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

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\begin{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\textsubscript{2}. 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\textsubscript{2}, 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 < 2.71 × 10\textsuperscript{−3} $/m\textsuperscript{3} sweet natural gas was estimated to achieve the separation requirement of < 2.5 mol\% CO\textsubscript{2} in purified NG together with captured high purity CO\textsubscript{2} (> 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\textsubscript{2}O/CH\textsubscript{4} selectivity at high pressure should be pursued to promote the application of the designed membrane system for subsea natural gas dehydration and sweetening.
\end{abstract}

1. Introduction

Natural gas (NG) is considered as one of the less carbon-intensive energies, and the demand 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\textsubscript{2}O/CH\textsubscript{4} separation in natural gas dehydration were reviewed in the literature [2,6], and very high H\textsubscript{2}O/CH\textsubscript{4} selectivity (several thousands) for some membranes such as polyimide (Kapton) cellulose acetate, and Nafion\textsuperscript{®} 117 were reported. It should be noted that the reported performances (both H\textsubscript{2}O permeability

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and \( \text{H}_2\text{O}/\text{CH}_4 \) 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 \( \text{H}_2\text{O}/\text{CH}_4 \) separation at a low feed pressure of 2 bar [6], but a reasonable water permeance of 0.055 \( \text{m}^3 \) (STP)/\( \text{m}^2\cdot\text{h}\cdot\text{bar} \) and a low \( \text{H}_2\text{O}/\text{CH}_4 \) selectivity of 47 at 30–60 bar were reported from their field testing [7]. Thus, membrane performance (especially \( \text{H}_2\text{O}/\text{CH}_4 \) 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 \( \text{H}_2\text{O}/\text{CH}_4 \) 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 \(-40 ^\circ \text{C}\) in natural gas. Moreover, natural gas sweetening is also required to remove \( \text{CO}_2 \) and reach the content of \(< 2.5 \text{ mol}\% \) in the product gas [8]. Although chemical (amine-based) absorption is the commercial technology for \( \text{CO}_2 \) removal in 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 separation membrane is considered as a promising technology for this environment with a second pollution for amine-based absorption. 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 \( \text{CO}_2/\text{CH}_4 \) selectivity over 34 at 75 °C [10]. Recently, the fixed-site-carrier (FSC) membranes have been reported for \( \text{CO}_2 \) removal from natural gas [9,11]. This type membrane showed a good \( \text{CO}_2/\text{CH}_4 \) separation performance at moderate pressures [9,11]. However, the challenge on getting good performance at higher pressure > 40 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 cellulose-derived hollow fiber carbon membranes presented a good selectivity of \( \text{CO}_2/\text{CH}_4 \) with a lower \( \text{CO}_2 \) permeance (usually < 30 GPU) due to a symmetric structure with a thickness of ca. 30 \( \mu \text{m} \) [16–24]. Recently, carbon membranes made from polyimide precursors were tested at high pressure of up to 120 bar for \( \text{CO}_2/\text{CH}_4 \) separation [15]. They reported a \( \text{CO}_2 \) permeance of 100 GPU (i.e., 0.28 \( \text{m}^3 \) (STP)/(\( \text{m}^2\cdot\text{h}\cdot\text{bar} \))) and a \( \text{CO}_2/\text{CH}_4 \) separation factor of 60 based on a 50% \( \text{CO}_2/\text{50% CH}_4 \) 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, \( \text{H}_2\text{S}, \text{CO}_2 \) and helium. Among them, water and acid gases need to be removed to meet gas grid specifications. A hybrid membrane system for natural gas dehydration and sweetening, illustrated in Fig. 1, was designed to reach the low impurity requirements of water (dew point < \(-40 ^\circ \text{C}\)) and \( \text{CO}_2 \) (< 2.5 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 \( \text{CO}_2 \). 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 \( \text{H}_2\text{O} \) and part of \( \text{CO}_2 \) and \( \text{HCs} \) permeate through the membranes. The dried gases containing less permeable components (mainly \( \text{HCs} \) and \( \text{CO}_2 \)) 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 \( \text{CO}_2 \), and produce sweet natural gas (i.e., HC content of > 97.5 mol%) in the retentate as the product gas. Part of the captured \( \text{CO}_2 \) from the \( \text{CO}_2 \) 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 \( \text{NG} \) in gas phase will be burned out before venting (this is the major HC loss). Moreover, the rest of the captured \( \text{CO}_2 \) will be further compressed and injected back to gas reservoirs for enhanced gas recovery (EGR)-this is to displace natural gas by injecting \( \text{CO}_2 \) in the supercritical phase. Implementation of \( \text{CO}_2 \)-EGR can potentially enhance the recovery of natural gas and store \( \text{CO}_2 \) underground to reduce \( \text{CO}_2 \) emissions [25,26]. It should be noted that the influence of \( \text{CO}_2 \) for EGR is not clarified [27], but a high purity of \( \text{CO}_2 > 95\% \) 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 × 10^5 \( \text{m}^3 \) (STP)/h natural gas plant (10 mol% \( \text{CO}_2 \) in water-saturated natural gas, see the detailed gas composition in Table 1) at 80 °C. The \( \text{CH}_4 \) purity of 97.5 mol%, the \( \text{CO}_2 \) purity of 95 mol% and the gas dew point of < \(-40 ^\circ \text{C}\) 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 \( \text{C}_2-\text{C}_5+ \) permeances of carbon membranes were estimated to 0.001 mol(STP)/(\( \text{m}^2\cdot\text{h}\cdot\text{kPa} \)) 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
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\(_2\) recycling ratio, etc.) were adjusted to investigate their influences on natural gas processing cost. The membrane areas in the dehydration and CO\(_2\) 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\(^2\) 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\(^S\), $/m\(^3\) sweet NG) [28] is estimated by:

\[
C^S = \frac{\text{CRC} + \text{OPEX}}{\text{Annual Total sweet NG production}}
\]  

The C\(^S\) 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 1-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\(_2\) 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\(_2\)). The stream P\(_2\) is then compressed and fed into the 3rd-stage (carbon) membrane unit to produce high purity CO\(_2\) in the permeate side (> 95 mol\%) where feed pressure (P\(_3\)) and permeate pressure (p\(_3\)) were varied. Part of the captured CO\(_2\) (CO\(_2\)-Sweep) is recycled back to the sweep side of the dehydration unit, and the rest CO\(_2\) (Stream CO\(_2\)-EGR) is re-compressed for enhanced gas recovery. The process operating parameters such as the permeate pressures of the streams P\(_2\) and P\(_3\), the feed pressure of the stream F\(_3\) as well as the recycling ratio of capture CO\(_2\) 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.
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 CO₂ 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 × 10⁻³ $/m³ at the optimal permeate pressure (p², opt) of 4 bar, corresponding to a pressure ratio of 15. One should expect that a lower permeate pressure (p₂) 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

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

Table 4

| 2nd-stage permeate pressure, bar | Total compressor power demand, kW | 2nd-stage membrane area, m² | 3rd-stage membrane area, m² |
|---------------------------------|----------------------------------|-----------------------------|----------------------------|
| 1                               | 1.1 × 10³                        | 5.2 × 10⁴                   | 7.1 × 10⁴                   |
| 2                               | 9.1 × 10³                        | 6.5 × 10⁴                   | 6.9 × 10⁴                   |
| 3                               | 8.6 × 10³                        | 8.1 × 10⁴                   | 6.9 × 10⁴                   |
| 4                               | 8.2 × 10³                        | 9.6 × 10⁴                   | 6.6 × 10⁴                   |
| 5                               | 8.0 × 10³                        | 1.2 × 10⁵                   | 6.2 × 10⁵                   |
| 6                               | 7.8 × 10³                        | 1.4 × 10⁵                   | 5.7 × 10⁵                   |
| 7                               | 7.7 × 10³                        | 1.6 × 10⁵                   | 5.2 × 10⁵                   |
| 8                               | 7.6 × 10³                        | 1.8 × 10⁵                   | 4.8 × 10⁵                   |
| 9                               | 7.4 × 10³                        | 2.1 × 10⁵                   | 4.0 × 10⁵                   |
| 10                              | 7.3 × 10³                        | 2.3 × 10⁵                   | 3.2 × 10⁵                   |

The dehydration membrane area is 1.6 × 10⁴ m².

Table 3

| Scenario | The 2nd-stage permeate pressure, p₂ (bar) | The 3rd-stage feed pressure, p₃ (bar) | The 3rd-stage permeate pressure, p₃ (bar) | The recycling ratio of captured CO₂, R (-) |
|----------|------------------------------------------|--------------------------------------|----------------------------------------|------------------------------------------|
| A        | 1–10 bar                                 |                                      |                                        |                                          |
| B        | p₂,opt (Optimized from the scenario A)   | p₃ (60)                              |                                        |                                          |
| C        | p₂,opt                                   | p₃,opt (Optimized from the scenario B) |                                        |                                          |
| D        | p₂,opt                                   | p₃,opt                               |                                        |                                          |

Fig. 4. Dependence of the NG processing cost on the 2nd-stage permeate 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 CO2 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 (p3) from 4 to 60 bar was simulated at the constant 3rd-stage permeate pressure (p2 = 1 bar) and the captured CO2 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 ($p_{3,\text{opt}}$) of 15 bar. One should expect that higher feed pressure can significantly reduce the required membrane area with the increased driving force, and thus deceases 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 (p3) 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 ($p_{3,\text{opt}}$) 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% CO2). It should be noted that further purification of CO2 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 CO2 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 CO2 recycling ratio as shown in Fig. 7. Without CO2 recycling (i.e., R = 0), the minimum specific cost is $2.71 \times 10^{-3}$ $/m^3$ purified NG. It should be noted that CO2 emissions will be higher if more captured CO2 is used as sweep gas in the 1st-stage dehydration unit, and for EGR is less. Injection of CO2 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 CO2 injection rate and time on the CO2-EGR performance, and the optimal recycling ratio of captured CO2 should be identified based on the balance of carbon tax and CO2 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 H2O/CH4 selectivity) is crucial to reduce the overall HC loss. Even though membranes with H2O/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 simulation 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...
conducted at the $p_{2, \text{opt}}$ of 4 bar, the $p_{3, \text{opt}}$ of 15 bar, the $p_{3, \text{opt}}$ of 2 bar and the CO$_2$ 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$_2$ recycling ratio of 0.5, which indicated that novel membrane materials should have the H$_2$O/CH$_4$ selectivity of at least 530 at 60 bar. For the low CO$_2$ 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$_2$ recycling ratio (e.g., 0.5) is benefitted to reach the requirement of lower HC loss, but the captured CO$_2$ for EGR is significantly reduced (only 36%), which causes much higher CO$_2$ emissions compared to the scenario with lower CO$_2$ recycling ratio (e.g., 0.1). Therefore, the balance of CO$_2$ emissions and HC loss should be well considered for membrane system design. 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$_2$ 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$_2$/CH$_4$...
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 CO2 content may require a different process configure (especially CO2 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 CO2-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 interstage 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 × 10⁻³ $/m² purified natural gas was theoretically estimated without captured CO2 for EGR, however, this will influence CO2 emissions and CO2-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 CO2/CH4 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

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