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Mica-based Composite Compressive Seals for SOFC

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Introduction

Pacific Northwest National Laboratory is developing glass – mica "hybrid" composite compressive seals for high temperature applications such as solid oxide fuel cell stacks. The seals are easy to fabricate, and offer stable, low leak rates.

Technology

One of the critical challenges facing planar solid oxide fuel cell (SOFC) developers is the need for reliable sealing technology. Seals are required which will offer long-term stability in the high temperature SOFC environment and also maintain their integrity during thermal cycling. Several different approaches for sealing SOFC stacks are under development, including glass or glass-ceramic seals, metallic brazes, and compressive seals. Compressive seals potentially offer a significant and unique advantage over the other approaches by providing a means of mechanically "de-coupling" adjacent stack components, thereby minimizing the need for closely matching the coefficients of thermal expansion (CTE) of the various SOFC stack components. In an attempt to help the SOFC industry overcome sealing challenges, PNNL is developing mica-based hybrid compressive seals which exhibit leak rates 2 to 3 orders of magnitude lower than obtained with simple mica gasket seals.

Materials and Fabrication

The hybrid seals consist of commercially available Phlogopite mica paper sandwiched between layers of a proprietary SOFC glass-ceramic seal material (US Patents 6,430,966; 6,532,769). The seals are fabricated by inserting the mica paper between polymer tapes (prepared by conventional tape casting techniques) which contain the glass-ceramic powder. Sealing is accomplished by placing the tape/mica/tape structure between the stack components to be sealed, followed by heat treatment (typically to 830ºC) to soften the glass sufficiently to cause bonding to the component surfaces. Final seal thickness is typically ~100-200 μm. (Figure 1)

Testing Procedures

For leak rate tests, the seals were placed between an Inconel600 pipe and an alumina substrate. A compressive load was applied

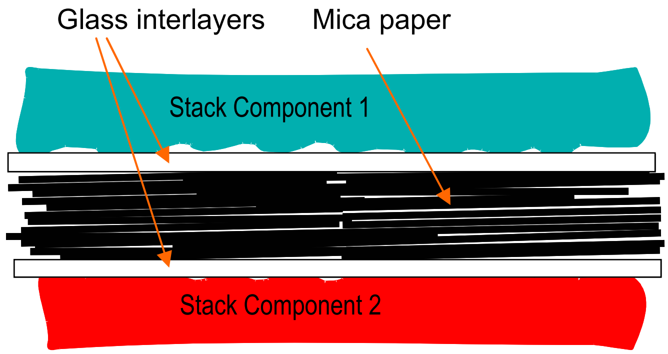

Figure 1. Schematic illustration of hybrid mica seal. Mica paper is in center of seal (black) with glass layers (white) on each side to fill mica/stack component interfaces.

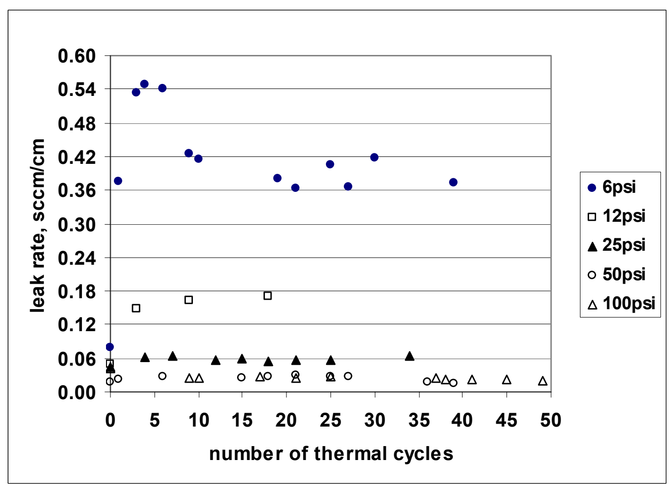

Figure 2. Leak rates at 800ºC of a 2" x 2" hybrid Phlogopite mica seal pressed under various compressive loads.

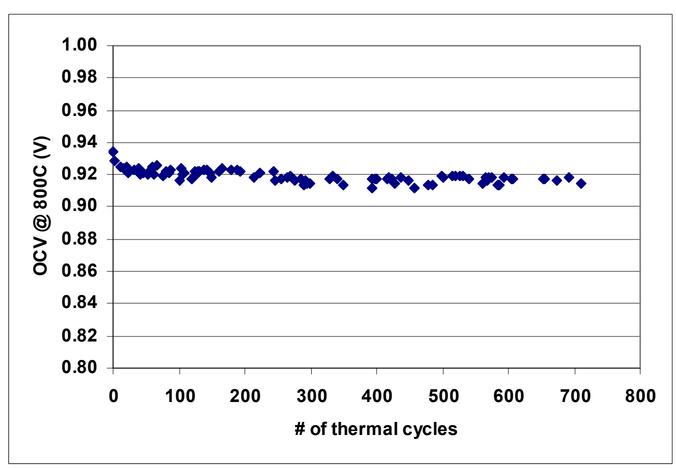

Figure 3. OCV results for thermal cycle tests on 2"x 2" 8YSZ plate with hybrid Phlogopite mica seal compressed at 100 psi.

throughout the tests, including the heating and cooling cycles. The leak rates were determined with high-purity helium using a 2 psi differential. Thermal cycling was conducted between 100°C and 800°C, with 2 hour dwells at 800ºC. Open circuit voltage (OCV) tests were also conducted, using electroded dense 8YSZ plates pressed between an Inconel cap and an alumina base support; in these tests the mica seals were located between the 8YSZ and the Inconel fixture.

Performance

Results for thermal cyclic leak rate testing of a 2"x 2" hybrid seal (1 layer of mica paper sandwiched between 2 layers of glass-ceramic) are shown in Figure 2. When compressed at ~25 psi or more, the seals exhibited low leak rates and excellent stability during repeated thermal cycling. It is important to emphasize that, in this test, the materials adjacent to the seal had a significant CTE mismatch (Inconel600, with a CTE of ~16-17 ppm/K vs. alumina, with a CTE of ~8 ppm/K). As a point of comparison, similar tests using a glass-ceramic seal alone between these materials resulted in seal failure after a single thermal cycle.

Results for OCV testing of a 2"x 2" seal compressed at 100 psi are shown in Figure 3. The OCV measurements were conducted using a dilute moist hydrogen "fuel" (2.55-2.72% H_2 / balance Ar) / 3% H_2O) vs. air, for which the calculated Nernst voltage at 800ºC is 0.932-0.934 V. Over 800 thermal cycles (heated from 100°C to 800°C in 30 minutes), the OCV decreased by only 2%.

Testing was also performed using lower applied compressive stresses. Figure 4 shows OCV as a function of thermal cycles and fuel flow rate for a 3.5"x 3.5" seal compressed at only 12.5 psi. These OCV measurements were conducted using moist pure hydrogen (97% H_2 / 3% H_2O) vs. air, for which the calculated Nernst voltage is 1.10 V at 800ºC. As an approximate reference point to help interpret the flow rate results, one can assume that the 3.5" x 3.5" footprint would correspond to a cell with an active area of ~55 cm^2. Assuming a current density of 0.7 A/cm^2 and 80% fuel utilization, a fuel flow rate of ~270 sccm would be required; for flow rates in that range, the measured OCV was essentially identical to the calculated Nernst voltage.

Note that the leak rates shown (Fig. 2) were measured at 2 psid; reduction of

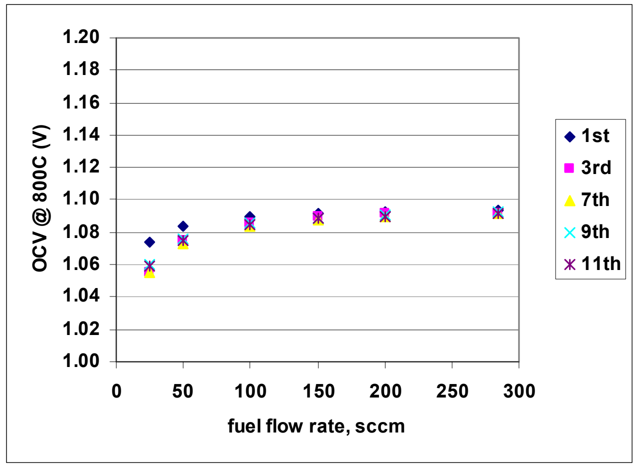

Figure 4. OCV of 3.5"x 3.5" hybrid Phlogopite mica seal as a function of fuel flow rate and thermal cycles, using reduced applied compressive stress (12.5 psi).

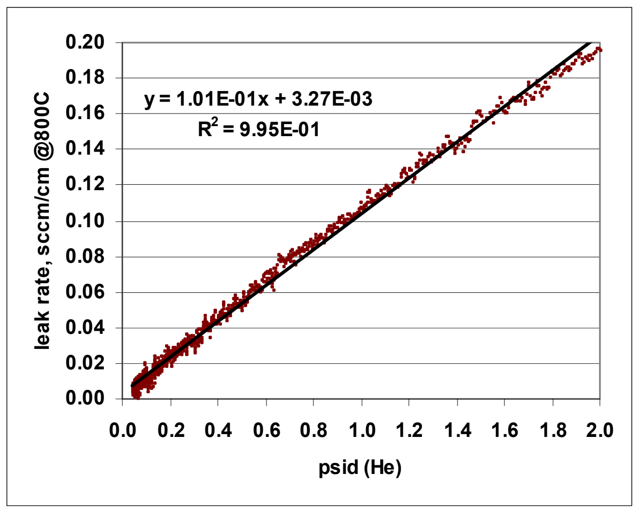

Figure 5. Effect of differential pressure on the leak rate of a 2"x2" hybrid Phlogopite mica seal pressed at 12.5 psi. Measurement was taken after 19 thermal cycles.

the pressure drop across the seal resulted in a linear decrease in leak rate. A typical example is shown in Figure 5 for a 2"x 2" hybrid Phlogopite mica pressed at 12.5 psi; the data in Figure 5 were taken after 19 thermal cycles. For pressure drops of 0.1-0.2 psid, the seal leak rate was ~0.02 sccm/cm.

Contacts

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