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Fabrication and operating characteristics of a flat tubular segmented-in-series solid oxide fuel cell unit bundle

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ABSTRACT

A unit bundle of a flat tubular segmented-in-series (SIS)-solid oxide fuel cell (SOFC) for intermediate temperature (650–800 °C) operation was fabricated and operated in this study. We fabricated flat tubular ceramic supports through an extrusion process and analyzed the basic properties of the flat tubular ceramic support: the visible microstructure, porosity, mechanical strength, and pore size distribution. After that, we manufactured a flat tubular SIS-SOFC single cell using screen printing and a vacuum slurry dip-coating method for the electrode/interconnect and electrolyte. In addition, to make a unit bundle for a flat tubular SIS-SOFC, five SIS-SOFC single cells with an effective electrode area of 0.8 cm^2 were coated onto the surface of the prepared ceramic support and were connected in series using an Ag + glass interconnect between each single SIS-SOFC cell. The performance of the 5-cell unit bundle for a flat tubular SIS-SOFC in 3% humidified H_2 and air at 800 °C had a maximum power of 2.5 W.

1. Introduction

The solid oxide fuel cell (SOFC), which combines heat and electrical power, was developed for next generation power systems. Recently, the SOFC has received more attention due to its higher efficiency, waste-heat utilization and fuel flexibility – all of which are the major advantages for replacing the conventional power generating systems. Nonetheless, the SOFC still has some disadvantages in its actual application to a power system. Above all, the durability and cost competitiveness of the SOFC are recognized as the technical barriers that hinder the entry of SOFCs into the commercial market. Therefore, most R&D activities on SOFCs are focused mainly on the development of commercially viable SOFC technology with higher performance and longer durability, system integration [1–4].

There are two main SOFC configurations, tubular and planar. Planar SOFC performance is theoretically higher than that of tubular, because of reduced in-plane ohmic resistance. In addition, tape casting and other mass production techniques, for example plasma-spray, can easily be applied for planar SOFC production and fabrication, thus making possible continuous production and cost reduction. On the other hand, the tubular configuration, because of its geometry, is capable of solving problems related to cracking, thermo-cycling, quick start-up time and sealing [5–7].

In addition to these general SOFC geometries, recently, segmented-in-series (SIS)-solid oxide fuel cells have been attracting more and more attention [8]. The advantages of segmented-in-series solid oxide fuel cells (SIS-SOFCs) over tubular SOFCs include simplification of the externally electric and mechanical connection, decrease of the current path and increase of the energy density per unit volume [4,9,10]. Being light and compact, it is very efficient for practical SOFC application [7,11,12].

There are two main configurations of SIS-SOFC: banded tubular SIS-SOFC and cone-shaped anode supported SIS-SOFC. Banded tubular SIS-SOFC, composed of a porous insulating substrate and anode/electrolyte/cathode tri-layers, have excellent redox stability and extended range of coke-free operation conditions due to the thinness of the anode membrane and the porous substrate barrier layer [13–16]. Fig. 1 shows a schematic diagram of banded tubular SIS-SOFC. Barnett's research group prepared banded tubular SIS-SOFC on partially stabilized zirconia (PSZ) flattened-tube supports. They obtained suitable performance by decreasing the width of each cell unit [17]. However, the decrease in the cell width complicates the fabricating process and restricts the total output power [18]. Moreover, the fabrication process, specifically the gel casting for the insulating substrate, may not be suitable or acceptable for continuous, mass production of ceramic supports [19].

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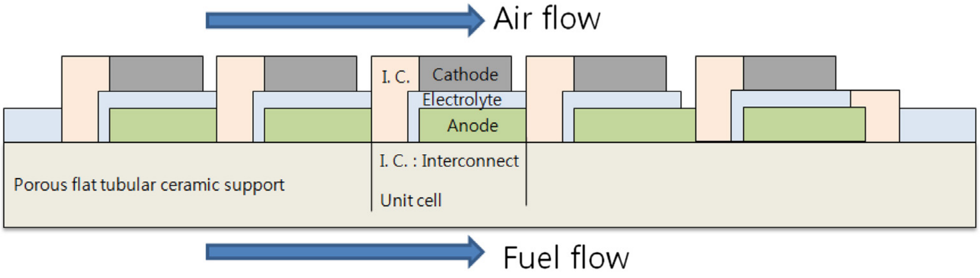

Fig. 1. Schematic diagram of the unit bundle for the flat tubular segmented-in-series (SIS)-SOFC.

A cone-shaped, anode-supported SIS-SOFC stack is a new design proposed by Refs. [18,20]. This unique SIS-SOFC stack is composed of several cone-shaped single SOFC cells connected in an electrical, gas flowing series. However, gas leakage at an interconnect composed of silver paste has somewhat reduced the open-circuit voltage (OCV) of the total cone-shaped SIS-SOFC stack and induced performance degradation of continuous operation of the SIS-SOFC [11,12,18,20,22].

Almost all of the current literature on SIS-SOFC has focused exclusively on the anode-support cone-shaped SOFC [11,12,18,20,22] and tubular SIS-SOFC configurations based on a complex fabrication method such as the gel-casting process [9,10,17,21]. There are few studies on the flat tubular SIS-SOFC based on a continuous, mass fabrication method such as the extrusion process. In order to evaluate a flat tubular SIS-SOFC, the best design and fabrication of both the ceramic support and the unit bundle for a flat tubular SIS-SOFC are necessary and important.

Therefore, in this work, we present an easy and simple fabrication procedure for the flat tubular segment-in-series (SIS)-SOFC unit bundle and an optimized design for current collection of SIS-SOFC electrode, and the sealing & interconnection method between SIS-SOFC single cells. Based on our results, we fabricated and analyzed the basic properties and operating variables of a ceramic support and a unit bundle for flat tubular SIS-SOFC.

2. Experimental

2.1. *Preparation of the flat tubular ceramic support and fabrication of the flat tubular segmented-in-series-SOFC*

The flat tubular ceramic tube as a support of the segmented-in-series (SIS)-SOFC was fabricated by the extrusion method [8]. The powder for the flat tubular ceramic tube (3 mol.% yttria-stabilized ZrO_2(3YSZ), TERIO, China) and activated carbon (YP-50F, Kuraray Chemical., Japan) as a pore former were well weighed and mixed in ethanol by ball milling for one week and then dried. An organic binder and distilled water were added to the dried powder, and then the well-dispersed paste was extruded in the form of a flat tubular support. The extruded flat tubes were dried in a constant temperature and humidity dryer (TH-G 300, DB Hi-tech., brand, Korea) at 80 °C for 24 h, and then pre-sintered at 1100 °C. After the thermal treatment of the ceramic support, its basic characteristics, the real picture, cross-section of SEM picture, porosity, pore size distribution (PSD), and mechanical strength were measured and analyzed.

Screen printing was employed to deposit the anode, cathode, and interconnect layers, and vacuum slurry dip-coating was used to coat the electrolyte layer on the surface of the anode functional layer (AFL) and flat tubular ceramic support. The screen printing pastes were prepared after first ball milling the relevant powders, and then combining them with a high speed mixer (HKSM-50, Korea) and pre-treating them in a three-roll mill (EXAKT, Germany). Prints with excellent uniformity and well-defined patterns were suitably obtained. Details of the screen printing, such as the pastes of solids loading and rheology, adjustment of the printing parameters, and surface flatness requirements, have been described by Pillai et al. [17], Pi et al. [23], Pi et al. [24].

The AFL material was NiO-ScSZ cermets. NiO (J.T. Baker, USA) and Ce_1ScSZ_{10} (DAICHI KIGENSO KAGAKU KOGYO Co., LTD, Japan) were mixed in the weight ratio 1:1 before the making of paste with solids loading of 55–60% by weight.

The ScSZ (AnaKasei Co., LTD, Japan) electrolyte layer and GDC ($Ce_{0.9}Gd_{0.1}O_{1.95}$, Fuelcell Material (FCM), USA) buffer layer were coated onto the surface of the flat tubular ceramic support and AFL using a vacuum slurry dip-coating method to form a thin and crack-free layer, which was then co-fired at 1400 °C [8,25,26]. The multi-layered cathode was composed of an LSCF($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}$)/GDC composite, LSCF, and an LSCF/LSCo ($La_{0.6}Sr_{0.4}CoO_3$) composite, and was coated onto the cosintered flat tubular ceramic support and suitable layer including the electrolyte, and then sintered at 1150 °C for 3 h [27].

The Ag + glass composite interconnect was made from a mixture of Ag (Kojundo Chemical Lab. Co., Ltd, Japan) and glass (Daejoo Electronic Materials Co., Ltd, Korea) in a weight ratio of 9:1. The detailed properties of the interconnect has been well reported in our group's paper [23,24,30]. The interconnect conductivity was measured separately using unpatterned layers at each operating temperature using a four-point geometry [9,23]. The overall flow chart of the manufacturing process for flat tubular SIS-SOFC is well shown in Fig. 2.

2.2. *Fabrication and operation of the unit bundle for the flat tubular segmented-in-series (SIS)-SOFC*

Fig. 3 is a real photograph of the unit bundle for the flat tubular SIS-SOFC, which is composed of five single cells for a flat tubular SIS-SOFC fabricated by a screen printing and vacuum slurry dip-coating method using a unique masking method. As can be seen from Fig. 3, the single SIS-SOFC cell is fabricated onto the surface of the ceramic support to form a unit bundle for the flat tubular SIS-SOFC. Each single SIS-SOFC cell is connected with electrical and gas flow in series. The interconnect serves as the sealing and electrical connection between the anode of one single cell and the cathode of the next single cell. Ag wire was used as the current collector for both electrodes of the unit bundle for the flat tubular SIS-SOFC. Both ends of the unit bundle for the flat tubular SIS-SOFC were connected to the metal cap using ceramic glue as a sealing and joining material (Ceramabond, Aremco, USA) to prevent leakage of fuel. Then the as-prepared unit bundle was heated at 500 °C for 2 h with a heating rate of 2 °C $min.^{-1}$ to evaporate the solvent and binder in the Ag–glass composite paste, in order to densify the connection. Hydrogen (300 ml min^{-1}) saturated with water was used as fuel at the anode side and ambient air (1500 ml min^{-1}) was used as an oxidant at the cathode side. The 5-cell unit bundle was