Understanding Graphite Behavior in Nuclear Reactor Environments for Lifetime Predictions

Anne A. Campbell, Ph.D.
Oak Ridge National Laboratory
Oak Ridge, TN
campbellaa@ornl.gov

US-NRC Advanced Non-Light Water Reactors – Materials and Component Integrity Workshop, December 9-11

ORNL is managed by UT-Battelle, LLC for the US Department of Energy

Irradiation Effects in Graphite

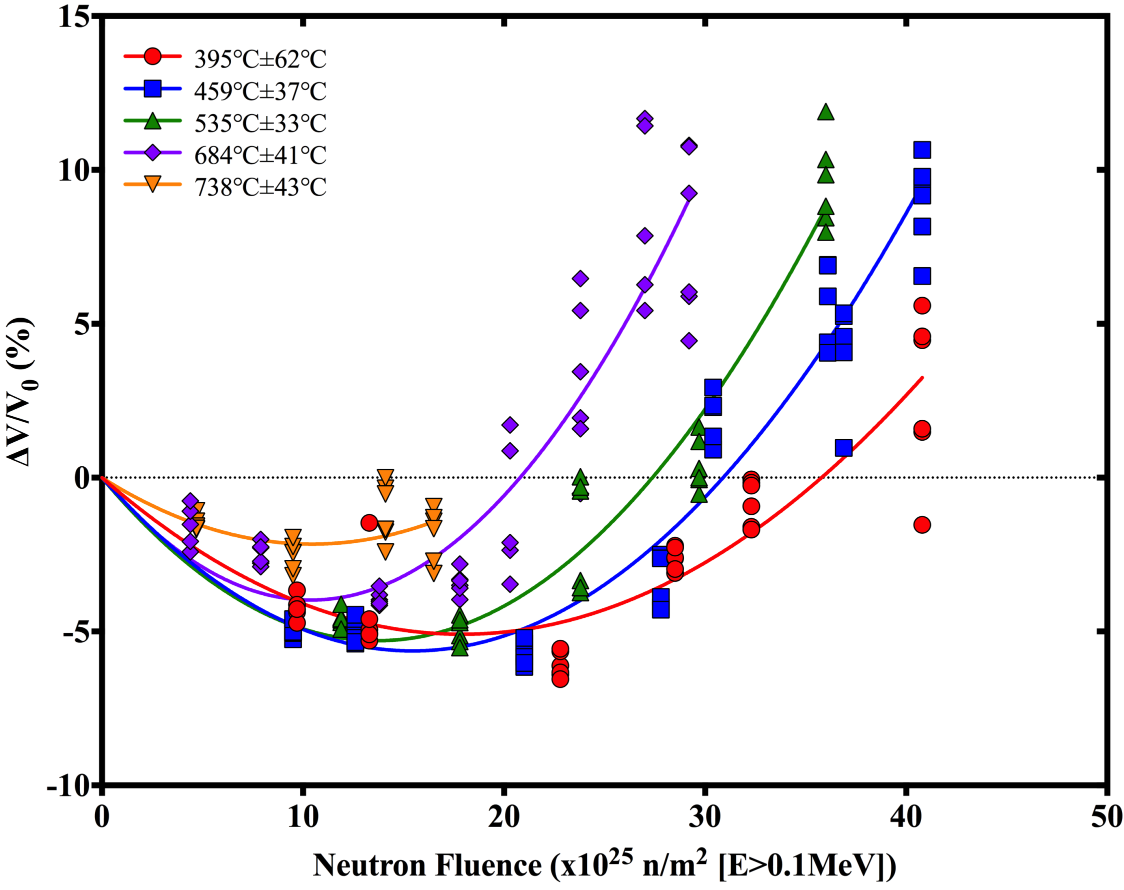
- Irradiation damage in graphite causes changes to all the physical, thermal, and mechanical properties of graphite
- Property change trends are similar between grades, but the net change is different for each
  - Example is the plot of volume change to the right showing 4 different grades

See Fig.1 from following paper: Heijna et al. *Journal of Nuclear Materials*, v492 (2017) p148

Heijna et al. *JNM* v492 p148

Volumetric Change

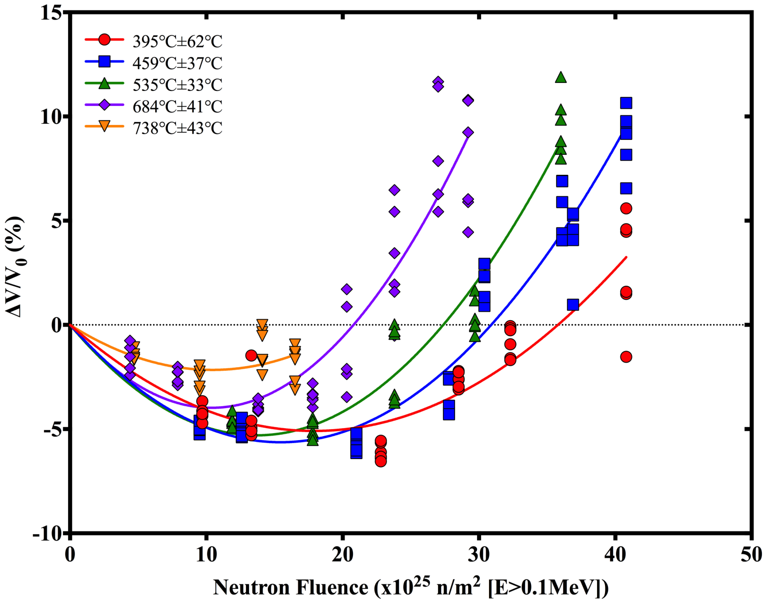
- Graphite dimensional changes are a result of crystallite dimensional change and graphite texture.
- Swelling in c-direction is initially accommodated by aligned microcracks that form on cooling during manufacture.
- Therefore, the a-axis shrinkage initially dominates and the bulk graphite exhibits net volume shrinkage.
- With further irradiation, incompatibilities in crystallite strains causes the generation of new porosity and the volume shrinkage rate falls eventually reaching zero.

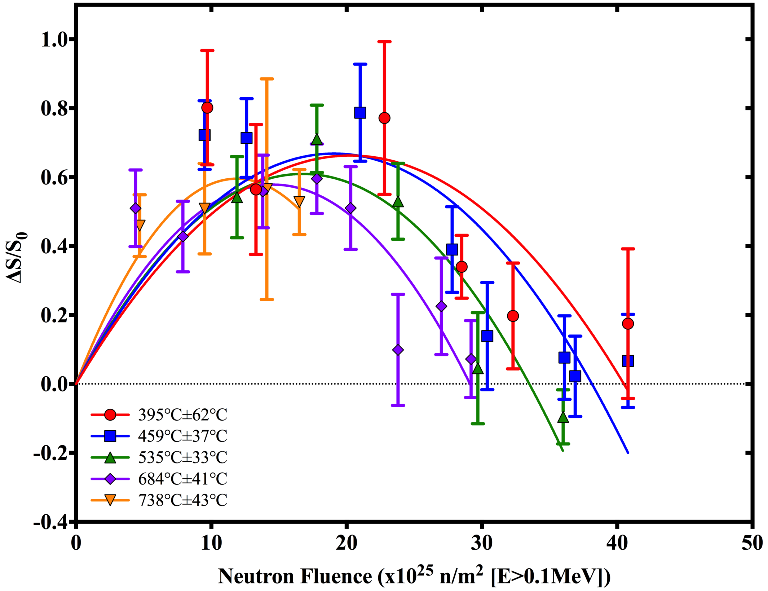


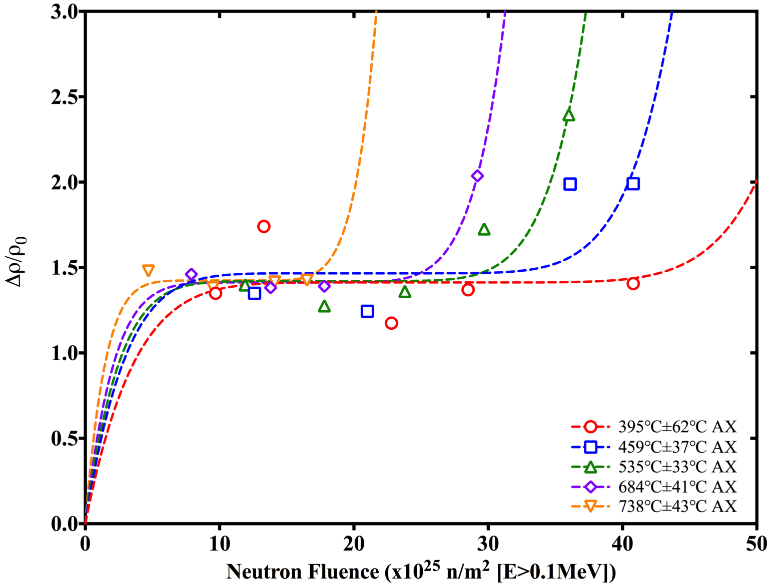
Campbell, et al., *Carbon*, 109 (2016) 860-873

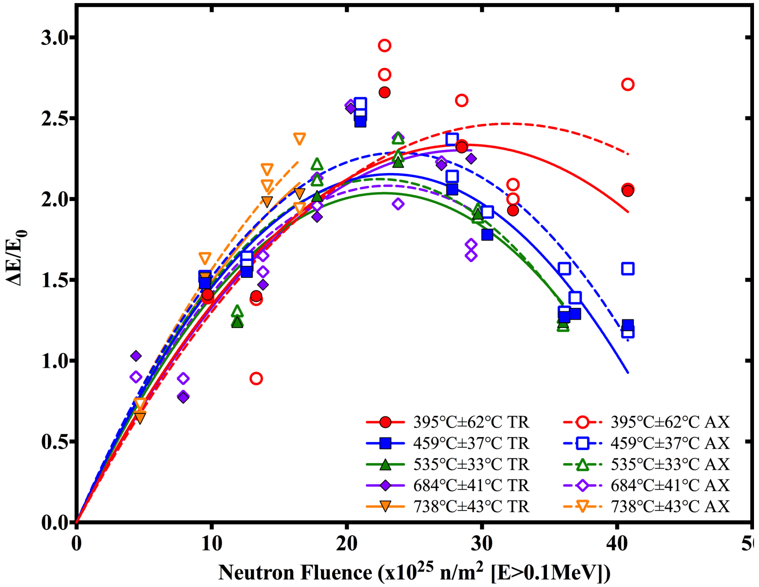
Neutron Fluence and Temperature Effects on Properties

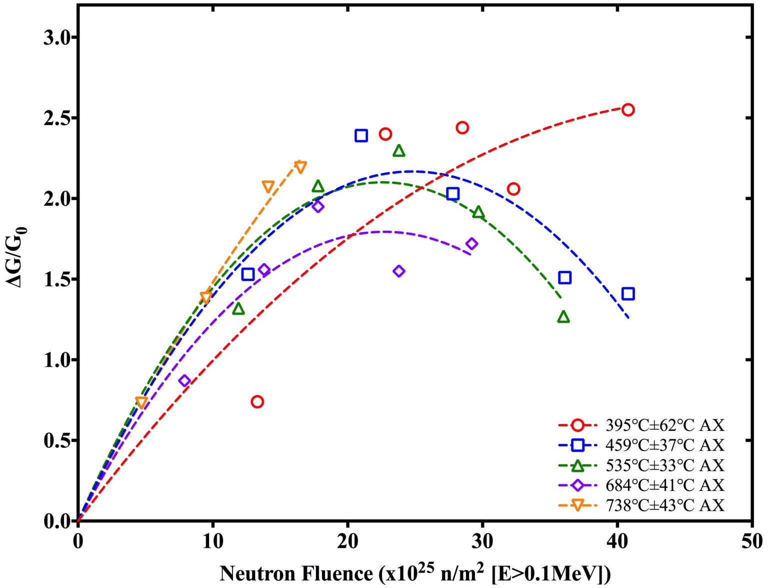
- Volume change follows semi-parabolic shape for increasing fluence
  - Higher temperature reduces net contraction and time for swelling onset
- Strength, Young's modulus, and shear modulus have trends that appear to mimic the density change
  - Following density curve is seen for isotropic fine-grained graphite and is different behavior than seen in coarser grades
  - Poisson's ratio does not change







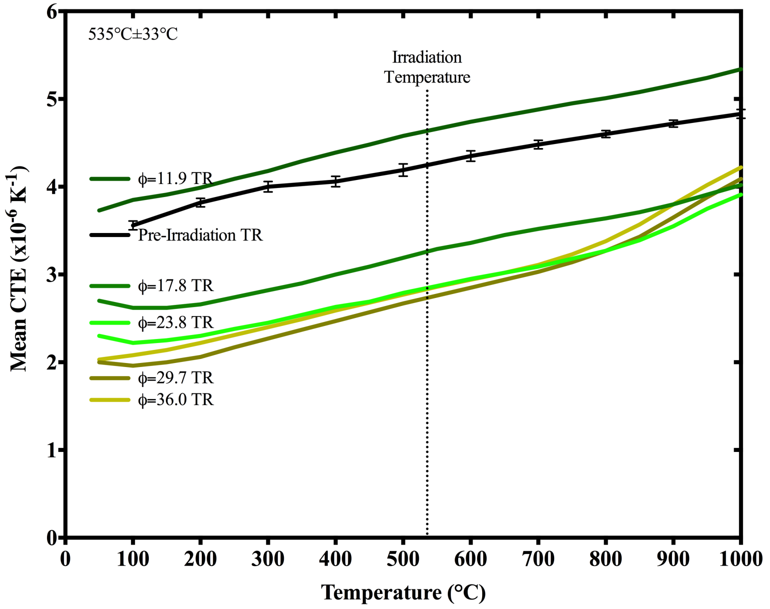


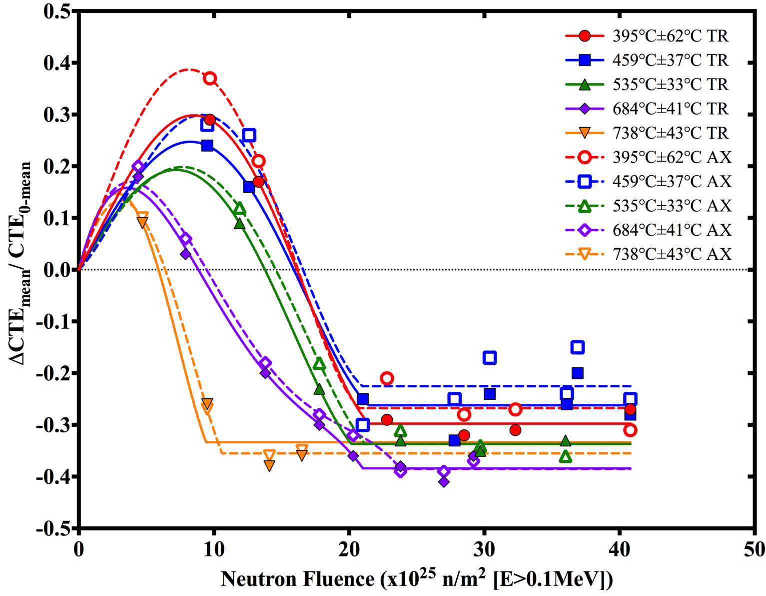


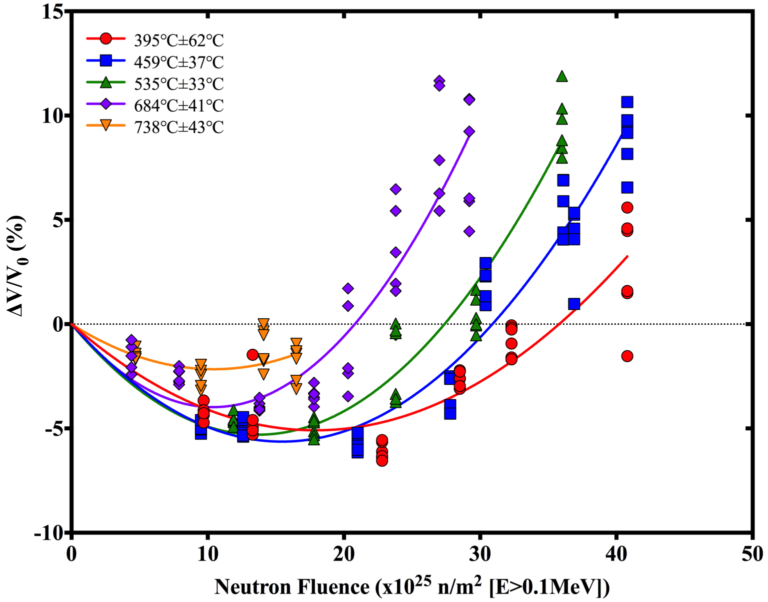
Campbell, et al., *Carbon*, 109 (2016) 860-873

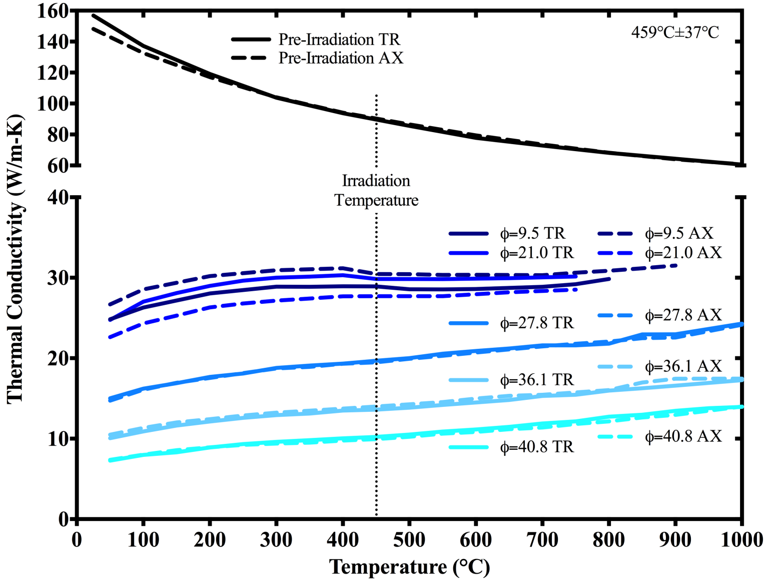
Neutron Fluence and Temperature Effects on Properties

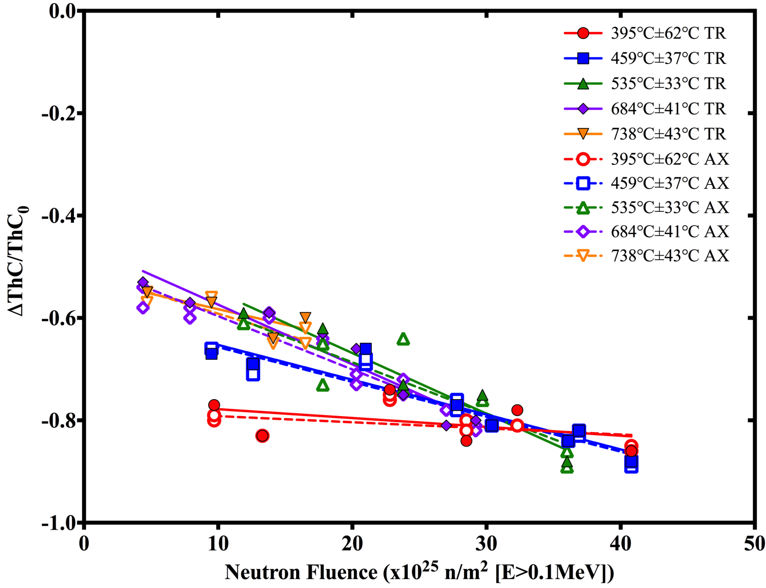
- Thermal conductivity
- Coefficient of thermal expansion
- No density dependence like mechanical properties











Campbell, et al., *Carbon*, 109 (2016) 860-873

Irradiation-Induced Creep

- For most nuclear applications irradiation-induced creep is not desirable
- For graphite the internal stress buildup is a function of neutron fluence and localized temperature
  - Would quickly surpass the UTS of graphite (15-25 MPa)
  - Process is actually internal stress relaxation, unlike irradiation creep of torqued baffle bolts

Irradiation Creep/Stress Relaxation

- Volume change is controlled by irradiation temperature and flux
- In large components the temperature and flux gradients can be quite severe
  - Internal stress formation and buildup
  - Can quickly surpass the graphite strength
- Irradiation-induced creep acts as a stress relaxation mechanism

See Figure 6 from following paper:
Tsang, B.J. Marsden, *Journal of Nuclear Materials*, 350 (2006), 208-220

See Figure 8 from following paper:
Tsang, B.J. Marsden, *Journal of Nuclear Materials*, 350 (2006), 208-220

See Figure 9 from following paper:
Tsang, B.J. Marsden, *Journal of Nuclear Materials*, 350 (2006), 208-220

D.K.L. Tsang, B.J. Marsden, *JNM*, 350 (2006), 208-220

Creep Effects on Dimensional Change

See Figure 42, from the following report:
G. Haag, Properties of ATR-2E Graphite and Property Changes due to Fast Neutron Irradiation", Jül-4183 (2005) available at:
http://juser.fz-juelich.de/record/49235/files/Juel_4183_Haag.pdf

ATR-2E Graphite (WG), T_{irr} = 550ºC, 5 MPa compressive stress

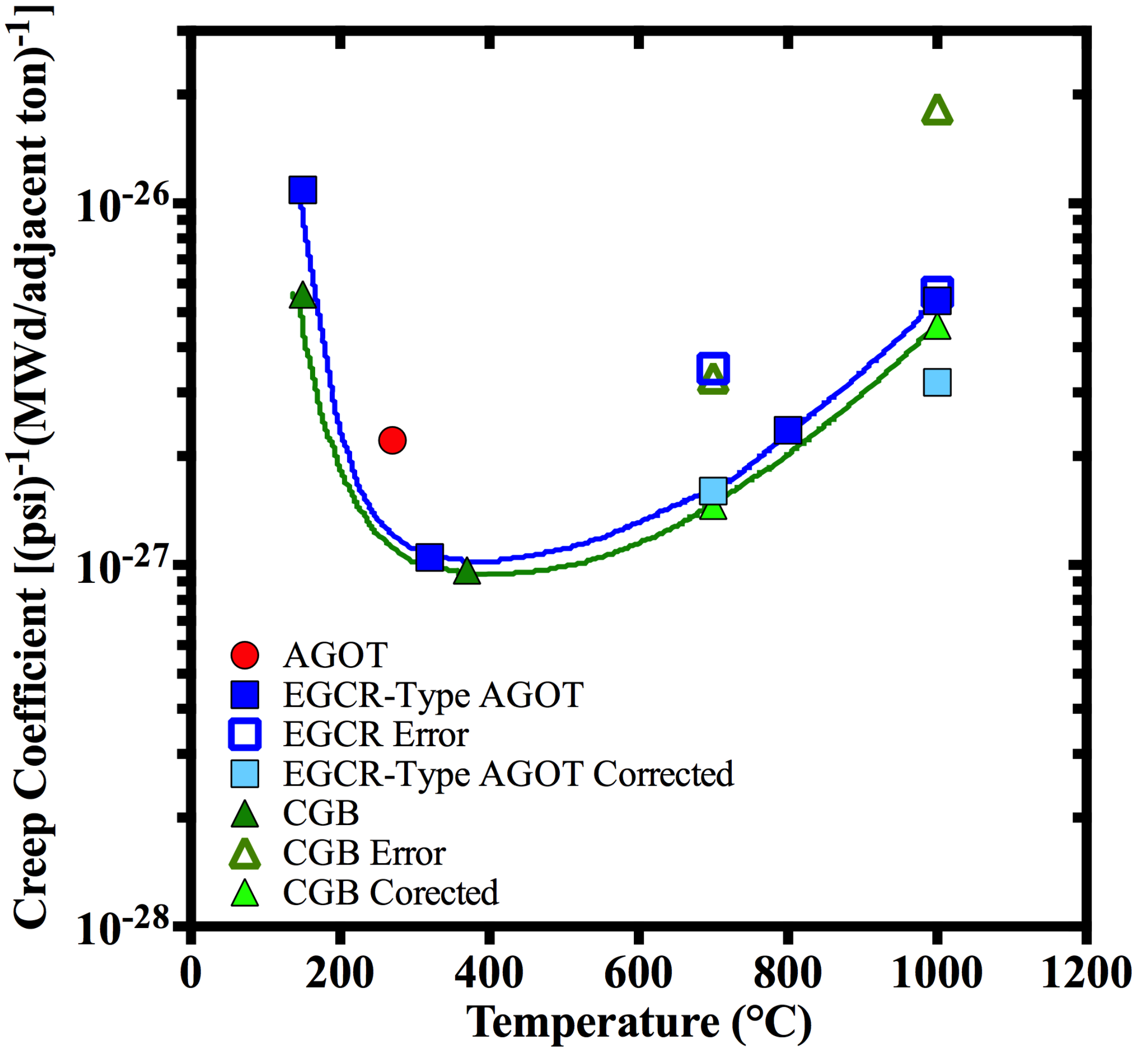
See Figure 43, from the following report:
G. Haag, Properties of ATR-2E Graphite and Property Changes due to Fast Neutron Irradiation", Jül-4183 (2005) available at:
http://juser.fz-juelich.de/record/49235/files/Juel_4183_Haag.pdf

ATR-2E Graphite (WG), T_{irr} = 500ºC, 5 MPa tensile stress

Haag, G., Jül-4183, (2005).

Irradiation Creep Behavior in Graphite

- Complex to measure
  - Need both a stressed specimen and a reference specimen to account for non-stressed dimensional change
- Behavior is also complex
  - Atypical temperature dependence



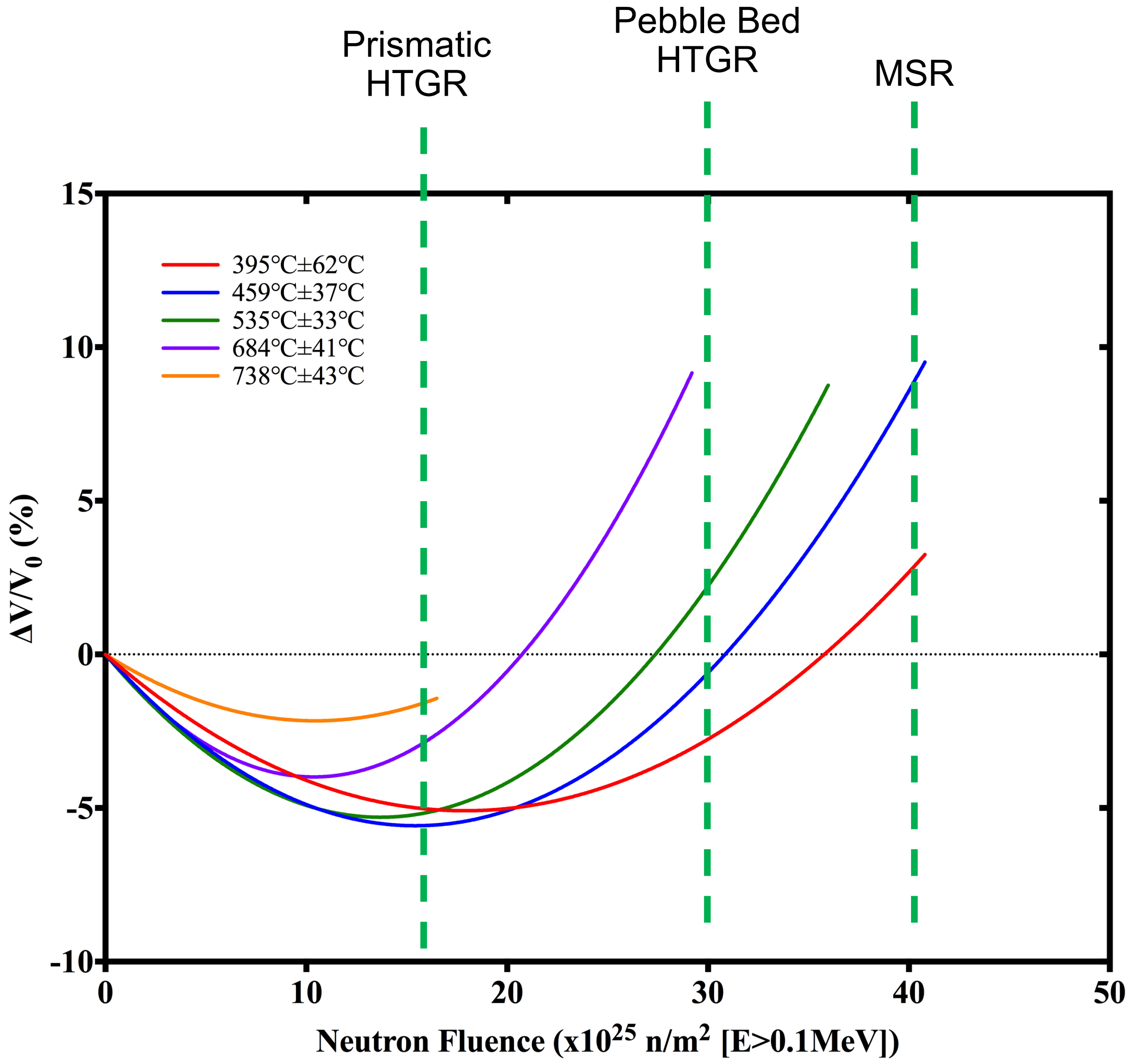
Campbell, *Carbon*, 139 (2018) 279

Lifetime Limiting Questions

- Does graphite need to be removed from the core?
  - Refueling
  - Replacement
- Is strength life limiting property? Is dimensional change life limiting property?
  - ASME B&PV Section III Division 5 Code states "Material that exceeds this fluence limit is considered to provide no contribution to the structural performance (stiffness and strength) of the Graphite Core Component. This fluence limit shall be set to the fluence at which the material experiences a +10% linear dimensional change in the with-grain direction."
  - Maybe final tensile strength should be limit.
- Is life limiting the same for each reactor type?

What Limits Graphite "Lifetime" in Nuclear Reactors?

- Need to remove graphite from core is most likely life limit for prismatic HTGR, since fuel is located in blocks
- Pebble bed HTGR may only need to replace graphite if strength cannot hold up the block stack in the reflector region
- Molten salt lifetime may be when flow channels cannot be maintained



Campbell, et al., *Carbon*, 109 (2016) 860-873

Concerns with Graphite in MSRs

- Salt wetting into pores
  - Localized changes in temperature, mismatch of graphite and salt thermal/physical/mechanical properties
  - Fuel intrusion with salt – makes reactor simulations and source terms difficult to define
  - $LiF-BeF_2$ (Capillary experiments (ORNL-4254))
    - No intrusion in 0.8 mm hole
    - <1 mm intrusion into 1.6 mm hole
    - 1.1 cm intrusion in 3.2 mm hole
    - 0.7 cm intrusion into 6.35 mm hole
  - Treatment of salt with HF or metallic Zr decreased wetting by 30%
- Salt, graphite, and metal reactions/corrosion
- Irradiation effects on graphite size, strength, thermal properties
- Fission gas retention
  - ^{3}H top concern with any salt with Li from $^{6}Li(n,^{3}H)He$ and $^{7}Li(n,^{3}H)He+n$ reactions
  - ^{135}Xe – neutron poison, main concern in breeder reactors
    - Reduce gas permeability below 10^{-8} cm^2/s of He at STP (ORNL-4812)
    - Requires surface pores diameter <0.01μm
    - Multiple surface preparation techniques suggested to reduce permeability
- Salt intrusion may locally effect graphite temperature, strength, and lifetime
  - Localized hot/cold spots reduces graphite lifetime
  - Salt has different physical/mechanical/thermal properties than the graphite

Understanding Graphite Behavior in Nuclear Reactor Environments for Lifetime Predictions

Anne A. Campbell, Ph.D.
Oak Ridge National Laboratory
Oak Ridge, TN
campbellaa@ornl.gov

US-NRC Advanced Non-Light Water Reactors – Materials and Component Integrity Workshop, December 9-11