Engineered Glass Seals for SOFCs

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Oak Ridge National Laboratory

Acknowledgments

- Valerie Garcia-Negron, Dana McClurg, Beth Armstrong, Rosa M. Trejo, Hannah Stokes and John Henry (ORNL).
- Matt Chou, Jeff Stevenson (PNNL).
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- Rin Burke and Briggs White for guidance and support.

Outline

- Background
- Engineered Glass Seals
  - Characterization
  - Routes to low-cost manufacturing
- Summary

Background

Requirements for SOFC seals

- Simultaneous fulfillment of thermal, physical, chemical, mechanical and electrical property requirements.
- Phase stability and chemical compatibility without substantial property degradation for 40,000 hours in oxidizing and wet reducing environments.

- Address potential lack of flatness and/or parallelism of cells with large active area

Objective

- To develop engineered glass seals for SOFCs.
- Identify low-cost manufacturing processes

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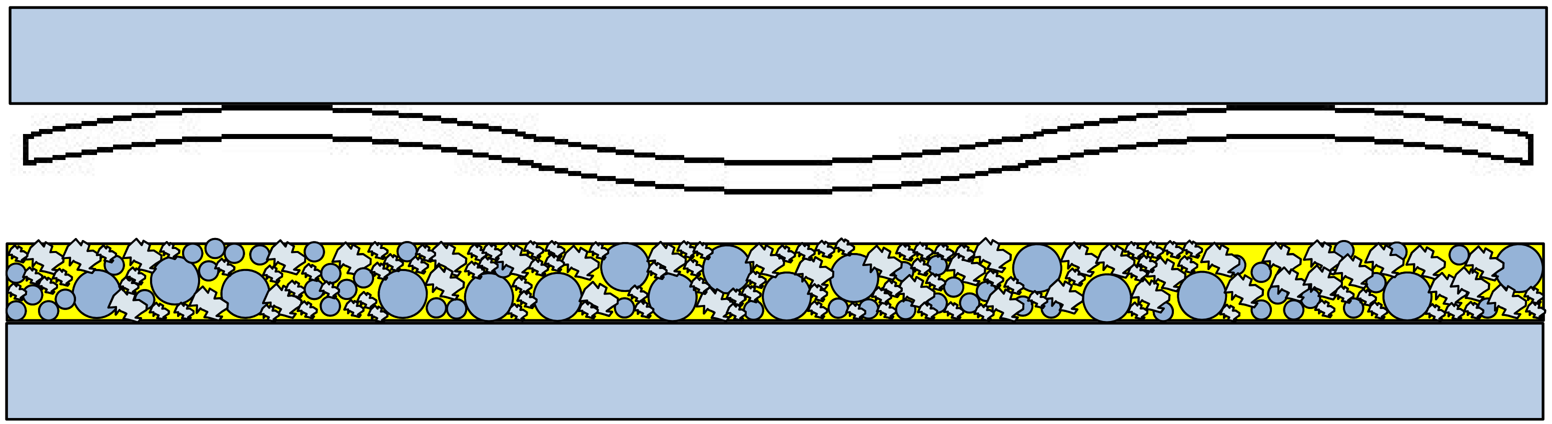

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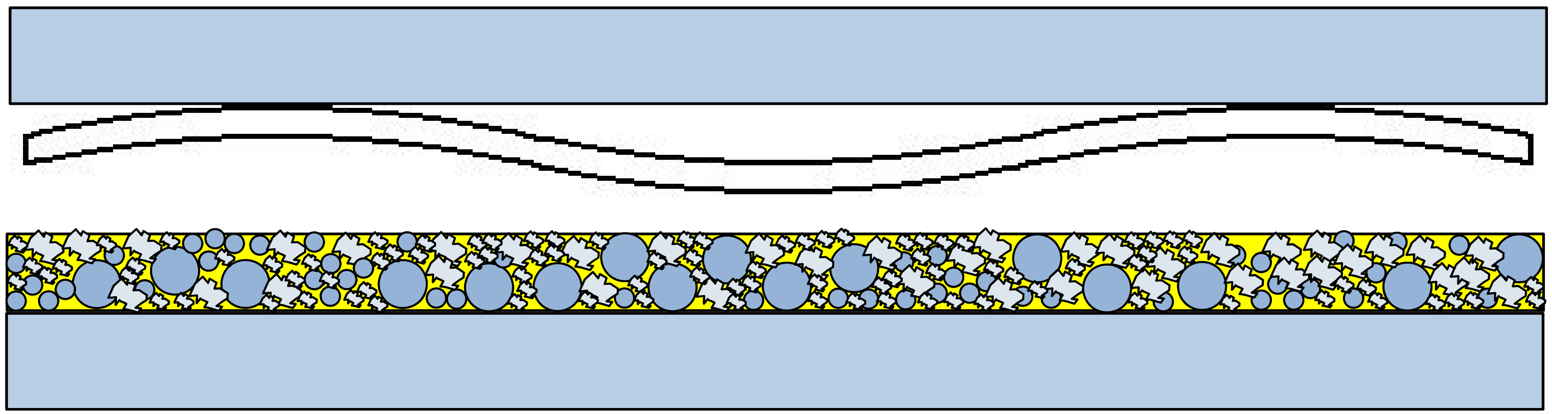

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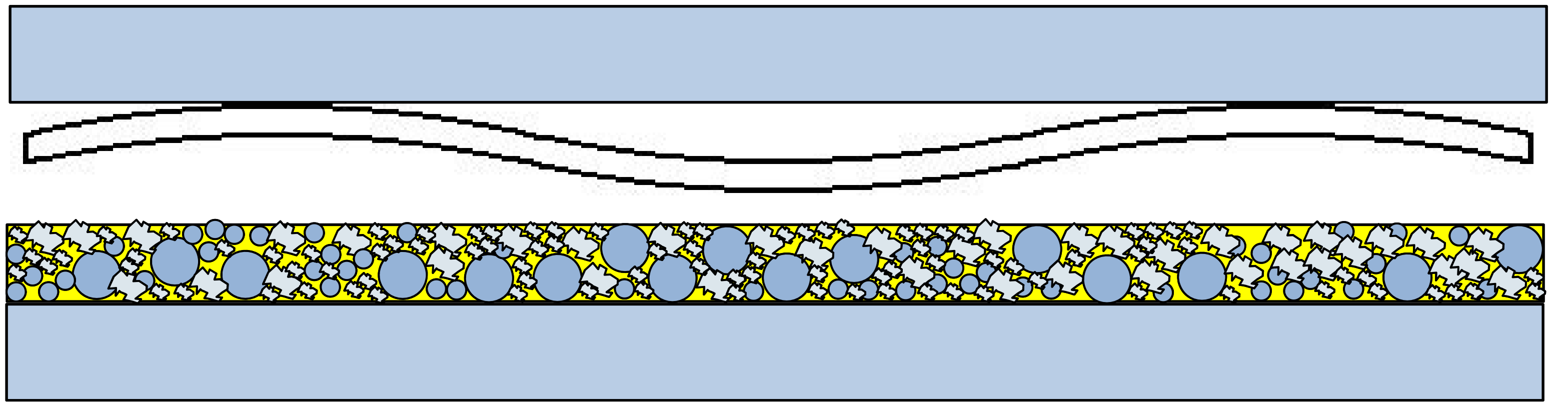

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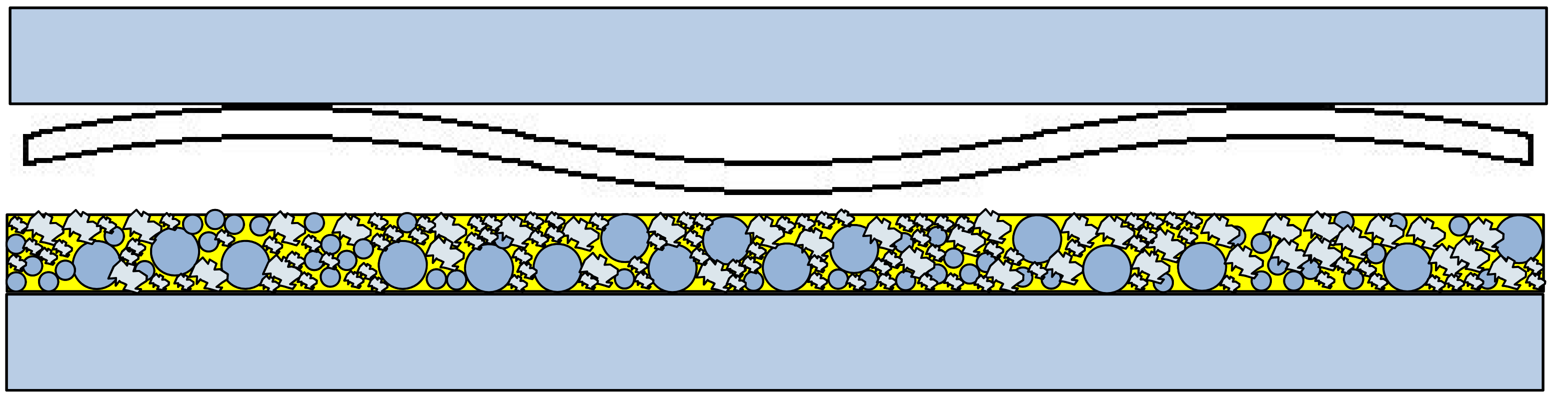

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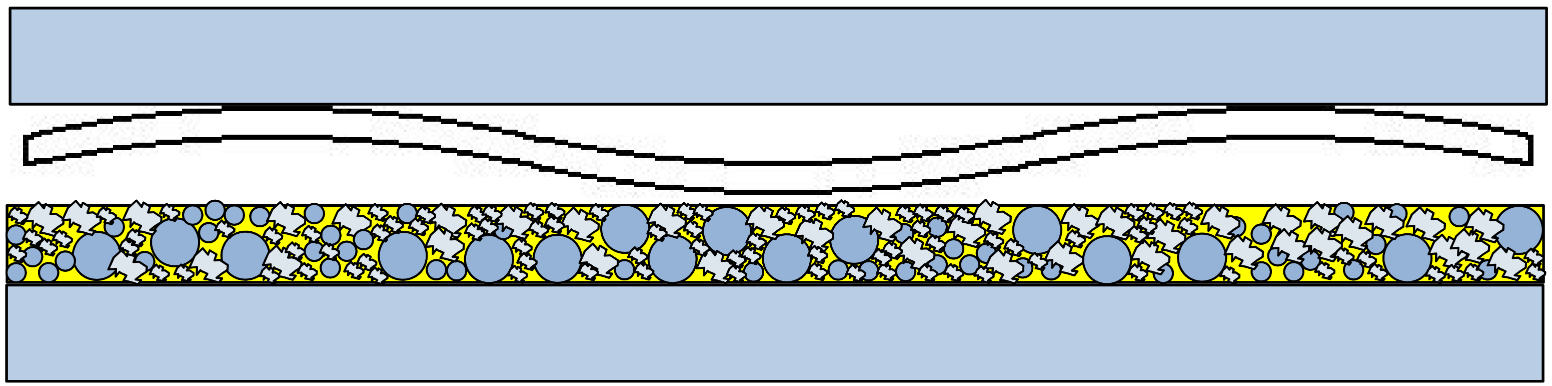

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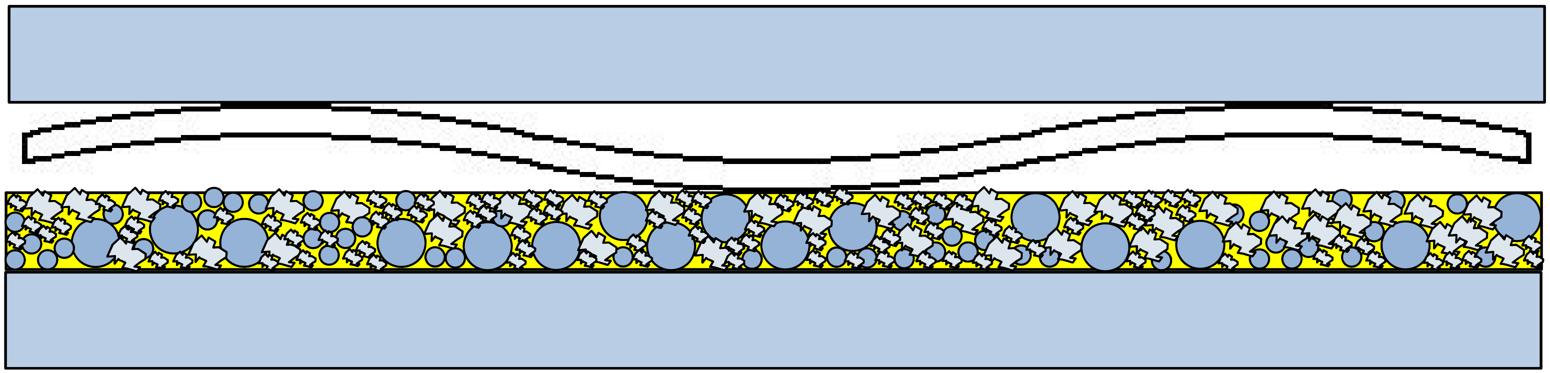

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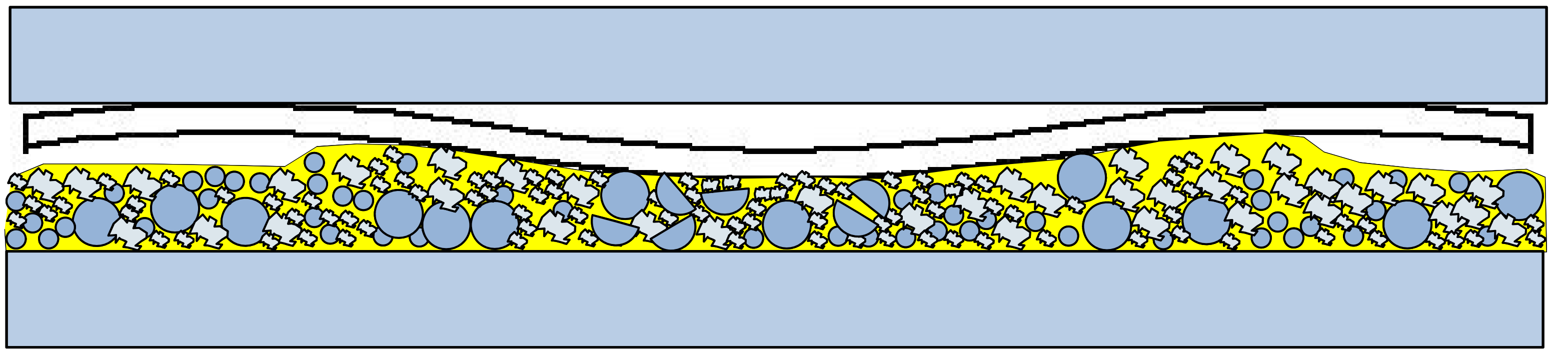

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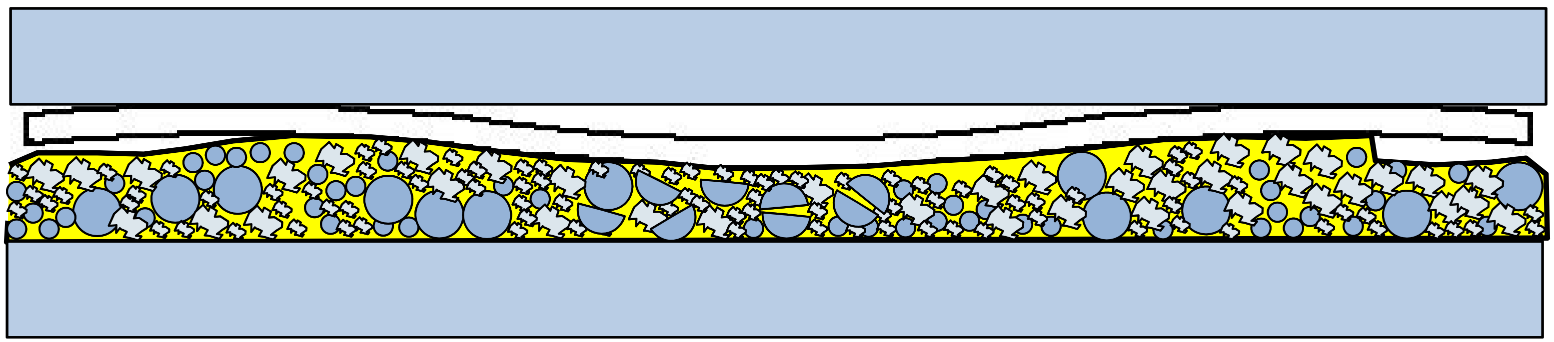

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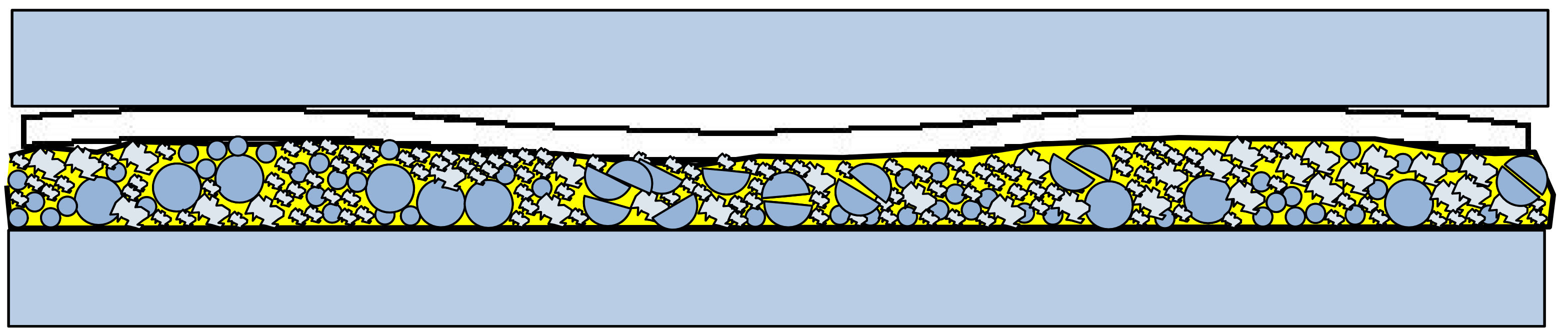

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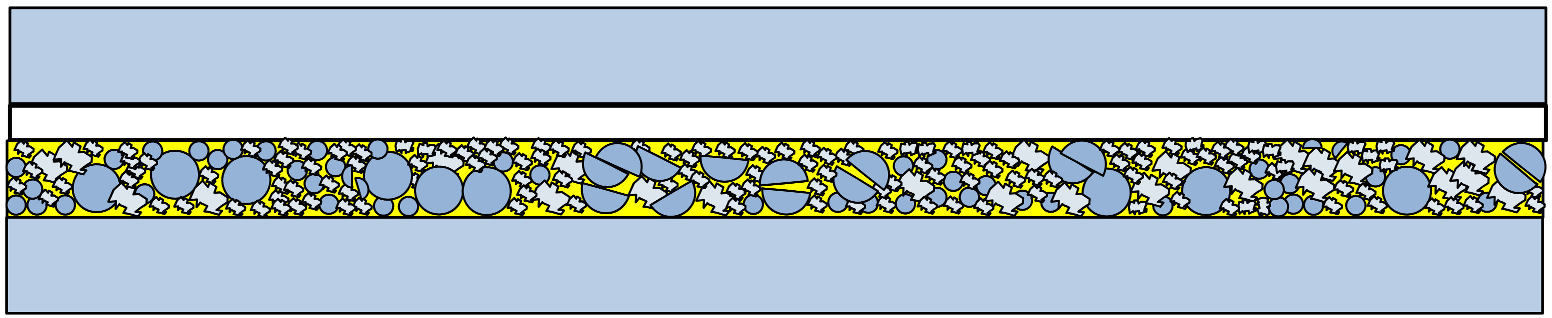

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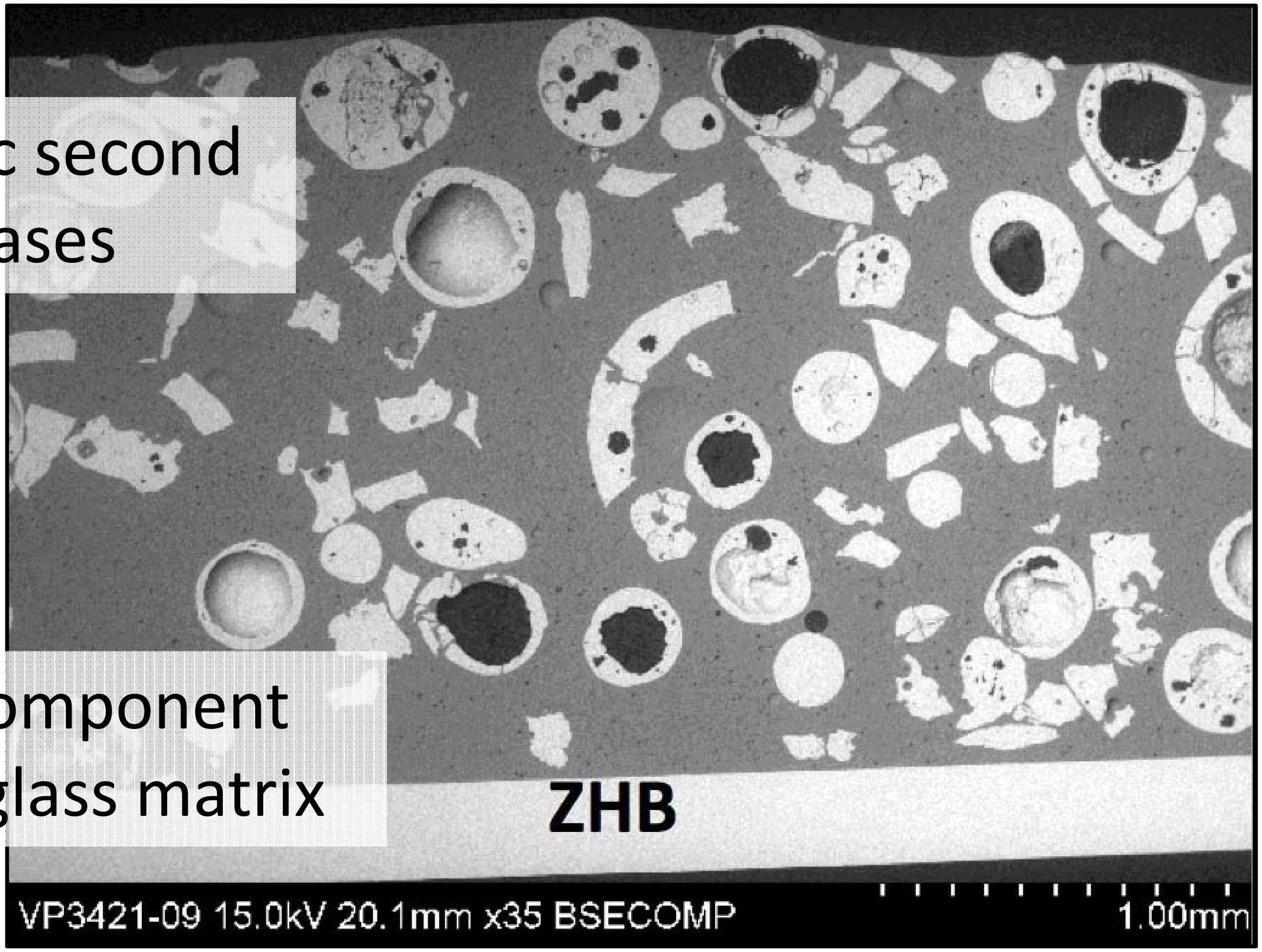
Engineered Glass Seals

ceramic second phases

Multicomponent silicate glass matrix



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Composition SCN Glass

As sintered

Element	at %
O	57.13
Si	25.64
K	2.85
Ba*	2.00
Na	3.84
Ca	2.32
Al	2.64
Mg	1.21
Ti	0.06
B	0.04
Zn	0.01

Atom Probe Tomography

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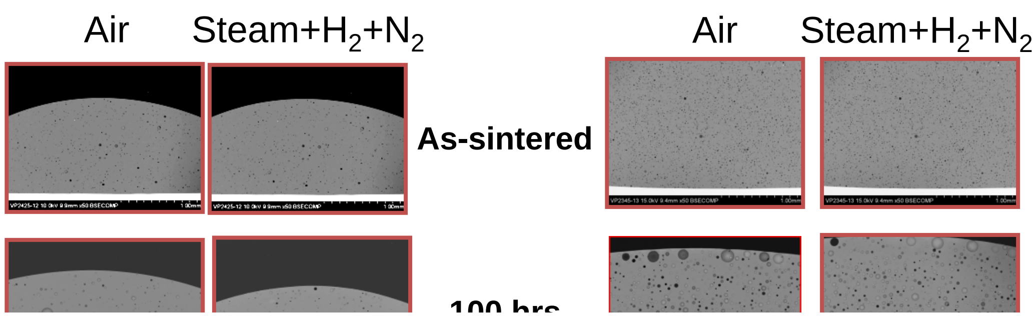
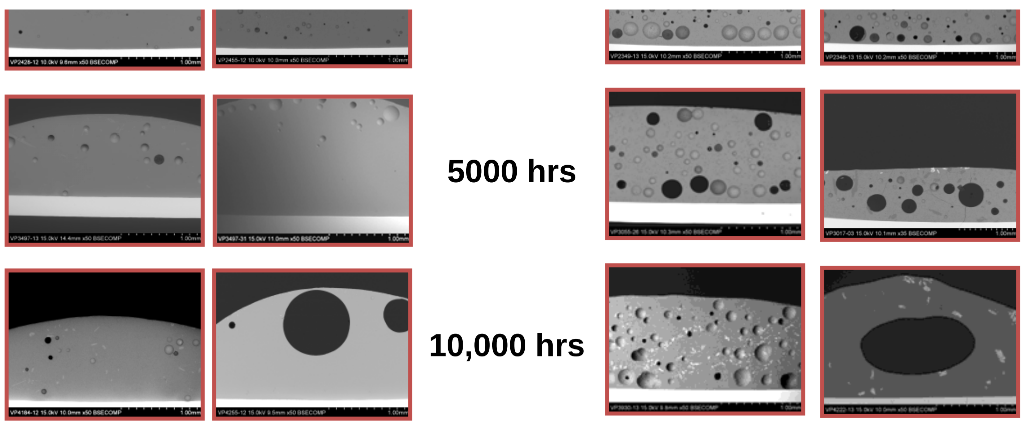
Microstructural Evolution of multicomponent silicate glasses



Ongoing exposure tests have surpassed 25,000 hrs



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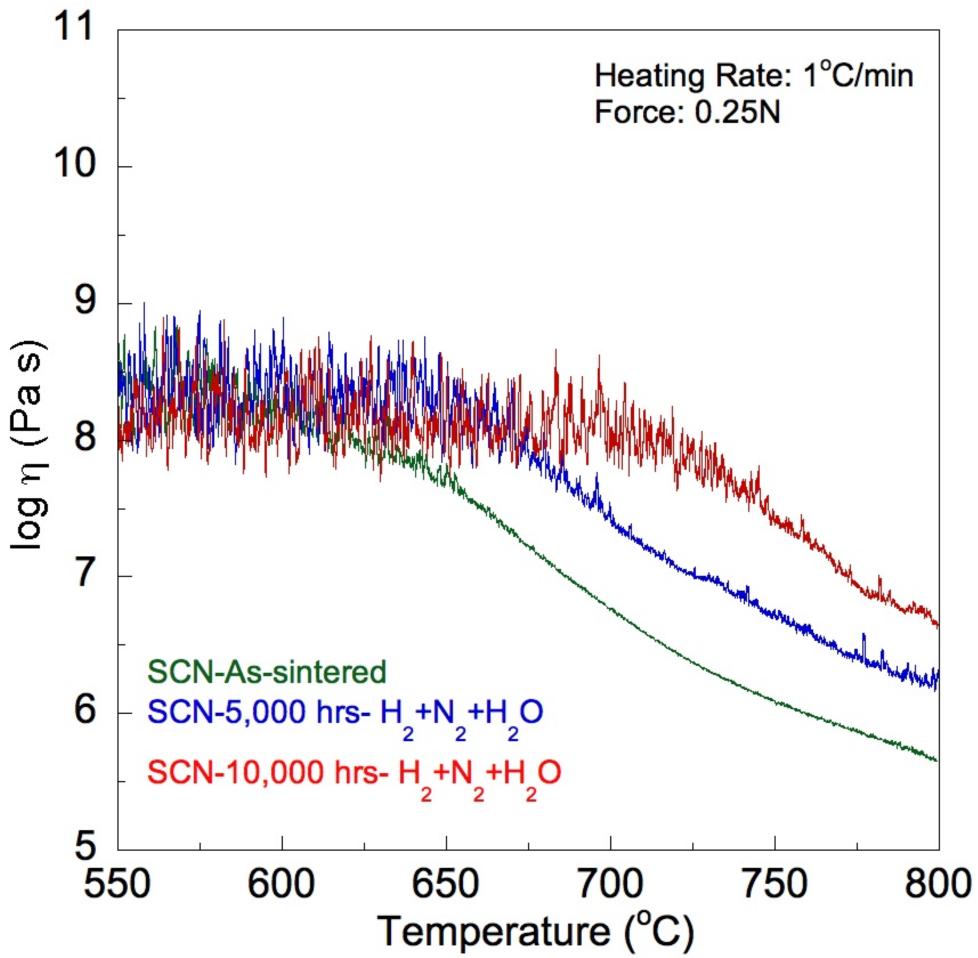
Viscosity of SCN Glass

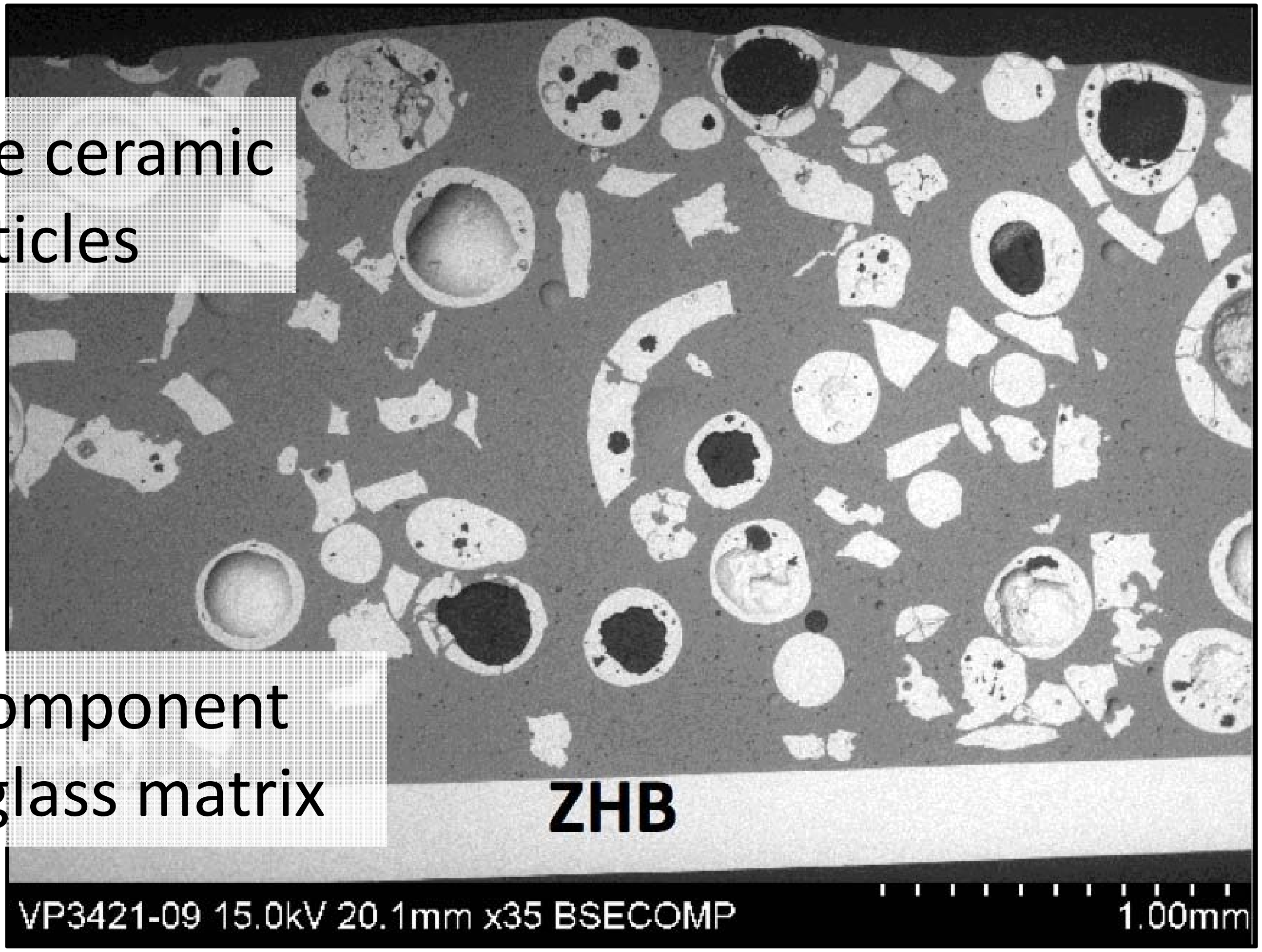


- Viscosity decreases with temperature and increases with time of exposure.
- Increase in viscosity could be explained by precipitation of crystalline phases.

Engineered Glass Seals

Frangible ceramic particles

Multicomponent silicate glass matrix



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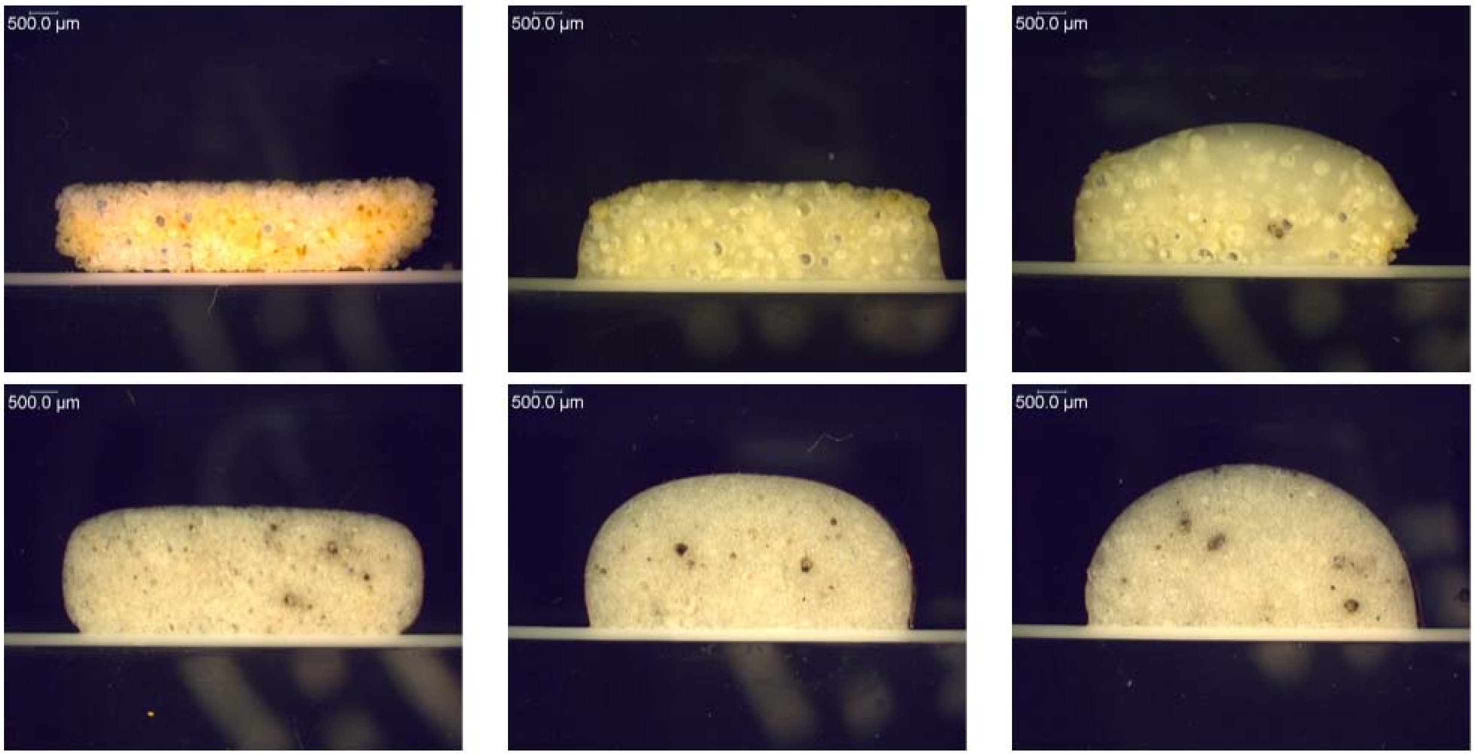
Wetting Behavior of Engineered Glass Seals

1:1 1:2 1:3



ZHB

Agsco

SCN glass

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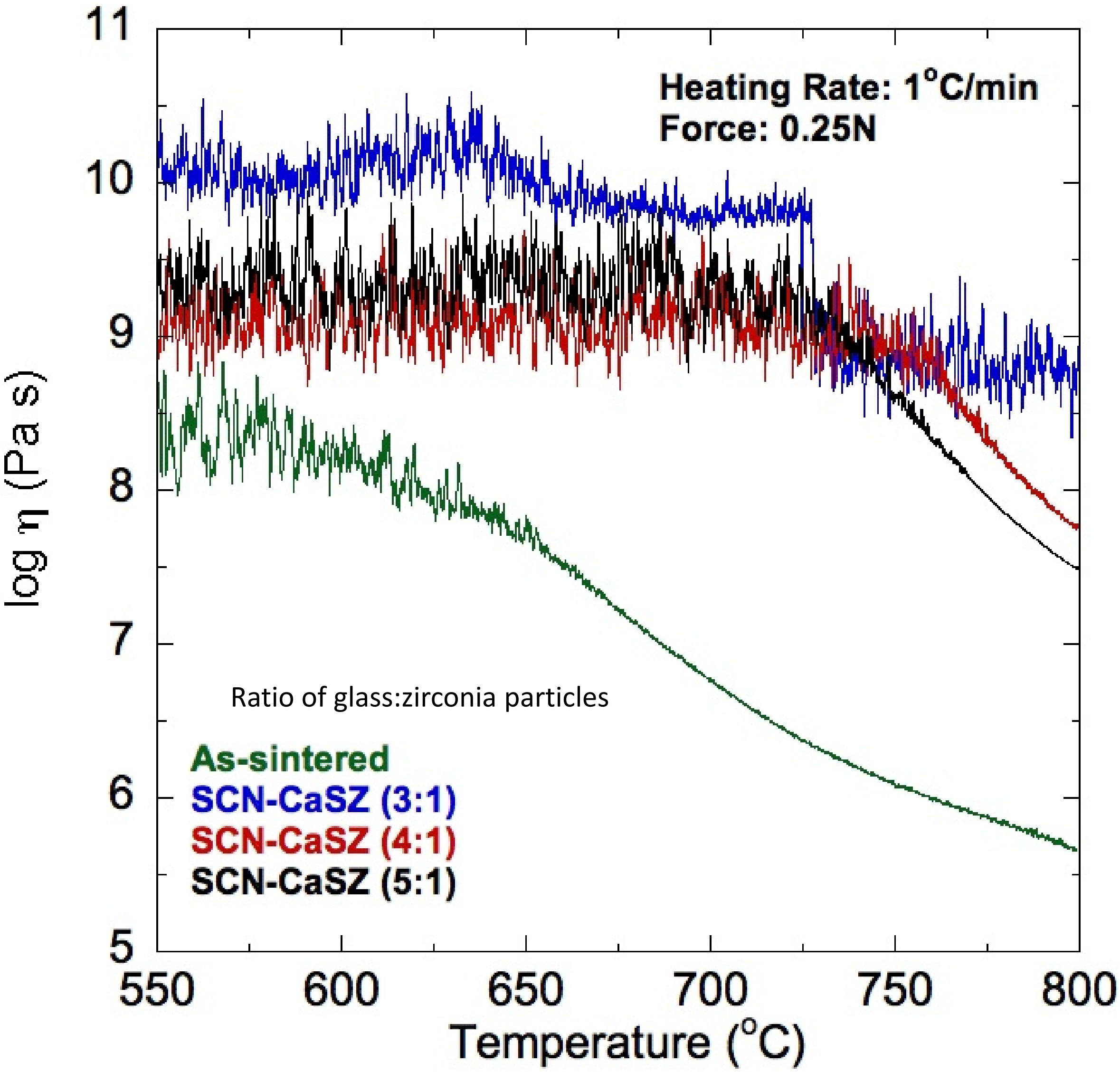
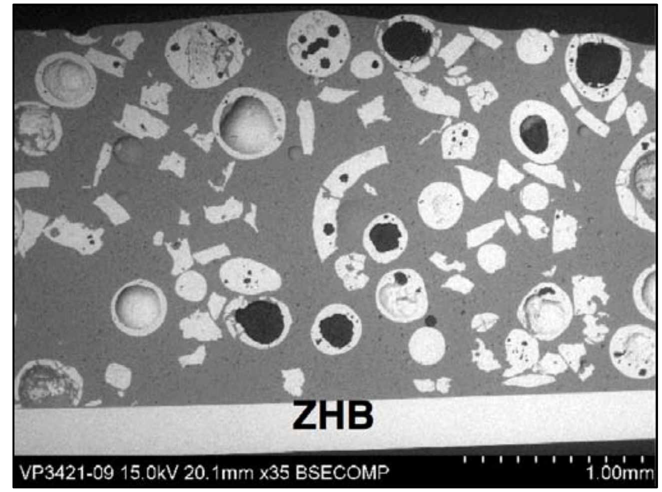
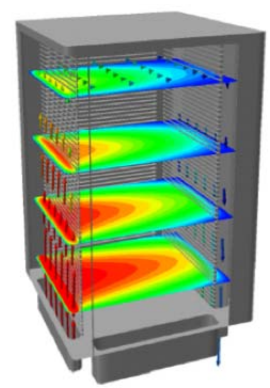
Viscosity of SCN glass containing zirconia hollow spheres





Frangible calcia-stabilized zirconia particles in SCN glass matrix

The viscosity of the seal can be tailored to accommodate the large temperature gradients in SOFCs during transients and steady state operation.



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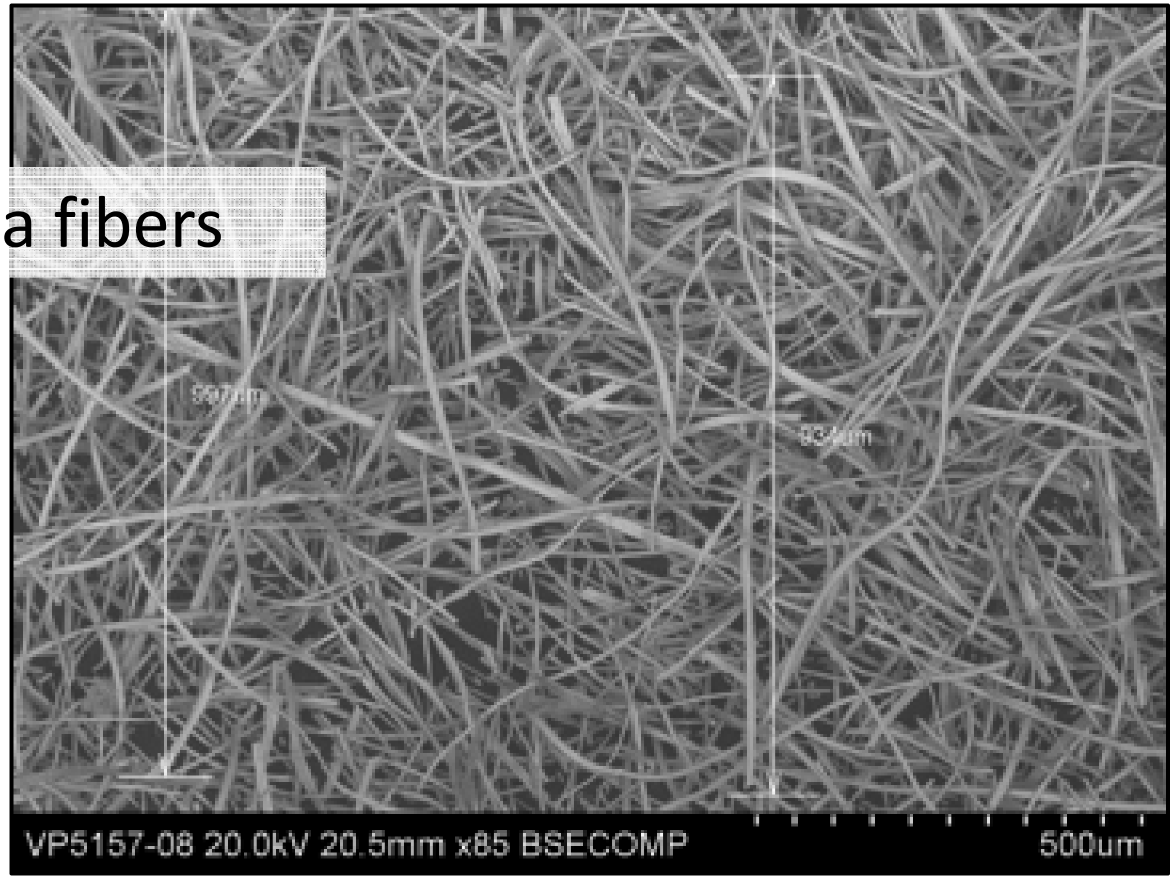
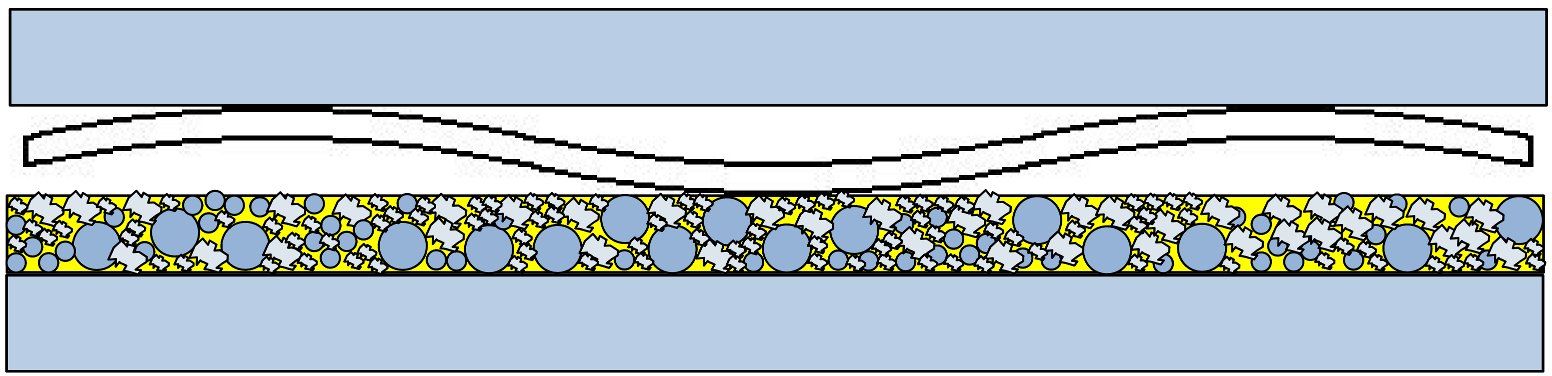
Engineered Glass Seals

zirconia fibers





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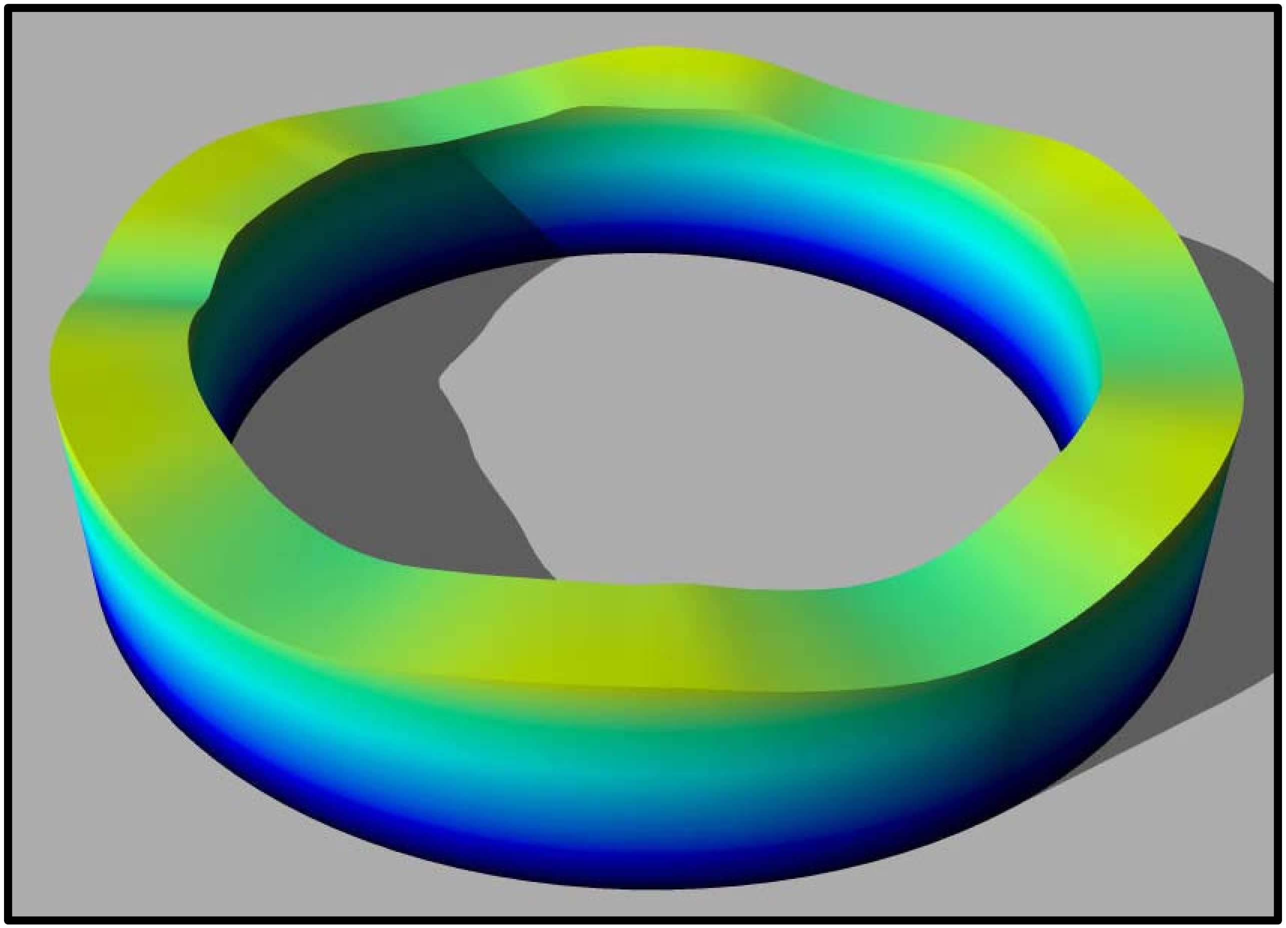
Zirconia Tube with Machined Sinusoidal Pattern



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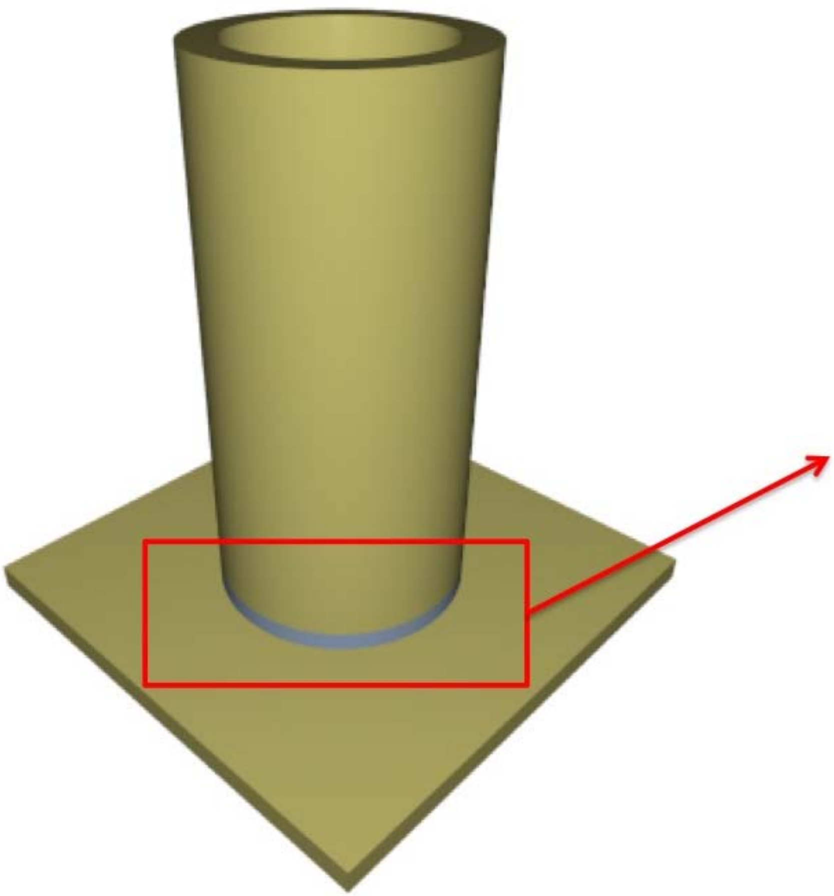
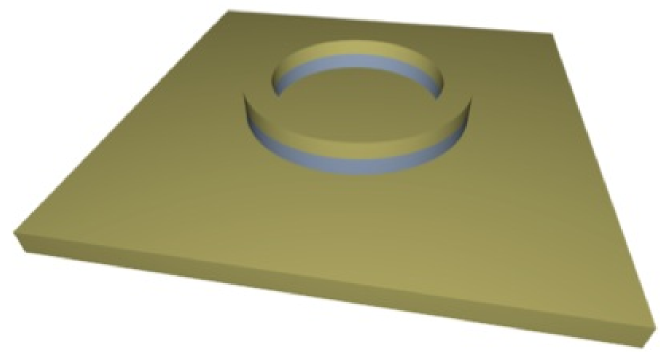
Zirconia tubes have been bonded to zirconia plates using engineered glass seal. This is the specimen configuration for testing in dual environments



Zirconia tube is cut and ground to enable imaging of bonded area using IR imaging.



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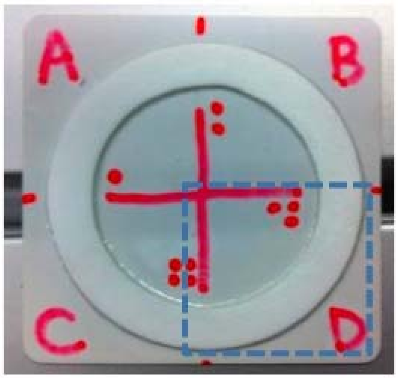
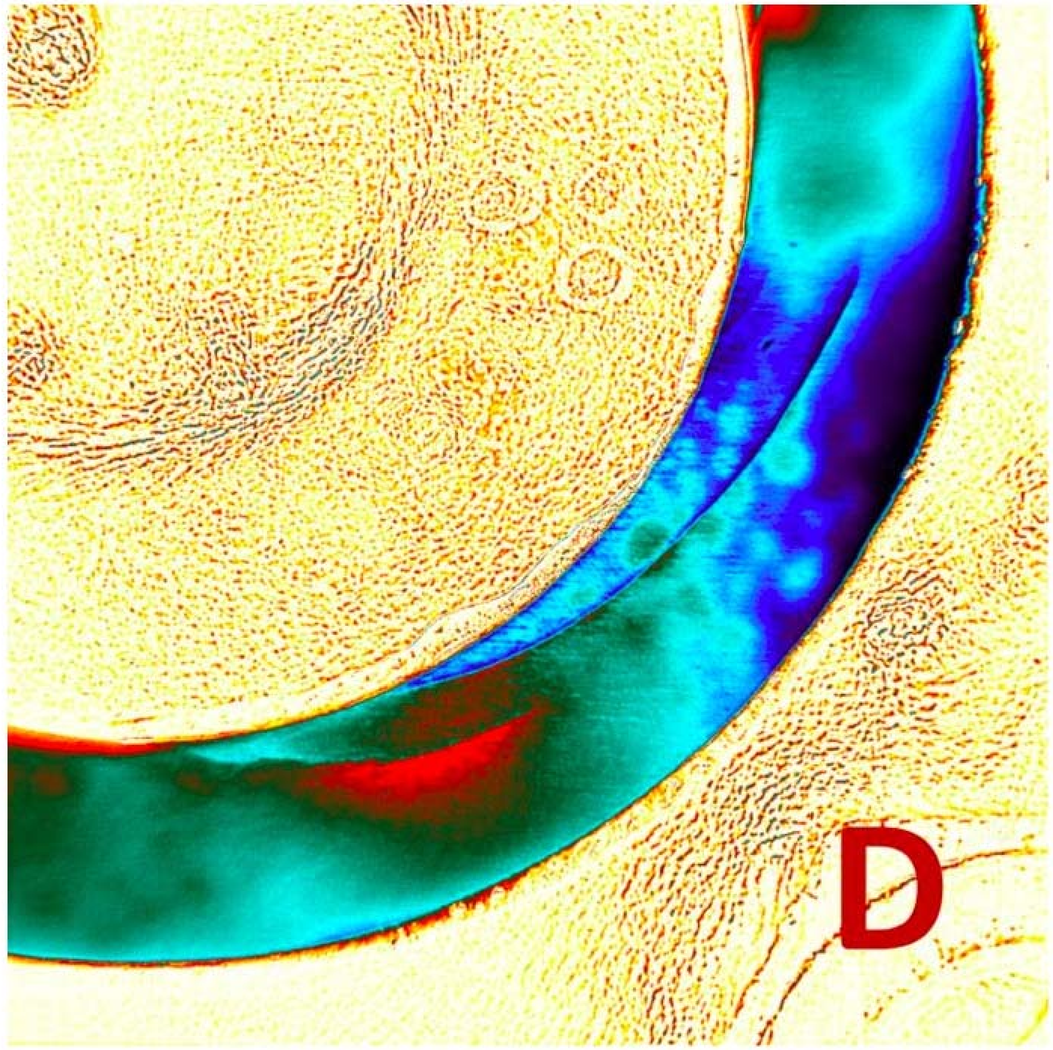
NDE of engineered glass seals





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Routes to low-cost manufacturing

- Tape casting
- Screen printing
- Fused deposition (3D Printing)

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Routes to low-cost manufacturing

- Tape casting

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Tape Casting

- SCN glass
- Hollow zirconia spheres
- Organic binder

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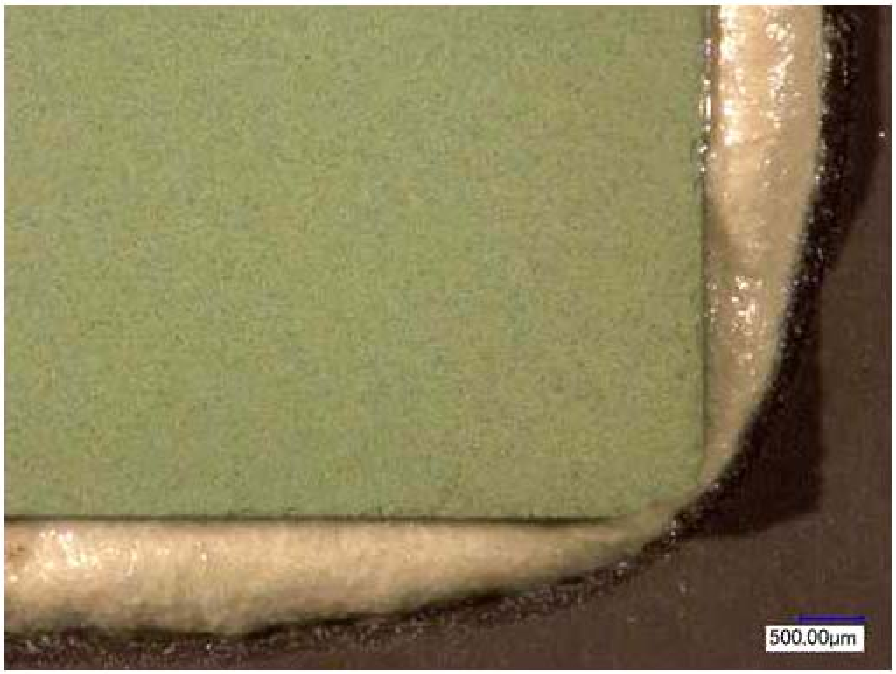
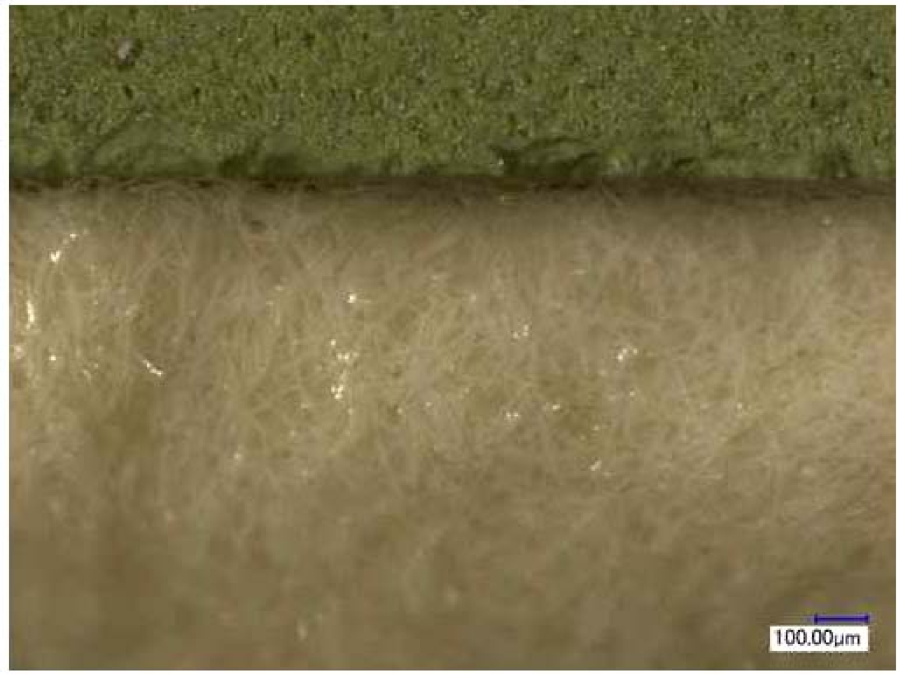
Post-mortem analysis of SCN-1 glass with 15V% ZrO_2 fibers after 12 thermal cycles

- No iso-propanol penetration along sealing edges or through bilayer
- No micro-cracks on sealing glass





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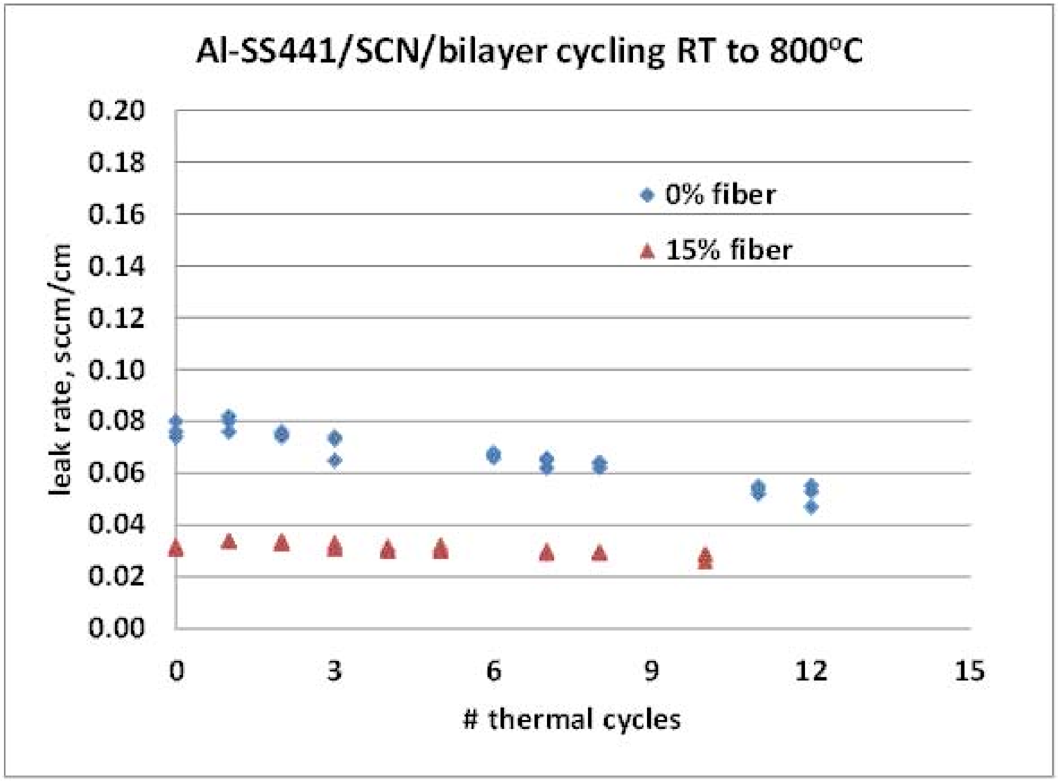
Thermal cycling of SCN-1 glass with ZrO_2 fibers

- ~40°C to 800°C in 3h, held for 3h at 800°C then furnace cooled to ~40°C in ambient air, 1 cycle/day.
- Constant leak rates suggested hermetic seal (observed leakage was from perimeter mica seal)
- Post-mortem check with iso-propanol showed no penetration



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Routes to low-cost manufacturing

- Tape casting
- Screen printing

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Screen-printed engineered glass seals

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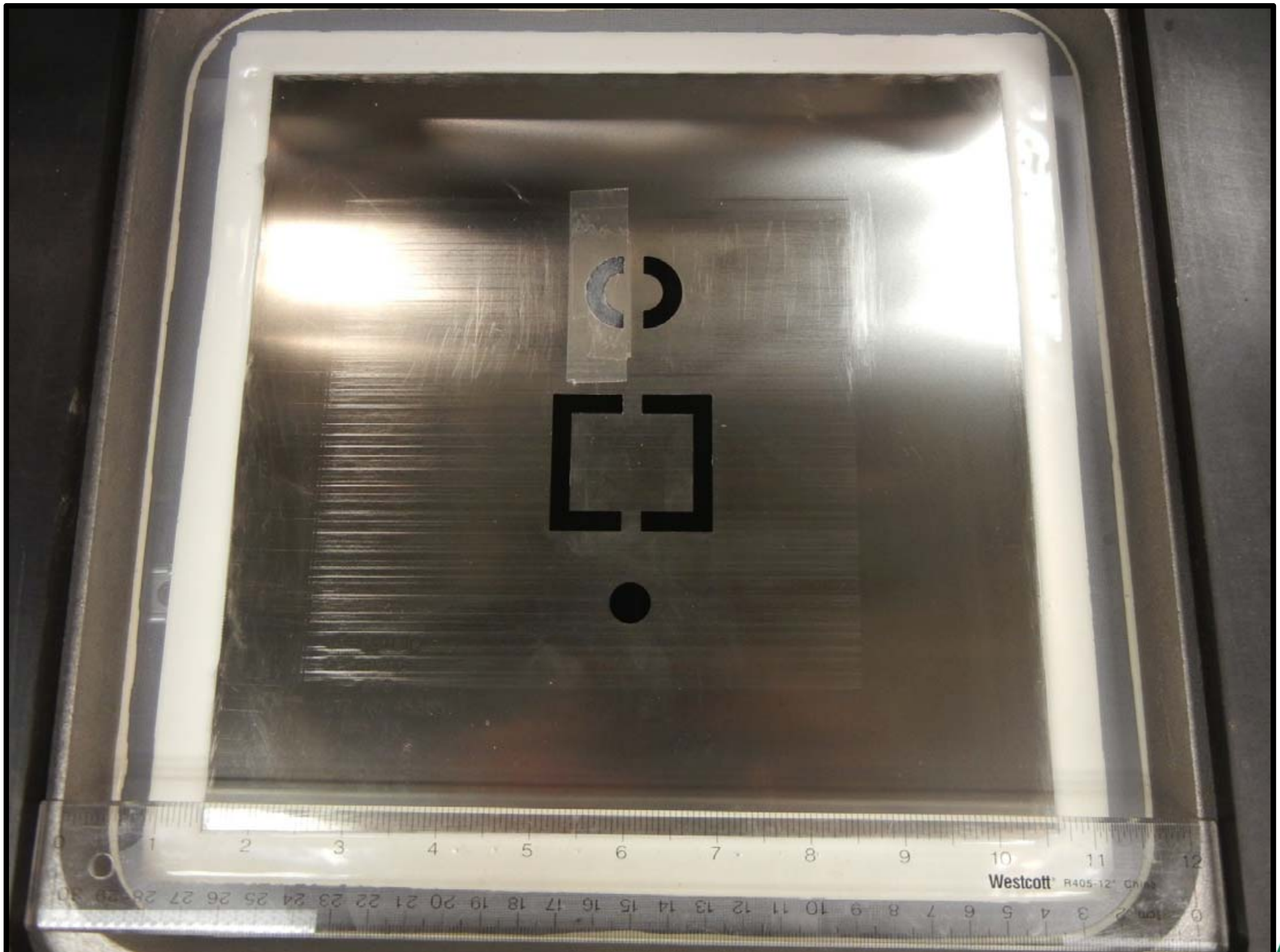
Screen-printed engineered glass seals



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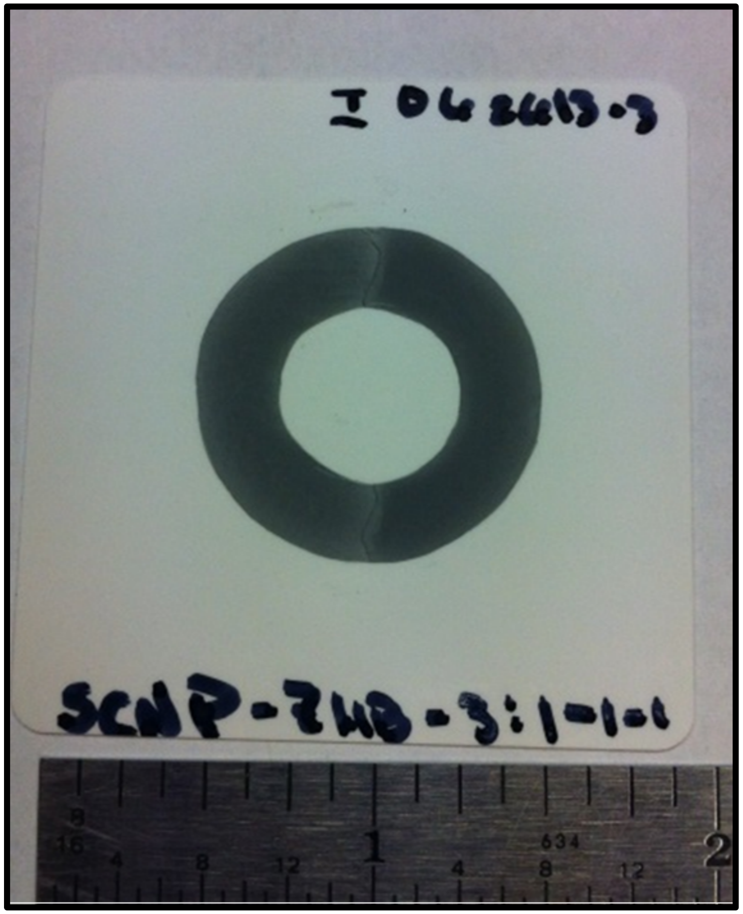
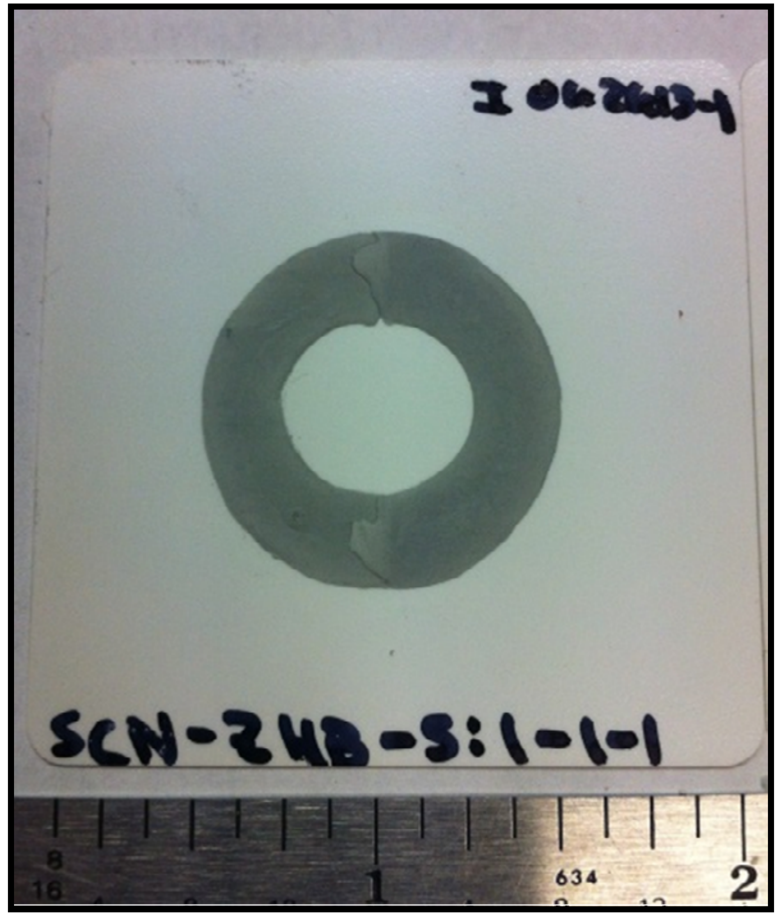
Screen-printed engineered glass seals





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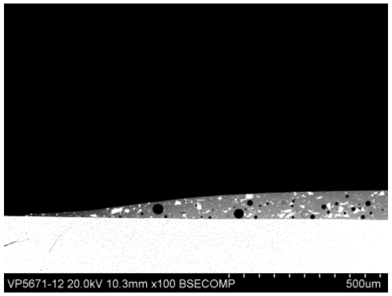
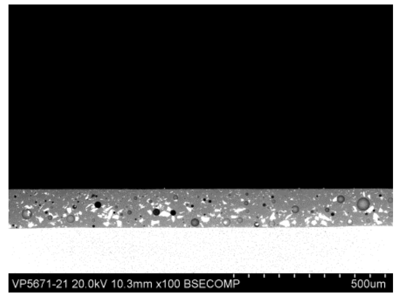
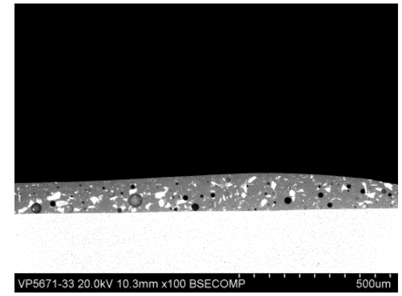
Screen-printed engineered glass seals

Left Side | Middle Side | Right Side

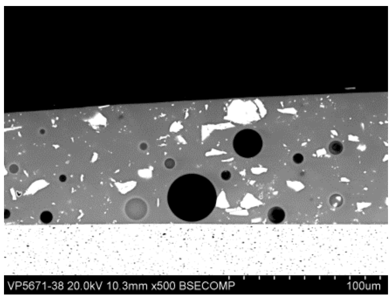
100X

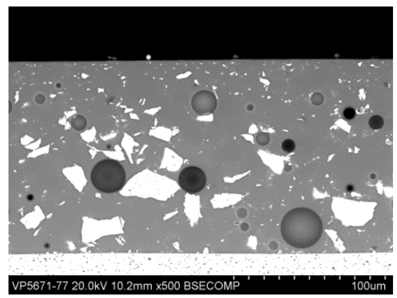


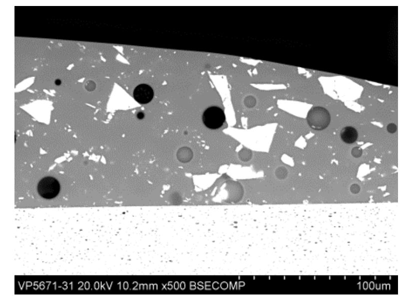




500X







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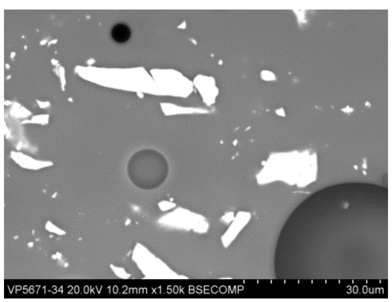

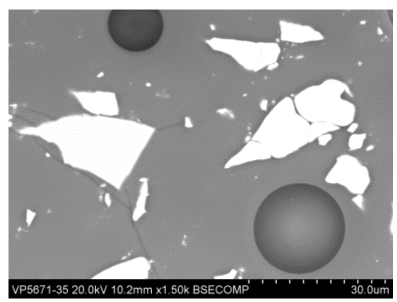

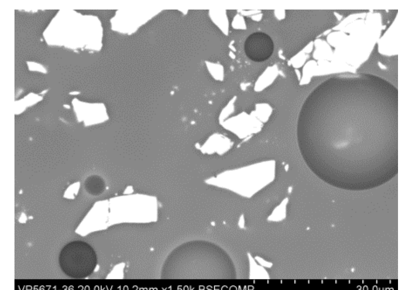

SCN-ZHB-(3:1)

Cross-sectional view

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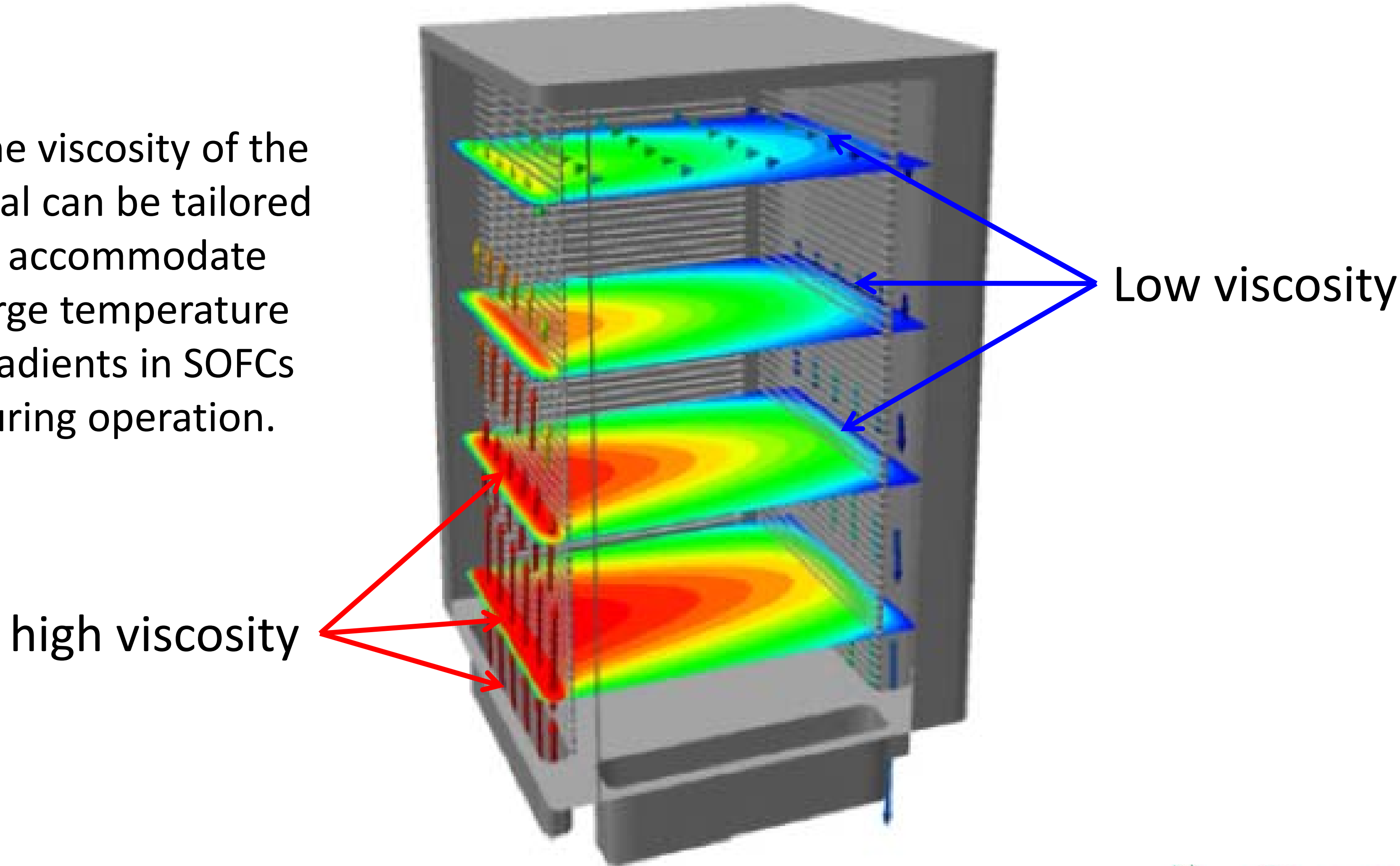
Viscosity of engineered glass seals

The viscosity of the seal can be tailored to accommodate large temperature gradients in SOFCs during operation.



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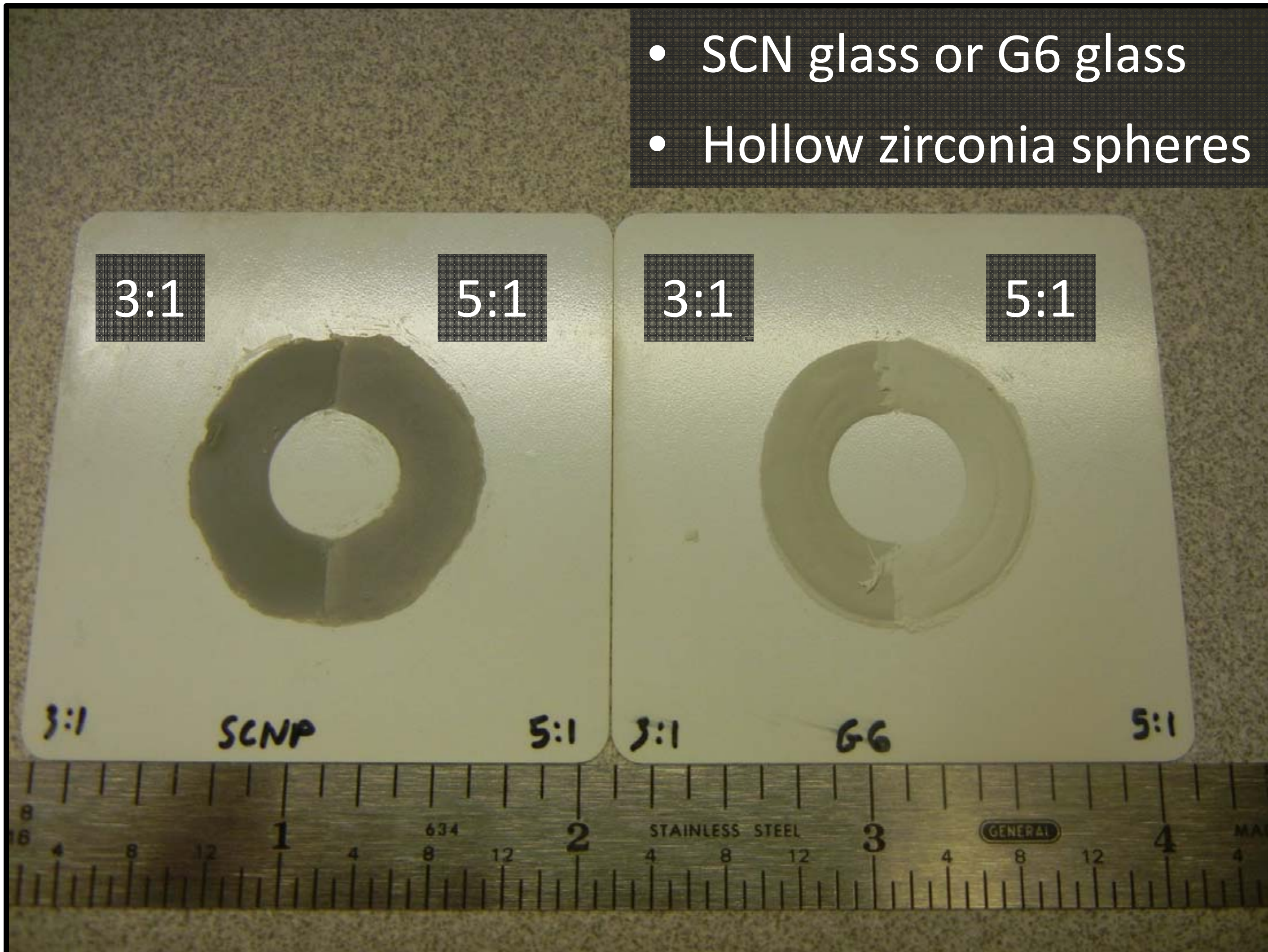
Screen-printed engineered glass seals



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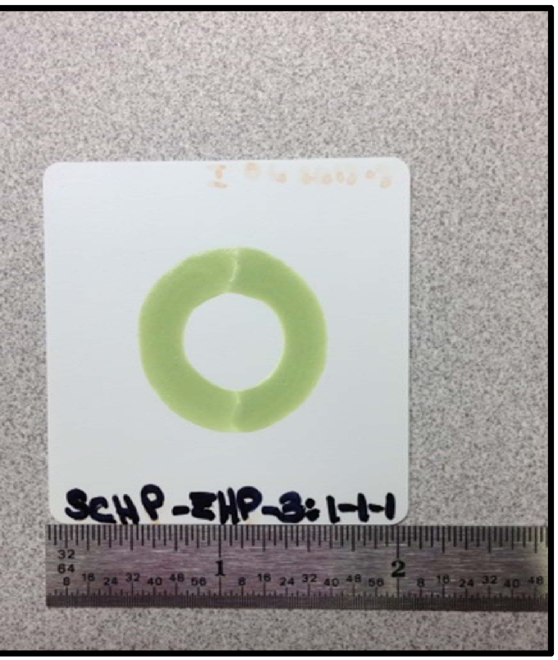
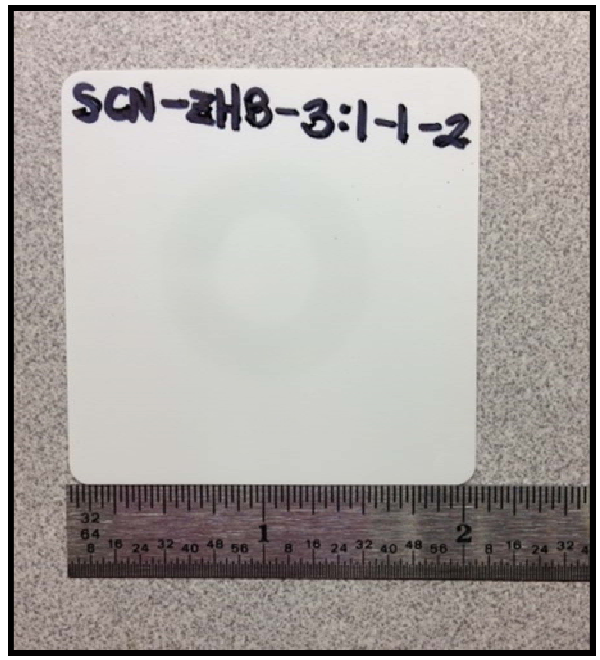
een-printed engineered glass seals

After Sintering





Sintering Process:

Up to 600 °C in 6 hrs, then up to 850 °C to 2 hrs.

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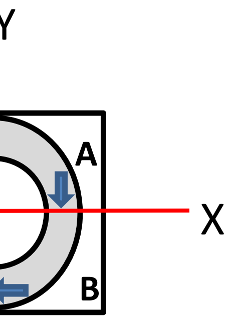
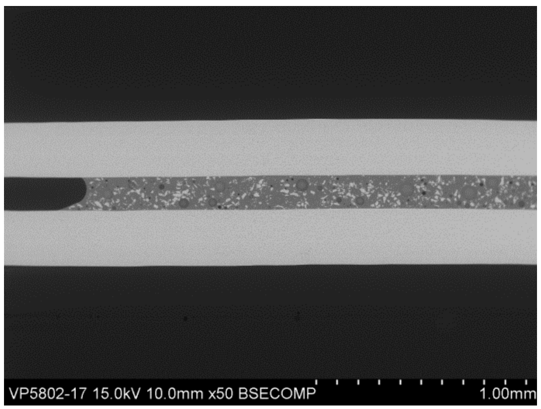
een-printed engineered glass seals

SCN-ZHB-3:1-5:1 (sandwich)



Left Edge



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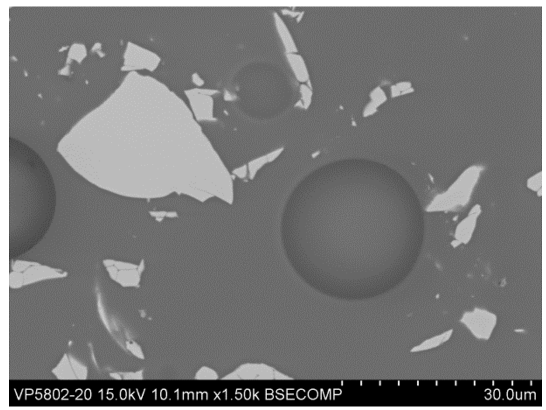

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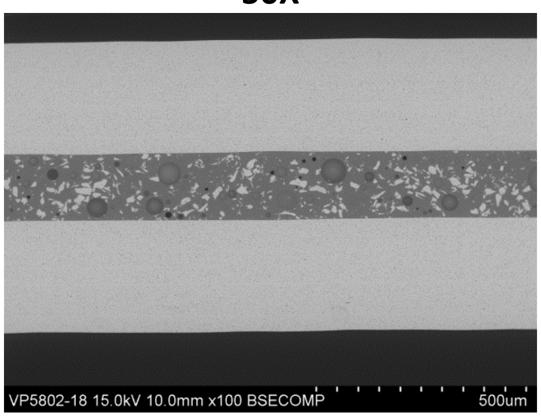
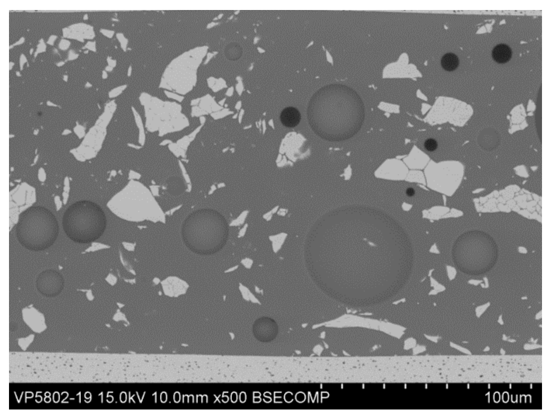


100X



500X

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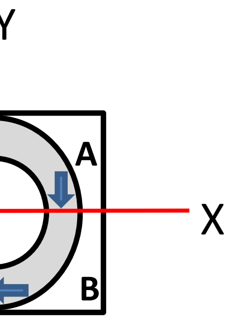
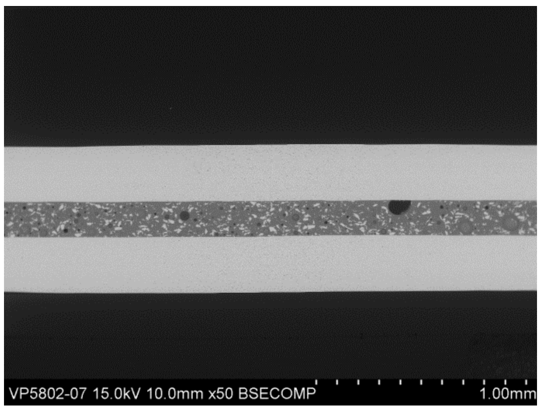
een-printed engineered glass seals

Middle Side

SCN-ZHB-3:1-5:1 (sandwich)





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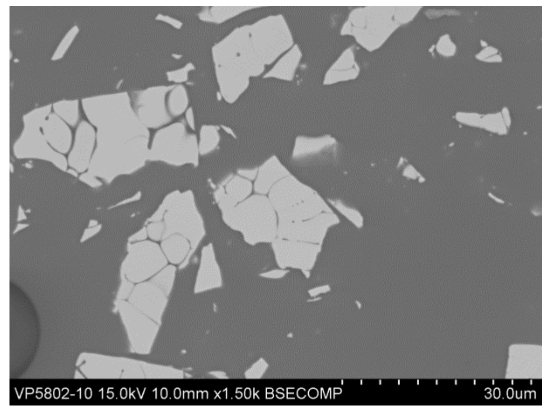

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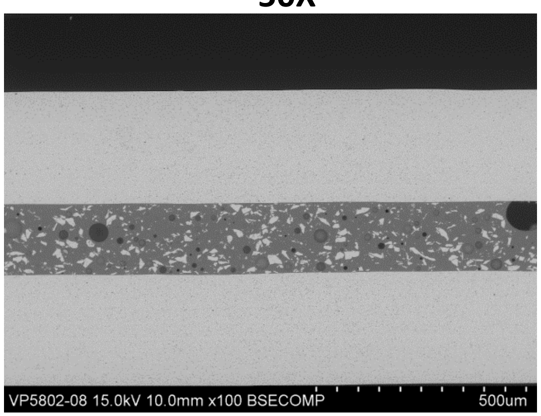

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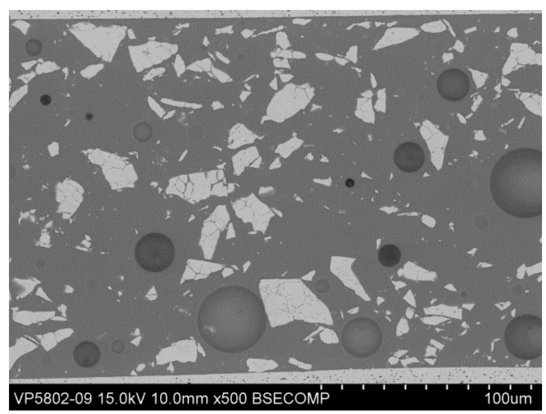

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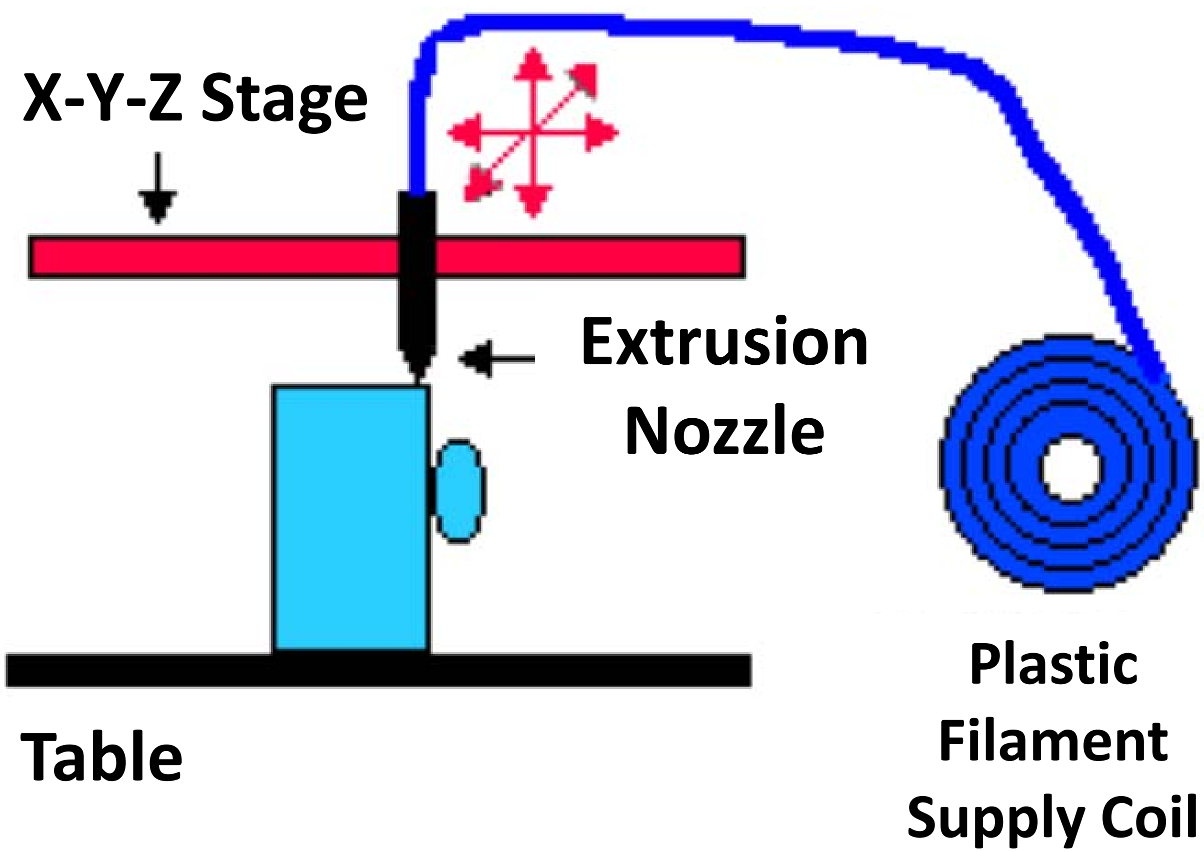
utes to low-cost manufacturing

Tape casting

Screen printing

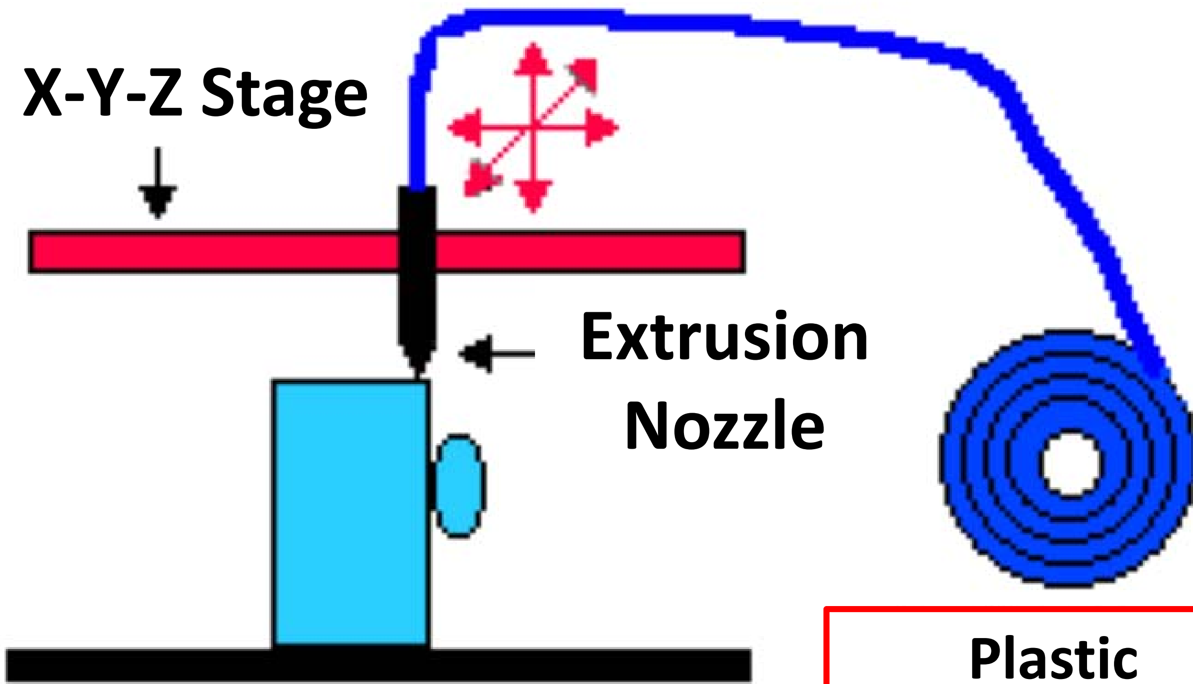
Fused deposition (3D Printing)

sed Deposition (3D Printing)



ttp://www.additive3d.com

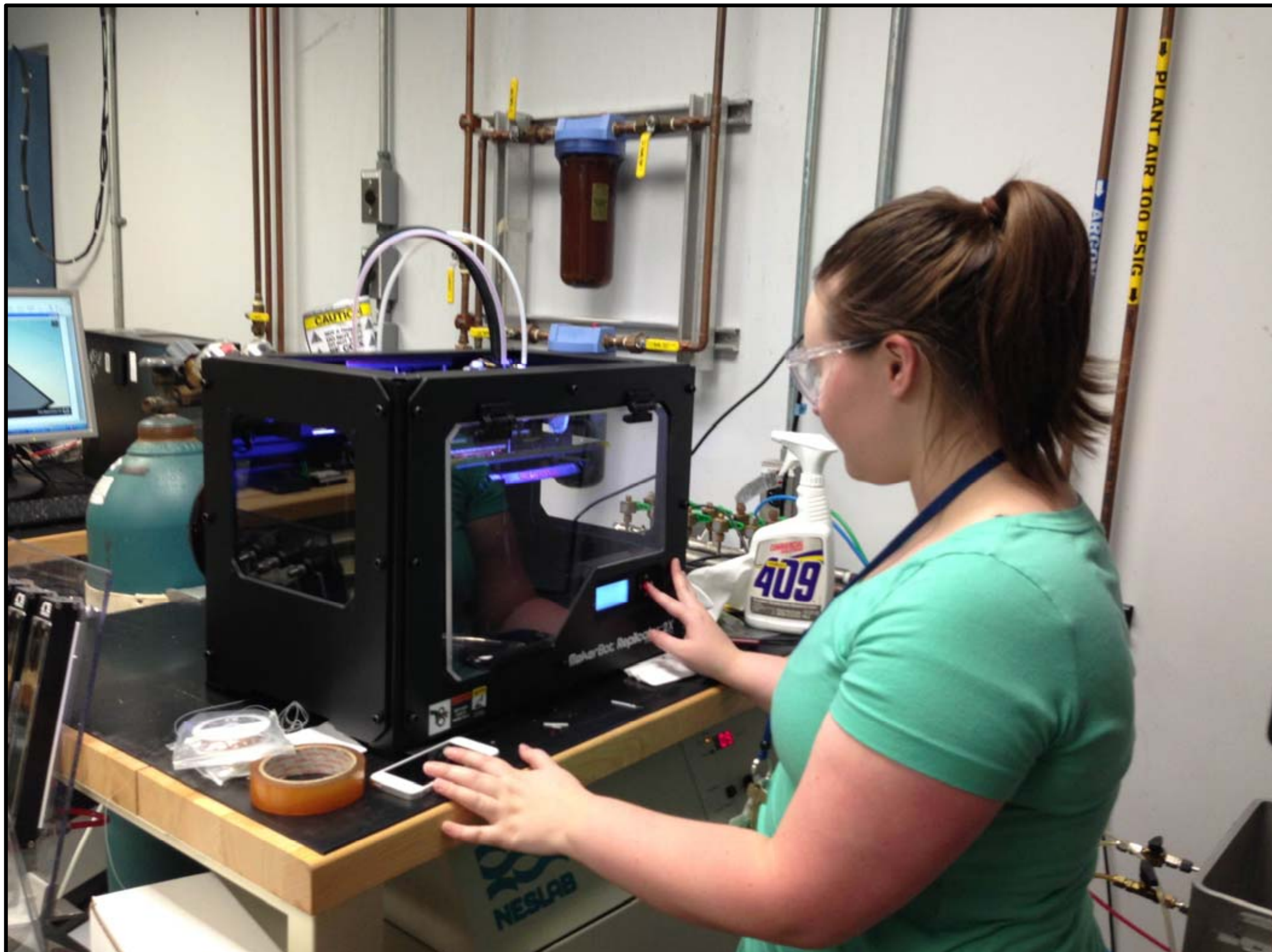
sed Deposition (3D Printing)



Plastic Filament Supply Coil

omposite mixture of glass,
ramic 2nd phase and binder

sed Deposition (3D Printing)



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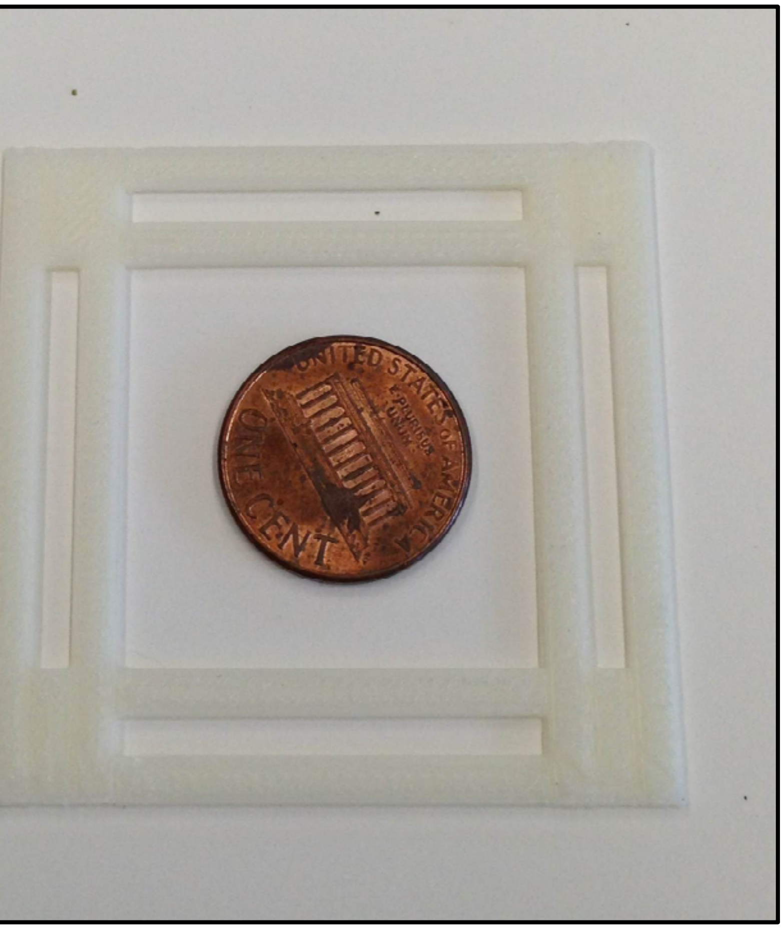
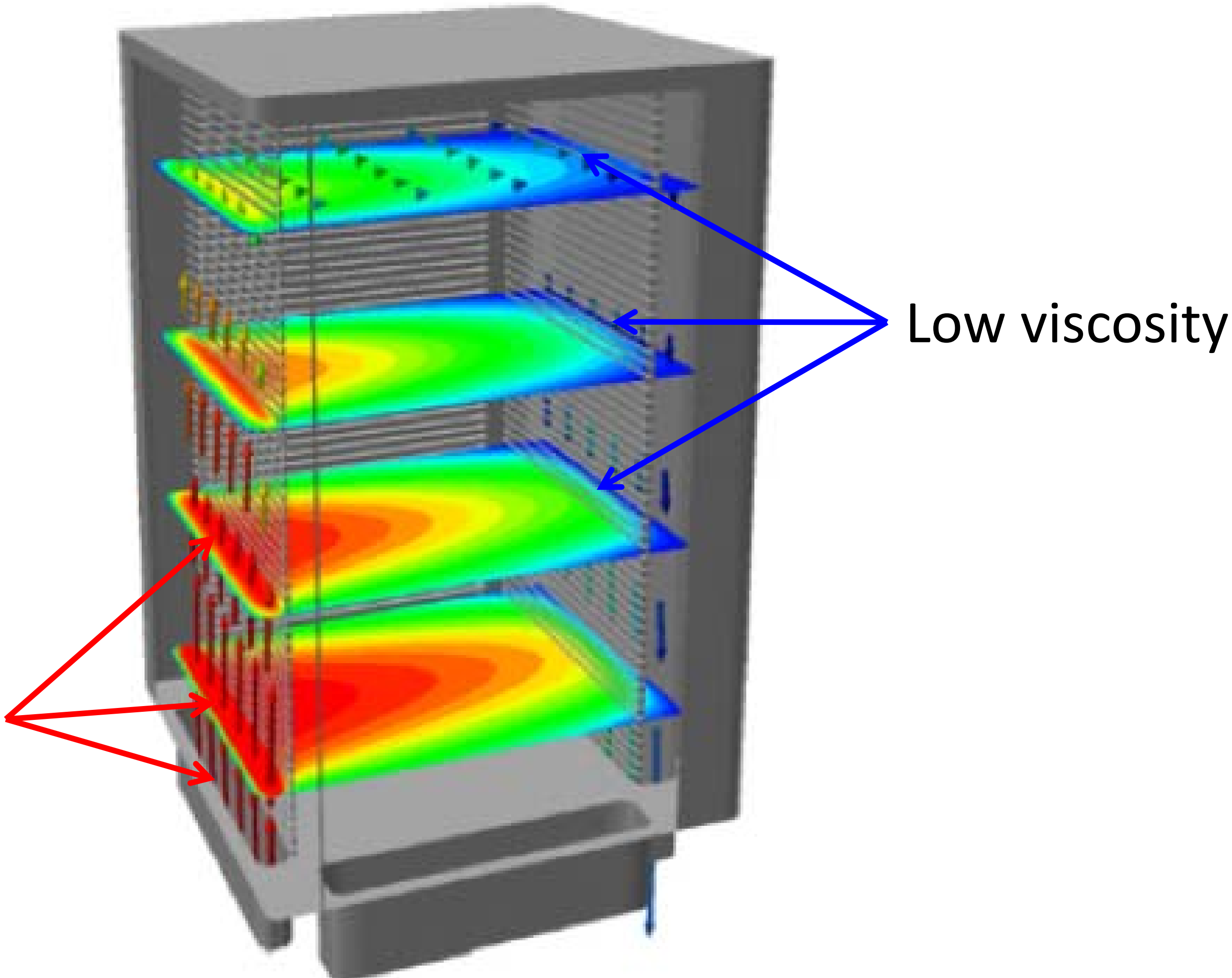


- We are using a 3-D printer to manufacture engineered glass seals with predetermined concentration values of glass and ceramic particles/ceramic fibers.
- The concentration of second phases and distribution of sizes would depend on topographic features of the cell and maximum local temperature.

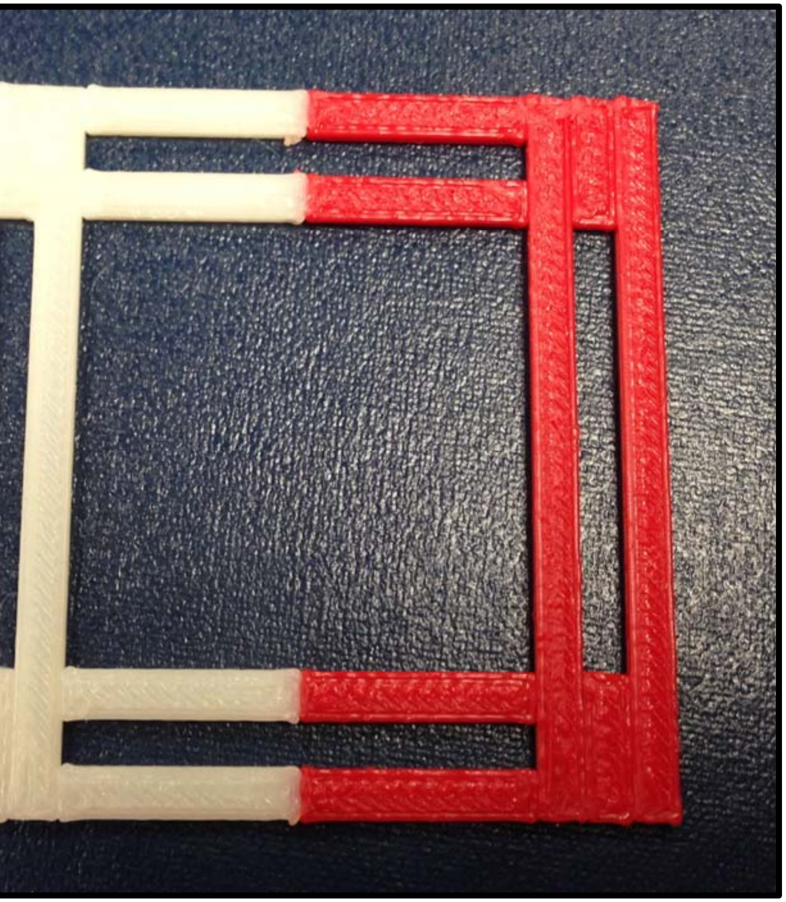
OAK

Viscosity of SCN glass containing zirconia particles

iscosity of the seal
e tailored to
mmodate the large
erature gradients
FCs during
ients and steady
operation.



sed Deposition (3D Printing)



- We are using a 3-D printer to manufacture engineered glass seals with predetermined concentration values of glass and ceramic particles/ceramic fibers.
- The concentration of second phases and distribution of sizes would depend on topographic features of the cell and maximum local temperature.

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Engineered glass seals are being developed for SOFCs.

Designs consist of a multi-component silicate glass matrix and a ceramic second phase (hollow spheres, fibers, particles).

The effectiveness and durability of these seals are being investigated.

Low-cost manufacturing processes are being developed.

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RIDGE