Residual stress analysis of a micro-tubular solid oxide fuel cell

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Abstract:

A two-dimensional axisymmetric micro-tubular solid oxide fuel cell (MT-SOFC) model is developed to study the influence of component (anode, cathode or electrolyte) thickness on residual stress at the room temperature. The results show that the anode is mainly subjected to tension stress while electrolyte and cathode are mostly exposed to compressive stress. However, the stress in a component can be reduced by increasing its thickness with other components unchanged. Then the Weibull analysis method is adopted to calculate the failure probability of the anode, it is found that the anode is prone to failure with a thinner anode or thicker cathode and electrolyte; the electrolyte thickness has a more significant effect on the anode failure probability than the cathode; and the component thickness must be coordinated in order to maximize the mechanical performance. The formula of the minimum anode thickness under the failure probability criterion $1E-6$ is given, which brings considerable convenience to the design of MT-SOFC.

Key words: Micro-tubular solid oxide fuel cell (MT-SOFC); Residual stress; Electrode thickness; Failure probability.

1. Introduction

The solid oxide fuel cell (SOFC) has been recognized as an alternative power generation device for its high efficiency, low emission, fuel flexibility and all-solid-state characteristic [1-4]. Among various SOFC designs, planar and tubular are the most maturely developed types [5, 6]. For the tubular configuration, the Micro-tubular SOFC (MT-SOFC) possesses great advantages over conventional tubular SOFC (T-SOFC) and planar SOFC (P-SOFC), such as higher power density, higher mass transfer efficiency, stronger mechanical property and lower thermal stress [7, 8].

However, the MT-SOFC development still encounters a lot of issues: on the one hand, the cell's mechanical strength is disturbed due to the fine micro-tubular structure, which will limit the output power; on the other hand, it is a challenging work to fabricate and assemble the cells because of its micro-scale. One of the main reasons for MT-SOFC's short lifetime is the internal stress, which would finally crack and damage the cell during a long-time impact. So the fracture and failure of MT-SOFC can be predicted by analyzing the internal stress.

The thermal stress of SOFC has been widely studied in experiments [9-12] and numerical model [13-16], especially for the P-SOFC: Chen et al. [17] constructed a three-dimensional finite element analysis model and investigated the status of thermal stress in a single P-SOFC induced by a uniform temperature field. Nakajo et al. [18] established a multi-physics model and studied the stress of SOFC when they are in operation state and cool-down phase. Yakabe et al. [11, 19] built a three-dimensional

model of anode-supported P-SOFC with STAR-CD to analyze residual stress in electrolyte. Selimovic et al. [20] focused on the stress in P-SOFC caused by the thermal expansion incompatibility under high temperature gradients during steady and dynamic operation. Fan et al. [21] researched the thermal stress distribution in the positive electrode-electrolyte-negative electrode (PEN) via a finite element model. And Weil et al. [22] investigated the stress performance of the new bonded compliant seal (BCS) utilizing the ANSYS. However, in contrast to the intensive reports on mechanical property of P-SOFC, the studies about MT-SOFC's stress are few and recent. Serincan et al. [23] and Cui et al. [24] reported a MT-SOFC multi-physics model, then investigated the internal stress based on the temperature obtained from the thermal-fluid model; Xu et al. [25] did some researches aiming at the influence of operation temperature and temperature gradient on the state of the stress. Nevertheless, the effect of MT-SOFC component (anode, cathode or electrolyte) thickness on the stress distribution and the failure probability hasn't been studied in detail and the component thickness is irregularly chosen with little concern for stress stability, which is unfavorable for the long term stability of MT-SOFC [26-29].

In this study, a two-dimensional (2D) axisymmetric model is established and validated to analysis the effect of component thickness on the residual stress and the failure probability, then the minimum anode thickness under the failure criterion $1E-6$ is carefully commented.

2. Numerical Model

2.1 Structural mechanics relation

It is assumed that the MT-SOFC materials in this model meet the linear elastic theory. The total strain consisting of elastic strain, thermal strain and initial strain can be written as:

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}_{el} + \boldsymbol{\varepsilon}_{th} + \boldsymbol{\varepsilon}_0 \quad (1)$$

where $\boldsymbol{\varepsilon}$ is the total strain, $\boldsymbol{\varepsilon}_{el}$ is the elastic strain, $\boldsymbol{\varepsilon}_{th}$ is the thermal strain, $\boldsymbol{\varepsilon}_0$ is the initial strain. In which, the thermal strain can be expressed as:

$$\boldsymbol{\varepsilon}_{th} = \alpha(T - T_f) \quad (2)$$

where α denotes the thermal expansion coefficient of the material; T refers to local temperature where thermal stress is determined; T_f represents the stress-free temperature corresponding to thermal stress of zero.

Therefore, the stress could be expressed as:

$$\boldsymbol{\sigma} = D\boldsymbol{\varepsilon}_{el} + \boldsymbol{\sigma}_0 = D(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}_{th} - \boldsymbol{\varepsilon}_0) + \boldsymbol{\sigma}_0 \quad (3)$$

where $\boldsymbol{\sigma}$ is the stress vector, D represents the elasticity matrix, $\boldsymbol{\sigma}_0$ is the initial stress. For isotropic material, elasticity matrix D is:

$$D = \frac{E}{(1+\nu)(1-2\nu)} \begin{bmatrix} 1-\nu & \nu & \nu & 0 \\ \nu & 1-\nu & \nu & 0 \\ \nu & \nu & 1-\nu & 0 \\ 0 & 0 & 0 & \frac{1-2\nu}{2} \end{bmatrix} \quad (4)$$

where E is the Young's modulus and ν is the Poisson's ratio.

2.2 Geometric model

An anode-supported MT-SOFC three-dimensional model is schematically showed in **Fig. 1a**, which is composed of cathode, electrolyte and anode from outside to inside. In this manner, the fuel goes through the inner anode tube and the air

contacts with outer cathode in order to supply oxygen advantageously. However, considering the axisymmetric structure, the computational domain can be simplified into a 2D section as shown in **Fig. 1b**.

The detailed input parameters of the model are summarized in **Table 1**.

2.3 Mechanical property of materials

This study here focused on the most typical MT-SOFC in configuration of porous composite Ni-YSZ anode/dense YSZ electrolyte/porous composite LSM-YSZ cathode, the effective properties of the porous composite can be calculated by following method [30].

The bulk modulus K_{com} and shear modulus G_{com} of the dense composite material are attained by:

$$K_{com} = K_2 + \frac{\varphi_1}{1/(K_1 - K_2) + 3\varphi_2/(3K_2 + 4G_2)} \quad (5)$$

$$G_{com} = G_2 + \frac{\varphi_1}{1/(G_1 - G_2) + 6\varphi_2(3K_2 + 2G_2)/(5G_2(3K_2 + 4G_2))} \quad (6)$$

where K_i and G_i are the bulk and shear modulus of material i , respectively, φ_i denotes the volume fraction of material i .

The Young's modulus of the dense composite materials E_{com} is calculated by:

$$E_{com} = \frac{9K_{com}G_{com}}{3K_{com} + G_{com}} \quad (7)$$

The thermal expansion coefficient of the two-phase composite dense material α_{com} is given by:

$$\alpha_{com} = \frac{\alpha_1\varphi_1K_1 + \alpha_2\varphi_2K_2}{\varphi_1K_1 + \varphi_2K_2} \quad (8)$$

The effective Young's modulus E_{eff} and shear modulus G_{eff} of the porous electrodes are calculated by:

$$E_{eff} = E_{com} \frac{(1-\phi_g)^2}{1+\phi_g(2-3\nu_{com})} \quad (9)$$

$$G_{eff} = G_{com} \frac{(1-\phi_g)^2}{1+\phi_g(11-19\nu_{com})/(4+4\nu_{com})} \quad (10)$$

where ϕ_g is the porosity, the subscript “com” represents the effective properties of dense material.

The effective Poisson's ratio of the porous electrode ν_{eff} is:

$$\nu_{eff} = \frac{1}{4} \frac{4\nu_{com} + 3\phi_g - 7\nu_{com}\phi_g}{1 + 2\phi_g - 3\nu_{com}\phi_g} \quad (11)$$

Density of the porous electrode ρ_{eff} :

$$\rho_{eff} = (\rho_1\rho_1 + \rho_2\rho_2)(1-\phi_g) \quad (12)$$

The properties of dense material at the room temperature are listed in **Table 2**.

2.4 Failure probability analysis

Ceramic material fractures easily under certain stress intensity due to the brittle nature, its failure probability can be determined by Weibull method. For MT-SOFC, the failure probability mainly depends on three principal stresses because the MT-SOFC is subjected to multidirectional stresses, so it can be written as:

$$P_f = 1 - \prod_{i=1}^3 \exp\left(-\int_V \left(\frac{\sigma_i}{\sigma_0}\right)^m \frac{dV}{V_0}\right) \quad (13)$$

where V_0 is the reference volume, V is the volume of the SOFC component, σ_0 represents the Weibull strength, m signifies the Weibull modulus and σ_i ($i=1, 2, 3$) is the first, second, and third principal stress, respectively. According to the

definition of principal stress, there is $\sigma_1 > \sigma_2 > \sigma_3$, meanwhile, a positive σ_i represents tensile stress and a negative σ_i means compressive stress. It is noteworthy that the Weibull method can only be applied to the tensile stress.

2.5 Model validation

The model is verified by comparing the distribution and magnitude of the three principal radial stresses at room temperature with model data from literature [30]. The first and second principal stresses are shown in the **Fig. 2a**, and the third principal stress is presented in **Fig. 2b**. It is readily noticeable that the distribution of the principal stresses from this model are in good agreement with the literature data, which proves the validity of the model and qualifies the model for a reliable further study.

3. Result and discussion

3.1 Residual stress

During the sintering process of the MT-SOFC, the anode and electrolyte are assumed to have the same stress free temperature of 1473 K, and that of cathode is 1373 K [30]. The residual stress arises from different thermal expansion coefficient between materials while the MT-SOFC cools down from sintering temperature to room temperature. The maximum residual principal stress at room temperature is shown in **Fig. 3** in which the positive value represents tensile stress and the negative one represents compressive stress. It easy to point out that the anode is mainly subjected to tension stress while electrolyte and cathode are mostly exposed to compressive stress, which is because that the thermal expansion coefficient of anode

is greater than that of electrolyte and cathode. The influence of component thicknesses on residual stress is systematically investigated via parameter sweep.

The effects of anode thickness on the residual stress in anode, electrolyte and cathode are demonstrated in **Fig. 3a**. The tensile stress in anode is decreasing with the increase in anode thickness due to the stress release in the thick anode. However, the compressive stresses in the electrolyte and cathode are increasing. With anode thickness of 0.2 mm, the maximum residual principal stress in anode, electrolyte and cathode are 5.19×10^7 Pa, -7.36×10^8 Pa, -7.94×10^7 Pa, respectively. When the anode thickness increases to 1.0 mm, the compressive stresses in electrolyte and cathode reaches 8.58×10^8 Pa and 9.56×10^7 Pa, respectively, both are on the verge of their compressive strength 1 GPa [19] and 100 MPa [31]. As a consequence, the anode thickness in the MT-SOFC should be appropriate to avoid the stresses in electrolyte or cathode exceeding their compressive strength.

Shown in **Fig. 3b** is the impact of electrolyte thickness on the residual stress. A thick electrolyte tends to raise the tensile stress in the anode, but the compressive stresses in the electrolyte and cathode can be reduced. In other words, increase in electrolyte thickness can enhance the mechanical stability of the electrolyte and cathode. The tensile stress in the anode increased from 5.19×10^7 Pa to 1.40×10^8 Pa along the electrolyte thickness from 10 μm to 50 μm . Nevertheless, the ceramic electrodes is quite sensitive to the tensile stress, so the electrolyte also cannot be too thick and usually selected in the range of 10 μm -20 μm [24, 28, 29, 32-42].

The plot in **Fig. 3c** shows the evolution of residual stress with the thickening of

cathode. The stresses in the electrolyte and cathode are inversely proportional to the cathode thickness, but the anode stress is opposite. In brief, the mechanical property of electrolyte and cathode can be enhanced with the thick cathode. With the cathode increased from 10 μm to 80 μm , the compressive stress in the cathode and electrolyte decreased by 1.18×10^7 Pa and 6.20×10^7 Pa respectively. As a comparison, in **Fig. 3b**, with the electrolyte increased from 10 μm to 50 μm . The decrements are 4.07×10^7 Pa and 2.60×10^8 Pa for cathode and electrolyte respectively. As a conclusion, the electrolyte has a more significant impact on the residual stress than cathode.

As a brittle material, ceramic is more sensitive to tensile stress than compressive stress because its compressive strength is much higher than tensile strength. A SOFC can be judged failure if the compressive stress exceed compressive strength [30]. Therefore, it is vital to keep the maximum compressive stress in electrolyte and cathode lower than respective compressive strength. However, the life time of the whole MT-SOFC is more dependent on the anode with its tensile stress. For the ceramic material in tensile state, Weibull method is an effective tool to analyze the failure probability. So in this paper, the failure probability is used to evaluate the mechanical performance of the tensile stress suffered anode.

3.2 Failure probability of the anode

Since the tensile stress suffered anode is more likely to failure than electrolyte and cathode, here Weibull analysis was performed to analyze its failure probability. The criterion of the failure probability is elected as $1\text{E-}6$ according to the practical application of SOFC [43]. In order to investigate the effect of component thickness on

the failure probability of the anode, the failure probability of anode was calculated independently via changing the thickness of anode, electrolyte and cathode.

The failure probabilities of anode under different electrolyte thicknesses are displayed in **Fig. 4**. The result reveals that the failure probability of anode increases with increasing the electrolyte thickness, e.g. for a 0.2 mm thick anode, the failure probability increased by two orders of magnitude from 1.54×10^{-6} to 4.92×10^{-4} when the electrolyte increased from 10 μm to 20 μm . Under the adopted failure probability criterion $1\text{E-}6$, the minimum thicknesses of the anode are 0.263 mm, 0.554 mm, 1.024 mm corresponding to the electrolyte thickness 10 μm , 20 μm , 30 μm , which indicates that a thick electrolyte needs to match with thick anode, but it may result in a high Ohmic polarization. Therefore, in order to prevent the MT-SOFC from failure, the electrolyte thickness should be appropriately determined.

Fig. 5 shows influence of cathode thickness on the failure probabilities of anode. The increase of cathode thickness will cause the increase of the failure probability for anode, but the effect is not as significant as that of electrolyte thickness, e.g. the failure probability of anode only increased from 1.54×10^{-6} to 3.39×10^{-6} when the cathode thickness increased from 10 μm to 20 μm under the anode thickness of 0.2 mm, the increase is much smaller compared to that introduced by electrolyte thickness. The minimum anode thicknesses of the MT-SOFC should be 0.235 mm, 0.263 mm, 0.292 mm, 0.321 mm, 0.350 mm, 0.380 mm corresponding to the cathode thickness 10 μm , 20 μm , 30 μm , 40 μm , 50 μm , 60 μm , an approximately linear increase in accordance with criterion $1\text{E-}6$. But thick cathode also needs to match with thick

anode, which raises the cost and is not favorable to the commercialization of MT-SOFC.

3.3 The relationship among the thicknesses of the anode, electrolyte and cathode

According to study of the residual stress and the failure probability, it is obvious that the thickness of the anode, electrolyte and cathode should meet a certain relationship to make the MT-SOFC has higher mechanical stability and long life.

The relationship among the thicknesses of anode, electrolyte and cathode when the failure probability equals to 1E-6 is shown in **Fig. 6**. The range of electrolyte thickness is usually in 10-20 μm [24, 28, 29, 32-42] in the literature with respect to the MT-SOFC. According to straight line plots, the relationship between anode thickness and cathode thickness is approximately linear at a fixed electrolyte thickness.

Therefore, the minimum anode thickness is related to the cathode thickness via following correlation:

$$t_{\min-\text{anode}} = A + B \times t_{\text{cathode}} \quad (14)$$

where A is intercept and B is slope. The values of A and B with different electrolyte thickness are shown in **Fig. 7**. In this fashion, the minimum anode thickness can be easily determined.

As a further study of **Fig. 7a**, it can be found that the A and the electrolyte thickness are in linear relationship, which can be expressed as:

$$A = 0.0287 t_{\text{electrolyte}} - 0.0839 \quad (15)$$

Moreover, **Fig. 7b** displays the relation between the B and electrolyte thickness.

It is clear that the value of B increases from 0.0029 to 0.0047 with increase of the electrolyte thickness from 10 μm to 20 μm , so the B must be related to electrolyte thickness. Then the relationship can be considered as:

$$B = 1.2 \times 10^{-5} t_{\text{electrolyte}}^2 - 3.04 \times 10^{-4} t_{\text{electrolyte}} + 0.0048 \quad (16)$$

As a result, the relationship among the thicknesses of the anode, electrolyte and cathode can be attained by:

$$t_{\text{min-anode}} = (0.0287 t_{\text{electrolyte}} - 0.0839) + (1.2 \times 10^{-5} t_{\text{electrolyte}}^2 - 3.04 \times 10^{-4} t_{\text{electrolyte}} + 0.0048) t_{\text{cathode}} \quad (17)$$

where the unit of the $t_{\text{min-anode}}$ is [mm] and $t_{\text{electrolyte}}$ and t_{cathode} are in [μm].

Once the thicknesses of two layers are given, the thickness of the third one can be acquired precisely by this equation, which brings considerable convenience to the design of MT-SOFC. But it is important to keep in mind that Eq. 17 is derived under the failure probability criterion of 1E-6.

4. Conclusions

A well verified MT-SOFC 2D axisymmetric model has been established in this study to investigate the effect of component thickness on residual stress and failure probability. Based on this model, parametric study and Weibull analysis are implemented to study the residual stress and failure probability, respectively. The most important conclusions are summarized as follows:

- (1) The anode mainly undergoes tension stress while electrolyte and cathode mostly suffer compressive stress.
- (2) For a chosen component (which means the dimensions of other components are fixed), the stress decreases with increasing its thickness.

(3) The failure probability of anode decreases with the increase of anode thickness or the decrease of electrolyte and cathode thicknesses.

(4) The influence of cathode thickness on the failure probability is weaker than electrolyte thickness.

(5) A simply relationship among the thicknesses of anode, electrolyte and cathode under the failure probability criterion $1E-6$ can be written as:

$$t_{\min-anode} = (0.0287t_{electrolyte} - 0.0839) + (1.2 \times 10^{-5}t_{electrolyte}^2 - 3.04 \times 10^{-4}t_{electrolyte} + 0.0048)t_{cathode}$$

The conclusions derived from this study have practical significance to improve mechanical stability and prolong the lifetime of MT-SOFC, which will benefit the application of the MT-SOFC.

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Table 2 Mechanical properties of dense material

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Fig. 1(a) Schematic of the MT-SOFC; **(b)** Schematic of the 2D axisymmetric MT-SOFC model

Fig. 2 Principal radial stress distribution of cell at room temperature

Fig. 3 Maximum principal stress in the PENs with various thicknesses:

(a) $t_{electrolyte} = 10 \mu\text{m}$, $t_{cathode} = 20 \mu\text{m}$;

(b) $t_{anode} = 0.2 \text{ mm}$, $t_{cathode} = 20 \mu\text{m}$;

(c) $t_{anode} = 0.2 \text{ mm}$, $t_{electrolyte} = 10 \mu\text{m}$.

Fig. 4 The anode failure probability with various electrolyte thicknesses

Fig. 5 The anode failure probability with various cathode thicknesses

Fig. 6 The relationship among the thicknesses of anode/electrolyte/cathode

Fig. 7(a) The relation between the A and electrolyte thickness

Fig. 7(b) The relation between the B and electrolyte thickness

Table 1

Input parameters to model	Value
Inner diameter of MT-SOFC tube (mm)	0.85
Volume fraction of Ni (φ_{Ni})	0.445
Volume fraction of LSM (φ_{LSM})	0.479
Anode porosity (ϕ_g)	0.4
Cathode porosity (ϕ_g)	0.4
Anode length (mm)	30
Electrolyte length (mm)	20
Cathode length (mm)	11.8
Anode thickness (mm)	0.15
Electrolyte thickness (μm)	12
Cathode thickness (μm)	20
Anode and electrolyte stress-free temperature (K)	1473
Cathode stress-free temperature (K)	1373
Room temperature (K)	298

Table 2

Material	Young's modulus (GPa)	Poisson's ratio	TEC (10^{-6} K^{-1})	Density (g/cm³)
YSZ	215	0.317	10.8	6.04
Ni	219	0.313	16.5	8.90
LSM	95	0.320	12.4	6.57

Table 3

Component	Weibull modulus	Weibull strength (MPa)	Reference volume (mm³)
Anode (298K)	11.8	187	0.587
Electrolyte (298K)	7.0	446	0.350
Cathode (298K)	7.0	52	1.210

Fig. 1

Fig. 2(a)

Fig. 2(b)

Fig. 3(a)

Fig. 3(b)

Fig. 3(c)

Fig. 4

Fig. 5

Fig. 6

Fig. 7(a)

Fig. 7(b)