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ORIGINAL ARTICLE

Parametric sensitivity analysis to investigate heptane reforming in circulating fast fluidized bed membrane reactors

M.E.E. Abashar

Department of Chemical Engineering, College of Engineering, King Saud University, P.O. Box 800, Riyadh 11421, Saudi Arabia

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Abstract In this paper, we present mathematical modeling and numerical simulation tools in searching the high parameter space of steam reforming of heptane for the key design parameters, which have the potential to give high heptane conversion, high hydrogen yield and hydrogen to carbon monoxide ratio within the industrial limits for the syngas used as a feedstock for the gas to liquid processes (GTL). The system under consideration is the novel circulating fast fluidized bed membrane reactor (CFFBMR). The simulation results show that the hydrogen membrane has a significant role in the displacement of the thermodynamic equilibriums of the reversible reactions and production of ultraclean hydrogen, which can be used as a fuel for the fuel cells. Also the results of the sensitivity analysis show that the best performance of the CFFBMR can be obtained by a proper selection of combination of several parameters of high feed temperatures, high steam to carbon feed ratios, high reaction side pressures coupled with a large permeation area of a composite thin film membrane. These parameters are interacting in a very complex manner to give 100% conversion of heptane and 496.94% increase in hydrogen yield compared to the reformer without hydrogen membrane. It was found that under these selected operating conditions a low H₂/CO ratio of 1.15 is achieved satisfying the practical recommended industrial range.

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1. Introduction

The recent high crude oil prices, growing environmental concerns, high energy demand for the rapid growth of the industry and progressive depleting of the fossil fuels have imposed

strong challenges on research and industry to pursue the potential of alternative fuels such as hydrogen and liquid fuels (gasoline and diesel) from syngas (H₂, CO, CO₂). Hydrogen has been known as a clean fuel that does not harm the environment (Abashar, 1990, 2004; Elnashaie and Abashar, 1993; Barbieri and Di Maio, 1997; Abashar et al., 2007, 2008). The potential of the utilization of syngas as a feedstock for the gas to liquid processes (GTL) such as the Fischer–Tropsch (FT) has attracted much interest in recent years (Mark, 1999; Olusola et al., 2010).

Hydrogen and syngas are produced in commercial scale by the conventional steam reforming of methane due to the

E-mail address: mabashar@ksu.edu.sa

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Nomenclature

A_c	free cross-sectional area of the reactor for catalyst circulation, m^2	Re_p	particle Reynolds number, [-]
d_{H_2}	diameter of hydrogen membrane tube, m	T	temperature, K
d_c	diameter of catalyst particle, m	T_f	feed temperature, K
d_c^*	dimensionless diameter of catalyst particle	u_o	superficial gas velocity, m/s
F_j	molar flow rate of component j, kmol/h	u^*	dimensionless gas velocity
g	gravitational acceleration, m/s^2	z	dimensionless length of the reactor
k_i	rate coefficient of reaction i	<i>Greek letters</i>	
K_2, K_4	equilibrium constant of reactions (2) and (4), kPa^2	δ_{H_2}	thickness of hydrogen membrane, μm
K_3	equilibrium constant of reaction (3), [-]	ε	void fraction
L	reactor length, m	μg	viscosity of gas, $kg/m.s$
N_{H_2}	number of hydrogen membrane tubes	ρ_c	catalyst density, kg/m^3
P	total pressure, kPa	ρ_g	gas density, kg/m^3
P_j	partial pressure of component j, kPa	<i>Superscript</i>	
Q_{H_2}	permeation rate of hydrogen, kmol/h	p	permeation side
r_i	rate of reaction i, kmol/kgcat h	r	reaction side
R	gas constant, kJ/mol K		

availability of the natural gas. This process is very expensive due to the excessive heat used in the furnaces to shift the thermodynamic equilibriums for the endothermic reactions for high conversion and yield. Moreover, this process suffers from high diffusion limitations (very low effectiveness factors) and the destructive effect of the elevated temperatures on the catalyst and the reformers (Abashar, 1990; Elnashaie and Abashar, 1993). All these factors have shifted the focus of the research and industry toward new innovative production routes and technologies (Kaihu and Hughes, 2001; Venkataraman et al., 2003; Levent et al., 2003; Prasad and Elnashaie, 2003; Chen et al., 2003a,b; Abashar et al., 2007, 2008).

In last few years, considerable attention has been paid to application of palladium based membranes in the steam reforming industry due to their significant impact on shifting the thermodynamic equilibrium and separation of hydrogen. Substantial improvement in methane conversion and hydrogen yield has been achieved by employing palladium based membranes. An effective method to enhance the hydrogen permeation flux is to employ a composite membrane in which a very thin layer of palladium or palladium alloy is deposited on the surface of a porous thermostable substrate (Shu et al., 1994, 1995; Gobina et al., 1995; Dittmeyer et al., 2001; Hughes, 2001). Techniques such as magnetron sputtering, chemical vapor deposition, solvated metal atom and electroless plating have been successfully employed to deposit very thin palladium films (2–10 μm) on mechanically stable supports (Gobina et al., 1995; Hughes, 2001).

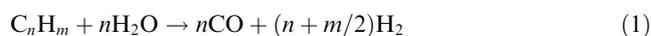
Elnashaie and co-workers have published a series of papers indicating that circulating fast fluidized bed membrane reactors (CFFBMR) have the potential to be the next generations of the reformers for efficient hydrogen production (Prasad and Elnashaie, 2003; Chen et al., 2003a,b; Abashar et al., 2007, 2008). These reactors have many good hydrodynamic characteristics such as: good solid contact, fine catalyst particles are used and high gas throughputs per unit cross-section (Brereton, 1987; Kunii and Levenspiel, 1991, 1997, 2000; Brereton and Grace, 1993; Luan et al., 2000; van der Meer

et al., 2000; Prasad and Elnashaie, 2003; Chen et al., 2003a,b; Abashar et al., 2007, 2008).

Chemical kinetics of higher hydrocarbon such as heptane has been developed and used by many researchers (Phillips et al., 1969; Rostrup-Nielsen, 1973, 1997; Tottrup, 1982; Christensen, 1996; Chen et al., 2003b; Nah and Palanki, 2009; Rakib et al., 2010). It is of a great surprise that there are only a few reported studies in modeling and simulation of steam reforming of heptane in the CFFBMR (Chen et al., 2003b). Also, the experimental data in the literature are very scarce. In the present modeling and simulation study a composite very thin layer of hydrogen membrane of thickness 3 μm of palladium-alloy deposited on a porous support is employed rather than the thick hydrogen membrane used by the earlier study (Chen et al., 2003b). The theme of the study is to investigate further in more detail the large parameter space of the steam reforming of heptane in order to identify the most effective key parameters that influence the performance of the CFFBMR for efficient production of hydrogen and syngas. Furthermore, a special attention has been paid to the H_2/CO ratio, which is a very important factor for the syngas used as a feedstock for the GTL processes. The study locates in this high parameter space, the parameter regions at which the H_2/CO ratio is within the recommended industrial limits.

2. Rate of reactions

Steam reforming of hydrocarbons can be represented in a general form by:



In the case of steam reforming of heptane ($n = 7, m = 16$). Usually this reaction is accompanied by many reactions. Many investigators have proposed that the steam reforming of heptane over a nickel based catalyst is accompanied by methanation reaction, water gas shift reaction and methane overall steam reforming reaction as follows (Phillips et al., 1969; Rostrup-Nielsen, 1973, 1997; Tottrup, 1982; Christensen,

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