Journal of Power Sources 300 (2015) 318–324

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Design of a dual-layer ceramic interconnect based on perovskite oxides for segmented-in-series solid oxide fuel cells

Beom-Kyeong Park [a, b], Dae-Wi Kim [a], Rak-Hyun Song [a], Seung-Bok Lee [a], Tak-Hyoung Lim [a], Seok-Joo Park [a], Chong-Ook Park [b], Jong-Won Lee [a, c, *]

[a] New and Renewable Energy Research Division, Korea Institute of Energy Research, 152 Gajeong-ro, Yuseong-gu, Daejeon, 305-343, Republic of Korea
[b] Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology, 335 Gwahak-ro, Yuseong-gu, Daejeon, 305-701, Republic of Korea
[c] Department of Advanced Energy Technology, Korea University of Science and Technology, 217 Gajeong-ro, Yuseong-gu, Daejeon, 305-350, Republic of Korea

HIGHLIGHTS

- Dual-layer interconnect design for segmented-in-series solid oxide fuel cells.
- The interconnect comprises two perovskite oxides, $Sr_{0.7}La_{0.2}TiO_3$ and $La_{0.8}Sr_{0.2}FeO_3$.
- The interconnect exhibits low resistance and high stability in a dual atmosphere.
- The fuel cell with the interconnect films shows a power density of 340 mW cm^{-2}.

ARTICLE INFO

Article history:
Received 26 February 2015
Received in revised form
19 July 2015
Accepted 22 September 2015
Available online 29 September 2015

Keywords:
Solid oxide fuel cell
Segmented-in-series
Interconnect
Perovskite
Coating

ABSTRACT

A segmented-in series (SIS) SOFC consists of segmented unit cells connected in electrical series and shows improved stack efficiency over conventional SOFCs. In this design, a thin interconnect film provides both electrical contact and sealing between the anode of one cell and the cathode of the next; thus, it should have high conductivity and chemical/structural stability in both reducing and oxidizing atmospheres as well as impermeability to gases. Here, we report a dual-layer interconnect film for SIS–SOFCs comprising perovskite-type oxides, $Sr_{0.7}La_{0.2}TiO_3$ (exposed to a reducing atmosphere) and $La_{0.8}Sr_{0.2}FeO_3$ (exposed to an oxidizing atmosphere). The interconnect film is not only very dense but also highly conductive and stable under SOFC operating conditions; in particular, it shows an area-specific resistance of 19.6 mΩ cm^2 at 800 °C, which is much lower than the generally accepted limit for SOFCs. A flat-tubular SIS–SOFC fabricated using these interconnect films exhibits a power density as high as 340 mW cm^{-2}, which proves the feasibility of the dual-layer interconnect design.

1. Introduction

A solid oxide fuel cell (SOFC) is an electrochemical energy conversion device that can generate electricity directly from a wide range of fuels. As compared to low-temperate fuel cells, SOFCs operating at relatively high temperatures (600–900 °C) have a number of attractive features, such as high energy conversion efficiency, fuel flexibility, and rapid reaction kinetics [1–3]. A segmented-in-series (SIS) SOFC consists of multiple unit cells (anode/electrolyte/cathode) on a single porous support, which are connected in electrical series by interconnects [4]. This design offers many advantages over conventional SOFCs, such as (*i*) high output voltages at low currents, leading to improved stack efficiency; (*ii*) high tolerance to redox cycling owing to the stability of the ceramic support in both reducing and oxidizing atmospheres; (*iii*) reduced sealing areas; and (*iv*) no requirement of additional components between the segmented unit cells (*e.g.*, metallic current collectors) [5–8].

The SIS–SOFC requires a thin interconnect film sandwiched between the anode of one unit cell and the cathode of the adjacent

* Corresponding author. New and Renewable Energy Research Division, Korea Institute of Energy Research, 152 Gajeong-ro, Yuseong-gu, Daejeon, 305-343, Republic of Korea.
E-mail address: jjong277@kier.re.kr (J.-W. Lee).

http://dx.doi.org/10.1016/j.jpowsour.2015.09.082
0378-7753/

cell. The interconnect should be able to fulfill a dual function, namely, the electrical connection of the unit cells and the separation of fuel and oxidant gases [9,10]. The interconnect film should have high conductivity and stability in both reducing (on the anode side) and oxidizing (on the cathode side) atmospheres as well as negligibly low gas permeability (high film density). The material requirements for the interconnects are therefore considered the most demanding among the SIS–SOFC components. Furthermore, making the interconnect film is the most difficult step of cell fabrication because the thin and dense film must be coated on porous media.

Interconnect films for SOFCs are usually fabricated using perovskite-type oxide materials based on either chromites (*e.g.*, $La_{1-x}Ca_xCrO_3$, $La_{1-x}Sr_xCrO_3$, and $LaCr_{1-x}Mg_xO_3$) [10–13] or titanates (*e.g.*, $Sr_{1-x}La_xTiO_3$ and $Sr_{1-x}Y_xTiO_3$) [14–17]; however, there are still some critical issues to be addressed. Doped chromites or titanates display a dominant *p*-type or *n*-type conducing property, respectively, which results in high resistances when exposed to a dual atmosphere [10,13–17]. In addition, the poor sinterability of $LaCrO_3$ makes it difficult to fabricate dense interconnect films on a porous support. It has been known [10–13] that $LaCrO_3$ sintering proceeds mainly by evaporation and condensation of gaseous Cr–O species rather than by solid-state mass transport and that transient liquid phases ($CaCrO_4$ and $SrCrO_4$) formed during sintering soak into the underlying porous substrate.

In recent years, a dual-layer design has been proposed as a promising strategy to solve the problems associated with conventional interconnect materials [18–21]. Huang and Gopalan [18] first suggested the concept of a dual-layer interconnect comprising an *n*-type conductor on the anode side and a *p*-type conductor on the cathode side. Later, Xu et al. [19,20] and our group [21] demonstrated the feasibility of dual-layer interconnects having *n*-type (Sr,La)TiO_3 (SLT) and *p*-type (La,Sr)MnO_3 (LSM). In particular, we reported that a dense interconnect film could be fabricated on a porous anode support by the simple screen-printing and co-sintering of nano-sized $Sr_{0.7}La_{0.2}TiO_3$ and $La_{0.8}Sr_{0.2}MnO_3$ powders even without any sintering aids [21]. The fabricated SLT/LSM interconnect exhibited low resistances (*e.g.*, 47 mΩ cm^2 at 800 °C) and high stability in a dual atmosphere; hence, when applied to a flat-tubular SOFC, it caused no considerable performance loss compared to a metallic Ag–glass interconnect. As will be shown later, however, SLT/LSM-based interconnects are unsuitable for SIS–SOFCs, which would require improved patterning precision of the interconnect films. It turned out that Mn diffusion and evaporation from the LSM layer during high-temperature sintering hinder the formation of well-defined boundaries between the interconnects and the other cell components.

With the objective of avoiding the LSM-induced problems, in this study, we designed and fabricated a dual-layer interconnect based on *n*-type $Sr_{0.7}La_{0.2}TiO_3$ (SLT) and *p*-type $La_{0.8}Sr_{0.2}FeO_3$ (LSF) for SIS–SOFCs. Dense SLT/LSF interconnect films with various thicknesses were fabricated on a porous substrate, and their microstructures and electrical properties were examined. Then, a flat-tubular SIS–SOFC with five segmented unit cells was fabricated and tested using the dual-layer (SLT/LSF) interconnect films. Our experimental results demonstrate that the interconnect film is not only very dense but also highly conductive and stable in a dual atmosphere and that the combination of the SLT and LSF layers allows for the formation of well-defined interconnect films for SIS–SOFCs.

2. Experimental

2.1. Preparation of perovskite oxides and screen-printing pastes for interconnect coating

Perovskite oxide materials for interconnects, such as $Sr_{0.7}La_{0.2}TiO_3$ (SLT), $La_{0.8}Sr_{0.2}FeO_3$ (LSF), and $La_{0.8}Sr_{0.2}MnO_3$ (LSM), were synthesized by the Pechini method using citric acid, as reported elsewhere [17,21]. The requisite precursors, $Sr(NO_3)_2$ (≥99.0%, Aldrich), $La(NO_3)_3 \cdot 6H_2O$ (99.99%, Kanto), $Ti(OCH(CH_3)_2)_4$ (99.999%, Aldrich), $Fe(NO_3)_3 \cdot 9H_2O$ (≥98%, Aldrich), and $Mn(CH_3COO)_2 \cdot 4H_2O$ (99%, Aldrich), were dissolved in a mixture of distilled water and ethylene glycol (99.8%, Aldrich). To make a viscous gel, citric acid (≥99.5%, Aldrich) was added to the solution, followed by aging at 120 °C. The gel was dried at 250 °C and then calcined at 800 °C for 5 h to obtain pure perovskite oxides. The screen-printing pastes for interconnect coating were prepared by mixing the synthesized powder with ethyl cellulose (Junesi) and α–terpineol (Junsei).

2.2. Fabrication and performance evaluation of a flat-tubular SIS–SOFC with the dual-layer interconnect films

The flat-tubular SIS–SOFC fabricated in this study has five segmented unit cells on a support of 3 mol.% Y_2O_3-stabilized ZrO_2 (3YSZ), and each segment consists of the following components: (*i*) a composite anode of NiO and 10 mol.% Sc_2O_3-stabilized ZrO_2 (ScSZ), (*ii*) a dual-layer electrolyte of ScSZ (on the anode side) and 10 mol.% Gd-doped CeO_2 (GDC) (on the cathode side), (*iii*) a dual-layer cathode of $La_{0.6}Sr_{0.4}CoO_3$–GDC and $La_{0.6}Sr_{0.4}CoO_3$, and (*iv*) a dual-layer interconnect of SLT (on the anode side) and LSF (on the cathode side). Fig. 1 illustrates a schematic diagram of the flat-tubular SIS–SOFC with 5 segmented unit cells along with the cross-sectional view of the unit cell. The 3YSZ support has sixteen fuel flow channels with a diameter of 1.5 mm. The thicknesses of the SIS–SOFC components (support, anode, electrolyte, cathode, and interconnect) are indicated in Fig. 1(b). In the present study, 3YSZ (Tosoh) was used as a support material for the SIS–SOFCs due to its excellent mechanical strength and high stability as well as its good thermal expansion compatibility with other cell materials. First, a flat-tubular support was fabricated by extruding a mixture of 3YSZ, activated carbon (YP–50F, Kuraray), and an organic binder (YB-131D, Yuken). Here, the activated carbon was employed as a pore-former to obtain the desired porosity. The extruded support was then pre-sintered at 1100 °C for 3 h in air. Second, 5 segmented anodes of NiO (J. T. Baker) and ScSZ (Daiichi Kigenso Kagaku Kogyo) were coated on the pre-sintered support by a screen printing technique. After this, the anodes were partially covered with masking tape for interconnect coating. Third, segmented electrolytes of ScSZ and GDC (Rhodia) were formed by a vacuum slurry coating process. Fourth, segmented dual-layer interconnects were coated onto the anodes by the consecutive screen-printing processes of SLT and LSF. The support, anodes, electrolytes, and interconnects were co-sintered at 1400 °C for 5 h in air. Finally, segmented cathodes were screen-printed onto the electrolytes and were then sintered at 1100 °C for 3 h in air. For electrochemical measurements, the fabricated SIS–SOFC was mounted with two metal tubes for H_2 supply using a ceramic adhesive. For current collection, one Ag wire was attached to the anode of the first segmented cell using an Ag–glass composite paste, whereas the other was attached to the cathode of the fifth segmented cell. The polarization (cell voltage *vs.* current density) data of the SIS–SOFC were collected by using an automated fuel cell test station. During electrochemical testing, H_2 gas with 3 vol.% H_2O was supplied to the channels of the support *via* a metal tube, while air was allowed to flow over the cathode.

2.3. Characterizations of the dual-layer interconnects

To determine its electrical properties, a dual-layer interconnect was fabricated on a porous NiO–8YSZ support by the screen