



Development of tubular anode-supported solid oxide fuel cell cell and 4-cell-stack based on lanthanum gallate electrolyte membrane for mobile application

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HIGHLIGHTS

- Tubular LSGM membrane electrolyte SOFCs cell/stack are successfully developed.
- Cell/stack exhibits good performance fueled with bubbled hydrogen and methane.
- Stability test of stack is investigated at constant current densities fueled with CH₄.

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ABSTRACT

Anode-supported tubular solid oxide fuel cells and 4-cell-stack with lanthanum-doped ceria barrier layer and strontium, magnesium doped lanthanum gallate tri-layer electrolytes membrane are investigated. The single cell exhibits peak power densities of 1061 mW cm⁻² and 917 mW cm⁻² at 800 °C fueled with bubbled hydrogen and methane (3 vol % water), respectively. Energy dispersive X-ray detector result demonstrates that no measurable carbon is detected in the anode directly operated in methane during the whole testing process.

The 4-cell-stack assembled with single cells provides maximum output power of 1.66 W and open circuit voltage of 3.13 V at 800 °C with hydrogen as fuel. While the 4-cell-stack presents maximum output power of 1.38 W and open circuit voltage of 3.88 V at 800 °C fueled with methane. A stability test of each cell in 4-cell-stack is developed at constant current densities ($J = 0 \text{ A cm}^{-2}$, $J = 0.1 \text{ A cm}^{-2}$, $J = 0.4 \text{ A cm}^{-2}$) directly fueled with 75 ml min⁻¹ methane at 800 °C. The results show that the voltages of each cell in 4-cell-stack stabilize during the whole durability test. The results demonstrate that the stability and dependability of 4-cell-stack is good, and it is promising for mobile application of intermediate temperature solid oxide fuel cells.

1. Introduction

LSGM (strontium, magnesium doped lanthanum gallate) with perovskite type is considered as one of alternative electrolyte material for intermediate temperature solid oxide fuel cells (IT-SOFCs) applications. LSGM has some advantages such as high oxygen ionic conductivity, good chemical stability and good mechanical strength [1–6]. And it has not been thoroughly investigated nowadays, especially for the application of LSGM thin electrolyte membrane on anode-supported SOFCs. But the reactions of LSGM with the cathode and anode materials have been a major problem. The performance of the cell or stack may degrade with a LSGM electrolyte membrane for La diffusion at the anode/electrolyte interface and inter-diffusion of Ga and Co across the cathode [7,8]. It has been proved that introducing LDC isolated layer can

effectively minimize reactions between LSGM electrolyte membrane and the anode with Ni or the cathode with Co [9–11].

Exciting results about the thin LSGM electrolyte SOFCs with high power densities have been reported [7,12–15]. Lin [14] has explored the use of co-firing to prepare anode-supported SOFCs with LDC/LSGM/LDC electrolytes thin film and obtained the peak power density of 1.12 W cm⁻² at 750 °C with hydrogen as fuel. Bi [15] has fabricated anode-supported SOFCs with LDC-LSGM bilayer electrolytes thin membrane. Maximum power densities of 1.1 W cm⁻² and OCV of 1.02 V are achieved at 800 °C with hydrogen as fuel, respectively. Guo [7] has successfully prepared anode-supported SOFCs with LDC/LSGM/LDC tri-layer electrolytes membrane using centrifugal casting and gained good electrochemical performance with 1.23 W cm⁻² at 800 °C with hydrogen as fuel.

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However, up to now, many reports have been concentrated on the planar or disc SOFCs with LSGM electrolyte membrane including the reports mentioned above [16,17]. The tubular SOFCs, due to their geometry, are able to solve the problems of cracking, thermal stability, thermal shock resistance, and so on [18–21]. Furthermore, because of the all solid components, tubular SOFCs can be directly operated on a large number of fuels such as hydrocarbon fuels which is easily available [22]. Methane is cheap and it is readily available. And there is no pre-reforming for the cell or stack directly running on methane. Therefore, it is a widely used fuel for IT-SOFCs in hydrocarbon fuels [23–25]. Tubular SOFCs directly operated on methane are one of the most promising demonstration units for portable and mobile applications in future. There is very few literature on the tubular SOFCs with LSGM thin electrolyte membrane directly operated on methane. Also, there have been no reports or demonstrates about SOFCs stack preparation with LSGM thin electrolyte membrane using methane as fuel, to our knowledge.

In the present work, tubular anode-supported SOFCs cell and 4-cell-stack with tri-layer LDC/LSGM/LDC electrolytes membrane directly running on methane were fabricated and investigated. This kind of IT-SOFCs cell and stack with LSGM electrolyte thin film will accelerate the mobile application of SOFCs and it is hoped to substitute for the cell or stacks with YSZ electrolyte thin film in future.

2. Experimental

2.1. Powder preparation

NiO powder was synthesized by ammonia precipitation method [26]. Ammonia (analytical reagent, A.R.) is added to $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (A.R.) solution drop by drop to obtain nickel hydroxide deposition. Then, the as-prepared deposition is aged at 80 °C for several hours. After washed by alcohol and dried at 110 °C, the remainder is fired at 400 °C for 2 h to get nickel oxide powder.

$\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ (SDC) powder was prepared by citrate-nitrate process [27]. Stoichiometric amounts of $\text{Sm}(\text{NO}_3)_3$ (A.R.) and $\text{Ce}(\text{NO}_3)_3$ (A.R.) were dissolved in deionized water and citric acid (A.R.) was added as the complexant. The total molar ratio of cation to citric acid was 1:1.2. The solution was evaporated at 85 °C to form a transparent gel. After drying at 100 °C for 12 h, the transparent gel changed to dry gel. Finally, the dry gel was fired at 1000 °C for 4 h to get SDC powder.

LSCF ($\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_3$) powder was produced by a combined citrate-EDTA complexing method [7]. Stoichiometric amounts of $\text{La}(\text{NO}_3)_3$, $\text{Sr}(\text{NO}_3)_2$, $\text{Co}(\text{NO}_3)_2$ and $\text{Fe}(\text{NO}_3)_3$ are dissolved in water and followed by the addition of citric acid, EDTA and NH_3 . The pH value of the system was controlled around 6 during the entire evaporation process. The reaction mixture is then heated under constant stirring to obtain a purple gel. Afterwards the gel is pretreated at 150 °C for several hours to form a solid precursor, and then calcined at 1000 °C for 6 h to get the pure perovskite phase.

$\text{La}_{0.4}\text{Ce}_{0.6}\text{O}_{1.8}$ (LDC) and $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{3-\delta}$ (LSGM) powder were synthesized by using a solid-state method [28,29]. Stoichiometric amount of lanthanum oxide and ceria were mixed with ethanol by ball milling. The mixture after ball-milled was dried and calcined at 1250 °C for 21 h. And then the powders were ball-milled with ethanol and media in further for 3 days to gain LDC powders. A rotation speed of 200 rpm was used during ball milling. The synthesis process of lanthanum oxide is the same as that of LDC. The original powder of LSGM and LDC were both ball-milled for about 24 h before using in order to obtain the suitable grain.

2.2. Cell and stack fabrication

The NiO-SDC (weigh ratio of 6:4) tubular anode substrates and LDC/LSGM/LDC tri-layer electrolyte membrane were prepared by dip coating technique [30,31]. The technique is simple, fast and cost-

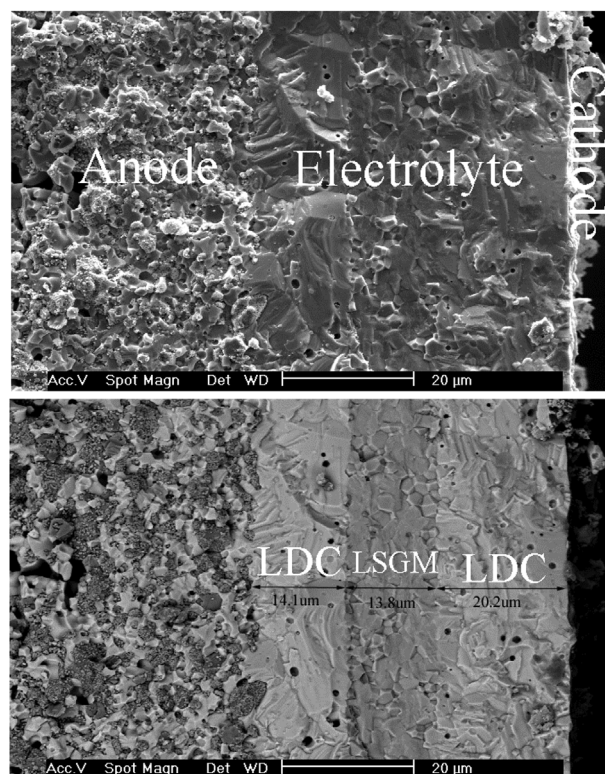


Fig. 1. SEM and BSE photograph of cross-sectional view of anode/electrolyte membrane/cathode (SEM: Scanning Electron Microscope; BSE: Back Scattered Electron Imaging).

effective. Then, the NiO-SDC anode and LDC/LSGM/LDC tri-layer electrolyte membrane were sintered at 1400 °C for 4 h to densify the tri-layer electrolyte film. LSCF-LSGM/LSCF bi-layer cathode were applied onto the LSGM electrolyte membrane by slurry coating process [30,31] and then sintered at 1000 °C for 3 h in air. The method of cell and 4-cell-stack assembling was similar to that described in our previous work [31–33]. A 4-cell-stack was assembled by connecting four cone-shaped tubular single cells in electrical and gas flow series. The smaller end of the first cell is closed, while the bigger end of the first cell and both ends of the other cells are open. Silver paste (Shanghai Research Institute of Synthetic Resins, Shanghai, China) serves as sealing and electrical connection was applied in the joint part to connect the each two single cell units. Then 4-cell-stack was heated to evaporate the solvent and binder in the silver paste in order to densify the connection.

2.3. Cell and stack testing

The method of cell and 4-cell-stack testing was similar to that described in our previous work [31–33]. A four-probe testing set-up was adopted to eliminate the ohmic loss in the silver wires. The single cell and 4-cell-stack were attached to one end of an alumina tube by using silver paste as sealing and jointing material, respectively. Fuel saturated with water at room temperature (3 vol % water) was used at the anode side at a flow rate of 75 ml min⁻¹ and ambient air was used as oxidant at the cathode side. The cell and 4-cell-stack performances and electrochemical impedance spectroscopy were measured using Iviumstat electrochemical analyzer (Ivium Technologies B.V., Netherlands). The current-voltage (I–V) curves were obtained by linear sweep voltammetry at a scanning rate of 5 mV s⁻¹. The impedances were measured in the frequency range of 100 kHz–0.1 Hz with signal amplitude of 5 mV under open-circuit condition. After electrochemical test, the cell/stack was fractured and examined using a scanning electron microscope (SEM) and Back Scattered Electron Imaging (BSE).

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