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Review

Preliminary design and simulation of a microstructured reactor for production of synthesis gas by steam methane reforming



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A B S T R A C T

The present study considers the potentials of the well-known production of syngas by steam methane reforming (SMR), by operation within microstructured reactors. The model of a microchannel reactor is developed, including very fast kinetic reaction rates on the coated catalytic walls of the reactor module. By varying the characteristic dimensions of the channels, and considering technical constraints on the design and operating conditions, the results demonstrate that the SMR reactor can be drastically miniaturized while maintaining its productivity without any additional pressure drop. Furthermore, by reducing the channel characteristic dimensions, it is possible to suppress heat and mass-transfer limitations enabling SMR reactor operation at thermodynamic equilibrium. A fast method for preliminary design of microstructured heat-exchanger reactors is developed, that enables to identify the optimal channels number and heat power needed to reach process specifications.

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Keywords: Microstructured reactor design; Methane reforming; Syngas; Hydrogen; Process intensification

Contents

1. Introduction.....	1729
2. Model description.....	1729
2.1. Considered chemical reactions.....	1729
2.2. Microchannel reactor.....	1730
2.3. Reactor model.....	1730
2.3.1. Heat transfer and external mass transfer.....	1731
2.4. Internal mass-transfer investigation.....	1732
2.5. Characteristic times analysis.....	1732
2.6. Choice of the catalyst wash-coat thickness.....	1733
2.7. Constraints on reactor design.....	1733
3. Results.....	1733
3.1. Model validation with literature results.....	1733
3.2. Reference geometry.....	1734
3.3. Influence of heat supply conditions and numbers of channels upon H ₂ production.....	1735
3.4. Comparison of various reactor designs.....	1738

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4. Conclusion	1738
Acknowledgement	1738
Appendix A. Supplementary data	1738
References	1738

Nomenclature

C_{CH_4}	methane concentration (mol/m ³)
$C_{T,g}$	total concentration in the gas phase (mol/m ³)
D_m	molecular diffusivity (m ² /s)
D_h	hydraulic diameter of the microchannel (m)
E_1	activation energy for SMR reaction (J/mol/K)
E_2	activation energy for WGS reaction (J/mol/K)
F_0	total molar flow rate of reactants (mol/s)
$k_{d,j}$	mass-transfer coefficient between the gas and the catalytic wall (m/s)
k_h	heat-transfer coefficient between the gas and the walls (W/m ² /K)
L	the reactor length (m)
P	total pressure (Pa)
R	universal gas constant (J/mol/K)
r_1	rate of the SMR reaction (mol/m ³ _{catalyst} /s)
r_2	rate of the WGS reaction (mol/m ³ _{catalyst} /s)
T_g	gas temperature (K)
T_s	reactor temperature (K)
u_c	gas velocity (m/s)
$y_{g,j}$	gas phase molar fraction of species j
$y_{s,j}$	molar fraction of species j in the catalyst
z	axial position along the channel (m)

Greek notations

α	ratio between the height and the width of the microreactor
μ	dynamic gas viscosity (Pa s)
$\nu_{i,j}$	stoichiometric coefficient of species j in reaction i
λ	thermal conductivity of the gas (W/m/K)

Dimensionless numbers

Gz_{th}	thermal Graetz number
Gz_m	mass Graetz number
Nu	Nusselt number
Pr	Prandtl number
Re	Reynolds number
Sc	Schmidt

1. Introduction

Process intensification is currently a promising strategy to answer the chemical industry needs in terms of competitiveness and respect of new environmental rules. Indeed, this methodology enables to reduce significantly the size of process units, their energetic consumption and environment impacts, while maintaining their productivity. The development of structured catalytic reactors should make possible to surpass the performances of traditional reactors and provide innovative demonstrators, operating safely within new working conditions. Currently, hydrogen production (280 ton/day) requires process units of considerable size (12 m × 17 m × 19 m) and contact times larger than one second (Rostrup-Nielsen et al., 2002). To make this process more efficient, during the last decade, several experimental and

simulation studies of syngas production by using microreactor have been performed. The main motivation is the high surface-to-volume ratio of microreactors, which provides a high heat transfer and allows utilizing the full potential of catalyst during highly endothermic or exothermic reactions. Furthermore, the small characteristic dimensions of microreactors enable to suppress heat and mass transfer resistances and to avoid hot or cold spot formation (Kolb and Hessel, 2004).

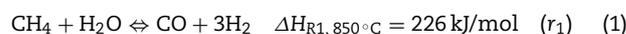
In the frame of process intensification, this study deals with the analysis of new process units for production of syngas by steam methane reforming in microstructured reactors. The main objective of this work is to identify the characteristic dimensions and general design of a microstructured reactor for reaching the maximum productivity. To reach that goal, a numerical model of the structured reactor is developed including a fast kinetic reaction rate. The reactor receives a uniform heat flux for providing heat to the endothermic reaction. Heterogeneous mass transfer between the reactants in the gas phase and the catalyst, as well as heat transfer, are taken into account to quantify potential limitations that are likely to appear. As the reactor design is the only objective of this study, without any attempt to modify the process upstream and downstream, its operation is subject to the general constraints of operating temperature and pressure of the current industrial process. Including these assumptions in a model and additional constraints related to the current operating conditions of the industrial process enables to calculate the performances of a microreactor module. Varying the characteristic dimensions of this module finally makes possible to analyze the optimum design parameters and operating conditions.

2. Model description

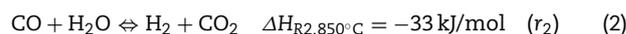
2.1. Considered chemical reactions

When performing steam methane reforming, the main reactions to be considered are:

- the endothermic steam methane reforming (SMR)



- the exothermic water gas shift reaction (WGS)



In industrial practice, these reactions are performed in the presence of nickel or rhodium catalyst at high pressure (20 bars) and high temperature (from 650 to 950 °C) (Stefanidis and Vlachos, 2010; Zeppieri et al., 2010).

The main drawback of steam methane reforming reaction is the risk of carbon formation, which leads to catalyst deactivation and metal dusting (De Groot and Froment, 1995).

In order to avoid carbon formation due to methane cracking, Boudouard reaction and CO reduction, that are harmful for the operation of production units, we use in this study an excess of H₂O (oxidizing agent) corresponding to a steam-to-carbon ratio of 3. The two reactions described above are

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