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Barium carbonate nanoparticle to enhance oxygen reduction activity of strontium doped lanthanum ferrite for solid oxide fuel cell

Tao Hong [a], Fanglin Chen [b], Changrong Xia [a,*]

[a] CAS Key Laboratory of Materials for Energy Conversion, Department of Materials Science and Engineering, University of Science and Technology of China, Hefei, Anhui 230026, China

[b] Department of Mechanical Engineering, University of South Carolina, Columbia, SC 29208, USA

HIGHLIGHTS

- $BaCO_3$ nanoparticles enhance the oxygen reduction activity of LSF cathode.
- $BaCO_3$ increases oxygen chemical exchange coefficient by a factor of ~10.
- $BaCO_3$ nanoparticles are very stable upon heating at 700 °C for more than 300 h.

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ABSTRACT

$BaCO_3$ nanoparticles are demonstrated as outstanding catalysts for high-temperature oxygen reduction reaction (ORR) on the $La_{0.8}Sr_{0.2}FeO_{3-\delta}$ (LSF) cathode for solid oxide fuel cells (SOFCs) based on yttria-stabilized zirconia (YSZ) electrolytes. Thermal gravitational and X-ray diffraction measurements show that $BaCO_3$ is stable and chemically compatible with LSF under the fabrication and operation conditions of intermediate-temperature SOFCs. The $BaCO_3$ nanoparticles can greatly reduce the interfacial polarization resistance; from 2.96 to 0.84 Ω cm^2 at 700 °C when 12.9*wt*% $BaCO_3$ is infiltrated to the porous LSF electrode on the YSZ electrolyte. Electrochemical impedance spectroscopy shows that there is about one order of magnitude decrease in the low-frequency resistance, indicating that $BaCO_3$ nanoparticles can greatly enhance the surface steps for ORR. Electrical conductivity relaxation investigation indicates about one order of magnitude increase in the chemical oxygen surface exchange coefficient when $BaCO_3$ is applied, directly demonstrating significant increase in the kinetics for ORR. In addition, LSF cathodes with infiltrated $BaCO_3$ nanoparticles have shown excellent stability and substantially enhanced cell performance as demonstrated with single cells, suggesting $BaCO_3$ nanoparticles are very effective in enhancing ORR on LSF.

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1. Introduction

Oxygen reduction reaction (ORR) in the cathodes dominates the performance of a solid oxide fuel cells (SOFCs) [1]. The cathode performance could be greatly improved using impregnation/infiltration technique, which resulting in nanostructured electrodes that could increase the surface area of the electrocatalyst and/or enlarge the three-phase boundaries (TPB), where ORR is believed to take place [2–6]. Basically, two strategies are developed since a cathode is usually a composite consisting of two phases, an ionic conducting electrolyte and an electronic conducting electro catalyst [6]. One is infiltrating the electro catalyst to a porous structure/electrode to increase the catalytic activity by increasing the surface area. For example, when $PrBaCo_2O_{5+\delta}$ (PBC) is infiltrated into a composite electrode of PBC-SDC ($Sm_{0.2}Ce_{0.8}O_{2-\delta}$), the area specific interfacial polarization resistance at 600 °C is only 0.082 Ω cm^2, much lower than 0.25 Ω cm^2 for the PBC-SDC electrode on SDC electrolyte [7]. Improvement is also reported for typical electrocatalysts including Pd, Pt, Ag, $La_{0.8}Sr_{0.2}CoO_{3-\delta}$ (LSC) and $La_{0.8}Sr_{0.2}MnO_{3-\delta}$ [8–14]. In addition, enhanced stability upon thermal cycle is reported for the LSC infiltrated electrode comparing with the composite structure [14].

The other strategy is infiltrating the electrolyte to a porous structure to enlarge the TPB length [3], which exhibits the highest

* Corresponding author.
E-mail address: xiacr@ustc.edu.cn (C. Xia).

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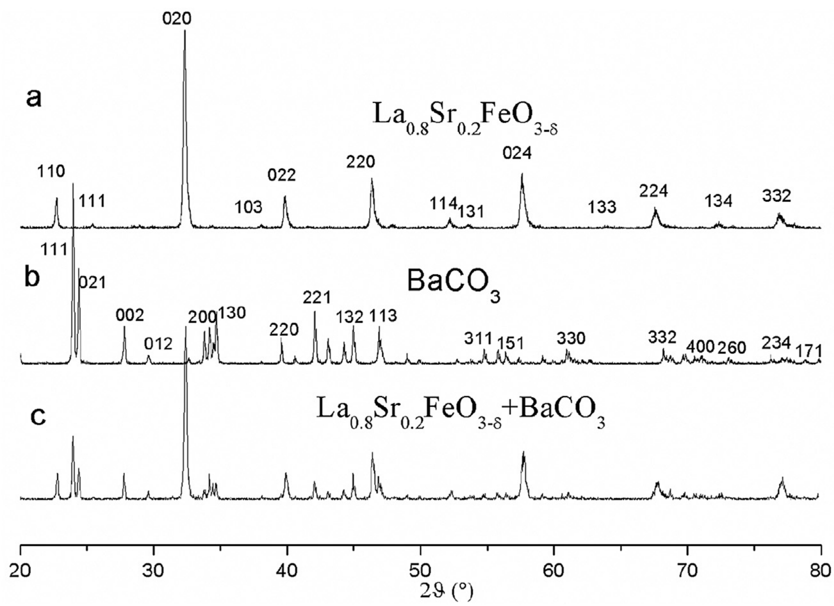

Fig. 1. XRD patterns for (a) LSF powder heated at 700 °C for 2 h. (b) $Ba(Ac)_2$ powder heated at 800 °C for 2 h and (c) LSF + $Ba(Ac)_2$ powders heated at 800 °C for 10 h.

value at a certain electrolyte loading that has been clearly demonstrated by model approach [15–17]. Typical electrolyte materials such as doped ceria, yttria stabilized zirconia (YSZ), and even stabilized bismuth oxides are investigated to improve the cathode performance [18–21]. The improvement generally increases to the highest and then decreases with the amount of infiltrated electrolyte [22,23]. The consistent in model prediction and experimental results demonstrates that the improvement is caused by the ionic conductivity of the infiltrated electrolyte.

In addition to the materials with electronic/ionic conductivity, some low or even none electronic/ionic conductive materials have also been infiltrated to improve the cathodic electrochemical performance although the reason for such improvement is not clear yet [24]. In this work, $BaCO_3$ nanoparticles are investigated to reveal their effect on the ORR for a cobalt free cathode material, $La_{0.8}Sr_{0.2}FeO_{3-\delta}$ (LSF). $BaCO_3$ is not an electronic/ionic conductor and has never been reported to as an oxygen reduction catalyst. This work demonstrates that $BaCO_3$ nano-particles, which are prepared by the infiltration method, can greatly enhance the performance of LSF cathodes on YSZ electrolytes, thus show significant catalytic activity for oxygen reduction. And $BaCO_3$ shows great thermal stability and chemical compatible.

2. Experimental section

2.1. Electrode and cell fabrication

$La_{0.8}Sr_{0.2}FeO_{3-\delta}$ (LSF) and $Zr_{0.85}Y_{0.15}O_{2-\delta}$ (YSZ) powders were synthesized by a glycine–nitrate process [25,26]. Symmetrical cells were composed of dense YSZ electrolyte substrates and porous LSF

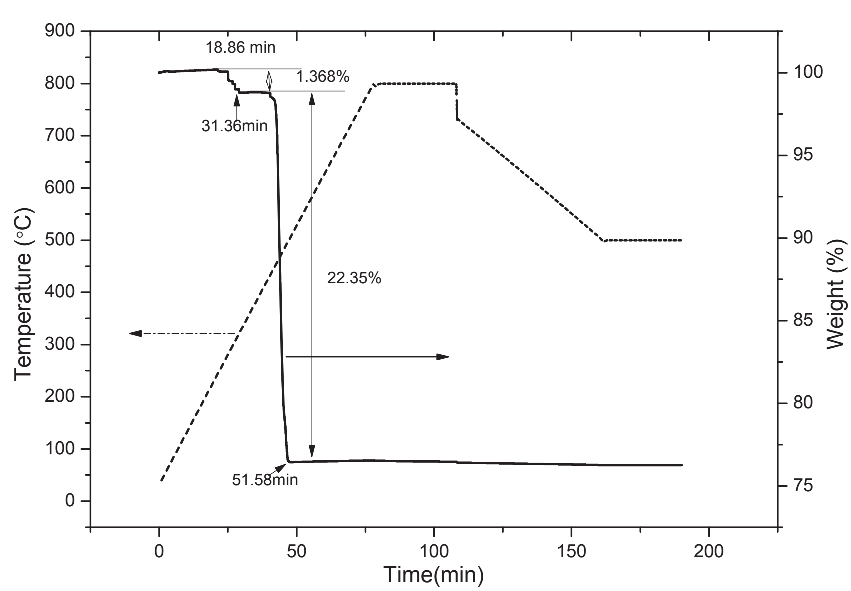

Fig. 2. Thermal gravitational curve for $Ba(Ac)_2$. The measurement is conducted in air atmosphere with a flux of 75 ml min^{-1}.

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