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Microchanneled anode supports of solid oxide fuel cells

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ABSTRACT

A novel anode with microchanneled structure has been prepared by a mesh-assisted phase-inversion process. The uniform microchannels templated by mesh apertures cross the anode, and act as efficient gas diffusion pathways, with one end open and the other end stopped with a less porous layer, which functions as a support for thin electrolyte film and provides anode reaction sites. The solid oxide fuel cell supported on the microchanneled anode produced the maximum power densities of 550, 772 and 936 mW/cm^2 at 750, 800 and 850 °C respectively, and no concentration polarization was found, suggesting there is efficient gas diffusion within the anode support.
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1. Introduction

Electrode-supported solid oxide fuel cells (SOFCs) with thin electrolyte films have widely replaced original electrolyte-supported SOFCs to achieve high power output via reducing the thickness of the electrolyte, which dominates the ohmic resistance of the SOFCs. However, gas (fuel or oxygen) diffusion resistance within the supporting electrodes causes concentration polarization resistance [1,2], which does not occur within thin electrodes of the electrolyte-supported SOFCs. To form a thin dense electrolyte layer on the supporting electrode, the two layers are co-sintered at high temperatures (up to 1400 °C), which results in a low porosity of electrodes, and therefore high gas diffusion resistances.

A phase-inversion process has been used to prepare ceramic membranes, and a hierarchically porous structure is formed during the phase-inversion: finger-like pores are embedded within the membranes, and sponge-like pores are distributed around the finger-like pores [3,4]. The finger-like pores act as gas diffusion channels where gas is delivered and distributed into the layer with the sponge-like pores, which acts as a functional layer, for example for catalytic reactions [5]. Compared with the pores templated by conventional pore former particles such as carbon spheres [6], the pores formed by the phase-inversion have better connectivity because the pore channels are formed by the convection of solvent and coagulant [7].

However, the finger-like pores are embedded within the ceramic membranes because a skin layer is formed at the surface during the phase-inversion [8]. The skin layer has a lower porosity compared with the other part of the ceramic, which might limit the gas diffusion into the porous ceramics and therefore cause concentration polarization [9]. Recently, we have invented a technology to remove the skin layer by using stainless steel mesh, and the mesh apertures also templated uniform microchannels, opening at one end and stopping with a layer at the other end [10]. In this study, novel microchanneled anode supports will be prepared by the method, and SOFCs with the microchanneled anodes will be tested in power generation from hydrogen fuel.

2. Experimental

2.1. Anode support preparation

The green bodies of anode supports were prepared as described in our previous study [10]. Typically, 2.46 g of Polyethersulfone (PESF, Radel-A300) and 0.21 g of Polyvinylpyrrolidone (PVP, MW = 40000, Sigma-Aldrich, Australia) were dissolved in 13.69 g of NMP (99%, Acros organics) to form a uniform solution. Then, the solution was mixed with 30 g of NiO/$Gd_{0.2}Ce_{0.8}O_2$ (GDC) power (Fuel Cell Materials, Ohio, USA) in a teflon jar for 48 h by a planetary ball miller. To form anode green bodies, the obtained slurry was cast into an alumina module to form disc-shape anodes, and stainless steel mesh with the aperture size of 45 μm was put just below the slurry surface. Water was applied on the top of slurry to induce phase-inversion. After the phase-inversion for 20 min, the mesh was lifted off to remove the skin layer. Finally, the anode green bodies were taken out from the module and rinsed with water to wash out NMP solvent, followed by drying at 100 °C in an oven.

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2.2. SOFC preparation and test

The green bodies of anode supports were pre-sintered at 1100 °C for 2 h to burn off the polymer and obtain a certain mechanical strength. Then, the bottom of anode supports (away from the channel side) were dipped into a $Sc_{0.1}Ce_{0.01}Zr_{0.89}O_2$ (Fuel Cell Materials, Ohio, USA) slurry using ethanol as a solvent to form a coating on the support, and the two layers were co-sintered at 1400 °C for 5 h to form a dense electrolyte film on the anode supports. Using ethanol as a solvent, GDC (Fuel Cell Materials, Ohio, USA) powder was mixed with $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_3$ powder prepared by a combustion method [11], and the slurry was sprayed on the electrolyte film to form a cathode layer after sintering at 950 °C for 2 h [12]. The final cells are about 0.9 mm in thickness and the cathode area is about 0.2 cm^2.

The fuel cells were tested with a home-made apparatus. The fuel cells were sealed on a stainless steel tube with ceramic adhesive (552-VFG, Aremco Products Inc., USA). Hydrogen was flowed into the tube as a fuel, and other side of the fuel cell was exposed to air as an oxidant. The fuel cell performances were tested with an electrochemical workstation (VersaSTAT4, Princeton Applied Research). The microstructure of the fuel cells was observed using a scanning electron microscope (Zeiss Neon 40EsB FIBSEM).

3. Results and discussions

To increase the porosity of the porous ceramics, fugitive pore formers are generally distributed into the green bodies of ceramics during ceramic formation and then burned off during ceramic sintering. Thereby, the pore formers template the pores within the ceramics. Nevertheless, it is inefficient in improving gas diffusion within porous ceramics. Pore formers such as graphite, polymer or carbon spheres and starch are particulate, and therefore generally template separated pores, as shown in Fig. 1a. The low pore connectivity restrains gas diffusion through these pores. Adding more pore former can improve the pore connectivity. However, it is not practical to prepare ceramic supports via slurry such as tape-casting and gel-casting because adding more pore formers into the slurry greatly increases slurry viscosity and makes it uncastable [13]. Moreover, the mechanical strength of the supports is decreased as the amount of pore former is increased.

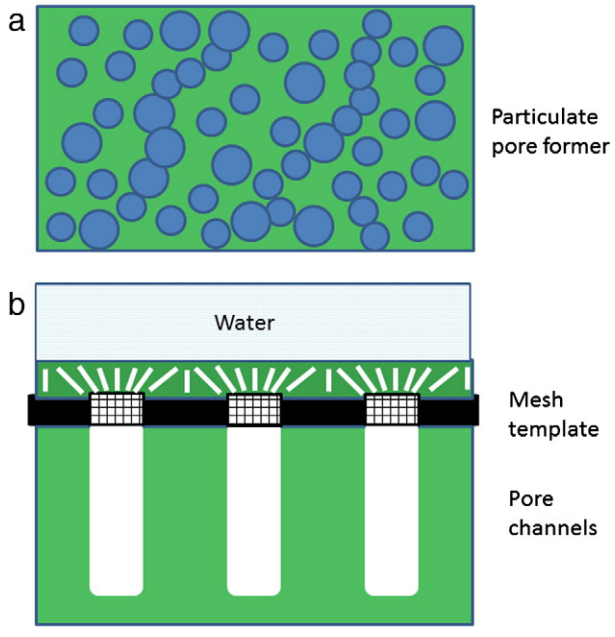

Fig. 1. Schematic representations of the formation of porous structures by (a) using conventional particulate pore former and (b) a mesh-assisted phase-inversion process.

Pore channels (instead of separated pores) are formed via the convection of solvent and coagulant during phase-inversion [7]. As shown in Fig. 1b, water (coagulant) is applied on top of the slurry, and small pore channels are formed when water diffuses into the slurry, driven by the miscibility between solvent and water. The pore channels grow as water and solvent continuously diffuse into the channels. When the small pore channels approach the mesh, they are combined into large channels. The large channels keep growing beyond the mesh aperture, and the growth is driven by the convection of solvent and water. Simultaneously, small pores (or sponge-like pores) within the channel wall are formed due to the slow convection of solvent and water. The pore channels will grow until no solvent is available to be extracted from the slurry, and stop before reaching the other side of the anodes. Thus, the microchanneled structure formed by phase-inversion will show higher efficiency in gas diffusion compared with the pore structure templated by pore formers in term of pore connectivity and pore tortuosity [14].

The untemplated phase-inversion process has been used in the preparation of SOFC anodes [2,4]. However, the less porous skin layer restrains the gas diffusion into the anodes. Chen et al. had identified the concentration polarization caused by the skin layer via comparing the cells with and without the skin layer [9]. Furthermore, the pore channels under the skin layer are irregular. The mesh plays a two-fold role in the formation of the new structure: removing the skin layer and templating pore channels. Fig. 2a shows the anode surface, and all pore channels are open for gas diffusion into them. From the trace of mesh wires, one mesh aperture templates one pore channel. Under the surface (see Fig. 2b), uniform channels with diameters around 30 μm cross the anode thickness and stop before reaching the other side of the anode, forming a layer with a conventional porous structure (approximately 100 μm in thickness). The less porous layer acts as a functional layer to support a thin electrolyte film and conducts electrochemical reactions within the anodes. Accordingly, gas can quickly diffuse through the long pore channels and distribute into the functional layer with a long triple-phase boundary [15]. The uniform open microchannels would facilitate the delivery of catalyst precursors to the function layer by the conventional impregnation process, which is not easy to achieve on conventional anode-supported SOFCs. For example, coking-resistant catalysts are delivered through the microchannels to the anode active sites for methane-fuelled SOFCs [16]. Therefore, the integrated structure produced by templating phase-inversion is attractive as a porous support of SOFCs.

As shown in Fig. 2c, a thin electrolyte film (about 8 μm in thickness) was successfully prepared by a simple dip-coating process. Numerous channels were soaked with a large amount of solvent during the dip-coating, which improved the coating efficiency. Moreover, the highly porous green body underwent a large sintering shrinkage (about 24%), which was beneficial for the densification of the electrolyte coating via sintering. There are two types of pore within the anode functional layer: small pores and large pores. Small pores are attributed to ceramic particle sintering and NiO reduction; large pores are formed during the phase-inversion.

The SOFC supported on the new anode was tested for power generation using hydrogen as a fuel. From Fig. 3, the maximum power densities are 550, 772 and 936 mW/cm^2 at 750, 800 and 850 °C with the electrode polarization resistances of 0.25, 0.12 and 0.06 Ω.cm^2 at open circuit voltage (OCV) respectively. The small electrode polarization resistances are attributed to the integrated anode structure. From the current–voltage curves, it is seen that no concentration polarization was found even when increasing the testing temperature to 900 °C. If concentration polarization does occur, the voltage will sharp decrease at high currents, which indicates cell performances are limited by concentration polarization.

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