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Optimizing the fluidized-bed reactor for synthesis gas production by tri-reforming

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A B S T R A C T

Methane tri-reforming is considered as a highly efficient process for syngas production. In this paper, a fluidized-bed tri-reformer reactor (TR), which is a suitable alternative for conventional steam reformer (CSR) and auto thermal reformer (CAR), is optimized. A mathematical heterogeneous model is applied to simulate the reactor and the model is validated using the plant data. After determining the optimal operating conditions through differential evolution (DE) method, the performance of the proposed reactor is assessed through evaluating the methane conversion, hydrogen production and desired H₂/CO ratio. Methane conversion, hydrogen yield and H₂/CO ratio are 99.4%, 2.1 and 1.84, respectively.

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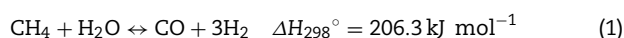
Keywords: Fluidized-bed; Tri-reformer; Optimization; Differential evolution; syngas production

1. Introduction

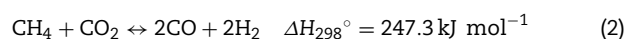
Methanol is considered as a key chemical which could be obtained from a wide variety of resources, mainly from synthesis gas (CO and H₂) (Choudhary and Mamman, 2000; Choudhary and Mondal, 2006; Feng et al., 2009; Ahmed and Gupta, 2009, 2011). The predominant production method of syngas includes steam reforming of natural gas. In this method, obtaining the desired H₂/CO ratio with high methane conversion and limiting coke deposition are among the most important operational issues. It is worth mentioning that the suitable H₂/CO ratio highly varies according to the intended use for the syngas that is 1.5–2 for the methanol production (Jiang et al., 2007).

In the conversion of natural gas to syngas, following basic catalytic reactions present:

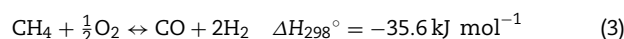
- Steam reforming of methane (SRM):



- Carbon dioxide reforming of methane (CDR) or dry reforming of methane:



- Partial oxidation of methane (POM):



In the previous decade, Song (2001) suggested tri-reforming of methane (TRM) as a novel syngas production procedure. This process refers to simultaneous reforming of oxidative CO₂-steam from natural gas, which involves synergetic combination of CO₂ reforming, steam reforming, and methane oxidations. In order to improve the performance of this process, O₂ and H₂O are used to decrease coke formation on the catalysts and the H₂/CO ratio is maintained around 2 for better syngas production. Considering the fact that reforming reactions are endothermic and oxidation reaction is exothermic, combination of these reactions contributes to energy saving because the produced heat in the partial oxidation of methane is consumed in the reforming reactions. From environmental

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Nomenclature

Symbol

A	cross section area of reactor [m^2]
Ar	Archimedes number [-]
a_b	specific surface area of bubble [m^{-1}]
C_p	specific heat of the gas at constant pressure [$\text{J mol}^{-1} \text{K}^{-1}$]
C_t	total concentration [mol m^{-3}]
D_i	inner diameter [m]
d_p	particle diameter [m]
d_b	bubble diameter [m]
F_t	total molar flow [mol s^{-1}]
F^b	molar flow in bubble side [mol s^{-1}]
F^e	molar flow in emulsion side [mol s^{-1}]
k_1	reaction rate constant for the first rate equation [$\text{mol kg}^{-1} \text{s}^{-1}$]
k_2	reaction rate constant for the second rate equation [$\text{mol kg}^{-1} \text{s}^{-1}$]
k_3	reaction rate constant for the third rate equation [$\text{mol kg}^{-1} \text{s}^{-1}$]
k_{4a}	first reaction rate constant for the fourth rate equation [$\text{mol kg}^{-1} \text{s}^{-1}$]
k_{4b}	second reaction rate constant for the fourth rate equation [$\text{mol kg}^{-1} \text{s}^{-1}$]
K_{bei}	mass transfer coefficient for component i in fluidized-bed [m s^{-1}]
L	reactor length [m]
P_i	partial pressure of component i in reaction side [bar]
r_i	reaction rate of component i [$\text{mol kg}^{-1} \text{s}^{-1}$]
r_{bi}	reaction rate of component i in bubble phase [$\text{mol kg}^{-1} \text{s}^{-1}$]
r_{ei}	reaction rate of component i in emulsion phase [$\text{mol kg}^{-1} \text{s}^{-1}$]
R_1	first rate of reaction for steam reforming of CH_4 [$\text{mol kg}^{-1} \text{s}^{-1}$]
R_2	second rate of reaction for steam reforming of CH_4 [$\text{mol kg}^{-1} \text{s}^{-1}$]
R_3	water–gas shift reaction [$\text{mol kg}^{-1} \text{s}^{-1}$]
R_4	rate for total combustion of CH_4 [$\text{mol kg}^{-1} \text{s}^{-1}$]
R	universal gas constant [$\text{J mol}^{-1} \text{K}^{-1}$]
T	bulk gas phase temperature [K]
T_w	wall temperature of steam reformer [K]
u	superficial velocity of fluid phase [m s^{-1}]
u_b	velocity of rise of bubbles [m s^{-1}]
U_w	overall heat transfer coefficient between tube wall and reaction side streams in steam reformer [$\text{W m}^{-2} \text{K}^{-1}$]
y_{ib}	mole fraction of component i in the bubble phase [-]
y_{ie}	mole fraction of component i in the emulsion phase [-]
z	axial reactor coordinate [m]

Greek letters

α	activity of catalyst (where $\alpha = 1$ for fresh catalyst) [-]
ρ_B	density of catalytic bed [kg m^{-3}]
ρ_e	density of emulsion phase [kg m^{-3}]
ρ_g	density of fluid phase [kg m^{-3}]
ρ_p	density of catalyst [kg m^{-3}]

$\Delta H_{f,i}$	enthalpy of formation of component i [J mol^{-1}]
ΔH_{298}	enthalpy of reaction at 298 K [J mol^{-1}]
δ	bubble phase volume as a fraction of total bed volume [-]
ε_{mf}	void fraction of catalytic bed at minimum fluidization [-]
η	catalyst effectiveness factor [-]
μ	viscosity of fluid phase [$\text{kg m}^{-1} \text{s}^{-1}$]
γ	volume fraction of catalyst occupied by solid particle in bubble [-]
φ_s	sphericity of catalyst [-]

Subscript

0	entrance of reactor
i	component i
j	reaction number index
p	particle
g	gas phase
b	bubble phase
e	emulsion phase
mf	minimum fluidization

point of view, in this process, CO_2 (which exists in effluent gases) is consumed as a co-reactant and is turned into syngas that is largely used in methanol, dimethyl ether and other valuable components.

After the introduction of methane tri-reforming (TRM) in laboratory scale by Song (2001) in 2001, researchers have investigated this process extensively (Song et al., 2002; Hong-tao et al., 2007; Lee et al., 2003a,b; Tomishige, 2004). In recent years, tri-reforming process has been applied to di-methyl-ether (DME) production in a pilot plant and demonstration unit in Korea (Cho et al., 2009).

Arab Aboosadi et al. (2011) used differential evolution (DE) procedure to optimize tri-reformer reactor and studied the influence of various parameters including steam/ CH_4 on gas temperature, inlet temperature, O_2/CH_4 , CO_2/CH_4 , methane conversion, H_2/CO ratio and hydrogen production. The results showed that the conversion of methane enhanced by 3.8% compared to industrial reformers in a single reactor. García-Vargas et al. (2013) studied the effect of the feedstock composition on methane conversion, the H_2/CO molar ratio of the synthesis gas obtained by tri-reforming of methane and the heat released or supplied to the system with a Ni/ β -SiC catalyst. Lo Faro et al. (2013) optimized the coupling of a biogas-fed solid oxide fuel cell (SOFC) with an external tri-reforming system over ceria supported Ni catalyst (1.75-wt.% Ni/ CeO_2). They showed that this process is a promising process for small and medium sized stationary power systems. Rahimpour et al. (2012) investigated syngas production in fixed-bed tri-reformer reactor. They used a hydrogen and oxygen selective membrane for methanol production.

Non-convex, non-linear, and constrained optimization problems are considered as the most frequent industrial optimization problems. An optimum design, which includes the most suitable condition, mostly contributes to the lowest cost per unit of time or the highest profit per unit of production. Recently, quite a few number of search algorithms techniques have been suggested in the fields of evolutionary programming (EP) (Viecco and Caram, 2002), evolution strategies (ES) (Rechenberg, 1973), genetic algorithms (GA) (Holland, 1975)

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