

Methane-fueled IT-SOFCs with facile in situ inorganic templating synthesized mesoporous $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ as catalytic layer

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Received 19 January 2007; received in revised form 10 April 2007; accepted 17 April 2007

Available online 22 April 2007

Abstract

Mesoporous $\text{Ce}_{0.8}\text{Sm}_{0.2}\text{O}_{1.9}$ (SDC) oxide with high surface area was prepared by a novel glycine-nitrate combustion process with in situ created nickel oxide as template, and applied as the catalytic layer for methane-fueled solid-oxide fuel cells (SOFCs) operated at reduced temperatures. The weight ratio of nickel oxide to SDC in the synthesis process was found to have significant effect on both the crystallite size and the textural properties of the resulted SDC powder. In particular, when it was at 9, the thermally stable and well-crystallized SDC powder showed a mesoporous structure with narrow pore-size distribution, high surface area ($77 \text{ m}^2 \text{ g}^{-1}$) and large pore volume ($\sim 0.2276 \text{ cm}^3 \text{ g}^{-1}$), even after the calcination at 700°C for 3 h. The mesoporous SDC was found to favor free gas diffusion with no gas diffusion polarization occurred even at high current density both for hydrogen and methane fuels. The SOFC with Ru impregnated mesoporous SDC catalytic layer displayed promising performance with a peak power density of $\sim 462 \text{ mW cm}^{-2}$ at 650°C .

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Keywords: Doped ceria; Mesoporous; Methane; Catalyst; Solid-oxide fuel cells

1. Introduction

Fuel cells are attractive power-generation technologies that directly and efficiently convert chemical energy to electrical power via the silent and environmental friendly electrochemical way. For most of the fuel cells, high-purity hydrogen is usually required as the fuel. However, due to the lack of public hydrogen production, storage and transportation infrastructure, the wide spread of hydrogen fuel cell may not be so optimistic within the next few decades. The much more matured infrastructure of gasoline and home gas system makes the hydrocarbon fuel cell more realistic in the near future. Solid-oxide fuel cells (SOFCs) are such kind of fuel cells that can operate directly with hydrocarbon fuel without the need of external reforming process. However, because of the less electrochemical activity of hydrocarbon than hydrogen over the typical nickel-based anode, a better performance electrochemical catalyst for hydrocarbon oxidation is necessary for practical application purpose. Ceria oxide has been found to be an excellent high-active catalyst for

hydrocarbon oxidation and cracking [1–5]. Electrochemical oxidation of hydrocarbons using ceria-based anode has also been successfully reported [6–9]. For example, it demonstrated that the addition of CeO_2 to the anode of Cu–Ytria stabilized zirconia/SDC cermets significantly enhanced the cell performance with hydrocarbon fuels [6].

Recently it was recognized that the ceria in mesoporous state could greatly improve the catalytic activity for hydrocarbon oxidation as compared to the bulk ceria [10–12]. Nonperiodic hierarchical mesoporosity coupled with nanocrystallinity has the potential to enhance mass transport, ionic and electronic mobility, and in particular maximize the three-phase-boundary (TPB) length in cathode or anode [12]. Significant improvement of fuel cell performance by applying mesoporous ceria catalytic layer has been demonstrated [13,14].

Up to now, undoped ceria was mostly applied as the electrocatalyst in SOFCs. Due to the poor ionic conductivity ($< 10^{-4} \text{ S cm}^{-1}$), it mainly functioned as the reforming/oxidation catalyst with improved gas diffusion properties but negligible contribution to the extension of the TPB length. The doping of CeO_2 with Sm_2O_3 or Gd_2O_3 leads to the substantially increase of the ionic conductivity to a level of $\sim 0.01 \text{ S cm}^{-1}$ at 600°C and $\sim 0.1 \text{ S cm}^{-1}$ at 800°C . Therefore, an increase

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of the fuel cell performance is possible from the extension of the TPB length by applying the doped ceria as the catalyst. For example, it was reported that the coating of the pores of Ni/ScSZ (scandia-stabilized zirconia) anode with 2 wt.% nano-sized $Gd_{0.2}Ce_{0.8}O_{1.9}$ conducting particles yielded much higher power density than that without the ceria modification [13]. The combination of high ionic conductivity and mesoporosity of the doped ceria may result in the substantially increase of the fuel cell performance. Therefore, in this study we are aimed at the synthesis of mesoporous SDC and the application of it as the electrocatalyst for reduced temperature methane-fueled SOFCs.

In literature, several methods have been used for the preparation of mesoporous ceria powders [15–19]. In those methods, fine and mesoporous ceria powders were produced by applying relatively expensive surfactants and organic templates. Furthermore, the synthesized powders always showed poor thermal and mechanical stability, which greatly limited their application in SOFCs. Here, we reported an innovative in situ inorganic templating process for the facile synthesis of mesoporous nano-crystalline SDC powder with high thermal stability. A glycine-nitrate combustion process (GNP) with in situ created nickel oxide as the agglomeration inhibitor and template was applied for the synthesis of mesoporous SDC powders. The method takes the advantages of cheap raw material, fast synthesis, recyclable inorganic template, high thermally stable and well-crystallized SDC. The as-prepared mesoporous SDC was loaded with Ru and applied as the catalytic layer for methane-fueled SOFCs operated at intermediate temperature with promising performance.

2. Experimental

2.1. Synthesis of SDC powder and fabrication of fuel cell MEA (Membrane-electrode-assembly) with catalyst layer

A modified glycine-nitrate process was applied for the synthesis of mesoporous SDC, which was schematically shown in Fig. 1. Stoichiometric amount of ceria nitrate ($Ce(NO_3)_3 \cdot 6H_2O$, >99%), samarium nitrate ($Sm(NO_3)_3 \cdot 6H_2O$, >99%) and nickel nitrate ($Ni(NO_3)_2 \cdot 6H_2O$, >99%), according to the aimed ratio, was prepared into a mixed aqueous solution in a Pyrex breaker. Glycine (NH_2-CH_2-COOH , 99%) was added to the mixed solution at the mole ratio of glycine:total metal ions at 2:1. The solution precursor was then heated at $\sim 90^\circ C$ over a hot plate under vigorous magnetic stirring until a gel was obtained, which was then transferred to a heated oven at $250^\circ C$ for a self-sustaining combustion. After the reaction, the primary products were further calcined at $700^\circ C$ under stagnant air for 3 h at the heating/cooling rate of $5^\circ C \text{ min}^{-1}$ to remove the carbon residues in the powders and form the well-crystallized powders. The as-obtained powders were then soaked with a $3 \text{ mol L}^{-1} HNO_3$ aqueous solution and heated at $90^\circ C$ for several hours under stirring. The nickel oxide in the powder was dissolved by the HNO_3 to leave a porous SDC powder, which was then filtered and washed with deionized water and ethanol. The obtained nickel nitrate solution can be recycled and applied as the raw material for the next synthesis.

The anode-supported cells with SDC as electrolyte were prepared by a dual dry-pressing process. Anode powder consisting

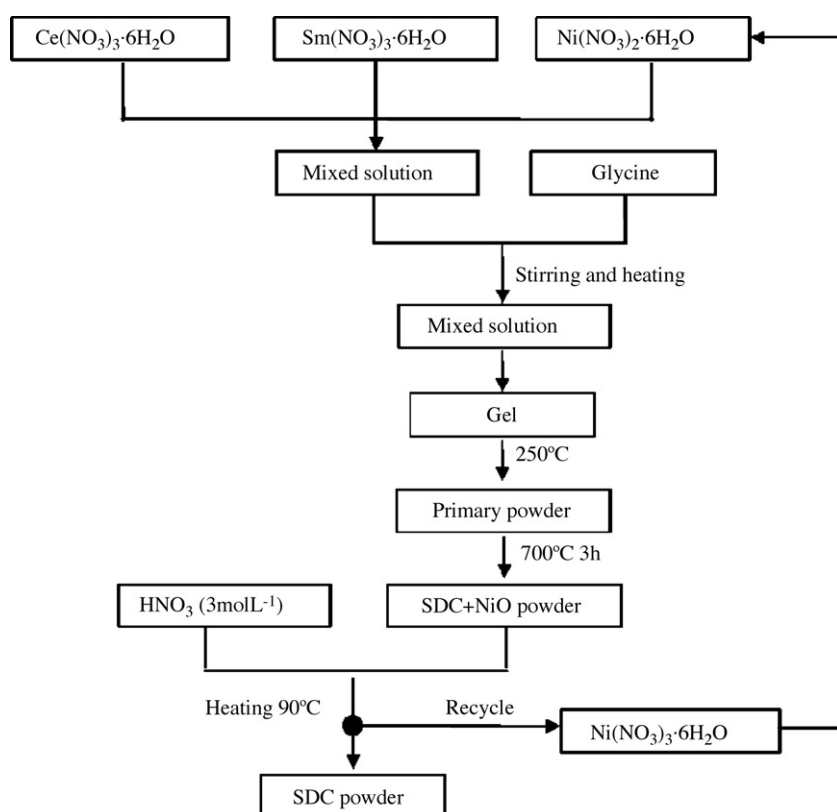


Fig. 1. Schematic diagram for the preparation of mesoporous SDC powder via the modified GNP method with in situ created NiO as template.

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