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The effect of ceria content in nickel–ceria composite anode catalysts on the discharge performance for solid oxide fuel cells

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ABSTRACT

Optimum ceria content in nickel–ceria composite anode catalyst from the point of discharge performance is discussed. The ohmic loss increased when the ceria content was higher than 30 mol%. Even though the electrical conductivity of the anode decreased with increasing ceria content in the anode catalyst in association with decreasing nickel content, the ohmic loss was kept low until the ceria content was ≤ 30 mol% because the semiconducting ceria compensated for the decreased current path owing to the decreasing nickel content. The lowest activation loss was observed when the ceria content in the nickel anode catalyst was 30 mol% and the maximum activation loss was obtained for ceria content of 2 mol%. Ceria content in nickel anode influenced microstructure of the anode matrix. When the CeO₂ content was 2 mol%, sintering of anode catalyst was evident and the porosity of anode matrix was almost 57% - highest in this study. Whereas sintering of anode catalyst was not evident and the porosity of anode matrix was 46% when the ceria content in the nickel anode catalyst was 30 mol%. Activation loss was strongly influenced by microstructure of anode matrix, and highest activation loss when the CeO₂ content was 2 mol% was owing to the inappropriate microstructure for electrochemical reaction: sintering of the anode catalyst and excessive porosity of the anode.

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Introduction

One of the most attractive features of solid oxide fuel cells (SOFCs) is the flexibility to use a wide range of fuels owing to internal reforming and electrochemical oxidation of hydrocarbons [1,2]: this flexibility allows both conventional

hydrocarbons and renewables (e.g., biogas, syngas from biomass) to be used as fuels. However, metal nickel, the most common anode catalyst used in SOFCs, is susceptible to carbon deposition [2] and nickel depletion [3] when hydrocarbon fuels are used. Because the deposition of carbon or the depletion of nickel can cause irreversible deterioration of the

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Nomenclature

J	Current density [$A\ cm^{-2}$]
V	Voltage [V]
Z'	Real part of impedance [$\text{ohm}\ cm^2$]
Z''	Imaginary part of impedance [$\text{ohm}\ cm^2$]
Z_{ohm}	Ohmic loss [$\text{ohm}\ cm^2$]
Z_{act}	Activation loss [$\text{ohm}\ cm^2$]

nickel anode, these constraints must be overcome before their practical application in a direct hydrocarbon solid oxide fuel cell; to overcome these constraints, extensive efforts have been made during the last few decades [2,4–6].

For electrode catalysts, two types of alternative materials to metal nickel have been investigated: composite oxides [7–12] and metal alloys [2,13–15]. However, even though these materials are highly durable with hydrocarbon fuel, their discharge performance is not very high or there are other limitations for their use.

Recently, nickel and functional oxide composites have received attention as alternative anode materials. Chung et al. [16] used a nickel–manganese oxide composite anode catalyst and evaluated its discharge performance in direct-methane SOFCs: coking tolerance was improved and negligible coke formation was observed when a $Ni_{0.9}(MnO)_{0.1}$ anode catalyst was used. Zhao et al. evaluated the effect of $Mn_{1.5}Co_{1.5}O_4$ -SDC composite catalyst layer over Ni/SDC anode on stability and discharge performance [17], then their anode supported single cell showed excellent performance. In addition, Patil, et al. [18] and Wang [19] used Ni- Al_2O_3 composite as anode catalyst for a hydrocarbon fueled SOFC. Yang applied a MgO-modified Ni cermet anode for hydrocarbon fueled solid oxide fuel cells [20]. Islam et al. added barium oxide into Ni/YSZ anode by microwave irradiation, and the anode showed lower carbon accumulation [21]. Takeguchi et al. used MgO, CaO, SrO and CeO_2 as additive for Ni-YSZ cermet, and they showed CaO addition was effective in suppressing carbon deposition [22]. Other cases are reviewed by Faes et al. [23]. Nickel–ceria composite anode catalysts have also been applied in direct methane SOFCs [24,25]. Namioka et al. [26] used a nickel–ceria composite as an anode catalyst for SOFCs and investigated the influence on discharge performance of trace amounts of light hydrocarbons in wet hydrogen: the terminal voltage between the anode and the reference electrode was stable and nickel depletion was suppressed relative to those in nickel electrode catalysts.

Metal oxide composite anode catalysts display other interesting properties. For example, the discharge performance of composite anode catalysts is improved compared with that of pure nickel catalysts. Y. S. Chung et al. [16] showed that the power density of the SOFCs increased when manganese oxide was used in the anode; the manganese oxide enhanced electrochemical oxidation and could control the progression of nickel sintering. Zhao et al. also evaluate the performance of $Mn_{1.5}Co_{1.5}O_4$ modified Ni-SDC anode. They showed that $Mn_{1.5}Co_{1.5}O_4$ modification was effective for reducing polarization resistance due to increasing porosity [27]. He et al. [28] used various lanthanide oxides to prepare

nickel and functional oxide composites and evaluated their discharge performance: ceria was the best functional oxide, which they attributed to the optimized microstructure by increasing the porosity. Qiao et al. [29] used a nickel–ceria composite electrode catalyst prepared by an impregnation method and evaluated its discharge performance: the performance was influenced by the nickel–ceria ratio. Although they proposed that ceria facilitated the adsorption and dissociation of hydrogen and prevented the sintering of nickel particles, they showed little evidence for this.

As described above, the addition of functional oxides, especially ceria, facilitates the improvement of the discharge performance of anodes used in SOFCs. Several research groups have reported that ceria is an excellent catalyst for the electrochemical oxidation reaction of hydrocarbons through a reversible CeO_2 – Ce_2O_3 transition [6,30]; however, the reason for the improvement of the discharge performance of the nickel–ceria composite catalyst compared with that of the metal nickel catalyst is still not understood.

In this study, we evaluate the effect of the nickel–ceria ratio on the discharge performance of the anode and discuss the reason for the change in the discharge performance in terms of the electrical conductivity, microstructure, and morphology of the anode.

Material and methods

Anode catalyst preparation

One kind of nickel oxide (NiO) particles and six kinds of nickel oxide–ceria (NiO– CeO_2) composite particles were used as raw materials for the anode catalyst. The NiO powder was fabricated from nickel nitrate (Kanto Chemical Co., Inc., Tokyo, Japan) with glycine (Sigma–Aldrich Japan, Tokyo, Japan) solution through a glycine nitrate process (GNP). NiO– CeO_2 composite particles were also fabricated through a GNP from $Ni(NO_3)_2 \cdot 6H_2O$, $Ce(NO_3)_3 \cdot 6H_2O$, and glycine (both from Sigma–Aldrich Japan, Tokyo, Japan). Both kinds electrode catalyst particles were heated to 1073 K in air for 5 h to eliminate any carbonaceous residues.

The molar ratios of NiO and CeO_2 in the powders were confirmed by X-ray fluorescence (RIX2100, RIGAKU, Tokyo, Japan): the obtained results (data not shown) corresponded almost exactly to the target composition.

An X-ray powder diffractometer (RINT2100, Rigaku Co., Tokyo, Japan) was used for phase identification and to estimate the crystallite diameter with Scherrer's equation. The samples for X-ray diffraction analysis were calcined at 1573 K for 10 h and reduced with hydrogen at 1173 K for 2 h. These calcination and reduction conditions correspond to the fabrication conditions of the single cells used for the discharge experiments. All the peaks obtained were identified as Ni and CeO_2 , and no other peaks including complex oxides were detected [26].

Cermet anode material and composition

The cermet anode used in this study was a mixture of anode catalyst and 10Sc1CeSZ (ScSZ) electrolyte particles

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