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Sandia National Laboratories

# Stress Dependent Phase Transition in Cristobalite Containing Glass-Ceramics

*Presented by:*
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2022 Glass and Optical Materials Division
Baltimore, MD
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U.S. DEPARTMENT OF ENERGY | NNSA

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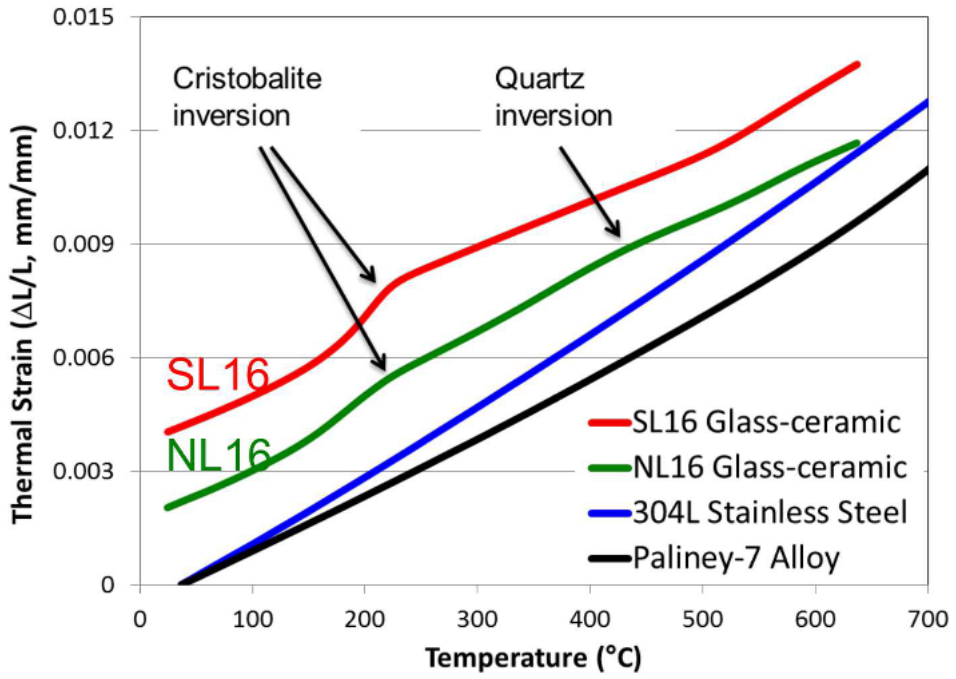
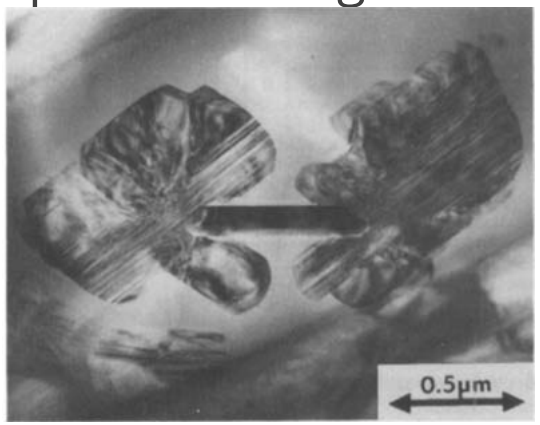
# Outline

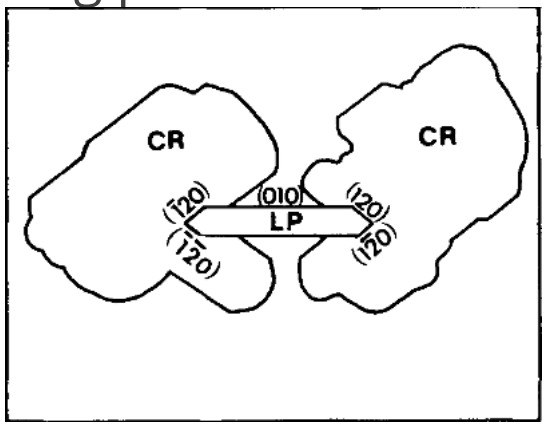
- Motivation
- Background
- Experimental Procedure
  - Glass-ceramic processing
  - Material Characterization
    - Microstructure – SEM
    - Phase Analysis – XRD
    - Phase Transition – DSC and Thermal Expansion
  - Applied pressure thermal expansion Measurements
- Results
- Discussion
- Summary and Future Work

# Motivation – Glass Ceramic-to-Metal Seals

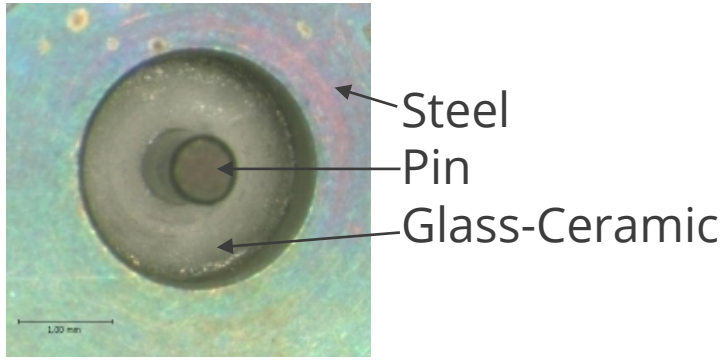
- Lithium alumino-silicate glasses are known to form high CTE phases such as Quartz and Cristobalite.
- Ability to tailor the CTE by controlling the thermal profile during the sealing process.
  - Ideal for creating a matched CTE GCtM seal.



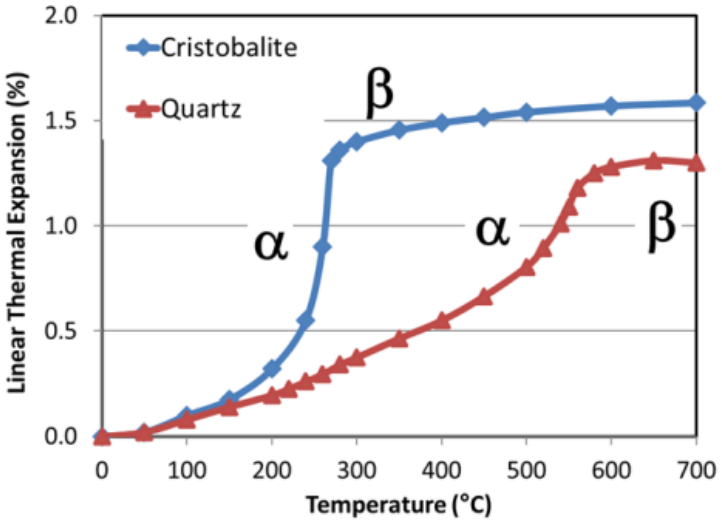

Headley, T. J., and R. E. Loehman. "Crystallization of a glass - ceramic by epitaxial growth." *J. Amer. Cer. Soc.* **67** [9] (1984) p. 620-625.



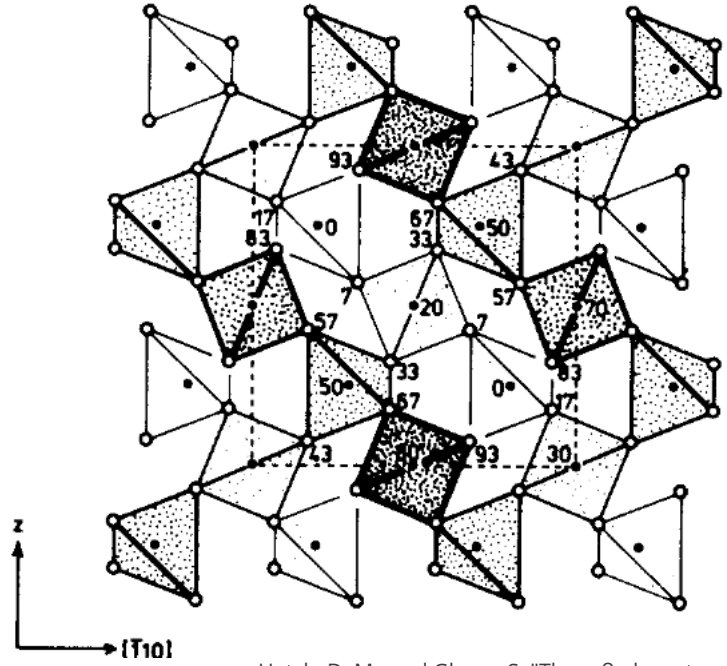

Question? – Can we model the non-linear behavior of the Glass-Ceramic? Does applied stresses cause a change in the non-linear behavior?

# Background – Cristobalite Phase Transition

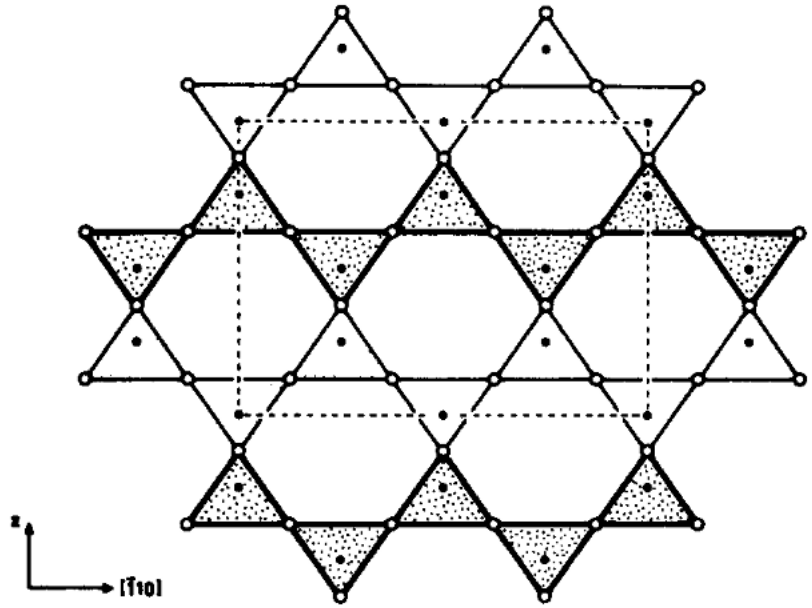
- Cristobalite goes through a first-order phase transition that is reversable.
- Several mechanisms can cause the phastransition temperature to shift.
  - Chemical impurities, crystallographic defects and applied stress.




α - tetragonal




β - cubic




Hatch, D. M., and Ghose, S. "The α-β phase transition in cristobalite, SiO2." *Phys. and Chem. of Min.* **17** [6] (1991) p 554-562.

# Experimental Procedure Sample Fabrication

## Glass Composition

| Oxide | wt% | | | mol% |
|---|---|---|---|---|
| | Min | Max | Target | |
| $SiO_2$ | 65 | 80 | 74.35 | 69.44 |
| $B_2O_3$ | 0.5 | 7 | 1.20 | 0.97 |
| $Li_2O$ | 8 | 16 | 12.70 | 23.85 |
| $Al_2O_3$ | 2 | 8 | 3.80 | 2.09 |
| $K_2O$ | 1 | 8 | 2.95 | 1.76 |
| $P_2O_5$ | 1 | 5 | 3.15 | 0.62 |
| ZnO | 0 | 5 | 1.85 | 1.28 |

Glass network former

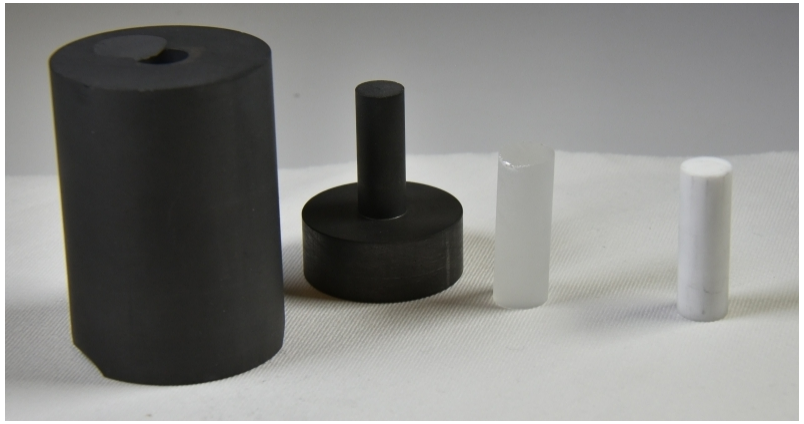
Glass network modifier

→ $Li_3PO_4$ nuclei for crystallization

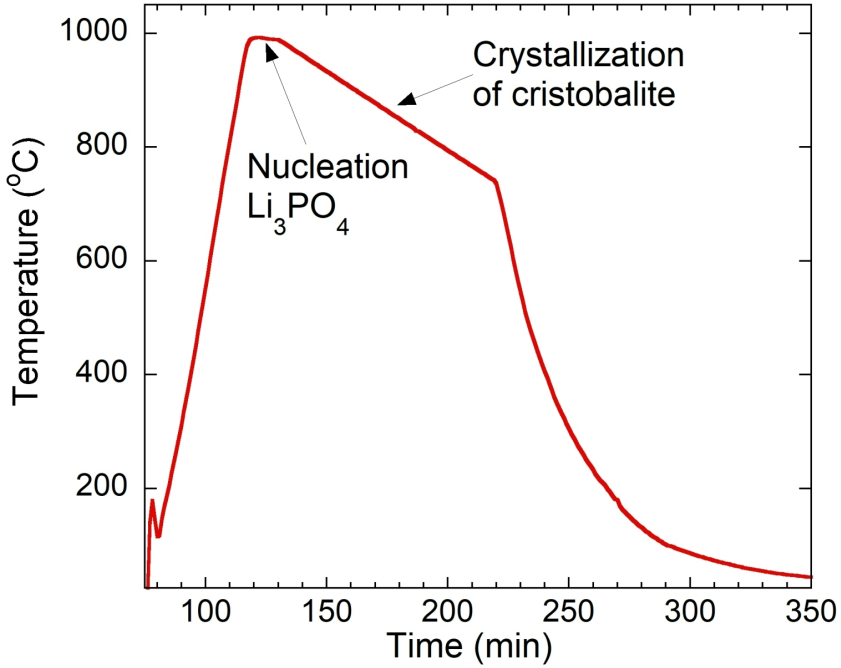
→ Corrosion resistance

| Constituent | α (x10$^{-6}$ mm/mm-°C) for 20-600°C |
|---|---|
| Cristobalite | 27.1 |
| Lithium Metasilciate | 13 (20-300°C) |
| Lithium Disilicate | 11 |
| Quartz | 23.3 |
| Tridymite | 14.4 |

Sample Dimensions:
9.53mm OD x 25 mm H



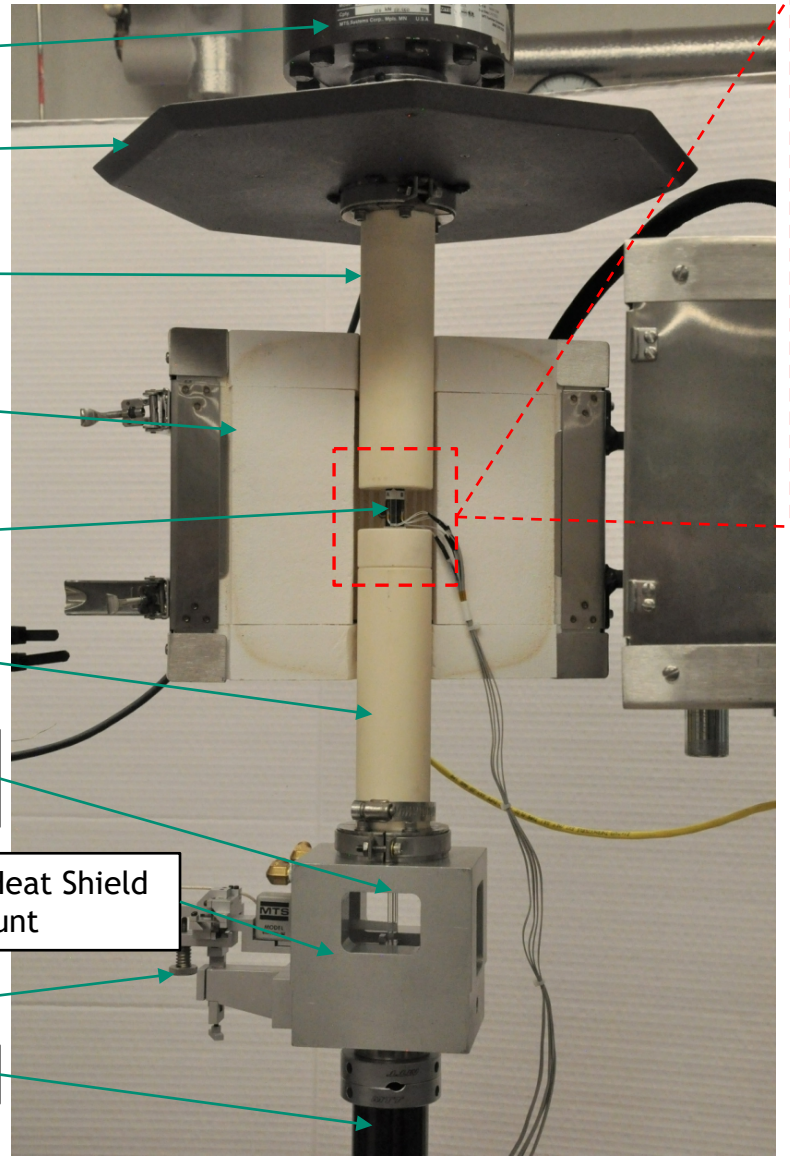
Temperature Profile

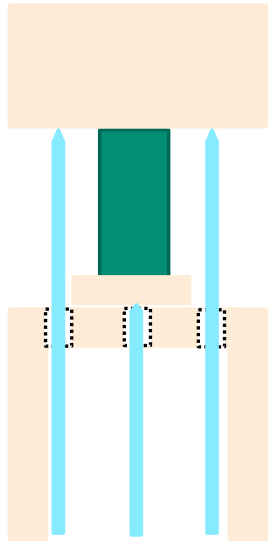


# Experimental Procedure – Material Characterization

- XRD
  - 5 - 100° 2θ (0.02 ° 2θ step size and 10 second dwell)
  - Pseudo-Void Fit, Rietveld Analysis
- SEM (General Microstructure Observation)
- DSC (TA Instruments DSC Q2000)
  - 10 °C/min
- Thermal Expansion (Netzsch TMA )
  - 1 °C/min

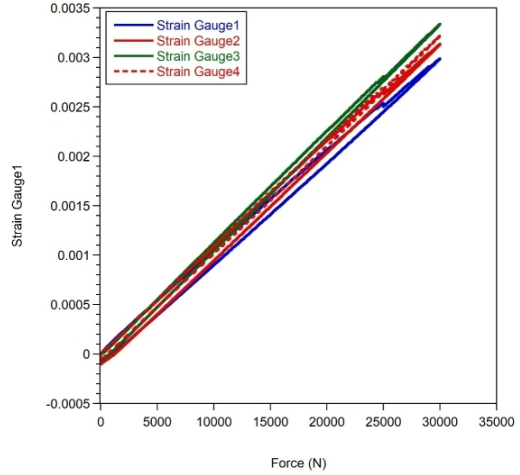
# Experimental Procedure – Pressure Expansion Measurements



## Alignment

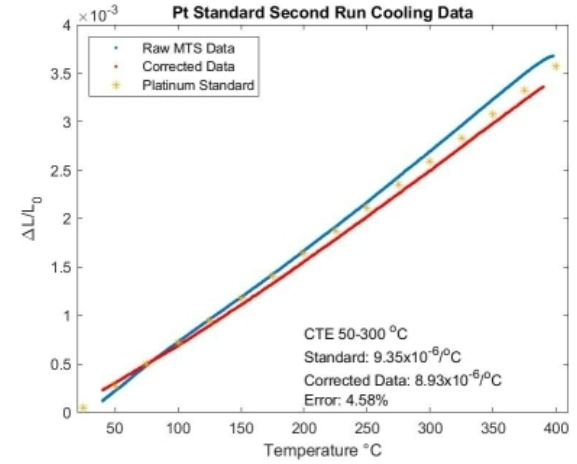
Glass Cylinder with 4 strain gauges.




## Correction and Verification

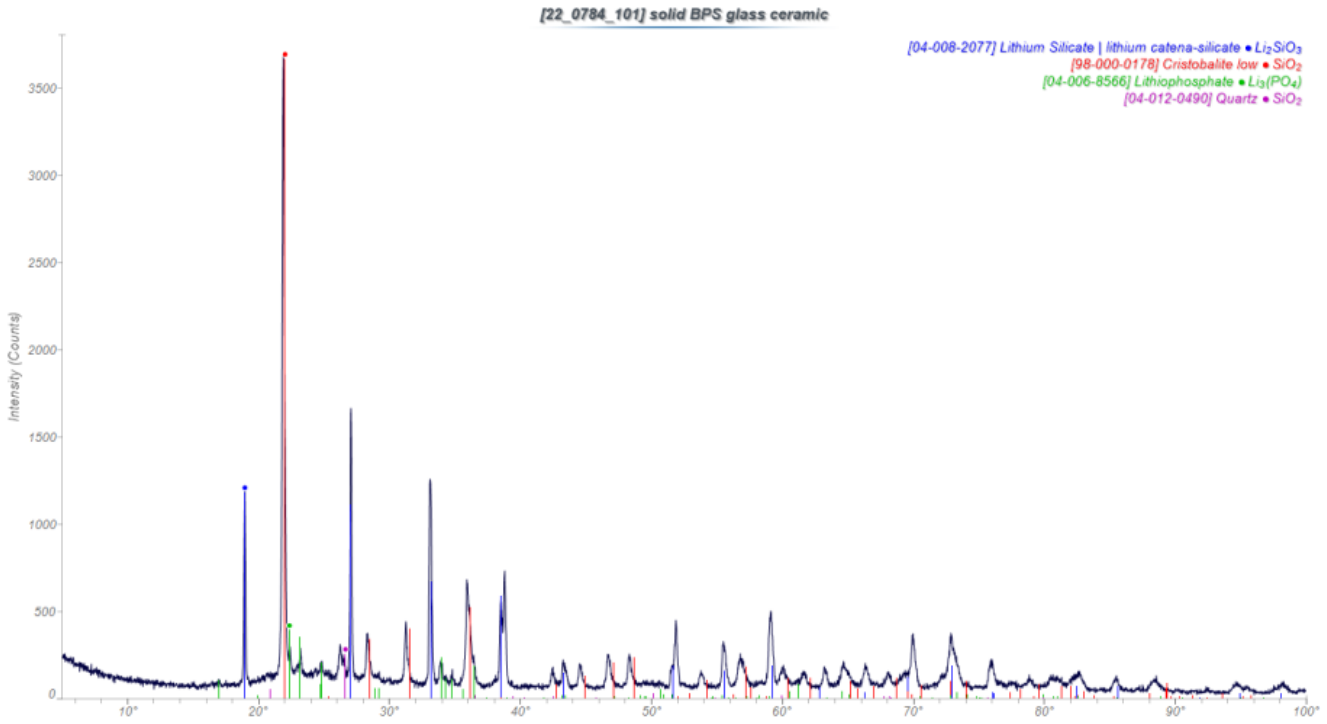
$$\frac{\Delta L}{L_0}(T)_{Correction} = \frac{\Delta L}{L_0}(T)_{\substack{Raw \\ Data\ Std.}} - \frac{\Delta L}{L_0}(T)_{\substack{Known \\ Standard}}$$

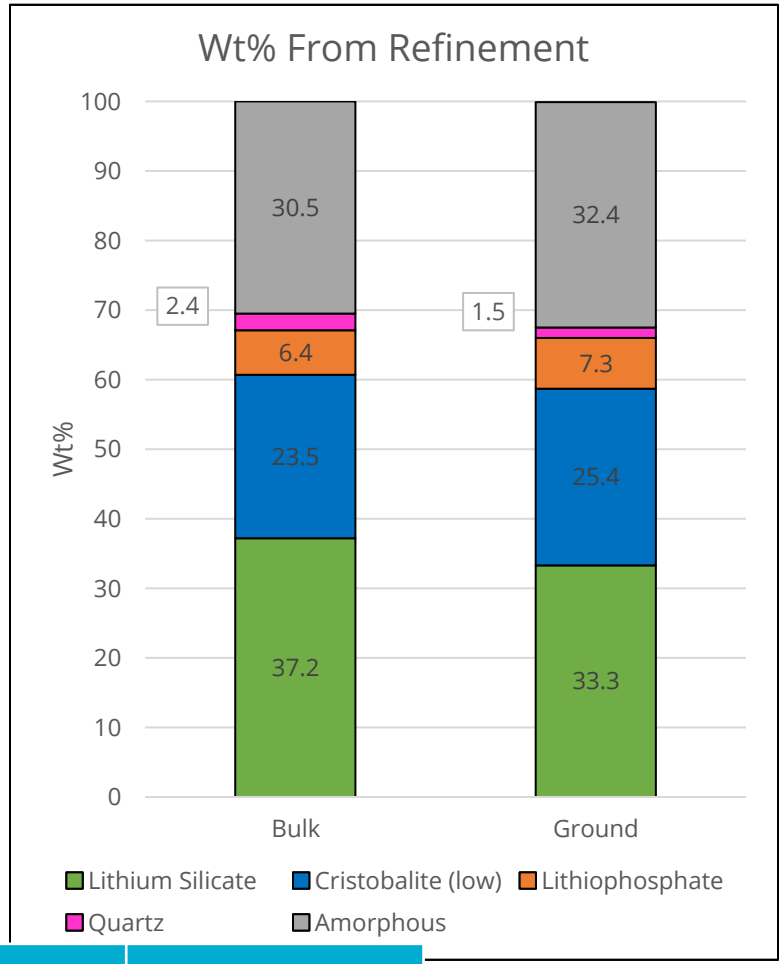
$$\frac{\Delta L}{L_0}(T)_{\substack{Corected \\ Raw\ Data}} = \frac{\Delta L}{L_0}(T)_{\substack{Raw \\ Data}} - \frac{\Delta L}{L_0}(T)_{Correction}$$




| | CTE $10^{-6}$/°C | Error (%) |
|---|---|---|
| Heat | 9.01 | 3.65 |
| Cool | 8.93 | 4.58 |
| Standard | 9.35 | - |

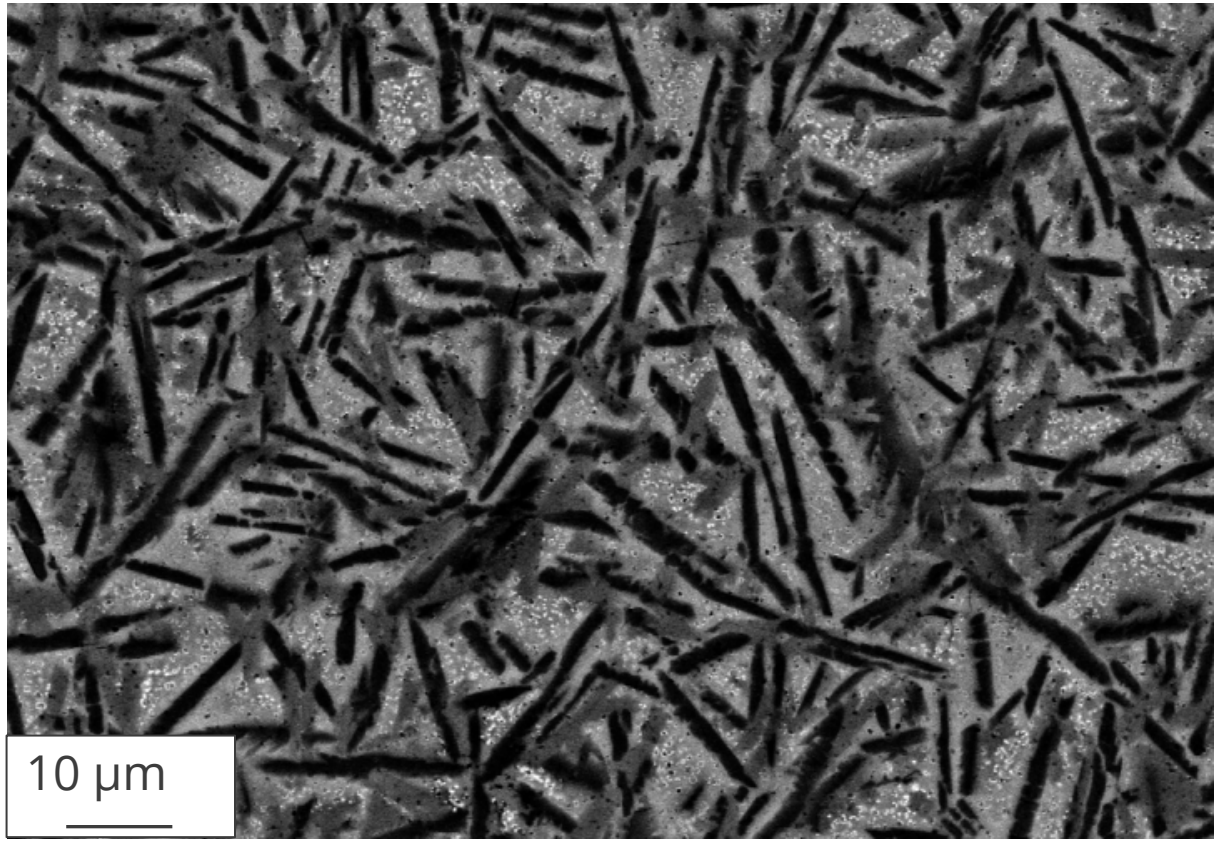
# Results- XRD

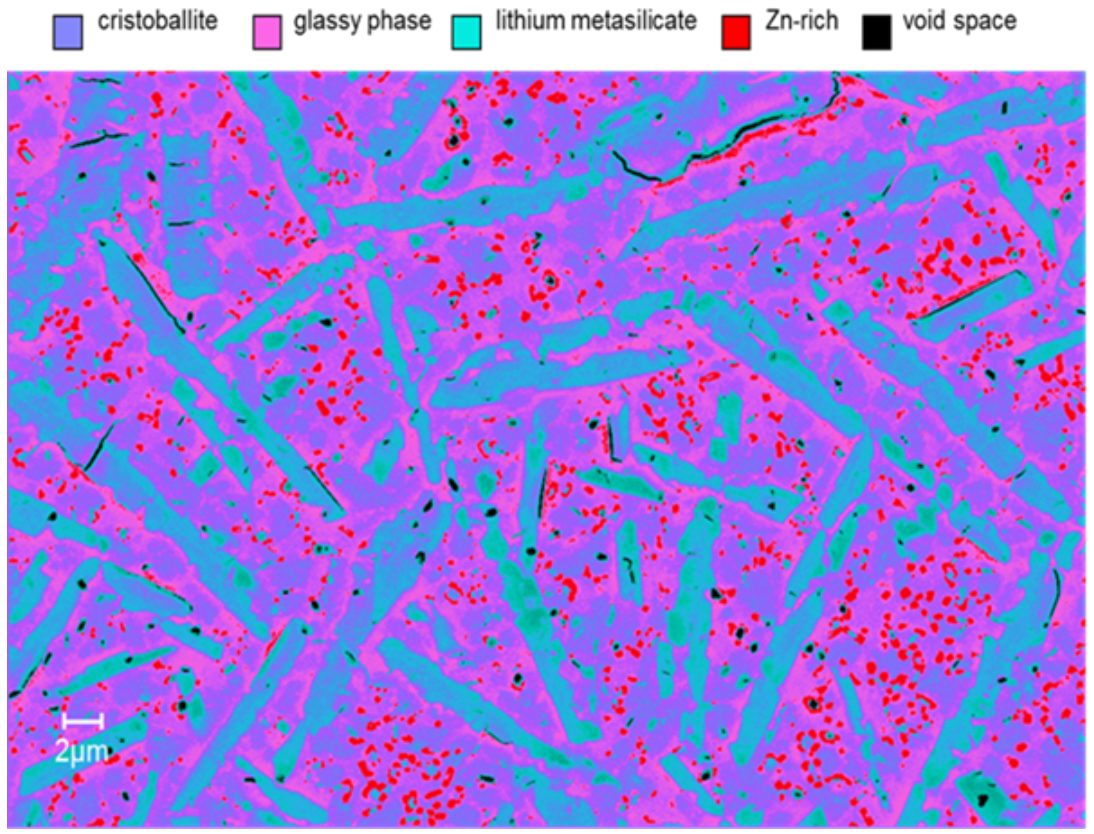



| Phase | Stoichiometry | Wt.% |
|---|---|---|
| Lithium Silicate | $Li_2SiO_3$ | 37.2 |
| Cristobalite | $SiO_2$ | 23.5 |
| Lithium Orthophosphate | $Li_3(PO_4)$ | 6.4 |
| Quartz | $SiO_2$ | 2.4 |
| Residual Glass | - | 30.5 |

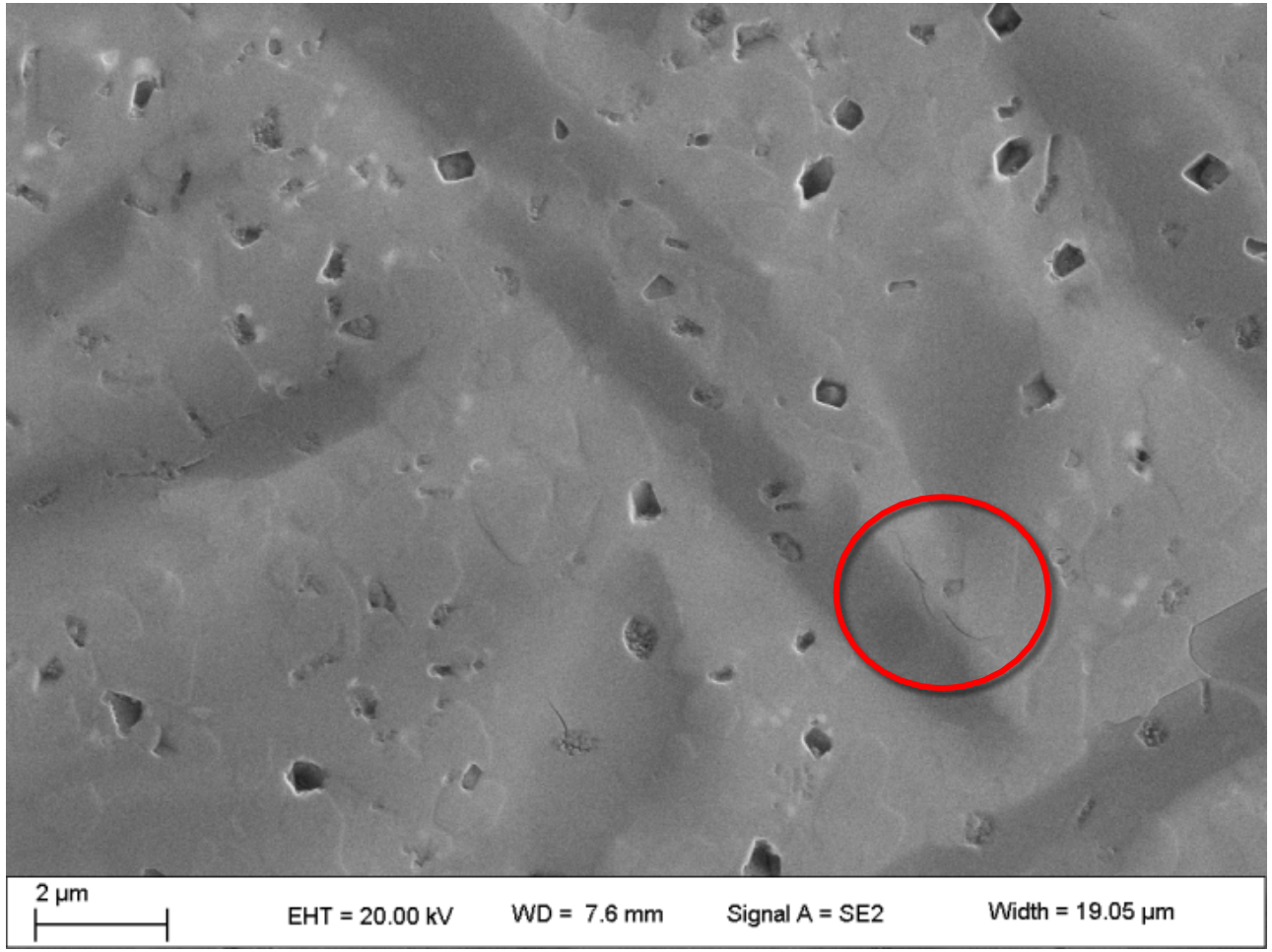
# Results- SEM




Ref. SAND report

# SEM - Microstructure



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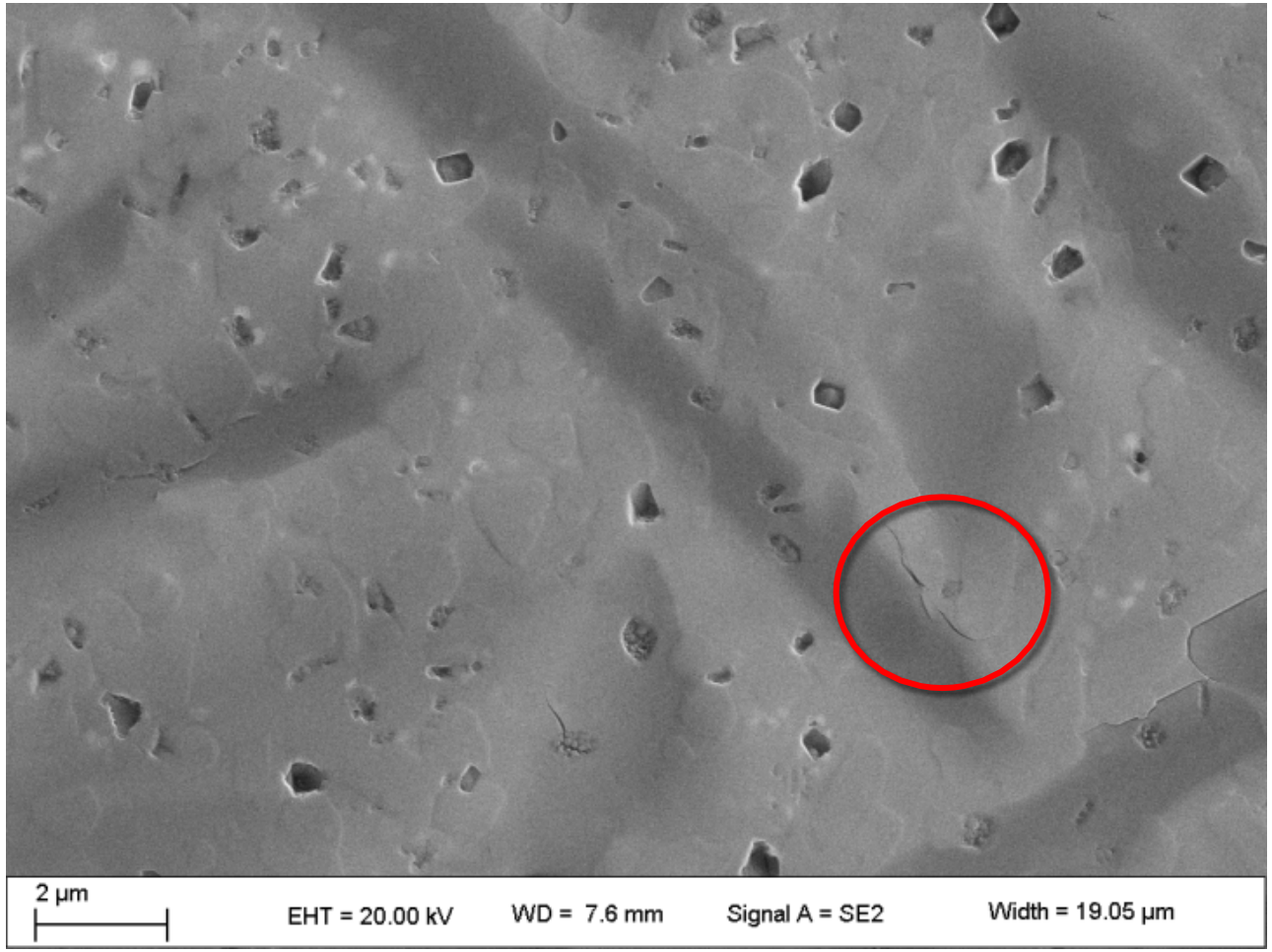
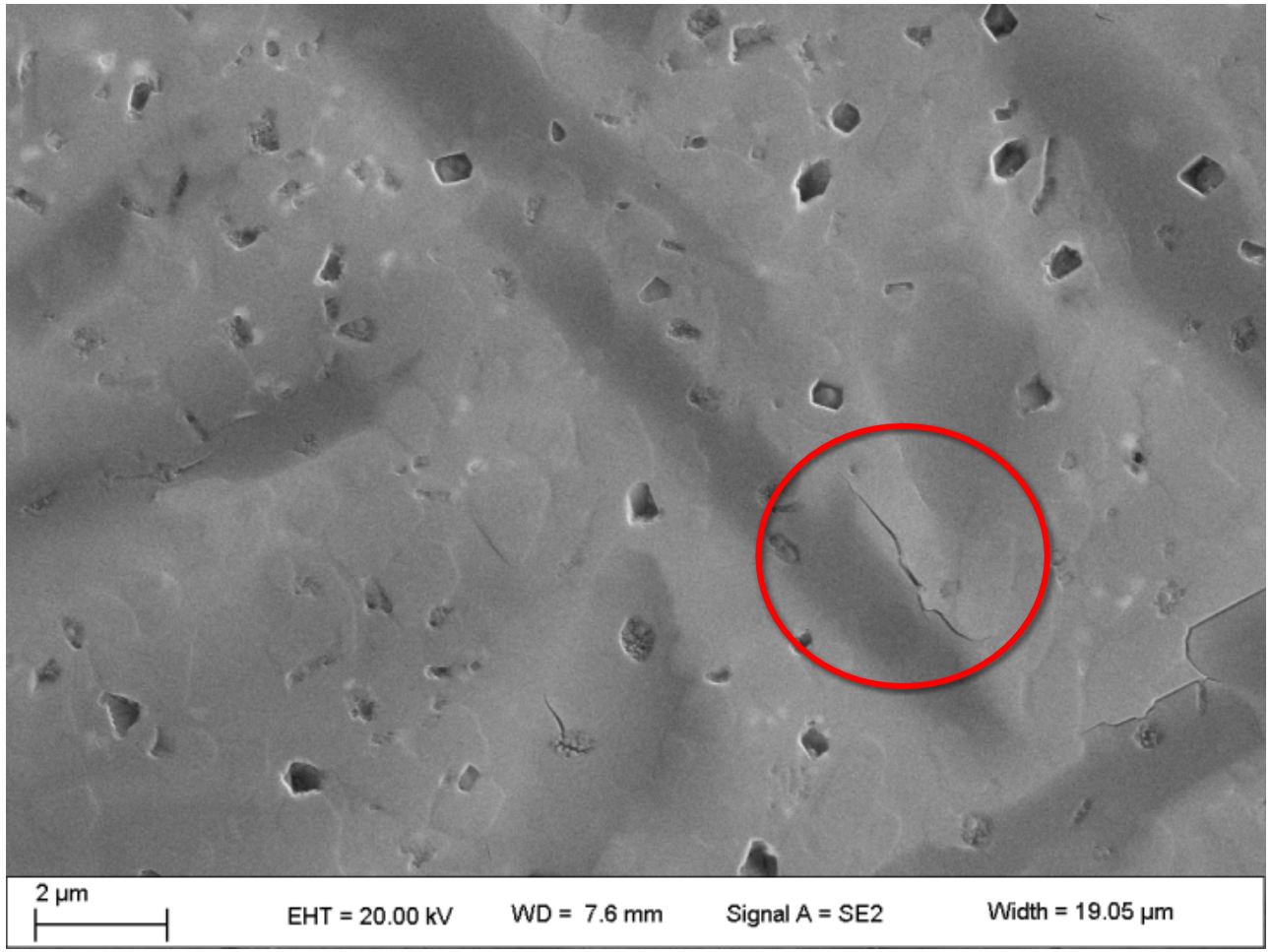


# SEM - Microstructure



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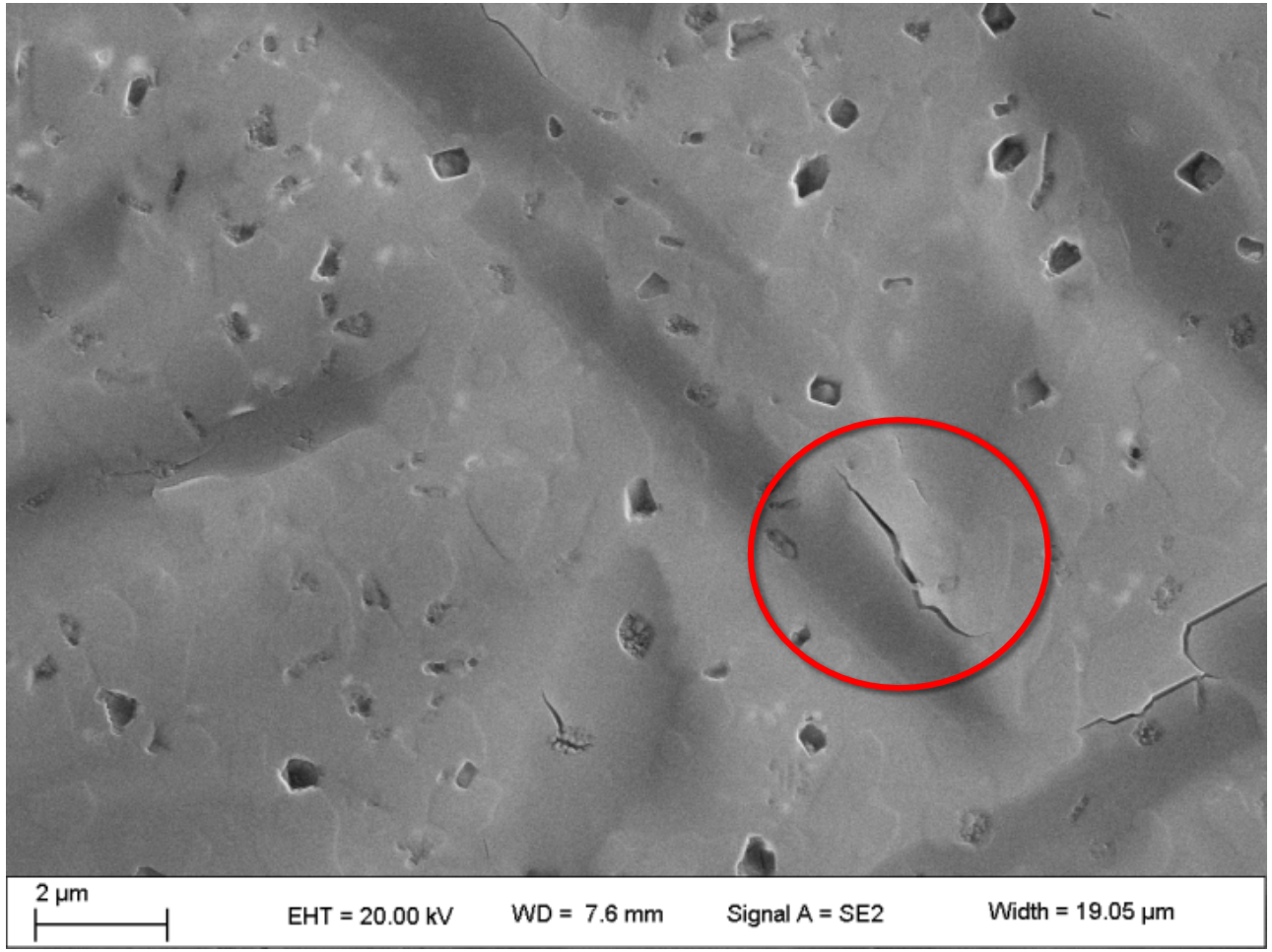
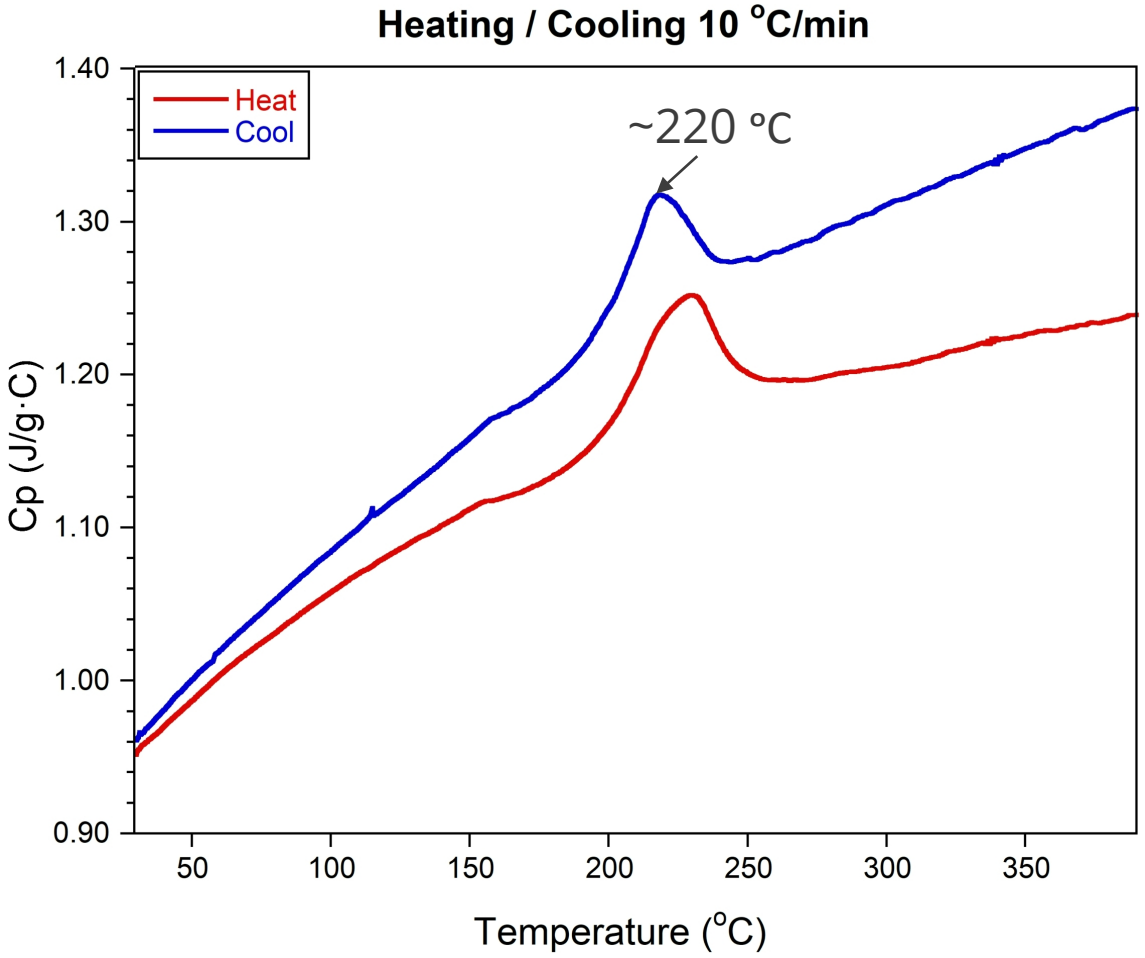
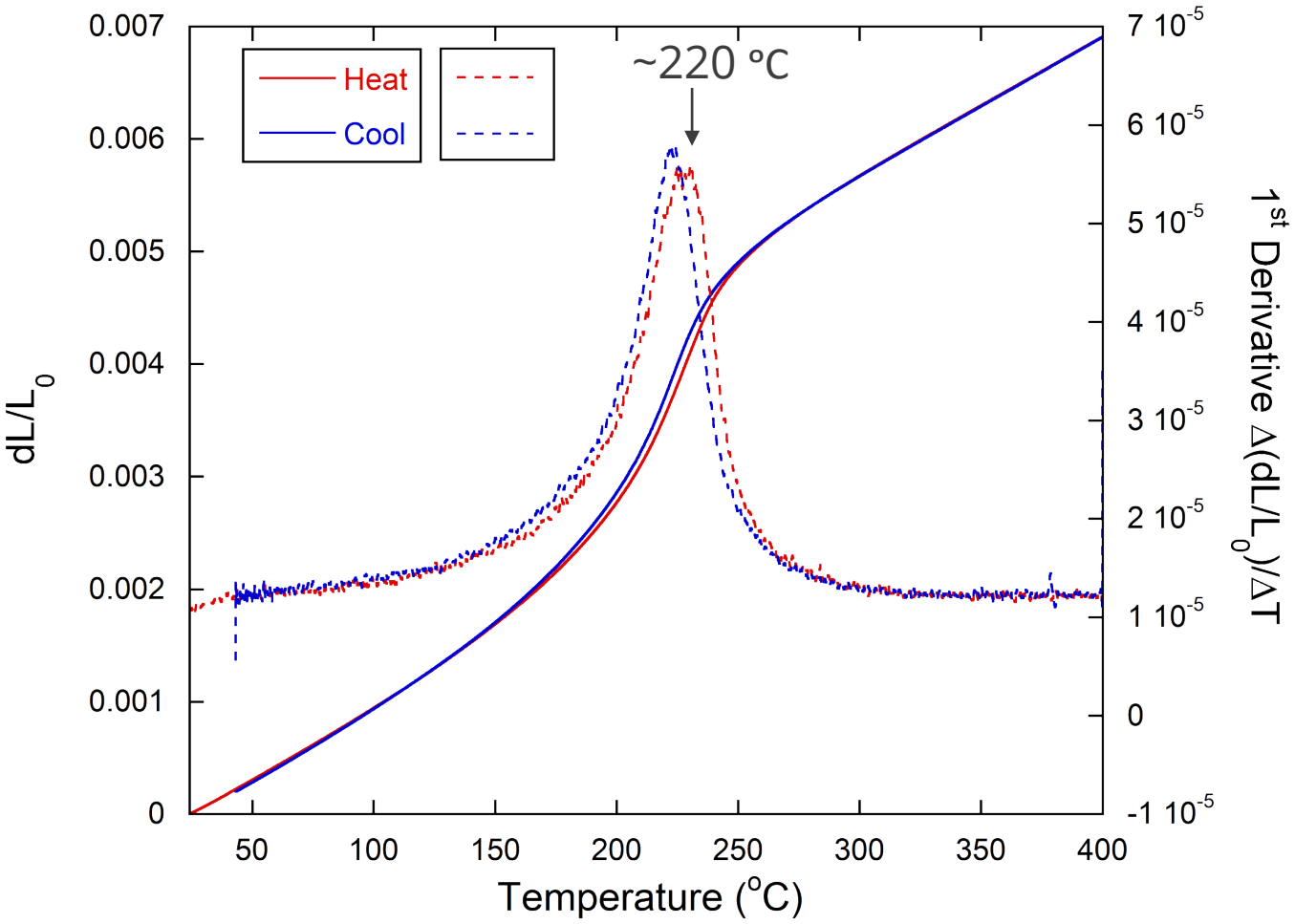



**Significant amount of residual stress in these materials**

# Material Characterization – DSC / Dilatometry

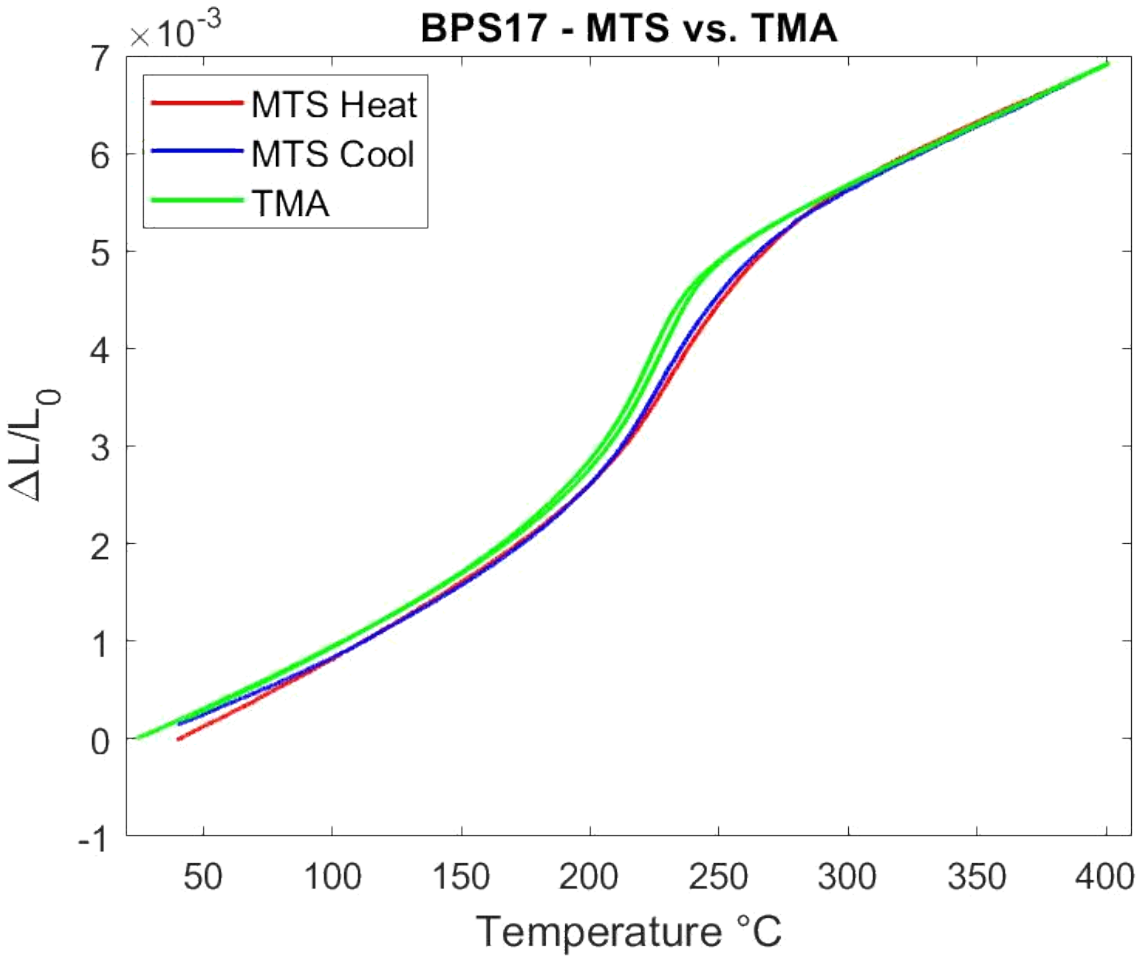


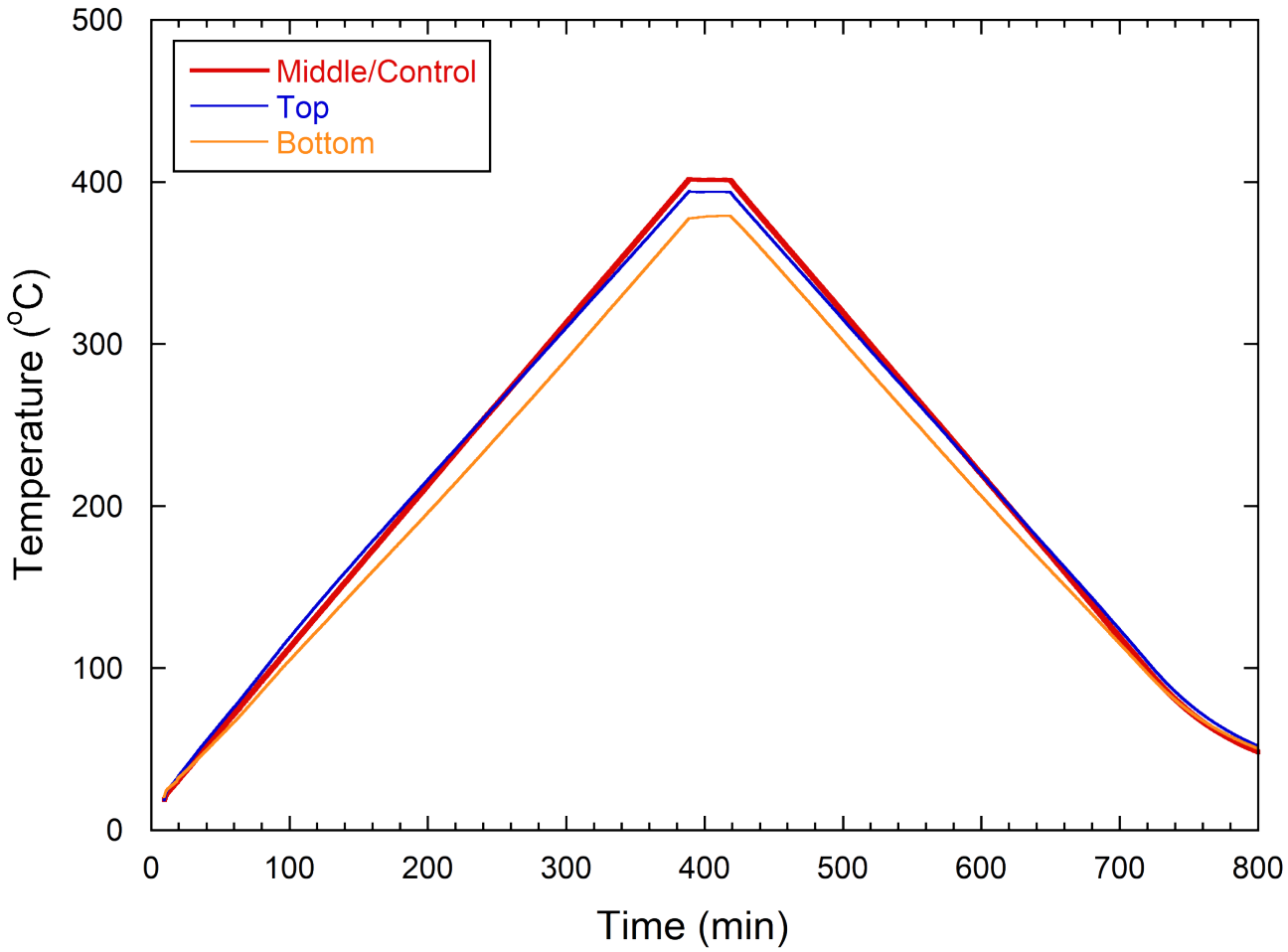




Theoretical Phase Transition Temperature - ~270 °C.

Swainson, I. P., and Martin T. Dove. "Molecular dynamics simulation of alpha and beta-cristobalite." *J. Phys.: Cond. Matter* **7** [9] (1995) p 1771.

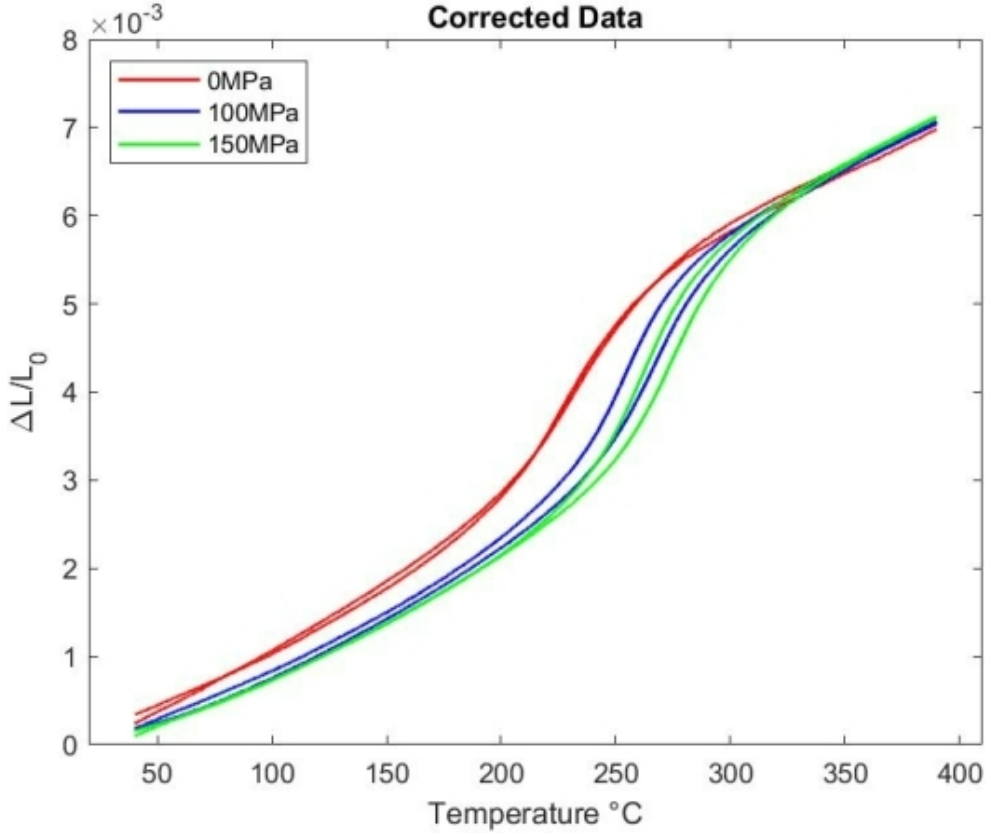
# Results – Comparison with TMA

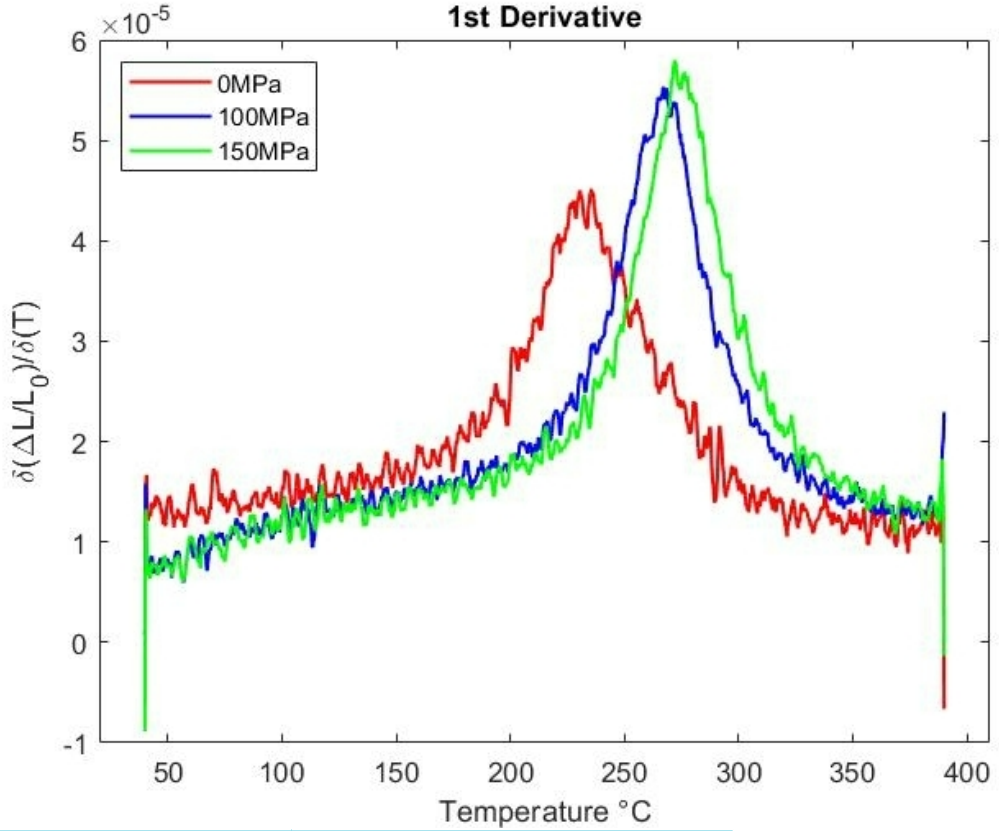



Phase transition region is “stretched” in the load frame data due to the thermal gradient along the specimen.

# Results – Applied Pressure Experiments

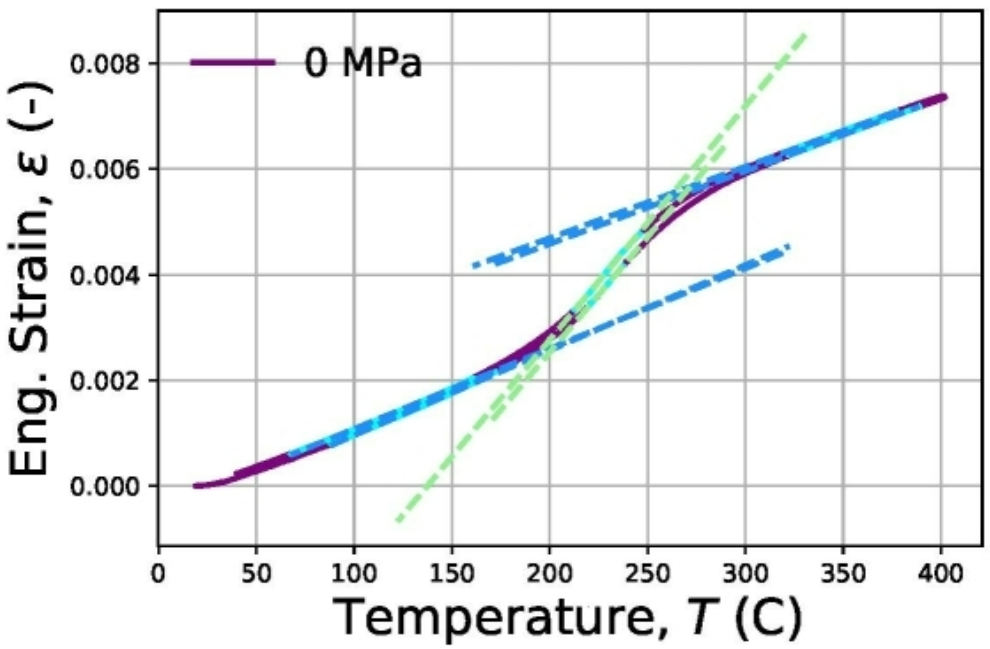


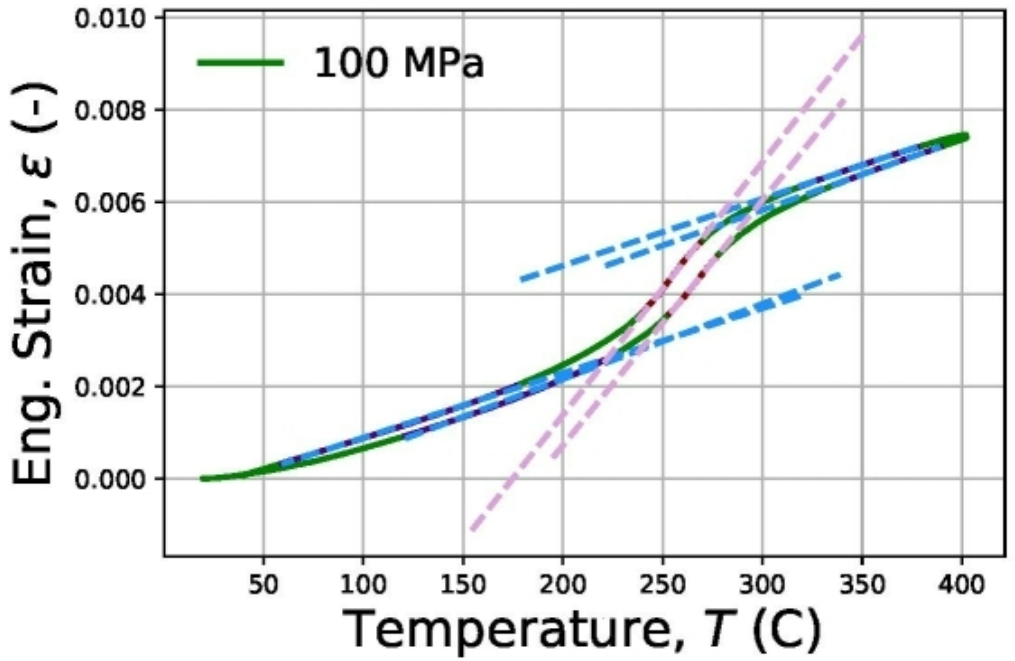



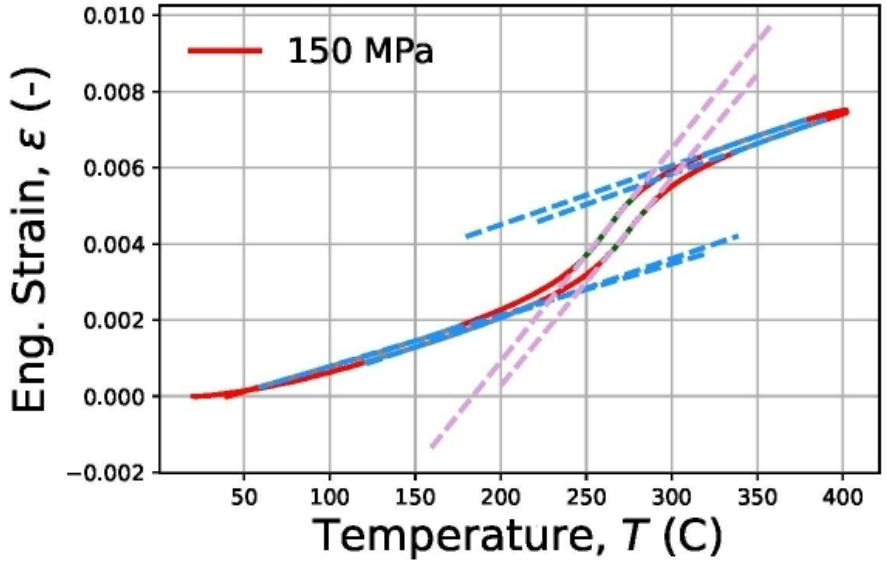

| Applied Stress (MPa) | Transformation Temperature (°C) | |
|---|---|---|
| | Heat | Cool |
| 0 | 231 | 227 |
| 100 | 268 | 253 |
| 150 | 273 | 262 |

Transition temperature shifts to higher temperatures with applied uniaxial compressive stress.

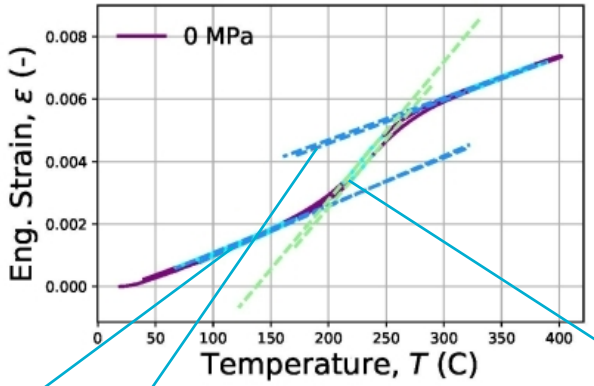
# Discussion – Data Interpretation

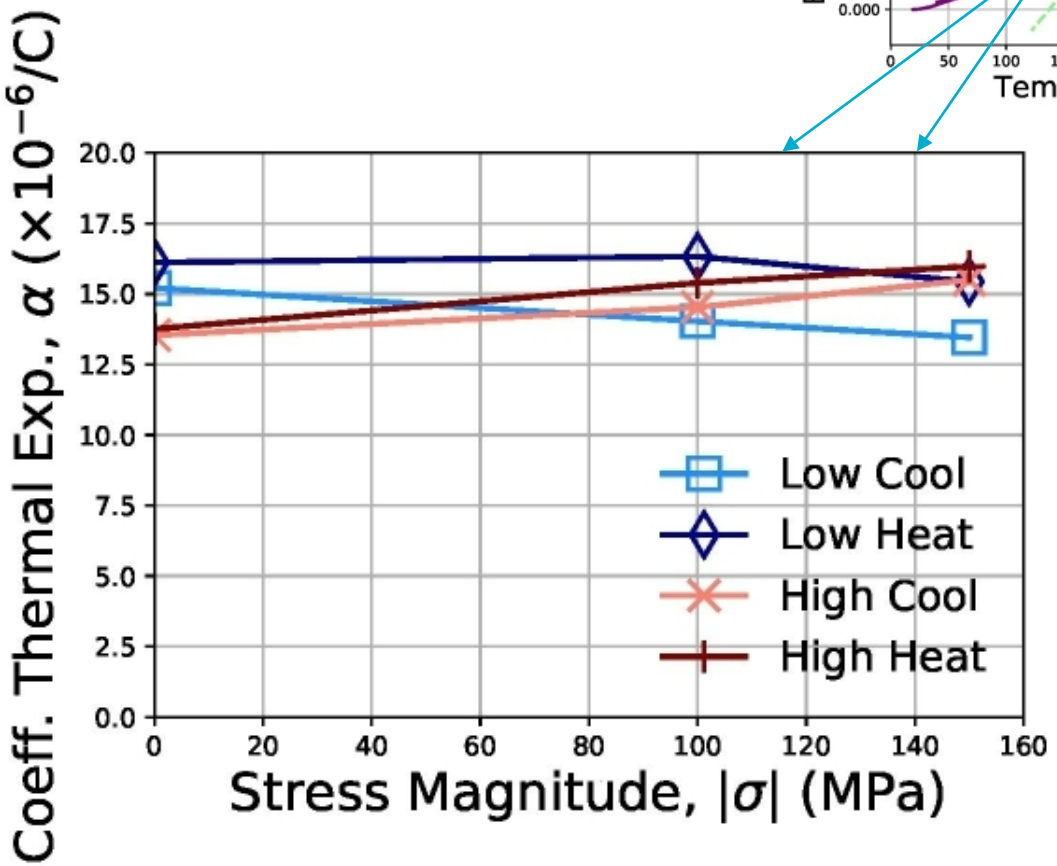


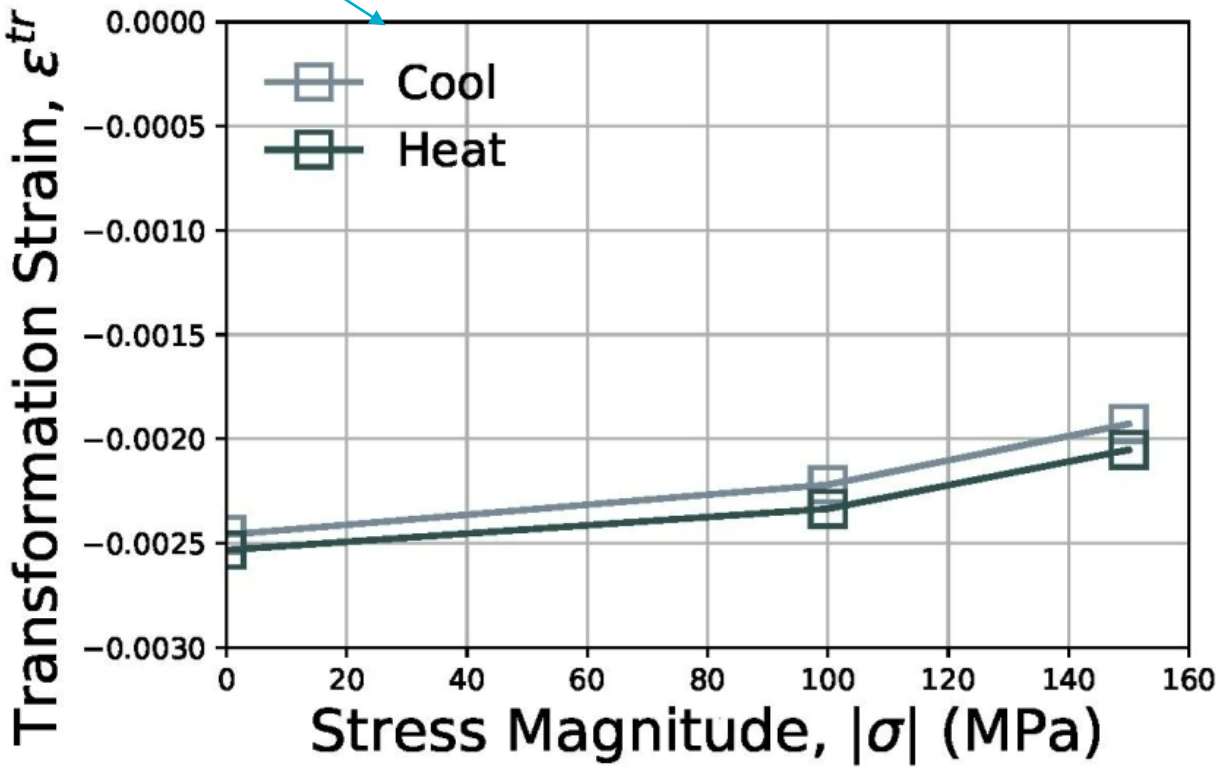



# Discussion – Data Interpretation

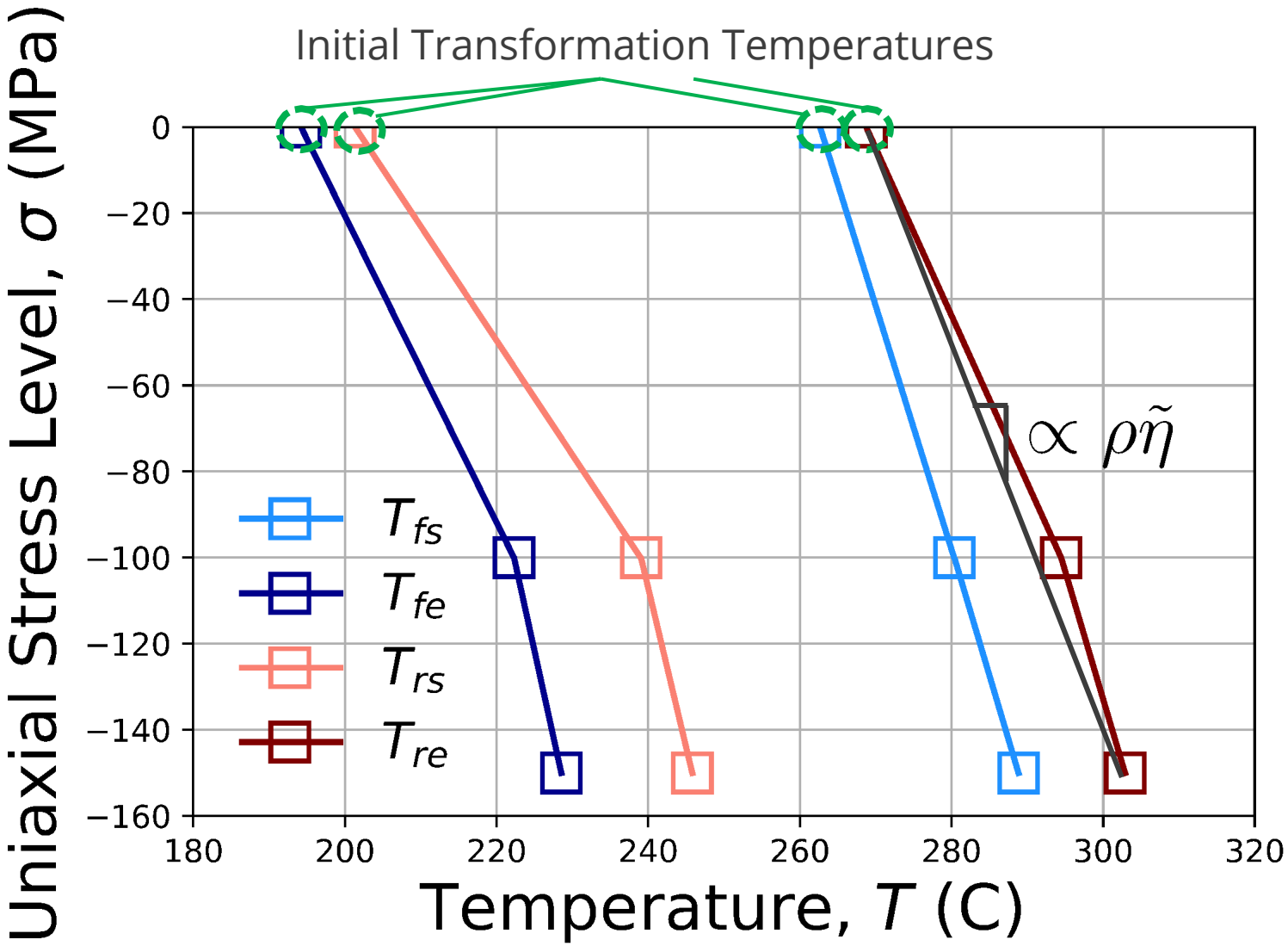


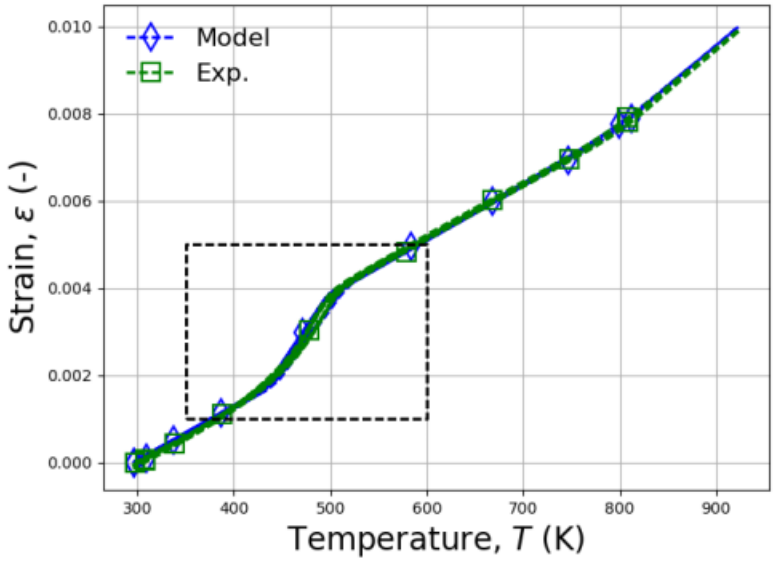
# Phase Diagram Characterization

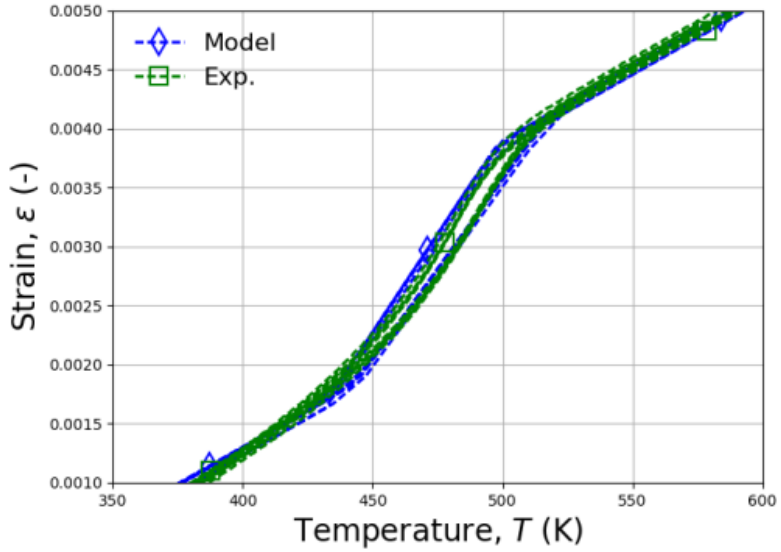
- Determination of phase-diagram is essential for model development and calibration
- Informs flow rule
- Essential parameters readily identifiable
  - Initial transformation temperatures
  - Stress-temperature slope



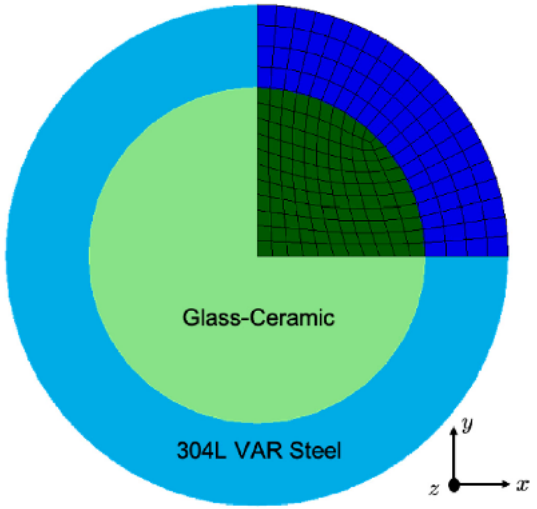
# Current Model Development

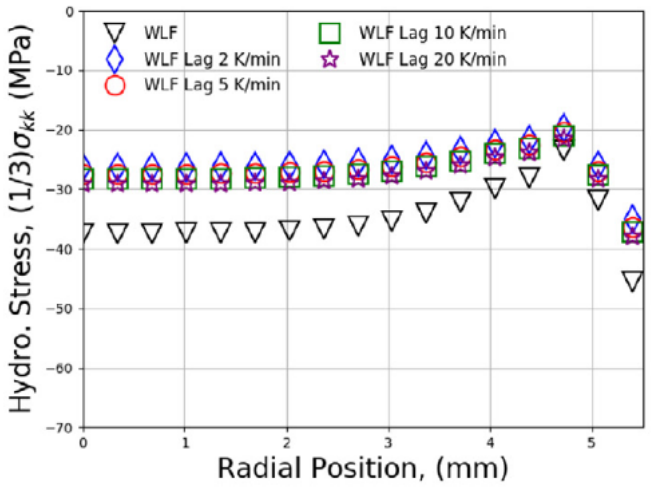
Modeling results compare with experimental results with no applied stress.

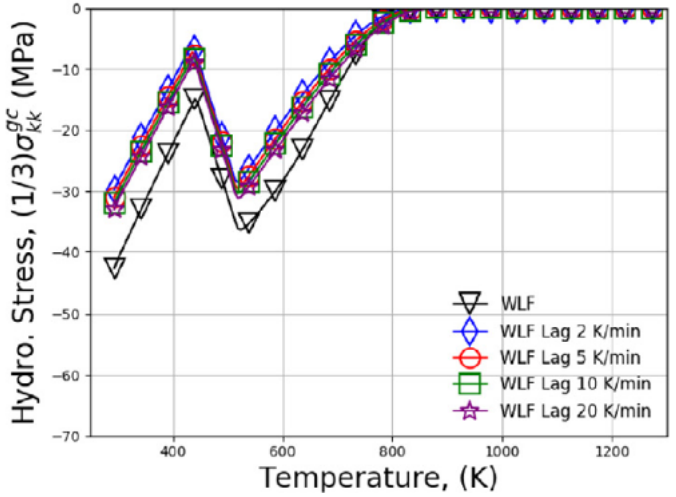



Unknown if material model will accurately predict changes in transformation temperature will applied stress (e.g. from a concentric steel shell)




B.T. Lester, K.N. Long; "A constitutive model for glass–ceramic materi
*Mechanics of Materials;* **158** (2021)

# Summary

- Measured the applied pressure dependent phase transition of cristobalite containing glass-ceramics with custom built thermal expansion setup.
- Future experiments will focus on having a controlled and uniform heated zone.
- Attempt to measure the deviatoric strain vs. temperature.

## Acknowledgements

Mark Rodriguez

Mia Blea

# Questions?

**Ruptured Panel**
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**By: Daniel Vuono**

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