BNL Irradiation and Characterization Studies

Summary Report on HP Accelerator Material Research

Reporting on (ONLY):

- **Irradiation and micro- macro-characterization of Beryllium**
- **Irradiation Damage and Assessment of Graphite**
- **Irradiation and Characterization of Ti-alloys (Ti6Al4V and Gum Metal)**
- **Irradiation and behavior of Tungsten (plus Ta, Mo)**

N. Simos

and new member of team: M. E

May 19, 2015

Materials linked with:

Neutrino Factory, LHC, LBNE, Next Gen Fusion/fission Reactors

Materials:

Steels, Inconel, S-Invar, Gum Metal, Ti-6Al-4V, Cu, Glidcop, W, Ta
Graphite (s), C/C composites, SiC/SiC
Mo, Mo-GR, Cu-CD
Interfaces (Cu-Ti-Graphite, Al_2O_3-Ti6Al4V

Facilities Utilized/integrated:

200 MeV BNL Linac/BLIP, Tandem, Isotope Extraction Facility
National Synchrotron Light Source (NSLS) and NSLS II
Center of Functional Nanomaterials

BNL Studies on Beryllium

BNL Be Studies OVERVIEW

Beryllium was of interest in Neutrino Factory as a low-Z target material and also Fusion and Next Generation Reactors

Special samples of Be (along with **AlBeMe**t) were originally irradiated with 200 MeV protons, and followed with fast neutron irradiation (no post analysis yet)

Macroscopic post-irradiation analyses confirmed the overall properties including dimensional stability, ductility and strength and thermal conductivity

Recent versitile EDXRD and XRD experiments on irradiated Be using the NSLS Synchrotron focused on

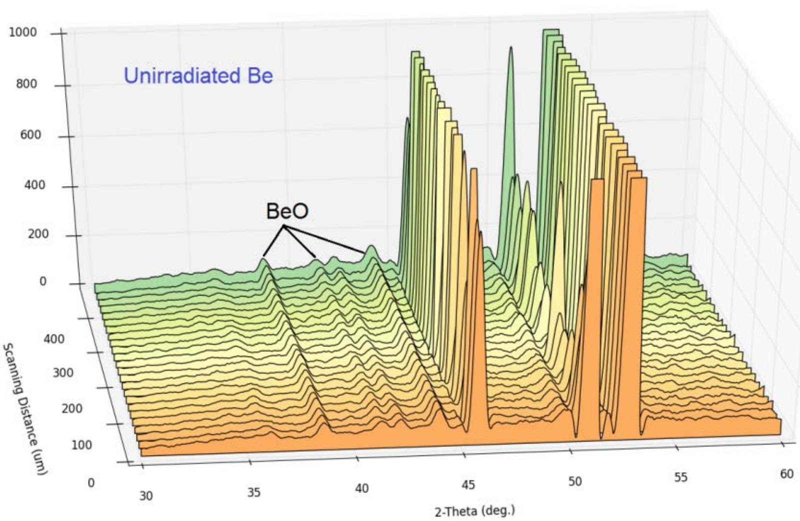
- The effects of irradiation damage on the Be microstructure
- The Be anisotropy in compression and tension
- The controlling deformation mechanisms (such as twinning in the Be h.c.p. lattice) which will explain its plasticity behavior and also its fracture characteristics

As in all hexagonal close-packed (HCP) structural materials Be also exhibits limited activation of different slip mechanisms results in alternative deformation mechanisms, such as twinning and c+a mode deformation mode, which become relevant to plasticity.

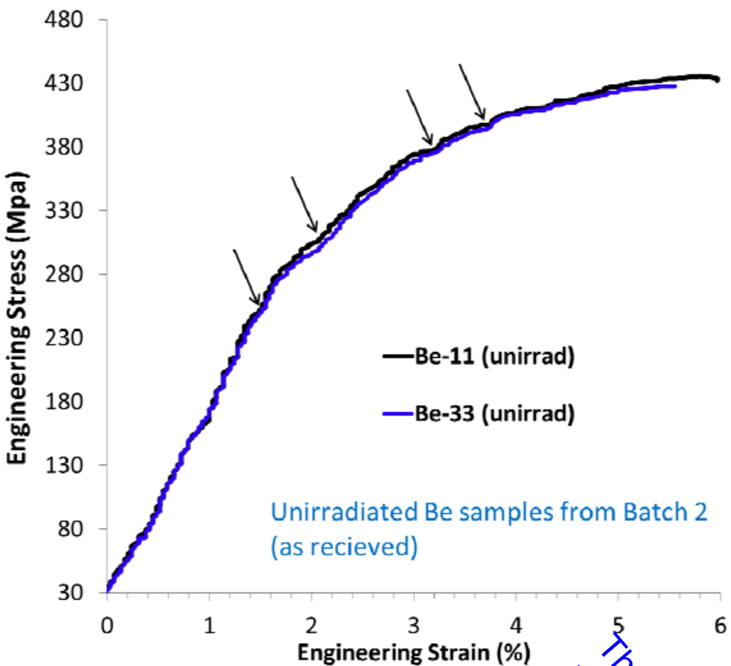
Table shows the great variability of Beryllium
BNL received 2 bunches from Brush-Wellman for its studies:

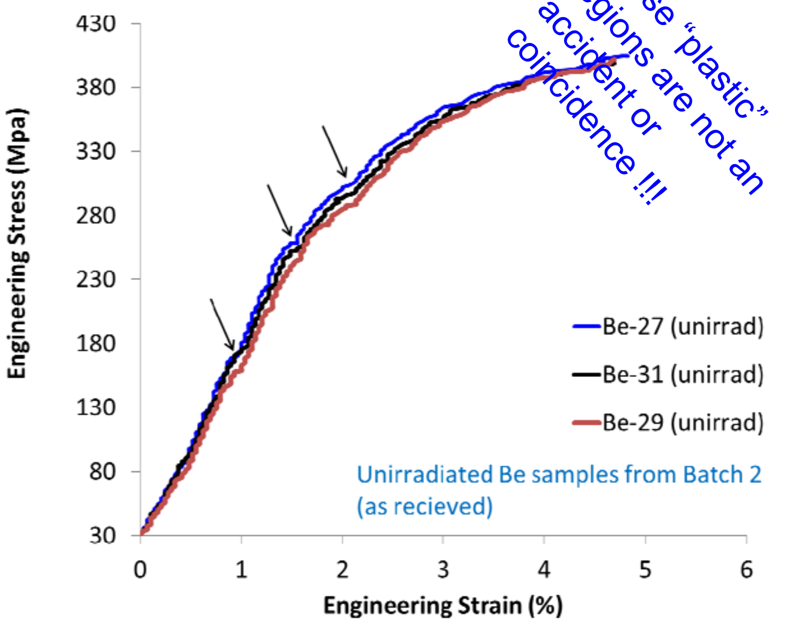
Manufacturer's properties: Ultimate Tensile = 492 MPa, Yield Strength =329 MPa and Tensile Elongation = 7.3%

Material	Grain size, μm	Ultimate tensile strength (UTS) (a), MN/m²	Yield strength (b), MN/m²	Tensile elongation, %
Rolled electrolytic flake	80	112	131 (c)	~4
Complexed, worked, high-purity ingot	13	?	596 (d)	7.5 (e)
Upset-forged high-purity ingot	70	120	45	1.5
Diffusion-bonded rolled ingot laminates	16	221	284	18
Commercial purity (Brush QMV), extruded	6	492	329	7.3
High purity (Pechiney CR), extruded	20	166	382	9.7
Hot pressed, forged and aged, high purity	~20 (powder)	373	219	18
High purity, hot isostatically pressed (HIP),	1.6	676	484	12.7
	2.8	586	305	27.5
...1.	?	284	315	13.6
	10	321	108	4.8
	7.0	284	215	6.7
	?	238	214	7.1
	3.8	420	205	5.9
	7.9	240	250 (c)	5.7

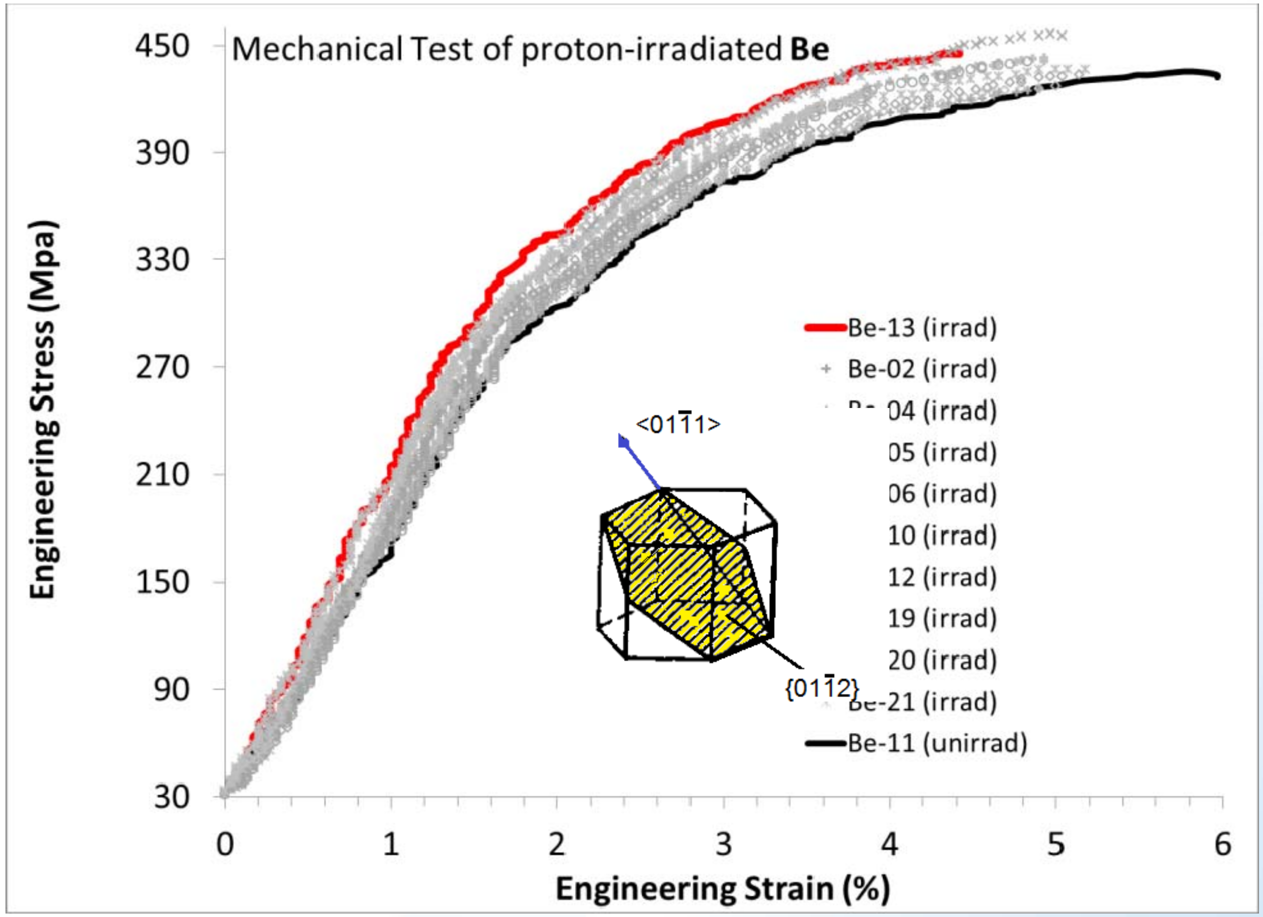


Mechanical Tests confirmed manufacturer's data, revealed ductile behavior of un-irradiated and irradiated Be as well as the intriguing behavior of twinning under tension!!!





These "plastic" regions are not an accident or coincidence !!!



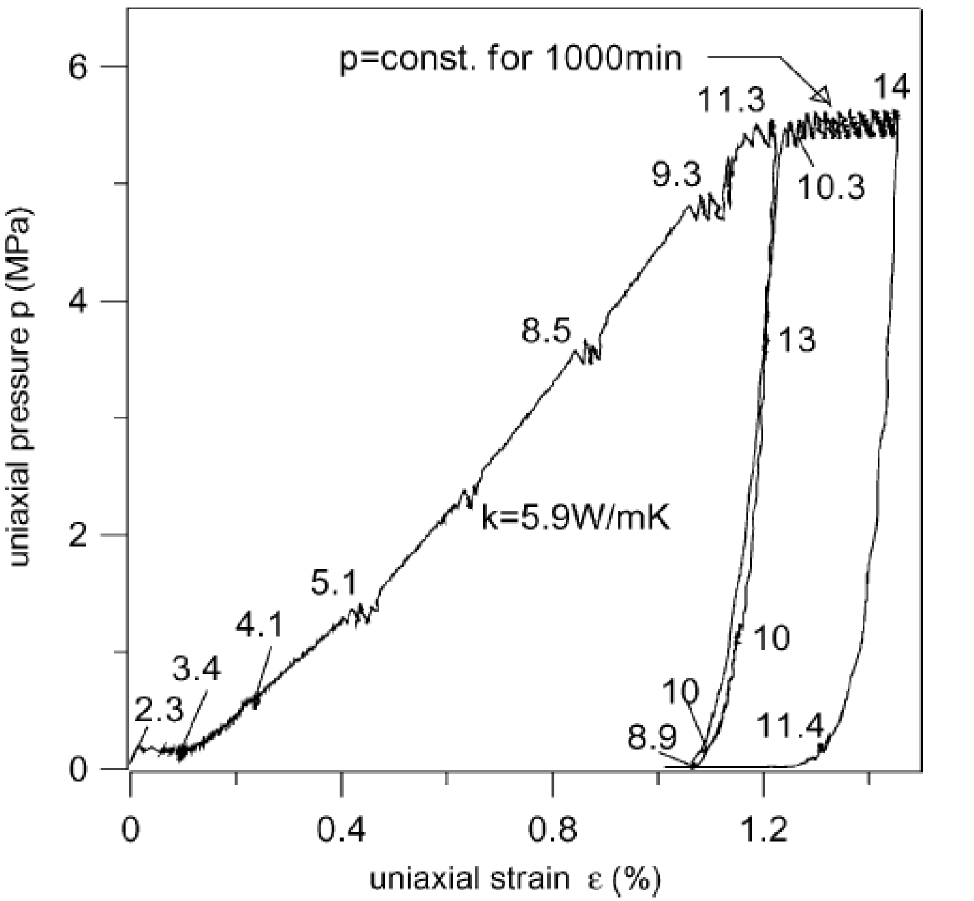
For c/a < √3 Compression NORMAL and tension parallel to c-axis will trigger twinning.

What we have here is c/a = 1.5682< √3 in tension (with some compression in the polycrystalline normal to tensile load). So twinning under tension primarily should appear!!

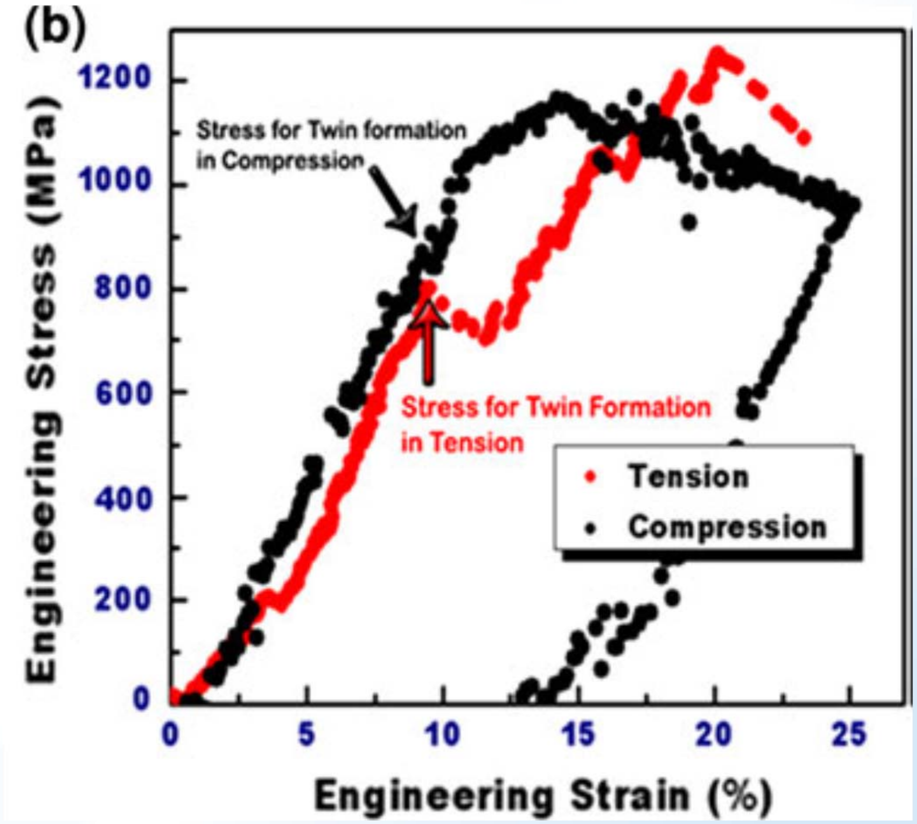
Kinks in the elastic regime of Be or of other h.c.p. structures have been seen by other researchers.

On left a increase in conductivity is observed over these micro-plastic deformation zones under compression.

On right the "asymmetry" between compression and tension at nano-level



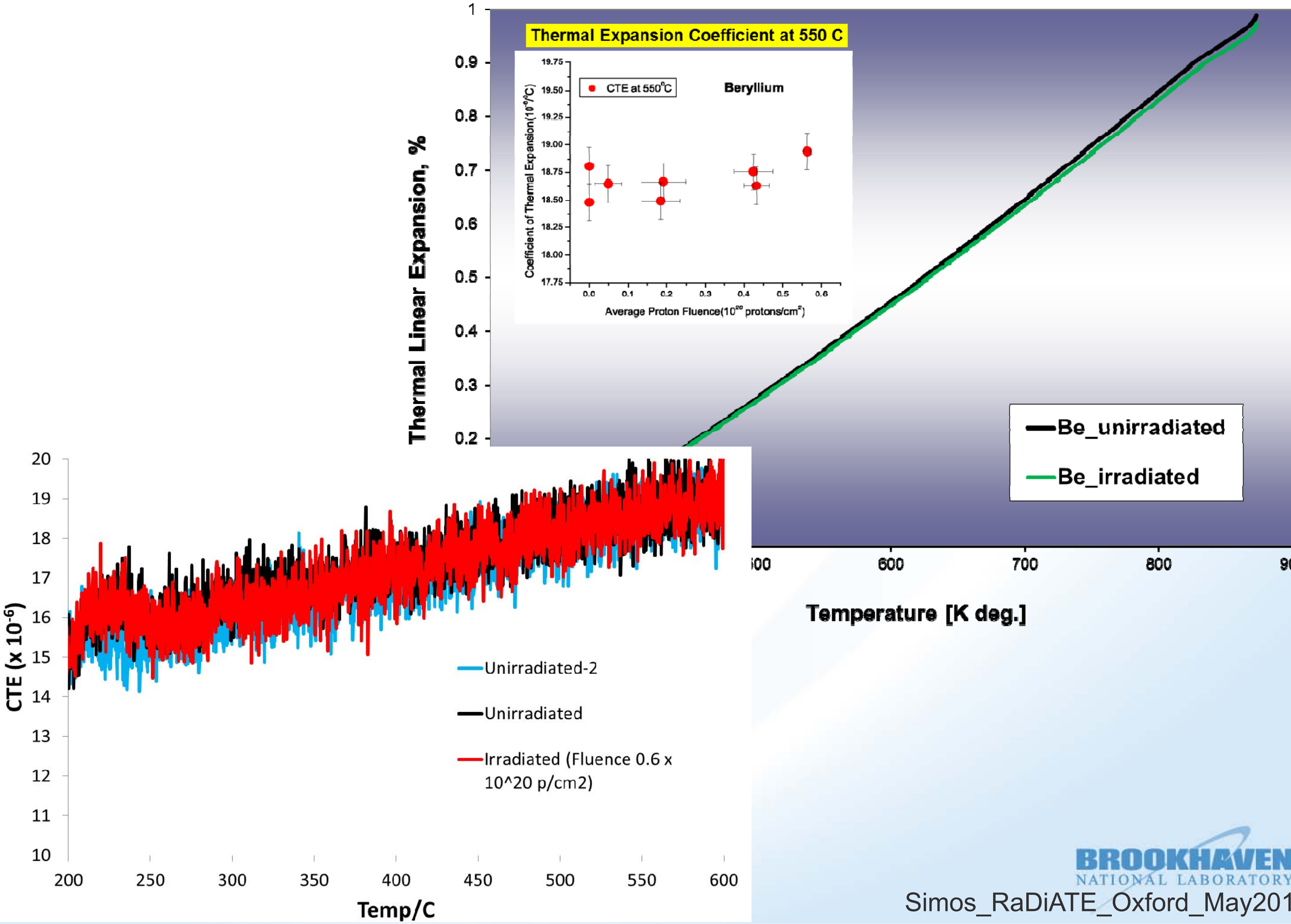
Beryllium for fusion application – recent results
Journal of Nuclear Materials 307–311 (2002) 630–637



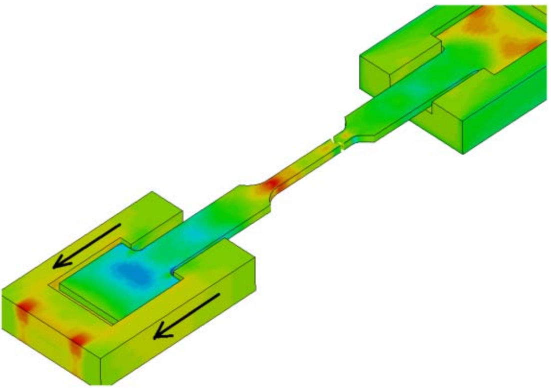
Comparison of the stress–strain curves from a nano-compression and a nano-tensile test in submicron Mg samples. Strain softening was observed in the nano-compression test, while strain hardening was observed in the nano-tensile test

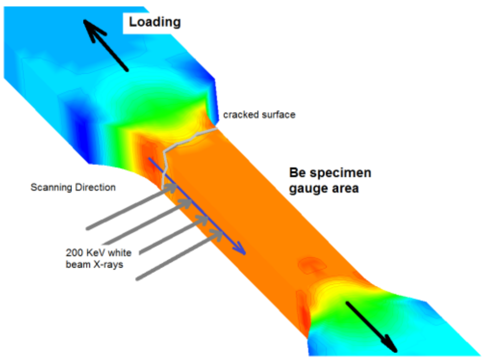
The Effect of Size on the Deformation Twinning Behavior in Hexagonal Close-Packed Ti and Mg, QIAN YU, et al

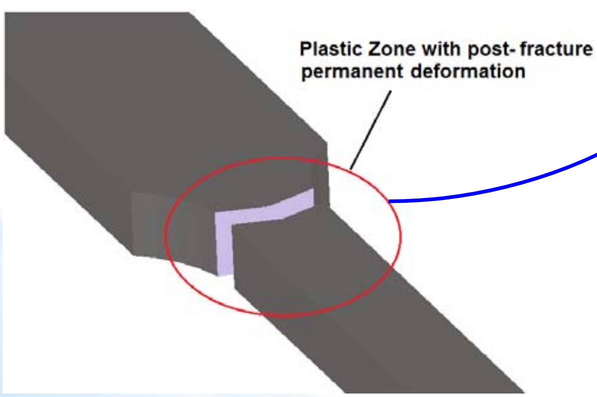
Thermal Stability Analysis of Irradiated Be

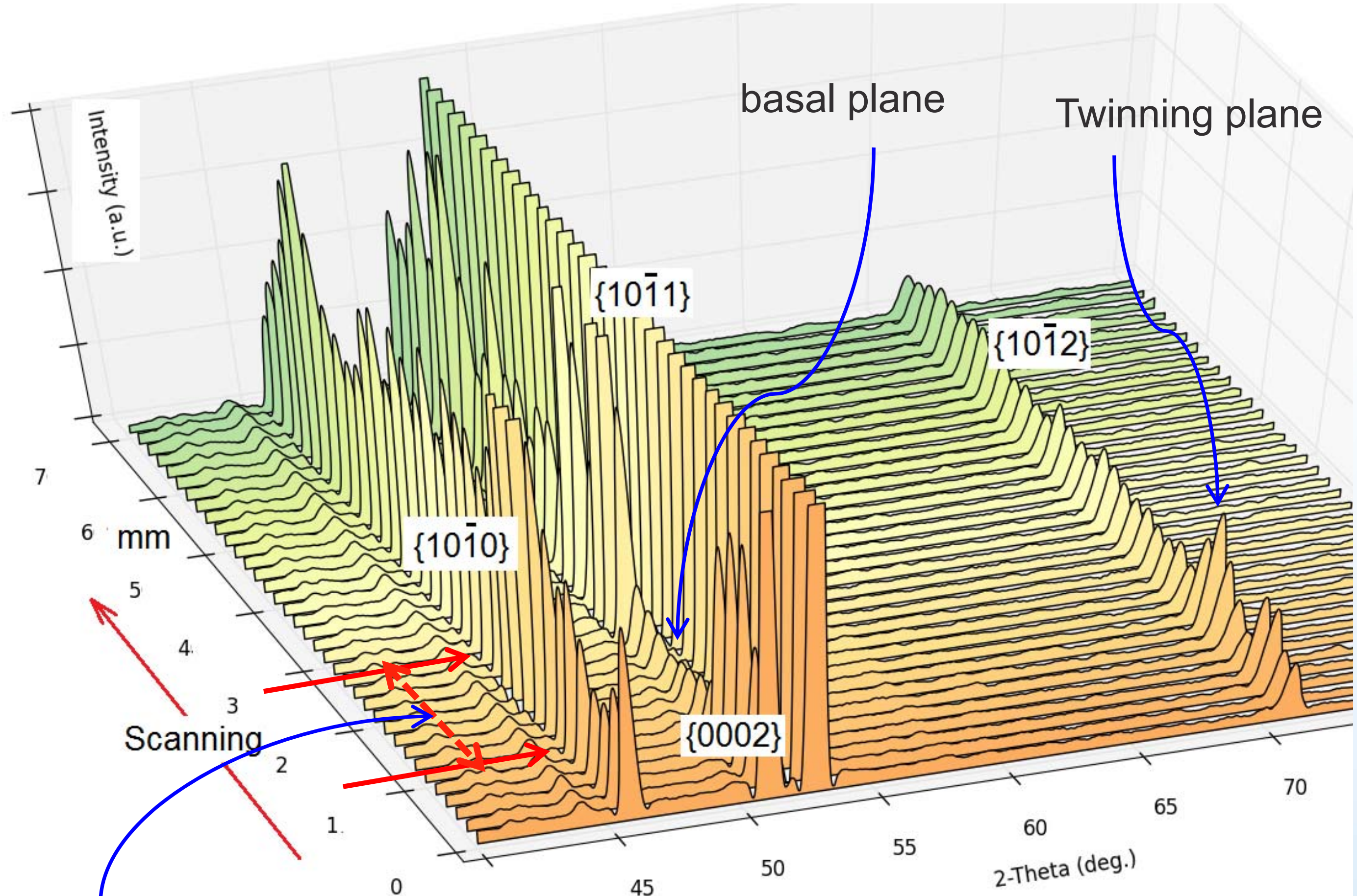


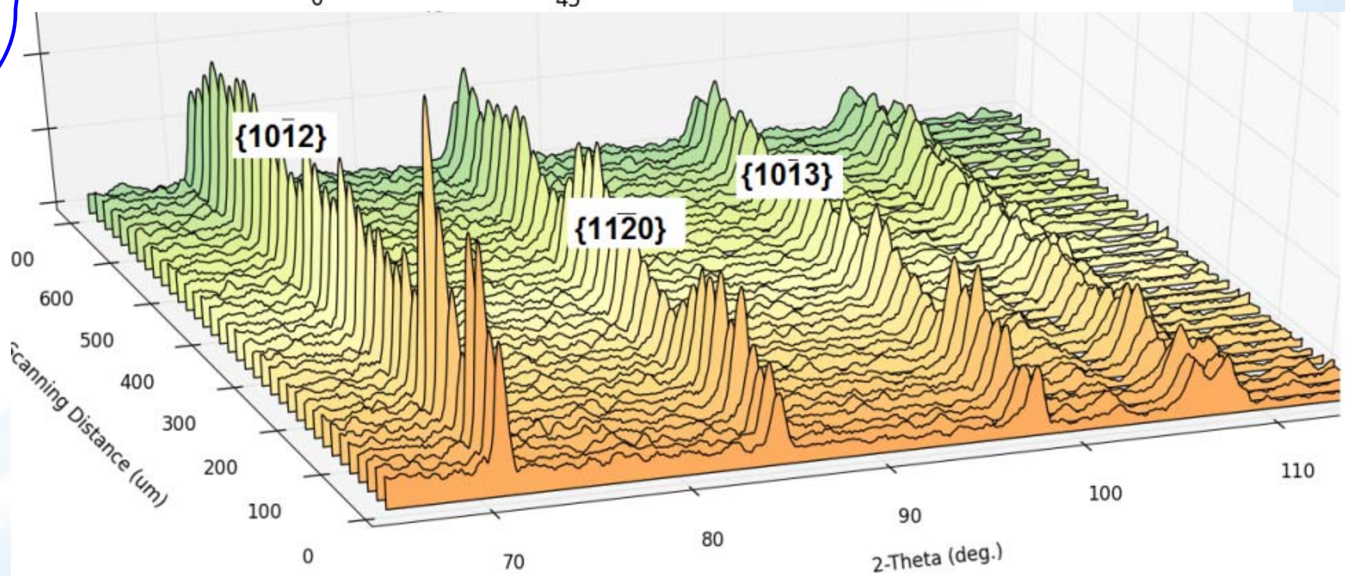
Irradiated Be X-ray Diffraction Experiments





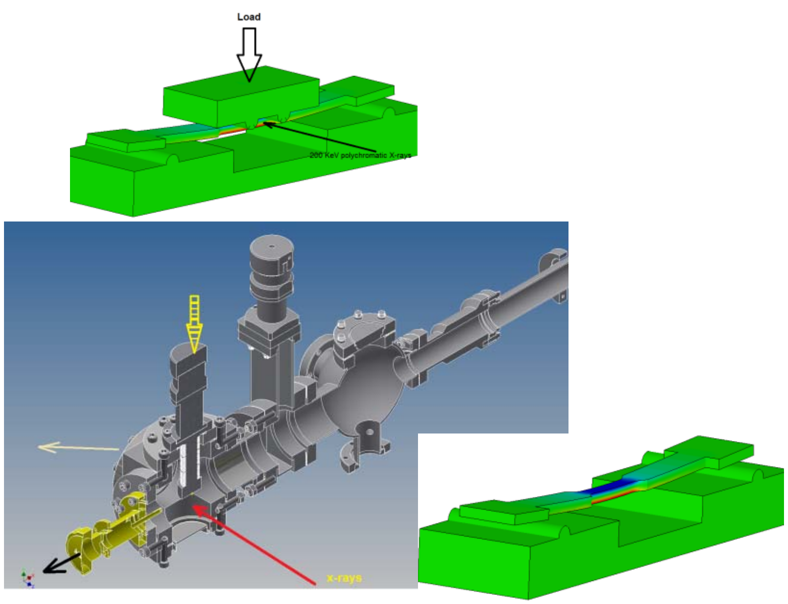


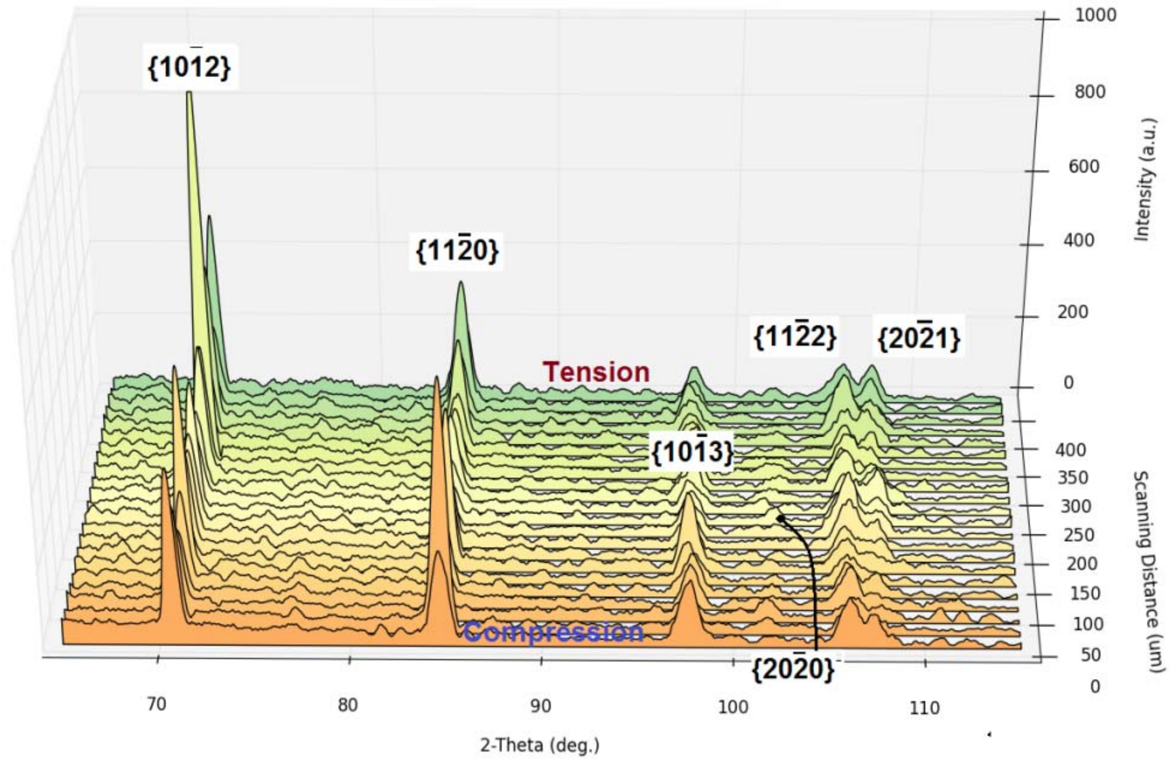


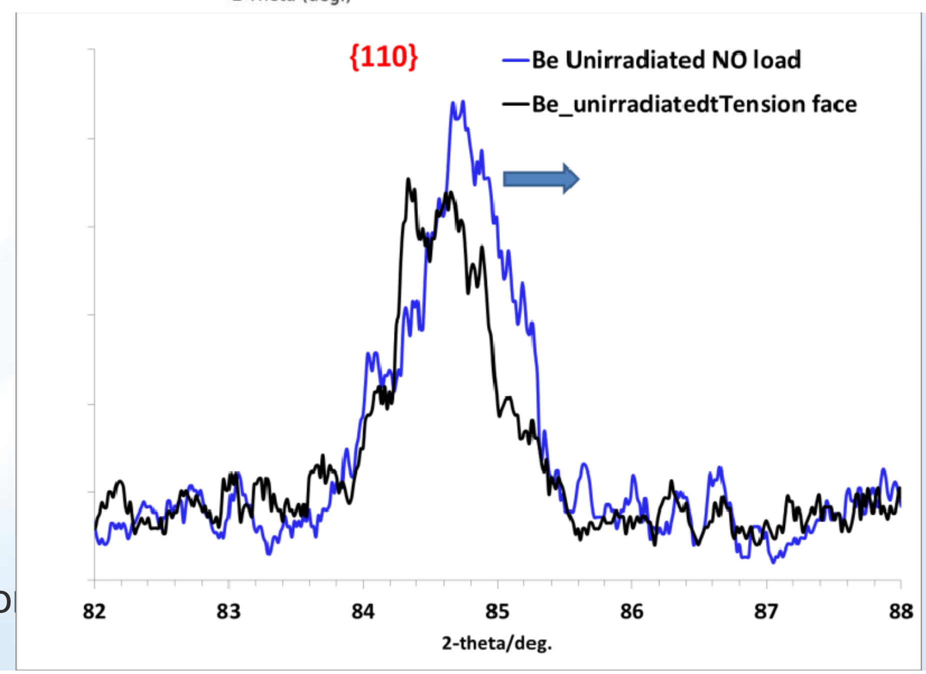


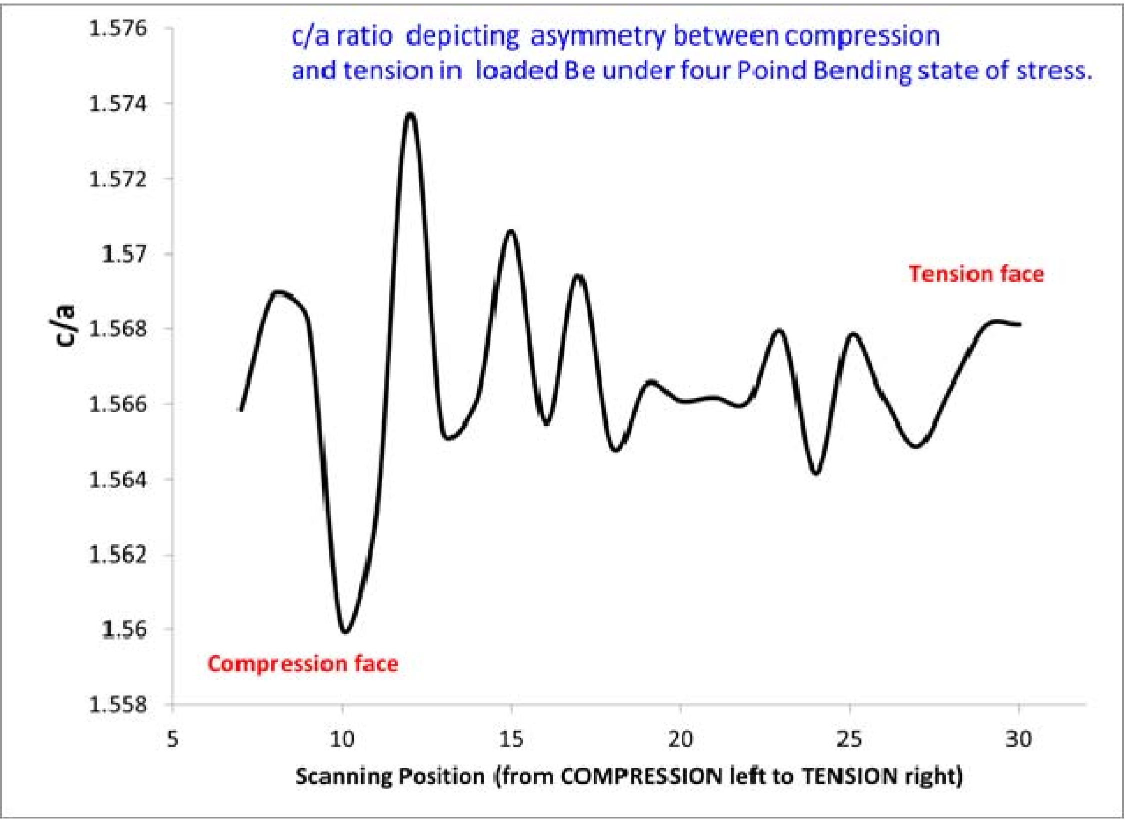
Studying Tension-Compression Asymmetry in Be

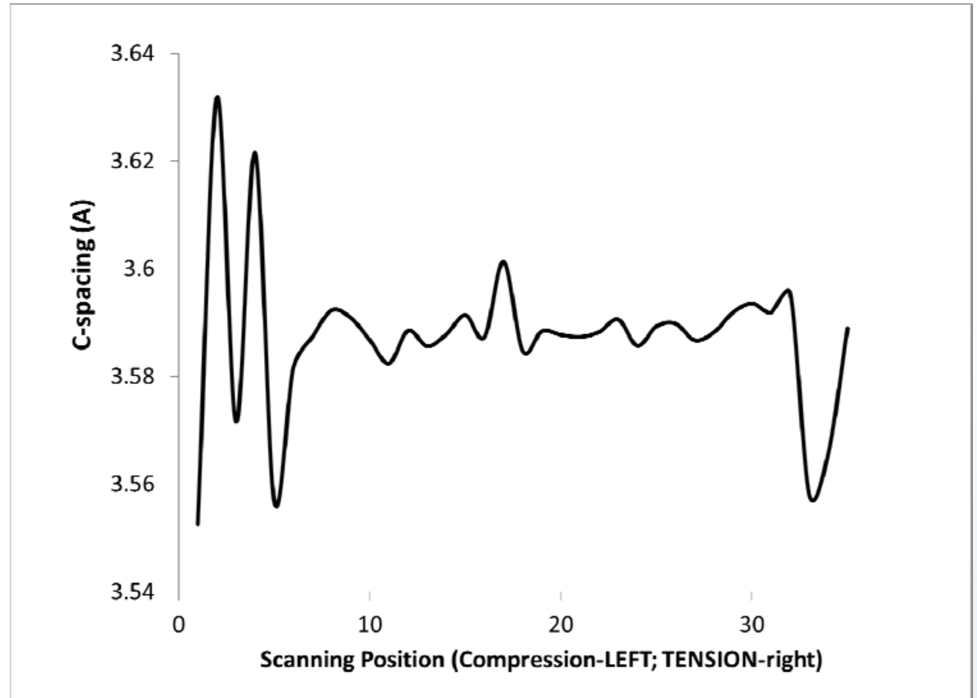
EDXRD study under Four-Point Bending











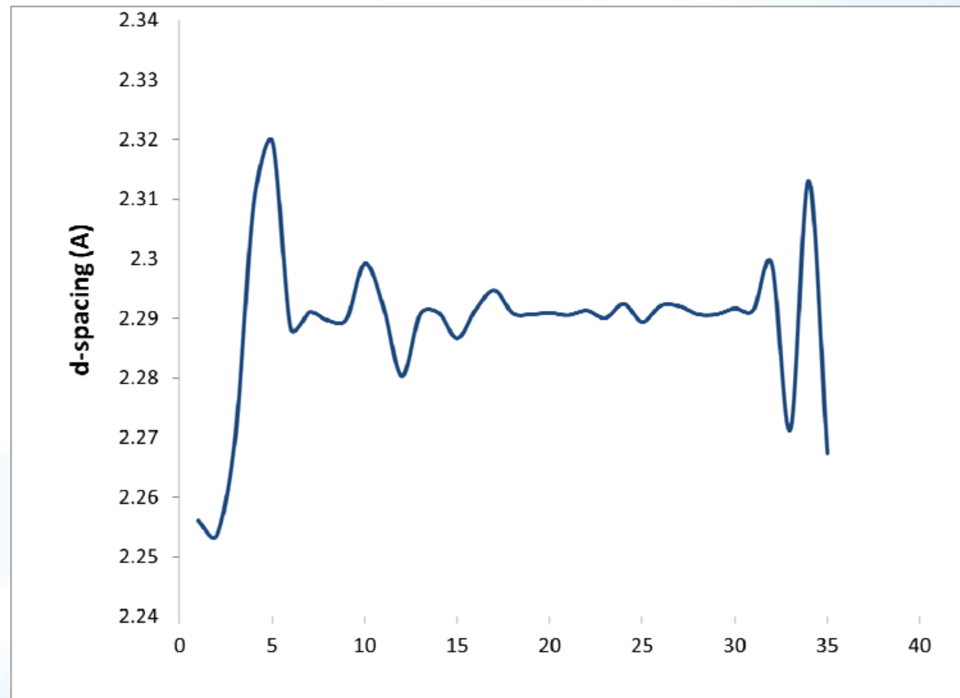
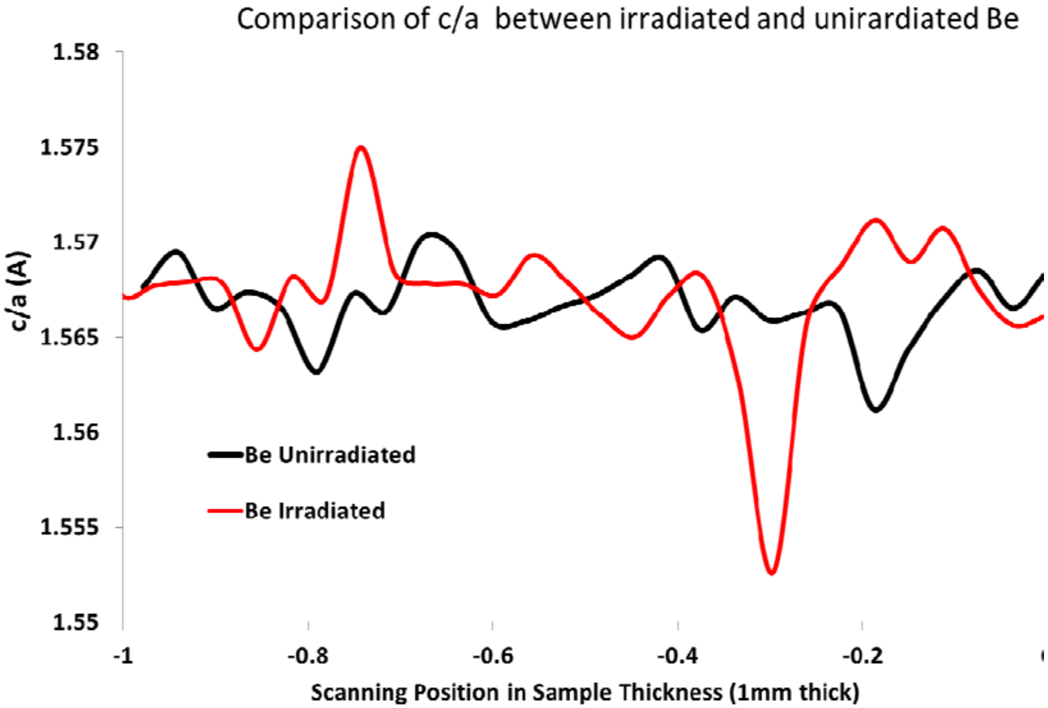


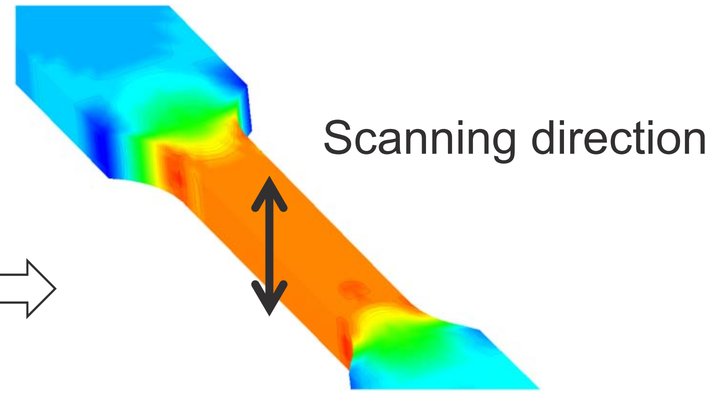
Table 4.1 Atomic/crystal structure/ characteristic of beryllium

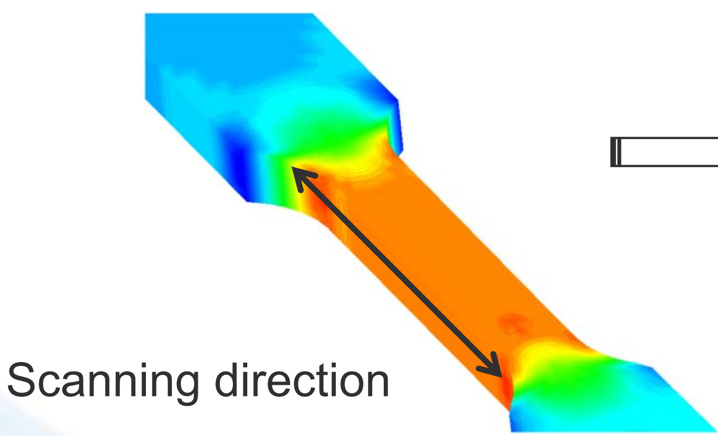
Atomic number	4
Atomic radius, Å	1.125
Atomic volume, cm³/g · atom	4.96
Atomic weight, g	9.01218
Density—pure, g/cm³	1.8477
Density—commercial grades, g/cm³	1.82–1.85
Electronic structure	$1s^2 2s^2$
Crystal structure, <1250 °C	Hexagonal close-packed (α-Be)
Crystal structure, >1250 °C	Body-centered cubic (β-Be)
Lattice constants, Å	a = 2.286; c = 3.583; c/a = 1.568; ideal = 1.633

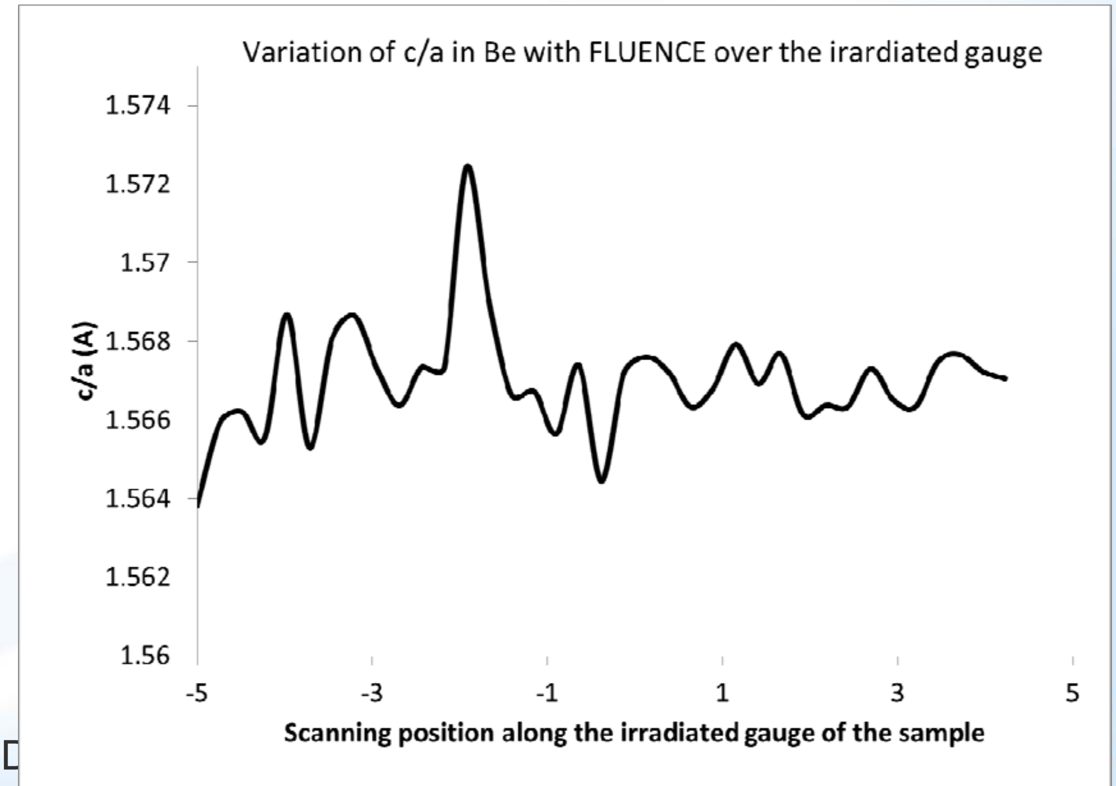
Table 4.2 Variation of lattice parameters with temperature

Temperature, °C	a-axis	c-axis	c/a
50	2.287	3.585	1.5679
100	2.288	3.587	1.5676
200	2.292	3.591	1.5670
300	2.296	3.596	1.5664
400	2.300	3.601	1.5658
500	2.304	3.607	1.5652
600	2.309	3.613	1.5646
800	2.319	3.625	1.5633
1000	2.330	3.639	1.5620


Comparison of c/a between irradiated and unirardiated Be
1.58
1.575
1.57
1.565
1.56
1.555
1.55
c/a (A)
Be Unirradiated
Be Irradiated
-1
-0.8
-0.6
-0.4
-0.2
Scanning Position in Sample Thickness (1mm thick)


Scanning direction


Scanning direction


Variation of c/a in Be with FLUENCE over the irardiated gauge
1.574
1.572
1.57
1.568
1.566
1.564
1.562
1.56
c/a (A)
-5
-3
-1
1
3
5
Scanning position along the irradiated gauge of the sample

Irradiation Damage to Graphite

Graphite & Carbon-based Material Irradiation/Characterization

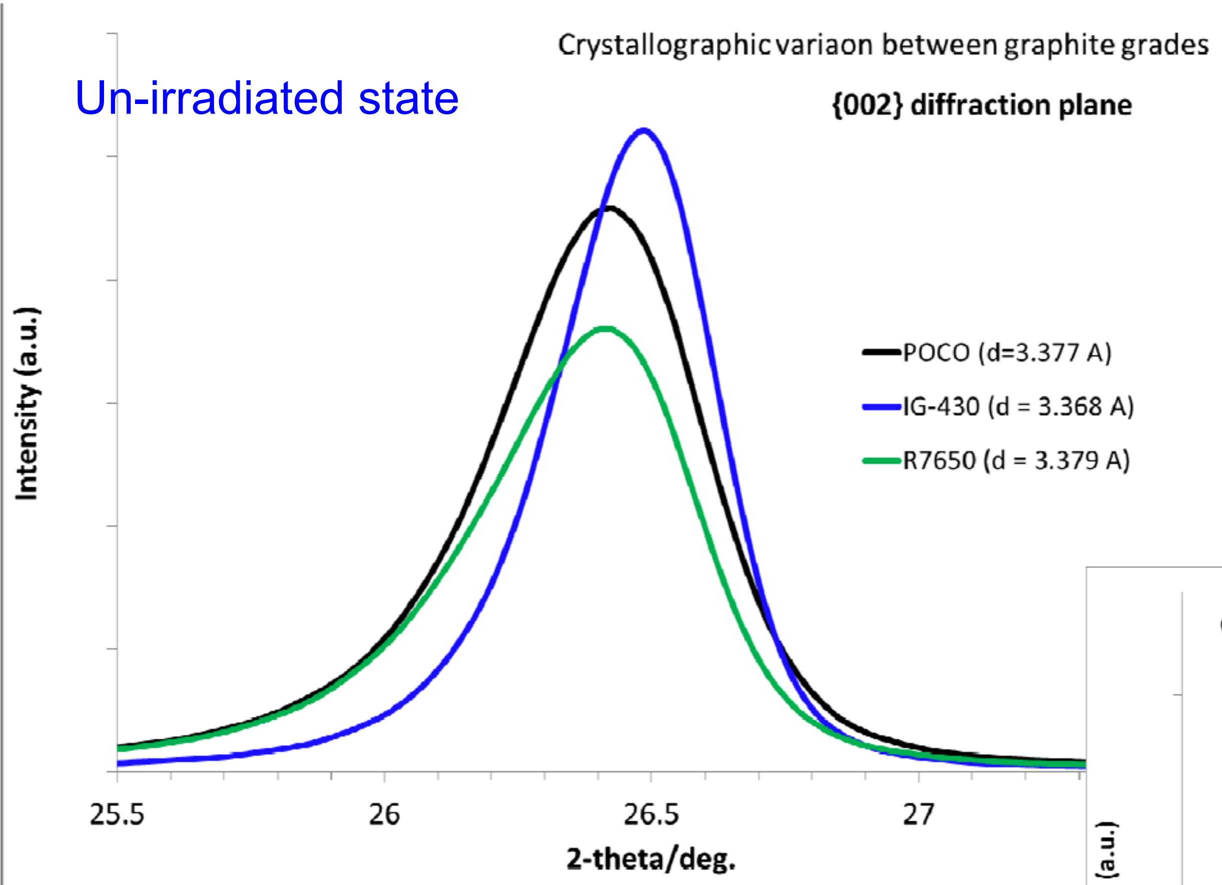
- An array of irradiation damage and post-irradiation characterization studies have been under way at BNL for graphite and carbon-based structures
- Brookhaven has a long history in the study of nuclear graphite
- BNL accelerator complex facilities (200 MeV Linac/BLIP and Tandem accelerator) provide proton, spallation fast neutron and ion irradiation beams)
- Macroscopic post-irradiation characterization utilizes the Isotope Extraction Facility (hot cells, remote handling and testing)
- Microscopic post-irradiation is performed at the BNL Synchrotron facilities (NSLS using white and monochromatic x-ray beams and now NSLS II) aided by multi-faceted characterization at the Center of Functional Nanomaterials

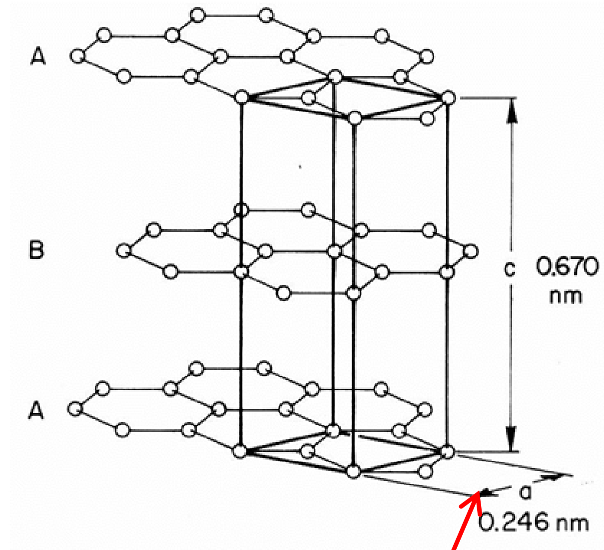
Graphite & Carbon-based Materials

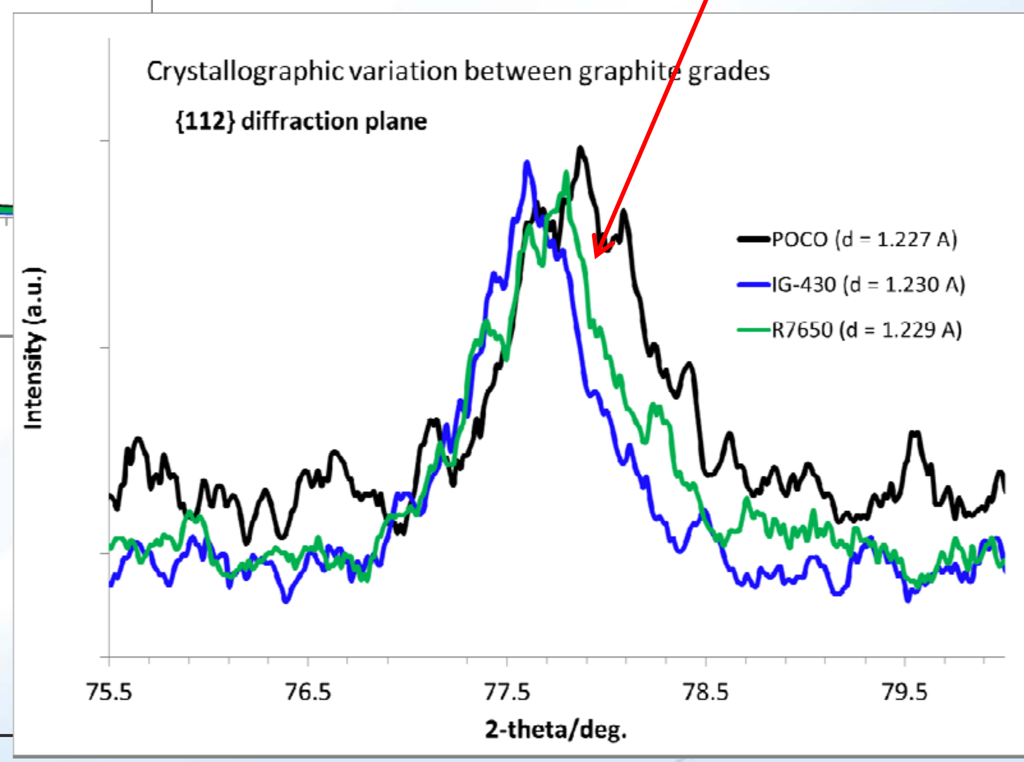
- Reactor-grade graphite (IG-43, IG-430) under fast neutrons and protons
- Carbon fiber composites (2D C/C and 3D C/C) + SiC/SiC
- HP Target bound graphite (LBNE) – 4 grades (POCO, IG-430, Carbone and R7650)
- Newly developed structures such as Mo-GR

Difference in crystal structure➔ macroscopic behavior?

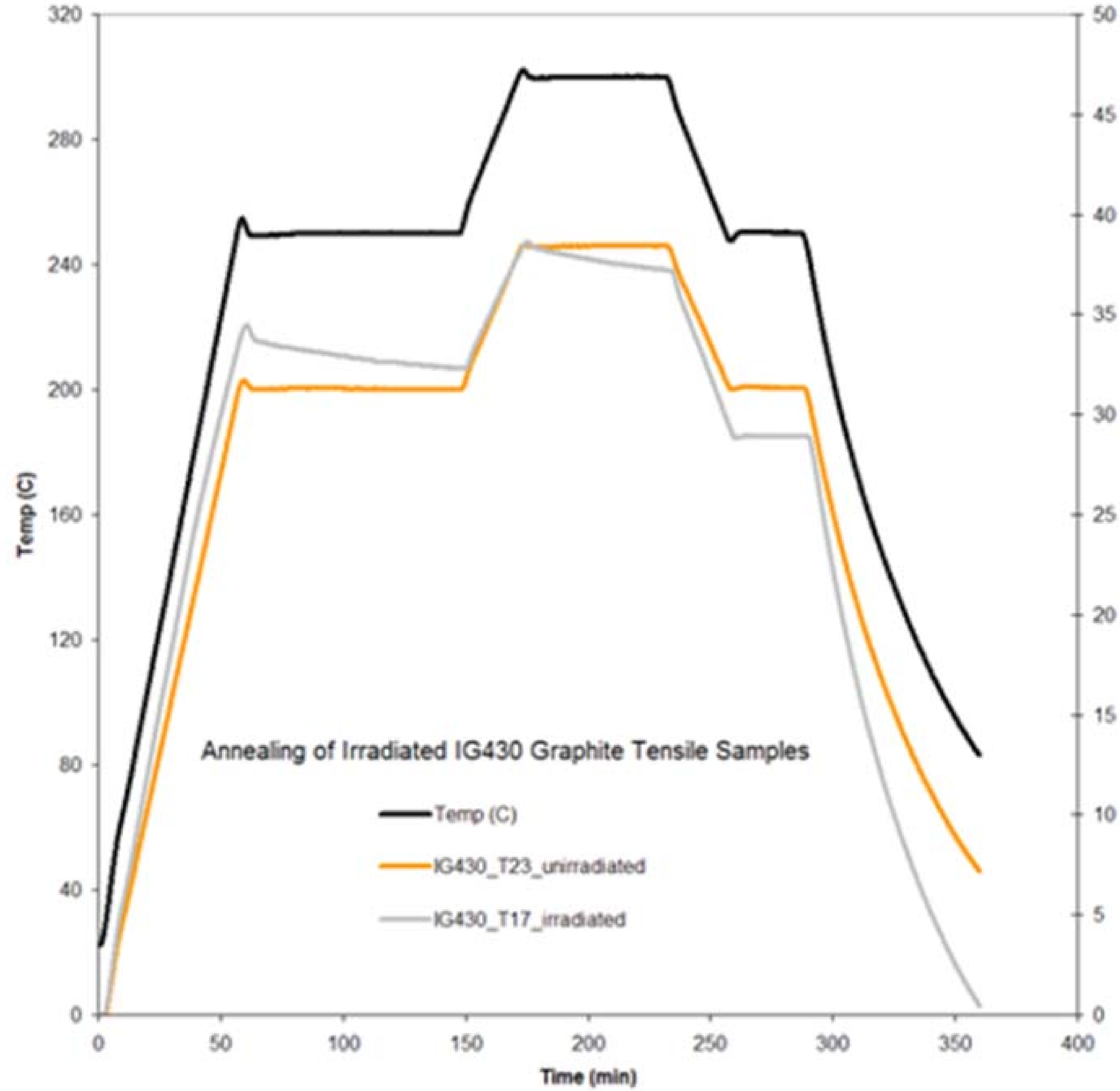
Un-irradiated state



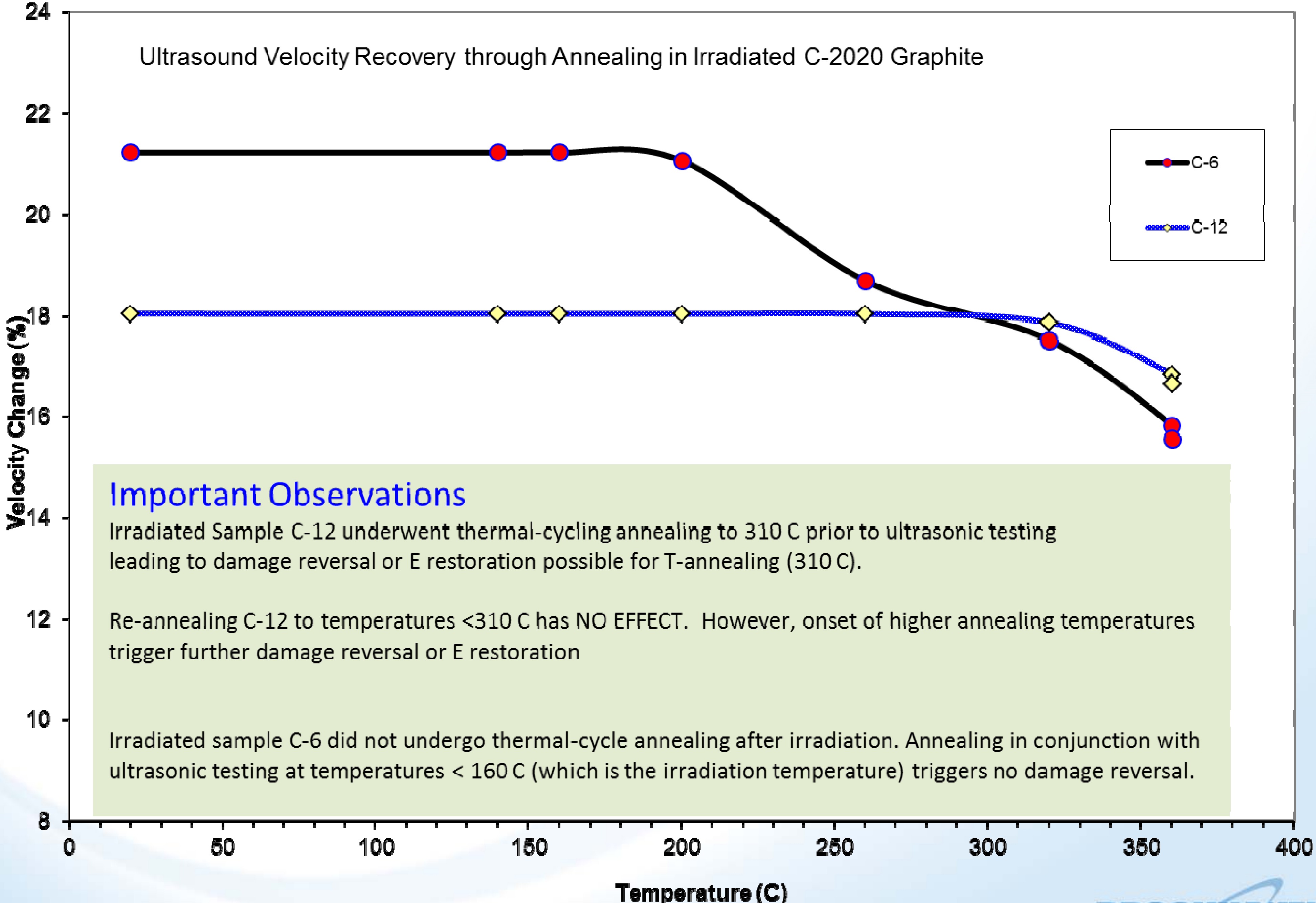




Annealing of Graphit



Annealing of Graphite - OBSERVATIONS

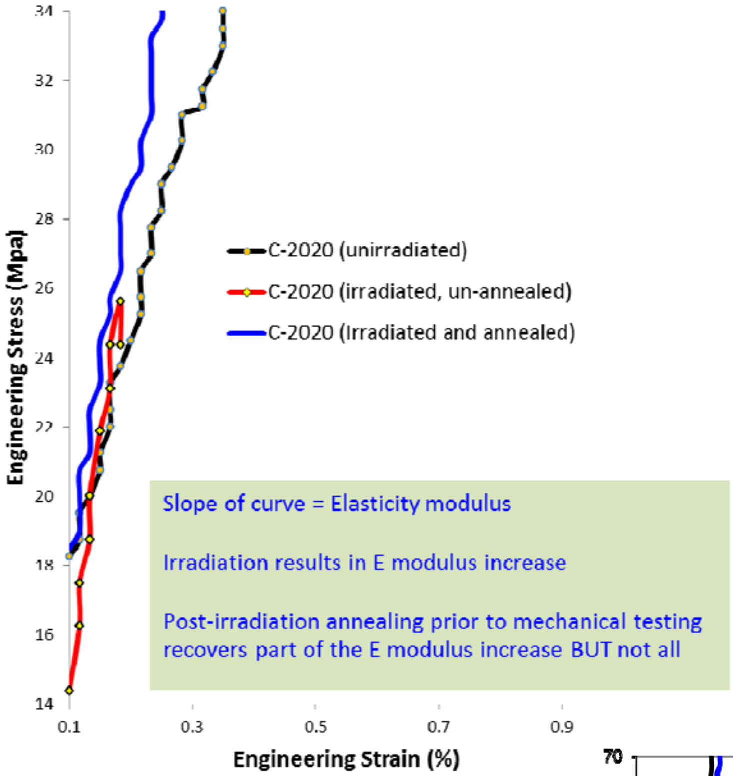


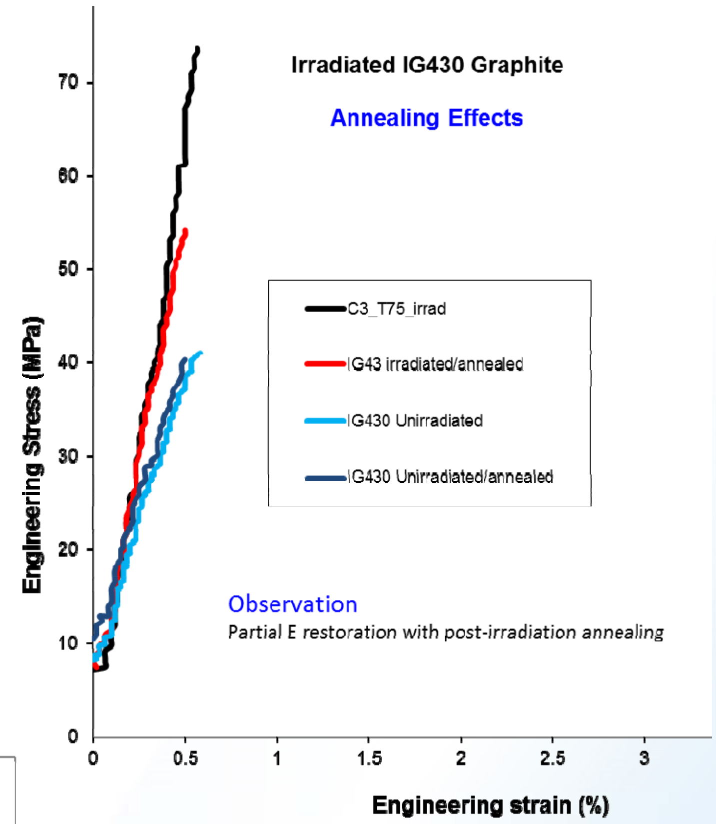
Important Observations

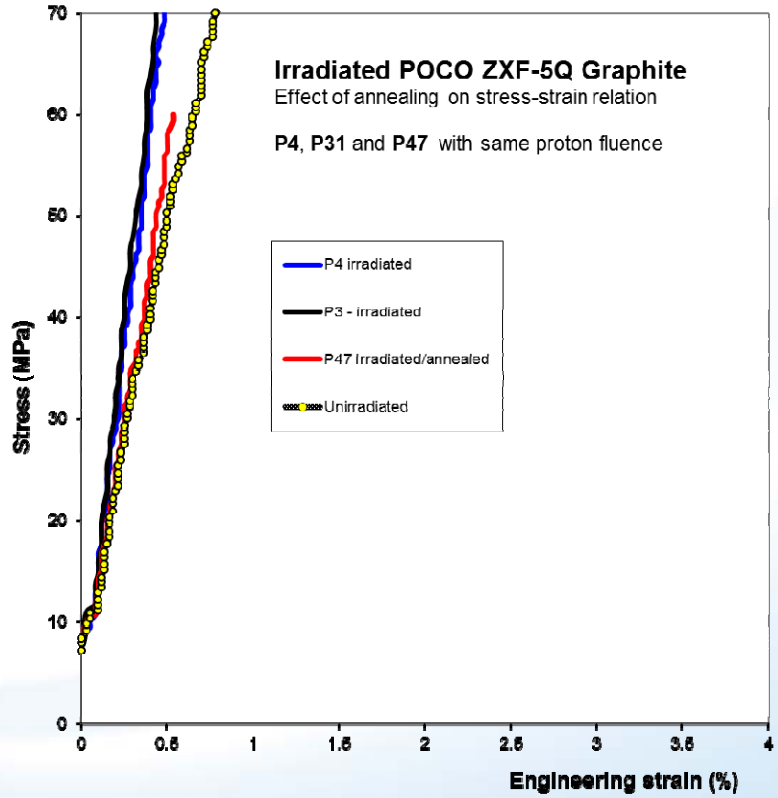
Irradiated Sample C-12 underwent thermal-cycling annealing to 310 C prior to ultrasonic testing leading to damage reversal or E restoration possible for T-annealing (310 C).

Re-annealing C-12 to temperatures <310 C has NO EFFECT. However, onset of higher annealing temperatures trigger further damage reversal or E restoration

Irradiated sample C-6 did not undergo thermal-cycle annealing after irradiation. Annealing in conjunction with ultrasonic testing at temperatures < 160 C (which is the irradiation temperature) triggers no damage reversal.

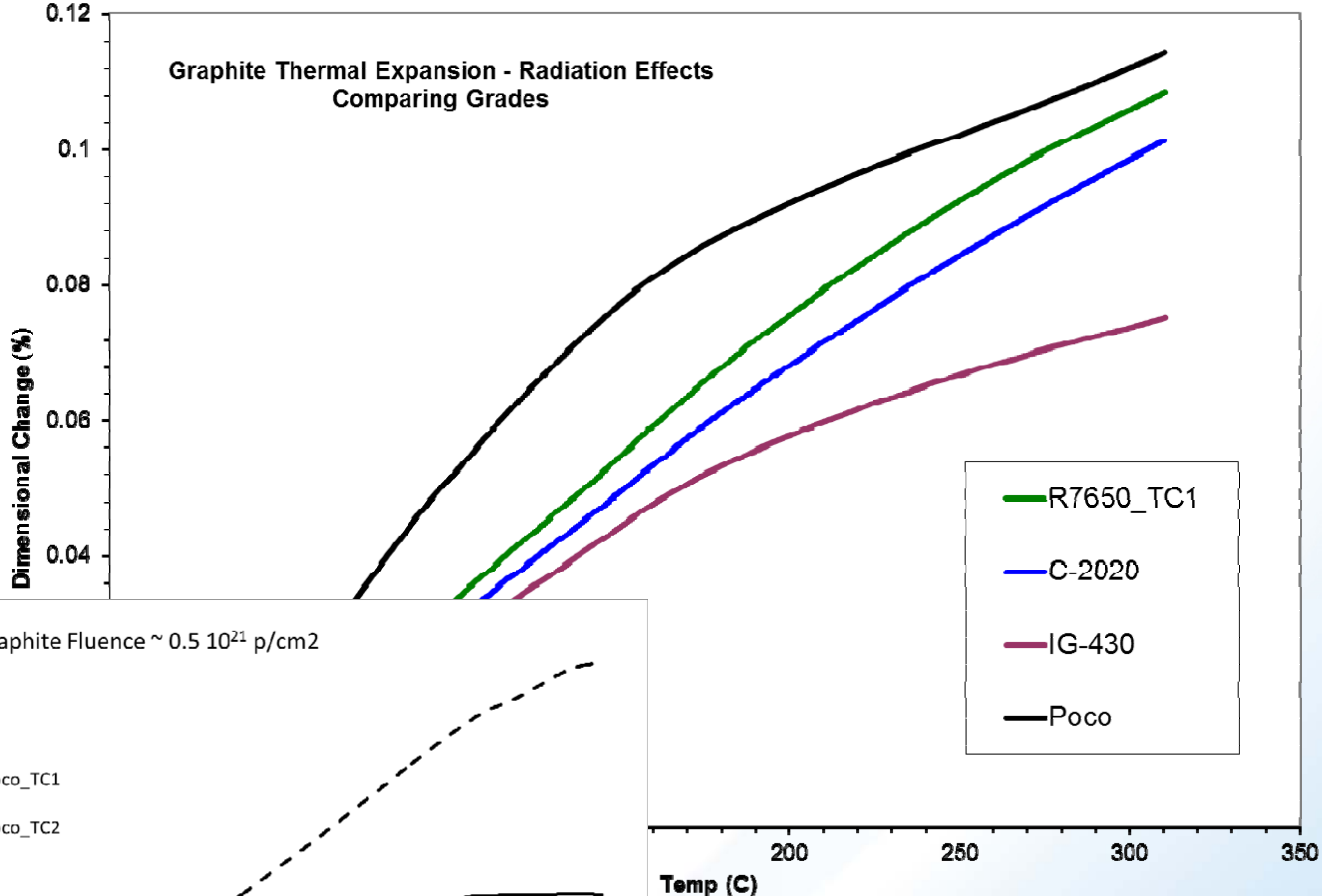


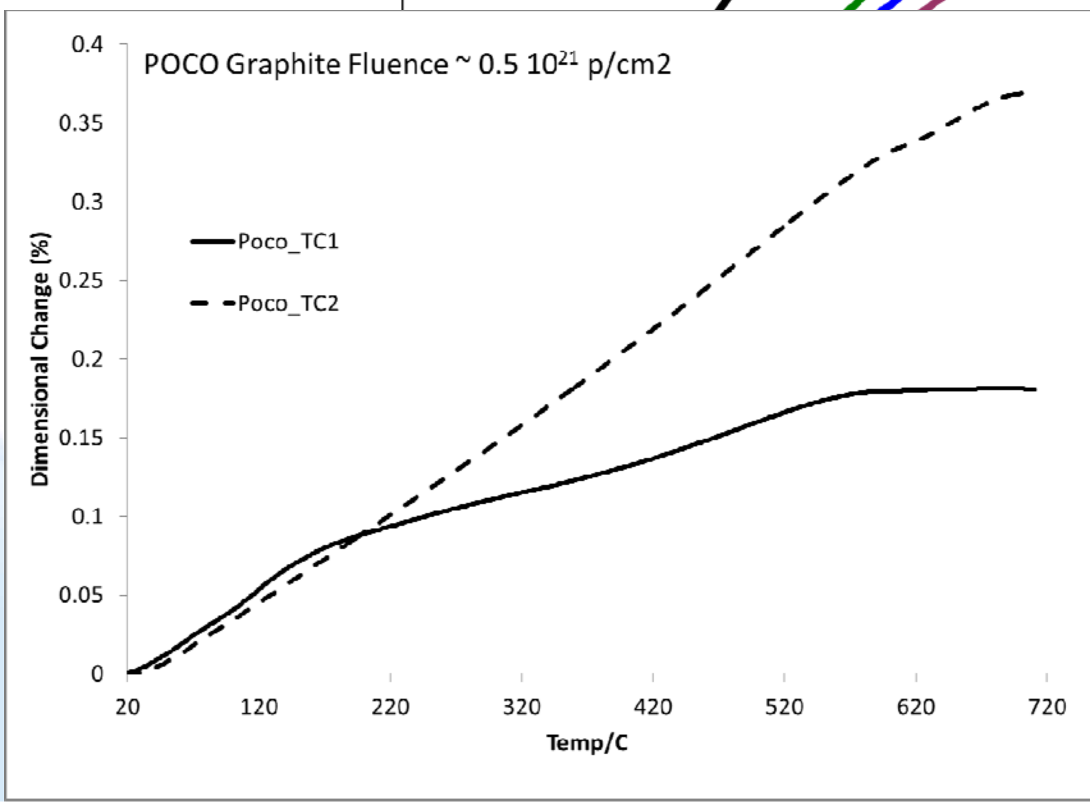




Observed in all grades is the increase in E due to dislocation pinning and "tightening up" of the aggregate structure due to irradiation growth.

Change in elastic modulus cannot be annealed out. Observations are in agreement with reactor neutron-induced deformation of graphite.





180 MeV proton-induced dimensional stability effects on LBNE graphite. Fig. 1depicts dimensional change variations between grades during the 1st post-irradiation annealing cycle which reverses damage

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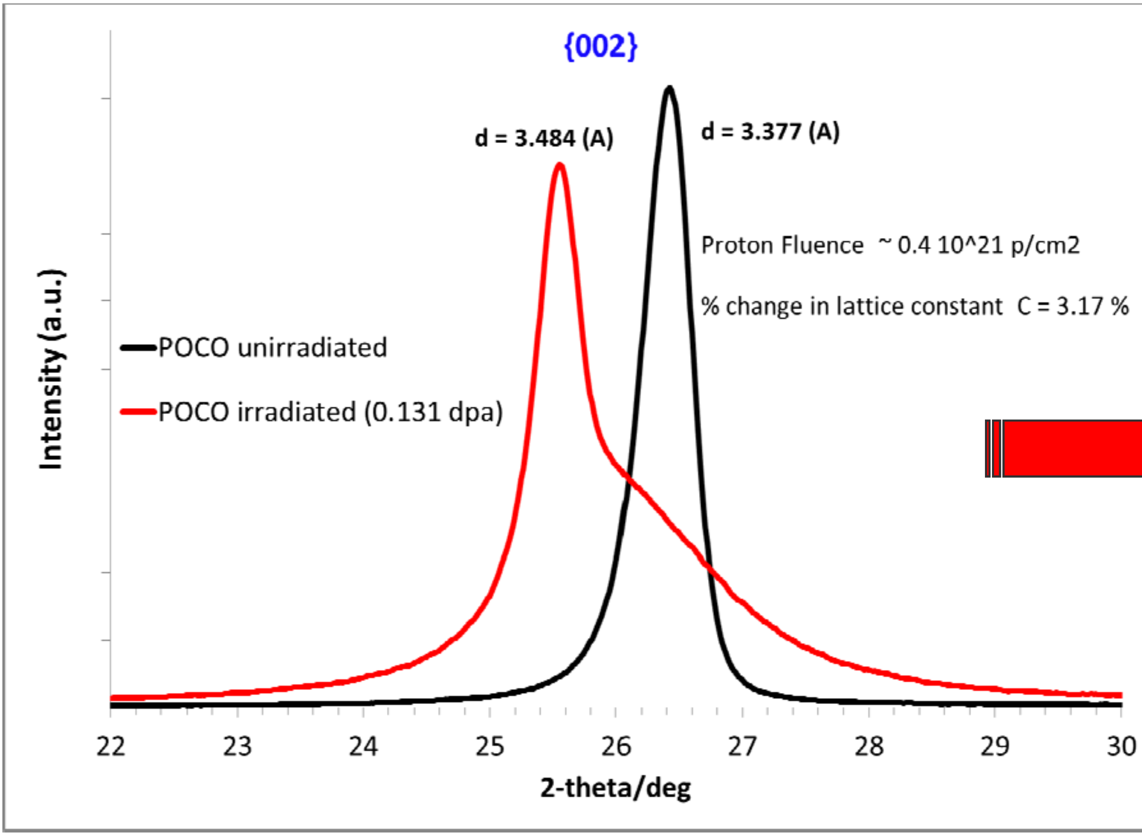
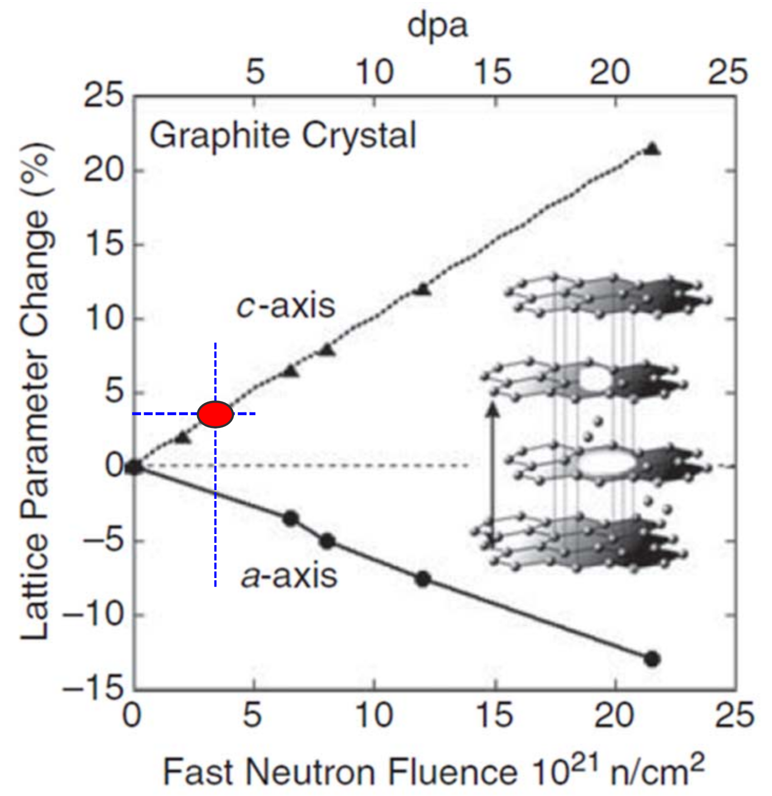
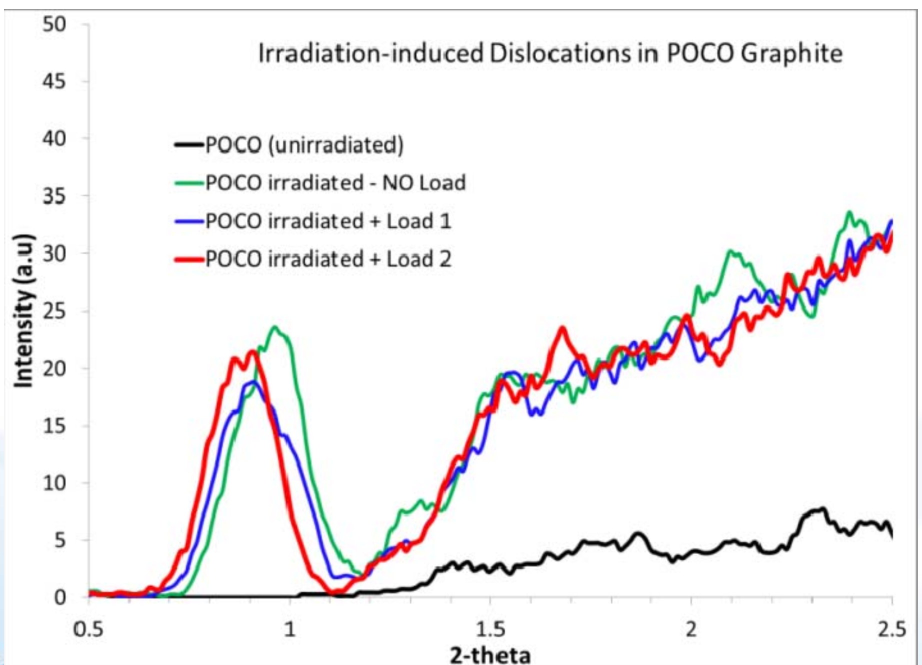
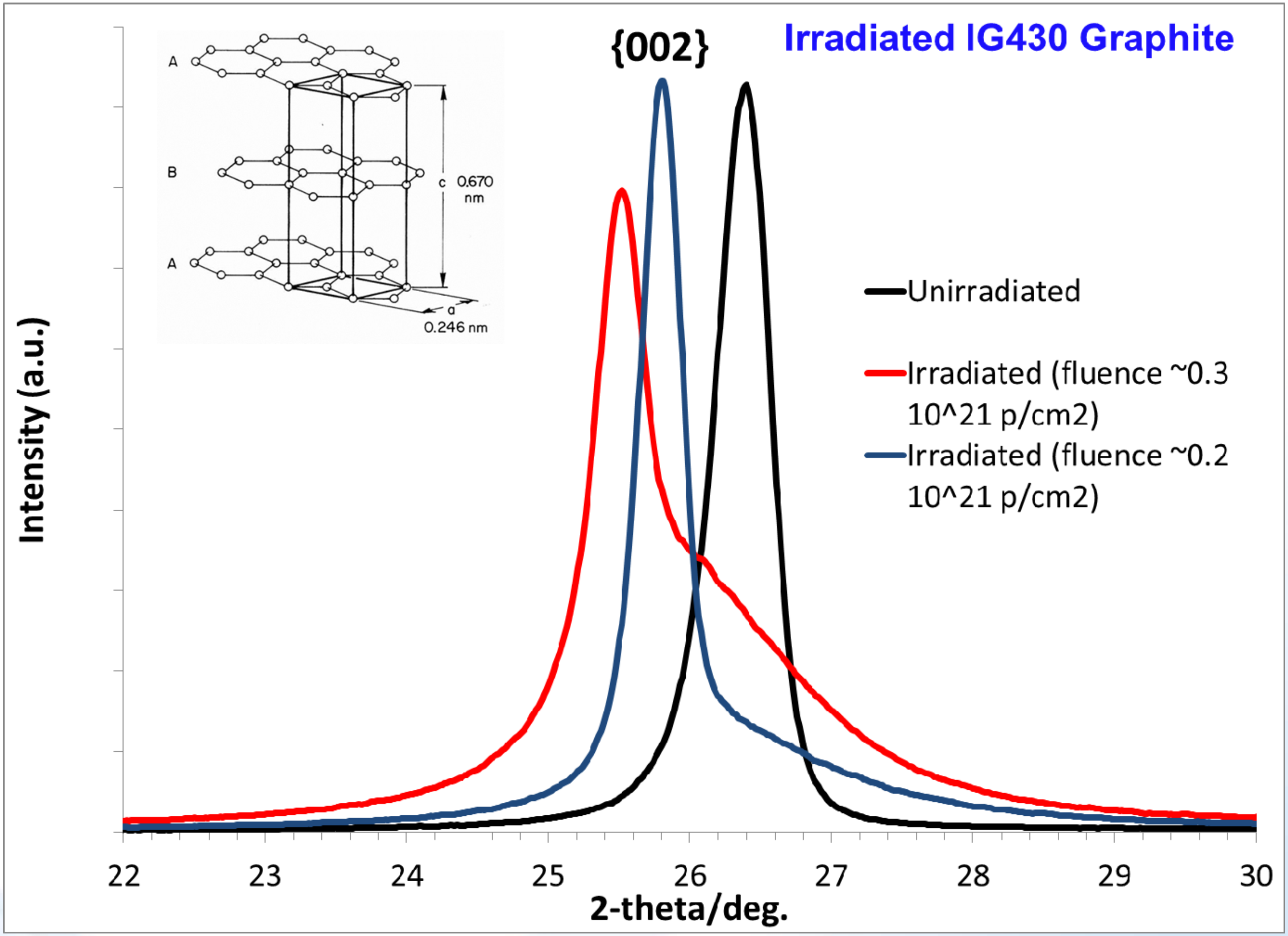




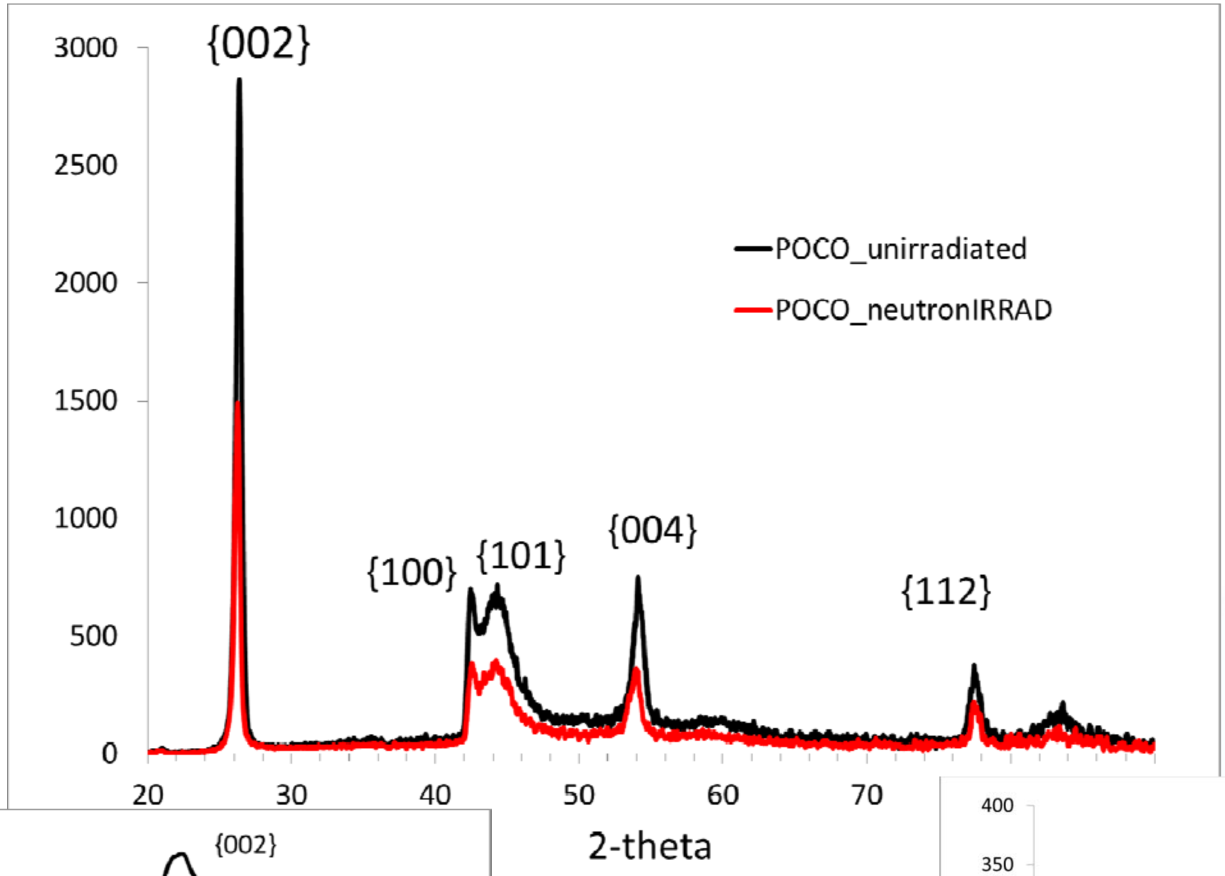


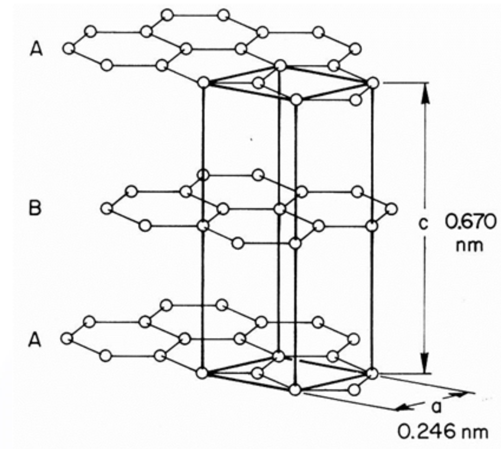
BNL EDXRD study on irradiated graphite revealed the following important correlation:
Damage expressed in terms of MEASURABLE quantities (i.e. crystal lattice changes) is achieved much faster and at much lower FLUENCE or DPA by energetic protons than fast neutrons.
BNL finding is set to a factor of ~10

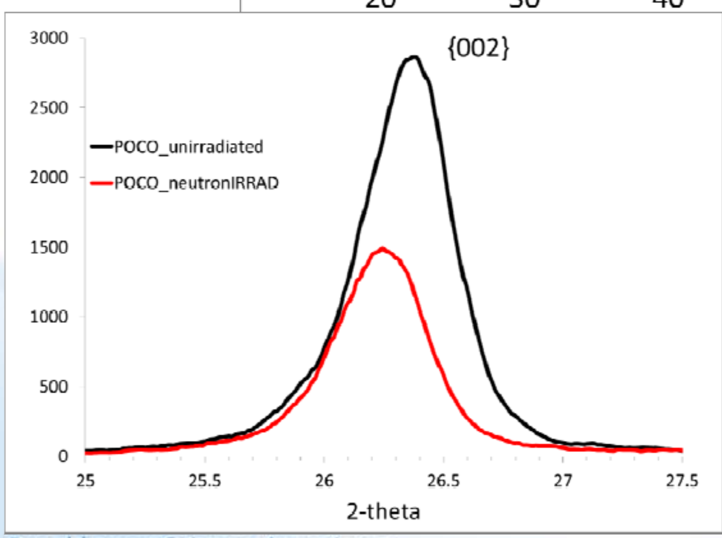


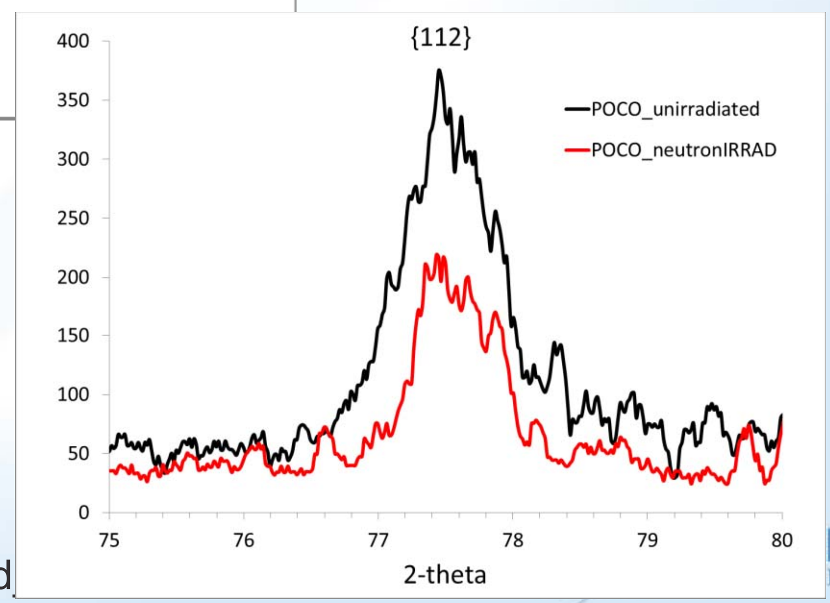
{002}
Irradiated IG430 Graphite
A
B
A
c 0.670 nm
a 0.246 nm
Unirradiated
Irradiated (fluence ~0.3 10^21 p/cm2)
Irradiated (fluence ~0.2 10^21 p/cm2)
Intensity (a.u.)
22
23
24
25
26
27
28
29
30
2-theta/deg.

Spallation Neutron Irradiation - Microstructural Changes







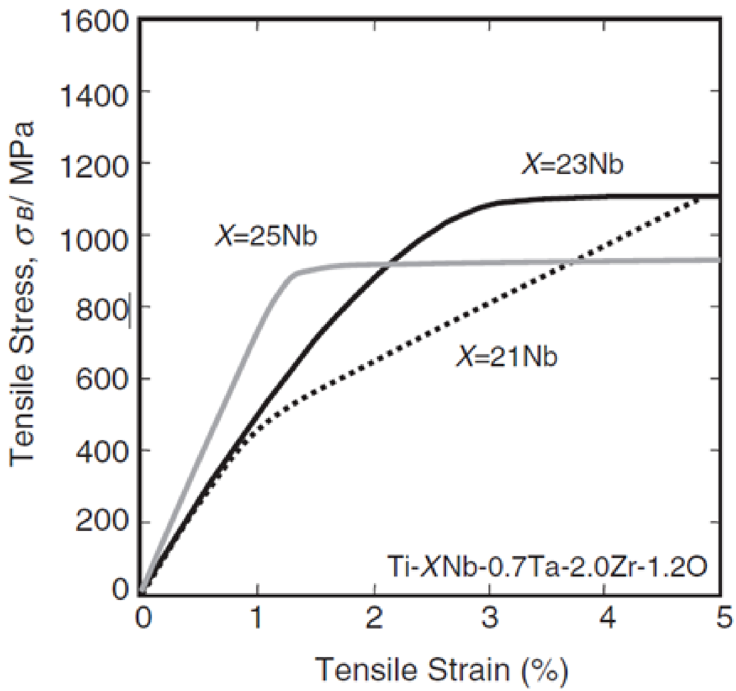


Ti alloys

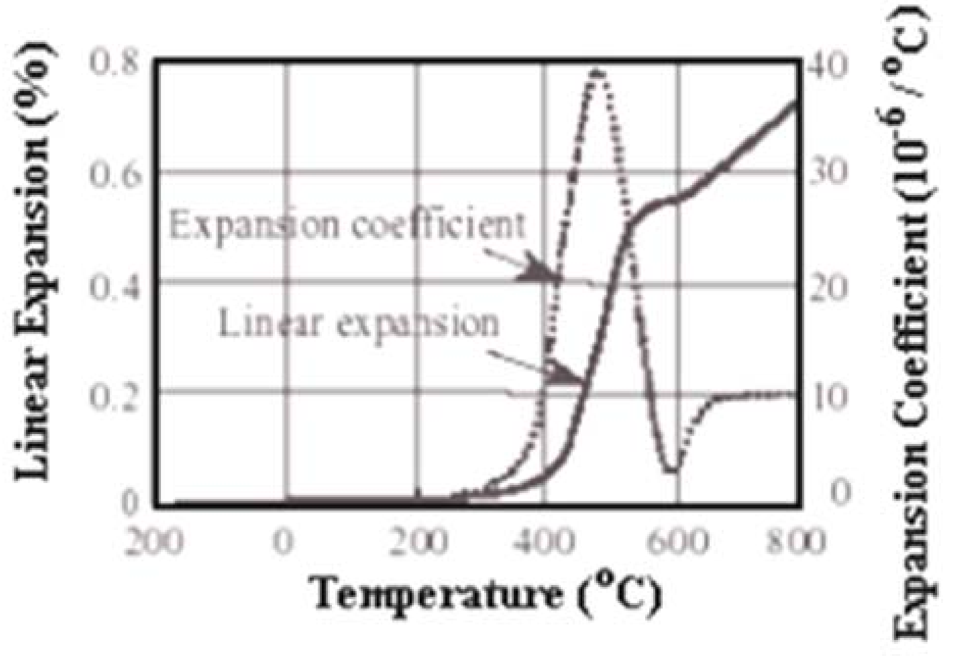
The (α + β) Ti-6Al-4V alloy
The β-titanium alloy Gum metal (Ti-21Nb-0.7Ta-2.Zr-1.2O)

Titanium Alloy Irradiation/Characterization Studies at BNL

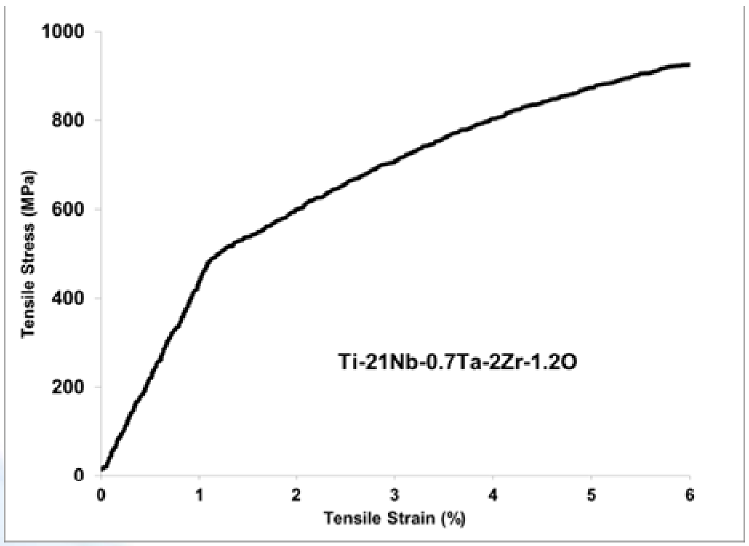
- An array of irradiation damage and post-irradiation characterization studies have been under way at BNL for Ti-alloys that include
  - (α + β) Ti-6Al-4V alloy
  - β-titanium alloy Gum metal (Ti-21Nb-0.7Ta-2.Zr-1.2O)
- Both alloys were investigated as candidates for HP targets in the Neutrino Factory initiative
- The (α + β) Ti-6Al-4V has also been studied as a substrate of ceramic nano-structured coatings for potentially nuclear applications (fast neutron and elevated temperatures)
- 200 MeV protons and spallation generated fast neutrons at the BNL complex were used for irradiation induced damage
- Macroscopic post-irradiation and EDXRD/XRD studies at the BNL synchrotrons were employed to study microstructural changes and damage



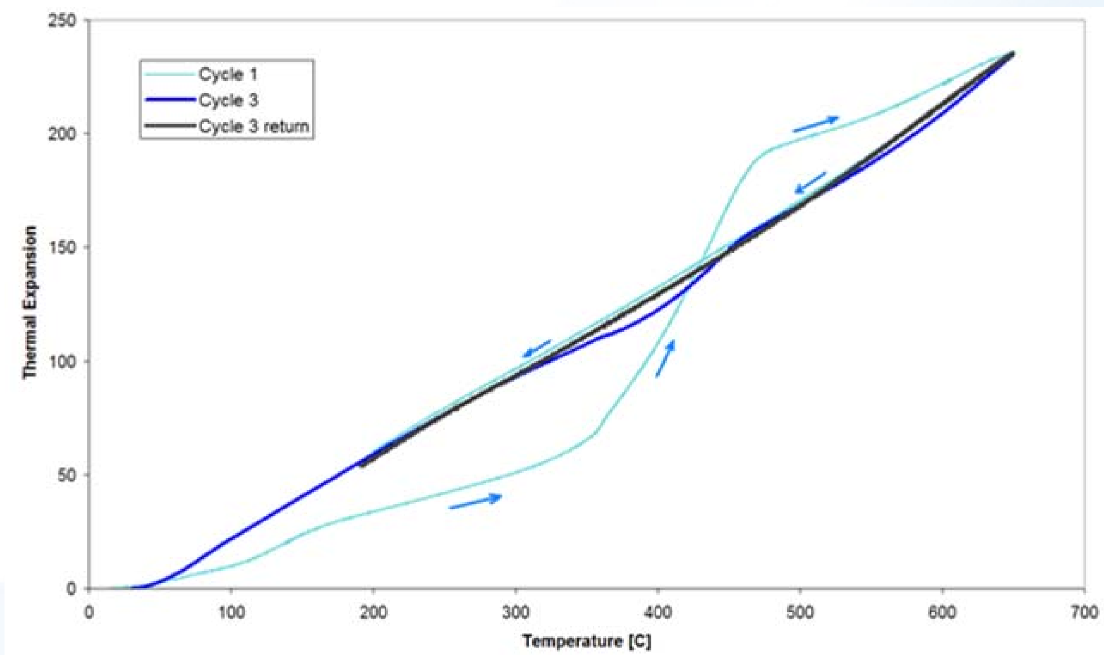
Tensile Stress, σB/ MPa
Tensile Strain (%)
X=23Nb
X=25Nb
X=21Nb
Ti-XNb-0.7Ta-2.0Zr-1.2O



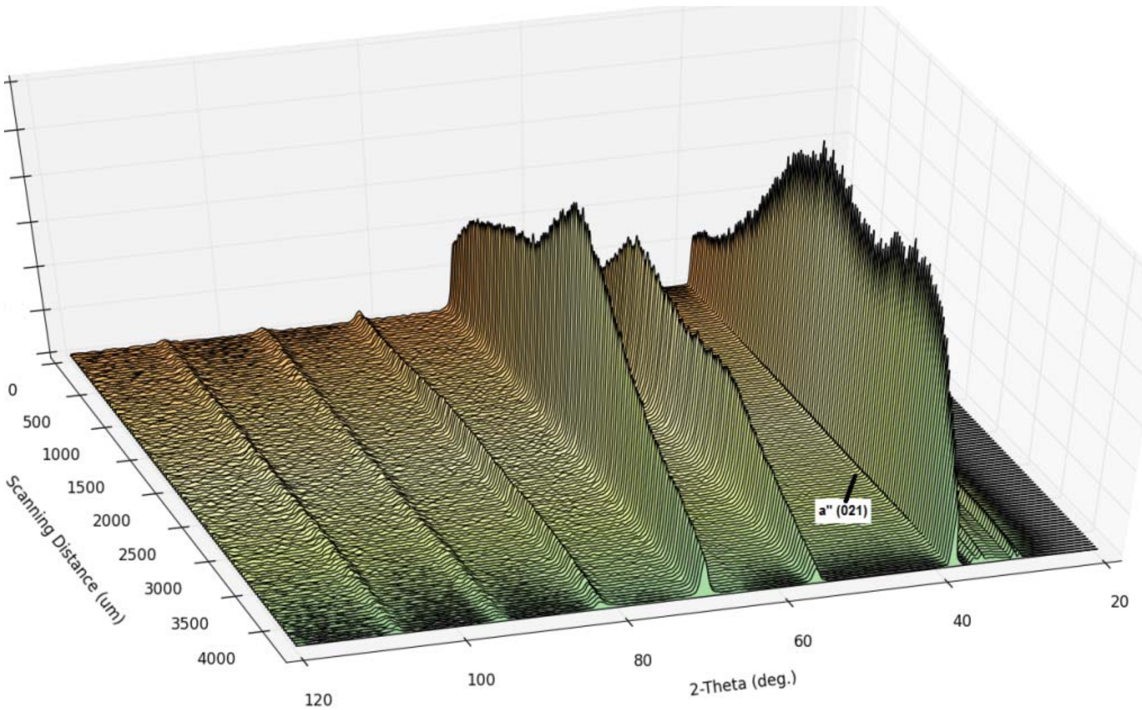
Linear Expansion (%)
Expansion Coefficient (10^-6 /°C)
Expansion coefficient
Linear expansion
Temperature (°C)

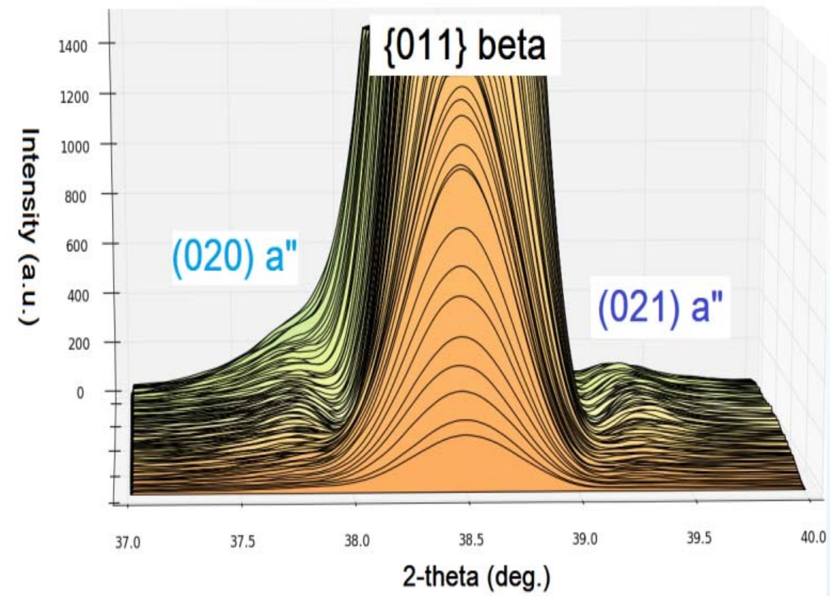


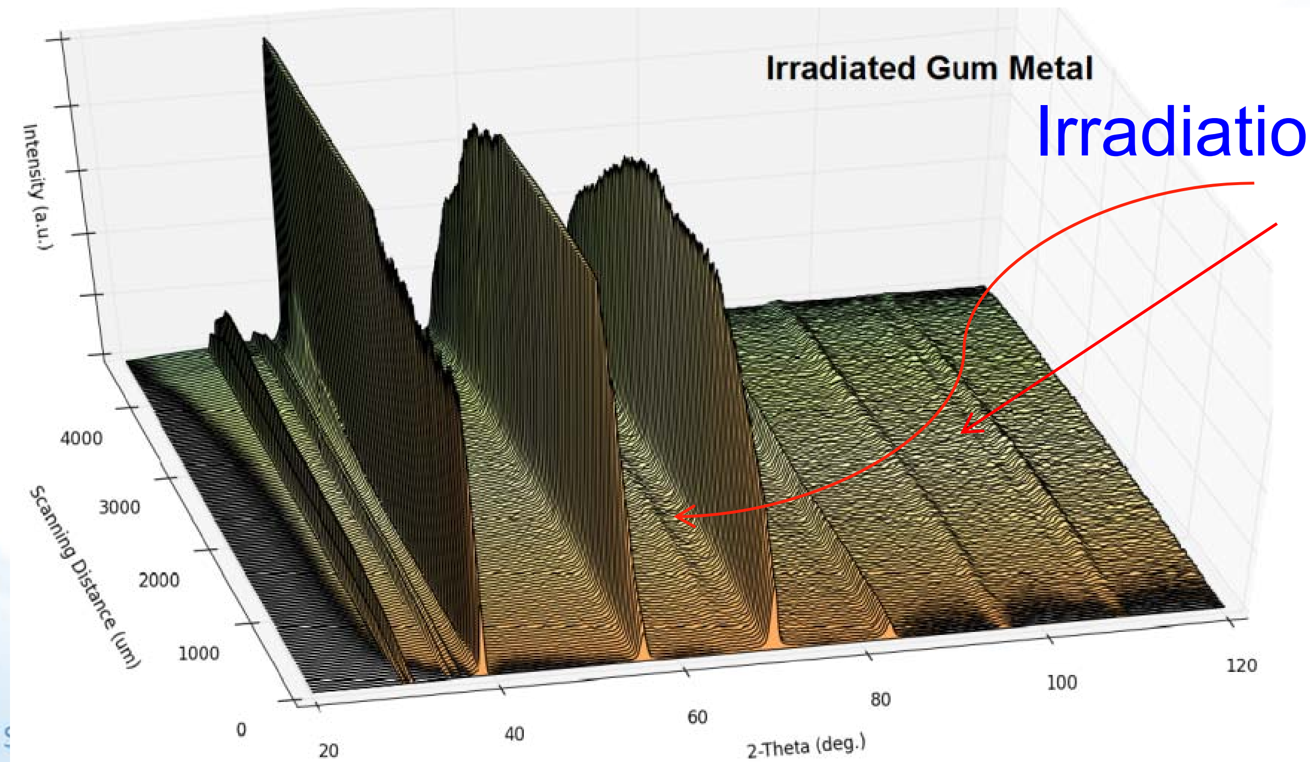
Tensile Stress (MPa)
Tensile Strain (%)
Ti-21Nb-0.7Ta-2Zr-1.2O



Cycle 1
Cycle 3
Cycle 3 return
Thermal Expansion
Temperature [C]







Irradiation-induced phases

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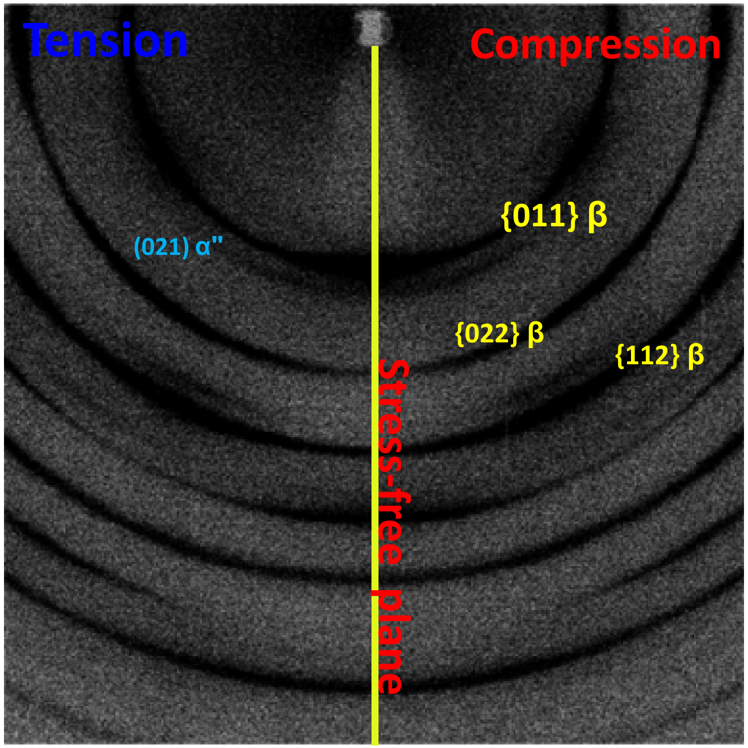
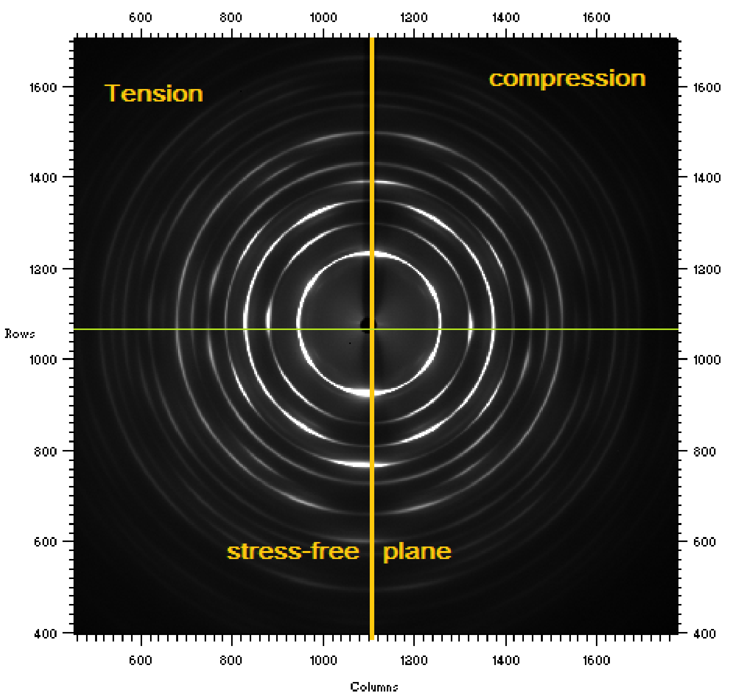
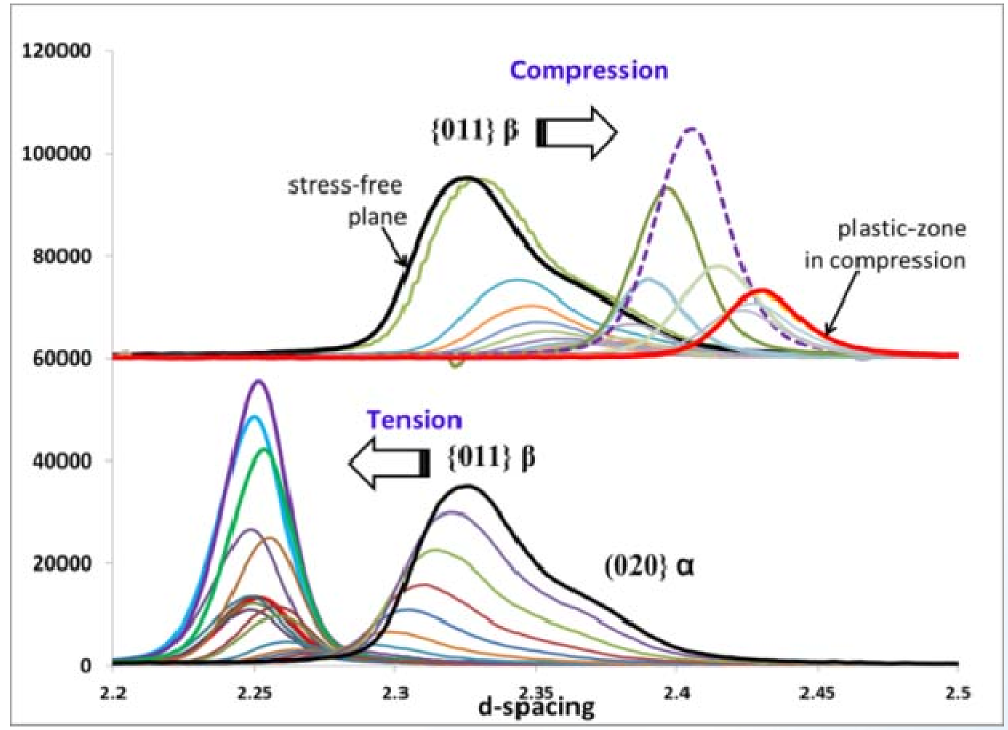
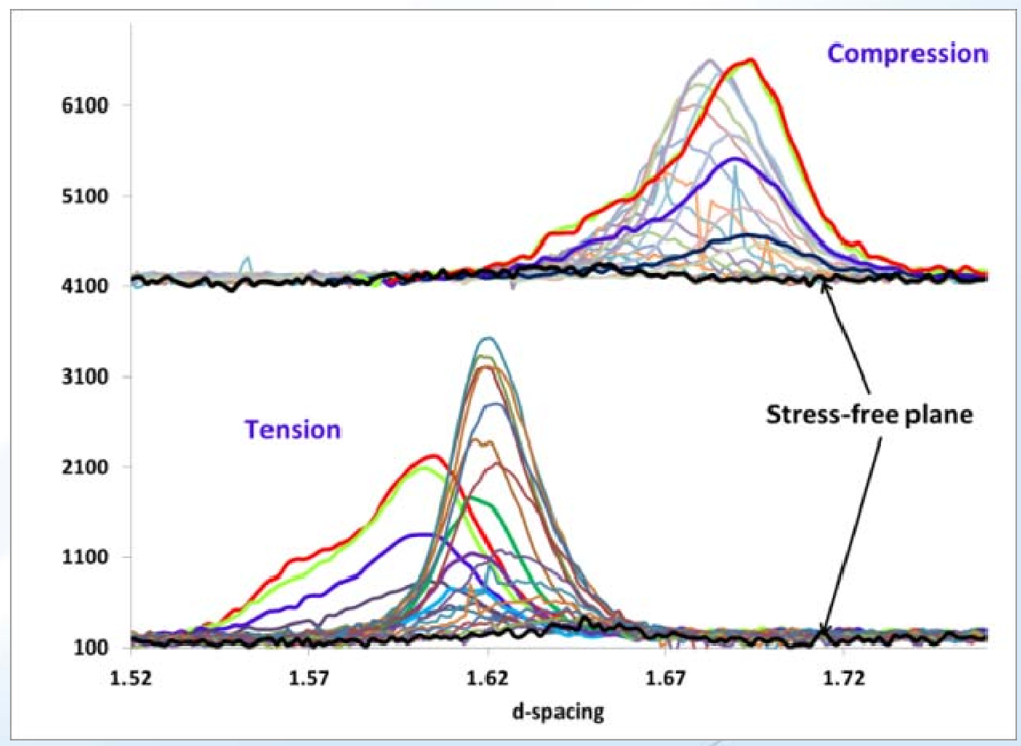
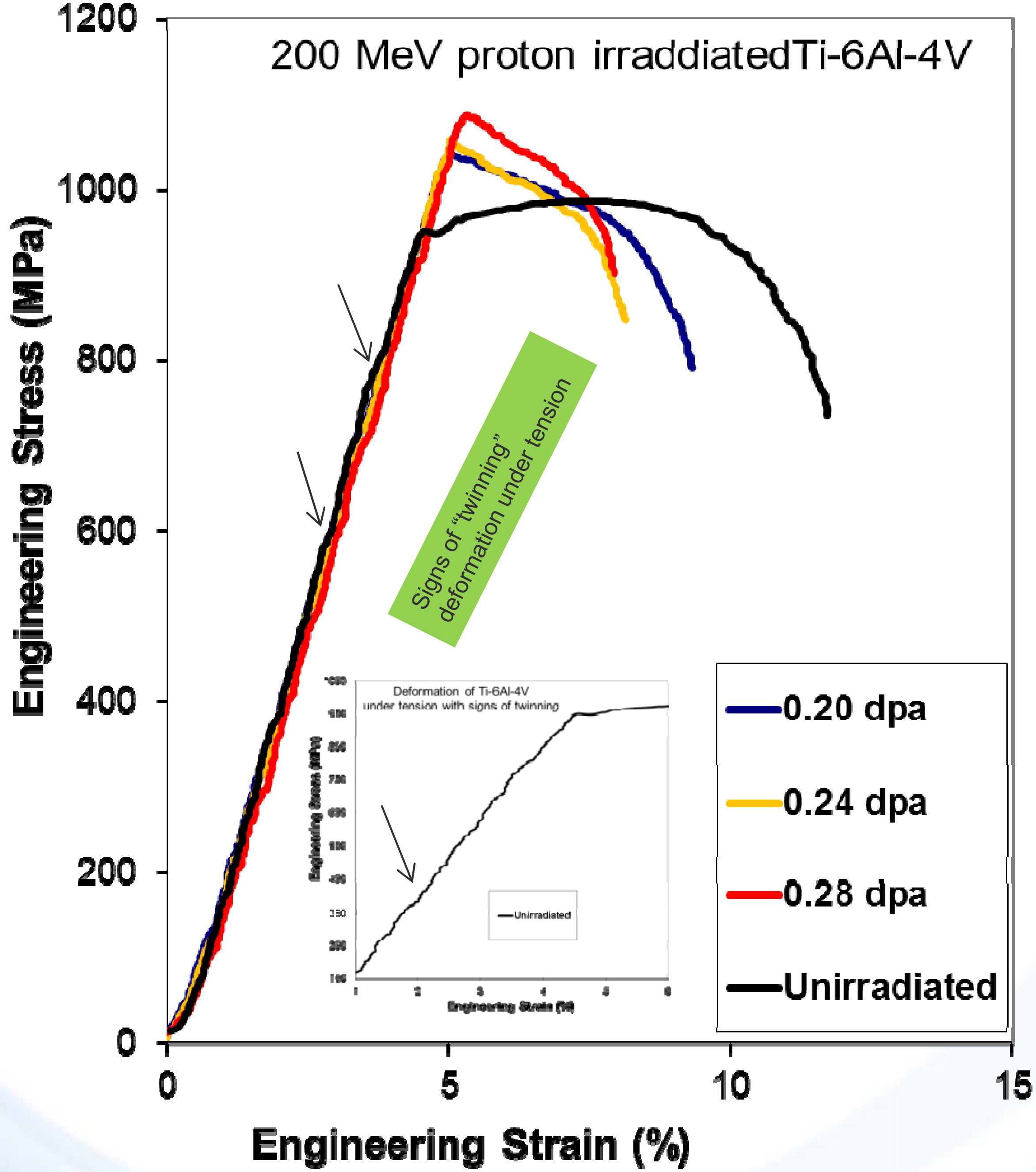

Tension
Compression
(021) α"
{011} β
{022} β
{112} β
Stress-free plane

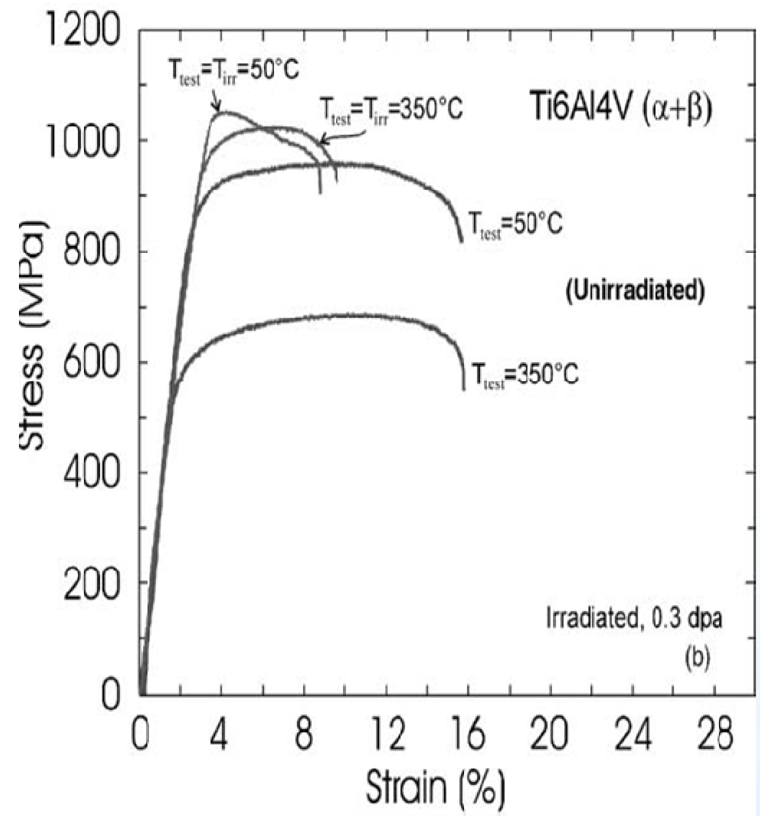

Tension
compression
stress-free plane
Rows
Columns


Compression
{011} β
stress-free plane
plastic-zone in compression
Tension
{011} β
(020) α
d-spacing


Compression
Tension
Stress-free plane
d-spacing

Bro
Sim



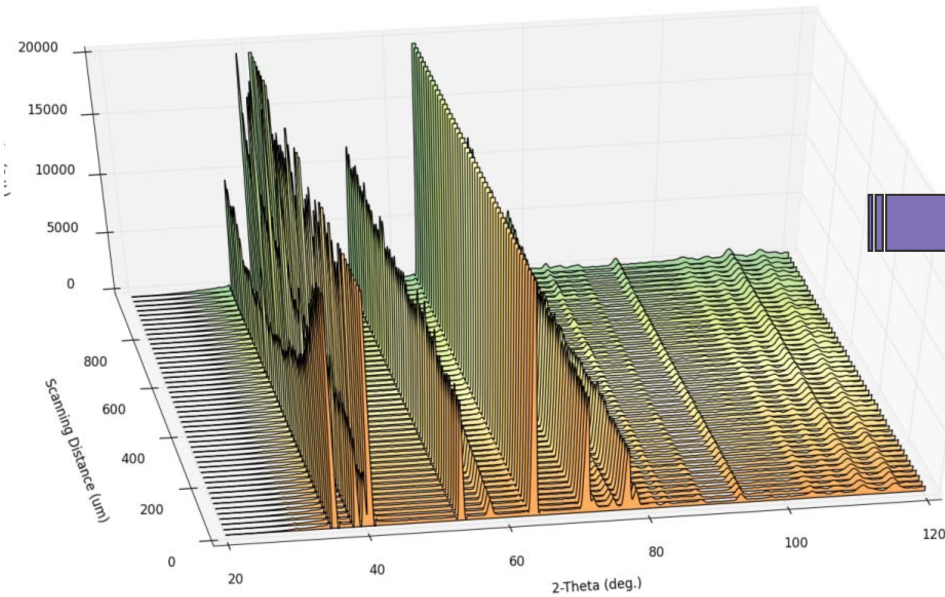


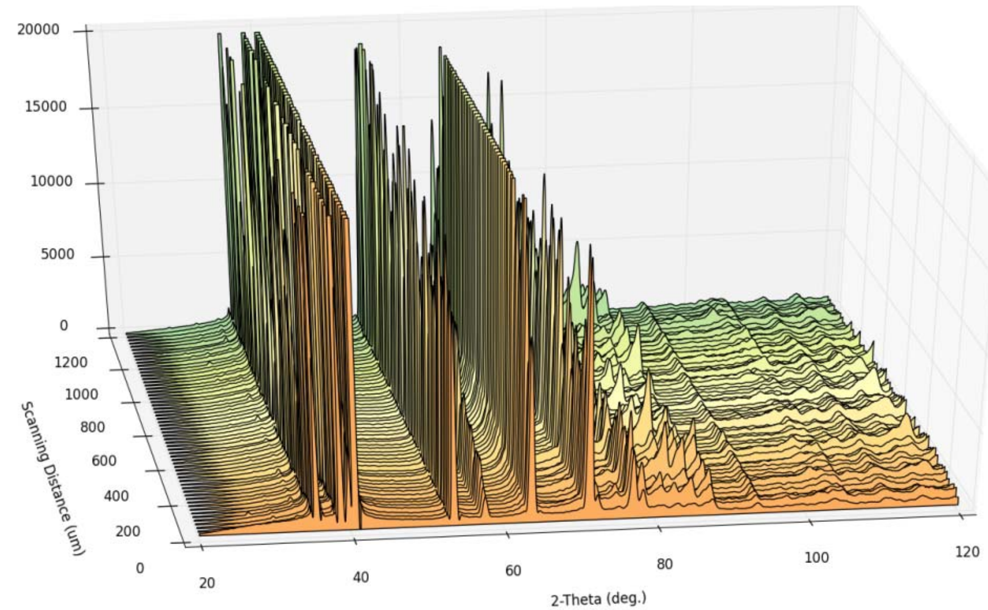
Tensile and fracture toughness properties of unirradiated and neutron irradiated titanium alloys
S. T€ahtinen a,*, P. Moilanen a, B.N. Singh b, D.J. Edwards c

Journal of Nuclear Materials 307–311 (2002) 416–420

Pristine Ti-6Al-4V

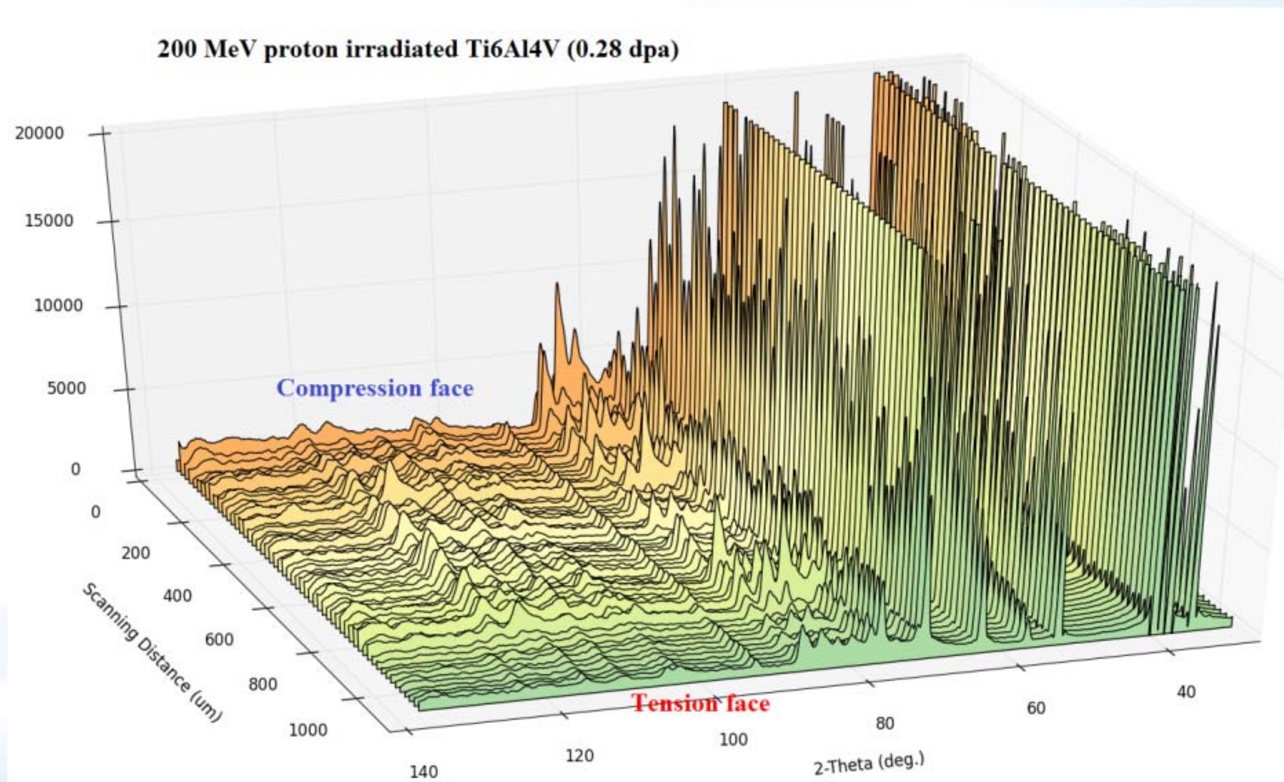
Proton-irradiated Ti-6Al-4V

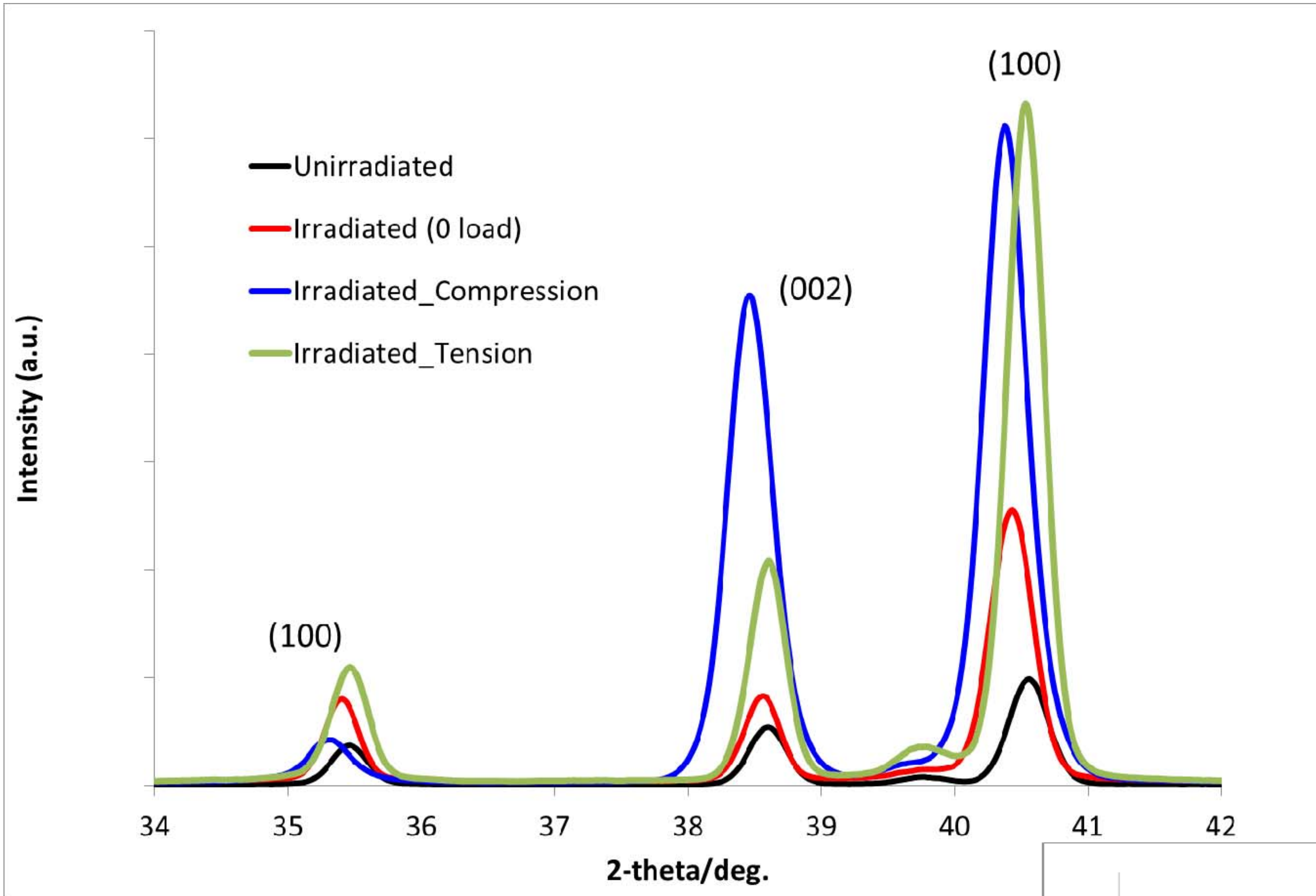


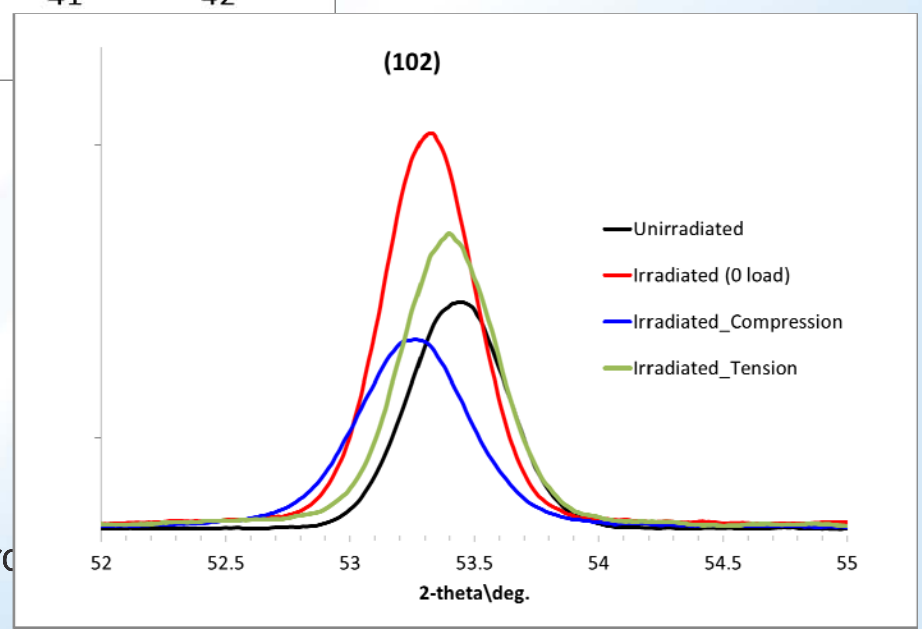


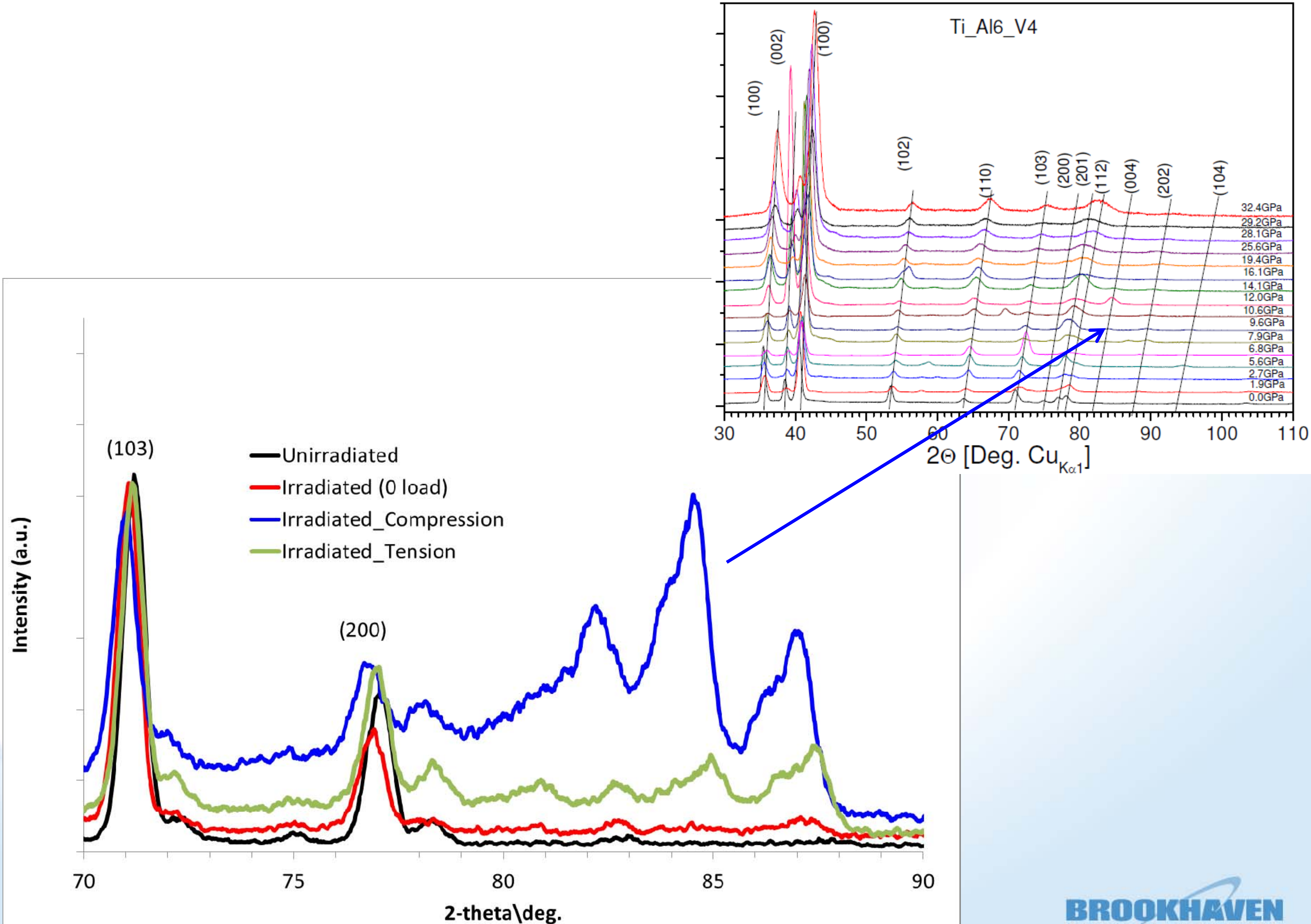
Proton-irradiated Ti-6Al-4V
Under four-point bending state

NOTE the distinct difference in diffraction between tension and compression!




(100)
(002)
(100)
Unirradiated
Irradiated (0 load)
Irradiated_Compression
Irradiated_Tension
Intensity (a.u.)
34
35
36
37
38
39
40
41
42
2-theta/deg.


(102)
Unirradiated
Irradiated (0 load)
Irradiated_Compression
Irradiated_Tension
52
52.5
53
53.5
54
54.5
55
2-theta\deg.


Ti_Al6_V4
(100)
(002)
(100)
(102)
(110)
(103)
(200)
(201)
(112)
(004)
(202)
(104)
32.4GPa
29.2GPa
28.1GPa
25.6GPa
19.4GPa
16.1GPa
14.1GPa
12.0GPa
10.6GPa
9.6GPa
7.9GPa
6.8GPa
5.6GPa
2.7GPa
1.9GPa
0.0GPa
30 40 50 60 70 80 90 100 110
2Θ [Deg. Cu$_{K\alpha 1}$]
(103)
(200)
Unirradiated
Irradiated (0 load)
Irradiated_Compression
Irradiated_Tension
Intensity (a.u.)
70 75 80 85 90
2-theta\deg.

Tungsten and other Refractory Metals

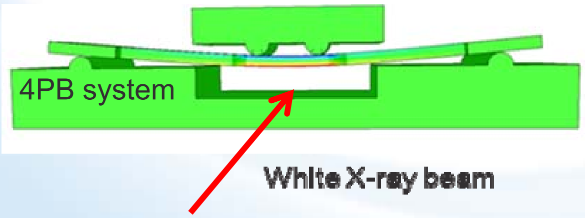
N. Simos, R. Bennett, et al

W and other refractory metals Irradiation/Characterization SUMMARY

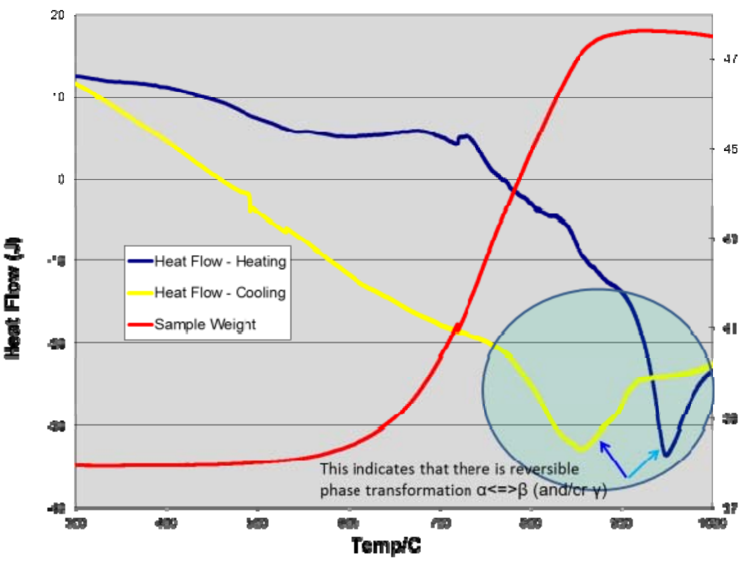
- Irradiation damage and post-irradiation characterization studies have been under way at BNL on refractory metals
  - **Tungsten,** Tantalum and Molybdenum (HP Targets , LHC collimators and Fusion Reactors)
- 200 MeV protons at the BNL Linac/BLIP and 28 MeV proton at Tandem (Mo) were used for irradiation induced damage
- Macroscopic post-irradiation analysis addressed:
  - Irradiation-induced ductility loss and strength increase
  - Thermal stability and exhibited “anomalies”
  - Oxidation and erosion in irradiated W under cooling water

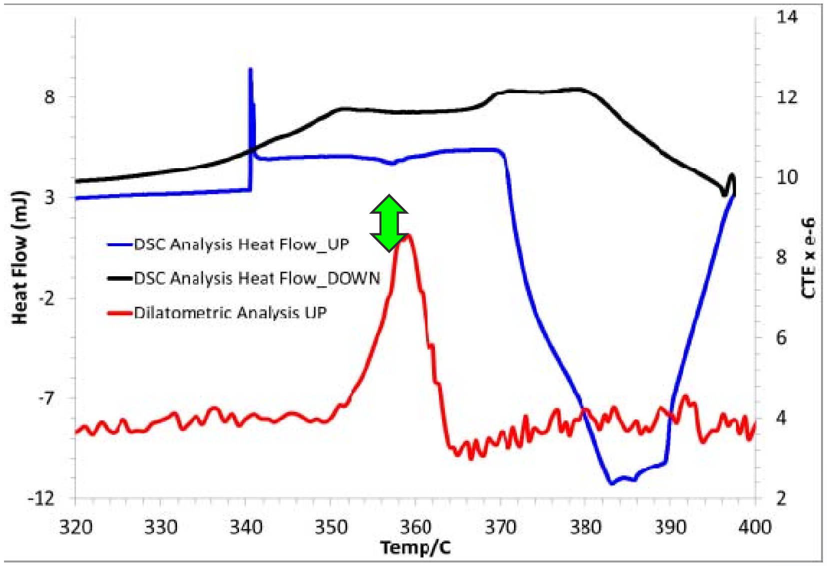
W specimen seriously eroded in water cooling while in proton beam

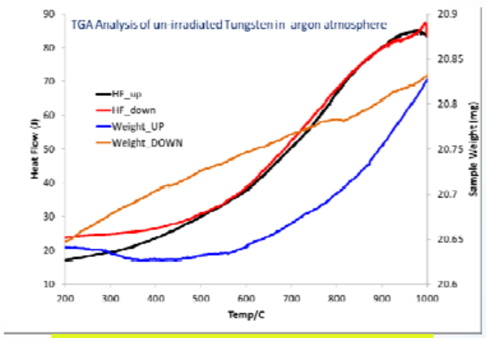
- Microscopic assessment/annealing behavior techniques employed
  - Thermal annealing
  - Scanning Electron Microscopy and EDS
  - DSC and TGA
- EDXRD/XRD studies at the BNL synchrotrons were employed to study irradiation-induced microstructural changes with in-situ loading



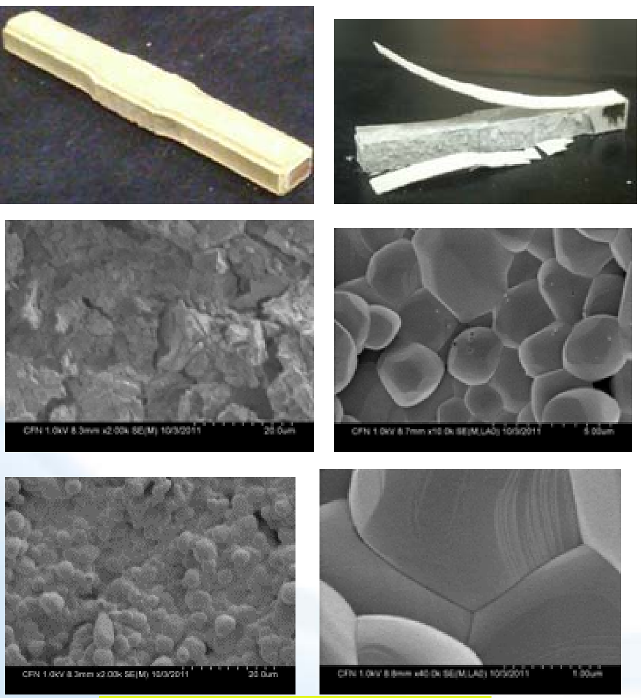
Thermo-mechanical Characterization W behavior (W ➔ WO_3) to temperatures reaching 1050 C



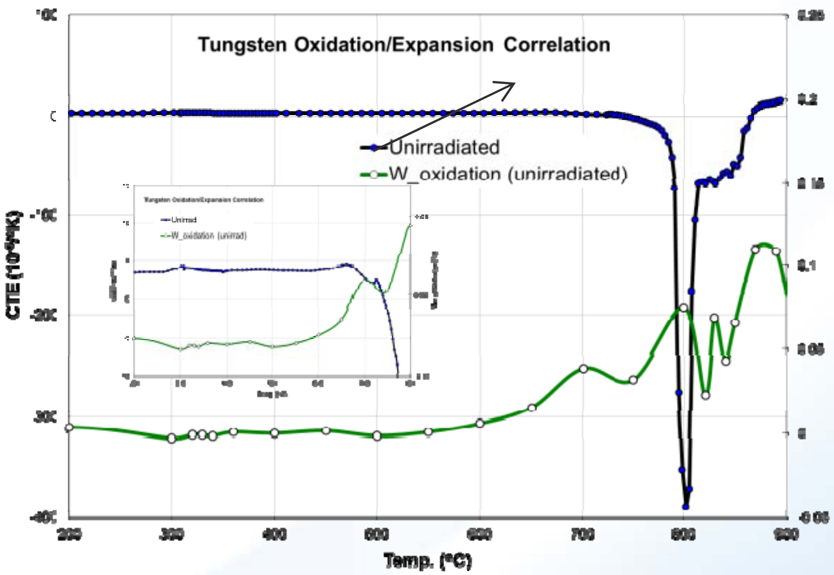


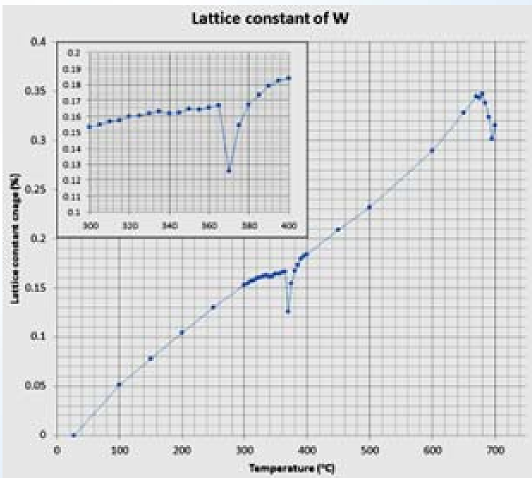


Asymmetry in weight gain/loss
Heat Flow asymmetry < 500 C



Evolution of W oxidation





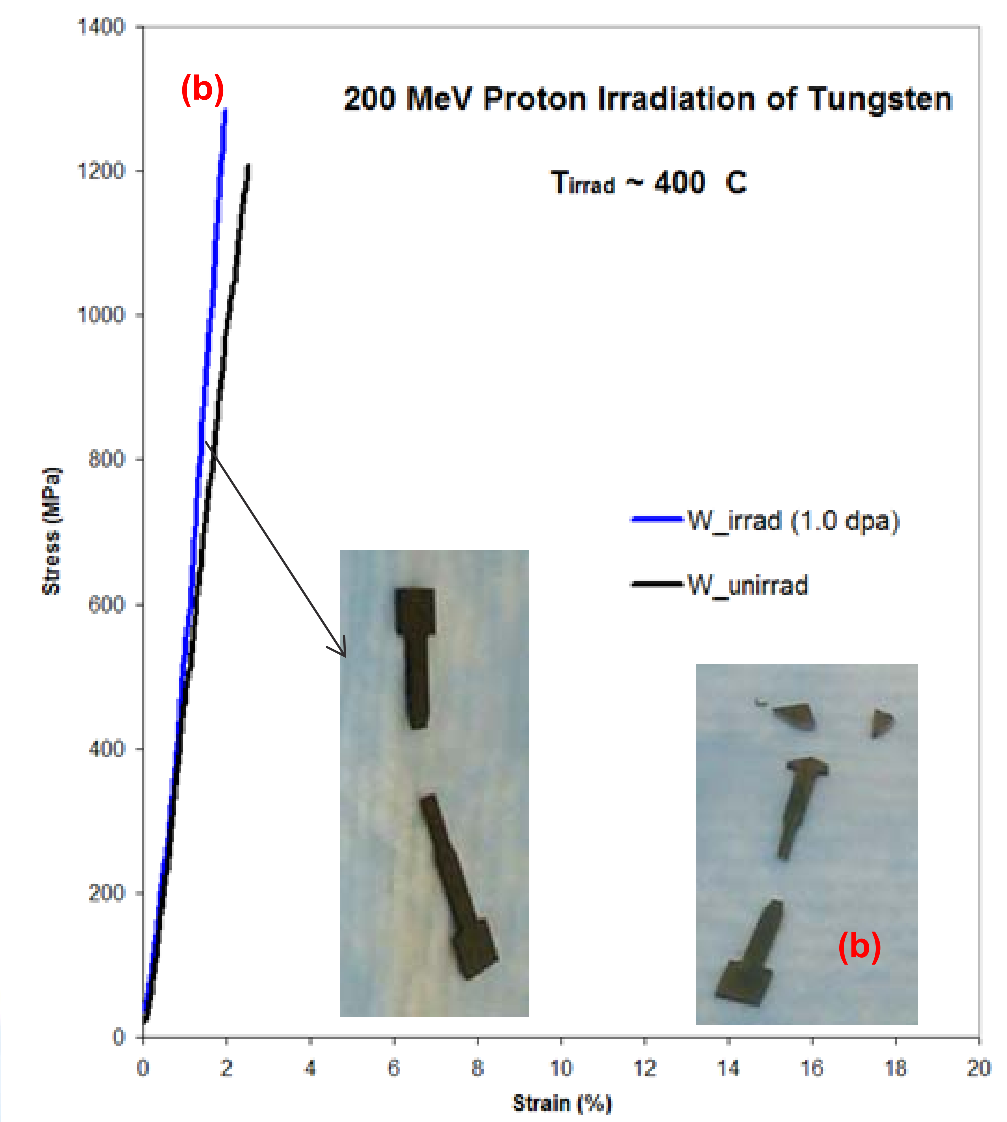
W X-ray analysis with in-situ heating measuring Lattice constant and confirming the presence of "anomalies"

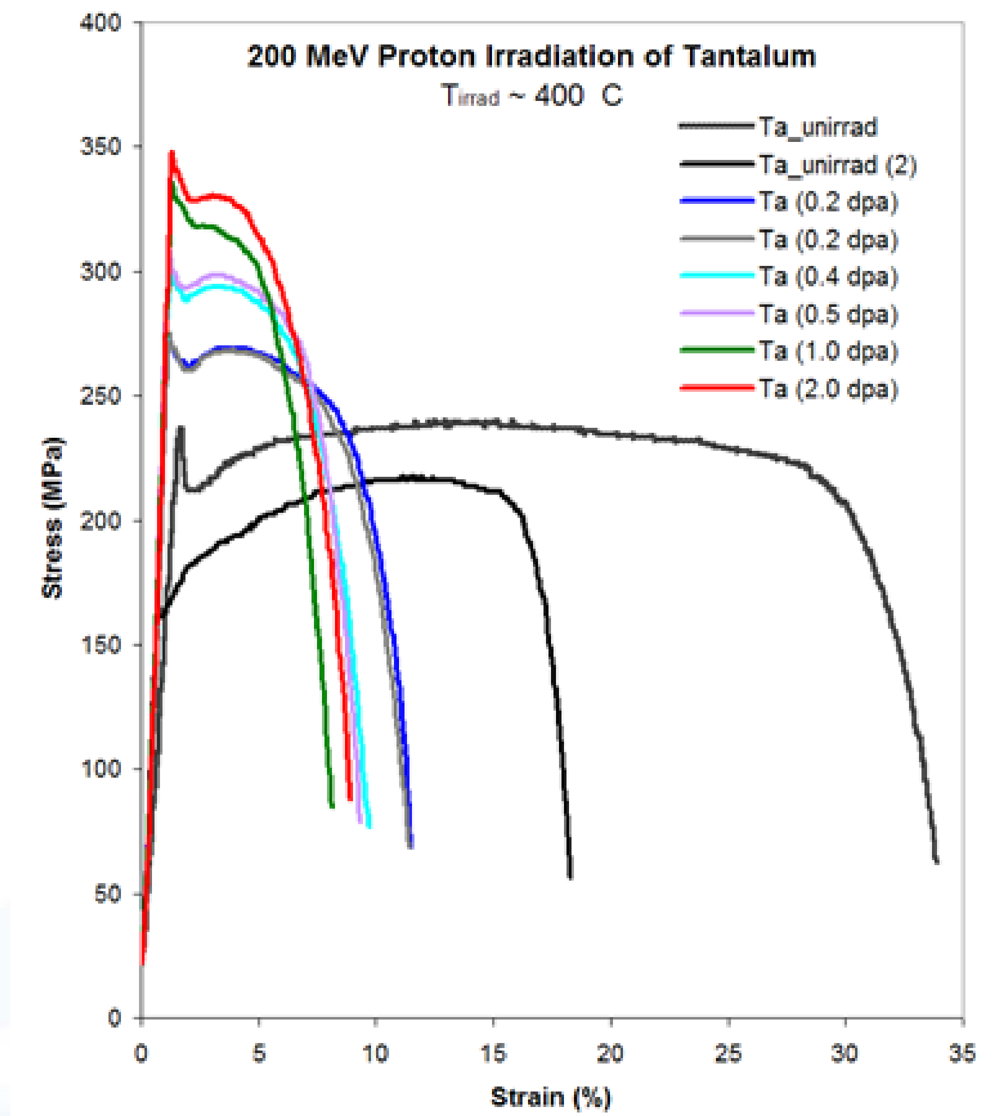
BROOKHAVEN
NATIONAL LABORATORY

W Studies- BNL-Simos

Thermo-mechanical Characterization and Fracture

Study of ductility loss and gain of strength in comparison with Ta





Fractured W specimens

Irradiated sample (b) experienced what appears as shock-induced fracture at head

EDXRD Studies of Irradiated W

EDXRD studies at BNL NSLS synchrotron of irradiated W under stress revealed that with the stable **α-tungsten** with the bcc lattice the **metastable β-tungsten** phase with cubic lattice is also present.

The metastable β-tungsten converts to α-tungsten at T>600 C while the fcc γ-tungsten converts to α-tungsten at T>700 C (traces present).

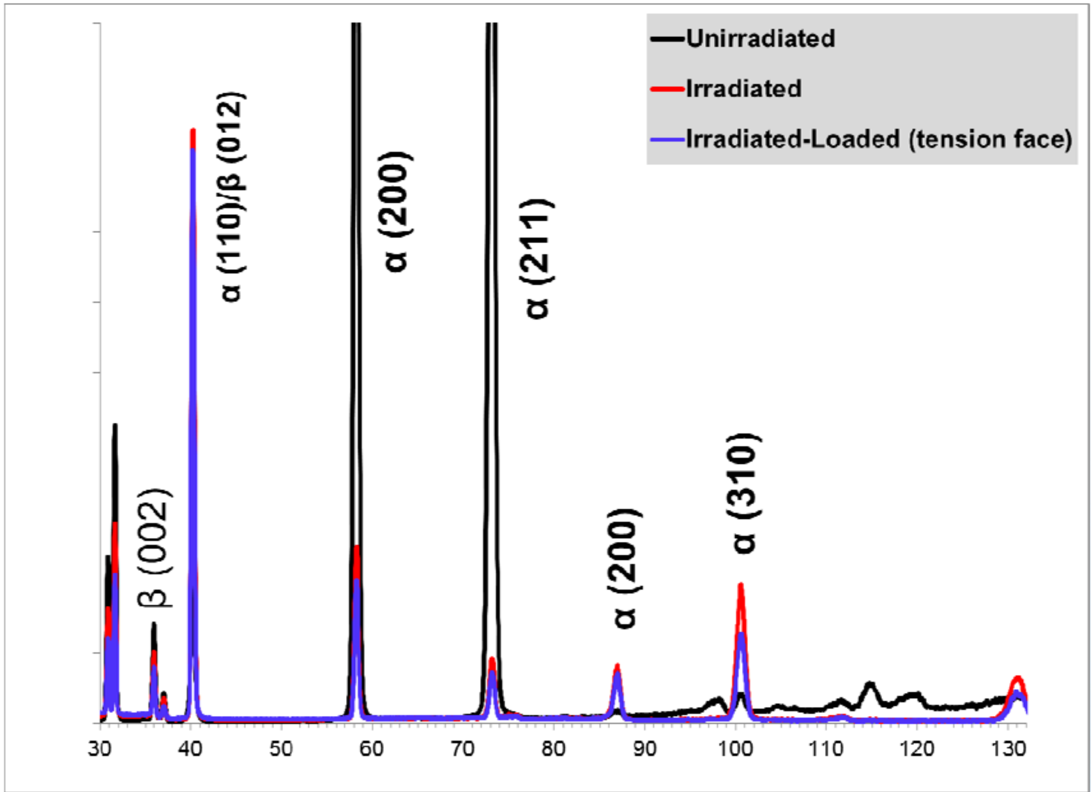
The presence of the two other phases (β-tungsten primarily) and their transition to α-tungsten **EXPLAINS** the various "anomalies" observed over these ranges.

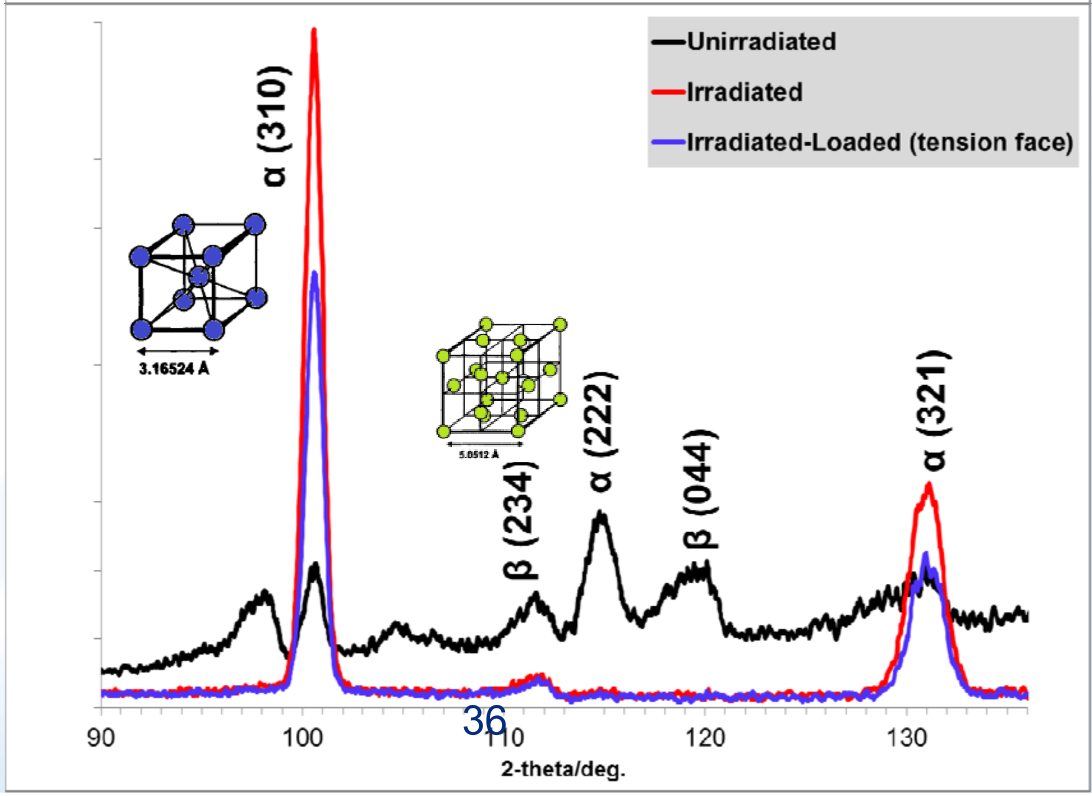
The lattice parameter for α-W reported as **3.16524 (A)**

In the BNL studies = 3.1615 A (un-irradiated)
= 3.1416 A (irradiated)

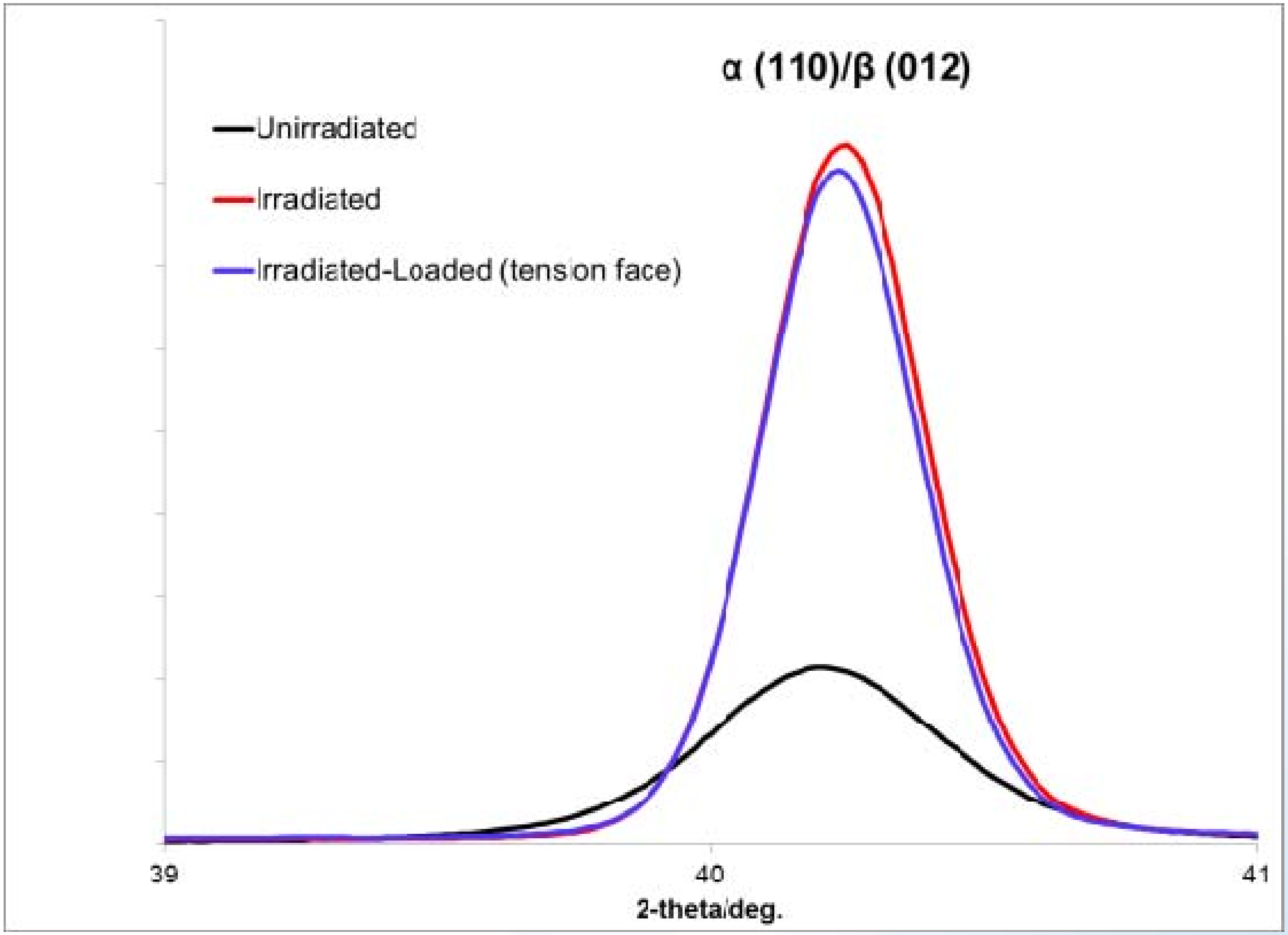
β-W reported lattice parameter **5.0512 (A)**

BNL study: 5.0227 (un-irradiated W)
4.9935 (irradiated W)





Tungsten and other Refractory Metals

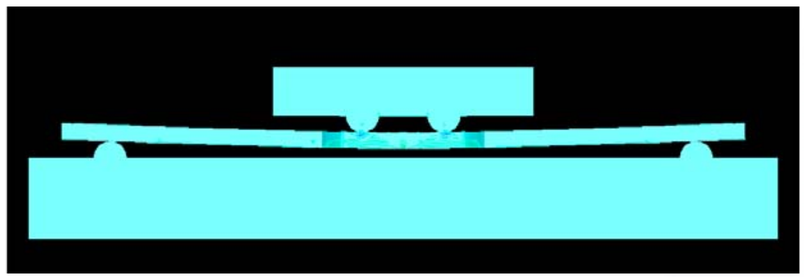


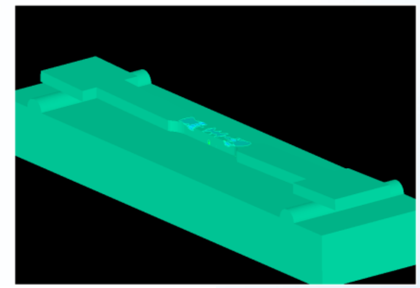
Effect of high stress on
X-ray diffraction of W

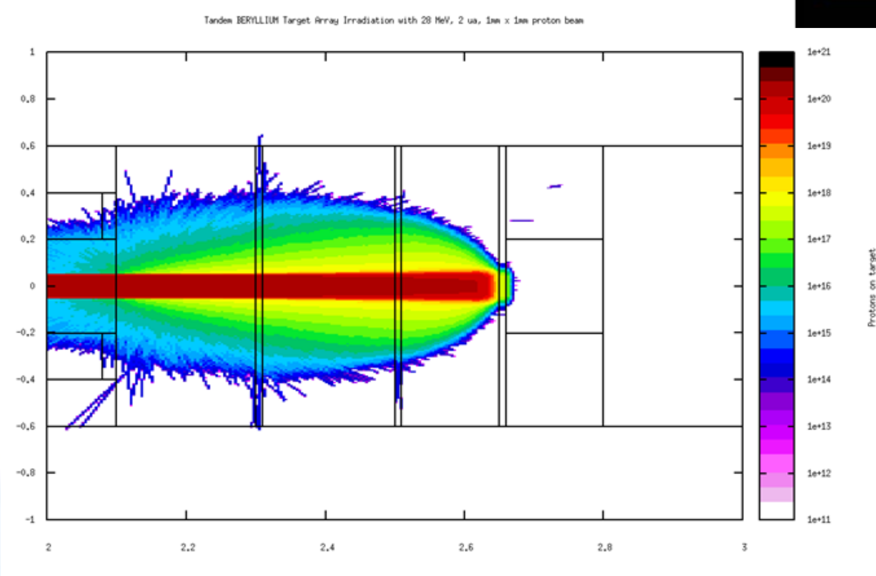
Near-future Plans

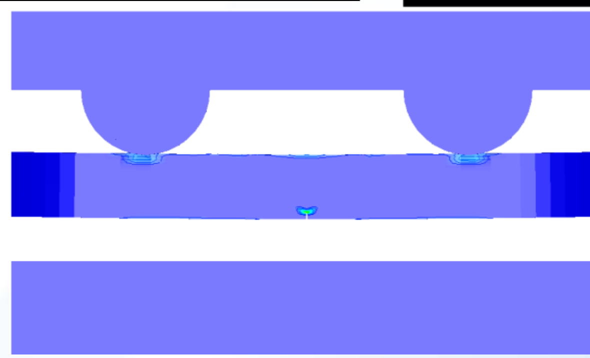
Focused-beam (28 MeV and heavy ions) experiments at BNL Tandem looking into

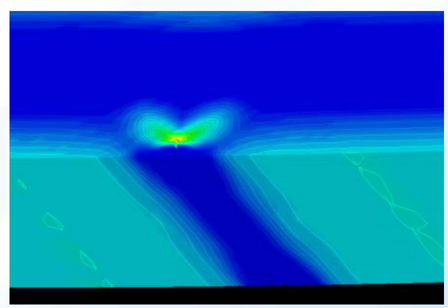
- Fracture Toughness
- Crack Propagation and crack tip phase transformations

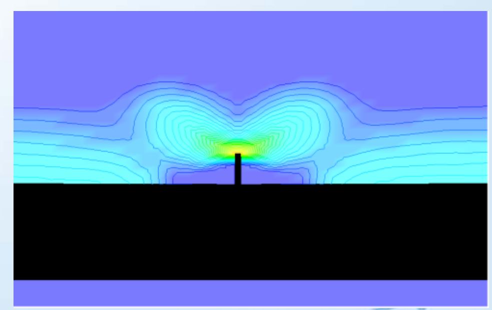




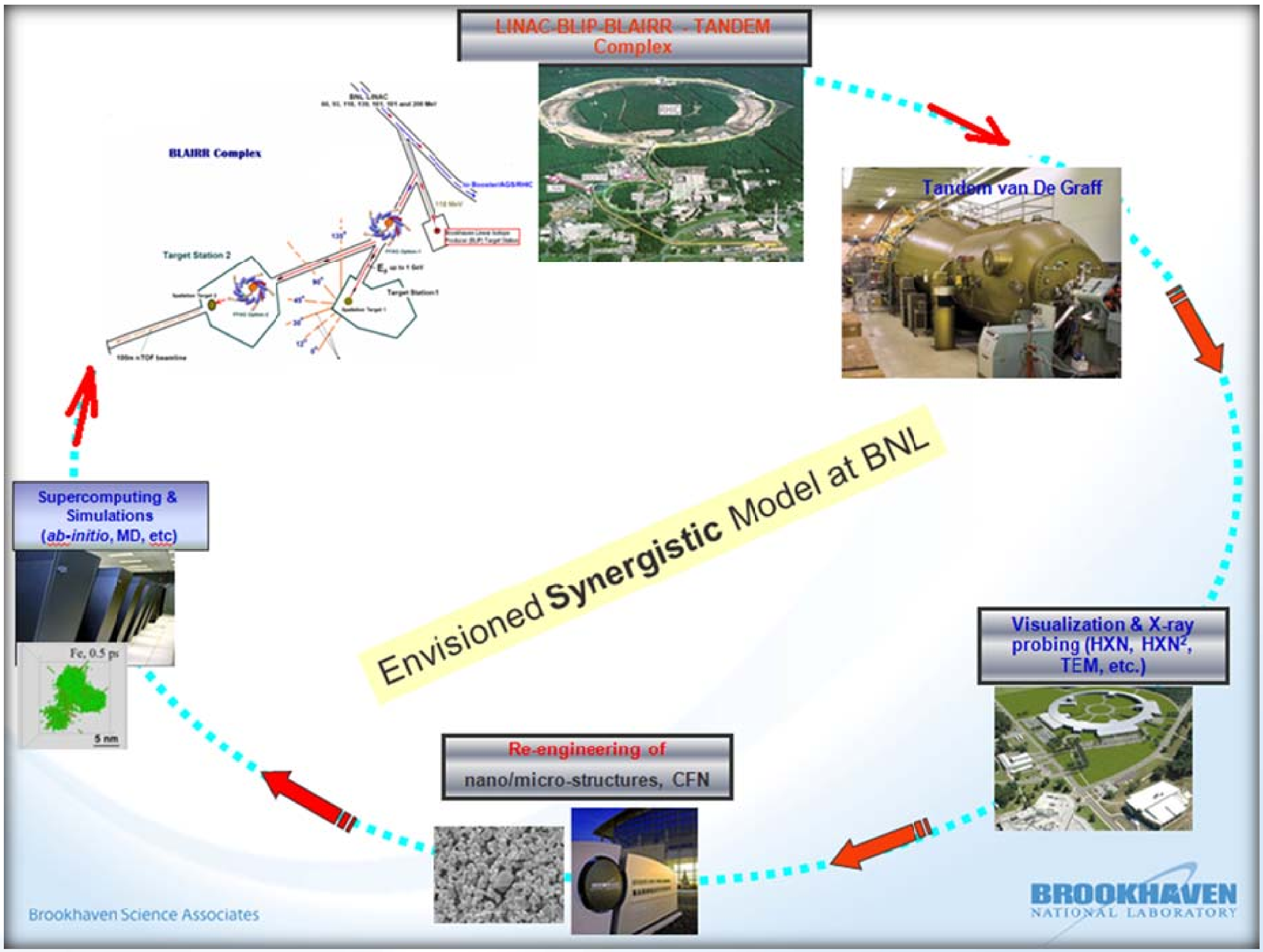






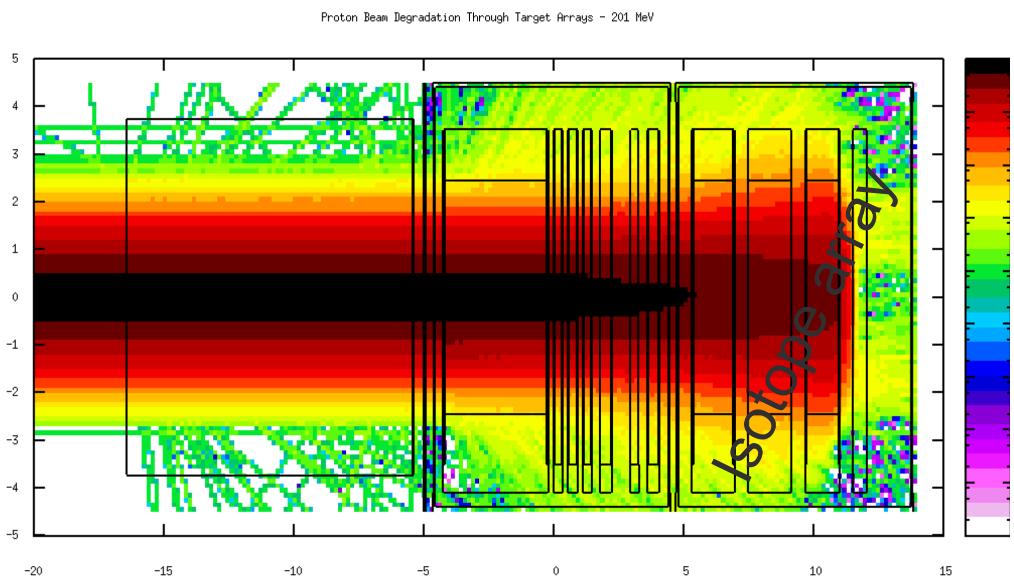


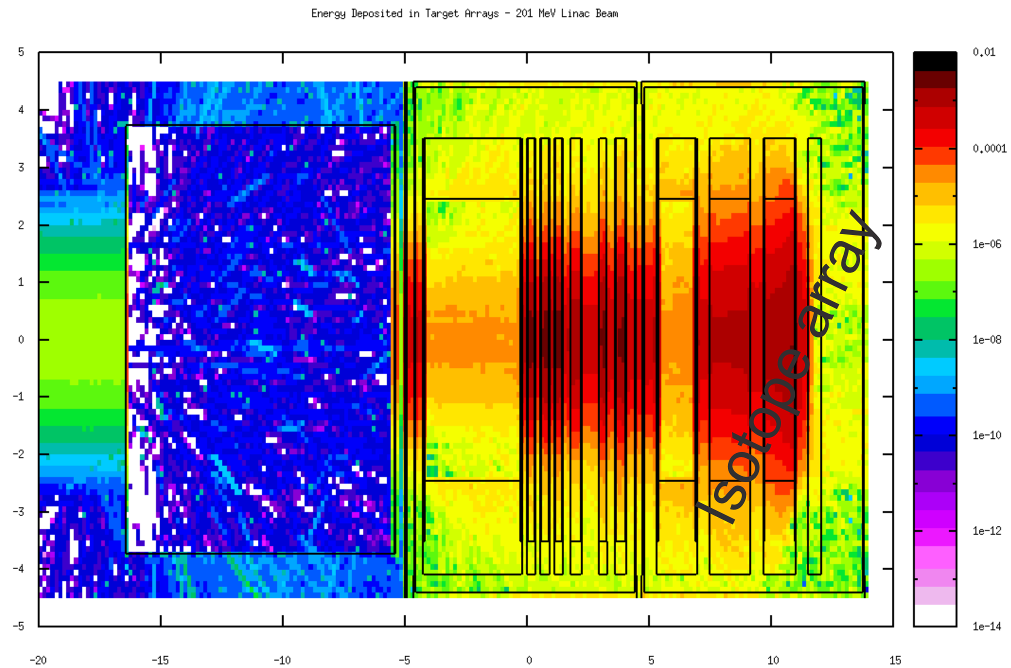
B-UP Slides

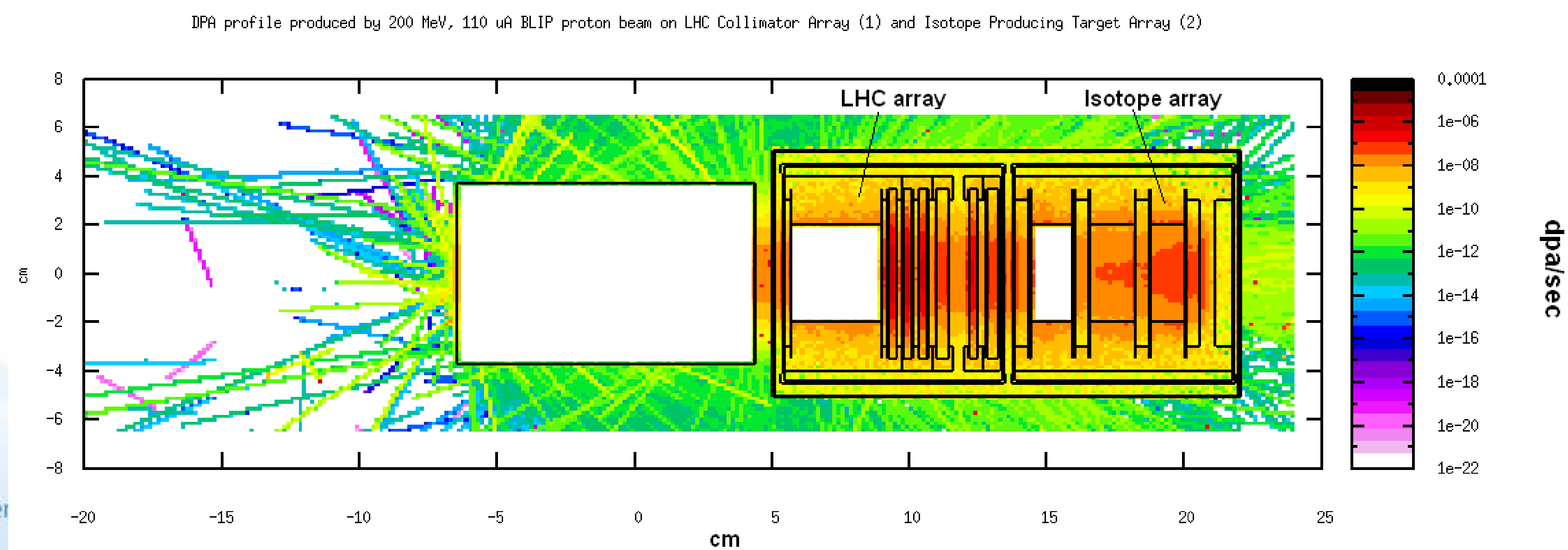

LINAC-BLIP-BLAIRR - TANDEM Complex
BLAIRR Complex
Target Station 2
Tandem van De Graff
Envisioned Synergistic Model at BNL
Supercomputing & Simulations (ab-initio, MD, etc)
Fe, 0.5 ps
5 nm
Visualization & X-ray probing (HXN, HXN², TEM, etc.)
Re-engineering of nano/micro-structures, CFN
Brookhaven Science Associates
BROOKHAVEN NATIONAL LABORATORY

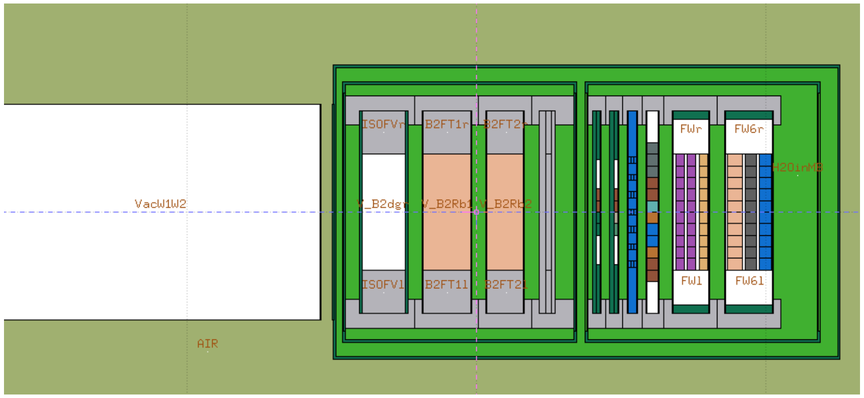
Material Irradiation Damage Studies for:

- Large Hadron Collider (CERN)
- Long Baseline Neutrino Experiment
- Neutrino Factory

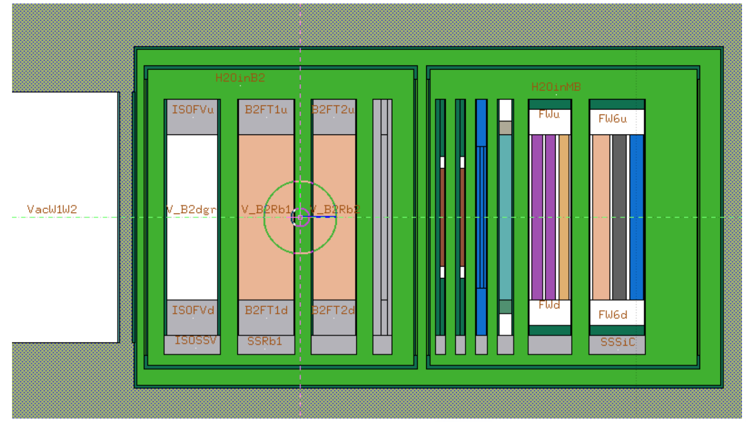




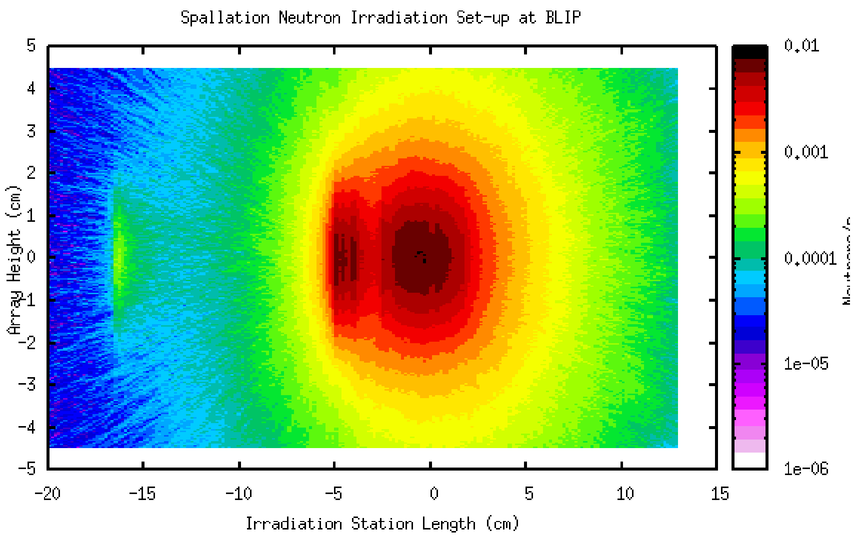




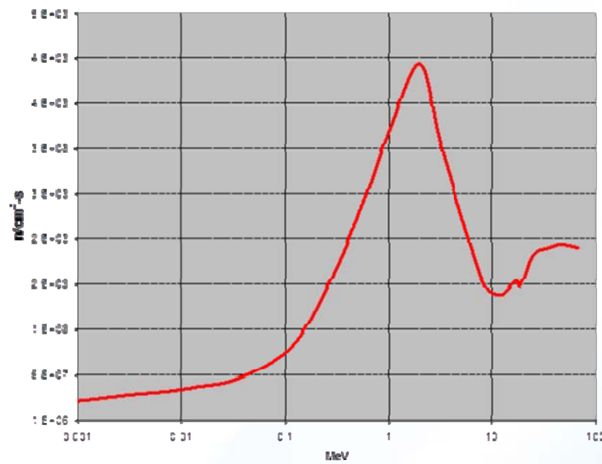
VacW1W2
AIR
ISOFVr
B2FT1r
B2FT2r
ISOFVl
B2FT1l
B2FT2l
V_B2dgr
V_B2Rb1
V_B2Rb2
FWr
FW6r
FWl
FW6l
H2OinMB



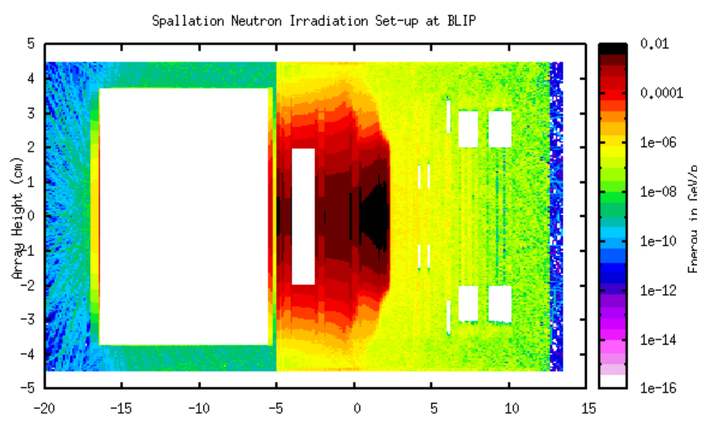
VacW1W2
H2OinB2
H2OinMB
ISOFVu
B2FT1u
B2FT2u
V_B2dgr
V_B2Rb1
V_B2Rb2
ISOFVd
B2FT1d
B2FT2d
ISOSSV
SSRb1
FWu
FW6u
FWd
FW6d
SSSiC



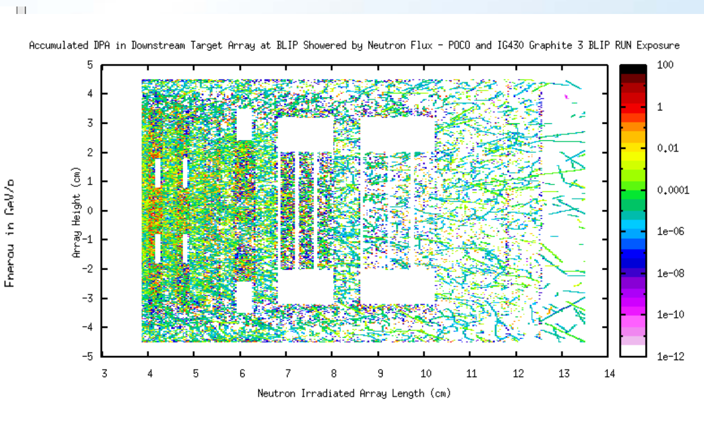
Spallation Neutron Irradiation Set-up at BLIP
Array Height (cm)
Irradiation Station Length (cm)
0.01
0.001
0.0001
1e-05
1e-06



MeV



Spallation Neutron Irradiation Set-up at BLIP
Array Height (cm)
0.01
0.0001
1e-06
1e-08
1e-10
1e-12
1e-14
1e-16



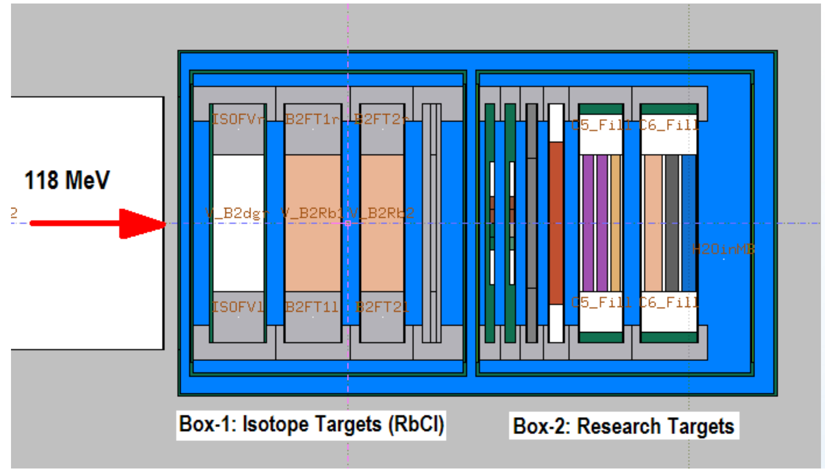
Accumulated DPA in Downstream Target Array at BLIP Showered by Neutron Flux - POCO and IG430 Graphite 3 BLIP RUN Exposure
Array Height (cm)
Neutron Irradiated Array Length (cm)
Accumulated DPA

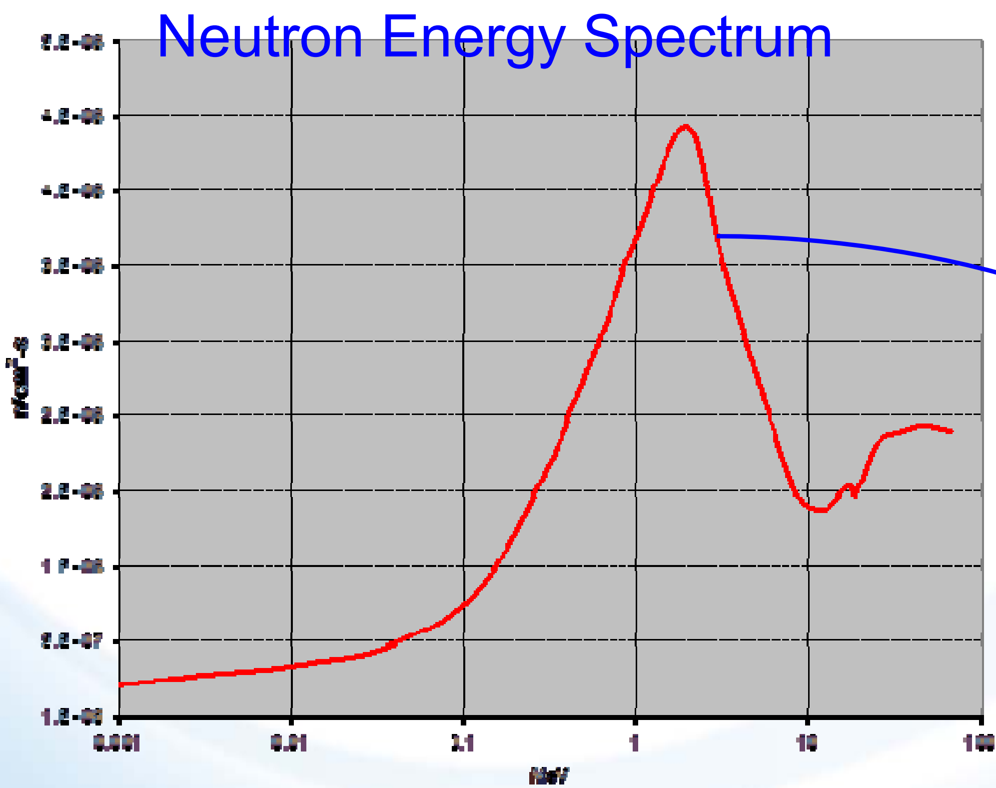
Spallation-induced Fast Neutron Irradiation at BLIP

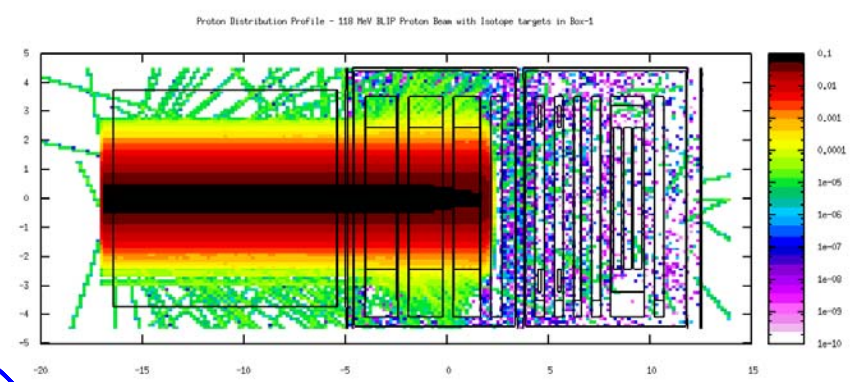
Irradiation damage studies from mixed spectrum (dominated by fast neutrons)

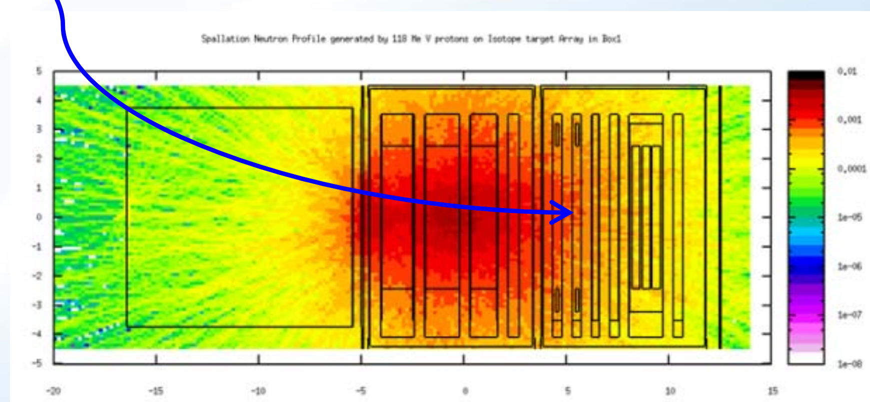
Studies:

- Fusion Reactor Materials and Composites
- DOE-NE materials (super-alloys, ceramic and amorphous coatings on reactor steels, etc.)

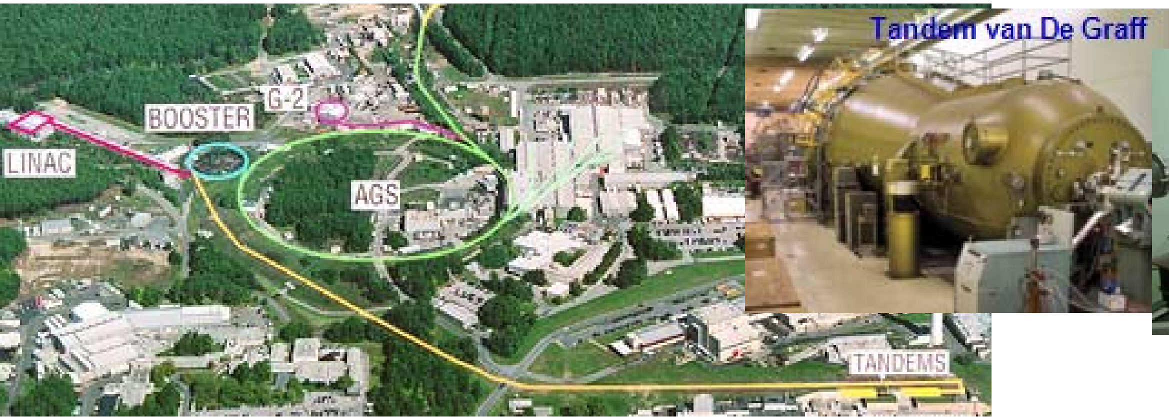




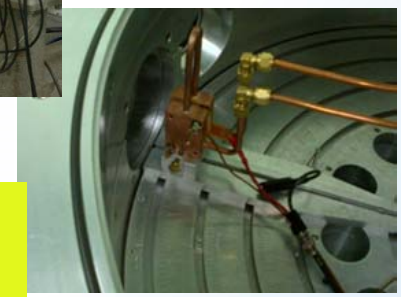




28 MeV Proton & Heavy ion irradiation at Tandem



Target Irradiation Beamline



IONS Available at Tandem

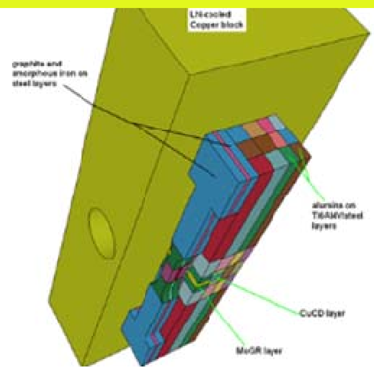
Flux can be in the range of 1 particle/cm^2/sec to greater than $1 \cdot 10^6$ particles/cm^2/sec.

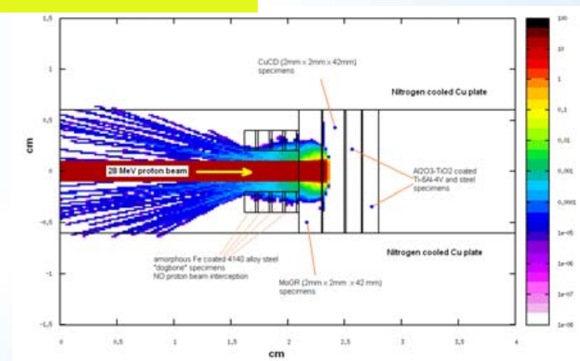
High LET Summary | Low LET Summary | High LET Summary | Low LET Summary

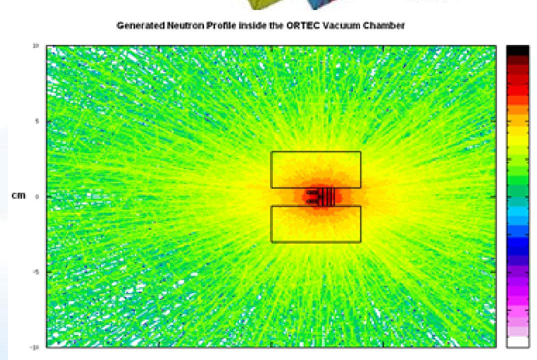
How To Use The Charts Below

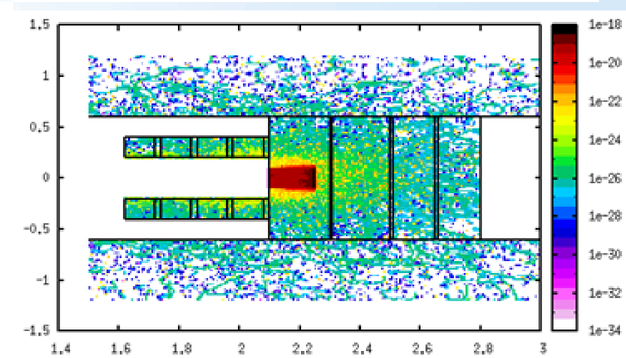
Z	Symbol	Mass AMU	Max Energy MeV	Max Energy MeV/AMU	Surface LET MeV mg/cm²	Range Microns	Surface LET MeV mg/cm²	Range Microns
1	^{1}H	1.0079	28.75	28.52	0.0153	4550	0.0118	2610
3	^{7}Li	7.0160	57.2	8.15	0.369	390	0.273	240
5	^{11}B	11.0093	85.5	7.77	1.08	206.13	0.754	132.55
6	^{12}C	12.0000	99.6	8.30	1.46	180.43	1.03	115.82
8	^{16}O	15.9994	128	8.00	2.61	137.78	1.83	88.9
9	^{19}F	18.9954	142	7.48	3.51	118.88	2.45	77.12
12	^{24}Mg	23.9927	161	6.71	6.01	84.16	4.17	55.13
14	^{28}Si	28.0855	187	6.66	7.81	77.16	5.42	50.66
17	^{35}Cl	34.9688	212	6.06	11.5	64.41	7.93	42.71
20	^{40}Ca	39.9753	221	5.53	15.8	51.89	10.9	34.7
22	^{48}Ti	47.9479	232	4.84	19.6	47.8	13.4	32.36
24	^{52}Cr	51.9405	245	4.72	22.3	45.86	15.3	31.06
26	^{56}Fe	55.9349	259	4.63	25.1	44.24	17.2	30.09
28	^{58}Ni	57.9353	270	4.66	27.9	44.56	19.1	30.47
29	^{63}Cu	62.9296	277	4.40	30.1	42.06	20.6	28.79
32	^{72}Ge	71.9221	273	3.80	35.9	37.94	24.4	26.25
35	^{81}Br	80.9163	287	3.55	41.3	37.50	28.0	26.11
41	^{93}Nb	92.9060	300	3.23	47.5	36.32	32.1	25.4
47	^{107}Ag	106.9051	313	2.93	59.2	32.48	39.9	22.89
53	^{127}I	126.9045	322	2.54	66.9	32.54	45.0	23.17
79	^{197}Au	196.9665	337	1.71	84.6	29.21	56.2	21.18

Recent 28 MeV proton + spallation neutron irradiation experiment at sub-zero temperatures at Tandem (Simos, et al)



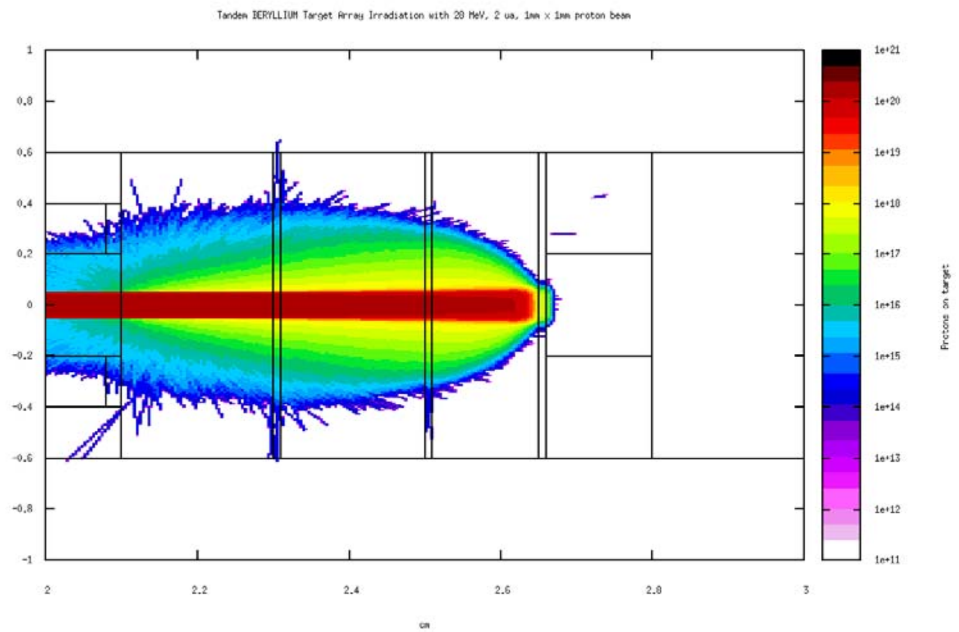


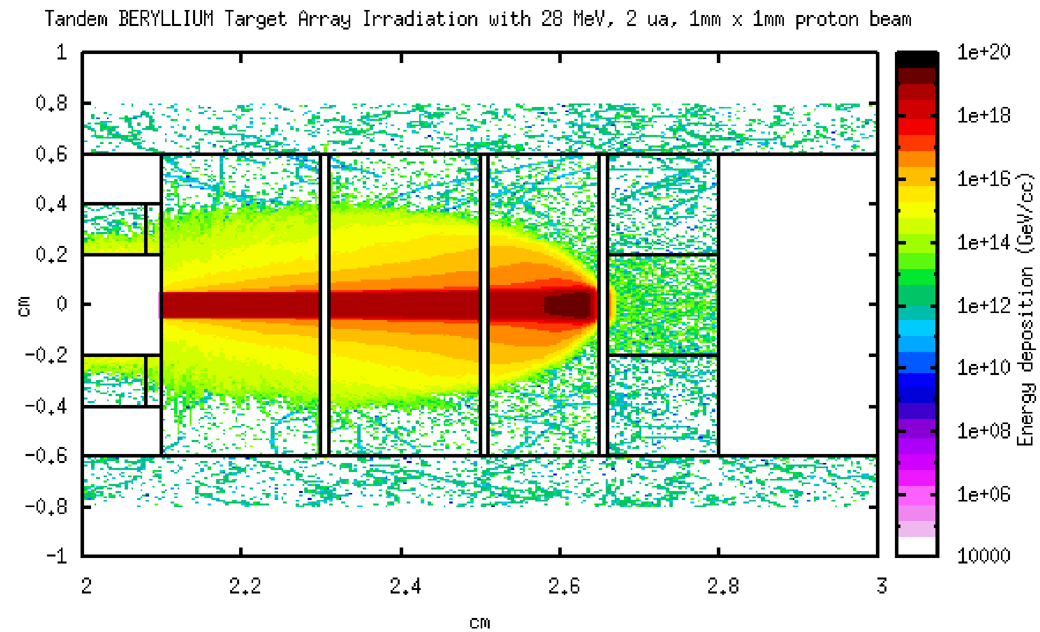


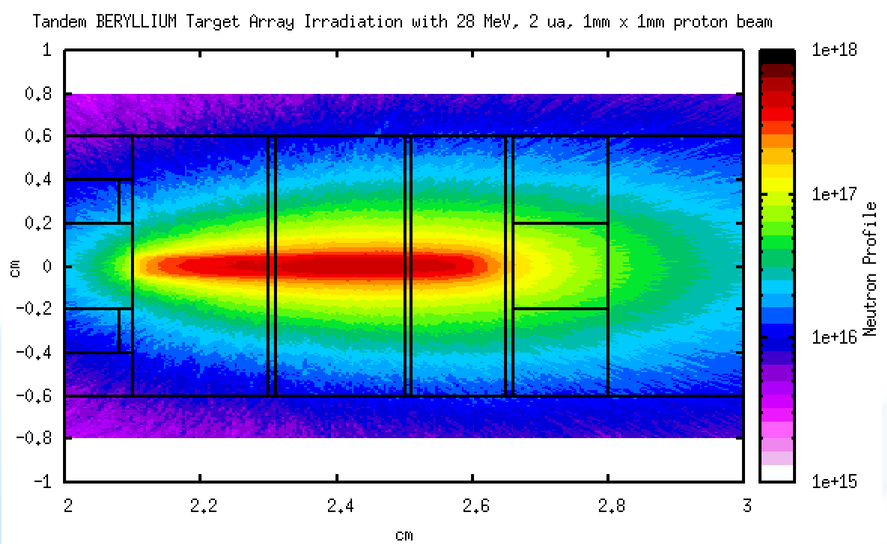


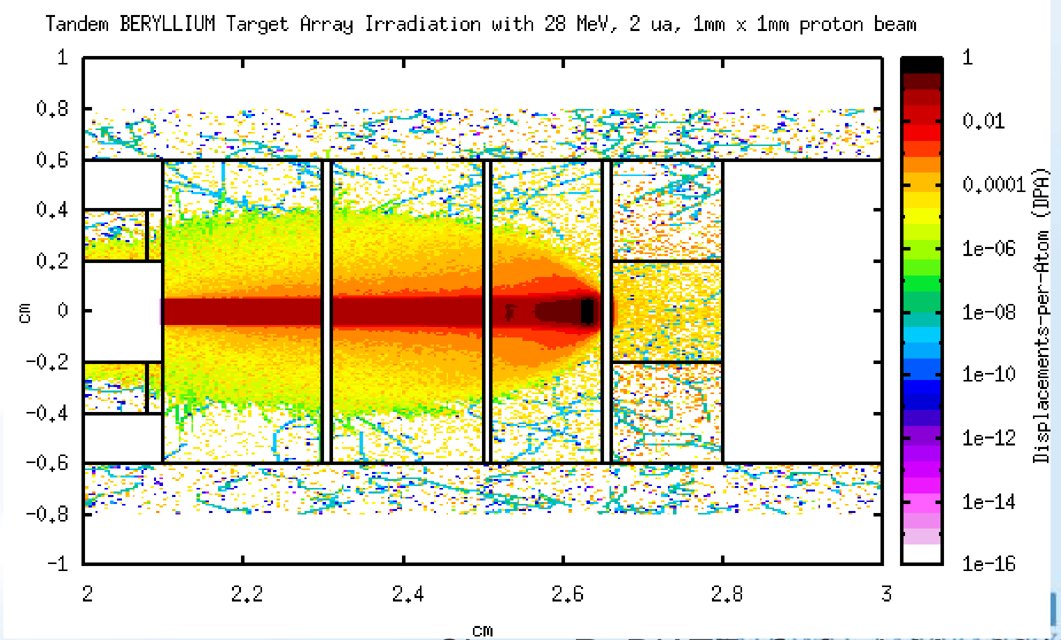
What Damage Can One Achieve at Tandem?

28 MeV protons on BERYLLIUM target array









^{56}Fe ion on Be target Array