DEVELOPMENT OF A PLANAR SOLID OXIDE FUEL CELL MODULE AT SANYO

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ABSTRACT

A large-sized cell and bipolar plate, and stacking technology have been developed for a 1 kW class planar solid oxide fuel cell (SOFC) module. A maximum output power exceeding 1 kW and a power density of 0.22 W/cm² were achieved with a 160 cm² × 30-cell stack. The endurance of the module, in particular, the effect of a cathode second layer, was investigated by operating the module for 1870 hours. As a result, the module output power degradation rate was 4.4 %/1000 hours at 1030 hours on the basis of the current density of 0.3 A/cm². By means of supplying both fuel and oxidant gas on the upper side of a stack, the effective diffusion of the heat in the upper part of the module was demonstrated. In addition, the combined cell stacked module concept was reported.

INTRODUCTION

Our current R&D goal for 1995 is to achieve a few kW output power with a combined cell stacked planar solid oxide fuel cell (SOFC). This type of module can be expected to have a compact body, a simple system, high reliability and low cost. Many kinds of single cell stacked modules have previously been studied. In 1993, an output power exceeding 500 W was achieved with a 130 cm² × 20-cell stack, a 500 W class module(1). These modules were investigated for the development of a large-sized cell and stacking technology, in other words, for the uniformity of individual cell performance and that of individual cell operating condition. On the other hand, the composite electrode and the surface treatment of an alloy bipolar plate have been studied with single cells(2,3). By coordinating the set of results, a 1 kW class module has been developed and operated for the development of a combined cell stacked module.

1 KW CLASS MODULE

Stack design
The stack of a 1 kW class module was made of 30 sheets of 150 mm \times 200 mm cell with an active electrode area of 160 cm\(^2\). An inner manifold and counter-flow configuration and the oxidant gas flow configuration with side exhaust, as shown in Figure 1, were adopted as well as a 500 W class module. The authors reported that the oxidant gas pressure decreased to a level sufficient for large-sized cells by adopting the oxidant gas flow configuration with side exhaust\(^{(1)}\).

Figure 2 shows a schematic view of the gas routes of a 1 kW class module and of a 500 W class module. In the former, both fuel and oxidant gases were supplied on the stack upper side, though in the latter, only fuel gas was supplied on the stack lower side. These gases entering the top plates were preheated.

In addition, each bipolar plate is provided with two or eleven side measuring holes to monitor its temperature. The main measuring points were provided on the fuel inlet and the outlet side\(^{(2)}\).

Cell and bipolar plate fabrication

The stack components of a 1 kW class module are specified in Table I. As a self-supported type cell was adopted, the electrolyte thickness was 200 \(\mu\)m. The anode slurry of a mixture of anode materials, terpineol and poly vinyl butyral was screenprinted on the surface of an electrolyte, and was then sintered for 2 hours at 1300 \(^\circ\)C. Similarly, the cathode slurry was coated on the other surface and sintered for 4 hours at 1100 \(^\circ\)C.

A heat resistant alloy bipolar plate was made by a cutting process. Concerning the 1 kW class module, electrical discharge machining was used to shape the bipolar plate gas flow channel instead of cutting. This was because the strain which occurred on the bipolar plate from electrical discharge machining was smaller than that from cutting. As a result, 1 kW class module bipolar plates, which were larger than those

| Components     | Material                        | Thickness / \(\mu\)m |
|----------------|--------------------------------|---------------------|
| Electrolyte    | 3 mol% Y\(_2\)O\(_3\)-ZrO\(_2\)  | 200                 |
| Anode          | NiO-YSZ                         | 40                  |
| Cathode        | La\(_{0.9}\)Sr\(_{0.1}\)MnO\(_3\)-YSZ | 50                  |
| Cathode 2nd Layer | La\(_{0.9}\)Sr\(_{0.1}\)MnO\(_3\)-10 wt% La\(_2\)O\(_3\) | 50                  |
| Bipolar plate  | Inconel 600                    | 5000 ~ 6000        |
for a 500 W class module, could be prepared within the allowable bipolar plate surface tolerance. In addition, electrical discharge machining may reduce SOFC cost in mass production due to shorter work time needed for shaping the gas flow channel.

A heat resistant alloy, i.e., Inconel 600, must offer good performance concerning electrical conductivity, heat conductivity, gas-tightness, mechanical strength and ease of manufacturing, but it has been pointed out that chromium diffuses from the alloy to the cathode at the operating temperature, i.e., 1000°C, and subsequently degrades the properties of the cathode(3). It was found that the amount of diffused chromium can be suppressed with heat treatment and polishing to remove the bipolar plate surface oxidant layer(3). In a 1 kW class module, a cathode second layer was adopted for the suppression of chromium instead of alloy surface treatment. La2O3 included in the cathode second layer, as shown in Table I, reduces the CrO3 vapor pressure and consequently suppresses the amount of diffusing chromium. Figure 3 shows test cell (0.5 cm²) performances with the cathode second layer that reduces the influence of chromium diffusion from the alloy. To accelerate the phenomenon, a higher chromium concentration alloy, HRN-X (Cr 22 %, Fe 18.5 %, Mo 9 %, Co 1.5 %, W 0.6 %, C 0.1 %, Ni Bal.) was used. This figure indicates that the second layer including 10 wt% La2O3 can remove the influence of chromium diffusion for at least 700 hours. Hence La0.9Sr0.1MnO3-10wt%La2O3 was used for the 1 kW class module. The cathode second layer fabrication process was as well as anode or cathode, but it was sintered in situ for good adhesion of the layer to the bipolar plate rib.

Initial performance

A 1 kW class module was operated with H2 fuel gas humidified at room temperature and air oxidant gas. The V-I and power characteristics of the module at a fuel utilization (Uf) of 50 % and an oxidant utilization (Uox) of 30 or 35 % are shown in Figure 4. The maximum output power of 1064 W was achieved at the current density of 0.35 A/cm². As the maximum 1 kW class module power density of 0.22 W/cm² is a little superior to that of a 500 W class module, it can be considered that the module performance does not exhibit any reduction from using the large-sized cell and the increase of stacked cells except for the gas utilization characteristics. Fuel utilization characteristics at a current density of 0.3 A/cm² were gained at a Uf up to 50 %, because some gas cross leakage occurred in the module. The cell voltage distribution at the maximum output power is given in Figure 5. The uniform gas flow distribution among cells was suggested in terms of the average cell voltage of 0.633 V and the relatively uniform distribution.

Endurance test

The endurance of a 1 kW class module is evaluated from the result of operating it for more than 1800 hours. It is shown in Figure 6. After measuring various
characteristics, the module was operated at a constant current density of 0.3 A/cm² with $U_f / U_{ox} = 30\% / 15\%$ for about 600 hours. Continuous operation was then undertaken at a different $U_f$ of 40\%, but unexpected gas supply system problems occurred at about 1100 hours. Because a few cells of the module were damaged, the current density was decreased to 0.2 A/cm² up to a life time of 1870 hours. The module output power degradation rate was 4.4 \%/1000 hours at 1030 hours on the basis of the current density of 0.3 A/cm² with $U_f / U_{ox} = 30\% / 15\%$ at 101 hours. The evaluation indicates that the module endurance is relatively high in spite of some gas cross leakage.

The investigation of the individual cell endurance of the module suggests that the degradation rates of the cell near the middle of the stack were great. Figure 7 shows cross-section SEM images of anodes before operation (a), after operation (No.4 cell) (b), and after operation (No.18 cell)(c). The pore ratio was evaluated by image processing to separate the anode particle and the space (pore) on the cross-section. The pore ratio, especially that of No.18 cell, increases after operation. The result suggests that the Ni particles, especially those of the cell near the middle of the stack, sintered during the operation. No remarkable distribution was obtained by the similar evaluation of the pore ratio. Therefore the individual cell endurance of the module mainly depended on the operating temperature because of anode sintering.

It is considered that the module endurance is mainly attributed to the cathode second layer. Figure 8 shows the EPMA chromium analysis results of two cathode samples of the module after 1870 hours of operation and one of a test cell after degradation without a second layer. The chromium intensity was small in the case of the module (b) (c), though it was great at the interface between the cathode and the electrolyte in the case of the test cell(a). The results indicate that the cathode second layer offers the advantage of improved cell endurance by suppressing chromium diffusion to the interface between the cathode and the electrolyte. It is interesting that the intensity was markedly small in the case of the No.18 cell. As mentioned below, the temperature of the No.18 cell was higher than that of the No.4 cell, so that the second layer effect appears to depend on temperature.

**Temperature distribution**

Figure 9 shows the temperature distribution in the stack of a 1 kW class module and that of a 500 W class module under operation in a steady state. The peak of the temperature distribution of a 1 kW class module shifted to the middle of the stack, while the peak of a 500 W class module occurs in the upper part of the stack. The projection of the No.8 bipolar plate temperature was due to the cross leakage. So far the temperature difference in a stack increased in proportion to the up-scaling of the module scale(2). However, the temperature difference between all data of a 1 kW class module, except for No.8 data, was approximately 80 °C, which was almost the
same as that of a 500 W class module. These results demonstrated effective diffusion of the heat of the upper part of the module by changing the gas routes.

As mentioned above, the wide temperature distribution caused a great difference in the cell performance and endurance in the stack. Therefore, the temperature uniformity is emphasized in the development of a SOFC module.

**COMBINED CELL STACKED MODULE CONCEPT**

On the basis of the successful development of a 1 kW class module, a 2 kW class module has also been developed. An attempt was made to apply the combined cell stacked module concept to a 2 kW class module. This concept is illustrated in Figure 10. One layer of the stack contains 4 cells and one bipolar plate. Hence, high capacity but low cost SOFCs can be attained through mass production. Mechanical and thermal stress in one layer can be reduced in terms of separation of each cell in one layer. Especially, the combined cell stacked module is provided with a heat exchanging part at its center. As shown in the lower figure, the oxidant gas flows through the module before entering each layer, and consequently is expected to decrease the temperature difference across the layer and among the layers.

**SUMMARY**

As the large-sized cell and bipolar plate, and stacking technology were successfully developed, an output power exceeding 1 kW and a power density of 0.22 W/cm² were achieved with a 160 cm² X 30-cell stack. As a result of the 1 kW class module operating for 1870 hours, the module output power degradation rate was 4.4 %/1000 hours at 1030 hours on the basis of the current density of 0.3 A/cm² with Uf / Uox = 30 % / 15 %. By means of EPMA and SEM analysis, it was suggested that the cathode second layer was effective and cell endurance depended on operating temperature. By means of both fuel and oxidant gas supplied on the upper side of a stack, effective diffusion of the heat of the upper part of the module was demonstrated. For a 500 W class module, the temperature difference among cells was approximately 80 °C. In addition, the concept of a 2 kW class combined cell stacked module was reported.

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Figure 1. 1 kW class module.

Figure 2. Gas route schematic view of a 1 kW class module (a) and a 500 W class module (b).

Figure 3. Test cell performances with the cathode second layer that reduce the influence of chromium diffusion from the alloy.

Temperature : 1000°C
Oxidant : AIR
Fuel : H₂+3% H₂O
Current density : 0.3 A / cm²
Alloy : HRN-X
Cathode : La₀.₉Sr₀.₁MnO₃ (1 nm)
          80 wt% + YSZ (0.5 μm)
          20 wt%
Cathode 2nd. layer :
          La₀.₉Sr₀.₁MnO₃ (1 nm)
          (100-x) % + La₂O₃ x %
Cathode 2nd. layer thickness: 50 μm
Figure 4. The V-I and power characteristics of a 1 kW class module.

Figure 5. The cell voltage distribution of a 1 kW class module at the maximum output power.

Figure 6. Endurance of a 1 kW class module.
Figure 7. Cross-section SEM images of anodes in a 1 kW class module before operation (a); after operation (No.4 cell) (b); and after operation (No.18 cell) (c).

Figure 8. EPMA chromium analysis results of a test cell (a) and two cathode samples of a 1 kW class module (b) (c) after operation.
Figure 9. Temperature distribution in stacks under operation in a steady state.

Fuel: H₂ (humidified at R.T.)
Oxidant: Air
Current density: 0.3 A/cm²
Uₘ/Uₘₐₓ = 30/15

Figure 10. Concept of a 2 kW class combined cell stacked module.