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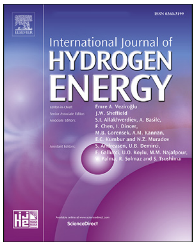

Two-dimensional thermal analysis of radial heat transfer of monoliths in small-scale steam methane reforming

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ABSTRACT

Monolithic catalysts have received increasing attention for application in the small-scale steam methane reforming process. The radial heat transfer behaviors of monolith reformers were analyzed by two-dimensional computational fluid dynamic (CFD) modeling. A parameter study was conducted by a large number of simulations focusing on the thermal conductivity of the monolith substrate, washcoat layer, wall gap, radiation heat transfer and the geometric parameters (cell density, porosity and diameter of monolith). The effective radial thermal conductivity of the monolith structure, $k_{r,eff}$, showed good agreement with predictions made by the pseudo-continuous symmetric model. This influence of the radiation heat transfer is low for highly conductive monoliths. A simplified model has been developed to evaluate the importance of radiation for monolithic reformers under different conditions. A wall gap as thin as 0.05 mm significantly decreased $k_{r,eff}$, while the radiation heat transfer showed limited improvement. A pseudo-homogenous two-dimensional model combined with the symmetric model has been developed for a quick evaluation of geometric parameters for a monolith reformers. Monolithic reformers based on highly conductive substrates e.g., Ni and SiC showed great potential for small-scale hydrogen production.

Introduction

Steam methane reforming (SMR) is well-established method and is currently the preferred route for large-scale hydrogen production which is used as the raw material for the production of ammonia and methanol and for hydrotreating in refineries [1,2]. For the distributed generation of hydrogen on a smaller scale (e.g., for hydrogen fueling stations), SMR is also economically attractive since it takes advantage of the existing natural gas supply infrastructure [3]. More attentions have been paid to the study on small-scale SMR reformers for hydrogen production in the power system (combined with fuel cells) for commercial and residential application [4–7].

Unlike large-scale SMR, small-scale SMR requires a smaller and more compact reformer in cramped conditions with limited space (e.g., for the use of micro-combined heat and power systems in a home) and a quick response for frequent start-up, shut-down, and transient operations [8,9]. These requirements can be better fulfilled by structured reactors such as monolith reactors than by fixed-bed reactors, which are typically applied to large-scale SMR [10]. Monolith reactors

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Nomenclature

a	(-) Adsorption coefficient
Cd	cells/cm^2 Cell density of monolith structure
d	m Diameter of monolith; side length of square channel
E	J/mole Activation energy
F_{kj}	(-) View factor
f	(-) Enhancement factor of radiation heat transfer
f_{wash}	(-) Influence of washcoat layer on the G factor
G	(-) Factor for evaluate the effective thermal conductivity
G_{rad}	(-) G factor considering radiation heat transfer
G_{gap}	(-) G factor considering the wall gap
$G_{gap,rad}$	(-) G factor considering radiation and the wall gap
ΔT	K Temperature difference $T_w - T_c$
ΔT_m	K ΔT value for monolith structure
ΔT_s	K ΔT value for solid structure
ΔT	K Temperature difference
k	W/m·K Thermal conductivity
$k_{CH4,T}$	mole·m^{-3}·s^{-1}·bar^{-1} Reaction constant
k_f	W/m·K Thermal conductivity of gas phase
k_s	W/m·K Thermal conductivity of monolith substrate
$k_{r,eff}$	W/m·K Effective radial thermal conductivity
$k_{r,eff,gap}$	W/m·K Effective radial thermal conductivity considering the wall gap
k_w	W/m·K Thermal conductivity of washcoat
n	(-) refractive index
P_{CH4}	bar Partial pressure of CH_4
q	W/m^2 Heat flux
$q_{out,k}$	W/m^2 Energy fluxes leaving surface k
Q_m	W Total heat transfer rate through monolith structure
$Q_{mono,avg}$	W/m^3 Average reaction heat for the bulk monolithic bed
Q_s	W Total heat transfer rate through pure solid
Q_{wash}	W/m^3 Reaction heat in the washcoat
$Q_{wash,ref}$	W/m^3 Reference reaction heat in the washcoat
R_{wash}	mole·m^{-3}·s^{-1} Reaction rate
R	J·mole^{-1}·K^{-1} Gas constant
S_{ch}	m^2 Surface area of the monolith channels
S_R	W/m^3 Volumetric heat source
T	K Temperature
T_c	K Temperature at the center
T_{ref}	K Reference temperature
T_w	K Temperature at the outer wall
V_{mono}	m^3 Monolith volume
W	m Overall cell dimension

Greek letters

δ_{wash}	µm Washcoat thickness
δ'_{wash}	µm Virtual washcoat thickness
ε	(-) Porosity of a bare monolith without washcoat layer; emissivity of channel surface
ε_k	(-) Emissivity of surface k
λ	µm Thermal conductivity
ξ	(-) Volume fraction of washcoat
ρ_k	(-) Refractivity of surface k
σ	W/m^2·K^4 Stefan-Boltzmann constant
φ	(-) Volume fraction of the gas phase

Subscripts

avg	average
c	Center
ch	Monolith channel
CH_4	Methane gas
f	Gas phase
gap	Wall gap
k	Surface *k*
m	monolith
mono	monolith
rad	Radiation heat transfer
ref	reference
s	Solid; monolith substrate
w	Outer wall; washcoat
wash	washcoat

have been successfully used in environmental applications, especially for the treatment of exhaust gas in automobiles. In recent years, these reactors have gained more interests due to their potential applications in fuel processing, such as steam reforming of hydrocarbons for small-scale hydrogen production [10,11].

A catalytic monolith has a honeycomb structure with parallel channels (Fig. 1) and regular channel shapes (e.g., square, triangle, rectangle, or hexagon). The geometric parameters of the monolith include cell density, porosity, and dimensions of the monolith segment and thickness of the catalyst layer. The substrate material for the monolith can be ceramic or metallic and usually contains a thin layer of catalytic washcoat on the surface of the monolith channels.

Compared with conventionally packed pellets in a fixed-bed reactor, metallic monoliths have a larger void fraction, resulting in a pressure drop of up to two orders of magnitude lower, better heat transfer performance by using monolith substrate with a higher thermal conductivity, and a higher catalyst effectiveness factor (lower diffusional resistance due to thin catalyst layers on monolith surface) [12,13]. These advantages, especially the good heat transfer performance by

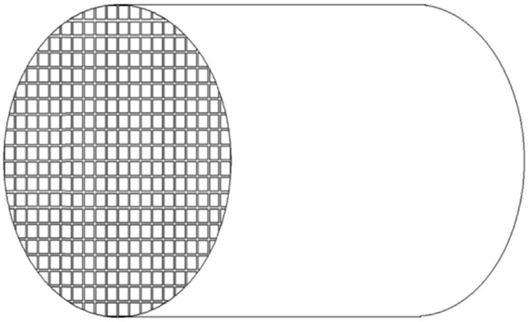

Fig. 1 – Schematic of a typical monolith structure.