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Short communication

Synthesis and characterizations of composite particles for solid oxide fuel cell anodes by spray pyrolysis and intermediate temperature cell performance☆

Mitsunobu Kawano[a,*], Koji Hashino[a], Hiroyuki Yoshida[a], Hiroshi Ijichi[b], Seiji Takahashi[c], Seiichi Suda[c], Toru Inagaki[a]

[a] *Energy Use R&D Center, The Kansai Electric Power Co. Inc., 11-20 Nakoji 3-chome, Amagasaki, Hyogo 661-0974, Japan*
[b] *Kanden Power-tech Co. Ltd., 2-1-1800, Benten 1-chome, Minato-ku, Osaka 552-0007, Japan*
[c] *Japan Fine Ceramics Center, 4-1 Mutsuno 2-chome, Atsuta-ku, Nagoya, Aichi 456-8587, Japan*

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Abstract

$NiO–Ce_{0.8}Sm_{0.2}O_{1.9}$ (SDC) composite particles were synthesized using starting solutions containing the components for NiO–SDC and various amounts of nitric acid. It was found that the particles had a different surface morphology and specific surface area depending on the pH values of the starting solutions. SOFC single cell using the composite particles as an anode electrode was examined at an intermediate temperature to clarify the relationship between particle morphology and cell performance. High and consistent cell performance was obtained when the composite particles were synthesized using the solutions containing large amounts of nitric acid. It was considered that the morphology and the specific surface area of NiO–SDC composite particles played an important role realizing a high cell performance anode.

Keywords: Spray pyrolysis; Solid oxide fuel cell; Intermediate temperature; Composite particle

1. Introduction

The operation of solid oxide fuel cells (SOFCs) at intermediate temperatures between 600 °C and 800 °C provides several advantages (i.e. extensive selection of low-cost and high-performance component materials, high flexibility of SOFC structure, etc.). Since lowering the operation temperature increases not only the ohmic loss but also the polarization loss at the anode and the cathode, it is necessary to develop highly active electrodes that show sufficiently low polarizations at intermediate temperatures. We have developed a $Ni–Ce_{0.8}Sm_{0.2}O_{1.9}$ (SDC) cermet anode that shows high cell performance at temperatures below 800 °C, by using highly dispersed NiO–SDC composite particles synthesized by a spray pyrolysis method [1–3]. The synthetic conditions of the NiO–SDC composite particles, however, should be investigated further to realize the optimized anode properties. It was reported that the presence of additional anion species in the starting solutions for spray pyrolysis had significant influence on the properties of the particles [4,5]. In this paper, we investigate the effects of nitric acids added to the starting solution for spray pyrolysis on the properties of NiO–SDC composite particles. And, we describe the relationship between the properties of various NiO–SDC composite particles and the cell performance using these particles for the anode electrode.

2. Experimental

2.1. Synthesis of NiO–SDC composite particles by spray pyrolysis method

NiO–SDC composite particles were synthesized by spray pyrolysis method as shown schematically in Fig. 1. Aque-

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* Corresponding author. Tel.: +81 6 6494 9715; fax: +81 6 6494 9705.
E-mail address: kawano.mitsunobu@e2.kepco.co.jp (M. Kawano).

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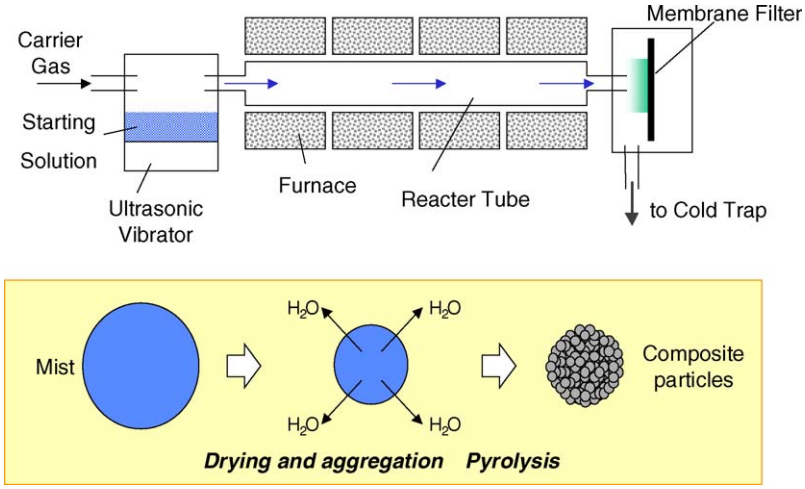

Fig. 1. Synthesis of composite particles by spray pyrolysis.

ous starting solutions containing the desired composition of corresponding cations were prepared by dissolving nickel acetate, cerium nitrate, samarium nitrate and various amounts of nitric acid (60 mass%) in the 500 ml starting solutions. These solutions were atomized with an ultrasonic vibrator operating at 1.65 MHz. The droplets were transported into a reaction furnace using air as a carrier gas at a fixed flow rate of 1 $dm^3\,min^{-1}$. The reaction furnace consisted of four independent heating zones, of which temperatures were set at 200 °C, 400 °C, 800 °C and 1000 °C, respectively. The NiO–SDC particles were collected using a membrane filter.

2.2. Characterization of NiO–SDC composite particles by spray pyrolysis method

X-ray diffraction (XRD, Shimadzu XRD-6000) analysis was carried out to confirm that the composite particles possessed the individual NiO and SDC crystal structures. Specific surface area of the NiO–SDC composite particles was measured by means of specific surface analyzer (YUASA-IONICS NOVA 2000). The microstructure of the particles was observed by scanning electron microscope (SEM, Hitachi S-2380N). The pH values of the solutions were measured with a pH meter (YOKOGAWA Model PH82) at room temperature. The pH values of the solutions were constant during the spray pyrolysis.

2.3. Cell fabrication process

For the electrolyte, we selected a lanthanum gallate-based electrolyte, which possesses high oxide ion conductivity [6–8]. Commercially available oxide powders (La_2O_3, $SrCO_3$, Ga_2O_3, MgO and CoO) were proportionally mixed, ball-milled and calcined in air to obtain the LSGMC powder with the chemical formula of $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.15}Co_{0.05}O_{3-\delta}$. The product of the calcination process was re-ground and mixed with an organic binder to be tape-casted into green sheet. Disks were then cut out and sintered in air at 1400–1500 °C for 6 h to obtain a 200 μm thick LSGMC electrolyte with relative density greater than 98%.

For the anode, the slurry containing the NiO–SDC composite particles was screen-printed onto the electrolyte, so as to give an effective electrode area of 2 cm^2, and then fired in air at 1280 °C for 3 h.

For the cathode, we selected samarium cobaltite compounds, which have been demonstrated to show very small polarization [9]. The slurry made of $Sm_{0.5}Sr_{0.5}CoO_{3-\delta}$ (SSC) powder was screen-printed onto the electrolyte to give the same effective electrode area of 2 cm^2. The final sintering was performed in air at a temperature of 1100 °C for 3 h to obtain the porous cathode.

2.4. Single cell performance test

The single cell tests were carried out at 750 °C. Air was used as an oxidant (31 ml min^{-1}), and dry hydrogen gas was used as a fuel (7.9 ml min^{-1}) (Fig. 2). NiO in the anode was reduced to Ni under the fuel atmosphere prior to measuring the power generation characteristics. For the electrochemical characterization, the current-interruption technique was

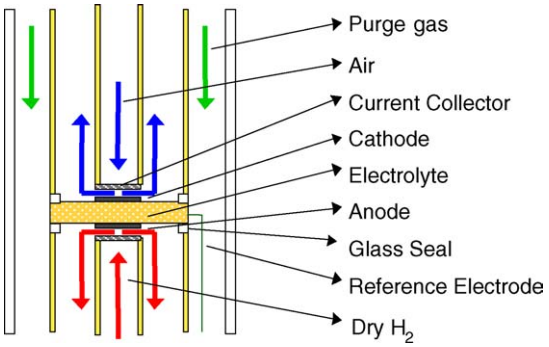

Fig. 2. Schematic view of testing apparatus for Ni–SDC/LSGMC/SSC.