



Optimization of strontium molybdate based composite anode for solid oxide fuel cells



Zhihong Du ^a, Hailei Zhao ^{a, b, *}, Chunyang Yang ^a, Yongna Shen ^a, Chunlin Yan ^a, Yang Zhang ^a

^a School of Materials Science and Engineering, University of Science and Technology Beijing, Beijing 100083, China

^b Beijing Key Lab of New Energy Material and Technology, Beijing 100083, China

HIGHLIGHTS

- Enhanced electrochemical performance of SrMoO₃ by electrolyte incorporating.
- SrMoO₃/GDC composite exhibits the lowest polarization resistance.
- SMO–60GDC composite electrode shows optimized anode performance.
- Perfect chemical compatibility of SrMoO₃ with YSZ, LSGM and GDC electrolytes.

ARTICLE INFO

Article history:

Received 25 August 2014

Received in revised form

10 October 2014

Accepted 12 October 2014

Available online 18 October 2014

Keywords:

Strontium molybdate

Electrochemical performance

Compatibility

Anode materials

Solid oxide fuel cells

ABSTRACT

Perovskite-type SrMoO₃ (SMO) is composited with commonly used electrolytes Y_{0.08}Zr_{0.92}O₂ (YSZ), La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O₃ (LSGM) and Gd_{0.1}Ce_{0.9}O₂ (GDC) to form dual-phase materials, which are characterized as potential anode materials for solid oxide fuel cells. Introduction of any of the electrolytes YSZ, LSGM and GDC can remarkably enhance the electrochemical performance of SrMoO₃. The polarization resistance of the composite anode in a symmetrical cell decreases in the sequence of SMO–LSGM, SMO–YSZ and SMO–GDC. The content of GDC in SMO–xGDC composite anode has a considerable effect on the catalytic activity of the electrode. The SMO–60GDC composite shows the lowest polarization resistance of 0.107 Ω cm² at 850 °C. With SMO–60GDC composite as anode, the maximum power densities of 584, 452 and 257 mW cm⁻² are achieved at 900, 850 and 800 °C, respectively, for LSGM electrolyte supported cell with La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O₃ as cathode. The cell exhibits considerably stable performance under a constant voltage of 0.71 V at 800 °C in a short-term cycle test.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

Solid oxide fuel cells (SOFCs) have been recognized as promising energy generation devices, which convert chemical energy of fuel directly into electricity with high efficiency and fuel flexibility [1]. The high operating temperature allows for the utilization of a variety of different fuels, such as hydrogen, syngas, natural gas, biogas, or even solid carbon [2,3]. The traditional nickel-based cermet anode is capable of providing excellent power output

using H₂ and CO fuels. However, it suffers from many problems, which can lead to cell performance degradation during long-term operation, such as Ni coarsening, poor redox stability, and deactivation by carbon deposition/sulfur poisoning when hydrocarbons are directly utilized [4–6]. Therefore, alternative anode materials with strong structural stability and good tolerance to carbon deposition/sulfur poisoning are urgently in demand.

Recent years, perovskite oxides have been widely investigated as potential anode materials and displayed promising performance. Most of these anodes are mixed conductors, which could yield superior kinetics reactions for hydrogen oxidation. In the reports on such anodes, La and Y doped SrTiO₃ materials exhibit high electrical conductivity in fuel atmosphere and good dimensional stability, however, the electrocatalytic activity is not sufficient to serve as an alternative anode compared to nickel-based cermet [7–9]. Hence,

* Corresponding author. School of Materials Science and Engineering, University of Science and Technology Beijing, Beijing 100083, China. Tel./fax: +86 10 82376837.

E-mail address: hlzhao@ustb.edu.cn (H. Zhao).

various extra modifications are attempted to improve the anode performance, such as decorating with CeO₂, Ni and Pd nanoparticles [10,11]. La_{0.75}Sr_{0.25}Cr_{0.5}Mn_{0.5}O₃ (LSCM) is another attractive anode material, which performs excellent redox stability in both oxidizing and reducing atmosphere and shows high resistance to carbon deposition when using hydrocarbon as fuel [12,13]. Many efforts were devoted to improving its electronic conductivity and electro-catalytic activity for efficient application at intermediate temperatures [14–16]. Recent years, double perovskite Sr₂MMoO₆ (M = Mg, Mn, Fe, Co, Ni) materials draw much attention because of their good cell performance and strong sulfur and carbon deposition resistance [17–19]. To cater the requirements of practical application, many properties need to be further enhanced in terms of electrical conductivity, catalytic activity and redox stability [20–22].

Scheelite-type SrMoO₄ is an insulating impurity commonly observed in preparation process of double perovskite materials, which, fortunately, can easily be converted to a good electronic conductive phase SrMoO₃ under anode atmosphere [23]. The SrMoO₃ oxide with perovskite structure exhibits one of the highest electrical conductivity values ($\sim 10^4$ S cm⁻¹) at room temperature [24]. With its high electronic conductivity and good structural stability in reducing atmosphere, SrMoO₃ has been proved to be a potential anode material for SOFCs [25–28]. However, this material possesses very low oxygen-ion conductivity [26] and poor catalytic activity towards fuel oxidation, which will limit the electrode performance in fuel cell.

An approach to overcome these problems is to substitute Mo with aliovalent transition metal elements (like Fe, Cr and Co) [26–28]. Incorporation with electrolyte materials is another effective way to enhance the ionic conductivity and catalytic activity of perovskite anode. Several composite anode materials, such as La_{0.2}Sr_{0.8}TiO₃-Gd_{0.2}Ce_{0.8}O_{2- δ} , LSCM/(Gd,Ce)O_{2- δ} and Sr_{0.92}Y_{0.08}TiO_{3- δ} -Sm_{0.2}Ce_{0.8}O_{2- δ} , have been reported to show remarkably improved electrochemical properties with the addition of electrolyte component [29–31]. The anode performance of SrMoO₃-yttria-stabilized zirconia composite was also further enhanced by impregnating Gd_{0.2}Ce_{0.8}O₂ electrolyte [32].

In the present work, different electrolytes Y_{0.08}Zr_{0.92}O₂ (YSZ), La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O₃ (LSGM) and Gd_{0.1}Ce_{0.9}O₂ (GDC) are incorporated into SrMoO₃, respectively, to improve its electrochemical performance. The effects of electrolyte species and electrolyte/SrMoO₃ ratio on the electrochemical performance of the SrMoO₃-based composite anode were investigated with symmetrical and single cells.

2. Experimental

2.1. Materials preparation

Scheelite-type oxides SrMoO₄ (SMO) powder was synthesized as precursor for SrMoO₃ perovskite by a citric-nitrate combustion process using Sr(NO₃)₂ (99.9%, Sinopharm), (NH₄)₆Mo₇O₂₄·4H₂O (99.9%, Sinopharm) as starting materials while the citric acid monohydrate (C₆H₈O₇·H₂O, Guangdong Xilong) as the chelating agent. The molar ratio between the total amount of metal ions and citric acid monohydrate was 1:2. The obtained solution was heated in water bath at 80 °C to form a gel and then the obtained gel was slowly heated in an oven until self-combustion occurred at 250 °C. The combustion product was ground and subsequently calcined at 800 °C for 6 h with an intermediate stay at 400 °C for 2 h to remove the organic residues and obtain the single phase simultaneously. Corresponding LSGM, GDC, La_{0.4}Ce_{0.6}O₂ (LDC) and La_{0.58}Sr_{0.4}Co_{0.2}-Fe_{0.8}O₃ (LSCF) powders were also synthesized by the same method. The latter two powders were applied as buffer layer and cathode materials in single-cell preparation, respectively.

The SrMoO₄ powder was mixed with 50 wt.% YSZ (TZ-8Y, Tosoh Corporation), LSGM (calcined at 1400 °C for 8 h) and GDC (calcined at 650 °C for 6 h) electrolyte powders to form the composite anodes SMO-YSZ, SMO-LSGM and SMO-GDC, respectively. The SMO-xGDC composites with different percentages of GDC powder ($x = 40, 50, 60, 70$ wt. %) were prepared and referred to as SMO-40GDC, SMO-50GDC, SMO-60GDC, and SMO-70GDC, respectively. The SrMoO₃/electrolyte composite anodes were reduced *in situ* at 850 °C in H₂ for 4 h during cell test before data collection.

2.2. Cell fabrication

The LSGM pellets of 19 mm diameter were sintered at 1450 °C for 8 h and then polished to a thickness of 400 μ m. LDC was pasted on the electrolyte and sintered at 1400 °C for 2 h, which was employed as buffer layer between the anode and the electrolyte to prevent the interdiffusion of ionic species [33]. The electrode slurries were prepared by homogeneously mixing the electrode powders with α -terpineol solution of 6 wt.% ethylene cellulose in a weight ratio of 2:1. For symmetrical cells, the composite slurries were screen-printed on both sides of LDC coated LSGM electrolyte symmetrically with an active area of 0.5 cm², followed by calcining at 1200 °C for 2 h. The cell performance of the SMO-xGDC composites anode was tested with the cell configuration of SMO-xGDC|LDC|LSGM|LSCF. SMO-xGDC composite anodes were screen-printed on the LDC layer and then sintered at 1200 °C for 2 h. The LSCF, which was synthesized at 1000 °C for 6 h, was finally applied as cathode by screen-printing on the other side of LSGM and firing at 1050 °C for 2 h. Ag paste was used as the current collector, which was painted as grid on both sides of the calcined cells and fired at 700 °C for 0.5 h. Then the obtained cells were sealed on an alumina tube using a ceramic-based material (Cerama-bond 552-VFG, Aremco). The humidified H₂ was fed as fuel to the anode side with a flow rate of 40 ml min⁻¹ and air was conducted to the cathode side at a flow rate of 100 ml min⁻¹ as oxidant. Before fuel cell performance testing, the anode was reduced at 850 °C for 4 h to get SrMoO₃/electrolyte composite electrode.

2.3. Characterization

Phase composition and crystal structure of the samples were identified by X-ray diffraction (XRD, Rigaku TTRIII) with CuK α radiation. To examine the chemical compatibility of SMO with YSZ, LSGM and GDC electrolytes, the SrMoO₄ powder was mixed uniformly with electrolyte powders in 1:1 mass ratio, and then pressed and calcined at 1250 °C for 6 h in air. The calcined pellets were crushed and examined by XRD. The scanning electron microscope (SEM, LEO-1450) was used to observe the microstructure of the cells. The Impedance measurement of the symmetrical cells and single-cells was performed on Solartron 1260 impedance gain/phase analyzer in combination with 1287 electrochemical interface in the 0.1–10⁶ Hz frequency range with perturbation amplitude of 10 mV. The V - I (voltage-current) curves were recorded in the range of 800–900 °C by using a Solartron 1287 Electrochemical interface controlled by CorrWare software.

3. Results and discussions

3.1. Structural and chemical characterization

The SrMoO₄ powders are synthesized through a citric-nitrate combustion process with a post heat-treatment at 800 °C in air. Fig. 1 displays the XRD pattern and particle morphology of the

Download English Version:

<https://daneshyari.com/en/article/7734242>

Download Persian Version:

<https://daneshyari.com/article/7734242>

[Daneshyari.com](https://daneshyari.com)