Long baseline neutrino experiment (LBNE) target material radiation damage studies using energetic protons of the Brookhaven linear isotope production (BLIP) facility*

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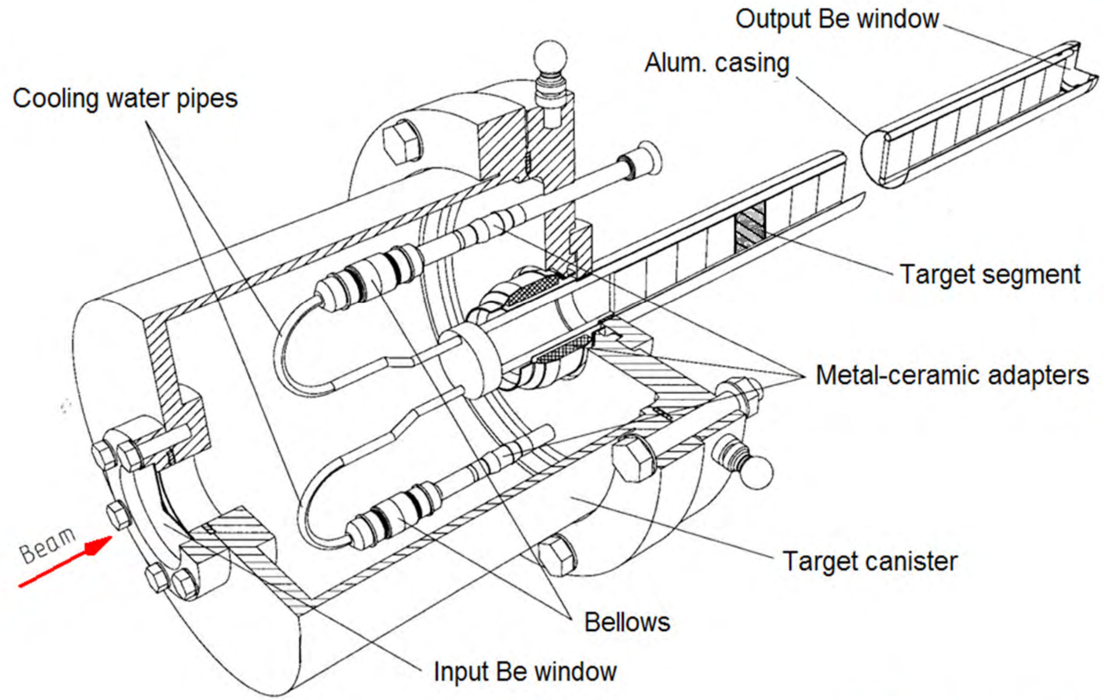
Objective

Concerns from past studies, including 2D-CC LHC collimator material, that energetic protons are much more damaging to graphite and other carbon-based structures prompted new studies on low-Z materials for LBNE targets

Verify the C/C limitations and assess the influence of operating environment (for high power water in contact cannot be excluded)

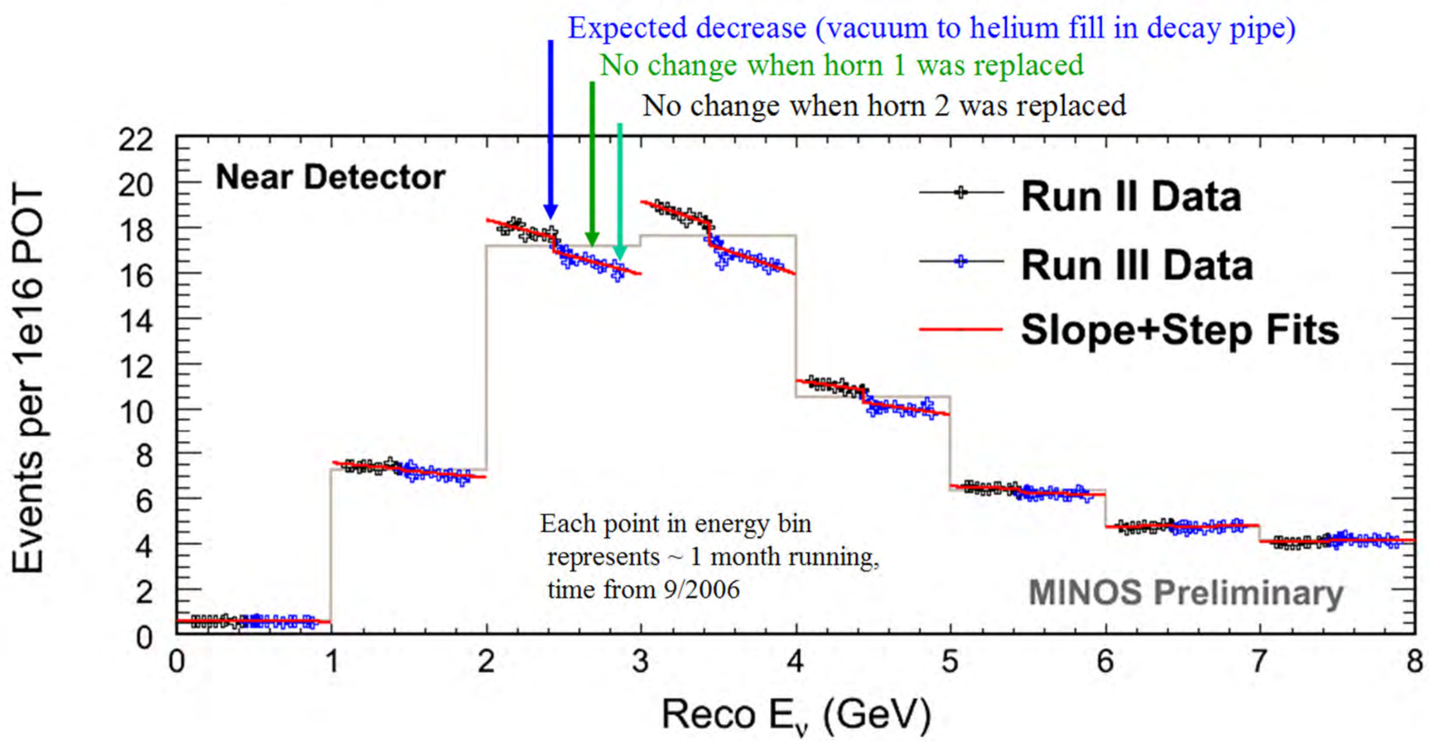
Evaluate graphite across different grades by addressing damage thresholds, annealing, dimensional change, strength, etc.

Explore h-BN and AlBeMet and Be



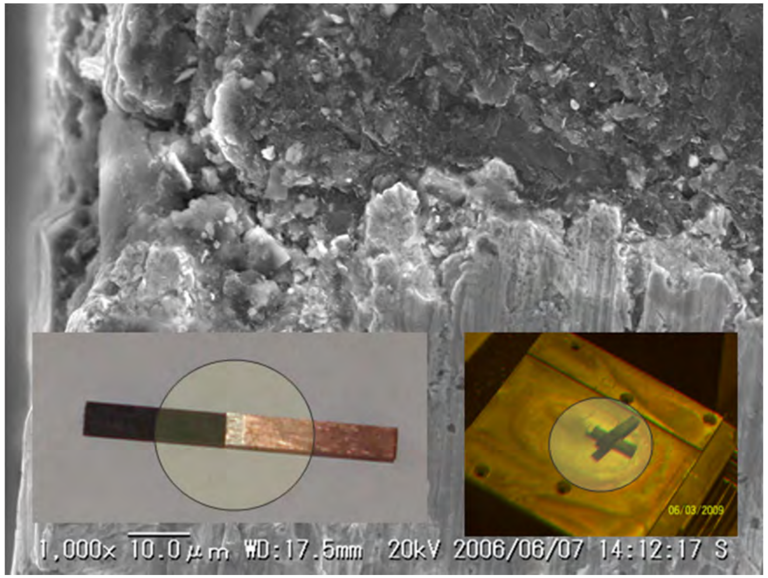
NUMI Target (ZXF-5Q amorphous graphite) Experience

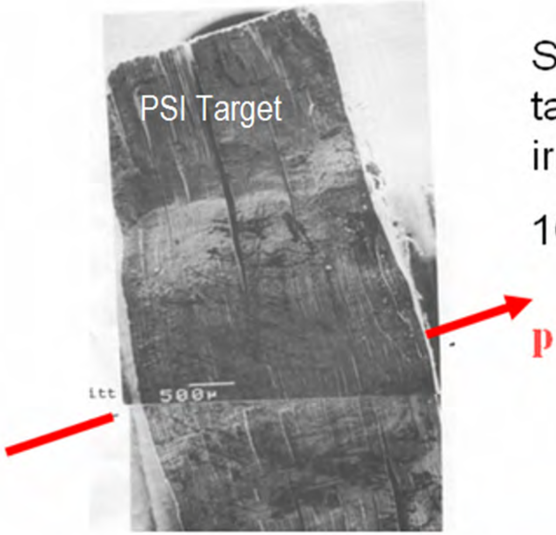
Gradual neutrino rate decrease attributed to target radiation damage



Experience of graphite damage from energetic protons < 1 dpa !!!

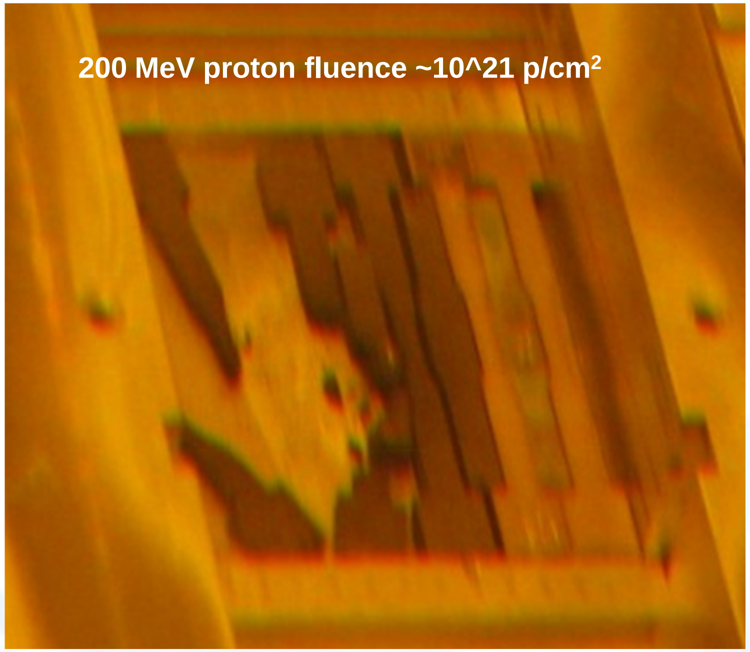
On the contrary, thermal neutron irradiation has achieved tens of dpa.





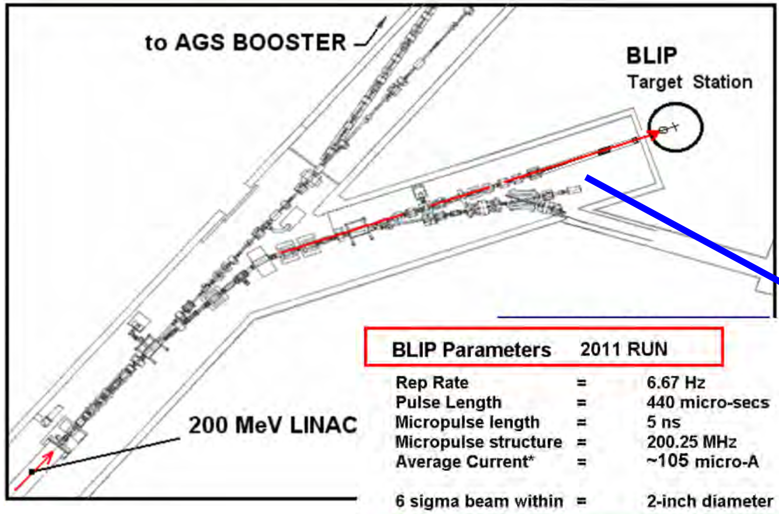
Swelling of the target after irradiation

10^{22} p/cm2

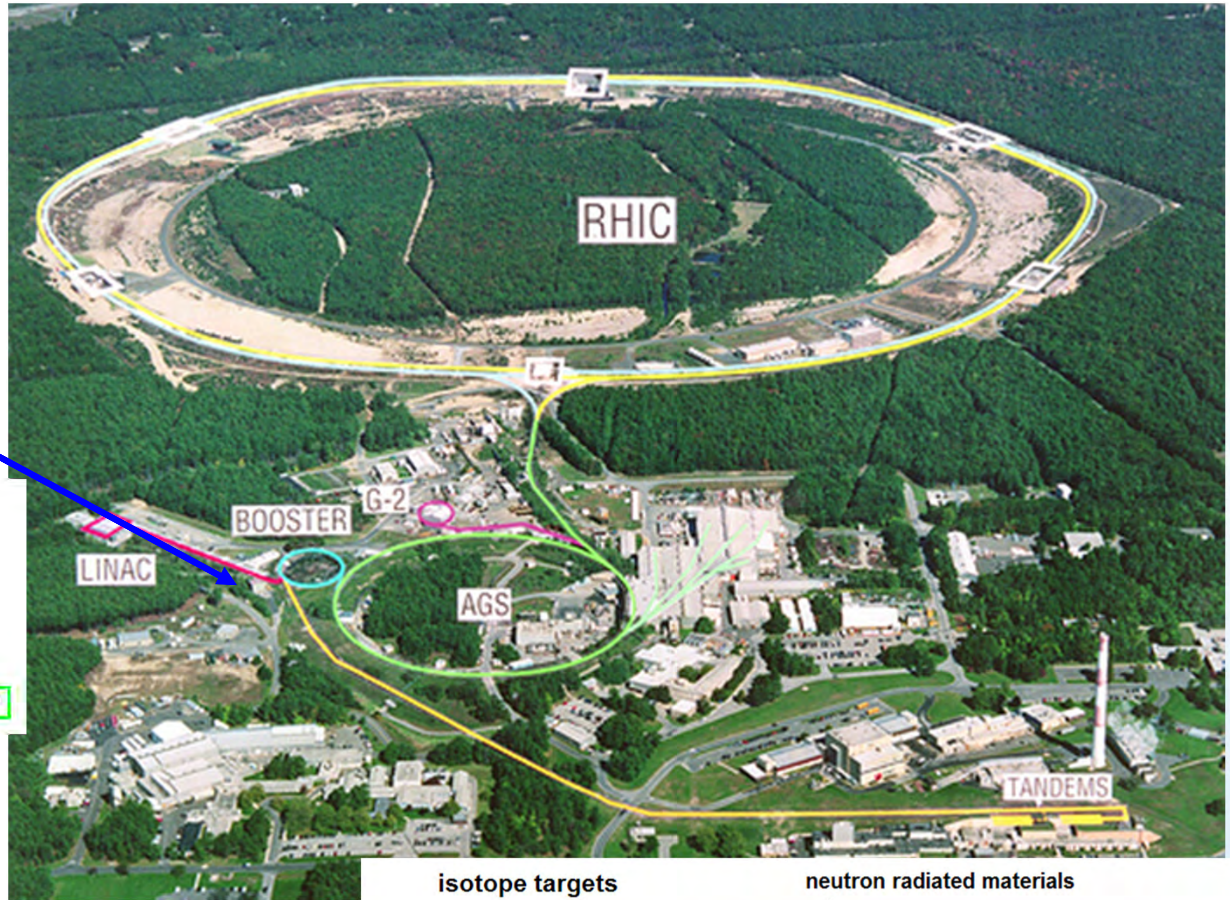


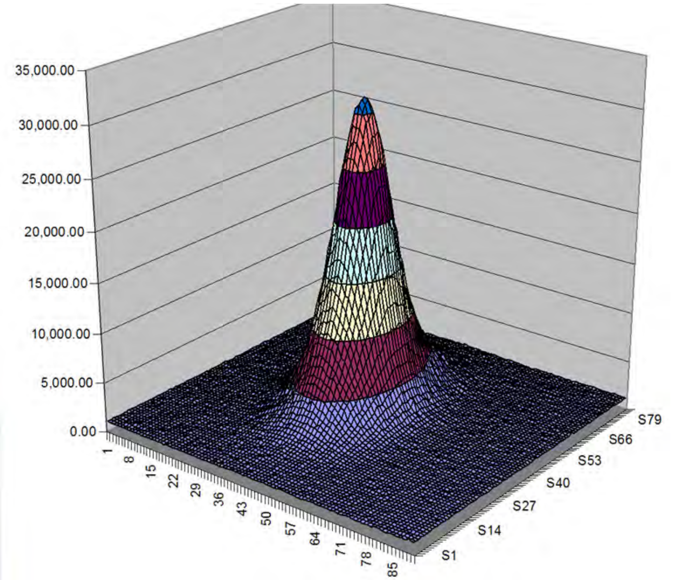
Irradiation at the BNL Accelerator Complex

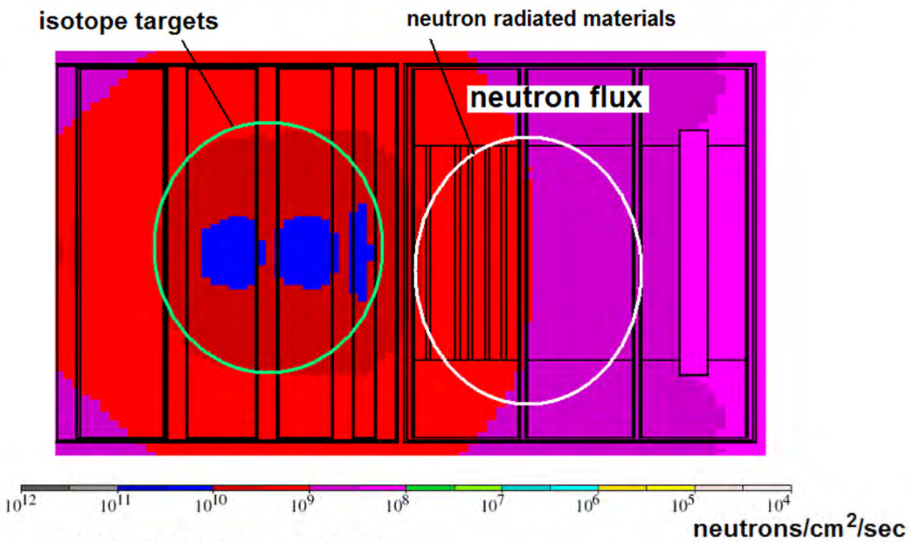
Irradiation at BLIP
(up to 200 MeV or spallation neutrons from 112 MeV protons)



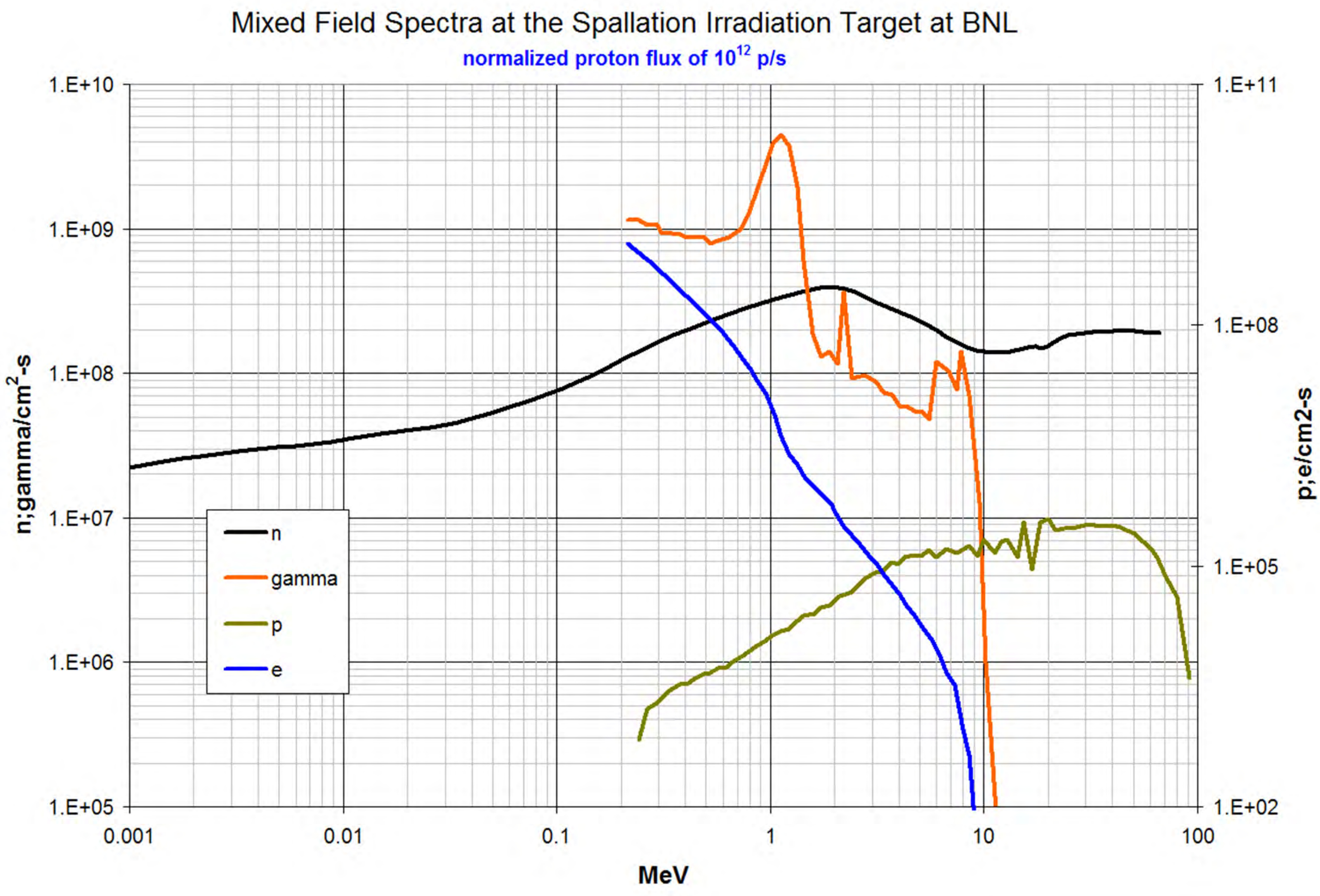
BLIP Parameters		2011 RUN
Rep Rate	=	6.67 Hz
Pulse Length	=	440 micro-secs
Micropulse length	=	5 ns
Micropulse structure	=	200.25 MHz
Average Current*	=	~105 micro-A
6 sigma beam within	=	2-inch diameter
Beam Gaussian	==>	1 sigma = 4.233 mm

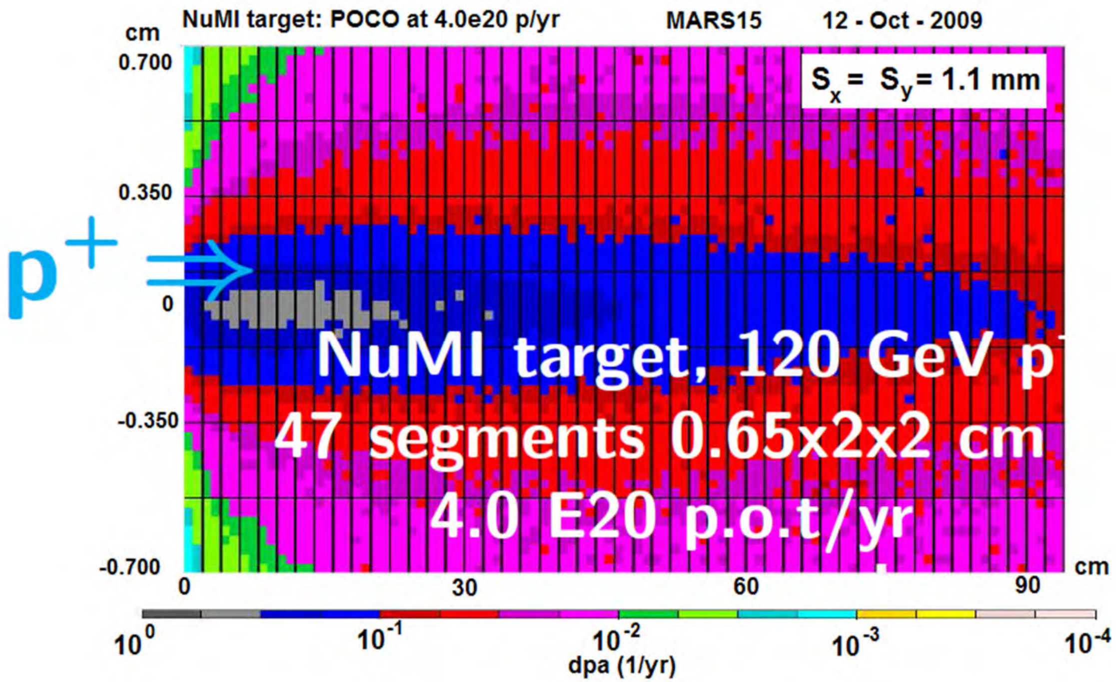






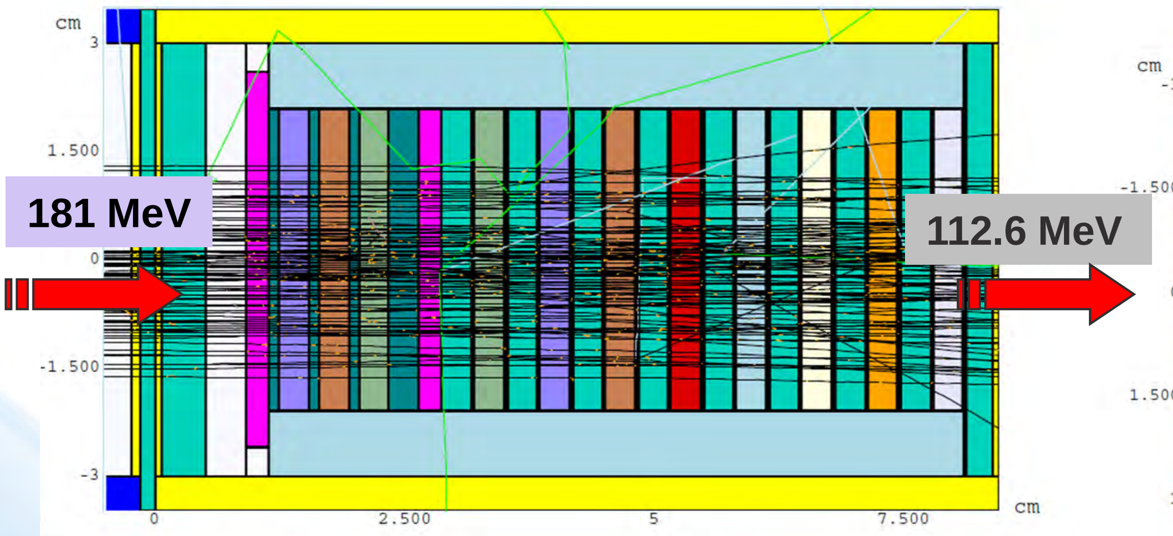
NORMALIZED neutron flux
at BLIP target station (by N. Mokhov, FNAL)

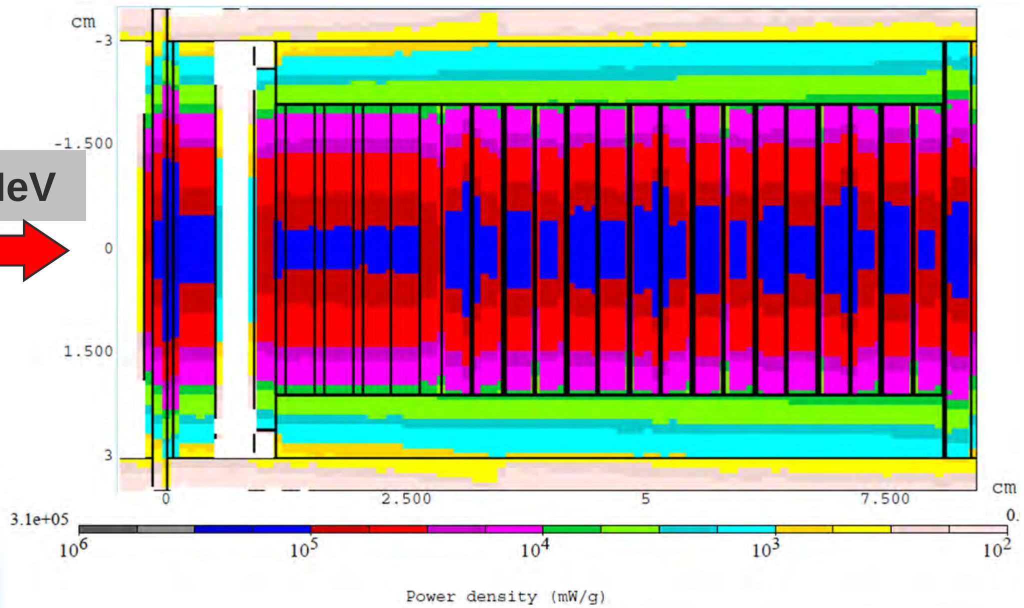

Mixed Field Spectra at the Spallation Irradiation Target at BNL
normalized proton flux of 10^12 p/s
n;gamma/cm2-s
p;e/cm2-s
MeV
1.E+10
1.E+09
1.E+08
1.E+07
1.E+06
1.E+05
1.E+11
1.E+08
1.E+05
1.E+02
0.001
0.01
0.1
1
10
100
n
gamma
p
e

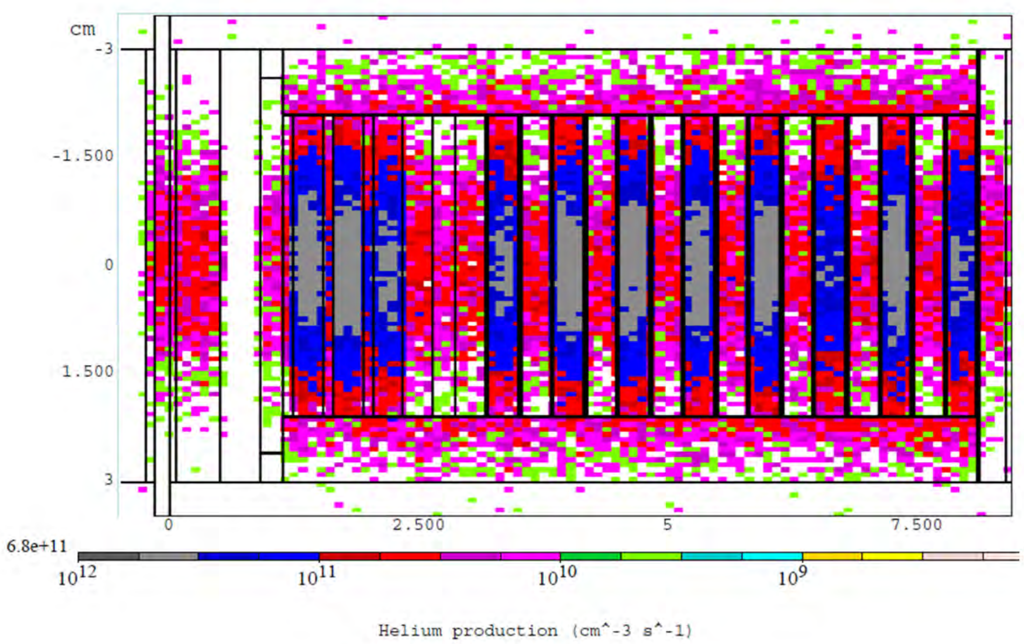


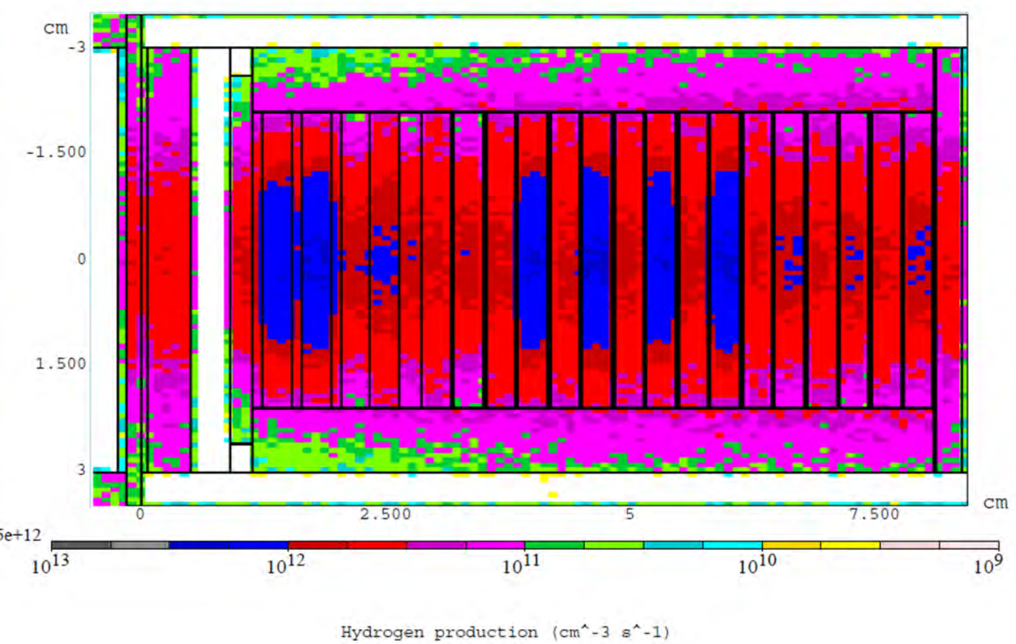
NuMI target simulation with MARS-15

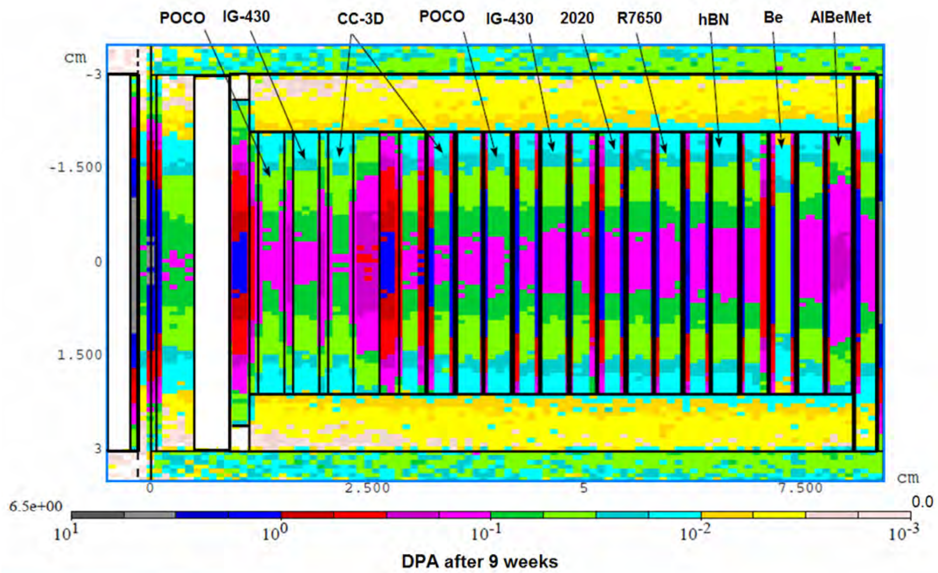
BLNL_BLIP target simulation with MARS-15











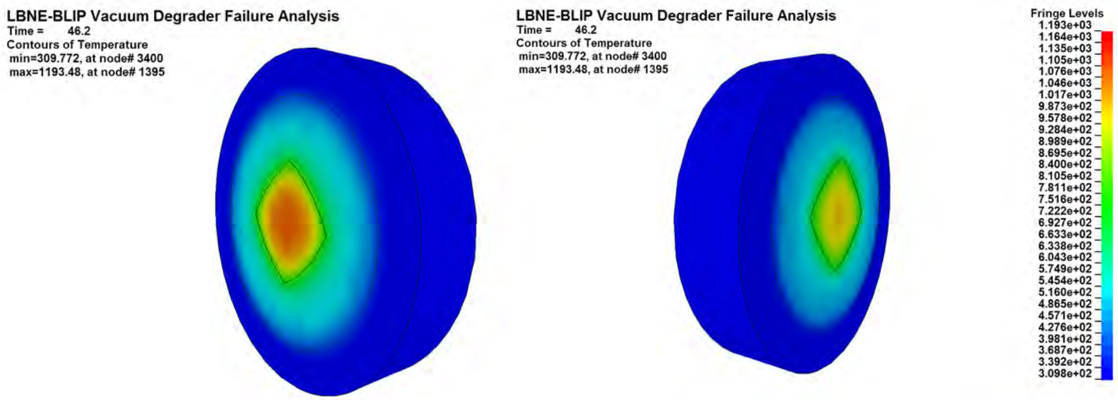
MARS-15 analysis confirmed what has been anticipated/observed in the various studies prior, that damage (dpa portion) is greater at the lower energies

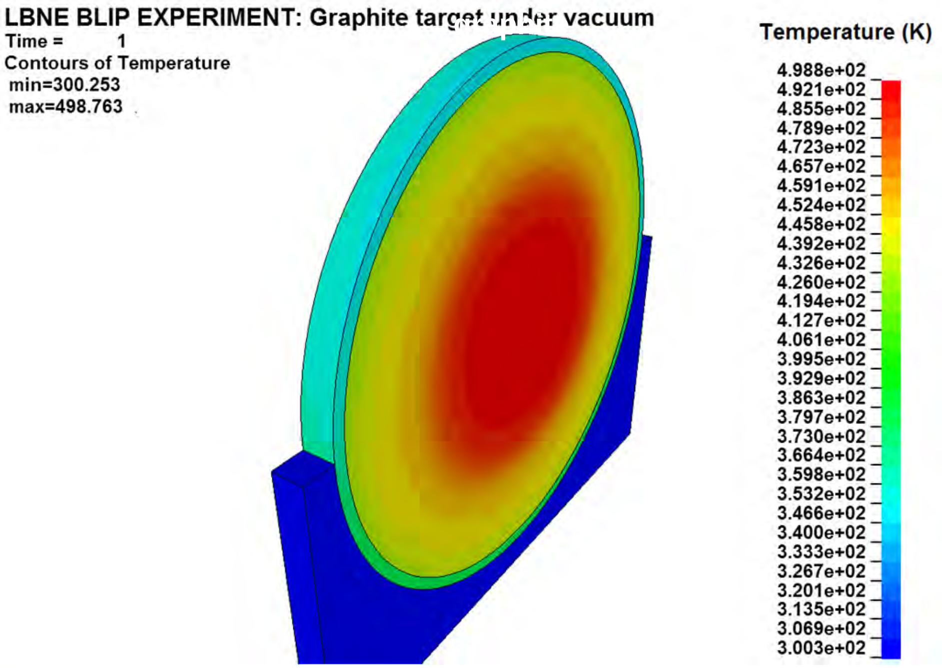
Physics process contribution (%) at beam axis:
z=15 cm (NuMI) and Box 2 POCO graphite (BLIP)

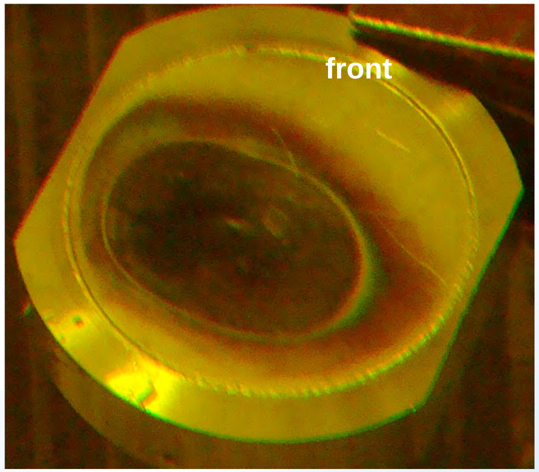
Target	Nuclear	EM elastic	L.E. neutrons	e±
NuMI	50.8	43.3	1.5	4.4
BLIP	43.5	53	3.5	0.02

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

LBNE Target Temperature Predictions







While a difficult thermal problem, the predictions on graphite T_{irr} were very close to the actual test values

LBNE Target Radionuclide Production Analysis using MCNPX

From the entire spectrum of nuclides produced by the interaction of the 181 MeV with the target array, attention was focused on a short list and in particular those that are expected to be contributing to the dose registered around BLIP.

Nuclide	Half-life	Nuclides per proton from water in LBNE targets	TOTAL # of nuclides per proton [targets + water]
O-14	70.6 s	7.50 e-05	7.50 e-05
O-15	122.2 s	1.51 e-03	1.52 e-03
N-13	9.97 min	2.90 e-04	3.01 e-04
C-11	20.3 min	6.67 e-04	5.45 e-03
Be-7	**53.28 days**	**2.71 e-04**	**2.61 e-03**

Based on BLIP average beam current of 104 μA which corresponds to ~ 6.4945 10^{14} protons/sec the following estimates are deduced:

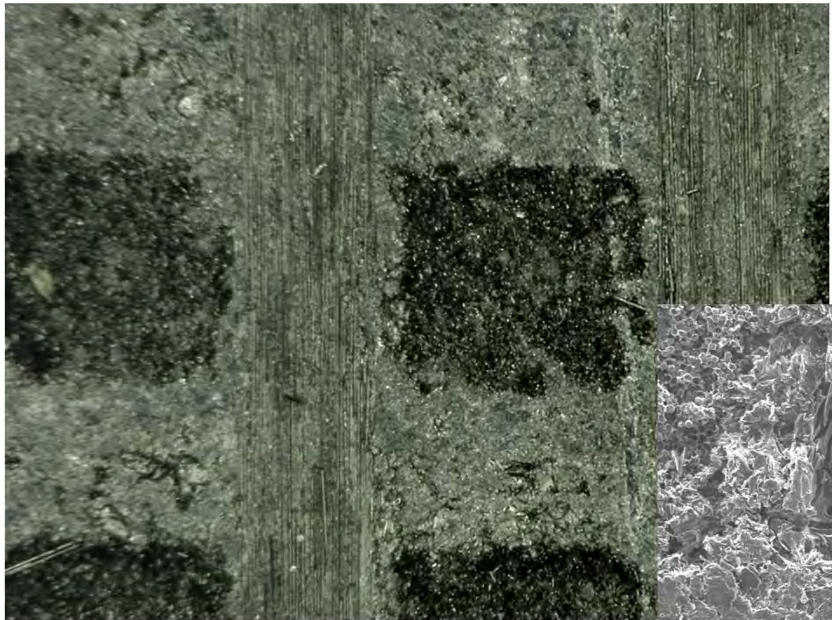
O-14	$1.99 \cdot 10^{-17}$ Ci/proton
O-15	$2.32 \cdot 10^{-16}$ Ci/proton
N-13	$9.07 \cdot 10^{-18}$ Ci/proton
C-11	$1.02 \cdot 10^{-17}$ Ci/proton

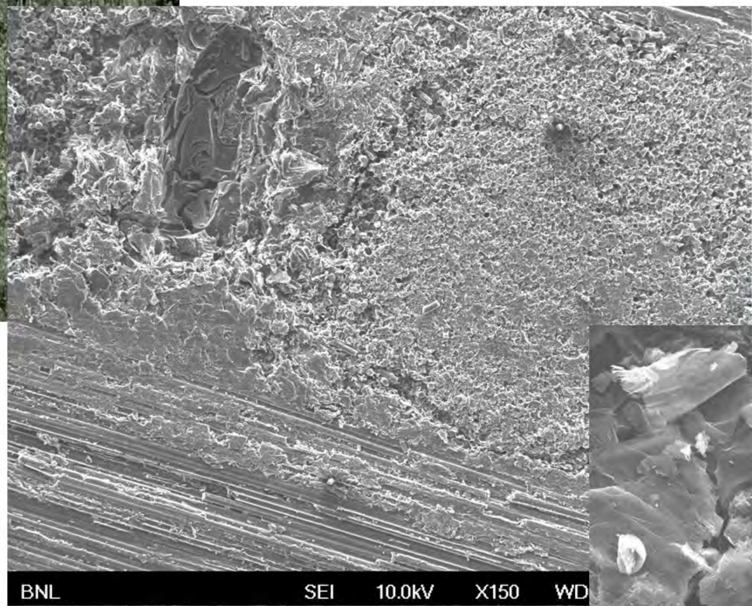
for current of 104 μA average at BLIP translate the production rate from the LBNE target water:

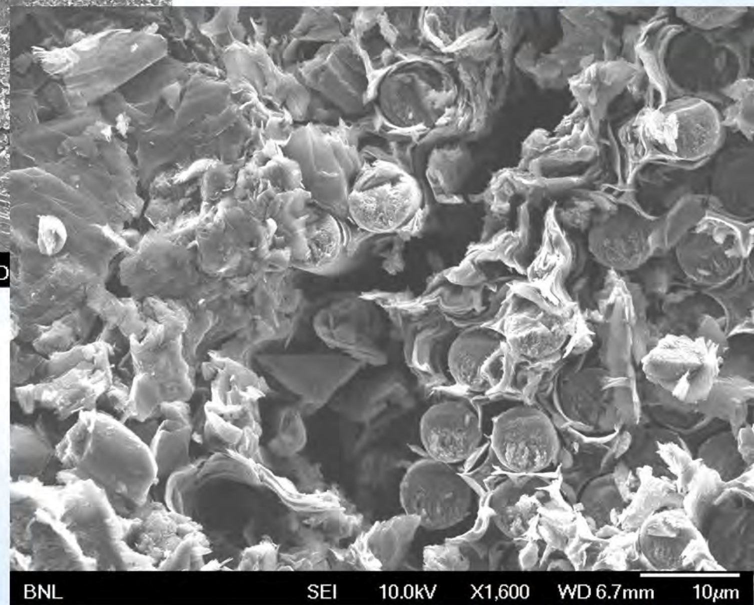
O-14	1.30 · 10-2 Ci/sec
O-15	1.50 · 10-1 Ci/sec
N-13	5.89 · 10-3 Ci/sec
C-11	6.66 · 10-3 Ci/sec

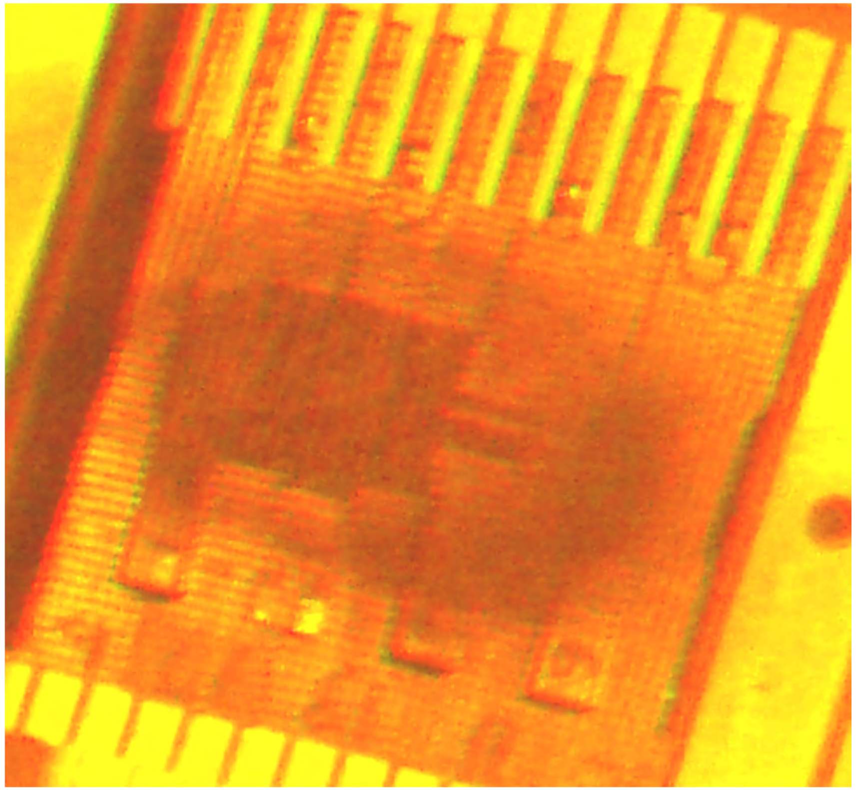
Material	Motivation
C-C Composite (3D)	Observed damage at low dose at BNL BLIP
POCO ZXF-5Q	NuMI/NOvA target material
Toyo-Tanso IG-430	Nuclear grade for T2K
Carbone-Lorraine 2020	CNGS target material
SGL R7650	NuMI/NOvA baffle material
St.-Gobain AX05 h-BN	Hexagonal Boron Nitride

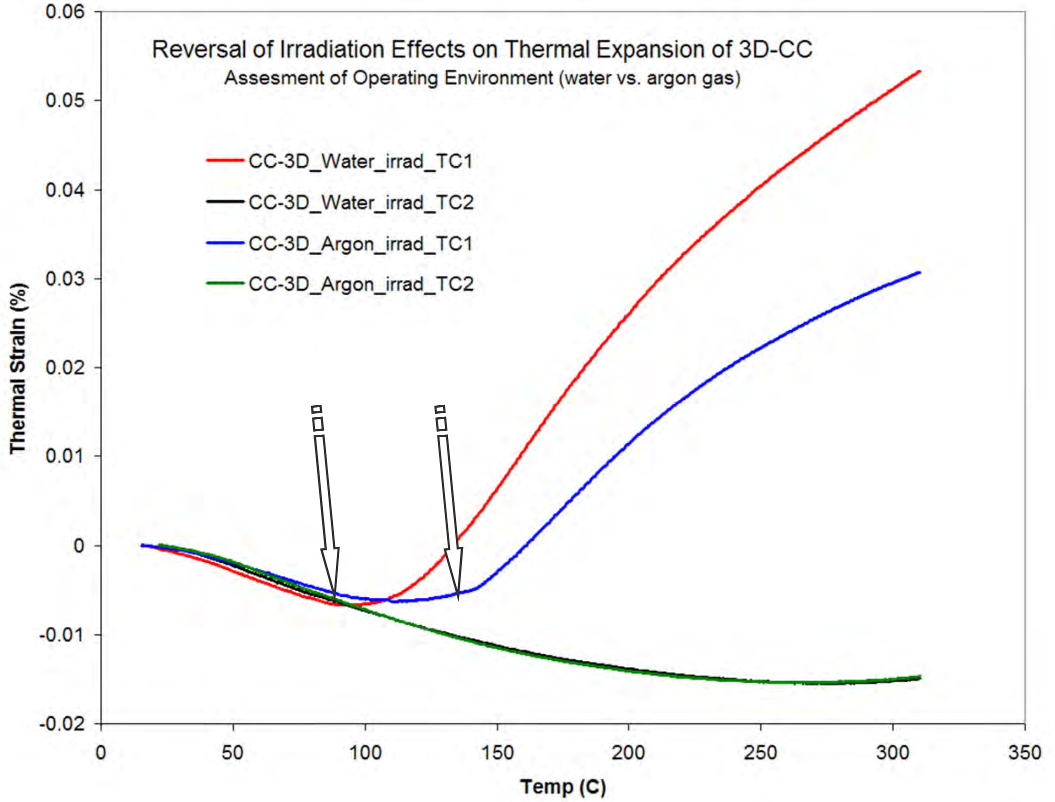
Carbon/Carbon Composite

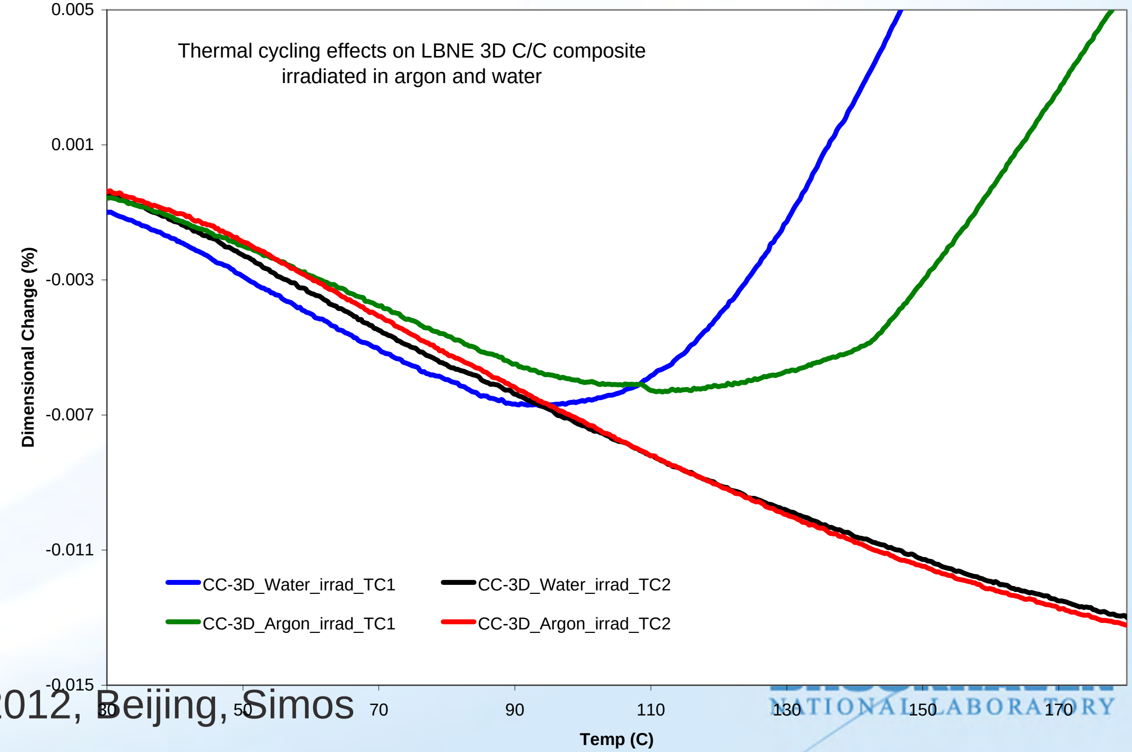









Reversal of Irradiation Effects on Thermal Expansion of 3D-CC
Assesment of Operating Environment (water vs. argon gas)
CC-3D_Water_irrad_TC1
CC-3D_Water_irrad_TC2
CC-3D_Argon_irrad_TC1
CC-3D_Argon_irrad_TC2
Thermal Strain (%)
Temp (C)
0.06
0.05
0.04
0.03
0.02
0.01
0
-0.01
-0.02
0
50
100
150
200
250
300
350

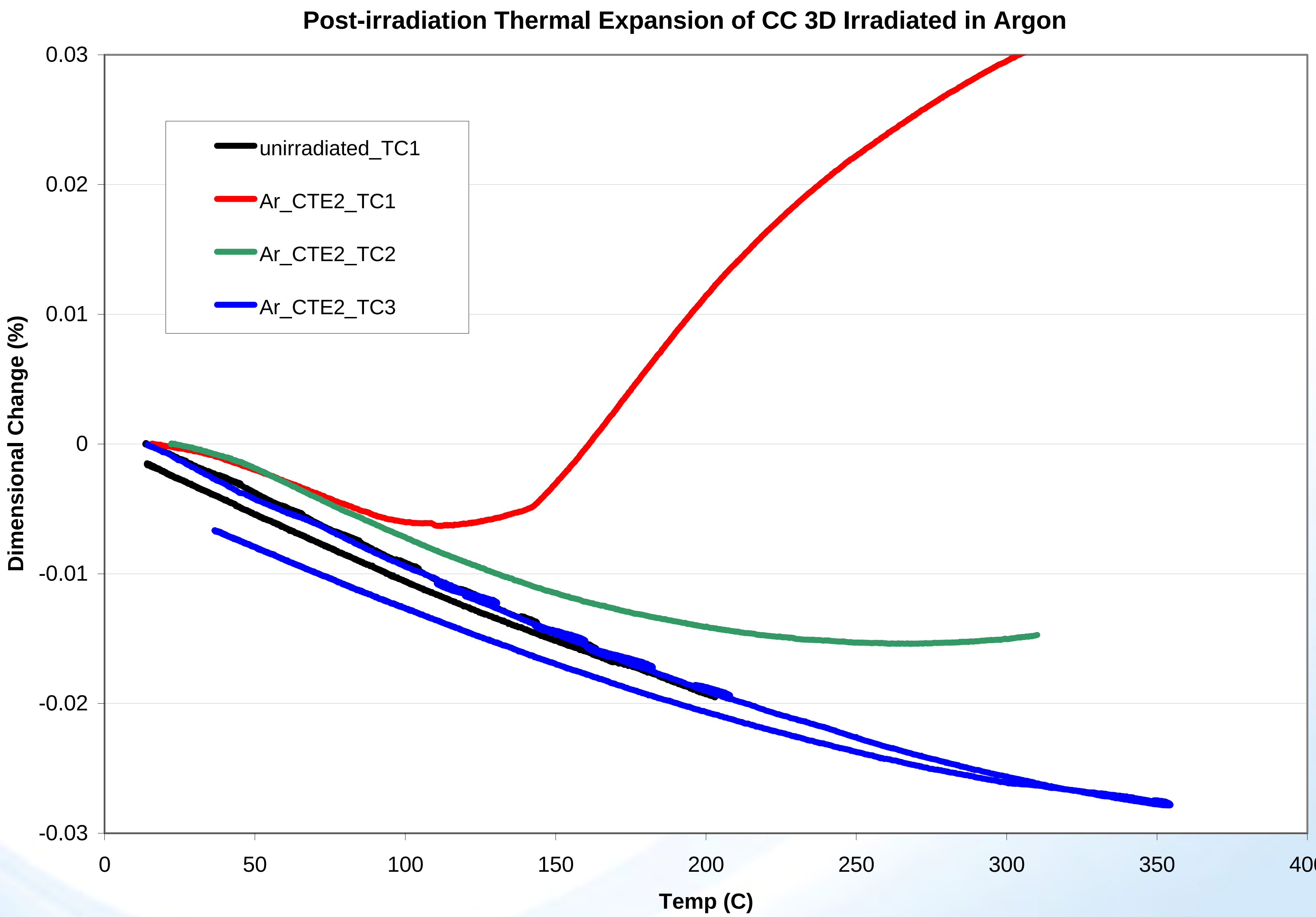

Thermal cycling effects on LBNE 3D C/C composite
irradiated in argon and water
CC-3D_Water_irrad_TC1
CC-3D_Water_irrad_TC2
CC-3D_Argon_irrad_TC1
CC-3D_Argon_irrad_TC2
Dimensional Change (%)
Temp (C)
0.005
0.001
-0.003
-0.007
-0.011
-0.015
30
50
70
90
110
130
150
170

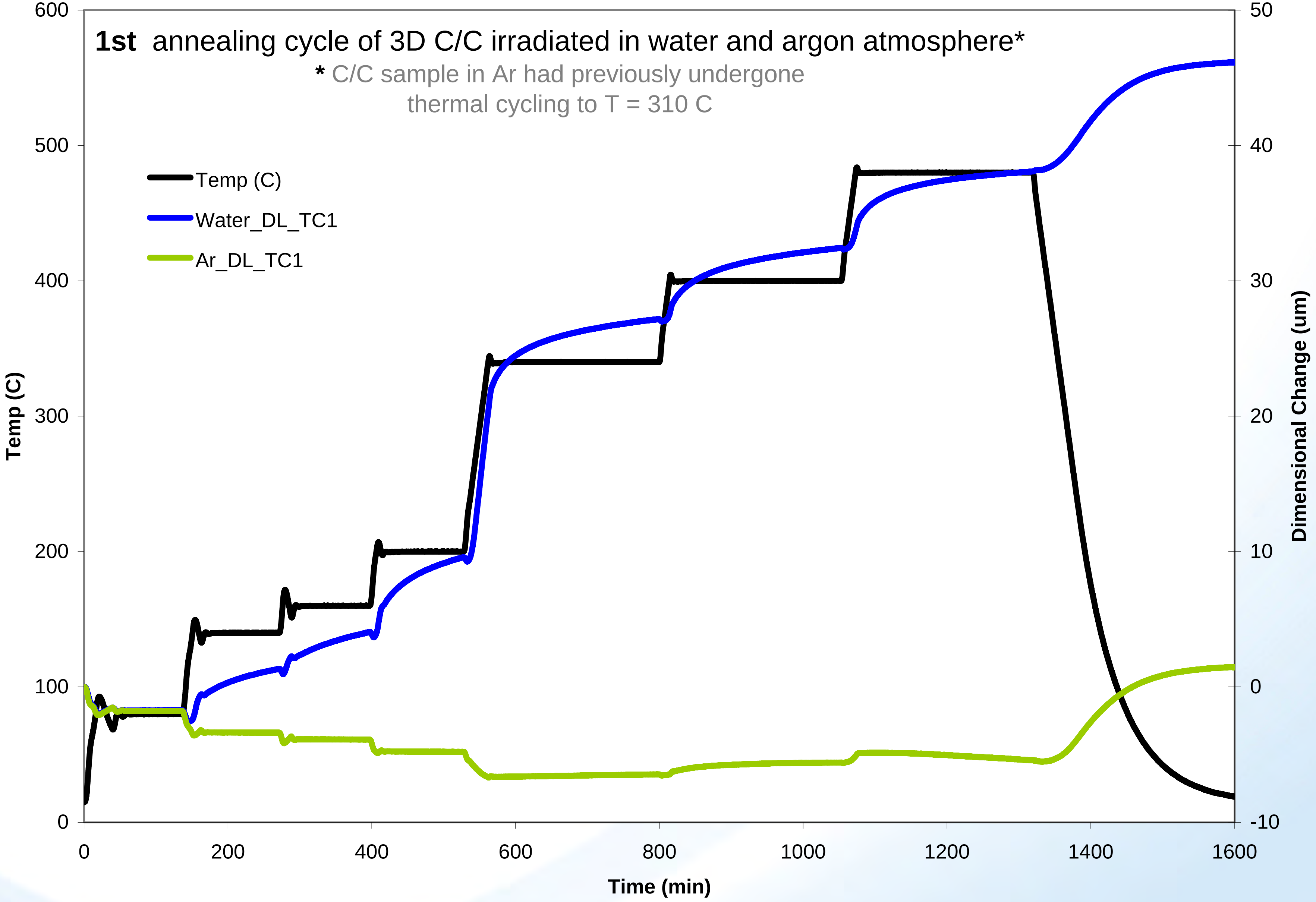
Weight Loss Measurements of Irradiated C/C Composite

	Water-cooled C/C	**Argon Environment C/C**
Unirradiated mean weight	0.708 grams	0.708 grams
Temperature (^oC)	W(g)	W(g)
RT (irradiated)	0.4938	0.7157
475 (1)	0.4834	0.7106
475 (2)	0.4488	0.6837
670	0.4010	0.6341

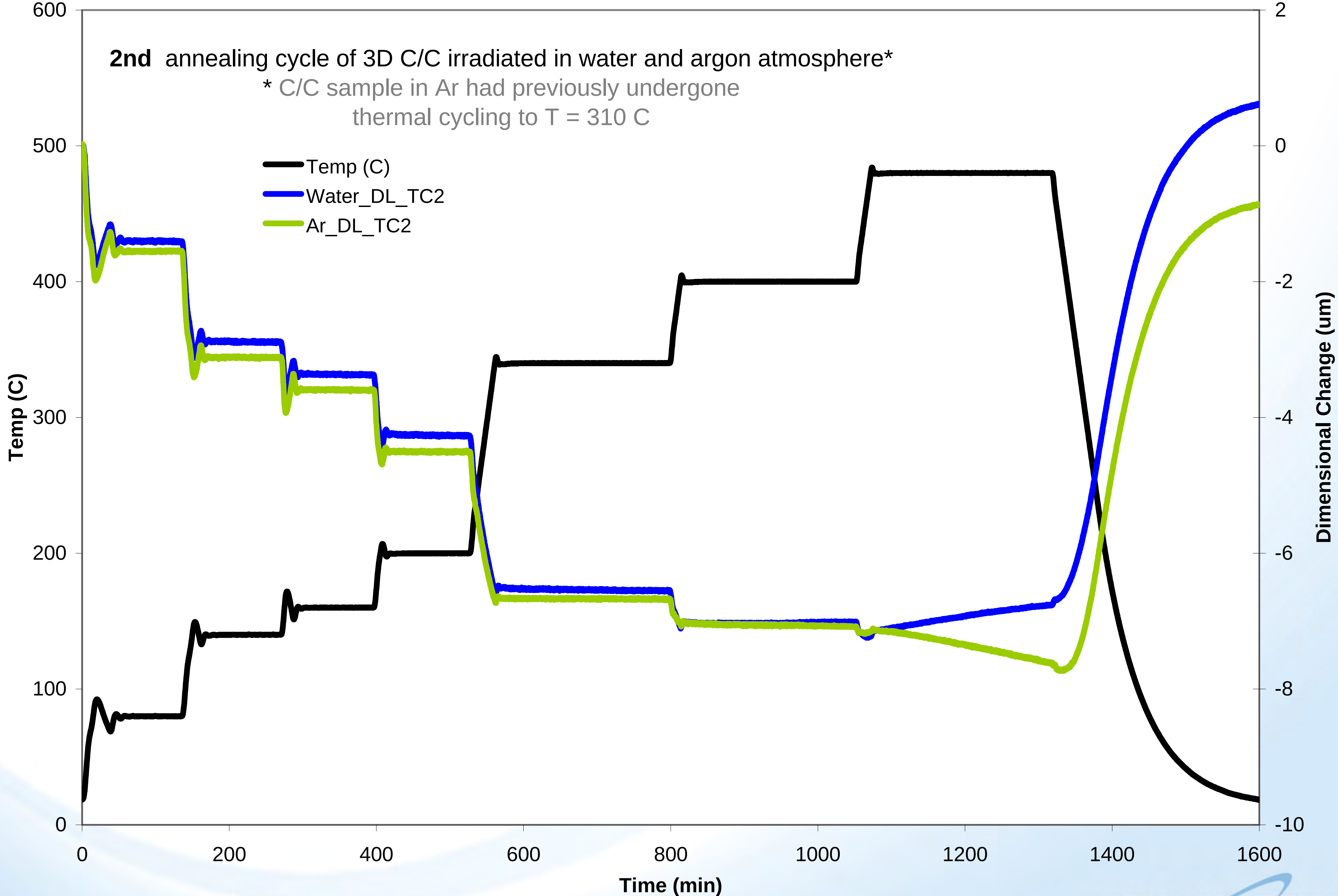
981 μCi ➔ 0.7157 grams

3,110 μCi ➔0.7214 grams

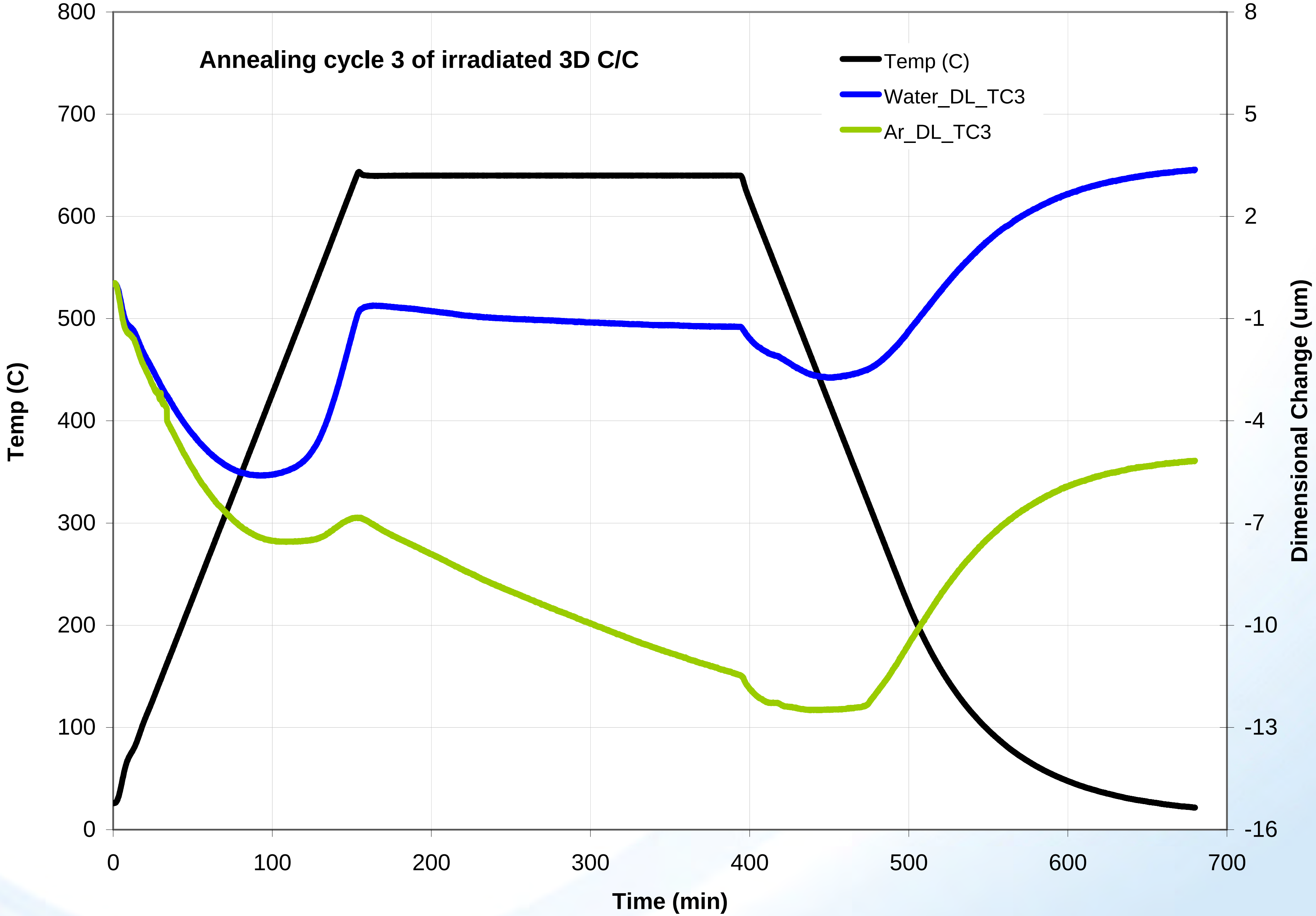

Post-irradiation Thermal Expansion of CC 3D Irradiated in Argon
Dimensional Change (%)
Temp (C)
0.03
0.02
0.01
0
-0.01
-0.02
-0.03
0
50
100
150
200
250
300
350
400
unirradiated_TC1
Ar_CTE2_TC1
Ar_CTE2_TC2
Ar_CTE2_TC3

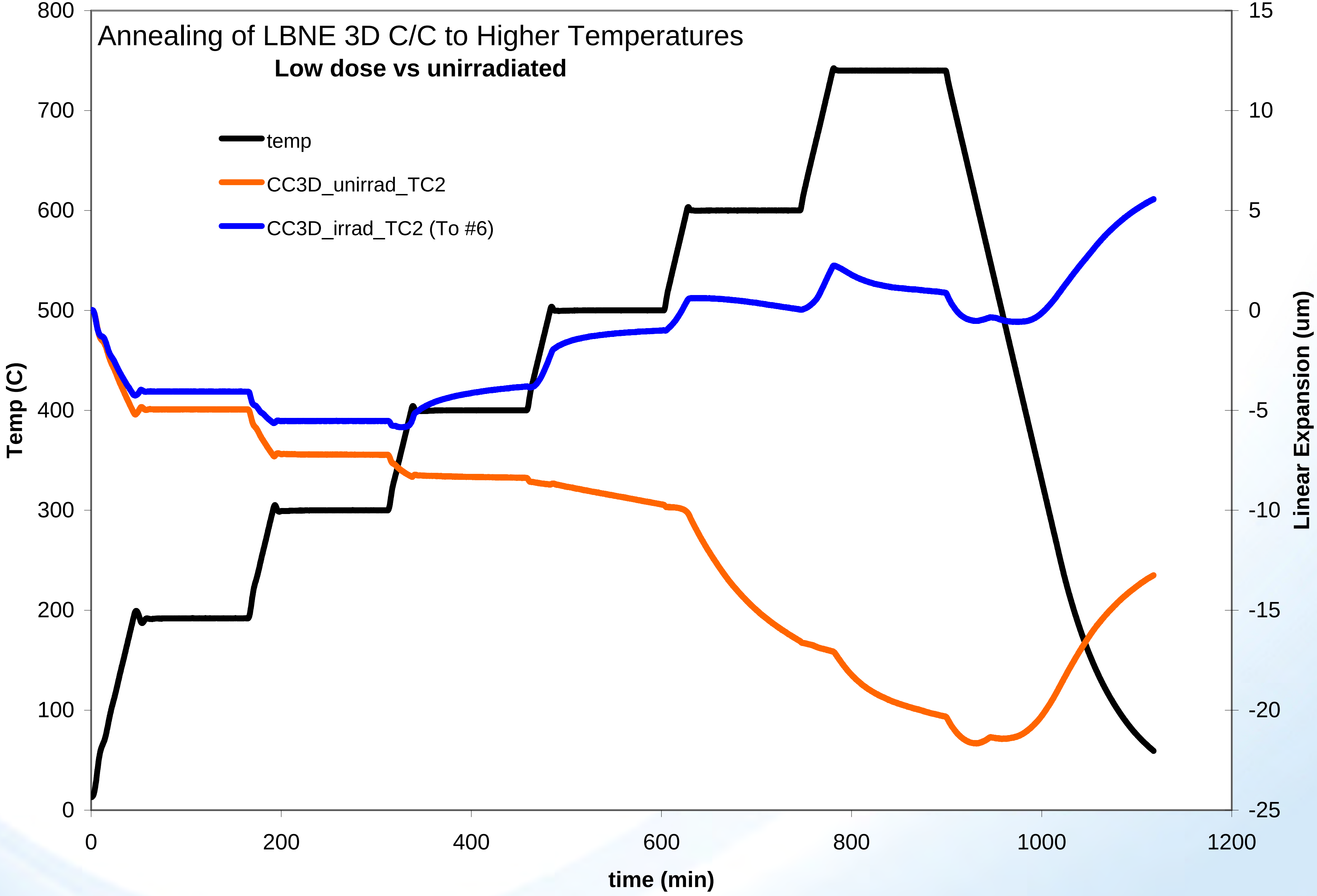


1st annealing cycle of 3D C/C irradiated in water and argon atmosphere*
* C/C sample in Ar had previously undergone
thermal cycling to T = 310 C
Temp (C)
Water_DL_TC1
Ar_DL_TC1
600
500
400
300
200
100
0
50
40
30
20
10
0
-10
0
200
400
600
800
1000
1200
1400
1600
Time (min)
Dimensional Change (um)



2nd annealing cycle of 3D C/C irradiated in water and argon atmosphere*
* C/C sample in Ar had previously undergone thermal cycling to T = 310 C
Temp (C)
Water_DL_TC2
Ar_DL_TC2
Temp (C)
Dimensional Change (um)
Time (min)
0
100
200
300
400
500
600
2
0
-2
-4
-6
-8
-10
0
200
400
600
800
1000
1200
1400
1600


Annealing cycle 3 of irradiated 3D C/C
Temp (C)
Water_DL_TC3
Ar_DL_TC3
800
700
600
500
400
300
200
100
0
8
5
2
-1
-4
-7
-10
-13
-16
0
100
200
300
400
500
600
700
Time (min)
Dimensional Change (um)



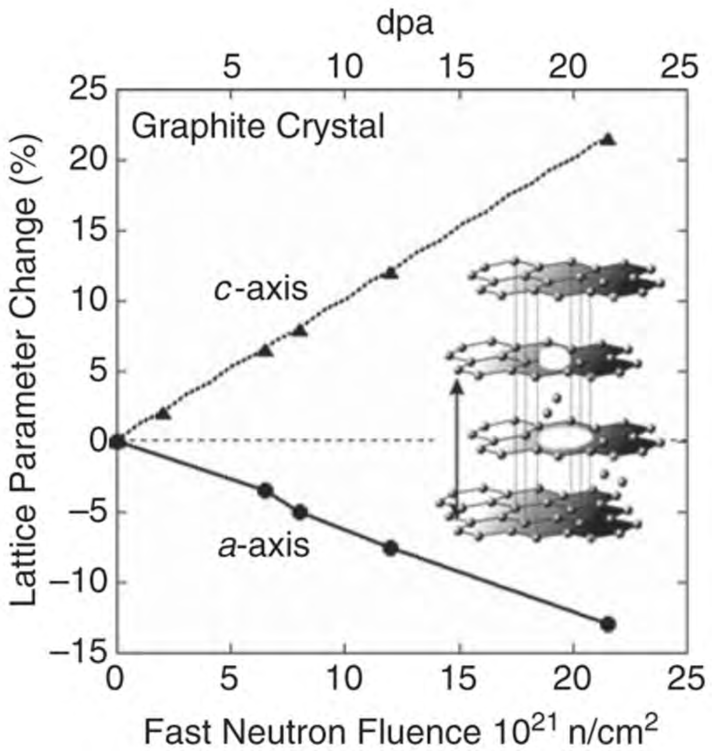
Annealing of LBNE 3D C/C to Higher Temperatures
Low dose vs unirradiated
temp
CC3D_unirrad_TC2
CC3D_irrad_TC2 (To #6)
Temp (C)
Linear Expansion (um)
time (min)
800
700
600
500
400
300
200
100
0
15
10
5
0
-5
-10
-15
-20
-25
0
200
400
600
800
1000
1200

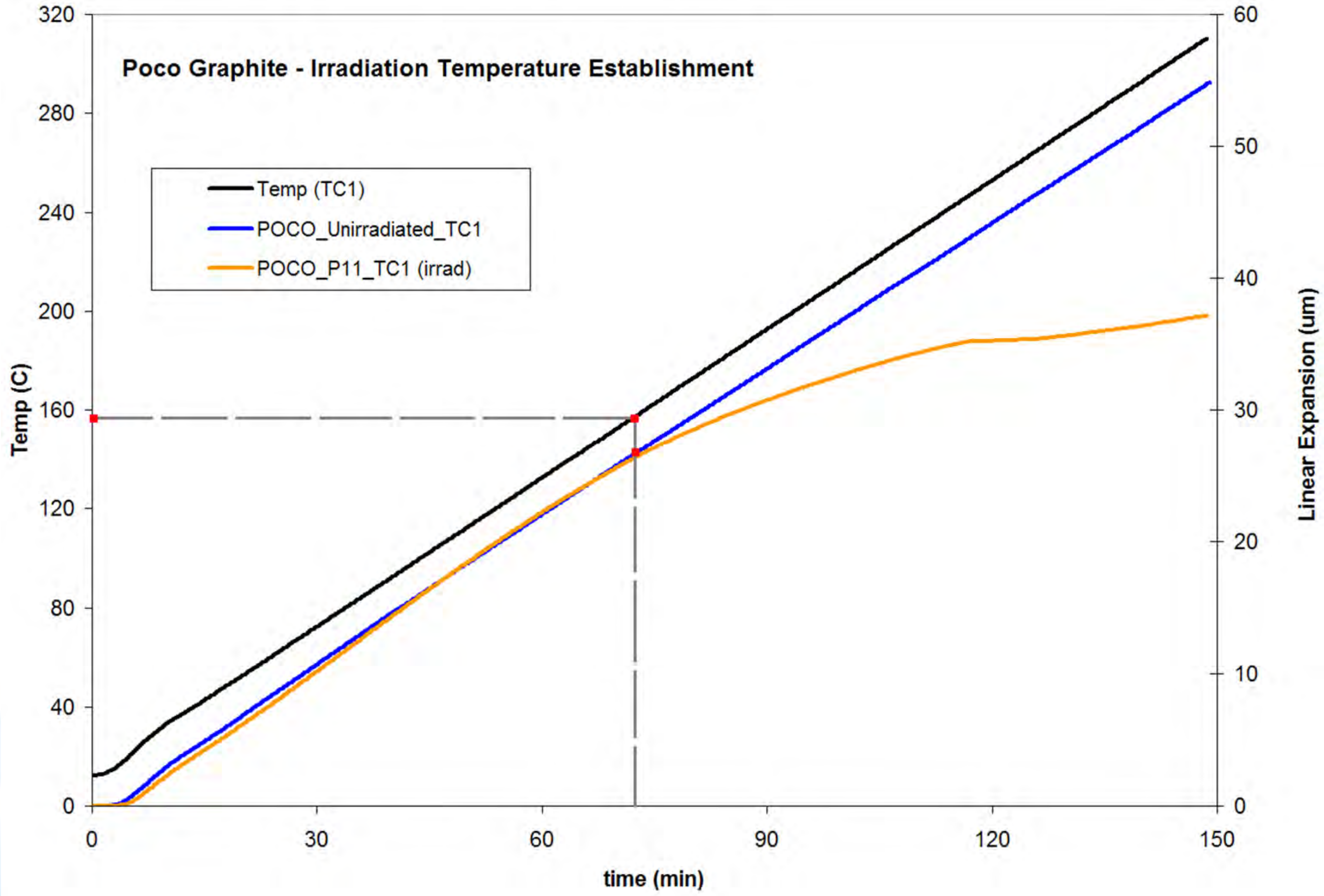
Graphite – Irradiation Damage & Annealing

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 ..")

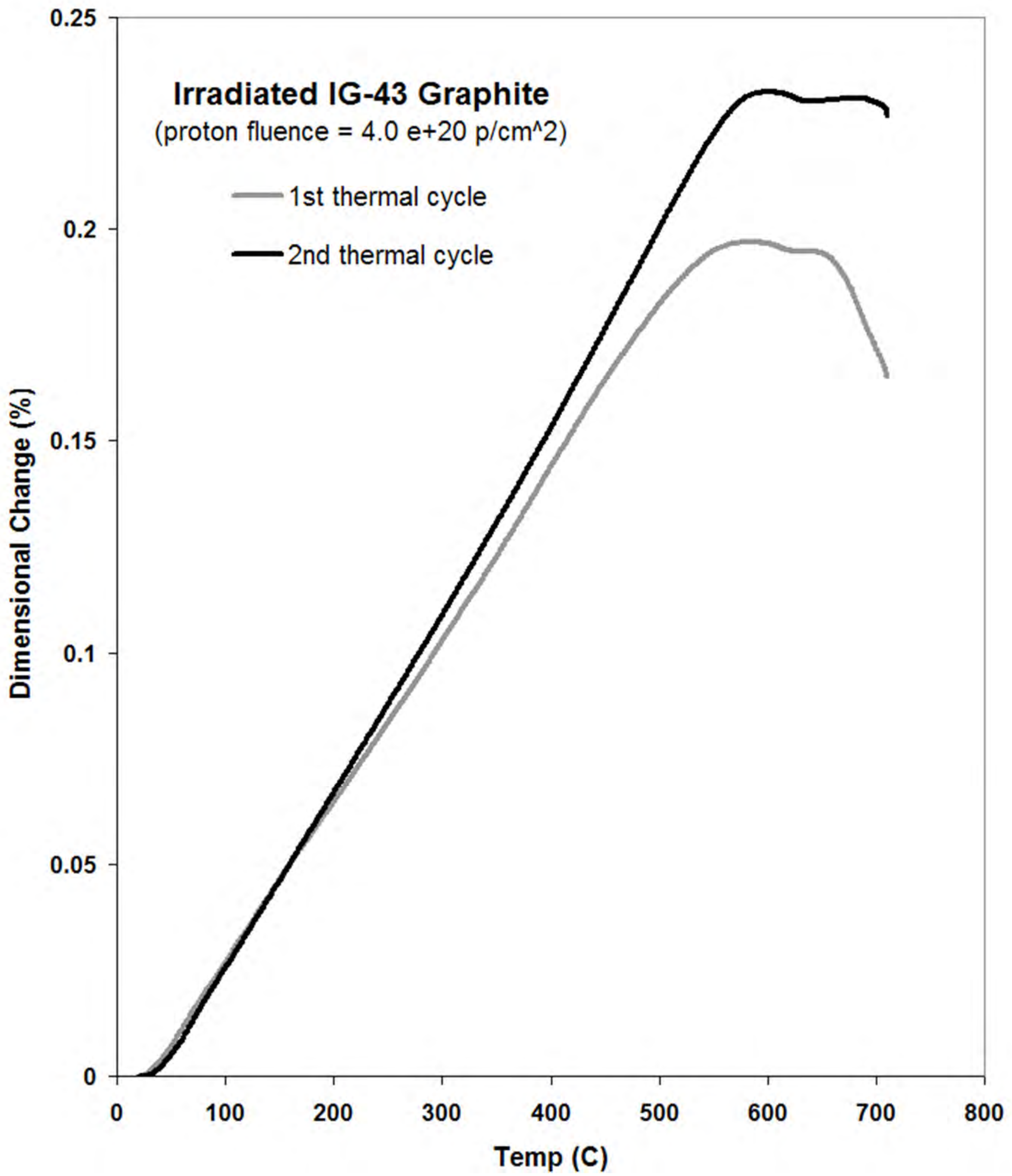
A displaced interstitial will undergo many collisions until its energy is reduced to values corresponding to lattice temperature. In process some interstitials remain in stable configuration and some anneal immediately. Those that do not anneal cause an increase in the dimensions of the sample.

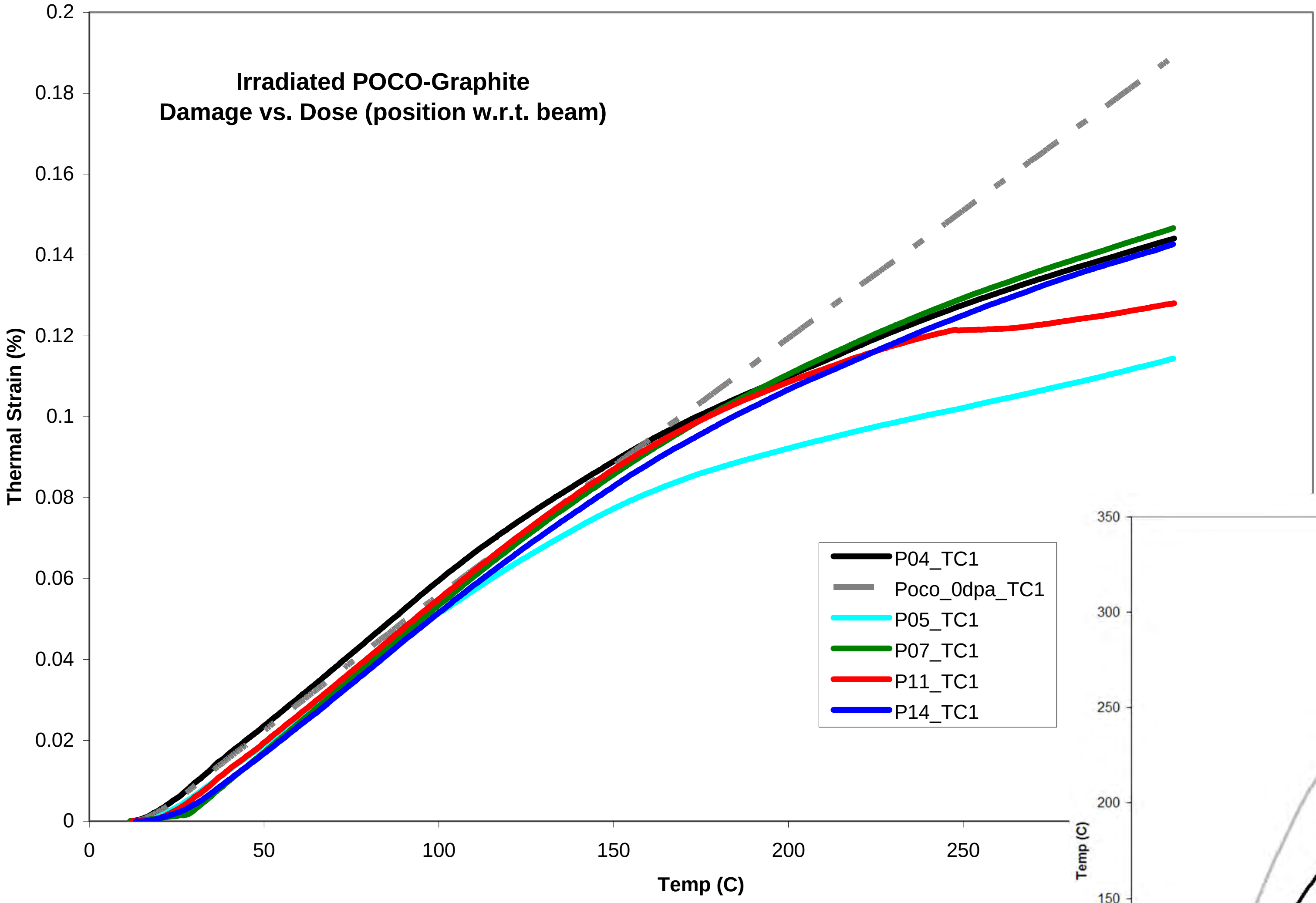
As shown here, when annealing above the irradiation temperature the dimensional change dips because more "stable" interstitials are leaving the temporary locations between lattice planes and return to them.



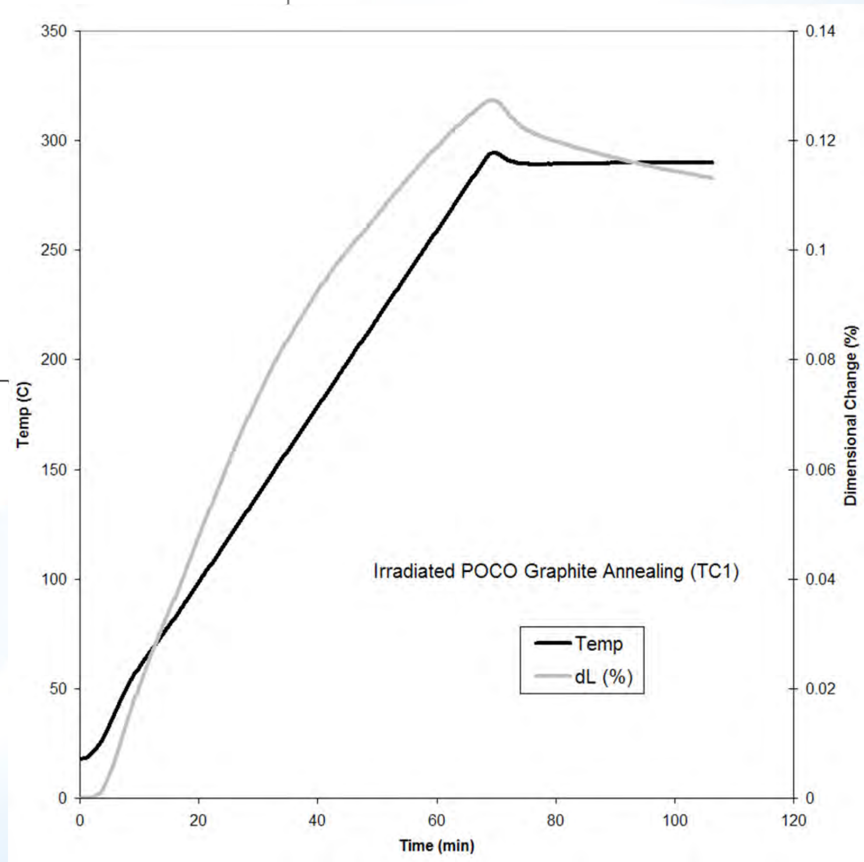


High temp. annealing

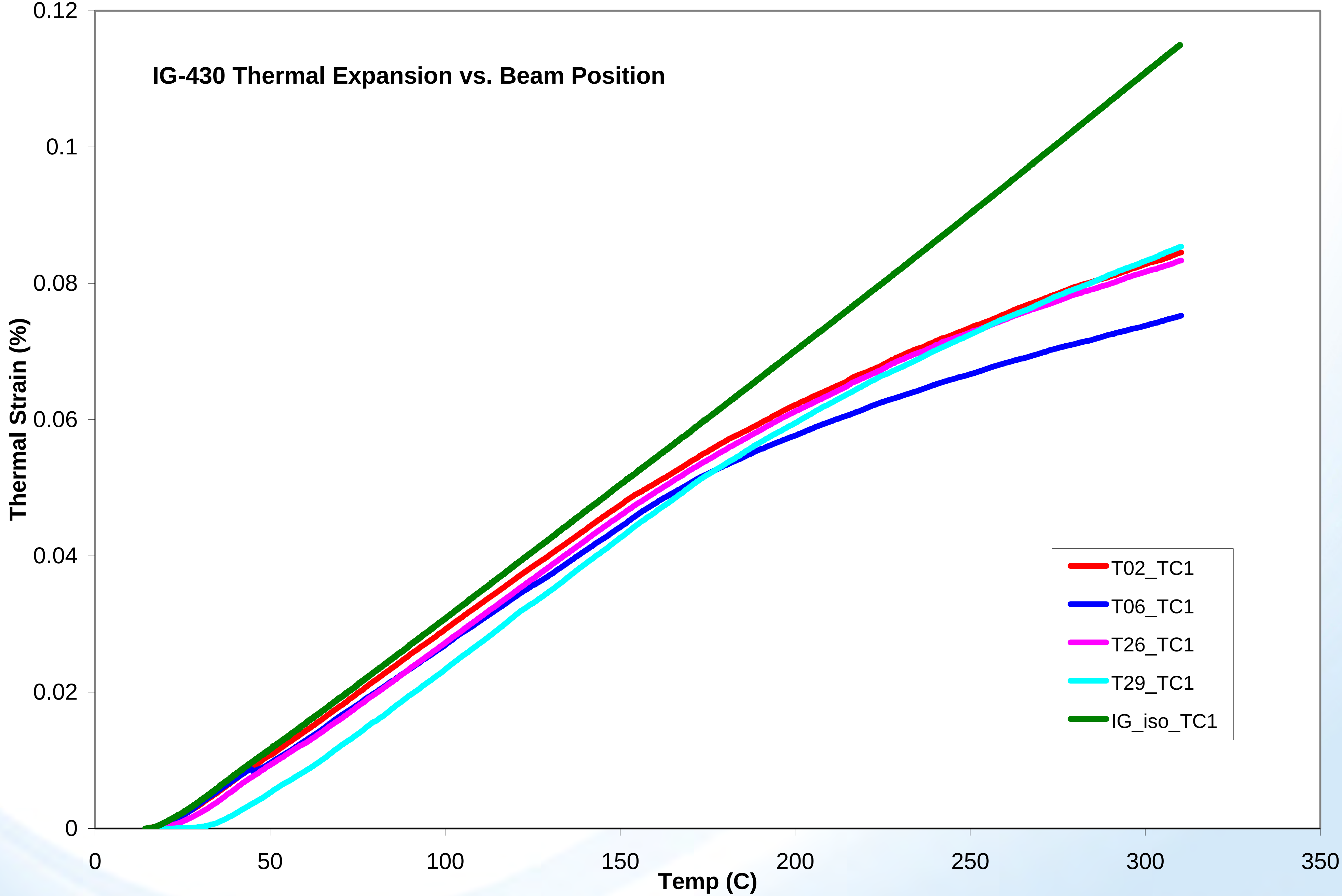


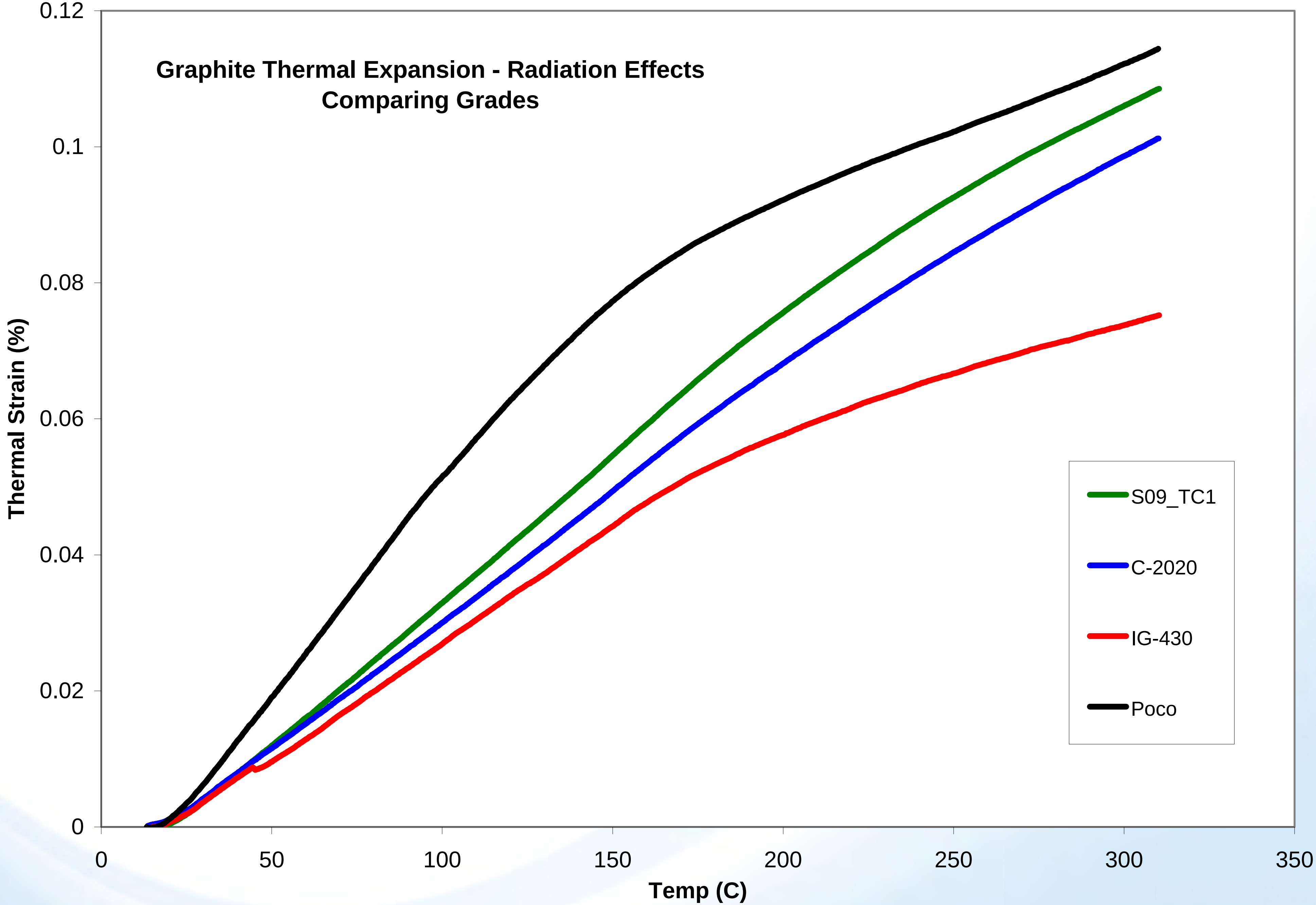


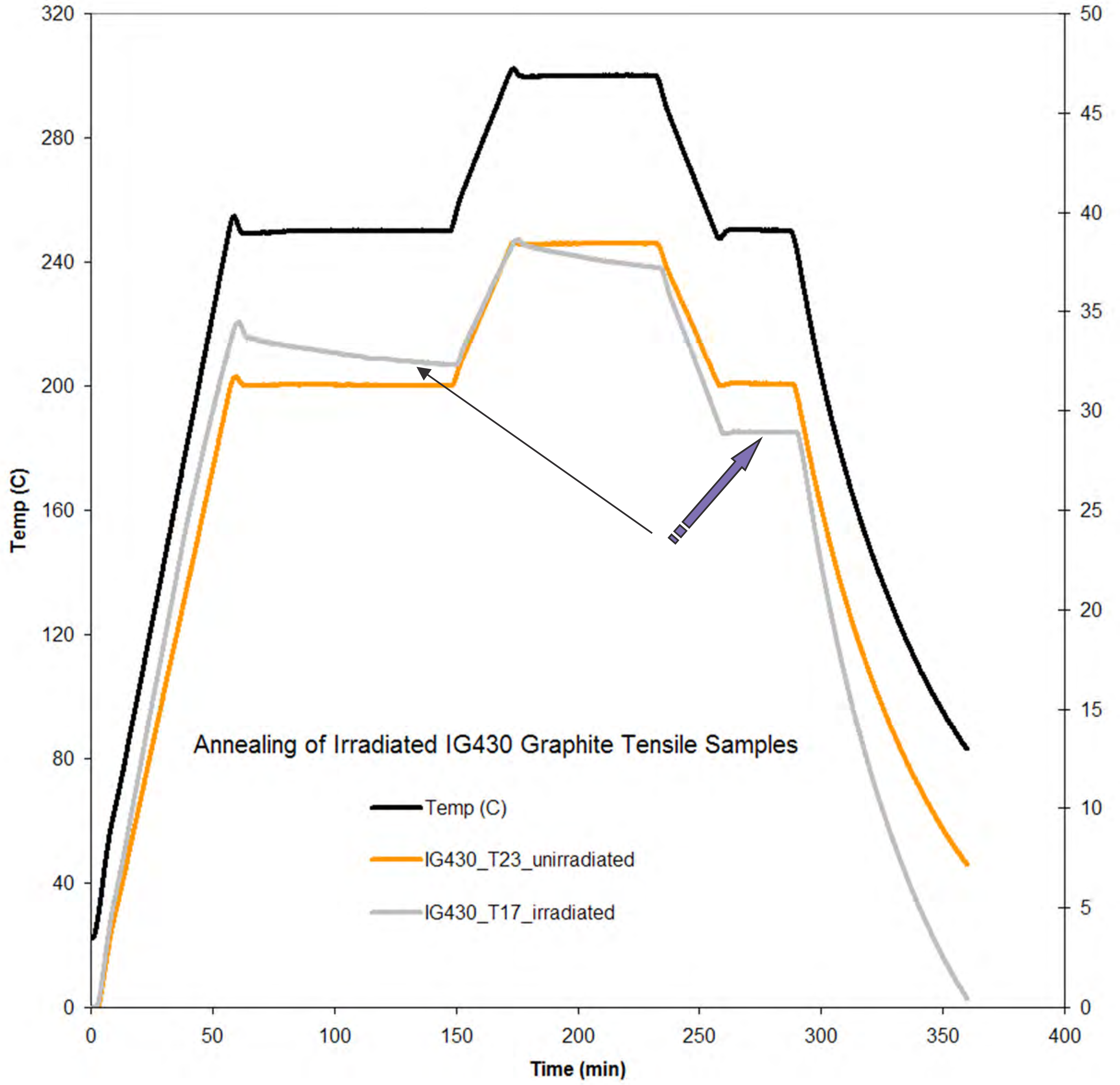
Irradiated POCO-Graphite
Damage vs. Dose (position w.r.t. beam)
Thermal Strain (%)
Temp (C)
P04_TC1
Poco_0dpa_TC1
P05_TC1
P07_TC1
P11_TC1
P14_TC1



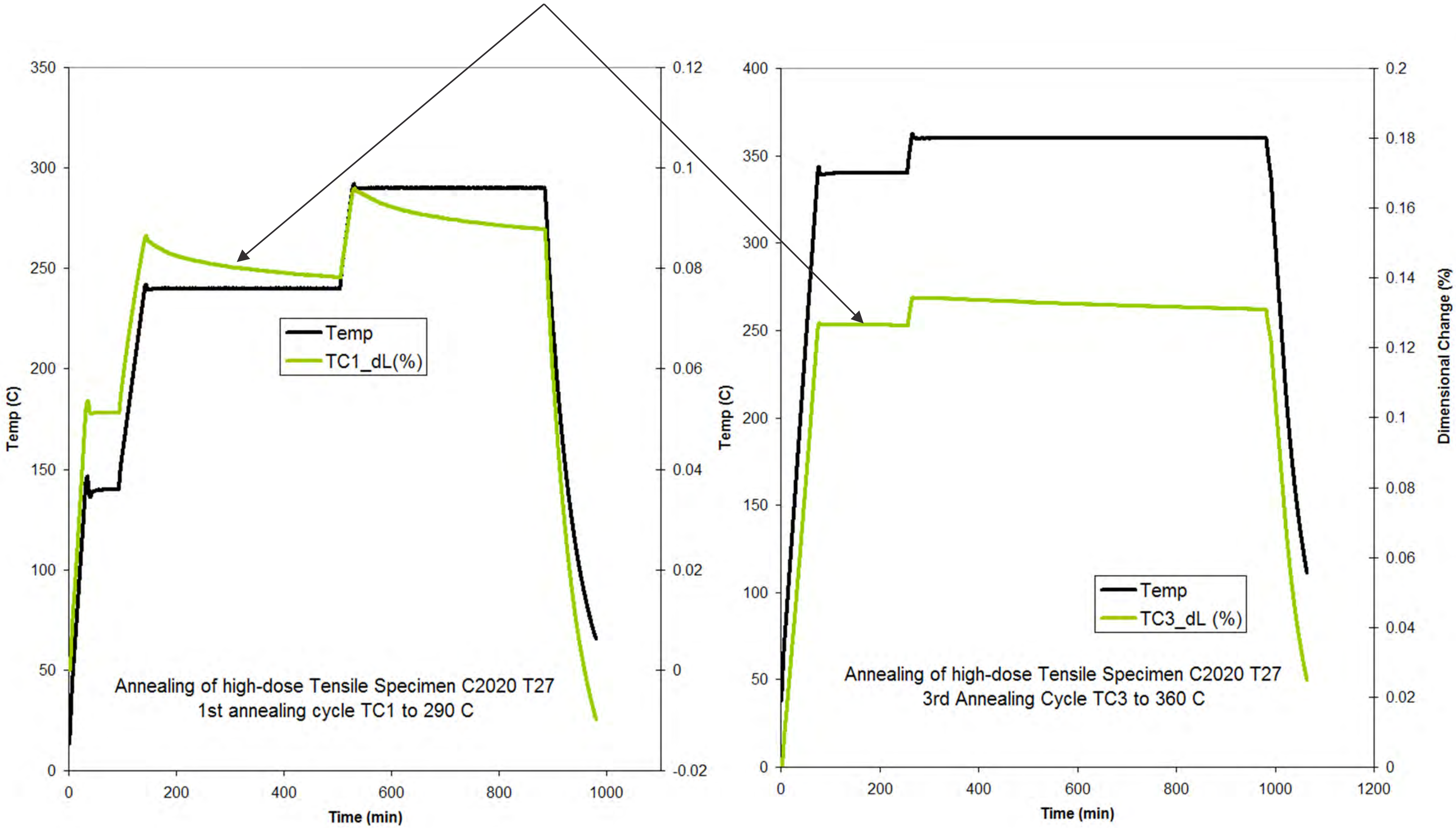
Irradiated POCO Graphite Annealing (TC1)
Temp (C)
Dimensional Change (%)
Time (min)
Temp
dL (%)


IG-430 Thermal Expansion vs. Beam Position
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)
T02_TC1
T06_TC1
T26_TC1
T29_TC1
IG_iso_TC1

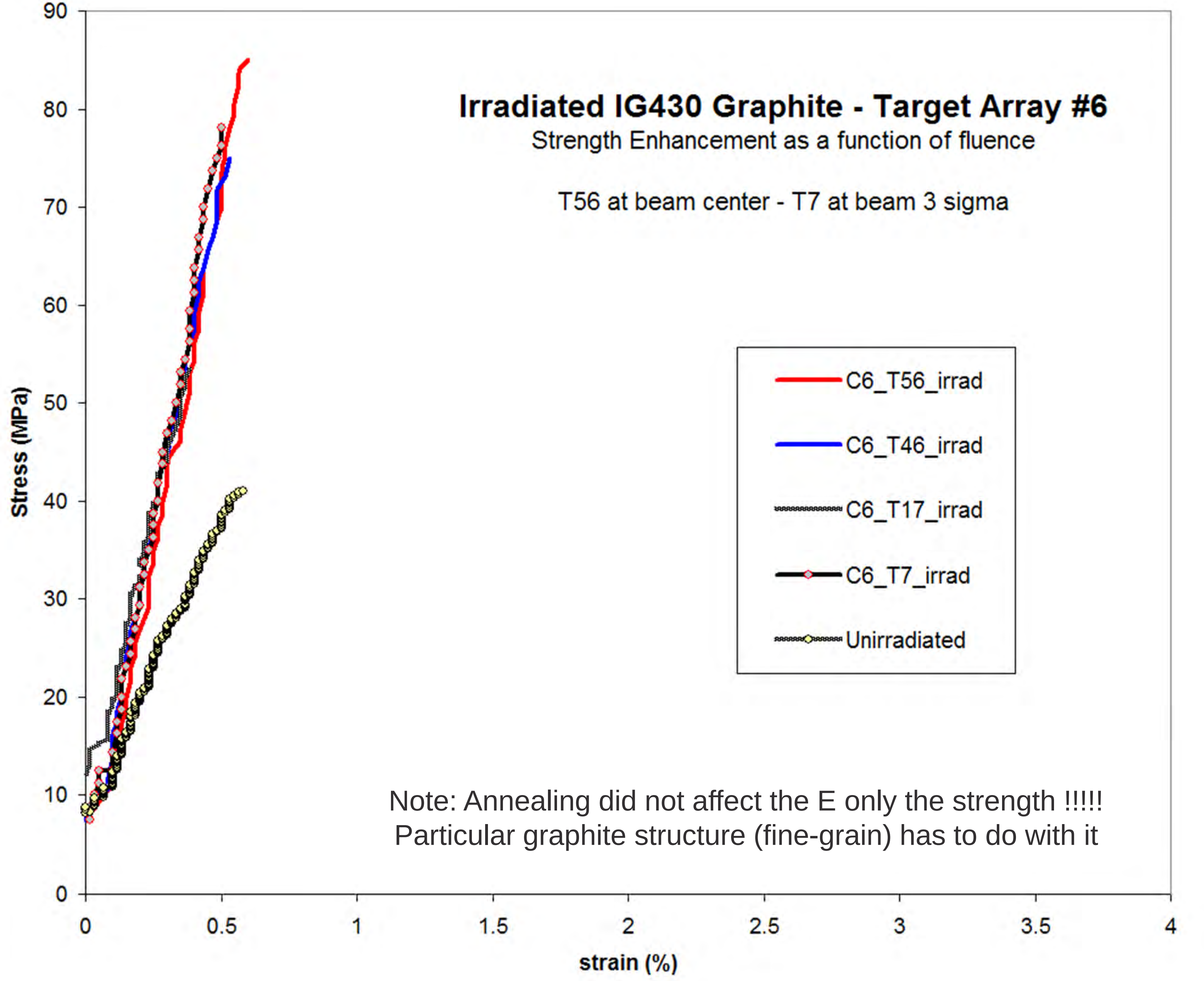

Graphite Thermal Expansion - Radiation Effects
Comparing Grades
Thermal Strain (%)
0.12
0.1
0.08
0.06
0.04
0.02
0
0
50
100
150
200
250
300
350
Temp (C)
S09_TC1
C-2020
IG-430
Poco



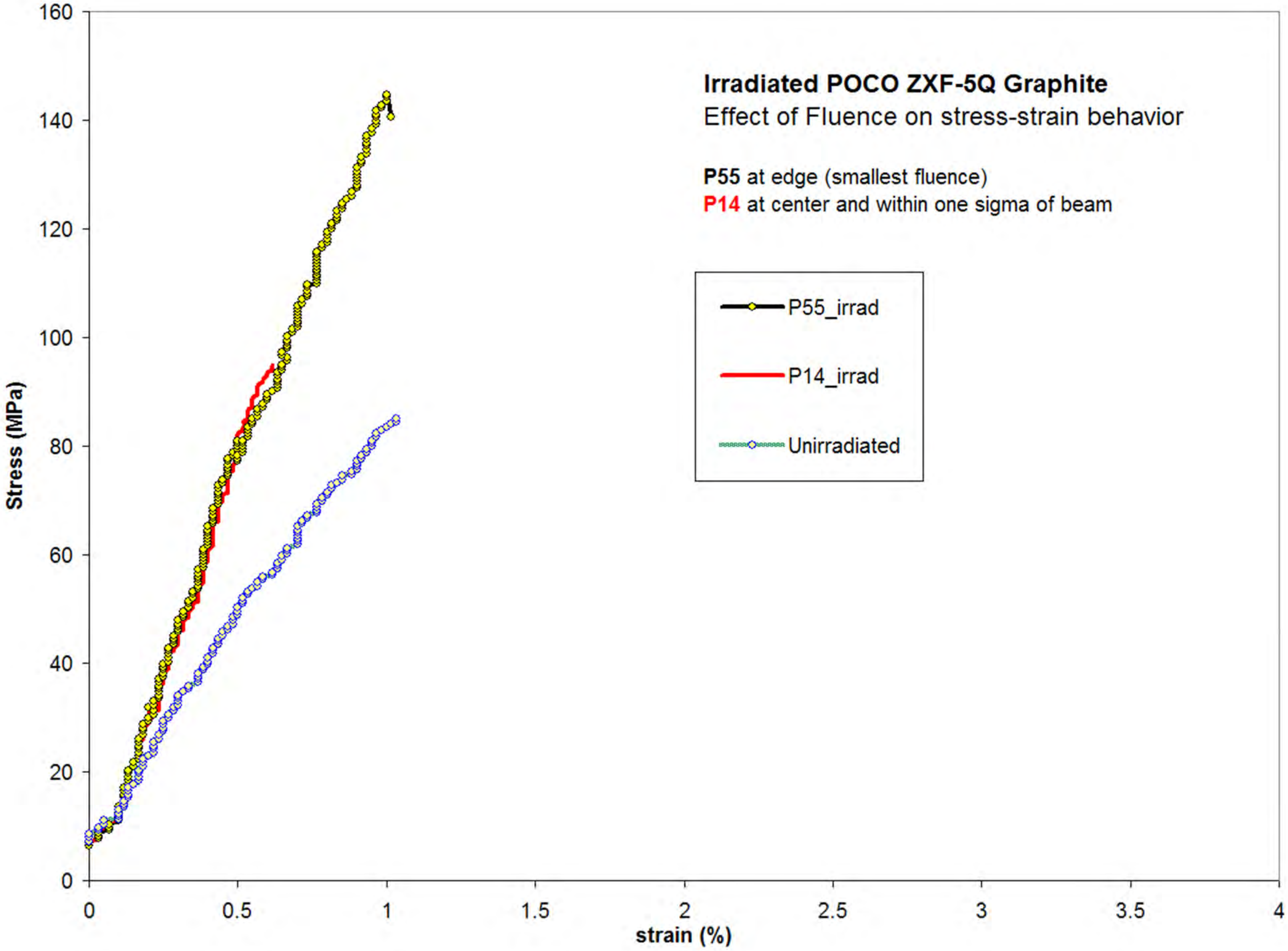
Annealing of Irradiated IG430 Graphite Tensile Samples
Temp (C)
IG430_T23_unirradiated
IG430_T17_irradiated
Time (min)
0
50
100
150
200
250
300
350
400
40
80
120
160
200
240
280
320
5
10
15
20
25
30
35
40
45
50



Temp
TC1_dL(%)
Annealing of high-dose Tensile Specimen C2020 T27
1st annealing cycle TC1 to 290 C
Temp (C)
Time (min)
Temp
TC3_dL (%)
Annealing of high-dose Tensile Specimen C2020 T27
3rd Annealing Cycle TC3 to 360 C
Temp (C)
Dimensional Change (%)
Time (min)



Irradiated IG430 Graphite - Target Array #6
Strength Enhancement as a function of fluence
T56 at beam center - T7 at beam 3 sigma
C6_T56_irrad
C6_T46_irrad
C6_T17_irrad
C6_T7_irrad
Unirradiated
Note: Annealing did not affect the E only the strength !!!!!
Particular graphite structure (fine-grain) has to do with it
Stress (MPa)
strain (%)
0
10
20
30
40
50
60
70
80
90
0
0.5
1
1.5
2
2.5
3
3.5
4


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 (%)
0
20
40
60
80
100
120
140
160
0
0.5
1
1.5
2
2.5
3
3.5
4

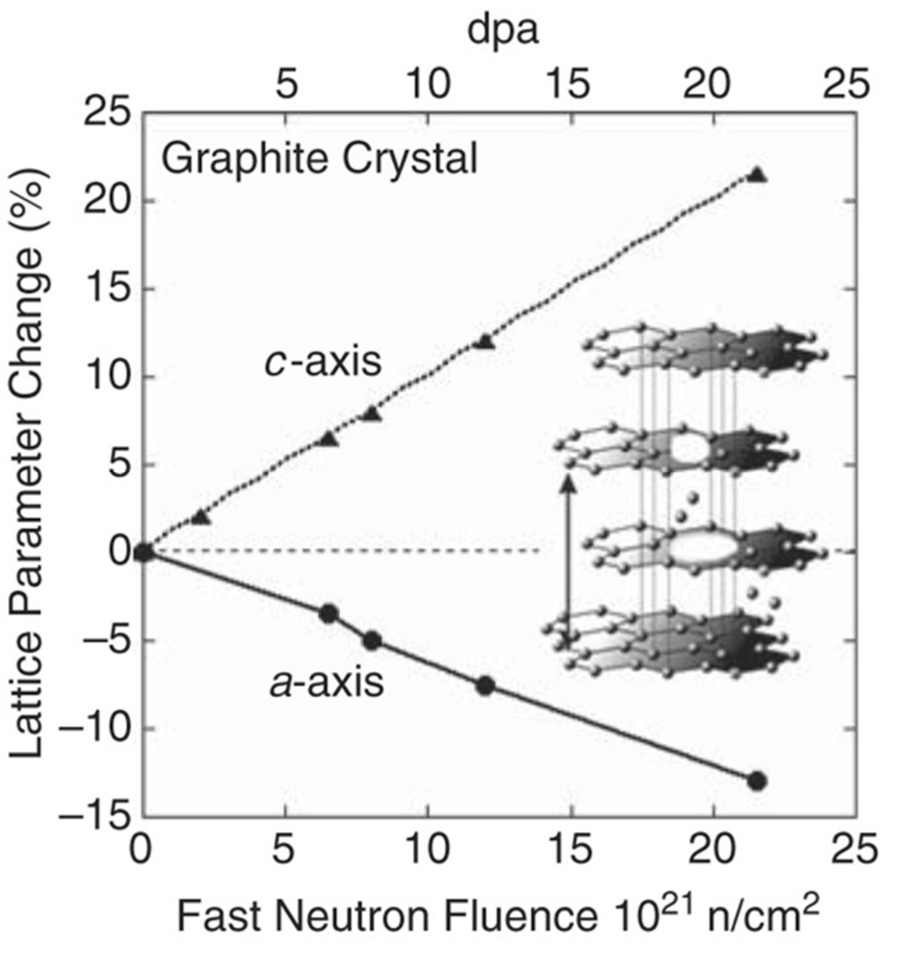
Non-destructive testing of graphite damage “annealing” with ultrasound

Observe through annealing

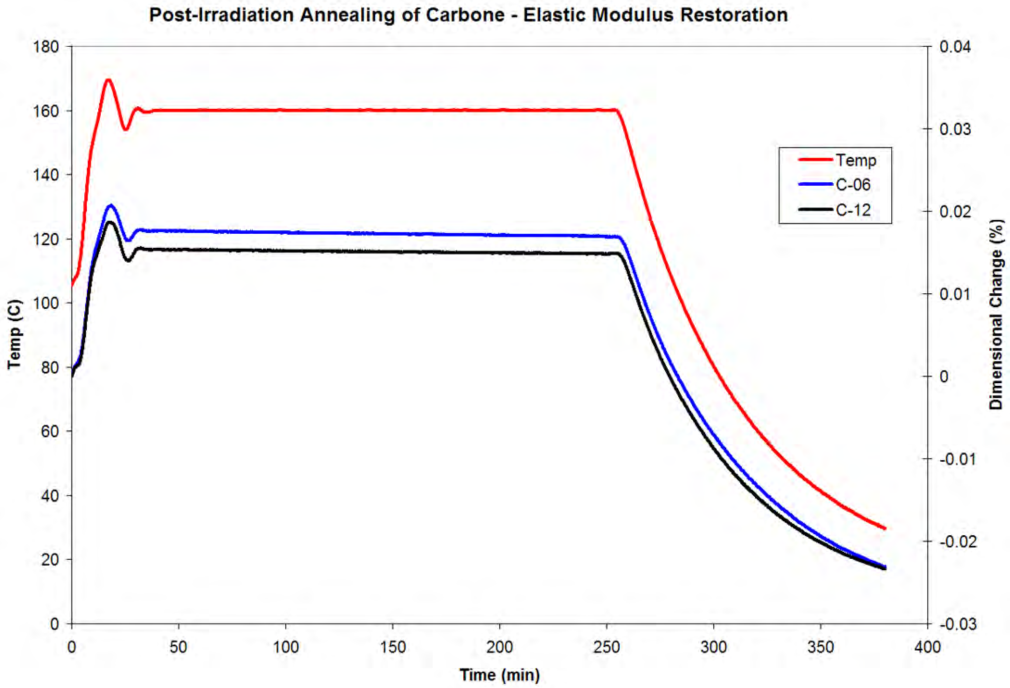
speed of sound change

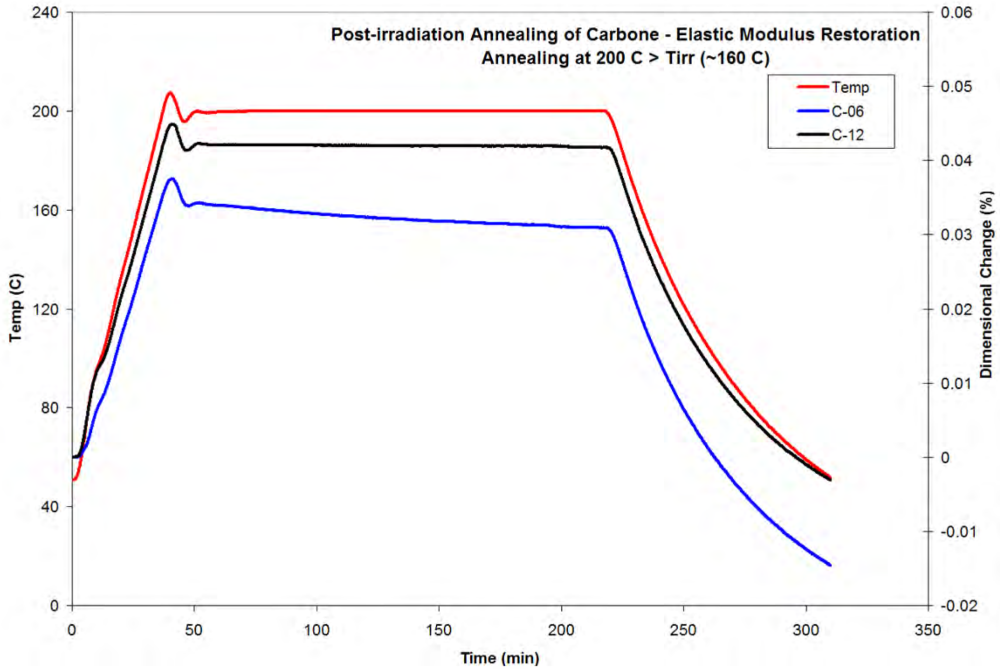
dimensional change

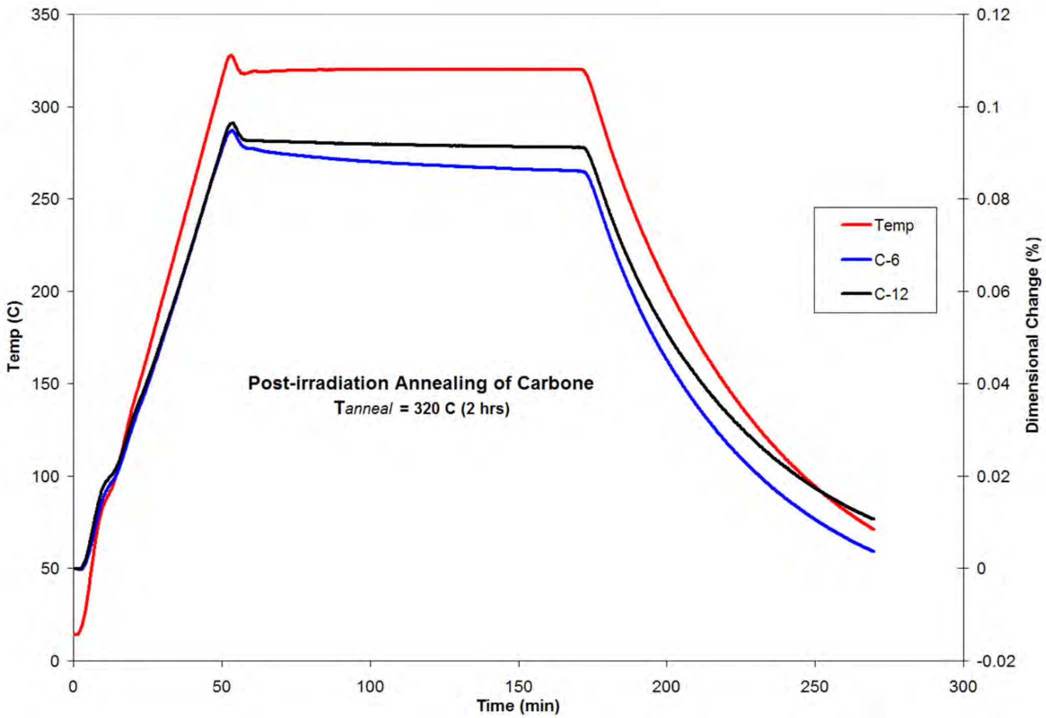
weight change

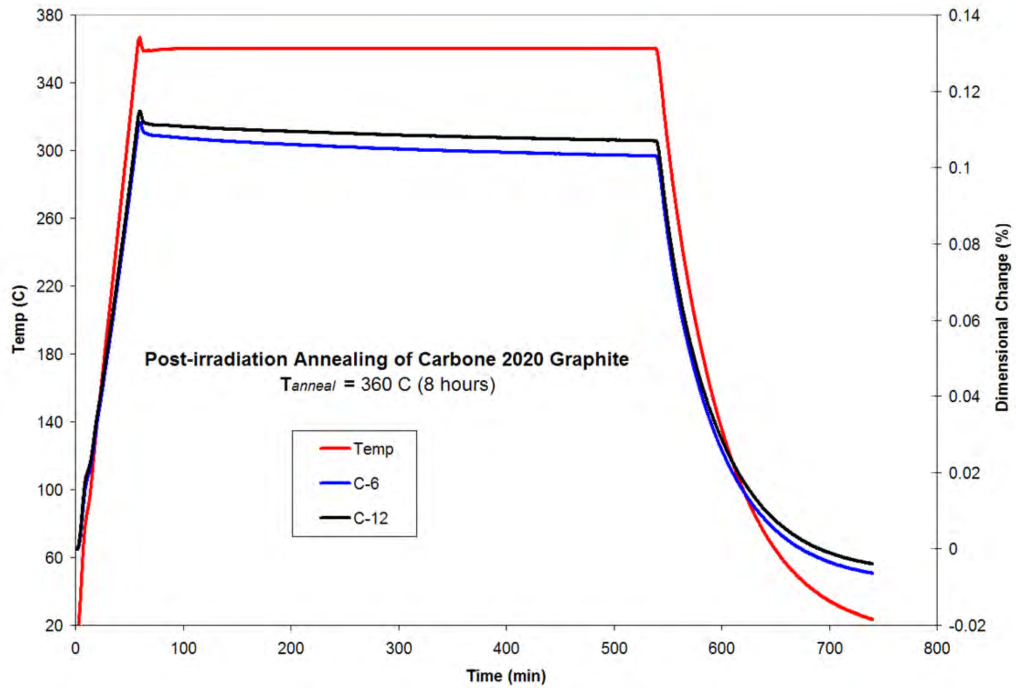


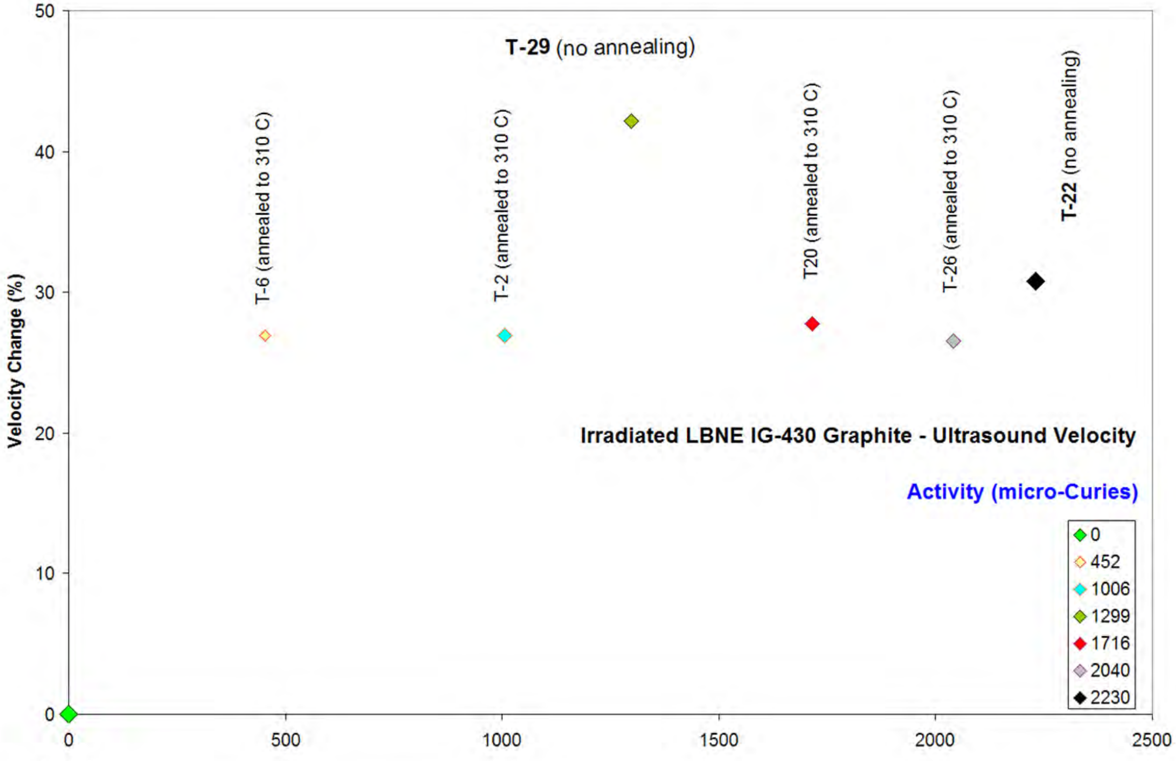
Multi-stage thermal annealing of irradiated Carbone-2020

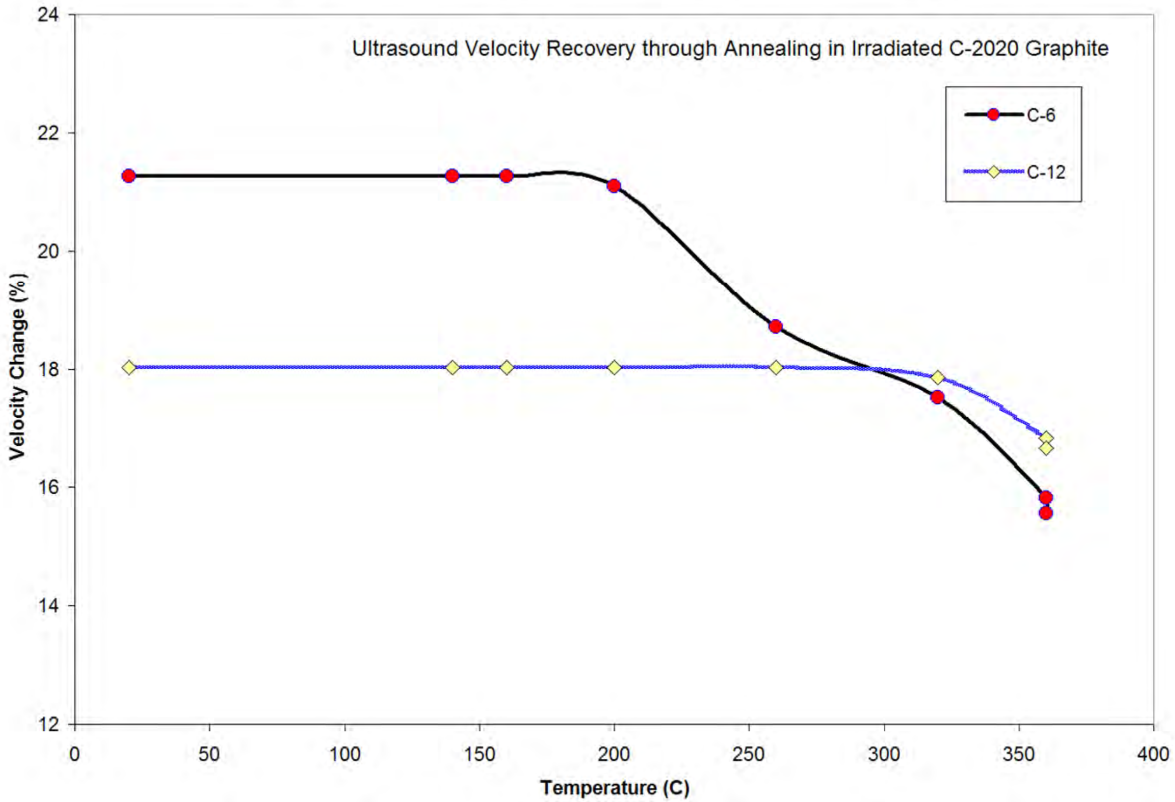












While for a complete determination of the combined influence of irradiation and temperature induced changes in graphite strength will require a series of additional well controlled processes such as:

- measurements of shear wave velocities that will help determine Poisson's ratio v,
- wave propagation principles through the porous structure of graphite to account for wave-pore interaction and
- quantification of contribution on dimensional changes from irradiation and from temperature so the density variation can be accounted for

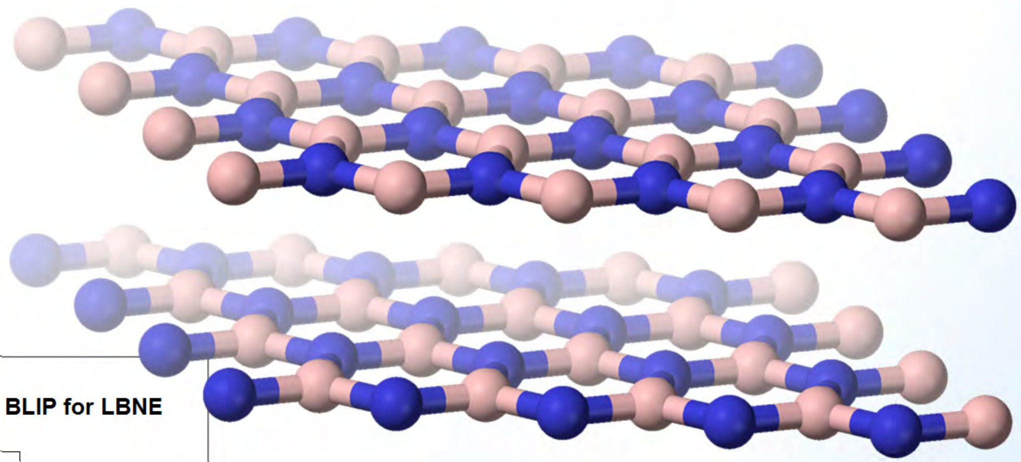
a first level evaluation based on the changes of ultrasonic velocity is a good indicator of its strengthening due to irradiation and its softening due to annealing to $T > T_{irr}$

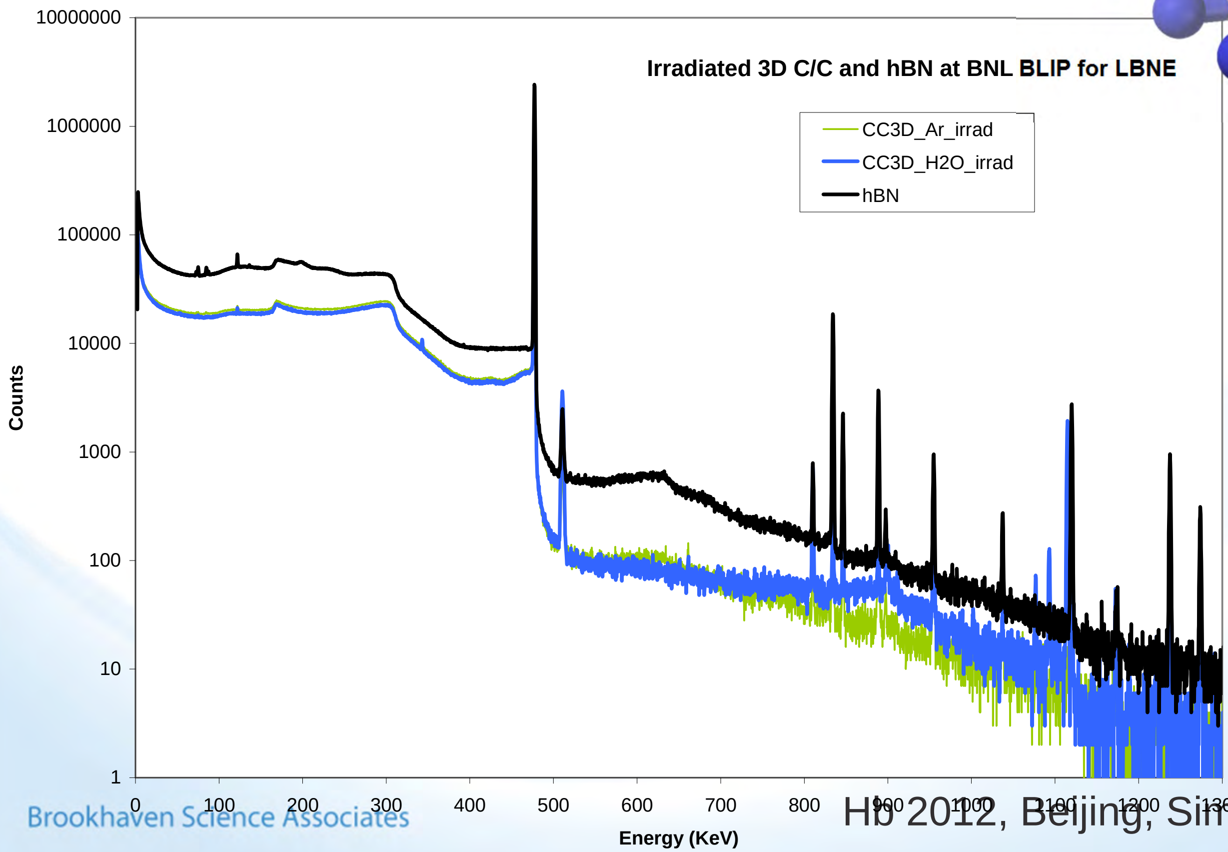
Simos

BROOKHAVEN NATIONAL LABORATORY

h-BN Irradiation

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.

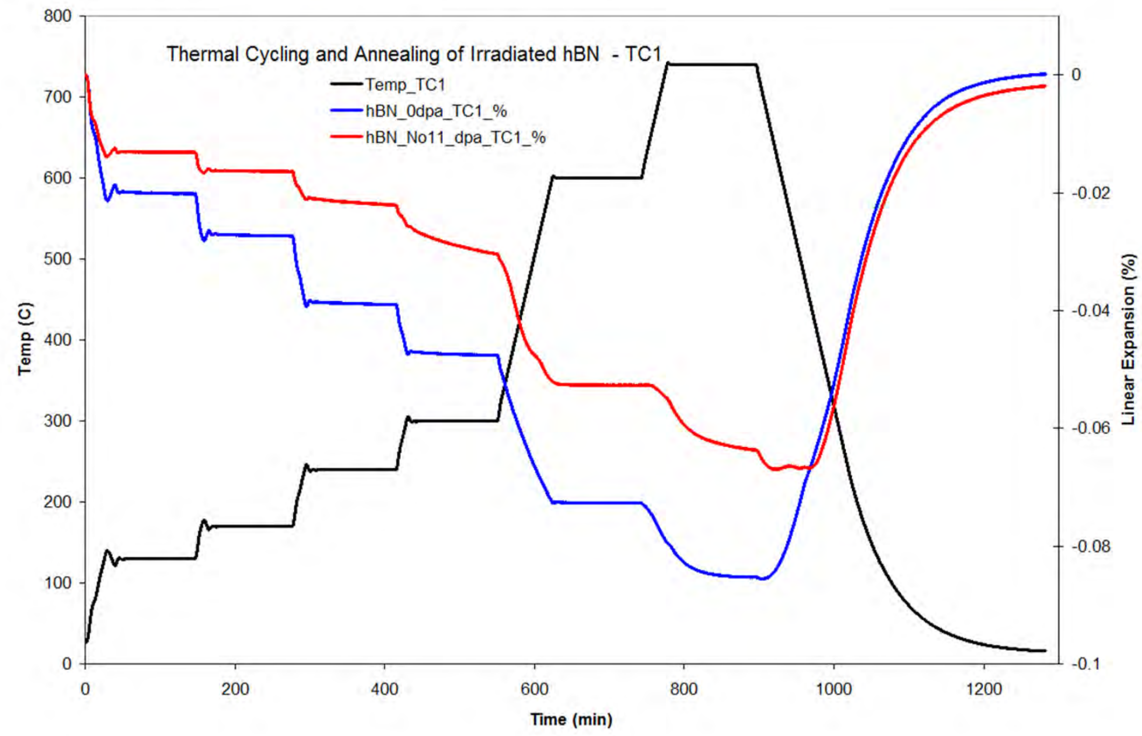




Things to note:

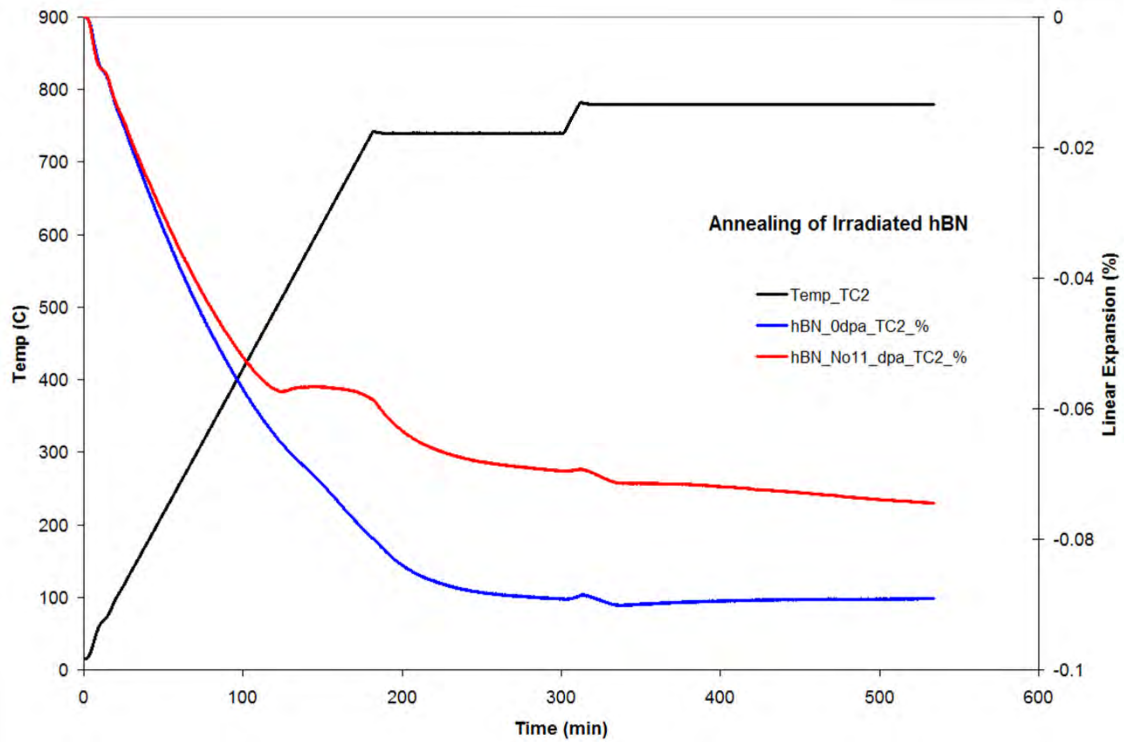
h-BN has a negative thermal expansion for the temperature range studied (<800 ^oC) and it appears to continue on the negative side at even higher temperatures.

Irradiation induces changes in the expansion behavior and in similar way to the behavior in the direction parallel to the fiber planes in C/C and not similar to graphite that is a close neighbor!! This indicates that h-BN is highly oriented into parallel planes and not just its lattice.



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

The new study confirmed that energetic proton fluence thresholds are a reality for carbon-based lattices

Graphite continues to be very intriguing in spite of its simple lattice structure

There is significant variability between graphite grades in the way that graphite responds to irradiation

Non-destructive testing has shown great potential in assessing damage annealing

Fast neutron exposure to fluences that approach some of the proton irradiation tests has just been completed and will provide a good correlation between the different energetic irradiating species