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Low temperature preparation and characterization of solid oxide fuel cells on FeCr-based alloy support by aerosol deposition

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ABSTRACT

Dense electrolyte and porous cathode coatings by room-temperature operating aerosol deposition process are applied and optimized for metal-supported solid oxide fuel cell fabrication. Porous metal support of FeCr-based alloy including (La, Sr)TiO₃ diffusion barrier and (Ce, Gd)O_{2-δ}-Ni anode were prepared using tape-casting process and co-fired in reducing atmosphere. Dense (Zr, Y)O_{2-δ} electrolyte and porous (La, Sr)(Co, Fe)O_{3-δ} cathode were prepared using aerosol deposition on it. The La_{0.2}Sr_{0.8}TiO₃ diffusion barrier effectively suppressed the reaction between the FeCr-based alloy support and Ni in the anode during co-firing at 1300 °C. Room-temperature deposition of the electrolyte and cathode layers in low vacuum conditions effectively prevented metal support degradation and cathode decomposition. Microstructural analysis of the anode, electrolyte, and cathode layers is presented. An open circuit voltage of 1.08 V and maximum power density of 0.71 W/cm² were achieved at 750 °C.

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Introduction

Solid oxide fuel cells (SOFCs) with porous metal support based on FeCr steel alloys are expected to have high potential for commercialization due to a number of factors, including low material cost, mechanical stability, and manufacturability benefits [1]. The main obstacles in SOFC fabrication on metal support are the limitations of sintering conditions for the high-temperature corrosion resistance of the metal support, decomposition of the cathode materials in inert or reducing

atmospheres, and the diffusion of Fe, Cr, and Ni between the ferritic FeCr steel and nickel-containing anode [2–4].

The corrosion problem of the metal support enforce lower-temperature sintering in a reducing atmosphere, which is not suitable for full densification of electrolyte materials such as (Zr, Y)O_{2-δ} (yttria stabilized zirconia, YSZ) and (Ce, Gd)O_{2-δ} (gadolinia doped ceria, GDC). Also, it limits the sintering of widely used cathode materials such as (La, Sr)Mn_{3-δ} (LSM) and (La, Sr)(Co, Fe)O_{3-δ} (LSCF) because sintering in a reducing atmosphere results in the development of unstable states and cation precipitation in those cathode materials, which causes

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significant degradation of the cell performance. To overcome these issues, several processing techniques have been proposed. One of them is a coating technique of thermal plasma spray on the porous FeCr-alloy support [5,6]. However, the techniques still face some challenges, such as difficulty in obtaining a thin and dense electrolyte, insufficient porosity of the electrode, thermal damage of thin metal supports, and high processing cost. Another technique is the infiltration of active materials into pre-sintered porous structure at relatively low temperature [7,8]. The SOFCs fabricated by this technique showed promising cell performance, even though the structural and electrochemical stabilities of the infiltrated nano-sized active electrode material remain in question.

Aerosol deposition (AD), a dense ceramic spray-coating process at room-temperature under low vacuum conditions was recently proposed as an alternative preparation technique for the electrolyte and cathode [9,10]. The process uses micron-sized ceramic particles mixed with a carrier gas in an aerosol flow that is accelerated by the pressure difference between the aerosol and the deposition chamber. The aerosol is ejected through a nozzle with high kinetic energy to collide with a substrate, forming a dense ceramic film without external heating. Dense YSZ, GDC, (La, Sr) (Ga, Mg)O_{3-δ} (LSGM) and porous LSCF layers prepared by this technique were successfully fabricated and verified for SOFC components [11–15]. The AD technique can fabricate thin, dense film (5–10 μm), which is hard to achieve in plasma spray-coating, and the porosity can be controlled in a wide range using ceramic-polymer composite powder [15]. In this study, the low temperature ceramic coating technique was adopted for fabrication of a dense electrolyte and porous cathode layer for high-performance SOFCs on a porous FeCr-based metal support.

Experimental procedure

A metal support including a diffusion barrier layer (DBL) and anode layer was fabricated by tape-casting, laminating, and co-firing in a reducing atmosphere. For metal tape fabrication, Crofer22APU powder (~36 μm, H.C. Starck) was mixed with a commercial binder system (B73225, Ferro). For preparation of the diffusion barrier layer, La_{0.2}Sr_{0.8}TiO₃ (LST) powder was made by mixing La₂O₃, SrCO₃, and TiO₂ (all reagent grade, Aldrich) raw powders in an appropriate ratio and calcining at 1200 °C for 6 h. For anode layer preparation, NiO (AFL grade, K ceracell) and GDC (ULSA grade, Rhodia) powders were mixed with a 7:3 weight ratio. The prepared powders were also mixed with a commercial binder for tape-casting slurry preparation using planetary milling machine with zirconia ball. Weight ratio of powder and commercial binder system for Crofer22-APU, LST, and NiO-GDC slurry fabrication was 10/3, 4/6, and 5/5 (powder/binder system), respectively. The thickness of the prepared tapes of the Crofer22APU, LST, and NiO-GDC layers were 120, 10, and 25 μm, respectively. 4 layers of Crofer22APU, 1 layer of LST, and 2 layers of NiO-GDC tapes were laminated and warm-pressed at 50 °C in a 25 mm diameter mold for 10 min with 5 ton. Binder burn-out of the laminated structure was conducted in ambient atmosphere at 500 °C for 2 h. Then, the sample was sintered in flowing H₂ conditions at 1300 °C for 2 h.

Electrolytes were deposited using commercially available YSZ (FYT13.0-005H, Unitec Ceramics) as a starting powder. The dried YSZ powder was mixed with carrier gas to form an aerosol flow in the aerosol chamber. The aerosol flow was transported through a tube to a nozzle before being accelerated and ejected into the deposition chamber through a rectangular nozzle with a 25 × 0.8 mm² orifice, which was evacuated by a rotary pump with a mechanical booster. Compressed air was dried through a dehumidifying filter and used as the carrier gas at a flow rate of 15 l/min. The pressures in the aerosol and deposition chambers were 600 and 1 torr, respectively. The accelerated YSZ particles collided with the prepared anode/DBL/metal support structure which was 5 mm from the nozzle. Cathode layers were then prepared on the YSZ film using commercially available LSCF powder (LSCF-S, K ceracell), which was heat-treated and mixed with polyvinylidene fluoride (PVDF, Aldrich) at a weight ratio of 19:1. The PVDF polymer decomposed during evaluation of the resulting SOFCs to form nano-sized porous structures [16]. The deposition equipment is described in detail elsewhere [10]. Fig. 1 summarizes the sample images of each preparation step for full cell fabrication. The cell diameter, thickness, and cathode area were 24.5 mm, 0.8 mm, and 1.0 cm², respectively.

The surface and fractured cross section microstructures of the films were examined by a scanning electron microscope (SEM, JSM-5800, JEOL) and scanning transmission electron microscope (STEM, JEM-2100F, JEOL) equipped with an energy dispersion X-ray (EDX) spectrometer. The gas permeability of the YSZ electrolyte was assessed using a digital flow meter (ADM2000, Agilent Technologies). For electrochemical testing of the cell, Pt mesh with small amounts of Pt paste attached as current collectors were employed at both the cathode and anode sides. A Pyrex ring was used for gas sealing between the electrolyte and the alumina tube of the sample holder. Air oxidant and dry H₂ fuel were used at flow rates of 1000 and 200 ccm, respectively. The cells' current–voltage (I–V) characteristics at 750 °C were recorded with a scan rate of 10 mA/s, and impedance spectra were measured using an electrochemical impedance spectroscopy analyzer (SP300, Biologic) over the frequency range of 30 mHz–1 MHz with a potentiostatic mode of 0.7 V.

Results and discussion

XRD patterns of the metal support (Crofer22APU), metal support/DBL/anode structure (Crofer22APU/LST/Ni-GDC), YSZ electrolyte deposited on Crofer22APU/LST/Ni-GDC structure, and LSCF-PVDF cathode deposited on YSZ/Crofer22APU/LST/Ni-GDC structure were shown in Fig. 2. The deposited YSZ film showed cubic fluorite structure with smaller and broader peaks and high FWHM (full width at half maximum) in their spectra due to the smaller crystallites that formed during deposition. The particle size of the YSZ film calculated by Scherrer's equation was ~10 nm. The LSCF film containing PVDF showed perovskite structure with sharp and smaller FWHM, and a particle size of >50 nm, probably because the deformation of PVDF polymer absorbed kinetic energy during collision and suppressed the fracture of the LSCF ceramic powder.

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