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Catalytic partial oxidation of methane for the production of syngas using microreaction technology: A computational fluid dynamics study

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ABSTRACT

Catalytic partial oxidation of methane in high temperature environments under extremely short contact time conditions has emerged as a very promising reaction pathway for the production of syngas. This paper addresses the issues related to the favorable operating conditions for the process. Computational fluid dynamics simulations were performed to gain insight into the underlying mechanism and the key factors affecting primary reaction products. Particular emphasis was given to the role of homogenous and heterogeneous reaction pathways in determining the distribution of reaction products. The effect of preheating temperature, pressure, feed composition, and reactor dimension was investigated in order to identify conditions that will maximize the yield of syngas. Comparisons were made between air-feed and oxygen-feed systems. The relative importance of homogeneous and heterogeneous reactions was assessed, and the reaction pathways responsible for the production of syngas were identified. It was shown that there is a strong interplay between gas-phase and surface chemistry due to the competitive oxidation reactions occurring simultaneously in the system. The contribution of homogeneous and heterogeneous reaction pathways is highly dependent on the operating conditions. Gas-phase chemistry is favored at high preheating temperatures, high pressures, and large reactors, whereas surface chemistry is favored at low preheating temperatures, low pressures, and small reactors, with a tendency to shift towards higher syngas yields. It is particularly beneficial to utilize air instead of oxygen as the oxidant, especially at industrially relevant pressures, thereby inhibiting or avoiding the onset of undesired gas-phase chemistry.

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Nomenclature

A	pre-exponential factor, (cm, mol, s), Table 2
$A_{catalyst}$	catalytically active surface area, m^2, Equation (11)
$A_{geometric}$	geometric surface area, m^2, Equation (11)
$C_{i,interface}$	concentration of the i-th species at the fluid-washcoat interface, mol/m^2, Equation (13)
$c_{p,k}$	specific heat at constant pressure of the k-th gaseous species, J/(kg·K), Equation (7)
d	separation distance between the two plates, m, Fig. 1 and Table 1
d_{pore}	mean pore diameter of the catalyst, m, Equation (16) and Table 1
D	diffusion coefficient, m^2/s, Equation (6)
D^T	thermal diffusion coefficient, m^2/s, Equation (6)
$D_{i,eff}$	effective diffusion coefficient of the i-th species in the catalyst washcoat, m^2/s, Equation (13), as defined by Equation (15)
$D_{k,m}$	mixture-averaged diffusion coefficient of the k-th gaseous species, m^2/s, Equation (6)
Ea	activation energy, kJ/mol, Table 2
$F_{cat/geo}$	catalyst/geometric surface area, m^2/m^2, Table 1, as defined by Equation (11)
$F_{s\text{-}\infty}$	view factor for solid-ambient, unity, Equation (18)
hh_k^o	specific enthalpy, specific enthalpy of the k-th gaseous species at reference temperature, J/kg, Equation (7)
h_o	external heat loss coefficient, $W/(m^2{\cdot}K)$, Equation (18) and Table 1
$\Delta_r H_m^\Theta$	standard molar enthalpy of reaction, kJ/mol, Equation (19)
K_g	number of all gaseous species, Equation (4)
K_s	number of all surface species, Equation (8)
l	reactor length, Table 1
m	number of all gaseous and surface species, Equation (8)
p	pressure, Pa, Equation (2) and Table 1
q	heat flux, W/m^2, Fig. 1 and Equation (17)
R	ideal gas constant, J/(mol·K), Equation (7)
$\dot{s}_{eff}$	effectiveness rate of appearance of a heterogeneous product, $mol/(m^2{\cdot}s)$, Equation (12)
$\dot{s}$	rate of appearance of a heterogeneous product, $mol/(m^2{\cdot}s)$, Equation (8)
T	absolute temperature, K, Equation (4) and Table 1
T_{amb}	ambient temperature, K, Equation (18) and Table 1
T_o	reference temperature, K, Equation (7)
$T_{w,o}$	temperature at the external surface of the solid wall, K, Equation (18)
u	streamwise velocity component, m/s, Equation (1) and Table 1
v	transverse velocity component, m/s, Equation (1)
V_k	diffusion velocity of the k-th gaseous species, m/s, Equation (5)
$\vec{V}_k$	diffusion velocity vector of the k-th gaseous species, m/s, Equation (6)
W_k	relative molecular mass of the k-th gaseous species, dimensionless, Equation (5)
$\overline{W}$	relative molecular mass of the gas mixture, dimensionless, Equation (6)
X	streamwise coordinate, Fig. 1 and Equation (1)
Y	transverse coordinate, Fig. 1 and Equation (1)
Y_k	mass fraction of the k-th gaseous species relative to all species in the gas mixture, Equation (4)

Greek variables

Γ	site density for surface phase, mol/m^2, Equation (8) and Table 1
Γ	catalytically active surface area per washcoat volume, m^2/m^3, Equation (13), as defined by Equation (14)
δ	wall thickness, m, Fig. 1 and Table 1
$\delta_{catalyst}$	thickness of the catalyst washcoat, m, Equation (13)
ε	emissivity, Equation (18) and Table 1
ε_p	catalyst porosity, dimensionless, Equation (15) and Table 1
η	effectiveness factor, Equation (10), as defined by Equation (12)
Θ_i	surface coverage of species i, Table 2
Λ	thermal conductivity, W/(m·K), Equation (4) and Table 1
M	dynamic viscosity, kg/(m·s), Equation (2)
P	density of the gas mixture, kg/m^3, Equation (1)
Σ	Stefan-Boltzmann constant, $W/(m^2{\cdot}K^4)$, Equation (18)
σ_m	site occupancy of the m-th surface species, Equation (8)
τ_p	catalyst tortuosity factor, dimensionless, Equation (15) and Table 1
φ	inlet methane-to-oxygen molar ratio, Table 1
Φ	Thiele modulus, dimensionless, Equation (12), as defined by Equation (13)
$\dot{\omega}$	rate of appearance of a homogeneous product, $mol/(m^3{\cdot}s)$, Equation (5)

Subscripts

Amb	ambient, Equation (18)
G	gas, Equation (4)
I	species index, Equation (12)
In	inlet, Table 1
K	gaseous species index, Equation (4)
M	surface species index, Equation (8)
O	outer, Equation (18)
Rad	radiation, Fig. 1 and Equation (17)
S	solid, Equation (9)
W	wall, Equation (18)
X	streamwise component, Equation (4)
Y	transverse component, Equation (4)