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Fabrication of composite cathode by a new process for anode-supported tubular solid oxide fuel cells

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ABSTRACT

Anode-supported tubular solid oxide fuel cells (SOFCs) fabricated by traditional method suffered from the charge exchange constraint on the interface of electrolyte and cathode. In this work, we introduced a new sintering process to obtain the dense and porous electrolyte bi-layer structure on anode-supported tubular SOFCs. The cathode was built by infiltrating nano-catalyst $La_{0.6}Sr_{0.4}Fe_{0.9}Sc_{0.1}O_{3-\delta}$ (LSFSc) into the porous electrolyte, which showed better electrochemical activity than traditional LSM cathode. The functional fuel cells showed the maximum power density of 292.3mW·cm^{-2} at 800 °C with H_2 as fuel and air as oxidant. This unique performance was due to the porous electrolyte provided more specific surface area for cathode and the nanostructured cathode catalyst showed high electro-catalysis for oxygen reduction reaction. Moreover, a long term test was carried out, demonstrating the excellent stability which was extremely important for application in the future.

1. Introduction

Solid oxide fuel cells (SOFCs) are one of the efficient energy conversion device and the most expected to be commercialized due to the high demand from the energy society[1]. Tubular solid oxide fuel cells are particularly attractive due to special advantages compared to planar type, such as higher mechanical, thermal stability, simpler seal requirement and higher volumetric power density[2]. Recent researches focused on the anode-supported tubular SOFCs due to the reduced resistances compared to the electrolyte-supported tubular cells and better mechanical strength than the cathode-supported tubular cells[3]. Despite the promise of anode-supported tubular SOFCs, the traditional fabrication method based on printing or dip-coating for cathode resulted in some potential problems, such as low performance and degradation [4,5]. Liu used the reference electrode to investigate the source of high resistance, and concluded that the ohmic resistance of the cathode/electrolyte interface and the polarization resistance of the cathode dominated the total resistance of the fuel cells[5]. Considering the high sintering temperature for $La_{0.8}Sr_{0.2}MnO_{3-\delta}$(LSM) cathode, e.g., 1200 °C, undesirable chemical reactions between ZrO_2-based electrolyte and LSM cathode usually happened and produced insulating impurities such as $La_2Zr_2O_7$ and $SrZrO_3$ [6]. In addition, cathode particles in cathode active layer became excessive coarsening when undergoing the high temperature sintering[7]. These two conclusions were thought to be the most critical reasons resulted in the high polarization resistance and the low power density.

Recently, an effective method for enhancing SOFC cathode performance by surface modification based infiltration has been proposed[8]. The method was able to fabricate the nanostructured cathode at reduced temperature and increased the triple phase boundaries(TPBs) for oxygen reduction reaction(ORR)[9]. Liu et al. fabricated LSM cathode framework by dip-coating and ion-impregnated scandia-stabilized zirconia (SSZ) into the porous LSM matrix for anode-supported tubular SOFCs which showed the improved performance, 433mW·cm^{-2} at 850 °C [10]. Liu et al. reported that the catalytic activity of the $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$-$Ce_{0.9}Gd_{0.1}O_{1.95}$ (LSCF-GDC) cathode can be significantly improved by introducing the electrochemically catalytic Ag using an impregnation method, showing 1.05W·cm^{-2} at 550 °C [11,12]. However, Kim reported that fuel cell coated with perovskite

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cathode was more stable in the long operation time than other metal catalyst, such as Ag and Pt which increased polarization resistance due to metal particle sintering[13]. In the recent reports, $La_{0.6}Sr_{0.4}Fe_{0.9}Sc_{0.1}O_{3-\delta}$ (LSFSc) was approved to be a great promising SOFCs cathode material with high oxygen reduction catalytic activity [14,15]. Moreover, the high valence Sc cations occupied in B-site of the perovskite structure resulted in the good chemical stability[14].

In this work, we introduced a new sintering process to obtain a dense and porous SSZ electrolyte bi-layer structure on anode-supported tubular SOFCs. The anode-supported tubular SOFCs consisting of NiO-$Zr_{0.92}Y_{0.08}O_2$(8YSZ) anode supporting substrate, NiO-$Zr_{0.89}Sc_{0.1}Ce_{0.01}O_{2-x}$ (SSZ) anode active layer, dense SSZ electrolyte, porous SSZ layer and $(La_{0.8}Sr_{0.2})_{0.95}MnO_3$ (LSM95) current collection layer were fabricated by slip-casting, dip-coating, pre-sintering, co-sintering and infiltration techniques. The porous SSZ electrolyte which had largely specific surface areas worked as the active cathode after few times infiltrating of LSFSc catalyst. Functioning fuel cells were fabricated and electrochemically characterized to evaluate anode-supported tubular SOFCs with the composite cathode. In addition, a long term test was carried out to evaluate the stability of the new designed tubular SOFCs.

2. Experiments

2.1. Cell fabrication

Nickel oxide (NiO, INCO, Japan) powders and Commercial 8 mol % Y_2O_3 stabilized ZrO_2 (YSZ, Tosoh, Japan) powders were used as the anode supporting substrate. Commercial $Zr_{0.89}Sc_{0.1}Ce_{0.01}O_{2-x}$ (SSZ, Daiichi Kigenso Kagaku Kogyo, Japan) was adopted for electrolyte. A-site deficient $(La_{0.8}Sr_{0.2})_{0.95}MnO_3$ perovskite oxide powders synthesized by traditional solid-state reaction method were used as cathode current collection material [16,17].

As a comparison, we fabricated tubular fuel cells with the traditional cathode structure of NiO-YSZ|NiO-SSZ|SSZ|LSM95-

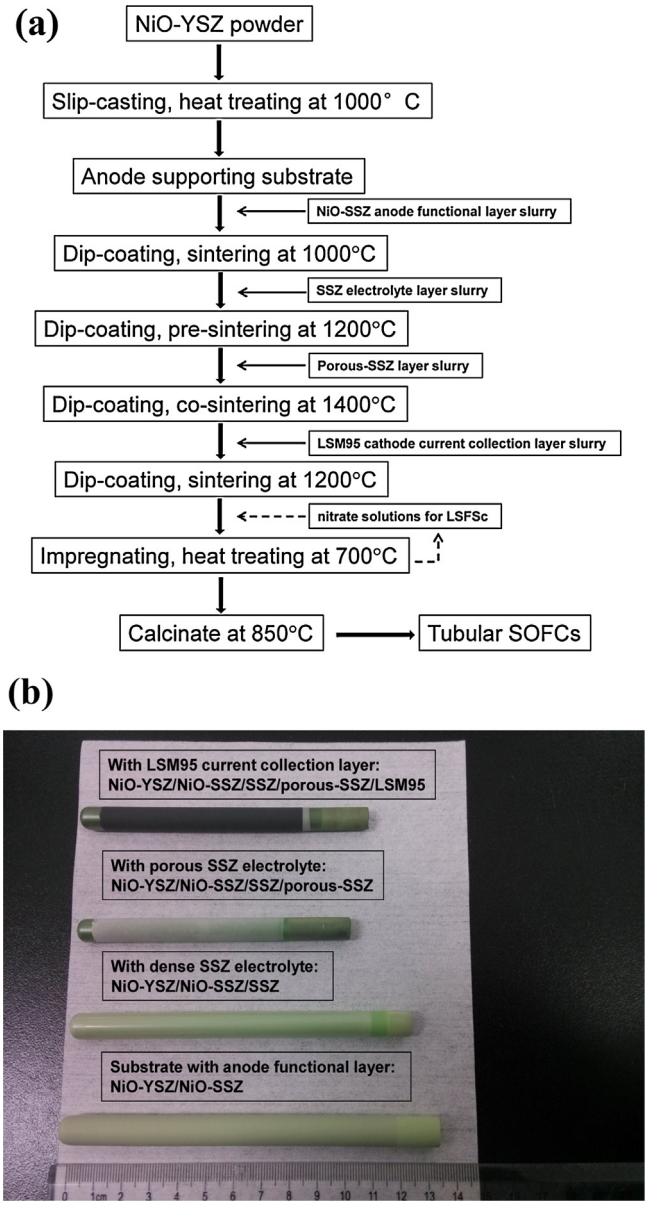

Fig. 1. (a) Fabrication processes of anode-supported tubular fuel cell-II (b) Sample photographs for every important fabricating step of cell-II.