



# Conceptual process design and simulation of membrane systems for integrated natural gas dehydration and sweetening



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## ABSTRACT

Subsea natural gas processing attracts increased interest due to the smaller environmental footprint. Natural gas (NG) dehydration and sweetening are the main processing steps to avoid pipeline plugging and corrosion caused by the presence of water and CO<sub>2</sub>. Triethylene glycol (TEG) and amine absorption are the commercial technologies for these applications. However, membrane technology is considered as promising solutions for alternative subsea gas processing technologies, which provides unmanned operations without the requirements for rapidly periodical maintenance. In this work, a hybrid membrane process was designed for integrated dehydration and sweetening of a saturated natural gas containing 10 mol% CO<sub>2</sub>, and the process operating parameters such as inter-stage feed and permeate pressures are investigated. The simulation results indicated that the optimal permeate pressure in the 2nd -stage unit is 4 bar, and the optimal 3rd-stage feed and permeate pressures are 15 bar and 2 bar, respectively. The minimum specific cost of <math>2.71 \times 10^{-3}</math> \$/m<sup>3</sup> sweet natural gas was estimated to achieve the separation requirement of <math>< 2.5</math> mol% CO<sub>2</sub> in purified NG together with captured high purity CO<sub>2</sub> (> 95 mol%) for enhanced gas recovery. However, due to the relatively low water selectivity of the dehydration membranes at high pressure of 60 bar used in the simulation, the hydrocarbon loss is still quite higher. Thus, advanced membranes with high H<sub>2</sub>O/CH<sub>4</sub> selectivity at high pressure should be pursued to promote the application of the designed membrane system for subsea natural gas dehydration and sweetening.

## 1. Introduction

Natural gas (NG) is considered as one of the less carbon-intensive energies, and the demand of has been increased in the recent decade [1]. However, natural gas produced from gas wells must be dehydrated and sweetened before entering distribution pipelines as water-saturated sour NG flows in a gas pipeline can cause serious issues related to the increase of pressure drop, the reduction of gas flow or the plugging of pipeline, and severe corrosions. Thus, natural gas dehydration and sweetening are mandatory in any natural gas plant. Subsea natural gas processing can avoid the offshore and on-shore treatment, and directly transport produced hydrocarbons (HCs) from reservoir to market, and also improves the operational safety regarding fire and explosions [2]. However, implementation of unmanned operations is crucial in subsea natural gas processing without the requirements for rapidly periodical maintenance. Although glycol (e.g., triethylene glycol (TEG) or monoethylene glycol (MEG)) absorption is the state-of-the-art technology for natural gas dehydration, it faces increasing environmental restrictions since the separation units can emit hazardous volatile organic

compounds (VOCs), especially BTEX (benzene, toluene, ethylbenzene, and xylene), due to the decomposition of glycol from regeneration reboiler [3]. Moreover, glycol system operation is also complex and requires solvent storage, replacement and disposal, which increases operating cost due to the factors such as large heating requirements and the need for continuously attention and maintenance. Membrane contactor has advantages of high interface between gas and liquid phases and no flooding or foaming, which was reported to be feasible for natural gas dehydration [2,4]. However, solvent regeneration/recycling is an energy-intensive process. Membrane gas or vapor separation systems are often compact with a small footprint, and without moving parts and the need of on-site attention when operated in remote/subsea locations [2,5,6], which enables a higher potential application in subsea natural gas processing comparing to absorption technology. Different types of membranes tested for H<sub>2</sub>O/CH<sub>4</sub> separation in natural gas dehydration were reviewed in the literature [2,6], and very high H<sub>2</sub>O/CH<sub>4</sub> selectivity (several thousands) for some membranes such as polyimide (Kapton) cellulose acetate, and Nafion® 117 were reported. It should be noted that the reported performances (both H<sub>2</sub>O permeability

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and H<sub>2</sub>O/CH<sub>4</sub> selectivity) were mostly obtained at a low membrane operating pressure. Among them, the commercial polymeric membranes of PEEK-SEP™ (Air Liquide) used for natural gas dehydration has low BTEX emission and less required pre-treatment. Moreover, the Pebax® thin film composite membranes developed by MTR Inc. presented good performance for H<sub>2</sub>O/CH<sub>4</sub> separation at a low feed pressure of 2 bar [6], but a reasonable water permeance of 0.055 m<sup>3</sup> (STP)/(m<sup>2</sup>·h·bar) and a low H<sub>2</sub>O/CH<sub>4</sub> selectivity of 47 at 30–60 bar were reported from their field testing [7]. Thus, membrane performance (especially H<sub>2</sub>O/CH<sub>4</sub> selectivity) will be significantly influenced in the real natural gas processes compared to lab-scale synthetic gas permeation testing. Although many polymeric and inorganic membranes were developed for H<sub>2</sub>O/CH<sub>4</sub> separation, the process feasibility of such membrane systems has not been conducted to document the potential for natural gas dehydration at large-scale, especially to achieve the specific low water content (e.g., dew point of  $-40\text{ }^{\circ}\text{C}$ ) in natural gas. Moreover, natural gas sweetening is also required to remove CO<sub>2</sub> and reach the content of  $< 2.5\text{ mol}\%$  in the product gas [8]. Although chemical (amine-based) absorption is the commercial technology for CO<sub>2</sub> removal from natural gas plants, the energy consumption and environmental issues with a second pollution for amine-based absorption directs to the development of advanced separation technologies. Gas separation membrane is considered as a promising technology for this application even though it has only 5% of the market today. However, the commercially available polymeric membranes (e.g., cellulose acetate (CA), cellulose triacetate (CTA) and polyimide (PI)) used for CO<sub>2</sub> removal from natural gas have low separation performances related to membrane compaction and plasticization at high-pressure operations [9]. These issues lead to the increase of capital cost due to the larger required membrane surface area together with the reduced material lifetime, which indicates the need of development of advanced membranes materials. Matrimid hollow fiber membranes were reported to present a good performance with CO<sub>2</sub>/CH<sub>4</sub> selectivity over 34 at 75 °C [10]. Recently, the fixed-site-carrier (FSC) membranes have been reported for CO<sub>2</sub> removal from natural gas [9,11]. This type membrane showed a good CO<sub>2</sub>/CH<sub>4</sub> separation performance at moderate pressures [9,11]. However, the challenge on getting good performance at higher pressure  $> 40\text{ bar}$  remains, which can be addressed by using carbon membranes due to the strong mechanical strength for tolerating high pressure operations. Carbon membranes with graphitic or turbostratic structure are usually made from polyimides (PI) and cellulose derivatives [12–16]. The cellulosic-derived hollow fiber carbon membranes presented a good selectivity of CO<sub>2</sub>/CH<sub>4</sub> with a lower CO<sub>2</sub> permeance (usually  $< 30\text{ GPU}$ ) due to a symmetric structure with a thickness of ca. 30 μm [16–24]. Recently, carbon membranes made from polyimide precursors were tested at high pressure of up to 120 bar for CO<sub>2</sub>/CH<sub>4</sub> separation [15]. They reported a CO<sub>2</sub> permeance of 100 GPU (i.e., 0.28 m<sup>3</sup>(STP)/(m<sup>2</sup>·h·bar)) and a CO<sub>2</sub>/CH<sub>4</sub> separation factor of 60 based on a 50% CO<sub>2</sub>/50% CH<sub>4</sub> gas mixture permeation testing, which may have the potential for natural gas sweetening. Thus, the techno-economic feasibility analysis was conducted to document the advance of the hybrid membrane process for subsea natural gas dehydration and sweetening in this work. Moreover, process operating parameters were investigated by HYSYS simulation to identify the optimal condition for achieving minimum NG processing cost. The sensitivity analysis on membrane performance especially selectivity was also conducted to document the required material performances to reach a low HC loss.

## 2. Methods

### 2.1. Process description and design

The sour natural gas produced from gas/oil reservoirs contains different hydrocarbons, and the impurities of water, H<sub>2</sub>S, CO<sub>2</sub> and helium. Among them, water and acid gases need to be removed to meet gas grid specifications. A hybrid membrane system for natural gas

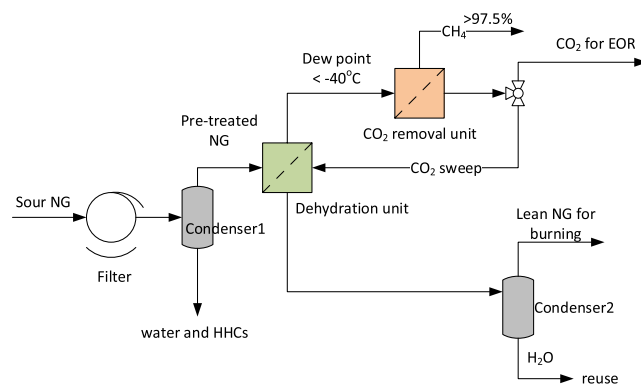


Fig. 1. Illustration of a hybrid membrane process for integrated natural gas dehydration and sweetening.

dehydration and sweetening, illustrated in Fig. 1, was designed to reach the low impurity requirements of water (dew point  $< -40\text{ }^{\circ}\text{C}$ ) and CO<sub>2</sub> ( $< 2.5\text{ mol}\%$ ) in the sweet natural gas. The designed process uses water-selective membranes (dehydration unit) to remove water vapor from the pre-treated natural gas and followed by carbon membrane systems to remove CO<sub>2</sub>. The sour natural gas was initially filtered to remove any trapped liquids, aerosols, particles and sands, and then enters into the water-selective membrane modules (using MTR Pebax® membranes [7]), where H<sub>2</sub>O, and part of CO<sub>2</sub> and HCs permeate through the membranes. The dried gases containing less permeable components (mainly HCs and CO<sub>2</sub>) remain in the retentate at the processing pressure (the pressure drop in the feed side of dehydration membrane unit is ignored), which are then sent to the subsequent carbon membrane units to remove CO<sub>2</sub>, and produce sweet natural gas (i.e., HC content of  $> 97.5\text{ mol}\%$ ) in the retentate as the product gas. Part of the captured CO<sub>2</sub> from the CO<sub>2</sub> removal unit is used as sweep gas in the dehydration unit to provide higher driving force for water permeation and reduce HC loss. Moreover, using sweep gas in the membrane dehydration unit can also avoid water condensation inside the membrane unit by keeping all the permeated water molecules in the vapor phase. The permeate stream is then condensed to recover water, and the lean NG in gas phase will be burned out before venting (this is the major HC loss). Moreover, the rest of the captured CO<sub>2</sub> will be further compressed and injected back to gas reservoirs for enhanced gas recovery (EGR)-this is to displace natural gas by injecting CO<sub>2</sub> in the supercritical phase. Implementation of CO<sub>2</sub>-EGR can potentially enhance the recovery of natural gas and store CO<sub>2</sub> underground to reduce CO<sub>2</sub> emissions [25,26]. It should be noted that the influence of CO<sub>2</sub> for EGR is not clarified [27], but a high purity of CO<sub>2</sub>  $> 95\%$  is usually required for re-compression and re-injection back to gas reservoirs.

### 2.2. Simulation basis

The designed hybrid membrane system was used to evaluate the technology feasibility of the combined natural gas dehydration and sweetening from a  $5 \times 10^5\text{ m}^3(\text{STP})/\text{h}$  natural gas plant (10 mol% CO<sub>2</sub> in water-saturated natural gas, see the detailed gas composition in Table 1) at 80 °C. The CH<sub>4</sub> purity of 97.5 mol%, the CO<sub>2</sub> purity of 95 mol% and the gas dew point of  $< -40\text{ }^{\circ}\text{C}$  were defined as the separation targets given in Table 2, the HC loss was not specified. The gas separation performances of Pebax membranes [7] and carbon membranes [15] reported in the literature were used as the simulation basis in this work. It should be noted that the C<sub>2</sub>-C<sub>5</sub> + permeances of carbon membranes were estimated to 0.001 mol(STP)/(m<sup>2</sup>·h·kPa) as described in Table 1. Moreover, all gas permeances were assumed to be independent of feed gas composition and process condition. The pressure drops of coolers and heat exchangers were neglected in all the simulation scenarios, and a counter-current flow pattern was used for all

**Table 1**  
Feed gas composition and gas permeance of membranes.

Component	Feed NG composition (mol %)	Gas permeance of Pebax membranes (mol(STP)/(m <sup>2</sup> ·h·kPa)) [7]	Gas permeance of carbon membranes (mol(STP)/(m <sup>2</sup> ·h·kPa)) [15]
CO <sub>2</sub>	10 <sup>§</sup>	0.069	0.095
Methane	Balanced	0.023	0.002
Ethane	7	0.046	0.001 <sup>#</sup>
Propane	3	0.071	
Butane	1.9	0.075	
C <sub>5</sub> +	0.1	0.116	
H <sub>2</sub> O	0.78	1.217	0.95 <sup>*</sup>

<sup>§</sup> A 10 mol% CO<sub>2</sub> in NG was selected as a case study in this work.

<sup>#</sup> Estimated gas permeance for hydrocarbons of C<sub>2</sub>-C<sub>5</sub> +.

<sup>\*</sup> Water permeance is assumed to be 10X of CO<sub>2</sub> for carbon membranes.

**Table 2**  
The simulation basis of a hybrid membrane process for natural gas dehydration and sweetening.

Parameters	Values
NG feed flow rate, m <sup>3</sup> (STP)/h	5 × 10 <sup>5</sup>
Raw NG composition (mol%)	See table 1
NG pressure (bar)	60
Temperature (°C)	80
HC purity of sweet NG (mol%)	> 97.5
Captured CO <sub>2</sub> purity (mol%)	> 95
Water content in dried NG (dew point, °C)	< -40
Membrane area (m <sup>2</sup> )	Calculated
Compressor power demand (kW)	Calculated
Compressor adiabatic efficiency (%)	75

three membrane units. The adiabatic efficiency of 75% was applied for the estimation of power demands for compressors.

Fig. 2 illustrates the simulation workflow of the hybrid membrane system for integrated NG dehydration and sweetening. The input variables such as flow configuration, membrane area, feed flow rate, temperature, etc. were set in the HYSYS simulation environment. The design variables (e.g., feed and permeate gas pressures, CO<sub>2</sub> recycling ratio, etc.) were adjusted to investigate their influences on natural gas processing cost. The membrane areas in the dehydration and CO<sub>2</sub> removal units were adjusted to achieve the separation targets defined in Table 2. The simulation results of the required membrane area, the compressor power, and the purified NG flow rate were obtained for estimation of the NG processing cost.

### 2.3. Cost model

Cost estimations for compressors and membrane units were conducted based on the cost model reported by Chu et al. [28]. The axial compressors (450–3000 kW) with stainless steel materials were selected for high-pressure operation. The membrane cost of \$50 per m<sup>2</sup> was used to estimate the Pebax and carbon membrane unit cost, and a membrane lifetime of 5 years was also applied [23,28]. It should be noted that carbon membrane cost may be higher compared to polymeric membranes, and sensitivity analysis on different membrane materials cost was also investigated. Other equipment such as heat exchanger, cooler and mixer were not included as those unit costs are expected to be much lower compared to that of the compressors and the membrane units. An interest of 7% was used for the estimation of annual capital related cost (CRC) [29]. The electricity price of 0.05 \$/kWh was applied to estimate the operating expenditure (OPEX) of the major driving equipment of compressors, and labor cost was not included in this work. The specific NG processing cost (C<sup>S</sup>, \$/m<sup>3</sup> sweet NG) [28] is estimated by:

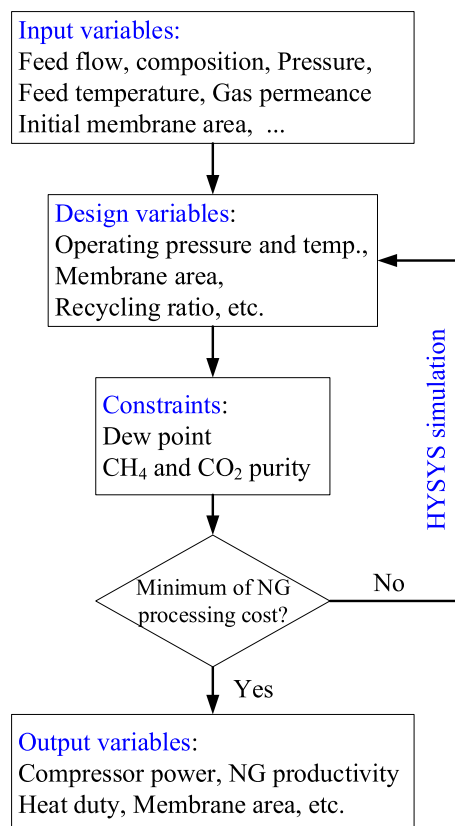


Fig. 2. Illustration of simulation workflow.

$$C^S = \frac{CRC + OPEX}{\text{Annual Total sweet NG production}} \quad (1)$$

The C<sup>S</sup> is used as the objective function for process optimization.

### 2.4. Process parametric study

Process simulation was conducted by HYSYS integrated with ChemBrane (the customized membrane unit [23,30]), and the flow sheet is shown in Fig. 3. The sour NG is cooled down and sent to a gas-liquid separator (Sep-1 in Fig. 3) to knock out condensed water and heavy hydrocarbons. The pre-treated NG is further fed to the 1st-stage membrane dehydration unit to remove most of water and control the dew point of the dehydrated gas (R1) to < -40 °C using the adjuster of ADJ-1. The stream R1 is then sent to the two-stage carbon membrane units for CO<sub>2</sub> removal. The HC purity in the retentate (R2) of the 2nd-stage (carbon) membrane unit should achieve 97.5 mol% by adjusting membrane area and the permeate pressure (p<sub>2</sub>). The stream P2 is then compressed and fed into the 3rd-stage (carbon) membrane unit to produce high purity CO<sub>2</sub> in the permeate side (> 95 mol%) where feed pressure (P<sub>3</sub>) and permeate pressure (p<sub>3</sub>) were varied. Part of the captured CO<sub>2</sub> (CO<sub>2</sub>-Sweep) is recycled back to the sweep side of the dehydration unit, and the rest CO<sub>2</sub> (Stream CO<sub>2</sub>-EGR) is re-compressed for enhanced gas recovery. The process operating parameters such as the permeate pressures of the streams P2 and P3, the feed pressure of the stream F3 as well as the recycling ratio of capture CO<sub>2</sub> are identified as the major variables that may influence the required power consumption for compressors, and membrane areas. Thus, the process parametric study by tuning the parameters listed in Table 3 was conducted to identify the optimal operating condition to reach the minimum specific NG processing cost.

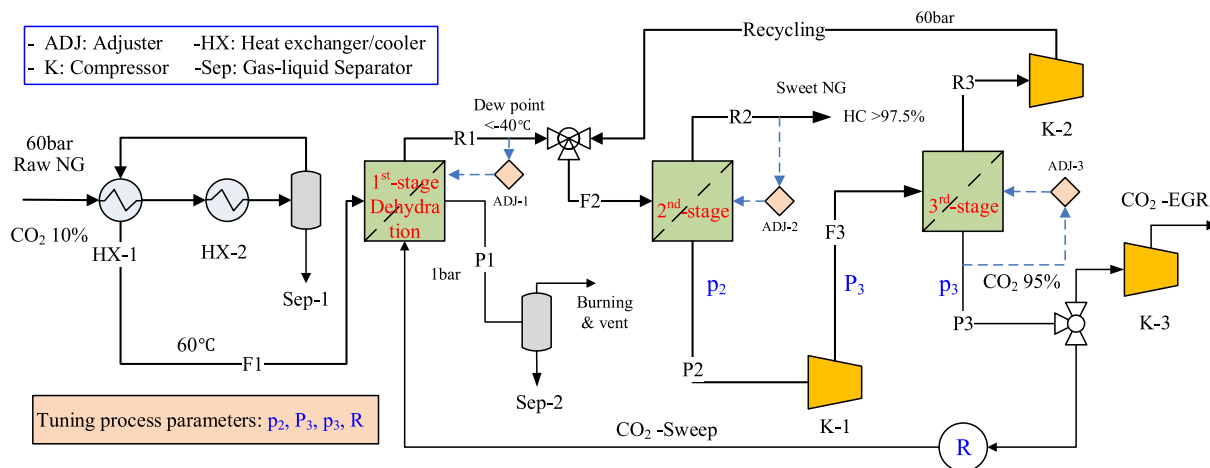


Fig. 3. Process flow diagram of a hybrid membrane system for natural gas dehydration and sweetening.

3. Results and discussion

The designed integrated NG dehydration and sweetening system was used for process simulation by HYSYS (see the process flow diagram in the supporting information) and cost estimation, and the influences of process operating parameters (listed in Table 3) on the NG processing cost were investigated.

3.1. Effect of the 2nd-stage permeate pressure

For the scenario A, the permeate pressure of the 2nd-stage membrane unit was varied from 1 to 10 bar while the other parameters were kept constant. The proposed membrane system, according to the process simulations, is technically feasible to produce dried natural gas (dew point < -40 °C at 60 bar) with high HC purity of > 97.5 mol% by adjusting the membrane areas of the dehydration and CO<sub>2</sub> removal units. The key output variables-compressor power demands and membrane areas are listed in Table 4. It can be found that the 2nd-stage membrane area is much larger compared to that of the dehydration unit and the 3rd-stage. The annual CRC and OPEX were estimated based on the cost model described in Section 2.3, and Fig. 4 shows the simulation results. It can be found that the minimum specific NG processing cost is  $3.06 \times 10^{-3}$  \$/m<sup>3</sup> at the optimal permeate pressure ( $p_{2,opt}$ ) of 4 bar, corresponding to a pressure ratio of 15. One should expect that a lower permeate pressure ( $p_2$ ) increases driving force and reduces the required membrane area of the 2nd-stage. However, the power demand for the re-compression of the stream P2 before feeding to the 3rd-stage membrane system will be higher. Based on the principle of increase of entropy, power demand is decreased by reducing re-compression of the permeate gas so that the entropy production can be minimized as reported by Magnanelli et al. [31]. Therefore, maintaining a certain pressure in the permeate side is favorable for natural gas processing with a lower cost. however, higher permeate pressure of > 4 bar

Table 3 Process parametric study.

Scenario	The 2nd-stage permeate pressure, $p_2$ (bar)	The 3rd-stage feed pressure, $P_3$ (bar)	The 3rd-stage permeate pressure, $p_3$ (bar)	The recycling ratio of captured CO <sub>2</sub> , R (-)
A	1–10 bar	60	1	1
B	$p_{2,opt}$ (Optimized from the scenario A)	$[p_{2,opt}, 60]$	1	1
C	$p_{2,opt}$	$P_{3,opt}$ (Optimized from the scenario B)	$[1, P_{3,opt}]$	1
D	$p_{2,opt}$	$P_{3,opt}$	$p_{3,opt}$ (Optimized from the scenario C)	0–1

Table 4 The key output variables of the simulation results.

2nd-stage permeate pressure, bar	Total compressor power demand, kW	2nd-stage membrane area, m <sup>2</sup>	3rd-stage membrane area, m <sup>2</sup>
1	$1.1 \times 10^4$	$5.2 \times 10^4$	$7.1 \times 10^3$
2	$9.1 \times 10^3$	$6.5 \times 10^4$	$6.9 \times 10^3$
3	$8.6 \times 10^3$	$8.1 \times 10^4$	$6.9 \times 10^3$
4	$8.2 \times 10^3$	$9.6 \times 10^4$	$6.6 \times 10^3$
5	$8.0 \times 10^3$	$1.2 \times 10^5$	$6.2 \times 10^3$
6	$7.8 \times 10^3$	$1.4 \times 10^5$	$5.7 \times 10^3$
7	$7.7 \times 10^3$	$1.6 \times 10^5$	$5.2 \times 10^3$
8	$7.6 \times 10^3$	$1.8 \times 10^5$	$4.8 \times 10^3$
9	$7.4 \times 10^3$	$2.1 \times 10^5$	$4.0 \times 10^3$
10	$7.3 \times 10^3$	$2.3 \times 10^5$	$3.2 \times 10^3$

The dehydration membrane area is  $1.6 \times 10^4$  m<sup>2</sup>.

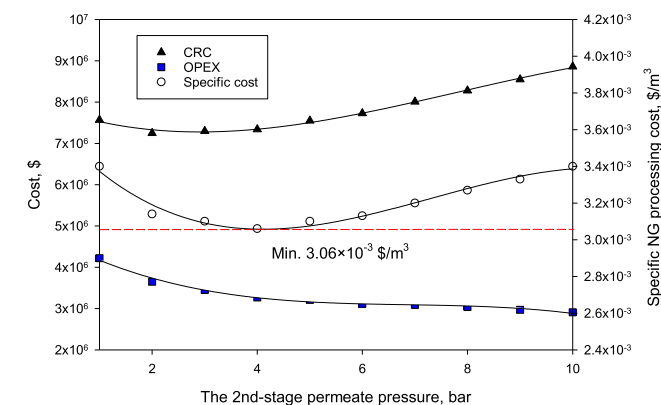


Fig. 4. Dependence of the NG processing cost on the 2nd-stage permeate pressure.

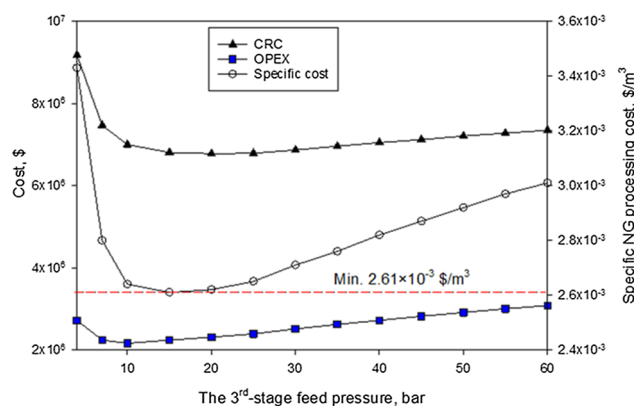


Fig. 5. Dependence of the NG processing cost on the 3rd-stage feed pressure.

increases the specific cost, which is mainly caused by the dramatically increased membrane area due to the decrease of pressure ratio of the 1st-stage carbon membrane unit, and thus a significant increase of CRC. Moreover, it was also found that CRC is dominating the specific cost while OPEX has a relatively less influence, which indicates that reduction of capital cost can significantly bring down the natural gas processing cost. Thus, future research should be focused on the development of advanced membrane materials with higher gas permeance without significant loss of selectivity.

### 3.2. Effect of the 3rd-stage feed pressure

In order to reduce HC loss, the 3rd-stage CO<sub>2</sub> removal unit is applied to recover part of HC, the retentate in the 3rd-stage (R3) is recycled back to the 2nd-stage feed. The stream P2 is re-compressed before sending into the 3rd-stage unit. According to the results in Section 3.1, the optimal permeate pressure in the 2nd-stage is identified to 4 bar, and the scenario B with the variation of the 3rd-stage feed pressure ( $P_3$ ) from 4 to 60 bar was simulated at the constant 3rd-stage permeate pressure ( $p_3 = 1$  bar) and the captured CO<sub>2</sub> recycling ratio of 1. The simulation results are shown in Fig. 5, it can be seen that the specific NG processing cost decreases with the increase of the 3rd-stage feed pressure up to 15 bar, and increases afterwards. It was found that the minimum specific cost is  $2.61 \times 10^{-3}$   $\$/m^3$  at the optimal feed pressure ( $P_{3,opt}$ ) of 15 bar. One should expect that higher feed pressure can significantly reduce the required membrane area with the increased driving force, and thus decreases the membrane unit cost and CRC dramatically. Further increasing feed pressure, the process requires additional compressors and higher power consumption. The reduced membrane area cannot offset the increased cost from compressors, which eventually increases the specific NG processing cost. Moreover, a higher pressure ratio applied to the 3rd-stage membrane unit will lose more work due to the increased entropy production. It is more energy efficient to compress the 3rd-stage retentate (R3) for recycling back to the 2nd-stage unit instead of directly compressing the feed gas (stream F3) to the same pressure, and the optimal 3rd-stage feed pressure of 15 bar is identified.

### 3.3. Effect of the 3rd-stage permeate pressure

Based on the obtained optimal 3rd-stage feed pressure, the scenario C with the variation of the 3rd-stage permeate pressure ( $p_3$ ) from 1 to 6 bar was simulated, and the results are shown in Fig. 6. The lowest specific cost of  $2.55 \times 10^{-3}$   $\$/m^3$  was identified at a permeate pressure ( $p_{3,opt}$ ) of 2 bar. The influence of permeate pressure on specific cost is not significant up to 2.5 bar, but dramatically increases afterwards due to the significant increase of the required membrane area for completing the specific separation requirement (i.e., 95% CO<sub>2</sub>). It should be

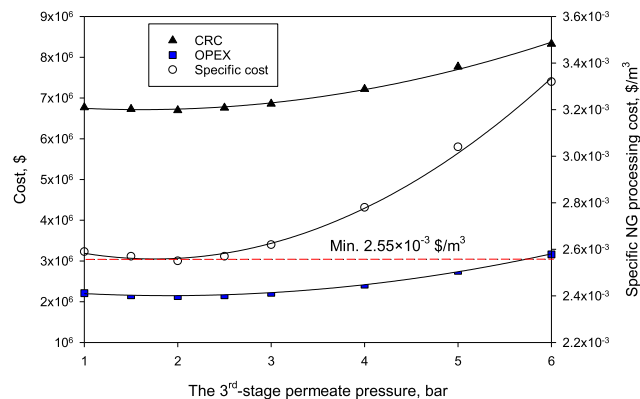


Fig. 6. Dependence of the NG processing cost on the 3rd-stage permeate pressure.

noted that further purification of CO<sub>2</sub> requires a relatively high pressure ratio to maintain a high driving force over the whole membrane area. Pursuing a very low pressure ratio of  $< 6$  (i.e., the 3rd-stage permeate pressure is high than 2.5 bar) will dramatically increase the required membrane area, which leads to the increase of both CRC and specific NG processing cost as shown in Fig. 6. Therefore, identifying the optimal pressure ratio for membrane unit is crucial to bring down NG processing cost.

### 3.4. Effect of recycling ratio

By keeping the optimal operating pressures, the scenario D by varying the CO<sub>2</sub> recycling ratio from 0 to 1 was investigated. It can be found that both specific NG processing cost and HC loss slightly decrease with the increase of the CO<sub>2</sub> recycling ratio as shown in Fig. 7. Without CO<sub>2</sub> recycling (i.e.,  $R = 0$ ), the minimum specific cost is  $2.71 \times 10^{-3}$   $\$/m^3$  purified NG. It should be noted that CO<sub>2</sub> emissions will be higher if more captured CO<sub>2</sub> is used as sweep gas in the 1st-stage dehydration unit, and for EGR is less. Injection of CO<sub>2</sub> into gas wells can enhance natural gas productivity, especially for those gas reservoirs at the late operation stage. Khan et al. [25] investigated the influences of CO<sub>2</sub> injection rate and time on the CO<sub>2</sub>-EGR performance, and the optimal recycling ratio of captured CO<sub>2</sub> should be identified based on the balance of carbon tax and CO<sub>2</sub> breakthrough -this has not been included in the current work. It is worth noting that the HC loss obtained from the simulation is still quite high ( $> 11\%$ ), which is usually not acceptable in most NG plants due to the great greenhouse effect of methane and its economic value. Therefore, reducing HC loss should be further investigated by developing highly selective membranes especially the dehydration unit.

### 3.5. Sensitivity analysis of membrane performance

The major HC loss comes from the dehydration unit based on the process simulations. Thus, development of high-performance dehydration membranes (especially H<sub>2</sub>O/CH<sub>4</sub> selectivity) is crucial to reduce the overall HC loss. Even though membranes with H<sub>2</sub>O/HC selectivity at high pressure operation have not been fully developed, the sensitivity analysis on the influence of dehydration membrane selectivity on HC loss was conducted to document the potential and future research directions on advanced membrane materials development. By keeping constant permeances of the other gas species, various water vapor permeances up to 12X of the referenced Pebax membrane (given in Table 1) were simulated to investigate the influence on HC loss. It should also be noted that the required membrane area is inversely proportional to gas permeance when selectivity is kept constant. The influence of gas permeance on membrane area and cost is quite straight forward, and thus is not discussed here. Process simulations were



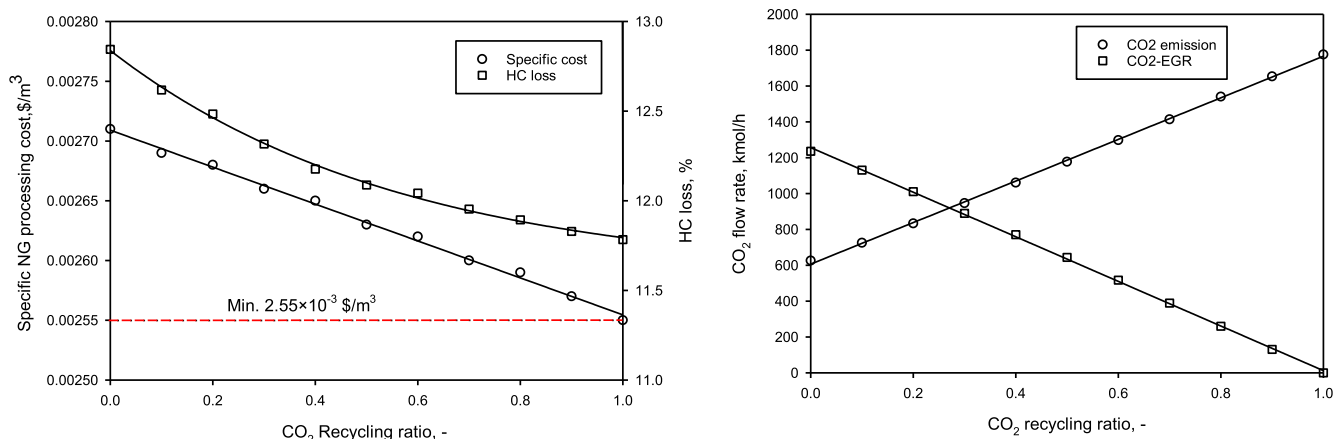


Fig. 7. The influence of CO<sub>2</sub> recycling ratio on NG processing cost and HC loss (Left) and CO<sub>2</sub> emissions (Right).

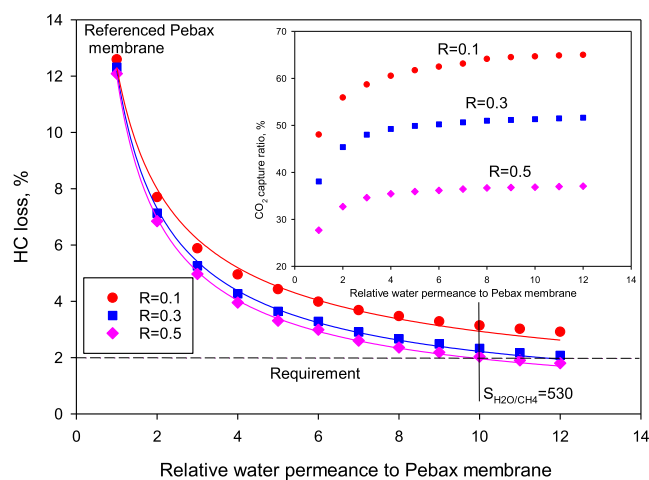


Fig. 8. Dependence of HC loss on the assumed water permeance (2X-12X to the referenced Pebax membrane listed in Table 2) at different CO<sub>2</sub> recycling ratios (0.1–0.5); inset is the dependence of CO<sub>2</sub> capture ratio for EGR on the assumed water permeance at different CO<sub>2</sub> recycling ratio (0.1–0.5).

conducted at the  $p_{2,opt}$  of 4 bar, the  $p_{3,opt}$  of 15 bar, the  $p_{3,opt}$  of 2 bar and the CO<sub>2</sub> recycling ratio of 0.1–0.5, and the results are shown in Fig. 8. It can be found that water permeance of 10X higher compared to the referenced Pebax membrane should be achieved to reach a HC loss of < 2% at a CO<sub>2</sub> recycling ratio of 0.5, which indicated that novel membrane materials should have the H<sub>2</sub>O/CH<sub>4</sub> selectivity of at least 530 at 60 bar. For the low CO<sub>2</sub> recycling ratio of 0.1, achieving the required HC loss of < 2% is extremely challenging. Thus, Future researches on developing highly water-selective membranes at high pressure conditions should be pursued. Moreover, a high CO<sub>2</sub> recycling ratio (e.g., 0.5) is benefitted to reach the requirement of lower HC loss, but the captured CO<sub>2</sub> for EGR is significantly reduced (only 36%), which causes much higher CO<sub>2</sub> emissions compared to the scenario with lower CO<sub>2</sub> recycling ratio (e.g., 0.1). Therefore, the balance of CO<sub>2</sub> emissions and HC loss should be well considered for membrane system design. It is worth noting that carbon membrane performance influences have already been reported in the literature [28], and they concluded that the increase in membrane performance (i.e., CO<sub>2</sub> permeance and CO<sub>2</sub>/CH<sub>4</sub> selectivity) could bring down the specific cost, and high selectivity will have a more significant contribution on reducing energy consumption-this has not been discussed in this work. Moreover, it should also be noted that the material selectivity of a membrane is often collected under ideal conditions. When operated in the field, that process selectivity (separation factor) can suffer even

50% losses due to non-idealities, feed composition, etc.- this should be taken into account for techno-economic feasibility analysis.

### 3.6. Sensitivity analysis of carbon membrane cost

The carbon membrane cost of 50 \$/m<sup>2</sup> might be a bit low due to a more complex preparation process compared to polymeric membranes. Thus, the sensitivity analysis of carbon membrane cost on the specific NG processing cost was also conducted. Since the required carbon membrane area is much larger compared to the dehydration membranes as shown in Table 4, only the carbon membrane cost (30–150 \$/m<sup>2</sup>) was investigated in this work, and the results are shown in Fig. 9. It can be found the specific NG processing cost increases with the increase of carbon membrane material cost. It is worth noting that the optimal permeate pressure for the 2nd-stage unit may change to a lower value when the carbon membrane cost increases, which indicates that the decrease of the operating cost for the re-compression of the permeate gas (feed to the 3rd-stage unit) may not offset the dramatically increased membrane unit cost. Moreover, it was also found the optimal pressures for the 3rd-stage feed and permeate as well as the CO<sub>2</sub> recycling ratio are the same at different carbon membrane costs (see the supporting information). Which is mainly due to the much smaller membrane areas for the dehydration and the 3rd-stage units. One should expect that membrane unit contributes to the major investment cost for membrane technology, and reducing membrane material cost can enhance the competition of membrane technology for gas separation/purification. Therefore, future work on developing high performance membranes at a low cost for high-pressure CO<sub>2</sub>/CH<sub>4</sub>

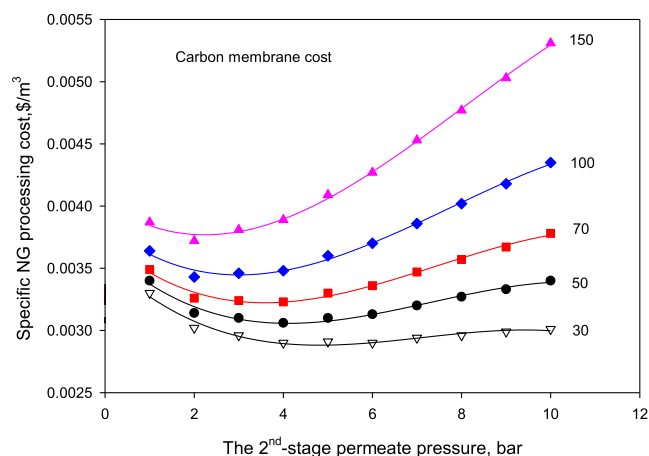


Fig. 9. The influence of carbon membrane cost on the specific NG processing cost at different 2nd-stage permeate pressure.

separation is still essential.

It should be noted that the techno-economic feasibility analysis is much dependent on the cost model employed, and selection of cheaper centrifugal compressors will reduce the specific cost, and the optimal operating condition may be different. Moreover, simulation using process parameter (especially pressure) dependent gas permeances should be conducted in the future work, but more experiment on membranes for natural gas dehydration and sweetening at high pressure condition should be conducted before moving the designed hybrid membrane system to commercial application. Moreover, processing a natural gas with high CO<sub>2</sub> content may require a different process configure (especially CO<sub>2</sub> removal unit as reported by Chu et al. [28]), and the optimal process operation condition discussed above will also be different-this has now been included in the current work.

#### 4. Conclusions

The designed hybrid membrane systems for integrated natural gas dehydration and sweetening using both water-selective and CO<sub>2</sub>-selective membranes are feasible to achieve the separation requirement with HC purity of > 97.5 mol% and dew point of < -40 °C, but HC loss is relatively high. The process operating parameters such as inter-stage feed pressure and permeate pressure of the membrane units were found to have great effects on membrane system performance. The optimal pressure ratio of the 2nd-stage membrane unit is identified as 15, while the 3rd-stage permeate pressure around 2 bar is required to maintain a relatively low natural gas processing cost. Pursuing a very low pressure ratio will increase membrane unit cost dramatically, and thus increase the total capital cost. The lowest specific cost of  $2.55 \times 10^{-3}$  \$/m<sup>3</sup> purified natural gas was theoretically estimated without captured CO<sub>2</sub> for EGR, however, this will influence CO<sub>2</sub> emissions and CO<sub>2</sub>-EGR performance. Moreover, novel dehydration membranes with higher water/HC selectivity at high pressure should be developed in the future work, and advanced membranes with high performance for high pressure CO<sub>2</sub>/CH<sub>4</sub> separation is also required. Nevertheless, process design and optimization should be well considered to achieve a high separation performance for the whole membrane system.

#### CRedit authorship contribution statement

**Xuezhong He:** Conceptualization, Investigation, Writing - original draft, Funding acquisition. **Izumi Kumakiri:** Methodology, Writing - review & editing, Funding acquisition. **Magne Hillestad:** Conceptualization, Writing - review & editing, Supervision.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.seppur.2020.116993>.

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