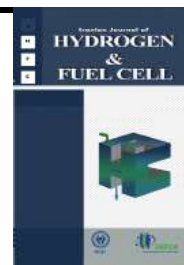


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Silica membrane performance for hydrogen separation from methanol steam reforming products: Assessment of different multistage membrane schemes

Abbas Aghaeinejad-Meybodi^{1,*}, Kamran Ghasemzadeh², Angelo Basile³

¹ Chemical Engineering Department, Faculty of Engineering, Urmia University, Urmia, Iran.

² Chemical Engineering Faculty, Urmia University of Technology, Urmia, Iran.

³ ITM-CNR, c/o University of Calabria, via P. Bucci cubo 17/C, 87036 Rende (CS), Italy

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Abstract

The aim of this work is a theoretical study of multistage silica membrane configurations for hydrogen purification by methanol steam reforming (MSR) products. Four membrane schemes including single permeator, CMC (continuous membrane column), ISMC ("in series" membrane cascade), and CRC (countercurrent recycle membrane cascade) were considered for this purpose. The modeling results showed that silica membranes have a high potential for high purity (more than 99.9%) hydrogen production. The lowest amounts of compressor duty and the required total membrane area were considered as the objective functions to select the optimal design and amount of hydrogen purification. A comparison of our simulation results of the different multistage membrane schemes showed the CRC configuration was more efficient than the other configurations. The modeling results show that increasing the retentate side pressure from 2 to 5 bar reduced the total silica membrane area for the CRC scheme by almost 13 times (30.67 and 2.37 cm² silica membrane area for a retentate side pressure of 2 and 5 bar, respectively).

1. Introduction

Today, hydrogen is known as a clean energy carrier for fuel cells. [1]. The stream of hydrogen that is used

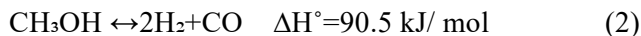
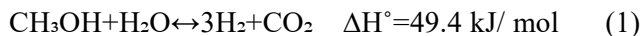
as feedstock for fuel cells must be of high purity. Also, the amount of carbon monoxide in it must have a concentration of less than 10 ppm because it is considered as a poison of the anodic catalyst [2].

*Corresponding Author's Fax: +98 4432773591

E-mail address: a.aghaeinejad@urmia.ac.ir

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Methanol has been proposed as a promising fuel to produce hydrogen, especially for fuel cell applications, because of its capability to transport, easy accessibility, and reaction simplicity [3, 4]. One of the important advantages of methanol fuels compared to hydrocarbon fuels is that it is free from sulfur, hence, does not require sulfur removal in the processing. It should be mentioned that the methane refining process is carried out at a high temperature of between 800-1000 °C, but methanol reforming requires only a relatively low temperature of 240-300 °C. When the methanol steam reforming (MSR) reaction is performed in a conventional reactor, the following chemical reactions occur [5, 6]:



Hydrogen should be purified to be used in the products of the MSR reaction for fuel cells. An appropriate solution to achieve high purity hydrogen is to use a membrane separation process. In recent decades, membrane separation processes have been considered by many researchers as an alternative to other conventional separation methods such as distillation, extraction, etc. [7, 8]. Among the different membranes, palladium (Pd) membranes have the highest selectivity for hydrogen. The disadvantages of Pd-based membranes include low hydrogen permeability and their high cost [9]. Therefore, cheaper silica membranes that have a higher permeance of hydrogen can be introduced as an alternative to Pd-based membranes. But, these membranes exhibit lower selectivity for hydrogen separation compared to Pd-based membranes [10]. Modeling of membrane separation processes is an essential step in analyzing and developing their performance at both industrial and pilot scales. Most membrane-based studies focus on single stage systems [11, 12]. Nonlinear ordinary differential equations (ODE) governing the membrane processes

of gases separation have been solved by many researchers by various techniques [11-14]. Shindo et al. examined multicomponent gas separation processes [12]. They considered five flow patterns, complete mixing, one-side mixing, cocurrent flow, countercurrent, and cross flow, for development of models. Then, they proposed an approximate solution for the governing differential equations. Kaldis et al. used the orthogonal collocation method to solve the mathematical model describing separation of a two-component gas mixture using hollow fiber membranes [15, 16]. In our previous work [14], the modified operational line method (MOLM) was presented to solve the governing differential equations of the separation process of a binary gas mixture using membranes. In this method, a linear relationship is considered between the molar fraction of components on the permeate and feed sides.

A convenient way to achieve high-purity products is through the use of multistage membrane schemes. Similar to the distillation column, multistage membrane schemes have been used in industrial applications to achieve high purity products [17]. The first studies on membrane multistage cascades were carried out by Pan and Habgood [18]. They considered six membrane stages in their studies. Stern et al. [19] investigated continuous membrane columns (CMC) and a single stage membrane with permeate recycle. Avgidou et al. [17] analyzed separation of LPG paraffins and olefins using multistage membrane configurations including CMC and countercurrent recycle cascade (CRC) schemes. Unfortunately, no scientific literature has been published on the design of an optimal membrane multistage system for hydrogen separation using silica membranes.

To the best of our knowledge, there is no point of view about hydrogen purification performance using multistage silica membrane configurations. It should be noted that only a limited number of modeling studies have been done on the analysis of silica membranes performance in hydrogen separation as well as in membrane reactors for hydrogen production [20-32].

In this work, the cocurrent flow pattern has been

considered for each stage of multistage membrane cascades for hydrogen separation from the products stream of the MSR reaction. Therefore, as a first step in this theoretical study, silica membrane performance in hydrogen separation from MSR products in a traditional reactor (TR) was examined for a single-stage permeation and three multistage membrane cascades, namely; countercurrent recycle cascade (CRC), "in series" membrane cascade (ISMC), and two-types of continuous membrane columns (CMC). The compressor duty and required membrane area for the desired hydrogen separation were calculated for each configuration. Finally, the effect of the retentate side pressure on membrane surface area and compressor duty for the optimum scheme was evaluated.

2. Model development

In this work, a mathematical model (black box model) was used to model the separation of a multicomponent gas stream in a membrane [12]. A schematic of a MSR reactor in series with an hydrogen selective membrane system is shown in Fig. 1. In this modeling study, the silica membrane is considered to be housed in the membrane separator. The gas flow from the reformer reactor (containing H_2 , CO , CO_2 , H_2O and CH_3OH) enters an ice bath

before being sent to the membrane stage. In this step, unreacted methanol and water are separated. The flow into the membrane stage includes H_2 , CO , and CO_2 . In this modeling study, a cocurrent flow pattern is considered for the membrane stage. The proposed model is suitable for computer programming. The suggested method for multi-component systems is appropriate for any number of components. The equations can also be developed for countercurrent and cross flow patterns.

The following assumptions were considered for the proposed model:

- Fick's law is considered for calculation of permeation rates.
- The permeability of each component in the gas stream is considered independent of pressure and equal to the value of the pure gas.
- The effective thickness of the membrane is assumed to be constant throughout the membrane.
- The concentration in each membrane side (retentate and permeate) is constant in the radial (permeation) direction.
- The pressure drops of the gas streams are considered zero for the permeate and retentate sides.
- The plug flow pattern is considered for the permeate and retentate sides.

In this model, mass balance equations for membrane systems were used. More details of the governing equations of this model can be found in the research

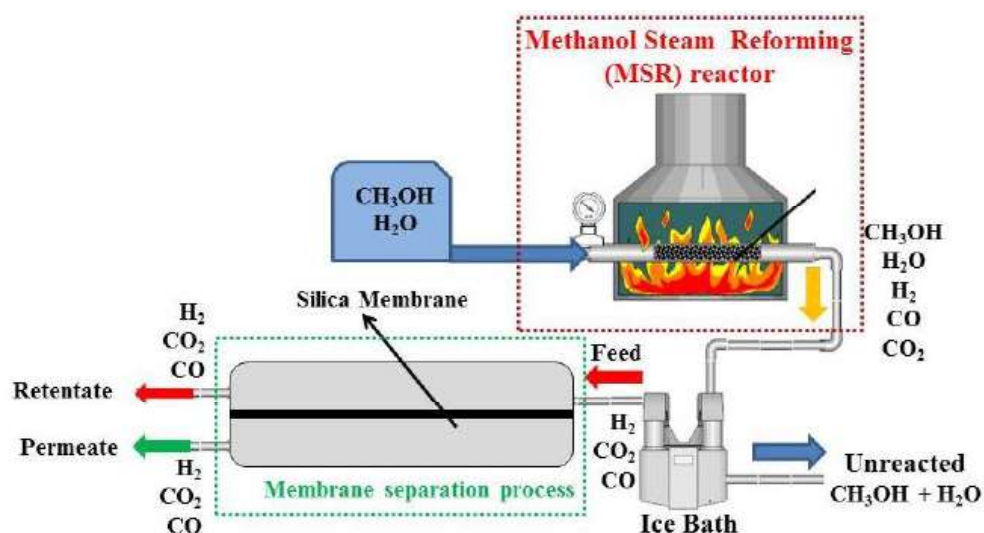


Fig.1. The scheme of the hybrid plant including a MSR reactor and a membrane separator

work of Shindo et al. [12].

2.1. Governing mathematical equations

In Fig. 2, a schematic of a membrane cell is shown with a cocurrent flow pattern. The equations of material balances (overall and partial) for the differential element (dA) are written as follows:

$$-dL = dV = dA \sum_{i=1}^n P_{e_i} (P_R x_i - P_p y_i) \quad (4)$$

$$-dx_i L_i = dy_i V = dA P_{e_i} (P_R x_i - P_p y_i) \quad (5)$$

where V is the permeate side molar flow rate (mol/s), L is the feed side molar flow rate (mol/s), P_{e_i} is the permeance of component i (mol/m².s.Pa), A is the membrane surface area (m²), P_p is the total permeate side pressure (Pa), P_R is the total retentate side pressure (Pa), y_i is the molar fractions of permeate side, and x_i is the molar fractions of the retentate side.

The next equations can be obtained by integrating Eqs. 4 and 5:

$$dx_i = \frac{-dA}{L} \left[P_{e_i} (P_R x_i - P_p y_i) - \sum_{i=1}^n P_{e_i} (P_R x_i - P_p y_i) \right] \quad (6)$$

$$y_i = \frac{x_{Fi} L_F - x_i L}{L_F - L} \quad V \neq 0 (i = 1, \dots, n-1) \quad (7)$$

$$y_i = \frac{P_{e_i} (P_R x_i - P_p y_i)}{\sum_{i=1}^n P_{e_i} (P_R x_i - P_p y_i)} \quad \text{at } V = 0 \quad (8)$$

where x_{Fi} is the feed composition and L_F is the feed

flow rate (mol/s).

Using Eqs. (1, 3-5), calculations of membrane separation processes can be performed for the multi-component mixture of gases for a cocurrent arrangement. To start the calculation loop, an initial guess should be considered for calculation of modeling problems.

The following initial guesses were used to start the loop of calculations:

$$\text{at } A=0, L=L_F, \text{ and } x=x_{Fi} \quad (9)$$

It should be noted that the local driving force is considered for mass transfer flux of each component through the membrane. Also, all components are permeable to the membrane. In general, the mass transfer equation for the permeation of any component through the membrane can be written as follows:

$$J_i = P_{e_i} (p_{i,\text{retentate}} - p_{i,\text{permeate}}) \quad (10)$$

where J_i is the permeating flux (mol/m².s) of component i , $p_{i,\text{permeate}}$ is the partial pressure (Pa) of component i in the permeate side, and $p_{i,\text{retentate}}$ is the partial pressure (Pa) for component i in the retentate side.

Cocurrent flow pattern equations were used in every stage in modeling the multistage membrane cascades (CMC, ISMC and CRC).

2.2. Numerical solution procedure

Boundary conditions should be used to solve the

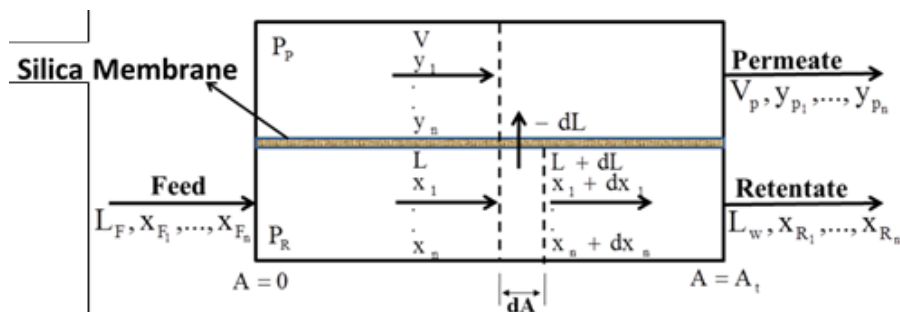


Fig.2. Schematic diagram of a cocurrent multicomponent gas permeator

governing differential equations of the membrane-based gas separation processes using numerical methods. For this purpose, the 4-order Runge-Kutta method has been used to solve the differential equations.

The no-mix condition was used in modeling of the multistage membrane cascades (CMC, and CRC) as follows:

“The compositions of streams are assumed equal to the feed composition at the feed point for CMC schemes. But, the compositions of head and tail streams that forming the feed of every stage are considered the same for multistage CRC schemes [17, pp 27].”

For each multistage membrane scheme, the compressor duty of process (Pa.mol/s) is calculated as follows:

$$\text{Compressor duty} = (p_p - p_R) \times (\text{total inlet flow rates to compressors}) \quad (11)$$

The feed stage was considered as the starting point for calculations in the CRC scheme. It should be noted that the calculations continue until the desired concentrations are achieved in the enrichment and stripping parts.

For the silica membrane, the compositions of the feed stream (Table 1) were considered based on experimental data from the outlet stream of a MSR reactor set-up obtained by the Institute on Membrane Technology–National Research Council (Italy). The values of hydrogen permeance of a silica membrane and ideal selectivities of components are summarized in Table 2.

Table1. The feed compositions of a MSR reactor for simulation at 2 bar and 280 °C.

Components	Mole fraction
H ₂	0.7420
CO	0.0433
CO ₂	0.2137
Total flow rate (mL/min)	51.40

Table 2. The values of ideal selectivities and permeance used in the simulation [33]

Membrane type	Permeability of H ₂	Selectivity (H ₂ /CO ₂)	Selectivity (H ₂ /CO)
Silica membrane	(mol/ s. m ² . Pa)	50	60

3.Results and Discussion

Validation of the model used in this study showed good agreement with the experimental data, details on the black box model is described in our previous work [13].

3.1.Evaluation of membrane schemes

Evaluations of four different membrane schemes with a cocurrent flow pattern in each stage have been considered in this study. These schemes are:

- (i) Single stage membrane
- (ii) “In series” membrane cascades (ISMC)
- (iii) Continuous membrane column (CMC)
- (iv) Countercurrent recycle membrane cascade (CRC)

To do this, the total surface area of the membranes and compressor duty are calculated for the same separation for each scheme using the discussed model.

For a comparative study between the single stage, CMC, ISMC and CRC configurations, the final composition of the retentate stream (for a stage cut of 0.65 in the first stage) for all of configurations and the permeate composition for the CRC, ISMC and CMC configurations were set at the same values. However, it should be mentioned that the objective function was considered as “ $y_p(\text{H}_2) > 0.99$ ” for the modeling and calculation of variables of the CRC scheme. According to this objective function and a stage cut of 0.65 in the first stage, the values of $y_{\text{H}_2}=0.9990$, $y_{\text{CO}_2}=0.0009$, and $y_{\text{CO}}=0.0001$ for the final permeate stream, and $x_{\text{H}_2}=0.4592$, $x_{\text{CO}_2}=0.4456$, and

$x_{CO}=0.0952$ for the final retentate product were obtained for the CRC scheme. Also, these values were used for modeling of the CMC, ISMC, and single stage configurations.

The model used a stage cut of 0.65 for a single stage system with a cocurrent flow pattern.

Fig. 3 shows the results of the modeling for a single stage membrane system with a cocurrent flow pattern. As shown in this figure, highly purity hydrogen can not be obtained in the permeate stream using a single stage membrane with a cocurrent flow pattern, making this system an inappropriate membrane system; therefore, the manipulation should be assigned using multistage membrane cascades. Highly pure hydrogen product can be obtained using CRC, ISMC, and CMC schemes. In the CMC(1) scheme (Fig. 4a) the entire permeate streams from the enriching and stripping sections are mixed and a fraction of this stream is compressed and used as feed in the enriching section. In the CMC(2) scheme (Fig. 4b) all of the permeate stream from the stripping section and a fraction of the enriching section are compressed and set as feed to the enriching section. Therefore, a multicomponent mixture can be theoretically separated into two components of any desired degree of purity in a CMC operated at a sufficiently high reflux ratio and low stage cut.

A straightforward method to increase the product purity is to connect a suitable number of permeators in series (ISMC), as shown in Fig. 5. In the enriching

section, the permeate stream from each permeator is recompressed and used as feed in the next permeator, while in the stripping section the retentate stream is fed to the next stage. In this study, to allow for a comparative study between all of the schemes, only the enriching section is considered for the ISMC configuration.

In a CRC cascade, the intermediate side streams are recycled (Fig. 6). Therefore, the CRC mode achieves significantly higher yields compared to non-recycle cascades. The recycle flow pattern is characteristic of a general countercurrent recycle cascade (CRC), that is, the tails stream (retentate) from stage $i+1$ and the heads stream (permeate) from stage $i-1$ combined to make up the feed to a general stage i .

Compositions of components and stream flow rates of the feed, residue, and permeate streams for CMC(1), CMC(2), ISMC, and CRC schemes are shown in Figs. 4, 5, and 6, respectively. Tables 3 and 4, show the simulation results using the CMC, ISMC and CRC schemes.

As shown in Tables 3 and 4, the CMC(1) and CMC(2) schemes are less efficient for the specified separation, requiring about 17.75 and 2.5 times more membrane area and 83 and 7.5 times more compressor duty, respectively, than the CRC scheme. It should be noted that although the required membrane area and compressor duty for the CRC scheme is greater than the ICMC scheme, the CRC scheme produces



Fig. 3. Modeling results of a single stage of silica membrane for hydrogen separation

Table. 3 Simulation results for CMC(1) and CMC(2) schemes.

	Membrane Area (cm ²)			Stage Cut		Compressor Duty (mol/atm.s ⁻¹)
	Stripping Sec.	Enriching Sec.	Total	Stripping Sec.	Enriching Sec.	
CMC(1)	24.58	529.86	544.44	0.65	0.9970	213×10 ⁻⁴
CMC(2)	24.58	49.74	74.32	0.65	0.9670	19.36×10 ⁻⁴

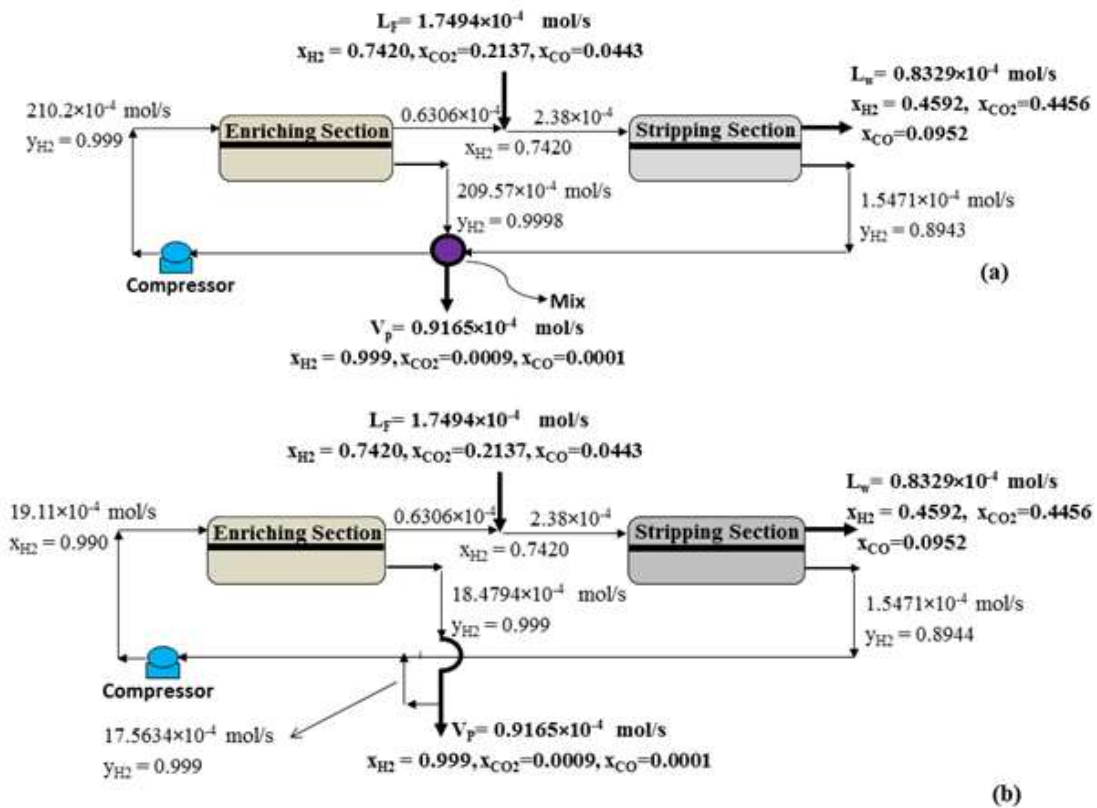


Fig. 4. Compositions and stream flow rates of components calculated for the a) CMC(1) and CMC(2) schemes for hydrogen separation using silica membrane

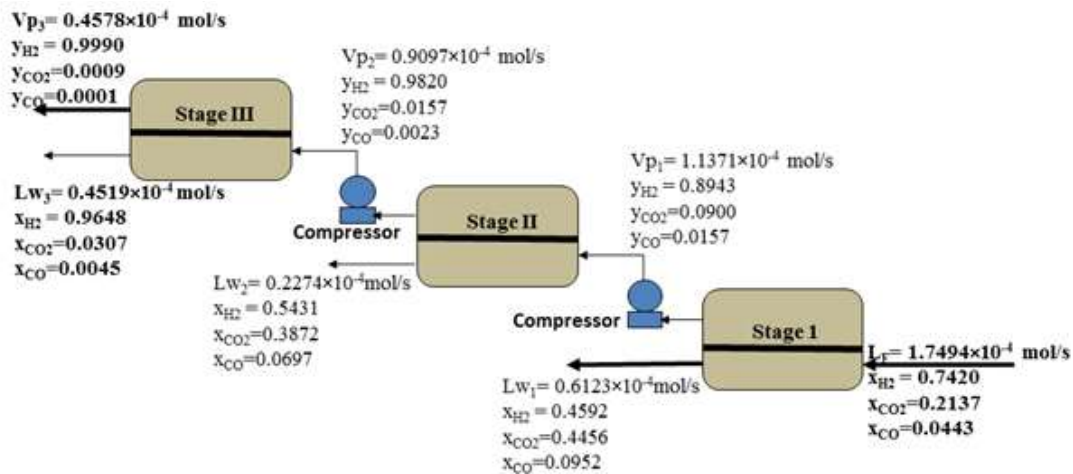


Fig. 5. Compositions and stream flow rates of components calculated for the ISMC scheme for hydrogen separation using a silica membrane.

about 2 times high purity product of hydrogen (more than 99.9%) than the ISMC configuration. A serious disadvantage of the ISMC scheme is the production of many side-products which cannot be utilized, and the desired final permeate and retentate products constitute only a small fraction of the feed. Therefore, it is evident that the non-recycle cascade

schemes involve the discharge and non-utilization of a considerable amount of the starting material. The loss increases sharply when high product purities are require; thus, many membrane stages are necessary. In cases where the starting material is of high value, recycling of the side-products is mandatory to improve product recovery.

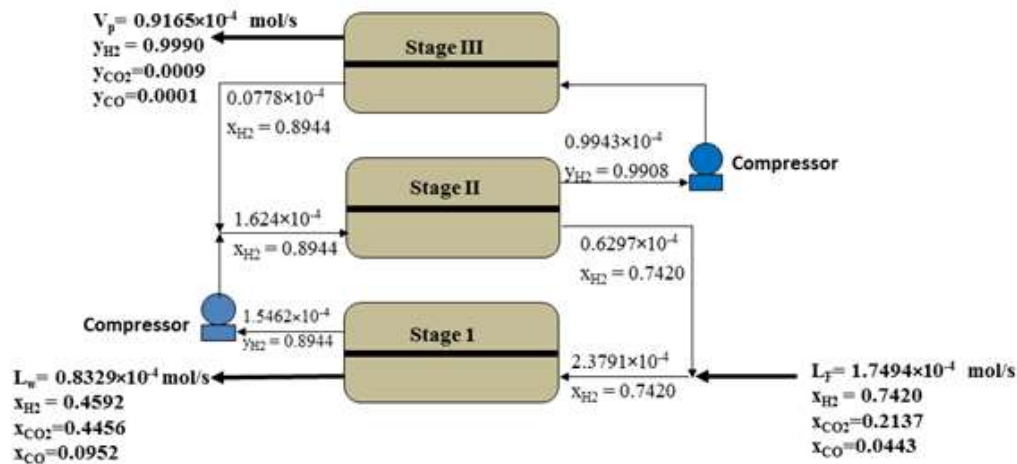


Fig. 6. Compositions and stream flow rates of components calculated for the CRC scheme for hydrogen separation using a silica membrane.

Table 4. Simulation results for CRC and ISMC schemes

Configuration	Stage	Membrane Area (cm ²)	Stage Cut	Total Membrane Area (cm ²)	Compressor Duty (mol/atm.s ⁻¹)	Separated Hydrogen with purity of 0.999 (%)
CRC	I(feed stage)	24.62	0.6500	30.67	2.57×10^{-4}	70%
	II	3.64	0.6120			
	III	2.41	0.9217			
ISMC	I(feed stage)	18.10	0.6500	23.60	2.07×10^{-4}	35%
	II	4.33	0.8000			
	III	1.20	0.5032			

According to the results of the modeling and simulations, the countercurrent recycle membrane cascade (CRC) is more efficient than the other configurations for hydrogen separation from MSR products to produce high purity hydrogen using a silica membrane.

The effect of retentate side pressure in the CRC scheme is presented in the next section.

3.2. Effect of retentate side pressure

A parameter that strongly affects the membrane performance is the feed/retentate side pressure. According to the gas separation driving force in molecular sieving silica membranes, increasing the retentate side pressure results in a higher H₂ pressure gradient with respect to CO. As shown in Fig. 7, the hydrogen molar fraction in the permeate stream increases when the retentate side pressure in each membrane stage increases, while the CO molar

fraction decreases when the retentate side pressure increases in each stage.

As shown in Fig. 7, a purity of about 98% for hydrogen in the permeate product can be obtained in an operating pressure of 5 bar in the single stage mode. The effect of retentate side pressure on the membrane area of each stage is presented in Fig. 8. According to this figure, the required membrane area for each membrane stage decreases as the retentate side pressure increases because of the enhancement of the separation driving force. The modeling results show that by increasing retentate side pressure from 2 to 5 bar, the total silica membrane area for the CRC scheme will be reduced almost 13 times (30.67 and 2.37 cm² silica membrane area for retentate side pressure of 2 and 5 bar, respectively).

The effect of ratio of feed to permeate pressure on the total membrane area and compressor duty is shown in Fig. 9.

A graphical representation confirms an optimum

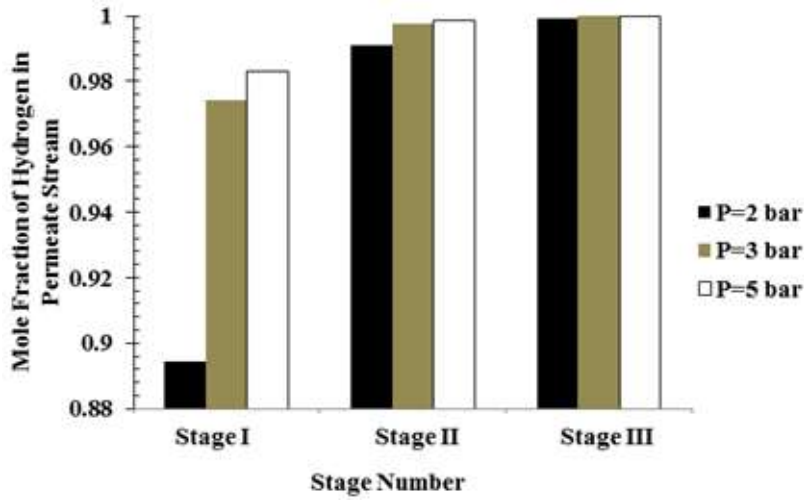


Fig. 7. The H₂ molar fraction in the permeate side of each membrane stage at different absolute retentate side pressure

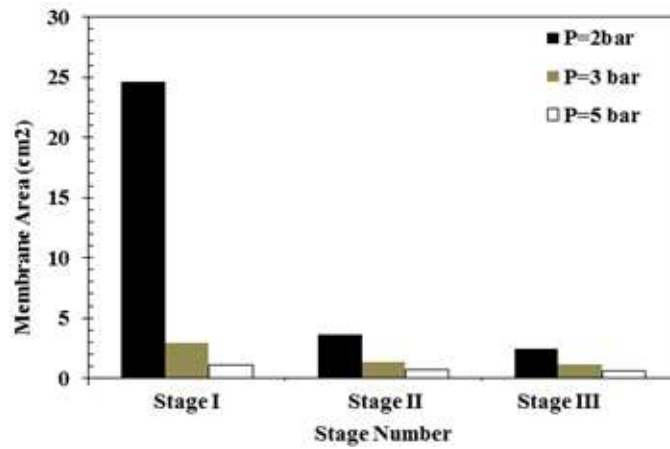


Fig. 8. Effect of retentate side pressure on required silica membrane area for each stage of the CRC scheme

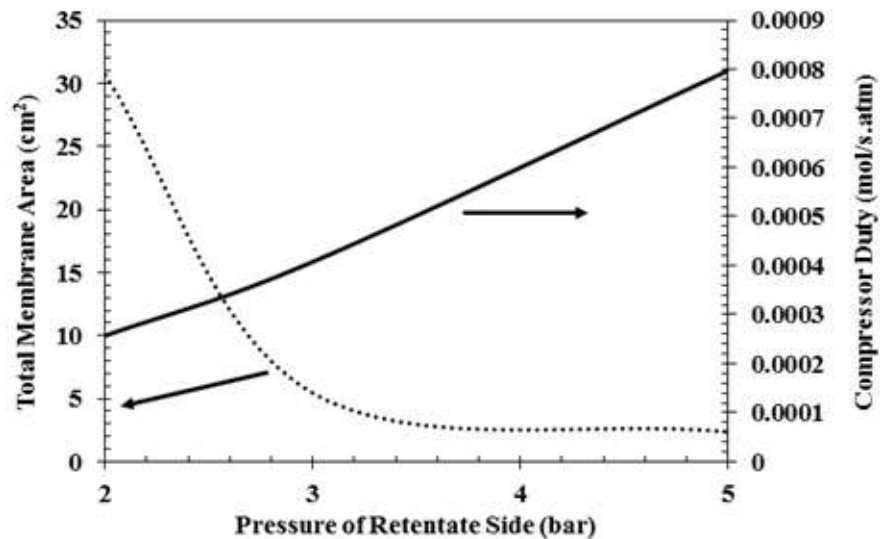


Fig. 9. Effect of feed to permeate pressure ratio on total membrane area and compressor duty in CRC scheme

pressure ratio. As shown in this figure, the total membrane area decreases with increasing pressure ratio, while the compressor duty becomes lower when a smaller pressure ratio is used. The optimum economic condition is located where the sum of membrane area cost and compressing cost becomes minimum.

4. Conclusion

In this work, four types of single/multi stage silica membrane configurations were examined, i.e. single stage membrane, CMC, ISMC, and CRC, assuming cocurrent flow pattern in each stage for hydrogen separation from MSR products to produce high purity hydrogen. The results showed that one-stage membrane systems were not suitable for hydrogen separation. For this purpose, the use of multi-stage membrane cascades is proposed for hydrogen purification from MSR products. Comparative studies between multi-stage cascades showed that the CRC scheme is the most efficient for specified hydrogen separation, requiring about 17.75 and 2.5 times less membrane area and 83 and 75 times less compressor duty in the CMC(1) and CMC(2) schemes, respectively. Moreover, the modeling results indicated that the silica membrane gave a notable performance in producing high purity hydrogen (more than 99.9%). In addition, simulation results showed that the CRC configuration is more efficient than the other configurations. By increasing retentate side pressure, the total membrane area decreases, while the compressor duty increases with the increase of retentate pressure. However, it should be noted that from economic and optimum design viewpoints, both the membrane area and compressing duty must be minimized.

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