



A new symmetric solid-oxide fuel cell with $\text{La}_{0.8}\text{Sr}_{0.2}\text{Sc}_{0.2}\text{Mn}_{0.8}\text{O}_{3-\delta}$ perovskite oxide as both the anode and cathode

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Abstract

A novel perovskite-type $\text{La}_{0.8}\text{Sr}_{0.2}\text{Sc}_{0.2}\text{Mn}_{0.8}\text{O}_3$ (LSSM) oxide was synthesized and evaluated as the electrode material of a symmetric solid-oxide fuel cell. Characterization was done by electrical conductivity, crystal structure stability, redox stability, catalytic activity for methane oxidation and oxygen electro-reduction. LSSM shows greater electrical conductivity than the typical $\text{La}_{0.8}\text{Sr}_{0.2}\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_3$ (LSCM) perovskite oxide under both anode and cathode operating conditions. It also shows excellent chemical and structural stability due to the backbone effect of Sc^{3+} for the perovskite lattice structure. A symmetric electrolyte-supported cell with 0.3 mm thick scandium-stabilized zirconia electrolyte and LSSM as cathode and anode shows peak power densities of 310 and 130 mW cm^{-2} at 900 °C, respectively, when operating on wet H_2 and wet CH_4 . Stable performance is demonstrated.

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1. Introduction

Fuel cells, which electrochemically convert chemical energy to electrical power with much greater efficiency and lower emission of greenhouse gases than well-established technologies based on traditional fossil fuel combustion, have attracted growing attention for the development of renewable energy supplies [1–3]. Among the many types of fuel cells, proton-exchange-membrane fuel cell (PEMFC) and solid-oxide fuel cell (SOFC) are the two most explored types. A SOFC is an all-solid device which typically operates at 500–1000 °C. The high operating temperature improves fuel conversion efficiency and electrode activity [4,5]. Consequently, much cheaper metals or metal oxides can be applied as electrode materials to replace precious Pt, which is the main component of PEMFCs' electrode catalyst [6,7]. The high operating temperature also

accepts a wide range of fuels; besides hydrogen, other more accessible chemicals such as methane, gasoline and coal are all potential fuels for SOFCs [8–14]. The high fuel flexibility is a practical advantage since the infrastructure of hydrogen production, storage and transportation is still far from mature. Despite the above benefits, progress towards the commercialization of SOFCs is still much less than that of PEMFCs. The main obstacles include inefficient materials, complicated fabrication processes and poor long-term operating stability.

A SOFC is composed of a dense oxide electrolyte sandwiched between two porous electrodes, which typically have been sintered differently and have different thermal expansion coefficients (TEC). To perform as an ideal electrolyte, the oxide should have high ionic conductivity but negligible electronic conductivity, favorable thermal and chemical stability under both oxidizing and reducing atmospheres, and high mechanical strength. Ytria- or Scandia-stabilized zirconia (YSZ or ScSZ) are the most well developed ionic conductors and meet most of the

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requirements of SOFC electrolytes [4,5]. Exploiting proper electrode materials is, however, still an area of great challenge. Because they are exposed to atmospheres with significantly different oxygen potentials, materials with different compositions are usually applied to the anode and cathode. Recently, a new SOFC configuration of using the same material for both electrodes, named symmetric SOFC, has been proposed and investigated [15–20]. The application of this new concept leads to a number of advantages. Possible coke formation over the anode can be easily eliminated by facilely switching the gas flow without causing damage to the cell. In the same way, sulphur poisoning of the anode can also be effectively alleviated. Based on this concept, the life-span of fuel cell is also doubled. Furthermore, the strains induced during the fabrication of SOFC due to the mismatch of TECs between different cell components are greatly reduced since only one type of electrode–electrolyte interface exists. The identical composition of anode and cathode also allows them to be fired in a single step during cell fabrication; consequently, fabrication cost could be effectively reduced.

As the electrode of a symmetric SOFC, it should meet the requirements of a cathode and an anode simultaneously, i.e., good chemical and thermal stability and high electronic conductivity under both oxidizing and reducing atmospheres, and high electrochemical activity for oxygen reduction and for hydrogen or hydrocarbon oxidation. The state-of-the-art anode is composed of Ni/YSZ (Ni/ScSZ) cermet [21,22], which shows excellent electro-catalytic properties for hydrogen oxidation and favorable electronic conductivity ($>1000 \text{ S cm}^{-1}$). The classic SOFC cathode is composed of a perovskite-type oxide, such as a pure electronic conductor of $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ (LSM) [23], or mixed ionic and electronic conductors of $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ (LSCF) [24,25] and $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ (BSCF) [26,27], which show excellent cathodic performances at high and intermediate temperatures, respectively. Since nickel easily loses its activity and electronic conductivity under cathodic atmospheres due to oxidation of nickel, LSCF- and BSCF-based perovskite oxides are chemically unstable under the reducing atmosphere of the anode; thus, all of these materials are not applicable as the electrode of a symmetric SOFC.

Up to now, only very limited numbers of materials have successfully been demonstrated as electrodes of symmetric SOFCs. $\text{La}_{0.75}\text{Sr}_{0.25}\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_3$ (LSCM) represents one of the most investigated materials [15–18]. As an anode material, LSCM showed wonderful chemical stability but relatively low electronic conductivity in the fuel atmospheres. More importantly, LSCM had relatively poor oxygen reduction activity as a cathode material. On the other side, as mentioned previously, a classic cathode material of LSM is chemically unstable under the reducing atmosphere of the anode. Therefore, it is urgently to develop an alternative electrode for symmetric solid-oxide fuel cell application. Previously, we demonstrated that $\text{La}_{0.8}\text{Sr}_{0.2}\text{Sc}_y\text{Mn}_{1-y}\text{O}_3$ possesses not only better cathode performance

in oxidizing atmosphere, but also higher chemical and structural stability than LSM under reducing atmospheres [28]. Both characteristics are the most fundamental requirements of a potential electrode for symmetric SOFCs.

In this study, for the first time, $\text{La}_{0.8}\text{Sr}_{0.2}\text{Sc}_{0.2}\text{Mn}_{0.8}\text{O}_3$ (LSSM) perovskite oxide was systematically evaluated as the electrodes of a symmetric SOFC with zirconia-based electrolyte operating on hydrogen or methane fuel. Investigations were made into the phase stability, electrical conductivity under both reducing and oxidizing atmospheres, electro-catalytic activity for oxygen reduction, catalytic activity for methane oxidation, and single cell performance.

2. Experimental procedures

LSSM oxide powder was synthesized by a combined EDTA-citrate complexing sol–gel process [29,30]. $\text{La}(\text{NO}_3)_3 \cdot x\text{H}_2\text{O}$, $\text{Sr}(\text{NO}_3)_2$, Sc_2O_3 and $\text{Mn}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$, (all A.R. grade) were used as the metal–ion sources. $\text{La}(\text{NO}_3)_3 \cdot x\text{H}_2\text{O}$ was first prepared in an aqueous solution around 1 M with its precise concentration determined by the standard EDTA titration technique. Sc_2O_3 was prepared in 1 M aqueous $\text{Sc}(\text{NO}_3)_3$ solution by dissolving it in 6 M nitric acid under heating and then diluted with a proper amount of de-ionized water. Required amounts of $\text{La}(\text{NO}_3)_3$, $\text{Sr}(\text{NO}_3)_2$, $\text{Sc}(\text{NO}_3)_3$, and $\text{Mn}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$ were then prepared in a mixed aqueous solution. EDTA- $\text{NH}_3 \cdot \text{H}_2\text{O}$ solution and citric acid solid were added in sequence under stirring and heating at a mole ratio of total metal ions to EDTA to citric acid of 1:1:2. The water was evaporated from the solution by heating at 90°C until a transparent gel was obtained, which was pre-fired at 250°C and further calcinated at 950°C in air for 5 h to get the final products with the desired lattice structure.

$(\text{Sc}_2\text{O}_3)_{0.1}(\text{ZrO}_2)_{0.9}$ (ScSZ) was used as the electrolyte material, also prepared by the EDTA-citrate complexing process. To fabricate a single cell, the ScSZ powders were pressed into disk-shape pellets and then fired in air at 1500°C for 5 h to achieve densification of the electrolytes. The sintered cells had a diameter of $\sim 13 \text{ mm}$ and a thickness of $\sim 0.3 \text{ mm}$. LSSM powders were dispersed in a pre-mixed solution of glycerol, ethylene glycol and isopropyl alcohol to form a colloidal suspension using a high-energy ball miller (Fritsch, Pulverisette 6) at a rotational speed of 400 rpm for 1 h. The suspension was then symmetrically painted on both central surfaces of the dense ScSZ pellets. The whole cell was then calcinated at 1150°C for 2 h in air to fabricate complete symmetric SOFCs.

The phase structure of synthesized powders was characterized by an X-ray diffractometer (XRD, Bruker D8 Advance) equipped with Cu K α radiation ($\lambda = 1.5418 \text{ \AA}$). For phase stability tests, about 0.2 g samples were placed in a U-type quartz tube reactor and treated at high temperature (typically 850°C) under various atmospheres for 24 h. The oxides were cooled to room temperature under the same atmosphere and subjected to phase structure examination.

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