Comparisons between methane and methanol steam reforming in thermally integrated microchannel reactors for hydrogen production: A computational fluid dynamics study

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ABSTRACT

This paper addresses the issues related to the design and operation of steam reforming combined with catalytic combustion in thermally integrated microchannel reactors for hydrogen production. Comparisons were made between methanol and methane steam reforming, representing a low and a high temperature process respectively, under the same operating conditions to determine whether methanol-based thermally integrated systems can be more energy-efficient than methane-based ones. Computational fluid dynamics simulations were performed to gain insight into the reactor performance and thermal behavior. The effect of various design parameters was investigated to identify suitable ranges of operating conditions, and an analysis of heat and mass transfer was performed to design a highly efficient system. It was shown that steam reforming of both fuels is feasible in millisecond reactors under a variety of conditions, but very careful design is necessary. Methanol reforming can be more efficient, offering a better solution not only to simplify design but also to improve power and efficiency. The wall thermal conductivity is essential to the design and optimization of these systems, as it can significantly affect the overall energy balance. There is no significant difference in reactor performance between different channel heights at the same flow rate. The ratio of the flow rates on opposite sides of the reactor is an important design parameter and must be carefully adjusted to improve efficiency and eliminate hot spots. Finally, a simple operating strategy was proposed to achieve variable power output, and design recommendations were made.

Introduction

In recent years, there has been an increasing interest in the development of portable fuel processing systems for hydrogen production [1—4]. Since conventional systems are often limited by physical transport effects, microreactor technology has the potential to greatly improve overall performance for fuel cell applications [4—12]. The major techniques used to produce hydrogen from hydrocarbon fuels are steam

Nomenclature

A	surface area
$C_{fuel,in}$	initial concentration of the fuel
c_p	specific heat capacity at constant pressure
d	gap distance between the plates
d_{pore}	mean pore diameter
D	diffusivity
D_{eff}	effective diffusivity
D_m	mixture-averaged diffusivity
D^T	thermal diffusivity
F	catalyst/geometric surface area
$F_{s-\infty}$	view factor for solid-ambient
h	specific enthalpy
h_o	overall heat transfer coefficient
$\Delta_c H_m^\theta(T)$	standard enthalpy of combustion at the specified temperature
$\Delta_r H_m^\theta(T)$	standard enthalpy of reaction at the specified temperature
K	number of species
l	length
m	total number of species
p	pressure
q	heat flux
$\dot{Q}$	molar flow rate
R	ideal gas constant
$\dot{s}$	rate of appearance of a heterogeneous product
S_{BET}	Brunauer-Emmett-Teller surface area
S/V	surface-to-volume ratio
T, T_o	absolute temperature, reference temperature
$T_{w,o}$	temperature at the external surface of the solid wall
u, v	streamwise velocity component, transverse velocity component
V_{pore}	catalyst pore volume
$V, \vec{V}$	diffusion velocity, diffusion velocity vector
$W, \overline{W}$	relative molecular mass, relative molecular mass of the gas mixture
x, y	streamwise coordinate, transverse coordinate
Y	mass fraction

Greek variables

Γ	surface site density
δ	thickness
ε	emissivity
ε_m	porosity
$\varepsilon_{s-\infty}$	effective emissivity for solid-ambient
η	thermal efficiency
$\eta_{reforming}$	thermal efficiency of the reforming process
λ	thermal conductivity
λ_m	porous medium effective thermal conductivity
λ_{eff}	effective thermal conductivity
μ	dynamic viscosity
ν	stoichiometric coefficient
ρ	density
σ	Stefan-Boltzmann constant
σ_m	site occupancy of the m-th surface species
τ	time-scale
τ_m	tortuosity factor
$\dot{\omega}$	rate of appearance of a homogeneous product

Subscripts

$(+)$	just above the interface
$(-)$	just below the interface
amb	ambient
c	catalyst
g	gas
in	inlet
i, j, k, m	species index
o	outer
rad	radiation
s	solid
w	wall
x, y	streamwise component, transverse component

reforming, partial oxidation, and autothermal reforming [13−16]. Steam reforming is typically the preferred process for hydrogen production in industry [3]. Due to the importance of this reaction, substantial efforts are being made to ensure stable and efficient operation of portable fuel processing systems [17−24].

Unfortunately, there exist a number of key challenges for the realization of these systems [5,25]. The management of heat within a highly compact device is generally regarded as the most crucial challenge [26,27]. Thermal coupling of exothermic and endothermic reactions is needed to reduce heat losses and improve thermal efficiency. Additionally, mass-transport effects are usually negligible in these systems and thus intrinsic kinetics tend to dominate, but in reality this is not always the case [5,28]. Furthermore, the overall performance of these portable fuel processing systems is highly dependent on the reactor dimensions [29,30] and operating conditions [18,31,32]. In particular, the dependence of kinetics with temperature tends to be the dominant factor in design and operation, and must be clearly established. It is therefore of great significance to clarify the role of operating temperature in these portable fuel processing systems.

The steam reforming along with water-gas shift reaction is kinetically limited at low temperatures but thermodynamically limited at high temperatures [33,34]. From the point of view of reduced carbon monoxide content and increased hydrogen production, it is desirable to conduct the reforming process at low temperatures [33]. However, to achieve the necessary reaction rates, higher temperatures are often required [34]. Unfortunately, higher-temperature processes require more insulation and thermal integration. An ideal condition would be the use of a catalyst active enough to operate at low temperatures where equilibrium is very favorable [33,34].

The operating temperature during the process of steam reforming is mainly determined by the fuel used. Alcohol fuels