



RAPID COMMUNICATION

High-performance Ni nanocomposite anode fabricated from Gd-doped ceria nanocubes for low-temperature solid-oxide fuel cells



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Received 19 November 2013; received in revised form 4 March 2014; accepted 6 March 2014

Available online 18 March 2014

KEYWORDS

Ceria;
SOFCs;
Hybrid materials;
Nanocrystals;
Aero-sol;
Microstructures

Abstract

Gadolinium-doped ceria (GDC) nanocubes with highly reactive {001} facets were synthesized as an anode material for solid-oxide fuel cells by organic-ligand-assisted hydrothermal treatment with a water-soluble amino acid, 6-amino hexanoic acid (AHA). An aerosol technique was applied to fabricate a NiO-GDC nanocube composite with water as a green solvent. The NiO-GDC nanocube composite was easily sintered even at a temperature of 1100 °C, while the conventional NiO-GDC composite covered with the most stable {1 1 1} facets was sintered at 1300 °C. Sintering at such a low temperature inhibited undesirable coarsening of NiO and GDC particles, resulting in an enlarged, triple-phase boundary (TPB). The NiO-GDC nanocube composite anode with the enlarged TPB exhibited a rather low area specific resistance of 0.14 Ω cm² compared with the conventional NiO-GDC composite anode's resistance of 0.58 Ω cm² when operated at 600 °C.

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Introduction

Solid-oxide fuel cells (SOFCs) have received much attention as a promising next-generation electric power generation

systems, because of the former's high generation efficiency with direct conversion of chemical energy into electricity. The latest critical issue regarding SOFCs is the reduction of the operation temperature from 800–1000 °C to 600 °C or lower. Low-temperature operation enables shorter startup time and suppression of the degradation of SOFC components. For operation at lower temperatures, many researchers have reported the development of new SOFC materials

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[1-4], fabrication of a thinned electrolyte film [5-8], and microstructural control of the anode and cathode [9-13]. The sintering temperatures of anode materials such as Ni-GDC and Ni-YSZ cermet typically reach 1300 °C. Sintering at such a high temperature causes significant decrease in the triple-phase boundary (TPB) as a result of coarsening of the controlled size and microstructure of NiO and GDC. Therefore, simple ball milling is applied to fabrication of the anode structure, with grinding and mixing of commercial NiO and GDC powders in many cases [14-16]. Nanotechnologies using solution processes, especially for control of the crystallographic planes, are thus not employed for the synthesis of anode materials.

Zhang et al. have reported supercritical hydrothermal synthesis of tailor-made ceramic nanocrystals with organic ligands selectively adsorbed on particular crystal planes [17-19]. In their studies, CeO₂ nanocubes with highly reactive {0 0 1} crystal facets were synthesized by supercritical hydrothermal treatment with a carboxylic acid such as decanoic acid and hexanoic acid, and the {0 0 1} facets participated in extra-low-temperature oxygen storage capacity [19]. In our study, we focused on the singular functional material as a novel anode component, which was expected to contribute to the lowering of the working temperature because of the enlarged TPB caused by low-temperature anode fabrication.

We examined the organic-ligand-assisted hydrothermal synthesis of Gd³⁺-doped CeO₂ (GDC) nanocubes with a water-soluble amino acid, 6-amino hexanoic acid (AHA), for the control of crystal facets and the development of hydrophilicity. The NiO-GDC nanocube composite was fabricated by an aerosol process for homogeneous hybridizing of the GDC nanocubes and NiO nanoparticles. Fabrication of high-performance anode or cathode composites, such as NiO-Sm-doped ceria (NiO-SDC) and NiO-Sr- and Mg-doped LaGaO₃ (NiO-LSGM), or Sr-doped LaCoO₃ (LSC) and Sr-doped LaMnO₃-Y-stabilized zirconia (LSM-YSZ), with homogeneous hybridization and ideal fine microstructures using an aerosol process has been reported [20-22]. It is also an appropriate procedure for homogeneous hybridizing of highly dispersible GDC nanocubes and NiO.

Experimental section

Synthesis of GDC nanocubes and fabrication of NiO-GDC nanocube composite

Ce_{0.9}Gd_{0.1}O_{1.95} (GDC) nanocubes were synthesized by organic-ligand-assisted hydrothermal treatment. Ce(NO₃)₃·6H₂O (2.7 mmol, Wako Pure Chemical), Gd(NO₃)₃·6H₂O (0.3 mmol, Kanto Chemical), and AHA (18 mmol, Tokyo Chemical Industry) were dissolved in distilled water (29.5 mL). In addition, *tert*-butylamine (0.5 mL) was quickly added to the mixed solution with vigorous stirring. After 5 min of stirring, the GDC nanocube sol precursor was placed in a Teflon-lined stainless steel autoclave (total volume was 50 mL), where it was then heated at 180 °C for 24 h for the growth of {001} facets with coordination of AHA. The GDC nanocube precipitate was isolated by centrifuging and then washed three times with distilled water.

Fabrication of NiO-GDC nanocube composite by aerosol process

Fabrication of the NiO-GDC nanocube composite was carried out by a simple aerosol process [20-22]. The undried GDC nanocube specimen was dispersed in an aqueous solution of Ni(CH₃COO)₂ (0.05 M, 300 mL; molar ratio Ni/GDC=3.63:1), and the pH value was adjusted to 8 with aqueous NH₃. After 10 min of sonication, the suspension was atomized with an ultrasonic vibrator that was operated at 1.7 MHz. The atomized water droplets were transported to the reaction furnace using air as a carrier gas at a fixed flow rate of 1 L min⁻¹. The three reaction furnace temperatures were set at 200, 400, and 600 °C, respectively. The sample powders from the furnace outlet were collected on a membrane filter. For comparison, a NiO-GDC composite was prepared by a conventional aerosol process from an aqueous solution of Ni, Ce, and Gd nitrates.

Cell fabrication and cell performance tests

A NiO-GDC nanocube composite powder prepared by the aerosol process was mixed with PEG #400 that served as a binder. The paste was deposited by screen printing onto dense GDC electrolyte pellets (thickness: 400 μm, AGC Seimi Chemical) prepared by sintering at 1500 °C for 4 h. The NiO-GDC nanocube composite anode was prepared by sintering at various heating temperatures. After that, La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ} (LSCF) powder was prepared by the aerosol process as the cathode was prepared on the flip side by sintering at 900 °C for 2 h. The effective anode and cathode areas were both 0.282 cm² (6 mm in diameter). Pt wires were used as the reference electrode, and they were wound around the electrolyte pellets and fixed with Pt paste. Pt meshes were attached on top of each electrode to serve as the current collectors. The anode and cathode compartments were separated by a Pyrex glass ring gasket. The single-cell performance tests were carried out at 600 °C by supplying 3% moisturized hydrogen gas (H₂+3% H₂O) and air to the sides of the anode and cathode, respectively. The flow rates at both sides were adjusted to 50 cm³ min⁻¹. Current-voltage (*I*-*V*) measurements and electrochemical impedance spectroscopy (EIS) measurements were performed using an Electrochemie Autolab PGSTAT302N analyzer, with a frequency range of 0.01-10⁵ Hz and an amplitude of less than 30 mV. All measurements within this study were performed under open-circuit conditions.

Results and discussion

All samples for the current study were synthesized by organic-ligand-assisted hydrothermal treatment at 180 °C for 24 h with AHA. X-ray diffraction results confirmed that they had a fluorite structure irrespective of the amount of Gd-doping, and the diffraction peaks were also observed to shift to higher angles with increasing Gd concentration (Supporting information Figure S1). The morphologies of the CeO₂ nanocube and GDC nanocube particles were characterized by transmission electron microscope (TEM) analysis. As shown in Figure 1a, the CeO₂ nanocubes exhibited a highly dispersed cubic shape with an average

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