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Development of envelope-type solid oxide fuel cell stacks

H. Yoshida*, H. Yakabe, K. Ogasawara, T. Sakurai

Technical Research Institute, Tokyo Gas Co., Ltd., 16–25, Shibaura, 1-chome, Minato-ku, Tokyo 105-0023, Japan

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Abstract

Anode-supported planar solid oxide fuel cells (SOFCs) have been developed at Tokyo Gas. Recently, to achieve a tight gas seals and sufficient electrical contacts between each cell, a new method for stacking the cells has been evolved. The former system was assembled by stacking planar cells and rigid planar interconnectors alternately. This study reports a new system, in which constituent units with an envelope configuration are piled vertically. Each cell is wrapped in a thin electrically conductive alloy and connected to the next cell through a flexible metal interconnector. Thus, the assembly is called an envelope-type SOFC stack. By using a thin alloy foil and a flexible metal interconnector, tight gas seals and sufficient electrical contact between each cell are achieved. Tests are conducted on 7- and 20-cell stacks and an improvement in performance is obtained.

Keywords: Alloy envelope; Flexible metal interconnector; Cell warp; Stacking; Planer solid oxide fuel cell; Performance improvement

1. Introduction

Planar solid oxide fuel cells have been developed at Tokyo Gas since 1989. Initially, electrolyte-supported cells were developed, but in 1998 the main development was shifted to anode-supported cells. The main challenge was how to stack the cells with a tight gas seals and sufficient electrical contacts. Recently, the method for stacking cells has been improved in an envelope-type stack, and the performance of this stack has been tested. This paper reports the history of development of planar SOFCs at Tokyo Gas and the latest progress on the envelope-type SOFC stack.

As mentioned above, initial work commenced on electrolyte-supported cells stacks were fabricated using $LaCrO_3$ interconnectors and $MgAl_2O_4$ frames. A schematic representation of the configuration is given in Fig. 1. In 1998, 1.7 kW of power were obtained from a pair of 48-cell stacks (shown in Fig. 2) that operated on methane reformed gas at 1000 °C. Although this was the world's first test of a several kW system with a directly-internal methane-reforming anode, the stacks were immediately damaged and destroyed. The main reason for the destruction of the stack was a lack of mechanical reliability and thermal durability of the electrolyte-supported cells under operation at the high temperature. A test of the durability of a 10-cell stack against thermal stress was conducted. The stack was operated in an electric furnace at 1000 °C and internal heating was generated by an increase in the current flow. With an internal heat of 0.24 W cm^{-2}, the stack was destroyed, as shown in Fig. 3. The test conditions were simulated using a computer calculation; the simulated temperature and corresponding stress distribution in the electrolyte are shown in Figs. 4 and 5, respectively. The temperature of the cell located in the middle part of the stack rose to more than 1400 K. The thermal stress at the edge part of the cell became 215 MPa, which was in excess of the strength of the electrolyte, and thus the cell was damaged [1]. The tests revealed that the electrolyte-supported cell was very weak against thermal stresses and had to operated carefully.

In 1998, SOFC development was shifted from the electrolyte-supported to the anode-supported design. The latter has certain advantages compared with the electrolyte-supported type, notably it is markedly resilient to thermal stresses. A compressive residual stress remains in the electrolyte part of the cell, and it can lower the thermal tensile stress caused by the temperature distribution during operation. Accordingly, the anode-supported cell can withstand thermal stresses under practical operation. Another advantage is that the lower ohmic loss in the thin electrolyte can reduce the operating temperature. As a result, metallic components can be used to assemble the stack. In the early stages of development, an electrical conversion efficiency of more than

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* Corresponding author. Tel.: +81 3 5484 4284; fax: +81 3 3453 2674.
E-mail address: yhideki@tokyo-gas.co.jp (H. Yoshida).

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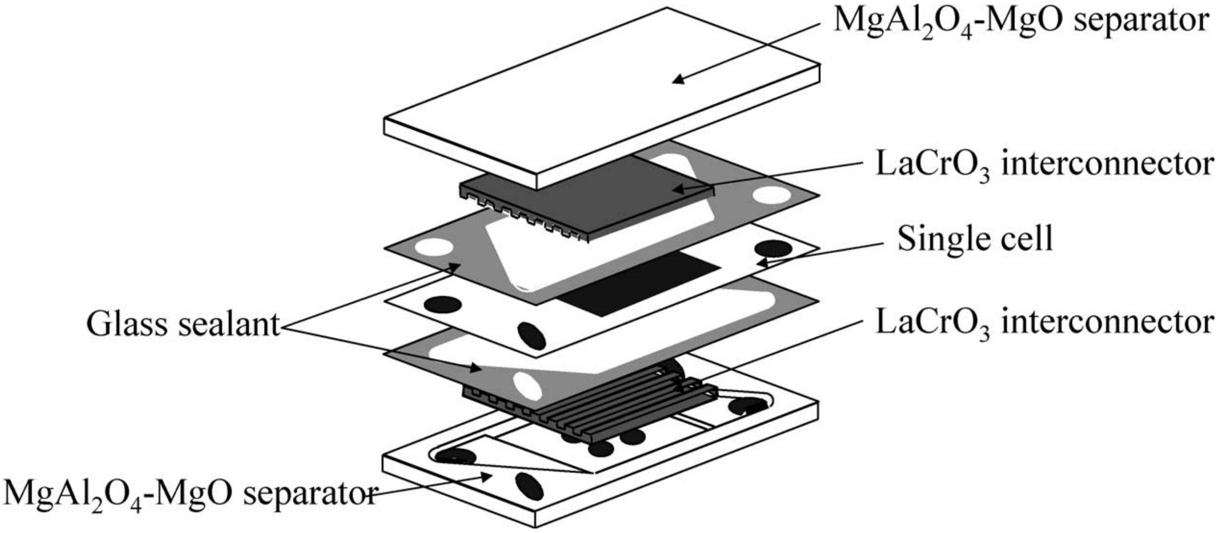


Fig. 1. Schematic illustration of stack structure for electrolyte-supported type SOFC, using $LaCrO_3$ interconnectors and $MgAl_2O_4$ frames.

50% (higher heating value) at around 750 °C was obtained with a small singl-cell stack [2].

Next, an attempt was made to scale up the cell from 5 × 5 cm to 13 × 13 cm and construct a stack. Unfortunately, the performance of the enlarged cell was extremely low. The performance of the 5 × 5 cm and 13 × 13 cm cells are compared in Fig. 6. Increase in cell size induces a new problem, namely, warping. The origin of cell warp is the difference in the thermal expansion coefficients (TECs) between the anode and the electrolyte. Morever, it is difficult to obtain sufficient electrical contacts between extremely warped cells. It is also difficult to achieve a tight seal of the air and the fuel at the edges of the cell, and thus, cross-leaks occur. Two methods were investigated to solve these problems: (i) modification of the cell fabricating process to reduce the magnitude of warping; (ii) employing a new stack design, i.e., the envelope-type stack.

Fig. 2. Photograph of a pair of 48-cell stacks for an electrolyte-supported type SOFC.

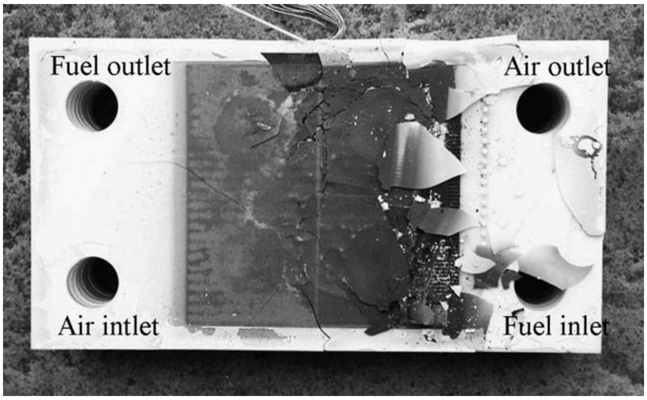


Fig. 3. Photograph of destroyed 10-cell stack after durability test.

2. Experimental

Anode-supported cells were fabricated by co-firing both the electrolyte and the anode; details of the fabrication process are described elsewhere [3]. YSZ slurry was dip-coated on a pressed green substrate of NiO/YSZ and then both were co-fired. A cathode was screen printed on the electrolyte and fired. The single cells were scaled up to 13 × 13 cm, but the enlargement caused a warping of the cell, as diplayed in Fig. 7(a). The large size cell warped convexly to the electrolyte side by more than 2 mm. The thermal expansion coefficients (TECs) of NiO/YSZ and YSZ do not match well, and this mismatch induces warping and a large compressive stress of several hundreds MPa in the electrolyte part [4]. Since the stack performance becomes extremely low, it is essential to reduce this warp to obtain a high performance. Accordingly, a two-step co-firing process was adopted for the anode–electrolyte part. First, the green substrate of the anode was pre-fired and, second, the YSZ slurry was coated and the

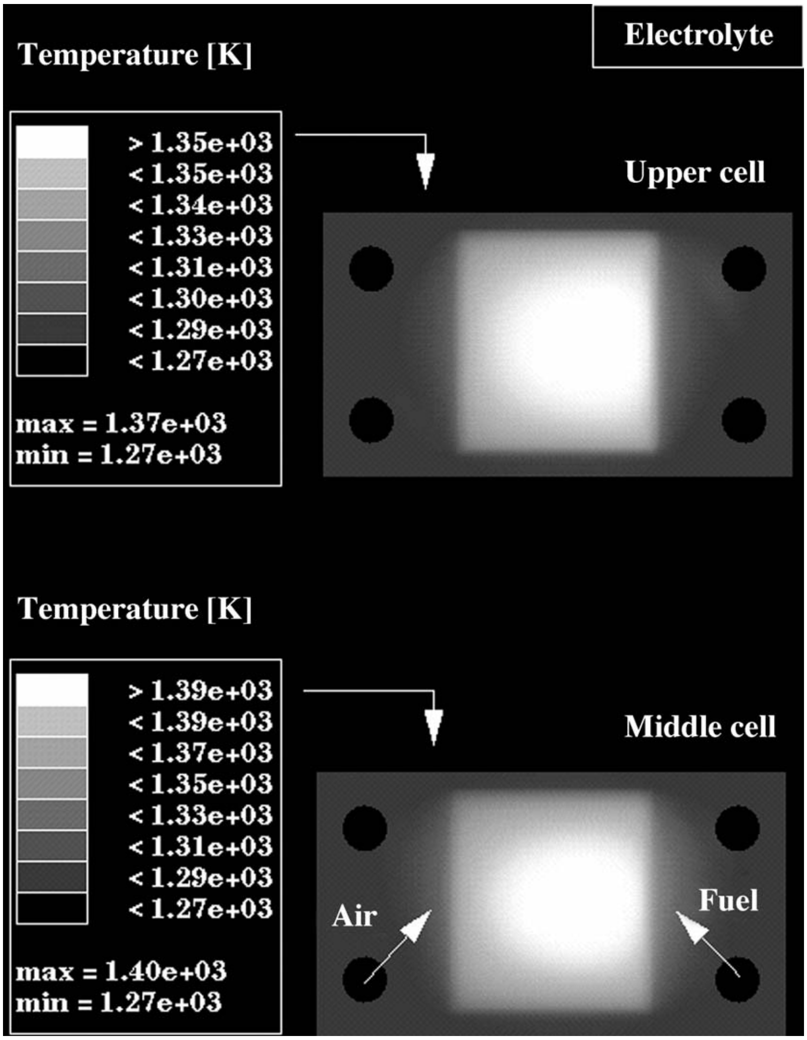


Fig. 4. Simulated temperature distribution in electrolyte of cell located at the upper part and the middle part of 10-cell stack.

product was co-sintered. With this modification, the warp was reduced to less than 1.5 mm, as illustrated in Fig. 7(b).

Cross-sectional and the schematic illustrations of the stack structure developed in the early stages of the research programme are given in Fig. 8(a and b), respectively. The single cell was held to an alloy manifold using thin and flexible alloy support foil to mitigate thermal stress. The manifold and the cell were sandwiched by rigid planar interconnectors which had a grooved structure for the gas channels. As displayed in Fig. 8(b), the fuel gas enters the stack from the left internal manifold, passes over the grooved structure on the inteconnector of the anode side, and exits through the right internal manifold. Similar to the fuel channel, the air comes in the front internal manifold, passes over the cathode, and goes out through the back side. All the components, except the cell, were made of an alloy based on a ferritic stainless-steel. Glass was used to seal the cell and alloy foil, and mica sheets for the other parts to obtain a tight seal of gases and electrical isolation. A serious issue for this stack was that if the single cell warped greatly to the electrolyte side, the contacting area was small, and thus the contacting resistance was increased.

Recently, a new stack design was employed to decrease the contact resistance and obtain a tight gas seal, simultaneously. Cross-sectional and schematic illustrations of the envelope type stack structure are given in Fig. 9(a and b), respectively. In this new stack, a single cell is held to an alloy manifold through a thin alloy support foil as well as the preceding stack, and the new characteristic is that the anode side of the cell is covered with the same alloy foil. The interconnecter is also made of an alloy foil with a corrugated shape and inserted between each unit. By covering the anode side with a foil, the parts to be sealed are decreased and an excellent gas seal is achieved. In addition, by using a flexible alloy foil as the interconnector, the contact area between the cathode and the interconnector can be increased and the contact resistance decreased. In addition to these advantages, the use of thin foil can greatly reduce the weight of the stack. Similar to the former stack, the fuel gas comes in from the left internal manifold and goes out through the right one, but the flow channel of the air is considerably different. In the new stack, the air is blown in directly from the stack side.

An evaluation has been made of the difference in the contact area between the old and the new stack designs. The

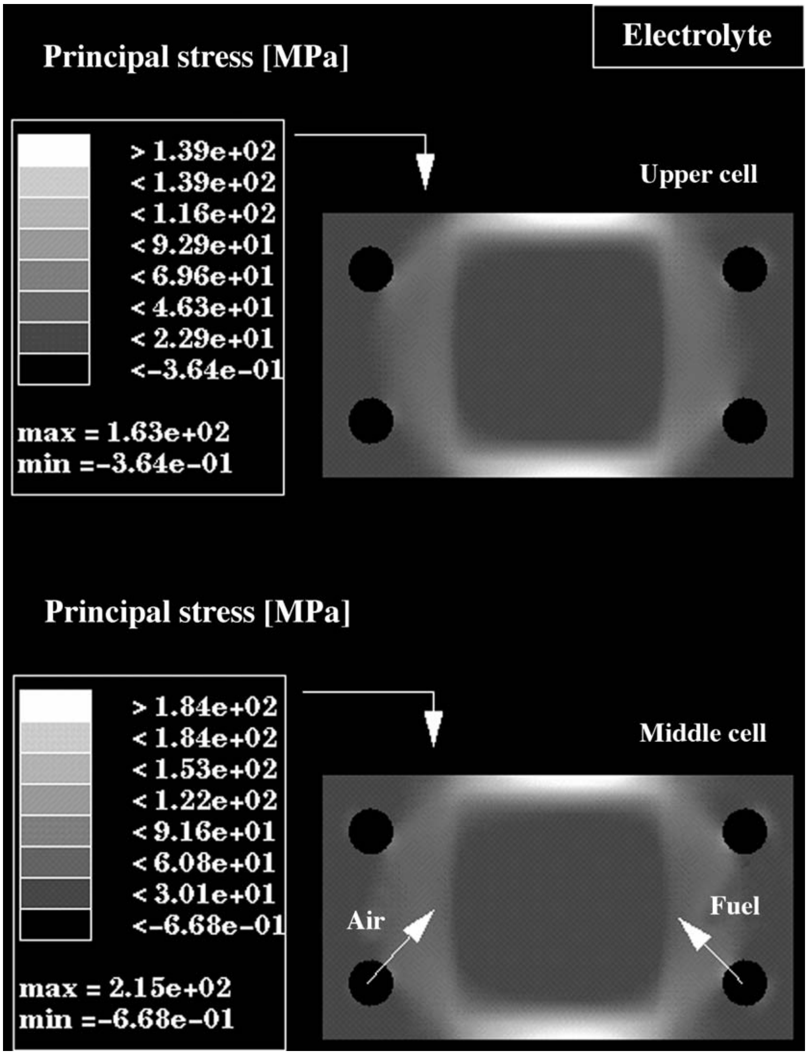


Fig. 5. Simulated stress distribution in electrolyte of cell located at upper part and middle part of 10-cell stack.

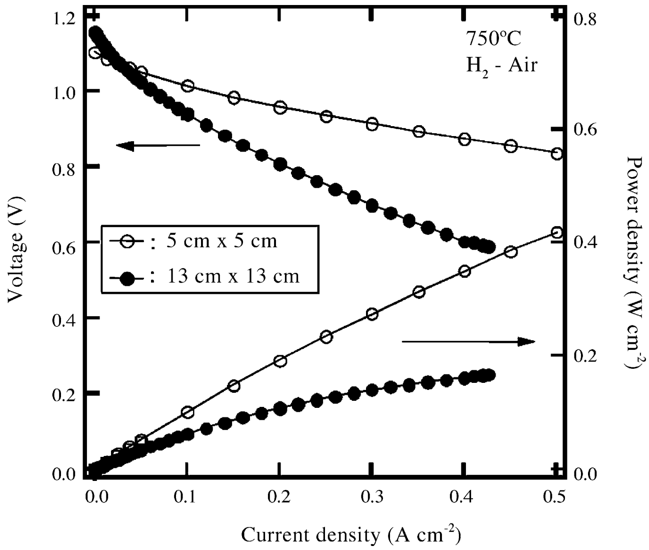


Fig. 6. Comparison of performance of single-cell stacks for 5 × 5 cm and 13 × 13 cm cells at 750 °C with hydrogen fuel.

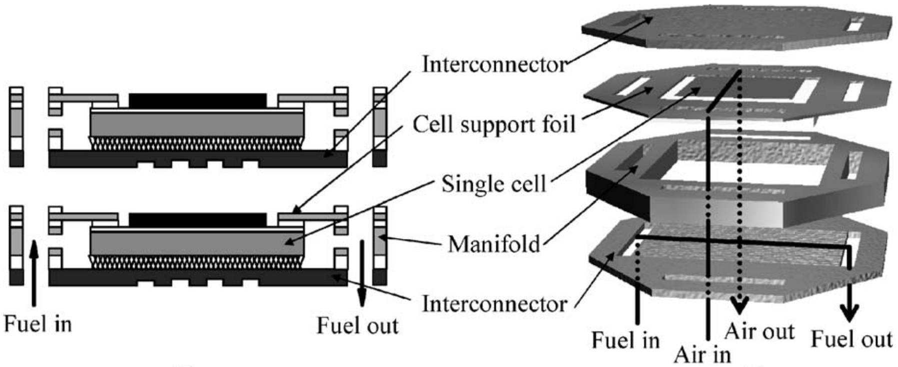
configuration in the test is presented in Fig. 10. A warped cell is sandwiched between two rigid plates and a pressure sensor sheet is inserted between the cathode side of the cell and the plate (Fig. 10(a)). A load was put on the upper plate and the change in contact area was measured for an increasing load. In this test, the solid plate played the same role as the rigid planar interconnector of the old type of stack. The same test for the new type of stack was also carried out. Two unit-stacks of the envelope-type were sandwiched between two rigid plates and a pressure sensor sheet was inserted between one of the cathodes and the interconnector, see Fig. 10(b).

3. Results and discussion

From the load test, it is possible to evaluate the change in the contact area in a new design stack compared with the old one. A comparison of the contact area in the two test patterns mentioned above in Section 2 is shown in Fig. 11. The upper figures correspond to the results of the test for the old stack. The pressure concentrates only in the central part of the cell under a load of 50 N. With increasing the load, the contact area

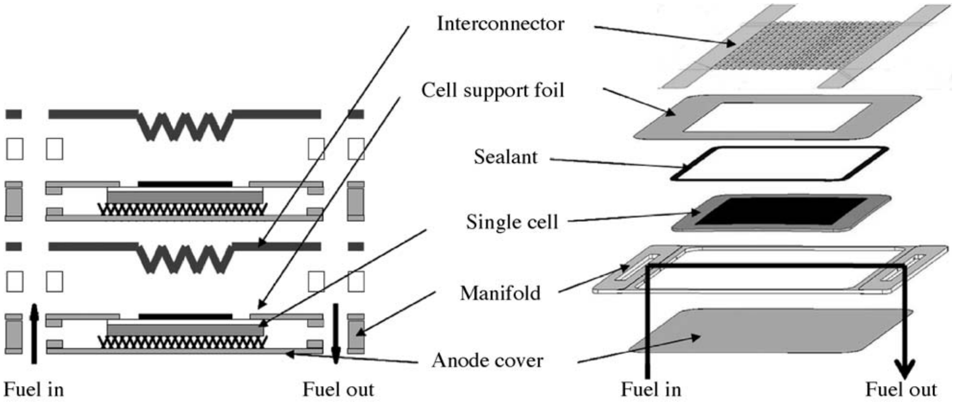
(a) (b)

Fig. 7. Appearance of 13 × 13 cm single cells: (a) before modification; (b) after modification of fabrication process.



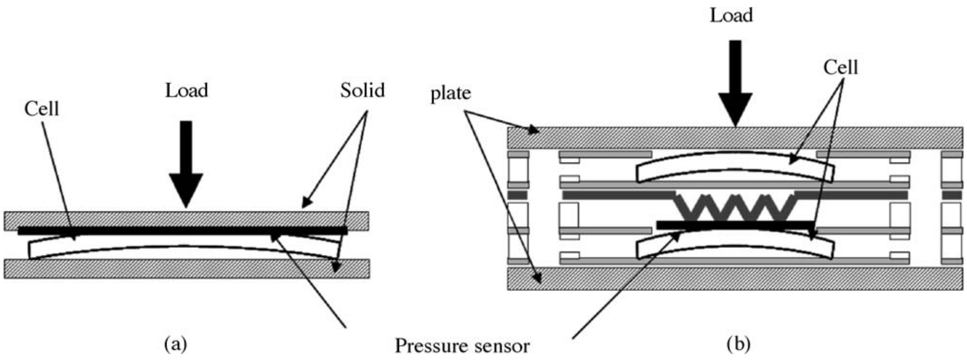
(a) (b)

Fig. 8. (a) Cross-sectional and (b) schematic illustrations of anode-supported type SOFC stack structure.



(a) (b)

Fig. 9. (a) Cross-sectional and (b) schematic illustrations of envelope-type stack structure.



(a) (b)

Fig. 10. Configuration of load test: (a) warped cell and pressure sensor sheet are inserted between plates; (b) two unit-stacks of envelope type and pressure sensor sheet are inserted between plates.

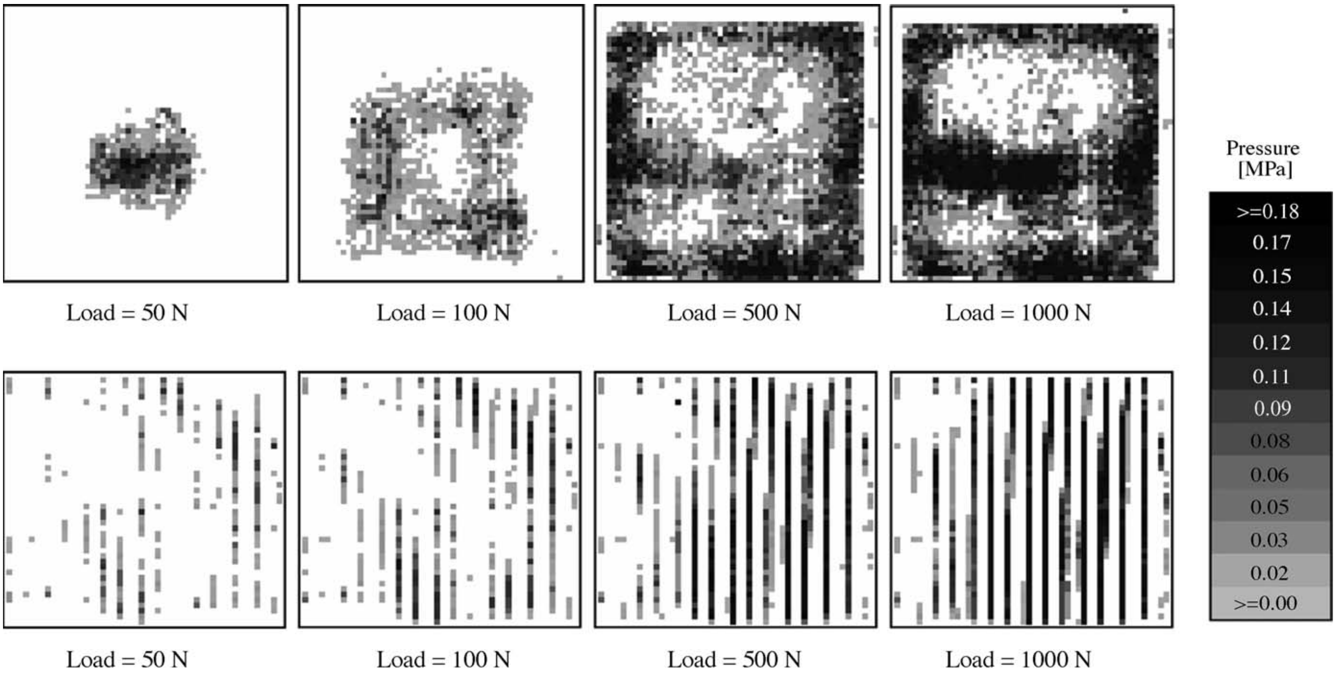


Fig. 11. Comparison of contact area between two test patterns. Upper and lower images correspond are for the old and new stacks, respectively.

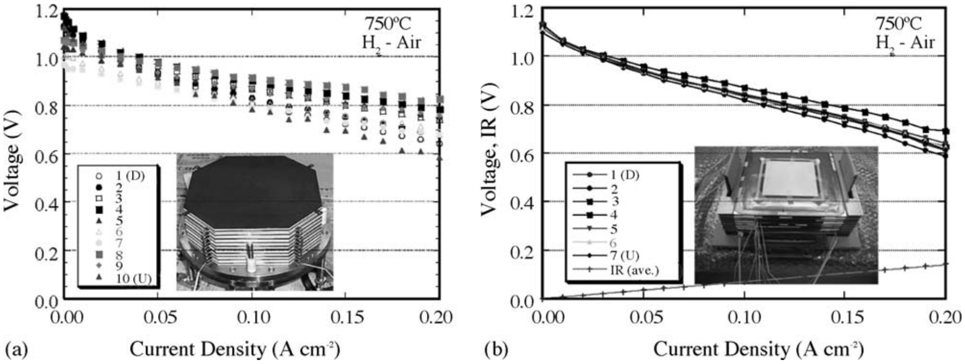


Fig. 12. Current–voltage characteristics of (a) 10-cell stack of old design and (b) 7-cell stack of new design at 750 °C with a hydrogen fuel. Terminal voltage measured for each cell. The cell number shows the order from the bottom of the stack.

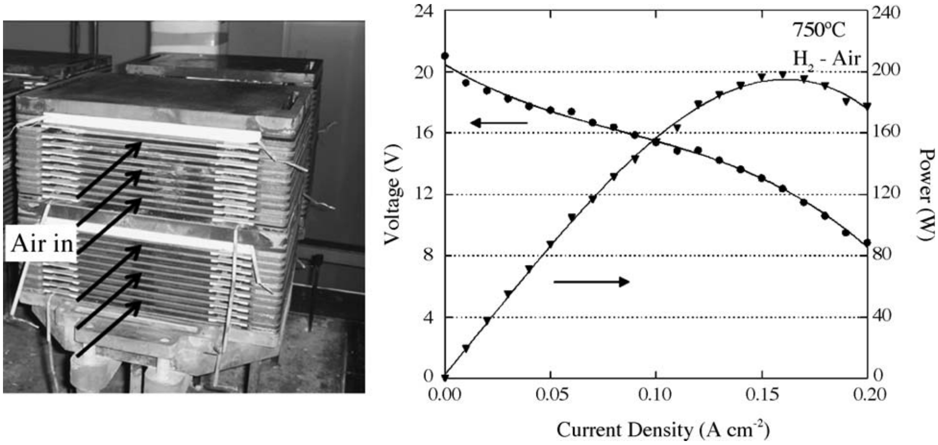


Fig. 13. (a) Photograph of 20-cell envelope-type stack and (b) current–voltage characteristic at 750 °C with hydrogen fuel.

spreads [5]. This test demonstrates clearly that the contact area is small under a low load, in other words, only the top area of the cathode could contact with the flat interconnector. Accordingly, the ohmic loss would be large for the old type stack. The lower images in Fig. 11 are the results obtained from the new design of stack. Reflecting the shape of the interconnector, the contact area with the cathode takes the form of a stripe. In this case, the contact area spreads over the cathode even under a low load, and thus, a low contact resistance between the cathode and the interconnector is expected for the new stack.

Using the new envelope-type units and the flexible interconnectors, a 7-cell stack was assembled and its performance evaluated. The result is compared with that of a 10-cell stack of the old design in Fig. 12. For the old design stack, the cells with a small warp, i.e., less than 1.5 mm, were selected to obtain a high performance. On the other hand, for the new design stack, cells were not selected and thus had various degrees of warping. In spite of the disadvantage of the large warp, the new design of stack gave a high performance compared with the old design. The individual performances of the cells were almost uniform although those of the old designed were unequal. The reason for the observation with the new stack is that the contact between the cathode and the interconnector is excellent even when using extremely warped cells. Thus, the advantage of the new design of stack compared with the old design.

Recently, a 20-cell stack of the envelope type has been fabricated and subjected to thermal-cycle testing and measurement of the current–voltage (*I*–*V*) characteristics. The *I*–*V* characteristics were investigated at 750 °C with hydrogen fuel; the results are shown in Fig. 13. Although around 200 W of power could be obtained, this performance per layer was lower than that of the 7-cell stack. The most likely reason for the lower performance for the 20-cell stack is an unequal distribution of the supplied air. As mentioned above, air is blown into the stack directly from the stack side in the new design of stack. As a result, most of the introduced air will not flow inside the stack, but pass through the outside of the stack. The *I*–*V* plot shows a falling curve, which suggests a concentration overvoltage at a high current density. This drop in voltage may be due to a depletion of air at the cathode owing to the geometry for the air supply. A sufficient air supply to the cathode is a novel feature of for the new design stack.

In the thermal-cycle test, the stack temperature was varied from room temperature to 750 °C, at a heating rate of 200 $^{\circ}\mathrm{C\,h^{-1}}$, for five times. After the test, there are no cracks in the cells in all layers of the stack and no falls in open circuit voltage (OCV) [6]. These observations confirm the durability of the new design of stack against thermal-cycle operation.

4. Conclusions

The performance of an anode-supported planar SOFC has been improved through: (i) a change in the cell fabrication process to reduce cell warping; (ii) change in the stack design to obtain sufficient electric contact between each cell. By measuring the contact pressure between the cathode and the interconnector, it is confirmed that the contact between them spreads over the whole cathode. A 20-cell stack delivers 200 W at 750 °C and withstand thermal cycling.

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