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Graphite Irradiation Data Analysis

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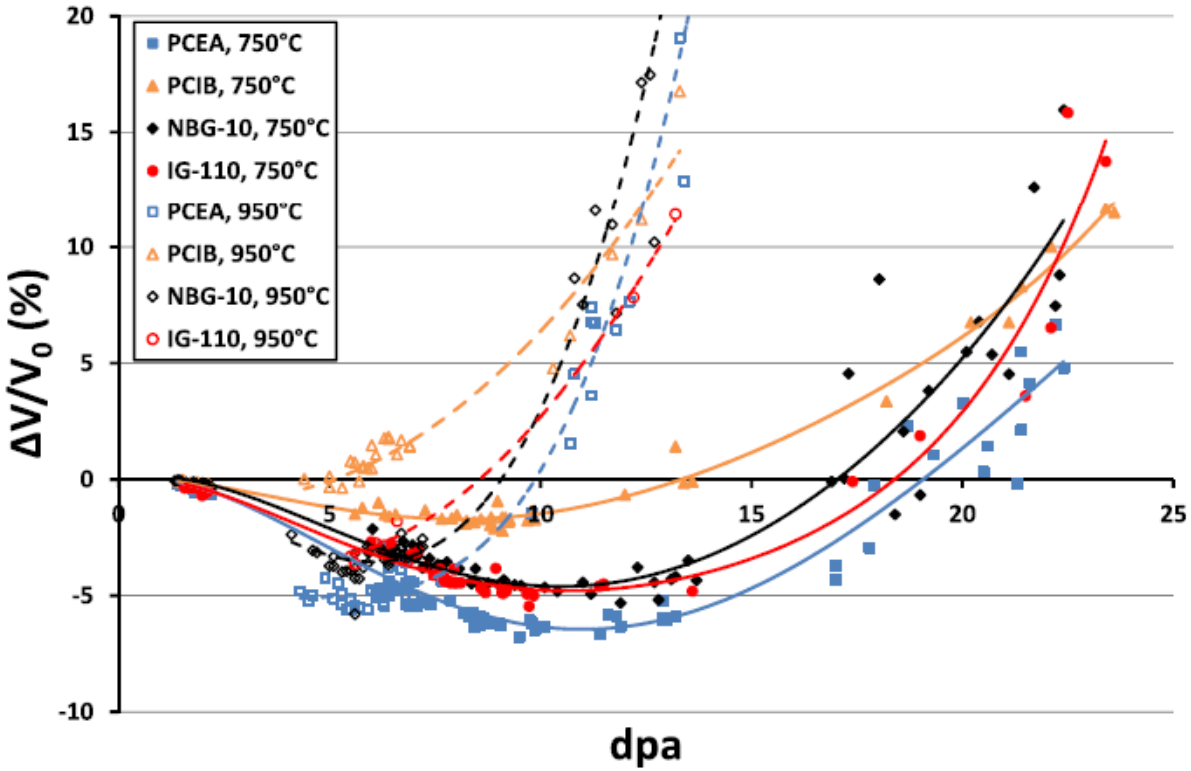
Graphite Irradiation Data Analysis

Semi-Empirical Modeling of Turnaround Behavior

ART Advanced Reactor Technologies
GAS-COOLED REACTORS

Future ASME Code Development and Reactor Deployment

- 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.
- Irradiation temperature is a key parameter effecting turnaround dose.
- Dependent on coke source, and manufacturing process, each nuclear graphite has a unique response.



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Define a Universal Response for Nuclear Graphite

- Qualifying each candidate grade of nuclear graphite at various temperature regimes is unreasonable.
  - New irradiation campaigns are time consuming
  - Can be extremely expensive (millions)
- For development of ASME code rule, and to inform vendors, can a universal response be defined?

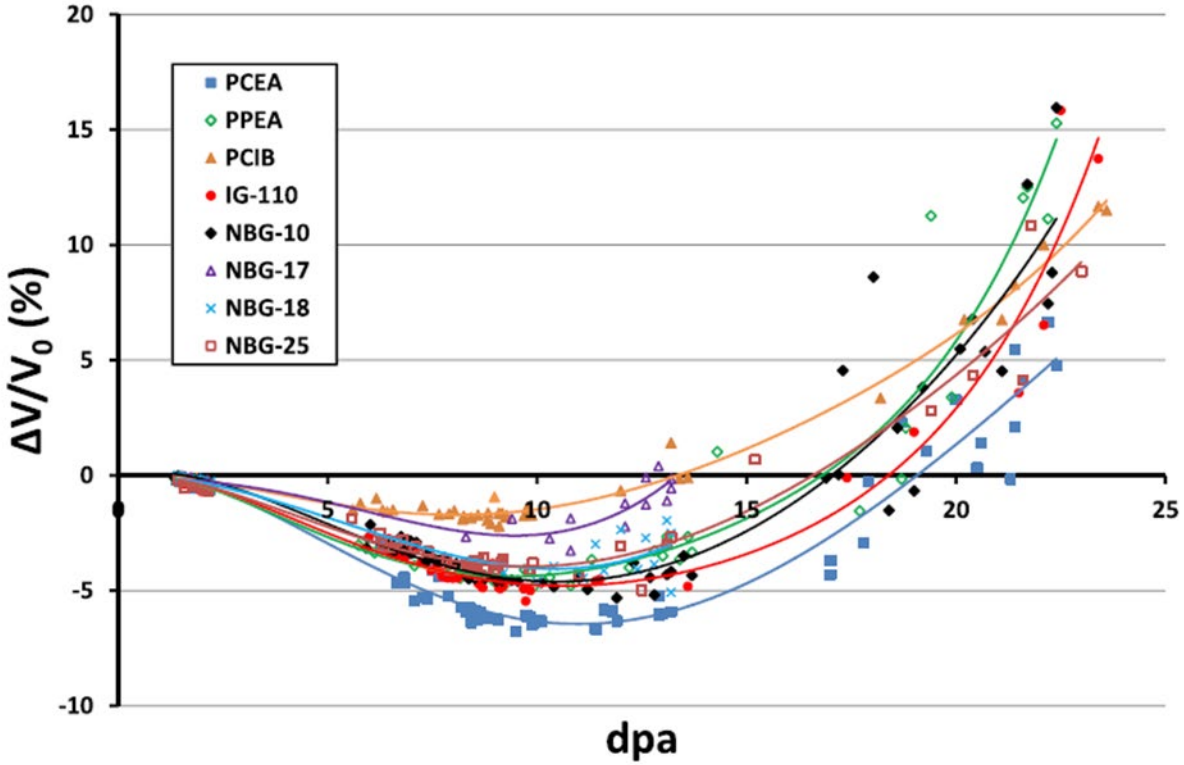

Fig. 2. Dimensional change as function of dpa at 750 °C.

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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)

γ: Fast neutron fluence (n/m^2)

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: $\boldsymbol{G_x(\gamma) = A_x G_c(\gamma) + (1 - A_x) G_a(\gamma) + F_x(\gamma)}$

Linear

Non-linear

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

Dimensional Change Theory

$$G_x(\gamma) = A_x G_c(\gamma) + (1 - A_x) G_a(\gamma) + F_x(\gamma)$$

If the porosity function is assumed to be quadratic (i.e., porosity decreases then increases with increasing dose), the semi-empirical relationship is given by:

$$G_x(\gamma) = a_1 \gamma^2 + a_2 \gamma$$

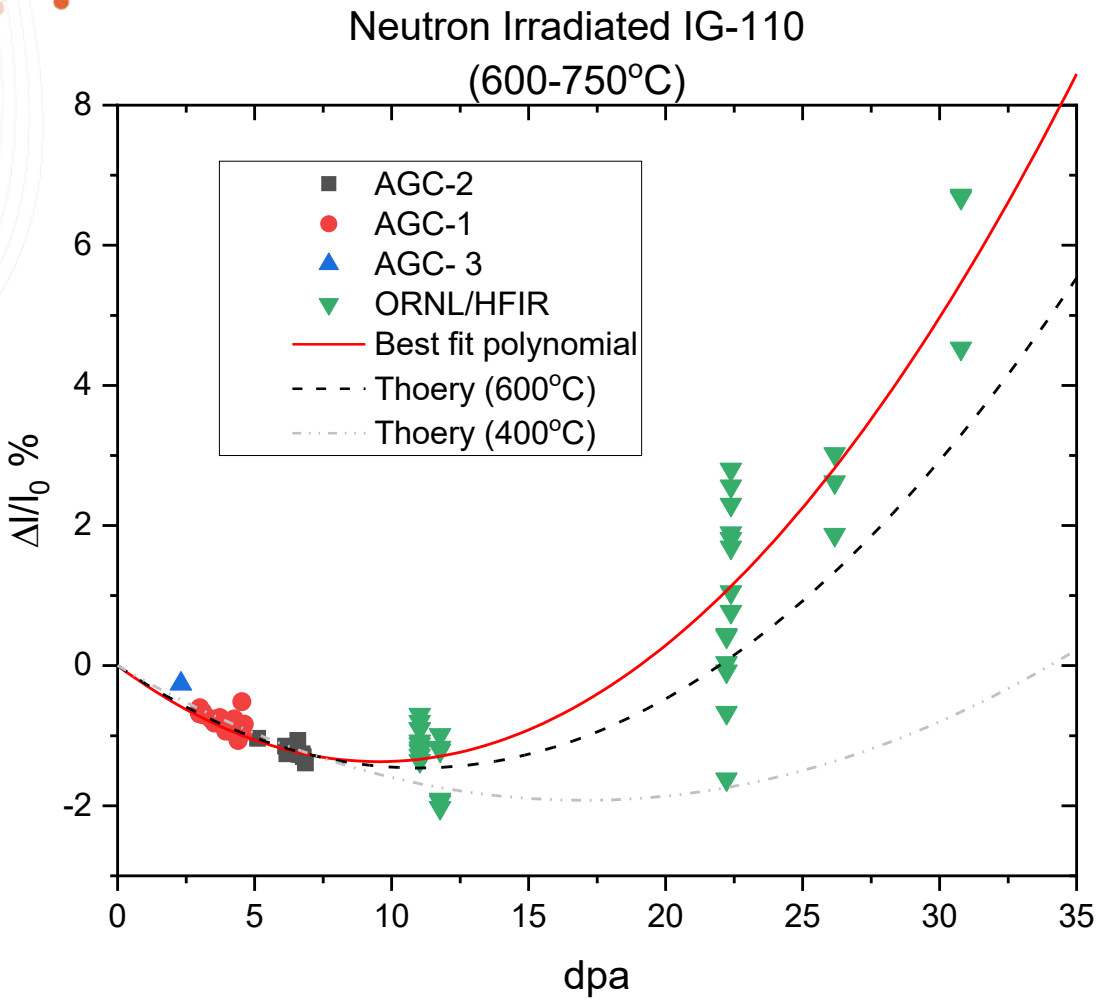
Where a_1 and a_2 are temperature dependent constants.

For IG-110, literature gives the following values:

Grade	**IG-110**	
Irradiation temperature (°C)	a_1	a_2
400	0.279	-1.64
600	0.450	-1.86
800	0.821	-2.19

S. Mohanty, S. Majumdar, M. Srinivasan, U.S.N.R. Commission HTGR Graphite Core Component Stress Analysis Research Program – Task 1 Technical Letter Report (2011)

Theoretical vs Empirical Modeling



- Between best fit polynomial, theory at 400 and 600°C, there is little to no deviation *before* turnaround dose.
- Shown is that the methodology, theoretical or purely empirical, is not significant until after turnaround.
- Proposed is to empirically fit all available data to inform ASME code development.

All Graphites Behave Similar – Can we Predict the %Δ?

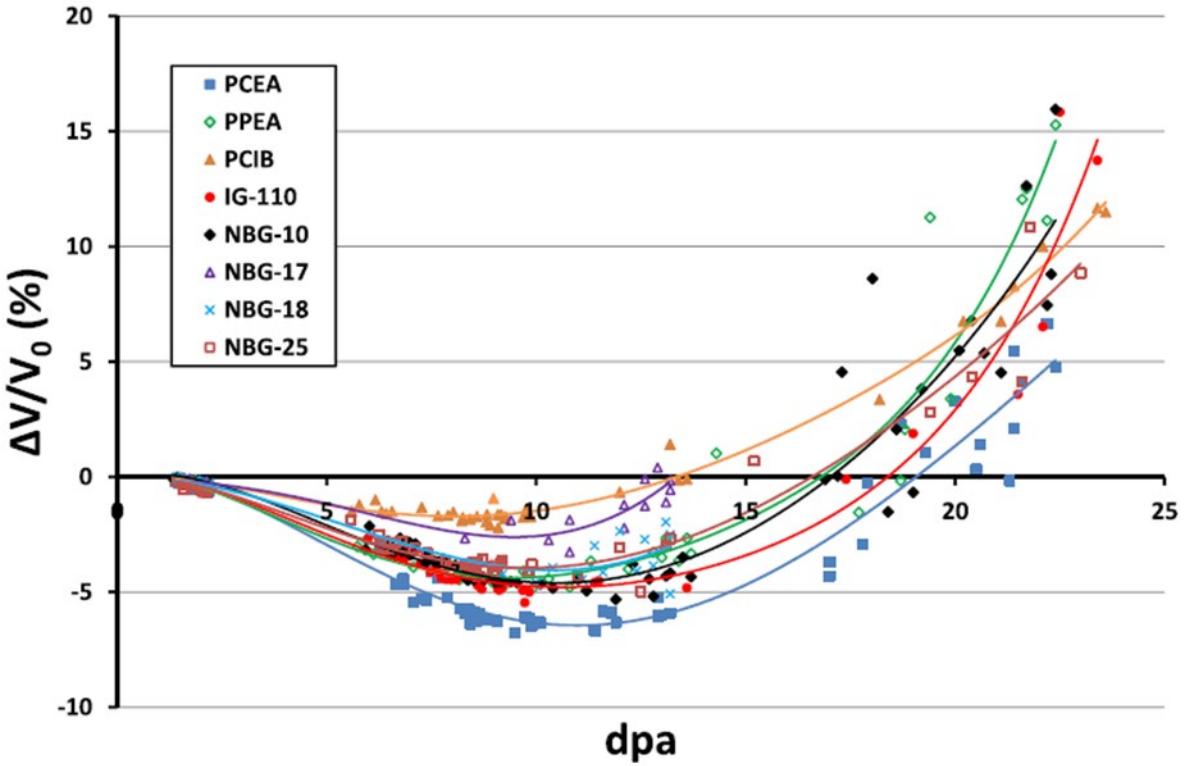


Fig. 2. Dimensional change as function of dpa at 750 °C.

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

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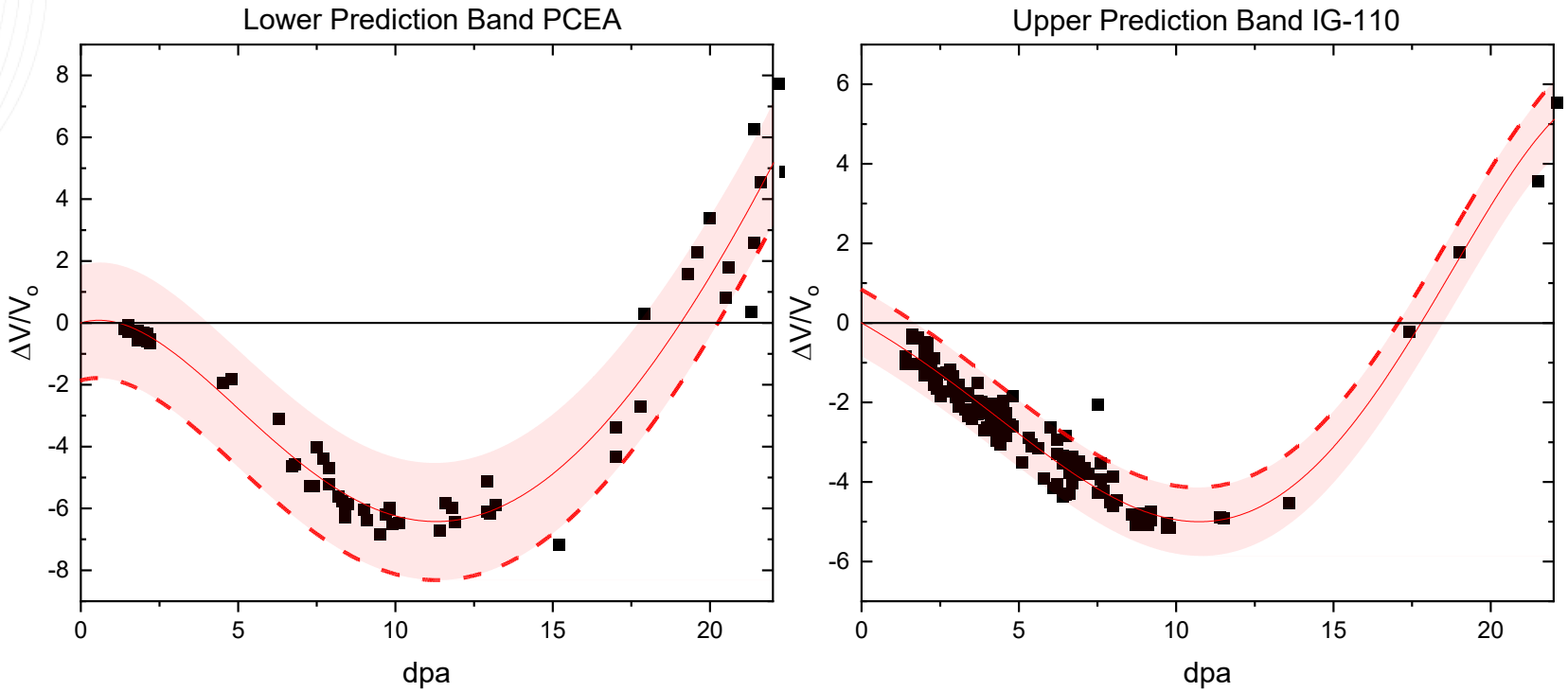
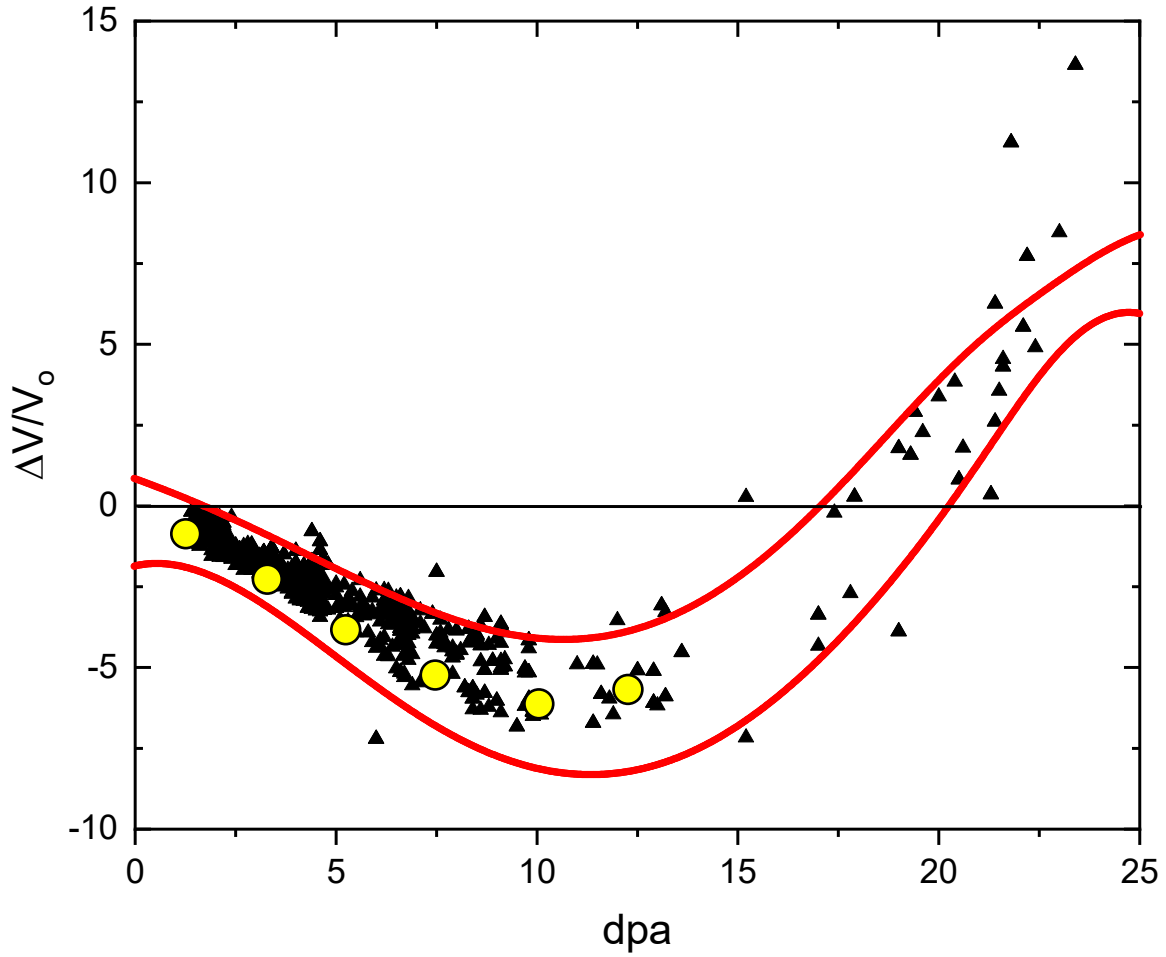
Irradiation Data From AGC1-3 and InnoGraph



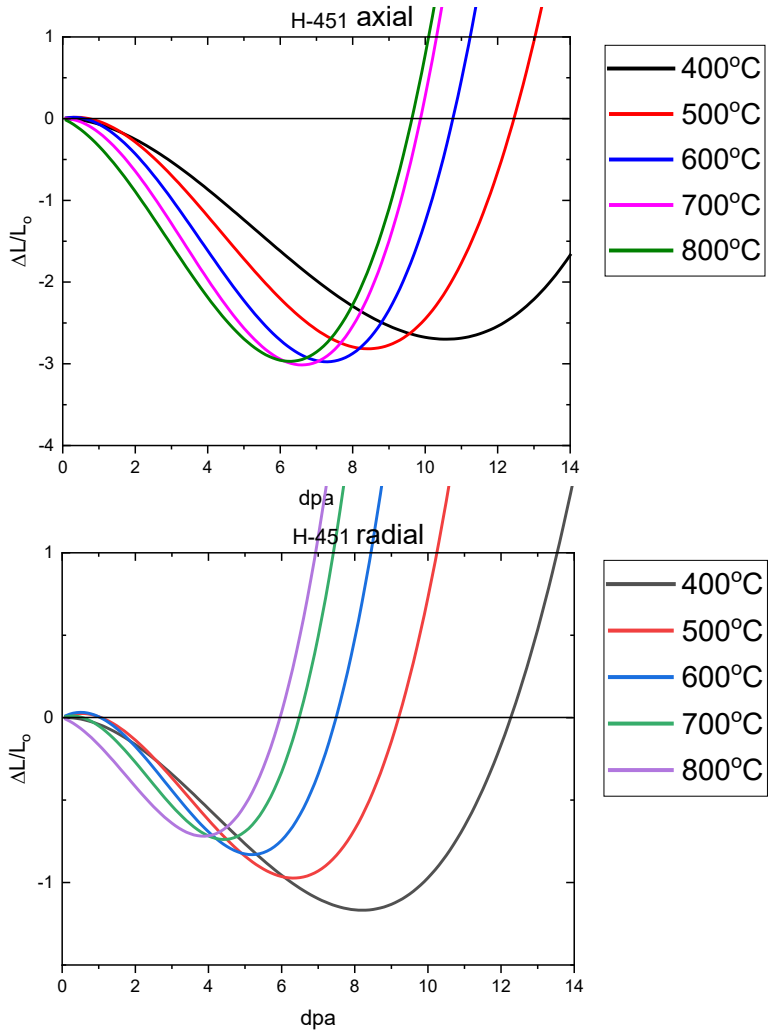
- Define a 95% lower prediction band with PCEA.
  - Medium grained, extruded
- Define a 95% upper predication band with IG-110
  - Fine grained, isomolded

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.
  - Ex. by small, medium and large grained graphites.

Turnaround Dose is a Function of Temperature



- Historical grade H-451 dimensional change model (3rd order polynomial).
- Identify turnaround dose as a function of temperature.
- Turnaround is a temperature dependent response (thermally activated).
- Define an Arrhenius function

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

- All graphites should have the same activation energy (E_a).

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

Assume E_a is Constant for all Nuclear Graphites

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

$$A(E_o, A_x, \rho_o, CTE)$$

E_o= Elastic Modulus

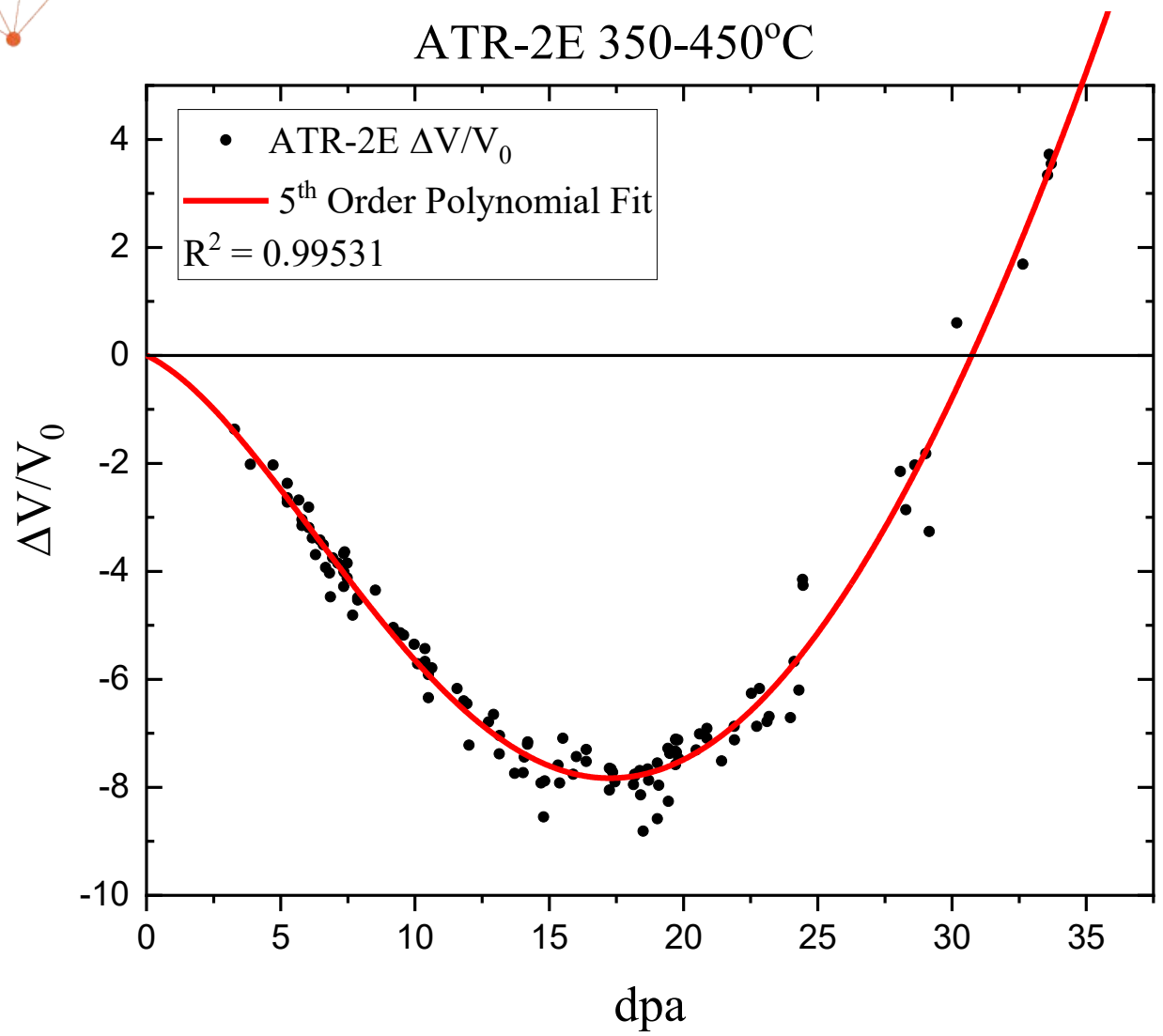
A_x= Anisotropic Factor

ρ_o= Density

CTE = Coefficient of Thermal Expansion

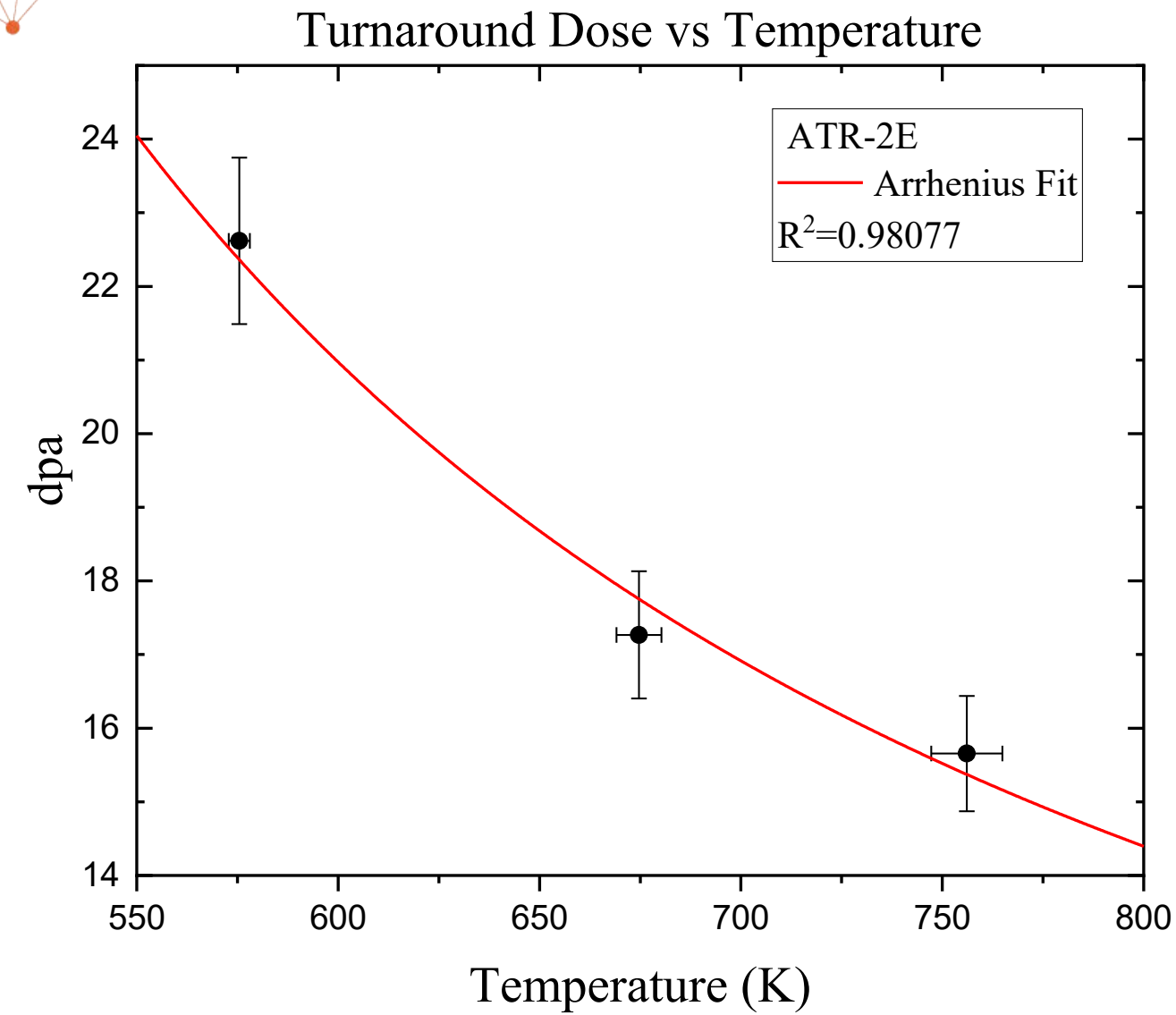
- Fundamentally, on the atomic scale, all nuclear graphites are the same. Sp_2 bonded Carbon with some degree of disorder.
- Variation in the irradiation response amongst grades comes from differences in the meso – macroscale features.
- The value of the pre-exponential factor could be defined by *baseline* properties specific to grade.

Historical Graphite Irradiation Data



- Complete ‘sets’ of data needed for Arrhenius models.
  - Enough data points to accurately fit dimensional change behavior.
  - At least 3 temperature regimes of irradiation.

Arrhenius Fitting



- For each turnaround point, polynomial fits must have R^2 values above .90.
- Horizontal error bars represent one standard deviation of the mathematical average of temperature data.
- Vertical error bars assume 5% error in dose calculation.

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

Arrhenius Fits for Complete 'Sets' of Data

Best fits allowing E_a and A to vary.

Graphite Grade	E_a / k_b	A	R^2
ASR-1R SQ75	-1151.5655	2.62588	0.99996
ASR-2R	-1074.5024	2.95306	0.9877
ATR-2E	-923.7902	4.51307	0.98619
V356	-827.45755	4.54242	0.96885
V483	-899.3156	4.17444	0.99996
G347A	-1011.99749	2.83742	0.9992
	Ave. = -981.4381		
	St.Dev.= 109.7843		

Arrhenius Fits for Complete 'Sets' of Data

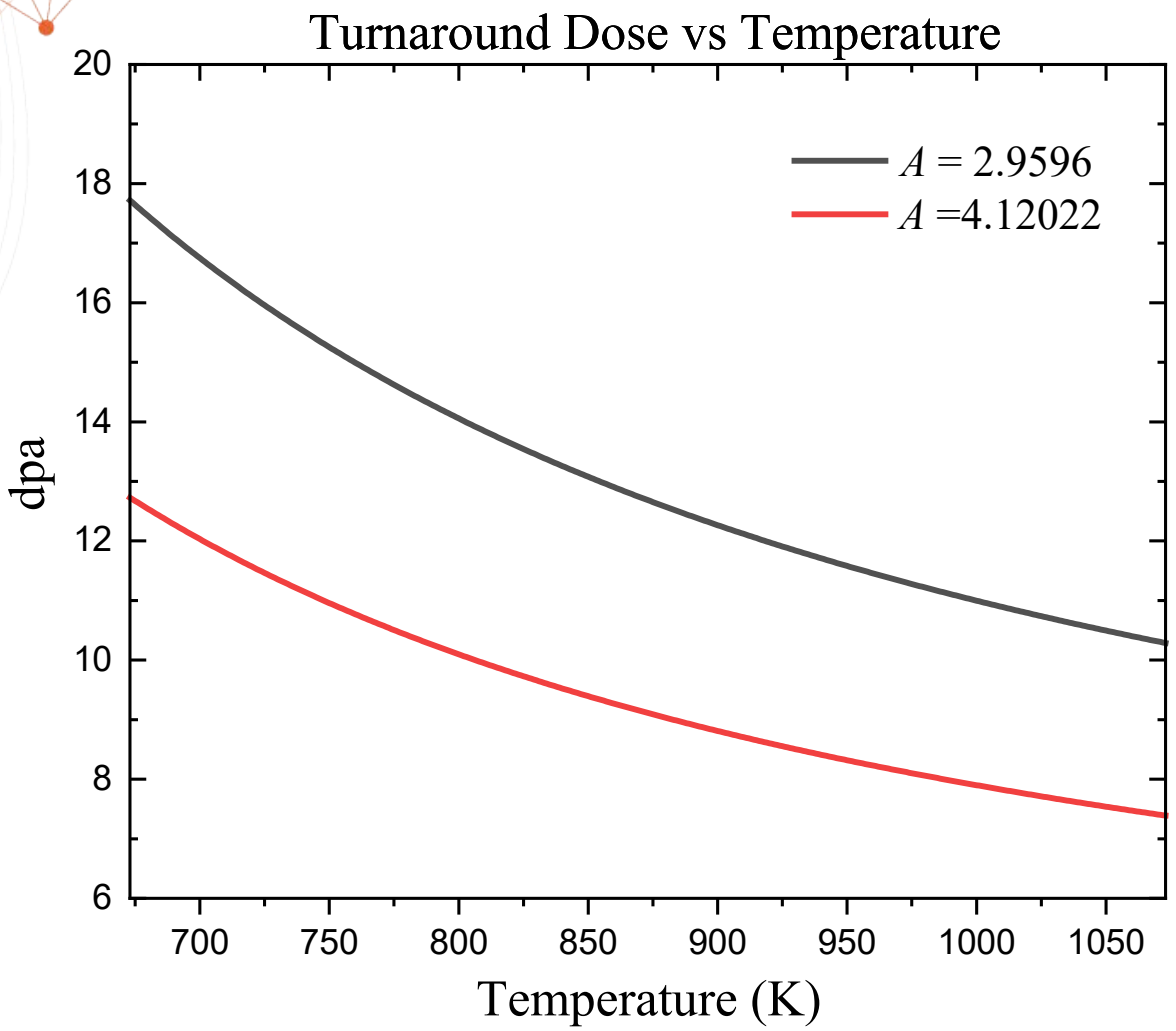
Best fits holding E_a constant and allowing A to vary.

Graphite Grade	E_a / k_b	A	R^2
ASR-1R SQ75	-981.4381	3.42823	0.97875
ASR-2R	-981.4381	3.41626	0.9805
ATR-2E	-981.4381	4.12022	0.98233
V356	-981.4381	3.56761	0.93602
V483	-981.4381	3.99176	0.99176
G347A	-981.4381	2.9596	0.99832
	E_a = 0.085 eV		

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

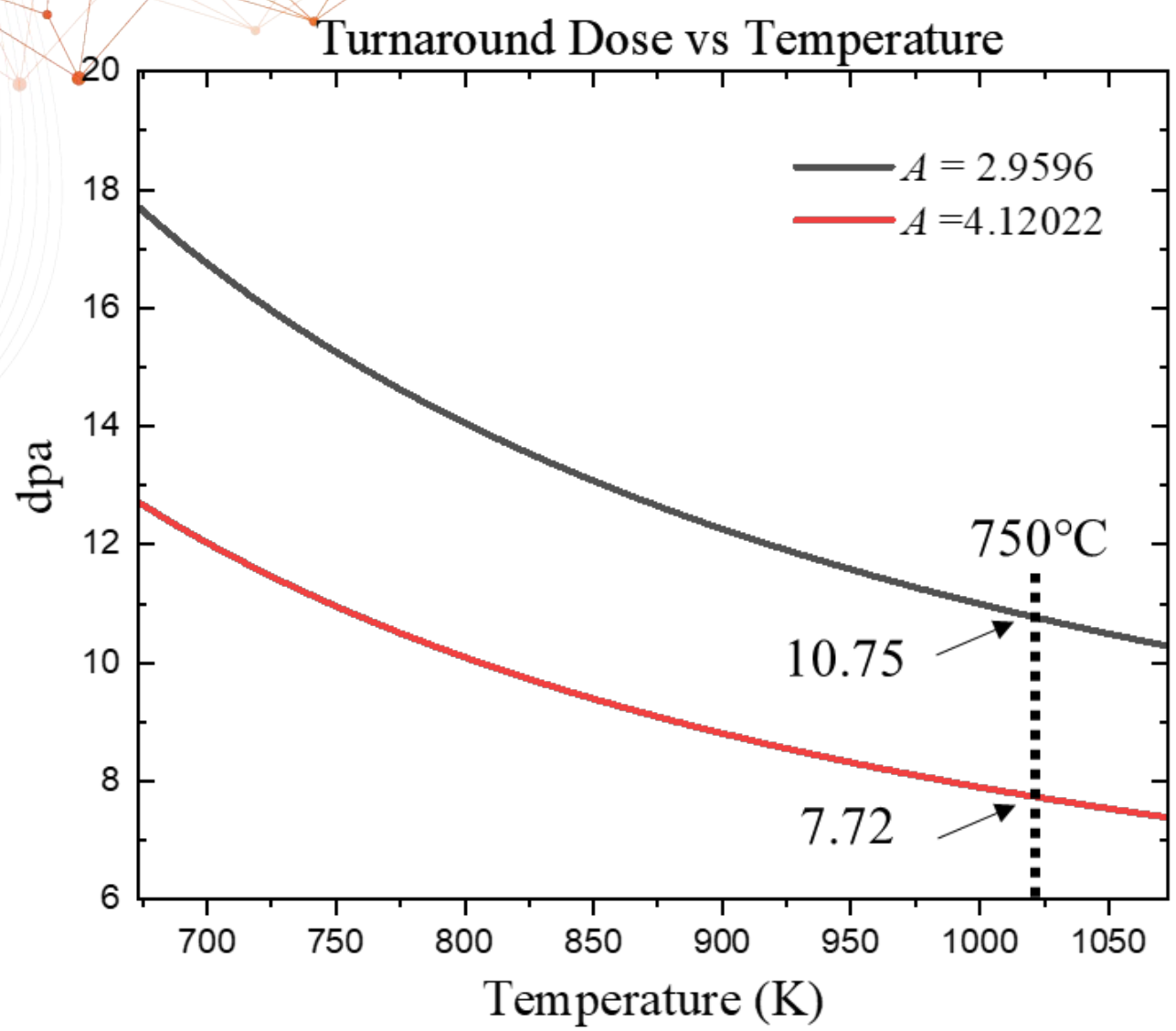
$$A(E_o, A_x, \rho_o, CTE)$$

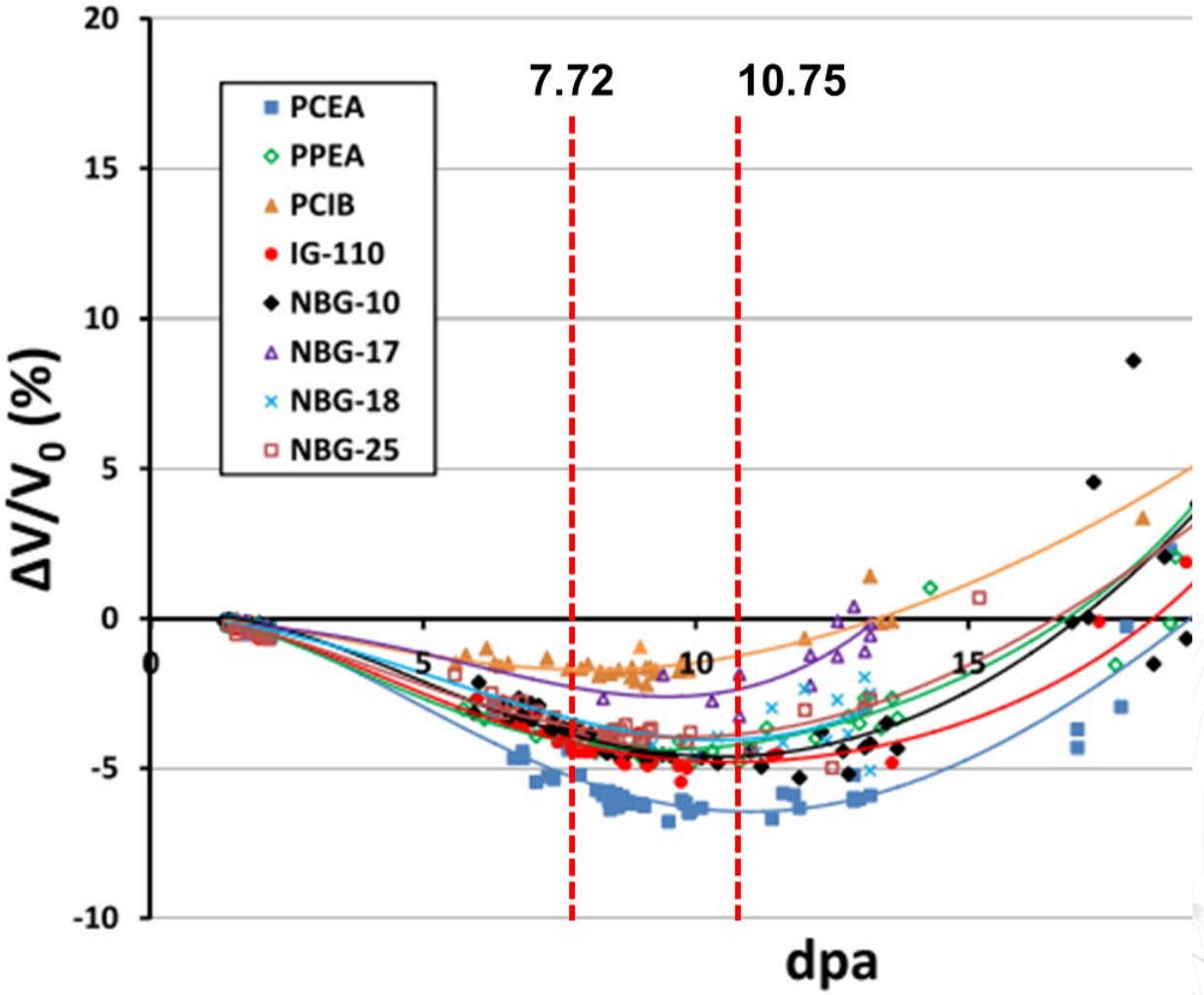
Universal E_a with Upper and Lower Pre-Exponential Values



- The values of the pre-exponential range from ~ 3-4.
- High *A* correspond to a higher turnaround dose.
- Low *A* correspond to a lower turnaround dose.
- The rate of dimensional change is a function of the '*accommodating porosity*'.
- I.e., that which allows for irradiation induced *c*-axis expansion.
- A function pore size, porosity, density, grain size, and anisotropy.

Arrhenius Model Compared to Current Grades





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Conclusions

- To support ASME code development and reactor deployment, a significant effort is being made to define universal code rules for all graphites and their irradiation property changes.
- The amount of dimensional change may be reasonably predicted for all grades using empirical models; however, data needs to be refined by 100°C increments (400-800 °C).
- The turnaround fluence is reasonably predicted for all grades assuming a constant E_a. Analysis and defining the pre-exponential by baseline properties is anticipated (pore size, porosity, density, grain size, and anisotropy).
- This code rule development should allow vendors to conduct relatively small irradiation campaigns to validate their graphite of choice.

Thank You!

Questions?

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