Experimental Investigation on Potential Effect of Cell Shape and Size on the Residual Stress in Solid Oxide Fuel Cells

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In the manufacturing process of solid oxide fuel cells (SOFCs), the residual stresses and curvature are developed in components due to the differences in material properties of cell layers. Residual stress may lead to the crack formation in the cell layers and facilitates cell fracture. In this work, the changes of the residual stress in the electrolyte layer of the anode-supported planar solid oxide fuel cells are experimentally determined at room temperature. The “sin^2ψ” technique of X-ray diffraction method is employed to measure the residual stress in the half-cell samples. Investigation on the changes of the residual stress and curvature state in the scaling-up process of the cell is crucial for commercial use. Therefore, several cells with different sizes and shapes are investigated to evaluate the potential impact of cell size and cell shape on the residual thermal stress. Values of about −610 MPa are determined for the electrolyte layer on an oxidized ~400 μm thick anode substrate. The results reveal that despite the effect of size and shape on the radius of curvature, these parameters have no significant impact on the residual stress level.

Keywords: solid oxide fuel cell, residual stress, radius of curvature, cell size, cell shape, X-ray diffraction

1 INTRODUCTION

A solid oxide fuel cell (SOFC) is a high-efficient electrochemical device that directly converts the chemical energy of fuels such as hydrogen and natural gas into electrical energy. In various configurations and designs, anode-supported planar SOFCs are widely used due to their higher power density and lower manufacturing cost (Ettler et al., 2010; Fan et al., 2014; Du et al., 2018; Shin et al., 2020; Padinjarethil et al., 2021).

Residual stresses are developed during the manufacturing process of the cells due to the thermo-elastic mismatch between the cell layers (Selçuk et al., 2001; He et al., 2011; Molla et al., 2013; Wei et al., 2018; Shang et al., 2019; Frandsen et al., 2021). These residual stresses can result in performance degradation (Li et al., 2020; Liu et al., 2021) as a result of delamination (Selçuk et al., 2001) or the formation of cracks in the cell layers (Selçuk et al., 2001; Ettler et al., 2010; Li et al., 2010) and in the extreme case may lead to the complete deterioration of the cell (Steinbrech, 2008; Zhang et al., 2008; Villanova et al., 2010).

In the case of the anode-supported cells, much attention has been paid to the residual stress distribution after the cell fabrication process at room temperature and/or during its high-temperature operation, especially in the electrolyte and anode layers. It should be noted that because of low stiffness and low thickness of cathode in the anode-supported cells, the cathode layer has little effect on the stresses in the other components (Sun et al., 2009; He et al., 2011).
Figure 1 shows a schematic cross-section of the residual stress distribution in an anode-supported cell at room temperature. The stress is compressive within the electrolyte layer, and its value does not change significantly through the electrolyte thickness. The anode experiences tensile stress towards the electrolyte and compressive stress towards its free surface. The stress level at the free surface is lower than that at the interface (Severson and Assadi, 2013; Xiang et al., 2014; Wei et al., 2018).

For conventional anode (Ni/YSZ) and electrolyte (YSZ) materials, depending on the fabrication procedure and cell design, the maximum compressive stress in the electrolyte is as high as 500–700 MPa (Yakabe et al., 2004; Fischer et al., 2005; Laurencin et al., 2008; Zhang et al., 2008; Malzbender et al., 2009; Severson and Assadi, 2013; Xiang et al., 2014; Wei et al., 2018). The maximum tensile and compressive stresses in the anode are respectively about 20–100 MPa and 10–50 MPa (Laurencin et al., 2008; Zhang et al., 2008; Severson and Assadi, 2013; Greco et al., 2014; Xiang et al., 2014; Wei et al., 2018).

Figure 2 shows a similar trend for residual stress distribution in an anode-supported cell at operating temperature of 800°C. However, the operating temperature reduces the absolute stress level by about 50% (Fischer et al., 2005; Laurencin et al., 2008; Xiang et al., 2014) or even higher (Frandsen et al., 2021).

During cell operation, chemical stresses (e.g., redox and chemical expansion) and thermally induced stresses are also superimposed to the residual stresses. These stresses are the primary cause of the failure of the cell (Kakaç et al., 2007; Delette et al., 2012) and should not exceed the strength of the materials (Fischer et al., 2005; Malzbender and Steinbrech, 2007; Malzbender et al., 2008, Malzbender et al., 2009). Since the residual stress significantly affects the magnitude and distribution of stresses in the cell at operating conditions (Clague et al., 2012), estimation of residual stress at room temperature would be beneficial to calculate the stress in the anode-supported cells under operating conditions (Yakabe et al., 2004; Zhang et al., 2008; Lin et al., 2009).

The effects of various factors such as layers thickness, using an additional layer, applying different additives, sintering temperature, cell configurations and fabrication method on the residual stress of anode supported cells have been studied by researchers.

Zhang et al. (2008) developed an analytical model to predict the residual thermal stresses in a single solid oxide fuel cell. They investigated the influence of the thickness of each layer on the residual stress distributions in the cell. In a similar study, Fan et al. (2014) calculated the relationship between the residual stresses and the thicknesses of different cell components when the cell is cooled down to the room temperature. Laurencin et al. (2008) proposed a numerical model to study the risk of cell failure due to residual stresses and investigated the effect of electrolyte thickness on the risk of anode failure. Severson and Assadi (2013) developed a structural model to analyze residual stresses in anode- and electrolyte-supported planar SOFCs and they studied residual stress distribution for different thickness combinations.

Malzbender et al. (2006) showed that applying an additional layer, as a support for the anode layer, compensates the cell curvature. However, the average residual tensile stress in the anode increases, which could lead to a more significant fracture probability of the anode layer. Sun et al. (2009) compared the effect of thermal cycling on residual stress and distortion in a standard 3-layer cell and one with an additional layer. Charlas et al. (2015) analyzed and discussed the influence of an additional layer on the residual stresses in 4 layers half-cells.

He et al. (2011) showed that adding Al₂O₃ in NiO-YSZ support materials affected the thermal expansion mismatch and reduced the residual stress in the cell. Cologna et al. (2010) tailored the electrolyte composition by adding a fraction of fine powders to coarse powders to reduce the sintering stresses.

Yakabe et al. (2004) studied the effect of sintering temperature on the calculated residual stress in the electrolyte at room
temperature with X-ray measurements. Malzbender et al. (2008) determined residual stress of half-cells with oxidized anode as a function of temperature.

Fujita et al. (2011) and Somekawa et al. (2013) estimated and compared residual stresses in the electrolytes of segmented-in-series solid oxide fuel cells (SIS-SOFCs) and usual anode-supported cells at room temperature by X-ray diffraction. Nakajo et al. (2011) used a model based on the Euler–Bernoulli theory to study the residual stresses in the cell layers. Using the temperature-dependent mechanical properties of materials has enabled the study of the residual stress in several anode-supported SOFC configurations. Menzler et al. (2014) developed a novel route to fabricate anode-supported solid oxide fuel cells, and they measured the residual stresses in the electrolyte after sintering, before and after flattening. The stress level is significantly reduced compared to the data obtained for half-cells manufactured via the classical route.

Fischer et al. (2005) showed that the flattening procedure of the SOFCs to remove the warp, essentially does not change the residual stress level; however, it reduces the in-plane fluctuation. Moon et al. (2011) concluded that applying compressive force during co-firing can affect the residual stress distribution. They fired a cell under optimal pressure showed homogeneous stress distribution. Shin et al. (2020) showed that the roll calendaring process could produce cells with lower and more uniform residual stress than the conventional uniaxial press.

Many researchers have developed equations for calculating the residual stress and radius of curvature during the sintering process based on fundamental stress-strain relations (Lee et al., 2004; Malzbender et al., 2006; Chang et al., 2008; Cologna et al., 2010; Xiang et al., 2014). Using these equations, the impact of different parameters on residual stress and radius of curvature were investigated, including layer thickness, cell configuration, sintering temperature, and material properties. These equations do not consider the dimensions and geometry of cells and implicitly assume that the size and geometry do not affect residual stress and radius of curvature.

However, some experiments showed that the dimension and geometry of cells affect the radius of curvature. Aguilar-Arias et al. (2013) showed that the increase in cell diameter increases the radius of curvature. Converting deflection data of Moon et al. (2011) and Orui et al. (2008) into the radius of curvature demonstrated that the radius of curvature increases with increasing dimension. Mücke et al. (2009) showed that the radius of curvature of the horizontally sintered specimens is three times higher than the free-hanging samples. This difference is attributed to the effect of gravity and the weight of the samples. Molla et al. (2013) presented an improved model that effect of the weight of the sample (gravity) on the kinetics of distortion is considered. Even under identical conditions, large cells are received more force due to the gravity effect. Malzbender (2010) also investigated the effect of cell geometry on the curvature radius of cells.

On the other hand, the commercial cells are usually much larger than the samples made in the laboratories or used in the research experiments. So, while researchers use small-size cells to conduct preliminary studies on the effect of various parameters on cell performance, it is still necessary to enlarge the cell size for practical and commercial use. Any changes of the residual stress and curvature state in this scaling-up process should be investigated. Regarding the effect of dimension on the radius of curvature and the relationship between curvature and residual stress, we experimentally investigated the potential effects of the shape and size of ceramic cells on the residual stress.

In this work, we report our evaluation of the residual stresses in the electrolyte of anode-supported solid oxide fuel cells, which reveals the effect of the shape and size of cell on residual stresses. All cells have been fabricated by the conventional tape casting and co-sintering method. The conditions of the co-firing and tape-casting processes were identical for all specimens. We used the X-ray diffraction method to measure the residual stresses in the electrolyte of the anode-supported cell. This technique has been widely used to evaluate the residual and thermal stress in solid oxide fuel cells (Yakabe et al., 2004; Fischer et al., 2005; Sumi et al., 2006; Malzbender et al., 2009; Huang and Harter, 2010; Yang et al., 2013; Wei et al., 2018).

2 MATERIALS AND METHODS

2.1 Sample Preparation

Commercial 8YSZ (FCM, United States) powder with a surface area of 6.7 m² gr⁻¹ and NiO-YSZ cermet powder (FCM, United States) with a surface area of 2.4 m² gr⁻¹ were used for the fabrication of the electrolyte and the anode layers, respectively. The electrolyte slurry was cast on a Si-coated Mylar sheet using a homemade tape caster. After drying, the anode slurry was cast on the electrolyte layer. Dried green half-cell was cut in suitable geometries to acquire the desired sizes and shapes after sintering. Figure 3 shows the small half-cells after this step. Further experimental details of the fabrication process are described in previous works (Azari et al., 2015; Azari et al., 2016).

Burn-out process was performed at 1000°C for 2 h in air atmosphere. After the burn-out procedure, for obtaining the desirable mechanical properties, pre-sintered half-cells were sintered at 1400°C for 4 h in air atmosphere. Figure 4 displays the large half-cells after this step.
Anode and electrolyte thicknesses of all samples are about 400 μm and 30 μm, respectively. The open porosity of the sintered anode was measured by the Archimedes method using distilled water as the immersion medium. The open porosity of the anode was 14.4 Vol% to total anode volume. Specifications of nine fabricated half cells are shown in Table 1.

### 2.2 Residual Stress Measurement

The residual stresses in the electrolyte of the anode-supported half cells were estimated using an X-ray Philips X'Pert diffractometer. The conventional sin²ψ method and iso-inclination technique were used, and the Cu-Kα X-ray source was selected. In this method, a diffraction peak at a high 2θ angle is used because, in this case, applying a large ψ range is possible. (Yakabe et al., 2004). For this purpose, a preliminary scan was first conducted from 90 to 140° (2θ), which is displayed in Figure 5. Based on the results of the initial scan, the reflection plane (5 3 1) was selected. The diffraction angle 2θ is about 125°.

The residual stresses in the electrolyte were estimated using the following equation (Yakabe et al., 2004; Fujita et al., 2011):

\[
\sigma = \frac{1}{d_0} \times \frac{E}{1 + \nu} \times \frac{\partial d_0}{\partial \sin^2 \psi}
\]

where \(d_0\) is the interplanar spacing under a stress-free condition, \(\psi\) the tilt angle, \(E\) is Young’s modulus of 215 GPa, and \(\nu\) is Poisson’s ratio of 0.3 (Fischer et al., 2005; Fujita et al., 2011; Menzler et al., 2014).

The stress was evaluated assuming a plane unidirectional stress field. Diffraction measurements were taken at 9 ψ-angles (negative and positive inclinations) at equal distances in sin²ψ from 0 to 0.5, using a step of Δθ = 0.05° in the neighborhood of 125°.

### 3 RESULTS AND DISCUSSION

#### 3.1 Effect of Cell Shape on the Residual Stress

The shift of the diffracted (5 3 1) peak of YSZ with the change of ψ for the sample c3 is illustrated in Figure 6. For all samples, the peak position shifted to a higher angle with increasing ψ.

For all samples, the corresponding d-spacing of various tilt angles ψ were determined from the detected YSZ (5 3 1) diffraction peak positions. Figure 7 shows the d–sin²ψ diagram for sample c3. As seen, the d-spacing decreases with the increase of sin²ψ, implying that the residual stress is compressive in this specimen. The residual stresses are estimated from the d–sin²ψ diagram using Eq. 1.

For small cells, the residual stresses in the electrolyte were calculated and listed in Table 2. The average value of 610 MPa was calculated for residual stress in the electrolyte layer in this work. It can be seen that the estimated values are in agreement with the values reported in the literature for similar anode-supported cells. Yakabe et al. (2004) reported 774 MPa for similar cells with anode and electrolyte thicknesses of 2000 μm and 30 μm, respectively. According to Yakabe simulation (Yakabe et al., 2004), for similar cells with different shapes, the residual stresses in the electrolyte were calculated.

#### 3.1.1 Table 1: General specifications of the half cells.

| Specimen No. | Shape     | Dimension (mm) | Area (cm²) | Size |
|--------------|-----------|----------------|------------|------|
| r-1          | Rectangle | Length = 29.5, Width = 18 | 5.3 | Small |
| r-2          | Rectangle | Length = 29.5, Width = 18 | 5.3 | Small |
| s-1          | Square    | Side = 23      | 5.3 | Small |
| s-2          | Square    | Side = 23      | 5.3 | Small |
| c-1          | Circle    | Diameter = 26  | 5.3 | Small |
| c-2          | Circle    | Diameter = 26  | 5.3 | Small |
| c-3          | Circle    | Diameter = 26  | 5.3 | Small |
| C-1          | Circle    | Diameter = 105 | 86.6 | Large |
| C-2          | Circle    | Diameter = 105 | 86.6 | Large |
et al., 2004), a decrease of anode layer thickness from 2000 μm to 400 μm (with a constant electrolyte layer thickness of 30 μm) can reduce electrolyte stress level by about 150 MPa.

The results in Table 2 reveal that the shape of the cells has little effect on the residual stresses and can be ignored. This is not unexpected, because the main cause of the residual stress is the difference in the material properties of the layers, and any potential effect of other parameters should be studied.

It should be noted that all samples have been fabricated and sintered under identical conditions and all the effective parameters in the cell production route are the same. They are cut out of the same tape and sintered together in the furnace. However, the results of Table 2 also show that the samples with the same shape, have different residual stresses. This fluctuation in residual stress under the same experimental conditions has already been reported (Yakabe et al., 2004; Fischer et al., 2005; Sun et al., 2009; Fujita et al., 2011). Because, even under apparently identical conditions of the production process, microstructure may be inhomogeneous (e.g. density and distribution of the crack). Inhomogeneity in the microstructure of ceramic can affect the material properties, and properties of different regions would be different. For example, the thickness of the tape as an effective factor on the residual stress level is not equal in length of a tape.

Therefore, the X-ray technique is unable to precisely detect the residual stresses due to changes in the cell shape. Although the stress is lowest for square cells, by taking into account the intrinsic errors of the method and the errors generated by assumptions (i.e. plane unidirectional stress field), no absolute conclusion can be
made. In other words, the conventional \( \sin^2 \psi \) method cannot measure the probable effect of cell shape on the residual stresses.

### 3.2 Effect of Cell Size on the Residual Stress

The X-ray stress evaluation method was also used to study the effect of cell size on the residual stress. The XRD patterns for residual stress measurement of a large cell (C1) are shown in Figure 8.

Figure 9 shows the \( d - \sin^2 \psi \) diagram for sample C1. The estimated residual stress in the electrolyte was compressive stress of 610 MPa for sample C1 and 618 MPa for sample C2.

Comparing this result for large cells with small cells data (Table 2) shows that the residual stress is independent of the cell size. This is also consistent with the previous simulations for anode-supported cells (Yakabe et al., 2004). Simulation results reported by Yakabe and co-workers (Yakabe et al., 2004) show that when the cell size is larger than 5 mm, the calculated stress is almost independent of the cell size.

Results of this section revealed that scaling up the cell size does not affect the residual stress state. However, this does not mean that scaling up the cells to larger sizes does not affect the probability of cell fracture. Because the increase in cell size is associated with the statistical probability of larger defects which can decrease the fracture stress of large cells in comparison with smaller specimens in an identical residual stress state (Fischer et al., 2005).

On the other hand, due to the mismatch between the material properties of the individual layers, residual stresses and warpage are evolved during the manufacturing process. Some modifications, such as using additives or the decrease of sintering temperature, can reduce both residual stress and warpage (Yakabe et al., 2004; He et al., 2011; Menzler et al., 2014). Some modifications, such as using a compensation layer, can reduce warpage, but increases residual stress (Malzbender et al., 2006). Our results demonstrate that despite the effect of cell size on the warpage behavior, cell size does not affect residual stress.

### 4 CONCLUSION

Using large-scale SOFCs is very crucial for practical and commercial use. Changes in the residual stress and curvature state in the scaling up process of cells should be investigated. This work was an effort to address the stress state in the electrolyte layer of the anode-supported planar solid oxide fuel cell. The residual stress of the YSZ electrolyte was evaluated using the
X-ray sin²ψ stress measurement method. The changes in the residual stress were measured to investigate the impact of cell size and cell shape on the residual stress.

The XRD measurements were carried out at room temperature in the electrolyte of half-cells. In the case of a ~30 μm thick electrolyte layer on an oxidized ~400 μm thick anode substrate, the estimated residual stresses in the electrolyte were compressive stresses of around 600 MPa. For our specimens, the change of cell shape without changing other parameters did not alter the stress states and the relative distributions. Regarding the intrinsic errors of the method and the errors generated by assumptions, the probable effect of shape is not large enough to be detected by the conventional sin²ψ method.

It was also found that the residual stress in the electrolyte of the anode-supported planar SOFC remained almost constant during the scale-up process, and the change of cell size did not affect the residual stress level.

Based on the results of this study, it can be concluded that despite the effect of size and shape on the radius of curvature, these parameters do not affect the residual stress level.

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DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

KA: Conceptualization, methodology, experimental tests, writing—original draft, writing—review and editing, funding acquisition HA: Conceptualization, investigation, writing—review and editing MT: Investigation, experimental tests, writing—review and editing SB: Investigation, writing—review and editing.

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