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Modeling and simulation of microwave double absorption on methanol steam reforming for hydrogen production

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ABSTRACT

Methanol steam reforming (MSR) accompanied by methanol decomposition (MD) in an environment with microwave heating is modeled and simulated numerically in the present study. In the developed method, the governing equations simultaneously consider the continuity, momentum, energy, species and Maxwell's equations. Meanwhile, the double absorption of microwaves by both the reactants and the catalyst bed in the reactor is also taken into account. The heating processes of microwaves on the reactants and the catalyst bed are described by establishing two sets of complex relative permittivity in the non-porous and porous region. The permittivity consists of a dielectric constant and a dielectric loss factor. The obtained results indicate that the developed model can predict MSR and MD accurately. With microwave irradiation, it is found that varying dielectric loss factor in the non-porous region has a significant impact on hydrogen production, revealing that the preheating of the reactants plays a prominent role in determining the performance of MSR. Regarding microwave power, it is observed that increasing power intensifies H₂ yield, especially at 500 W where methanol is depleted completely. However, an increase in power also makes MD tend to grow in that the CO₂ selectivity decreases.

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1. Introduction

Fuel cells are likely to become the important devices of the future to directly convert chemical energy of fuel into electric energy because the devices are characterized by energy conversion with high efficiency [1]. On the other hand, when low-temperature fuel cells, such as proton exchange membrane fuel cells (PEMFCs), are employed for power and electricity generation, hydrogen has to be fed into the fuel cells as the fuel [2]. In addition to high efficiency, hydrogen-fed fuel cells also possess the merit of low emission in that water is the only byproduct and, unlike internal combustion engines, no other air pollutants are liberated. Before PEMFCs can be extensively used as power generators, hydrogen production becomes an essential and vital task.

Currently, hydrogen is mainly produced from natural gas or methane by means of the thermochemical procedure of steam reforming (SR) [3,4], namely, steam methane reforming (SMR). This arises from the facts that natural gas is a commonly and extensively used fossil fuel and the infrastructure of natural gas is available. Instead of the use of methane, methanol has been considered as a potential feedstock for hydrogen production, especially for hydrogen production on board used in mobile fuel cells [5]. The primary advantages using methanol as the feedstock are: (1) methanol is a liquid fuel so the energy density per unit volume of fuel is high; (2) it is easier to store and deliver methanol compared to gaseous methane from the transport point of view; (3) methanol is featured by its good miscibility with water; and (4) unlike SMR which is usually carried out at high temperature

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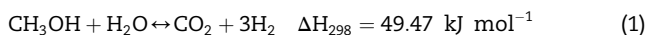
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Nomenclature			
c	molar concentration, mol m ⁻³	w	velocity, m s ⁻¹
c_p	gas mixture specific heat, J kg ⁻¹ K ⁻¹	X_i	molar fraction of species i , dimensionless
D	diffusion coefficient, m ² s ⁻¹	Greek letters	
D_p	mean particle diameter, m	γ	porosity, dimensionless
E	electric field intensity, V m ⁻¹	ϵ_0	Free space permittivity, 8.854 × 10 ⁻¹² F m ⁻¹
f	frequency, Hz	ϵ'	dielectric constant (F m ⁻¹)
H	magnetic field intensity, A m ⁻¹	ϵ''	dielectric loss factor, F m ⁻¹
k_{dc}	rate constant of decomposition reaction, mol kg ⁻¹ s ⁻¹	ϵ_r	complex relative permittivity, dimensionless
k_{eff}	effective thermal conductivity, W m ⁻¹ K ⁻¹	ϵ'_r	relative dielectric constant, dimensionless
k_f	fluid phase thermal conductivity, W m ⁻¹ K ⁻¹	ϵ''_r	relative dielectric loss factor, dimensionless
k_s	solid medium thermal conductivity, W m ⁻¹ K ⁻¹	μ	viscosity, Pa s
k_{sr}	rate constant of reforming reaction, m ³ kg ⁻¹ s ⁻¹	μ_0	free space permeability, 4π × 10 ⁻⁷ T m A ⁻¹
K	catalyst-layer permeability, m ⁻²	μ_r	Relative permeability, dimensionless
M_i	molar mass of species i , kg mol ⁻¹	ρ	gas mixture density, kg m ⁻³
N	number of species	ρ_s	Catalyst density, kg m ⁻³
p	pressure, Pa	ϕ	steam-methanol molar ratio, dimensionless
p_{atm}	atmospheric pressure (1.013 × 10 ⁵ Pa)	ω	angular frequency, Rad s ⁻¹
Q_{mw}	energy source term due to microwave heating, J m ⁻³	Subscript	
$Q_{reaction}$	energy source term due to chemical reaction, J m ⁻³	CH ₃ OH	methanol
R	universal gas constant, 8.314 m ³ Pa K ⁻¹ mol ⁻¹	CO	carbon monoxide
R_i	reaction rate of species i , mol m ⁻³ s ⁻¹	CO ₂	carbon dioxide
R_{dc}	reaction rate of decomposition reaction, mol m ⁻³ s ⁻¹	f	fluid
R_{sr}	reaction rate of steam reforming reaction, mol m ⁻³ s ⁻¹	H ₂ O	water
T	temperature, K	i	species i
$\tan\delta$	loss tangent, dimensionless	in	inlet
V	velocity, m s ⁻¹	N ₂	nitrogen
V_s	seepage velocity, m s ⁻¹	out	outlet
		sr	steam reforming
		dc	decomposition
		steam	steam
		w	wall

(750–1000 °C) and high pressure (30–40 bar) environments [6], MSR is usually carried out at atmospheric pressure and temperatures ranging from 200 to 300 °C [7]. In consequence, MSR is a simple and efficient way of producing hydrogen.

Methanol steam reforming (MSR) is expressed as the following



From the preceding equation, it is clear that MSR inherently is an endothermic reaction. Therefore, when hydrogen is produced from water and methanol via SR, heat must be supplied from the surrounding to activate the reaction. Reviewing past studies of triggering chemical reactions, a number of heating routes, such as conventional electric heating [8,9], plasmas [10–12], microwaves [7,13] and heat recovery [14], with the aid of catalysts have been employed to drive hydrogen production.

When one is concerned with microwave heating, basically, the heating mechanisms are different from those of conventional heating. Specifically, in conventional heating processes, thermal energy is delivered into materials by means of conduction, convection and radiation in the presence of thermal gradient and temperature difference [15]. However, in microwave-heating processes, when materials

containing dielectrics are exposed to the environment with microwave irradiation, microwave energy will be transferred into the materials through molecular interactions in the electromagnetic field [16]. That is to say, electromagnetic energy is converted into thermal energy through dielectric heating [17,18]. On account of direct transport and conversion of energy, the microwave heating has been regarded as an important tool to rapidly and effectively activate chemical reactions. Apart from minimizing heating time, the microwave heating also possesses the merits of providing uniform temperature distribution, high energy efficiency and improvements in product quality for various industrial applications [19].

To date, two different methods have been developed to describe microwave-heating processes; they are Lambert's law and Maxwell's equations [19,20]. Lambert's law describes microwave power reduction as a function of distance when microwaves travel into a sample [19,21]. It has been stated that Lambert's law is valid only for large sample dimensions and high loss dielectric materials [22,23]. In contrast, for small or low dielectric samples, the spatial variations of electromagnetic fields and microwave power absorbed within samples must be required by a complete solution of Maxwell's equations [19,20].

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