



Multi-objective optimization of industrial membrane SMR to produce syngas for Fischer-Tropsch production using NSGA-II and decision makings



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ABSTRACT

Membrane reactors are an advanced technology with vast application capacities for equilibrium limited endothermic reactions. The main propose of this study is to offer an optimized packed-bed membrane steam methane reforming (SMR) tubular reactor for sustainable CH₄ conversion by implementing triple-objective optimization model based on optimum H₂/CO ratio for low temperature Fischer-Tropsch (F-T) process. In this study a one dimensional pseudo-homogeneous model based on mass, energy, and momentum conservation laws is used to simulate the behavior of a packed-bed membrane reactor for production of syngas by SMR. In the optimization section, the proposed work explores optimal values of various decision variables that simultaneously maximize CH₄ conversion, H₂ selectivity, and CO selectivity by applying elitist non-dominated sorting genetic algorithm (NSGA-II). Pareto optimal frontier between triple objectives is obtained in three spaces and best optimal value is selected by using LINMAP, TOPSIS, Shannon's entropy and Fuzzy Bellman-Zadeh decision making methods. The final optimal solutions illustrate that the membrane reactor presents higher CH₄ conversion which can be operated under milder conditions than the conventional reactor.

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1. Introduction

Syngas as a mixture of H₂, CO and CO₂ is the building block in the synthesis of petrochemical industries such as ammonia, methanol, Fischer-Tropsch (F-T) fuels and other fine chemicals (Christensen and Primdahl, 1994). The gas composition varies with the intended use of the syngas; methanol synthesis and high temperature (F-T) synthesis require the syngas composition which is specified by a stoichiometric number ($S_R = (H_2 - CO_2)/(CO + CO_2)$) which should be closed to 2. On the other hand, for the low temperature (F-T) synthesis, the syngas should have a H₂/CO ratio close to 2 (Larentis et al., 2001), Or H₂/CO ratio close to 1 is suitable for manufacture of oxygenated hydrocarbons and polycarbonate products. These different syngas compositions are attained by applying various types of reactor technology and by varying the amount of feed composition.

Nowadays the SMR is considered the main process used to produce syngas. However the conventional industrial SMR guarantee appropriate CH₄ conversion but it leads to a high H₂/CO ratio between 3 and 6 (depending on feedstock) in the outlet stream. Using Pd-based membrane SMR reactor is a technology with huge application potentials for break equilibrium limited reforming reactions by removing the product H₂ from the reaction mixture. The idea of process intensification of SMR by using a membrane reformer could permit syngas to be produced in different ratios of H₂/CO viz. 1, 2 or 3, depending on the purpose for which it has to be used (Mohanty, 2006).

Two different configurations of membrane SMR reactors have been experienced and applied: a packed bed membrane reactor and fluidized bed membrane reactor. Several research efforts have been made to investigate the two various membrane SMR reactors and enhance the production of the desired products.

For instance, in fluidized bed membrane reactors for SMR, Adris et al. developed a comprehensive mathematical model for the fluidized bed membrane reactor (FBMR) system. They concluded that the membrane capacity and their distribution between the dense catalyst bed and the freeboard region are essential design

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parameters in FBMR (Adris et al., 1997). Abashar et al. investigated the performance of concurrent and countercurrent fluidized bed membrane reactors for SMR. Experimental and theoretical results of this study indicated that the countercurrent fluidized bed membrane reactors display substantial improvement over the equilibrium conversions (Abashar et al., 2003). Patil et al. studied experimentally a novel multifunctional fluidized bed membrane reactor for ultrapure H₂ production via SMR. They concluded that the reactor performance can be enormously improved with the use of perm-selective H₂ membranes in terms of improved CH₄ conversion, decreased CO selectivity, and improved H₂ product yield and power output (Patil et al., 2007). Dehkordi et al. presented a mathematical model to describe the flow pattern of gas within the dense zone of a tapered membrane-assisted fluidized-bed reactor, in the bubbling mode of operation for SMR under wall heat flux. They investigated effects of various operating and design parameters such as the tapered angle, bed operating temperature and pressure, and feed temperature on the CH₄ conversion and the total yield of H₂ (Dehkordi et al., 2011).

Also in packed bed membrane SMR reactors, Oklany et al. simulated the SMR using catalytic membrane reactors for two types of a dense Pd–Ag composite membrane and a series of microporous membranes. They determined that Pd–Ag membranes gave the better performance, for all parameters investigated including temperature, pressure, sweep ratio, and membrane thickness (Oklany et al., 1998). Gallucci et al. developed a simulation study of the SMR in a dense tubular membrane reactor, and considering the influence of the different parameters on CH₄ conversion. They concluded that using membrane reactor with optimum operational conditions makes possible to take the same conversion of conventional SMR reactor but operating at a lower temperature, with a resulting energy saving, and keeping the pure H₂ production from the permeable side stream (Gallucci et al., 2004). Bottina et al. presented a mathematical model of a non-adiabatic SMR membrane reactor, to investigate the influences of operational variables on the membrane area and the energy required by the process, which in turn affect fixed and operating costs (Bottino et al., 2006). Ohmori et al. carried out comparative studies via numerical simulation to analyze the performances of a porous ceramic membrane reactor for SMR using nitrogen and steam as sweep gases. As a result, the use of steam as the sweep gas showed the better performance in terms of CH₄ conversion and H₂ recovery yield (Yu et al., 2008). Marín et al. studied numerical simulation of 2D model of membrane packed bed SMR, which is formed by Ru–SiO₂ catalyst particles. In this study, the optimum operating conditions, corresponding to a maximum H₂ permeation rate, were determined, and the behavior of the optimized reactor was analyzed in detail (Marín et al., 2012).

For many complicated problems, the multi-objective optimization (MOO) is more accurate and real compared to the single-objective approach. MOO problems satisfy a number of different and even conflicting objectives at the same time, which is known as Pareto optimal solutions (Deb, 2005; Liu, Reynolds). In this regard, the MOO has been studied for different membrane reactors and engineering problems (Isebor and Durlinsky, 2014; Taghdisian et al., 2015). For instance, Cheng et al. carried out the triple-objective function optimization for the catalytic membrane reactors, for methanol synthesis as well as hydrogen generation, respectively. In this study, for the two catalytic membrane reactors, the optimization problem is developed to define the operating conditions that maximize the major product rate $F_{\text{CH}_3\text{OH}} & F_{\text{H}_2}$ and minimize both the major reactant rate $F_{\text{H}_2} & F_{\text{CH}_4}$ and the exergy loss (Cheng et al., 2008). Sharma et al. investigated MOO of a membrane distillation (MD) System for desalination of sea water. In this study, MOO is performed for design of MD module to explore the trade-off

between conflicting high water production rate and lower energy consumption objective functions (Sharma et al., 2012). In another study, Soleimani et al. presented experimental investigation, modeling and optimization of membrane separation of oily wastewaters using artificial neural network and multi-objective optimization. In this study, by applying genetic algorithm, an optimization tool was created to predict the optimum parameters for desired permeation flux (i.e. maximum flux) and fouling resistance (i.e. minimum fouling) behavior (Soleimani et al., 2013).

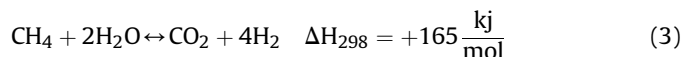
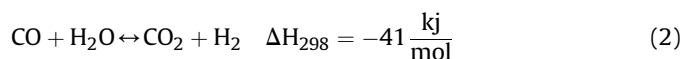
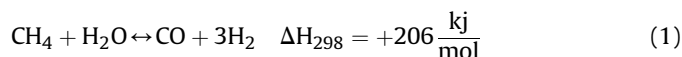
In this work, we point out our attention toward the influence of the membrane on CH₄ conversion with approaching H₂/CO ratio to optimal value of 2, by implementing MOO model that simultaneously maximizes CH₄ conversion, H₂ and CO selectivity. Controlling the H₂/CO ratio is important for F-T process and influences hydrodynamic products distribution.

In this regard, as a starting point in Section 2, the packed bed SMR in the conventional and membrane form are explained. In Section 3, the reaction rate equations and mathematical model of the hydrogen-permselective membrane SMR reactor are presented. In Section 4, the mathematical procedure to develop the multi-objective function for optimization of the process by NSGA-II algorithm is described. Furthermore, generating the optimal Pareto set, applying decision-making methods such as LINMAP, TOPSIS, Shannon's entropy and Fuzzy Bellman-Zadeh, the use of numerical solution method and providing steady state model validation of base case are explained. In Section 5, the optimum operating conditions corresponding to MOO formulation are determined. Also, the behavior of the optimized membrane reactor is analyzed in detail. Finally the conclusion is presented in Section 6.

2. Process description

2.1. Conventional reactor

In the conventional steam reforming process, since sulfur acts as a poison for reforming catalysts, it is removed from natural gas over a zinc oxide catalyst. Then, before in letting the reformer the natural gas is mixed with excess steam and then prepared steam reformer feed is preheated to higher temperatures. The preheated mixture is entered to the catalytic section of the reformer, which are implemented in top fired furnace. The reformer reactor is conventional vertical tubes, which are packed with the catalyst pellets. The reactor tubes are about 10–12 m long, with diameters of about 10–12 cm. The following catalytic reactions are the main reactions which are taking place in tubular fixed bed reactor of low diameter-to-height ratio to ensure efficient heat transport in radial direction:



The reactions taking place here are the two endothermic steam reforming reactions (R₁ and R₃) of CH₄ and the parallel exothermic water gas shift reaction (R₂), respectively. Equilibrium conversions of both reforming reactions benefit from high temperatures and low pressures, whereas the water gas shift reaction (R₂), being exothermic and having no change in the number of moles, benefits thermodynamically from lower temperatures and is independent of pressure. Although a high pressure negatively affects the CH₄

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