Characterization of radiation damage in graphite induced by GeV heavy ion irradiation

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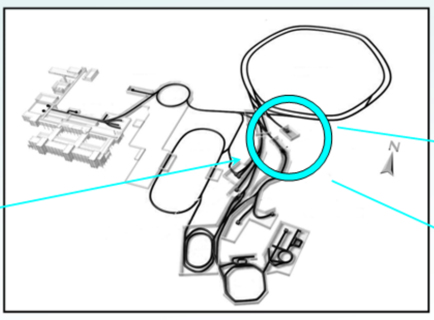
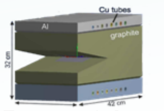
Motivation:

Radiation damage in graphite is mostly investigated for neutrons and little is known about damage induced by swift heavy ions. For the planned Facility for Antiproton and Ion Research (FAIR) - at GSI, new materials for accelerator components exposed to high-intensity primary beams are needed. Graphite and carbon/carbon composites are used due the good thermo-mechanical properties and the low Z minimizing energy deposition.

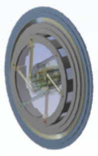
FAIR needs:

- Materials for extreme conditions resistant to:
  - radiation
  - high temperature
  - thermal shock
  - thermal stress
- Radiation damage plays a mayor factor in the lifetime reduction for components like production targets and beam catchers



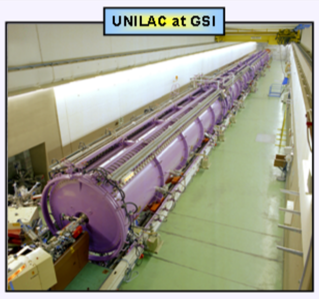


Beam catcher

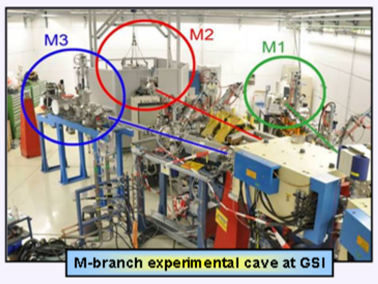


Production Targets

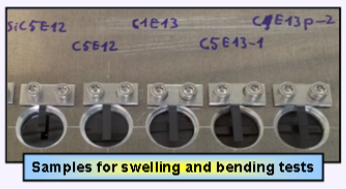
Irradiation condition



UNILAC at GSI



M-branch experimental cave at GSI



Samples for swelling and bending tests

All samples have been irradiated at the M-branch, Material Research beam line at GSI, using GeV energies.

The UNILAC at GSI is able to accelerate a wide range of particles (from Protons up to U) to energies ranging between 3,6 MeV / u and 11,4 MeV / u.

Iradiation parameters

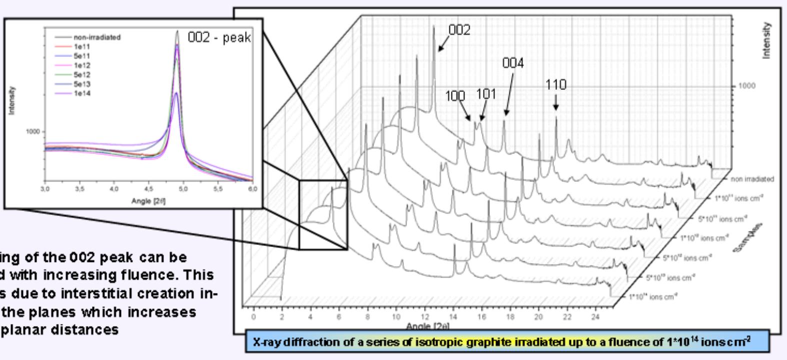
Gold 79 Au 196.97

Energies: from 3,6 to 11,4 MeV/u
Fluxes: 1×10^9 ions cm^{-2} s^{-1} to 1×10^{10} ions cm^{-2} s^{-1}
Fluences: up to 1×10^{14} ions cm^{-2}
Charge state: + 25

Structural analysis

X-Ray diffraction

Structural changes in isotropic fine-grained graphite were analyzed with X-ray diffraction



X-ray diffraction of a series of isotropic graphite irradiated up to a fluence of $1*10^{14}$ ions cm^2

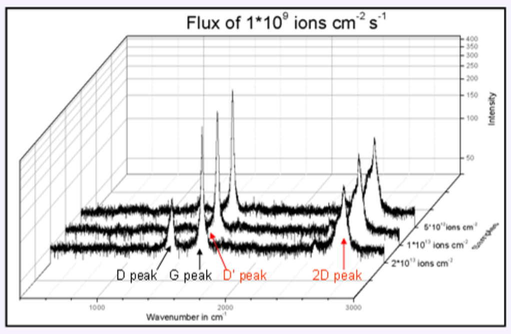
Broadening of the 002 peak can be observed with increasing fluence. This change is due to interstitial creation in-between the planes which increases the inter planar distances

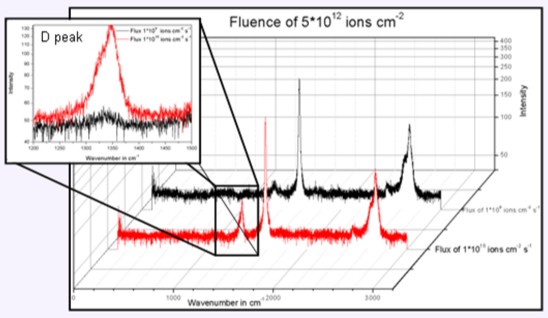
Investigation of irradiated isotropic graphite at the DESY PETRA3 beam line, using a synchrotron X-ray source, show the influence of radiation damage on the structure.

A general broadening of all the peaks can be observed, which is related to the change of the inter lattice distances in the corresponding planes.

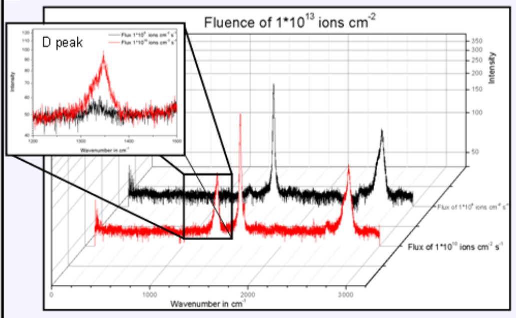
Raman spectroscopy

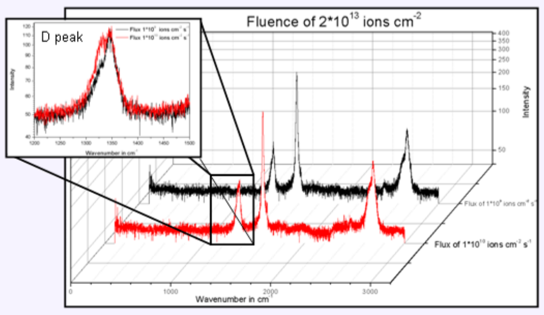
Structural changes in monocrystalline HOPG were analyzed with the Raman spectrometer





Raman spectroscopy analysis of irradiated HOP G shows the increase of the D peak in graphite (associated to structural disorder and defects in the lattice) with increasing fluence. For Fluxes of 1×10^9 ions cm^{-2} s^{-1} the D peak is only faintly present below the critical fluence for track overlapping at around 2×10^{13} ions cm^{-2}. Above the critical fluence the D peak becomes distinct.



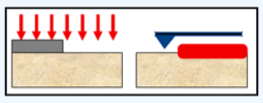


Further investigation shows a dose rate dependence of the defect creation and clustering in the material. For Fluxes of 1×10^{10} ions cm^{-2} s^{-1} the D peak is prominent below the threshold for track overlapping. This is due to a higher temperature induced during high flux irradiation at which the die vacancy clusters become mobile and migrate to create larger clusters. This phenomenon has been investigated by positron annihilation and it has been determined that the increase in the D peak can be related to the clustering of vacancies.

Analysis of change in function and properties

Swelling

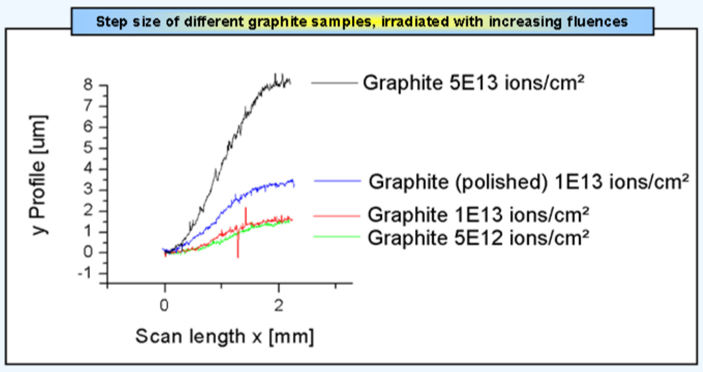
Problems:
-Radiation leads to swelling in the material
-Swelling leads to deformation
-Deformation influences the functionality of components (e.g. contact to cooling system)



Experimental setup:

-masked sample is irradiated-non-irradiated and irradiated surface is measured with a DEKTAK profilometer

Step size of different graphite samples, irradiated with increasing fluences

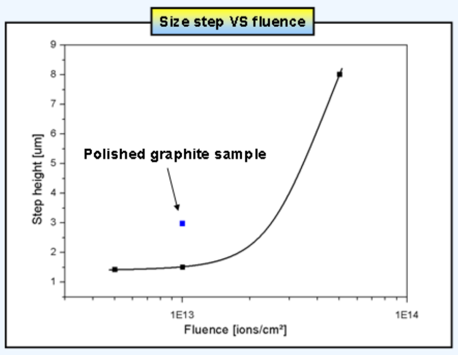


Investigation of irradiated isotropic graphite samples shows a radiation induced swelling of the sample. Analysis of the experiment shows that below a critical fluence of around 2×10^{13} ions cm^{-2} the swelling effects are small.

The experiment with an unpolished and polished sample, both exposed to fluences of 1×10^{13} ions cm^{-2}, shows larger swelling for the polished sample, indicating a higher sensitivity of the polished surface to ion disordering.

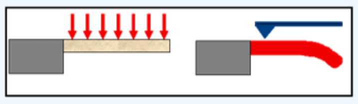
Fluence in ions/cm²	H in Gpa
5,00E+12	1,4325
1,00E+13	1,5174
5,00E+13	8,0197

Size step VS fluence



Induced stress

-Radiation induced swelling leads to stress
-stress results in bending
-bending can be measured
-stress is calculated from measured data



Experimental setup:

-Free hanging cantilever sample (fixed on one side) gets irradiated

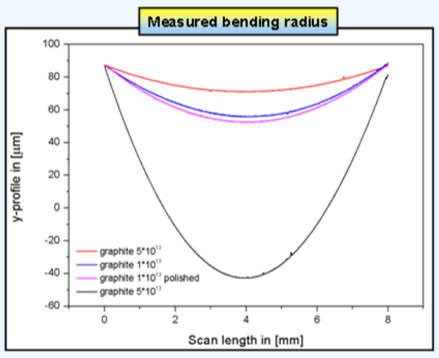
-Bending radius of the irradiated sample is measured using a DEKTAK profilometer

Stoney's formula

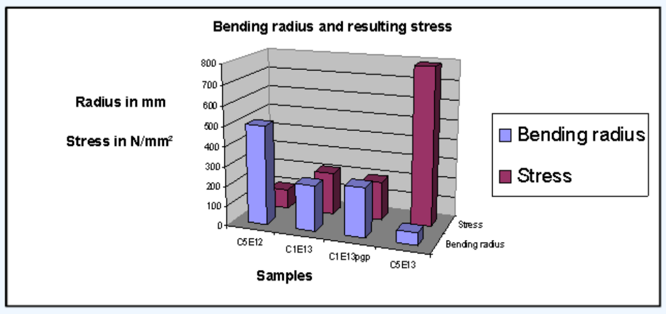
$$\sigma_{rr}^{f} \simeq -\frac{E_s h_s^2}{6(1-\nu_s)h_f}\frac{1}{R_r}$$ [1]

E_s = elastic modulus
h_s = height of sample
h_f = range of the ion in cantilever
R_r = bending radius
ν_s = Poisson's number
σ^f_{rr} = stress

Measured bending radius



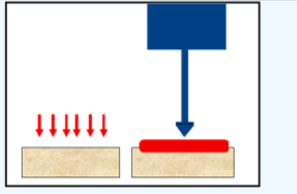
Analysis of irradiated isotropic graphite cantilevers with a profilometer show the results of the radiation induced stress in the form of a bending of the samples. Above a critical threshold of a fluence about 2×10^{13} ions cm^{-2} the swelling induced stress is significantly increased. This is indicated by the increase of the bending radius.



Swelling induced stresses were calculated with the obtained data from the DEKTAK profilometer using Stoney's formula [1].

Hardening

-Radiation induces hardening
-hardening leads to embrittlement
-embrittlement endangers mechanical stability and integrity of components

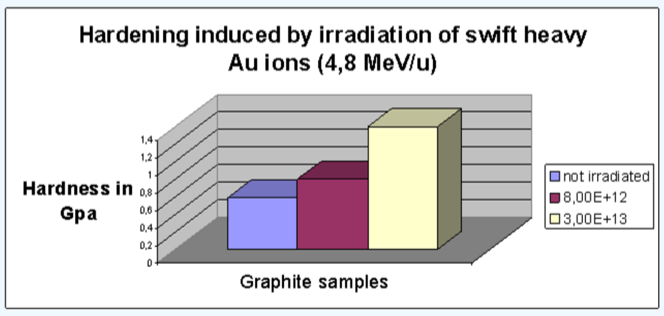


Experimental setup:

-isotropic graphite samples get irradiated

-Hardness is measured using a nano indenter

Investigations of irradiated isotropic graphite samples show an increase in hardness of irradiated material. The increase in hardness is higher above fluences of 2×10^{13} ions cm^{-2}.



The material hardens under the swift heavy ion irradiation due to the aggregation of structural defects and grain and domain boundaries which are obstacles for dislocation[3]. Hardening also points to the formation of other carbon structures like glassy carbon inside the swift heavy ion irradiated graphite[3].

Sample	H (Gpa)
not irradiated	0,59
8,00E+12	0,8
3,00E+13	1,4

Conclusion and outlook

The performed experiments showed the presence of a critical threshold in fluence for the swift heavy ion graphite material. This fluence is for Au ions of around 2×10^{13} ions cm^{-2} corresponding to the track overlapping of the ion tracks inside the material.

The variation of the flux during the irradiation experiment showed dose rate dependence of the creation of vacancy clusters. This is due to a temperature increase in the material during high flux irradiation which promotes the mobility of the vacancies and creation of bigger vacancy clusters inside the material.

Future experiments aim towards further characterization of the changes of the thermo mechanical properties in graphite materials induced by irradiation of swift heavy ions. Planned experiments include:
-Nano indentation at high strain rates to characterize the impact resistance changes induced by swift heavy ions
-Thermal conductivity investigation by laser flash methods
-In-situ monitoring of electrical resistivity during heavy ion irradiation
-The role of creep behavior in swift heavy ions induced deformation

References

[1] - N.Schwarzer, F.Richter – On the determination of film stress from substrate bending: Stoney´s formula and its limits

[2] - 2007 - Pimenta et al. - Studying disorder in graphite-based systems by Raman spectroscopy

[3] - I. Manika, J. Maniks, R. Zabels, J. Gabrusenoks, M. Krause, M. Tomut & K. Schwartz (2012): Nanoindentation and Raman Spectroscopic Study of Graphite Irradiated with Swift 238U Ions, Fullerenes, Nanotubes and Carbon Nanostructures, 20:4-7, 548-552

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