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Two-dimensional simulation of gas concentration impedance for a planar solid oxide fuel cell

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ABSTRACT

This paper presents a two-dimensional model for a planar solid oxide fuel cell (SOFC) anode in order to simulate the steady-state performance characteristics as well as the electrochemical impedance spectra. The developed model couples the mass transport with the electrochemical kinetics. The transient conservation equations (momentum and species equations) are solved numerically and the linear kinetic is used for the anode electrochemistry. In order to solve the system of the nonlinear equations, an in-house code based on the finite volume method is developed and utilized. A parametric study is also carried out and the results are discussed. Results show a capacitive semicircle in the Nyquist plot which is identical to the gas concentration impedance. The simulation results are in good agreement with published data.

1. Introduction

Fuel cell is an electrochemical device that converts stored chemical energy of fuel into the electrical energy.

Solid oxide fuel cells (SOFCs) are the safest type of high temperature fuel cells that generates electricity. In order to improve the efficiency of the fuel cells, better understanding of the electrochemical reactions and mass transport in the fuel cell is essential. Therefore, it is useful to numerically investigate the SOFC performance. Various studies with different levels of sophistication on SOFC modeling have been published [1–6].

In addition to study the steady state behavior and dynamic performance of fuel cells, impedance response modeling appears as a useful tool for both analyzing the experimental results of electrochemical impedance spectroscopy (EIS) and study the fuel cells performance. EIS is a powerful technique to describe electrochemical systems [7]. This method plays an important role in the SOFC studies.

Hu et al. [8] presented a steady state, two-dimensional model for mass transport and polarization phenomena of polybenzimidazole fuel cells based on AC impedance technique. Hassel et al. presented the theoretical viewpoint of the impedance model in order to investigate the polarization effects on the electrode processes [9,10]. Adler et al. presented a polarization impedance model for a composite cathode [11]. In this study the total impedance of cell was expressed by tortuosity, porosity, interface area, oxygen self-diffusion coefficient and oxygen coefficient. Wagner et al. measured the electrochemical impedance spectrum of the SOFC and the PEMFC [12]. Primdahl and Mogensen used the EIS method to study the SOFC anodes [13]. Bieberle and Gauckler investigated the reaction mechanism inside the anode experimentally and theoretically [14,15]. They presented a six-step mechanism for the electrochemical oxidation reaction of hydrogen. Takano et al. studied the impedance corresponding to the mass transfer or the gas diffusion impedance using a 1D model [16]. Jasinski et al. studied the mechanism of a single- chamber SOFC by impedance spectroscopy method [17]. Lang et al. used the EIS method to measure the different resistances that affects the area specific resistance (ASR) [18]. Bessler presented a new computational method for the impedance simulations of SOFCs [19]. In two other studies, Bessler applied his previous proposed model for two different SOFC geometries using a 1D model [20,21].

Based on the mentioned researches, the experimental impedance spectra of the SOFC anode show two dominant features: a high-frequency process (1–10 kHz) associated with electrochemistry and a low frequency process (~10 Hz) associated with gas transport processes in the anode channels [12,13]. This feature results in a capacitive semicircle in Nyquist plot which is called 'concentration impedance' [20,21]. The present study investigates the SOFC anode impedance by analyzing the latter feature. Most of the mentioned studies on impedance modeling of SOFC anode,

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Nomenclature

C_g	gas concentration capacitance (F)
D_{ij}	binary diffusivity ($m^2\ s^{-1}$)
F	Faraday's constant (96845 C mol^{-1})
f	Frequency (Hz)
f_g	relaxation frequency (Hz)
H_{ch}	channel height (m)
i	current density ($A\ m^{-2}$)
j	mass diffusion flux ($mol\ m^{-2}\ s^{-1}$)
L	cell length (m)
M	molar mass ($kg\ mol^{-1}$)
$\dot{m}$	mass flux ($kg\ s^{-1}$)
P	pressure (Pa)
R	universal gas constant ($J\ mol^{-1}\ K^{-1}$)
R_g	gas concentration resistance ($\Omega\ cm^2$)
R_{ct}	electrode charge- transfer resistance ($\Omega\ cm^2$)
T	temperature (K)
W	unit cell width (m)
W_{ch}	channel width (m)
W_{rib}	rib width (m)
t	time (s)
u	velocity in x direction (m/s)
v	velocity in y direction (m/s)
y_i	mass fraction of specie i
Y	complex admittance (Ω^{-1})
Y'	real part of admittance (Ω^{-1})
Y''	imaginary part of admittance (Ω^{-1})
Z	complex Impedance (Ω)

Greek symbols

ρ	gas density ($kg\ m^{-3}$)
η_{act}	activation overvoltage (V)
η_{anode}	anode overvoltage (V)
η_{conc}	concentration overvoltage (V)
η_{steady}	steady state overvoltage (V)
$\eta_{excitation}$	excitation overvoltage (V)
μ	viscosity (Pa s)
τ	time period (s)

considered lumped assumptions [12,13] or assumed 1D uniform flow in the anode channel [16,19,20] to avoid calculation complexity. However, concentration losses may take place at different directions in the fuel channel. Therefore, two-dimensional modeling of gas flow in the anode channel is essential in order to obtain a more accurate concentration impedance model. The purpose of the current study is then to develop a two-dimensional transient model in order to simulate the impedance spectra. This model is an extension of a 1D impedance model that was previously developed by the authors [22] to investigate the effects of gas flow in two dimensions on the impedance spectra. Furthermore, the velocity, concentration and partial pressure of the gas flow and the produced current density of the anode are presented and analyzed. A parametric study is also carried out using the simulation tool developed and the results are discussed.

2. Modeling and simulation approach

Fig. 1 depicts the schematic diagram of a planar SOFC and the computational domain. As shown, the hydrogen fuel passes through the anode channel.

In this study the electrolyte and cathode overvoltages are ignored while the SOFC anode overvoltage (η_{anode}) is analyzed. In addition, the anode ohmic overvoltage is neglected due to the high anode electronic conductivity [23–25]. Furthermore, concentration overvoltage within the porous anode is also ignored, since in thin anodes ($\leqslant$30 μm) concentration overpotentials due to porous transport are negligible [26]. Tables 1 and 2 show the cell geometry, as well as the assumed SOFC operational conditions.

2.1. Electrochemical model

The overall SOFC reaction and the electrochemical oxidation reaction at the anode for hydrogen fuel cell are as follows, respectively:

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \tag{1}$$

$$H_2 + O^{2-} \rightarrow H_2O \tag{2}$$

The anode overall loss is given by:

$$\eta_{anode} = \eta_{act} + \eta_{conc} \tag{3}$$

where the anodic concentration overvoltage is [22]:

$$\eta_{conc,a} = \frac{RT}{2F}\ln\left(\frac{P^{in}_{H_2}P^{ra}_{H_2O}}{P^{in}_{H_2O}P^{ra}_{H_2}}\right) \tag{4}$$

where P^{ra} refers to the reactant and product partial pressure at the reaction sites. The activation overvoltage is determined from the linear equation as follows [28]:

$$i(x) = \frac{\eta_{act}}{R_{ct}} \tag{5}$$

where $i(x)$ is current density and R_{ct} is the electrode charge-transfer resistance. The electrode charge-transfer resistance at open-circuit conditions and 97% inlet H_2 is equal to 0.1 $\Omega\ cm^2$ as observed in experiments [13].

2.2. Mass transfer model

The transient momentum and mass transfer governing equations inside the 2-D anode channel are used as follows, respectively:

$$\frac{\partial\rho}{\partial t} + \frac{\partial(\rho u)}{\partial x} + \frac{\partial(\rho v)}{\partial y} = 0 \tag{6}$$

$$\frac{\partial\rho u}{\partial t} + \frac{\partial\rho uu}{\partial x} + \frac{\partial\rho uv}{\partial y} = -\frac{\partial P}{\partial x} + \frac{4}{3}\frac{\partial}{\partial x}\left(\mu\frac{\partial u}{\partial x}\right) + \frac{\partial}{\partial y}\left(\mu\frac{\partial u}{\partial y}\right) + \frac{\partial}{\partial y}\left(\mu\frac{\partial v}{\partial x}\right) - \frac{2}{3}\frac{\partial}{\partial x}\left(\mu\frac{\partial v}{\partial y}\right) \tag{7}$$

$$\frac{\partial\rho v}{\partial t} + \frac{\partial\rho uv}{\partial x} + \frac{\partial\rho vv}{\partial y} = -\frac{\partial P}{\partial y} + \frac{\partial}{\partial x}\left(\mu\frac{\partial v}{\partial x}\right) + \frac{4}{3}\frac{\partial}{\partial y}\left(\mu\frac{\partial v}{\partial y}\right) + \frac{\partial}{\partial x}\left(\mu\frac{\partial u}{\partial y}\right) - \frac{2}{3}\frac{\partial}{\partial y}\left(\mu\frac{\partial u}{\partial x}\right) \tag{8}$$

$$\frac{\partial(\rho y_i)}{\partial t} + \frac{\partial(\rho u y_i)}{\partial x} + \frac{\partial(\rho v y_i)}{\partial y} = \nabla\cdot(\vec{j_i}) \tag{9}$$

where ρ is flow density, y_i is the species mass fraction, μ is dynamic viscosity which is obtained by Reichenberg formulation [29], and $\vec{j_i}$ represents mass diffusion flux that is modeled by Fick's law. The binary diffusivity is determined by Fuller et al. relation [29]. Due