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Ni–YSZ-supported tubular solid oxide fuel cells with GDC interlayer between YSZ electrolyte and LSCF cathode

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ARTICLE INFO

Article history:
Received 10 May 2014
Received in revised form
17 June 2014
Accepted 19 June 2014
Available online 14 July 2014

Keywords:
Gadolinia doped ceria
GDC interlayer
Carbonate coprecipitation
Tubular solid oxide fuel cell
Yttria stabilized zirconia
$La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$

ABSTRACT

Highly sinterable gadolinia doped ceria (GDC) powders are prepared by carbonate coprecipitation and applied to the GDC interlayer in Ni–YSZ (yttria stabilized zirconia)-supported tubular solid oxide fuel cell in order to prevent the reaction between YSZ electrolyte and LSCF ($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$) cathode materials. The formation of highly resistive phase at the YSZ/LSCF interface was effectively blocked by the low-temperature densification of GDC interlayer using carbonate-derived active GDC powders and the suppression of Sr diffusion toward YSZ electrolyte via GDC interlayer by tuning the heat-treatment temperature for cathode fabrication. The power density of the cell with the configuration of Ni–YSZ/YSZ/GDC/LSCF–GDC/LSCF was as high as 906 mW cm^{-2}, which was 2.0 times higher than that (455 mW cm^{-2}) of the cell with the configuration of Ni–YSZ/YSZ/LSM($La_{0.8}Sr_{0.2}MnO_{3-\delta}$)–YSZ/LSM at 750 °C.

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Introduction

Solid oxide fuel cells (SOFCs)—electrochemical devices that convert chemical energy into electrical energy—can be divided into two categories according to their shape: planar type and tubular type. Tubular SOFCs are close to commercialization owing to simple gas sealing, good mechanical strength, stability during the thermal cycle, and easy scale up [1]. The operation of SOFCs in low- (≤600 °C) and intermediate-(600–800 °C) temperature regimes is advantageous to realize long-term stability, quick start-up, long lifetime of the stack, and the use of cost-effective gas sealants and interconnector materials.

The coating of a thin yttria stabilized zirconia (YSZ) electrolyte with excellent stability in a highly reducing atmosphere on tubular Ni–YSZ supports is a probable design for low- and intermediate-temperature SOFCs. To date, $La_{1-x}Sr_xMn_{3\pm\delta}$ (LSM) has been one of the most widely used representative cathode materials. The cathode polarization of

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http://dx.doi.org/10.1016/j.ijhydene.2014.06.103

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LSM, however, increases significantly as the operation temperature decreases down to 500–700 °C, which deteriorates the performance of SOFCs [2]. Accordingly, Co-containing perovskite materials with high electrical conductivity, such as $La_{1-x}Sr_xCoO_{3-\delta}$ (LSC), $Sm_{1-x}Sr_xCoO_{3-\delta}$ (SSC), $Sr_{1-x}Ce_xCoO_{3-\delta}$ (SCC), $Ba_{1-x}Sr_xCo_{1-y}Fe_yO_{3-\delta}$ (BSCF), $Ba_{1-x}Pr_xCo_{1-y}Fe_yO_{3-\delta}$ (BPCF) and $La_{1-x}Sr_xCo_{1-y}Fe_yO_{3-\delta}$ (LSCF), have been widely explored as cathode materials [3–8]. However, the heat treatment of a cathode layer at elevated temperatures (≥1000 °C) often deteriorates the cell performance owing to the formation of highly resistive second phases such as $La_2Zr_2O_7$ and $SrZrO_3$ [9] at the interface between YSZ and cathode layers [10–12]. A thin layer of doped ceria such as gadolinium-doped ceria (GDC), samarium-doped ceria (SDC), and yttrium-doped ceria (YDC) with high ionic conductivity [13] and low reactivity to Co-containing cathodes [14] can be placed between the YSZ electrolyte and cathode layer to enhance the cell performance by preventing the reaction between the cathode and YSZ [15,16]. The densification of a GDC interlayer at >1200 °C, unfortunately, can induce other adverse effects due to the formation of a resistive second phase at the GDC/YSZ interface by the interdiffusion of cations [17,18].

In planar-type SOFCs, various physicochemical routes have been employed to deposit thin and dense GDC interlayers, including screen printing [19], dip coating [20], magnetron sputtering [21], aerosol deposition [22], plasma-beam sputtering deposition [23], and electrophoretic deposition [24]. However, in tubular SOFCs, it is difficult to use various deposition methods that are carried out at low- and mid-temperature regimes because of the limitation in the support morphology; thus, the dip coating of slurry and subsequent sintering is typically used to form the GDC interlayer. Suzuki et al. [25] prepared a Ni–YSZ anode-supported (diameter: 1.7 mm) microtubular SOFC with maximum power density (0.39 W cm^{-2} at 600 °C) by inserting GDC interlayer between YSZ electrolyte and LSCF–GDC cathode. They also reported the high power density (0.57 W cm^{-2} at 750 °C) of the Ni–YSZ anode-supported (diameter: 1.93 mm) microtubular SOFC with scandia stabilized zirconia electrolyte and GDC interlayer [26]. Wu et al. [27] fabricated the tubular SOFC with configuration of Ni–YSZ/YSZ/SDC/$Sm_{0.5}Sr_{0.5}CoO_3$-SDC on glass tube (diameter: 10 mm, thickness: 0.4 mm). They combined dip coating and impregnation processes in order to increase the density of SDC interlayer. Note that low-temperature sintering of the GDC interlayer is essential to block the interdiffusion between YSZ and cathode layers.

In the present study, GDC nanopowders were prepared by carbonate precipitation that is known as a promising chemical route to decrease the sintering temperature [28,29] and applied to the cathode interlayer in Ni–YSZ-supported tubular SOFCs with the cell configuration of Ni–YSZ/YSZ/GDC/LSCF–GDC/LSCF. The formation of a resistive phase at the YSZ/LSCF and YSZ/GDC interfaces could be successfully prevented by the low-temperature sintering of the GDC interlayer and the control of the cathode-forming temperature. By employing this method, a high power density of 906 mW cm^{-2} was achieved at 750 °C. The main focus of this study was directed at the design of high-performance Ni–YSZ-supported tubular SOFCs using a thin YSZ electrolyte through the introduction of a GDC interlayer.

Experimental

Powder preparation

The $Gd_{0.1}Ce_{0.9}O_{1.95}$ (GDC) powders were prepared by carbonate coprecipitation [28]. $Ce(NO_3)_3{\cdot}6H_2O$ (Kanto Chemical Co. Ltd., Japan) and $Gd(NO_3)_3{\cdot}6H_2O$ (Kanto Chemical Co. Ltd., Japan) were used as the source materials, and $(NH_4)_2CO_3$ (Sigma–Aldrich, USA) was used as the precipitant. The precursors were prepared by the addition of mixed source solution to the precipitant solution. Stoichiometric amounts of each component of GDC were dissolved in 250 ml of distilled water, subsequently, the mixed aqueous solution ($[Ce^{3+}]$ = 0.09 M, $[Gd^{3+}]$ = 0.01 M) was dripped slowly into 250 ml of $(NH_4)_2CO_3$ aqueous solution ($[(NH_4)_2CO_3]$ = 0.5 M). During the precipitation, the solution was stirred mildly and the solution temperature was kept at 60 °C. The precipitates were washed and then dried for 24 h at room temperature. After pulverization, the precipitates were calcined at 700 and 900 °C for 2 h.

Cell fabrication

NiO–YSZ composite powders (NiO:YSZ = 40:60 by vol%) were prepared by mixing YSZ powders (8 mol% yttria stabilized zirconia, TZ-8Y, Tosoh Co., Japan) and NiO powders (J. T. Baker Co., USA). The NiO–YSZ composite powders and activated carbon (pore former) were weighed and mixed in ethanol by ball milling for 14 days and then dried. An organic binder and distilled water (25 wt%) were added to the dried powders and the mixture was homogenized to form a paste. The paste was extruded in the form of a tube and dried at 120 °C for 12 h. A NiO–YSZ tubular support was prepared by the heat treatment of the dried tubular structure at 1100 °C.

A YSZ (TZ-8Y, Tosoh Co., Japan) electrolyte layer was coated onto the porous NiO–YSZ tubular support by dip coating the support in slurry. The slurry for YSZ coating was prepared according to the following procedure: YSZ powders (15.0 g) were added to solvent mixtures of toluene (21.0 g) and 2-propanal (35.4 g). After addition of sorbitan trioleate (dispersant, 0.15 g) and Triton X-100 (surfactant, 0.3 g), the slurry was ball-milled for 24 h. Subsequently, di-*n*-butyl phthalate (1.5 g) and polyvinyl butyral (2.0 g) were added as a plasticizer and binder, respectively, and the slurry was ball-milled again for 24 h. The NiO–YSZ tubular support was immersed in YSZ slurry for 10 s, pulled out slowly, and dried at room temperature. The dipping and pulling speeds for dip coating were 100 and 10 mm min^{-1}, respectively. The resultant cell was sintered at 1400 °C for 5 h.

In the tubular SOFCs using a $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF) cathode, the GDC interlayer was coated onto the YSZ electrolyte layer by dip coating. For this, GDC powders prepared via carbonate coprecipitation were used. The sintered NiO–YSZ/YSZ tubular cell was immersed in GDC slurry for 10 s, pulled out slowly, dried at room temperature, and heat-treated at

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