GEN IV International Forum
Expertise | Collaboration | Excellence

Overview of Nuclear Graphite R&D in Support of Advanced Reactors

Dr. Will Windes

Idaho National Laboratory, USA

5 April 2023

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Meet the Presenter

Dr. Windes has over 35 years' experience in extreme materials research with the majority being in nuclear materials. His material interests range widely from solid oxide fuel cell development to space nuclear propulsion systems to spent nuclear fuel issues. However, his focus for the past 20 years has been in the areas of nuclear graphite and carbon-based composite materials for the new High Temperature Reactor design. As the Advanced Reactor Technologies graphite program technical lead, he has overseen the large Advanced Graphite Creep (AGC) irradiation experiment at INL, developed one of the largest unirradiated nuclear graphite material property databases, is the current chair in developing ASME graphite code, and has numerous interactions with the NRC, international organizations, and commercial HTR vendors on graphite related issues. Dr. Windes holds a doctorate in Material Science from the University of Idaho and a Master and Bachelor in Nuclear Engineering from the University of Illinois and UC Santa Barbara, respectively.

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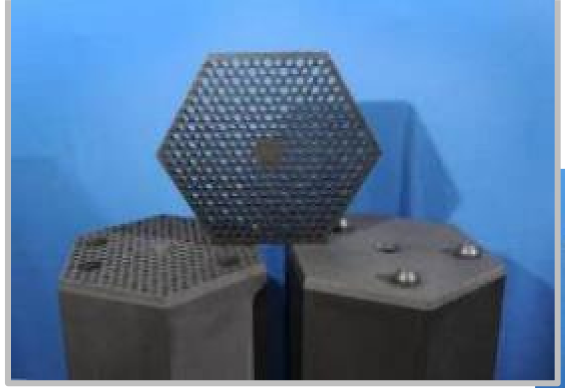
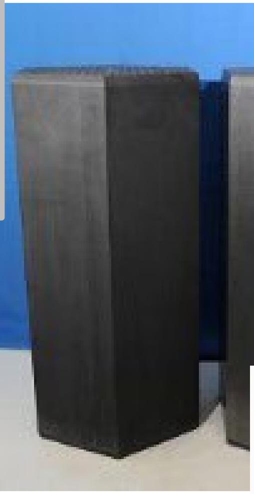
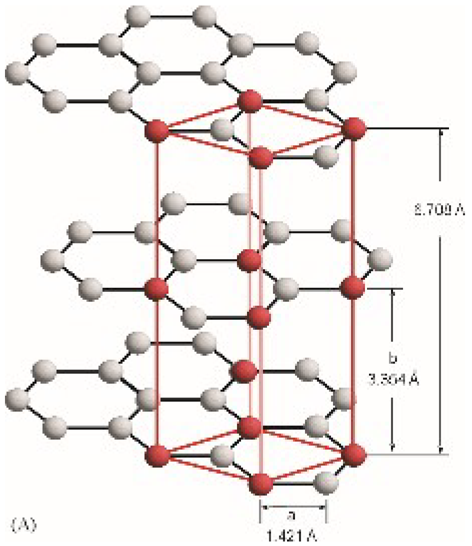
Wilna Geringer geringerjw@ornl.gov	ASME, Composites, Graphite

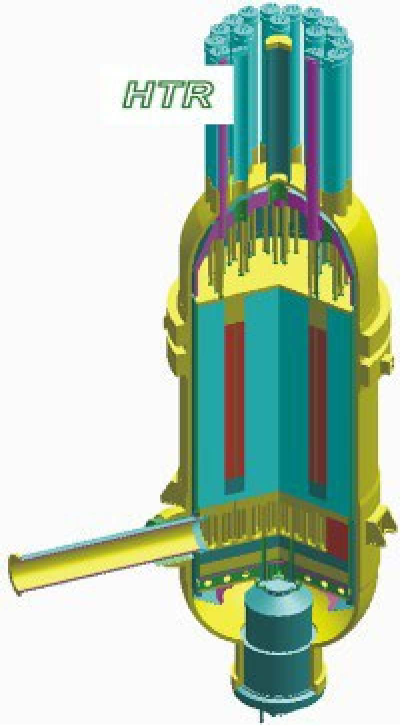
Discussion Points

- Why are we talking about graphite?
- What makes graphite tick?
  - Graphite crystal and microstructure (not to detailed)
  - Anisotropy and pores
  - Engineered composite
- Graphite behavior
  - Irradiation
  - Oxidation
- Some speculation on graphite behavior
  - Interesting irradiation mechanisms
  - Why graphite can not, will not burn
  - Molten salt interactions (a prelude)









The Nuclear Renaissance

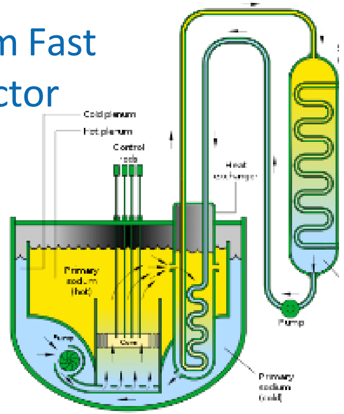
(Since about 2001) the term nuclear renaissance has been used to refer to a possible nuclear power industry revival:

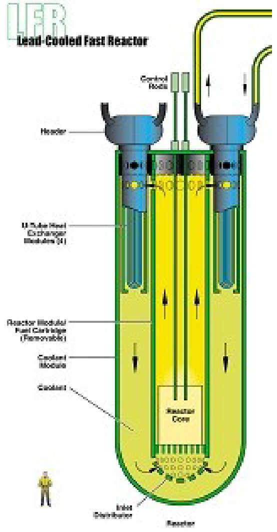
- Rising fossil fuel prices
- Limiting greenhouse gas emission

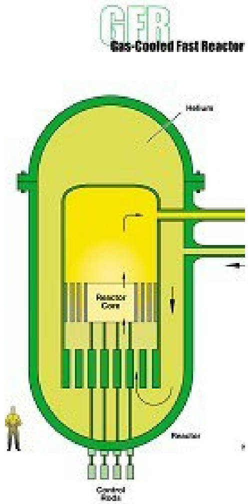
Generation IV reactor designs

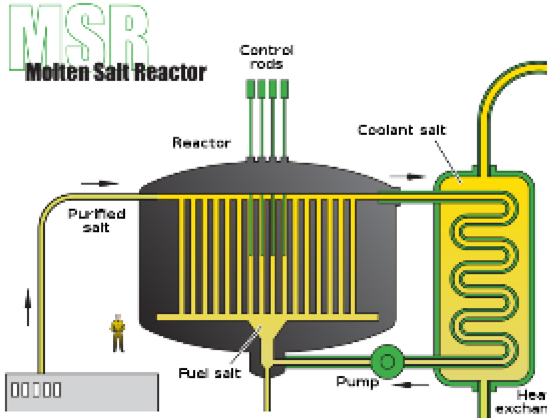
- Inherently & Passively safe
  - Natural shutdown and cooling from design
- New designs = new uses
  - Process heat
  - Small modular designs
  - Variety of coolants and fuels

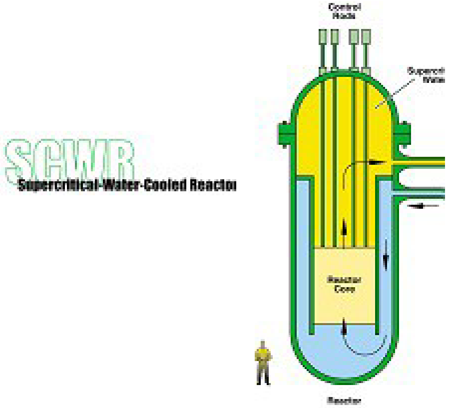
Sodium Fast Reactor

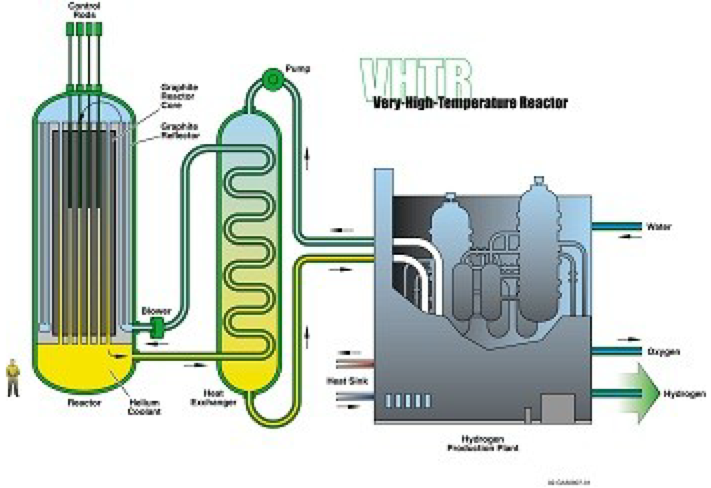






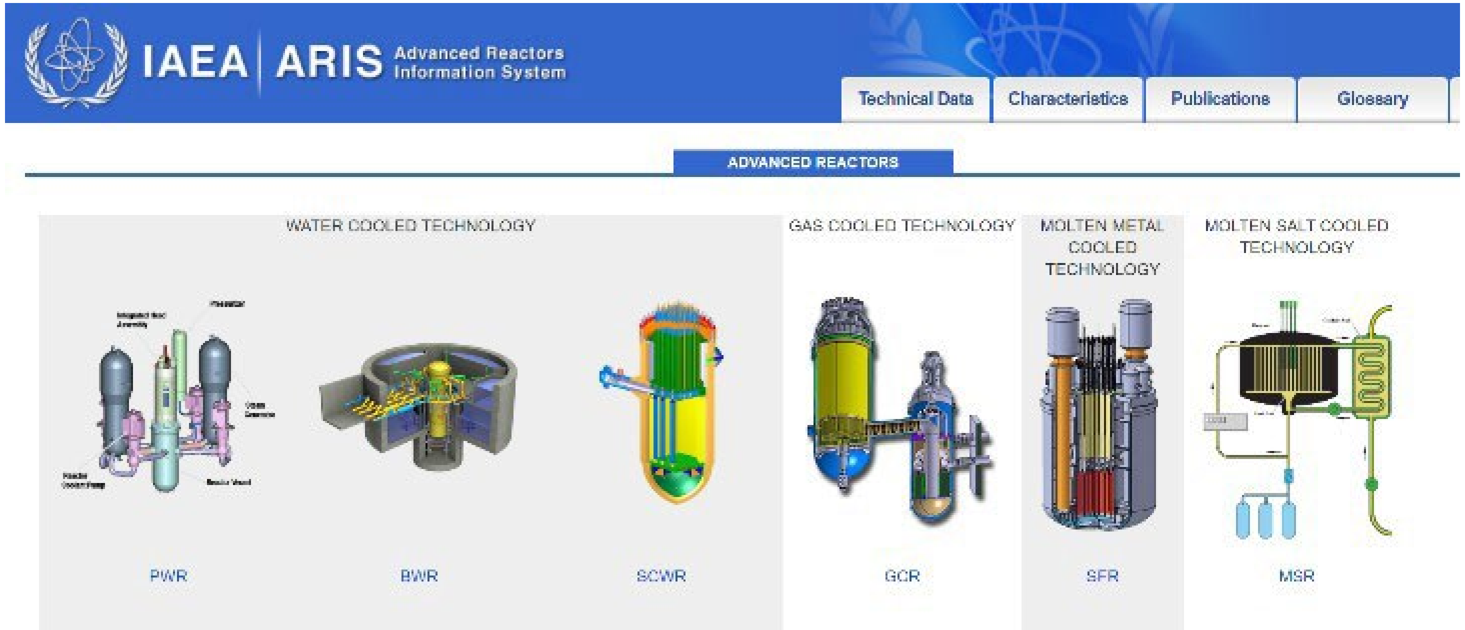


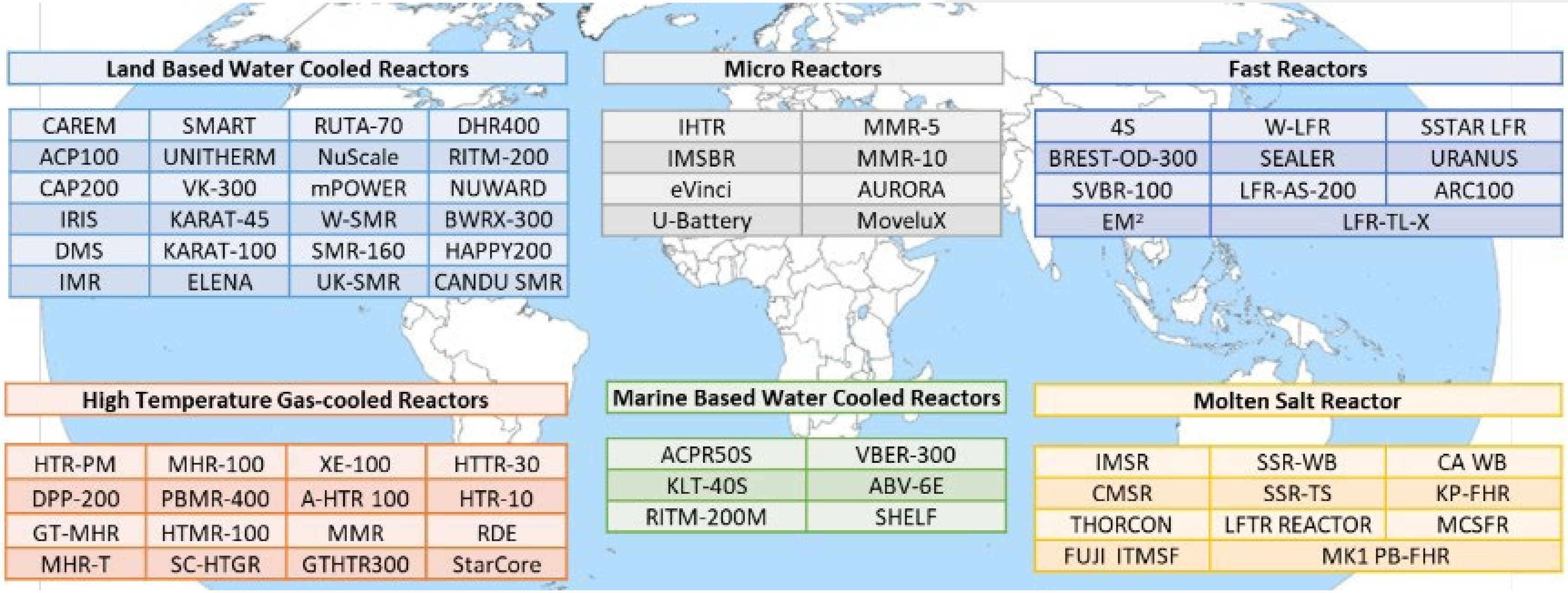




Advanced Reactors Worldwide

More than 70 advanced reactor projects worldwide under development





Land Based Water Cooled Reactors

CAREM	SMART	RUTA-70	DHR400
ACP100	UNITHERM	NuScale	RITM-200
CAP200	VK-300	mPOWER	NUWARD
IRIS	KARAT-45	W-SMR	BWRX-300
DMS	KARAT-100	SMR-160	HAPPY200
IMR	ELENA	UK-SMR	CANDU SMR

Micro Reactors

IHTR	MMR-5
IMSBR	MMR-10
eVinci	AURORA
U-Battery	MoveluX

Fast Reactors

4S	W-LFR	SSTAR LFR
BREST-OD-300	SEALER	URANUS
SVBR-100	LFR-AS-200	ARC100
EM^2	LFR-TL-X	

High Temperature Gas-cooled Reactors

HTR-PM	MHR-100	XE-100	HTTR-30
DPP-200	PBMR-400	A-HTR 100	HTR-10
GT-MHR	HTMR-100	MMR	RDE
MHR-T	SC-HTGR	GTHTR300	StarCore

Marine Based Water Cooled Reactors

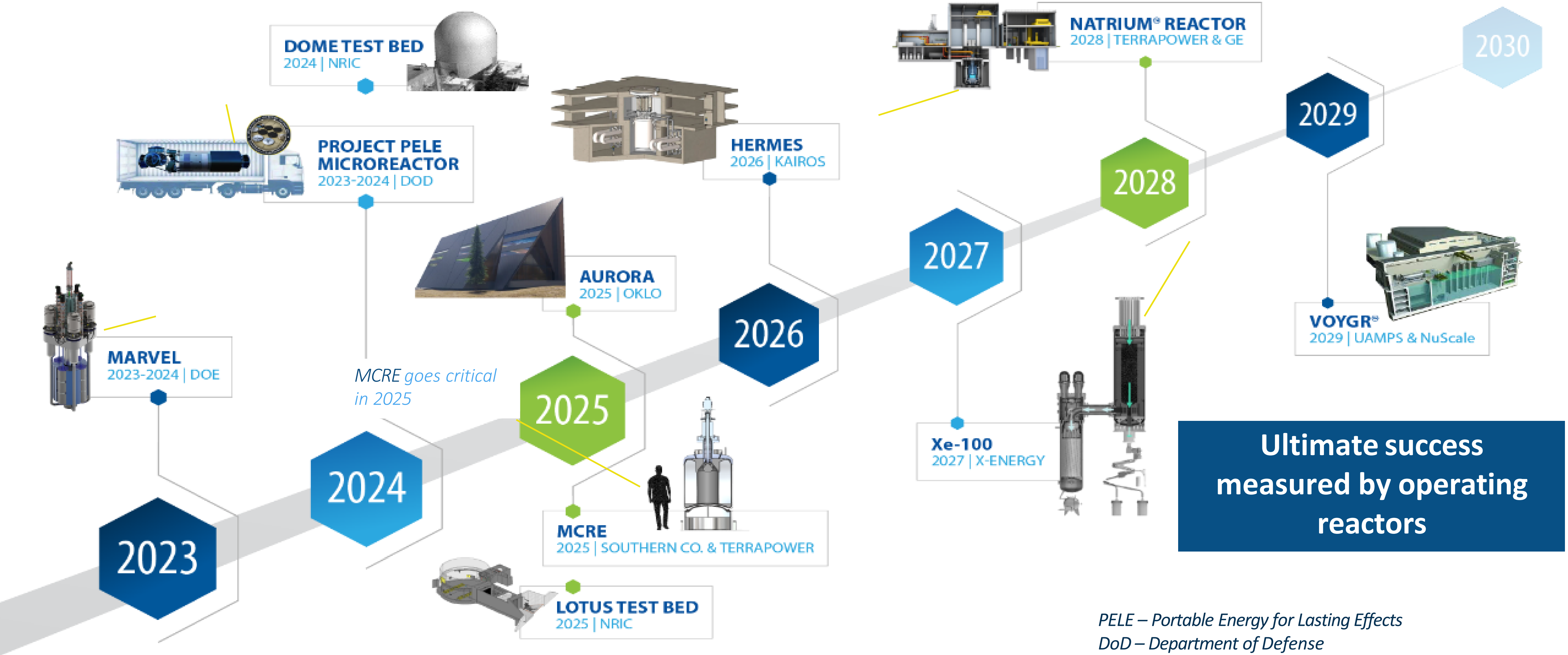
ACPR50S	VBER-300
KLT-40S	ABV-6E
RITM-200M	SHELF

Molten Salt Reactor

IMSR	SSR-WB	CA WB
CMSR	SSR-TS	KP-FHR
THORCON	LFTR REACTOR	MCSFR
FUJI ITMSF	MK1 PB-FHR	

https://aris.iaea.org/

On schedule for deployment in next few years



MARVEL
2023-2024 | DOE

PROJECT PELE MICROREACTOR
2023-2024 | DOD

DOME TEST BED
2024 | NRIC

MCRE goes critical in 2025

AURORA
2025 | OKLO

MCRE
2025 | SOUTHERN CO. & TERRAPOWER

LOTUS TEST BED
2025 | NRIC

HERMES
2026 | KAIROS

Xe-100
2027 | X-ENERGY

NATRIUM® REACTOR
2028 | TERRAPOWER & GE

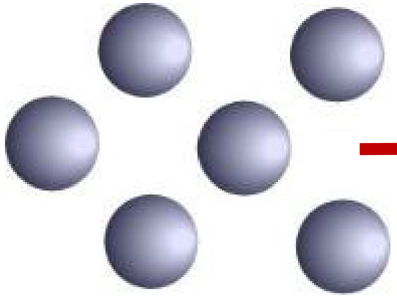
VOYGR®
2029 | UAMPS & NuScale

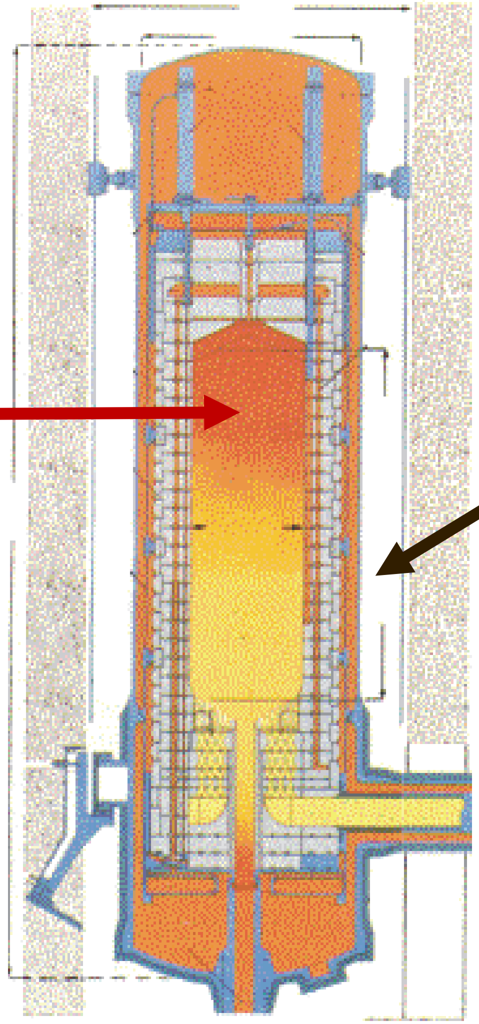
Ultimate success measured by operating reactors

PELE – Portable Energy for Lasting Effects
DoD – Department of Defense
DOME – Demonstration of Microreactor Experiments
NRIC – National Reactor Innovation Center
LOTUS – Laboratory for Operating and Testing in the U.S.

(Very) High Temperature Reactor (HTR)

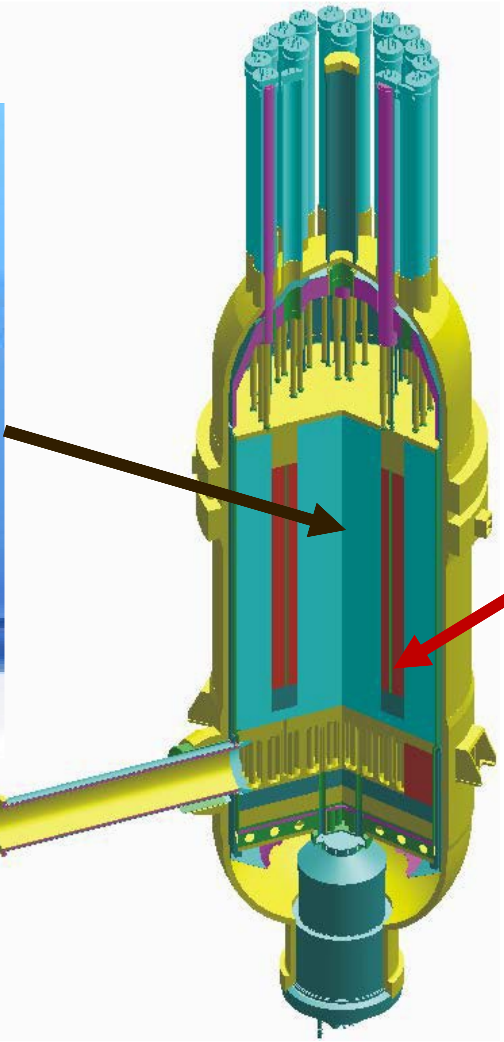
(Graphite)
Fuel Pebbles





Graphite Core

Gas or molten
salt coolant

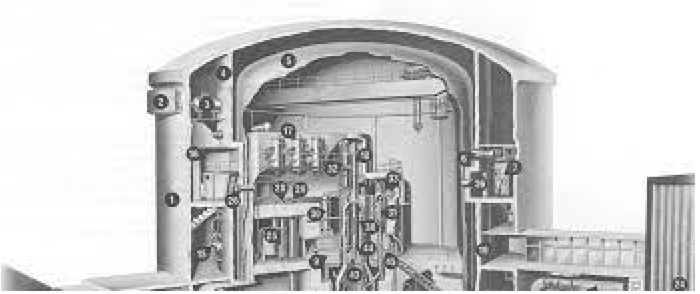


Graphite Fuel
Element

Pebble Bed

Prismatic

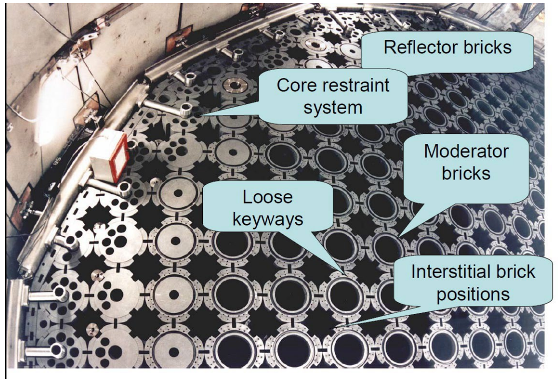
High Technology Readiness (Previous graphite Rx)



Dragon Reactor
1965 to 1976

Magnox Reactors
1956 to 2015

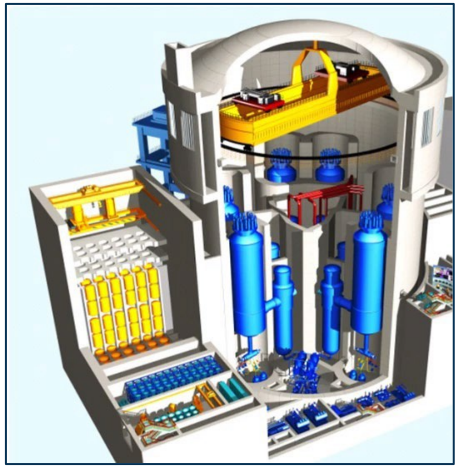
Fort St Vrain
1979 to 1989



Advanced Gas Reactors
1967 to present

AVR
1967 to 1988

THTR
1983 to 1989



HTR-PM
2022 to present

Peach Bottom
1966 to 1974

HTTR
2002 to present

HTR-10
2003 to present

Renaissance is happening now

What started in 2001 as government funded R&D projects has evolved into numerous commercial enterprises world-wide

A wide variety of different designs

- Coolant
  - Water, inert gas, molten salt, fueled salt, heat pipe
- Fuel type
  - TRISO, UO2, UN, other
- Size
  - SMR & micro ⟶ large "conventional"

Xenergy

AREVA

Kairos Power

moltex clean energy

ThorCon

HolosGen™

BWXT
BWX Technologies, Inc.

Westinghouse

flibe ENERGY

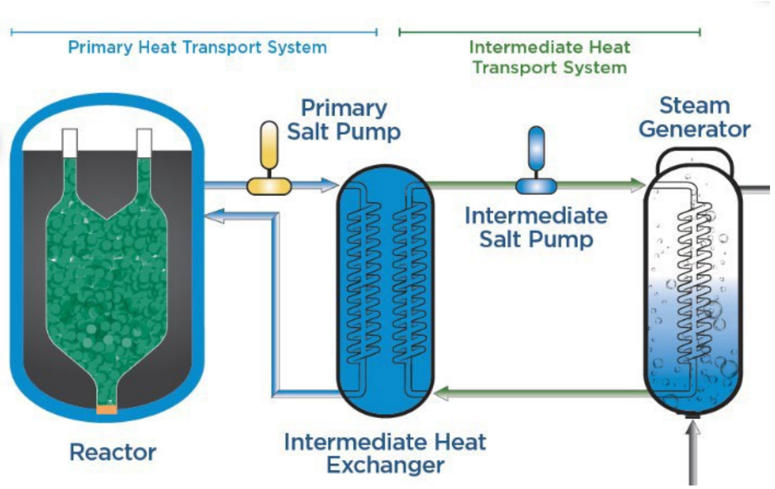
RADIANTPOWERCORP
A HEICO COMPANY

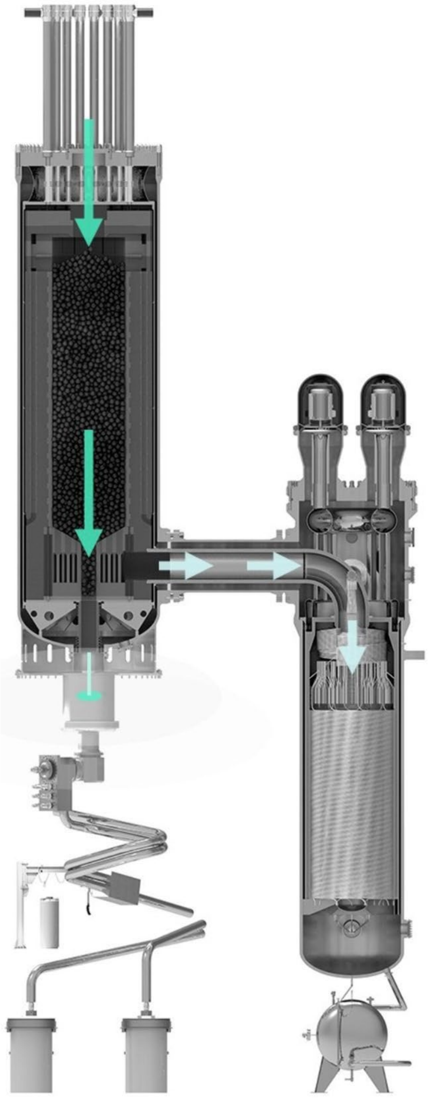
Ultra Safe Nuclear

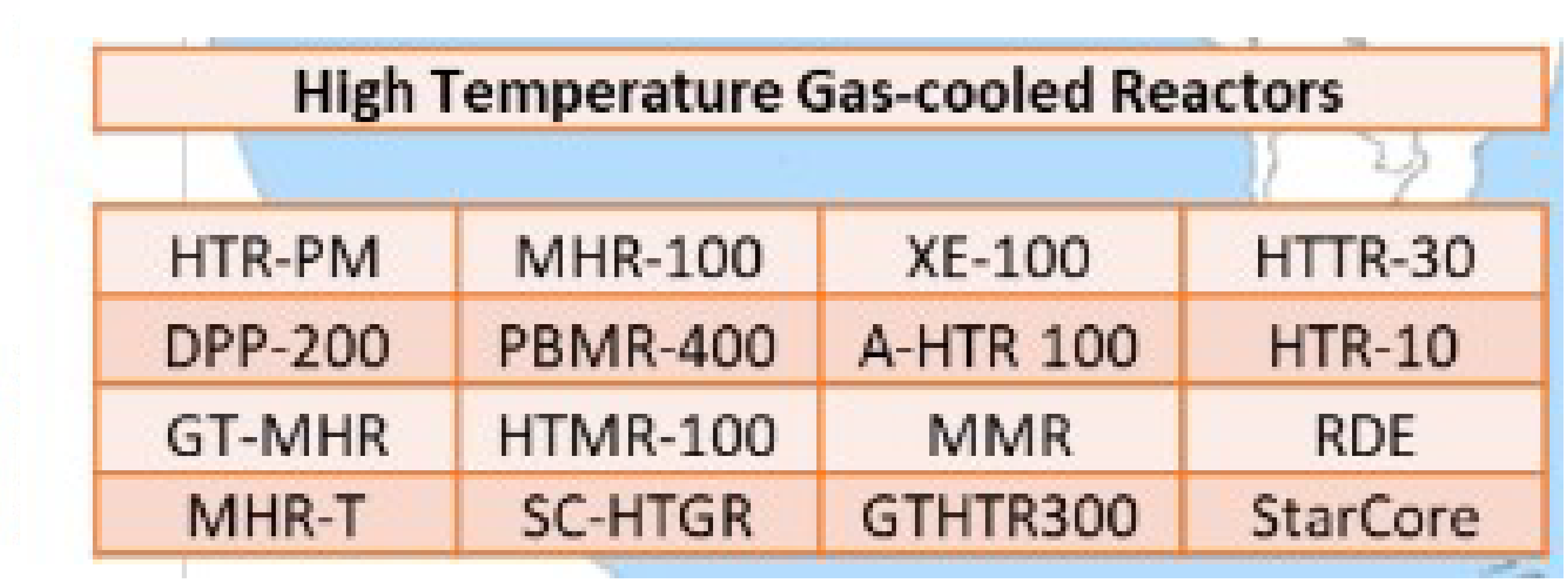
Graphite is important

(V)HTR: one of the 1st GEN IV Designs

- Highest technology readiness
- ***Graphite*** is important material for GEN IV design







High Temperature Gas-cooled Reactors

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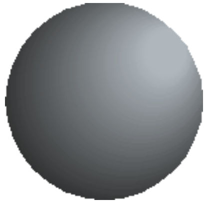
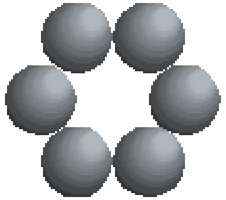
Graphite’s unique structure key to behavior

Begins with benzene-like ring of carbon atoms – covalent bonds

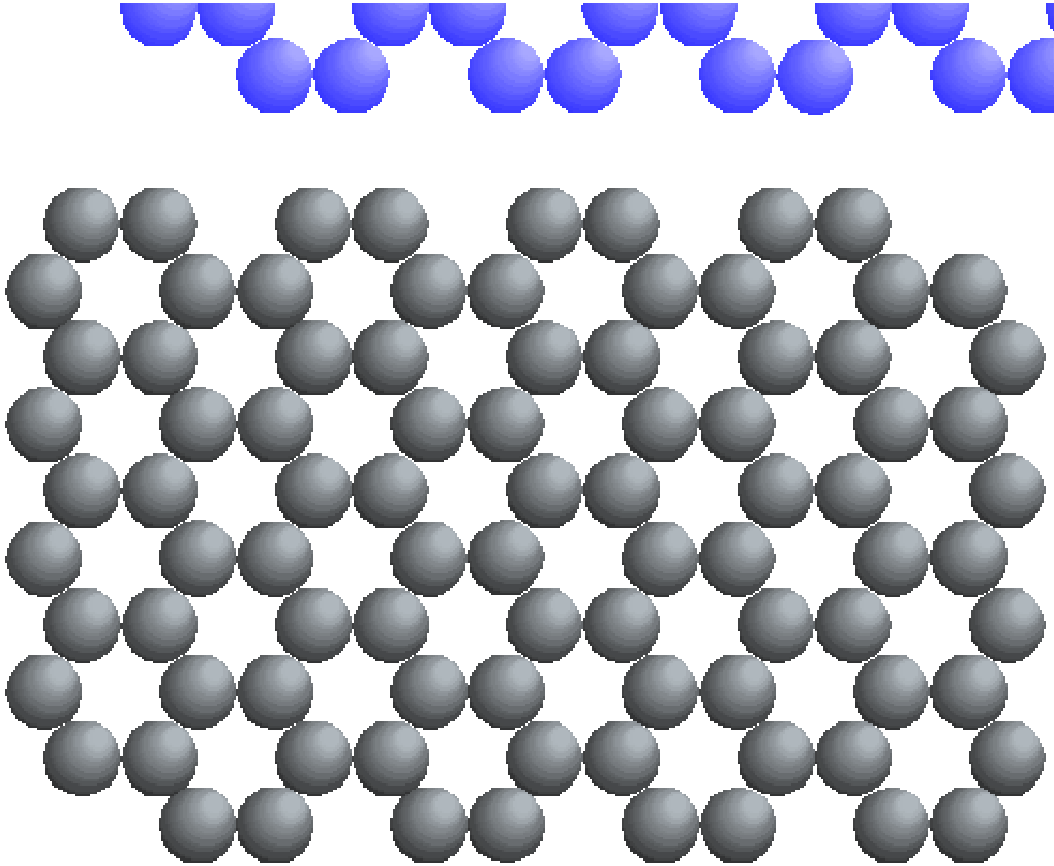


Graphene basal planes – a 2D “chicken-wire”



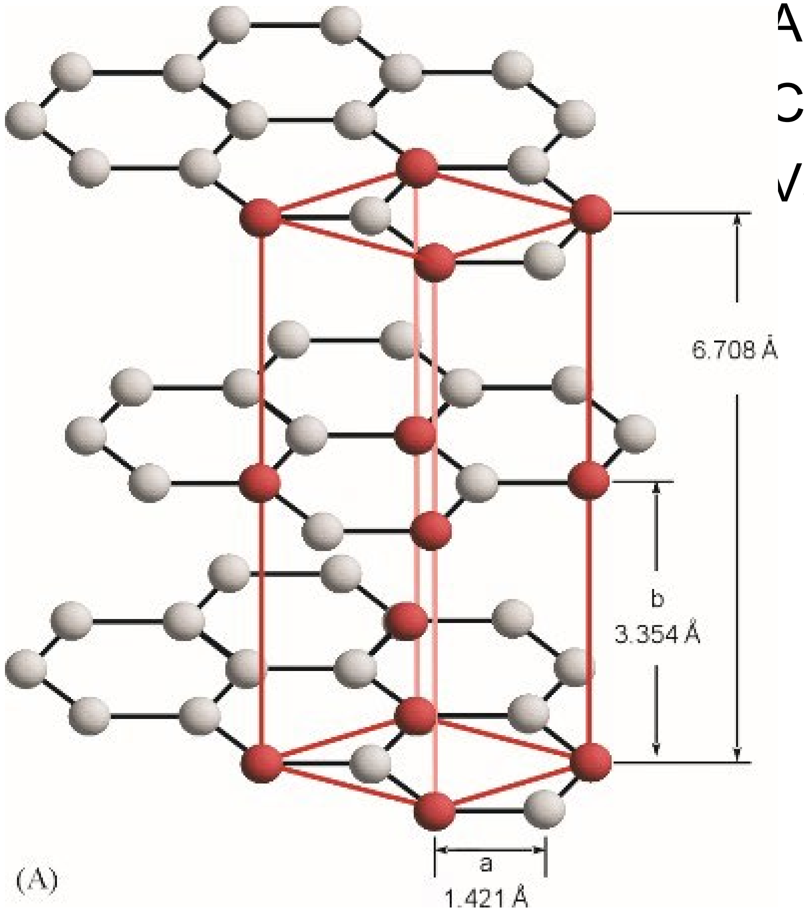
Stack up covalently bonded graphene planes

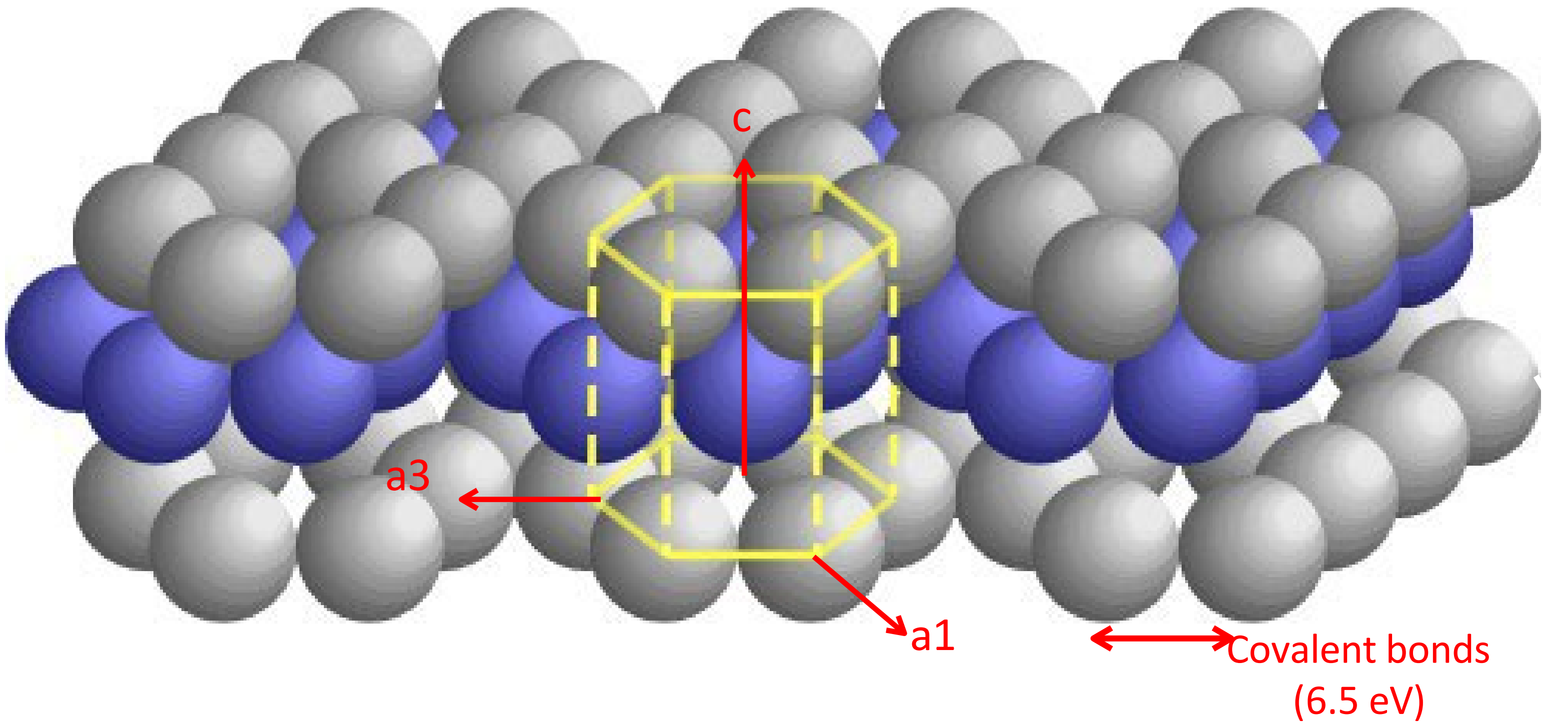
Basal planes stacked to form an … ABAB… stacking structure



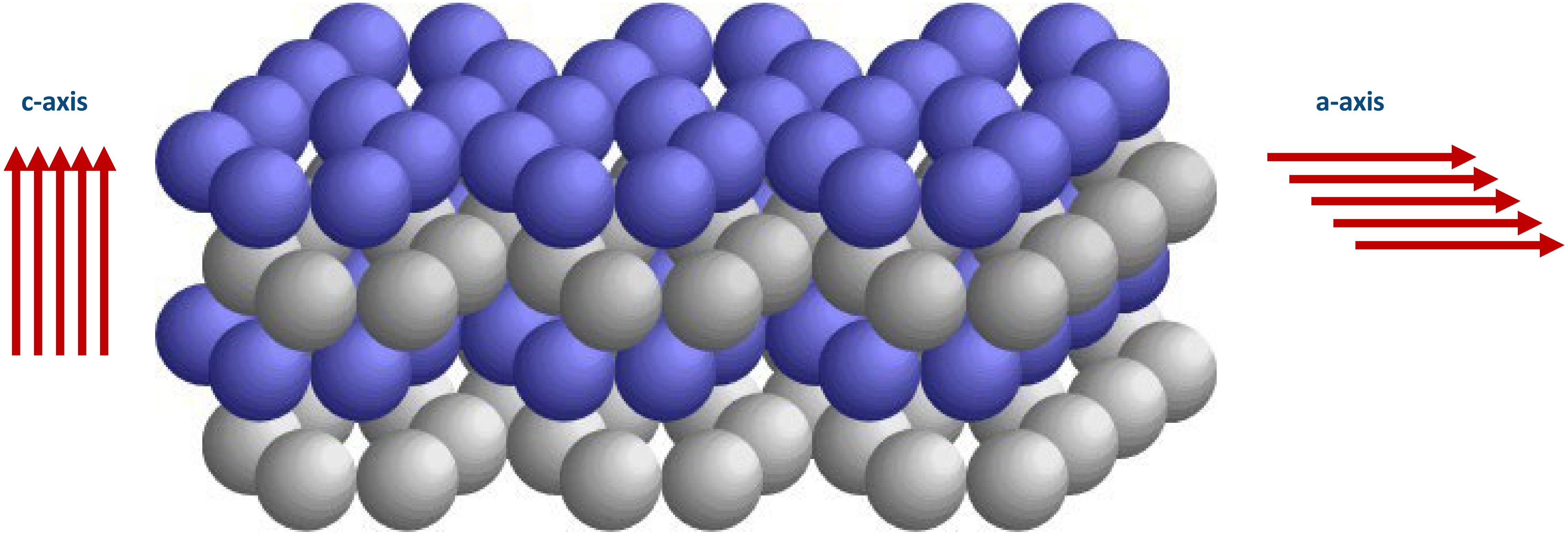
Graphite Crystal Structure

- Hexagonal close pack (HCP)
  - A-B-A-B stacking of basal planes
  - Covalent within basal planes,
  - Van der Waals (electronic) between basal plane



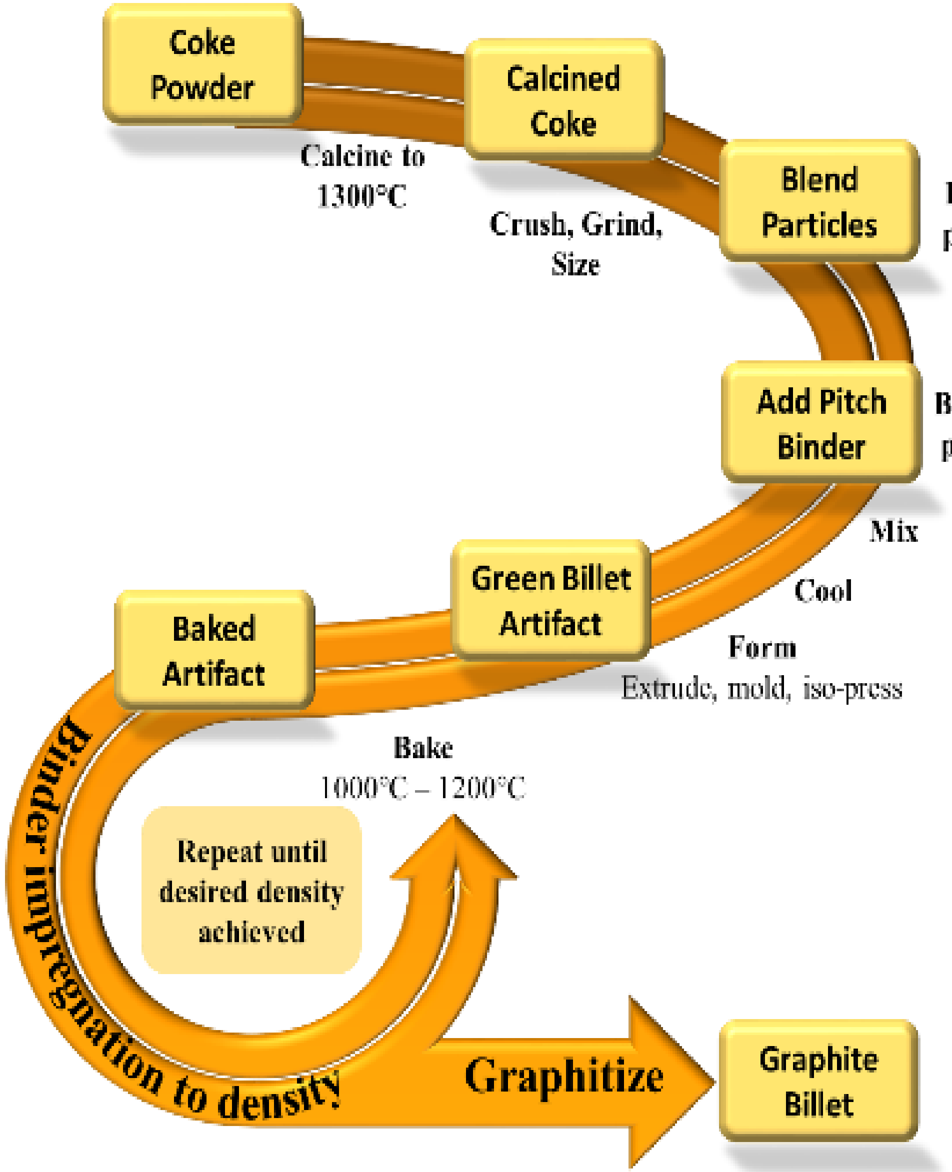


Completely anisotropic



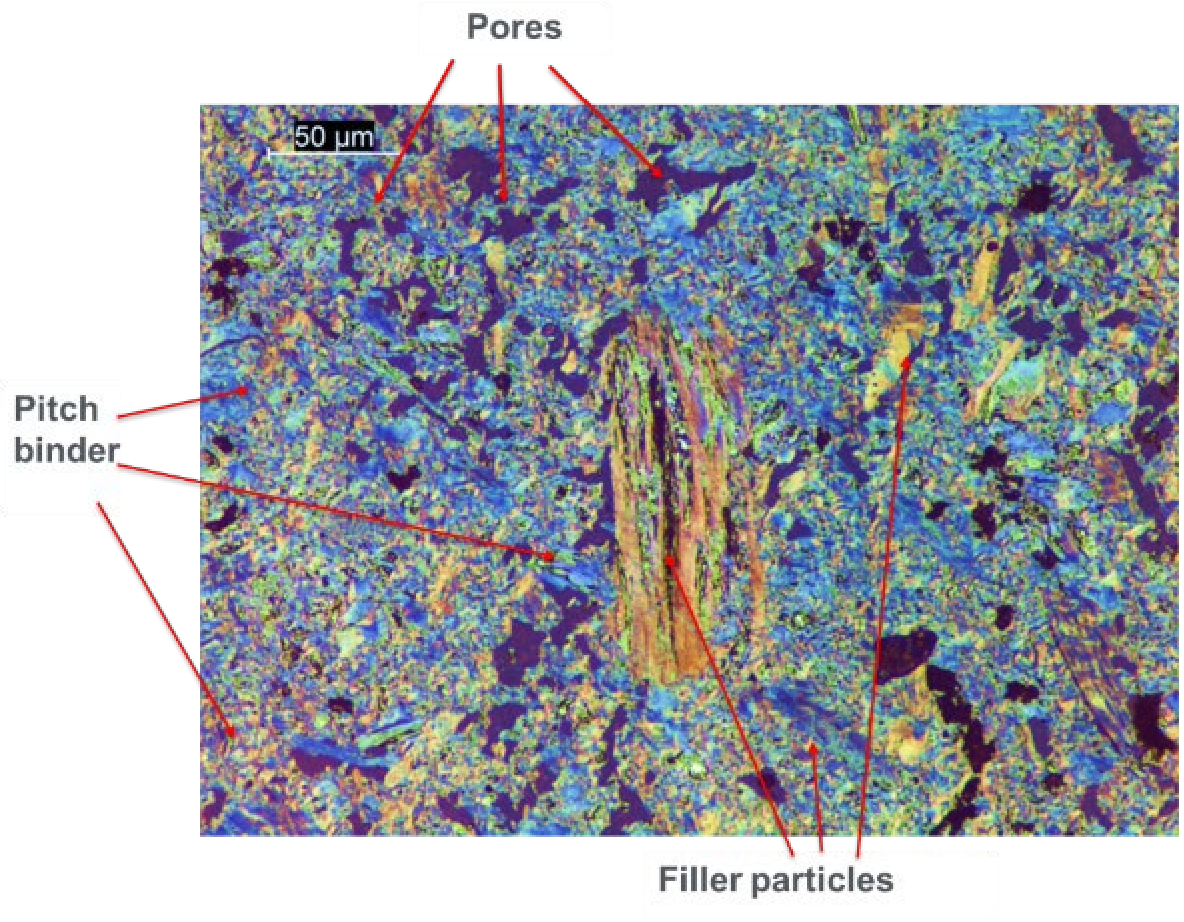
- Huge differences between c-axis and a-axis directions
  - c-axis CTE ~ 26 x 10^{-6} /°K a-axis CTE ~ 1 x 10^{-6} /°K
  - c-axis thermal conductivity ~ 6.8 W/m K : a-axis conductivity ~ 2000 W/m K

What do we do? Make a composite



- Long fabrication process
  - Filler particles: petroleum coke/pitch coke
    - Grind filler particles to desired size – grain size
  - Bind the particles: pitch-based liquid binder
    - Mix filler (grains) randomly in liquid binder
  - Form into final billet
    - Fabrication by extrusion, vibrationally molded, or isostatically molded
  - Baked green billet is heterogenous mix of filler particles bound by carbonaceous binder phase
    - Multiple pitch impregnations to increase density
  - Graphitize at >2200 °C
    - Most grades are heat treated to >3200 °C

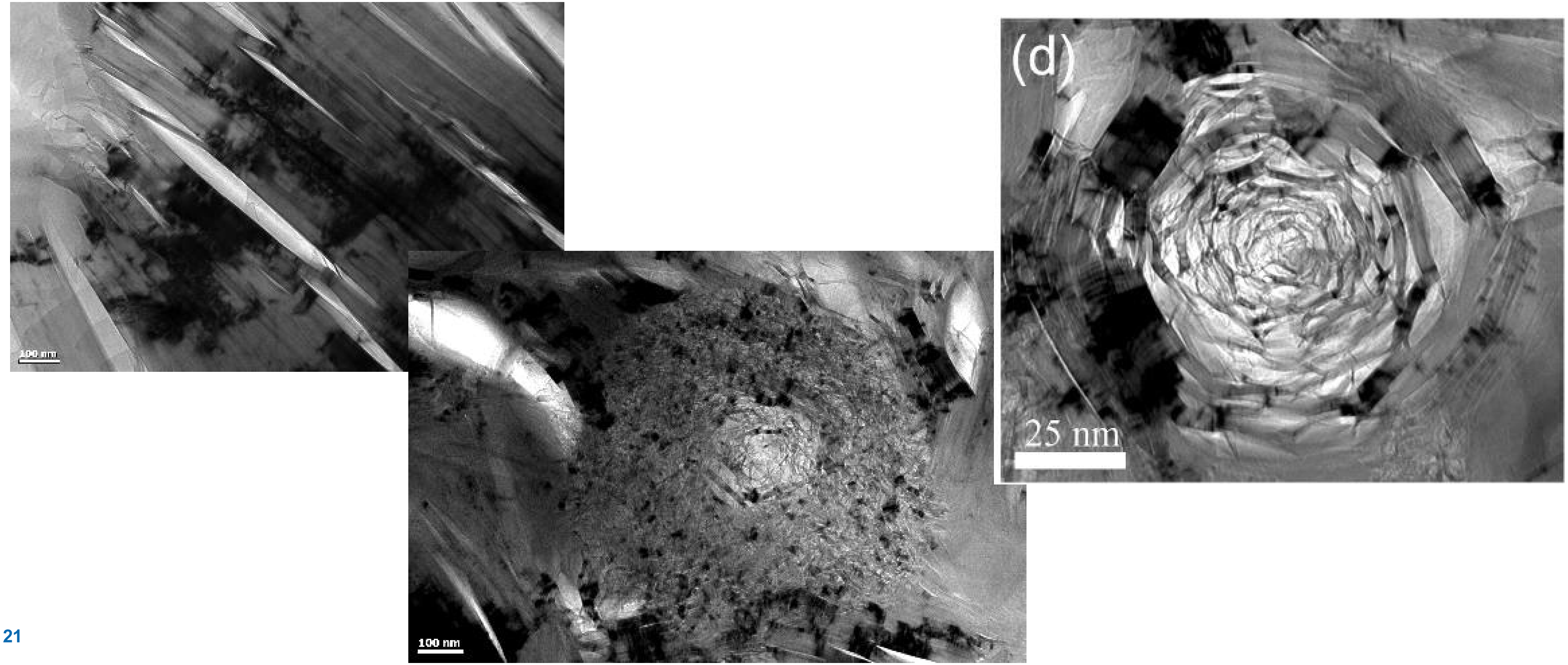
Microstructure of synthetic graphite



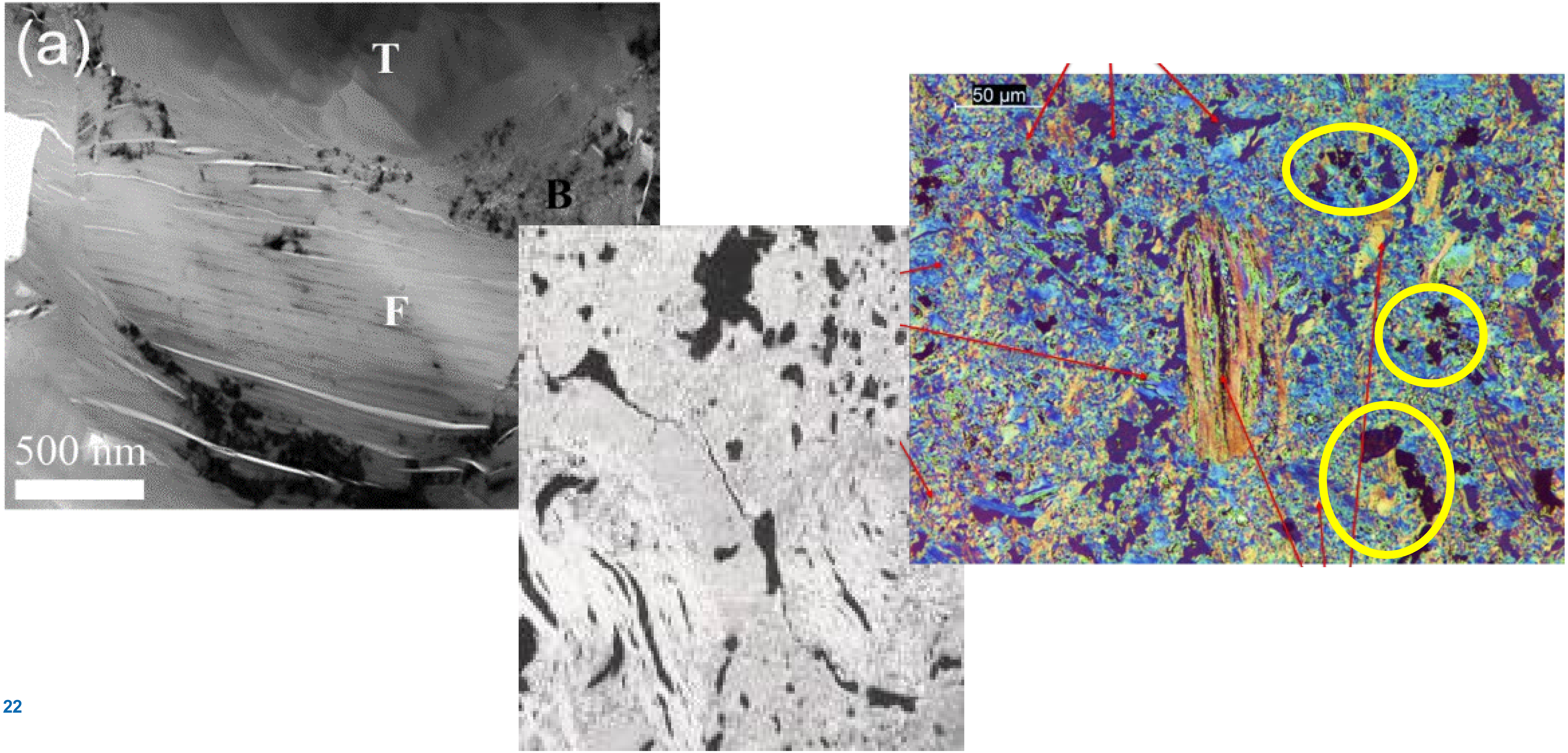
- Three phases in graphite microstructure
  - **Filler phase**
  - **Binder phase**
  - And Pore phase
- Nuclear graphite grades ~20% total porosity
  - Pores and pore structure define graphite behavior
  - Pore size range : nm to mm

- **Must. Have. Pores**
  - More accurately : must have accommodating porosity
  - Pores occur at all length-scales

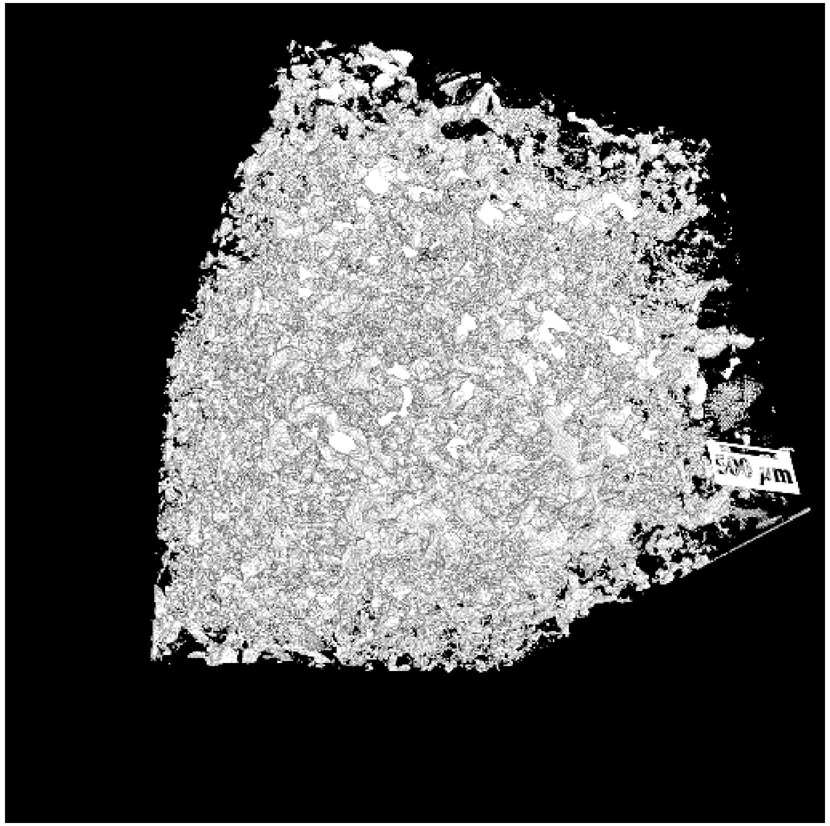
Crystal length-scale

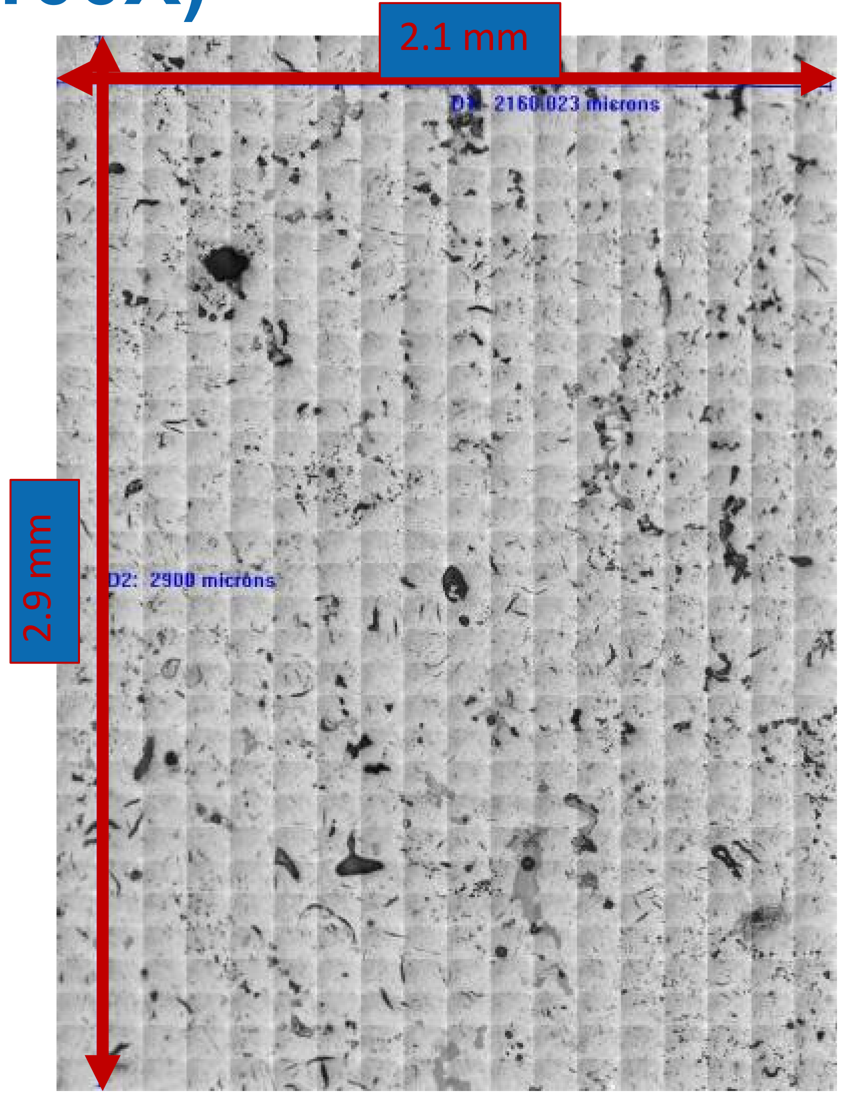


Grain-size length-scale



To the sample and component level (100X)





A “near” isotropic material

Material Properties

- Near-isotropic material response
- High thermal stability > 3000°C
  - *Well above any accident temperatures*
- High heat capacity (thermal sink)
- High thermal conductivity (better than metal)
- Density: 15% - 20% porosity
- Purified graphite: Low activation (Medium waste)
- Chemically inert (Molten salt)
- Neutron moderator (thermal designs)
- Easy machinability / cheap material
- High compressive / Low tensile strength
  - *Ceramic composites for tensile*
- Ceramic like material response
  - *Low fracture toughness (~ 1-2 MPa √m)*
  - *Quasi-brittle cracking*

Component Behavior

Decent irradiation response

- Smooth dimensional change
  - ○ *Life-limiting mechanism*
  - ○ *Multiple decades of safe operation*
  - ○ *And **even longer** at lower temperatures*
- Generally gets stronger with irradiation
- Isotropy stays relatively constant
- Thermal stability and capacity are unaffected

Oxidation and molten salt intrusion

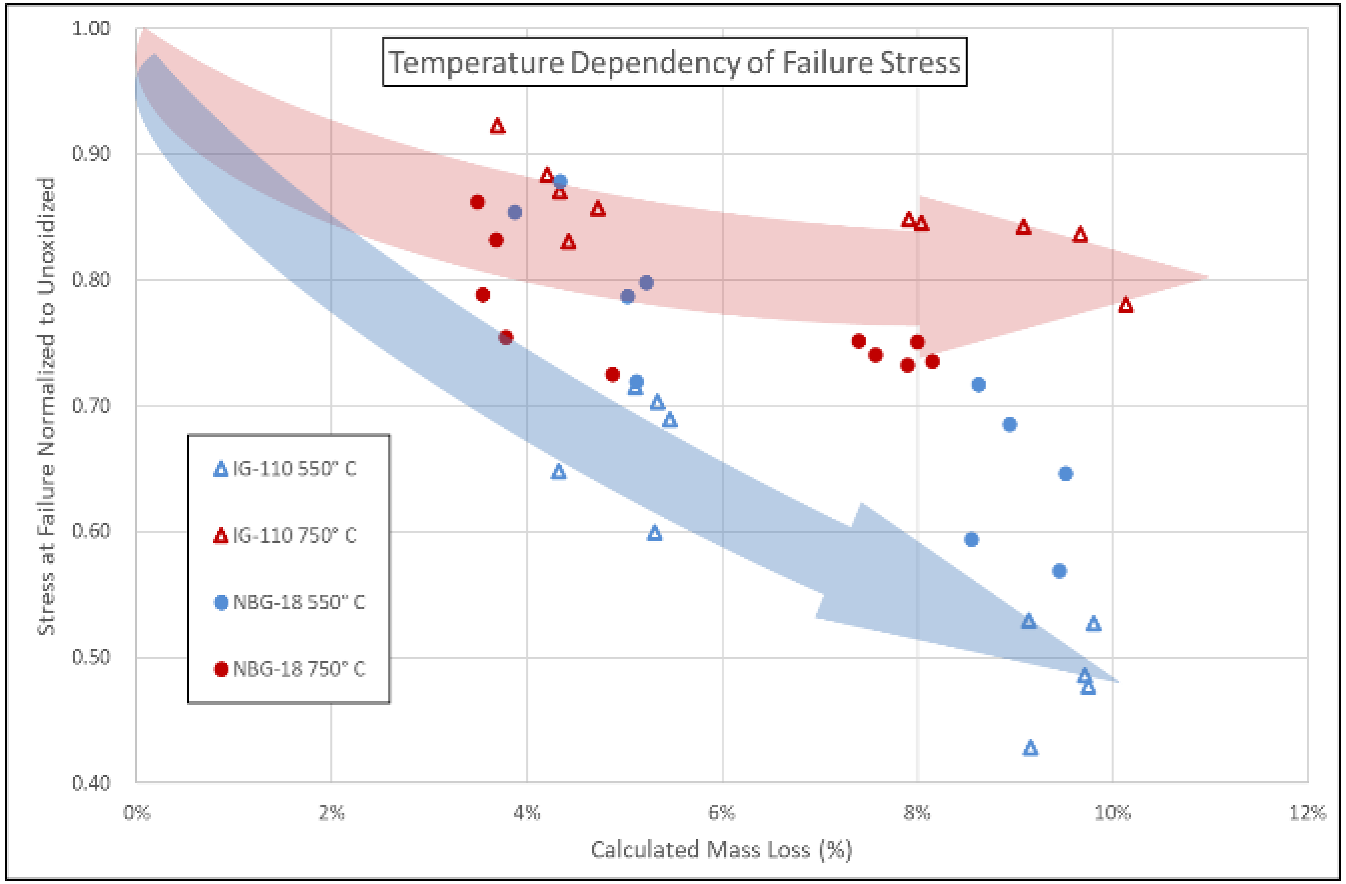
- Graphite does oxidize at all temperatures

  But it does not burn!
- Oxidation and molten salt behavior depends on pore structure

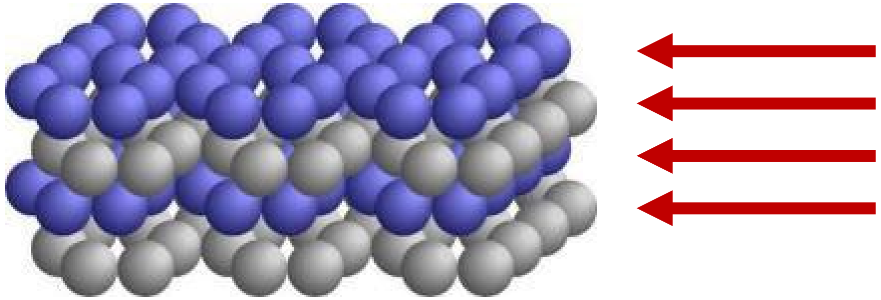
There is **no** “nuclear graphite” fabrication standard
All grades are proprietary

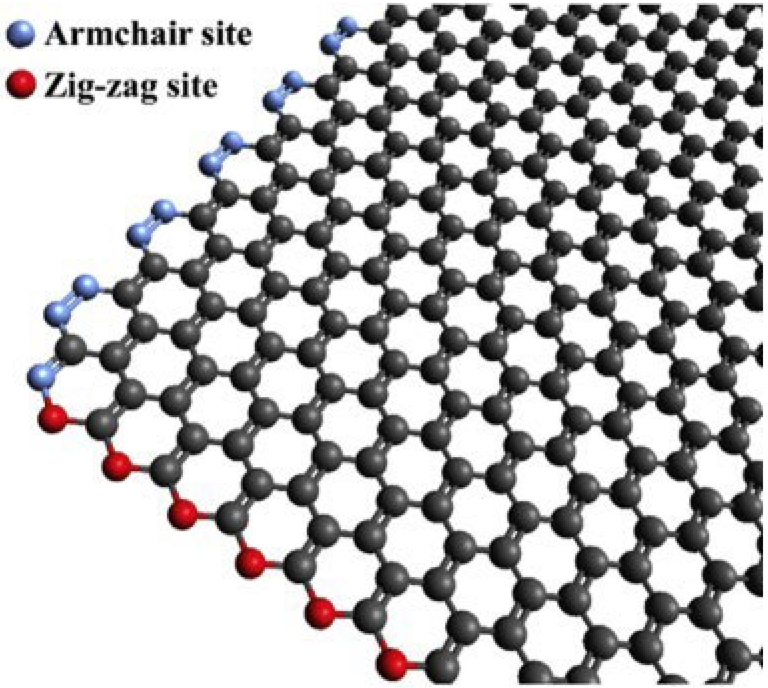
Degradation: Oxidation Behavior

- Internal Oxidation:
  - Air/oxygen into the pore structure
  - Oxidation rate is temperature dependent
    - *Oxidation occurs at all temperatures*
- Oxidation Effects
  - Internal oxidation has larger effect on residual strength
  - High temperature oxidation only attacks outside of component



Oxidation mechanisms



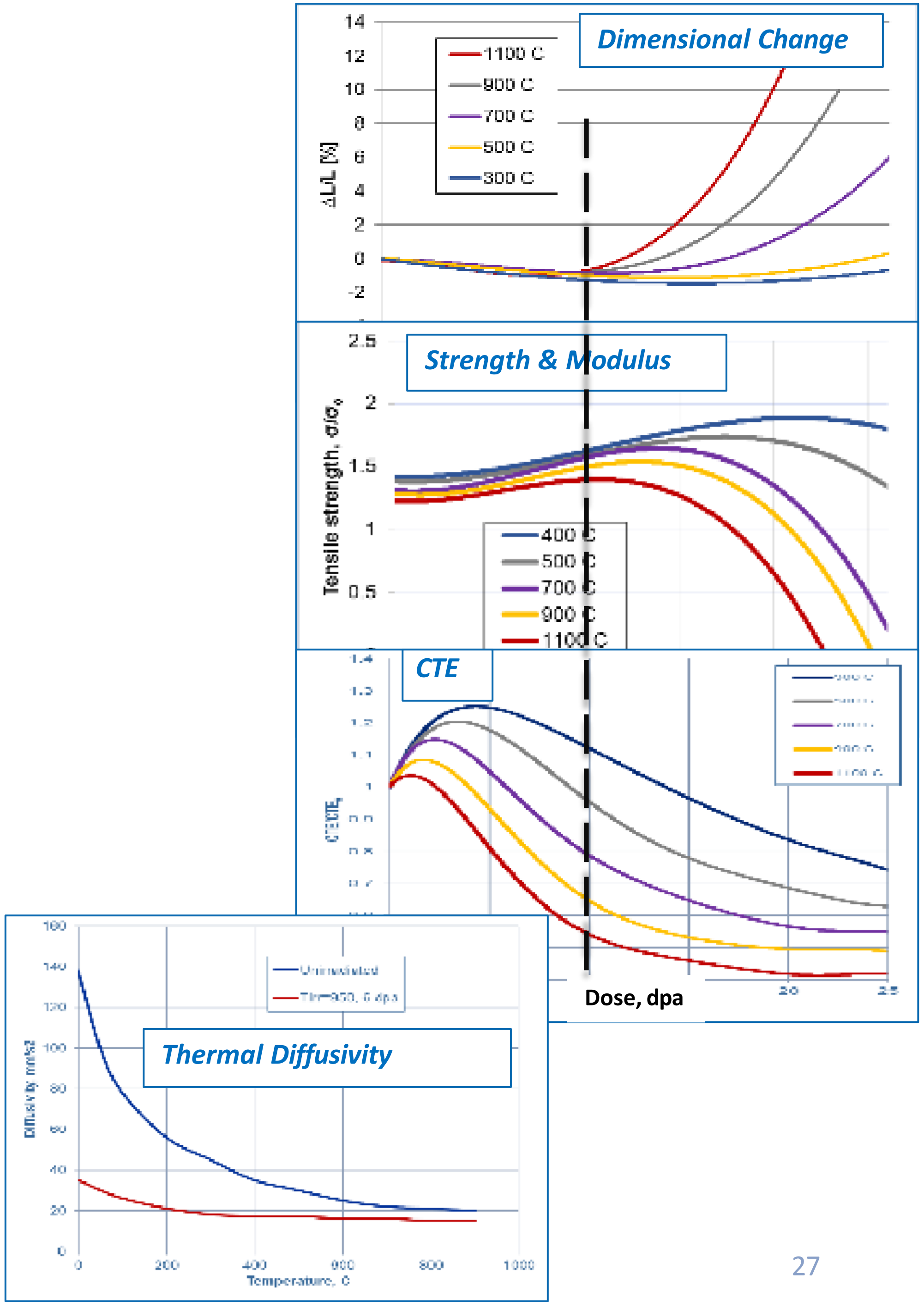


- **Only on the Edges**
  - **Oxygen chemical reaction occurs only on the edge of basal planes**
  - **Covalent bonds within planes are too energetic**
- **RSA sites**
  - **Reactive Surface Area**
  - **Most chemically reactive**
    - *Fission product and other impurities end up here*
- **Can not sustain reaction**
  - **No fires!**

Irradiation Behavior

Significant changes occur during normal operation:

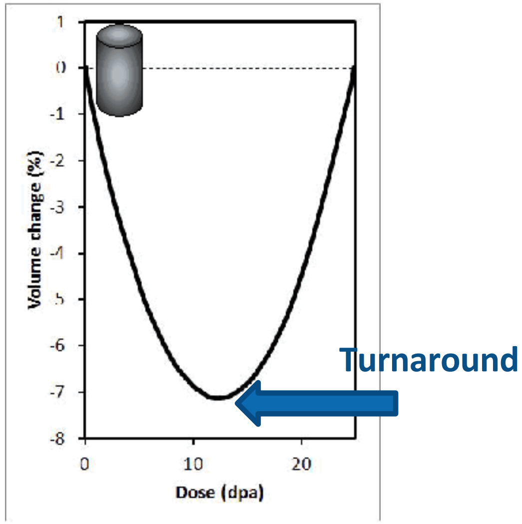
- Density - Densification
  - *Graphite gets denser with irradiation until Turnaround dose*
  - *After Turnaround density decreases (volumetric expansion)*
  - *Formation of microcracks (molten salt consideration)*
- Dimensional change
  - *Turnaround dose is key parameter*
  - *Highly temperature dependent*
- Strength and modulus
  - *Graphite gets stronger with irradiation …*
  - *Until Turnaround dose is achieved. It then decreases*
- Coefficient of thermal expansion
  - *Initial increase but then reduces before Turnaround*
  - *CTE is why properties are so temperature dependent*
- Thermal diffusivity
  - *Decreases immediately to ~30% of unirradiated values*
  - *At high temperatures it is same as unirradiated conductivity*



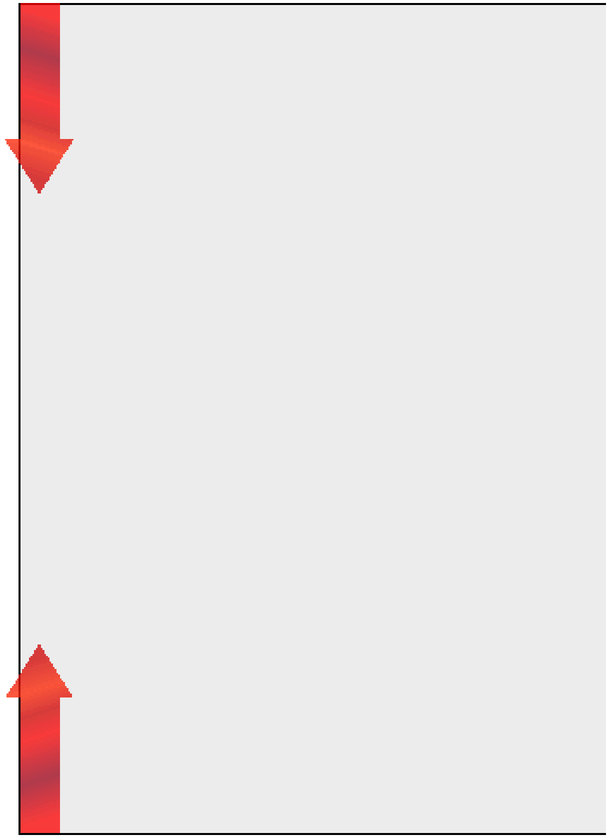
Irr. Dimensional change → life limiting mechanism

Internal stresses for crack formation develop from irradiation dimensional change

- Densification to volumetric expansion
  - *As dose increases, dimensional change creates internal stresses*
  - *Temperature accelerates dimensional change*
  - *Internal stress buildup at interface of dimensional change*
  - *Changes after turnaround most limiting*

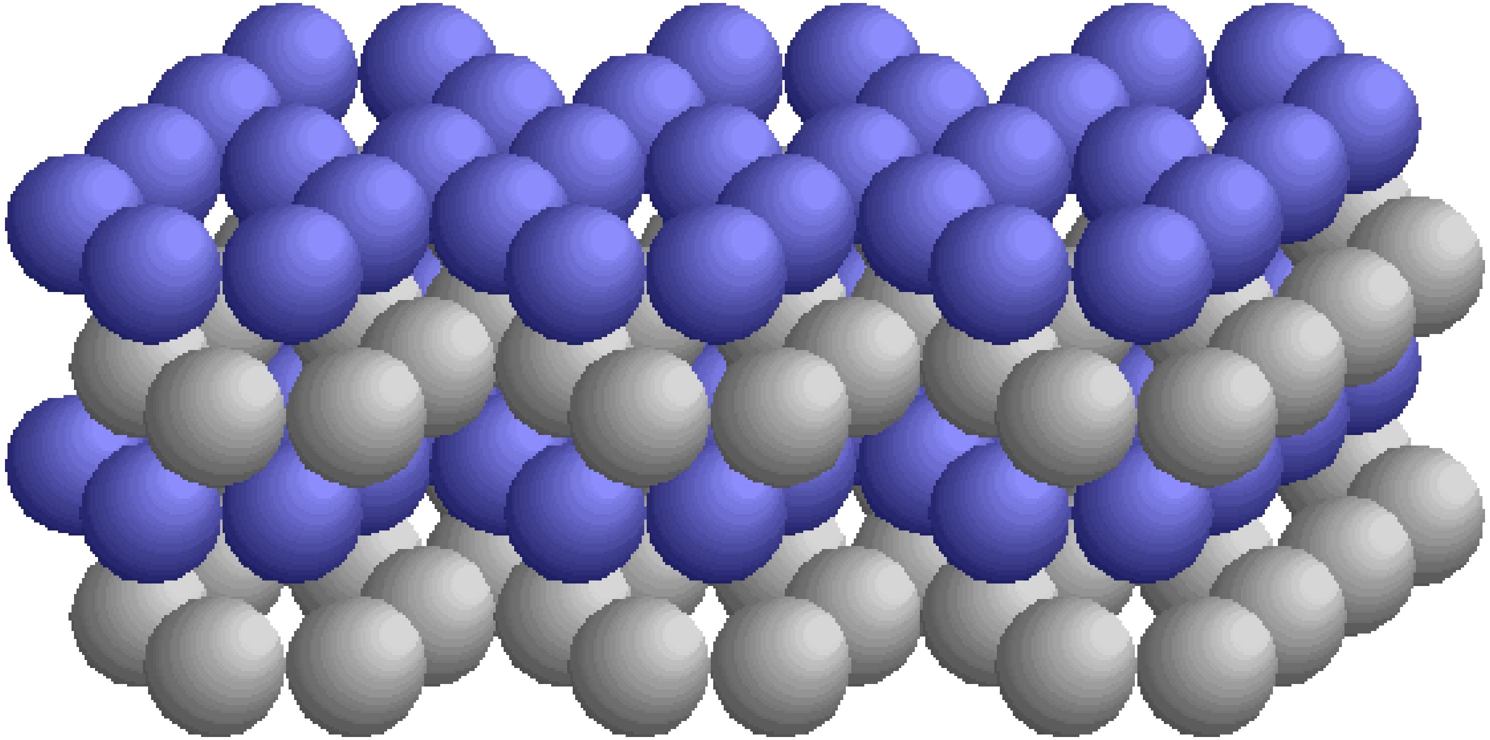


G. Haag,” ***Properties of ATR-2E Graphite and Property Changes due to Fast Neutron Irradiation”, Juel-4183, 2005***



Mechanisms underlying irradiation damage

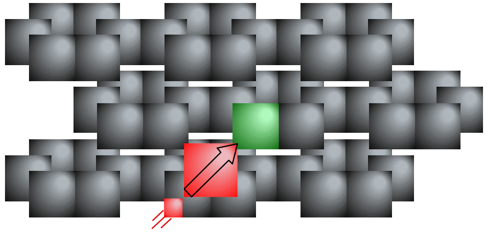
- Ballistic event physically displaces atoms for lattice position
- Sub-plane formation, vacancy clusters

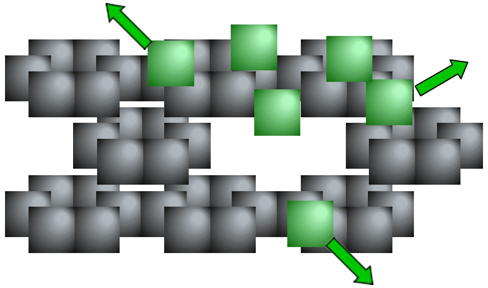


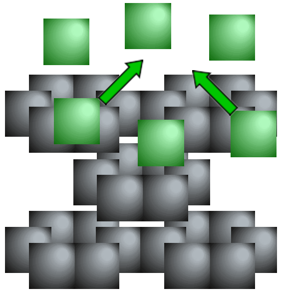
Model describing irradiation damage

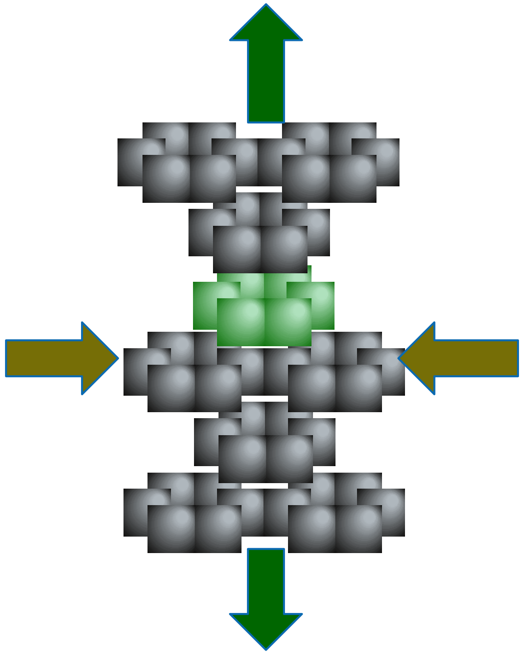
- Ballistic event physically displaces atoms for lattice position
- Sub-plane formation, vacancy clusters

- Sub-plane formation between basal planes
- Vacancy cluster/loop collapse

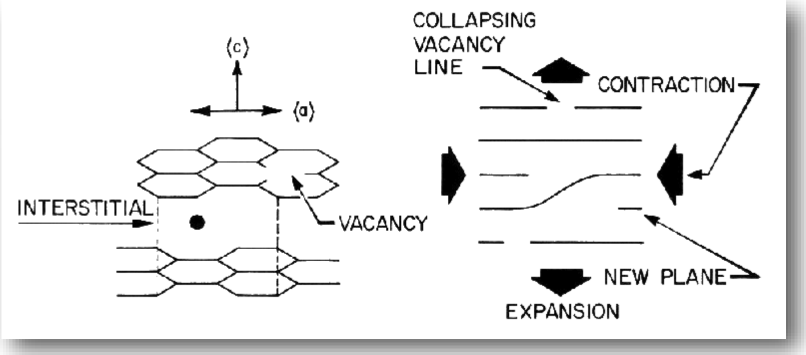




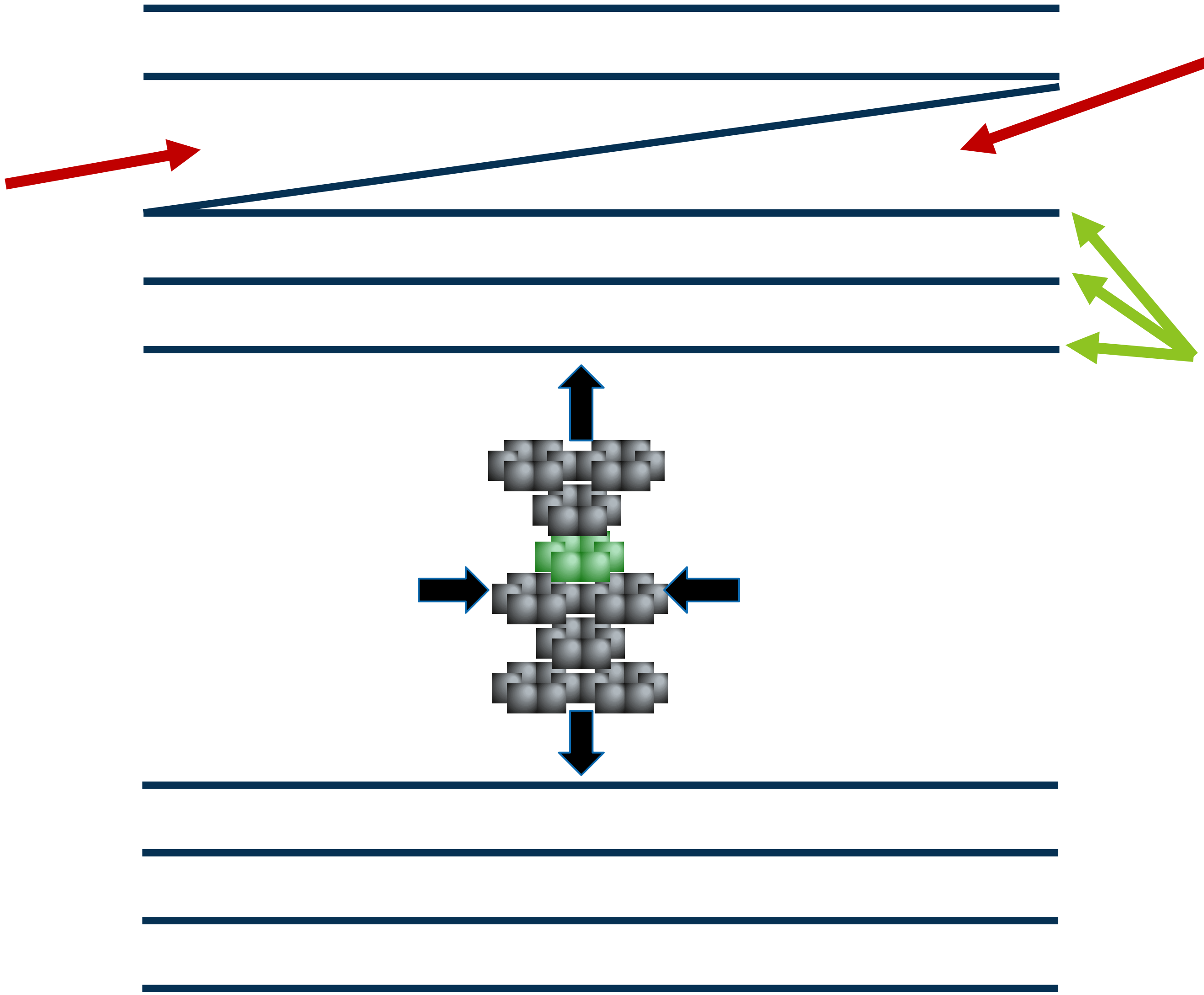




- **Crystallites shrink parallel to basal planes**
- **They expand perpendicular to planes**



Model describing irradiation dimensional change

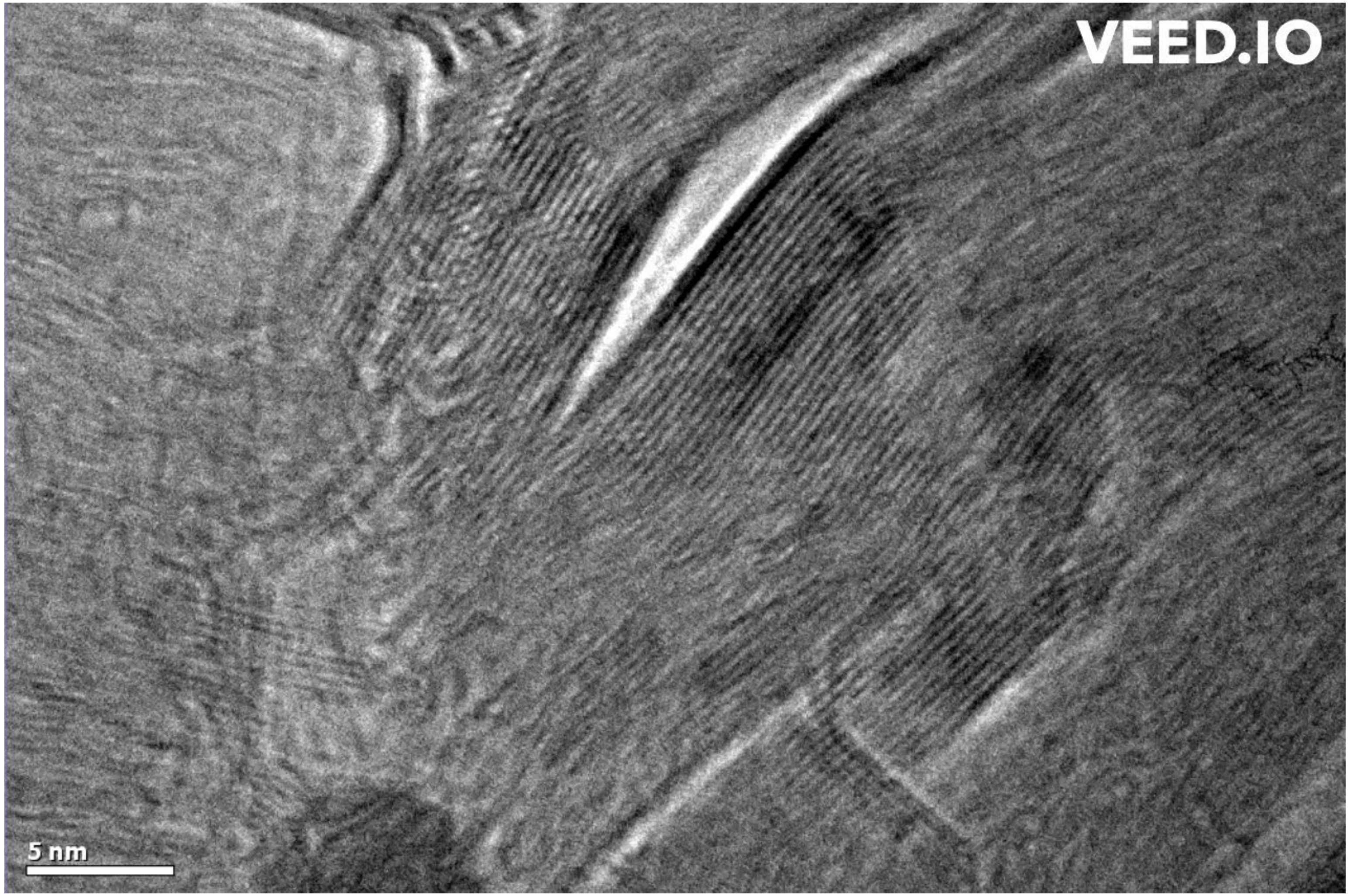


Mrozowski cracks
(Accommodating
porosity)

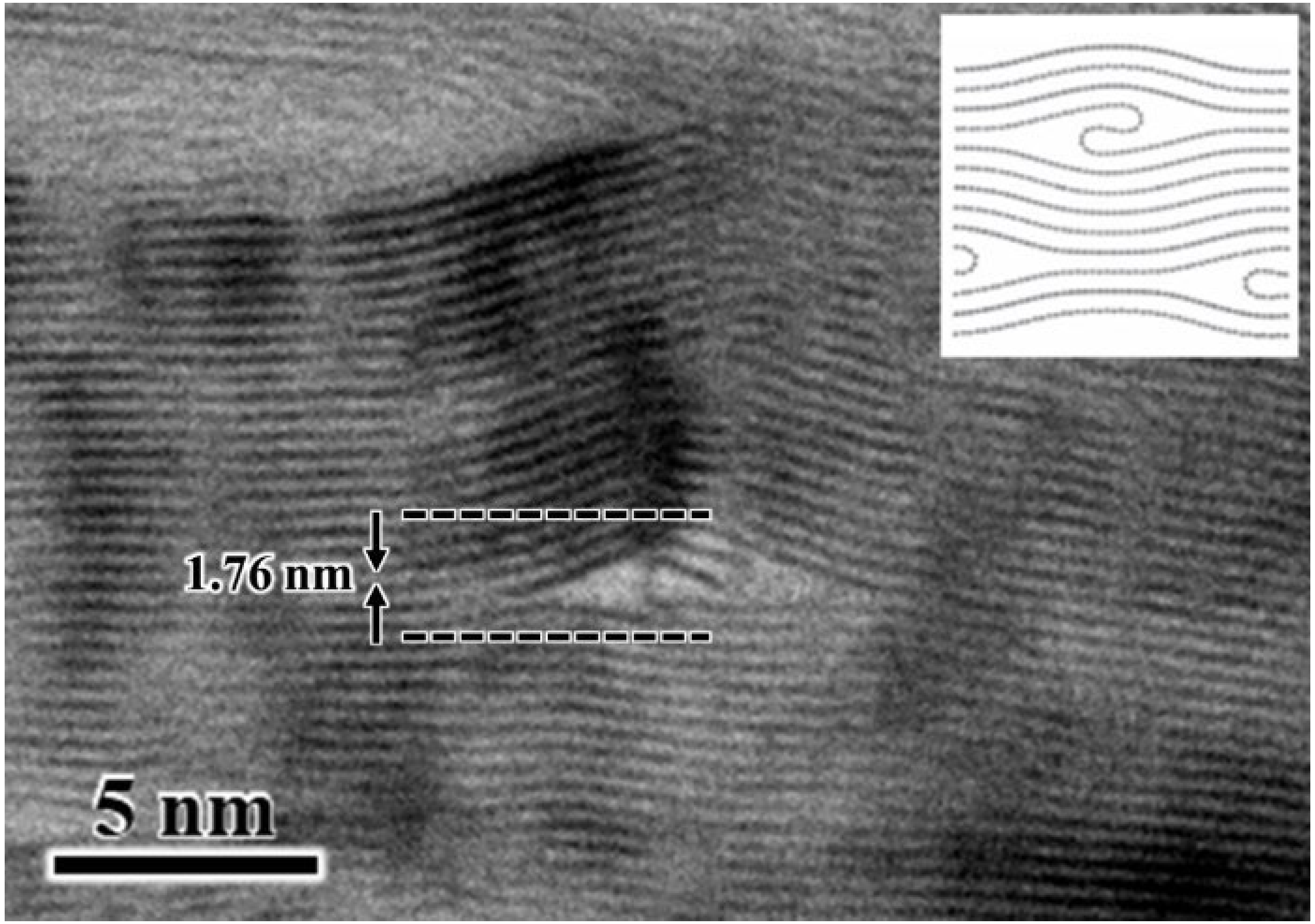
Mrozowski cracks
(Accommodating
porosity)

Basal planes
(Graphene planes)

Observations: Crack closure



Observations: c-axis expansion



- Buckle, ruck & tuck
  - Buckling of basal planes.
  - Not sub-plane formation.
- New techniques offer new data for mechanisms
  - Defect formations
  - Dimensional change
  - Property changes
- But we must understand limitations of new techniques

Graphite does not, can not burn

Graphite does not, can not burn

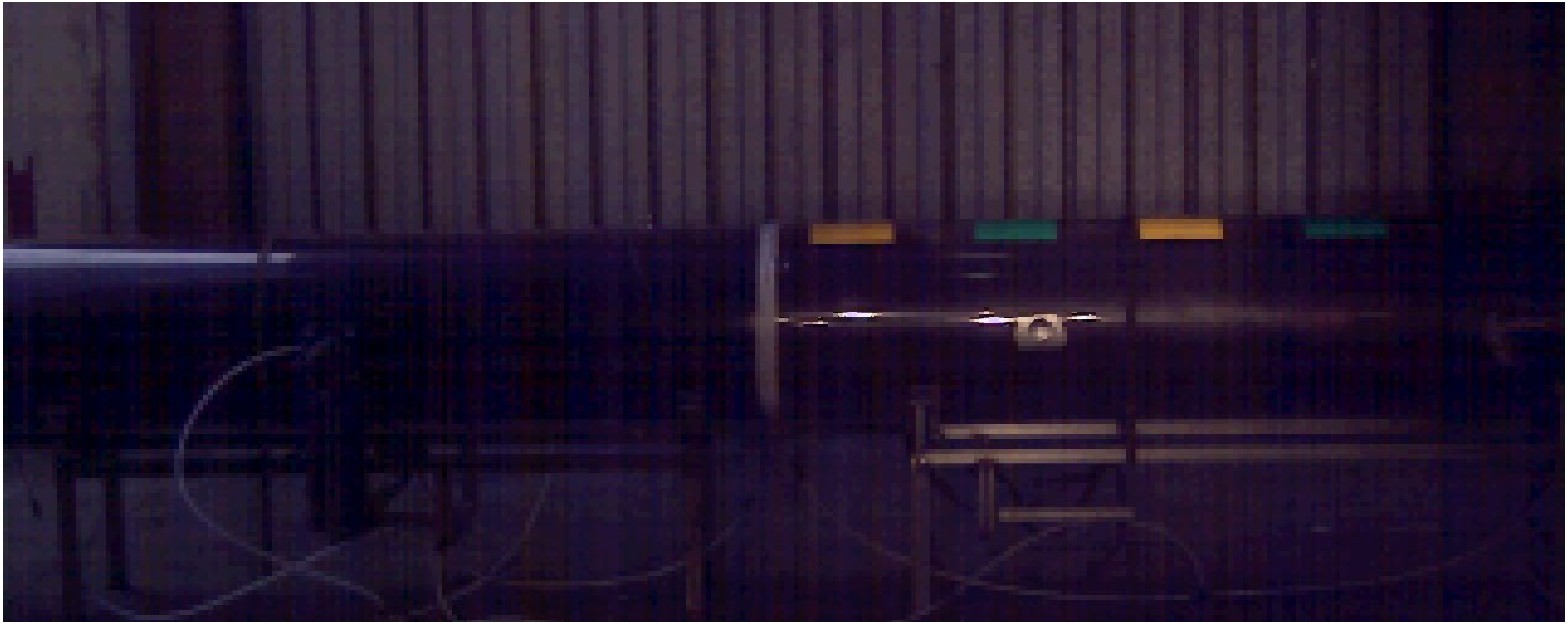
It oxidizes but can not sustain the reaction

- ***Only outer edges of crystals can react***
  - *Analogy: burning a thick paper book*
- ***Reactive Surface Area (RSA) sites on the edges***

Graphite "Fires" and "Explosions"

Maize Dust

Graphite “Fires” and “Explosions”



Graphite Dust

(Potential) Molten Salt Issues

Dr. Nidia Gallego on 24 May 2023

Large molten salt tests are being initiated

- Salt impregnation into graphite pores
  - Physical damage/cracks
  - “Hot spots” from fueled molten salt
- Wear/abrasion/erosion
  - Molten salt has higher density than graphite
  - Liquid flow over soft graphite has potential
- Chemical coupling with metallic systems
  - Graphite – MS is inert
    - *Fluorination questions remain.*
  - There are questions when a metallic component is added to the MS system

Before immersion in FLiNaK

Salt residue

After immersion in FLiNaK

In summary

- **Graphite is an important material for future GEN IV reactor designs**
  - ***Carbon-based materials***
  - ***Ceramic composites***
  - ***Ceramics***

- **Much work still remains in characterizing nuclear graphite**
  - ***Chronic oxidation behavior***
  - ***Irradiation behavior***
  - ***Molten salt interactions***
  - ***New coolant and fuel systems for advanced reactor concepts***

INL

Idaho National Laboratory

Upcoming Webinars

Date	Title	Presenter
24 May 2023	Graphite-Molten Salt Interactions	Dr. Nidia Gallego, Oak Ridge National Laboratory, USA
21 June 2023	Panel Session: International Knowledge Management and Preservation of SFR	Joel Guidez, CEA (retired), France; Hiroki Hayafune, JAEA, Japan; Ron Omberg, PNNL, USA; Cal Doucette, ARC Energy, Canada; and Patrick Alexander, Terra Power, USA
26 July 2023	Off-Gas Xenon Detection and Management in Support of MSRs	Dr. Hunter Andrews, ORNL Dr. Praveen Thallapally, PNNL, USA

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