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Nuclear Graphite and Graphite Matrix for HGTR's

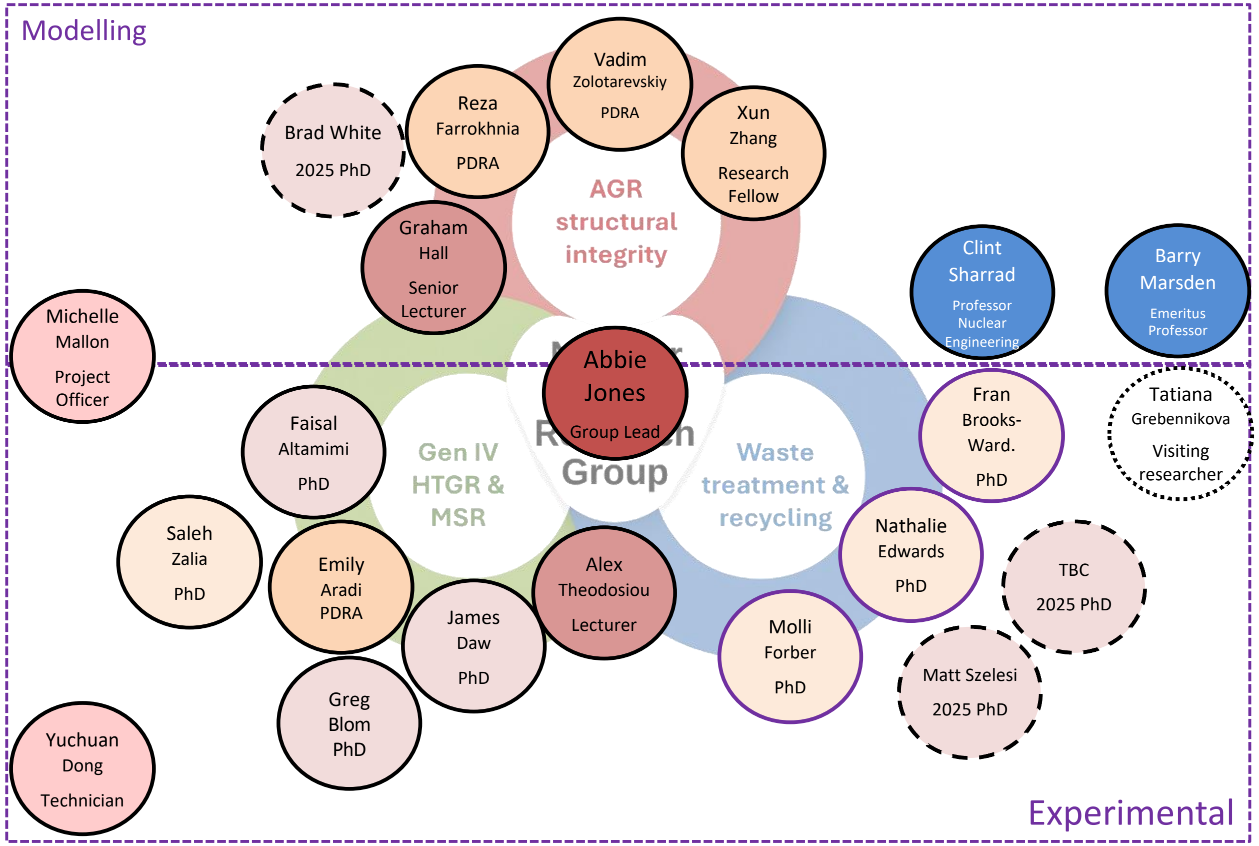
Prof. Abbie Jones

Nuclear Graphite Research Group
The University of Manchester, UK

Nuclear Graphite Research Group

- NGRG founded in September 2001, we are an internationally recognised nuclear research group based at the University of Manchester
- Major Partnerships >£30M in funding and collaborations:
  - Nuclear Regulator (ONR)
  - National and International Facilities
  - Nuclear Industry
- Dissemination and Impact
  - Over 160 open literature publications
  - Over 300 industrial reports

Nuclear Graphite Research Group



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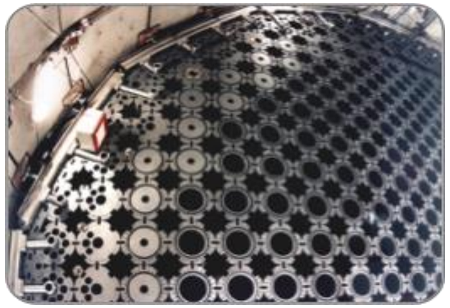
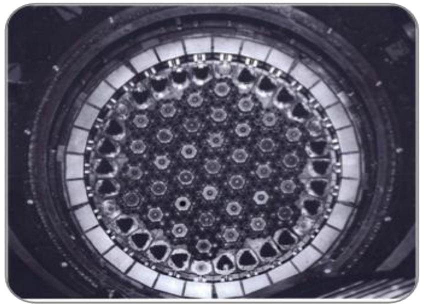
The challenges of nuclear graphite…





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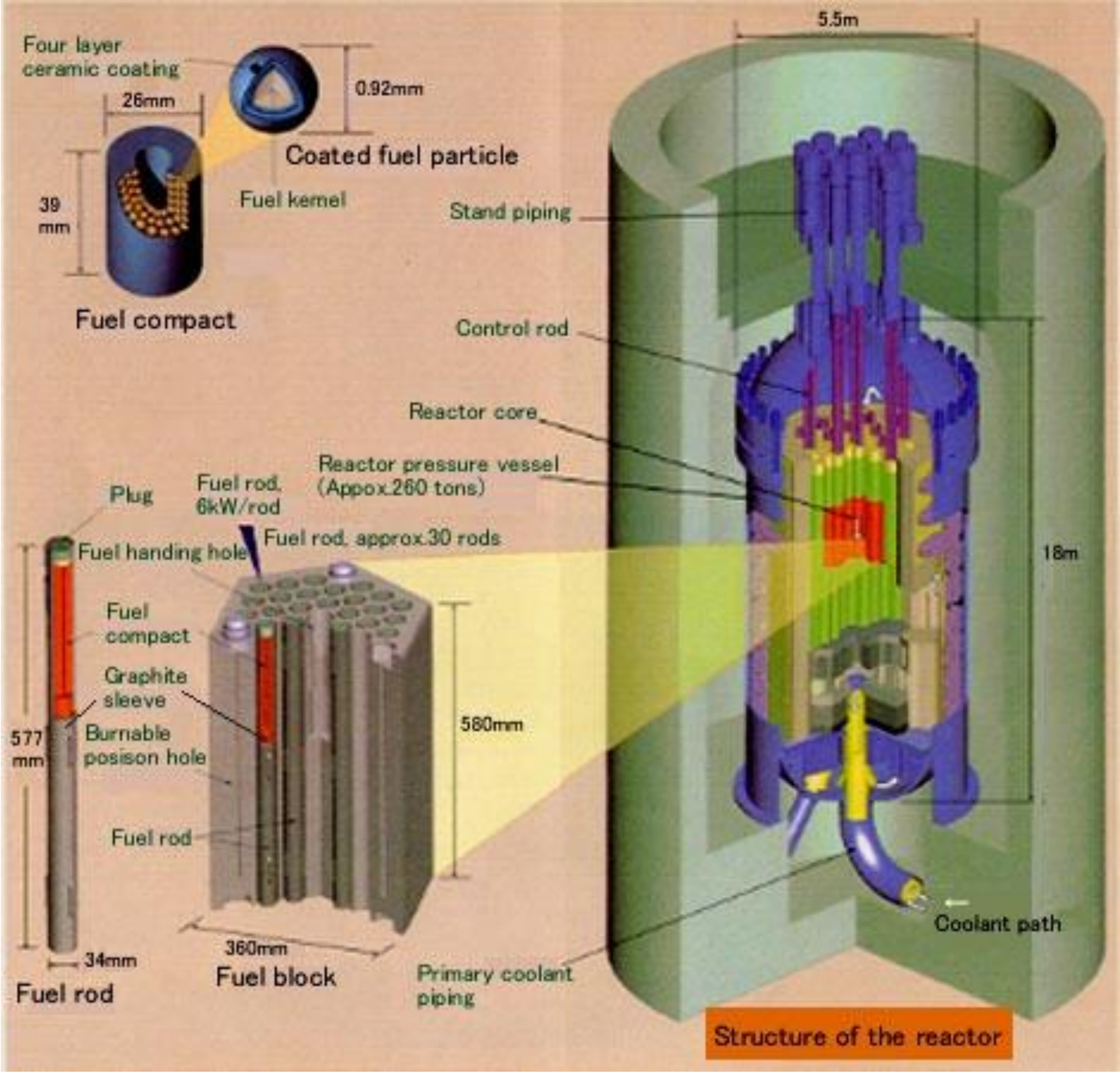
HTGR NUCLEAR GRAPHITE -MODERATOR / REFLECTOR MATERIAL - REQUIREMENTS

HTGR GRAPHITE USE



Nuclear graphite

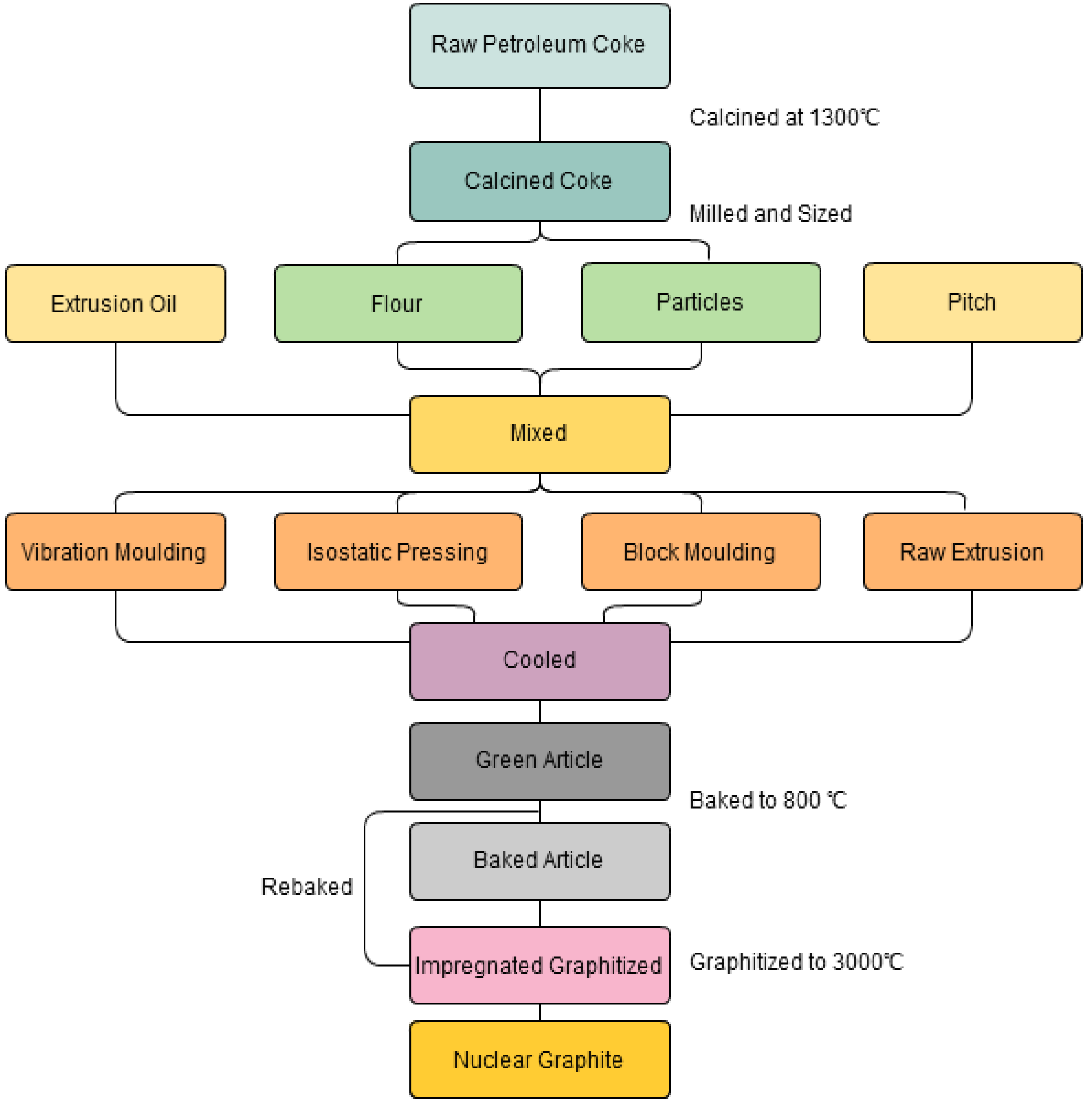
- Nuclear graphite is an artificially produced product
- Essentially a purified version of electrodes used in the steel industry or more refined products used in the electronic and other industries
- Graphite is based on a coke and a binder, which is produced as a bi-product of oil or pitch tar refining
- Non-irradiated (virgin) nuclear graphite is a polygranular crystalline material that has an initial density in the range 1.6 - 1.8 g cm^{-3}.
- The type & size of coke used and the manufacturing route largely determines the virgin graphite material physical properties
- Graphite is batch-produced in large quantities involving very high temperatures

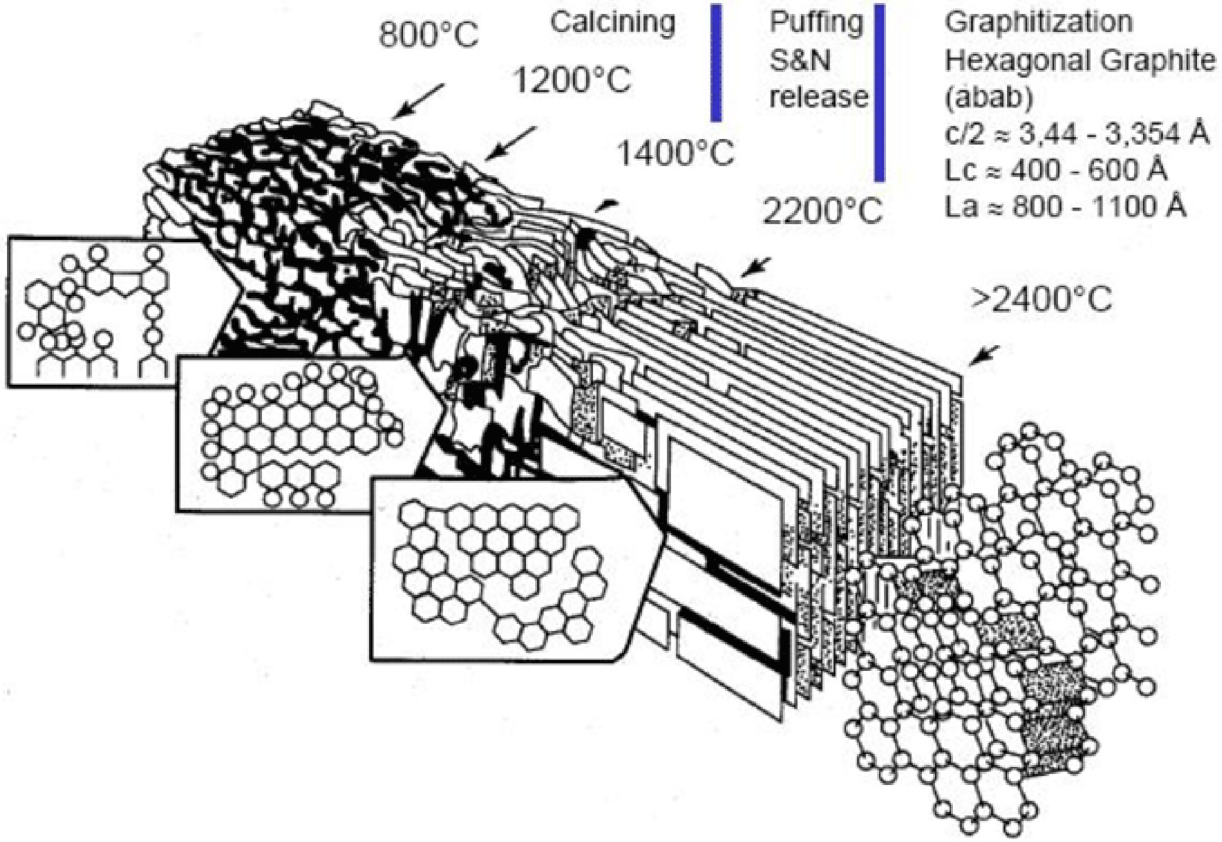
Nuclear Graphite Requirements

Nuclear Graphite designer requires:

- Near-isotropic
  - (1.1) Defined by Coefficient of Thermal expansion (CTE) in orthogonal directions
- High density
- Optimum material properties
- High thermal conductivity
- High purity (neutronic and waste point of view)
- Dimensional stability under irradiation, associated with high CTE ~4 x 10^{-6} K^{-1} (20 – 120 °C)

The Production of Nuclear Graphite





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BAKING PROCESS

[Ref 5]

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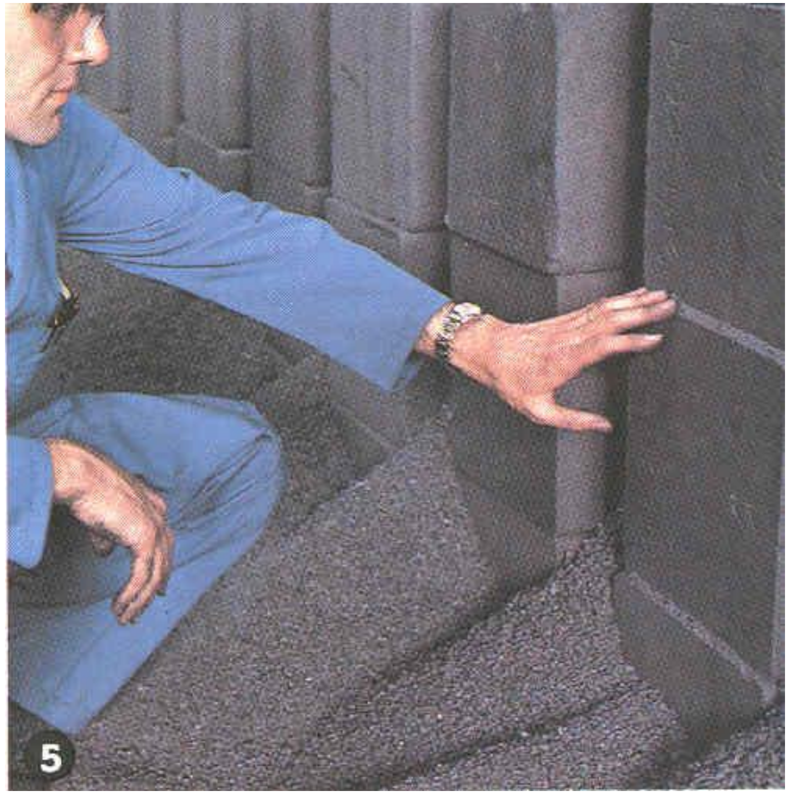
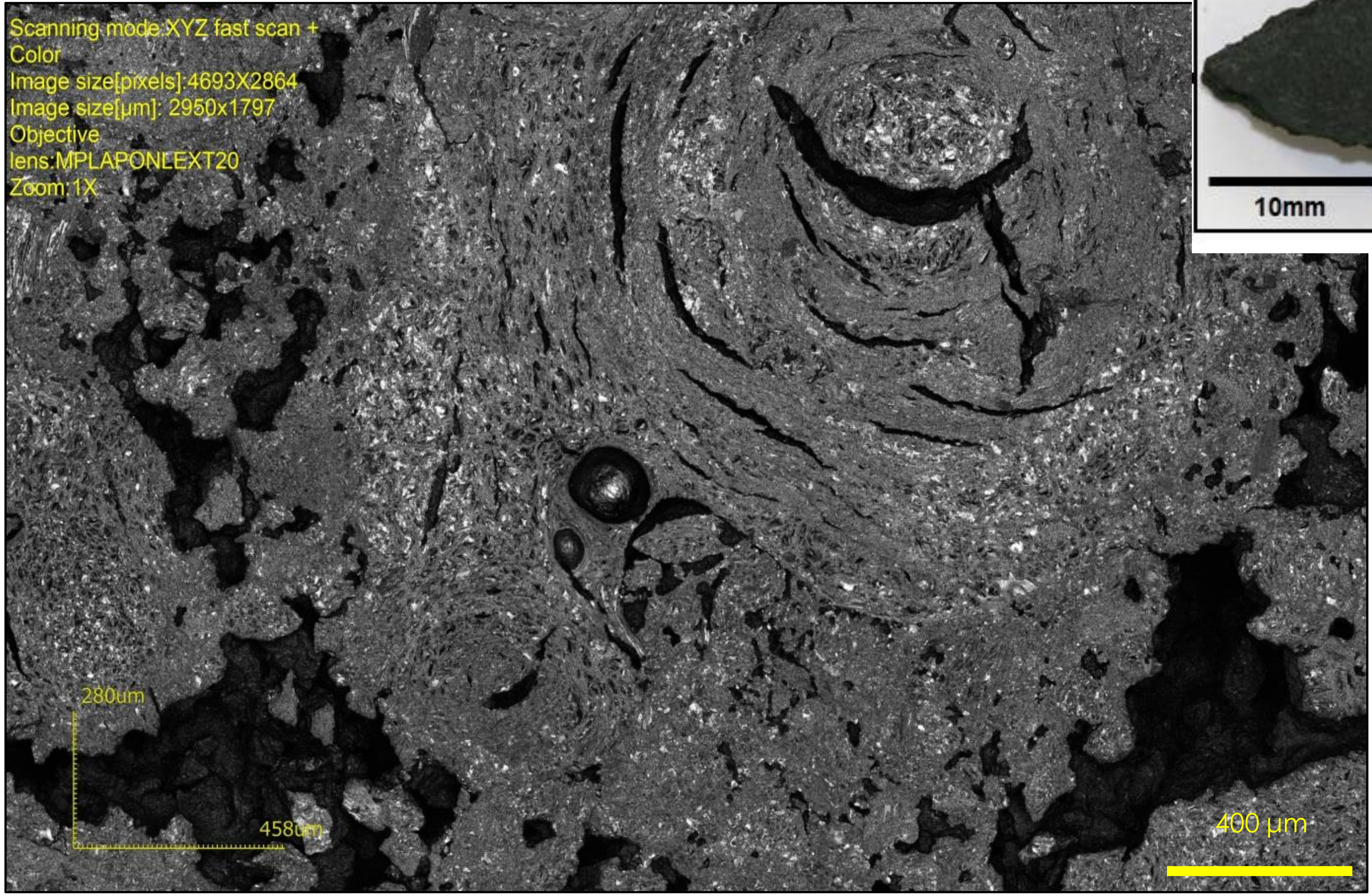
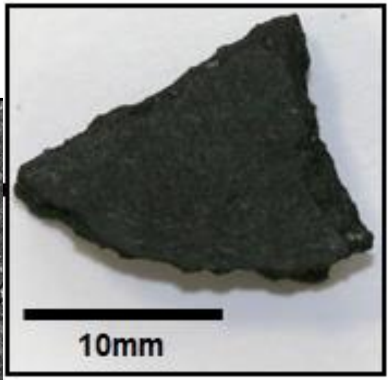


- Green Article baked in a Furnace
- Packed with sacrificial coke to reduce oxidation
- ~800°C, ~4 weeks

Baked carbon





Laser confocal image of baked carbon [3]

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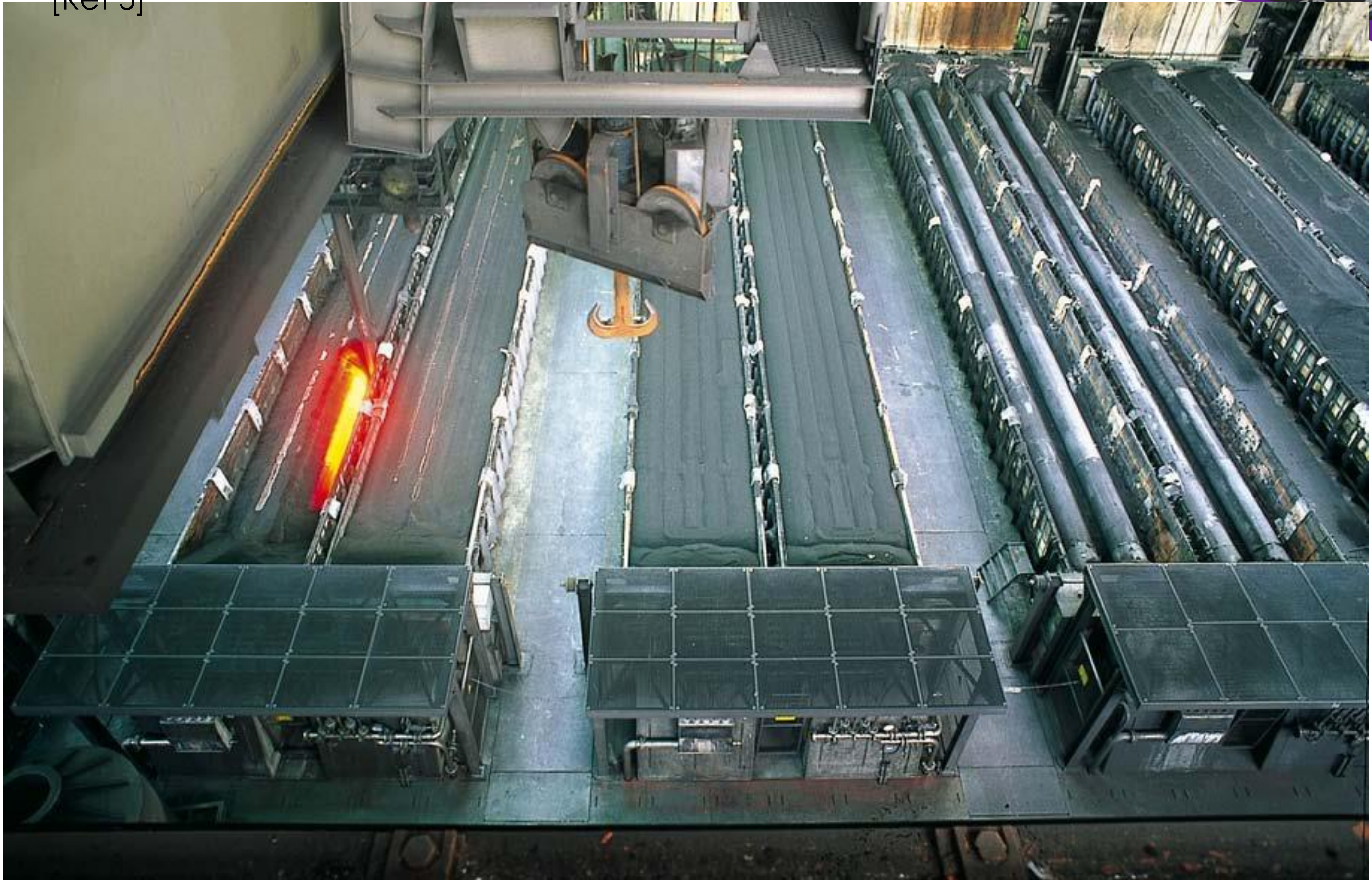
GRAPHITISATION

[Ref 5]



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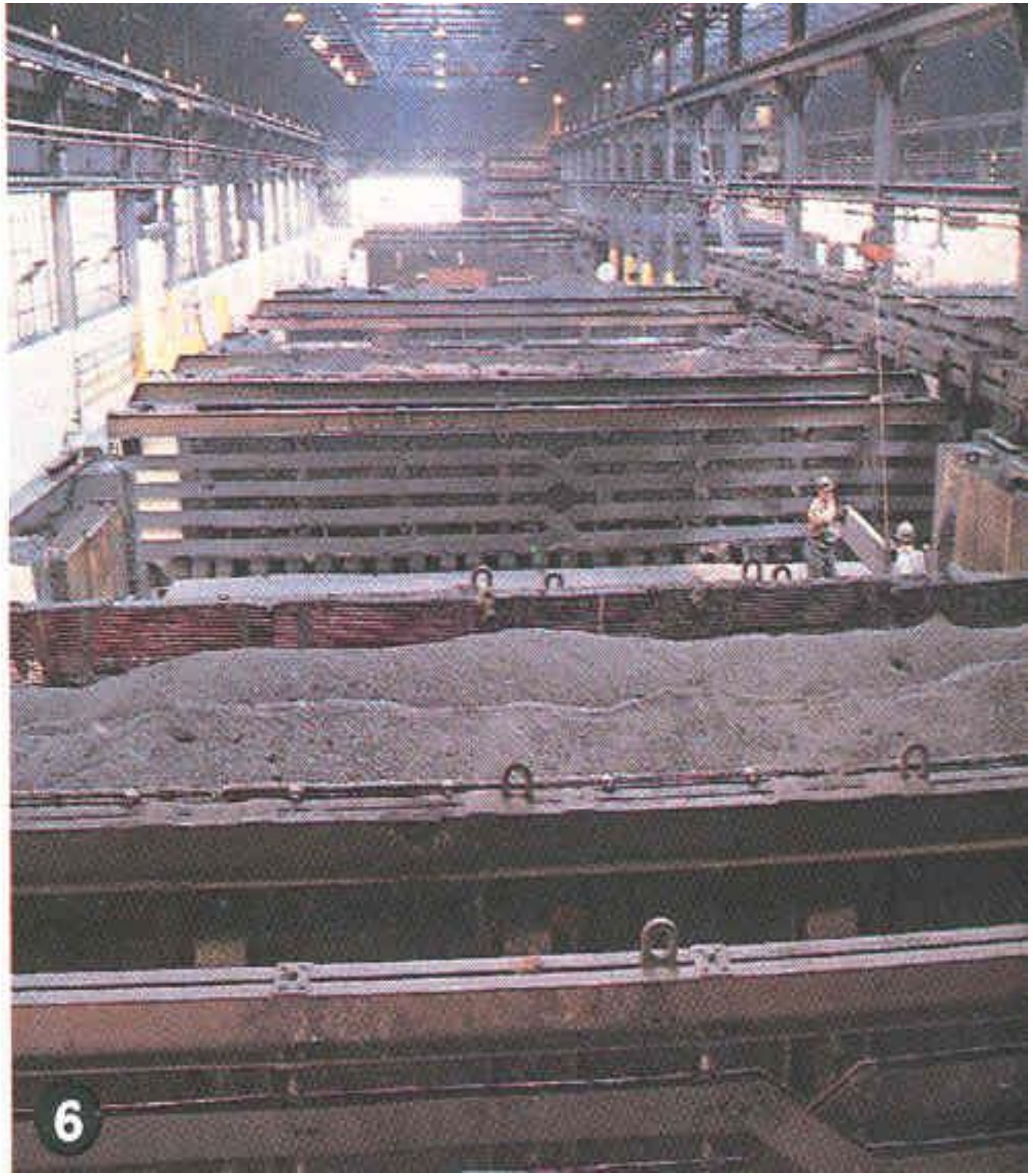

- Acheson Type Graphitisation Furnaces
- ~2800°C,
- ~4 weeks

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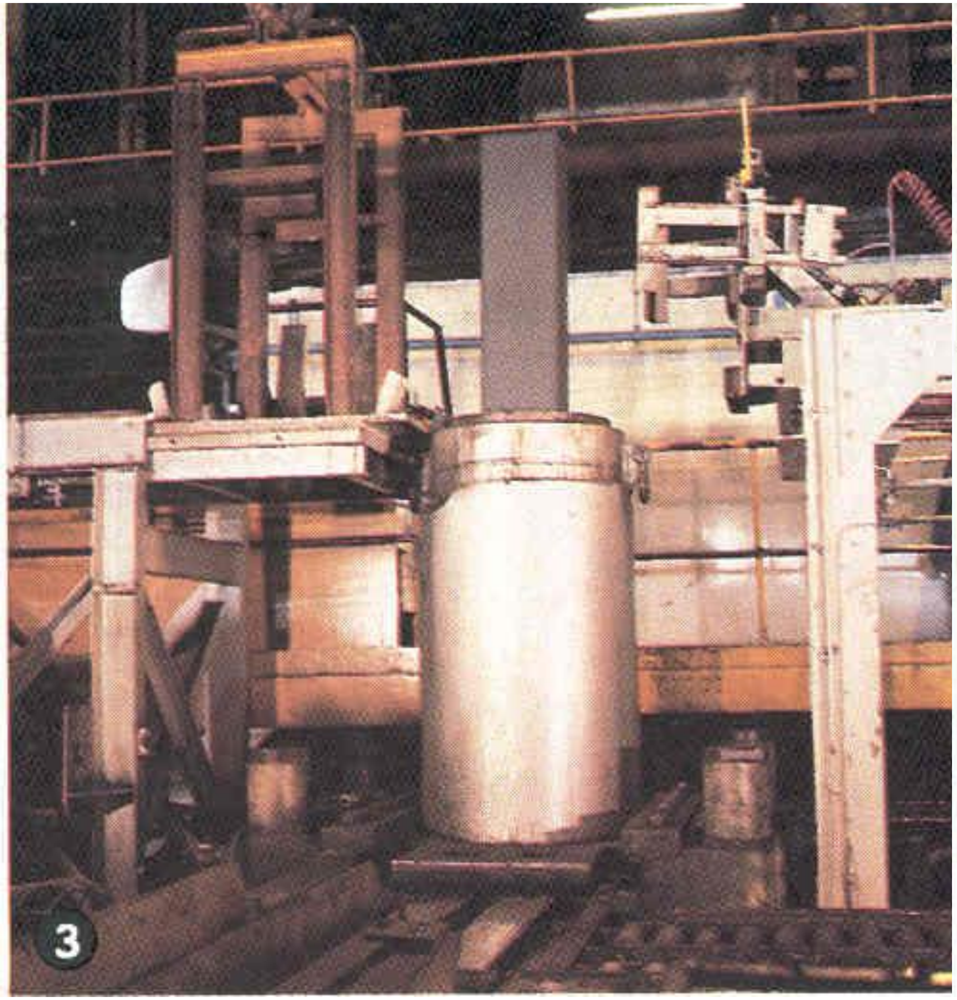
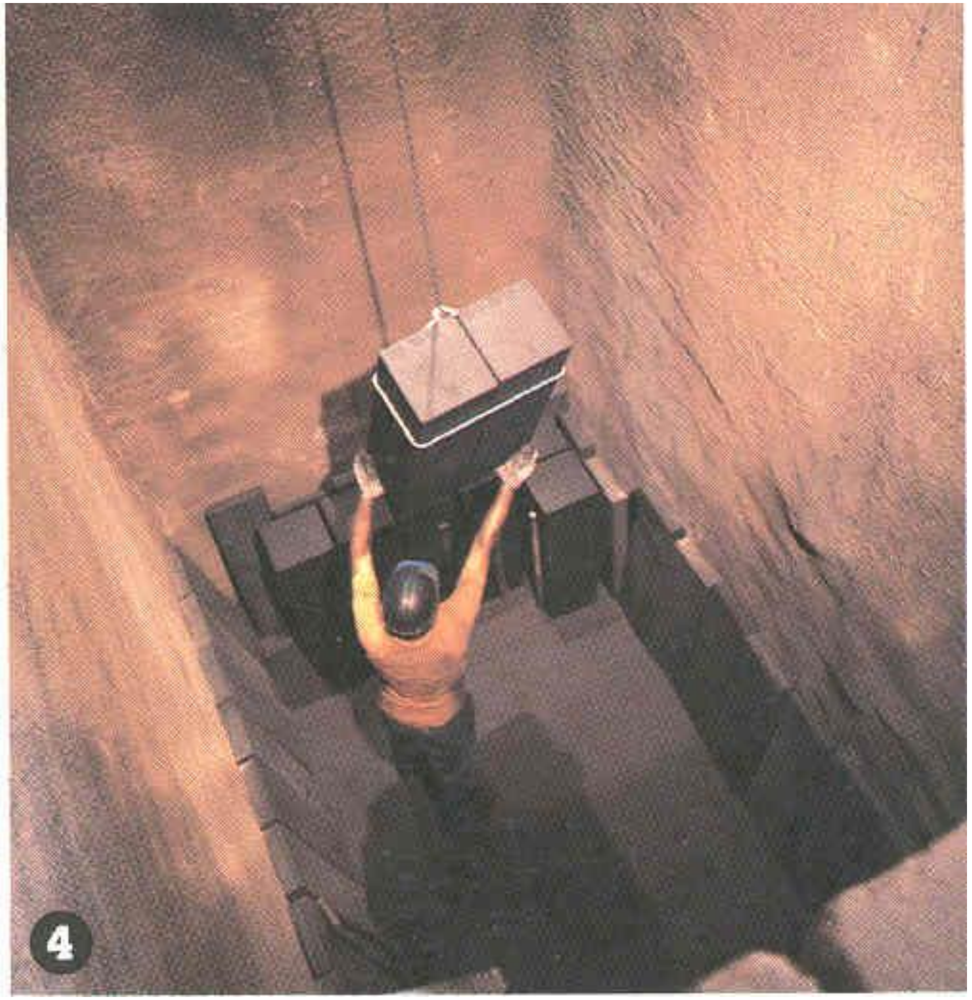
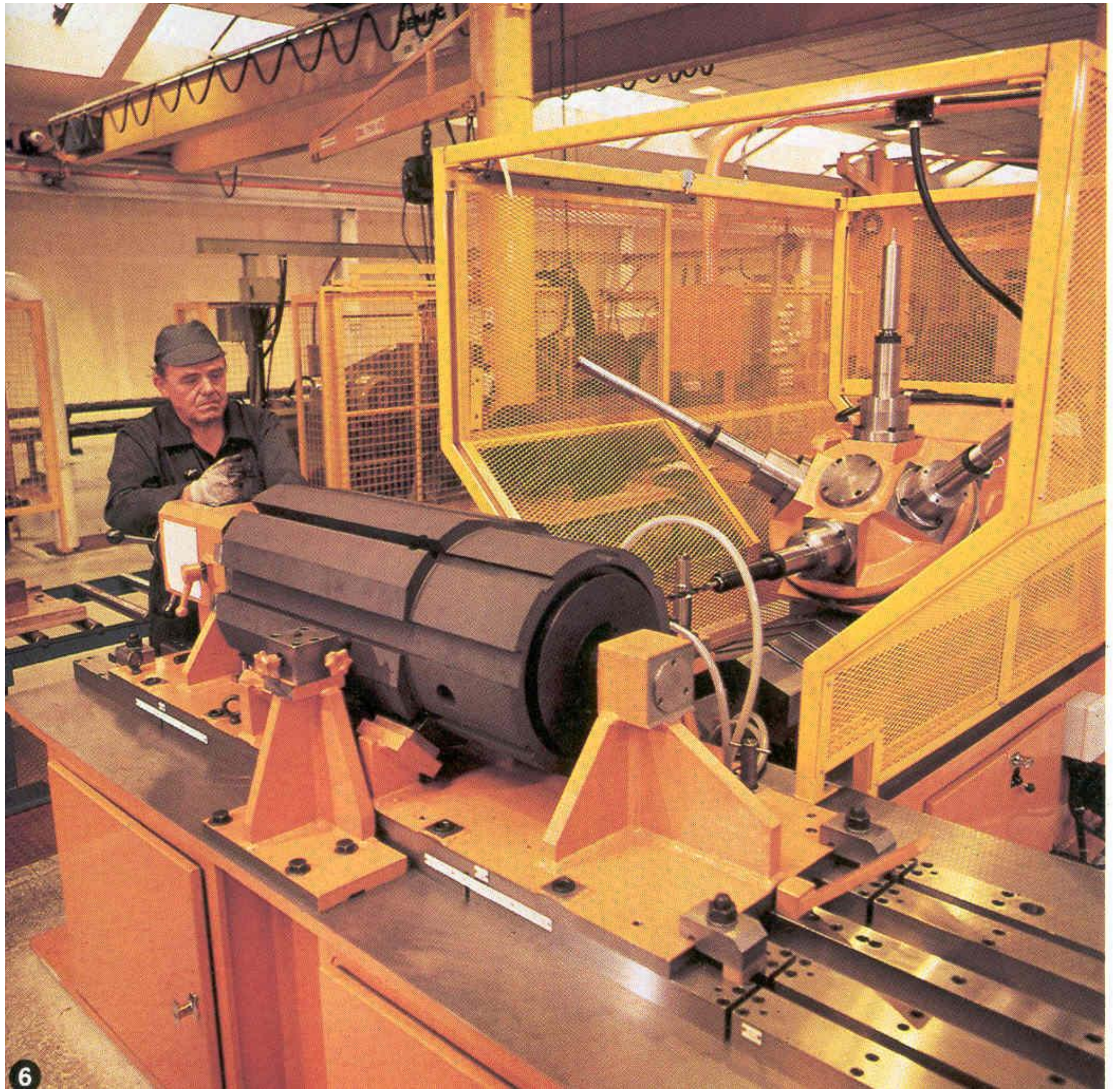
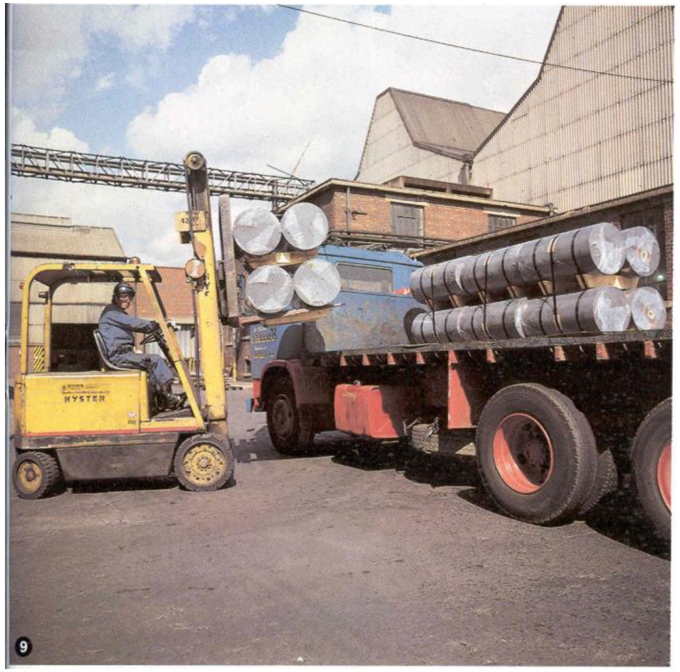




Autoclave Impregnation





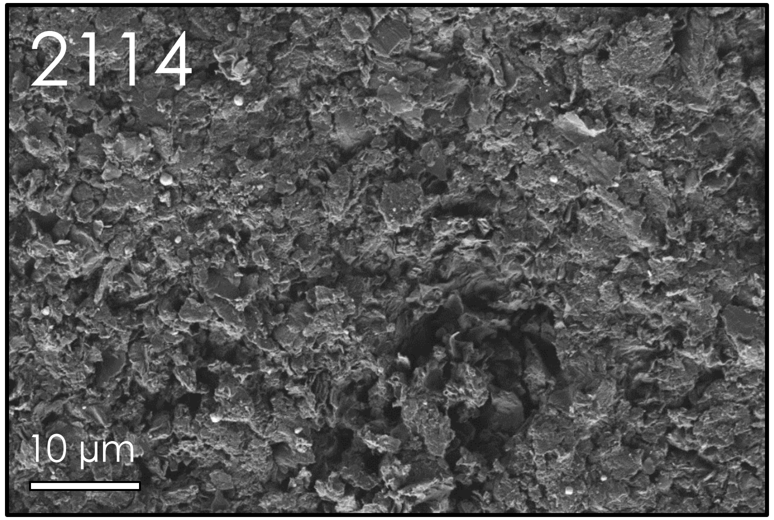
Manufacture, Packing and Transport

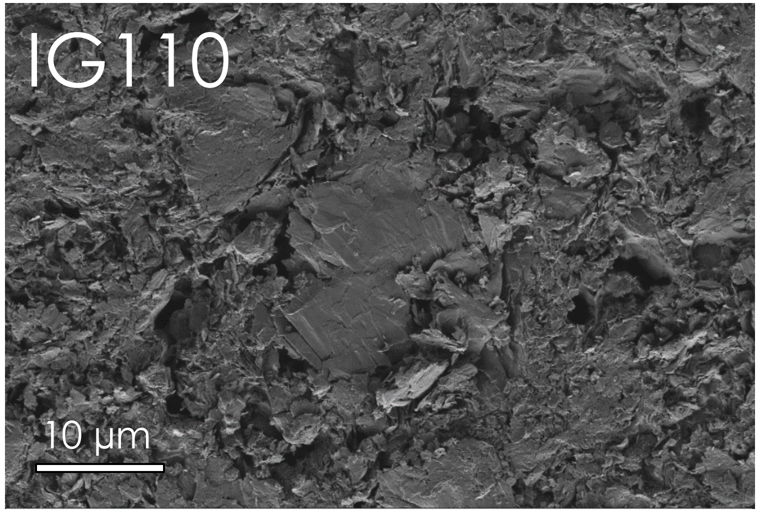
- SGL Carbon Process Technology (reprinted with permission), 20

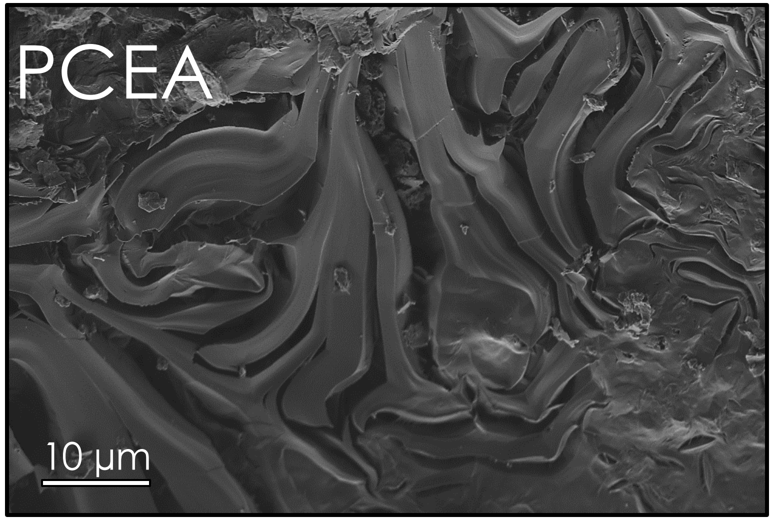
Final Product

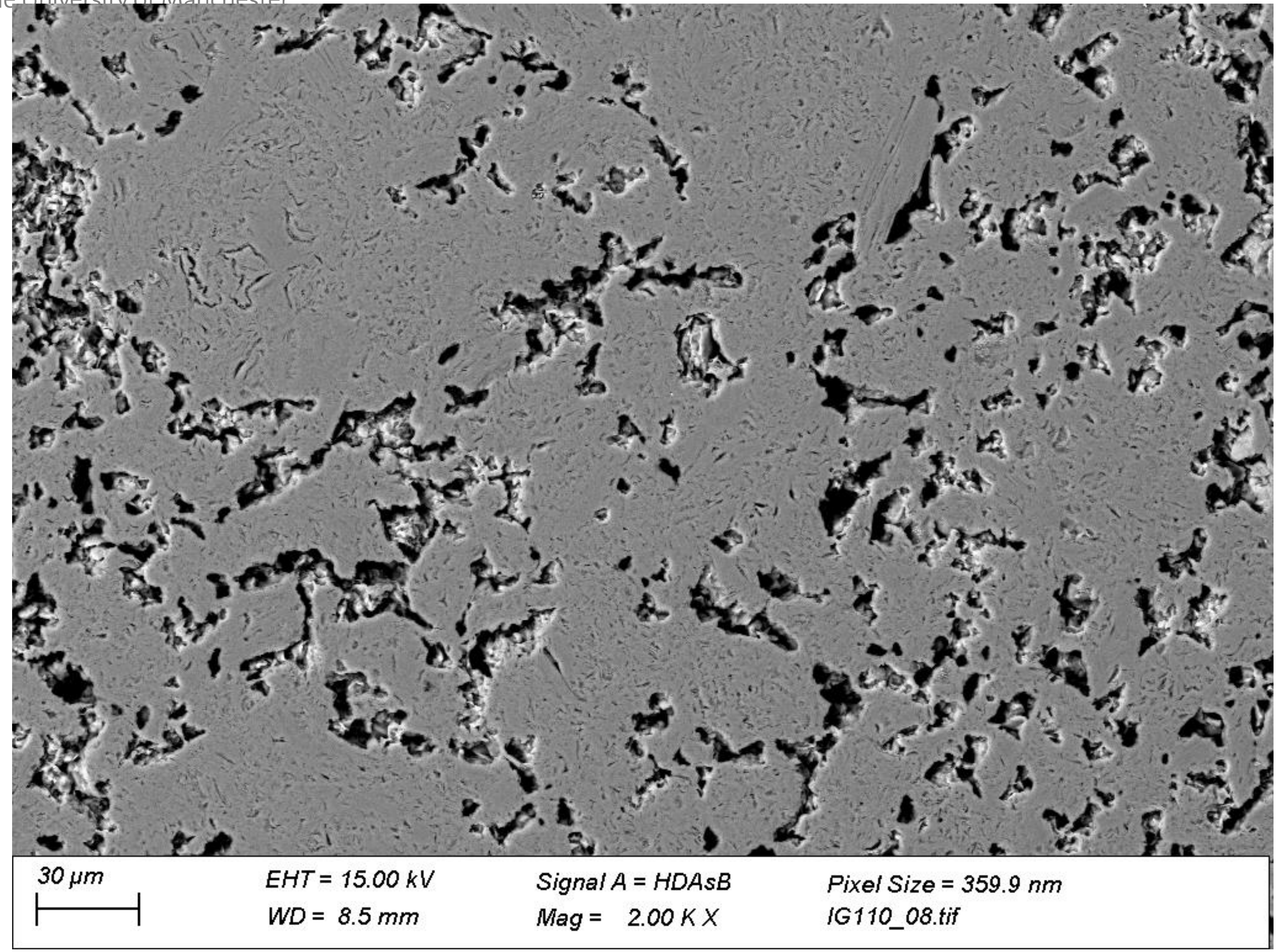
- Either anisotropic or near-isotropic product
- Significant porosity ~20%
  - >10% open porosity, <10% closed porosity
  - Typical Density 1.72 – 1.8g/cm^3
  - High purity – impurities measured in parts per million (ppm)
    - Less than 2ppm Boron EBC

HTGR microstructure example grades



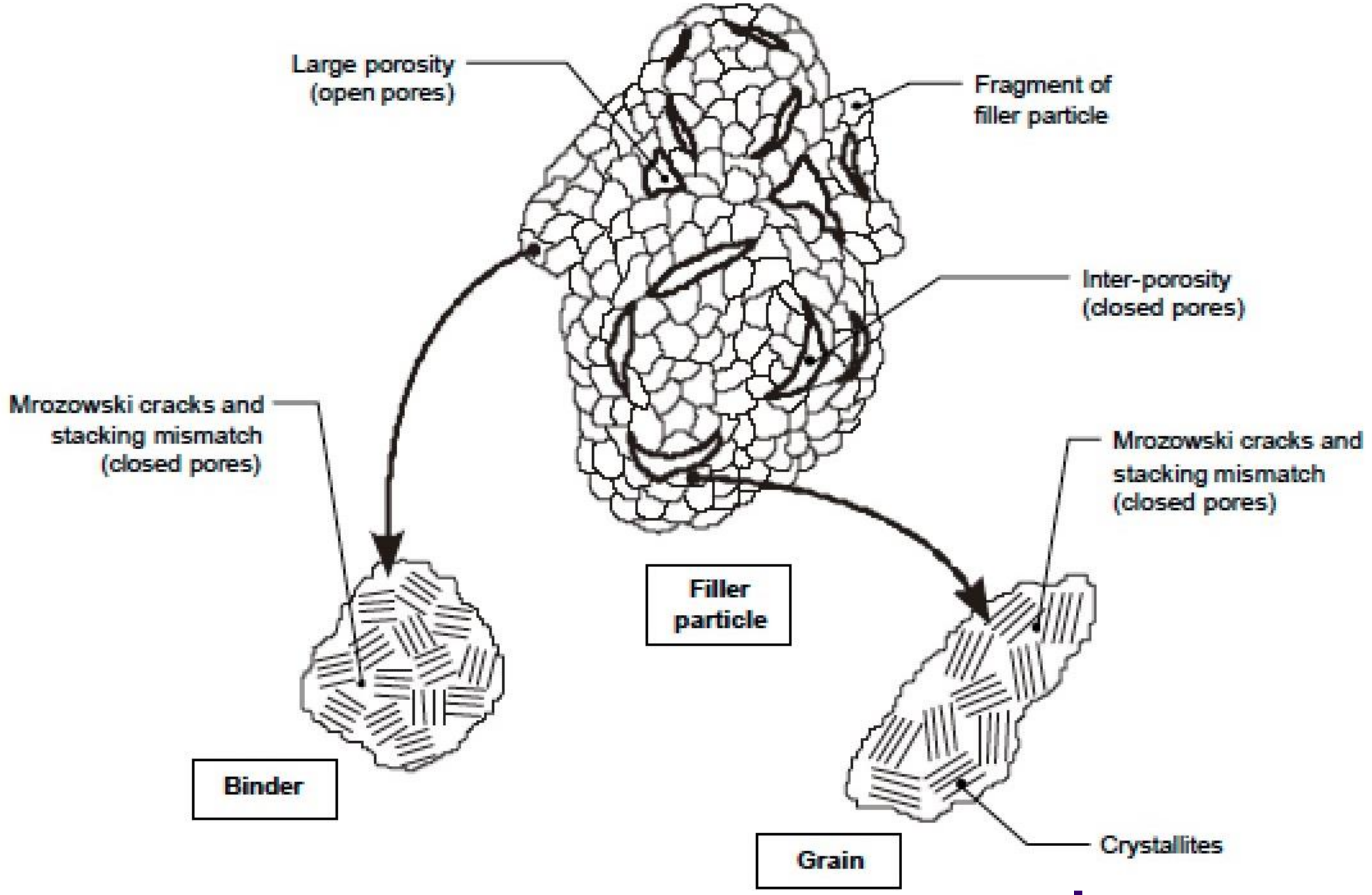


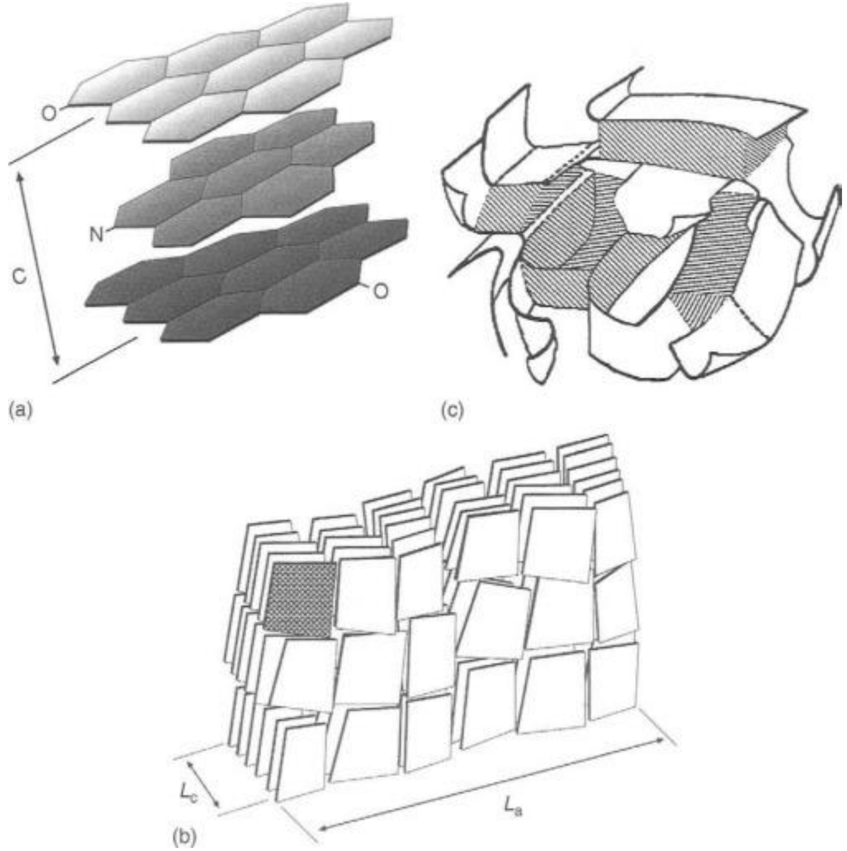




IG110 polished sample showing the pore distribution within the material.

Filler Coke Particle Structure





1. Hall, G., Microstructural modeling of Nuclear grade graphite, 2004
2. Engle G. Irradiation behavior of nuclear graphites at elevated temperatures. Carbon N Y 1971;9:539–54. doi:10.1016/0008-6223(71)90076-5.
3. Activated Carbon, 2006 - https://doi.org/10.1016/B978-0-08-044463-5.X5013-4

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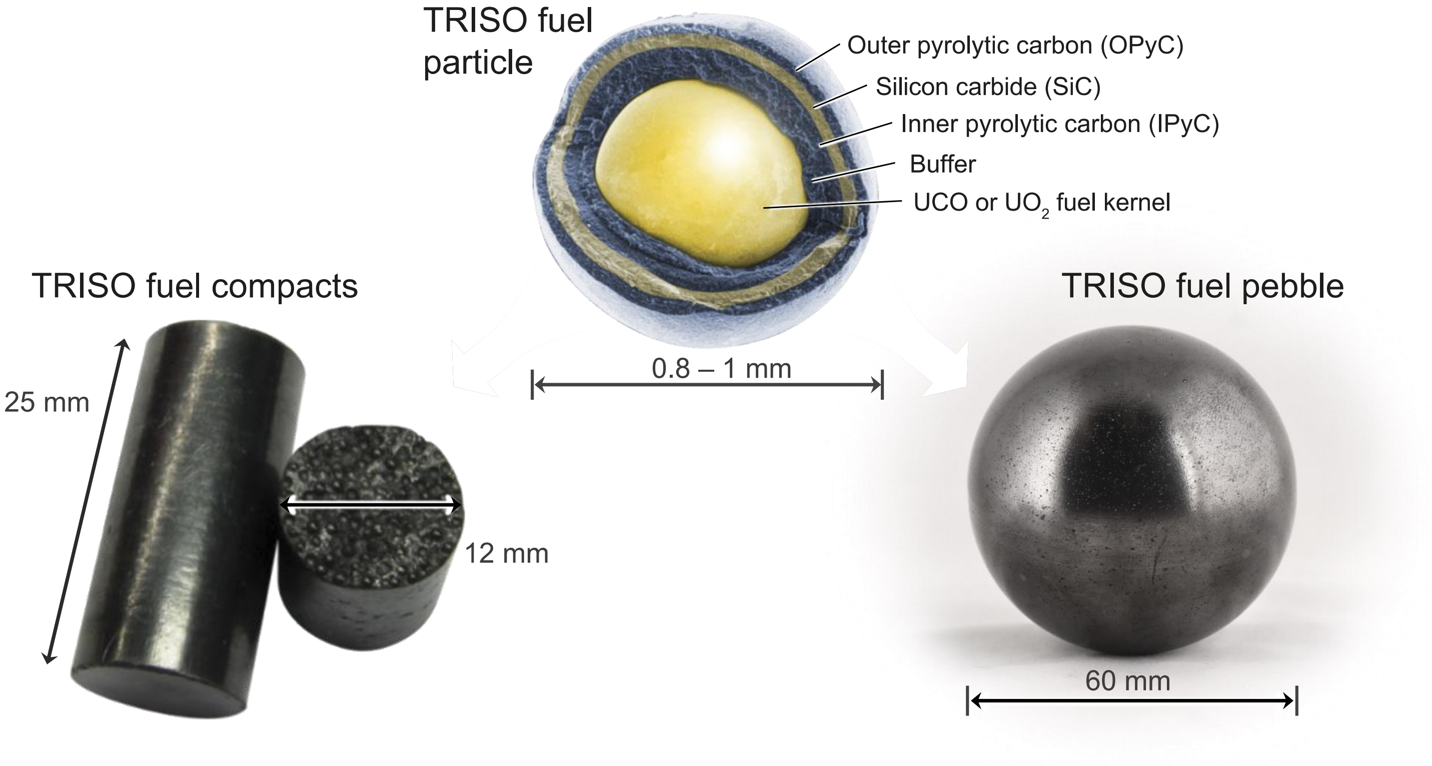
GRAPHITE MATRIX MATERIALS



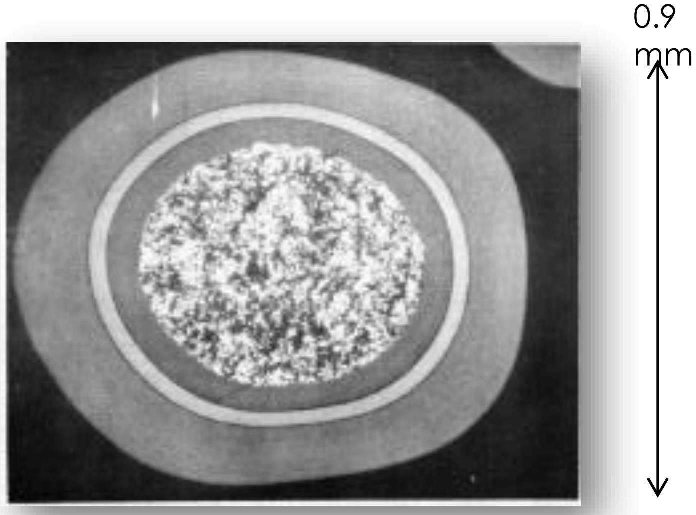
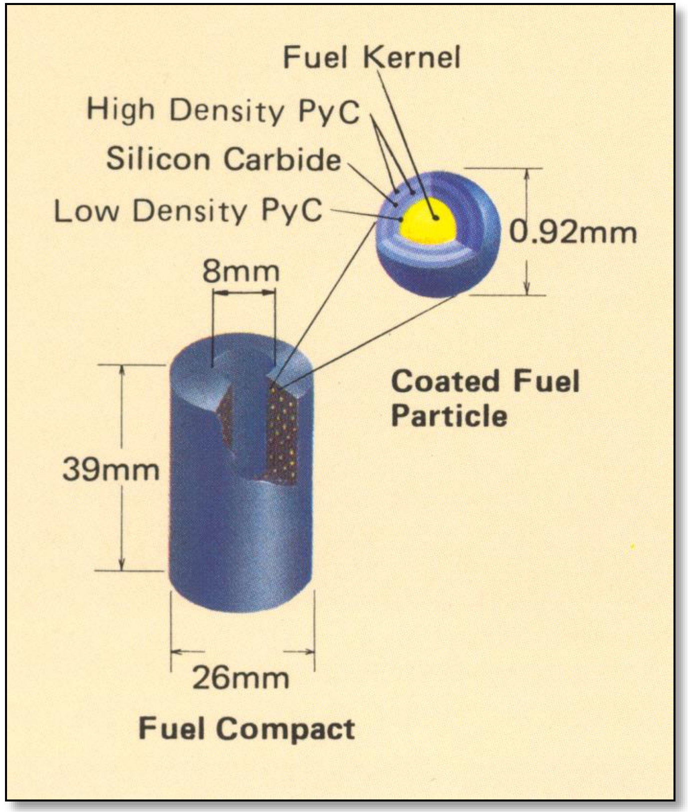


Fig 2: Typical TRISO PyC/SiC/PyC Particle

Fuel Compacts

- In HTRs fuel compact may be spherical or annular in shape, in the Dragon reactor annular compacts were used, although some spherical compacts were used for short experiments.
- A typical fuel compact used in the DRE consists of approx. 10'000 fuel particles embedded within a graphitic matrix material (powder).
- An average compact contains by volume 20% fuel particle, **80% Graphite / PyC matrix**
- Fuel particles were uniformly distributed throughout the matrix compact to avoid any hotspot formation.
- Thousands of these TRISO particles were embedded within a graphite matrix and pressed into an annular shaped fuel compact.



Schematic section through a TRISO particle and fuel compacts.

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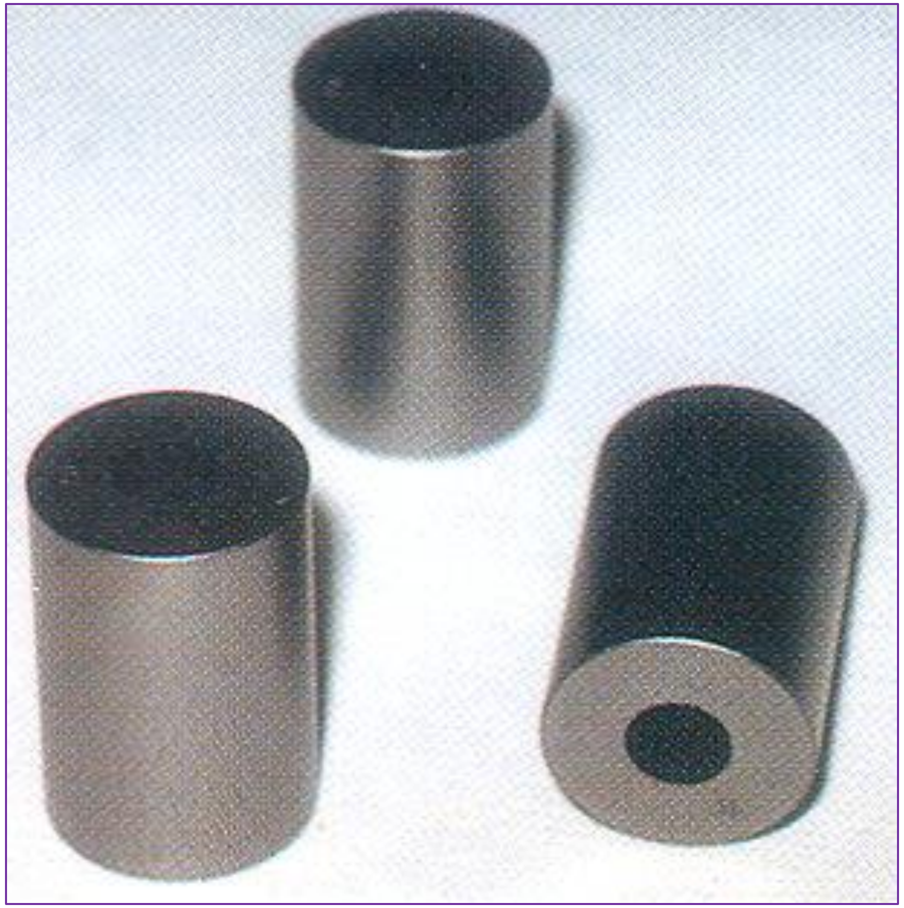
Manufacture of Fuel Compacts

TRISO Particle Preparation	• Uranium-based fuel kernels are coated with multiple layers (buffer PyC, IPyC, SiC, OPyC) to form TRISO particles.
Matrix Powder Preparation	• Graphite powder is selected and mixed with binders (e.g., phenolic resin, polyvinyl butyral). • Wet milling and pulverisation are used to ensure uniform particle size and distribution.
Mixing	• TRISO particles are mixed with the graphite-binder matrix to form a homogeneous mixture.
Compaction	• The mixture is pressed into cylindrical or spherical shapes using isostatic or uniaxial pressing.
Carbonisation	• The green compacts are heated in an inert atmosphere (180 ^{o}C) to carbonise the binder, forming a rigid structure.
Heat Treatment	• Final high-temperature treatment (~1800 ^{o}C), outgasses compacts, improves mechanical strength and removes residual volatiles.
Inspection & Quality Control	• Dimensional checks, density measurements, and mechanical testing ensure the compacts meet specifications.

Manufacture of Fuel Compacts

- This process generally gave a compact with around 6 %wt of carbonised resin with a matrix density of around 1.7 - 1.8 g/cm^3.



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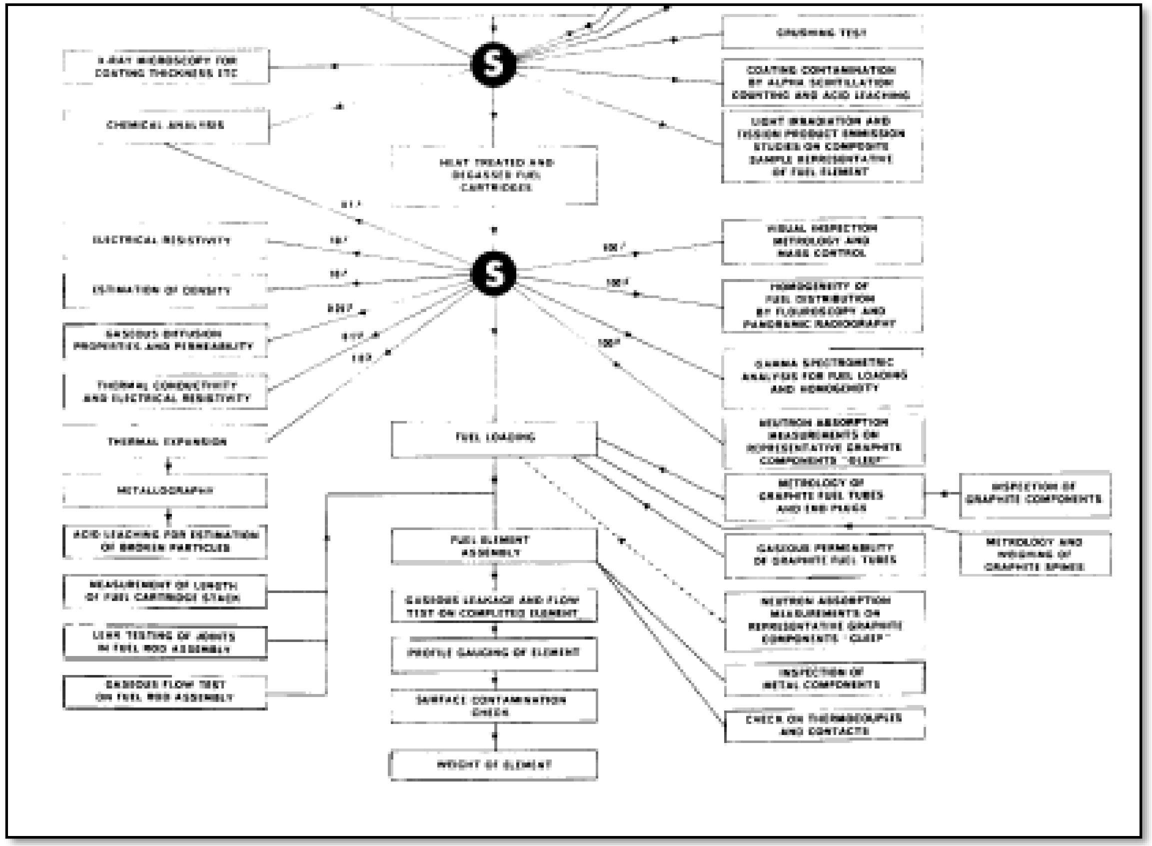
Quality Control -Past Experience DFR

Extensive programme of testing

- Assessment of the fuel particles focused on:
  - Metallographic Structure
  - Density
  - Crushing Strength
  - Contamination
  - Fission Product Release
- Particular attention made to the particle diameter, porosity density and thickness of coatings.
  - For DFR An optical particle size analyser was used for this which could accurately measure 50 particles per second.

Example of QC inspection and sampling points

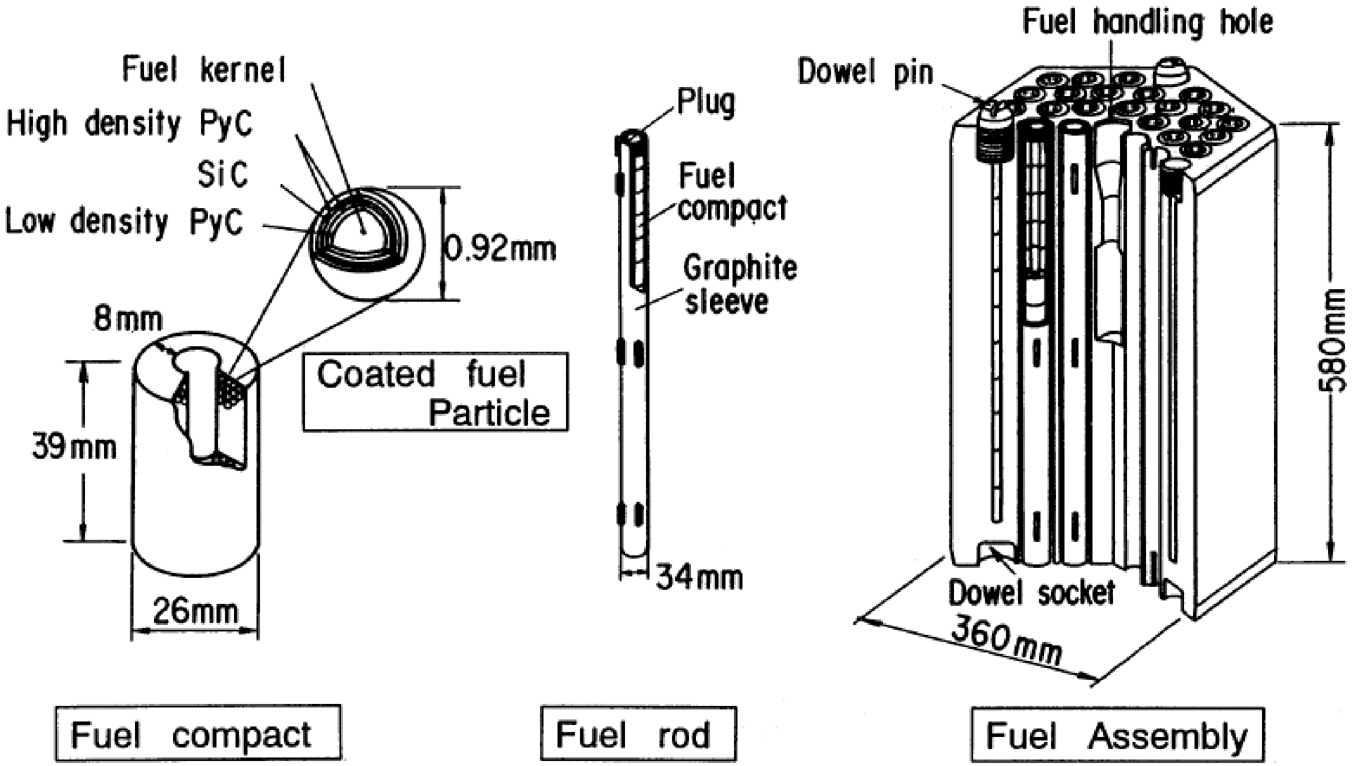


Quality Control (ii) -Past Experience DFR

- Metallographic Structure
  - The crystallite orientation of the PyC layers was assessed using the reflection of a beam of polarised light, the intensity of which was dependant on the crystallographic axes of the graphite crystal.
- Density
  - The PyC and SiC densities were measured using suitable gradient density columns.
  - The kernel density and density of the porous buffer layer was assessed through the particle size analyser.
- The purpose of the coatings was to provide porosity and containment for retention of fission products released from the kernel material;
  - Coating layers were typically 0.1 - 0.2 mm thickness.

Fuel Compact Fabrication

- HTTR process
  - Coated fuel particles are dispersed in a graphite matrix and sintered to form an annular fuel compact as
  - Fuel compacts are contained in a graphite sleeve to form a fuel rod.
  - These fuel rods are inserted into vertical holes bored into the prismatic graphite block.



Pin-in-block prismatic fuel element for the High Temperature Engineering Test Rector (HTTR) in Japan. REF IAEA IAEA-TECDOC-1645

Function of Fuel Compacts

- The primary function of the matrix material was to provide a conducting path for heat generated in the coated particle to the helium cooled graphite structure.
- Further functions include:
  - Local mechanical support to the coated particle.
  - Provide protection against irradiation induced stresses.
  - Must have sufficient strength, irradiation creep, Young's modulus and CTE characteristics.
  - Should act as a sacrificial layer in the case of chemical attack via any oxidants in the coolant i.e. should protect the fuel particles.
  - Should act as a chemically passive sink for any emitted fission products from damaged or defective fuel particles.

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Dragon Fuel Compacts pre-irradiation properties

- Initial Young's Modulus and Strength to fracture of fuel compacts Type A containing 20% by volume of coated particles.

Initial Young's Modulus and Strength of Fuel Compacts (Type A)		
	Young's Modulus (E)	Ultimate Strength (S)
Axial	2.2 GN/M^2	21 MN/M^2 (compressive)
Circumferential	5.8 GN/M^2	12 MN/M^2 (Tensile)

Young's Modulus and Strength of Gilsocarbon		
	Young's Modulus (E)	Ultimate Strength (S)
Gilsocarbon	10.85 GN/M^2	17.5 MN/M^{2}– (compressive) 70 MN/M^2 (tensile)

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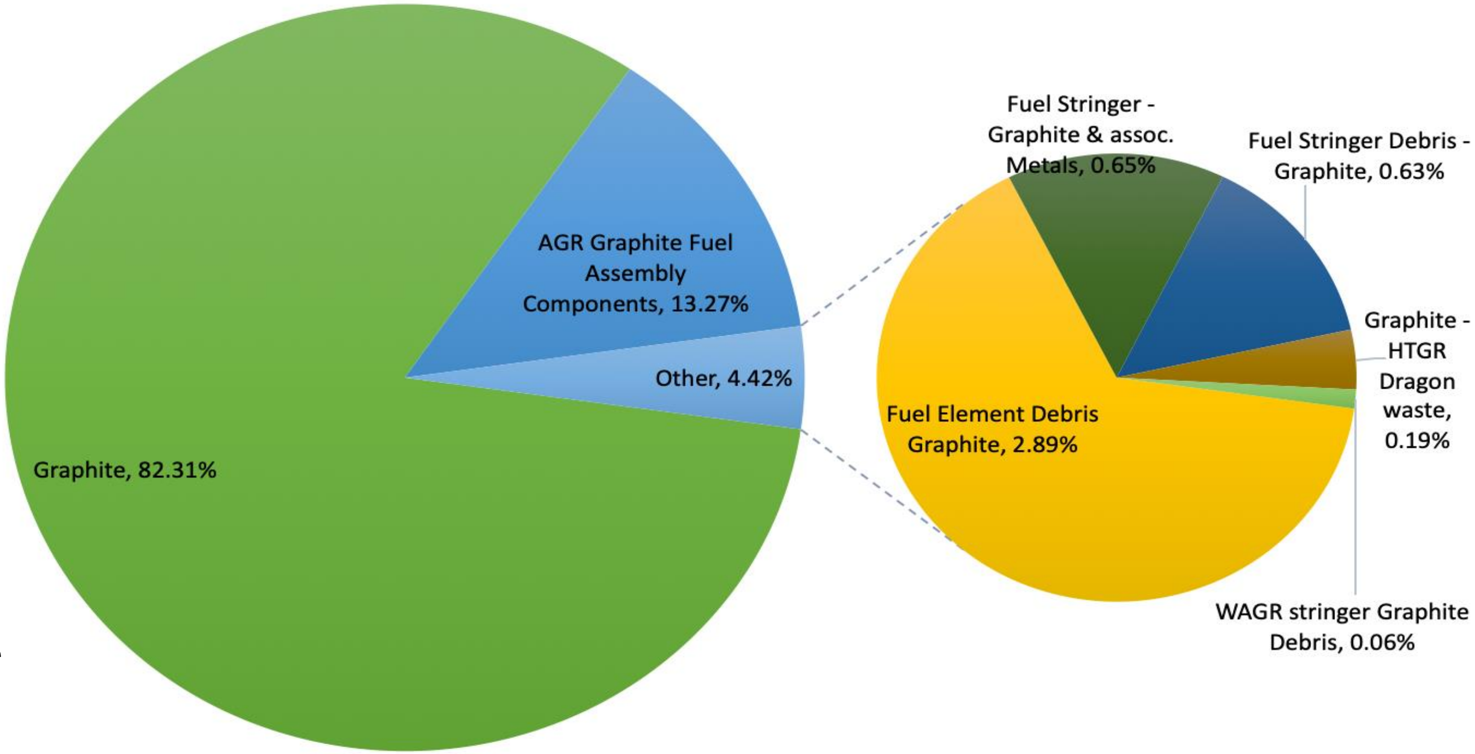
NUCLEAR GRAPHITE WASTE

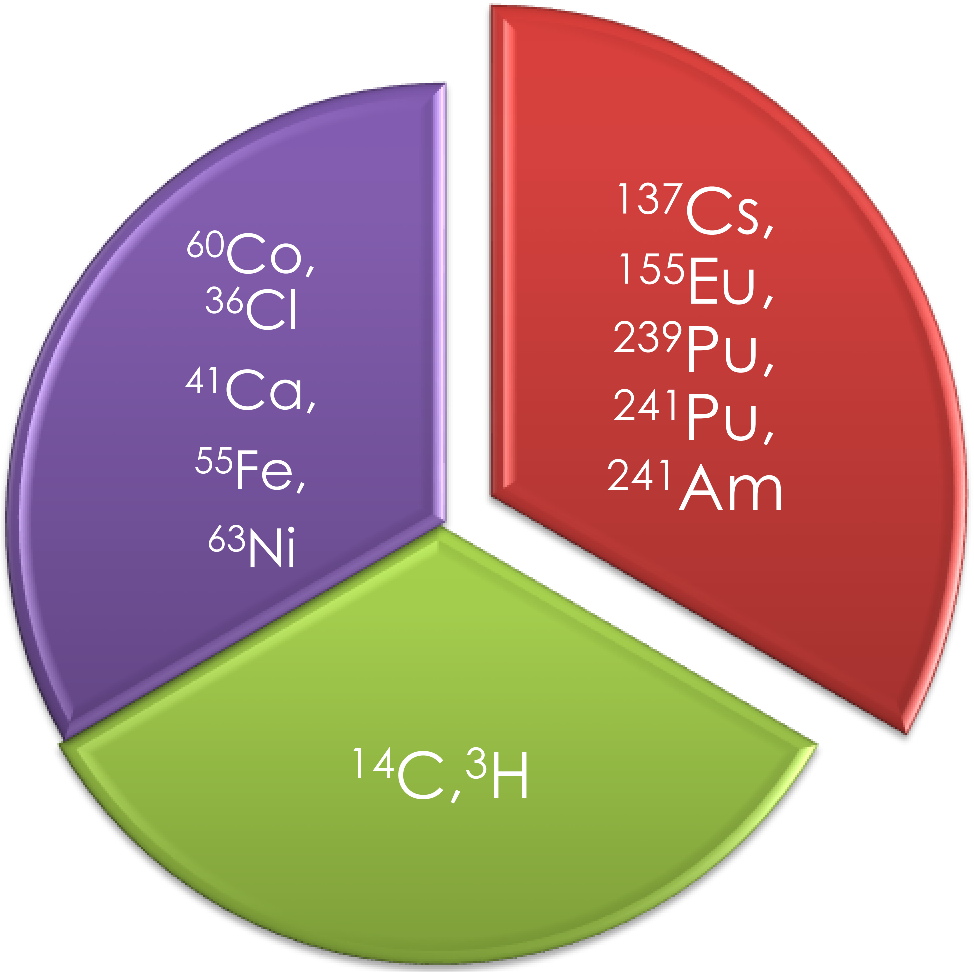
Graphite waste

- ~ 100,000Te of material making it one of the largest ILW streams for GDF disposal
- Magnox, AGR Reactors (operating sites) plus smaller research units & legacy waste
- R&D UK to underpin & optimise Baseline (disposal) and develop treatment and/or re-use options
- The economic benefit to the UK, the estimates from reduced waste storage and diversion from GDF would significant benefits on disposal costs (£ billions)
- The potential volumes of arising future graphite waste may be significant



Graphite Waste and Treatment

- Complexities due to graphite grades, varied operational environment, oxidation and irradiation damage
- Challenges around removal, retrieval, treatment and long term behaviour in a Geological Disposal Facility (GDF)
- Potential to clean, reuse and recycle graphite
- Main impurities of importance include boron, cadmium, nitrogen, iron, cobalt, nickel, and chlorine



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International Graphite Waste Programmes

2006–2012

EPRI "Graphite Decommissioning Initiative"
six comprehensive reviews on all aspects of i-graphite disposal

2007

IAEA Technical Meeting "**Progress in Radioactive Waste Management**", Manchester, UK. -CRP

2008–2013

EU FP7 CARBOWASTE Project
Treatment and Disposal of Irradiated Graphite and Other Carbonaceous Waste

2011–2014

IAEA Collaborative Research Programme "Treatment of Irradiated Graphite to meet Acceptance Criteria for Waste Disposal"
IAEA TECDOC-1790, and CD-ROM, IAEA, Vienna (2016).

2013–2016

EU FP7 CAST Project
('Carbon-14 Source Term')

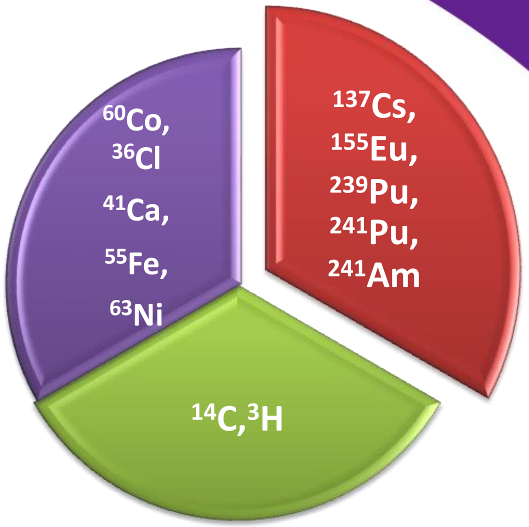
2016–2020

IAEA Collaborative Research Programme on Irradiated Graphite Processing Approaches (GRAPA)
IAEA TECDOC 2072, with appendices, IAEA, Vienna (2024).

2023 –

IAEA Collaborative Research Programme Management of Spent Fuel (Pebble Beds and Prismatic) from High Temperature Gas Cooled Reactors

Graphite Treatment



Graphite treatment could enable the reclassification a significant volume of graphite from ILW to LLW via development of a process to allow for successful decontamination

Scale up the development of graphite treatment technologies with potential opportunities to export technologies worldwide

UK can avoid disposal costs for the large amount of existing and future arising irradiated nuclear graphite ‘waste’, by potentially recycling it into new reactor material, or further potential graphite products for the nuclear industry leading to additional manufacturing savings

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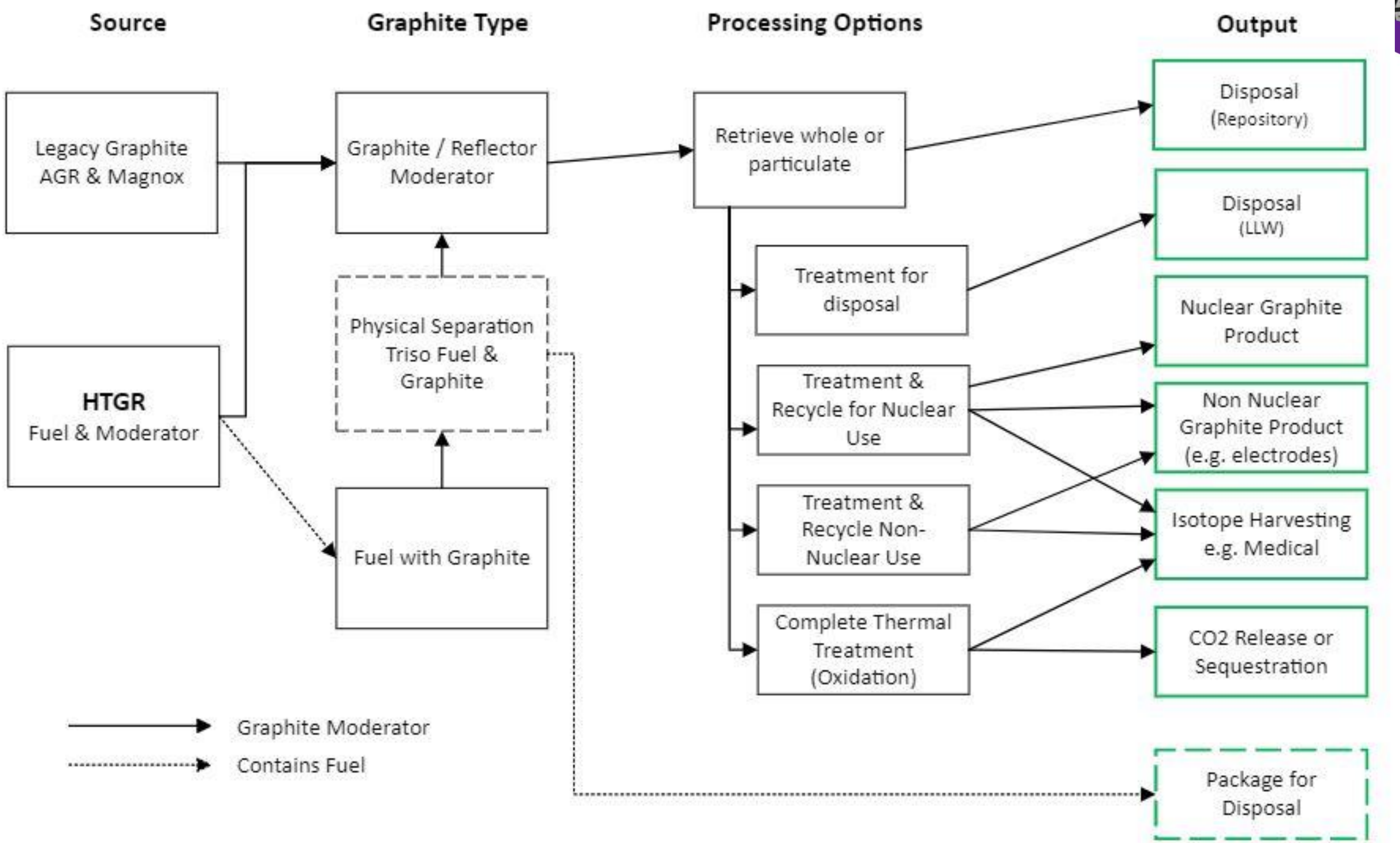
Graphite Treatment

- Many treatment methods previously explored by the nuclear graphite waste community focus on the reduction of graphite radioisotopic content to potentially change the waste category
- However, with our UK (NWS/NRS) revised strategy for rolling Magnox reactor decommissioning and the emergence of HTGR and other Gen IV technologies, this has created opportunities to develop and improve technologies and innovative approaches to the retrieval, storage and treat reactor graphite
- TRL levels for the majority of graphite treatments are low, without further RD&I the UK may not be able to demonstrate the benefit of these technologies achieve decontaminations factors required to reclassify this large volume of ILW material

Bulk Oxidation	Thermal treatment in an oxidising atmosphere	Thermal decontamination	Chemical Treatment	High-Temperature Molten Salt Treatment (HTMST)	Direct re-use of irradiated graphite

6 - 4 → TRL → 2 - 1



Source
Graphite Type
Processing Options
Output
Legacy Graphite AGR & Magnox
HTGR Fuel & Moderator
Graphite / Reflector Moderator
Physical Separation Triso Fuel & Graphite
Fuel with Graphite
Retrieve whole or particulate
Treatment for disposal
Treatment & Recycle for Nuclear Use
Treatment & Recycle Non-Nuclear Use
Complete Thermal Treatment (Oxidation)
Disposal (Repository)
Disposal (LLW)
Nuclear Graphite Product
Non Nuclear Graphite Product (e.g. electrodes)
Isotope Harvesting e.g. Medical
CO2 Release or Sequestration
Package for Disposal
Graphite Moderator
Contains Fuel

Graphite Waste R&D Programmes

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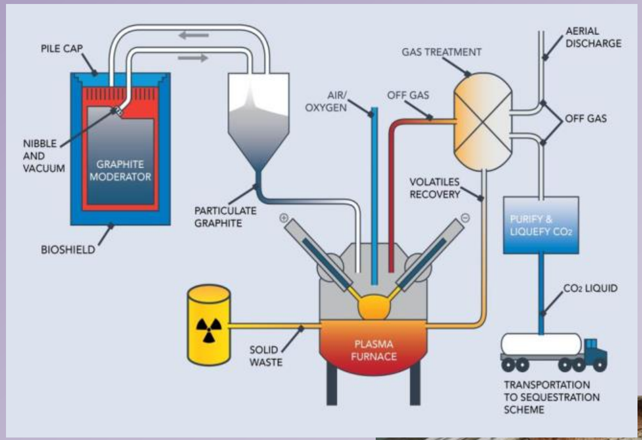
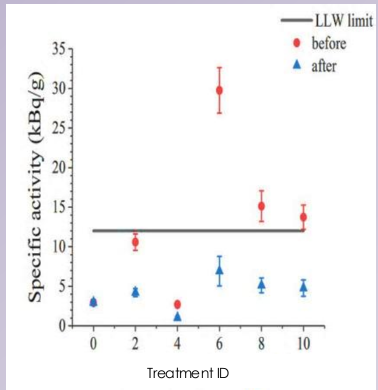
Thermal treatment

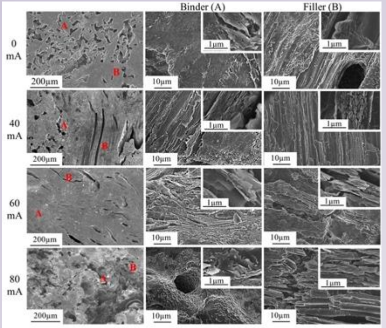


From core to capture

500 Kg graphite treated at Pilot scale

Clean-up using molten salts





*Patent submitted for MS treatment

[1] Grebennikova T, Jones AN, Sharrad CA Energy Environ Sci 2021. doi:10.1039/d1ee00332a.

[2] Worth RN, Theodosiou, Wickhamd AJ, Jones AN, J Nucl Mater 2021; doi:10.1016/j.jnucmat.2021.153167.

[3] Theodosiou A, Jones AN, J Nucl Mater 2018. doi:10.1016/j.jnucmat.2018.05.002.

[4] Theodosiou A, Jones PLoS One 2017;12:1–19. doi:10.1371/journal.pone.0182860.

UKRI Innovate UK

IAEA International Atomic Energy Agency

EPSRC Pioneering research and skills

Thank you – Happy to answer any questions

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