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Build and Performance Test of a 3-cell Solid Oxide Fuel Cell Stack

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ABSTRACT

A 3-cell stacked anode-supported solid oxide fuel cell was designed and fabricated to achieve a complete gas seal and the facile stacking of components. The stack was assembled with a unit cell with 10×10 cm^2 area, and each cell was interconnected by a stainless steel 430 separator using a proprietary sealant sheet. The stack performance was examined at various gas flow rates of H_2+3.5 vol% H_2O, and air at a fixed temperature of 800°C. No gas leakage was found from the sealing between cells and interconnects within a measurement system in this research during a prolonged time of 500 h in operation. The test resulted in an open circuit voltage of 3.12 V, a peak power of 149 W, and a power density of 0.61 W/cm^2, while the long term durability of the power showed 19.1% degradation during the prolonged time of 500 h when tested at 800°C.

Key words ***: Solid Oxide Fuel Cell (SOFC), Metallic interconnects, Planar-type stack, Stack performance, Sealant***

1. Introduction

The solid oxide fuel cell (SOFC) has received tremendous amounts of attention due to its potential in clean power generation systems. SOFCs are known to be associated with less pollution, and have higher efficiency than any other type of fuel cell. Particularly, the planar-type design offers greater possibilities for high power density and cost-effectiveness compared with the tubular type. However, the planar-type SOFC has not yet shown reliability in long-term operations, and is thus limited to several thousands of hours in contrast to tubular SOFCs, which have been reported to endure tens of thousands of hours.[1-4] The reliability and cost reduction considerations of a SOFC system have been the key issues of current research from the view point of commercialization, especially as related to the electrochemical long-term stability of the stack.[5-9]

In the planar-type SOFC, two unit cell models are employed currently: the electrolyte-supported cell and the electrode-supported cell. The electrolyte-supported cell involves a somewhat simple fabrication procedure; however, the higher ohmic resistance due to its thick layer of electrolyte ranging over 100~200 μm requires a high operating temperature of approximately 1,000°C.[10,11] Among all electrode-supported cells, the anode-supported type exhibits superior electrical advantages compared with the electrolyte-supported type, as the lower ohmic loss in the thin electrolyte layer can induce a reduction of the operating temperature. The anode-supported cell with an over-coated electrolyte layer of approximately 10 μm was reported to lower the operation temperature to below 800°C,[12,13] which provides a realistic level for use with metallic interconnects in the stack. Employing metal interconnects in stack enables the SOFC to operate at a low cost, with the freedom of different design choices. Owing to the above advantages of the anode-supported SOFC, many companies such as FZJ, Delphi/PNNL (Battelle), GE, FCE and Tokyo Gas have commenced development of prototype multi-stacks and systems for anode-supported planar SOFCs employing metallic interconnects.[14-17]

Essentially, SOFC performance with an anode-supported cell depends on the thickness and material of the electrolyte, the porous state of the anode substrate, and the electrochemical activity of the cathode. Consequentially, in order to improve the cell performance, a reduction of the thickness of the electrolyte layer is essential.

The present study was carried out in terms of building a multi-cell stack without gas leakage combining with sufficient electrical contact and the facile stacking of components in a full scale. A stack composed of three anode-supported cells with a thin electrolyte of 5 μm was assembled employing metallic interconnects using an appropriate sealant, and the performance was characterized over 500 h at 800°C.

2. Experimental Procedure

The anode-supported cells were purchased and used. The cell has a dense electrolyte layer that was 5 μm thin, which allows the cell to operate at an intermediate temperature in

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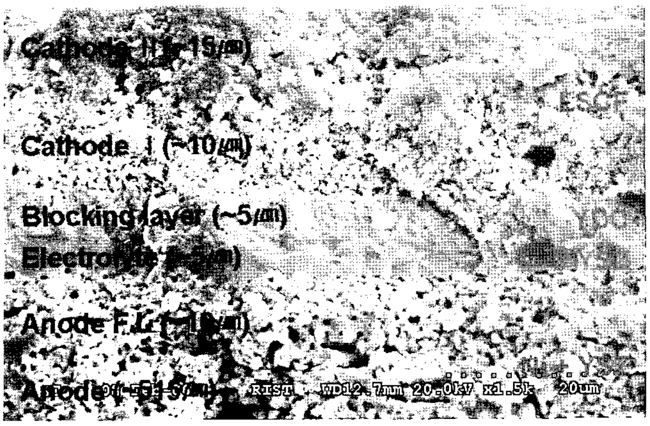


Fig. 1. Microstructure showing a cross-section of the unit cell used.

a range of 750 - 850°C. The appropriate interconnect and the sealant used to assemble the 3-cell stack were developed in house at RIST. The developed interconnect was designed from a stainless steel (STS) 430 and was composed of bipolar/end-plate that was machined to have a cross flow of gases for complete sealing of every stacking component. Sealant was formed by open molding casting into the shape of a manifold sheet with a thickness of 1.5 mm, for which slurry was prepared by mixing glass powder with an optimized organic binder. The stack was composed of three cells 10×10 cm^2 in size with a 9×9 cm^2 active area, and the assembly was done by placing each cell in a central position on the sealant sheet separated via sandwiching of metallic interconnects one by one.

To evaluate the developed 3-cell stack, a testing system that involved temperature controls, mechanical loading, gas supplies, and the acquisitioning of data was fabricated. In addition, an electronic load used for measuring and dissipating the power generated by cells was necessary. To intensify the cohesion force between the cell and the interconnect, a pneumatic piston was used to press the stack. Output power was obtained by a current density-voltage (I-V) curve as a function of the fuel gas flow at 800°C. Air as the oxidant and H_2 + 3.5 vol% H_2O gas as the fuel were used. The long-term durability of the stack was evaluated by measuring the degradation of the power during an operation time greater than 500 h, and the I-V curve was then obtained after the run of 500 h.

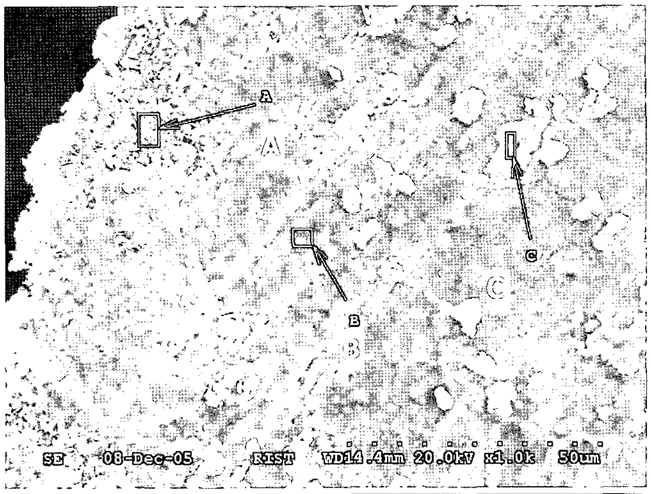
3. Results and Discussion

3.1. Fabrication of the Stack Components

Fig. 1 shows a SEM image of the cross-section of the cell termed ASC2 tested in this research. This cell was purchased from InDEC. Y_2O_3-stabilized ZrO_2(YSZ) electrolyte used appeared nearly pore-free and appeared to be well adhered to the NiO/YSZ anode. The total thickness of the used cell was approximately 560 μm, consisting of a 525 μm NiO/YSZ anode, a 5 μm electrolyte of YSZ, a 5 μm blocking layer of Y_2O_3-doped CeO_2 (YDC), and a 25 μm cathode of $La_{1-x}Sr_xCo_{1-x}Fe_xO_3$ (LSCF). In fact, the cathode consisted of two sub-layers that included one layer of 15 μm LSCF with a low pore density for improving the interfacial contact with the interconnect and another layer of more porous 10 μm LSCF. The YDC layer served to prevent the reaction between the cathode and the YSZ. It is known that a double-layer cathode has lower polarization resistance inside the cathode layer compared to a single-layer LSCF composite cathode. The anode layer was also multilayer, as it was intended to facilitate the electrochemical reaction with the electrolyte and the fuel gas.[18]

In order to maintain the structural stability, the sealant must maintain its chemical compatibility with the metal interconnect and must sustain a similar coefficient of thermal expansion (CTE). The CTE of metal is generally in the range of 11 ~ 13 ppm/K, which is higher than that of most basic glasses. The appropriate composite compound compatible with a metal interconnect consisting of BaO-SiO_2-B_2O_3-Al_2O_3-ZrO_2-based glass was investigated, and an organic binder was specifically prepared for the SOFC stacking. The sheet forming the sealant was always followed by permeance test by flowing N_2 gas at the operation temperature. The sealant sheet exhibited good adhesion to the interconnect in spite of the existence of a number of pores inside.

To minimize the degradation of the stack performance during the operation, the chemically stable anti-corrosive interconnect STS 430 was used. Fig. 2 shows the surface area of the sealant sheet after it was used for 500 h, in



	Fe	Si	Cr	Ba	O	Remains
A	-	9.8	50.2	4.4	26.8	8.8
B	-	20.0	23.7	11.3	33.6	11.4
C	53.2	4.3	9.5	6.0	24.0	3.0

Fig. 2. Surface mapping of elements obtained from the sealant sheet interfaced with the interconnect after the test operation.

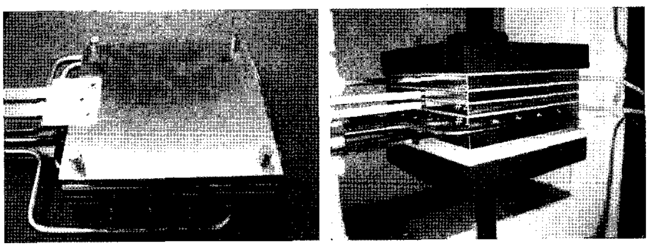
which the element Cr was detected at the three points A, B, and C. This likely resulted from Cr evaporation from the STS 430. Each point showed a relatively high content of Fe, which appears to have been an oxidized scale peeled from the STS 430 interconnect.

The interconnect in a planar SOFC plays two roles. First, it ensures the physical separation between the air and fuel gas, and second it provides an electrical connection adjoining the anode and cathode of the stack. Accordingly, the interconnect alloy is well known to have a significant effect on the long-term durability of the stack. However, metallic interconnects have two basic problems. The first is Cr evaporation leading to the decomposition of the cathode material, and the second is the formation of oxide scales resulting in significant ohmic losses. To minimize the above problems in this research, the cathode sides of the bipolar and end plate were coated with a $La_{0.85}Sr_{0.15}MnO_3$ (LSM) layer, which prevented the deterioration by Cr-evaporation from the STS 430 interconnect, as shown in Fig. 2, and also allowed improved electrical contact with the electrode. STS 430 plates of $15\times15\times1$ cm were trimmed to the dimension of $14\times14\times0.8$ cm and then machined to the final design of the cross flow and internal manifold. The final shape of the interconnect was confirmed to be perfect for a facile stacking and was found to be free from gas leakage.

3.2. Stack Assembly

A planar SOFC stack must be designed so that no gas leakage occurs. At the same time, a uniform distribution of fuel and air across the active area of the cell is desired. The present research proposes a cost-effective design based on a planar stack fabricated using anode supported cells interconnected via the STS 430 plates with a internal manifold type for high volumetric power density. Fig. 3 shows the stack developed in this research along with the test set-up.

In a test of the initial performance and the long term durability, the stack was sandwiched between an insulated ceramic and a mica plate, as shown in Fig. 3(b). It was then placed into a loading fixture composed of Inconel 600 plates in an electric furnace. An axial load of 3 kg/cm^2 was applied on the top of the stack during the testing. Although three configurations of gas flow for the planar SOFC, including the cross-flow, co-flow, and counter-flow configurations are known, the present design chose the cross-flow with the gas



(a) (b)

Fig. 3. Photographs of the tested (a) 3-cell stack and (b) set-up for the performance.

manifold of an integral configuration aiming for a low pressure drop caused by the uniform gas distribution. As the total flow rate was low although a difference in flow rate existed between the fuel H_2+3.5 vol% H_2O and oxidant air, the manifold was machined to have symmetrical and identical dimensions for the inlet and outlet of the gas. A sealant sheet was applied to ensure perfect tightness and to relieve the thermal stress caused by the coefficient mismatches of the thermal expansion between the individual components of the SOFC stack during the operation.

3.3. Performance Test

During the Open Circuit Voltage (OCV) test, a failure generally occurs as a result of gas leakage from inside to outside the stack and/or internal gas flow crossing over due to cracks or holes in electrolyte layer or at the interface between the sealant and the cell. Gas crossing over in the stack is generally more serious than gas leakage toward the outside. Once the gas crosses over, a significant drop in the

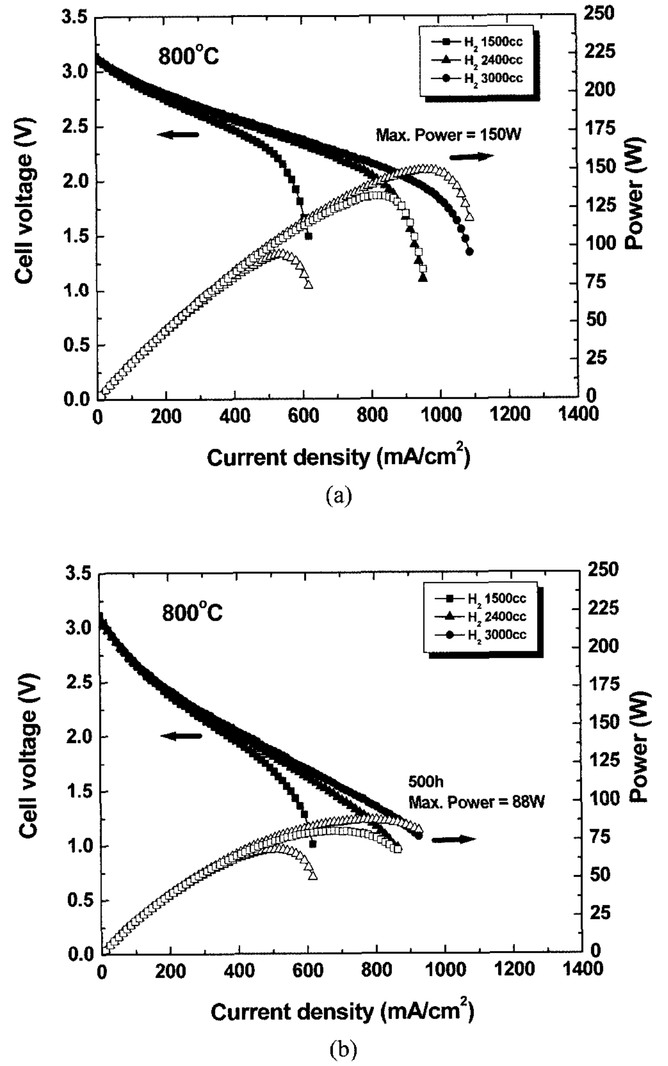


Fig. 4. I-V curves as a function of the air flow and H_2 fuel measured at 800°C during (a) the initial test and (b) after running for 500 h.

Table 1. Electrical Characteristics of the 3-cell Stack

H_2 Flow mass /cc·min^{-1}	Current density / mA·cm^{-2}		Voltage of 3 cell / V		Fuel utilization coefficients(U_f) / %		Electrical efficiency(η_{el}) / %		Max power / W	
	Initial	Final	Initial	Final	Initial	Final	Initial	Final	Initial	Final
1,500	543.2	506.2	2.15	1.67	67.0	62.4	38.4	27.8	94.6	68.5
2,400	827.2	679.0	1.98	1.47	63.8	52.4	33.7	20.5	132.7	80.6
3,000	975.3	765.4	1.89	1.42	60.2	47.2	30.3	17.8	149.3	88.0

OCV takes place, and the measured value is much lower than the calculated OCV at the same temperature. To check the gas leakage in the present study, two different tests were performed by monitoring the gas flow and initial OCV. Variation of the gas flow rate of H_2 and air was not found at either the inlet or the outlet. Moreover, the initial OCV of the 3-cell stack was 3.12 V, which is close to the theoretical OCV value. Therefore, gas leakage was not believed to have occurred in the stack in this study.

Basically, a current density-voltage curve, the I-V curve, reveals the performance of a stack in terms of the cell design, component arrangement and fabrication of the stack. If the stack is in a normal state, the maximum power density along the I-V curve varies greatly with increasing fuel flow rate and/or increasing the operation temperature. Accordingly, the I-V curves were measured at a constant operation temperature of 800°C by varying the H_2 flow rates of 1500, 2400, and 3000 cc/min with the corresponding air gas rates of 3750, 6000, and 7500 cc/min, respectively. Fig. 4(a) shows the I-V curves measured at the beginning of the test, and Fig. 4(b) illustrates the I-V behaviors after running 500 h in identical conditions. The maximum power generated at initial stage increases with increasing the gas flow rate from 94.6, 132.7 to 149.3 W, and the corresponding power density increases from 0.39, 0.55 to 0.61 W/cm^2, respectively. Peak power was obtained at 975.3 mA/cm^2, as shown in Fig. 4(a). After running for 500 h, the I-V behaviors were greatly deteriorated resulting in the power to decrease to 68.5, 80.6 and 88.0 W as shown in Fig. 4(b). To investigate the I-V behaviors of each cell composing the developed stack, the initial I-V curves were separated and plotted, as shown in Fig. 5. The first cell denoted by #1 in the figure was confirmed to exhibit superior power generation; it tends to decrease its generation by moving to the upper cell, as shown along the respective curves #2 and #3 in the figure. The decrease of the generating power appears to have resulted from the disequilibrium of the H_2 supply with moving to upper cell.

The utilization coefficients (U_f) of fuel H_2 were calculated as a function of the flow rate by the ratio of H_2 reacting in the stack and according to the H_2 input to the cell. The electrical efficiency (η_{el}) of the unit cell as shown in equation (1) below can be expressed as V_{cell}, U_f, and LHV, which are respectively the single cell voltage, the fuel utilization coefficient, and the Lower Heating Value.

$$\eta_{el} = U_f \frac{V_{cell}}{1.25} 100\% \qquad (1)$$

Table 1 presents the evaluated electrochemical properties of the developed stack at the initial stage before the durability test and the final stage after running for 500 h. By increasing the fuel H_2 flow rate to 1500, 2400, and 3000 cc/min, the electrical power clearly increased while η_{el} and U_f decreased gradually at the same time. U_f was measured in the range of 60~67% initially and at 47~62% after running for 500 h. η_{el}

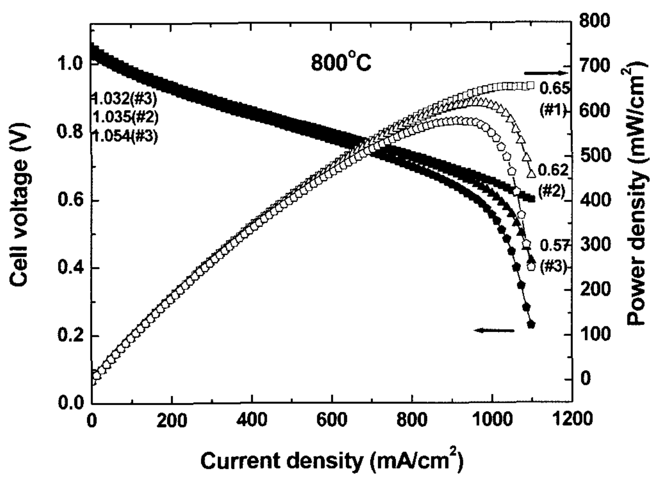


Fig. 5. I-V curves obtained from each cell in the 3-cell stack tested initially with H_2 flow of 3,000 cc/min.

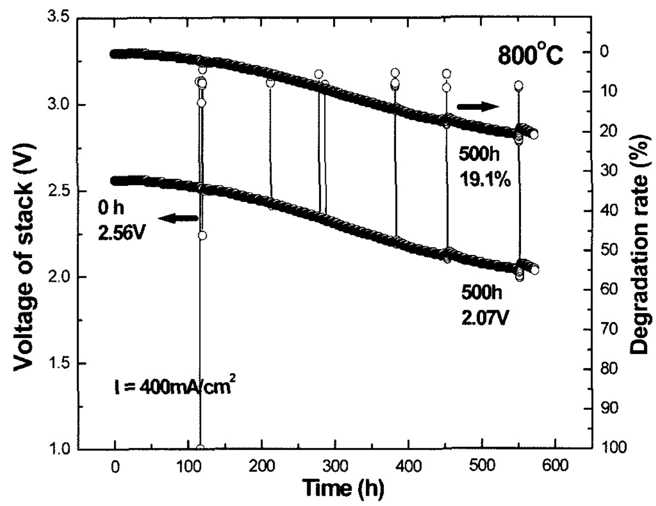


Fig. 6. Power durability of the developed stack prolonged for 500 h.

was calculated at 30~38% initially and at 18~28% after running for 500 h. A η_{el} of value 38% was obtained at the highest coefficient (U_f) of 67% during the initial test.

To examine the long-term durability of the stack, the test was prolonged for more than 500 h under a load of 400 mA/cm^2 at 800°C. The power degradation related to this test is presented in Fig. 6. During the operation up to 100 h, the power tended to decrease slowly. The degradation rate then accelerated past this time, and a power drop of 19.1% was determined at the end of the test. The corresponding I-V curve is shown in Fig. 4(b). As a result, the final powers corresponding to H_2 flow rates of 1500, 2400, and 3000 cc/min were obtained, respectively, as 72, 61, and 59 % of the initial power. This remarkable deterioration in the power must have been due to the increase of the contact resistance that took place at the interface between the electrodes and interconnect. Increased resistance can be caused by the diminution of the contact area at the interface due to the deformation of the machined metallic interconnect at the operation temperature as well as by decomposition of component materials such as the cathode and the interconnect.

4. Conclusions

A proprietary metallic interconnect and appropriate sealant were developed for the test of a planar-type SOFC composed of three cells with an active area of 9×9 cm^2. The cells used in this study were anode-supported type, and the interconnect was machined to have an internal manifold that allowed the gas to flow cross-directionally. The interface of the interconnect facing the cathode layer was coated with LSM to prevent Cr evaporation and to enhance the electrical contact, leading to high efficiency.

A performance test of the stack was carried out at 800°C as function of the fuel gas flow of H_2 and air. Long-term durability was evaluated by measuring the power degradation while running over 500 h. The open circuit voltage (OCV) revealed that no gas leakage took place in the proposed measurement system, and that the OCV of the 3-cell stack measured 3.12 V. The peak power generated and power density of the stack were 149.3 W and 0.61 W/cm^2, respectively, at the initial stage when the flow rate of H_2 was 3,000 cc/min and that of air was 7,500 cc/min. Under those conditions, the maximum U_f and η_{el} values were 67% and 38%, respectively. The degradation rate after 500 h of operation was found to be nearly 19.1%, which appears slightly higher than earlier results. It is believed that this resulted from the deformation of metallic interconnect that took place during the prolonged test at a high temperature. In addition, an electrical insulating oxidation of the interconnect was suggested by surface analysis of the stack components after the tests. Conclusively, a more careful design must be developed in order to prevent any deformations of the metallic interconnect while avoiding oxidation in an effort to maintain a perfect contact interface.

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