Nuclear Graphite - Fission Reactor Brief Outline of Experience and Understanding

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Overview

- Nuclear Graphite – Use, Manufacture, Microstructure
- Irradiation Damage to Crystal Structure
- Radiolytic Oxidation
- Physical Changes – to Polycrystalline Graphite due to Fast Neutron Damage and Radiolytic Oxidation
- Irradiation Creep

Use of Graphite in the Nuclear Industry

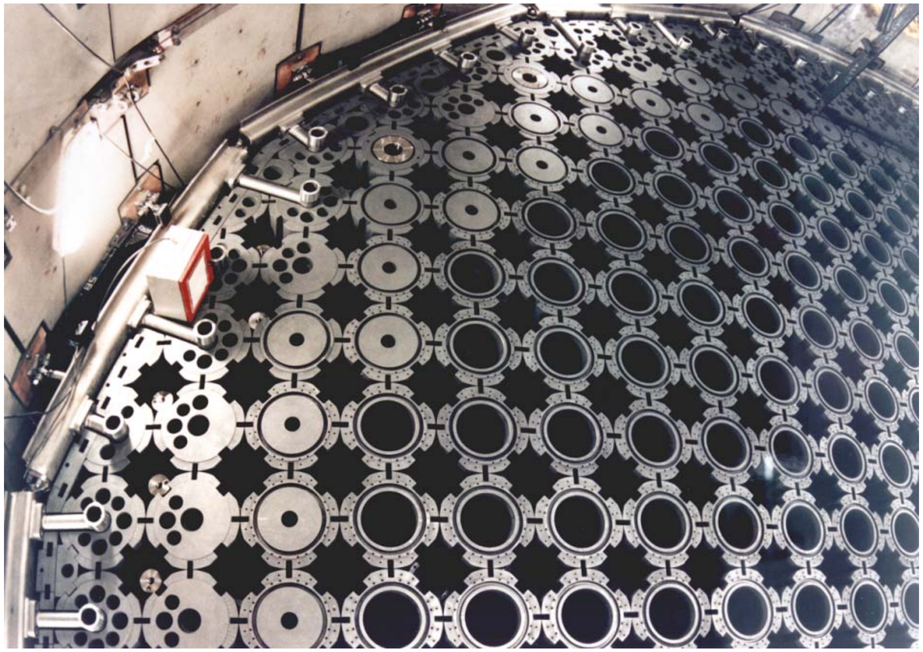
- Moderator
  - Slow down neutrons by scattering
  - High scatter cross-section
  - Low absorption cross-section
- Reflector
  - Reflects neutrons back into the core
  - Protect surrounding supports structure and pressure vessel
- Major Structural Component
  - Provided channels for control rods and coolant gas
- Neutron Shield
  - Boronated graphite
- Thermal columns in research reactors
- Moulds for casting uranium fuel

Type of Graphite Moderated Reactors

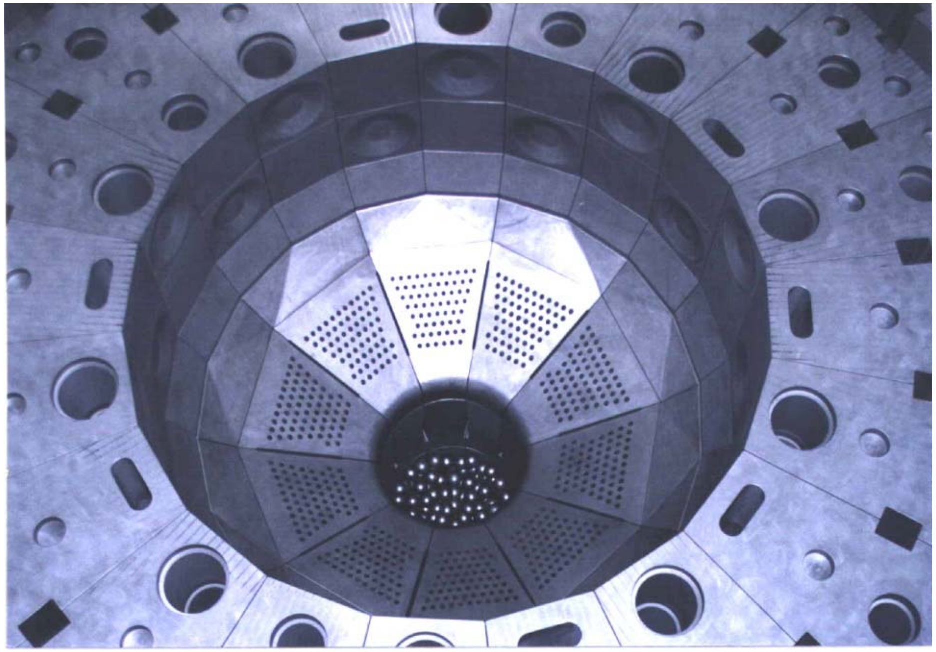
- Air-cooled
    - Chicago Pile, GLEEP, BEPO, Windscale Piles, G1-France
- Light Water-cooled Graphite Moderated
    - Hanford, Russian-PPR, RBMK
- Carbon Dioxide Cooled
    - UK and French Magnox reactors, AGR
- Helium Cooled
    - Dragon, Peach Bottom, Fort St. Vrain, THTR, AVR
    - HTR, HTR-10 China, HTTR Japan, PBMR South Africa
    - Generation IV - VHTR

Chicago Pile 1

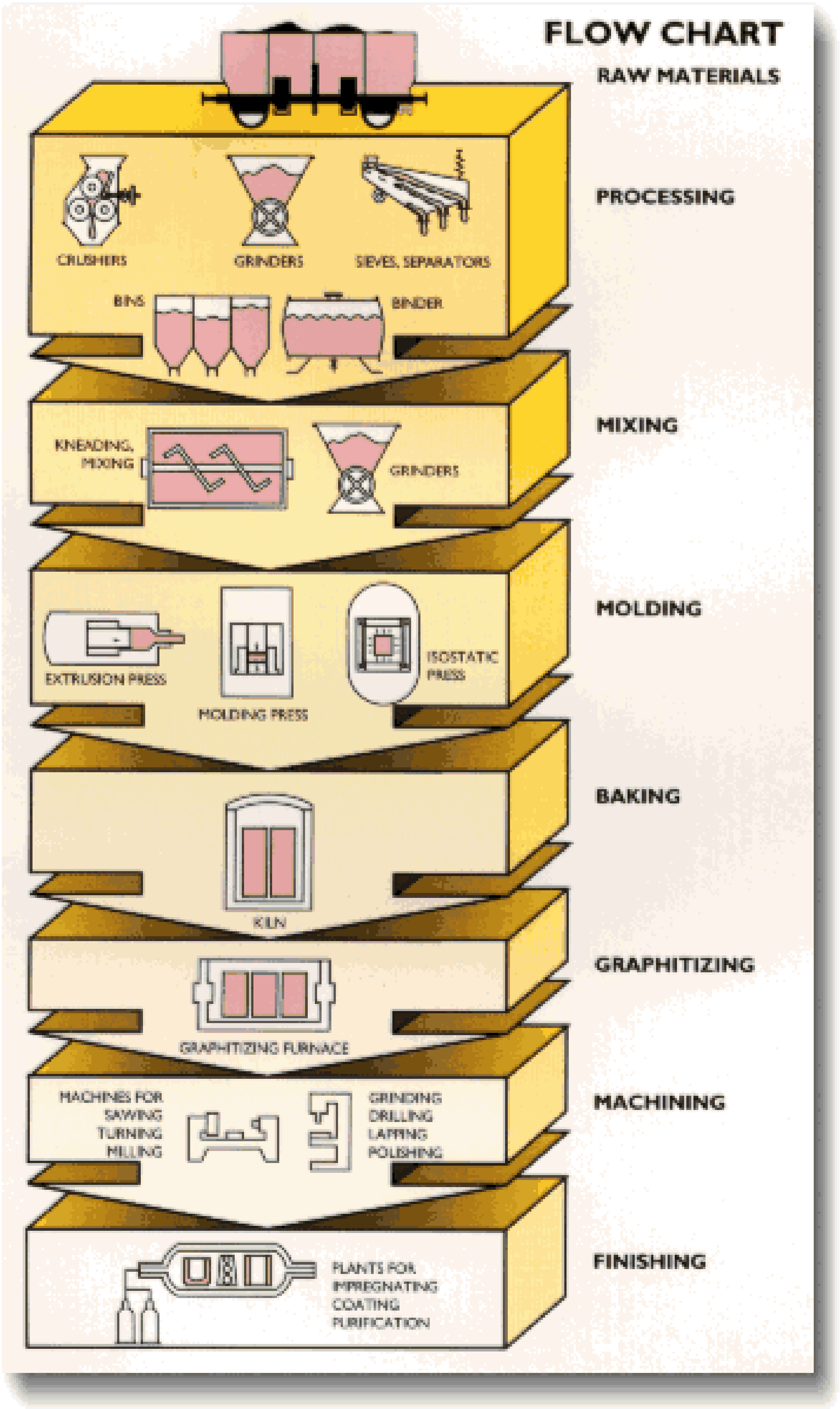
Typical Graphite Components

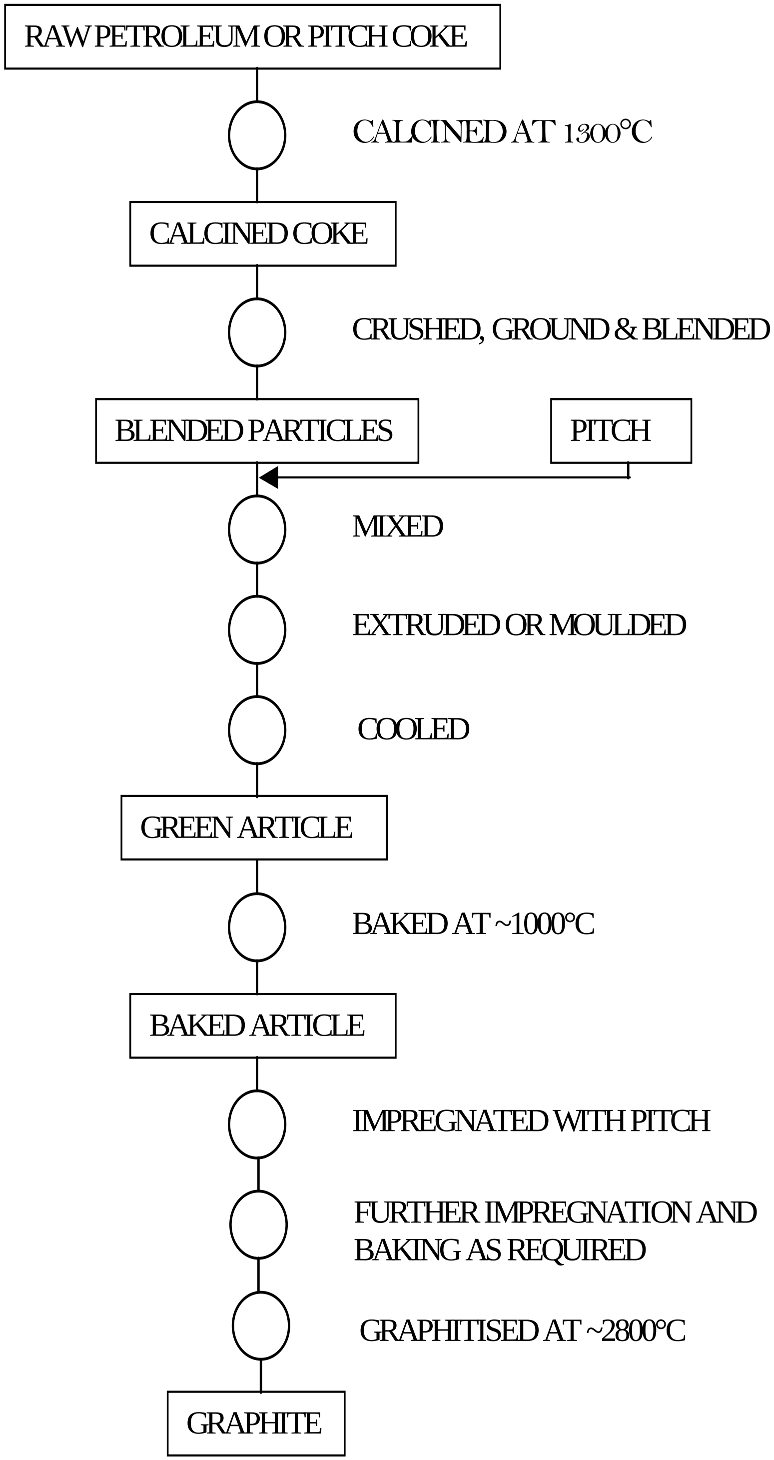


Torness Core – During Construction



HTR-10 During Construction in China





RAW PETROLEUM OR PITCH COKE

CALCINED AT 1300°C

CALCINED COKE

CRUSHED, GROUND & BLENDED

BLENDED PARTICLES

PITCH

MIXED

EXTRUDED OR MOULDED

COOLED

GREEN ARTICLE

BAKED AT ~1000°C

BAKED ARTICLE

IMPREGNATED WITH PITCH

FURTHER IMPREGNATION AND BAKING AS REQUIRED

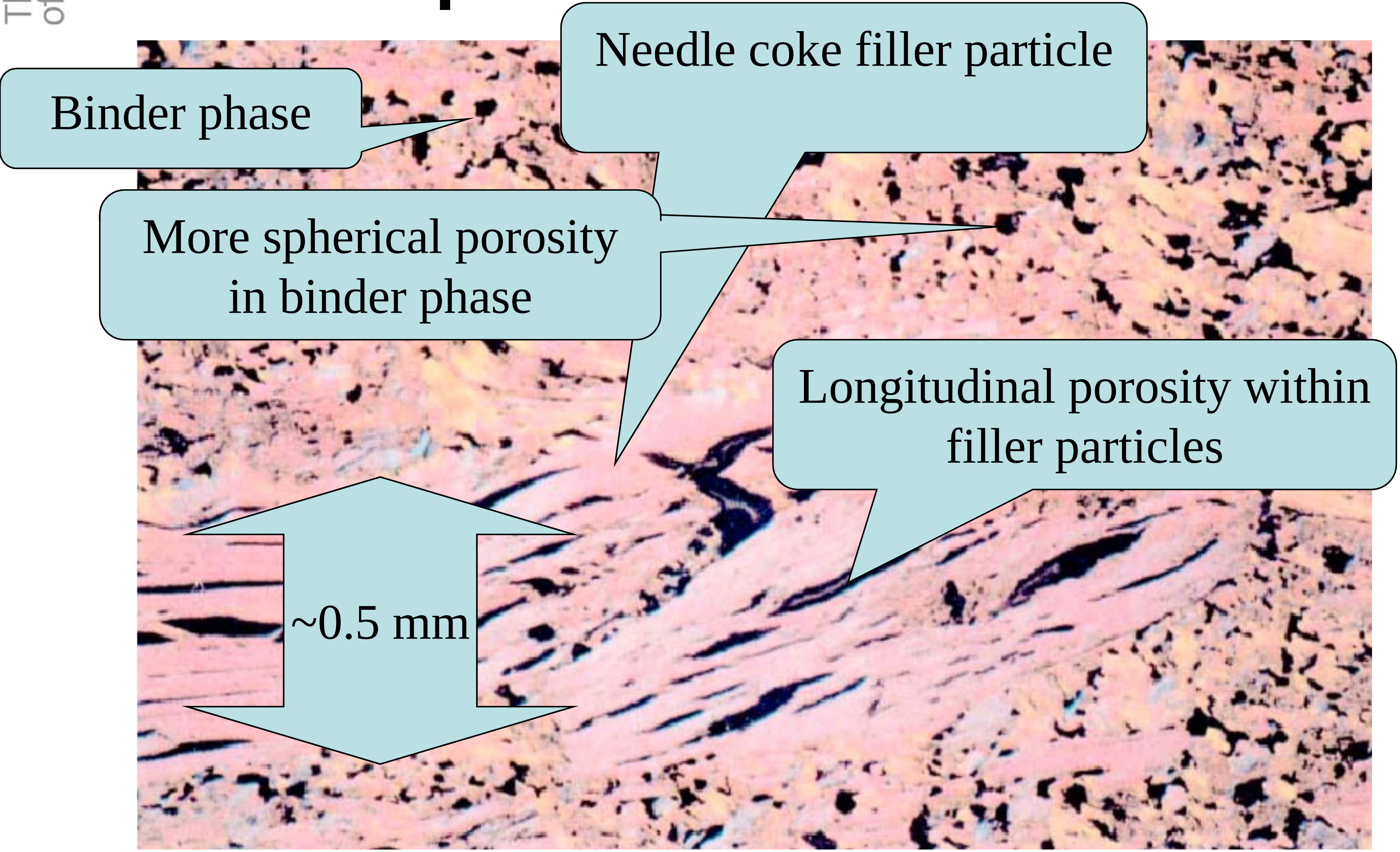
GRAPHITISED AT ~2800°C

GRAPHITE

Final Product

- Either anisotropic or semi-isotropic product
  - Modern reactors use graphite with semi-isotropic properties
- Significant porosity ~20%
  - ~10% open porosity, ~10% closed porosity
  - Density 1.72 – 1.8g/cm^3 compared to 2.26g/cm^3 for perfect graphite crystal
- High purity – impurities measured in parts per million (ppm)
- Nuclear designer requires
  - Semi-isotropic 1.1 Defined by Coefficient of Thermal expansion (CTE) in orthogonal directions
  - High density
  - Optimum material properties
  - High thermal conductivity
  - High purity (neutronic and waste point of view)
  - Dimensional stability under irradiation, associated with high CTE ~4 x 10^{-6} K^{-1} (20-120^{o}C)

Pile Grade A Microstructure – Anisotropic



Needle coke filler particle

Binder phase

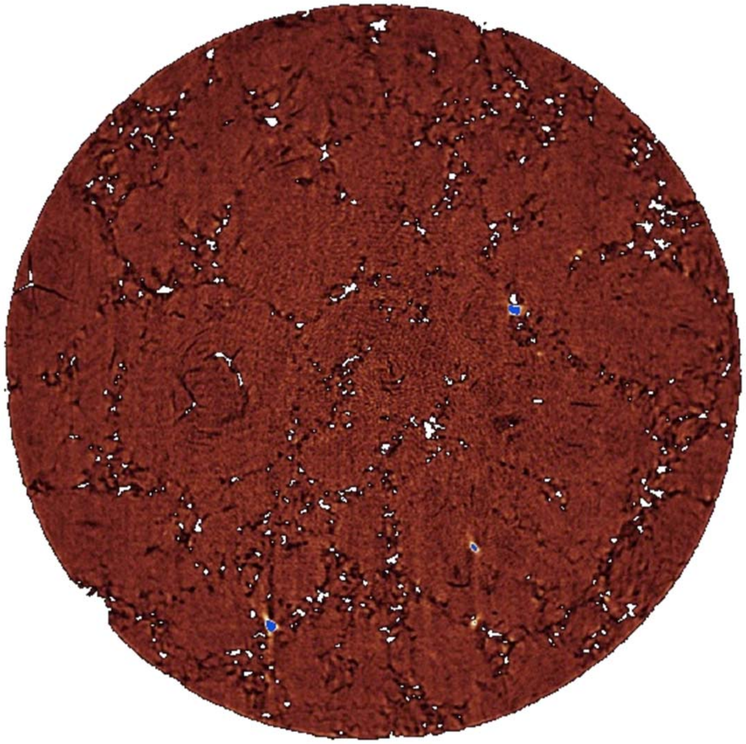
More spherical porosity in binder phase

Longitudinal porosity within filler particles

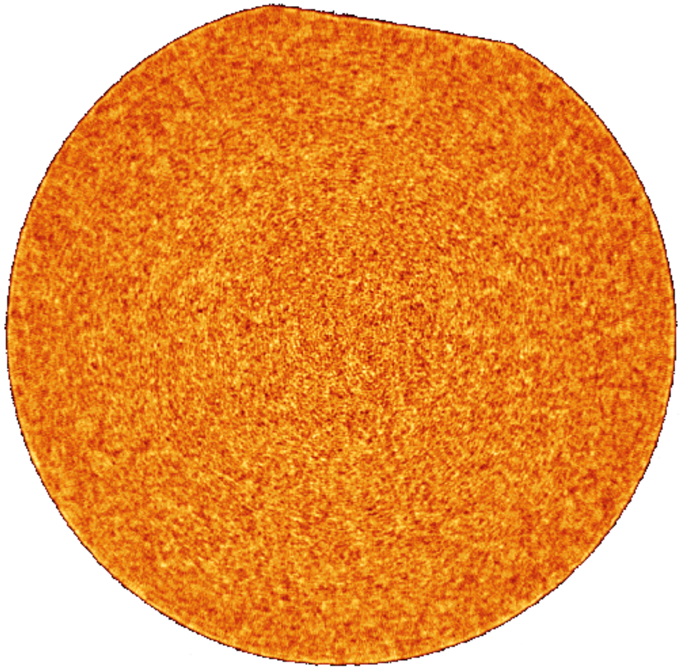
~0.5 mm

Grade	ZXF-5Q	AXF-5Q	Gilsocarbon	IG-430	IG-110
Comment	candidate	similar to AXF-8Q1 (US historical experience)	UK AGR experience	Japanese & EU experience	Japanese & EU experience
Particle size (µm)	1	5	500	10	20
Pore size (µm)	0.3	0.8	42	-	16
Density (g/cm^3)	1.78	1.78	1.81	1.82	1.77
Comp. strength (MN/m^2)	175	138	70	97	79
Flex. strength (MN/m^2)	112	86	23	52	40
Tensile strength (MN/m^2)	79	62	18	38	27
Modulus (GN/m^2)	14.5	11.0	10.8	10.8	9.7
CTE (10^{-6} K^{-1})	8.1	7.9	4.3	4.5	4.0
Thermal conduct. (W/m K)	70	95	131	143	135

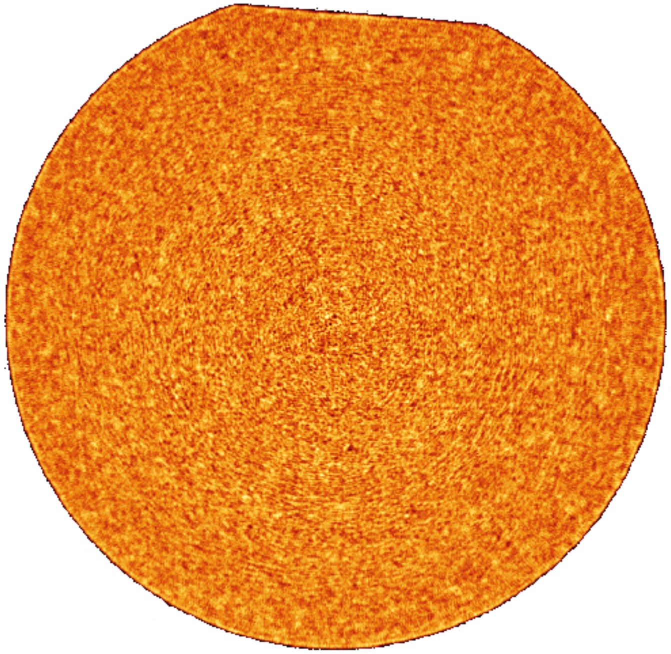
Computed X-ray tomography images of various grades of graphite



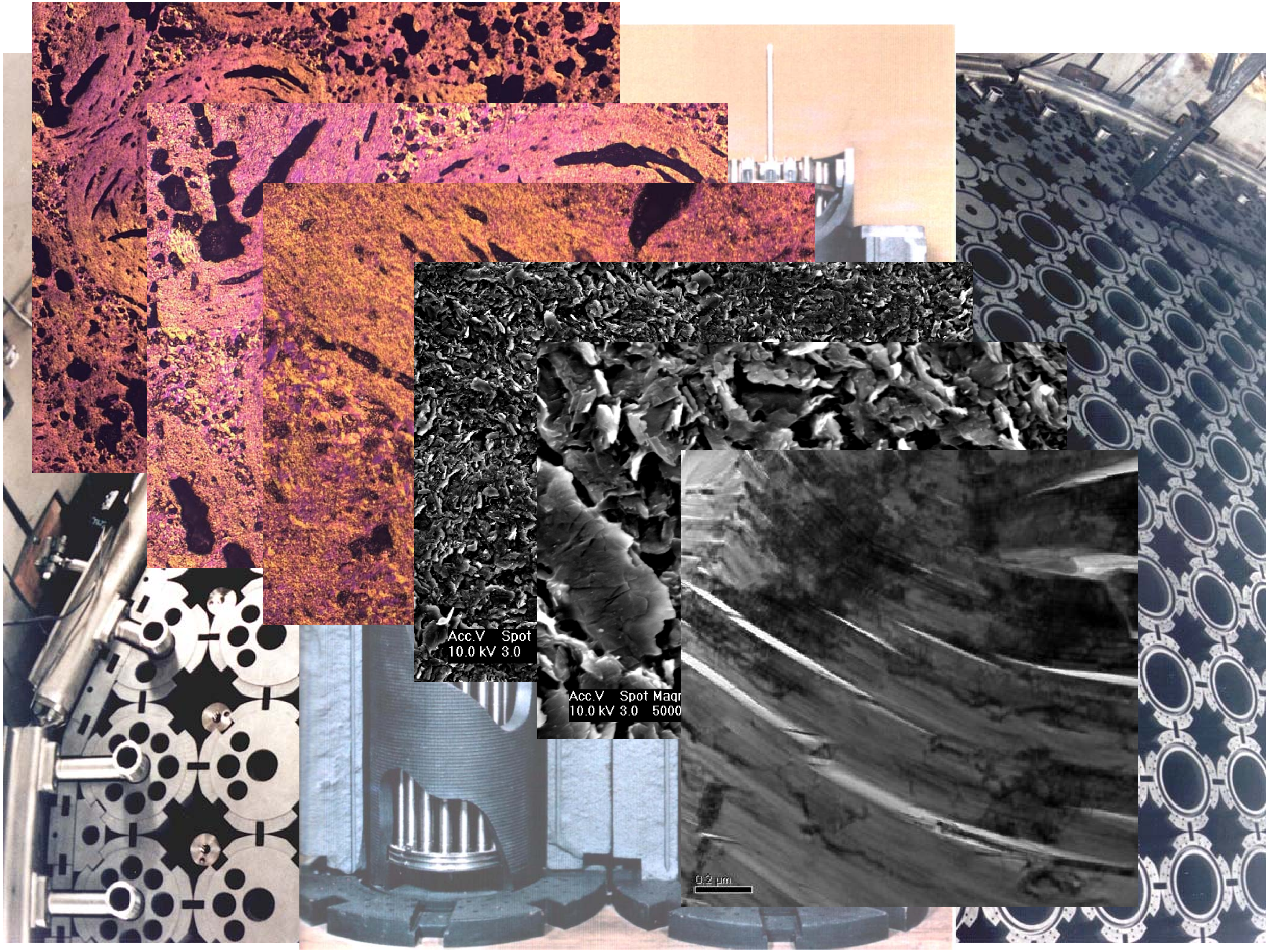
Gilsocarbon



IG-110

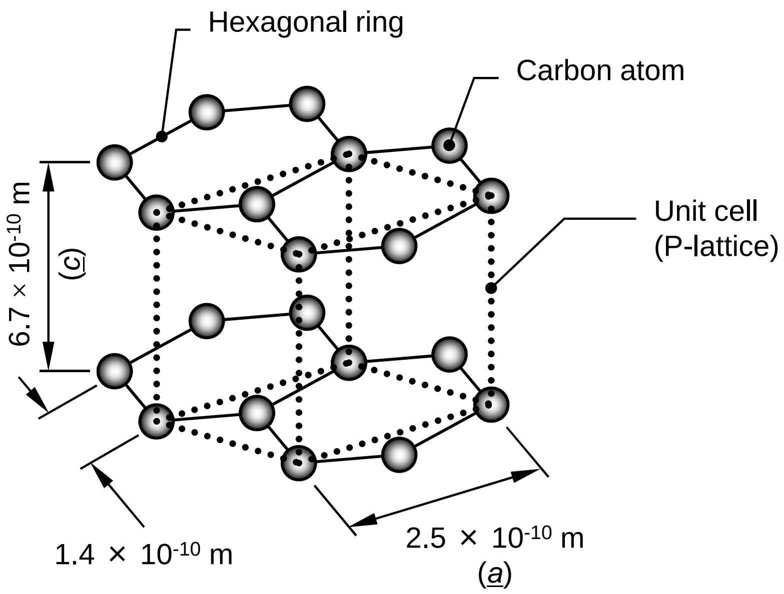


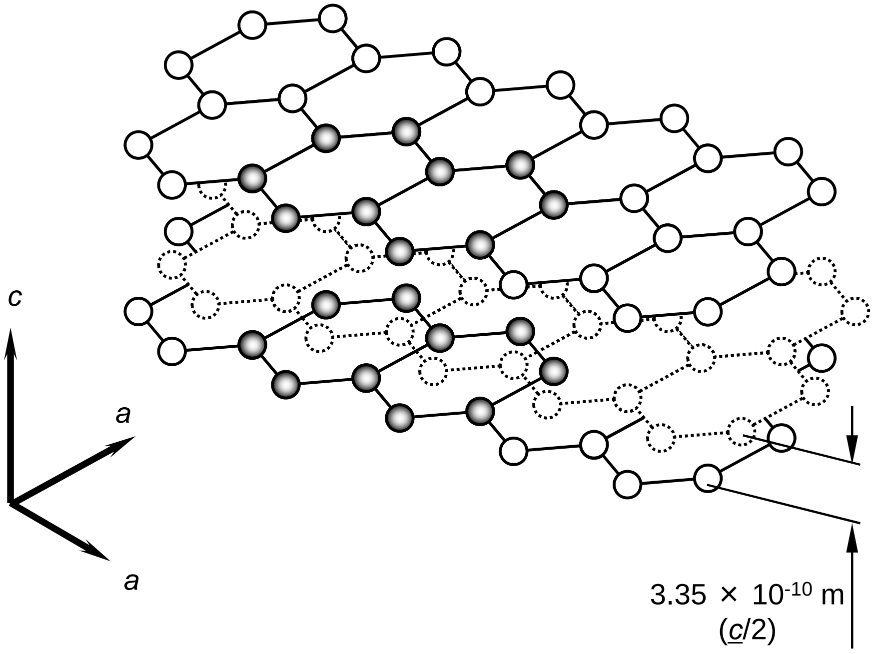
IG-430


Acc.V Spot
10.0 kV 3.0
Acc.V Spot Mag
10.0 kV 3.0 5000
0.2 µm

Crystal structure

- lattice spacing
  - $a = 2.4612 \times 10^{-10}$ m
  - $c = 6.7079 \times 10^{-10}$ m
- alternately stacked planes
  - 335×10^{-12} m
- density
  - 2.66 g/cm^3
- CTE
  - $\alpha_a = -1.25 \times 10^{-6}\ \mathrm{K}^{-1}$ (20-120ºC)
  - $\alpha_c = 26 \times 10^{-6}\ \mathrm{K}^{-1}$ (20-120ºC)





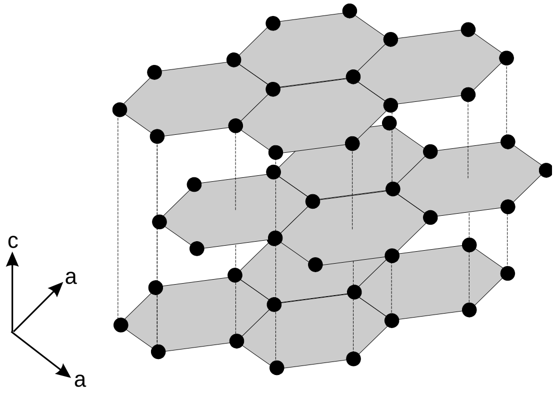
Irradiation damage to graphite Crystallites

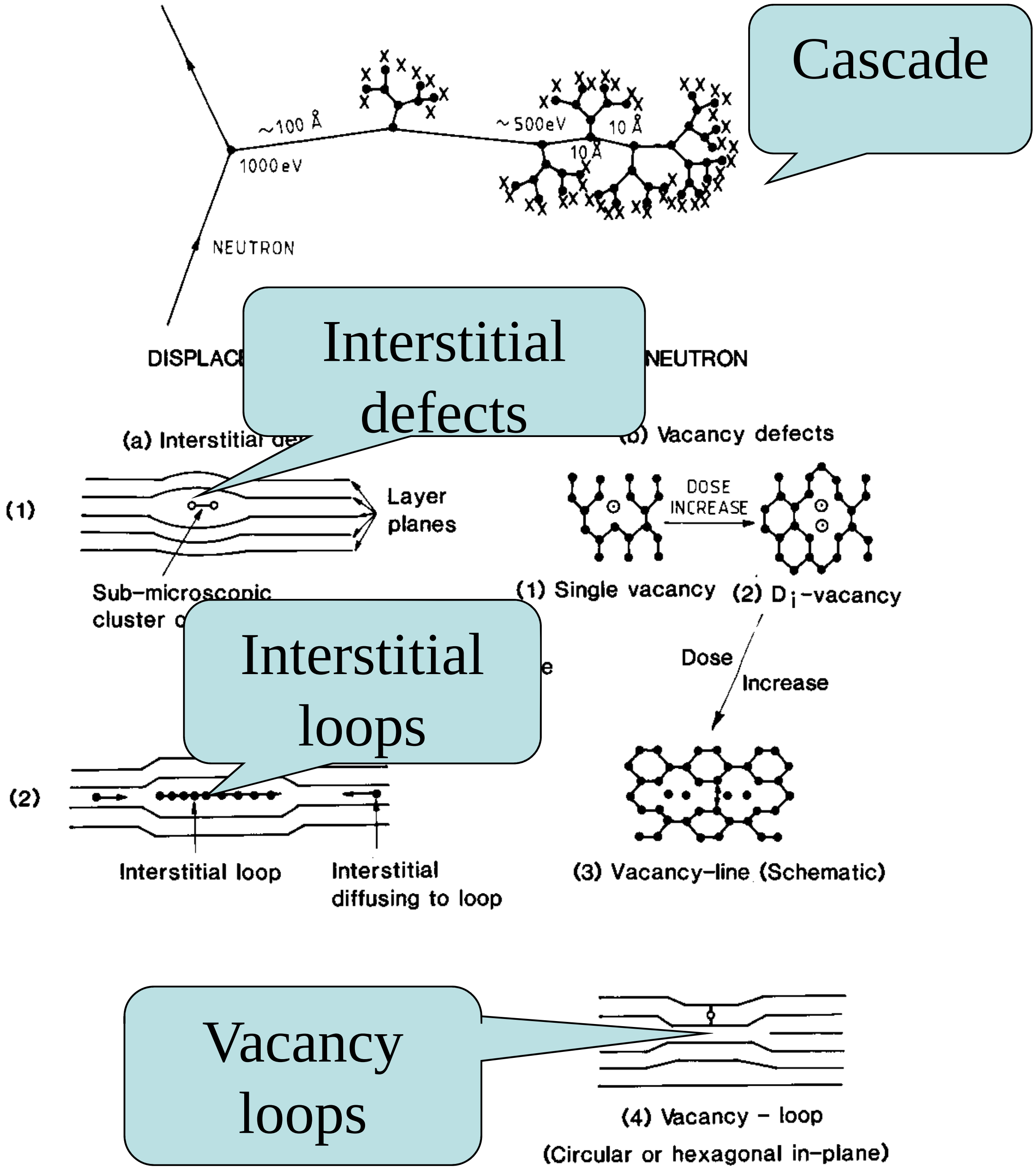
- Damage leads to crystal changes:
  - Stored energy (Significant below irradiation temperatures 150^{o}C, insignificant above 350^{o}C)
  - Dimensional changes
  - Thermal conductivity changes
  - Modulus changes
  - Strength changes?
  - No Coefficient of Thermal Expansion (CTE) changes above ~300^{o}C
  - Irradiation creep (when under stress)

Fast Neutron Damage

- Thermal reactor neutron energies up to 10MeV, average 2MeV
- About 60eV to permanently displace a carbon atom from the lattice
- Most damage due to fast neutron energies > 0.1 MeV
- Cascade caused by primary and secondary knock-ons
- Interstitial and vacancy loops are formed
- Size of loops depends on irradiation annealing
- Change in crystallite behaviour at an irradiation temperature of about 250°C
- A measure of damage is irradiation “dose” of “fluence” units:
- displacements per atom “dpa”
  - n/cm^2 - Equivalent DIDO Dose (EDND)
  - n/cm^2 – with energies greater than 0.18MeV (En>0.18MeV)
  - nvt – neutron velocity time

Formation of interstitial and vacancy loops





Cascade

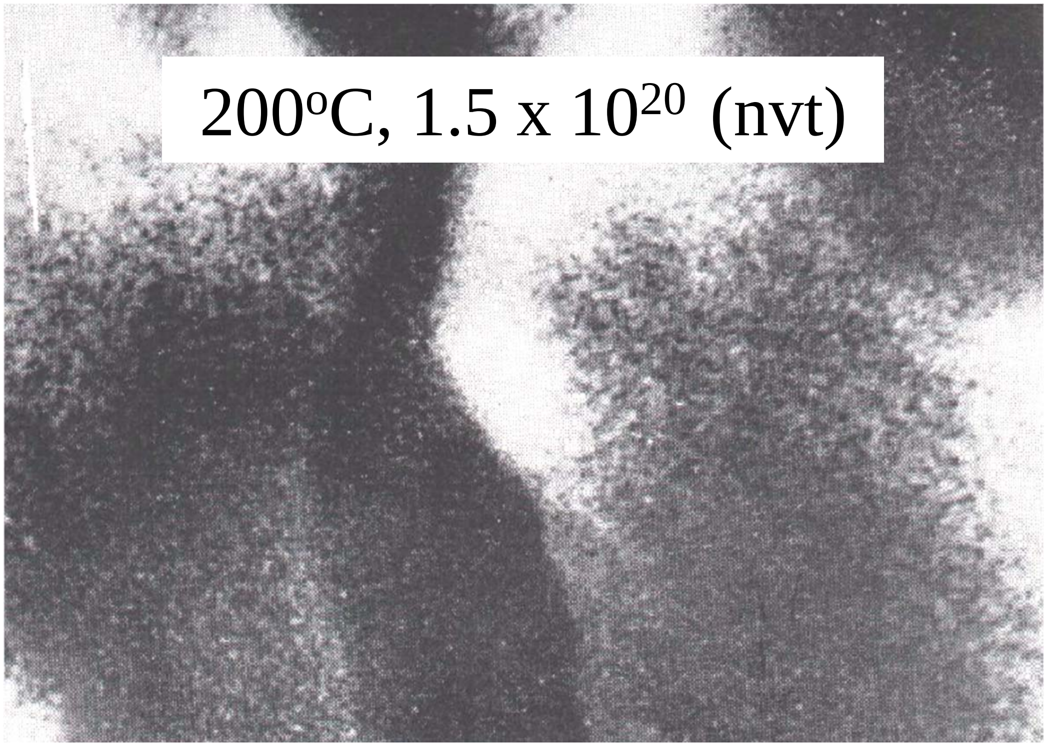
Interstitial defects

Interstitial loops

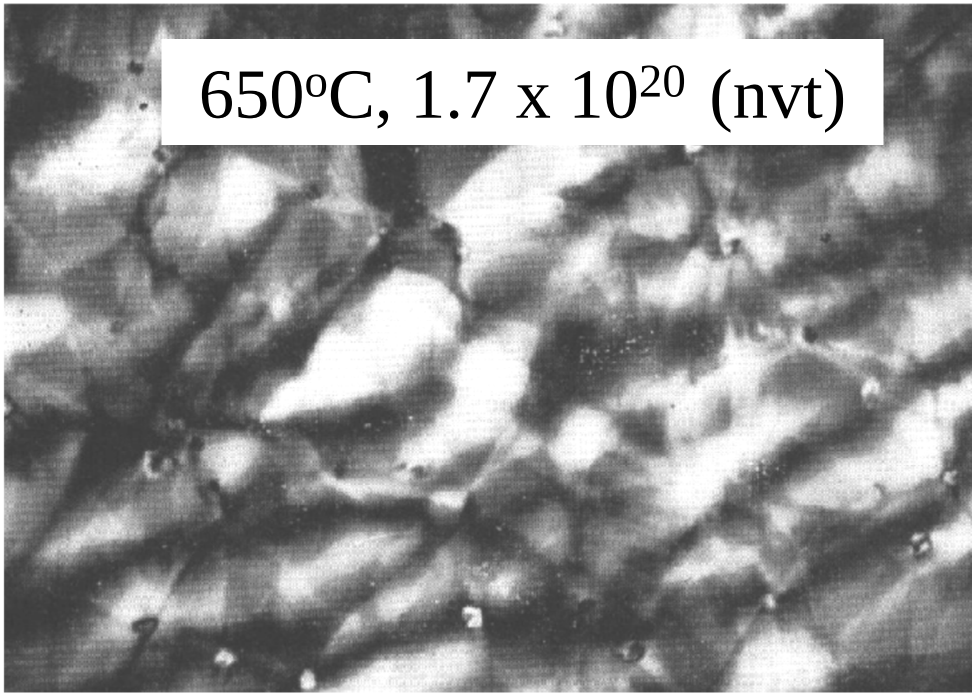
Vacancy loops

Irradiation defects in graphite crystals (HOPG)

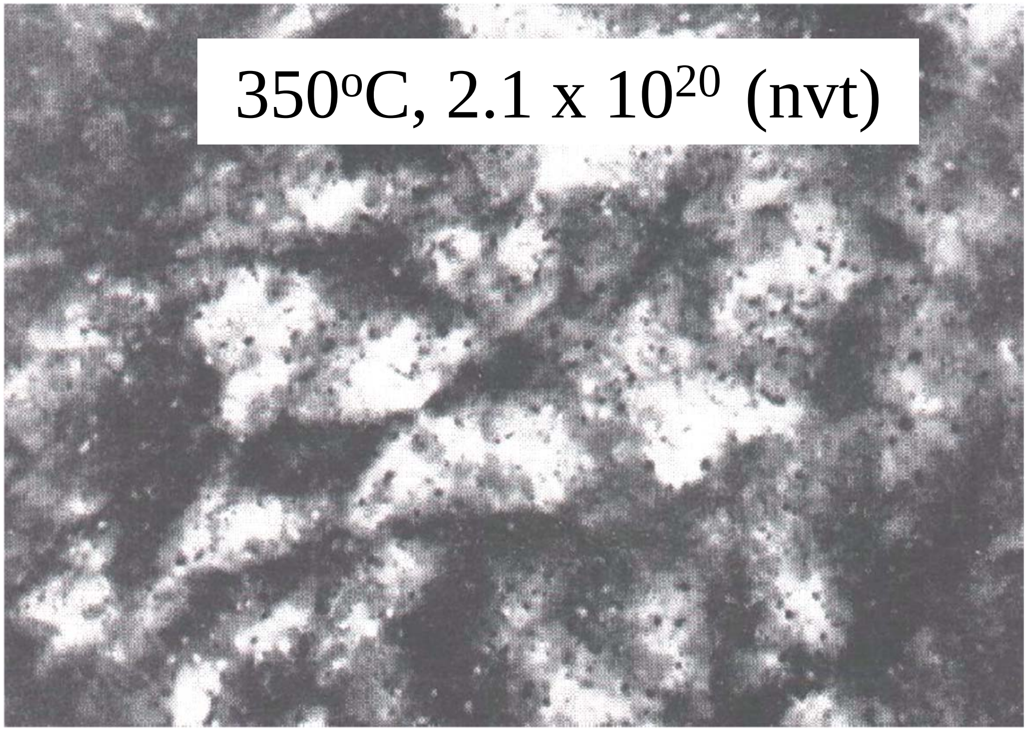
(x 20,000)



200^{o}C, 1.5 x 10^{20} (nvt)



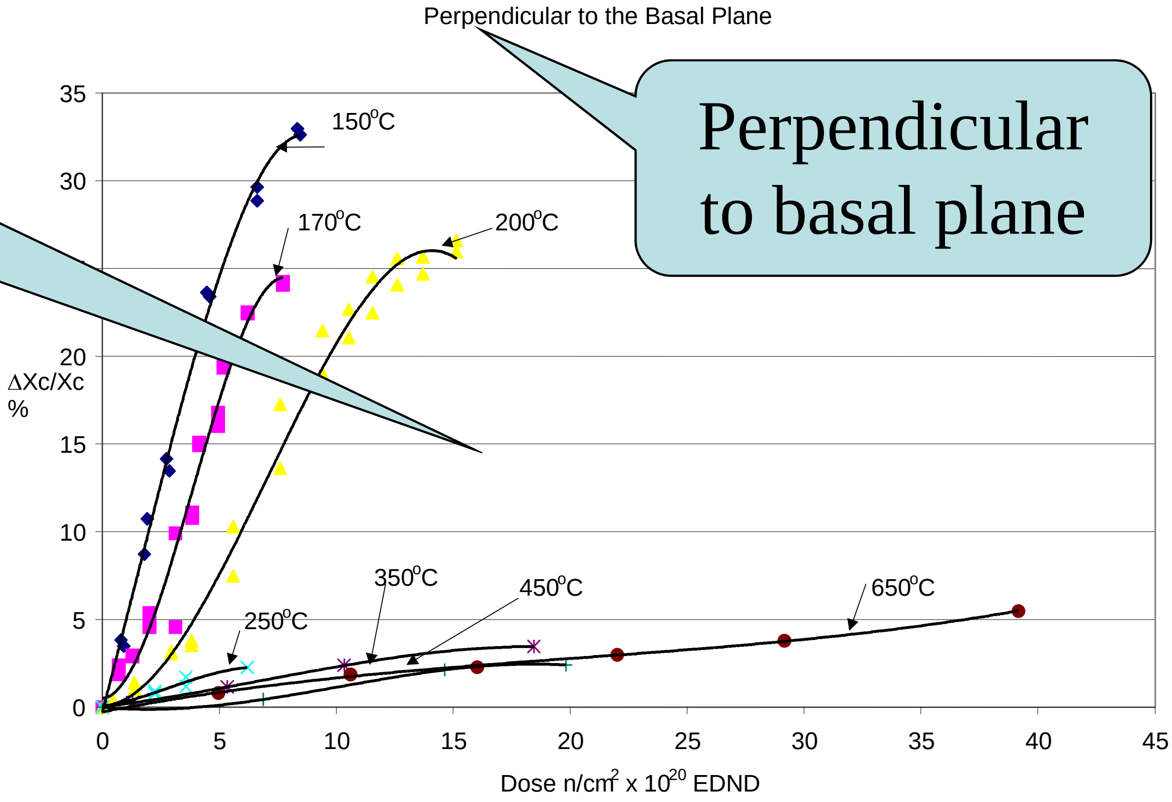
650^{o}C, 1.7 x 10^{20} (nvt)



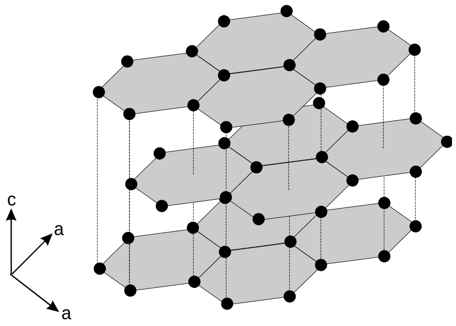
350^{o}C, 2.1 x 10^{20} (nvt)

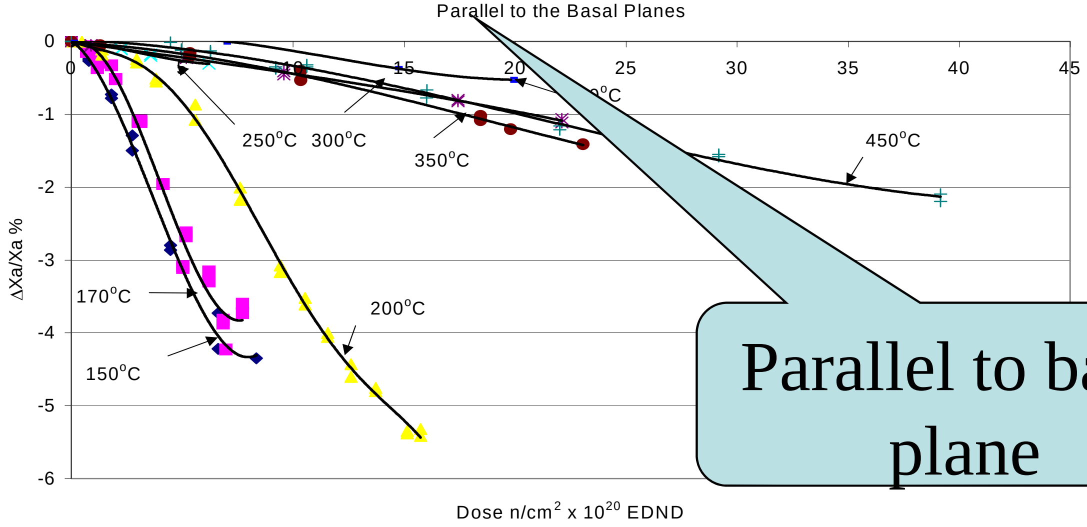
Significant change in rate between 200 and 250°C

Crystal Dimensional Changes measured in HOPG with increasing Dose



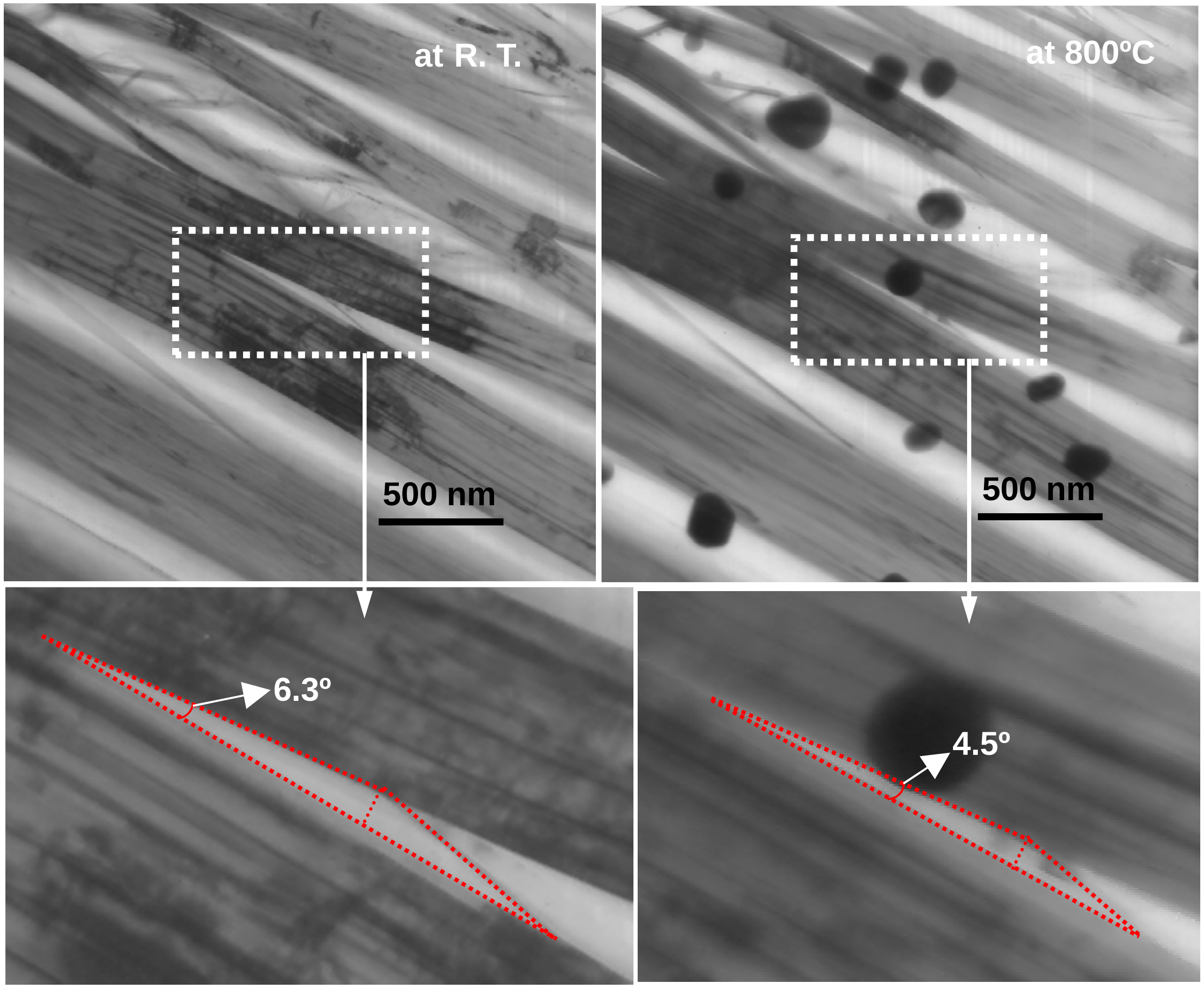
Perpendicular to basal plane





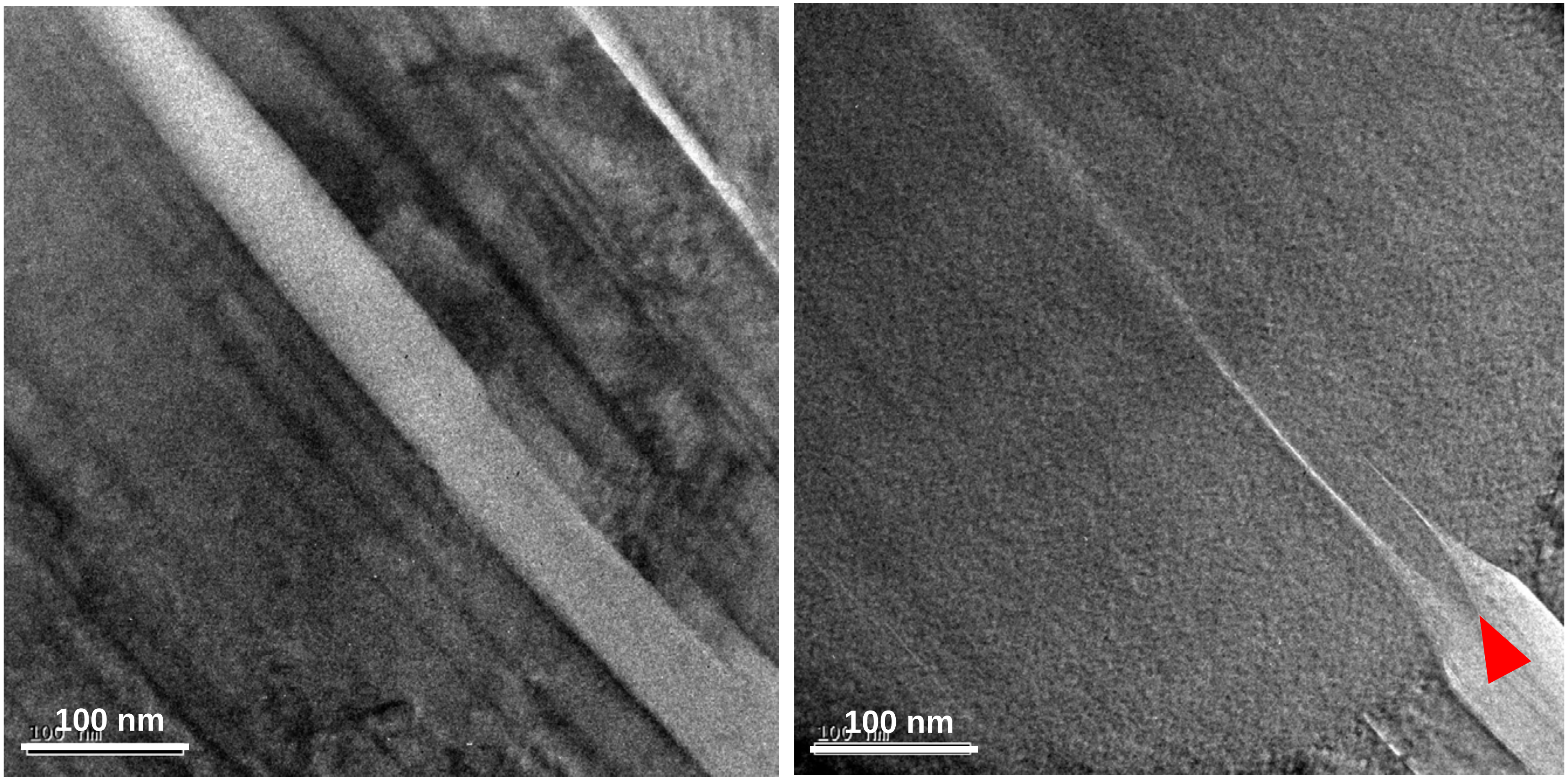
Parallel to basal plane

TEM: *In situ* heating



Upon heating, a gradual closure of cracks was observed because of the thermal expansion of the graphite crystallites surrounding the cracks.

TEM *In situ* electron irradiation



Closure of a crack in Gilsocarbon after In-situ electron irradiation.
The feature with bright contrast does not disappear completely.
Note a small part of crack (indicated by arrow), which was covered by the electron beam has not closed completely

Radiolytic Oxidation

- Two types of oxidation can occur in CO_2.
  - Thermal oxidation is a purely chemical reaction between graphite and CO_2.
  - Reaction is endothermic, is negligible below about 625^{o}C and is not important up to 675^{o}C.
  - Only an issue for HTRs
- Radiolytic oxidation occurs when CO_2 is decomposed by fast neutron and gamma radiation (radiolysis) to form CO and an active oxidising species which attacks the graphite porous structure.
  - Radiolytic oxidation occurs predominantly within the graphite pores.
  - Overall component geometry stays essentially the same

Radiolytic Oxidation

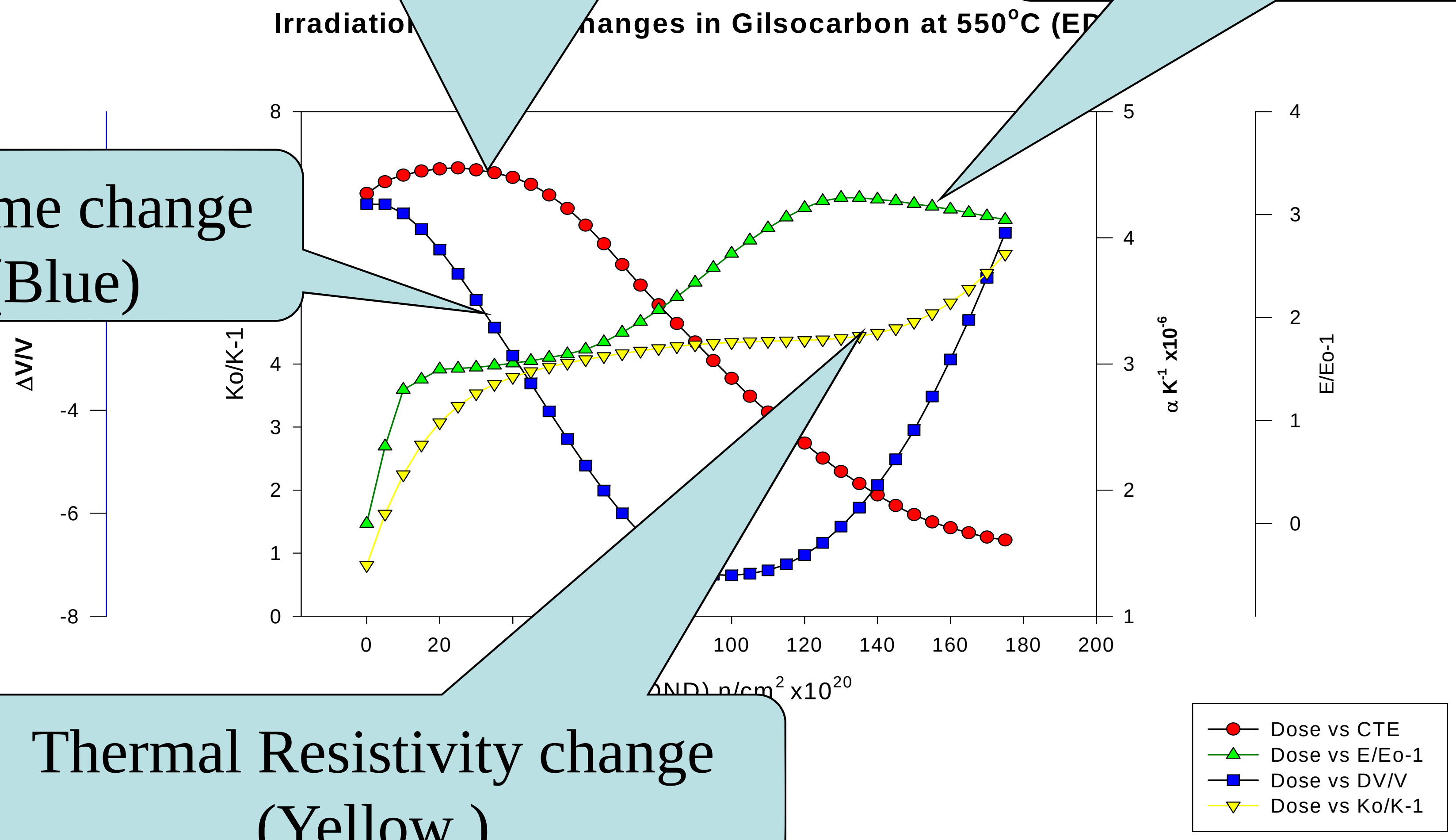
- The mechanism of radiolytic oxidation is:
- Gas Phase
- CO_2--------radiation -----> CO+O*
- CO+O*---------------------> CO_2
- Graphite Pore Surface
- O*+C----------------------> CO
- Definition
  - G_{-c} is the number of carbon atoms gasified by the oxidising species produced by the absorption of 100eV of energy in the CO_2 contained within the graphite pores.

Oxidising species

Irradiation Damage in Polycrystalline Graphite

- Crystal changes modify polycrystalline dimensions and properties through the microstructure
  - Stored Energy – Only significant below 150^{o}C, negligible at 350^{o}C
  - Dimensional changes
  - CTE
  - Young’s modulus
  - Strength
  - Thermal conductivity
  - Irradiation creep (when under stress)
- Radiolytic oxidation further modifies these properties
- Semi-isotropic graphite is considered in the next section

Graphite Irradiation Behaviour – Isotropic Gilsocarbon irradiated at 550°C



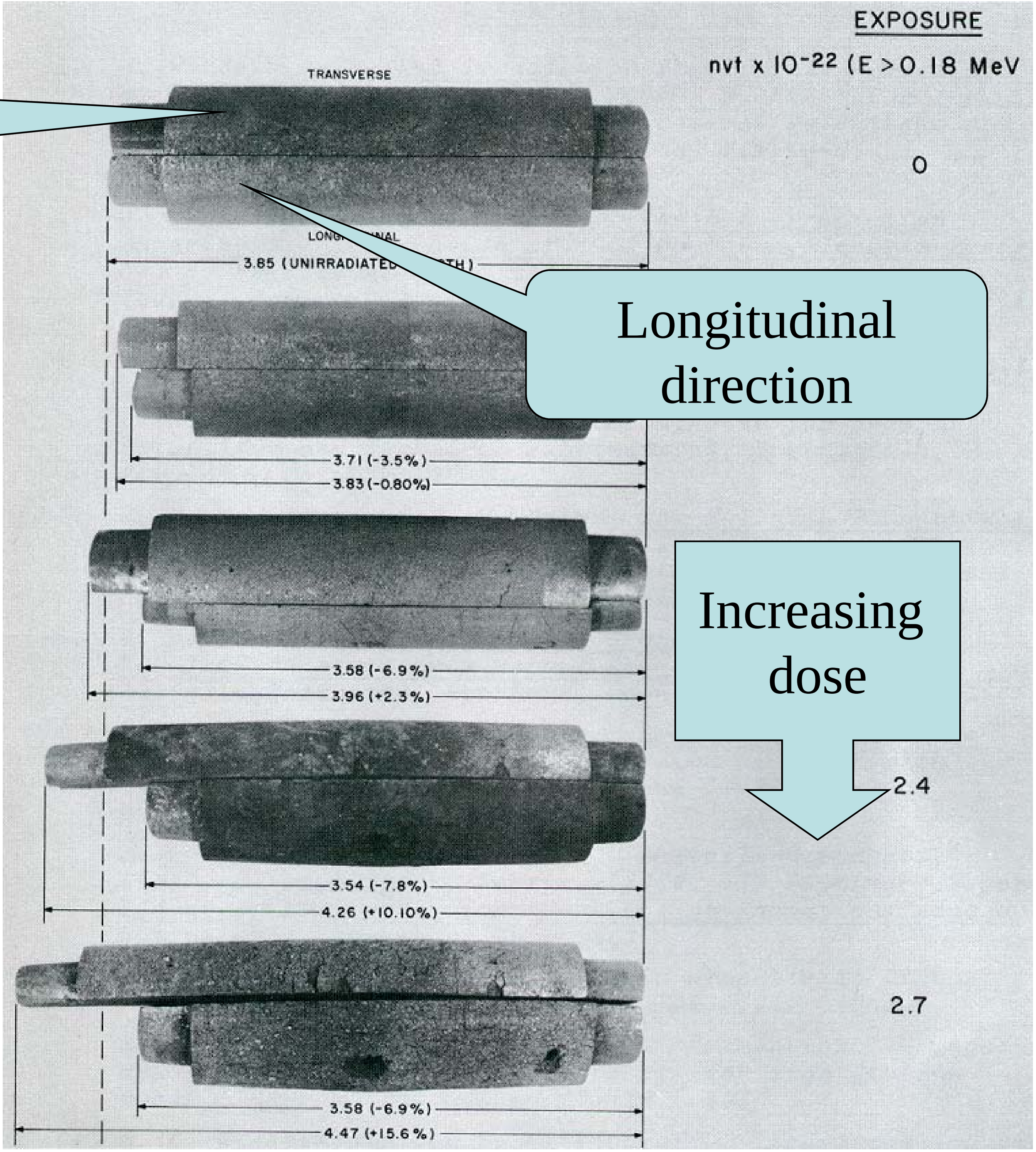
Coefficient of Thermal Expansion change (Red)

Young’s modulus change (Green)

Volume change (Blue)

Thermal Resistivity change (Yellow)

Shrinkage of CSF Graphite Irradiated at 800^{o}C to various Irradiation Doses

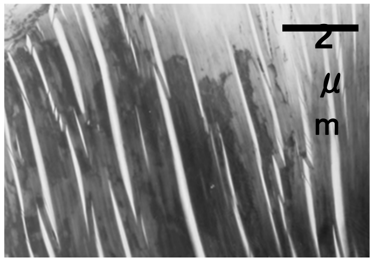


Transverse direction

Longitudinal direction

Increasing dose

Gilsocarbon Dimensional Changes



Mrozowski cracks

Gilscarbon Dimensional change

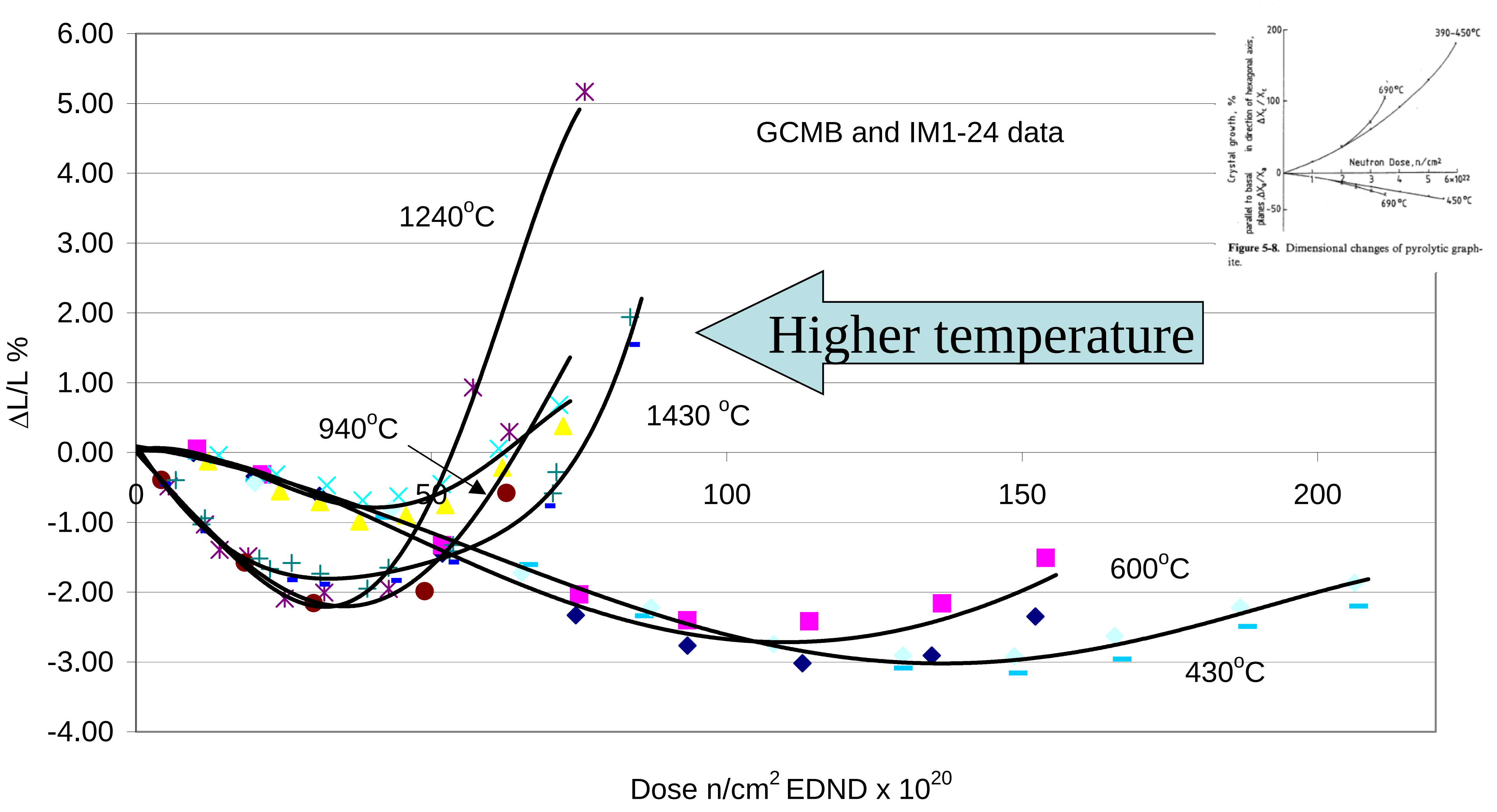


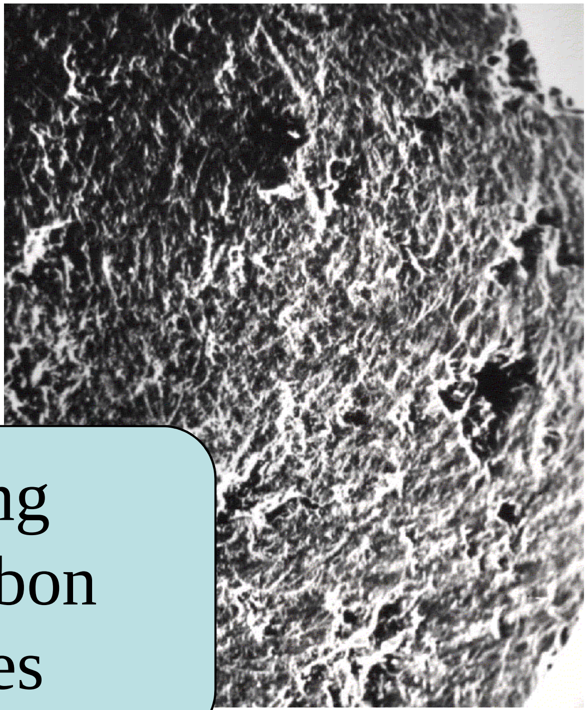
Figure 5-8. Dimensional changes of pyrolytic graphite.

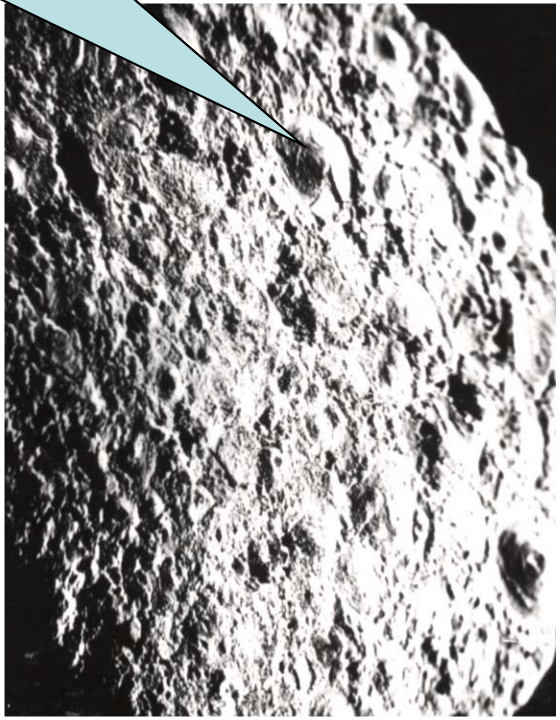
Unirradiated Gilsocarbon Specimens 7mm long by

m di

Swelling Gilsocarbon particles

Swelling Gilsocarbon particles

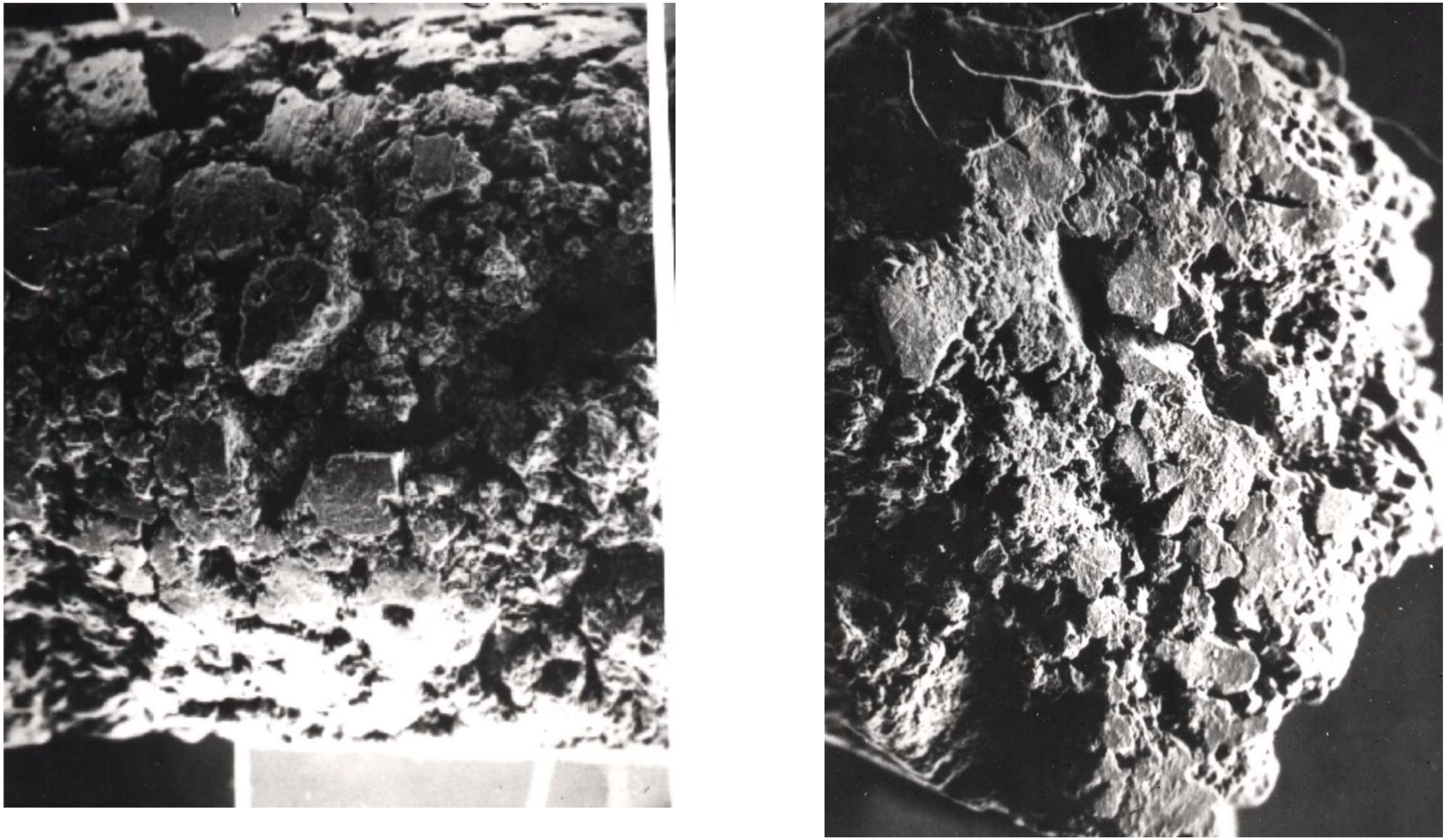




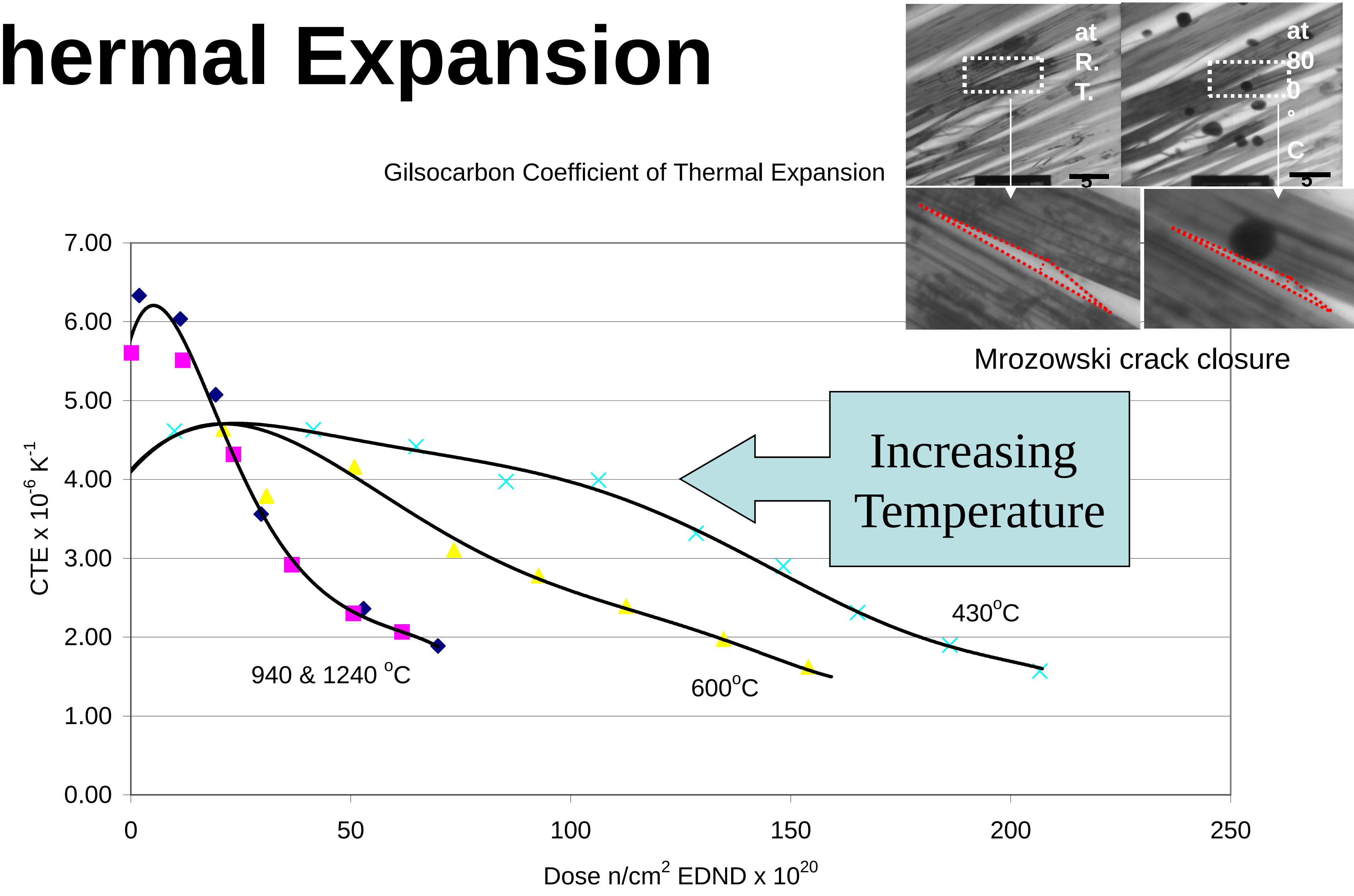
285×10^{20} n/cm^2 EDND

$+0.9\% \ \Delta V/V_o$

Gilsocarbon irradiated to 271 x 10^{20} n/cm^2 EDND 33% $\Delta V/V_o$



Gilsocarbon Coefficient of Thermal Expansion

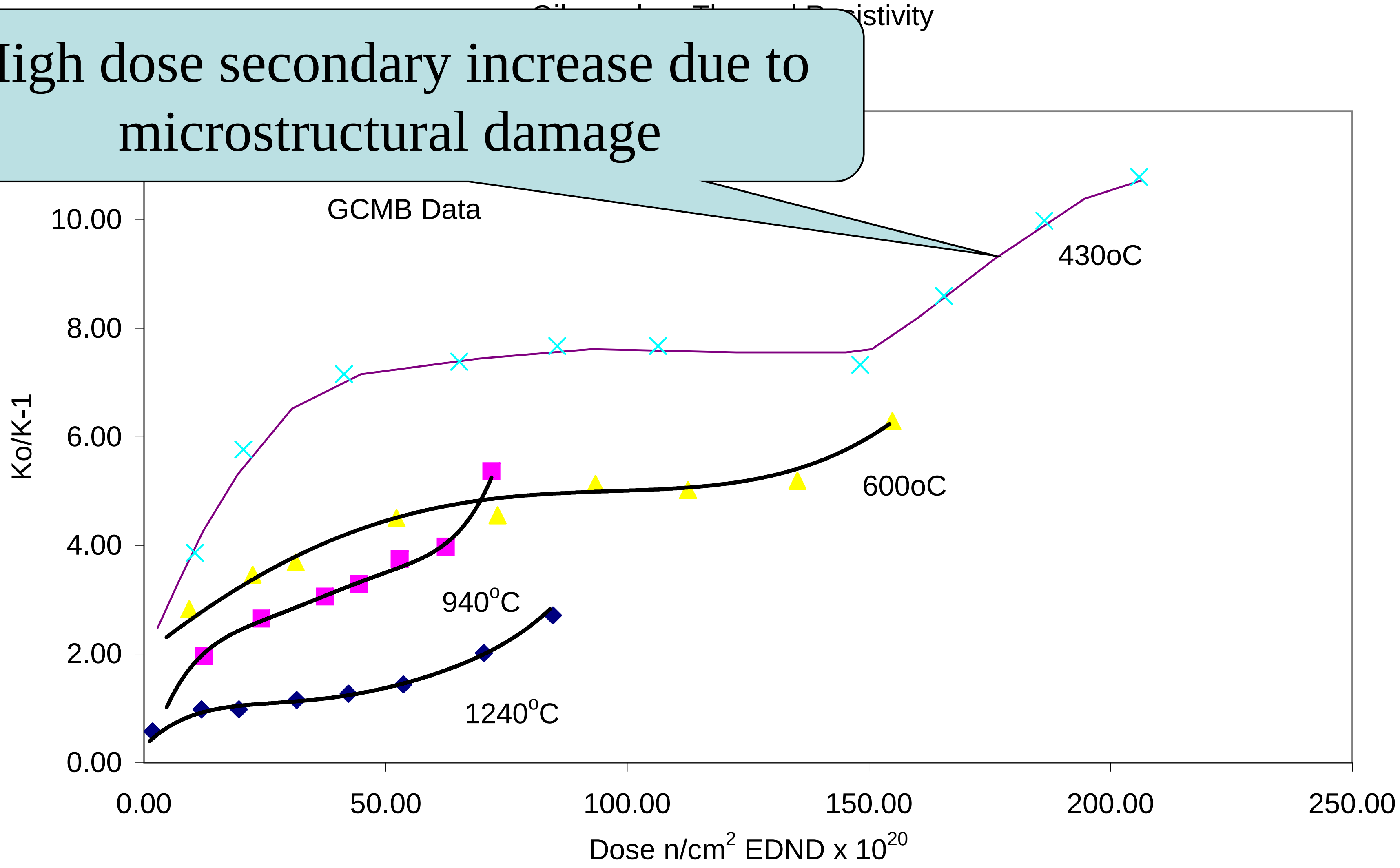


Mrozowski crack closure

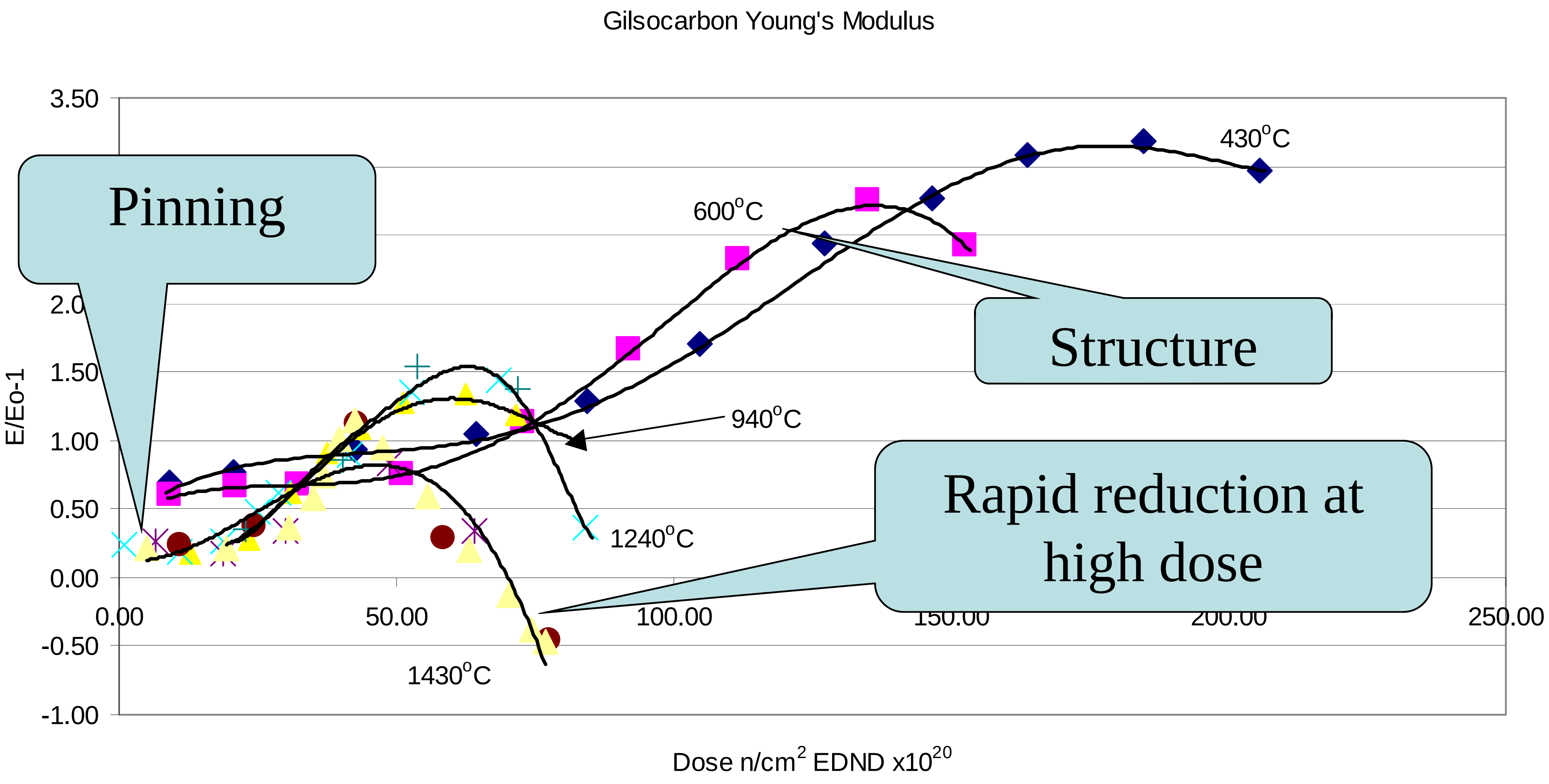
Increasing Temperature

Gilsocarbon Thermal Resistivity

High dose secondary increase due to microstructural damage



Gilsocarbon Change Young’s Modulus



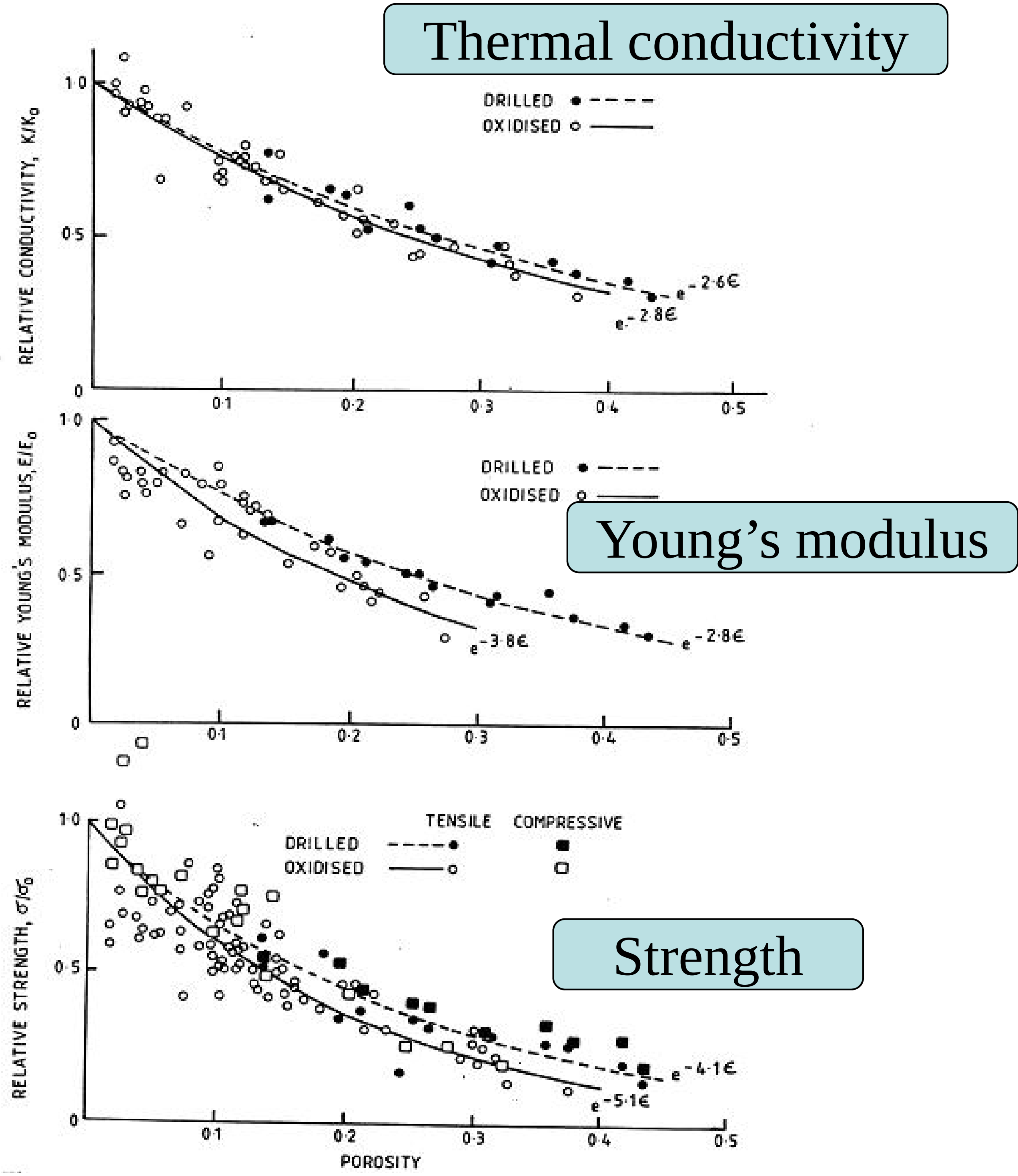
Pinning

Structure

Rapid reduction at high dose

Reduction in properties due to radiolytic oxidation

- The black symbols are drilled specimens indicating the loss of section is a major factor



Thermal conductivity

Young's modulus

Strength

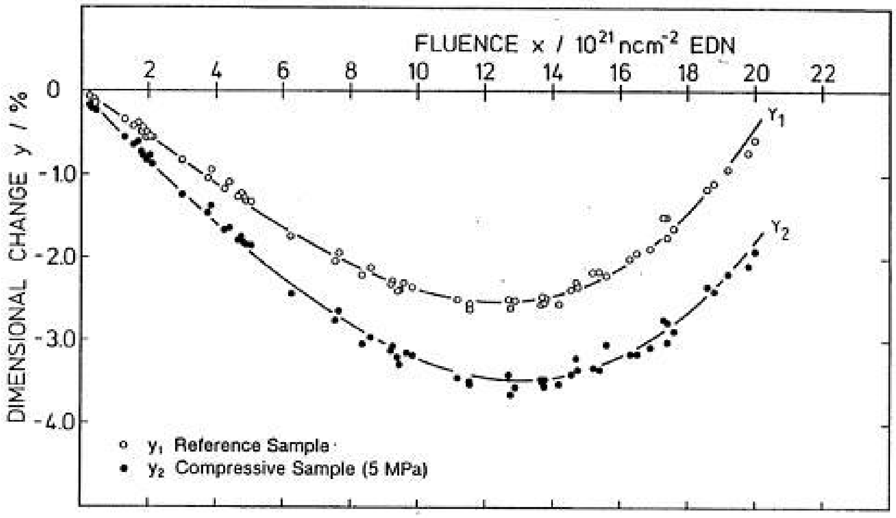
Irradiation Creep in Graphite

- Due to fast neutron irradiation
- Significantly reduces stresses in nuclear graphite components
- Definition
  - The difference in dimensions between a stressed sample and a sample having the same properties as that sample when unstressed

Dimensional Change Under Load

Example ATR-2E Graphite

- Under compressive load shrinkage is increased
  - Upper right
- Under tensile load shrinkage is decreased
  - Lower right
- There is also a lateral (Poisson's) effect
  - Below



DIMENSIONAL CHANGES OF ATR-2E AT 500°C UNDER COMPRESSIVE LOAD

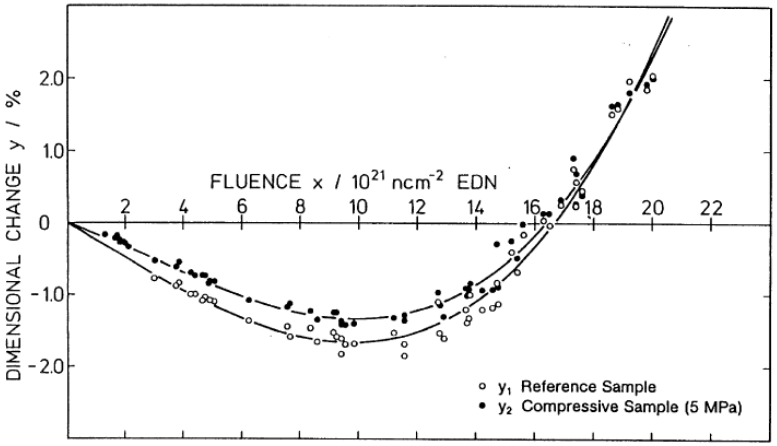
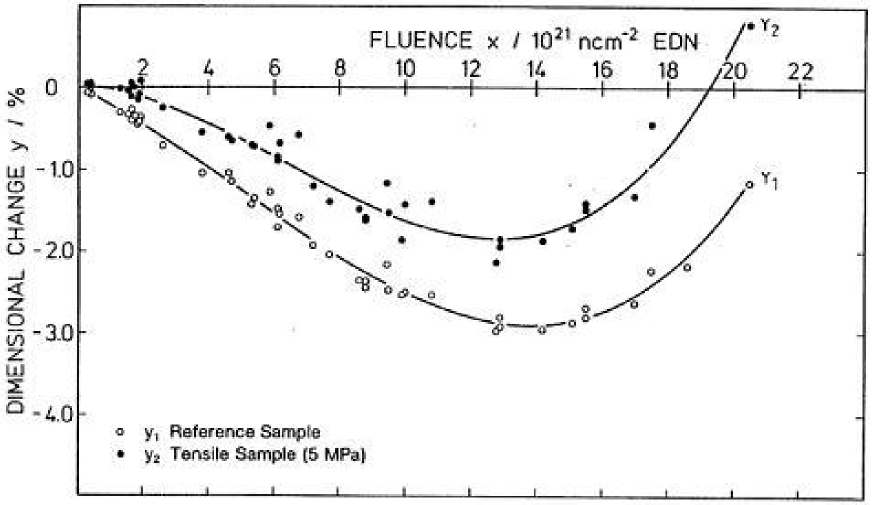


FIG. 18. DIMENSIONAL CHANGES OF ATR-2E AT 500°C UNDER COMPRESSIVE LOAD (TRANSVERSE DIRECTION) (REF. 37)

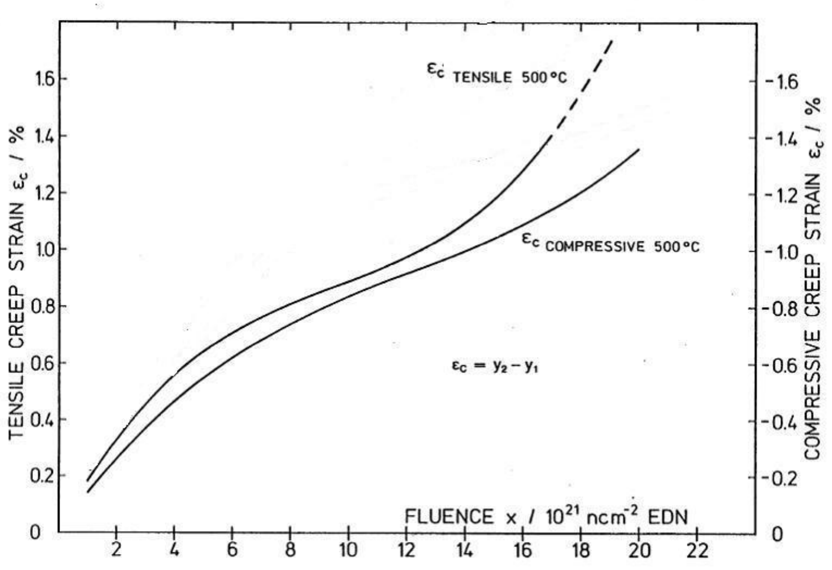


DIMENSIONAL CHANGES OF ATR-2E AT 500°C UNDER TENSILE LOAD

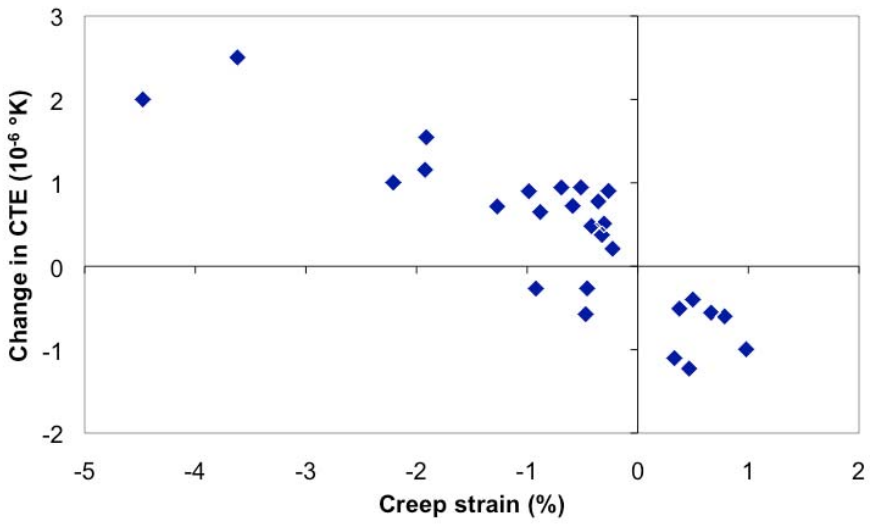
Irradiation Creep Curves

Example ATR-2E (500ºC)

- Irradiation creep curve can be simply obtained by subtraction of the unloaded dimensional change curve from the crept dimensional change curve
- However, for assessments this would require data for a range of temperatures and fast neutron fluence covering all the expected conditions.
- In addition changes to the Coefficient of Thermal Expansion (CTE) and Young’s modulus have been observed.



CREEP STRAINS OF ATR- 2E UNDER 5 MPa LOAD



Issues to consider

- Properties
  - thermal conductivity
  - thermal shock resistance
  - modulus of elasticity
  - tensile strength
  - CTE
  - dimensional change & irradiation creep
    - initial compressive stress
- Protons versus neutrons
  - dose rate effect (pulsed versus continuous)
  - helium production
- POCO
  - historical experience