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Model-oriented cast ceramic tape seals for planar solid oxide fuel cells

Shaobai Sang[a,b], Jian Pu[a], Chi Bo[a], Li Jian[a,*]

[a] College of Materials Science and Engineering, State Key Laboratory of Material Processing and Die & Mould Technology, Huazhong University of Science and Technology, 1037 Luo Yu Lu, Wuhan, Hubei 430074, PR China
[b] State Key Laboratory Breeding Base of Refractories and Ceramics, Wuhan University of Science and Technology, Wuhan 430081, PR China

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ABSTRACT

A straight capillary model is developed to estimate the mass leak rate of the cast ceramic tape seals for planar solid oxide fuel cells (SOFCs), which is further rectified with consideration of microstructure complexity including the tortuosity, cross-section variation and cross-link of leak paths. The size distribution of the leak path, effective porosity and the microstructure complexity are the main factors that influence the leak rate of the cast tape seals. According to the model, Al_2O_3 powders are selected for preparation of the seals by tape casting, and the leak rate is evaluated under various compressive stresses and gauge pressures. The results indicate that Al_2O_3 powder with D_{50} value about 2 μm and specific surface area near 5 $m^2\,g^{-1}$ can be used for the cast tape seals; and the obtained leak rate can satisfy the allowable leak limit.

1. Introduction

In planar solid oxide fuel cells (SOFCs), sealing has been regarded as one of the most critical issues that limit the long-term operation of a SOFC stack. The seal material needs not only to be stable in the dual oxidizing and reducing environments but also to be electrically insulating and chemically compatible with other fuel cell components [1–3]. Conventional glass or glass-ceramic seals tend to transform in phases and react with cell components and interconnect materials under SOFC operating conditions in a long run, due to their intrinsic thermodynamical instability [4–6]. The deformable metallic seals, such as ductile silver [7,8] and corrugated or C-shaped super alloy gaskets [9], are limited by their high electronic conductivity. Although the hybrid mica-based seals have demonstrated excellent hermetic property, potassium element is still a potential concern for electrode poisoning.

In order to overcome the disadvantages presented in the aforementioned sealing materials, investigation of alternative sealing concepts has been the focus for years. Ceramic felts [10,11], ceramic papers [12] and cast ceramic tapes [13] with materials that are chemically stable in SOFC environments were reported as sealing components for planar SOFCs. The density of ceramic felts is usually low; pre-densification is needed before their applications. The cast ceramic tape seals are flexible and compressible to some extent; gapless contact can be maintained under moderate compressive stresses.

In the present study, flow mechanics modeling and experimental verification are carried out with cast Al_2O_3 tape seals. The purpose is to develop a physical model to estimate the leak rate of cast ceramic tape seals; accordingly, the factors that control the leak rate can be identified and analyzed and a preferred sealing microstructure can be suggested for achieving improved sealing property.

2. Model analysis

2.1. List of symbols

a	length of ceramic tape seal
b	thickness of ceramic tape seal
D	diameter of capillary or dimension of flow
D_0	expectation value of capillary diameter distribution
D_l	diameter of large capillary
D_s	diameter of small capillary
g	gravitational acceleration
Kn	Knudsen number
L	sealing width
L_e	actual length of leak path
l_l	length of large capillary
l_s	length of small capillary
$\dot{M}$	mass flow rate
$\dot{M}_{VD}$	mass flow rate in a capillary with variable diameters
$\dot{M}_{D_s}$	mass leak rate in a capillary with a constant diameter D_s
N	total number of capillary in tape seal
P	pressure

* Corresponding author. Tel.: +86 27 87557694; fax: +86 27 87558142.
E-mail address: lijian@hust.edu.cn (L. Jian).

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P_i	pressure inside SOFC stack
P_o	pressure outside SOFC stack
Q	total flow rate across a capillary
R	radius of capillary
r	radial distance
T	temperature
t	time
V	gas velocity
V_s	slip boundary velocity
α	rarefaction coefficient
λ	gas molecular free path
μ	viscosity coefficient
ρ	gas density
σ	deviation of capillary diameter distribution
τ	tortuosity
ϕ	effective porosity of tape seal

Subscript

r	component in radial coordinate direction
θ	component in azimuth coordinate direction
x	component in x coordinate direction
y	component in y coordinate direction
z	component in z coordinate direction

2.2. Modeling

At room temperature, the cast ceramic tape seal contains organic binder and additives, such as dispersant, plasticizer and lubricant, and can be placed in contact with the sealing surfaces perfectly with the aid of a small amount of solvent. At operating temperatures, the added organic binder and additives are burnt out, leaving interconnected pores in the tape seal. Therefore, it is considered that the leak through sealing interfaces is manageable, and the infiltration leak through the seal body is the primary concern in this model. Because the microstructure is complicated with various sizes and shapes of interconnected pores, it is difficult to define the boundary conditions of the leaking gas flow. Usually, the gas flow in such a microstructure can be idealized as that in a closed pipe network or a regular array of solid particles; however, a viable calculation of the leak rate remains problematical [14,15]. The approach used in the current model is to simplify the porous microstructure to straight capillaries with specific size distributions. The leak rate is calculated based on the simplified capillary structure, and then is corrected according to the realistic microstructure.

Three basic assumptions are made in the model: (1) all interconnected pores in the ceramic seal are visualized as straight capillaries lying in the direction of gas leak flow, and the diameter of the capillaries is in the range of 0.01–200 μm according to experimental observations and experiences; (2) in a ceramic seal with a length a, a thickness b and a sealing width L as shown in Fig. 1, the capillary diameter D obeys the Gaussian distribution:

$$f(D) = \frac{1}{\sqrt{2\pi}\sigma} e^{-(D-D_0)^2/2\sigma^2} \tag{1}$$

and (3) the probability for a diameter smaller than 200 μm must be greater than 99.8%, that is

$$P\{D < 200\} = \Phi\left(\frac{200 - D_0}{\sigma}\right) > 0.998 \tag{2}$$

where Φ is the standard function of normal distribution.

2.3. Calculation of mass flow rate

H_2 is chosen as the gas of investigation. H_2 leaks through the capillaries under the condition that the pressure inside the SOFC stack P_i is higher than that outside P_o. Similar to the case of interface leak in mica-based seals [16], the flow characteristics of the high-temperature gas should be considered, as the gas flow is confined in such micro-channels. The average molecular free path of H_2 gas is 0.60 μm at 750 °C which is the operating temperature of the planar SOFC studied. The pressure-driven leak flow can be considered to be a continuum laminar flow in capillaries with diameters in the range of 60–200 μm; and the flow falls into the slip or transition flow regime if the diameter of the capillary is smaller than 60 μm, according to the value of the Knudsen number ($Kn = \lambda/D$, defined as the degree of gas rarefaction). In subsequent sections, these two situations are considered individually:

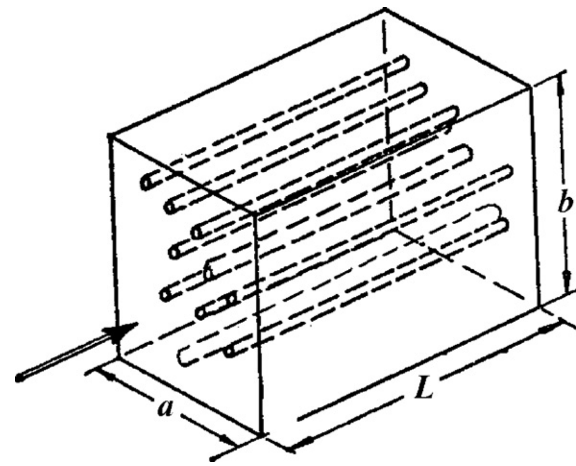

Fig. 1. Schematic drawing of the simplified porous microstructure in which capillaries with various diameters lying in leak flow direction represents interconnected pores. The diameter of the capillaries obeys the Gaussian distribution.

(1) Laminar flow in capillaries with $60 \le D \le 200$ μm.
Fig. 2 shows a capillary referenced in both cylindrical and Cartesian coordinates. The pressure-driven H_2 leak flow is regarded as a continuum laminar flow that can be described by the Navier–Stokes equations:

$$\begin{cases} \dfrac{\partial V_r}{\partial t} + (V \cdot \nabla)V_r - \dfrac{V_\theta{}^2}{r} = g_x - \dfrac{1}{\rho}\dfrac{\partial P}{\partial r} + \nu\left(\nabla^2 V_r - \dfrac{2}{r^2}\dfrac{\partial V_\theta}{\partial \theta} - \dfrac{V_r}{r^2}\right) \\ \dfrac{\partial V_\theta}{\partial t} + (V \cdot \nabla)V_\theta - \dfrac{V_r V_\theta}{r} = g_\theta - \dfrac{1}{\rho r}\dfrac{\partial P}{\partial \theta} + \nu\left(\nabla^2 V_\theta + \dfrac{2}{r^2}\dfrac{\partial V_r}{\partial \theta} - \dfrac{V_\theta}{r^2}\right) \\ \dfrac{\partial V_z}{\partial t} + (V \cdot \nabla)V_z = g_z - \dfrac{1}{\rho}\dfrac{\partial P}{\partial z} + \nu \nabla^2 V_z \end{cases} \tag{3}$$

where ν is defined as μ/ρ. Considering the leak flow of H_2 in a planar SOFC stack, it is further assumed that:
(i) the flow is isothermal and steady;
(ii) H_2 gas is incompressible, and the gravity effect of H_2 is ignored, $g_x = g_\theta = g_z = 0$;
(iii) the velocity slip is neglected and the flow velocity at the capillary wall is zero.

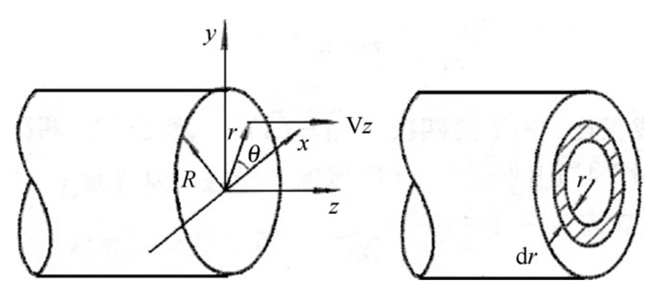

Fig. 2. Cylindrical and Cartesian coordinates showing the capillary model of the lamellar flow.