



# Improving the electrochemical properties of SSZ electrolyte-supported solid oxide fuel cells

He Miao\*, Guanghui Liu, Yi Zhang, Changrong He, Yifeng Zheng, Wei Guo Wang\*,<sup>1</sup>

Division of Fuel Cell and Energy Technology, Ningbo Institute of Materials Technology & Engineering, Chinese Academy of Sciences, Ningbo 315201, PR China

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## Abstract

Electrolyte-supported solid oxide fuel cells (NiO-1Ce10ScZr11Ce10ScZr1LSM-1Ce10ScZr) were prepared in this study. The effects of co-sintering temperatures of cathode and anode with electrolyte and the additions of metal oxide ( $\text{Bi}_2\text{O}_3$  and  $\text{CeO}_2$ ) in the anode on the microstructures and electrochemical properties were investigated. The carbon deposition behaviors of the anode with added  $\text{CeO}_2$  were also evaluated. Results indicated that the maximum power density ( $P_{\text{max}}$ ) initially increased from  $0.28 \text{ W/cm}^2$  ( $1150 \text{ }^\circ\text{C}$ ) to  $0.35 \text{ W/cm}^2$  ( $1250 \text{ }^\circ\text{C}$ ), and then decreased to  $0.33 \text{ W/cm}^2$  ( $1300 \text{ }^\circ\text{C}$ ) with the increase in co-sintering temperature from  $1150 \text{ }^\circ\text{C}$  to  $1300 \text{ }^\circ\text{C}$ . The variation tendency of  $P_{\text{max}}$  was attributed to a complex combination of the bond strength increase of the interfaces and the densification of the electrodes with the increase of the co-sintering temperature. Adding proper  $\text{CeO}_2$  in the anode effectively improved the electrochemical properties of the  $\text{ZrO}_2$  doped with  $\text{Sc}_2\text{O}_3$  electrolyte-supported single cells. This phenomenon could be related to the increase of the dimension of three-phase boundary. The  $P_{\text{max}}$  of the SSZ electrolyte-supported single cell with the anode mixed with 1.0 wt%  $\text{CeO}_2$  could reach  $0.42 \text{ W/cm}^2$ . The mixture of 1.0 wt%  $\text{CeO}_2$  in the anode could also effectively suppress the carbon deposition of  $\text{CH}_4$  on the anode surface.

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## 1. Introduction

Current research efforts are focused on solid oxide fuel cells (SOFCs) because of their high efficiency and significant environmental benefits [1,2]. Among the various cell structures, the electrolyte-supported SOFC (ESC) is well known because of its high stability during thermal cycle and oxidation–reduction cycle [3]. The electrolyte material for ESC should have pure ionic conductivity and chemical stability in oxidizing and reducing atmospheres. The 8.0 mol% yttrium-stabilized zirconia (8YSZ) is the most commonly used electrolyte for ESC [4]. However, the ionic conductivity of

this ceramic is not sufficiently high for utilization at operating temperatures.

Compared with YSZ electrolyte,  $\text{ZrO}_2$  doped with  $\text{Sc}_2\text{O}_3$  (SSZ) electrolyte displays higher oxygen ionic conductivity, but these two electrolytes show good chemical compatibility and similar thermal expansion coefficients [5–7]. The cubic phase in the SSZ system has the highest ionic conductivity, so it is suitable for use as a solid electrolyte for SOFCs [8]. However, the cubic phase is not stabilized at lower temperatures (below  $650 \text{ }^\circ\text{C}$ ), which has been causing an abrupt decrease in ionic conductivity during cooling in this SSZ system [9–11]. The SSZ system has the highest ionic conductivity among the zirconia-based solid electrolytes, but it cannot be extensively adopted because of its undesirable phase instabilities.

To suppress an undesirable phase transition, most studies have focused on the doping level of the SSZ system from 8 mol% to 12 mol%  $\text{Sc}_2\text{O}_3$ , where the cubic phase is the main phase. Additional dopants, such as  $\text{Al}_2\text{O}_3$  [8,12],  $\text{Yb}_2\text{O}_3$  [13],

\*Corresponding authors. Tel.: +86 574 8668 5097; fax: +86 574 8668 5702.

E-mail addresses: [miaoh@nimte.ac.cn](mailto:miaoh@nimte.ac.cn) (H. Miao), [wgwang@nimte.ac.cn](mailto:wgwang@nimte.ac.cn) (W.G. Wang).

<sup>1</sup>Tel.: +86 574 8791 8363; fax: +86 574 8668 5702.

and  $\text{CeO}_2$  [5,9,11], are effective in reducing or prohibiting this undesirable phase transition. Based on the overall investigations of the electrical properties and long-term stability of SSZ system materials,  $\text{ZrO}_2$  co-doped with 1 mol%  $\text{CeO}_2$  and 10 mol%  $\text{Sc}_2\text{O}_3$  (1Ce10ScZr) is an optimal candidate electrolyte material for SOFCs. However, few reports on full cells with 1Ce10ScZr electrolyte are available.

In the present study, electrolyte-supported SOFCs (NiO-1Ce10ScZr/1Ce10ScZr/LSM-1Ce10ScZr) were developed. The effects of the co-sintering temperatures of the cathode and anode with electrolyte and the additions of metal oxide ( $\text{Bi}_2\text{O}_3$  and  $\text{CeO}_2$ ) in the anode on the microstructures and electrochemical properties were discussed.

## 2. Materials and methods

The original powders of 1Ce10ScZr (99.9%;  $D_{50}=0.5 \mu\text{m}$ ) and NiO (99.9%;  $D_{50}=0.8 \mu\text{m}$ ) were purchased from Qingdao Tianrao Industry Co. Ltd., China and Tongxiang Tianyuan Chemical Co., Ltd., China, respectively.  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Mn}_{1.1}\text{O}_{3-\delta}$  (LSM) was synthesized through combined citric acid and EDTA complexing method and calcined at  $800^\circ\text{C}$  for 5 h to obtain a pure perovskite phase. The starting materials consisted of lanthanum(III) nitrate [ $\text{La}(\text{NO}_3)_3$ ; 99.5%], strontium nitrate [ $\text{Sr}(\text{NO}_3)_2$ ; 99.5%], manganese nitrate [ $\text{Mn}(\text{NO}_3)_4$ ; 99.5%], acrylic acid (99.5%), and ethylenediamine tetraacetic acid (99.5%), which were supplied by Sinopharm Chemical Reagent Co. Ltd., Shanghai, China. The metal oxide additives of  $\text{CeO}_2$  ( $D_{50}=20 \text{ nm}$ ; 99.99%) and  $\text{Bi}_2\text{O}_3$  ( $D_{50}=20 \text{ nm}$ ; 99.99%) in the anode were obtained from Aladdin Chemical Reagent Co., Ltd., Shanghai, China.

1Ce10ScZr powder (0.7 g) was pressed into pellets with diameter of 22.5 mm and thickness of 0.65 mm. The pellets were sintered at  $1500^\circ\text{C}$  in an open air for 5 h. NiO and 1Ce10ScZr powders with weight ratio of 1:1 were mixed by ball milling. Proper amount of binder and solvent were added into the mixed powder to make anode slurry. The slurry was silkscreened on one side of the 1Ce10ScZr electrolyte pellets. Similarly, LSM and 1Ce10ScZr powders were mixed by ball milling with weight ratio of 1:1 to make the cathode slurry. The cathode slurry was silkscreened on the other side of the

electrolytes. Full cells were then fired at a temperature of  $1150^\circ\text{C}$  to  $1300^\circ\text{C}$  for 2 h. The cathode active area was  $1 \text{ cm}^2$  after sintering. To investigate the effects of oxide additives on the microstructures and electrochemical properties of the cells,  $\text{CeO}_2$  and  $\text{Bi}_2\text{O}_3$  powders with designated substitution ratios for NiO were added in the anode. The microstructures of the cells were investigated using FESEM (Hitachi S-4800).

An SOFC single-cell pellet was attached on the ceramic tube. Silver wires with a diameter of 0.5 mm were attached to the anode and cathode as current leading wires. Hydrogen with 3%  $\text{H}_2\text{O}$  was fed through the ceramic tube to the anode side of the SOFC single cell with a flow rate of 100 sccm. Air was used as the oxidant to the cathode side with a flow rate of 300 sccm. The  $I$ - $V$  properties of the single cells were measured using Solartron SI 1287 electrochemical interface at  $800^\circ\text{C}$ . Electrochemical impedance spectra (EIS) measurements were carried out using the electrochemical interface with a 1260B frequency response analyzer at  $800^\circ\text{C}$ . The EIS of the full cells were obtained in a frequency range of 100 kHz to 10 mHz, with an AC amplitude of 10 mV under open circuit condition.

To measure the carbon deposition behaviors of the anode, anode slurries with different percentages of  $\text{CeO}_2$  additives were silkscreened and fired on the 1Ce10ScZr electrolyte via the same method. The half cells were then exposed to  $\text{CH}_4$  atmosphere for 30 min with a flow rate of 25 sccm. Then, they were cooled to room temperature with  $\text{N}_2$  protection. The

Table 1  
Electrochemical properties of SSZ electrolyte-supported SOFCs at  $800^\circ\text{C}$ .

ESCs	$P_{\text{max}}$ ( $\text{W}/\text{cm}^2$ )	ASR ( $\Omega \text{ cm}^2$ )	$R_s$ ( $\Omega$ )	$R_p$ ( $\Omega$ )	$R_p-R_s$ ( $\Omega$ )
Co-sintered at $1150^\circ\text{C}$	0.28	1.61	0.8	2.23	1.43
Co-sintered at $1200^\circ\text{C}$	0.31	1.28	/	/	/
Co-sintered at $1250^\circ\text{C}$	0.35	1.09	0.6	2.05	1.45
Co-sintered at $1300^\circ\text{C}$	0.33	1.13	0.5	2.18	1.68
Added with 0.5% $\text{CeO}_2$	0.36	0.95	0.57	1.95	1.38
Added with 1.0% $\text{CeO}_2$	0.42	0.88	0.55	1.24	0.69
Added with 1.5% $\text{CeO}_2$	0.37	0.96	0.55	1.4	0.85
Added with 1.0% $\text{Bi}_2\text{O}_3$	0.29	1.43	0.68	3.4	2.72

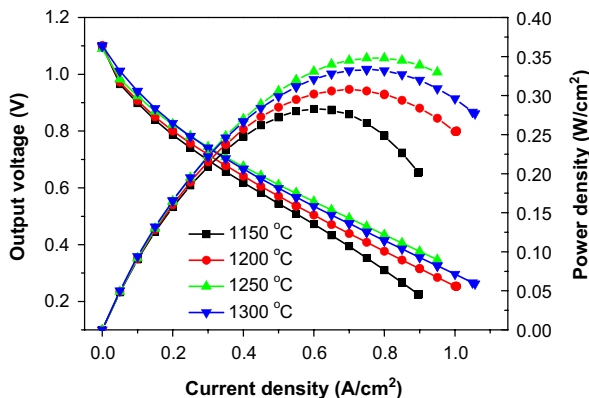


Fig. 1.  $I$ - $V$  and  $I$ - $P$  curves of SSZ electrolyte-supported SOFCs with anode and cathode co-sintered with electrolyte at temperatures of  $1150^\circ\text{C}$  to  $1300^\circ\text{C}$ .

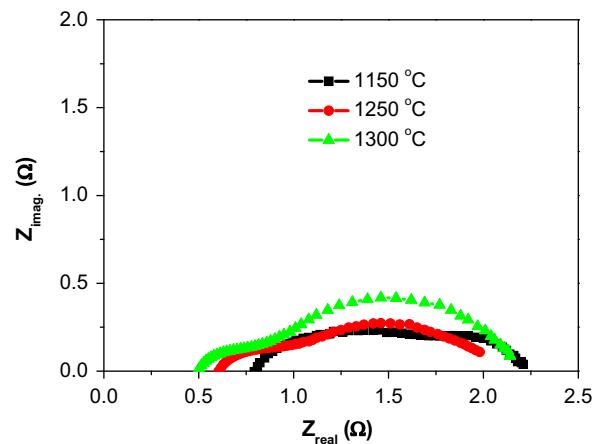


Fig. 2. EIS curves of SSZ electrolyte-supported SOFCs with anode and cathode co-sintered with electrolyte at  $1150^\circ\text{C}$  to  $1300^\circ\text{C}$ .

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