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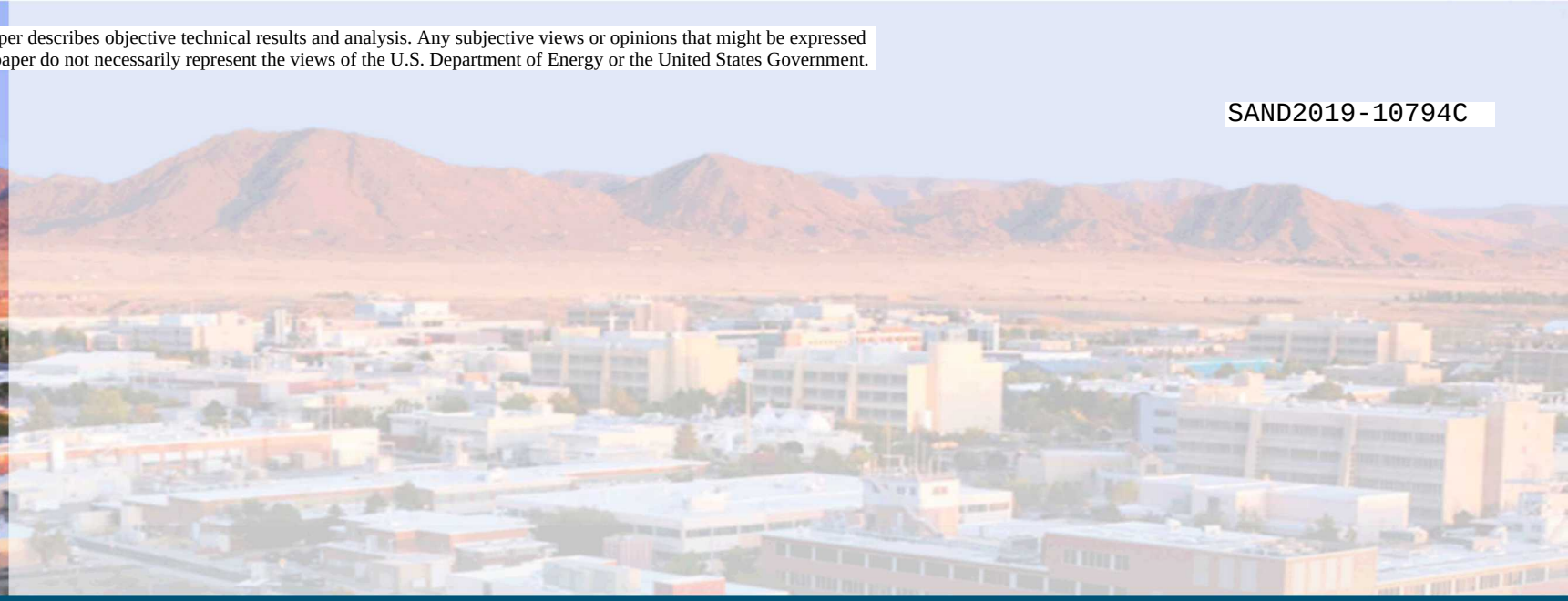

Abnormal Crystallization in Glass-Ceramic to Metal Seals

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Rio Grande Symposium for Advanced Materials

16 September 2019

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Sandia National Laboratory

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MetLab
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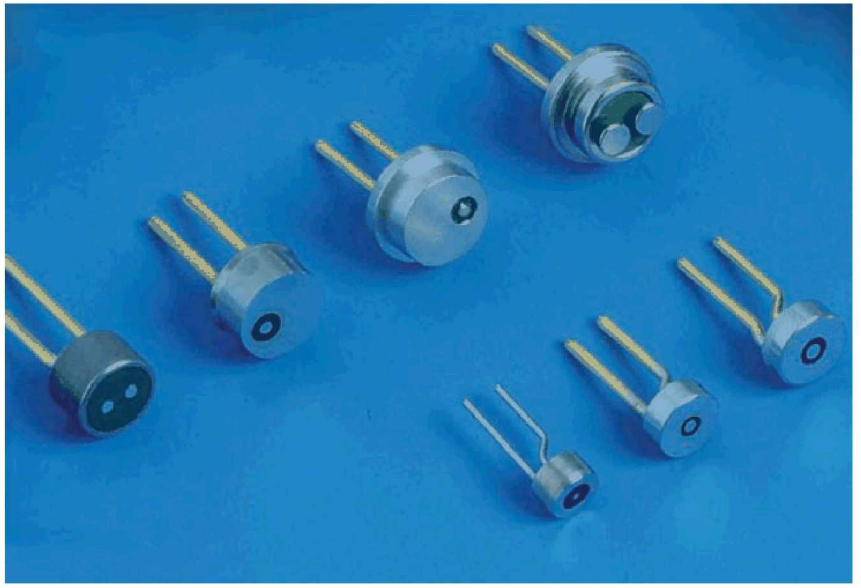
Electrical Contact Pin Assemblies

Glass is used to bond or join materials in compression seals

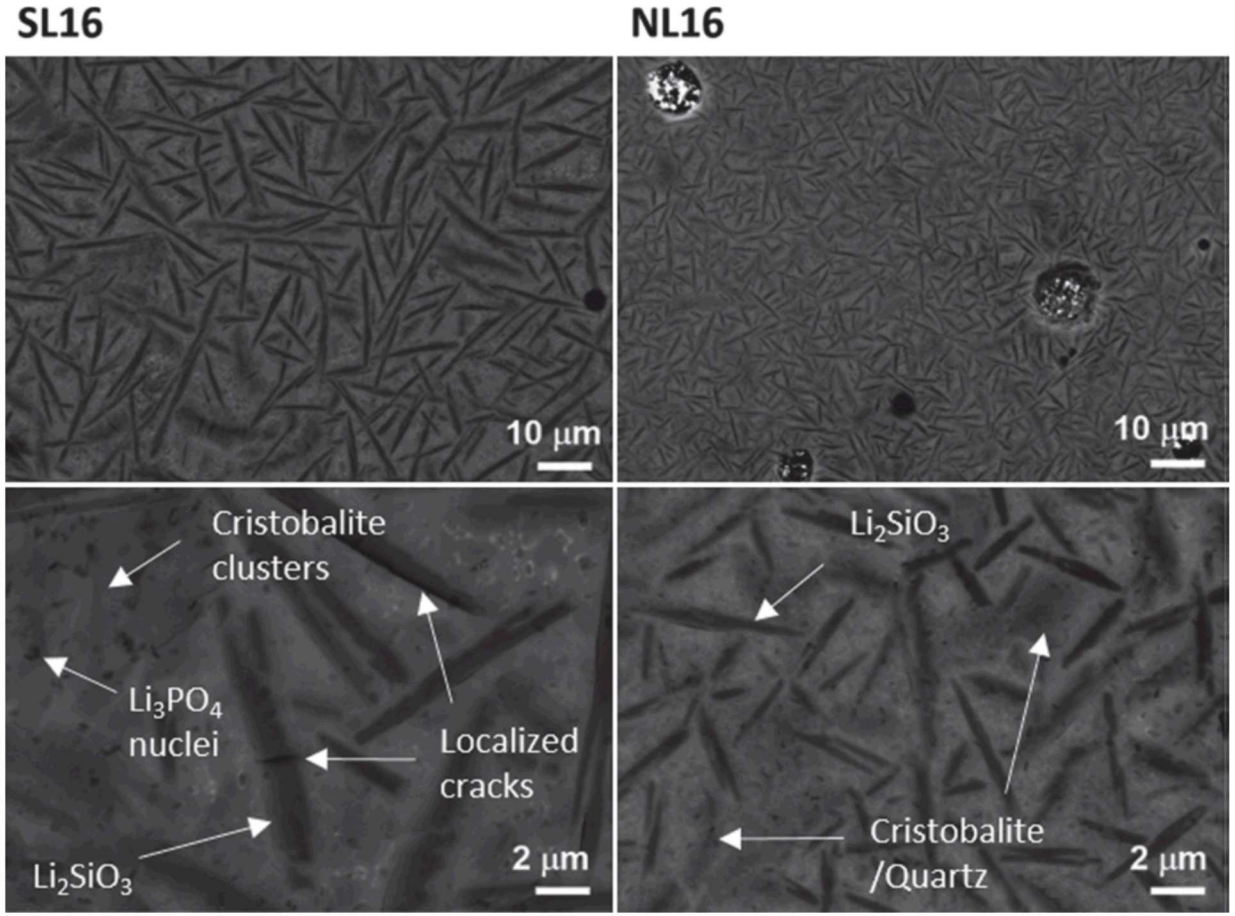
- In this case, an precious metal pin to a stainless steel shell

Glass-ceramic is an improvement over traditional glass seals because it offers tunability with thermal expansion coefficients and improved mechanical properties

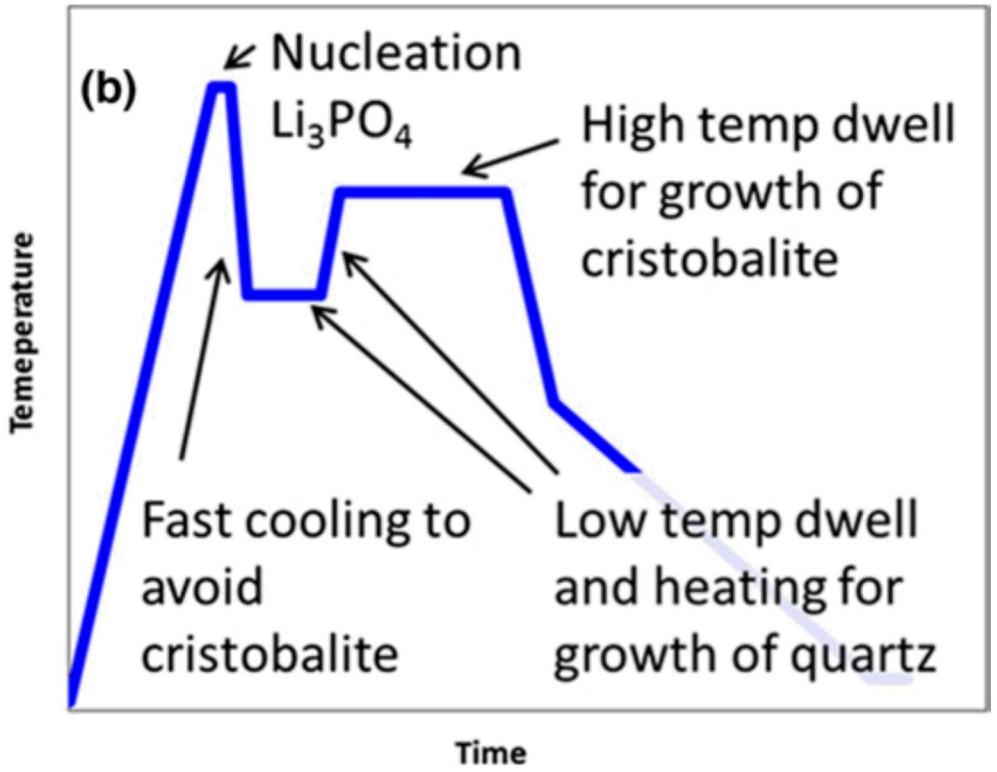


Thermal Profile and Glass-Ceramic Phases

"Near linear" CTE profile better matches the pin and shell (small steps)



Rodriguez, Griego, Dai. J. Am. Ceram. Soc., 99 {11}, 2016.



Dai, Rodriguez, Griego. J. Am. Ceram. Soc., 99 {11}, 2016.

Questions

How common is abnormal crystallization?

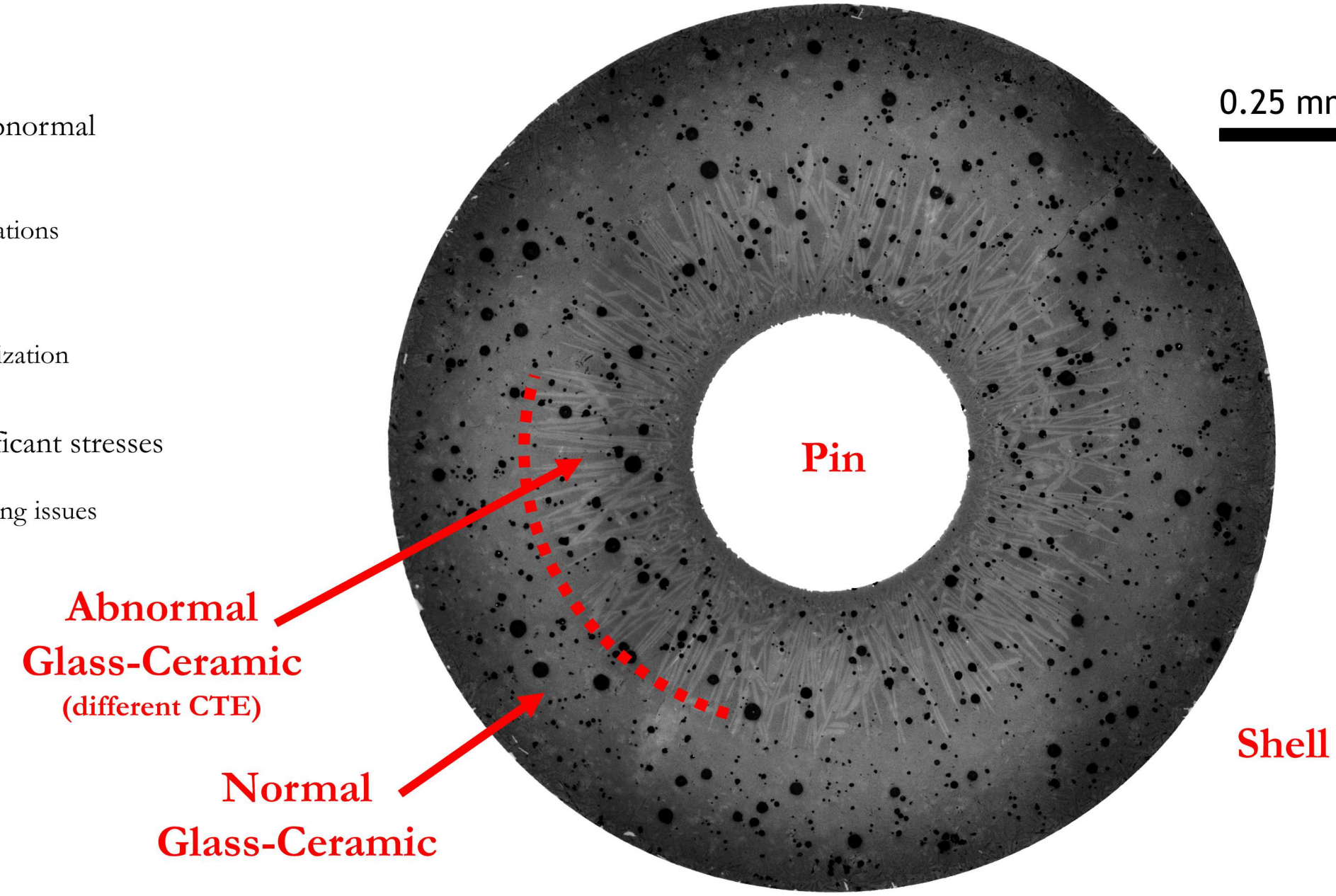
- Survey
- Area/length calculations

What causes it?

- Interface characterization

Does it cause significant stresses within the seal?

- Hermeticity/cracking issues
- FEA

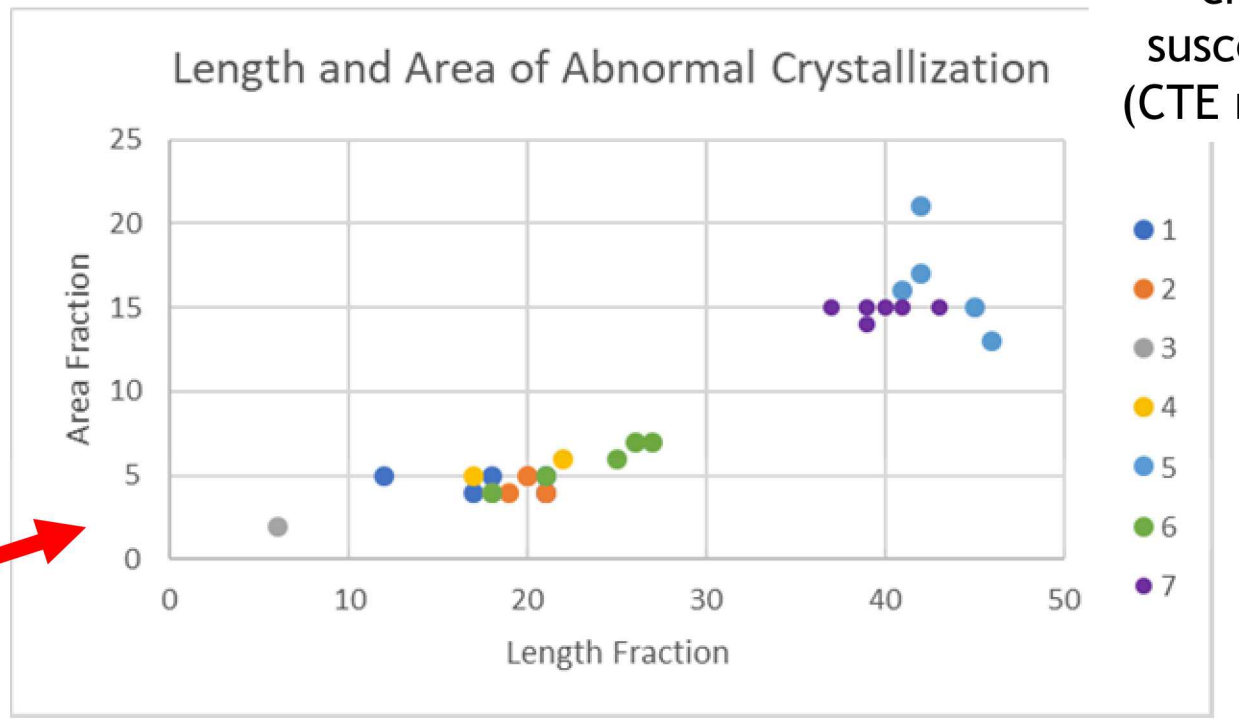


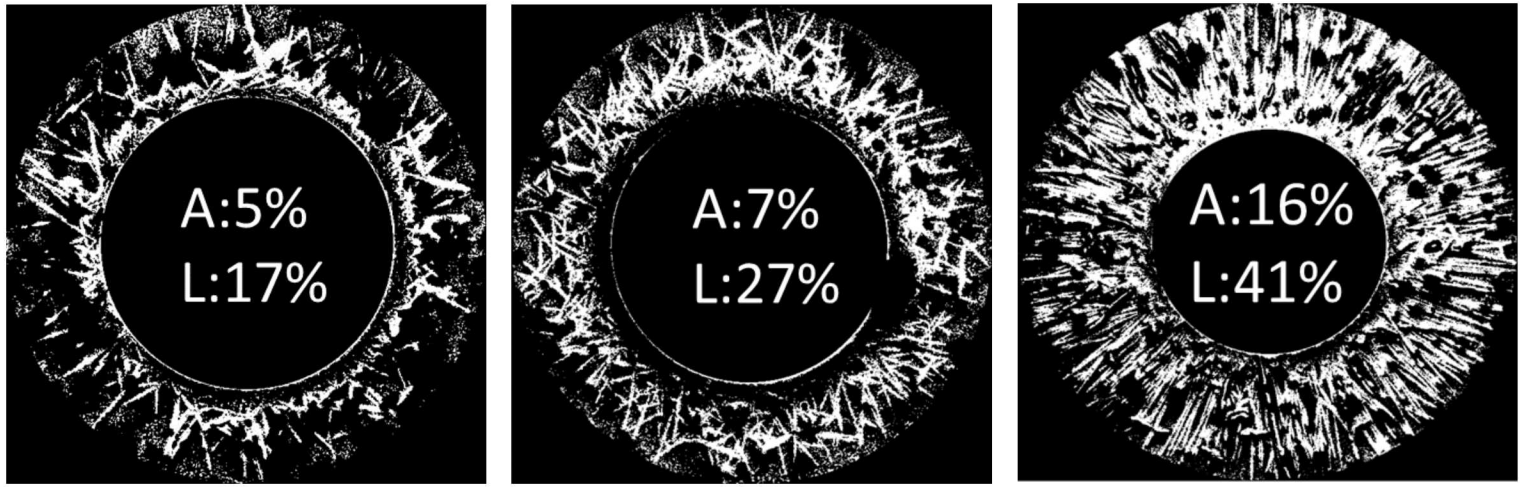
Abnormal Crystallization Survey

32% of headers surveyed show some degree of abnormal crystallization

Of those with abnormal crystallization, there is significant variation in area and length fraction

Increased cracking susceptibility (CTE mismatch)

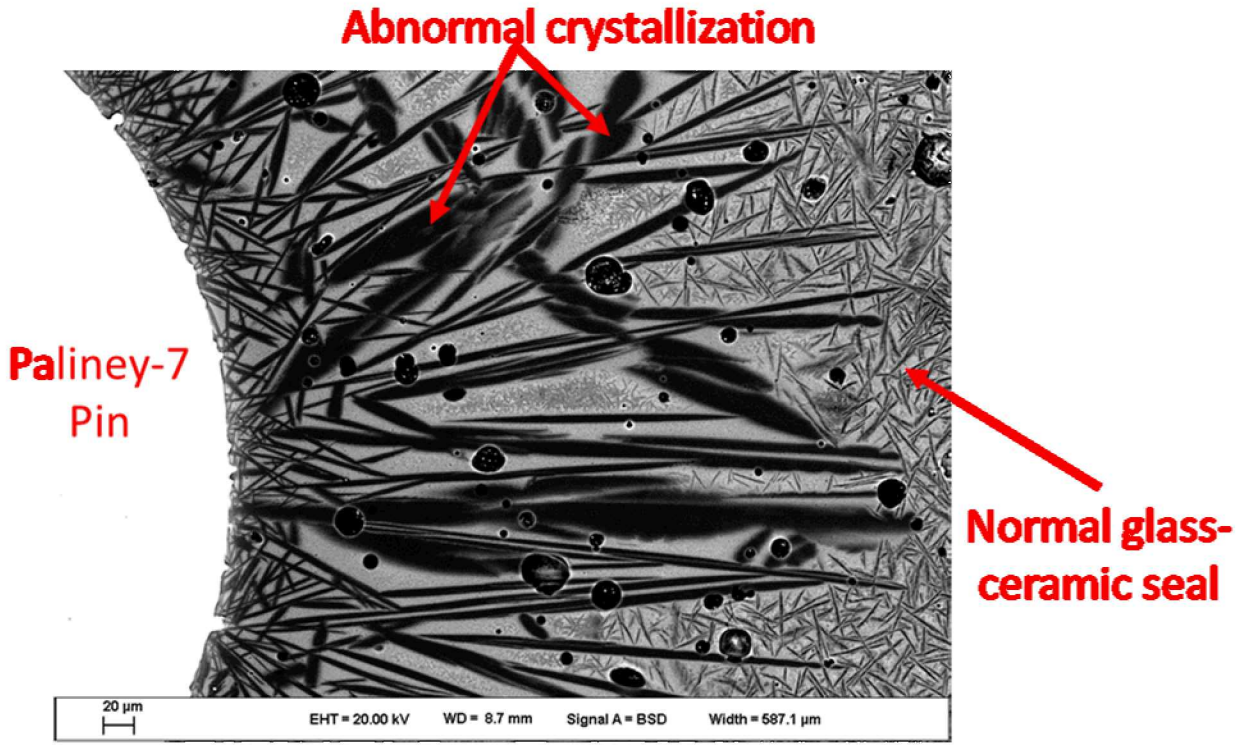




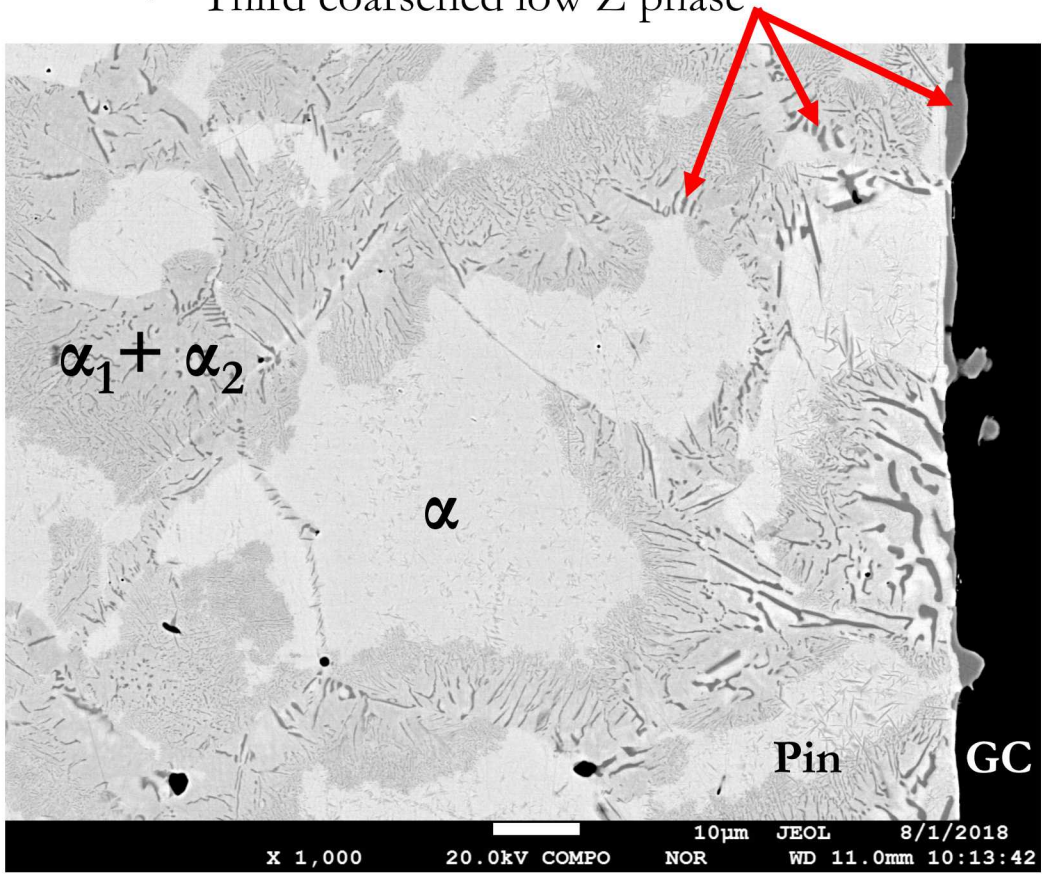
ImageJ Threshold Analysis

Pin to Glass-Ceramic Interface Characterization

What causes abnormal crystallization?

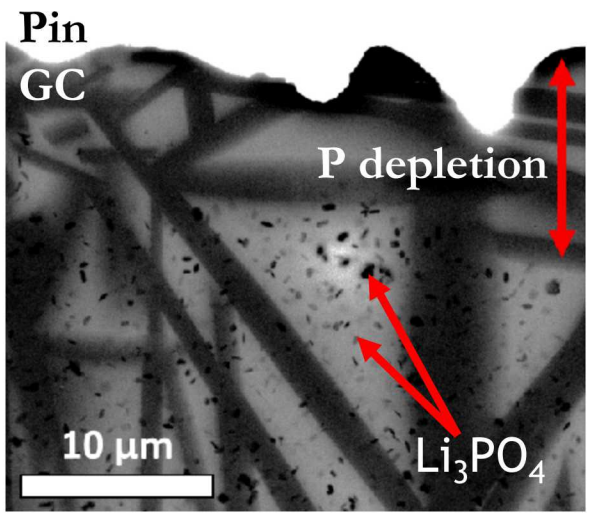


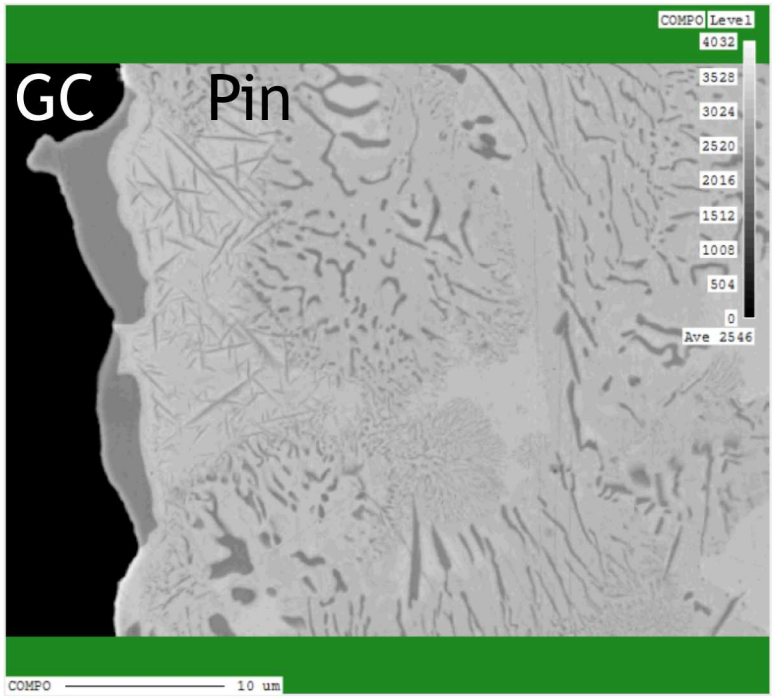
- Two FCC $\alpha_1 + \alpha_2$ – Ag rich, Cu rich (discontinuous precipitation from parent α)
- Third coarsened low Z phase

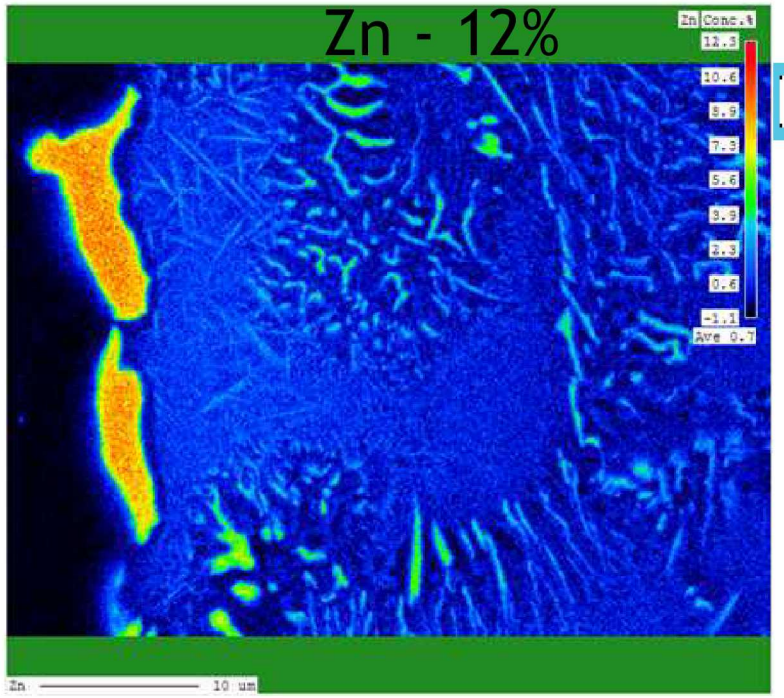


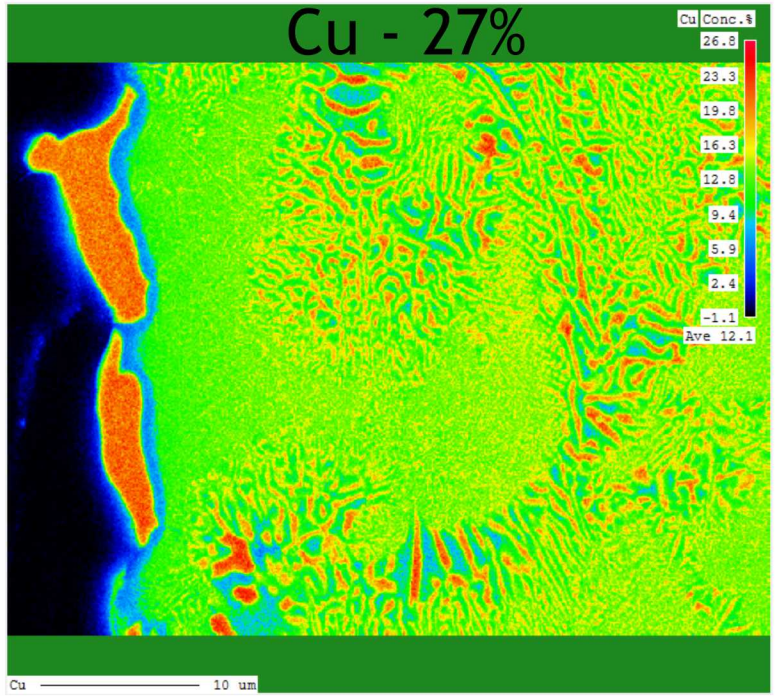
Interfacial Third Phase: WDS

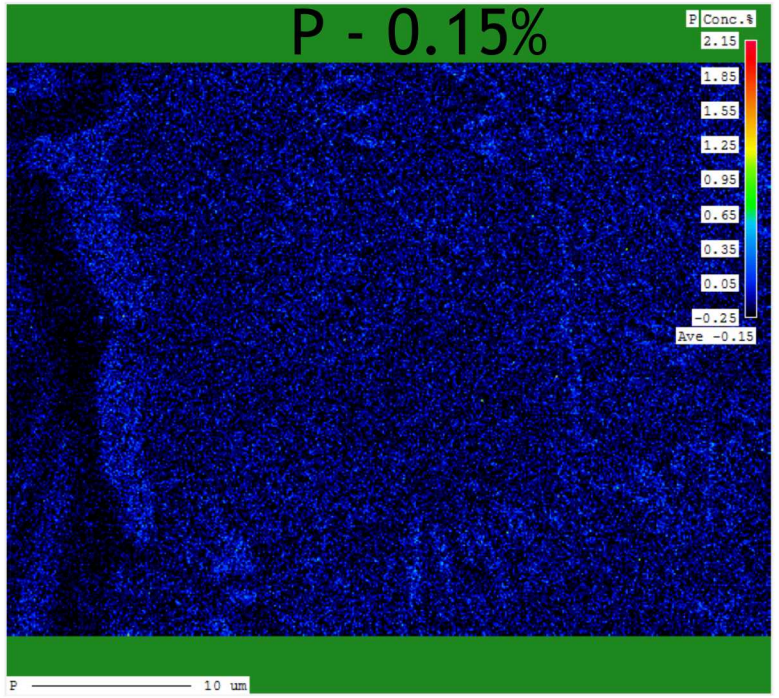
- WDS shows the interfacial phase is rich in Zn, Cu, and P
- P is needed to form Li_3PO_4, the nucleating agent in the glass-ceramic



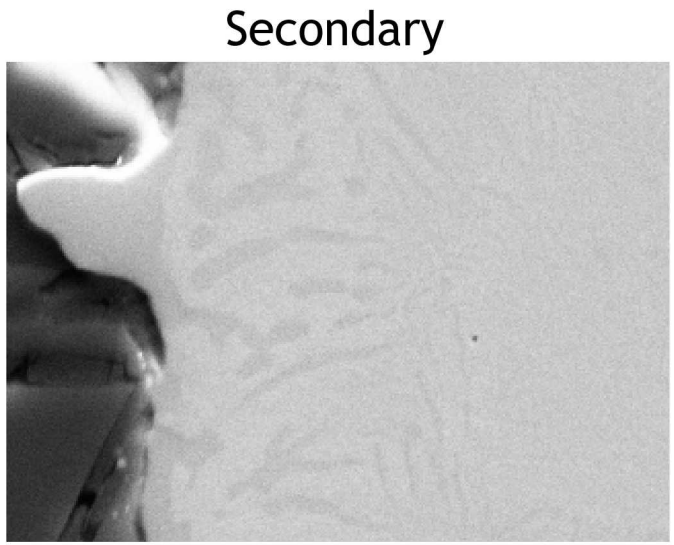


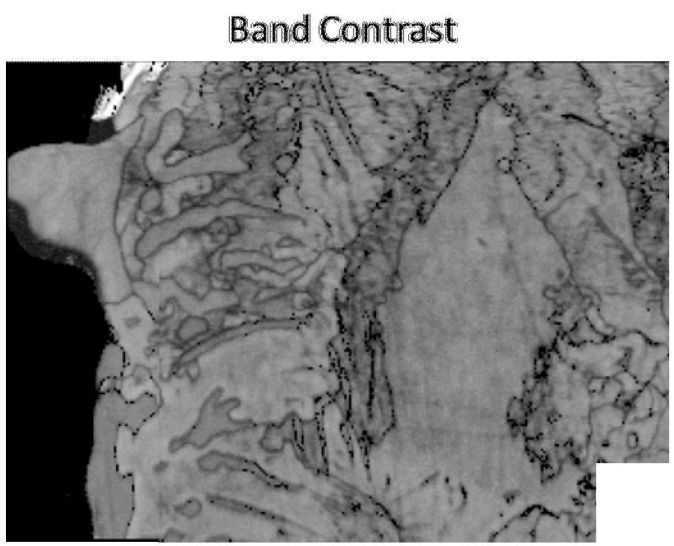


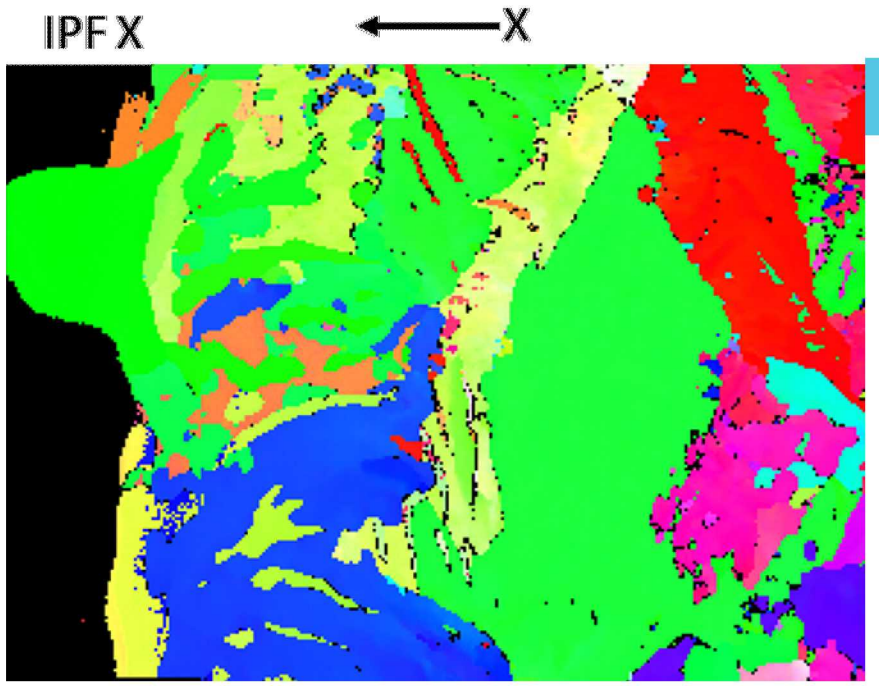


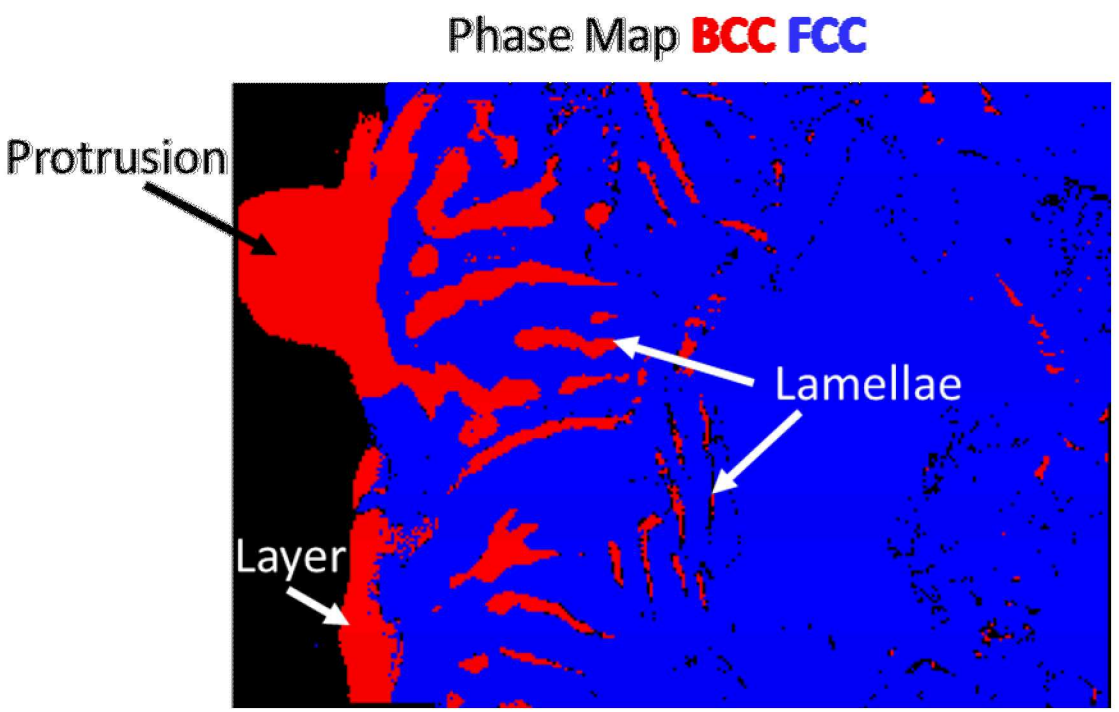


Interfacial Third Phase: EBSD





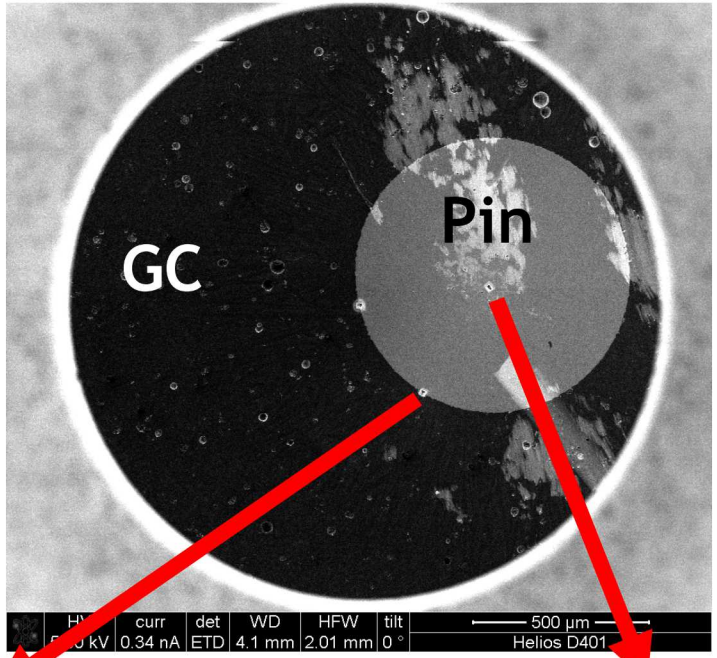




- EBSD shows the interfacial phase is BCC (red), also confirmed by XRD → New phase!
- There is not a preferred growth direction into the GC, but BCC texture appears to relate to parent FCC

- BCC phase was also found near the inside of the pin and on pins that hadn't been sealed, but is more abundant near the glass-ceramic interface

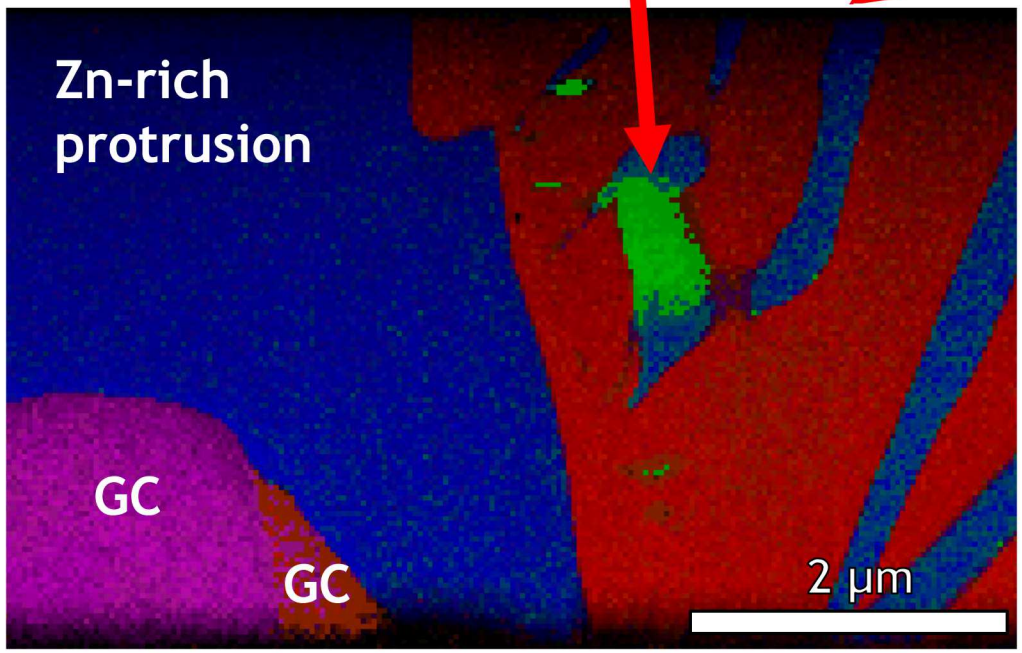
Interfacial Phase: TEM



- Zn-rich BCC is morphologically similar to the Cu-rich FCC
- Zn-rich BCC appears to form at the expense of Cu-rich FCC

FCC-BCC transformation

Interface

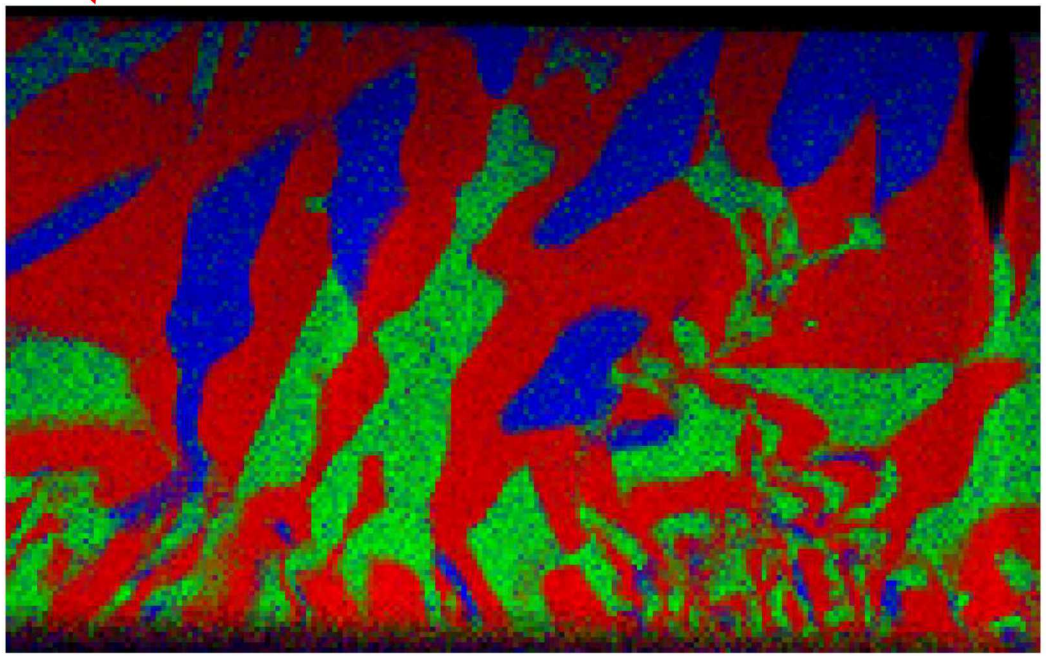


Zn-rich (BCC)

Cu-rich (FCC)

Ag-rich (FCC)

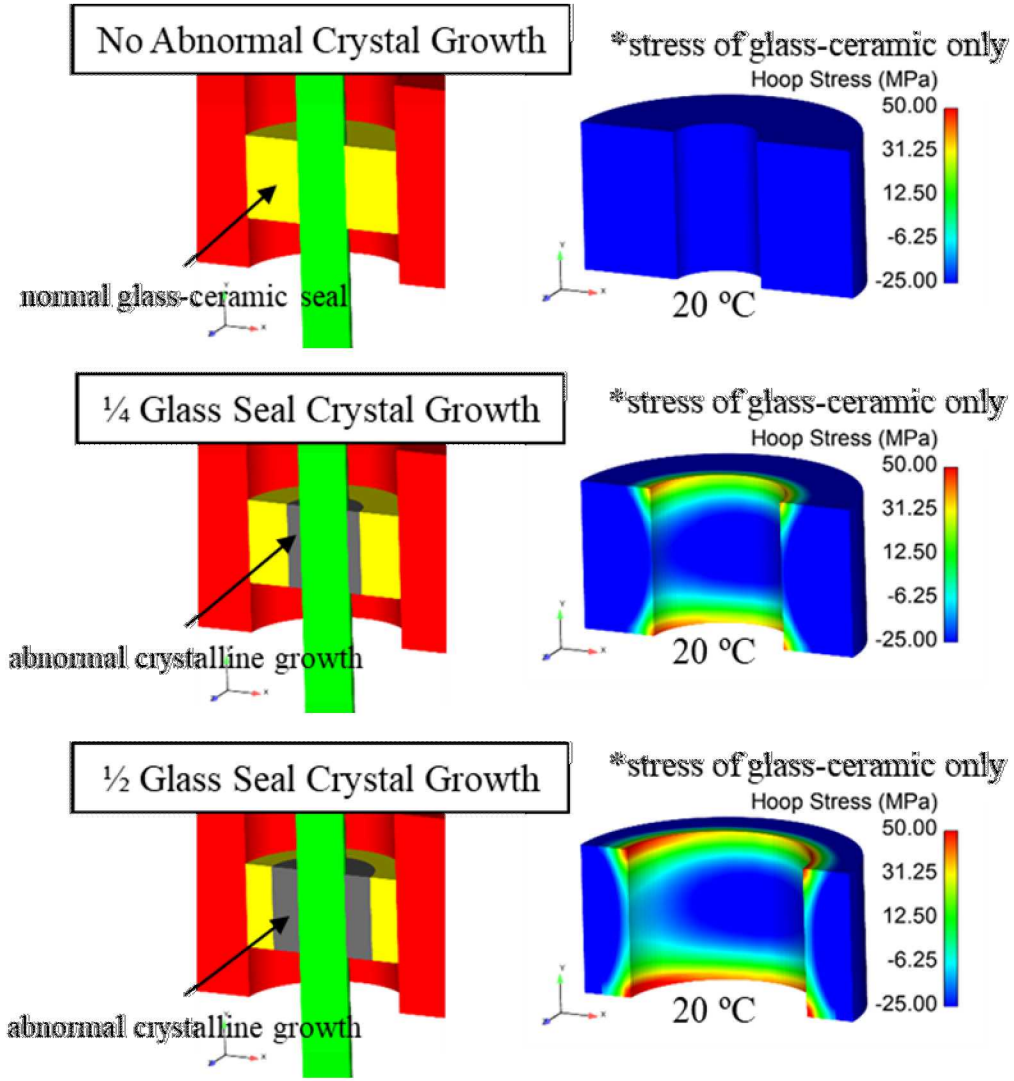
Center



Abnormal Crystallization FEA

Brenton Elisberg

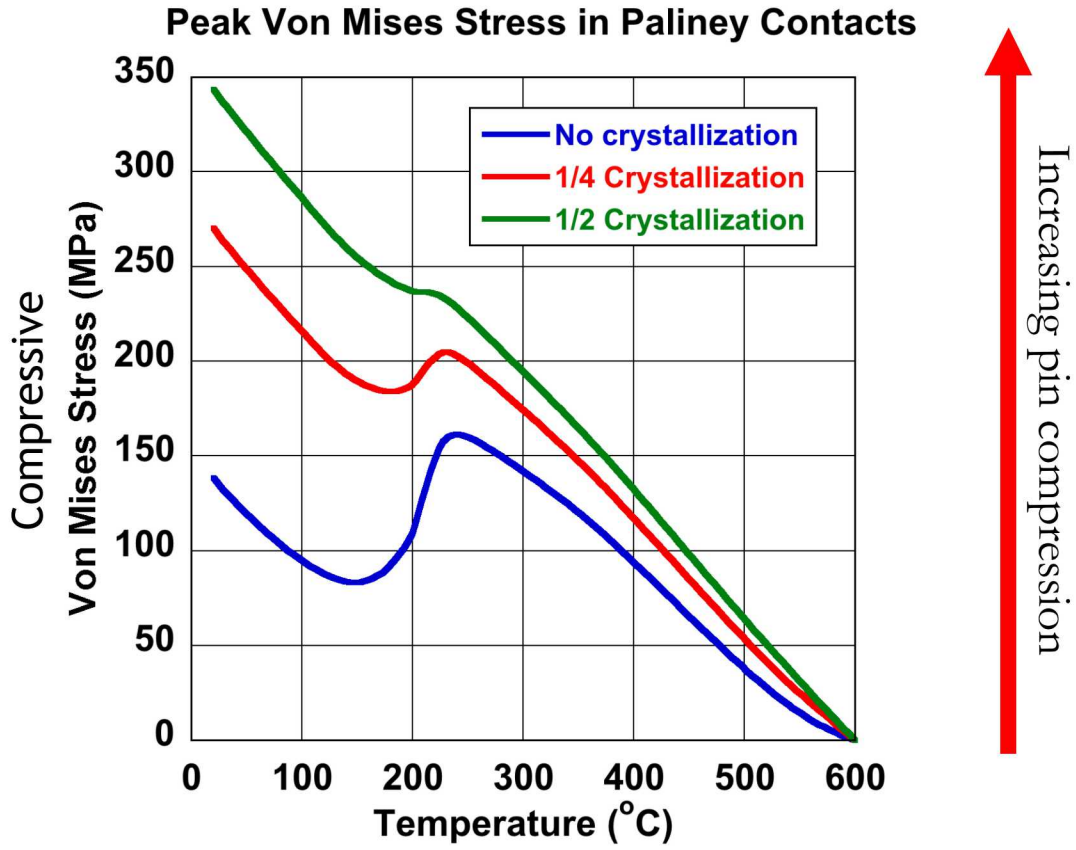
Increasing hoop tensile stress



Abnormal crystallization results in tensile hoop stresses within the glass, though we're unsure when the glass-ceramic would begin to crack

Also results in increased compression on the pin, though we're unsure when it would begin to deform

Our survey showed some headers with ~¼ crystallization, where we would be concerned about cracks forming



Increasing pin compression

Conclusions and Future Work

32% of surveyed headers show some extent of large crystals, though the severity varies

Possible causes related to interdiffusion between the pin and glass-ceramic: segregation of P from nucleating agent, or segregation of phase stabilizing elements

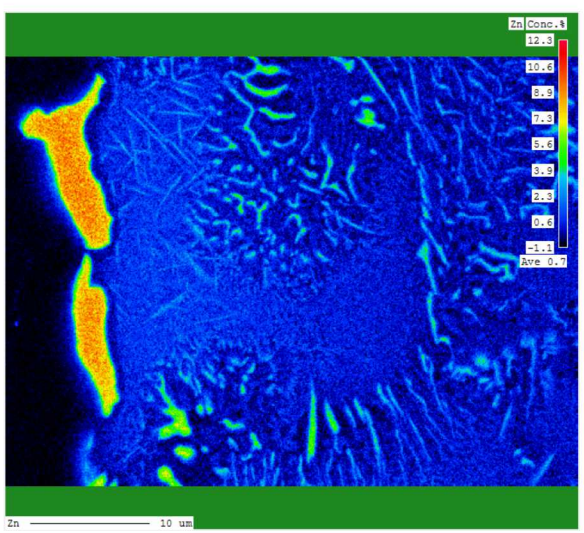
- Interdiffusion results in BCC phase rich in Zn, Cu, and P → New phase!

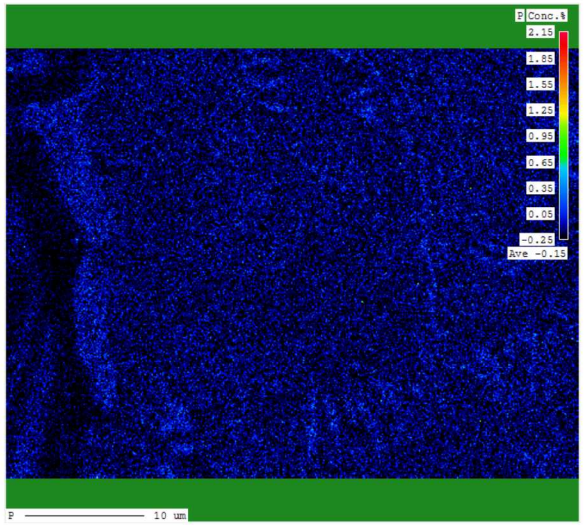
It appears that not all abnormal crystallization will generate enough stress to see an impact, perhaps only the highest area fractions

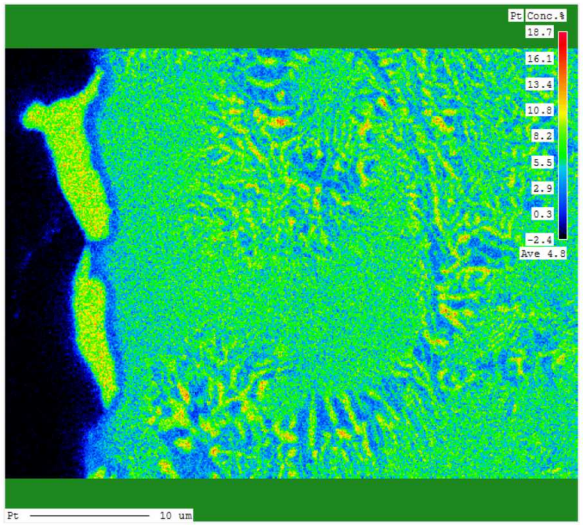
WDS

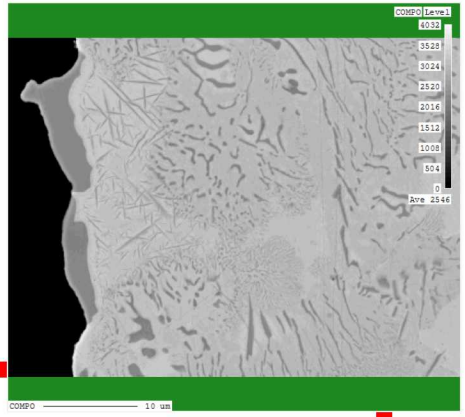
Zn, Cu and P enrichment in lamella and surface features

Depletion zones of Zn and Cu nearby – enriched specifically in Ag??

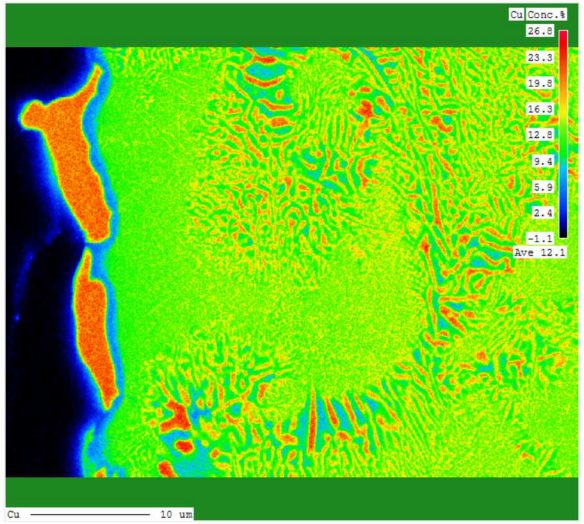


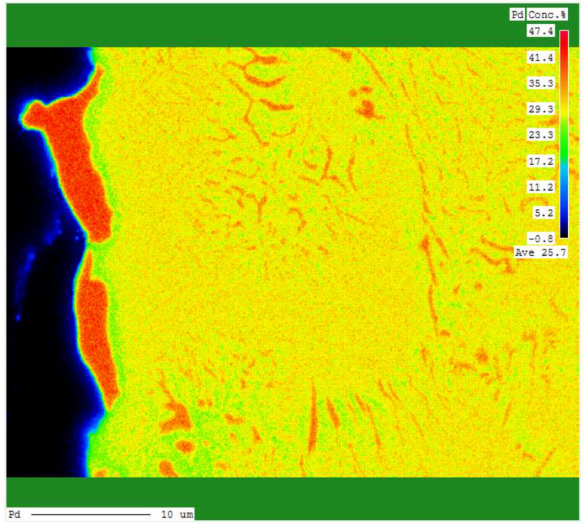


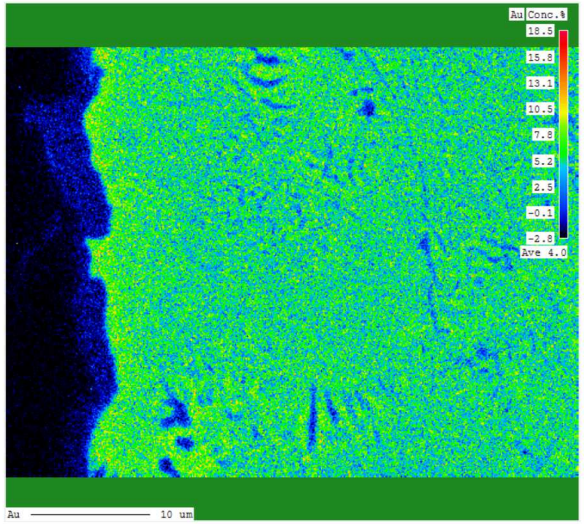


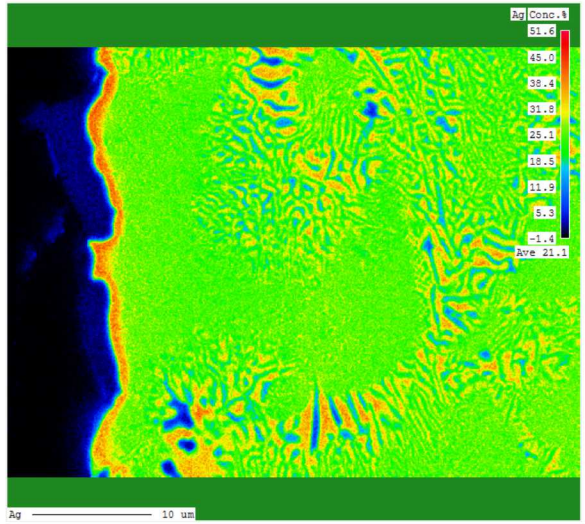


Ag









Some areas rich in Cu but not Zn – this is the second FCC phase (see TEM)