SOFC seal development at PNNL

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Outline

- Compressive mica seal

1. Origin of major leak path
2. Hybrid micas
3. Long-term thermal cycling
4. Combined aging and thermal cycling
5. Long-term aging
6. Effect of temperature gradient
7. Issue of recyclable

- Conclusions

Current status of compressive seal

Final goals:
>40,000 hrs stability
$>10^2$ or 10^3 cycle
No degradation to mating mat'l
Low stresses
Low cost in SOFC stack

Hybrid micas showed low leakage over 18,650h @800ºC 12 psi

6"x6" tested

Hybrid micas survived combined 4000hr, 119 cycles @12 psi 0.03-0.04 sccm/cm @0.2psi

Hybrid micas showed low leakage at T gradient

Hybrid micas showed low leakage @ 6 psi and Nernst OCV

Hybrid micas survived 88 cycles@12.5 psi

Hybrid micas survived 1026 thermal cycle and 2052 hrs @800C, ~2.7%H_2/Ar+3% H_2O and 100 psi

Glass-mica composites

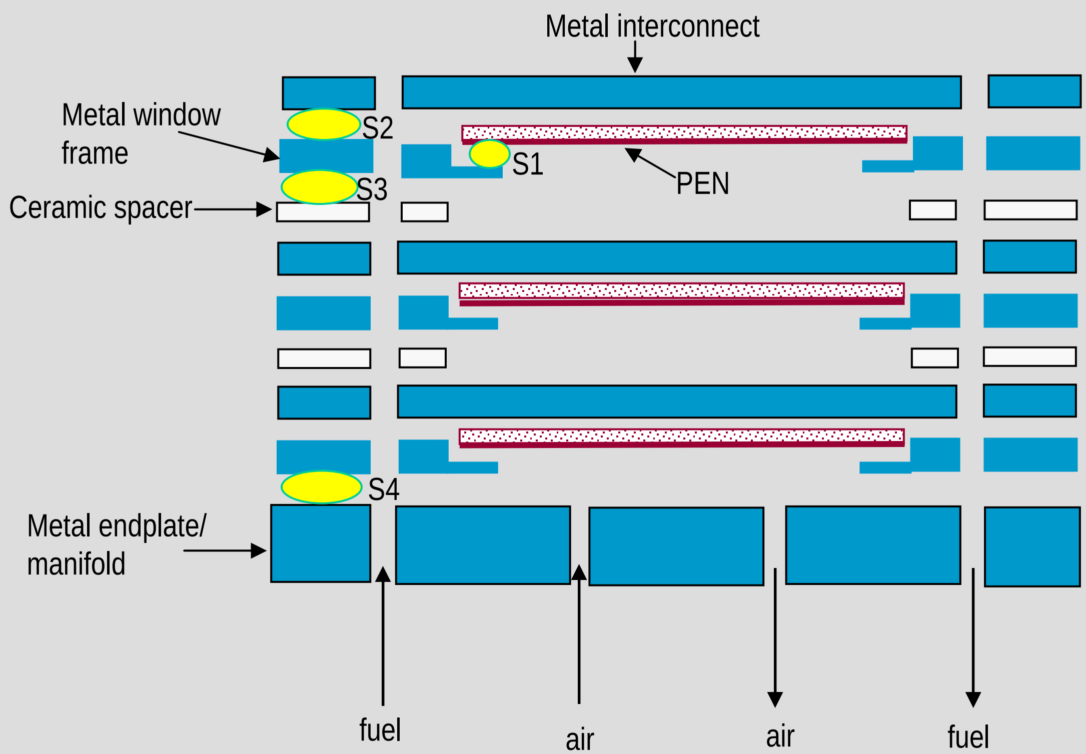
Infiltrated micas

Hybrid micas

Plain mica paper

Plain Muscovite mica (monolithic)

Multiple seal sections in planar SOFC stack



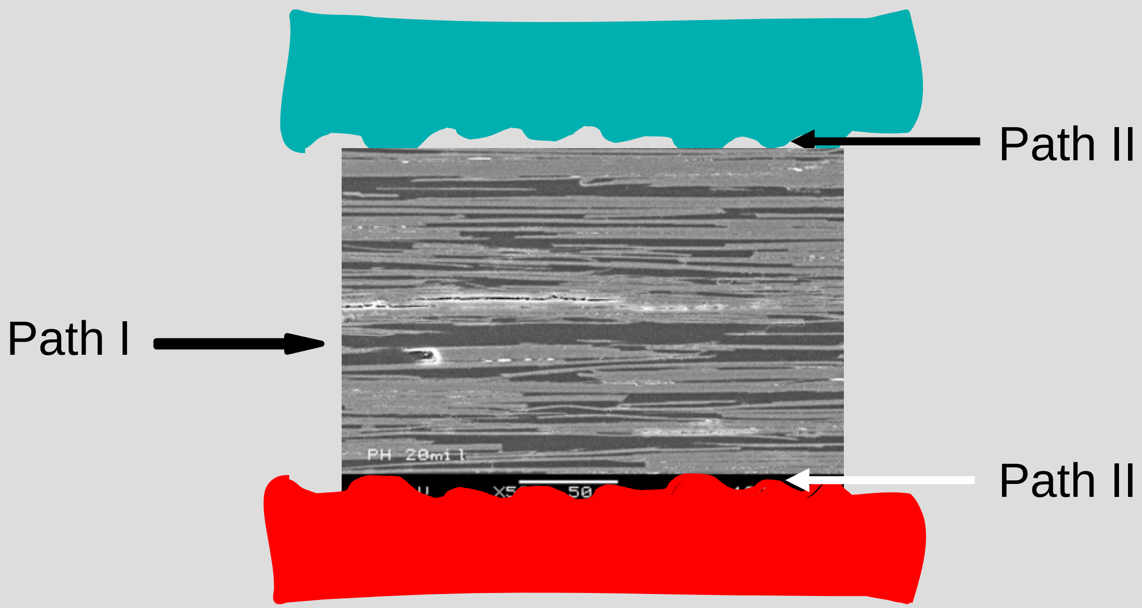
4 Sections to be sealed:

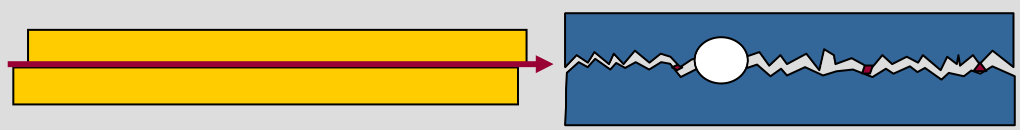
- Electrolyte to metal frame
- Metal frame to metal IC
- Metal IC to ceramic spacer
- Metal frame to metal endplate

Possible sealing approaches:

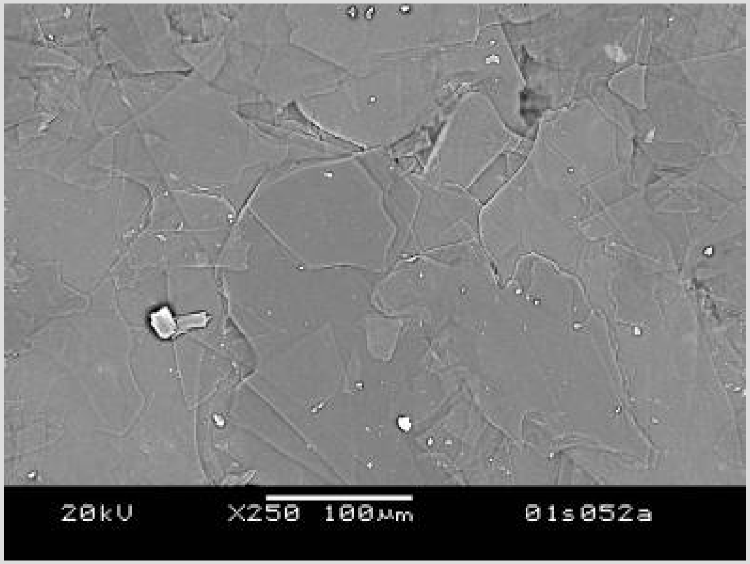
- Rigid glass (glass-ceramics)
- Braze
- Compressive (mica) seals
- welding

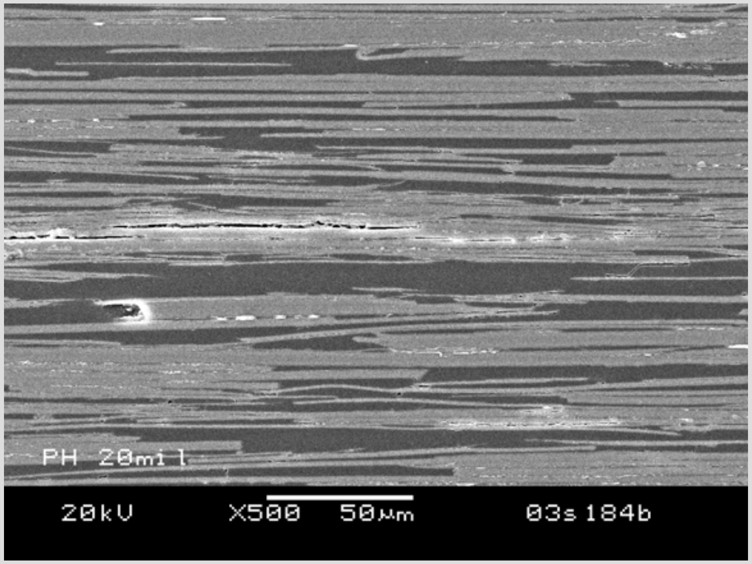
Origin of leak paths



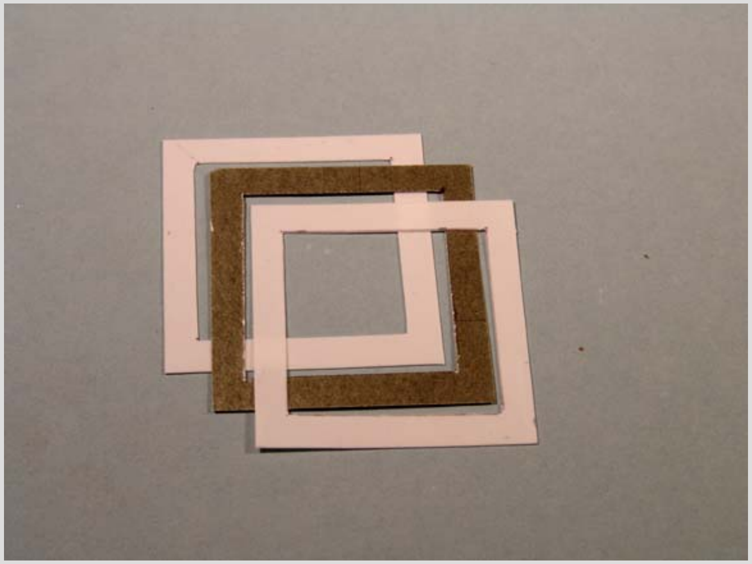


Hybrid micas



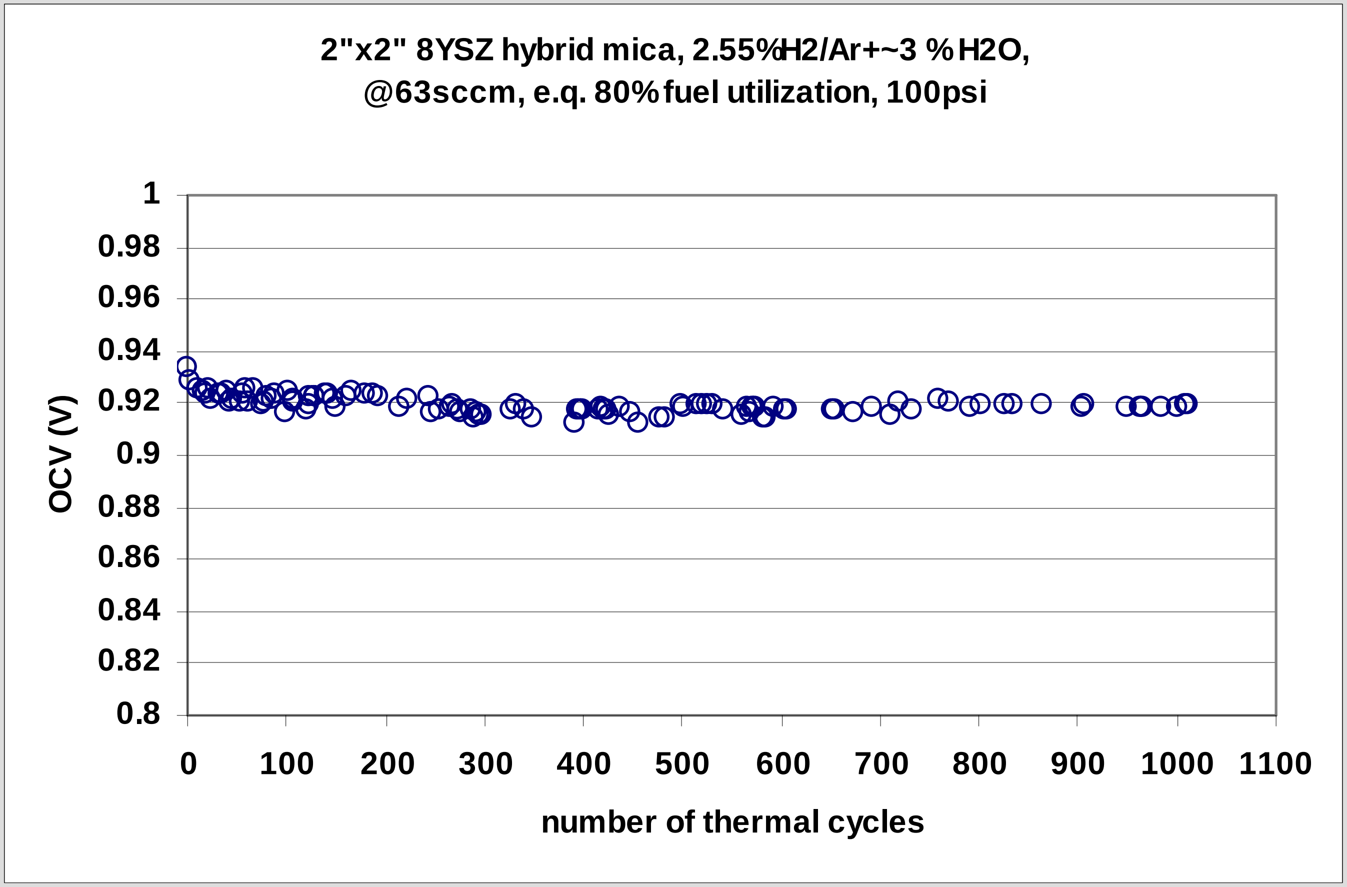


Compliant layers
1. Glass
2. Ag



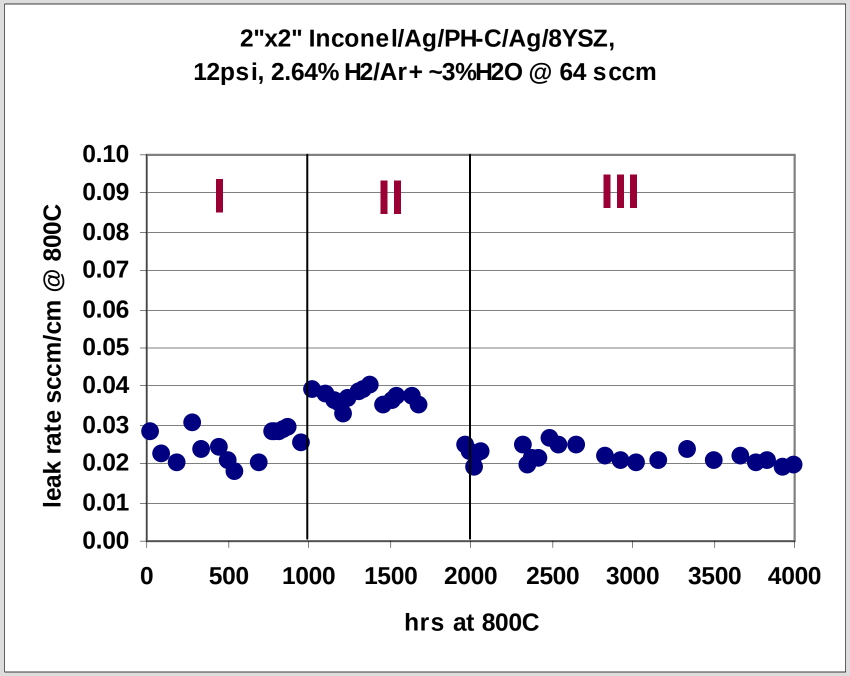
Long-term thermal cycling and OCV tests

Loss of ~1.4% (OCV=0.919 V) after 1026 thermal cycles (@ 100 psi)
At 63 sccm of 2.55% H_2/Ar + ~3% H_2O versus air

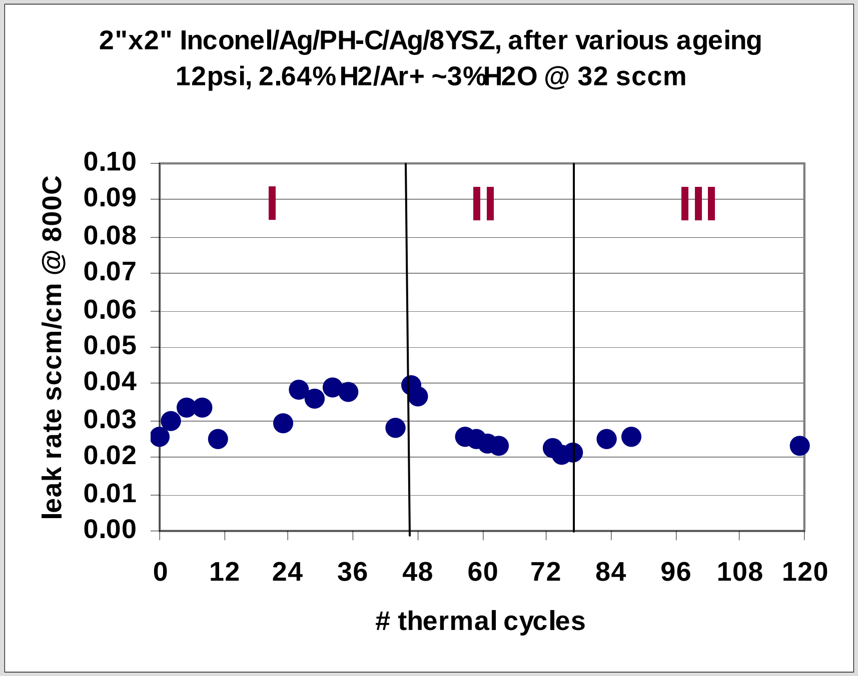


Combined aging and thermal cycling

Inconel/Ag/Phlogopite/Ag/8YSZ @12psi



Isothermal ageing

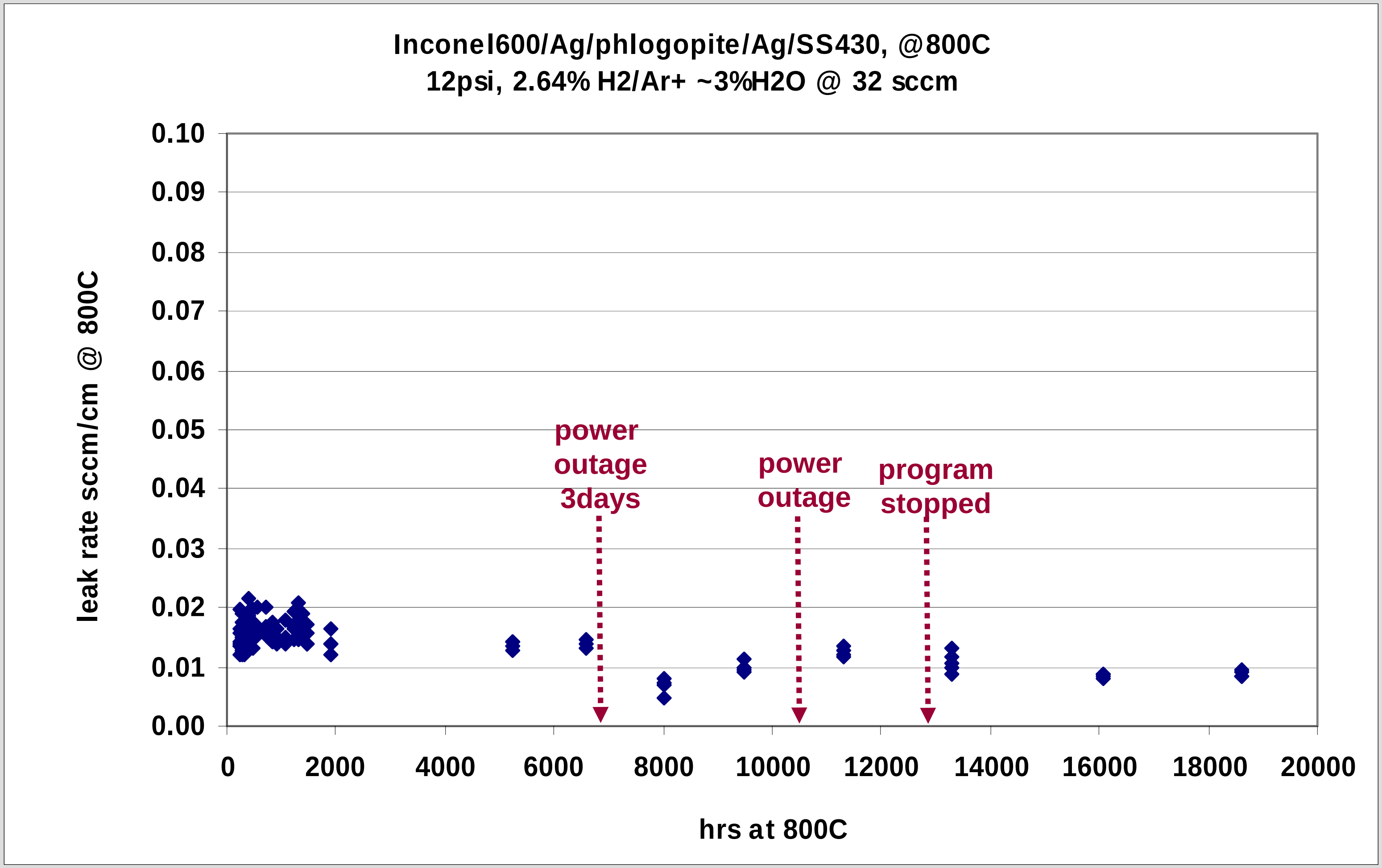


Thermal cycling after ageing

fuel loss = 0.2% @0.03 sccm/cm, 0.2 psid, 0.7V, 0.5 W/cm^2, 800^{o}C, 80% fuel utilization of pure hydrogen of a 6"x6" SOFC cell

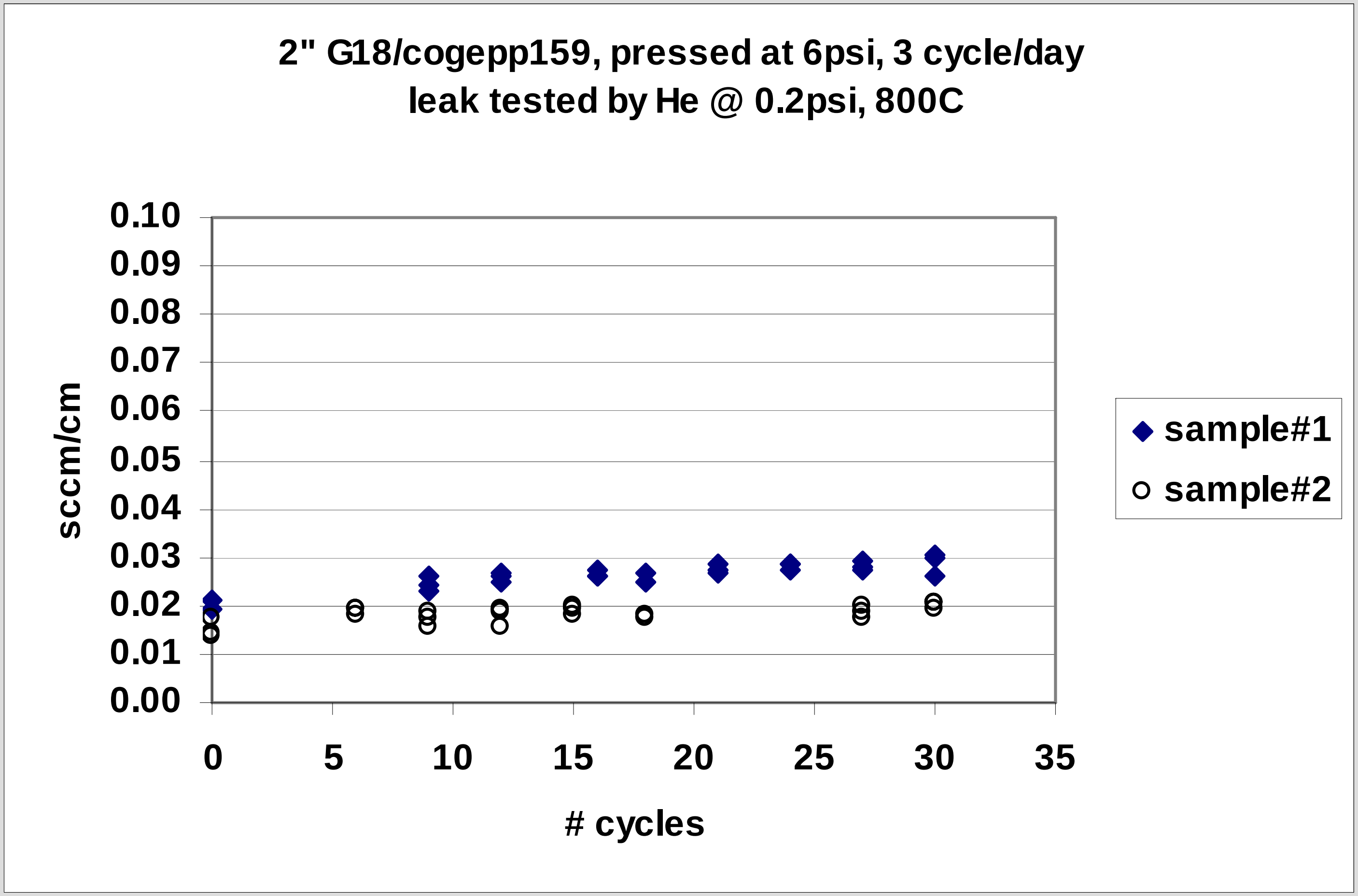
SECA target: fuel loss <1% @ 0.1 psid after 10 thermal cycles for 6"x6"

Long-term ageing at 800ºC

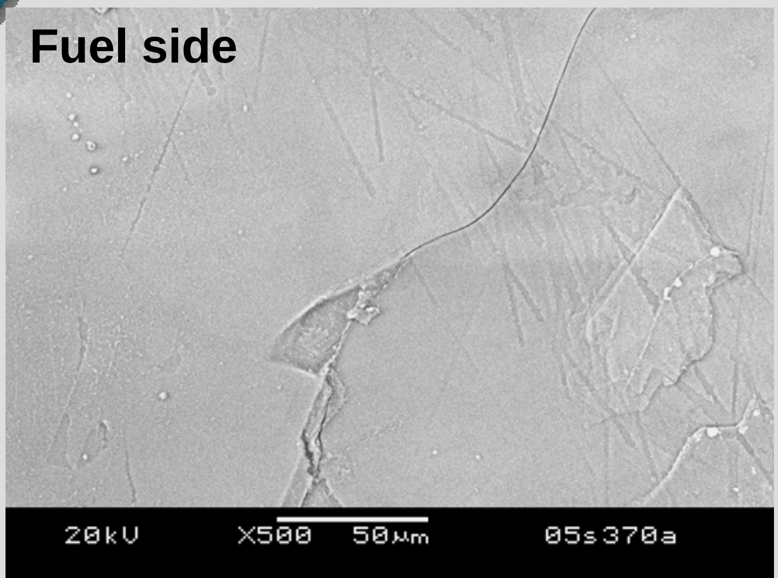


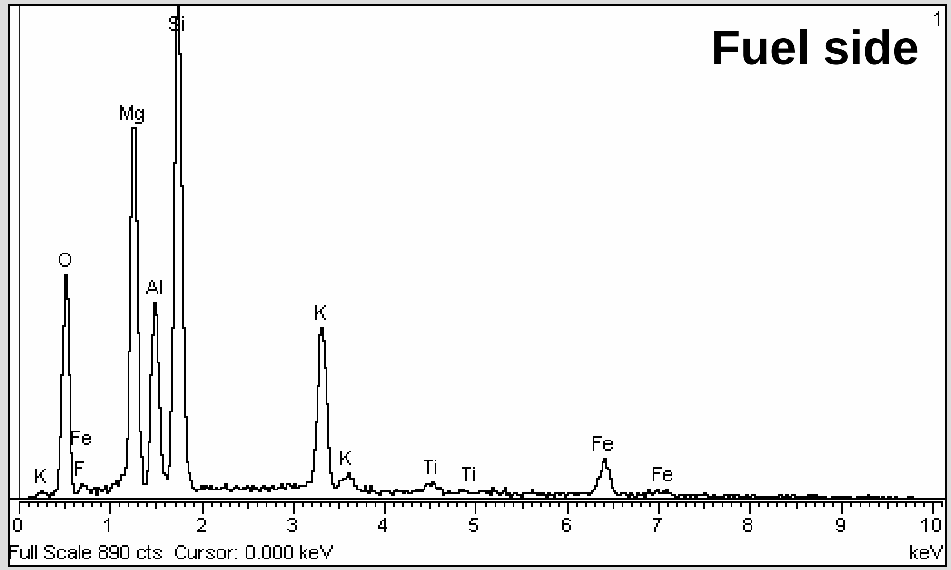
Successfully tested on 6”x6” stacks

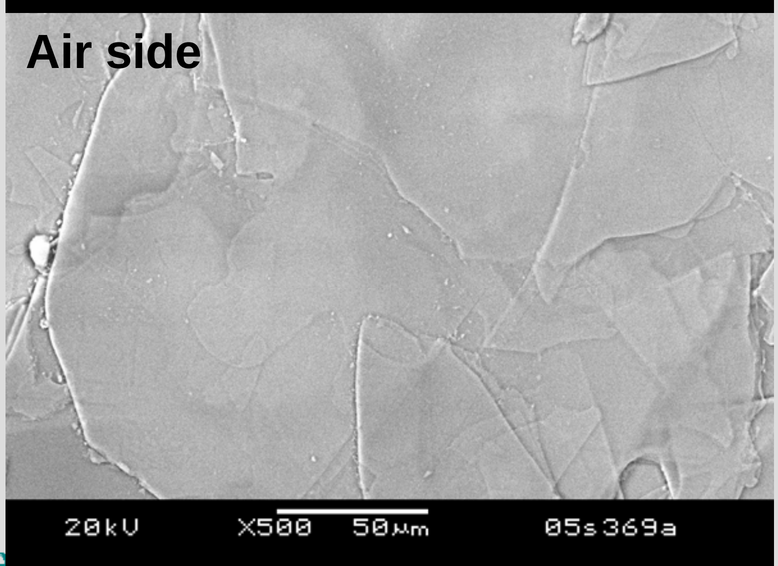
Good thermal cycle stability at very low stresses @ 6 psi

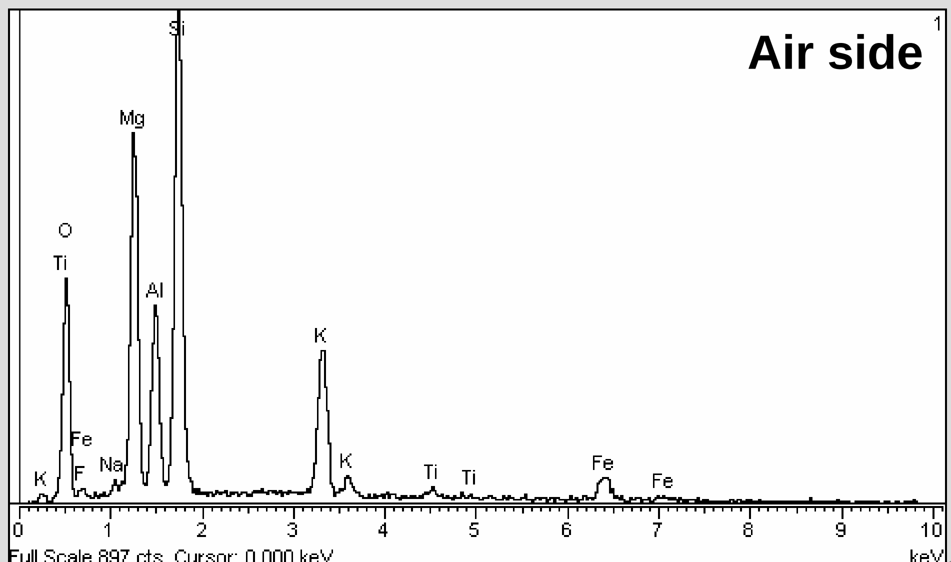


No mica degradation after 800ºC/4000 hrs and 119 thermal cycles





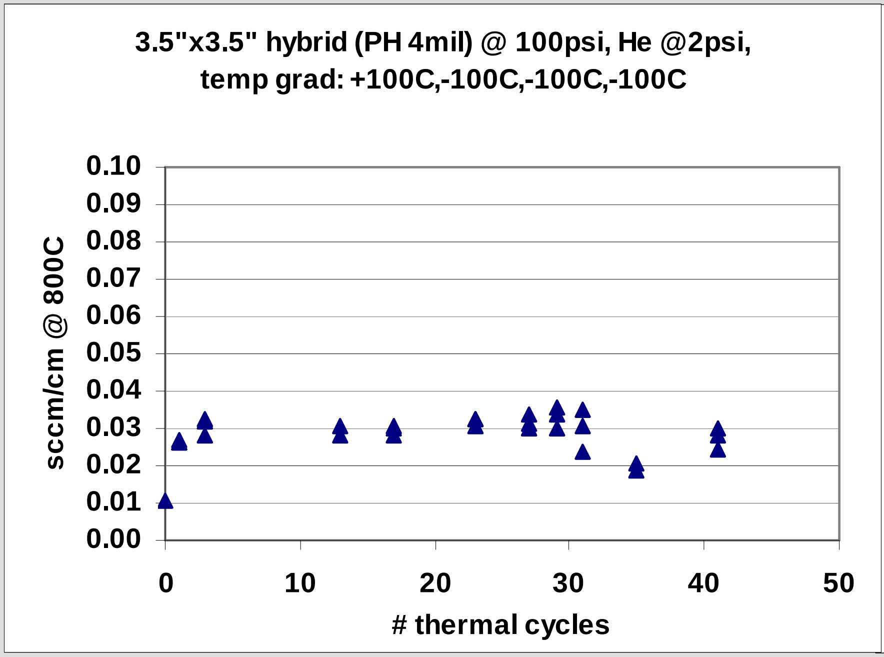




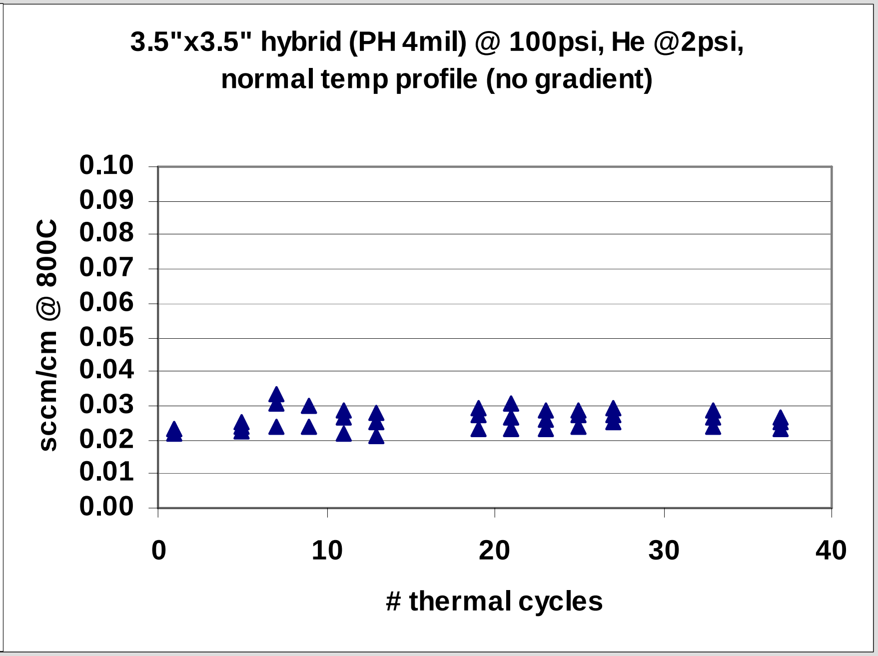
Effect of temperature gradient

3.5”x3.5” hybrid mica @100psi and cycling between 200-800^{o}C With T gradient of 30-100^{o}C

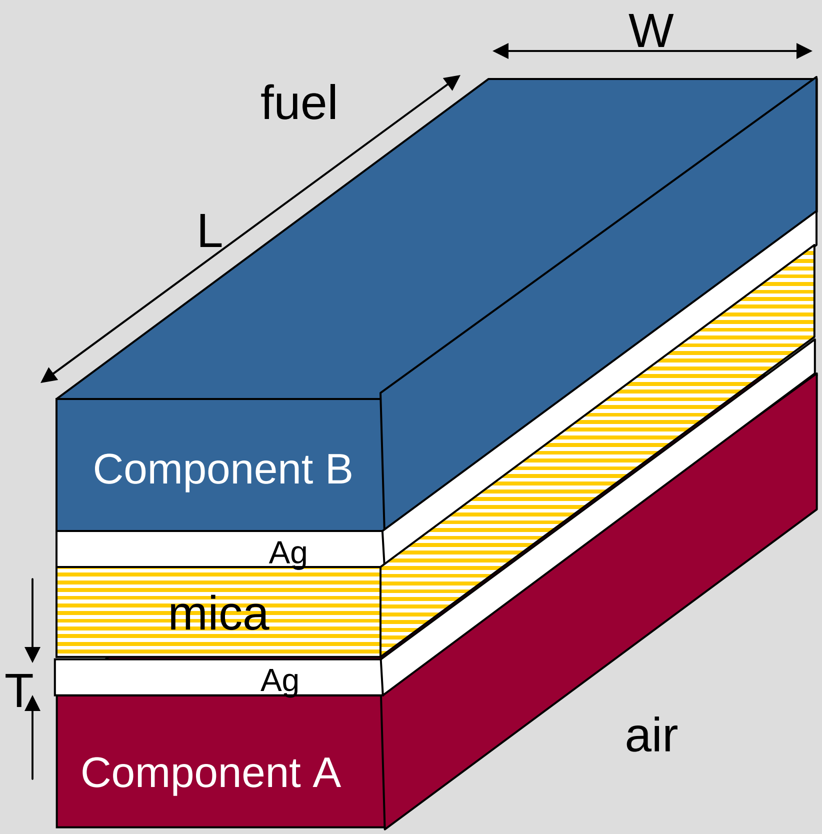
With T gradient



Without T gradient



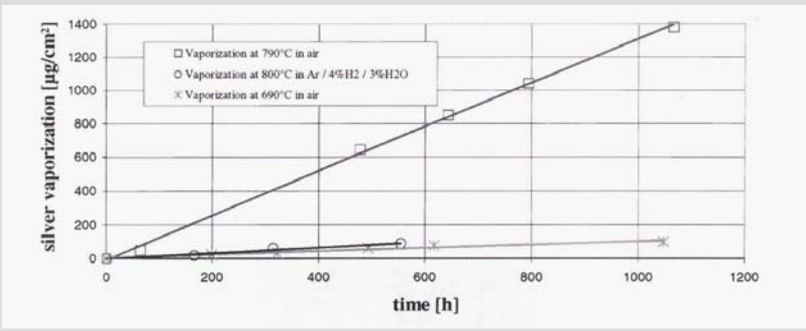
Insignificant loss of Ag in hybrid mica assembly @ 790-800^{o}C



For a width (W) = 0.5 cm
ρ(Ag) = 10.5 g/cc

Ag loss on fuel side
=40,000(aTL)/(ρTWL) = 0.12%

Ag loss on air side
=40,000(bTL)/(ρTWL) = 0.98%



From Meulenberg etal
J. mater. Sci., 36 [6] 3189-3195 (2001)

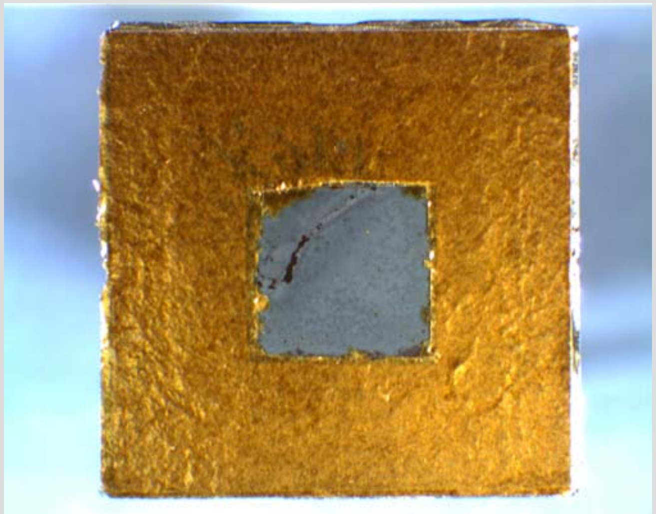
690^{o}C/air: 0.094 μg/cm^2/h

790^{o}C/air: 1.29 μg/cm^2/h

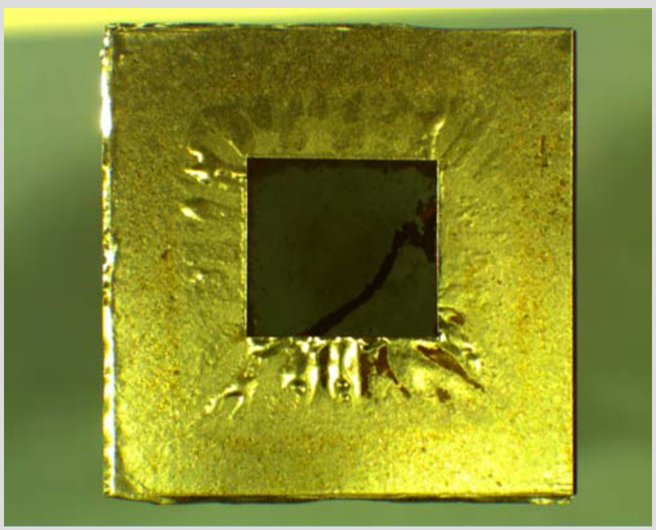
800^{o}C/Ar/H_2/H_2O: 0.161 μg/cm^2/h

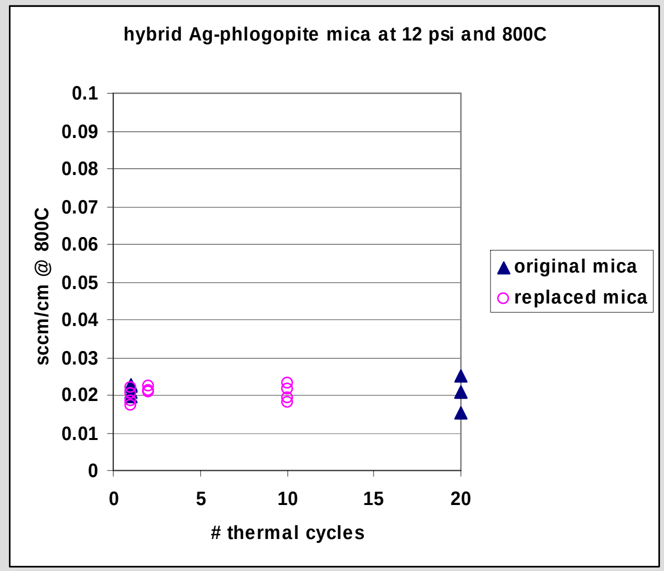
Hybrid mica makes SOFC recyclable

2"x2"
after 20
cycles



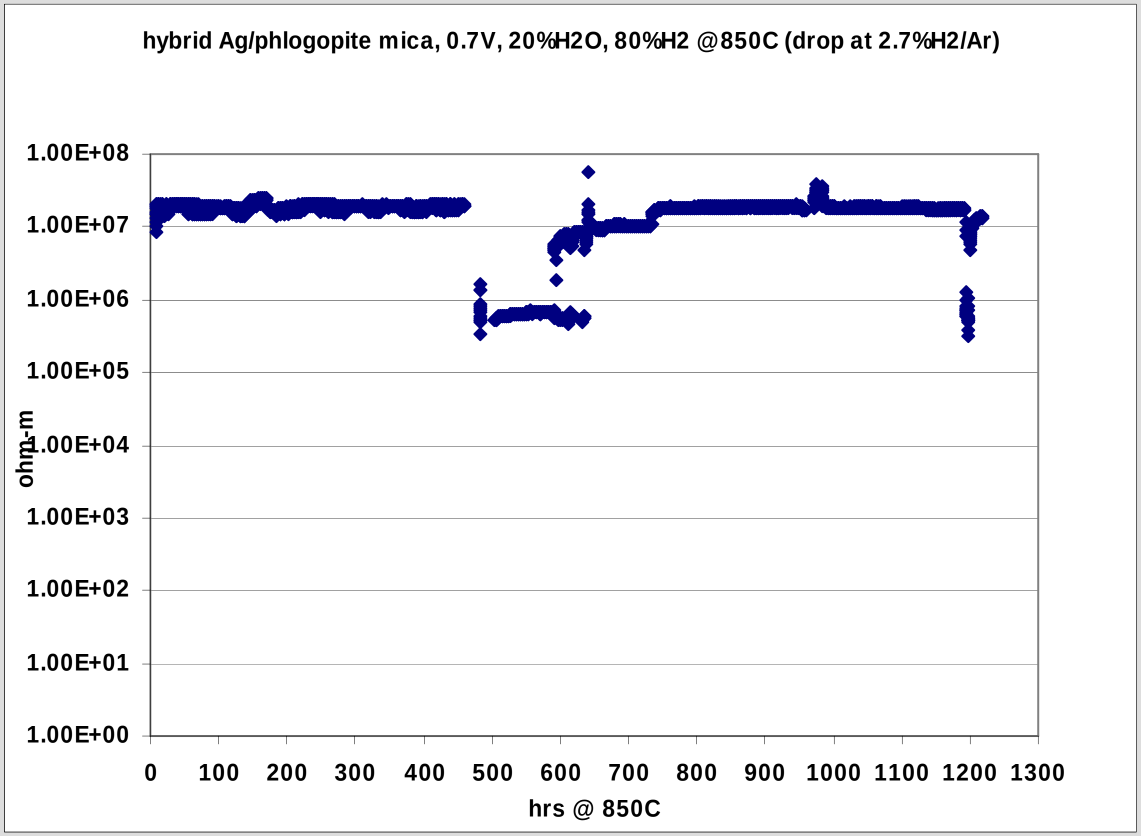
After air
sprayed





Electrical stability

Hybrid phlogopite with Ag under 12 psi @ 850°C and 0.7 V
Resistivity change when switching pure H2 to 2.7%H2/Ar



Conclusion

- Concept of hybrid mica was developed and tested using glass and/or Ag as compliant inter-layers.
- Hybrid mica successfully demonstrated desirable long-term (>18,000h) thermal stability, long-term thermal cycle (1026 cycles) stability, combined thermal cycling and aging test with constant leakage at low compressive stresses in SOFC environments.
- Phlogopite mica showed minute materials degradation during long-term aging. Vaporization loss of Ag was also insignificant.
- Hybrid mica is low cost, easy processing and offers potential for recycle and reuse of SOFC components.