



Effects of cathode materials on the characteristics of electrolyte supported micro-tubular solid oxide fuel cells



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HIGHLIGHTS

- Properties of tubular SOFC were improved by LSCF–GDC cathode and GDC interlayer.
- The fabrication was improved by carefully monitoring formulation and sintering.
- An LSCF cathode had a power density 44% greater than an LSM cathode.
- Drop in ASR value of LSCF cathode was >30% compared to that of LSM cathode.

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ABSTRACT

The effects of the GDC–LSCF ($\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{0-\delta}\text{–La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$) cathode layer and the GDC interlayer on the electrochemical performance of the ScSZ ($\text{Zr}_{0.8}\text{Sc}_{0.2}\text{O}_{2-\delta}$) electrolyte supported ($\approx 270\ \mu\text{m}$) micro-tubular SOFC cells are investigated in this study. Material formulation and sintering profile for fabricating the micro-tubular SOFC cells are developed to avoid physical defects caused by the large sintering shrinkage mismatch among the layers. Cell B (with the LSCF–GDC composite cathode layer and the GDC interlayer) reports an ohmic resistance slightly higher than that of Cell A (with the GDC– $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{3-\delta}$, i.e. LSM, composite cathode), while its polarization resistance emerges to be significantly smaller than that of Cell A. In terms of cell performance, Cell B demonstrates an OCV value ($> 1.07\ \text{V}$) similar to that of Cell A and a maximum power density ($0.26\ \text{W cm}^{-2}$) 44.4% greater than that of Cell A ($0.17\ \text{W cm}^{-2}$) at $850\ ^\circ\text{C}$. It can thus be concluded that using the LSCF–GDC composite-cathode layer and inserting the GDC interlayer help reduce the total cell impedance, thereby improving the power density of the tubular cells.

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1. Introduction

Solid oxide fuel cell (SOFC) has attracted worldwide interest with its promising commercialization potential thanks to major advantages like high energy conversion efficiency, structural integrity, fuel flexibility, and non-reliance on noble metals of its electrodes [1,2]. To date, planar and tubular designs remain the two most common SOFC configurations. Compared to their planar counterparts, tubular SOFCs are known for superb thermal resistance, secure sealing, solid mechanical strength, rapid heat cycling, and stable performance. The drawbacks, on the other hand, are smaller current density and complex fabrication process [1,3–5]. Considerable efforts have accordingly been invested to reduce cell

size and fabricate anode-supported SOFCs with thin electrolytes for raising volumetric power density [6–10]. However, anode-supported tubular SOFCs have often encountered mechanical failure during operation mainly due to the large volume change (around 40 vol.%) of the anode during the reduction and re-oxidation cycles, which may easily crack the thin electrolyte layer and delaminate between the electrode and electrolyte to decrease the cell open circuit voltage (OCV) [11–16].

Using extrusion and dip-coating to prepare electrolyte-supported micro-tubular SOFCs (T-SOFCs), a previous study found the NiO/NiO–ScSZ/ScSZ/GDC–LSM cell exhibiting fine flexural strength (190 MPa), and the micro-tubular SOFCs, after thermal recycling, showed no delamination and retained good mechanical integrity [17]. Yet, the maximum power density (MPD) of the micro-tubular SOFCs reached only $0.23\ \text{W cm}^{-2}$ at $900\ ^\circ\text{C}$ due to the cells' high ohmic and polarization resistances. The ohmic resistance can be reduced by using $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{0-\delta}$ (GDC) and $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{3-\delta}$ (LSGM) as the

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electrolyte, which further helps lower the operating temperature [18]. However, the cell performance may be degraded by the impaired mechanical strength or the electronic conduction in the electrolyte. It was shown that the Ce^{4+} ions could be reduced to Ce^{3+} ions under a reducing atmosphere, thereby causing some electronic conduction in the electrolyte and resulting in OCV drop [19]. The high polarization resistance could be reduced by using a single-phase mixed ionic and electronic conductor (MIEC), such as $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF), to extend the triple-phase boundary (TPB) into the whole cathode. Unfortunately, LSCF may react with the ZrO_2 -based electrolyte over $900^\circ C$, leading to the formation of the performance-degrading $SrZrO_3$ and $La_2Zr_2O_7$ compounds [20–23].

In this study, electrolyte-supported micro T-SOFCs using ScSZ electrolyte and LSCF cathode were fabricated by extrusion and dip-coating. To improve the chemical compatibility between ScSZ and LSCF, a GDC interlayer was inserted between the electrolyte and cathode layers of the NiO/NiO–ScSZ/ScSZ/GDC–LSCF cells [23,24]. The extruded green ScSZ tubes were pre-sintered at $1100^\circ C$ and the anode dip-coated onto the inner surfaces of the electrolyte tubes and then sintered at $1400^\circ C$. A NiO current collector layer and a GDC interlayer were then coated respectively onto the inner and outer surfaces of the microtubes before co-sintering at $1350^\circ C$. The fired microtubes consisting of an anode layer, an electrolyte layer, and an interlayer were subsequently dip-coated with the GDC/LSCF cathode and fired at $1200^\circ C$. The NiO/NiO–ScSZ/ScSZ/GDC/GDC–LSCF micro T-SOFCs were built and characterized through microstructural and electrochemical performance studies. The study further investigated and compared the electrochemical performance of the ScSZ-supported micro T-SOFCs using LSCF as the cathode with the electrochemical performance of those with an LSM cathode.

2. Experimental

Fig. 1 presents the schematic drawing of the electrolyte-supported micro T-SOFCs with $La_{0.8}Sr_{0.2}MnO_{3-\delta}$ (LSM) and $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF) cathodes. The two designs of micro T-SOFCs used the same half-cell micro-tubes incorporating a $Zr_{0.8}Sc_{0.2}O_{2-\delta}$ (ScSZ) electrolyte tube and a two-layer anode comprising an anode functional layer (AFL) of NiO–ScSZ composite (60 vol.:%:40 vol.:%) and a current collector layer (outer layer) of pure NiO. For the two designs, Cell A used a $Ce_{0.8}Gd_{0.2}O_{2-\delta}$ (GDC)–LSM composite and Cell B a GDC–LSCF composite as the cathode layer. A GDC interlayer was inserted between the ScSZ electrolyte tube and

the GDC–LSCF cathode layer in Cell B to prevent the formation of $SrZrO_3$ and $La_2Zr_2O_7$.

Commercially available raw $Zr_{0.8}Sc_{0.2}O_{2-\delta}$ (ScSZ; $d_{50} = 0.09 \mu m$; Fuel Cell Materials) was mixed with binder (Methyl cellulose, MC, Tsair Yu Technology), lubricant (Oil, Tsair Yu Technology, Taiwan), surfactant, and D.I. water. The mixtures were extruded into the micro-tubes with an in-house designed die (diameter = 5 mm). After cutting and drying, the green ScSZ tubes were pre-sintered at $1100^\circ C$. The inner surfaces of the electrolyte tubes were then dip-coated with a NiO (Fuel Cell Materials, $d_{50} = 0.8 \mu m$)–ScSZ slurry and then co-fired at $1400^\circ C$. For the preparation of Cell A, the current collector layer of NiO was dip-coated onto the surfaces of the anode functional layer of NiO–ScSZ and subsequently sintered at $1350^\circ C$. The GDC–LSM (20 vol.:%:80 vol.:%) cathode layer were then dip-coated onto the outer surfaces of the half-cells of the electrolyte-anode micro-tubes in a suspension composed of GDC (Fuel Cell Materials; $d_{50} = 0.09 \mu m$) and LSM (Fuel Cell Materials; $d_{50} = 1.19 \mu m$) powders, and then post-fired at $1100^\circ C$ for 2 h. For the preparation of Cell B, the GDC interlayer was first dip-coated on the outer surfaces of the electrolyte micro-tubes with a green NiO layer in a GDC suspension and then co-sintered at $1350^\circ C$. Finally, the GDC–LSCF (20 vol.:%:80 vol.:%) cathode were dip-coated onto the surfaces of the GDC interlayer in a GDC–LSCF (Fuel Cell Materials; $d_{50} = 0.99 \mu m$) suspension and then post-fired at $1200^\circ C$. Details of the cell preparation are presented elsewhere [18]. In order to evaluate the sintering shrinkage mismatch with respect to temperature for the GDC interlayer and the NiO coated on ScSZ electrolyte tube, dilatometric analysis on the green NiO, green GDC and pre-sintered ScSZ compacts was performed, using a dilatometer (NETZSCH DIL 402C) in air and at a heating rate of $5^\circ C \text{ min}^{-1}$. The fired ScSZ compact was pre-sintered at $1400^\circ C$ for 2 h.

Scanning electron microscopy (SEM, Joel JSM-6510LV) associated with energy dispersive spectroscopy (EDS, INCA X-ACT) was used to conduct chemical analysis and examine the microstructures of the fracture surfaces. The electrochemical performances of the micro T-SOFCs were evaluated using an in-house designed setup. Ag wire and Ni foam were respectively used as cathode and current collector. The micro T-SOFCs were mounted to an alumina tube with hydrogen flowing inside using a sealant (Aremco products, Zirconia 885). The anode was first reduced in H_2 at $700^\circ C$ for 1 h. The cell voltage and the power density as a function of cell current density were determined using a potentiostatic instrument (Jiehan ECW-5000) at 800 – $900^\circ C$ at intervals of $50^\circ C$. The impedance analysis was measured by an electrochemical impedance analyzer

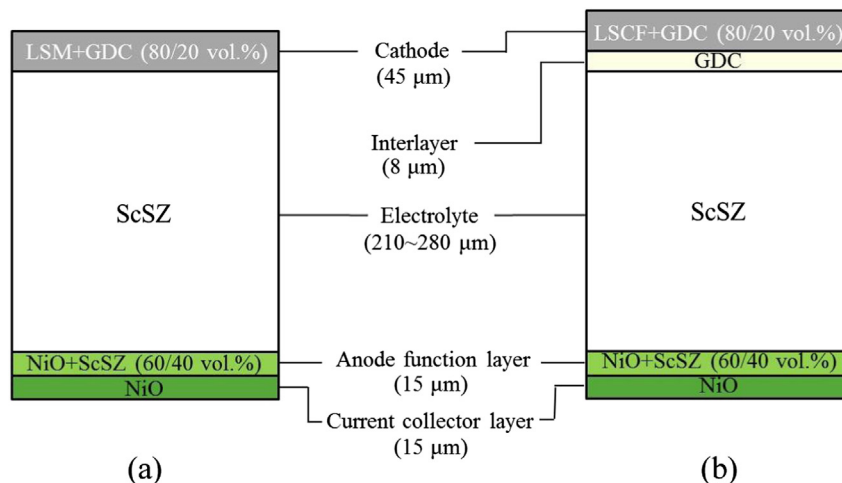


Fig. 1. Schematic drawing of the electrolyte-supported cell structure of micro T-SOFCs: (a) LSM–GDC/ScSZ/NiO–ScSZ/NiO (Cell A); (b) LSCF–GDC/GDC/ScSZ/NiO–ScSZ/NiO (Cell B).

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