Development of High Performance Seals for Solid Oxide Fuel Cells

Ronald E. Loehman

Advanced Materials Laboratory
Sandia National Laboratories
Albuquerque, NM

SECA Core Technology Program Review, Albany, NY Sept. 30 - Oct. 1 , 2003

Supported by DOE Contract DE-AC04-94AL85000 at Sandia National Laboratories

SOFC Seals Present Formidable Technical Challenges

- long-term HT stability in oxidation and reduction (decomposition, vaporization, phase transitions)
- low reactivity with environment and other components
- strength and toughness at the use temperature
- thermal shock resistance
- ability to accommodate CTE mismatch
- hermeticity

All for lifetimes of up to 10,000 hours

There is No Universally-Applicable Seal Material or Process

- Seal design is an engineering optimization that requires frequent tradeoffs
- Sealing techniques are specific to the application
- Rational design must be based on a thorough understanding of fundamental science, e.g.,

  - materials synthesis

  - reaction thermodynamics and kinetics

  - interface structure and properties

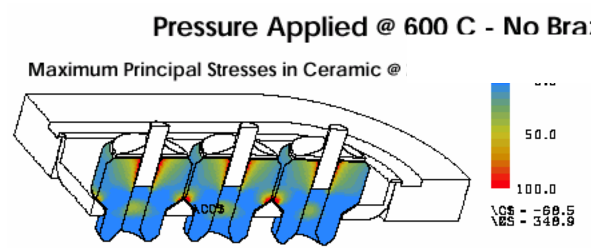
  - micromachanics of interfaces

Multidisciplinary Approaches to Seal Design and Manufacture Have Proved Successful

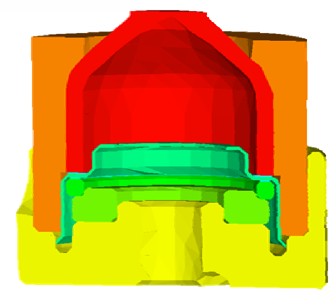
- Experimental Materials & Process Science
- Computational Modeling
  (Thermal, Fluid Flow, Mechanical, & Interface)
- Interfacial, Microstructural & Compositional Analyses
- Design & Production Feedback
  (Development, Characterization, Qualification, & Failure Analysis)

MC4277 Tube

The Multidisciplinary Approach Has Produced Ceramic Seals for a Wide Variety of Components



- switch tubes
- neutron tubes
- stronglinks
- thermal batteries
- storage containers
- plasma facing components
- feedthrough connectors/headers
- auxiliary power units
- liquid metal thermoelectric cells
- electrochemical potential sensors



Sealing Requirements for Solid Oxide Fuel Cells

- Leak tight at 800-1000°C operating temperature
- Withstand thermal cycling to room temperature
- Accommodate thermal expansion mismatch
- Stability in oxidizing and reducing environments
- Adhesion to cell components
- Sealant must wet but not react with stack materials
- Gas tight seal

SOFC Sealing Techniques Must Address Stresses From Thermal Gradients and CTE Mismatch

Cell designs with seals to materials with similar CTEs

- thermal strains only from temperature gradients
- can minimize with mechanical design process controls

Cell designs with seals to materials with dissimilar CTEs

- strains from both materials and thermal sources
- mitigated with materials and process development and component design

Composite Sealing Concept

Assume: SOFC components in intimate contact

CTE mismatch

Stress on thermal cycling

Approach: A deformable seal based on glass flow above its T_g

Control viscosity with powder additive

Slight flow to relieve stress, heal cracks

Rigid enough to remain in joint

Composite CTE and Viscosity Vary With Relative Amounts of Glass and Ceramic

Composite CTE depends on individual CTE and modulus values

$$\alpha = \frac{\alpha_1 K_1 V_1 + \alpha_2 K_2 V_2}{K_1 V_1 + K_2 V_2}$$

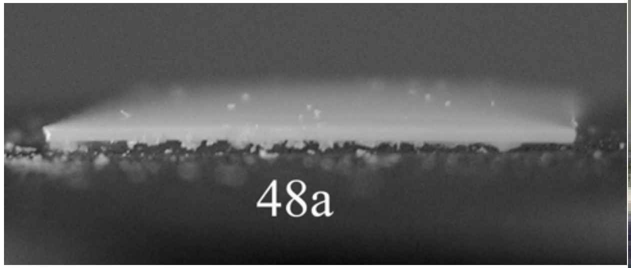
K_i = modulus, V_i = volume fraction

Viscosity increases with decreasing filler particle size and with increasing filler concentration

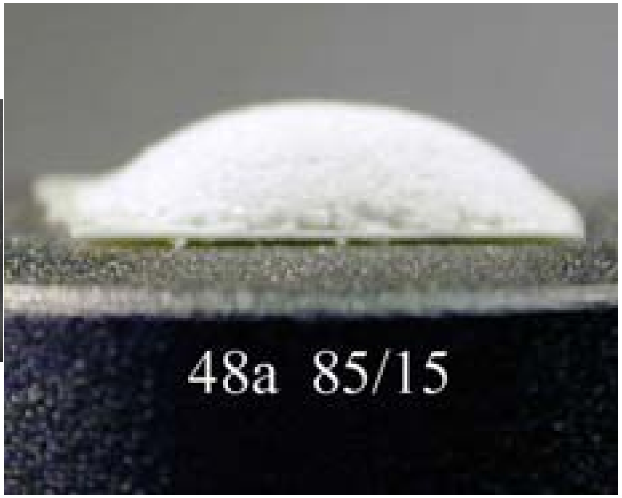
$$\eta = \left(1 + \frac{\kappa \phi}{1 - \left(\phi / \phi_{\max} \right)} \right)^2$$

κ = 1/particle size, ϕ = particle packing density

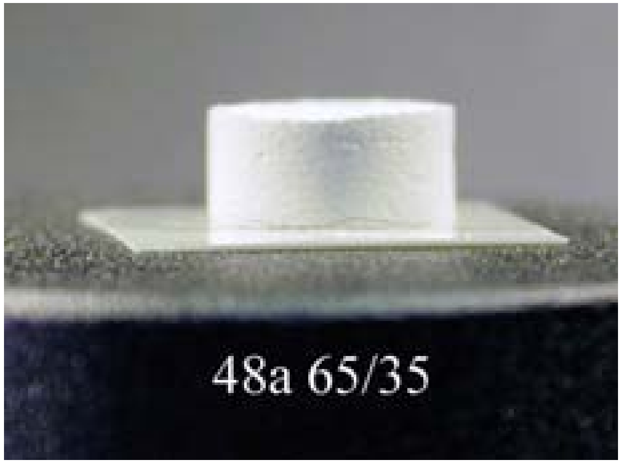
Heating Experiments Show Effect of Glass/Ceramic Powder Ratio on Composite Viscosity



Glass on YSZ



85 vol% Glass/15%YSZ Powder

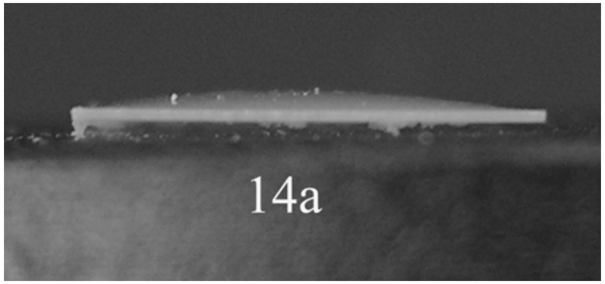


65 vol% Glass/35% YSZ Powder

Thermal Cycle: 10°/min to 1100°C, hold 12 min., cool to 850°C, hold 60 min.

Glass Properties: CTE = 6.1×10^{-6}/°C, Tg = 623°C

Heating Experiments Show Interplay of Glass Tg and Ceramic Powder Content on Composite Viscosity



Glass on YSZ

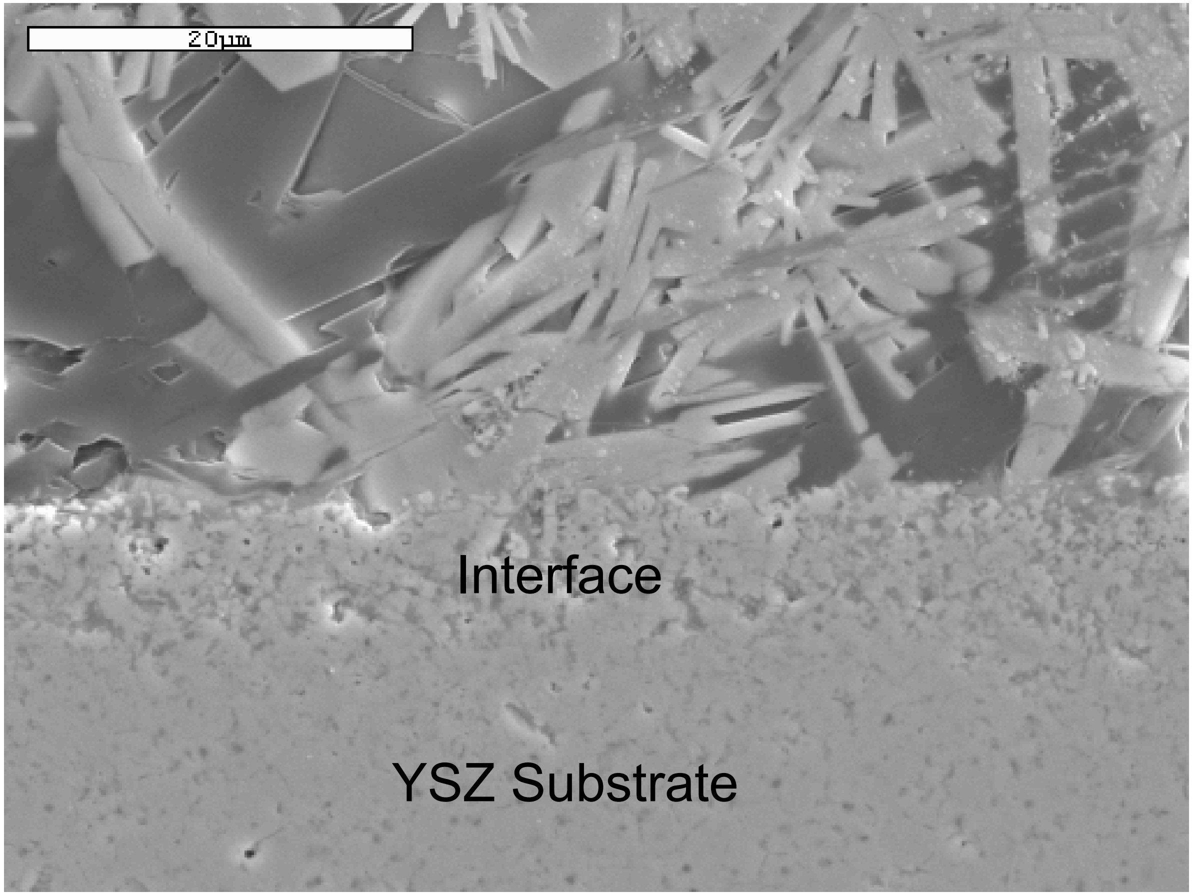
85 vol% Glass/15%YSZ Powder

65 vol% Glass/35% YSZ Powder

Thermal Cycle: 10°/min to 1100°C, hold 12 min., cool to 850°C, hold 60 min.

Glass Properties: CTE = 9.1×10^{-6}/ºC, Tg = 576°C

SEM Analysis Shows Good Adhesion at Composite-YSZ Interface



14a 85/15 @ 2000x

Work Plan for New Core Technology Project

- glass synthesis (Dick Brow, UMR)
- determine glass properties
- make composite sealants with range of properties
- make test seals, measure strengths
- determine stability of joints, interfacial analysis
- measure joint bi-layer deformation in situ (Garino, SNL)
- calculate stresses for candidate seals with 3D FEM computations
- Apply measurements and calculations to design workable SOFC seals