MEASUREMENT OF GAS LEAKS THROUGH THE CONTACTS BETWEEN HEAT RESISTANT ALLOY DISKS AND YSZ PLATES

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ABSTRACT

Gas seal technology for high temperature apparatus is essential to develop high performance planar SOFC stacks, and quantitative investigations of the gas leaks between electrolytes and separators are required. We designed an apparatus for measuring the gas leaks through the contact between heat resistant alloy disks and YSZ plates. By using the apparatus, gas leak rates of nitrogen and hydrogen gases through the contact between the materials were measured from room temperature to 1000°C. The gas leak rates were measured for various contact pressure and contact areas. As a result, it was found that when the temperature was raised from room temperature to 1000°C, gas leak rates of both nitrogen and hydrogen gases decreased considerably. The gas leak rate for hydrogen gas at 1000°C was about 1/3 of that at room temperature.

INTRODUCTION

Improvements in conventional gas seal technology are required to develop advanced planar SOFCs, especially the control of gas leak between electrolytes and separators is one of the most important subjects. The gas leak process at high temperature is complicated, because the different thermal expansions between separators and electrolytes induce shear stresses in the contact plane. Few data have been reported on the gas leak rate at high temperatures. In this paper, we designed an apparatus for measuring the gas leaks through the contact between heat resistant alloy disks and YSZ plates. By using the apparatus, gas leak rates of nitrogen and hydrogen gases through the contact between the materials were measured from room temperature to 1000°C.

EXPERIMENTAL

Leak test disk and YSZ plate

Fig. 1 shows leak test disks made of heat resistant alloy and YSZ plates. The YSZ
plate (Y$_2$O$_3$-8mol%ZrO$_2$ TOSOH Co.) is 90mm in diameter and 2mm thick. The contact surfaces of alloy and YSZ were polished. The heat resistant alloy disk is 80mm in diameter and 10mm thick and made of Hastelloy (Cr:42mol%, Fe:36mol%, Mo:17mol%, MITSUBISHI MATERIALS Co.). It has a hollow which is 60mm in diameter and 5mm in depth, and has an inconel pipe for gas inlet. The heat resistant alloy disk was placed on the YSZ plate as shown in Fig. 1 (a), and the measurements were made.

**Apparatus for measuring gas leak rates**

Fig. 2 shows a schematic of the apparatus for measuring the gas leak rates. The leak test disk is placed in a furnace. A disk is pushed against a YSZ plate by weights through a refractory pipe, and is supported with a ceramic ball for weighting homogeneously. The apparatus is divided into a measurement side and a reference side. Reservoirs of 1000cm$^3$ are located at each side. The reservoirs prevent the fluctuations caused by the change of air temperature and atmospheric pressure. The manometer at measurement side is used for measuring the pressure of the system, and the micro manometer in Fig. 2 measures the pressure difference between the two reservoirs.

**Experimental procedure**

The leak test disk was placed in the furnace, and temperatures were raised. The measurement side was connected with the reference side and gases were introduced to the leak test disk and the reservoirs. Then the stop valve was shut and each side was connected with the micro manometer. Gas pressure in the reference side was constant, but in the measurement side, gas pressure decreases by the gas leak at the leak test disk and YSZ plate. The pressure difference between two sides was measured for 2~25min, and the curves were analyzed.

**RESULTS**

Fig. 3 shows pressure reduction curves for N$_2$ gas at different contact pressure at room temperature. The initial gas pressure is 50mmH$_2$O. It was found that when contact pressure increased, the pressure reduction rate decreased. We define a gas leak rate as the gas leakage volume (cm$^3$) divided by time (s), gas pressure (mmH$_2$O) and the length of contact area (cm). The dimension of the gas leak rate is cm$^2$(s·mmH$_2$O)$^{-1}$. Gas leak rates of ①, ② and ③ are 2.28x10$^{-6}$cm$^2$(s·mmH$_2$O)$^{-1}$, 1.84x10$^{-6}$ and 1.51x10$^{-6}$ respectively.

Fig. 4 shows pressure reduction curves for N$_2$ and H$_2$ gases at room temperature. Contact pressure was 110.3g/cm$^2$. It was found that the pressure reduction rate of H$_2$ was much larger than that of N$_2$. The gas leak rate of H$_2$ was 8.19x10$^{-6}$cm$^2$(s·mmH$_2$O)$^{-1}$ and
that of N\textsubscript{2} was 1.51x10\textsuperscript{6} cm\textsuperscript{2}(s\cdot mmH\textsubscript{2}O)\textsuperscript{-1}.

Fig. 5 shows heat resistant alloy disks with different contact areas. The contact areas of the heat resistant alloy disks were 22.0 and 11.8 cm\textsuperscript{2}, respectively. By using these disks, the gas leak rates for different contact areas with a weight of 2500 g were measured at room temperature (Table I). When H\textsubscript{2} was used, the gas leak rate of the disk with large contact area was 3/4 of that of the small contact area disk. When N\textsubscript{2} was used, the leakage ratio was 1/2. It was found that when contact area increased, gas leak rate decreased for both H\textsubscript{2} and N\textsubscript{2}.

Fig. 6 shows pressure reduction curves at different temperatures for N\textsubscript{2}. Before the measurements, the leak test disk was heated to 1000°C and the contact surface was covered with oxide film. First, the measurement was made at room temperature, then it was conducted at 1000°C, and the system was cooled to room temperature, then the last measurement was made. The leak test disk in the furnace was pressed at 2500 g, through the three measurements. When the temperature was raised from room temperature to 1000°C, gas leak rate decreased considerably. When the temperature was lowered from 1000°C to room temperature, gas leak rate increased again. The gas leak rates of ①, ② and ③ are 2.46x10\textsuperscript{-6} cm\textsuperscript{2}(s\cdot mmH\textsubscript{2}O)\textsuperscript{-1}, 9.59x10\textsuperscript{-7} and 4.83x10\textsuperscript{-6}, respectively.

Fig. 7 shows pressure reduction curves at different temperatures for H\textsubscript{2}. They were measured in the same condition as for N\textsubscript{2}. The result showed the same tendency as that of N\textsubscript{2}. The gas leak rates of ①, ② and ③ are 8.48x10\textsuperscript{-6} cm\textsuperscript{2}(s\cdot mmH\textsubscript{2}O)\textsuperscript{-1}, 2.52x10\textsuperscript{-6} and 1.64x10\textsuperscript{-5}, respectively.

**DISCUSSION**

As shown in Table I, the ratio of contact area for two disks was 1:2. As the weight was equal, the contact pressure of the large contact area disk was half of the pressure of the small contact area disk. The results in Table I show that the gas leak rates are affected by contact area rather than contact pressure.

It is noted that the gas leak rates decreased at high temperature as shown in Fig. 6 and 7. The origin of the reduction of the gas leak at high temperature may be related to the considerable lowering of the Vickers hardness of YSZ from 1300 kgf(mm\textsuperscript{2})\textsuperscript{-1} at room temperature to 400 kgf(mm\textsuperscript{2})\textsuperscript{-1} at 1000°C. In other words, it can be presumed that the decrease of gas leak was caused by the closer contact of ductile materials.
CONCLUSIONS

By the investigation of the gas leak rate through the contacts between heat resistant alloy disks and YSZ plates, the following results were obtained. When the contact pressure increases, gas leak rate decreases at room temperature. The gas leak rate of H₂ is about five times as large as that of N₂ at room temperature. When the contact area increases, gas leak rate decreases at room temperature. It was found that the gas leak rate of N₂ at 1000°C is about 1/3 of that at room temperature, and the gas leak rate of H₂ at 1000°C is about 1/3 of that at room temperature.

Table I  Gas leak rates of N₂ and H₂ for different contact areas

| Contact area (cm²) | Contact pressure (g/cm²) | H₂ (cm²·s⁻¹(mmH₂O)⁻¹) | N₂ (cm²·s⁻¹(mmH₂O)⁻¹) |
|-------------------|--------------------------|------------------------|------------------------|
| 22.0              | 114                      | 8.19x 10⁻⁶             | 1.51x 10⁻⁶             |
| 11.8              | 212                      | 1.12x 10⁻⁵             | 3.12x 10⁻⁶             |

Weight: 2500g

Fig. 1  Leak test disks of heat resistant alloy and YSZ plates.
Fig. 2 Schematic of the apparatus for measuring gas leak rates.

Fig. 3 Reduction of N₂ gas pressure as a function of contact pressure, initial gas pressure: 50mmH₂O.

| Contact pressure (g/cm²) | Variation of gas pressure (mmH₂O) |
|-------------------------|-----------------------------------|
| 13.4                    | 1                                 |
| 41.1                    | 2                                 |
| 110.3                   | 3                                 |
Fig. 4 Reduction curves for N$_2$ and H$_2$ gases, initial gas pressure: 50mmH$_2$O, contact pressure: 110.3g/cm$^2$.

Fig. 5 Heat resistant alloy disks with different contact areas.
Fig. 6 Pressure reduction curves for N\textsubscript{2} at different temperatures, initial gas pressure: 50mmH\textsubscript{2}O, contact pressure: 110.3g/cm\textsuperscript{2}.

Fig. 7 Pressure reduction curves for H\textsubscript{2} at different temperatures, initial gas pressure: 50mmH\textsubscript{2}O, contact pressure: 110.3g/cm\textsuperscript{2}.