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Effect of nanostructured anode functional layer thickness on the solid-oxide fuel cell performance in the intermediate temperature

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ABSTRACT

Effect of anode functional layer thickness on the performance of solid-oxide fuel cells (SOFCs) has been investigated in the intermediate temperatures of 600–650 °C. Three types of cells with different thickness (0, 4, 10 micron) of nanostructured anode functional layer (AFL) consisting of Ni-ScSZ (Scandia stabilized zirconia) are prepared. The SOFCs consist of Ni-3YSZ (3 mol% yttria stabilized zirconia) anode tube support with the AFL, ScSZ electrolyte, and LSCF (lanthanum strontium cobalt ferrite) and GDC (gadolinium doped ceria) mixture cathode. It is shown that the performance of the cell is improved as the thickness of the anode functional layer increases. Power densities of the cell with 10 micron thick AFL at 600 and 650 °C are shown to be 0.22 and 0.27 W/cm² at 0.75 V, respectively. According to impedance spectroscopy, improvement of both ohmic and polarization resistances has been observed by increasing the thickness of the AFL, suggesting that the AFL also acts as a better contact layer between the electrolyte and the anode support, and the effectiveness of the AFL by optimizing the thickness.

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Introduction

Solid oxide fuel cells (SOFCs) are high-temperature-operated fuel cell, typically operated at 700–800 °C, in turn, they have advantages such as high energy conversion efficiency and fuel flexibility [1–7]. Currently, enormous efforts are poured into the development of SOFCs for commercialization. In Japan, 700 W micro combined heat and power (CHP) system utilizing SOFCs, “ENE-Farm”, for households has been released in the

market [8]. Portable power devices for consumer electronics (from several W) have also been of interest for long life-time usage compared to battery. This is enabled by utilizing high energy density fuel such as butane (13.6 Wh/g), where the energy density of typical Li –battery ranged around 0.1–0.2 Wh/g [9].

Current challenges for SOFC development mainly focuses on improving the fuel cell performance for lowering the cost of the system and the operating temperature, improving the

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durability and redox performance, and user friendly features such quick start-up capability [10–13]. Among them, improving durability of SOFCs is one of the very important issues for commercialization and this factor can be improved by optimizing SOFC materials, as well as the cell and stack/module structure. Lowering the SOFC operating temperature leads to reduce the start-up time of the system, as well as the cost of materials used for SOFC system components. Typically, the operating temperature of SOFCs can be lowered by introducing either ultra-thin electrolytes using conventional materials [14], or new materials with higher ionic conductivity [15]. Both methods have been reported to be effective for lowering operating temperature even below 500 °C.

For realizing low cost, low temperature operable SOFCs, we have been focusing on the development of cost effective SOFC fabrication processes for realizing high performance micro-tubular and micro-honeycomb SOFCs [16–18], by utilizing powder-based ceramic slurry and extrusion technology. In our studies, so far, high performance micro-tubular and honeycomb SOFCs operable at 600 °C have been developed using conventional SOFC materials.

For further improvement of fuel cell performance, a variety of cell structures were proposed and demonstrated, and one of effective methods to improve the fuel cell performance was to introduce an anode functional layer between anode and electrolyte interface [19–21]. They reported that introducing anode functional layer (AFL thickness \approx 5 μ m) improved the fuel cell performance 1.5 to 2 times, however the electrode overpotential increased with increasing AFL thickness up to 20 μ m.

In this study, the effect of the AFL thickness was investigated in the micro-tubular supported cells at the intermediate temperatures below 700 °C. Micro-tubular cells with different thickness of anode functional layer were fabricated, consisting of Ni-3YSZ (3 mol% yttria stabilized zirconia) anode micro-tube support with Ni-ScSZ (scandia stabilized zirconia) nanostructured AFL, ScSZ electrolyte, and LSCF (lanthanum strontium cobalt ferrite) and GDC (gadolinium doped ceria) mixture cathode. The cell performance was investigated in the intermediate temperatures of 600 and 650 °C using impedance spectroscopy and correlated to the microstructure of the anode.

Experimental

Fabrication

The micro-tubular SOFC's we report here consist of Ni – 3 mol % yttria stabilized zirconia (3YSZ, TZ3Y grade, Tosoh) for the anode support, Ni -10 mol% Scandia stabilized zirconia (ScSZ, 10Sc1CeSZ grade, Daiichi-Kigenso Kagaku Kogyo Co., Ltd.) for the anode functional layer, ScSZ for the electrolyte, gadolinium doped ceria (GDC, Ce_{0.9}Gd_{0.1}O_{1.95} grade, Shin-Etsu Chemical Co., Ltd.) for the interlayer and La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-y} (LSCF) – GDC for the cathode. Anode green tubes were prepared using nickel oxide (NiO) powder (Sumitomo Mining Co., Ltd.), 3YSZ powder, carbon powder (Showa Denko Co., Ltd.), and cellulose binder (Yuken Kogyo Co., Ltd.) with a NiO:3YSZ ratio of 60:40 wt%. These powders were mixed for

1 h by a mixer, 5DMV-r (Dalton Co., Ltd.), and after adding the appropriate amount of water, were stirred for 30 min in a vacuum chamber. The mixture that was prepared from these powders was left for over 1 day for aging. Using the aforementioned mixture, the tubes were extruded from a metal mold (6.0 mm diameter with 4.0 mm diameter pin) using a piston cylinder type extruder (Ishikawa-Toki Tekko-sho Co., Ltd.). An anode functional layer (NiO:ScSZ = 60:40 wt%) and electrolyte layer was then coated onto each green anode tube by dip-coating slurries consisting of these materials, respectively, at the pulling speed of 1 mm/s. These slurries were prepared by mixing the designated powders, solvents (toluene and ethanol), binder (poly vinyl butyral), dispersant (polymer of an amine system) and plasticizer (dioctyl phthalate) for 48 h. The coated ScSZ layer was dried in air, and co-sintered with the anode tube at 1400 °C for 2 h in air. A GDC slurry was then dip-coated onto the electrolyte surface in the same manner, and sintered at 1250 °C for 2 h in air. Finally, a cathode slurry, mixture of La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-y} (LSCF) powder and the GDC powder (LSCF:GDC = 70:30 wt%), were dip-coated onto the GDC interlayer, and after drying in air, the cell was sintered at 1000 °C for 1 h. The final cell diameter was approximately 4.0 mm with an anode thickness of 0.3 mm. The thickness of the AFL varies from 0, 4, 10 micron, which were controlled by using slurries with different amount of powder content. For comparison, Ni-ScSZ anode tube supported cell without the AFL was also fabricated at 1350 °C for 2 h, followed by the same manner described above except the coating part of the AFL. Table 1 summarized the composition of each cell.

Characterization

The electrochemical performance of the cells was investigated using a potentiostat (Solartron 1287) and impedance analyzer (Solartron 1255B), at the cell temperature of 600 and 650 °C, which was monitored using a thermocouple placed close to the sample. The size of the cell was 4.0 mm in diameter and 50 mm in length with a cathode length of 40 mm, and an effective electrode area of 5 cm². Ag wire, which was fixed using Ag paste, was used for collecting current from the anode and cathode. The current from the anode side was collected only from an edge of the anode tube, whereby no conducting pastes were applied inside the tube. The current from the cathode side was collected from the whole cathode area. Hydrogen with 3% H₂O flowed inside the tubular cell at a flow rate of 50 mL/min. Air was supplied to the

Table 1 – Composition of the cells used in this study.

Cell	Anode support	AFL	Electrolyte	Cathode
AFL(0 μ m)	Ni-3YSZ	n/a	ScSZ	LSCF-GDC
AFL(4 μ m)	Ni-3YSZ	Ni-ScSZ	ScSZ	LSCF-GDC
AFL(10 μ m)	Ni-3YSZ	Ni-ScSZ	ScSZ	LSCF-GDC
Cell _{ref}	Ni-ScS	n/a	ScSZ	LSCF-GDC

AFL: anode functional layer, YSZ: yttria stabilized zirconia, ScSZ: Scandia stabilized zirconia, LSCF:LaSrCoFeO₃, GDC: gadolinium doped ceria.

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