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Electrochemical performance of a new structured low temperature SOFC with BZY electrolyte

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ABSTRACT

A symmetrical solid oxide fuel cell (SOFC) with a novel microstructure of BaZr_{0.9}Y_{0.1}O_{3-δ} (BZY) as the electrolyte is investigated in this study. The cell with the Ni_{0.8}Co_{0.15}Al_{0.05}LiO₂ (NCAL)-foam Ni/BZY/foam Ni-NCAL structure is prepared by a co-pressing method. The maximum obtained power density is 735.6 mW cm⁻² in H₂ at 550 °C, which is comparable to the results obtained using Ni cermet anode-supported SOFCs with an extremely thin electrolyte. The ionic conductivity of the BZY electrolyte prepared in this study is much higher than that of the conventional BZY electrolyte. The activation energy of ionic conduction is much lower than that of traditional oxygen ion or proton conduction. Electrochemical impedance spectra (EIS) results of the cell with the BZY electrolyte measured in different atmospheric conditions and the results of oxygen ion filtration experiments for the cell using the BZY/Ce_{0.9}Gd_{0.1}O₂ (GDC) bilayer electrolyte indicate that oxygen ion is one of the carriers in the BZY electrolyte prepared in this study. According to the results of X-ray photoelectron spectroscopy (XPS) and Fourier Transform infrared spectroscopy (FTIR), an interfacial O²⁻ conduction mechanism at the interface of BZY particles in the electrolyte is discussed.

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Introduction

A solid oxide fuel cell (SOFC) is a device that directly converts chemical energy into electric power. An SOFC has the advantages of high energy conversion efficiency, environmentally friendly operation and large-scale elasticity, and is recognized as the green energy device of the 21st century. Reducing the SOFC operating temperature below 600 °C is an internationally recognized technical route for SOFC [1]. Low ionic or proton conductivity of the electrolytes at low

temperatures is one of the main factors limiting the electrochemical performance of SOFCs operated at low temperatures [2]. While thinning the electrolyte can solve the problem of the low ionic conductivity, this approach greatly increases the SOFC cost and has a negative effect on the long-term stability of the SOFC stack [2,3].

Alternative materials that show high conductivity at low temperatures or materials with new ionic/proton conduction mechanisms have been investigated [4–10]. Recently, the ionic interfacial diffusion phenomenon has attracted increasing attention [11–14]. Barriocanal et al. reported a

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colossal ionic conductivity at the interfaces of epitaxial $\text{ZrO}_2\text{:Y}_2\text{O}_3/\text{SrTiO}_3$ (YSZ/STO) heterostructures [11]. Lee et al. reported that the ionic conductivity of YSZ nanocolumns with 20 nm diameter strain coupled to an STO matrix with thick epitaxial nanocomposite films is two orders of magnitude more than that of the plain YSZ films [12]. Maier indicated that ionic transport properties are drastically changed at interfaces and may dominate the ionic conductivity of the entire material [13]. Unlike conventional ionic transport mechanisms, interfacial conduction mainly depends on the interface of the materials. Therefore, the dependence of the interfacial conduction on the electrolyte microstructure is different from that of the conventional electrolyte. While conventional electrolytes mainly rely on bulk diffusion and grain boundary diffusion, interfacial conduction of the electrolyte requires the presence of an interface that provides ionic conduction channels [15–18]. Therefore, the interfacial conduction electrolyte is generally not treated by very high temperature densification. The interfacial conduction phenomenon has been demonstrated in many low-temperature SOFCs (LTSOFCs) [15–18]. Many materials with very poor bulk ionic conductivity or with high electronic conductivity were used as electrolytes in interfacial conduction SOFCs, and remarkable electrochemical performances were obtained at low temperatures [15–18].

High-temperature proton conductors (HTPCs) have attracted increasing attention for intermediate-temperature applications (300–600 °C) because they exhibit high conductivities at low temperatures [4–7]. In the family of HTPC materials, BaCeO_3 - and BaZrO_3 -based materials show the best proton conduction characteristics. Y-doped barium cerate (BCY) electrolyte shows the highest conductivity among all HTPCs at low temperatures (10^{-2} S cm^{-1} at 600 °C), but it is easy to react with CO_2 and water vapour [1,5]. Compared with BCY, yttrium-doped barium zirconate (BZY) shows better bulk proton conductivity and chemical stability in fuel cell operation conditions [7,19]. It has been reported that the bulk proton conductivity of BZY exceeds the conductivity of the best oxide ion conductors at temperatures below approximately 700 °C [19]. The proton conductivity of BZY is generally reported to be between 10^{-3} S cm^{-1} and 10^{-2} S cm^{-1} at 600 °C [20,21]. The range of the conductivity values is mainly due to the contribution of the grain boundary conductivity of BZY, which is significantly affected by the sintering characteristics of the BZY pellet [22,23].

Nevertheless, the 10^{-2} S cm^{-1} conductivity of the BZY electrolyte below 600 °C cannot meet the requirements of LTSOFCs. For the cell with traditional sintered BZY electrolyte, the electrolyte must be very thin, and the cell can achieve good electrochemical performance. The thin film process of electrolytes will significantly increase the cost of cell preparation. In this study, $\text{BaZr}_{0.9}\text{Y}_{0.1}\text{O}_{3-\delta}$ (BZY) was used as the electrolyte to construct an SOFC with interfacial conduction. A low cost co-press method was used to prepare a symmetric cell with BZY as electrolyte. The electrochemical performance of the symmetrical cell composed of Ni foam pasted by $\text{Ni}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{LiO}_2$ (NCAL) electrodes and BZY electrolyte with novel structure was investigated, and the ionic conduction mechanism in such BZY electrolyte was discussed.

Experimental

$\text{BaZr}_{0.9}\text{Y}_{0.1}\text{O}_{3-\delta}$ (BZY) powders were synthesized by a sol-gel method [22]. $\text{Ba}(\text{NO}_3)_2$ (99.5%), $\text{Zr}(\text{NO}_3)_4$ (99.99%), $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99%), $\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$ (99.5%), and $\text{C}_{10}\text{H}_{16}\text{N}_2\text{O}_8$ (99.5%) were used as precursors in the synthesis. The details of the method are as follows: First, $\text{Ba}(\text{NO}_3)_2$, $\text{Zr}(\text{NO}_3)_4$, and $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ were dissolved in deionized water according to the stoichiometric ratio and stirred until they were dissolved and mixed evenly. $\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$ and $\text{C}_{10}\text{H}_{16}\text{N}_2\text{O}_8$ were added to the solution according to the mole proportions of 1:1.5:1 of metal cation/citric acid/EDTA acid. $\text{NH}_3 \cdot \text{H}_2\text{O}$ was used to adjust the pH to approximately 9, and the solution became clear. Then, after the solution was stirred for 14 h and then continuously stirred and heated for several hours to evaporate the water, a dark gel was obtained. The gels were calcined at either 1000 or 1200 °C in air for 5 h to form BZY powder. NCAL powder was purchased from Tianjin Bamo Science & Technology Joint Stock Ltd., China, for use as electrode material. Ni foam with thickness of 2 mm was used as the current connector and cell support. The BZY phase structure was characterized by an advanced X-ray diffractometer (SMART LAB 9 kW, Rigaku), using $\text{Cu K}\alpha$ radiation ($\lambda = 1.54060$ Å). The angular range of 2θ is between 20° and 90°, and the scanning rate was 10°min^{-1} . The cross-sectional structural morphologies of the cell were measured by an SSX-550 field emission scanning electron microscope (FE-SEM, Japan). The surface compositions and the oxidation state of the BZY powders before and after the performance test were analyzed by FTIR absorption spectra and X-ray photoelectron spectroscopy (XPS). The XPS spectra were recorded on a Thermo Scientific ESCALAB 250Xi XPS system with a monochromatic Al/Mg binode X-ray source. The bonding energy and oxidation state of the oxygen can be obtained by high-resolution scanning. FTIR spectra were recorded at room temperature in the mid-IR range ($400\text{--}4000 \text{ cm}^{-1}$) by the Nicolet-380 Fourier transform infrared spectrometer from Thermo, USA.

Commercial NCAL powder was added into glycerol and ethanol and stirred to make a slurry. The slurry was pasted on one side of Ni foam and desiccated at 120 °C for 20 min. The Ni foam with NCAL was cut into a circular pellet with a diameter of 13 mm. The BZY powder was sandwiched between two Ni foam-NCAL pellets and compressed at 360 MPa into a single cell with diameter of 13 mm. The effective area of the as-prepared symmetrical cell Ni-NCAL/BZY/NCAL-Ni was 0.64 cm^2 . Electrochemical performance of the cell was tested with H_2 fuel and air oxidant. The flow rate of H_2 and air were both 150 ml min^{-1} . The impedance of the cells was measured between 0.1 Hz and 1 MHz using a Princeton electrochemical system (VersaSTAT 3), an electrochemical workstation with AC voltage with amplitude of 10 mV under open-circuit conditions. The IV curves of the cells were measured using computerized instruments (IT8511 + 120 V/30 A/150 W).

Results and discussion

Fig. 1 shows the XRD patterns of the as-prepared BZY powder samples sintered at either 1000 or 1200 °C for 5 h. The

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