Performance Variation with SDC Buffer Layer Thickness

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ABSTRACT

The performance of anode-supported solid oxide fuel cells was investigated as the SDC buffer layer thickness was varied between $\sim 0.4 \mu\text{m}$ and $\sim 2.3 \mu\text{m}$. The thickness of the buffer layer has a significant effect with the peak performance varying in magnitude by a factor of almost three. A peak power density of 1106 mW.cm^{-2} was achieved at 800°C and an optimal SDC buffer layer thickness of $\sim 1.5 \mu\text{m}$. The performance variation was complex due to a balance between ohmic and polarization losses, triple phase boundary area, pin holes and interfacial reactions between the BSCF + SDC cathode, SDC buffer layer, and YSZ electrolyte. Understanding this variation is essential in order to compare two fuel cells having a different porous buffer layer thickness.

Keywords: Intermediate temperature solid oxide fuel cell; Buffer layer; Interlayer; Anode supported solid oxide fuel cell

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1. INTRODUCTION

Solid oxide fuel cells (SOFCs) are promising electrochemical energy conversion devices that boast high efficiency and fuel flexibility [1-3]. SOFCs often use a yttria-stabilized zirconia (YSZ) electrolyte due to its mechanical strength and performance at high temperatures ($\sim 1000^{\circ}\text{C}$). Despite the advantages while operating at high temperatures, there are additional challenges including reduced lifetime, increased maintenance, material degradation and seal failures [4,5]. Recent efforts have focused on reducing the operating temperature to intermediate temperatures from $600\text{--}800^{\circ}\text{C}$. Two different techniques have been used extensively to improve the performance at intermediate temperatures; 1) reduce the electrolyte thickness and/or 2) use perovskite cathode materials with higher ionic and electronic conductivity at intermediate temperatures. The use of the anode-supported SOFC (AS-SOFC) architecture has allowed for thin film YSZ electrolytes with enhanced performance due to a reduction in the ohmic losses of the fuel cell [6-10]. Further reductions in the fuel cell resistance are possible with perovskite materials like $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ (BSCF) [11,12]. This cathode material is known for higher oxygen permeability and electro-catalytic activity compared to the Sr-doped LaMnO_3 (LSM) cathode used with YSZ electrolytes, especially at intermediate temperatures [13,14].

Despite the potential for enhanced performance by reducing the electrolyte thickness and using a BSCF cathode, poorly conductive layers of BaZrO_3 and SrZrO_3 form in between BSCF and YSZ at the temperatures required for fabrication [4,15-18]. For example, BaZrO_3 is detected after co-sintering YSZ and BSCF powders at 900°C or higher [15]. SrZrO_3 is detected

after sintering BSCF on YSZ substrates at 1000°C [4], but has also been detected after sintering at 900°C or higher in other cathode materials containing Sr [19]. The chemical incompatibility between these materials resulted in a call for an interlayer or buffer layer of doped-ceria material [16,20-26] such as Sm-doped CeO₂ (SDC) that is more compatible with BSCF and can eliminate the interfacial reactions and reduce the interfacial resistance [27,28]. Unfortunately, Zr – Ce interdiffusion also results in interfacial reactions between YSZ and the SDC buffer layer [28] and occurs while co-sintered YSZ-SDC substrates in air at 1100°C or higher [16]. The formation of the resistive (Ce,Sm)₂Zr₂O₇ phase is highly dependent on the sintering temperature [15,16,29,30]. As a result, low sintering temperatures are often employed, which do not allow for full densification of the SDC buffer layer and result in diffusion of Zr into SDC [31]. In order to prevent the YSZ-SDC reaction, advanced techniques such as pulsed laser deposition and chemical solution deposition have been employed that create dense SDC layers and help prevent Zr diffusion into the SDC [4,31,32]. This has led to peak power densities of ~2.0 W.cm⁻² for the fully dense SDC buffer layer with BSCF cathode [4]. Despite the potential with these advanced techniques, techniques such as wet powder spraying and screen-printing are cost effective and scalable to industry [5,9,11,13,33,34], regardless of the porous SDC layer. Furthermore, the porous, rough SDC layer can be advantageous in increasing the effective surface area of the triple phase boundary and enhancing the fuel cell performance [6,35]. These reasons are considered partially responsible for the peak performance (~1.9 W.cm⁻²) of a porous SDC buffer layer occurring at a sintering temperature of 1350°C despite the detrimental Zr – Ce interdiffusion that occurs [36].

Despite comparisons between different fabrication techniques [4], effective means of comparing porous buffer layers with dense buffer layers is significantly hindered by the wide range of buffer layer thicknesses used. For example, the buffer layer thickness often ranges from 150 nm to 6 μm with most studies being conducted with only a single thickness of the SDC buffer layer [4,6,11,37,38]. Recently, Constantin *et. al.* demonstrated the effects of varying dense buffer layer thickness [39]. In the case of the porous SDC buffer layers, there are many competing factors including pinholes in the buffer layer, interfacial reactions and improved triple phase boundary. To our knowledge, no studies have investigated the tradeoffs between the thickness of the porous SDC buffer layer and the fuel cell performance, resistance, and interfacial reactions.

In the present study, wet powder spraying is employed to create a porous SDC buffer layer. The thickness of the buffer layer is varied between 0.4 μm and 2.3 μm and the effect on performance, resistance, and interfacial reactions is investigated.

2. EXPERIMENTAL

2.1 Fuel cell fabrication

The anode and electrolyte of the SOFCs were prepared by tape casting NiO + YSZ (60:40 w/w, NexTech materials) anode and the YSZ (NexTech materials) electrolyte. The green body was then laminated and co-sintered [40] at 1200°C for 2 hrs. After sintering, an SDC buffer layer was deposited by wet powder spraying. Six different samples were prepared by spraying 20, 40, 60, 80, 100 and 120 layers of SDC onto the pre-sintered substrates. The substrates were then sintered at 1400°C for 4 hrs for a final anode thickness of ~250 μm and electrolyte

thickness of ~10 μm . The sintered buffer layer thickness was ~0.4 μm , ~0.8 μm , ~1.1 μm , ~1.5 μm , ~1.9 μm and ~2.3 μm for the 20, 40, 60, 80, 100 and 120 layers sprayed, respectively. The BSCF+SDC (70:30 w/w) cathode was then spray deposited and the cells were sintered at 1000°C for 5 hrs for a final cathode thickness of ~4 μm . The prepared substrates were 15 mm in diameter with a cathode area of 0.7 cm^2 .

2.2 Characterization

The fuel cell performance was measured using the current voltage (IV) method. A digital SourceMeter (Keithley 2420) was interfaced with a computer for measurement and recording of the fuel cells open circuit voltage (OCV), polarization and power density. Electrochemical impedance spectroscopy (EIS) of the fuel cells was conducted with a Solartron 1260 A frequency response analyzer with a Solartron 1287 Potentiostat under OCV conditions. EIS was recorded over a frequency range of 10^6 to 0.1 Hz with a signal amplitude of 10 mV. The fuel cells cross sectional morphology was assessed with a scanning electron microscope (SEM) and energy dispersive X-ray spectroscopy (EDX). The SEM and EDX analysis was performed at 8 kV and a working distance of 15 mm. The magnification for the EDX analysis was set to 85,000x and the probe size estimated for these conditions is 1 cubic micron.

2.3 Testing setup

Testing using the IV method and EIS occurred in a tubular furnace, which was heated, to 800°C at 5 °C per minute. The fuel cells were tested with hydrogen at a flow rate of 100 mL min^{-1} . The flow of hydrogen was regulated with a mass flow controller and LabView software. Air was used as the oxidant and was supplied by natural convection in the furnace. The

prepared fuel cell was sealed on a quartz tube with silver paste. Silver paste was applied to the cathode and silver wire was used as the current collector for both the anode and cathode. The fuel cell temperature was monitored with a K-type thermocouple placed on the outside of the quartz tube.

3. RESULTS AND DISCUSSION

3.1 Fuel cell performance

Each of the fuel cells tested at 800°C achieved significant performance despite the reaction between the YSZ and SDC layers which is known to occur at 1400°C [37]. However, a definite trend was observed in fuel cell performance and polarization as the thickness of the SDC layer was varied. **Fig. 1** shows a comparison of the fuel cell performance for the 20, 40, 60, 80, 100 and 120 layers of SDC sprayed. Initially, the fuel cell performance improved with increased buffer layer thickness. The peak power density for the 20, 40, 60 and 80 sprayed layers was 398, 478, 948 and 1106 mW.cm⁻², respectively. With further increase of the buffer layer thickness, the performance declined. With 100 and 120 layers of SDC sprayed the fuel cell only achieved 884 and 845 mW.cm⁻², respectively. The trend is surprising because the preparation for the cells was identical except for the variation in the number of SDC layers sprayed and the resulting buffer layer thickness. In fact, increasing the thickness of an electrolyte layer is known to cause increased ohmic resistance and reduced fuel cell performance. The exact opposite occurs for the fuel cell initially, followed by a decrease in performance beyond a certain thickness of the SDC buffer layer. The variation in performance

for a buffer layer of $\sim 0.4 \mu\text{m}$ (20 layers) to a buffer layer of $\sim 1.5 \mu\text{m}$ (80 layers) differs by nearly a factor of three, demonstrating the importance of controlling the buffer layer thickness.

Figure 1: Fuel cell performance at 800°C with 20, 40, 60, 80, 100, and 120 layers of SDC sprayed onto the substrate.

To further understand this unusual behavior, EIS was conducted on the fuel cell. **Fig. 2** shows the results of the EIS with the ohmic resistance, polarization resistance and total resistance variations plotted against the number of SDC layers sprayed. As expected, the ohmic resistance of the fuel cell increases with SDC thickness as long as 40 layers of SDC or more were sprayed on the substrate. This trend is expected because increasing the SDC thickness results in a longer path for ionic conduction of oxygen ions. However, the ohmic resistance of

the fuel cell with 20 layers of SDC sprayed is much higher than the ohmic resistance of any other fuel cell except the 120 layers of SDC. The ohmic resistance was 0.361, 0.335, 0.349, 0.355, 0.356, and 0.364 $\Omega\cdot\text{cm}^2$ for the 20, 40, 60, 80, 100 and 120 layers of SDC, respectively. Despite the variations in ohmic resistance, the total resistance was effected only slightly because the variations in ohmic resistance are small compared to the total resistance. The total resistance follows the polarization resistance much more closely, beginning at 1.28 $\Omega\cdot\text{cm}^2$ for 20 layers of SDC sprayed and decreasing steadily with increasing layers of SDC sprayed to 0.819 $\Omega\cdot\text{cm}^2$ at 80 layers. A decrease in polarization losses appears to result from an improved triple phase boundary. The SDC buffer layer does not densify fully after sintering at 1400°C as shown in Fig. 3, resulting in an average pore size of 0.16 μm . This pore size is comparable to the thickness of the 20 and 40 layers sprayed, which resulted in an SDC buffer layer thickness of ~0.4 and ~0.8 μm , respectively. Due to the porosity of the SDC buffer layer, which is pre-sintered at 1400°C before applying the SDC + BSCF cathode, the triple phase boundary can improve with the SDC + BSCF cathode infiltrating into the SDC buffer layer. This infiltration is primarily a result of applying the SDC + BSCF cathode with the wet powder spraying technique, which results in a wet application of the cathode that can flow into the pores prior to drying fully. For the 20 and 40 layers, the cathode can potentially flow through most of the buffer layer because the buffer layer is thin relative to the average pore size. This enhanced triple phase boundary can result in decreased polarization losses, which are observed between 20 and 80 layers of SDC and should affect the 100 and 120 layers of SDC as well. However, with an average pore size of 0.16 μm the cathode is not expected to infiltrate through

the entire buffer layer when the thickness increases to $\sim 1.9 \mu\text{m}$ and $\sim 2.3 \mu\text{m}$ for the 100 and 120 layers of SDC, respectively. As a result, the triple phase boundary can only be enhanced to a certain extent. The trend of decreasing polarization losses reverses as the polarization losses began to increase for the 100 layers of SDC up to $1.37 \Omega\cdot\text{cm}^2$ for the 120 layers of SDC. The cause of the peak performance at 80 layers of SDC is clearly related to both ohmic and polarization losses and is dependent upon the cathode-buffer layer and buffer layer-electrolyte interfaces of the fuel cell. Variations in the anode-electrolyte interface did not contribute because the preparation was identical for these layers.

Figure 2: Ohmic resistance, polarization resistance and total resistance as the number of sprayed SDC layers is varied from 20 to 120 layers.

Figure 3: SEM morphologies of the SDC buffer layer surface sintered at 1400°C.

3.2 SEM and EDX analysis

To gain a more comprehensive understanding of the performance variation with SDC thickness, the microstructure of the fuel cells was assessed with SEM. **Fig. 4** is a comparison of the fuel cell cross sectional morphologies as the SDC buffer layer thickness is varied. With only 20 layers sprayed of SDC, the buffer layer is extremely thin between the cathode and electrolyte and is porous as shown in **Fig. 3**. The thin, porous buffer layer at 20 layers is likely to result in a reaction between BSCF and YSZ. EDX analysis of the YSZ-SDC interface is shown in **Fig. 5**. Indeed, both Ba and Sr were detected at the YSZ-SDC interface. The detrimental formation of BaZrO_3 and SrZrO_3 is not prevented with only 20 layers of SDC and the buffer layer was ineffective due to pin holes. The presence of Ba and Sr at the YSZ-SDC interface explains the initially high ohmic resistance of the fuel cell with 20 layers of SDC sprayed. As described in the introduction, the formation of BaZrO_3 and SrZrO_3 is known to create high interfacial resistance and poor performance. **Fig. 6** is the EDX analysis of the YSZ-SDC interface of the fuel cell with 40 layers of SDC sprayed. Also shown is the EDX

analysis within the SDC buffer layer. As shown, no Ba or Sr were detected in the fuel cell at the interface. The buffer layer was thick enough to prevent direct contact between the electrolyte and the cathode. The ohmic resistance of the fuel cell reduced to its minimum at 40 layers of SDC and then steadily increases with thickness of the SDC layer, as expected. Unfortunately, the porous buffer layer with 40 layers sprayed is not effective at preventing Zr diffusion into the SDC layer. **Fig. 6** indicates that significant Zr is detected within the SDC buffer layer where Ba and Sr are also present. Reactions can still occur to form BaZrO_3 and SrZrO_3 within the buffer layer, but the reactions were prevented at the YSZ interface directly, which reduced the ohmic resistance of the fuel cell for this condition.

Figure 4: Cross sectional morphologies of the fuel cell at a) 20 b) 40 c) 60 d) 80 e) 100 and f) 120 layers of SDC sprayed.

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Figure 5: EDX analysis at the YSZ-SDC interface with 20 layers of SDC sprayed.

Figure 6: EDX analysis of the YSZ-SDC interface and within the SDC layer with 40 layers of SDC sprayed.

Fig. 4 shows the growth of the SDC layer with the number of layers of SDC sprayed. However, Fig. 4e and 4f show an increasingly defined layer between the YSZ and SDC buffer layer. This layer appears as an intermediate gray color in comparison to the lighter YSZ layer below and the darker SDC layer above. This layer, although present in the earlier samples, is becoming thicker. **Fig. 7** is the EDX analysis of the interface between this grey area and the darker SDC layer above for 80 layers of SDC sprayed. As shown, no Ba or Sr were detected at this interface. However, Zr, Y, Ce, Sm and O were detected. This grey area is a layer of interfacial reaction between the YSZ-SDC layers. As discussed in the introduction, YSZ and SDC can also react at

high sintering temperatures to form the resistive $(\text{Ce,Sm})_2\text{Zr}_2\text{O}_7$ phase. This resistive phase is growing as shown by the SEM. As this layer grows, the increasing interfacial resistance contributes to the polarization and ohmic resistance of the fuel cell. **Fig. 8** shows a similar analysis of the 120 layers of SDC. For this cell, no Ba or Sr were initially detected at the YSZ-SDC reaction interface. However, the resistive $(\text{Ce,Sm})_2\text{Zr}_2\text{O}_7$ phase cannot be prevented with Zr, Ce, Sm and O all present. Fortunately, this resistive phase is not as detrimental to the fuel cells performance as BaZrO_3 and SrZrO_3 [39], which causes the performance to initially improve with thickness until the $(\text{Ce,Sm})_2\text{Zr}_2\text{O}_7$ phase becomes too thick and the performance degrades. The increasing region of YSZ-SDC interfacial reactions appears to alter the morphology and influence the activation and mass transport losses of the fuel cell by decreasing O_2 and O^{2-} transfer to the YSZ layer. This results in an increase in polarization losses and a decrease in performance with a thicker SDC buffer layer.

Figure 7: EDX analysis of the YSZ-SDC reaction interface and SDC-BSCF interface with 80 layers of SDC sprayed.

Figure 8: EDX analysis of the YSZ-SDC reaction interface and SDC-BSCF interface with 120 layers of SDC sprayed.

The EDX analysis shown in **Fig. 7** and **Fig. 8** also show that Zr is present at the SDC-BSCF interface. There is not as much Zr present at this interface as the YSZ-SDC reaction interface, but the formation of both BaZrO_3 and SrZrO_3 is still possible in the region near the SDC-BSCF interface. The overall effect of these interfacial reactions is an increase in the ohmic and polarization resistance, which contribute to a gradual decline in fuel cell performance. These reactions cause the decrease in fuel cell performance as the SDC buffer layer becomes thicker.

4. CONCLUSION

The thickness of the SDC buffer layer has a significant effect on the overall fuel cell performance. An optimal porous SDC buffer layer thickness of $\sim 1.5 \mu\text{m}$ (80 layers of SDC sprayed) was uncovered with a peak power density of 1106 mW.cm^{-2} at 800°C . The peak power density was nearly 3 times higher than the peak power density of the 20 layers sprayed. For layers thinner than $1.5 \mu\text{m}$, the reaction between BSCF and YSZ as well as the decreased triple phase boundary contribute to reduced performance. For layers thicker than $1.5 \mu\text{m}$, the increased ohmic resistance due to the buffer layer thickness and the reactions between YSZ and SDC and between BSCF and YSZ contribute to decreased performance. An optimal buffer layer thickness results, which balances these contributions.

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FIGURE CAPTIONS

Figure 1: Fuel cell performance at 800°C with 20, 40, 60, 80, 100, and 120 layers of SDC sprayed onto the substrate. (1 column image)

Figure 2: Ohmic resistance, polarization resistance and total resistance as the number of sprayed SDC layers is varied from 20 to 120 layers. (1 column image)

Figure 3: SEM morphologies of the SDC buffer layer surface sintered at 1400°C. (1 column image)

Figure 4: Cross sectional morphologies of the fuel cell at a) 20 b) 40 c) 60 d) 80 e) 100 and f) 120 layers of SDC sprayed. (1.5 or 2 column image)

Figure 5: EDX analysis at the YSZ-SDC interface with 20 layers of SDC sprayed. (1 column image)

Figure 6: EDX analysis of the YSZ-SDC interface and within the SDC layer with 40 layers of SDC sprayed. (1 column image)

Figure 7: EDX analysis of the YSZ-SDC reaction interface and SDC-BSCF interface with 80 layers of SDC sprayed. (1 column image)

Figure 8: EDX analysis of the YSZ-SDC reaction interface and SDC-BSCF interface with 120 layers of SDC sprayed. (1 column image)