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The Influence of High Energy Proton Irradiation on Fine-Grained Isotropic Graphite Grades: A Summary of Recent RaDIATE Results

P. Hurh – RaDIATE Collaboration Program Coordinator

Contribuitors: N. Simos (BNL); K. Ammigan, V. Sidorov, J. Hylen (FNAL); D. Senor, A. Casella (PNNL); D. Liu (Oxford); T. Davenne (STFC)

High Power Targetry Workshop, 04 June 2018

RaDIATE Collaboration

Radiation Damage In Accelerator Target Environments

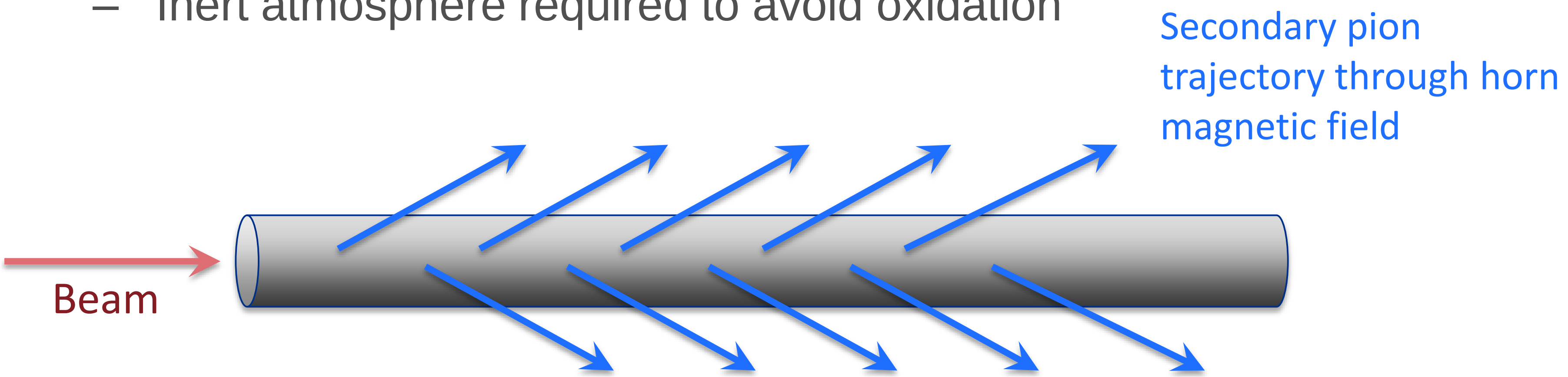
http://radiate.fnal.gov

- To generate new and useful materials data for application within the accelerator and fission/fusion communities;
- To recruit and develop new scientific and engineering experts who can cross the boundaries between these communities;
- To initiate and coordinate a continuing synergy between research in these communities, benefitting both proton accelerator applications in science and industry and carbon-free energy technologies

In 2017, MoU revision has counted J-PARC (KEK+JAEA) & CERN as official participants

Graphite Advantages for a Nu Target

- Physics: Low Z (Atomic Number) higher yield of low energy Nu's
  - Although it means a longer target, the low Z results in less re-interaction of the secondary pions on the way out of the sides of the target (long, but narrow target is an advantage, especially for low-energy neutrino experiments)
- Thermal Shock Resistance
  - Very low effective modulus of elasticity mean stresses from thermal shock are 3x's less than metallic counterparts (beryllium)
- High temperature operation
  - Inert atmosphere required to avoid oxidation



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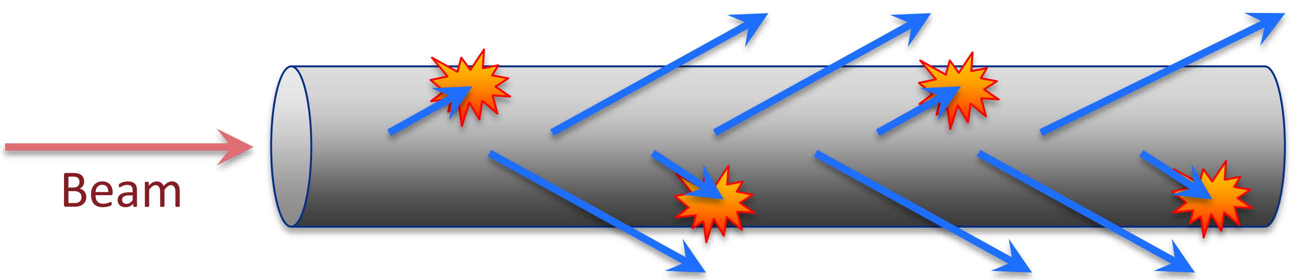


Some Secondary pions Interact with target before exiting the target

Non-irradiated properties of graphite vs Be

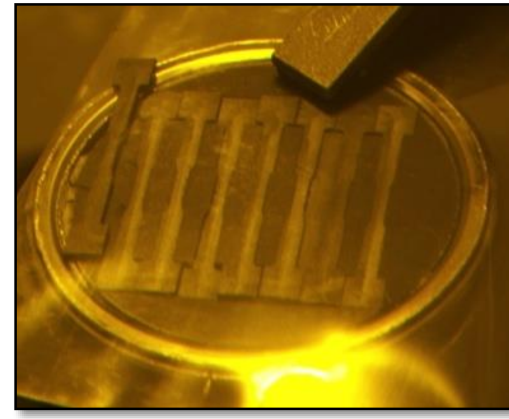
Property	POCO ZXF-5Q	ToyoTanso IG-43(0)	Be S200F
Comp Strength (MPa)	175	97	-
Tensile Strength (MPa)	79	38	345
Elastic Modulus (GPa)	14.5	10.8	309
CTE (10^{-6} K^{-1})	8.1	4.5	11.5
Specific Heat (J/Kg/K)	710	630	1829
Thermal Cond (W/m/K)	70	143	183
Thermal Shock Resist	0.48	0.49	0.18
Application	NuMI	T2K	Beam windows

Thermal Shock Resistance = (UTS*C) / (CTE * E)

- What about radiation damage from high energy protons?

RaDIATE Graphite Studies

- 2010 – 2012 LBNE Graphite Study at BLIP, BNL
  - 4 Grades of graphite
  - C-C Composite
  - Irradiation Temp 120 – 180 ˚C
  - 0.1 DPA
- NT-02 NuMI-MINOS Graphite Target Fin Study
  - Dave Senor et al., PNNL
  - Dong Liu, Oxford
  - Nick Simos et al., NSLS-II, BNL
  - Irradiation Temp 90 – 300 ˚C
  - 0.6 DPA
- MET-01 NuMI-NOvA Graphite Target Fin Study
  - Visual observation only
  - Irradiation Temp 300 – 700 ˚C
  - 1.1 DPA



Graphite micro-structure

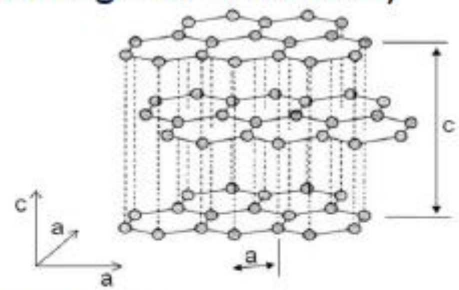
Slide by Dong Liu, Oxford

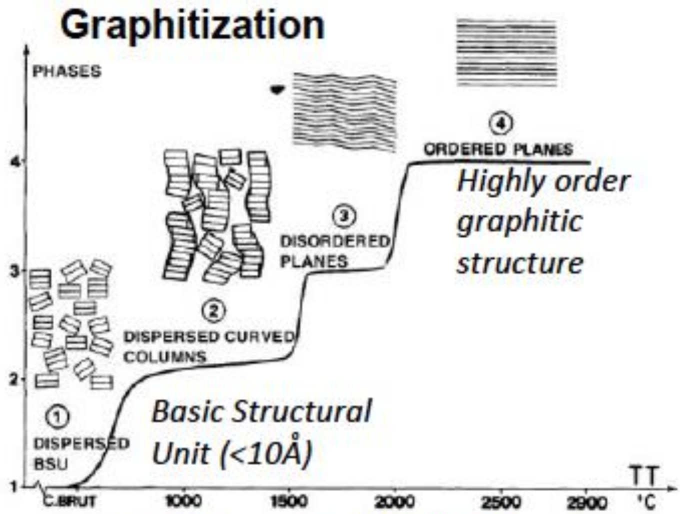
Desired properties in nuclear-grade graphite:

- High purity; High density; High thermal conductivity; Low CTE
- High irradiation stability (pitch, coke and manufacture process);
- High strength (flexural: 20-30MPa);
- Low anisotropy, (less than 1.1, defined by CTE in orthogonal directions)
- Low elastic modulus (~11 GPa for Gilsocarbon)

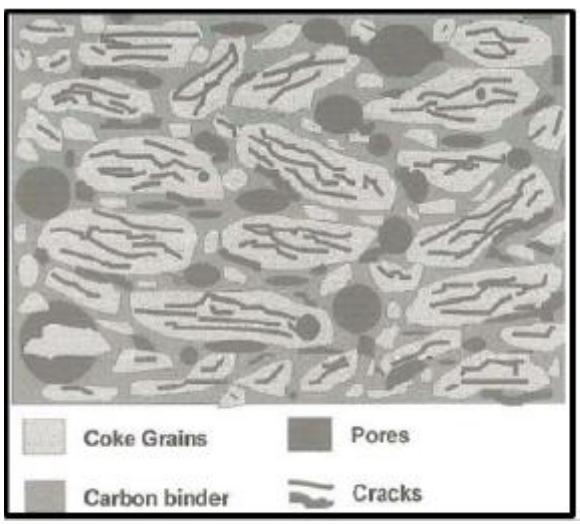
Facts:

- Damage and fracture
- Microstructure, porosity
- Residual stresses





Oberlin A. Carbon N Y 1984;22:521-41



Graphite micro-structure

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OxfordMaterials

Desired pro
- High pur
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- Low anis
- Low elas

Facts:
- Damage
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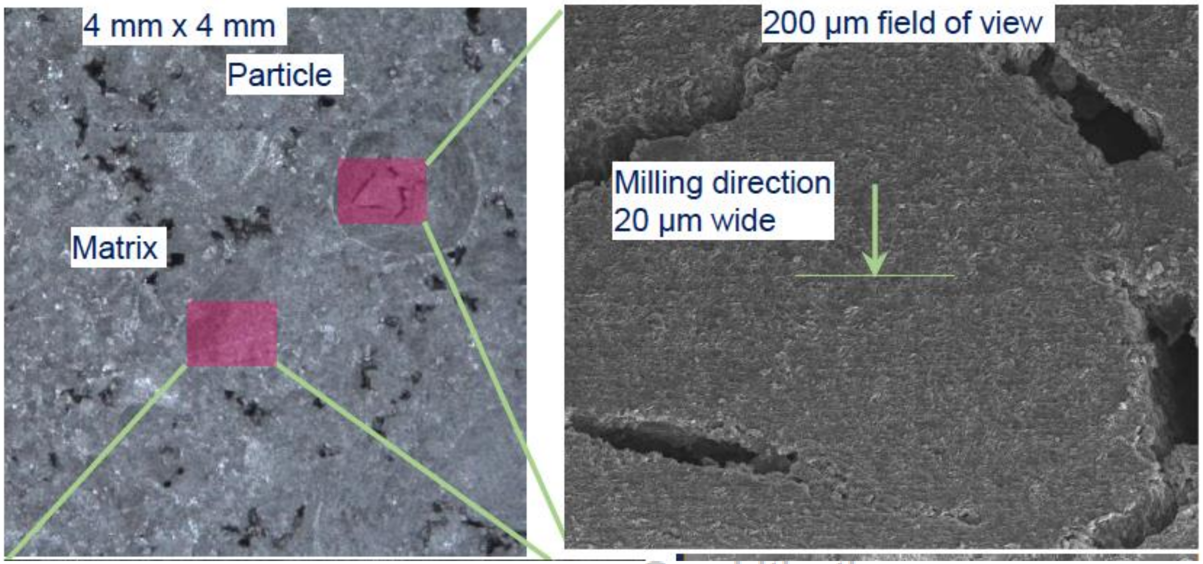
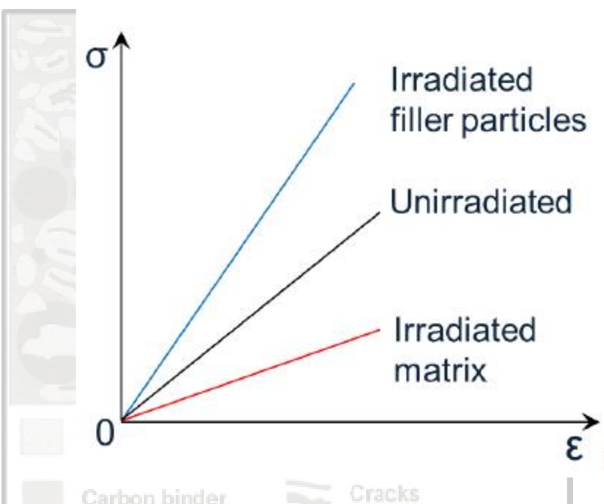
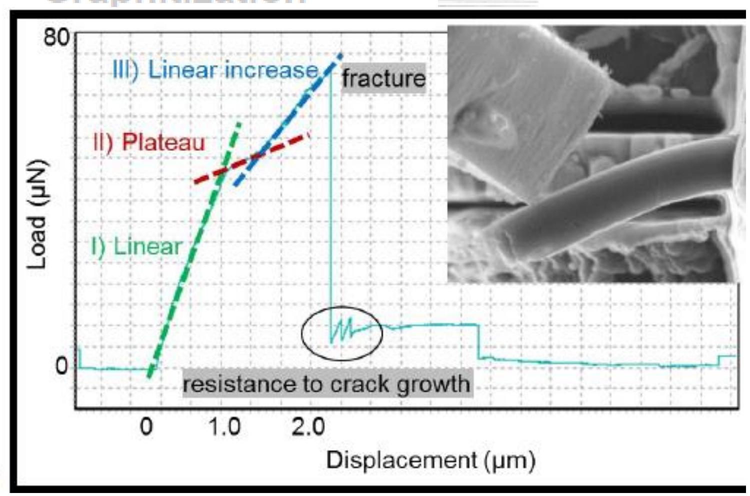


Graphitization

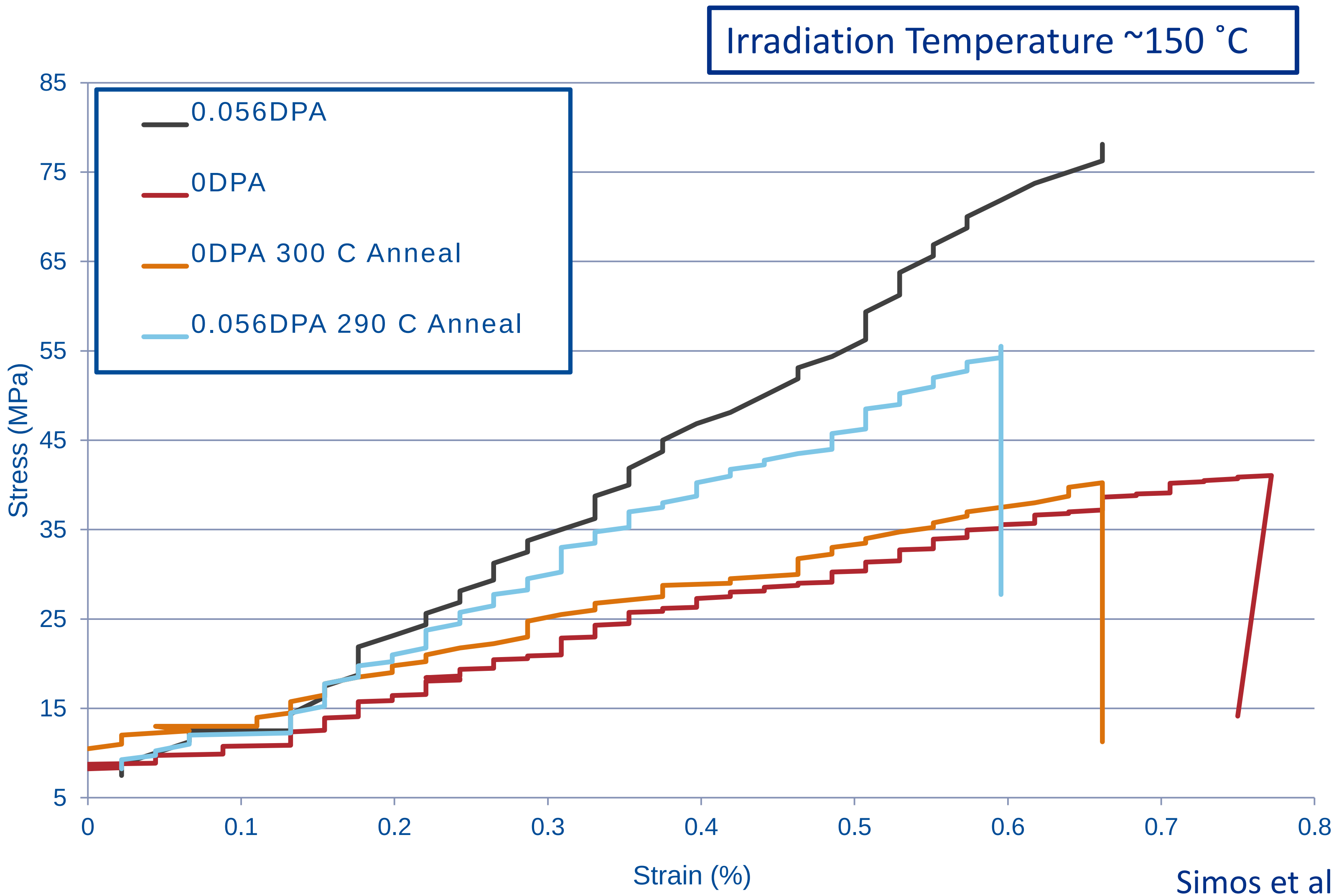


Liu *et al*. Carbon, 2017



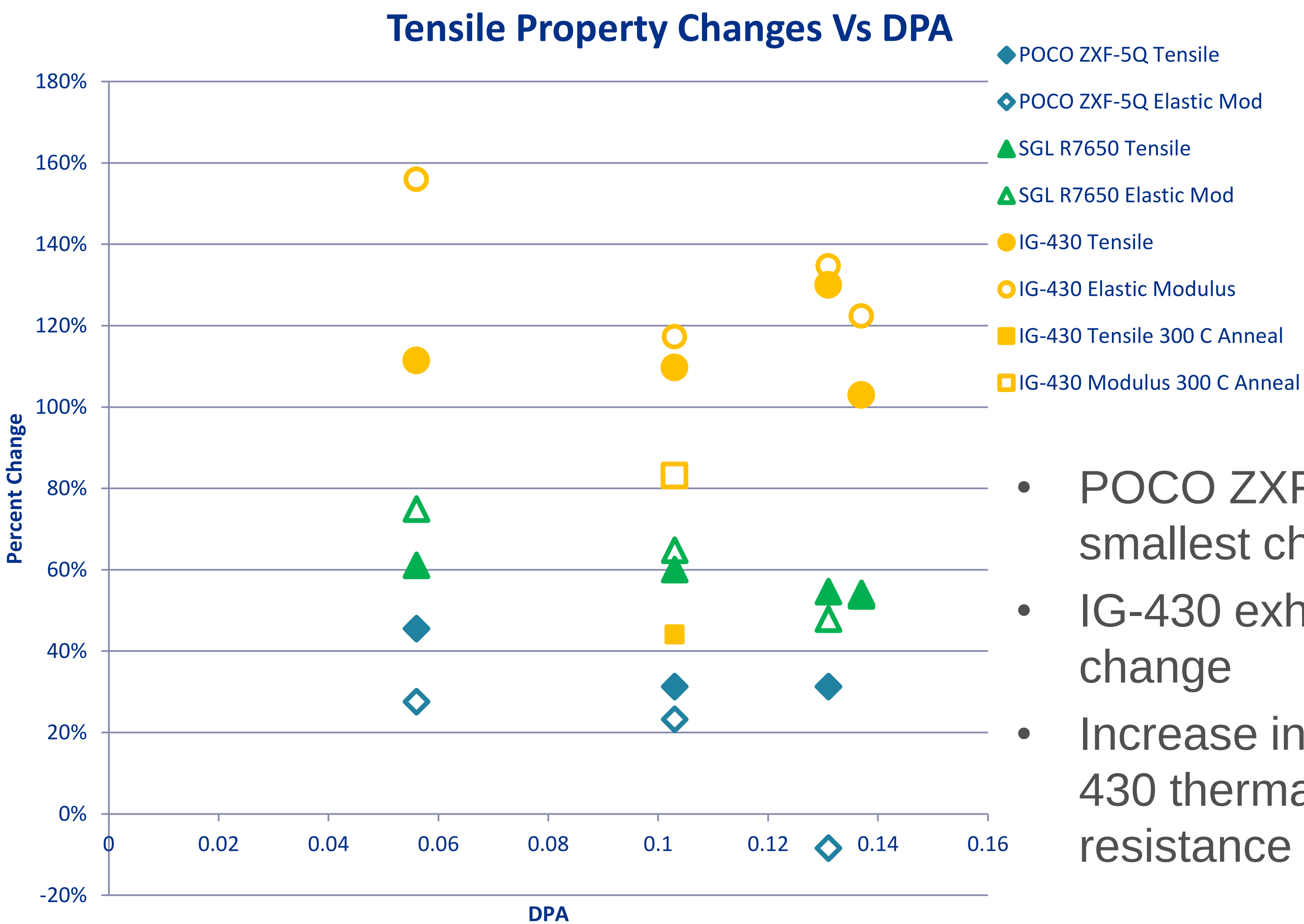
Oberlin A. Carbon N Y 1984;22:521-41

Results – Typical Tensile Properties (IG-430)



Simos et al

Results – Tensile Properties Summary Plot



- POCO ZXF-5Q exhibited smallest change
- IG-430 exhibited largest change
- Increase in E lowers IG-430 thermal shock resistance

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Results – Sonic Velocity

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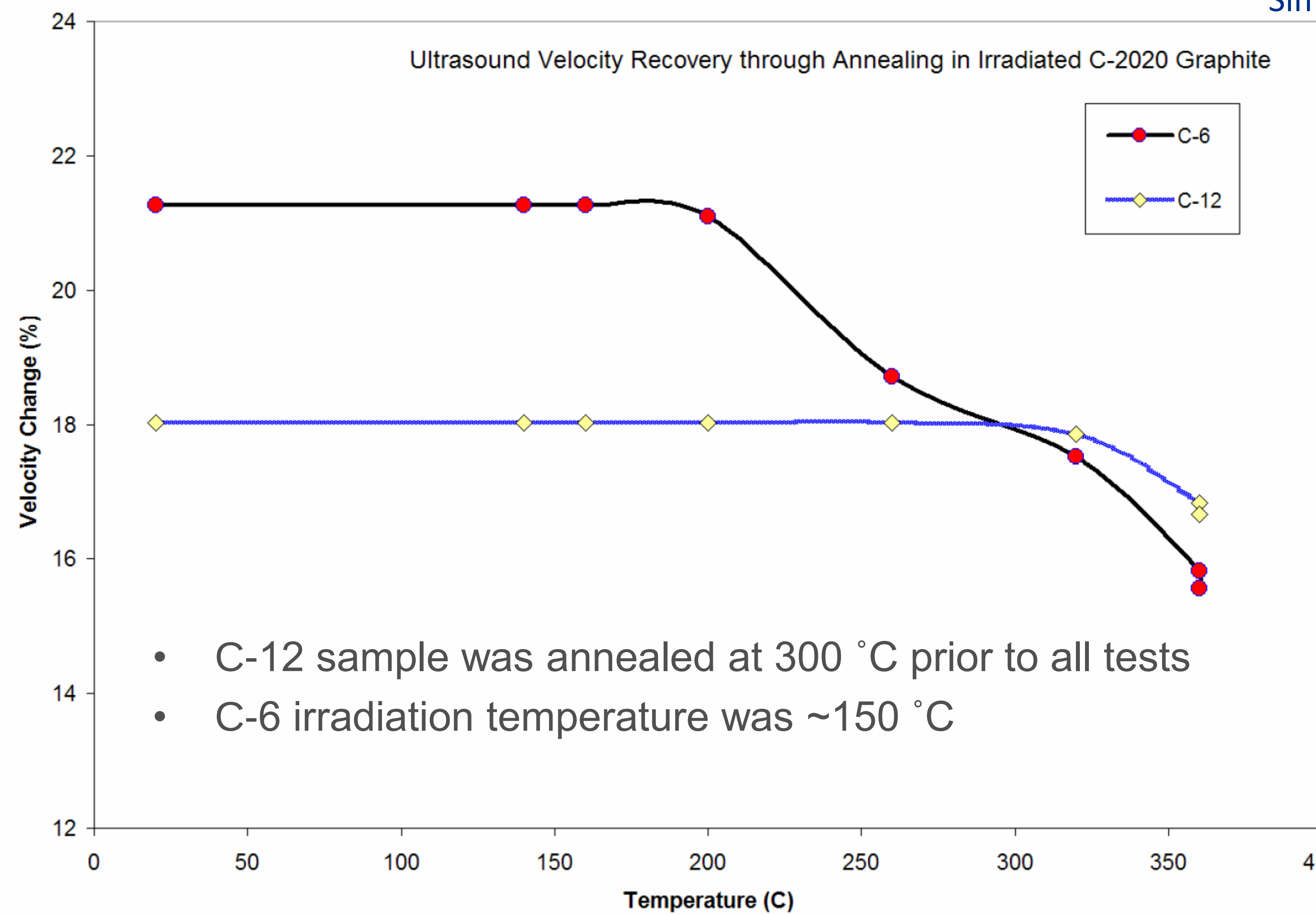
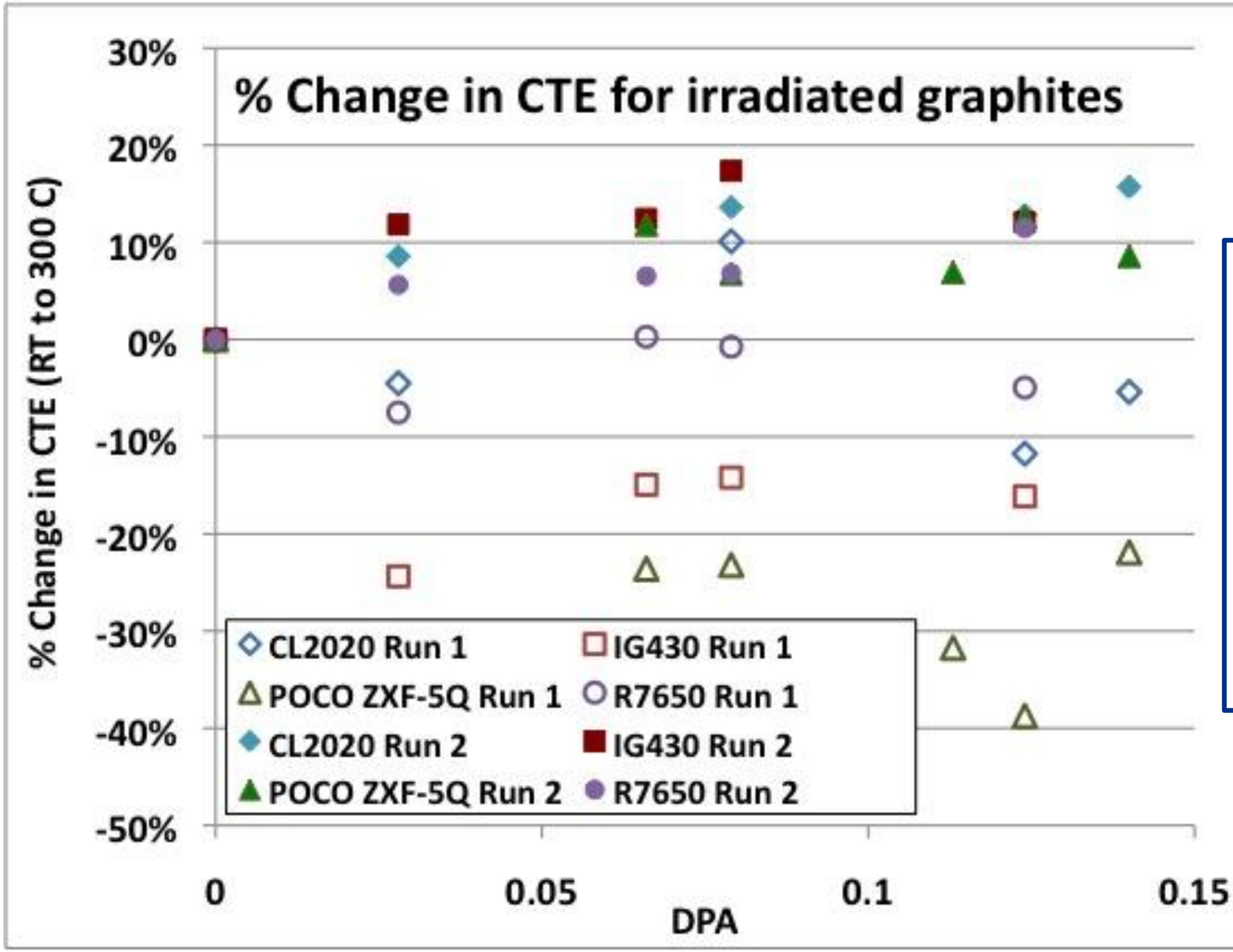


- C-12 sample was annealed at 300 ˚C prior to all tests
- C-6 irradiation temperature was ~150 ˚C

Figure 3.3.50: Ultrasound velocity recovery in irradiated/annealed Carbone-2020

Results – CTE and dimensional changes



- During 1st run annealing, specimens shrunk
- 2nd run, all graphites exhibited ~10% increase in CTE

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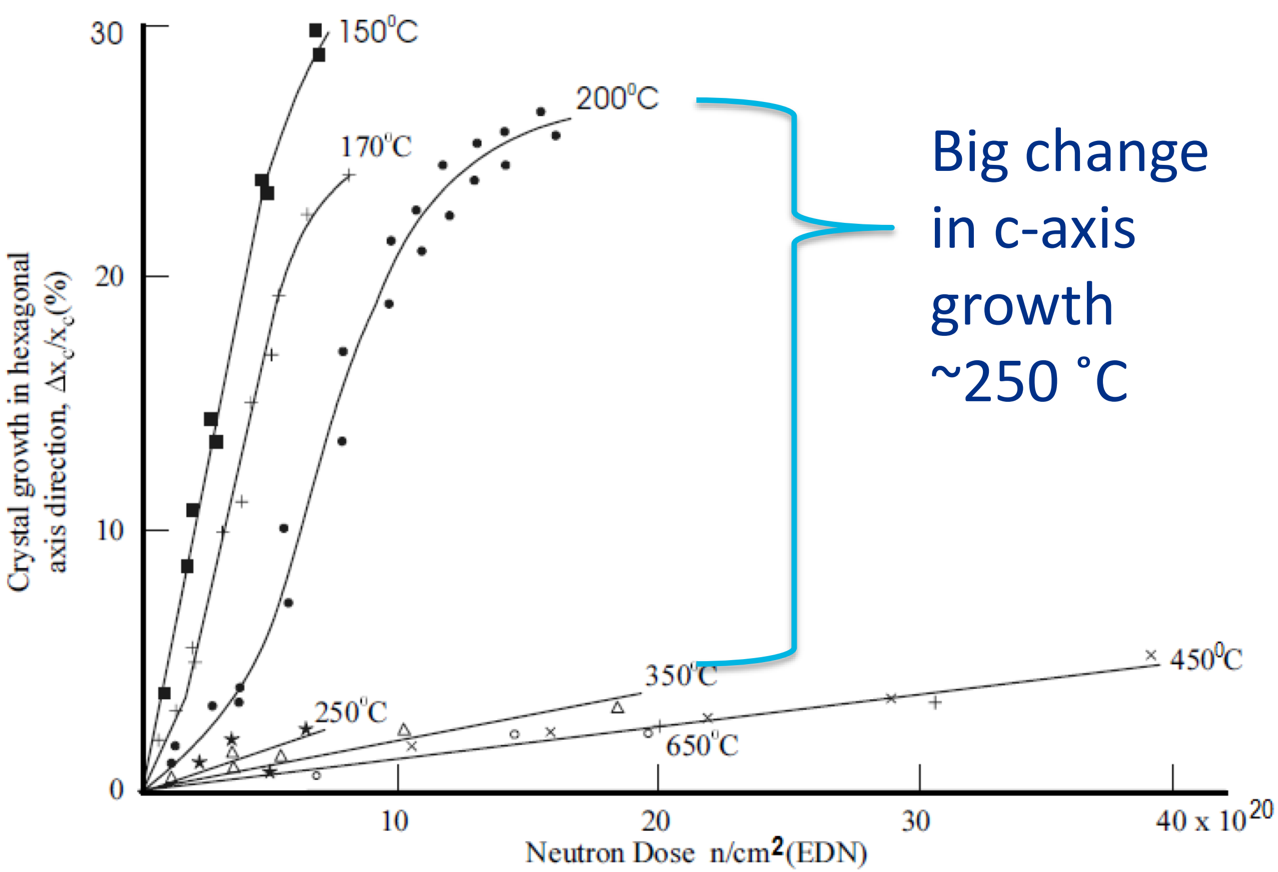
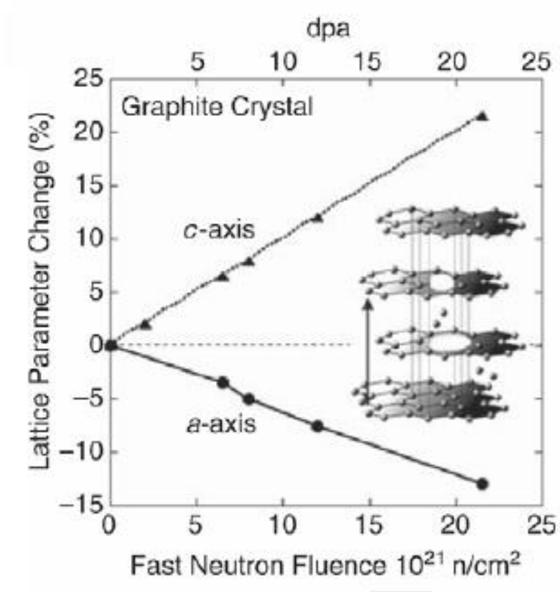
Neutron irradiated graphite dimensional changes

- B.J. Marsden, “Irradiation Damage in Graphite due to fast neutrons in fission and fusion systems,” IAEA-TECDOC-1154, 2000

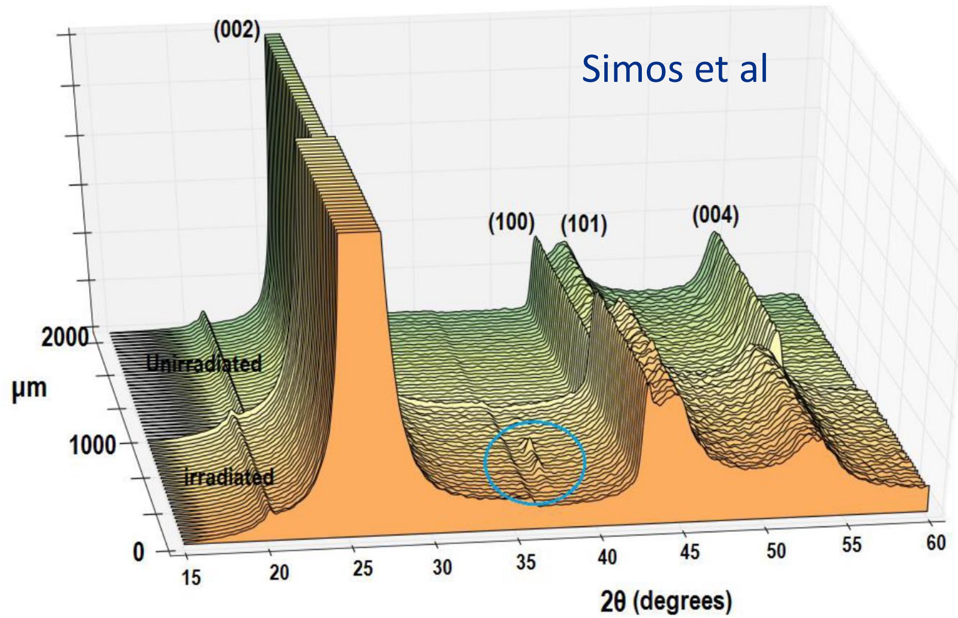


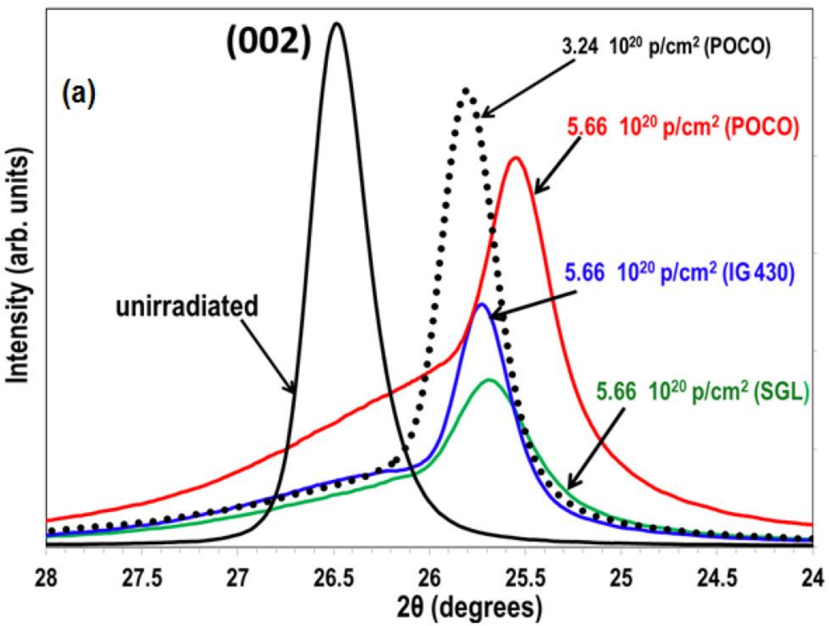
Big change in c-axis growth ~250 ˚C

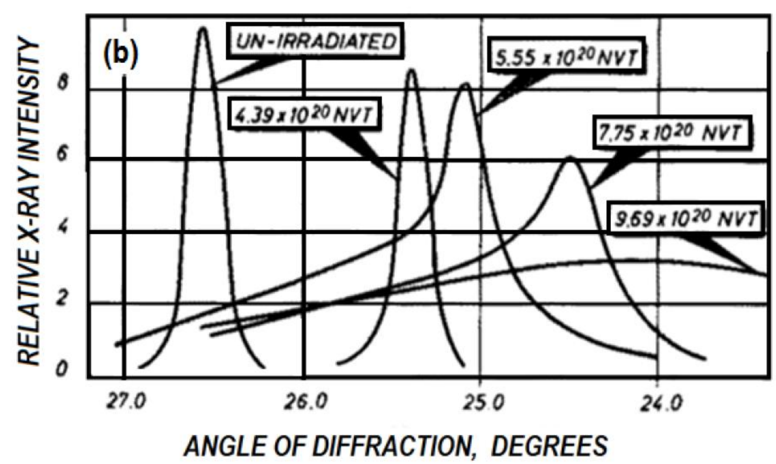


Results – X-ray diffraction

XRD on BLIP irradiated POCO graphite indicates agreement with c-axis lattice growth results from neutron irradiation







W. Bollmann. “Electron-microscopic observations on radiation damage in graphite” Phil. Mag., 5(54):621-624, June 1960.

NT-02 Graphite Fin Studies

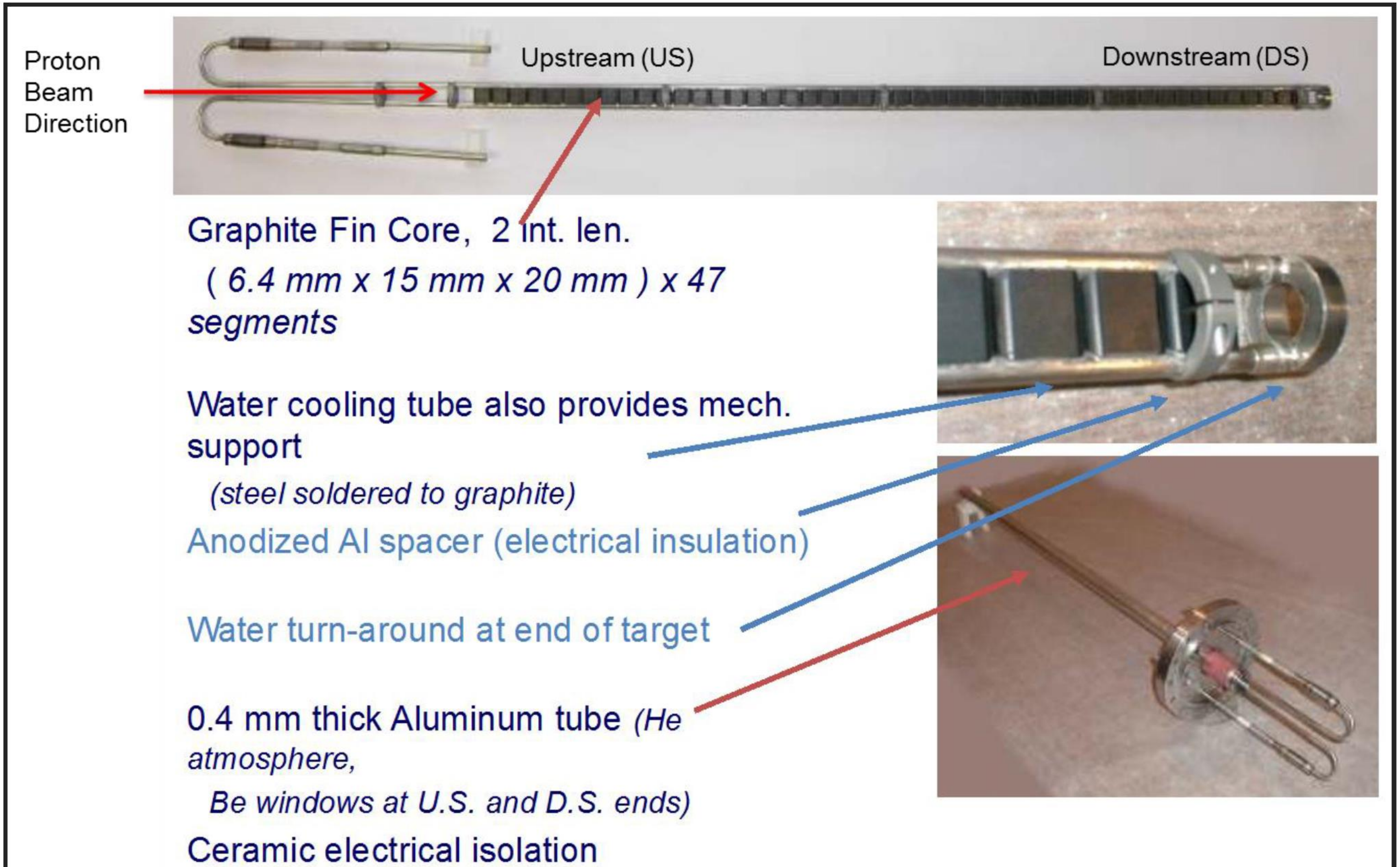


Figure 1.2. Description of the NT-02 target (Hylen 2009, Senor et al. 2016).

NT-02 Graphite Fin Fracture

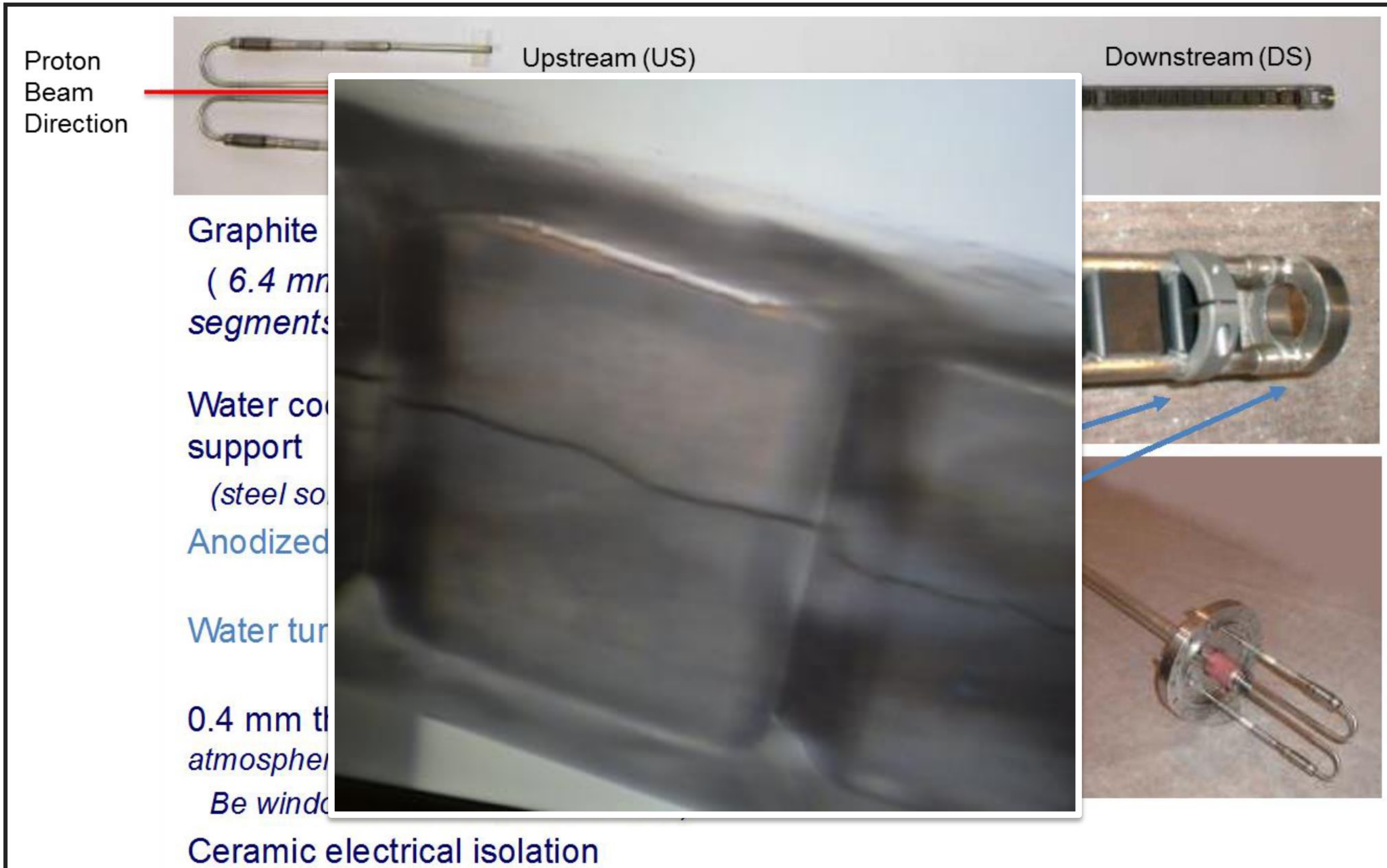


Figure 1.2. Description of the NT-02 target (Hylen 2009, Senor et al. 2016).

Results – NT-02 Evidence of Swelling

- Micrometer measurements revealed 2 – 4% swelling in the fin thickness in the beam center area
- TEM imaging did not show evidence of displacement damage (black spots, dislocation loops)

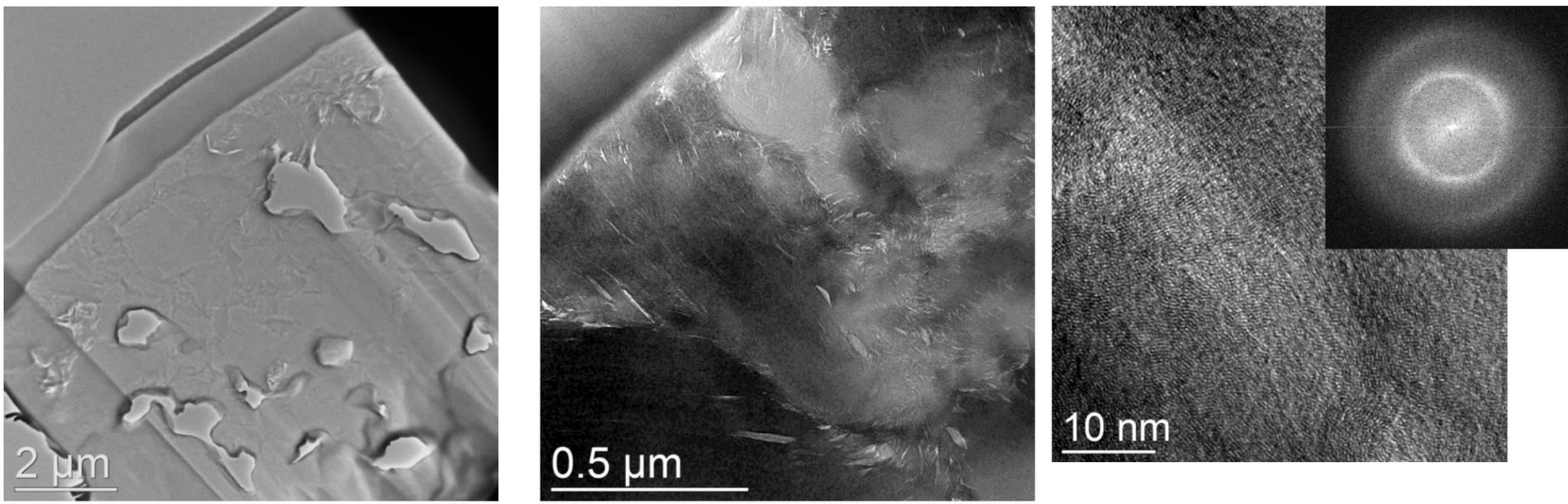
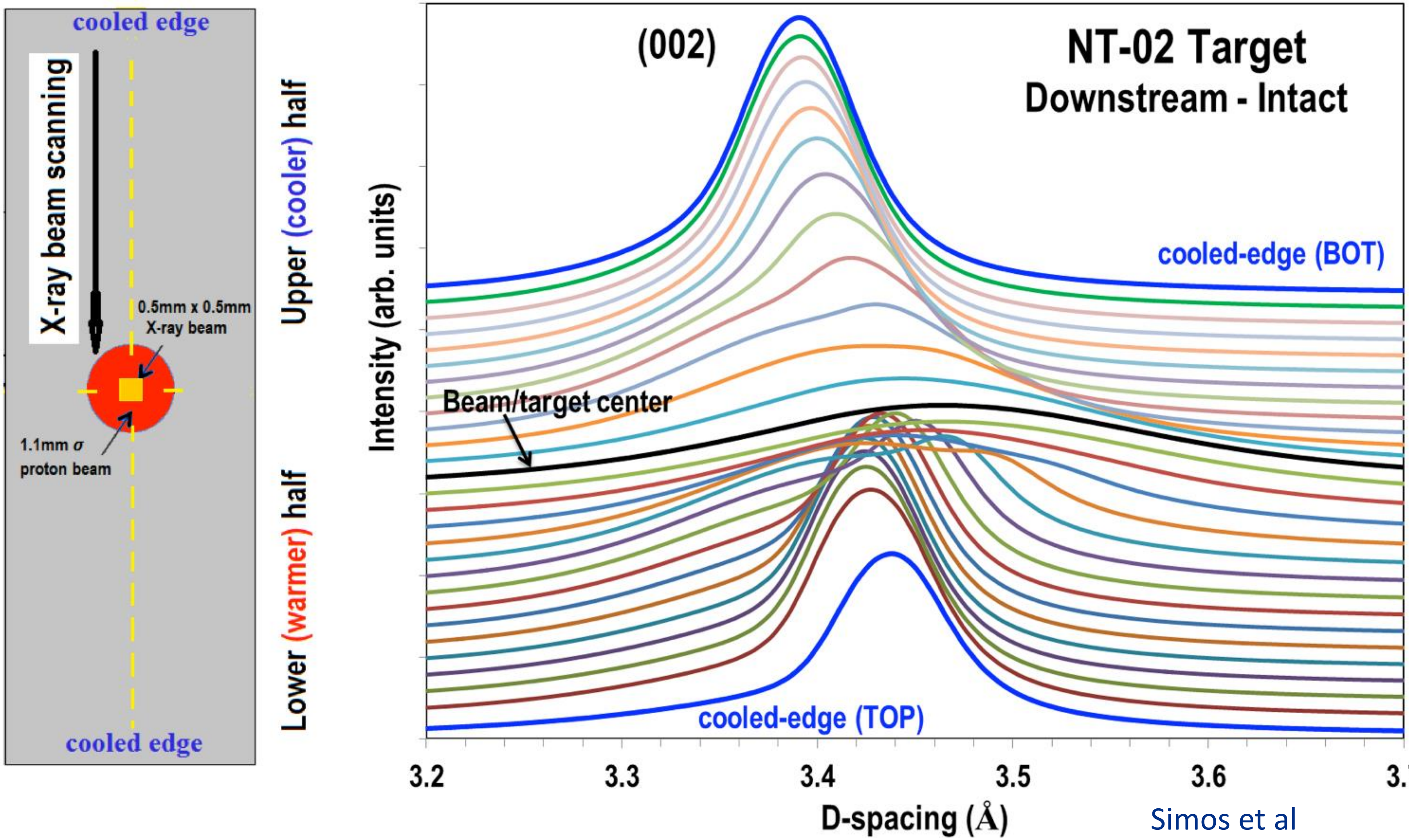


Figure 2.59. TEM imaging of DS ½ fin sample 16F017B. Overview of the sample shows porosity and a mix of nanocrystalline (darker) and amorphous (lighter) regions. Mrozowski cracks are prevalent in this sample, often at the interface between the two phases or in the nanocrystalline region.

Casella et al

Results – X-ray diffraction shows lattice growth and amorphitization at beam center



Simos et al

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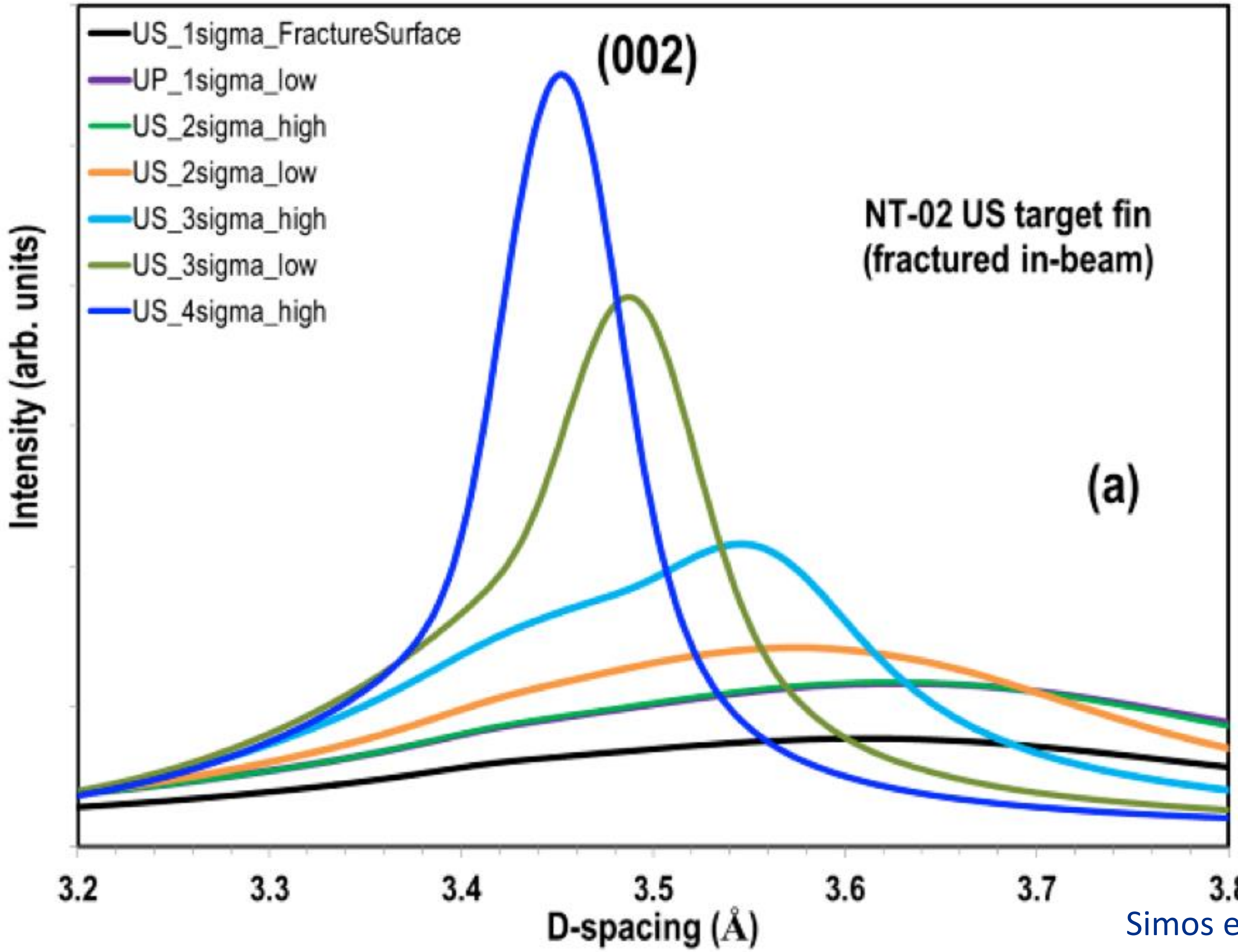

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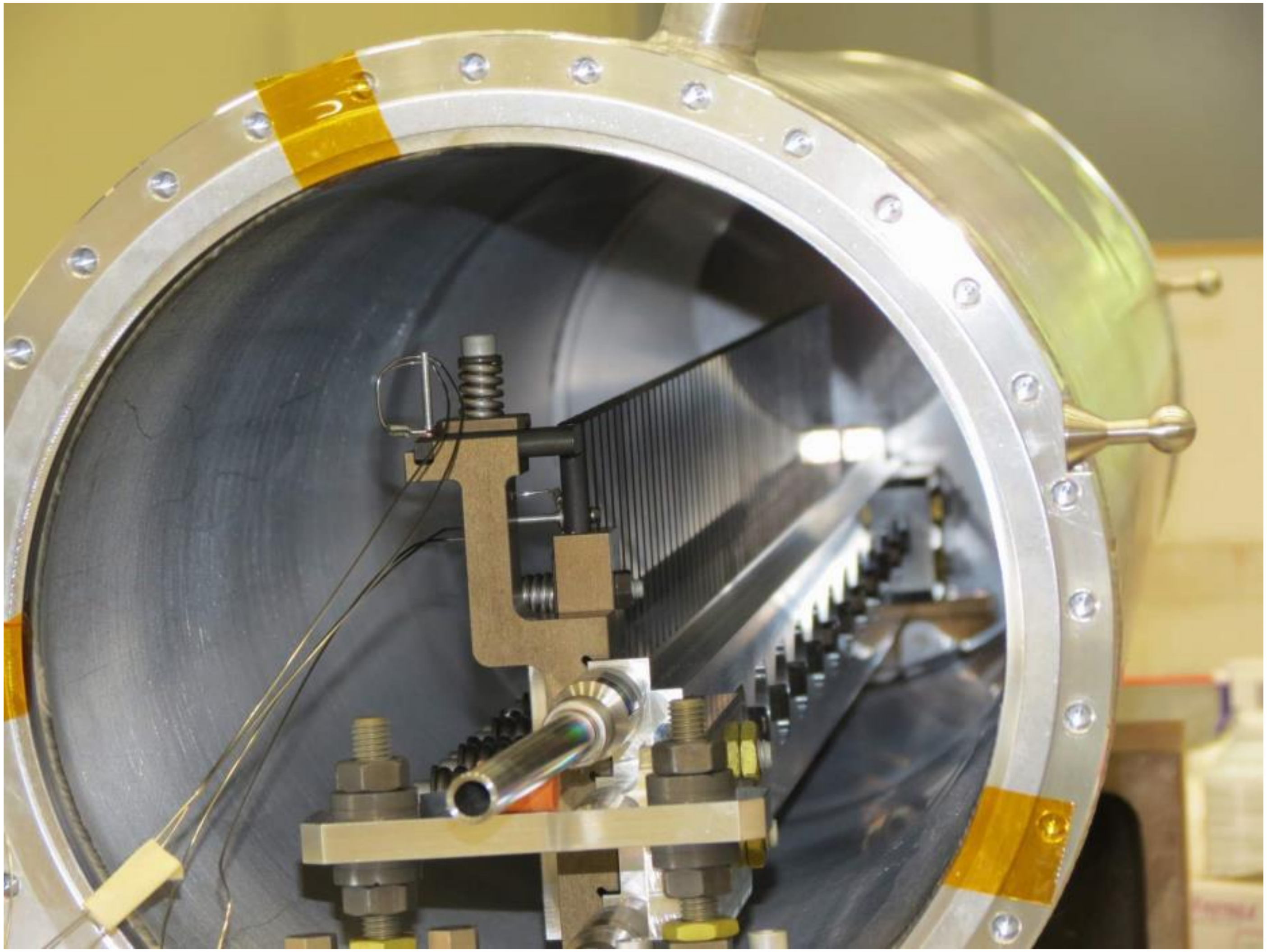
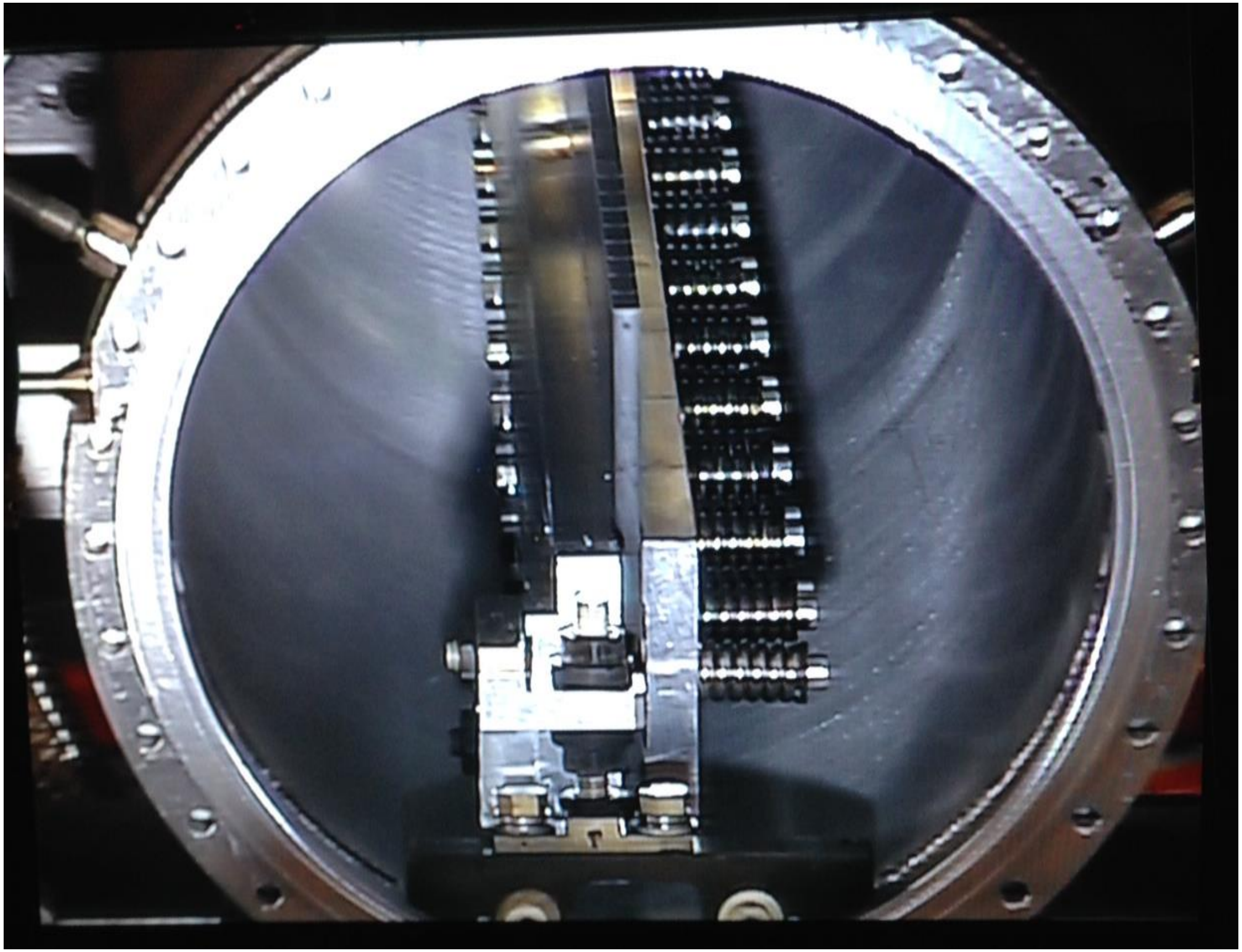
NOvA Target (MET-01) Autopsy



Before Irradiation (US end)

NOvA Target (MET-01) Autopsy



After Irradiation (DS end)

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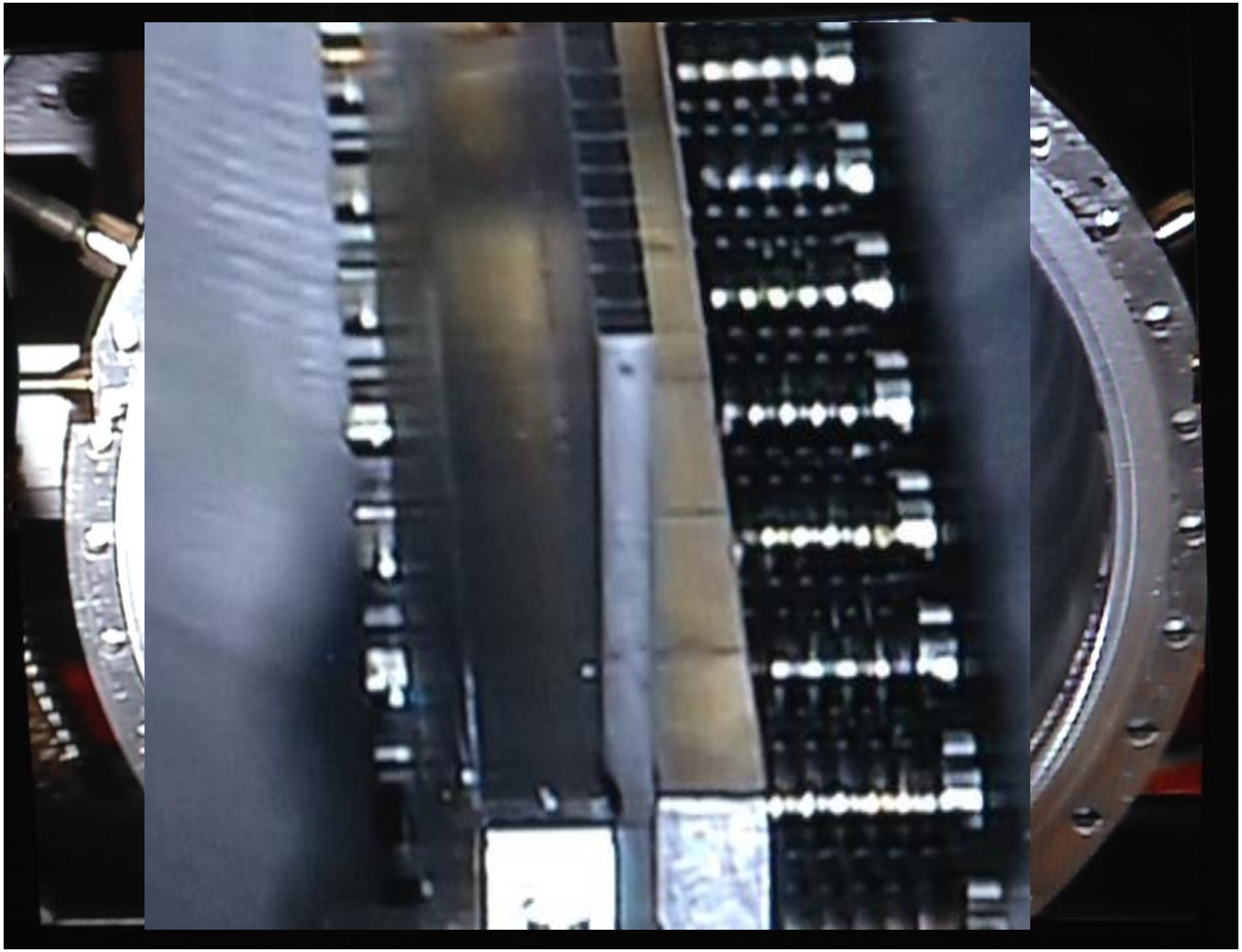
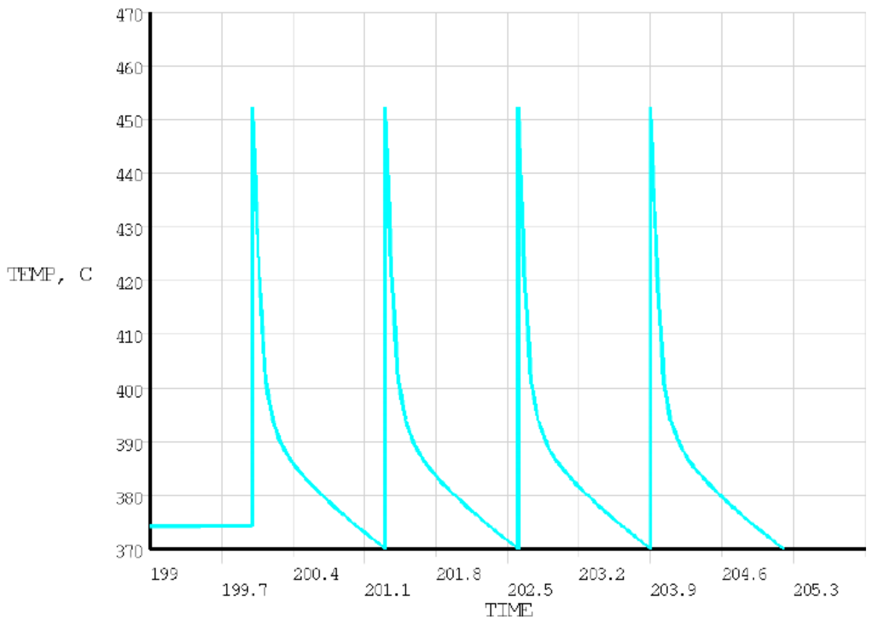
NOvA Target (MET-01) Autopsy

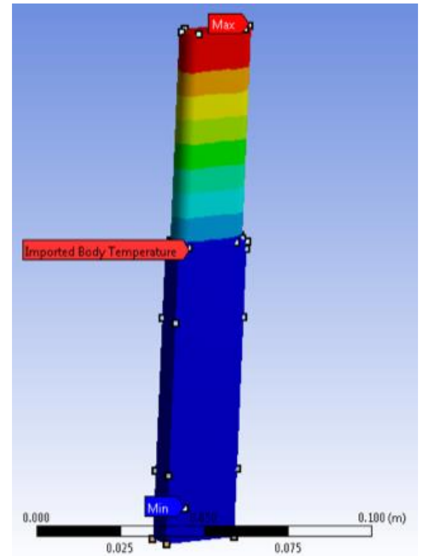


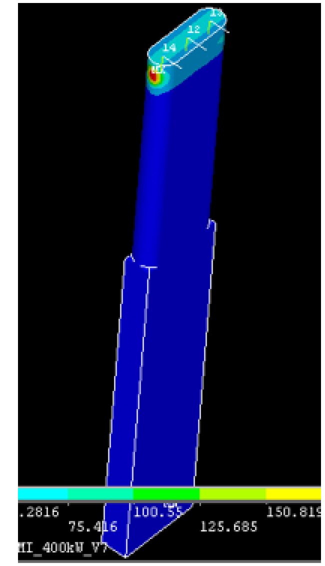
After Irradiation (DS end)

Sidorov et al

Thermal Comparison NT-02 to NOvA MET-01







Thermal Loading	NT-02 (350 kW)	MET-01 (550 kW)
Quasi-static Temp (˚C)	84	533
Peak Temp (˚C)	304	711
Time Average Mean (˚C)	139	578
Beam sigma (mm)	1.1	1.3

Davenne et al

Conclusion

- Significant changes in material properties with high energy proton irradiation at moderate temp (especially elastic modulus)
- High dependence upon irradiation/annealing temperature, especially for swelling (which exhibits a threshold at ~250 ˚C)
- No dislocation defects visible at dose up to 0.6 DPA and irradiation temperatures <~150 ˚C
- Failure of NT-02 graphite
  - Possibly swelling, internal stresses, loss of structure due to low temperature irradiation
  - Possibly oxidation or other contaminant
- Success of MET-01 graphite
  - Higher temperature irradiation
  - Better maintained quality of environment
- Future work
  - MET-01 and MET-02 PIE
  - Low energy ion irradiation to mimic high energy proton irradiation effects