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Short communication

Evaluation of micro flat-tube solid-oxide fuel cell modules using simple gas heating apparatus

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HIGHLIGHTS

- Cost effective 1 mm thick micro flat-tube SOFCs were fabricated.
- Five and ten cell modules of the micro flat-tube SOFCs were evaluated.
- Simple gas heaters were utilized for the SOFC module operation.
- The testing unit realized quick startup operation of 20 min.

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ABSTRACT

Micro flat-tube solid-oxide fuel cell (SOFC) modules consisting of 1 mm thick, 1.2 cm wide micro flat-tube SOFCs, gas manifold, and insulator have been fabricated and evaluated using simple gas heating apparatus. The cell consists of NiO – yttria stabilized zirconia (YSZ) as an anode (flat-tube support), scandia stabilized zirconia (ScSZ) as an electrolyte, gadolinia doped ceria (GDC) for an interlayer, and (La, Sr)(Fe, Co)O_3 (LSCF) – GDC as a cathode, which has been fabricated using cost effective extrusion technique and dip-coating technique. The cell has been investigated between 600 and 650 °C operating temperature and showed the power density at 0.75 V of 0.19 and 0.385 W cm^{-2}, respectively. Using the cell, a five and ten-series modules were assembled and stored in insulator with small gas heaters powered by a 24 V power source for start-up. The module successfully operated using hydrogen and methane fuel.

1. Introduction

Fuel cells have been intensively studied in recent years as a key technology of the future energy sources. Among various type of fuel cells, solid-oxide fuel cells (SOFCs), operated at high temperatures of 700–1000 °C, have been shown to be the most effective fuel cells due to their highest energy conversion efficiency [1–7]. In addition, SOFCs allow using direct hydrocarbon fuel by selecting appropriate anode materials, and thus they are more suitable for variety of application use [8–11]. Up to date, in order to meet the demand of increasing the life-time of the cell/stack/module for lowering the cost of the system, many studies have been focused on lowering the operating temperature of the SOFCs by developing new electrode and electrolyte materials [12–22] as well as the structures of the cell/stack/module. Enabling quick startup/shutdown operation is another key issue for broadening the application of the SOFCs, and micro-tubular type cells, which were proved to be ideal shape for quick-startup, have been well investigated [23–28].

In this study, we report newly developed 1 mm thick micro flat-tube SOFCs using the conventional Ni-zirconia based anode and zirconia based electrolyte. Micro flat-tube SOFCs are expected to have desirable characteristics which planar and micro-tubular SOFCs have, larger electrode area and higher durability for quick heating. Simple testing apparatus for its module has also been designed using simple small gas heaters powered by a 24 V power source for quick start-up, and the modules were evaluated using hydrogen fuel and methane + air mixed fuel.

2. Experimental

2.1. Single cell fabrication and evaluation

The micro flat-tube SOFCs developed in this study consisted of an anode (NiO – 8 mol% yttria stabilized zirconia (YSZ) as a flat-tube type support), an anode functional layer (NiO – YSZ), an

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electrolyte (10 mol% Scandia stabilized zirconia (ScSZ)), an interlayer (10 mol% gadolinium doped ceria (GDC)), and a cathode ($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-y}$ (LSCF) − GDC).

Anode micro flat-tubes of 1 x 16 channels were made from NiO (Sumitomo Metal Mining Co., Ltd.), YSZ (Tosoh Co., Ltd.), poly methyl methacrylate beads (PMMA) (Sekisui Plastics Co., Ltd.), and cellulose (Yuken Kogyo Co., Ltd.). These powders were mixed for 1 h, and after adding the correct amount of water, were stirred for 30 min in a vacuumed chamber. The mixture that was prepared from these powders was left over 15 h for aging. The flat-tubes were extruded, using the aforementioned extrudate, from a metal mold (1.2 mm thick, 15 mm wide with 0.8 mm square 16 pins) by using a piston cylinder type extruder (Ishikawa-Toki Tekko-sho Co., Ltd.). A slurry for dip-coating the anode functional layer was prepared by mixing the YSZ and NiO powders, solvents (toluene and ethanol), binder (poly vinyl butyral), dispersant (polymer of an amine system) and plasticizer (dioctyl phthalate) for 24 h. The same procedure was applied for preparing the electrolyte using the ScSZ powder (Daiichiki-genso Co., Ltd.), the interlayer and the cathode slurries. The anode tubes were first dipped in the anode functional layer slurry and then, in the electrolyte slurry after drying the first coated layer in air. The coated films were dried and co-sintered at 1350 °C for 1 h in air. An interlayer slurry of the GDC powder (Shinetsu kagaku, Co., Ltd.) was dip-coated on the electrolyte layer of the flat-tube and sintered at 1200 °C. A cathode slurry of LSCF and the GDC powder (LSCF-GDC) were dip-coated on the interlayer. The SOFCs were completed by sintering at 1050 °C. Appearance of the cell is shown in Fig. 1. As can be seen, the fabrication process developed in this study allows mass-production since it only requires simple cost effective extrusion and dip-coating techniques and multiple sintering processes. The length of the cell can be determined depending upon the size and design of the stack/module, but technically the length can be limited by the size of the furnace for sintering process. The width and thickness of the cell are 12 mm and 1 mm, respectively and it can provide the electrode area of 2.8 cm^2 per 1 cm cell length, which has the volume of 0.12 cc per 1 cm cell length. As can be seen, equal thickness of 0.25 mm anode wall was successfully constructed with sixteen fuel gas channels of 0.25 × 0.25 mm^2. A crack-free dense electrolyte with a thickness of about 10 μm has prepared on the anode functional layer by co-sintering method. This developed fabrication process also allows controlling of the anode microstructure for the improvement of the cell performance [29–31].

The electrochemical performance of the single cell was investigated by using a potentiostat (Solartron 1296) and impedance analyzer. The size of the cell was 1 mm thick, 1.2 cm wide and 40 mm in length with cathode length of 10 mm, and an effective electrode area of 2.4 cm^2. Ag wire was used for collecting current from the anode and cathode sides, which were both fixed using Ag paste. The current collection from the anode side was made from an edge of the flat-tube, and the collection from the cathode side was made from the whole cathode area. Diluted hydrogen (20% H_2 in Ar) was flowed inside of the cell at a flow rate of 47–140 mL min^{-1} (H_2 flow rate: 3.8–11 mL min^{-1} per electrode area of 1 cm^2). Air was supplied at the cathode side at a flow rate of 100 mL min^{-1}.

2.2. Module and gas heater apparatus

Fig. 2a and b shows images of the cell and the five-cell module, and the ten-cell module using micro flat-tube SOFCs (cathode length: 1.5 cm, electrode area: 3.6 cm^2 per cell). The cells are integrated into metal (2a) or ceramic (2b) gas manifolds with a seal glass. Ag wire was used as interconnect fixed to the electrodes with Ag paste.

Fig. 3a shows the chart of the experimental apparatus including gas heaters and power unit, and a DC–DC converter for demonstration of LED light-up. It consists of the module that are stored in a ceramic insulator (maximum wall thick = 3 cm), a 100 W platinum gas heaters (SAH24V-100W, Fintech, Co., Ltd. Kobe, Japan) (Fig. 3b) with a 24 V power source. Advantages of use of the gas heater are that the gas temperature is easily controlled by changing applied voltage and it can be operable using battery, for application use. Note that there is a variety of gas heaters available depending up on the size of stack/modules. Fig. 3c shows the closer look of the mounted module in an insulator with thermocouples for monitoring the temperature of fuel gas at the cell outlet ($T_{fuel,output}$) and the temperature of module at the cathode of the cells ($T_{cell,cathode}$). Fuel and air are supplied to the module after heated by the gas heaters. The modules were tested using diluted hydrogen (20% H_2 in Ar) at the flow rate of 3 or 5 L min^{-1} and diluted methane mixed with the air for partial oxidation (10% methane in Ar: 3.3 L min^{-1} + air: 1 L min^{-1}). Air was feed to the module with the flow rate of 3 or 5 L min^{-1} Fig. 3d shows the appearance of module testing, showing that the surface temperature of the insulator was just room-temperature, while inside the insulator was about 600–800 °C.

3. Results and discussion

3.1. Single cell performance

Performance of the single cell obtained at various gas flow rates was shown in Fig. 4 for (a) 600 and (b) 650 °C operating

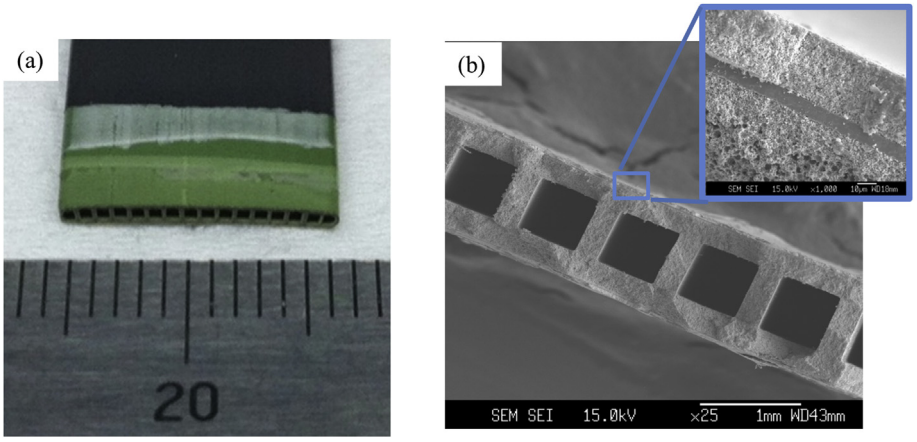

Fig. 1. (a) Image of the micro flat-tube SOFC (b) Cross-sectional SEM images of the micro flat-tube SOFC.