Fabrication of 1.3kW Class Anode-Support Flat Tubular SOFC Stack

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ABSTRACT

KIER has fabricated anode-supported flat tubular SOFC stack for the intermediate temperature (700~800°C) operation. For this purpose, we have first fabricated anode-supported flat tubular cells by the optimization between the current collecting method and the induction brazing process. After that we designed the compact fuel & air manifold by adopting the simulation technique to uniformly supply fuel & air gas and the unique seal & insulation method to make the more compact stack. For making stack, the prepared anode-supported flat tubular cells with effective electrode area of 108cm2 connected in series with 37 bundles, in which one bundle consists of two cells connected in parallel. The performance of stack in 3% humidified H2 and air at 800°C showed maximum power of 1.3kW

INTRODUCTION

The SOFC has two type structures of the tubular and planar designs. The tubular design has been applied from small to large-scale power generation system. The planar design has also been studied for long time and was developed for the small power system. For the commercialization of SOFC technology, there are strong needs for establishing key technologies such as cell components and fabrication technology, high performance cell, and stacking technology, etc. In order to increase the durability and to allow cost reduction by less expensive materials such as metallic interconnect, many researchers are currently concentrating on development of SOFC operating at reduced temperatures below 800°C.

We developed first an anode-supported flat tubular SOFC (1-3) stack for the intermediate temperature operation. After our development, many research groups including Acumentrics started the development of the anode-supported tubular SOFC (4-7). In this work, we reported an anode-supported flat tubular SOFC stack to improve the power density of SOFC cell and make the compact stack system. The 1.3kW class anode-supported flat tubular SOFC stack was fabricated by the optimized fabrication method and its performance was evaluated.

EXPERIMENTAL

The extruded flat tubular anode support was served as fuel electrode and other cell components were fabricated in thin layers onto it. The 40 vol. % Ni – 8YSZ(8 mol. % Y_2O_3-stabilized ZrO_2) anode powder was prepared by mixing 8YSZ (TZ-8Y, Tosoh co.) and nickel oxide powders (J.T. Bakers co.). Anode powder and activated carbon as pore former were weighed and mixed in ethanol by ball milling for 14 days and then dried. Organic binder and 25 wt. % distilled water were added to the dried powder, and then the well dispersed paste was extruded in the form of flat tube. The extruded flat tubes were dried and in the drying oven at 120 °C for 12h. It was pre-sintered at 1100 °C. The YSZ electrolyte layer was coated on the pre-sintered anode tube by vacuum slurry dip coating to form a dense layer and co-fired at 1400 °C. Cathode material, LSM($(La_{0.85}Sr_{0.15})_{0.9}MnO_3$) and LSCF($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$) were prepared by solid state powder reaction method by weighing in a selected proportion and mixed in ethanol by ball milling for 10 days. The synthesized powder was calcined at 1000 °C for 5h. The multi-layered cathode composed of LSM/8YSZ composite, LSM, and LSCF were coated onto the co-sintered flat tube by slurry dip process and sintered at 1200 °C.

The flat tubular cell has four ribs inside the tube, which is considered to increase the mechanical strength and the electrical conductivity of the anode tube. The flat tubular cell was brazed with a metal cap by induction brazing process as shown in figure 1.

Fig. 1. Brazed anode-supported flat tubular cell with metallic cap by using BNI2 filler metal

The metal cap plays roles of the fuel flow path and the anode current collector. Two brazed flat-tube cells are connected in parallel, which is defined as a bundle. In one bundle, two brazed anode-supported flat tubular cell was connected in parallel. The anode-supported flat tubular

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SOFC was connected in-parallel with 2 cells and in-series again. The two-cell unit bundle was placed in air chamber. 3% humidified hydrogen as fuel was supplied through the chamber connected with brazing cap. The air was supplied through the air distributor. In the case of the fabrication for compact unit bundle, we designed the compact fuel & air manifold by adopting the simulation technique to uniformly supply fuel & air gas and the unique seal & insulation method to make the more compact unit bundle (Figure 2).

Fig. 2. Optimized fuel and air chamber design by using CFD simulation technique

The equipment of performance test was composed of a gas control system, a heating unit, a preheating system, humidified chambers, an electronic load, and a data acquisition system. The stack performance was evaluated by adjusting the electronic load. Using the same concept as that of 2 cell-unit bundle, the 1.3kW class stack (Figure 3) was designed and fabricated. The stack has the 37 bundles connected in series, that is, number of cells is 74 cells. Each active area of the flat tube SOFC cell is 108 cm^2. Total volume of SOFC stack including air & fuel chamber is about 15L and the stack performance was evaluated using hydrogen with 3 % water and air.

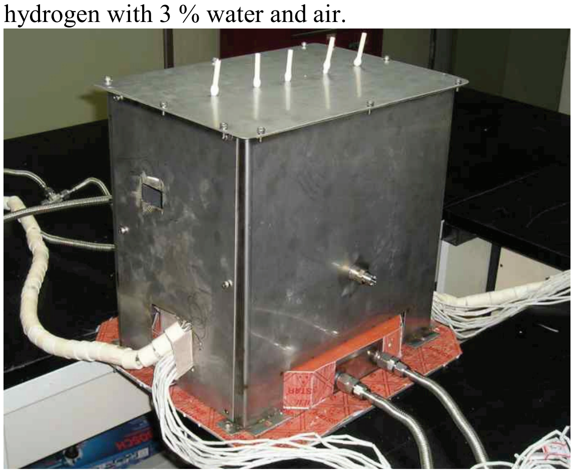

Fig. 3. Anode-supported 1.3kW class flat tubular SOFC stack

RESULT AND DISCUSSION

An electron current path of the cylindrical tubular type SOFC structure is geometrically determined by half circle of the cell, so the power per mass or per volume is much lower than the planar type. On the other hand, the flat-tube structure is possible to increase the power density per unit volume by giving the electron current path through the ribs. Its mechanical strength is higher as compared to the conventional planar type.

The thickness of the anode-supported flat-tube was 1.9 mm, and each of electrolyte and cathode layers had thickness of 7 (Figure 4), 25 µm, respectively. The electrolyte showed a dense microstructure without any detrimental defects.

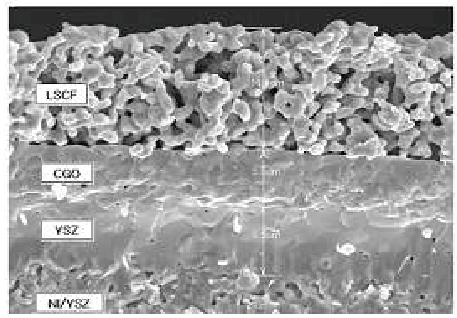

Fig. 4. SEM picture of the cross section between anode support and electrolyte and cathode and surface of electrolyte

Gas Permeability tests were conducted to estimate the gas-tightness of electrolyte. The gas permeability for cells as a function of differential pressure by using He gas was measured. It has shown the value below 4 x 10^{-8} $Lcm^{-2}sec^{-1}$ to the pressure difference of 3 atm.(Figure 5).

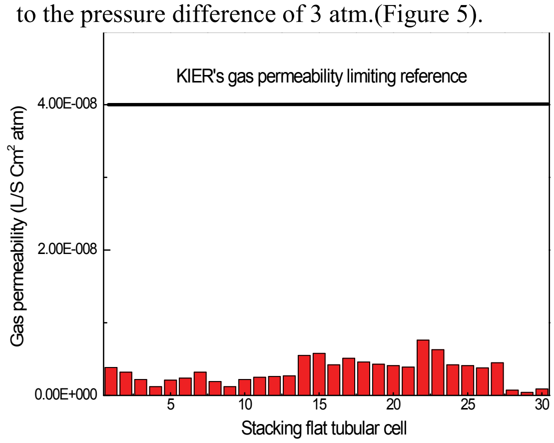

Fig.5 Gas Permeability of flat tubular SOFC cell

The prepared anode-supported flat tubular cells were brazed by using induction brazing process, in which the metal cap was bonded to the end of the flat tubular cell without cathode layer. The cathode current-collection was made by the wiring of silver wires. The induction brazed anode and metal cap play a role of current-collecting of anode.

The 1.3kW class anode-supported flat tubular SOFC stack was manufactured on the basis of the experimental date of 2 cell-unit bundle. The stack has the pre-heater in air and fuel inlets and during the stack operation the temperature of the preheated gases was set at 550°C. Figure 6 shows a performance of the 1.3kW class anode-supported flat tubular SOFC stack. The stack performances of 1.3kW in 3 % humidified H_2 and air were obtained. The stack furnace was set at 800°C but the measured stack temperatures were in the range of 790 to 810°C.

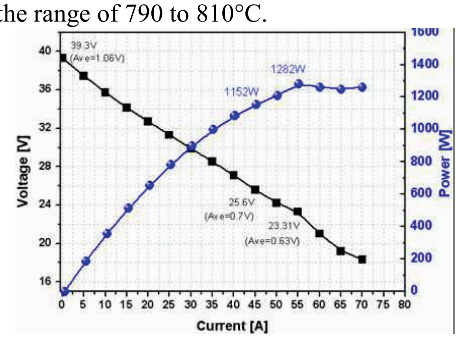

Fig. 6. Performance characteristic of the 1.3kW class anode-supported flat tubular SOFC stack

CONCLUSIONS

We have developed successfully the 1.3kW class anode-supported flat tubular SOFC stack. The end of the flat tube cell was connected with the metal cap by an induction brazing process. The brazed parts had efficient gas tightness. The metal cap played two roles of the anode current collection and the fuel supply path. The 1.3kW class anode-supported flat tubular SOFC stack with 74 cells (2x37) was designed and fabricated, which howed performances of 1.3kW in air at 800°C. The improvement of the stack is in progress.

REFERENCES

1. Song R-H., Kim E-Y., Shin D-R., Yokokawa H. Fabrication and Characteristics of Anode-Supported Tube for Solid Oxide Fuel Cell. In: Singhal SC, Dokiya M, editors. Proceedings of the International Symposium on Solid Oxide Fuel Cells VI. Honolulu, 17-22 October. New Jersey: The Electrochemical Society, Inc., 1999. p.845-850.
2. Kim E-Y., Song R-H., Lim Y-E., Fabrication and Characteristics of Anode-Supported Tube for Solid Oxide Fuel Cell by Wet Process. Korean J. Mater. Res. 2000; 10; 659-664.
3. Song R-H., Shin D-R., Kim E-Y., Yokokawa H. Fuel Electrode-Supported Tubular Solid Oxide Fuel Cell and Method of Manufacturing The Same. U.S. Patent 6,436,565 B1. Aug. 20, 2002.
4. Sammes N-M., Du Y.. The mechanical properties of tubular solid oxide fuel cells. J. Material Sci. 2003; 38: 4811-4816.
5. Du Y., Sammes N-M., Fabrication and properties of anode-supported tubular solid oxide fuel cells. J. Power Sources 2003; 136: 66-71.
6. Sammes N-M., Du Y., Bove R. Design and fabrication of a 100 W anode-supported micro-tubular SOFC stack. K. Power Sources 2005; 145: 428-434.
7. Nguyen T-L., Honda T., Kato T., Iimura Y., Kato K., Negishi A., Nozaki KShiono., M., Kobayashi A, Hosoda K., Cai Z., Dokiya M., Fabrication and characterization of anodesupported tubular SOFCs with zirconia-based electrolyte for reduced temperature operation. J. Electrochem. Soc. 2004; 151(8): A1230-A1235.J. Doe and R. Hill, *J. Electrochem. Soc.*, **137**, 1902 (1990).