Update of post-irradiation examination of collimator materials irradiated at GSI

C. Accettura

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M. Tomut, A. Prosvetov, P. Simon, P. Bolz (GSI)

ColUSM #119 13/09/2019
Outline

• Introduction
• Electrical resistivity measurement
  • DC
  • H011 cavity
• Microscopic analysis
Irradiation conditions

- The irradiation took place between the 27th of March and the 1st of April 2019 in the M3 line of the M-branch
- 113h of beamtime granted
- Ca ions of 4.8 MeV/u to reach higher DPA
- Peak DPA in Mo coating equal to the one in HL-LHC (~1e-3DPA)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Species</td>
<td>Ca48</td>
</tr>
<tr>
<td>Charge state</td>
<td>+10</td>
</tr>
<tr>
<td>Energy</td>
<td>4.8 MeV/u</td>
</tr>
<tr>
<td>Flux</td>
<td>5÷8.5 ion/cm²·s</td>
</tr>
<tr>
<td>Time pulse</td>
<td>1.8÷5.2 ms</td>
</tr>
<tr>
<td>Frequency</td>
<td>5 Hz</td>
</tr>
<tr>
<td>Beam spot</td>
<td>2.5x2.5 cm² or 2.7x2.7 cm²</td>
</tr>
</tbody>
</table>
Irradiation conditions

• Movable sample holder with 4 target stations
• Possible to irradiate 1 single station (4 materials) → reach 4 different fluences → relation between fluence (DPA) and properties degradation
• 2 samples geometries (optimized for PIE)

<table>
<thead>
<tr>
<th>Fluences [ions/cm²]</th>
<th>Peak DPA coating</th>
<th>Peak DPA bulk</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 \cdot 10^{12}$</td>
<td>$\sim 2.8 \cdot 10^{-6}$</td>
<td>$\sim 1.1 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>$1 \cdot 10^{13}$</td>
<td>$\sim 2.8 \cdot 10^{-5}$</td>
<td>$\sim 1.1 \cdot 10^{-3}$</td>
</tr>
<tr>
<td>$7 \cdot 10^{13}$</td>
<td>$\sim 1.9 \cdot 10^{-4}$</td>
<td>$\sim 7.8 \cdot 10^{-3}$</td>
</tr>
<tr>
<td>$4 \cdot 10^{14}$</td>
<td>$\sim 1.1 \cdot 10^{-3}$</td>
<td>$\sim 4.4 \cdot 10^{-2}$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Peak DPA collimator HL-LHC life</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mo coating</td>
</tr>
<tr>
<td>MoGr secondary</td>
</tr>
<tr>
<td>MoGr primary</td>
</tr>
</tbody>
</table>
Holder description-materials

- MoGr Nanoker (Nb8304Ng) → production grade
- Graphite SGL R4550
- Nb8304Ng+ Mo coating DTI
- R4550+ Mo coating DTI

- CFC Tatsuno FS140
- MoGr Brevetti (Mg6541Fc) → with fibers, tested in HRTM36
- Mg6541Fc+ Mo coating CERN
- Nb8304Ng+ Cu coating DTI

• All the samples irradiated at 4 different fluences:

<table>
<thead>
<tr>
<th>1e12</th>
<th>1e13</th>
<th>7e13</th>
<th>4e14</th>
</tr>
</thead>
</table>
Visual inspection

No visual signs of mechanical damage both in coating and bulk
Activation after the test

- Just after the irradiation ~200µSv/h at ~20cm
- Just after the irradiation (inside the box) ~31µSv/h
- After 1 day (inside the box) ~8µSv/h

End of irradiation

~ 2 months

Shipping at CERN

- Sample shipped at CERN on the 11\textsuperscript{th} of June (dose rate<0.1µSv/h)
- Measurements ongoing in the mechanical lab (376-R-003) classified as a controlled area since the 25\textsuperscript{th} of June
Outline

• Introduction

• Electrical resistivity measurement
  • DC
  • H011 cavity

• Microscopic analysis
Electrical resistivity measurements

- Four probes method set-up
- Parallel resistance model (2 or 3-layers)
- Minimized sample thickness to see the contribution of the irradiated layer
- Radiation penetration depth from FLUKA simulation

\[
R_{\text{irradiated}} = ?
\]

\[
R_{\text{bulk}} = \rho_{\text{pristine}} \frac{L}{d_1 \cdot t_{\text{bulk}}}
\]

\[
R_{\text{coating}} = ?
\]

\[
R_{\text{bulk}} = \frac{R_{\text{bulk, pristine}} \cdot R_{\text{bulk, irradiated}}}{R_{\text{bulk, pristine}} + R_{\text{bulk, irradiated}}}
\]

Four probe methods measurements @ Impedance meeting:
https://indico.cern.ch/event/816840/
Electrical resistivity measurements-bulks

- All the materials increase the resistivity with fluences
- Graphite starts from a much more disordered state → reach saturation before MoGr
- Electrical conductivity in MoGr systematically higher than Gr in spite of stronger DPA effect
- **Threshold effect**: for a DPA 10 times higher then in secondary collimators the MoGr will have a conductivity ~0.6-0.7 MS/m
- MoGr with fibers more radiation resistant
- High scattering in CFC values → Estimated value at 4e14 with min R|| not with the average:

\[
R_{\text{irr}} = \frac{R_{\text{pristine}} \cdot R_||}{R_{\text{pristine}} - R_||} < 0
\]
Electrical resistivity measurements-coatings

- No clear trend with fluences
- Final values for Mo ~6-13 MS/m → factor 2-3 worst then initial values

Possible explanation:
- Not possible to see the trend because of the high uncertainty related to multi-layer model
- Annealing induced by temperature at higher fluences
- 2 layer model not accurate (conductivity of the irradiated bulk different with respect to the case of uncoated → different DPA distribution → not sensitive to the irradiated layer conductivity
Impedance measurement with H011 cavity

- Uncoated samples
  - No relevant changes in the measured Q factor
  - Coherent with DC: average DPA in the layer investigated with the cavity (skin depth ~4µm) << then the average DPA in the whole irradiated layer

<table>
<thead>
<tr>
<th>Fluence [ions/cm²]</th>
<th>Average DPA over 50µm (DC)</th>
<th>Average DPA over 10µm (H011)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1e12</td>
<td>5.5E-05</td>
<td>5.5E-07</td>
</tr>
<tr>
<td>1e13</td>
<td>5.5E-04</td>
<td>5.5E-06</td>
</tr>
<tr>
<td>7e13</td>
<td>3.8E-03</td>
<td>3.8E-05</td>
</tr>
<tr>
<td>4e14</td>
<td>2.2E-02</td>
<td>2.2E-04</td>
</tr>
</tbody>
</table>

→ No relevant Δσ for DPA<2e-4 : coherent with DC!
Impedance measurement with H011 cavity

• Coated samples
  • Sensitivity of the cavity too low to detect small $\Delta \sigma$ (within the error bar of the measurements) in the region of interest (flat curve)
  • Possible improvement with the smaller cavity

$Q$ factor variation in function of material resistivity (coated and uncoated sample)

Thanks to Nicolò and Adnan!
Outline

• Introduction
• Electrical resistivity measurement
  • DC
  • H011 cavity
• Microscopic analysis
Raman Spectroscopy of MoGr

- Penetration of the red laser ($\lambda \sim 632.8\text{nm}$) in graphite $\sim 100\text{ nm} \rightarrow$ investigation of the irradiated region only
- Mapping over 20 points to increase the statistic

**Table:**

<table>
<thead>
<tr>
<th>Fluence [ions/cm²]</th>
<th>Grain size before [nm]</th>
<th>Grain size after [nm]</th>
<th>Normalized grain reduction</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00E+12</td>
<td>146</td>
<td>180</td>
<td>-26%</td>
</tr>
<tr>
<td>1.00E+13</td>
<td>140</td>
<td>150</td>
<td>-7.14%</td>
</tr>
<tr>
<td>7.00E+13</td>
<td>410</td>
<td>150</td>
<td>63.41%</td>
</tr>
<tr>
<td>4.00E+14</td>
<td>350</td>
<td>72</td>
<td>79.43%</td>
</tr>
</tbody>
</table>

$La[\text{nm}] = (2.4\times 10^{-10})\lambda^4 \left(\frac{I_G}{I_D}\right)$

- Important change of grain at 7e13 (coherent with electrical resistivity)
- Results to be cross-checked with the same device used before irradiation (different laser wavelength)

**Equation:**

$La[\text{nm}] = (2.4\times 10^{-10})\lambda^4 \left(\frac{I_G}{I_D}\right)$
Raman Spectroscopy of Graphite

- Disorder increase with the fluences
- Grain size change less that MoGr, but final grain smaller (same as for electrical conductivity)

<table>
<thead>
<tr>
<th>Fluence [ions/cm²]</th>
<th>Grain size before [nm]</th>
<th>Grain size after [nm]</th>
<th>Normalized grain reduction</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00E+12</td>
<td>47</td>
<td>44</td>
<td>5.7%</td>
</tr>
<tr>
<td>1.00E+13</td>
<td>49</td>
<td>43</td>
<td>11.62%</td>
</tr>
<tr>
<td>7.00E+13</td>
<td>57</td>
<td>43</td>
<td>24.13%</td>
</tr>
<tr>
<td>4.00E+14</td>
<td>60</td>
<td>28</td>
<td>52.25%</td>
</tr>
</tbody>
</table>

Measurements performed in GSI
SEM Mo on MoGr

Mo on MoGr irradiated at 4e14 ions/cm² → Max DPA in the coating corresponding to the peak DPA in the coating at the end of the HL-LHC lifetime

Many thanks to A. Peres, A. Baris (EN/MME)
SEM Mo on MoGr

Mo on MoGr irradiated at 4e14 ions/cm² → Max DPA in the coating corresponding to the peak DPA in the coating at the end of the HL-LHC lifetime

Before irradiation

After irradiation

- We are comparing different samples but same bulk, surface preparation and coating process
- Irradiated sample present more discontinuous microstructure: possible indication of grain size reduction → to be checked with XRD
- Discontinuities could explain the reduction in electrical conductivity
FIB Mo on MoGr
Mo on MoGr irradiated at $4 \times 10^{14}$ ions/cm$^2$ → Max DPA in the coating corresponding to the peak DPA in the coating at the end of the HL-LHC lifetime

- The coating-bulk interface is continuous, no evidence of coating detachment
- No crack in the coating
Mo on Gr irradiated at 4e14 ions/cm² \( \rightarrow \) Max DPA in the coating corresponding to the peak DPA in the coating at the end of the HL-LHC lifetime
FIB Mo on Gr
Mo on Gr irradiated at 4e14 ions/cm² → Max DPA in the coating corresponding to the peak DPA in the coating at the end of the HL-LHC lifetime

- The coating-bulk interface is continuous, no evidence of coating detachment
- No crack in the coating
- Mo diffusion in the bulk thought the porosities (probably before irradiation)
Thermal diffusivity

- 2-layers model + reverse engineering
- Short and long measurement to study heating up and cooling down (along plane and through plane conductivity studies)

Very thin sample \(\rightarrow\) lateral conduction
Thermal diffusivity

- 2-layers model + reverse engineering
- Short and long measurement to study heating up and cooling down (along plane and through plane conductivity studies)

- The through-plane conductivity seems to be higher after irradiation (faster) → analysis ongoing
Conclusions

- 80 samples of different materials/coatings and size have been irradiated with Ca ion
- No visible sign of radiation-induced mechanical damage of coatings/bulks
- Electrical conductivity degradation in the bulk in line with expectations (higher for more order materials)
- Threshold effect at 7e17ions/cm² highlighted by electrical conductivity and Raman measurements
- Coating degradation seems to be independent on the fluences (very low threshold): factor of 2 with respect to the pristine value for both Mo and Cu
- SEM and FIB investigation revealed a possible change in the coating microstructure, but the adherence seems good also at the maximum fluences (corresponding to DPA expected in HL-LHC)
- XRD measurement completed → data to be analyzed to understand microstructural changes
Thank you
General information on irradiation-design
## Ion vs proton irradiation

<table>
<thead>
<tr>
<th></th>
<th>High energy proton</th>
<th>Light ion (Ca/C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Penetration</td>
<td>Deep</td>
<td>Superficial</td>
</tr>
<tr>
<td>Gas production</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Activation</td>
<td>High</td>
<td>Zero-low</td>
</tr>
<tr>
<td>DPA rate</td>
<td>Medium</td>
<td>High</td>
</tr>
</tbody>
</table>

- **Penetration**: Deep for proton, superficial for light ion (Ca/C).
- **Gas production**: Yes for proton, no for light ion (Ca/C).
- **Activation**: High for proton, zero-low for light ion (Ca/C).
- **DPA rate**: Medium for proton, high for light ion (Ca/C).

### Notes
- Need of multi-layer model, less accurate.
- How relevant is for collimator? Simulation ongoing.
- Cheap measurement.
- High DPA reached in tens of hours → possible increase of damage.

For a coating with similar structure to HIPIMPS, no blistering until $3 \times 10^{18}$ ions/cm$^2$.

Simulation of gas production

• The ion irradiation is characterized by
  • High dose rate → conservative with respect p+
  • No gas production → important to investigate!
• Simulate gas production with H/He implantation
  • Where? → MIAMI (University of Huddersfield): 10keV, ~100nm
    • Possible to regulate the beam to have certain appm/DPA
    • Online TEM monitoring
    • Thermophysical characterization would not be possible in such a small layer, but a threshold for blistering can be established
• How much?
  • FLUKA simulation (appm/DPA production)
  • Measurement of diffusion coefficient with TDS
• With the ion irradiated samples (at different DPA) we could investigate the influence of the DPA on the gas effects
Flux: $5 \times 10^9$ ions/cm$^2$/s (Ca-48, 4.8 MeV/u)

- MoGr Nb8304Ng (2.40 g/cm$^3$)
- MoGr MG6541Fc (2.48 g/cm$^3$)
- CIC FS140 (1.75 g/cm$^3$)
- Graphite R4550 (1.83 g/cm$^3$)
- Graphite R4550 + 0.5µm Ti + 3µm Cu
- Graphite R4550 + 0.5µm Ti + 3µm Cu (DPA)

DPA per hour of irradiation

Depth (µm)
Temperature profile

- 2D geometry
- 1 imp/s

\[W_{out} = 2.16 \, W, \, W_{in} = 3 \, W\]
\[\rightarrow \text{Close to equilibrium}\]

! Repeat the analysis for electrical conductivity sample
! Repeat with right power/imp and frequency
Holder description-geometry

- Samples: 20x5x0.15mm
- Geometry chosen for electrical and thermal conductivity measurements (2-3 layers)
- All the sample have been characterized before to know the reference value
Holder description-geometry

- Disk: 10x1mm
- Geometry chosen for microscopy (FIB, XRD, Raman), indentation and adhesion

100um flexible graphite in the groove and below the mask
Holder description-matals

• Hybrid holder (beamtime left) to investigate intermediate fluences

<table>
<thead>
<tr>
<th>2e13</th>
<th>1.4e14</th>
<th>2e13</th>
<th>1.4e14</th>
</tr>
</thead>
</table>

- MoGr Nanoker (Nb8304Ng)\rightarrow production grade
- Graphite SGL R4550
- Nb8304Ng+ Mo coating DTI
- R4550+ Mo coating CERN (no more spares of DTI coating)
Pristine sample characterization
Thermal diffusivity

- Simulation indicates that the influence of the damaged layer should be seen with thin sample

- LFA of thin sample required reverse engineering to find the values of conductivity
Indentation–Bulk

- Each sample indented in 25 points with penetration-controlled (nano) and load controlled (micro)
- Microindentation allows a reduction of the standard deviation → useful to observe radiation-induced hardening and increase of the elastic modulus
- Differences with respect other method (e.g. IET) related to factors such as anisotropy and non-linearity.

<table>
<thead>
<tr>
<th>Material</th>
<th>Average E [GPa]</th>
<th>Standard deviation [GPa]</th>
<th>IET [GPa]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Graphite R4550</td>
<td>6.9</td>
<td>1.7</td>
<td>11</td>
</tr>
<tr>
<td>MG6541Fc</td>
<td>12</td>
<td>1.9</td>
<td>5</td>
</tr>
<tr>
<td>Nb8304Ng</td>
<td>8.1</td>
<td>1.6</td>
<td>4</td>
</tr>
<tr>
<td>CFC FS 140</td>
<td>4.9</td>
<td>3.3</td>
<td>3</td>
</tr>
</tbody>
</table>
Coating adhesion-Scratch test

- An increasing load is applied by the nanoindenter, possible coating failure can be detected with:
  - Optical microscopy images → difficult for material roughness and coating/bulk colours
  - Abrupt variation in the load curve → irregularities due to roughness already present
  - Acoustic emission → not available in the used set-up
PIE-additional information
Electrical resistivity measurements

- Bulk resistivity changes in function of fluences (DPA)
- Graphite starts from a much more disordered state → reach saturation before MoGr
- Electrical conductivity in MoGr systematically higher than Gr in spite of stronger DPA effect
- Values coherent with previous test

For a DPA 10 times higher than in secondary collimators the MoGr will have a conductivity ~0.6-0.7 MS/m
- Important to check the area affected by 0.3 DPA in TCP

Irradiation test at GSI 2015-2016
Electrical resistivity measurements-coatings

- Coating degrades at the lowest fluences and then stays constant
- Final values ~6-13 MS/m $\rightarrow$ factor 2-3 worst then initial values
- Irradiated bulk value assumed equal to the one found for uncoated IRRADIATED sample (see slide before): same average DPA but different distribution $\rightarrow$ possible improvement if we put multi-layer model with different layers at different conductivity

$$ R_{\text{coating}} = ? $$

$$ R_{\text{bulk}} = \frac{R_{\text{bulk pristine}} \cdot R_{\text{bulk irradiated}}}{R_{\text{bulk pristine}} + R_{\text{bulk irradiated}}} $$

- Additional analysis ongoing
- Important to study grain size to understand if the trend is reasonable (XRD)
- Potential annealing at high fluence (T~100-150°C, to be checked)
- Annealing cycle at the bake-out temperature planned to check possible improvement
<table>
<thead>
<tr>
<th>Holder</th>
<th>Fluence</th>
<th>DPA peak</th>
<th>Material</th>
<th>$\sigma_{\text{layer irradiated}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.00E+12</td>
<td>1.11E-04</td>
<td>graphite</td>
<td>0.053</td>
</tr>
<tr>
<td>1</td>
<td>1.00E+13</td>
<td>1.11E-03</td>
<td></td>
<td>0.047</td>
</tr>
<tr>
<td>5</td>
<td>2.00E+13</td>
<td>2.22E-03</td>
<td></td>
<td>0.054</td>
</tr>
<tr>
<td>1</td>
<td>7.00E+13</td>
<td>7.77E-03</td>
<td></td>
<td>0.031</td>
</tr>
<tr>
<td>5</td>
<td>1.40E+14</td>
<td>1.55E-02</td>
<td></td>
<td>0.026</td>
</tr>
<tr>
<td>1</td>
<td>4.00E+14</td>
<td>4.44E-02</td>
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<td>0.025</td>
</tr>
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<td>1</td>
<td>1.00E+12</td>
<td>1.11E-04</td>
<td>MoGr</td>
<td>0.575</td>
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<td>1.11E-03</td>
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<td>1</td>
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<td>0.185</td>
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<td>1.55E-02</td>
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<td>0.047</td>
</tr>
<tr>
<td>1</td>
<td>4.00E+14</td>
<td>4.44E-02</td>
<td></td>
<td>0.033</td>
</tr>
</tbody>
</table>
Thermal diffusivity

- 6 out of 40 samples measured (analysis ongoing)
- 2-layers model + reverse engineering
- Short and long measurement to study heating up and cooling down (along plane and through plane conductivity studies)

Very thin sample → lateral conduction