# EFFECT OF SEAL COMPOSITION ON THE PROPERTIES OF GLASS-MATRIX COMPOSITES FOR SOFC APPLICATIONS

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## 1 Introduction

A solid oxide fuel cell (SOFC), which consists of a number of basic cells, is an electrochemical device that converts chemical energy into electric power or that produces fuel gases. In a cell stack, the five essential components are the cathode, anode, interconnect, electrolyte, and seal. Seals are generally applied to the cell edges between the ceramic electrolyte and the metal interconnect. The seals used for SOFCs require low gas leak rates at elevated operating temperatures (700−900°C). Fused glasses or glass ceramics applied to stack components such as metal interconnects have been known to provide a good hermetic seal [1]. Glass matrix composites as sealing materials have been known to provide design flexibility, e.g., by controlling properties such as viscosity and seal operating temperatures [2]. In this work, the seal performance of a composite seal was studied according to gas leak rates and glass-based seal viscosity at elevated temperatures.

## 2 Experimental Procedures

### 2.1 Materials

A sealing glass (55 $SiO_2$, 10 $B_2O_3$, and 35 $La_2O_3$-SrO by mol%), was prepared by melting reagent-grade raw materials in a 50 ml Pt/Rh crucible at 1450°C for 2 h. The transformation range viscosities ($10^{10}$ to $10^{14}$ dPa·s) of the glass-based seals were measured according to the beam-bending method using a lab-constructed beam-bending viscometer.

### 2.2 Preparation and Evaluation of the Seals

Disc samples (~1 mm thickness and 21 mm diameter) were prepared through uniaxial pressing of the mixtures with varying amounts of glass powder and nano-sized alumina powder as a filler. The glass powder was obtained by passing the powdered glass through a 325-mesh screen. The seal samples used for seal tests, as well as for strength tests, were prepared by sintering the powder compacts at 800°C. A high-temperature leak testing apparatus was used to evaluate the sealing performance of the seal sample. To measure the leak rate in the temperature range of 740~800°C, an as-sintered seal was placed between a 17 mm–diameter Inconel 600 tube and an Inconel 600 block support. The applied compressive stress exerted vertically on the seal sample and the gauge pressure at which nitrogen gas was fed inside the tube were about 0.04−0.18 MPa and 0.5 kg/cm$^2$, respectively. Leak rates were calculated from the exponential decay curves obtained by plotting the pressure of the gas inside the tube as a function of the elapsed time.

## 3 Results and Discussion

### 3.1 Viscosity of the Sealing Glass

Figure 1 shows the temperature T and the viscosities $\eta$ of the pure sealing glass. The viscosity values, obtained using the beam-bending method, were fitted to the Vogel–Fulcher–Tammann (VFT) equation:

$$\text{Log}\,\eta = A + B/(T - T_o) \qquad (1)$$

From this fit, we obtained the values of the constants in the viscosity equation ($A$, $B$, and $T_o$). The continuous line in Fig. 1 was calculated using these values. The fit of the experimental values to the VFT equation allows us to estimate the viscosity in the range of log $\eta \approx 4$ to 10, within which it is difficult to experimentally determine the viscosity of the glasses. As seen in the figure, the viscosity of the bulk sealing glass gradually decreases as the temperature increases and is well fitted to the VFT equation. It can be seen that the viscosity of the sealing glass at 800°C is equal to $2.5 \times 10^8$ dPa·s (log $\eta$ = 8.4), which is two times lower than that of the glass seal at 780°C. It has been known that the sealing glass should have viscosity in the range of $10^7 \sim 10^{13}$ dPa·s in sealing temperature range.

3.2 Leak Rate of the Seals

Figure 2 shows the gas pressure drop of the composite seal containing 10% nano-sized alumina powder as a function of the logarithm of the elapsed time at 770°C. The leak rate L of the seal sample was calculated using the exponential decay law, which describes the characteristics of a gas leak within a closed vessel. If the initial pressure inside the cavity is $P_i$ and the pressure outside is $P_o$, the pressure will obey the following exponential equation:

$$P(t) = P_o + (P_i - P_o) \exp(-Lt/V) \qquad (2)$$

where L is the leak rate dependent on the seal composition, t is the time, and V is the volume of the closed vessel. By fitting Eq. (2) with the measured experimental data of pressure as a function of time, the leak rate L of the seal sample was calculated. As shown in Fig. 2, the pressure decay curves for the composite seal showed a leak rate of 0.005 sccm/cm at 770°C, which satisfies the requirement of SOFC seal, i.e., < 0.01 sccm/cm.

## 4 Conclusions

In this work, it was shown that the micron-sized alumina powder added to the glass have a certain effect to increase high temperature viscosity of glass. Based on the results of leak tests, it was concluded that the leaking properties of a composite seal containing 10 vol% alumina nano-powder satisfies the requirement of SOFC seal, i.e., < 0.01 sccm/cm.

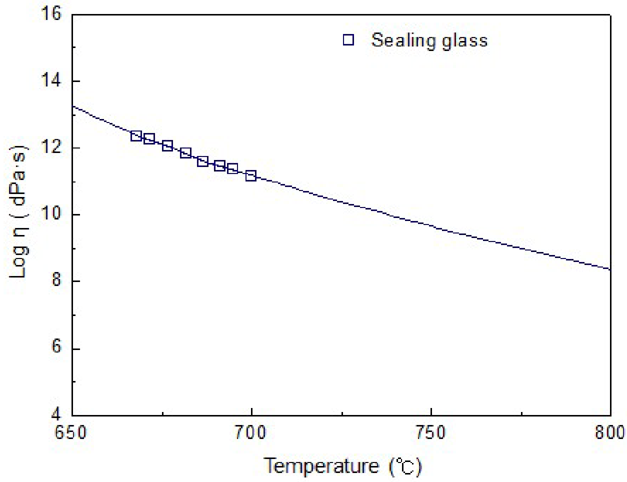


Fig.1. Viscosity curves of the bulk sealing glass.

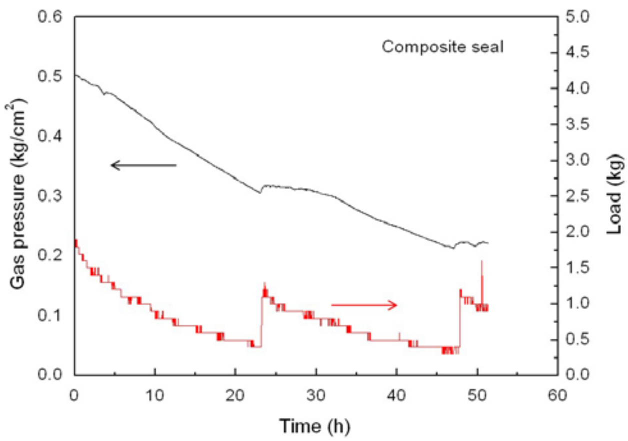


Fig.2. Pressure decay and the applied load curves for the composite seal at 770°C.

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## References

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