

Fuel cell studies of perovskite-type materials for IT-SOFC

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Abstract

The electrochemical performance of solid oxide fuel cells (SOFCs) based on perovskite-type materials (ABO_3) was investigated. $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM) ceramics were used as electrolyte and a composite containing $La_{0.8}Sr_{0.2}MnO_3$ (LSM) as cathode. $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (BSCF) was also used as cathode and $La_{0.75}Sr_{0.25}Cr_{0.5}Mn_{0.5}O_{3-\delta}$ (LSCM) as anode materials. Furthermore, fluorite-type $Sm_{0.15}Ce_{0.85}O_{2-\delta}$ (SDC) material was used as buffer layer between the electrolyte and the anode to avoid possible interfacial reactions. The maximum power density value of BSCF/LSGM/LSCM with 1.5 mm thick electrolyte supported cell was 160 mW cm^{-2} at 1073 K, using moist H_2 diluted with N_2 as fuel and air as oxidant.

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1. Introduction

The versatile perovskite-type structure (ABO_3) is an interesting option to develop new materials for intermediate temperature solid oxide fuel cells (IT-SOFCs). It is well known that the ionic conductivity of $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{2.85}$ (LSGM) [1–5] is higher as compared to yttria-doped zirconia (YSZ) at intermediate temperatures (773–1073 K). This material has an ionic conductivity of about 0.1 S cm^{-1} at 1073 K, good chemical stability and negligible electronic conduction over a broad range of oxygen partial pressures ($1\text{--}10^{-20}$ atm) [6]. On the other hand, LSGM has some disadvantages as electrolyte, such as undesired segregation of impurities during its synthesis, e.g. $LaSrGa_3O_7$ and $LaSrGaO_4$ [7], and chemical incompatibility with Ni-based anode materials [8,9]. Ni reacts with LSGM to form $LaNiO_3$ [10]. However, the synthesis can be optimized to reduce undesired phases and alternative anode material without nickel or a buffer layer, between electrolyte

and Ni-based anode, can be used to avoid interfacial reactions [11].

Results from LSGM electrolyte supported cells with $La_{0.8}Sr_{0.2}MnO_3$ (LSM) as cathode and $La_{0.75}Sr_{0.25}Cr_{0.5}Mn_{0.5}O_{3-\delta}$ (LSCM) [12,13] as anode were reported in a previous work [14]. Dense LSGM samples were prepared by tape casting. The power density reached a value of 570 mW cm^{-2} at 1073 K with moistened H_2 and O_2 as oxidant. However, the output power density drastically decreased at 873 K, mainly due to the high polarization resistance of LSM and higher ohmic losses of the electrolyte. Therefore, different strategies were envisaged to improve the performance of LSGM-based cells using LSCM as anode material at low temperature: reducing the electrode overpotential and avoiding the possible interfacial reactions between the electrodes and the electrolyte.

For the first option, a composite of LSM and LSGM in a 30–70% molar ratio (LSM–LSGM) was used. A lower cathodic overpotential had been reported using LSM–LSGM as composite than with LSM only, both on LSGM electrolyte [15]. In addition, the recently proposed $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (BSCF) material was used in this study as alternative cathode, due to the good properties for electrochemical reduction of oxygen in the range of 773–973 K [16]. For the second option,

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$\text{Sm}_{0.15}\text{Ce}_{0.85}\text{O}_{2-\delta}$ (SDC) was used as buffer layer between the electrolyte and the anode material.

LSM/LSGM/LSCM, LSM/LSGM/SDC/LSCM, LSM–LSGM/LSGM/LSCM and BSCF/LSGM/LSCM cells were prepared and tested in this work and their performances compared.

2. Experimental

2.1. Electrodes and electrolyte preparation

LSGM electrolyte, LSM–LSGM cathode and LSCM anode materials were prepared by conventional solid state reaction, using powders of La_2O_3 (Fluka, 99.98%), Ga_2O_3 (Fluka, 99.99%), MgO (Fluka, 99%), SrCO_3 (Aldrich, 99.9%), Mn_2O_3 (Aldrich, 99.999%) and Cr_2O_3 (ABCR, 99.999%). La_2O_3 and MgO were fired at 1273 K for more than 3 h before weighing to remove water and impurities, in order to ensure the correct stoichiometry. Commercial powders of LSM obtained via combustion spray pyrolysis (SSC Inc. Worthington, USA, 99.9%) were used to prepare the LSM cathode specimens.

BSCF cathode and SDC interlayer materials were prepared via sol–gel, using powders of $\text{Ba}(\text{NO}_3)_2$ (Fluka, 99.95%), $\text{Sr}(\text{NO}_3)_2$ (Fluka, 99%), $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (Fluka, 98%), $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (Merck, 99%), $\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (Alfa Aesar, 99.9%) and $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (Alfa Aesar, 99.9%).

The ceramic route was done by ball-milling stoichiometric quantities of the reagents with acetone. The resulted mixture was dried and fired in air up to 1173 K for 10 h and ground again. Dense samples of LSGM were obtained by uniaxial pressing into a disk at 140 MPa, for 5 min ($\varnothing = 3.0$ cm) of the synthesized powders and sintered at 1673 K for 6 h. LSCM and LSM–LSGM powders were obtained after treatment at 1373 K for 10 h and 1473 K for 6 h, respectively.

The sol–gel route was carried out, using citric acid and ethylenediaminetetraacetic acid (EDTA) as complex agents to prepare BSCF and SDC materials. Stoichiometric cation solution of the corresponding nitrates was prepared with distilled water. An ammonium solution of EDTA, 1.5 wt.% in excess, with a ratio ligand/metal (L/M) = 1 was added. Afterwards, the citric acid solution, 1.5 wt.% in excess and in a ratio L/M = 1 and 2/3 for trivalent and divalent cations, respectively, was also added. The pH was adjusted up to 9 with addition of ammonia solution (20%, v/v). A gel was formed with continuous stirring and low heating (~ 338 K). It was dried at room temperature for more than 8 h. The dried gels were fired at 573 K for 30 min.

The obtained powders were ground in an agate mortar and fired in air at 923 and 723 K for BSCF and SDC, respectively, during 5 h, ground again and finally fired at 1173 K for 5 h in air.

Electrode slurries were prepared with ethyl cellulose as binder and diethyleneglycol monobutylether acetate as solvent. The composition (wt.%) of the slurries was 75% of electrode powders, 5% of binder and 20% of solvent.

2.2. Material characterization

X-ray diffraction (XRD) patterns were recorded using a Siemens D5000 with $\text{K}\alpha_1$ copper radiation wavelength of 0.15406 nm. The scans were performed in the 2θ range ($5\text{--}120^\circ$) with 0.02° step and 10 s step^{-1} . XRD studies were also carried out to investigate the chemical compatibility of the electrolyte with the electrodes. Powder mixtures of LSGM with BSCF, LSGM with SDC and LSCM with SDC, 1:1 (wt.%) were ground in agate mortar and fired at 1173 and 1473 K for 10 h.

The morphology and composition of ceramic materials were observed by scanning electron microscopy (SEM) (JEOL JSM-6300 and LEO 1530) and energy dispersive X-ray spectroscopy (EDS) (NORAN Vantage) before and after the tests.

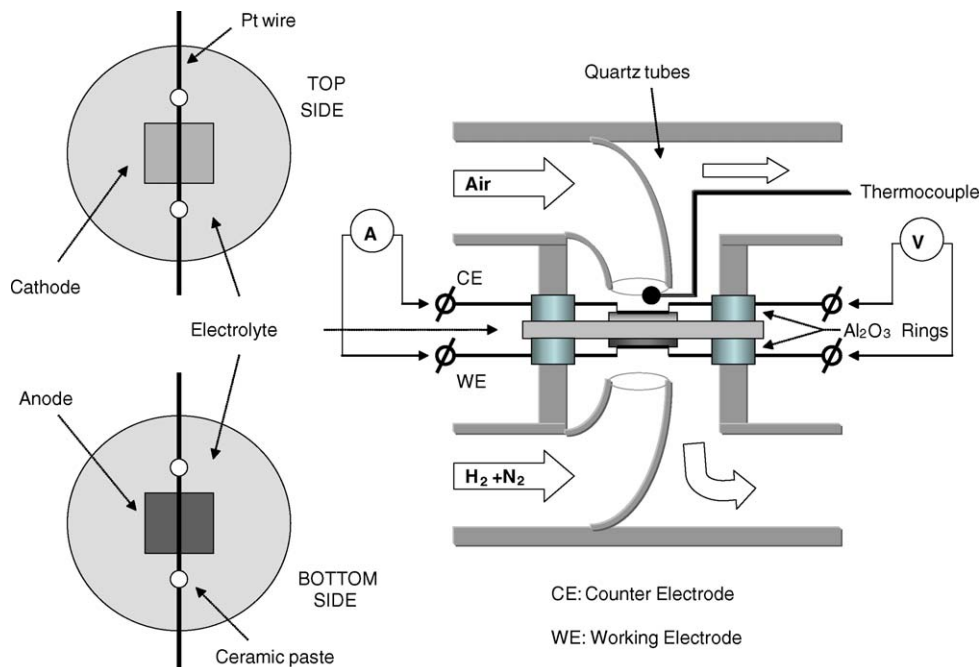


Fig. 1. Scheme of the setup for fuel cell tests.

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