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Graded porous solid oxide fuel cells fabricated by multilayer tape casting and co-firing progress

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ABSTRACT

Solid oxide fuel cells (SOFCs) with graded porous cathode were fabricated via a multilayer tape casting and co-firing technique, which is a low-cost and reproducible fabrication process. The effects of pore formers on the electrochemical performance of graded cathode are studied to minimize cathodic polarization resistance. Examination of the microstructures reveals that the multilayer single cell had no delamination, observable cracks, or other large defects. The cell prepared by the co-firing process exhibits low interfacial polarization resistance and high power density at the operating temperature range of 700–800 °C. This simple fabrication technique can be used for optimization of electrode microstructures and cost-effective fabrication of high-performance SOFCs.

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Introduction

Solid oxide fuel cells (SOFCs) have gained remarkable interest in recent years owing to their high efficiency, low emission, and fuel flexibility [1–3]. However, the cost of current SOFC systems is still prohibitive for many practical applications. To be economically competitive, both the cost of materials and the cost of fabrication for SOFC systems must be dramatically reduced [4,5]. One effective approach to cost reduction is to reduce the operating temperature of SOFCs. As a consequence, however, reduction of operating temperature considerably increased electrode polarization resistance, especially the resistance to oxygen reduction at the cathode. To address this problem, many studies have been conducted to develop new electrode materials or composite electrodes with high catalytic activity at relatively low temperatures [6]. Nevertheless, for a given electrode material, the electrode performance can be drastically altered by changing its microstructure, including pore and gain sizes, size distribution, and porosity.

Among various fabrication methods, tape casting is a well-established technique for fabricating large, thin, and flat ceramic tapes with a thickness range of 1–1000 μm. The scalability and longstanding success of the tape casting process makes it a legitimate technique for low-cost manufacturing of SOFC components, such as electrolyte membranes, anode supports, and interconnect layers [7–9]. Recently, tape casting and co-firing techniques have been applied to the fabrication of SOFC single cells and have proven to be scalable and cost-effective. A peak power density of 250 mW cm^{-2} at 900 °C has been obtained in an anode-supported cell with a LSM cathode YSZ electrolyte by screen printing and co-firing [10]. Wang et al. [8] fabricated Ni/ScSz

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anode-supported SOFCs by a multilayer tape casting and co-sintering technique. With the use of a LSCF/GDC composite cathode sintered at 1100 °C, they obtained a maximum power density of 0.85 W cm^{-2} and an area-specific polarization resistance of 0.336 Ω cm^2 at 850 °C. H. Moon [11] developed NiO/YSZ anode-supported SOFCs with an anode active layer via a tape casting and lamination technique. After pressing isostatically for 10 min, the half cell with the anode and electrolyte was sintered at 1350 °C for 3 h. The prepared SOFC with a LSM/YSZ cathode exhibited high power density and low cell resistance of 0.73 W cm^{-2} and 0.35 Ω cm^2, respectively, at 700 °C.

Materials in the (La, Sr) (Co, Fe)$O_{3-\delta}$ family have been extensively investigated and utilized as a cathode for SOFC due to their high ionic and electronic conductivity and good catalytic activity for the oxygen reduction reaction [12,13]. There have been various processes used for the preparation of porous LSCF electrode such as screen printing [14], dip coating [15] and spray painting [16]. To date, most studies on LSCF cathode reported in the literature focus on cathodes of uniform microstructures. In this communication, we report our findings on fabrication and characterization of porous $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ cathodes with graded microstructures fabricated by tape casting. The effect of the amount of pore formers on the performance of the graded cathode was determined. Meanwhile, an anode-supported SOFC cell was fabricated by tape casting and co-firing process to produce a cost-effective single cell that operates at intermediate temperatures. The power density and area specific resistance of the resulting SOFC were evaluated.

Experimental process

Cathode preparation

$La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ powders with varying particle sizes were prepared by citrate method [15] and gelcasting technique [17], respectively. The LSCF powders synthesized by the citrate method (S.A. 14.38 m^2 g^{-1}) and gelcasting (S.A. 8.07 m^2 g^{-1}) were used for active layer (inner layer) and central layer, respectively. For current collector (top layer), commercial $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ powders (S.A. 1.36 m^2 g^{-1}, Inframat Advanced Materials) were applied. Two pore formers were selected: graphite (Alfa Aesar) and corn starch (Sigma-Aldrich). The graphite and the starch were applied for inner layer and top layer and the amounts were from 5wt% to 20wt% and from 25wt% to 40wt%, respectively. For central layer, 15vol% of graphite and 15vol% of starch were added to the LSCF slips. In a first step, the LSCF powders and pore formers were ball milled for 12 h with dispersant in a mixture of xylene and ethanol to form a stable slurry. In a second step, two plasticizers and a commercial bonder were added to confer adequate flexibility and strength to the tapes, and left for further milling for another 12 h. The resulting homogeneous slurry was then cast under doctor blade. The inner layer was cast first on the carrier film and allowed to dry in air for several minutes, then the central layer and the top layer were prepared similarly. The thickness of each layer is 20 μm. After drying overnight at room temperature, the three layer green tape was cut into disks with diameter of 15 mm. For comparison, 60 μm thick single-layer LSCF cathodes of uniform microstructure were also fabricated using the same method.

Fabrication of symmetric cell and anode supported cell

In order to characterize the electrochemically behavior of these cathodes with different microstructures, electrolyte-supported symmetric cells with a configuration of LSCF|SDC|YSZ|SDC|LSCF were used. The YSZ electrolyte membranes were fabricated from 8 mol% Y_2O_3-doped ZrO_2 (Tosoh, Japan) using tape casting and sintering at 1400 °C for 5 h. To avoid the reaction between LSCF cathode and YSZ electrolyte, a $Sm_{0.2}Ce_{0.8}O_{1.95}$ (SDC) buffer layer was applied by drop coating SDC slurry on both sides of the YSZ pellets. The slurry was a mixture of the SDC powder and additives dispersed in an organic solvent. Then the LSCF green tapes were bonded to the buffer layer. After dried at 80 °C for 1 h, the symmetric cells were heated in a furnace at a rate of 3 °C/min up to 1080 °C.

The button cell in this study had Ni-YSZ anode supports, Ni-YSZ active layer, YSZ electrolyte, SDC buffer layer and graded LSCF cathodes (shown in Fig. 1). Electrolyte thin film

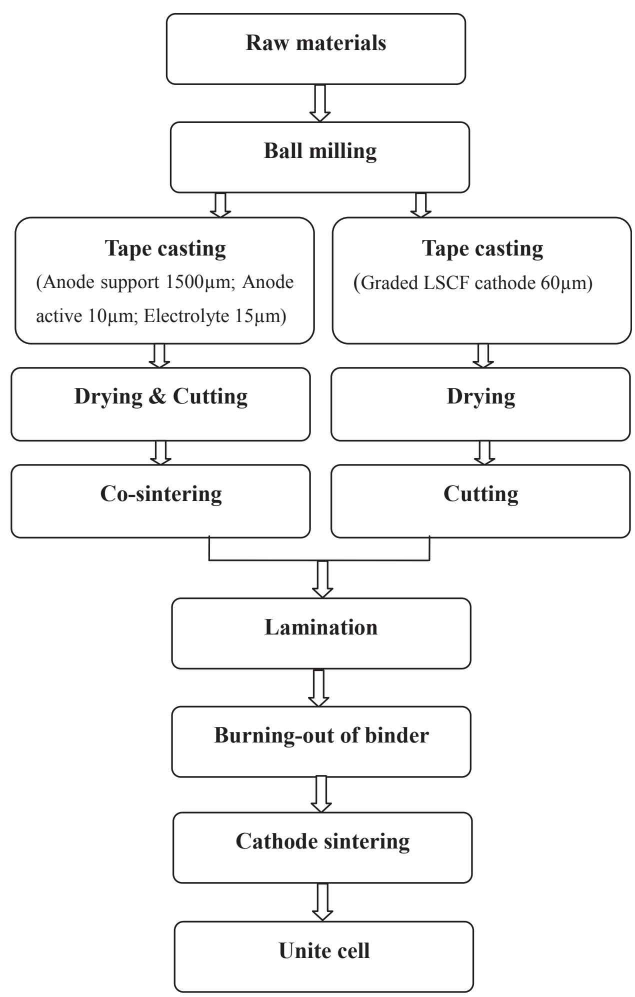

Fig. 1 – Flow chart of the manufacturing procedure for unit cell.

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