July 26, 2023

#### Will Windes (for Steve Johns)

Directorate Fellow Idaho National Laboratory

# **ASME Irradiation Model**

How to deal with irradiation data in ASME code rules

**DOE ART Gas-Cooled Reactor (GCR) Review Meeting** Virtual Meeting July 25 – 27, 2023



# Nuclear graphite has a significant challenge (problem)

- There is not enough irradiation data to qualify graphite for nuclear application
  - For all grades
  - At all irradiation temperatures
  - Over entire range of anticipated dose
- Problem is the variety of different grades
  - No single "nuclear" grade
  - Historical grades with irradiation experience no longer available
  - Irradiation data across available temperature range is limited
- We don't have time/room in available MTRs to get all the required data



# ARTICLE HHA-II-2000 MATERIALS DATA SHEET - ASME BPVC.III.5-2021

- ASME code rule modification for support of new reactor concepts and commercial vendors.
- Irradiation induced property change.
  - 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

ASME MDS Sheet for qualification of HTR graphite for NRC licensing.

Irradiated Graphite			
Property	Units	WG	AG
Dimensional change [.] ③			
Creep coefficient [.] ③			
Coefficient of thermal expansion [.] 🚳			
Strength [.] 3			
Elastic modulus [.] 🚳			
Thermal conductivity [.] 36			
GENERAL NOTES: (a) WG, AG refers to the with- and against-grain direction of the material. (b) [.] indicates a dimensionless quantity.		NOTE: (1) If the maximum intended use temperature exceeds 1,832°F, then the temperature dependent data shall be extended to cover the property values at the maximum intended use temperature.	
(07/21)			

# **Irradiation** Behavior

Significant changes occur during normal operation:

- Density Densification
  - Graphite gets denser with irradiation until Turnaround de
  - 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. It then decreases
- Coefficient of thermal expansion
  - Initial increase but then reduces before Turnaround
  - CTE is why properties are so temperature dependent
- Thermal conductivity
  - Decreases almost immediately to ~30% of unirradiated values
  - At temperatures it is same as unirradiated conductivity





12

(%) 1/7 4

### Leveraging the data generated by GIF countries last 20 years



So long as "new" data falls within data cloud we can use all grades to predict behavior

- AGC Experiment = 20+ years
- We need figure out a way to get needed irradiation data without new "AGC" tests
- Why not leverage all existing data?
  - Assume all grades behave similarly
  - At least until turnaround dose is achieved
- If we can demonstrate that all graphites behave similarly we can create engineered limits
- Let's see if we can do this ...

#### **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<sup>2</sup>)

 $A_x$ : 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: 
$$G_x(\gamma) = A_x G_c(\gamma) + (1 - A_x) G_a(\gamma) + F_x(\gamma)$$
 Linear  
Non-linear

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

### All Graphites Behave Similar – Can we Predict the % $\Delta$ ?



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

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

- Proposed is empirical polynomial fitting *up to* turnaround.
- To be used as a reference for ASME code / commercial vendors, for dimension change (%∆) at a given dose.
- Shown is 5<sup>th</sup> 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).

#### 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.
  - Example. by small, medium and large grained graphites.

## **Turnaround Dose is a Function of Temperature**

- Identify turnaround dose (TAD) as a function of temperature.
- Turnaround is a temperature dependent response (thermally activated).
- Define an Arrhenius function

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

Assume all grades have similar defects. **Thus, same activation energy** ( $E_a$ ).

DOE-HTGR-88111, Graphite Design Handbook, General Atomics Company, CA, 1988

- Fundamentally, on the atomic scale, all nuclear graphites are the same. sp<sub>2</sub> bonded Carbon with some degree of disorder.
- Variation in the irradiation response amongst grades comes from differences in the meso – macroscale features.

All other graphite differences are in  $A \propto (grain \ size, \rho_o, CTE)$ 

#### **Arrhenius Fits for H-451 Dimensional Change Data**

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

- During the first fitting, the activation energy and preexponential were allowed to vary.
- A second fitting was conducted by setting the activation energy as an average from axial and radial data.



### **TAD Extrapolated with 5<sup>th</sup> Order Polynomial**



#### Irradiation temperature is critical

- "Same" activation energy implies "same" defects
  - Defects form at different temperatures
  - Defects can anneal out at higher temperatures

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





- An Arrhenius approach to predict the turnaround behavior may be a viable solution for the ASME MDS requirements for all nuclear graphites.
- This model is anticipated to include the elastic modulus and electrical resistivity.
- A draft manuscript to be submitted to a referred journal is anticipated by the end of FY23.