



The beneficial effects of straight open large pores in the support on steam electrolysis performance of electrode-supported solid oxide electrolysis cell



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HIGHLIGHTS

- Electrode with straight pores were prepared by phase inversion tape casting method.
- Cathode-supported cell exhibited much improved steam electrolysis performance.
- High current density of 1.42 A cm^{-2} was achieved at 1.3 V, 750 °C and 50%RH.
- Straight pores in electrode could reduce concentration and activation polarization.

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ABSTRACT

This study is aimed at improving the electrochemical performance of electrode-supported solid oxide electrolysis cells (SOECs) by optimizing the pore structure of the supports. Two planar NiO–8 mol% yttria-stabilized zirconia supports are prepared, one by the phase-inversion tape casting, and the other by conventional tape casting method using graphite as the pore former. The former contains finger-like straight open large pores, while the latter contains randomly distributed and tortuous pores. The steam electrolysis of the cells with different microstructure cathode supports is measured. The cell supported on the cathode with straight pores shows a high current density of 1.42 A cm^{-2} and a H_2 production rate of $9.89 \text{ mL (STP) cm}^{-2} \text{ min}^{-1}$ at 1.3 V and 50 vol % humidity and 750 °C, while the cell supported on the cathode with tortuous pores shows a current density of only 0.91 A cm^{-2} and a H_2 production rate of $6.34 \text{ mL cm}^{-2} \text{ min}^{-1}$. It is concluded that the introduction of large straight open pores into the cathode support allows fast gas phase transport and thus minimizes the concentration polarization. Furthermore, the straight pores could provide better access to the reaction site (the electrode functional layer), thereby reducing the activation polarization as well.

1. Introduction

Global warming and limited availability of fossil fuels have stimulated intensive interest in hydrogen as a new carbon-free energy carrier. Currently, hydrogen is predominantly produced from steam reforming of methane or other hydrocarbons [1,2]. Transitioning to a low-carbon economy poses an urgent demand for an alternative route for hydrogen production that is sustainable and environmentally benign. Solid oxide electrolysis cells (SOECs), combining the heat and electricity from renewable energy or advanced nuclear reactors, allow for efficient production of hydrogen via steam electrolysis at high temperatures [3–7].

SOECs are reversely operated solid oxide fuel cells (SOFCs) [8–11]. In order to operate SOECs at intermediate temperatures ranges, an asymmetric configuration is commonly adopted in which a thin electrolyte layer is supported on a thick porous electrode [12–16]. The porous electrode support is typically prepared by using pyrolyzable pore formers such as graphite or starch. The pore former is introduced during the preparation of green body, and subsequently burned-out at elevated temperatures. The as-obtained pores are tortuous and may thus impose a large resistance to the gas phase mass transport [17–20]. Recently, it has been reported that porous electrode supports can be produced by the phase-inversion [21–25] or freeze-drying [26–28]

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method. The as-introduced pores are finger-like, and aligned along the thickness direction of the supports. The resulting SOFCs showed much improved electrochemical performance. In particular, no sign of concentration polarization was observed at high current densities [22,29,30].

Our previous study has shown that a new variant of tape casting method, which involves solidification of the slurry via phase inversion mechanism, could be successfully applied to fabrication of a Ni-YSZ anode comprising a thin anode functional layer and an open finger-like porous support layer [24]. Therefore, the activation resistance and the concentration resistance in the anode could be simultaneously decreased, and the SOFCs supported on the as-prepared composite anodes exhibited much improved electrochemical performance compared with the conventional anode-supported SOFCs [24]. The present study was intended to investigate the effects of the pore structure on the electrolysis performance of the electrode-supported SOECs. It was demonstrated that the cell comprising a support with the finger-like large pores exhibited much better steam electrolysis performance than the one comprising a support with randomly distributed and tortuous pores. The presence of the straight open pores in the electrode support is beneficial not only to mitigation of the concentration polarization, but also to reduction of the activation polarization.

2. Experiments

2.1. Preparation of SOECs

The cathode substrates were prepared by the modified phase-inversion co-tape casting with two types of slurries. The first slurry was composed of NiO (36.0 wt%) (NOVAMET, USA), 8 mol% yttria-stabilized zirconia (YSZ) (36.0 wt%) (Tosoh Corporation, Japan), *N*-methyl-2-pyrrolidone (23.1 wt%) (CP, Sinopharm Chemical Reagent Co.), polyethersulfone (PESf) (3.9 wt%) (RadelA-100, Solvay Advanced Polymers) and polyvinylpyrrolidone (PVP) (1.0 wt%) (K30, Sinopharm Chemical Reagent Co.). The second slurry consisted of NiO (37.8 wt%), 3mol% yttria doped tetragonal zirconia polycrystal (TZP) (25.2%wt%) (Tosoh Corporation, Japan), graphite (7.0 wt%) (Shanshan Technology Co.), NMP (24.8 wt%), PESf (4.2 wt%) and PVP (1.0 wt%). The two slurries were ball-milled for 24 h, degassed for 10 min, and then co-tape cast onto a Mylar sheet using a device as detailed in the previous studies [24], which were subsequently transferred into a water bath for 12 h to complete the solidification process. The as-formed green tapes comprised two layers: one free of graphite and the other containing graphite. The thickness of the graphite-free layer was varied by adjusting the blade gap. The green cathodes were punched into pellets with a diameter of about 18 mm, and dried in an oven at 80 °C for 24 h (coded as Cathode-1). For comparison, another cathode of the same composition was fabricated by using the conventional tape casting method with graphite as the pore former (coded as Cathode-2). Both cathodes were pre-fired at 1050 °C for 2 h. Thin films of YSZ (Tosoh, Japan) electrolytes were dip-coated onto the cathode surfaces. After co-sintered at 1400 °C for 5 h, A-site non-stoichiometric $(\text{La}_{0.8}\text{Sr}_{0.2})_{0.95}\text{MnO}_{3-\delta}$ (LSM)-YSZ anode slurry at a weight ratio of 50:50 was screen-printed onto the YSZ electrolytes, followed by firing at 1180 °C for 2 h. The cells supported on Cathode-1 and Cathode-2 were denoted as Cell-1 and Cell-2, respectively.

2.2. Characterization of cathodes and SOECs

The porosity and gas transport property of the as-obtained Ni-YSZ cathodes were measured. The cathodes were first sintered at 1400 °C for 5 h in air and then reduced at 800 °C for 2 h in hydrogen. The porosity was determined by the Archimedes method in water, and gas permeability was measured at varied pressures.

Fig. 1 shows the schematic diagram of the experimental set-up used in this study. The single cell was sealed to the alumina ceramic tube

using a conductive silver paste (DAD-87, Shanghai Synthetic Resin Institute). Another silver paste (Sino-platinum Metals CO., Ltd, China) was used as the current collector on both anode and cathode surfaces with Ag wire as the voltage and current leads. The anode was exposed to static air. Nitrogen was used as an inert carrier gas for purging air in the cathode. After reduction in hydrogen, steam was fed into the cathode side of the cell by bubbling hydrogen through a heated water bath. The hydrogen flow rate was held at 40 mL min⁻¹ through a mass flow controller. The current-voltage characteristics were measured from the open circuit voltage to 1.6 V with a scanning rate of 20 mV s⁻¹. AC impedance measurements were collected on the Solartron1287/1260 electrochemical system with an amplitude of 30 mV over the frequency range of 10⁶–10⁻¹ Hz. The microstructures of the cathodes and single cells were examined by SEM (JSM-6390LA, JEOL, Japan).

3. Results

3.1. Microstructure, porosity and gas permeability of cathodes

Fig. 2 presents the SEM micrographs of two cathodes that were sintered in air at 1400 °C and then reduced in hydrogen at 800 °C. Cathode-1 possessed an asymmetric two-layered structure with a total thickness of ~920 μm (Fig. 2a). The top layer had a thickness of ~20 μm, and was well adhered to the porous support (Fig. 2b). The support layer contained finger-like pores of several tens micrometer in diameter, as produced in the phase inversion step and survived through the sintering step; small pores were also present in the walls of the large pores as produced from the removal of graphite during the sintering step (Fig. 2c). The bottom surface (the surface of skin layer) contained micrometer-sized pores, which were also derived from the graphite pore former, revealing that the finger-like large pores were opened up to the outer surface (Fig. 2d). Fig. 2e–h are the SEM micrographs of Cathode-2 as fabricated by the conventional tape casting. The total thickness of Cathode-2 and the thickness of its top layer are almost the same as those of Cathode-1. Different from Cathode-1, the bulk and bottom surface of the support of Cathode-2 possessed a uniform and random pore structure with an average pore size of ~10 μm, as produced from the burn-out of graphite as the pore former (Fig. 2g and h). The porosity was measured to be 57.5% for Cathode-1 and 50.5% for Cathode-2.

Fig. 3 compares the gas permeation behavior of two cathodes as measured using N₂ at room temperature, showing a significantly higher gas permeation rate for Cathode-1 than for Cathode-2. In particular, the gas permeation rate at a pressure difference of 0.6 bar was 6.73 × 10⁵ Lm⁻²h⁻¹ for Cathode-1, which was about 3.3 times as high as that for Cathode-2. Although Cathode-1 exhibited a slightly higher porosity than Cathode-2, such a large difference in gas permeability should be ascribed to the presence of the finger-like large pores in Cathode-1 that were highly conducive to gas transport.

3.2. Microstructure and electrochemical performance of the cells

Fig. 4a and b shows the cross-sectional SEM micrographs of the single cells supported on Cathode-1 and Cathode-2. It can be seen that dense electrolytes well adhered to the cathode functional layer without any distinctive cracking or delamination. The electrolytes and the anodes were largely the same for both cells. The thickness was 13 μm for the electrolytes and 24 μm for the anodes.

Fig. 4c and d presents the curves of current-voltage (I-V) and the corresponding power density of the two cells tested in the fuel cell mode. The open circuit voltage (OCV) of the two cells was above 1.0 V, indicative of good gas tightness of the electrolyte and reasonable cell sealing. Cell-1 produced maximum power densities of 1200, 980 and 715 mW cm⁻² at 800, 750 and 700 °C, respectively. In contrast, Cell-2 produced much smaller maximum power densities of 904, 487 and

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