Understanding and Improving Graphite for Nuclear Fission

Background

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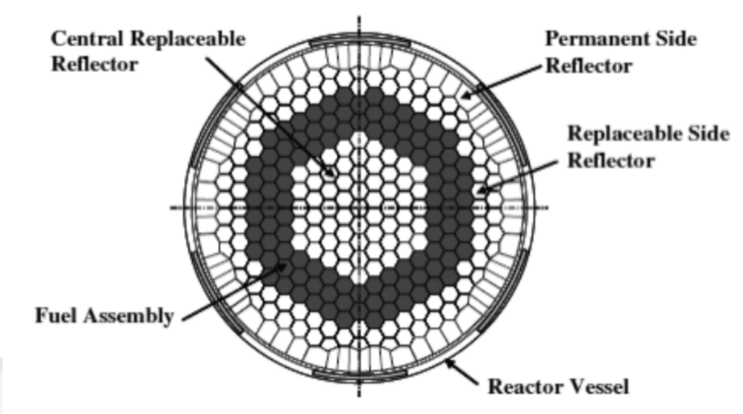
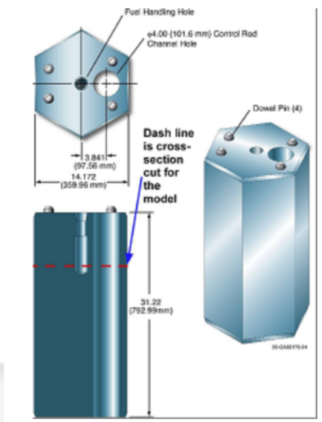
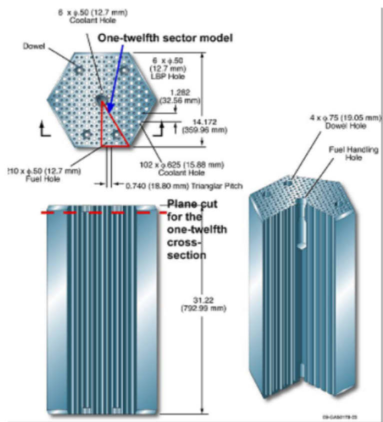
Why graphite?

- A key structural material in advanced nuclear reactors for electricity and process heat generation
- Moderator & reflector to slow down fast neutrons
  - Fast neutron flux reduce exponentially with distance → thermal flux↑
  - Neutron flux and thermal gradient → internal deformations
- Typical components – quite intricate





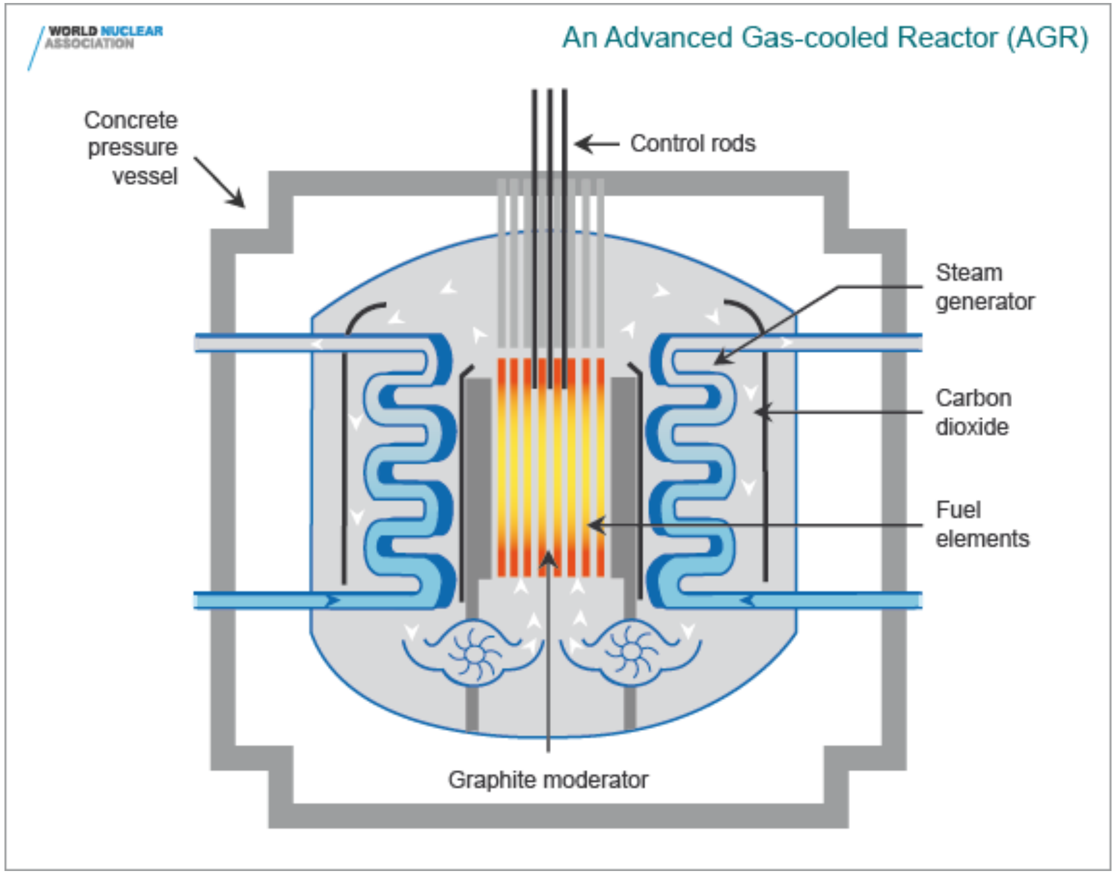


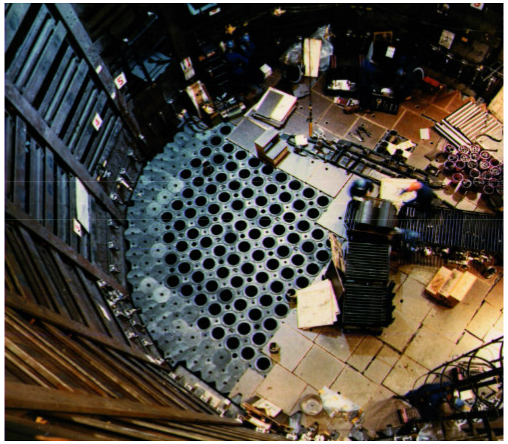
Advanced designs – heading to Gen IV

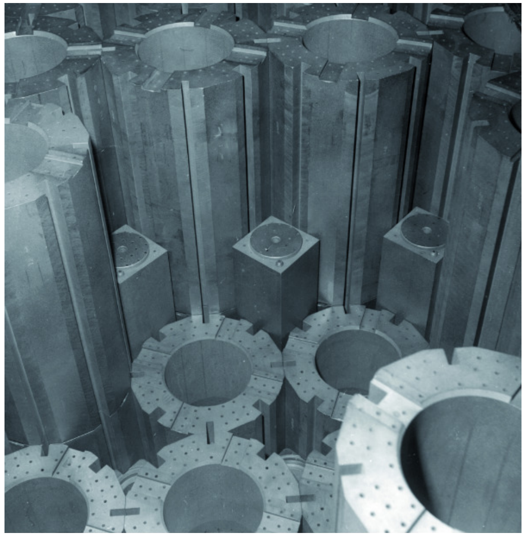
- HTR development: Dragon (OECD), AVR, THTR
- Peach Bottom, Fort St Vrain
- HTR-10, China;
- HTTR, Japan
- PBMR development, S. Africa
- HTR-PM development, China
- Other HTR concepts: France, Russia, USA, NGNP (Areva)
- VHTR

All HTRs use graphite as reflector. Prismatic core design employs graphite for fuel matrix; pebble-bed design employs graphite in fuel ball matrix

Example graphite reactor – AGR







- Operating temp. ~400^{o}C

Example graphite reactor – HTR-10

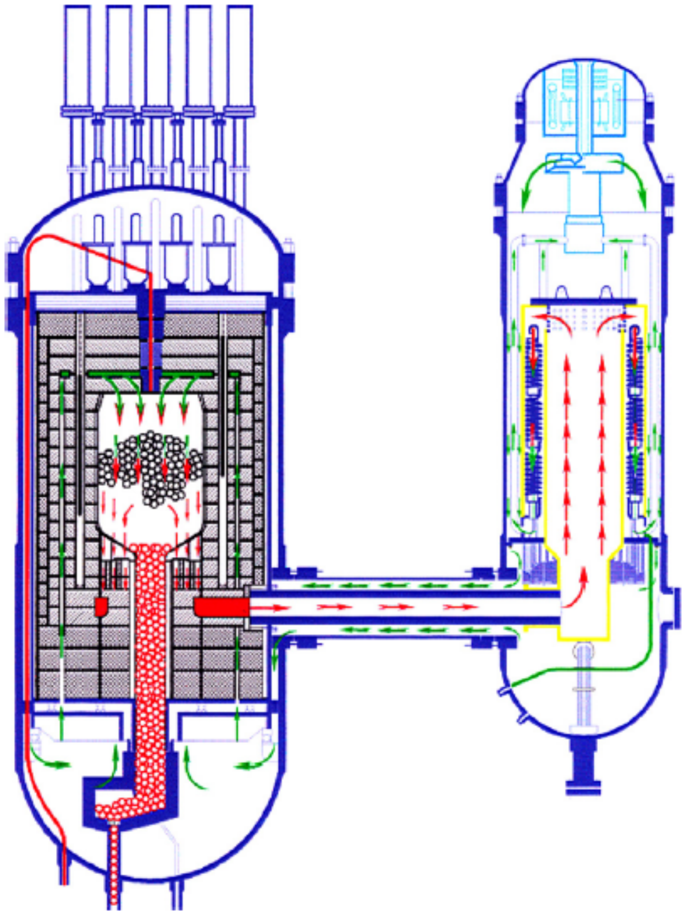
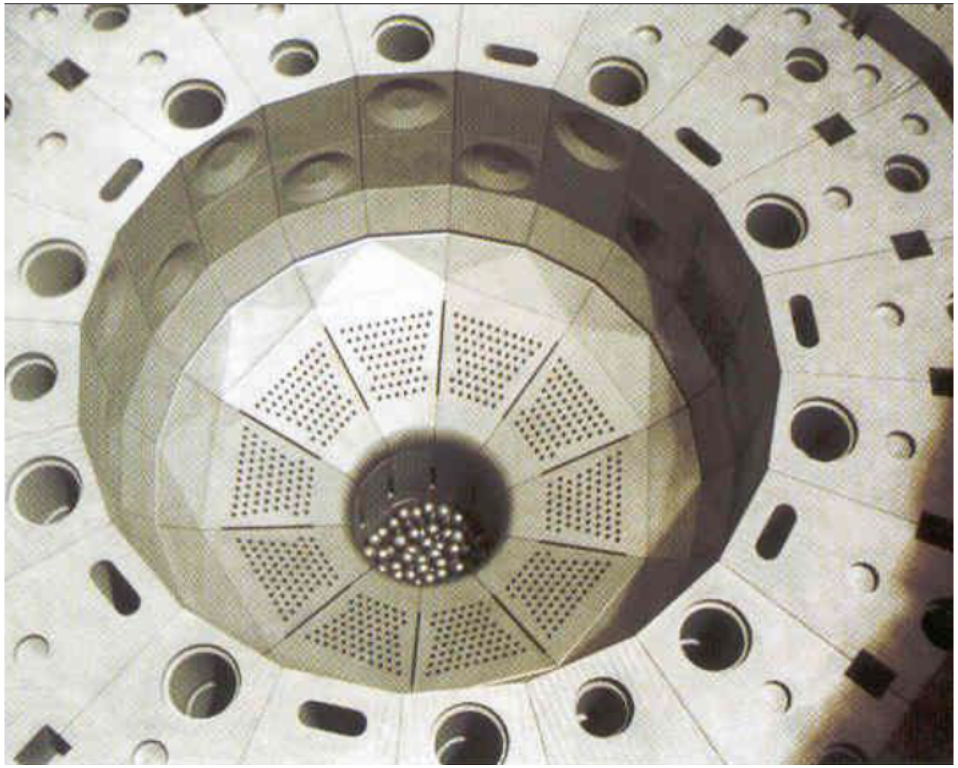
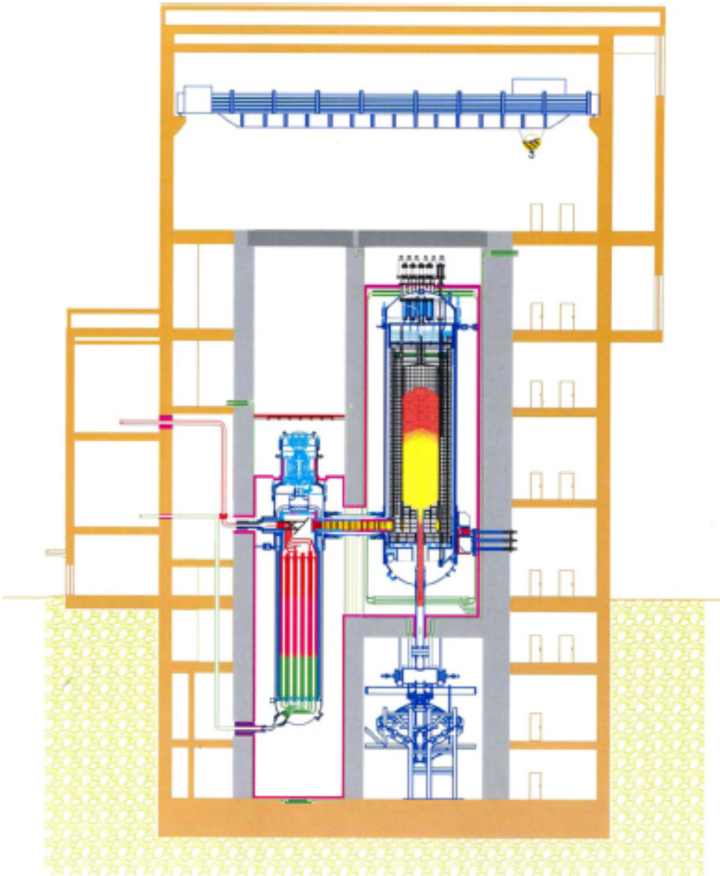


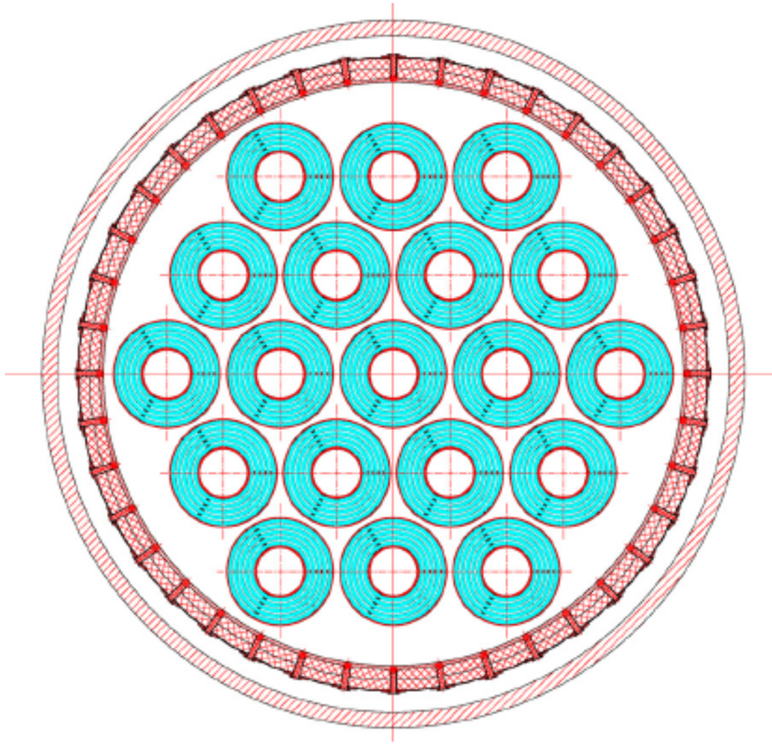
Fig. 1. Chinese 10 MW high-temperature gas-cooled test reactor (HTR-10).



Z. Zhang, etc., Nuclear Eng & Design 239(2009) 1212-19

Example graphite reactor – HTR-PM





Z. Zhang, etc., Nuclear Eng & Design 239(2009) 1212-19

- Operating temp. ~750^{o}C, potentially 900^{o}C

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Harsh environment inside HTGR

- Working environment
  - Pressure of He : 11-70 bar
  - Temperature: Inlet temp. >250^{o}C; outlet temp. >700^{o}C
  - Oxidants: leaking water and air
  - Radiation: neutrons
- Material property change induced by irradiation
  - Internal shrink stress
  - Thermal stress
  - Irradiation-induced swelling:
    - Reduction of modulus, strength, etc.

Why iso-statically moulded graphite

- High strength
- High reliability
- Good thermal conductivity
- Large components possible
- Delicate features possible
- Tend to be more brittle
  - Low crack propagation resistance
  - High sensitivity on flaw
  - Fatigue could be concerned

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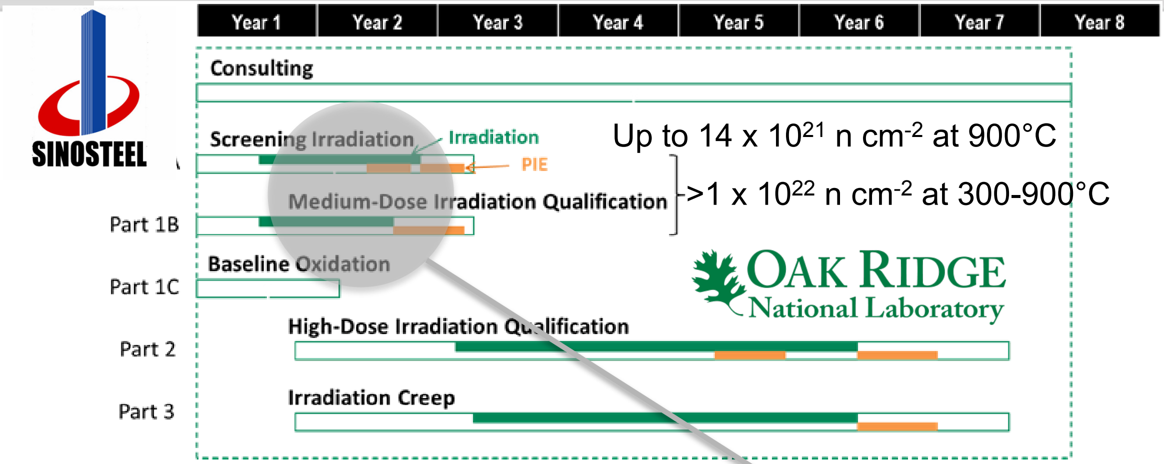
Understanding and Improving Graphite for Nuclear Fission

Loughborough: Houzheng Wu, Roger Smith

Bristol: Peter Flewitt, Keith Hallam

Oxford: James Marrow

Opportunity and Methodology



Sinosteel/ORNL Irradiation Programme (Year 1 = 2015)

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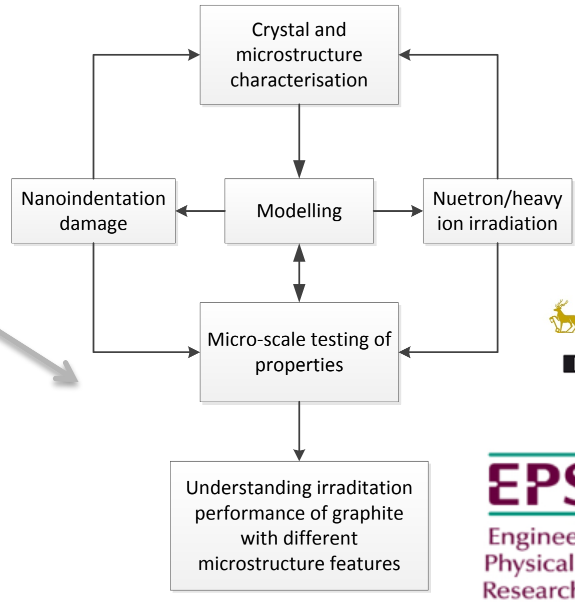
SINOSTEEL
- Producer of graphite for the Chinese HTR-PM programme,
- Funding the irradiations at ORNL to select graphites



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GSI

EPSRC Engineering and Physical Sciences Research Council

Eight Graphites (different Filler and Binder)
- Characterisation over length scales:
  - Dimensional change, Young's modulus, thermal expansion, tensile strength, toughness
  - HRTEM, nano-indentation, Raman, EELS
- Modelling at the meso-scale
  - MD atomistic simulations (up to ~50 nm^3) to predict physical and mechanical properties
- Testing at the meso-scale
  - In situ nano-indentation, pillar compression and Nano-XCT
  - In situ TEM testing with DIC and diffraction strain measurement

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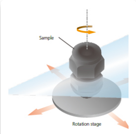
Samples and methods

- 8 grades of nuclear graphite
- Isostatically pressure-moulded
- Different in coke size, amount of binder, sources of raw materials, impregnation/graphitisation conditions, density, porosity

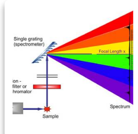


X-Ray Tomography

- Micro-porosity
- Grain sizes

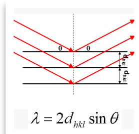
(S)TEM

- Microstructural features
- Lattice defects
- Distribution of filler particles
- Porosity
- Chemical bonding



Raman spectroscopy

- Defect information
- Disordering (basal planes)
- Filler/Binder
- Polarising



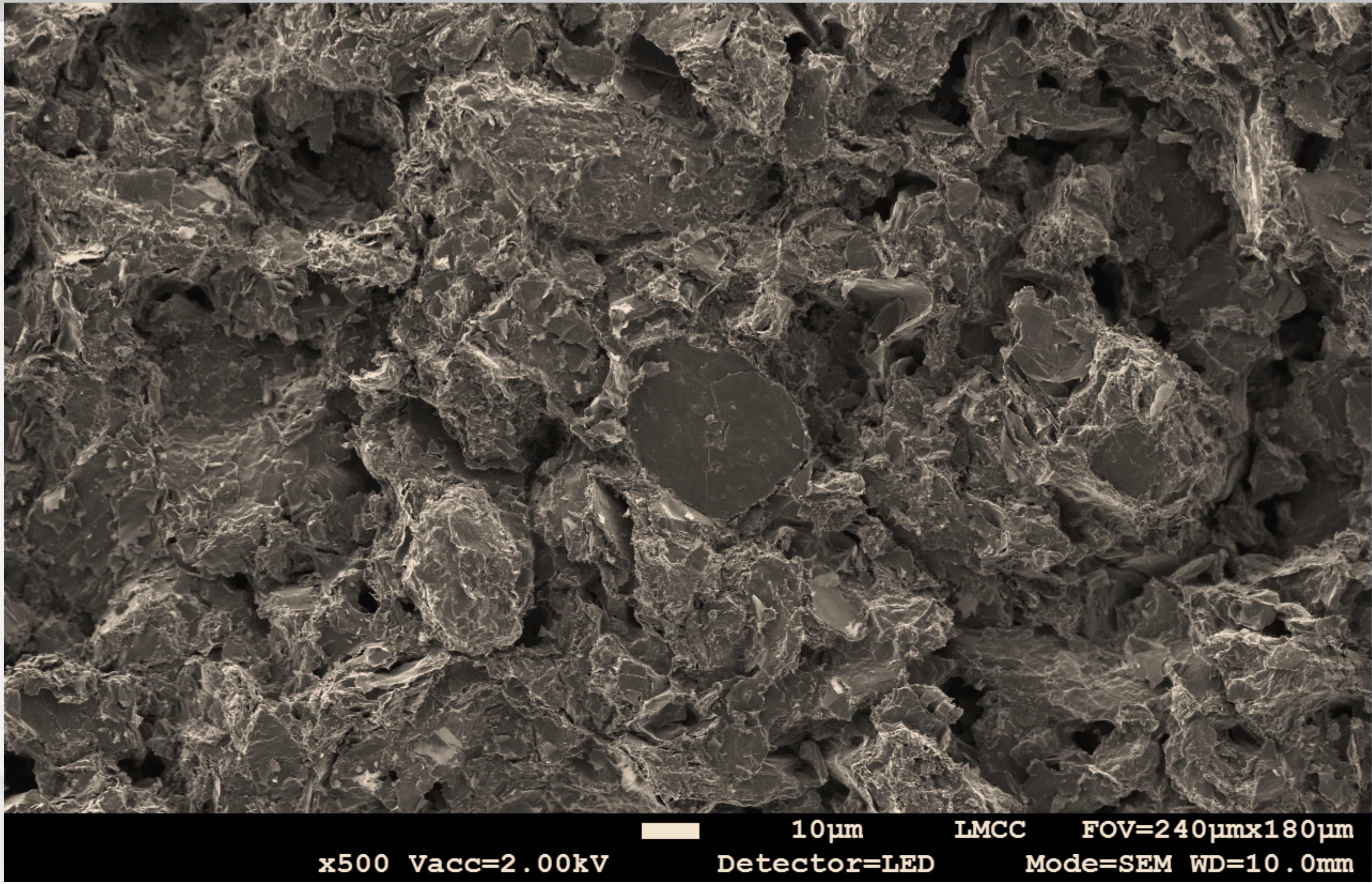
X-Ray Diffraction

- Grain sizes
- Lattice parameters
- High-temp. behaviour

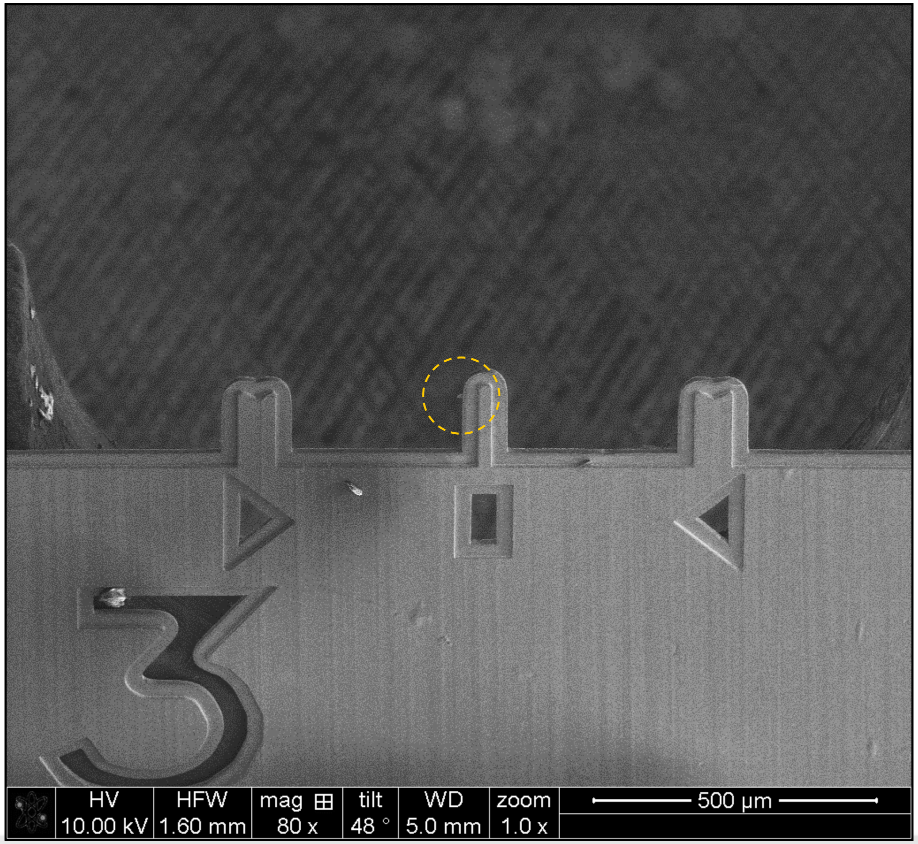
Progress

- RA on microstructure characterisation at L'boro started in March 2016
- RA on microscale testing at Bristol/Oxford started in August 2016
- RA on modelling at L'boro started in September 2016
- Heavy ion irradiate graphite done by GSI
- All virgin graphite samples are ready
- 1st batch of neutron irradiated graphite at 900^{o}C is available now, and 2nd batch in April 2017.

Fracture surface of virgin iso-graphite



FIB Sample preparation workflow



1. Deposition of protective layer
2. Trench production and cross-section cleaning
3. Tilt and cut off lamella
4. Transfer to TEM grid

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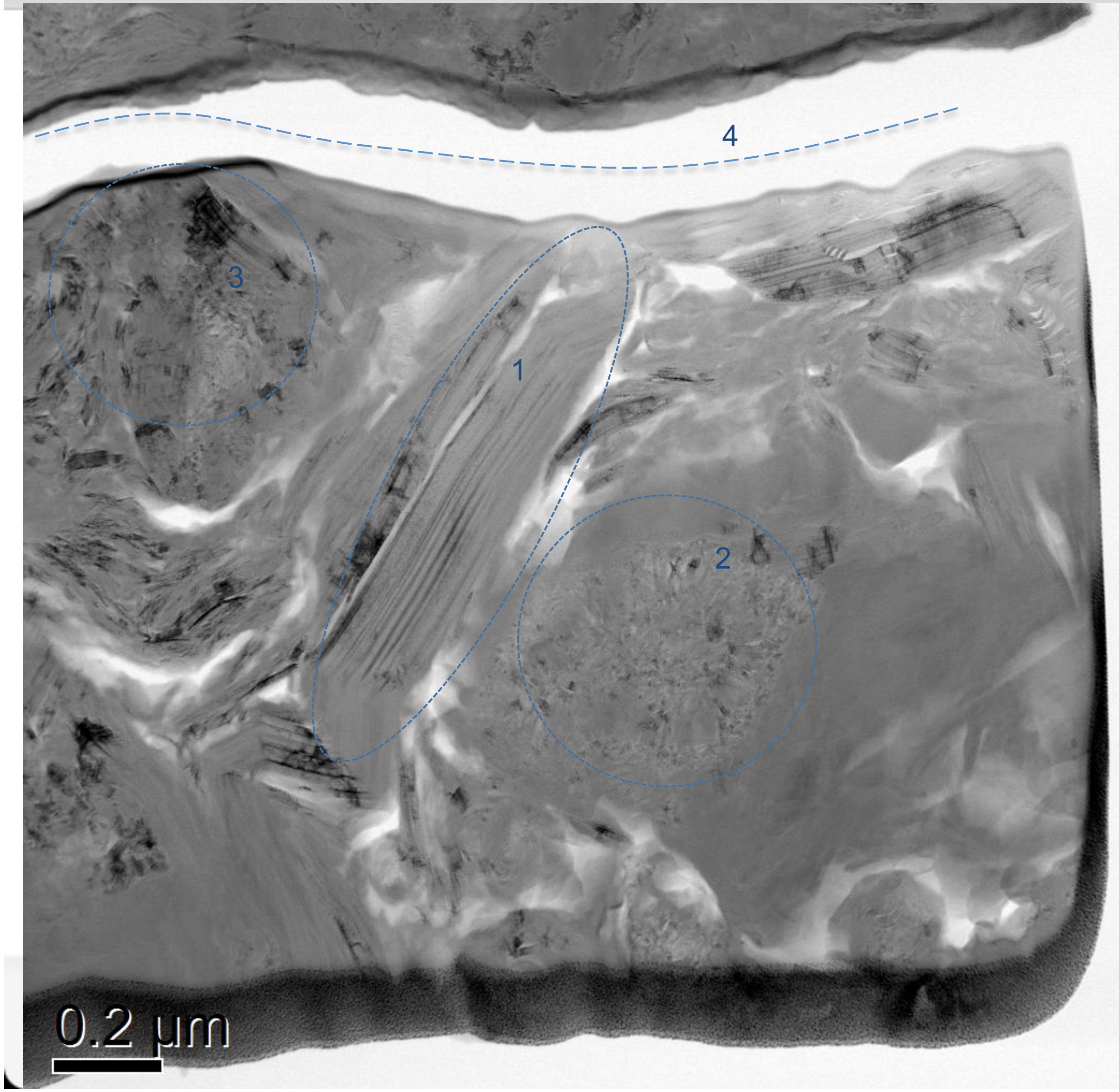
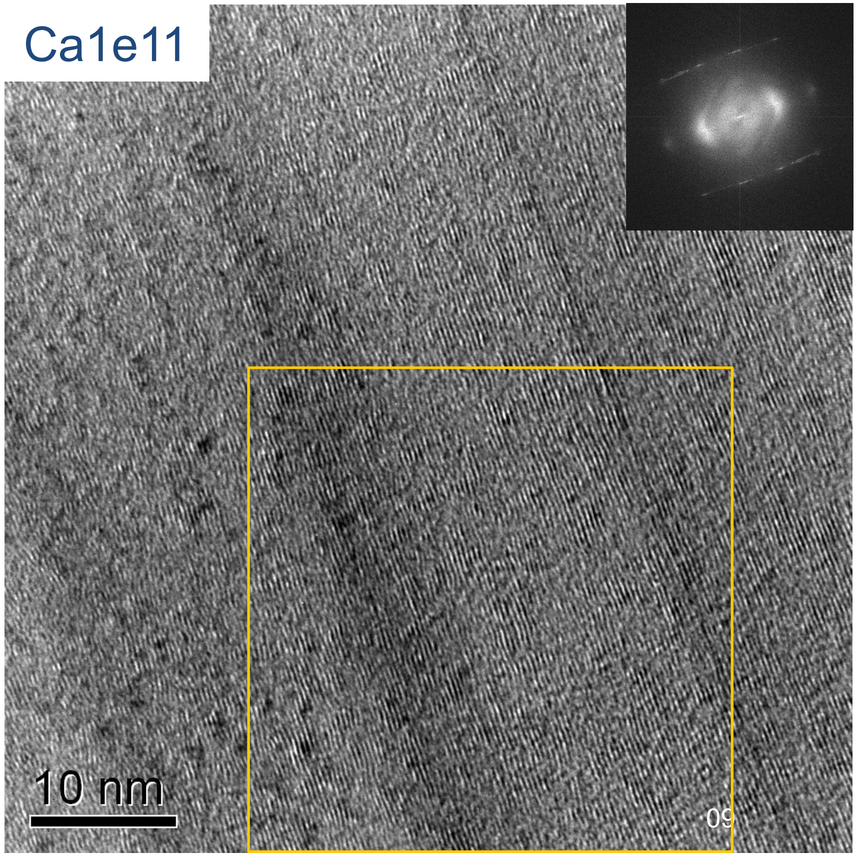
Microstructure of binder

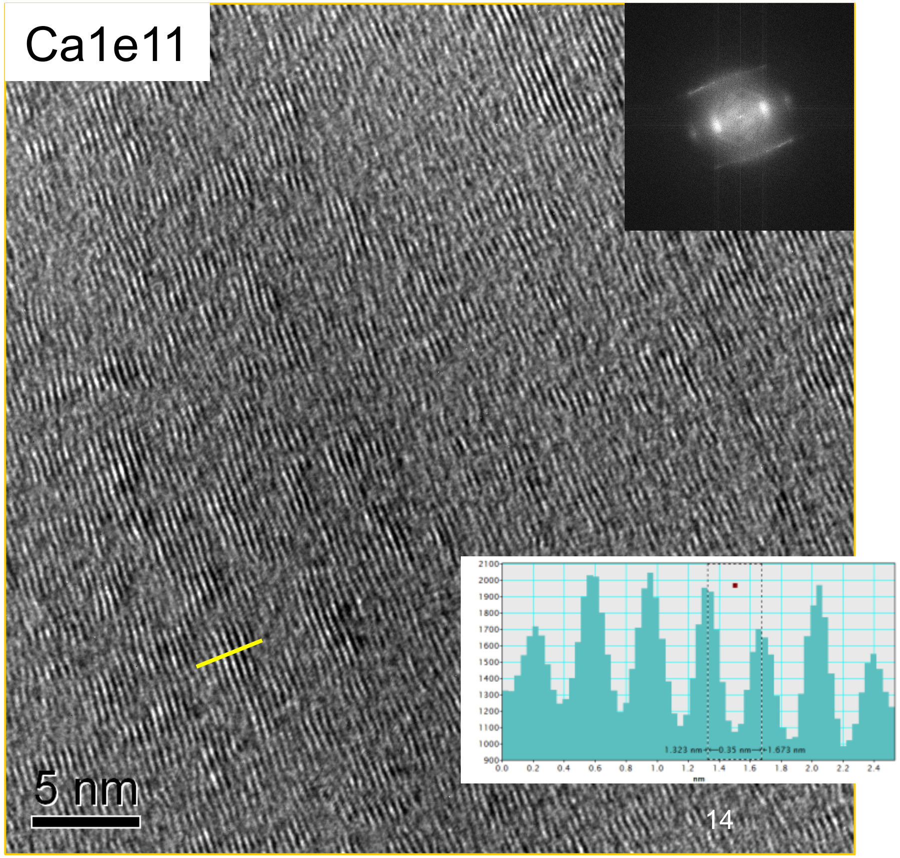


1. Graphitised area/ particle shows Mrozowski cracking
2. Chaotic structure
3. Quinoline insoluble
4. Crack

Virgin sample #2, STEM-BF

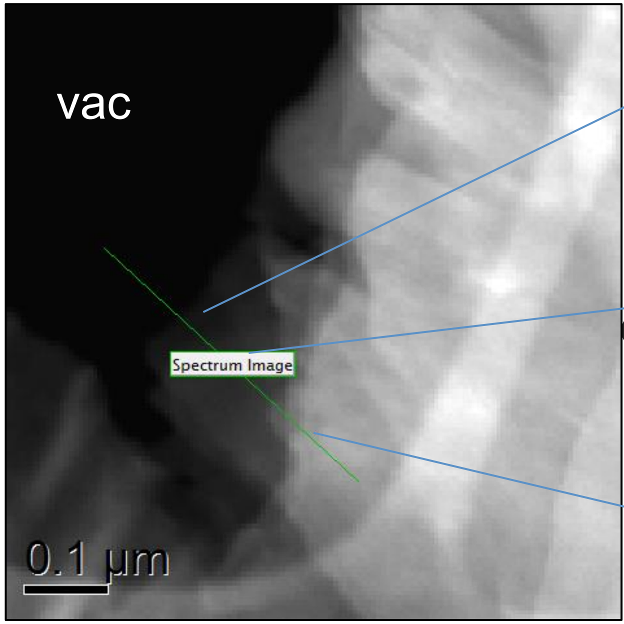
Ca ion irradiated graphite





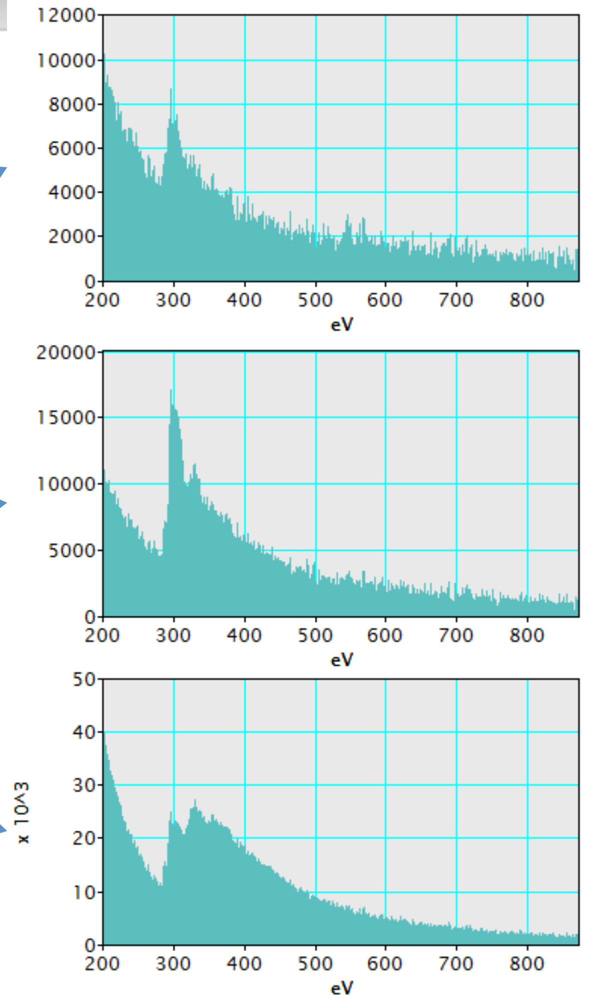
Ca ion irradiated graphite

• ctroscopy



SNG623C1e11

- species
- concentration
- el. structure



core-loss spectra extracted from line scan

Carbon K-edge

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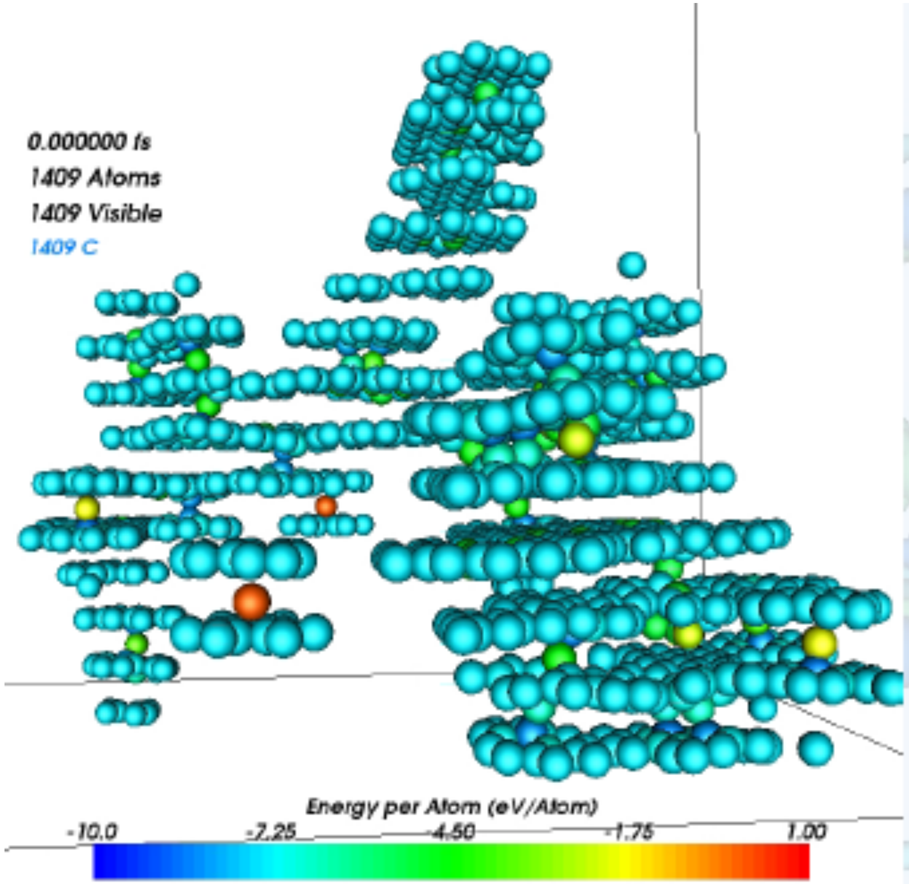
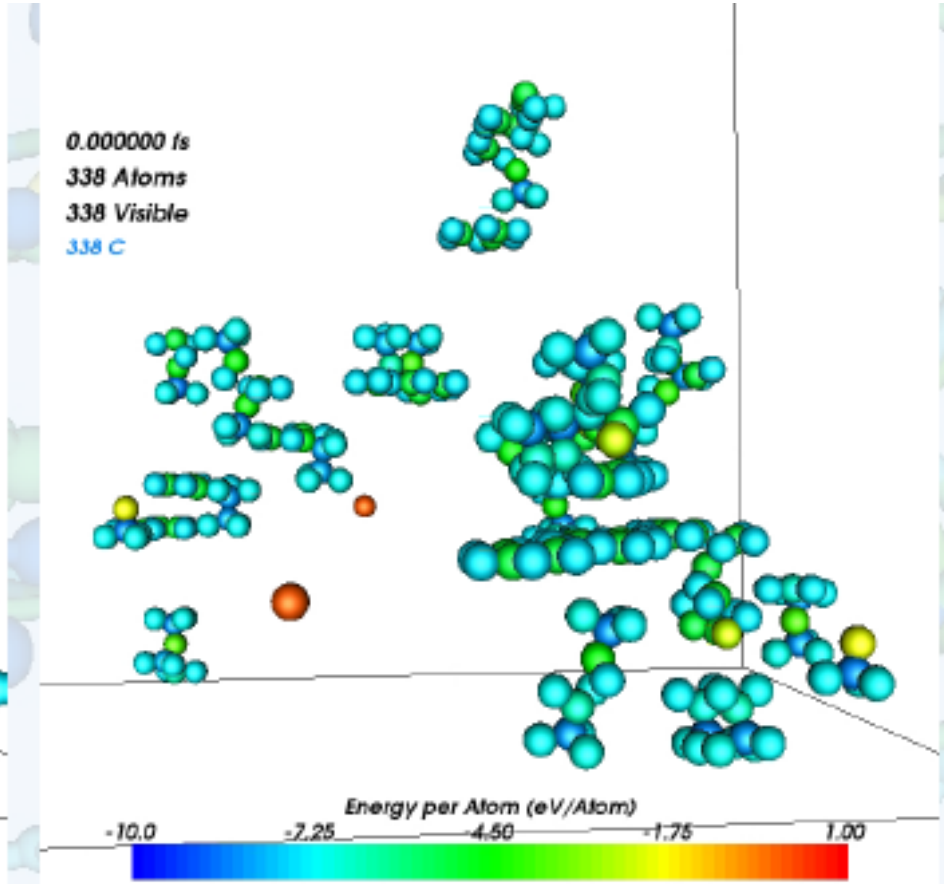
Molecular dynamic modelling the neutron knock on effect in graphite



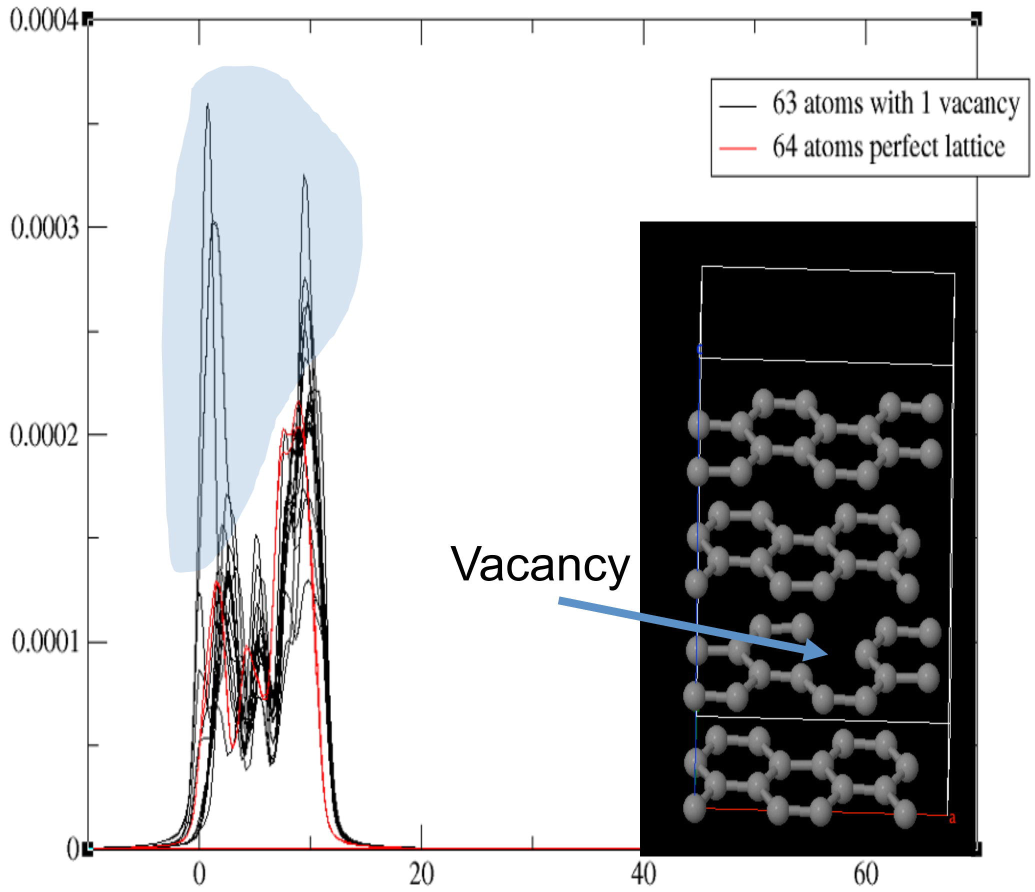


EELS simulations on vacancies & interstitials

- Goal:
  - Understand how many and what kind of defects in a graphite
  - Provide scientific base for de-convoluting EELS spectrum

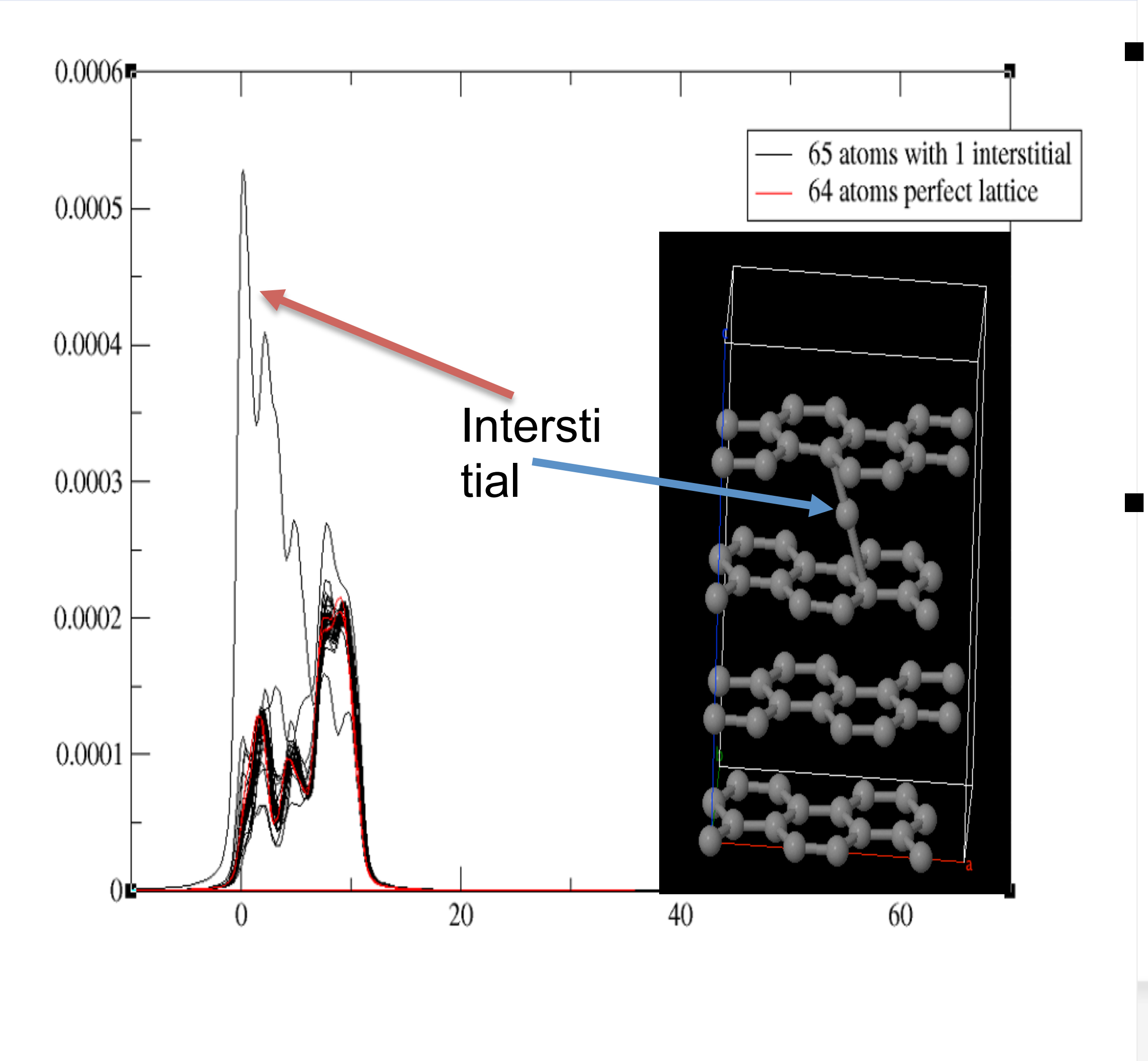
- Method:
  - Use DFT to simulate EELS spectrum on graphite systems
    - DFT code: CASTEP (http://www.castep.org/)
  - Provide coordinates of a graphite system
  - Yield EELS spectrum for each atom in the system

EELS spectrum with 1 vacancy



- Little effect on atoms which are far away from the vacancy.
- Spectrums for atoms close to the vacancy (shaded) are quite different from others.

EELS spectrum with 1 interstitial (1)

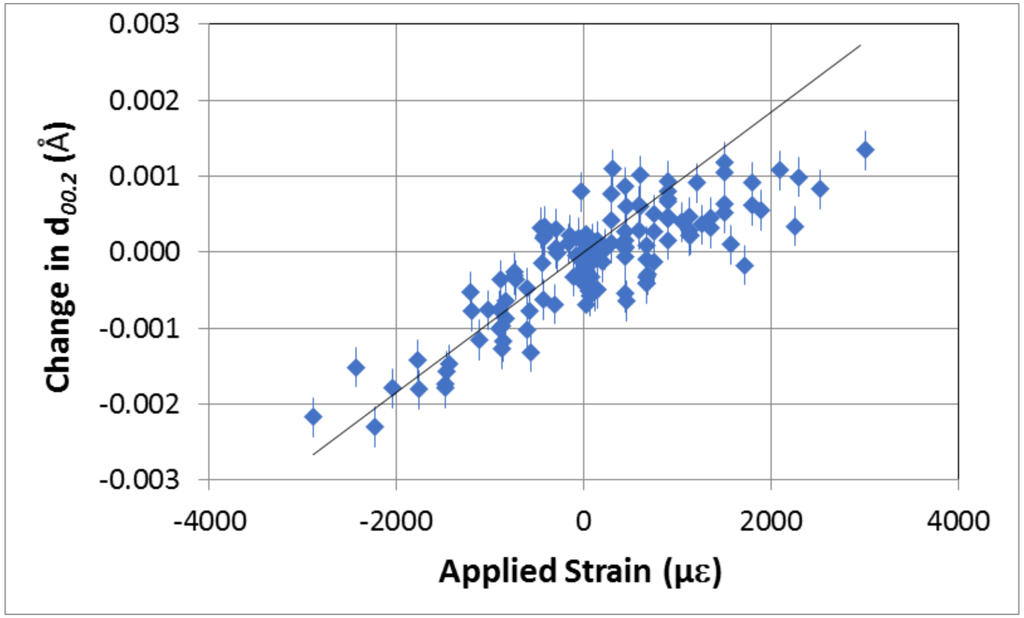


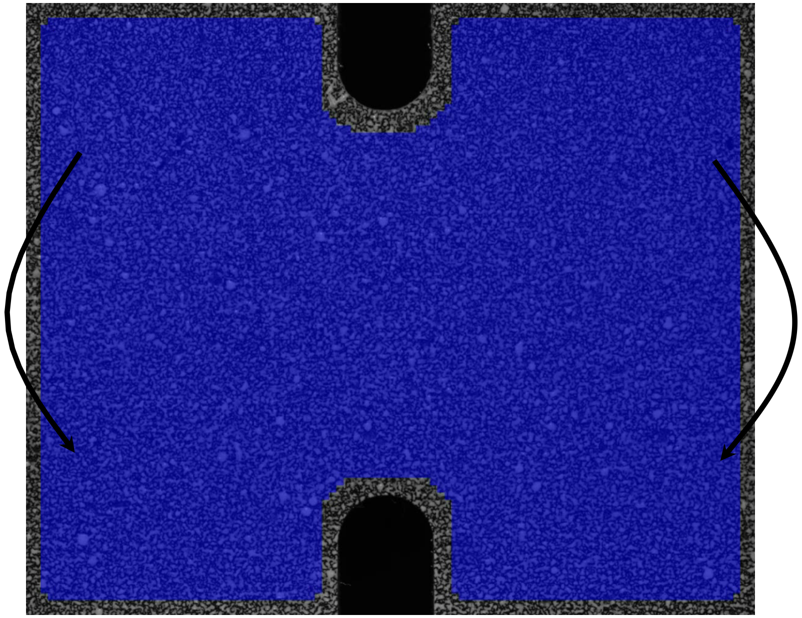
- There are many kinds of interstitials. In some cases, the interstitials have little effect on other atoms.

- The spectrum for some interstitial atoms is well separated from others.

Multi-scale deformation & fracture (by Liu etc.)

- Nano-scale (*in situ* neutron diffraction)

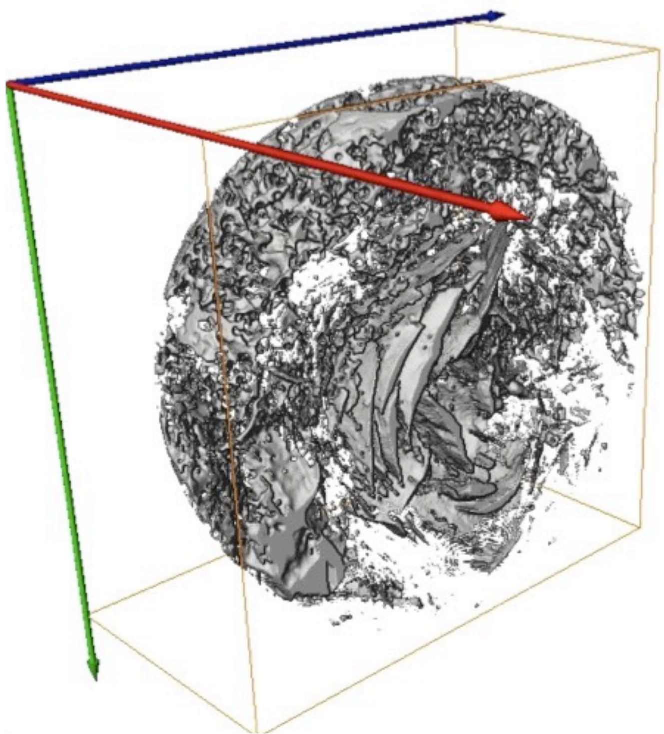




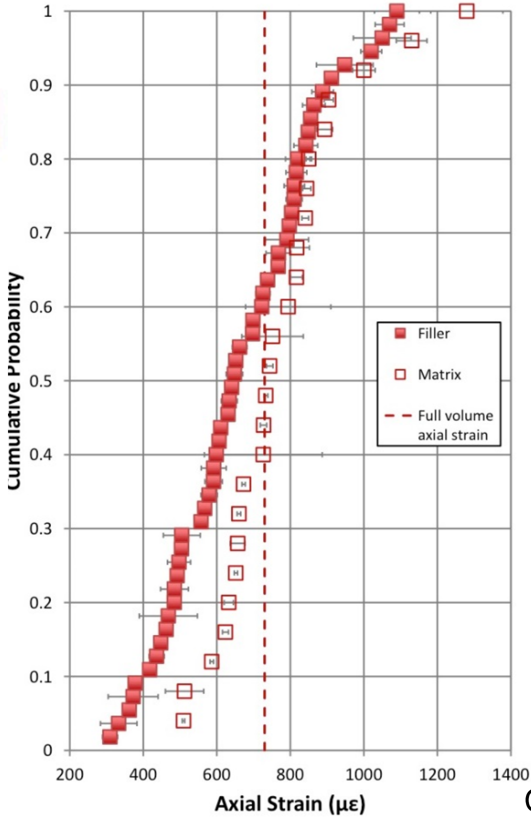
- Lattice strain is an order of magnitude less than bulk strain, i.e. the microstructure accommodates the local strain
- Non-linear relation at higher tensile strain

Multi-scale deformation & fracture (by Liu etc.)

- Nano-scale (*in situ* X-ray and neutron diffraction)



Section of spherical particle ~ 1 mm dia.



Different part of the microstructure accommodates different amount of strain, i.e. compliance variance across the loaded volume

Non-uniform strain distribution

- Local strain concentration
- Local damage initiation

Courtesy of Dr. Vertyagina (Oxford University)

Multi-scale deformation & fracture (by Liu etc.)

- Micro-scale (*in situ* micro-cantilever testing)
  - System calibrated using Si;
  - Larger deformation than macro-scale
  - Progressive fracture
  - Unload leads to permanent deformation