

Multi-objective Optimization of Mixed Membrane Reactors for Autothermal Reforming of Methane

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Abstract

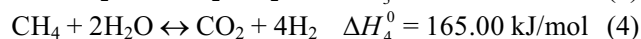
Autothermal reforming (ATR) of methane, which supplies the heat for endothermic steam reforming by internal combustion of methane, is an important process for synthetic gas production. The axial-distributed feeding of oxygen via a packed bed inert membrane reactor (MR) can reduce the peak temperature and improve the reactor performance. A modified MR, called mixed membrane reactor (MMR), combines permeable membrane tube wall and non-permeable tube wall provides extra degrees of freedom for reactor design and operation. For MR and MMR, this study presents the ternary-objective optimization analysis for maximizing hydrogen production rate, non-combustion selectivity and conversion of methane, using a 1D pseudo-homogeneous reactor model and the NSGA-II algorithm. Compared to MR, MMR can be operated under significantly higher oxygen permeation flux without violating the maximum temperature constraint. The non-combustion selectivity and conversion of methane of MR and MMR are close, however, the hydrogen production rate of MMR can be as high as 200% of MR.

Key Words: Methane Reforming, Membrane Reactor, Mixed Membrane Reactor, Multi-objective Optimization

1. Introduction

Synthesis gas (syngas, $\text{CO} + \text{H}_2$) forms the basis for a wide range of industrial products, such as ammonia, methanol, dimethyl ether and hydrogen [1]. Worldwide demand for syngas is rising, in large part driven by a growing need for hydrogen in petroleum refineries, petrochemical industry, transport fuel in fuel cells for electric vehicles [1]. Syngas from steam reforming of methane (SRM) is growing in popularity as the spread of hydraulic fracturing contributions to a fall in the price of natural gas. In SRM, methane is converted with steam on supported Ni catalysts in a highly endothermic reaction to yield H_2/CO . The SRM reaction is conducted in large tubular reactors to achieve high temperatures needed for considerable syngas yields, making steam reforming a

major energy consumer. An alternative process to SMR is the autothermal reforming (ATR) process, which supplies the reaction heat by catalytic combustion of part of the methane using pure oxygen or air [2]. The overall chemical reactions taking place in the ATR include both exothermic reactions, i.e. combustion (Eq. (1)) and water gas shift (Eq. (3)), and endothermic reactions of steam reforming (Eqs. (2), (4)).



In ATR, the distribution of the oxygen feed along the reactor length is critical to the amount of methane consumed through combustion reaction versus reforming reaction. Packed bed inert membrane reactor (PBIMR) is a

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multifunctional process intensification design of reactors which combines the functions of membrane separation and reaction [3]. In PBIMRs, the reaction species penetrate through an inert membrane, either adding into or extracting from the packed bed reactors. PBIMRs have been used successfully as oxygen distributors in a number of oxidation processes, including methane oxidative coupling and the production of olefins and oxygenates from oxidation of alkanes [4]. For the oxidative dehydrogenation of ethane, a theoretical study comparison of a PBIMR with a traditional packed bed reactor [5] shows that in addition to the enhancement of reaction selectivity and conversion, the use of the PBIMR can prevent oxygen accumulation, reduce the occurrence of ‘hot spots’ within the reaction bed and improve the process safety. Simulation study [6] also indicates that PBIMR is a promising alternative to carrying out ATR of methane at milder conditions than those commonly employed in conventional reactors.

A modified membrane reactor has been proposed and experimentally tested for the oxidative dehydrogenation of ethane to produce ethylene [7]. The modified membrane reactor is a packed bed inert membrane reactor (PBIMR) followed by a conventional packed bed reactor (PBR) and is called a mixed membrane reactor (MMR). Using the MMR, higher conversions of ethane was obtained, while maintaining a good selectivity. Compared to PBIMRs, extra degree of freedoms are provided by MMRs for the adjustment of the reactant species permeation.

For the performance enhancement of methane reforming processes, many papers have presented the parametric study and optimization study for various methane reforming reactors using simulation models. For example, in [8], a thermodynamic equilibrium model was used to investigate the effects of operational parameters, such as temperature, pressure, steam to carbon ratio and oxygen to carbon ratio, on the hydrogen yield and hydrogen to carbon monoxide ratio. For an industrial steam methane reformer, design of experiment and response surface methodology were employed for the optimization of hydrogen production or methane conversion [9]. Incorporating membrane separation for hydrogen product removal to a SMR reactor is an effect approach for enhancement of reactor performance. Optimal solutions of a packed

bed membrane steam methane reforming tubular reactor for simultaneously maximizing methane conversion, hydrogen selectivity and carbon monoxide selectivity were obtained [10] by applying elitist non-dominated sorting genetic algorithm (NSGA-II) [11].

This study is aimed at obtaining optimal solutions of PBIMR and PBIMMR for methane autothermal reforming. The packed bed membrane reactor and mixed membrane reactor, hereafter referred as MR and MMR, for ATR of methane to be investigated in this study are illustrated in Figure 1. The tubular reactors are packed with catalyst particles. For MR, the tube wall is the oxygen permeable membrane. For MMR, the tube wall is composed of repetitive sections. In this study, each section of the MMR comprises an oxygen permeable membrane part followed by a non-permeable part with the same length. The shell side of the reactors is the oxygen gas flow channel. The reactant gas and the oxygen gas flows are co-current. For these reactors, multiple performance criteria must be taken into account and they are determined by multiple operating and design variables. In the present study, multi-objective optimization analysis of MRs and MMRs for ATR of methane obtained using the genetic algorithm, NSGA-II [11], are presented and compared. In the following sections, one-dimensional pseudo-homogeneous models for MR and MMR are presented first, followed by the definitions of the optimization problems and finally the discussion of the pareto optimal solutions.

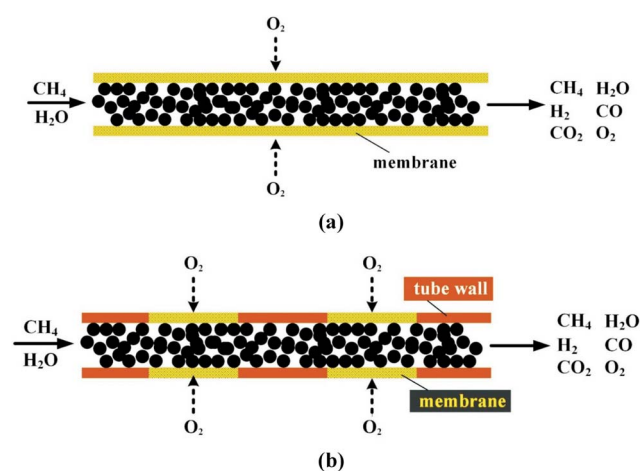


Figure 1. Schematic illustrations of (a) membrane reactor (MR) and (b) mixed membrane reactor (MMR) for autothermal reforming (ATR) of methane.

2. Mathematical Model

A one-dimensional, pseudo-homogeneous, steady state model is used to represent the ATR of methane in a multi-tubular packed bed MR and MMR. The tube-side feed includes methane and steam. The concurrently fed shell-side gas is pure oxygen and part of it will permeate through the tubular porous membrane into the tube-side. The assumptions of the model include:

- Radial variations of temperature and concentration are neglected because of the small tube diameter,
- Axial dispersion of mass and energy of fluid and the external heat and mass transport limitations of catalyst particles are neglected because of the high flowrates,
- Internal mass transport limitations of catalyst particles are accounted for by means of constant effectiveness factors [12] for the four reactions,
- Internal heat transfer limitation of catalyst particles is neglected,
- Bed porosity is assumed to be constant,
- The tube-side and shell-side fluids follow ideal gas law,
- The mass transfer resistances on both sides of the membrane wall are neglected,
- The heat loss from the reactor to the environment is neglected,
- The pressure drop of the tube-side gas is estimated using Ergun equation [13] and the pressure drop of the shell-side gas is neglected.

For the reaction gas in the catalyst tube, the mass and energy balance equations take into account the reactions (Eqs. (1)–(4)), the permeation flux (for oxygen) and the heat transfer between the shell-side and tube-side gases are:

$$\frac{dF_j}{dz} = A_T \rho_B \sum_{i=1}^4 v_{i,j} r_i \eta_i + J_j \pi d_T \quad (5)$$

$j = 1 \sim 6$ (CH_4 , H_2 , H_2O , CO_2 , CO , O_2)

$$\frac{dT}{dz} = \frac{A_T}{\sum_{j=1}^6 F_j C_{p,j}} \left[\rho_B \sum_{i=1}^4 r_i (-\Delta H_{r_i}) \eta_i - \frac{4}{d_T} (J_{\text{O}_2} C_{p_{\text{O}_2}} + U)(T - T_s) \right] \quad (6)$$

For the pure oxygen gas in the shellside, the mass

balance and energy balance equations take into account the permeation flux of oxygen and the heat transfer between the shell-side and tube-side gases are:

$$\frac{dF_s}{dz} = -J_{\text{O}_2} \pi d_T n_T \quad (7)$$

$$\frac{dT_s}{dz} = \frac{\pi U d_T n_T (T - T_s)}{F_s C_{ps}} \quad (8)$$

The reaction rates (r_i) of reactions (1)–(4) for autothermal reforming of methane using the common Ni-based catalysts can be calculated using the reaction kinetics reported in the literature [6,12,14]. The constant effectiveness factors (η_i) for the four reactions reported in [12] were used.

The ceramic inert porous membrane described in [7, 12] is adopted for this study. In their study, the commercial tubular microfiltration membranes (SCT, Inocermic) were modified by deposition of silica in order to attain the desired permeation characteristics and to minimize the back permeation of species in the tube side. The membranes were applied in [6,7,15–17] for the productions of ethane and maleic anhydride. The membrane is assumed to be selective to oxygen, hence the permeation fluxes of the species other than oxygen are zero. The permeation flux of oxygen (J_{O_2}) was calculated considering the Knudsen diffusion and Poiseuille flow inside the membrane pores. The values of membrane thickness and permeation parameters were given in [16].

$$J_{\text{O}_2} = -\frac{1}{RT_m} \Delta P_{mem} \left[\left(\frac{D_{k,\text{O}_2}}{\delta} \right) + \left(\frac{B_0 P_m}{\delta \mu_{\text{O}_2}} \right) \right] \quad (9)$$

For the MMR, a sharp step change of oxygen permeation flux causes convergence difficult. A smooth change of oxygen permeation flux was hence used by employing a logistic function as:

$$f(x) = r_A \left(1 - \frac{1}{(1 + e^{-k(x-x_0)})} \right) \quad (10)$$

The function describes the permeation profile for one segment, which includes a permeable section and a non-permeable section, of the MMR. In the logistic function, x represents the relative position within a reactor segment which includes a permeable section followed by a non-permeable section. The parameters of the logistic

function, including r_A , k and x_0 , can be adjusted. The term r_A is used to make the total flux of the reactor section of MMR equal to the total flux of the same length of MR. In this study, x_0 is fixed at 0.5. The profiles with different k values are shown in Figure 2. The step function can be approached by using high k values. The oxygen permeation flux determined from Eq. (9) was multiplied by the logistic function value (f) determined from Eq. (10).

The overall heat transfer coefficient (U) was estimated using the correlations for packed beds [18].

The boundary (inlet) conditions are:

$$F_j = F_{j,in}, F_S = F_{S,in}, T = T_{in}, T_S = T_{S,in}, P = P_{in}, P_S = P_{S,in} \quad (11)$$

The performance of the reactor evaluated include the hydrogen production rate, the conversion of methane and the non-combustion selectivity of methane reacted. The conversion and selectivity are defined as:

$$X_{CH_4} = 1 - \frac{F_{CH_4,out}}{F_{CH_4,in}} \quad (12)$$

$$S_{non\ comb} = 1 - \frac{F_{total\ O_2\ consumed} / 2}{F_{CH_4,in} - F_{CH_4,out}} \quad (13)$$

The models of MR and MMR were solved using the ode15s solver of Matlab, which is effective for stiff differential equations and DAEs (differential-algebraic equations). The model was first verified for the conventional packed bed reactor reported in [12]. The reactor performance obtained from simulation are fairly close to the real plant data as listed in Table 1. For the membrane reactor, the simulation results obtained from this study are also in consistent with those presented in Rodríguez's paper [6].

3. Optimization Problems

In the present study, the task is to find the optimal solutions for the MR and MMR with given reactor sizes, catalyst packing and membrane characteristics, as listed in Table 2. The reactor is composed of 90 tubes with inside diameter of 0.0156 m and the shell with inside diameter of 0.3048 m. The catalyst particle has a diameter of 1

mm and the bed porosity is 0.4. The catalyst packing density is 1413 kg/m³ bed. The membrane thickness is 1.75 mm.

The optimization problems for MR and MMR are both defined for maximizing the three objective functions, including the production rate of hydrogen ($F_{H_2,out}$), the non-combustion selectivity of methane ($S_{non\ comb}$) and the conversion of methane (X_{CH_4}). The decision variables include the tube-side feed rates of methane and steam, the shell-side pure oxygen feed rate, the tube-side inlet pressure and temperature, the trans-membrane pressure drop,

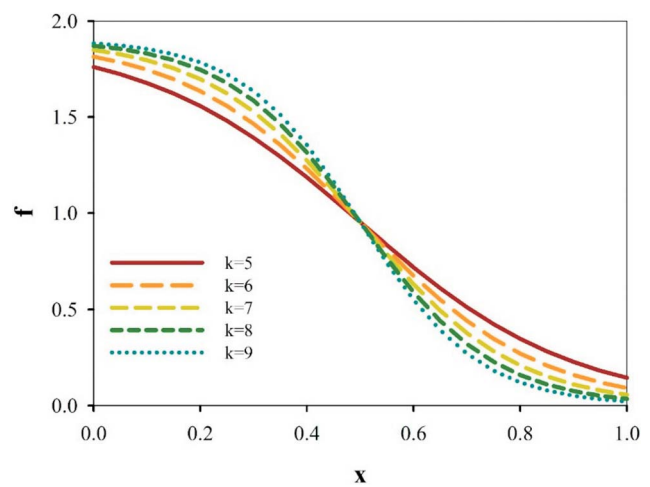


Figure 2. Logistic function.

Table 1. Model verification

Inlet conditions		
P_{in} (atm)	25	
T_{in} (°C)	535	
$F_{CH_4,in}$ (Nm ³ /h)	3483	
$F_{O_2,in} / F_{CH_4,in}$	0.598	
$F_{H_2O,in} / F_{CH_4,in}$	1.4	
Product conditions	Industrial reactor [12]	This study
T_{out} (°C)	950	934
y_{O_2}	0	6.09×10^{-6}
y_{CH_4}	0.008	0.0034
y_{H_2}	0.456	0.452
y_{CO}	0.160	0.175
y_{CO_2}	0.07	0.0495
y_{H_2O}	0.306	0.319
F_{H_2} (Nm ³ /h)	6976	6882
F_{CO} (Nm ³ /h)	2444	2678

the reactor length and the catalyst packing density. For the MMR, the extra decision variable is the number of the mixed wall segments. Note that each segment is composed of a permeable section and a non-permeable section with equal length. The constraints are the lower and upper bounds of the decision variables. In addition, considering the thermal stress on the reactor materials and the damage on the catalyst, a maximum temperature ($T_{\max}$) constraint was included [6,12]. The formulation of the optimization problem can be expressed as:

$$\text{Maximize } (F_{H_2, \text{out}}, S_{\text{non comb}}, X_{CH_4}) \quad (14)$$

Subject to:

$$1 \text{ kmol/h} \leq F_{CH_4, \text{in}} \leq 5 \text{ kmol/h}$$

$$1 \leq F_{H_2O, \text{in}} / F_{CH_4, \text{in}} \leq 5$$

$$1 \text{ kmol/h} \leq F_{S, \text{in}} \leq 4 \text{ kmol/h}$$

$$0.5 \text{ bar} \leq \Delta P_{\text{mem}} \leq 2 \text{ bar}$$

$$500 \text{ }^\circ\text{C} \leq T_{\text{in}} \leq 700 \text{ }^\circ\text{C}$$

$$24 \text{ bar} \leq P_{\text{in}} \leq 30 \text{ bar}$$

$$1 \text{ m} \leq L \leq 4 \text{ m}$$

$$1200 \text{ kg}_{\text{cat}}/\text{m}^3 \leq \rho_B \leq 1800 \text{ kg}_{\text{cat}}/\text{m}^3$$

$$T_{\max} < 1200 \text{ }^\circ\text{C}$$

The NSGA-II (non-dominated sorting genetic algorithm) developed by Deb et al. [11] was employed for the multi-objective optimization search. The gamultiobj solver of Matlab was used and the number of generations and populations were set to be 200 and 50, respectively. With a pareto fraction of 0.35, 18 pareto solutions can be obtained.

4. Results and Discussion

With the mathematical models of MR and MMR, the performance and internal profiles of the two types of reactors operated under the base case conditions as listed in Table 2 can be obtained. For the MMR, the logistic function parameters r_A , n , k and x_0 were specified as 1.9, 1, 8 and 0.5. The temperature profiles of MR and MMR are similar, as presented in Figure 3(a), and both show a peak temperature of the tube-side gas at a location near 0.6 m. The hydrogen production profiles shown in Figure 3(b) for MR and MMR are also similar and reveal that the steam reforming reaction does not occur until the tube-side temperature has become high enough by the

oxidation reaction. The start of the endothermic steam reforming reaction at about 0.6 m then results in a sharp decrease of temperature as well as a steep rise of hydrogen production. In the following section of the reactor, the temperature and the hydrogen production keep increasing slightly in MR, but remain relatively constant in MMR. These results can be explained by the trans-membrane oxygen flux profiles shown in Figure 3(c). In MR, the oxygen fluxes at the front and rear sections of the reactor both remain constant with lower flux at the rear section due to the higher temperature. In MMR, the oxygen flux profile is mainly determined by the logistic function and the flux at the rear section is very small. The dip of the oxygen flux profile at 0.6 m in MR is due to the peak temperature as shown in Figure 3(a). The sustaining of the oxygen flux in the rear section of MR allows the temperature and hydrogen production stay increasing. On the contrary, the very low oxygen flux in the rear section of MMR results in much less increase in temper-

Table 2. Reactor specifications and the base case operation conditions and performance of MR and MMR

Reactor specifications	MR	MMR
React length (m)	1	
Tube diameter (m)	0.0156	
Shell diameter (m)	0.3048	
Number of tubes (-)	90	
Catalyst particle diameter (m)	0.001	
Catalyst packing density (kg _{cat} /m ³ bed)	1413	
Bed porosity (-)	0.4	
Membrane thickness (m)	0.00175	
r _A /n/k/x ₀	-	1.9/1/8/0.5
Base case operation conditions		
F _{CH4,in} (kmol/h)	1.641	
F _{H2O,in} (kmol/h)	2.298	
F _{S,in} (kmol/h)	3	
P _{in} (atm)	25	
ΔP _{mem,in} (atm)	0.7	
T _{in} (°C)	535	
T _{S,in} (°C)	535	
Base case performance		
F _{H2,out} (Nm ³ /h)	61.67	41.87
T _{max} (°C)	1024.2	918.9
L(T _{max}) (m)	0.575	0.606
S _{non comb} (-)	0.725	0.732
X _{CH4} (-)	0.730	0.451

ature and hydrogen production. The profiles of the oxygen mole fraction in the tube-side are also shown in Figure 3(b). In both MR and MMR, the increase of gas mole flow due to steam reforming reactions causes the sharp decrease of the oxygen mole fraction.

The performance data of MR and MMR listed in Table 2 indicate that (1) the maximum temperature of MMR is significantly lower than that of MR by about 100 °C, (2) the hydrogen production and methane conversion of MR are higher than those of MMR, and (3) the non-combustion selectivity of methane of MMR is slightly higher than that of MR. Note that the base case operation conditions are not the optimal ones for either MR or MMR. The optimal solutions for MR and MMR will be discussed in the following.

Because the logistic function is employed for the simulation of MMR, a binary optimization for maximizing $F_{H_2,out}$ and $S_{non-comb}$ were carried out first to determine the best logistic function parameter k and the segment number n . The values of r_A and x_0 were fixed at 1.9 and 0.5. The effects of k and n on the optimal solutions are shown in Figure 4(a) and (b), respectively. The optimal solutions obtained by different k values are fairly close. However, solutions with higher objective functions were obtained when using only one segment. Therefore, $k = 8$ and $n = 1$ were adopted in the ternary-objective optimization analysis.

The distributions of the ternary-objective optimal so-

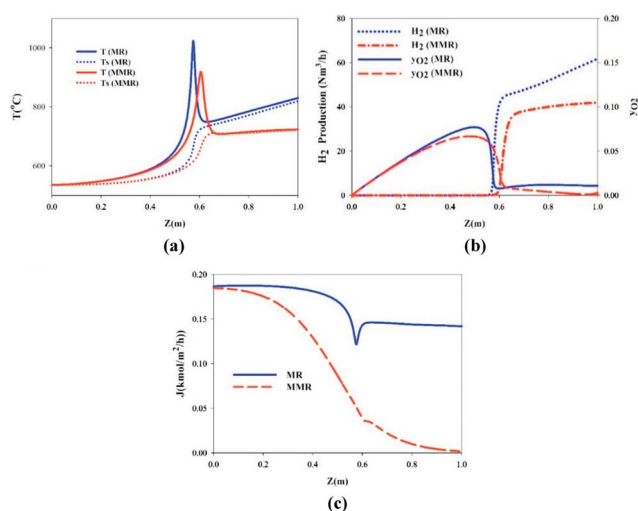


Figure 3. Comparisons of (a) temperature, (b) hydrogen production and oxygen concentration, and (c) transmembrane oxygen flux for the base case operation of ATR-MR and ATR-MMR.

lutions for MR using different catalyst packing densities are summarized in Figure 5. In Figure 5(a), it is shown that most of the solutions simultaneously having higher methane conversion and higher hydrogen production. That means the two objective functions are not in a trade-off relationship. On the contrary, the non-combustion selectivity of methane and the hydrogen production are in a close to linear disproportional relationship as shown in Figure 5(b). The non-combustion selectivity and the conversion of methane are also reversely related as shown in Figure 5(c), but solutions with nearly complete conversion can have a wide-spread selectivity values. These figures show that the values and spread of the objective functions of the optimal solutions for different catalyst packing density are close, however, the values of decision variables employed by those optimal solutions for different catalyst packing density are quite different, as

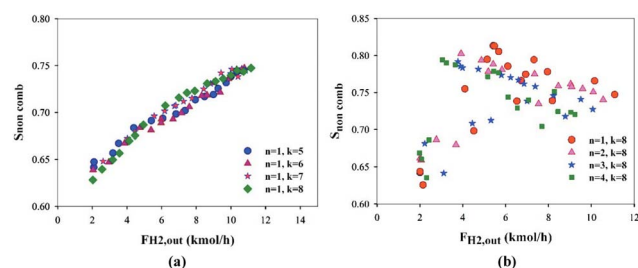


Figure 4. Effect of logistic function (a) k and (b) n parameter of on the solutions of binary-objective optimization ($F_{H_2,out}$ and $S_{noncomb}$) for MMR.

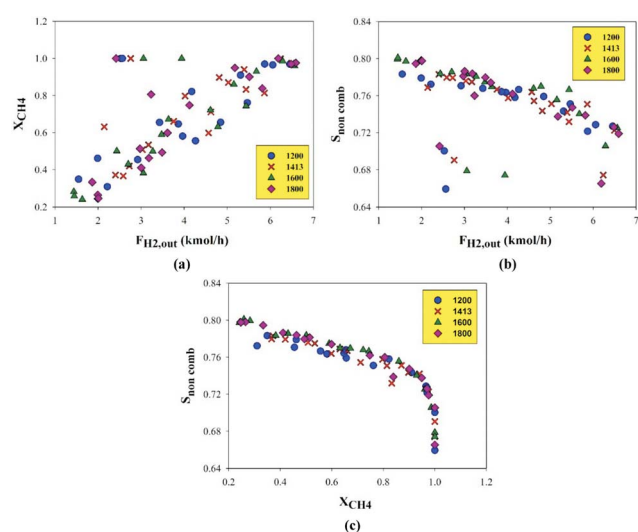


Figure 5. Objective functions of ternary-objective optimization ($F_{H_2,out}$, $S_{non comb}$ and X_{CH_4}) for ATR-MR (Figure legend: catalyst loading in kg_{cat}/m³).

shown in in Figure A1 of the Appendix.

For the ternary-objective optimization of MMR using different catalyst packing densities, the distributions of optimal solutions are shown in Figure 6. Compared to the optimal solution distributions of MR shown in Figure 5, the relationships among the three objective functions are the same based on the major part of the solutions. However, in Figure 6(a) there are more solutions hold a close to 100% methane conversion. In Figure 6(b), the relationship between hydrogen production and non-combustion selectivity of those solutions is reversed from other solutions. Same as the MR, the effects of catalyst packing density values and spread of the objective functions of the optimal solutions of MMR are not substan-

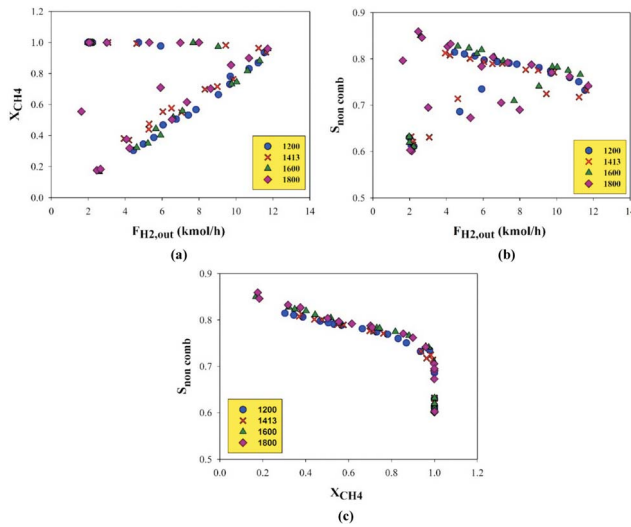


Figure 6. Objective functions of ternary-objective optimization ($F_{H_2,out}$, $S_{non\ comb}$ and X_{CH_4}) for ATR-MR (Figure legend: catalyst loading in kg_{cat}/m^3).

tial. Furthermore, also same as the MR, the values of decision variables employed by those optimal solutions for different catalyst packing density are quite different, as shown in in Figure A2 of the Appendix.

For in-depth comparison, the three optimal solutions MR and MMR of with the highest non-combustion selectivity of methane (TO-1), highest conversion of methane (TO-5) and highest hydrogen production (TO-9) are discussed. The objective functions and decision variables are listed in Table 3. The internal profiles of temperature and trans-membrane oxygen flux are shown in Figure 7. For TO-1 and TO-9, the target objective functions of MMR are higher than those MR, For TO-5, the conversions of MR and MMR are both equal to 1. The results in Table 3 and Figure 7 indicate that for both MR and MMR, the de-

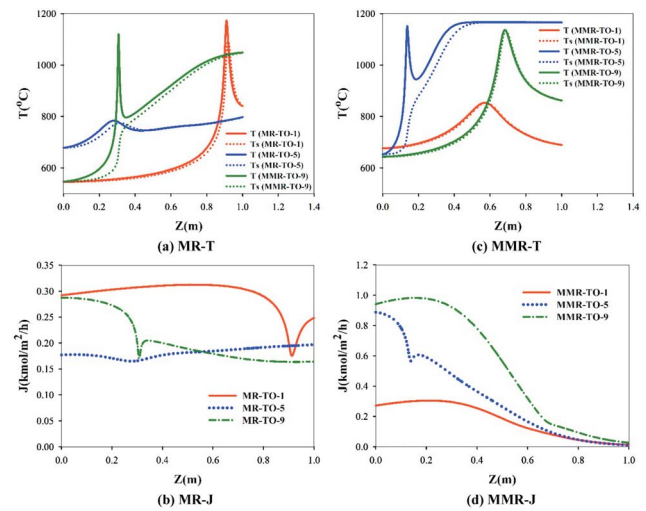


Figure 7. Internal profiles of some solutions of ternary-objective optimization ($F_{H_2,out}$, $S_{non\ comb}$ and X_{CH_4}) for MR (a-b) and MMR (c-d).

Table 3. Comparison of selected optimal solutions for MR and MMR¹

Optimal solutions	MR-TO-1	MMR-TO-1	MR-TO-5	MMR-TO-5	MR-TO-9	MMR-TO-9
Objective functions						
$F_{H_2,out}$ (kmol/h)	3.77	5.42	2.06	2.14	5.43	11.09
X_{CH_4}	0.518	0.381	1	1	0.753	0.876
$S_{non\ comb}$	0.772	0.813	0.684	0.626	0.735	0.747
Decision variables						
$F_{CH_4,in}$ (kmol/h)	2.87	4.76	1.00	1.07	2.99	5.00
$F_{H_2O,in} / F_{CH_4,in}$	1.32	2.33	1.97	3.47	2.08	3.65
$F_{S,in}$ (kmol/hr)	1.33	1.11	1.92	3.46	1.87	2.90
ΔP_{mem} (bar)	0.82	0.68	1	1.987	1	2
T_{in} (°C)	678	676	548	653	546	643
P_{in} (bar)	26.5	25.7	29.0	28.0	29.4	29.4

¹ The reactor lengths of MR and MMR are both 1 m.

cision variables and the internal profiles of the three optimal solutions are very different. From Figure 7(b) and (d), it is found that MMR can be operated under much higher oxygen fluxes without violating the maximum temperature constraint.

The ranges of objective function values of the optimal solutions for MR and MMR using different catalyst packing densities are summarized in Table 4. As has been shown in Figure 5 and Figure 6, the effect of catalyst packing density is not significant. For both the conversion and the non-combustion selectivity of methane, the value ranges of MR and MMR are pretty close but the value ranges of MMR are slightly wider than those of MR. However, significantly higher hydrogen production rates can be obtained using MMR than MR. Compared to MR, the highest hydrogen production rates of MMR are approximately twice as great.

5. Conclusions

For the autothermal reforming of methane to produce synthesis gas, this study has compared the performance of the packed bed membrane reactor (MR) and the packed bed mixed membrane reactor (MMR), which uses a mix of permeable membrane and non-permeable wall. Incorporation of the one-dimensional pseudo-homogeneous mathematical membrane reactor models and

the multi-objective optimization analysis provides a theoretically sound and practical approach for the evaluation and comparison of MR and MMR. Multiple solutions were obtained for the optimization of ternary objective functions, including conversion of methane, non-combustion selectivity of methane and hydrogen production rate, using NSGA-II for MR and MMR. For both the conversion and the selectivity of methane, the distributions of optimal solutions are close for MR and MMR. However, compared to MR, about 100% increase of the hydrogen production rate can be obtained by employing MMR. The internal profiles reveal that the optimal operations of MMR can be characterized by much higher trans-membrane oxygen fluxes compared to MR and the temperature can be kept lower than the maximum allowed temperature. This study has demonstrated the benefit of employing the packed bed mixed membrane reactors.

Acknowledgments

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Nomenclature

A_T cross-sectional area of the tubes, m^2

Table 4. Comparison of optimal performance for MR and MMR

Reactor performance	MR	MMR
Catalyst packing density (kg/m^3 bed)		1200
$F_{H_2,out}$ (kmol/hr)	1.548-6.454	2.079-11.537
$S_{non\ comb}$	0.659-0.783	0.611-0.814
X_{CH_4}	0.310-1.000	0.303-1.000
Catalyst packing density (kg/m^3 bed)		1413
$F_{H_2,out}$ (kmol/hr)	2.143-6.496	2.089-11.613
$S_{non\ comb}$	0.674-0.783	0.614-0.813
X_{CH_4}	0.371-1.000	0.370-1.000
Catalyst packing density (kg/m^3 bed)		1600
$F_{H_2,out}$ (kmol/hr)	1.438-6.563	1.980-11.282
$S_{non\ comb}$	0.674-0.801	0.610-0.850
X_{CH_4}	0.241-1.000	0.168-1.000
Catalyst packing density (kg/m^3 bed)		1800
$F_{H_2,out}$ (kmol/hr)	1.865-6.594	1.632-11.719
$S_{non\ comb}$	0.665-0.798	0.601-0.859
X_{CH_4}	0.246-1.000	0.318-1.000

B_0	geometric parameter, m^2
C_P	specific heat, $kJ/(kmol\ ^\circ C)$
D_{k,O_2}	Knudsen diffusivity of oxygen, m^2/s
d_T	tube diameter, m
f	value of logistic function
F_j	molar flow of component j , $kmol/h$
F_S	molar flow of shell-side gas, $kmol/h$
J_j	permeation flow of component j , $kmol/(h\ m^2)$
k	parameter of logistic function
L	tube length, m
MR	membrane reactor
MMR	mixed membrane reactor
n	number of sections of mixed membrane reactor
n_T	number of tubes
P	reactor (tube-side) pressure, bar
r_A	parameter of logistic function
r_i	reaction rate of reaction i , $kmol/(kg_{cat}\ h)$
R	gas constant, $m^3\text{-Pa}/(K\text{-kmol})$
$S_{non\ comb}$	non-combustion selectivity of methane
T_m	mean temperature of tube-side and shell-side, $^\circ C$
T	reactor (tube-side) temperature, $^\circ C$
T_S	shell-side temperature, $^\circ C$
U	overall heat transfer coefficient, $kJ/(h\ ^\circ C\ m^2)$
x_0	parameter of logistic function
X_{CH_4}	conversion of methane
y_i	reaction gas (tube-side) mole fraction of component i
z	axial coordinate, m
ΔP_{mem}	trans-membrane pressure difference, bar
δ	membrane thickness, m
ε	bed porosity
η_i	effectiveness factor of reaction i
μ_{O_2}	viscosity of oxygen, $Pa\ s$
ν_{ij}	stoichiometric coefficient with i reaction and j component
ρ_B	catalyst packing density, kg_{cat}/m^3

Subscript

i	reaction i
j	component j
in	inlet
max	maximum
out	outlet
S	shell-side

Appendix

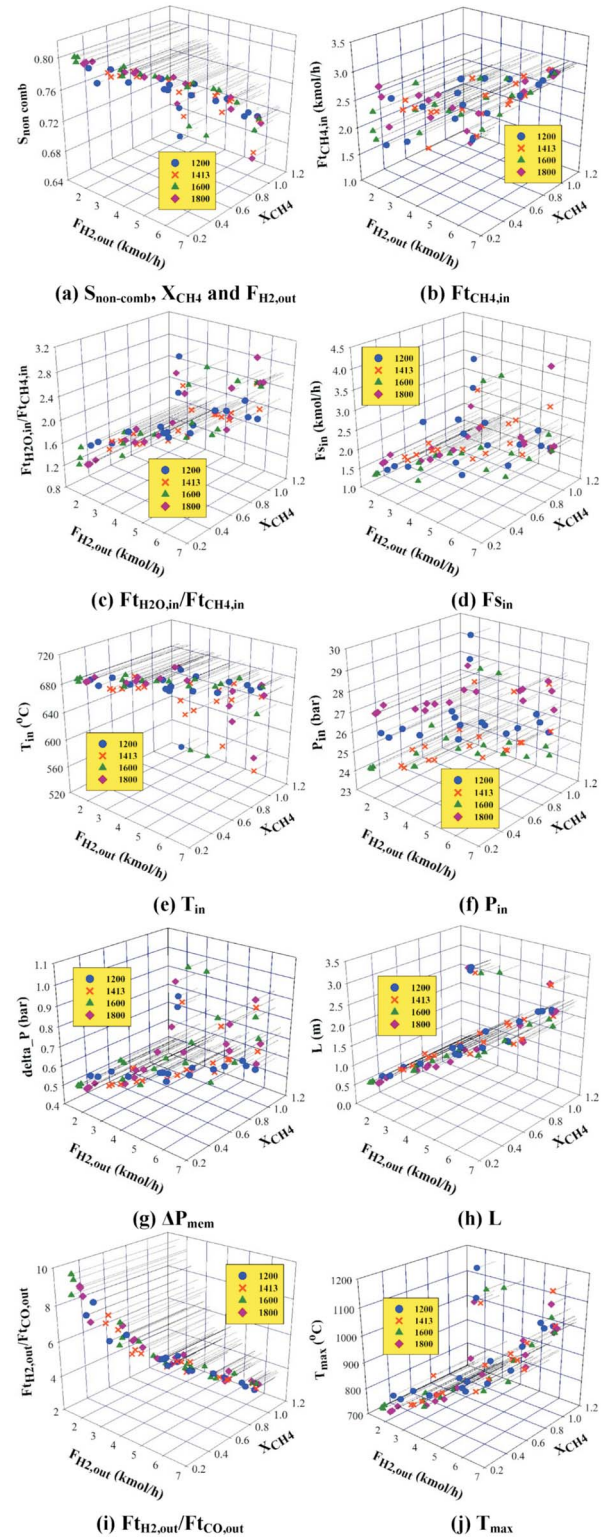


Figure A1. Objective functions (a), decision variables (b-h) and outcomes (i-j) of ternary optimization solutions for ATR-MR (Figure legend: catalyst loading in kg_{cat}/m^3).

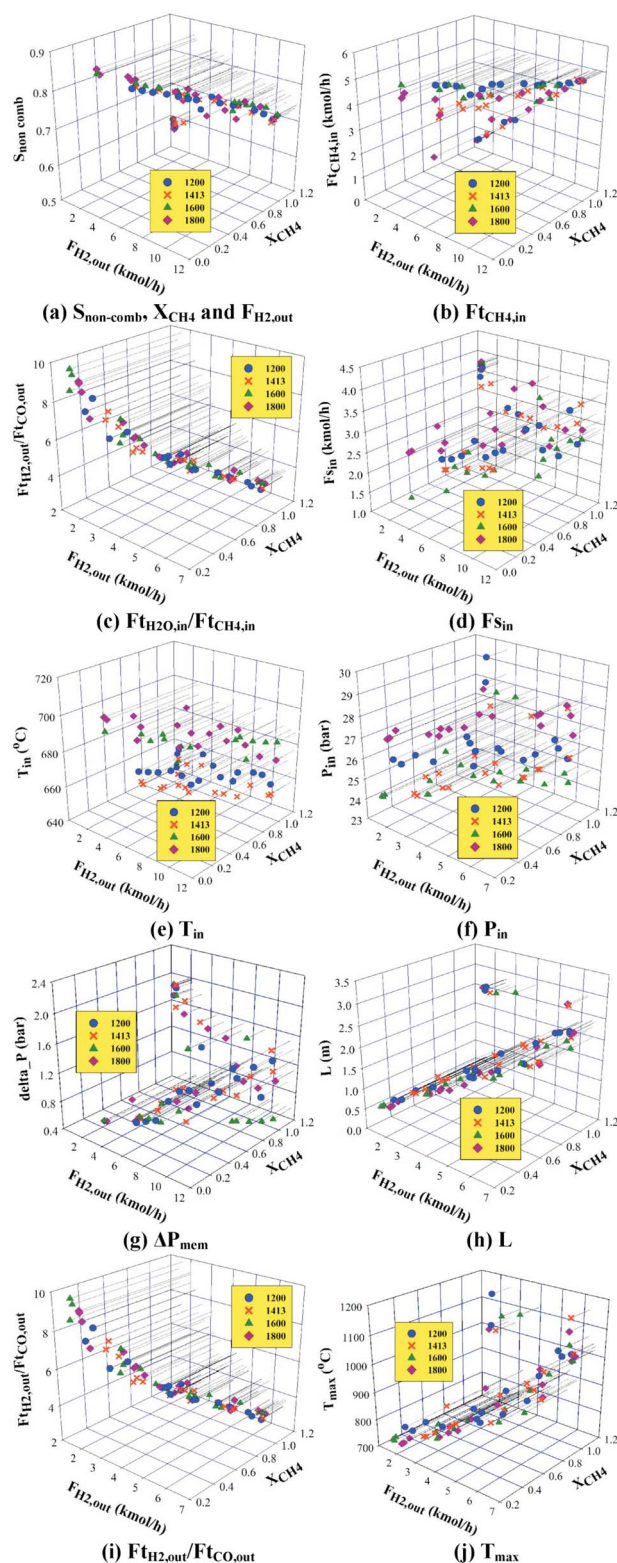


Figure A2. Objective functions (a), decision variables (b-h) and outcomes (i-j) of ternary optimization solutions for ATR-MMR ($k = 8$, $n = 1$) (Figure legend: catalyst loading in $\text{kg}_{\text{cat}}/\text{m}^3$).

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IMPORTANT DATES

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| • Abstract submission | March 1, 2019 |
| • Full paper submission | May 1, 2019 |
| • Acceptance notification | June 1, 2019 |
| • Final paper submission | June 31, 2019 |
| • Early bird registration | July 7, 2019 |
| • Hotel registration | July 7, 2019 |