Materials and Design 93 (2016) 53–62

Contents lists available at ScienceDirect

Materials and Design

journal homepage: www.elsevier.com/locate/matdes

Growth and residual stresses in the bonded compliant seal of planar solid oxide fuel cell: Thickness design of window frame

Wenchun Jiang [a,*], Yu-Cai Zhang [a,b], W.Y. Zhang [a], Y. Luo [a], W. Woo [c], S.T. Tu [b]

[a] State Key Laboratory of Heavy Oil Processing, College of Chemical Engineering, China University of Petroleum (East China), Qingdao 266555, PR China
[b] Key Laboratory of Pressure System and Safety (MOE), School of Mechanical and Power Engineering, East China University of Science and Technology, Shanghai 200237, PR China
[c] Neutron Science Division, Korea Atomic Energy Research Institute, Daejeon 305-353, South Korea

ARTICLE INFO

Article history:
Received 19 July 2015
Received in revised form 19 December 2015
Accepted 23 December 2015
Available online 24 December 2015

Keywords:
Solid oxide fuel cell
Bonded compliant seal
Growth stress
Residual stress
Neutron diffraction measurement

ABSTRACT

Bonded compliant seal (BCS) is a new sealing method for planar solid oxide fuel cell. The BCS design uses a thin foil to bond the cell and window frame, which generates a multilayer structure. However, the high temperature bonding generates large residual stresses that greatly affect the fracture. This paper presents a numerical method and neutron diffraction measurement to study the residual stress, and effect of window frame thickness has been discussed. A grain boundary diffusion model incorporated with a power-law creep constitutive model is developed to calculate the growth stress in the oxide film. Then, the thermal elasto-plastic finite element method is applied to calculate the thermal stress. A neutron diffraction experiment is performed to measure the through-thickness stresses. A good agreement is found between the calculation results and the neutron diffraction measurements. Compressive stress is generated in the oxide scale because of the substrate constraint. Furthermore, a competition exists between the generation of growth stress and the creep relaxation in the oxide layer. The residual stresses in the oxide layer decrease with the decrease in the substrate thickness. The thicknesses of the window frame and foil are designed to be 500 and 50 μm, respectively.

1. Introduction

The planar solid oxide fuel cell (SOFC) is a promising clean energy device that converts chemical energy into electricity by electrochemical reaction [1]. The cathode and anode sides of the planar SOFC operate in oxidizing atmosphere and wet reducing gas, respectively. This device undergoes several thermal cycles at a high temperature (~750 °C). Such hostile conditions require a good hermetic sealant to prevent the leakage of air and fuel and to effectively isolate the fuel from the oxidant.

Two main sealing methods are available for use in planar SOFCs: rigid-bonded and compressive seals [2]. A rigid-bonded seal [3] uses metal or glass-ceramic as sealant material. Glass or glass-ceramic is very popular because metal brazing (silver and gold) is highly expensive. However, its coefficient of thermal expansion (CTE) is different from that of the SOFC components; thus, this material is prone to thermal stresses and cracks [4]. Improving the life of glass-ceramic sealing by using new composite sealants that can decrease the difference of the CTE is therefore important [5]. A compressive seal [6,7] applies a pressure load on the stack to maintain tightness; this pressure load allows some extent of deformation because the components are not rigidly bonded together. However, the applied load can be relaxed at a high temperature, making this seal unsuitable for long-term operation [8]. Therefore, developing new sealing technologies for SOFCs remains crucial.

Weil et al. recently developed a new sealing method called the bonded compliant seal (BCS), which combines the advantages of rigid and compressive seals [9–10]. The BCS uses a thin foil to bond the cell and the window frame, thereby mitigating a large amount of thermal stress in the cell through elastic or plastic deformation within the sealing foil [11,12]. The material of the thin foil is FeCrAlY alloy with good creep resistance. It contains a high concentration of aluminum that can transform into a thin alumina layer that prevents further oxidation at a high temperature. As a result, a thin multilayer structure that includes an oxide film, a foil, a filler metal, and a window frame is generated. The oxide film contains growth stresses generated by the formation of new oxide within the existing oxide layer [13,14]. Thermal stresses are generated because of the mismatching CTEs [15] between the components. As the structure cools to room temperature, residual stresses are generated by the contraction and plastic deformation. These stresses have a significant influence on fracture [16]. Therefore, an accurate calculation of the two types of stresses must be achieved to ensure safety. In this study, a finite element method (FEM) is developed to predict the growth and thermal stresses in the BCS structure and is subsequently verified by neutron diffraction measurement. The thicknesses of the foil and BNi2 filler metal are set to 80 [17] and 75 μm [18], respectively. However, the design of the window frame thickness is still unclear. In

---

* Corresponding author.
E-mail address: jiangwenchun@126.com (W. Jiang).

http://dx.doi.org/10.1016/j.matdes.2015.12.145
0264-1275/

this paper, we discuss the effect of window frame thickness on residual stress to ultimately provide a reference for thickness design.

Table 1
Chemical composition of FeCrAlY (wt.%).

Fe	Cr	Al	Y	Mn	C	Si
≥69.62	22	4.8	0.3	≤0.04	≤0.08	≤0.70

2. Experimental

2.1. Sample preparation

Fig. 1 shows the cross section of the BCS structure in a planar SOFC. A sealing foil is used to bond the cell and the window frame. The cell is composed of an anode, an electrolyte layer, and a cathode layer. In the present study, we focus on the joint of the sealing foil-to-window frame because of its significance in stress evolution. Therefore, the composite cell was assumed to be the same as the anode material [9–11]. The materials of the sealing foil, filler metal, and window frame were FeCrAlY, BNi2, and Inconel alloy C276, respectively. Their chemical compositions are listed in Tables 1, 2, and 3 [17–19], respectively. Neutron diffraction measurement is an effective method for measuring through-thickness stress, but using it in this study was difficult because the BCS structure is too thin and because the neutron beam only has a mm-scale gauge volume [20]. Therefore, an enlarged sample was prepared, and the residual stress was calculated and compared by neutron diffraction measurement. Then, the validated FEM was used to predict the residual stress in the thin BCS structure.

The specimen was 4 mm thick, 100 mm long, and 100 m wide (Fig. 2). The materials for the top, bottom plates, and filler metal were FeCrAlY, Inconel alloy C276, and BNi2, respectively. The bonding temperature cycle is shown in Fig. 3. First, the specimen was heated up to 850 °C and then held for 30 min. Second, it was heated up to 1100 °C within 30 min and held for 30 min. Finally, it was cooled to ambient temperature.

2.2. Neutron diffraction measurement

Neutron diffraction experiments were performed using the residual stress instrument at the HANARO reactor of the Korea Atomic Energy Research Institute [20]. The Si (220) bent perfect crystal was selected for the monochromator and produced neutrons with the wavelength of 1.46 Å at take-off angles ($2\theta_M$) of 45°. The configuration enabled us to measure the (110) peak at the scattering angles ($2\theta_S$) of 42.2° for the bcc FeCrAlY alloy and the (111) peak at 41.0° for the fcc Inconel alloy C276. The section of the gauge volume (GV) in the diffraction plane was defined by 1 mm-wide cadmium slits on the incident and diffracted beams. The height of the GV was also defined by the 1 mm height of the incident beam. Thus, the GV was ~$1 \times 1 \times 1$ mm^3 for the three orthogonal directions of the specimen. A total of 12 points of the diffraction peaks were measured through the thickness of the specimen at the middle of the plate width and length along the line *P* marked in Fig. 2. Two direction theodolite and levelers were used to fit the exact location for the measurements with the accuracy of about 0.1 mm. Vertical and horizontal alignments were fitted by using procedure (ISO/TS 21432:2005) [21]. Thus, after one component measurement and rotating horizontally 90°, we measured the second component. Finally we vertically rotated 90° and then measured the third strain component. All three components at the exactly same point with the accuracy of 0.1 mm were applied to calculate the stress components. Boron has very strong neutron absorption, but here the BNi2 layer is only 0.075 mm and the measurement is located in the base metal. The peak comes mostly from the chemical components from the gauge volume ($1 \times 1 \times 1$ mm^3).

The measurement of the residual stress using neutron diffraction was based on Bragg's law, which is given by

$$n'\lambda = 2d'\sin\theta \tag{1}$$

where *n'* is an integer, *d'* is the lattice spacing, and 2θ is the diffraction angle. Strain (ε) is determined by measuring the scattering angle (θ) of a material under stress and the scattering angle (θ_0) of the same material that is free of stress.

$$\varepsilon = \frac{\Delta d'}{d'} = -(\theta - \theta_0)\cot\theta \tag{2}$$

The lattice spacing is measured in the direction bisecting the incoming and diffracted neutron beams. The measured peak was fitted by the Gaussian peak fitting method and the peak center was selected to determine the peak shift location consistently. Given that strain and stress have tensor dimensions, measuring the strains in at least three mutually orthogonal directions (x, y, and z) at a certain location is necessary to obtain the normal stress components. The three orthogonal stress components can be calculated using the generalized Hook's law by converting the elastic strains ($\varepsilon_{xx}, \varepsilon_{yy}, \varepsilon_{zz}$) to residual stresses ($\sigma_x$, σ_y, σ_z) along the three orthogonal directions.

$$\sigma_{ii} = \frac{E_{hkl}}{1+\nu_{hkl}}\left[\varepsilon_{ii} + \frac{\nu_{hkl}^{hkl}}{1-2\nu_{hkl}}(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})\right] \tag{3}$$

where $i = x, y$ or z, E_{hkl} and ν_{hkl} are the elastic modulus and Poisson's ratio, respectively. In this study, the diffraction elastic constant (E_{110}) of 225.5 GPa and Poisson's ratio (v) of 0.28 for bcc alloy and the E_{111} of 247.9 GPa and v of 0.24 for fcc alloy were used. The selection of the peak to represent bulk properties is the key issue. The procedure of the neutron residual stress experiment recommends the peak (111) for fcc, (110) for bcc structures because it is known not to obtain anisotropy of the bulk materials.

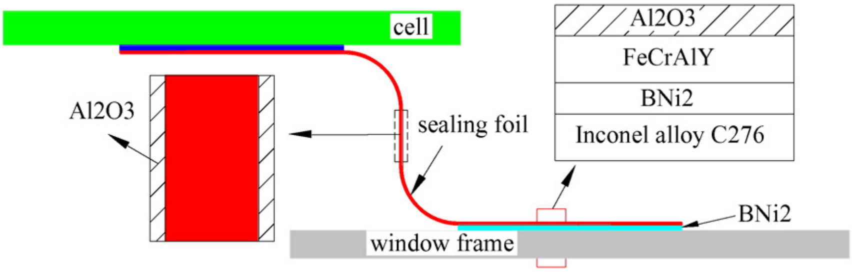

Fig. 1. The cross section of the BCS structure in planar SOFC.