



Numerical analysis of electrical power generation and internal reforming characteristics in seal-less disk-type solid oxide fuel cells

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ARTICLE INFO

Article history:

Received 4 September 2008
Received in revised form 22 October 2008
Accepted 24 October 2008
Available online 8 November 2008

Keywords:

Solid oxide fuel cell
Simulation
Internal reforming
Diffusion
Seal-less
Concentration profile

ABSTRACT

For seal-less type solid oxide fuel cells, its power generation characteristics and distribution of the gas composition depend on not only the electrochemical reaction, but also complex kinetics and transport phenomena, because the internal reforming reaction and the water-gas shift reaction take place together with reverse diffusion of the ambient gas from the surroundings of the cell. The purpose of this paper is to theoretically explain the experimental results of the anodic concentration profile of gaseous species previously reported in a practical seal-less disk-type cell which used pre-reforming methane with steam as a fuel. A numerical model that takes into account the transport phenomena of the gaseous species and the internal reforming reaction with the water-gas shift reaction together with the assumption of the cell outlet boundary condition was constructed to numerically analyse the gas composition distribution and power generation characteristics. Numerical analyses by the model were conducted for the several cases reported as the experiment. The calculated results in the anode gas concentration profile and in the voltage–current characteristics show good agreement with the experimental data in every case, and then the validity of the simulation model was verified. Therefore, the model is useful for a seal-less disk-type cell which is operated by a fuel including non-reformed methane.

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1. Introduction

Solid oxide fuel cells (SOFCs) have attracted much attention due to their high efficiency and cleanliness, and they are expected to be one of the most favorable electrical power generation technologies in the near future. Recently, SOFC systems driven by town gas have been intensively developed by several companies using seal-less cell structure, where the outlet edges of the gas channels of the anode and cathode are open to the ambient, owing to its advantageous features in gas sealing and thermal stress problems [1–3].

In the SOFC systems, an internal reforming, i.e., reforming of the fuel in the cell, is often employed in conjunction with pre-reforming due to its high operating temperature. Generally, the power generation characteristics of cells depend on the gas composition on the anode side and cathode side of the cells. In the seal-less type

cells, the internal reforming reaction and the water-gas shift reaction take place together with reverse diffusion of the ambient gas from the surroundings of the cell, i.e., diffusion opposite to gas flow direction. Therefore, the distribution of the gas composition in the cell and power generation characteristics depends on not only the electrochemical reaction, but also complex kinetics. Many simulation studies have been conducted to aid in development of SOFCs [4–19].

To the best of our knowledge, however, very few numerical results have been reported on the power generation characteristics compared to the measurement of the gas composition distributions because the concentration profile of the gaseous species in a cell has been seldom measured. For the seal-less disk-type cell using pure hydrogen as a fuel, it has been reported by Momma et al. [17] that the influence of the reverse diffusion of the ambient gas is clarified based on both the experimental and theoretical analyses, the results of which have shown good agreement. On the other hand, there has never been such practically useful report on the cell using reformed gas as the fuel.

In one of the recent papers dealing with seal-less disk-type SOFCs, Momma et al. [20], reported the experimental results for the concentration profile of gaseous species along the flow direction in the anode gas channel of the single cells which were performed

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Nomenclature

c	molar density (mol m^{-3})
c_{CO}	CO concentration in the mixture gas (mol m^{-3})
$c_{\text{H}_2\text{O}}$	H ₂ O concentration in the mixture gas (mol m^{-3})
\dot{c}_j	mole reaction quantity due to cell reaction for each unit time and unit volume ($\text{mol s}^{-1} \text{m}^{-3}$)
\dot{c}_{ref}	mole reaction quantity due to reforming reaction for each unit time and unit volume ($\text{mol s}^{-1} \text{m}^{-3}$)
\dot{c}_{sf}	mole reaction quantity due to water-gas shift reaction for each unit time and unit volume ($\text{mol s}^{-1} \text{m}^{-3}$)
D_{AB}	mutual diffusion coefficient of species A and B ($\text{m}^2 \text{s}^{-1}$)
d_{an}	thickness of anode current collector
d_{ca}	thickness of cathode current collector
D_e	effective diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)
$D_{i,m}$	diffusion coefficient of species i in multi-component mixture gas ($\text{m}^2 \text{s}^{-1}$)
D_{1j}	diffusion coefficient of the pair of $1-j$ species in a binary mixture ($\text{m}^2 \text{s}^{-1}$)
D_{1m}	diffusion coefficient when element 1 diffuses in the mixture gas m
Ea_r	activation energy (J mol^{-1})
ΔEa_{ref}	activation energy ($=57,840 \text{ J mol}^{-1}$)
ΔEa_{sf}	activation energy ($=95,000 \text{ J mol}^{-1}$)
$\Delta G_{\text{H}_2\text{O}}^\circ$	Gibbs free energy change in reaction $\text{H}_2 + \text{O}_2 = \text{H}_2\text{O}$ at the standard pressure (0.1 MPa) and reference state (J mol^{-1})
F	Faraday constant (C mol^{-1})
J_e	electronic current density which flows in electrolyte (A m^{-2})
J_i	ionic current density which flows in electrolyte (A m^{-2})
J_o	current density which flows to an external circuit (A m^{-2})
k	Boltzmann constant ($=1.3807 \times 10^{-23} \text{ J K}^{-1}$)
K_{sf}	equilibrium constant of water-gas shift reaction
M_i	molecular weight of species i (kg mol^{-1})
P	pressure (Pa)
p_{CH_4}	partial pressure of methane in the mixture gas
p_i	partial pressure of chemical species i
R	gas constant ($=8.314 \text{ J mol}^{-1} \text{ K}^{-1}$)
r_{cell}	radius of the cell
R_{cell}	cell resistance ($\Omega \text{ m}^2$)
R_{cell_0}	cell resistance at 1073 K ($\Omega \text{ m}^2$)
R_e	ohmic resistance of electrolyte by electronic conduction ($\Omega \text{ m}^2$)
R_i	ohmic resistance of electrolyte by ionic conduction ($\Omega \text{ m}^2$)
R_r	reactive resistance of the electrodes ($\Omega \text{ m}^2$)
R_s	series resistance of the electrodes ($\Omega \text{ m}^2$)
r_{CH_4}	rate of the reforming reaction ($\text{mol s}^{-1} \text{ kg}_{\text{cat}}^{-1}$)
r_{CO}	rate of the water-gas shift reaction ($\text{mol s}^{-1} \text{ kg}_{\text{cat}}^{-1}$)
T	temperature (K)
T_{cell}	temperature of the cell
T_{ref}	outlet gas temperature of pre-reformer (K)
V_{cell}	cell operating voltage (V)
u	mass average velocity (m s^{-1})
U_f	fuel utilization

Greek letters

α_{ref}	utilization factor by which the catalyst in the anode current collector actually contributes to the reforming reaction
α_{sf}	utilization factor by which the catalyst in the anode current collector actually contributes to the water-gas shift reaction
ε_{an}	porosity of the anode current collector
ε_{ca}	porosity of the cathode current collector
ε_{po}	porosity
σ	characteristic diameter (\AA)
ρ	total mass density (kg m^{-3})
ρ_{anc}	density of anode current collector (kg m^{-3})
$\rho_{\text{cat,ref}}$	density of catalysts which actually contribute to the reforming reactions (kg m^{-3})
$\rho_{\text{cat,sf}}$	density of catalysts which actually contribute to the water-gas shift reactions (kg m^{-3})
τ	twist factor
χ	molar fraction
Ω	collision integration
ω_i	mass fraction of species i

Subscripts

an	fuel (anode gas)
ca	air (cathode gas).

by partially reformed methane as the fuel. From the compositional analysis of the anode gas, their experimental results indicated that the molar ratio of the hydrogen and carbon elements, i.e., H/C, is not a constant value between the inlet and outlet of the anode gas channel, and that nitrogen, which is not surely present in the inlet, is significantly detected near the cell edge. The following conclusions are suggested: (1) there are different diffusion velocities, or different species velocities, between each chemical species that composes the anode gases, and (2) reverse diffusion of ambient air from the periphery of the cell takes place.

In this paper, to theoretically clarify these phenomena, a numerical model that takes the convection of the gas and diffusion into account was constructed for a seal-less disk-type SOFC which is operated by a fuel including non-reformed methane. Therefore, we compared the numerical results with the experimental data such as the concentration distributions of the gaseous species and voltage–current characteristics using the calculation corresponding to the experimental conditions, and then examined these phenomena. This model can explain the influence of an internal reforming reaction and a water-gas shift reaction in the anode gas flow together with the reverse diffusion from the periphery of the cell. Furthermore, this model suggests the distributions of an electromotive force and a current because it deals with the distributions of the concentration profile of gaseous species along the flow direction, thereby the voltage–current characteristics are predicted.

2. Modeling

2.1. Simulation model and assumption

Fig. 1 shows a schematic diagram of a seal-less disk-type SOFC single cell as a simulation model used to explain the data obtained from the experiment of Momma et al. [20]. The model consists of a composite disk made of a solid electrolyte with electrodes on both sides, and current collectors and metal separators, each for the positive and negative electrodes.

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