

Design and Optimization of Membrane System for Gas Separation

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Abstract

Membrane separation may substitute conventional energy intensive technologies, and it could have cost benefits and lower environmental footprints. Several membrane processes have been developed to achieve higher purity and recovery of products. This study uses a generic membrane superstructure (or system) that facilitates all possible inter- and intra-connections among different membrane stages (or units) which are arranged in series-parallel configurations. The developed mathematical model for the membrane system is a mixed integer non-linear programming (MINLP) problem. The mathematical model is implemented in AMPL, and BARON solver is used to solve it. The MINLP model of the membrane system can choose membrane from a Membrane Database, which has a number of polymeric and inorganic (graphene, carbon molecular sieve, zeolite and metal-organic frameworks) membranes. In this work, two industrial case studies of gas separation are considered: post-combustion CO₂ capture and biogas upgradation by CO₂ removal. The selection of CO₂ removal technology depends on plant location, production capacity and product quality specifications. The chosen applications have challenges in terms of energy consumptions, economics and environmental burden. The separation performance of the membrane system is evaluated and compared for same membrane in all membrane stages. Two optimization problems were solved for each membrane: minimization of total area of membranes and minimization of total mechanical power. For both applications, best performing membranes were identified to target the minimum separation cost.

Keywords: Membrane System, Post-combustion CO₂ Capture, Biogas Upgradation.

1. Introduction

Membrane separation is one of the emerging technologies that has the potential to replace traditional energy intensive separation technologies. In process industry, solvent absorption (amine absorption), solid adsorption (pressure swing adsorption) and cryogenic distillations are used to separate gas mixtures (Tock, 2013; Leung et al., 2014). Recently, gas separation using membranes has received considerable attention for industrial applications, namely air separation, syngas ratio adjustment, hydrogen recovery in refinery, post-combustion CO₂ capture and biogas upgradation by CO₂ removal (Ismail, 2015). Membrane separation has several advantages over conventional gas separation technologies, e.g., no use of chemicals, mild operating conditions, simple installation and easier operation, and flexibility to integrate with other separation technologies.

Several studies have explored post-combustion CO₂ capture from coal and natural gas power plants, using membranes (Kárászov et al., 2020). Zhang et al. (2014) studied post combustion carbon capture, and amine-based capture system had higher energy

consumptions and environmental impact compared to the membrane process. Arias et al. (2016) optimized performance of multi-stage membrane superstructure for capturing CO₂ from flue gases. Lee et al. (2018) optimized membrane superstructure for CO₂ capture from coal power plants, and showed the benefits of using different membranes in different membrane stages. Scholz et al. (2013) performed detailed analysis of biogas upgradation into biomethane using several types of membranes. Finally, Sun et al. (2015) reviewed several biogas upgrading technologies, including cryogenic separation, physical and chemical absorptions, pressure swing adsorption, membrane separation, hydrate formation and biological methods.

Post-combustion CO₂ capture and biogas upgradation have several challenges such as energy consumptions, capital and operating costs. The selection of CO₂ removal technologies depends on plant location, production capacity, product quality specifications, availability of financial resources, environmental regulations and energy integration with CO₂ emitting plant or industrial site. In order to achieve the required purity and capture rate, membranes are arranged in complex series-parallel configurations. This arrangement gives numerous degrees of freedom for membrane system design. In this study, a generic superstructure of membrane modules/units, with all possible inter- and intra-connections, is used (see Figure 1). A mixed integer non-linear programming problem of membrane superstructure has been developed in AMPL (A Mathematical Programming Language). A database of several membranes has been used, and optimization method can choose any membrane from the database.

Two important industrial case studies of gas separation are considered: post-combustion CO₂ capture and biogas upgradation by removing CO₂. There are eight (M1-M8) and seven (m1-m7) membranes respectively in the membrane databases for post-combustion CO₂ capture and biogas upgradation. The separation performance of the membrane system was evaluated and compared for all membranes. For each membrane, two optimization problems were solved: minimization of total area of membrane (TAM), and minimization of mechanical power (TP). The optimization results allow to identify best performing membranes for both applications. In the mathematical model of the membrane system, different membranes can also be used in different membrane stages to improve the separation performance. For both applications, the final solution always contains same membranes in both membrane stages, as Membrane Database has limited number of membranes. In case of large number of membranes in the Membrane Database, the proposed approach can identify best performing membrane clusters, based on the membrane permeability and selectivity. These findings could be useful to the membrane researchers for further improving the performances of their membranes.

2. Membrane Superstructure Model

Figure 1 presents generic membrane superstructure or system. Fresh feed is compressed [$C(F)$] and cooled-down [$HE(F)$] before it enters the membrane system. The fresh feed can go to any membrane stage or unit in the membrane system. Figure 1 shows i^{th} membrane stage of the membrane system, inside the dotted line. Each membrane stage has a membrane module [$MEM(i)$], a mixer [$M(f,i)$], two splitters [$S(r,i)$, $S(p,i)$] on retentate and permeate sides, a compressor [$C(i)$] and cooler [$HE(i)$] for permeate stream. The membrane stage mixer [$M(f,i)$] is used to mix the fresh feed and retentate and/or permeate recycled from the same or different membrane stages. Finally, there are two mixers [$M(r)$, $M(p)$] for both product streams. A turbine [$T(P)$]

and a heater $[HE(P)]$ are used to recover the mechanical power, from the retentate side product.

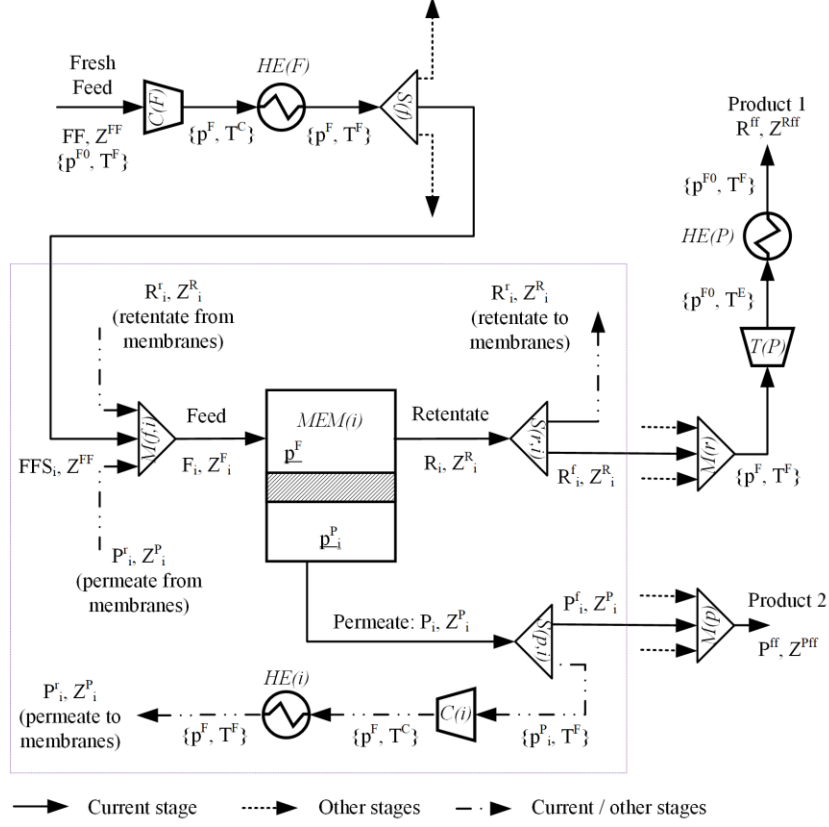


Figure 1: Membrane superstructure for gas separation

The mathematical model for the membrane superstructure is a mixed integer non-linear programming (MINLP) problem. Table 1 summarizes balances and equations for different units in membrane system.

Table 1: Summary of MINLP model for membrane system

| Units | Balances or Equations |
|--|---|
| Membrane system | Mass and component balances |
| Splitters $[S(F), S(r,i), S(p,i)]$ | Mass balance |
| Mixers $[M(f,i), M(p), M(r)]$ | Mass and component balances |
| Membranes $[MEM(i)]$ | Mass and component balances, membrane transport |
| Compressors, turbines $[C(F), C(i), T(P)]$ | Equations for calculating outlet temperatures and powers ($\eta_C = 0.8$, $\eta_T = 0.85$) |
| Heaters, coolers $[HE(F), HE(i), HE(P)]$ | Heat balance equation for calculating heat duties |
| Constraints | Limits on the product purities |

3. Post-combustion CO₂ Capture

The flue gases contain mainly N₂, O₂, CO₂ and H₂O. This work considers separation of water prior to the use of membrane separation for CO₂ capture. It is assumed that the feed contains 14% CO₂ and remaining 86% N₂. The CO₂ and N₂ mixture have a flow rate of 10 mol/s at 1 bar pressure and 40 °C temperature. The membrane database has eight membranes (M1-M8), and Figure 2(c) shows CO₂ permeance and CO₂/N₂ selectivity for different membranes. M1-M3 are polymeric membranes whereas M4-M8 are inorganic (graphene, carbon molecular sieve, zeolite, metal-organic frameworks) membranes. The feed side pressure for membrane units can vary between 5 and 13 bar (Minh et al., 2008). The CO₂ and N₂ streams from the membrane system have 95% purities.

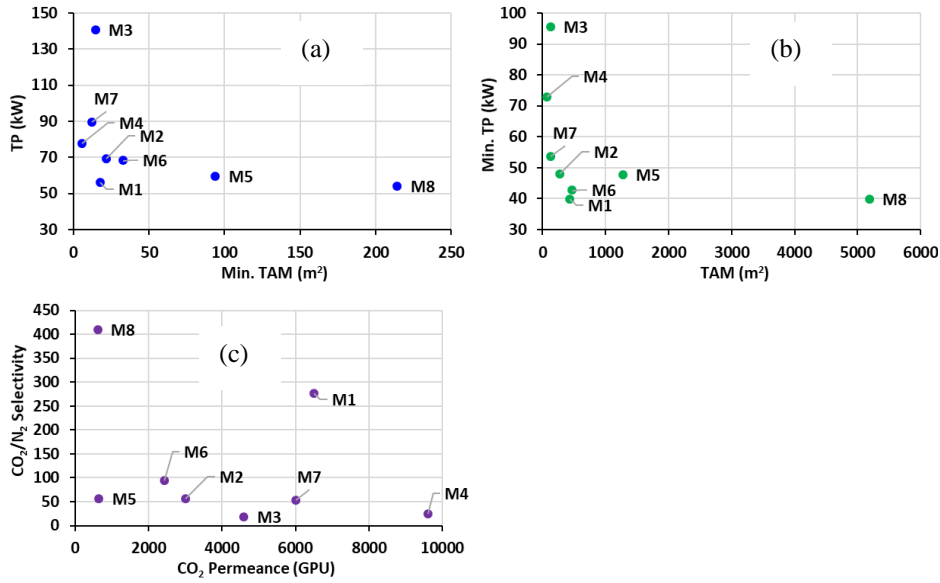


Figure 2: Post-combustion CO₂ capture using different membranes: (a) minimization of total area of membranes, (b) minimization of total mechanical power, (c) CO₂ permeance versus CO₂/N₂ selectivity for all membranes.

For this study, the membrane system has two membrane stages. For each membrane in the membrane database, two optimization problems were solved: minimization of total area of membranes, and minimization of total mechanical power (feed compression + stage compression – product expansion). The optimization problems have 252 variables and 266 constraints, and they were solved using BARON (v21.1.13) solver in AMPL (v20210220), with a maximum solution time of 30 minutes. Figure 2(a) presents optimization results for minimum total area of membranes. This figure also presents related values of total mechanical power obtained for different solutions. Similarly, Figure 2(b) presents results for minimum total mechanical power (2nd optimization problem) for all membranes. To minimize the total cost of separation, both total area of membranes and total mechanical power are equally important. It can be seen from Figures 2(a) and 2(b) that membrane M1 has the best performance for separating a mixture of CO₂ and N₂. Further, membrane M1 has best compromise between permeance and selectivity. Membrane M4 has very high permeance but low selectivity, whereas membrane M8 has very high selectivity but low permeance.

4. Biogas Upgradation by CO₂ Removal

The biogas contains 38% CO₂ and remaining 62% CH₄. The biogas has a flow rate of 10 mol/s at 1 bar pressure and 40 °C temperature. The membrane database for biogas upgradation has seven membranes (m1-m7), and Figure 3(c) shows CO₂ permeance and CO₂/CH₄ selectivity for different membranes. m1-m3 are polymeric membranes whereas m4-m7 are inorganic (graphene, carbon molecular sieve, zeolite, metal-organic frameworks) membranes. Two membrane stages were considered for separating CO₂ and CH₄ mixture. The feed side pressure for membrane units can vary between 5 and 13 bar. The CO₂ and CH₄ streams from the membrane system have 95% purities.

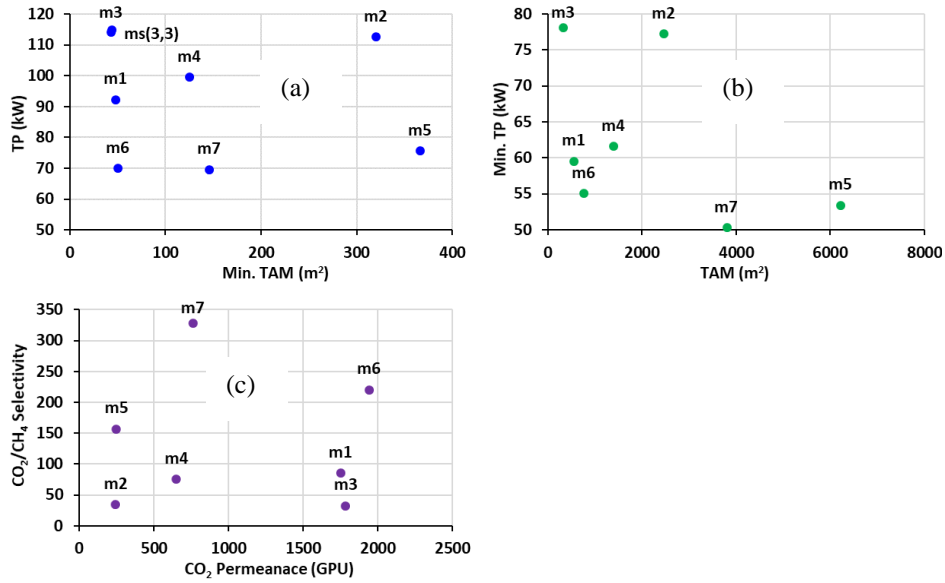


Figure 3: Biogas upgradation by CO₂ removal using different membranes: (a) minimization of total area of membranes, (b) minimization of total mechanical power, (c) CO₂ permeance versus CO₂/CH₄ selectivity for all membranes.

Similar to previous case study, two optimization problems were solved for all the membranes: minimization of total area of membranes, and minimization of total mechanical power. Both optimization problems have 252 variables and 266 constraints, and they were solved using BARON solver in AMPL, with a maximum solution time of 30 minutes. Figure 3(a) presents optimization results for minimum total area of membranes. This figure also present values of total mechanical power obtained for different solutions. Figure 3(b) presents results for minimum total mechanical power for all membranes, along with related total area of membranes. For minimum total cost of separation, solutions obtained for membrane m6 are better than other membranes, as these solutions are nearer to the corner. Membrane m6 has best compromise between permeance and selectivity, as shown in Figure 3(c).

The mathematical model of membrane system can use different membranes in different stages to improve the separation performance. For biogas upgradation, we minimized total area of membranes, and this optimization problem has 250 variables and 268 constraints. The optimal solution uses membrane 3 in both stages (see solution ms(3,3) in Figure 3(a)). Figure 4 provides detail of solution ms(3,3). If we have many membranes

in the Membrane Database, it is possible to identify best performing membrane clusters, for separating a gas mixture.

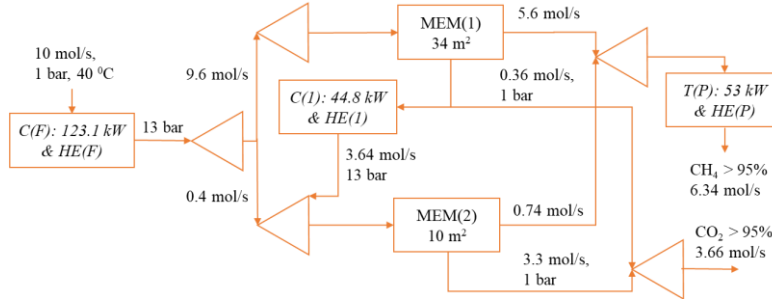


Figure 4: Details of solution ms(3,3); membrane 3 was used in both membrane stages

5. Conclusions

This study develops a mathematical model for multi-stage membrane superstructure. The model is a mixed integer non-linear programming problem that has been implemented in AMPL, and solved using BARON solver. Two case studies, namely post-combustion carbon capture and biogas upgradation were solved using the developed mathematical optimization problem. The optimization results present optimal flows and pressure levels inside the membrane system, and also allow to identify best performing membranes. The proposed approach can identify best performing membrane clusters, based on the membrane permeability and selectivity. This knowledge could be useful to the membrane developers for further improving the performances (selectivity versus permeance) of membranes. The future studies will focus on separating a gas mixture with three components, using different number of membrane stages.

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