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Effect of Feed Location on the Performance of Single-Stage Membrane Permeators

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Abstract

The effect of feed location on the performance of single-stage membrane permeators was determined based on the minimum unit compressor load (recycle ratio). Since certain feed locations correspond to several well-known permeator configurations (e.g., simple recycle permeator, continous membrane column), it is possible to characterize the relative performance of these configurations for separating binary gas mixtures. For separations involving oxygen, nitrogen, and carbon dioxide, it was found that the location of feed introduction was related to the apparent difficulty of separation. For binary seprations of low to moderate difficulty, the optimum feed location was at a dimensionless axial distance of 0.6 to 0.75 from the top of the column. This feed location corresponded to the continuous membrane column configuration. For difficult separations, the optimum feed location was at the top of the column which corresponded to the simple recycle permeator. Based on this study, the simple recycle permeator configuration outperforms the continuous membrane column for the most difficult separations such as in the separation of oxygen from air. However, the continuous membrane column configuration can be used effectively for less difficult gas separations which cannot be accomplished by a membrane permeator without recycle, but do not require high recycle ratios to achieve the desired separation.

INTRODUCTION

Membrane separations are of increasing importance in a variety of industrial applications, especially in the separation of gases (*1–3*). Several review articles (*4*, *5*) and texts (*6*, *7*) give a broad overview of membrane technology. In particular, the Prism separator developed by Monsanto demonstrates the commercial feasibility of industrial gas separation by membrane proceses (*8*, *9*). As shown in Fig. 1, membrane permeators come in a variety of configurations including single-stage or cascade; countercurrent, cocurrent, or well-mixed; single membrane or multiple membrane; with or without recycle; and combinations of these configurations. Regardless of the specific permeator configuration chosen, the objective is to obtain one or more high purity fractions in sufficient quantity at a reasonable cost. Thus, a good membrane separator must maximize product specificity and throughput while minimizing membrane area and compression cost.

The residue product leaving a membrane permeator can be stripped completely of the more permeable gas* if the permeator is of sufficient length (*10*). Thus, it is possible to obtain the less permeable gas in essentially pure form in the residue stream. However, it is not possible to obtain a pure overhead product stream since the product composition cannot exceed the maximum composition of the more permeable gas passing through the membrane. The enrichment limit in a conventional stripper is given by

$$Y = \frac{X}{1 + X} \tag{1}$$

where

$$X = \alpha\left(\frac{x}{x - 1}\right) \tag{2}$$

With this enrichment limitation, a conventional stripper may be unable to produce a sufficiently pure overhead product stream. The enrichment capability of a single-stage membrane permeator may be increased by recycling. By extracting a portion of the enriched product and returning the remainder back to the permeator, the enrichment potential is increased significantly. The degree of enrichment obtainable is a function of membrane selectivity, cut, membrane area, unit com-

*It should be noted that permeability is a property of the membrane rather than of the gas; however, it is common to refer to the "more permeable" gas as the gas of which the membrane has the greatest permeability. Likewise, the "less permeable" gas is the gas of which the membrane has the least permeability. The convention will be used in this paper.

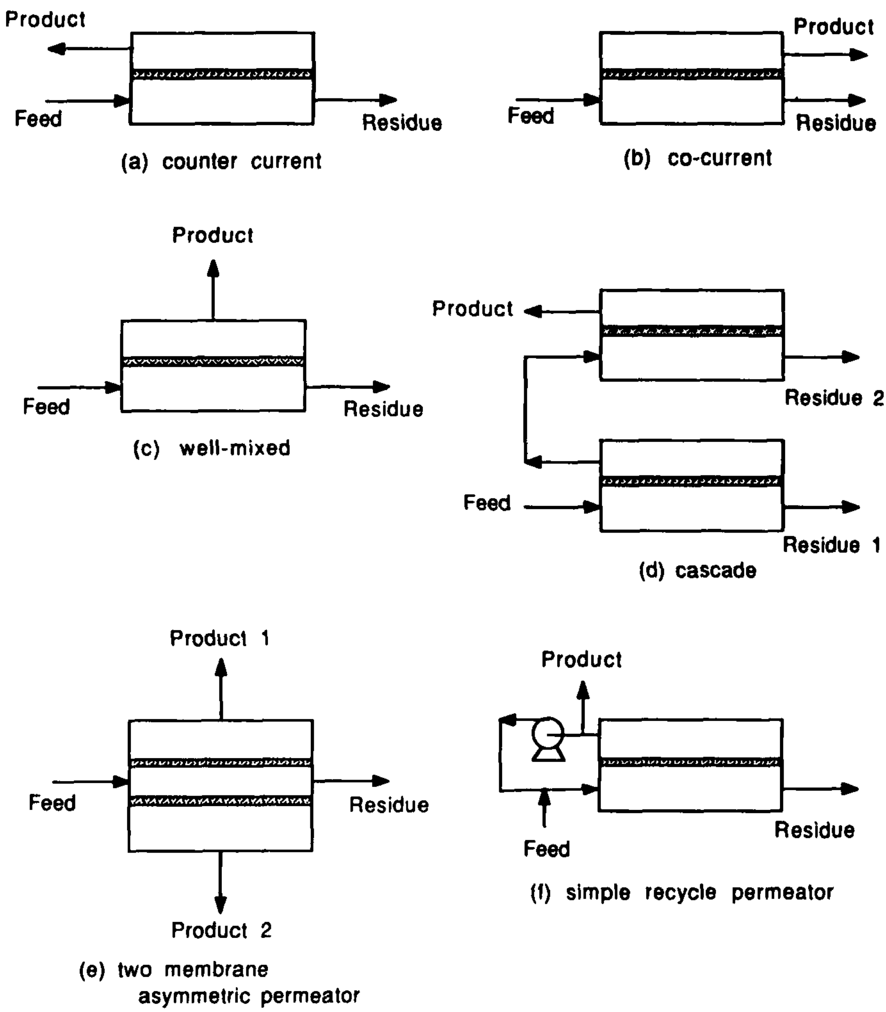

FIG. 1. Membrane permeator configurations.

pressor load or recycle ratio, and location of feed introduction. The unit compressor load (or recycle ratio), defined as the ratio of the compressor flow rate to the overhead product flow rate, increases as the product composition increases.

There has been much discussion regarding the optimum recycle permeator configuration for a given separation. Numerous investigators have studied the performance characteristics of the most common recycle permeator configurations: the simple recycle permeator, the complete continuous membrane column (CMC), the CMC enricher, and others (*11–15*). As pointed out by Stern et al. (*13*), most of the single membrane recycle permeators can be characterized largely by the location of the feed stream as shown in Fig. 2. Although the CMC is able to achieve very high product compositions and high product recovery, it has been criticized for its high unit compressor load. Virtually all comparisons among recycle membrane permeators have been based solely on their ability to separate oxygen from room air (21% O_2–79% N_2). Few studies have evaluated permeator performance for other gas mixtures.

This study investigates the effect of feed location on the unit com-

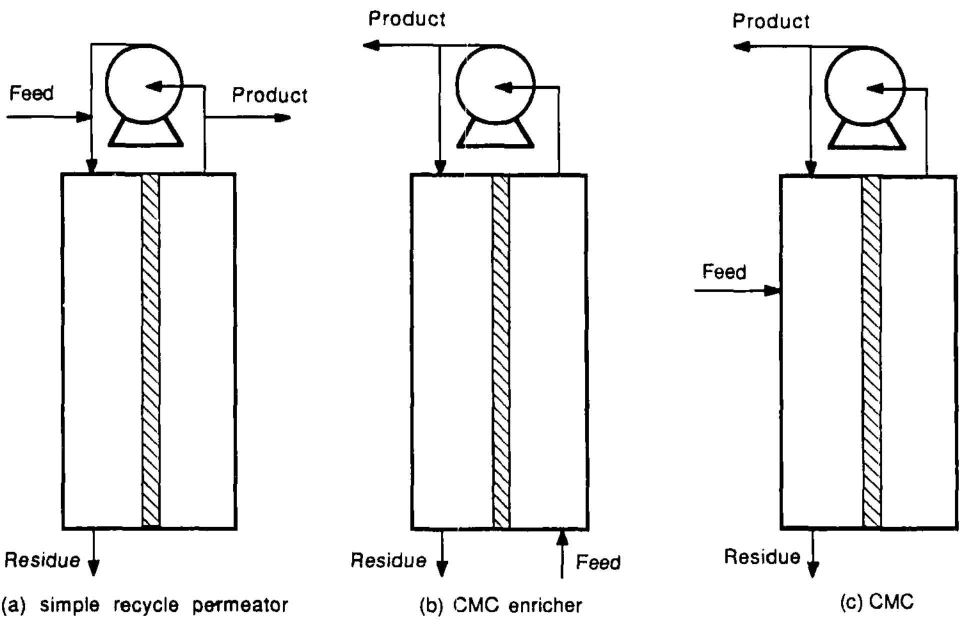

FIG. 2. Membrane permeators with recycle.

pressor load for single membrane permeators with recycle for CO_2–N_2, CO_2–O_2, and O_2–N_2 gas mixtures. Based on Stern's observation, knowledge of the feed location which minimizes the unit compressor load for these gas mixtures can be used to characterize the relative performance of single-stage membrane permeators with recycle. For example, a simple recycle permeator configuration is favored if the minimum unit compressor load occurs when feed is introduced at the top (i.e., near the compressor end) of the permeator. Similarly, the CMC configuration is favored if the minimum unit compressor load occurs when the feed is introduced at some point in the middle of the membrane permeator. Since the energy cost of compression is the principal operating cost of a membrane permeator, it is reasonable to optimize a membrane permeator with recycle on the basis of the minimum unit compressor load for a given membrane area. This study was limited to single-stage, single-membrane series-type permeators. No comparisons are intended with the multiple membrane asymmetric permeator (*16*) or parallel-type permeators (*17*).

MEMBRANE COLUMN SIMULATIONS

Earlier models of the total continuous membrane column neglected the effect of axial diffusion, and the models were not expected to predict behavior at low Peclet number (*10–12*). Low Peclet number situations can be expected at low flow rates and near the closed end of a membrane permeator. Several investigators have included an axial diffusion term, resulting in the introduction of a second-order differential equation (*19–21*). In terms of the low pressure side variables, the species material balance equation with axial diffusion for a binary gas system is given by

$$\frac{d^2 y_A}{dz^2} - \mathrm{Pe}\,\frac{dy_A}{dz} = f(y_A) \tag{3}$$

where

$$f(y_A) = \left\{\frac{C_3}{D_{AB}}\right\}\{K_A(y_A - 1)(y_A P^* - x_A) + K_B(1 - y_A)[(1 - y_A)P^* - (1 - x_A)]\} \tag{4}$$

$$C_3 = \left[\frac{2NL^2RT}{\ln\left(\frac{r_1}{r_0}\right) P^* r_{eq}^2} \right] \tag{5}$$

The Peclet number and the dimensionless pressure are given by

$$\mathrm{Pe} = \frac{GRTL}{PP^* \pi r_{eq}^2 D_{AB}} \tag{6}$$

$$P^* = P/P_0 \tag{7}$$

The low pressure side flow rate as a function of axial position is obtained from

$$\frac{dG}{dz} = \left\{ \frac{2\pi N}{\ln\left(\frac{r_1}{r_0}\right)} \right\} \{K_A(y_A P_0 - x_A P) + K_B[(1 - y_A)P_0 - (1 - x_A)]\} \tag{8}$$

The low pressure side pressure is assumed constant at P_0, and the Hagen-Poiseuille equation has been shown to be a good approximation of the pressure drop on the high pressure side of the membrane

$$\frac{dP}{dz} = \left\{ \frac{8\mu RTq}{\pi NPr_1^4} \right\} \tag{9}$$

The high pressure side flow rate q and composition x are related to G and y by material balance equations taken around the top portion of the membrane permeator,

$$q = q_F + G - q_P \tag{10}$$

$$x = \left(\frac{q_F x_F + Gy - q_P x_P}{q_F + G - q_P} \right) \tag{11}$$

Application of these equations to the continuous membrane column enricher and the simple recycle permeator requires only that the appropriate feed location and the boundary conditions be properly specified when using Eqs. (3) and (8).

The model equations for the membrane permeator were solved numerically using the method of orthogonal collocation on finite elements. This method has been successful in solving convective-diffusion type equations (*22–24*) and in modeling membrane permeators (*18–21*). Details of the orthogonal collection method are discussed in several texts (*25–27*). Numerical simulations were performed based on binary gas mixtures of carbon dioxide, nitrogen, and oxygen.

The model was verified using experimental data collected by Thorman for silicone rubber capillary membranes (*10*). The membrane permeators were assumed to be in a shell-and-tube configuration composed of 35 silicone rubber capillary membranes. The inner and outer radius of each capillary membrane were 1.19×10^{-4} m and 3.04×10^{-4} m, respectively. The inner radius of the shell was 3.175×10^{-3} m. The equivalent radius for flow, r_{eq}, was 2.62×10^{-3} m. The membrane permeabilities for silicone rubber were obtained as a function of temperature and pressure from Thorman (*10*). The mass diffusivity were estimated from the Wilke-Lee equation (*28*) using the collision integral as estimated by the method of Neufeld (*29*) and Hattikuder and Thodos (*30*). Wilke's approximation (*28*) was used to estimate gas mixture viscosities based on the pure component properties. The simulation conditions for the three systems are given in Table 1. In all cases, at least one product composition was chosen to exceed the enrichment limit attainable in a conventional single-stage permeator without recycle. For a 21% O_2–N_2 feed mixture with a separation factor of 2.1, the maximum oxygen enrichment is 36%. For a 57% CO_2–O_2 feed mixture with a separation factor of 5, the maximum CO_2 product composition is 87%. For a 53% CO_2–N_2 mixture with a separation factor of 11, the maximum CO_2 product concentration is 93%.

TABLE 1
Data Used in Membrane Column Simulations

Gas system	O_2–N_2	CO_2–N_2	CO_2–O_2
Feed composition	0.209	0.526	0.570
Product composition	0.417	0.946	0.883
Residue composition	0.196	0.082	0.226
Feed flow rate (μmol/s)	13.853	4.96	14.56
Product flow rate (μmol/s)	0.933	2.37	7.46
Column length (m)	4.24	5.12	5.12
Compressor pressure (kPa)	227.34	223.86	223.89
Ambient pressure (kPa)	98.59	98.78	99.27
Column temperature (K)	297.1	299.0	302.7

RESULTS AND DISCUSSION

The membrane permeator model with axial diffusion was solved numerically to obtain the compressor load as a function of feed location. The optimization results for the enrichment of a 21% O_2-N_2 mixture with a feed flow rate of 13.852 μmol/s and an overhead product flow rate of 0.933 μmol/s are shown in Fig. 3. Overhead product compositions simulated were 41.7, 65, and 90%. These compositions significantly exceed the maximum enrichment possible in a conventional stripper. As product composition increases, the difficulty of the separation increases. As a result, the required compressor load increases, although the increase in compressor load is not directly proportional to the increase in product composition. A 56% increase in product composition from 41.7 to 65% O_2 requires less than a 10% increase in compressor load. However, another 38% increase to 90% O_2 requires a 44% increase in compressor load. Each additional increase in the required product purity will lead to disproportionately greater and greater increases in compressor load. In general, the compressor load increases as the O_2-N_2 feed stream is introduced closer to the bottom of the column (i.e., $Z = 1$). A small local minimum is seen in the bottom curve at a dimensionless axial distance of 0.75; however, the minimum compressor load is obtained when the feed is introduced at the top of the permeator ($Z = 0$). Thus, a simple recycle permeator is indicated for this separation.

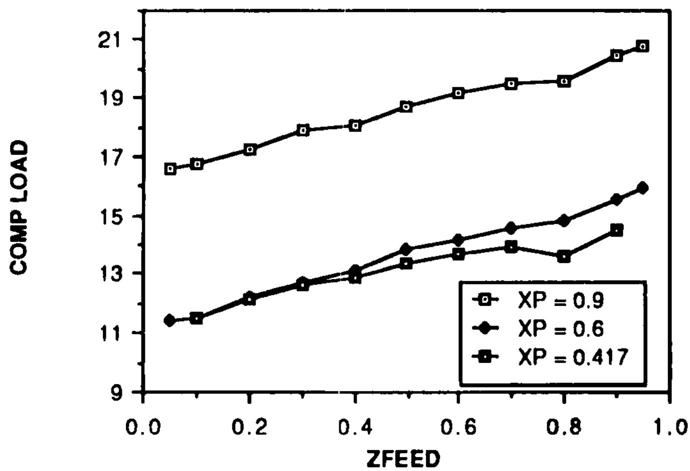

FIG. 3. Unit compressor load as a function of feed location for an O_2-N_2 system.

The simulation results for a 52.6% CO_2–N_2 mixture with a feed flow rate of 4.96 μmol/s and a product flow rate of 2.37 μmol/s are shown in Fig. 4. Product compositions investigated were 60, 80, and 95% CO_2. The shape of these curves is different from that seen in the O_2–N_2 system in Fig. 3. Most obvious is the minimum compressor load obtained when the feed is introduced from $Z = 0.65$ to 0.75 rather than at the top of the column. The compressor load remains relatively constant with respect to feed location over the top half of the permeator. As the product composition increases, the location of the minimum shifts higher in the column and the width of the minimum becomes narrower. The depth of the minimum also decreases. For this separation, a CMC would perform better than the simple recycle permeator.

Figure 5 shows the results for a 57% CO_2–O_2 feed mixture at a flow rate of 14.56 μmol/s and a product flow rate of 7.46 μmol/s. A broad minimum is seen when the overhead product is 65% CO_2, while no minimum is seen at all when the product composition is increased to 95% CO_2. In the region below the feed location of the minimum unit compressor load, the slope once again becomes rather steep. The curve representing a product composition of 83.3% CO_2 shows a smaller, narrower minimum at 0.7. Unlike the bottom curve, the compressor load steadily increases between 0.1 and 0.55. Its behavior over this region parallels that of the top curve. As the separation difficulty increases, the minimum becomes smaller and

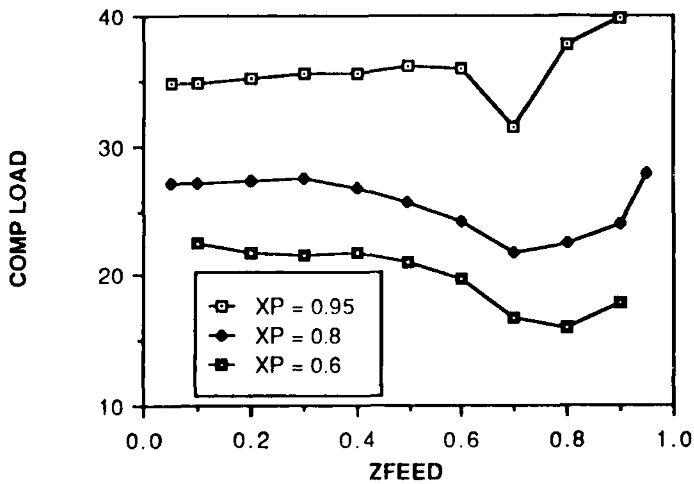

FIG. 4. Unit compressor load as a function of feed location for a CO_2–N_2 system.

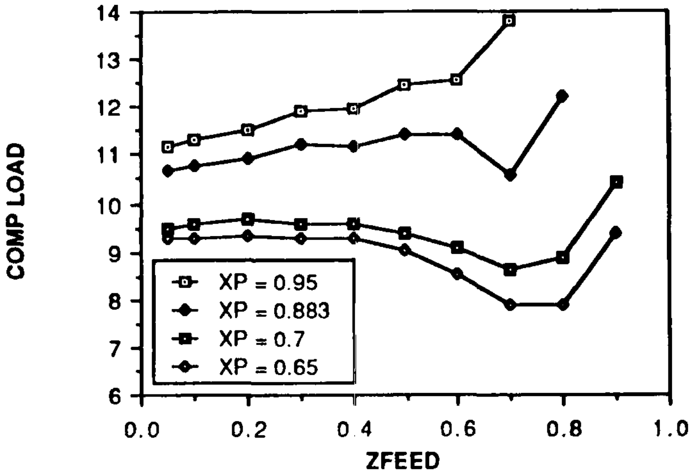

FIG. 5. Unit compressor load as a function of feed location for a CO_2-O_2 system.

smaller. Simultaneous with this, the slope of the curve becomes steeper until the minimum vanishes and a monotonically increasing function is reached at high product compositions.

The results of the optimization shown in Figs. 3–5 can be generalized roughly into easy separations, medium separations, and difficult separations. The difficulty of a separation is characterized by the separation factor and the product composition desired. Difficult separations are typically those involving systems with low separation factor and high product composition requirements. The O_2-N_2 separation with $\alpha = 2.1$ and a product composition exceeding 30–35% O_2 can be considered a difficult separation. The CO_2-N_2 separation with $\alpha = 11$ and a product composition below 90% is a relatively easy separation. The CO_2-O_2 system has a separation factor of 5 which is moderate. If high product compositions are required, those exceeding 85% would make the separation approach that of a difficult separation. Lower compositions would be of moderate separation difficulty.

Those separations we have termed difficult show an optimum feed location very near the top of the column. The slope of the curve shows a substantial positive slope, indicating that feed locations farther down the column will result in a large increase in unit compressor load. The O_2-N_2 curves and the top CO_2-O_2 curve fall into this category. For difficult separations of this type it is necessary to expose the feed mixture to as much membrane area as possible. As the feed location is moved farther and farther down the column, there is less and less membrane area through which to separate the gas mixture. If the feed is introduced too far down the membrane column, there is inadequate membrane area left so that a great increase in compressor load is required to compensate for the loss in separating ability. This is evidenced by the large increase in slope typical of these curves near the bottom of the column.

On the other hand, easier separations such as the CO_2–N_2 separation and the lower CO_2–O_2 separation show a distinct optimum in the region of 0.6 to 0.75. The easier separations show broad minima and indicate substantially reduced operating cost if the total membrane column is operated at its optimum feed location. In general, the feed location does not affect the unit compressor load very much if the feed location is above the optimum location. However, if the feed location is chosen below the optimum value, a large unfavorable effect on unit compressor load can be anticipated.

Finally, moderately difficult separations fall in between these two extremes. They have small, narrow local minima and represent regions where the optimum feed location may be at two widely different points depending on the depth of the minima. The CO_2–O_2 separation with 88.3% CO_2 is typical of this situation. A local minima occurs at the top of the column as well as at 0.7. Their magnitudes are approximately the same. In this situation, the optimum permeator configuration depends upon the range of product concentrations likely to be desired from the membrane permeator.

Figure 5 also shows how the membrane column responds to increases in product composition and underscores how the behavior of the membrane column can move from an "easy" region to a "hard" region for the same gas mixture. The general membrane column response is to reduce the width and depth of the minima at increasing product composition. At the same time, the slope of the unit compressor load curve becomes increasingly more positive, which serves to accelerate the reduction of the minima. With increasing product composition, a point is reached where the minima vanishes and a rapid increase in unit compressor load takes place. This same phenomenon is exhibited less dramatically in Figs. 3 and 4 for CO_2–N_2 and O_2–N_2, respectively.

In Figure 6 the minimum unit compressor load is shown as a function of product composition and separation factor. As product composition increases, minimum unit compressor load increases for all separation factors, as would be expected. The greatest increase in the minimum unit compressor load occurs at a low separation factor where increased recycle is necessary for enrichment; however, it also occurs at high

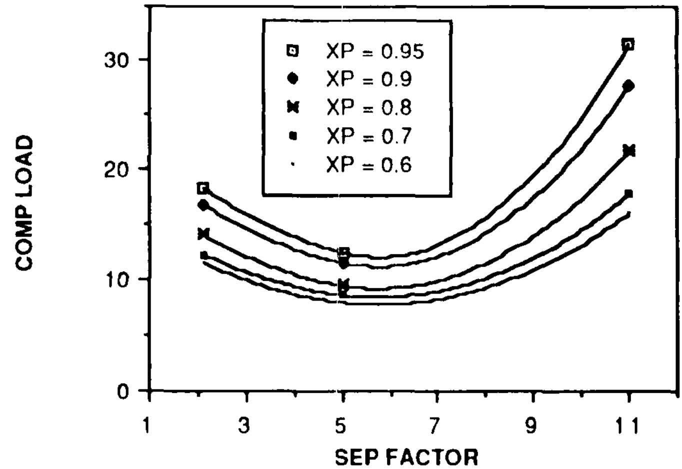

FIG. 6. Minimum compressor load as a function of separation factor.

separation factor. As the separation factor increases, the need for recycle decreases. The higher membrane throughput increases the compressor flow rate but does not significantly increase the enrichment potential of the permeator. As a result, the normalized compressor load increases rather than decreases for a high separation factor. For most high separation factor cases, the use of a conventional membrane permeator without recycle is favored.

CONCLUSIONS

This study shows that the position of the optimum feed location depends on the relative difficulty of separation. Difficulty of separation is a function of separation factor, product composition, and cut. Systems with a low separation factor have an optimum feed location near the top of the high pressure side of the membrane column. This feed location corresponds to the simple recycle permeator. For situations of low to moderate separation difficulty, a distinct minimum was found within the region of 0.6 to 0.75 for the separations simulated. This feed location indicates that the total continuous membrane column is a good method of operation for this case. For some gas mixtures, separation difficulty may change significantly as product composition increases, as in the case of the CO_2-O_2 system. For such cases a continuous membrane column may be used for low to moderate product compositions, but a recycle permeator may be optimal for higher product compositions. Finally, for many applications the cost of recycle is not justified and the use of a conventional membrane permeator without recycle is the configuration of choice.

NOMENCLATURE

D_{AB}	diffusivity of AB gas mixture
G	low pressure side flow rate
K_i	permeability of species i
L	length of permeator
N	number of tubes
P	high pressure side pressure
P_0	low pressure side pressure
P^*	dimensionless pressure
Pe	Peclet number
q	high pressure side flow rate
R	gas constant
r_{eq}	equivalent radius for flow around tubes
r_i	inner radius of tube
r_o	outer radius of tube
T	temperature
t	time
X	quantity in Eqs. (1) and (2)
x_i	high pressure side composition of species i
Y	quantity in Eq. (1)
y_i	low pressure side composition of species i
z	axial distance
z_F	feed location

Greek

α	separation factor
μ	viscosity
π	3.14159 . . .

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