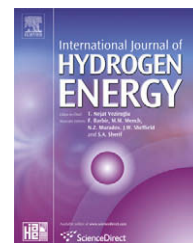


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Performance and effective kinetic models of methane steam reforming over Ni/YSZ anode of planar SOFC

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ABSTRACT

Under mass/heat transfer and species diffusion limitations, the detailed thermodynamic investigations and experimental measurements were carried out to study and evaluate the performance of methane steam reforming and to develop its effective kinetic models over Ni/YSZ anode of the planar solid oxide fuel cell developed by Central Research Institute of Electric Power Industry (CRIEPI) of Japan. Based on the kinetic models of methane steam reforming developed by Xu and Froment, the effective kinetic models were developed. Combining the experiment data with the iteration calculation, the effective factor (C_e) is found to be 5.0×10^{-4} . Using these effective kinetic models, the detailed distributions of temperature, species, methane conversion, and carbon formation and gasification activity are presented under wide range of operating conditions. The results show that the local temperature in the reactor and the methane conversion are sensitive to the operating parameters. The region of the lowest local temperature and the most possible region of carbon formation are at the front of the porous anode sample. Due to the limitation of contact time, heat transfer and species diffusion, the methane conversion appreciably decreases with an increase of S/C ratio.

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1. Introduction

Solid oxide fuel cell (SOFC) is a promising energy conversion unit that produces electric power and heat with higher efficiency, lower noise and pollutant emission than conventional heat engine, steam and gas turbines, and combined cycle [1,2]. Due to its high operating temperature (700 °C–1000 °C), SOFC offers the widest potential range of applications, the flexibility of fuel choices and the possibility for operation with an internal reforming [3–5]. The uses of various alternative fuels,

i.e. methane, methanol, ethanol, gasoline and other oil derivatives, in SOFCs have been widely investigated [6]. To date, methane is a promising fuel for SOFC system as it is an abundant component in natural gas (80–95% CH₄) [7]. The H₂/CO-rich gas, eventually used to generate the electrical energy and heat in the SOFC system, can be produced by catalytic heated steam reforming (HSR) or partial oxidation (POX) [8]. Nevertheless, the steam reforming is widely used in the industrial. Its main advantage compared to other fuel processing technologies is the high thermal efficiency.

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Nomenclature			
$a_{c,i}$	carbon activity	R	universal gas constant, $\text{J kg}^{-1} \text{K}^{-1}$
A_i	pre-exponential factor	r_i	reaction rate, $\text{kmol h}^{-1} \text{kg cat}^{-1}$
C_e	effective factor	S_ϕ	source or sink
$D_{i,e}$	overall effective diffusion coefficient of species i, $\text{m}^2 \text{s}^{-1}$	T	temperature K
D_{ij}	binary mass diffusion coefficient of a mixture species i and j, $\text{m}^2 \text{s}^{-1}$	\bar{U}	velocity, m s^{-1}
$D_{i,m}$	mass diffusion coefficient for species i in the mixture, $\text{m}^2 \text{s}^{-1}$	X_i	molar fraction of species i, kmol kmol^{-1}
D_K	Knudsen diffusion, $\text{m}^2 \text{s}^{-1}$	Y	mass fraction of species, kg kg^{-1}
d_p	pore diameter in porous medium, m	Greek symbols	
$E_{a,i}$	activation energy, J kmol^{-1}	k	turbulent kinetic energy, $\text{m}^2 \text{s}^{-2}$
$G_{i=1-5}$	flow rate of dry methane, ml min^{-1}	ε	turbulent dissipation rate, $\text{m}^2 \text{s}^{-3}$
H	total enthalpy, J kg^{-1}	ρ	density, kg m^{-3}
$\Delta H_{298,15}^0$	heat of reaction, J kmol^{-1}	λ	thermal conductivity, $\text{W m}^{-1} \text{K}^{-1}$
ΔH_i^0	heat of chemisorption, J kmol^{-1}	λ_f	mean free path length, m
$K_{eq,i}$	equilibrium constant	ν	special Fuller et al. diffusion volume
k_i	reaction rate constant	ε_p	porosity of porous medium
K_i	Adsorption constant of component i	τ_p	tortuosity of porous medium
K_n	Knudsen number $K_n = \lambda_f/d_p$	κ	permeability of porous medium
M	molecular weight, kg kmol^{-1}	ϕ_i	pore diameter, μm
P_i	partial pressure of component i, kPa	Γ_ϕ	diffusion coefficient, $\text{kg m}^{-1} \text{s}^{-1}$
		Subscripts	
		g	gas
		s	porous solid

The methane steam reforming is a quite complex process. It not only involves the transfer and diffusion of reactants and products between the bulk flow and the catalyst surface as well as within the catalyst, but also involves several reactions simultaneously in parallel or in series [9]. Therefore, there are several major problems remain to be solved before such SOFCs can be routinely operated on the direct feed of alternative fuels other than hydrogen. One of them is the carbon deposition on the anode, causing loss of active site and cell performance as well as poor durability. The growth of carbon filaments attached to anode crystallites can generate massive forces within the electrode structure leading to its rapid breakdown [10]. A number of efforts have been carried out to alleviate this problem. Usually, the approach to avoid carbon deposition is the addition of extra steam to the feed. High steam/carbon (S/C) ratios can be used to effectively suppress carbon formation. However, the use of high S/C ratio is unattractive as pressurizing and heating a large excess of water is expensive, and it lowers the electrical efficiency of SOFC by steam dilution of fuel. In addition, increasing the operating temperature also suppresses the carbon formation but it is restricted by the maximum temperature due to the thermal durability of the SOFC cell components [11]. Another of them is the internal stress in cell components arising from the non-homogeneous temperature distributions. The direct internal steam reforming causes the steep change of the local temperature on the anode due to providing large heat for the endothermic reaction. This not only causes larger internal stress and enhances the difficult of sealing, but also increases the possibility of carbon formation.

A considerable effort has been put into investigations of the kinetic models of methane steam reforming since the early 1950s [9,12,13]. Many studies of the kinetic models of methane steam reaction were carried out with the catalysts of

different compositions prepared by various methods and of different particle size, and over wide ranges of temperature and pressure. Therefore, there are many different mechanisms and kinetic models suggested. However, it impossible to develop generalized kinetic models, which can be applied to different catalyst with only a change in parameter to suit each catalyst. The main reasons are two-fold: (1) the change of catalyst composition changes not only the values of the parameters of the kinetic model, but it also changes the structure of the kinetic model via change in the mechanism [9]; (2) the most mechanism and the kinetic models for each type of steam reforming catalyst are obtained experimentally under no mass/heat transfer and species diffusion limitations. Whereas, in SOFC cell, mass/heat transfer and species diffusion in porous anode are significantly limited and affected by the operating conditions and anode properties, such as porosity, pore-size, tortuosity, permeability, thermal conductivity, etc. Furthermore, due to the limited contact surface area of porous medium, it is impossible that all catalyst in the porous anode contacts with gas molecules. Hence, it is necessary to study the effective kinetic models of steam reforming reaction for porous anode.

The objective of this work is to study and evaluate the performance of methane steam reforming and to develop its effective kinetic models over Ni/YSZ porous anode of the planar SOFC developed by CRIEPI of Japan, under heat/mass transfer and species diffusion limitation by the experiments and numerical thermodynamic investigation. Based on commercial CFD software (CFX4.4) and a routine developed by ourselves using the effective kinetic models of methane steam reforming, a fully three-dimensional mathematical model has been constructed to simulate the flow, heat and mass transfer, and steam reforming process in the experimental equipment. Within wide ranges of temperature, pressure and S/C ratio, the

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