Irradiation damage in nuclear graphite at the atomic scale

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Framework: decommissioning of graphite from "UNGG"

- Graphite = moderator in "UNGG" → neutrons, temperature
- In 11 years of exploitation **2.6 dpa**
- Activation of some impurities

i-graphite contains ^{14}C & ^{36}Cl ($\frac{1}{2}$ life 5 500 & 3×10^5 years)

Foreseen Solutions:

- Decommissioning under water
- Treatment to eliminate ^{14}C & ^{36}Cl , (^3H)

} interaction: graphite $\leftrightarrow$ H_2O , O_2 , CO_2 ...
depends on the **localization of ^{14}C & ^{36}Cl** after irradiation

- **Graphite microstructure evolution under irradiation?**
 - radiation damages and swelling
 - link between XRD - HRTEM images and atomic scale structure

cm

nm

Heggie et al (2011)

Scenario of swelling

1. point defects by irradiation
2. wrinkling
3. sliding
4. formation of dislocations

Based on:

- HRTEM observations
- Atomic scale (DFT - MD) calculations
- Wigner energy estimations

Our goals:

- Link between swelling - atomic configurations for irradiated single crystal
- Link between HRTEM images and atomic configurations for irradiated single crystal for poly-crystal

Primary ballistic damage by displacement cascade, with primary knocked on atom with 0.5 to 10 keV

if point defects then

Dose effect by Frenkel pairs accumulation

- Creation of Frenkel pairs every 2 ps
high dose rate
- Dose up to 10 dpa
- constant pressure and constant temperature

Frenkel pairs

HRTEM simulation (NCEMSS or Multi-slice code)

Graphite

XRD simulation (code Debye)

Scherrer's formula

Bragg's law

Topological analysis: local curvature criterion (graphite if curvature > 32°)

defects in blue

graphene sheets in red

Shrinking in the basal plane

defects in blue
graphene sheets in red

Swelling along c-axis

a and c from XRD - Bragg's law

1. Point Defects

interstitial

2. Pinning on small disordered cluster and Wrinkling of graphene sheets

swelling / shrinking

3. Growth of amorphous pockets

La and Lc - Scherrer's formula

Decrease of graphene size with increasing dose

→ small graphene sheets in "noodles shape"
= BSU (Basic Structural Units)

What simulations show:

graphite amorphizes

MD simulation

exp. J. Pageot (CEA)

Creation of 20 crystallites, stacked along c-axis, with angles randomly distributed between 0° and 20°

Scherrer $\rightarrow L_c = 7$ nm, $L_a = 22$ nm
 initial $\rightarrow L_c = 15$ nm, $L_a = 35$ nm

Close to experiments

HRTEM image shows dislocation
(edge or screw)

MD snapshot shows wrinkles

HRTEM image shows nano-pore
MD snapshot shows nano-pore

SIMULATION

HRTEM images
→ dislocations

but

MD snapshots
→ wrinkles

Irradiation damages in graphite single crystal: Frenkel Pair Accumulation method

➤ Scenario in 3 steps:

1. Point Defects
2. Pinning + Wrinkling of graphene sheets
3. Growth of amorphous pockets → Amorphisation

➤ Swelling due to both defects + wrinkling of graphene sheets

➤ Microstructure from XRD - HRTEM

1. Link dislocations - wrinkles ??
2. Basic Structural Units or amorphous??

Thanks for listening!

A. Chartier, L. Van Brutzel and J. Pageot, Carbon 133 (2018) 224.

A. Chartier, L. Van Brutzel, B. Pannier, and Ph. Baranek, Carbon 91 (2015) 395.

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0.01 dpa

Local curvature threshold

Remaining graphene sheets

Plans pinned by **disordered amorphous** zones

Graphene sheets are discriminated by **local curvature**

- average curvature radius
- size of graphene clusters

Graphite – displacement cascade

Graphite - Analysis with DXA, HRTEM and XRD

- Visualization of defects in graphene planes (MD and HRTEM)
- Visualization of wrinkles (MD analysis)
- Swelling from size of box (not easy from XRD)
- amorphization at 0.10 dpa (SAED and XRD)

Graphite – simulated XRD

5. nm

0.00 dpa

equivalent to 0.50 dpa

equivalent to 1.00 dpa

5. nm

equivalent to 1.2 dpa

equivalent to 2.50 dpa

equivalent to 3.50 dpa

Graphite – simulated HRTEM

5. nm

0.00 dpa

equivalent to 0.50 dpa

equivalent to 1.00 dpa

5. nm

equivalent to 1.2 dpa

equivalent to 2.50 dpa

equivalent to 3.50 dpa

Graphite – simulated SAED

0.00 dpa

equivalent to 0.50 dpa

equivalent to 1.00 dpa

equivalent to 1.2 dpa

equivalent to 2.50 dpa

equivalent to 3.50 dpa