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Promoted electrochemical performance of intermediate temperature solid oxide fuel cells with $Pd_{0.95}Mn_{0.05}O$ -infiltrated $(La_{0.8}Sr_{0.2})_{0.95}MnO_{3-\delta}-Y_{0.16}Zr_{0.84}O_2$ composite cathodes



Ao Wang ^a, Jian Pu ^{a, *}, Dong Yan ^a, Nan-Qi Duan ^a, Yuan Tan ^a, Lichao Jia ^a, Bo Chi ^a, Jian Li ^{a, b}

^a Center for Fuel Cell Innovation, State Key Laboratory of Material Processing and Die & Mould Technology, School of Materials Science and Engineering, Huazhong University of Science and Technology, Wuhan, Hubei 431074, China

^b Research Institute of Huazhong University of Science and Technology in Shenzhen, Shenzhen 518057, China

HIGHLIGHTS

• Pd_{0.95}Mn_{0.05}O-infiltrated LSM-YSZ cathode was applied in large size SOFC.

• Electrochemical performance of infiltrated cell was significantly improved.

• Thermal cyclicability and long-term stability was evaluated at 750 °C.

• The relativity between particle growth and performance degradation was discussed.

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ABSTRACT

 $Pd_{0.95}Mn_{0.05}O$ -infiltrated ($La_{0.8}Sr_{0.2}$)_{0.95} $MnO_{3-\delta}$ -8 mol.% Y_2O_3 stabilized ZrO_2 (LSM-YSZ) cathode is used to large size ($11 \times 11 \times 0.1$ cm) Ni-YSZ anode-supported planar cells for the first time and electrochemically evaluated in the intermediate temperature range from 650 to 800 °C with H₂ as the fuel and air as the oxidant. The initial open circuit voltage (OCV) of the cell is 1.15 V, and the achieved maximum power density increases from 328 to 734 mW cm⁻² with the increase of testing temperatures from 600 to 800 °C, which is almost 2.6 times higher than that of the cell with conventional LSM-YSZ cathode. After each thermal cycle between 750 and 300 °C, the OCV remains almost unchanged and the cell voltage decreases less than 0.007 V, indicating that the cell is capable of thermal cycling. The cell voltage at 310 mA cm⁻² and 750 °C declines linearly with testing time at a rate of 2.6 $\times 10^{-4}$ V h⁻¹ for the growth of the infiltrated $Pd_{0.95}Mn_{0.05}O$ size, resulting in reduction of the total surface area of the particles. The mechanism of performance degradation of the cell with $Pd_{0.95}Mn_{0.05}O$ -infiltrated LSM-YSZ composites cathode is discussed in detail.

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1. Introduction

Intermediate-temperature (600–800 °C) anode-supported solid oxide fuel cell (IT-SOFC) has received intensive attention due to the particular advantages of high efficiency of energy conversion, low emission of polluting substances, low cost of fabrication processes, and wide selection range of materials and fuels [1,2]. However, it is facing critical challenge on cathode materials for the oxygen

* Corresponding author. E-mail address: pujian@hust.edu.cn (J. Pu).

http://dx.doi.org/10.1016/j.jpowsour.2015.10.110 0378-7753/© 2015 Elsevier B.V. All rights reserved. reduction reaction (ORR) at intermediate temperatures. Sr-doped LaMnO₃ (LSM) is the state-of-the-art cathode material for hightemperature SOFCs. At temperatures near 1000 °C LSM possesses outstanding ORR catalytic activity, electronic conductivity, structural stability and compatibility with most widely used electrolyte Y_2O_3 stabilized ZrO₂ (YSZ) [3]; nevertheless, its catalytic activity reduces significantly in the intermediate temperature range because of the negligibly low oxygen ion conductivity [4,5]. It was reported that the power density of a button cell with LSM-YSZ composite cathode was around 200 mW cm⁻² at 750 °C with H₂ as the fuel and air as the oxidant, and the polarization resistance was 3.25 Ω cm² at 750 °C [6].

In previously studies, Pd has been proved to be an effective modifier to improve the electrochemical performance of cathodes in SOFCs [6–13]. The Pd-modified LSM-YSZ cathode by solution infiltration demonstrated a polarization resistance of 0.09 Ω cm² at 750 °C [9], which is significantly lower than those for the cathodes of LSM-YSZ (2.3 Ω cm² [9]), Gd-doped CeO2 (GDC)-infiltrated LSM $(0.11 \Omega \text{ cm}^2 \text{ [14]})$, LSM-GDC $(0.49 \Omega \text{ cm}^2 \text{ [15]})$ and single-phase LSCF $(0.1 \Omega \text{ cm}^2 \text{ [16]})$ at the same temperature; and a high power density of 1420 mW cm^{-2} was achieved [6]. Nevertheless, a significant growth of infiltrated Pd nanoparticles was observed possibly due to the decomposition of PdO to Pd under the testing condition [17,18], which raises the concerns for the performance durability of the cathode. Alloying elements, such as Mn [8,11,19,20], Co [8,11] and Ag [11], were used to stabilize the particle size of the infiltrated Pd and thus the cathode performance, without sacrificing the catalytic activity of Pd. Element alloying may cause chemical shift of the dband and decrease in the Fermi level, which weakens the adsorption of atomic oxygen on the surface sites of Pd to promote the ORR [21]. Alloyed by Mn, the particle size of Pd_{0.95}Mn_{0.05}O infiltrated into YSZ scaffold was stabilized at 750 °C in air, which resulted in unchanged low cathode overpotential of ~72 mV for at least 30 h [19].

It is noted that so far the performance enhancement of LSM-YSZ cathodes by Pd modifications has been achieved with small button cells as the vehicle, and the effectiveness of this approach needs to be further proved in large cells practically used for power generation. Therefore in the present study, $Pd_{0.95}Mn_{0.05}O$ -infiltrated LSM-YSZ composite cathode was employed for the first time in Ni-YSZ anode-supported cells with a dimension of $11 \times 11 \times 0.1$ cm and an active area of 9×9 cm², and investigated on the aspects of microstructure and electrochemical performance at the intermediate temperatures. Meanwhile, in order to demonstrate the improvement, a conventional Ni-YSZ anode-supported planar cell with LSM-YSZ composite cathode was evaluated comparatively under the same conditions.

2. Experimental

The anode-supported cells were fabricated by the tape castingscreen printing-firing process. Mixed powder consisting of NiO (Inco) and YSZ (Tosoh) at 57:43 mass ratio was ball-milled for 24 h with toluene/ethanol (Sinopharm Chemical Reagent, Inc.) as the solvent, fish oil (Richard Mistler, Inc.) as the dispersant and cornstarch (WuHan JinBao Food, Inc.) as the pore former. Polyvinyl butyral (Soultia, Inc.) and polyethylene glycol (Sinopharm Chemical Reagent, Inc.) were added then as the binder and plasticizer, respectively, followed by another 24 h ball milling. Such prepared homogeneous slurry was tape cast into sheet by using a tape casting machine (LY-500, Beijing Oriental Sun System Technology, Inc.). The sheet was dried for 48 h in air at room temperature, cut into the size of 13.8×13.8 cm and fully screen printed on one side consecutively with NiO-YSZ and YSZ pastes as the functional anode and electrolyte, respectively, before fired in air at 1390 °C for 3 h under the load of a porous ZrO₂ plate. Cathode paste, composed of $(La_{0.8}Sr_{0.2})_{0.95}MnO_{3-\delta}$ (LSM, Fuel Cell Materials) and YSZ at 50:50 weight ratio, was screen printed on the top of the fired YSZ electrolyte and fired at 1200 °C in air for 2 h to form a porous LSM-YSZ composite cathode, into which $Pd_{0.95}Mn_{0.05}O$ solution [19] was infiltrated using an in-house designed vacuumed vessel shown in Fig. 1 at 45 °C and 0.08 MPa, followed by calcination at 750 °C in air for 2 h to complete the process of cell fabrication. The loading of Pd_{0.95}Mn_{0.05}O was controlled to 15 wt.% of the cathode by repeated infiltrations. The final cell size was 11 \times 11 \times 0.1 cm and active cathode area was 9×9 cm². For comparison, a cell without infiltrating Pd_{0.95}Mn_{0.05}O into LSM-YSZ composite cathode was also



Fig. 1. Schematic drawing of in-house designed vacuumed vessel.

prepared. The cell without $Pd_{0.95}Mn_{0.05}O$ infiltration into LSM-YSZ composite cathode was designated as Cell-I, and that with the infiltration was designated as Cell-II.

Both Cell-I and Cell-II were tested at 750 °C using a stainless steel testing jig [22] in an SOFC testing station (SF-30, Wuhan Li Xing Testing, Inc) with H_2 as the fuel and ambient air as the oxidant at a flow rate of 2 L min⁻¹ to the anode and cathode side. The current density (I)–voltage (V)–power density (P) curves, electrochemical impedance, thermal cycling ability and performance durability were evaluated at the intermediate temperatures. X-ray diffraction (XRD, X'Pert PRO) and scanning electron microscopy (SEM, FEI Quanta200) were employed for cathode phase identification and cell microstructure examination.

3. Result and discussion

Fig. 2 shows the appearance of the anode-supported planar cell with a dimension of 11 \times 11 \times 0.1 cm and an active area of 9 \times 9 cm². The cross-sectional microstructure observation, as shown in Fig. 3, indicated that the functional anode, electrolyte and cathode were intimately contacted each other with a thickness of approximately 10–11 μ m (Fig. 3a) and the infiltrated Pd_{0.95}Mn_{0.05}O particles were uniformly distributed on the scaffold of LSM-YSZ (Fig. 3b). Fig. 4 shows the XRD result of the cathode in Cell-II at room temperature, demonstrating that the as-fired cathode consisted of the major phases of YSZ and LSM and infiltrated minor phase of Pd_{0.95}Mn_{0.05}O [19].

Fig. 5 shows the initial performance (I–V and I–P curves) of Cell-I (Fig. 5a) and Cell-II (Fig. 5b) at temperatures between 650 and



Fig. 2. Appearance of the anode-supported planar cell with a dimension of $11 \times 11 \times 0.1$ cm and an active area of 9×9 cm².

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