, He bubbles, precipitates.
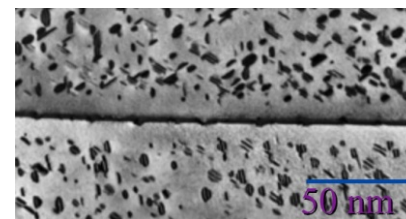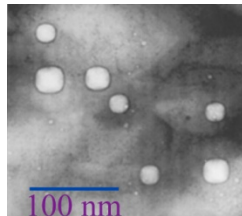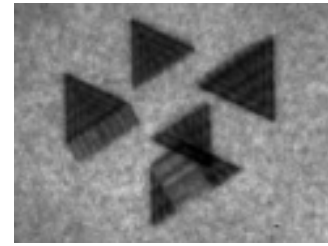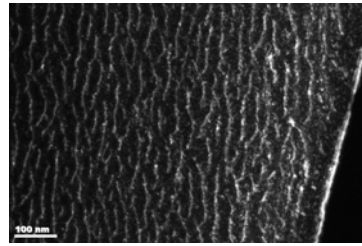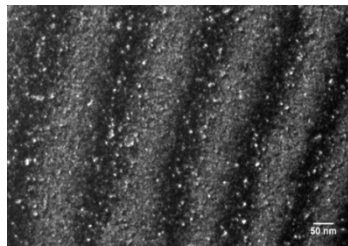
Irradiation-induced Amorphorization

- Crystalline-to-amorphous transition observed in ceramics during irradiation.
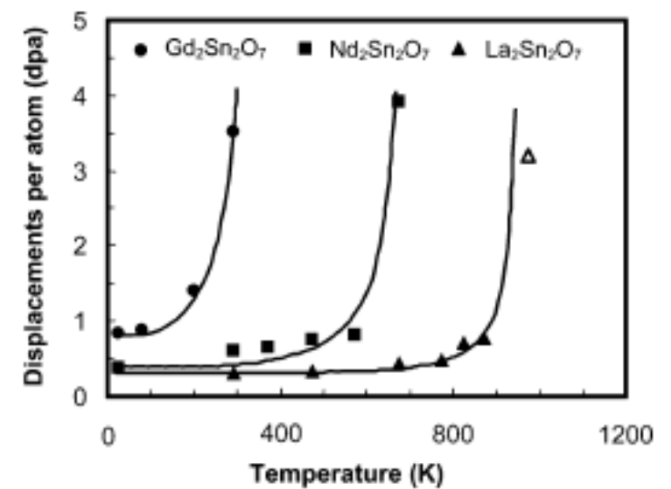

(R.C. Ewing, J. Lian, J. Zhang, L. M. Wang 2009).

Radiation-induced Property Changes

- Radiation-induced microstructural changes significantly degrade materials' properties
  - Degradation of physical properties (increase in electrical resistivity, decrease in thermal conductivity, etc.)
  - Radiation hardening and embrittlement
  - Irradiation creep
  - Void swelling
  - High temperature He embrittlement
  - Reduction in fatigue performance, irradiation-assisted stress corrosion cracking
- Synergistic effects of radiation, corrosive media, temperature, and stress
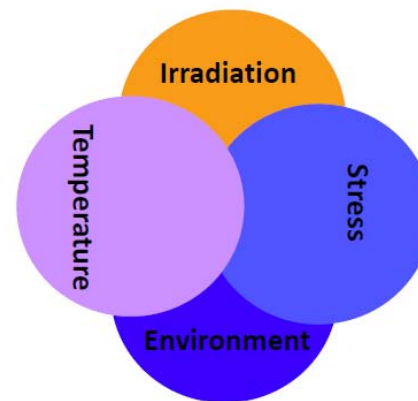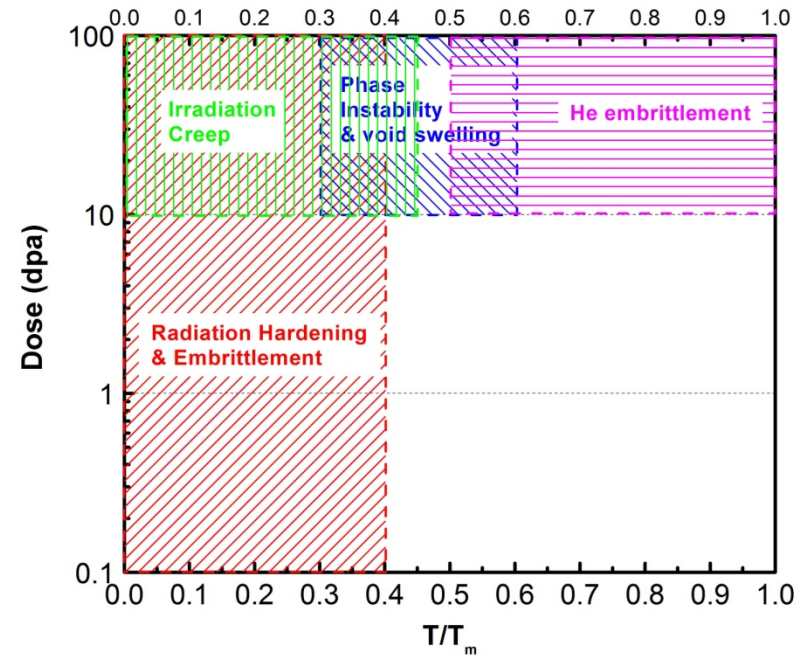
Electrical Resistivity of Irradiated Mo
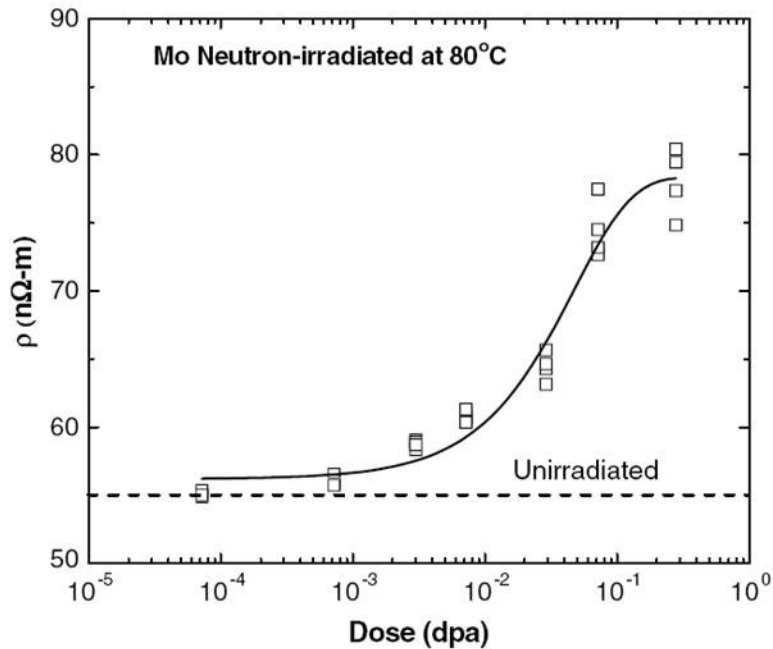
(Li et al. 2008)

Room-temperature electrical resistivity of neutron-irradiated Mo

- No measurable decrease in electrical conductivity at a very low dose of (~0.0001 dpa)

- Electrical conductivity decreased continuously with increasing dose.

Thermal Conductivity of Irradiated Graphite

- Neutron irradiation reduces thermal conductivity of graphite
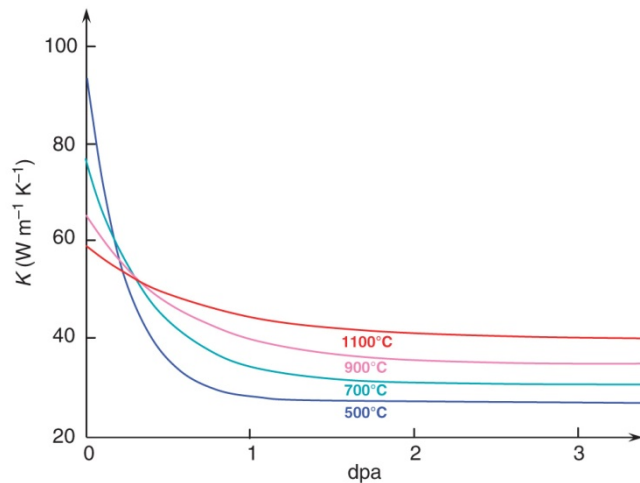
Thermal conductivity of irradiated graphite starts to decrease at very low neutron doses (10^{-3} dpa); As dose increases, the reduction in thermal conductivity tends to saturate (Bonal et al 2009)
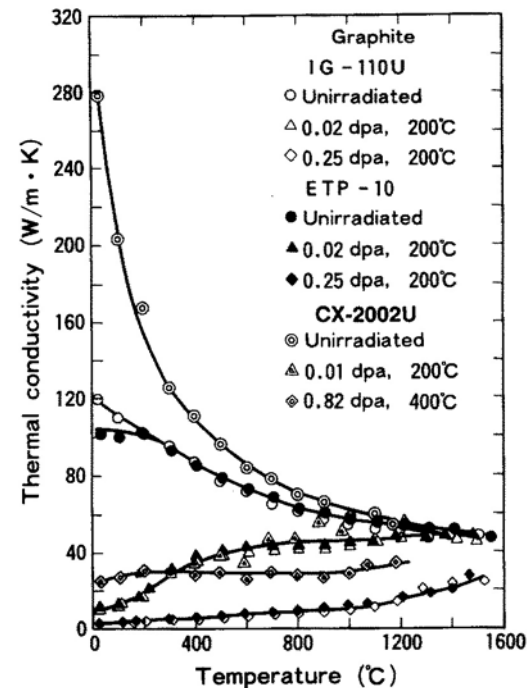
Thermal conductivity as a function of test temperature for neutron-irradiated graphite (Maruyama and Harayama 1992)
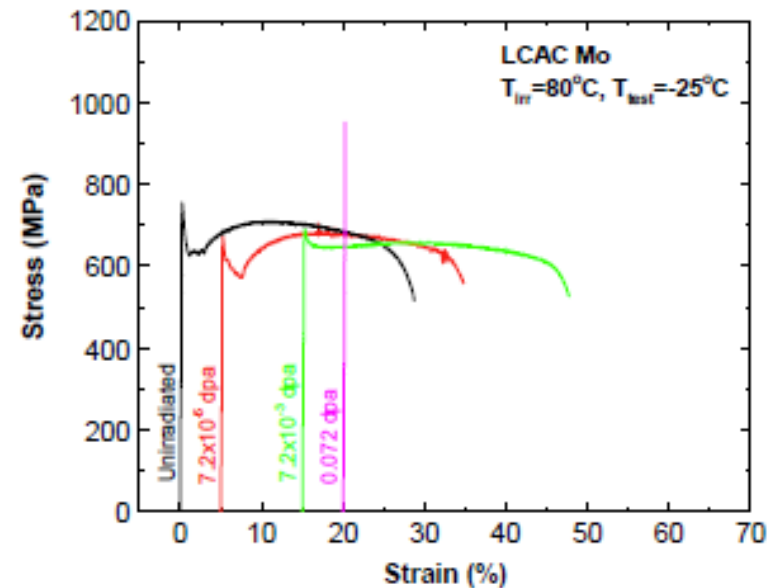
Radiation Hardening and Embrittlement

- Neutron irradiation leads to significant hardening, loss of strain hardening capability, and ductility loss.
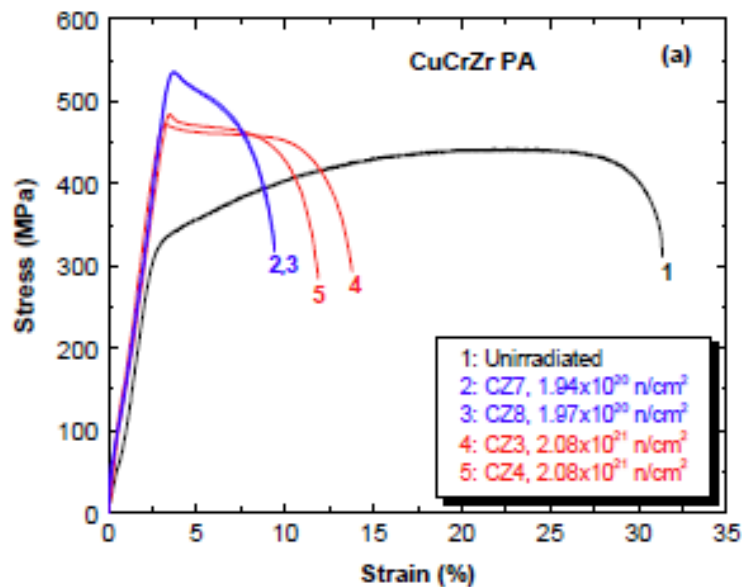
*(Li et al. 2010)*

(Li et al. 2008)

Damage Correlation

- Dpa is a most commonly-used damage correlation parameter. However, damage correlation and data extrapolation must consider other aspects and base on a fundamental understanding

- Damage correlation parameters
  - Irradiation particle type, energy
  - Energy spectra
  - Flux or dose rate (dpa/s)
  - Fluence or dose (dpa)
  - Irradiation temperature
  - Transmutation (e.g. He, H)
  - Pulsed irradiation vs. continuous irradiation

Energy Spectra and Damage Production

- Energy spectra in different irradiation sources vary significantly, and primary damage production is different

- Electron irradiations create single Frenkel defects (point defects of interstitials and vacancies); low-energy protons and light ions create similar defects as electron irradiations

- Neutrons and heavy ions produce cascade displacement damage

- Fusion neutrons create significant subcascades with very high energies PKAs

- Analysis of recoil spectra is a key to damage correlation
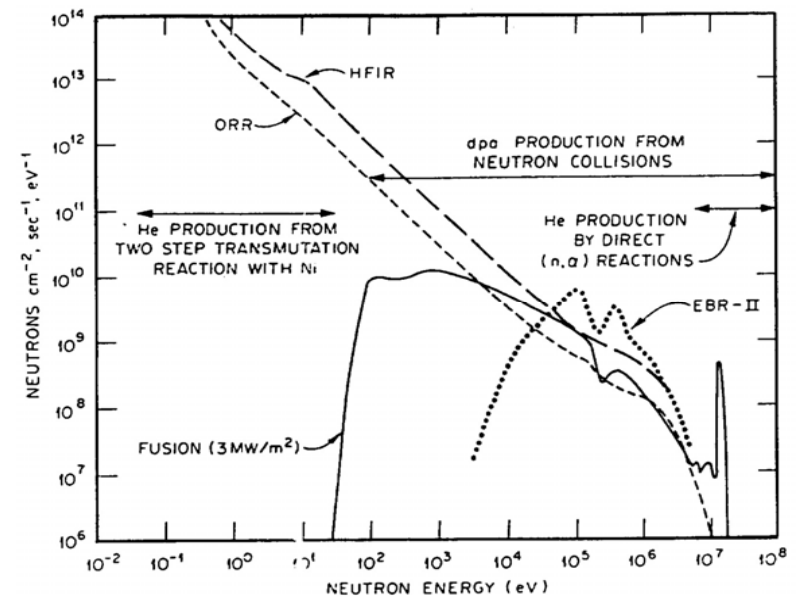
Energy spectra in various nuclear reactors

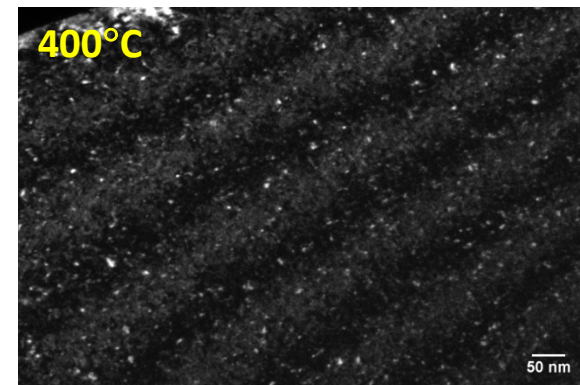(Abbromeit 1994)

Effect of Irradiation Temperature

- Irradiation at different temperatures can result in different defect structures

TEMPERATURE DEPENDENCE OF MICROSTRUCTURAL COMPONENTS IN NEUTRON-IRRADIATED AUSTENITIC STAINLESS STEEL

Zinkle, Maziasz & Stoller, J. Nucl. Mater. 206 (1993) 266

Dose = 0.64 dpa

20°C

Dose = 0.64 dpa

400°C
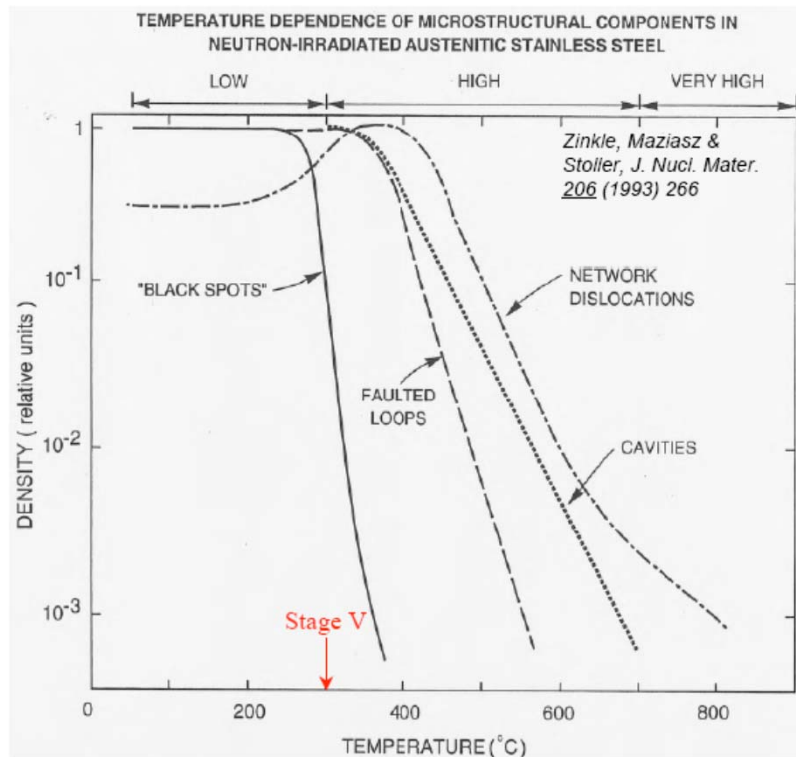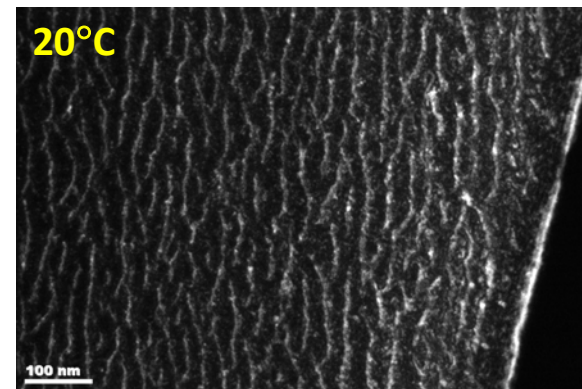
Recovery Stages

- The role of irradiation temperature is related to defect mobility
  - Stage I: self-interstitial atom (SIA) migration
  - Stage II: migration of SIA clusters and SIA-impurity complexes
  - Stage III: vacancy migration
  - Stage IV: migration of vacancy clusters and vacancy-solute complexes
  - Stage V: thermal dissociation of vacancy clusters

Correlated in-cascade recombination reduces surviving defect
fraction due to freely migrating interstitials
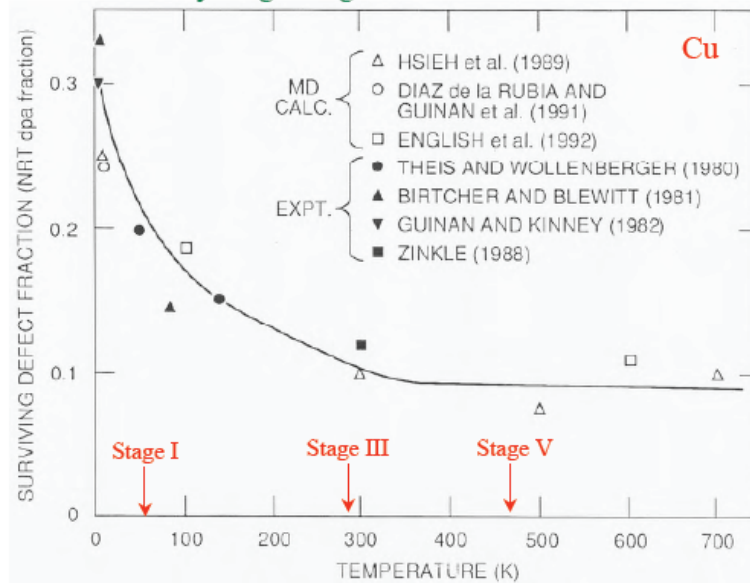
Zinkle and Singh, J. Nucl.
Mater. 199 (1993) 173

Dose Rate Effect

- Dose rate can significantly affect microstructural evolution and physical and mechanical properties

- Dose rate plays a critical role in irradiation-induced swelling, irradiation creep, and solute segregation.

Neutron Damage on Stainless Steel

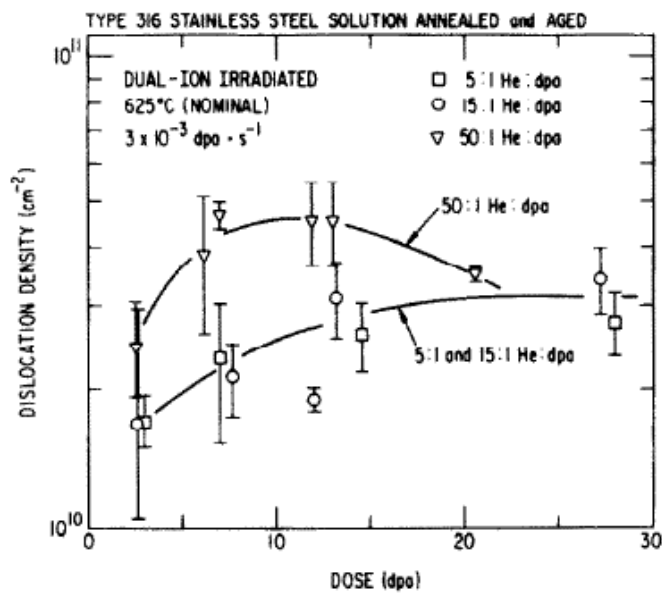100 nm
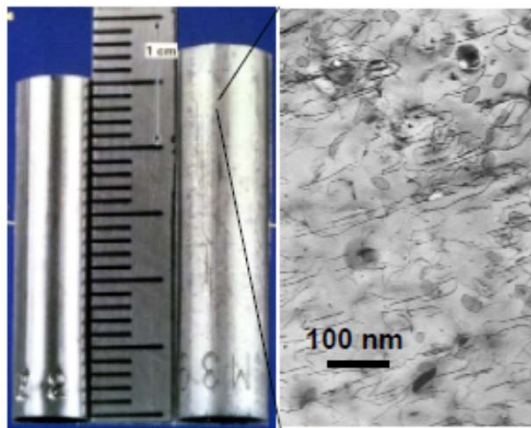
control   1.5 x 10^23 n cm^-2

20% CW 316 stainless steel, 796 K

(Straalsund et al, 1982).
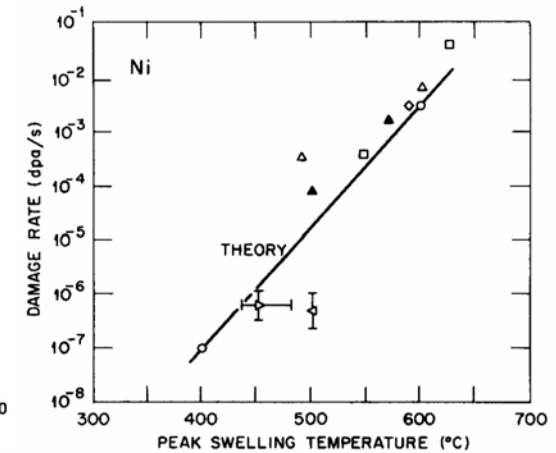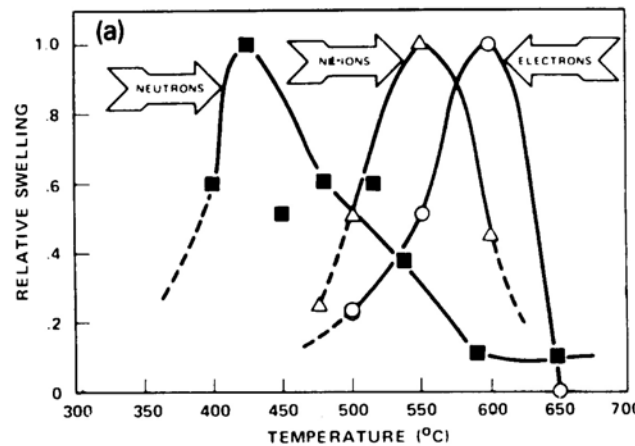
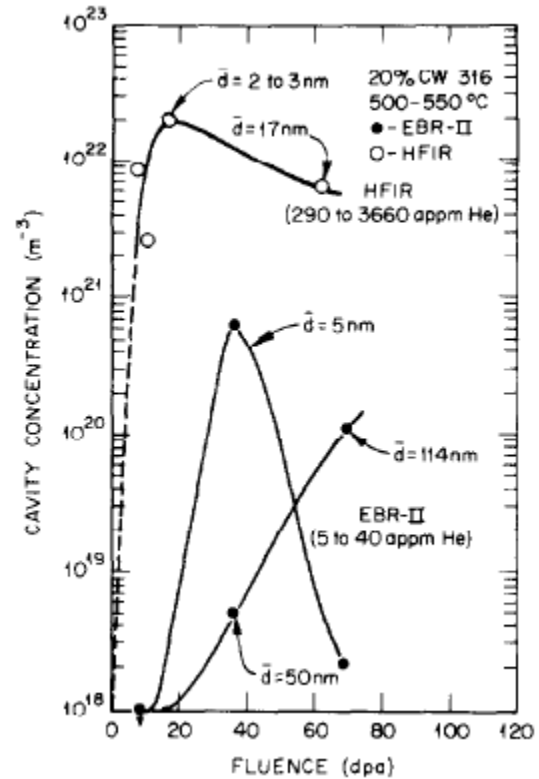The peak swelling temperature shifted as a function of dose rate. (Abromeit, Mansur 1994).

Effect of Helium

- Helium can significantly influence the microstructural evolution during irradiation

High concentration of He tends to increase dislocation density at lower irradiation doses (Ayrault et al. 1981)
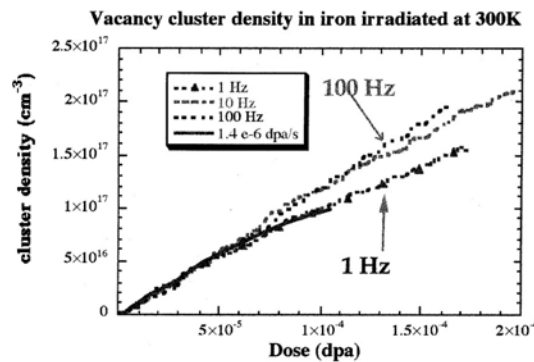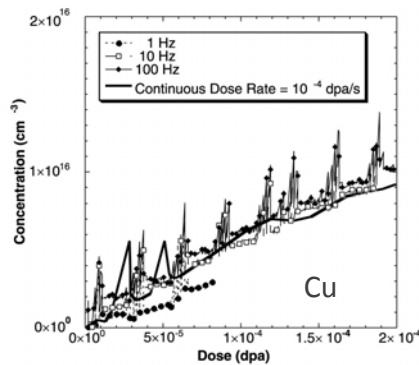
Higher He contents lead to higher cavity density (Stoller 1990)

Pulsed Irradiation vs. Continuous Irradiation

- Due to the pulsed nature of irradiation, the interplay of irradiation flux, temperature and pulse frequency can change the kinetics of irradiation damage accumulation compared to a steady-state continuous irradiation.

MD/kMC simulations showed evolution of radiation damage as a function of dose in Cu and Fe under pulsed irradiations and continuous irradiation (Caturla et al 2001)
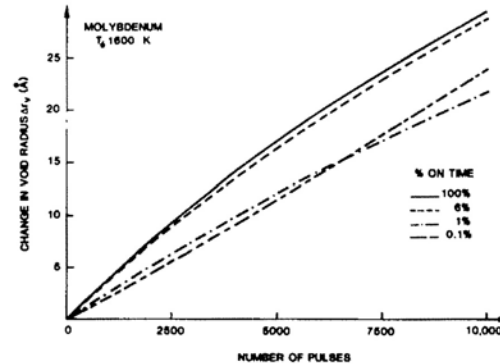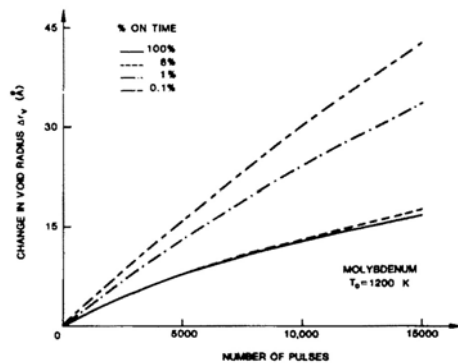

The pulse nature of irradiation is important when the characteristic pulse times are comparable to or greater than the vacancy and interstitial reaction times (Kmetyk et al 1981)

Radiation Modeling and Benchmark Experiments

- Effective radiation damage correlation requires close coordination between experimental, theoretical and computational studies.
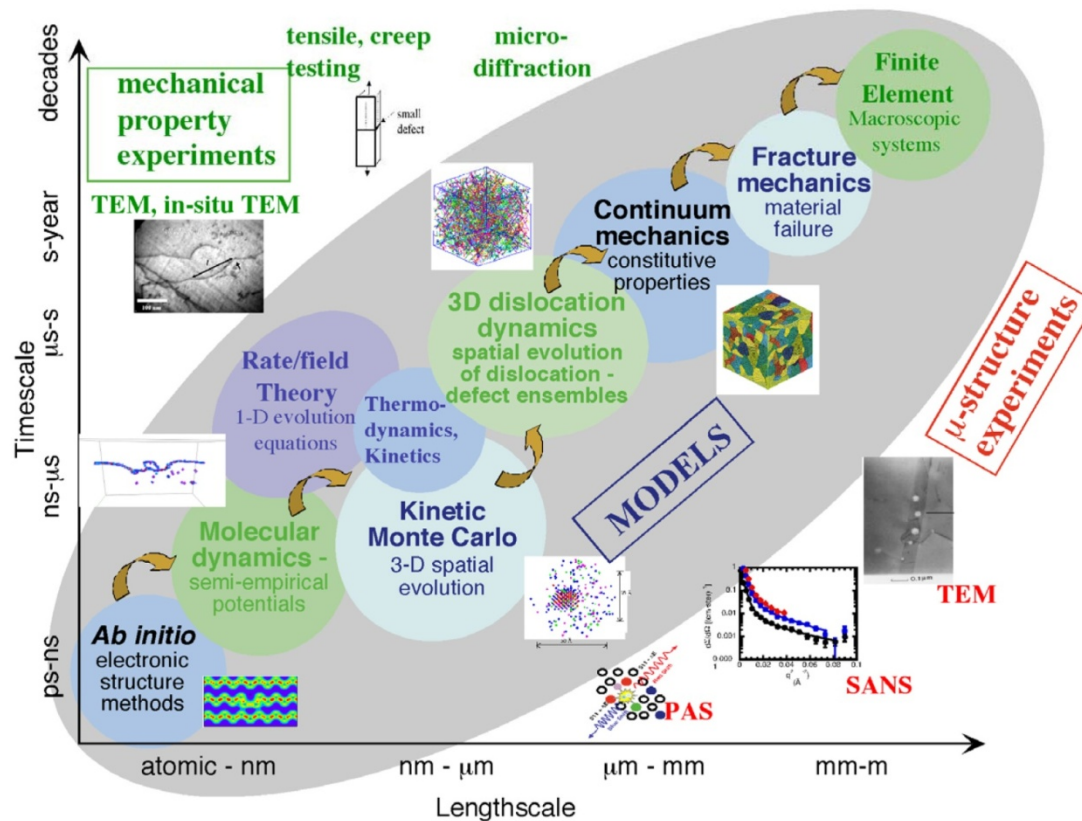
B. Wirth et al (2004)

Ion Irradiation and Implantation

- *In situ* TEM ion irradiation is a powerful tool for introducing disorders in materials and validate and verify computer models

  - Real-time observation of defect formation and evolution during irradiation

  - A wide range of techniques including imaging, electron diffraction, and spectroscopy

  - Well-controlled conditions (temperature, ion, ion energy, dose rate, dose)

  - High doses (e.g. 100 dpa) can be achieved in hours; irradiation dose rates can be varied over several orders of magnitude

  - Studies of single-parameter effects and synergistic effects of irradiation, temperature and stress

IVEM: Hitachi H-9000NAR, 100-300 kV, 0.2-1000 kx, spatial res 0.25 nm, time res 0.03 sec,

Irradiation: All ions, 1 MeV max.

Loading: 20 – 1273 K, straining stage

Damage Correlation between in situ Ion Irradiation and Neutron Irradiation

Molybdenum was irradiated at IVEM by 1 MeV Kr++ at 80°C

**Molybdenum was neutron irradiated in HFIR at 80°C**
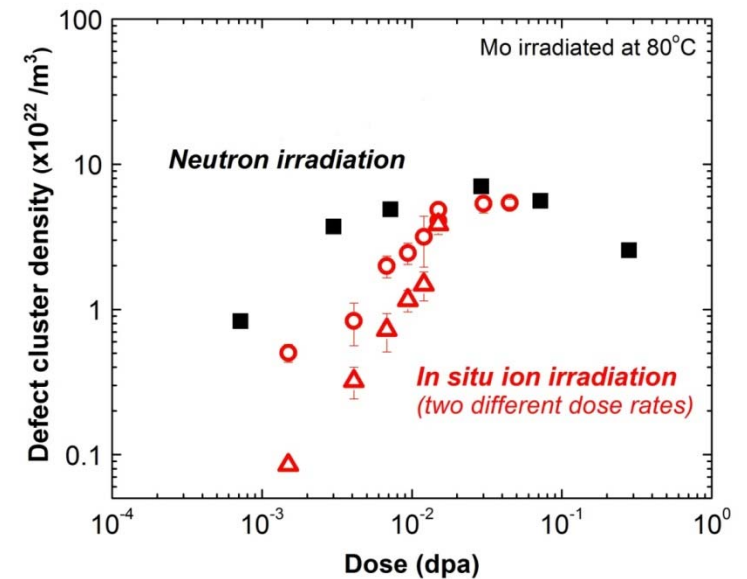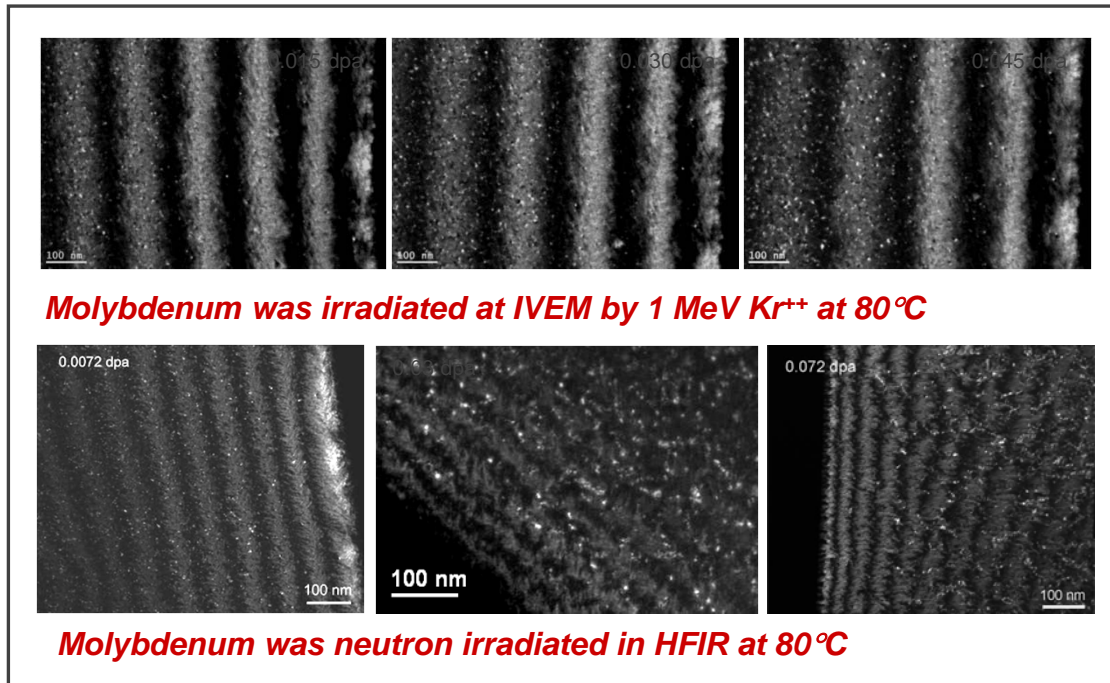
- A direct comparison of defect microstructure produced by neutron irradiation and in-situ ion irradiation was made by irradiating exactly the same material under equivalent irradiation conditions.
- A simple comparison based on equivalent dose (dpa) is inadequate. More specific parameters are proposed to allow detailed damage correlations by taking into account of the surface sink effect, displacement damage rate, and damage profile.
- Computer models verified and validated by in situ ion irradiation of thin foils can be used to predict neutron damage in reactor materials

(M. Li, M. Kirk, P. Baldo, D. Xu, B. Wirth)

In situ Ion Irradiation Experiments

- Conducted well-controlled *in situ* TEM ion irradiation of thin films provides a complete set of high-quality, 3-D, quantitative information used for improving and validating a computer model.
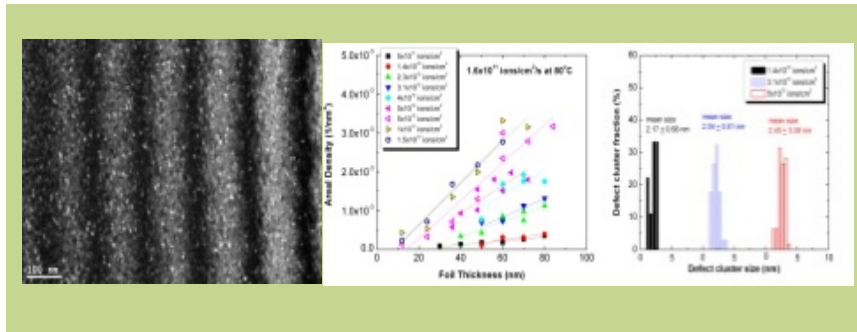

This set of experimental data described the defect evolution behavior at a level of detail unavailable before.
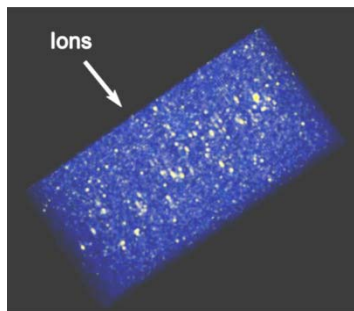
3D electron diffraction tomography measurements provided new view of irradiation defects in ion irradiated thin foils.

Computer Simulations

- Conducted multiscale modeling to simulate defect evolution from atomic-scale, pico-second events to nanometer-scale, hour evolution of defect structures.
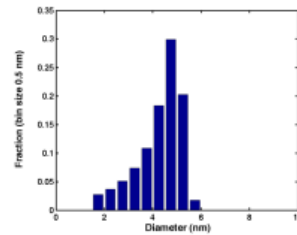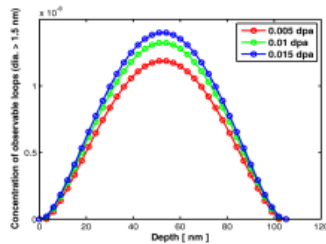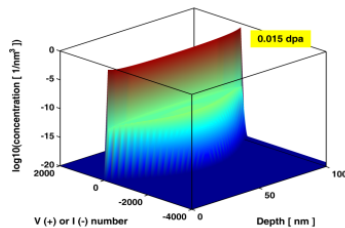

- *Combine molecular dynamics (MD) simulations of displacement cascades and the reaction rate theory to simulate defect evolution, taking into consideration of surface sink effects in TEM thin films under ion irradiation.*

- *Unknown material parameters, e.g. migration energy of defect clusters, were determined using the experimental data.*

Direct Comparison

■ Quantitative, absolute comparisons between experiments and modeling at the same spatial and time scales is leading to the establishment of an accurate, reliable computer model.
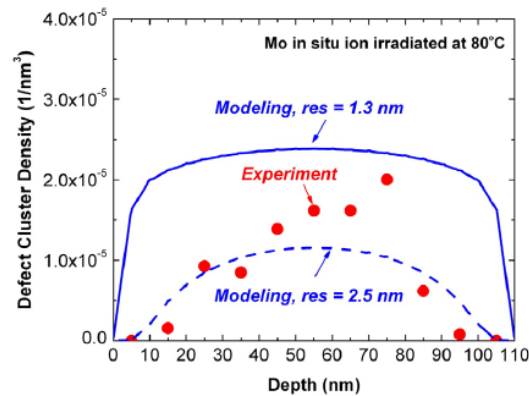
• The spatially-dependent cluster dynamics model captured the essential physics of damage in irradiated Mo thin films.

• Iterative refinement of key material parameters with in situ ion irradiation data led to a more accurate cluster dynamic model.
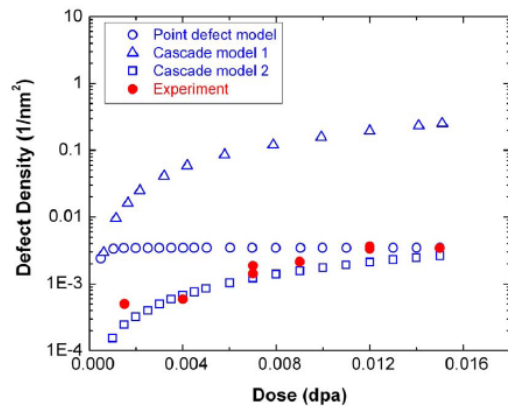
Predictions

- To demonstrate that a model built upon the fundamental physics of irradiation damage in ion-irradiated thin films can be used to predict neutron damage in bulk in extreme nuclear environments.

The model is being additionally validated by neutron irradiation data that were obtained on bulk materials with identical chemistry and metallurgy as used for in situ ion irradiations.
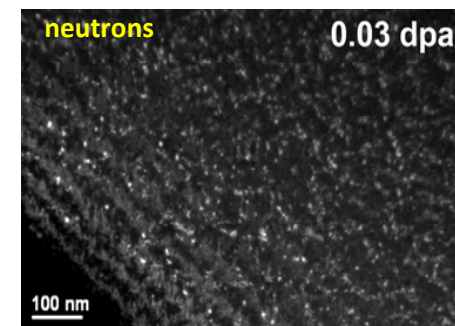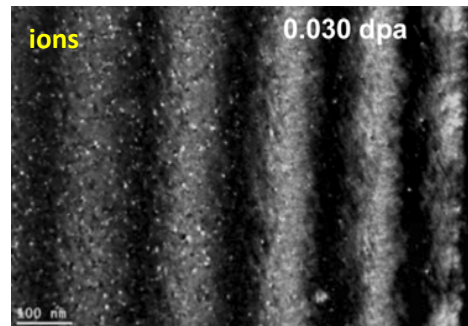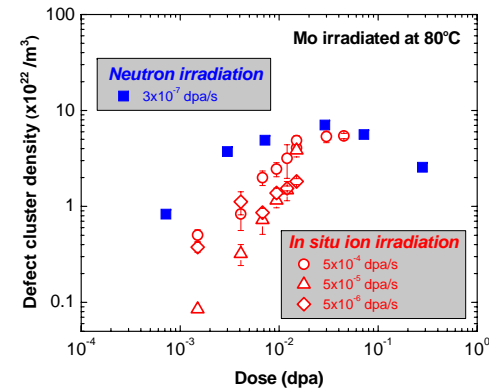
Summary

- Radiation damage by energetic particles can introduce displacement cascade damage and transmutation production of helium and hydrogen and other impurities.
- Radiation effects in materials have been studied using various irradiation sources, e.g. fission, fusion and spallation neutron sources, high-energy ions and electron beams, etc.
- Dpa is the parameter commonly used to correlate displacement damage. However, the extent of radiation damage cannot be fully characterized by a single parameter.
- Damage correlation between different types of irradiations should consider:
  - Primary recoil energy spectra
  - Displacement dose rate, dpa/s
  - Transmutation production rates, He/dpa and H/dpa
  - kinetics of irradiation-induced defect production and accumulation behavior due to pulsed irradiation
- Reliable damage correlation requires integration of theoretical, computational and experimental tools.