A Review of Stored Energy Release of Irradiated Graphite

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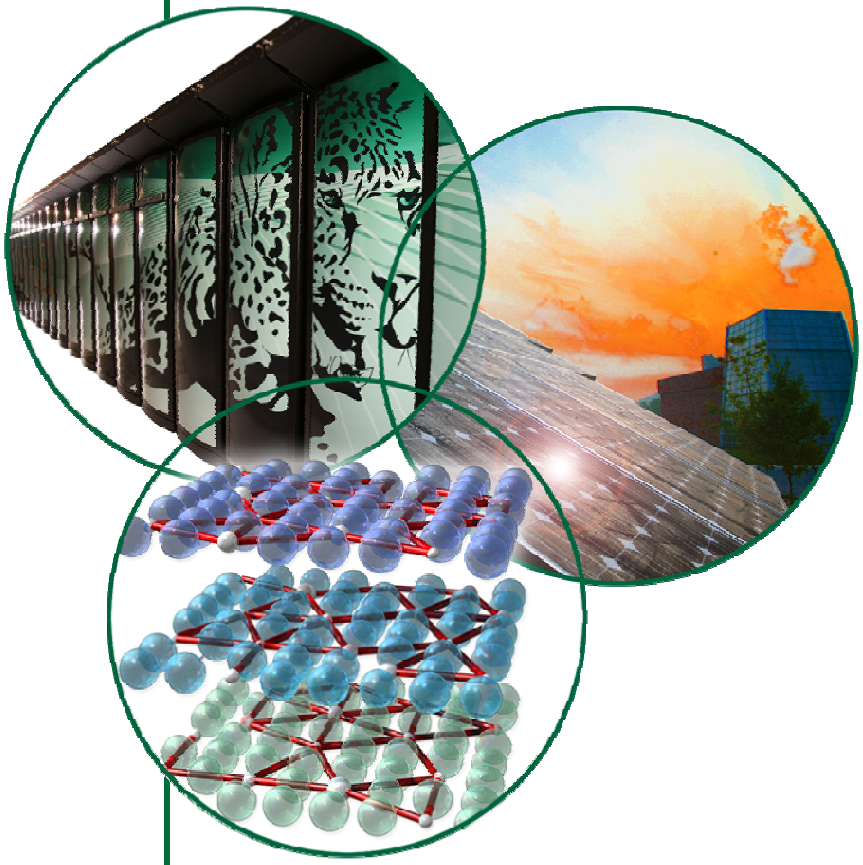

U.S. DEPARTMENT OF ENERGY

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Outline

- **Background**
  - Problem Definition
  - Irradiation-induced defects in graphite / Wigner energy
- **Prior work**
- **Objective of this study**
- **Relevant variables**
  - Cumulative dose and temperature
- **Experimental plan**
  - Current graphite inventory at ORNL
  - Study method

Role of Graphite in a Nuclear Reactor

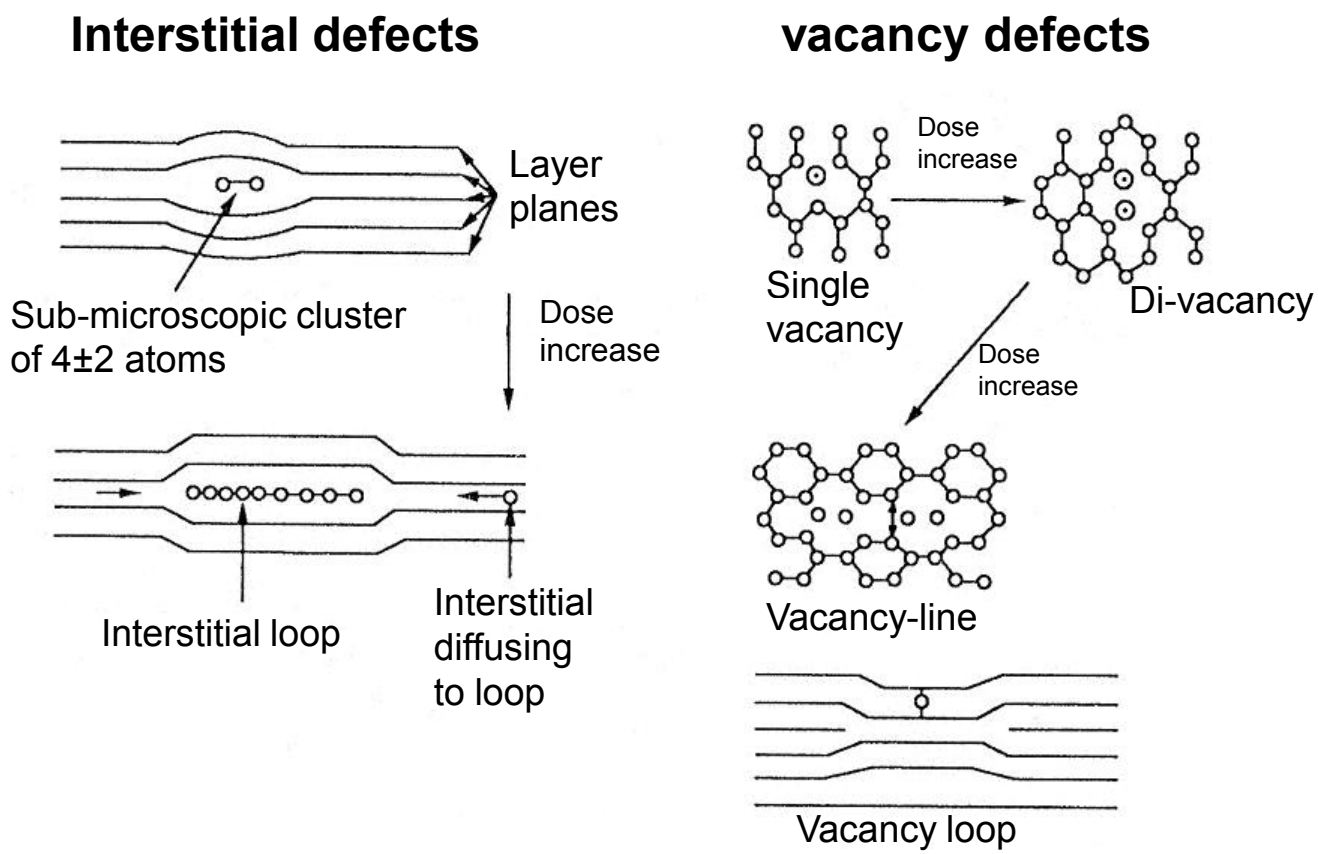
- Neutron moderator (carbon & graphite)
  - Thermalize fast neutrons to sufficiently low energies that they can efficiently fission $_{92}U^{235}$
- Neutron reflector – reflects neutrons to the active core
- Nuclear grade graphite has a low neutron capture cross section
- Withstand high temperature reactor operation

Role of Graphite in NGNP HTGRs

- Graphite is the reactor core structural material
- HTGR cores are constructed from graphite blocks
- In prismatic design the graphite fuel elements retain the nuclear fuel
- In a pebble bed the graphite reflector structure retains the fuel pebbles
- The graphite reflector structure contains vertical channels for reactivity control
- Reactivity control channels are also contained in prismatic graphite fuel elements

Wigner (stored) energy

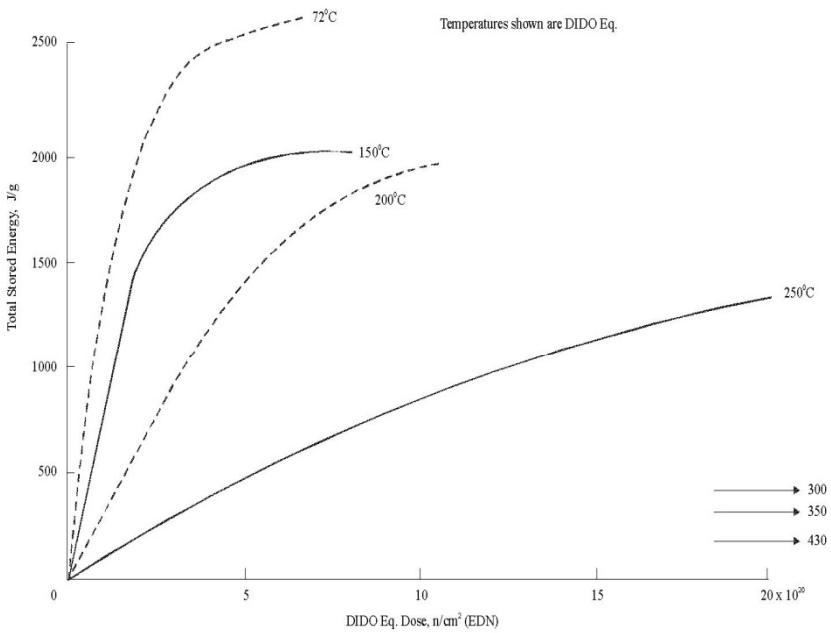
- Wigner (stored) energy accumulates in graphite under neutron irradiation because atoms are displaced from their normal positions into configurations of higher potential energy



Schematic of lattice defects introduced by radiation in graphite
(G.B. Engle and B.T.Kelly, J.Nucl. Mater., 122&123(1984) 122-129.)

Stored energy as a function of irradiation temperature

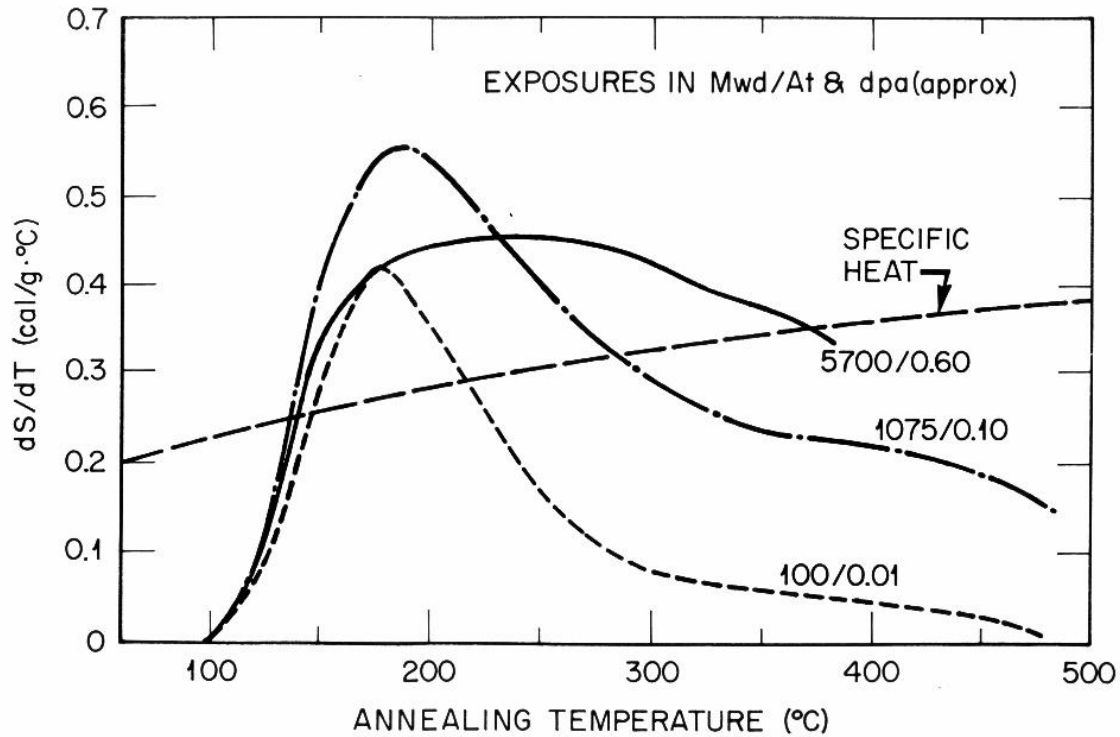
- In graphite irradiated with fast neutrons at **ambient temperature** relatively large amounts of energy can be stored as lattice defects.
- Values of up to 2700 $J.g^{-1}$ have been measured in graphite which, if released as heat under adiabatic conditions, could raise its temperature by ~ 1500°C.
- This behavior could be a potential technical safety issue requiring further study.



Accumulation of total stored energy in graphite at various irradiation temperatures

Source: IAEA-TECDOC-1154

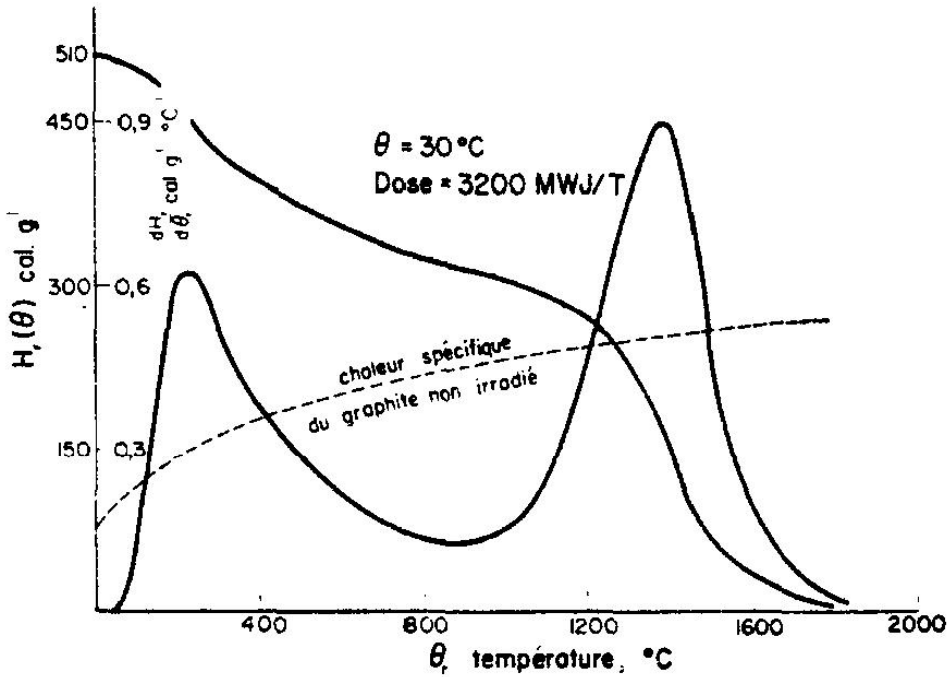
Stored energy release of graphite irradiated a low temperature



Stored energy release curves for CSF graphite irradiated at ~30C in the Hanford K reactor cooled test hole.

Source: Burchell T, Carbon Materials for Advanced Technologies, Chpt. 13 (1999) p. 429

High Temperature Stored Energy Release

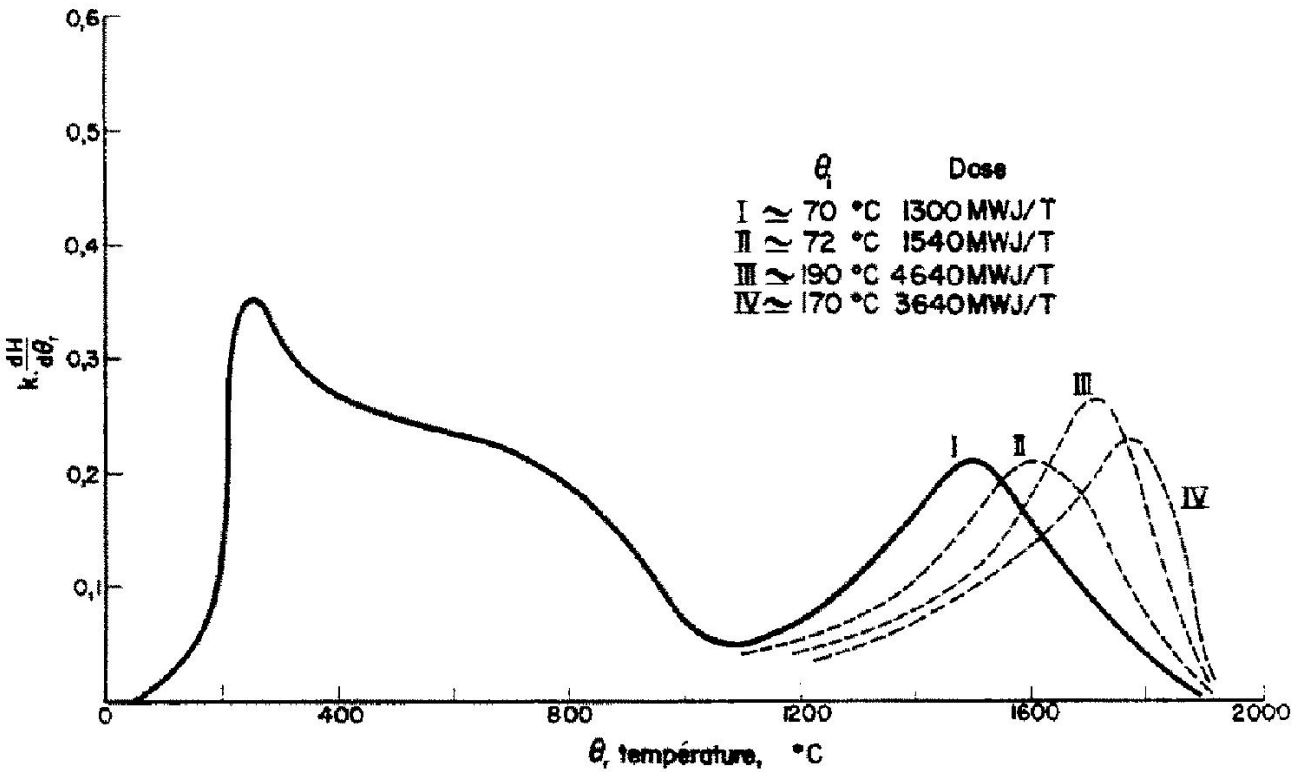


Stored energy release behavior for graphite irradiated at 30 °C compared with non-irradiated graphite C_p behavior

- A second release peak is observed at ~1400 °C in graphite irradiated at **low** temperatures
- Associated with the annealing of small interstitial clusters
- Immobile vacancies can coalesce at high temperature
- Release rates > C_p NOT seen in graphite irradiated at higher temperatures (>300°C)

Source: Rappeneau et al, CARBON 9 (1966) 115-124

High Temperature Stored Energy Release

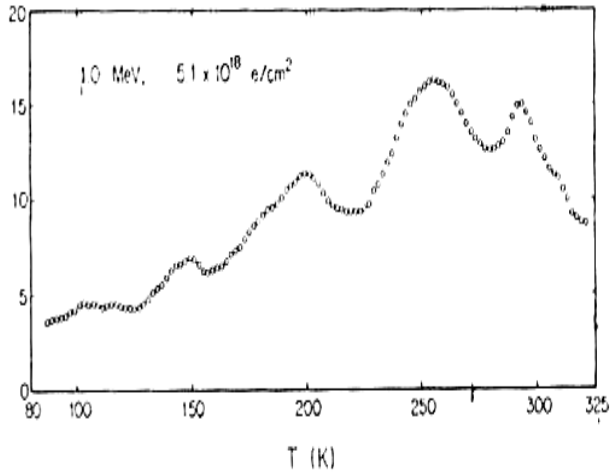


- High temperature release is due to a separate mechanism (confirmatory experiments planned)
- High temperature release rate does NOT exceed C_p

Source: Rappeneau et al, CARBON 9 (1966) 115-124

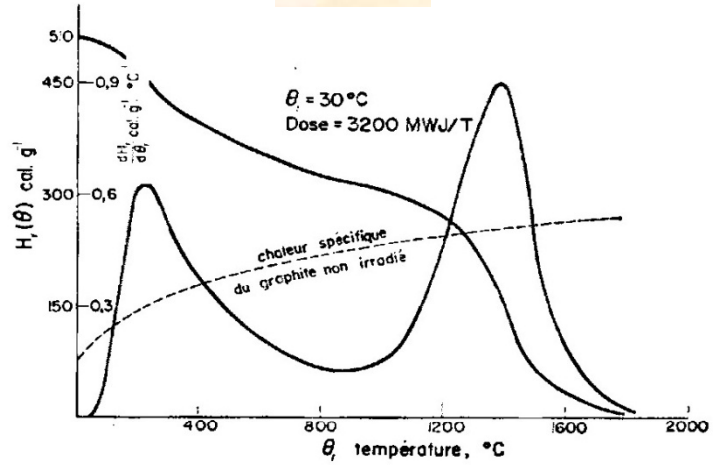
What is it known about Wigner energy release?

Low T

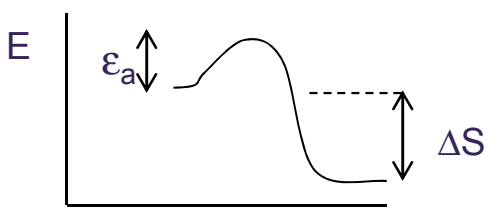


Iwata and Nihira (1976) *Jap. J. Appl. Phys.* **15** 575

High T



Rappenau *et al.* (1966) *Carbon* **4** 115



- Release curves dependent on irrad. T, irrad. species, dose, flux, previous history
- Peaks associated with particular defect processes: barrier energies
- Amount of heat released is ΔE between metastable and (more) stable state
- Peak broadening – process overlap and changes in local environment

Objectives

- Evaluate current knowledge of the effects of energy stored on previously irradiated nuclear graphite on potential accident scenario, such as loss of coolant;
- Assess this knowledge for the expected operational conditions of NGNP HTGR;
- Develop an experimental plan to study the stored energy, including an examination of available options with previously irradiated graphite, such as H-451, which may be at ORNL; conduct these experiments; and
- Analyze the data and provide new experimental information on potential for stored energy release on graphite irradiated at higher temperatures than previous studies
- Assess implications to NGNP HTGR operation.

NGNP assumptions

- Either a prismatic core or pebble bed core
  - Graphite – (compressive applications)
  - Ceramic composites – (tensile applications)
- No decision of specific graphite type
- 750 °C (to start with) - 900-950 °C (eventually)
- 400-600 MW_t
- He inert gas coolant (limited impurities)

- Funding Partnership with private sector
- Vendors = Westinghouse (PB), GA, & AREVA

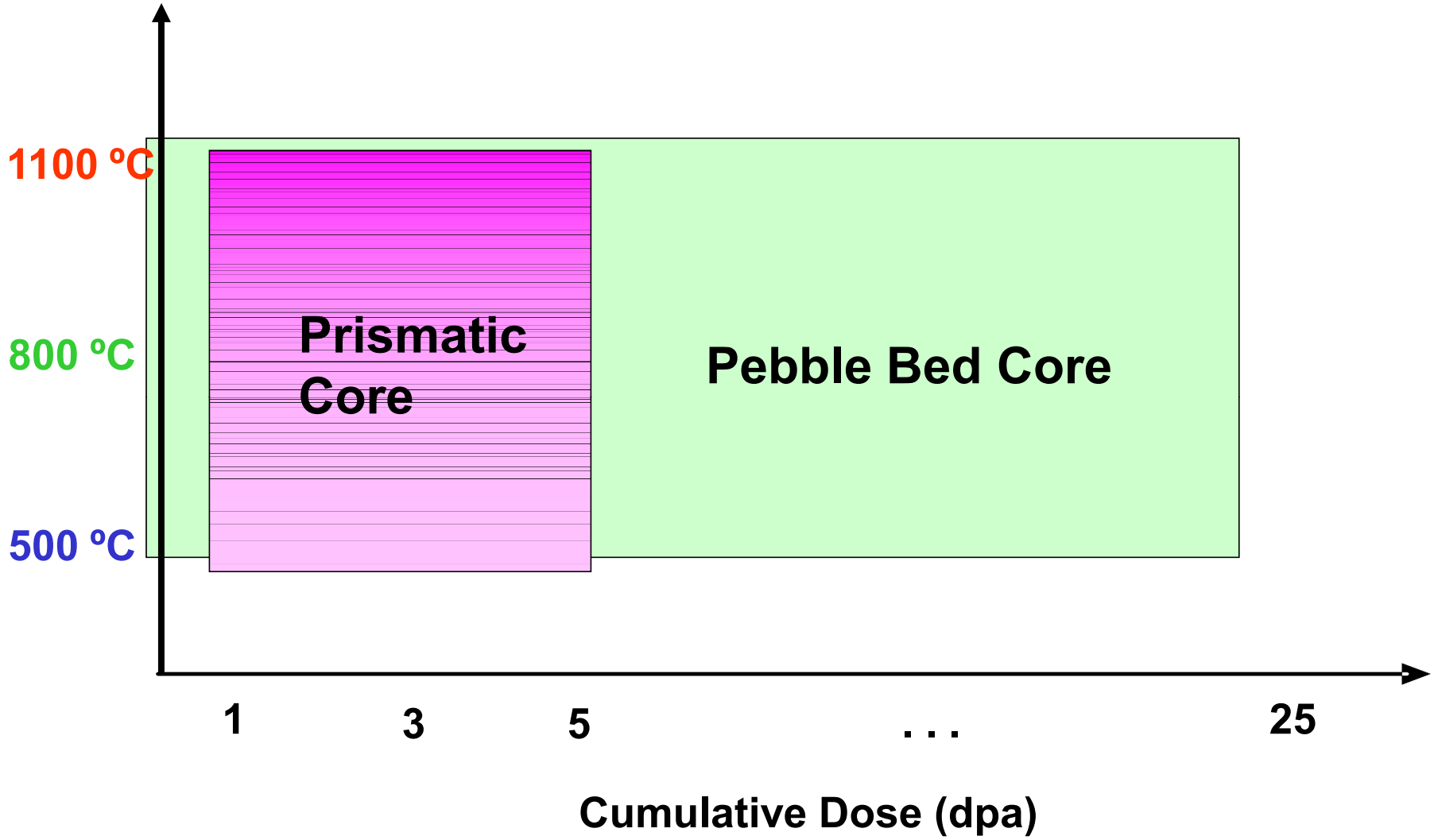
Source: ORNL/TM-2008/129

Candidates Graphite Materials for NGNP HTGR

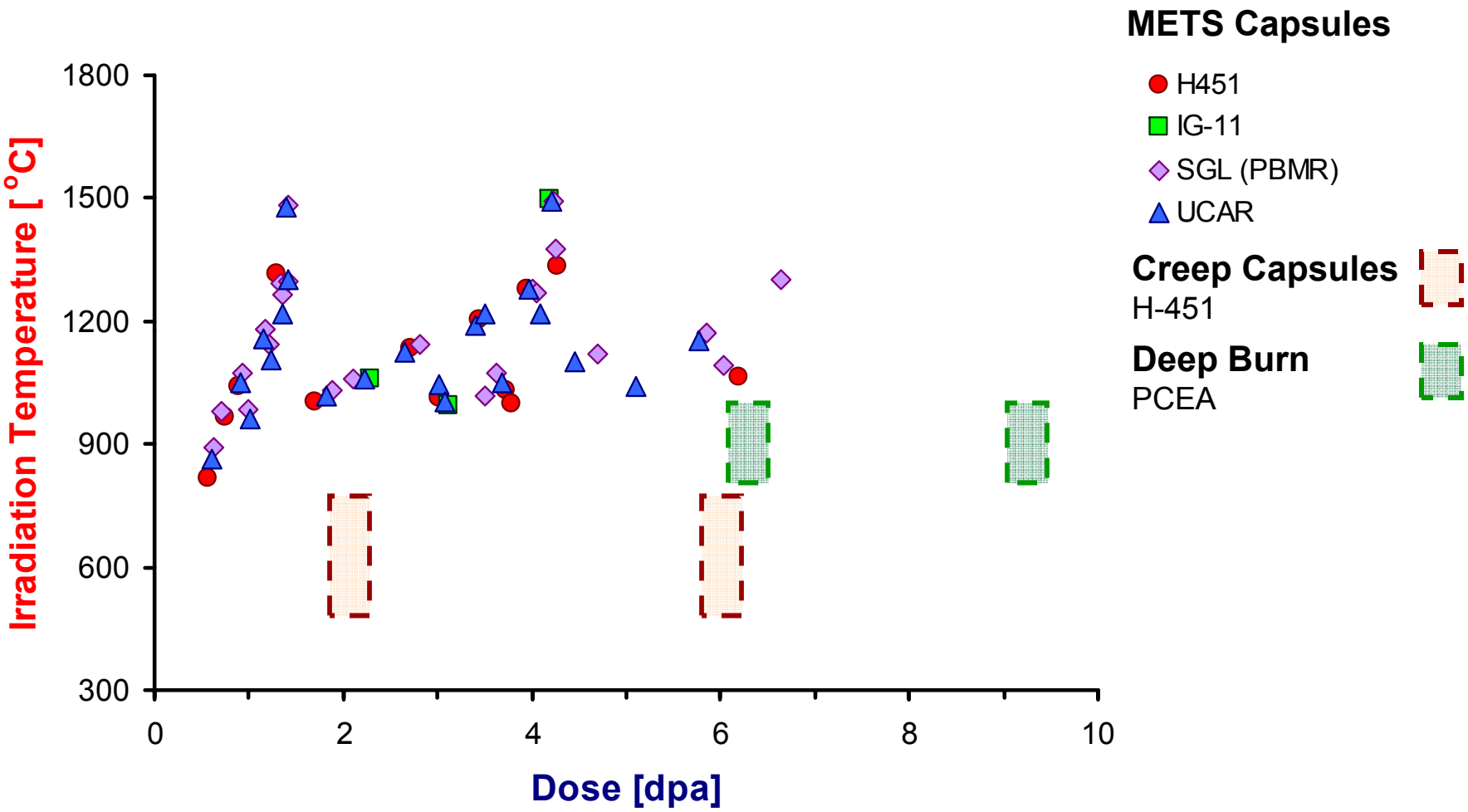
Grade	Manufacturer	Coke type	Comments	Grain size
IG-430	Toyo Tanso	Pitch coke	Isostatically molded; candidate for high dose region of NGNP	Fine
NBG-17	SGL	Pitch coke	Vibrationally molded; candidate for high-dose regions of NGNP prismatic core concepts	Medium
NBG-18	SGL	Pitch coke	Vibrationally molded; candidate for high-dose regions of NGNP pebble bed concepts; PBMR reflector graphite	Medium grain (1.6 mm max)
PCEA	GrafTech	Petroleum coke	Extruded; candidate or high-dose regions of NGNP prismatic core concepts	Medium
PGX	GrafTech	Petroleum coke	Large blocks for permanent structure in a prismatic core (used in HTTR)	Fine grain (~20 μm)
2020	Carbone of America	Petroleum coke	Isostatically molded; candidate for permanent structures in a prismatic core	Fine

Source: ORNL/TM-2008/129

NGNP Operating Conditions

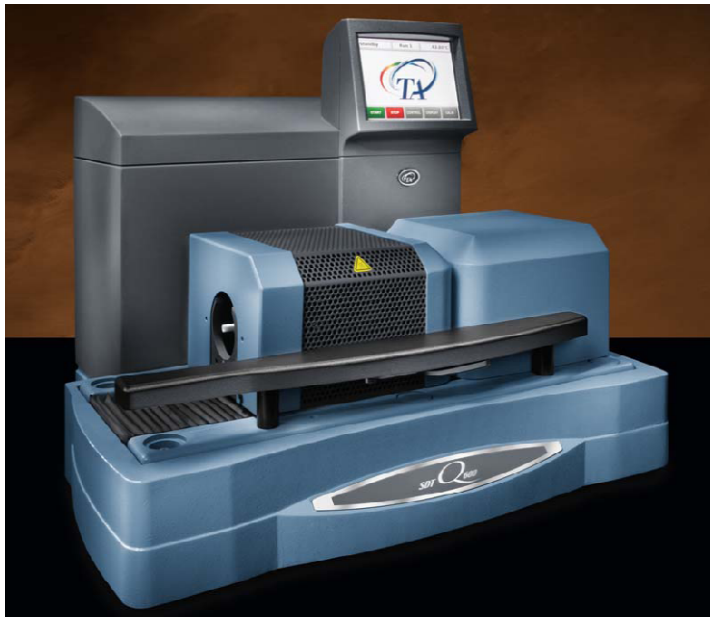


Current Inventory of Irradiated Graphites available at ORNL



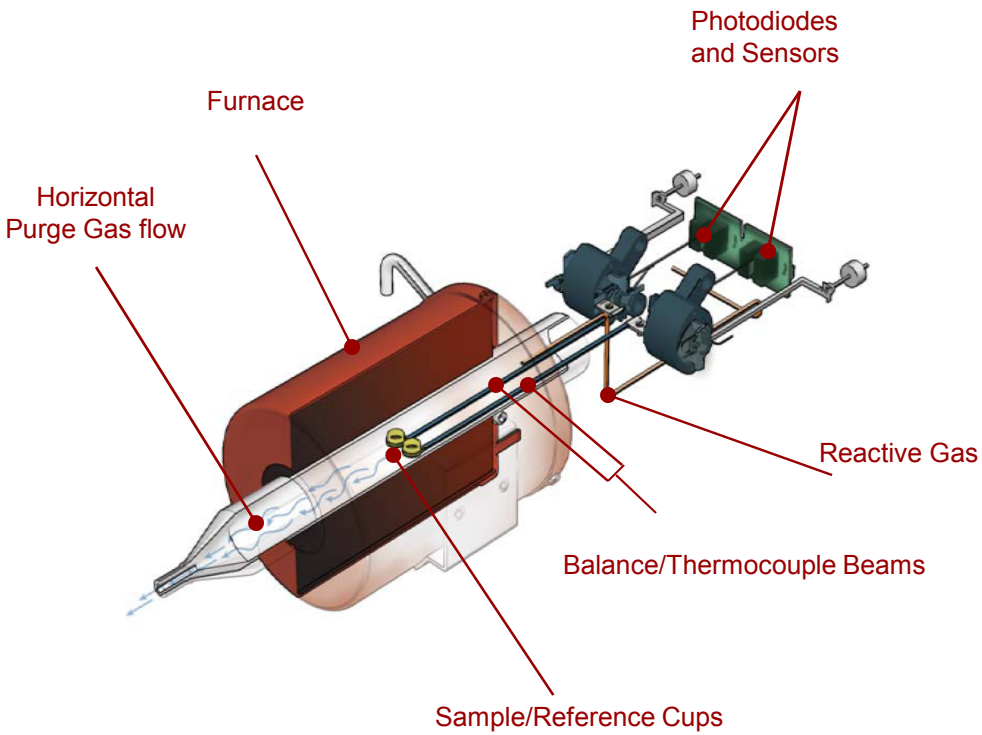
Our Experimental Set-up

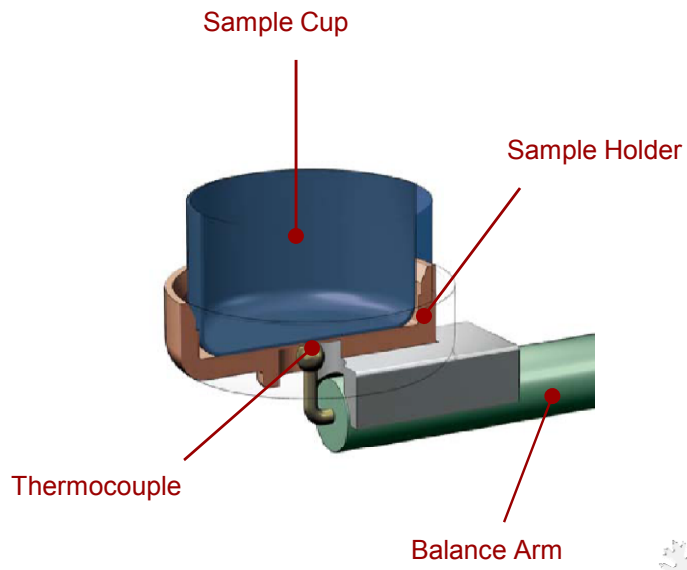
TA instruments Q600 Simultaneous DSC/TGA



- System Design: Horizontal Balance & Furnace
- Balance Design: Dual Beam (growth compensated)
- Sample Capacity: 200 mg
- Balance Sensitivity: 0.1 µg
- Furnace Type: Bifilar Wound
- Temperature Range: Ambient to 1500 °C
- Heating Rate
  - Ambient to 1000 °C: 0.1 to 100 °C/min
  - Ambient to 1500 °C: 0.1 to 25 °C/min
- Furnace Cooling: Forced Air (1500 to 50 °C in < 30 min, 1000 °C to 50 °C in < 20 min)
- Thermocouples: Platinum/Platinum-Rhodium (Type R)
- Temperature Calibration: Curie Point or Metal Standards (1 to 5 Points)
- DTA Sensitivity: 0.001 °C
- Calorimetric Accuracy/Precision: ± 2% (based on metal standards)
- Vacuum: to 7 Pa (0.05 torr)
- Sample Pans Platinum: 40 µL, 110 µl
- Alumina: 40 µL, 90 µL

Thermobalance and Furnace of the Q600





Instrument calibration

- Synthetic sapphire disk will be used as heat flow calibration standard
- Measurement of specific heat of non-irradiated graphite over the full temperature range (ambient to 1500°C);
  - currently available graphite grades:
    - H451
    - SGL NBG10
    - GrafTech PCEA
    - IG-110
  - Samples being machined in two directions: with grain / against grain
  - Two sample dimensions: (a) to match size of irradiated graphite; and (b) to match size of sample cup.

Current Challenges

- Procedure development for placing current irradiated graphite samples into instrument holders
  - Irradiated graphite samples (from METS) are disks of 5.8 mm diameter; vs. instrument's sample holders are 5.5 mm diameter.
  - Options include:
    - Machining excess of irradiated samples (not desirable)
    - Machining Alumina cups (being explored… not sure yet if possible)
    - Not using sample cup; placing graphite sample directly unto sample holder (TA technote vs. ASTM standard)
  - Samples from other capsules are larger sizes, some machining of irradiated samples will be necessary.
- Collecting full set of information on every sample of irradiated graphite to ensure that is appropriate for this study

Summary

- A stored-energy release study (SERS) of irradiated graphite has been initiated at ORNL.
- A literature review is in progress to evaluate the current knowledge of the effects of energy stored on previously irradiated nuclear graphite during potential accidents.
- An experimental plan to study the stored energy of irradiated graphite is under development; this includes an assessment of availability of irradiated graphite samples.