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Optimization-based design of spiral-wound membrane systems for CO_2/CH_4 separations

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Abstract

A systematic design strategy for spiral-wound gas separation systems is studied using a recently proposed algebraic permeator model (Qi and Henson, Approximate modeling of spiral-wound gas permeators, J. Membrane Sci. 121 (1996) 11–24). Nonlinear programming (NLP) is used to determine operating conditions which satisfy the separation requirements while minimizing the annual process cost. The design method is applied to the separation of CO_2/CH_4 mixtures in natural gas treatment and enhanced oil recovery applications. It is shown that a two-stage configuration with permeate recycle and a three-stage configuration with residue recycle are suitable for natural gas treatment, while a three-stage configuration with both permeate and residue recycle is appropriate for enhanced oil recovery. Parameter sensitivities are analyzed by changing operating conditions, membrane properties, and economic parameters. The optimization procedure is sufficiently robust to handle multi-stage configurations with very demanding separation requirements. The proposed NLP design method facilitates the development of mixed-integer nonlinear programming design strategies which allow simultaneous optimization of the permeator configuration and operating conditions. © 1998 Elsevier Science B.V.

Keywords: Spiral-wound membranes; CO_2/CH_4 separations; Process design; Optimization

Nomenclature

a	dimensionless constant defined by Eq. (5)
A	membrane area (m^2)
B	constant associated with the spacing materials inside the spiral-wound leaf (m^2)
b	dimensionless constant defined by Eq. (5)
C	dimensionless constant defined by Eq. (2)
C''	permeate-side pressure parameter defined by Eq. (3) ($MPa^2 s/mol$)
d	thickness of membrane skin (m)
d_m	membrane leaf thickness (m)
f_{cc}	annual capital charge (%)
f_{cp}	capital cost of gas-powered compressors ($/KW)
f_{hv}	sales gas gross heating value (MJ/m^3)
f_{mh}	capital cost of membrane housing ($/$m^2$ membrane)
f_{mr}	expense of membrane replacement ($/$m^2$ membrane)
f_{mt}	maintenance rate (%)
f_{sg}	utility and sales gas price ($/$km^3$)
f_{wk}	working capital rate (%)
F	annual process cost ($/$km^3$)
h	$=l/L$, dimensionless leaf length variable
l	membrane leaf length variable (m)
L	membrane leaf length (m)
n	permeator stage number in a configuration

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N number of quadrature points of the integral $I(\gamma, y_r')$
P feed-side pressure (MPa)
p permeate-side pressure (MPa)
p_0 permeate-side pressure at permeate outlet (MPa)
$p_{0,n}$ permeate outlet pressure for permeator n (MPa)
Q_1 permeability of the more permeable component (mol/m s Pa)
Q_2 permeability of the less permeable component (mol/m s Pa)
R dimensionless permeation factor defined by Eq. (8)
R_g ideal gas constant (m^3 Pa/kg-mol K)
t_m membrane life (years)
t_{wk} annual working time (days)
T temperature (K)
U_f feed gas flow rate for each permeator (mol/s)
$U_{f,0}$ feed gas processing capacity (mol/s)
U_0 residue gas flow rate for each permeator (mol/s)
u feed-side gas flow rate per unit length of membrane leaf (mol/s m)
u_f feed gas flow rate per unit length of membrane leaf (mol/s m)
u_r residue gas flow rate per unit length of membrane leaf (mol/s m)
V_0 permeate flow rate at permeate outlet (mol/s)
W membrane leaf width (m)
W_{cp} compressor power (kW)
w_j quadrature weights
x local feed-side concentration (mole fraction)
x_0 bulk residue stream concentration (mole fraction)
x_f feed concentration (mole fraction)
x_r local residue concentration along residue outlet of membrane leaf (mole fraction)
y_0 permeate concentration in bulk permeate stream at permeate outlet (mole fraction)
y' local permeate concentration on the membrane surface (mole fraction)
y_f' local permeate concentration along feed end of membrane leaf (mole fraction)
y_j' local permeate concentration at quadrature point j (mole fraction)
y_r' local permeate concentration along outlet end of membrane leaf (mole fraction)
α $=Q_1/Q_2$, membrane selectivity
γ $=p/P$, ratio of permeate pressure to feed pressure
γ_0 $=p_0/P$, ratio of permeate pressure to feed pressure at permeate outlet
μ viscosity of gas mixture (Pa · s)
θ_0 $=V_0/U_f$, ratio of permeate flow to feed flow at permeate outlet
ξ_j quadrature points
ϕ $=u/u_f$, dimensionless feed-side flow rate
ϕ_r $=u_r/u_f$, dimensionless feed-side flow rate at residue outlet
η_{cp} compressor efficiency (%)

Subscripts

j index of quadrature point
n index of membrane stage

1. Introduction

Gas separation membranes are widely used for hydrogen recovery, air separation, and natural gas processing [1–3]. The success of membrane systems in these applications stems from their inherent advantages of low capital cost, operational simplicity, space and weight efficiency, easy scalability, and (in some applications) low power consumption. Spiral-wound permeators have been successfully used to separate CO_2/CH_4 mixtures encountered in natural gas treatment [4–6], and enhanced oil recovery [7–9].

In natural gas treatment applications, the feed gas usually is obtained directly from gas wells with a wide range of pressures (normally 2–7 MPa) and compositions (5–50% CO_2). The typical product is a CH_4-enriched residue stream containing less than 2% CO_2, which is sold as pipeline fuel. The residue stream is produced with essentially no pressure loss, while the CO_2-enriched permeate is produced at low pressure (0.1–0.5 MPa) but contains an appreciable amount of CH_4. The permeate stream often has some value as a low heating value fuel. In most situations, membrane processes can offer lower capital investment than a traditional amine absorption process.

As compared to natural gas treatment, the major difference in enhanced oil recovery application is that both the permeate and residue streams have considerable value. The residue stream (greater than 98% CH_4) is used as a fuel, while the permeate stream (greater than 95% CO_2) is compressed and re-injected into the gas well. Membrane systems are quite successful because of their effectiveness at high CO_2 compositions and convenience in field use. To satisfy separation requirements for both residue and permeate streams, multi-stage membrane processes usually are necessary.

The economic viability of a gas membrane system can be significantly affected by process design. The design of a membrane separation process involves the determination of: (1) the configuration of the permeators; and (2) the operating conditions of the individual permeators. As shown in Fig. 1, a wide variety of permeator configurations have been proposed for CO_2/CH_4 separations. It is important to note that an individual separation stage may actually consist of several permeators arranged in parallel since commercial permeators are designed for a specific range of flow rates. Single-stage systems with/without recycle [configurations (a) and (b)] are appropriate for applications in which the product purity and recovery requirements are moderate [6,9,10]. Multiple stages with/without recycle [configurations (c)–(g)], are typically required for more demanding separations [2,4,7,11]. The design of multi-stage systems is very complex because it usually is unfeasible to enumerate and evaluate all possible permeator configurations [2,12].

Several investigators have considered the design of multi-stage permeation systems for CO_2/CH_4 separations [2,10,13–18]. The conventional approach is to select a small number of system configurations and then optimize the operating

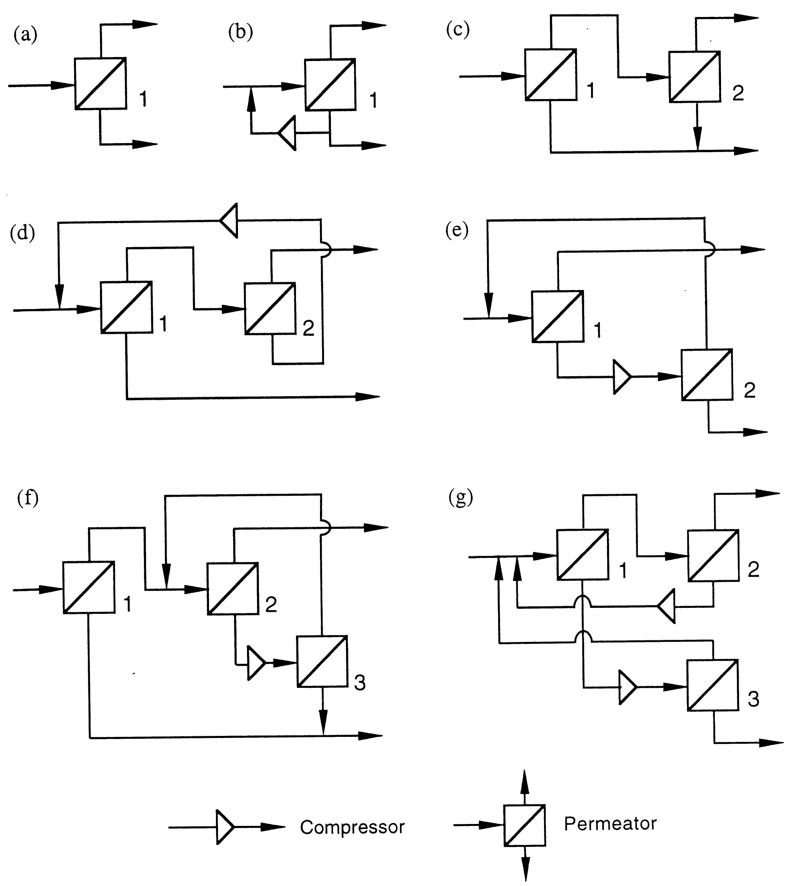

Fig. 1. Some proposed permeator configurations for CH_4/CO_2 separations: (a) single stage; (b) single stage with recycle; (c) two stage; (d) two stage with permeate recycle; (e) two stage with residue recycle; (f) three stage with residue recycle; and (g) three stage with permeate and residue recycle.

conditions of each network. The final design is chosen to be the membrane system which yields the most favorable economics. Spillman et al. [14] design membrane systems to separate CO_2/CH_4 mixtures encountered in natural gas treatment and enhanced oil recovery. The performance of each configuration shown in Fig. 1 is discussed, and a few configurations are optimized for a particular feed composition. However, the optimization method is not described in any detail. Babcock et al. [15] evaluate the economics of single-stage [configuration (a)] and three-stage [configuration (f)] membrane systems for a natural gas treatment application in which the residue stream must contain less than 2% CO_2. A variety of operating conditions are investigated, and the results are compared to those obtained with an amine treatment process. Despite these studies, more general comparisons of CO_2/CH_4 membrane system designs and more detailed description of the associated optimization methods are needed.

Bhide and Stern present detailed case studies of membrane separation systems for natural gas treatment [16,17] and oxygen enrichment of air [19,20]. By utilizing new optimization variables rather than the usual operating variables, a grid search method is used to optimize the operating conditions for several different configurations. However, the grid search procedure is much too inefficient for practical design problems. Pettersen and Lien [18] study the design of single-stage and multi-stage membrane systems using an algebraic model for hollow-fiber permeators. Because it neglects pressure drop inside the hollow fiber, the model does not yield accurate predictions when the feed has a mole fraction of the more permeable component greater than 0.3–0.5. As a result, the model is not appropriate for the design of multi-stage separation systems in which some permeators have a high feed CO_2 concentration.

Recently we proposed an approximate modeling technique for spiral-wound permeators separating binary [21] and multicomponent [22] gas mixtures. The development of the approximate models is based on simplifying basic transport models which include permeate-side pressure drop. The resulting models are ideally suited for process design because the nonlinear algebraic equations can be solved very efficiently and yield excellent prediction accuracy over a wide range of operating conditions. In this paper, we utilize the approximate binary model to develop a systematic design strategy for spiral-wound membrane processes. Nonlinear programming techniques are used to determine operating conditions which satisfy the separation requirements while minimizing the annual process cost. Our aim is to provide a general methodology for optimizing multi-stage gas membrane separation systems. The design procedure is applied to the separation of CO_2/CH_4 mixtures in natural gas treatment and enhanced oil recovery. We compare the performance of different permeator configurations and analyze parameter sensitivities. The results of these case studies have direct implications for other applications (e.g. hydrogen recovery, air separation) in which multiple permeation stages are employed.

2. Approximate spiral-wound permeator model

In this section, we describe the nonlinear algebraic equations which comprise the approximate permeator model. A more detailed discussion of the model is given elsewhere [21]. The model development is based on Fig. 2, which depicts

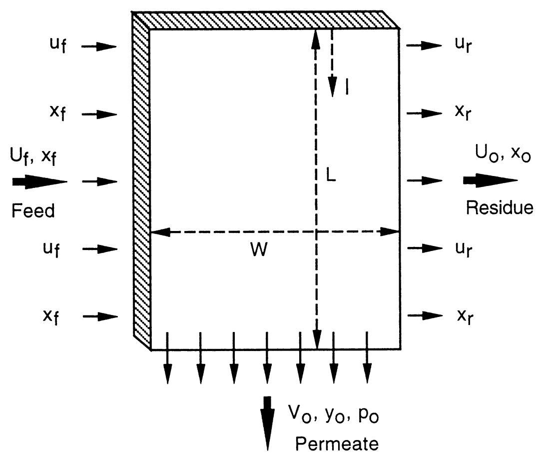

Fig. 2. Gas permeation through an extended spiral-wound membrane leaf.

permeation through an extended leaf of a spiral-wound permeator. The first equation describes the permeate-side pressure distribution,

$$\gamma^2=\gamma_0^2+\tfrac{1}{2}C(1-\phi_r)(1-h^2), \tag{1}$$

where γ is the ratio of the permeate-side and feed-side pressures; γ_0 is γ at the permeate outlet; ϕ_r is the dimensionless residue gas flow rate; h is the dimensionless membrane leaf length variable; and:

$$C=\frac{2R_g T\mu L U_f}{W d_m B P^2}. \tag{2}$$

The remaining variables are defined in the Nomenclature. The pressure coefficient C can be factored into a form involving the membrane area and the feed conditions:

$$C=C''\frac{U_f}{AP^2} \tag{3}$$

where U_f is the feed gas flow rate, P is the feed-side pressure, A is the membrane area, and C'' is a parameter that depends on the internal properties of the permeator. The second equation describes the effect of γ and the local permeate concentration y' on the ratio of local residue flow rate to the feed flow rate ϕ,

$$\phi(\gamma,y')=\left(\frac{y'}{y'_f}\right)^a\left(\frac{1-y'}{1-y'_f}\right)^b\left(\frac{\alpha-(\alpha-1)y'}{\alpha-(\alpha-1)y'_f}\right), \tag{4}$$

where y'_f is y' at the feed inlet, α is the membrane selectivity, and:

$$a=\frac{\gamma(\alpha-1)+1}{(\alpha-1)(1-\gamma)},$$

$$b=\frac{\gamma(\alpha-1)-\alpha}{(\alpha-1)(1-\gamma)}. \tag{5}$$

Therefore, the dimensionless feed-side flow rate at residue outlet can be written as,

$$\phi_r=\phi(\gamma,y'_r), \tag{6}$$

where y'_r is y' along the residue outlet of the membrane leaf.

The third equation describes the relation between the dimensionless permeation factor,

$$R=\frac{2WLQ_2P}{dU_f}=A\frac{Q_2}{d}\frac{P}{U_f}, \tag{7}$$

and the local permeate concentration along the residue outlet y'_r:

$$R=\frac{1}{\alpha(1-\gamma)}\{\alpha-(\alpha-1)y'_f-[\alpha-(\alpha-1)y'_r]\times\phi(\gamma,y'_r)-(\alpha-1)I(\gamma,y'_r)\}. \tag{8}$$

Here $I(\gamma,y'_r)$ is an integral function which is approximated using Gaussian quadrature [23],

$$I(\gamma,y'_r)=(y'_r-y'_f)\sum_{j=1}^{N}\phi(\gamma,y'_j)w_j, \tag{9}$$

where N is the number of quadrature points, w_j is the quadrature weight at quadrature point ξ_j, and:

$$y'_j=y'_f+\xi_j(y'_r-y'_f). \tag{10}$$

The fourth equation describes the relation between the local feed-side concentration x and the local permeate concentration y':

$$\frac{y'}{1-y'}=\frac{\alpha(x-\gamma y')}{1-x-\gamma(1-y')}. \tag{11}$$

Simultaneous solution of Eqs. (1), (6), (8) and (11) with $x=x_f$ and $y'=y'_f$ at the quadrature points yields $\gamma(h)$, $\phi_r(h)$, $y'_r(h)$, and $y'_f(h)$ [21]. The local residue concentration $x_r(h)$, which is the local feed-side concentration along the residue end of the membrane leaf, is obtained from Eq. (11) with $y'=y_r$.

The flow rate and concentration of the effluent permeate stream are calculated from integral expressions which can be approximated using Gaussian quadrature. For most operating conditions, a single quadrature point at $h_1=0.5$ is sufficient. In this case, the resulting equations are [21]:

$$\theta_0=1-\phi_r(h_1), \tag{12}$$

$$y_0=\frac{x_f-x_r(h_1)\phi_r(h_1)}{1-\phi_r(h_1)}. \tag{13}$$

The flow rate and concentration of the effluent residue stream are determined from an overall material balance about the permeator.

3. Nonlinear programming strategy

The optimal design of a gas membrane separation system involves a tradeoff between capital investment and operating expenses. Using the following nonlinear programming strategy and the approximate permeator model, we compare the system configurations shown in Fig. 1 for binary CO_2/CH_4 separations. Both natural gas treatment and enhanced oil recovery applications are considered. The design strategy is based on the following nonlinear optimization problem:

Minimize: annual process cost.
Subject to: operating requirements;
material balances for each mixing/splitting point;
model equations for each permeator;
non-negativity constraints.

An example of the mathematical formulation for a three-stage configuration [Fig. 1(g)] is shown in the Appendix. Other configurations in Fig. 1 are formulated similarly. The nominal economic costs and operating parameters are obtained from Spillman et al. [14], Babcock et al. [15], and Lee et al. [24,25].

- Operating conditions:
  natural gas processing capacity, $U_{f,\,0}=10$ mol/s (19 353 m^3/day);
  working time, $t_{wk}=300$ days/year;
  feed pressure, $P=3.5$ MPa;
  product permeate pressure, $p_0=0.105$ MPa;
  temperature, $T=40°C$;
  feed CO_2 concentration, $x_{f,\,0}=0.20$.

- Operating requirements:
  outlet residue CO_2 concentration less than 2% (both applications);
  outlet permeate CO_2 concentration greater than 95% (enhanced oil recovery only);
  outlet permeate pressure for each stage not less than 0.105 MPa (to avoid negative pressure operation).

- Membrane properties:
  CH_4 permeability: $Q_2/d=1.48\times10^{-3}$mol/MPa m^2s(0.0119m^3_{STP}/m^2h)
  CO_2/CH_4 selectivity, $\alpha=20$;
  pressure parameter, $C''=9.32$ MPa^{2}m^{2}s/mol.

- Capital investment:
  membrane housing, $f_{mh}=200$ \$/m^2 membrane;
  gas-powered compressors, $f_{cp}=1000$ \$/kW;
  compressor efficiency, $\eta_{cp}=70\%$;
  working capital, $f_{wk}=10\%$ of fixed capital investment;
  capital charge, $f_{cc}=27\%$ per year.

- Operating expenses:
  membrane replacements, $f_{mr}=90$ \$/m^2 membrane;
  membrane lifetime, $t_m=3$ years;
  maintenance, $f_{mt}=5\%$ of fixed capital investment;
  utility and sale gas price, $f_{sg}=35$ \$/km^3;
  sale gas gross heating value, $f_{hv}=43$ MJ/m^3;
  lost CH_4 is converted to sales gas value.

The gas volumes are calculated at standard conditions (0.102 MPa, 273 K). The pressure parameter C'' is based on an estimated value obtained from experiment data [21,24]. In this work, we assume the membrane area can be varied continously by changing the membrane width. This can be achieved by adding or removing membrane elements.

4. Natural gas treatment

For natural gas treatment, the residue CO_2 concentration must be less than 2%, while no constraint is placed on the permeate concentration. For the nominal operating conditions presented earlier, each configuration in Fig. 1 is optimized to minimize the annual cost of the membrane system. The results are shown in Table 1. The configuration with three stages and residue recycle (f) yields the lowest cost, while the two-stage system with residue recycle (e) is most expensive.

4.1. Effect of feed conditions

Fig. 3 shows the effect of feed composition on the process cost, total membrane area, compressor

Table 1
Optimal separation systems at nominal conditions for natural gas treatment

Configuration	F ($/Km^3)	n	A_n (m^2)	$W_{cp,n}$ (KW)	$p_{0,n}$ (MPa)	$U_{f,n}$ (mol/s)	$x_{f,n}$	$U_{0,n}$ (mol/s)	$x_{0,n}$	$V_{0,n}$ (mol/s)	$y_{0,n}$
a and b	11.874	1	352.75	—	0.1050	10.00	0.2000	6.51	0.0200	3.49	0.5353
c	11.692	1	142.15	—	0.1050	10.00	0.2000	8.07	0.0850	1.93	0.6769
		2	205.40	—	0.1050	8.07	0.0850	6.53	0.0200	1.53	0.3619
d	11.276	1	231.54	—	0.1050	11.08	0.2096	8.20	0.0567	2.88	0.6440
		2	157.96	9.86	0.1050	8.20	0.0567	7.12	0.0200	1.08	0.2984
e	12.747	1	424.30	37.96	0.1050	12.09	0.1954	7.93	0.0200	4.16	0.5300
		2	67.81	—	0.1050	4.16	0.5300	2.09	0.1735	2.07	0.8907
f	11.204	1	180.89	—	0.1050	10.00	0.2000	7.71	0.0659	2.29	0.6511
		2	184.97	11.90	0.1050	8.52	0.0660	7.22	0.0200	1.30	0.3209
		3	29.84	—	0.1050	1.30	0.3209	0.82	0.0678	0.49	0.7437
g	12.574	1	320.16	31.98	0.1050	12.10	0.1959	8.59	0.0367	3.51	0.5860
		2	101.15	5.89	0.1050	8.59	0.0367	7.95	0.0200	0.65	0.2429
		3	64.30	—	0.1050	3.51	0.5860	1.45	0.1465	2.05	0.8973

F, annual process cost; n, permeator stage number; A, membrane area; W_{cp}, compressor power; p_0, outlet permeate pressure; U_f, feed flow rate; x_f, feed concentration; U_0, residue flow rate; x_0, residue concentration; V_0, permeate flow rate; y_0, permeate concentration.

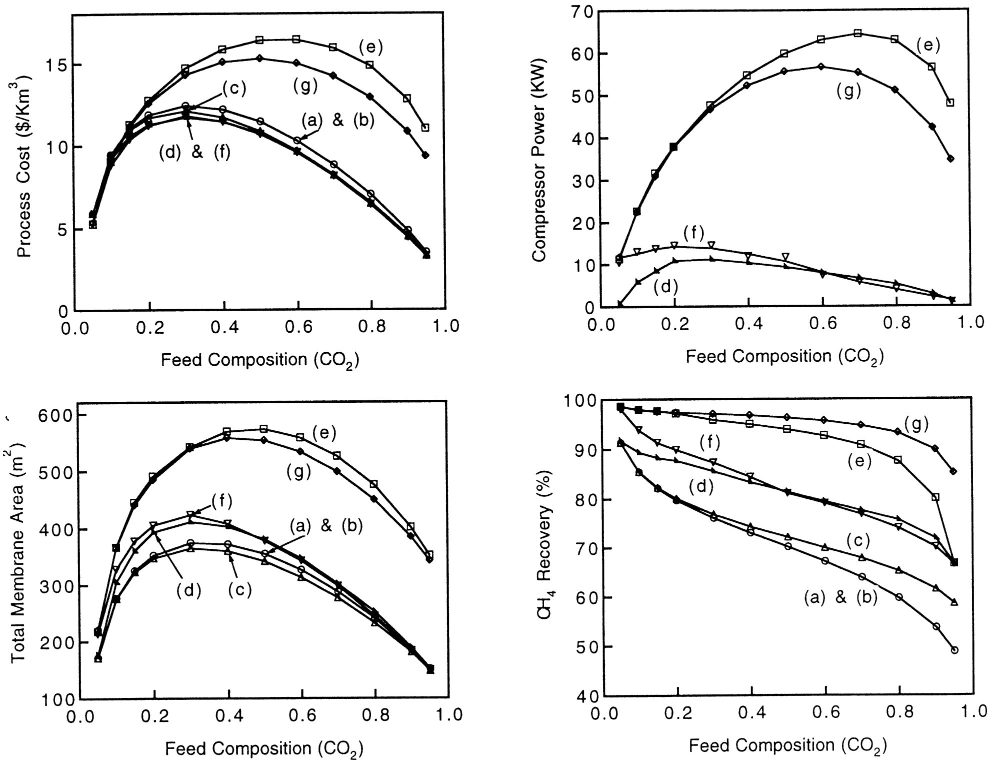

Fig. 3. Effect of feed composition on optimal separation system for natural gas treatment: (a) single stage; (b) single stage with recycle; (c) two stage; (d) two stage with permeate recycle; (e) two stage with residue recycle; (f) three stage with residue recycle; and (g) three stage with permeate and residue recycle.

power, and CH_4 recovery for each configuration in Fig. 1. Note that each configuration has a maximum cost with respect to the CO_2 concentration of the feed gas. Single stage with recycle [configuration (b)] always yields single stage without recycle [configuration (a)], which indicates permeate recycle is not favorable for single-stage systems. As expected, systems without recycle [configurations (a) and (c)] provide low CH_4 recoveries. However, they yield moderate process costs because small membrane areas and no compressors are needed. The two-stage system with permeate recycle [configuration (d)] and three-stage system with residue recycle [configuration (f)] have the lowest process costs, mainly because they can increase CH_4 recovery with moderate power consumption. The two-stage system with residue recycle [configuration (e)] and three-stage system with permeate and residue recycle [configuration (g)] provide unnecessarily high CH_4 recoveries at the expense of large membrane area and high power consumption. Therefore, the process costs for these two configurations are very high. These results are consistent with those obtained by Spillman et al. [14] and Babcock et al. [15].

The effect of feed pressure on different configurations is shown in Fig. 4. Only three configurations [(a), (d) and (f)] are investigated as a result of their relatively low process costs. It is important to note we have assumed there is no cost associated with increasing the feed pressure. Increasing feed pressure improves the process costs and the CH_4 recoveries, especially when the pressure is less than 40 MPa. Although not shown here, we also have investigated the effect of feed flow rate. Increasing feed flow rate increases the membrane area and compressor power linearly, while the process cost and CH_4 recovery essentially are unchanged. This behavior occurs because the permeator model does not account for concentration polarization [1].

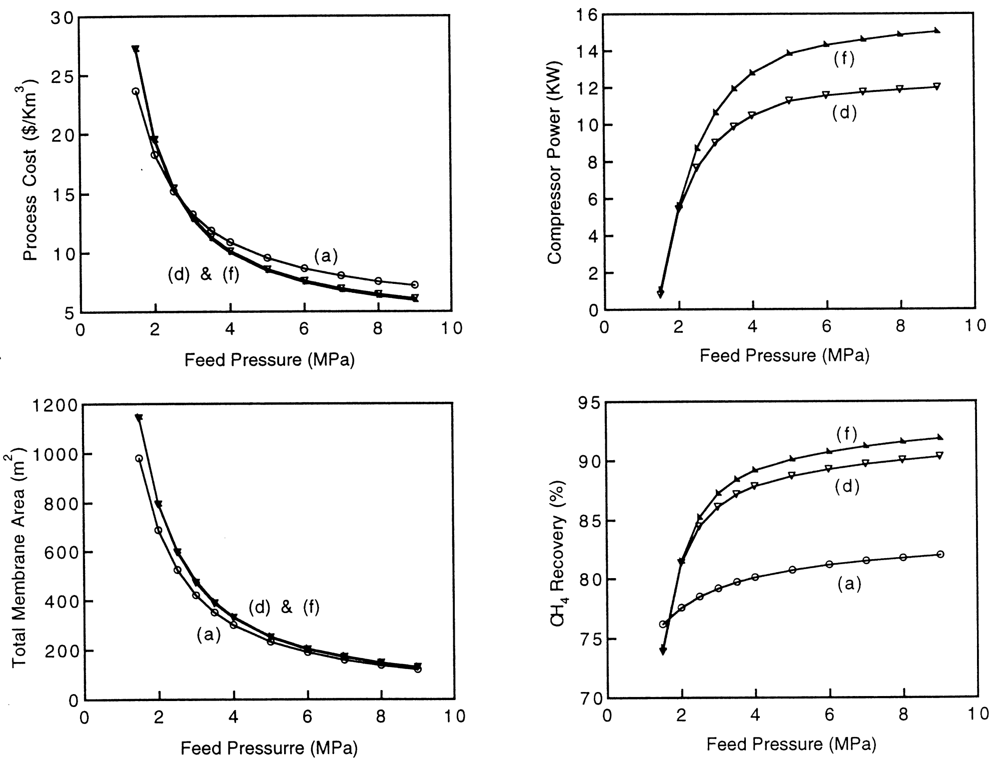

Fig. 4. Effect of feed pressure on optimal separation system for natural gas treatment: (a) single stage; (d) two stage with permeate recycle; and (f) three stage with residue recycle.

The feed flow rate has no effect on the optimal operating conditions, but it does change the size of the required equipment.

4.2. Effect of membrane properties

Fig. 5 shows the effect of membrane selectivity on three configurations [(a), (d) and (f)] with the most favorable economics. As expected, increasing selectivity significantly decreases the process cost and increases CH_4 recovery, especially when the selectivity is relatively low. Although not shown here, we have investigated the effect of parameter C'' from 0 to 30 $MPa^2 m^2 s/mol$ for the same three configurations. As expected, the process cost is the lowest if there is no permeate pressure drops (i.e. $C''=0$). As the permeate pressure drop increases, the process cost increases as more membrane area and compressor power are needed, while the CH_4 recovery decreases.

4.3. Effect of economic parameters

To investigate the effect of economic parameters on the optimal separation system, we consider variations in the capital cost of membrane housing and compressors, the expense of membrane replacement, and the price of the sales gas. For systems without recycle [configurations (a) and (c)], the operating conditions are determined solely by the membrane properties and separation requirements because no extra degree of freedom are available for economic optimization. For these configurations, variations in economic parameters change the process cost linearly and the operating conditions do not change. Therefore, we consider only the multi-stage configurations (d) and (f).

Varying membrane housing capital from 50 to 700 $\$/m^2$ membrane results in small changes in optimal operating conditions and CH_4 recoveries, while the process cost increases in linear fashion.

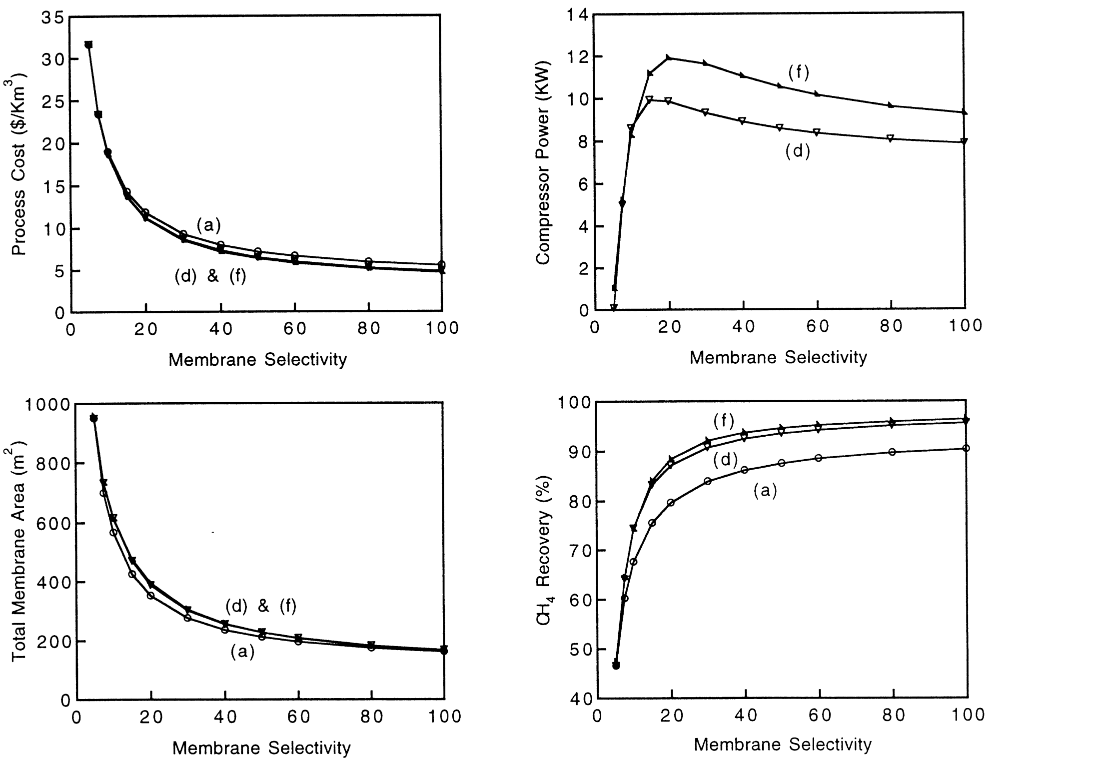

Fig. 5. Effect of membrane selectivity on optimal separation system for natural gas treatment: (a) single stage; (d) two stage with permeate recycle; and (f) three stage with residue recycle.

This indicates that membrane housing capital does not have a strong effect on the optimal separation system. The effect of compressor capital cost on the two configurations is shown in Fig. 6. The process cost increases with increasing compressor cost. However, the cost increase can be reduced substantially by optimizing the operating conditions. To balance the increased compressor cost, the compressor power and membrane area are reduced at the expense of decreased CH_4 recovery. Note that the second and third stages have very small membrane areas when compressor capital cost is very high, at which point both configurations effectively reduce to the single-stage configuration (a).

As shown in Fig. 7, the sales gas price has a strong effect on optimal operating conditions. Note that there is an intersection point where the process cost for the two configurations are identical. Configuration (d) is preferred when the sales gas price is low, while configuration (f) has better performance at high sales gas prices because it yields higher CH_4 recoveries. We also have investigated the effect of the membrane replacement expense from 30–150 \$/m^2 membrane. Although not shown here, these variations have no effect on the optimal operating conditions, while the process cost increases linearly with increasing replacement cost.

5. Enhanced oil recovery

The separation of CO_2/CH_4 in enhanced oil recovery differs from that in natural gas treatment because both the residue and permeate streams must satisfy concentration requirements. For single-stage configurations [(a) and (b)] and some

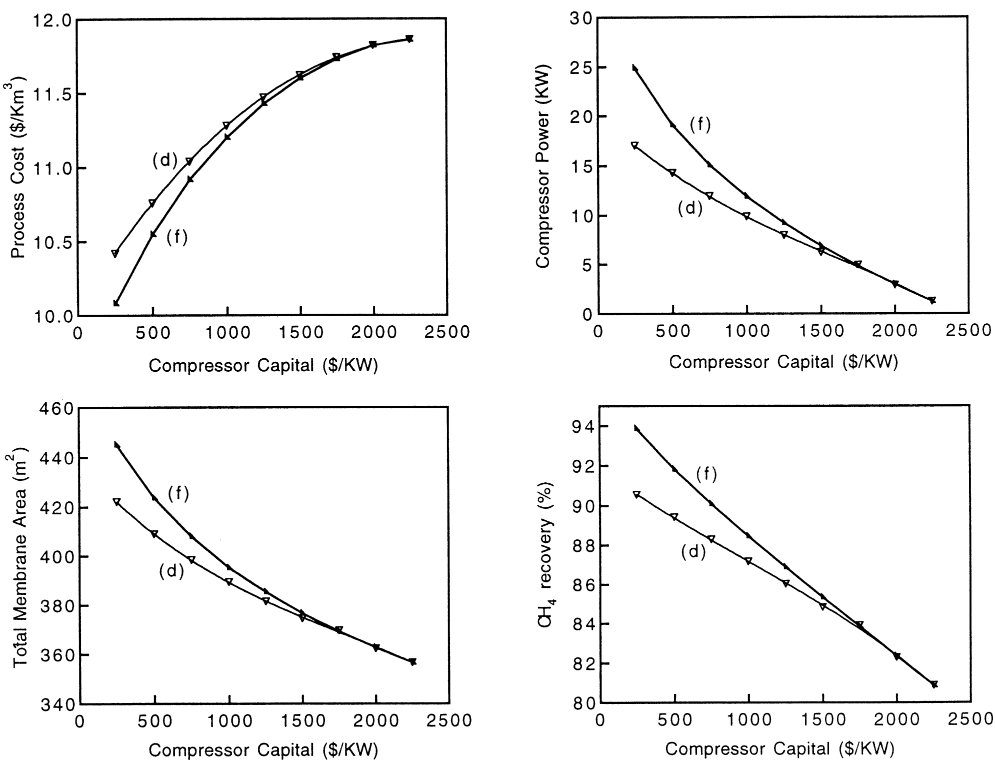

Fig. 6. Effect of compressor capital on optimal separation system for natural gas treatment: (d) two stage with permeate recycle; and (f) three stage with residue recycle.

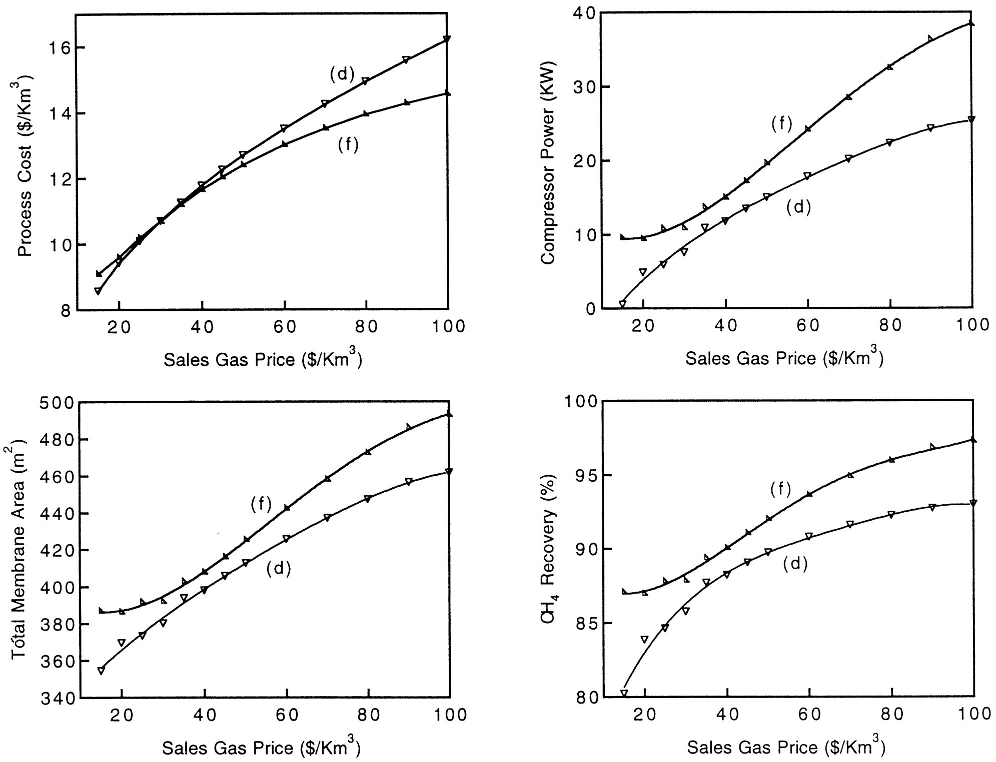

Fig. 7. Effect of sale gas price on optimal separation system for natural gas treatment: (d) two stage with permeate recycle; and (f) three stage with residue recycle.

two-stage configurations [(c) and (d)], the additional constraint that the permeate concentration must contain at least 95% CO_2 causes infeasibilities for a wide range of feed compositions. In fact, these configurations are feasible only when the feed CO_2 concentration is greater than 80%. Therefore, the subsequent analysis is restricted to configurations (e), (f) and (g). Table 2 gives the optimal separation system for these configurations at the nominal operating conditions.

5.1. Effect of feed conditions

Fig. 8 shows a comparison of the three configurations for different feed compositions. The three-stage system with permeate and residue recycle [configuration (g)] provides the lowest process cost for feed CO_2 compositions less than 50%. For higher feed concentration, the three-stage system with residue recycle [configuration (f)] becomes more cost effective. At feed compositions less than 40% CO_2, the first-stage membrane area of configuration (f) goes to zero, and the three-stage configuration becomes identical to the two-stage configuration (e). The effect of feed pressure at the nominal feed composition is shown in Fig. 9. As expected, increasing feed pressure always yields lower process cost, less membrane area, and less compressor power. It is interesting to note that configuration (g) has the lowest cost for all feed pressures considered.

5.2. Effect of economic parameters

The effect of membrane housing capital, compressor capital, sales gas price, and membrane replacement expense on the optimal operating conditions for the three configurations also has been investigated. It is interesting to note that variations in these parameters have much less

Table 2
Optimal separation systems at nominal conditions for enhanced oil recovery

Configuration	F (\$/Km^3)	n	A_n (m^2)	$W_{cp,n}$ (KW)	$p_{0,n}$ (MPa)	$U_{f,n}$ (mol/s)	$x_{f,n}$	$U_{0,n}$ (mol/s)	$x_{0,n}$	$V_{0,n}$ (mol/s)	$y_{0,n}$
e	15.355	1	530.69	56.48	0.1050	14.25	0.2780	8.06	0.0200	6.19	0.6142
		2	41.37	—	0.1050	6.19	0.6142	4.25	0.4614	1.94	0.9500
f	15.467	1	0.54	—	0.1050	10.00	0.2000	9.99	0.1994	0.01	0.7797
		2	533.84	57.13	0.1050	14.33	0.2808	8.06	0.0200	6.26	0.6166
		3	40.82	—	0.1050	6.26	0.6166	4.34	0.4682	1.93	0.9509
g	13.281	1	236.98	28.87	0.1272	13.14	0.2348	9.79	0.0775	3.35	0.6947
		2	236.30	15.74	0.1050	9.79	0.0775	8.06	0.0200	1.73	0.3465
		3	41.31	—	0.1050	3.35	0.6947	1.41	0.3446	1.94	0.9500

F, annual process cost; n, permeator stage number; A, membrane area; W_{cp}, compressor power; p_0, outlet permeate pressure; U_f, feed flow rate; x_f, feed concentration; U_0, residue flow rate; x_0, residue concentration; V_0, permeate flow rate; y_0, permeate concentration.

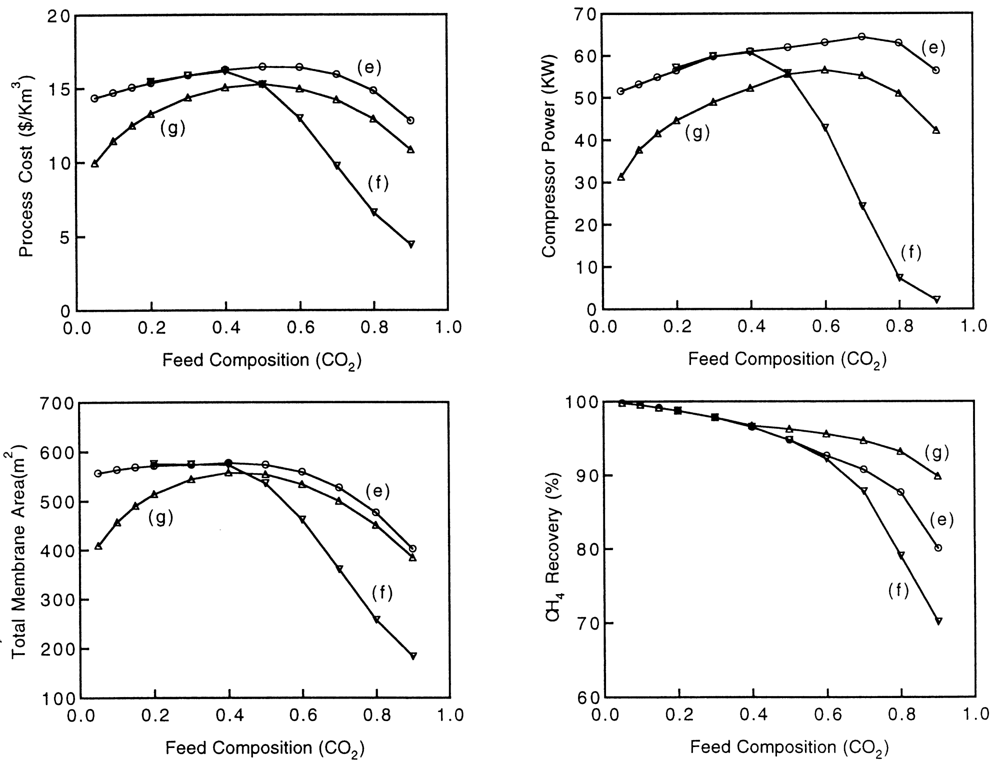

Fig. 8. Effect of feed composition on optimal separation system for enhanced oil recovery: (e) two stage with residue recycle; (f) three stage with residue recycle; and (g) three stage with permeate and residue recycle.

impact on process economics than in natural gas treatment. For each configuration, optimal operating conditions are not changed significantly by variations in membrane housing from 50–700 \$/m^2 membrane, compressor capital from 250–2500 \$/KW, sales gas price from 15–100 \$/km^3, and membrane replacement from 15–180 \$/m^2 membrane. The process cost increases linearly with the economic parameters, while the CH_4 recovery is always 98.79% for all configura-

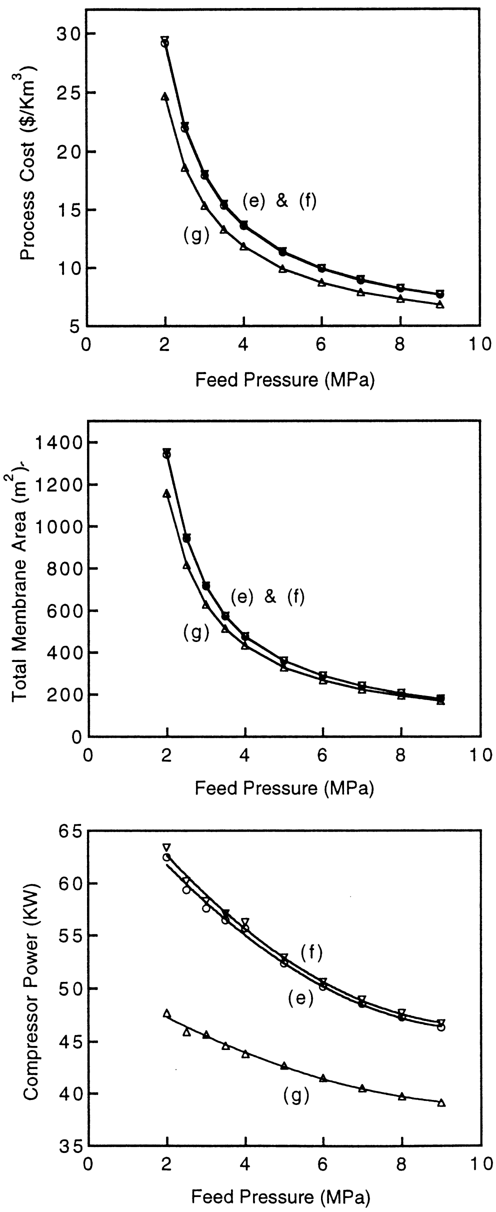

Fig. 9. Effect of feed pressure on optimal separation system for enhanced oil recovery: (e) two stage with residue recycle; (f) three stage with residue recycle; and (g) three stage with permeate and residue recycle.

tions. This is attributable to the fact that the optimal operating conditions are dominated by the separation requirements rather than economic parameters in the objective function.

5.3. *Effect of separation requirements*

For the three-stage system with permeate and residue recycle [configuration (g)], we investigate the effect of residue and permeate composition constraints on the optimal operating conditions for a feed CO_2 concentration of 20%. Fig. 10 shows the effect of the residue constraint (nominally 2%) on the optimal separation system. The process cost increases rapidly when the maximum CO_2 concentration in the residue is decreased as a result of increased membrane area and compressor power. It is interesting to note that the second-stage permeator and compressor are strongly affected, while the other permeators and the other compressor are effectively unchanged. The effect of the permeate composition constraint (nominally 95%) is shown in Fig. 11. The process cost increases significantly as the constraint becomes more stringent. As before, the second-stage permeator and compressor are most strongly affected by variations in this constraint. These results indicate that the proposed model and optimization procedure are sufficiently robust to optimize permeator systems with very stringent separation requirements.

6. Summary and conclusions

A systematic design strategy for spiral-wound membrane systems based on an algebraic permeator model and nonlinear programming has been proposed. Case studies for the separation of CO_2/CH_4 mixtures in natural gas treatment and enhanced oil recovery have been presented. Sensitivity of the proposed designs has been investigated by changing operating conditions, membrane properties, and economic parameters. It is shown that a two-stage configuration with permeate recycle and a three-stage configuration with residue recycle are suitable for natural gas treatment, while three-stage configurations with permeate and/or residue recycle are appropriate for enhanced oil recovery. The proposed design method provides an efficient tool for optimizing multistage membrane separation systems, including those with stringent separation requirements.

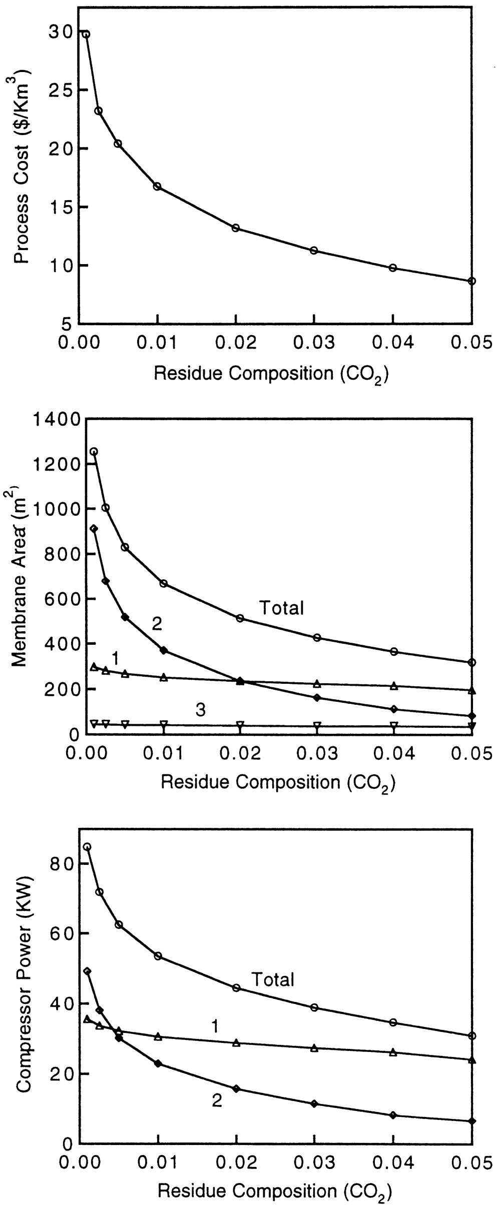

Fig. 10. Effect of residue composition constraint on optimal separation system for enhanced oil recovery.

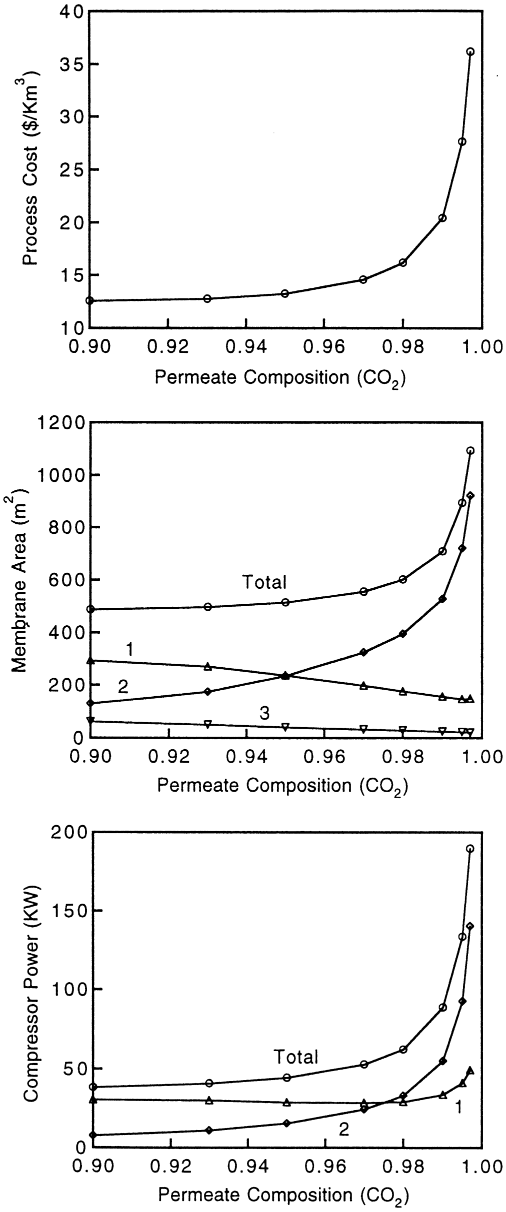

Fig. 11. Effect of permeate composition constraint on optimal separation system for enhanced oil recovery.

Future work will focus on incorporating the proposed method into a mixed-integer nonlinear programming design strategy which allows simultaneous optimization of the permeator configuration and operating conditions.

Acknowledgment

Financial support from Praxair is gratefully acknowledged.

Appendix

We present the formulation of the nonlinear optimization problem for a three-stage system with permeate and residue recycle [Fig. 1(g)] for the enhanced oil recovery application. The objective function represents the annual process cost of the membrane system. The operating requirements, material balances for each mixing point, and model equations for each permeator are posed as constraints. The nonlinear programming problem is solved with GAMS/CONOPT software running on an IBM RS-6000 workstation. It is important to note that the formulation usually yields a nonconvex optimization problem. As a result, the solution obtained represents a local optimum. This problem is addressed by checking several initial conditions. A single solution is typically obtained in a few seconds.

Annual process cost
The annual process cost is expressed as:

$$F=[f_{cc}(1+f_{wk})F_{fc}+F_{mr}+f_{mt}F_{fc}+F_{ut}+F_{pl}]/(U_{f,0}t_{wk}). \tag{A1}$$

Fixed capital investment:

$$F_{fc}=f_{mh}\sum_{n=1}^{3}A_n+f_{cp}W_{cp}/\eta_{cp}. \tag{A2}$$

Expense of membrane replacement:

$$F_{mr}=\frac{f_{mr}}{t_m}\sum_{n=1}^{3}A_n. \tag{A3}$$

Cost of utilities:

$$F_{ut}=f_{sg}t_{wk}W_{cp}/(f_{hv}\eta_{cp}). \tag{A4}$$

Value of lost gas:

$$F_{pl}=f_{sg}t_{wk}V_{0,3}(1-y_{0,3})/(1-x_{0,2}). \tag{A5}$$

Compressor power [26]:

$$W_{cp}=R_gT\sum_{n=1}^{2}V_{0,n}\ln\left(\frac{P}{p_{0,n}}\right). \tag{A6}$$

Operating requirements

$$x_{0,2}\le 0.02, \tag{A7}$$

$$y_{0,3}\ge 0.95, \tag{A8}$$

$$p_{0,n}\ge 0.105, \tag{A9}$$

$$p_{0,3}=0.105. \tag{A10}$$

Material balances for mixing point

$$U_{f,1}=U_{f,0}+V_{0,2}+U_{0,3}, \tag{A11}$$

$$U_{f,1}x_{f,1}=U_{f,0}x_{f,0}+V_{0,2}y_{0,2}+U_{0,3}x_{0,3}, \tag{A12}$$

$$U_{f,2}=U_{0,1}, \tag{A13}$$

$$x_{f,2}=x_{0,1}, \tag{A14}$$

$$U_{f,3}=V_{0,1}, \tag{A15}$$

$$x_{f,3}=y_{0,1}. \tag{A16}$$

Model equations for permeator n

$$U_{f,n}=U_{0,n}+V_{0,n}, \tag{A17}$$

$$U_{f,n}x_{f,n}=U_{0,n}x_{0,n}+V_{0,n}y_{0,n}, \tag{A18}$$

$$C_n=C''U_{f,n}/(A_nP^2), \tag{A19}$$

$$R_n=(Q_2/d)A_nP/U_{f,n}, \tag{A20}$$

$$V_{0,n}=U_{f,n}\theta_{0,n}, \tag{A21}$$

$$p_{0,n}=P\gamma_{0,n}, \tag{A22}$$

$$\gamma_n^2=\gamma_{0,n}^2+0.375C_n(1-\phi_{r,n}), \tag{A23}$$

$$a_n=[\gamma_n(\alpha-1)+1]/[(\alpha-1)(1-\gamma_n)], \tag{A24}$$

$$b_n=[\gamma_n(\alpha-1)-\alpha]/[(\alpha-1)(1-\gamma_n)], \tag{A25}$$

$$y'_{j,n}=y'_{f,n}+\xi_j(y'_{r,n}-y'_{f,n}), \tag{A26}$$

$$\phi_{j,n}=\left(\frac{y'_{j,n}}{y'_{f,n}}\right)^{a_n}\left(\frac{1-y'_{j,n}}{1-y'_{f,n}}\right)^{b_n}\left(\frac{\alpha-(\alpha-1)y'_{j,n}}{\alpha-(\alpha-1)y'_{f,n}}\right), \tag{A27}$$

$$I_n = (y'_{r,n} - y'_{f,n}) \sum_{j=1}^{3} \phi'_{j,n} w_j, \tag{A28}$$

$$\phi_{r,\,n} = \left(\frac{y'_{r,n}}{y'_{f,n}}\right)^{a_n} \left(\frac{1-y'_{r,n}}{1-y'_{f,n}}\right)^{b_n} \left(\frac{\alpha-(\alpha-1)y'_{r,n}}{\alpha-(\alpha-1)y'_{f,n}}\right), \tag{A29}$$

$$\alpha(1-\gamma_n)R_n = \alpha-(\alpha-1)y'_{f,\,n} - [\alpha-(\alpha-1)y'_{r,n}]\phi_{r,n} - (\alpha-1)I_n, \tag{A30}$$

$$y'_{f,\,n}/(1-y'_{f,n}) = \alpha(x_{f,n}-\gamma_n y'_{f,n})/[1-x_{f,n}-\gamma_n(1-y'_{f,n})], \tag{A31}$$

$$y'_{r,n}/(1-y'_{r,n}) = \alpha(x_{r,n}-\gamma_n y'_{r,n})/[1-x_{r,n}-\gamma_n(1-y'_{r,n})], \tag{A32}$$

$$\theta_{0,\,n} = 1-\phi_{r,n}, \tag{A33}$$

$$y_{0,\,n} = (x_{f,n}-x_{r,n}\phi_{r,n})/(1-\phi_{r,n}). \tag{A34}$$

Non-negativity constraints

$$U_{f,n}, U_{0,n}, V_{0,n}, p_{0,n}, A_n, R_n, C_n \geq 0, \tag{A35}$$

$$0 \leq x_{f,n}, x_{0,n}, y_{0,n}, y'_{f,n}, y'_{r,n}, y'_{j,n}, x_{r,n} \leq 1, \tag{A36}$$

$$0 \leq \theta_{0,n}, \gamma_{0,n}, \gamma_n, \phi_{r,n}, \phi_{j,n} \leq 1$$
$$n = 1,2,3;\; j = 1,2,3. \tag{A37}$$

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