Solid Oxide Fuel Cell Seal Development at NASA Glenn Research Center

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INTRODUCTION

Solid Oxide Fuel Cells (SOFCs) are being developed at NASA Glenn Research Center (GRC) to meet the power requirements for future aeronautic applications such as unmanned aerial vehicles (UAVs) and aircraft auxiliary power units (APUs). Benefits of fuel cell for aircraft APUs include potential for greater efficiency, lower emissions (e.g., NO_X, CO_2)[1] than turbine APUs, and the potential for higher power output at altitude. Aircraft applications demand high specific power densities (1.0-1.2kW/kg)[2] that are approximately five times greater than current fuel cells being developed for stationary power generation and transportation applications. The higher power density drives the cell design to be lighter weight, compact, and operate at higher operating temperatures (800-1000°C) than those proposed for other programs. Furthermore, reformed Jet A is the planned fuel. To achieve this objective, a demonstration of fuel cell stack durability over a 1000hr lifetime is planned. These requirements pose problems requiring advancements in anode/electrolyte/cathode material development, interconnects, cell design, and improved gas seals.

Planar SOFC fuel cells require high temperature hermetic seals to (1) prevent mixing of the fuel and oxidant within the stack, (2) prevent parasitic leakage of the fuel from the stack, (3) prevent contamination of the anode by air leaking into the stack, (4) electrically isolate the individual cells within the stack, and (5) mechanically bond the cell components. The sealing challenges are aggravated by the need to maintain hermetic boundaries between the different flow paths during transient (heating / cooling) operation with vibration loads.

The planar SOFC seal research program at GRC is divided into three primary elements, thermo-structural design and analysis, high-temperature seal and materials development, and experimental leakage evaluation.

THERMO-STRUCTURAL DESIGN AND ANALYSIS

Researchers at GRC are performing thermo-mechanical modeling of the fuel cell components to determine the requirements of candidate seal material systems.

Members of the Department of Energy's (DOE) Solid State Energy Conversion Alliance have developed a suite of software that can use chemical and flow analysis results as input for heat transfer and structural analysis solutions for SOFCs. NASA GRC team members have begun applying this software to define thermal and structural loads on the fuel cell seals to guide the required seal and material development.

While the analysis software serves as a tremendous asset to the effort, several challenges must still be addressed when modeling the SOFC operation. These challenges can be classified in terms of thermal analysis, material modeling (e.g., viscoelastic), and time dependent structural analysis.

The thermal fluctuations that the SOFC experiences throughout its operational lifetime can cause severe transient thermal stresses in both the seal and structural elements of the SOFC. This includes start up and shut down cycles, as these transients result in more severe thermal and mechanical stresses than steady state operation. Boeing[1] estimates that approximately a 40x decrease in SOFC start-up time is required for APU applications to compete with turbine APUs. Thermal profiles as a function of time will allow the team to capture the effects of thermal stresses that result from differing coefficients of thermal expansion (CTE) between the elements of the SOFC. By conducting case studies which consider different properties of the stack and seals, guidelines will be developed for material CTEs, material strengths, strains to failure, amongst others.

Glasses and glass composites are being considered for SOFC applications for several reasons, including the ability to tailor their expansion rates, their dielectric nature, and thin profile required to make a hermetic seal. However, glasses have radically varying stress-strain behavior as a function of temperature. At low temperatures, glasses tend to behave in a linear-elastic manner. As temperature increases, their behavior changes to viscoelastic behavior where strain in the material is a function of time and temperature. Current modeling efforts use viscoelastic material models to account for this behavior and achieve a more accurate representation of the seal response. Viscoelastic properties are not available for the glass / glass composites of interest for this project. As a result, GRC researchers will pursue obtaining these properties for select materials to satisfy modeling requirements.

By coupling the viscoelastic material properties of the seals and the time dependent thermal profile of the SOFC components, a time dependent structural solution is possible.

HIGH-TEMPERATURE MATERIALS DEVELOPMENT

Bonded seal materials that offer some measure of flexibility, such as glass, have shown the most promise of providing a hermetic seal for SOFC. However, previously formulated glasses possess less than adequate strength and fracture toughness properties to reliably seal throughout the lifetime of the SOFC. In order to improve these properties, researchers at GRC are investigating the effects of composite reinforcement.

A barium calcium aluminosilicate (BCAS) glass of composition 35BaO-15CaO-5Al2O3-10B2O3-35SiO2 (mol percent) has been developed by Pacific Northwest National Laboratory (PNNL) for use as sealing material for planar SOFCs. Since this composition has shown a tendency to crack upon thermal cycling, the glass was reinforced with one of two different additives: alumina platelets or yttria-stabilized zirconia (3-YSZ, mol percent) particulates.

Panels of glass containing 0 to 30 mol percent of the ceramic reinforcements were hot pressed and machined into test bars. Mechanical and physical properties including 4-point flexure strength, fracture toughness, elastic modulus, and density were measured at room temperature. Adding alumina or 3-YSZ significantly increased the composite flexure strength and fracture toughness (Figs. 1 and 2). For instance, adding either 30% alumina or 3-YSZ increased the flexure strength by 3.6 or 2.4 times, respectively, as compared to the parent glass (Fig. 1). Adding either 30% alumina or 3-YSZ increased fracture toughness by 4.5 or 2.2 times, respectively, as compared to the parent glass (Fig. 2). Elastic modulus (Fig. 3) also increased with increasing amount of reinforcement. The increase in elastic modulus was more predominant for alumina composites than for 3YSZ composites. Addition of alumina did not have much effect on the density whereas the glass density increased linearly with increasing 3YSZ content (Fig. 4). In summary, alumina platelets are much more effective in improving the strength, fracture toughness and elastic modulus of the sealing glass than 3YSZ, without affecting the density. These findings are important first steps in improving glass strength properties to arrive at a strong, durable, hermetic seal.

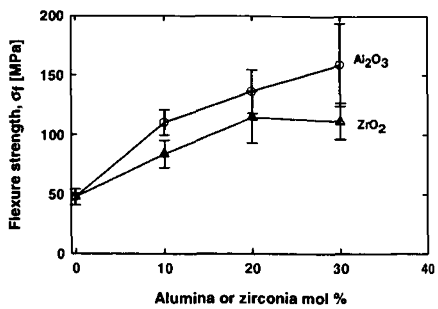

Fig. 1. Flexure strength of glass composites

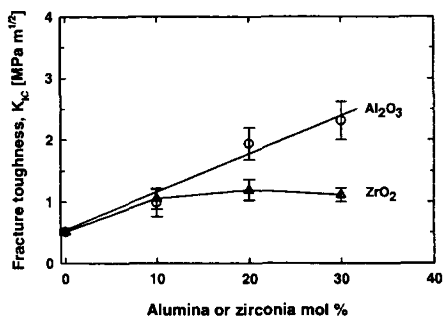

Fig. 2. Fracture toughness of glass composites

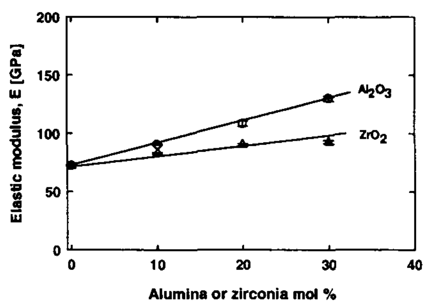

Fig. 3. Elastic modulus of glass composites

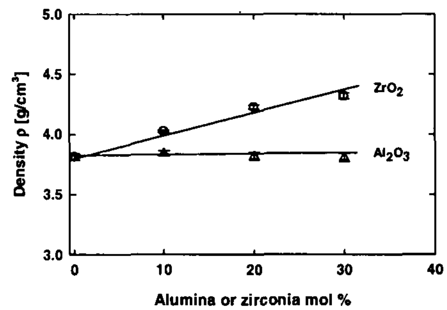

Fig. 4. Density of glass composites

EXPERIMENTAL LEAKAGE EVALUATION

The performance of candidate SOFC seals is characterized using a newly developed fuel cell seal test facility at GRC. The experimental setup (Fig. 5) includes a mechanical load frame, furnace, test fixtures, and gas supply. The system is capable of testing candidate seal concepts while subjecting the seals to representative mechanical loads, temperatures, temperature transients, and pressures of operating SOFCs.

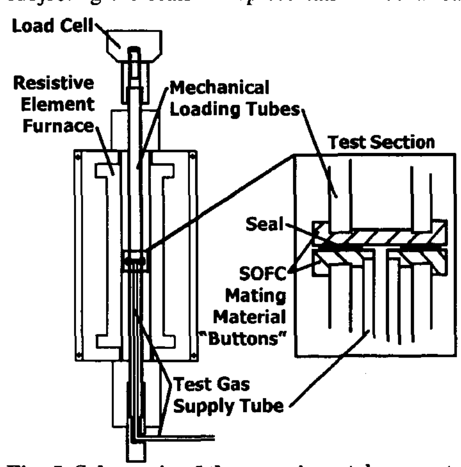

Fig. 5. Schematic of the experimental apparatus.

A unique specimen design is used to maximize test efficiency and permit testing of a large number of seal and adjoining materials. The seal is made between two 1-inch diameter "buttons" made of the candidate SOFC elements to be sealed. Depending on the sealing location being investigated, the buttons are made of metal, ceramic, or cermets. Seals range from glasses, glass composites, braze alloys, to compression seals.

In this unique test specimen design, the gas is supplied to the space between the seals using a supply tube hermetically attached to the lower button. The tube is connected to a gas reservoir using a system of high-vacuum (e.g., hermetic) tubing, fittings, and valves. Any pressure loss recorded in the gas reservoir can be attributed to leakage through the candidate seal, since the system is design to eliminate any background leakage. The decay of pressure in the reservoir is measured using a precision pressure transducer with an accuracy of ±0.05% (±0.00125psi). The leakage rate of the candidate seals is quantified using either Helium or air as the test gas. Due to the long duration of the leakage tests, both the reservoir temperature and barometric pressure are recorded to ensure accuracy of the leakage measurements.

An electromechanical load frame is used to apply a representative compression force to the candidate seal. The load is transmitted to the candidate seal assembly using quartz tubes extending from the load frame grips. A 1000 lbf capacity load cell is used to record the applied force to accuracies better than 0.03%.

The temperature of the test section is controlled using a quartz tube furnace manufactured from coaxially mounted glass tubes. A spirally oriented resistance element is mounted within the tubes to provide a circumferentially uniform heat distribution. The inner surface of the outermost tube is layered in gold, providing a mirror to reflect the infrared energy toward the test section. Due to the low thermal mass of the system, very rapid thermal ramp rates are possible with a maximum operating temperature of 1100°C. The thermal shock resistance of the furnace materials allows for very rapid cooling, thereby reducing the thermal cycling time and the overall test duration.

Post-test examination of the seal and the surrounding structure is used to characterize the microscopic features of the seal interfaces and to determine where possible leakage paths may have developed. Finally, post-test inspection data is combined with the structural analysis results to assess any failure mechanisms and to guide subsequent seal material and concept development.

NOVEL SEAL CONCEPTS

In addition to compressive, rigidly bonded, and compliant bonded seal materials, seals that offer some flexibility during operation are being considered. Novel seal concepts based on this approach are being developed at GRC that incorporate both advanced materials and mechanical structures in order to provide flexibility at the seal / component interface. Under investigation are concepts that do not rely solely upon matching the seal materials coefficient of thermal expansion, but in addition use the material properties (e.g., glass transition temperature and wettability) and structural design to create a hermetic seal.

SUMMARY

Researchers at NASA GRC are confronting the seal durability challenges of Solid Oxide Fuel Cells by pursuing an integrated and multidisciplinary development effort incorporating thermo-structural analyses, advanced materials, experimentation, and novel seal design concepts. The successful development of durable hermetic SOFC seals is essential to reliably producing the high power densities required for aerospace applications.

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[1] Daggett, D. "Fuel Cell APU for Commercial Airplane," 4th Annual Solid State Energy Conversion Alliance (SECA) Workshop, Seattle, Washington, April 15-16, 2003.

[2] Liang, A. "Emerging Fuel Cell Developments at NASA for Aircraft Applications," 4th Annual Solid State Energy Conversion Alliance (SECA) Workshop, Seattle, Washington, April 15-16, 2003.