# HiLuMi LHC Material Irradiation Studies at BNL

# STATUS

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**With help from Paola & Elena**

BROOKHAVEN NATIONAL LABORATORY
a passion for discovery
Office of Science
U.S. DEPARTMENT OF ENERGY

# Effort at BNL consists of:

## IRRADIATION Phase I

- 200 MeV proton irradiation at BNL Linear Isotope Producer (BLIP) of 4 candidate materials (Molybdenum, Glidcop, Mo-Graphite and Cu-CD (copper-diamond). Irradiation parameters: 8-weeks at 110 uA
- Neutron irradiation of Cu-CD from 118 MeV protons
- 28-MeV proton irradiation of Mo (including spallation neutron irradiation of Mo, Glidcop, Cu-CD and Mo-GR)

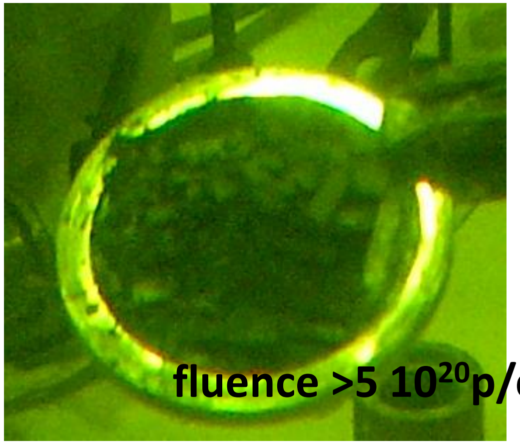
## IRRADIATION Phase II

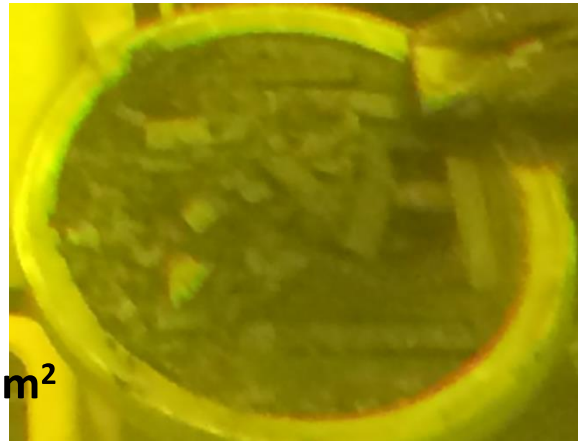
- 200 + 160 MeV proton irradiation at BNL Linear Isotope Producer (BLIP) of 2 newly developed MoGR grades and 2-D C/C composite (CFC)

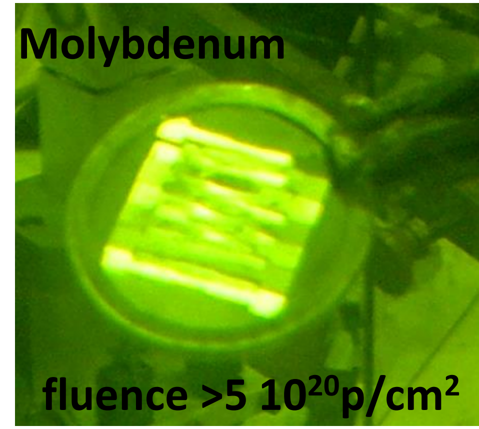
## PIE

- X-ray diffraction studies at NSLS X17B1/X17A beamlines (completed)
- X-ray diffraction studies at NSLS II XPD beamline – Approved Experiment August 2016
- Comprehensive macroscopic analysis at BNL Isotope extraction facility
- Spectral Analysis
- Nano- micro-structural characterization at Center of Functional Nanomaterials

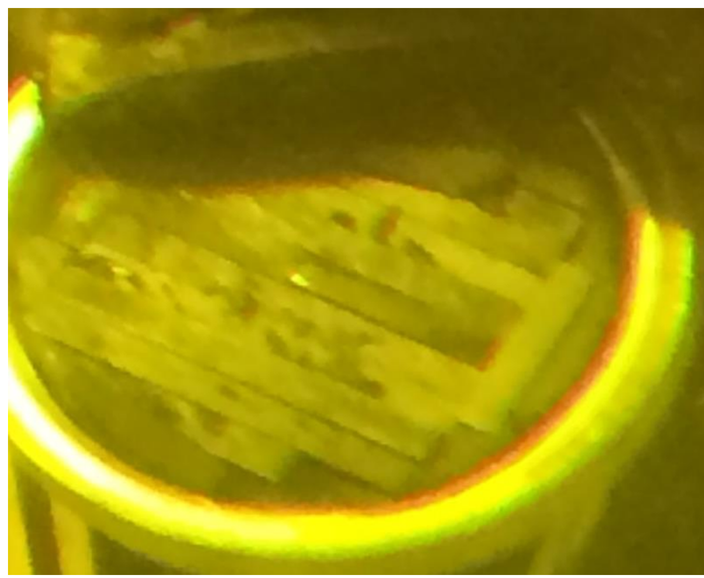
# Primary reason for conducting a follow-up irradiation to2014 RUN

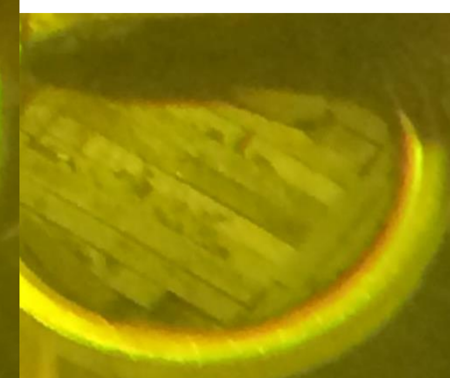


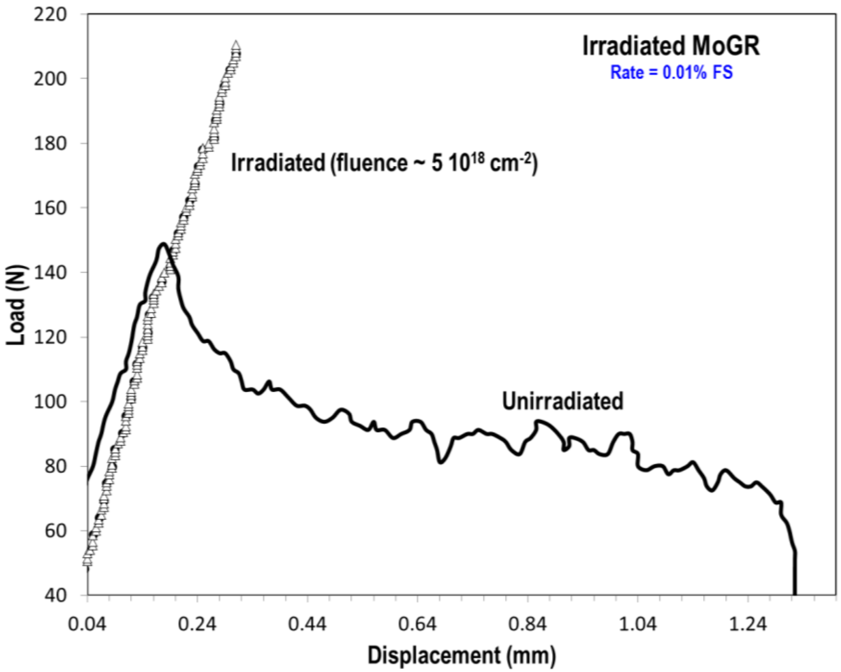



**QUESTION: at what fluence did structural integrity started to be seriously compromised??**

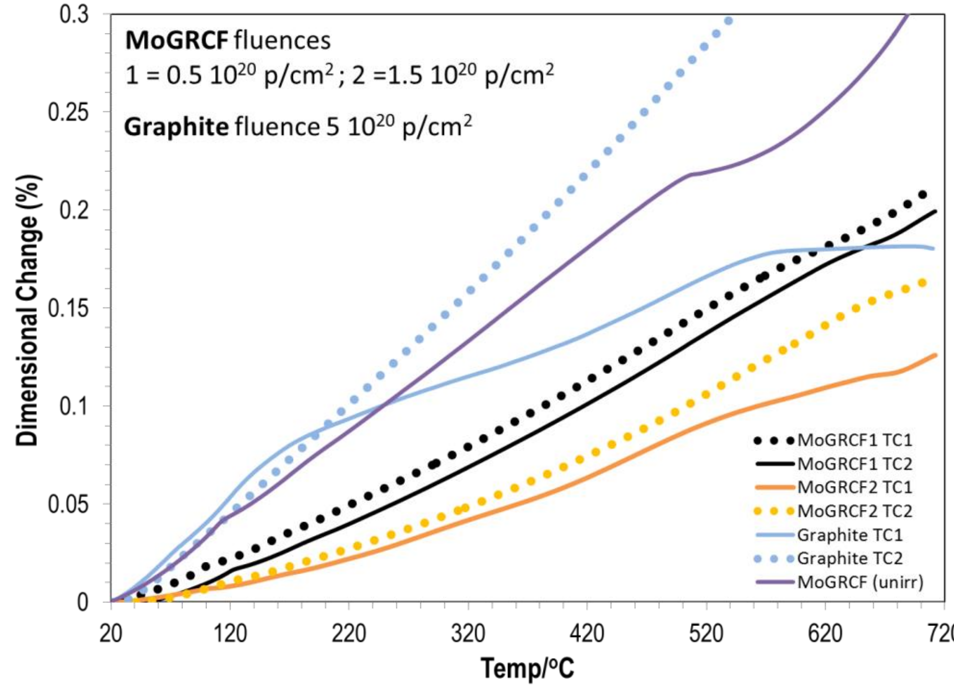




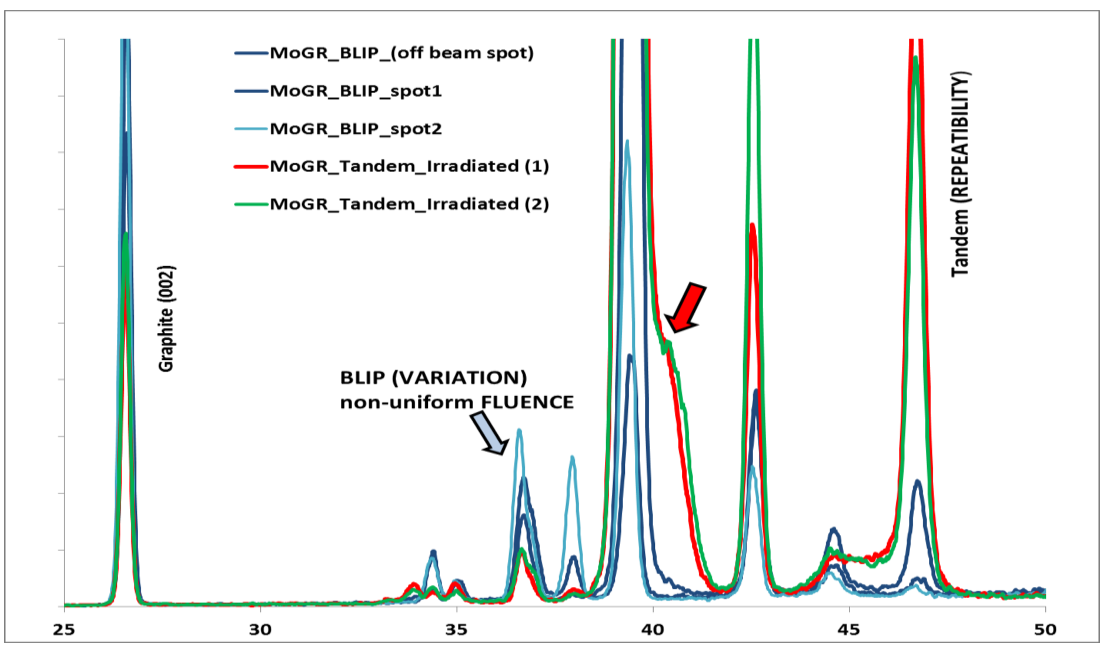
**Shows signs of distress even with minimum beam (fluence ~ 5 $10^{18}$ p/cm$^2$ + 5 $10^{18}$ n/cm$^2$**



Irradiated MoGR
Rate = 0.01% FS
Irradiated (fluence ~ 5 10$^{18}$ cm$^{-2}$)
Unirradiated
Load (N)
Displacement (mm)
220
200
180
160
140
120
100
80
60
40
0.04
0.24
0.44
0.64
0.84
1.04
1.24

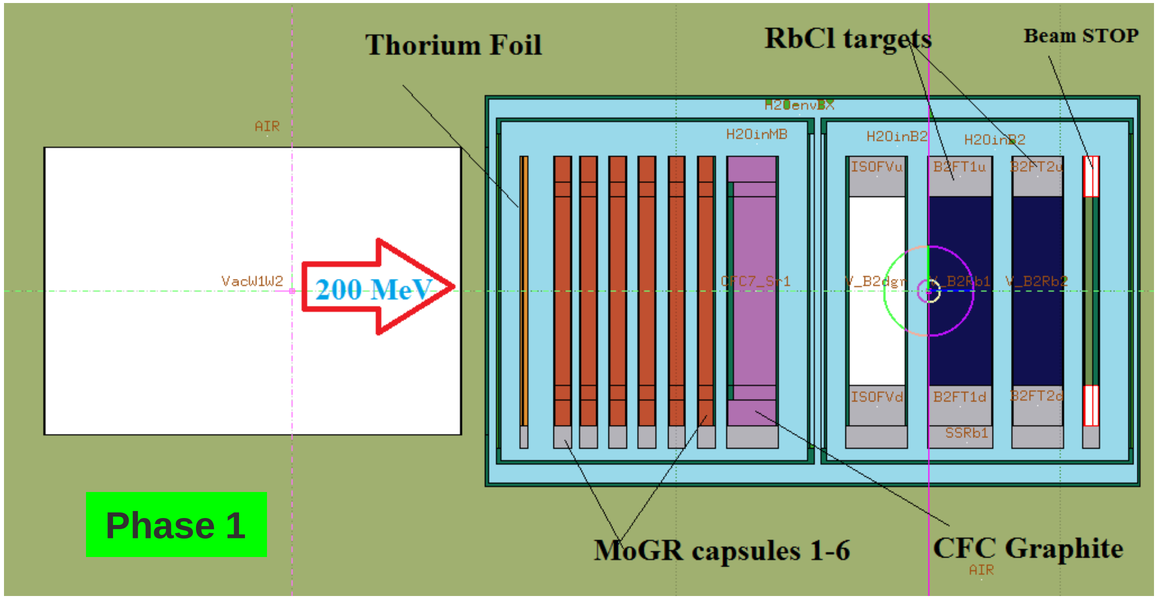



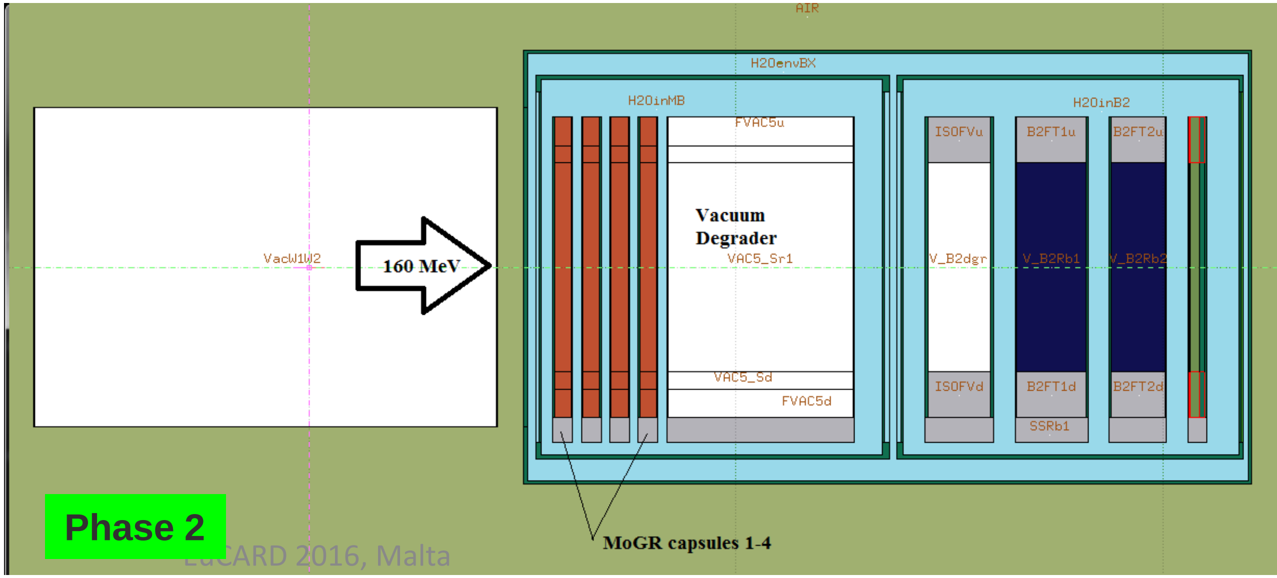
MoGRCF fluences
1 = 0.5 10$^{20}$ p/cm$^2$ ; 2 =1.5 10$^{20}$ p/cm$^2$
Graphite fluence 5 10$^{20}$ p/cm$^2$
Dimensional Change (%)
Temp/$^o$C
0.3
0.25
0.2
0.15
0.1
0.05
0
20
120
220
320
420
520
620
720
MoGRCF1 TC1
MoGRCF1 TC2
MoGRCF2 TC1
MoGRCF2 TC2
Graphite TC1
Graphite TC2
MoGRCF (unirr)

MoGR_BLIP_(off beam spot)
MoGR_BLIP_spot1
MoGR_BLIP_spot2
MoGR_Tandem_Irradiated (1)
MoGR_Tandem_Irradiated (2)
Graphite (002)
BLIP (VARIATION)
non-uniform FLUENCE
Tandem (REPEATIBILITY)
25
30
35
40
45
50

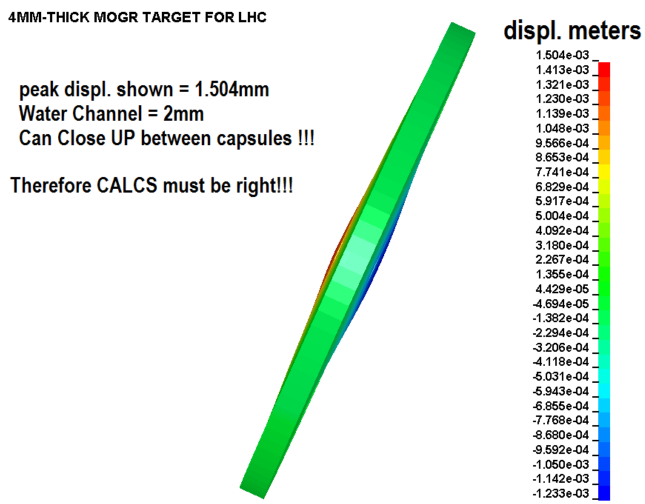
## 2016 RUN
## Explore the fluence region in between
## And for two new MoGR grades (less Mo content)



# Numerous Constraints & Challenges
# Of "parasitic" experiments:

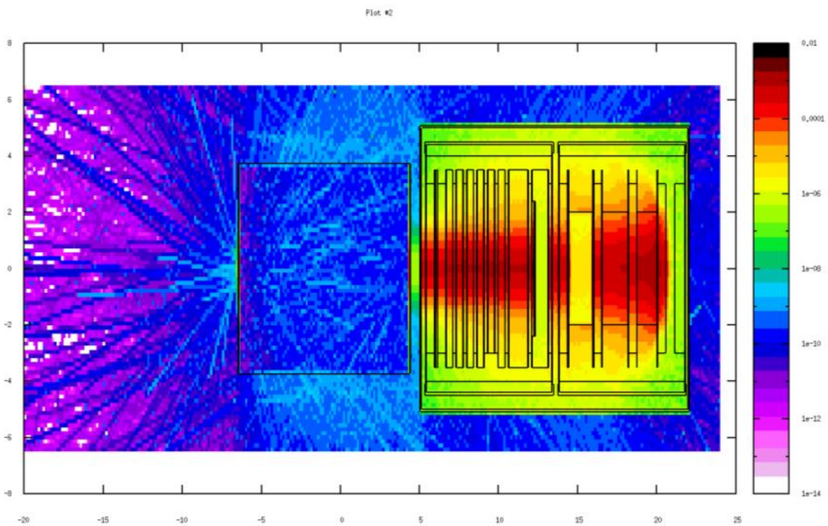
Isotope Production
Precise Energy Budget
Experimental Review Panel Parameters
Thorium Irradiation



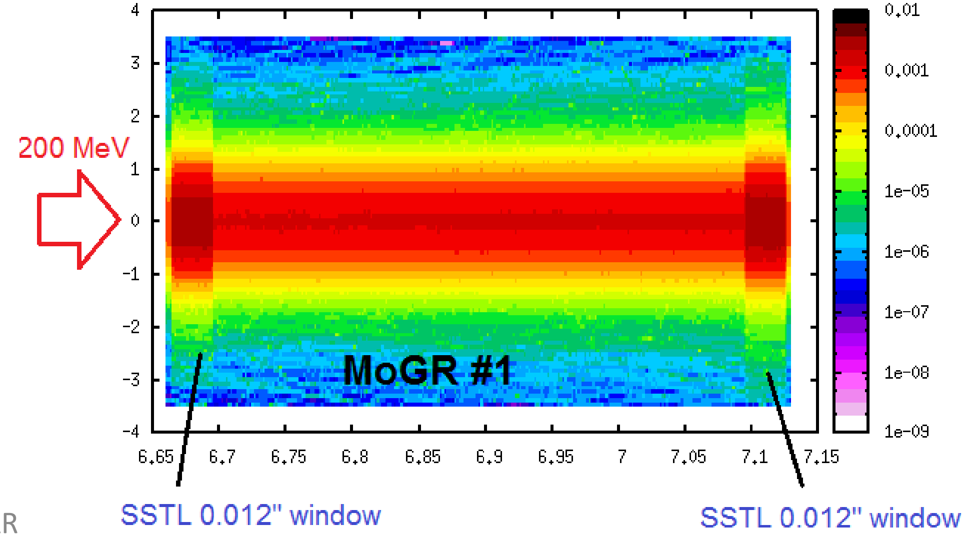

| | | | | | |
|---|---|---|---|---|---|
| CFC Capsule | Stainless Steel<br>CFC Graphite<br>Stainless | 8.0<br>8.0<br>1.67<br>8.0 | 0.012 / 0.3048<br>0.055/1.4<br>0.4567/11.6<br>0.012/0.3048 | 136.31 | 119.20 |
| **Water Gap** | **water** | **1.0** | **0.3102362/7.88** | **119.20** | **114.05** |
| upstream box window | stainless steel | 8.00 | 0.020 / 0.508 | 114.05 | 112.2 |
| **water gap between box windows** | **water** | **1.00** | **0.150 / 3.81** | **112.2** | **109.3** |
| Downstream box front window | stainless steel | 8.00 | 0.020 / 0.508 | 109.3 | 107.4 |
| **Water Gap** | **water** | **1.00** | **0.200 / 5.08** | **107.4** | **103.85** |
| Degrader window | stainless steel | 8.00 | 0.029 / 0.737 | 103.85 | |
| vacuum | vacuum | 0.00 | 0.596/15.14 | | |
| Degrader window | stainless steel | 8.00 | 0.029/0.737 | | 98.42 |
| **Water Gap** | **water** | **8.00** | **0.200 / 5.08** | **98.42** | **94.63** |
| RbCl-1 can window | Inconel | 8.43 | 0.012 / 0.3048 | 94.63 | 93.11 |
| RbCl salt | RbCl | 2.38 | 0.646 / 16.398 | 93.11 | 67.75 |
| can window | Inconel | 8.43 | 0.012 / 0.3048 | 67.75 | 66.47 |
| Water Gap | water | 1.00 | 0.200 / 5.08 | 66.47 | 61.57 |
| RbCl-2 can window | Inconel | 8.43 | 0.012 / 0.3[illegible] | 61.57 | 59.42 |
| RbCl salt | RbCl | 2.38 | 0.500 / 1[illegible]01 | 59.42 | 28.86 |
| RbCl-2 can window | Inconel | 8.43 | 0.012 / 0.3048 | 28.86 | 25.28 |
| cooling channel | water | 1.00 | 0.200 / 5.08 | 25.28 | 15.6 |

# 2016 RUN



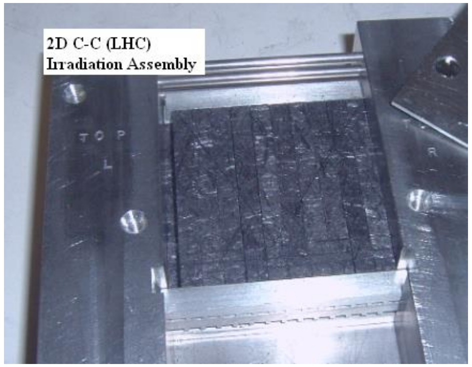


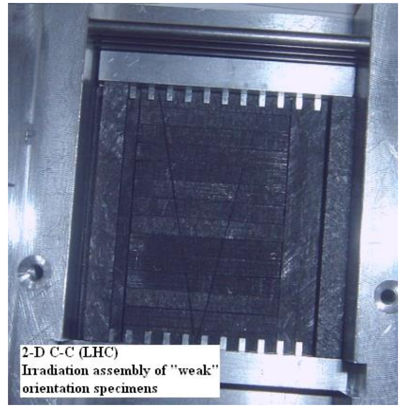
EuCAR

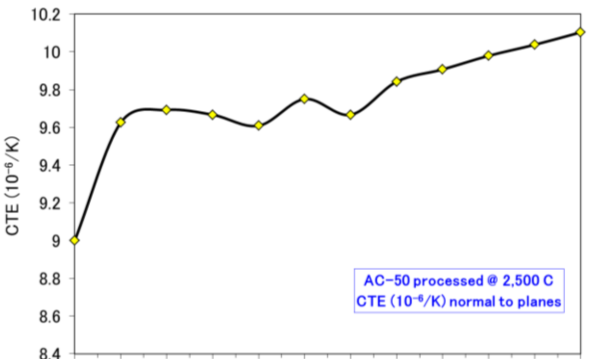


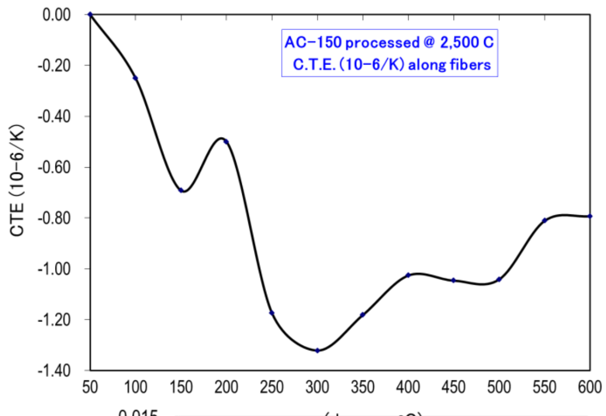
**CFC**

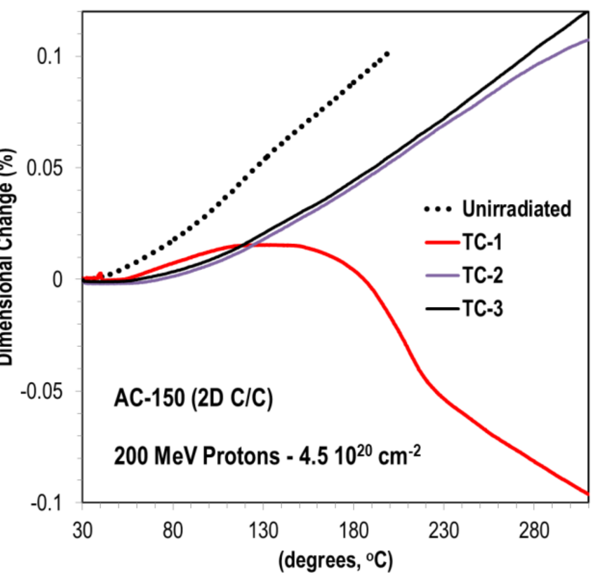
# 2D C/C structure (Toyo-Tanso AC-150)

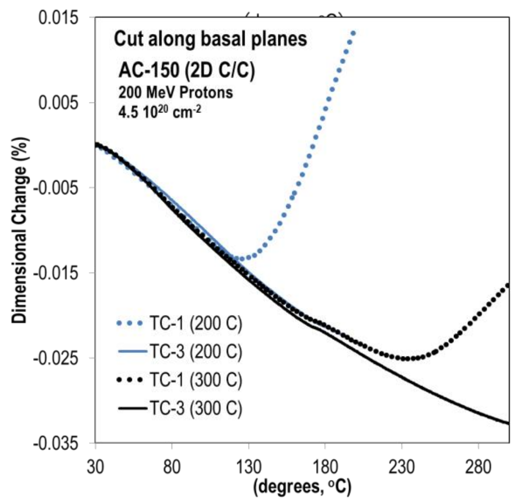


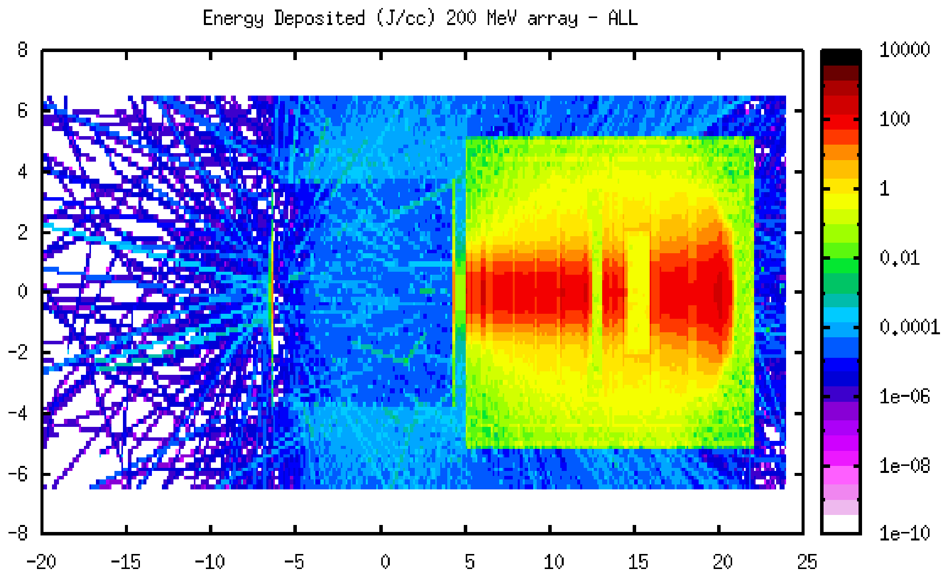



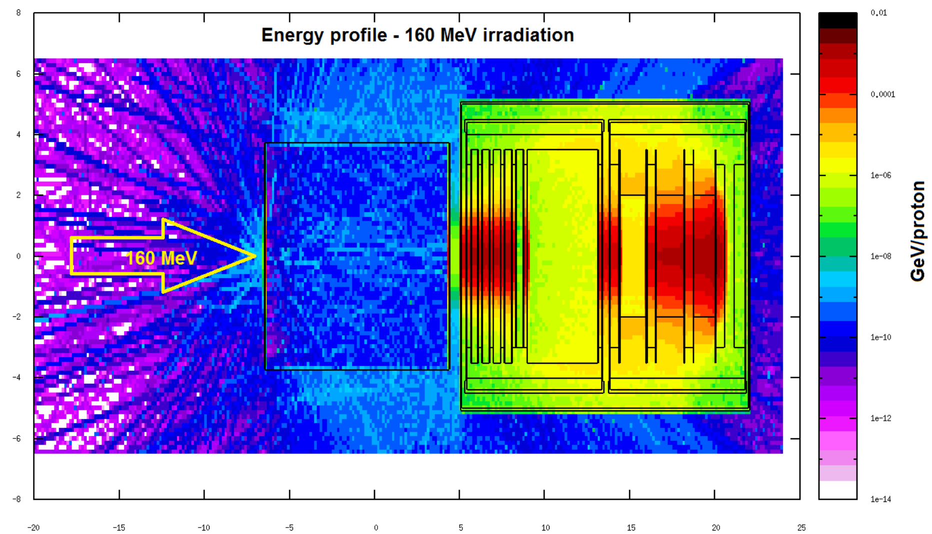



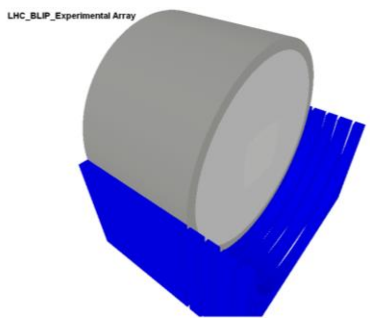



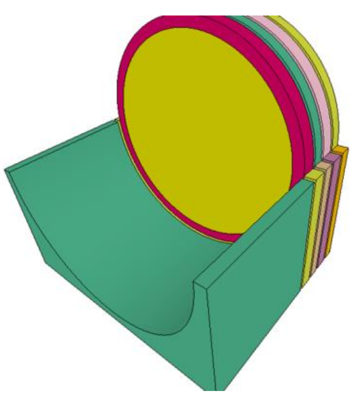



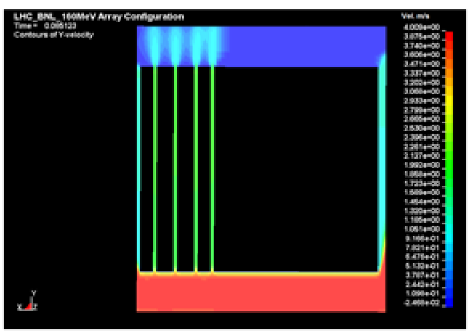



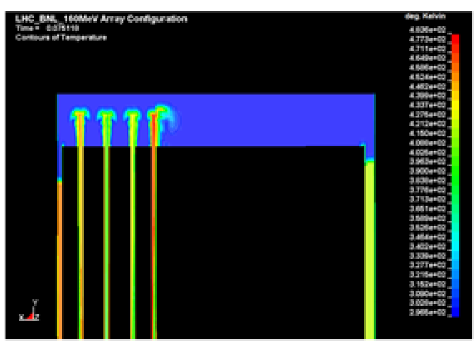


Energy Deposited (J/cc) 200 MeV array - ALL
10000
100
1
0.01
0.0001
1e-06
1e-08
1e-10

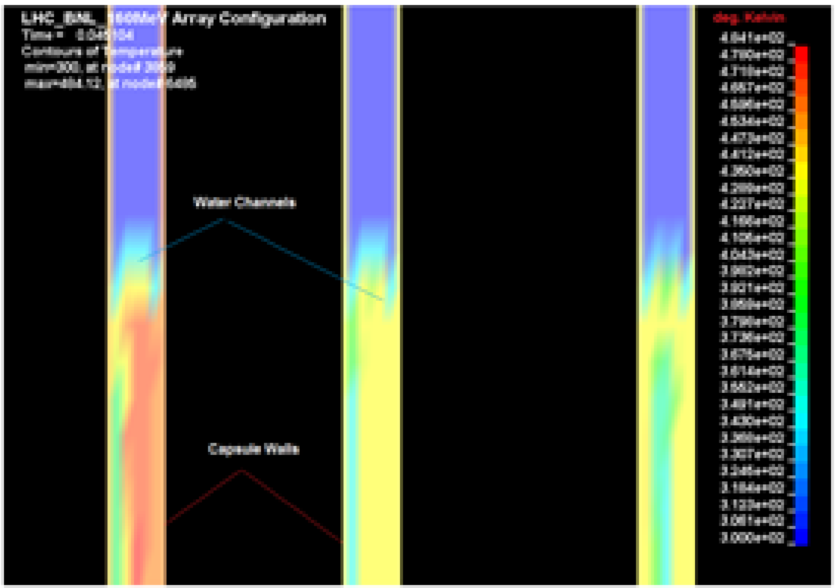

Energy profile - 160 MeV irradiation
160 MeV
GeV/proton


LHC_BLIP_Experimental Array

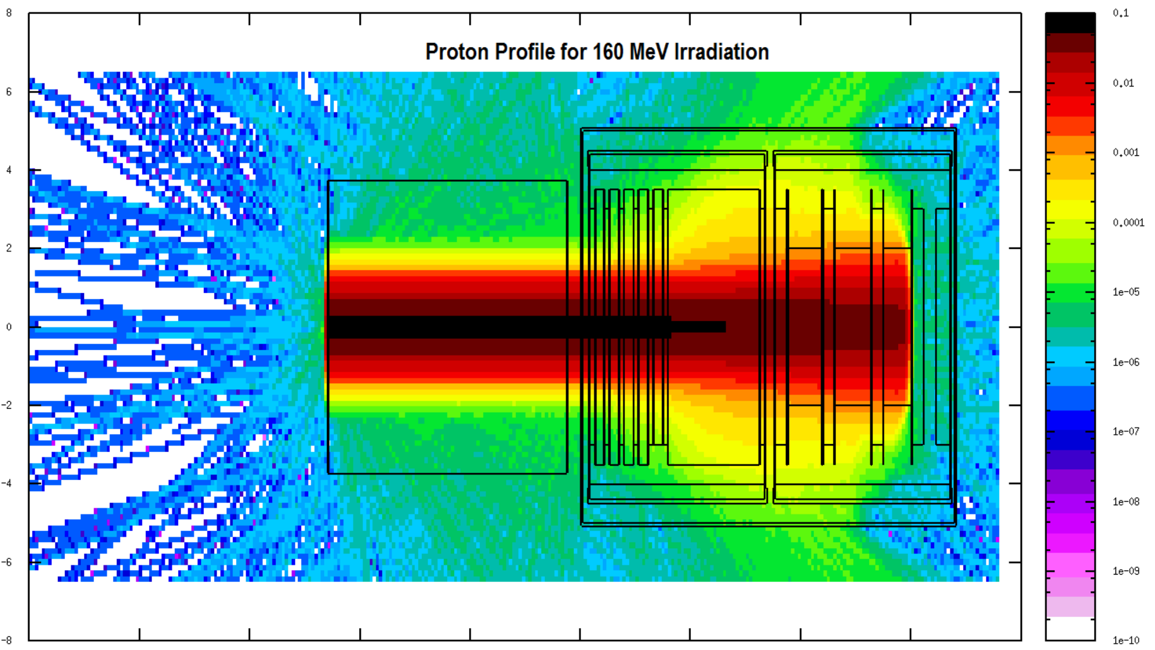



LHC_BNL_160MeV Array Configuration
Contours of Y-velocity

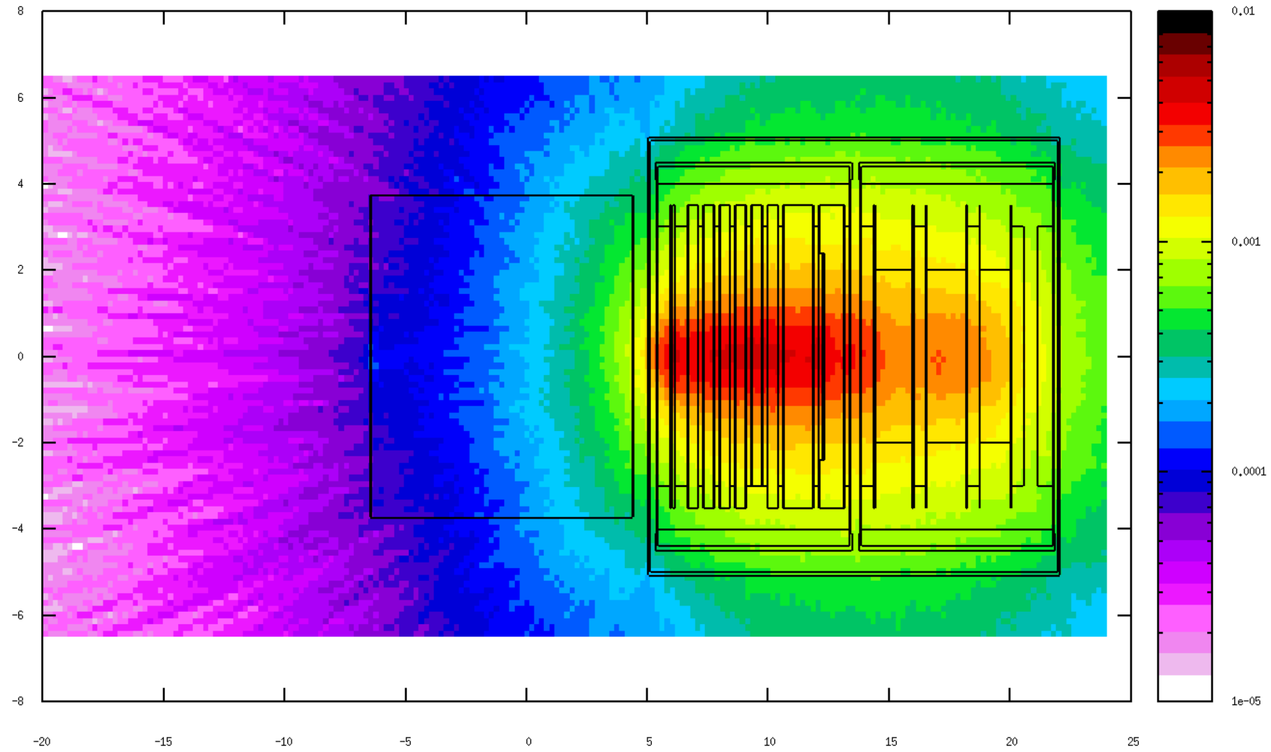

LHC_BNL_160MeV Array Configuration
Contours of Temperature
deg. Kelvin


LHC_BNL_160MeV Array Configuration
Contours of Temperature
deg. Kelvin
Water Channels
Capsule Walls

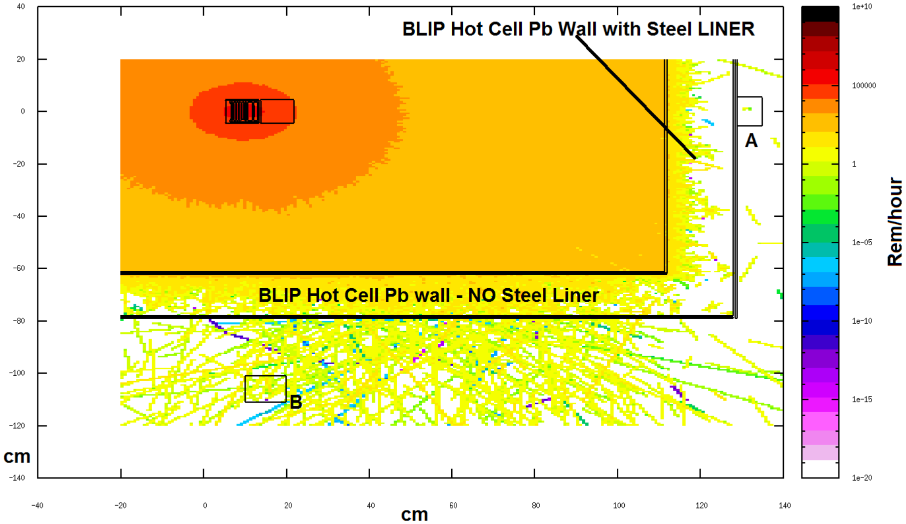
# 2016 RUN

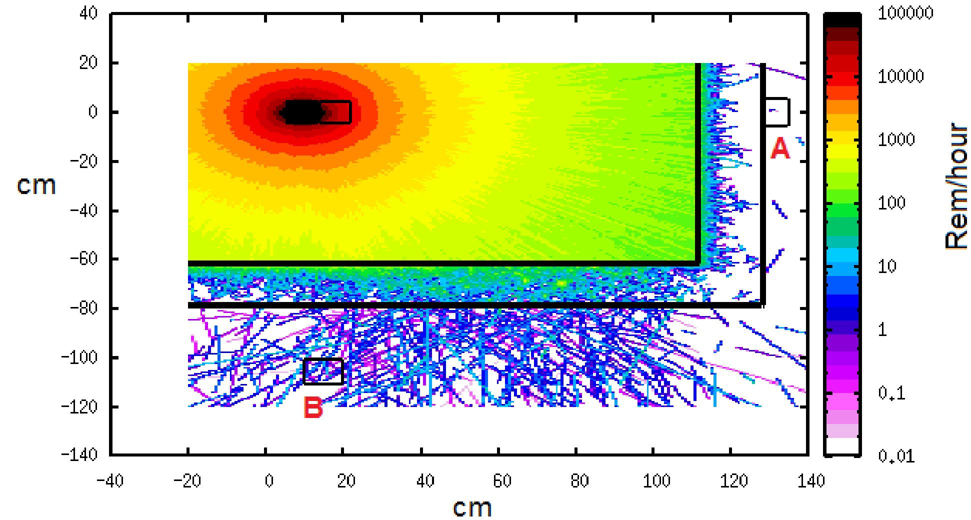


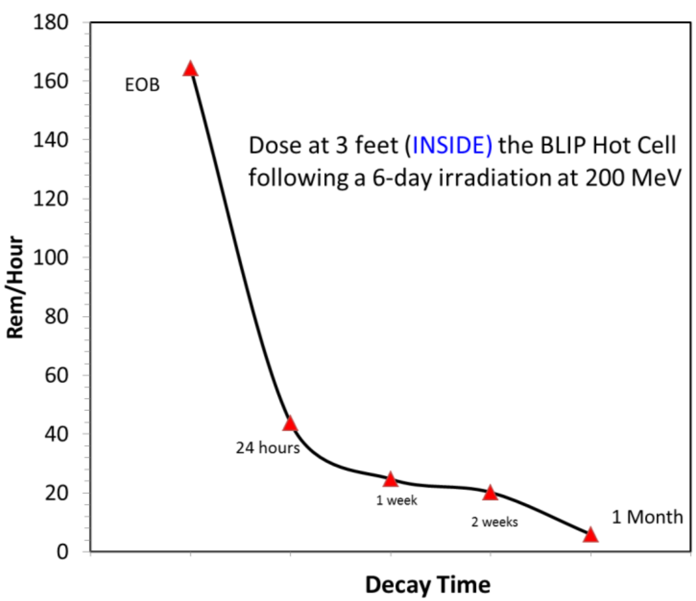
Proton Profile for 160 MeV Irradiation
8
6
4
2
0
-2
-4
-6
-8
0.1
0.01
0.001
0.0001
1e-05
1e-06
1e-07
1e-08
1e-09
1e-10

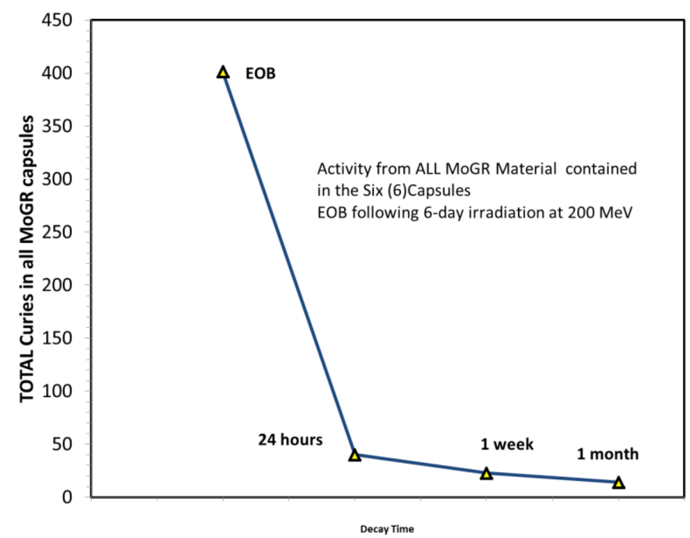



8
6
4
2
0
-2
-4
-6
-8
-20
-15
-10
-5
0
5
10
15
20
25
0.01
0.001
0.0001
1e-05

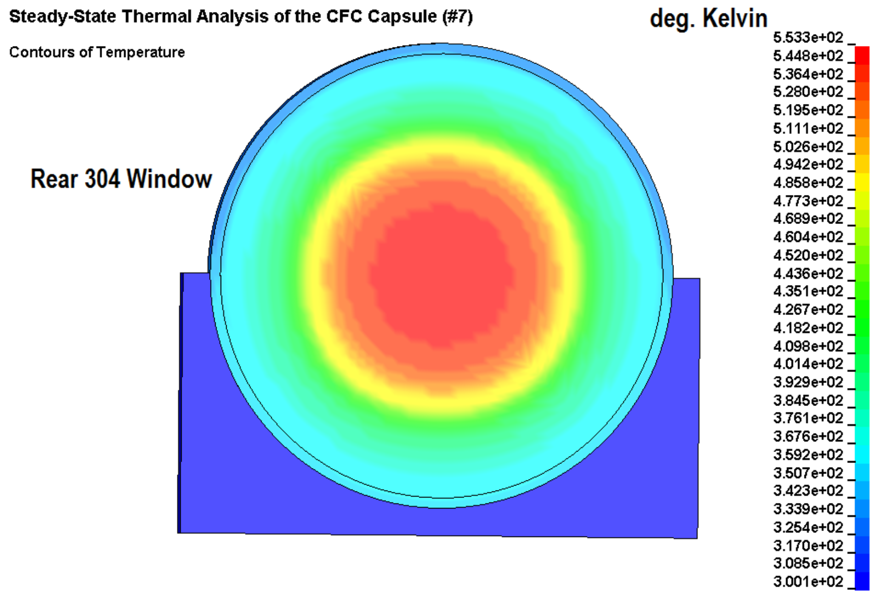

BLIP Hot Cell Pb Wall with Steel LINER
BLIP Hot Cell Pb wall - NO Steel Liner
A
B
cm
cm
Rem/hour


A
B
cm
cm
Rem/hour
100000
10000
1000
100
10
1
0.1
0.01


Dose at 3 feet (INSIDE) the BLIP Hot Cell
following a 6-day irradiation at 200 MeV
EOB
24 hours
1 week
2 weeks
1 Month
Rem/Hour
Decay Time


Activity from ALL MoGR Material contained
in the Six (6)Capsules
EOB following 6-day irradiation at 200 MeV
EOB
24 hours
1 week
1 month
TOTAL Curies in all MoGR capsules
Decay Time

# 2016 RUN



Steady-State Thermal Analysis of the CFC Capsule (#7)
Contours of Temperature
Rear 304 Window
deg. Kelvin
5.533e+02
5.448e+02
5.364e+02
5.280e+02
5.195e+02
5.111e+02
5.026e+02
4.942e+02
4.858e+02
4.773e+02
4.689e+02
4.604e+02
4.520e+02
4.436e+02
4.351e+02
4.267e+02
4.182e+02
4.098e+02
4.014e+02
3.929e+02
3.845e+02
3.761e+02
3.676e+02
3.592e+02
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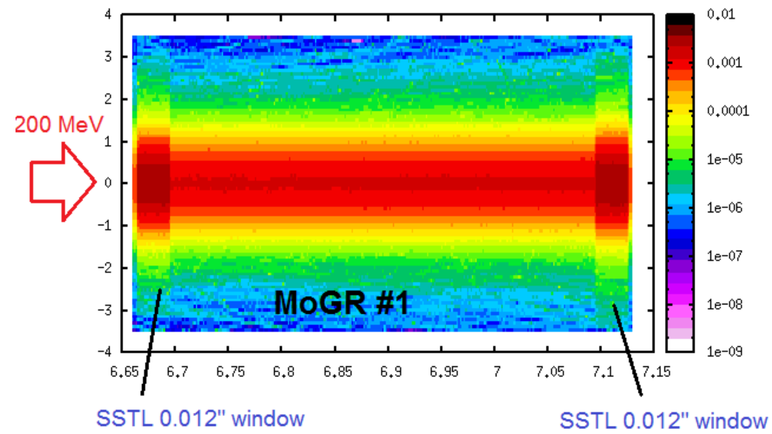
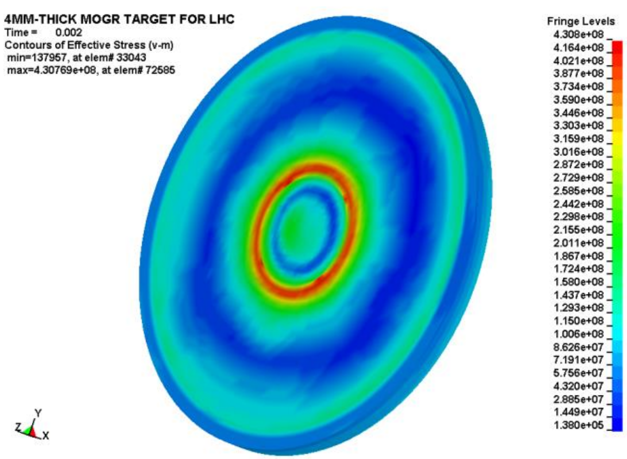
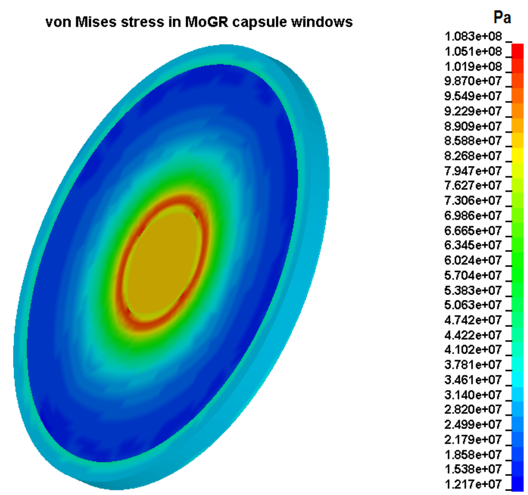
3.254e+02
3.170e+02
3.085e+02
3.001e+02

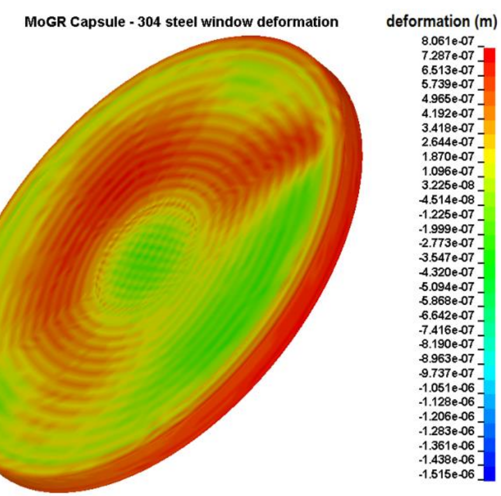
200 MeV
MoGR #1
4
3
2
1
0
-1
-2
-3
-4
6.65
6.7
6.75
6.8
6.85
6.9
6.95
7
7.05
7.1
7.15
0.01
0.001
0.0001
1e-05
1e-06
1e-07
1e-08
1e-09
SSTL 0.012" window
SSTL 0.012" window

4MM-THICK MOGR TARGET FOR LHC
Time = 0.002
Contours of Effective Stress (v-m)
min=137957, at elem# 33043
max=4.30769e+08, at elem# 72585
Fringe Levels
4.308e+08
4.164e+08
4.021e+08
3.877e+08
3.734e+08
3.590e+08
3.446e+08
3.303e+08
3.159e+08
3.016e+08
2.872e+08
2.729e+08
2.585e+08
2.442e+08
2.298e+08
2.155e+08
2.011e+08
1.867e+08
1.724e+08
1.580e+08
1.437e+08
1.293e+08
1.150e+08
1.006e+08
8.626e+07
7.191e+07
5.756e+07
4.320e+07
2.885e+07
1.449e+07
1.380e+05
Y
Z
X

von Mises stress in MoGR capsule windows
Pa
1.083e+08
1.051e+08
1.019e+08
9.870e+07
9.549e+07
9.229e+07
8.909e+07
8.588e+07
8.268e+07
7.947e+07
7.627e+07
7.306e+07
6.986e+07
6.665e+07
6.345e+07
6.024e+07
5.704e+07
5.383e+07
5.063e+07
4.742e+07
4.422e+07
4.102e+07
3.781e+07
3.461e+07
3.140e+07
2.820e+07
2.499e+07
2.179e+07
1.858e+07
1.538e+07
1.217e+07

MoGR Capsule - 304 steel window deformation
deformation (m)
8.061e-07
7.287e-07
6.513e-07
5.739e-07
4.965e-07
4.192e-07
3.418e-07
2.644e-07
1.870e-07
1.096e-07
3.225e-08
-4.514e-08
-1.225e-07
-1.999e-07
-2.773e-07
-3.547e-07
-4.320e-07
-5.094e-07
-5.868e-07
-6.642e-07
-7.416e-07
-8.190e-07
-8.963e-07
-9.737e-07
-1.051e-06
-1.128e-06
-1.206e-06
-1.283e-06
-1.361e-06
-1.438e-06
-1.515e-06

**2016 EXPERIMENT Phase 1 (200 MeV) was approved and was launched**

1$^{st}$ part of the 200 MeV (along Thorium) was completed last week.

**Experiment was a success.**

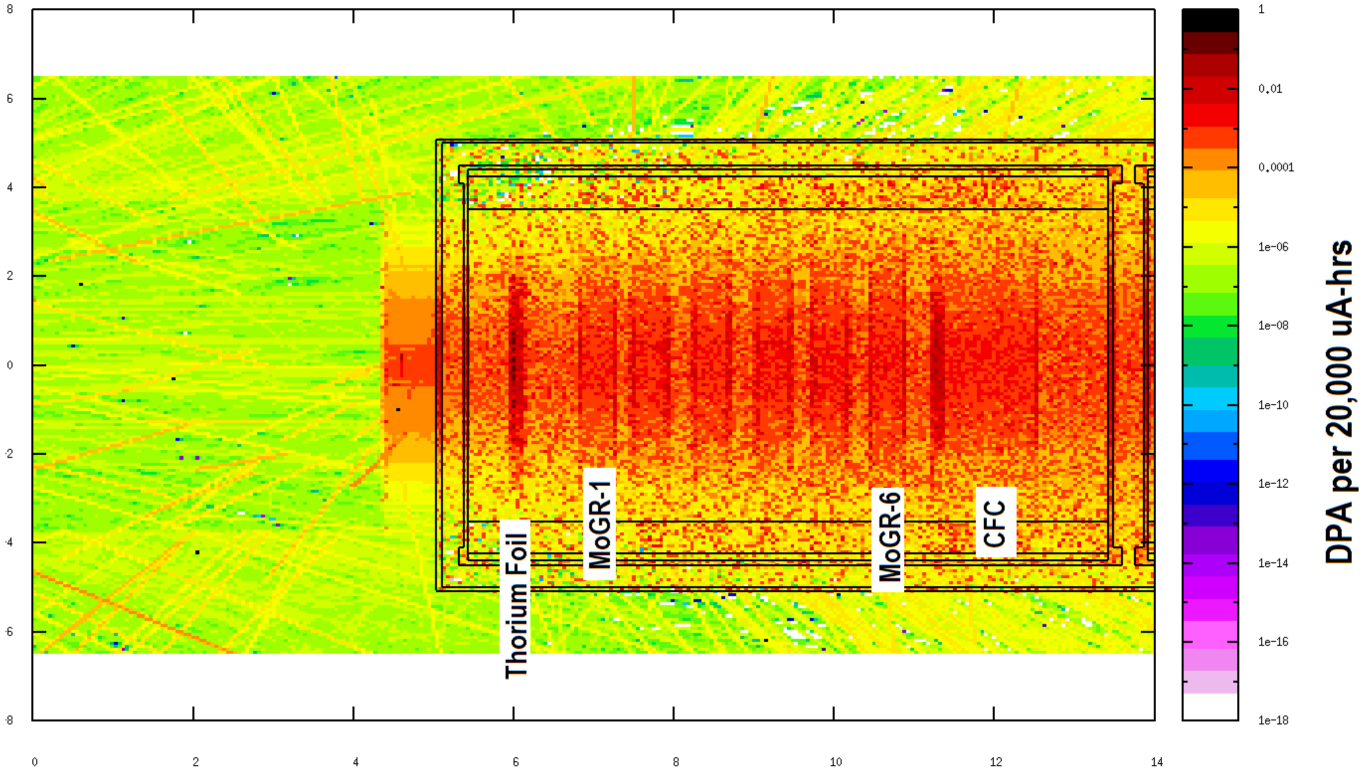
Beam Current was significantly higher (peak of 265 uA) than the 2014 campaign (ave. 105 uA)

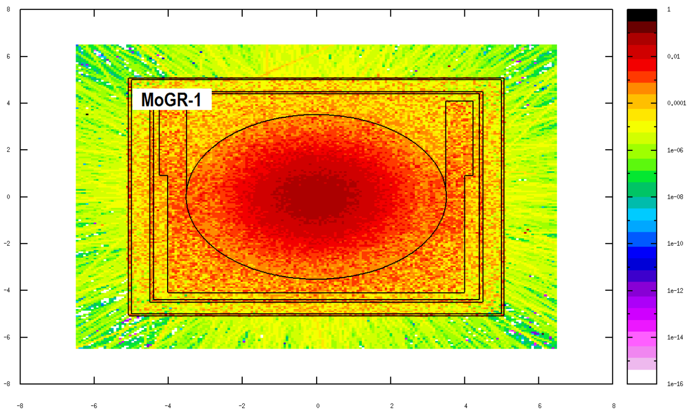
Approximate integrated current 20,000 uA-hours

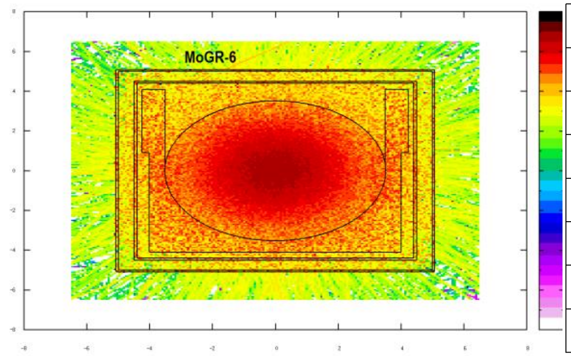
This translates to a fluence of ~ 7 $10^{19}$ p/cm$^2$

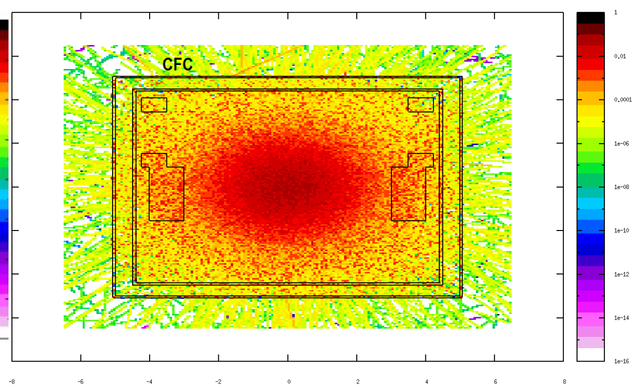
Target array is waiting inside the BLIP Hot Cell for next 200 MeV run along Thorium (dates TBD)

# 2016 RUN


Thorium Foil
MoGR-1
MoGR-6
CFC
DPA per 20,000 uA-hrs


MoGR-1


MoGR-6


CFC

## 2016 EXPERIMENT Phase – NEXT STEP

Complete 2nd part of the 200 MeV (along Thorium) – Approved (Date TBD)

**Some Concerns:**

Linac Experienced sparking at the end of run in Tank #6 which is needed to get the energy to 140 and above.

Current was lowered at the last day and sparking stopped

Decision was made by the Collider to not fix it during this year's run and disrupt Isotope Production (several days of downtime)

The Thorium + LHC 200 MeV run will go on as scheduled but at the level of current where no sparking is experienced (130-150 uA beam)

Running beyond that at approved 160 MeV will depend on two factors:

(a) State of MoGR (and CFC) after an additional 20,000 uA of beam (may render further radiation damage un-necessary)
(b) Provided the new MoGR grades survived, risk Isotope Production is willing to take in increasing the chance of blowing the key accelerator tank by operating at a higher energy than they need (117 MeV and tanks 1-5)

**2016 EXPERIMENT Phase – Optimizing the OUTPUT**

**STEP -1:**

**Retrieve 2 MoGR capsules NOW and replace them with identical MoGR for the next 200 MeV run (one of each grade)**

**This provides for examining the 2 grades at the 7 $10^{19}$ p/cm$^2$**

Structural state (signs of physical degradation)

Physical properties and mechanical behavior

Cool down and prepping for X-ray Diffraction (proposal approved for mid-August – but activation conditions stricter at NSLS II

**STEP-2:**

**Complete additional 200 MeV irradiation – Retrieve a pair that would have seen 40,000 uA-hrs and examine structural integrity, etc.**

**STEP -3:**

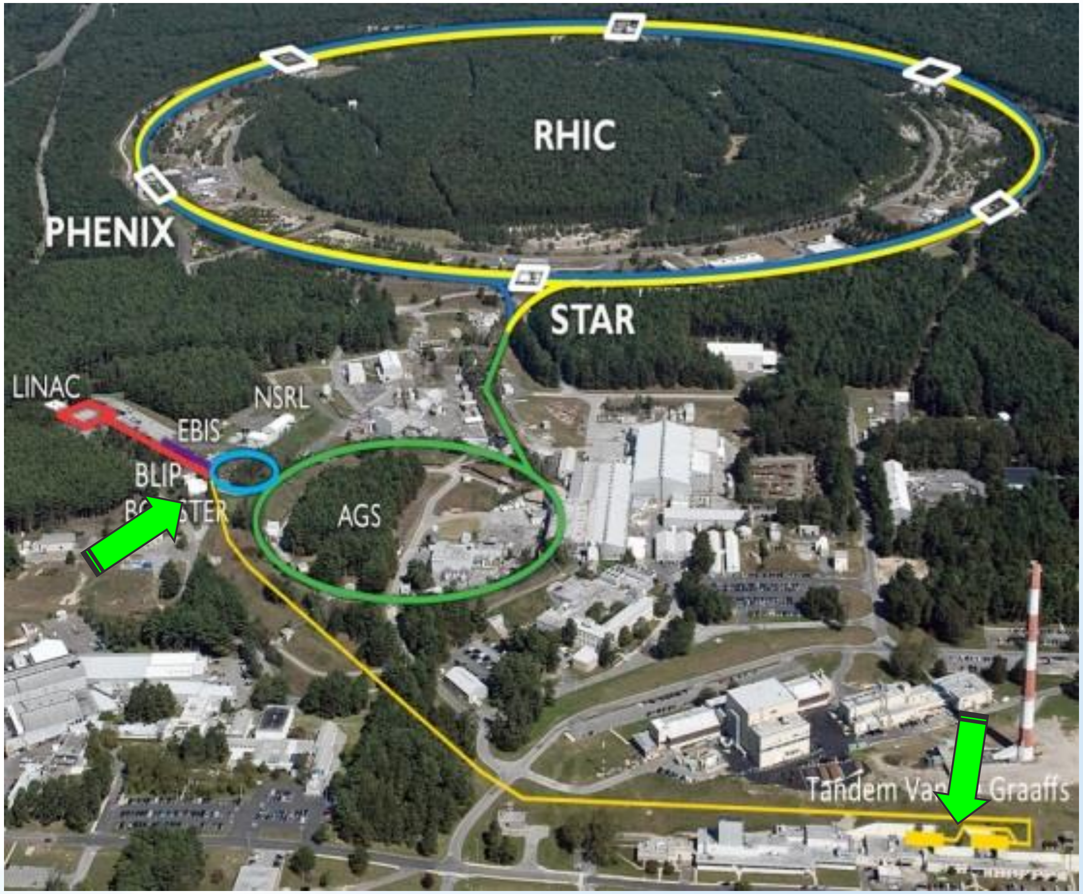
**Based on assessment of material state and the Isotope Program decision on risk taking switch to 160 MeV configuration and irradiate for another interval**

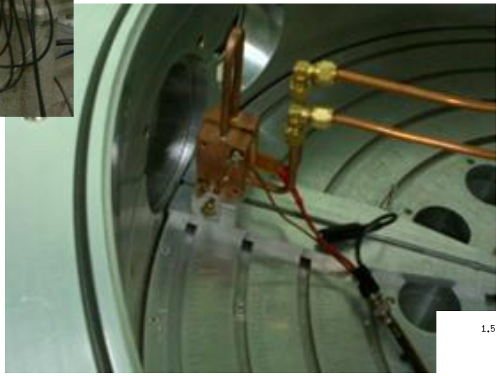
# EXCERPT Results from PIE of 2014 Irradiation Campaign

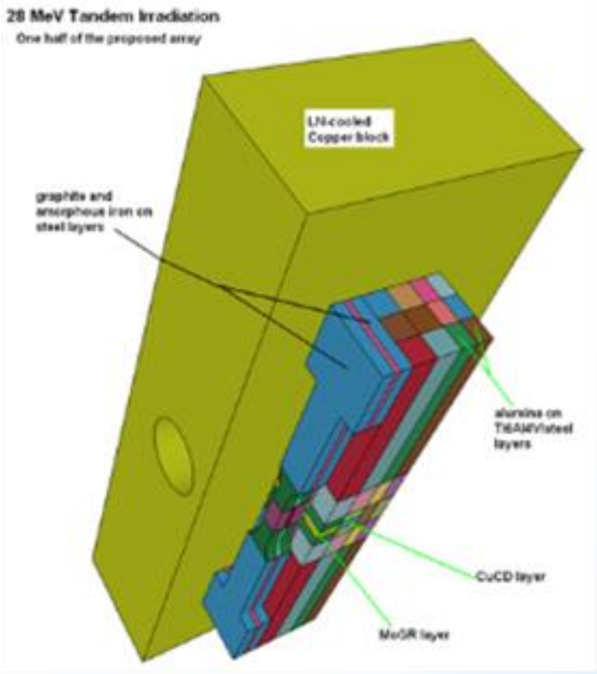
**Irradiations took place at BLIP (181 MeV) and Tandem (28 MeV)**

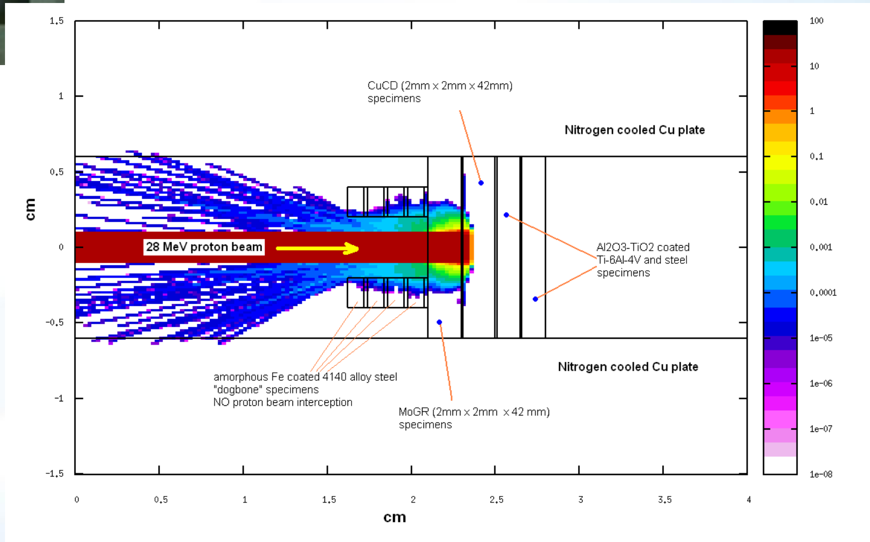
**At elevated temperatures a BLIP (MoGR ~420C; Mo ~1100 C)**
**At sub-zero temperatures at Tandem (spallation field from 28 MeV on Mo)**

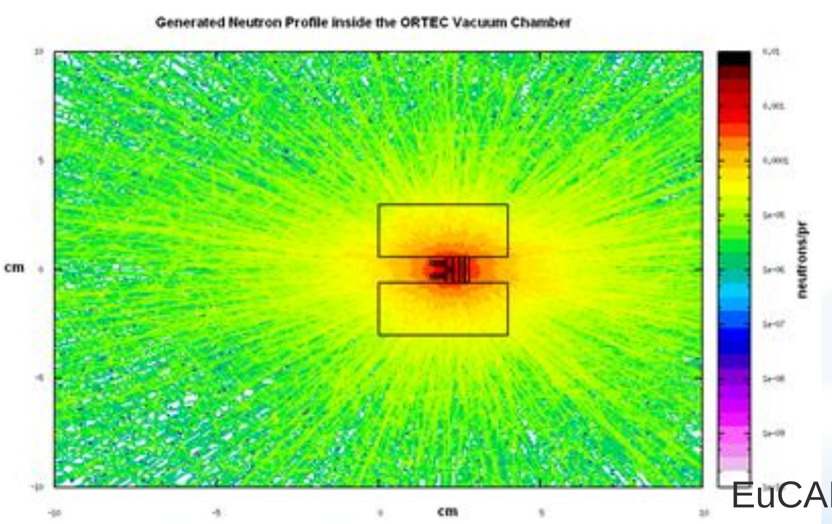
**MoGR also irradiated with protons + spallation neutrons to ~ (5+5) $10^{18}$**



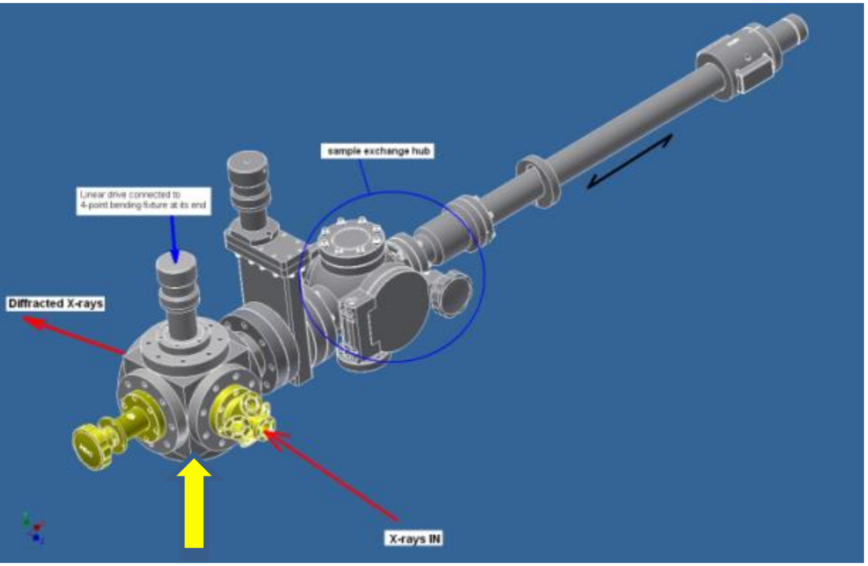


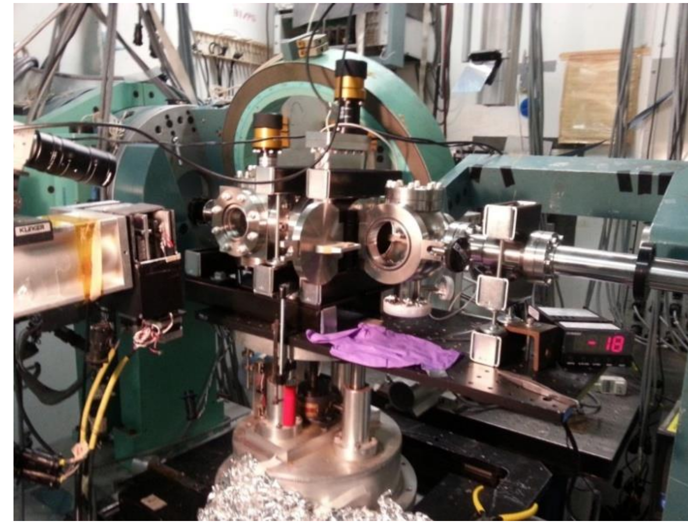

28 MeV Tandem Irradiation
One half of the proposed array
LN-cooled Copper block
graphite and amorphous iron on steel layers
alumina on Ti6Al4V/steel layers
CuCD layer
MoGR layer

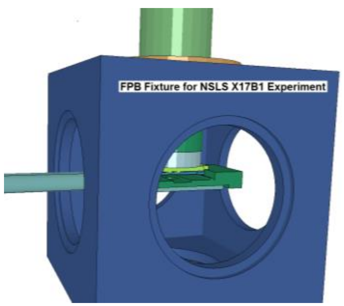

CuCD (2mm x 2mm x 42mm) specimens
Nitrogen cooled Cu plate
28 MeV proton beam
Al2O3-TiO2 coated Ti-6Al-4V and steel specimens
amorphous Fe coated 4140 alloy steel "dogbone" specimens NO proton beam interception
MoGR (2mm x 2mm x 42 mm) specimens
Nitrogen cooled Cu plate
cm
cm

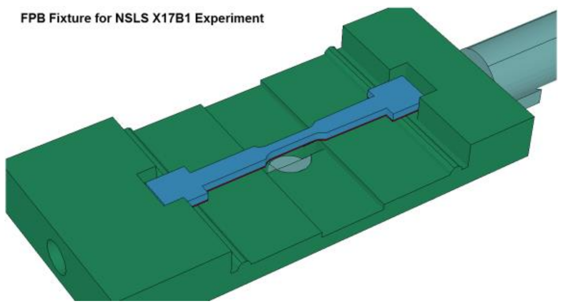

Generated Neutron Profile inside the ORTEC Vacuum Chamber
cm
cm
neutrons/pr

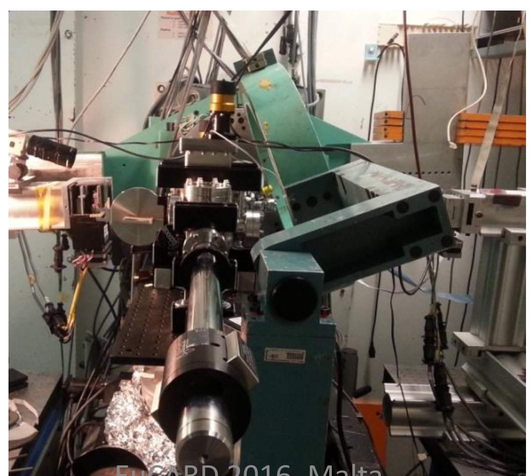
# EDXRD at NSLS

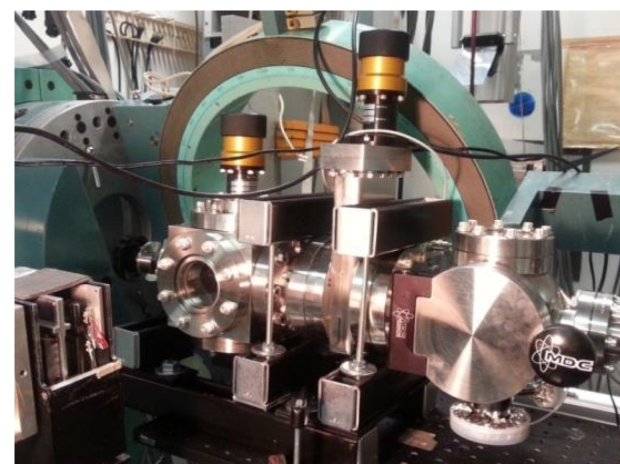




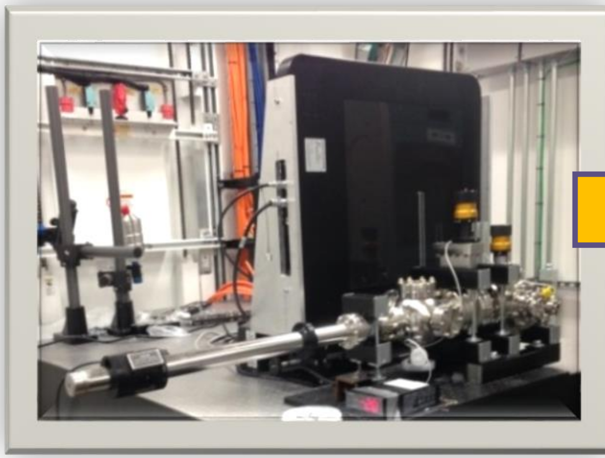



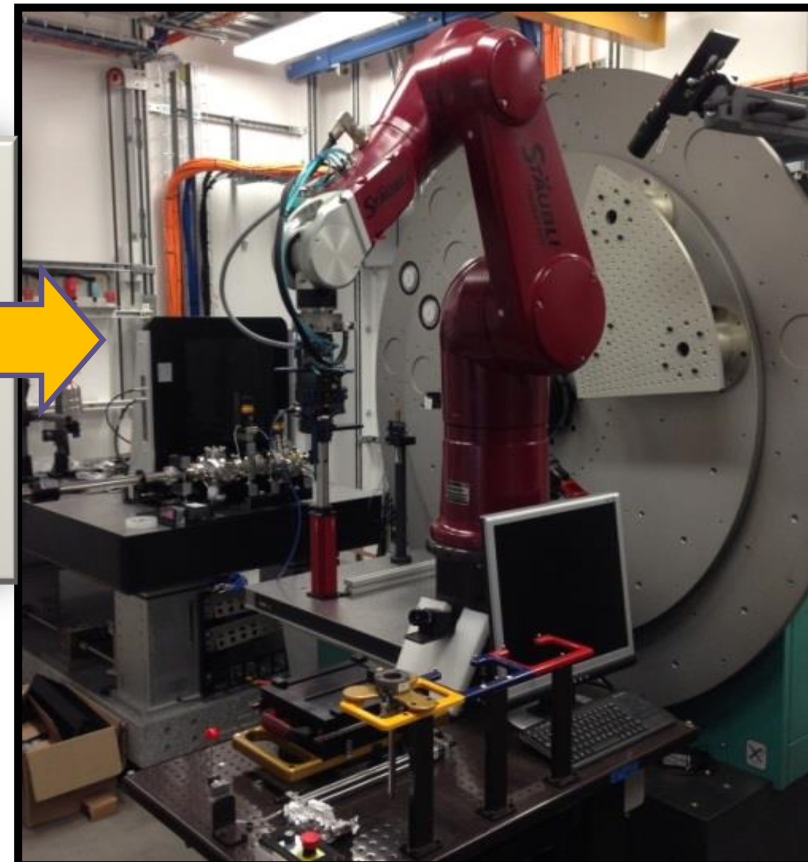



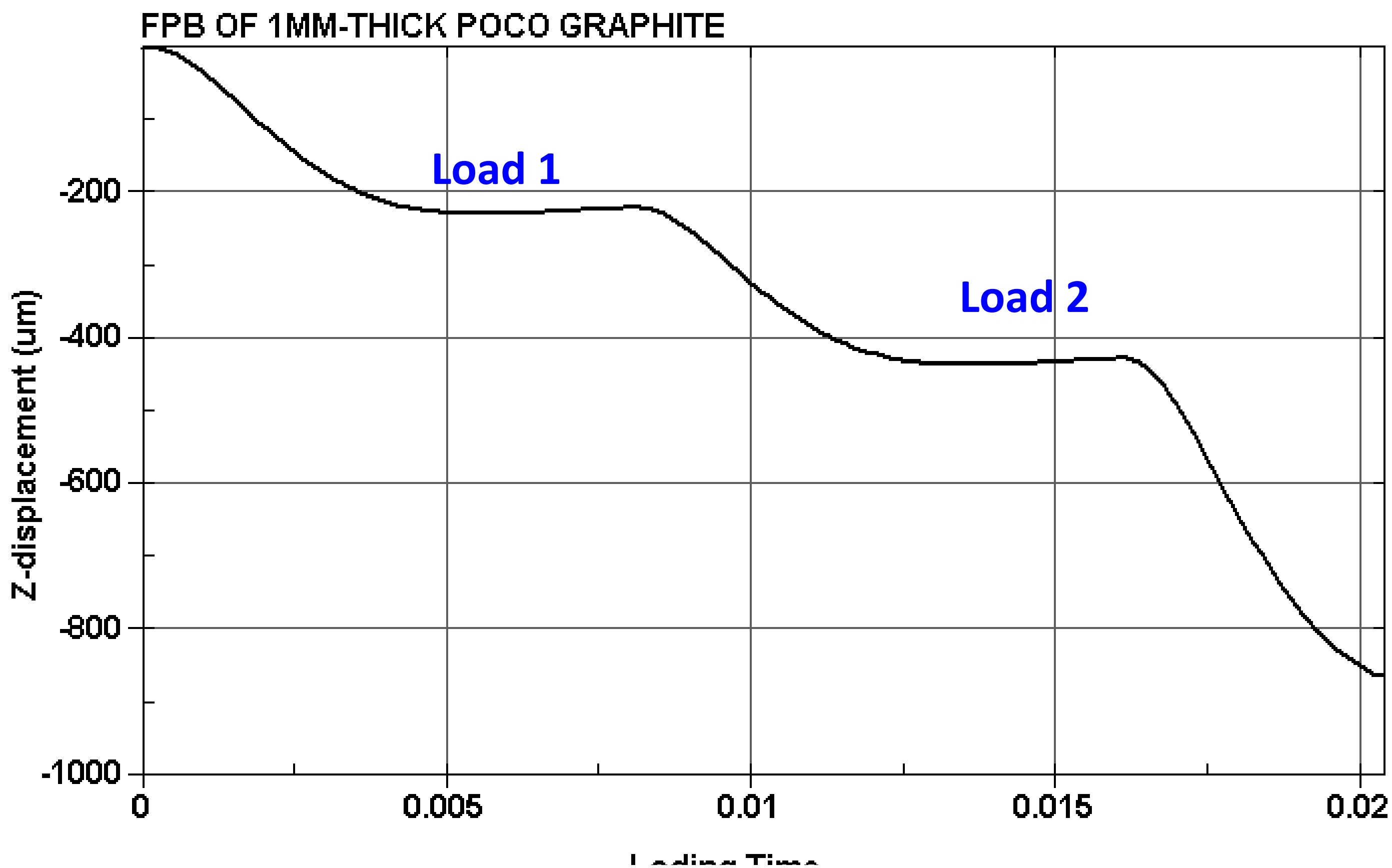


# XRD at NSLS (70 keV at X17A Beamline

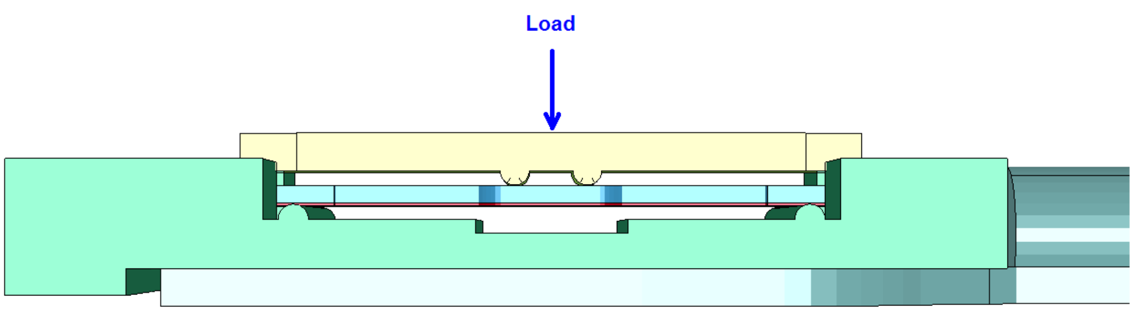
# Upcoming → XRD at XPD Beamline of NSLS II





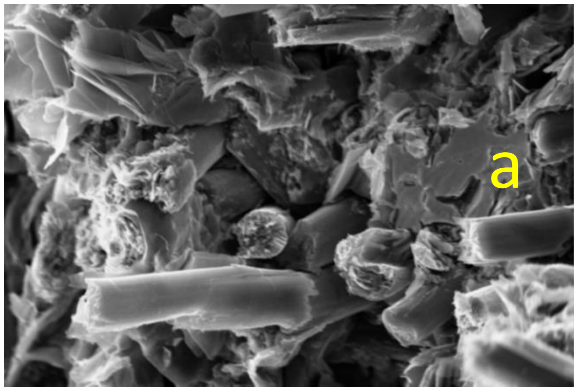


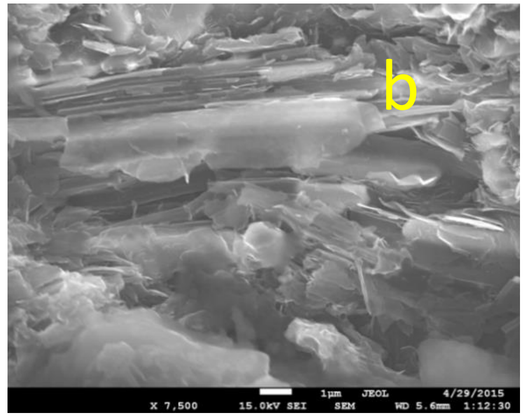

**FPB Fixture for NSLS X17B1 Experiment**

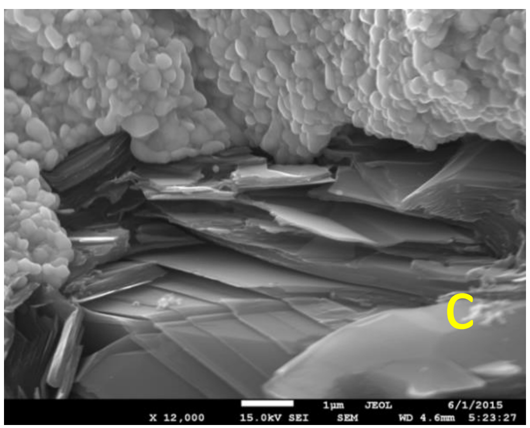


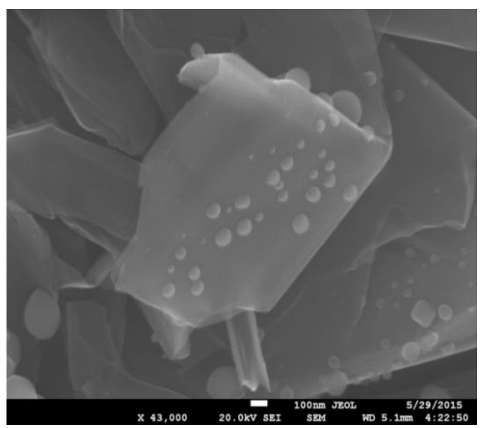

**GOAL: LATTICE strain and global strain are very different in magnitude**

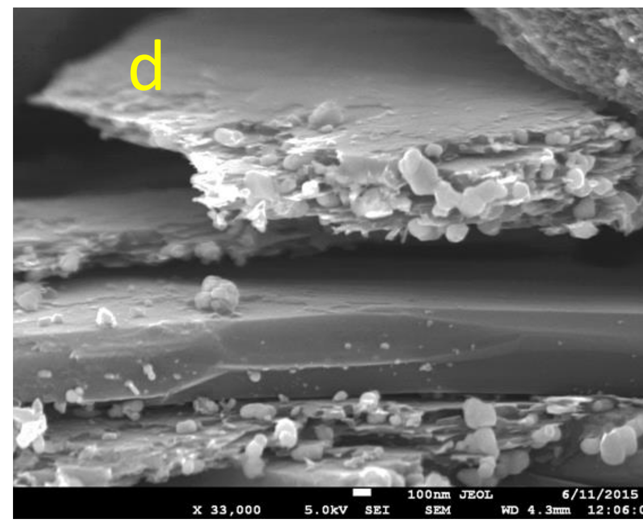
# Micro Evaluation of MoGR (original grade)
# How close to graphite behavior?



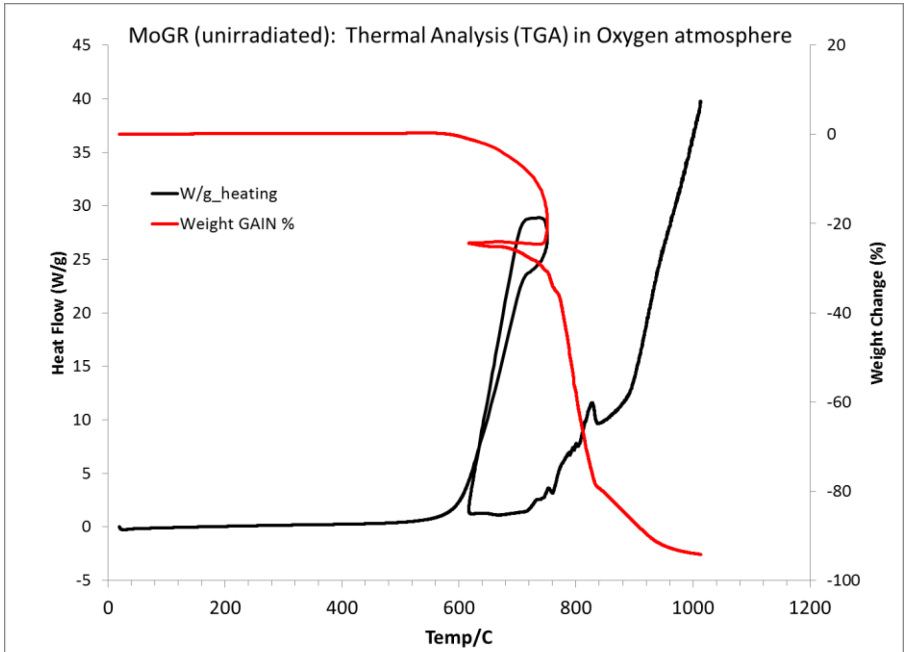



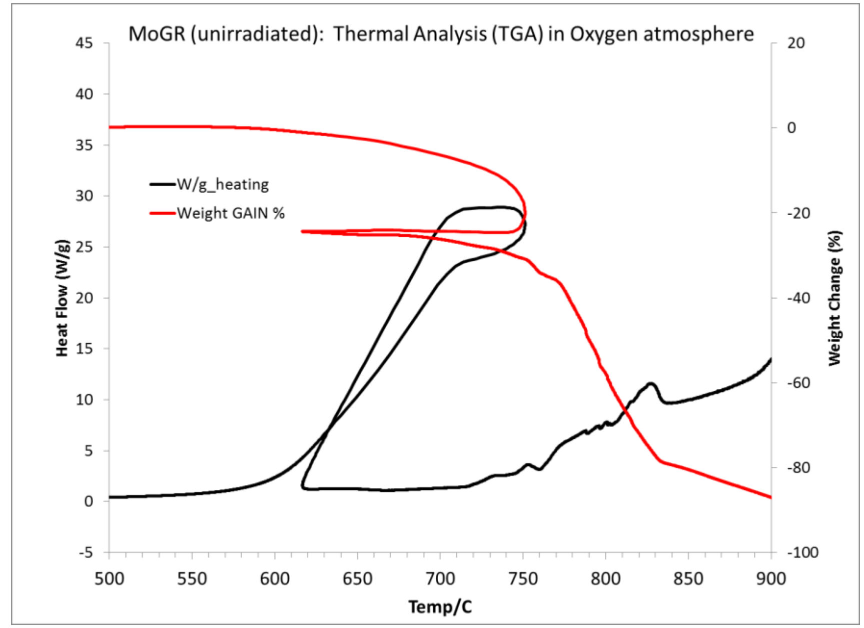



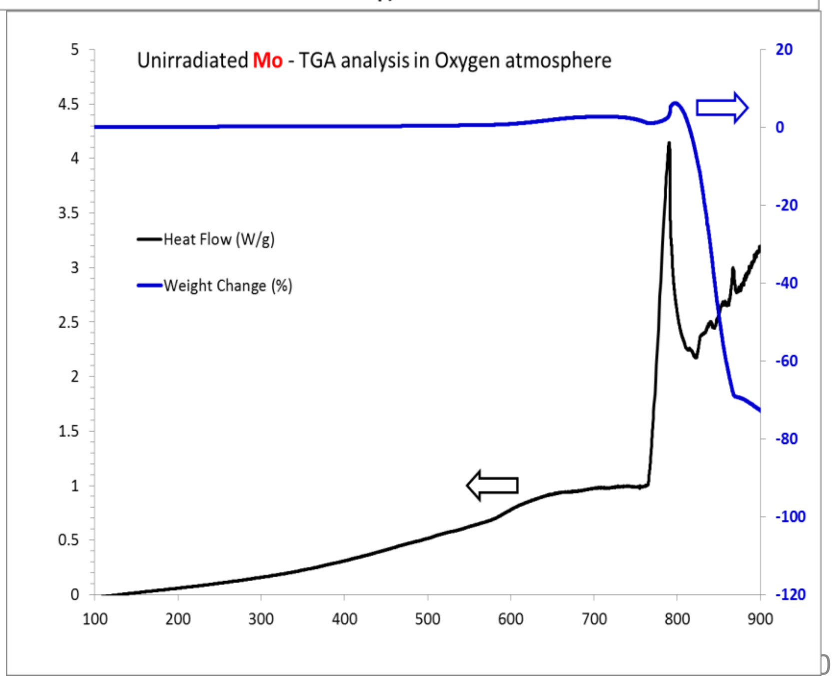



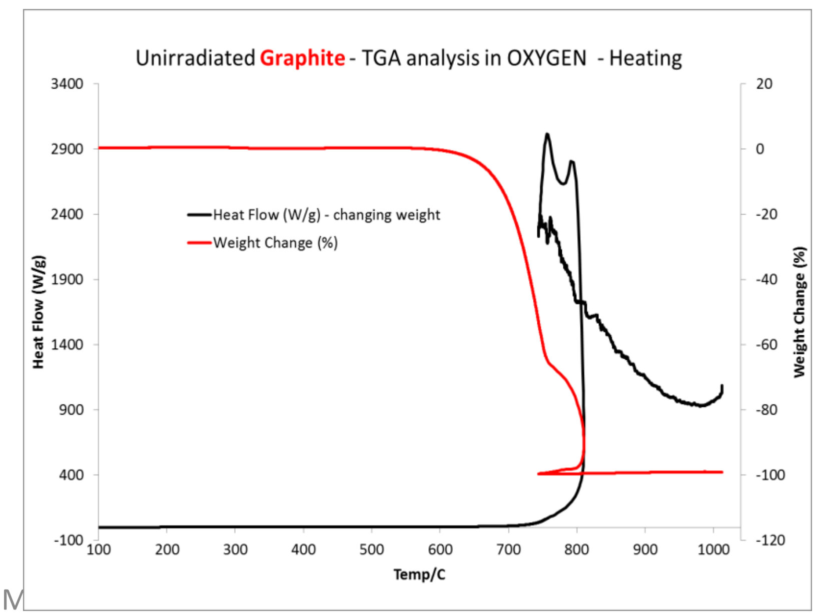




a = RT, b = 400°C, c = 720°C, d = 965°C

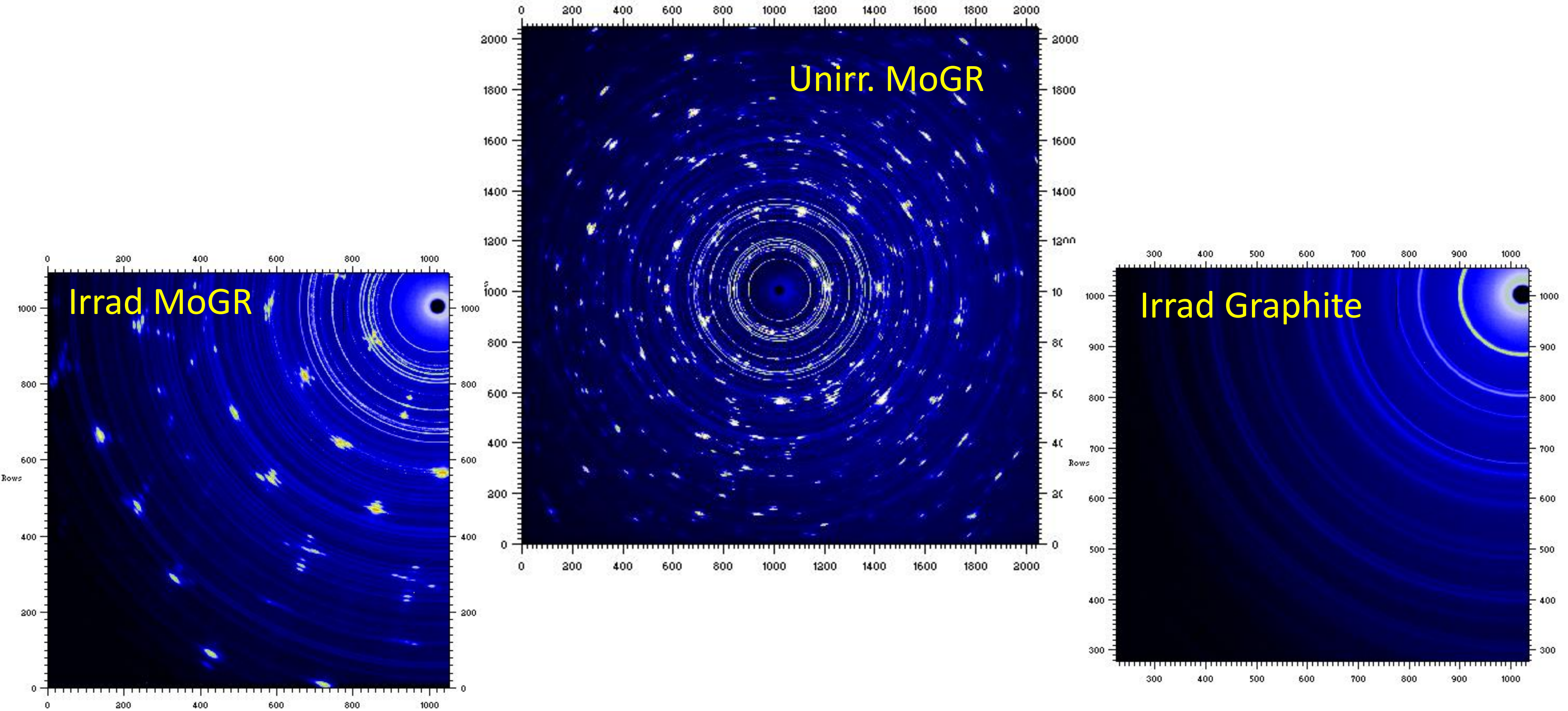

MoGR (unirradiated): Thermal Analysis (TGA) in Oxygen atmosphere
W/g_heating
Weight GAIN %
Heat Flow (W/g)
Weight Change (%)
Temp/C
45
40
35
30
25
20
15
10
5
0
-5
20
0
-20
-40
-60
-80
-100
0
200
400
600
800
1000
1200


MoGR (unirradiated): Thermal Analysis (TGA) in Oxygen atmosphere
W/g_heating
Weight GAIN %
Heat Flow (W/g)
Weight Change (%)
Temp/C
45
40
35
30
25
20
15
10
5
0
-5
20
0
-20
-40
-60
-80
-100
500
550
600
650
700
750
800
850
900


Unirradiated Mo - TGA analysis in Oxygen atmosphere
Heat Flow (W/g)
Weight Change (%)
5
4.5
4
3.5
3
2.5
2
1.5
1
0.5
0
20
0
-20
-40
-60
-80
-100
-120
100
200
300
400
500
600
700
800
900

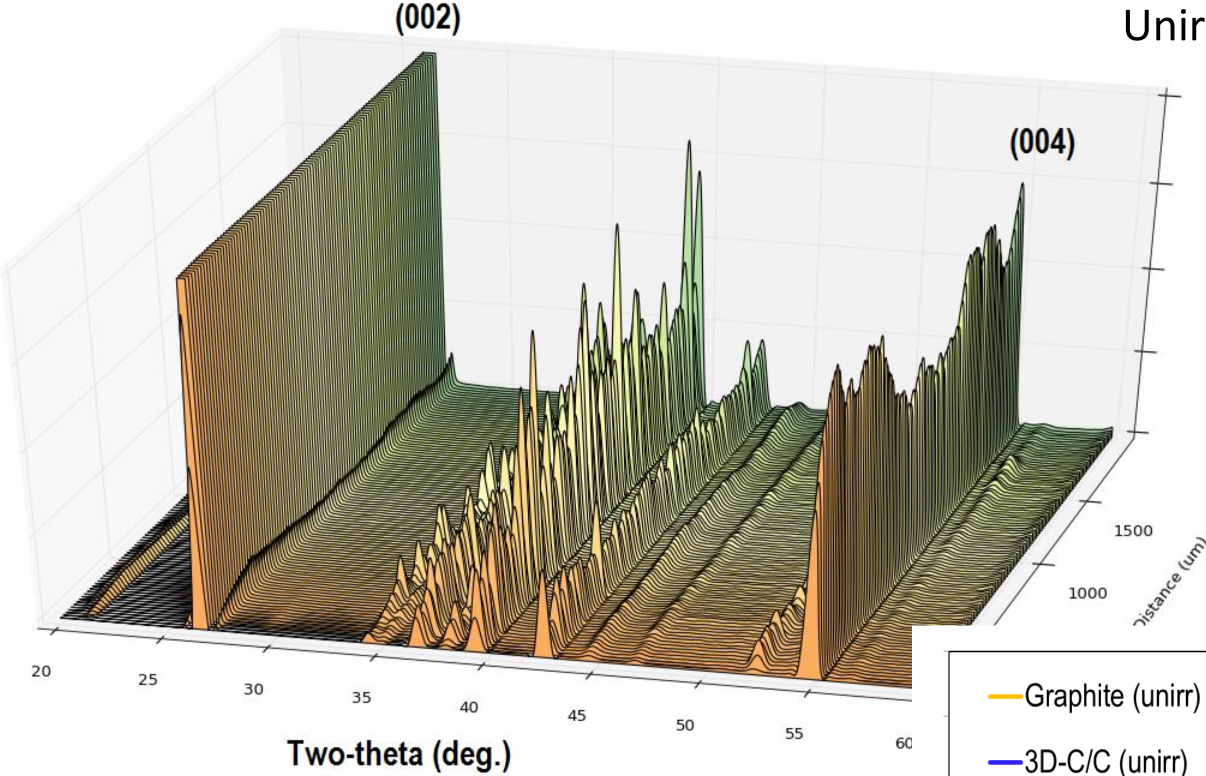

Unirradiated Graphite - TGA analysis in OXYGEN - Heating
Heat Flow (W/g) - changing weight
Weight Change (%)
Heat Flow (W/g)
Weight Change (%)
Temp/C
3400
2900
2400
1900
1400
900
400
-100
20
0
-20
-40
-60
-80
-100
-120
100
200
300
400
500
600
700
800
900
1000

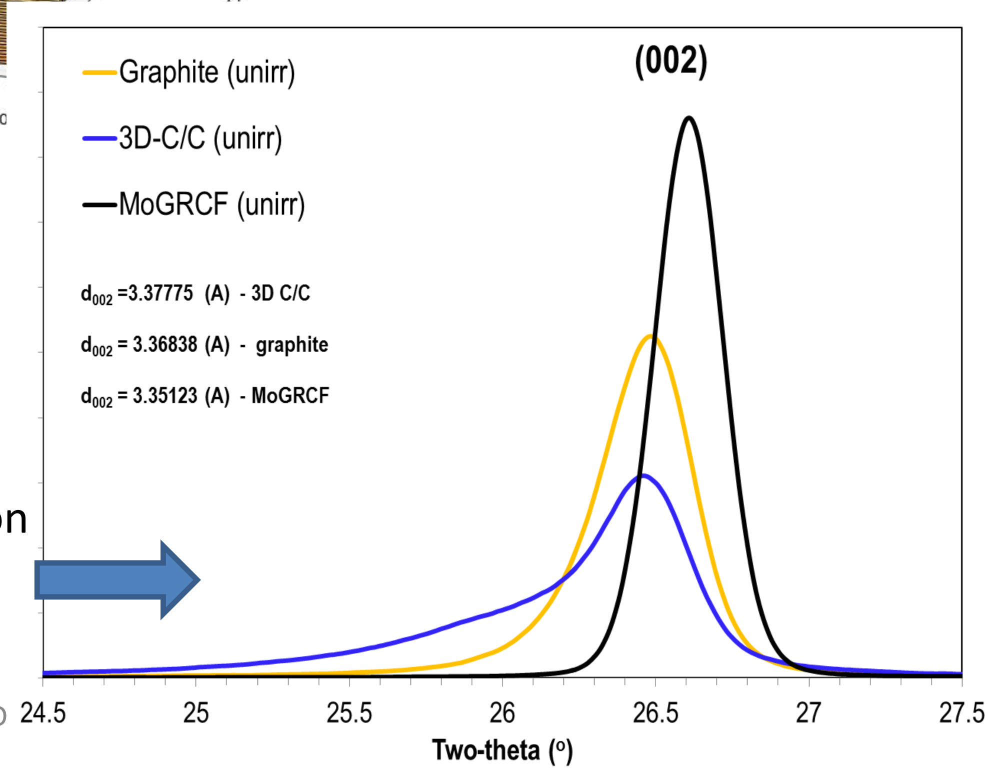
## XRD Study – 70 keV Monochromatic Beam at X17A



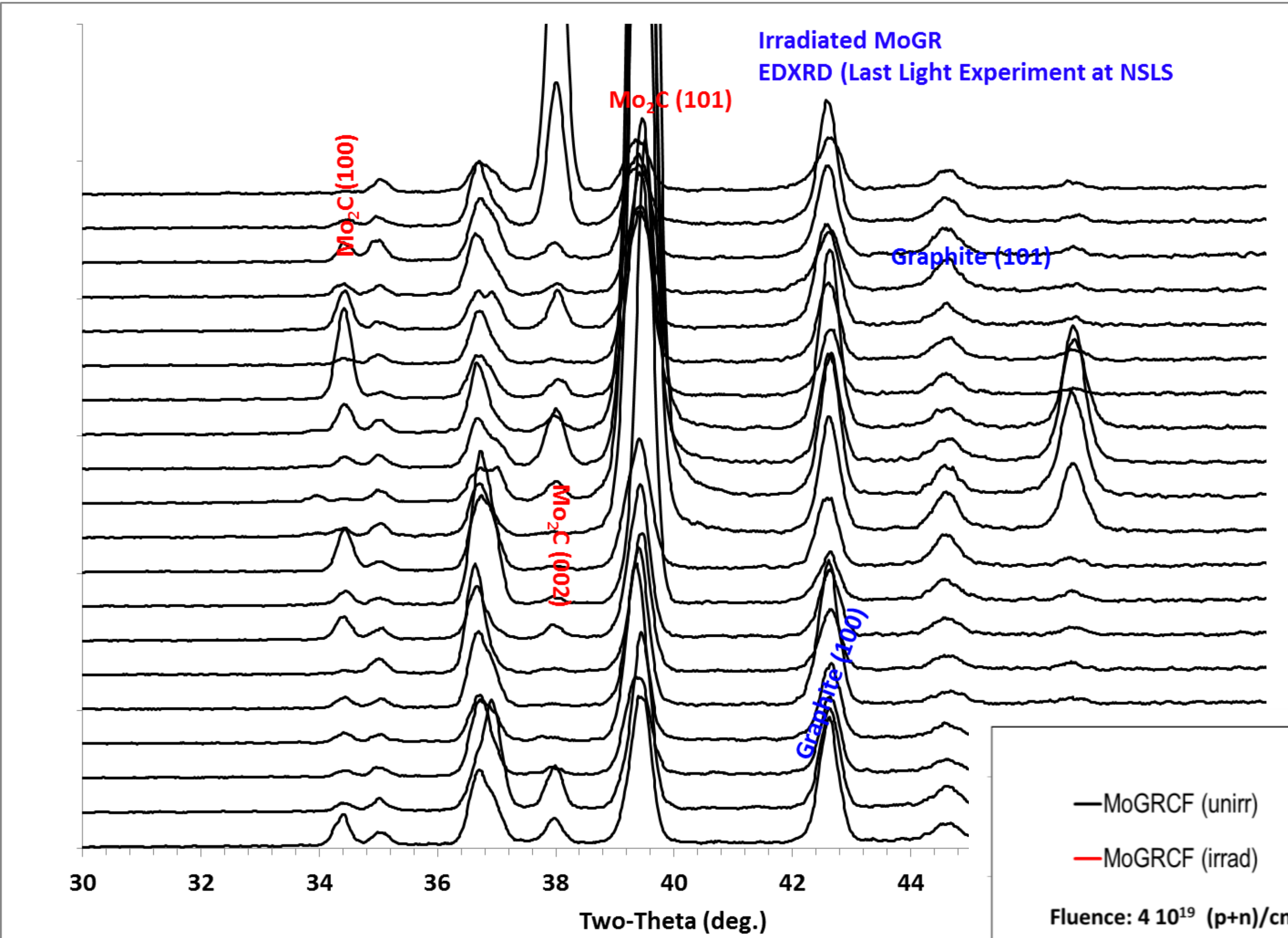

Diffraction pattern of (a) unirradiated MoGRCF (b) irradiated at modest fluence (~$10^{19}$ $cm^{-2}$) and (c) irradiated graphite

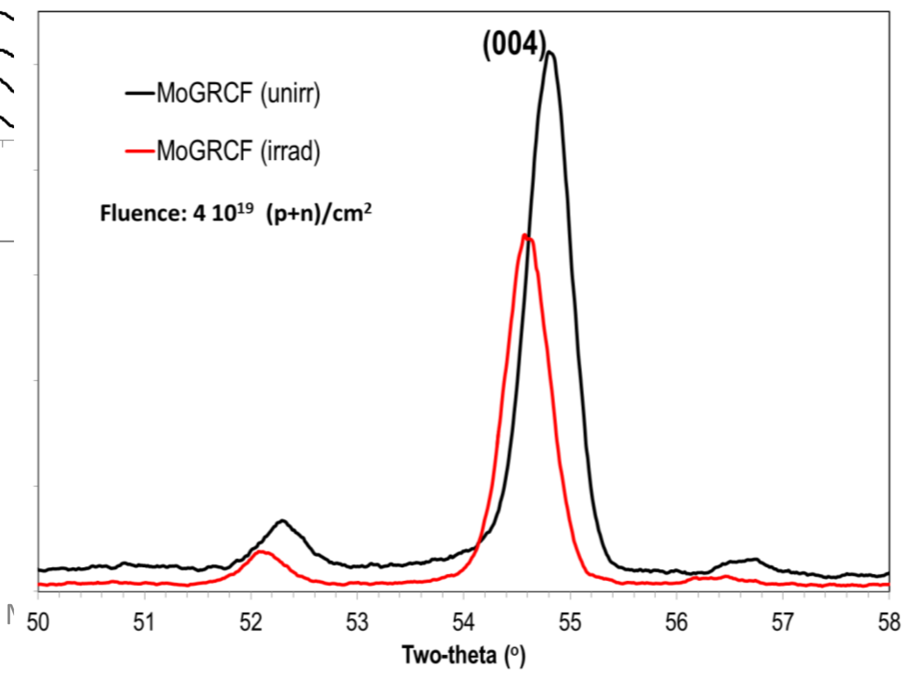
# Unirradiated MoGR 3D Phase Map



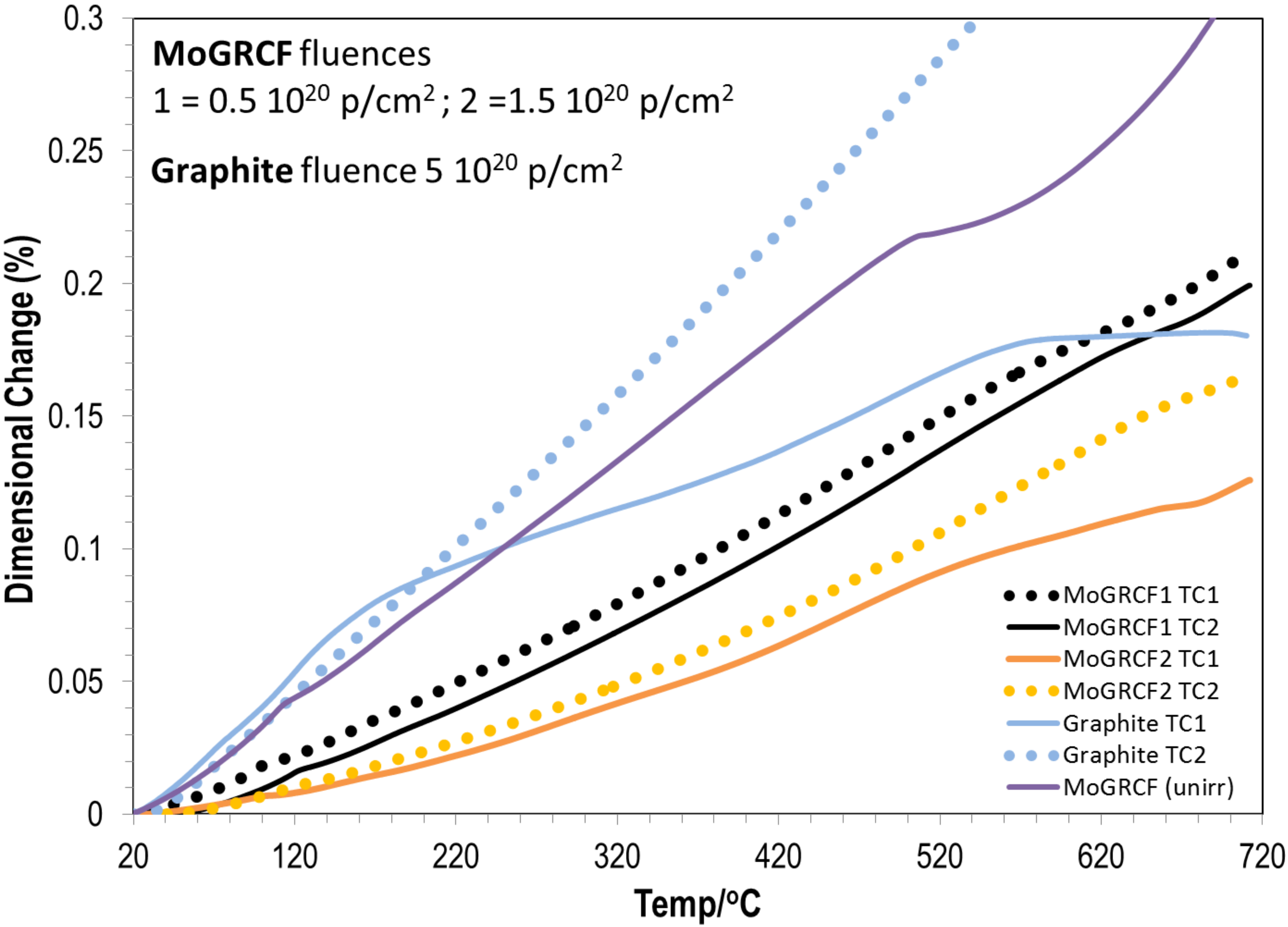




Company claim of achieving good graphitization in MoGR is valid

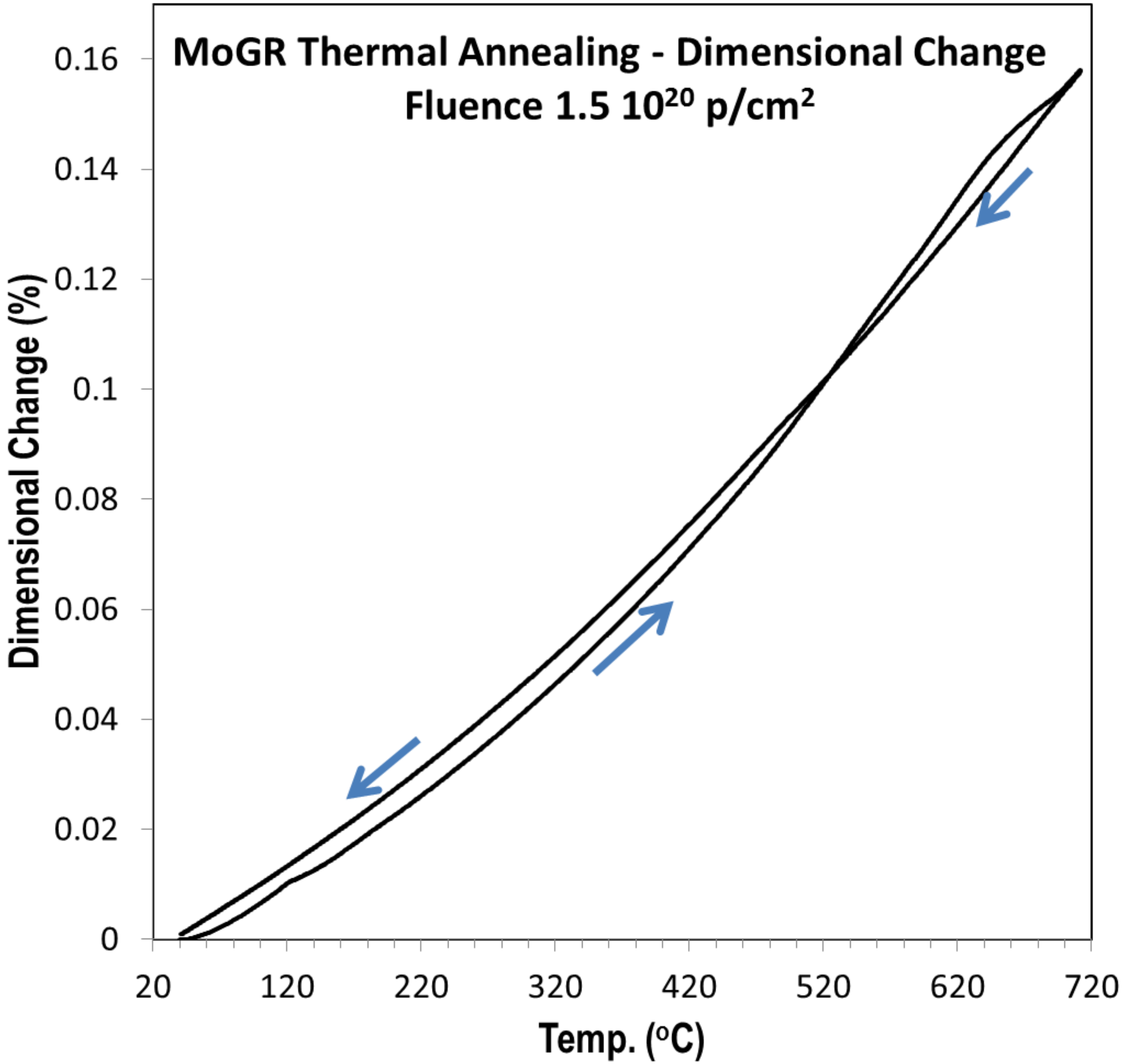


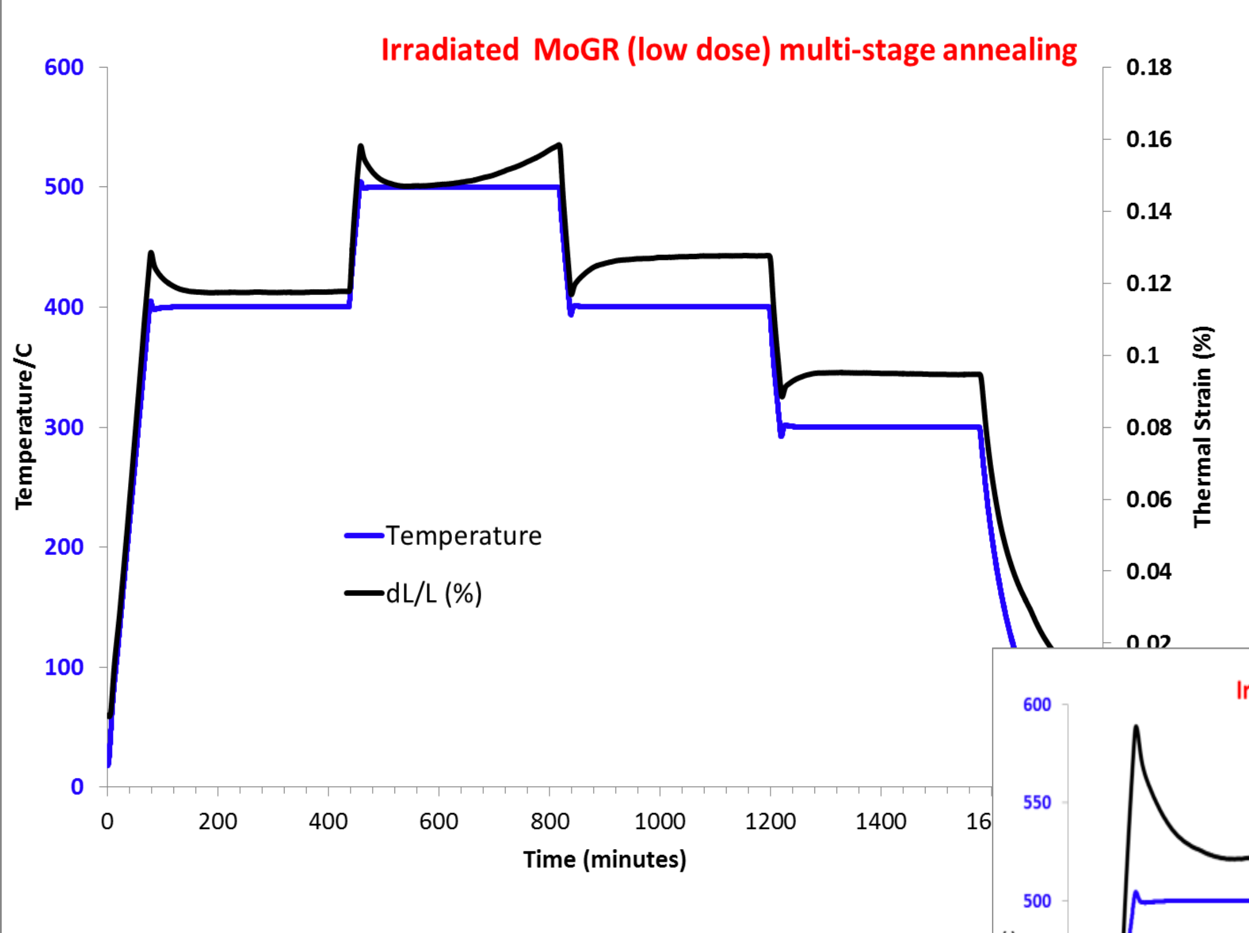




# LAST LIGHT NSLS Experiment
# MoGR received last NSLS photon !!!!

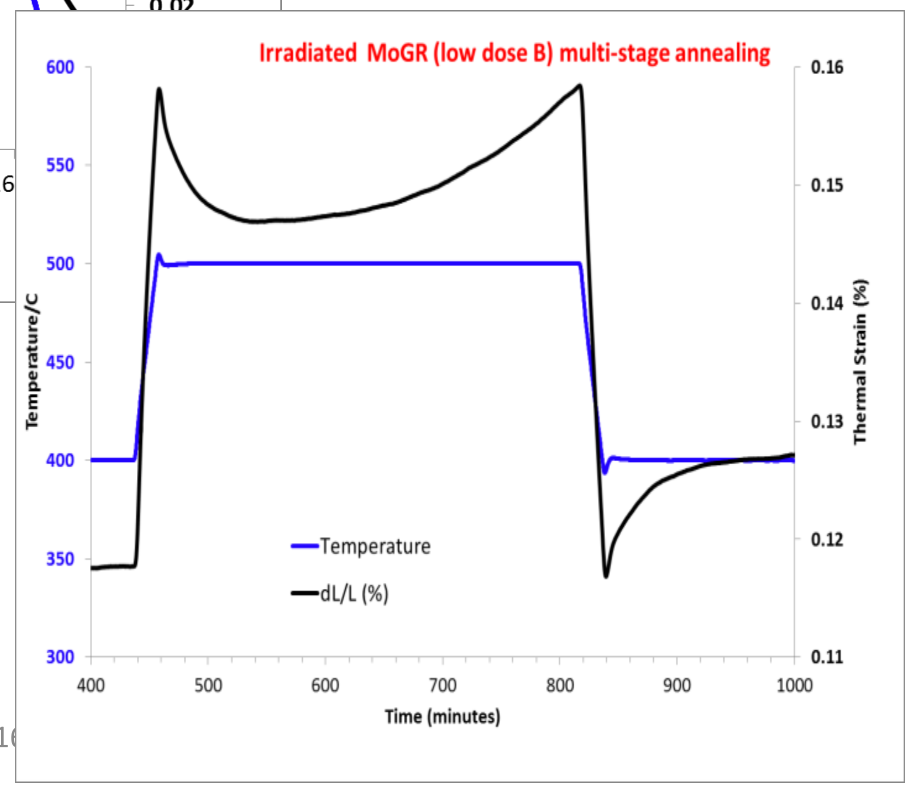


MoGRCF fluences
1 = 0.5 10^20 p/cm^2 ; 2 =1.5 10^20 p/cm^2
Graphite fluence 5 10^20 p/cm^2
Dimensional Change (%)
0.3
0.25
0.2
0.15
0.1
0.05
0
20
120
220
320
420
520
620
720
Temp/ºC
MoGRCF1 TC1
MoGRCF1 TC2
MoGRCF2 TC1
MoGRCF2 TC2
Graphite TC1
Graphite TC2
MoGRCF (unirr)

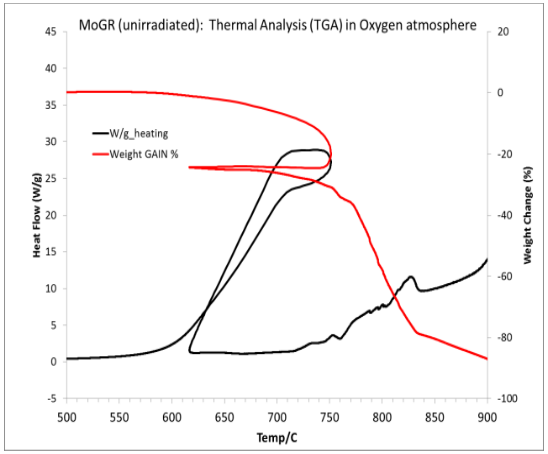

MoGR Thermal Annealing - Dimensional Change
Fluence 1.5 10^20 p/cm^2
Dimensional Change (%)
0.16
0.14
0.12
0.1
0.08
0.06
0.04
0.02
0
20
120
220
320
420
520
620
720
Temp. (°C)

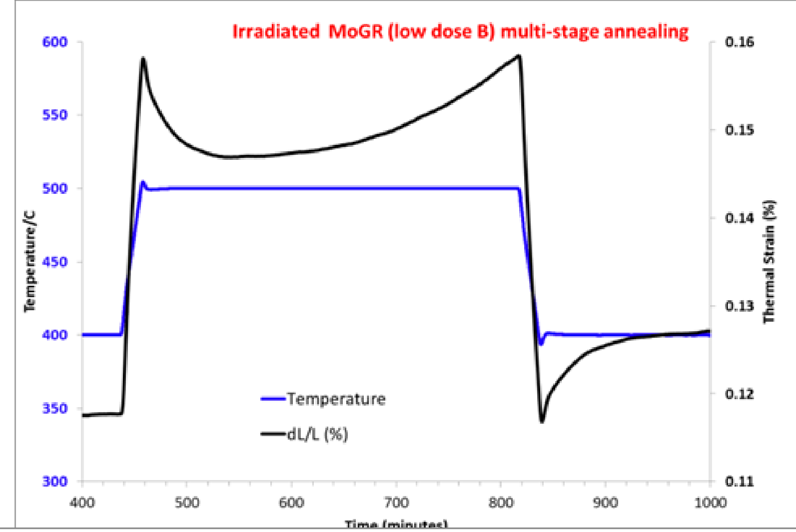


Irradiated MoGR (low dose) multi-stage annealing
Temperature/C
600
500
400
300
200
100
0
Thermal Strain (%)
0.18
0.16
0.14
0.12
0.1
0.08
0.06
0.04
0
200
400
600
800
1000
1200
1400
Time (minutes)
Temperature
dL/L (%)

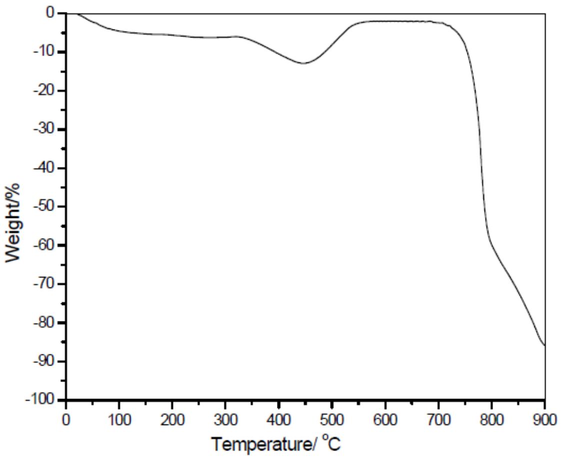



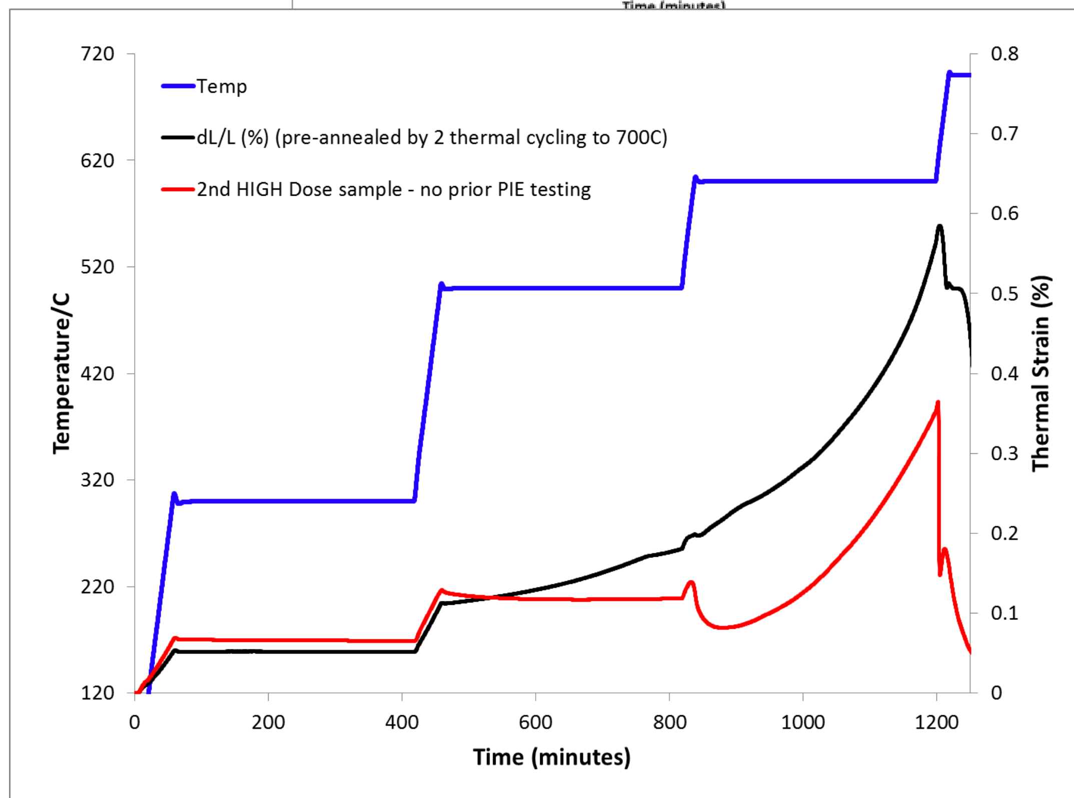
Irradiated MoGR (low dose B) multi-stage annealing
Temperature/C
600
550
500
450
400
350
300
Thermal Strain (%)
0.16
0.15
0.14
0.13
0.12
0.11
400
500
600
700
800
900
1000
Time (minutes)
Temperature
dL/L (%)

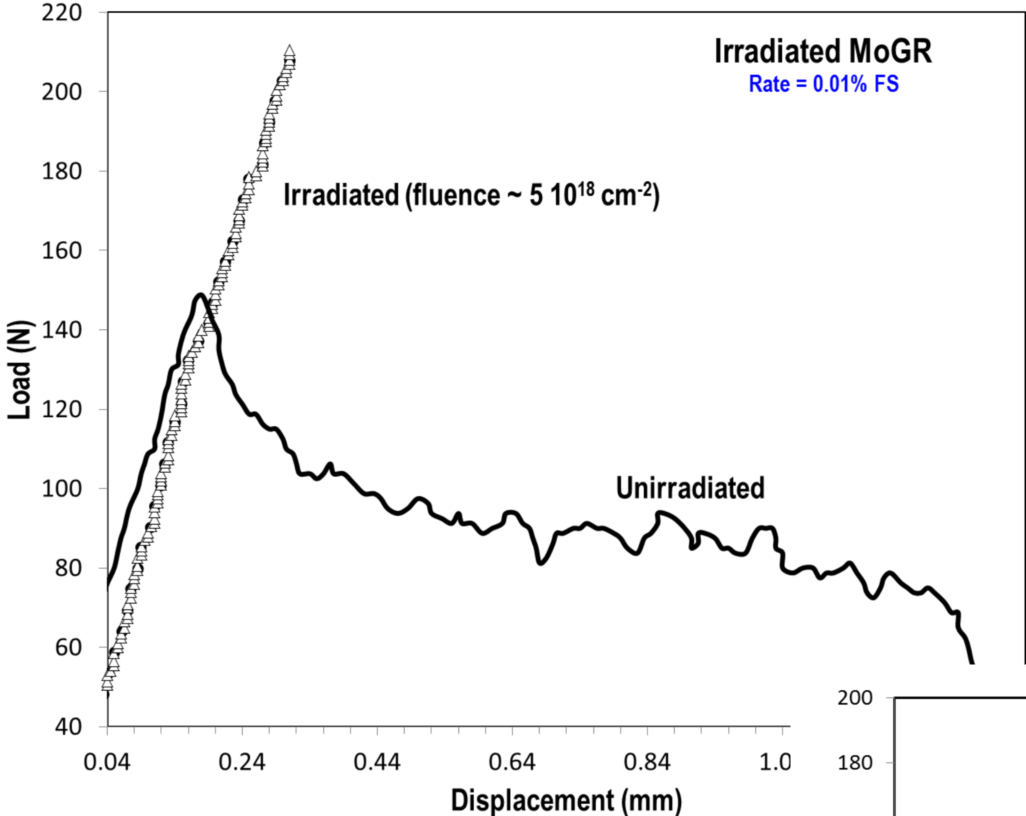


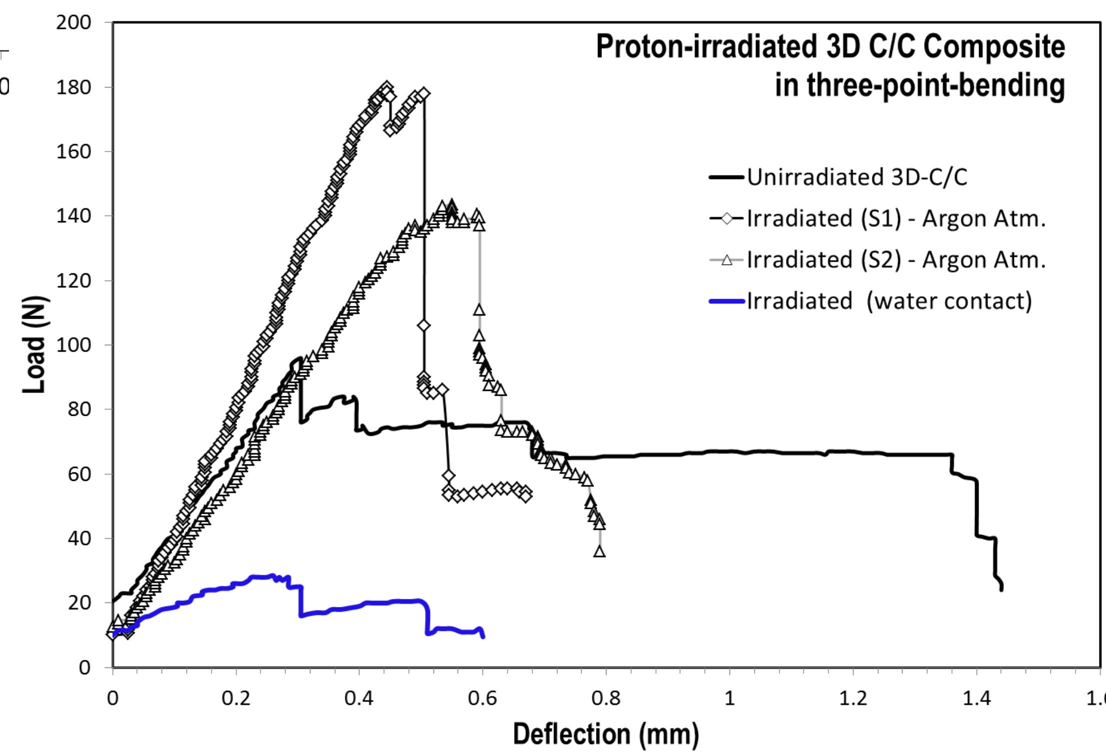

TGA curve of the as-prepared $Mo_2C$ sample in flowing air.



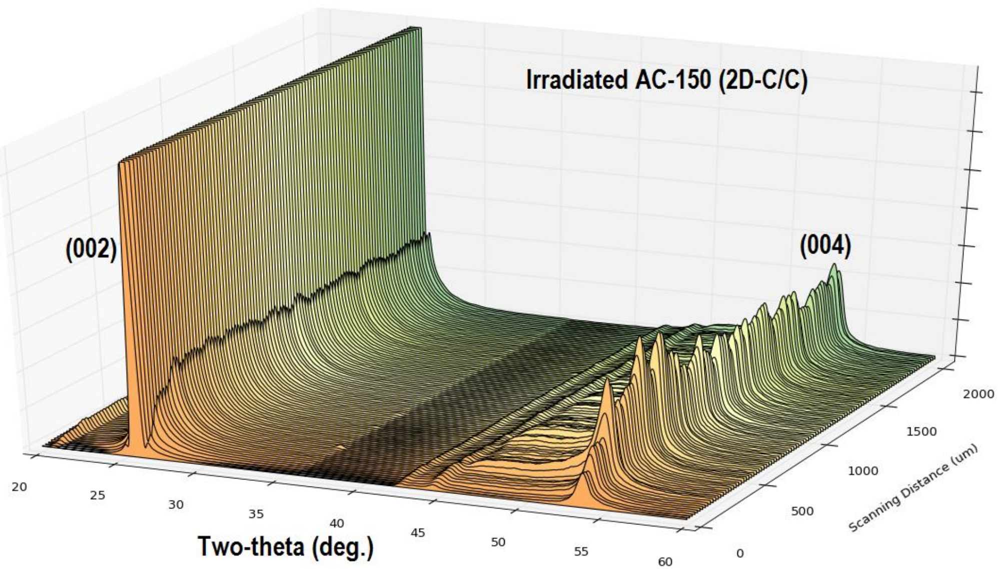


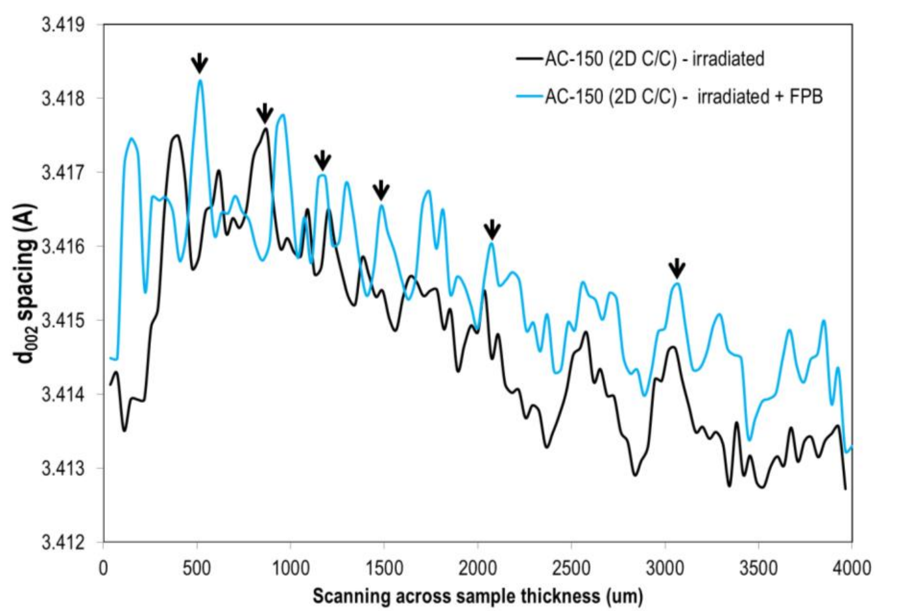
Irradiated MoGR
Rate = 0.01% FS
Irradiated (fluence ~ 5 10^18 cm^-2)
Unirradiated
Load (N)
Displacement (mm)
220
200
180
160
140
120
100
80
60
40
0.04
0.24
0.44
0.64
0.84
1.0

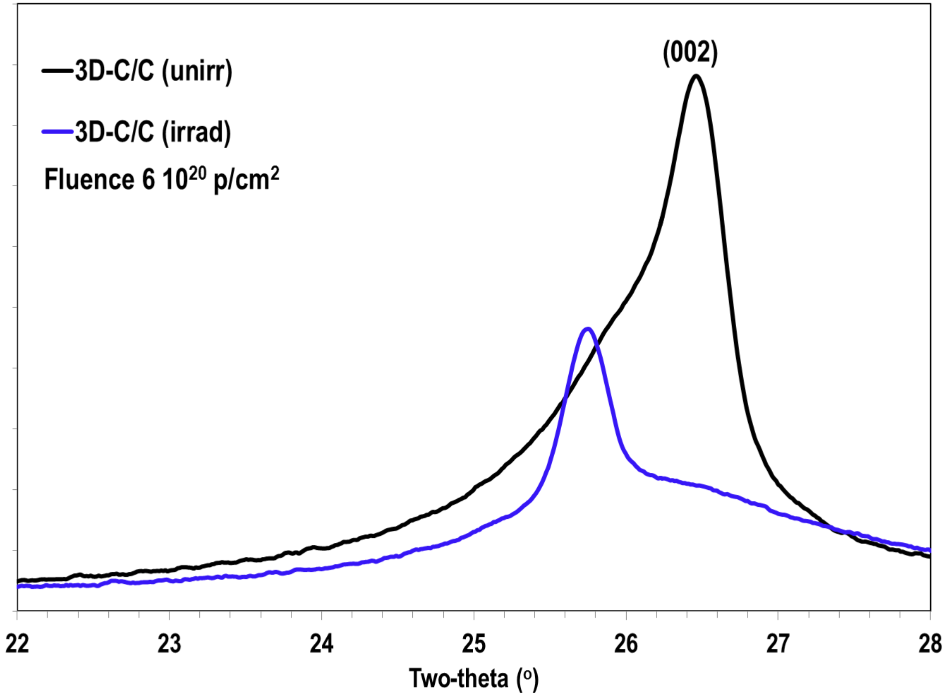
Proton-irradiated 3D C/C Composite
in three-point-bending
Unirradiated 3D-C/C
Irradiated (S1) - Argon Atm.
Irradiated (S2) - Argon Atm.
Irradiated (water contact)
Load (N)
Deflection (mm)
200
180
160
140
120
100
80
60
40
20
0
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6

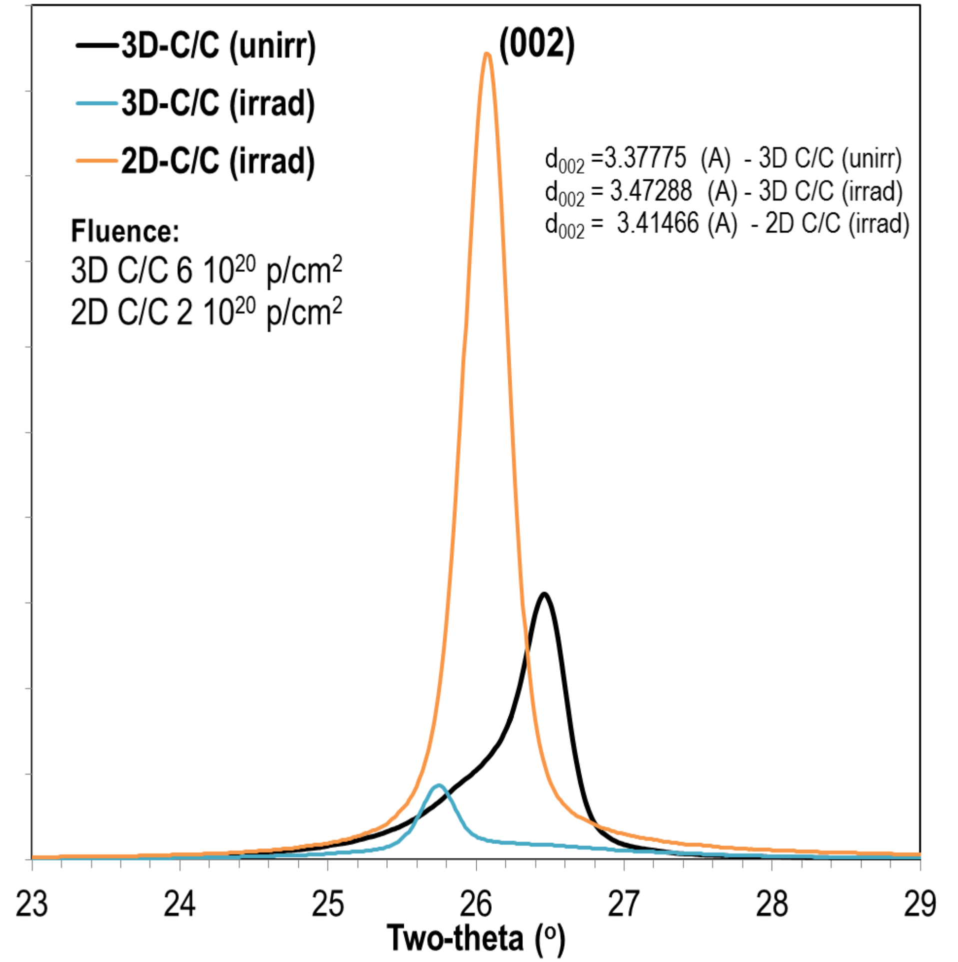
# CFC


Irradiated AC-150 (2D-C/C)
(002)
(004)
20
25
30
35
40
45
50
55
60
Two-theta (deg.)
0
500
1000
1500
2000
Scanning Distance (um)


AC-150 (2D C/C) - irradiated
AC-150 (2D C/C) - irradiated + FPB
3.419
3.418
3.417
3.416
3.415
3.414
3.413
3.412
$d_{002}$ spacing (A)
0
500
1000
1500
2000
2500
3000
3500
4000
Scanning across sample thickness (um)

# CFC


3D-C/C (unirr)
3D-C/C (irrad)
Fluence 6 10^20 p/cm^2
(002)
22
23
24
25
26
27
28
Two-theta (°)


3D-C/C (unirr)
3D-C/C (irrad)
2D-C/C (irrad)
(002)
$d_{002}$ =3.37775 (A) - 3D C/C (unirr)
$d_{002}$ = 3.47288 (A) - 3D C/C (irrad)
$d_{002}$ = 3.41466 (A) - 2D C/C (irrad)
Fluence:
3D C/C 6 10^20 p/cm^2
2D C/C 2 10^20 p/cm^2
23
24
25
26
27
28
29
Two-theta (°)

# SUMMARY

MoGR appears to be an intriguing compound

There appear to be signs of pre-mature degradation from irradiation as compared to its dominant constituent (graphite)

The on-going irradiation tests will reveal (we hope) the FLUENCE threshold at which the material begins to experience structural change.

New experiments will also provide the basis for assessing the effect of the composition of the compound on its stability

**Path Forward:**

- **Successfully complete upcoming crucial 200 MeV irradiation phase**
- **Prepare for the important PIE (following visual assessment) especially XRD at XPD beamline**