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EFFECT OF GLASS VISCOSITY ON GAS LEAK RATE IN GLASS SEALS FOR SOLID OXIDE FUEL CELL APPLICATIONS

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Abstract. Seals used for solid oxide fuel cells should have 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. Metal interconnects have a specific surface roughness in the nanometer-to-micron scale, depending on the manufacturing process employed to create the surface. Therefore, the viscosity of a fused glass at seal operating temperatures is expected to affect seal performance. In this work, the seal performance of two different glasses was compared by measuring gas leak rates and glass viscosity at elevated temperatures. It was shown that for sealing a metal with a rough surface, the working viscosity of the glass seal should be properly chosen to reduce leak rates at the glass/metal interface.

1. INTRODUCTION

A solid oxide fuel cell (SOFC), which is composed of a number of basic cells, is an electrochemical device that converts chemical energy into electric power or produces fuel gases [1]. In a cell stack, the cathode, anode, interconnect, electrolyte, and seal are five essential components. Seals are generally applied to the cell edges between the ceramic electrolyte and 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 [2]. The metal interconnect has a surface roughness in the nanometer-to-micron scale, depending on the manufacturing process employed to create the surface. For glass seals operating above the glass transition temperature, viscoelastic deformation occurs in the contact region at the glass seal/metal substrate interface. Therefore, the viscosity of a fused glass at seal operating temperatures is expected to affect seal performance. In this work, the seal performance of two different glasses was compared by measuring gas leak rates and glass viscosity at elevated temperatures. The objective of this study is to determine the optimal glass viscosity for sealing metals with a rough surface.

2. EXPERIMENTAL PROCEDURES

2.1. Materials

A sealing glass, designated as SLSB7-1 (55 SiO_2, 10 B_2O_3, and 35(La_2O_3-SrO) by mol.%), was pre-

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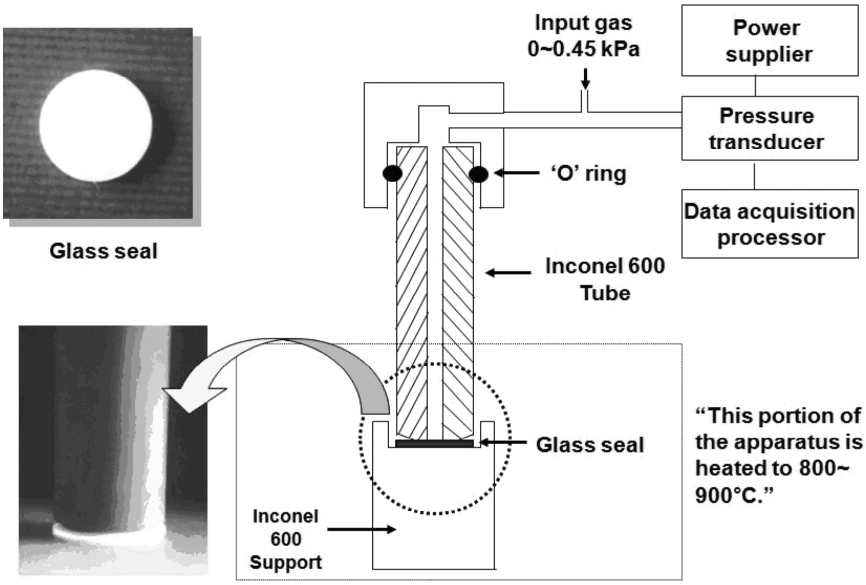

Fig. 1. Schematic of experimental setup for leak test.

pared by melting reagent grade raw materials in a 100-ml Pt/Rh crucible at 1450 °C for 2 h. The thermal properties of the bulk glass, i.e., transition point (T_g), dilatometric softening point (T_s), and the coefficient of thermal expansion, CTE (a), were measured using a dilatometer (Netzsch 402) at a heating rate of 10°C/min and a loading force of 15 cN. The glass transition temperature and CTE of the synthesized SLSB7-1 glass obtained by dilatometry were 652 °C and 11.8 ppm/K, respectively. Transformation range viscosities (10^{10} to 10^{14} dPa·s) of glass were measured by the beam-bending method using a homemade beam-bending viscometer [3]. The seals were prepared by passing powdered glass through a 325 mesh screen. In this work, commercial Pyrex 7740 glass was used for comparison because of its distinct temperature-dependent viscosity.

2.2. Preparation and evaluation of the seals

Disc samples (1.3-mm thickness and 21-mm diameter) were prepared by uniaxial pressing of the glass powders, SLSB7-1 and Pyrex. All samples used for seal tests were prepared by sintering the compacts at 850 °C for 1 h. As shown in Fig. 1, a high-temperature leak testing apparatus, similar to the one reported in literature, was built to evaluate the sealing performance of the seal samples [4]. To measure the leak rate in the temperature range of 810~910 °C, an as-sintered seal was placed between an Inconel 600 tube of 17-mm diameter and Inconel 600 block support. The support was then raised to force contact between the seal and the end of the Inconel tube at temperatures above the softening point of glass. The end of the Inconel tube used in this work had a rough oxidized surface texture. 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 0.32 MPa and 0.45 kg/cm^2, respectively. Leak rates were calculated from the exponential decay curves obtained by plotting the pressure of the nitrogen gas inside the tube as a function of the elapsed time [5,6].

3. RESULTS and DISCUSSION

3.1. Viscosity of the sealing glass

Fig. 2 represents the viscosity η of two glasses, SLSB7-1 and Pyrex, as a function of temperature T. The viscosity values obtained using the beam-bending method have been fitted to the Vogel–Fulcher–Tammann (VFT) equation [3]:

$$\log\eta = A + B/(T - T_0). \qquad (1)$$

From this fit, the values of the constants in the viscosity equation (A, B, and T_0) were obtained. The continuous line in Fig. 2 was calculated from these values. The fit of the experimental values to the VFT equation allows us to estimate the viscosity in the range $\log\eta \approx 6$ to $\log\eta \approx 10$, within which it is difficult to experimentally determine the viscosity of the glasses. As seen in the figure, the viscosity of the

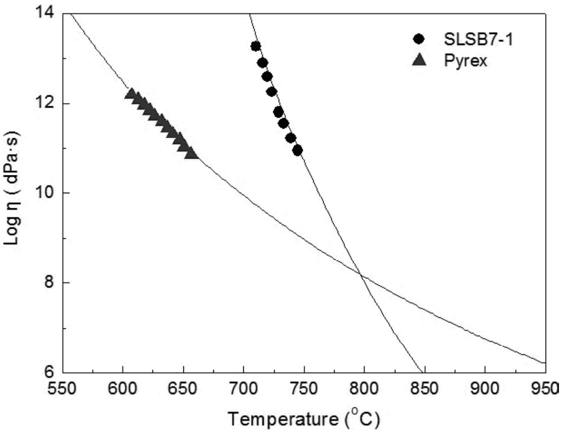

Fig. 2. Viscosity-temperature curves for SLSB7-1 and Pyrex glass.

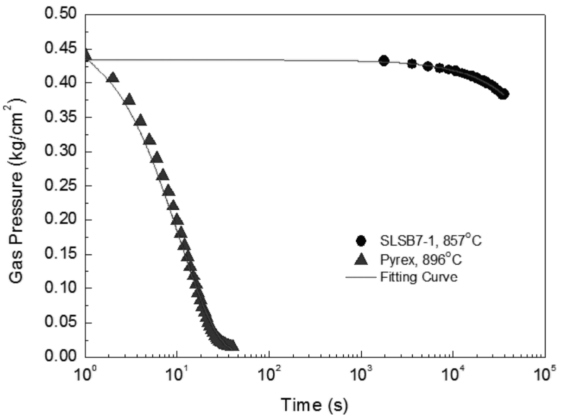

Fig. 3. Pressure decay curves for two different glass seals as a function of elapsed time.

SLSB7-1 sealing glass is higher than that of the Pyrex glass, until the temperature reaches 800 °C. For T > 800 °C, the SLSB7-1 sealing glass is less viscous than the Pyrex glass. The viscosity difference increases with temperature. It can be seen that at 850 °C, the viscosities of the SLSB7-1 and the Pyrex glass are equal to 1.3×10^6 dPa·s (logη = 6.1) and 2.5×10^7 dPa·s (logη = 7.4), respectively. It should also be noted that the viscosity of the Pyrex glass at 900 °C is equal to 6.3×10^6 dPa·s (logη = 6.8), which is still five times higher than that of the SLSB7-1 glass at 850 °C, i.e., 1.3×10^6 dPa·s (logη = 6.1).

3.2. Leak rate of the seals

In Fig. 3, the gas pressure drop of seal samples prepared using SLSB7-1 and Pyrex glass are plotted as a function of the logarithm of elapsed time. The leak rate L was calculated by using the exponential decay law, which describes the characteristics of a gas leak within a closed vessel [5,6]. 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:

$$P(t) = P_0 + \left(P_i - P_0\right)\exp\left(-Lt/V\right), \quad (2)$$

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. The calculated leak rate was converted to the standard leak (L, in standard cubic centimeters per minute at STP, sccm), which was further normalized with respect to the outer leak length (5.3 cm) of the Inconel tube. For example, as shown in Fig. 3, the pressure decay curves for a seal prepared using SLSB7-1 glass showed a leak rate of 0.0038 sccm/cm at 857 °C. The leak rate of the Pyrex glass seal was 90 sccm/cm at 896 °C, almost 250,000 times higher than that of the SLSB7-1 glass seal.

In Fig. 4, gas leak rates of the two seals are plotted as a function of sealing temperature. The gas leak rates of the SLSB7-1 glass seal varied from 0.001 to 0.006 sccm/cm over the temperature range 810~860 °C. For the Pyrex glass seal, gas leak rates varied from 43 to 80 sccm/cm over the temperature range 860~910 °C. The glass with lower viscosity employed in this work showed good sealing capability owing to its high spreading capability. However, it should be pointed out that the sealing property of a glass should be determined not only by its viscosity but also by its wetting behavior.

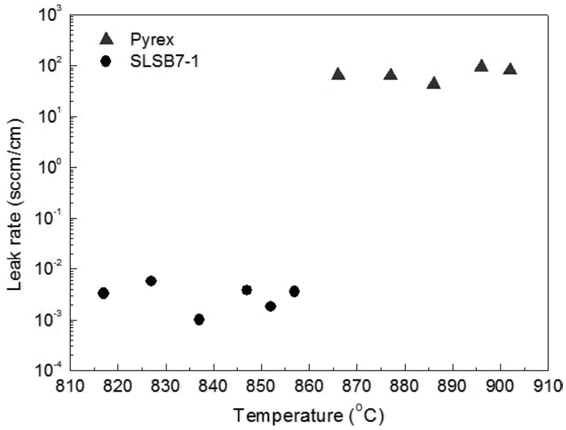

Fig. 4. Leak rates of two different glass seals as a function of temperature.

Glasses of different composition have different wetting properties, as well as viscosity. The con-

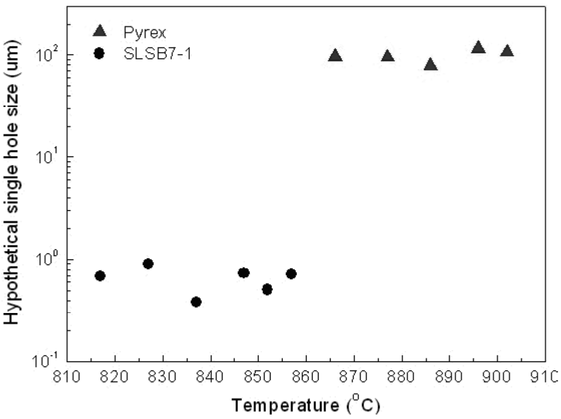

Fig. 5. Hypothetical single hole size for two different glass seals.

tact angle between fused glasses and a substrate represents wetting behavior. In most cases, a low contact angle less than 90° indicates good wetting and is expected to provide better sealing performance. However, as a sealing material for SOFCs, the contact angle needs to be greater than 90° to prevent fused glass from spreading to SOFC components [7]. The contact angles of the two glasses, SLSB7-1 and Pyrex, were measured on an Inconel 600 substrate at temperatures of 860 and 900 °C, respectively. The measurement results showed that both glasses had a contact angle greater than 90°; this means that the fused glasses used in this study do not appear to wet the substrate on a macro-scale. Therefore, it seems reasonable to conclude that the leaking properties shown in this work primarily depend on glass viscosity.

3.3. Hypothetical single hole size at the seal interface

Ideal leak assumes a gas leak through a circular hole of area *A*. The standard leak rate *L* derived using two well-known laws, the Ideal Gas Law and the Equipartition of Energy Law, is given by [5,6]:

$$L = A\left(RT/m\right)^{0.5}/2, \qquad (3)$$

where *R* is the gas constant, *T* is the temperature and *m* is the molar mass of the gas. This equation suggests that the leak rate is proportional to the square of the radius of a hypothetical single hole at the seal interface between the Inconel tube end and the glass seal. If we assume that the leak rate shown in Fig. 4 is achieved only by a hypothetical single hole, the estimated size ranges of the hypothetical single hole are, as shown in Fig. 5, 0.4~0.9 μm for SLSB7-1 glass seals and 80~115 μm for Pyrex glass seals. As mentioned earlier, the viscosity of the SLSB7-1 glass at 850 °C, i.e., 1.3×10^6 dPa·s (logη = 6.1) is five times lower than that of the Pyrex glass at 900 °C. This suggests that decreasing the viscosity of the sealing glass may decrease the size of the pore channels present at the seal interface. Otherwise, the surface roughness of the metal interconnects should be reduced to decrease gas leakage at the glass/metal interface.

4. CONCLUSIONS

In this work, the seal performance of two different glasses was compared by measuring gas leak rates and glass viscosities at elevated temperatures. The sealing property of the glass should be determined not only by its viscosity but also by its wetting behavior. Based on the results of contact angle measurements, it was concluded that the leaking properties shown in this work primarily depend on the glass viscosity.

For a Pyrex glass seal applied to an Inconel tube with a rough surface texture, the gas leak rates varied from 43 to 80 sccm/cm over the temperature range 860~910 °C. A synthesized glass, whose viscosity was five times lower than that of the Pyrex glass, reduced the gas leak rates to about 10^{-3} sccm/cm over the temperature range 810~860 °C. The size of the pore channels present at the seal interface could be reduced by decreasing the viscosity of the sealing glass. Alternatively, the surface roughness of metal interconnects should be reduced to decrease gas leakage at the glass seal/metal interface. Therefore, for sealing a metal with a rough surface, the working viscosity of the glass seal must be properly chosen to reduce leak rates at the glass/metal interface.

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