

Investigation of Reducing In-Plane Resistance of Nickel Oxide-Samaria-Doped Ceria Anode in Thin-Film Solid Oxide Fuel Cells

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Abstract: Metal/NiO-Samarium-doped ceria (SDC) nano-composite thin film anodes were deposited on anodic aluminum oxide by co-sputtering to enhance the in-plane current-collecting ability and investigated by varying the composition of metal materials (Pt and Au). Full fuel cells with these nano-composites were fabricated and tested at 500 °C. Columnar anodes with a sponge structure were fabricated by varying the DC sputtering source power and they were thermally stable at the operating temperature. By adding metal material, the ohmic resistance, including the current collecting resistance, was drastically reduced and the polarization resistance also decreased. The nano-composite electrode with a Pt content of 61 at% showed the highest performance, which is a maximum power density of 212.5 mW/cm² at 500 °C. In addition, Au was considered to reduce the current collecting resistance and the corresponding power density was 3 times higher than that with the NiO-SDC anode.

Keywords: NiO-SDC; co-sputtering; thin-film solid oxide fuel cell; anodic aluminum oxide

1. Introduction

Solid oxide fuel cells (SOFCs) are attracting great attention as a next-generation energy conversion device due to their high energy conversion efficiency, low pollution, and fuel flexibility [1]. However, due to the high operating temperature, it has been proven that it is difficult to overcome the problems of sealing, insulation, and material stability, which were big obstacles to move toward commercially viable products [2]. Therefore, many types of research have been studied to reduce the operating temperature of SOFCs to the low-temperature range below 500 °C. More research should be done on low-temperature SOFCs (LT-SOFCs) to make thinner electrolytes and make the ohmic resistance lower. Therefore, LT-SOFC with a sub-micrometer-thick electrolyte fabricated by a vacuum-based deposition process is called a thin-film solid oxide fuel cell (TF-SOFC) [3,4]. One of the

typical fabrication methods for TF-SOFC, in which a thin film is directly deposited on a nano-porous template, such as anodic aluminum oxide (AAO), has been extensively studied due to the simple fabrication steps by consecutive deposition processes [5–8].

Nickel-based cermetes have been typically used as a mixed ionic electronic catalyst at the anode to increase the triple phase boundaries (TPBs), which is an important junction directly related with cell performances. Nickel plays a role as an electron conductor and hydrogen oxidation reaction catalyst, while a combined oxide serves as an oxygen ion conductor. As a result, the nickel-based cermet extends TPB and enhanced the thermal stability of the Ni catalyst. There are two mainstream Ni-based cermet catalysts: Zirconia-based composites and ceria-based composites. Ni-YSZ is a typical zirconia-based composite and it is one of the most widely used anode materials for SOFCs. Yet, there is a low ionic conductivity issue for Ni-YSZ catalysts despite their high compatibility with the YSZ electrolyte. On the other hand, ceria-based nanocomposites, such as Ni-SDC, have higher ionic conductivity as well as electronic conductivity under the reducing environment [9]. Despite these factors, however, the properties of the nickel-based cermet in a relatively low operating temperature ($<500\text{ }^{\circ}\text{C}$) and very thin ($<200\text{ nm}$) film thickness increase the in-plane resistance at the anode in TF-SOFCs [10,11]. Among the vacuum-based deposition techniques, such as atomic layer deposition, pulsed laser deposition, and sputtering, sputtering is the most common method because it is an easy process with any target material and large area deposition by controlling the process parameters, including the gas pressures and tunable DC/RF powers [3,12]. To achieve superior electrical conductivity with a highly porous structure, the co-sputtering method is the best choice [13,14]. Lim et al. reported TF-SOFCs with an Ni-SDC cermet anode deposited by sputtering [14]. In the study, a cell with a carefully engineered Ni-SDC anode (178 mW/cm^2) outperformed the other cell with a Pt anode (113 mW/cm^2). Nevertheless, the Ni-SDC cell showed limitations in terms of the electric conductivity, which showed a higher ohmic resistance in electrochemical impedance spectroscopy (EIS) Nyquist plots than the Pt anode cell despite the higher power density. In this work, we investigated a new nano-composite anode consisting of Pt and Ni-based cermet by co-sputtering to enhance the electrochemical performances of the TF-SOFCs by improving not only the catalytic activity but also the electric conductivity.

2. Experimental Methods

Anodic aluminum oxide (AAO, Synkera, Longmont, CO, USA) was utilized as a TF-SOFC substrate, which is a dimension of $1 \times 1\text{ cm}^2$, a thickness of $100\text{ }\mu\text{m}$, and a pore size of 80 nm . NiO/Samaria-doped ceria (SDC) and Pt were deposited on the AAO using a commercial magnetron sputter (AT-12, A-Tech system Co., Incheon, Korea).

The sputtering conditions for the pure Pt anode were set as follows: The direct current (DC) sputtering power for Pt deposition, pressure of Ar gas, and deposition time were 100 W , 4 Pa (30 mTorr), and 13 min , respectively. On the other hand, the pure NiO-SDC anode was made by using the radio frequency (RF) sputtering method using NiO-SDC ($\text{NiO}-(\text{CeO}_2)_{0.9}(\text{SmO}_{1.5})_{0.1}$, 50:50 wt% (RND Korea Co., Seoul, Korea)) target. The RF sputtering power, pressure of mixed Ar/ O_2 gas (Ar: 80%, O_2 : 20%), and deposition time were 150 W , 4 Pa (30 mTorr), and 5 h , respectively. These anode deposition processes using the sputtering technique were done without substrate heating.

For the metal/NiO-SDC composite anode, the composition variance of Pt/NiO-SDC was prepared by varying the DC power of Pt. All of the sputtering process was done at room temperature. The RF power of NiO-SDC deposition was fixed at 150 W while the DC powers for Pt deposition were 15 , 30 , and 50 W . Here, we called the corresponding samples Pt15, Pt30, and Pt50, respectively. In addition to Pt, Au was also used for comparison, where a DC power of 15 W was applied for DC sputtering of Au. This Au cell was named Au15. All samples were the same thickness of 300 nm . The pressure of mixed Ar/ O_2 gas was 4 Pa (30 mTorr). The effective area of the electrode was 1 mm^2 . Figure 1 shows a schematic of the fabricated cell configuration.

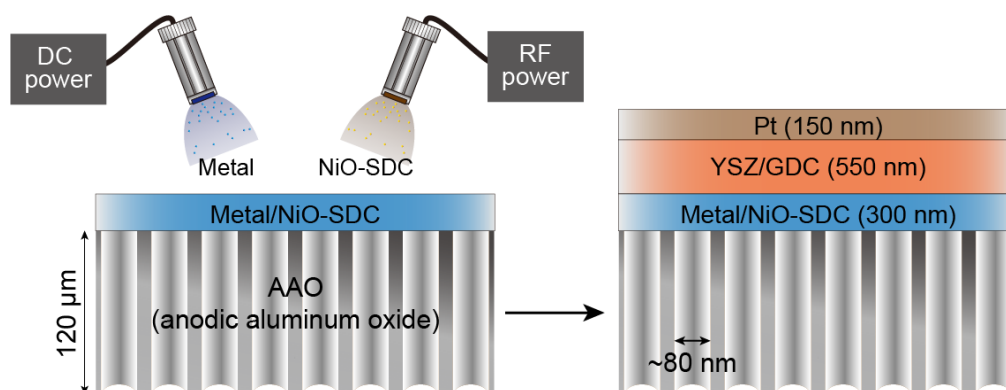


Figure 1. Schematic of the fabricated cell configuration. The membrane-electrode assembly (MEA) on the anodic aluminum oxide (AAO) consists of 300-nm-thick metal/NiO-SDC anode, 550-nm-thick YSZ/GDC bi-electrolyte, and 150-nm-thick Pt cathode.

Above the anode, the electrolyte consisted of, in the order of, a 450-nm-thick dense YSZ and a 100-nm-thick dense gadolinia-doped ceria (GDC), while the cathode was a 150-nm-thick porous Pt. A metal Y/Zr target (16% Y, 84% Zr, Advantec Korea Co., Seoul, Korea) and a ceramic GDC target $((\text{CeO}_2)_{0.9}(\text{GdO}_{1.5})_{0.1})$, RND Korea Co., Seoul, Korea) were RF sputtered to deposit YSZ and GDC thin film, respectively, and the process was also carried out at room temperature. The RF powers of 200 and 50 W were applied for the YSZ and GDC thin films, respectively and 0.67 Pa (5 mTorr) of Ar/O_2 (8:2) mixture gas was applied inside the sputter chamber.

Pt was used as a cathode material and deposited in a thin-film form using the sputtering method. The DC power source was utilized for the sputtering of the Pt target and the deposition process was carried out at room temperature. DC power of 100 W was applied and the atmosphere inside the chamber was maintained with 12 Pa (90 mTorr) of pure Ar gas.

Electrolyte was deposited on an 8 mm × 8 mm area on the AAO template, so a 1-mm-wide perimeter of the anode was exposed. The Pt cathode with a 1 mm × 1 mm active area was deposited on the electrolyte through sputtering using a shadow mask.

The TF-SOFCs were tested at 500 °C while the cells were fed with 100 sccm of 3% wet H_2 gas and exposed to ambient air on the other side. The anode was electrically connected to a stainless steel jig via Ag paste. Ceramic sealant was additionally applied on the Ag paste to ensure gas-tight sealing between the TF-SOFC and the jig. The external circuit was completed by contacting the cathode with a microprobe. The schematic and detailed explanation of the experimental set up for TF-SOFCs can be further referred to from the previously published literature from our research team [15].

The elements of an anode material were analyzed by X-ray fluorescence spectroscopy (XRF, XRF-1800, Shimadzu Co., Kyoto, Japan). Additionally, the surface morphologies of the anodes were observed by a field emission scanning electron microscope (FE-SEM) and focus ion beam-scanning electron microscope (FIB-SEM). The TF-SOFCs were electrochemically characterized by a commercial potentiostat, Solatron 1287/1260 (Solartron Analytical, Hampshire, UK). The current-voltage behaviors of the cells were measured through potentiodynamic mode with a scan rate of 30 mV/s. Electrochemical impedance spectroscopy (EIS) was measured with an amplitude of 30 mV, bias voltage of 0.7 V, and frequency range from 1 MHz to 2 Hz.

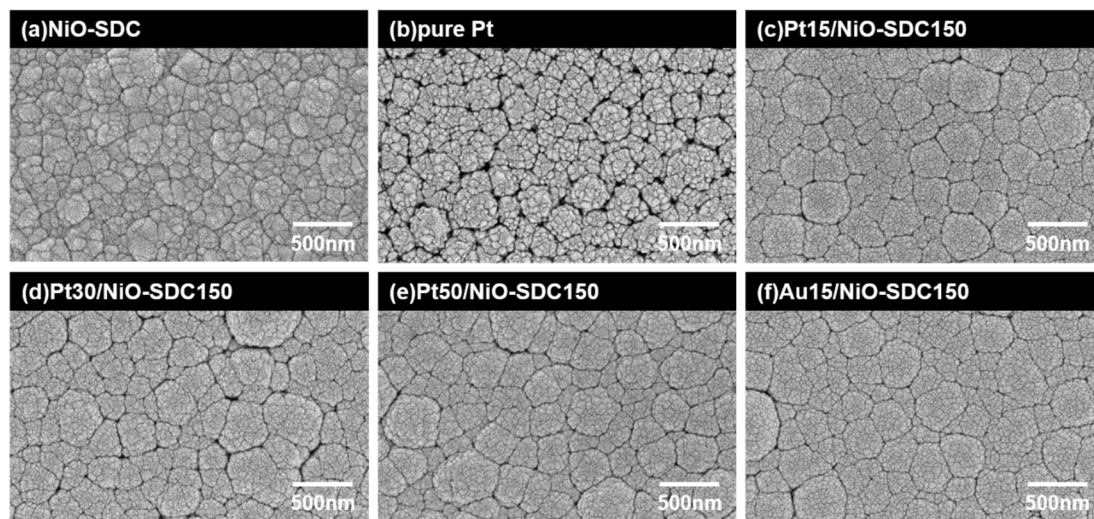
3. Results and Discussion

Table 1 shows the chemical compositions of the anode materials (Pt15, Pt30, Pt50, and Au15), analyzed by XRF. The atomic % of the deposited Pt increases with the rise of the DC sputtering power.

Table 1. The metal elements constituting the anodes analyzed by XRF (atomic %).

Sample	Pt	Ni	Ce	Sm
Pt15	60.7	24.9	11.1	3.3
Pt30	75.9	15.3	6.1	2.6
Pt50	85.2	9.1	4.2	1.5
Au15	63.7	22.1	10.3	3.9

Figure 2 shows the surface morphologies of the deposited anodes of Pt and NiO-SDC: (a) NiO-SDC only, (b) pure Pt, (c) co-sputtered Pt15/NiO-SDC, (d) co-sputtered Pt30/NiO-SDC, and (e) co-sputtered Pt50/NiO-SDC, and (f) co-sputtered Au15/NiO-SDC. As shown in Figure 2a, the pores are significantly clogged by the deposited NiO-SDC particles. Throughout the reduction reaction of the deposited NiO-SDC, it is believed that the microstructure of the deposited grains will transform, and thereby the nano-pores for hydrogen diffusion to TPBs will be gradually generated. However, the morphological structure of the pure Pt sample shows finely nano-sized pores of ~50 nm grown on the AAO template in Figure 2b. The other composite anodes, Figure 2c–e, have relatively denser surface structures than that of the pure Pt.

**Figure 2.** Surface morphologies of as deposited (a) NiO-SDC, (b) pure Pt, (c) Pt15/NiO-SDC150, (d) Pt30/NiO-SDC150, (e) Pt50/NiO-SDC150, and (f) Au 15/NiO-SDC150 on the AAO substrate.

The pure Pt anode (Figure 2b) has sub-50 nm pores grown on the AAO, and it is easy to supply hydrogen to the TPB interface between the anode and the electrolyte. In Figure 2c–e, Pt15, Pt30, and Pt50 have similar nano-porous TPB interfaces and also a similar porosity of 5%, which was analyzed by image J software. The grain sizes of Pt15, Pt30, and Pt50 were relatively larger than those of pure Pt, meaning a decreased TPB density.

Figure 3 shows the cross-sectional structures of the anodes after an hour operation at 500 °C. The porous anode also grows into a columnar structure, thereby the pores of the AAO are extended and formed in the anode. Figure 3a,b show some columnar structures merging as they grow, so the size of the pores between these columnar structures dwindles. However, Figure 3c,d,f show that the columnar structures in these anodes are well preserved. Small holes are found in each columnar structure, which can facilitate the fuel supply. As the metal ratio of the anode increases, the porosity of the anode decreases and finally the anode structure in Figure 3e is similar to the pure Pt anode structure in Figure 3b.

To investigate how the anode works, fuel cells prepared with different anode compositions were electrochemically tested at 500 °C. Figure 4a shows the polarization curves of the TF-SOFCs, and the

open circuit voltage (OCV), maximum power density, ohmic resistance, and polarization resistance are summarized in Table 2. Pt15 shows the highest maximum power density of 212.5 mW/cm² among the cells. The cell with the NiO-SDC anode exhibits a rapid voltage drop and the lowest maximum power density of 32.6 mW/cm². As shown in Table 2, the thin-film NiO-SDC anode suffers from a high current collecting resistance, and the steep slope of the polarization curve is caused by the high ohmic resistance of 4.92 $\Omega\cdot\text{cm}^2$ from the current collecting in the NiO-SDC anode. The measured OCVs of the NiO-SDC, pure Pt, Pt15, Pt30, and Pt50 were all above 1 V, but the pure Pt and Pt50 showed slightly lower OCV than any of the other cells, possibly due to minor pinhole defects within an electrolyte. Pinhole creation may result from two factors: One is contamination during the electrolyte deposition process, while the other one is that the agglomeration of the anodes during start up, which affects the microstructure of the electrolytes to make pinholes. Platinum suffers from microstructural degradation over time at the temperature caused by Ostwald ripening and coarsening [16,17].

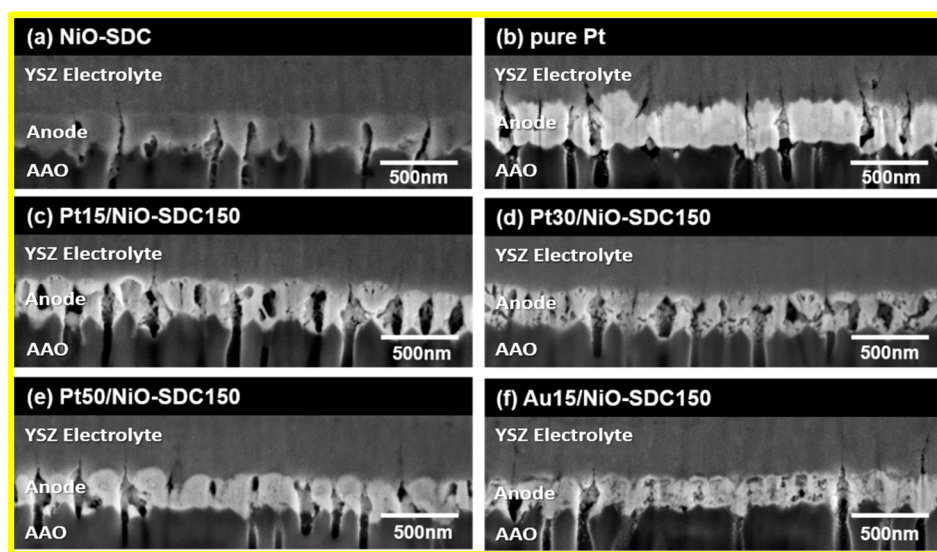


Figure 3. Cross-sectional structures of the anodes (a) NiO-SDC, (b) pure Pt, (c) Pt15/NiO-SDC150, (d) Pt30/NiO-SDC150, (e) Pt50/NiO-SDC150, and (f) Au15/NiO-SDC150 on the AAO substrate.

Figure 4b shows the EIS analysis with a 0.7 V bias voltage at 500 °C. Fitted curves were obtained by a least-square fitting (Z-Plot, Scribner Associates Inc., Southern Pines, NC, USA) to the equivalent circuit with the original EIS data of each cell. The equivalent circuit consisted of three series of elements and these elements are comprised of a parallel resistor (R) and a constant phase element (CPE) as shown in the inset of Figure 4b. Two slightly overlapped semicircles are observed in the Nyquist plot and the first x-intercept is considered as ohmic resistance mainly from the electrolyte and anode in-plane resistance, while the diameter of the first small semicircle in the high-frequency range is considered as an electrolyte grain response. The second large semicircle is likely to be a polarization response from an anode and a cathode in intermediate and low-frequency ranges [18,19].

Table 2. Electrochemical performances of the TF-SOFCs with different nano-composite anodes.

Case	OCV (V)	Max. Power Density (mW/cm ²)	Ohmic Resistance ($\Omega\cdot\text{cm}^2$)	Polarization Resistance ($\Omega\cdot\text{cm}^2$)
NiO-SDC	1.10	32.6	4.92	5.15
Pure Pt	1.00	189.3	0.34	1.38
Pt15	1.13	212.5	0.71	0.84
Pt30	1.09	192.7	0.64	0.96
Pt50	1.03	150.3	0.62	1.13
Au15	1.15	153.1	0.55	1.52

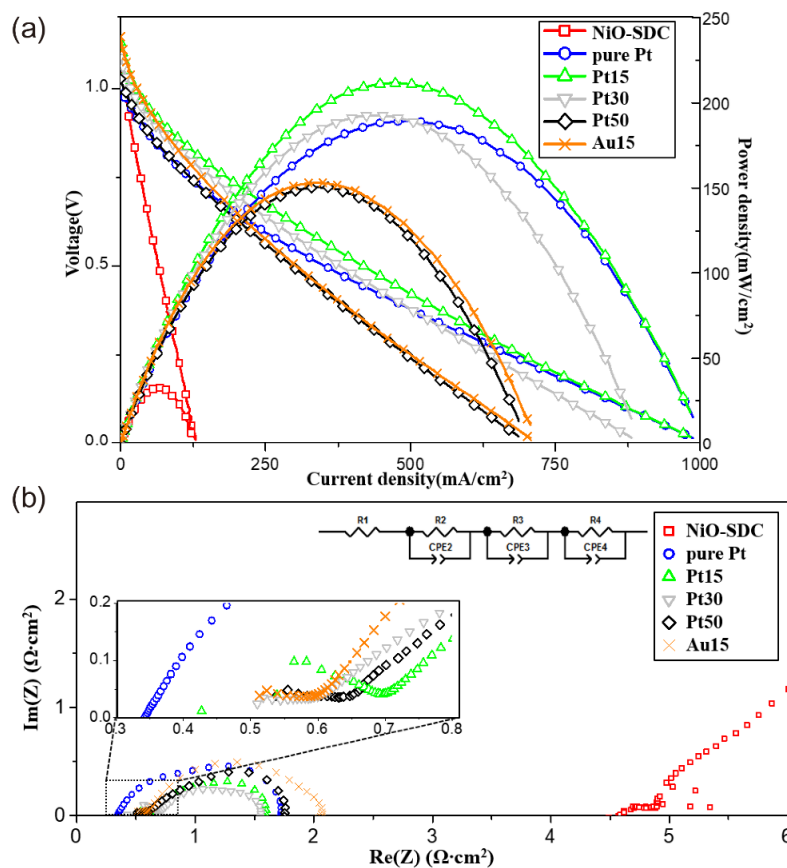


Figure 4. (a) Polarization curves of TF-SOFCs at 500 °C with anodes of NiO-SDC, pure Pt, Pt15/NiO-SDC, Pt30/NiO-SDC, Pt50/NiO-SDC, and Au15/NiO-SDC. (b) Nyquist plots of the impedance spectra with the corresponding equivalent circuit model at 0.7 V bias and 500 °C.

The ohmic resistance of the pure Pt anode is $0.34 \Omega\cdot\text{cm}^2$, which is only 0.06 times that of the NiO-SDC anode cell. Therefore, adding Pt into the NiO-SDC anode significantly reduces the in-plane resistance and enhances the hydrogen oxidation reaction rates, and consequently induces higher fuel cell performance. Furthermore, as it was confirmed in Figures 2 and 3, adding Pt to NiO-SDC enhances the porosity of the anodes, thereby contributing to lower polarization resistance since it secures a wider TPB area. In the case of Au15, the polarization resistance is about twice that of Pt15 due to an insufficient catalyst content for catalytic activity although Au has about four times higher electrical conductivity than that of Pt. It symbolizes that Pt affects not only the electric conductivity of the anode but also the catalytic kinetics of the anode. Besides, analysis of the impedance reveals that the higher the Pt contents in the anode, the higher the polarization resistance, and finally, it is closer to the pure Pt anode. This is likely due to the decrease of the TPB density resulting from morphological degradation as well as the different microstructure they have.

4. Conclusions

To reduce the high in-plane resistance of thin-film anodes composed of metal and NiO-SDC, Pt and Au, as the catalyst and electronic conductor, were co-sputtered with NiO-SDC. The Pt/NiO-SDC composite anode with Pt of 60.7 at% showed the highest power density due to the lowest polarization resistance. It was revealed that the higher the Pt contents, the lower the ohmic resistance and the higher the polarization resistance in Pt/NiO-SDC composite anodes. A high Pt content in the composite anode induced agglomeration of the Pt electrode and a reduction of the TPB area, thereby resulting in low fuel cell performances. In the Pt/NiO-SDC anodes and Au/NiO-SDC anode, each pillar of the anode

has a unique sponge-like structure with nano-scale holes. These unique structures were well preserved at an operating temperature of 500 °C and were expected to facilitate the gas supply.

Author Contributions: Y.K. and S.L. as the first author equally contributed to this work. They established cell design and conduct experiment for the current manuscript. G.Y.C., W.Y., and Y.L. gave their efforts to analyze experimental results and edit all figures and tables. I.C., J.D.B., and S.W.C. as co-corresponding authors provided helpful comments for improving the quality of the manuscript and contributed the supervision. All authors have read and agreed to the published version of the manuscript.

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