The Effect of Neutron Irradiation on Candidate First Wall and Diverter Materials

Materials Beyond ITER ?

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ARIES Town Hall Meeting

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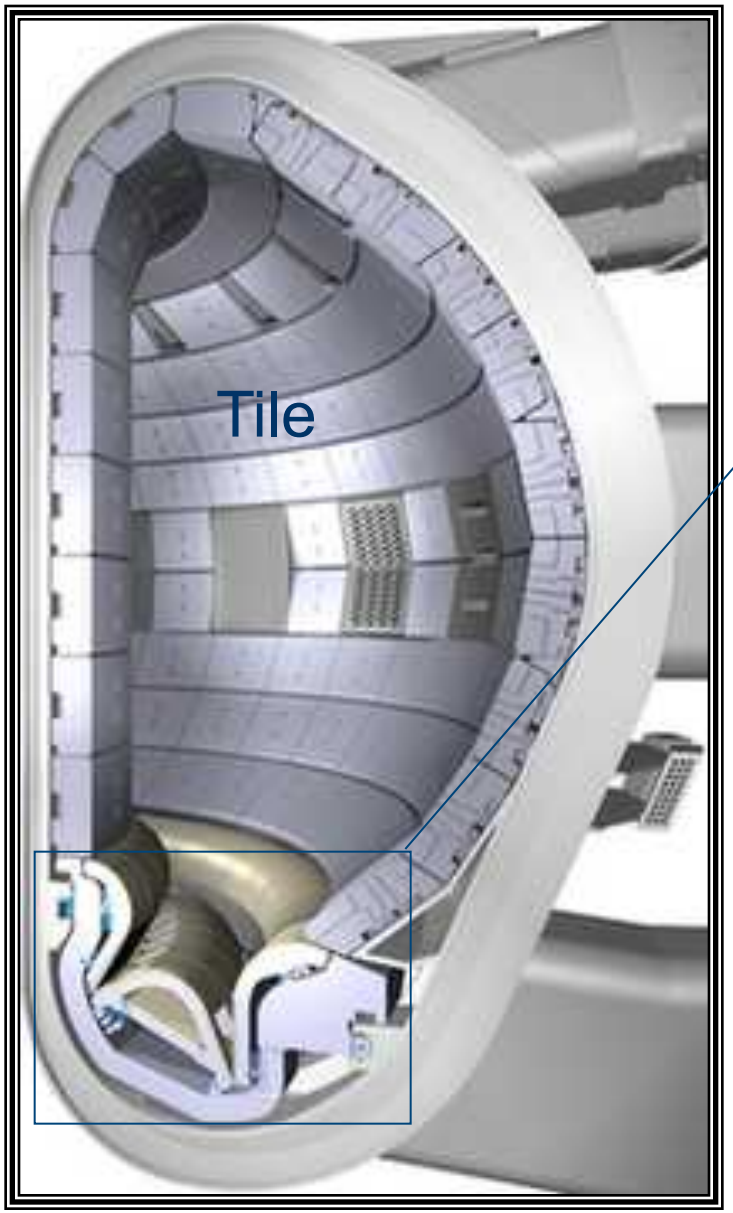
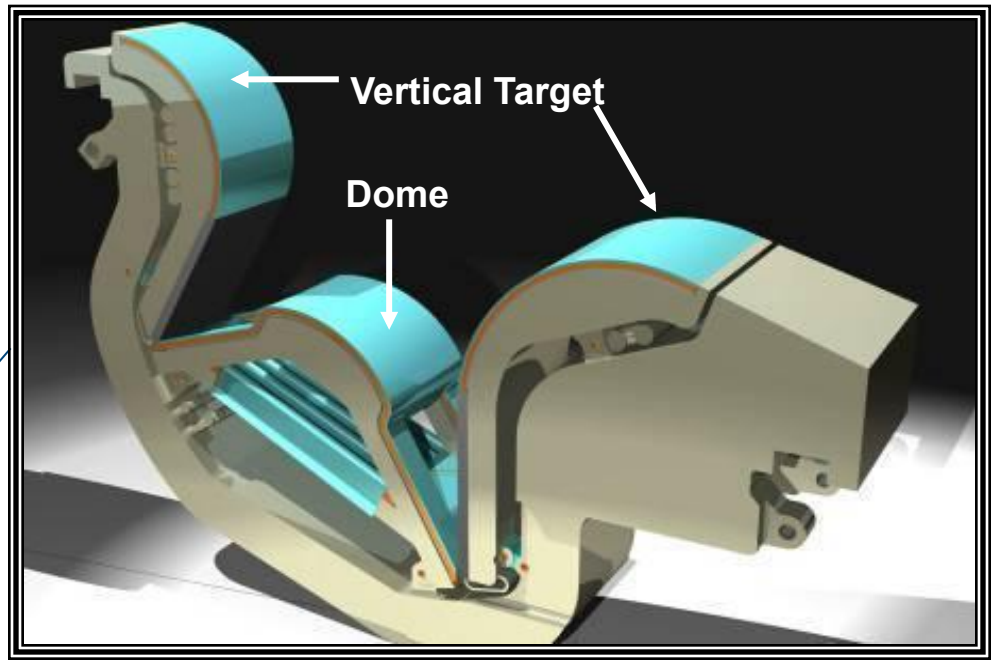
Thanks to : Yuri Osetskii (ORNL)
Maria Caturla (Universidad de Alicante)
Meimei Li (ANL)

OAK RIDGE National Laboratory

ITER Cutaway





Vertical Target

PFC : either W or Carbon Fiber Composite
Heat Sink : CuCrZr alloy
Structure : 316 LN(I)-G steel alloy

Dome

PFC : W
Heat Sink : CuCrZr alloy.
Structure : 316 LN(I)-G steel alloy

Tile

Pure Beryllium

Neutron Cascade Damage and Defect Evolution

Before we proceed……..a definition……

DPA = displacement per atom

Typical reactor application = 1-10 DPA / year

1 DPA Ceramic ~ 1 x 10^{25} n/m^2 E>0.1 MeV

Lattice Displacement Energies

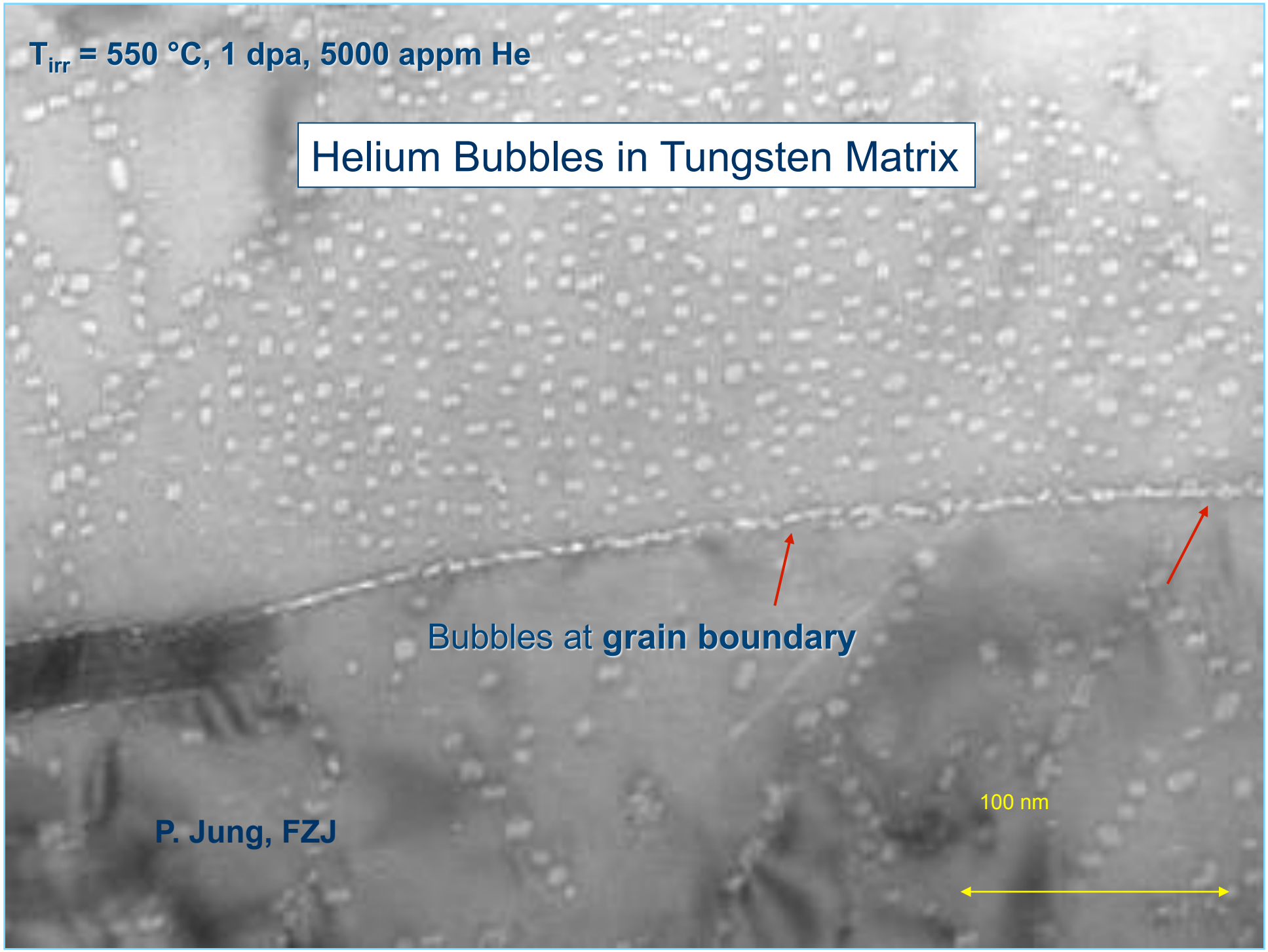
Graphite : 25 eV

Be 25 eV

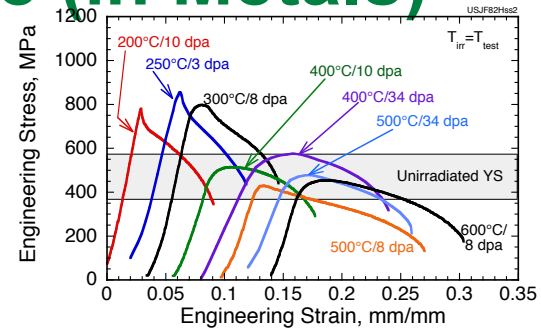
Iron : 40 eV

Tungsten : 80 eV

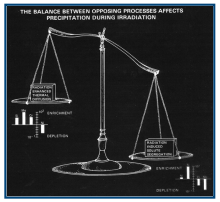
A 100 keV imparted energy
Will produce thousands of
displaced atoms !

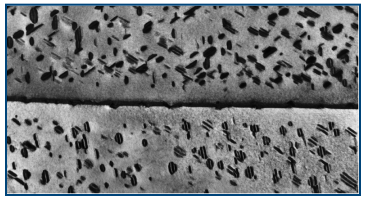

T_{irr} = 550 °C, 1 dpa, 5000 appm He
Helium Bubbles in Tungsten Matrix
Bubbles at **grain boundary**
100 nm
P. Jung, FZJ

Five Evils of Radiation Damage (in Metals)



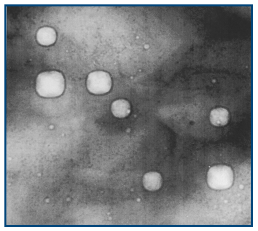
- **Radiation hardening & embrittlement ($<0.4\ T_M$, >0.1 dpa)**

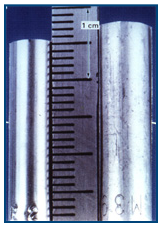




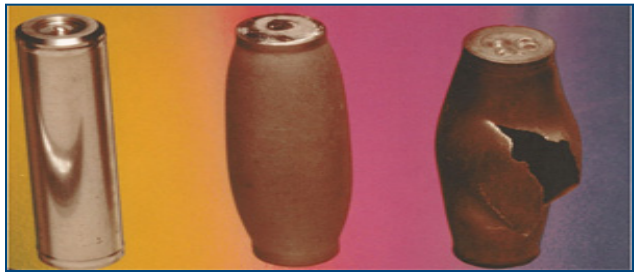
- **Phase instabilities from radiation-induced precipitation (0.3-$0.6\ T_M$, >10 dpa)**

- **High temperature He embrittlement ($>0.5\ T_M$, >10 dpa)**





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

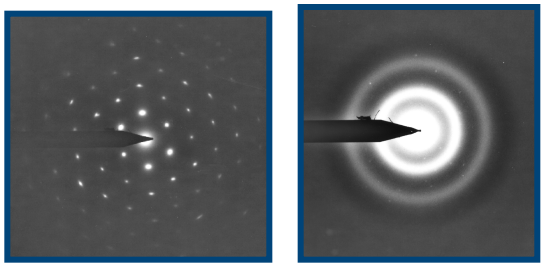


- **Irradiation creep ($<0.45\ T_M$, >10 dpa)**

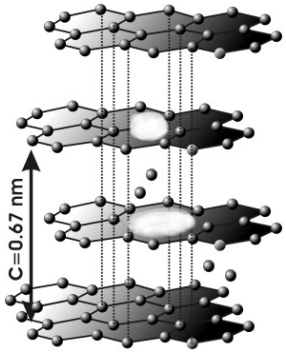
Evils of Radiation Damage (in Ceramics…)

Graphite Sublimes at > 3000 K

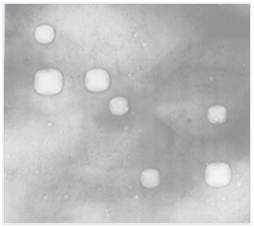
- **Point Defect Swelling and Amorphization (<0.3 T_M, < 5 dpa)**



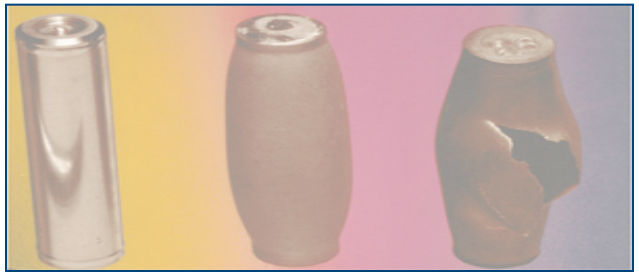
- **Radiation Induced Thermal Conductivity Degradation (<0.4 T_M, < 5 dpa)**



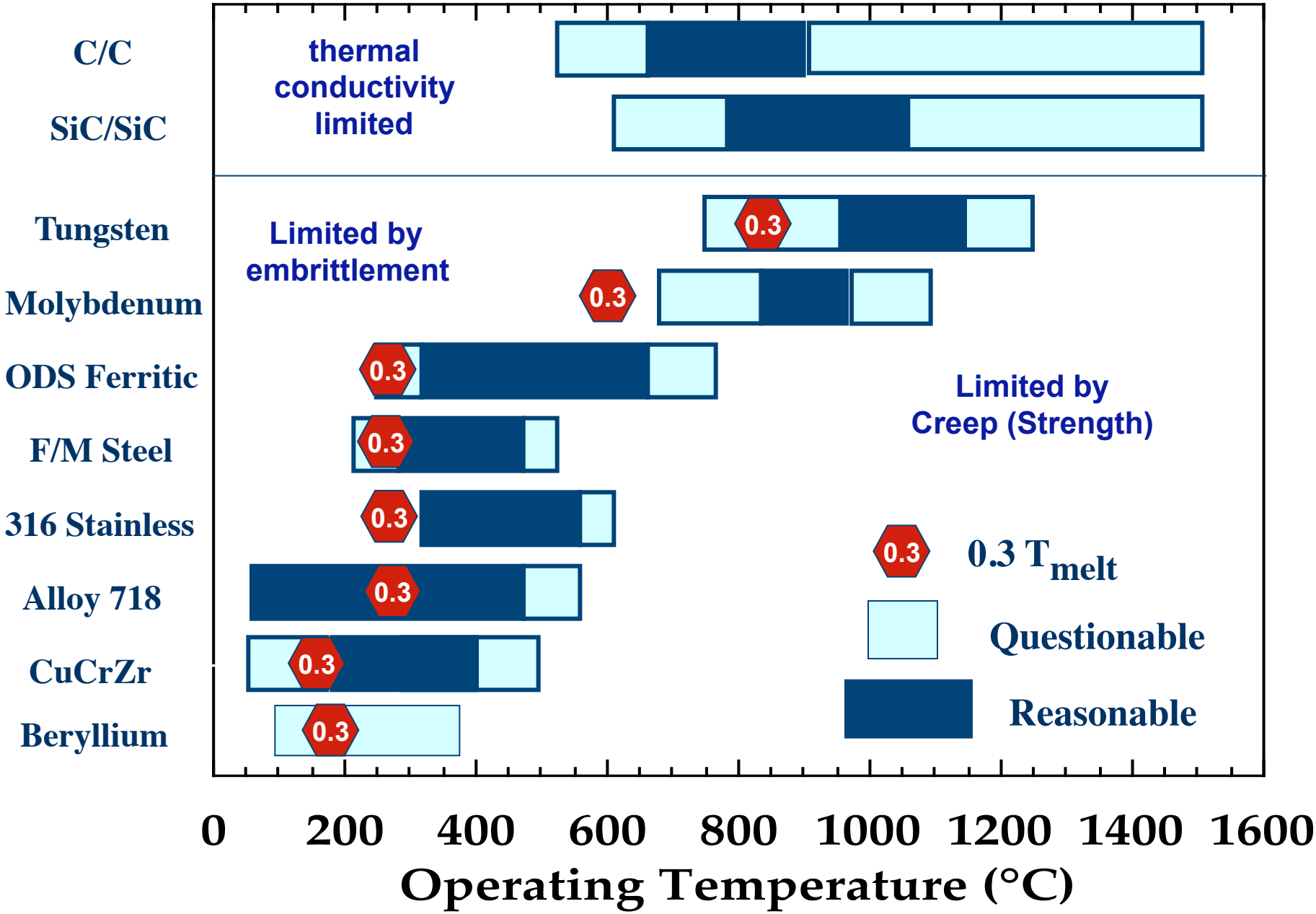
- **Volumetric swelling from void formation (0.4-0.6 T_M, >4 dpa)**



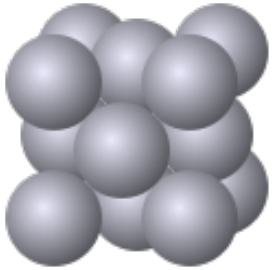
- **Irradiation creep (<0.45 T_M, >10 dpa)**



Operating Range, Irradiated Structural Materials



Copper for Nuclear Application



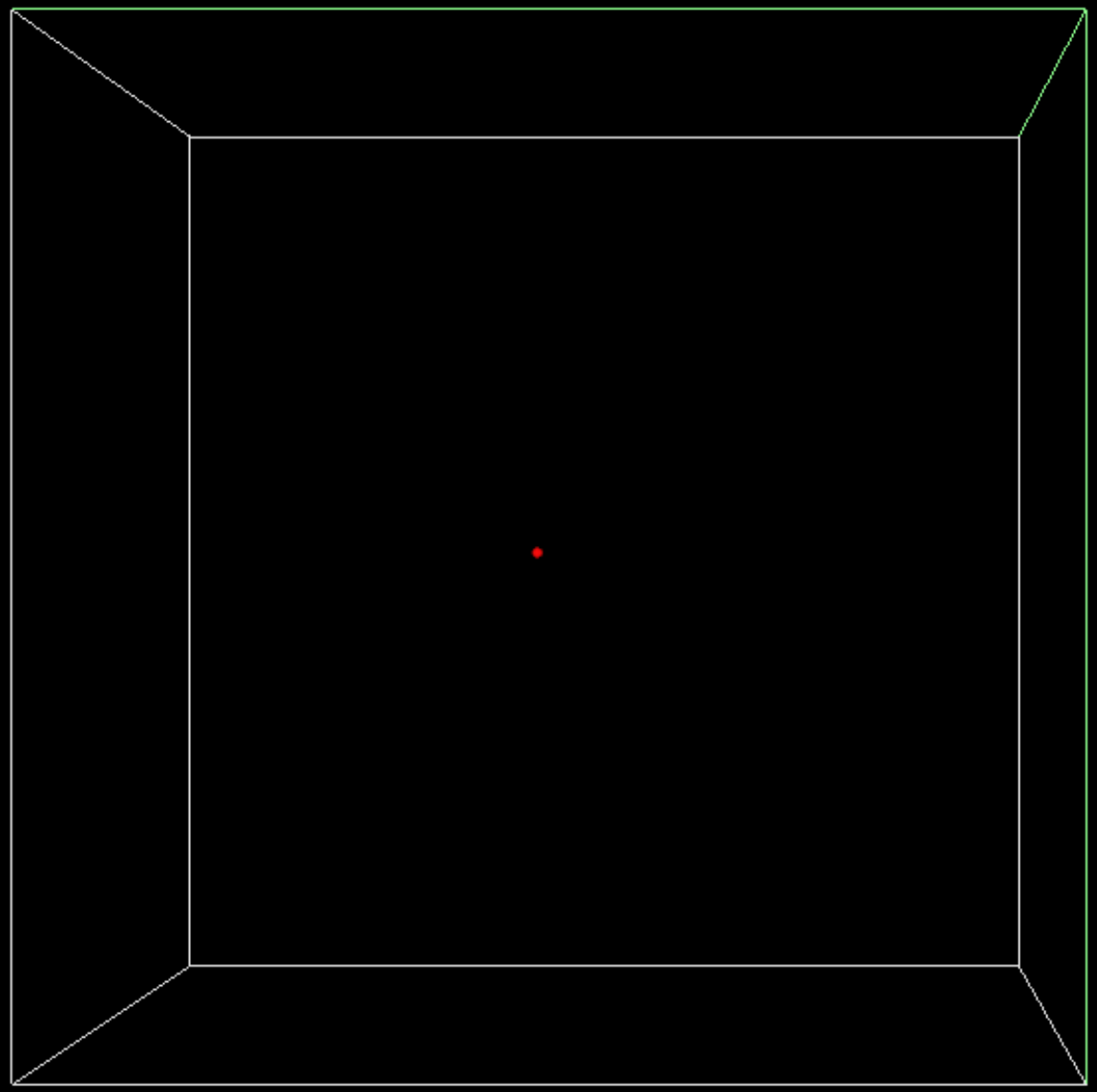
face center cubic

1357 K Melting Point

- Copper has been used extensively in tokamak application for heat sink application.

- Annealed, pure copper, is extremely weak at both room and elevated temperature. Solid solution, precipitation hardened, or dispersion strengthened copper alloys are used to increase strength.

- Neutron irradiation-produced defects cause significant embrittlement in copper.



Copper 25keV:

Cube faces (100)
Cube size = 25nm
2.048.000 atoms
Temperature=100K

time = 0.006ps

NFP = 1

Legend :

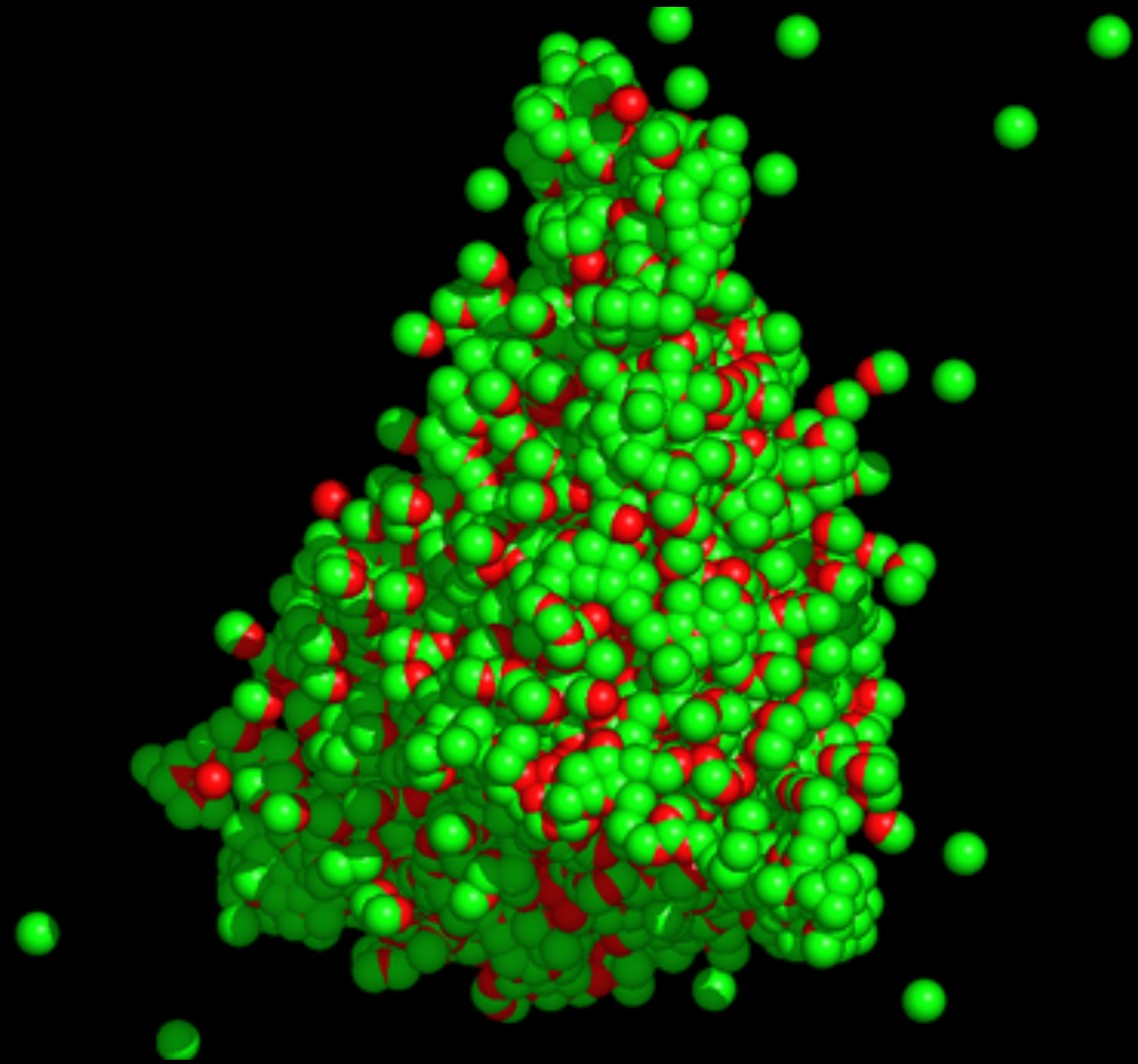
Displaced atoms

Vacant sites

Defect Production and Annihilation in Irradiated Materials

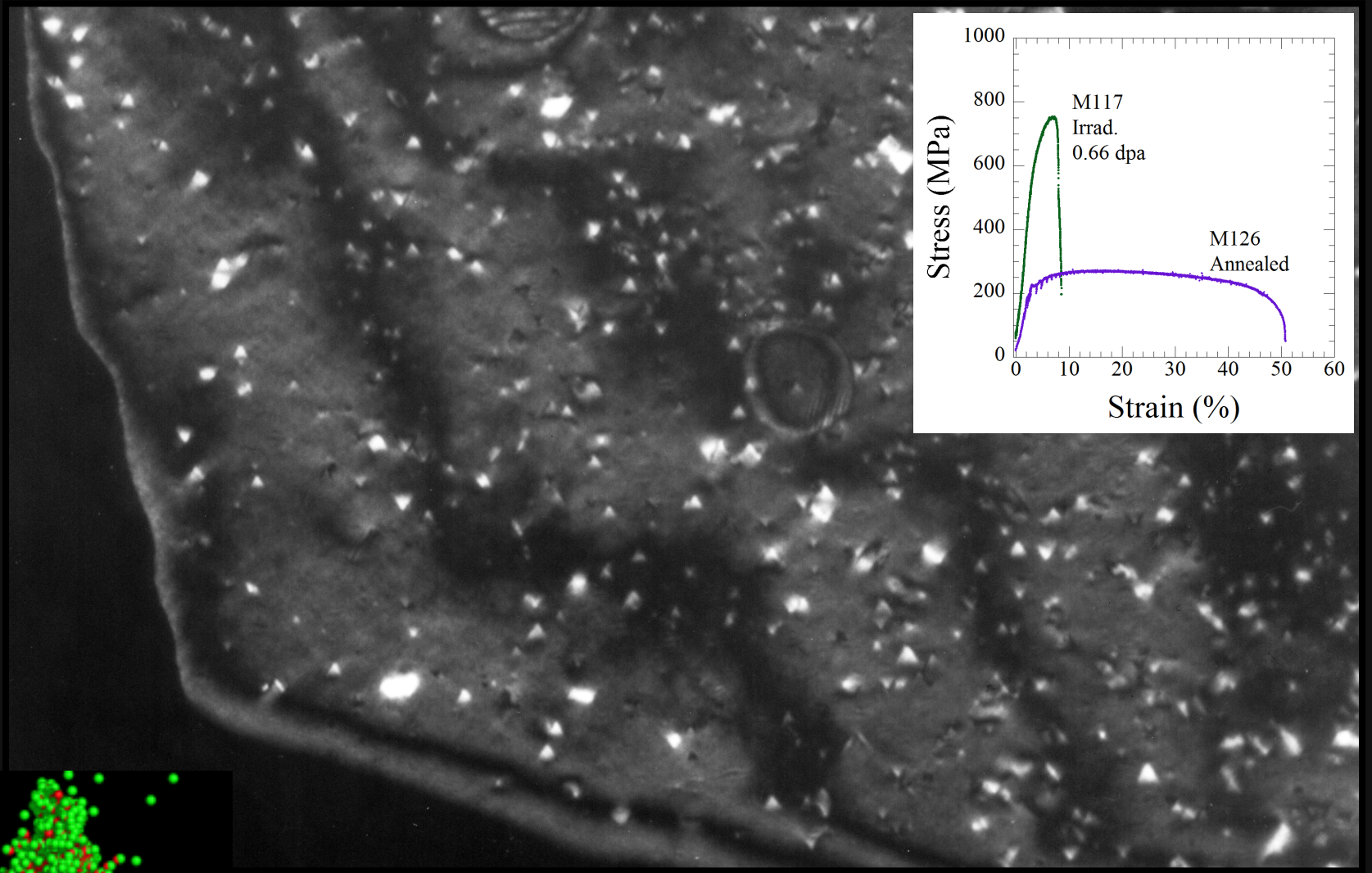
-strong function of material, time, and temperature… ○ vacancy ○ interstitial

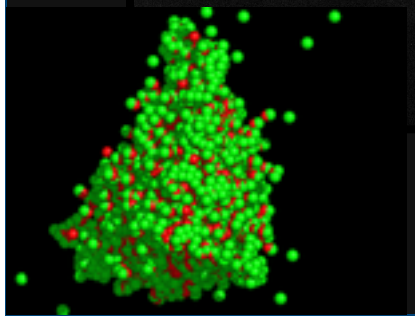
Condensation of Cascade in FCC Copper



A large non-perfect SFT and several SIA clusters/loops are formed

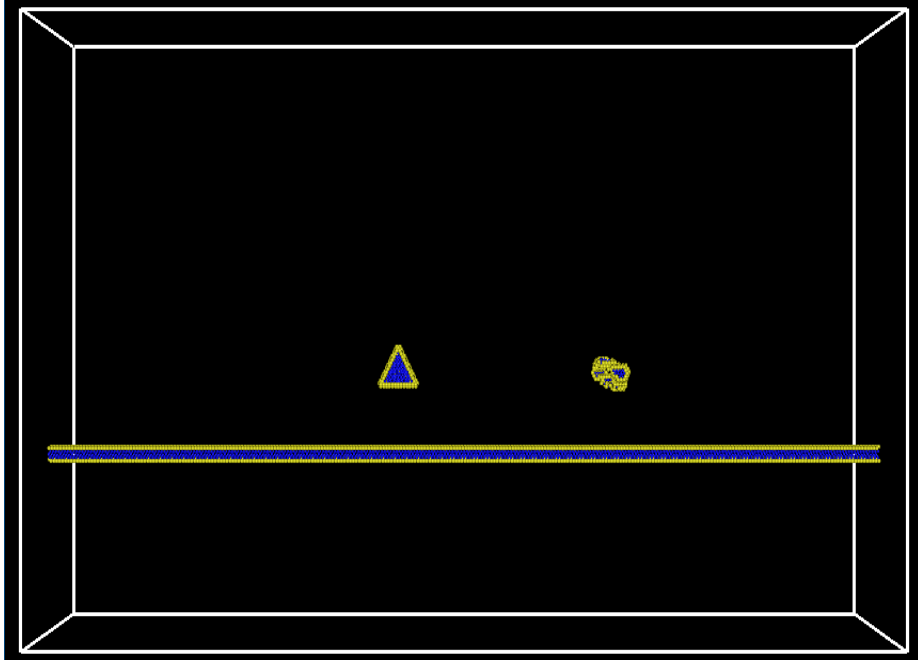
Defect cluster microstructure in Cu irradiated to 1 dpa at low temperature (~90˚C)

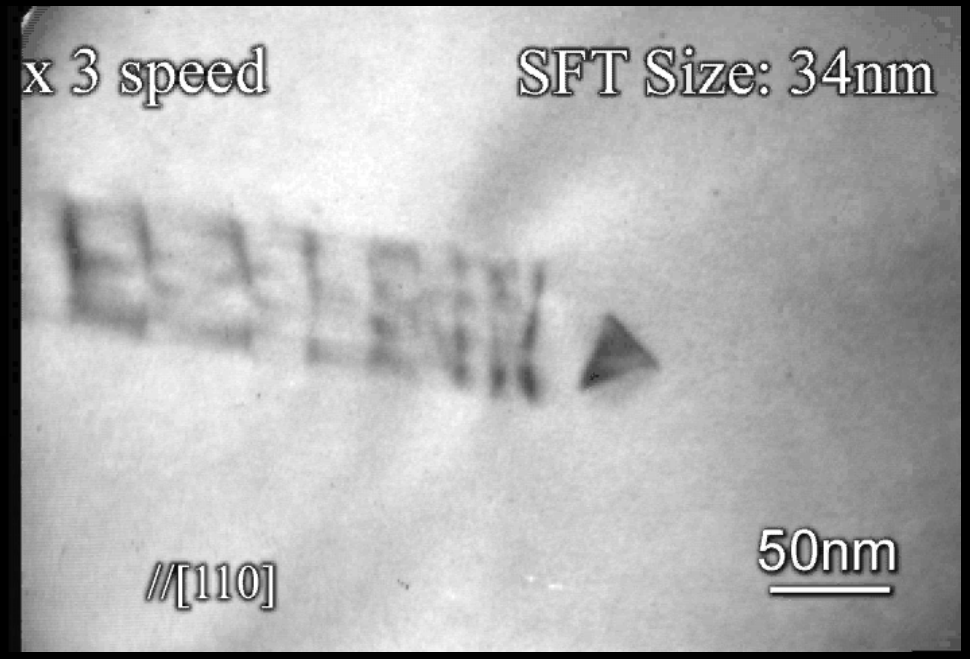




Zinkle et al., J. Nucl. Mater. (1994)

Dislocation dynamics and in-situ TEM of dislocation SFT interaction

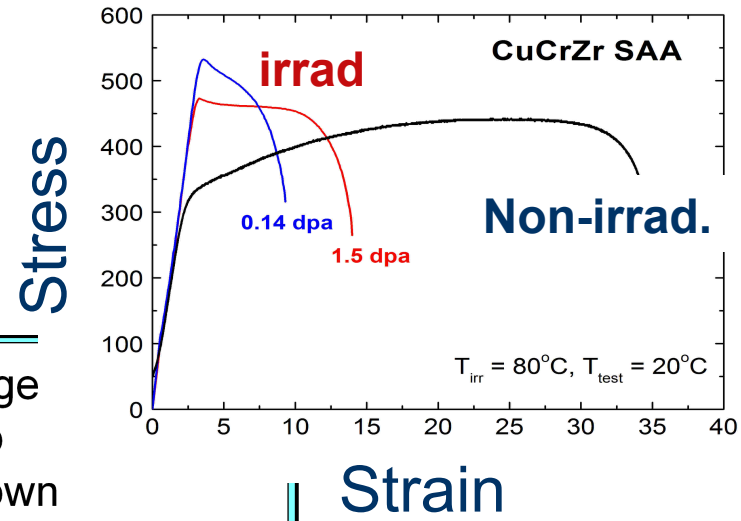


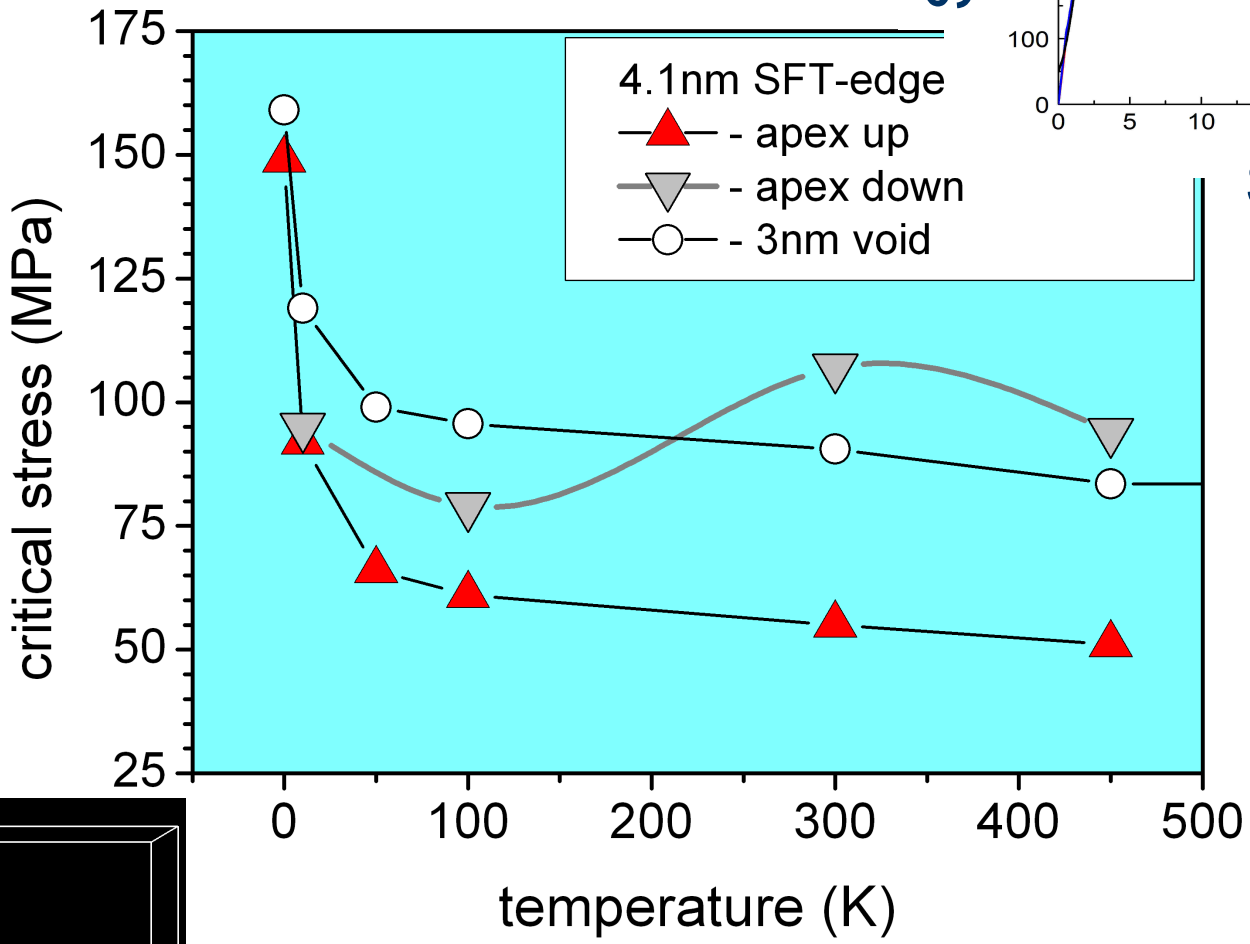


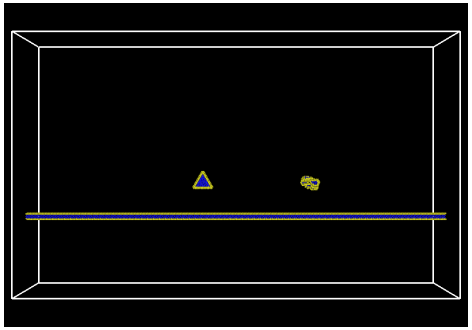
Yuri Osetskii, ORNL

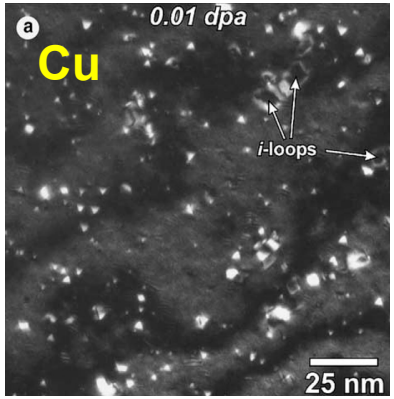
Yoshi Matsukawa, ORNL (now U. mich)

Irradiation Hardening/Embrittlement
- copper -







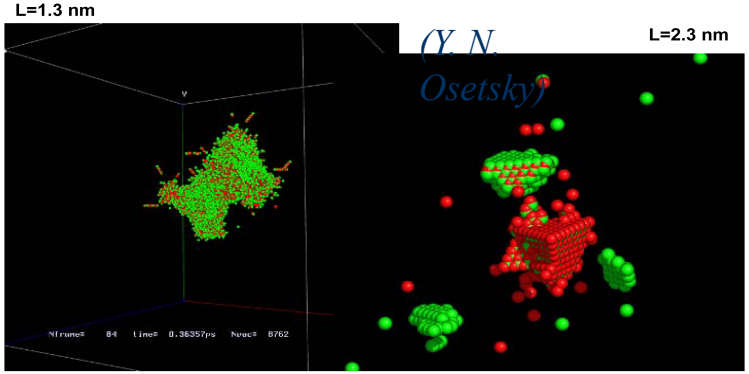


Irradiation Hardening and Embrittlement

- **Irradiation produced defects serve as "road blocks" to the dislocation motion required for deformation (plasticity.)**

- **Defects can be formed either directly within the cascade or can develop (mature) upon diffusion of interstitial/vacancy species following cascade:**

SFT in Cu
"in cascade"



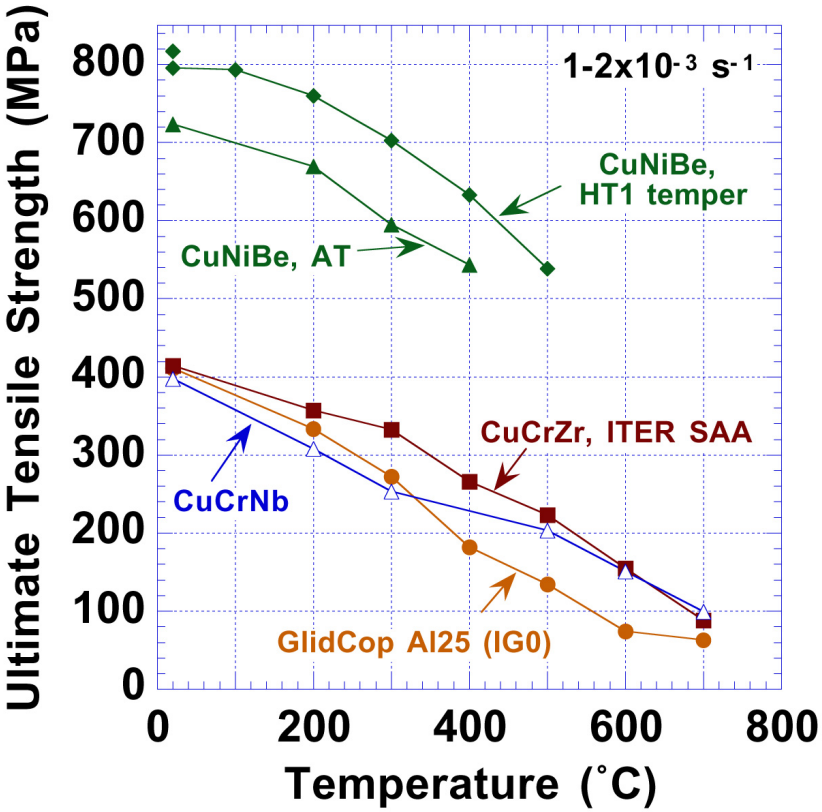
- **Immobile defects formed directly within the cascade are extremely troublesome, as we have no "metallurgically fix".**

  - typically, higher Z metals have higher fraction of "in cascade defects"

- **However, migrating defects (or transmutants such as He) can be dealt with by various means (alloying, dispersion strengthening, etc.)**

CuCrZr for ITER Application

- Copper alloys have significantly higher strength than pure copper, and still retain good conductivity

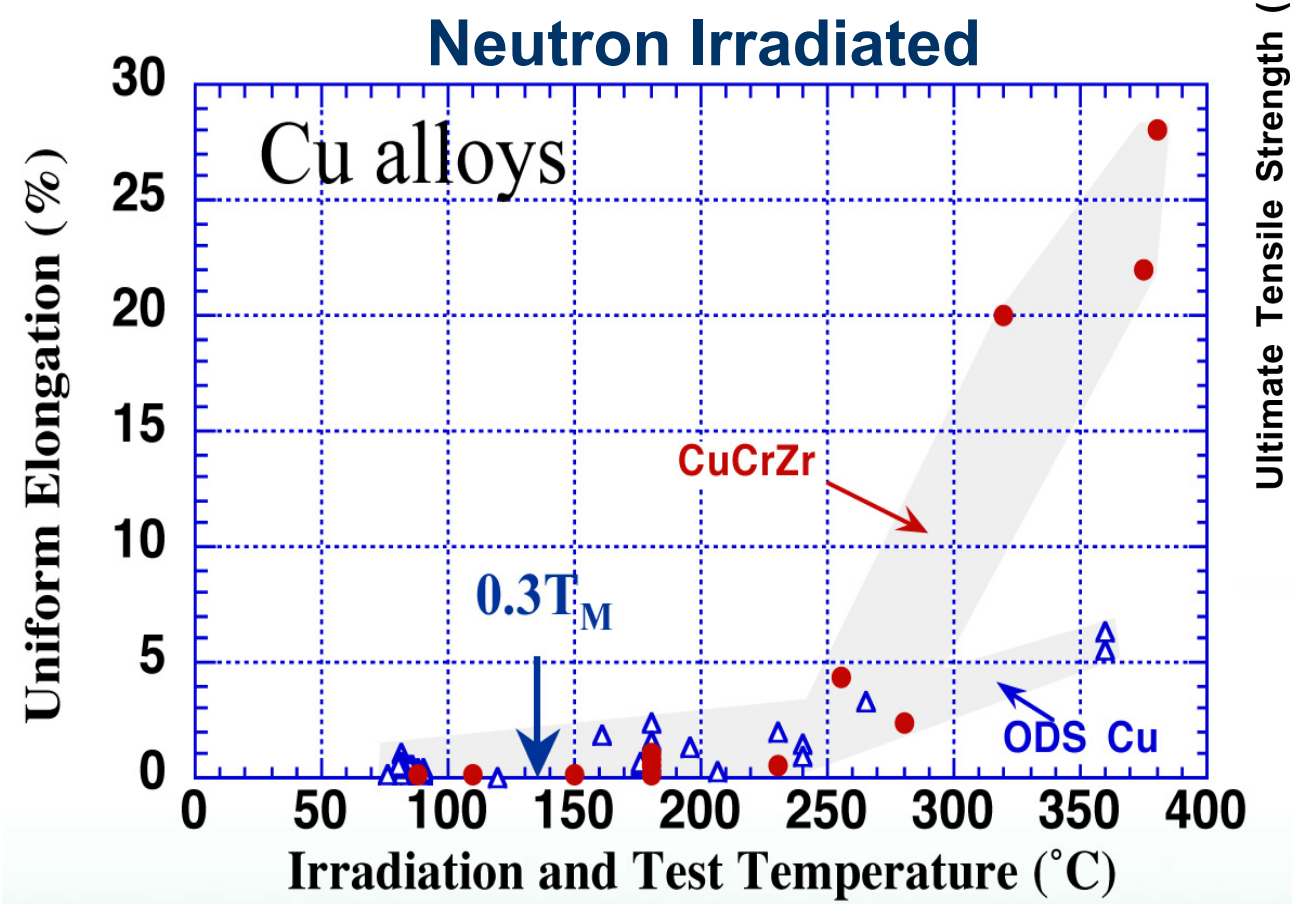


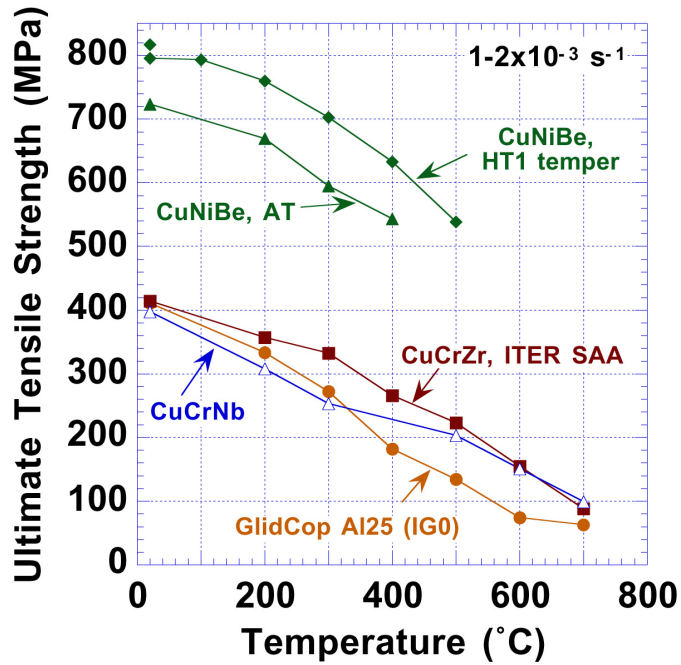
- Three copper alloys were considered:
  - Precipitation-hardened CuCrZr
  - Precipitation-hardened CuNiBe
  - Oxide-dispersion-strengthened (GlidCop Al-25)

- CuCrZr was selected due to its high fracture toughness, availability and low cost.

- ITER grade CuCrZr:
  - Cu – (0.60-0.90%)Cr – (0.07-0.15%) Zr

"Elevated" Temperature Irradiation Results for CuCrZr





- for irradiation in the 0.3-0.6 Tm range, copper embrittlement becomes less of an issue, however, alloy has little strength at these temperatures.

Biased Summary Comments on Copper Alloys

- Copper and copper alloys has been very well studied, both the fundamentals of the alloys and their irradiation effects. Improvements in both non-irradiated and as-irradiated performance will be incremental.

- Copper alloys are inherently limited in elevated temperature strength. For this reason they may <u>not</u> be attractive alloys beyond ITER.

Tungsten for Nuclear Application

body center cubic structure

3695K melting point

- Currently, tungsten is utilized as a plasma facing "tile" material with only moderate structural requirements. For ITER the requirement for ductility and irradiation resistance is limited.

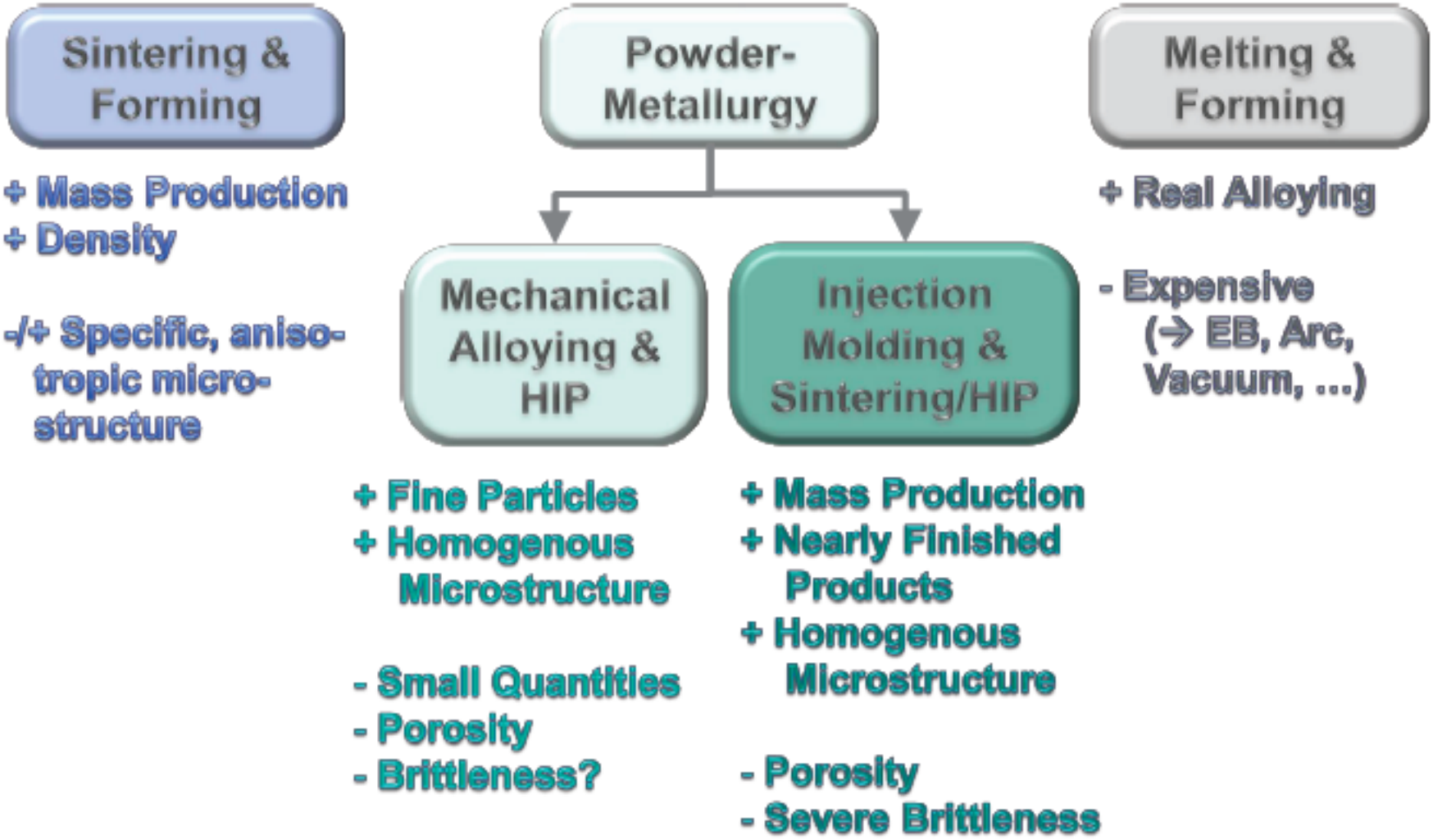
- As discussed already in this school, the primary advantage of tungsten lies in its good resistance to sputtering (high mass) and high melting temperature (3695 K). However, both of these benefits argue in direct opposition to good irradiation resistance:

  - high mass means higher fraction of defects formed directly within cascade (limiting metallurgical tools to mitigate irradiation effects)

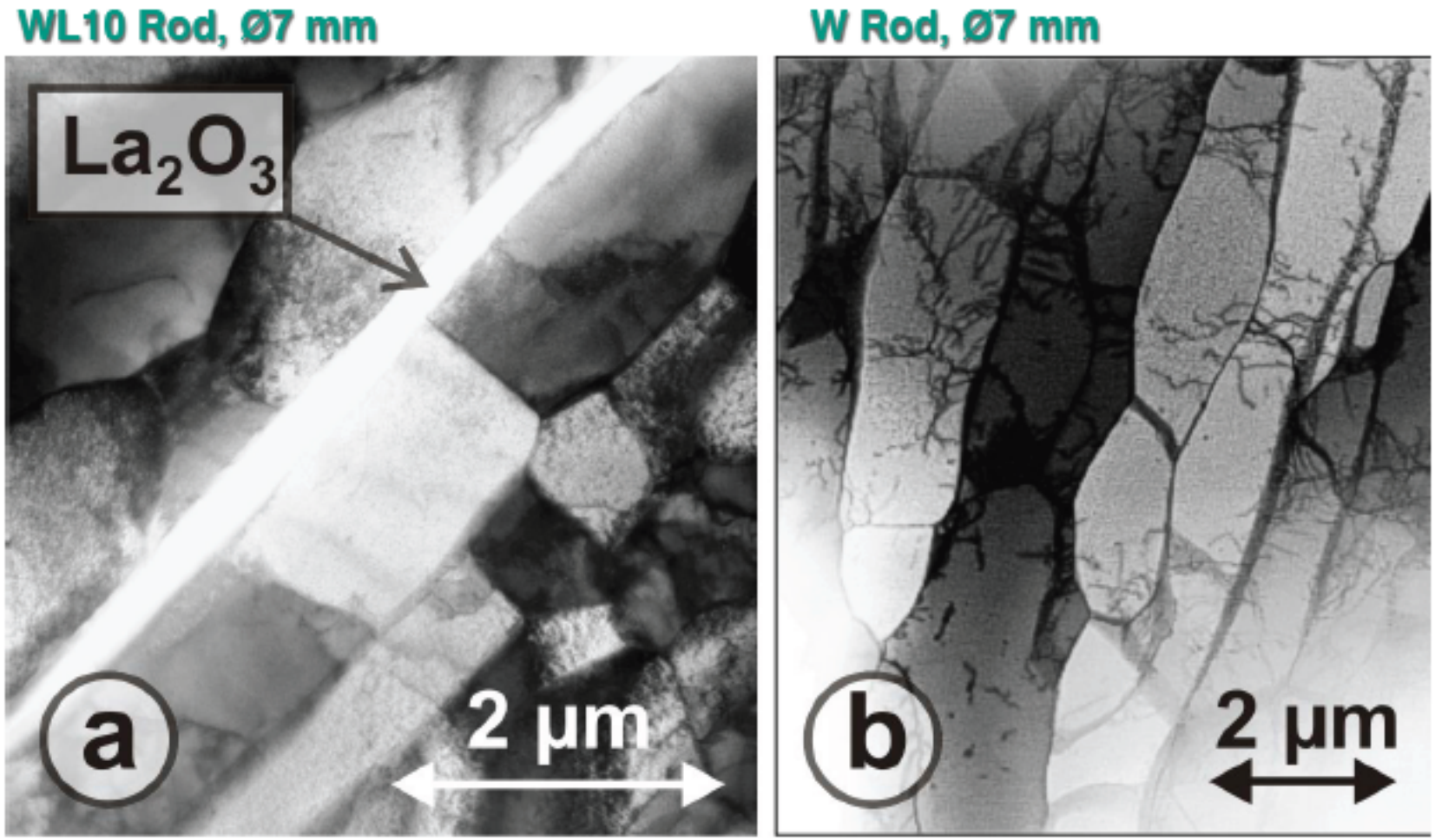
  - the application temperature (<1000°C) is < 0.4 Tm puts the material within the low temperature embrittlement regime

- Currently, the manufacturing technology for tungsten and tungsten alloys has struggled to produced "structural" tungsten

Manufacturing of Present Day Tungsten

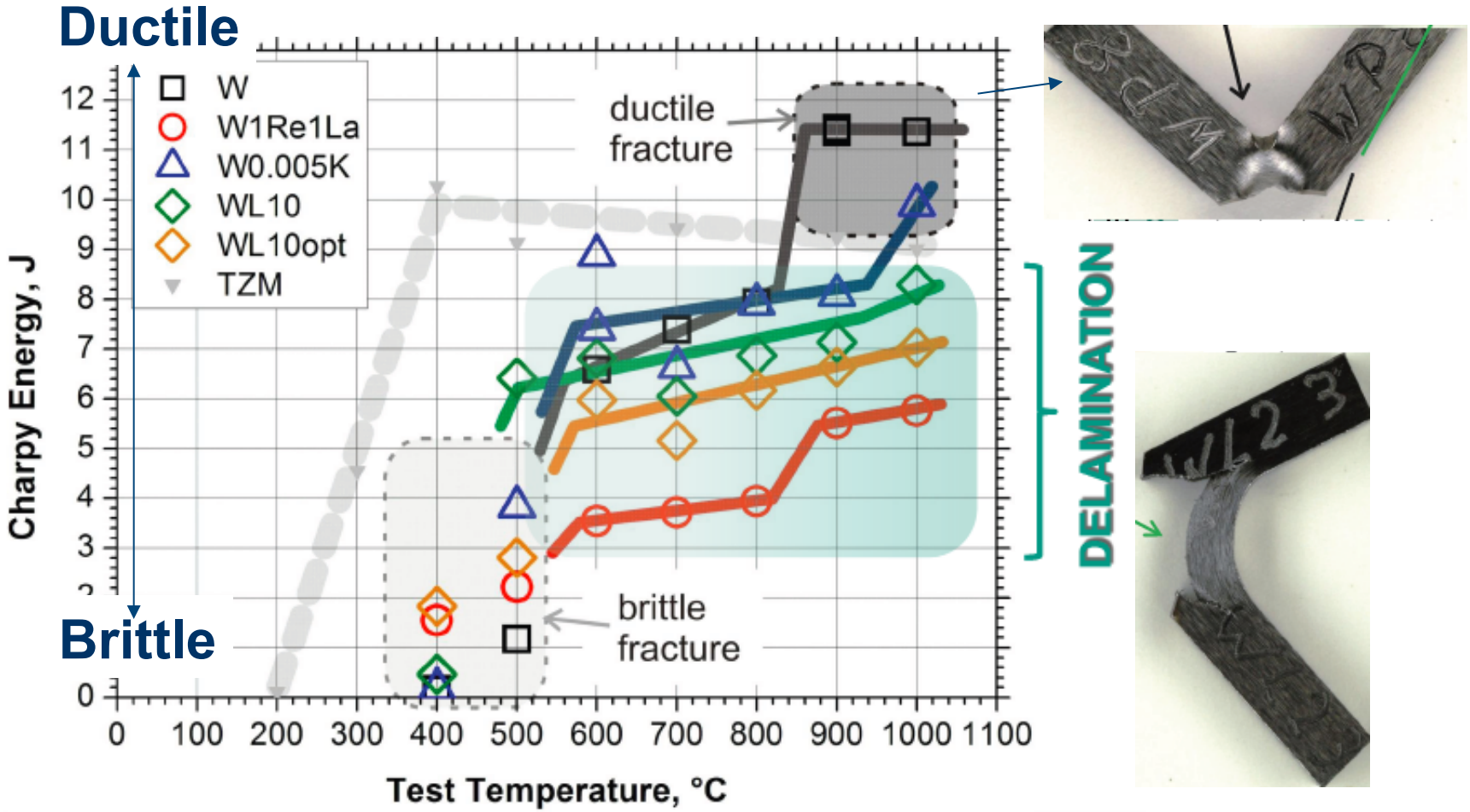


Microstructure



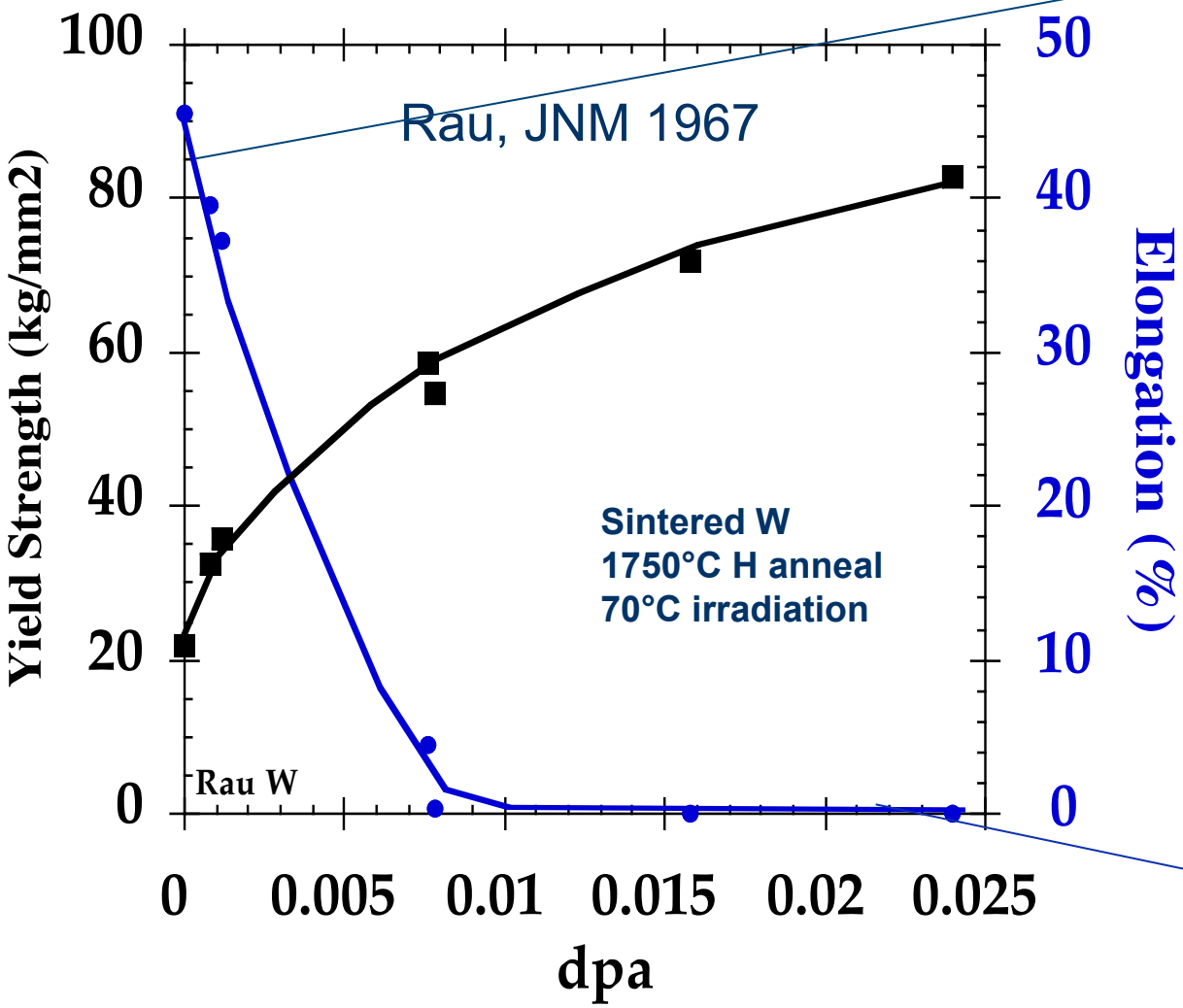
- properties can be highly non-uniform and process ductility has been gained by using additions of materials such as La_2O_3, which resides at grain boundaries.

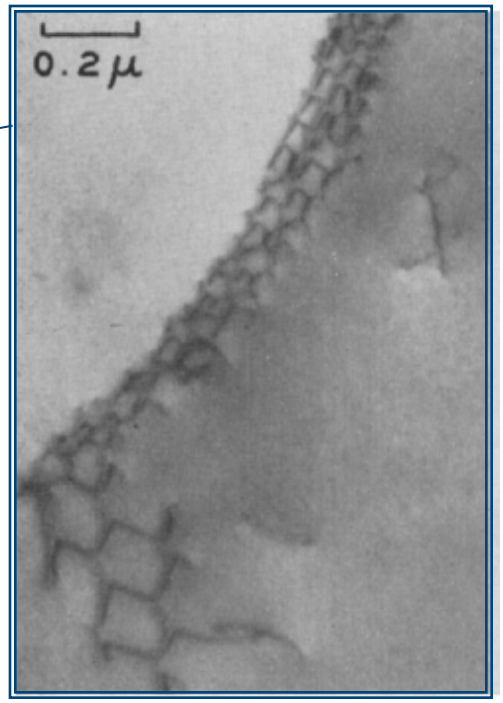
Fracture Characteristics of W Rod Materials

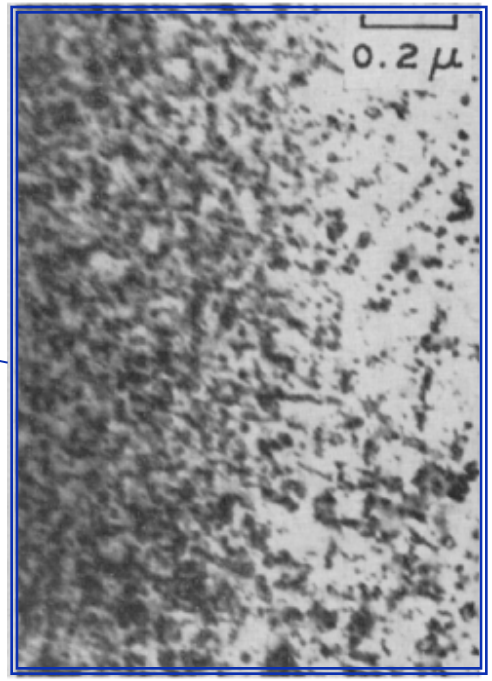


- Rod : Transition from brittle to ductile behavior for powder metallurgy processed materials, in the non-irradiated condition, is above room temperature. Upon irradiation, the transition temperature would further increase.
- Plate : Same situation, though properties are worse.

Tungsten under Irradiation

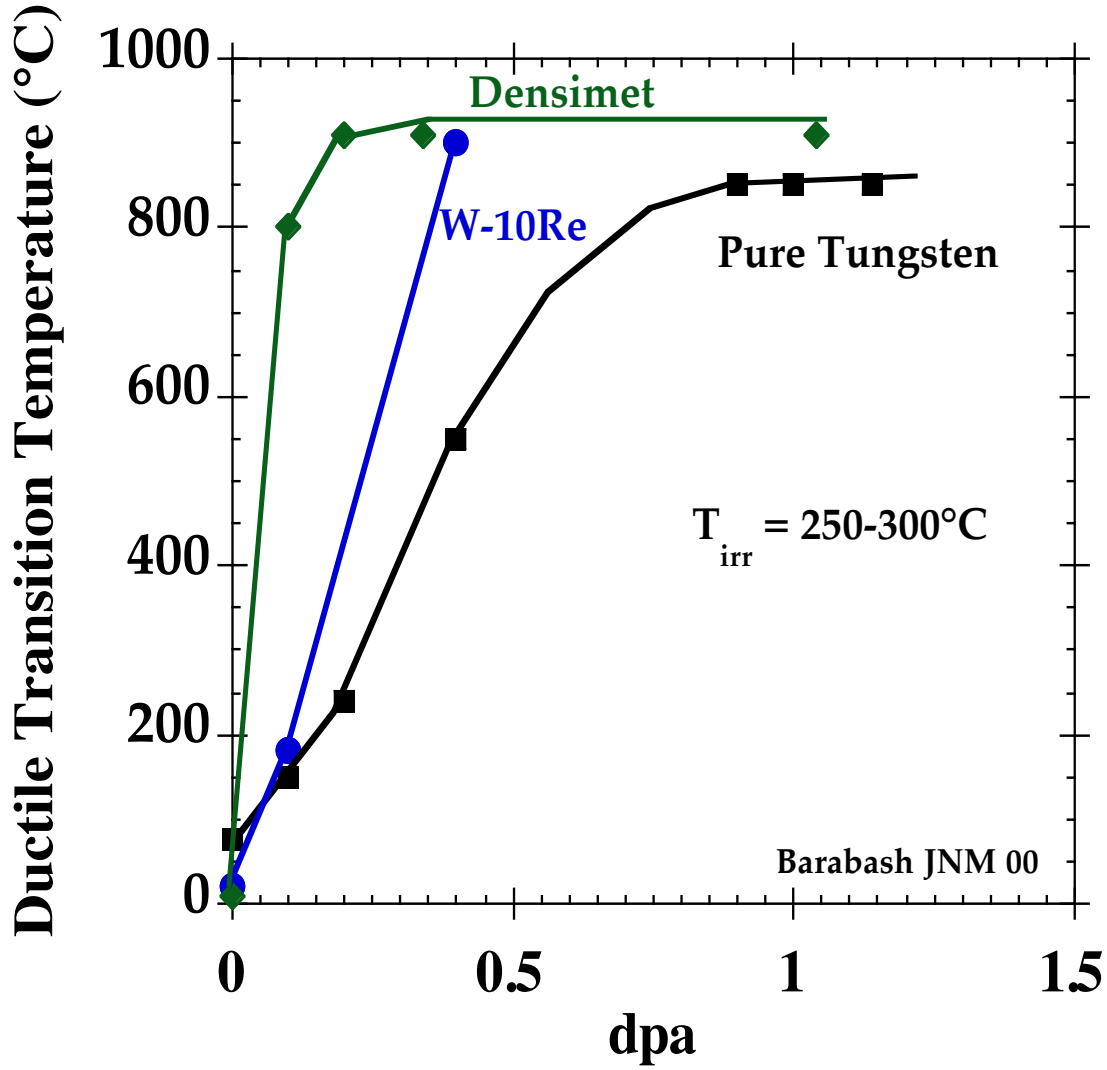






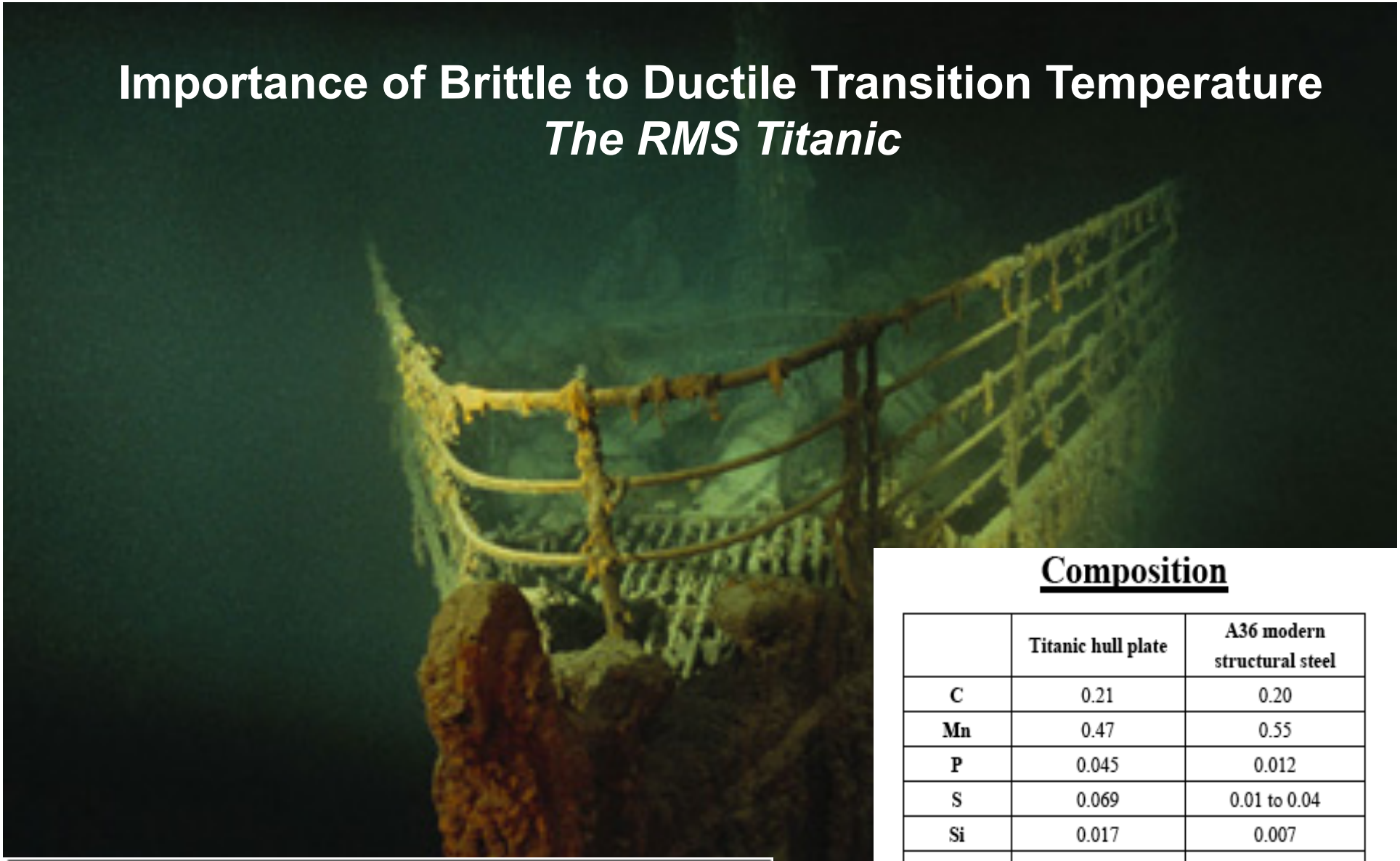
- Irradiation effects data on tungsten is rather limited, and typically on powder metallurgy samples. Results indicate severe embrittlement at low temperature.

Tungsten Under Irradiation



- Upon irradiation, the transition temperature for which tungsten goes from brittle to ductile behavior increased to well above room temperature.

Importance of Brittle to Ductile Transition Temperature
The RMS Titanic



	Titanic hull plate	A36 modern structural steel
Mn:S Ratio	7:1	15:1 (typical)

Composition

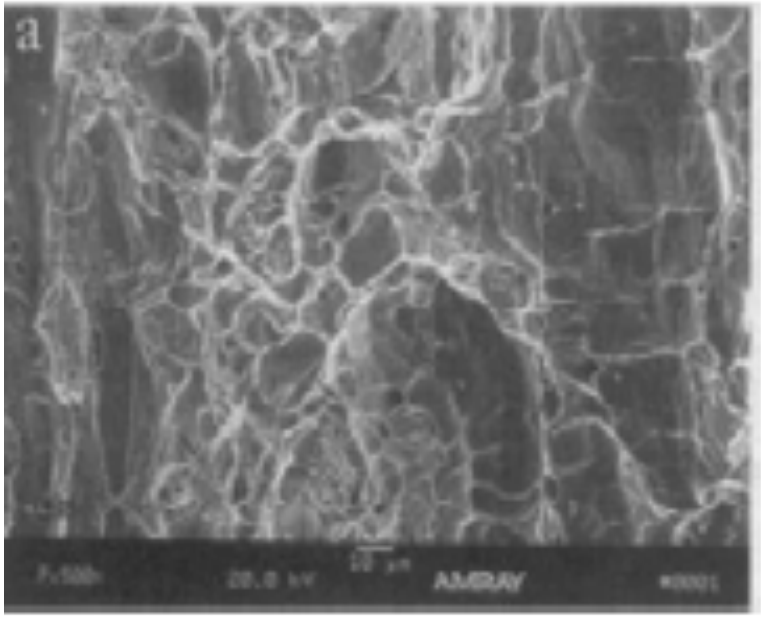
	Titanic hull plate	A36 modern structural steel
C	0.21	0.20
Mn	0.47	0.55
P	0.045	0.012
S	0.069	0.01 to 0.04
Si	0.017	0.007
Cu	0.024	0.01
O	0.013	-
N	0.0035	0.0032
Mn:S Ratio	7:1	15:1 (typical)

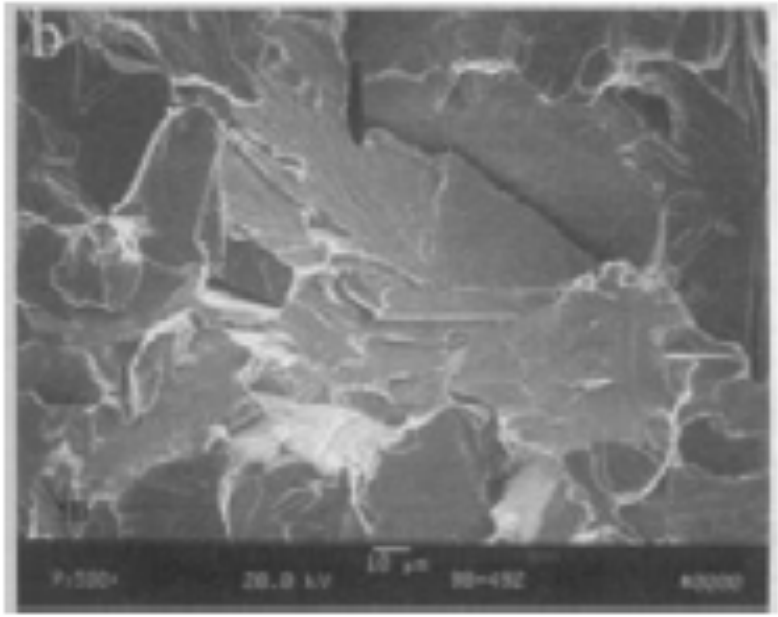
K. Felkins, H. P. Leighly, and A. Jankovic. JOM, 50(1), 1998, 12-18

Importance of Brittle to Ductile Transition Temperature

Fracture surface of Charpy specimens from Titanic plate

Longitudinal direction

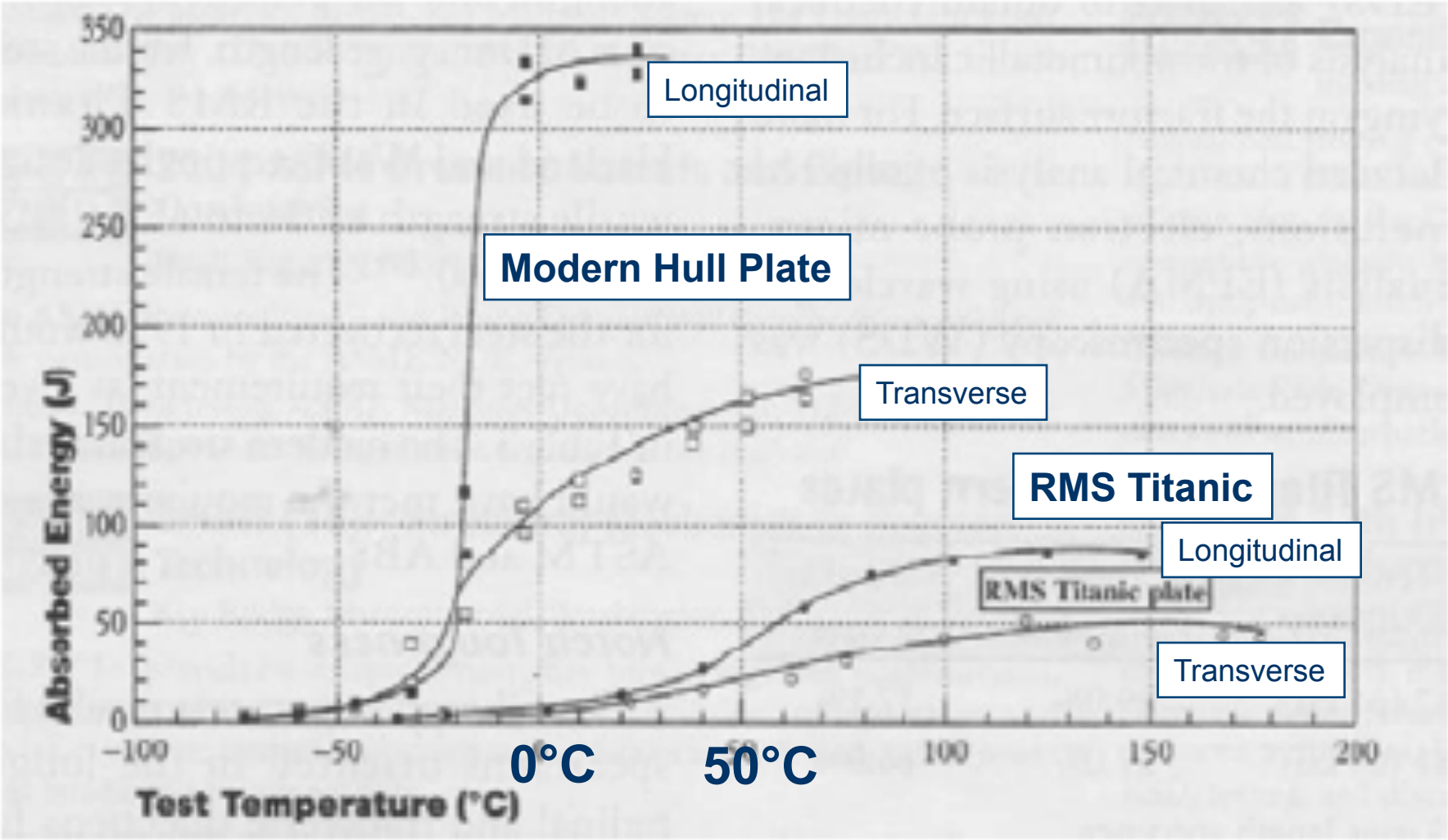




At 120 °C,
ductile fracture

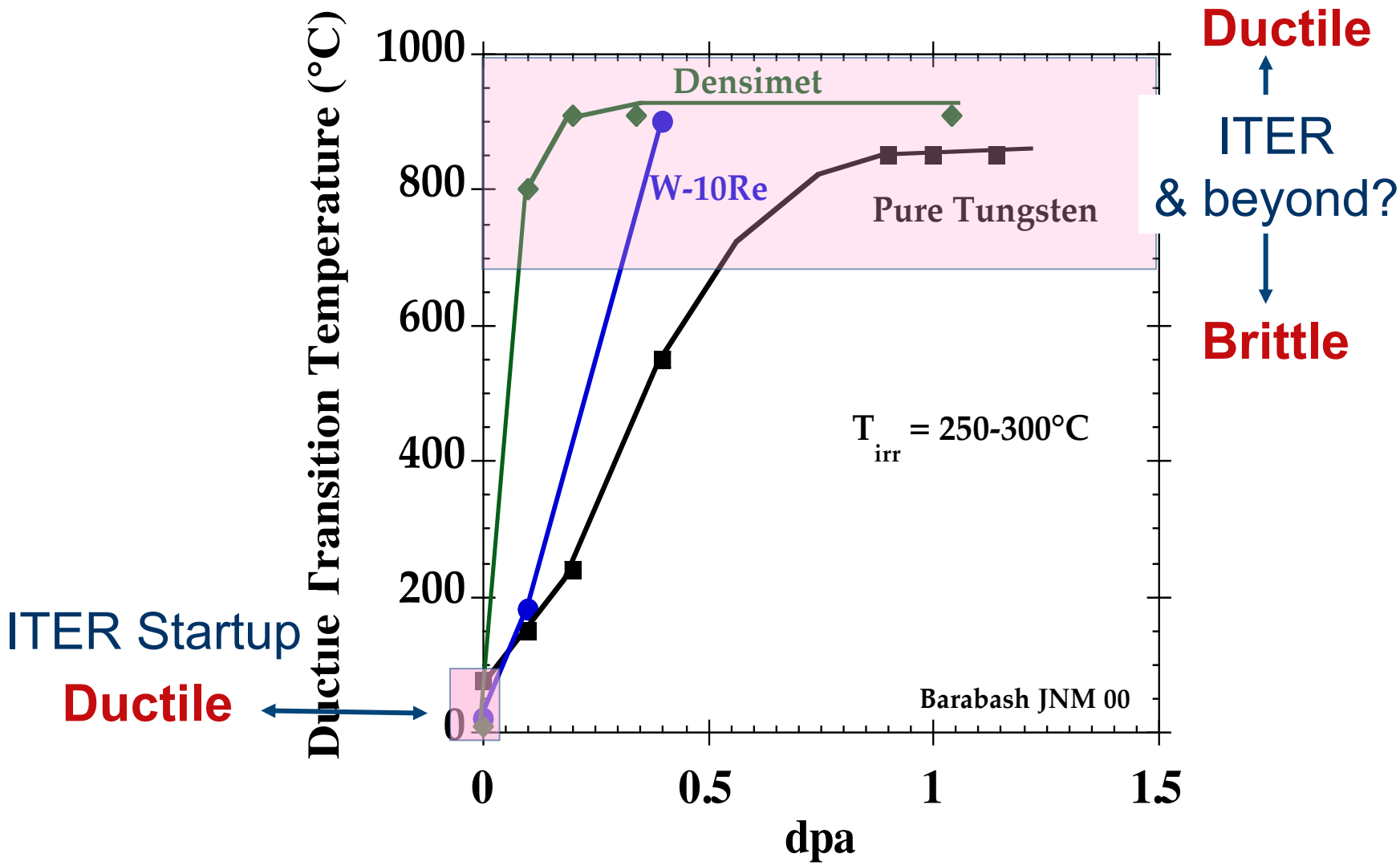
At -32 °C,
brittle fracture

Effect of temperature on toughness



H. P. Leighly, B. L. Bramfitt, and S. J. Lawrence. Practical Failure Analysis, 1(2), 2001

Tungsten Under Irradiation



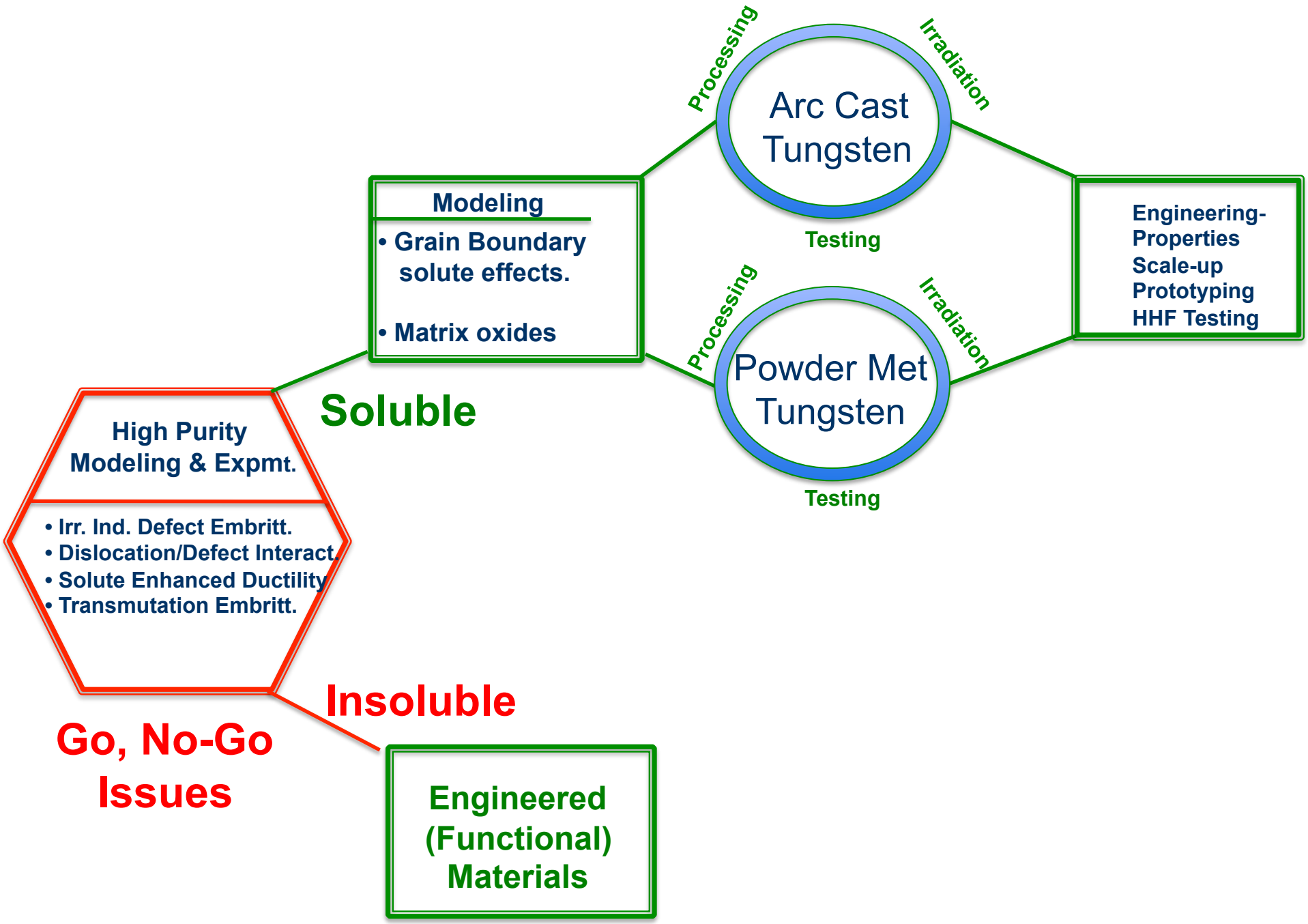
- Present forms of tungsten undergo significant embrittlement following low dose irradiation. It currently appears that for end of life ITER and higher dose fusion reactors, tungsten alloys will be fully brittle.

The Question of Structural Application of Brittle Materials and the Use of Engineered or Composite Structures

- The ITER project has pushed the lower limit of "engineering acceptability" of ductility (fracture toughness) to lower levels than in standard code use. Those levels are very likely still too high for the use of refractory metals (or ceramics…)

- An open question is whether designers can learn to design components with very low ductility materials and how these can be codified.

- If it is determined that this is not possible, and we make a judgment that we have taken materials as far as we can, then the next option is to move to engineered / composite structures.


Processing
Irradiation
Arc Cast Tungsten
Testing
Modeling
• Grain Boundary solute effects.
• Matrix oxides
Engineering-Properties
Scale-up
Prototyping
HHF Testing
Processing
Irradiation
Powder Met Tungsten
Testing
Soluble
High Purity Modeling & Expmt.
• Irr. Ind. Defect Embritt.
• Dislocation/Defect Interact.
• Solute Enhanced Ductility
• Transmutation Embritt.
Insoluble
Go, No-Go Issues
Engineered (Functional) Materials

Biased Summary Comment on Tungsten

- Tungsten is potentially a very high performance material. However, its current structural applications (any application) are very limited due to its extreme difficulty in fabrication and limited ductility.

- The poor ductility of tungsten is an issue of both the nature of tungsten crystal, and the nature of the processing used (which controls the grain boundary.) Improvements are possible, and the current understanding of how alloying may improve tungsten ductility, is not well understood.

- Irradiation will make the ductility situation even worse. What is currently needed is a combination of fundamental studies on the irradiation effects
  - are in-cascade formed defects a low temperature limiting issue?
  - can be use solid solution alloying to enhance ductility?
  - are the current nano-composited tungsten alloys going to be effective?

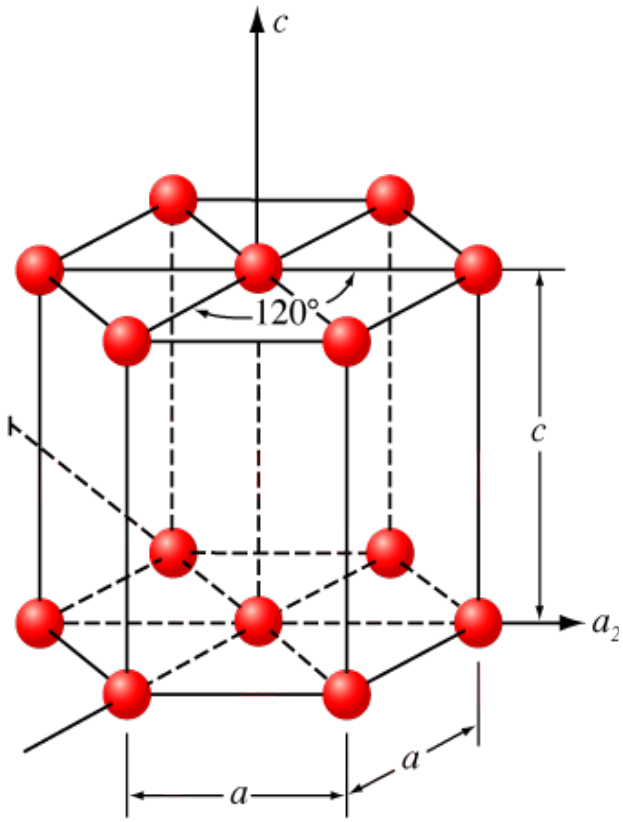
- We will likely (and should) push the envelope in both engineering design and materials development to define and extend performance limits and minimum requirement for materials.

Beryllium as an Engineering Material

- **Be has only two operating slip planes {0001} & {1010} significantly limiting the ductility of the metal.**
  - **T < 200°C, no {1010} slip, Ductility ~5%**
  - **200 < T < 500, {1010} & {0001}, Ductility to 50%**
  - **T > 500°C, grain boundary failure, Ductility < 20%**

- **For this reason key engineering properties such as fracture toughness, ultimate strength, and plastic elongation are poor.**

- **For the non-irradiated case, and especially in the neutron irradiated case, fabrication and design must consider working with a "brittle" material.**

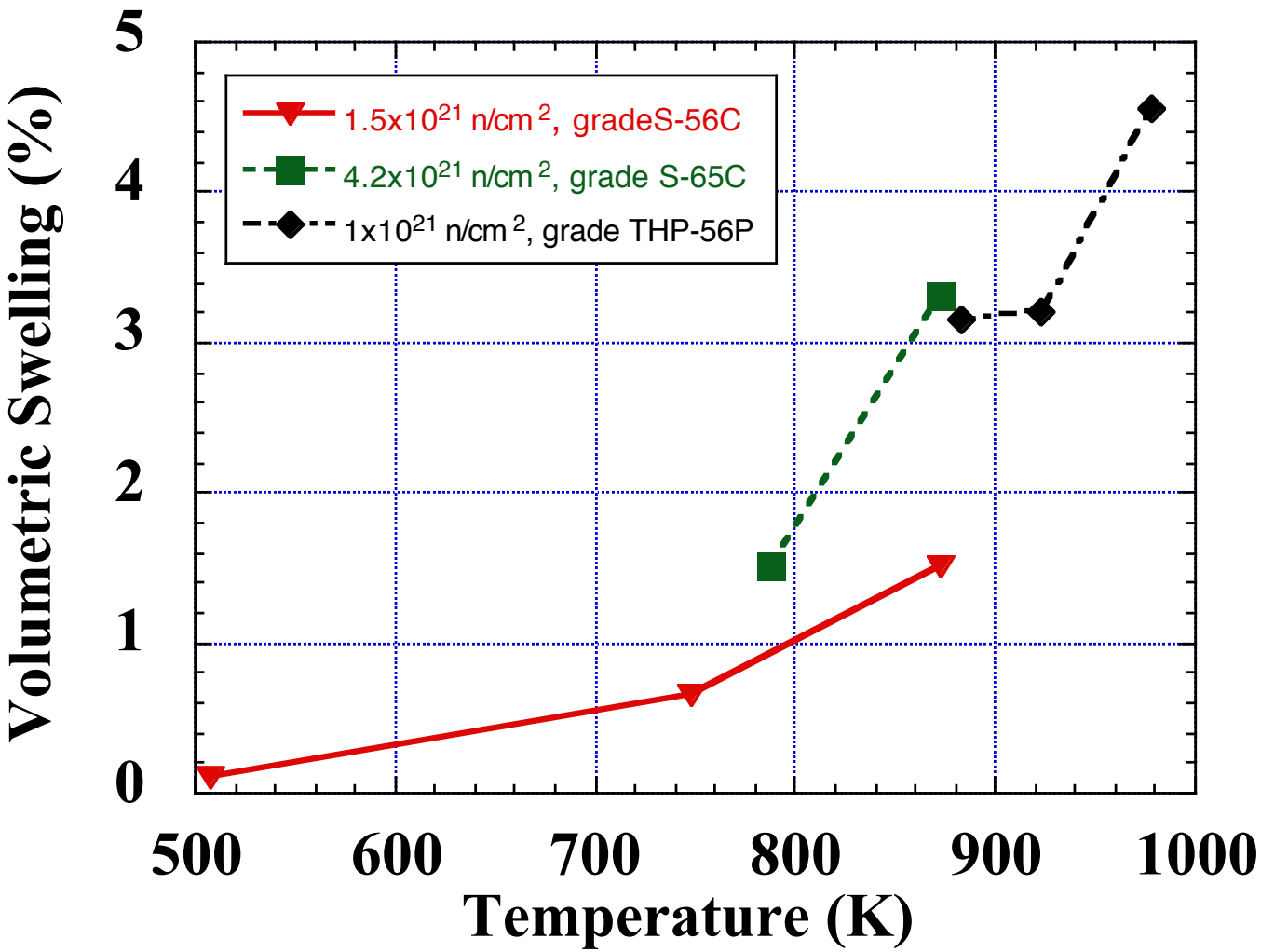


Hexagonal Close Packed Crystal

T_{melt}=1560°C

Effect of Neutron Irradiation on Be Metal

The primary effects of neutron irradiation on metallic Be: swelling and embrittlement



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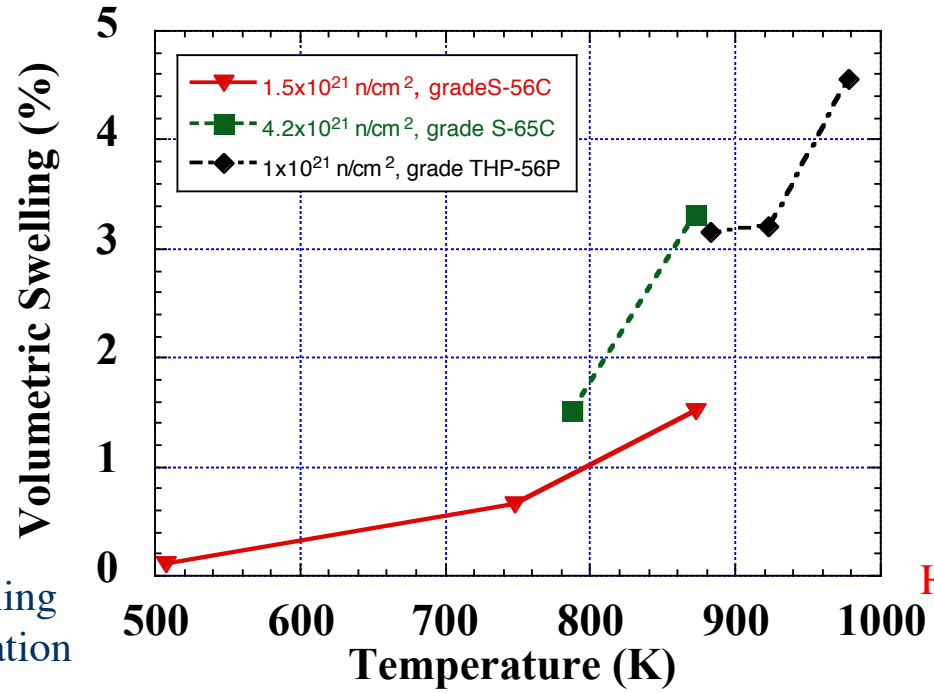
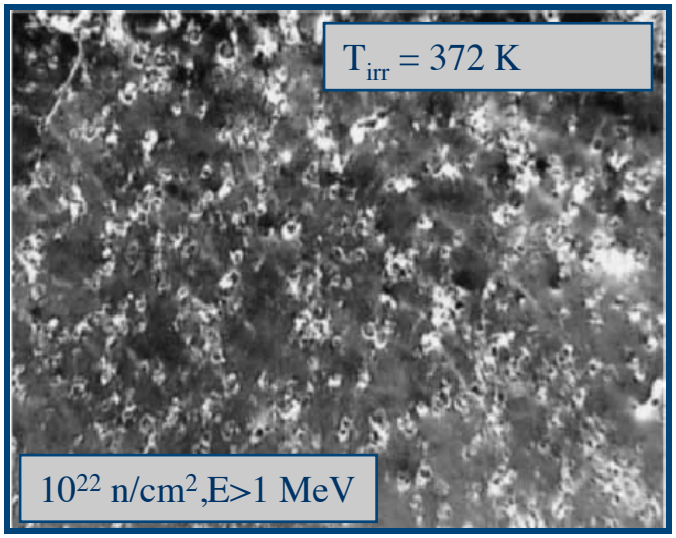


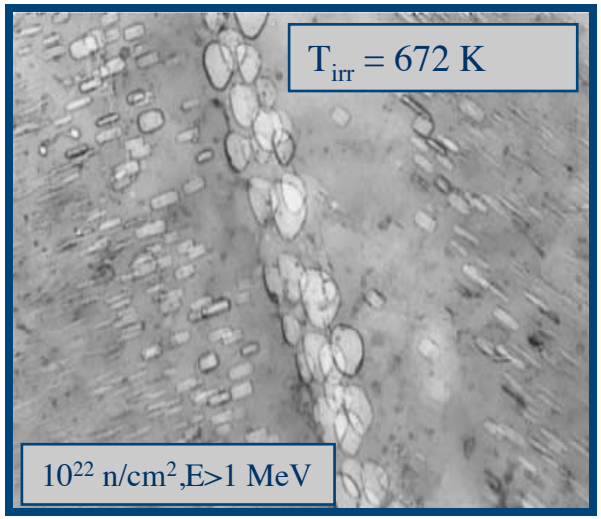
0.3 **< 0.3 T_{melt}**

Low Temperature Swelling due to Defect Accumulation

0.6 **0.6 T_{melt}**

High Temperature Swelling due to He Accumulation: --> B^9(n, He) reaction.



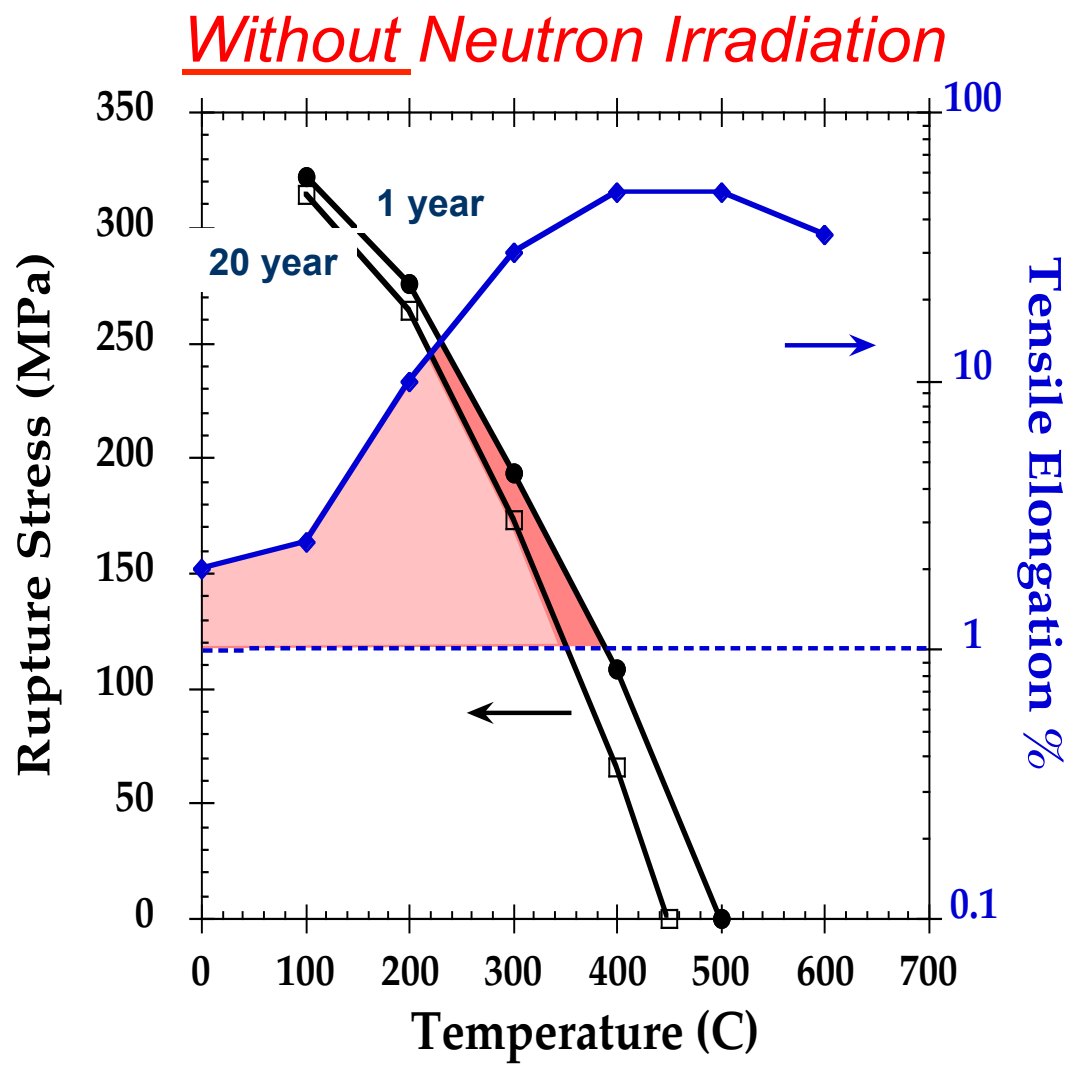


Defining an Operating Window for Structural Use of Be

- Operating windows are typically defined using following properties:
  - Creep
  - Fracture toughness
  - Fatigue
  - Ductility limits (plasticity, elongation)
  - Environmental effects

- There are no creep data on modern-day Be, though Larson Miller data exist to estimate the rupture lifetime.

- Design Space Assumptions:
  - Upper stress using extrapolated Larson Miller data.
  - Assume lower bound set by required elongation of 1% total.



Example Fracture Behavior of Beryllium : Fracture Surface

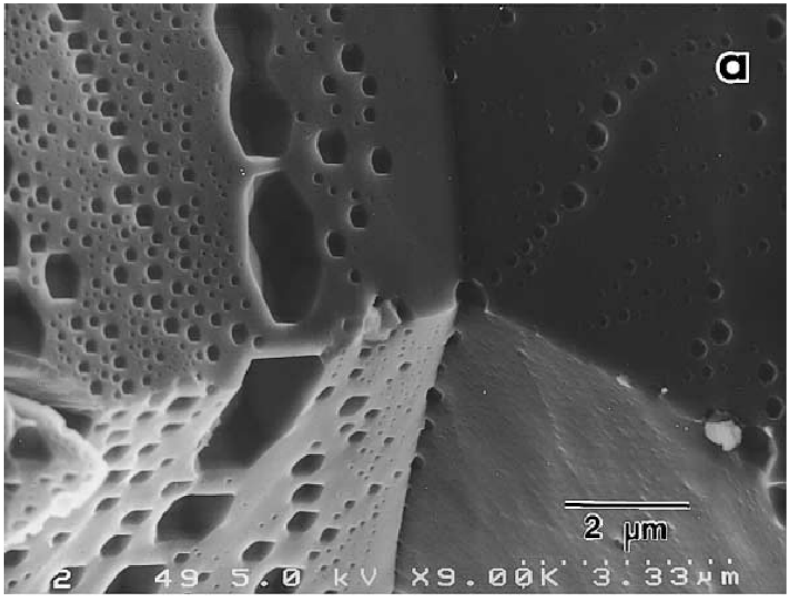
- At elevated temperature, neutron irradiation causes He bubbles to form and embrittle grain boundaries.

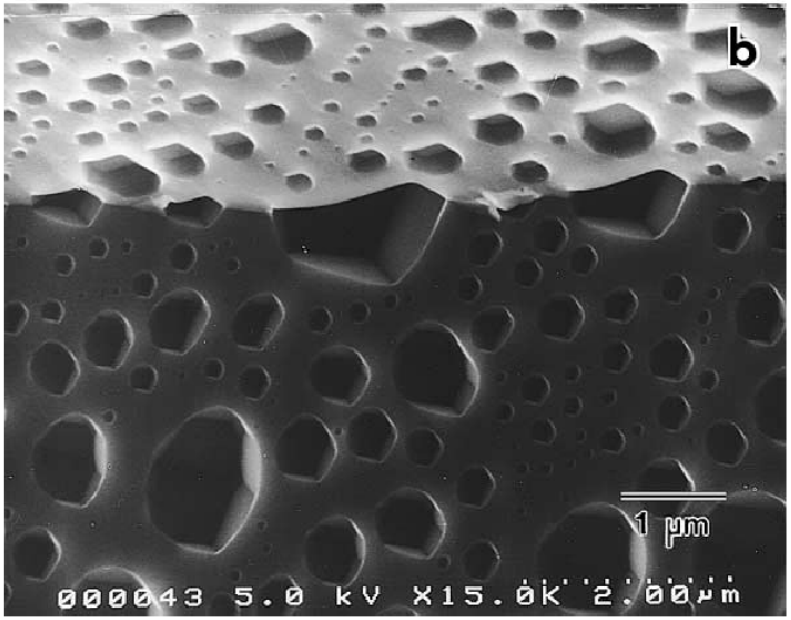
- Formation of bubbles and their catastropic effect on mechanical properties have been slightly improved by material purification, but significant embrittlement is unavoidable.

- Example of Improvement:

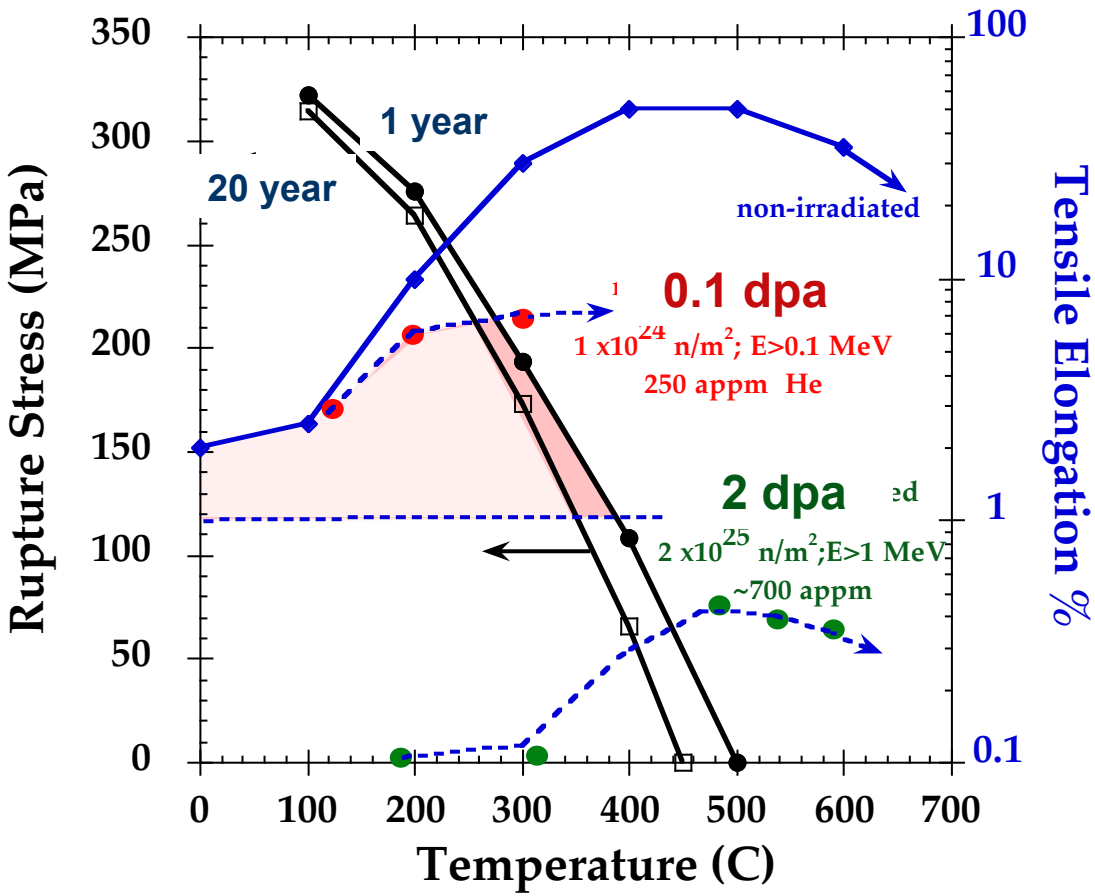
1960's vintage : 1×10^{24} n/m^2 E>0.1 MeV
total elongation < 0.2%

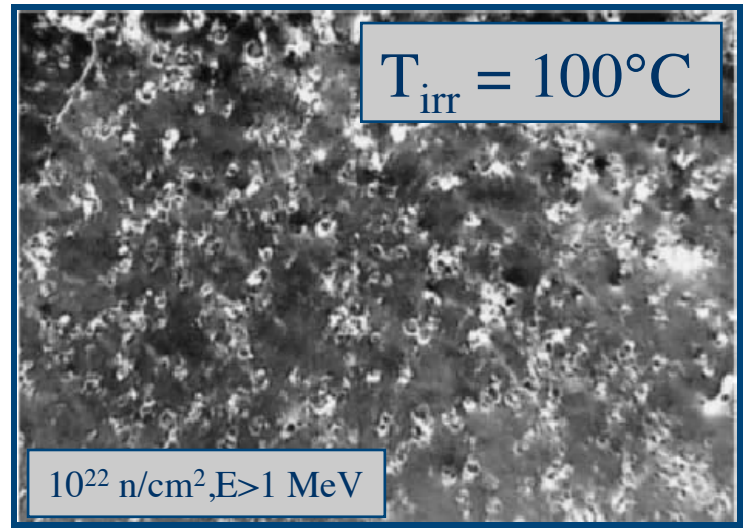
1990's vintage : 1×10^{24} n/m^2 E>0.1 MeV
total elongation few %

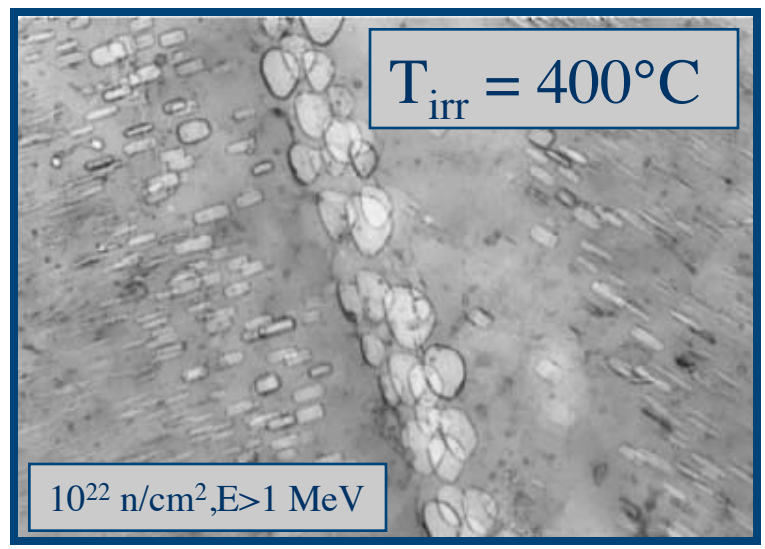




With Neutron Irradiation







< 0.001 dpa	0.01-1 dpa - ITER -	>1 dpa - beyond ITER-
Essentially non-irradiated	Embrittlement a factor Swelling no problem	Embrittlement and Swelling Unavoidable

Biased Summary Comments on Beryllium

- Beryllium has been well studied and its use as a plasma facing material requires very high purity materials. Alloying of beryllium will certainly improve mechanical properties, but the consequences on the plasma will likely be unacceptable.

- Improvement in recent years have been largely in the improvement in the grain boundary integrity (removal of oxygen.) Further, incremental improvement is possible, but low temperature embrittlement and helium swelling due to neutron irradiation is unavoidable.

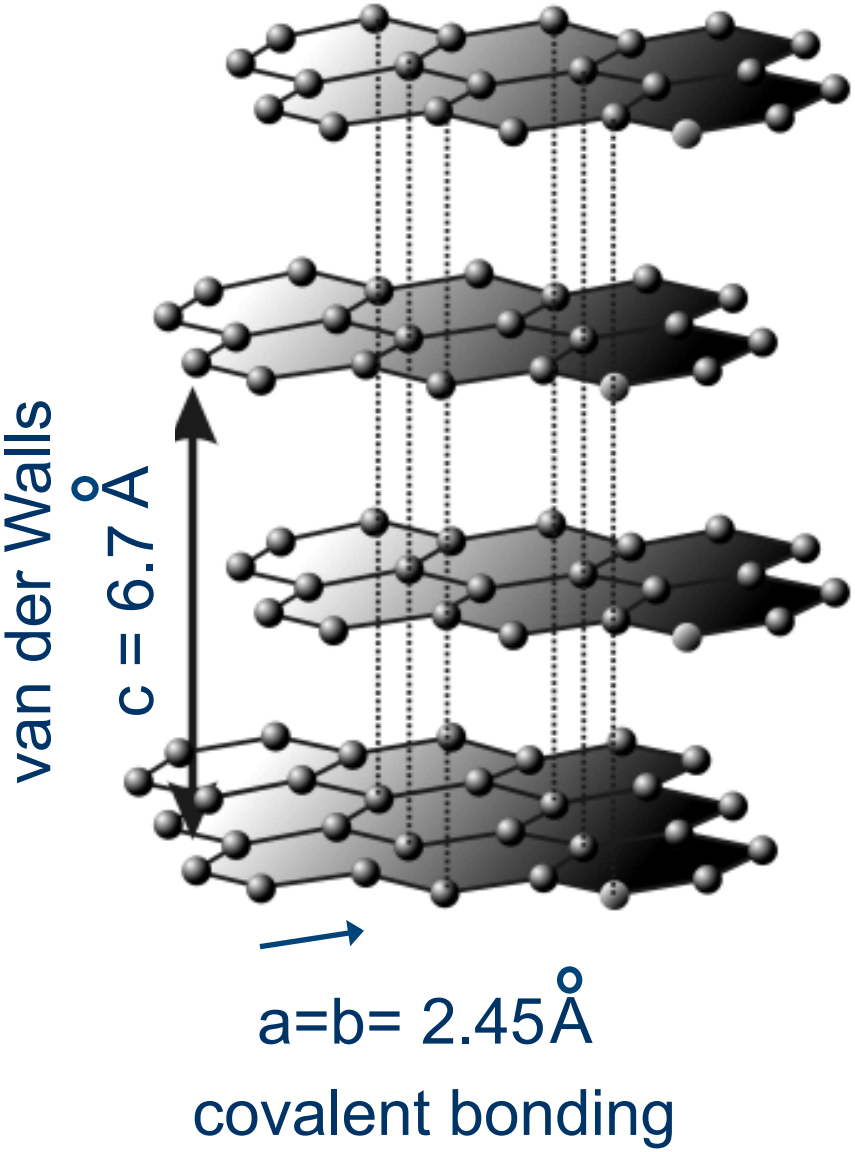
The Graphite Crystal

- The graphite crystal is an interpenetrating hexagonal "benzene" ring structure.

- Very weak bonding between planes, strong bonding in planes.

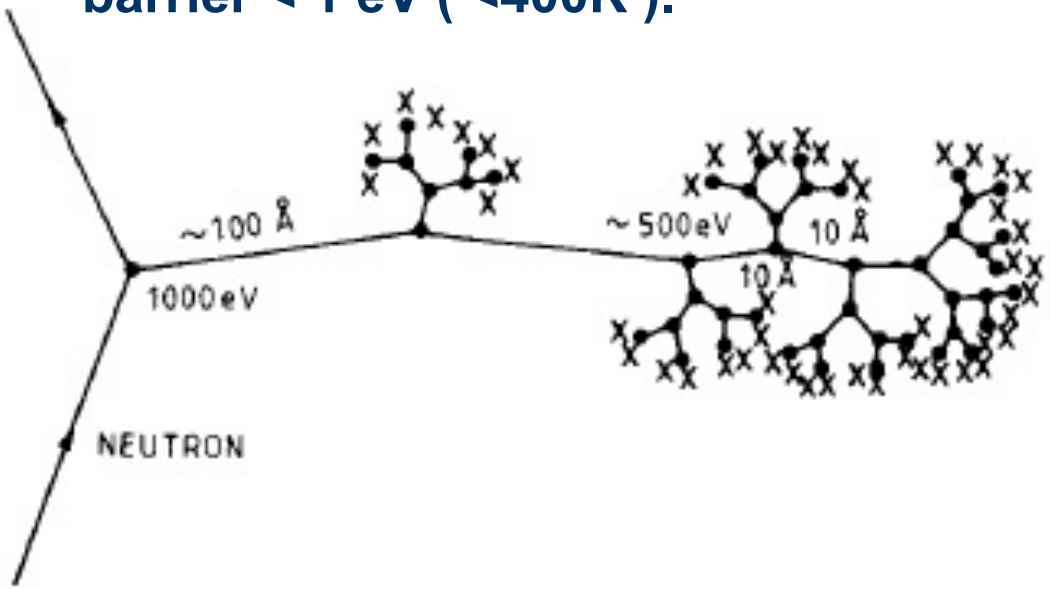
- Extraordinary in-plane properties, drastically different out-of-plane.

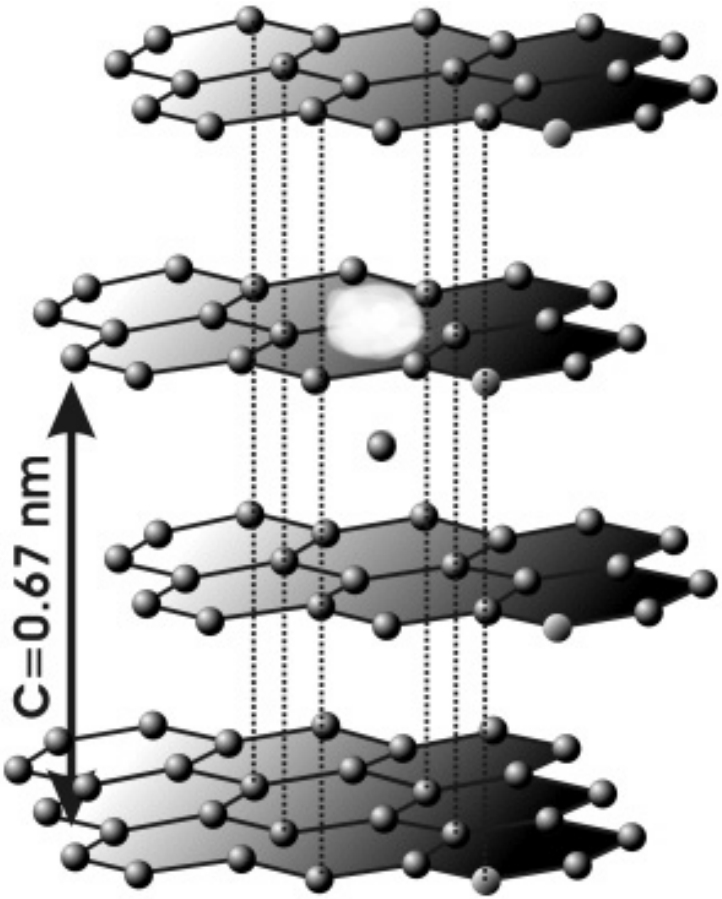
	In-plane	Out-plane
Thermal Conductivity W/m-K)	>2200 W	20
Thermal Expansion	0.5	6.5
Strength (MPa)	>1000	<1
Elastic Modulus (GPa)	20	<1



Irradiated Graphite Crystal

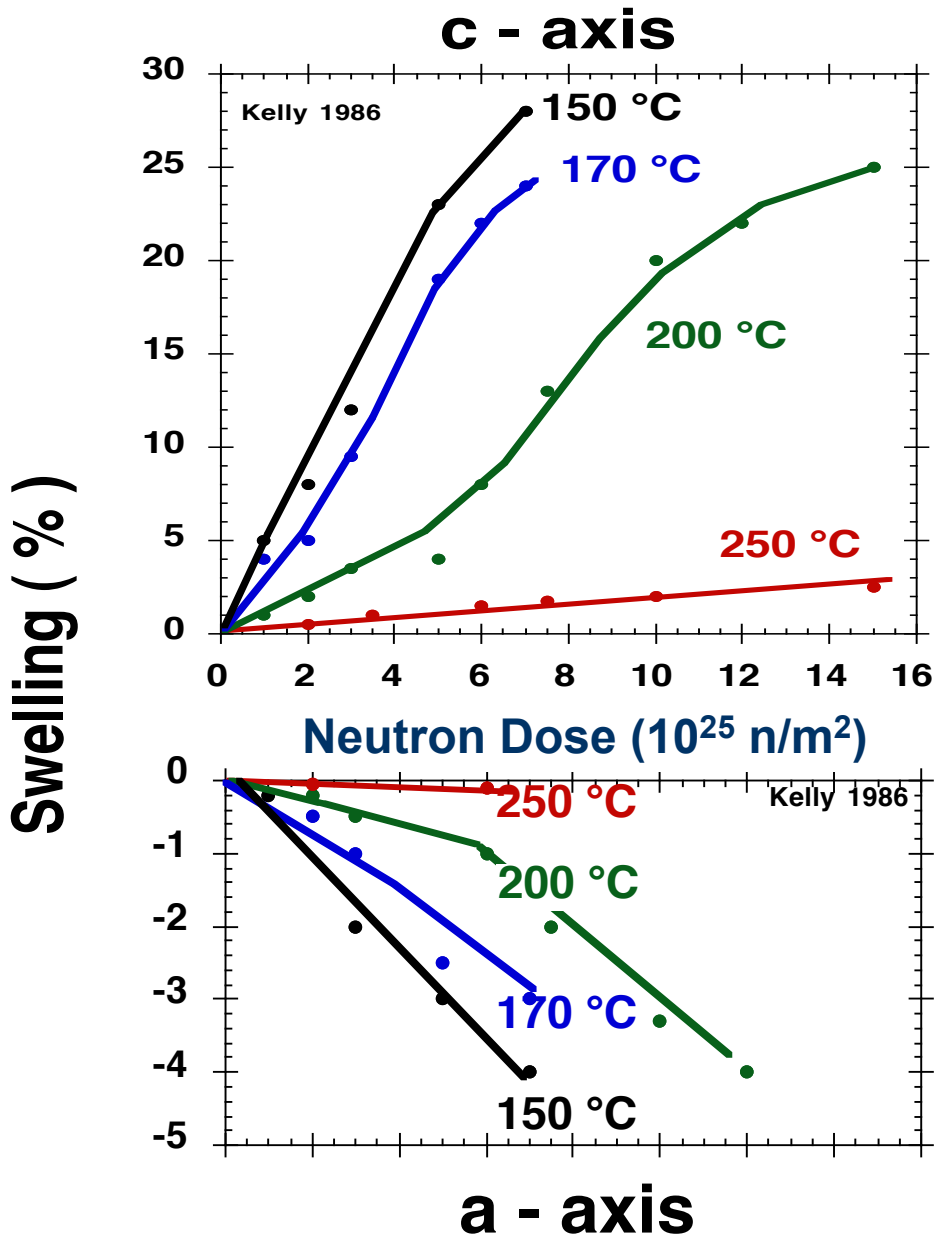
- Interstitials mobile > 70 K, move within the basal plane.
- Vacancies mobile > 1000 K, move freely between the basal plane.
- Interstitial-vacancy recombination barrier < 1 eV (<400K).

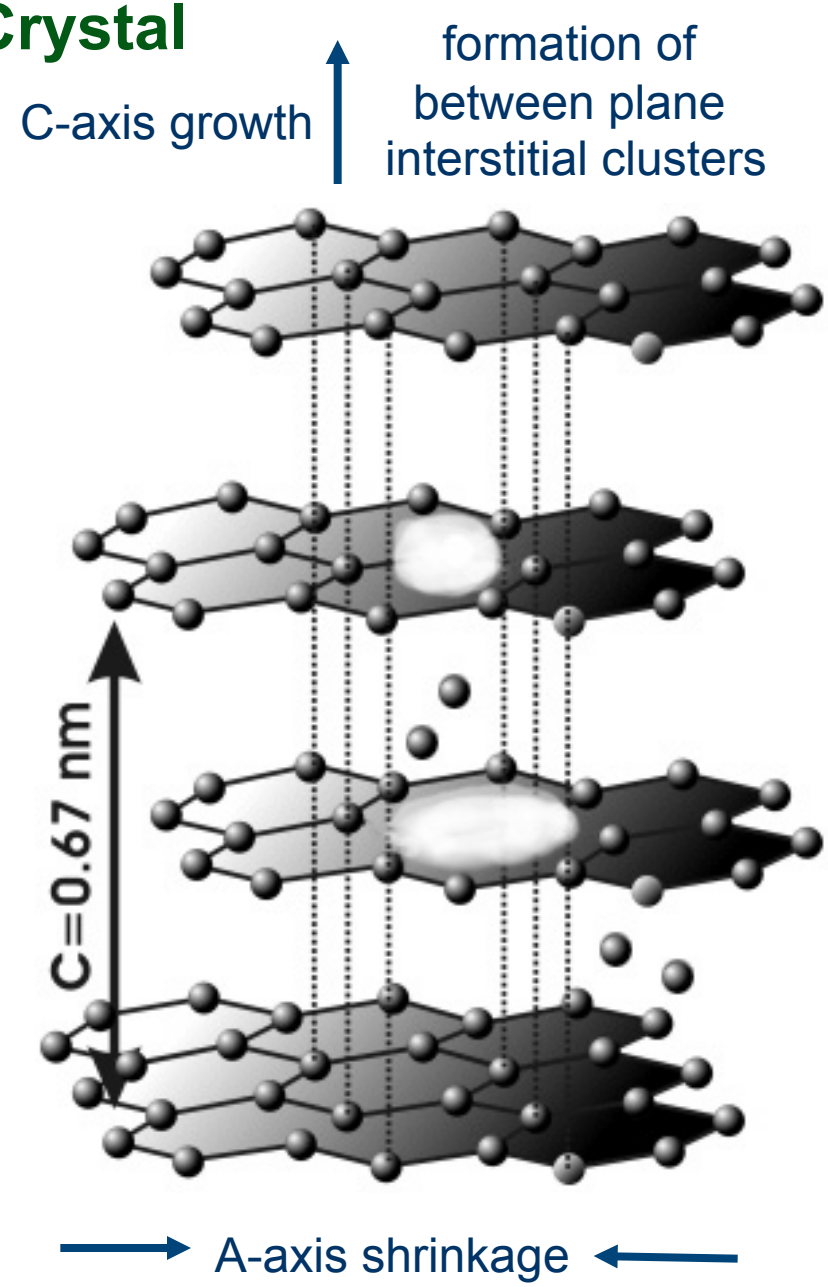




Dimensional Change in Graphite Crystal

Low Temperature

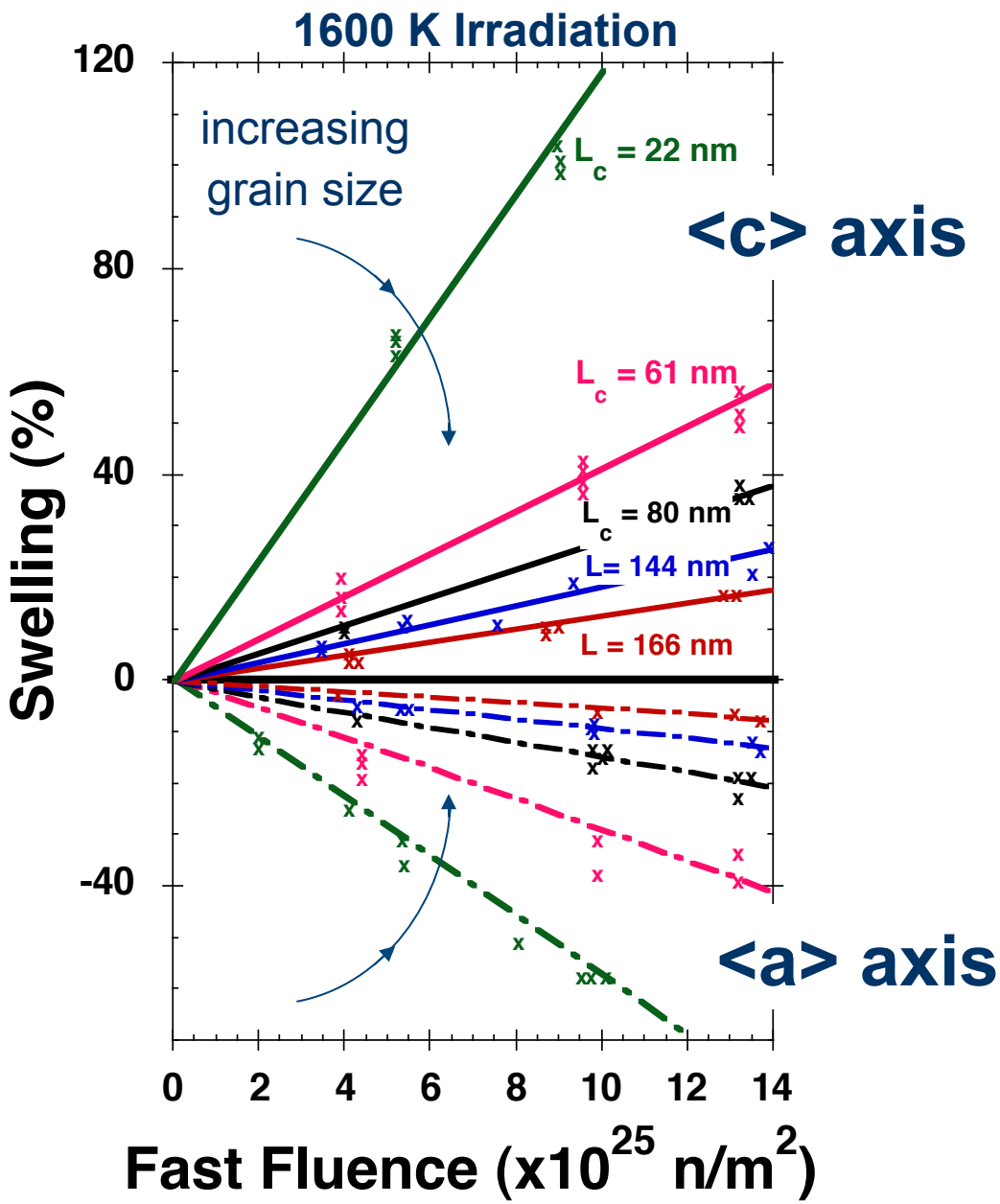




Overall Large Volume Increase

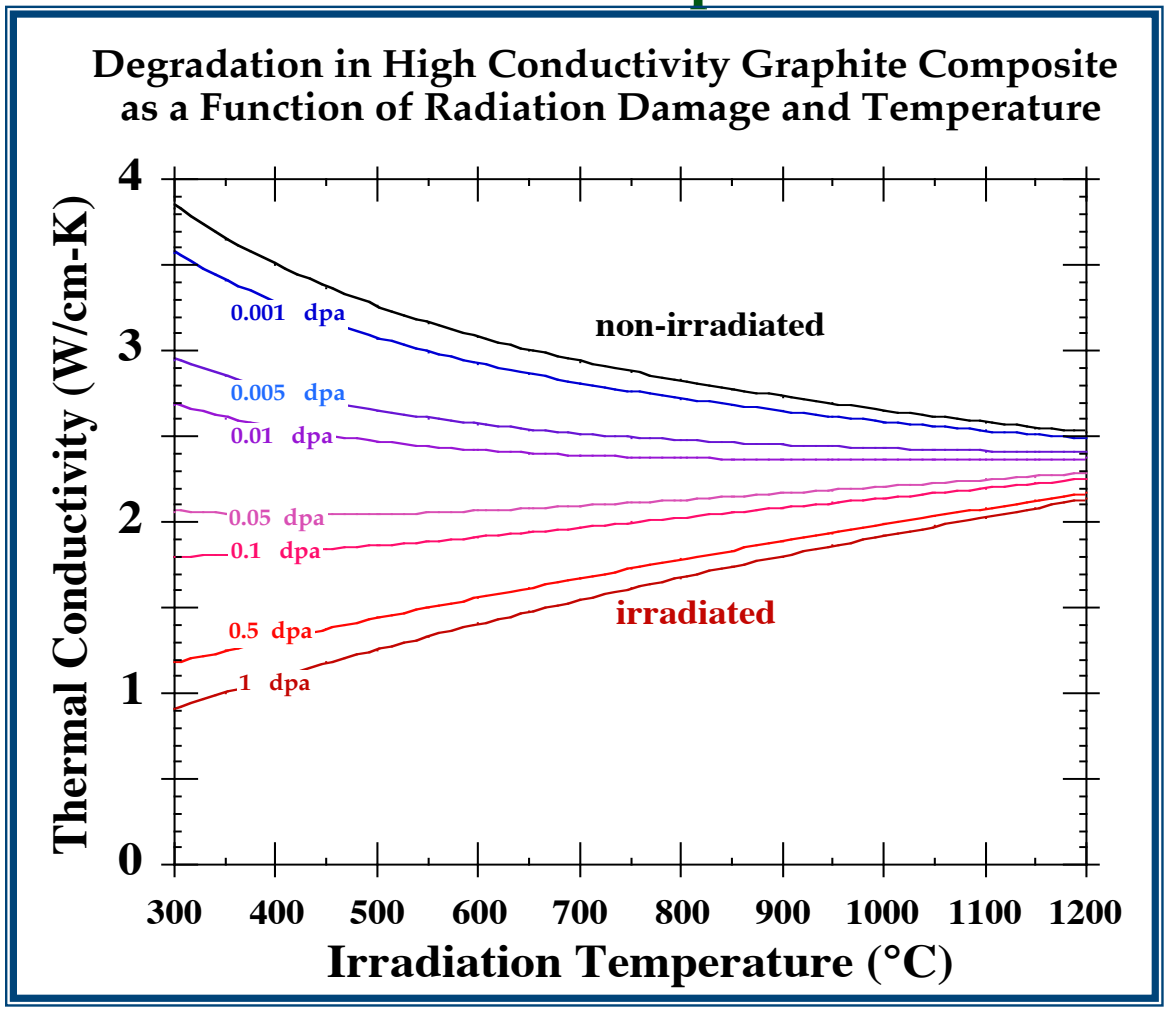
Dimensional Change in Graphite Crystal

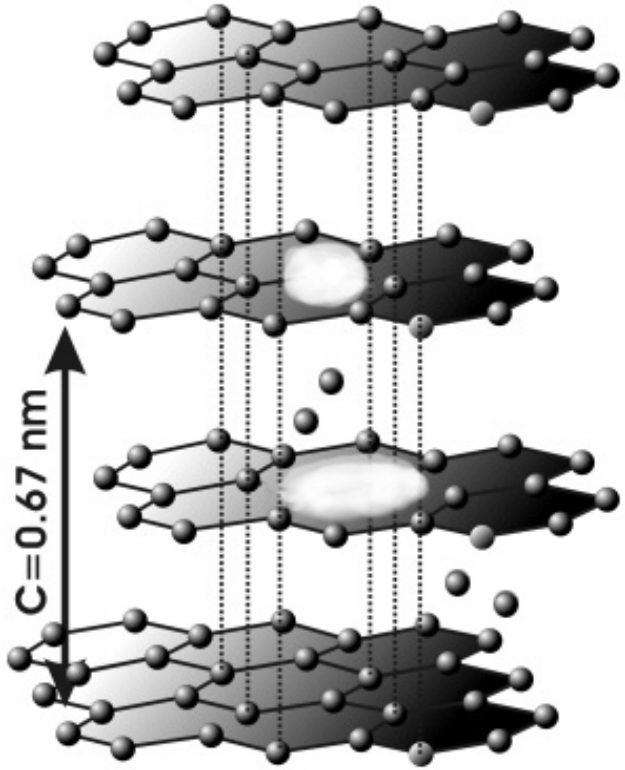
High Temperature



- Above ~1000 K both vacancy and interstitials are mobile.
- Dimensional change occurs to high dose <u>conserving volume</u>.
- Higher crystal perfection material suffers less dimensional change due to reduced pinning centers for migrating defects.

Irradiation-Degraded Thermal Conductivity
- Graphite -

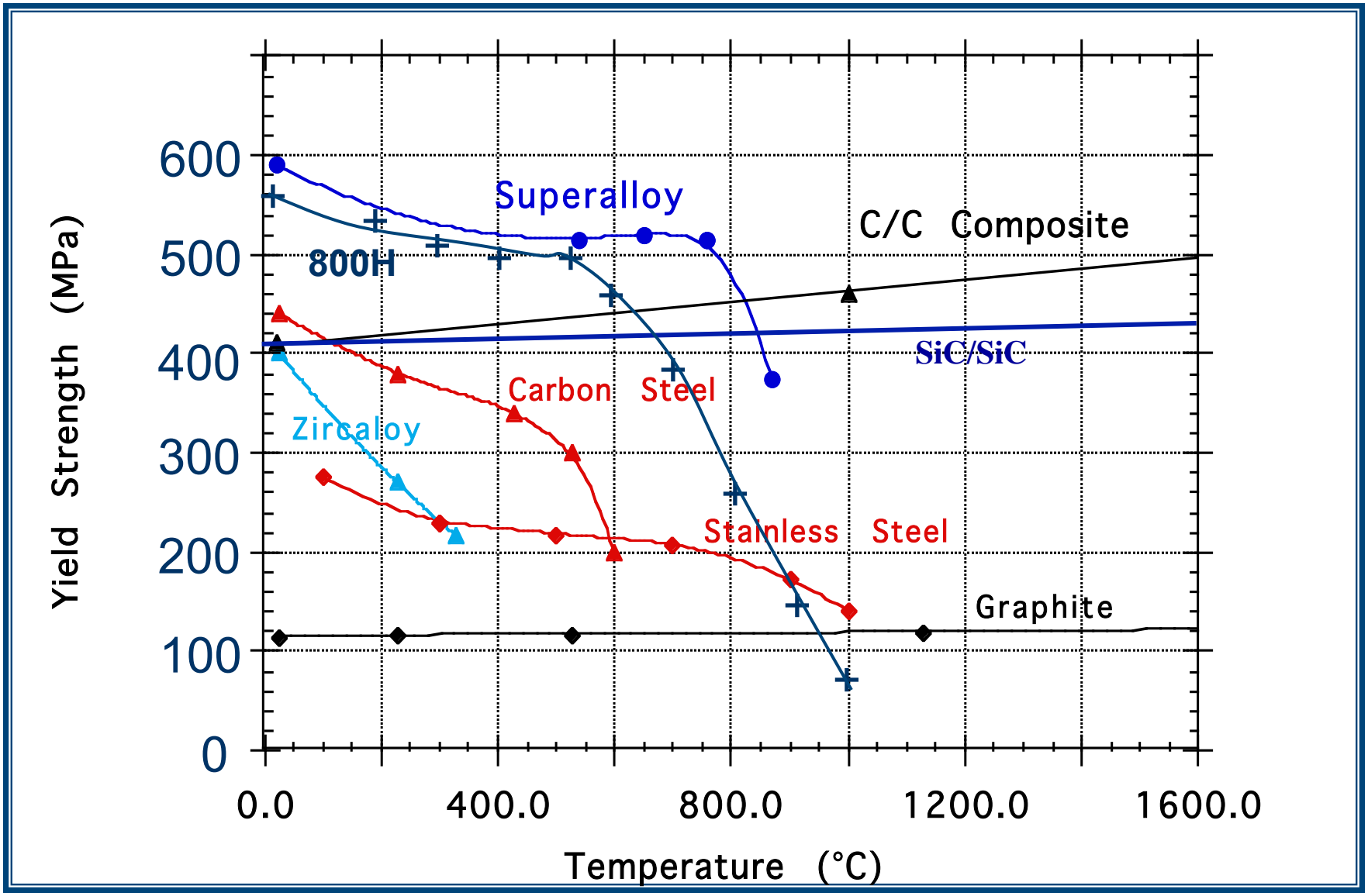




Conductivity reduction ascribed to formation of 4±2 vacancy clusters

- **Thermal conductivity in graphite is dominated by phonon transport.**
- **Vacancy complexes formed during irradiation are extremely effective at scattering phonons and degrading thermal conductivity.**

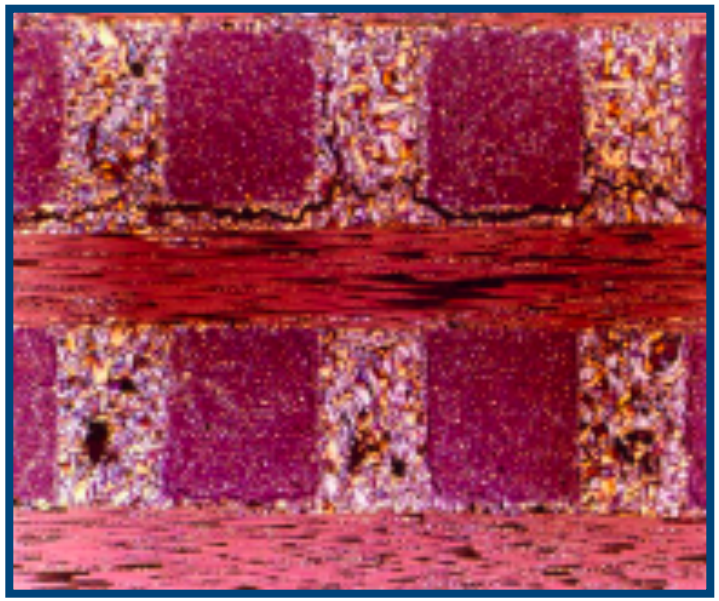
Yield Strength of Various Structural Materials

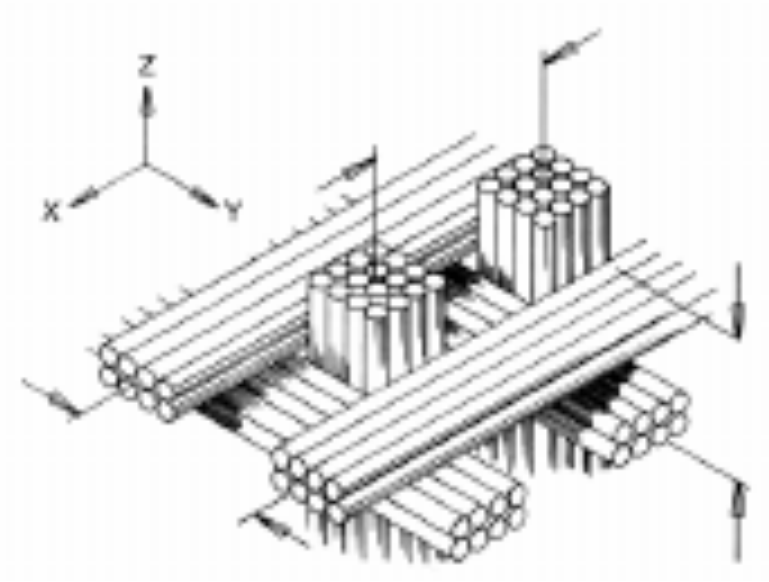


Carbon Fiber Composites

- Composites as being defined here are technically “continuous fiber reinforced composites,” the two most mature of which are Carbon (graphite) Fiber Composites (CFC’s) and Silicon Carbide fiber composites (SiC/SiC.)

- Of the two, the CFC is the more mature system, though they are similar in terms of processing status and cost.

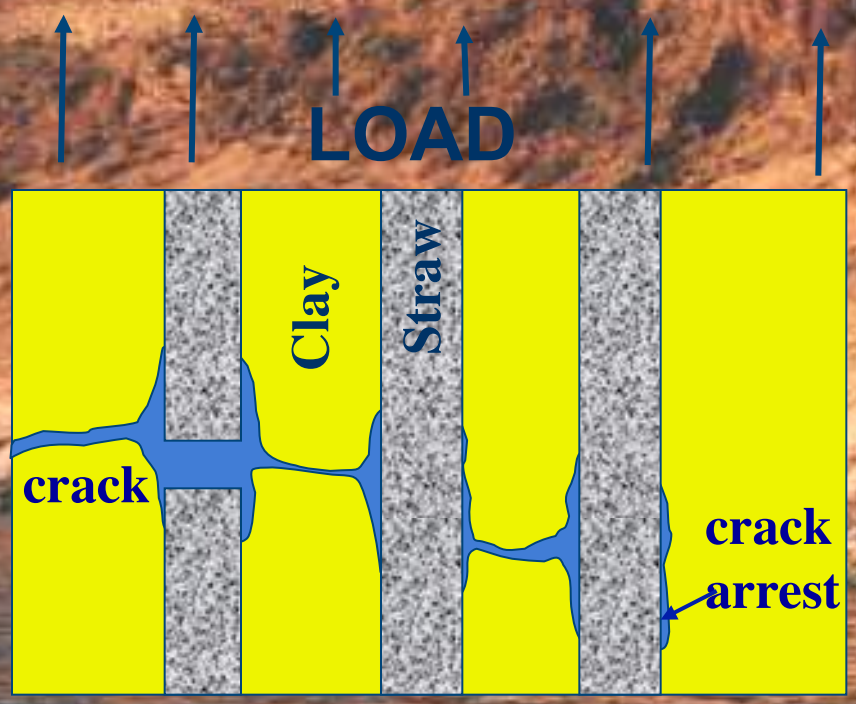


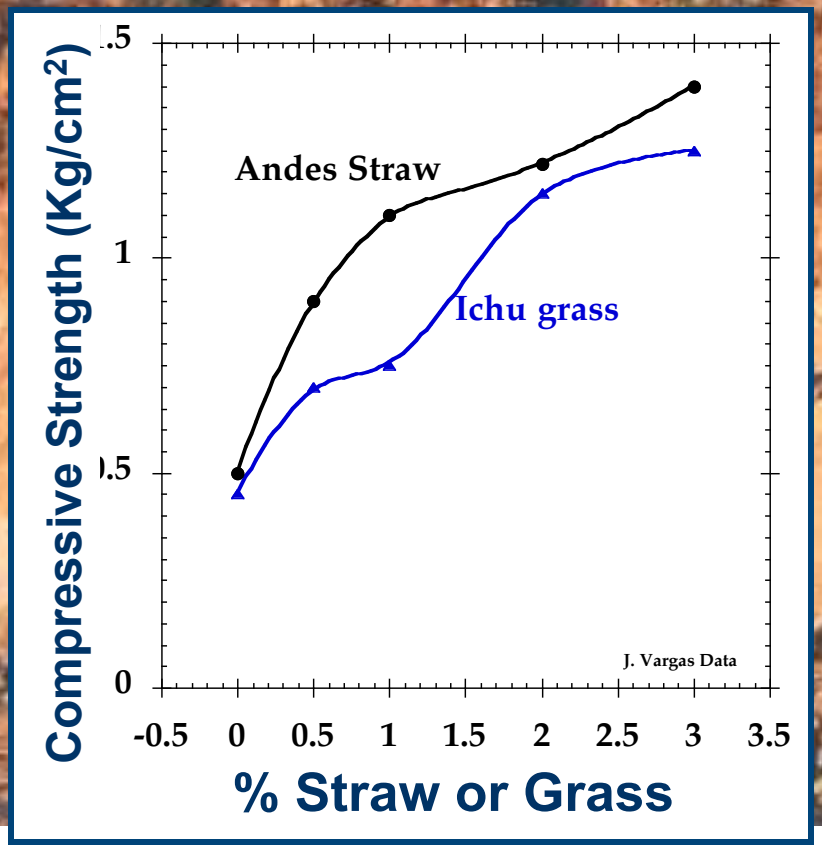


Reinforced Fired Adobe Composite

Inca city ~ 1500 AD

Present Day



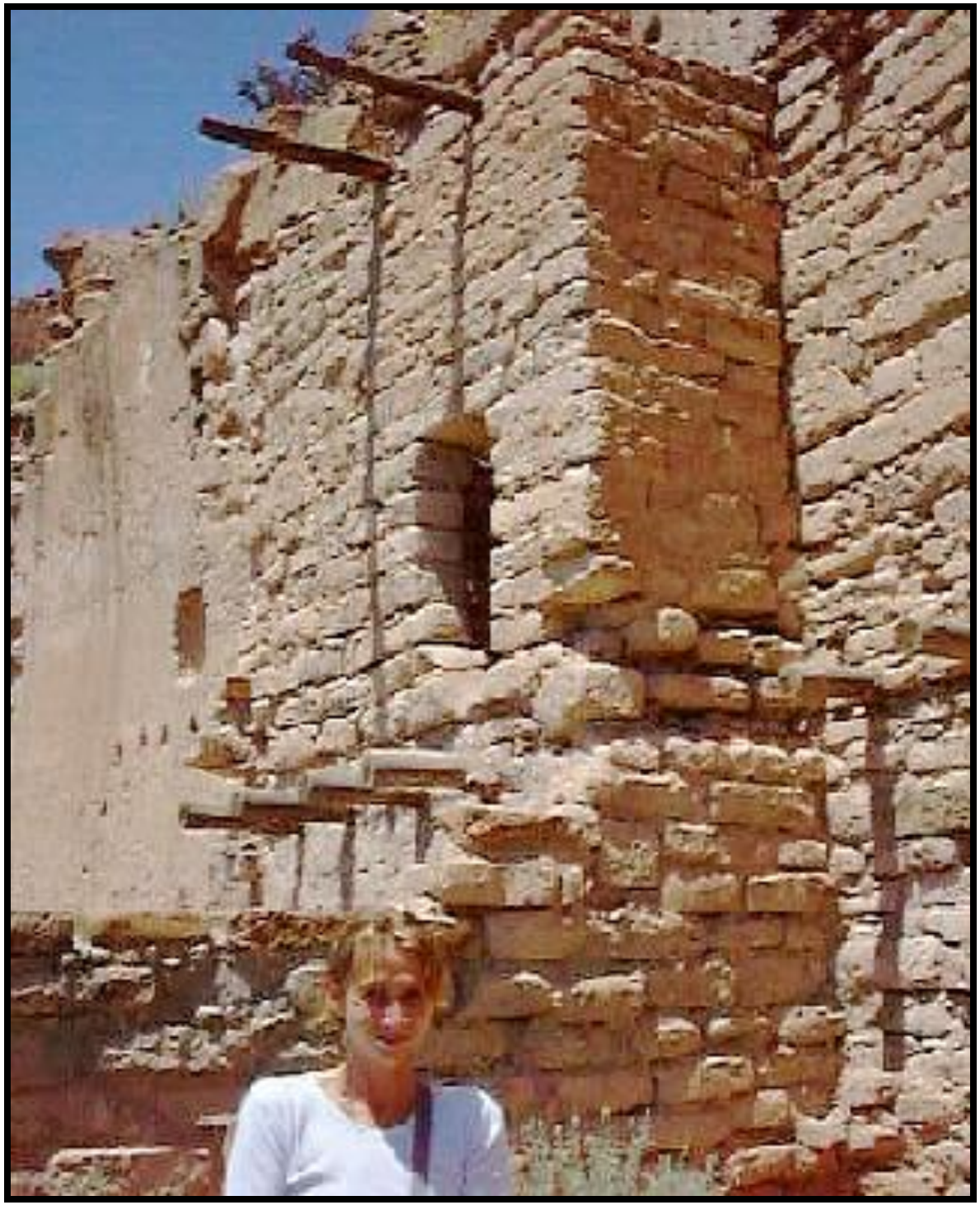


Fort Paramonga Chimu civilization ~1300 AD

Puye Cliff Dwelling

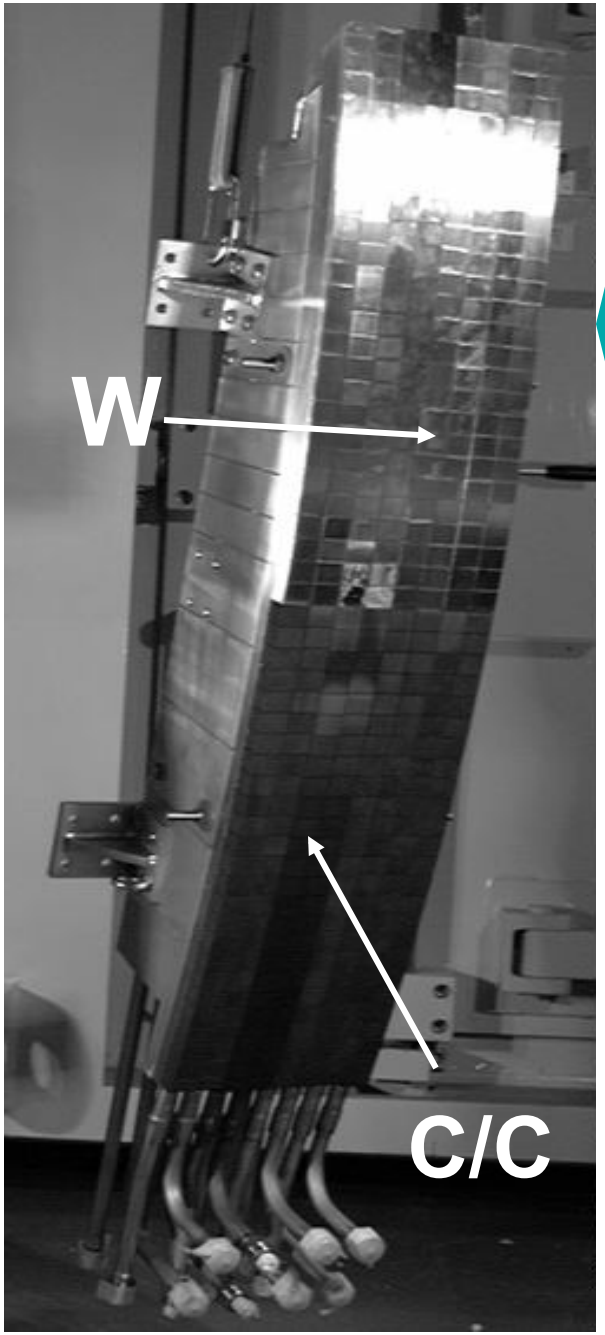
Anasaze Indians

1100-1580 AD



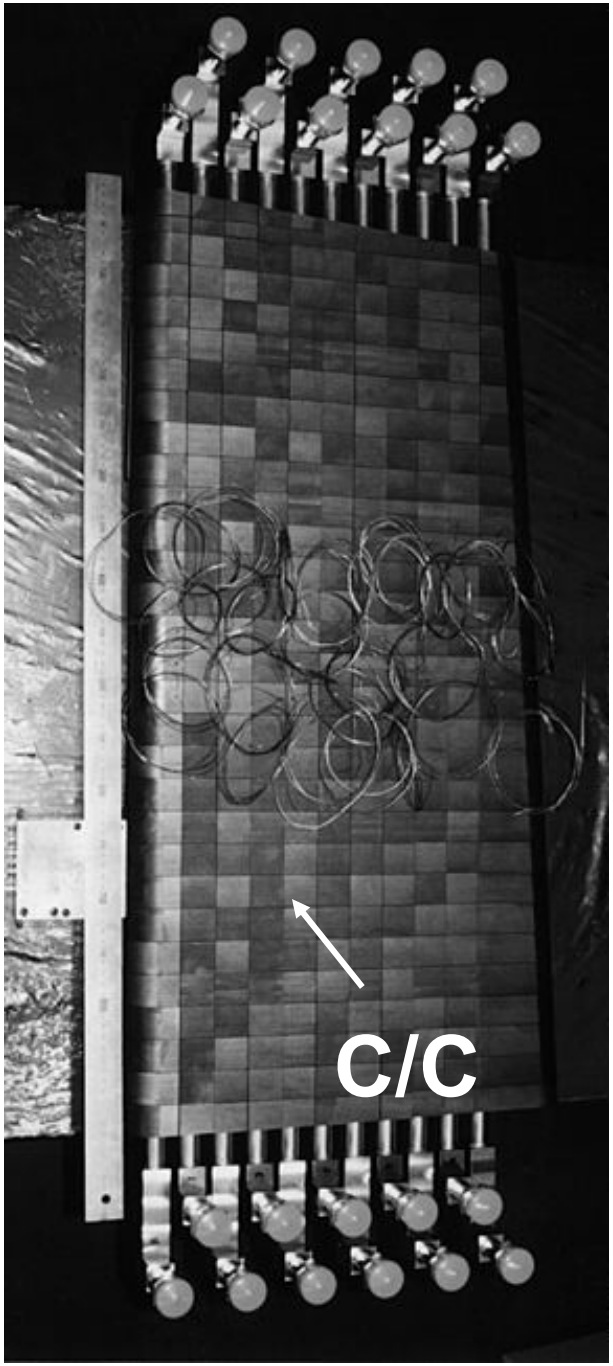
Tel-Dan Arch
~1600 BC

Divertor Mock-Ups Using C/C Composites



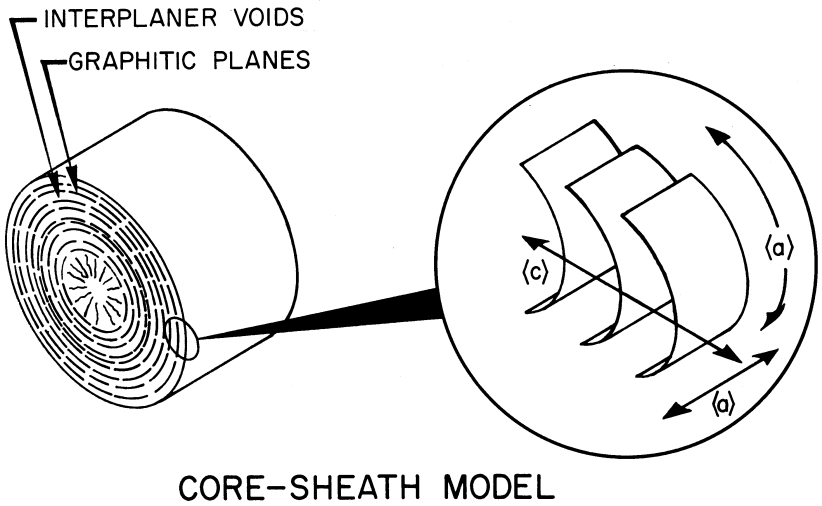
Full-scale vertical target armored mock-up uses a pure Cu clad DS-Cu tube armored with saddle-block C/C and CVD-W armors. (Hitachi Ltd., Japan)

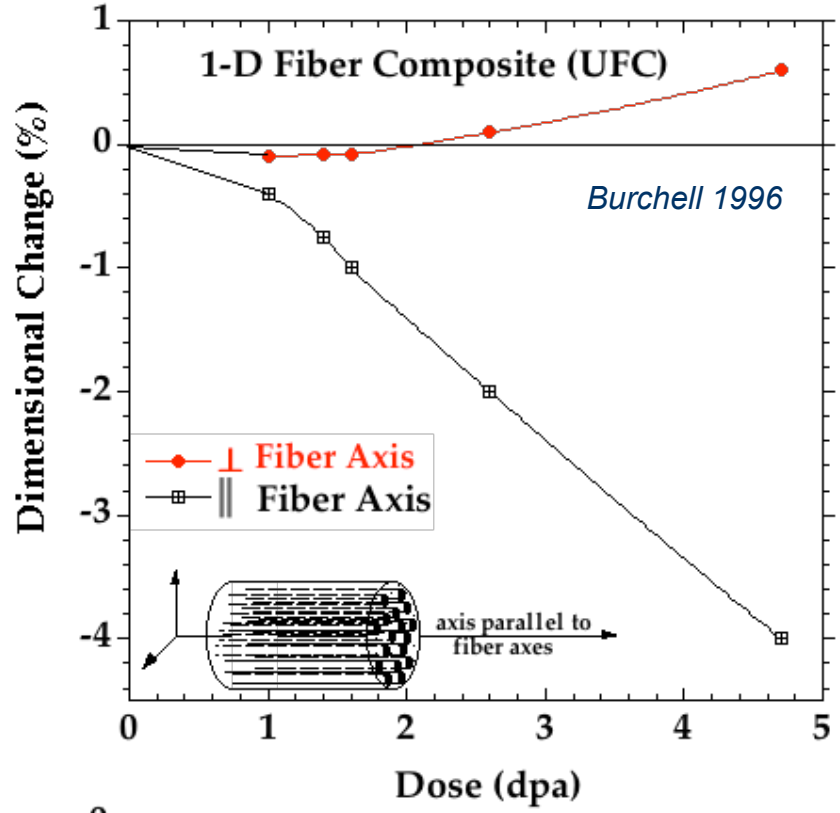
Pure Cu clad DS-Cu tube armored with C/C monoblocks. (Kawasaki Heavy Industries, Japan)

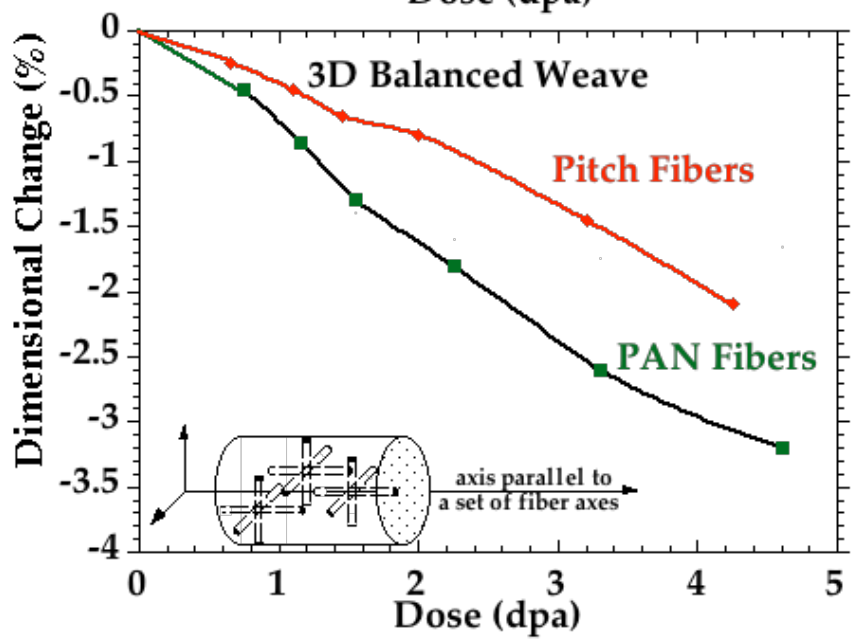


CFC's Under Irradiation

(HFIR , 600°C)



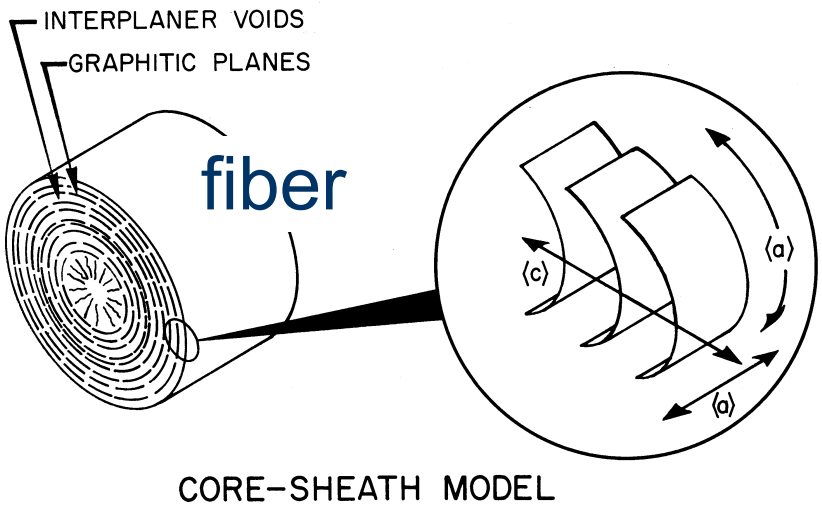


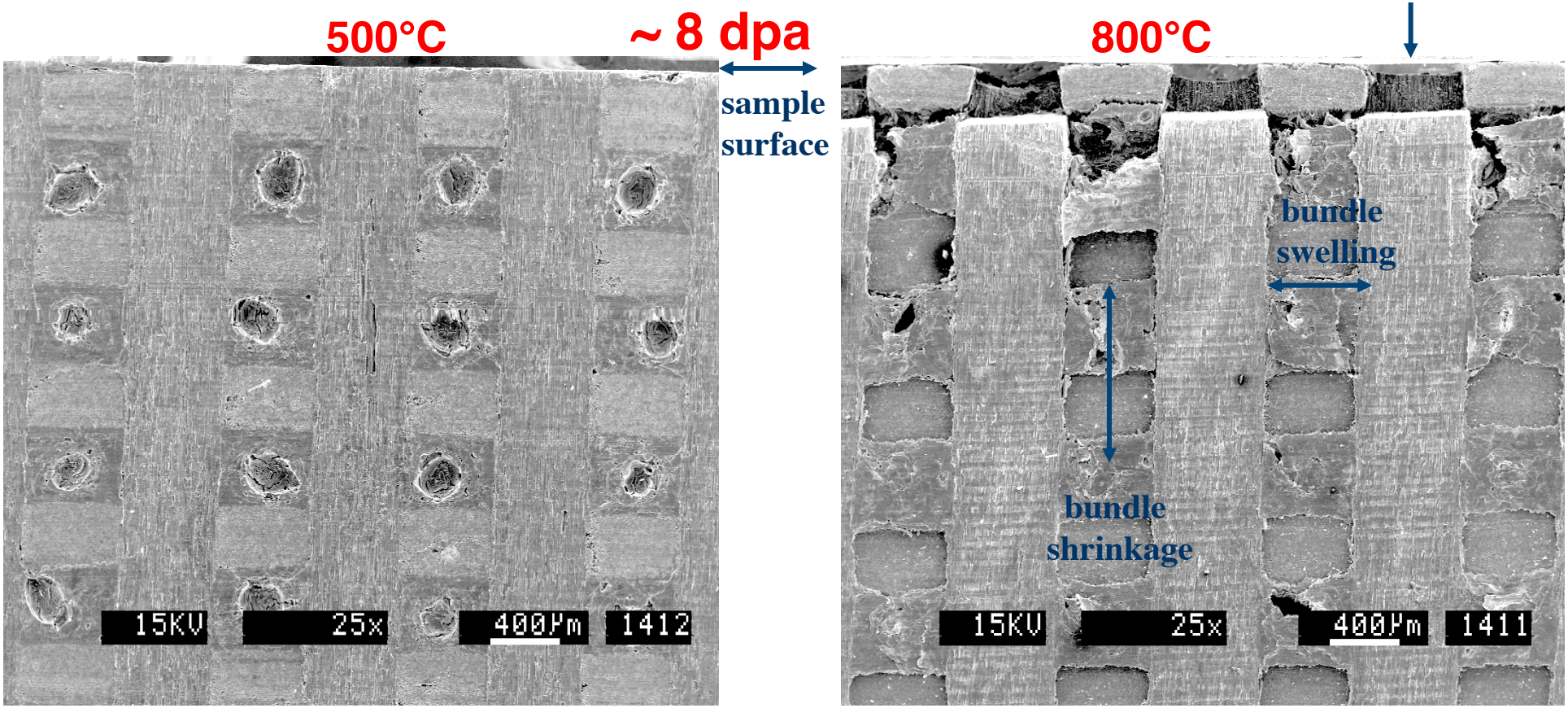


- **composite irradiation-induced dimensional changes explained by classic graphite model…**

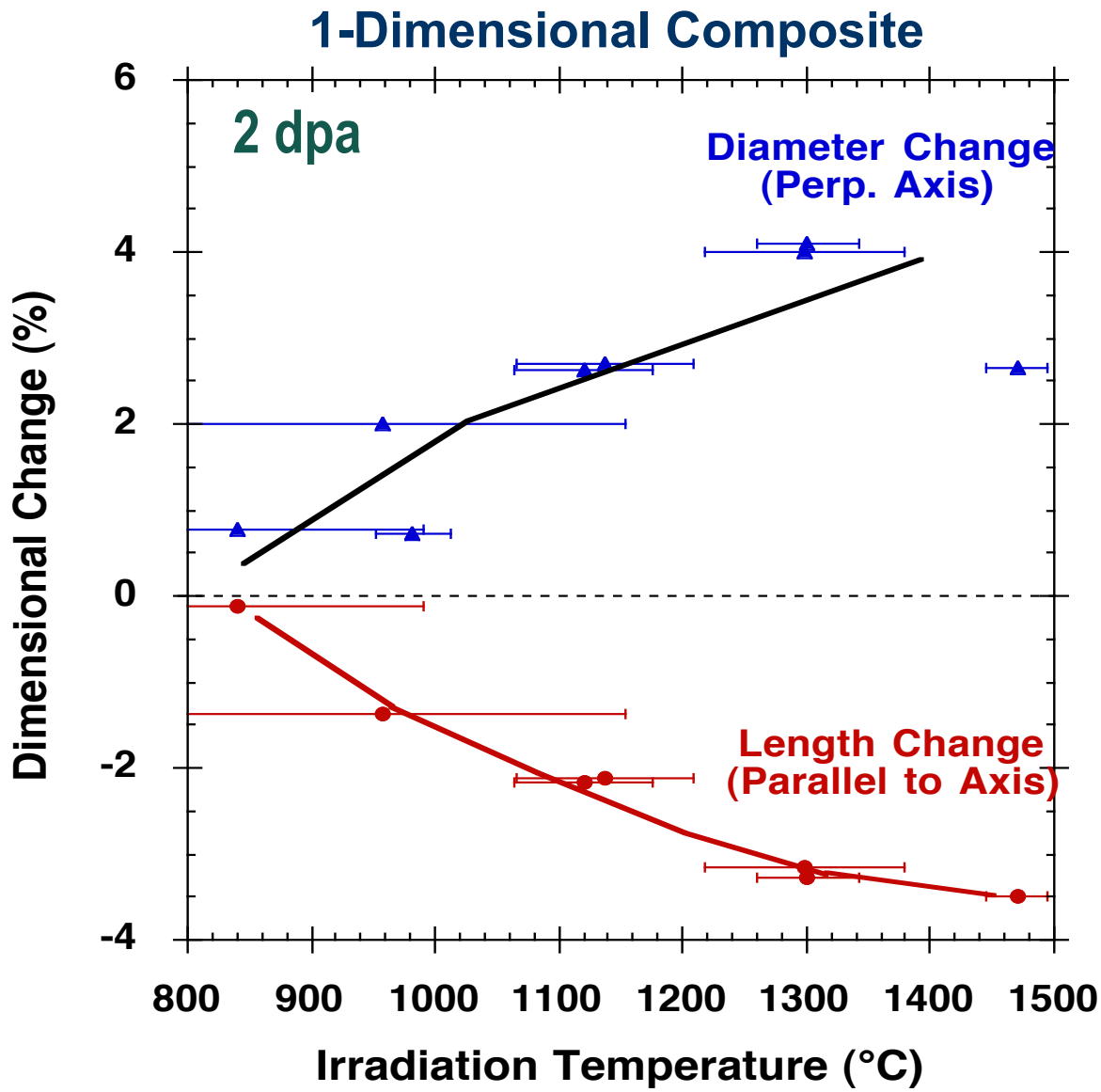
CFC's Under Irradiation

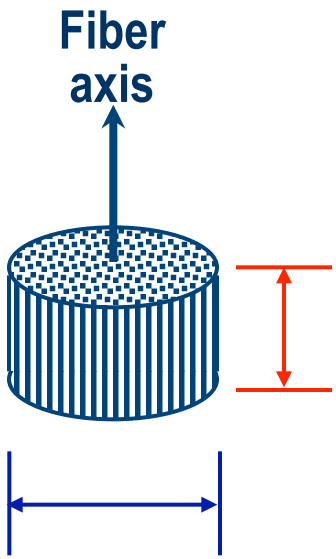
Elevated temperature irradiation begins to show severe degradation



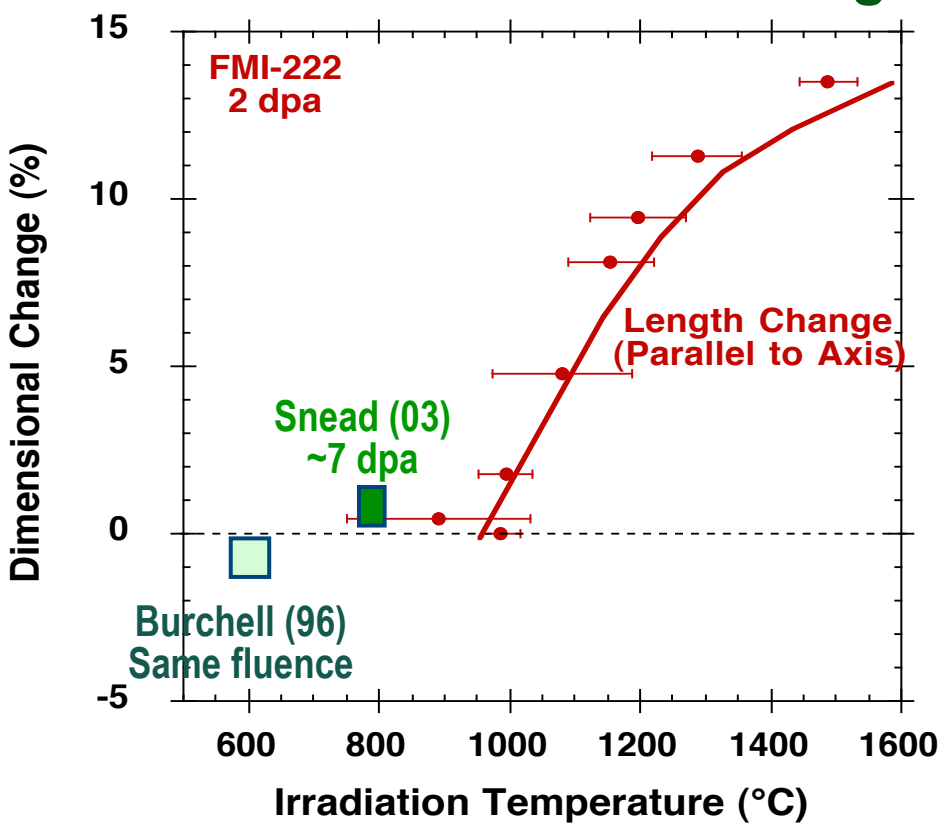


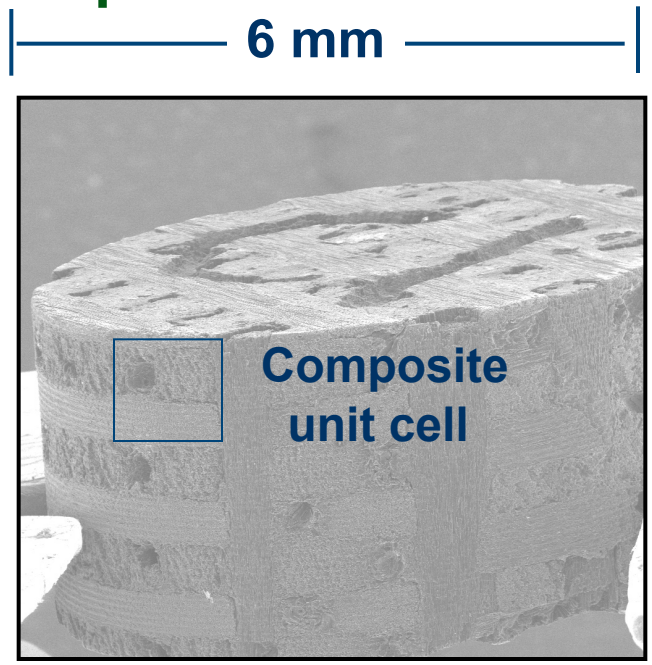
Dimensional Change in 1-D Composite

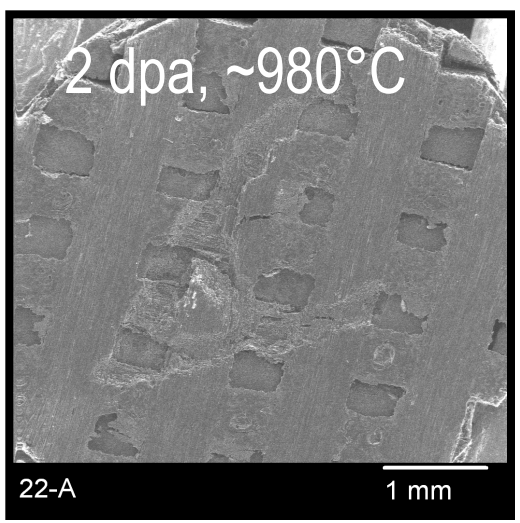


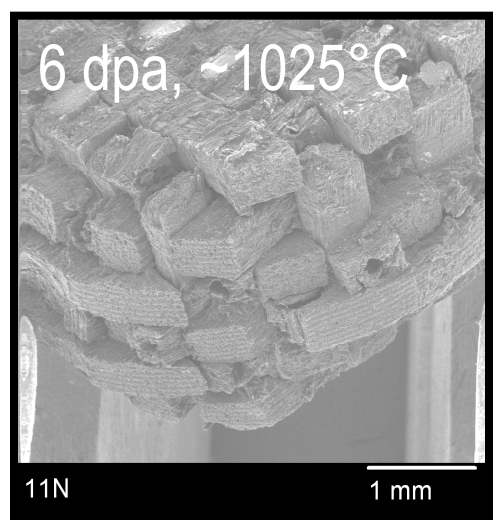


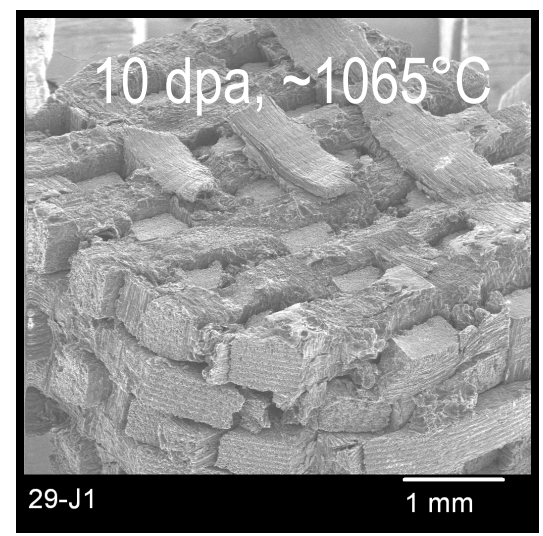
Dimensional Change in CFC Composite



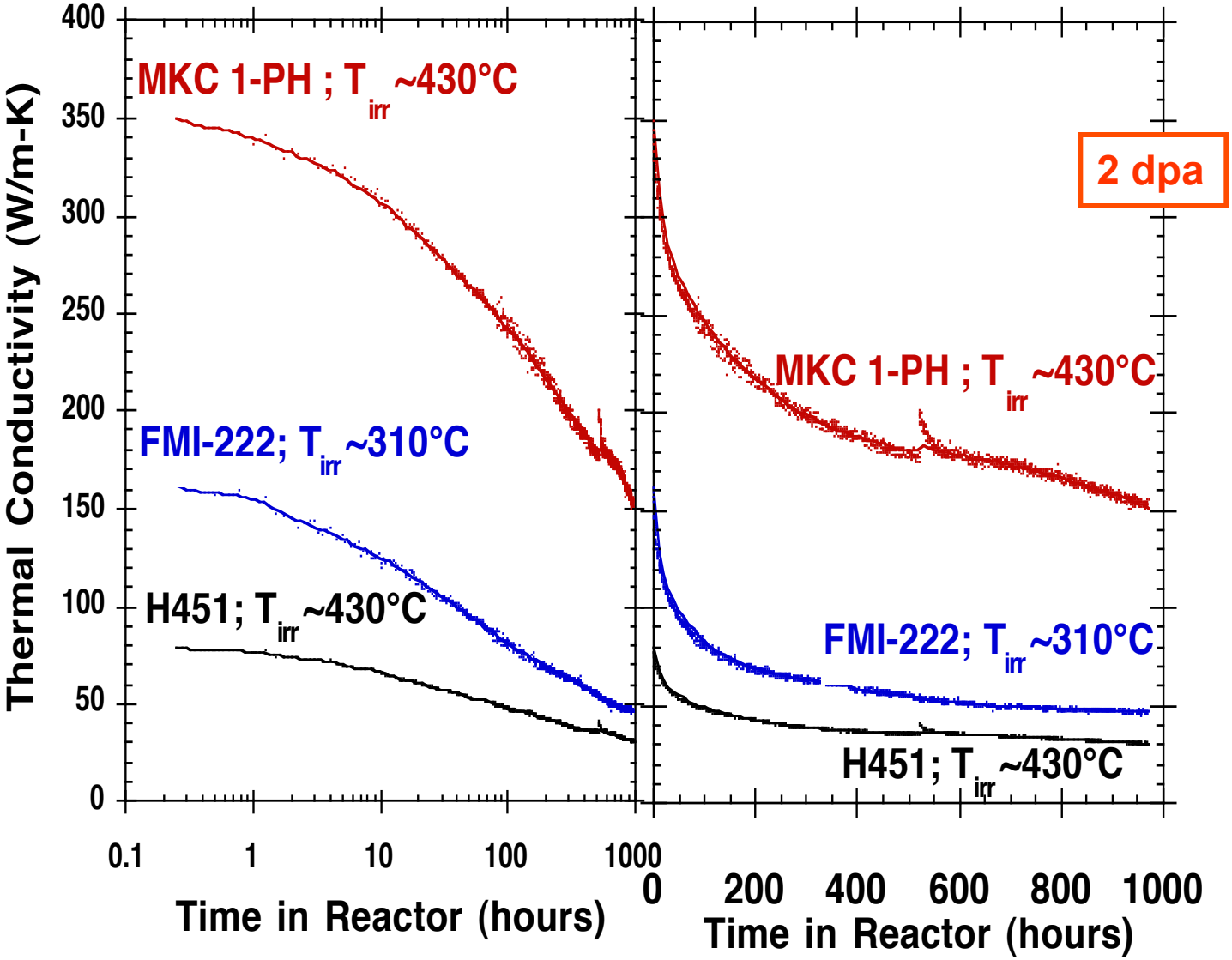








Comparison of Thermal Conductivity Degradation



Biased Summary Comments on Graphite and CFC's

- Graphite, like beryllium (and perhaps tungsten) suffers from "crystal-driven irradiation-induced changes. The effects can be somewhat mitigated, but strict lifetimes exist.

- CFC irradiation damage follows the same principals as for graphite.
  - Swelling perpendicular to basal planes, shrinkage within planes
  - Increase in strength and modulus up to composite lifetime.

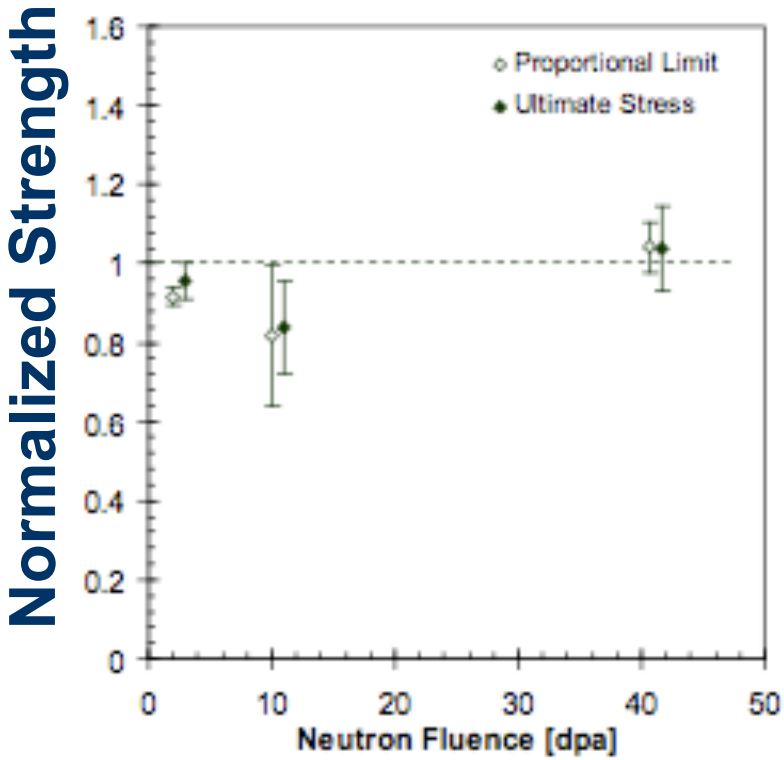
- The lifetime of the composite will depend sensitively on the irradiation temperature and dose. It appears that, due to the inherent perfection of graphite fibers, the lifetime of the composite may be lower than that of nuclear graphite, especially at high temperatures.

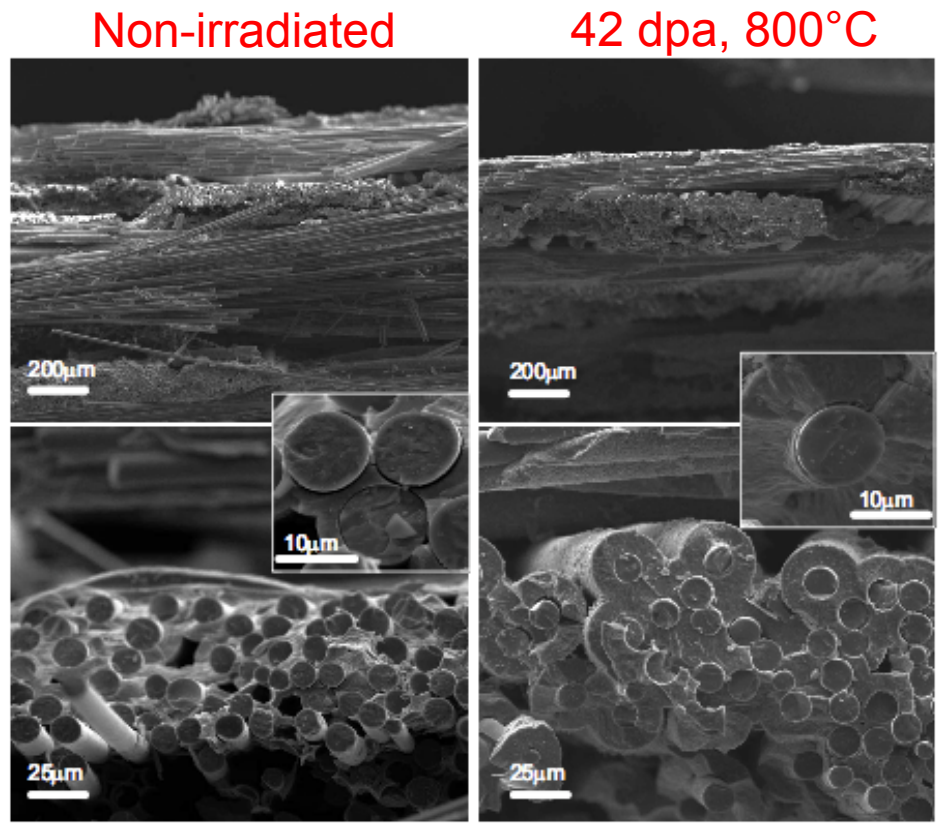
- The CFC materials studied to date have been selected based on high intrinsic perfection (thermal conductivity.) This selection may lead to lower lifetime. --> we may do much better with poorer materials…

Biased Comments on SiC/SiC : A Single Slide

- The SiC crystal does not undergo anisotropic swelling and is remarkably radiation stable. It does not have the tritium retention issues of graphite.

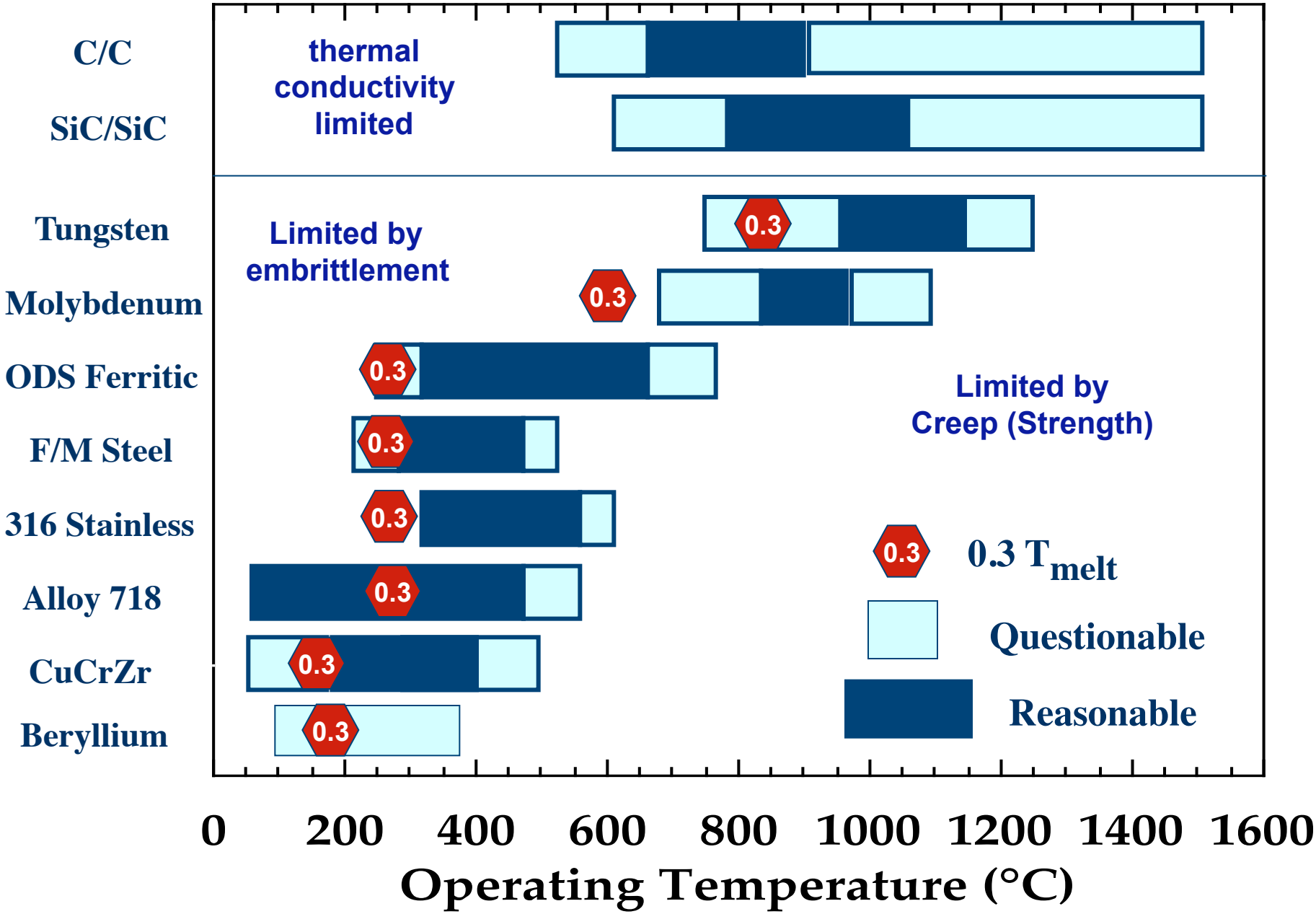
- Current data has shown nuclear grade composite to be mechanically and dimensionally stable to 42 dpa (the highest dose yet achieved.)





- However, thermal conductivity of both non-irradiated (20-30 W/m-K) and irradiated (2-5 W/m-K) will preclude it from first wall application.

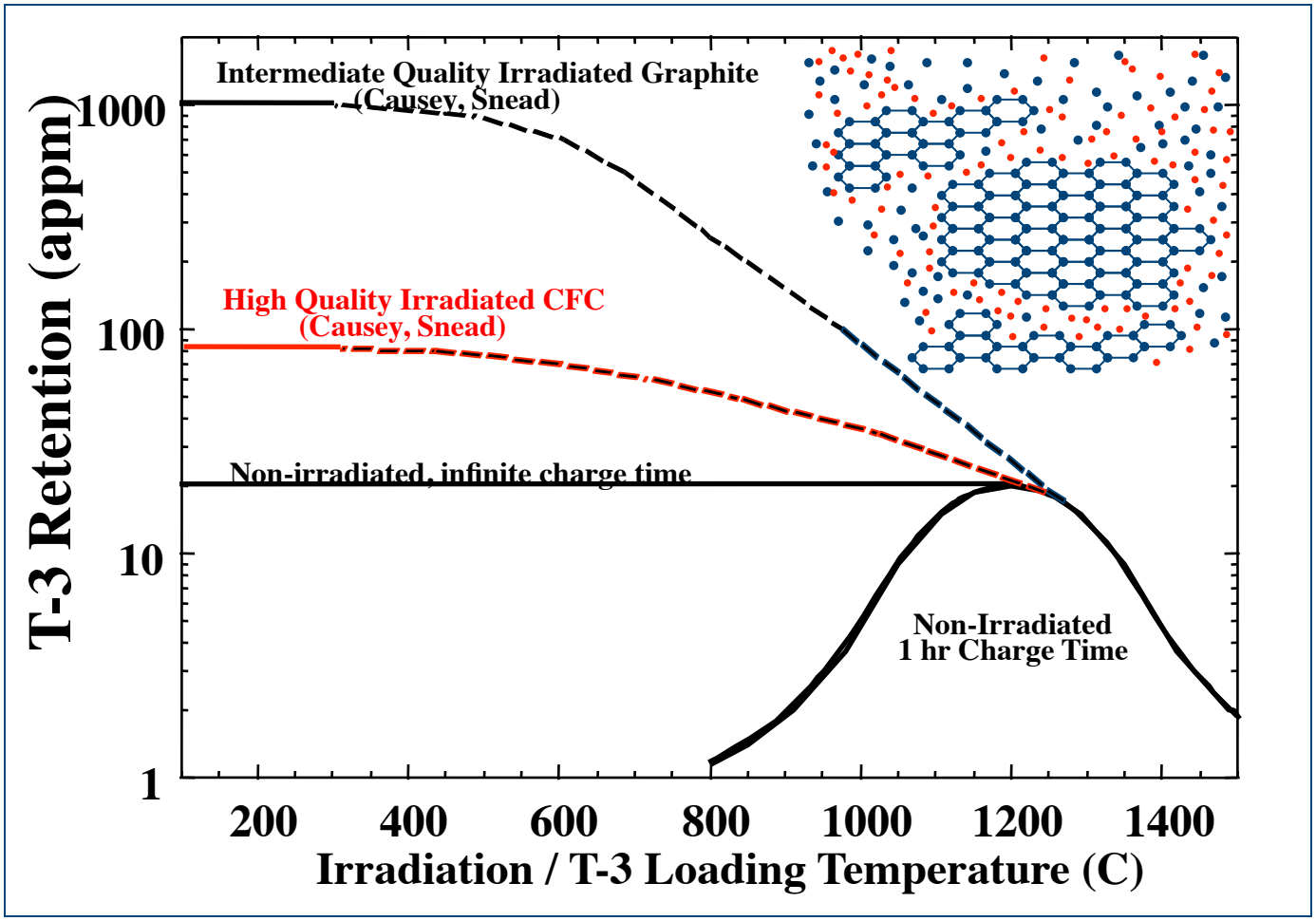
Operating Range, Irradiated Structural Materials



Questions ???

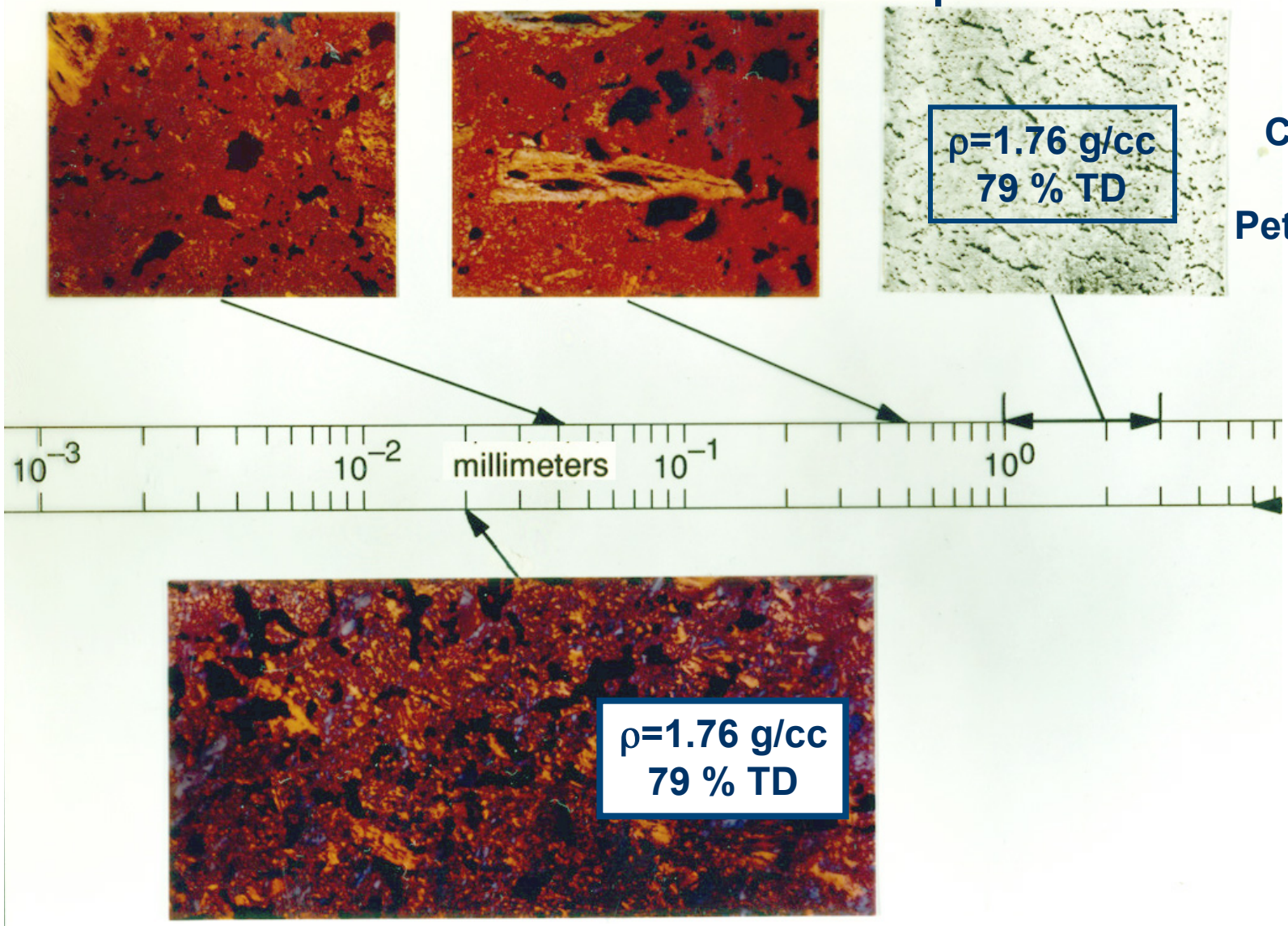
CFC's Under Irradiation : Tritium Retention

- T-3 attaches to basal plane edges and highly defected structure. More perfect material and/or high temperature allows less retention.



Nuclear Graphite

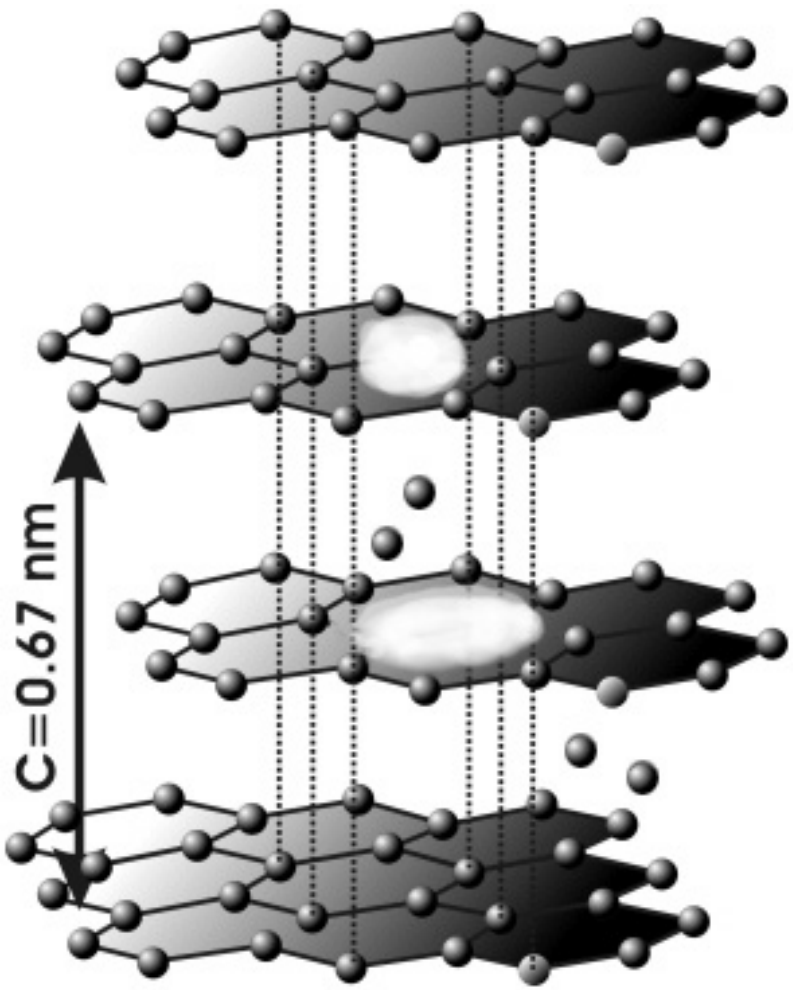
H-451 Extruded Nuclear Grade Graphite

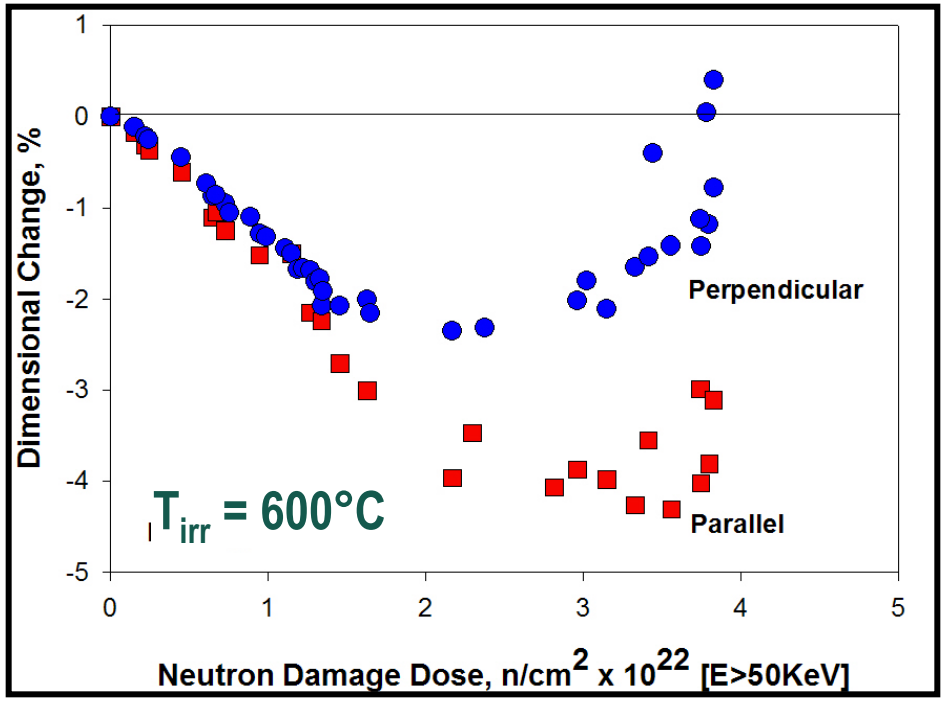


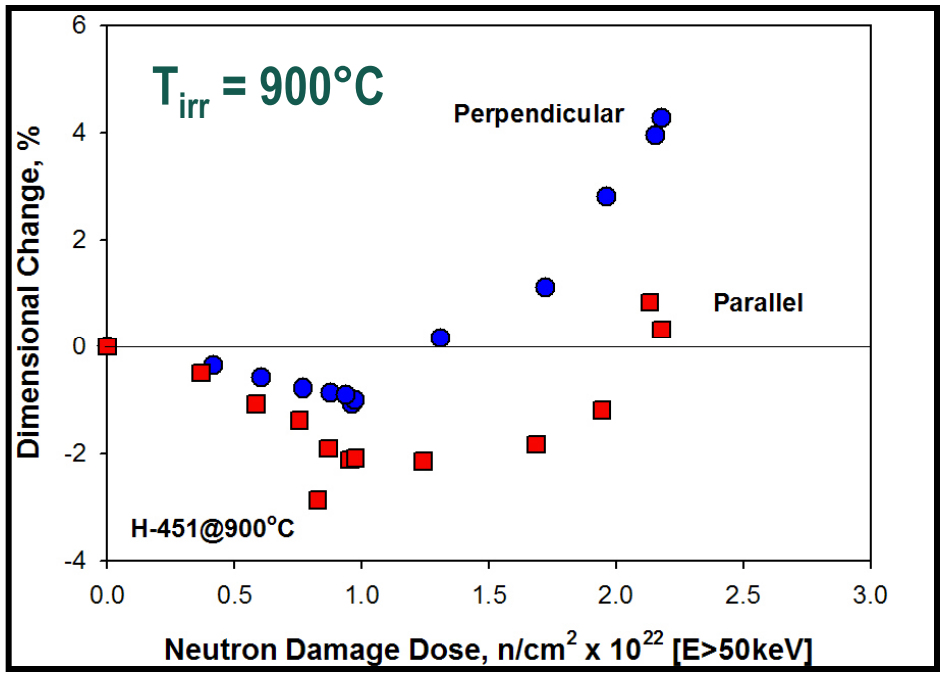
Graphite
Coal-tar pitch
Pitch coke
Petrolium binder

IG-11 Isomolded Nuclear Grade Graphite

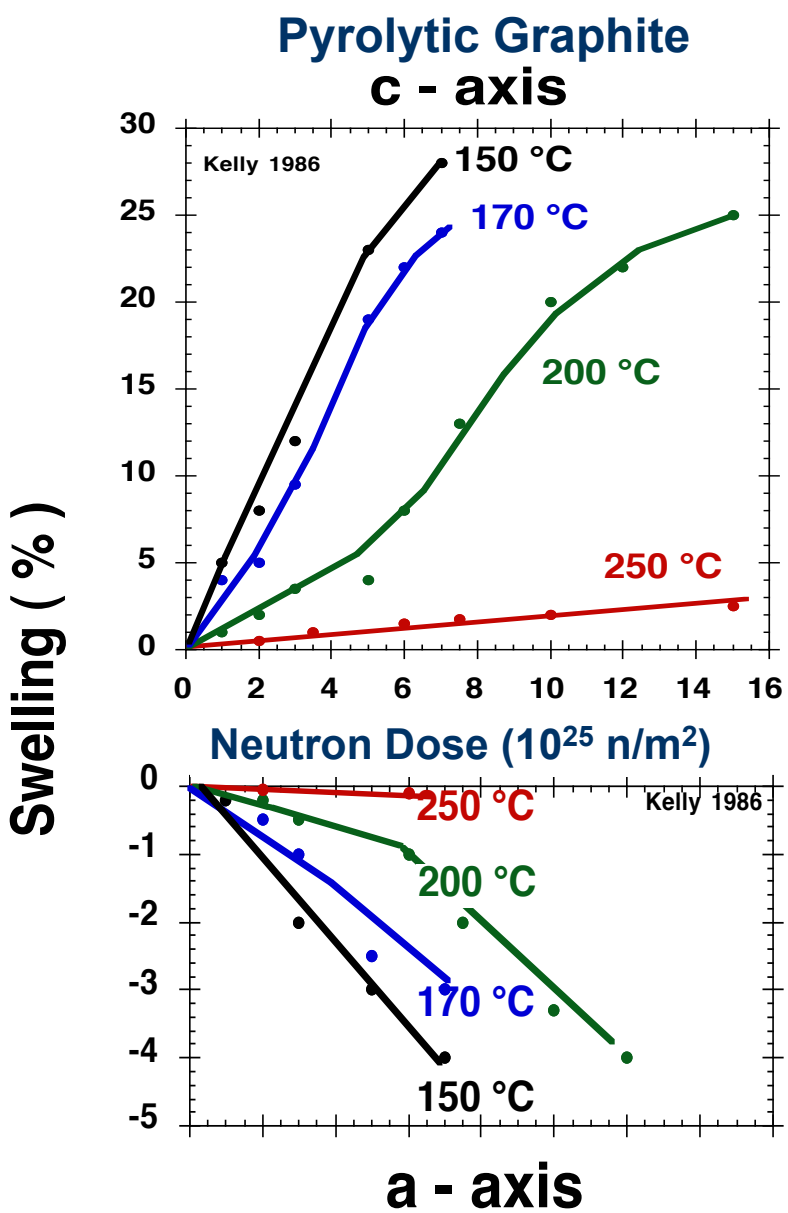
H-451 Nuclear Graphite

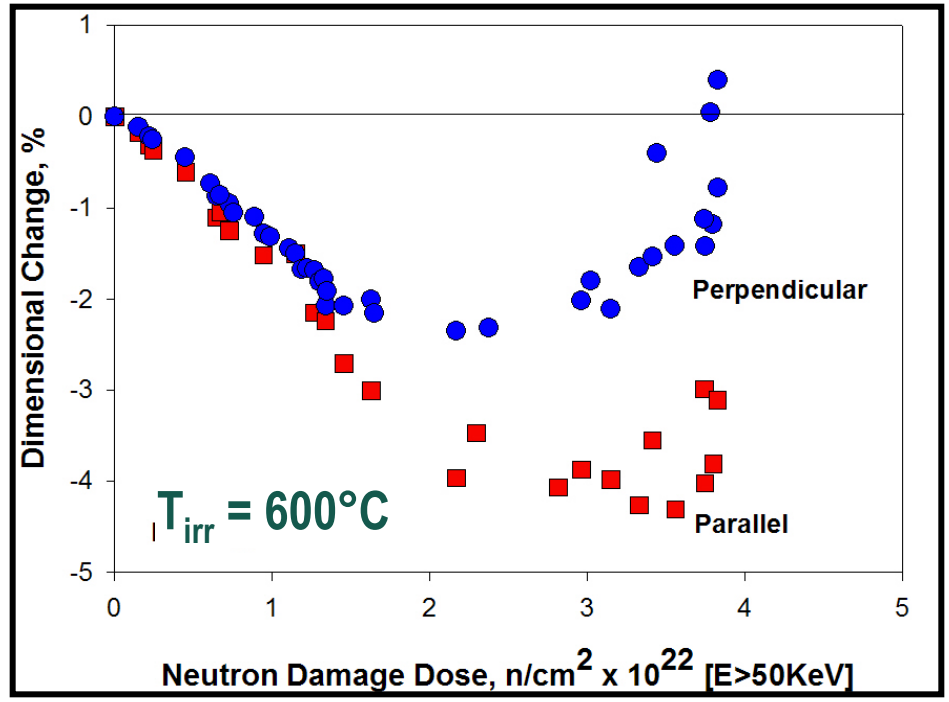


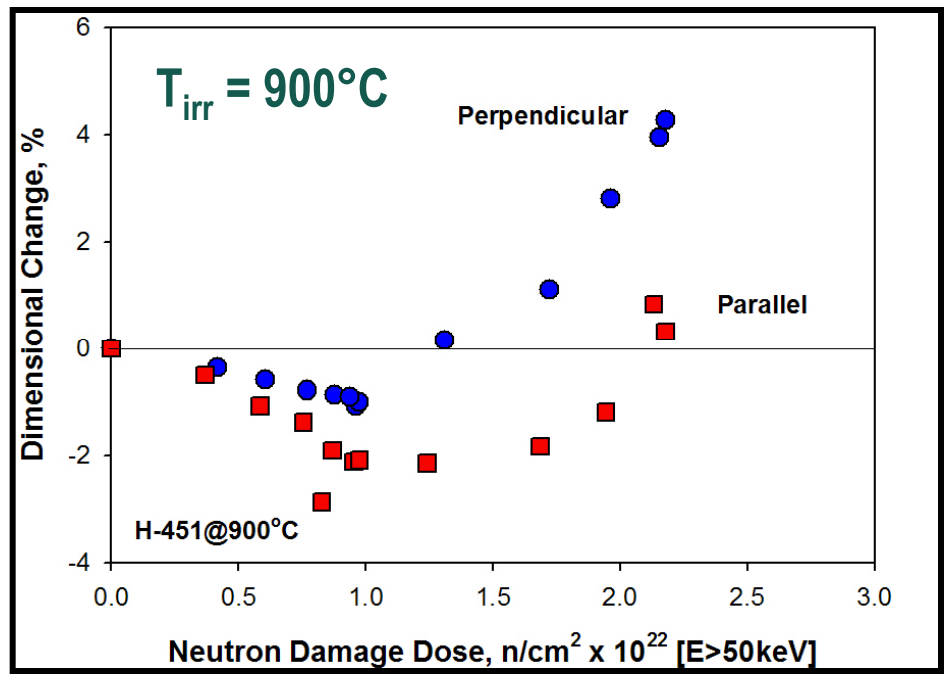




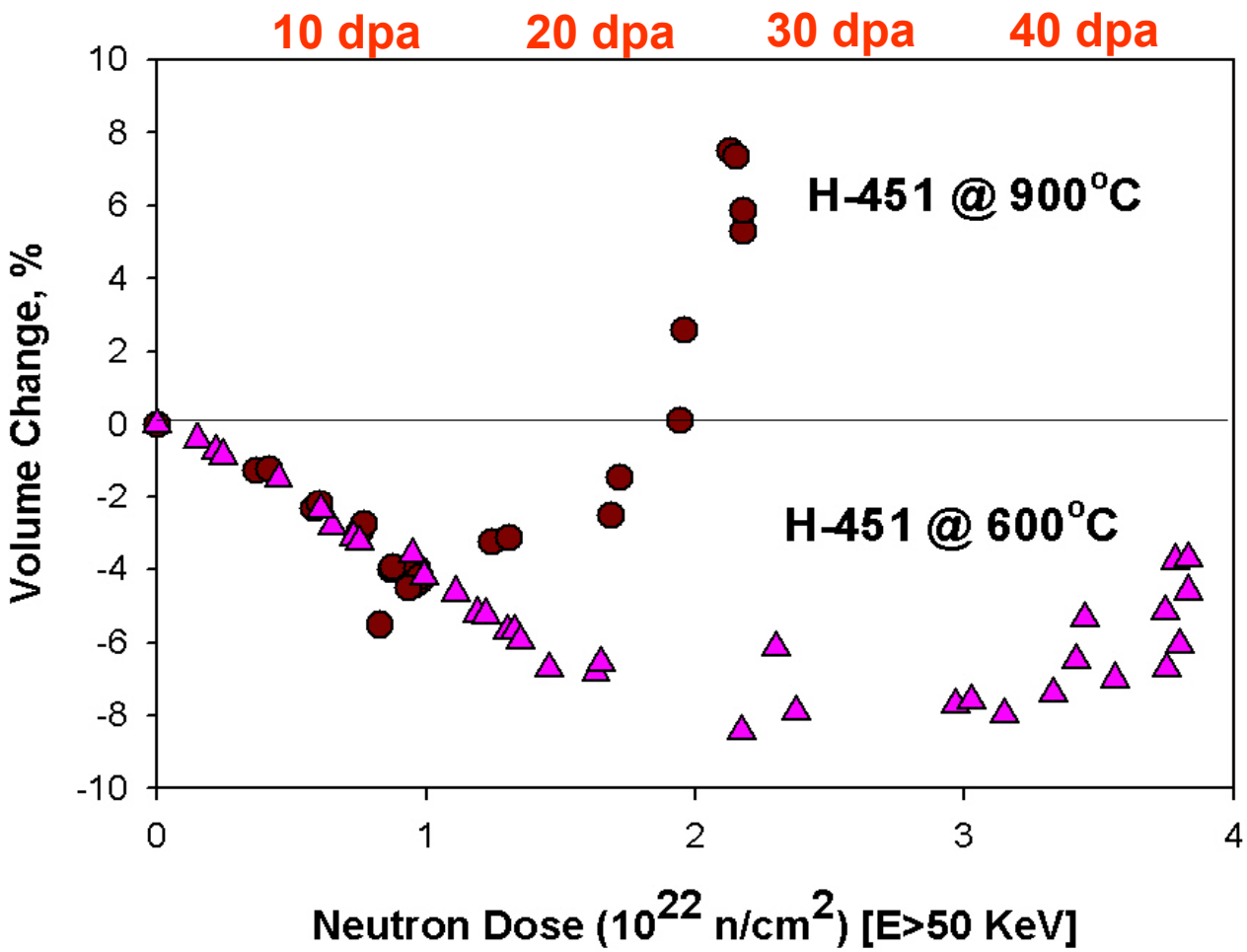
H-451 Nuclear Graphite





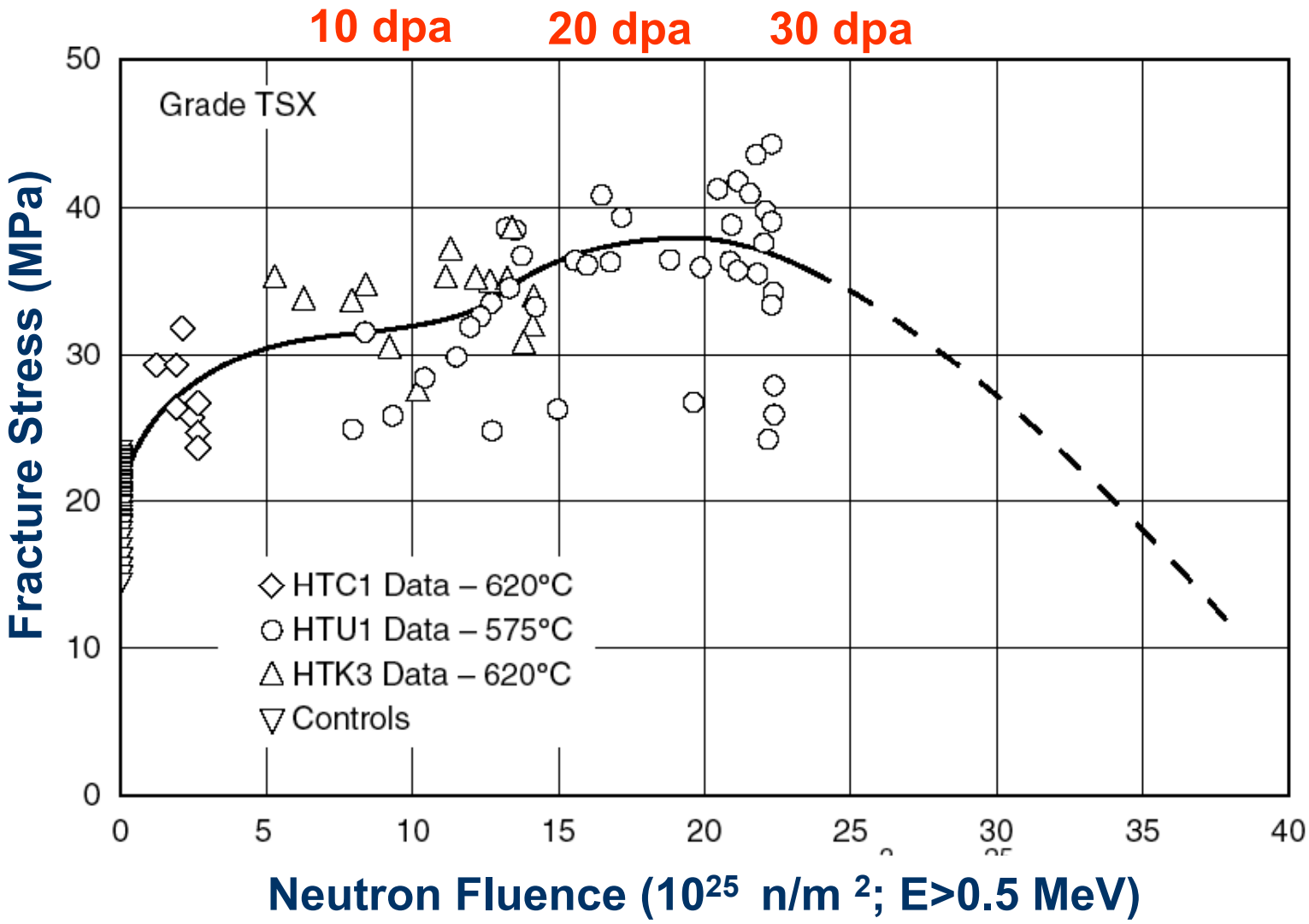


Effect of Temperature and Swelling of Nuclear Graphite

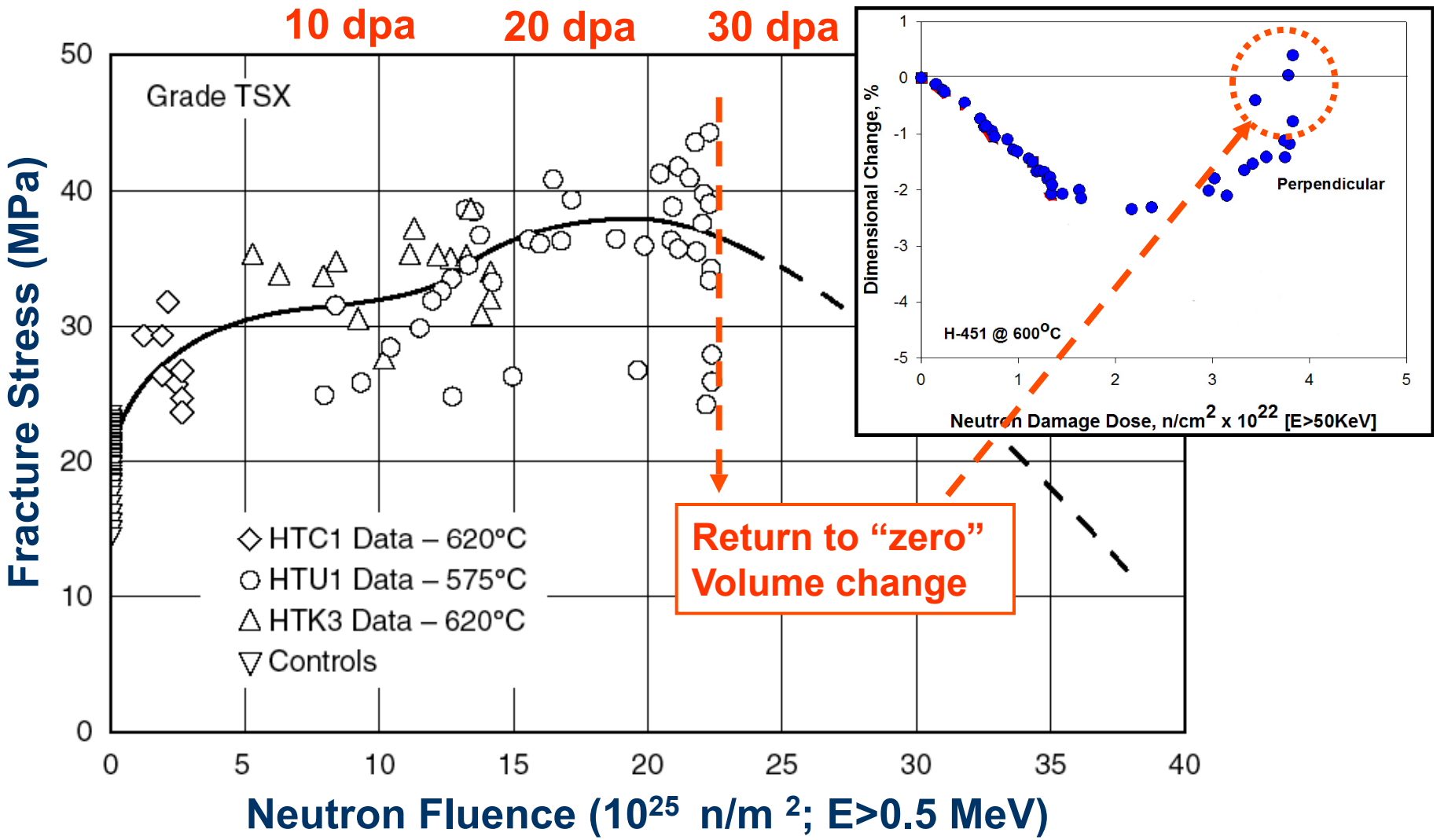
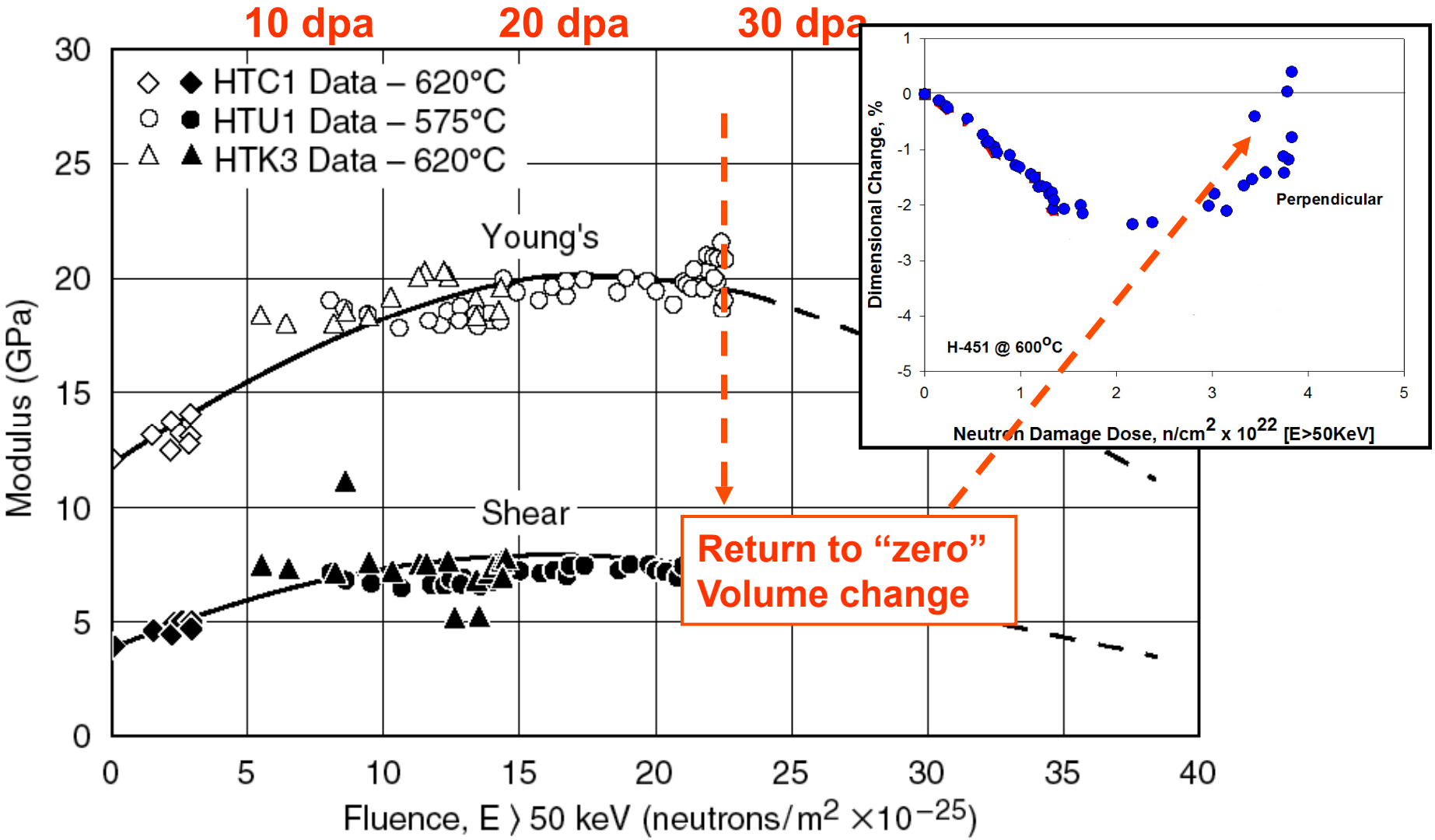


- initial <c> swelling accommodated by closure of intrinsic porosity.
- once porosity filled swelling can begin.
- less initial porosity for higher initial temperature (closure of intrinsic porosity.)

Effect of Irradiation on Strength of Nuclear Graphite



Effect of Irradiation on Strength of Nuclear Graphite



Effect of Irradiation on Elastic Modulus of Nuclear Graphite