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Hybrid Membrane-Distillation Separation Processes

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A novel synthesis approach is developed to evaluate and optimise various hybrid membrane-distillation schemes. The problem is formulated as an optimisation problem, where the objective is to minimise the total operating cost of the separation scheme. Interactions between the separation and refrigeration systems are considered and opportunities for heat integration are exploited. A case study, ethylene/ethane separation, is presented to demonstrate the synthesis approach. Schemes that apply a membrane in parallel to and in series with a distillation column are explored.
Shortcut models that account for multiple feeds and products are used to represent the distillation column. The distillation model predictions are shown to be in good agreement with results of more rigorous simulations carried out using HYSYS. Established membrane models (Shindo et al., 1985) are applied and shown to be valid for the system of interest. A systematic approach is developed to account for heat recovery between: i) column feeds and products; ii) the membrane feed and products and iii) the associated refrigeration system.
The optimisation results reveal that a facilitated transport membrane reported in the literature (Pinnau and Toy, 2001) used in parallel with a distillation column can reduce the condenser duty by about 33 %, compared to a conventional distillation column operating at the same pressure (20 bar). However, recompression and sub-ambient cooling are required for the permeate stream, incurring operating costs. The total operating cost of the heat-integrated parallel hybrid scheme may be reduced by 11 %, compared to a conventional distillation column.

1. Introduction

Separation processes represent 40 to 70 % of both capital and operating costs in industry. They also account for approximately 60 % of all the process energy used by the chemical and petroleum refining industries every year in the US (Eldridge et al., 2005). With industry's need to reduce operating costs and minimise environmental impact, extensive research has been performed to study new technologies which offer viable alternatives to traditional energy-intensive technologies.
Process integration and complex column configurations have proven to be very successful in reducing the energy costs for conventional distillation arrangements (Wang and Smith, 2005). Heat pumps systems can offer total annual costs savings of 37-39 % for olefin-paraffin separations (Kiss et al., 2012). Extractive distillation, molecular sieve adsorption and reactive absorption, and the combination of membrane and distillation technologies to form a hybrid system are also possible alternatives to energy-intensive conventional distillation (Motelica et al., 2012). Recently, systematic frameworks for process design and optimisation have been introduced. Marquardt et al. (2008) present a framework for the design of separation flowsheets which include hybrid membrane-distillation separations. In the proposed framework, different flowsheets are generated and then evaluated with shortcut methods. Finally the most energy-efficient alternatives are rigorously optimized using MINLP to calculate detailed information and obtain the most cost-effective design. Similar approaches to that of Marquardt et al. (2008) have been employed by Caballero et al. (2009) and Skiborowski et al. (2013) for the design of hybrid membrane-distillation systems. The important features of the approaches discussed above are that the advantages of shortcut methods and of rigorous simulation are exploited. The key shortcoming is the requirement to pre-specify the number of stages for the distillation column. Also, in the aforementioned frameworks, heat integration

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within the separation system and between the separation process and refrigeration system is not considered. There might be a greater opportunity to reduce the energy consumption of the system by introducing process-to-process heat recovery, and by recovering heat between the refrigeration system and the separation process (i.e. heat can be rejected to a heat sink within the process rather than to an external cooling utility). Therefore, a novel synthesis and design framework to screen, examine and optimise heat-integrated hybrid membrane-distillation separation process is developed, and presented in this work.

2. Design and optimisation methodology

Figures 1 and 2 present the approach used for the design and optimisation of heat-integrated hybrid membrane-distillation systems. The process simulation, which is the main component of the optimisation framework, applies established models for all the separation units, for the refrigeration system and for heat recovery.

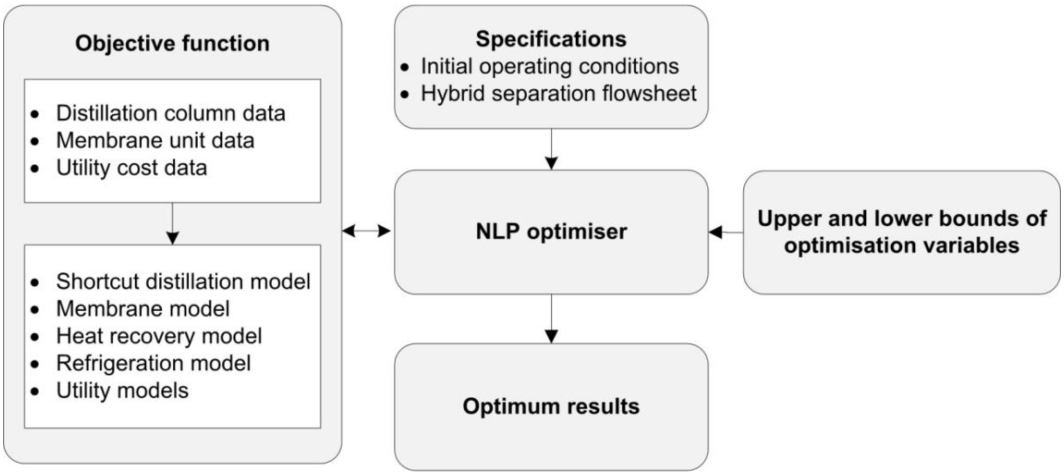


Figure 1: Overview of optimisation approach

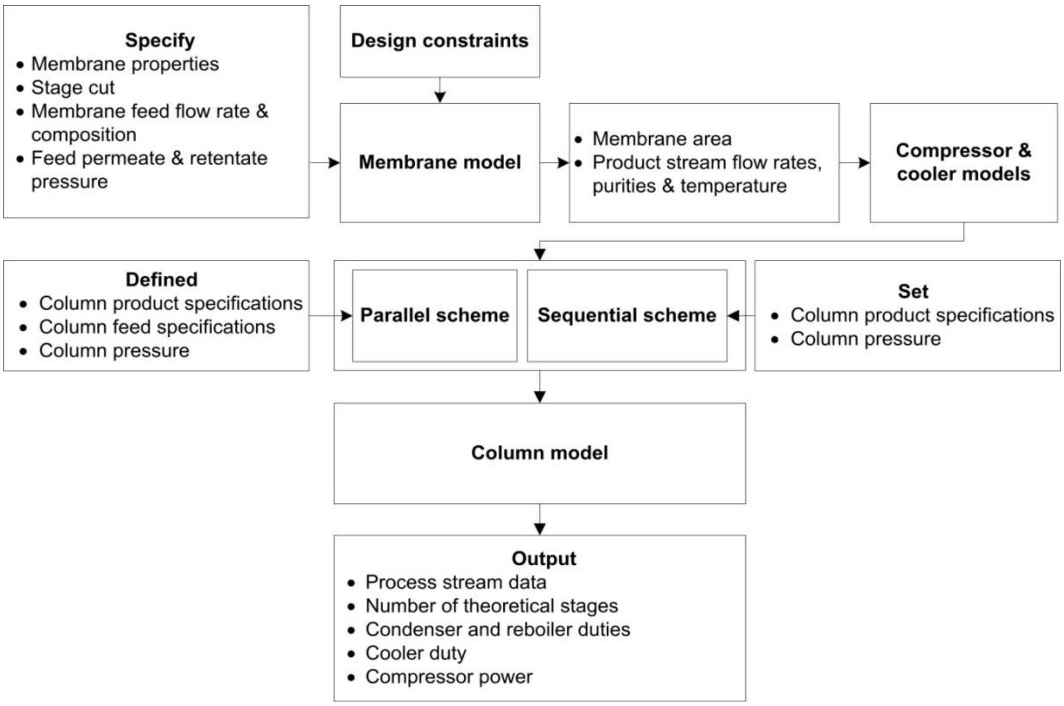


Figure 2: Simulation and design flowchart for the parallel and sequential hybrid system

Figure 2 shows the simulation and design flowchart for the parallel hybrid system and the sequential hybrid system. In the sequential hybrid configuration, a membrane module is placed in the process feed stream, and both the permeate and retentate feed the distillation column. In the parallel hybrid configuration, the membrane feed stream is withdrawn from an intermediate stage of the distillation column. The permeate stream leaving the membrane unit is recompressed and cooled. The retentate stream and recompressed permeate stream both re-enter the distillation column (at appropriate stages, where the stream composition matches the tray composition).

All the process simulation and optimisation is embedded in MATLAB. In this work, MATLAB is connected with HYSYS through an active client-server application for the calculation of thermodynamic and physical properties. To design and simulate the membrane, the modelling approach presented by Shindo et al. (1985) is employed. To design the distillation column, the Fenske-Underwood-Gilliland (FUG) shortcut method is modified to improve its performance particularly that of the Underwood method, which generally underestimates the minimum vapour flow. The vapour flow is only assumed constant in the pinch zone, and the minimum vapour flow in the top section of the column is calculated by performing an enthalpy balance.

The separation models are used to simulate and design the separation units, and to calculate the heating and cooling demands of the separation processes. The temperature and heat loads of the process streams requiring heating and cooling are inputs to the heat recovery model. In this work, the heat recovery model proposed by Farrokhpanah (2009) is modified and used. Farrokhpanah (2009) applied a matrix-based approach for assessing opportunities for heat integration. Synthesis and optimisation of refrigeration systems was the focus in the work of Farrokhpanah (2009); in contrast, in this work, a simple model for the refrigeration system is used to predict the coefficient of performance of the refrigeration system and the cost of indirect heat exchange (i.e. heat exchange between the separation process and the refrigeration system). The heat recovery model identifies opportunities to reuse heat rejected by hot streams to heat cold streams, including using heat rejected from a refrigeration cycle for process heating, in turn reducing the compression power demand of the refrigeration system.

The energy consumption of the overall system can be determined. In this work, the optimisation aims to minimise the total operating cost (TOC) of the separation system. The optimisation problem is formulated as a NLP and solved by the 'pattern search' method (Math Works Global Optimization Toolbox User's Guide, 2013).

The process models allow the performance (in terms of TOC) of the process to be determined; this value is input to the optimisation algorithm, which selects a new set of inputs to the process models. Iteration continues until a termination criterion is met; the optimum operating conditions and the optimal configuration are thus determined.

The total operating cost (the objective function) depends on the problem boundaries. In particular, the operating cost depends on whether heat recovery is taken into account. Eq(1) presents the case in which heat integration is not taken into account, where the compression power demand is given by Eq(2), and Eq(3) addresses the case in which heat recovery is considered:

$$TOC = WC^{elec} + \sum_{i=1}^{nsource-m} \frac{Q_{evapi}}{COP_i}\left(C^{elec} + C^{cw}\right) + \sum_{j=1}^{nsink} Q_{Hj} C_k^{steam} + C^{cw}\left(\sum_{i=1}^{nsource-m} Q_{evapi} + \sum_{i=1}^{m} Q_{Ci}\right) \quad (1)$$

$$W = W_P + W_R + W_F \quad (2)$$

Where W is the compressor shaft power (P: permeate; R: retentate; F: feed); Q is cooling or heating duty (evap: evaporator; H: heater; C: cooler); *COP* is the coefficient of performance of the refrigeration system; C represents cost values (elec: electricity; cw: cooling water; C_k^{steam} is the steam cost in the range of temperature k); m is the number of cold streams that use cooling water as the cooling utility.

$$TOC = WC^{elec} + \sum_{i=1}^{nsource} \sum_{j=1}^{nsink} Q_{ij} C_{ij} \quad (3)$$

Where W is the compressor shaft power; C^{elec} is the cost of electricity; C_{ij} is the utility cost of the heat recovery 'match' between source i and sink j per unit of heat load; Q_{ij} is the load on the match between source i and sink j; n_{source} is the number of source streams; n_{sink} is the number of sink streams.

The optimisation is subject to:

$$P_R = P_{MF} \tag{4}$$

$$P_{MF} = P_{COL} \tag{5}$$

$$P_P < P_{MF} \tag{6}$$

$$P_P^L \le P_P \le P_P^U \tag{7}$$

$$\theta^L \le \theta \le \theta^U \tag{8}$$

$$F_{MF}^L \le F_{MF} \le F_{MF}^U \tag{9}$$

$$y_{MF,i}^L \le y_{MF,i} \le y_{MF,i}^U \tag{10}$$

Where P is the pressure (R: retentate; P: permeate; MF: membrane feed; COL: column), Θ is the stage cut (L: lower bound; U: upper bound), F_{MF}: membrane feed molar flow rate, $y_{FM,i}$: mole fraction of component i in the membrane feed stream.

3. Case study

Option 1: Parallel hybrid scheme

To illustrate the implementation of the proposed approach for simultaneous structural and operational optimisation to separate a close-boiling mixture, the case study considers a mixture with feed composition and column specifications shown in Table 1. Cross-flow is assumed for the membrane. Table 2 lists the facilitated transport membrane properties of the solid polymer electrolyte composite membrane introduced by Pinnau and Toy (2001) for the separation of ethylene and ethane.

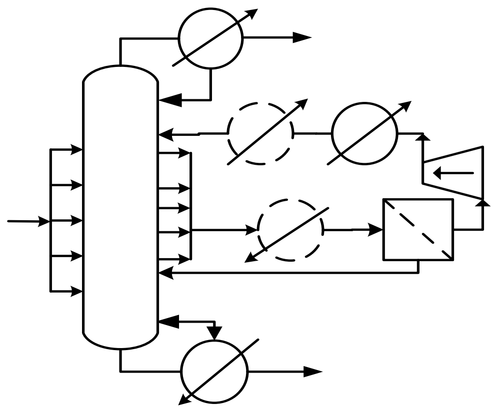
A superstructure representation for the 'parallel-hybrid flowsheet' optimisation is shown in Figure 3. The optimisation determines the optimal operating conditions and the optimal locations for the feed and side draw streams. A generalized pattern search method is used to solve the optimisation problem with the upper and lower bound values given in Table 3. The optimisation variables and their bounds are selected based on a preliminary sensitivity analysis (for which results are not presented).

Table 1: Column data

Stream	Feed	Distillate	Bottom
Temperature (°C)	-19.06	-29.21	-7.89
Pressure (bar)	20	20	20
Flowrate (kmol h^{-1})	100	53.82	46.18
C2H4 (mole fraction)	0.54	0.999	0.005

Table 2: Membrane properties

Membrane feed pressure (bar)	20
Membrane retentate pressure (bar)	20
Membrane feed temperature (°C)	23
Permeability of C_2H_4 (mol m s^{-1} m^{-2} Pa^{-1})	0.3×10^{-14}
Membrane thickness (m)	5×10^{-6}
Membrane selectivity	54



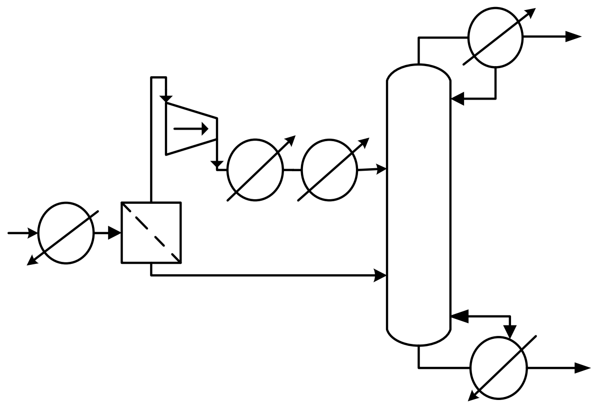


Figure 3: Superstructure representation for parallel hybrid schemes

Figure 4: Fixed sequential hybrid scheme

The objective is to minimise TOC of the separation flowsheet. The optimisation is carried out for two scenarios: (1) disallowing heat recovery; (2) with heat integration between the separation system and the associated refrigeration system. Utility costs are those presented by Farrokhpanah (2009). When designing the column using the shortcut model, the reflux ratio is set to be 1.05 R/R_{min}, as recommended by Ray et al. (1998) and Douglas (1988) for sub-ambient distillation. Soave-Redlich-Kwong is used to calculate thermodynamic properties because of its accurate prediction for describing small nonpolar hydrocarbons**.**

In the scenario without heat integration, heating and cooling is provided by external utilities. A propylene refrigeration cycle is used to provide cooling to the condenser and the below-ambient cooler. Compared to distillation alone, using a membrane can reduce the total power demand by about 26 %; the compression required for refrigeration decreases by 33 % but recompression of the permeate is required.

In the heat-integrated scenario, there is direct process-to-process heat exchange between the membrane heater and the permeate cooler. Therefore, the need for hot and cold utilities is reduced and the net shaft power is reduced by 14 % compared to heat-integrated distillation. Part of the condenser cooling duty is rejected to the reboiler. The rest of the cooling duty of the condenser and the second permeate cooler are heat-pumped to the ambient heat sink, cooling water. Thus the optimised hybrid system shows a reduction in TOC of around 11 %, compared to the heat-integrated distillation.

Option 2: Sequential hybrid scheme

In this option, the same product purities, column feed specifications and membrane properties given in Table 1 and 2 are used. In this case, the configuration is fixed as shown in Figure 4; the operating conditions (stage cut and permeate pressure) are optimised. The bounds for permeate pressure are 1.01 and 8 bar; the stage cut bounds are 0.01 and 0.7.

By applying heat integration between the hybrid separation system and refrigeration system, the total power requirement is reduced by 44 % compared to distillation alone, and no hot utility is needed. These savings result from heat recovery. The total operating costs (TOC) for Option 2 are presented in Table 4. It can be seen that TOC is reduced by 14 % compared to conventional distillation. More energy saving is achieved by applying heat integration but total operating cost is reduced by 4 % compared to heat-integrated distillation.

Table 3: Variables bounds for Option 1: Parallel hybrid scheme

Variable	Lower bound	Upper bound
Membrane permeate pressure (bar)	1.01	8
Fraction of feed permeated	0.01	0.75
Side stream composition of C_2H_4 (mol %)	0.3	0.7
Molar flowrate of membrane feed (kmol h^{-1})	10	110
Membrane permeate pressure (bar)	1.01	8

Table 4: Optimisation for Options 1 and 2 (Parallel and sequential hybrid schemes)

Cases	Without heat integration		With heat integration	
	Power, MW	TOC, £/y	Power, MW	TOC, £/y
Conventional column	0.38	178,000	0.22	83,000
Sequential hybrid scheme	0.33	152,000	0.21	80,000
Parallel Hybrid scheme	0.28	127,000	0.19	74,000
Saving relative to distillation				
Parallel Hybrid scheme	26 %	28 %	14 %	11 %
Sequential hybrid scheme	14 %	14 %	4 %	4 %

4. Conclusions

A new approach to design and optimise hybrid membrane-distillation separation processes is presented. Since heat integration between the separation process and refrigeration system bring benefits to the process, it should be considered. To demonstrate the performance of the proposed approach, it has been applied to an ethylene/ethane separation case study. Results show that the parallel hybrid scheme can reduce the condenser duty by about 33 %. The total operating cost of the parallel hybrid scheme is reduced by 11 %, compared to the heat-integrated column. Compared to the parallel hybrid case (Option 1), the sequential hybrid case (Option 2) brings fewer economic and energy benefits.

Further scrutiny of the results (not presented here) confirms that the distillation models used gives reasonable predictions, compared to rigorous simulation results obtained from HYSYS. A major advantage of the approach is that it provides a systematic and quick method for investigating design alternatives, while taking into account opportunities for heat recovery.

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