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Irradiation damage in nuclear graphite at the atomic scale

A. Chartier, L. Van Brutzel, and J. Pageot
Irradiated graphite in graphite-gas nuclear reactors

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 & $3 \times 10^5$ years)

Foreseen Solutions:
- Decommissioning under water
- Treatment to eliminate $^{14}$C & $^{36}$Cl, ($^3$H)
  interaction: graphite ↔ $H_2O$, $O_2$, $CO_2$...
  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
Microstructure / swelling by irradiation

 رسول close single crystals swell/shrink

Shen et al (2015)

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

Heggie & Telling (2007)
Modelling radiation damages

**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
Analysis of MD snapshots

**HRTEM simulation**
(NCEMSS or Multi-slice code)

![HRTEM simulation diagram](image)

**XRD simulation**
(code Debyer)

![XRD simulation graph](image)

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

![Topological analysis image](image)

Graphite

- Incident e-
- Transmitted e-
- Graphene sheets in red
- Defects in blue

**Scherrer's formula**

\[
L_c = \frac{K \lambda}{b \cos \theta}
\]

**Bragg's law**

\[
c \times 2 = n \lambda
\]

**FWHM**

**Peak position**

- X-ray pattern
- (002) - Pseudo Voigt
- (010) - Pseudo Voigt
- (101) - Pseudo Voigt

**Graphite**

- MD - FPA
- MD - NCG
- exp
- Balkur
- Monti
- Babu
- Park
- Zhou
- Freeman
- Huang
- Seo

**Defects in blue**

**Graphene sheets in red**

**MD from MD**

**Dose (dpa)**

- 0.0
- 0.08
- 0.16
- 0.24
- 0.32

**Truncation (%)**

- 0
- 5
- 10
- 15
- 20

**MD - Freeman**

**MD - Monirv**

**MD - Koike**
Frenkel pairs accumulation

Shrinking in the basal plane

Swelling along c-axis

10 nm

defects in blue

graphene sheets in red
1. Point Defects

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

3. Growth of amorphous pockets
Decrease of graphene size with increasing dose

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

What simulations show:

graphite amorphizes
Nano-crystalline graphite

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

Scherrer’s formula

Scherrer → $L_c = 7 \text{ nm}$, $L_a = 22 \text{ nm}$

initial → $L_c = 15 \text{ nm}$, $L_a = 35 \text{ nm}$

Close to experiments
HRTEM images of nano-crystalline graphite

HRTEM image shows dislocation (edge or screw)
MD snapshot shows wrinkles

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

70 nm
HRTEM images of nano-crystalline graphite

HRTEM images → dislocations

but

MD snapshots → wrinkles
Conclusion - summary

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!


Plans pinned by disordered amorphous zones

Graphene sheets are discriminated by local curvature
→ average curvature radius
→ size of graphene clusters

0.01 dpa
Local curvature threshold
Remaining graphene sheets
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
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

5. nm 5. nm 5. nm
Graphite – simulated HRTEM

- 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
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