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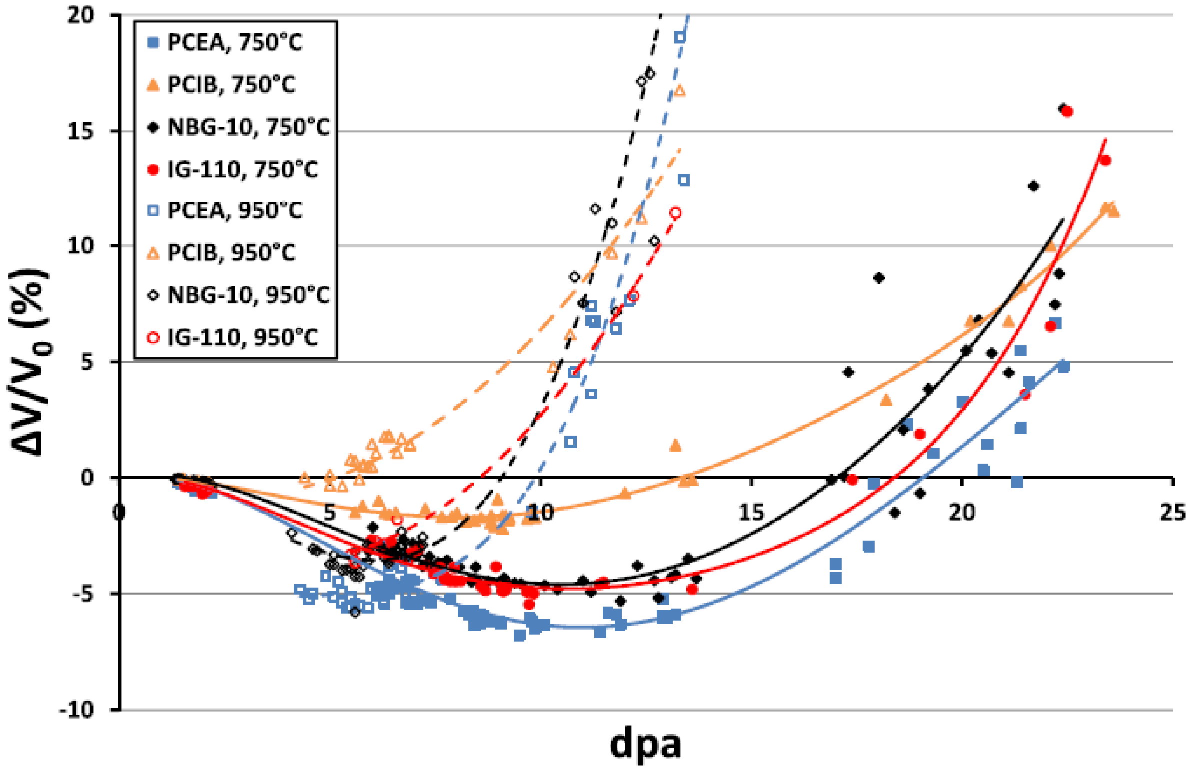
# Graphite Irradiation Data

Status Update

ART Advanced Reactor Technologies
GAS-COOLED REACTORS

# Future ASME Code Development and Reactor Deployment

- ASME code rule modification for support of new reactor concepts and commercial vendors
  - Key properties include, but is not limited to, the strength, elastic modulus, coefficient of thermal expansion, **dimensional change**, etc.
  - The turnaround dose signals when many other properties will significantly deteriorate.
  - Irradiation temperature is a key parameter effecting turnaround dose.
- Dependent on coke source, and manufacturing process, each nuclear graphite has a unique response.




M.C.R. Heijna et al. / Journal of Nuclear Materials 492 (2017) 148-156

# Can We Define a Universal Response for Nuclear Graphite?

- Qualifying each candidate grade of nuclear graphite, at various temperature regimes, is unreasonable.
  - New irradiation campaigns are time consuming
  - Can be extremely expensive (millions)

- For development of ASME code rule, and to inform vendors, can a universal response be defined?

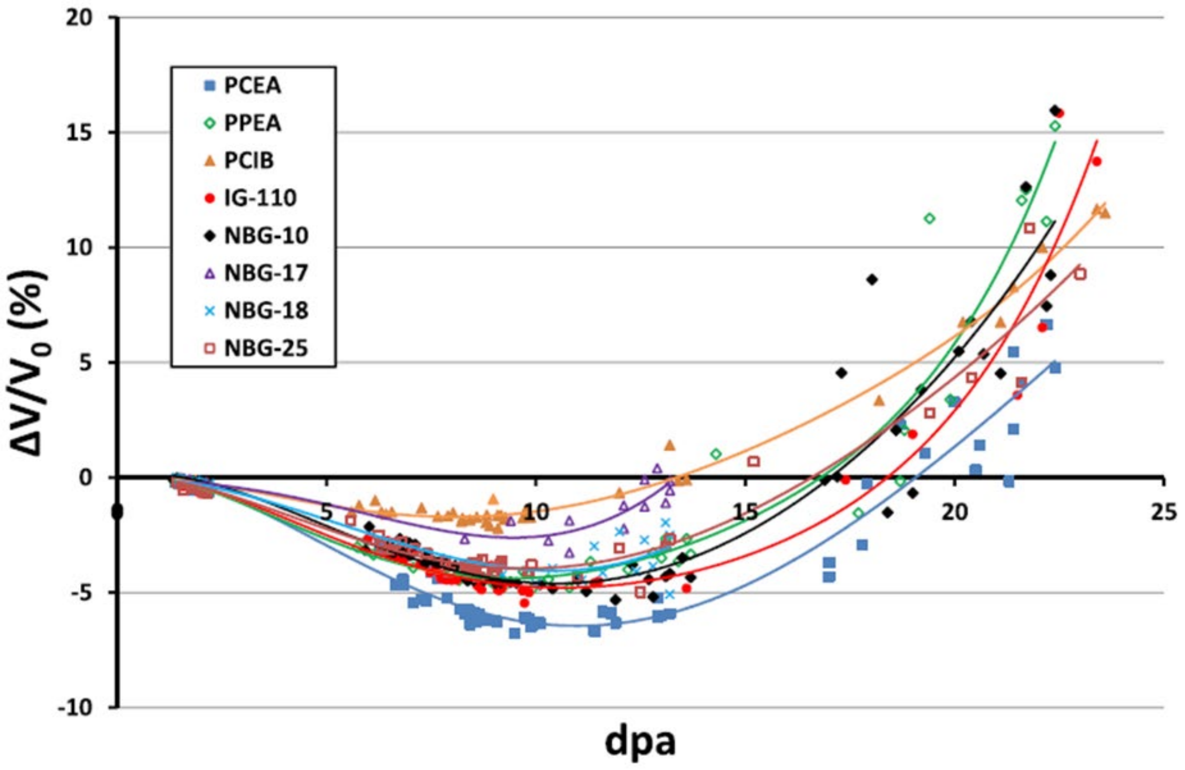



Fig. 2. Dimensional change as function of dpa at 750 °C.

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# Candidate Grades for Reactor Concepts and Obsolete Grades

- Past irradiations primarily focused for graphite use in prismatic reactor concepts.
- Since that time, many new reactor concepts and new candidate grades have emerged.
- Code development and manufactures still need irradiation data for property changes.
- New irradiation campaigns would be a significant financial burden and time consuming

## Major Grades in AGC – 1

| Graphite | Manufacturer | Coke Type | Grain Size | Forming Process | Remarks |
|---|---|---|---|---|---|
| IG-110 | Toyo-Tanso | Petroleum coke | Fine grain | Iso-molded | Prismatic fuel element, replaceable reflector, and core support pedestals |
| IG-430 | Toyo-Tanso | Pitch coke | Fine grain | Iso-molded | Prismatic fuel element, replaceable reflector, and core support pedestals |
| NBG-18 | SGL | Pitch coke | Medium grain | Vibramolded | Prismatic replaceable reflector Pebble bed reflector structure |
| NBG-17 | SGL | Pitch coke | Medium grain | Vibramolded | Prismatic fuel element and replaceable reflector. Pebble bed reflector structure and insulation blocks |
| PCEA | GrafTech | Petroleum coke | Medium grain | Extruded | Prismatic fuel and replaceable block. Pebble bed reflector and insulation blocks |
| H-451 | SGL | Petroleum coke | Medium grain | Extruded | Historical reference, no longer available. |

# Dimensional Change Theory

$$\frac{dG_x}{d\gamma} = A_x\left(\frac{1}{X_c}\frac{dX_c}{d\gamma}\right) + (1 - A_x)\left(\frac{1}{X_a}\frac{dX_a}{d\gamma}\right) + f_x$$

$x$: Direction (not specific)

$\gamma$: Fast neutron fluence (n/m$^2$)

$A_x$ : Structural factor: ration of grains to *c*-axis within x direction (i.e., purely isotropic $A_x = 0.5$)

$X_a, X_c$ : Fractional dimensional change to *a*- and *c*- axes

$f_x$: Fractional dimensional change from pores per neutron fluence

**Integration yields:** $\boldsymbol{G_x(\gamma) = A_x G_c(\gamma) + (1 - A_x) G_a(\gamma) + F_x(\gamma)}$

Linear

Non-linear

J. E. Brocklehurst, B. T. Kelly, Carbon, 31, 155–178 (1993).

# Dimensional Change Theory

$$G_x(\gamma) = A_x G_c(\gamma) + (1 - A_x) G_a(\gamma) + F_x(\gamma)$$

If the porosity function is assumed to be quadratic (i.e., porosity decreases then increases with increasing dose), the semi-empirical relationship is given by:

$$G_x(\gamma) = a_1 \gamma^2 + a_2 \gamma$$

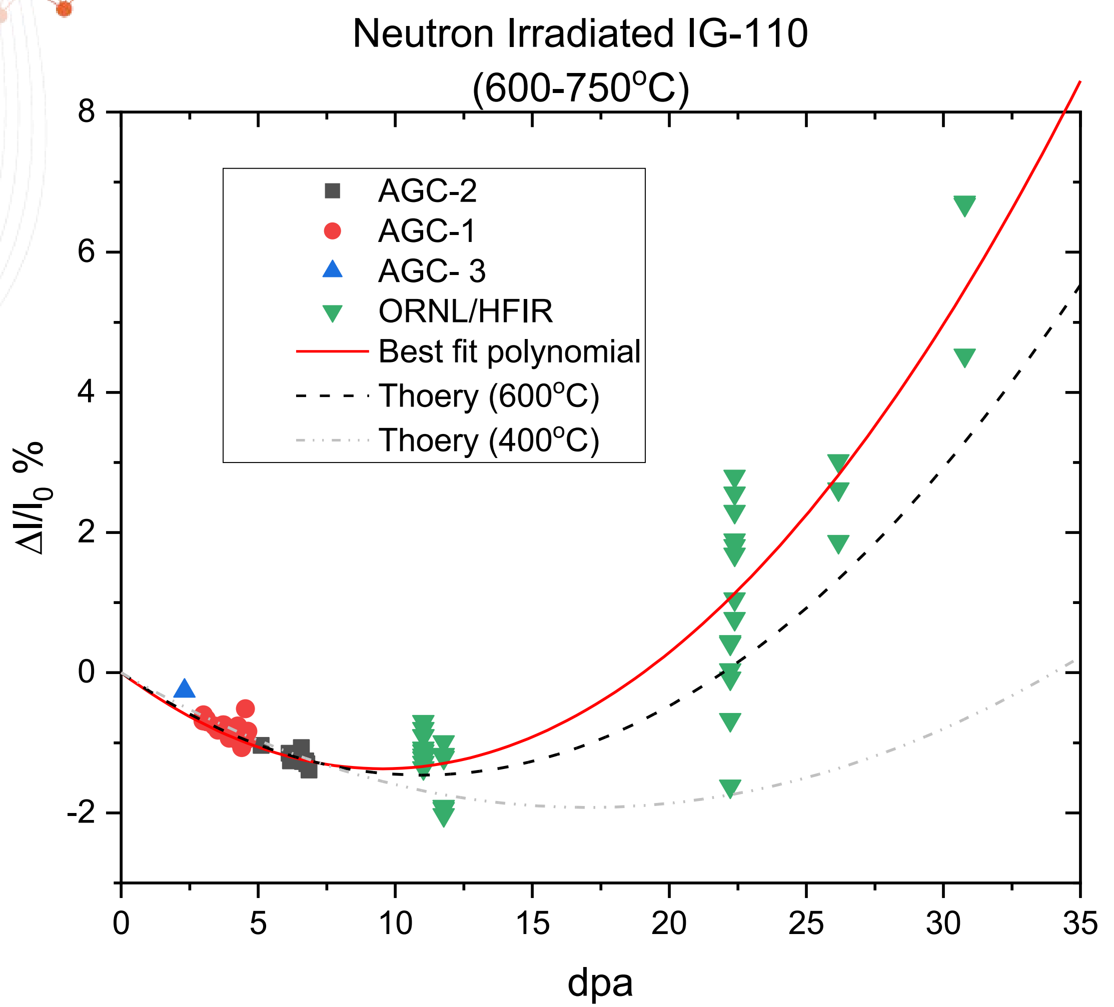
Where $a_1$ and $a_2$ are temperature dependent constants.

For IG-110, literature gives the following values:

| **Grade** | **IG-110** | |
|---|---|---|
| Irradiation temperature (°C) | $a_1$ | $a_2$ |
| 400 | 0.279 | -1.64 |
| 600 | 0.450 | -1.86 |
| 800 | 0.821 | -2.19 |

S. Mohanty, S. Majumdar, M. Srinivasan, U.S.N.R. Commission HTGR Graphite Core Component Stress Analysis Research Program – Task 1 Technical Letter Report (2011)

# Theoretical vs Empirical Modeling




- Between best fit polynomial, theory at 400 and 600°C, there is little to no deviation *before* turnaround dose.
- Shown is that the methodology, theoretical or purely empirical, is not significant until after turnaround.
- Proposed is to empirically fit all available data to inform ASME code development.

# All Graphites Behave Similar – Can we Predict the %Δ?

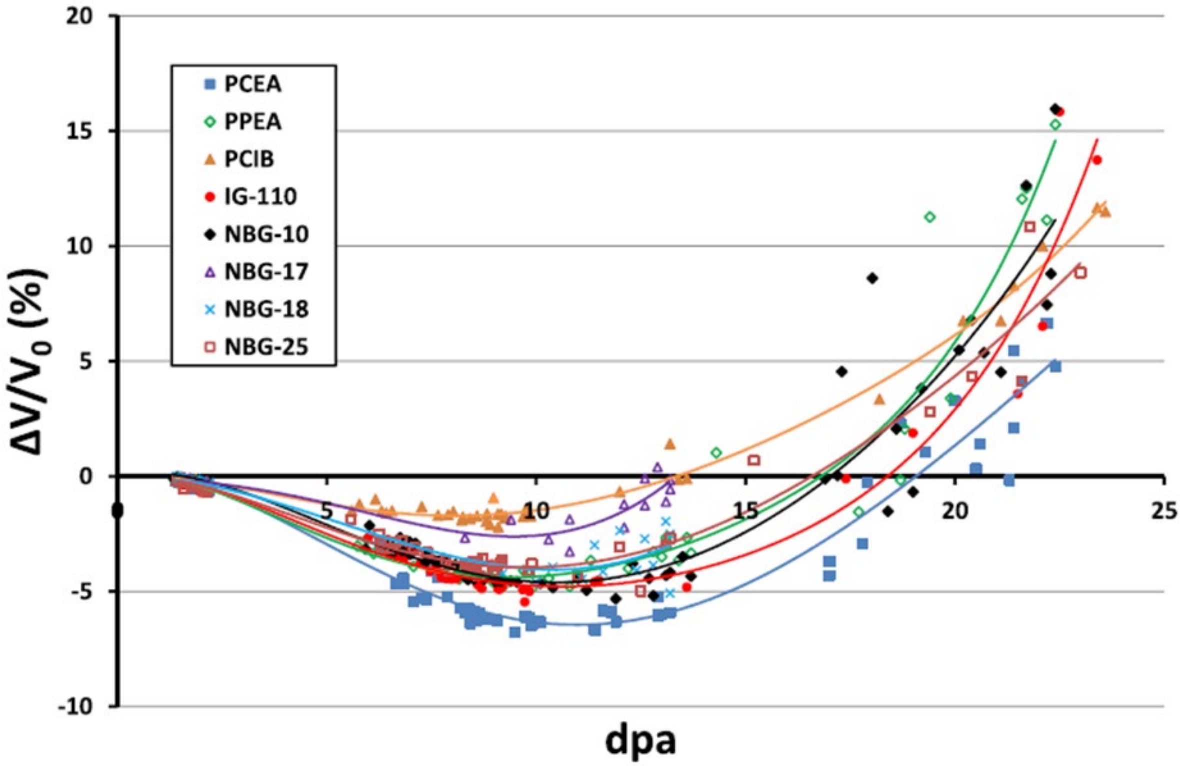



Fig. 2. Dimensional change as function of dpa at 750 °C.

- Proposed is empirical polynomial fitting *up to* turnaround.
- To be used as a reference for ASME code / commercial vendors, for dimension change (%Δ) at a given dose.
- Shown is 5$^{th}$ order polynomial fits, which accurately captures the delayed dimensional change response.
- Literature reviews suggest PCEA to have the largest dimensional change of candidate grades.
- Can PCEA data be used as a ‘lower bound’ for all candidate grades in the design code (% dimensional change).

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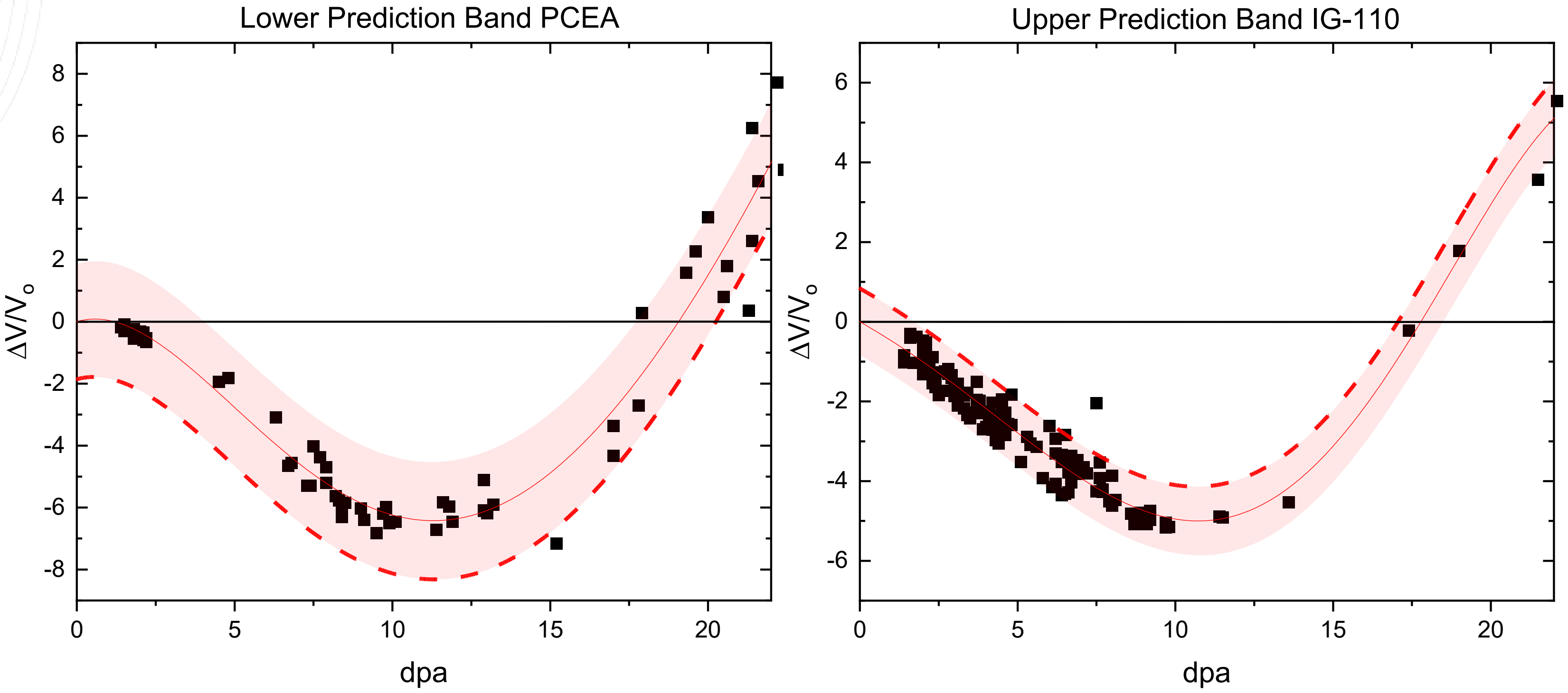
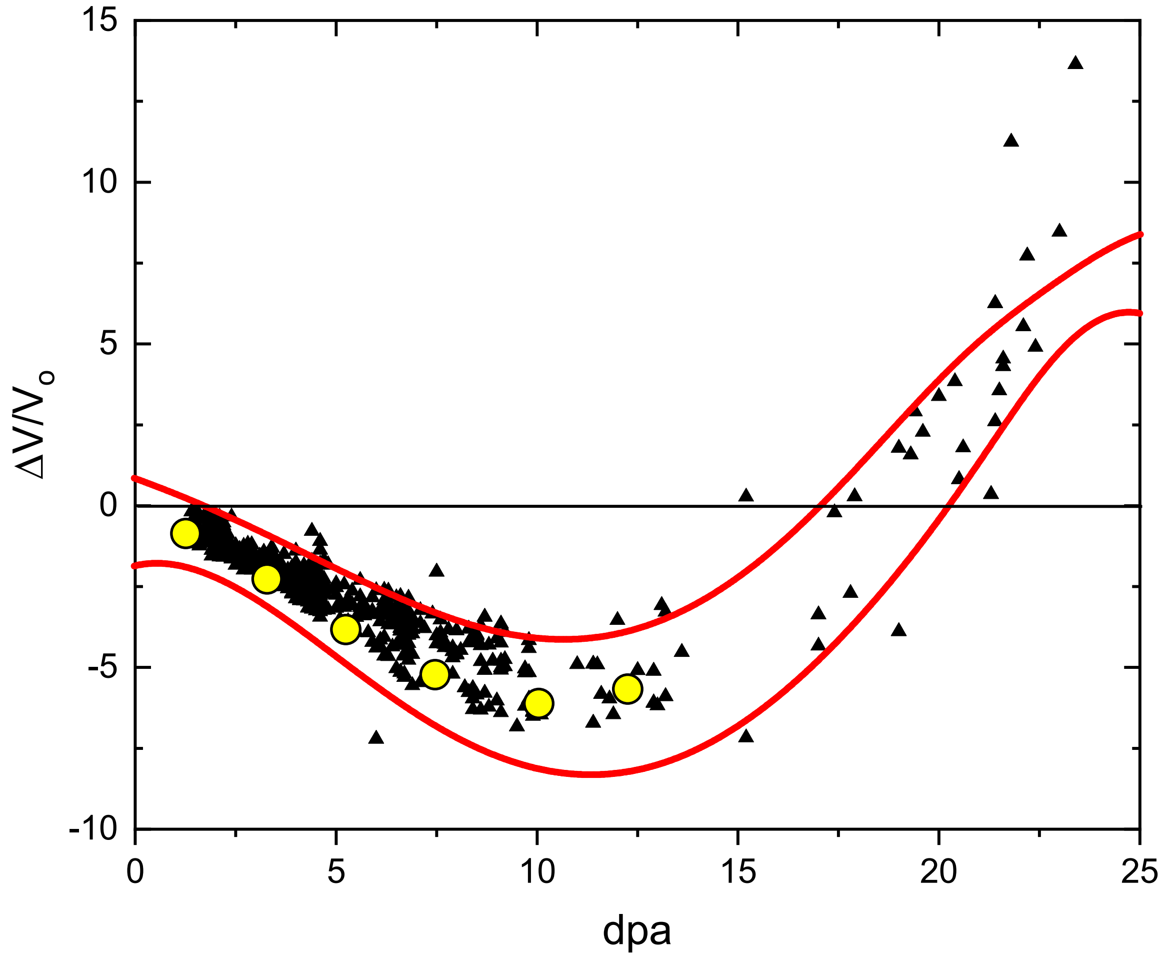
# Irradiation Data From AGC1-3 and InnoGraph



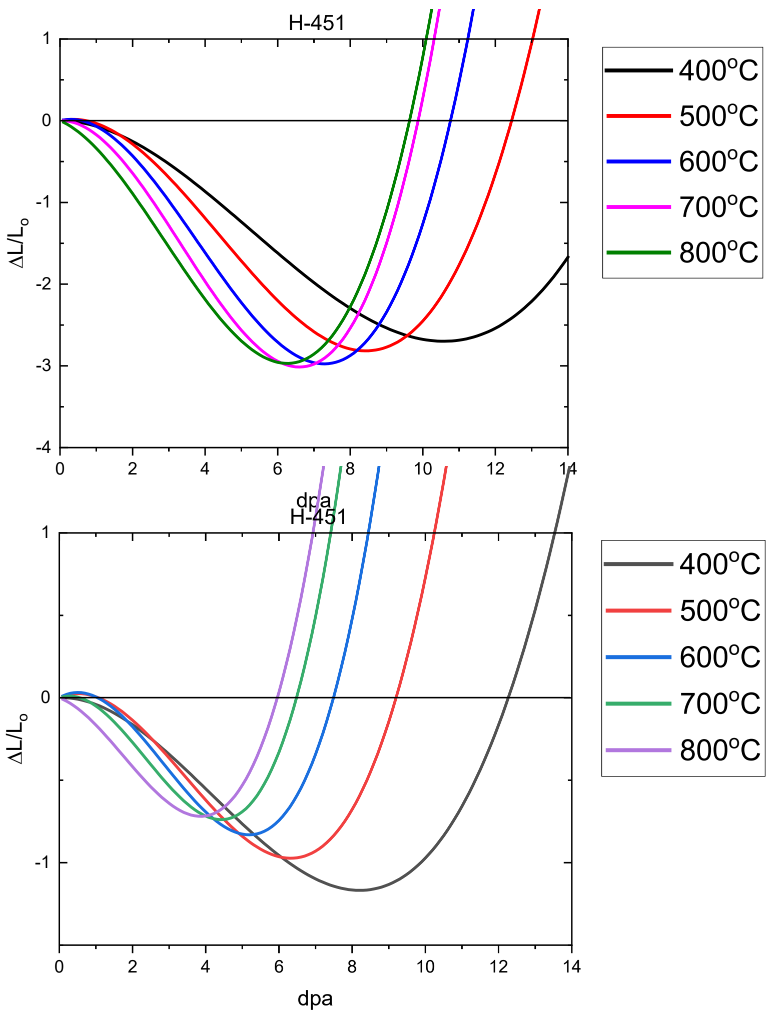

- Define a 95% lower prediction band with PCEA.
  - Medium grained, extruded
- Define a 95% upper predication band with IG-110
  - Fine grained, isomolded

# Irradiation Data From AGC1-3 and InnoGraph




- Nuclear graphite grades: NBG-10, NBG-17, NBG-18, NBG-25, PCEA, H-451, IG-110, and 2114.
- ***Needs*** to be refined by temperature.
- For adequate fitting, irradiation data was taken from 400-800°C.
  - Currently collaborating with ORNL to compile and produce open-source data for analysis.
- With enough data, additional refinements may be possible.
  - Ex. by small, medium and large grained graphites.

# Turnaround Dose as a Function of Temperature

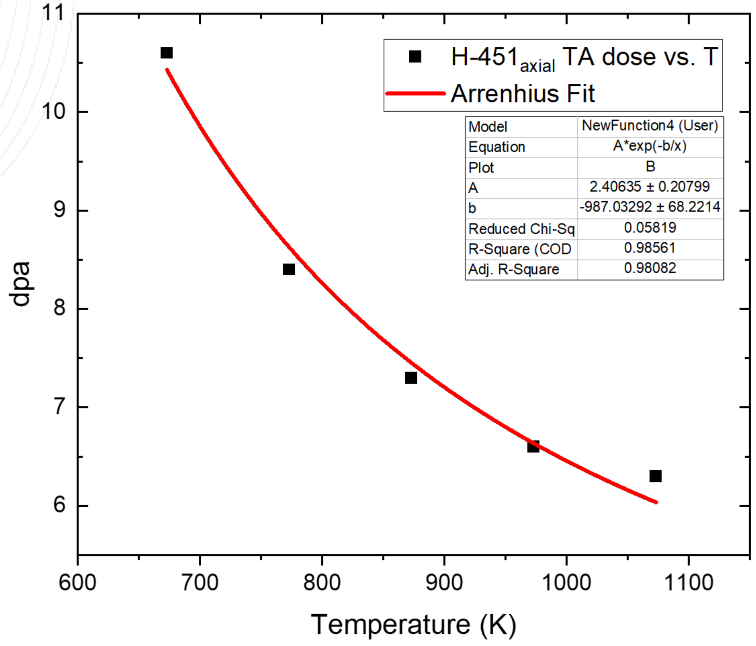


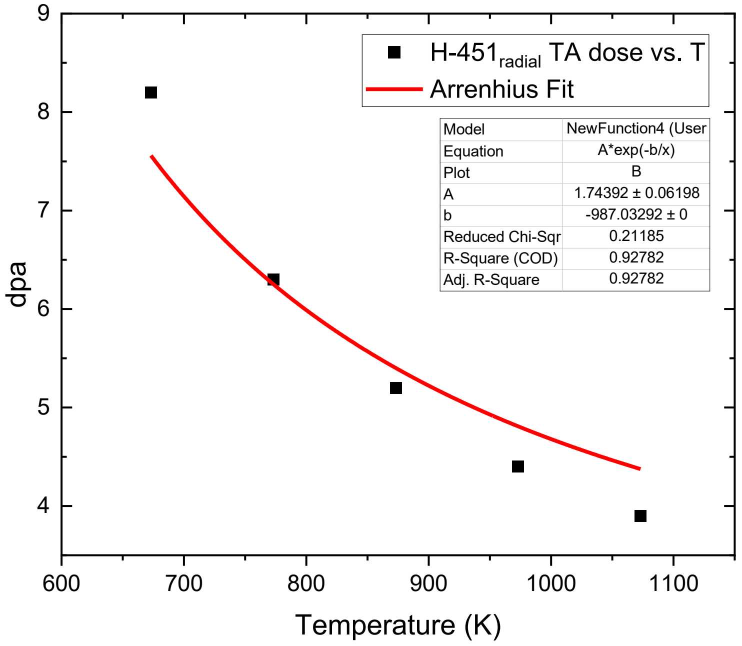

- Historical grade H-451 dimensional change model (3rd order polynomial).
- Identify turnaround dose as a function of temperature.
- Turnaround is a temperature dependent response (thermally activated).
- Define an Arrhenius function

$$\mathbf{TA}(T) = A\, exp\left(\frac{-E_a}{k_b T}\right)$$

- All graphites should have the same activation energy ($E_a$).

# Hold $E_a$ Constant and Allow Pre-exponential to Vary




- Plot and fit Arrhenius equation for H-451
- Activation energy should be constant and not orientation dependent.
- Allow the pre-exponential factor to vary.
- Assuming constant activation energy produces reasonable fit ($R^2$=0.92).
- To accurately estimate a universal activation energy, more data is needed.

# Pre-exponential Factor Could be Defined per Grade

$$\mathbf{TA}(T) = A\, exp\left(\frac{-E_a}{k_b T}\right)$$

$$A(E_o, A_x, \rho_o, CTE)$$

$E_o$= Elastic Modulus

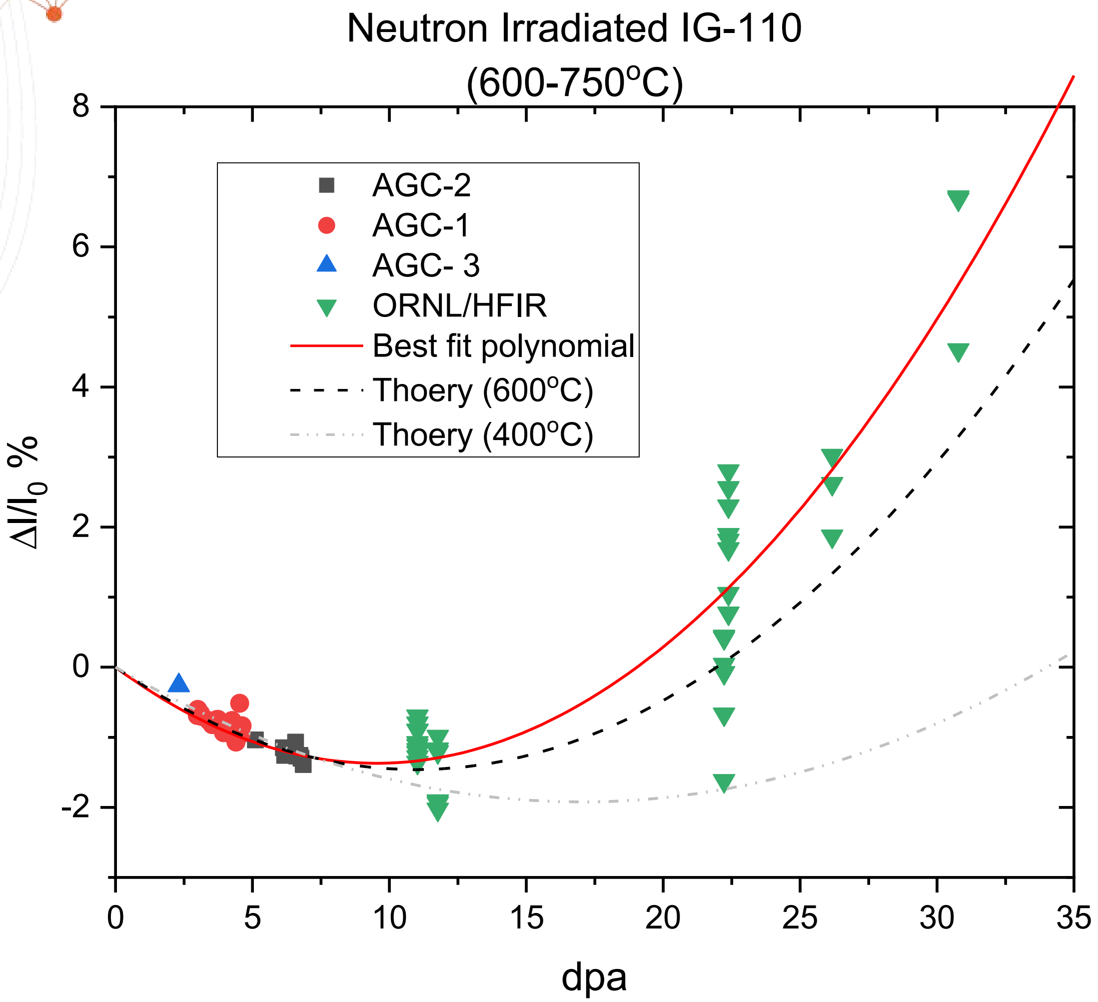
$A_x$= Anisotropic Factor

$\rho_o$= Density

$CTE$ = Coefficient of Thermal Expansion

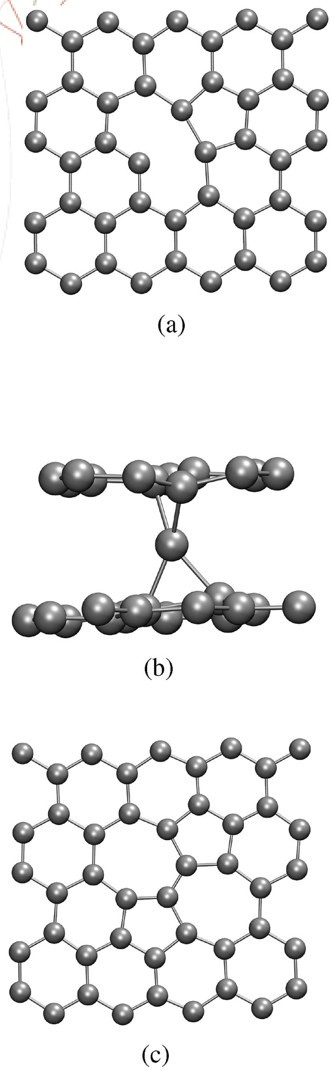
- Fundamentally, on the atomic scale, all nuclear graphites are the same. $Sp_2$ bonded Carbon with some degree of disorder.
- Variation in the irradiation response amongst grades comes from difference in the meso – macroscale features.
- The value of the pre-exponential factor could be defined by *baseline* properties specific to grade.
- Again, much more data is needed.

# Why Only Up to Turnaround Dose?

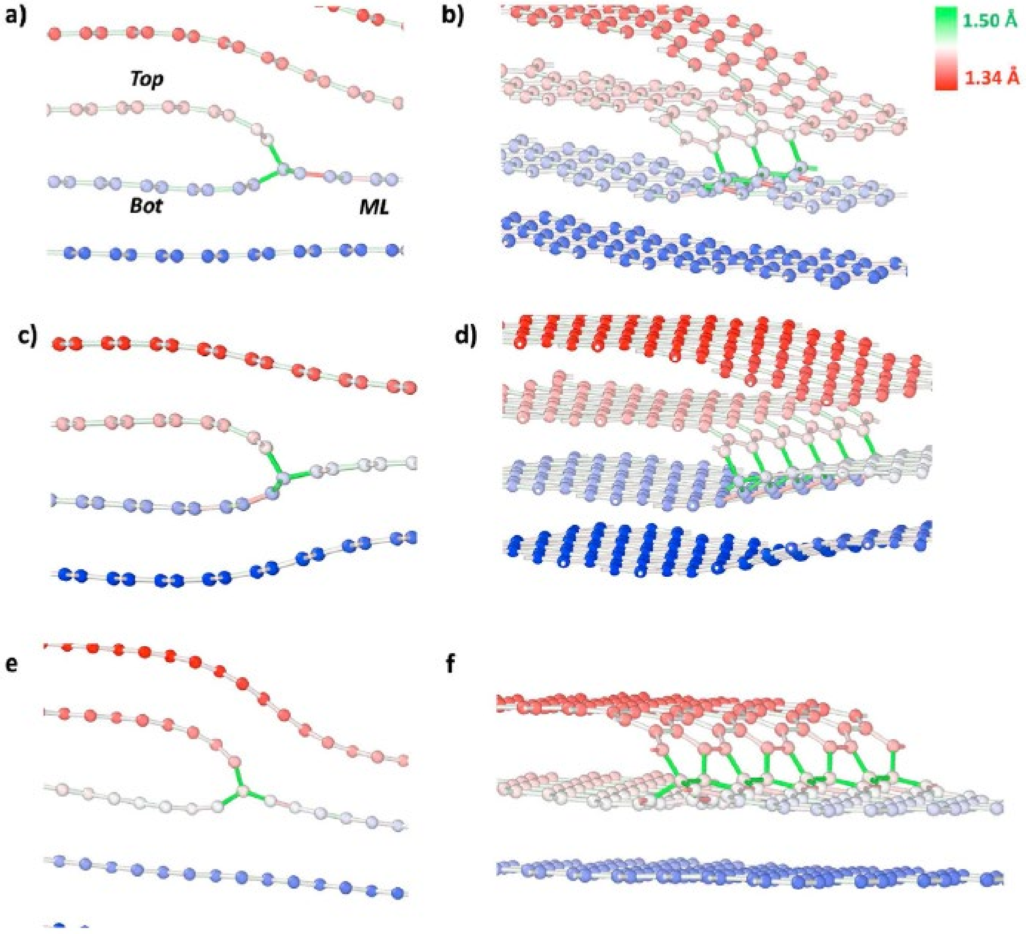



- Significant degradation to all properties beyond turnaround dose.
- Other properties such as creep must now be considered.
- The variance in data becomes significant.
- The amount of 'open-source' data is scant.
- To accurately describe the response of nuclear graphite past turnaround dose, a comprehensive understanding of the damage mechanisms is needed.

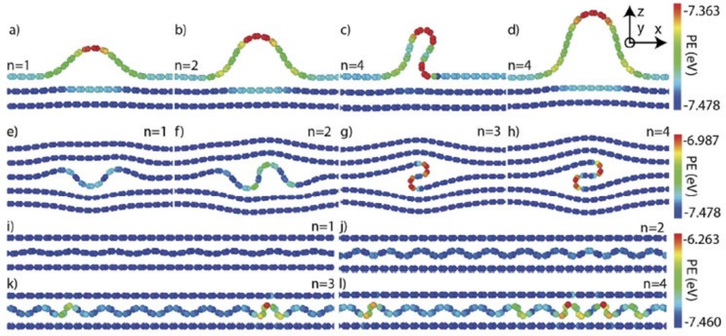
# Many Atomic Mechanisms are Energetically Favorable



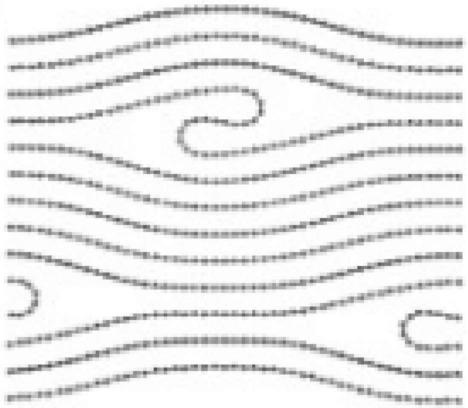

Mckenna, et al., Carbon, 99, 2016




McHugh, et al., Carbon, 188, 2022



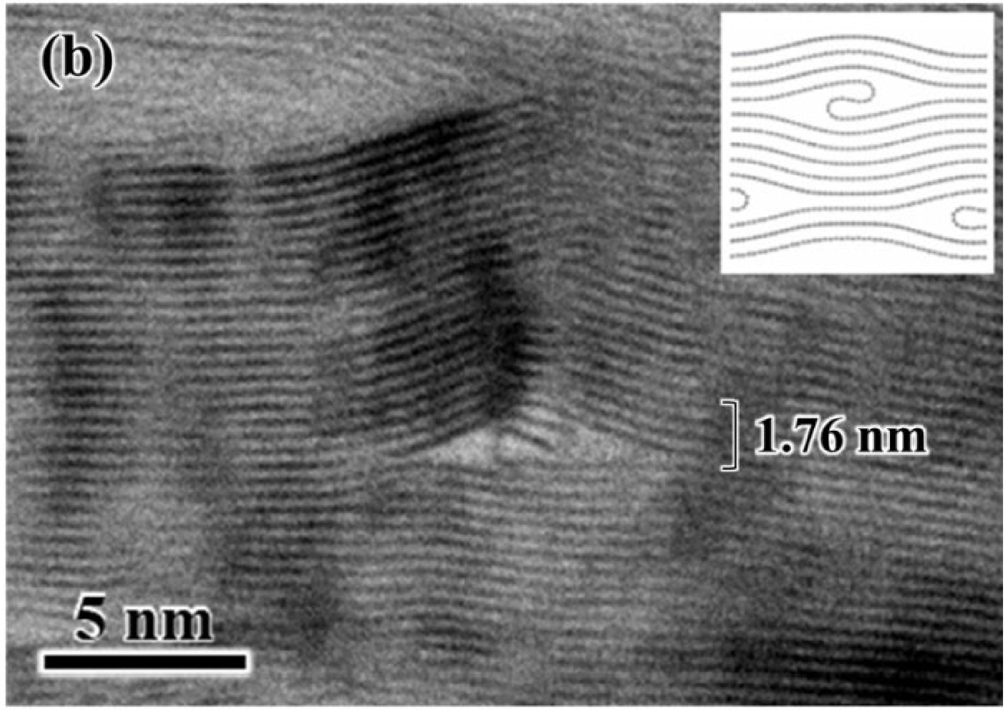

Gruber, et al., Scientific Reports, 6, 2016



Heggie, et al., Journal of Nuclear Materials, 413, 2011

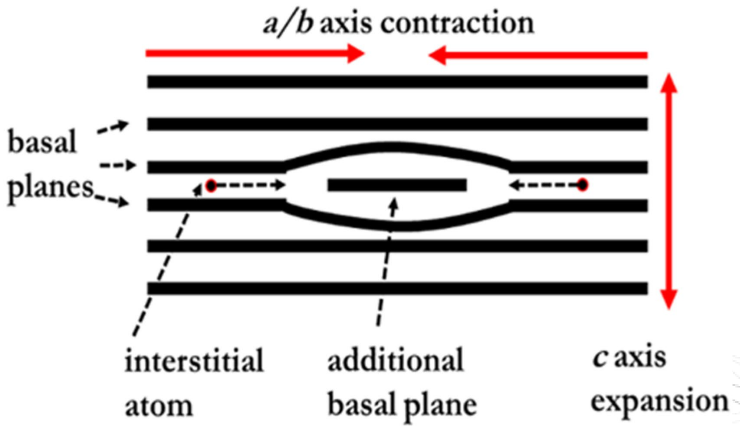
# First Evidence of ‘Ruck and Tuck’ Defect.

AGC-3 Specimen. IG-110 irradiated at ~800°C to 3.56 dpa.



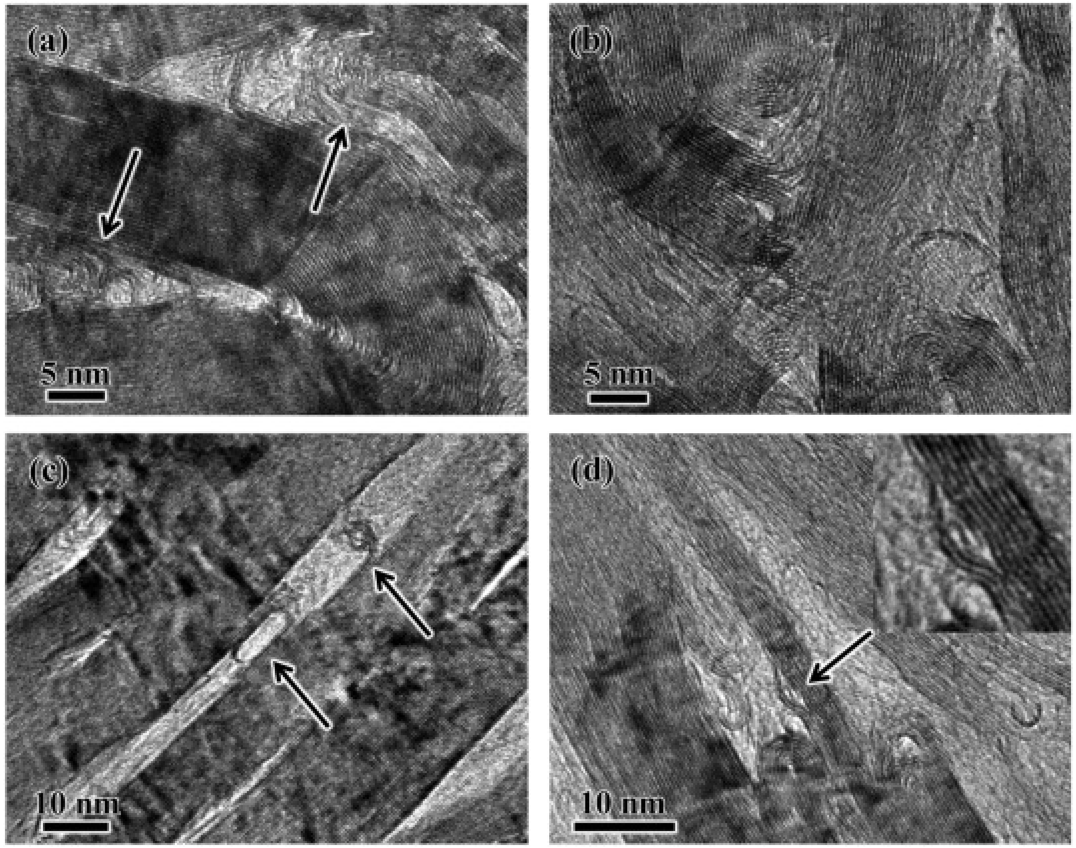

S.Johns, et al., Carbon, 159, 2020

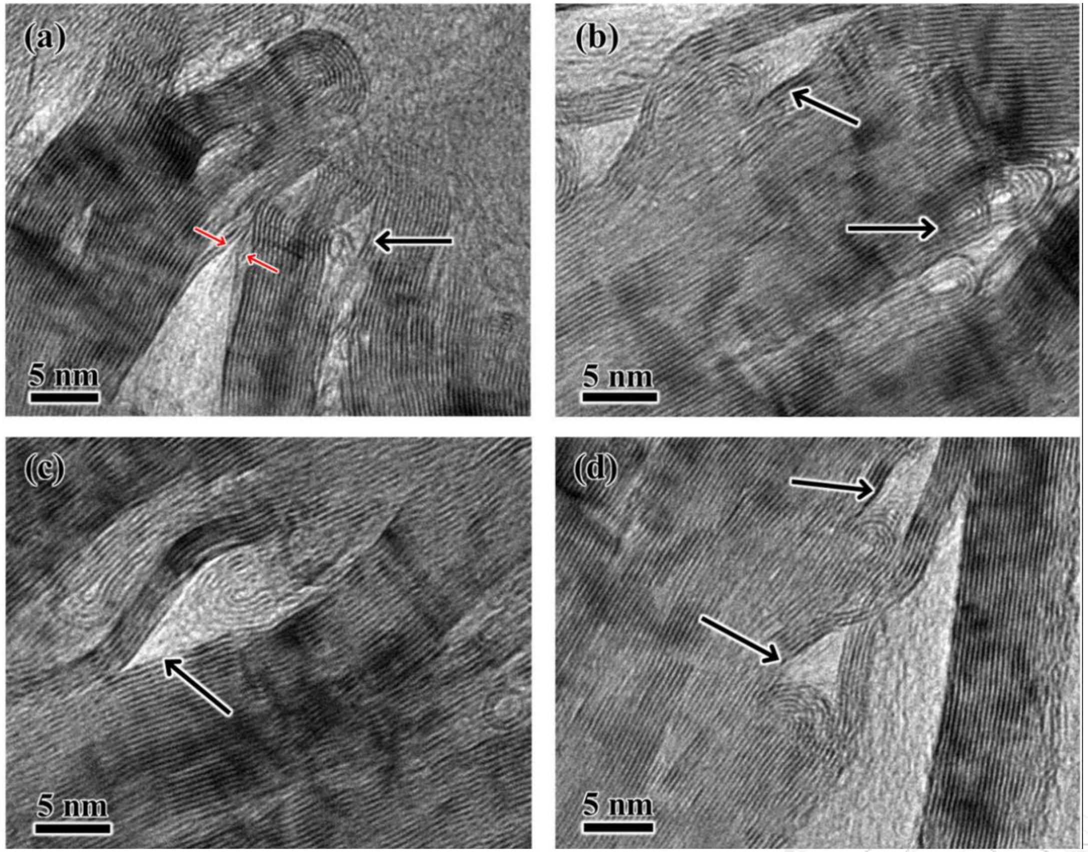
- First experimentally verified evidence of a ‘ruck and tuck’ defect in neutron-irradiated nuclear graphite.
- Provides an alternative theory to explain large *c*-axis expansion in which historical models failed.



# Zoo of Defects

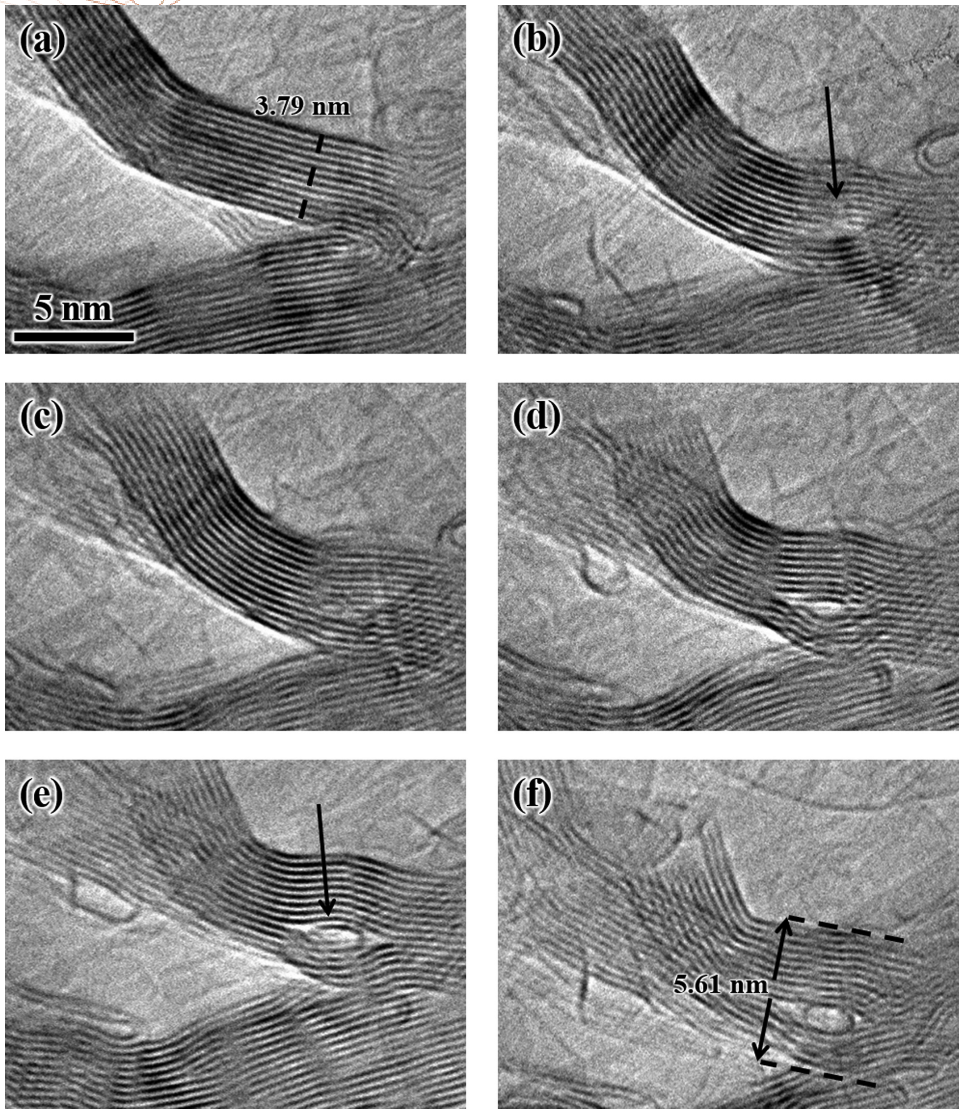
AGC-3 Specimen. IG-110 irradiated at ~800°C to 3.56 dpa.




S.Johns, et al., Carbon, 116, 2020

# Progress in Novel Experimental Techniques




S.Johns, et al., Carbon, 143, 2019

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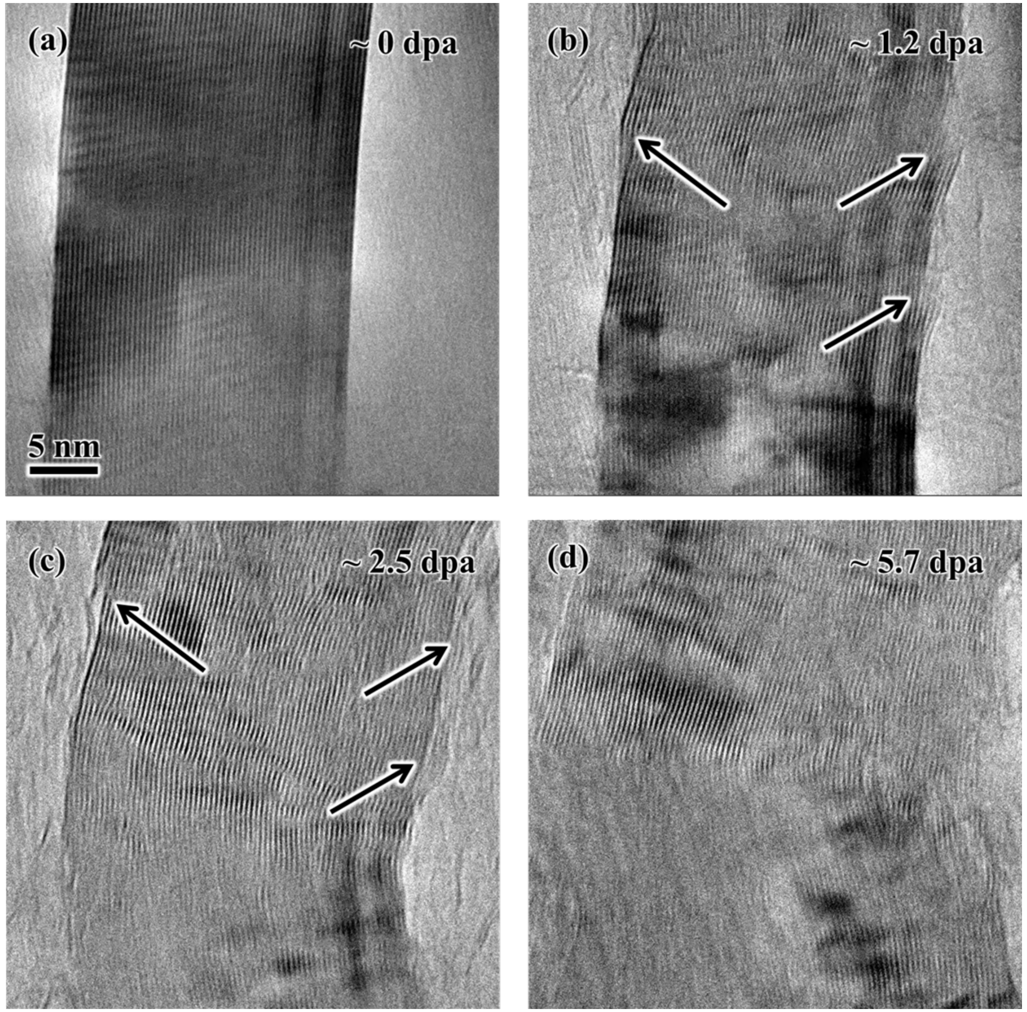
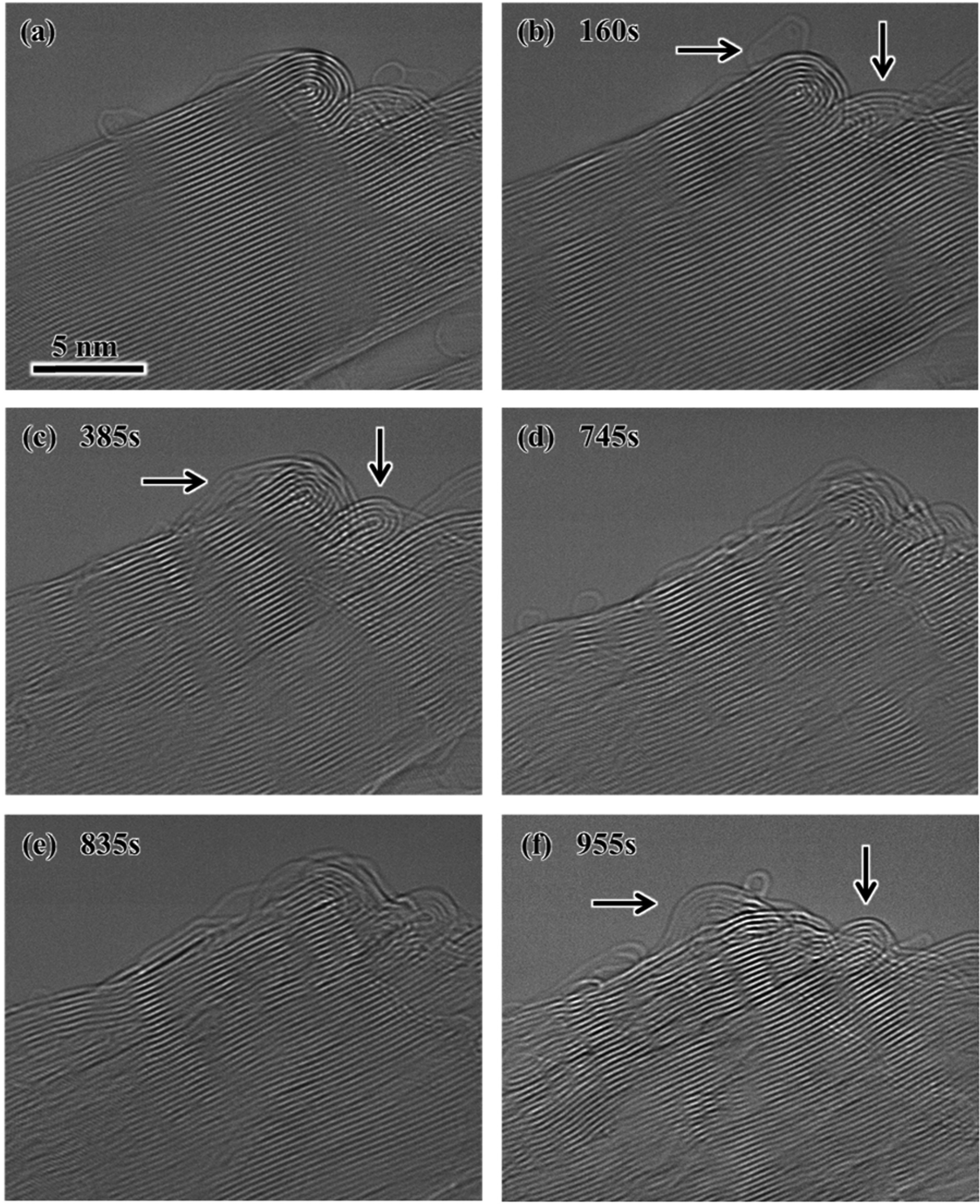
# Irradiation Induced Graphitization




- In-situ electron-irradiation conducted at 500°C.
- At lower doses, (b) & (c), additional layers are observed to 'template' onto the crystallite.
- At higher doses, (d), the crystallite becomes significantly disordered.

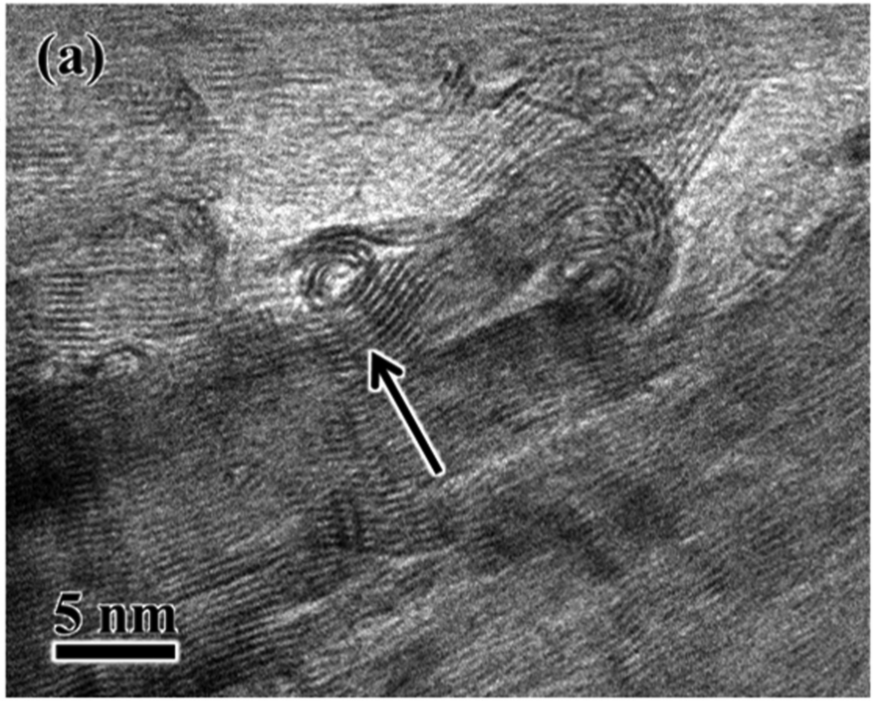
# In-situ Electron Irradiation

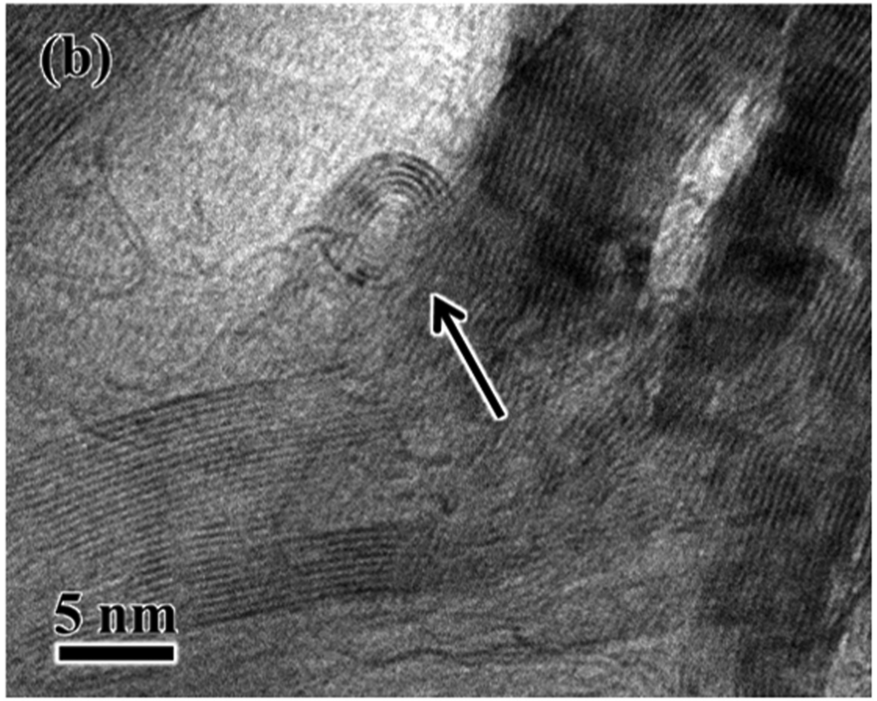
- Electron-irradiation conducted at 200keV at 800°C.
- Shown is the formation of fullerene-like defects.

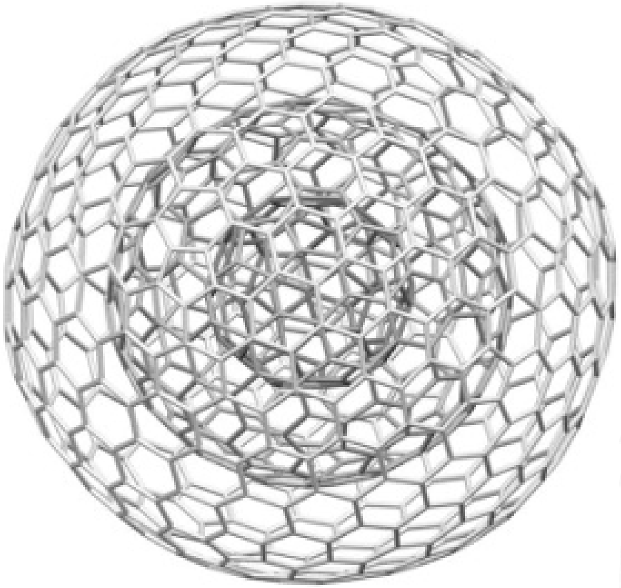



S.Johns, et al., Carbon, 116, 2020

# Carbon Onions in Neutron-irradiated Nuclear Graphite

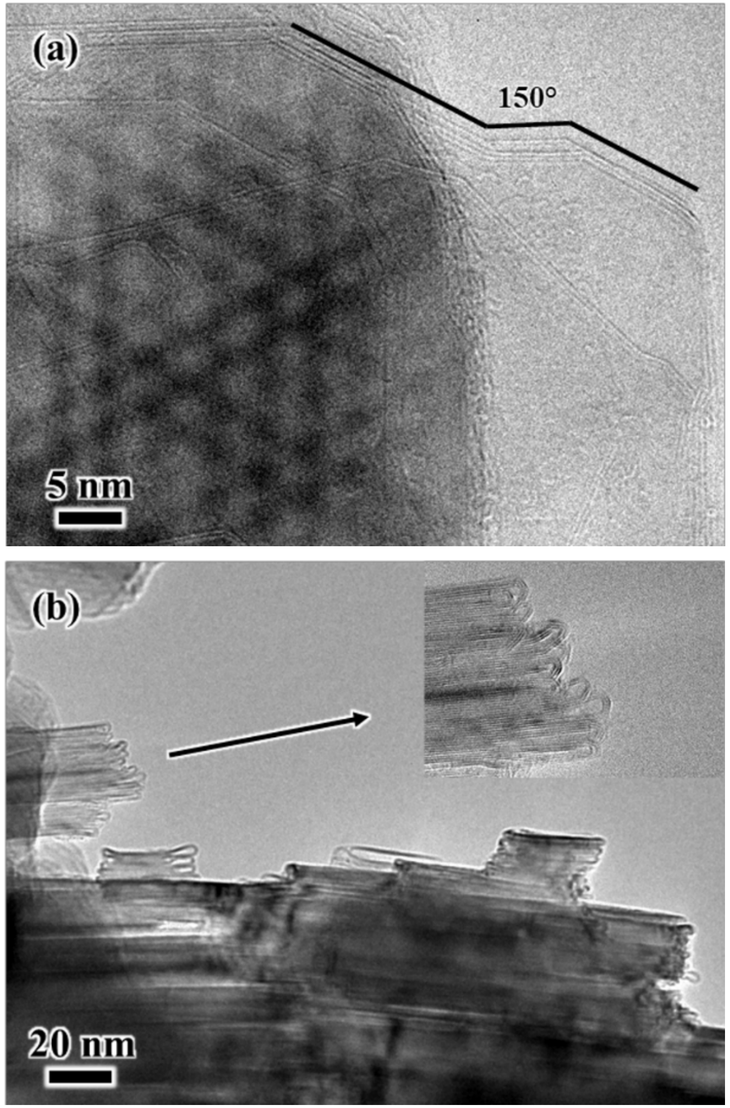


S.Johns, et al., Carbon, 116, 2020

# Effects of High-temperature Thermal Annealing




- Nuclear graphite IG-110 thermally annealed at 2500°C.
- All basal plane edges were observed to rearrange into fullerene-like loops.
- Loops were found to form at 150°.
- Suggesting, closed fullerene-like loops occur on alternating crystallographic directions (i.e., zigzag $\langle 11\bar{2}0 \rangle$ to armchair $\langle 10\bar{1}0 \rangle$).

# Conclusions