Enhancement of fuel transfer in anode-supported honeycomb solid oxide fuel cells

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Abstract. An anode-supported honeycomb solid oxide fuel cell can achieve high volumetric power density and improve thermo-mechanical durability at high temperatures. We have so far shown the promising power densities and investigated the effect of flow channel configurations on the cell performance in terms of the hydrogen partial pressure distributions in the cell under operation. In the present study, current-voltage characteristics of the cell depending on thicknesses of the porous anode substrate and forced convection in the substrate are studied under different flow rates of fed hydrogen to clarify the effect of 3-dimensional fuel transport in the porous anode substrate on the cell performance.

1. Introduction
Solid Oxide Fuel Cells (SOFCs) have advantages such as power generation with high efficiency and fuel flexibility owing to its high operating temperature. Moreover, high operating temperatures enable SOFCs to work without noble metal electrocatalysts. To date, planar and tubular type cells have been developed for SOFC systems. However, both types still have problems to be resolved. For example, planar type SOFCs need the development of the durability against the thermo-mechanical stress and the flow channel design, while tubular types are required to improve volumetric power density. Thus, improvements in the durability at high temperature and the volumetric power density of SOFC systems are of great interest. Honeycomb SOFCs are expected to give respectable durability and volumetric power density at high temperatures [1-8]. However there have been very few researches on anode-supported honeycomb SOFC [7, 8]. The anode-supported cell concept can decrease the ohmic loss by thin electrolyte layer and anode overpotential by increase in the anode surface area [9]. The aim of the present study is testing the performance of the anode-supported honeycomb cells fabricated in-house having different flow channel configurations under different hydrogen flow rates.

2. Experimental
2.1. Fabrication of the Anode-supported Honeycomb SOFC
An Ni/8YSZ (8 mol% yittria stabilized zirconia, NiO/YSZ: 65/35 wt%) honeycomb porous substrate with 3×3 flow channels (Repton Co. Ltd.) was used as the anode. The porosity was ca. 37% with the reduced Ni. It had 6 mm square channels with a wall thickness of 0.5 mm or 1.0 mm. The cell height was 18 mm. This anode substrate was coated with 8YSZ electrolyte slurry by dip-coating, being followed by co-firing at 1420 °C for 2 hours. Figure 1 shows the honeycomb cell after firing. The
The cathode slurry was a mixture of La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSM) and 8YSZ with a weight ratio of 10/3 [10]. The LSM-YSZ composite cathode slurry was applied on the electrolyte layer by brush coating and fired at 1150 °C for 2 hours. Different flow channel configurations were achieved by changing electrolyte applied channel with masking methods. Figure 2 shows the current collection method. “A” represents the anode flow channel and “C” represents the cathode flow channels, respectively. We coat electrolyte layer on the anode substrate, being followed by coating the cathode layer over the electrolyte layer. As shown in Figure 2, the anode and cathode are electrically connected by Pt wires, which were attached to the anode and cathode surfaces in the fuel inlet and outlet edges, separately [7, 8].

2.2. Performance Testing

We prepared four various honeycomb cells having two types of flow channel configuration as illustrated in Figure 3 with different porous anode substrate thicknesses of 0.5 mm and 1.0 mm shown in Figure 4. In the Type-A cell, five channels are anodes, while the other four are cathodes. Five anode channels are supplied with fuel and four cathode channels are supplied with air. In the Type-B cell,
five channels are anodes, while the other four are cathodes. Central anode channel is fed with fuel and outlet of the channel is closed. Four cathode channels are fed with air. Since H₂ diffuses inside the porous anode substrate, we regard the whole cathode surface as electrochemical active area. The active areas of the porous substrate thickness of 0.5 mm and 1.0 mm cells are calculated as 9.6 cm² and 8.5 cm², respectively.

**Figure 3.** Schematic drawings of the flow channel configurations of the (a) Type-A and (b) Type-B honeycomb SOFCs. A and C represent anode and cathode channels, respectively.

**Figure 4.** Anode-supported honeycomb SOFCs with different thickness of the porous anode substrates (a) 0.5 mm and (b) 1.0 mm.

Temperature of the honeycomb cell was maintained at 850 °C by a tubular electric furnace at open circuit voltage (OCV). Mass flow controllers (SEC-E40MK3, Horiba STEC) were controlled by LabView 8.5 (National Instruments Inc.) on a personal computer through an I/O device (NI USB-6008, National Instruments Inc.). Anode and cathode were supplied with H₂/N₂ mixture gas and air, respectively. The NiO anode was reduced to Ni by feeding H₂/N₂ mixture gas for two hours prior to measurements. During measurements, anode and cathode were fed in co-flow configuration with mixtures of H₂/N₂ and dried air at constant flow rates, respectively. For both types of cell, inlet gas flow rates for the anode were H₂/N₂: 40/40, 60/60, 80/80, 100/100, and 200/200 cm³/min in total, while that for the cathode was air: 400 cm³/min in total, respectively. Current voltage (I-V) characteristics of the cells were measured with an electric load (PLZ164WA, Kikusui Electronics Corp.).
3. Results and Discussion

Figure 5 shows the I-V characteristics of the two types of the honeycomb cells. One of the voltage losses of the SOFC is the concentration overpotential. When the fuel depletion occurs, the concentration overpotential becomes rather large. This leads to a significant decrease in the cell voltage with an increase in the current density. In the case of the low inlet gas flow rates, the fuel depletion becomes substantial, leading to the cell voltage drops shown in Figure 5.

Inlet gas flow rates, gas utilization at the maximum power, and maximum volumetric power density are listed in Tables 1 and 2. The maximum power densities for the Type-A05 and A10 cells under different gas flow rates derived from the I-V characteristics in Figure 5 are 0.03, 0.12, 0.21, 0.29 W/cm$^3$ at the hydrogen utilisations of 9, 19, 27, 23, 14 % and 0.04, 0.11, 0.11, 0.11, 0.25 W/cm$^3$ at 9, 19, 15, 13 %, respectively.

The thicker porous anode substrate exhibits lower power densities due to more hydrogen diffusing in the porous anode substrate in the upstream part and being consumed there. Thus hydrogen mole fraction is possibly smaller in the downstream, resulting in an increased voltage drop with the Nernst loss [11]. Furthermore, in the thinner cell, since hydrogen diffusion layer adjacent to the electrolyte is thinner, the diffusion flux becomes larger and performance becomes better. These power densities are promising compared to the electrolyte-supported honeycomb SOFCs considering the hydrogen utilizations [2-6]. This is due possibly to lower ohmic loss achieved by the thin electrolyte layer and 3-dimensional hydrogen transport in the porous anode-supported honeycomb cell.

![Figure 5. I-V characteristics of the (a) Type-A05 and (b) Type-A10 honeycomb cells at 850 °C, H$_2$/N$_2$: 40/40, 60/60, 80/80, 100/100 and 200/200 cm$^3$/min, Air: 400 cm$^3$/min in total.](image)
Table 1. Inlet gas flow rates and fuel utilizations for the Type-A05 cell at the maximum power.

| Flow rate (cm$^3$/min) [Utilization at the maximum power] | Maximum volumetric power density (W/cm$^3$) |
|--------------------------------------------------------|-------------------------------------------|
| H$_2$40 [9%], N$_2$40, Air400                           | 0.03                                      |
| H$_2$60 [19%], N$_2$60, Air400                          | 0.12                                      |
| H$_2$80 [27%], N$_2$80, Air400                          | 0.18                                      |
| H$_2$100 [23%], N$_2$100, Air400                        | 0.21                                      |
| H$_2$200 [14%], N$_2$200, Air400                        | 0.29                                      |

Table 2. Inlet gas flow rates and fuel utilizations for the Type-A10 cell at the maximum power.

| Flow rate (cm$^3$/min) [Utilization at the maximum power] | Maximum volumetric power density (W/cm$^3$) |
|--------------------------------------------------------|-------------------------------------------|
| H$_2$40 [9%], N$_2$40, Air400                           | 0.04                                      |
| H$_2$60 [19%], N$_2$60, Air400                          | 0.11                                      |
| H$_2$80 [19%], N$_2$80, Air400                          | 0.11                                      |
| H$_2$100 [15%], N$_2$100, Air400                        | 0.11                                      |
| H$_2$200 [13%], N$_2$200, Air400                        | 0.25                                      |

Figure 6 shows the I-V characteristics of the two types of the honeycomb cells. Decrease in the inlet gas flow rates also results in the fuel depletion, giving the cell voltage drops in Figure 6.

Inlet gas flow rates, gas utilization at the maximum power, and maximum volumetric power density are listed in Tables 3 and 4. The maximum power densities for the Type-B05 and B10 cell are 0.08, 0.07, 0.06, 0.21, 0.39 W/cm$^3$ at 23, 15, 8, 18, 20 % and 0.07, 0.08, 0.12, 0.10 W/cm$^3$ at 9, 13, 16, 13%, respectively, from the I-V characteristics in Figure 6.

Figure 6. I-V characteristics of the (a) Type-B05 and (b) Type-B10 honeycomb cells at 850 °C, H$_2$/N$_2$: 40/40, 60/60, 80/80, 100/100 and 200/200 cm$^3$/min, Air: 400 cm$^3$/min in total.
In the case of Type-B cell, the thinner porous anode substrate tend to give larger power densities with higher gas flow rate because of the smaller diffusion length, leading to the better performance with the smaller voltage drops by the larger hydrogen flux.

The performance of the Type-B cell is higher than that of Type-A cell at the high inlet flow rates. In the Type-B cell, hydrogen fed only into the central channel is transported to other anode channels through porous anode substrate, giving rise to the forced convection in the substrate because the outlet of central channel is closed. In addition, hydrogen diffusion length is smaller in the Type-B cell by the forced convection in the porous substrate. In the case of the low inlet gas flow rate, smaller inlet pressure possibly results in smaller convective flow rate and hydrogen partial pressure in the upstream parts of the substrate adjacent to the outer channels, so the performance becomes low. This effect is probably more pronounced in the thinner substrate under the small inlet gas flow rate conditions by the higher flow resistivity with the smaller cross sectional area. We will numerically evaluate the hydrogen mole fraction distributions in the honeycomb cell using finite element method in our future work.

| Flow rate (cm$^3$/min) | Maximum volumetric power density (W/cm$^3$) |
|------------------------|--------------------------------------------|
| $\text{H}_2\text{A} [23\%], \text{N}_2\text{A}, \text{Air}400$ | 0.08                                      |
| $\text{H}_2\text{A} [15\%], \text{N}_2\text{A}, \text{Air}400$ | 0.07                                      |
| $\text{H}_2\text{A} [8\%], \text{N}_2\text{A}, \text{Air}400$ | 0.06                                      |
| $\text{H}_2\text{A} [18\%], \text{N}_2\text{A}, \text{Air}400$ | 0.21                                      |
| $\text{H}_2\text{A} [20\%], \text{N}_2\text{A}, \text{Air}400$ | 0.39                                      |

Table 3. Inlet gas flow rates and fuel utilizations for the Type-B05 cell at the maximum power.

| Flow rate (cm$^3$/min) | Maximum volumetric power density (W/cm$^3$) |
|------------------------|--------------------------------------------|
| $\text{H}_2\text{A} [9\%], \text{N}_2\text{A}, \text{Air}400$ | 0.07                                      |
| $\text{H}_2\text{A} [13\%], \text{N}_2\text{A}, \text{Air}400$ | 0.08                                      |
| $\text{H}_2\text{A} [16\%], \text{N}_2\text{A}, \text{Air}400$ | 0.12                                      |
| $\text{H}_2\text{A} [13\%], \text{N}_2\text{A}, \text{Air}400$ | 0.10                                      |

Table 4. Inlet gas flow rates and fuel utilizations for the Type-B10 cell at the maximum power.

In the case of Type-B cell, the thinner porous anode substrate tend to give larger power densities with higher gas flow rate because of the smaller diffusion length, leading to the better performance with the smaller voltage drops by the larger hydrogen flux.

The performance of the Type-B cell is higher than that of Type-A cell at the high inlet flow rates. In the Type-B cell, hydrogen fed only into the central channel is transported to other anode channels through porous anode substrate, giving rise to the forced convection in the substrate because the outlet of central channel is closed. In addition, hydrogen diffusion length is smaller in the Type-B cell by the forced convection in the porous substrate. In the case of the low inlet gas flow rate, smaller inlet pressure possibly results in smaller convective flow rate and hydrogen partial pressure in the upstream parts of the substrate adjacent to the outer channels, so the performance becomes low. This effect is probably more pronounced in the thinner substrate under the small inlet gas flow rate conditions by the higher flow resistivity with the smaller cross sectional area. We will numerically evaluate the hydrogen mole fraction distributions in the honeycomb cell using finite element method in our future work.

4. Conclusions
Relations between the fuel transport and the performance change attributed to the difference of the porous anode substrate thickness and the forced convection are investigated. Moderately thin porous anode substrate exhibits higher volumetric power density, giving promising peak power compared with electrolyte-supported honeycomb cells. Since too much fuel diffusion to the outer channel is prevented in the upstream, fuel transport in the downstream is retained. Moreover, the performance becomes large by larger hydrogen diffusion flux with the thinner substrate. Forced convection by the closed outlet of the central anode flow channel gives better performance at high inlet fuel flow rate. Hydrogen diffusion flux is increased by the smaller diffusion length owing to the forced convection in the porous substrate. Hence the substrate thickness and flow channel configuration determine the
performance in terms of the hydrogen diffusion flux and the Nernst loss associated with the hydrogen partial pressure.

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