Irradiation Effects on Graphite, C/C and Be

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Objectives

Damage assessment to graphite and other carbon-based structures from energetic protons and its reversal

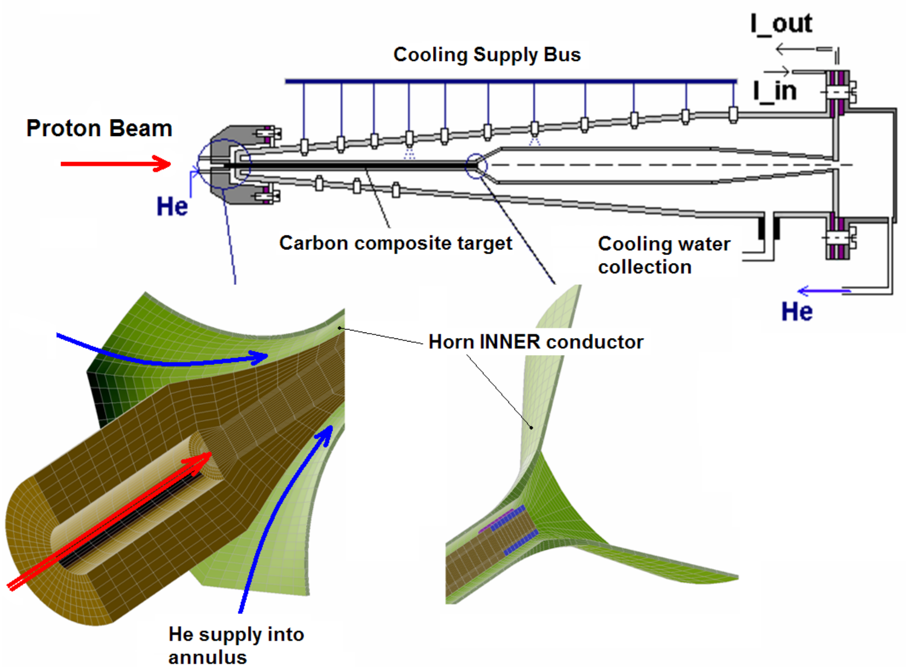
- Damage seen through dimensional stability/reversal and physio-mechanical property changes (strength, modulus, CTE, conductivity, etc.)
- Goals are the identification of the most radiation resistant/shock absorbent as well as the establishment of optimal operating temperature

Wealth of experience from reactor operations but still very intriguing lattice

Past studies and LBNE-related activities

Experience/study of Be (AlBeMet & h-BN)

Parenthesis: target-related background



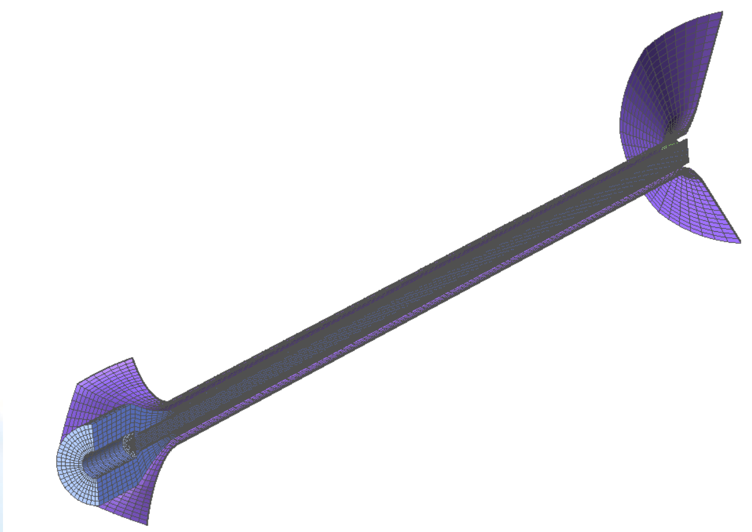
ESTIMATES OF HORN inner Conductor Heating

Joule Heat (conservative estimate) = 1.335 kW (for 2.5 Hz !!)

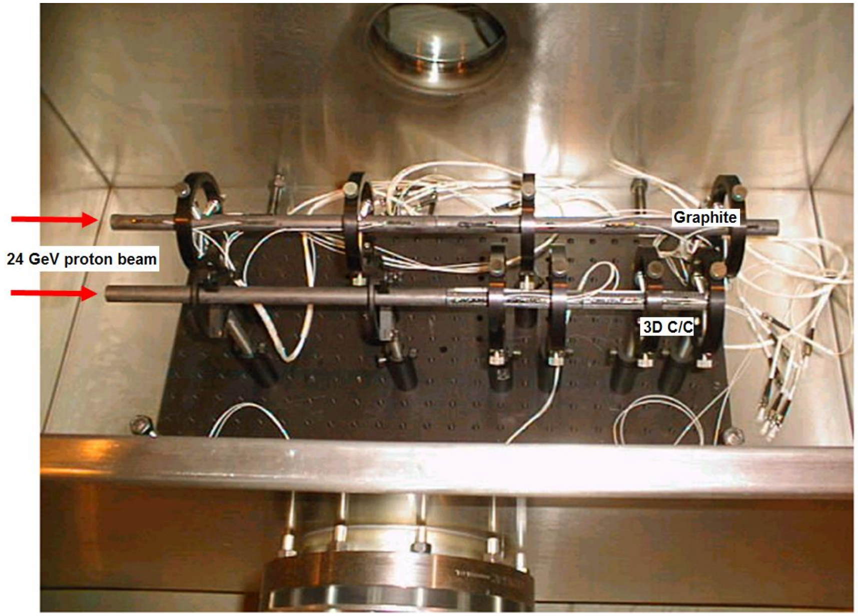
Heat from secondary particles = 10.3 kW

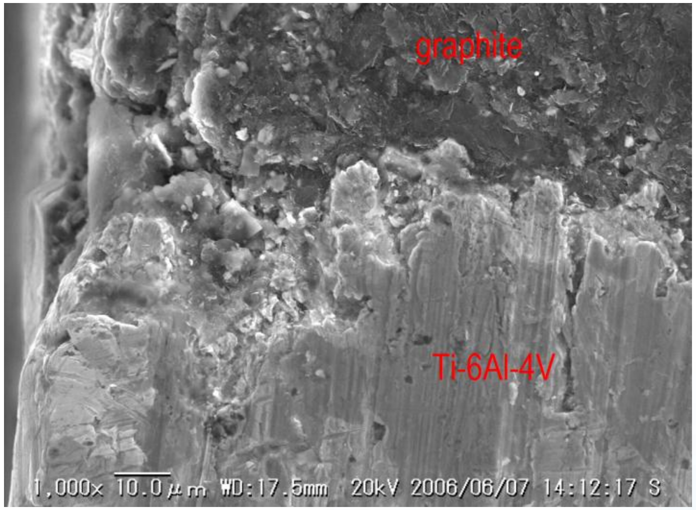
Radiation from target = 0.885 kW

TOTAL = 12.52 kW.

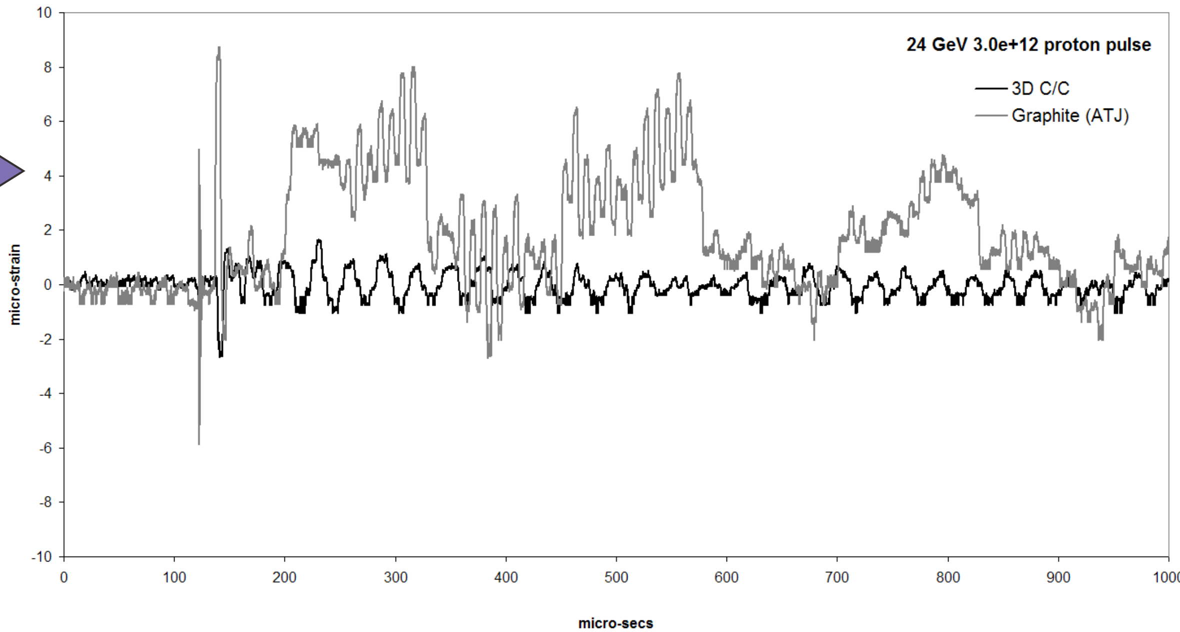


The removal of the generated heat using only the forced helium in the annulus, that is also cooling the target, high helium velocities will be required. Helium with inlet Temp of 144 K and with the surface temperature of the horn maintained at ~90 C, the required heat transfer film coefficient is 1624 W/m2-C requiring He velocities >150 m/s





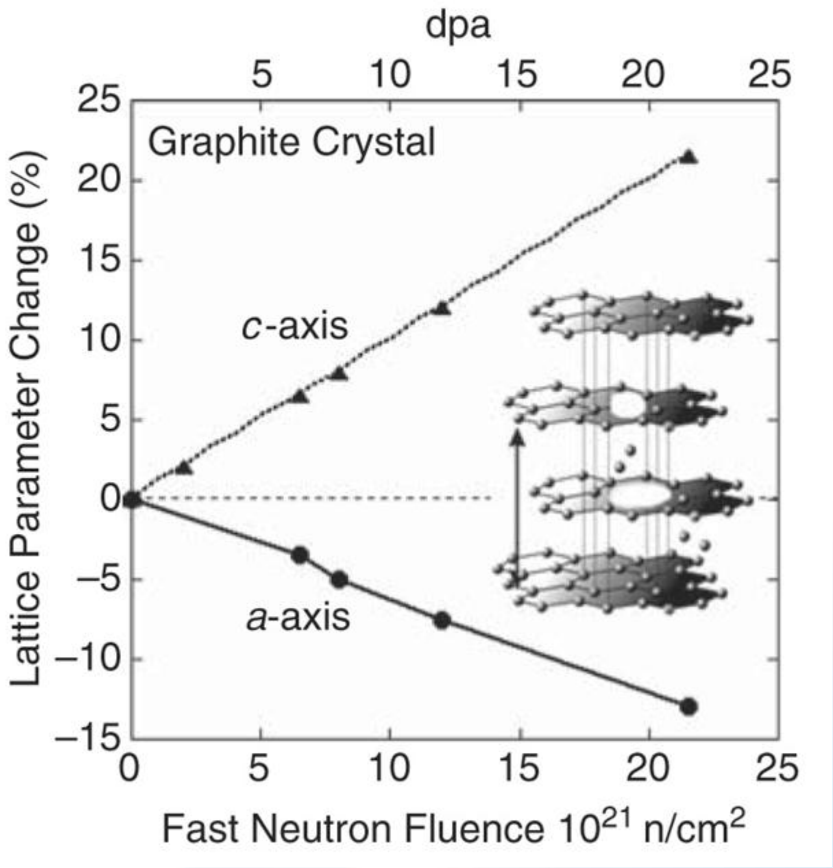
NOTE:
Using these graphite beam induced strain measurements and the results of irradiation (presented later) one can arrive at some realistic limits in terms of beam power that the target can sustain



Solid Targets – How far we think they can go?

1 MW ?	4 MW ?
Answer is **YES** for several materials **Irradiation damage is of primary concern** **Material irradiation R&D** pushing ever closer to anticipated atomic displacements while considering new alloys **is needed**	**Answer dependant on 2 key parameters:** 1 – rep rate 2 - beam size compliant with the physics sought A1: for rep-rate > 50 Hz **+** spot > 2mm RMS ➔ 4 MW possible (see note below) A2: for rep-rate < 50 Hz **+** spot < 2mm RMS ➔ Not feasible (ONLY moving targets) **NOTE:** While thermo-mechanical shock may be manageable, removing heat from target at 4 MW might prove to be the challenge. **CAN only be validated with experiments**

Graphite and Carbon Radiation Damage
What do we know?



After J-P Bonnal, A. Kohyama, L. Snead, MRS Bulletin, Vol. 34, 2009

Fission reactors (irradiation creep, low temperature irradiaton)
Accelerator targets (shock studies)

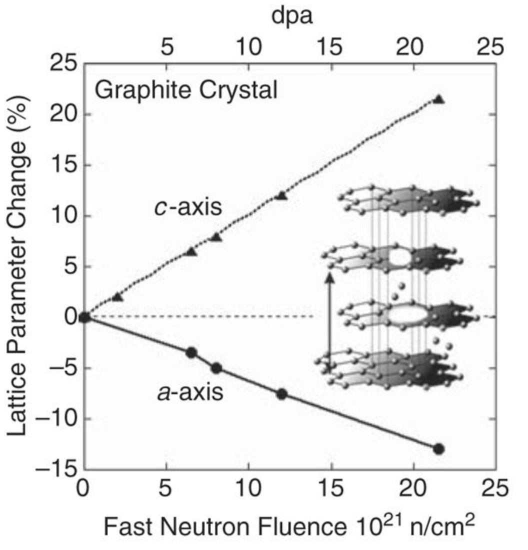
Graphite crystal and lattice (ordered basal planes or turbulent models, in the latter one observes the "effective" dimensional changes)

Unique structure ➔ interstitials, vacancies, activation energy and mobilization, Young's modulus and its partial recovery

Dimensional/volumetric changes (anisotropic) an important parameter that will cause high stresses in lattice (i.e. BeO) but graphite for up to some dose dilatation and shrinkage can be balanced!!

C/C composites - Graphite similarities and dissimilarities

Fibers dominated by basal graphite planes (that's why the high strength)



Graphite and Carbon Radiation Damage
Changes in the microstructure

For highly oriented pyrolytic graphite (crystal similarity)

Damage = lattice displacement and no difference between bombarding species should be small (that is good news so reactor experience can be utilized)

Early irradiation stages:

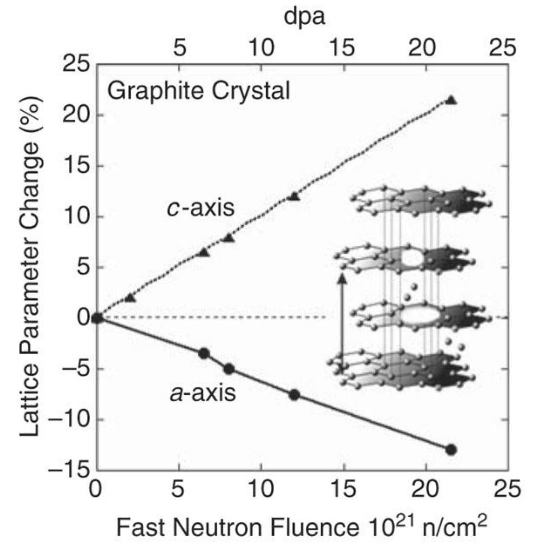
Defects nay not be limited to just vacancies and interstitials but also to changes in electronic structure ➔ changes in chemical nature (change in chemical bonding) ➔ significant increase of Young's modulus or hardness observed in irradiated graphite

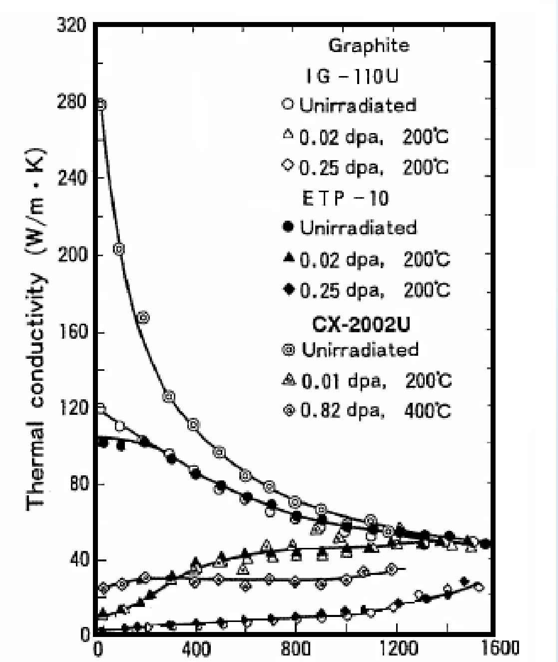
Higher irradiation ➔ turbulent basal plane structure/formation of 3D defect clusters (hard to recover with annealing)

see thermal conductivity in heavily irradiated due to the fact that conductivity is phonon conduction on basal planes

Other graphite grades are not as highly oriented in their microstructure

Radiation effects, especially dimensional are more "effective" than along a given direction (c, or a)

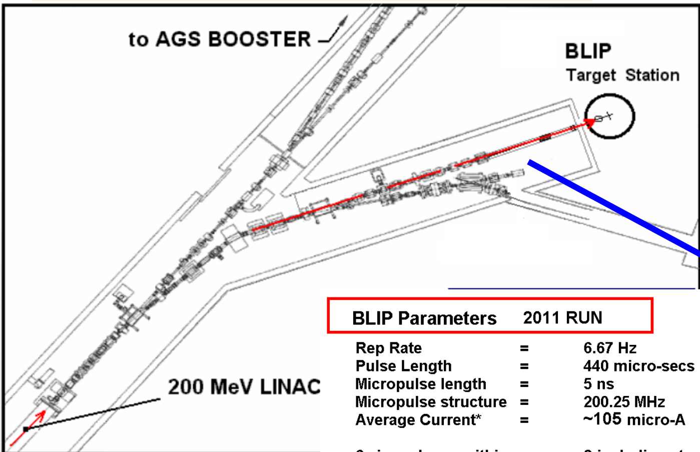




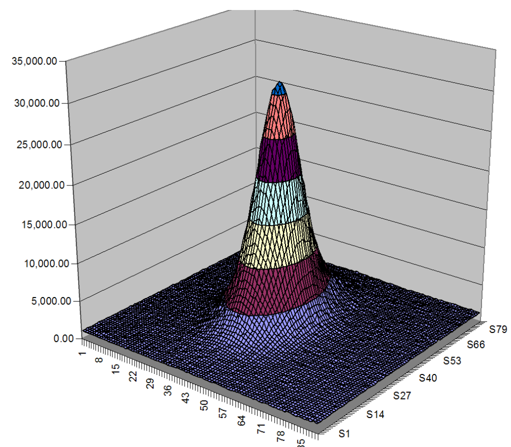
After T. Tanabe, Physica Scripta, Vol. T64, 7-16, 1996

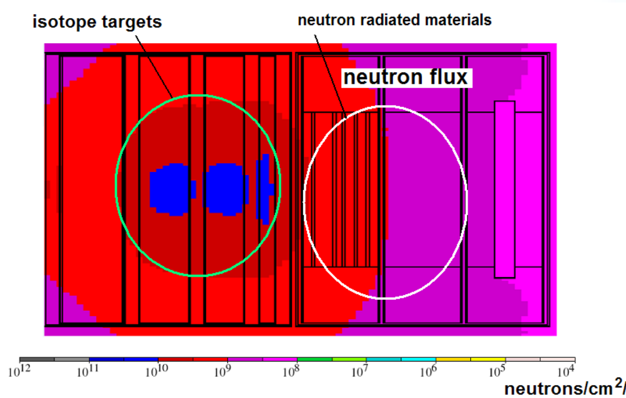
Irradiation at the BNL Accelerator Complex

Irradiation at BLIP
(up to 200 MeV or spallation neutrons from 112 MeV protons)

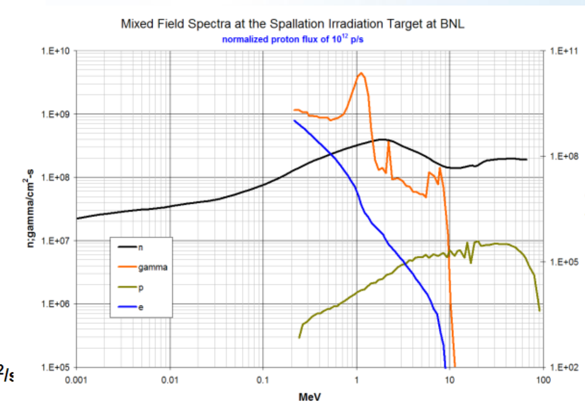


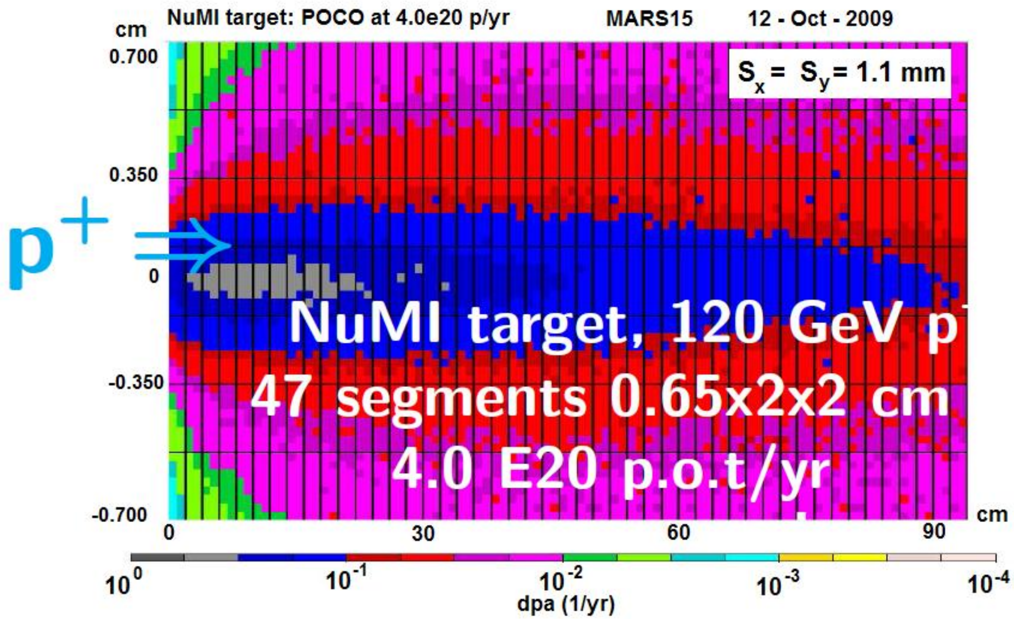
BLIP Parameters		2011 RUN
Rep Rate	=	6.67 Hz
Pulse Length	=	440 micro-secs
Micropulse length	=	5 ns
Micropulse structure	=	200.25 MHz
Average Current*	=	~105 micro-A
6 sigma beam within	=	2-inch diameter
Beam Gaussian	==>	1 sigma = 4.233 mm





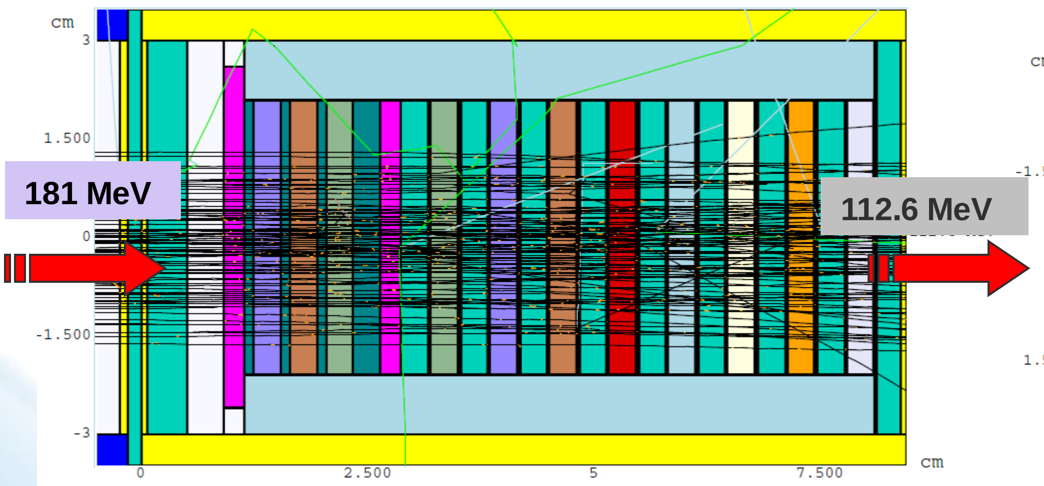
NORMALIZED neutron flux
at BLIP target station (by N. Mokhov, FNAL)

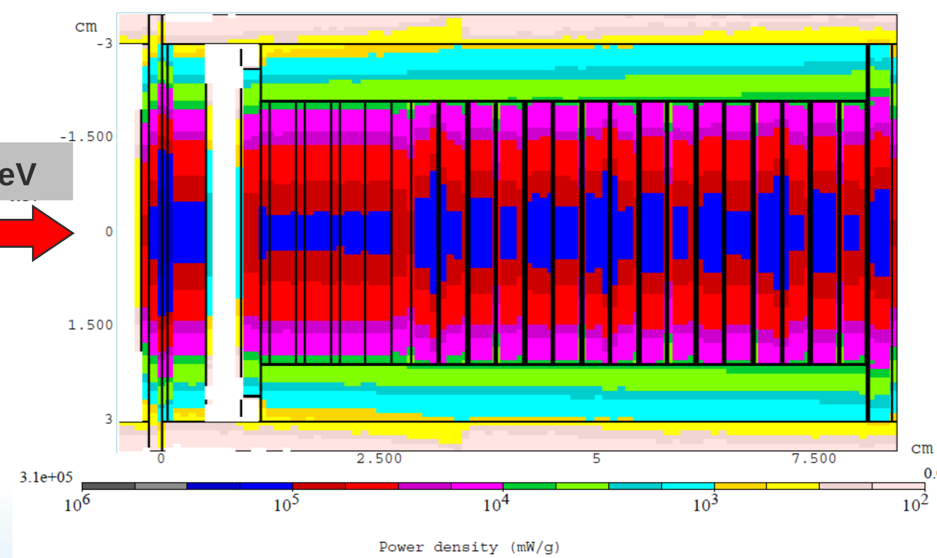


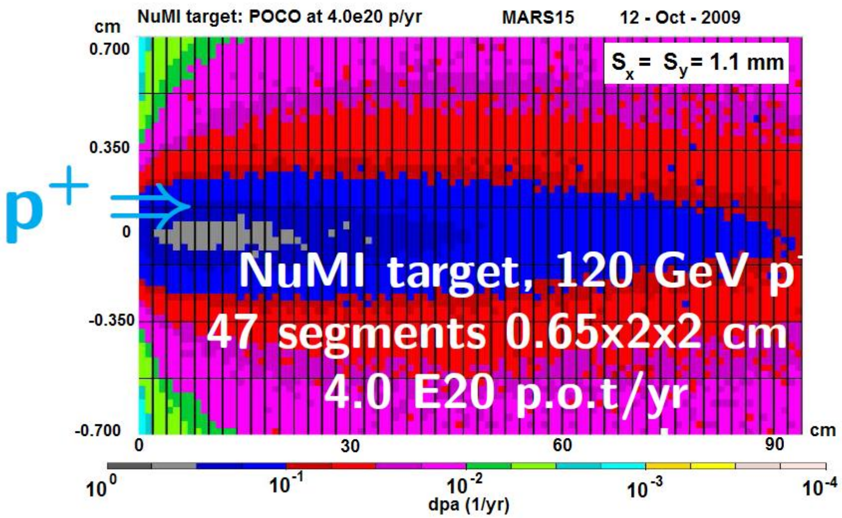


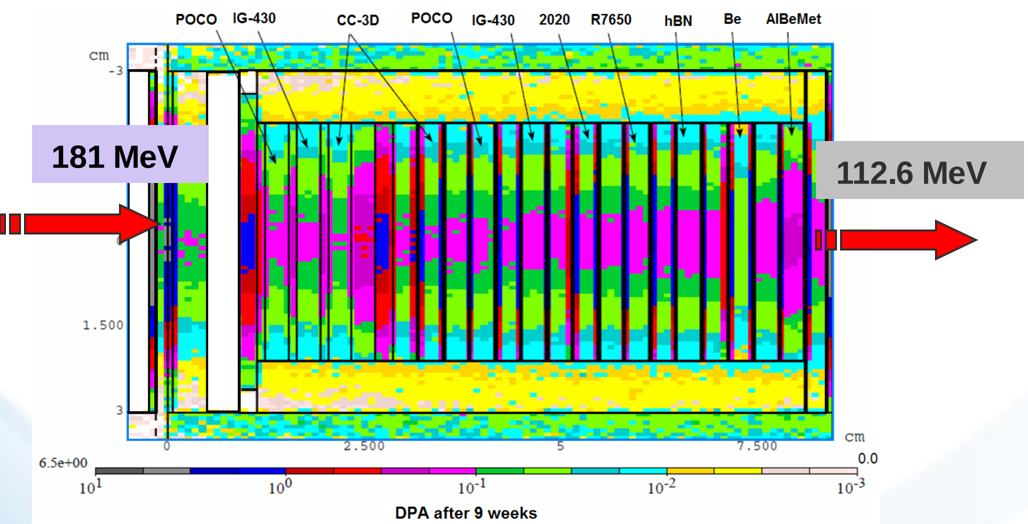
NuMI target simulation with MARS-15

BNL_BLIP target simulation with MARS-15









MARS-15 analysis confirmed what has been anticipated/observed in the various studies prior, that damage (dpa portion) is greater at the lower energies

Physics process contribution (%) at beam axis:
z=15 cm (NuMI) and Box 2 POCO graphite (BLIP)

Target	Nuclear	EM elastic	L.E. neutrons	$e^{\pm}$
NuMI	50.8	43.3	1.5	4.4
BLIP	43.5	53	3.5	0.02

Target	E_p (GeV)	Beam σ (mm)	N_p (1/yr)	DPA (1/yr)
NuMI/LBNE	120	1.1	4.0e20	0.45
BLIP	0.165	4.23	1.124e22	1.5

Graphite

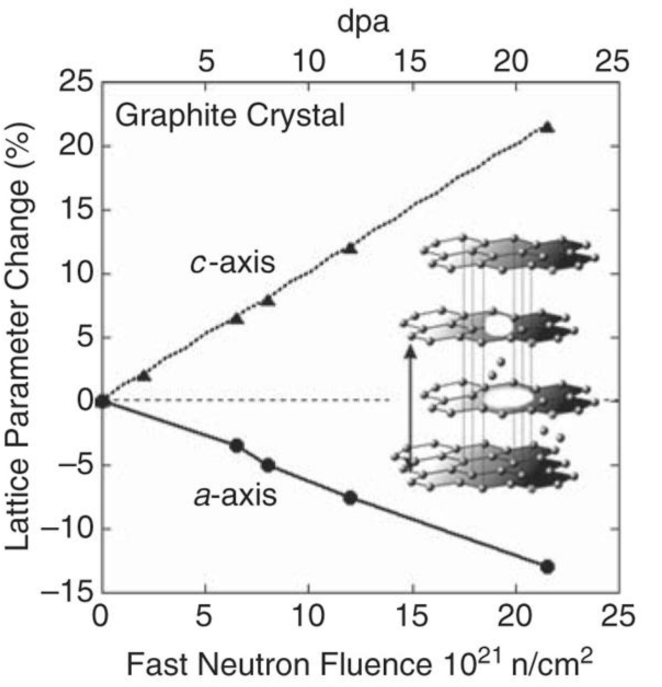
Irradiation Damage & Annealing prompted by LBNE Target Interest

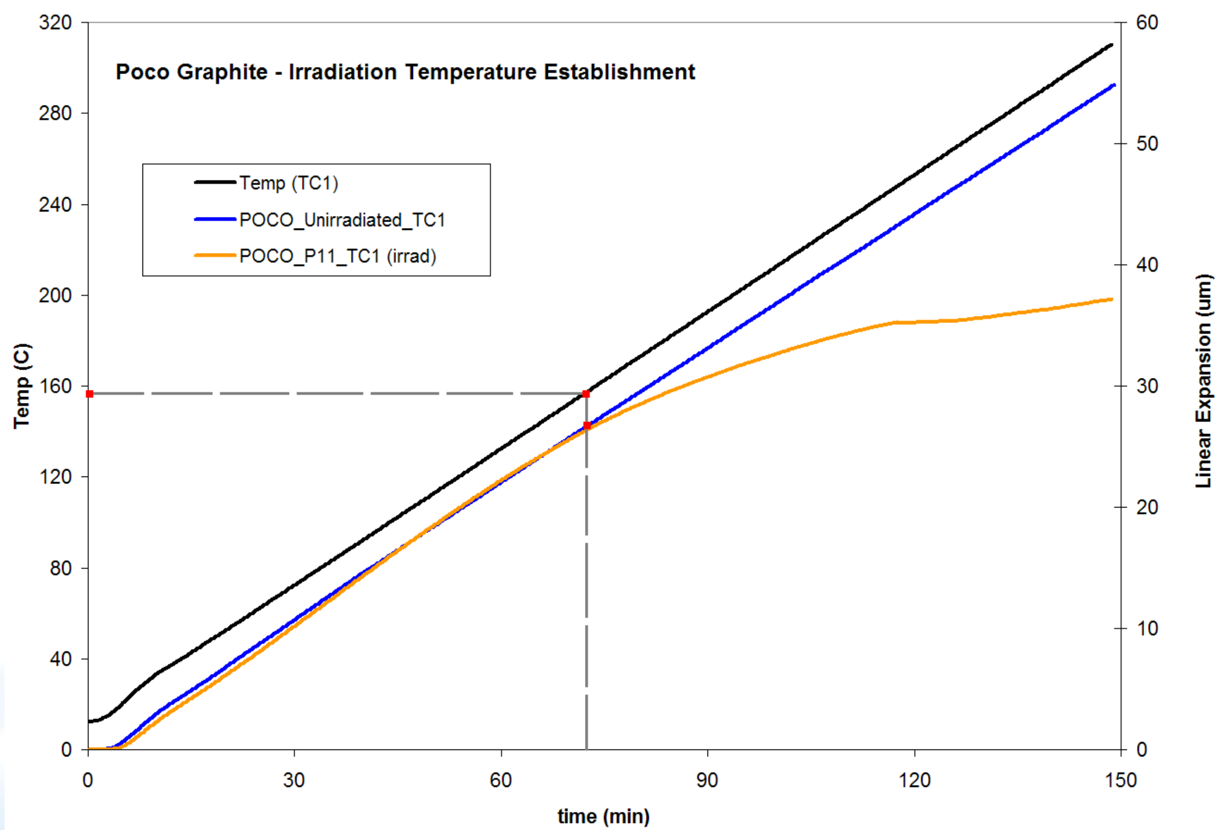
Material	Motivation
C-C Composite (3D)	Observed damage at low dose at BNL BLIP
POCO ZXF-5Q	NuMI/NOvA target material
Toyo-Tanso IG-430	Nuclear grade for T2K
Carbone-Lorraine 2020	CNGS target material
SGL R7650	NuMI/NOvA baffle material
St.-Gobain AX05 h-BN	Hexagonal Boron Nitride

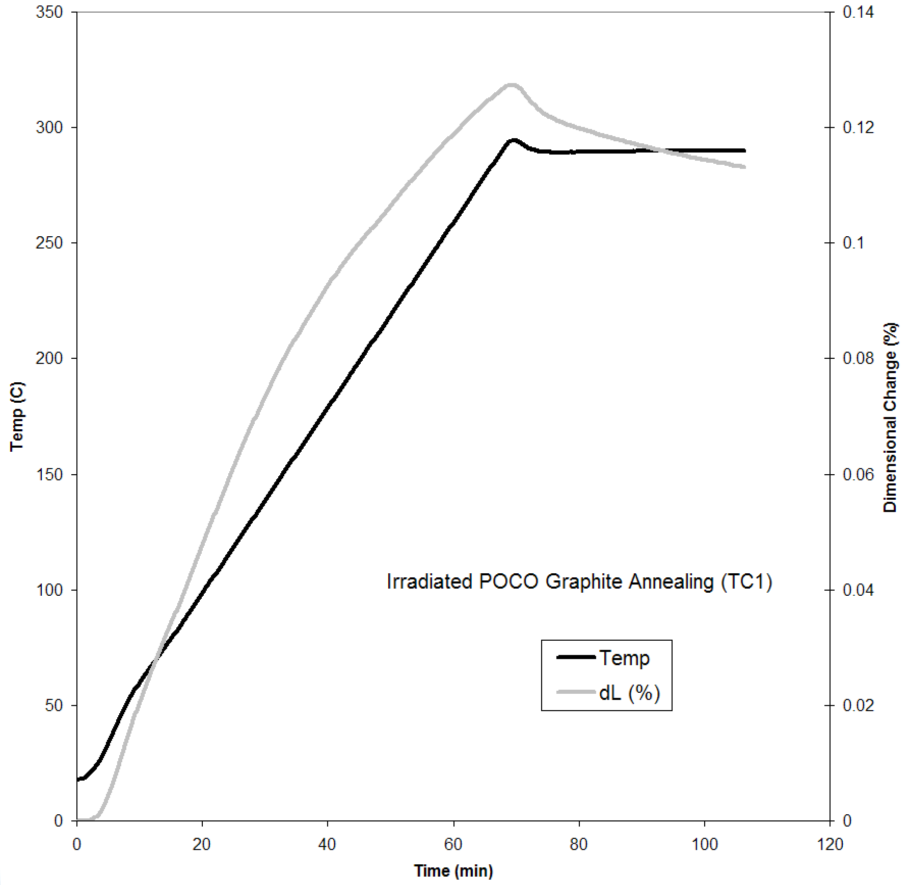
When high energy neutrons collide with graphite atoms the rate of displacement is flux-dependent and independent of the lattice temperature. (D. Switzer, BNL, from Physical Review “Activation energy for annealing single interstitials in neutron irradiated graphite ..”)

A displaced interstitial will undergo many collisions until its energy is reduced to values corresponding to lattice temperature. In process some interstitials remain in stable configuration and some anneal immediately. Those that do not anneal cause an increase in the dimensions of the sample.

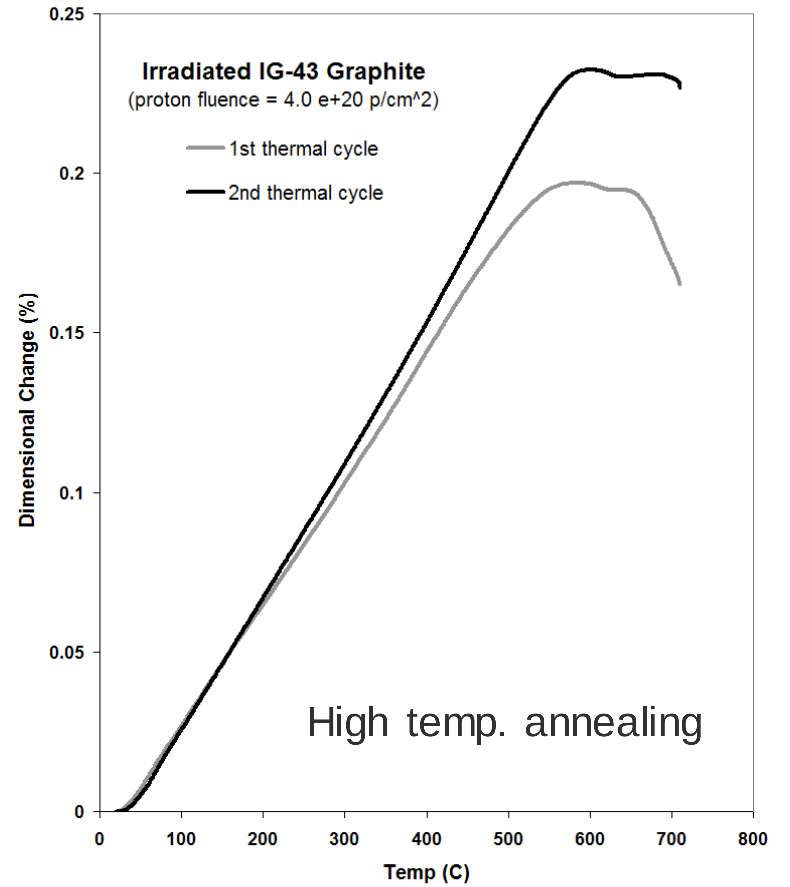
As shown here, when annealing above the irradiation temperature the dimensional change dips because more “stable” interstitials are leaving the temporary locations between lattice planes and return to them.



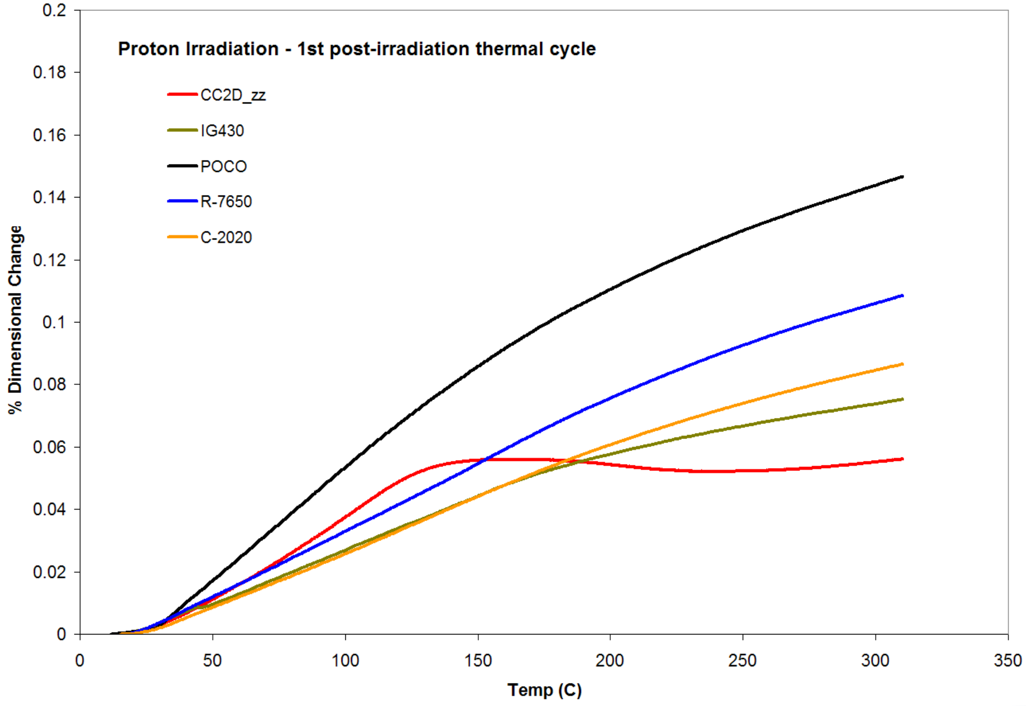


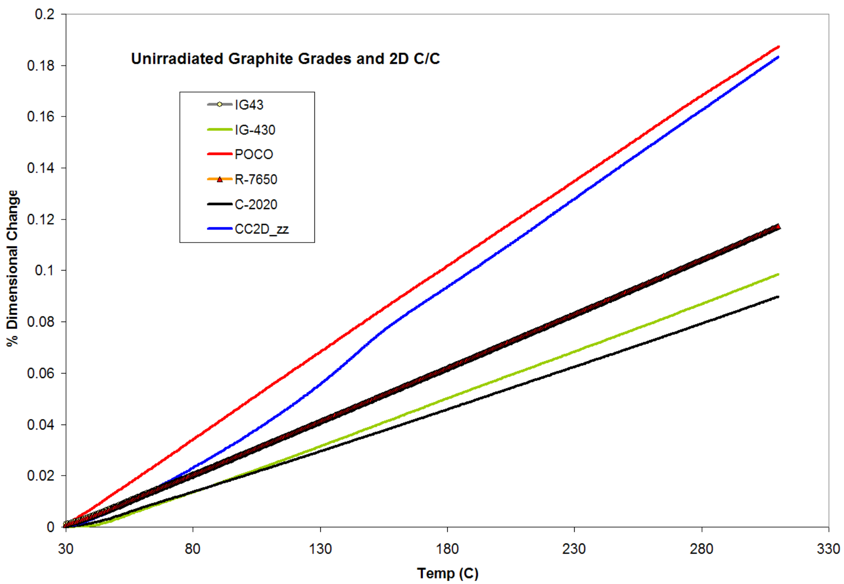


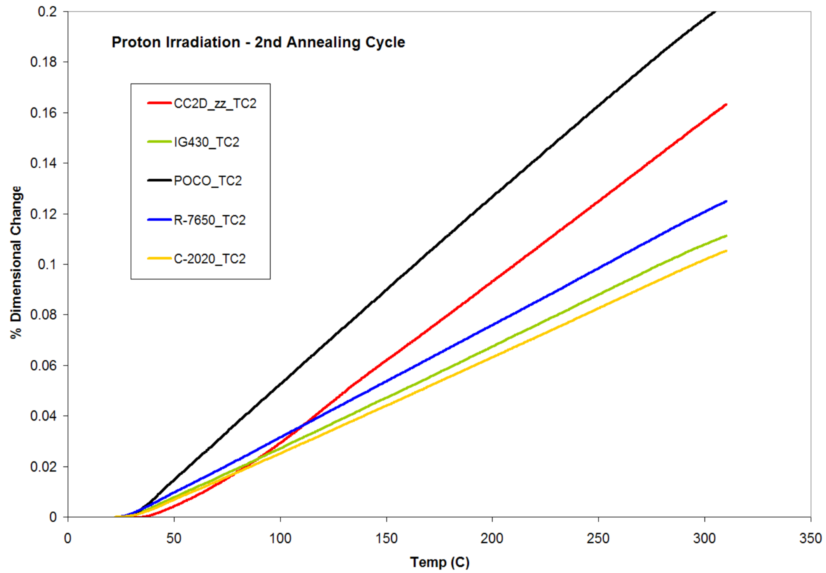
Irradiated POCO Graphite Annealing (TC1)
Temp
dL (%)
Temp (C)
Dimensional Change (%)
Time (min)

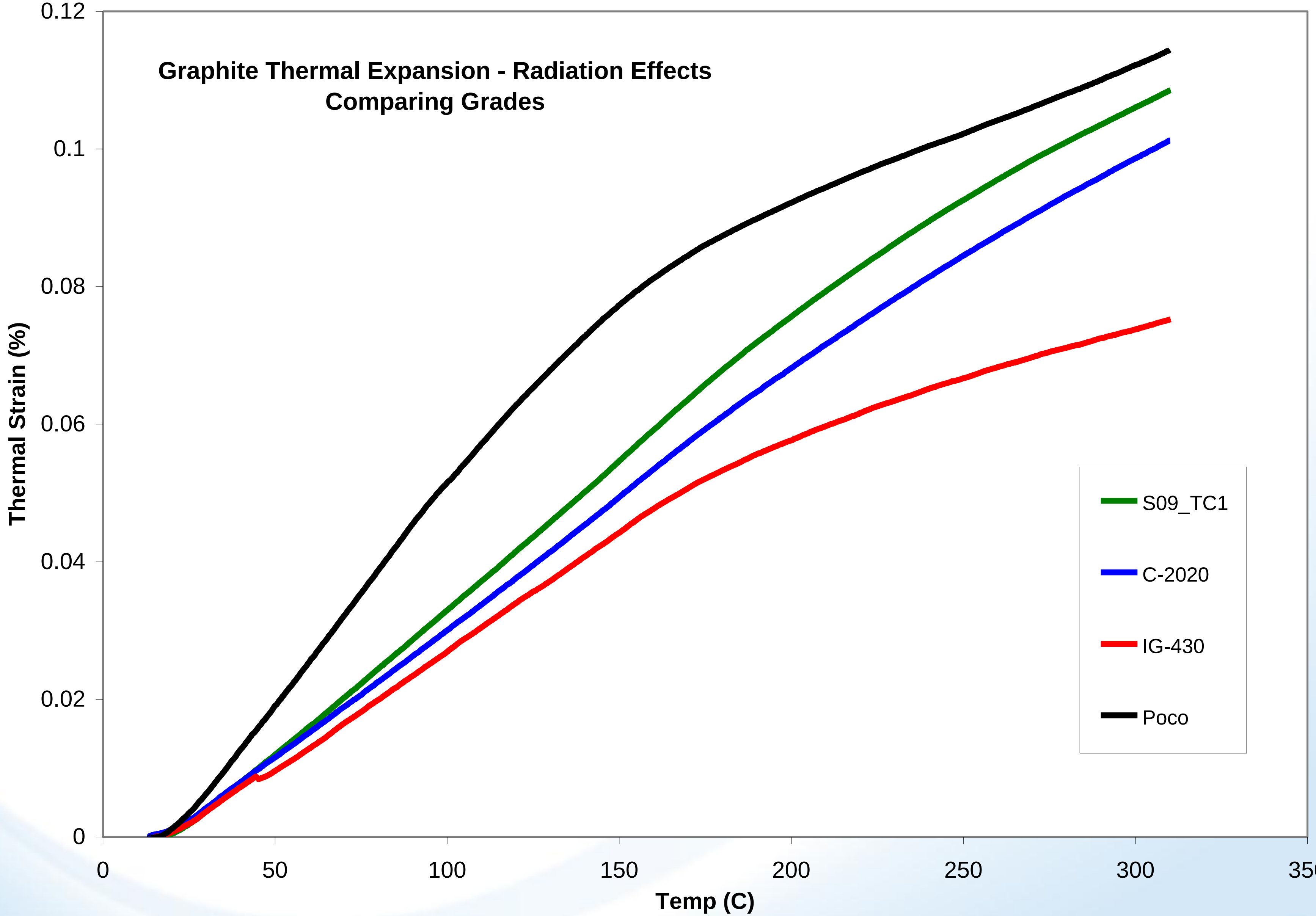


Irradiated IG-43 Graphite
(proton fluence = 4.0 e+20 p/cm^2)
1st thermal cycle
2nd thermal cycle
Dimensional Change (%)
Temp (C)
High temp. annealing

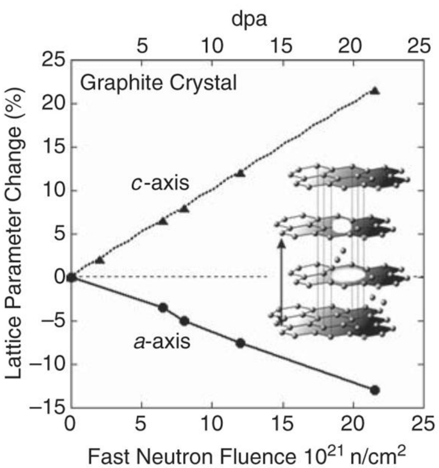

Proton Irradiation - 1st post-irradiation thermal cycle
CC2D_zz
IG430
POCO
R-7650
C-2020
% Dimensional Change
Temp (C)

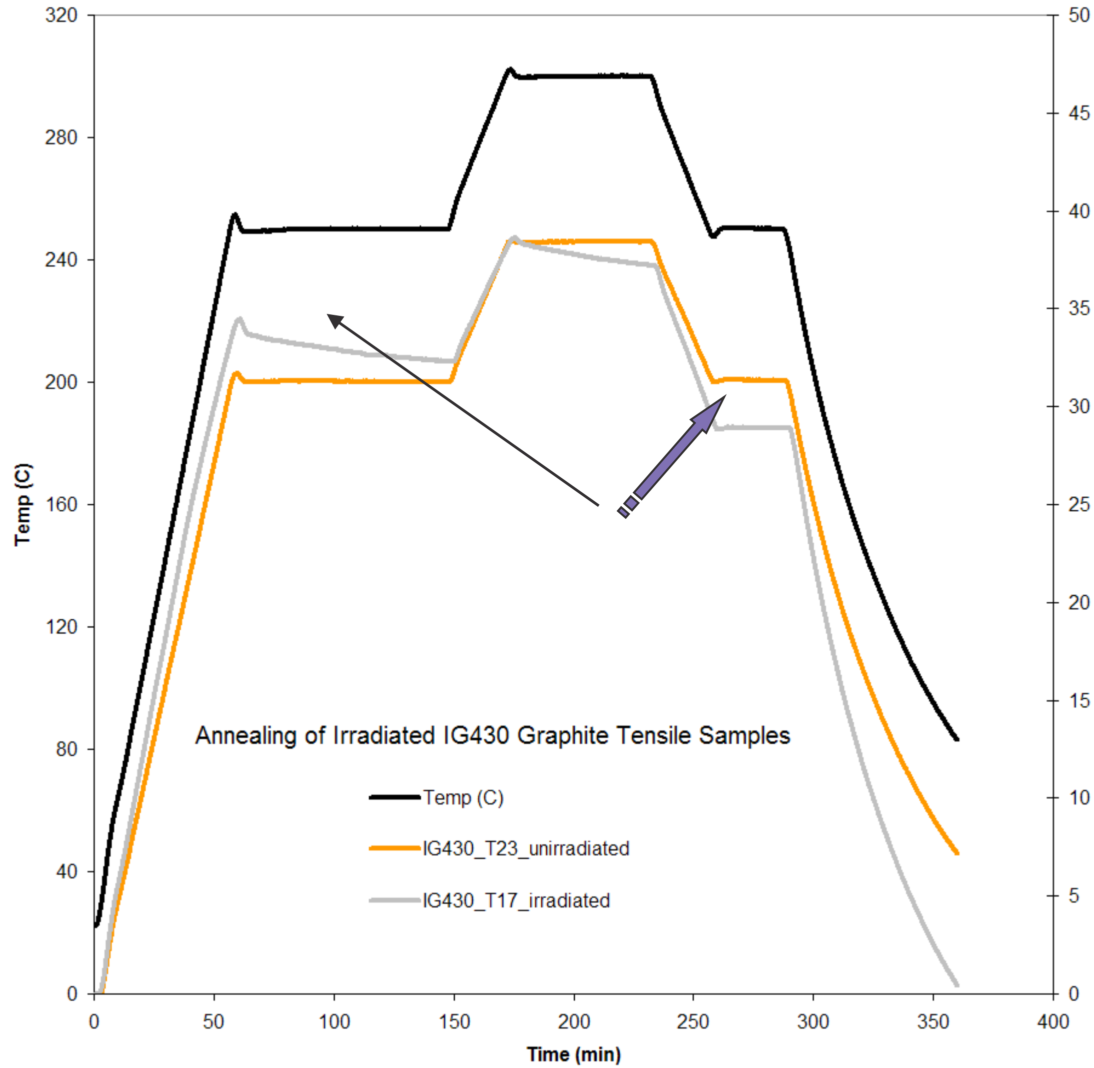

Unirradiated Graphite Grades and 2D C/C
IG43
IG-430
POCO
R-7650
C-2020
CC2D_zz
% Dimensional Change
Temp (C)

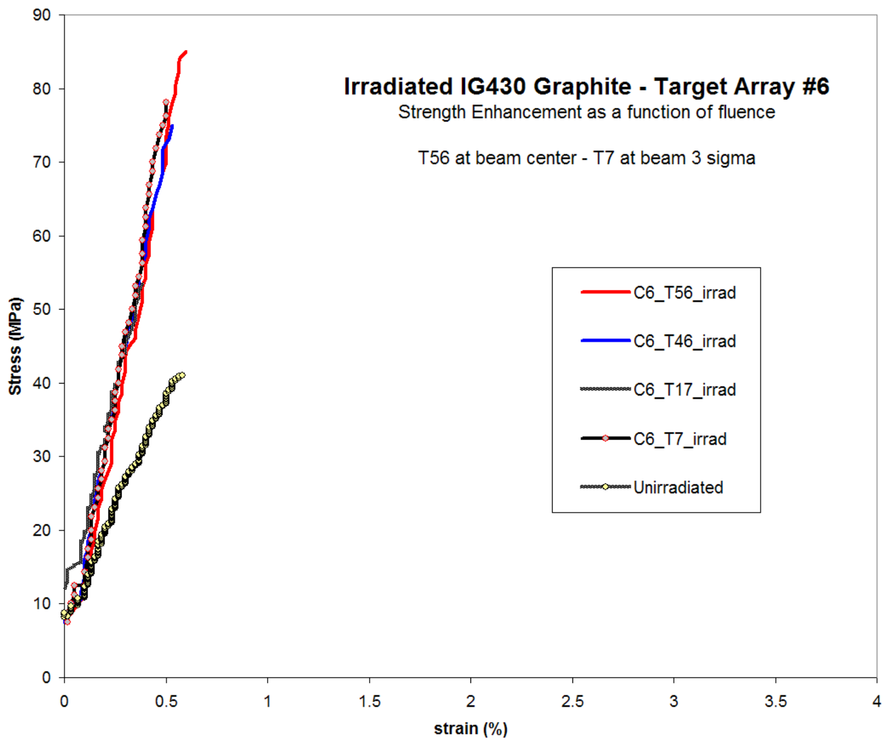

Proton Irradiation - 2nd Annealing Cycle
CC2D_zz_TC2
IG430_TC2
POCO_TC2
R-7650_TC2
C-2020_TC2
% Dimensional Change
Temp (C)

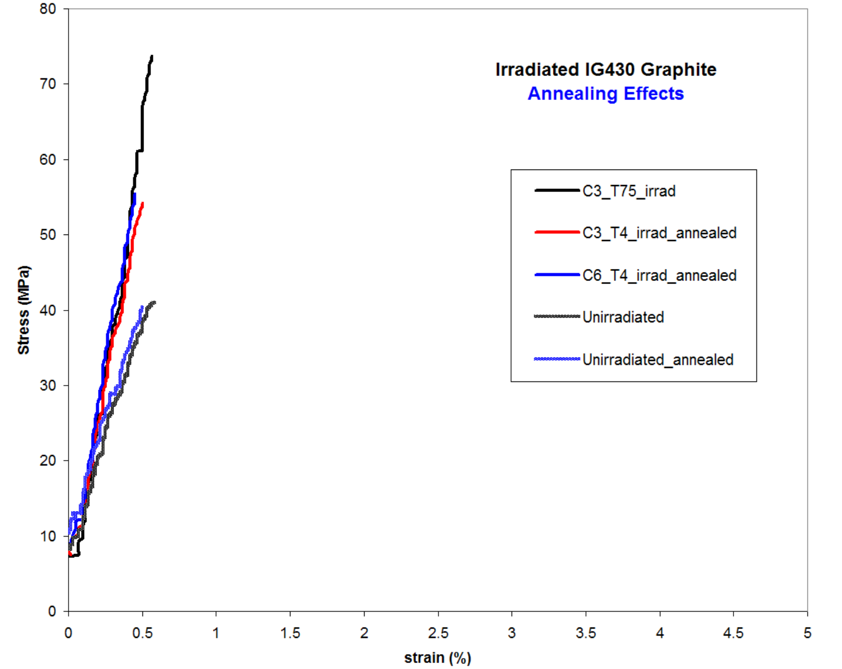

Graphite Thermal Expansion - Radiation Effects
Comparing Grades
Thermal Strain (%)
0.12
0.1
0.08
0.06
0.04
0.02
0
0
50
100
150
200
250
300
350
Temp (C)
S09_TC1
C-2020
IG-430
Poco

Atom vibration,
Activation energy
Interstitial mobilization









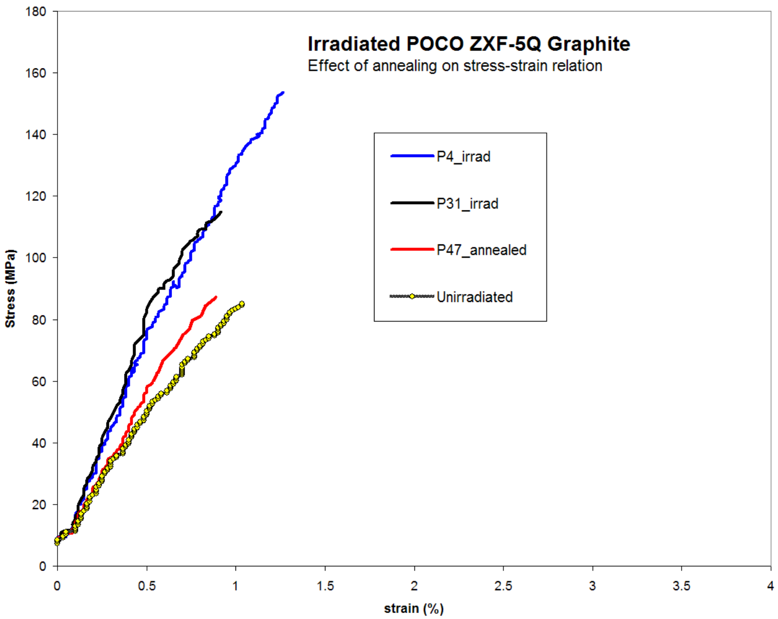
Comparison of measured and predicted creep strains in highly irradiated specimens
(After Kelly and Brocklehurst [38]. Courtesy of the authors.)

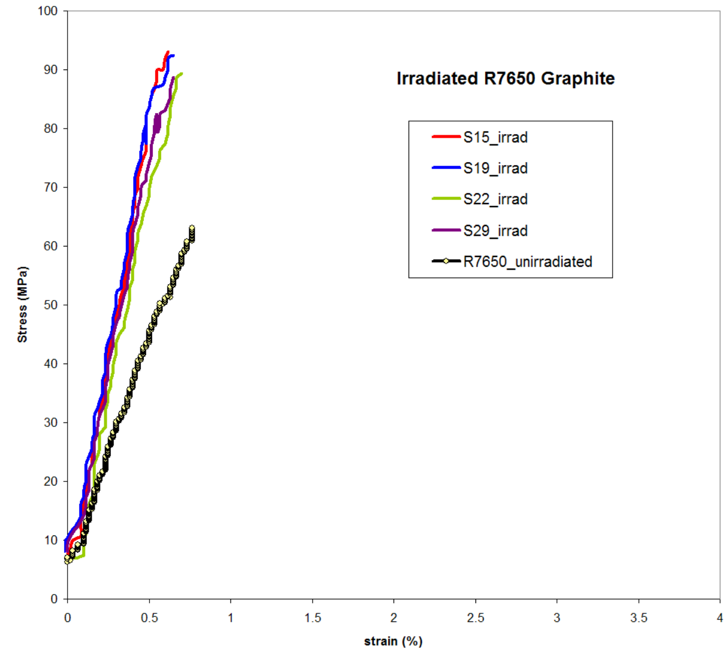
Graphite	Fast neutron dose, 10^{20} n/cm^2: Pre-irradiation	During creep	Stress σ MN/m^2	Modulus, MN/m^2, E_m: Unirradiated E_m	Irradiated and annealed (mean) E_{ms}	Creep strain ϵ, %: Predicted: Based on E_{ms}	Based on E_m	Measured
PV tensile	158	21·8	7·6	6500	12 300	0·37	0·71	0·40
compressive			8·3			0·40	0·77	0·43
compressive			6·2			0·30	0·58	0·38
PW compressive	147	40·0	6·2	7000	13 000	0·49	0·91	0·71

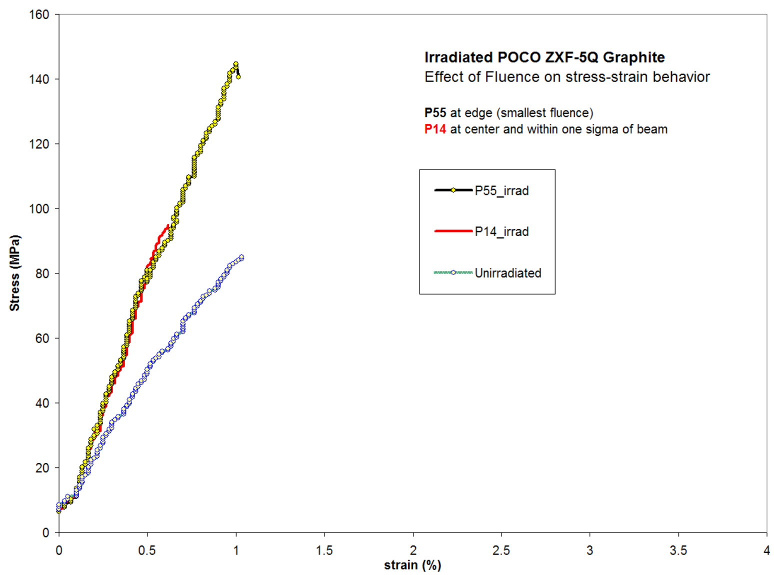
Reported by Gittus:

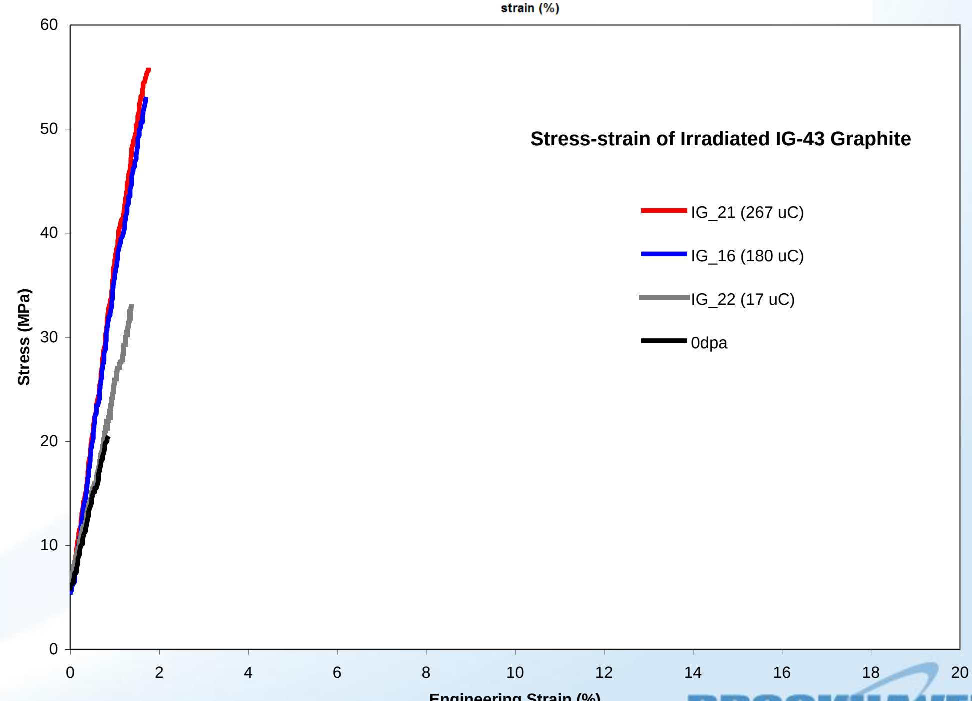
As fast neutron dose increases E increase reaches an ASYMPTOTE (as we see with IG430)

For negligible oxidation, porosity from manufacturing process is gradually reduced as individual graphite crystals undergo “irradiation growth” and grow into pores. The “tightening up” of the aggregate structure gives rise to continuous increase in E, **an increase that cannot anneal out.**

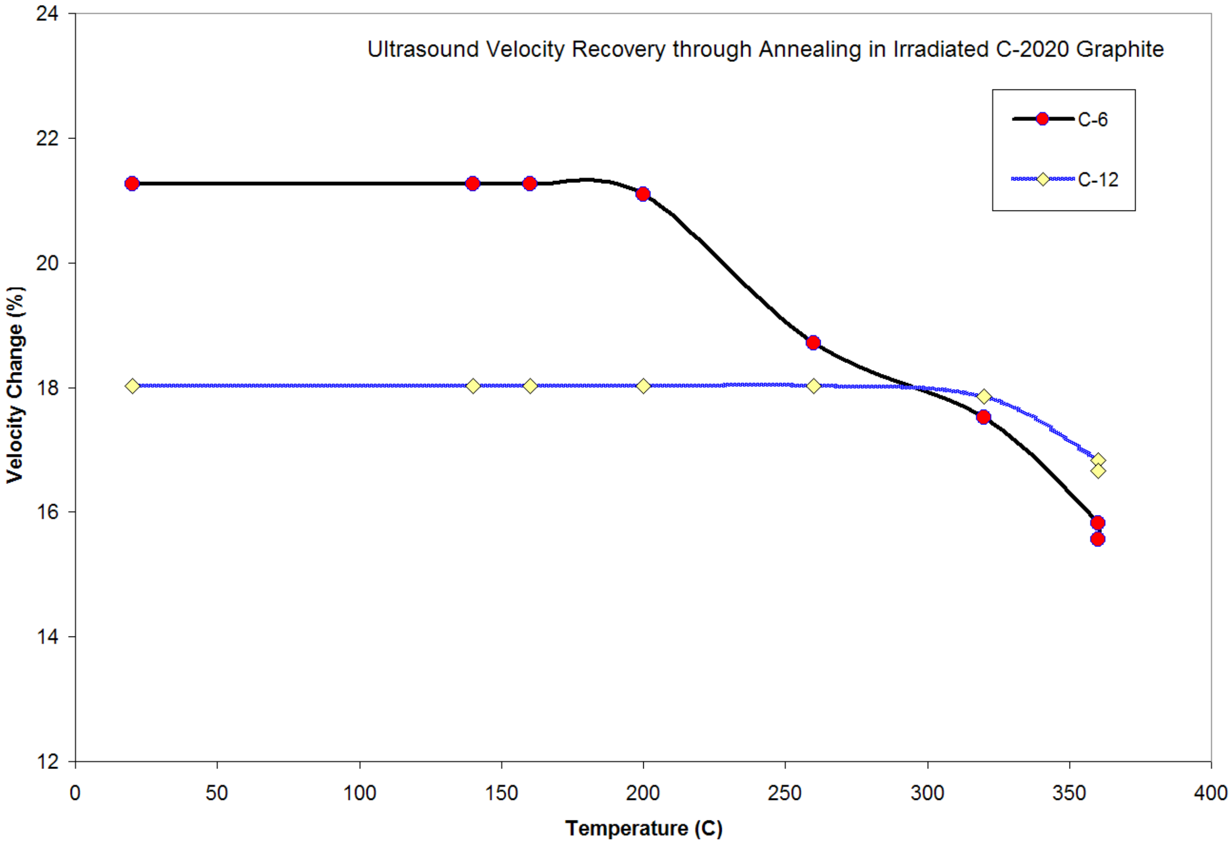

Irradiated POCO ZXF-5Q Graphite
Effect of annealing on stress-strain relation
P4_irrad
P31_irrad
P47_annealed
Unirradiated
Stress (MPa)
strain (%)


Irradiated R7650 Graphite
S15_irrad
S19_irrad
S22_irrad
S29_irrad
R7650_unirradiated
Stress (MPa)
strain (%)


Irradiated POCO ZXF-5Q Graphite
Effect of Fluence on stress-strain behavior
P55 at edge (smallest fluence)
P14 at center and within one sigma of beam
P55_irrad
P14_irrad
Unirradiated
Stress (MPa)
strain (%)


Stress-strain of Irradiated IG-43 Graphite
IG_21 (267 uC)
IG_16 (180 uC)
IG_22 (17 uC)
0dpa
Stress (MPa)
Engineering Strain (%)

Non-destructive testing of graphite damage “annealing” with ultrasound

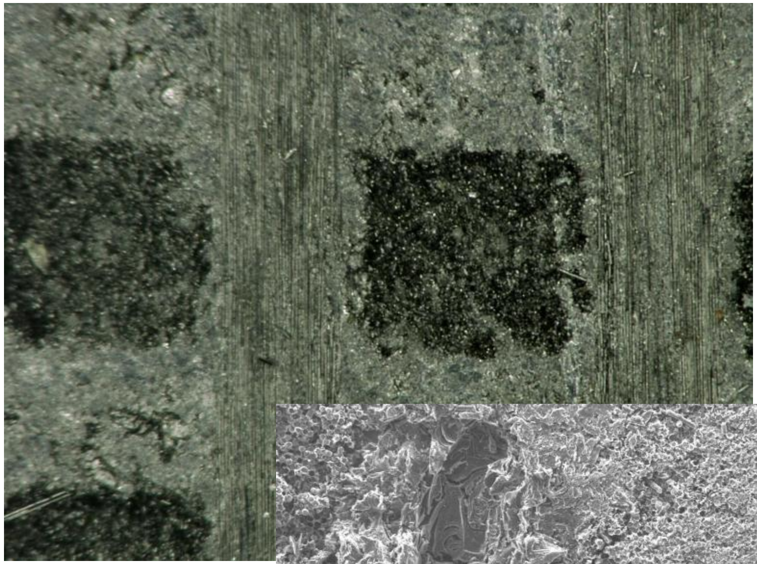


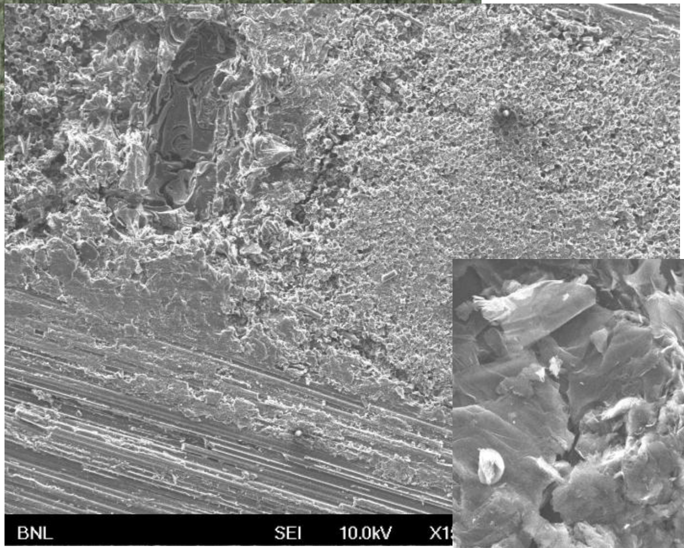
Incomplete !

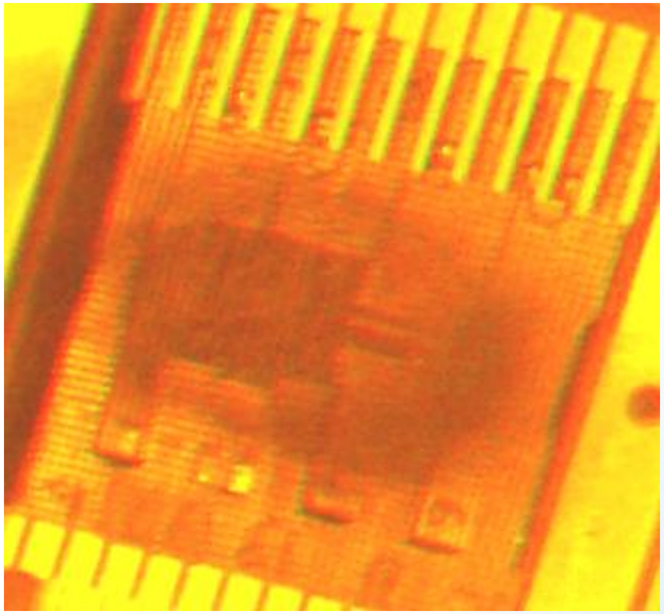
Task: anneal to higher temperatures and observe the residual E increase

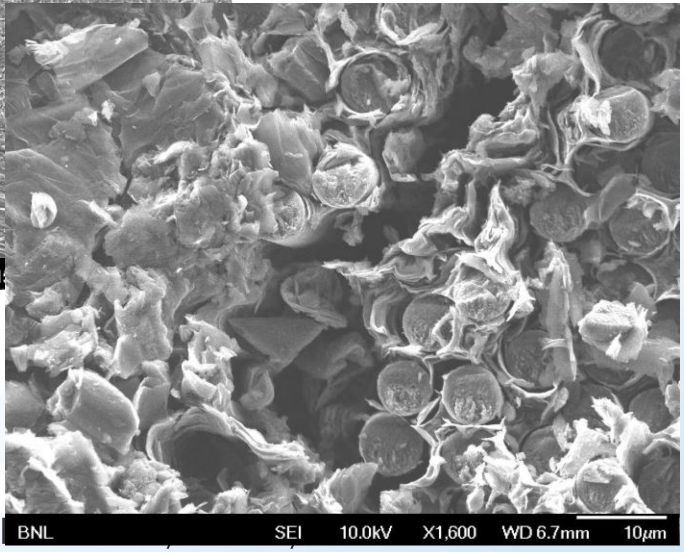
Verify the inability to anneal out and recover E

Carbon/Carbon Composite

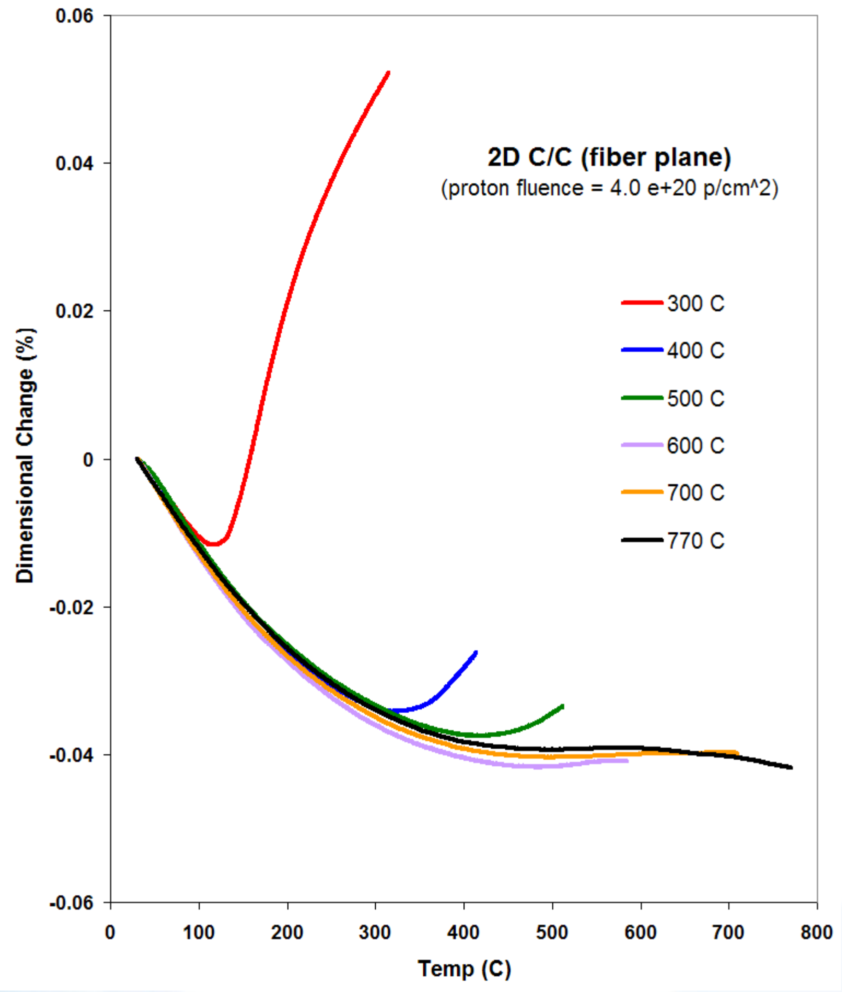


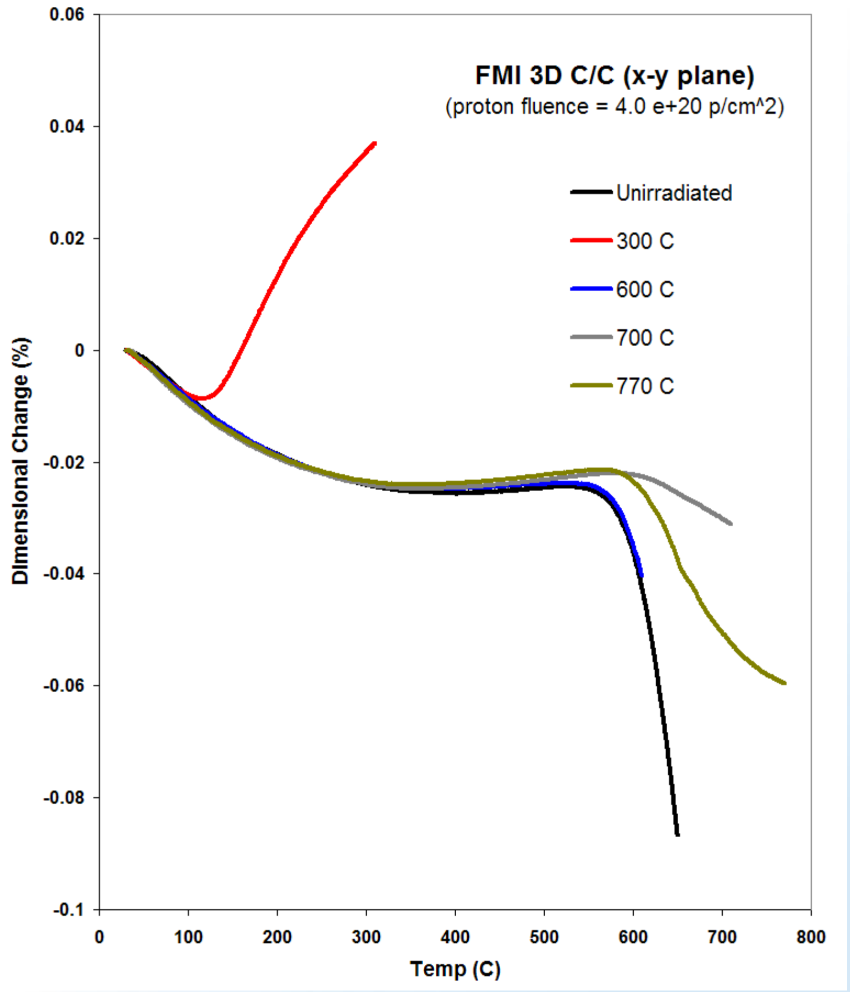




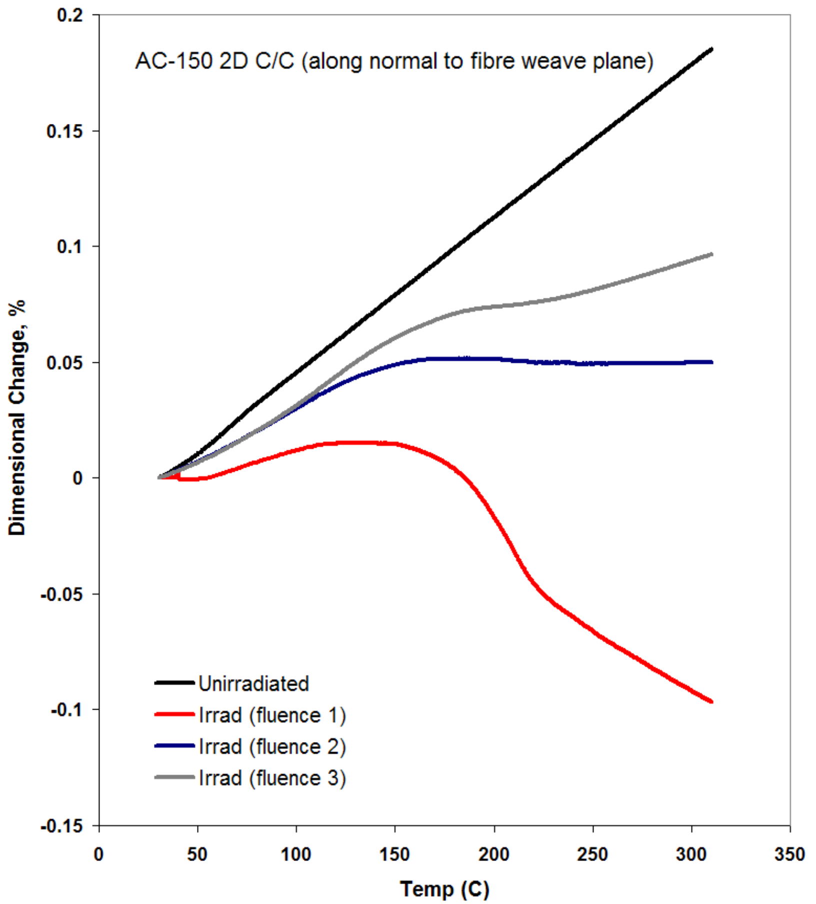


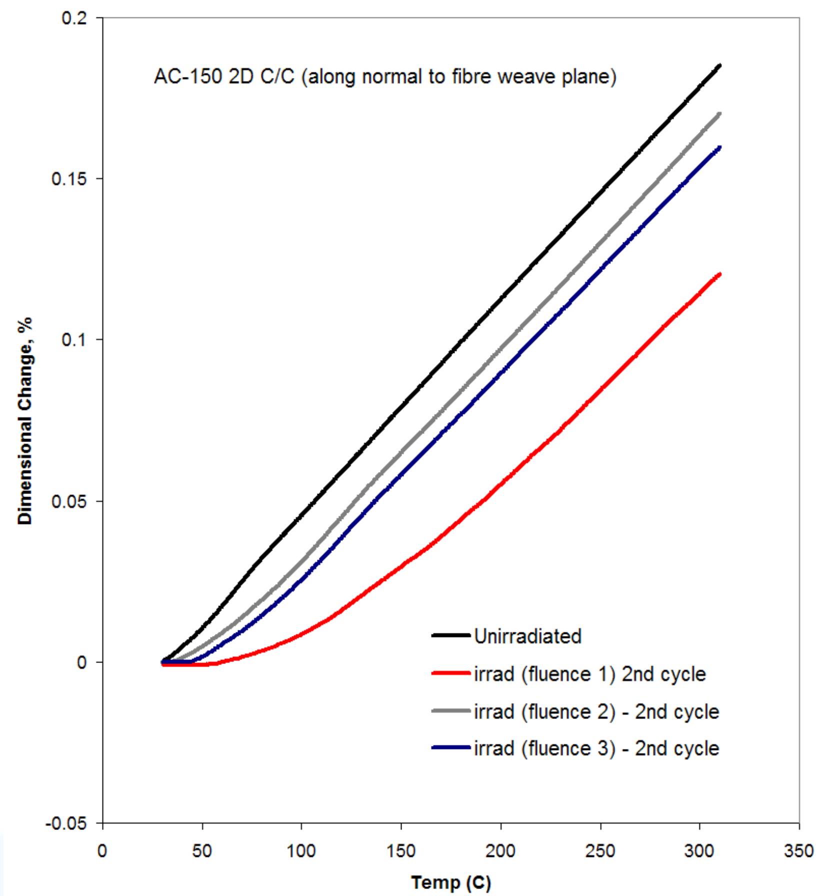
Carbon Fiber Composites
LHC Phase I primary collimator (2-D)

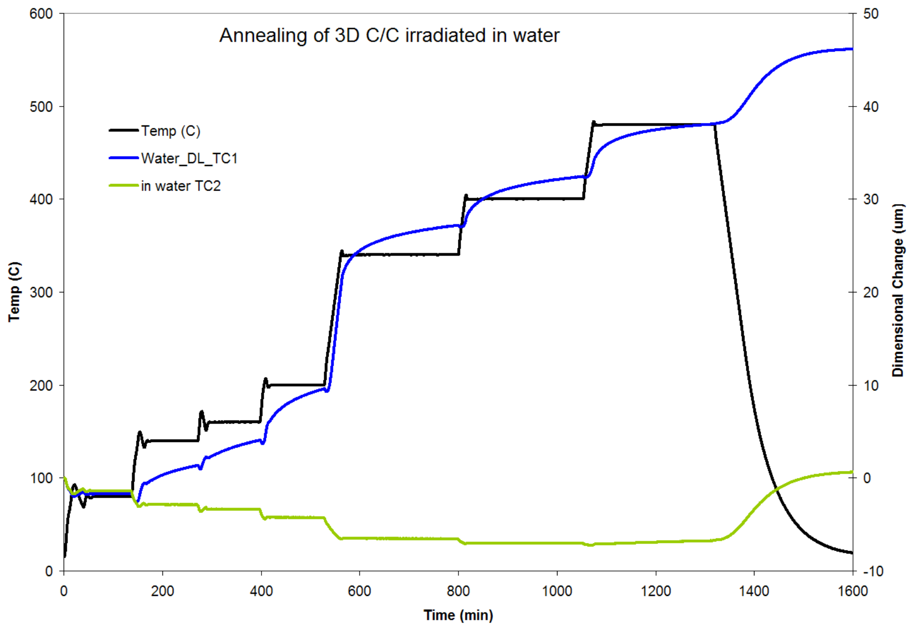


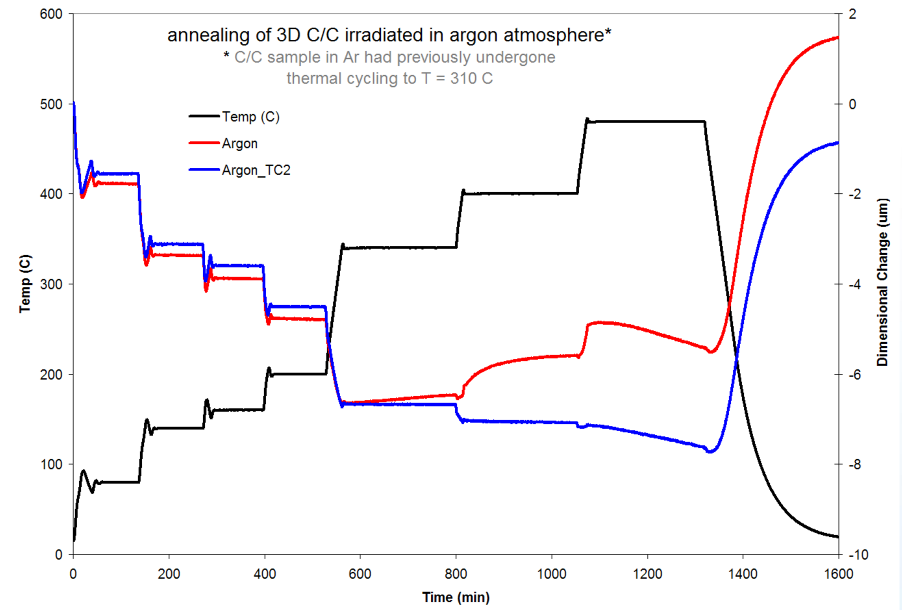


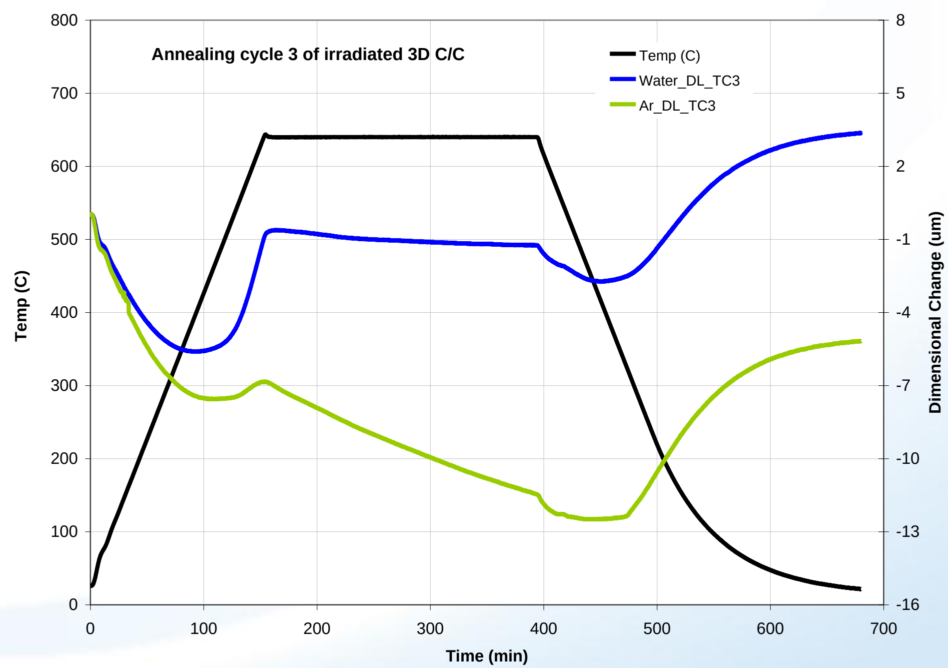
Carbon Fiber Composites
LHC Phase I primary collimator (2-D)

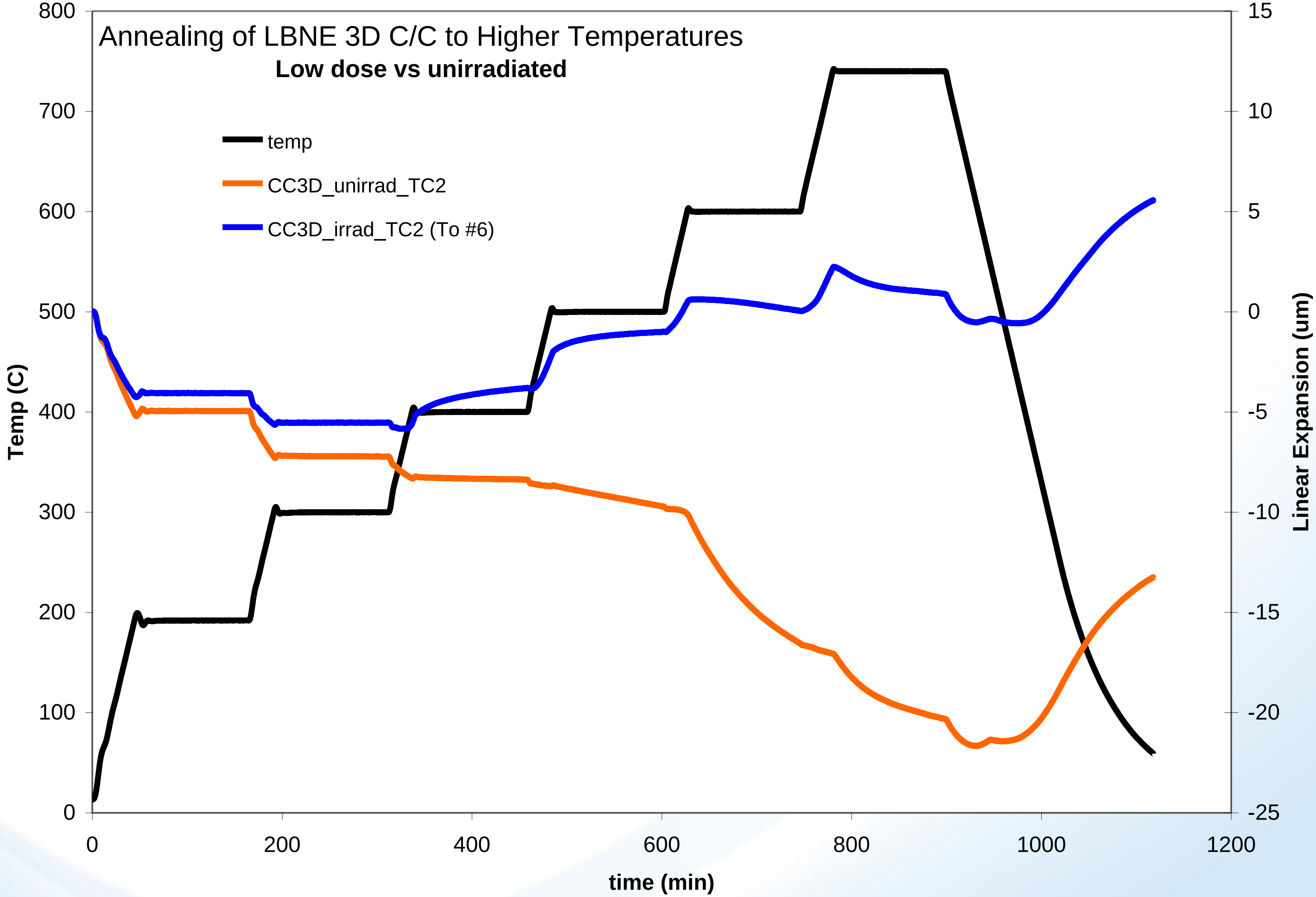





Annealing of 3D C/C irradiated in water
Temp (C)
Water_DL_TC1
in water TC2
Temp (C)
Dimensional Change (um)
Time (min)


annealing of 3D C/C irradiated in argon atmosphere*
* C/C sample in Ar had previously undergone
thermal cycling to T = 310 C
Temp (C)
Argon
Argon_TC2
Temp (C)
Dimensional Change (um)
Time (min)


Annealing cycle 3 of irradiated 3D C/C
Temp (C)
Water_DL_TC3
Ar_DL_TC3
Temp (C)
Dimensional Change (um)
Time (min)

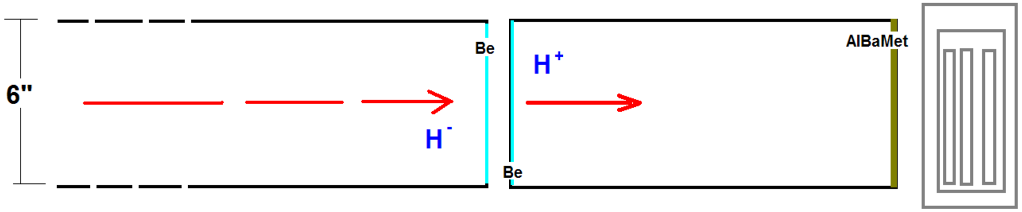

Annealing of LBNE 3D C/C to Higher Temperatures
Low dose vs unirradiated
temp
CC3D_unirrad_TC2
CC3D_irrad_TC2 (To #6)
Temp (C)
Linear Expansion (um)
time (min)
800
700
600
500
400
300
200
100
0
15
10
5
0
-5
-10
-15
-20
-25
0
200
400
600
800
1000
1200

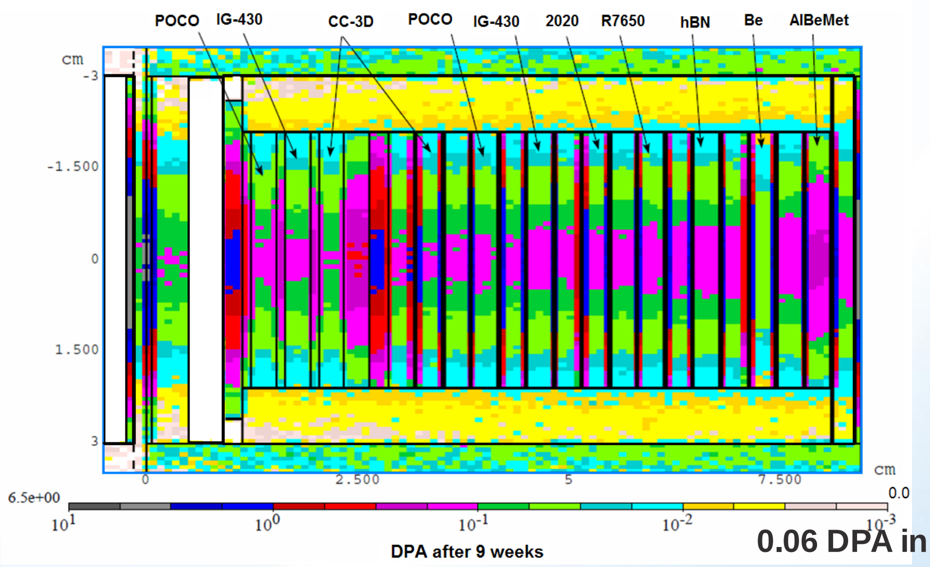
Beryllium (target and beam windows)

Be & BeO (issues in reactor applications at high doses because of the non-symmetric volumetric change)

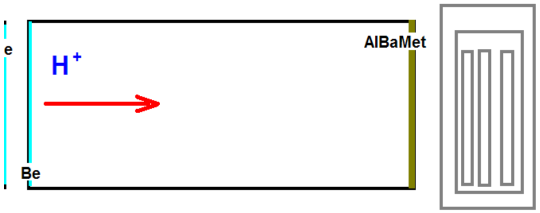
Experience of BNL BLIP Be Windows

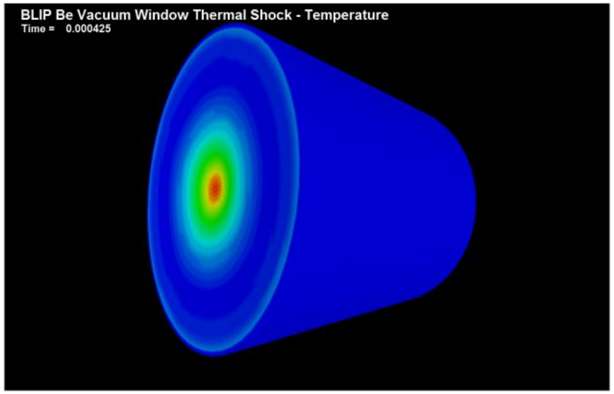
Shock-induced damage (energy reduction to 45 MeV, tightening of the spot and current halfing) led to total destruction after seeing significant beam (1,234,942 uA-hrs, in at 1/26/06 out/disappeared 3/31/10)

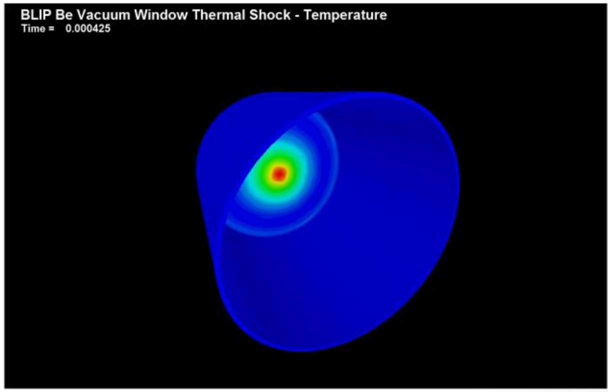


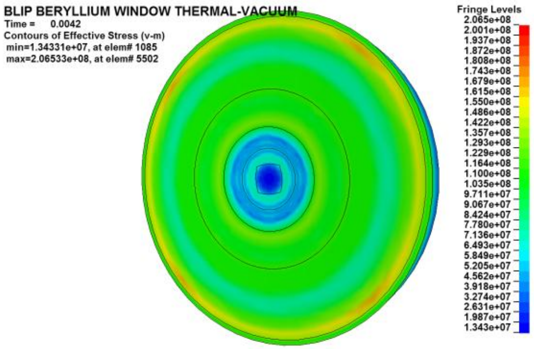


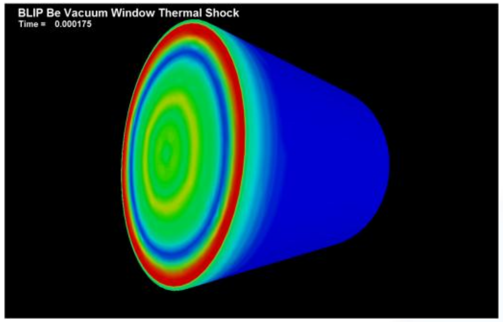
0.06 DPA in Be, and 0.3 DPA in AlBeMet

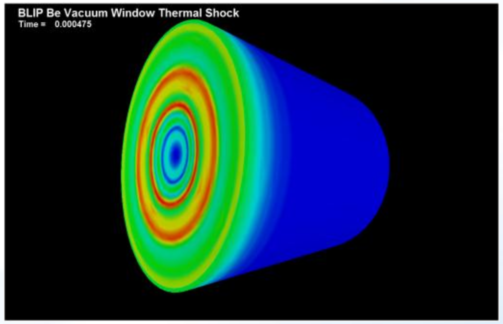

e
H +
AlBaMet
Be

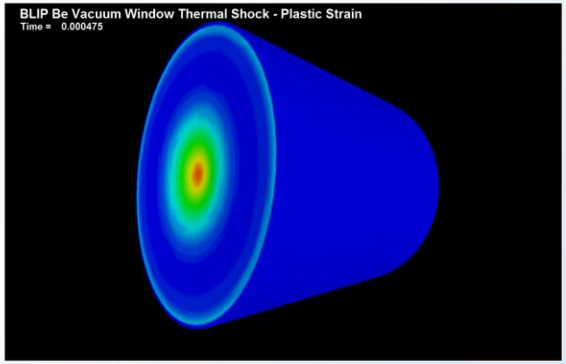

BLIP Be Vacuum Window Thermal Shock - Temperature
Time = 0.000425


BLIP Be Vacuum Window Thermal Shock - Temperature
Time = 0.000425

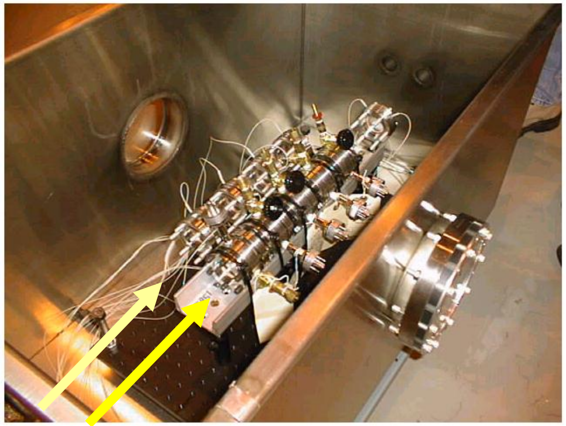

BLIP BERYLLIUM WINDOW THERMAL-VACUUM
Time = 0.0042
Contours of Effective Stress (v-m)
min=1.34331e+07, at elem# 1085
max=2.06533e+08, at elem# 5502
Fringe Levels
2.065e+08
2.001e+08
1.937e+08
1.872e+08
1.808e+08
1.743e+08
1.679e+08
1.615e+08
1.550e+08
1.486e+08
1.422e+08
1.357e+08
1.293e+08
1.229e+08
1.164e+08
1.100e+08
1.035e+08
9.711e+07
9.067e+07
8.424e+07
7.780e+07
7.136e+07
6.493e+07
5.849e+07
5.205e+07
4.562e+07
3.918e+07
3.274e+07
2.631e+07
1.987e+07
1.343e+07

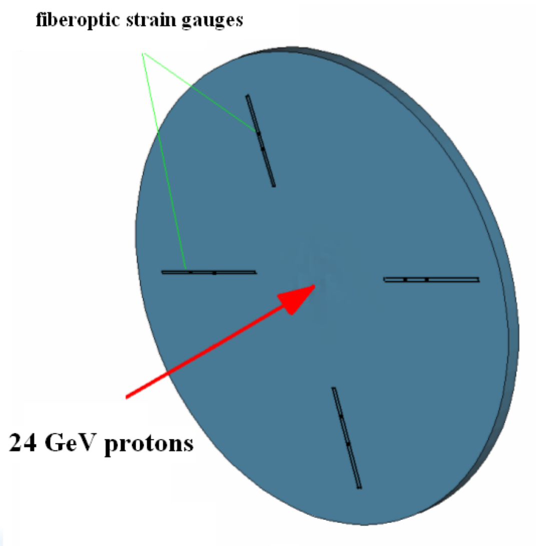

BLIP Be Vacuum Window Thermal Shock
Time = 0.000175

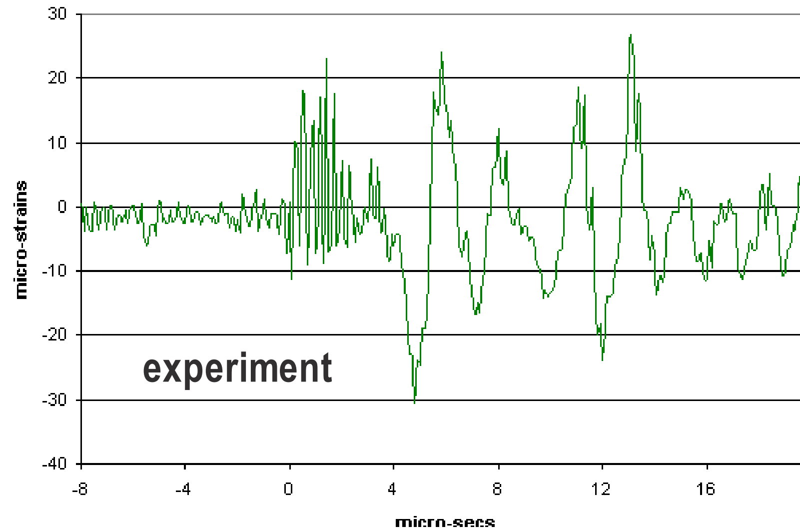

BLIP Be Vacuum Window Thermal Shock
Time = 0.000475

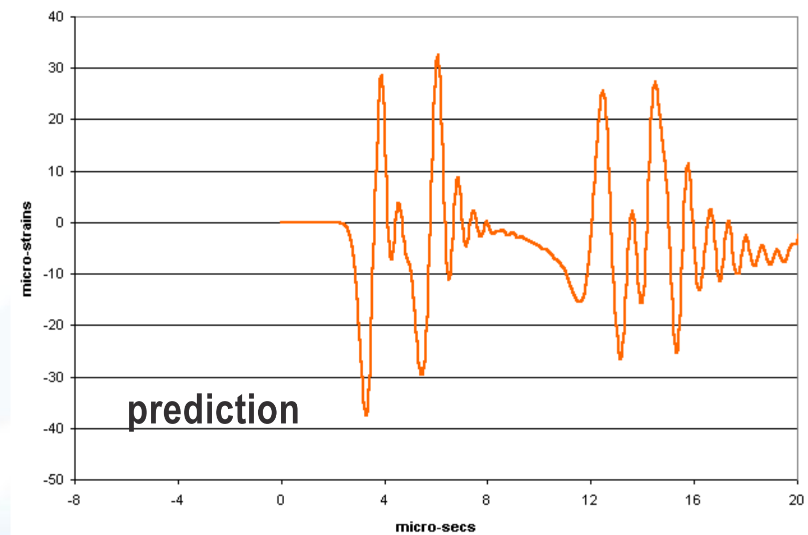

BLIP Be Vacuum Window Thermal Shock - Plastic Strain
Time = 0.000475

Beam-induced shock on thin targets

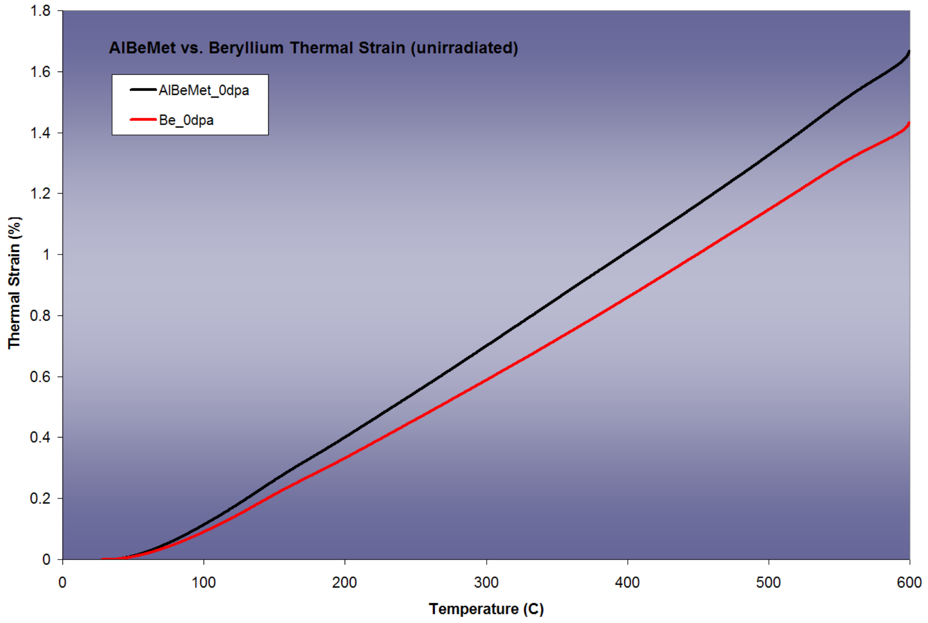


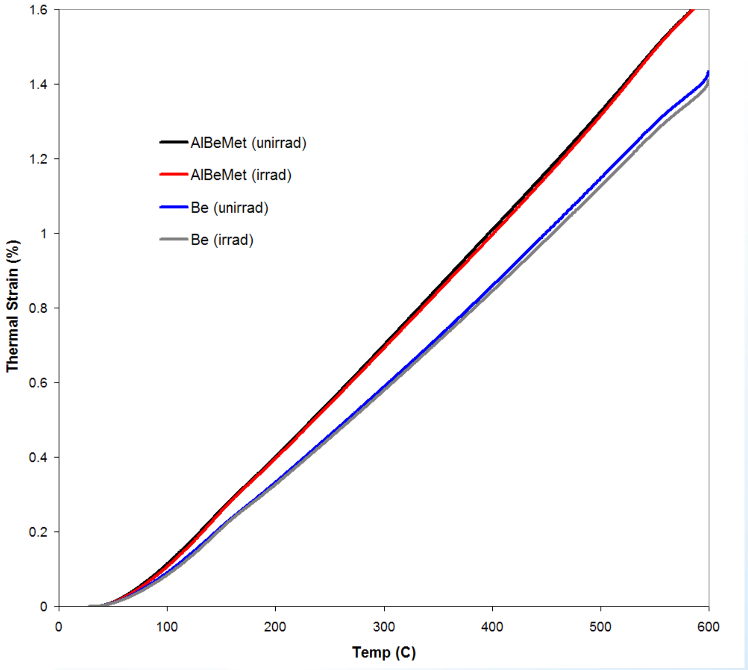




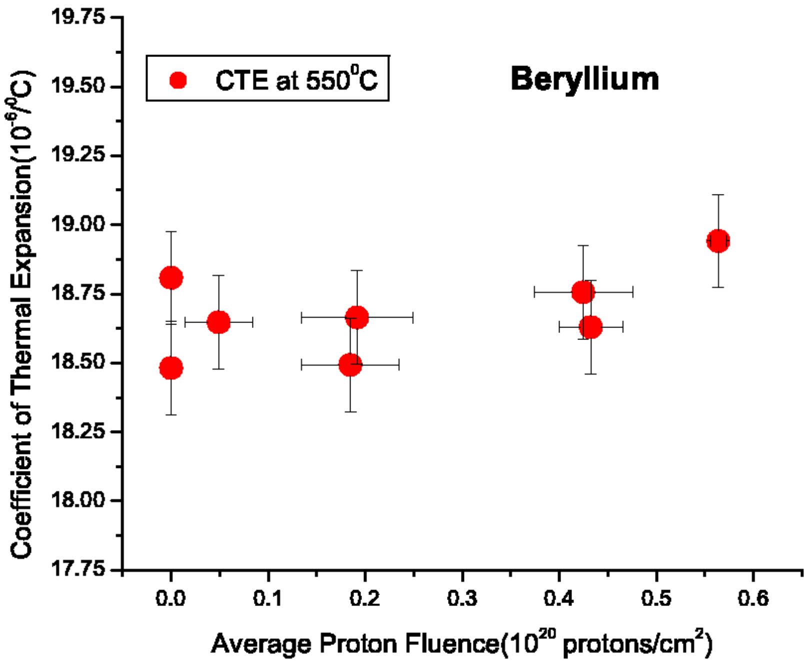


Beryllium

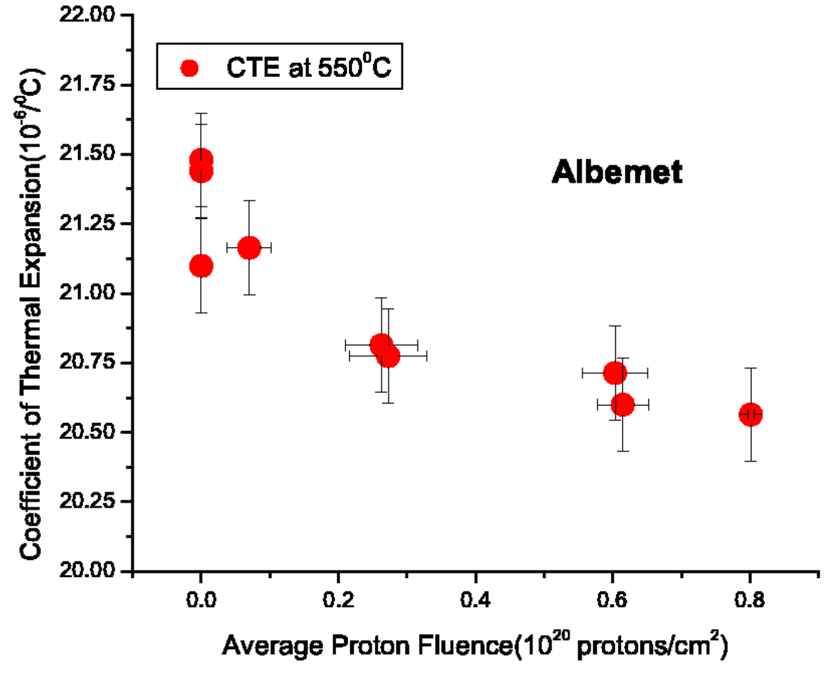

AlBeMet vs. Beryllium Thermal Strain (unirradiated)
AlBeMet_0dpa
Be_0dpa
Thermal Strain (%)
Temperature (C)
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
100
200
300
400
500
600


AlBeMet (unirrad)
AlBeMet (irrad)
Be (unirrad)
Be (irrad)
Thermal Strain (%)
Temp (C)
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
100
200
300
400
500
600

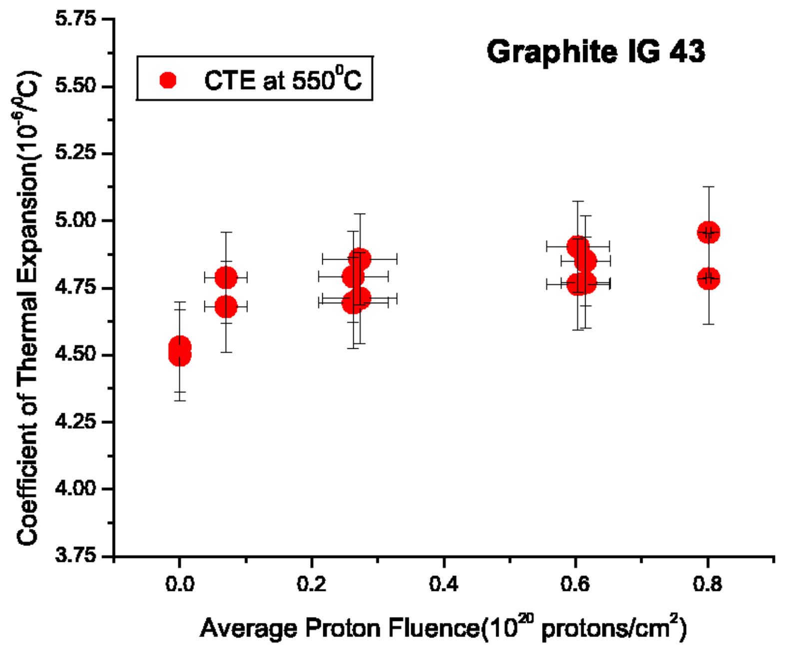
Be & AlBeMet



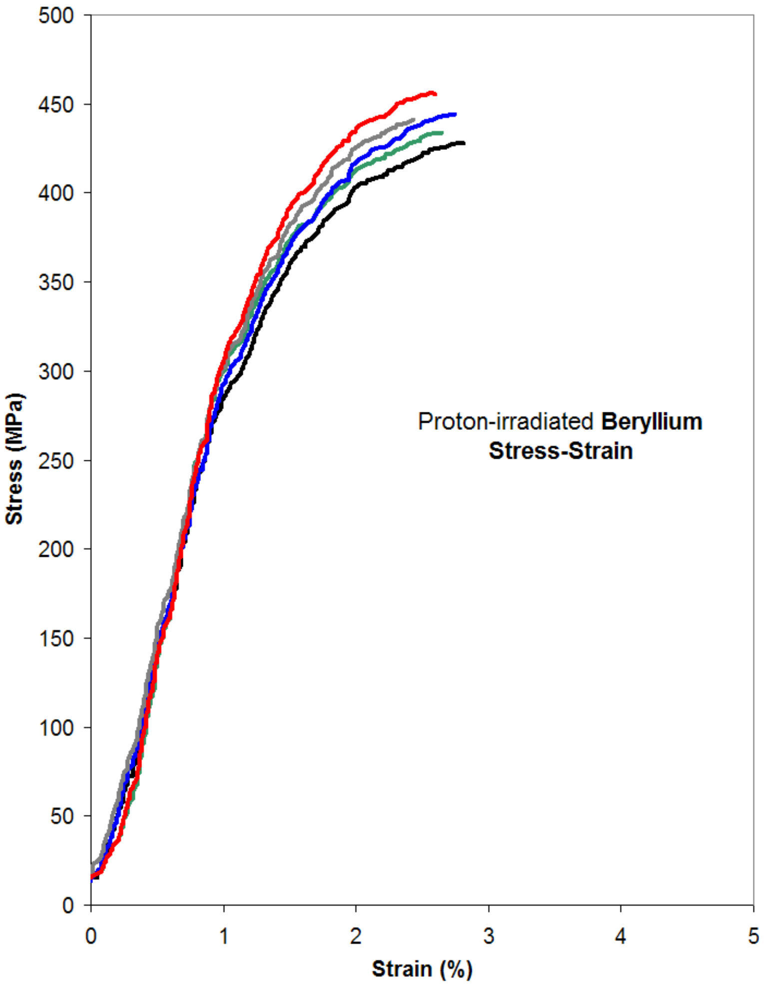
Beryllium
CTE at 550^{0}C
Coefficient of Thermal Expansion(10^{-6}/^{0}C)
Average Proton Fluence(10^{20} protons/cm^2)
19.75
19.50
19.25
19.00
18.75
18.50
18.25
18.00
17.75
0.0
0.1
0.2
0.3
0.4
0.5
0.6



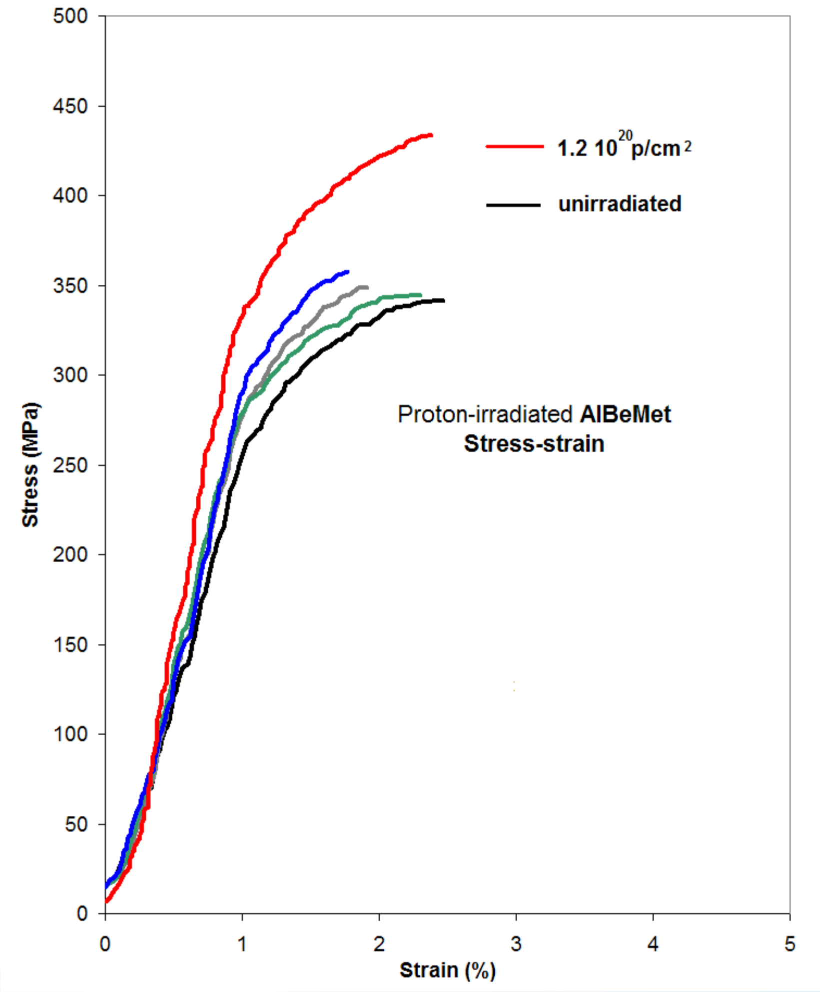
Albemet
CTE at 550^{0}C
Coefficient of Thermal Expansion(10^{-6}/^{0}C)
Average Proton Fluence(10^{20} protons/cm^2)
22.00
21.75
21.50
21.25
21.00
20.75
20.50
20.25
20.00
0.0
0.2
0.4
0.6
0.8



Graphite IG 43
CTE at 550^{0}C
Coefficient of Thermal Expansion(10^{-6}/^{0}C)
Average Proton Fluence(10^{20} protons/cm^2)
5.75
5.50
5.25
5.00
4.75
4.50
4.25
4.00
3.75
0.0
0.2
0.4
0.6
0.8



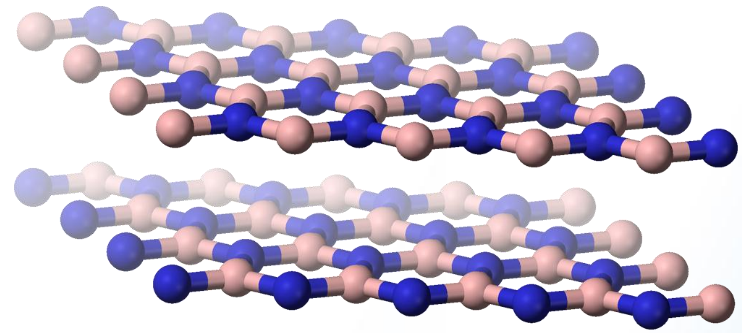
Stress (MPa)
Strain (%)
Proton-irradiated **Beryllium**
Stress-Strain

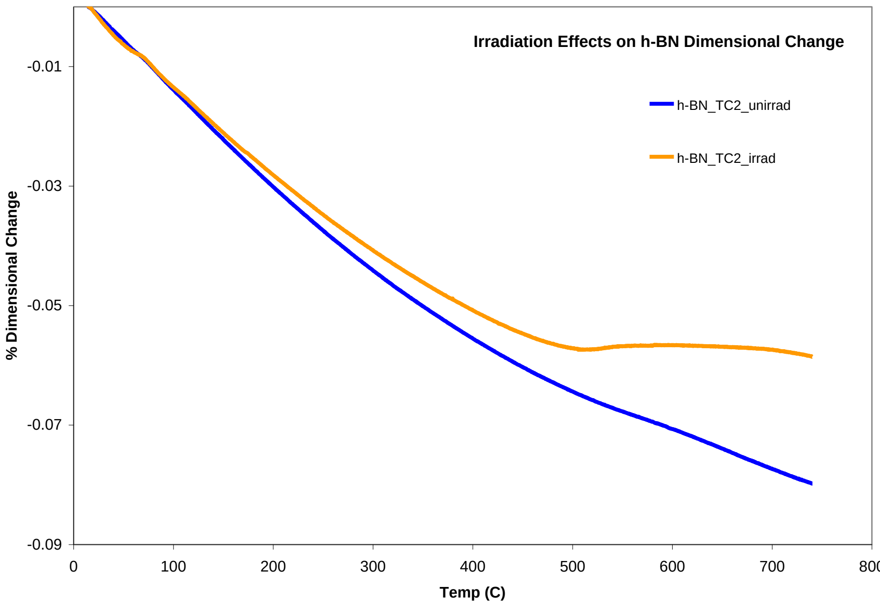


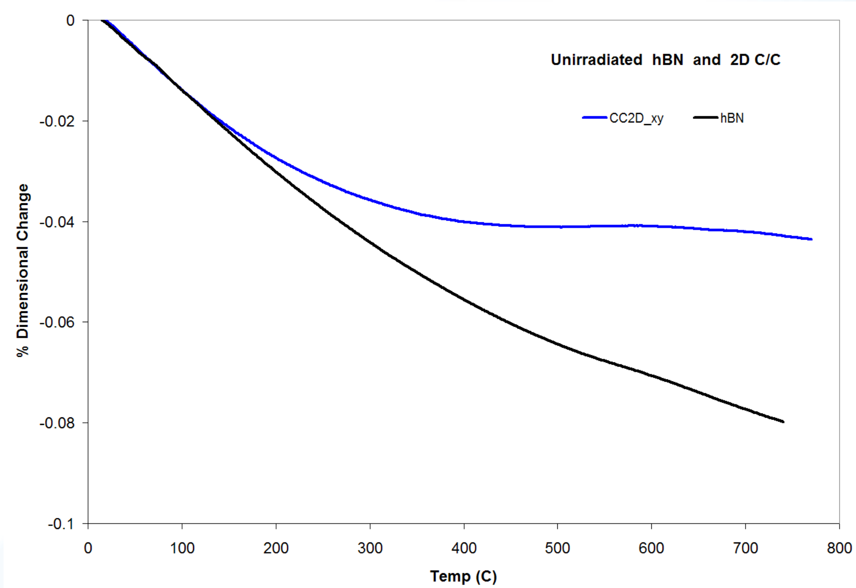
Stress (MPa)
Strain (%)
1.2 10^20 p/cm^2
unirradiated
Proton-irradiated **AlBeMet**
Stress-strain

hBN – Irradiation Damage Study

Because of the lattice structure of h-BN (similar to graphite) it is of interest to observe the dimensional changes as a function of radiation damage and temperature.







Irradiated specimens were very fragile!!!

A possible explanation is that the weakening is attributed **to the production of helium and hydrogen via the (n,α), (n,p), (p,α), and (p,p) reactions.** In the case of boron there is a particularly large (n,α) cross section for the boron-10 isotope, which makes up approximately 20% of natural boron.

Summary

Graphite especially and C/C are still intriguing and with studies that are being pursued we are attempting to understand their limitations better in accelerator targets

There is interest in Be and new initiatives are being formulated to study it further

Interest also exists in other low-Z materials and alloyed structures

The power demand in combination with radiation damage (and thus the useful life of the target) are the driver of these efforts