Radiation Damage and Radio-chemistry Issues

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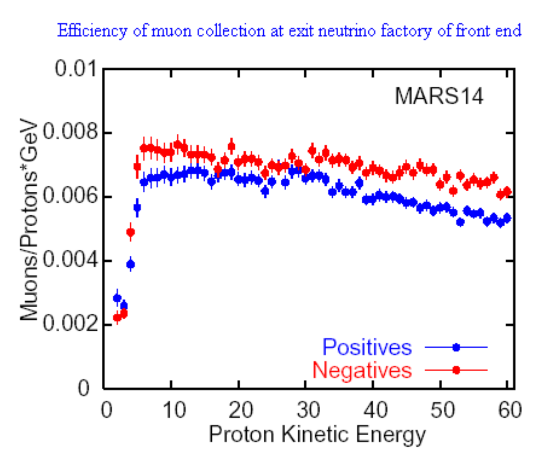
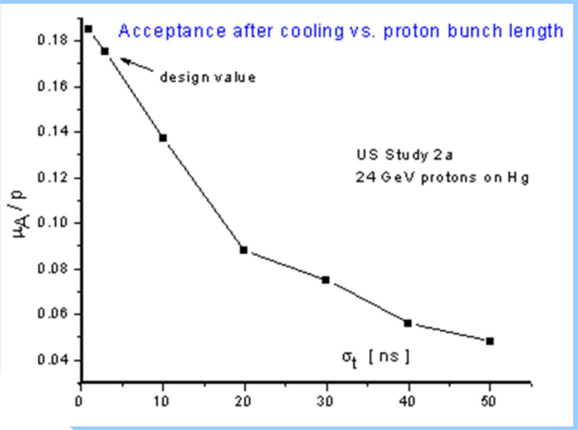
Parameter Space (damage is missing !!!)

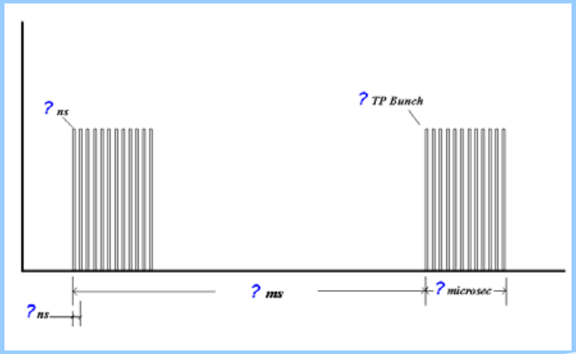
Protons per pulse required for 4 MW

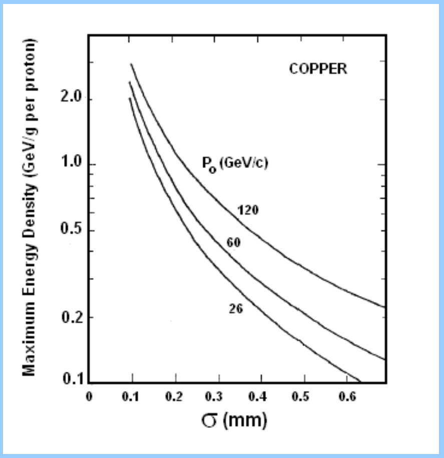
$$\bar{P}_{arc}(w) = E[eV] \times N \times e \times f_{rep}\,[Hz]$$

	10 Hz	25 Hz	50 Hz
10 GeV	250×10^{12}	100×10^{12}	50×10^{12}
20 GeV	125×10^{12}	50×10^{12}	25×10^{12}









Motivation:

Need for multi-MW level operations and beams associated with proposed long baseline neutrino experiments (i.e. LBNE)

Yield of useful pions/parents of the neutrinos of interest from low-Z materials is well matched to requirements of most neutrino experiments

Peak energy deposition in low-Z targets is lowest;

Graphite shown to exhibit superior thermo-mechanical performance confirmed by experience data from nuclear reactors

Identification of limitations (if any) of low-Z materials under intense proton beams in support of MW-level experiments

Background – Irradiation Damage & Accelerators

Extrapolation of nuclear reactor experience data on radiation damage extremely risky

- **Elastic collisions** (transferring of recoil energy to a lattice atom) leading to displaced atoms (dpa)
- **Inelastic collisions** ➔ transmutation products (H, He)

Series of experiments at BNL using the 200 MeV Linac on materials, some with excellent performance in reactors (tens of dpa damage) showed presence of proton fluence threshold (~10^21 p/cm2)

- (LHC beam collimation studies at BNL first revealed shortcomings of carbon based materials)

...... realization ⬇

For multi-MW level experiments, i.e. LBNE, apparent threshold **COULD** be a limiting factor

Threshold is within year of operation at LBNE

Radiation effects on materials

- Understand effects of irradiating species
- Energy
- Rate
- Operating Environment

The Radiation Damage Problem Simplified

- Material properties change significantly as they are irradiated due to microstructural disorder
- Design limits vary with exposure
- In general:
  - Strength increases
  - Ductility decreases
  - Thermal/Electrical conductivity decreases
- Not only targets, but also windows, collimators, instrumentation, etc…
- Data for LE neutron irradiation is relatively plentiful, not so for HE proton irradiation

The Radiation Damage Problem - Complicated

- Critical Mat'l properties sensitive to radiation damage include (note difference between LE neutron and HE proton):

- Irradiation particle, e.g. protons vs. neutrons
- Particle energy
- Flux or dose rate (dpa/s)
- Fluence or dose (dpa)
- Irradiation temperature
- Transmutation (e.g. He, H)
- Pulsed irradiation vs. continuous irradiation

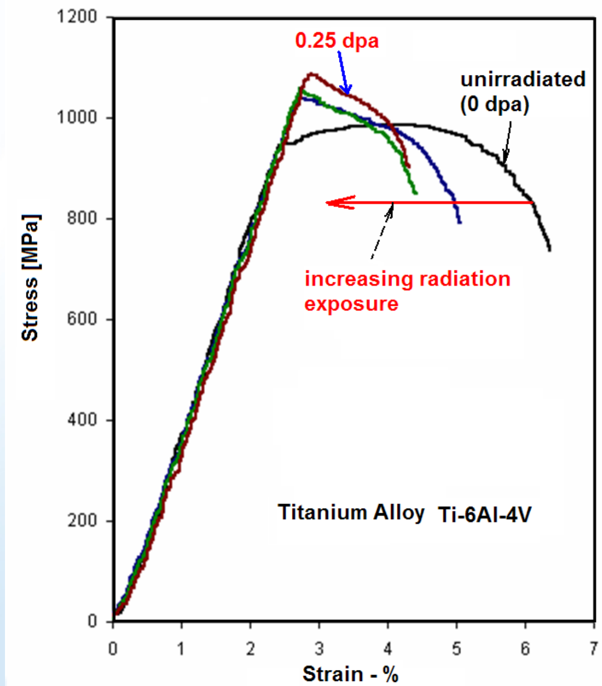
- These constitute "damage correlation parameters"
- Use these parameters to correlate damage from LE neutron to HE proton irradiation?
  - Could be powerful to harness the data from reactors for use in proton accelerator target facility designs

The Radiation Damage Problem Complicated

- Material property changes are sensitive to many parameters (not captured by DPA):

- Tensile and yield strengths
- Modulus of Elasticity
- Coef. of Thermal Expansion
- Heat Capacity
- Electrical and Thermal Conductivity
- Density/Dimensions (Swelling)
- Fracture Toughness
- Fatigue Strength
- Irradiation creep
- Hydrogen/Helium Embrittlement
- Sonic velocity
- Corrosion resistance

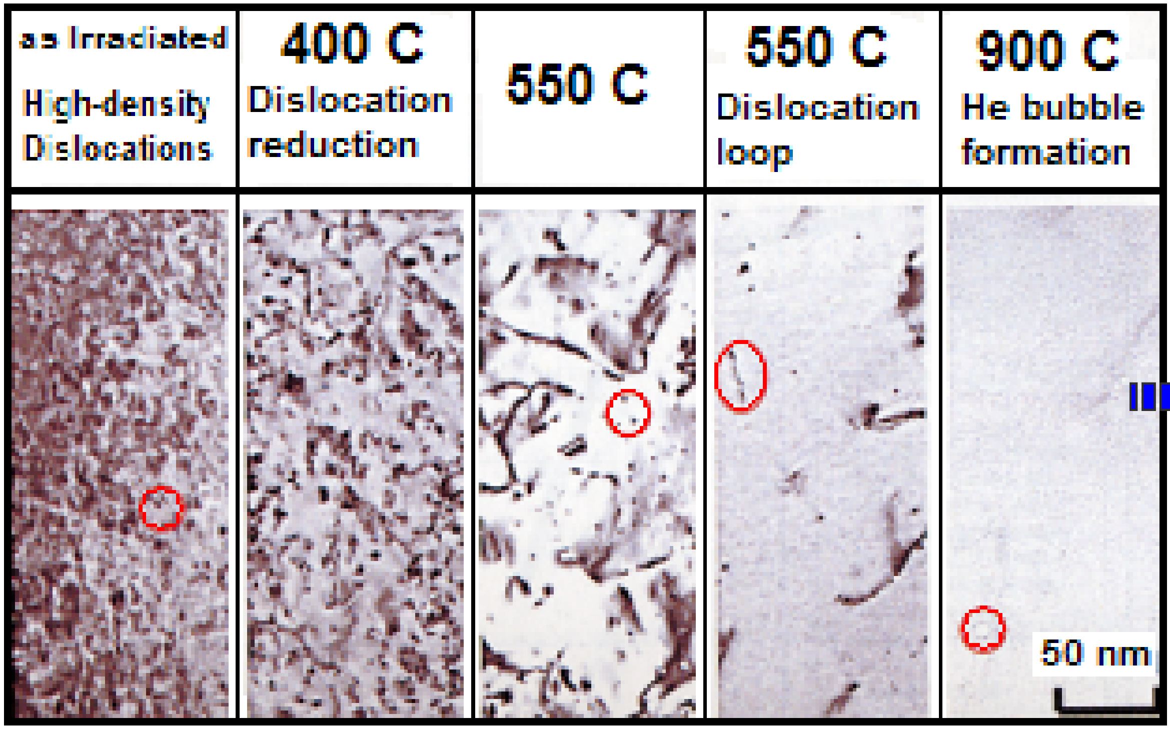
- "Threshold" of significant property change is material dependent
  - 0.02 DPA for graphite
  - 1-10 DPA for stainless steel



Irradiation damage & Reversal

➔ how does it manifest its self both in micro- and macro-scales?
➔ irreversible ?

Annealing or defect mobility at elevated temperature



Y. Ishiyama et al., J. Nucl. Mtrl. 239, 1996

Observed Behavior:
Increased defect density, damage acceleration upon re-exposure

Why and what is the responsible mechanism?

Does it apply with all alloys?

Radiation Accelerated Corrosion

- Ionization of air surrounding a target by primary and secondary particles can create a very aggressive, corrosive environment
- High strength steel may suffer hydrogen embrittlement (MiniBooNE, NuMI)
- Coupled with radiation damage of material, not only accelerates corrosion, but changes the nature of the corrosion morphology (localized pitting versus uniform layer; NuMI decay pipe window)

Radiation Accelerated Corrosion

- Al 6061 samples displayed significant localized corrosion after 3,600 Mrad exposure
- NuMI target chase air handling condensate with pH of 2
- NuMI decay pipe window concerns

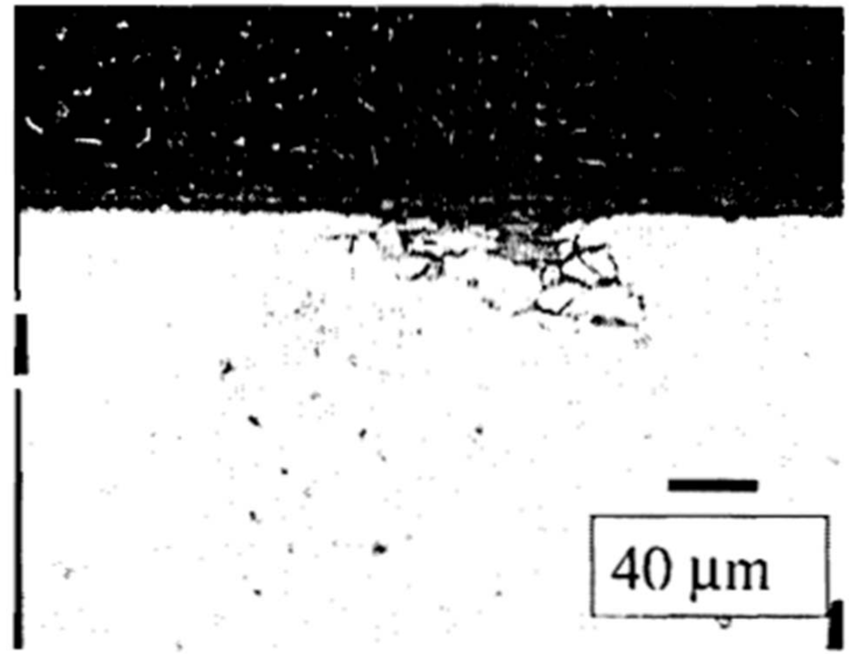
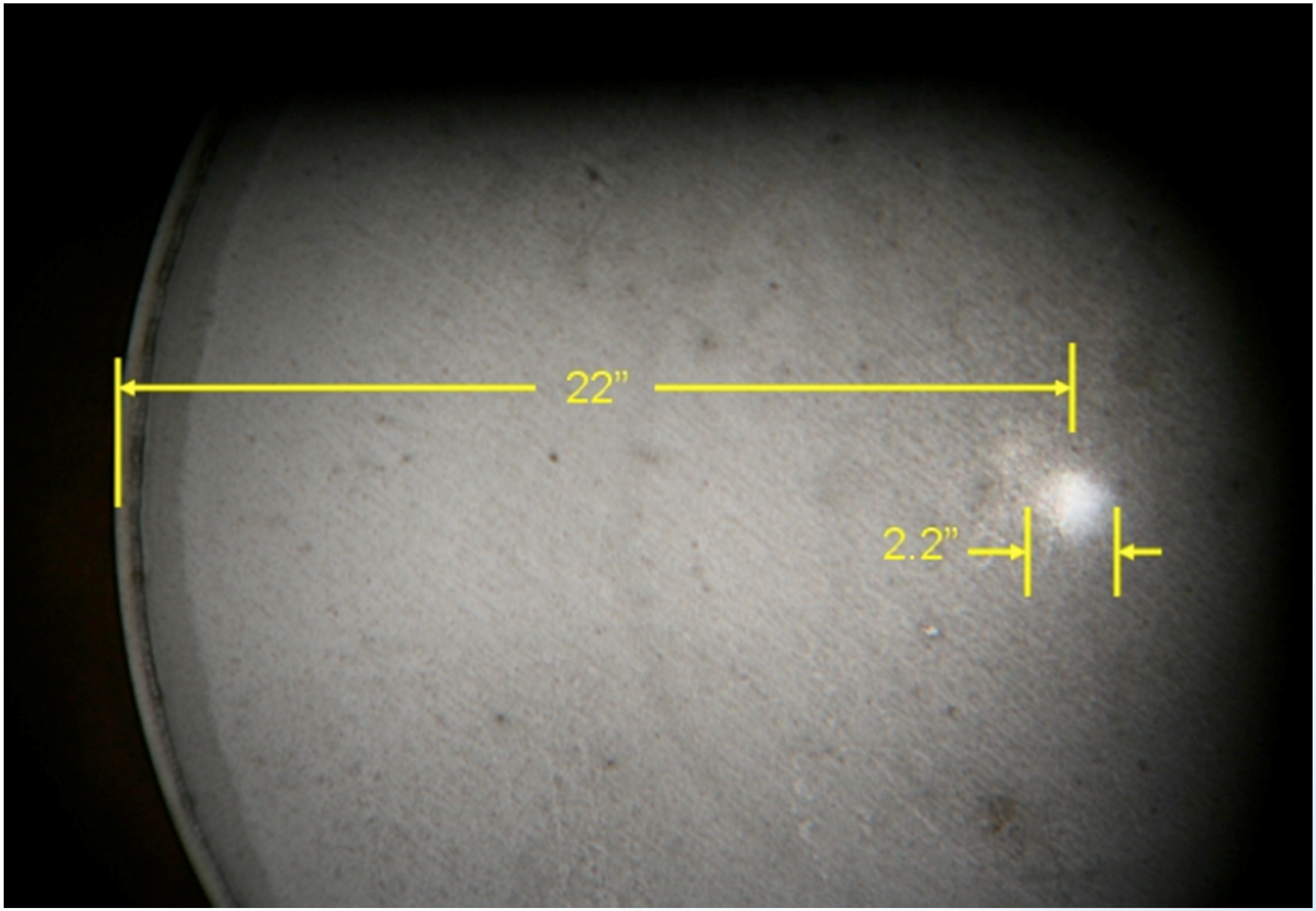


FIG. 8. Localized corrosion on 6061 Al sample exposed 12 weeks to saturated water vapor at 200°C and gamma irradiation.

R.L. Sindelar, et al., *Materials Characterization* 43:147-157 (1999).

Radiation Accelerated Corrosion

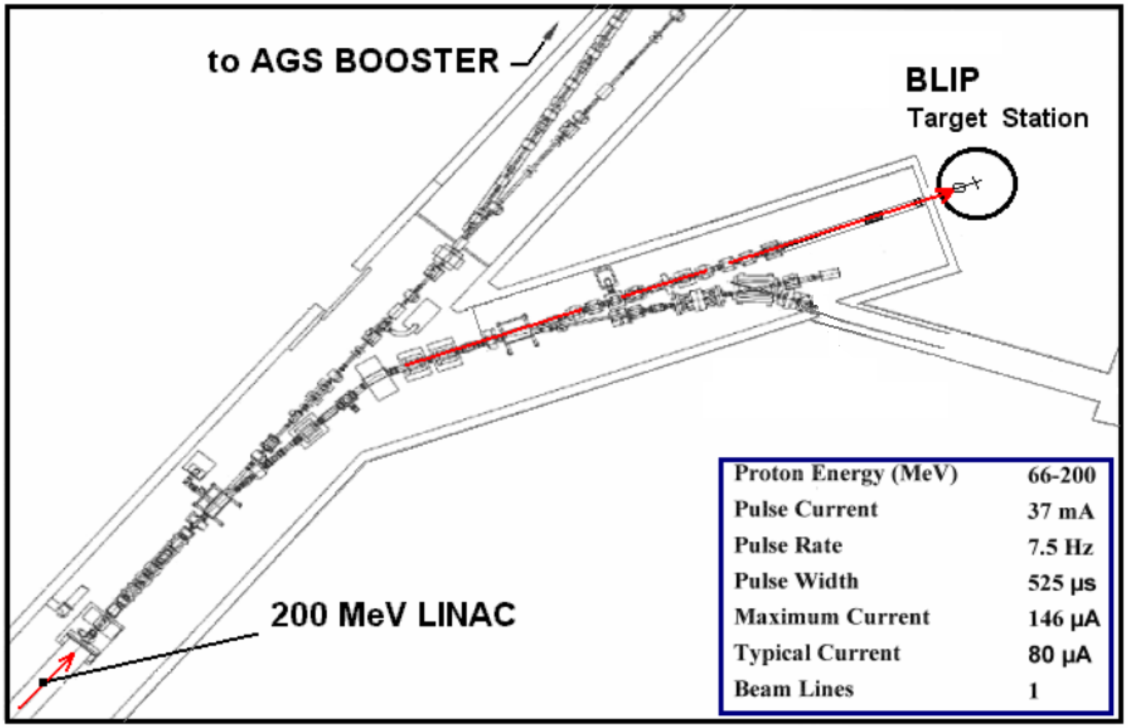


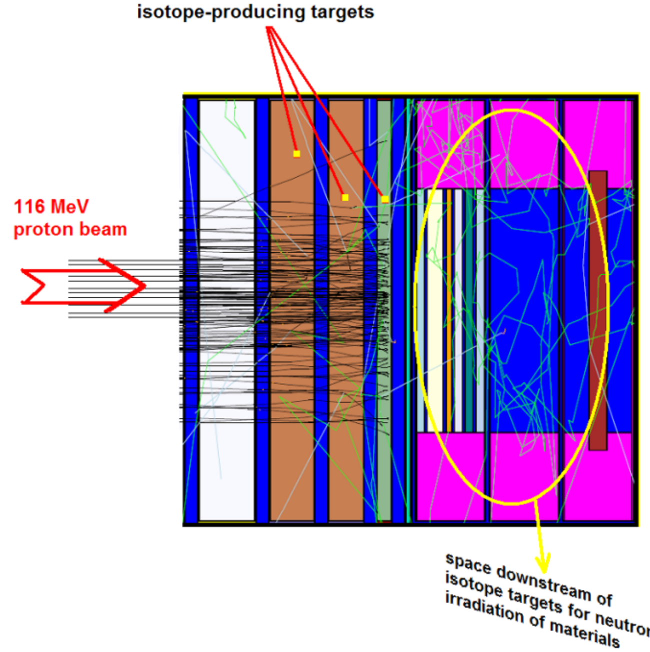
- Photograph of NuMI decay pipe US window showing corroded spot corresponding to beam spot

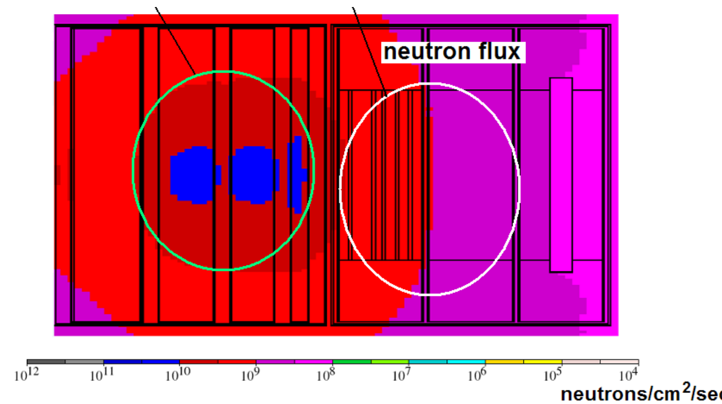
Any High Power Integrated Target System will be subjected to all that

Use of BNL Accelerator Complex for Material Damage

- Direct protons (130-200 MeV) on materials
- Spallation neutrons, gammas, electrons and secondary protons (isotope targets serving as spallation targets – 2011 beam run)





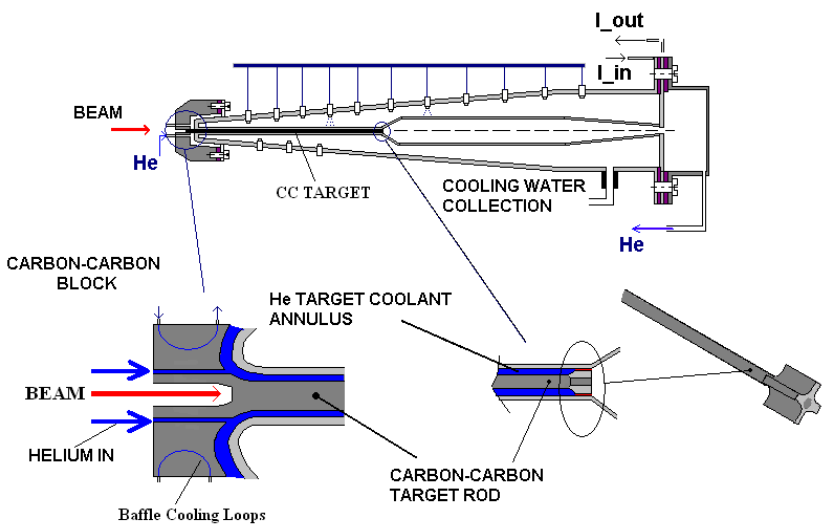


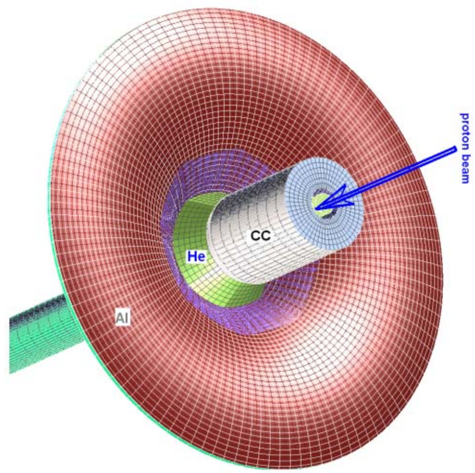
NORMALIZED neutron flux
at BLIP target station (by N. Mokhov, FNAL)

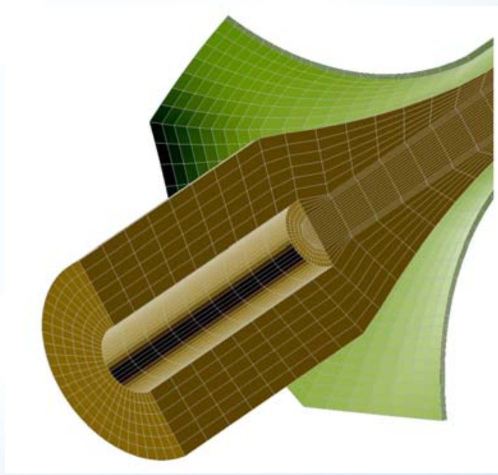
Why radiation damage at BLIP?

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

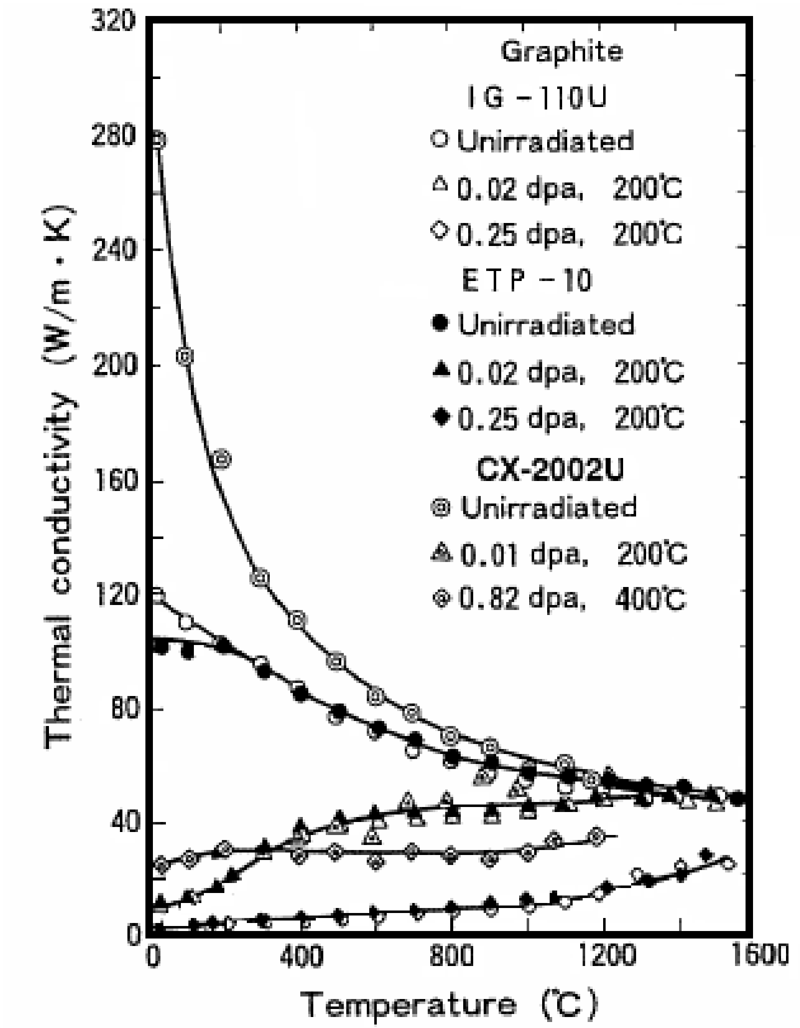
Graphite & Carbon Composites



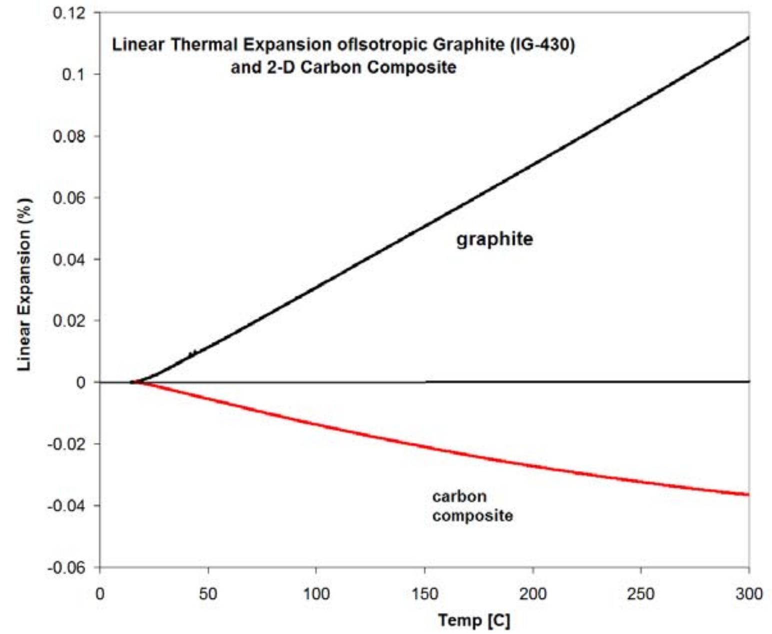


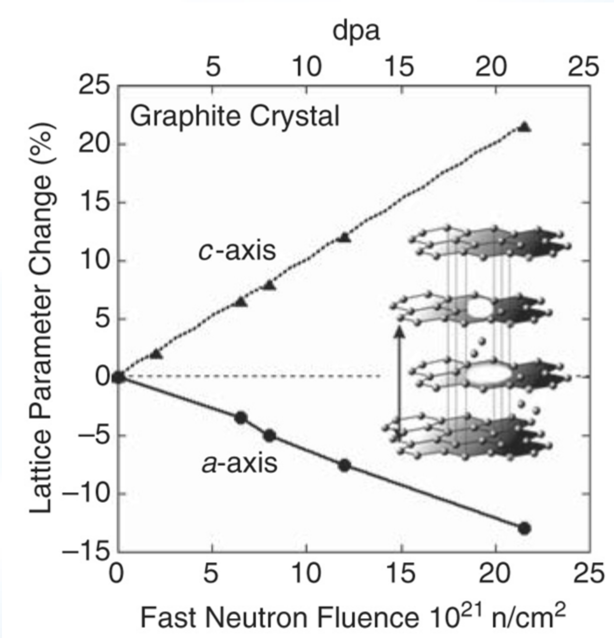


Graphite and C-C

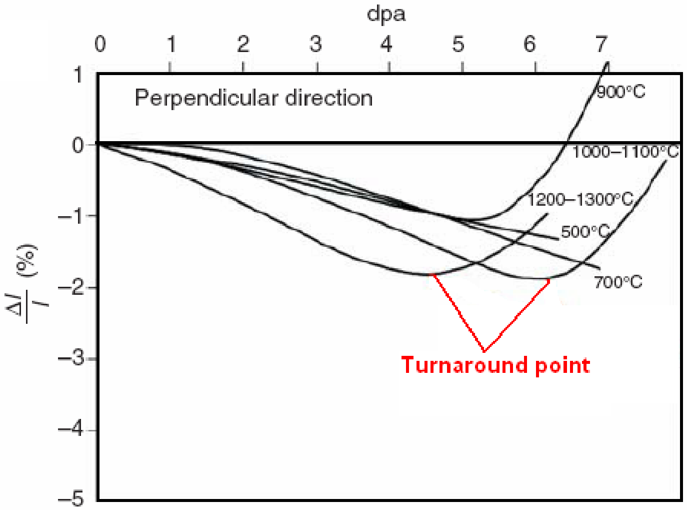


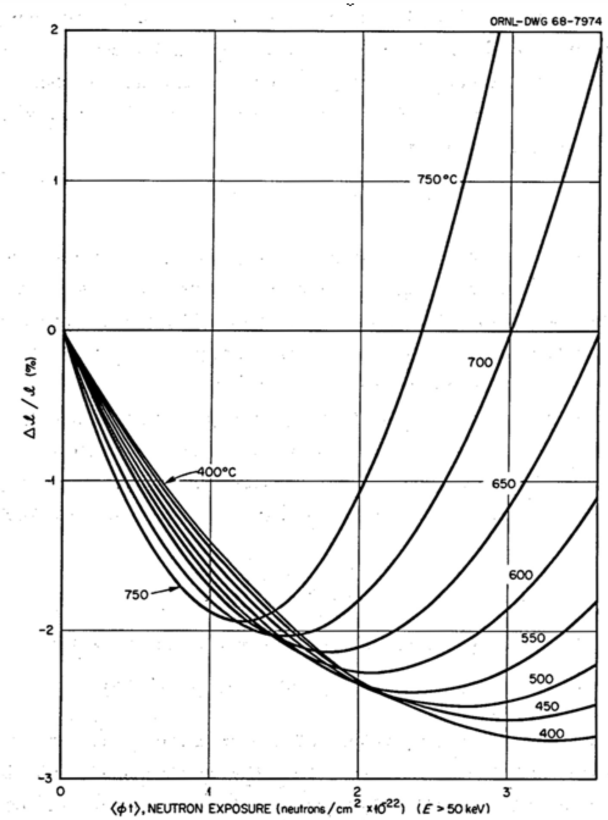
(Maruyama and Harayama 1992)

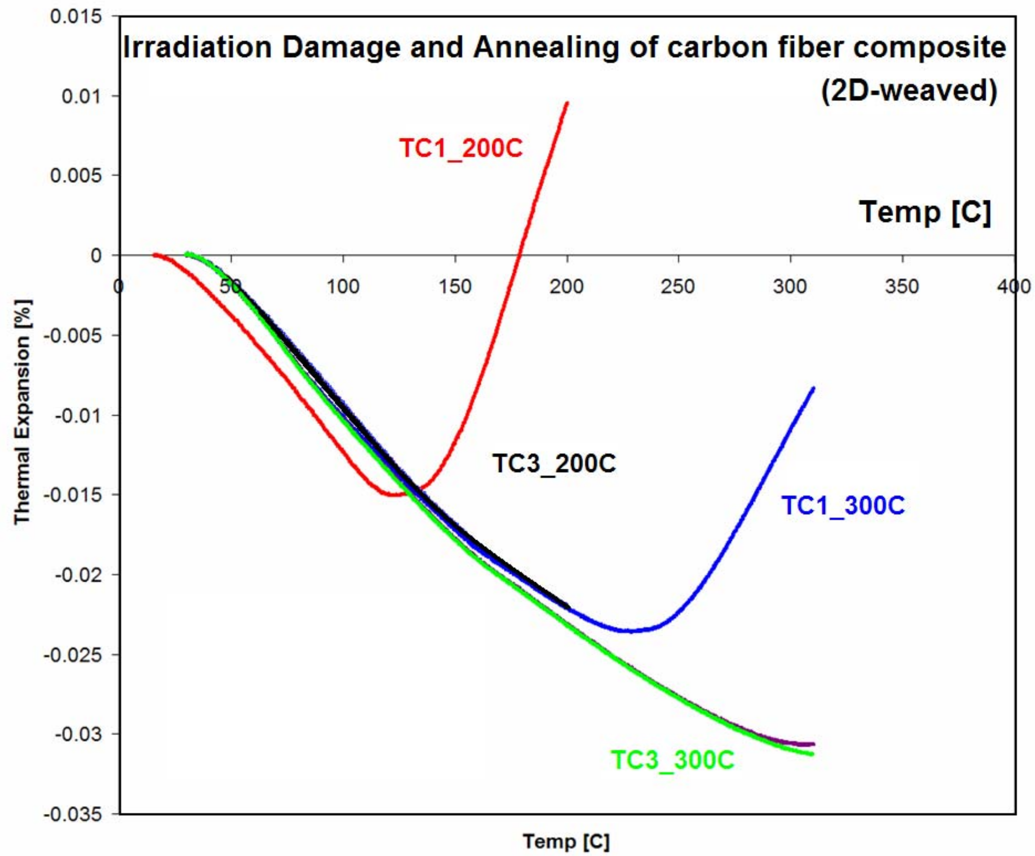




(Snead 2009)





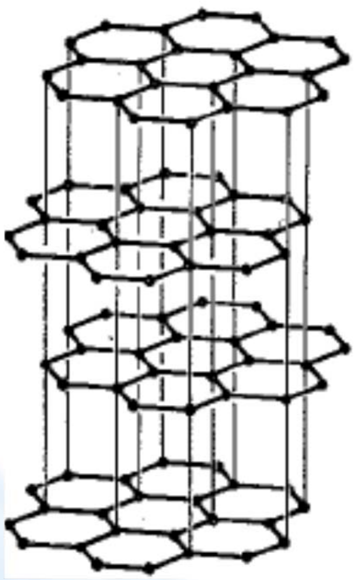


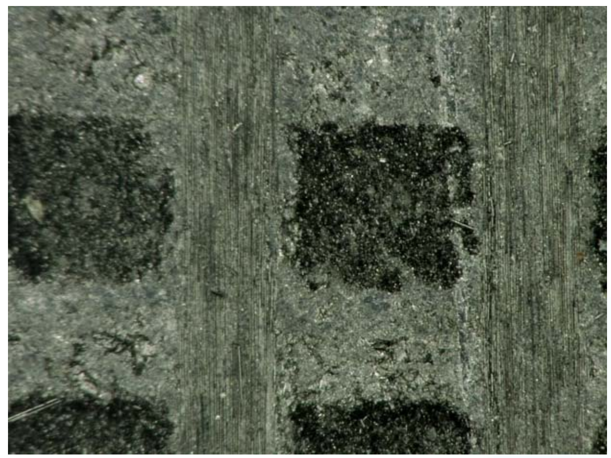
Neutron irradiation of polygranular graphite (shrinkage at low doses and expansion at high doses.(Kasten et al 1969).

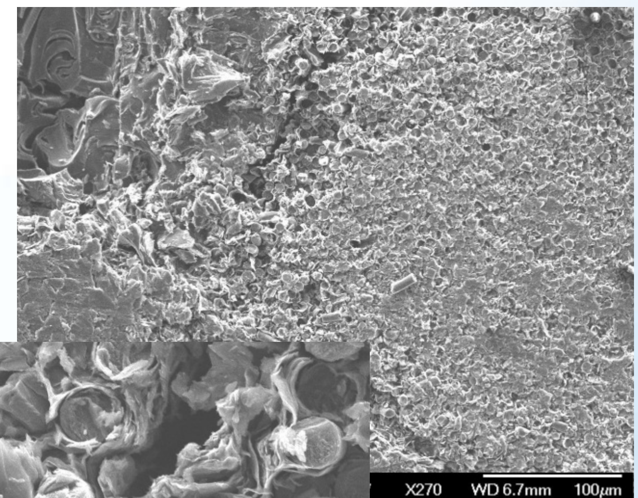
Low-Z Targets and LBNE

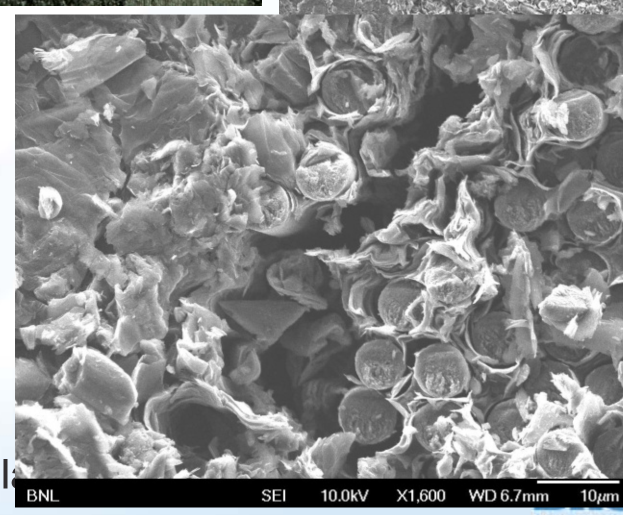
Graphite and Carbon-fiber composites

Graphite widely studied and used in reactors
Its behavior, however, remains elusive



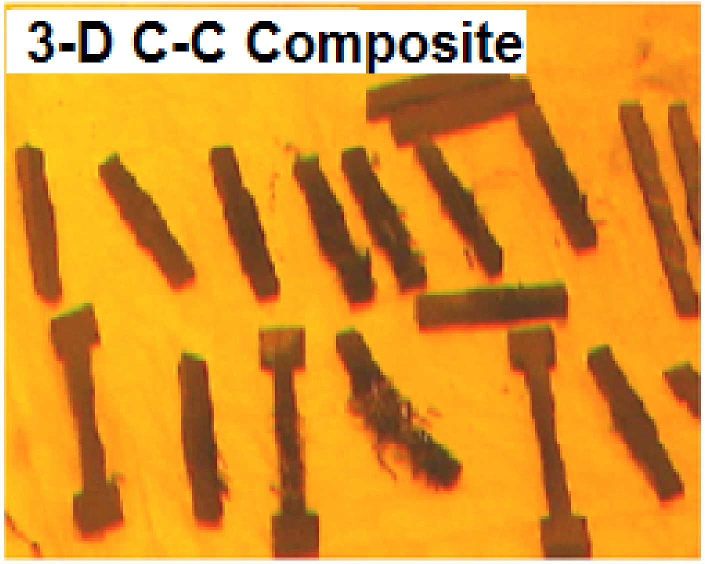


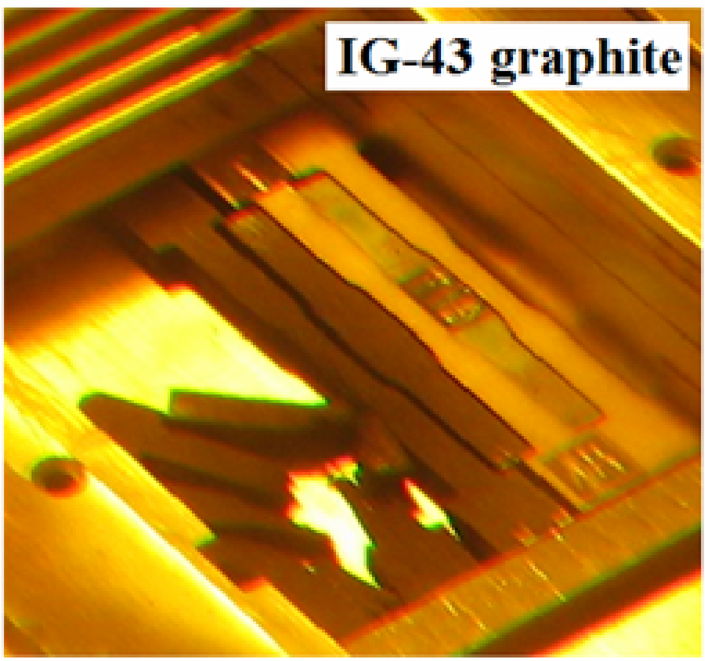


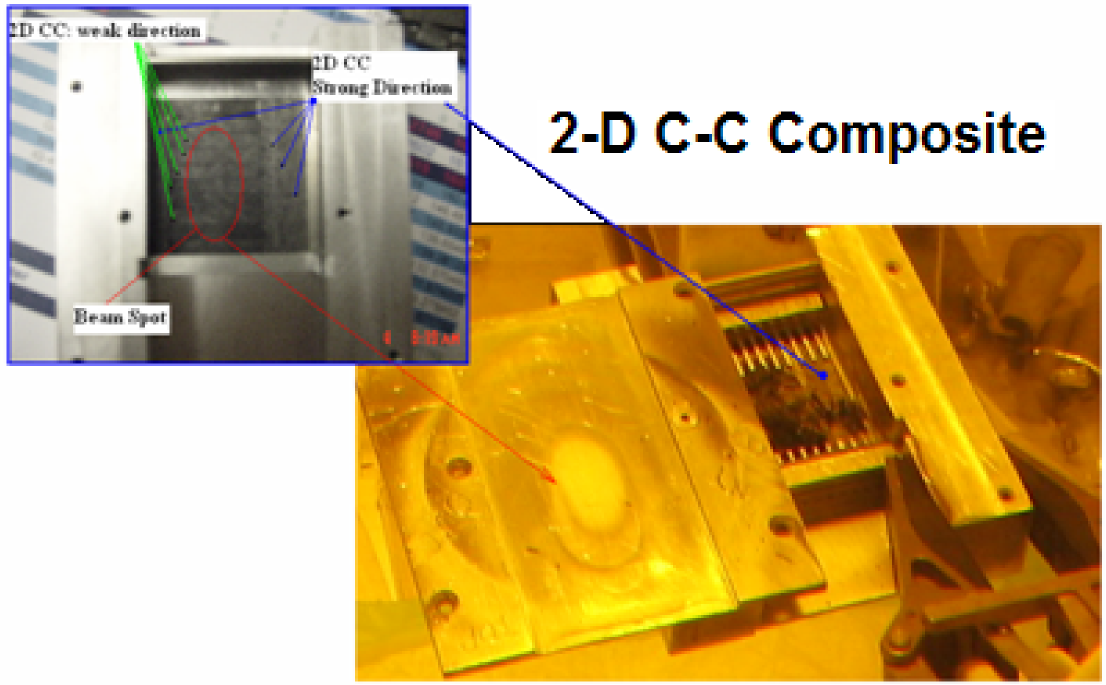


10^{21} p/cm^2 fluence → 0.2 dpa

what happened to the 10s of dpa seen in thermal reactors?

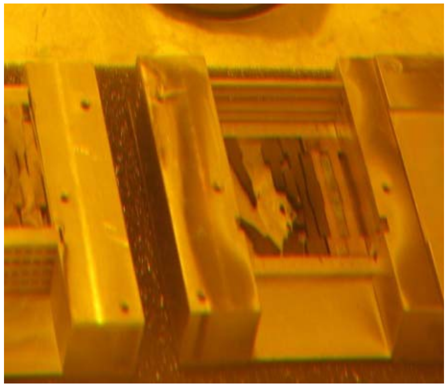


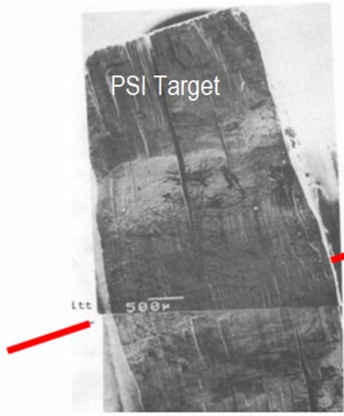




Observations were reproduced 3 times !!

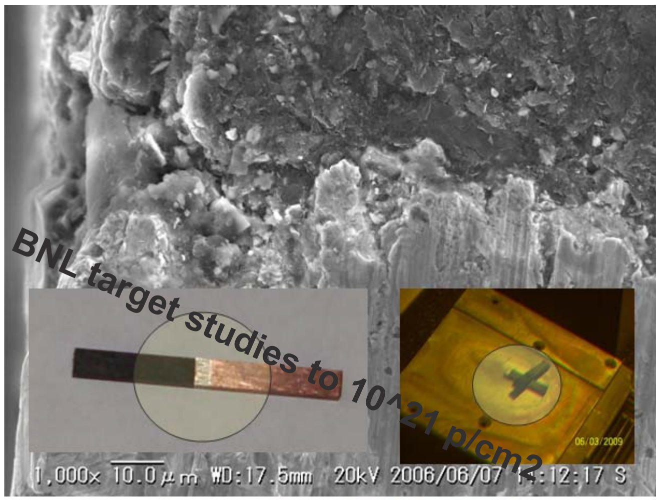
Graphite and carbon composites under high proton fluences

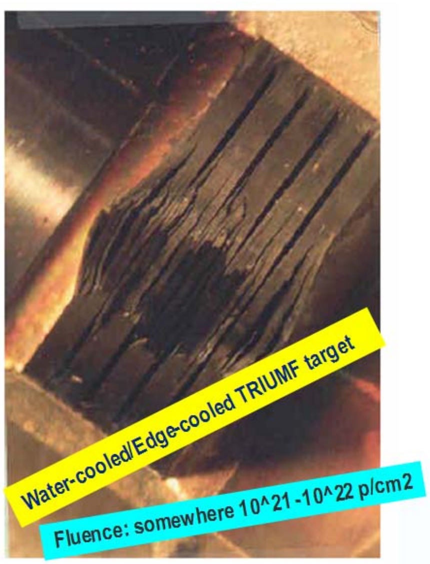


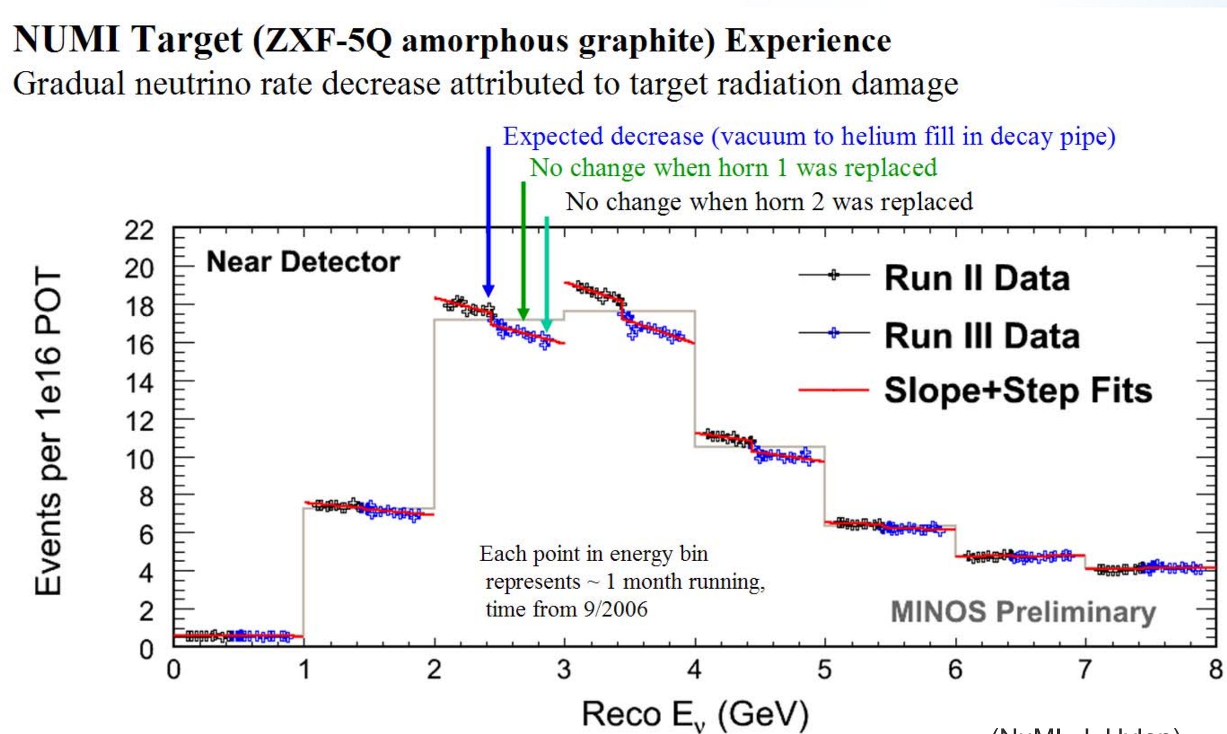


Swelling of the target after irradiation

10^{22} p/cm2







(NuMI, J. Hylen)

LBNE Target Experiment at BNL Linear Isotope Producer (BLIP)

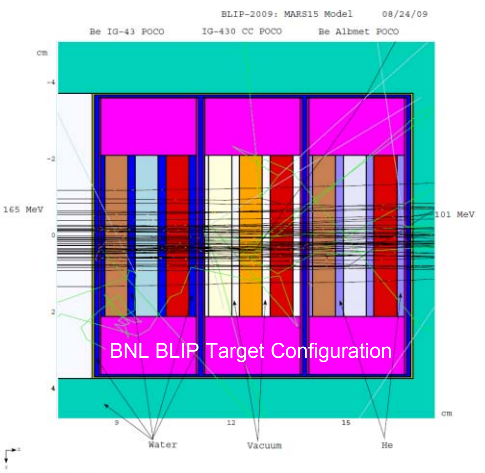
Experiment Objectives:

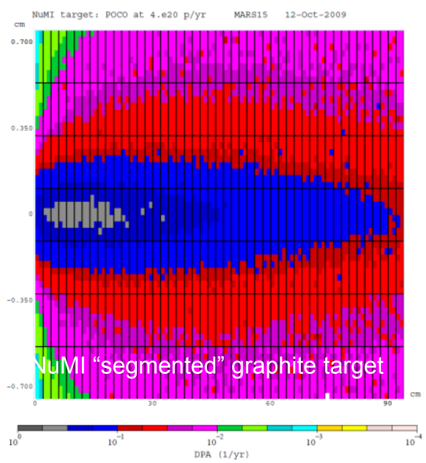
- Establish relation between limiting fluence threshold and operating environment
- Confirm damage & energy dependence (120 GeV vs. 180 MeV)
- Identify the optimal low-Z microstructure when exposed to similar fluence/temp/environment conditions
- Select the baseline target from material performance during LBNE test

MARS Analysis

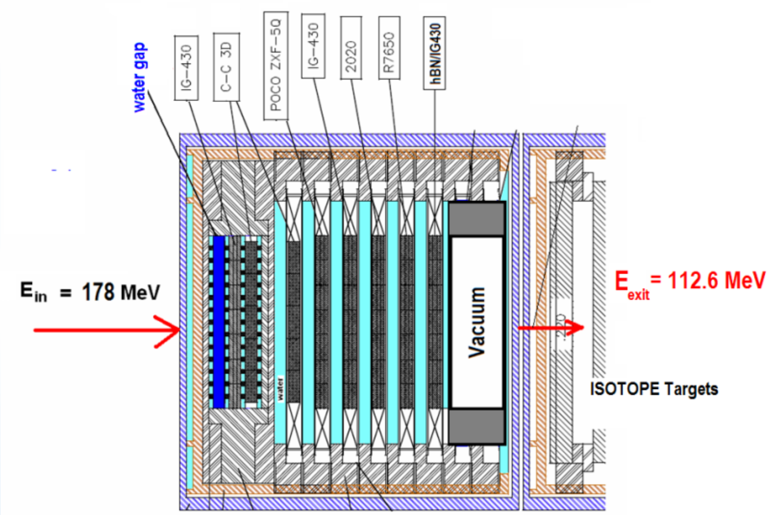
Material damage dependency on proton energy and irradiation rate

LBNE/NuMI & BNL BLIP Equivalence





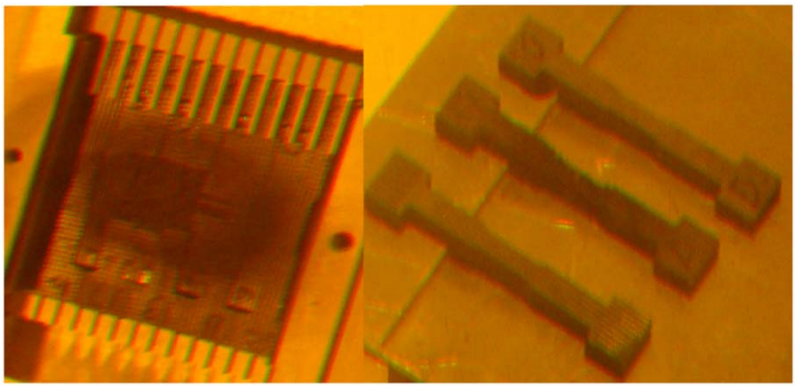
LBNE Target Array

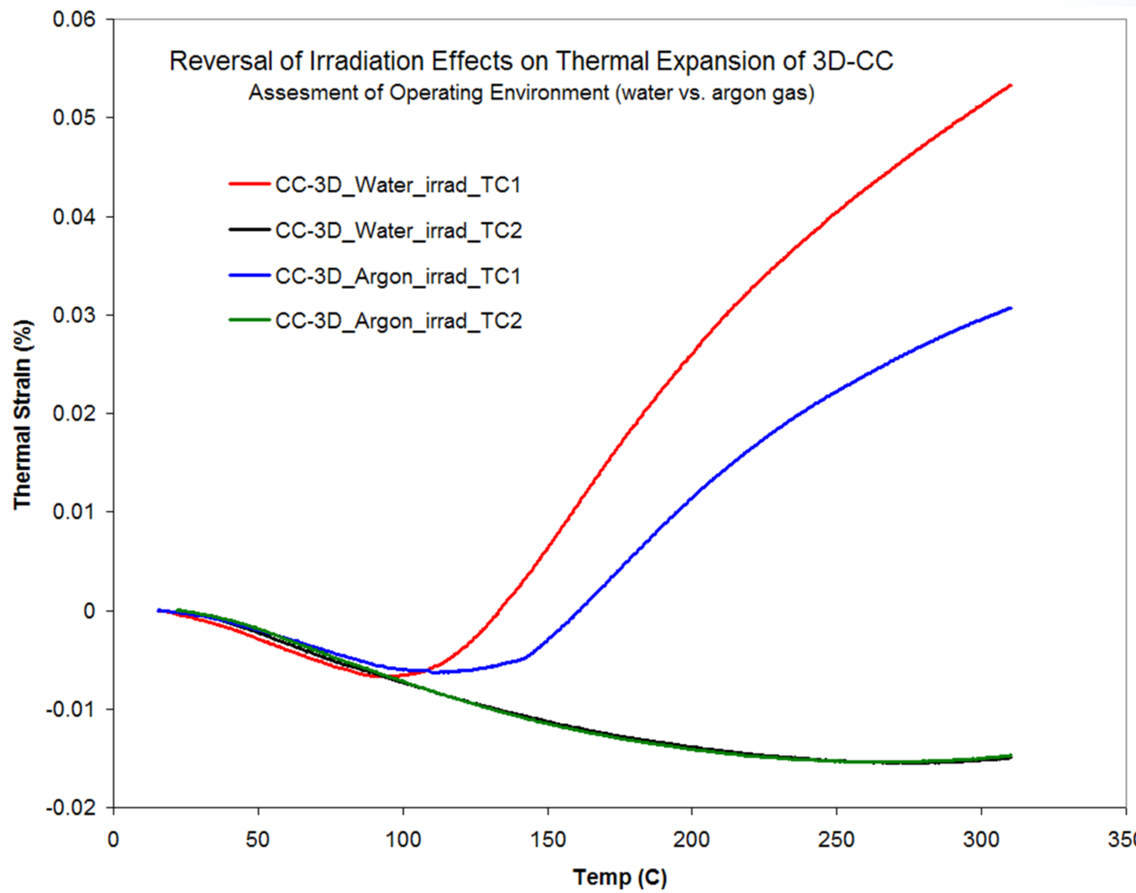


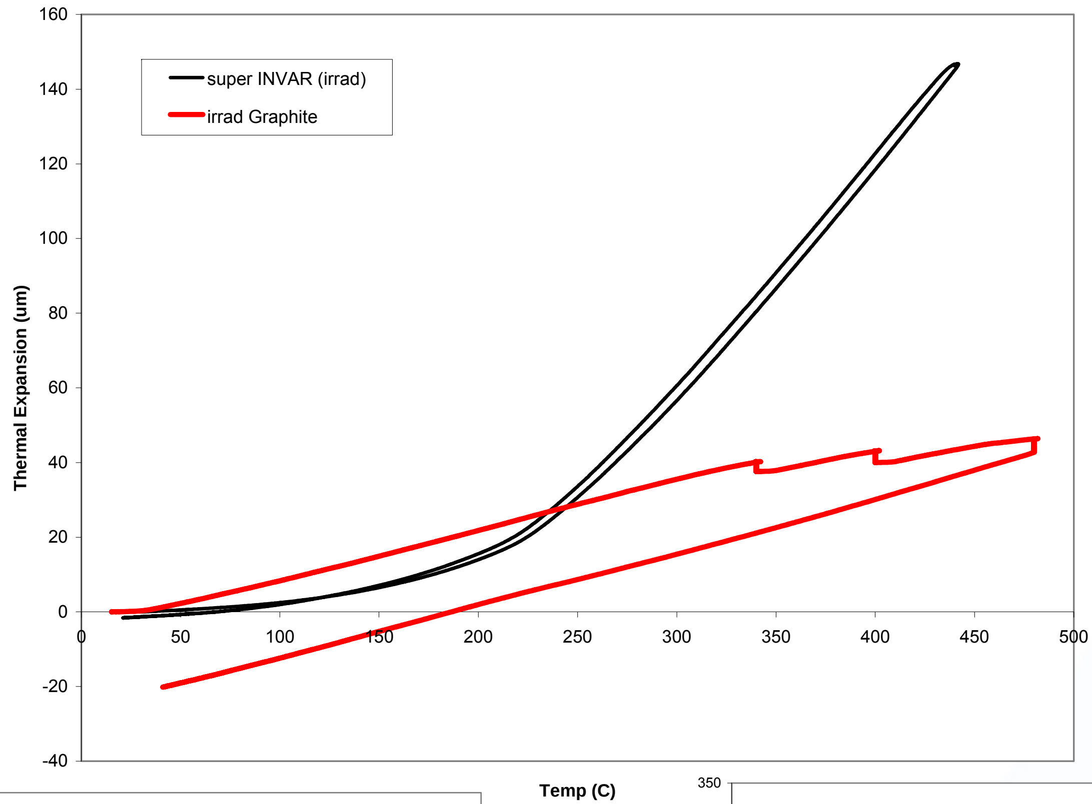
Target	E_p (GeV)	Beam σ (mm)	N_p (1/yr)	DPA (1/yr)
NuMI/LBNE	120	1.1	4.0e20	0.45
BLIP	0.165	4.23	1.124e22	1.5

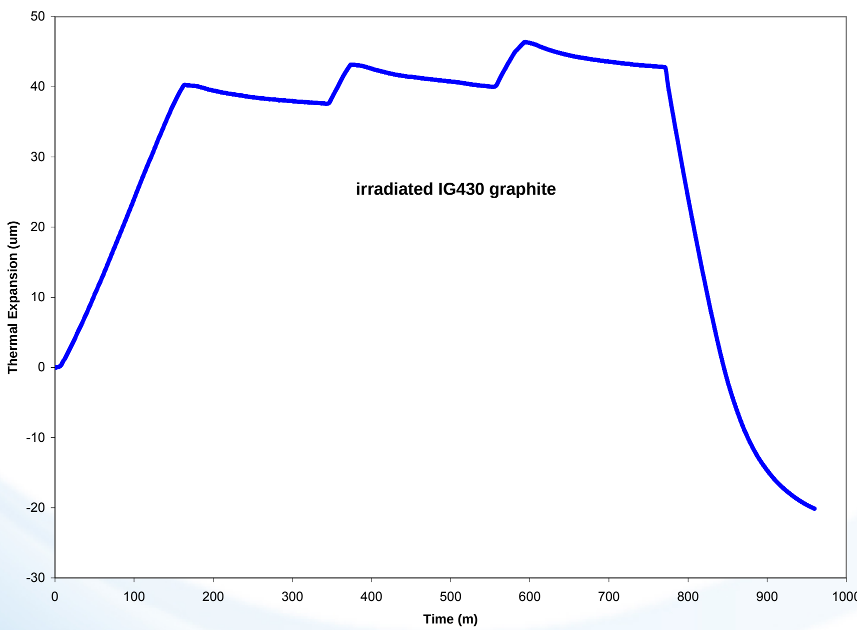
0.7-MW LBNE can be achieved at BNL BLIP over ~7 weeks

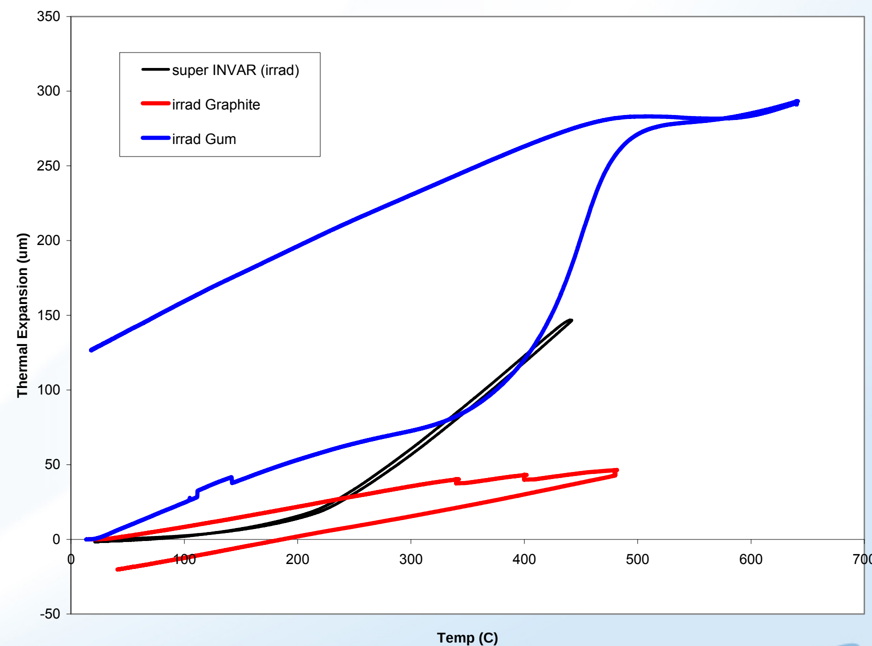
What have LBNE Radiation Damage Experiments have shown:

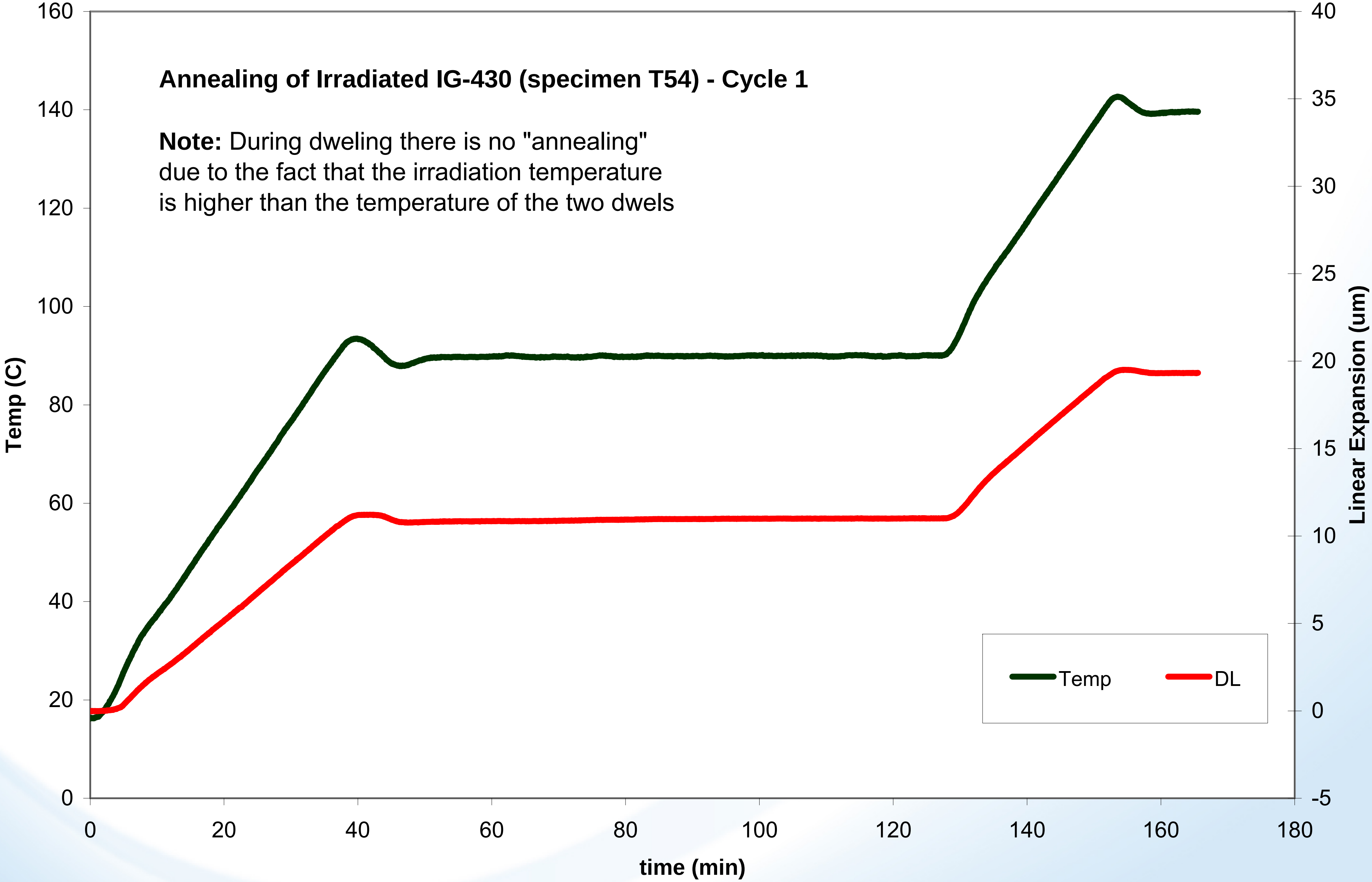


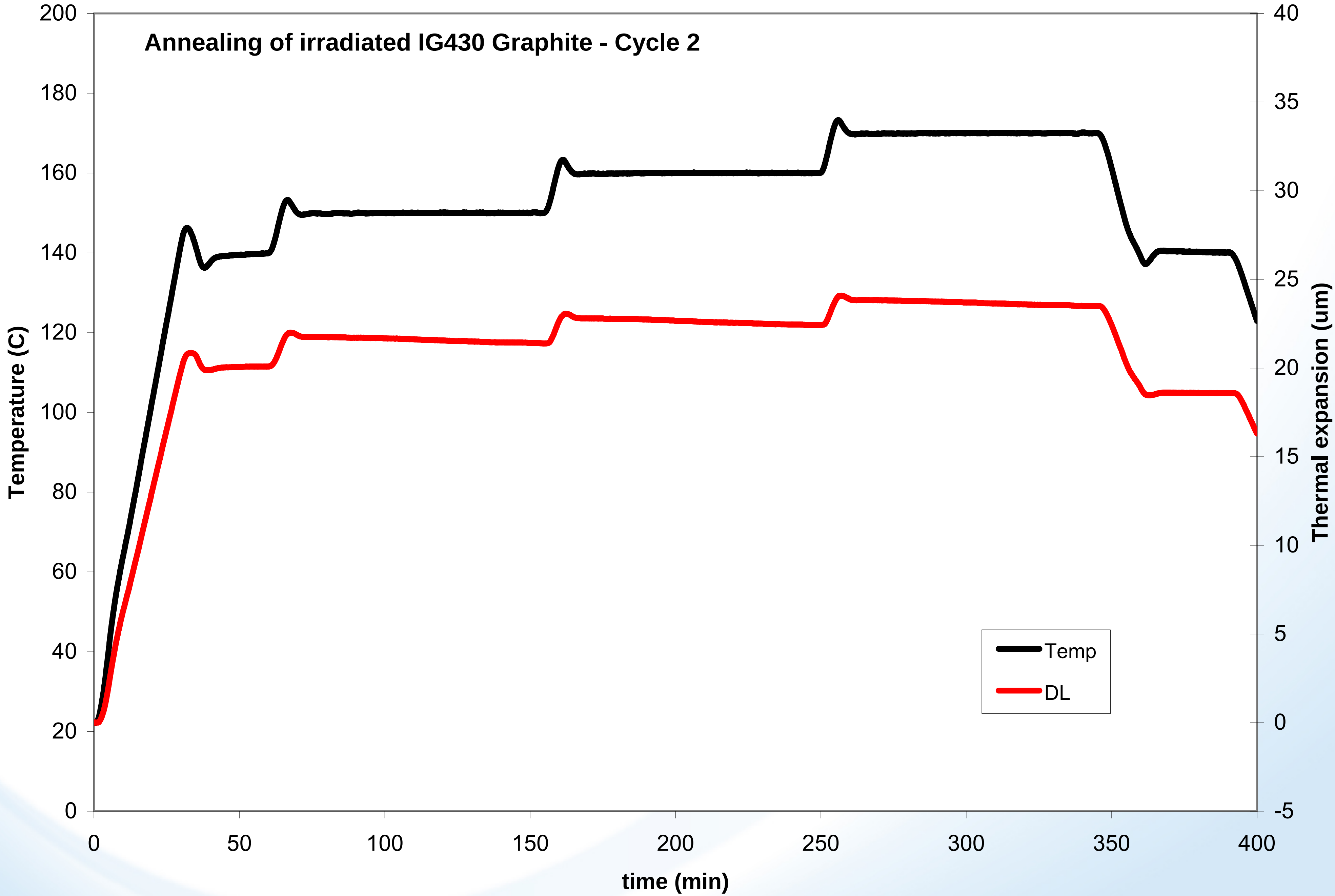



super INVAR (irrad)
irrad Graphite
Thermal Expansion (um)
Temp (C)
160
140
120
100
80
60
40
20
0
-20
-40
0
50
100
150
200
250
300
350
400
450
500


irradiated IG430 graphite
Thermal Expansion (um)
Time (m)
50
40
30
20
10
0
-10
-20
-30
0
100
200
300
400
500
600
700
800
900
1000


super INVAR (irrad)
irrad Graphite
irrad Gum
Thermal Expansion (um)
Temp (C)
350
300
250
200
150
100
50
0
-50
0
100
200
300
400
500
600
700

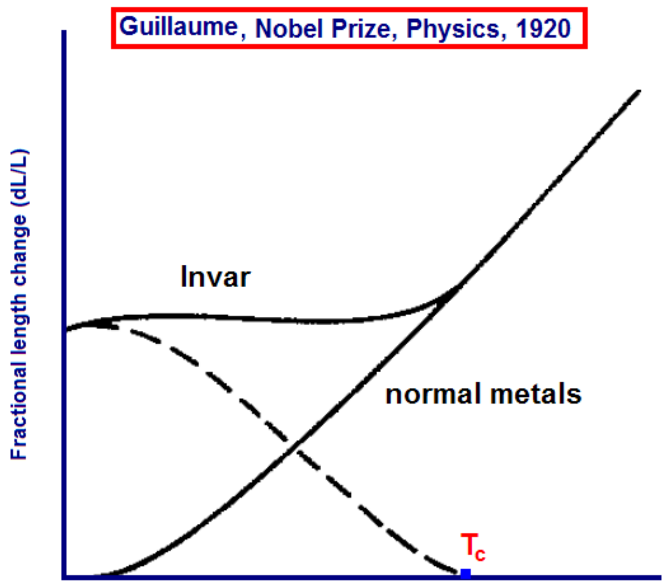

Annealing of Irradiated IG-430 (specimen T54) - Cycle 1
Note: During dweling there is no "annealing"
due to the fact that the irradiation temperature
is higher than the temperature of the two dwels
Temp (C)
Linear Expansion (um)
time (min)
Temp
DL
0
20
40
60
80
100
120
140
160
180
-5
5
10
15
25
30
35

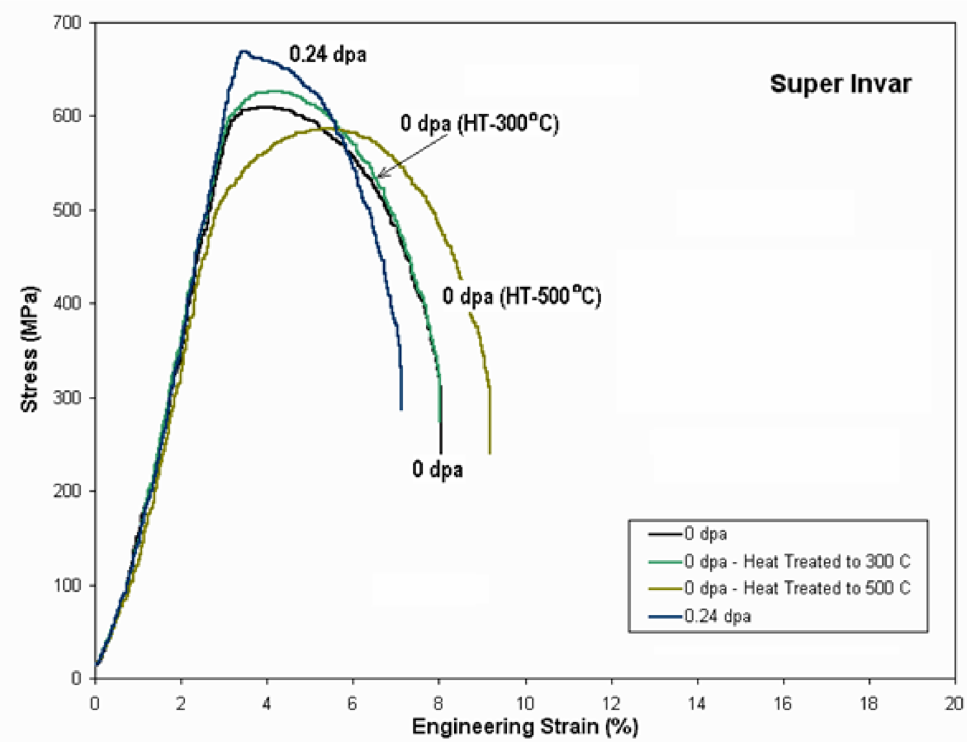


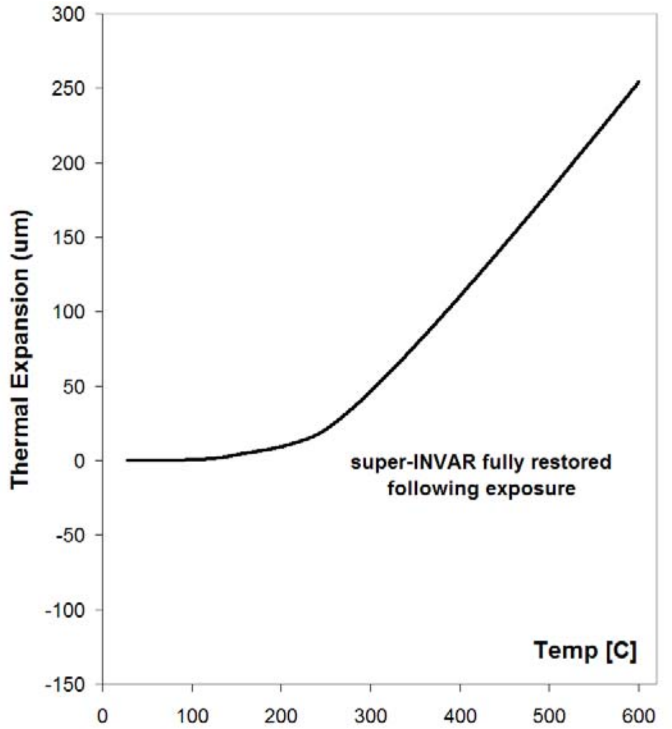
Annealing of irradiated IG430 Graphite - Cycle 2
Temperature (C)
Thermal expansion (um)
time (min)
Temp
DL
200
180
160
140
120
100
80
60
40
20
0
40
35
30
25
20
15
10
5
0
-5
0
50
100
150
200
250
300
350
400

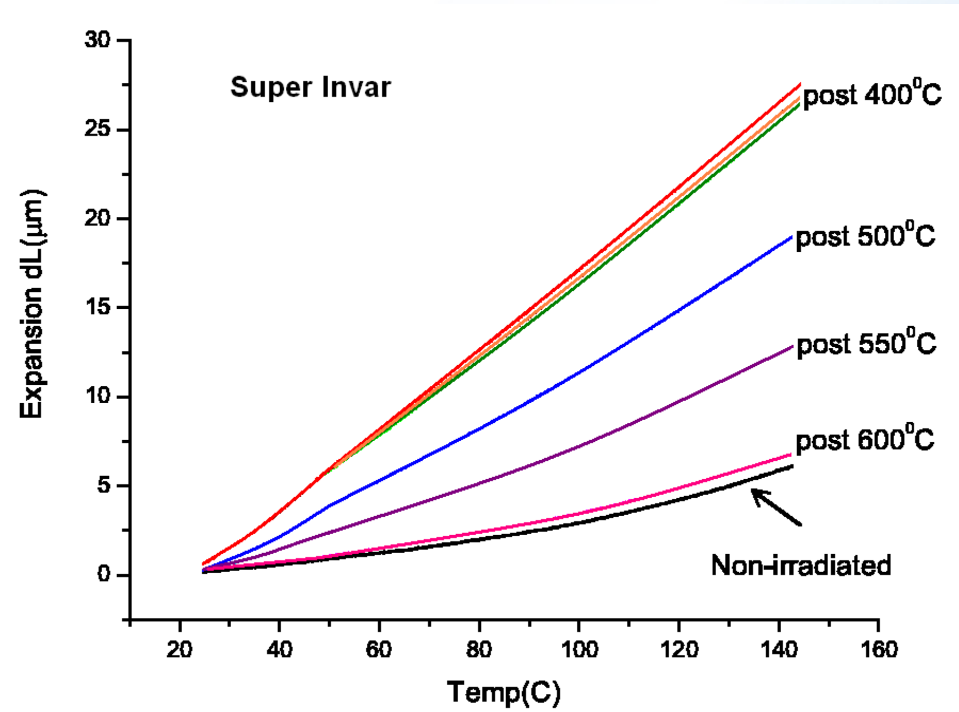
Super Alloys

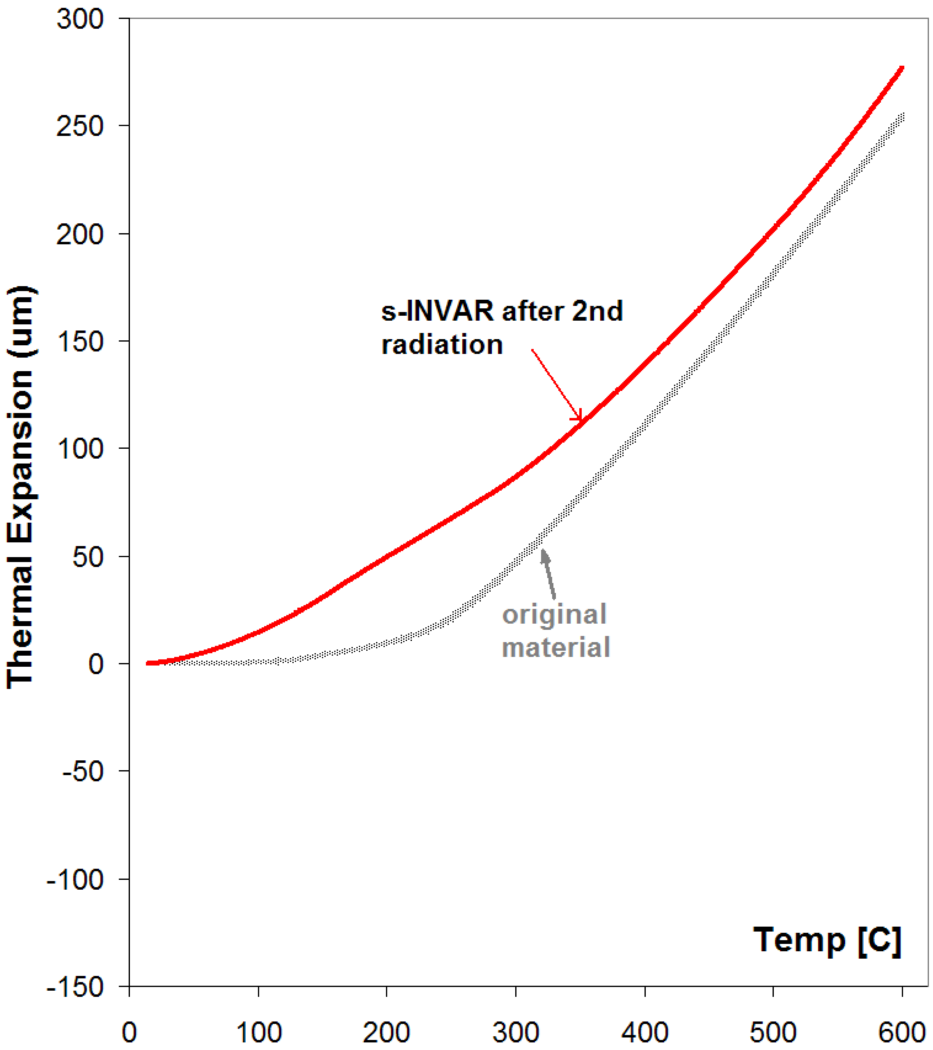
super-Invar

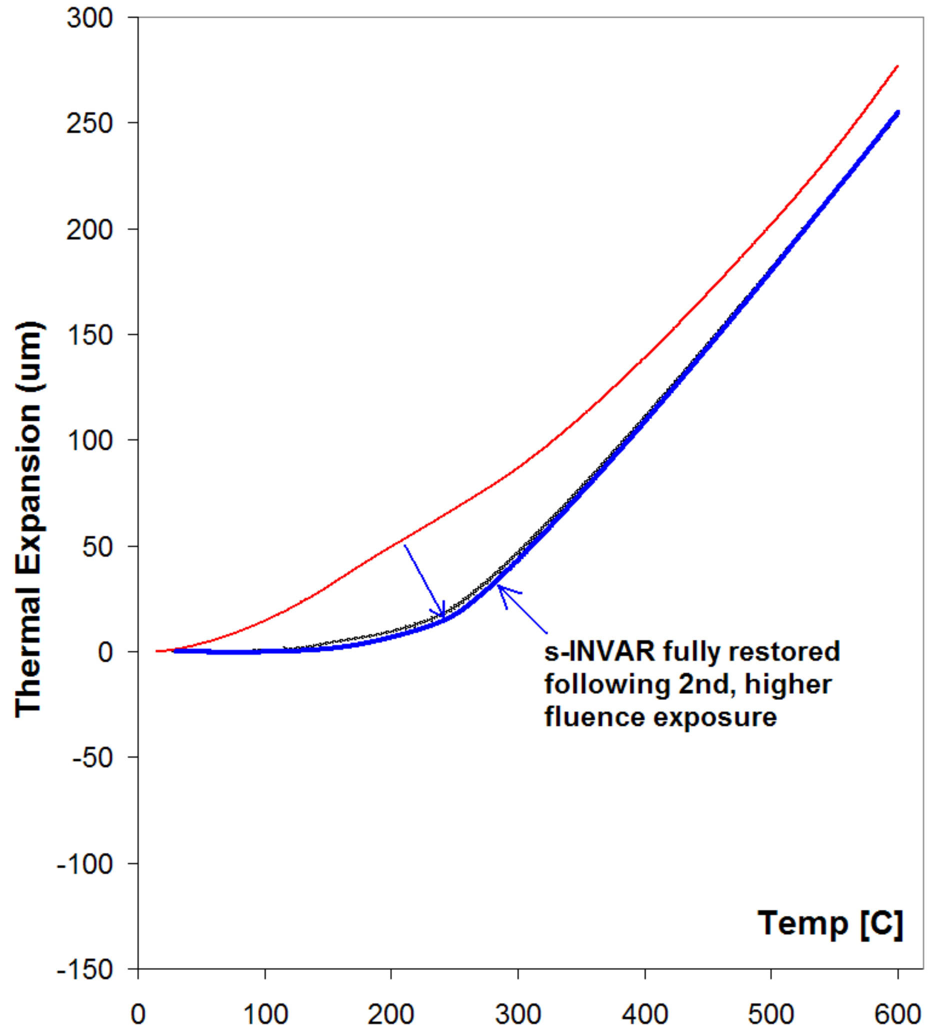

Guillaume, Nobel Prize, Physics, 1920
Fractional length change (dL/L)
Invar
normal metals
Tc

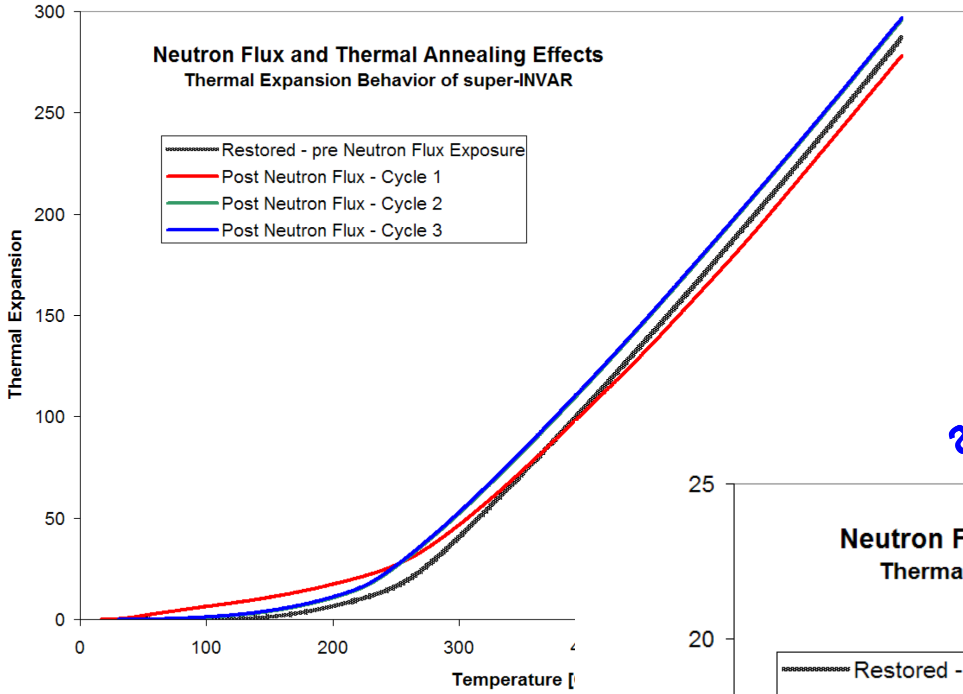

Super Invar
0.24 dpa
0 dpa (HT-300°C)
0 dpa (HT-500°C)
0 dpa
0 dpa
0 dpa - Heat Treated to 300 C
0 dpa - Heat Treated to 500 C
0.24 dpa
Stress (MPa)
Engineering Strain (%)
0
100
200
300
400
500
600
700
0
2
4
6
8
10
12
14
16
18
20


Thermal Expansion (um)
super-INVAR fully restored
following exposure
Temp [C]
300
250
200
150
100
50
0
-50
-100
-150
0
100
200
300
400
500
600

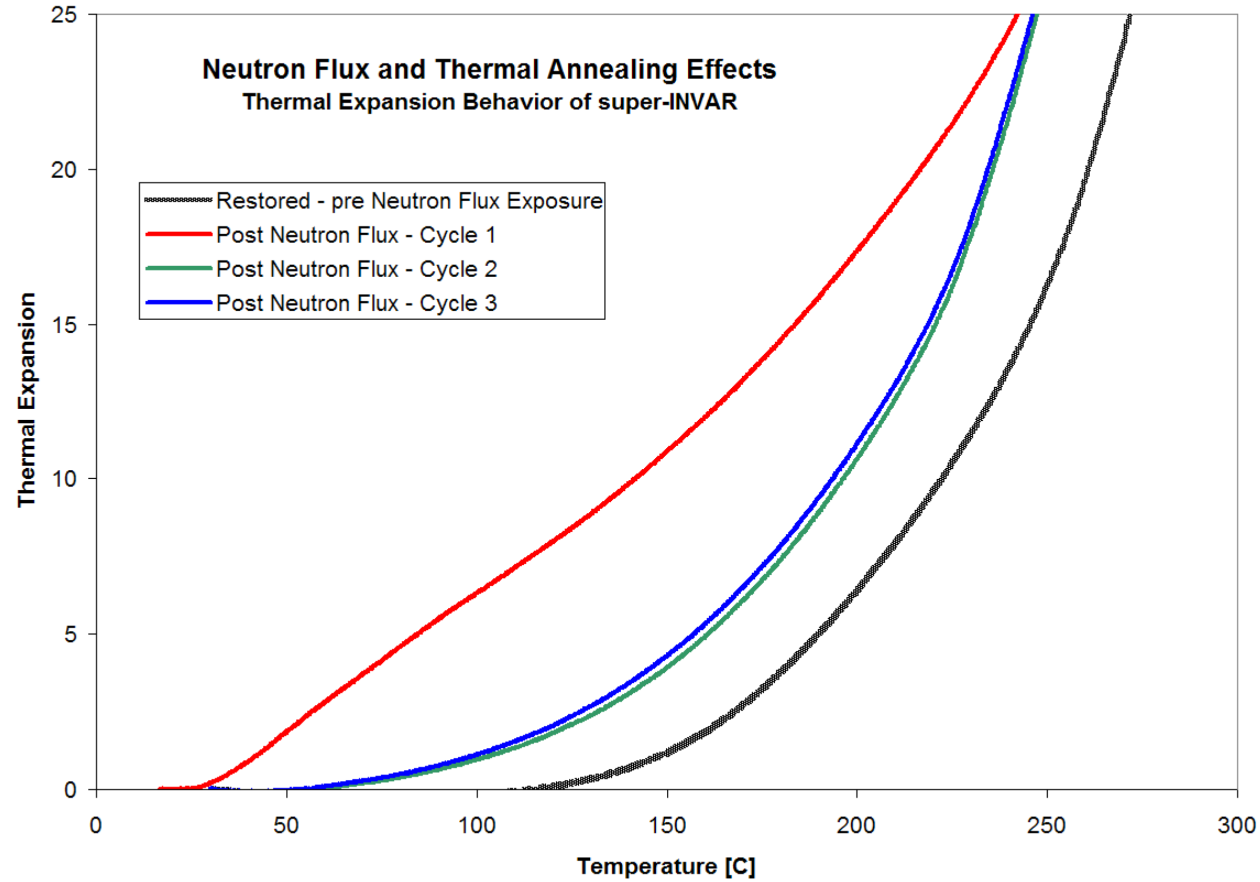

Super Invar
post 400°C
post 500°C
post 550°C
post 600°C
Non-irradiated
Expansion dL(µm)
Temp(C)
30
25
20
15
10
5
0
20
40
60
80
100
120
140
160


300
250
200
150
100
50
0
-50
-100
-150
Thermal Expansion (um)
s-INVAR after 2nd
radiation
original
material
Temp [C]
0
100
200
300
400
500
600

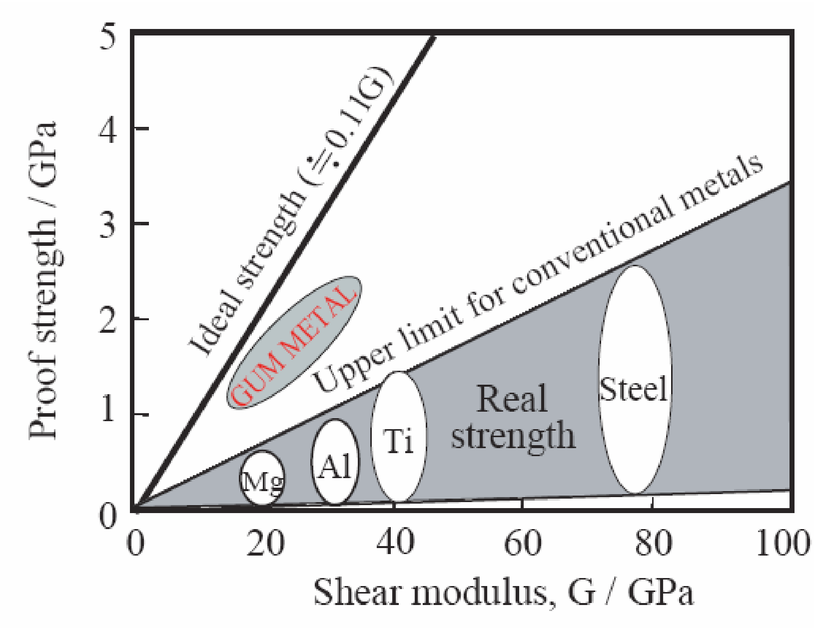

300
250
200
150
100
50
0
-50
-100
-150
Thermal Expansion (um)
s-INVAR fully restored
following 2nd, higher
fluence exposure
Temp [C]
0
100
200
300
400
500
600

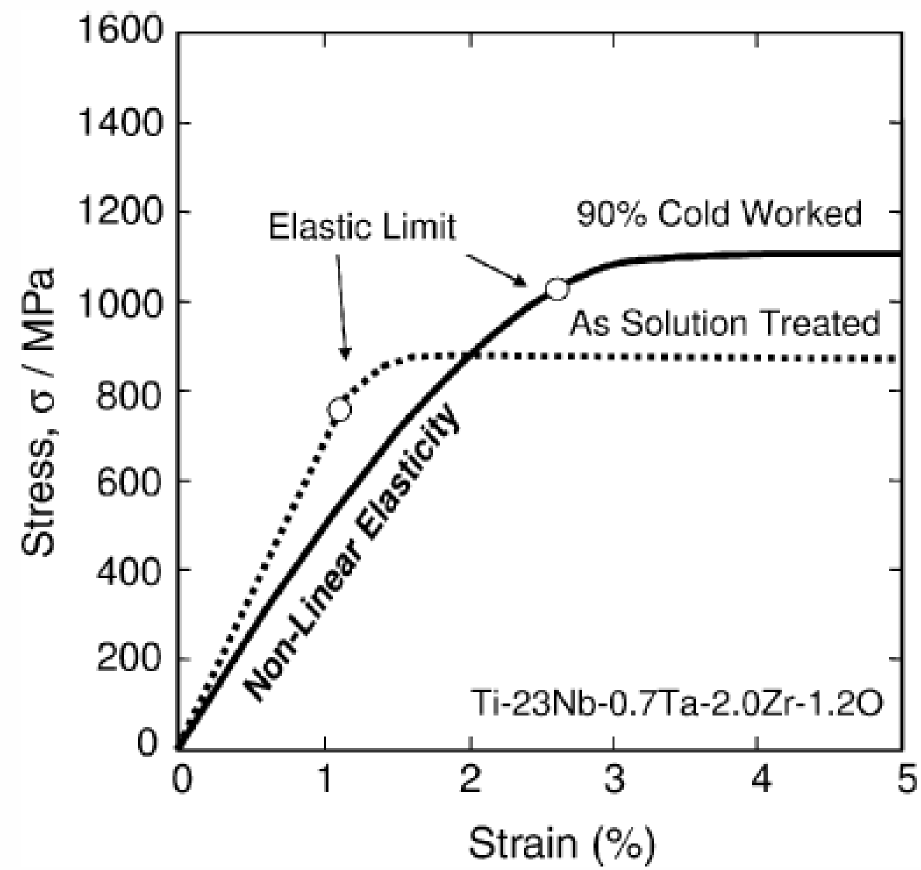


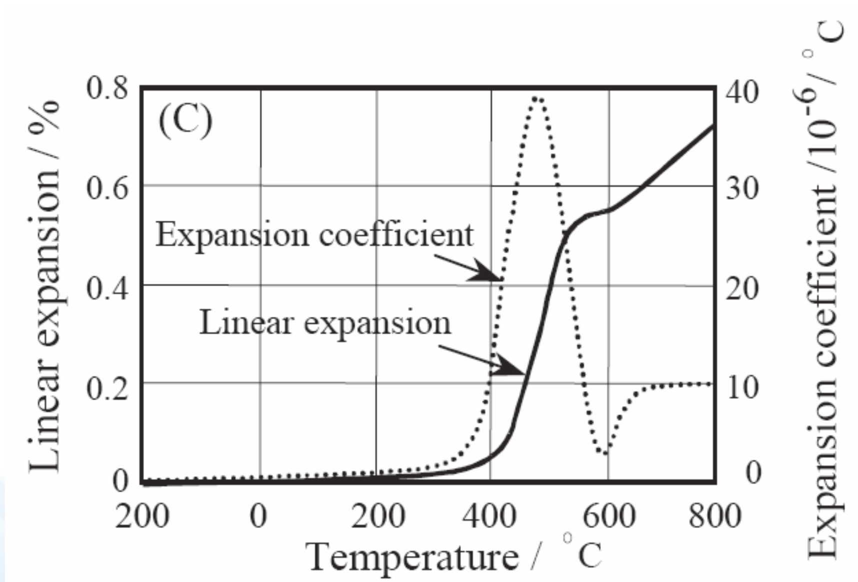
annealing following neutron irradiation



Ti-12Ta-9Nb-3V-6Zr-O ["gum" metal]

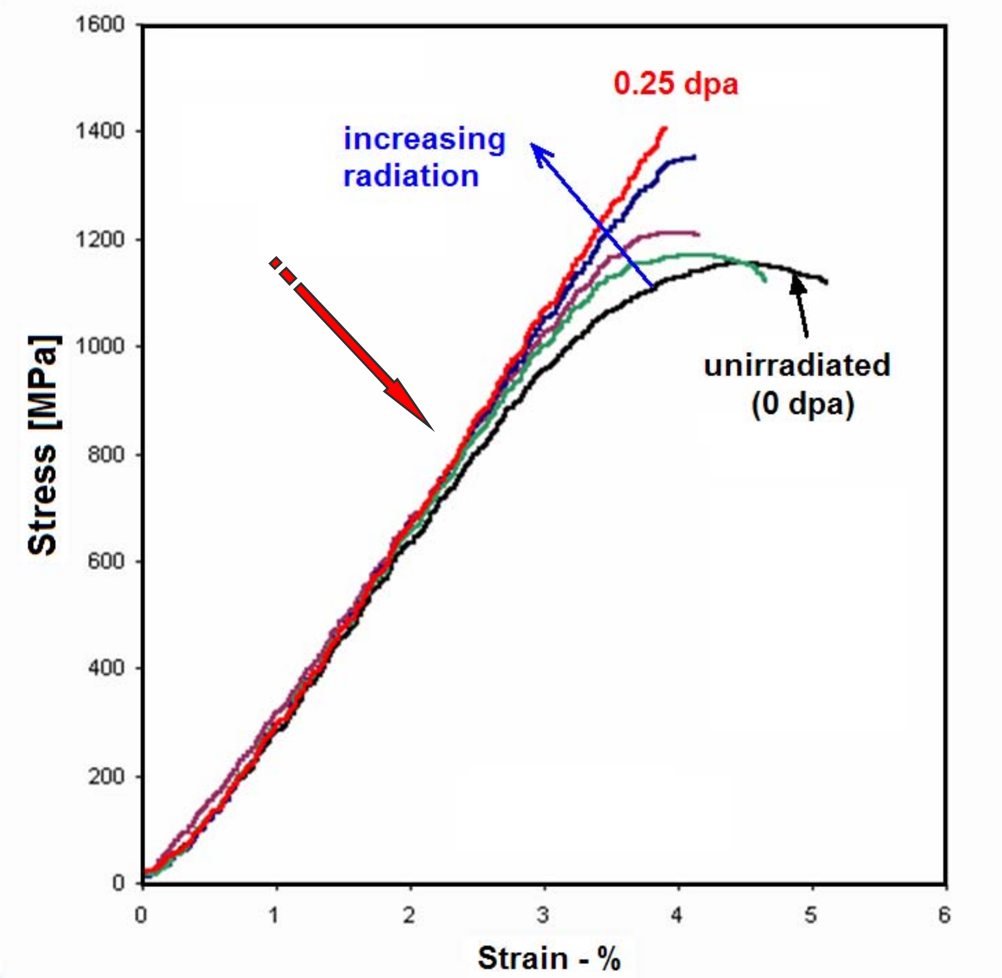


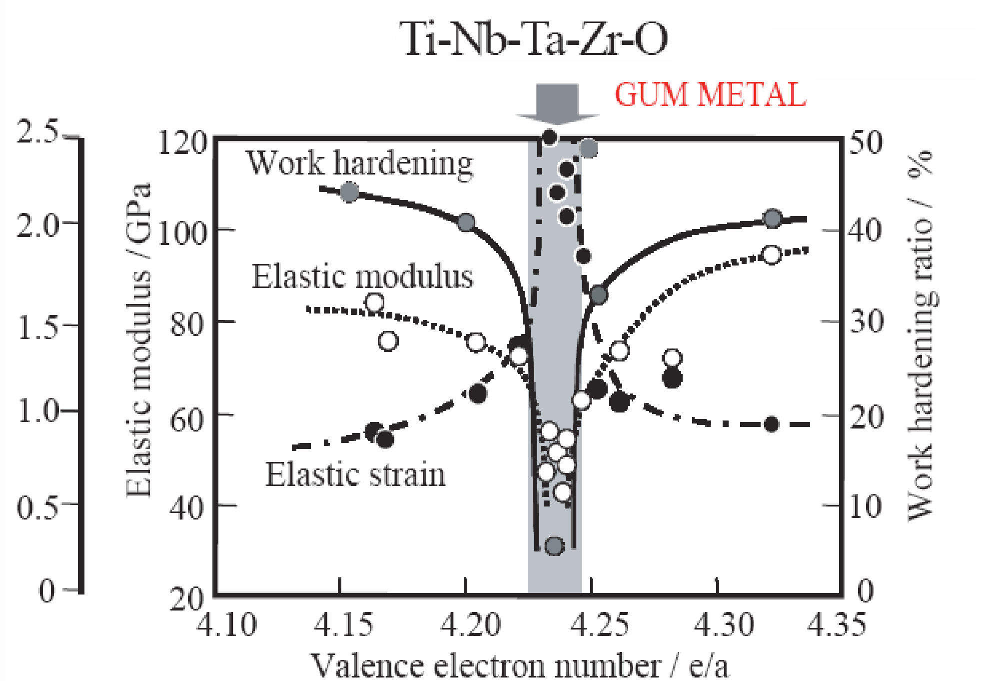




T. Saito, et al., Multifunctional Alloys Obtained via a Dislocation-Free Plastic Deformation Mechanism, Science, 300 (2003) 464

Radiation effects on Mechanical Properties of Gum Metal

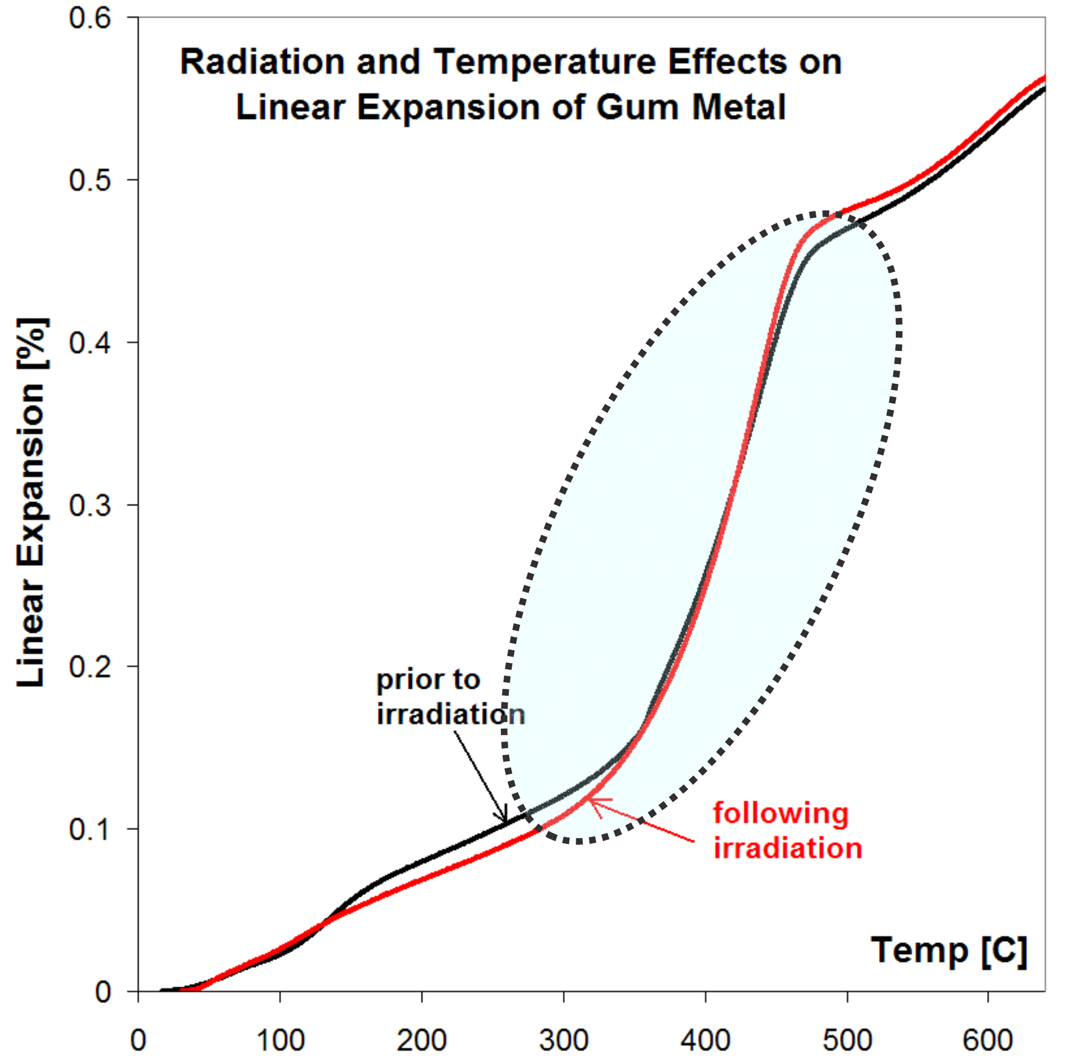


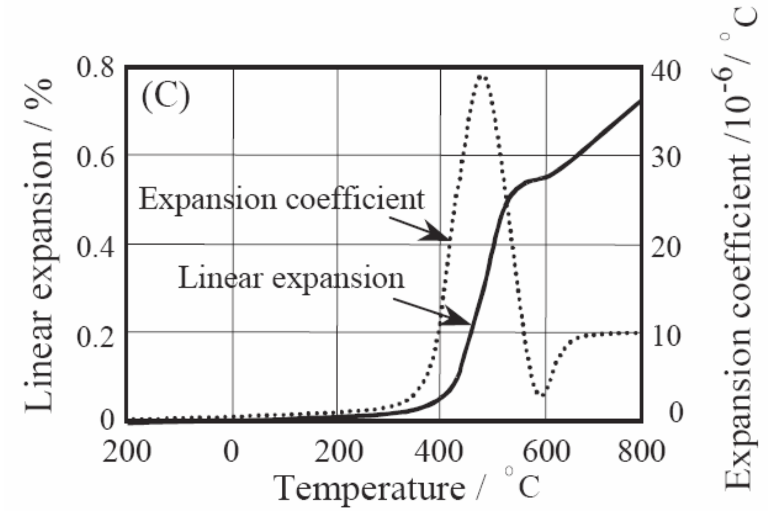


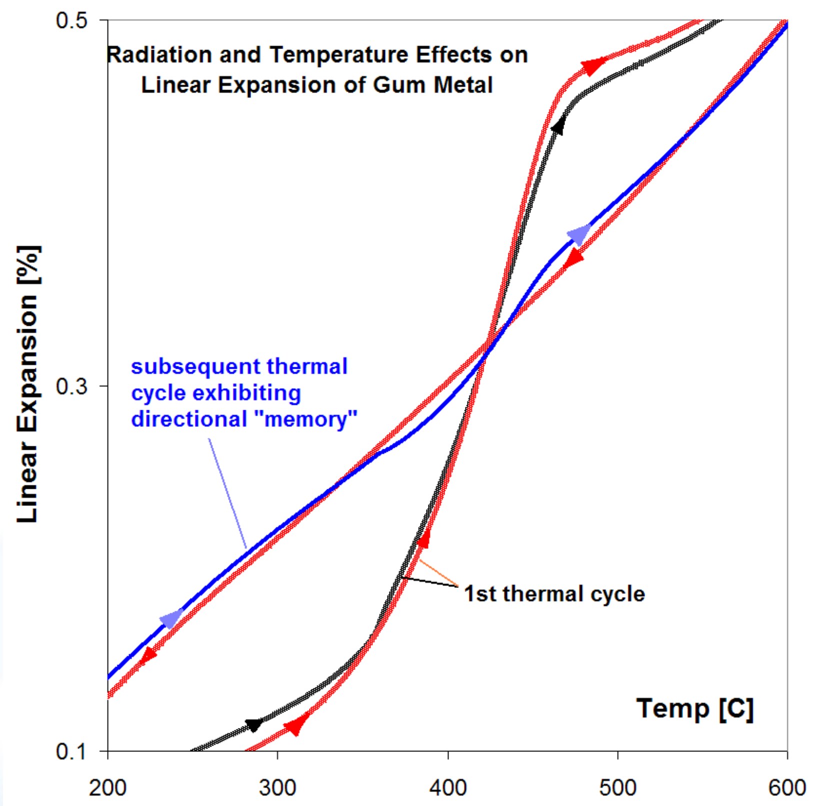
From super-ductile to TOTALLY brittle !
WHY is it so susceptible?

* bombardment of this “unstable” equilibrium
* **He** formed & retained in grain boundaries

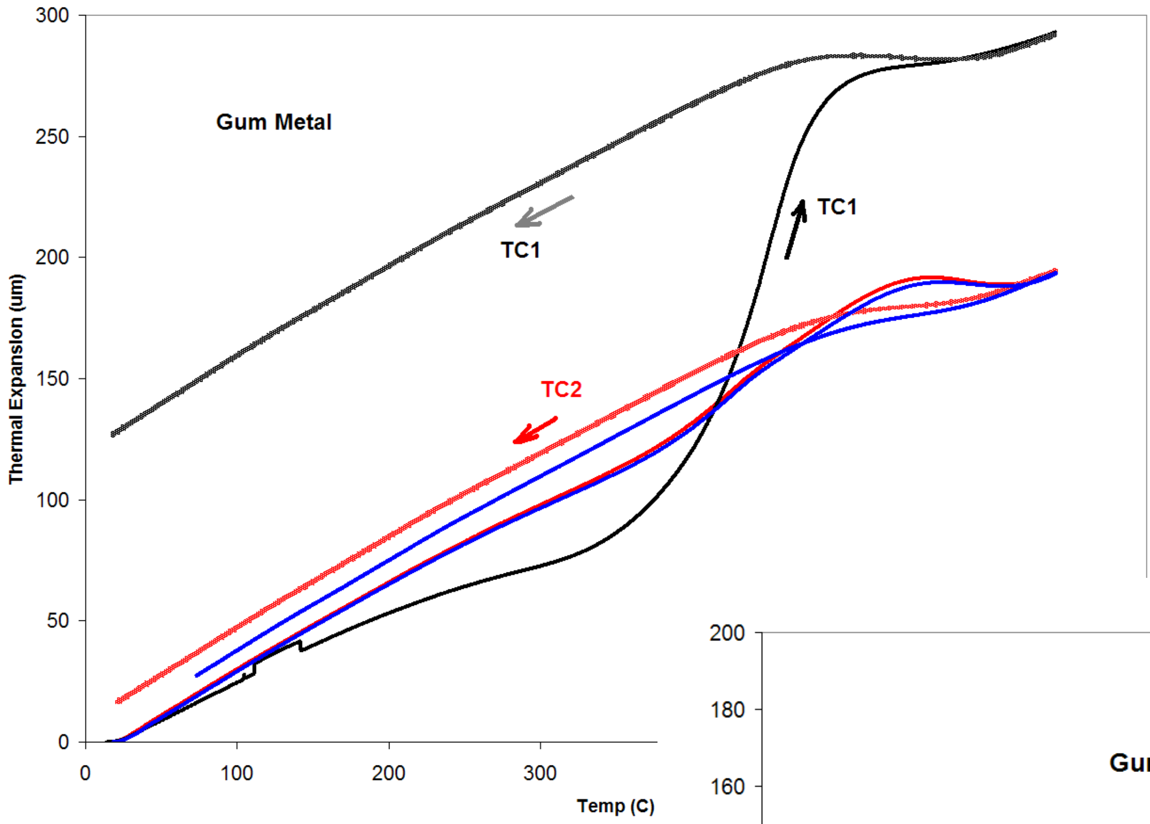
Unique gum metal linear expansion behavior

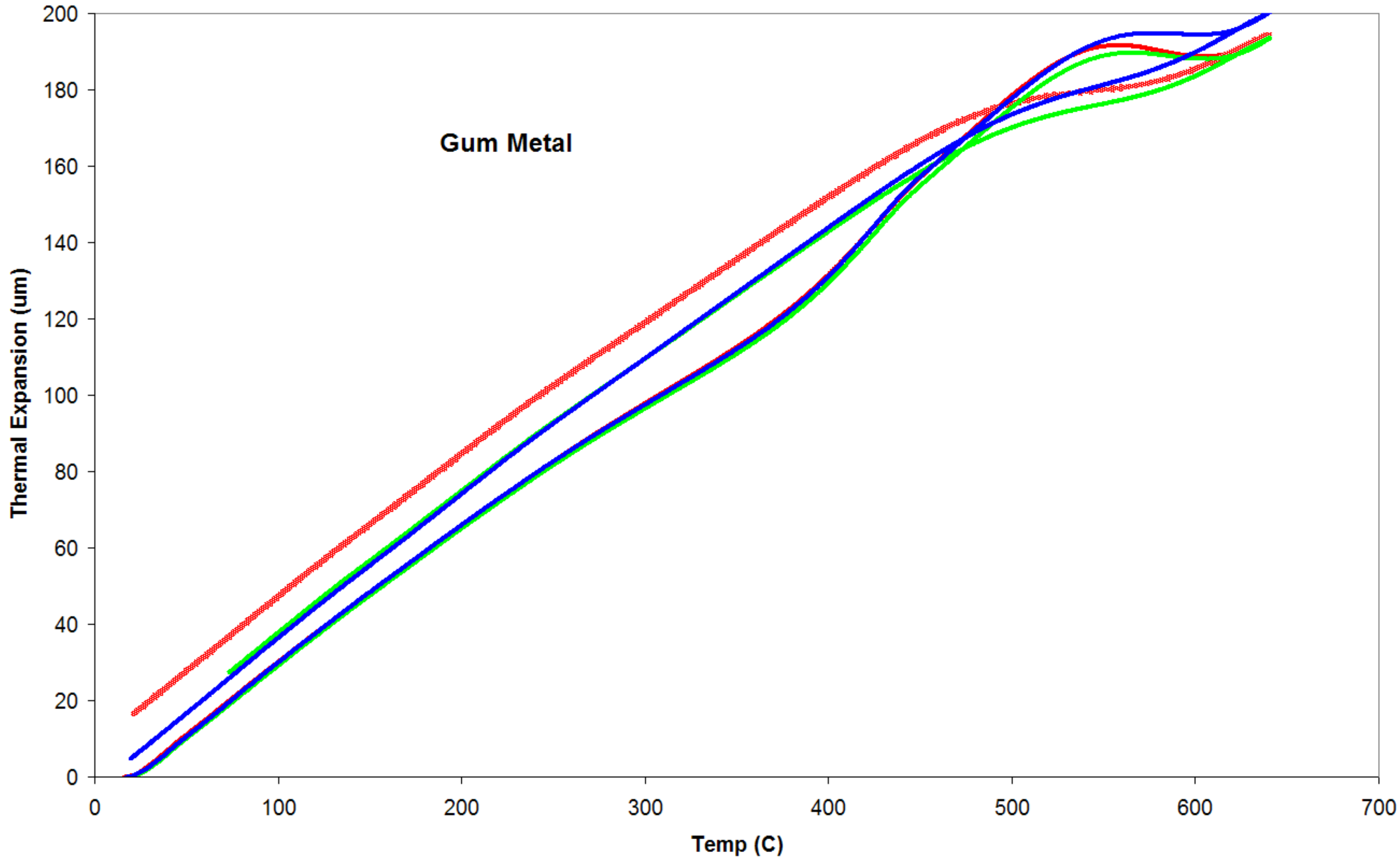




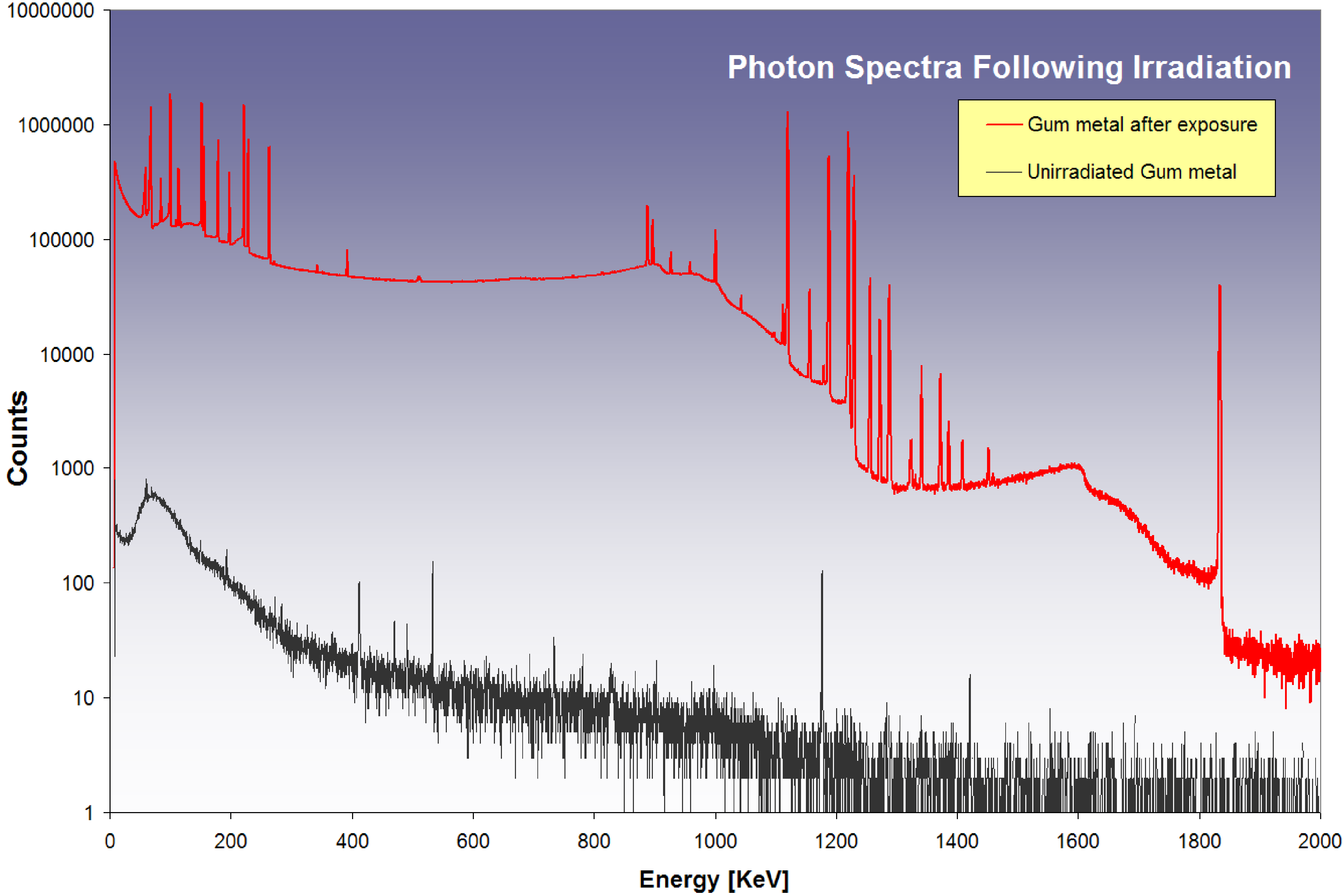


Irradiated Gum Metal

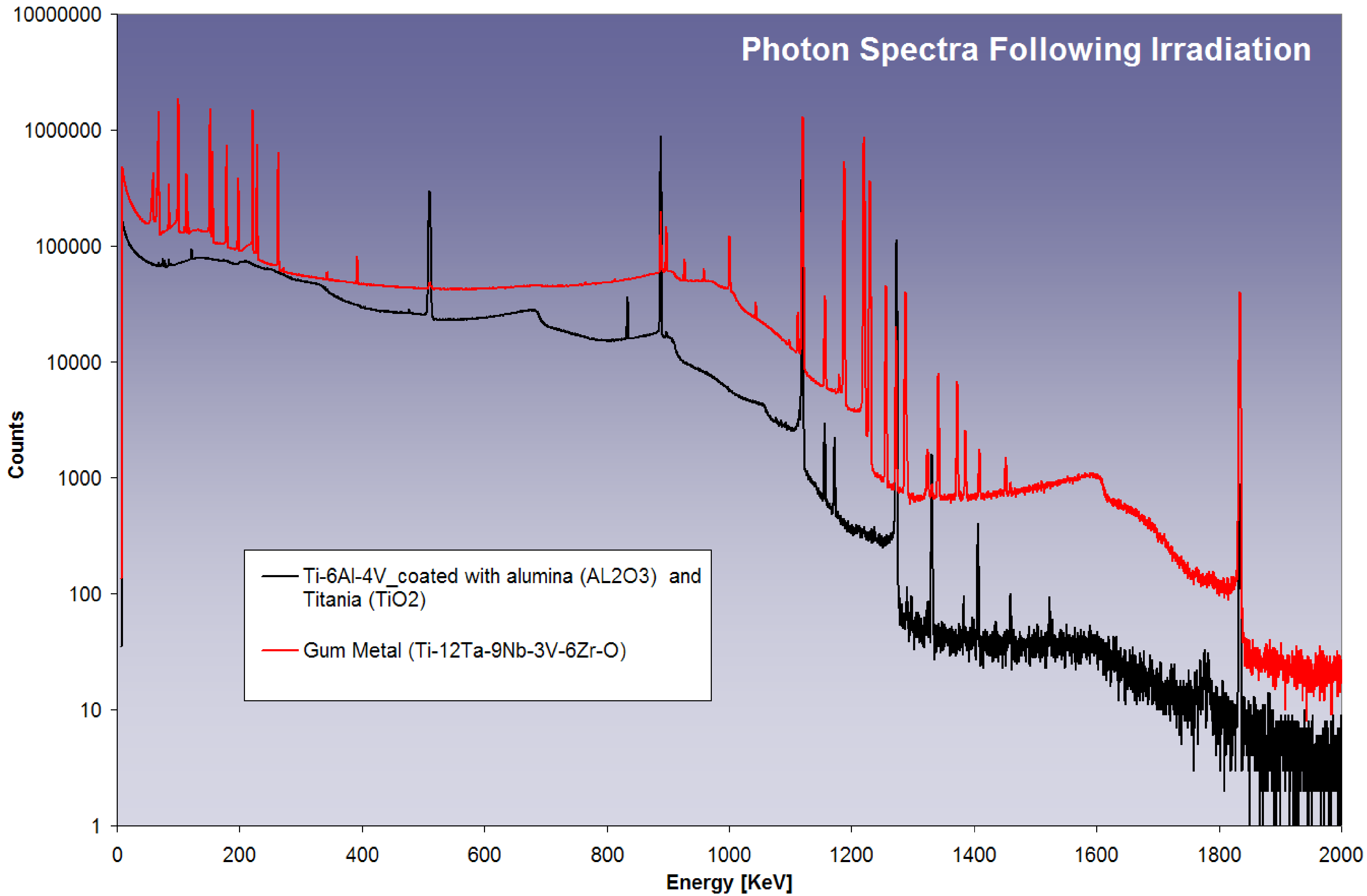


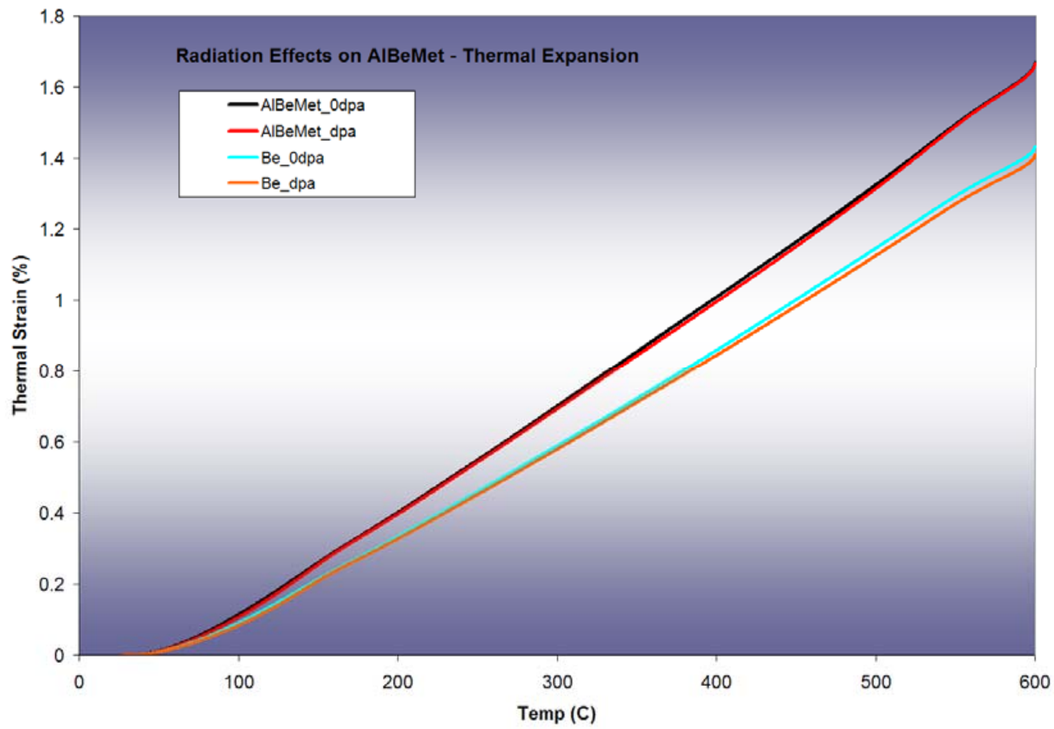


Characterization using Photon Spectra

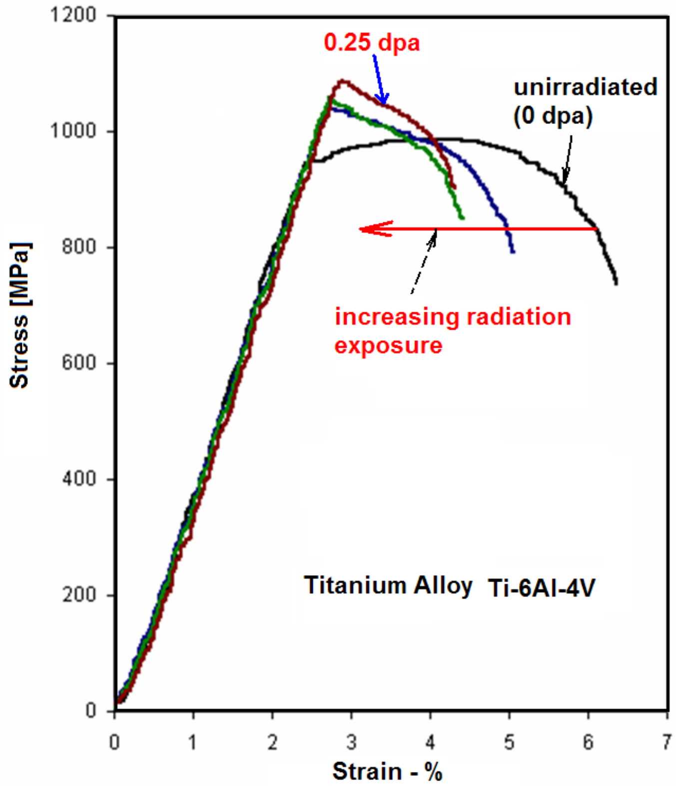


Characterization using Photon Spectra

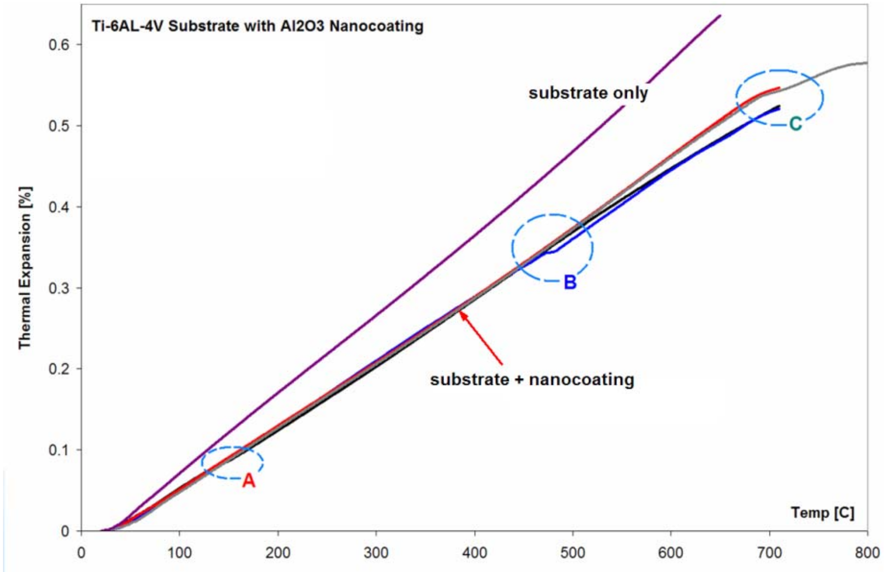



Radiation Effects on AlBeMet - Thermal Expansion
AlBeMet_0dpa
AlBeMet_dpa
Be_0dpa
Be_dpa
Thermal Strain (%)
Temp (C)
1.8
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
0
100
200
300
400
500
600

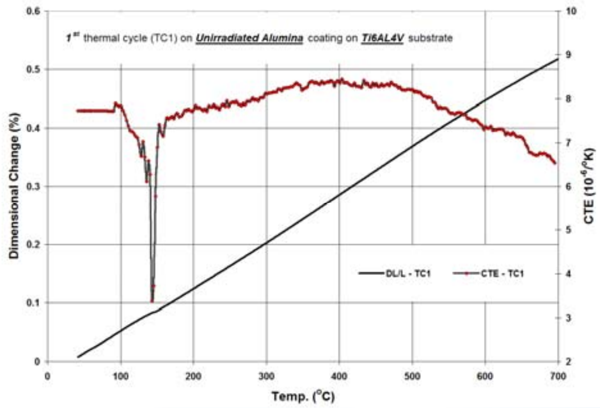
Ti-6Al-4V



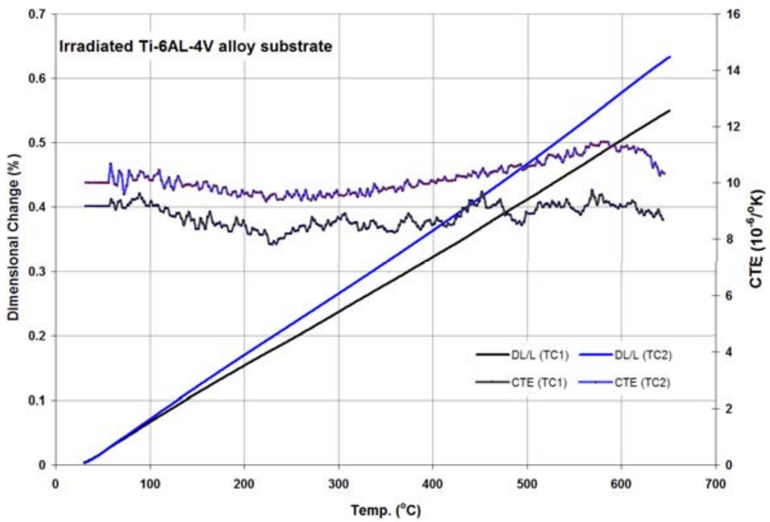
Stress [MPa]
Strain - %
0.25 dpa
unirradiated (0 dpa)
increasing radiation exposure
Titanium Alloy Ti-6Al-4V



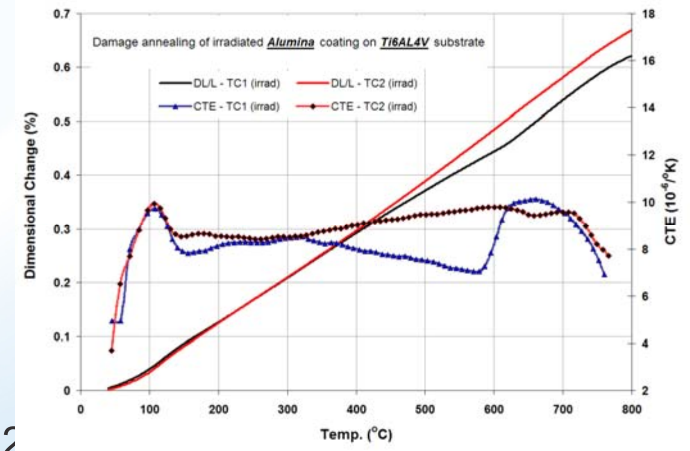
Ti-6AL-4V Substrate with Al2O3 Nanocoating
Thermal Expansion [%]
Temp [C]
substrate only
substrate + nanocoating
A
B
C



1st thermal cycle (TC1) on Unirradiated Alumina coating on Ti6AL4V substrate
Dimensional Change (%)
CTE (10^{-6}/°K)
Temp. (°C)
DL/L - TC1
CTE - TC1



Irradiated Ti-6AL-4V alloy substrate
Dimensional Change (%)
CTE (10^{-6}/°K)
Temp. (°C)
DL/L (TC1)
DL/L (TC2)
CTE (TC1)
CTE (TC2)

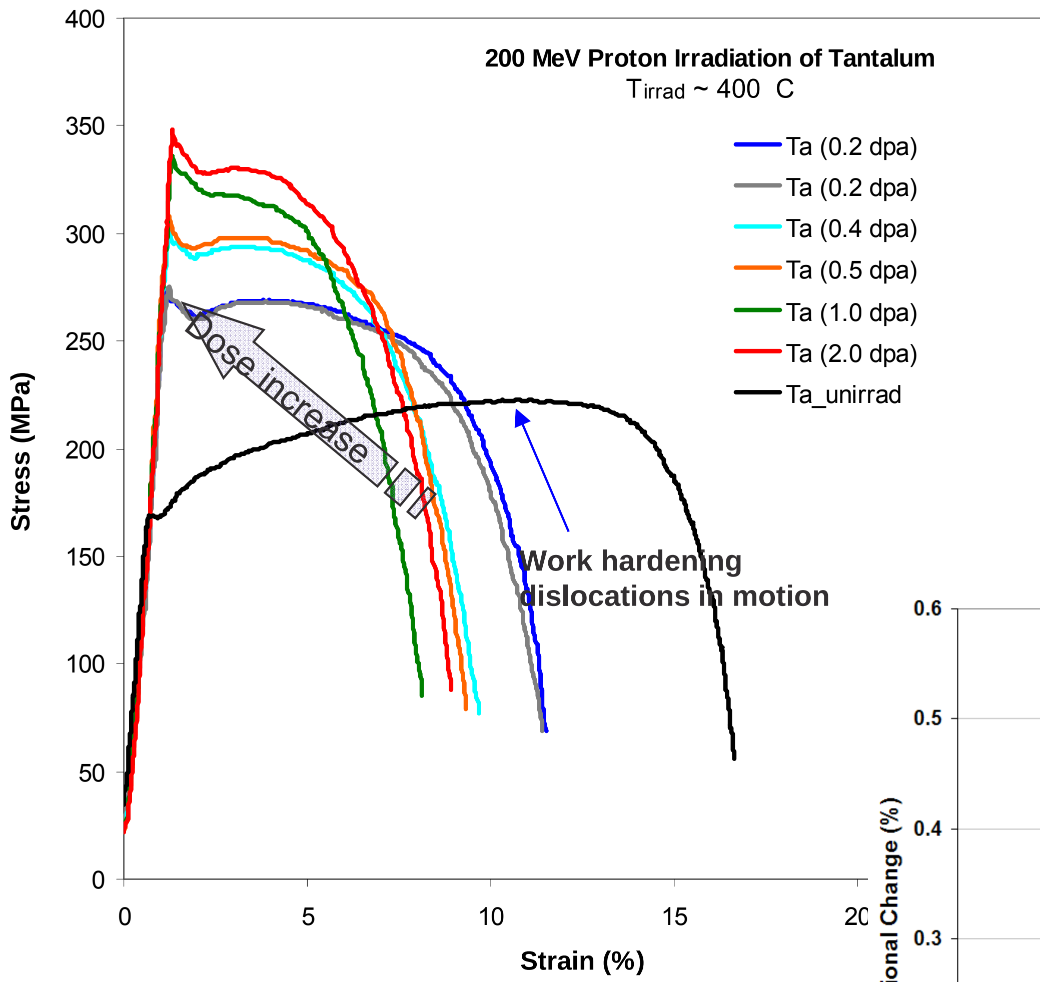


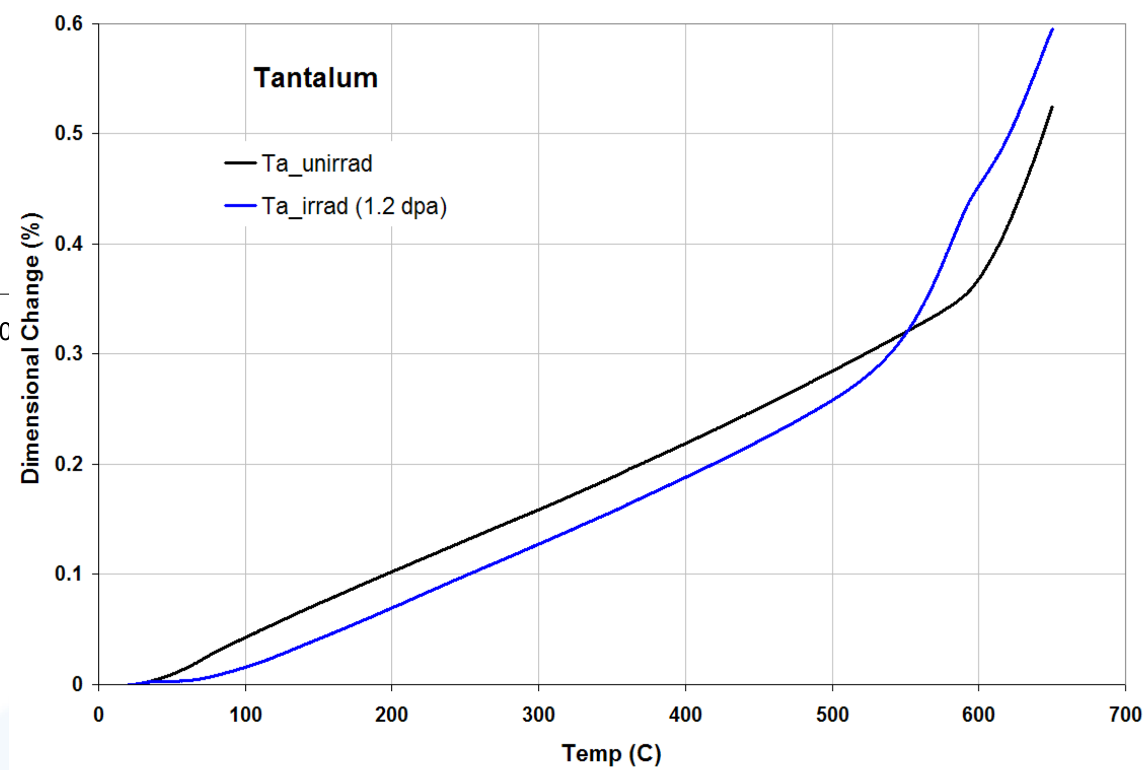
Damage annealing of irradiated Alumina coating on Ti6AL4V substrate
Dimensional Change (%)
CTE (10^{-6}/°K)
Temp. (°C)
DL/L - TC1 (irrad)
DL/L - TC2 (irrad)
CTE - TC1 (irrad)
CTE - TC2 (irrad)

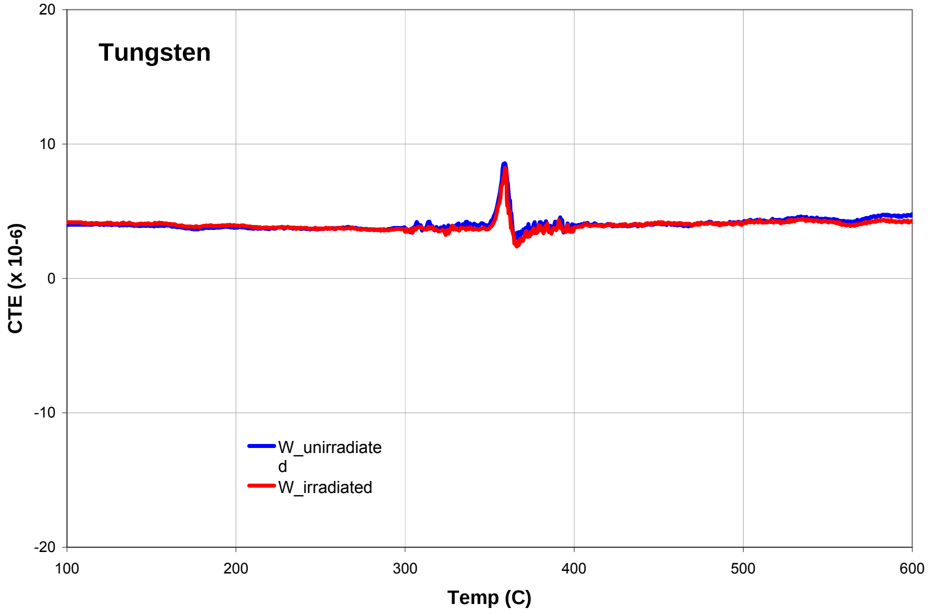
High-Z Materials and Irradiation

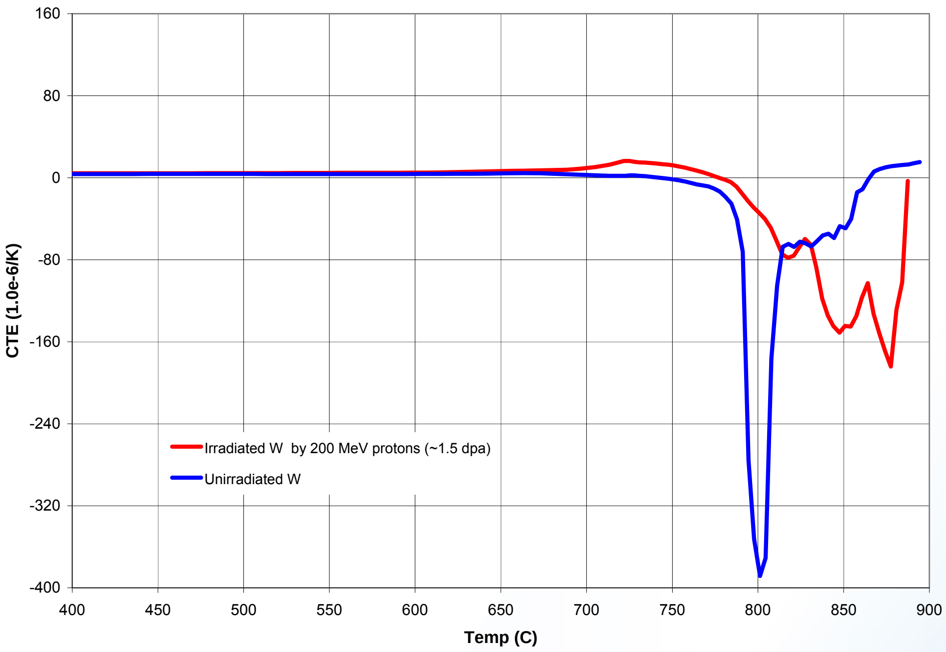
Experimental Set-Up addressing Oxidation/Volumetric Change (i.e. **tantalum)**

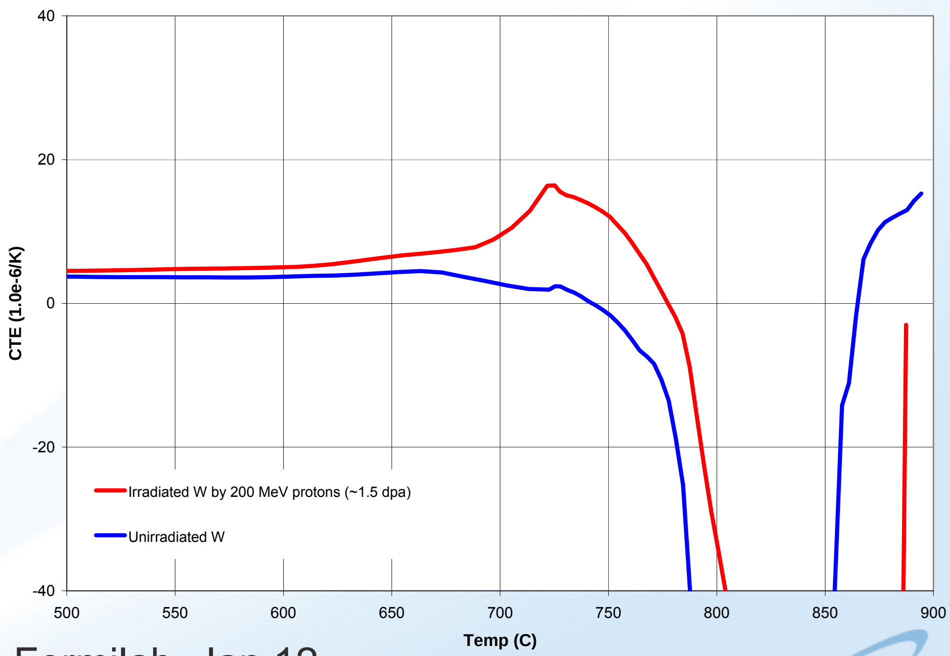
Accelerated Ta Oxidation:
Present of a third element
Radiation-induced oxidation acceleration ?


200 MeV Proton Irradiation of Tantalum
Tirrad ~ 400 C
Ta (0.2 dpa)
Ta (0.2 dpa)
Ta (0.4 dpa)
Ta (0.5 dpa)
Ta (1.0 dpa)
Ta (2.0 dpa)
Ta_unirrad
Dose increase
Work hardening
dislocations in motion
Stress (MPa)
Strain (%)
0
50
100
150
200
250
300
350
400
0
5
10
15
20

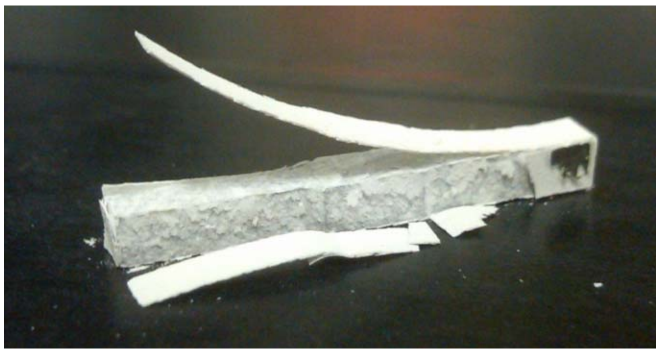

Tantalum
Ta_unirrad
Ta_irrad (1.2 dpa)
Dimensional Change (%)
Temp (C)
0
0.1
0.2
0.3
0.4
0.5
0.6
0
100
200
300
400
500
600
700

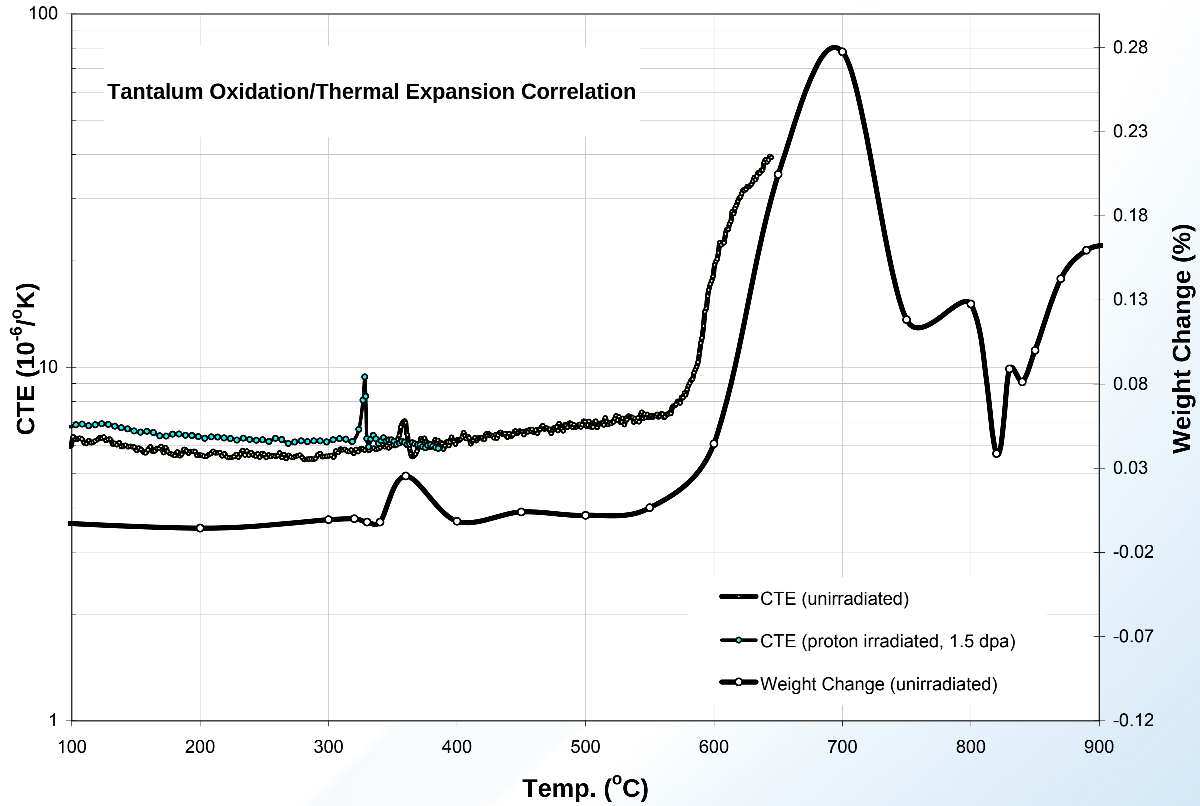

Tungsten
CTE (x 10-6)
Temp (C)
W_unirradiated
W_irradiated
20
10
0
-10
-20
100
200
300
400
500
600

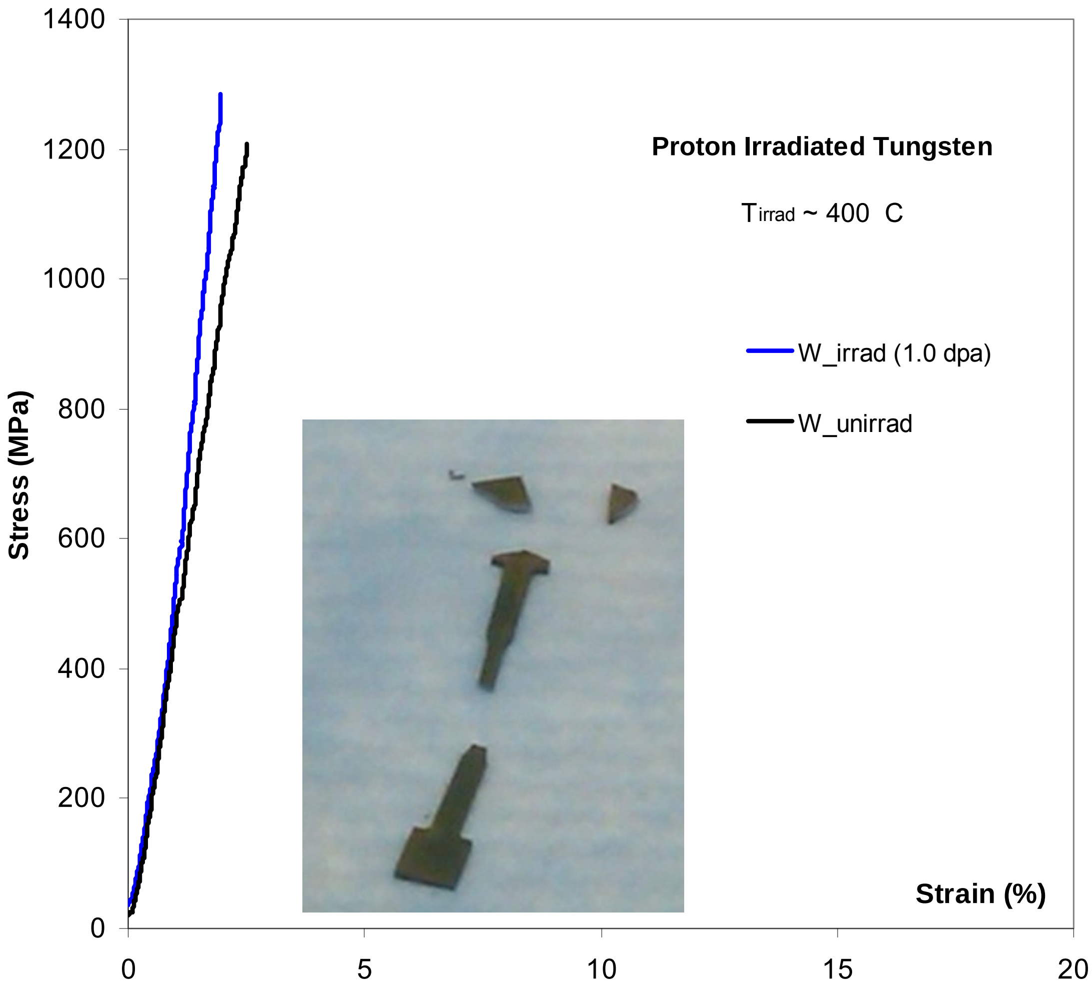

CTE (1.0e-6/K)
Temp (C)
Irradiated W by 200 MeV protons (~1.5 dpa)
Unirradiated W
160
80
0
-80
-160
-240
-320
-400
400
450
500
550
600
650
700
750
800
850
900


CTE (1.0e-6/K)
Temp (C)
Irradiated W by 200 MeV protons (~1.5 dpa)
Unirradiated W
40
20
0
-20
-40
500
550
600
650
700
750
800
850
900

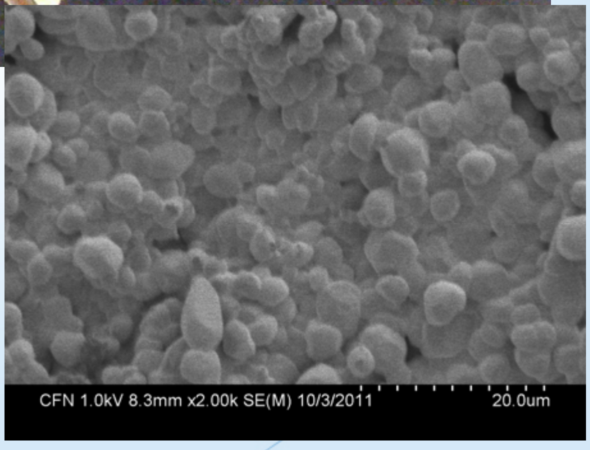

Ta40



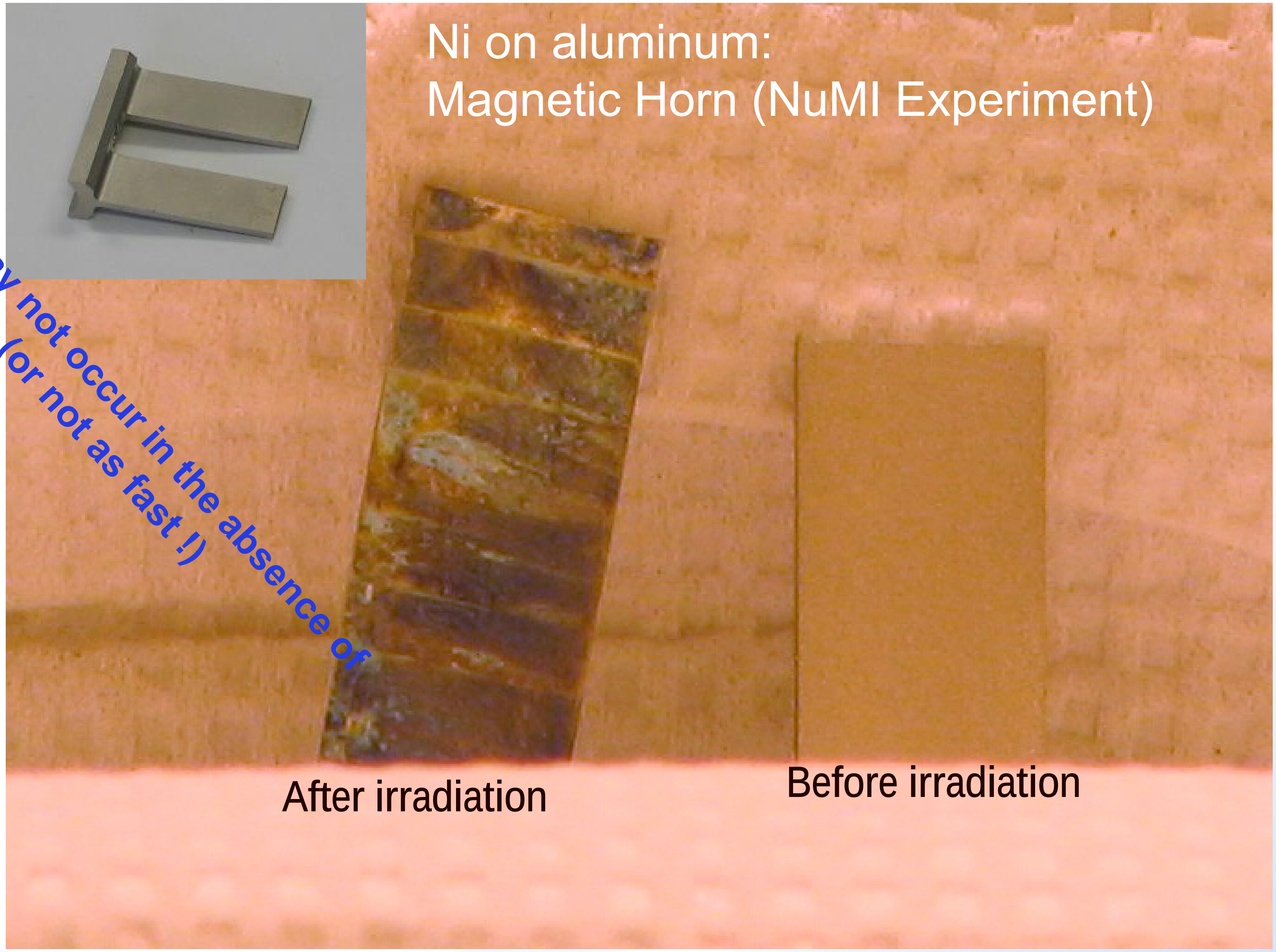

Tantalum Oxidation/Thermal Expansion Correlation
CTE (10^-6/°K)
Weight Change (%)
Temp. (°C)
100
10
1
0.28
0.23
0.18
0.13
0.08
0.03
-0.02
-0.07
-0.12
100
200
300
400
500
600
700
800
900
CTE (unirradiated)
CTE (proton irradiated, 1.5 dpa)
Weight Change (unirradiated)



2 dpa (70% mass loss)



Irradiation, temperature and corrosive environment on Ni film with AL substrate



This may not occur in the absence of irradiation (or not as fast !)

Way Forward

Radiation Damage R&D Opportunities

- Further irradiation and testing of promising target materials and **"super" materials**
  - Requires beam time, hot cell work & Material Science expertise
- Correlate LE neutron irradiation data to HE proton regime to take advantage of neutron irradiation data
  - Requires Material Science expertise & access to data
- Study **effects of gas production** in solid and liquid target materials
  - Requires Material Science, Radiochemistry, and Simulation expertise
  - May require testing

Radiation Damage R&D Opportunities

- Irradiation tests to validate DPA and gas production simulation tools
  - Requires Material Science and Simulation expertise
  - Requires beam time & hot cell work
- Participate in activities to characterize irradiated materials from operating facilities (such as the SNS target vessel testing)
  - Requires Mechanical Engineering, Material Science expertise

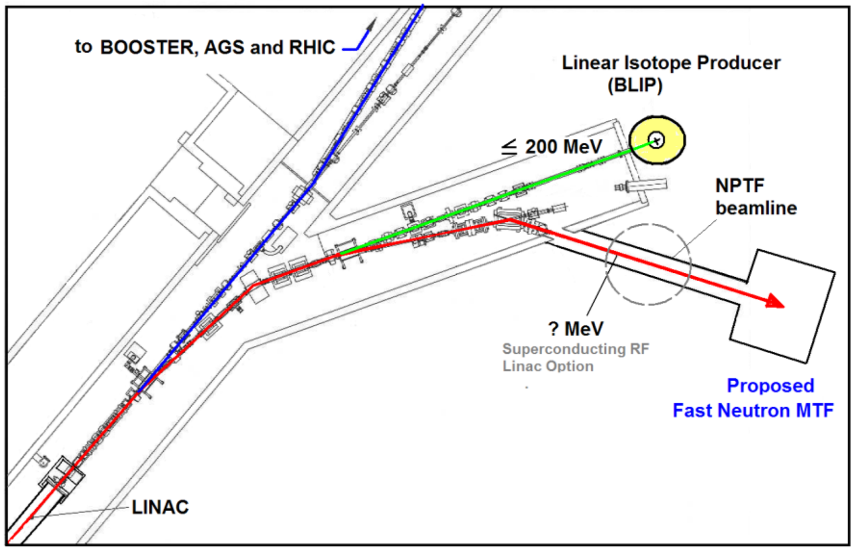
Radiation Damage R&D Opportunities

- Develop white paper outlining parameter space required for an irradiation test facility
  - Requires ME, Mat'l Science, Accelerator Physicist, and Simulation expertise
- Develop proposals for non-parasitic irradiation test sites at operating facilities (NuMI hadron absorber?)
  - Requires ME, Mat'l Science, Accelerator Physicist, and Simulation expertise

BLAIRR
The Brookhaven Linear Accelerator IRRadiation Test Facility

To provide a test bed for:

- material irradiation under protons and neutrons (thermal and fast)
- Neutron scattering and damage assessment



Unique Features

Proton Energy Optimization and Selectivity for target material damage (FFAG and coupled cavities are explored)

Selectivity of neutron spectra for material damage

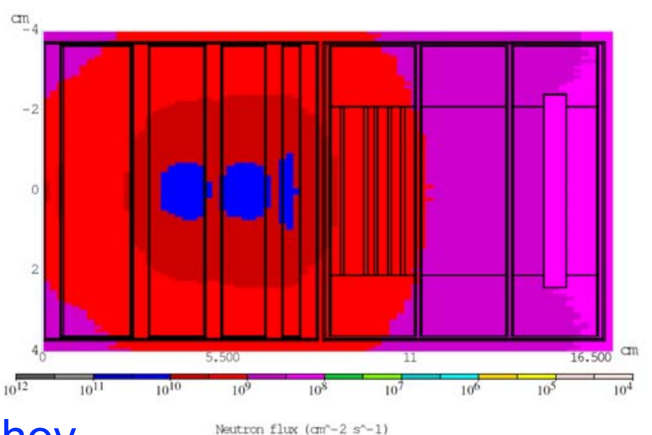
Neutron scattering for micro-scale damage evaluation

Brookhaven Linear Accelerator IRRadiation Test Facility

Present

Parameter Space
92-200 MeV Proton Beam
105 μA
Beam Sigma
Spallation neutron spectrum from 112 MeV Protons
Sharing Target Irradiation R

Activities
Material irradiation damage by proton irradiation
Spallation neutron material irradiation



N. Mokhov

Future

Dedicated Target Space customized/optimized

Synchronous proton and neutron irradiation damage

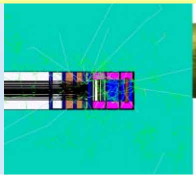
Utilization of Spatial properties of spallation neutron spectrum

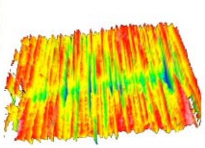
Tunable proton energy to several hundred MeV (optimized according to the primary target and the neutron spectra desired)

MOST importantly use spallation neutrons for neutron scattering studies of irradiation damage

BLAIRR – A Unique, ALL IN ONE OPERATION

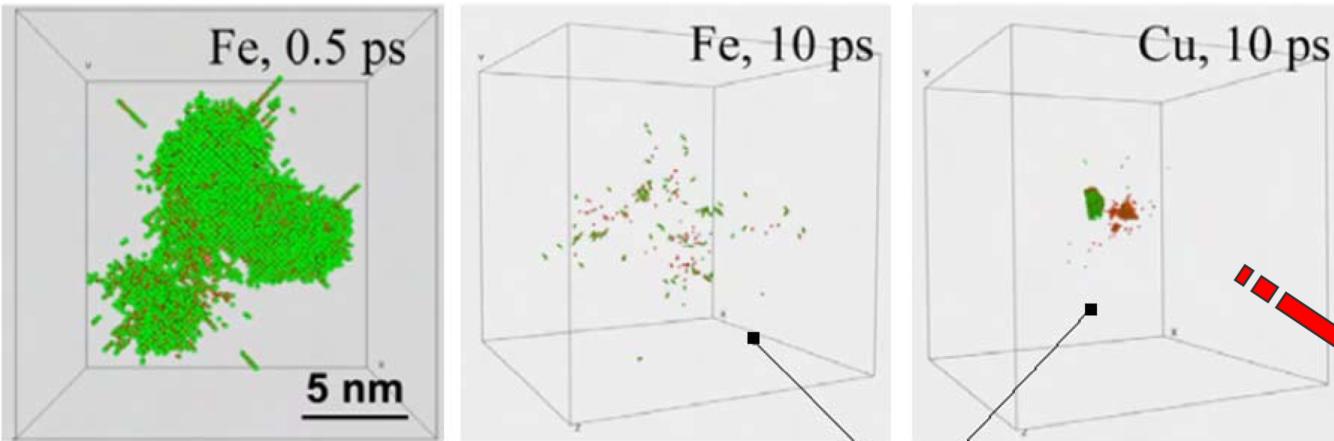
High Rate Irradiation Damage from Energetic Protons
Fast Neutron Irradiation or Fast and Fusion Reactor Materials
Crystal and Special Detector Performance Degradation
Neutron Scattering based Damage Assessment
Macroscopic Damage Assessment
Computational Damage Model Verification





MOLECULAR DYNAMICS SIMULATION

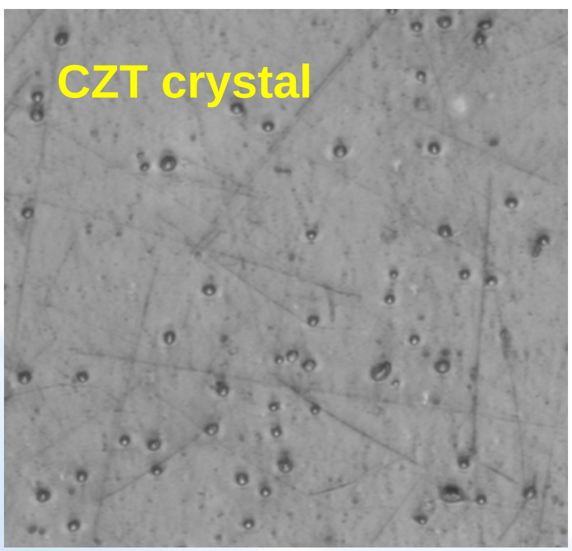
Atomic Dsisplacement Cascade

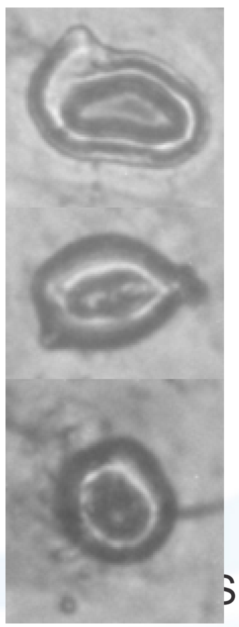


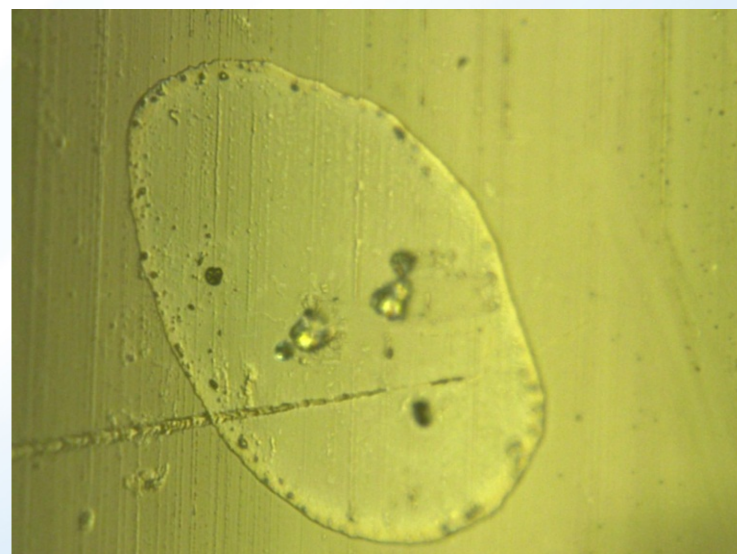
peak of displacement event

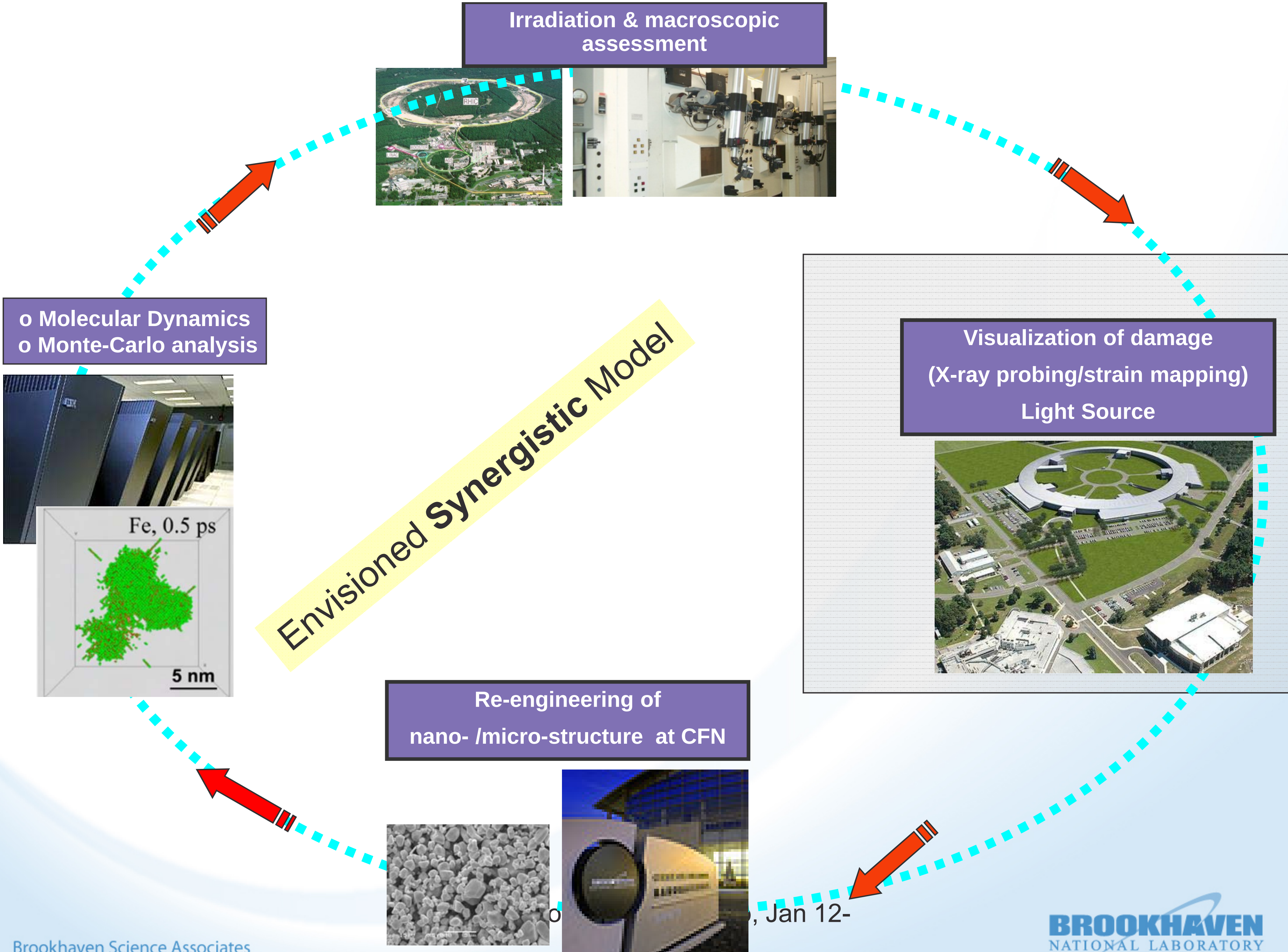
end of displacement cascade ("thermal spike")

Fragment track and **thermal spike** on the surface of fused silica bombarded with energetic particles








Irradiation & macroscopic assessment
RHIC
Visualization of damage
(X-ray probing/strain mapping)
Light Source
Re-engineering of
nano- /micro-structure at CFN
o Molecular Dynamics
o Monte-Carlo analysis
Fe, 0.5 ps
5 nm
Envisioned Synergistic Model