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Effect of Different Number of Coating for Anode Functional Layer on the Performance of Proton Ceramic Fuel Cell

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ABSTRACT

The effect of different numbers of anode functional layers (AFL) on the performance of an anode-supported proton ceramic fuel cell (PCFC) was investigated. Button cells of NiO-BCZY (50:50) | BCZY | LSCF (BCZY = $BaCe_{0.54}Zr_{0.36}Y_{0.1}O_{2.95}$) were fabricated with 3, 6, and 9 layers of AFL consisting of NiO-BCZY (10:90). Microstructural images clearly show that the button cell with 3 AFL layers exhibits better contact between the anode and electrolyte layers. Both the polarization resistance (RP) and ohmic resistance (Ro) of cell A (3 layers) were lower than those of cell B (6 layers) and cell C (9 layers). At 800 °C, the RP decreased from 79.6 to 19.3 Ω·cm² from cell C to cell A, respectively. Similarly, RP for cell B was 11.2 Ω·cm², while cell A showed only 9.2 Ω·cm². These results suggest that increasing the number of AFLs may reduce cell performance by hindering hydrogen ion diffusion. Therefore, three AFL layers provide the optimum structure for minimizing polarization resistance and maximizing PCFC performance.

1. INTRODUCTION

Towards the worldwide implementation of the hydrogen energy industry, upgrading hydrogen energy devices to increase their performance is the focus of various research groups. At the same time, the fabrication cost and operational parameters of hydrogen energy devices must be optimized. Efforts to reduce the operating temperature of high-temperature solid oxide fuel cells (HT-SOFCs) to intermediate and low-temperature ranges (IT-SOFC and LT-SOFC) have demonstrated considerable advantages for energy and cost management. Proton ceramic fuel cells (PCFCs) represent one such category, using a proton-conducting electrolyte instead of an oxide ion conductor. $BaCe_{0.54}Zr_{0.36}Y_{0.1}O_{2.95}$ (BCZY) is one such proton conductor that has delivered high power density in single-cell applications. One effective strategy to enhance SOFC performance is through the design of the anode functional layer (AFL). While AFL design has been well-studied for SOFCs with oxide ion conductors, studies on AFL design for BCZY-based PCFCs remain limited and underreported [1-2].

Conventionally, anode-supported SOFC was consecutively fabricated with three layers of anode substrate, electrolyte, and cathode. However, several problems such as cracks and disruptions of pores appeared at the interface of the anode substrate and electrolyte layer [3]. These issues motivated us to introduce the AFL between the two layers. The high ohmic resistance of the cell resulted from the poor contact resistance between the anode substrate and electrolyte layer, usually caused by large pores from the reduction of NiO to Ni in a hydrogen environment. The AFL that was fabricated as an interlayer usually has a smaller grain size than anode substrates and will reduce the size of the pores at the anode and electrolyte

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interface and this will reduce ohmic resistance [4]. Optimum AFL is fabricated to optimize the fuel gas transfer rate in the anode, at the same time preventing ohmic polarization loss from disrupted anode and electrolyte interface. The AFL can reduce anode polarization resistance by enhancing the length of Triple Phase Boundary (TPB), where protons will transfer through the interface of an anode and AFL and electrolyte layer, to the cathode layer and will meet oxygen ion-producing water and electrons. These electrons will be measured as current produced by the cell [3-5].

Varying the number of AFL does affect the cell performance in terms of microstructure and electrochemical properties [6]. An effort to find the optimum number of AFL for a specific cell's material is necessary. Some of the literature showed that a higher number of AFL will increase the open-circuit voltage (OCV) and decrease the local temperature near the electrolyte [7]. The optimization of the number of AFL plays an important role in two electrochemical reactions, which are the AFL load transfer response and the AFL diffusion-coupled load transfer response. [8-10]. Thus, this study aims to characterize fabricated button cells' microstructure and electrochemical performance at three different coating numbers.

2. Materials and Methods

2.1 Materials Synthesis and Slurry Preparation

The materials used in this work were previously synthesized in-house using the sol-gel method. These include yttrium-doped barium cerate–zirconate, $BaCe_{0.54}Zr_{0.36}Y_{0.1}O_{2.95}$ (BCZY), and iron-doped lanthanum strontium cobaltite, $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF), which were used as the electrolyte and cathode materials, respectively. For the anode substrate, commercial nickel oxide (NiO) (ACROS) was mixed and ground with sol-gel-synthesized BCZY using a mortar and pestle until a very fine composite anode powder was formed. All the slurries were prepared by suspending the powder materials in a polymer binder mixture and ethanol [11,12].

2.2 Button Cell Fabrication

The fabrication of the button cell with a NiO-BCZY | AFL | BCZY | LSCF configuration was performed as follows. Anode substrates were prepared by die-pressing NiO-BCZY powder (50:50 ratio) into pellets using a 13 mm stainless steel die. The anode functional layer (AFL), composed of NiO:BCZY at a 10:90 ratio, was applied using spin coating in three configurations: three (3) layers for Cell A, six (6) layers for Cell B, and nine (9) layers for Cell C. The electrolyte layer of BCZY was then spin-coated on the sintered anode and sintered at 1450 °C for 5 hours in ambient air. Finally, the cathode slurry was spin-coated onto the electrolyte surface and sintered at 950 °C to form the complete button cell.

2.3 Button Cell Characterization

The microstructure of the fabricated button cells was observed using a scanning electron microscope (SEM) (Phenom XL Benchtop). Comparisons were made with cells fabricated without the AFL. Electrochemical performance was evaluated using a ZIVE SP2 Electrochemical Workstation (ZIVE LAB, WonATech) at operating temperatures ranging from 600 °C to 800 °C in a fuel-cell environment. Impedance spectra were acquired over a frequency range from 1 MHz to 100 mHz and analyzed using ZMAN software. Distribution of relaxation times (DRT) analysis was performed using DRTtools to identify individual electrochemical processes within the cell.

3. Results and Discussion

3.1 Morphology and Elemental Composition Analysis

Figure 1 shows the cross-sectional SEM images and EDX elemental mapping of the button cells with 3, 6, and 9 layers of anode functional layer (AFL). An example of elemental mapping is shown in the inset of Figure 1(a), representing the 3-layer configuration. The cells display well-defined interfaces between the anode, electrolyte, and cathode layers, indicating successful fabrication. This separation is critical for avoiding undesired elemental diffusion, which may negatively impact electrochemical performance. Further analysis using field emission scanning electron microscopy (FESEM) is recommended to observe in more detail the effect of AFL thickness on microstructure.

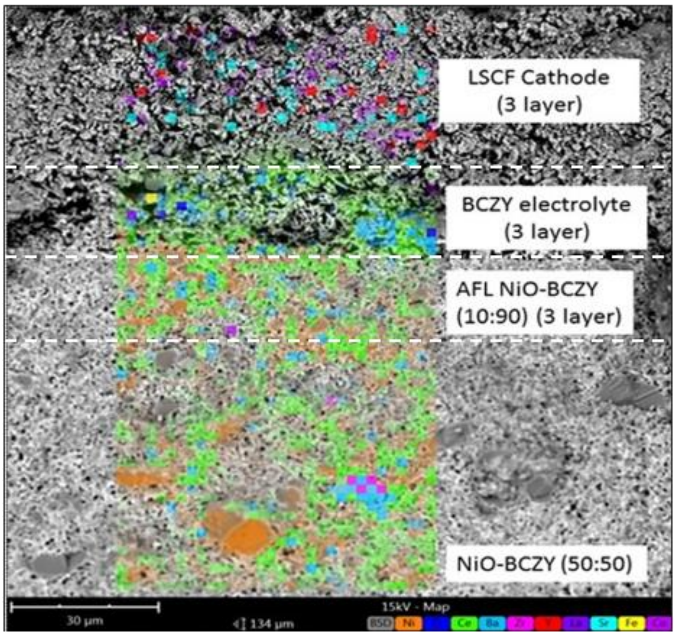

Fig. 1. Cross-sectional SEM and EDX mapping of button cell with three AFL layers

3.2 EIS and DRT Analysis

Figures 2 and 3 display the electrochemical impedance spectra (EIS) and the corresponding distribution of relaxation times (DRT) for cells with 3, 6, and 9 AFL layers measured at 800 °C. The EIS data were analyzed using the Complex Nonlinear Least Squares (CNLS) method and fitted using an equivalent circuit model consisting of Ls–Rs–R1|Q1–R2|Q2–R3|Q. This model yielded three distinct semicircular arcs corresponding to different electrochemical processes.

In contrast, the DRT analysis revealed up to six peaks, indicating additional electrochemical contributions not clearly distinguishable in the conventional Nyquist plots. The peaks can be correlated to charge transfer resistance, gas diffusion, interfacial resistance, and other internal processes. The enhanced resolution of DRT analysis demonstrates its advantage in identifying complex mechanisms and suggests the need for refining the equivalent circuit model to include more elements.

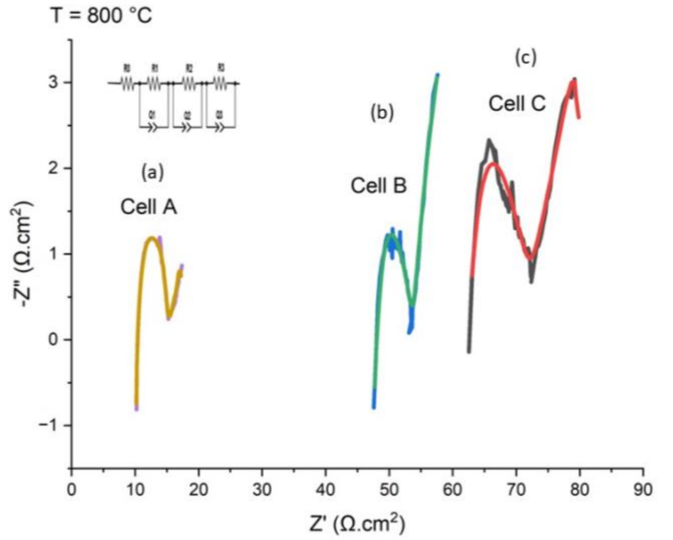

Fig. 2. Impedance spectra of button cells with (a) 3 AFL layers (Cell A), (b) 6 AFL layers (Cell B), and (c) 9 AFL layers (Cell C) at 800 °C

3.3 Resistance Analysis

Table 1 presents the ohmic resistance (Rs), individual polarization components (R1, R2, R3), total polarization resistance (Rpt = R1 + R2 + R3), and total resistance (Rt = Rs + Rpt) at 800 °C. Among all cells, the one with 3 AFL layers (Cell A) exhibited the lowest total resistance of 19.3 Ω·cm², while Cell B (6 layers) and Cell C (9 layers) showed significantly higher resistances of 58.8 Ω·cm² and 79.6 Ω·cm², respectively.

This result is in agreement with previous findings [7,8], where fewer AFL layers were associated with better electrochemical performance. This is likely due to reduced proton diffusion length and more efficient charge transport pathways in thinner AFLs. In thicker AFLs, the extended diffusion path and potential formation of defects or pore disruption can increase interfacial and bulk resistance.

Table 1. Electrical characteristics of button cells with different AFL layer numbers at 800 °C

Layers	Rs (Ω·cm²)	R1 (Ω·cm²)	R2 (Ω·cm²)	R3 (Ω·cm²)	Rpt = R1 + R2 + R3 (Ω·cm²)	Rt = Rs + Rpt (Ω·cm²)
3 (Cell A)	10.1	3.4	1.7	4.1	9.2	19.3
6 (Cell B)	47.6	5.1	2.3	3.8	11.2	58.8
9 (Cell C)	62.5	7.9	2.1	7.1	17.1	79.6

3.4 Performance Implications

Figure 4 illustrates the effect of the number of AFL layers on polarization resistance. A clear trend is observed: polarization resistance increases with the number of layers. Cell A (3 layers) recorded the lowest Rpt (9.2 Ω·cm²), whereas Cell C (9 layers) exhibited the highest (17.1 Ω·cm²). The observed performance degradation in thicker AFL configurations (Cells B and C) may be attributed to limited gas transport and reduced effective triple-phase boundary (TPB) density.

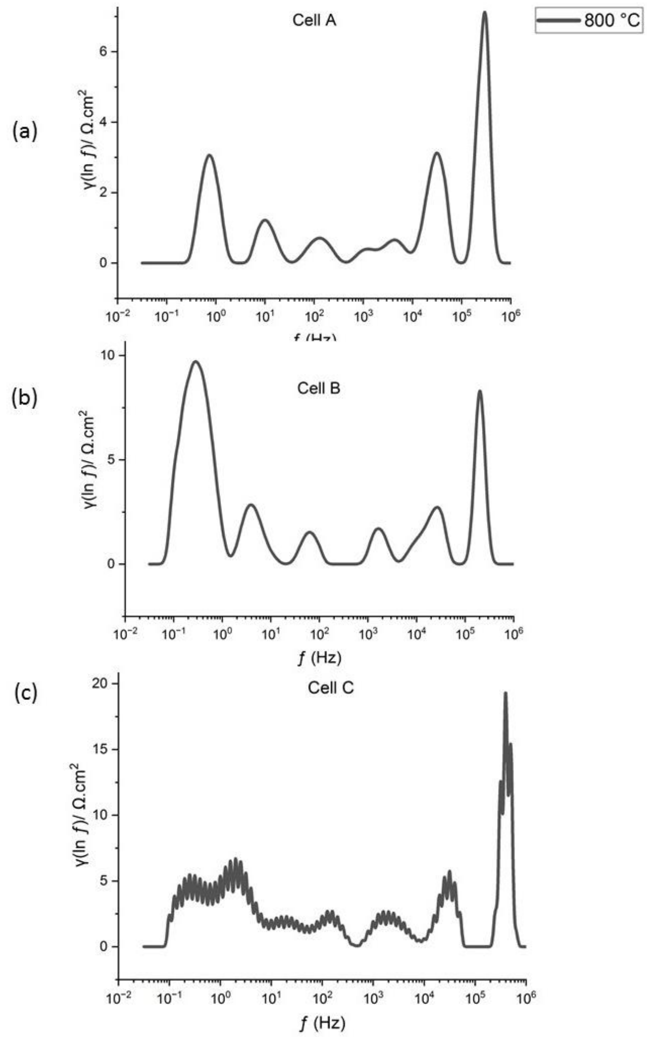

Fig. 3. DRT graphs of button cells with (a) 3 AFL layers (Cell A), (b) 6 AFL layers (Cell B), and (c) 9 AFL layers (Cell C) at 800 °C.

These findings emphasize that although additional AFL layers can enhance mechanical integrity, they may introduce excessive resistance due to longer hydrogen ion diffusion paths and interfacial mismatches. Optimization of AFL thickness is therefore crucial for balancing mechanical robustness and electrochemical efficiency.

4. CONCLUSION

The button cells with configurations of NiO-BCZY | AFL (NiO:BCZY) | BCZY | LSCF were successfully fabricated with 3, 6, and 9 layers of anode functional layers (AFL) and systematically characterized. Electrochemical impedance spectroscopy (EIS) results revealed that the button cell with 3 AFL layers (Cell A) demonstrated superior electrochemical performance, with the lowest total resistance and polarization resistance among all tested cells.

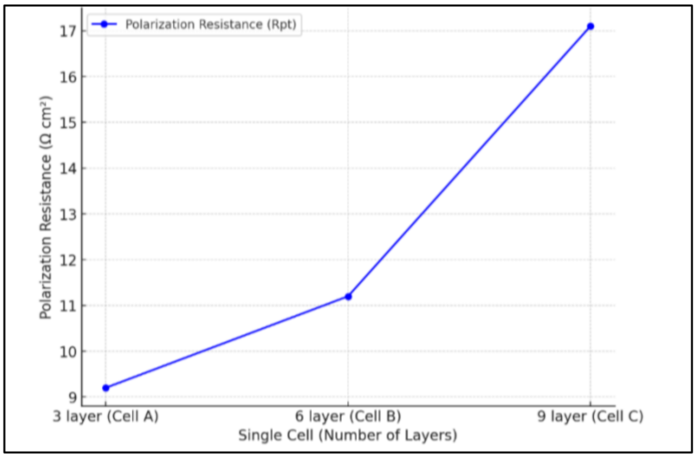

Fig. 4. Effect of number of AFL layers on polarization resistance of button cells at 800 °C

The reduced polarization resistance in the 3-layer configuration suggests improved charge transfer processes and effective triple-phase boundary (TPB) formation. In contrast, cells with more AFL layers showed increased resistance, likely due to longer diffusion paths and higher interfacial resistance. These findings indicate that the number of AFL layers has a significant impact on the electrochemical performance of proton ceramic fuel cells (PCFCs).

In summary, optimizing the AFL design is essential, and this study concludes that three AFL layers offer the most favorable performance at 800 °C. Future work should focus on the structural tuning of AFL layers and exploration of advanced materials and fabrication techniques to further enhance PCFC efficiency.

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