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Strontium-doped samarium manganite as cathode materials for oxygen reduction reaction in solid oxide fuel cells

W. Li [a, b], C.Y. Xiong [a], L.C. Jia [a], J. Pu [a], B. Chi [a], X. Chen [b], J.W. Schwank [b, *], J. Li [a, *]

[a] Center for Fuel Cell Innovation, School of Materials Science and Engineering, Huazhong University of Science & Technology, Wuhan, Hubei 430074, China
[b] Department of Chemical Engineering, University of Michigan, Ann Arbor, MI 48109, USA

HIGHLIGHTS

- SSM show better electrochemical performance than LSM82.
- SSM55 possesses high surface Mn^{4+}/Mn^{3+} and $O_{ad}/O_{lattice}$ concentration ratios.
- The surface oxygen exchange kinetics for SSM are faster than LSM82.

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ABSTRACT

$Sm_xSr_{1-x}MnO_3$ with x = 0.3, 0.5 and 0.8, denoted as SSM37, SSM55 and SSM82, respectively, have been prepared via a sol–gel route as materials for cathodes in solid oxide fuel cells. Their activities in the oxygen reduction reaction (ORR) have been evaluated in comparison with the state-of-the-art cathode material $La_{0.8}Sr_{0.2}MnO_3$ (LSM82) by electrochemical impedance spectroscopy (EIS), X-ray photoelectron spectroscopy (XPS) and thermogravimetry (TG). Among all the prepared cathodes, the SSM55 exhibits the lowest values, while the LSM82 exhibits the highest polarization resistance, at open circuit voltage (OCV) and temperatures from 650 to 800 °C. This result indicates that the prepared $Sm_xSr_{1-x}MnO_3$ is a promising replacement for LSM82 as cathode material for SOFCs, and the SSM55 represents the optimal concentration in $Sm_xSr_{1-x}MnO_3$ series. The remarkably high ORR activity of the SSM55 is ascribed to its high surface Mn^{4+}/Mn^{3+} and $O_{ad}/O_{lattice}$ ratios and fast surface oxygen exchange kinetics.

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1. Introduction

Solid oxide fuel cells (SOFCs) are considered as a green solution towards the increasing demand for clean energy and favored for their high efficiency in energy conversion, flexibility in fuel selection, and reduced impact on the environment. Typically, SOFCs with Sr-doped $LaMnO_3$ (LSM) cathodes are operated at high temperatures near 1000 °C. When the operating temperature is lowered to a commercially more attractive intermediate range between 600 °C and 800 °C, the increasing cathode overpotential becomes a critical issue. Therefore, exploring cathode materials with high electrocatalytic activity for the oxygen reduction reaction (ORR) within the intermediate temperature (IT) range is of great significance.

$Sm_{0.5}Sr_{0.5}MnO_3$, a material with relatively low energy barriers for oxygen adsorption and diffusion [1], has been investigated as a promising cathode material for IT-SOFCs [2–6]. It exhibits an excellent chemical compatibility with Y_2O_3 stabilized ZrO_2 (YSZ) electrolyte [2] and possesses an electrical conductivity higher than the requirement of 100 S cm^{-1} for SOFC cathode materials [3]. It has also been predicted by first-principles simulation that the presence of Sm leads to weak bonding between the $Sm_{0.5}Sr_{0.5}MnO_3$ surface and adsorbed oxygen species and a low diffusion barrier for oxygen ions, thus facilitating the ORR [1,4]. $Sm_{0.5}Sr_{0.5}MnO_3$ [5] was also proved to be a thermally stable cathode up to 1400 °C, with a thermal expansion coefficient of 10.7×10^{-6} K^{-1}, which is close to that of the most conventional electrolyte YSZ. Dong et al. [6] has also demonstrated that it performs better with YSZ electrolyte compared to $Sm_{0.2}Ce_{0.8}O_{1.9}$ (SDC). In the present study, we extended the investigation on $Sm_xSr_{1-x}MnO_3$ as a cathode material to different compositions with YSZ electrolyte, using X-ray diffraction (XRD), electrochemical impedance spectroscopy (EIS), X-ray photoelectron spectroscopy (XPS) and thermogravimetry as

* Corresponding authors.
E-mail addresses: schwank@umich.edu (J.W. Schwank), lijian@hust.edu.cn (J. Li).

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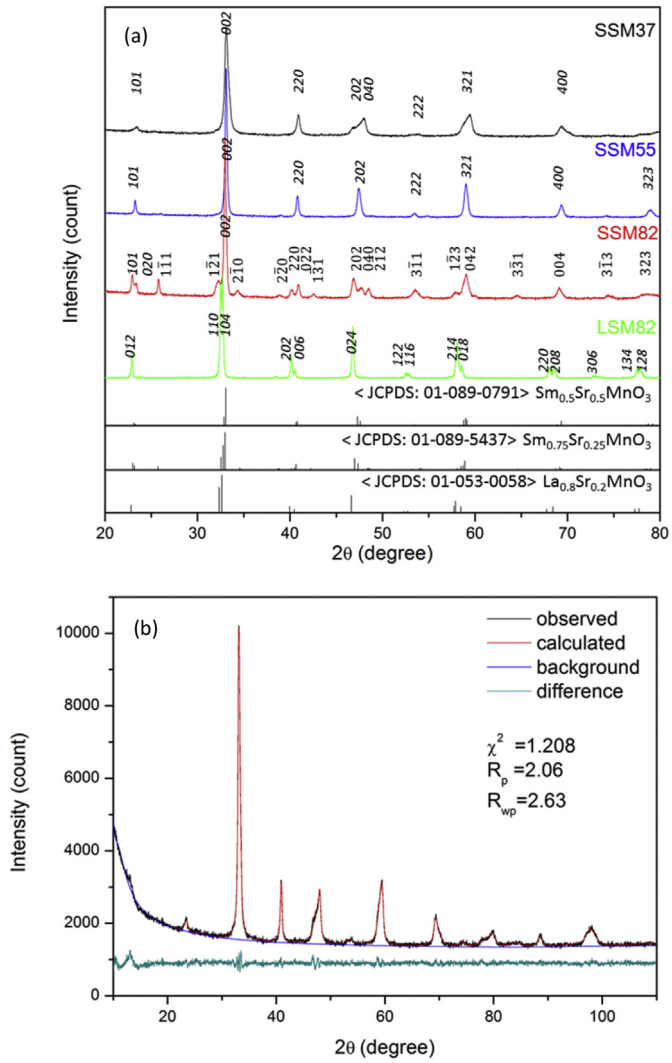

Fig. 1. Refined X-ray diffraction patterns for SSM37, SSM55, SSM82 and LSM82 powders (a) and the Rietveld refinement for SSM37 (b).

characterization methods. For comparison, the behavior of the state-of-the-art cathode material $La_{0.8}Sr_{0.2}MnO_3$ (LSM82) was also characterized accordingly.

2. Experimental

2.1. $Sm_xSr_{1-x}MnO_3$ synthesis

$Sm_xSr_{1-x}MnO_3$ (SSM) powders with x = 0.3, 0.5 and 0.8, designated as SSM37, SSM55 and SSM82, respectively, were synthesized by a sol–gel method in the aqueous solution. Stoichiometric amounts of metal nitrates of $Sm(NO_3)_3{\cdot}6H_2O$, $Sr(NO_3)_2$ and $Mn(NO_3)_3$ (Sigma Aldrich) were dissolved in distilled water, to which chelating agents citric acid and ethylene alcohol were then added sequentially to form a clear solution. This solution was evaporated at 80 °C in a water bath for 3 h to form a red colored gel, which was then held at 180 °C in an oven for 5 h. The obtained dry gel was ground and calcined at 800 °C for 2 h to form the SSM powders. The commercial LSM82 powder was provided by Fuel Cell Materials. The crystallographic phases of the SSM and LSM82 powders were determined by XRD (Rigaku Rotaflex RU-200B series) using CuKα radiation at 100 mA and 40 mV. The X-ray diffraction patterns of $Sm_xSr_{1-x}MnO_3$ compositions were refined by the Rieltveld method using GSAS software.

Table 1
Lattice parameters and unit cell volume of $Sm_xSr_{(1-x)}MnO_3$.

Composition	a (Å)	b (Å)	c (Å)	V (Å^3)
SSM37	5.43	7.59	5.45	224.73
SSM55	5.42	7.65	5.44	225.66
SSM82	5.42	7.64	5.56	230.42

2.2. Cell fabrication and evaluation

To prepare electrolyte-supported symmetric cells with the above cathode materials, YSZ powder (Tosoh) was die pressed and sintered in air at 1500 °C for 5 h to obtain dense YSZ electrolyte pellets with a dimension of Φ 20 × 1 mm. SSM37, SSM55, SSM82 and LSM82 slurries containing 5 wt.% ethyl cellulose in terpineol were spin-coated on both sides of the YSZ pellets, respectively, by using a Laurell WS-400A-6NPP/LITE spin coater. To avoid chemical reaction between the cathode materials and electrolyte and to adjust the porosity of cathode, the SSM- and LSM82-coated cells were sintered in air for 2 h at 1020 °C and 1150 °C, respectively. The active area of the symmetric cells was 1.8 cm^2. To prevent the contribution of current collecting material to the cathode performance, Au paste (Fuel Cell Materials), which is inert for the oxygen reduction reaction, was coated on the top of the cathodes as current collector for the EIS tests. These tests were carried out in air at

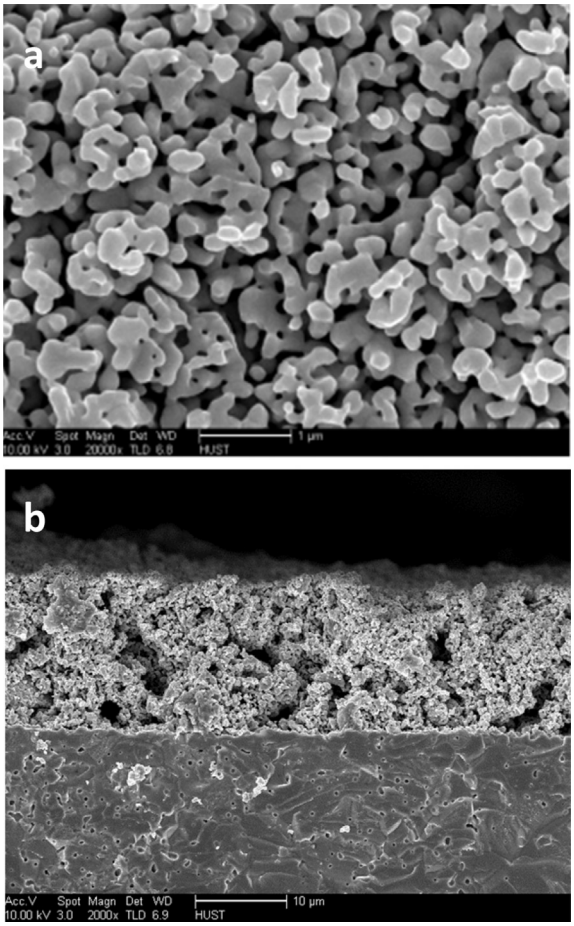

Fig. 2. SEM microstructure of SSM55 cathode (a) and SSM55/YSZ interface (b) sintered in air at 1020 °C for 2 h [5].

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