Electrical, thermal, and mechanical properties of spatially tailored fiber orientations in 3D printed carbon-carbon composites for EFAS/SPS tooling

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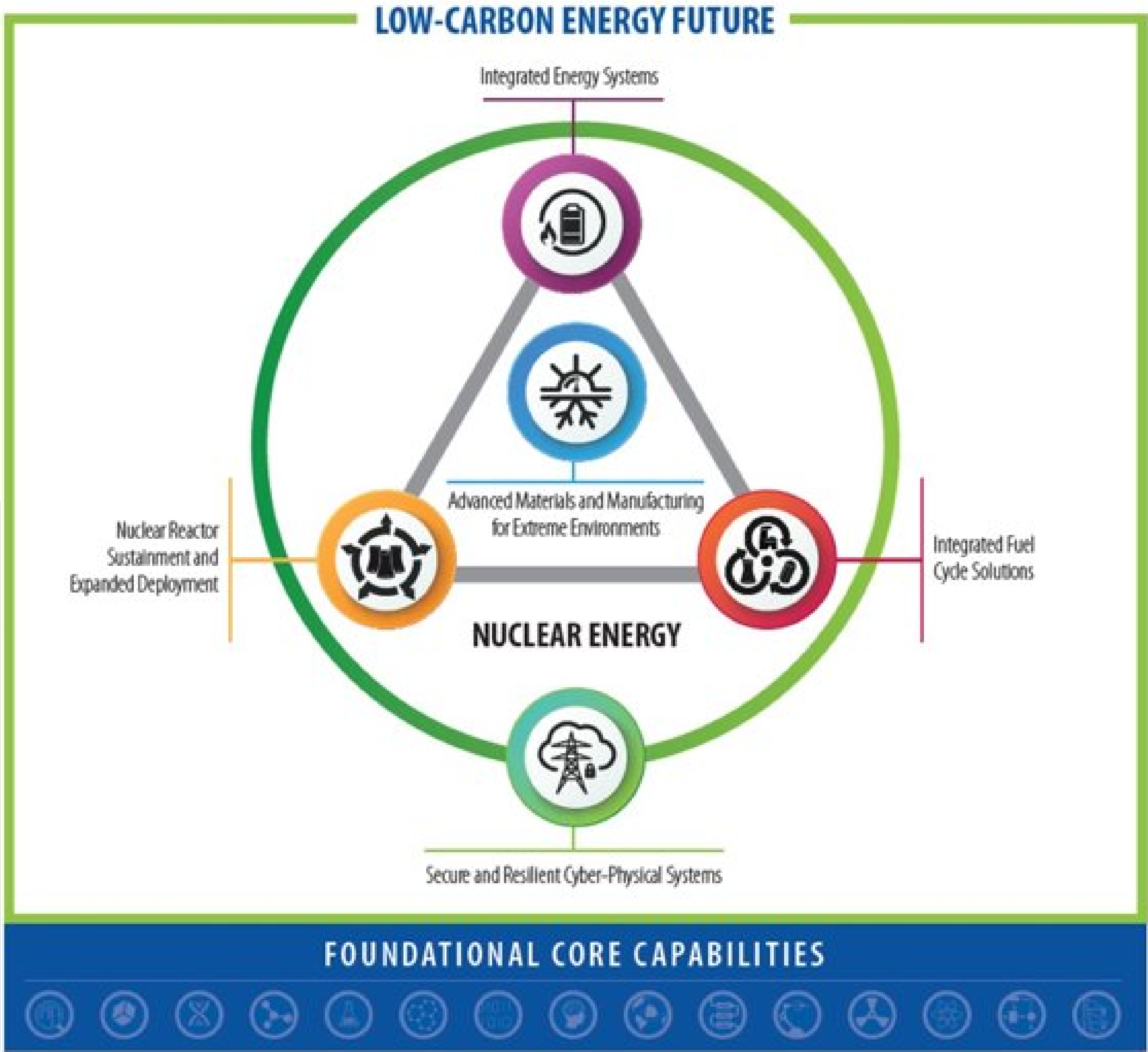
Overview

- Idaho National Laboratory
- EFAS tooling scale-up complications
- Carbon fiber reinforced carbon (C-C) for use as EFAS tooling
- 3D printed C-C anisotropic material properties
  - Engineering for greater efficiency
  - Experimental comparison to G535 graphite tooling.
- Modeling of energy usage and heat profiles
  - Tailoring fiber orientation for advanced thermal control

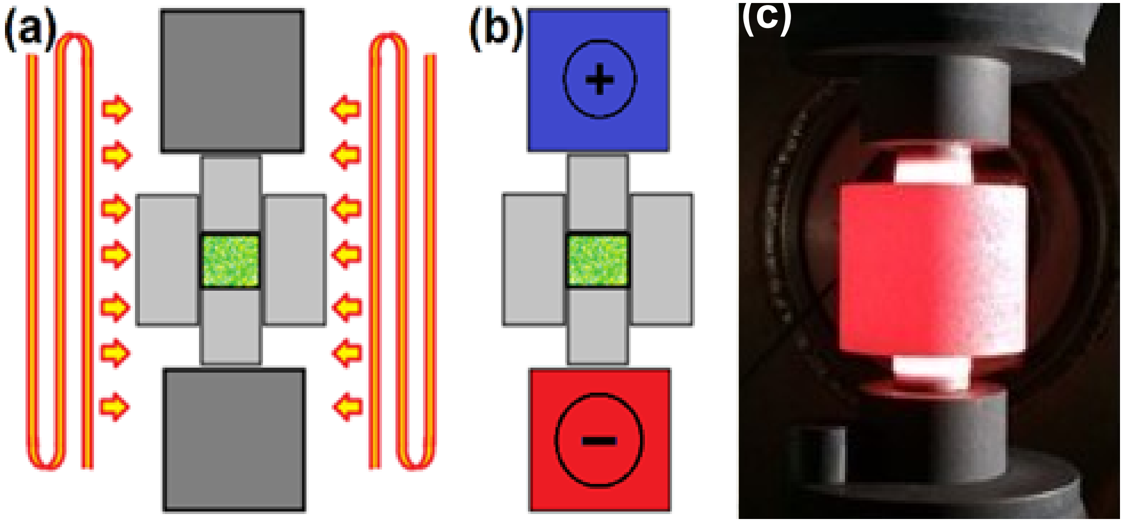
About Idaho National Laboratory

- INL is one of the U.S. Department of Energy's (DOE's) national laboratories. The laboratory performs work in each of DOE's strategic goal areas: energy, national security, science and environment.
- INL's Advanced Manufacturing efforts touch on every aspect of INL's mission.
- Home of the worlds largest experimentally available format of Electric Field Assisted Sintering instrument:
  - 800 tons uniaxial force
  - 150,000 Amps.



Electric Field Assisted Sintering (EFAS)

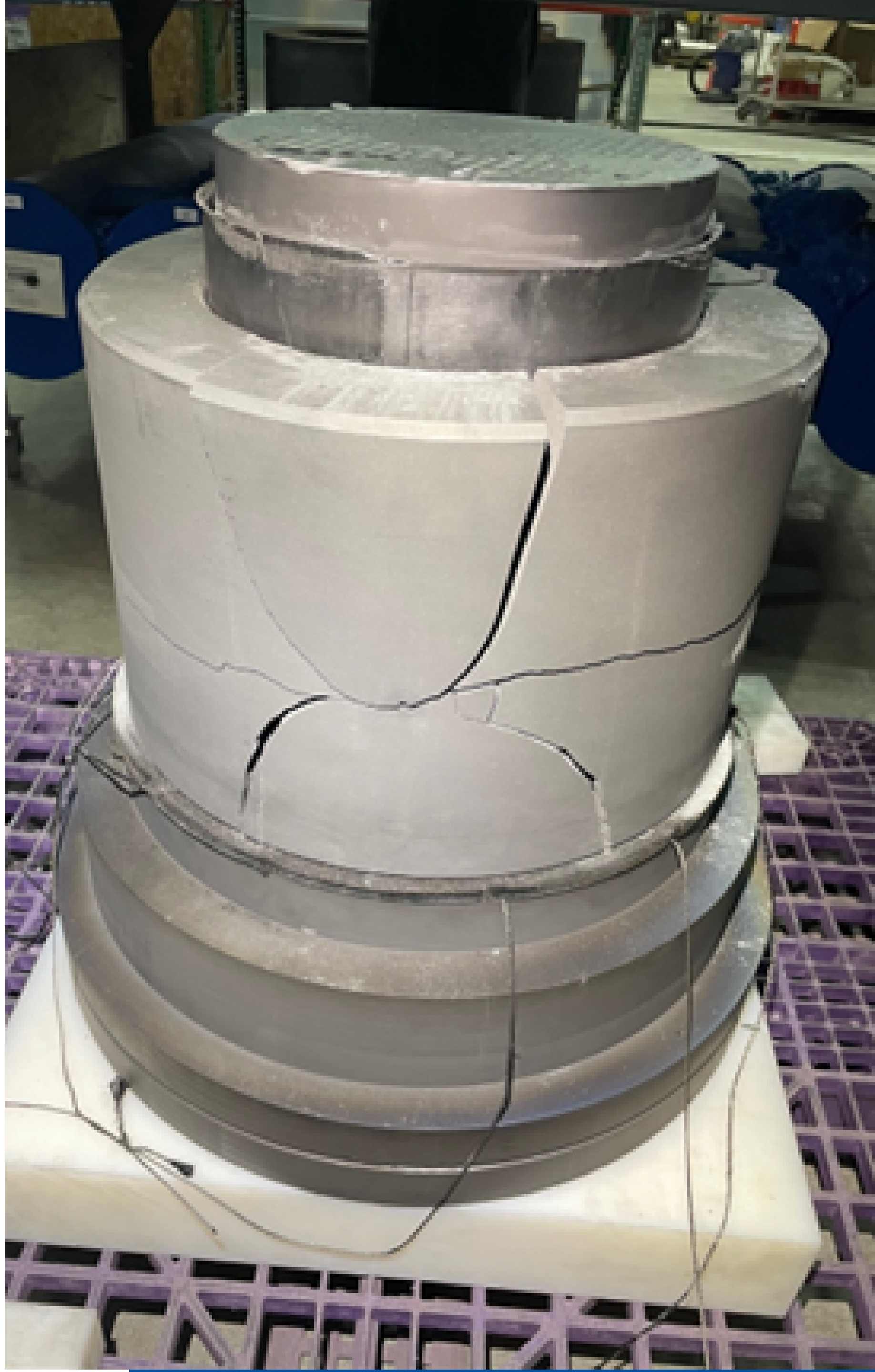
- EFAS, (or SPS) is a highly energy efficient sintering process.
  - Typically enables about 90% energy savings compared to hot pressing.
    (Musa et al. 2009, *J. Clean. Prod*)
    - Faster:
      - Increased ramp rates
      - Less time at temperature
    - More efficient heat localization:
      - Tooling is the heating element.



Schematics of a) Hot pressing, b) EFAS. c) EFAS tooling during heating

Large Scale Graphite Tooling

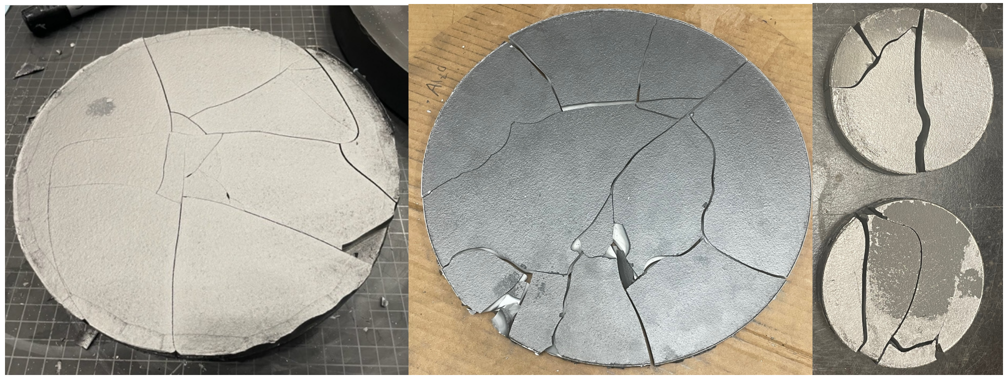
- However, scaling up to industrially relevant sizes brings challenges.
- Large graphite tooling:
  - CTE mismatch
  - Thermal gradients
  - Graphite availability/quality differs at largest sizes
  - LONG procurement lead times
  - Expensive.
  - Fails catastrophically (brittle fracture).



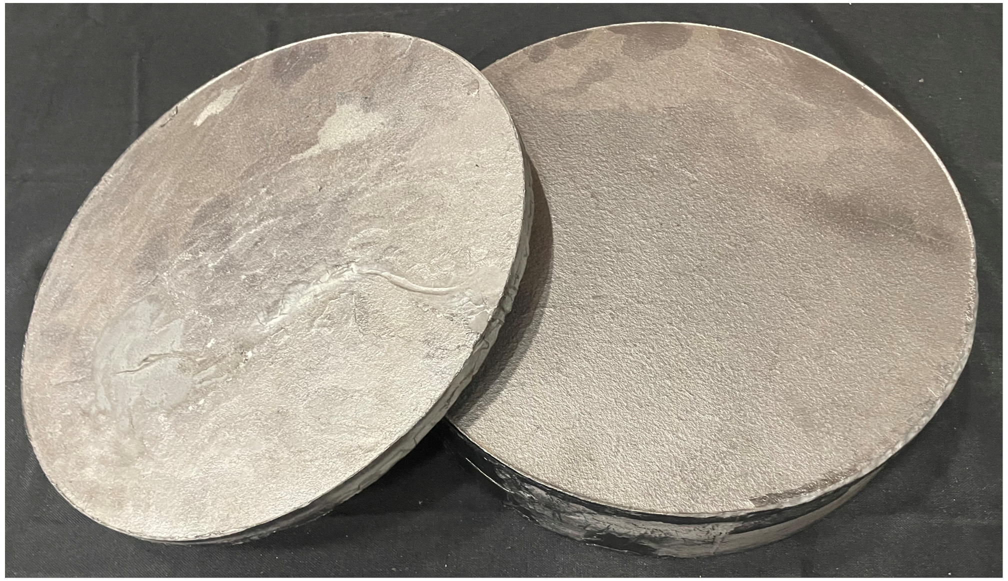
Thermal gradient example

- Poor heat distribution can cause thermal expansion and contraction mismatches which lead to stresses in ceramic parts
  - Has been observed in ceramic and metal parts from 3-12” in diameter
    - Can be solved with graphite tooling, but special processing and tooling configurations must be used.

12” 12” 3”

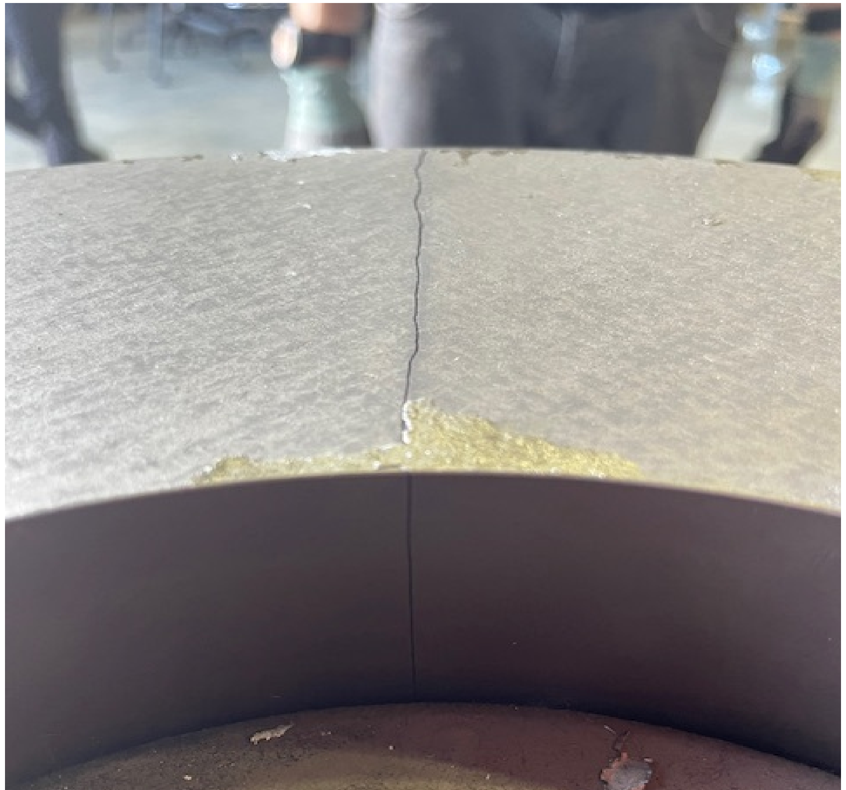


Intact 12” diameter ceramics



CTE mismatch example

- SiC sintered in 12” G535 tooling:
  - G535 Graphite CTE: 5.5 ppm/°C
  - Silicon Carbide CTE: 3-4 ppm/°C
  - SiC final diameter ~12.25”
  - Graphite die split during extraction.

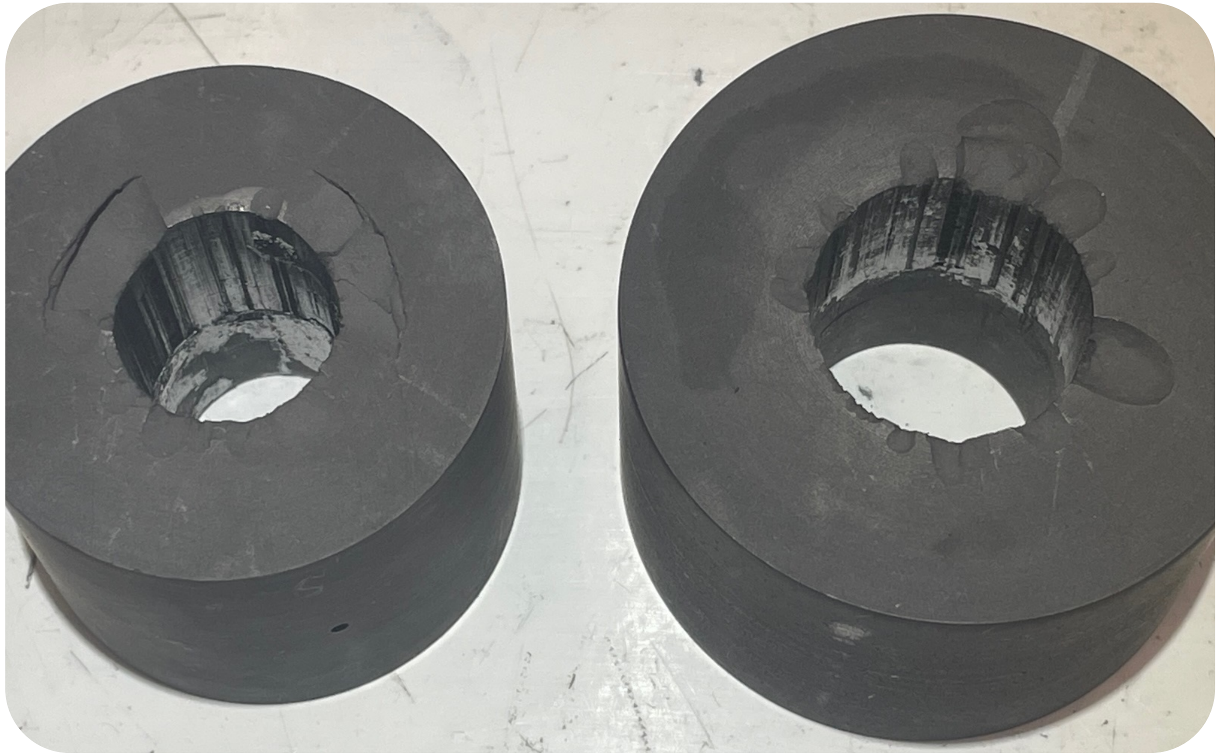


CTE mismatch example 2

- For materials with lower CTE than graphite, (such as SiC):
  - Difficult extraction and excessive tooling wear

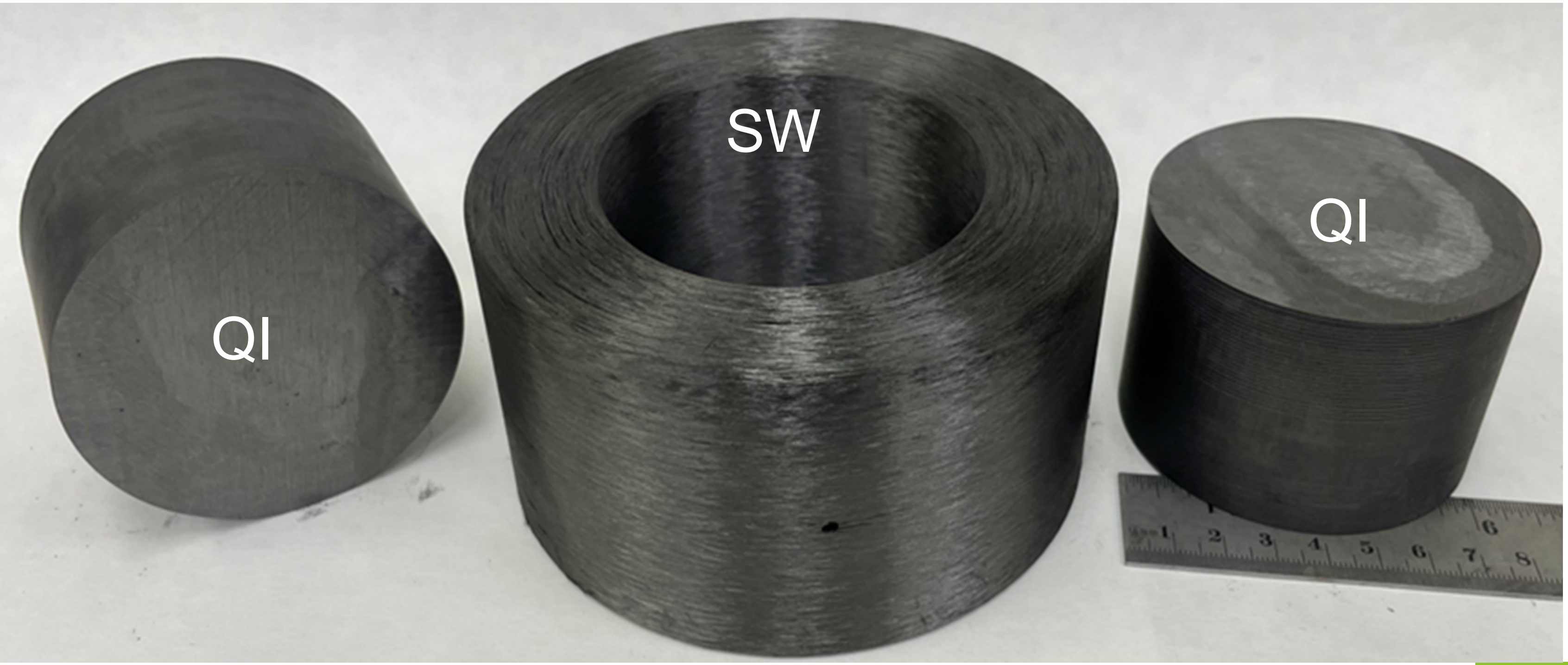
- Carbon fibers have extremely low coefficient of thermal expansion (CTE)
  - Toray T1000 CTE: -0.6 ppm/°C

  This can be very beneficial for a sintering mold, as most materials have higher CTE than C-C.
  - The samples contract more than the die during cooling, and the parts are easily extracted



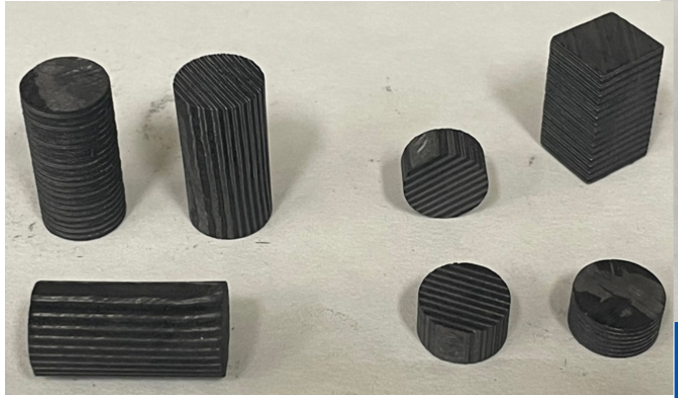
Carbon-Carbon (C-C) for EFAS Tooling

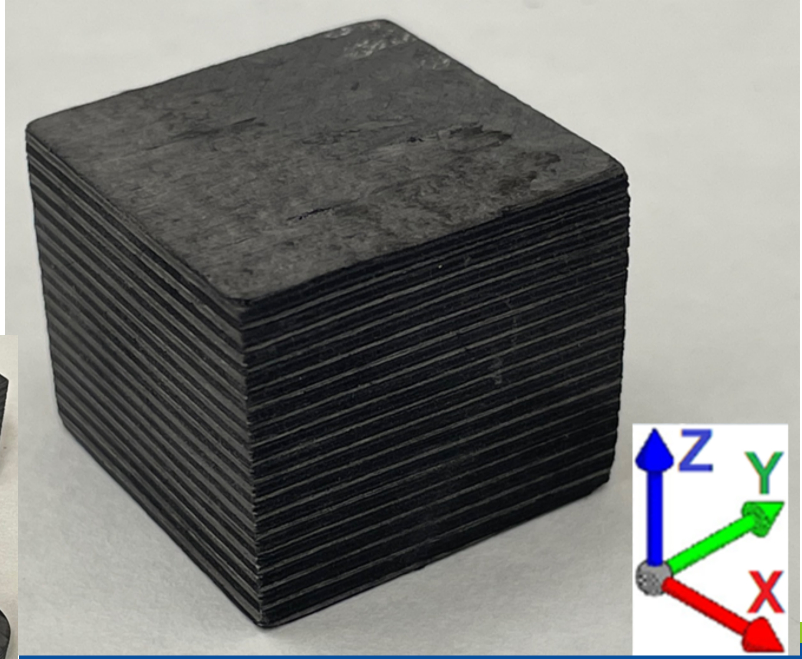
- C-C tooling can be designed for strength and thermal properties:
  - Fiber directionality determines mechanical, thermal, and electrical properties
  - Spirally wound (**SW**) fiber orientation for high hoop strength (dies)
  - Quasi-Isotropic (**QI**) fiber layering for high compressive strength (punches)



Carbon Fiber Reinforced Carbon (C-C)

- C-C was produced from continuous fiber 3D printed preforms
  - Continuous Composites, Coeur D'Alene, ID.
- Anisotropic material properties **with fiber axis**.
- Resistivity (ρ): [G535 Graphite: **17** μΩ*m]
  - **16.5** μΩm in X/Y (along fiber axes)
  - **121.2** μΩm in Z (across fiber diameters)
- Thermal Diffusivity (α): [G535 Graphite: **63** mm²/s]
  - **75** mm²/s in X/Y
  - **5** mm²/s in Z
- Mechanical properties greater than or equal to G535 Graphite

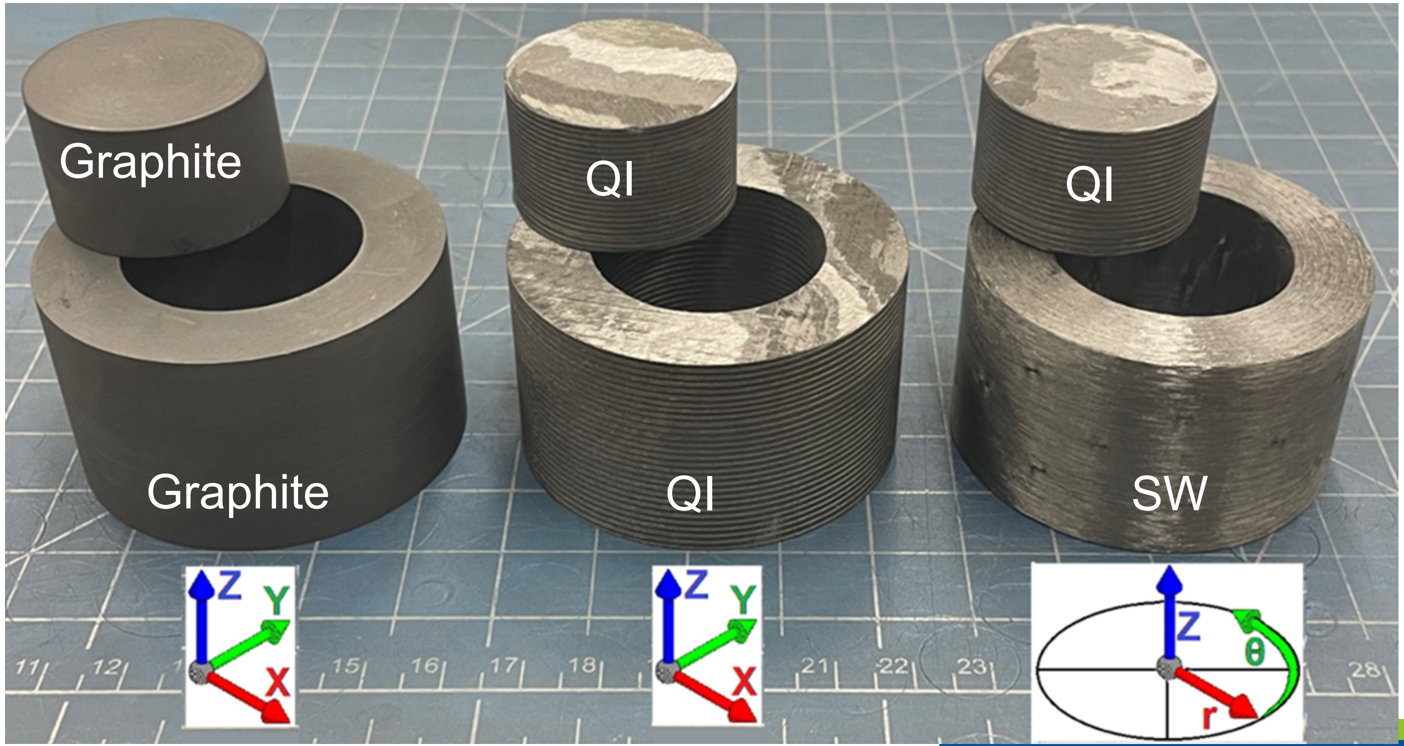




Engineering High Efficiency C-C Tooling for EFAS

- Joule heating equation: (H=heat, I=current, R=resistance)
  - Higher resistance = lower current required for a given heat.
- Resistance: (ρ=resistivity, L=length, A=cross-sectional area)
- Higher resistivity more efficiently converts current to heat.

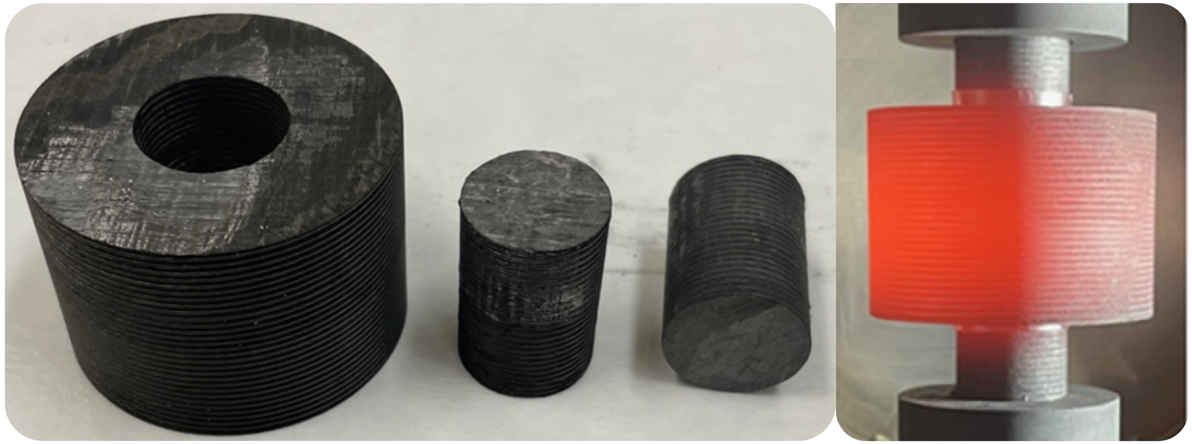
- Graphite
  - Uniform properties
  - X=Y=Z: Low resistivity
- Quasi-Isotropic Die
  - X=Y: Low resistivity
  - Z: **High resistivity**
- Spirally Wound Die
  - Θ: Low resistivity
  - Z=r: **High resistivity**



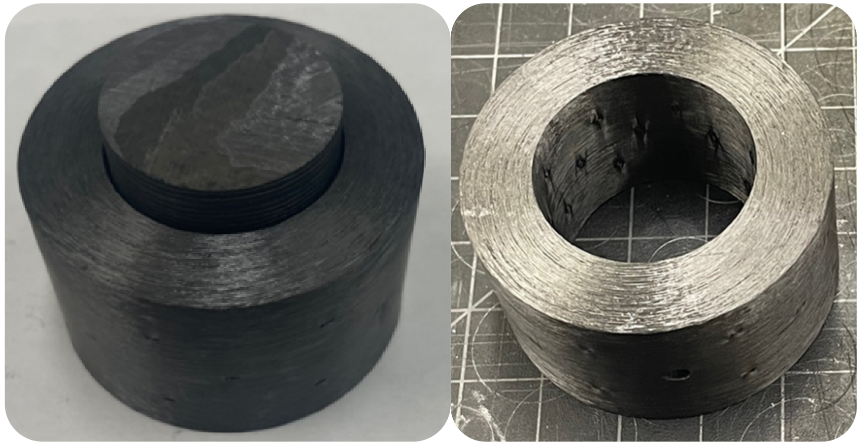
Engineering High Efficiency C-C Tooling for EFAS

- The high resistivity Z-direction of the C-C, when aligned with the axis of current flow, more effectively generates heat.
- Low thermal diffusivity of the Z-direction traps heat where it is generated

20 mm QI
C-C EFAS tooling:

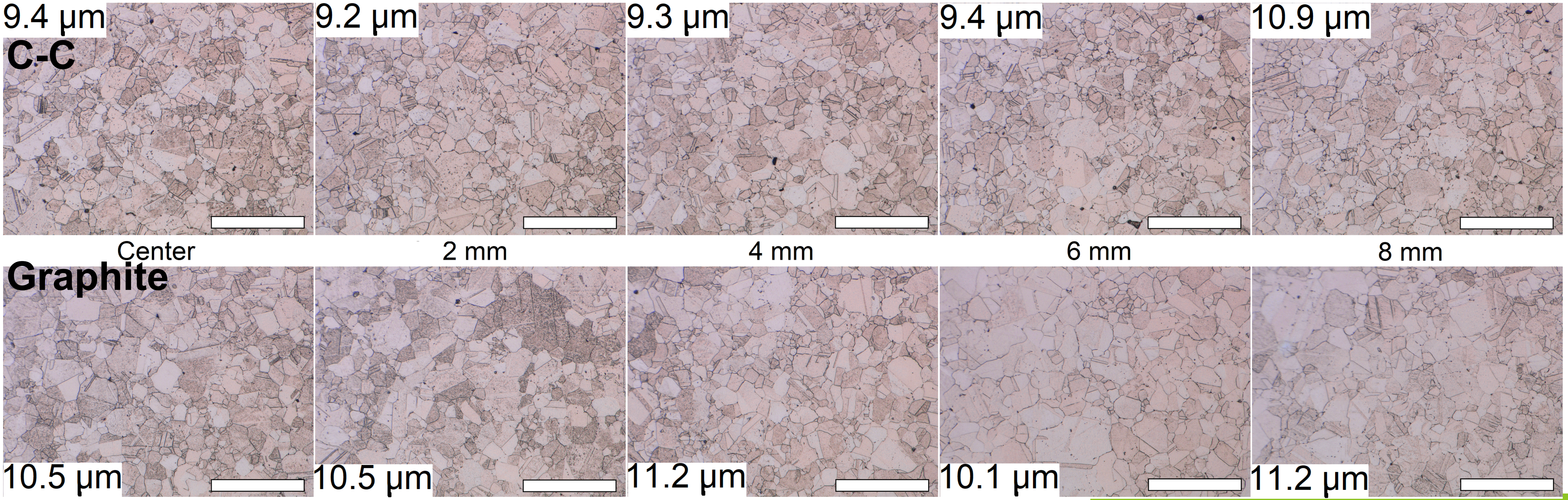


32 mm SW
C-C EFAS Die:
(punches remain QI type)



Engineering High Efficiency C-C Tooling for EFAS

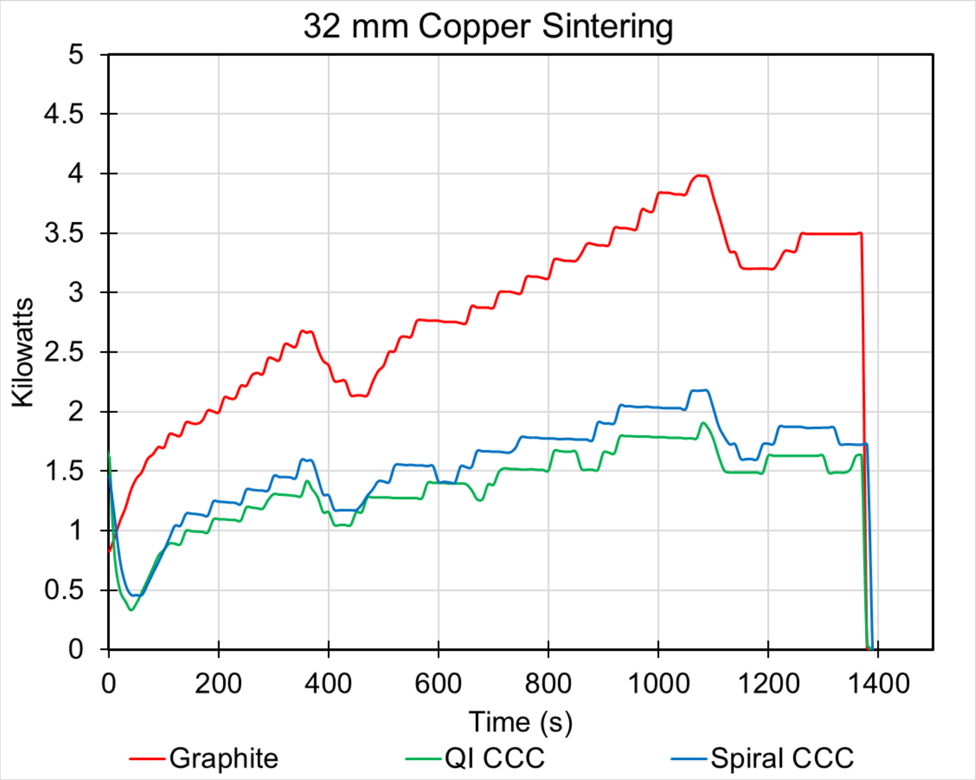
- High thermal diffusivity in the X/Y plane ensures temperature uniformity across the tooling
  - Copper microstructure and average grain sizes from center to edge
  - Top row: C-C tooling. Bottom row: G535 tooling.

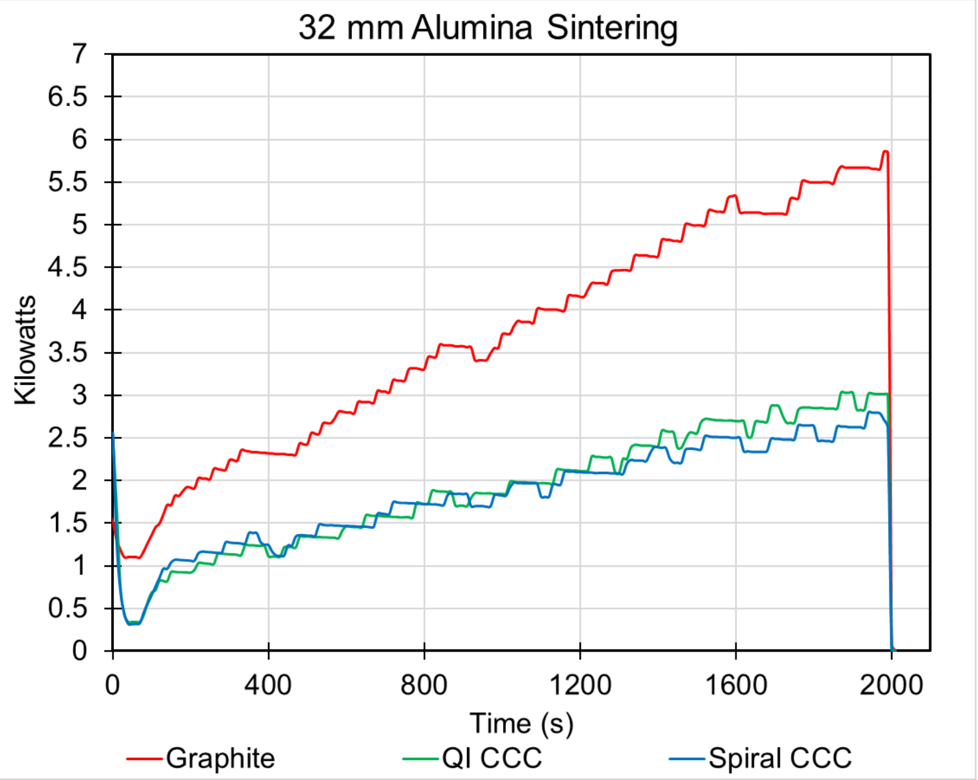


Carbon-Carbon Composite vs. Standard Graphite Tooling

20 and 32mm diameter tooling

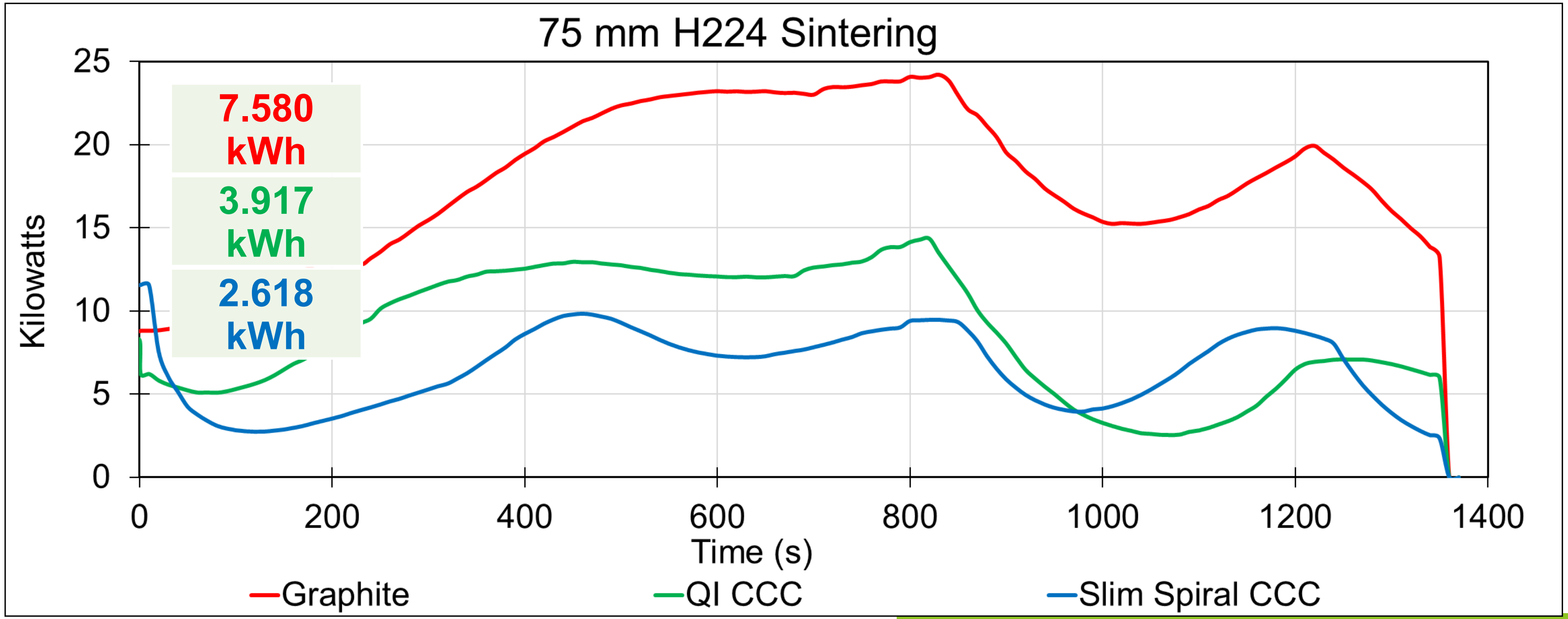
Average 48.3% less energy. Ram temperatures stay **37.3%** colder.





Carbon-Carbon Composite vs. Standard Graphite Tooling

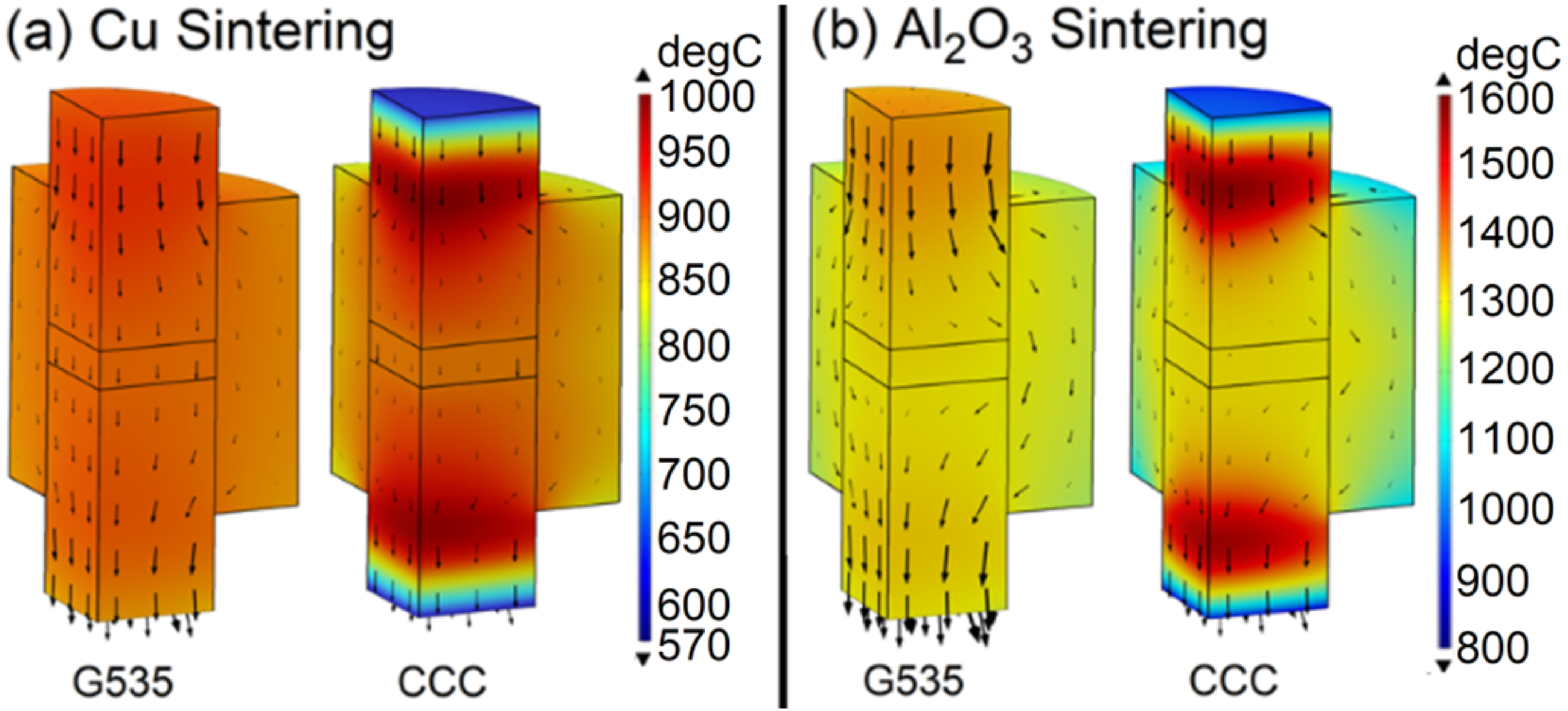
- 75 mm QI die made with same dimensions as graphite tooling.
- 75 mm SW die made with slimmer walls further reduces energy consumption:
  - QI compared to graphite reduces energy by 48.3%, Ram temp by 36.1%
  - **Slim SW** compared to graphite **reduces energy usage by 65.5%,** Rams by 51.0%



Carbon-Carbon Composite vs. Standard Graphite Tooling

Graphite vs C-C, with conductive and insulating sample types
Arrows indicate current density and pathway in parts

Cu
Sample region
@ 900 °C



1290 A

550 A

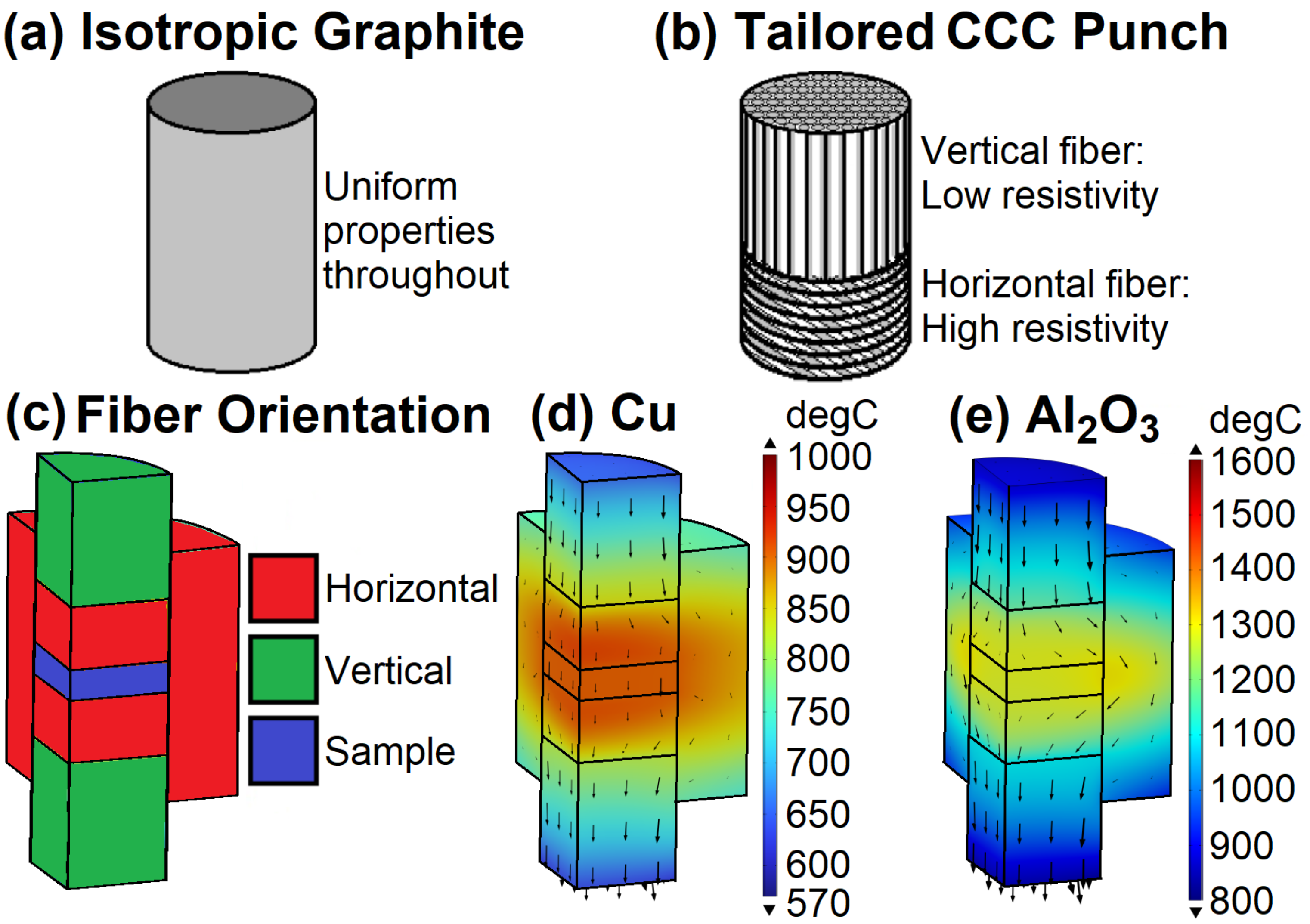
1500 A

783 A

Al_2O_3
Sample region
@ 1300 °C

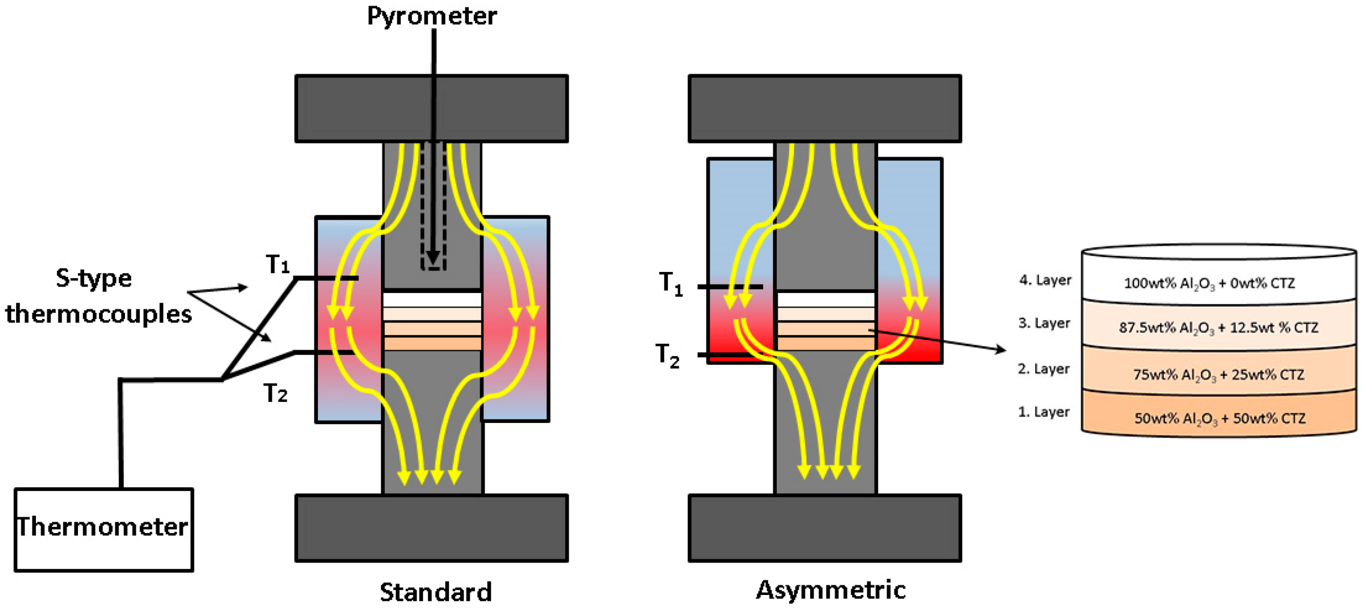
Advanced Possibilities – thermal management

- Leverage 3D printing capabilities:
  - Control current pathways
  - Further isolates the heated zone
- COMSOL modeling of
  - 32 mm simple tailored C-C EFAS punches in a QI Die



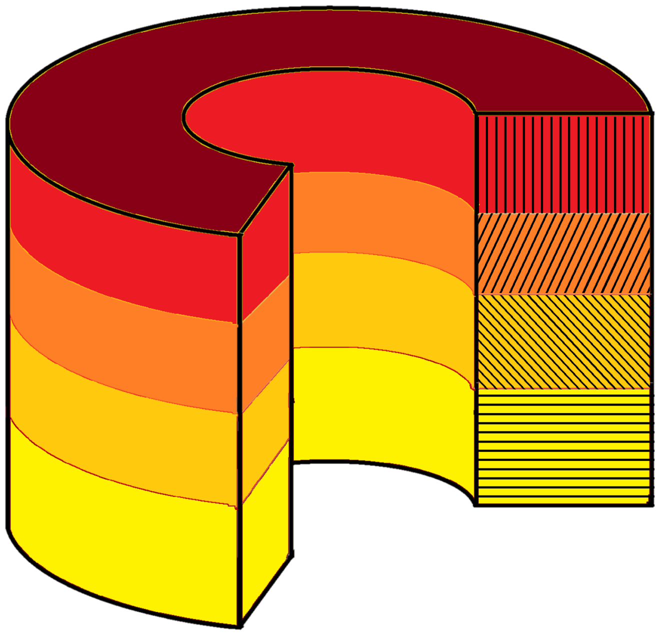
Advanced Possibilities – tailored gradients

Creating a thermal gradient
in standard tooling:



Bodis et al. 2022, *Materials.*

Tailored Anisotropy
Thermal Gradient Die



Functionally graded Al_2O_3 / CTZ (CeO$_2$ stabilized ZrO$_2$)

Summary

- Using C-C synthesized from continuous fiber 3D printed preforms, tooling was fabricated, tested, and compared to graphite.
- Leveraged the anisotropic properties to make stronger and more energy efficient tooling.
- Future work
  - Tailoring the fiber orientations to create targeted heating zones, thermal gradients.
  - Run and evaluate 150 mm SW tooling

150mm C-C tooling

Battelle Energy Alliance manages INL for the U.S. Department of Energy's Office of Nuclear Energy. INL is the nation's center for nuclear energy research and development, and also performs research in each of DOE's strategic goal areas: energy, national security, science and the environment.

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Supplementary Slides

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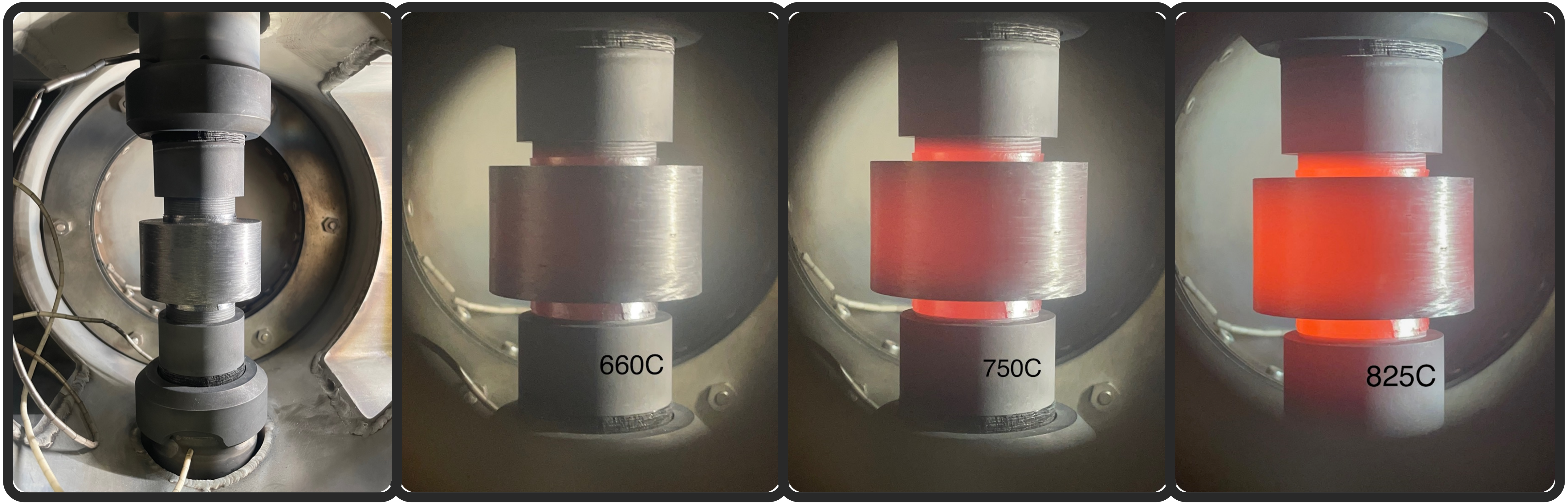
32 mm Spiral Die Heating; Cu Sintering

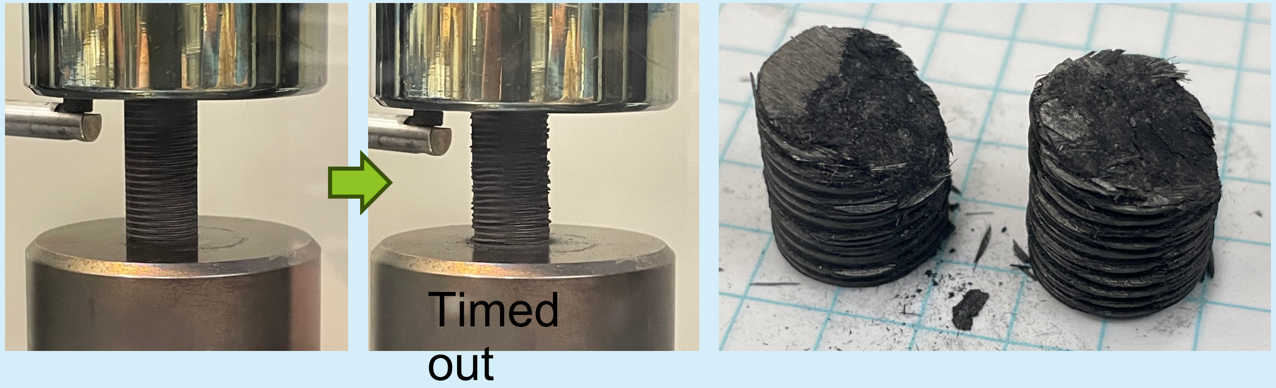


Mechanical Properties

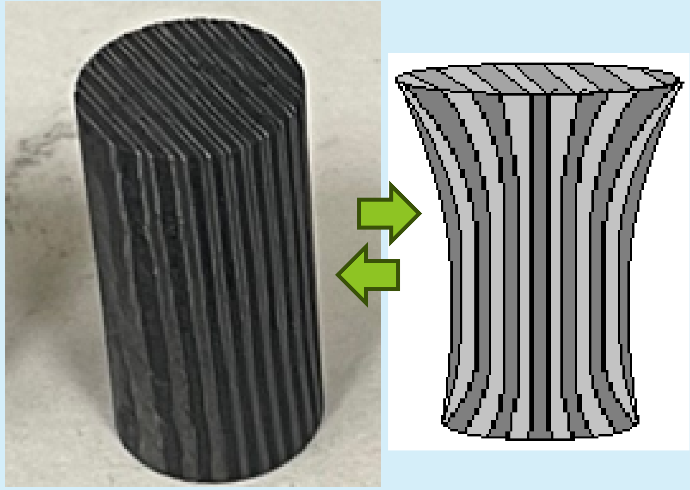
- Compression test (Z-axis) #1
  - Timed out but greater than 100 MPa
- Compression test (Z-axis) #2
  - Sample tilt error. >155 MPa
- Compression tests (X/Y)
  - Delamination failure at 48 MPa.
- Tension tests slipped in the gripping fixtures. >80 MPa
  - Carbon fibers have tensile strengths from 3.5 to 6 GPa.
  - 25% of the fibers in the tensile sample are aligned to the tension axis.
  - Reasonable to assume tensile strength is much higher than 80 MPa.

Z Compression

Sample tilted (not placed properly)

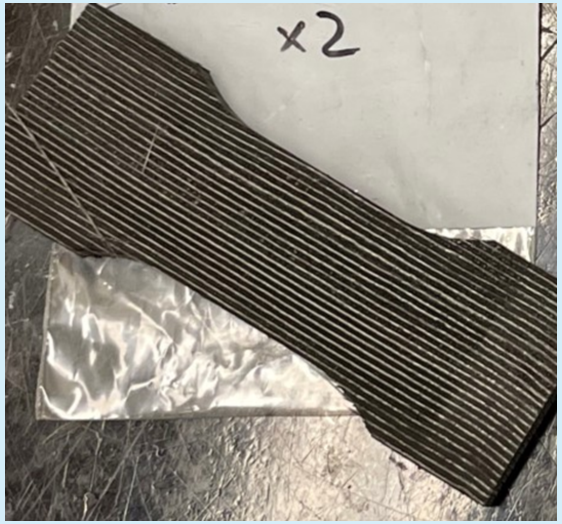


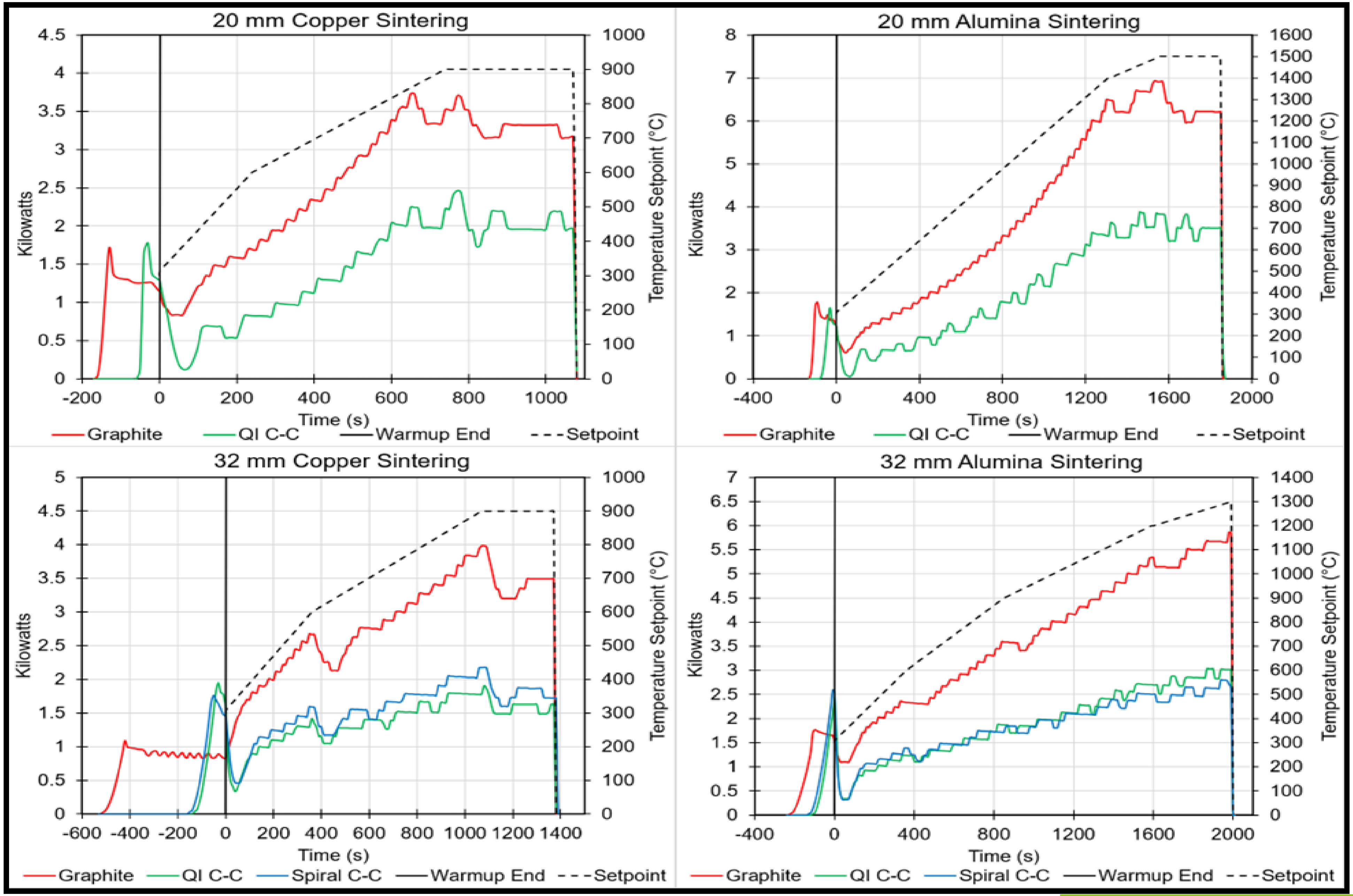
X/Y Compression



Failed at 48 MPa: delaminated, layers spring back after anvil removed.

Tensile




20 mm Copper Sintering
Kilowatts
Temperature Setpoint (°C)
Time (s)
Graphite
QI C-C
Warmup End
Setpoint
20 mm Alumina Sintering
Kilowatts
Temperature Setpoint (°C)
Time (s)
Graphite
QI C-C
Warmup End
Setpoint
32 mm Copper Sintering
Kilowatts
Temperature Setpoint (°C)
Time (s)
Graphite
QI C-C
Spiral C-C
Warmup End
Setpoint
32 mm Alumina Sintering
Kilowatts
Temperature Setpoint (°C)
Time (s)
Graphite
QI C-C
Spiral C-C
Warmup End
Setpoint