Single-step fabrication of an anode supported planar single-chamber solid oxide fuel cell

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Single-step fabrication of an anode supported planar single-chamber solid oxide fuel cell

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Abstract
We present single-step-co-sintering manufacture of a planar single-chamber solid oxide fuel cell (SC-SOFC) with porous multilayer structures consisting of NiO/CGO, CGO and CGO-LSCF as anode, electrolyte, and cathode, respectively. Their green tapes were casted with 20 μm thickness and stacked into layers of anode, electrolyte, and cathode (10:2:2), then hot-pressed at 2 MPa and 60°C for 5 minutes (deemed optimal). Subsequently, hot laminated layers were cut into 40 × 40 mm cells and co-sintered up to 1200°C via different sintering profiles. Shrinkage behavior and curvature developments of cells were characterized, determining the best sintering profile. Hence, anode-supported SC-SOFCs were fabricated via a single-step co-sintering process, albeit with curvature formation at edges. Subsequently, anode thickness was increased to 800 μm and electrolyte reduced to 20 μm to obtain SOFCs with drastically reduced curvature with the help of a porous alumina cover plate.

KEYWORDS
cell manufacturing, co-sintering, hot lamination, single-chamber, solid oxide fuel cell

1 | INTRODUCTION

Single-chamber solid oxide fuel cells (SC-SOFCs), a new configuration of SOFC, were introduced by researchers to eliminate certain problems faced in dual chamber SOFCs (DC-SOFCs), such as gas-tight sealing, complex gas manifolding, and flow field structures. Unlike conventional SOFCs, SC-SOFCs consist of only one gas compartment containing a gas mixture of fuel and oxygen. This characteristic property of SC-SOFCs enables them to have simplified structures, resulting in better thermal shock resistance and less start-up and shut down time than DC-SOFCs. If their potential is realized, the simplified structure could yield a significant reduction in the total system cost. Furthermore, the electrolyte layer does not have to be dense to stop gas crossover. However, the main issue for single-chamber structures is the low power output deriving from low fuel utilization due to a lack of catalytic selectivity of the electrode materials in a mixed gas condition. One of the other major reasons is the given restriction on fuel mixing ratios, at either very rich or very lean conditions, in order to avoid auto-ignition of the supplied fuel.

A co-sintering process of multi-layer structures yields significant benefits in time, effort, and energy savings, and so has been attracting increasing attention in a variety of research fields, including SOFCs. In the fabrication of conventional SOFCs, co-sintering has been usually applied to bi-layer structures, consisting of anode and electrolyte layer, as the similar sintering behavior of each layer prevents developments of any defects such as warping, cracking, and delamination during the process.

At present, anode-supported SC-SOFCs are fabricated using at least two sintering steps: co-sintering anode and electrolyte, followed by a separate deposition and sintering...
of the cathode.\textsuperscript{9,11} The rationale for a two-step sintering is that the materials for anode, electrolyte, and cathode require different sintering temperatures to achieve the expected microstructures. Though a two-step sintering process decreases possible reactions/interactions between electrolyte and cathode,\textsuperscript{11} there are several reasons why single-step co-sintering of SC-SOFCs are desirable, mainly pertaining to simplifying the process along with decreasing the processing time and input energy.\textsuperscript{12,13} These benefits of single-step co-sintering of SC-SOFCs reduce further the cost of SC-SOFCs fabrication and thereby improve commercial viability if employed in commercial scale.

In this study, a single-step co-sintering process is applied to fabricate anode-supported planar SC-SOFCs. Sintering the anode, cathode, and electrolyte in one step is difficult because each component has different sintering behaviors and microstructures.\textsuperscript{12,14-16} Therefore, each component of the cell must match in shrinkage behavior to avoid or minimize strain mismatches. Otherwise, it is highly possible to form crack or crack-like defects in the cell as a result of the mismatch stress. Furthermore, thermal expansion coefficient (TEC) mismatch between each layer becomes a dominant factor when materials have higher viscosity that is, during cooling process.\textsuperscript{6} Therefore, in order to achieve the single-step co-sintering of an anode supported SC-SOFC, the sintering temperature was decreased to obtain a preferable cathode microstructure, while still densifying the electrolyte.\textsuperscript{11,17,18} In addition, the sintering temperature of the cathode is generally less than that of the anode and electrolyte.\textsuperscript{11,19,20} Therefore, the cathode particle size was chosen to be greater than that of the anode and cathode, so as to retard the sintering kinetics of the cathode and thereby suppressing quicker densification, as well as to balance the final density of materials during the co-sintering process.\textsuperscript{5} Lowering sintering temperature is also helpful in reducing the TEC mismatch between each component and thereby mitigating possible crack or delamination in the cell during sintering.\textsuperscript{6} Furthermore, possible reactions between electrolyte and electrodes are reduced owing to decreased sintering temperature.\textsuperscript{14,21} However, lowering the sintering temperature of the electrolyte results in the electrolyte not being fully densified, which is unacceptable for DC-SOFCs since the fuel and oxidant have to be separated from each other. However, one of the benefits of using SOFC in a single-chamber condition is to tolerate a porous electrolyte structure.\textsuperscript{1,3} Therefore, the porous structure of electrolyte at low sintering temperature can be ignored, though the cell performance is decreased due to lower open circuit voltage (OCV) and higher electrolyte resistance.\textsuperscript{14,22,23} In addition, heating rate and cooling rate were also carefully defined to prevent any cracking, delamination and curvature formation due to the different shrinkage rate of anode cathode and electrolyte and thermal expansion co-efficient (TEC).\textsuperscript{6,24,25} For this purpose, an anode supported SC-SOFC, made of Ni-CGO anode, CGO electrolyte and LSCF-CGO cathode, were made and co-sintered at 1200°C.

The experimental results show that the anode supported planar cell for single-chamber working condition can be manufactured via single-step co-sintering, but have issues that must be tackled, such as sensitivity to mechanical stress (which must be reduced), as well as curvature formations at the cell edges. These problems must be overcome in order to manufacture cells by this fabrication method. Therefore, the thickness of anode was increased to increase the cell strength as well as to achieve a reduction in curvature evolution at the cell edges. Moreover, an alumina cover plate was used to help aid the removal of homogeneous additives and suppress curvature evolution. As a result, a curvature-free planar SC-SOFC was obtained via this fabrication method.

## EXPERIMENTAL PROCEDURE

### 2.1 Cell preparation

#### 2.1.1 Materials and slurry composition

The anode, cathode, and electrolyte green layers were purchased, (Maryland tape-casting\textsuperscript{26} made via tape-casting method, and were utilized to make the required thicknesses and corresponding thickness ratio of the electrodes and electrolyte for the anode-supported planar SC-SOFCs. The basic composition of the green tapes is summarized in Table 1 (for full slurry constituents see Table S1, appendix). The particle size of main constituents in the anode and electrolyte tapes is 0.3 μm, and in the cathode 1 μm, designed to retard its sintering.

#### 2.1.2 Green body preparation of cells

Multiple layers of green tapes were stacked together to reach the desired thickness of electrodes and electrolyte, and the thickness for each component is controlled by the number of layers, estimated based on the thickness of 20 μm for each tape. To make anode supported SC-

| TABLE 1 Green tape slurry composition and particle size of main constituent |
|-----------------|----------------|----------------|
| **Green tape layer** | **Composition** | **Particle size (μm)** |
| Anode | 60 wt%NiO; 40 wt%Ce\(_{0.8}\)Gd\(_{0.2}\)O\(_2\)\(_\gamma\) (CGO) (20%Gd) | 0.3 |
| Electrolyte | CGO Ce\(_{0.8}\)Gd\(_{0.2}\)O\(_2\)\(_\gamma\) (20%Gd) | 0.3 |
| Cathode | 50 wt%La\(_{0.6}\)Sr\(_{0.4}\)Co\(_{0.2}\)Fe\(_{0.8}\)O\(_3\) (LSCF); 50wt% CGO(20%Gd) | 1 |
SOFCS, the required number of anode green tapes were first layered upon each other to reach the desired thickness; secondly, the required number of layers of electrolyte tapes were stacked on the anode surface; thirdly, the required number of layers of cathode tapes were placed on the surface of electrolyte. The multi-layered stack was placed between two protective sheets (silicon coated PET release film) and pressed/laminated under different pressures and temperatures under a hot lamination machine (Carvel Heated Bench Top Hot Press, model: 3853CE-8).

It is important to determine suitable hot lamination conditions so as to obtain acceptable compactness, good adhesion between each layer, prevent over-pressing and avoid inhomogeneous cell area distribution. For example, high pressure and temperature of hot lamination might lead to (i) cell crushing or materials over penetrating into each other or (ii) give rise to the cell face sticking to the protective sheet and hence making it difficult to extract without damage; whereas overly low pressure and temperature results in delamination due to poor adhesion. Similarly, inhomogeneous cell area due to uneven pressing causes different shrinkage and thereby leads to stress in the cell during sintering.8

### 2.1.3 Hot lamination

To find optimum hot lamination conditions, nine examples of anode-supported cell were prepared at pressures ranging from 1-3 MPa and temperatures ranging from 50-70°C with a dwelling time of 5 minutes. All cells are 50 × 50 mm square with a thickness of 280 μm, including 200 μm (10 layers) for anode (A), 40 μm for electrolyte (E) and 40 μm for cathode (C), which give a thickness ratio of A to E to C as 10:2:2. All as-pressed green cells were placed on a dense alumina substrate and sintered at 1200°C for 1 hour with a heating rate of 0.5°C min⁻¹ from room temperature to 500°C, 2°C min⁻¹ from 500°C to 900°C, 1°C min⁻¹ from 900°C to 1200°C, and cooling rate of 2°C min⁻¹ from 1200°C to room temperature. Through visual assessment of the changes in shape, in conjunction with observation of dimensional changes, the optimum hot lamination conditions were determined.

### 2.1.4 Sintering

Sintering profile of cells in terms of the heating rate, dwelling and cooling rate, is one of the important aspects that should be taken into consideration so as to obtain cells with less defects by the single-step co-sintering fabrication method. In order to define the sintering profile, the debinding and shrinkage properties of each green layer should be known. This is so because, during the debinding, capillary force is created and this force leads to small shrinkage. If the solvents, binders or other additives in the green tapes are not removed in a controlled manner, that is, if the evaporation rate of these additives is too fast, the capillary force will be too high and cause stress in each layer and even result in some defects in the cells. These defects and stress in the cells at low temperature then cause cracking, delamination or curvature formation during the latest stage of the sintering.8,16 Similarly, if the main shrinkage areas of each layers are known then one can control these temperature areas in order to control stress occurring between each layer (anode-electrolyte and electrolyte-cathode interfaces) during sintering. In addition, the cooling rate during the cooling cycle is also important because each layer has different thermal expansion coefficient,27,28 which might give rise to cracks during cooling if the cooling rate is too high. Therefore, each component of cells’ thermal gravimetric analysis (TGA) and shrinkage measurement were taken to predict suitable sintering profiles. In addition to these, the pictures of separately sintered anode, cathode, and electrolyte were taken to monitor their figural behavior for predicting better sintering profile.

Thereafter five different sintering profiles were determined. In order to define the best sintering profile among these, five identical anode supported planar specimens, (40 × 40 mm, cut from 50 × 50 mm hot laminated cells) possessing 10:2:2 (AEC) thickness ratio and 200:40:40 μm thicknesses, respectively, were made and each sintered according to one of the five corresponding sintering profiles.

### 2.2 Preparation of SC-SOFC cells with different thicknesses of anode and electrolyte

The thickness of each component in a cell can have direct impact on co-sintering. In order to evaluate this phenomenon, cells were prepared with different thicknesses as detailed in Table 2, and same planar dimensions of 40 × 40 mm. They were hot laminated at 60°C and 2 MPa for 5 minutes, followed by sintering at 1200°C for 1 hour with a heating rate of 1°C min⁻¹ from room temperature to 500°C, 2°C min⁻¹ from 500°C to 900°C, 1°C

<table>
<thead>
<tr>
<th>Cell A</th>
<th>Cell B</th>
<th>Cell C</th>
<th>Cell D</th>
<th>Cell E</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness (μm)</td>
<td>200:40:40</td>
<td>400:40:40</td>
<td>400:20:40</td>
<td>800:20:40</td>
</tr>
</tbody>
</table>
**TABLE 3**  Hot lamination results for 9 anode supported cells, thickness ratio 10:2:2, thickness (200:40:400 μm), at different pressure and temperature

<table>
<thead>
<tr>
<th>Cells</th>
<th>Pressure (MPa)</th>
<th>Temperature (°C)</th>
<th>Time (minute)</th>
<th>After hot pressing (cell 50 x 50 mm)</th>
<th>After sintering Cell (40 x 40 mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cell 1</td>
<td>1</td>
<td>50</td>
<td>5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cell 2</td>
<td>2</td>
<td>50</td>
<td>5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cell 3</td>
<td>3</td>
<td>50</td>
<td>5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cell 4</td>
<td>1</td>
<td>60</td>
<td>5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cell 5</td>
<td>2</td>
<td>60</td>
<td>5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cell 6</td>
<td>3</td>
<td>60</td>
<td>5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cell 7</td>
<td>1</td>
<td>70</td>
<td>5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

(Continues)
min\(^{-1}\) from 900\(^\circ\)C to 1200\(^\circ\)C, and cooling rate of 3\(^\circ\)C min\(^{-1}\) from 1200\(^\circ\)C to room temperature. Cell A, B, C, and D were sintered without any constraint but Cell E was sintered with a porous alumina cover plate of 50 \(\times\) 50 mm planar dimensions, 1 mm thickness, 7.31 g weight and 40% porosity (ESL™ 42520-2) placed on top of it.

### 2.3 Characterizations and tests

Thermal gravimetric analysis (TGA) of as-received tapes of anode, cathode and electrolyte was performed by TA Instruments Q5000IR by heating up to 1000\(^\circ\)C at a heating rate of 5\(^\circ\)C min\(^{-1}\). In situ observations of the shrinkage behaviors of each layer with direction were done using long-distance microscope installed on a furnace (Infinity K-20, Infinity Photo-Optical Company). A small hole (30 mm diameter) was made in the furnace door and covered with an uncoated Sapphire window so as to enable pictures of the sample to be taken at different temperatures. Anode, cathode, and electrolyte layers were made separately with dimensions of 40 \(\times\) 40 mm and a thickness of 0.4 mm, and each of them was placed in the furnace with a reference alumina substrate. These samples were sintered at 1200\(^\circ\)C for one hour with a heating and cooling rate of 5\(^\circ\)C min\(^{-1}\). A photo was taken every 50\(^\circ\)C temperature increment up to 1200\(^\circ\)C. Each photo was analyzed in an image processing program (ImageJ program) with the help of reference alumina substrate to measure its shrinkage.

Scanning electron microscopy (SEM) (Zeiss 1530-VP FEGSEM) and X-ray diffraction (XRD) (Bruker D2 Phaser) were used to characterize microstructure of the cells and examine if any new phases formed during sintering. In addition, Porosity was estimated using the imageJ program.

### 3 RESULTS AND DISCUSSION

#### 3.1 Green bodies, hot lamination, and warpage monitoring after sintering

Table 3 illustrates the photos of green cells after hot lamination under various pressing conditions and their sintering results. Figure 1A shows the expansion on width (\(\Delta W\)), length (\(\Delta L\)), and the two diagonals (\(\Delta D\) and \(\Delta T\)) after hot lamination. In addition, Figure 1B depicts the size of the sintered cell on \(W\), \(L\), \(D\), and \(T\). In order to define if cells are hot laminated homogeneously, the cells’ sizes were measured. To achieve a homogeneous hot lamination, it is imperative that the measured cell dimensions satisfy the following criteria: (i) that \(\Delta W \approx \Delta L\); (ii) that \(\sqrt{\Delta W^2 + \Delta L^2} = \Delta D = \Delta T\). Similarly, it should follow that (iii) \(\Delta W \approx L\) and (iv) \(D \approx T\).

According to Table 3, cells that were hot laminated at 70\(^\circ\)C (Cell 7, 8, and 9) were over-pressed because of pressure and temperature mismatch. In addition, it is observed in Figure 1A that the intercept of these cells’ \(\Delta W\) and \(\Delta L\), and \(\Delta D\) and \(\Delta T\) are not on the line which has a slope of 1, which indicates that their expansion (either on width and length, or across diagonals) are different. This shows that there is inhomogeneous expansion in the cells during hot lamination, thus causing eccentric curvature formation and cracking during sintering. In order to avoid this over-pressing issue, hot lamination temperature was decreased to 50\(^\circ\)C. However, at this temperature, Cell 1, Cell 2, Cell 3 had cracks and curvature formation after sintering despite the inhibition of cell over-pressing (Table 3). This might be as a result of different expansion of cell(s) at different edges during hot lamination. As temperature may not be sufficient enough to allow particle packages in cell green
bodies to flow properly over protective sheet during hot lamination, this could have led to locally variable green body density in the cells. Moreover, Figure 1A shows that the expansions $\Delta W$ and $\Delta L$, and $\Delta D$ and $\Delta T$ of Cell 1, 2, and 3 are different, and support the claim that these cells were not pressed homogeneously. Increasing the hot lamination temperature from 50°C to 60°C had different effects on cells with regard to pressure change. Cell 4 and Cell 5 were pressed homogeneously and possess reasonable sintering results, whereas Cell 6 was over-pressed and resulted in strange curvature evolution. According to Figure 1A, the intercept of the $\Delta W$ and $\Delta L$ of hot laminated cell 5 are just about on the line. It shows that their expansion on width and length directions is almost the same, (0.42 mm and 0.46 mm for $\Delta W$ and $\Delta L$, respectively). Likewise, the intercept of the $\Delta D$ and $\Delta T$ of the same cell are also coincident on the line, thus implying that their expansion is nearly the same (0.628 mm and 0.623 mm for $\Delta D$ and $\Delta T$, respectively). In addition, the root of the sum of the squares of $\Delta W$ and $\Delta L$ of the cell 5 was measured as the same with dimension change $\Delta D$ and $\Delta T$. Furthermore, Cell 4 also had acceptable hot lamination results because

**FIGURE 1** A, dimensional changes in anode-supported planar SC-SOFC after hot lamination on $W$, $L$, $T$, and $D$ directions, and B, the value of $W$, $L$, $T$, and $D$ of the cell after sintering [Color figure can be viewed at wileyonlinelibrary.com]
the intercept of $\Delta W$ and $\Delta L$, and $\Delta D$ and $\Delta T$ of cell 4 was also on the line, Figure 1A, and suggesting that it is also hot laminated homogeneously. Figure 1B also illustrates that Cell 4 and Cell 5 possess almost homogeneous sintering results, although there is curvature at the edges. In order to ensure good adhesion between each layer, it is determined that the sintering condition of Cell 5 was to be used for this study rather than that of Cell 4.

Hot lamination temperature might also have effect on solvents in green tapes, such as leading to a small decrease in the amount of solvent in green tapes by evaporation during hot lamination. This then helps sinterability of the green tapes during firing because solvent evaporation becomes less pronounced during sintering, and thus reduces possible defects in the green tape during burnout or debinding. This is posited as one of the possible reasons why cells hot laminated at 60°C at reasonable applied pressures possess better sintering results in comparison to cells hot laminated at 50°C.

3.2 | Sintering analysis

3.2.1 | Debinding stage

Figure 2A shows the TGA profiles of green tapes of anode, cathode and electrolyte. The profiles are similar in weight loss versus temperature. Up to 400°C, almost all volatile constituents were burnt out. While cathode and anode had same weight loss of ~12.5%, electrolyte had slightly lower loss of 11.4%. At temperature from 400°C up to 800°C, all tapes had almost no weight change. From around 800 to 900°C, the cathode tape had further weight loss, making a total loss of 12.98%, while the electrolyte and anode ones had no more weight loss. These measurements imply that almost all polymer components such as solvent, binder, plasticizer, lubricant, and dispersant are completely burnt out before 400°C. The further loss for the cathode tape is likely due to the volatilization of one or more of the ceramic constituents; this weight loss could also be attributable to oxygen evolution from the cathode required to maintain equilibrium oxygen vacancy concentration, or the combination of the above two mechanisms.

3.2.2 | Sintering stage

Figure 2B,C depict the shrinkage and shrinkage rate of each material, respectively. The main shrinkage occurred at temperature above 900°C. Moreover, the cathode possesses the highest shrinkage and shrinkage rate (13.25%, 0.029%/°C, respectively) at a temperature of 1200°C and the electrolyte has the lowest (4.74%, 0.014%/°C, respectively). Furthermore, the shrinkage behavior of anode and electrolyte are a close match with each other before 1100°C, but shrinkage differences increase between the temperature of 1100°C and 1200°C.

These all are attributed to material properties and their grain size. In addition to these, Figure 3 shows the pictures taken of the anode, cathode, and electrolyte during the sintering at different temperatures, and after sintering. It can be clearly seen that cathode has serious issues during the debinding process when heating rate is defined high (5°C per minute), especially in the temperature range from

![Figure 2](https://example.com/figure2.png)
200°C to 500°C. This might be because of the fact that cathode has larger particle size, thus the voids between particles is high compared to anode and electrolyte. Therefore, these large voids lead to more additive to be located between particles. As a result, the amount of evaporated additives at specific places becomes higher and causes big ripples on the surface (Figure 3 temperature between 200°C and 500°C for cathode) and inhomogeneous sintering. These problems might bring about interior stress or small cracks in the material itself and thereby cause the cathode to break at the latest stage of sintering (Figure 3, temperature 900°C and after sintering for cathode). Similarly, electrolyte has inhomogeneous evaporation through the material itself (Figure 3, temperature from 200°C to 350°C for electrolyte) and becomes an issue at the early stage of sintering. However, this issue is not as severe as that of the cathode because it has smaller particle size and therefore the amount of additives between particles is less than that of the cathode. These factors result in better control of additives’ evaporation but still cause curvature formation (Figure 3, temperature from 200°C to 350°C for electrolyte). However, if the curvature formation of electrolyte at the early stage of sintering cannot be controlled then it can lead to permanent curvature formation of cell during sintering (Figure 3, after sintering for electrolyte). Conversely, the anode has no problem during sintering even though heating rate is high. This might be ascribed to the anode material composition (NiO-CGO).

In light of such information, five different sintering profiles were determined for the same five anode supported cells. All cells were placed on a porous alumina substrate, with the cathode adjacent to it (facing downward); through
this, the aim is to suppress possible curvature formation (deriving from the high shrinkage of the cathode) by utilizing the cell’s own weight against the shrinkage behavior. In addition, the densification of the cathode was further impeded by the porous structure of substrate. Figure 4 illustrates different sintering profile and Figure 5 shows sintering results of five cells. It can be seen that heating rate up to 500°C and between 900°C and 1200°C was carefully defined to prevent any cracks. Figure 5 also shows that, cells sintered at sintering 1, 2, and 3 conditions had cracks and severe curvature formation due to stress occurred. Decreasing dwelling time and cooling rate has no visible enhancement on sinterability of these cells. Decreasing heating rate after debinding, from 3°C min⁻¹ to 2°C min⁻¹ at the range from 500°C to 900°C and to 1°C min⁻¹ between 900°C and 1200°C, enhanced cell sinterability. Cells sintered at sintering conditions 4 and 5 have only uniform curvature evolutions due to the fact that stress occurred between each layers were carefully controlled by defining suitable heating rates during sintering, especially at the stage of debinding, shrinkage and cooling. Sintering 5 describes that increasing heating rate during debinding and cooling has no big effects on cell formation in comparison to sintering 4. However, it decreases sintering time, thereby saving energy. Therefore, sintering profile of cell 5 is chosen to be used for sintering of final cells used in this study, which comprises sintering at 1200°C for 60 minutes with a heating rate of 1°C min⁻¹ up to 500°C for debinding, 2°C min⁻¹ up to 900°C and 1°C min⁻¹ up to 1200°C while the cooling rate is 3°C min⁻¹.

The anode is required to possess enough thickness to provide mechanical support and to allow fuel reforming as well as electrochemical reactions for SC-SOFCs. Previous experimental results show that defining the optimum hot laminating condition and sintering profile is not enough to obtain planar anode-supported SC-SOFC. The curvature formation still occurs through crack and delamination issue were eliminated. Therefore, in the following work, the anode thickness was increased from 200 μm to 400 μm and to 800 μm, so as to examine the effect of anode thickness on cell sinterability by single-step co-sintering at the sintering conditions defined above and on curvature formation. Similarly, electrolyte thickness decreased from 40 μm to 20 μm for decreasing ohmic losses and to investigate its effect on single-step co-sintering. Therefore, five different SC-SOFCs were made; their dimensional properties are given in Section 2.2.

Figure 6 depicts the sintering results of Cell A, B, C, D, and E along with their SEM images. It can be clearly seen that increasing anode thickness from 200 μm to 400 μm decreased the curvature formation significantly in the cell (Figure 6; cell B, curvature height from 3.87 mm to 2.29 mm). This is likely because increasing anode thickness enhanced the cell strength and therefore led to the cell becoming more robust towards stress caused by thermal expansion mismatch between layers during sintering. In
Cell A 10:2:2 thickness ratio, 200:40:40 μm thickness.
Curvature -height: 3.87 mm
Estimated porosity:
Cathode: %24.17
Electrolyte: %16.04
Anode: %29.23

Cell B 20:2:2 thickness ratio, 400:40:40 μm thickness.
Curvature -height: 2.29 mm
Estimated porosity:
Cathode: %25.59
Electrolyte: %16.09
Anode: %30.36

Cell C 20:1:2 thickness ratio, 400:20:40 μm thickness.
Curvature -height: 1.97 mm
Estimated porosity:
Cathode: %24.98
Electrolyte: %15.76
Anode: %29.70

Cell D 40:1:2 thickness ratio, 800:20:40 μm thickness.
Curvature -height: 1.42 mm
Estimated porosity:
Cathode: %25.22
Electrolyte: %15.82
Anode: %30.34

Cell E 40:1:2 thickness ratio, 800:20:40 μm thickness, sintered with 5x5 cm porous (7.31 gr) cover plate on top of it.
Curvature -height: 0.26 mm
Estimated porosity:
Cathode: %26.32
Electrolyte: %15.67
Anode: %29.98

**FIGURE 6** Different anode supported planar cells. Cell A, B, C, and D sintered free but Cell E sintered with a porous cover plate (5 x 5 cm 7.31 gr on the top of it) SEM pictures [Color figure can be viewed at wileyonlinelibrary.com]
other words, the compressive and tensile stresses from the anode-electrolyte mismatch were decreased by increasing the cross-sectional area of the anode normal to the length direction. Therefore, the effect of cathode shrinkage (ie, the cathode-electrolyte mismatch) on overall cell curvature was mitigated by increasing anode strength. Furthermore, decreasing electrolyte thickness from 40 μm to 20 μm mitigated curvature evolution further (Figure 6; cell C, curvature height from 2.29 mm to 1.97 mm). This could be explained by that when the electrolyte is sintered separately, it is expected to have no curvature or defects after sintering as anode (Figure 3). However, one can observe that electrolyte has curvature at the edges due to interior stress caused by either nonuniform burnout of binders or inhomogeneity of electrolyte. Thus, reducing electrolyte thickness may have decreased stresses caused by electrolyte. When the anode thickness was increased from 400 μm to 800 μm, the curvature formation diminished some more but there was still a bit curvature at the cell (Figure 6, cell D, curvature height: 1.42 mm). Increasing anode thickness too much may have negative effects on the cell’s performance due to imposing diffusion/gas transport impediments, particularly related to product removal in the case of a cell being situated as a flow-through (perpendicular to the gas flow) arrangement in the fixture. Thus, it is important to explore different methods to eliminate curvature formation rather than continuously increasing anode thickness. Therefore, a porous alumina cover plate (7.31 g) was placed on the top of the stacked green layer specimens to improve homogeneous evaporation during debinding and oppress curvature evolution during sintering. Figure 6, cell E, depicts that there is far less curvature measured (0.26 mm vs 1.42 mm) when utilizing a cover plate on the top of the cell during sintering.

When inspecting the SEM images in Figure 6, all cells appear to have highly similar microstructures respective to their anode, electrolyte, and cathode layers, in terms of particle size and distribution. Across all cells, there is consistently good adhesion between anode-electrolyte, and cathode-electrolyte layers; however, they all have a porous electrolyte that is acceptable in the SC-SOFC configuration. The estimated porosity of the anode, electrolyte, and cathode increased slightly by increasing the anode thickness (Figure 6, anode thickness increment from Cell A to Cell B and from Cell C to Cell D). This might be as a result of the aforementioned cathode shrinkage being suppressed by increasing anode strength. In addition, the effect of cathode shrinkage on thicker anode sinterability became less. Moreover, putting a porous alumina cover plate on cell (Cell E) gave rise to slightly decreased anode and electrolyte porosities but increased that of cathode. This could be explained by the fact that particles in electrolyte and anode were forced by interior stresses caused by interface stresses to shrink on the longitudinal directions rather than causing curvature formation. This phenomenon might have increased anode and electrolyte sinterability but reduced that of cathode, hence resulting in increased porosity of the cathode. In addition, the cathode in all the cells possesses coarser microstructure than the anode as a result of over-sintering. Nonetheless, electrolyte porosity might be removed either by adding some sintering aids in order to reduce sintering temperature of electrolyte or reducing electrolyte particle size to improve its sinterability though these could affect the performance of the cell. Furthermore, the XRD result of the final planar cell with no curvature (specimen E) also shows that there is no third phase formation (see Figure 7).

4 CONCLUSIONS

In this study, an anode-supported SOFC for single-chamber working conditions was made via single-step co-sintering fabrication method. The results show that single-step co-sintering is possible to be applied to the area of SOFC
fabrication more generally, though there are certain problems to be overcome, such as obtaining dense electrolyte. The results also display that curvature formation can be suppressed by either defining better sintering conditions, changing the thickness of anode or electrolyte, and utilizing alumina porous cover plate on the top of the cell during sintering. Increasing the thickness of the anode led to an increase in cell strength as well as a reduction in curvature evolution at the cell edges. Decreasing electrolyte thickness also resulted in a decrease in curvature formation. However, increasing anode thickness and decreasing electrolyte thickness were not enough to obtain curvature-free anode supported SC-SOFCs. Therefore, a porous alumina cover plate (7.31 g) was used on top of the anode to suppress curvature formation during sintering. After determining optimum anode, electrolyte, and cathode thickness as well as utilizing alumina porous cover plate, an anode supported planar SC-SOFC was successfully made.

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CONFLICTS OF INTEREST

None.

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REFERENCES


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