Compressive Seal Development

SECA
Core Technology Program Review

Pacific Northwest National Laboratory
YS Matt Chou

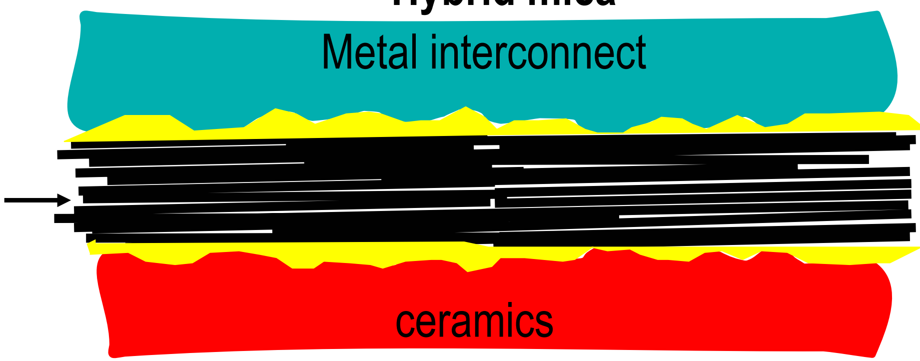
September 30-October 01, 2003
Albany, NY

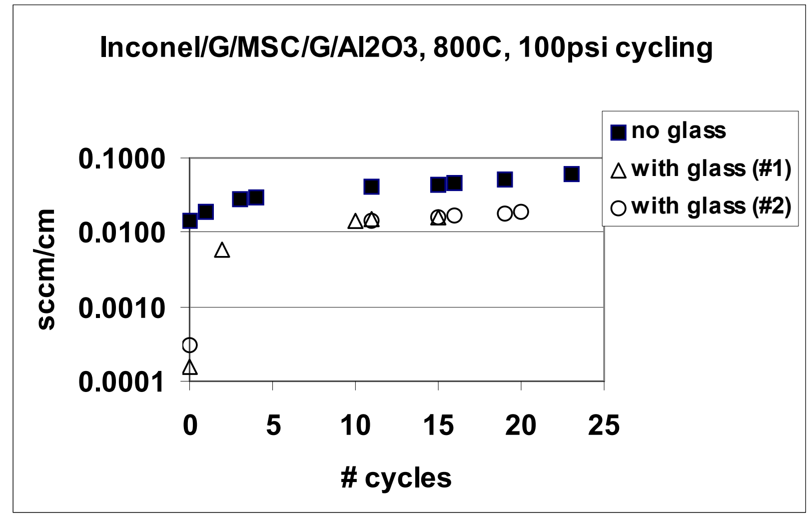
Outline

- Previous work and problem review
- Objectives
- Results and discussion
  - Effect of mica thickness on the leak rates during cycling
  - Concept of infiltration
  - Leak test of H_3BO_3-infiltrated Phlogopite mica
  - Leak test of Bi-nitrate-infiltrated Phlogopite mica for 3 metal couples
  - Leak test of glass-mica composite in hybrid form
  - OCV test of dense 8YSZ with various hybrid micas
  - OCV test and thermal cycling of 2”x2” 8YSZ with hybrid micas
- Summary
- Future work

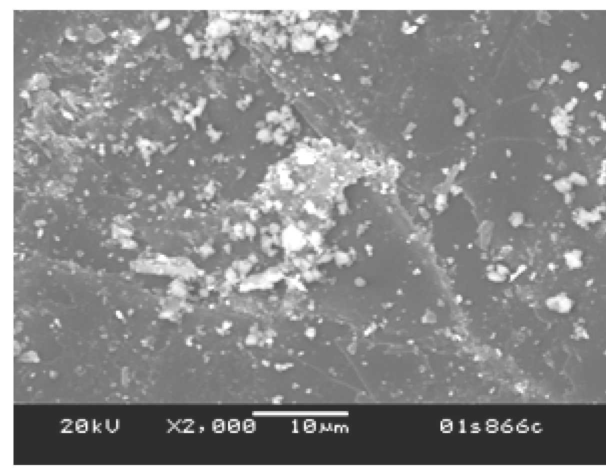
Previous work on monolithic Muscovite mica and problem review

Hybrid mica





Muscovite mica degradation: wear particle through-thickness crack

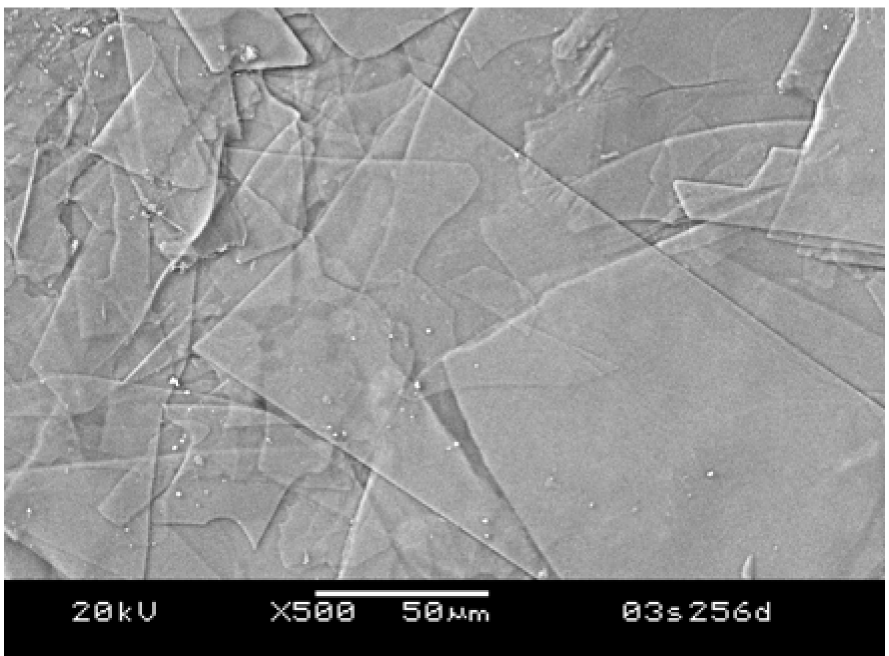


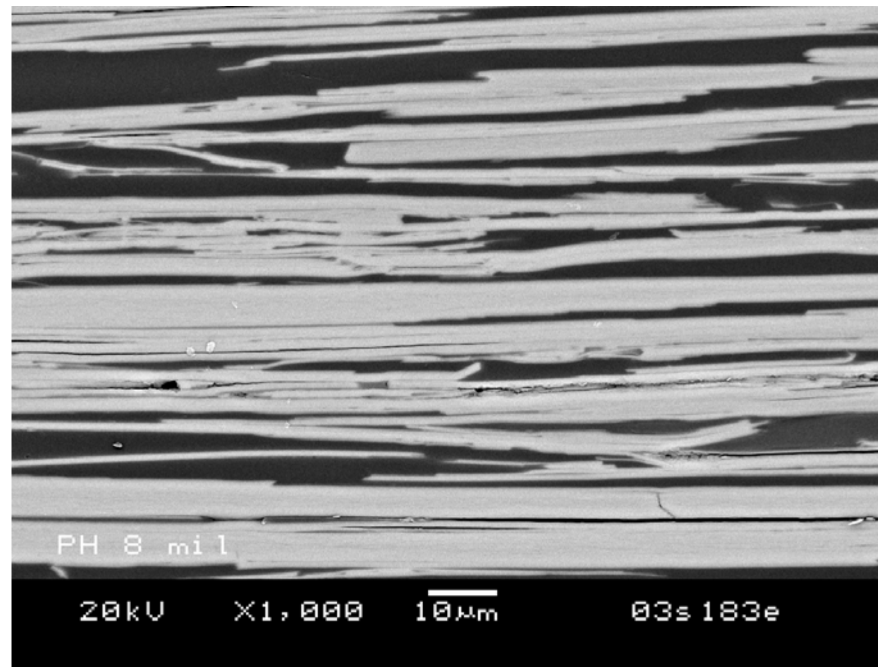
Objectives

- Evaluate a higher CTE mica (Phlogopite) as the compressive seals in terms of the thermal cycle stability.
- Test the concept of “infiltrated” Phlogopite micas for lower leak rates and the stability in thermal cycling.
- Assess the seal capability and cycling stability of hybrid Phlogopite micas through open circuit voltage measurement of medium-sized (2”x2”) dense 8YSZ plates.

Experimental: materials

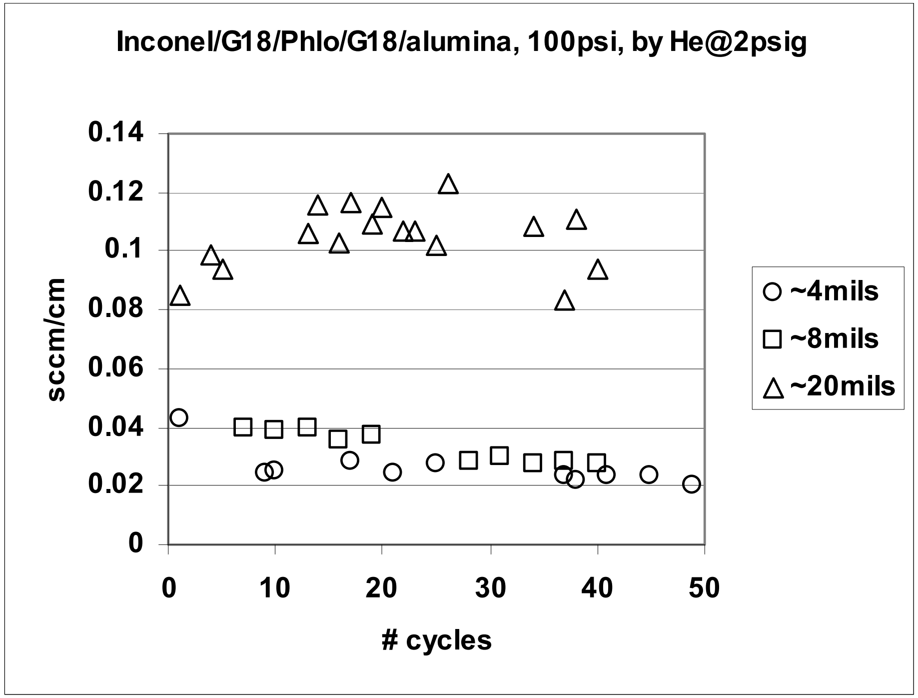
- Phlogopite mica not available in the monolithic (single crystal) form
- Paper form of various thickness: ~3mil, ~4mil, ~8mil, and ~20 mils
- Higher CTE (~11 ppm/^{o}C) than Muscovite (~7 ppm/^{o}C), but in paper form, contains leak path in 3-D

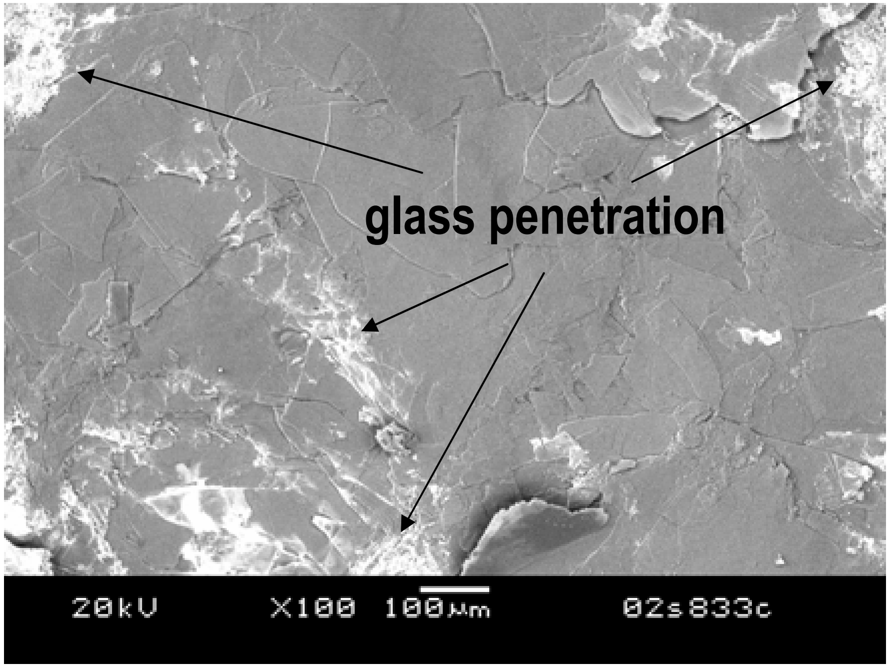




Thickness effect:
Lower leak rates for thinner Phlogopite micas

Mica paper pressed between Inconel600 pipe and alumina substrate at 100psi



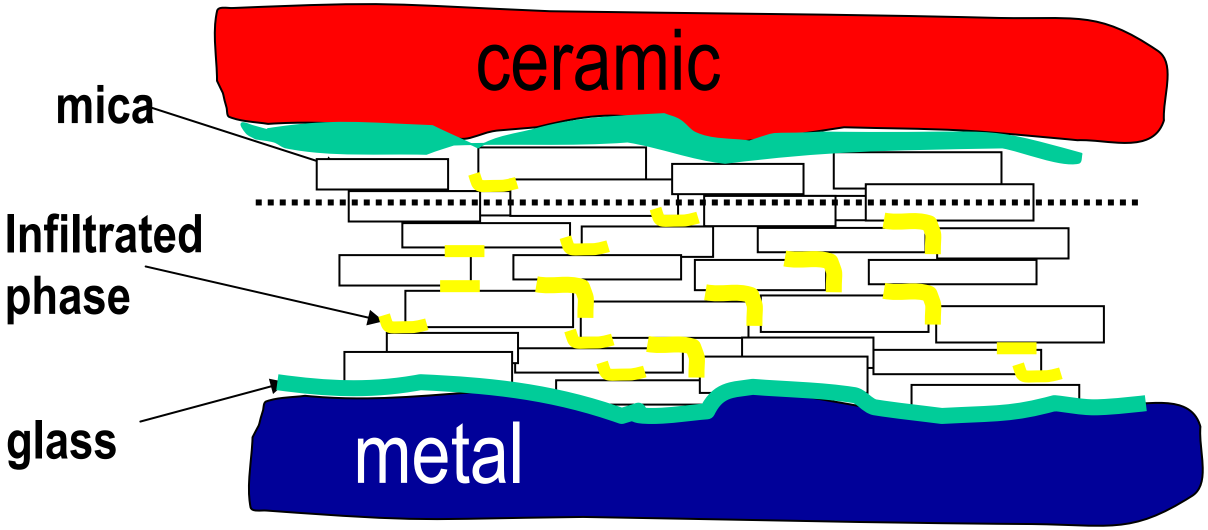


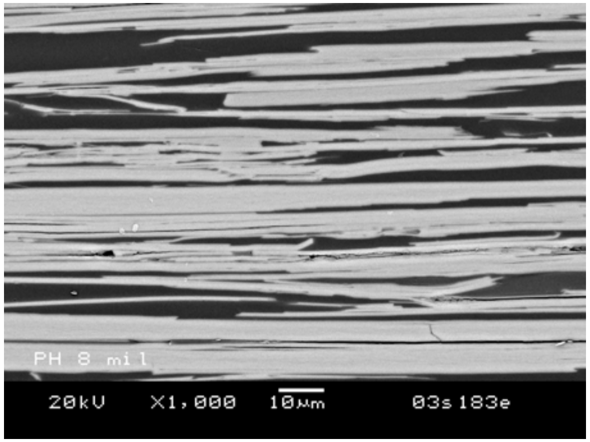
<u>Leak rates decrease with increasing thermal cycles for thinner micas</u>

Battelle

Infiltration Phlogopite mica paper with wetting or compliant phases

Two candidates: Bi_2O_3 (M.P.~813^{o}C), B_2O_3 (M.P.~450^{o}C)
Assumption: no severe reaction with the mica flakes

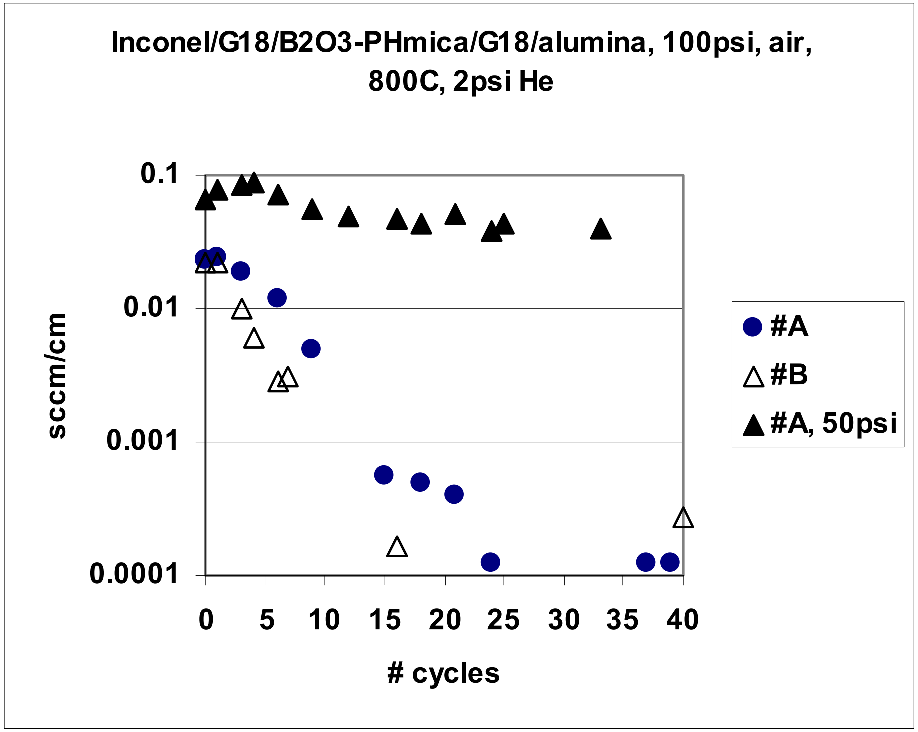




To minimize leak path from 3-D to 2-D by necking
or partial blocking paths between the discrete flakes

Leak test of H_3BO_3-infiltrated Phlogopite mica

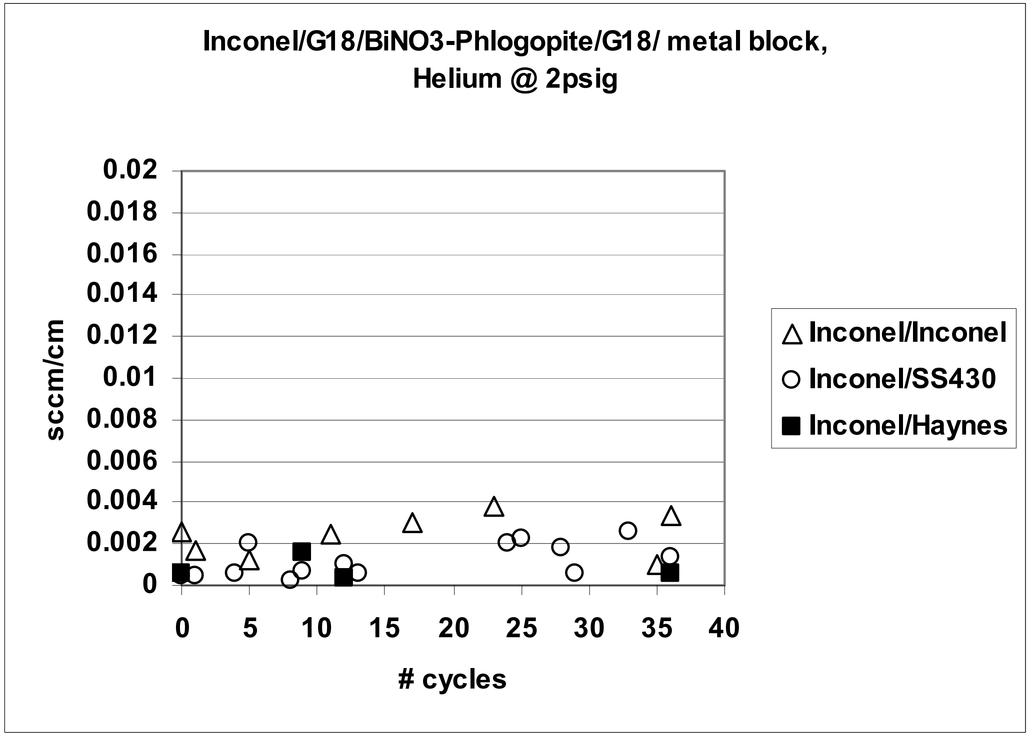
Two sources of Phlogopite mica paper used: ~3mil (#B) and ~4mil(#A) thick
Infiltrated with saturated solution at 70~90°C.
Minimum stress may be needed



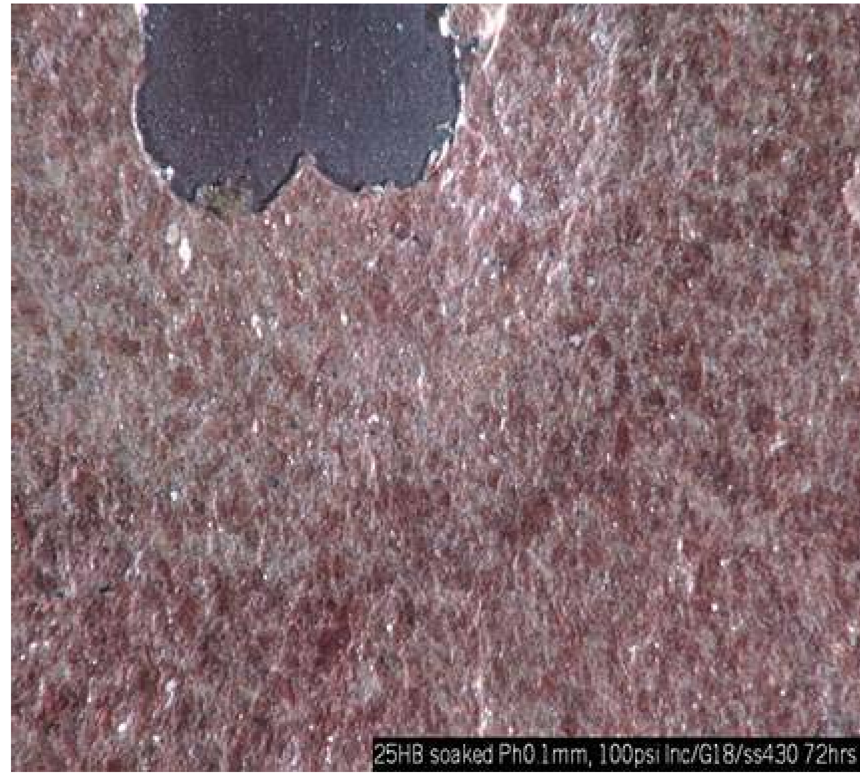
Battelle

Leak test of Bi-nitrate-infiltrated Phlogopite mica in 3 metal couples

Excellent thermal cycle stability with very low leak rates
3 metal pairs were tested: Inconel-Inconel, Inconel-SS430, Inconel-Haynes
Inconel (CTE 16-17 ppm/°C), Haynes (14~15 ppm/°C), SS430 (~12.7 ppm/°C)



Phlogopite mica appears stable in contact with molten Bi_2O_3 and B_2O_3

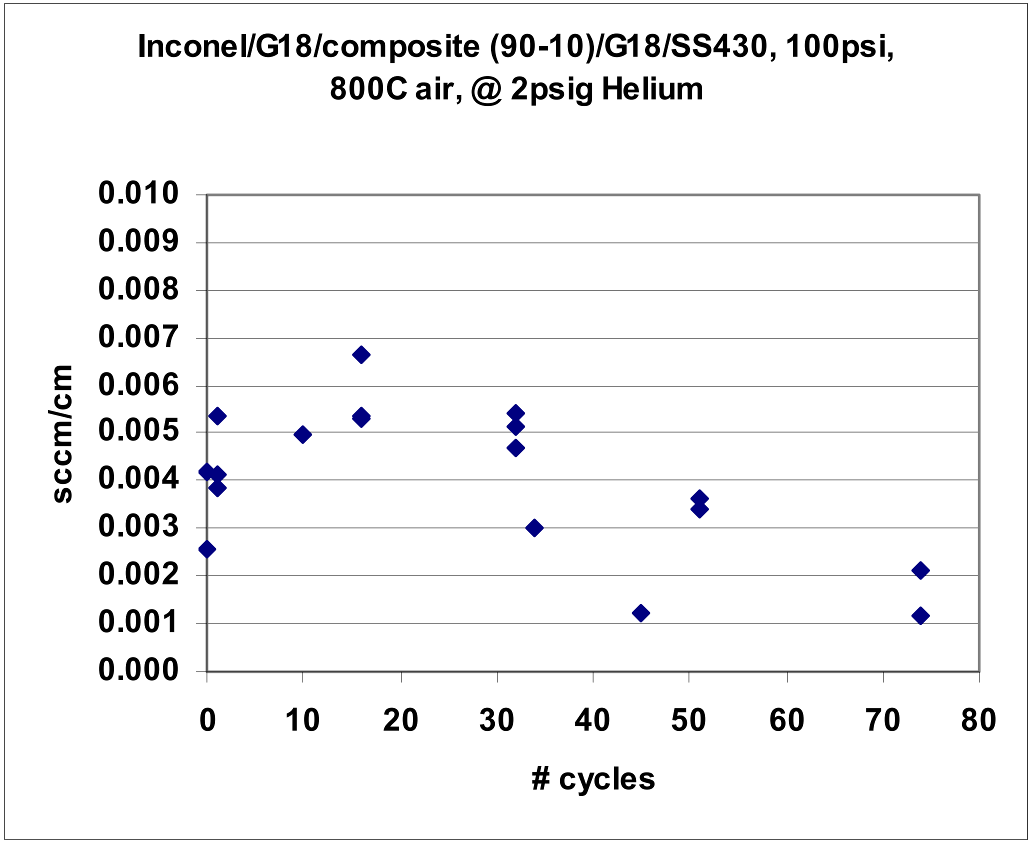


Bi_2O_3 after 37 cycles
Inc/G18/mica/G18/SS430

B_2O_3 after 72hrs @800^oC

Leak test of glass-mica composite in hybrid form

glass (10 v%)-mica (90 v%) in hybrid form @100psi

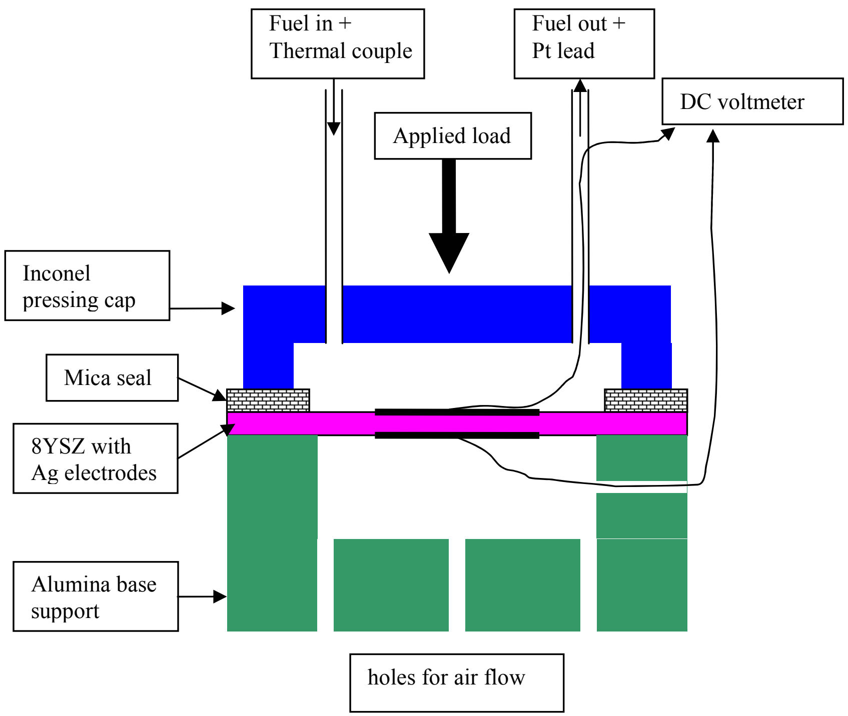


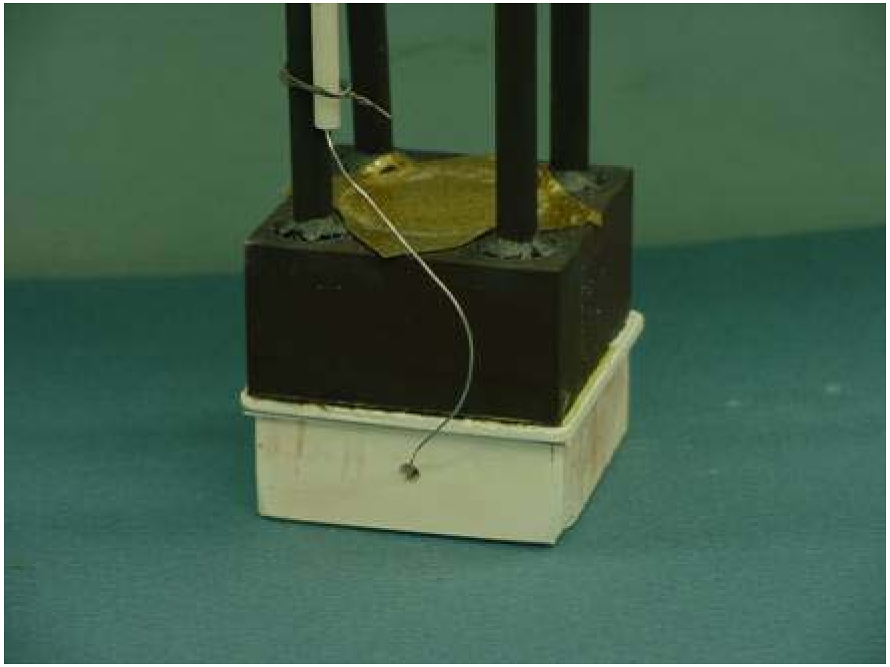
Battelle

U.S. Department of Energy
Pacific Northwest National Laboratory

Open circuit voltage testing

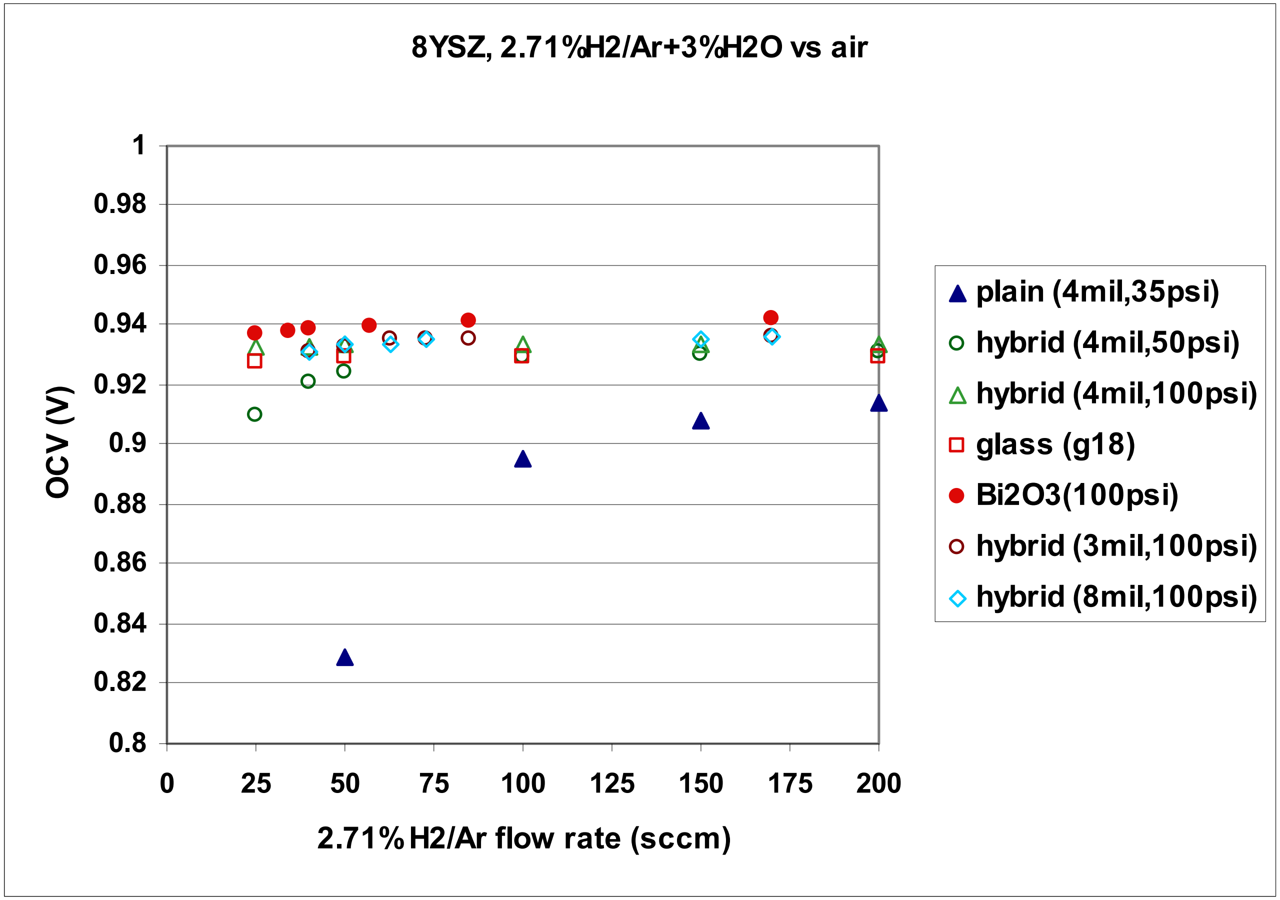
2”x2” 8YSZ dense plate with hybrid mica seals at 100 psi
Ag electrodes, Pt lead wire, and Inconel600 or SS430 block on alumina block





OCV test of 2”x2”dense 8YSZ plates with various hybrid mica seals

2.71% H_2/Ar + ~ 3% H_2O vs air, Theoretical (Nernst) OCV is 0.934 V at 800°C

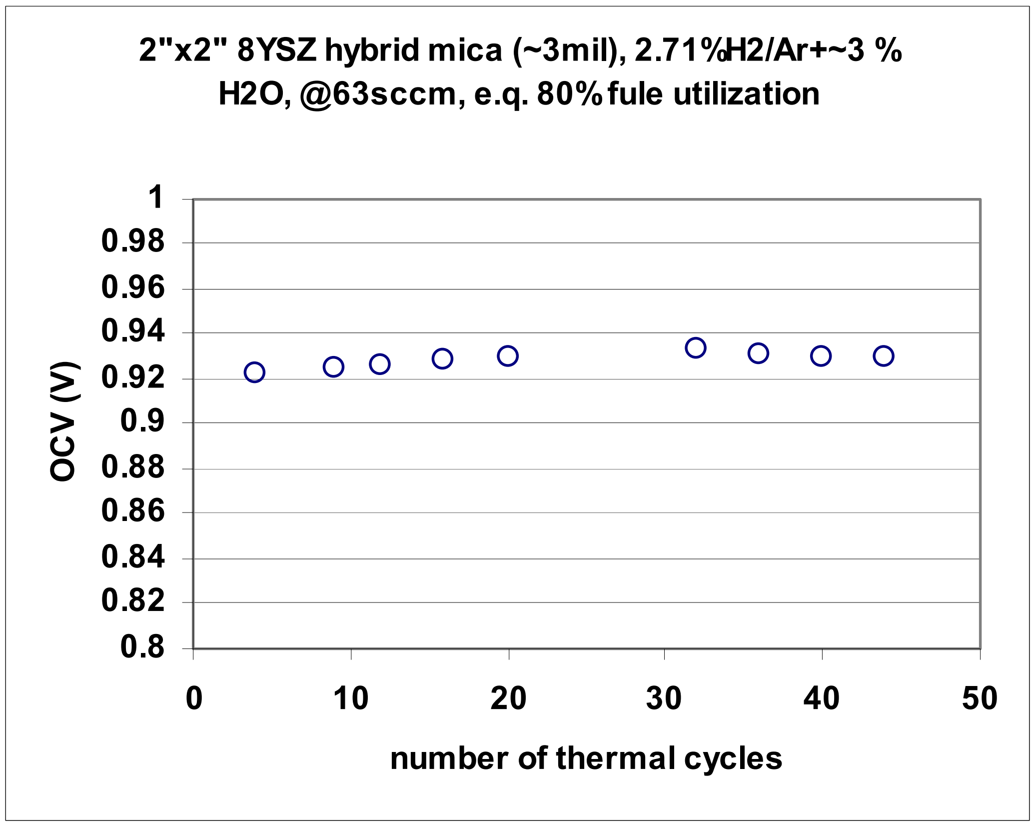


Battelle

U.S. Department of Energy
Pacific Northwest National Laboratory

OCV and thermal cycling of 2”x2” 8YSZ with hybrid Phlogopite mica (~3mils)

Equal to theoretical OCV, however, lost of contact after 45 cycles

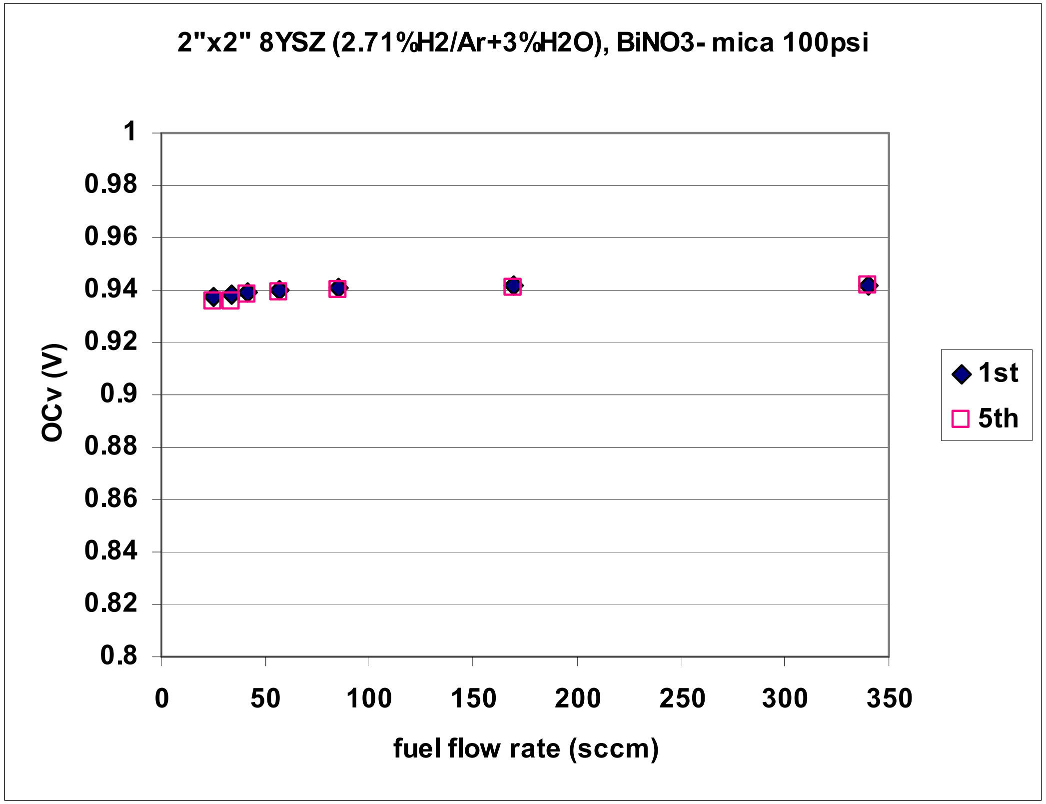


Battelle

U.S. Department of Energy
Pacific Northwest National Laboratory

OCV and thermal cycling of 2”x2” 8YSZ with Bi-nitrate-infiltrated mica in hybrid form

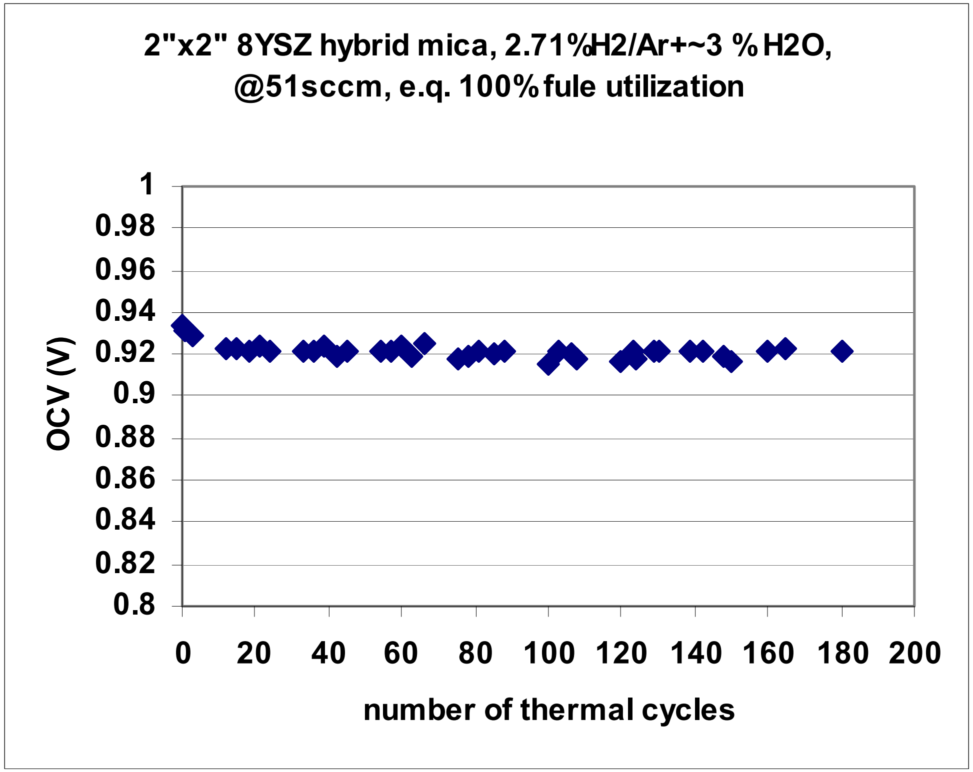
Equal to theoretical OCV, however, lost of contact after 5 cycles



Long-term thermal cycling and OCV of 2”x2” 8YSZ with hybrid Phlogopite mica (~4mils)

OCV=0.922 V after 180 cycles (~1.3% lower than Nernst value, 0.934V)

180 thermal cycles would be satisfactory for stationary applications



Heating @ ~20^{o}C/min

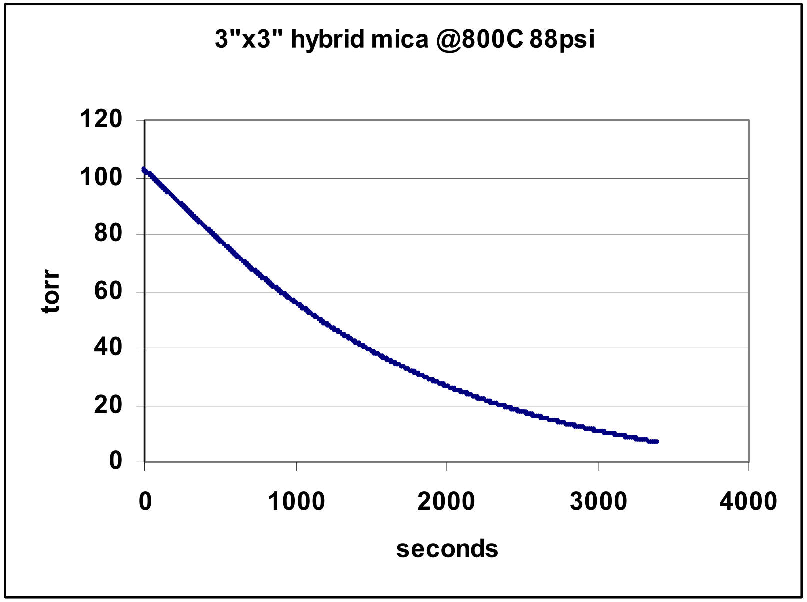
Cooling @ ~3^{o}C/min

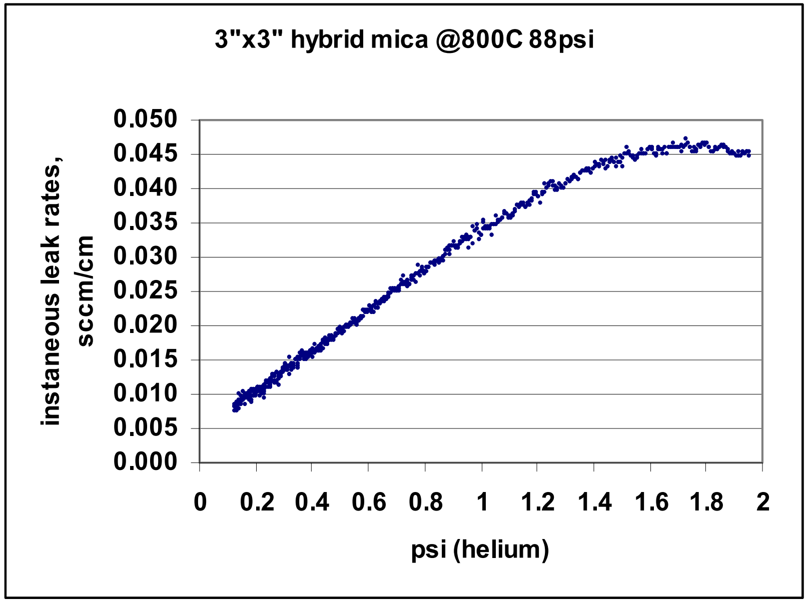
Effect of pressure on the leak rates

3"x3" hybrid Phlogopite mica (4 mils) pressed between Inconel and alumina substrate @ 800°C and 88 psi

→ Leak rates decreases linearly with decreasing pressure

→ No size effect of leak rates 0.045 sccm/cm (3"x3"), 0.043 sccm/cm (1.3"ϕ)





Minute fuel loss for compressive mica seals

For a 2”x2” cell of 0.5 W/cm^2
(@ 1.25”x1.25” active area, 0.7V, 0.75A/cm^2, and 800°C),

it needs pure hydrogen of

64 sccm @ 80% fuel utilization

Fuel loss due to mica leak (@ 0.2 psi)
= 0.01sccm/cmx20cm = 0.2 sccm ≈ 0.3%

Summary

- Thermal cycling of hybrid Phlogopite micas showed desirable behavior that the leak rates slightly decreased with thermal cycling. Thinner micas (4 and 8 mils) have lower leak rates than thicker ones (20 mils).
- Leak rates can be reduced 10-20 times to <0.004 sccm/cm when using "infiltrated" micas.
- Fuel loss due to mica leak will be very minute (~0.3%).
- OCV tests of various hybrid micas showed OCVs equal or close to the theoretical (Nernst) value.
- Excellent thermal cycle stability over 180 cycles were obtained on OCV test with hybrid mica seals.

Future work

- Continuing optimization hybrid mica and other advanced seals.
- OCV and cycling test of large-sized (3.5”x3.5”) 8YSZ of selected hybrid Phlogopite micas in pure hydrogen for ~30 cycles.
- Setup systematic and standardized leak tests for various sizes (1.3”ϕ, 2”x2”, 3.5”x3.5”, 6”x6”) of seals (mica and others). Leak and cycling test of actual-sized (6”x6”) single metal “dummy” plate with selected hybrid Phlogopite micas
- Leak and cycling (~30 cycles) test of actual-sized (6”x6”) multiple (10, 20 or 30) metal “dummy” plates with selected hybrid Phlogopite micas.
- OCV test with combined ageing (500 hrs @ 800^{o}C) and cycling (~30 cycles, 100-800^{o}C) of medium-sized (2”x2”) dense 8YSZ with selected hybrid Phlogopite micas.

Acknowledgement

The authors wish to thank Wayne Surdoval, Lane Wilson, and Don Collins (NETL) for their helpful discussions regarding this work.

This work was funded by the U.S. Department of Energy's Solid-State Energy Conversion Alliance (SECA) Core Technology Program.