Experience with Moving from Dpa to Changes in Materials Properties

Meimei Li, Argonne National Laboratory
N. V. Mokhov, Fermilab

46th ICFA Advanced Beam Dynamics Workshop
Sept. 27 – Oct. 1, 2010
Morschach, Switzerland

Outline

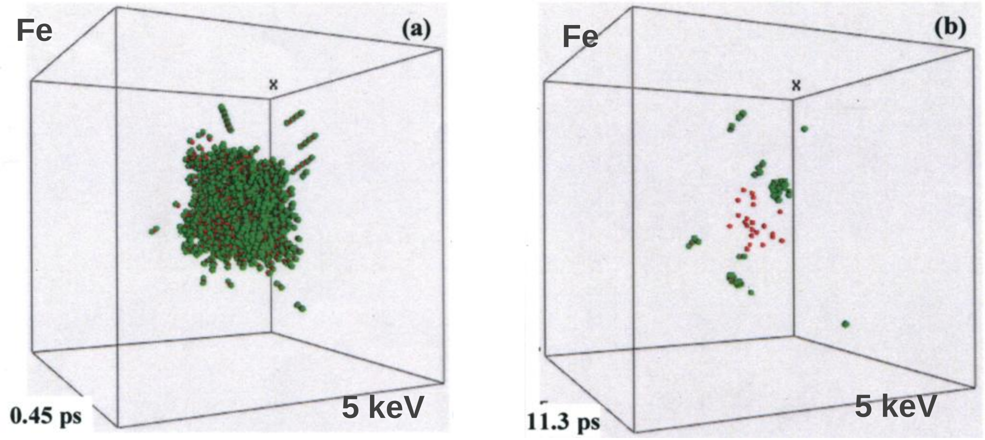
- Introduction
- Damage Correlation
- Damage Correlation between Neutron Irradiation and High Energy Proton Irradiation
- Radiation Effects in Graphite
  - Thermo-physical properties
  - Dimensional changes
  - Irradiation creep
  - Mechanical strength
  - Dose rate effects
  - Effects of helium
- Summary
- Recommended Future Work

Introduction

- Radiation damage is a serious concern in the target design in high energy particle accelerators
- Lack of prototype irradiation facilities require extrapolation of irradiation data obtained from different irradiation sources
- Damage correlation between different irradiation sources is critical in assessment of the target performance in high energy accelerators

Radiation Damage

- Radiation damage is produced by energetic particles, such as neutrons, ions, protons, or electrons, interacting with a crystalline solid.
- **Primary Radiation Damage: Cascade Displacement**
  - 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.
  - The displacement cascade event occurs within picoseconds.



t = 0.45 ps

t = 11.3 ps

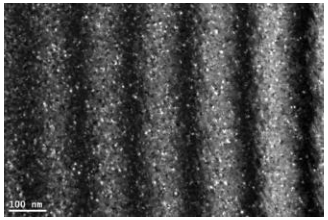
Molecular dynamic simulation of displacement cascade: Peak damage < 1 ps; stable configuration ~10 ps

(green: interstitials,
Red: vacancies)

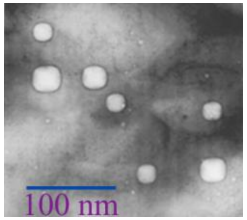
Stoller (1997)

Radiation Effects

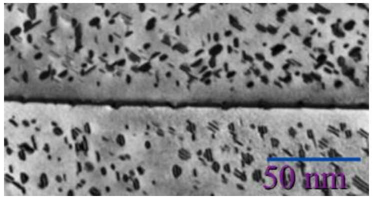
- A large population, different type of defects form during irradiation.



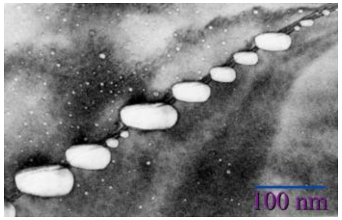
Dislocation loops



Voids



Precipitates



He bubbles

- Radiation-induced microstructural changes significantly degrade materials' properties
  - Degradation of physical properties
  - Dimensional instability
  - Radiation hardening and embrittlement
  - Irradiation creep
  - Helium embrittlement
  - Reduction in fatigue performance

Displacements Per Atom (DPA)

- Dpa, a dose unit, is the main parameter to compare radiation damage by different radiation sources

$$dpa = \int_0^{t_r} \phi_{tot}(t) \int_0^{\infty} \sigma_d(E) \varphi(E,t) dE dt$$

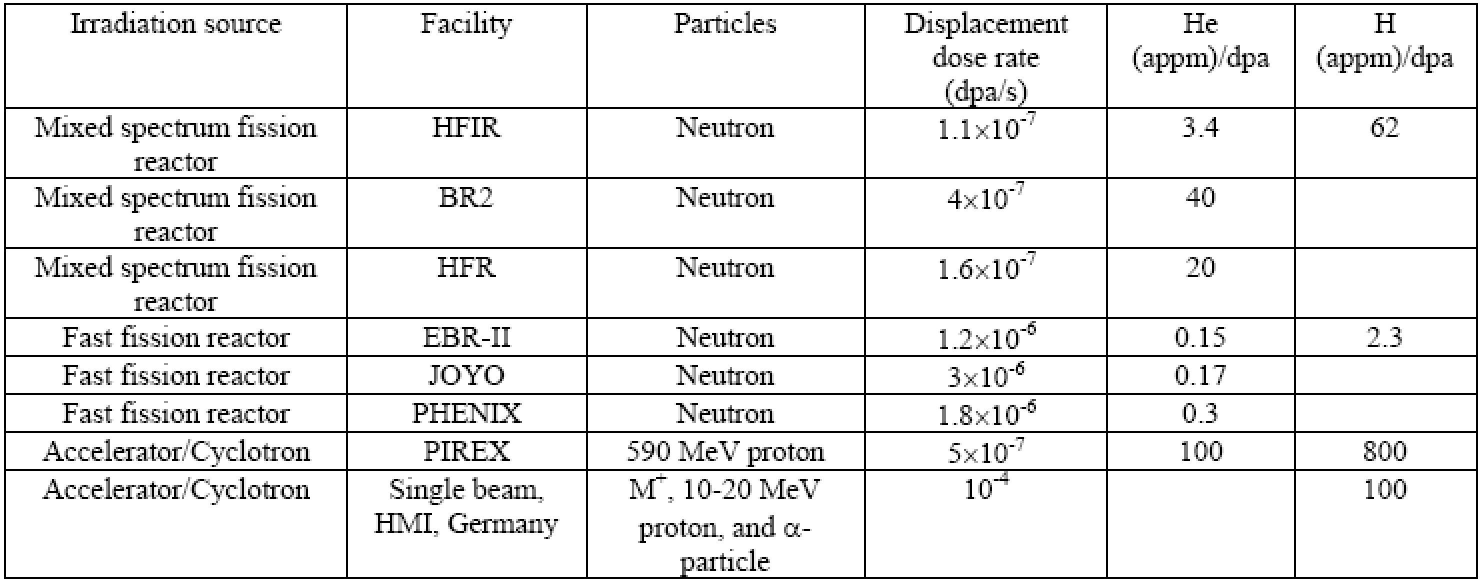
  - $\sigma_d(E)$ is the displacement cross section for an incident particle at an energy E,
  - t is the irradiation time,
  - $\varphi(E,t)$ is the fluence rate spectrum,
  - $\phi_{tot}(t)$ is the time-dependent fluence rate intensity.
- Irradiation-induced changes of material properties are measured as a function of dpa
- The extent of the radiation damage cannot be fully characterized by a single parameter
- Transmutation production rates (e.g. He/dpa, H/dpa) are important
- Radiation damage in terms of dpa and He/dpa, H/dpa under various types of irradiation environments can be calculated using theoretical models and computer codes and verified by experiments

Transmutation Products

- Nuclear transmutation reactions occur, producing He and H atoms and solid transmutation products
- Transmutation production rates (e.g. He/dpa, H/dpa) are important
- Radiation damage in terms of dpa and He/dpa, H/dpa under various types of irradiation environments can be calculated using theoretical models and computer codes and verified by experiments

Irradiation Sources

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



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)

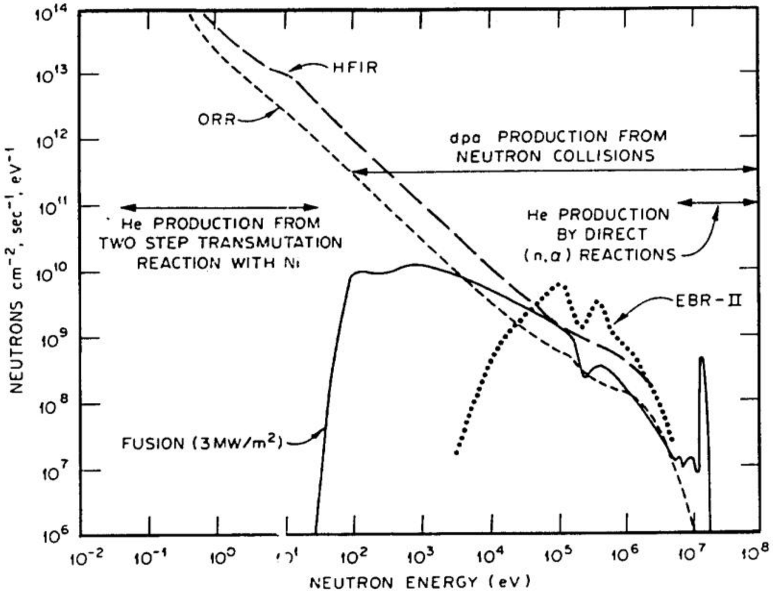
Damage correlation, extrapolation is necessary

Damage Correlation between Irradiation

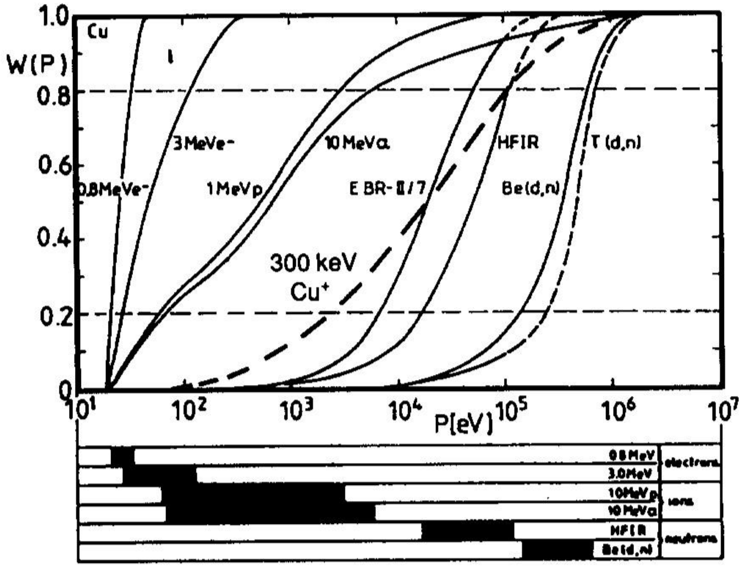
- Damage correlation parameters
  - Irradiation particle, e.g. protons vs. neutrons
  - Energy spectra
  - Flux or dose rate (dpa/s)
  - Fluence or dose (dpa)
  - Irradiation temperature
  - Transmutation (e.g. He, H)
  - Pulsed irradiation vs. continuous irradiation

Energy Spectra and Damage Production

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



Energy spectra in various nuclear reactors



Damage production by PKAs in copper by different irradiation sources

(Abbromeit 1994)

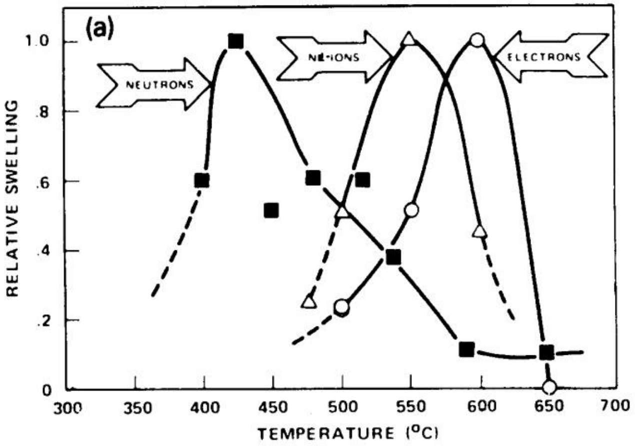
Defect Productions

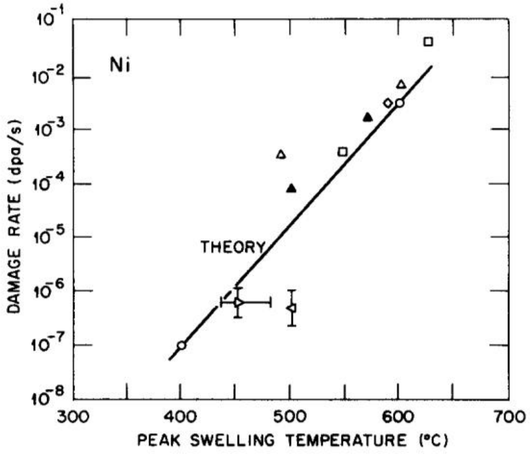
- Electron irradiations create single Frenkel defects (point defects of interstitials and vacancies)
- Low-energy protons and light ions create similar defects as electron irradiations
- Heavy ions and neutrons produce cascade displacement damage
- Fusion neutrons create significant subcascades with very high energies PKAs

Damage correlation by different types of irradiation should consider production of defects in single point defects and defect clusters

Displacement Dose Rate

- The dose rate for proton irradiations can be 2-3 orders of magnitude higher than neutron irradiation.
  - Thermal (or mixed spectrum) reactors: 10^{-7} dpa/s
  - Fast fission reactor: 10^{-6} dpa/s
  - Accelerators facilities: a wide range of dose rates, e.g. 10^{-3} dpa/s.
- Dose rate can significantly affect microstructural evolution and physical and mechanical properties

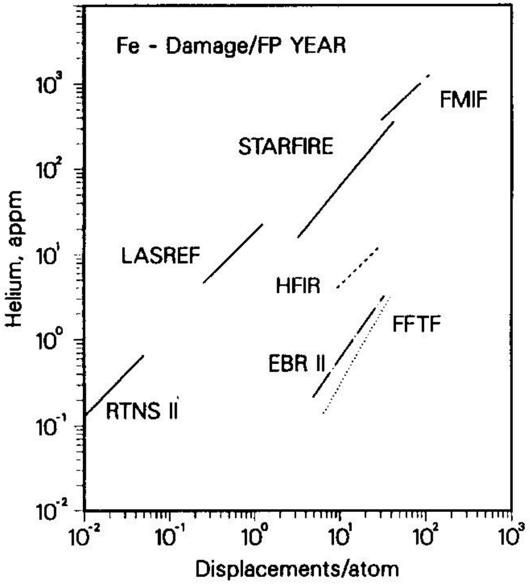




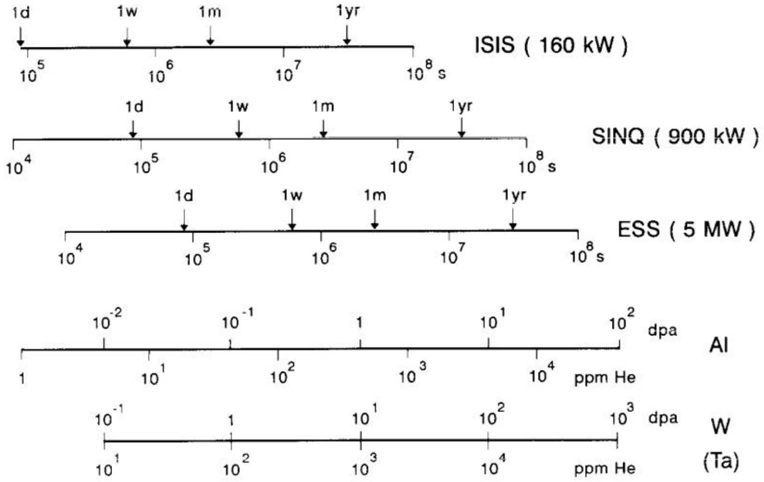
Dose rate plays a critical role in irradiation-induced swelling, irradiation creep, and solute segregation. The peak swelling temperature shifted as a function of dose rate. (Abromeit, Mansur 1994).

Transmutation Rates

- He and H gas atoms that can have pronounced effect on materials performance even at low concentrations
- The production rates of He, H can be exceptionally high under high energy proton irradiation compared to fission neutron irradiation



Comparison of displacement and He production in Fe in different irradiation facilities (Greenwood 1994)

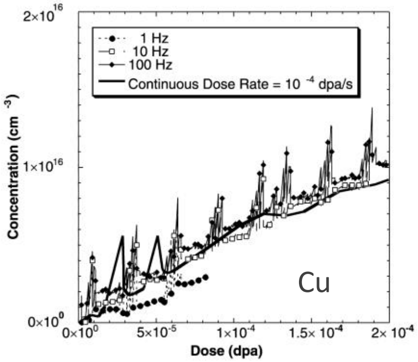


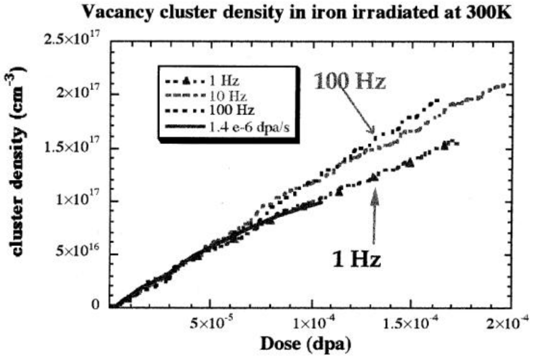
Displacement and helium production in Al and W, respectively as a function of service time for targets exposed to the proton beams of ISIS, SINQ and ESS

(Ullmaier and Carsughi 1995)

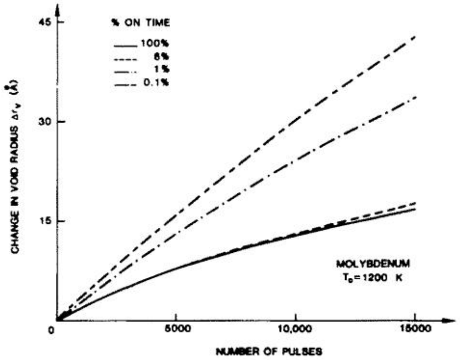
Pulsed Irradiation vs. Continuous Irradiation

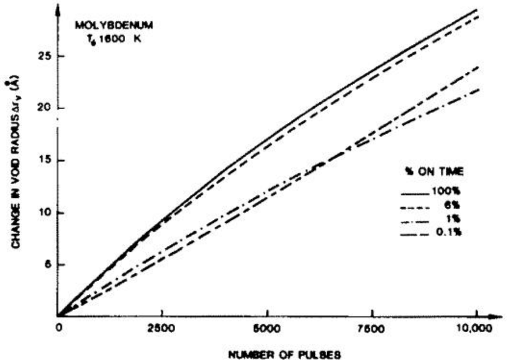
- Due to the pulsed nature of irradiation, the interplay of irradiation flux, temperature and pulse frequency can change the kinetics of irradiation damage accumulation compared to a steady-state continuous irradiation.





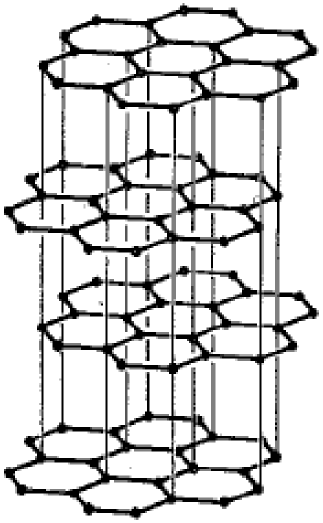
MD/kMC simulations showed evolution of radiation damage as a function of dose in Cu and Fe under pulsed irradiations and continuous irradiation (Caturla et al 2001)





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)

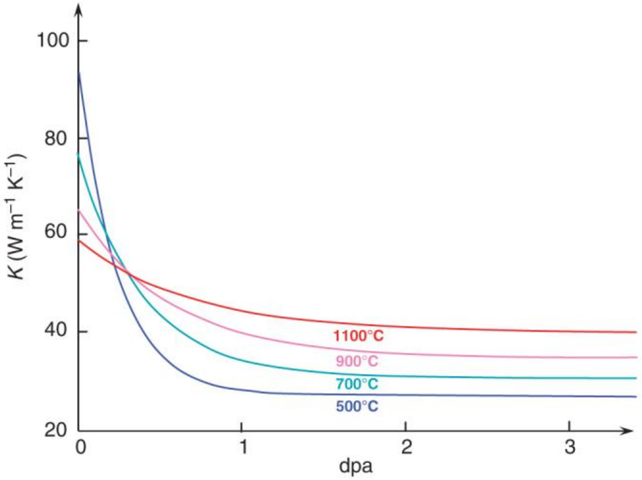
Radiation Effects in Graphite



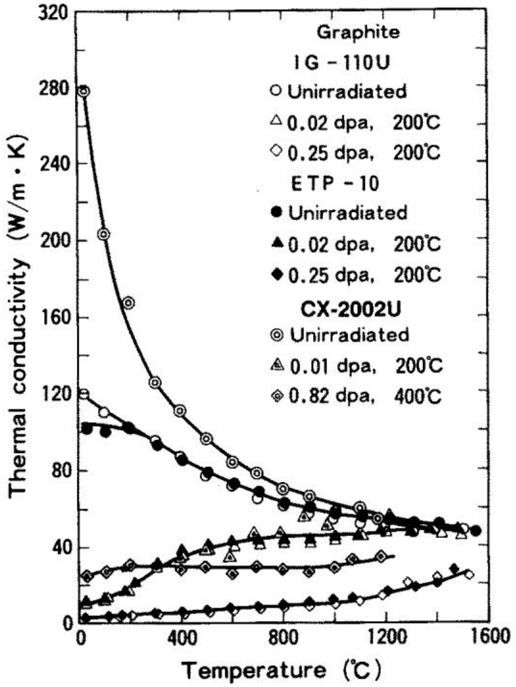
- Graphite is a lead candidate for the target design
- Neutron irradiation of graphite produces a large population of carbon interstitial atoms and vacancies, forming vacancies and interstitial clusters, dislocation loops or new graphitic planes
  - Interstitials move between layer planes and are mobile at temperatures as low as 70 K
  - Vacancies move in the layer planes and are mobile only at temperatures above 1000 K
  - Radiation effects in graphite are determined by the formation of defect structure.
- Radiation effect studies in graphite have primarily focused on displacement damage rather than helium generation; data on dose rate effects are scarce

Thermo-physical Properties of Irradiated Graphite

- Neutron irradiation reduces thermal conductivity of all types of graphite



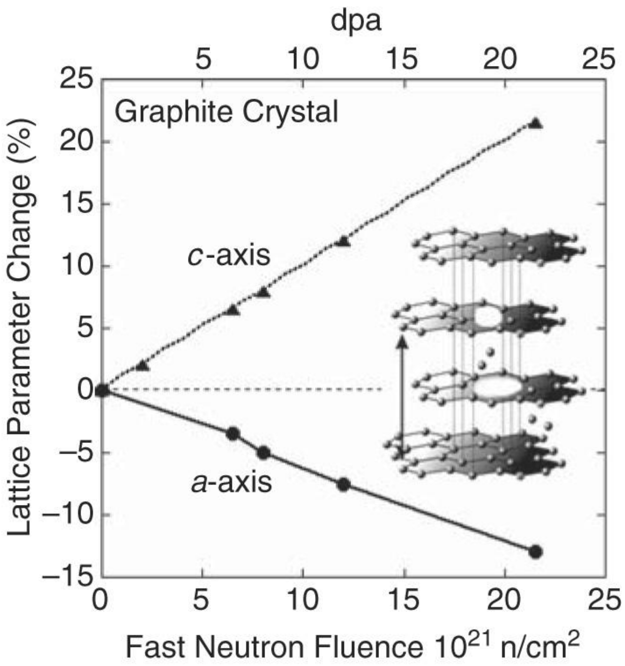
Thermal conductivity of irradiated polygranular graphite starts to decrease at very low neutron doses (10^{-3} dpa); As dose increases, the reduction in thermal conductivity tends to saturate (Bonal et al 2009)



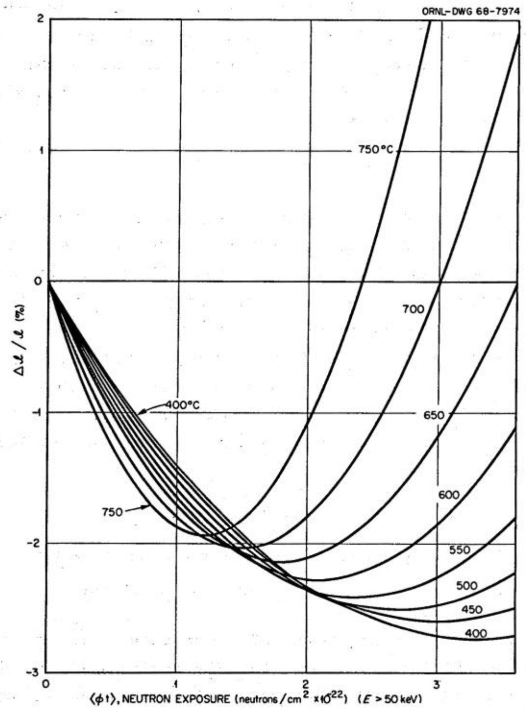
Thermal conductivity as a function of test temperature for neutron-irradiated graphite (Maruyama and Harayama 1992)

Irradiation-induced Dimensional Changes

- Neutron irradiation of graphite leads to significant dimensional changes over a wide range of irradiation conditions as a result of swelling and densification.



Irradiation-induced anisotropic dimensional change in graphite irradiated with neutrons at 710°C. The crystal expands in the c-axis direction and shrinks in the a-axis direction (Snead 2009).



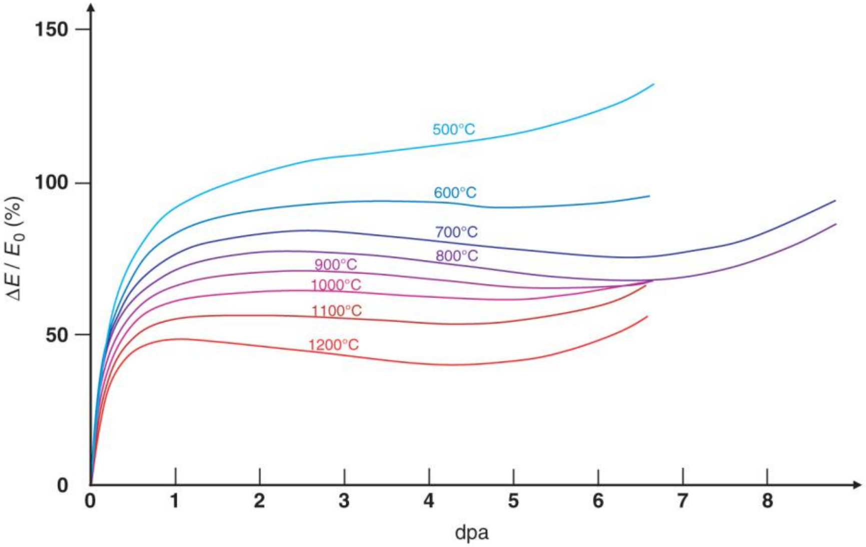
Neutron irradiation of polygranular graphite causes shrinkage at low doses and expansion at high doses. The threshold for the dimensional reversal is lower at higher irradiation temperatures (Kasten et al 1969).

Irradiation Creep

- Graphite experiences a time-dependent plastic deformation under irradiation and stress - irradiation creep
- Irradiation creep in nuclear graphite can occur at temperatures as low as 100°C
- Deformation caused by irradiation creep is much higher than thermal creep in graphite
- Irradiation creep in graphite relaxes stresses generated by gradients of irradiation-induced dimensional changes within components, and has significant impact on graphite cracking

Mechanical Strength of Irradiated Graphite

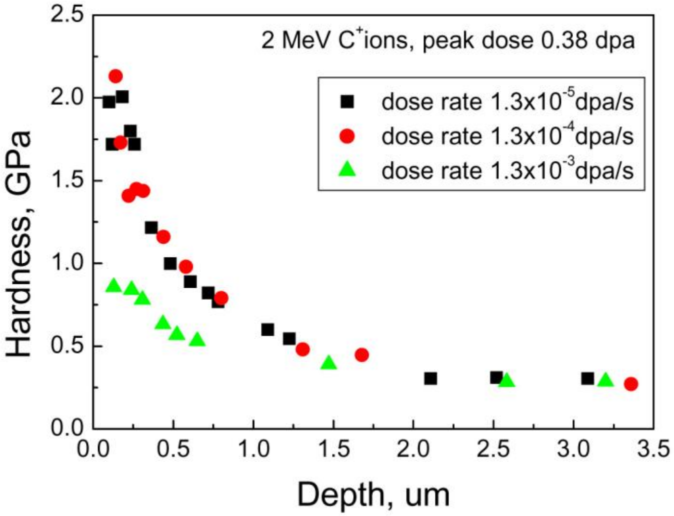
- Neutron irradiation significantly changes the mechanical properties of graphite.
- The strength of graphite increases under irradiation.
- Neutron irradiation initially increases the Young's modulus and mechanical strength substantially at low doses. This increase in the Young's modulus and mechanical strength is smaller at higher irradiation temperatures.



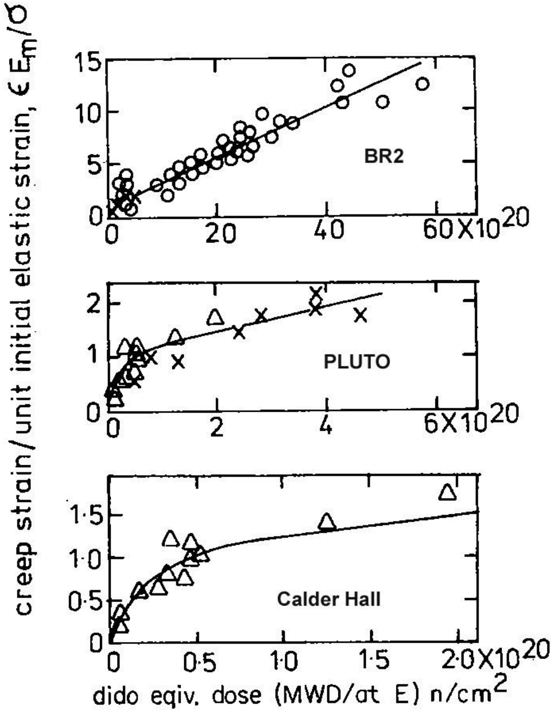
Effects of neutron irradiation on the Young's modulus of pitch coke graphite (Bonal et al 2009)

Dose Rate Effects in Graphite

- Very few studies have been done to investigate the effect of dose rate on radiation-induced property changes in graphite.



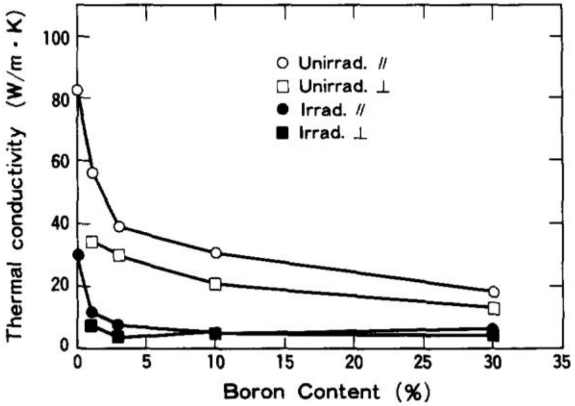
Increase in hardness increased with decreased dose rate in isotropic nuclear graphite irradiated to 0.38 dpa by three different dose rates with 2 MeV C ions (Chi et al 2004)



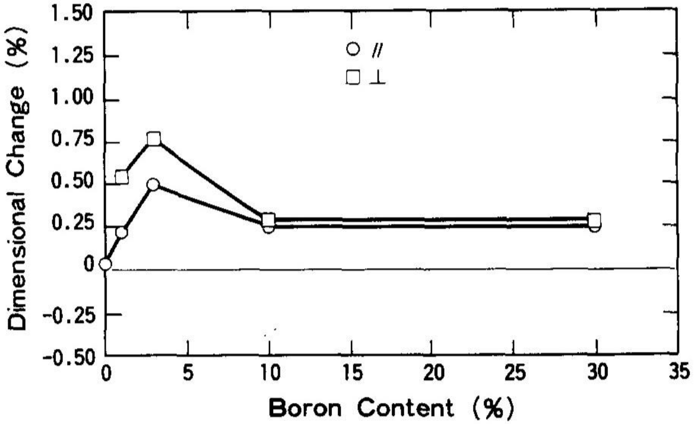
Comparison of irradiation creep data on graphite obtained in three different reactors, BR2, PLUTO and Calder Hall showed insignificant effect of dose rate on irradiation creep (Kelly and Brockklehurst 1970)

Effect of Helium in Irradiated Graphite

- High energy protons produce He and H to a much greater degree than fission neutrons
- Limited studies of high-level helium effects in graphite have been done in the past, primarily for fusion reactor applications
- Helium bubble formation and flaking observed in He-implanted ion-irradiated graphite
- Significant reduction in thermal conductivity and swelling in boron-doped graphite after neutron irradiation



Changes in thermal conductivity after neutron irradiation in bulk-boronized graphite (Maruyama and Harayama 1992)



Dimensional changes in bulk-boronized graphite by neutron irradiation (Maruyama and Harayama 1992)

Summary

- Radiation damage by high energy protons includes displacement cascades 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