2359-28

Joint ICTP-IAEA Workshop on Physics of Radiation Effect and its Simulation for Non-Metallic Condensed Matter

13 - 24 August 2012

Effects of neutron and gamma irradiation on degradation of nonmetallic materials for high temperature applications

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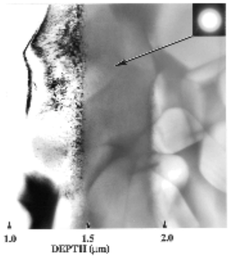
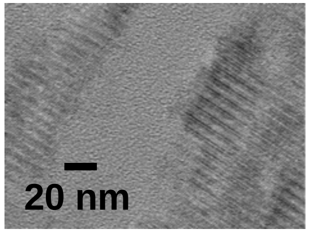
Trieste, Italy

August 13-24, 2012

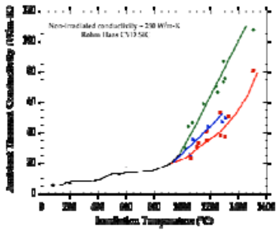
Radiation Damage can Produce Large Changes in Ceramic Materials

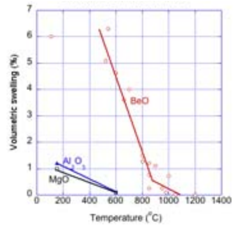
- **Amorphization and disordering (<0.2 T_M, >0.1 dpa)**



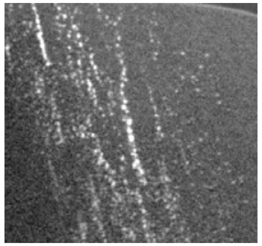


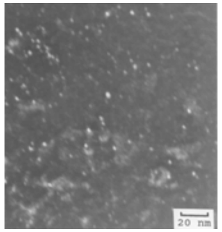
- **Thermal conductivity degradation and defect cluster swelling (<0.35 T_M, >0.01 dpa)**



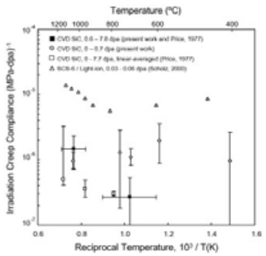


- **Phase instabilities from radiation-induced precipitation (0.3-0.6 T_M, >10 dpa)**

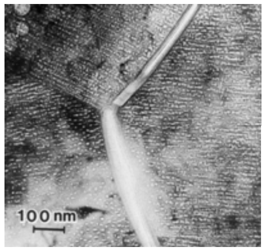


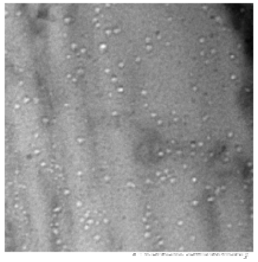


- **Irradiation creep (<0.45 T_M, >10 dpa)**



- **Volumetric swelling from void formation (0.3-0.6 T_M, >10 dpa)**

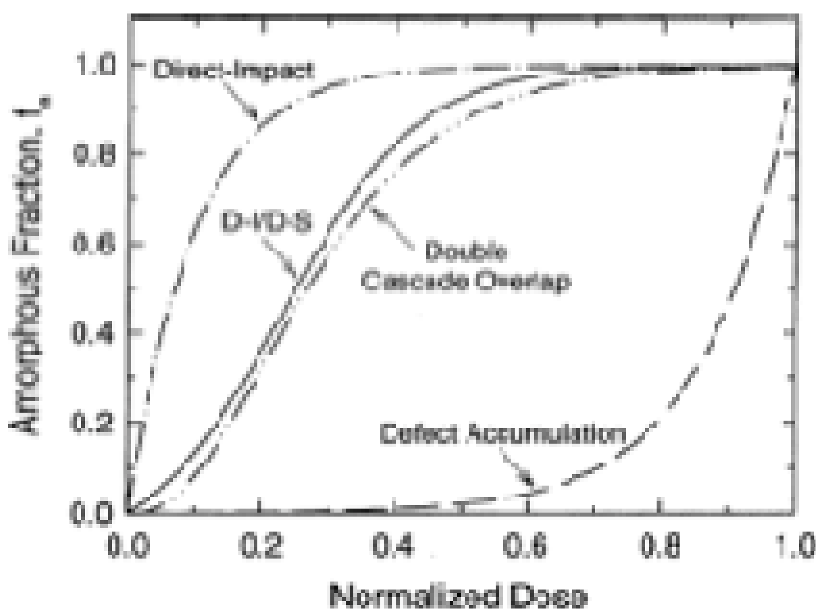




after S.J. Zinkle, Chpt. 3 in Comprehensive Nuclear Materials (Elsevier, 2012)

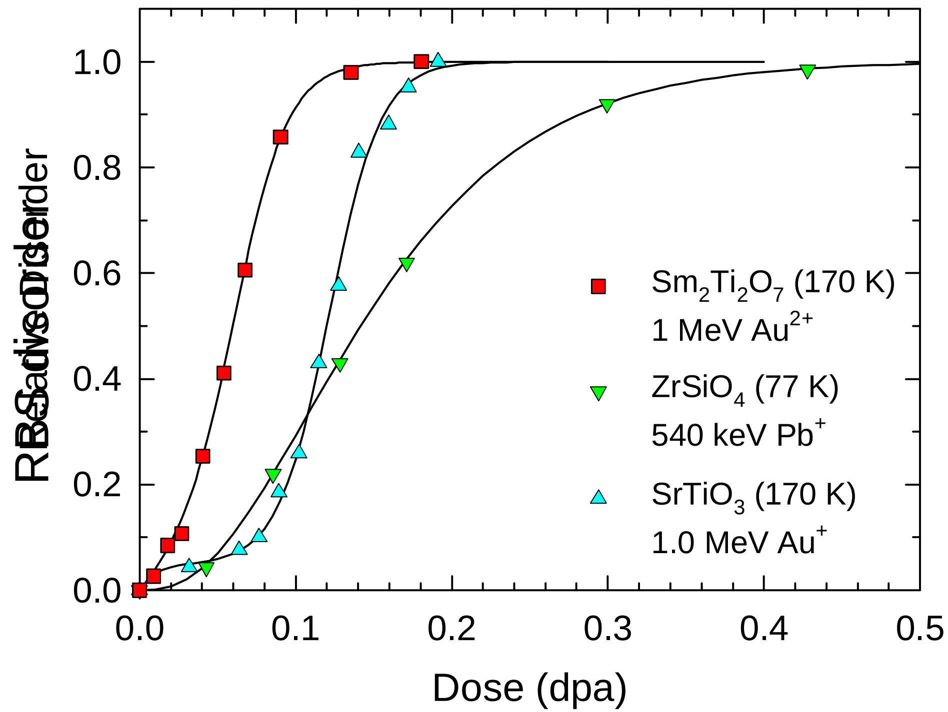
Overview of dose dependence for different amorphization mechanisms

Model predictions



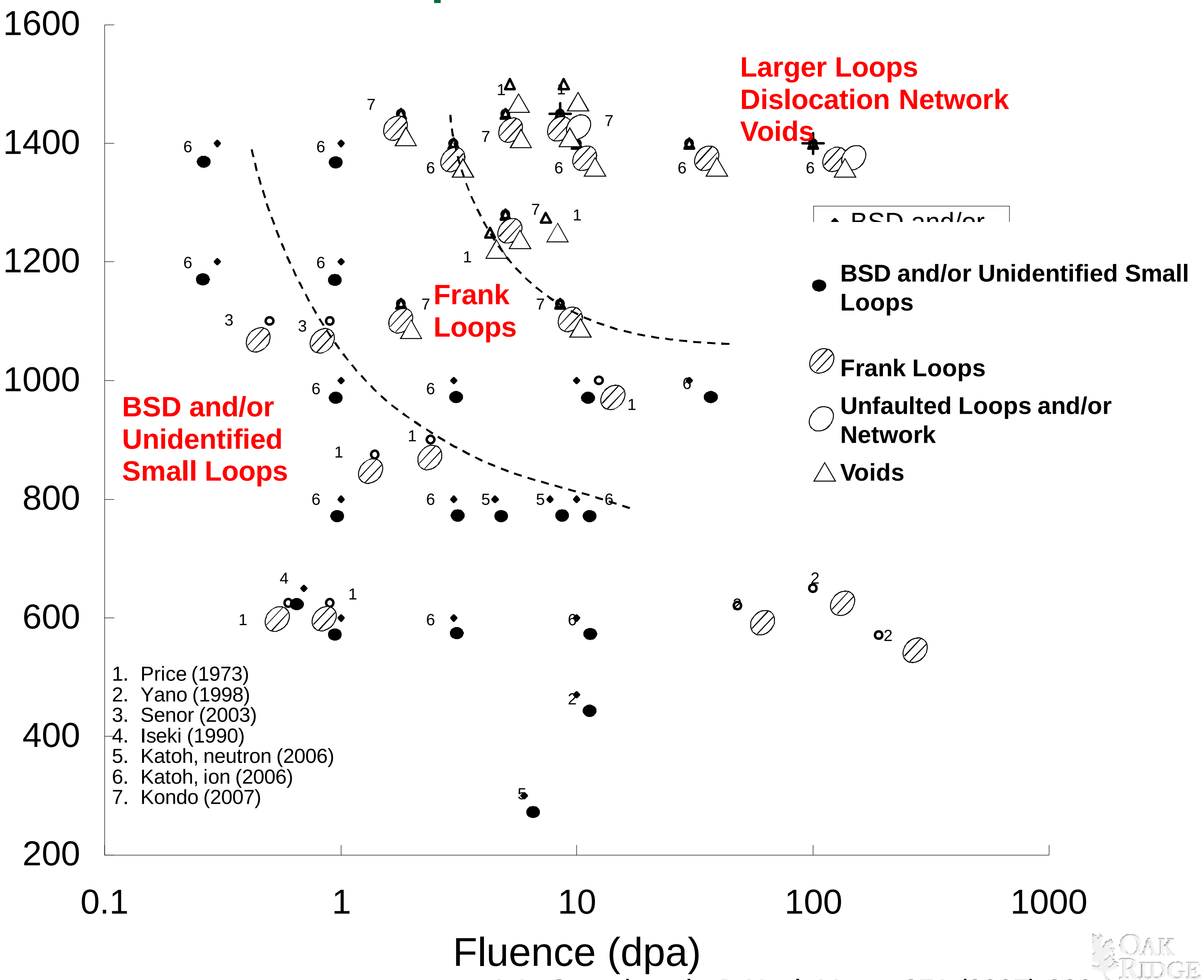
W.J. Weber, Nucl. Instr. Meth. B 166 (2000) 98

Experimental observations



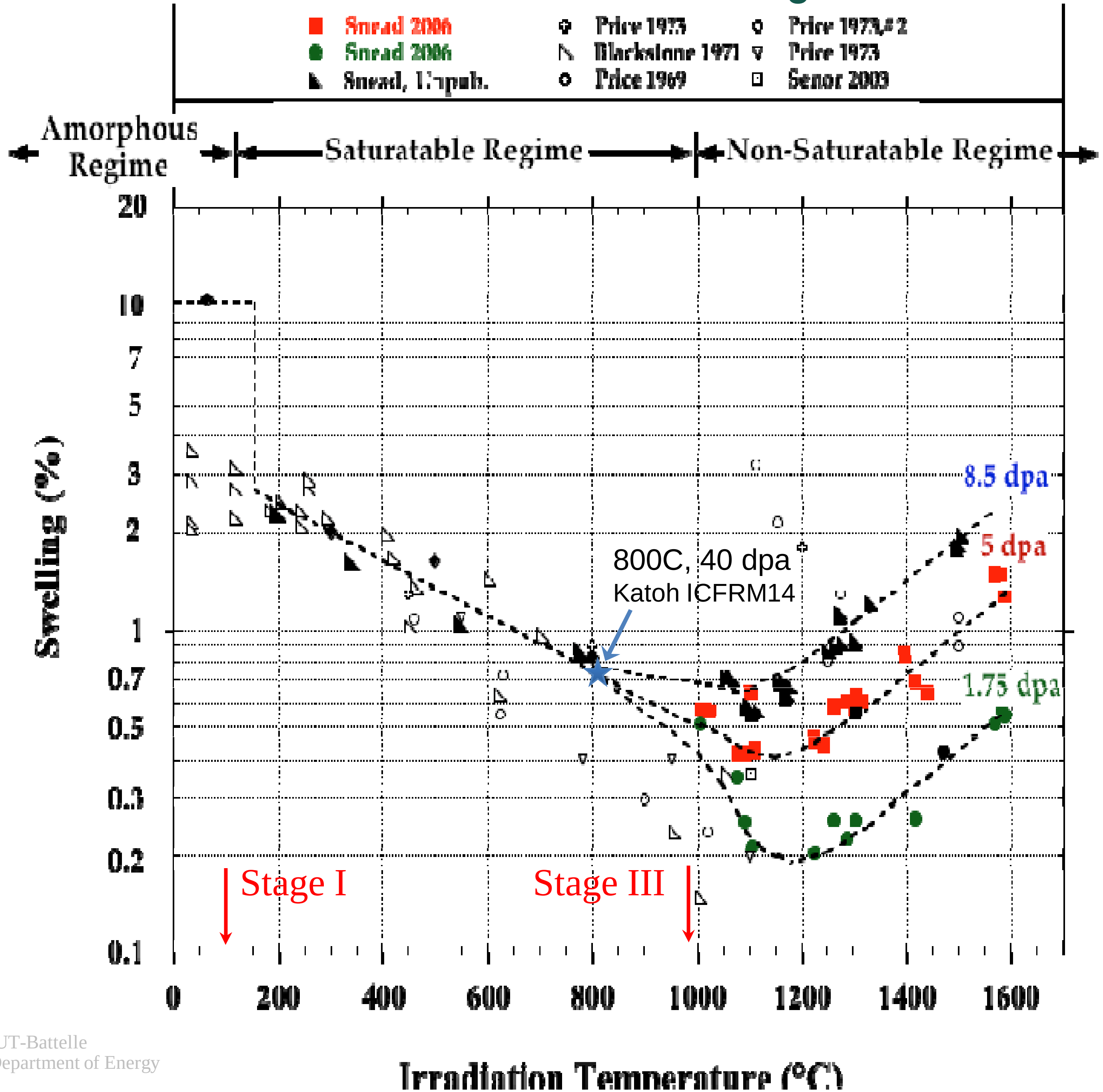
W.J. Weber

Microstructure Map for Irradiated CVD SiC



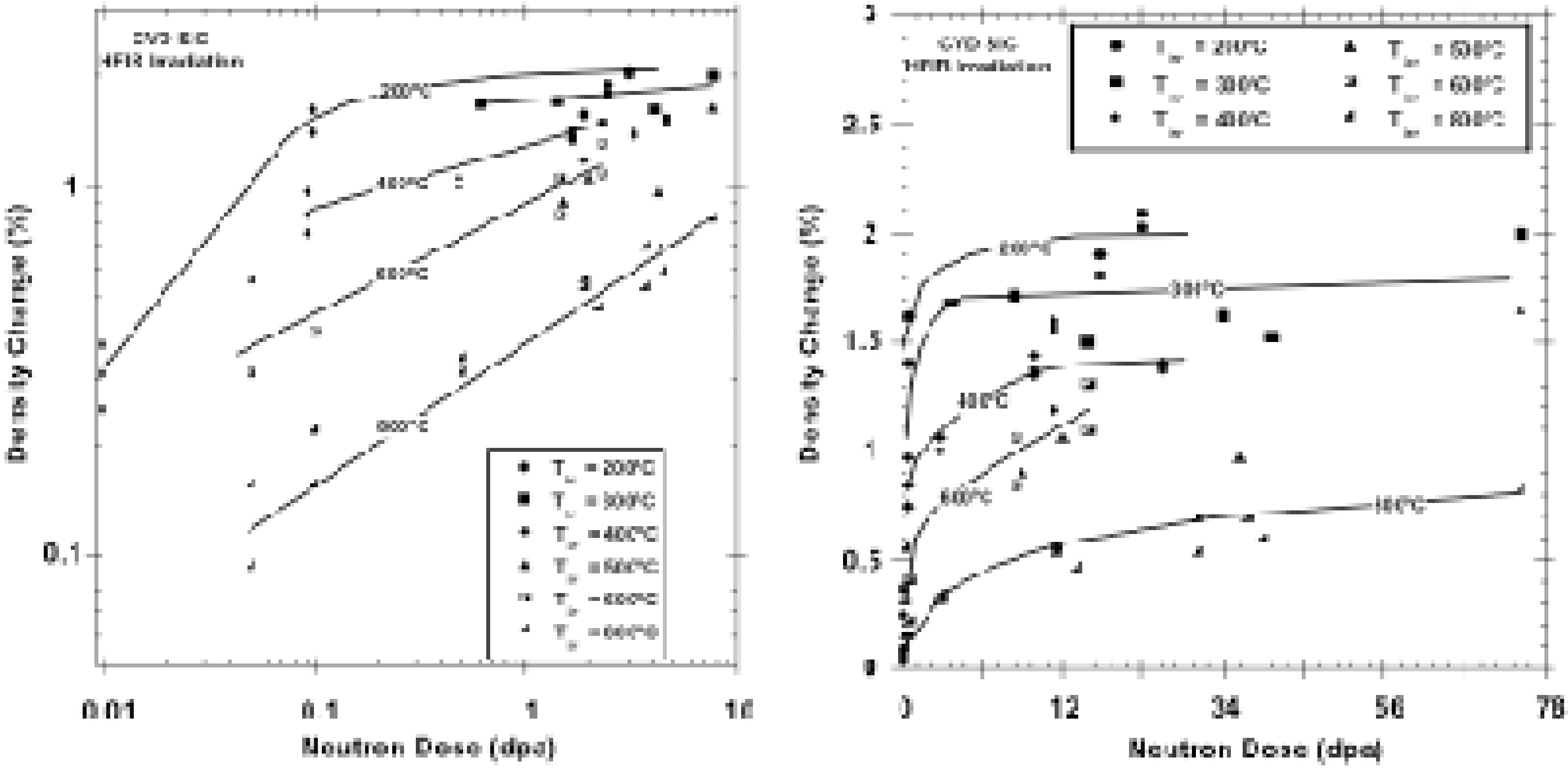
L.L. Snead et al., J. Nucl. Mater. 371 (2007) 329

Irradiation-induced swelling in SiC



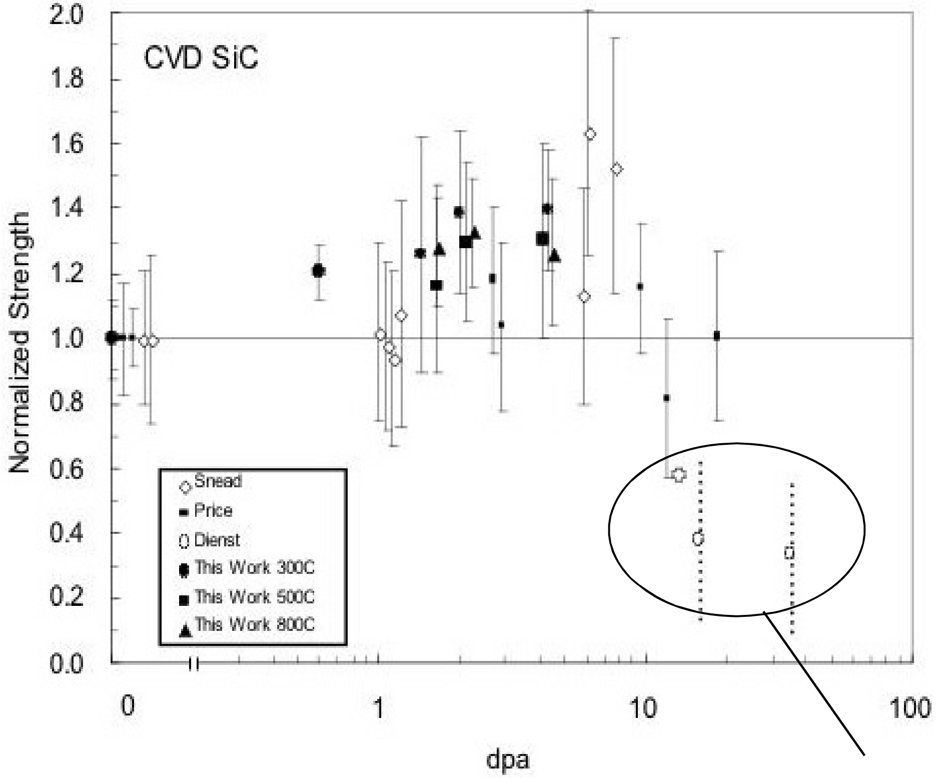
L.L. Snead

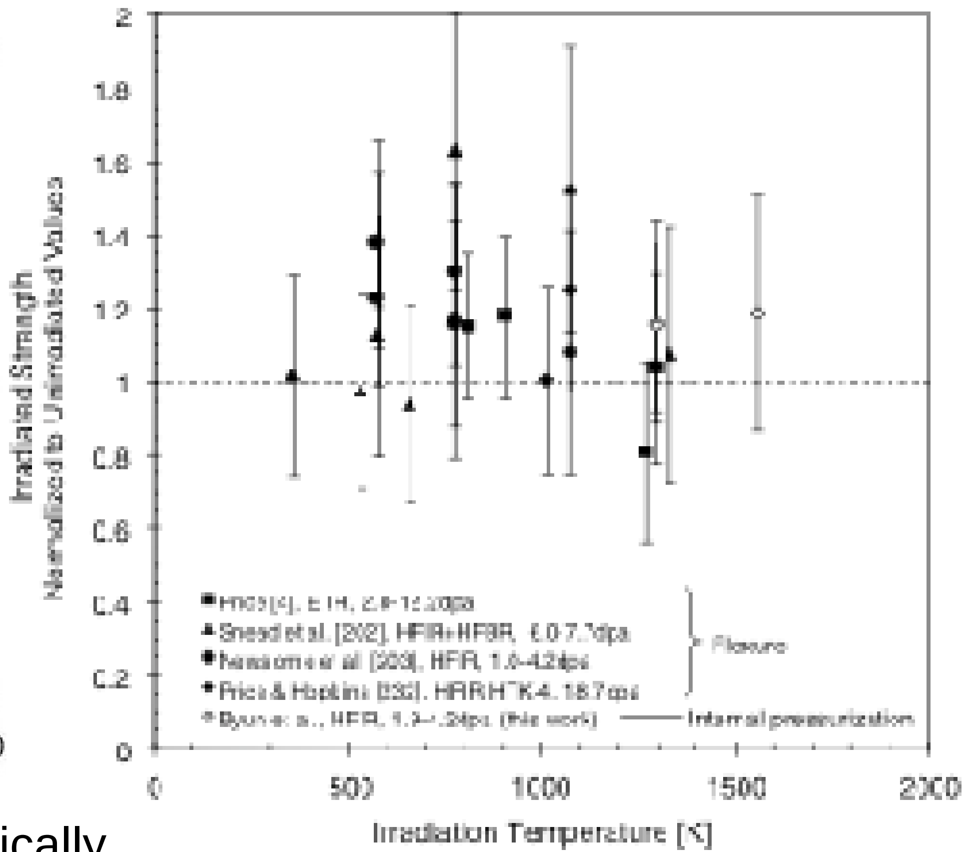
Effect of neutron irradiation on volumetric swelling of SiC



L.L. Snead et al., J. Nucl. Mater. 371 (2007) 329

Effect of neutron irradiation on room temperature flexural strength of SiC

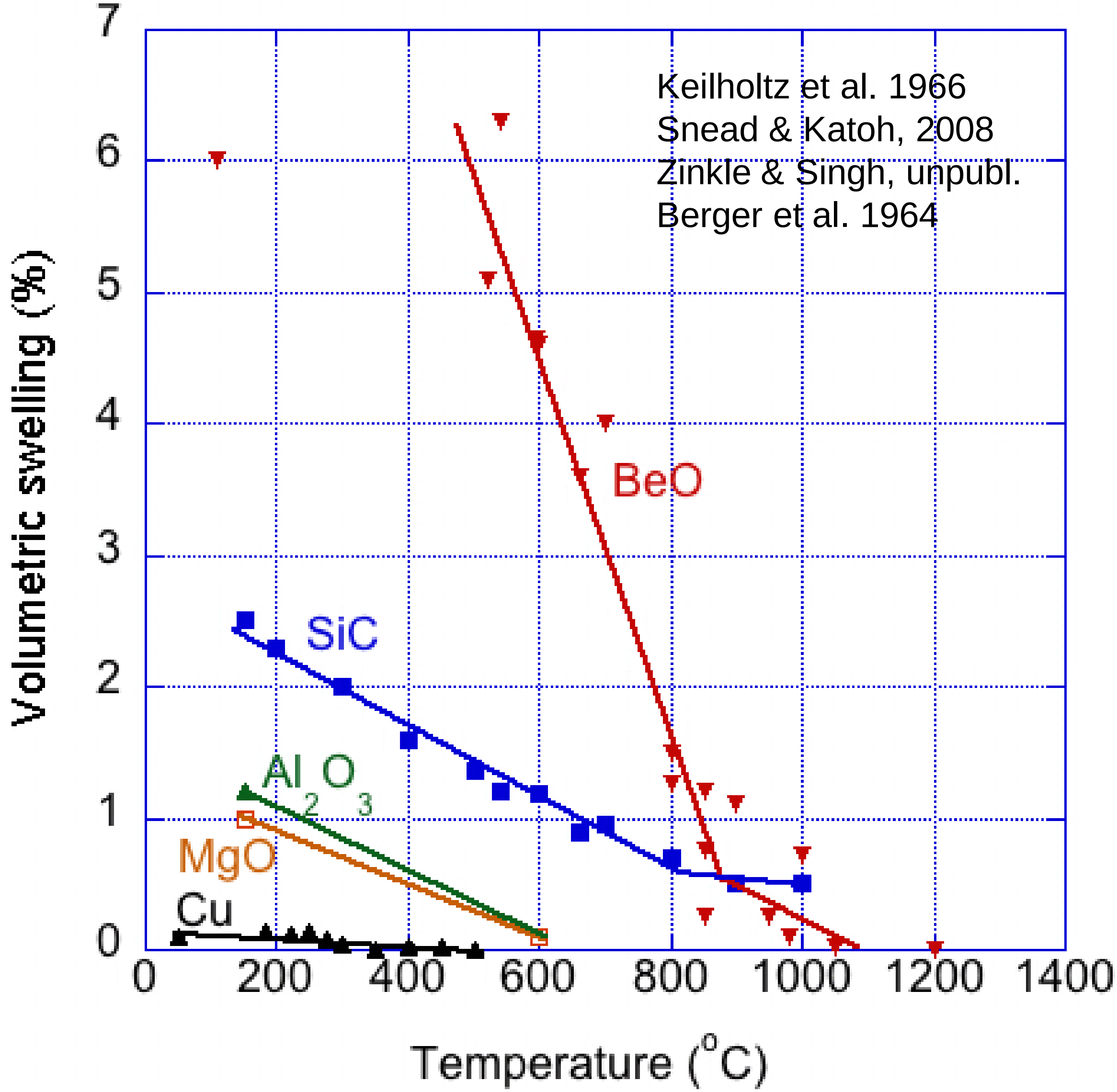




Statistically
Limited ?

L.L. Snead et al., J. Nucl. Mater. 371 (2007) 329

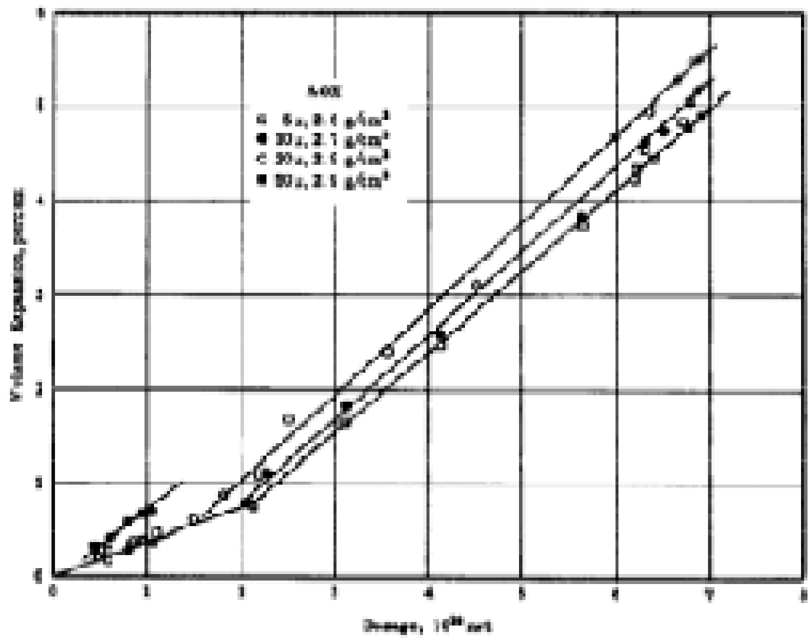
Comparison of point defect swelling behavior in irradiated metals and ceramics



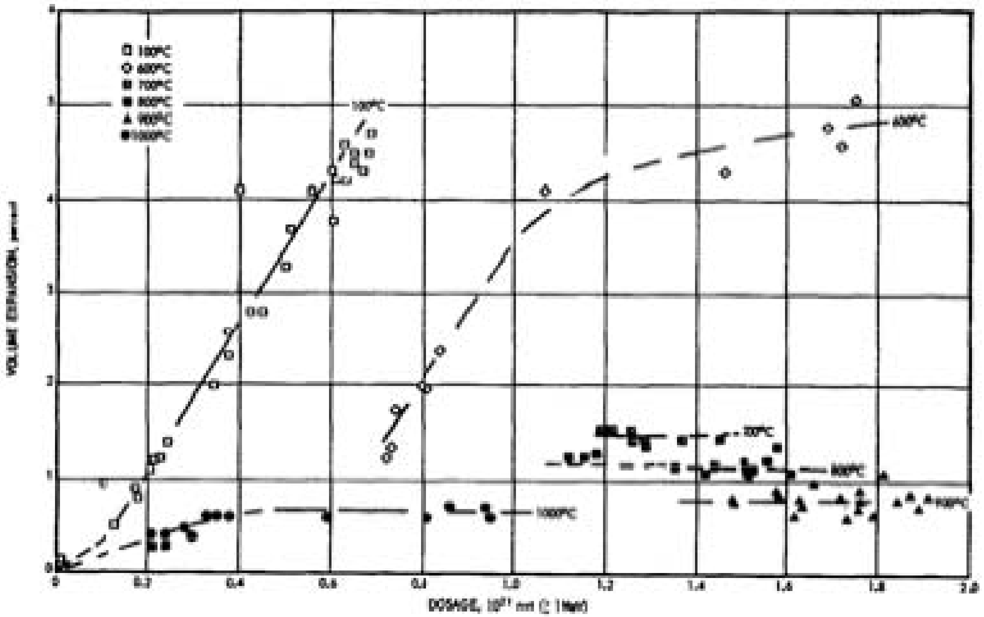
- Large variation in magnitude of point defect swelling in materials
- Metals typically have point defect swelling values near 0.1% (Al, Cu, Ag, Au), compared to 1-5% for ceramics
  - Implies shorter point defect recombination radii or reduced barriers for recombination in metals versus ceramics

Effect of low temperature neutron irradiation on volumetric swelling in BeO

T_{irr}=100°C

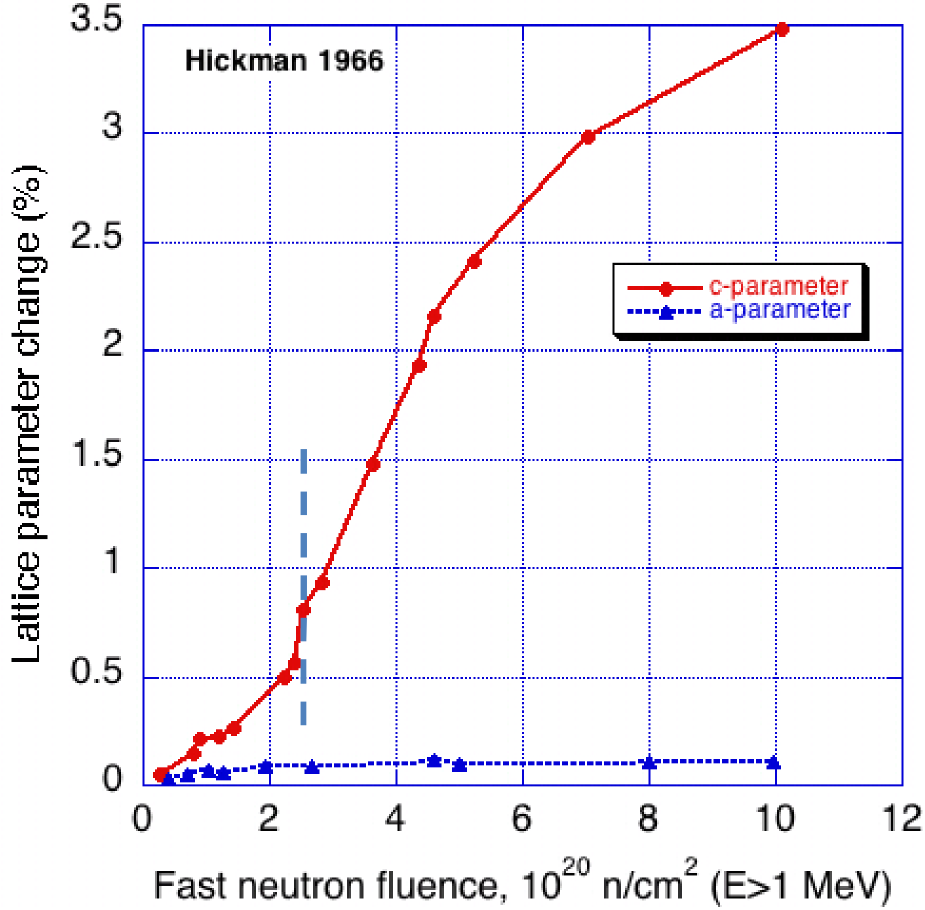


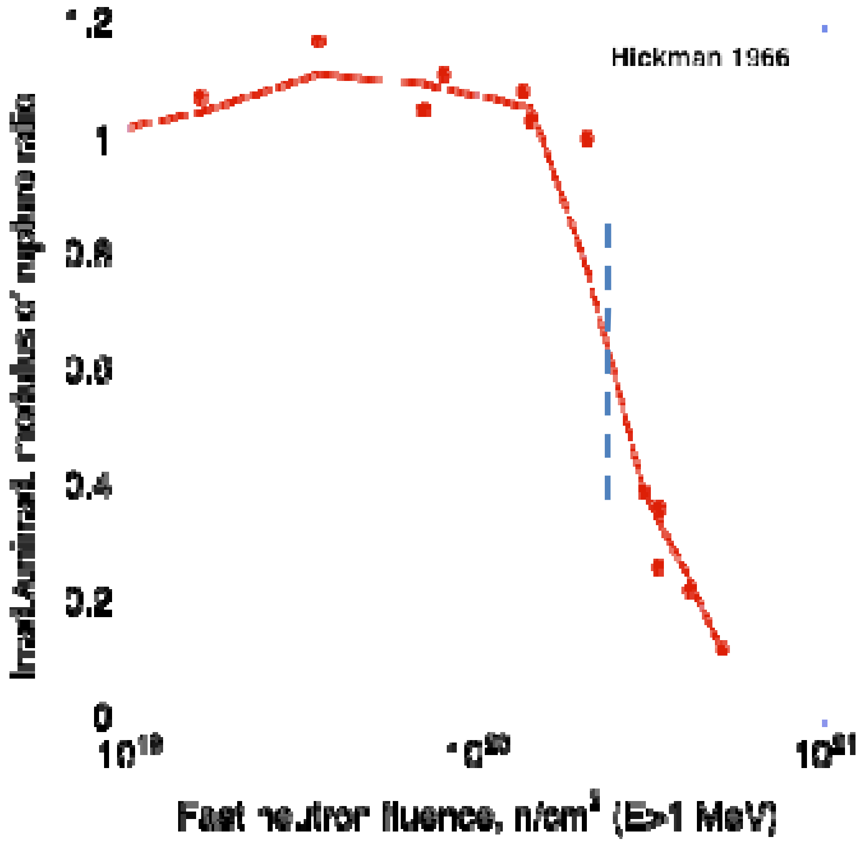
T_{irr}=100-1000°C



C.G. Collins, J. Nucl. Mater. 14 (1964) 69

Effect of fission neutron irradiation near 75°C on the anisotropic swelling and mechanical strength of BeO



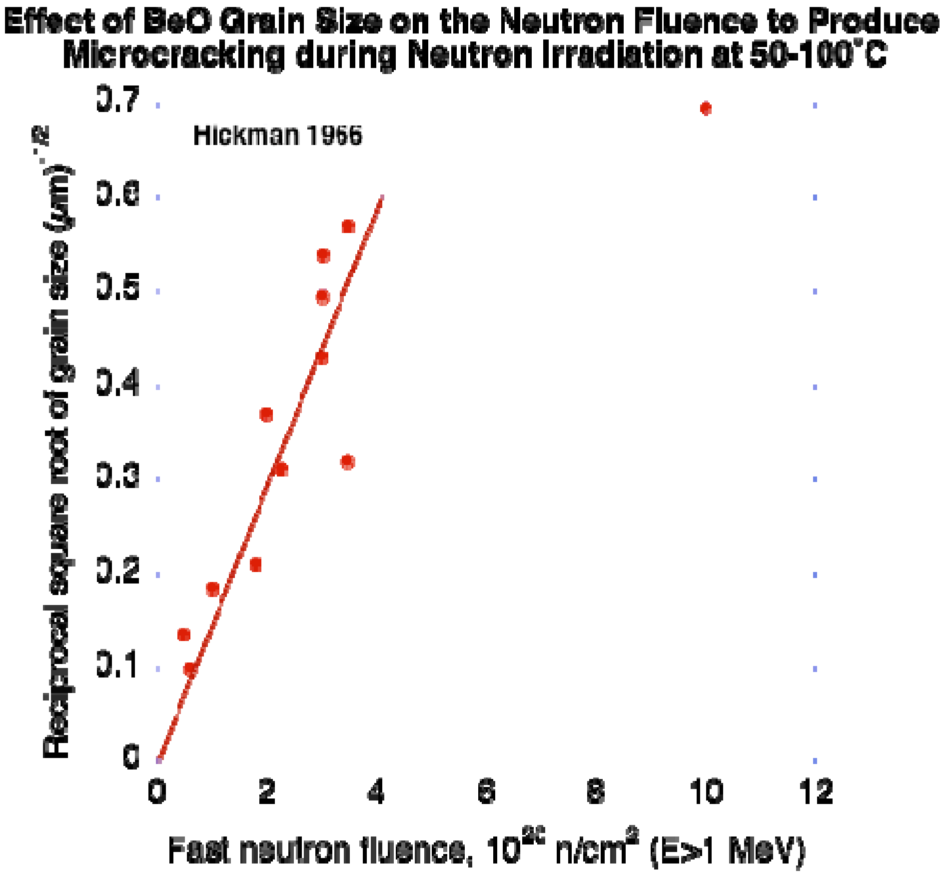


B.S. Hickman, in Studies in Radiation Effects, G.J. Dienes, Ed. (Gordon & Breach, 1966) vol. 1 p. 72

L.L. Snead & S.J. Zinkle, STAIF 2005, ed. M.S. El-Genk, AIP conf. proc. 746 (2005) p. 768

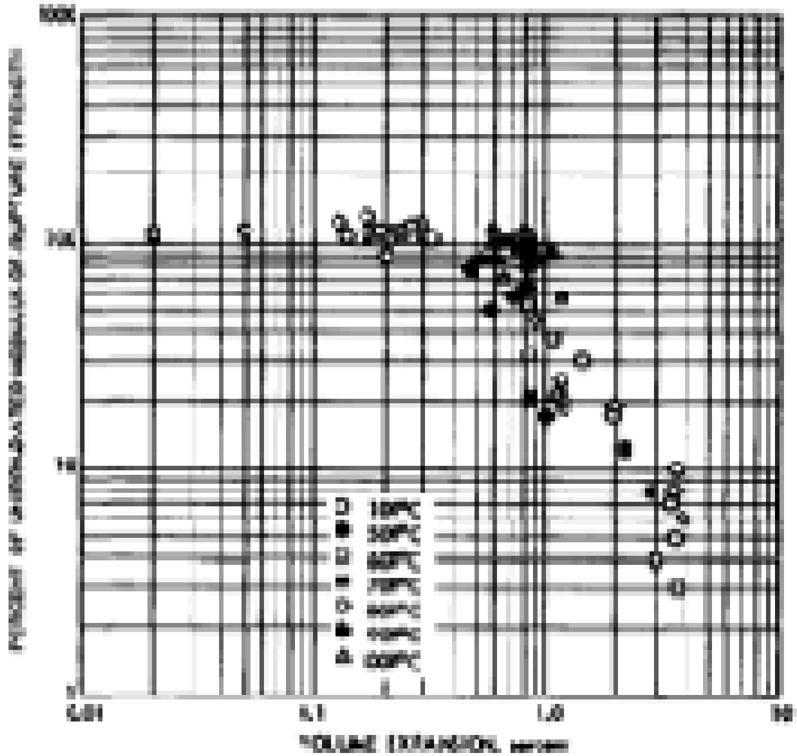
Pronounced strength reduction when macroscopic swelling exceeds ~0.5-1%

Effect of grain size and volumetric swelling on threshold microcracking in polycrystalline BeO



after L.L. Snead & S.J. Zinkle, STAIF 2005, ed. M.S. El-Genk, AIP conf. proc. 746 (2005) p. 768

Flexural strength of BeO vs. volumetric expansion (20 μm grain size, $1.5x10^{21}/m^2$) T_{irr}=100-1000°C

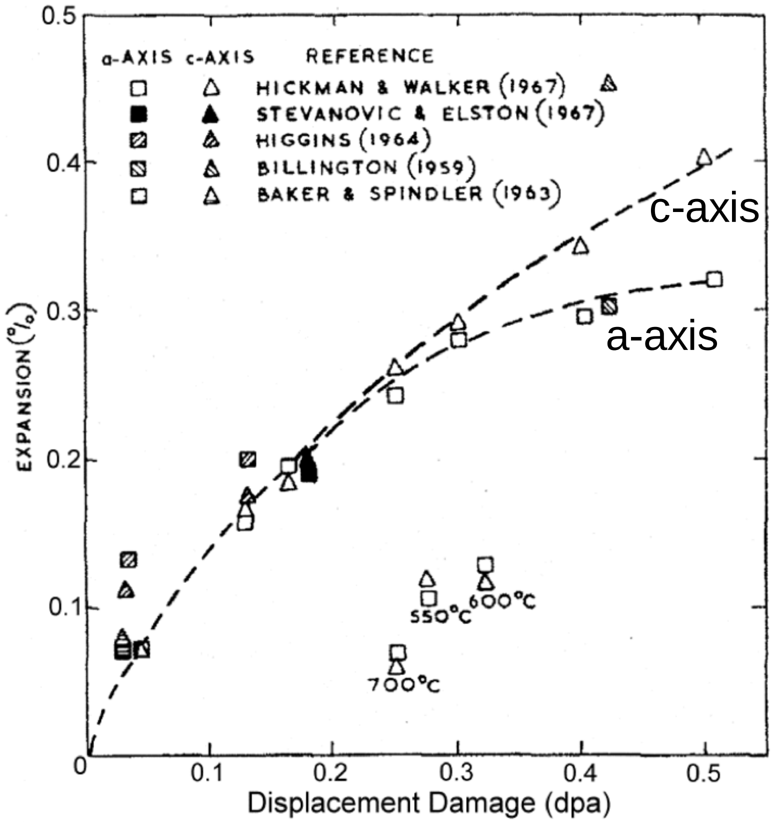


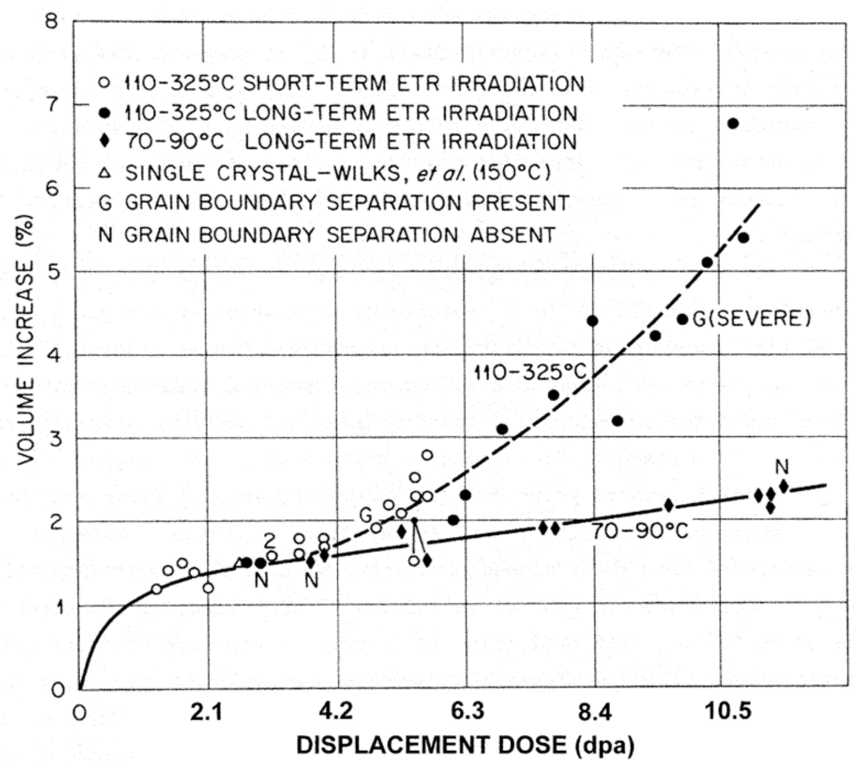
C.G. Collins, J. Nucl. Mater. 14 (1964) 69

Microcracking and strength degradation observed for volumetric swelling >1%

Anisotropic lattice expansion and grain boundary cracking in neutron-irradiated Al_2O_3

T_{irr}=75-100°C





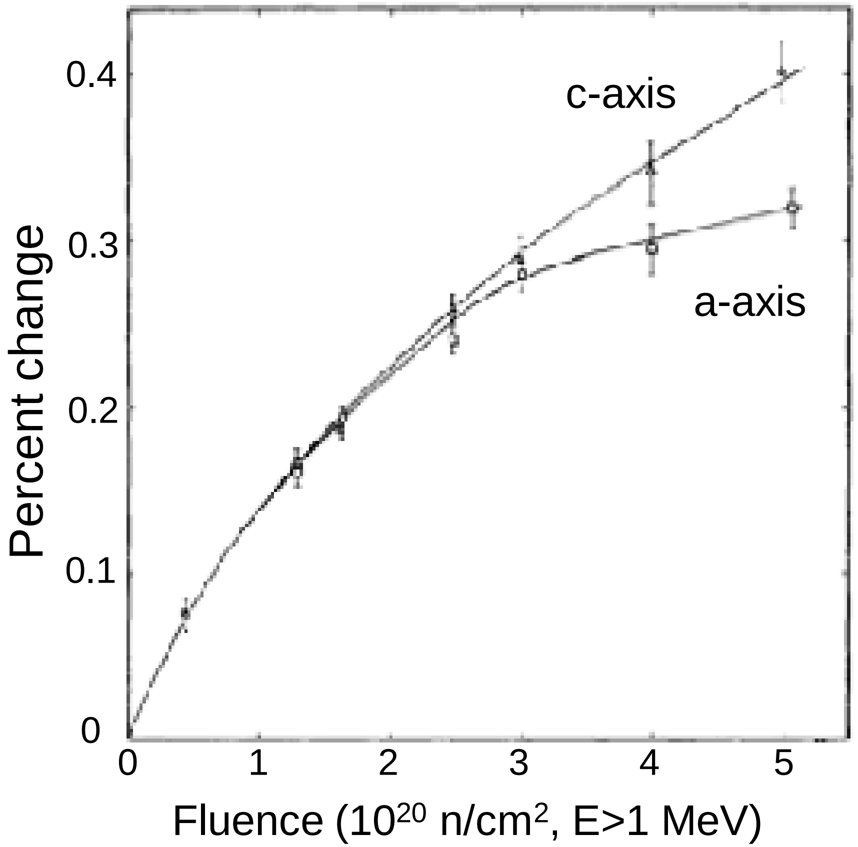
after B.S. Hickman & D.G. Walker, J. Nucl. Mater. 18 (1966) 197

G.W. Keilholtz et al., Nucl. Tech. 17 (1973) 234

Microcracking and strength degradation observed for volumetric swelling >3-4%

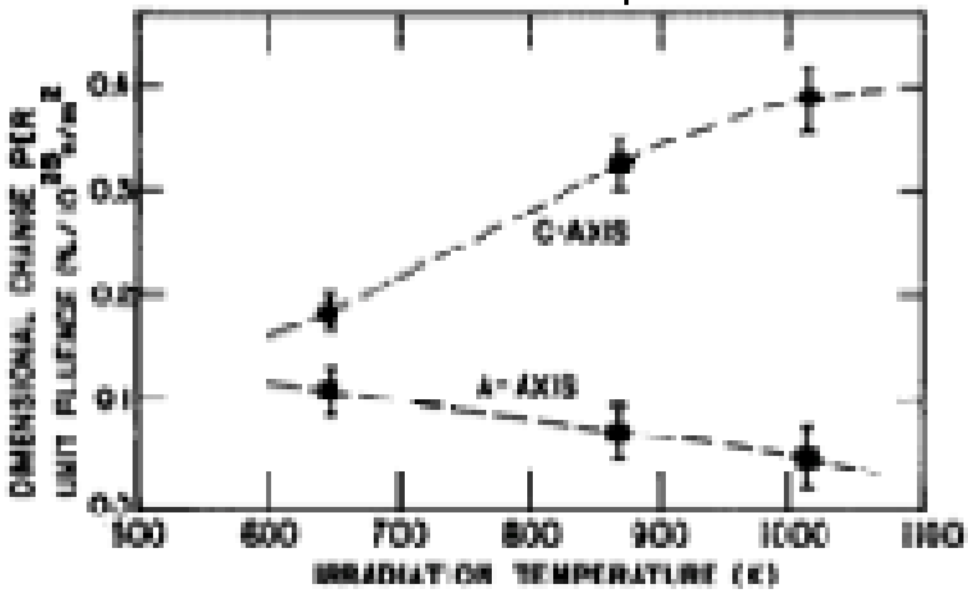
Neutron radiation-induced lattice expansion in Al_2O_3

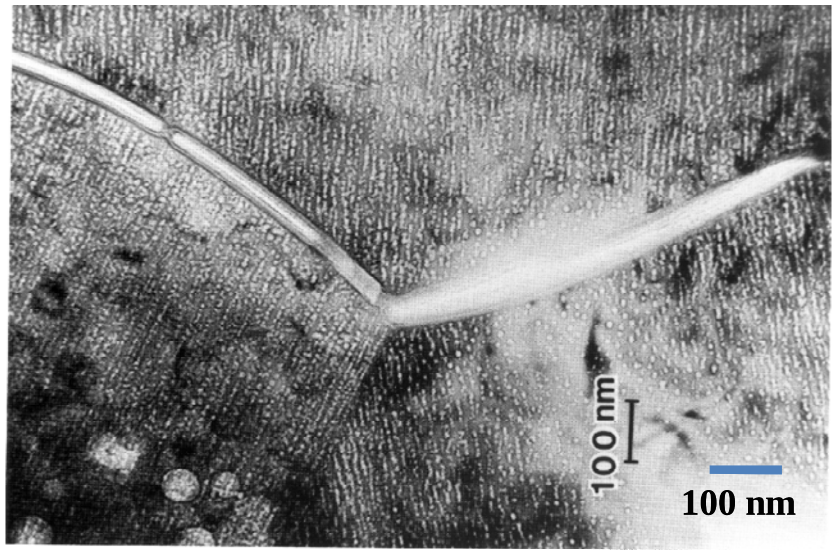
T_{irr}=75-100°C; max dose ~0.5 dpa



B.S. Hickman & D.G. Walker, J. Nucl. Mater. 18 (1966) 197

dose ~10 dpa

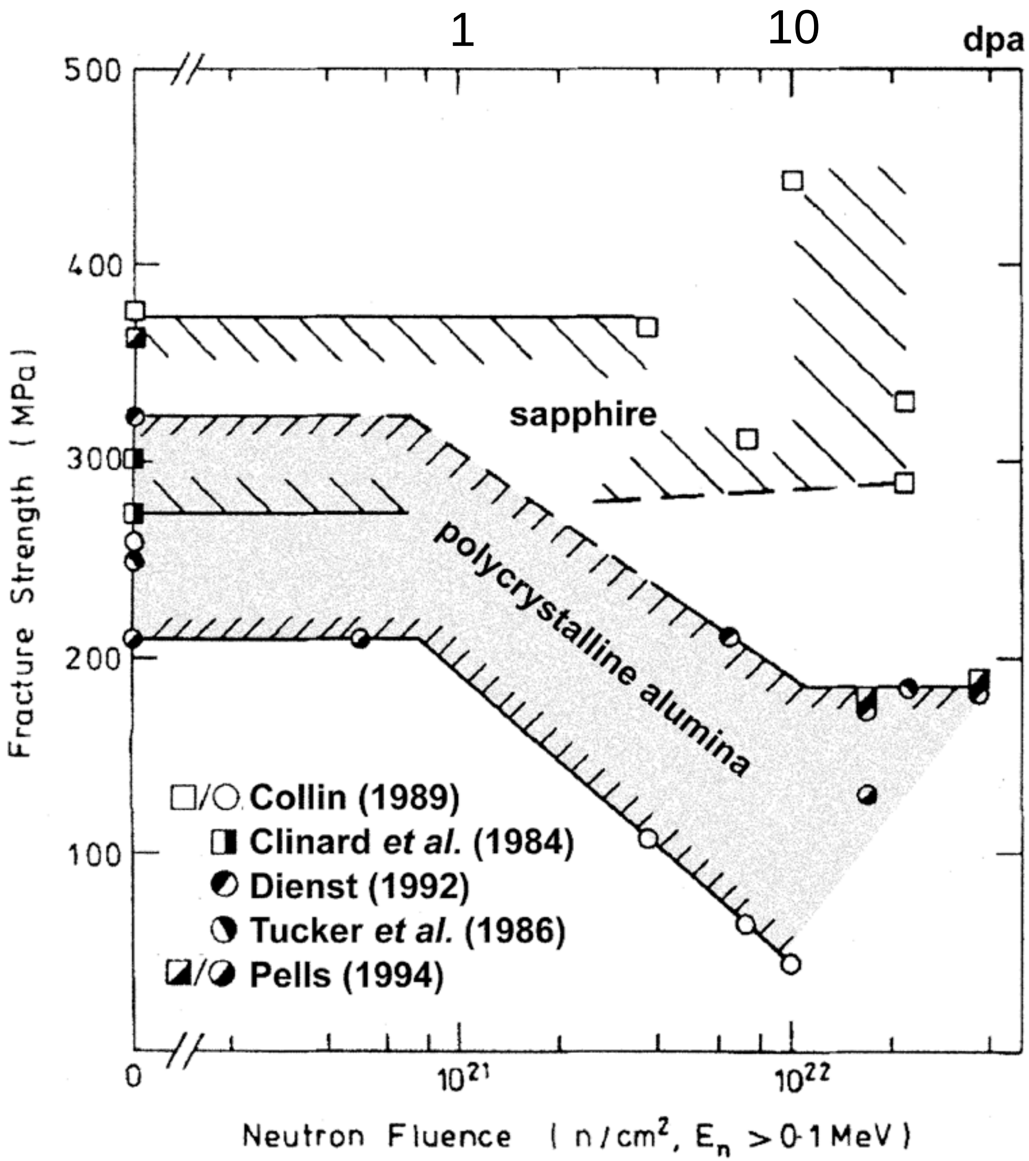




F.W. Clinard, Jr. et al., J. Nucl. Mater. 108&109 (1982) 655

Grain boundary microcracking occurs in Al_2O_3 at high temperatures and doses

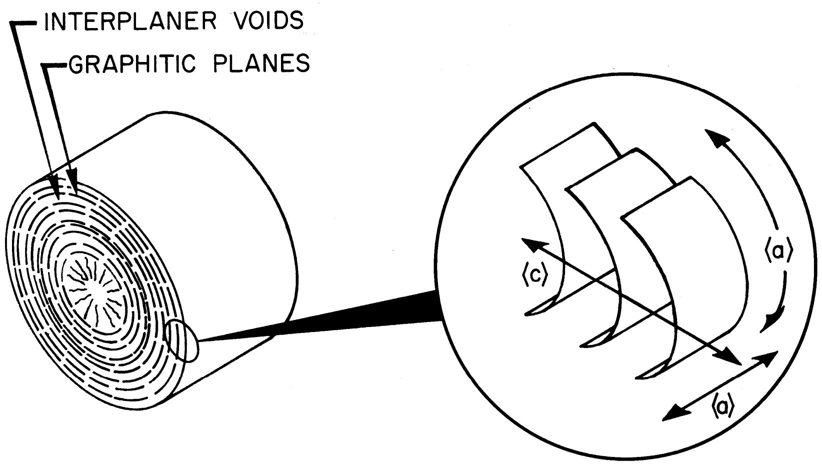
Effect of neutron irradiation on flexural strength of Al_2O_3



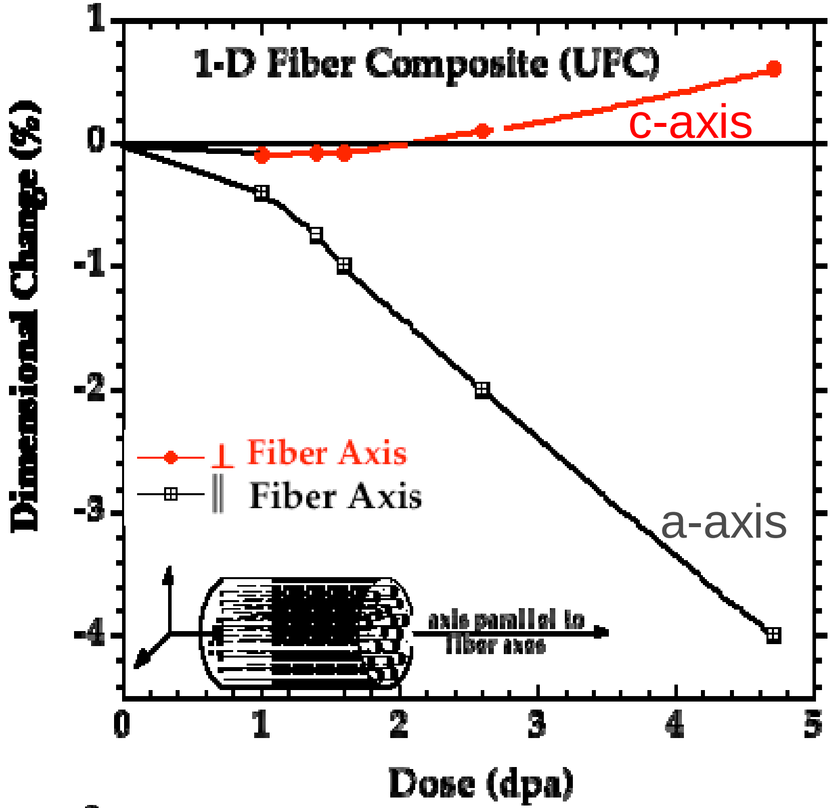
G.P. Pells, J. Amer. Ceram. Soc. 77 (1994) 368

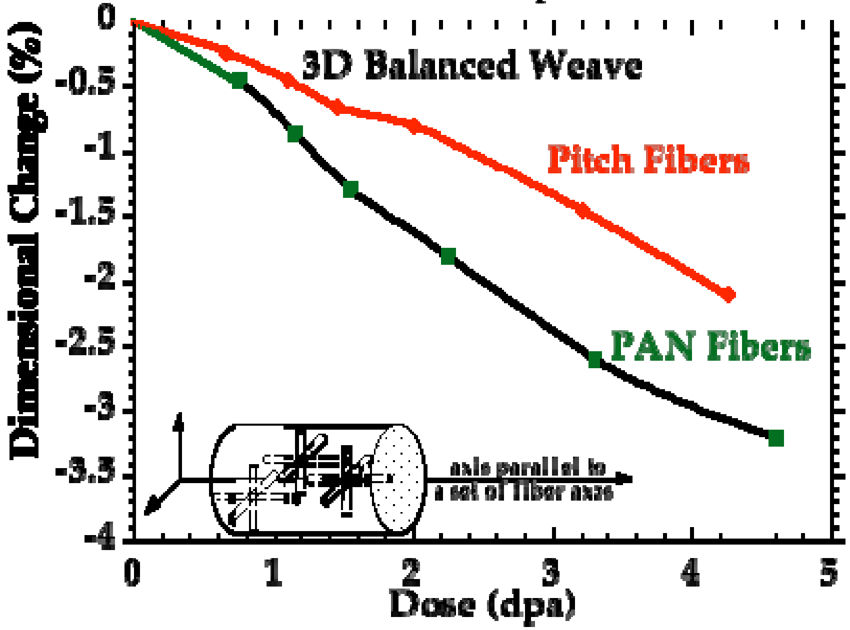
CFC's Under Irradiation

(HFIR , 600°C)

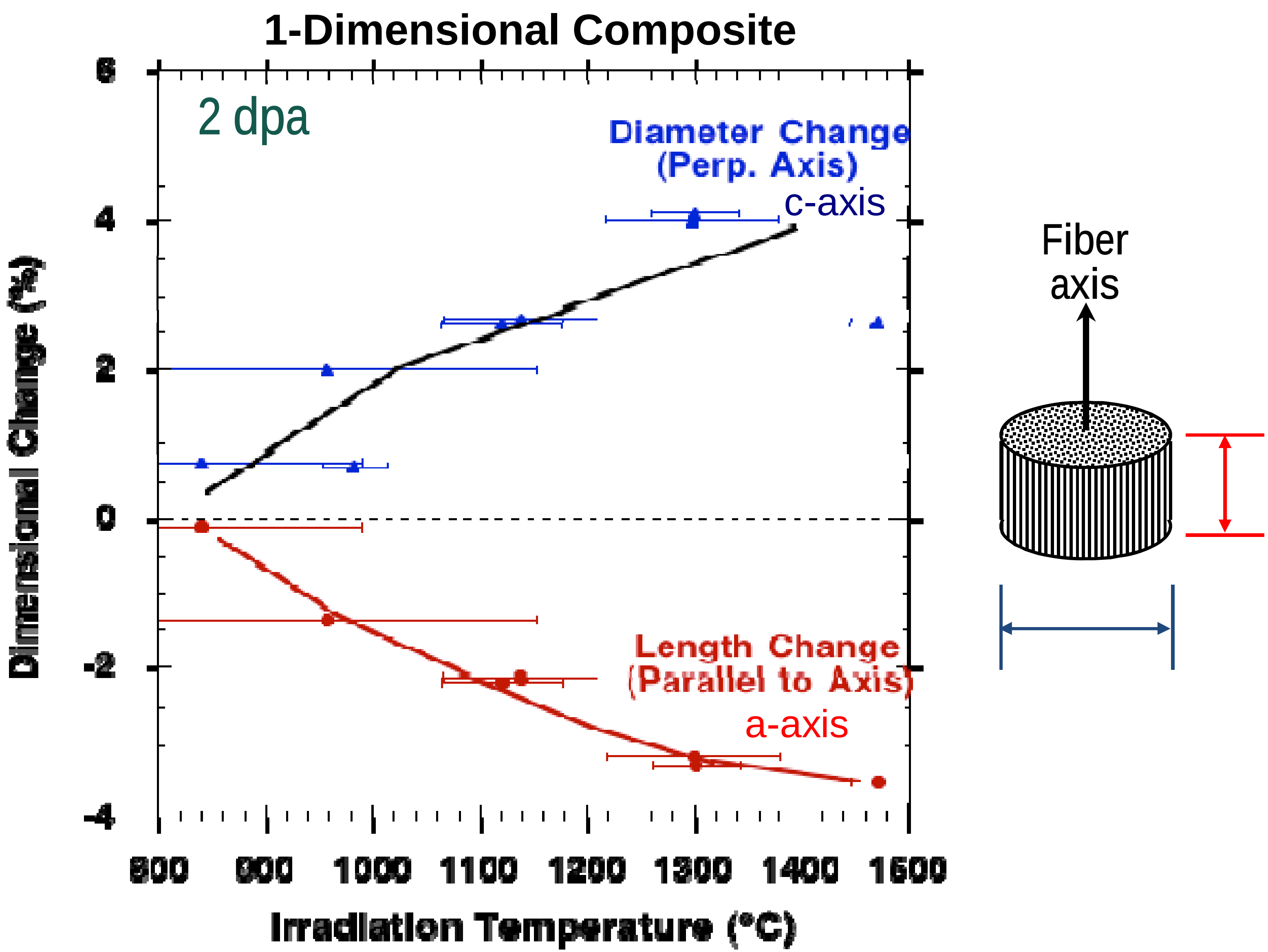


CORE–SHEATH MODEL

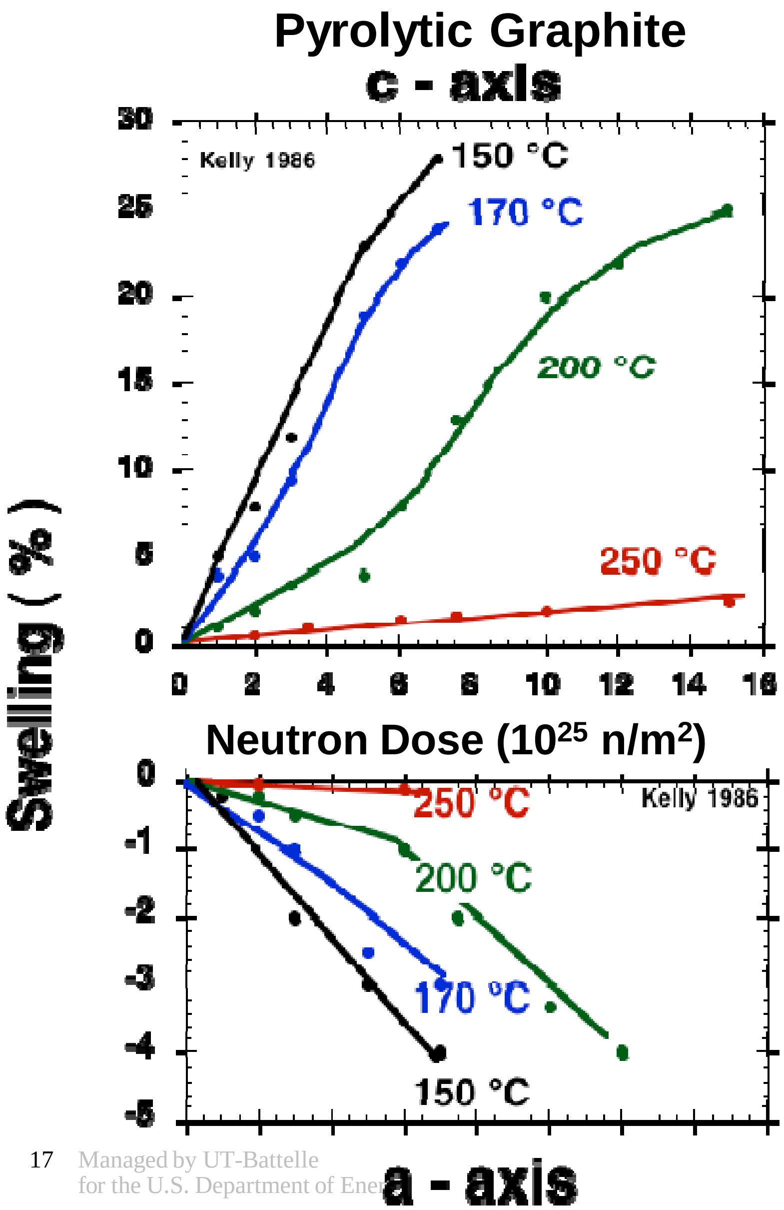


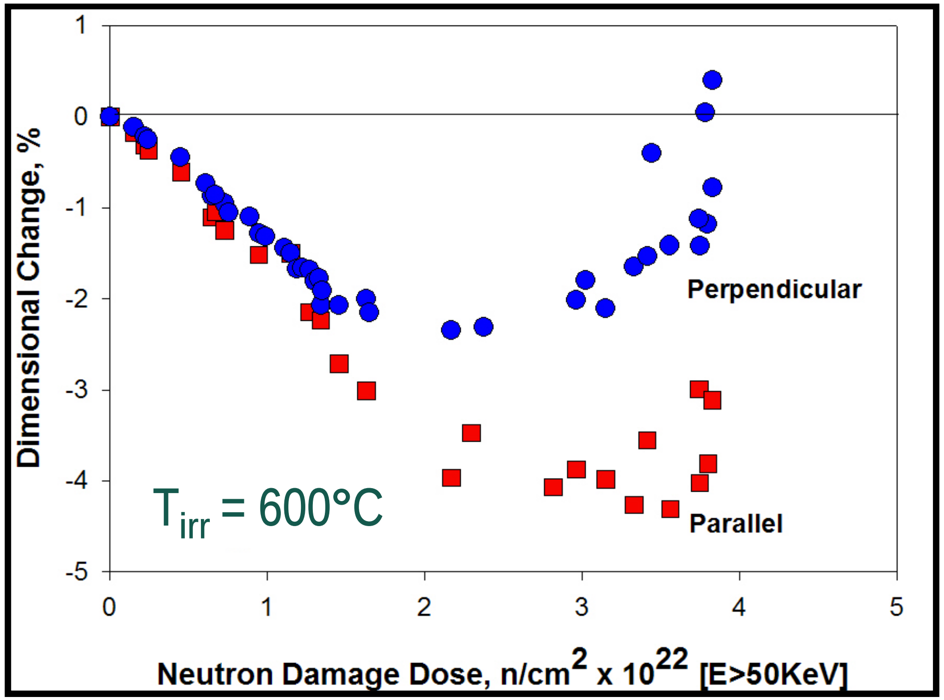


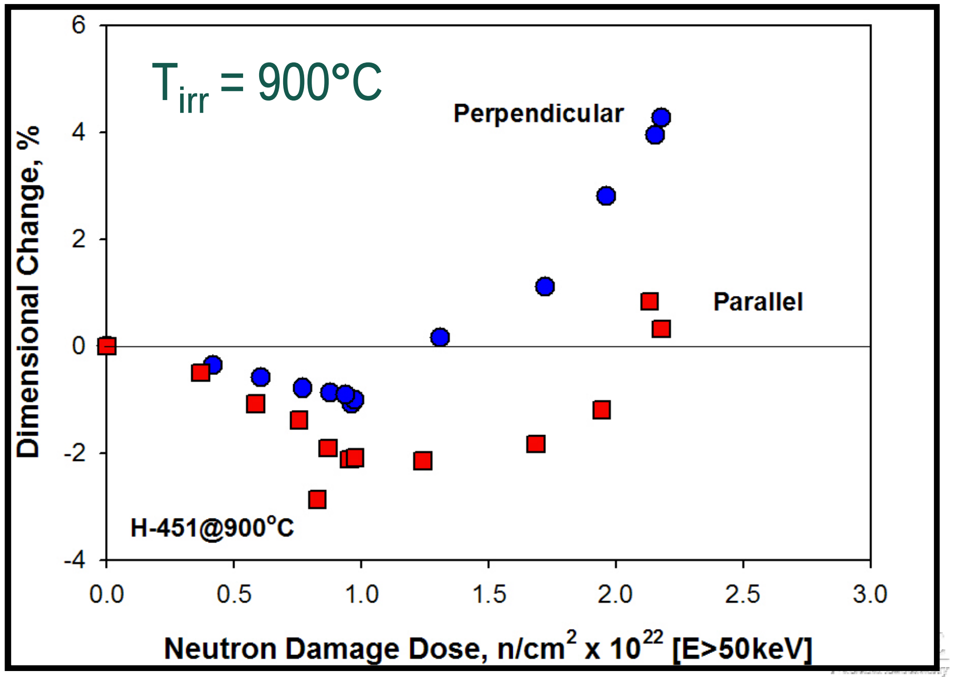
Dimensional Change in 1-D Graphite Composite



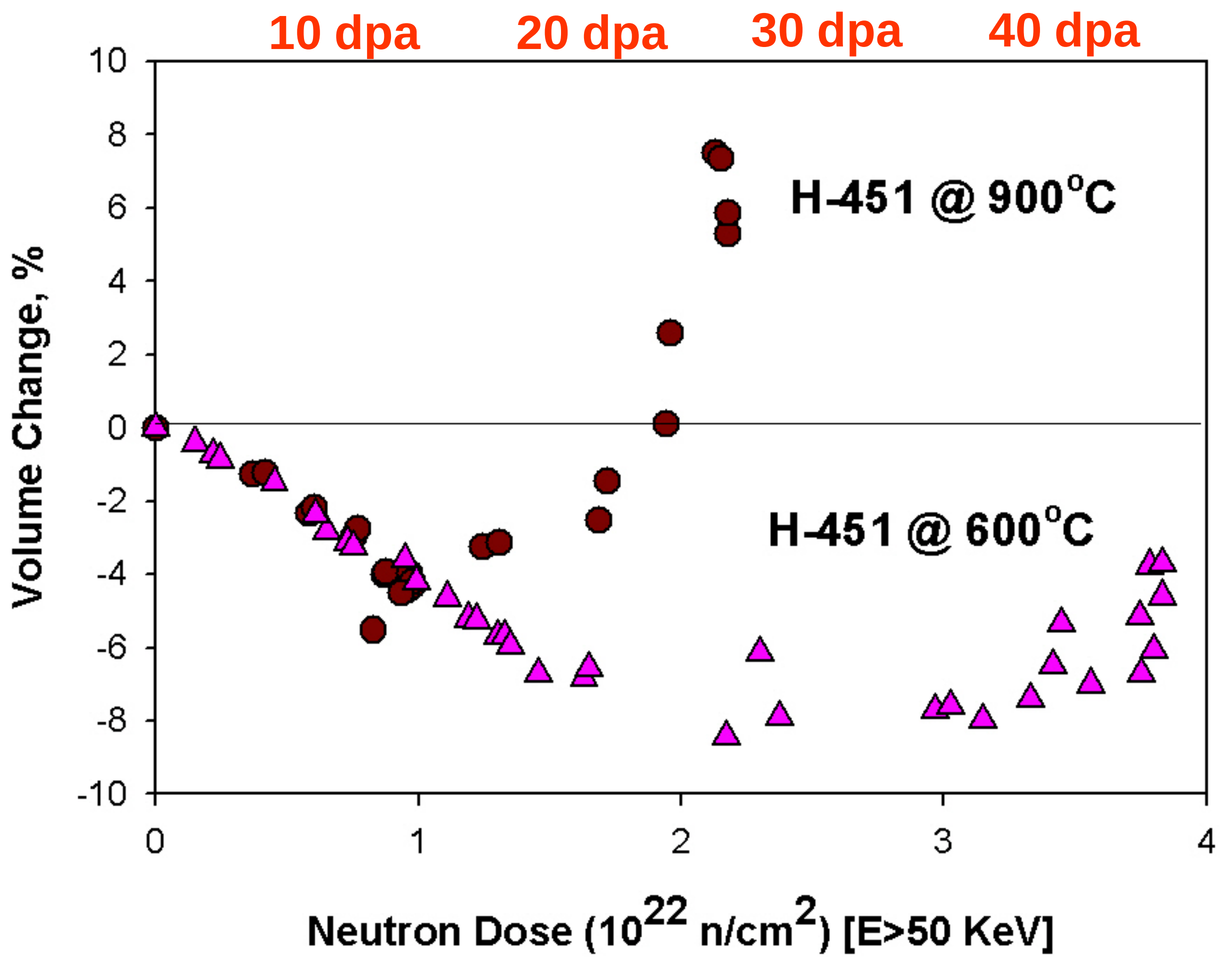
H-451 Nuclear Graphite





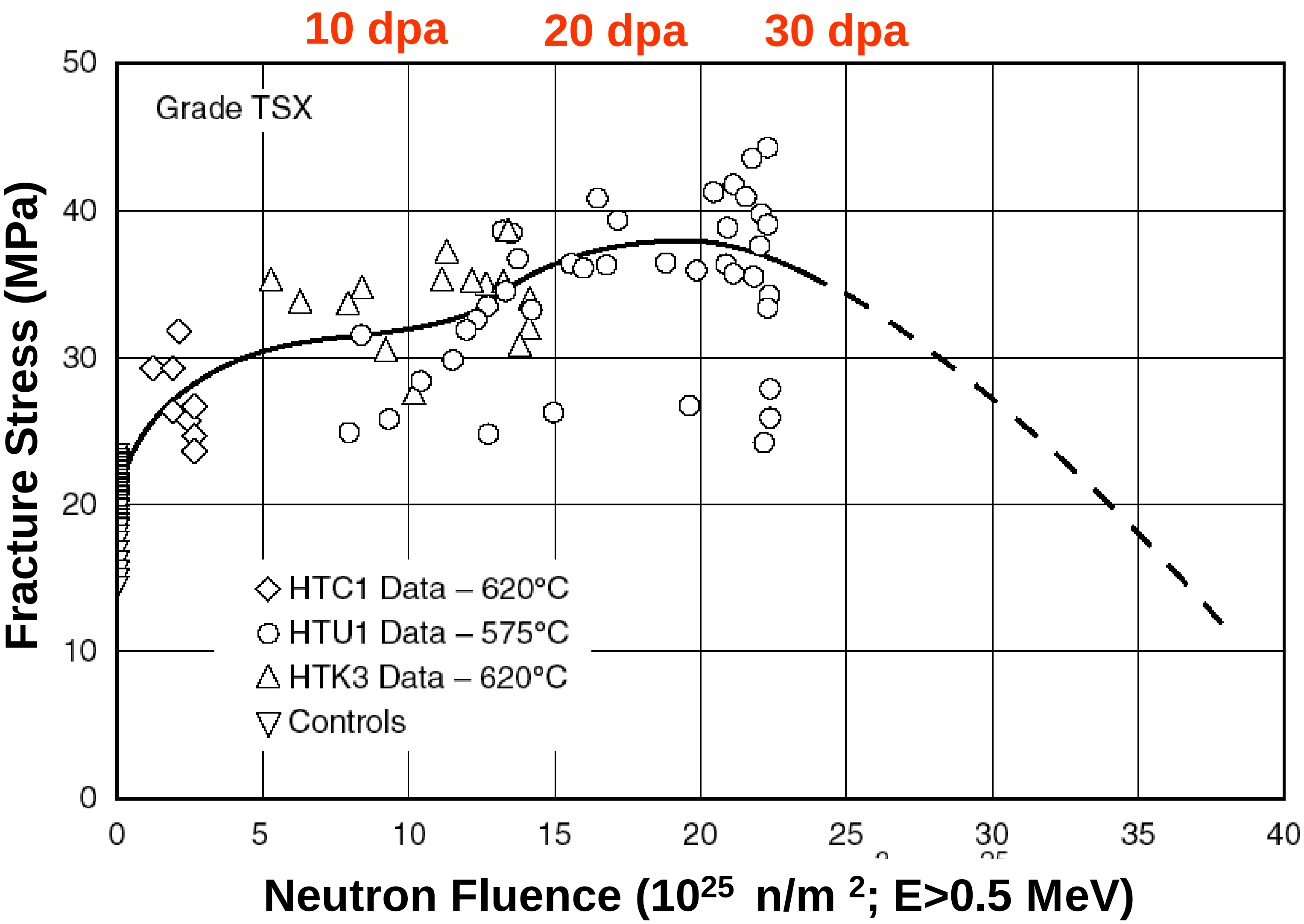


Effect of Temperature and Swelling of Nuclear Graphite



- initial <c> swelling accommodated by closure of intrinsic porosity.
- once porosity filled swelling can begin.
- less initial porosity for higher initial temperature (closure of intrinsic porosity.)

Effect of Irradiation on Strength of Nuclear Graphite



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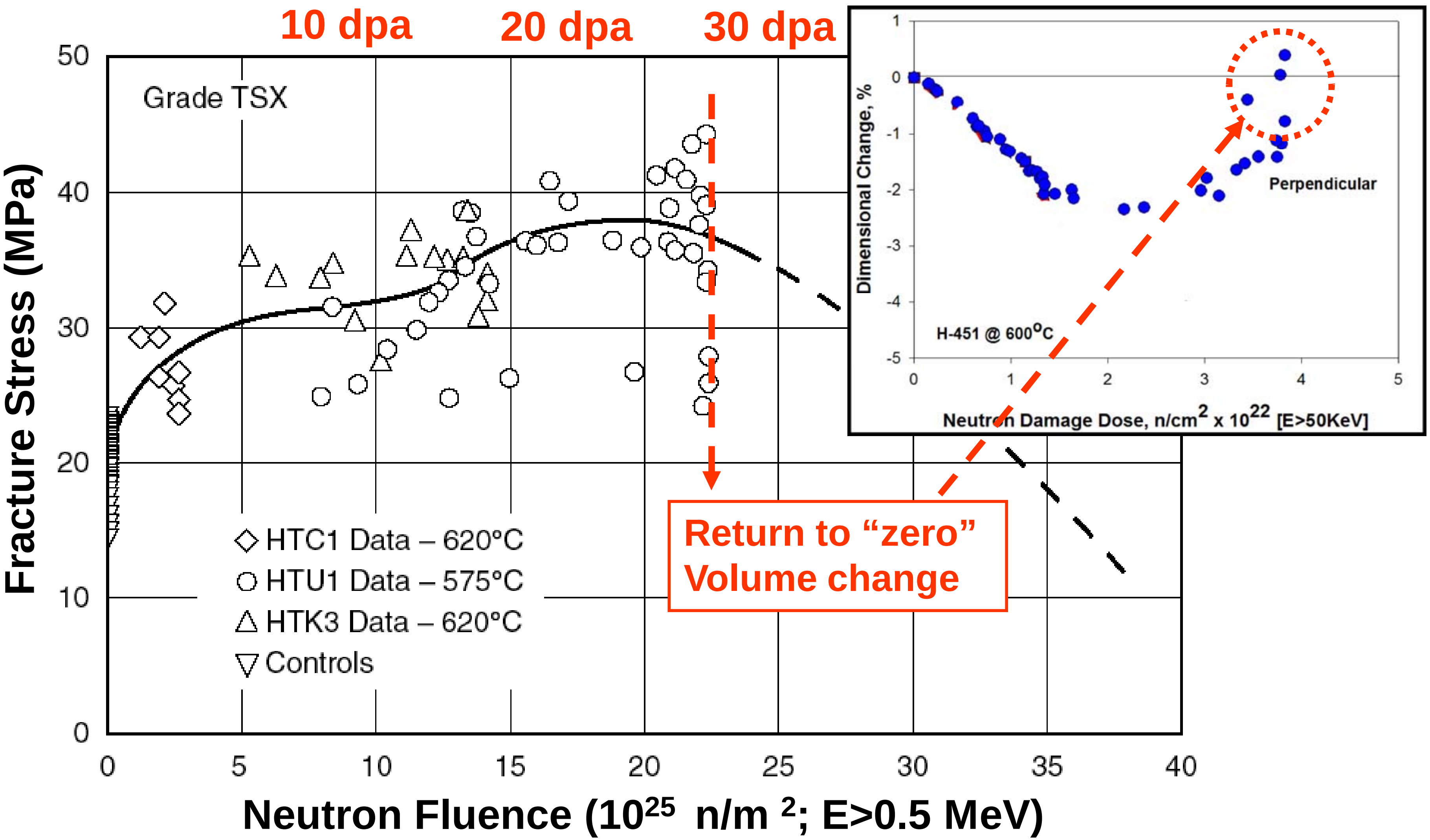
for the U.S. Department of Energy

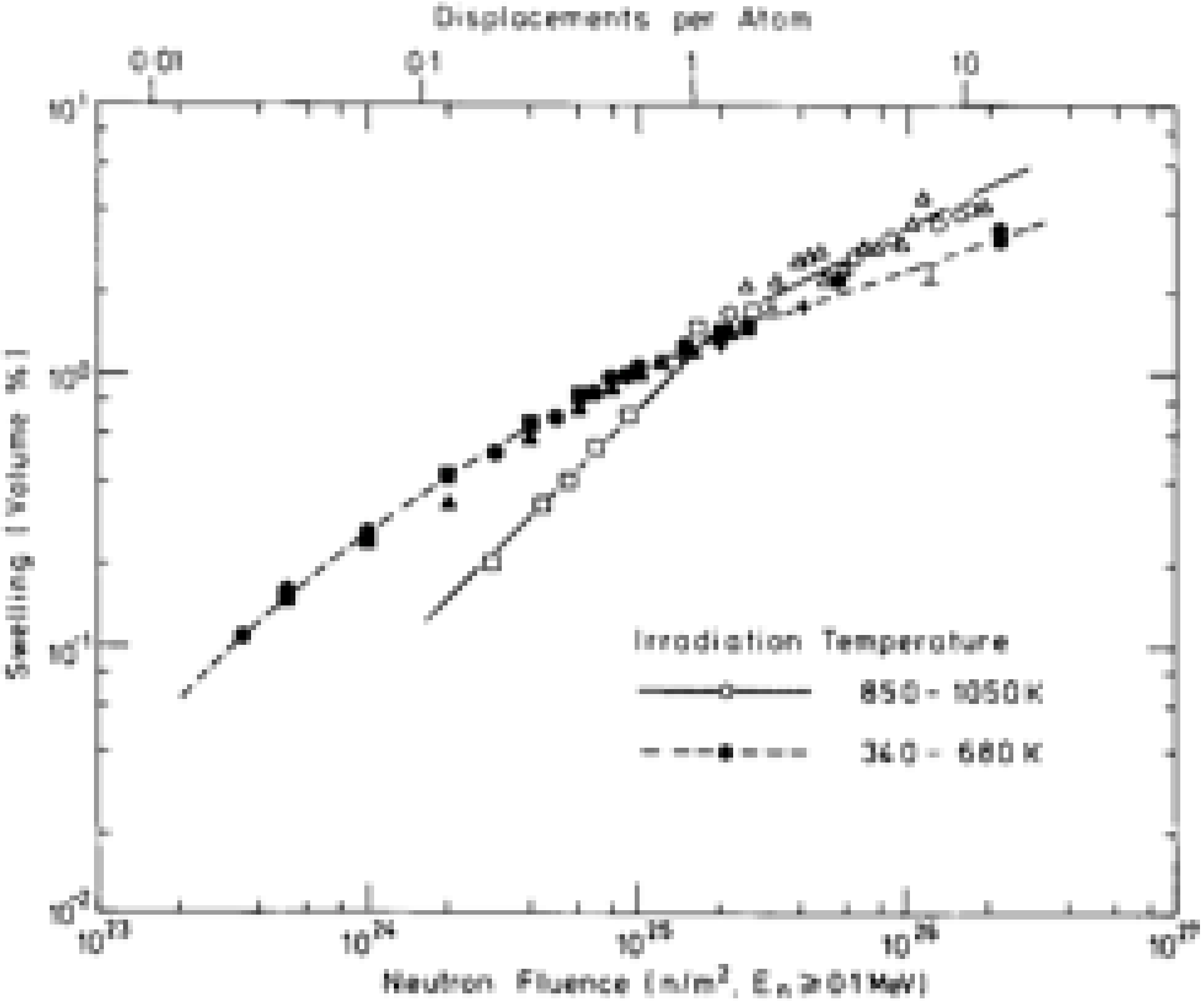
Effect of Irradiation on Strength of Nuclear Graphite



 Managed by UT-Battelle
for the U.S. Department of Energy

OAK RIDGE National Laboratory

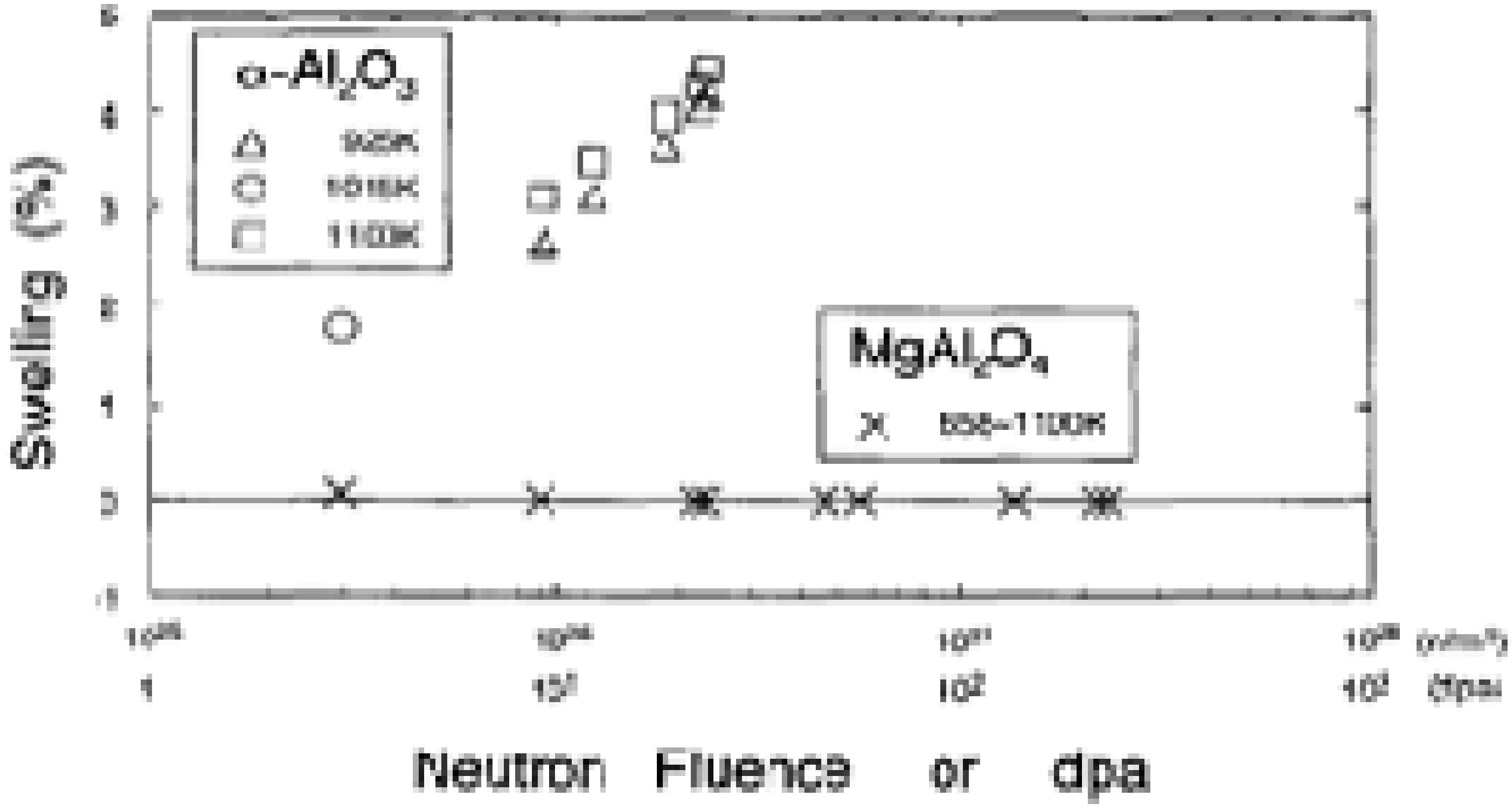
Effect of neutron irradiation on volumetric swelling of Al_2O_3



G.P. Pells, J. Amer. Ceram. Soc. 77 (1994) 368

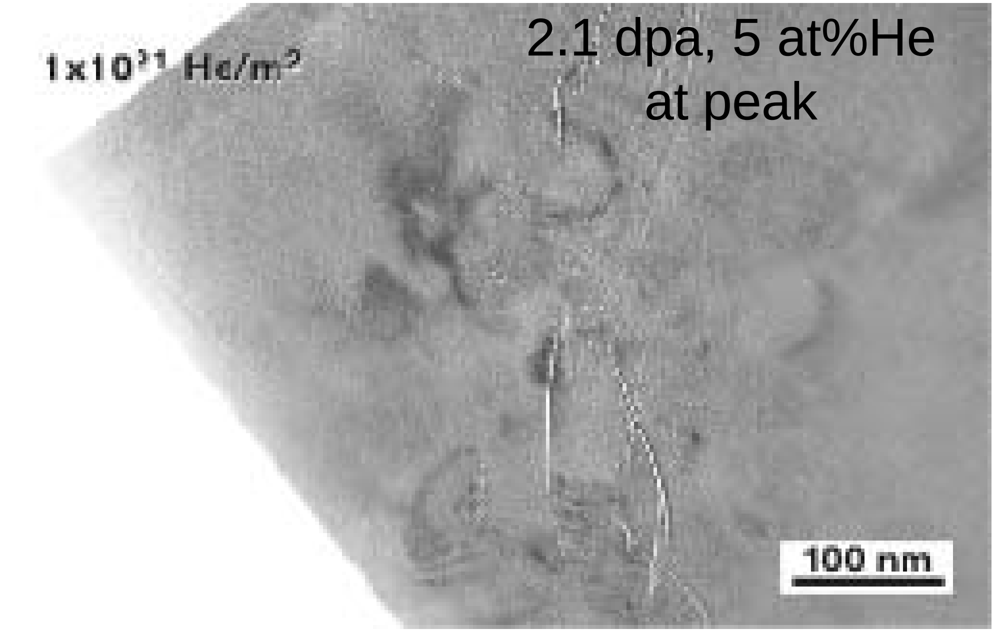
Effect of neutron irradiation on volumetric swelling of single crystal Al_2O_3 and $MgAl_2O_4$

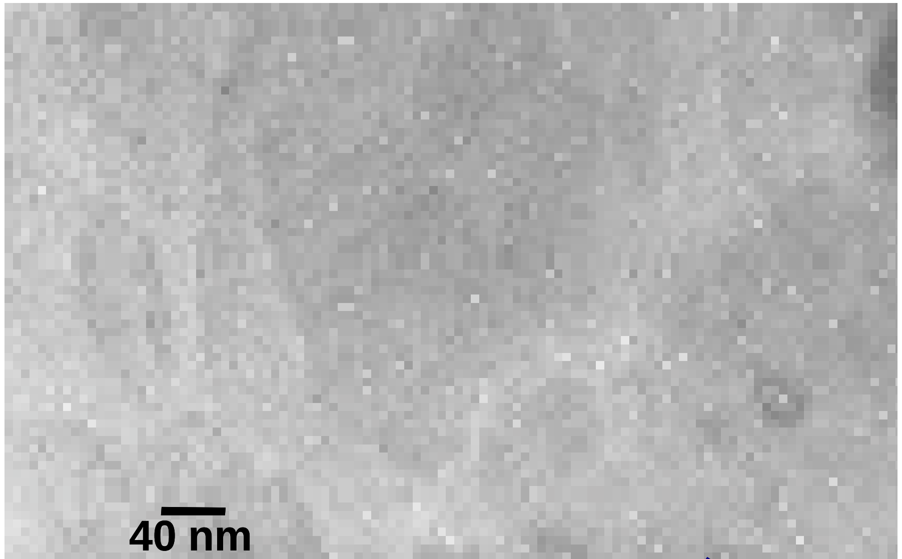
Based on data by Clinard et al. and Garner et al.

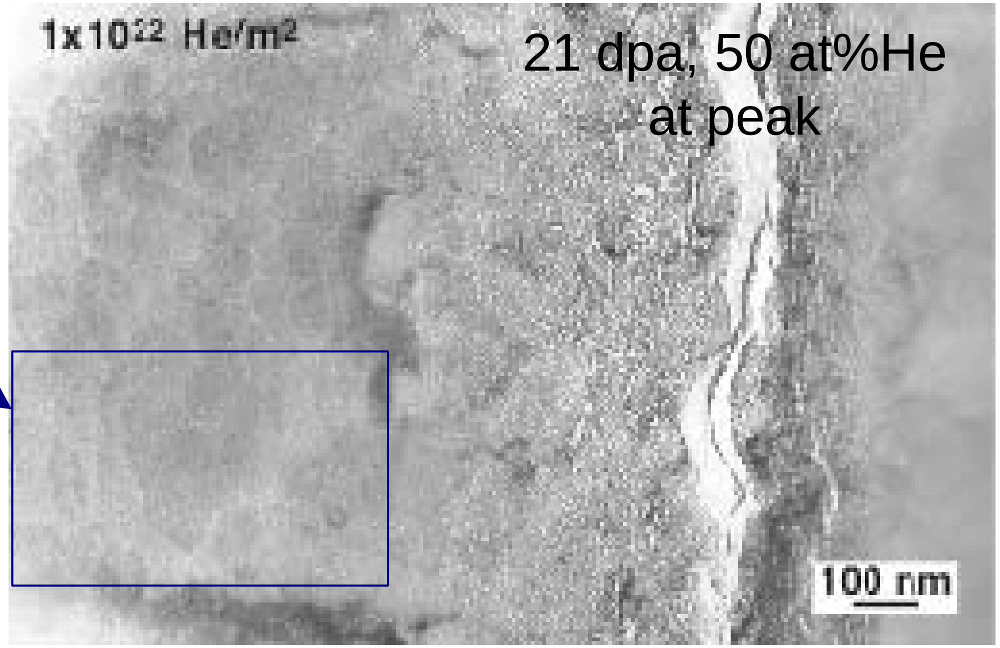


C. Kinoshita & S.J. Zinkle, J. Nucl. Mater. 233-237 (1996) 100

Cavity microstructure of 1 MeV He ion irradiated $MgAl_2O_4$ at 650^{o}C



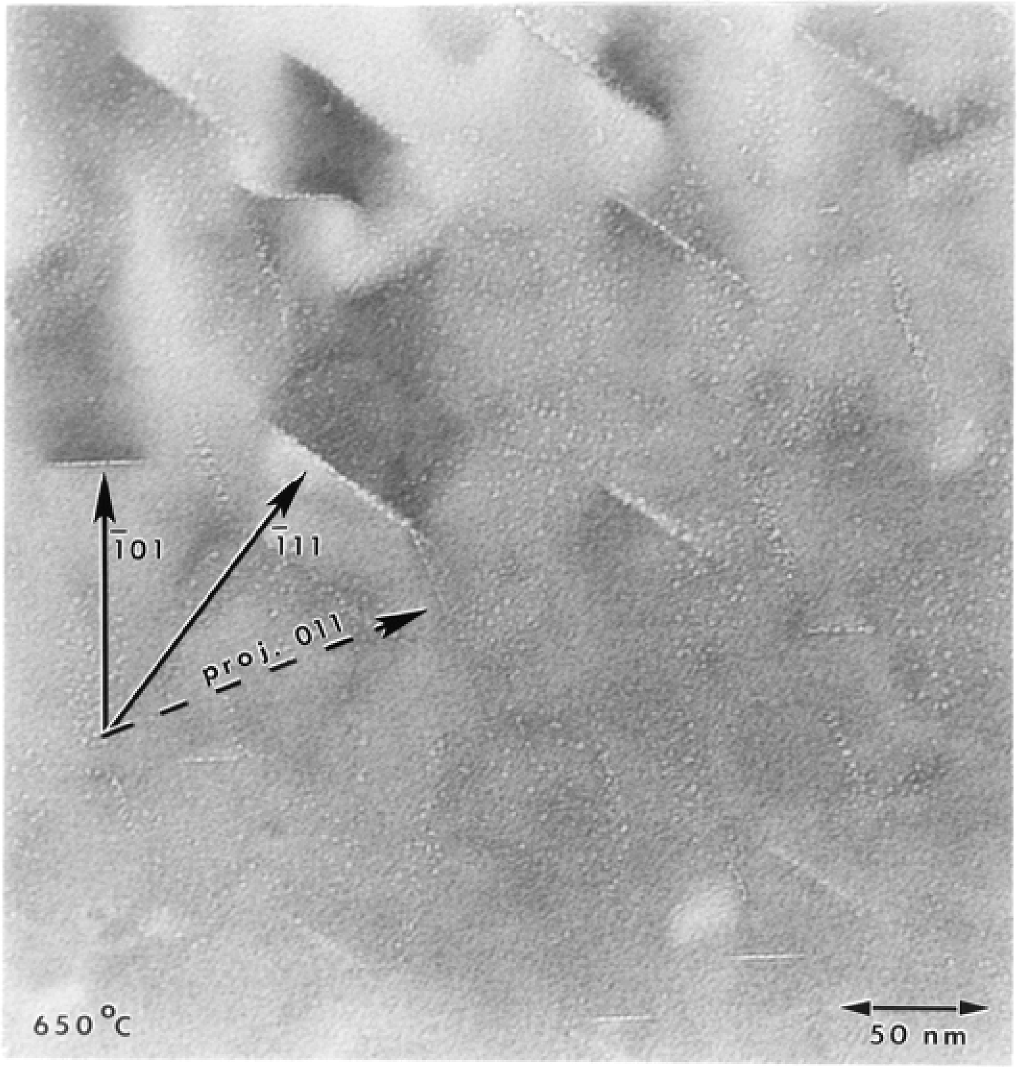




Cavity formation observed in irradiated midrange region >1µm from the peak implanted zone

S.J. Zinkle, Nucl. Instrum. Meth. B (2012) in press

Preferential cavity formation on dislocation loops in dual ion beam irradiated $MgAl_2O_4$ at 650°C

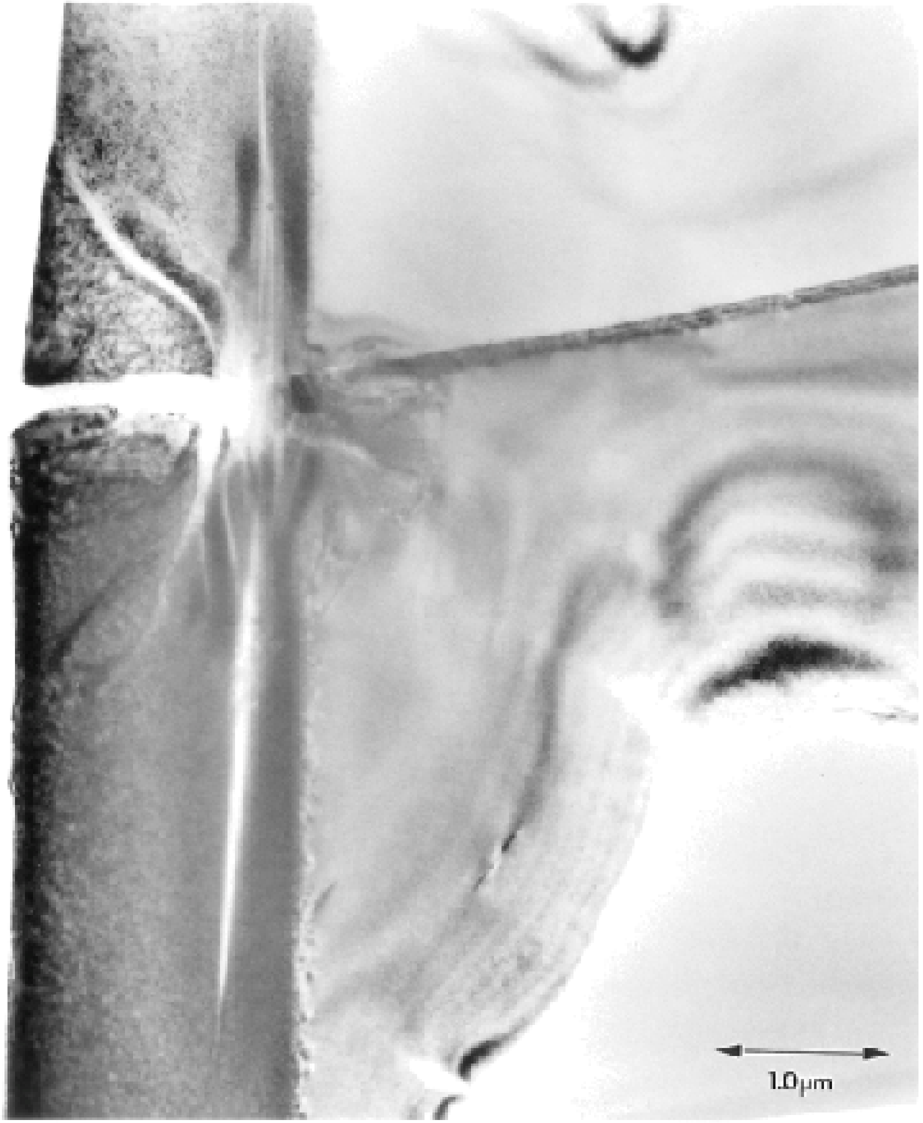


2 MeV Al +
200-400 keV He

26 dpa,
3900 appm He

S.J. Zinkle & S. Kojima, J. Nucl. Mater. 179-181 (1991) 395

Grain boundary cavitation in dual ion beam irradiated $MgAl_2O_4$ at 650^oC

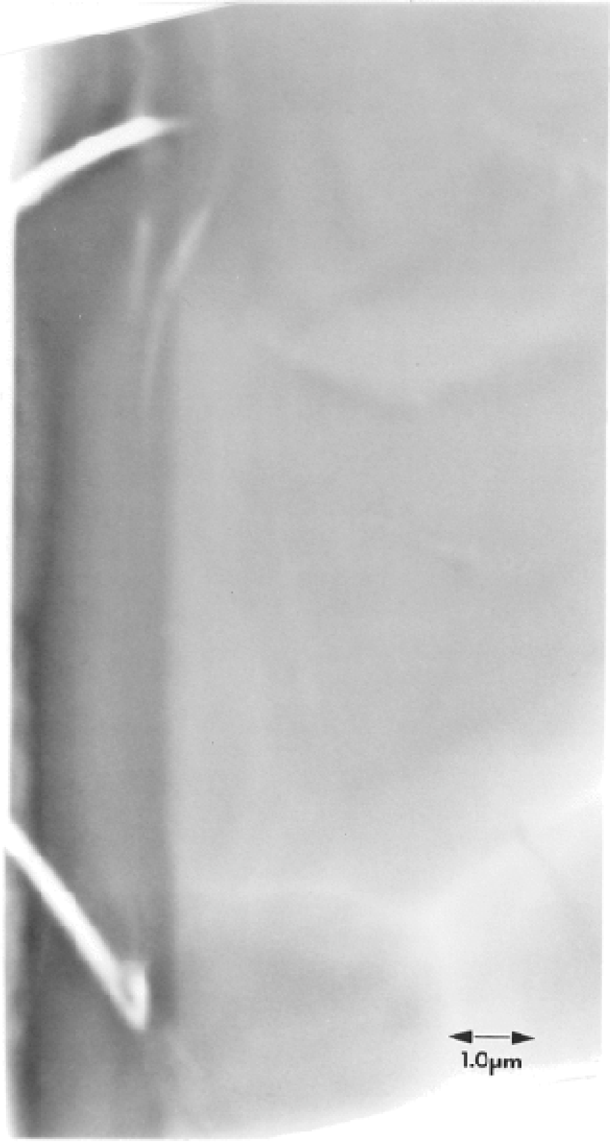


2 MeV Al +
200-400 keV He

26 dpa,
3900 appm He
at ~0.8 μm depth

S.J. Zinkle & S. Kojima, J. Nucl. Mater. 179-181 (1991) 395

Grain boundary cavitation in dual ion beam irradiated $MgAl_2O_4$ at 650°C

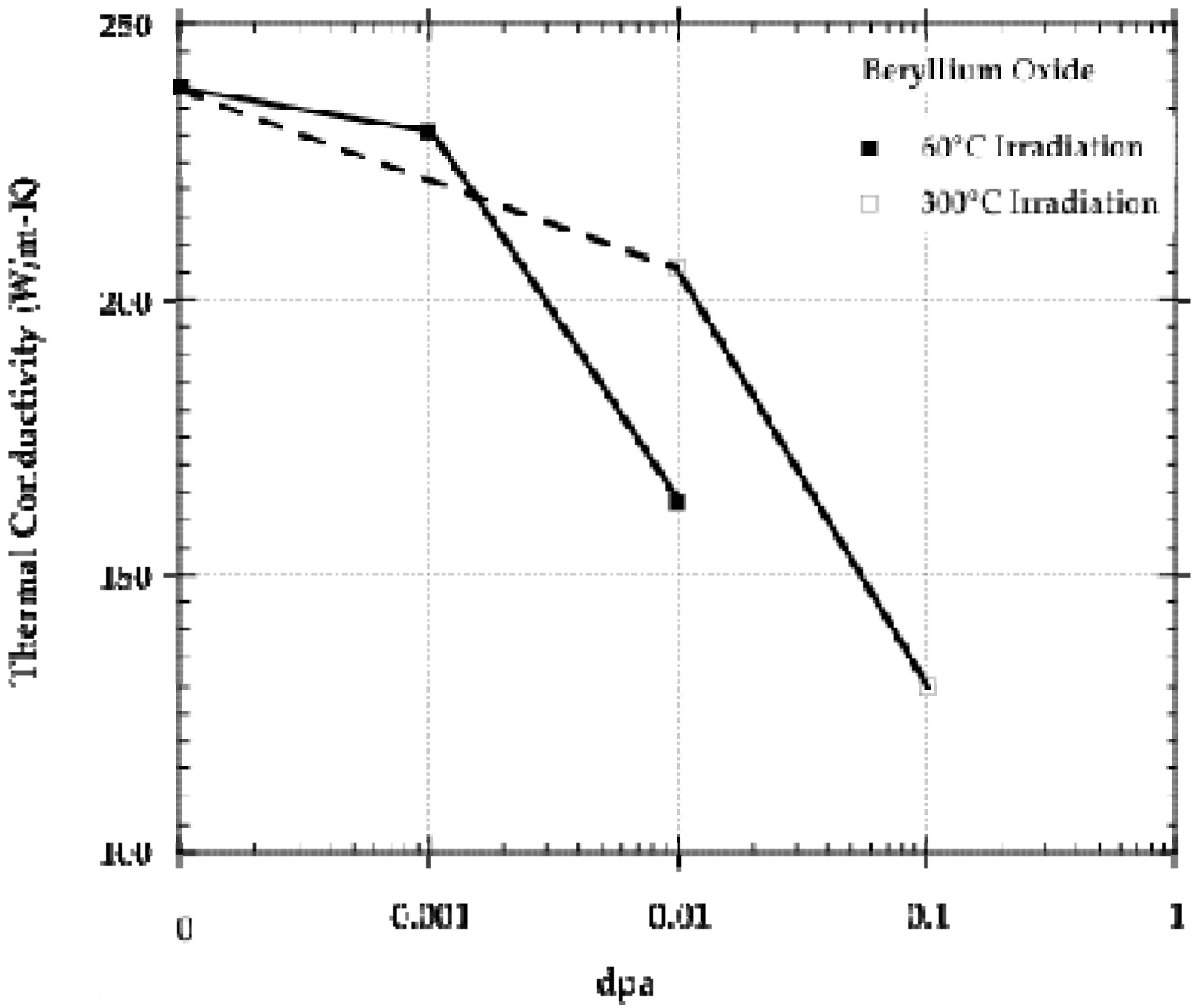


2 MeV Al +
200-400 keV He

26 dpa,
3900 appm He
at ~0.8 μm depth

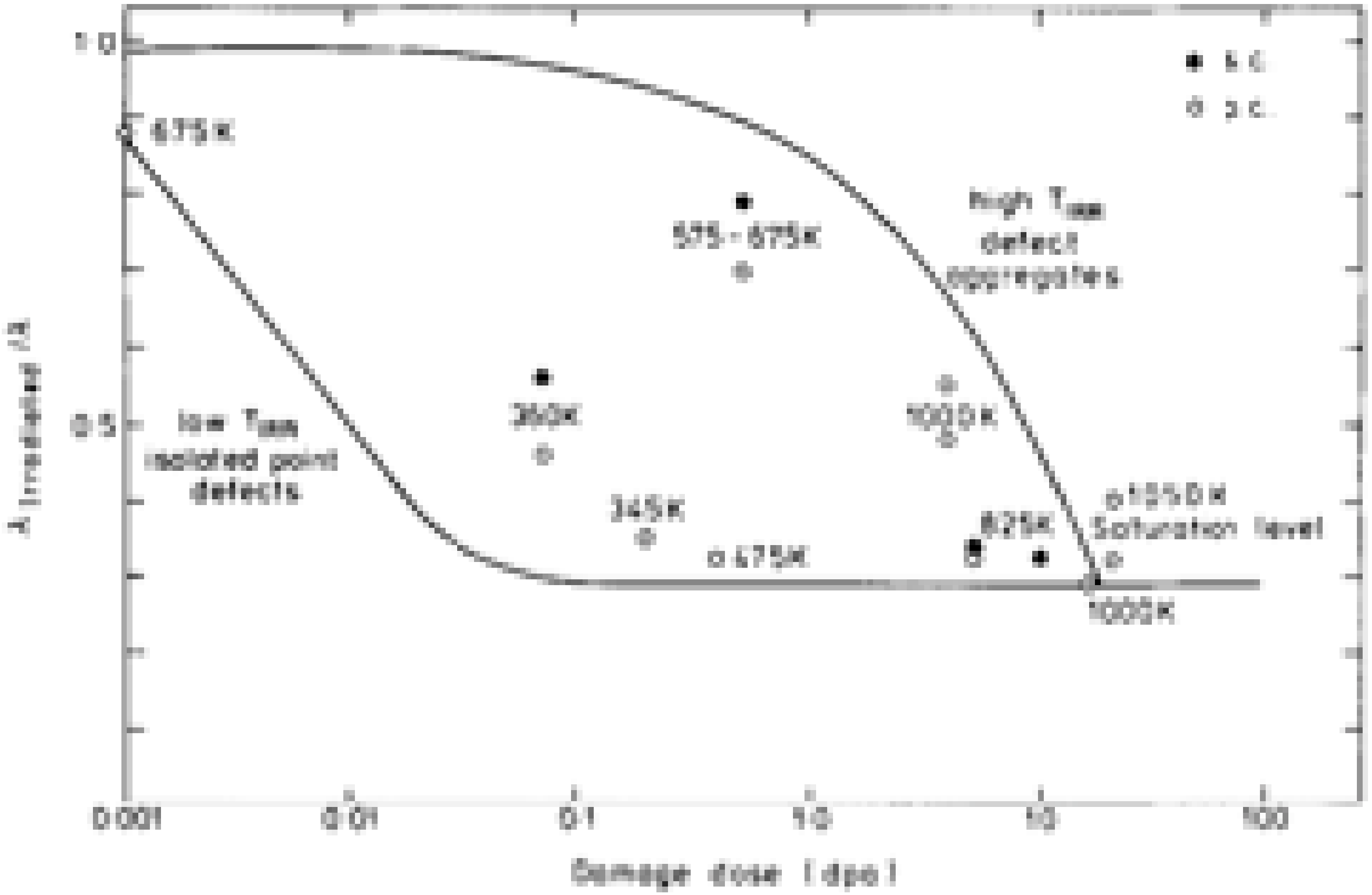
S.J. Zinkle & S. Kojima, J. Nucl. Mater. 179-181 (1991) 395

Effect of low temperature neutron irradiation on thermal conductivity of BeO



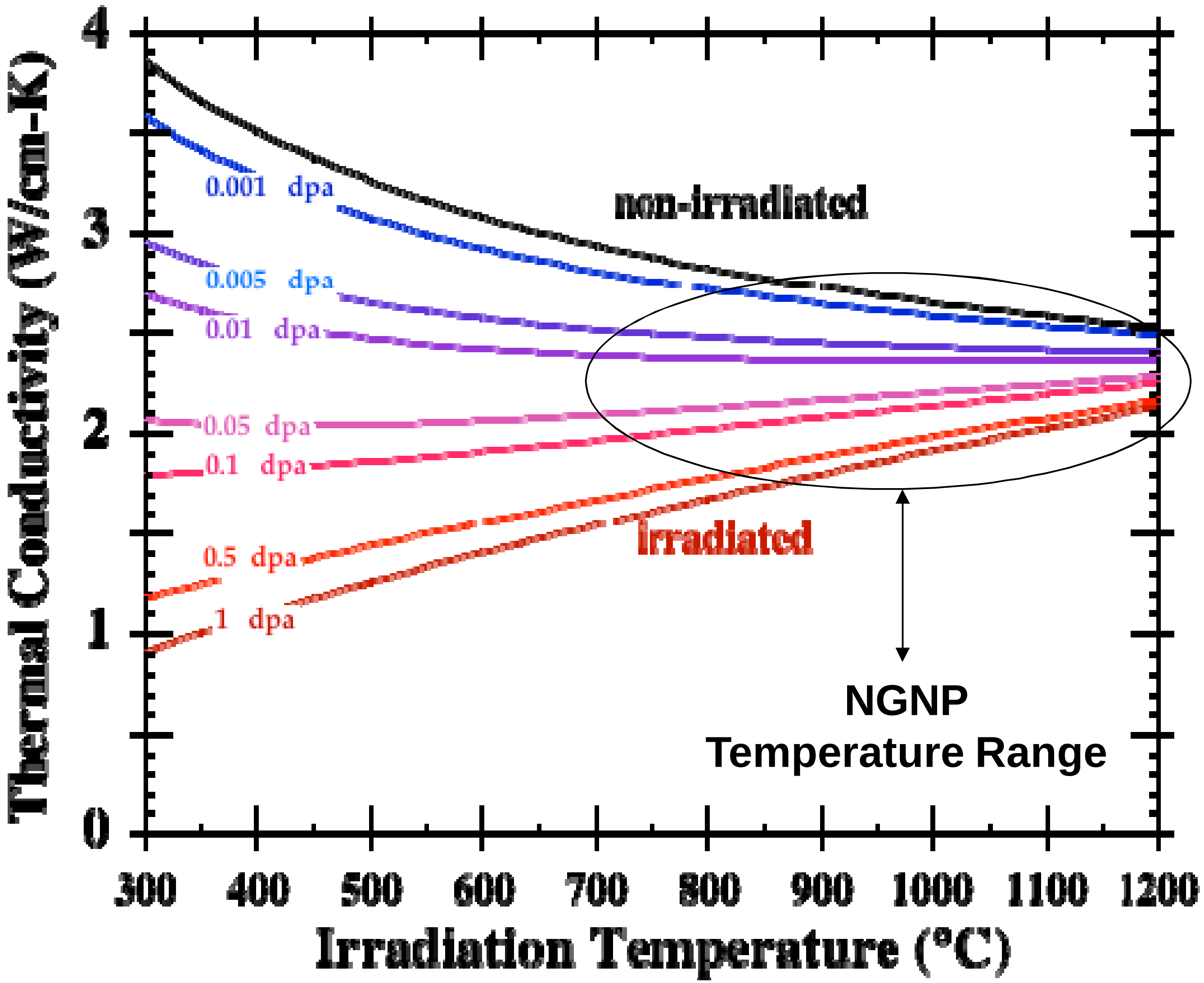
L.L. Snead, S.J. Zinkle & D.P. White, J. Nucl. Mater. 340 (2005) p. 187

Effect of neutron irradiation on thermal conductivity of Al_2O_3

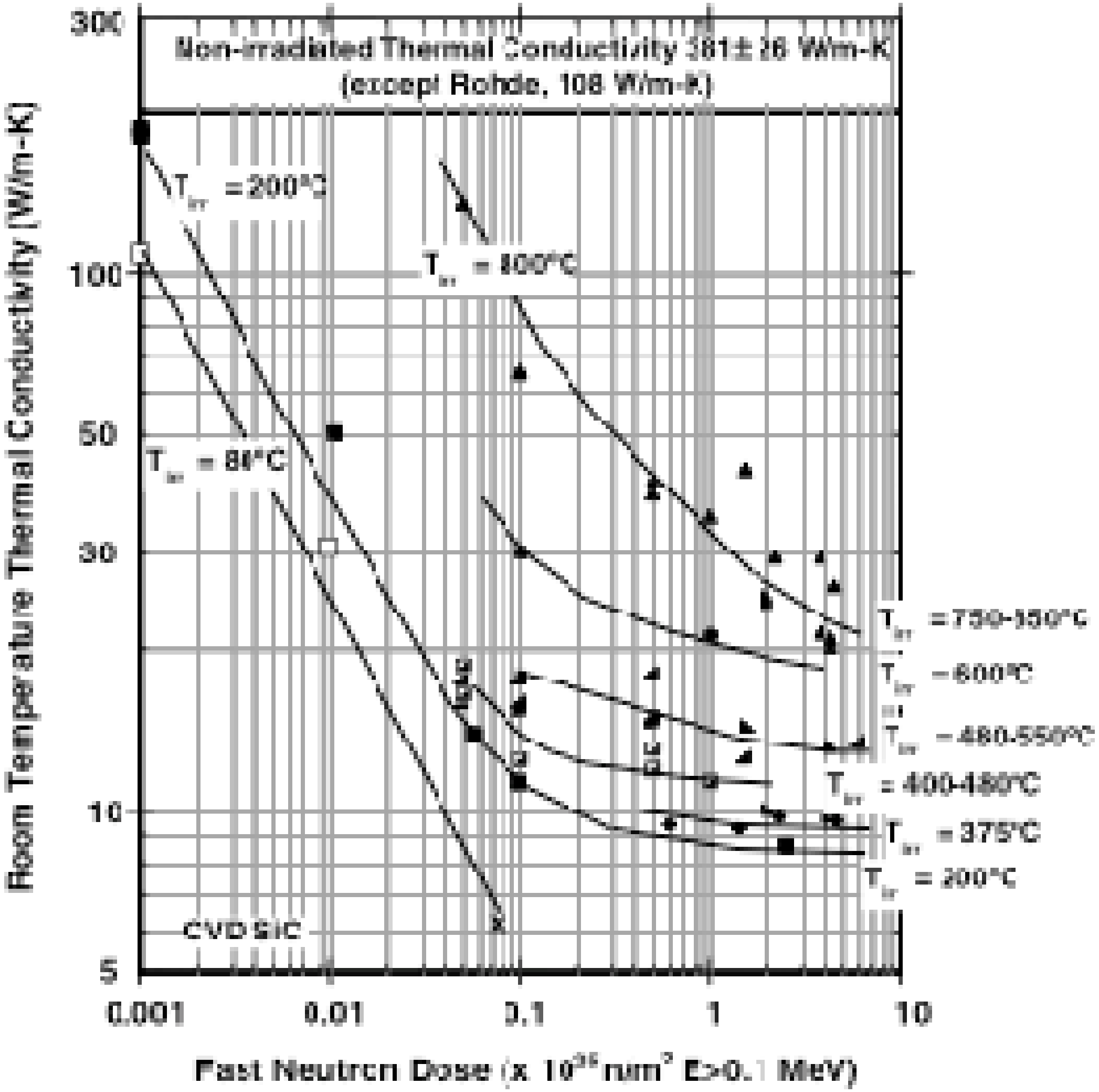


G.P. Pells, J. Amer. Ceram. Soc. 77 (1994) 368

Thermal Conductivity Degradation in High-conductivity Carbon Composite

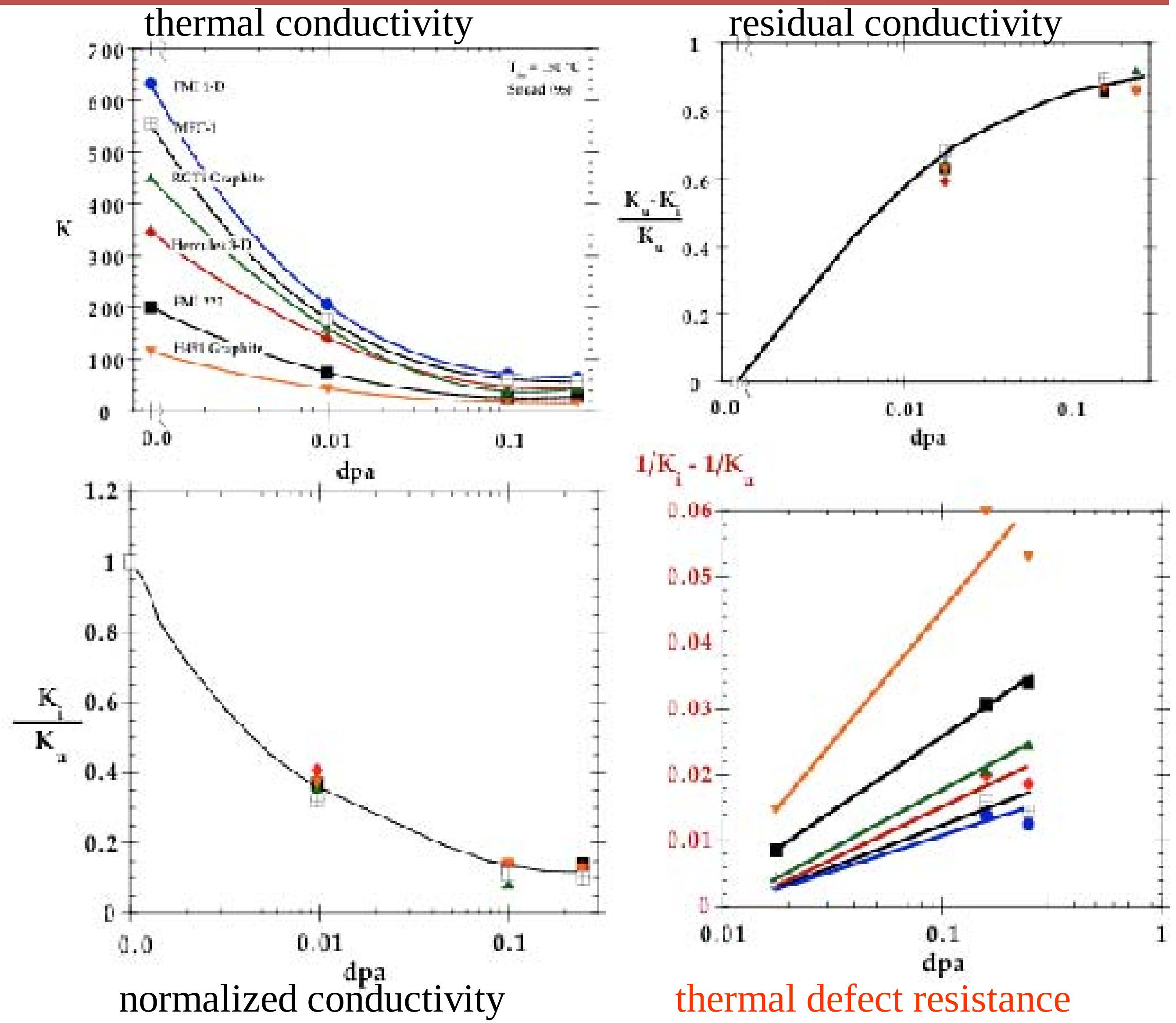


Effect of neutron irradiation on thermal conductivity of SiC

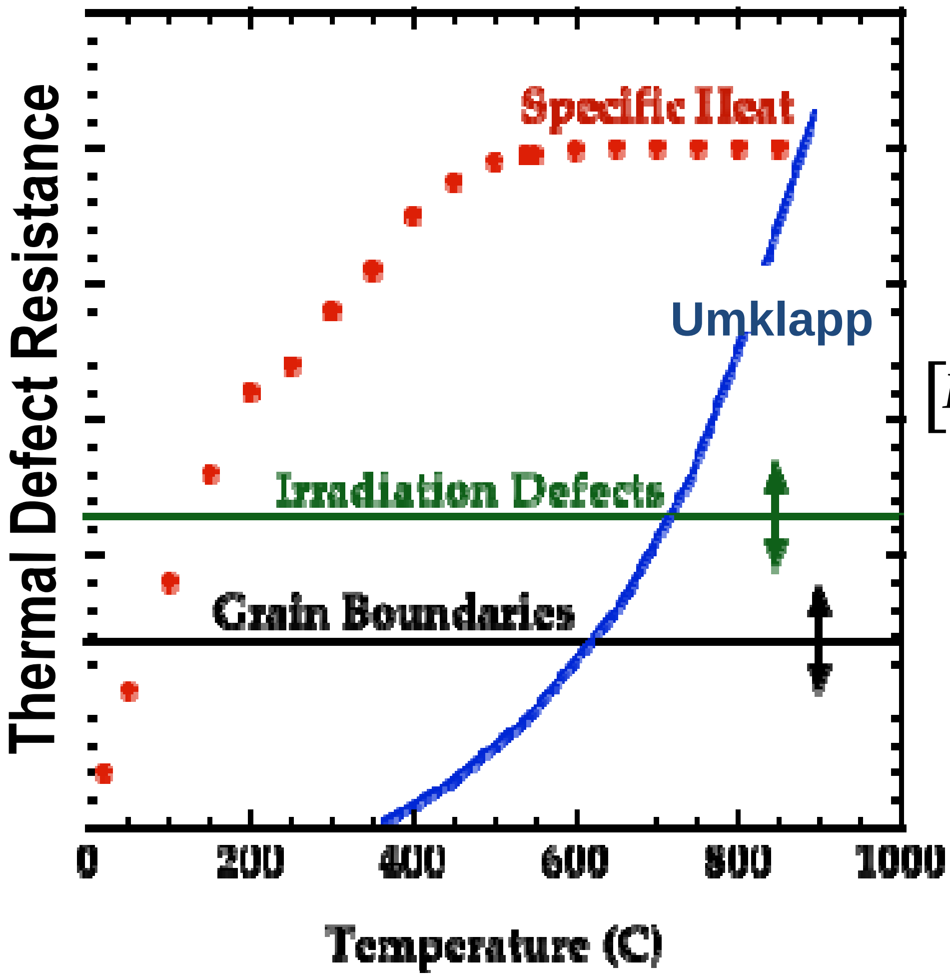


L.L. Snead et al., J. Nucl. Mater. 371 (2007) 329

What is the best parameter to characterize radiation-induced degradation of the thermal conductivity of ceramics?



Thermal Conductivity of Ceramics



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

radiation defects

intrinsic defects

boundaries

umklapp

L.L. Snead, S.J. Zinkle & D.P. White, J. Nucl. Mater. 340 (2005) p. 187

Adopting Thermal Defect Resistance

Thermal defect resistance

$$\frac{1}{K_{rd}} = \frac{1}{K_{irr}} - \frac{1}{K_{unirr}}$$

Thermal defect resistance due to low vacancy production

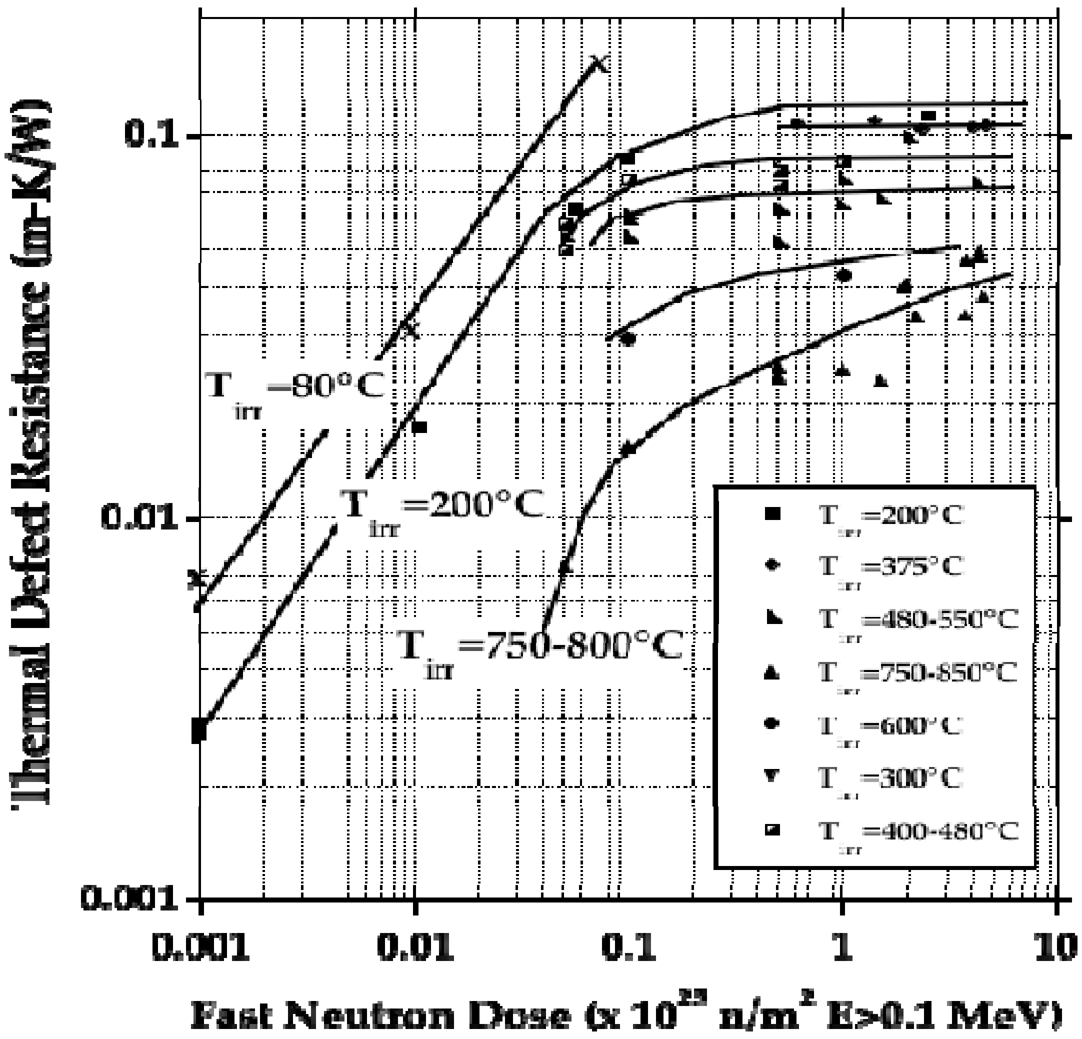
$$\frac{1}{K_{rd}} = \left(\frac{3\pi}{2k_B}\right)\left(\frac{\Omega\omega_D}{v^2}\right)C_v$$

The motivation for using thermal defect resistance is that radiation-induced defects, such as vacancies and clusters, have resistances proportional (or square root dependent) to their concentration and are additive. This gives an easy way to compare stability of ceramics under irradiation.

L.L. Snead, S.J. Zinkle & D.P. White, J. Nucl. Mater. 340 (2005) p. 187

$$\frac{1}{K_{tdr}} = \frac{1}{K_{irradiated}} - \frac{1}{K_{non-irradiated}}$$

TDR in Point Defect Swelling Regime



Vacancies

For Weak Scattering

$$\Delta\left(\frac{1}{K}\right)_{vac} = \frac{1}{3K_i}\left(\frac{\omega_D}{\omega_p}\right)^2 = \frac{3\pi\Omega\omega_D}{2k_B v^2}C_v$$

For Strong Scattering

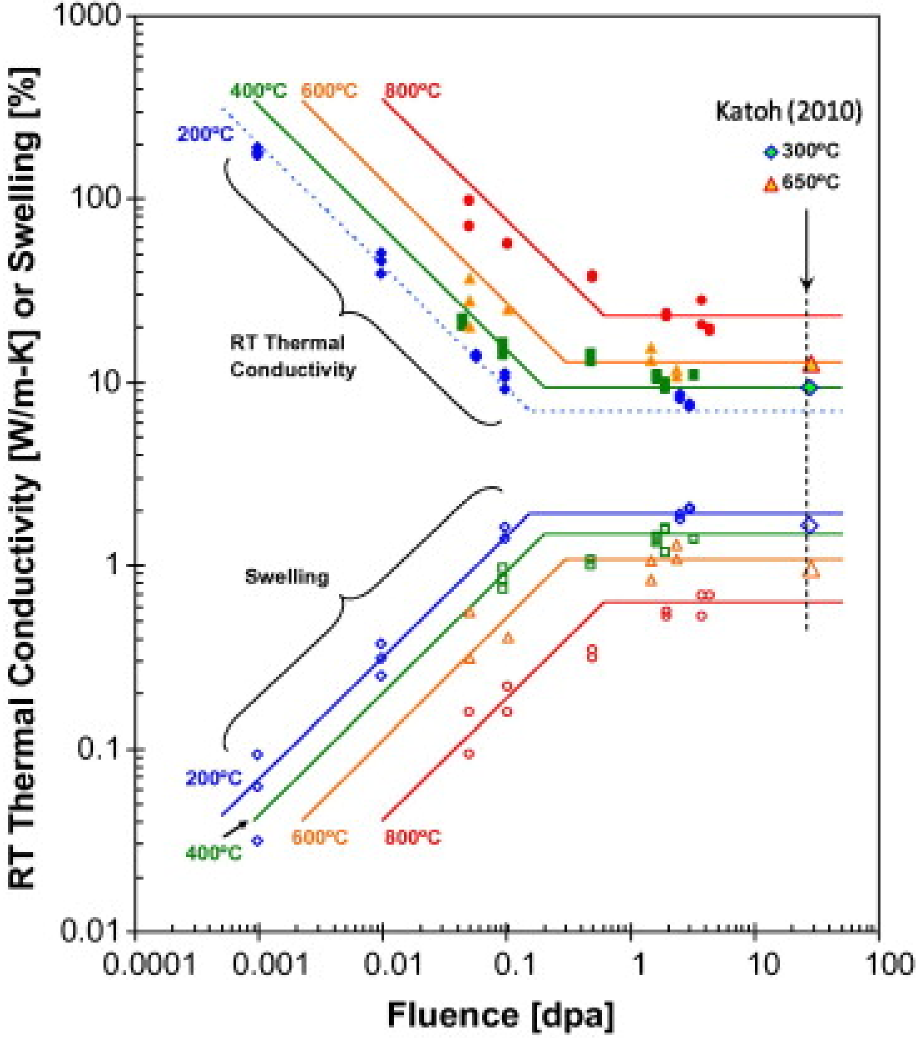
$$\Delta\left(\frac{1}{K}\right)_{vac} = \frac{2}{\pi K_i}\left(\frac{\omega_D}{\omega_p} - \frac{\pi}{2}\right)$$

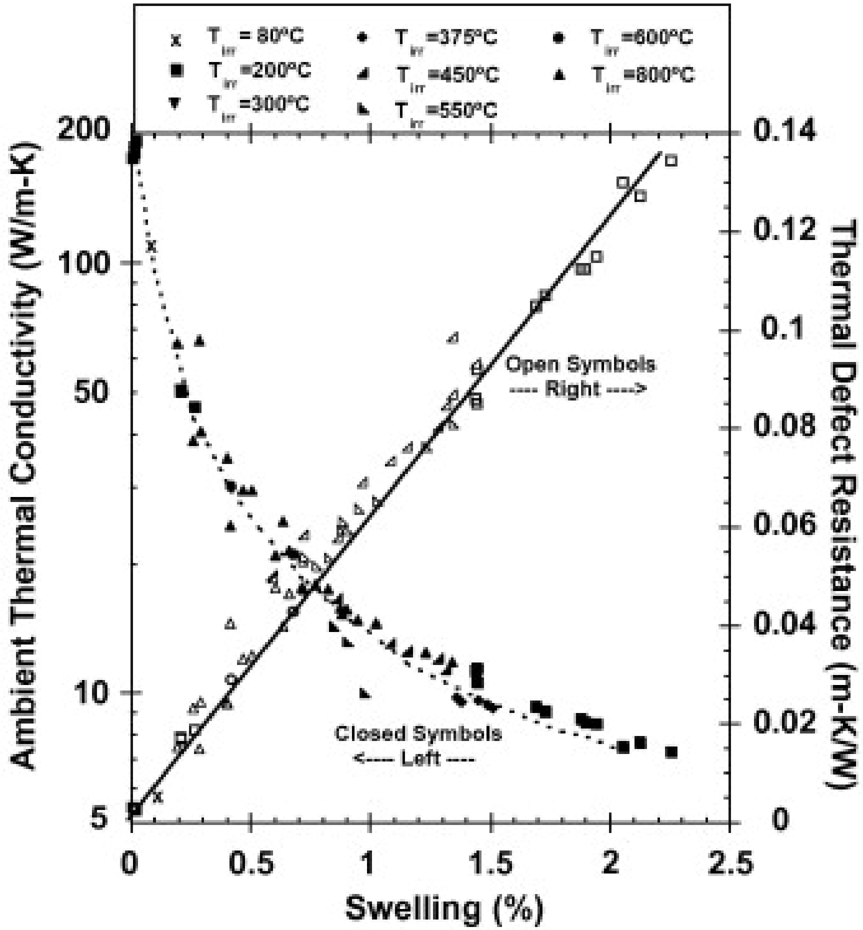
$$= \frac{6\pi^{1/2}}{k_B}\frac{1}{\omega_D}\left(\frac{\Omega}{aT_m}\right)^{1/2}(C_v T)^{1/2} - \frac{2\pi^2}{k_B}\frac{v^2}{aT_m\omega_D^3}T$$

Small Loops

$$\Delta\left(\frac{1}{K}\right)_{loop} = K_i\frac{C}{B} = \frac{24\pi h^2 R^2}{v}n_{loop}$$

Evolution of swelling and thermal conductivity in irradiated SiC

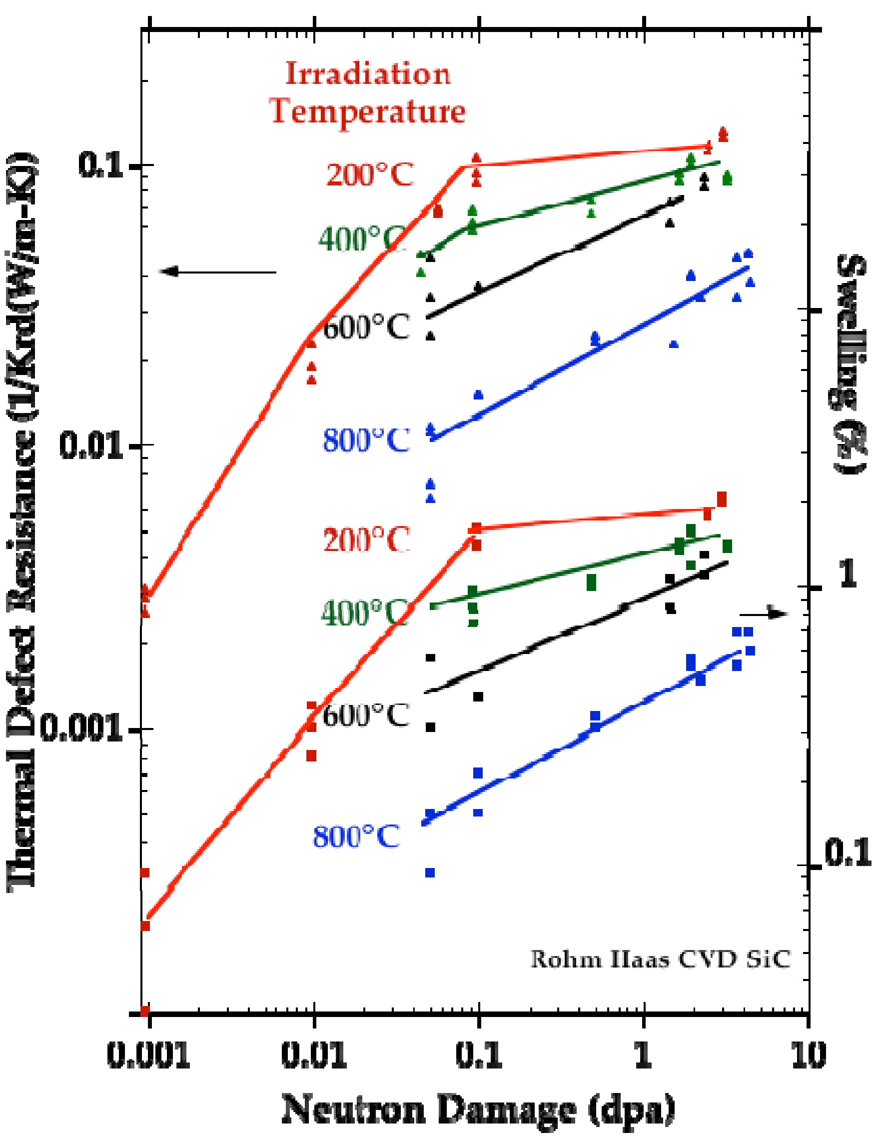


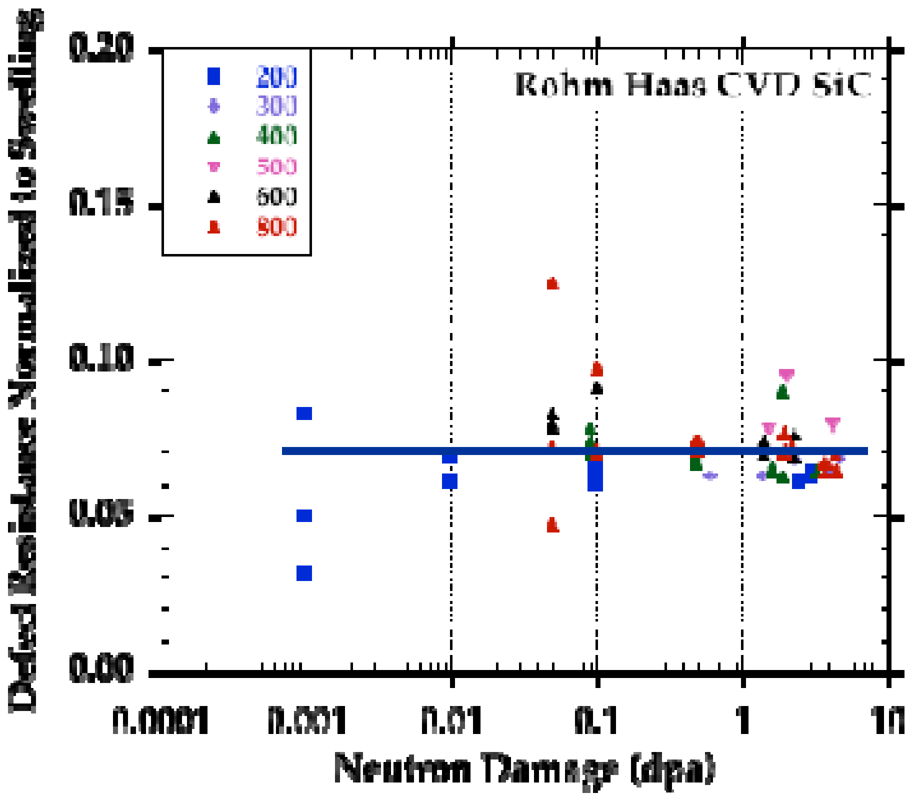


L.L. Snead et al., J. Nucl. Mater. 411 (2011) 330

L.L. Snead et al., J. Nucl. Mater. 371 (2007) 329

Irradiation-induced Thermal Defect Resistance in SiC

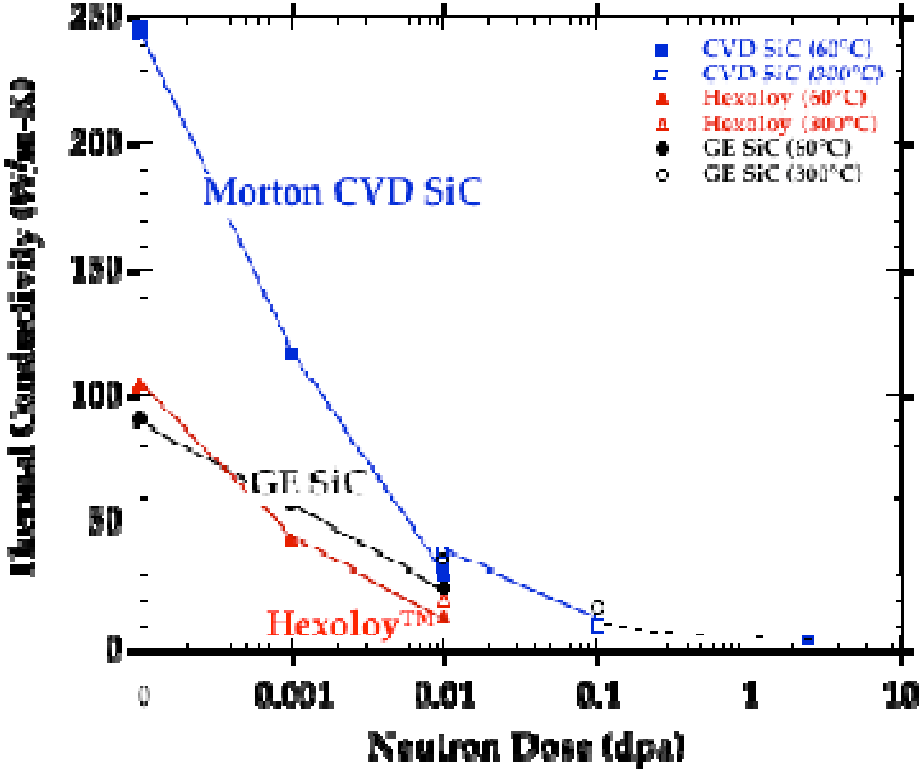


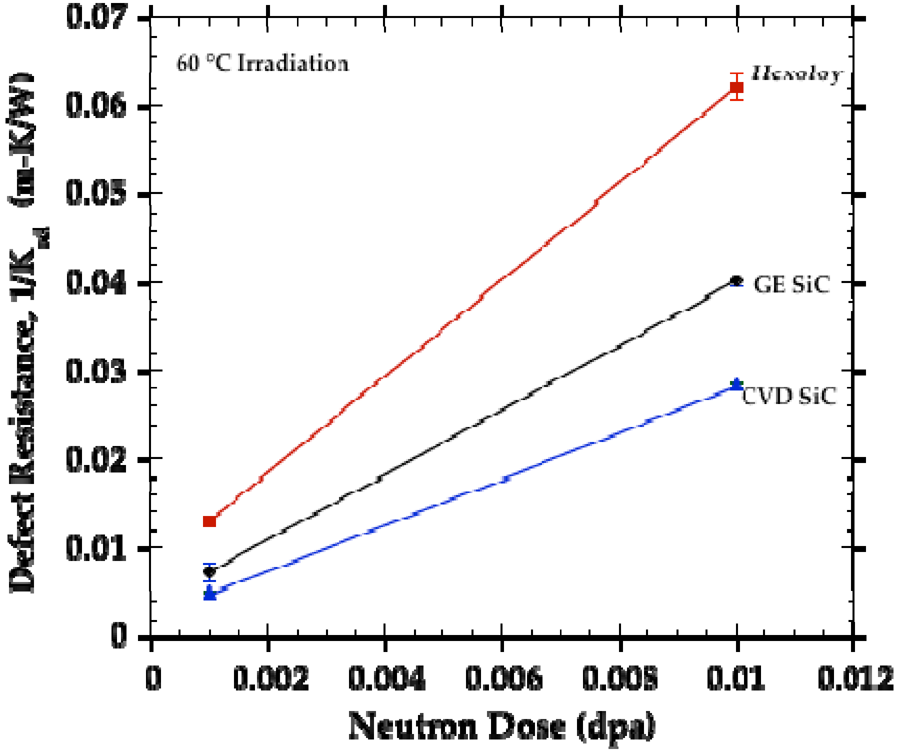


Thermal defect resistance normalized to irradiation-induced swelling is constant independent of irradiation temperature (200-800°C) suggesting single defect type controlling phonon scattering

L.L. Snead

Irradiation-Induced Thermal Defect Resistance of SiC

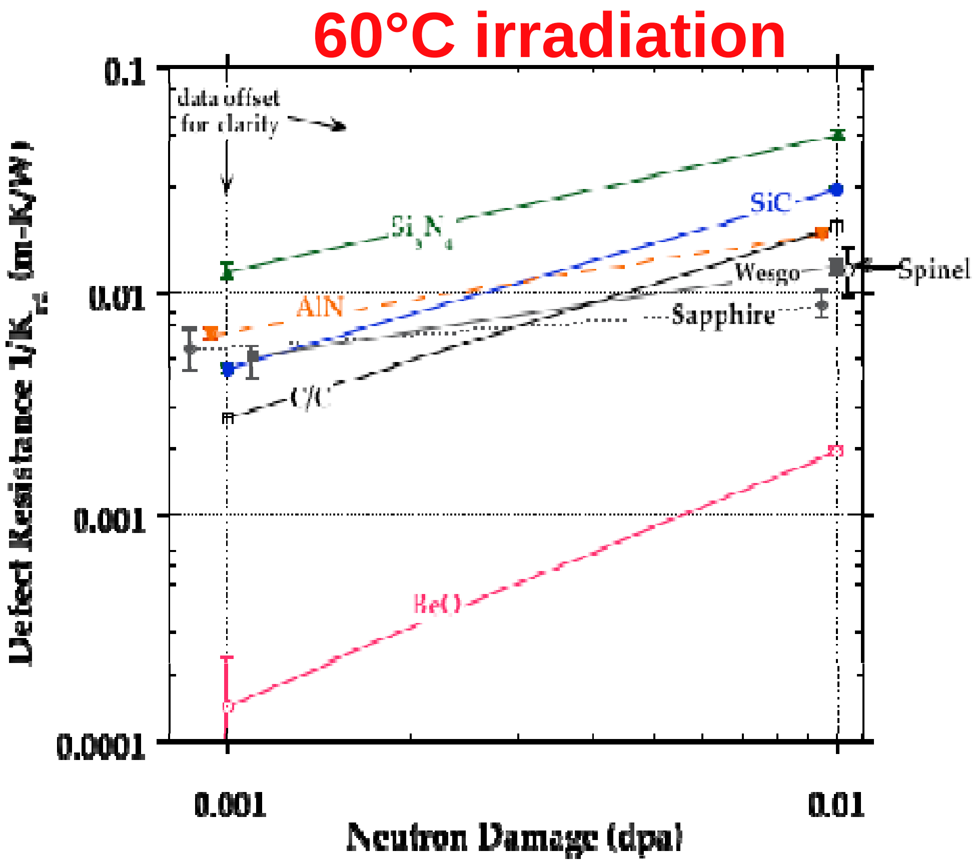


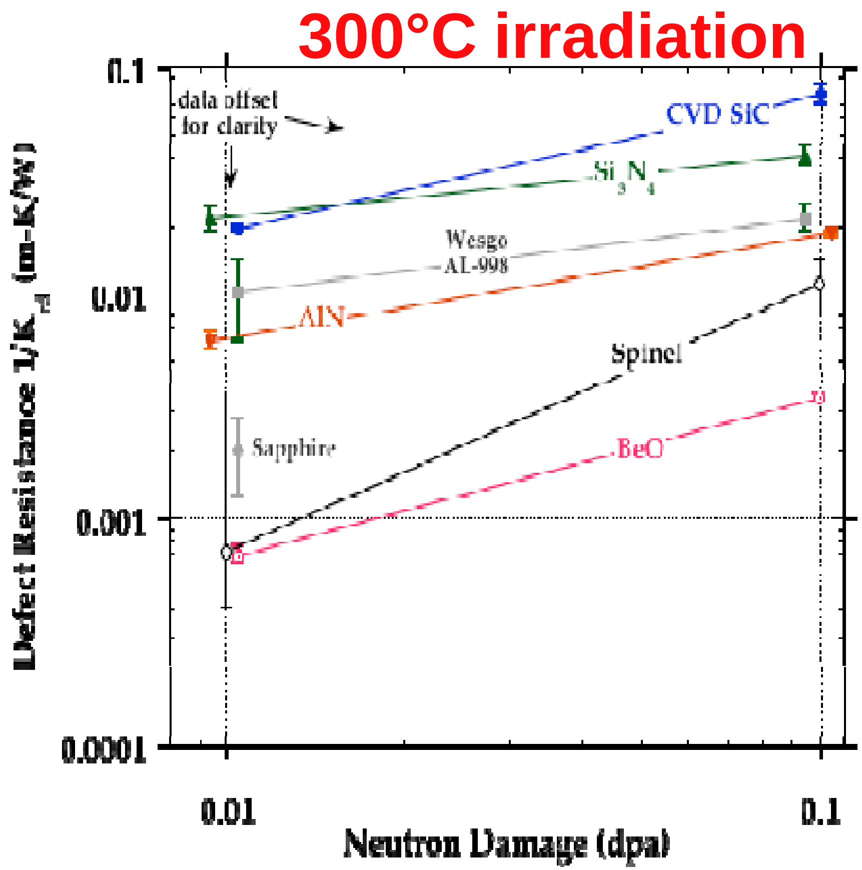


	CVD SiC	GE SiC
K_{unirr}-K_{irr}	2.35	0.7
% K_{unirr}	6%	22%
Defect Resistance	2.8	2.9

Hexoloy contains boron, which disrupts grain boundary under irradiation. GE SiC and CVD SiC are both high-purity SiC.

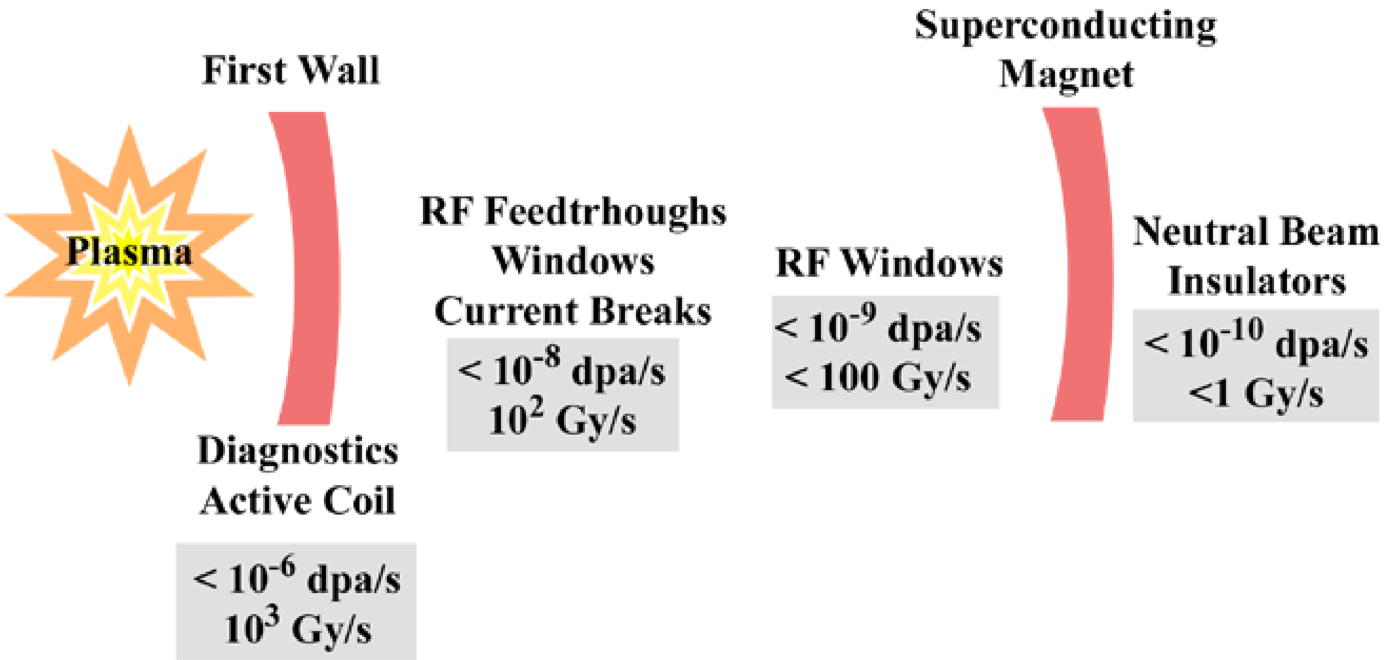
L.L. Snead, S.J. Zinkle & D.P. White, J. Nucl. Mater. 340 (2005) p. 187





L.L. Snead, S.J. Zinkle & D.P. White, J. Nucl. Mater. 340 (2005) p. 187

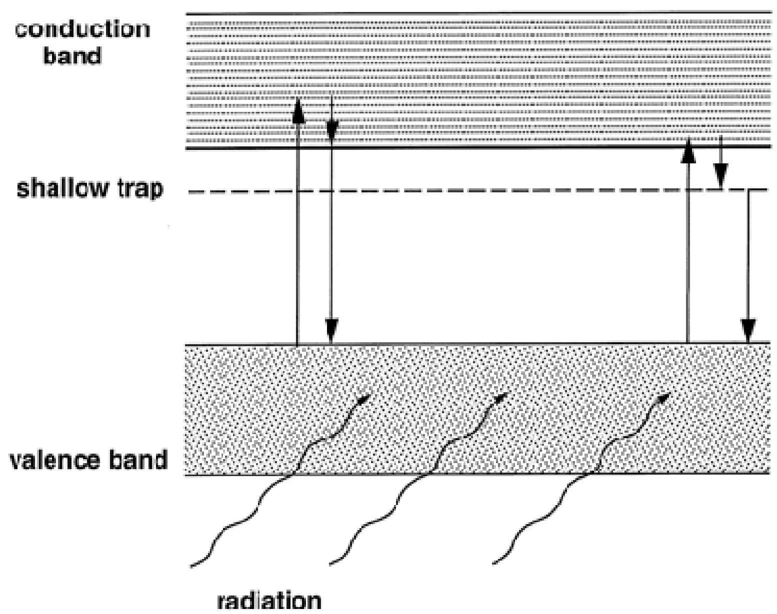
Radiation Environments Anticipated for Ceramic Components in the ITER

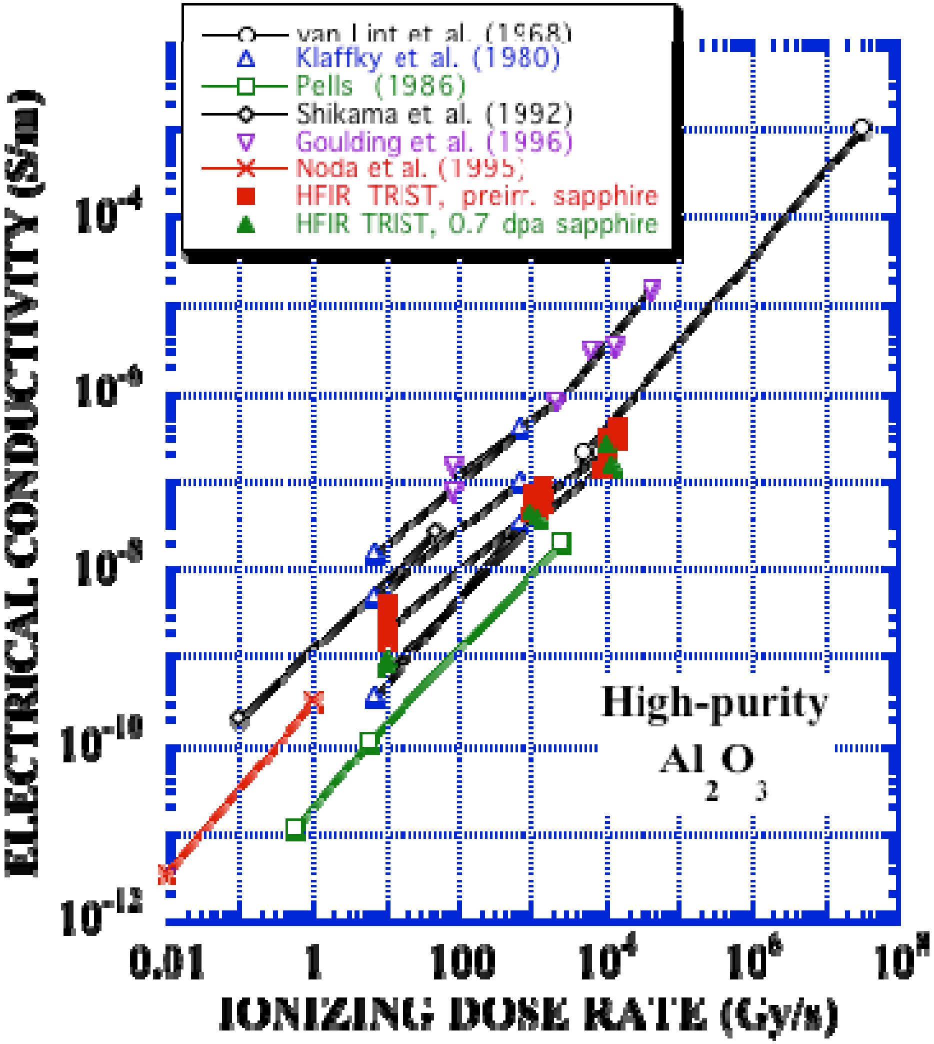


< 1×10^{18} n/m^2·s
< 1×10^{18} γ/m^2·s

Effect of ionizing irradiation on the electric conductivity of ceramic insulators

Radiation Induced Conductivity in Insulators



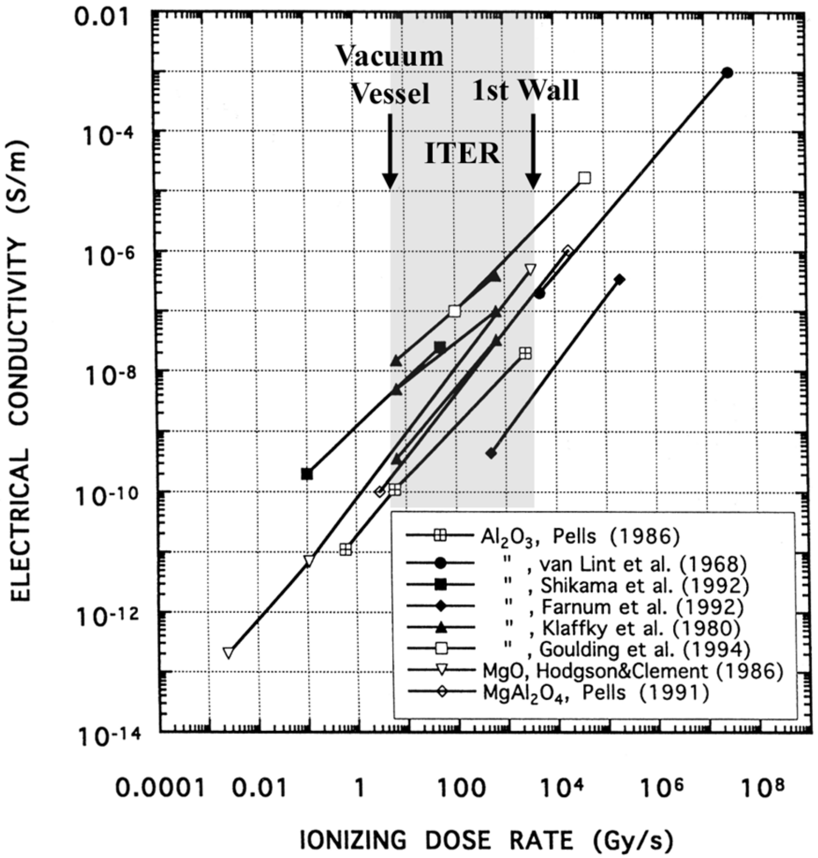


S.J.. Zinkle et al., DOE/ER-0313/22 (1997) p.188

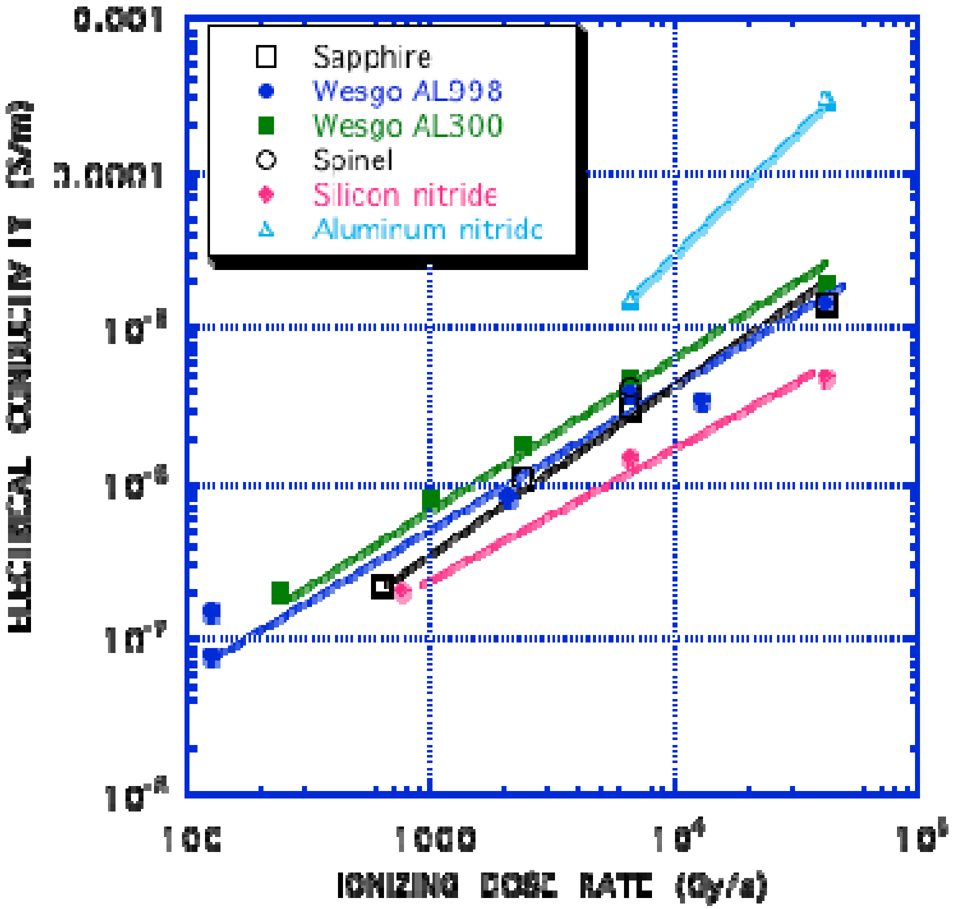
$$\sigma_{RIC}=\sigma_0+KR^{\delta}$$

$\delta\sim$ { 1 for insulators
<<1 for semiconductors and metals

Effect of ionizing irradiation on the electric conductivity of ceramic insulators



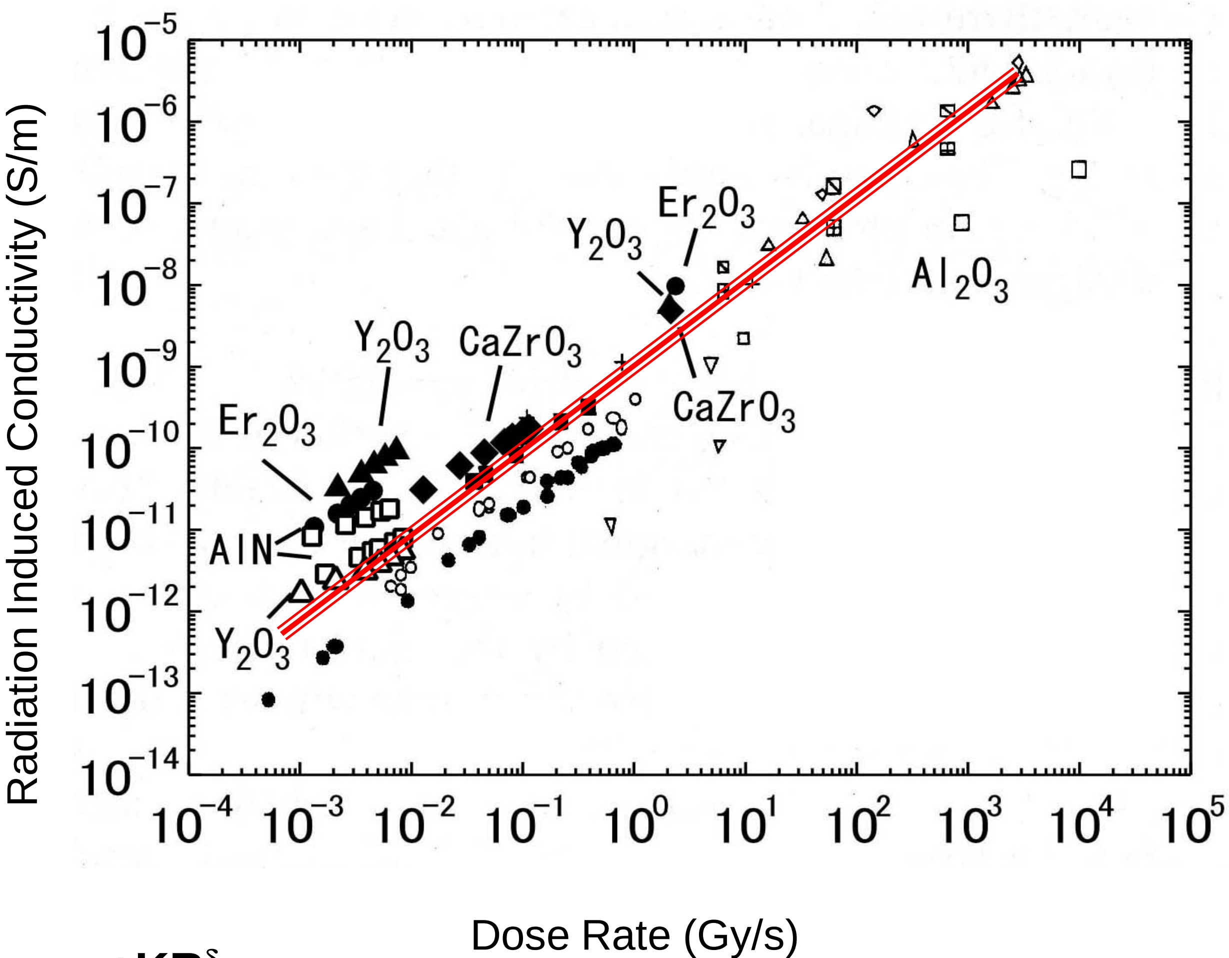
25^{o}C, pulsed neutron irradiation



L.W. Hobbs et al., J. Nucl. Mater. 216 (1994) 291

R.H. Goulding et al., J. Appl. Phys. 79 (1996) 2920

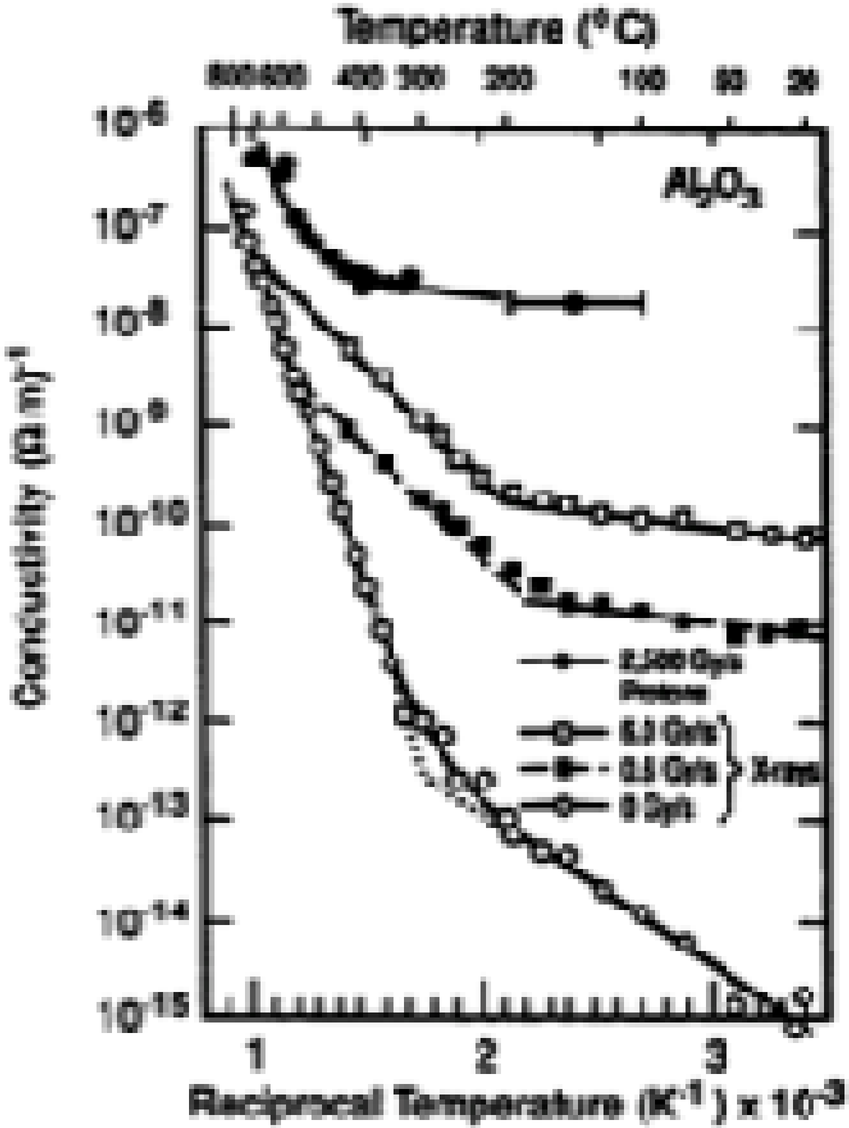
Effect of ionizing irradiation on the electric conductivity of ceramic insulators



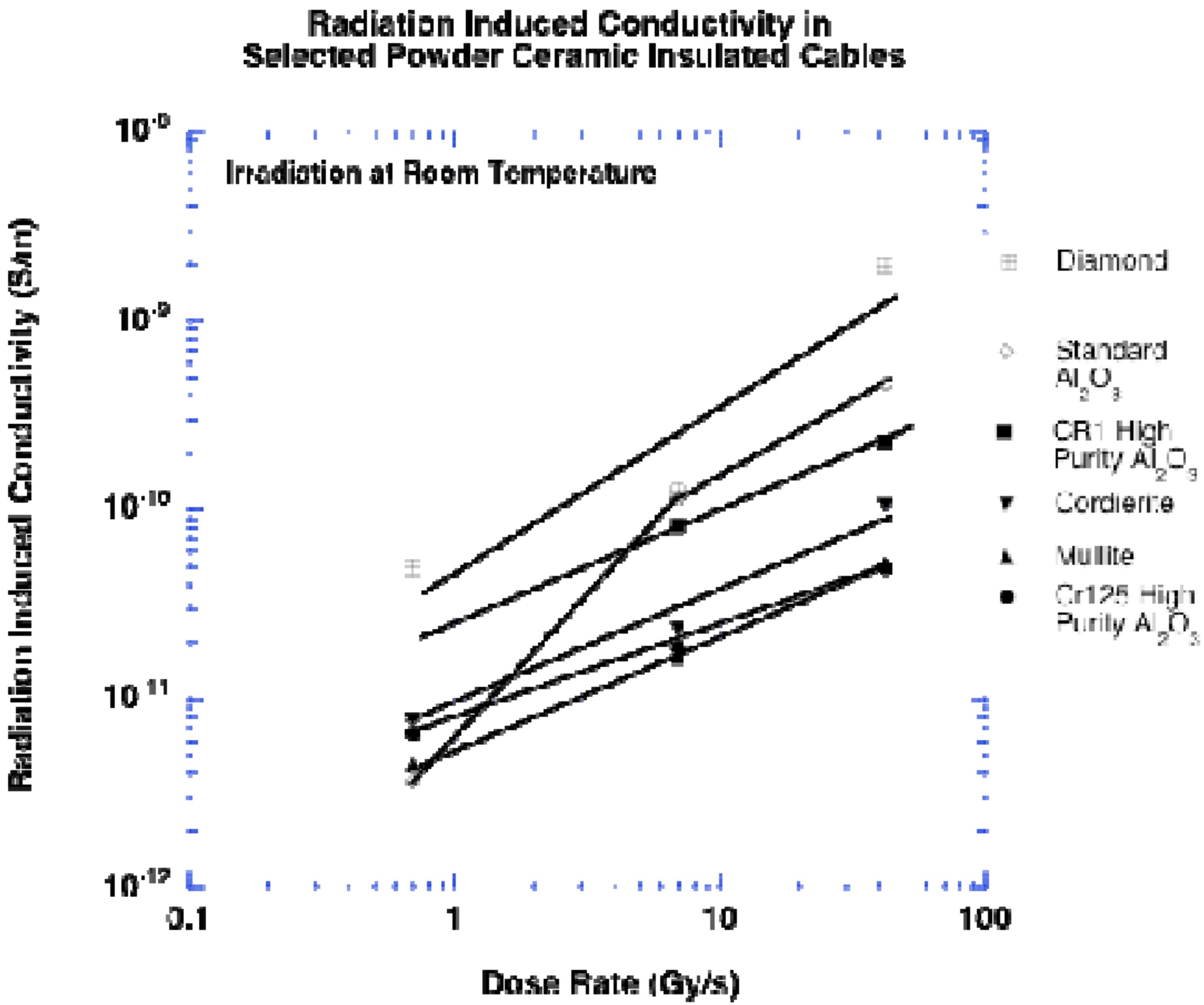
$\sigma_{RIC}=\sigma_0+KR^{\delta}$

$\delta\sim$ { 1 for insulators; <<1 for semiconductors and metals

Temperature-dependent electrical conductivity of Al_2O_3 with and without ionizing radiation



G.P. Pells, Rad. Eff. 97 (1985) 199


Radiation Induced Conductivity in
Selected Powder Ceramic Insulated Cables
Irradiation at Room Temperature
Radiation Induced Conductivity (S/m)
Dose Rate (Gy/s)
0.1
1
10
100
Diamond
Standard Al_2O_3
CR1 High Purity Al_2O_3
Cordierite
Mullite
Cr125 High Purity Al_2O_3

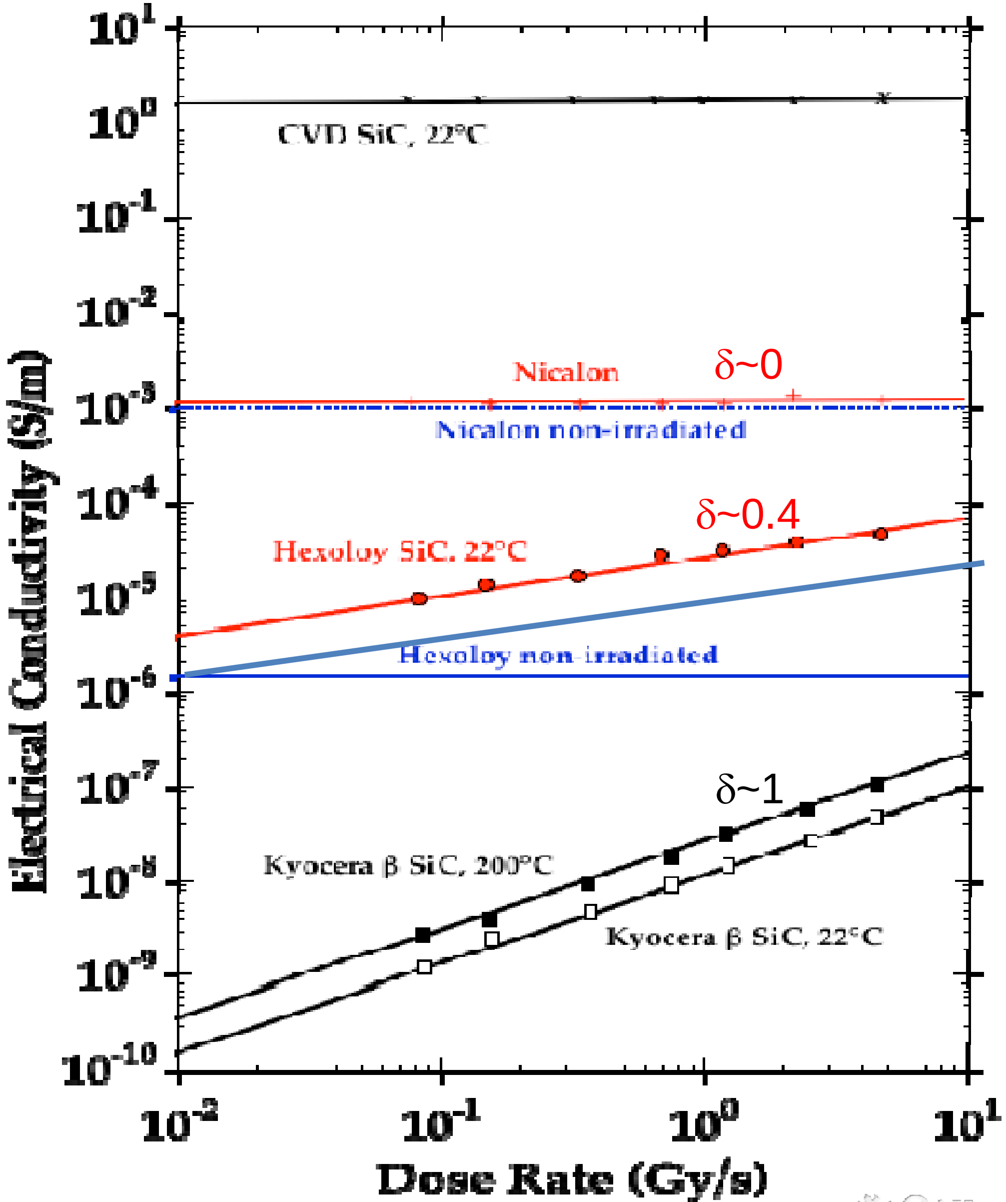
Radiation Induced Conductivity (In-situ) for different grades of SiC

- **Ionizing radiation excites electrons into the conduction band enhancing conductivity.**

$\sigma = \sigma_o + KR^{\delta}$

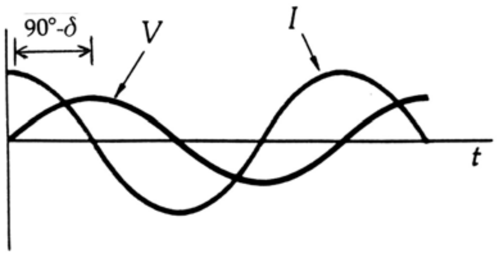
Dose rate

- **If base conductivity is high (>10^{-4} S/m), RIC is insignificant**

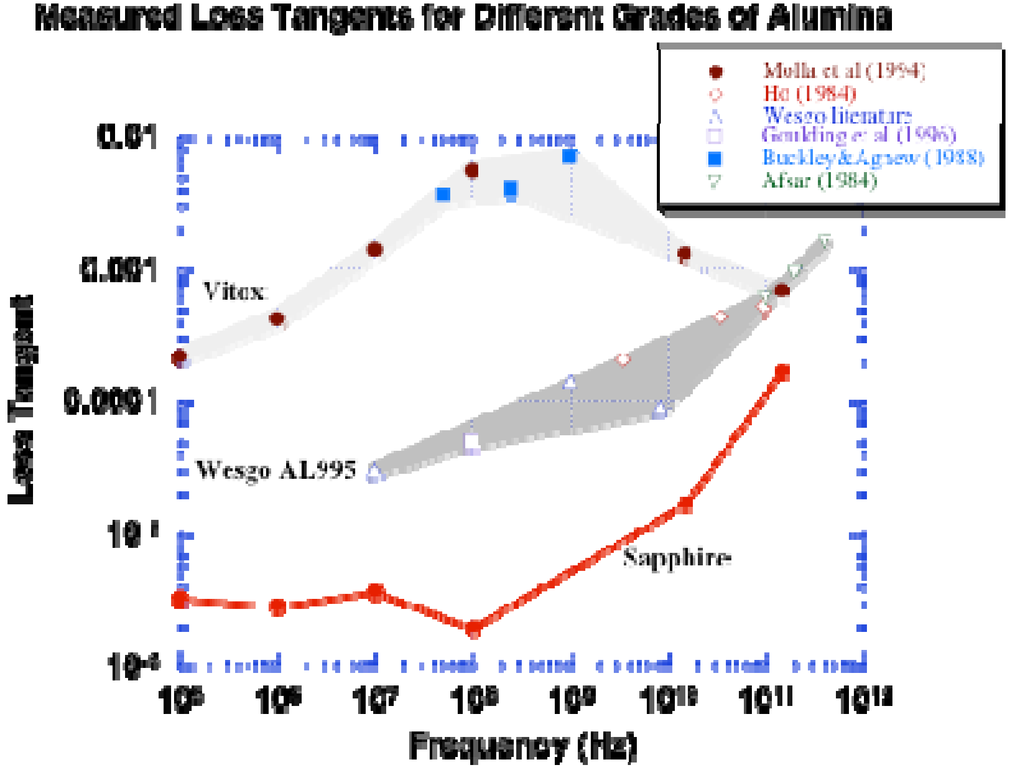


L.L. Snead, J. Nucl. Mater.329-333 (2004) 524

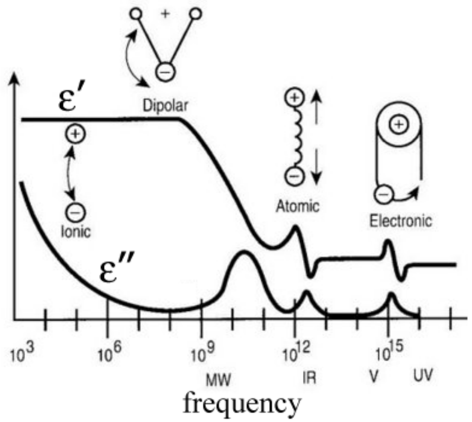
Dielectric loss tangent of ceramic insulators



Plots of current I and voltage V across the capacitor versus time. The voltage lags the current by 90°.



tanδ=ε''/ε' (ratio of imaginary to real parts of permittivity)



need low electrical conductivity (including during irradiation

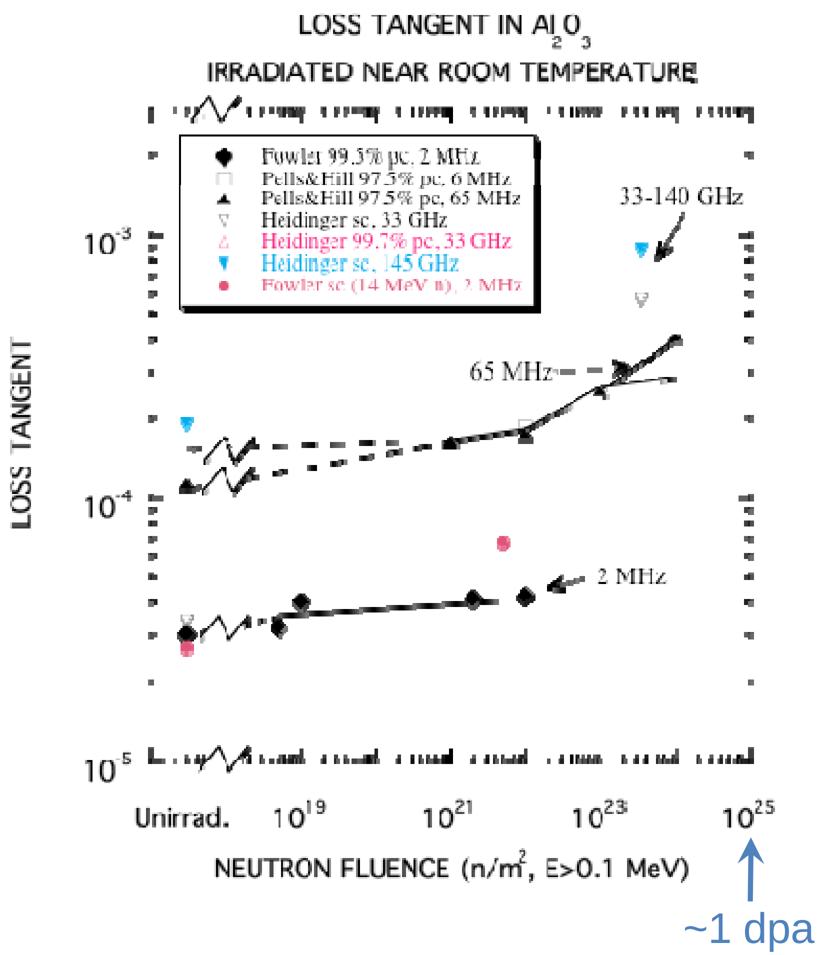
$$\tan\delta = \frac{\sigma_{DC}}{\omega\varepsilon'} + \frac{\chi''}{\varepsilon'/\varepsilon_0}$$

Polarization losses can be increased by irradiation-induced defects

Absorbed power: P=ωε' tanδ E^2

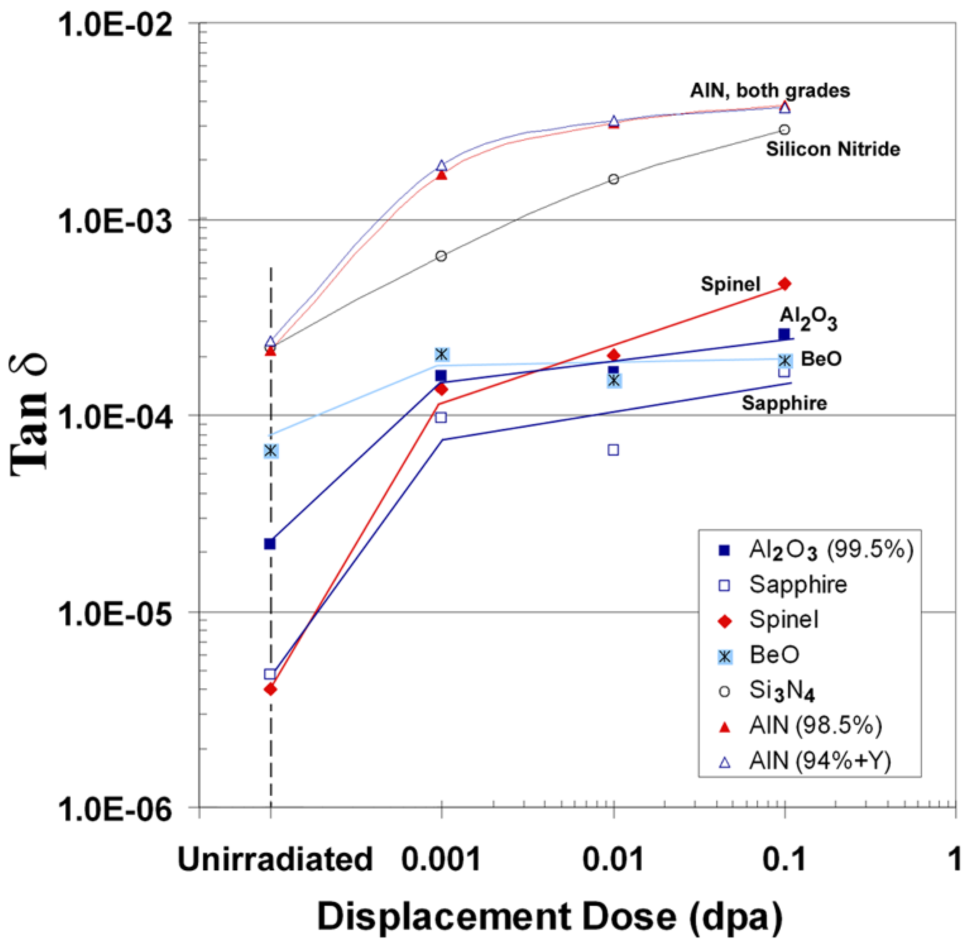
To avoid overheating, need tanδ<10^{-3} (ICH, 100 MHz) or <10^{-6} ECH, 100 GHz)

Effect of irradiation on the dielectric properties of ceramic insulators



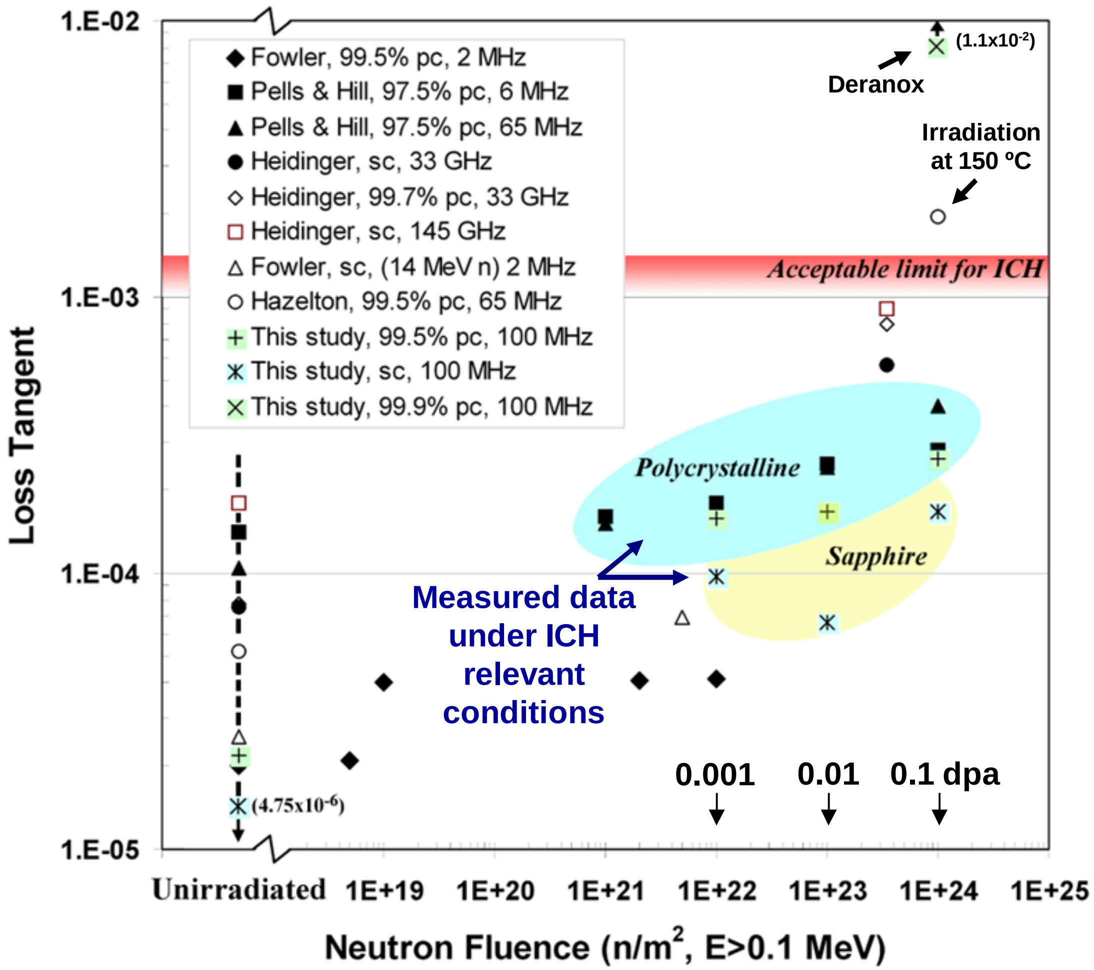
L.W. Hobbs et al., J. Nucl. Mater. 216 (1994) 291

100 MHz loss tangent after 70°C neutron irradiation



after R.H. Goulding & K.J. Leonard, ITER report G 51 TD 26 FU (2007)
and C. Hazeltine et al., J. Nucl. Mater. 253 (1998) 190

Loss Tangent Database of Irradiated Al_2O_3



- Neutron irradiation near room temperature for polycrystalline (pc) and single crystal (sc-sapphire)
- Acceptable limit for ion cyclotron heating (ICH), Tan $\delta \cong 0.001$
- Data from this study for Wesgo Al-995 and single crystal sapphire plotted
- Certain grades of polycrystalline Al_2O_3 suitable for use up to 0.1 dpa
- Deranox grade Al_2O_3 exceeds Tan δ limit for ICH application

Conclusions

- Neutron and gamma radiation can produce significant property changes (degradation) in ceramics
- Strength degradation typically occurs for anisotropic (e.g., HCP) crystal structures
  - Graphite is more resilient than BeO, Al_2O_3
- Most pronounced changes typically occur in physical properties (e.g., thermal conductivity)
- Electrical conductivity of insulators exhibits marked prompt increases during exposure to ionizing radiation
  - Effect is small for semiconductors