Fundamental Research on Ceramics for Fusion Energy

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Fusion Materials Sciences Peer Review Meeting

Santa Barbara, CA

August 27-28, 2001

Introduction

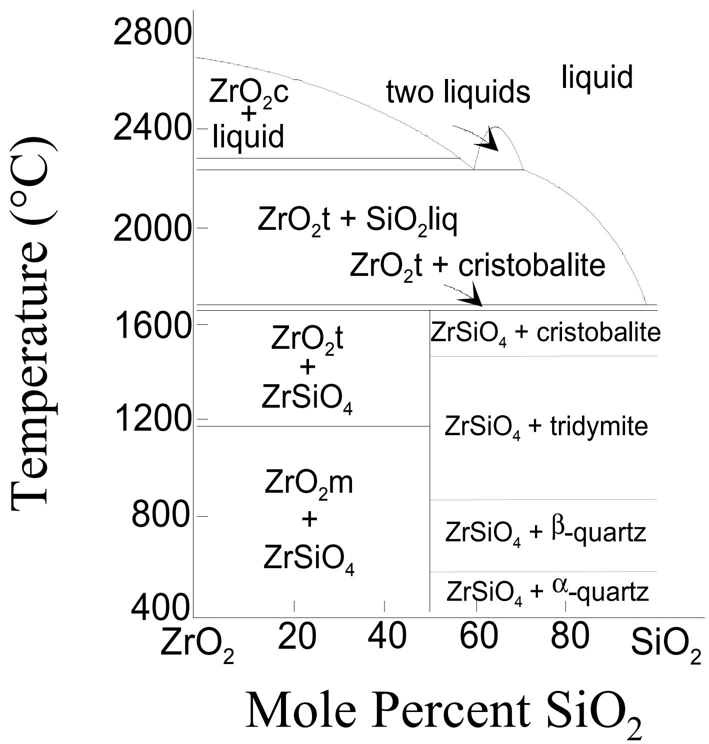
- Present-day and planned fusion power systems utilize ceramic materials, taking advantage of their unique range in properties

- Examples include:
  - Diagnostic Systems
  - Electrical Breaks and Insulator Coatings
  - RF Feedthroughs and Optical Windows
  - First Wall Tiles
  - Structural Components

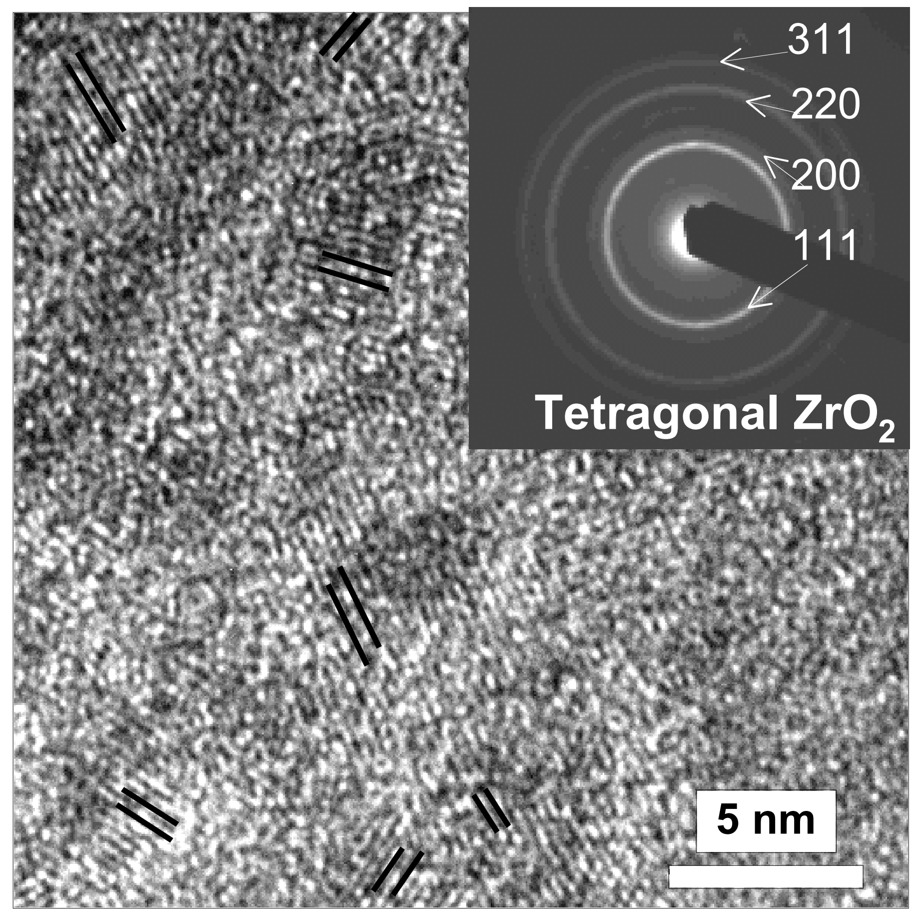
- Phenomena recently being studied include:
  - Point defect migration energies
  - Microstructure and property changes due to ionizing and displacive damage
  - Thermal transport
  - Irradiation-induced strain, amorphization and lattice recovery
  - Simulation of high helium levels on thermophysical properties

Experimental evidence for nanoscale melting during atomic collisions has been obtained

ZrO_2-SiO_2 Phase Diagram



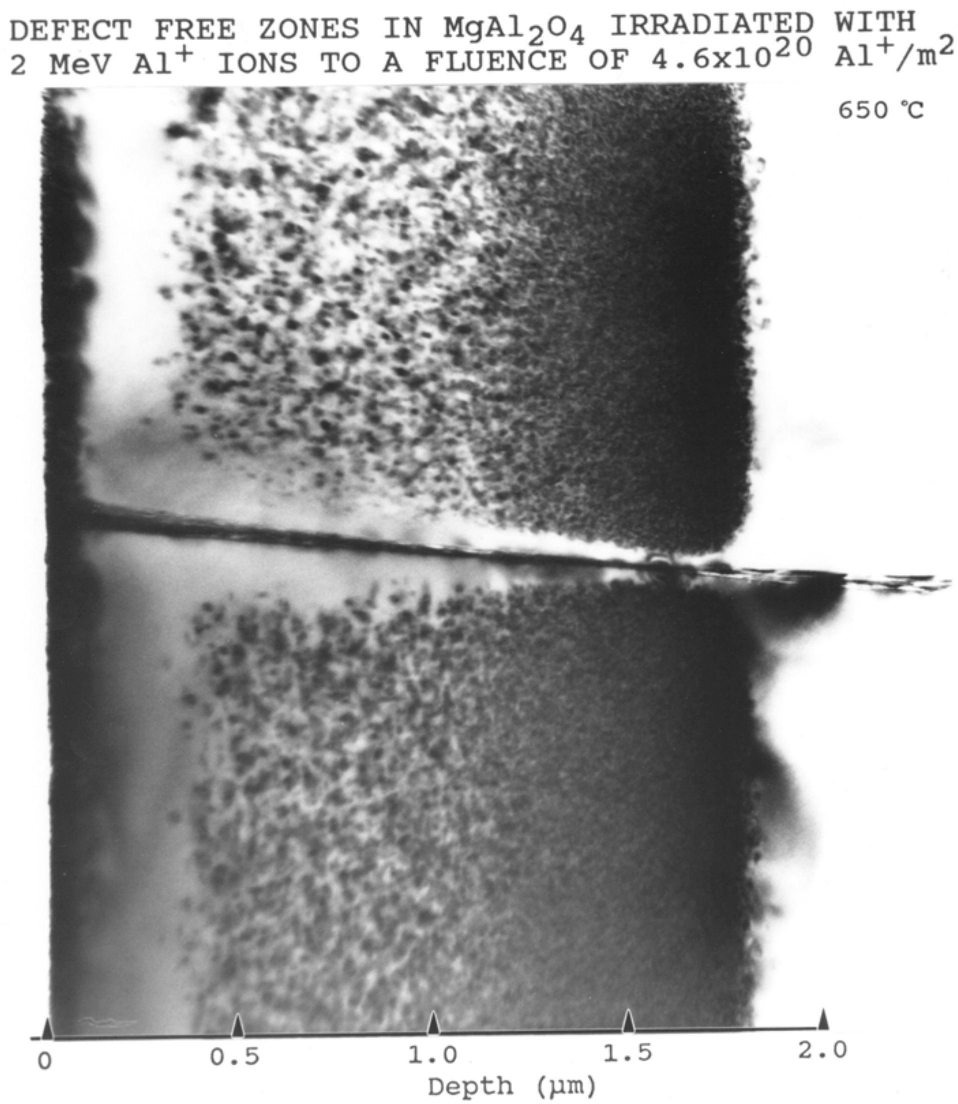
Microstructure of Zircon Irradiated at 800 °C

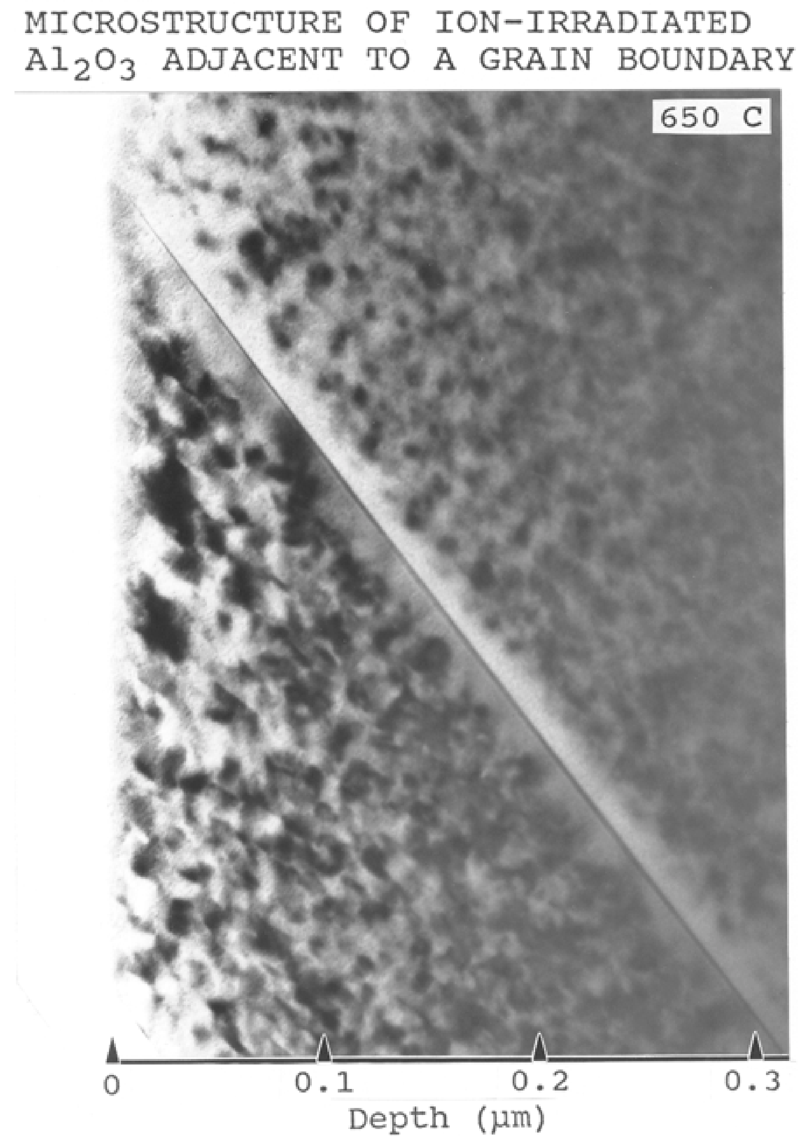


Nature, vol. 395, Sept 5. 1998, p. 56

Estimation of Interstitial Migration Energies in Ceramics

- Dislocation loops do not form near grain boundaries and free surface due to insufficient point defect supersaturation





Estimation of Interstitial Migration Energies in Ceramics

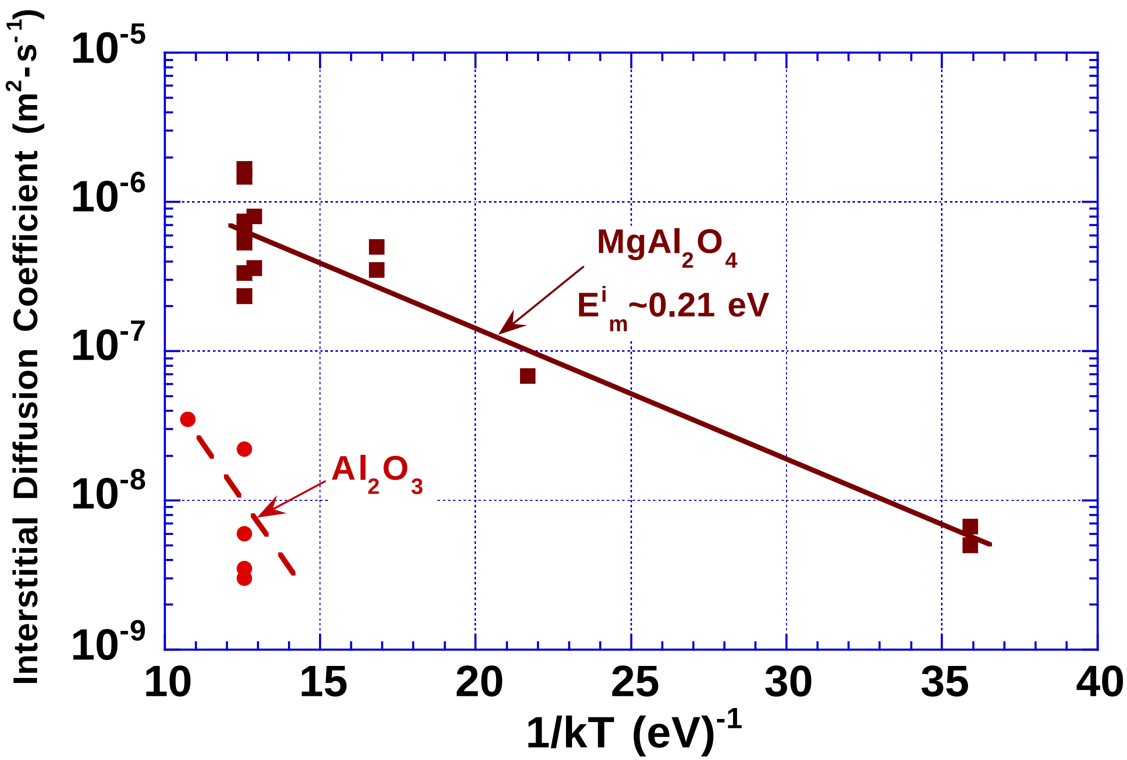
•Solve steady-state equations:

$$D_i \frac{d^2 C_i}{dx^2} - \alpha C_i C_v - D_i C_i C_s + P = 0$$

$$D_v \frac{d^2 C_v}{dx^2} - \alpha C_i C_v - D_v C_v C_s + P = 0$$

•For sink-dominant conditions, the defect-free zone near surfaces, g.b.'s is

$$L = \frac{1}{\sqrt{C_s}} \ln \frac{C_i^\infty}{C_i^\infty - C_i^{crit}}$$



Using $C_i^\infty = \frac{P}{D_i C_s}$

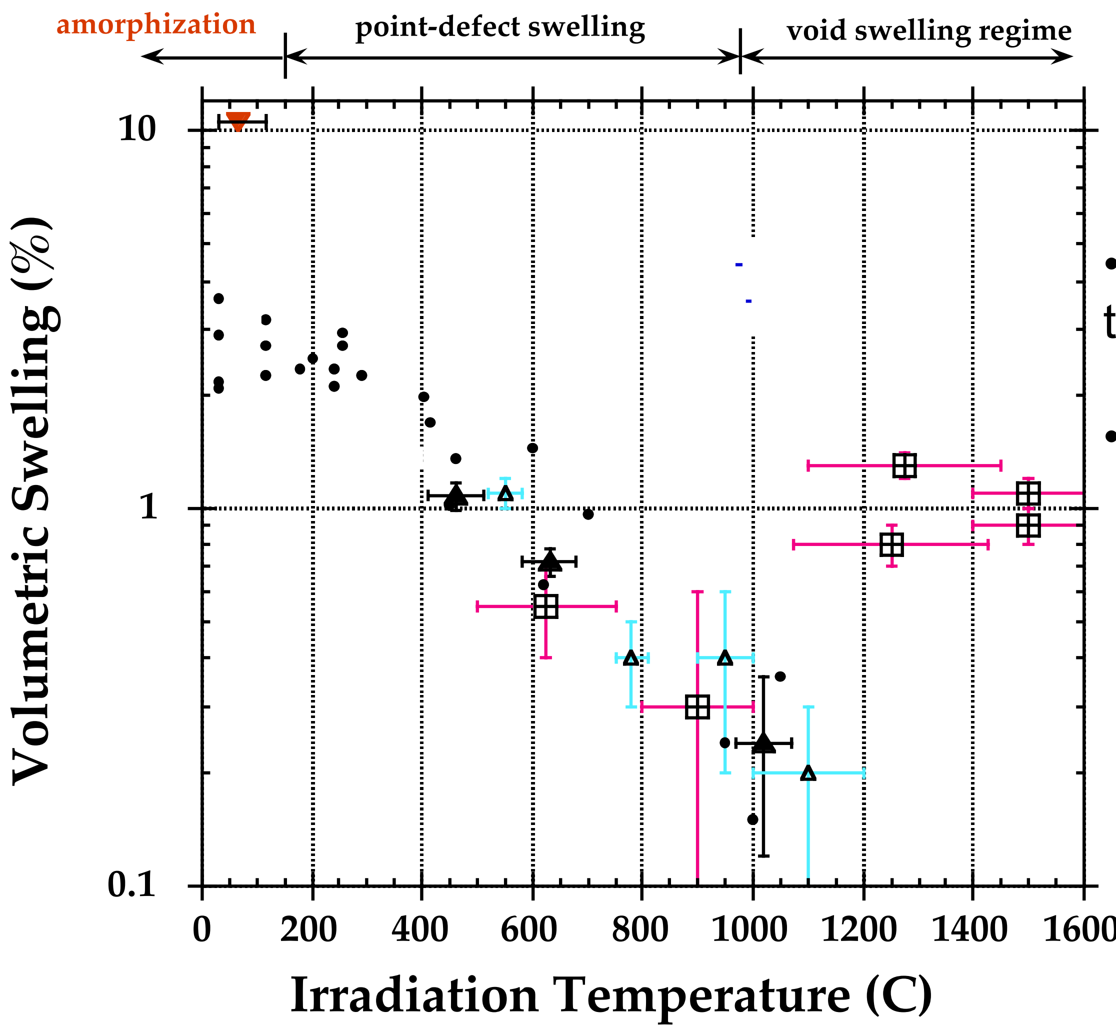
and assuming $C_i^\infty >> C_i^{crit}$:

$$D_i = \frac{L\ P}{C_i^{crit}\sqrt{C_s}}$$

$E_i^m \approx 0.21\ eV$ ($MgAl_2O_4$)

$E_i^m \approx 0.6\ eV$ (Al_2O_3)

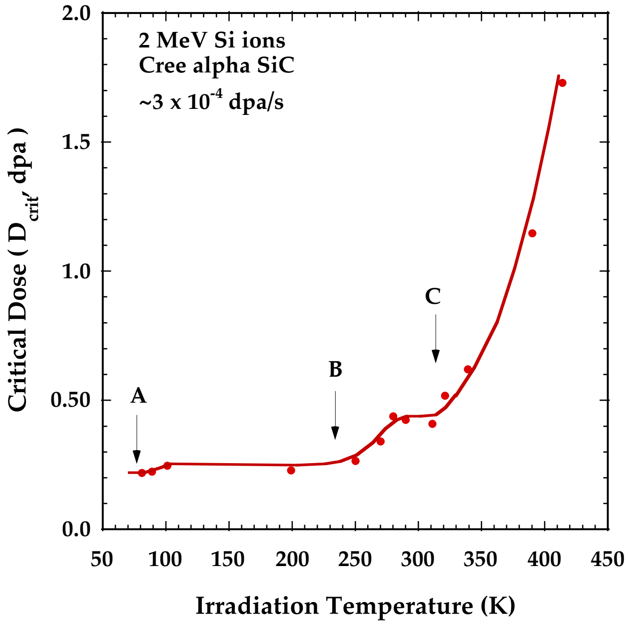
Irradiation Induced Strain and Amorphization

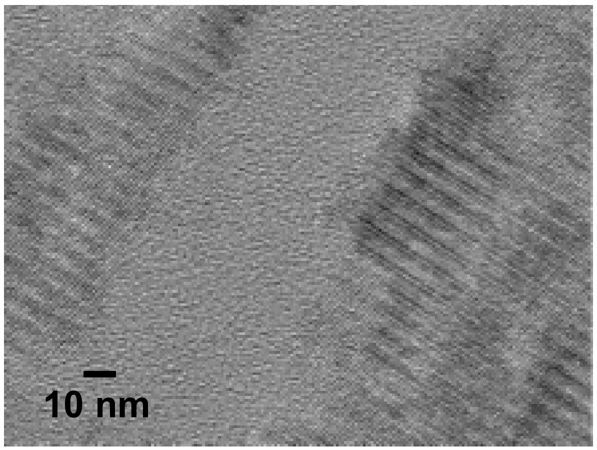


- Ceramics undergo swelling due to radiation-induced interstitial strain
- SiC swelling regimes:
  - Amorphization (T< 150°C)
  - Point Defect Swelling (<1000°C)
  - Void Swelling (?)

SiC Amorphization

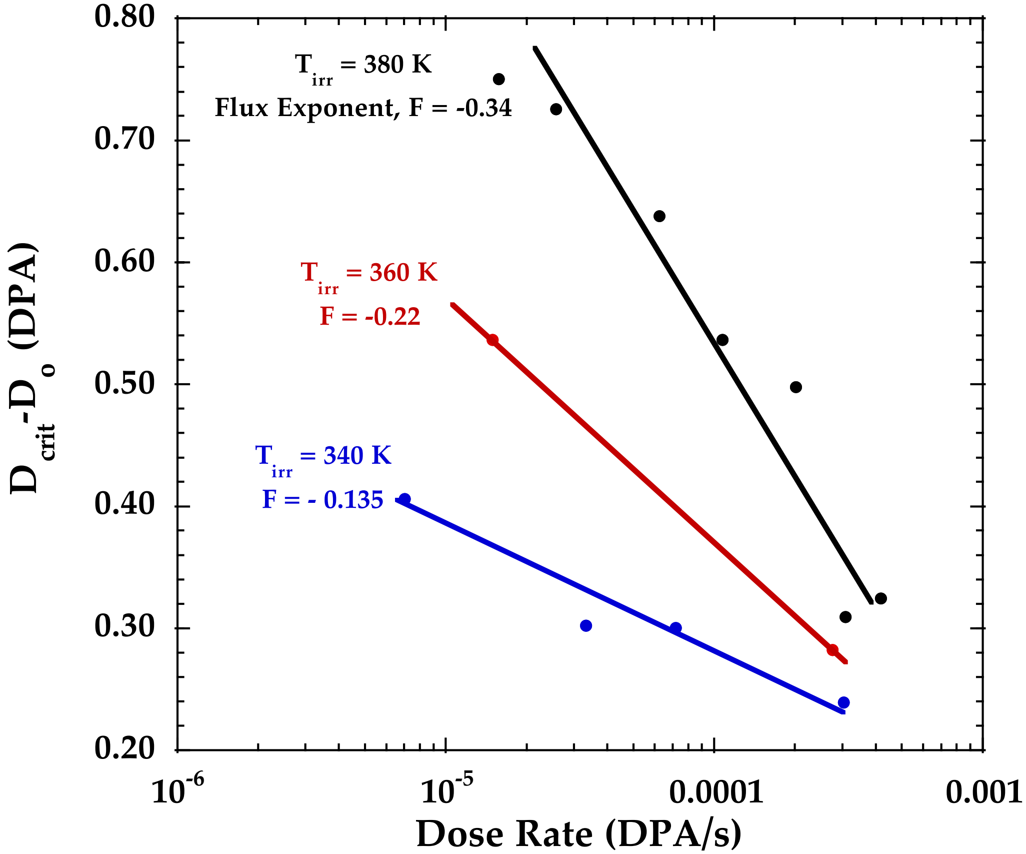
3 recovery substages are observed below 350 K





Flux dependence shows recovery stages are not associated with long range point defect migration (F<0.5 T< 380 K)

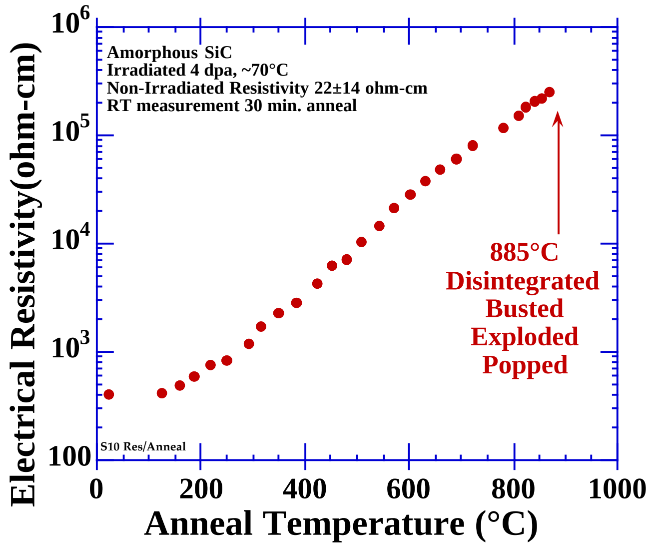
$$D_{crit}-D_o\ (DPA) = (A\Phi^F)e^{(-Em/2kT)}$$

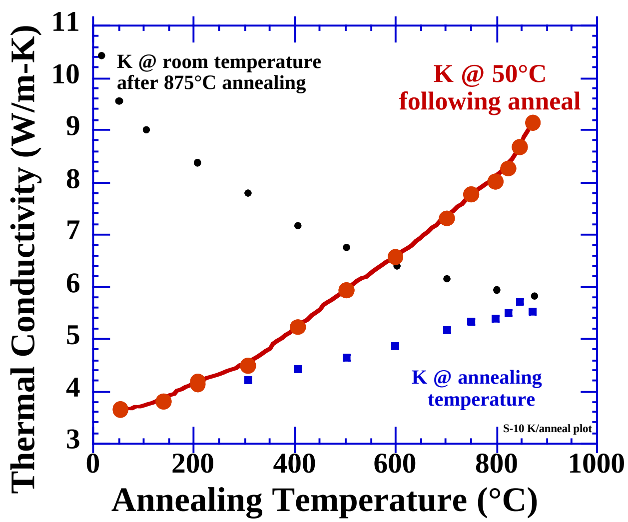


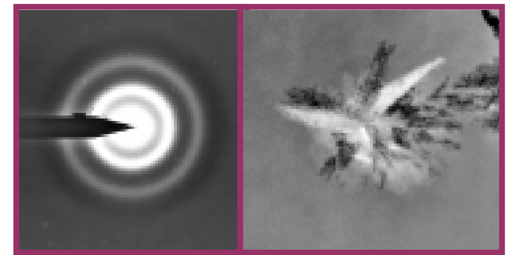
Unlike many ceramics, results imply immobility of vacancies and interstitials in SiC for T< 100˚C

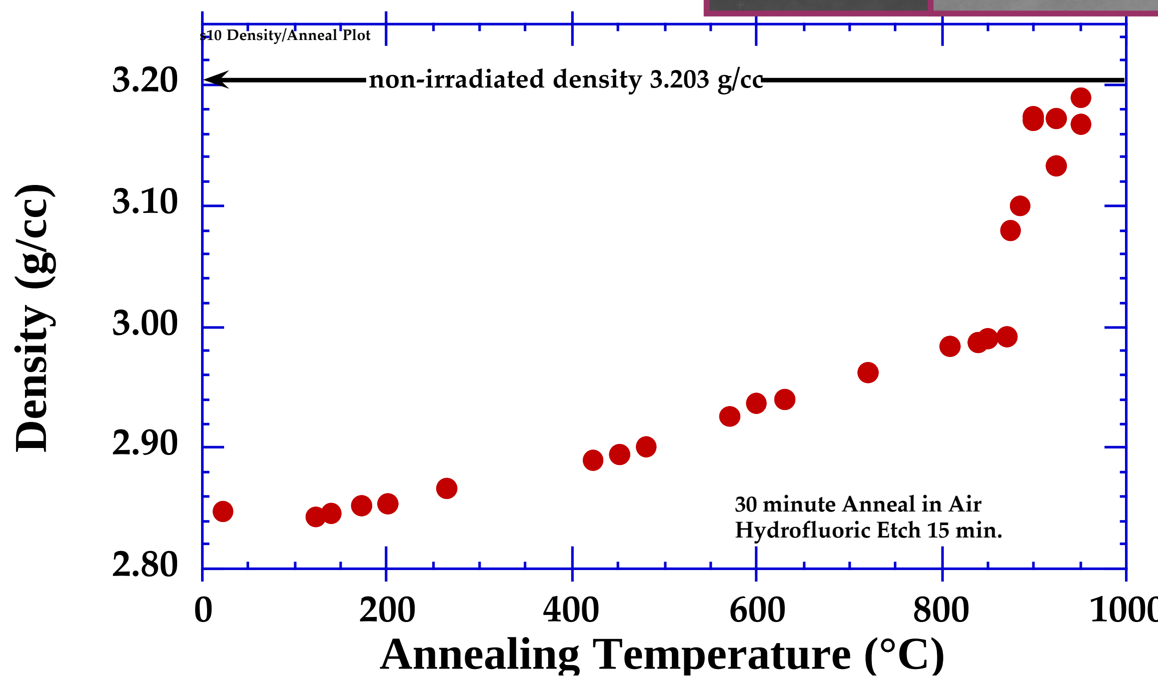
Effect of annealing on the properties of bulk amorphous SiC

- Amorphization causes large changes
  - --> density change, -10.8 %
  - --> hardness change, -46 %
  - --> elastic modulus change, -45 %









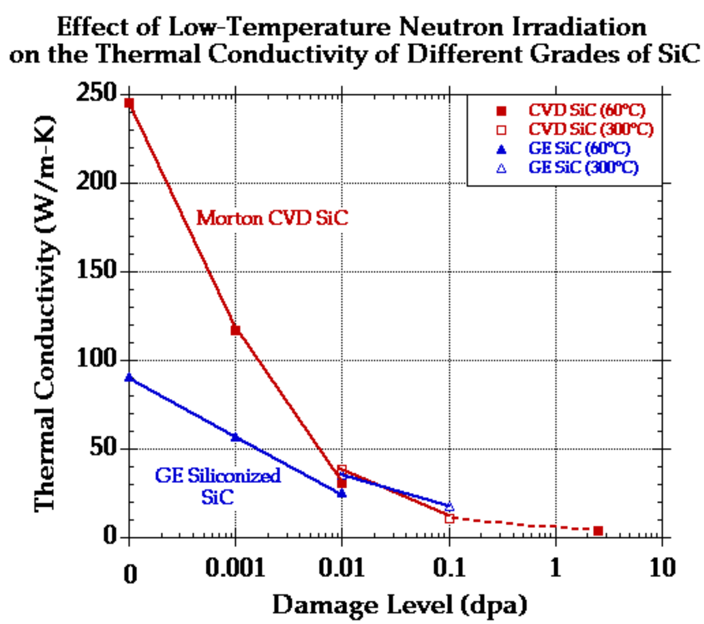
Physics of phonon transport & scattering are being investigated in neutron-irradiated ceramics

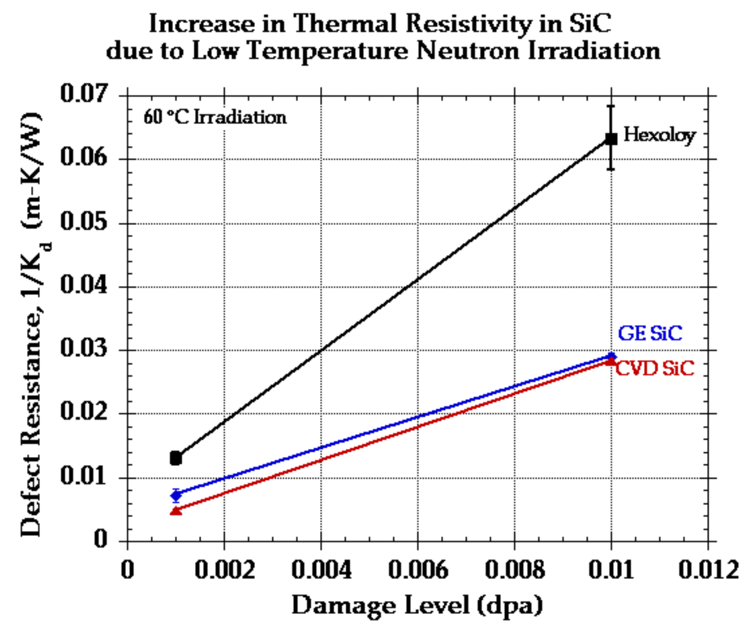
$$\left[K(T)\right]^{-1} = \left[\frac{1}{K_u(T)} + \frac{1}{K_{gb}(T)} + \frac{1}{K_{d0}} + \frac{1}{K_{rd}}\right]$$

Thermal resistance of different phonon scattering centers can be simply added if their characteristic phonon interaction frequencies are well-separated from one another

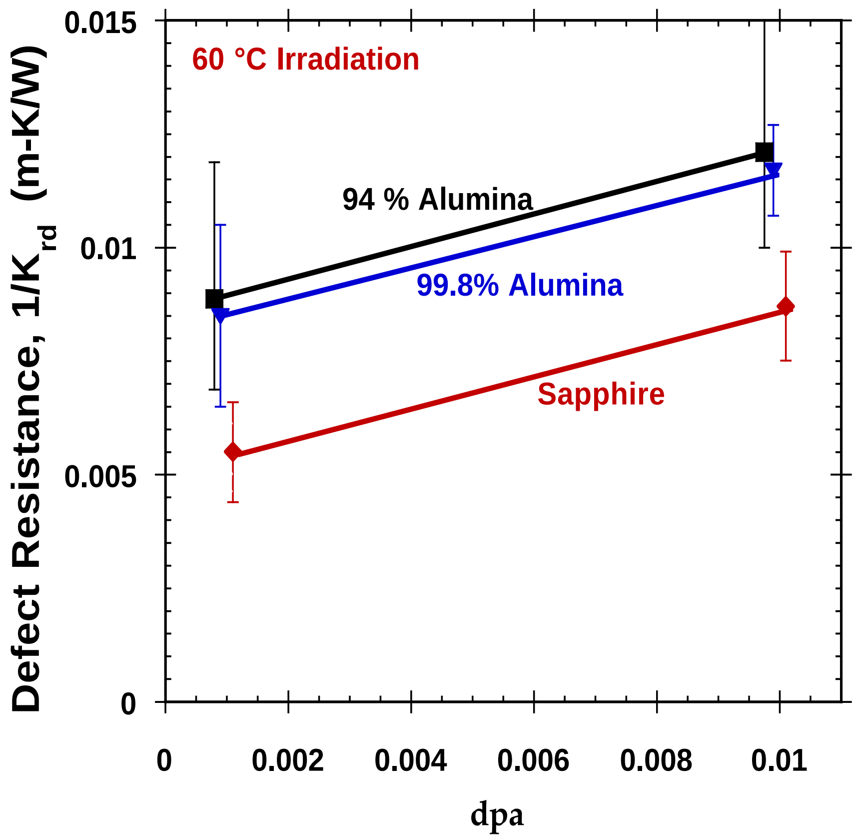
$$\frac{K_{irr}}{K_{unirr}} = \left(\frac{2h\upsilon^2}{18\pi^2\Omega\Theta_D K_{unirr} C_v}\right)^{1/2} \tan^{-1}\left(\frac{2h\upsilon^2}{18\pi^2\Omega\Theta_D K_{unirr} C_v}\right)^{-1/2}$$

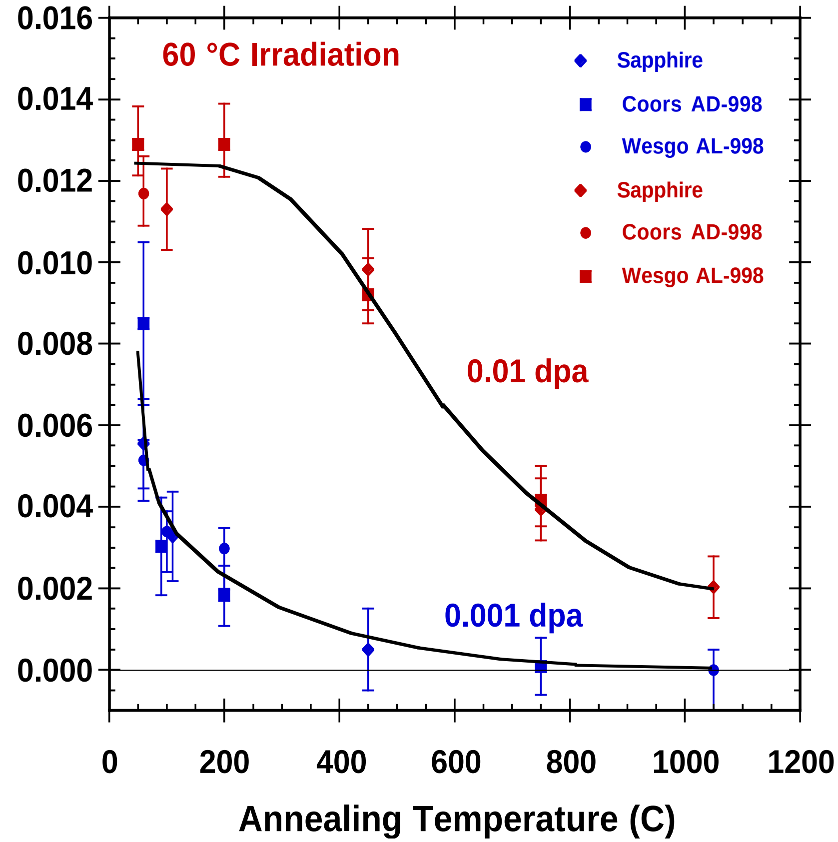
Thermal resistance due to radiation-induced defects (vacancies, dislocation loops, etc.) is proportional to their concentration



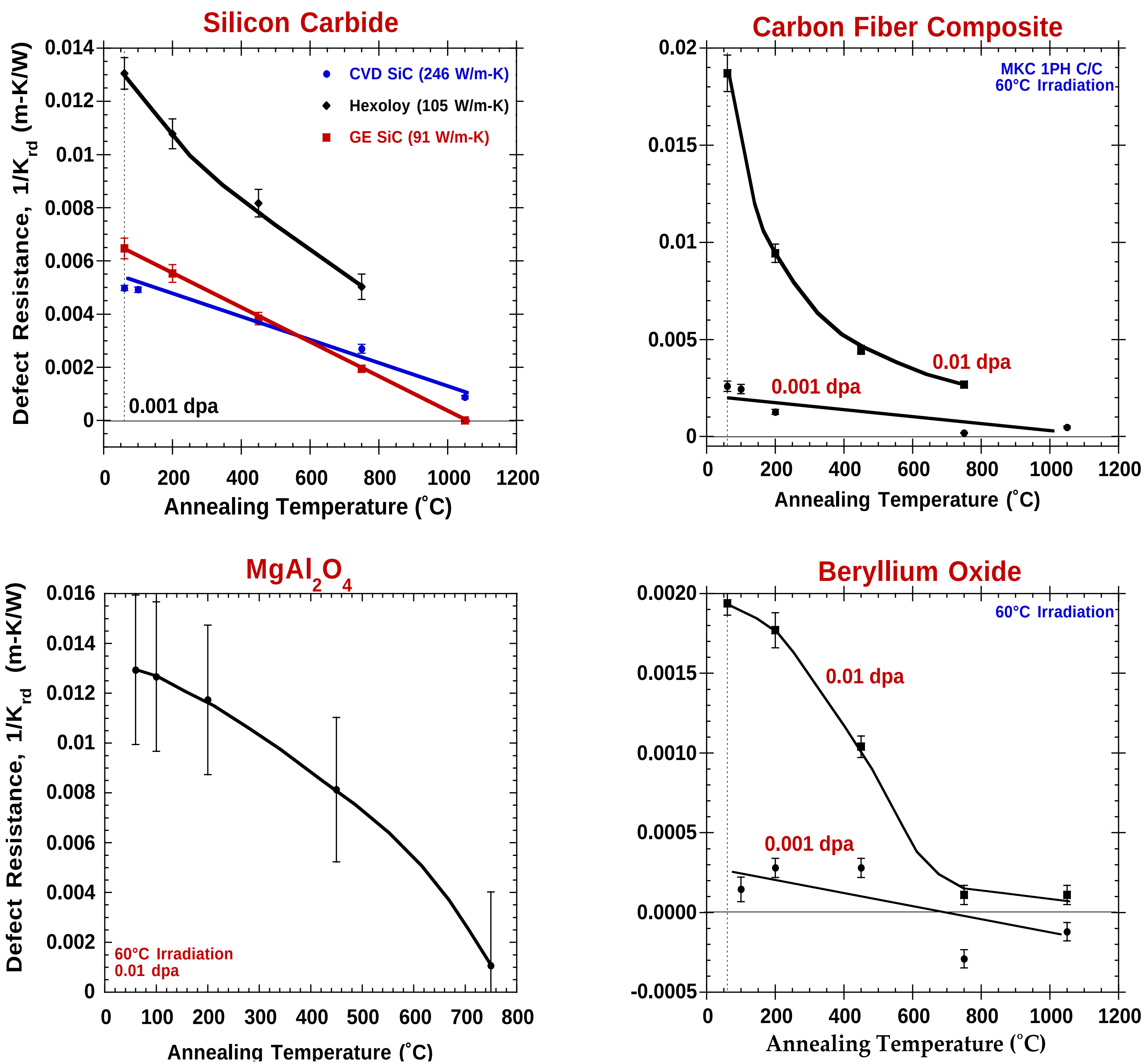


Insight into Defect Production and Thermal Stability





- Resistance is sublinear with dose;
  --> $1/K_{rd}$ increases by 2.5 times as dose is increased by 10 times
- Defects are more difficult to anneal as dose is increased.



Silicon Carbide
CVD SiC (246 W/m-K)
Hexoloy (105 W/m-K)
GE SiC (91 W/m-K)
0.001 dpa
Defect Resistance, 1/K_rd (m-K/W)
Annealing Temperature (˚C)
Carbon Fiber Composite
MKC 1PH C/C
60°C Irradiation
0.01 dpa
0.001 dpa
Annealing Temperature (˚C)
MgAl2O4
60°C Irradiation
0.01 dpa
Defect Resistance, 1/K_rd (m-K/W)
Annealing Temperature (˚C)
Beryllium Oxide
60°C Irradiation
0.01 dpa
0.001 dpa
Annealing Temperature (˚C)

Insight into Thermal and Defect Processes : Alumina

- $1/K_{rd}$ can be broken into vacancy, loop (and void) terms.

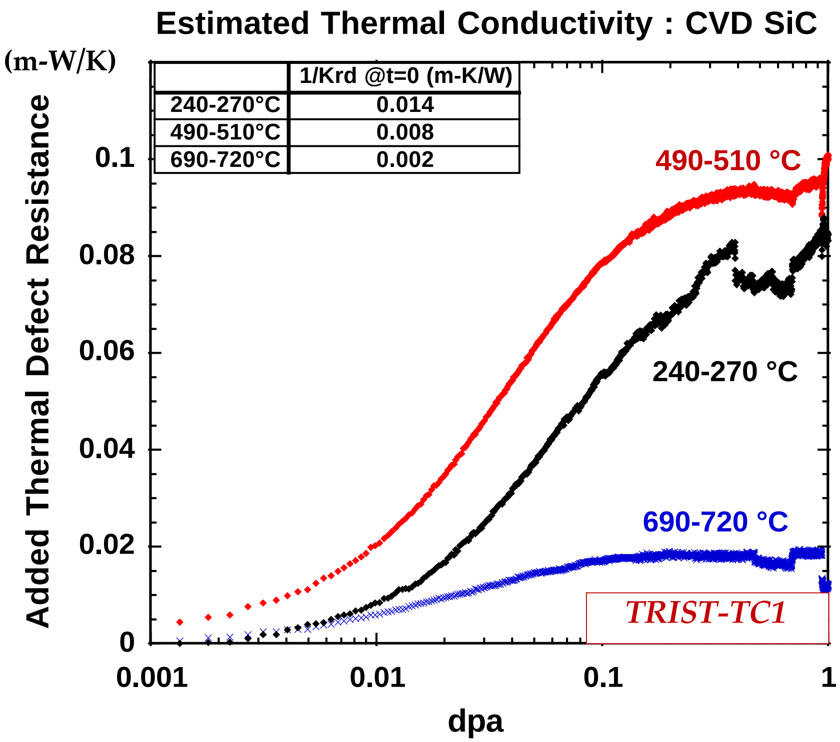
$$\frac{1}{K_{rd\ vac}} = \left(\frac{3\pi}{2K_B}\right)\left(\frac{\omega}{v^2}\right)C_{vac} \quad ; \quad \frac{1}{K_{rd\ loop}} \approx \frac{h^2 R^2 n_{loop}}{K_B}$$

- Following this analysis, **maximum** vacancy concentration can be calculated and compared with optical F-center measurements.

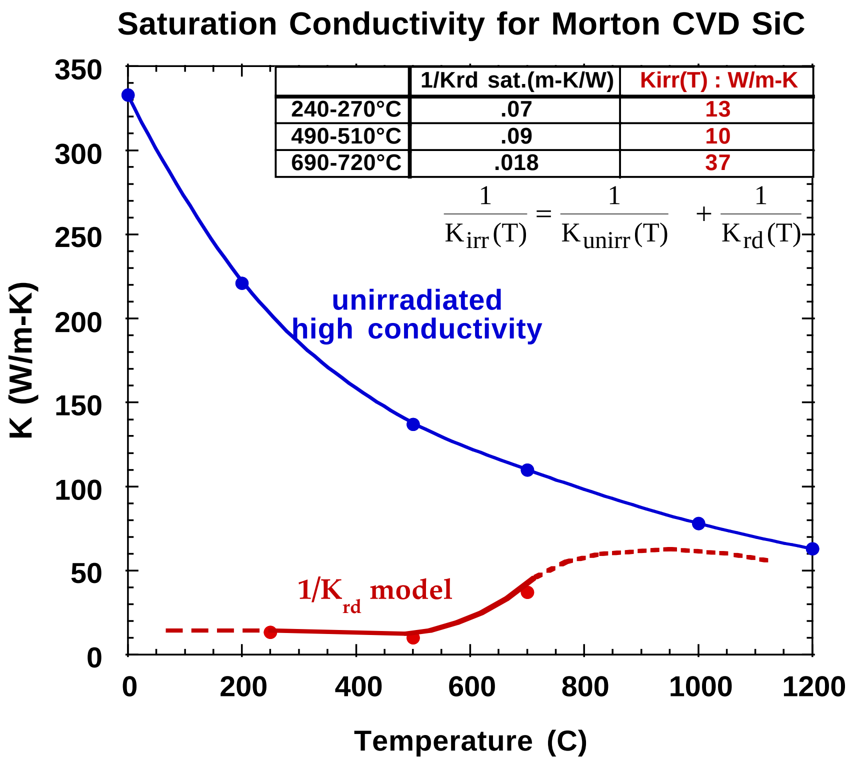
Vacancy (vppm) Concentration	0.001 dpa	0.01 dpa
From $1/K_{rd}$	1000	2000
From F+ Center (Atobe-87)	9	26

- Large discrepancy indicates that majority of thermal conductivity degradation in alumina (T_{irr} = 60°C) is dominated by loops. This is reinforced by increased difficulty in annealing of defects at higher doses.

Thermal Defect Resistance for Predicting Conductivity



	1/Krd @t=0 (m-K/W)
240-270°C	0.014
490-510°C	0.008
690-720°C	0.002



	1/Krd sat.(m-K/W)	Kirr(T) : W/m-K
240-270°C	.07	13
490-510°C	.09	10
690-720°C	.018	37

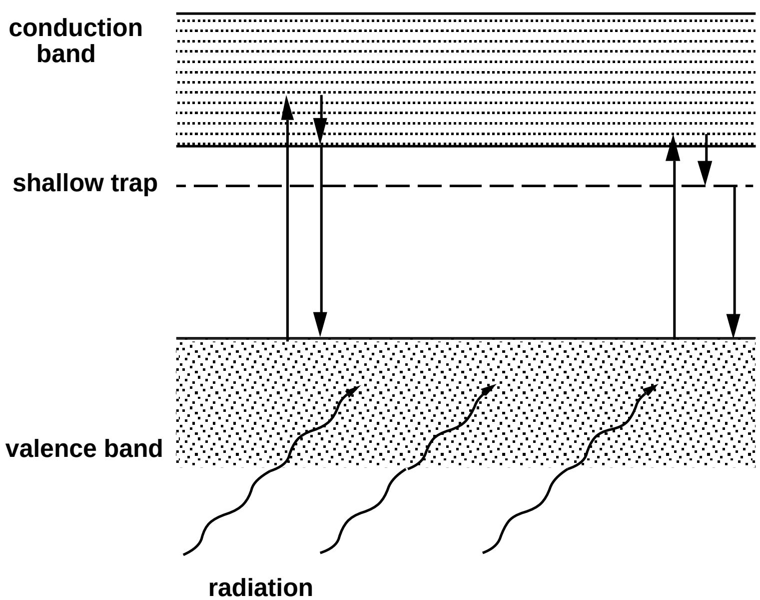
$$\frac{1}{K_{irr}(T)} = \frac{1}{K_{unirr}(T)} + \frac{1}{K_{rd}(T)}$$

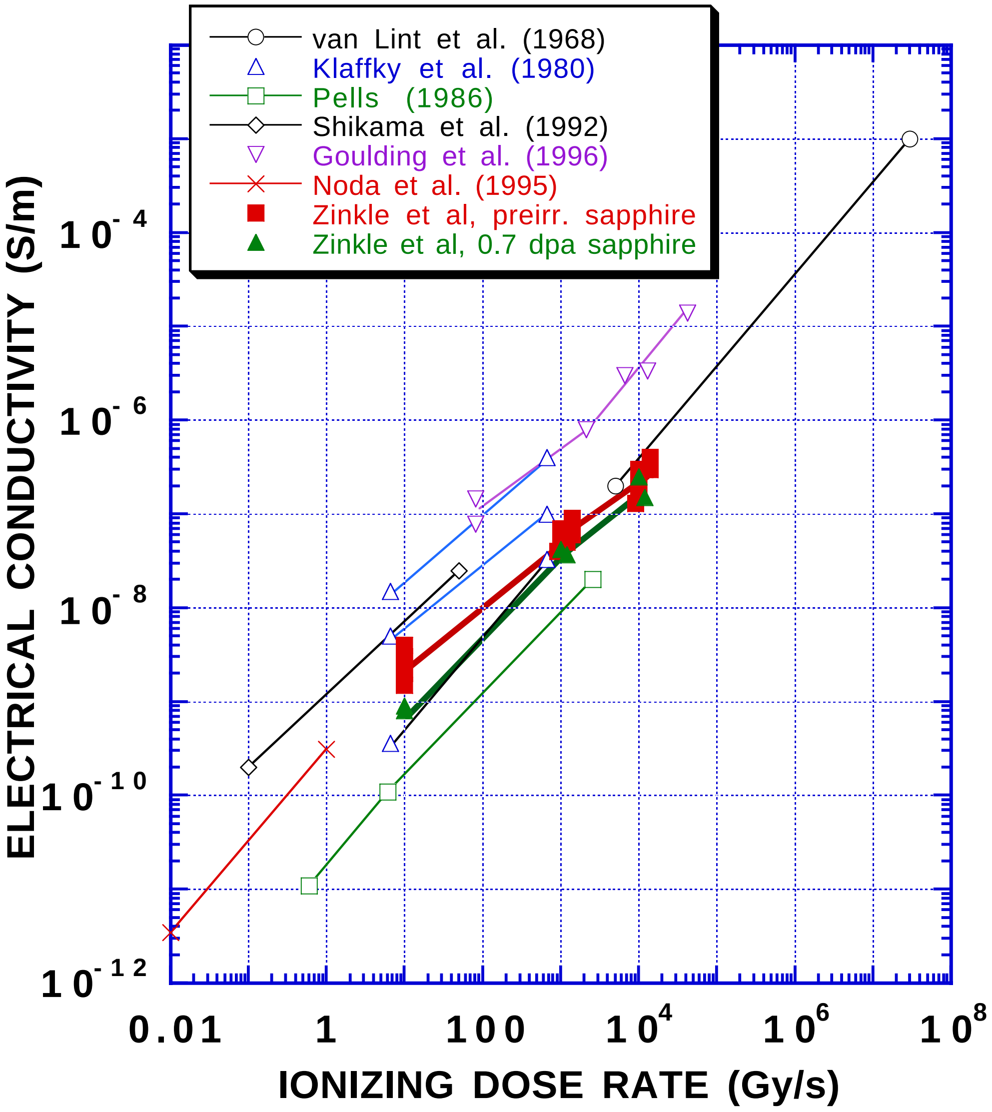
- Maximum thermal conductivity can be estimated for any material based on $1/K_{rd}$ measured from an “ideal” material.
- Maximum irradiated thermal conductivity for SiC is estimated to be ~ 10 W/m-K at 500°C, ~37 W/m-K at 700°C.

Ionizing Radiation can induce myriad effects in ceramics

- **Radiation-induced increases in electrical conductivity**
- **Defect annealing and coalescence (ionization-induced diffusion)**
  - **Athermal defect migration is possible in some materials**
- **Defect production**
  - **Radiolysis (SiO_2, alkali halides)**
  - **Ion track damage ("swift heavy ions")**

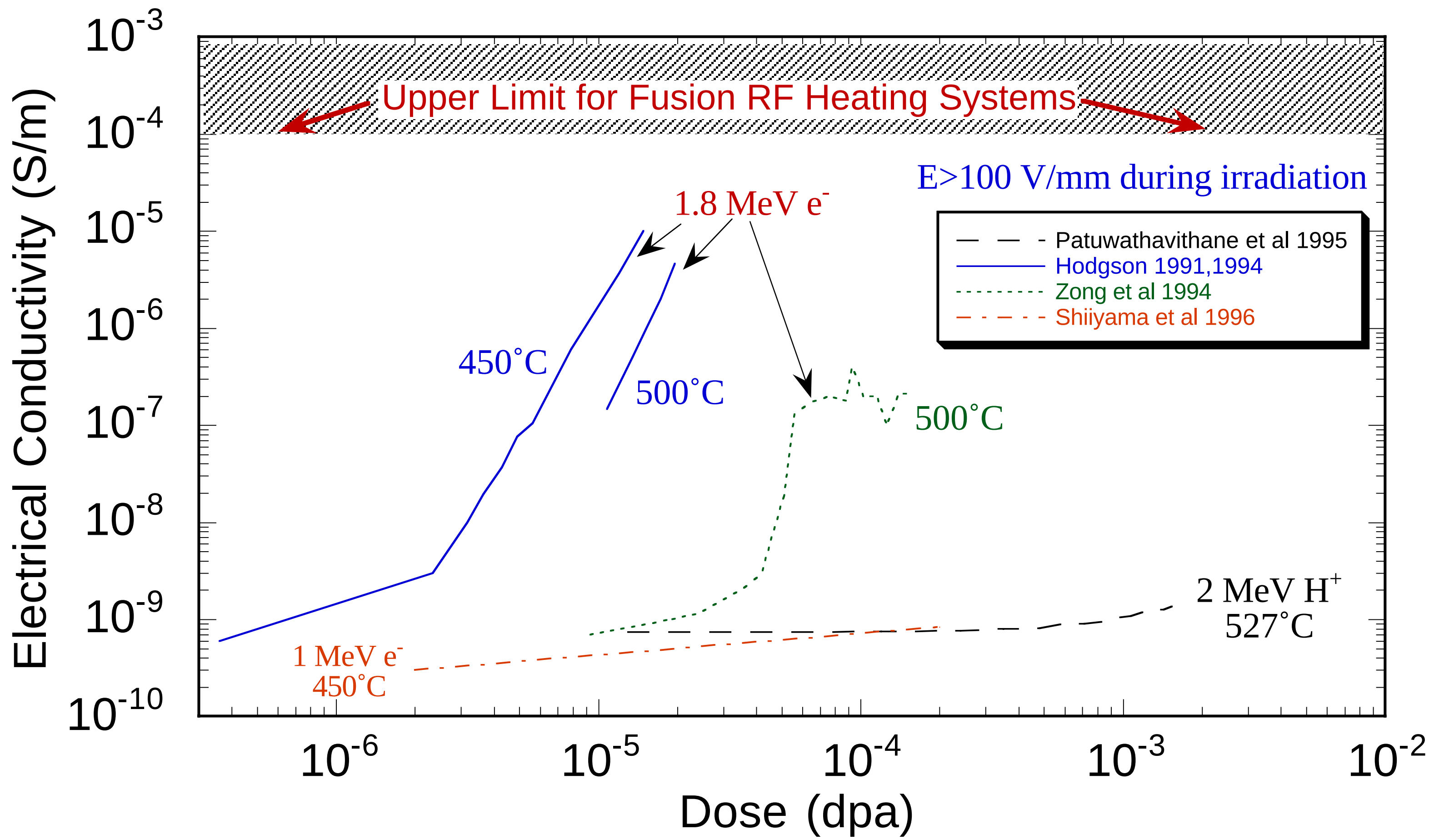
Radiation-induced conductivity





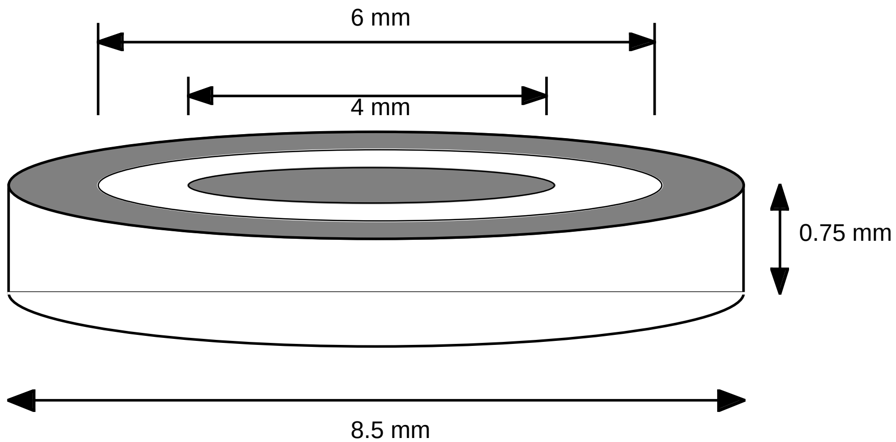
Radiation-Induced Electrical Degradation

RIED studies on single crystal alumina at 450-530˚C

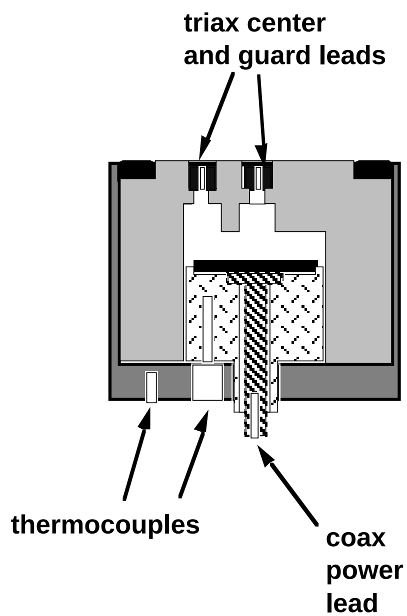


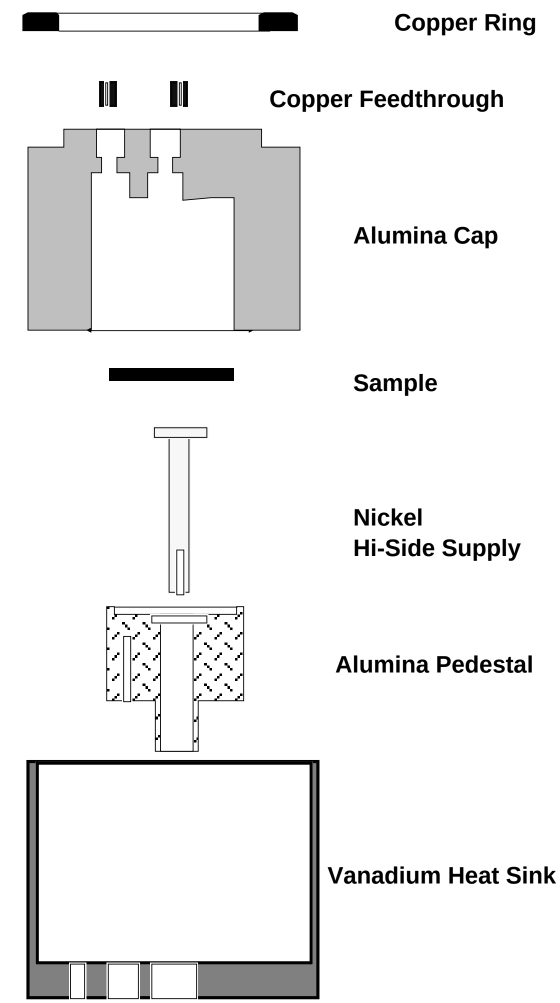
- **Studies performed in early 1990's suggested that catastrophic RIED might occur in ceramic insulators irradiated with an applied electric field >50 V/mm at 300-550˚C**
- **Fission reactor (HFBR and HFIR) irradiations were utilized to investigate behavior at higher doses (>10^{-3} dpa)**

Reactor in-situ test technique was in general conformance to ASTM standards for measuring resistance of insulating materials (D257-91)

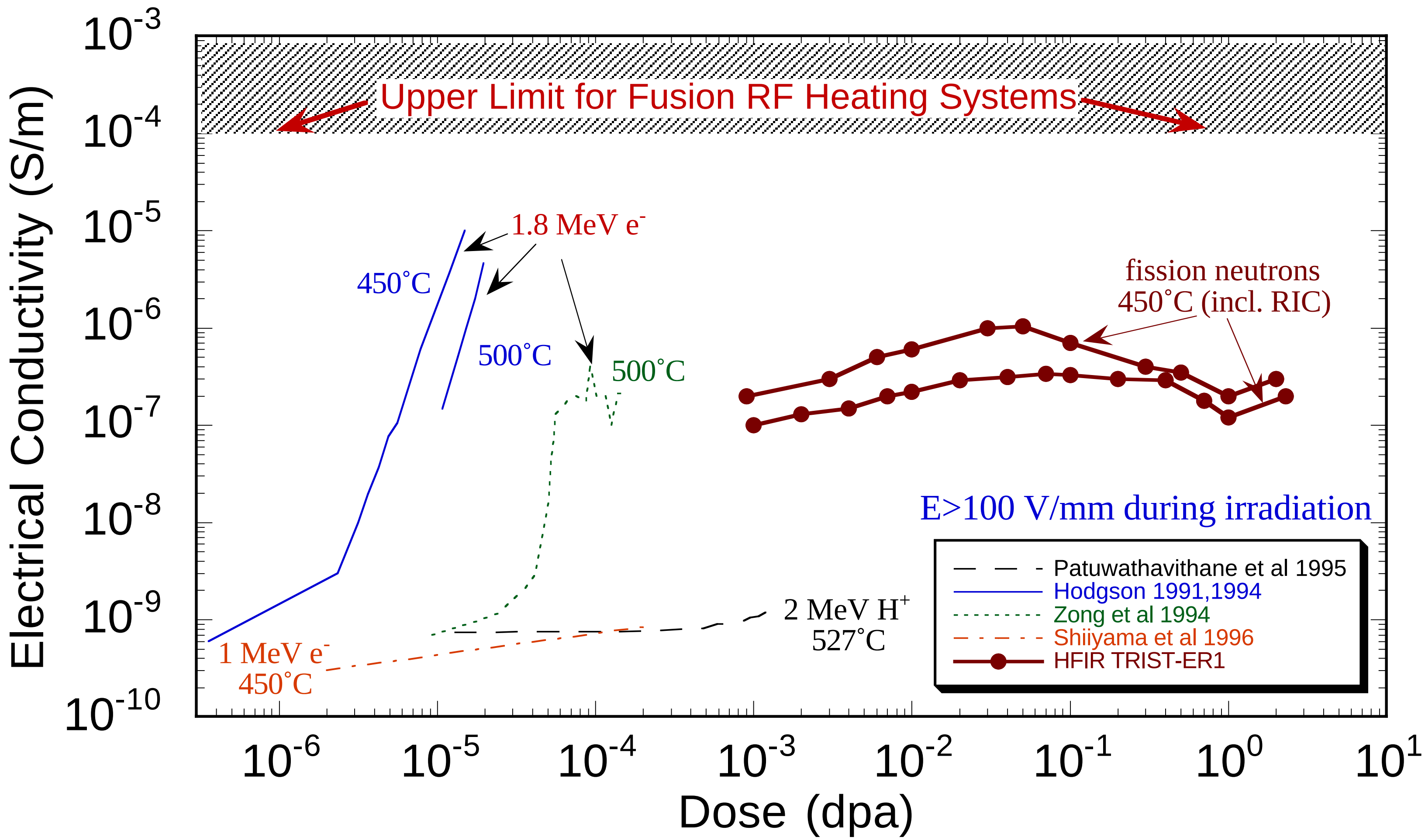


- Guard ring geometry
- Low-side measurement, triax guarded cables
- Resistivity determined from slope of I-V plot (±100 V)



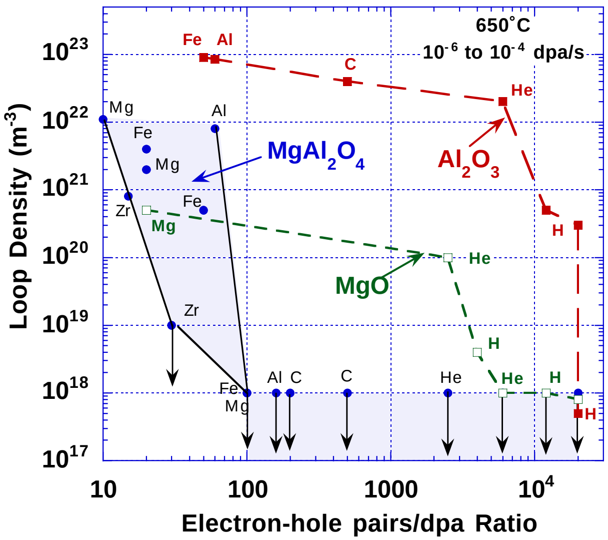


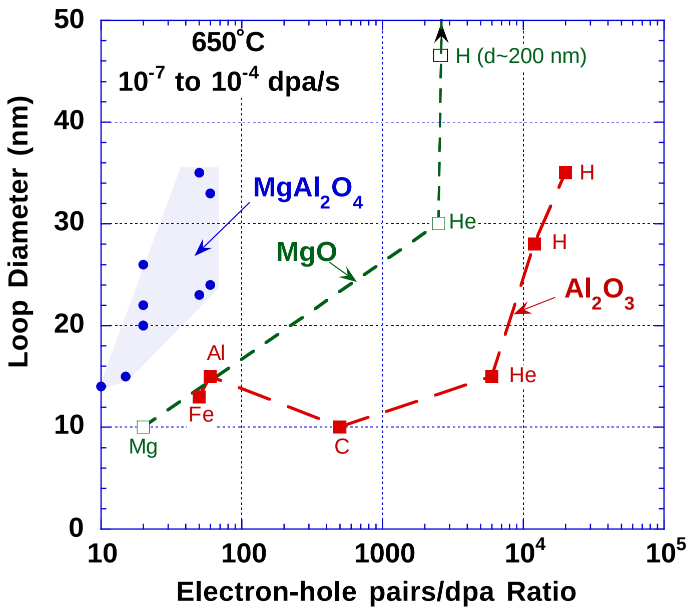
Permanent Electrical Degradation was not Observed in Al_2O_3 Specimens Irradiated up to Doses of ~3 dpa



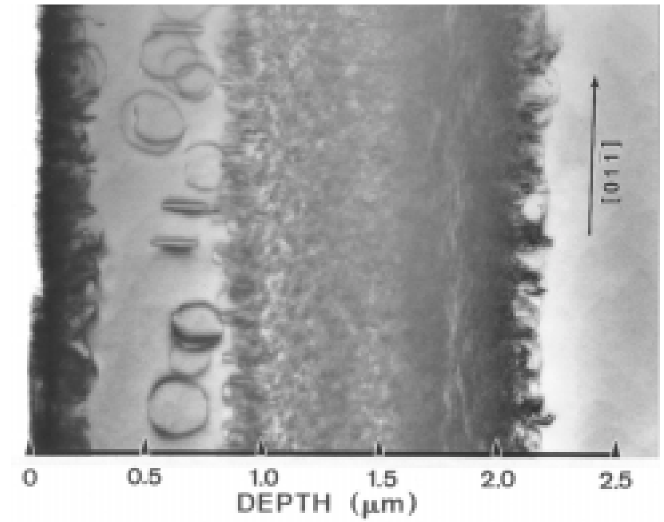
- Earlier reports of RIED are attributed to experimental artifacts (electron beam charging and surface conduction or microcracking effects)

Investigation of ionization-induced diffusion in ceramics

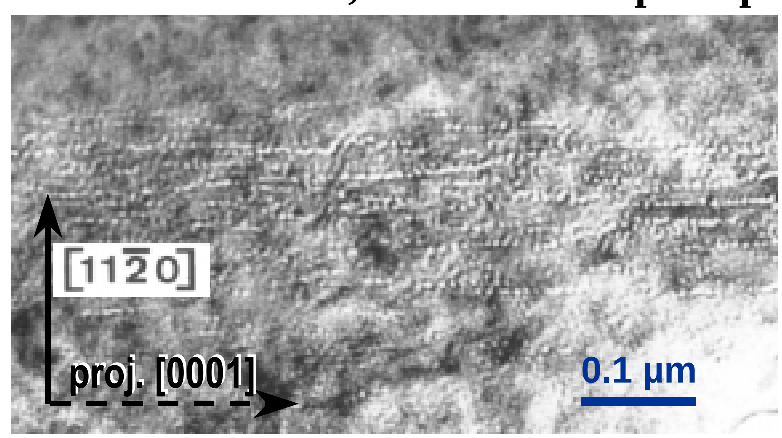




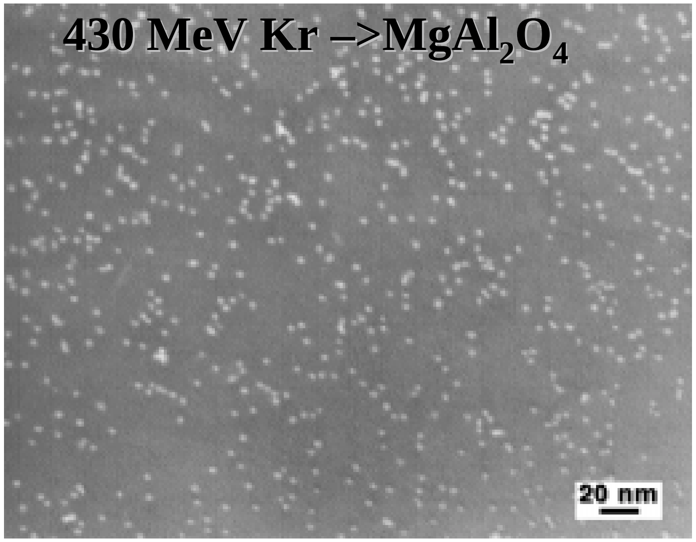
Large interstitial loops in $MgAl_2O_4$ ion-irradiated at 25˚C for regions with >100 eln.-hole pairs per dpa



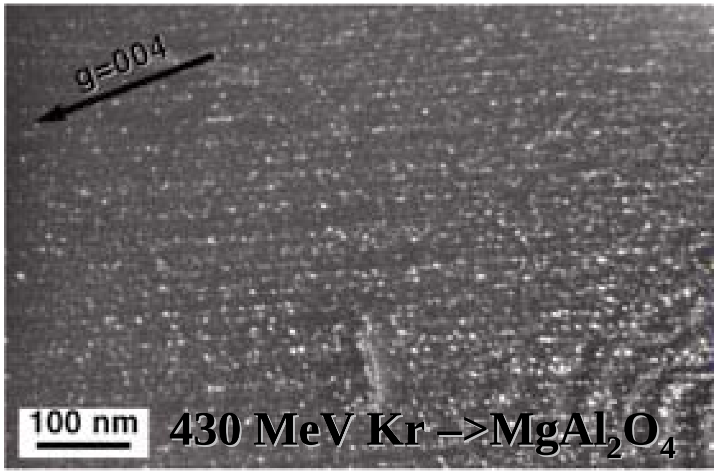
Aligned cavities in Al_2O_3 ion-irradiated at 25˚C (Al/O/He ion irradiation, >500 eln.-hole pairs per dpa)

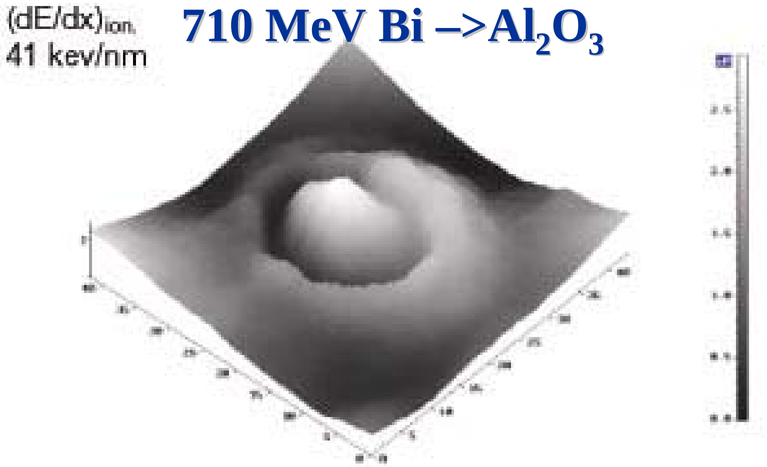


Highly ionizing radiation ($dE_{ioniz.}/dx > 7$ keV/nm) introduces new damage production mechanisms

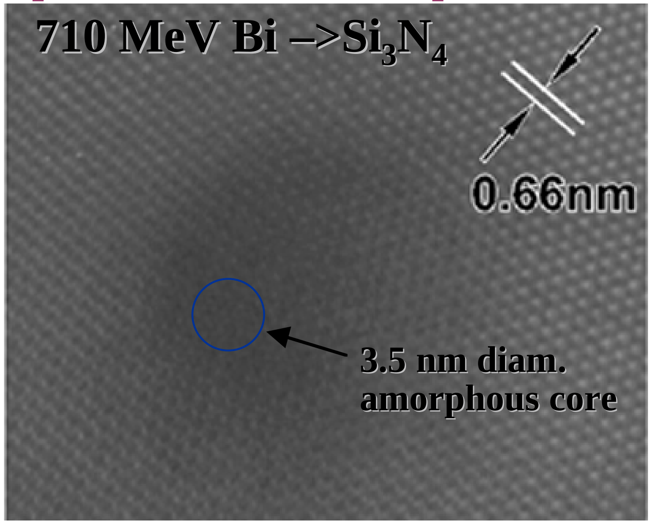


Ion tracks produce displacement damage via inelastic atomic events



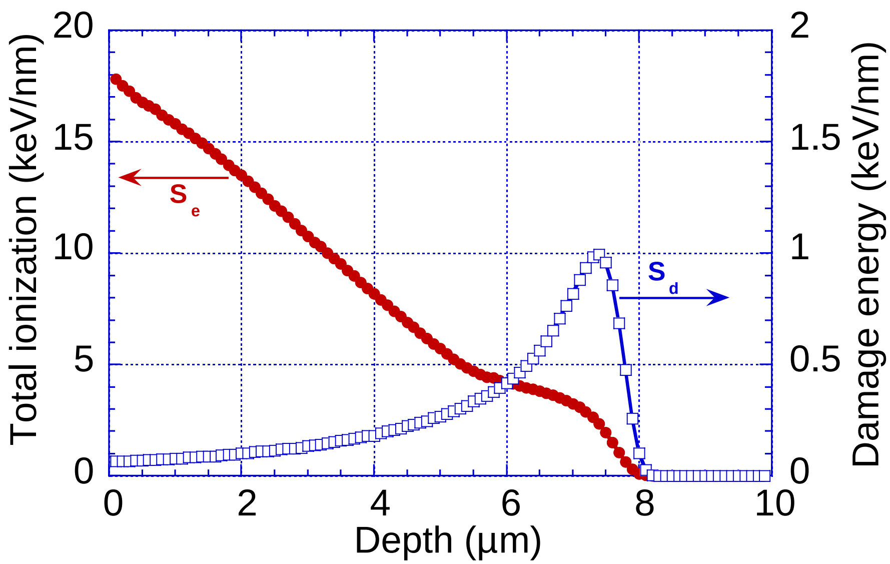


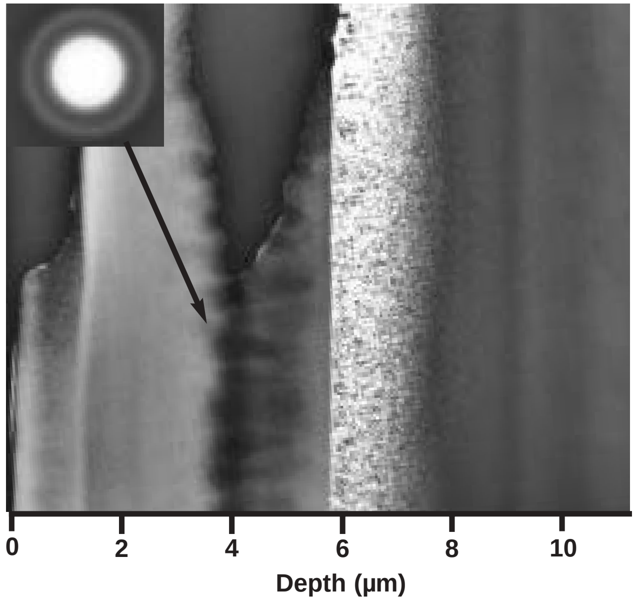
Swift heavy ions induce surface protrusions and amorphization



The high electronic stopping powers associated with fission fragment recoil ions can produce pronounced effects

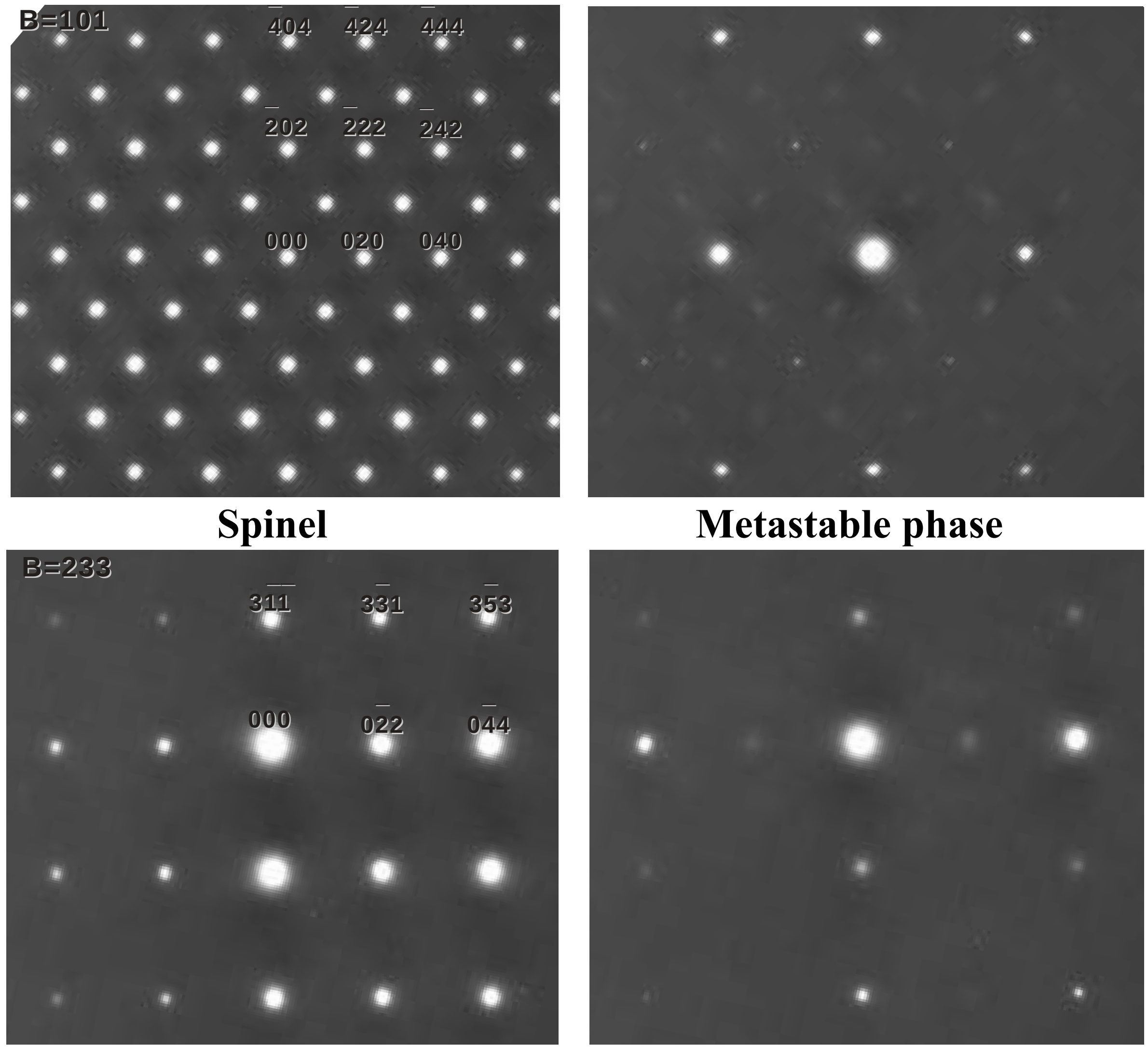
- **$MgAl_2O_4$ irradiated with 72 MeV I^+ ions experiences a crystalline phase change and then amorphization when $dE/dx)_e$>8 keV/nm**
  - **Volumetric expansion ΔV/V~35% due to amorphization will cause severe stresses and cracking**
  - **Amorphization occurs readily up to 500˚C**



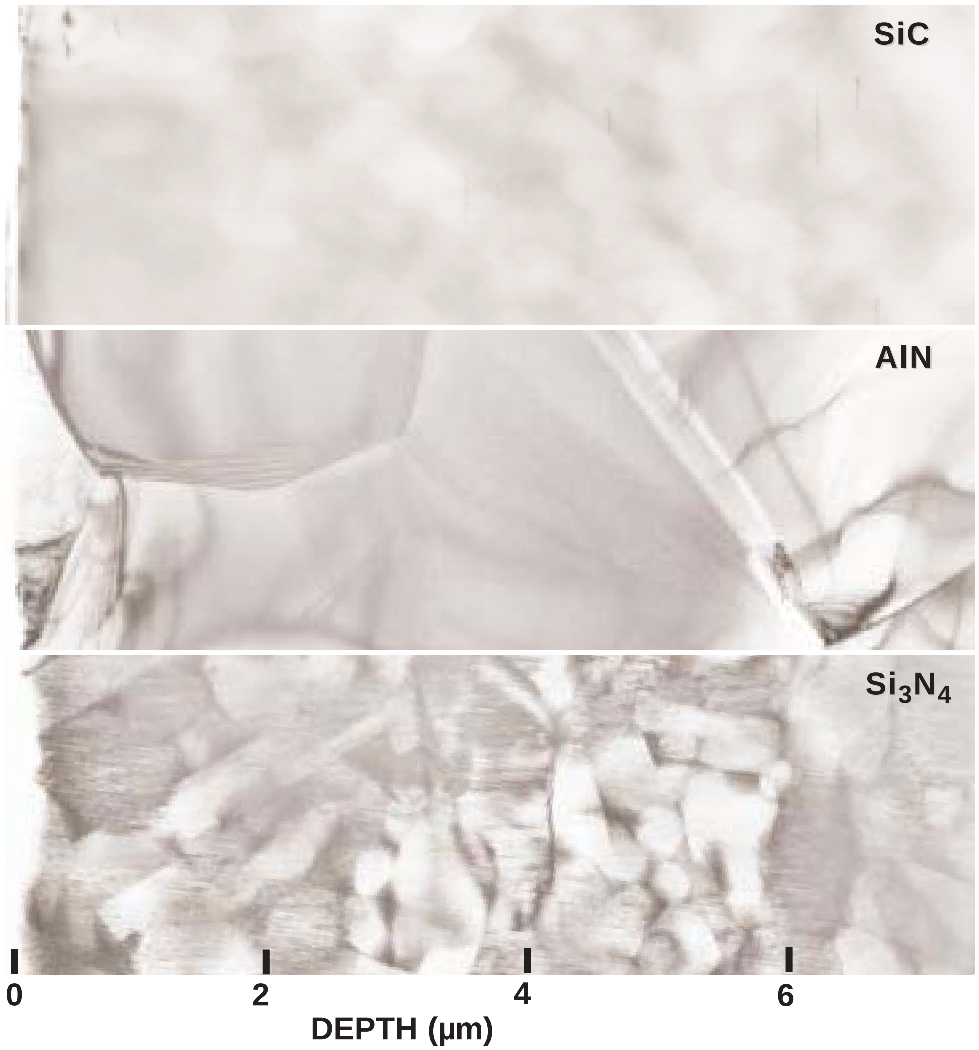


Will these ionizing radiation effects also occur in fusion reactors?

New (metastable) crystalline phase occurs in $MgAl_2O_4$ during 72 MeV ion irradiation



Comparison of low-magnification microstructures of ceramics irradiated with 710 MeV Bi ions



Ion track damage is visible in Si_3N_4, but not in SiC or AlN [max $dE/dx)_e$~34 keV/nm]

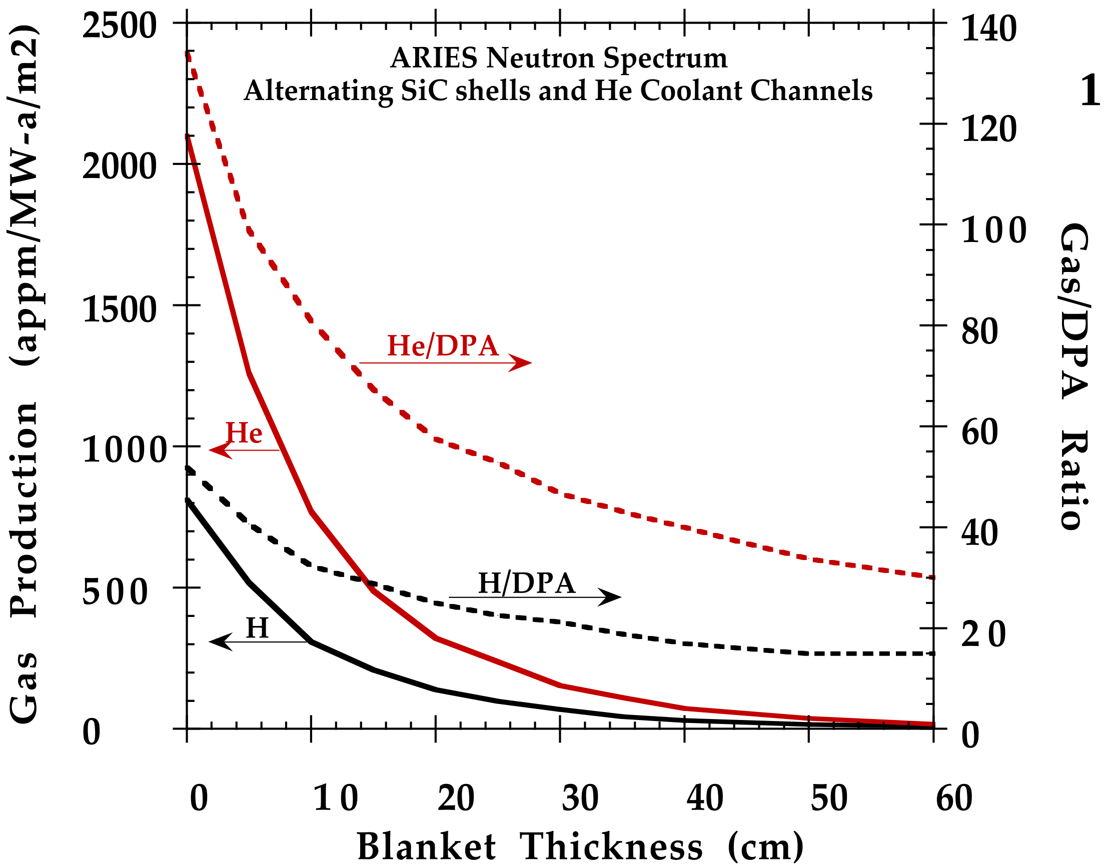
Summary of threshold ionizing radiation levels for defect production in ceramics

Material	Thermal conductivity (W/m-K)	Threshold dE/dx)$_e$ for ion track damage
$MgAl_2O_4$	20	8 keV/nm
β-Si_3N_4	29	15 keV/nm
Al_2O_3	32	~20 keV/nm
AlN	177	>34 keV/nm
SiC	350	>34 keV/nm

- Threshold dE/dx)$_e$ is comparable or higher than the ionizing radiation fields anticipated in fusion reactors
- Studies on effect of radiation-induced thermal conductivity degradation on threshold dE/dx for track formation are in progress
  - radiation-induced degradation of K_{th} may cause a reduction in the threshold dE/dx)$_e$ value

Helium Transmutation in Silicon Carbide

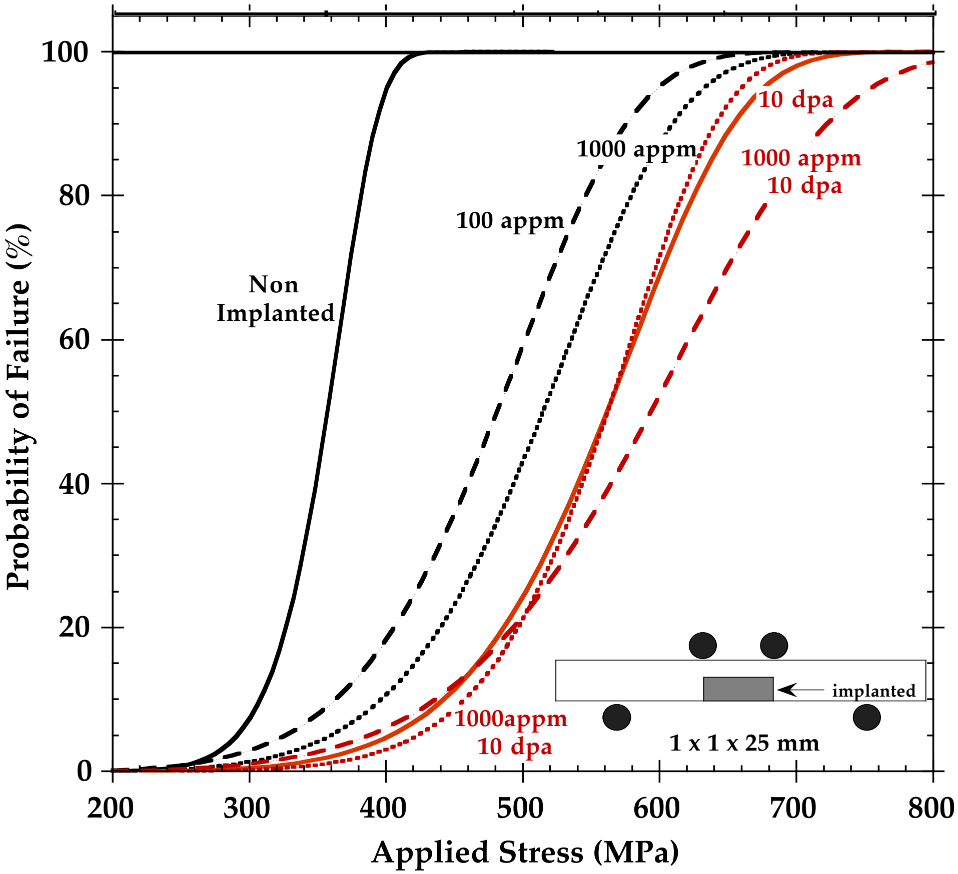
- High energy fusion neutron cause spallation He to be formed. **Up to 1 atom %.**
  ?? What is the effect of high helium concentration on properties of SiC ??

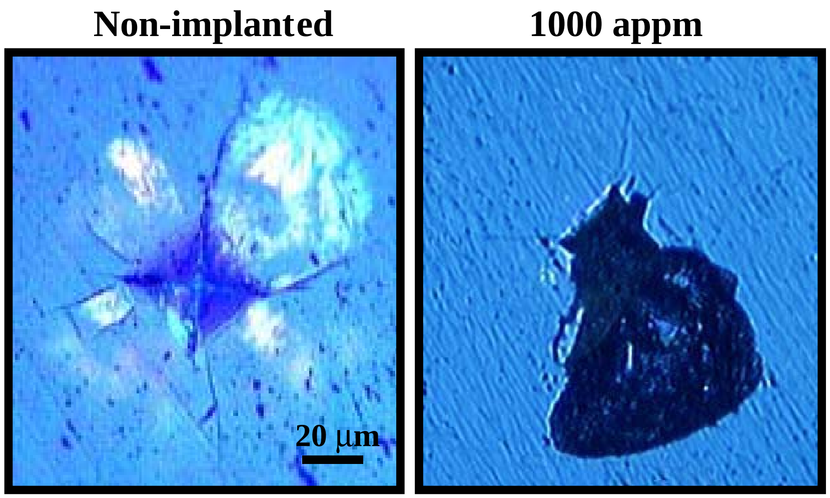


1 MW-yr/m2 at First Wall Produces

He :1600 appm
H : 440 appm
Mg : 458
Be : 234
Al : 72

Similar to neutron irradiation, He causes hardening and strengthening of SiC





Property	Virgin	100 appm	1000 appm	Neutron Irradiated (8 dpa)	100 appm and 8 dpa	1000 appm and 8 dpa
Swelling (%)	0		~0.5		~0.9	~0.9
Bend Strength (MPa)	353 ± 72	474 ± 127	532 ± 122	540 ±139	585 ± 162	554 ± 130
Indent Fracture Toughness (MPa/m$^{1/2}$)	1.4	1.2	0.9			
Vickers Hardness (GPa)	2257 ± 103	2381 ±120	2516 ±180			

SUMMARY

- Examination of irradiated ceramics is providing valuable fundamental materials science information
  - Point defect migration energies
  - Properties of amorphous materials
  - Phonon scattering physics
- Ionizing radiation can induce numerous phenomena in ceramics
  - Radiation induced conductivity
  - Ionization induced diffusion
  - Defect production
- Helium causes hardening in ceramics such as SiC at intermediate temperatures
  - He effects at higher temperatures (e.g., near 1000˚C for SiC) need further investigation (effect on cavity swelling, etc.)