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Analysis of Internally Multistaged Gas Permeator

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Abstract

Mathematical models have been developed for an internally multistaged gas permeator based on the perfect mixing case. A problem of oxygen enrichment from air has been selected to illustrate the performance of the permeator. It has been shown that the effect of the number of permeation stages on oxygen enrichment is significant when the number of permeation stages is increased from one to two or three, and its effect reduces with a further increase in stage number. The results further reveal that no matter how many permeation stages are employed, the higher degree of separation is only achievable at lower values of overall stage cuts compared to the conventional single-stage permeator.

INTRODUCTION

Gas separation by selective permeation through a nonporous membrane is an important unit operation which is now being widely applied in industry. Although a number of satisfactory membrane materials and separation schemes are presently available in practice, continuing efforts are being made toward the development of better membrane materials and more efficient separation schemes. One such scheme, shown in Fig. 1, is an internally staged permeation which makes use of the pressure difference between the feed and permeate side into two steps with two membranes of the same type. It can be seen from the figure that the permeate, enriched in a more permeable component, emerges from the first membrane at an intermediate pressure and is then applied as feed to the second membrane for the second permeation steps. This separation scheme was first studied by Sidhoum et al. (*1*) for a binary gas mixture in a separation device with two sets of hollow fiber. These authors demonstrated that the superior enriching performance is achievable at very low values of overall stage cuts compared to the conventional single-stage permeation schemes. While the work carried out by them was restricted to a fixed membrane area ratio

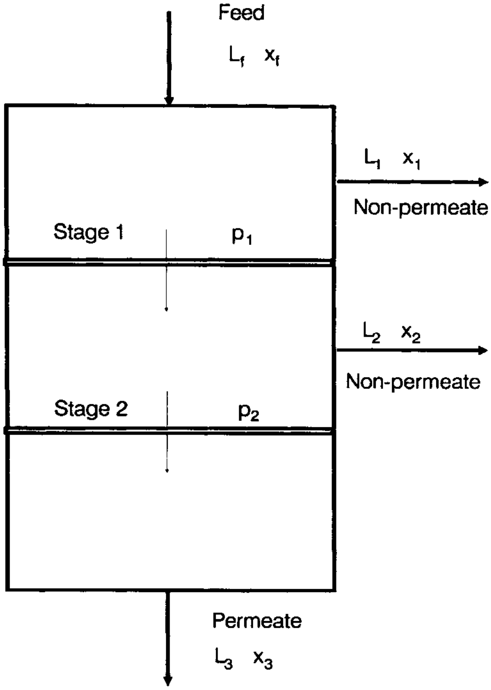

FIG. 1. Internally two-staged permeator.

between the stages, it was later analyzed further by Li et al. (*2*) who showed that the best or maximum degree of separation in such a permeator could be achieved if the membrane area ratio between the stages and the pressure ratio across each membrane are properly selected.

In the present investigation, the concept of the above-mentioned separation scheme has been extended to the multipermeation steps shown in Fig. 2 where the multilayer of membranes of the same type were housed in one device. The mathematical models developed for a binary gas mixture were based on the perfect mixing assumption. The example of oxygen enrichment from air has been chosen to illustrate the performance of such a separation scheme.

DEVELOPMENT OF MODELS

The models developed for the multipermeation stages are based on following assumptions:

1. All components in the feed stream are permeable.
2. The permeability of each gas component is the same as that of pure gas and is independent of pressure.

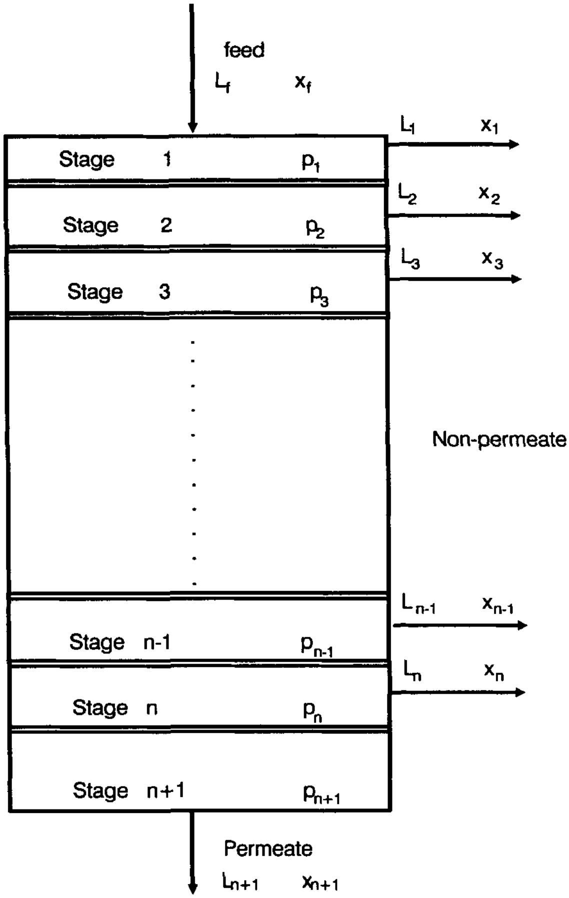

FIG. 2. Internally multistaged permeator.

3. A negligible gas-phase concentration gradient exists in the permeation direction, i.e., no concentration polarization.
4. Negligible pressure drop of the feed and permeate stream occurs along the flow path.
5. Perfect mixing condition occurs in each permeation stage.
6. Permeation obeys Fick's law.

With the above assumptions for a binary gas mixture such as O_2/N_2, the permeation equations will be given by

$$O_2: \quad L_f x_f - \sum_{i=1}^{n} L_i x_i = \frac{P_A A_i}{\delta}(p_i x_i - p_{i+1} x_{i+1}) \tag{1}$$

$$\text{N}_2: \quad L_f(1 - x_f) - \sum_{i=1}^{n} L_i(1 - x_i) = \frac{P_B A_i}{\delta}\,(p_i\,(1 - x_i) - p_{i+1}\,(1 - x_{i+1})) \tag{2}$$

and $i = 1, 2, 3, \ldots, n$, where i represents the number of permeation stages in the permeater.

In addition to the permeation equations, the material balances over the entire permeater are:

$$\text{Overall:} \quad L_f = \sum_{i=1}^{n} L_i + L_{i+1} \tag{3}$$

$$\text{O}_2: \quad L_f x_f = \sum_{i=1}^{n} L_i x_i + L_{i+1} x_{i+1} \tag{4}$$

$$\text{N}_2: \quad L_f(1 - x_f) = \sum_{i=1}^{n} L_i\,(1 - x_i) + L_{i+1}\,(1 - x_{i+1}) \tag{5}$$

and $i = 1, 2, 3, \ldots, n$.

The stage cut, i.e., fraction of feed allowed to permeate, defined for each permeation stage is

$$\phi_i = \frac{L_f - \sum_{i=1}^{n} L_i}{L_f - \sum_{i=1}^{n} L_{i-1}}, \qquad L_{i-1} = 0 \text{ when } i = 1 \tag{6}$$

and $i = 1, 2, 3, \ldots, n$.

According to Eq. (6), the overall stage cut is, therefore, obtained as

$$\Phi = \phi_1\phi_2\phi_3, \ldots, \phi_n = \frac{L_f - \sum_{i=1}^{n} L_i}{L_f} = \frac{L_{i+1}}{L_f} \tag{7}$$

and $i = 1, 2, 3, \ldots, n$

Rearranging the above equations leads to the following expressions:

$$\frac{L_f x_f - \sum_{i=1}^{n} L_i x_i}{L_f(1 - x_f) - \sum_{i=1}^{n} L_i (1 - x_i)} = \frac{P_A}{P_B} \frac{P_i x_i - p_{i+1} x_{i+1}}{p_i(1 - x_i) - p_{i+1}(1 - x_i)} \tag{8}$$

$$\frac{x_f}{1 - x_f} = \frac{\sum_{i=1}^{n} L_i x_i + L_{i+1} x_{i+1}}{\sum_{i=1}^{n} L_i(1 - x_i) + L_{i+1}(1 - x_{i+1})} \tag{9}$$

$$L_i = \left(L_f - \sum_{i=1}^{n} L_{i-1}\right)(1 - \phi_i), \qquad L_{i-1} = 0 \text{ when } i = 1 \tag{10}$$

and $i = 1, 2, 3, \ldots, n$

Li et al. demonstrated in their earlier study (*2*) that for an internally two-staged permeator, in order to maximize the degree of separation, the values of each stage cut and the pressure ratio across each membrane should be kept the same. Therefore, in this study, both the values of each stage cut and the pressure ratio across each membrane have been kept the same as shown in the following expressions:

$$\phi_1 = \phi_2 = \phi_3 = \ldots = \phi_n \tag{11}$$

$$\frac{p_1}{p_2} = \frac{p_2}{p_3} = \frac{p_3}{p_4} = \ldots = \frac{p_n}{p_{n+1}} \tag{12}$$

In order to determine the mole fraction of oxygen in the permeate stream and membrane area requirements, the following computational procedures were employed:

1. Specify the values of p_1 and p_{n+1}.
2. Assume a value of p_2 and calculate $p_3, p_4, \ldots, p_n$ and p_{n+1} from Eq. (12).

If the calculated value of p_{n+1} is different from that specified, assume a new value of p_2 and continue the iteration until the calculated value of p_{n+1} from Eq. (12) converges with the value of p_{n+1} specified.

3. Specify the values of x_f, L_f, P_A, P_B, δ, and ϕ_i.
4. Calculate L_i from Eq. (10).
5. Assume a value of x_1 and calculate the values of $x_2, x_3, \ldots, x_n$ and x_{n+1} from Eq. (8).
6. Use the calculated values of $x_2, x_3, \ldots, x_n$ and x_{n+1} from Eq. (8) to calculate the value of x_1 by using Eq. (9).

If the calculated value of x_1 from Eq. (9) is different from that assumed, Steps 5 and 6 are repeated until the calculated value of x_1 converges with the value assumed.

7. The membrane area requirement for each stage in the permeator is determined from Eq. (1).

Clearly, the above expressions reduce to the single-stage permeation case when $i = 1$.

RESULTS AND DISCUSSION

In this study, analysis of an internally multistaged membrane permeator was made for the case of oxygen enrichment using air as feed. The published values of permeabilities for an ethyl cellulose membrane (*3*) were used in the simulation. The operating conditions for the permeator and permeability values used for all the components involved are listed in Table 1. The feed pressure, p_1, was varied from 2600 to 7800 kPa, while the permeate pressure, p_{n+1}, was kept at 100 kPa in all cases.

In the parametric study, the oxygen enrichment, i.e., mole fraction of oxygen in the permeate stream as a function of permeation stage and overall stage cut Φ, is calculated using the perfect mixing model described above. The results of the study are presented in Figs. 3 and 4. In Fig. 3 the overall stage cut is plotted against oxygen mole fraction in the permeate stream for different numbers of permeation stages. It can be seen from the figure that as the number of permeation stages in the permeator is increased, oxygen enrichment is improved, especially at lower values of the overall stage cuts. It is, however, interesting to note that the extent of oxygen enrichment is not directly proportional to the number of permeation stages employed. As can be seen in the figure, the effect of permeation stages on oxygen enrichment is significant when it was increased from one permeation stage to two or three, but thereafter the enhancement effect gradually decreases. For instance, at an overall stage cut of 0.01 and one permeation stage in the permeator, the oxygen enrichment achieved was about 46% compared to 64, 69, 72, and 73% for permeation stages of 2, 3, 4, and 5, respectively, at the same overall stage cut value.

TABLE 1
Operating Conditions Used in Simulation

Feed rate, kmol/s	2.05×10^{-5}
Feed composition, mol fraction:	
O_2	21
N_2	79
Membrane thickness, m	3.81×10^{-5}
Permeability $\times 10^{15}$, $kmol \cdot m \cdot m^{-2} \cdot s^{-1} \cdot kPa^{-1}$:	
O_2	2.94
N_2	0.86

In Fig. 4 the effect of the number of permeation stages in the permeator on oxygen enrichment is shown for different overall stage cuts. It can be seen from the figure that at an overall stage cut of 0.01, the performance of oxygen enrichment is greatly enhanced upon introduction of multipermeation stages in the permeator. However, when the overall stage cut is increased, the advantage of employing multipermeation stages in the permeator is diminished. It can be seen from the figure that at an overall stage

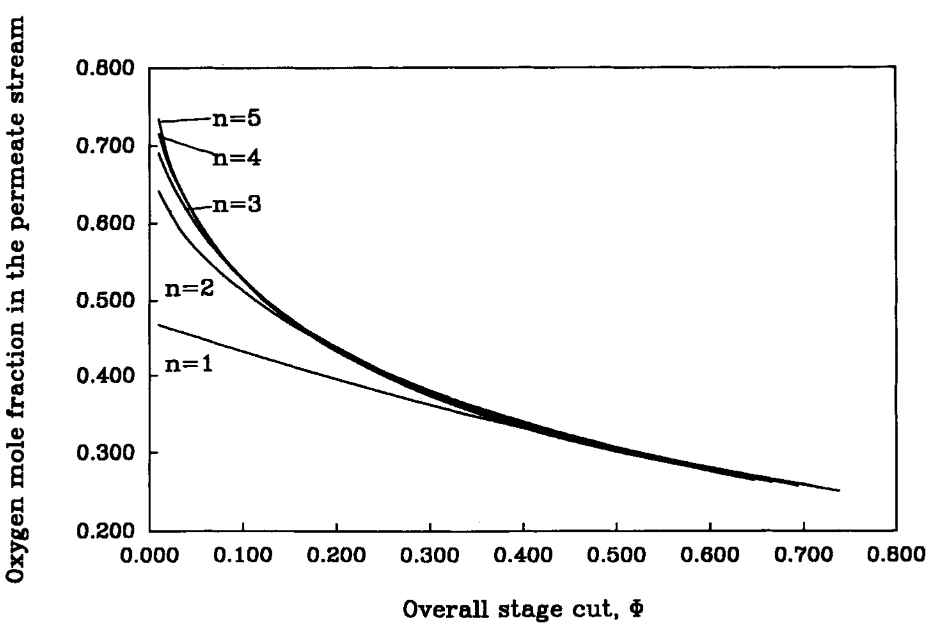

FIG. 3. Effect of overall stage cuts on oxygen enrichment for five different permeation stages (p_1 = 7800 kPa, p_{n+1} = 100 kPa).

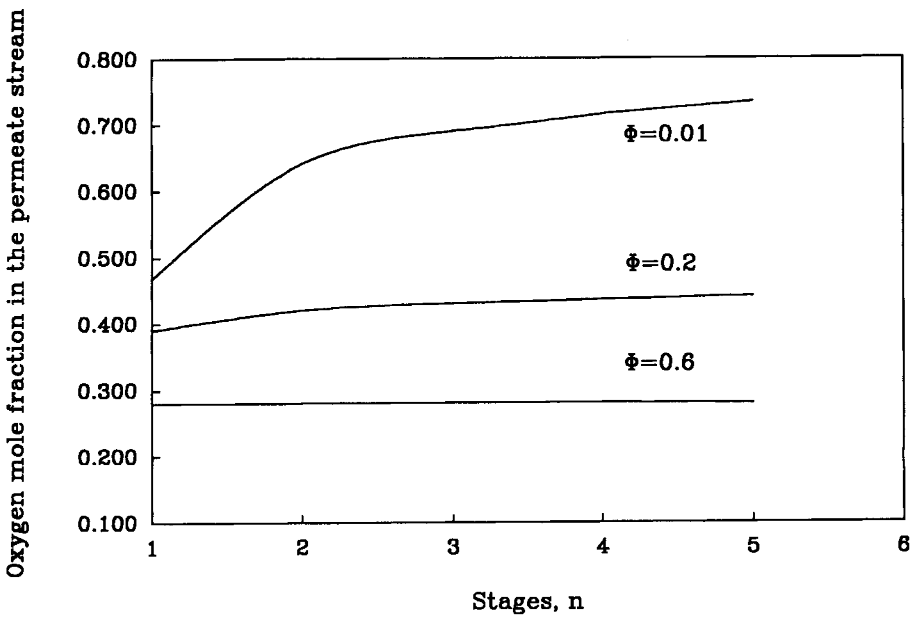

FIG. 4. Effect of permeation stages on oxygen enrichment (p_1 = 7800 kPa, p_{n+1} = 100 kPa).

cut of 0.2, the improvement of oxygen enrichment is gradually reduced, and at an overall stage cut of 0.6 an increase in permeation stages has no effect on oxygen enrichment. This clearly indicates that the introduction of multipermeation stages in a permeator will offer better performance in terms of oxygen enrichment at lower values of overall stage cuts. After a certain value of an overall stage cut (Φ = 0.6 in this study), the multi-permeation permeator does not give better performance compared to the conventional one-stage permeator. This behavior can also be seen in Fig. 3: at an overall stage cut of 0.6, the oxygen enrichment is about 0.3, regardless of the number of permeation stages employed.

The effect of the number of permeation stages on oxygen enrichment at different feed pressures is shown in Fig. 5. It can be seen from the figure that as the feed pressure is increased, better oxygen enrichment is achieved, assuming all other parameters remain unchanged. It is interesting to note that the degree of separation is almost proportional to the feed pressure after introduction of multipermeation stages. This behavior is certainly different from that of a one-stage permeator where it was demonstrated (*4*) that the effect of feed pressure is not directly proportional to the degree of separation and that an increase in feed pressure only results in a slightly better degree of separation, particularly in the high feed pressure region.

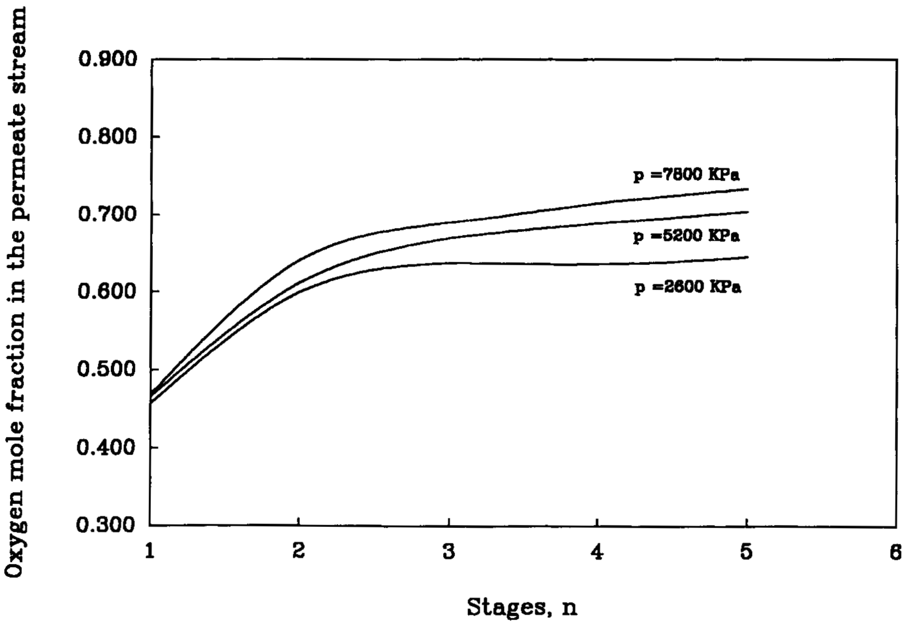

FIG. 5. Effect of permeation stages on oxygen enrichment for three different feed pressures (p_{n+1} = 100 kPa, Φ = 0.01).

Comparison of membrane area requirements for different values of overall stage cuts and different permeation stages as generated by three different feed pressures are illustrated in Figs. 6 and 7. It could be expected that the membrane area requirements increase with increasing permeation stages in the permeator as shown in Fig. 6. The required membrane area, however, could be reduced by applying high pressure on the feed side. It can be seen from Fig. 7 that considerable reduction in the membrane area requirement is achieved when the higher pressure is introduced on the feed side; for example, from 640.5 m^2 at a feed pressure of 2600 kPa to 240.5 m^2 at a feed pressure of 7800 kPa in the case of five permeation stages—a reduction of about 60%.

In Fig. 8, the oxygen recovery, defined as $R = L_{n+1}x_{n+1}/L_f x_f$, is plotted against the membrane area requirement for a different number of permeation stages in the permeator. The overall stage cut and the feed pressure are kept constant at 0.01 and 7800 kPa, respectively. It can be seen from the figure that with the same number of permeation stages, the oxygen recovery remains unchanged as the membrane area is increased. Introduction of multipermeation stages in a permeator will greatly improve the

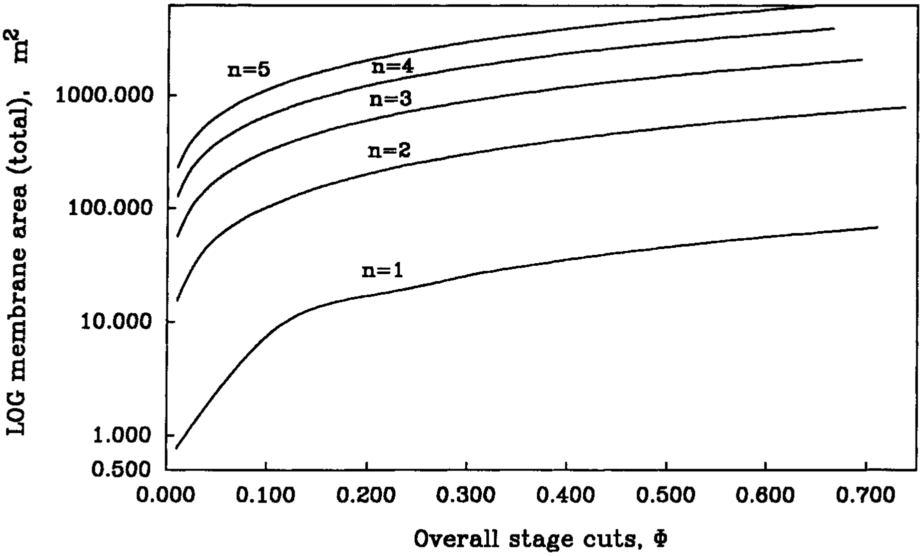

FIG. 6. Effect of permeation stages on total membrane area requirements (p_1 = 7800 kPa, p_{n+1} = 100 kPa).

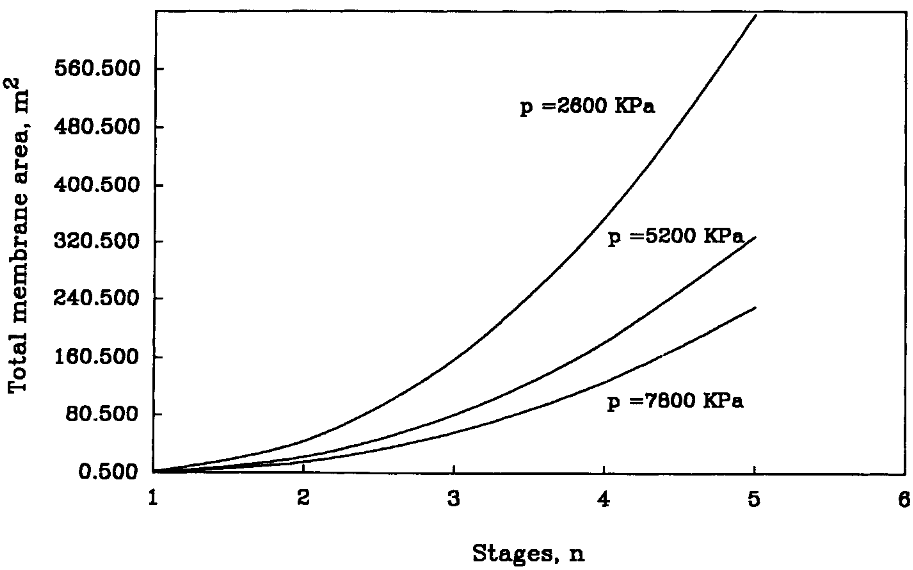

FIG. 7. Effect of feed pressure on total membrane area requirements (p_{n+1} = 100 kPa, Φ = 0.01).

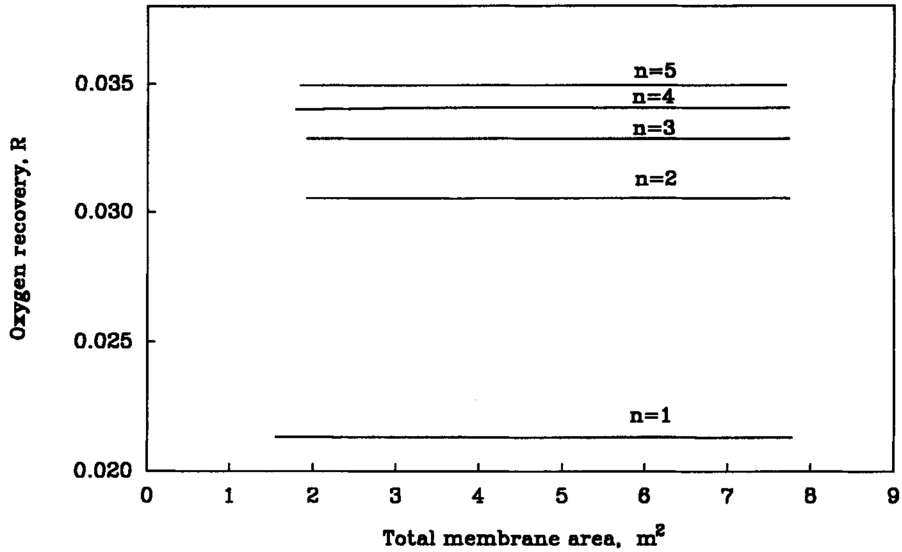

FIG. 8. Oxygen productivity versus membrane area for five different permeation stages (p_1 = 7800 kPa, p_{n+1} = 100 kPa, Φ = 0.01).

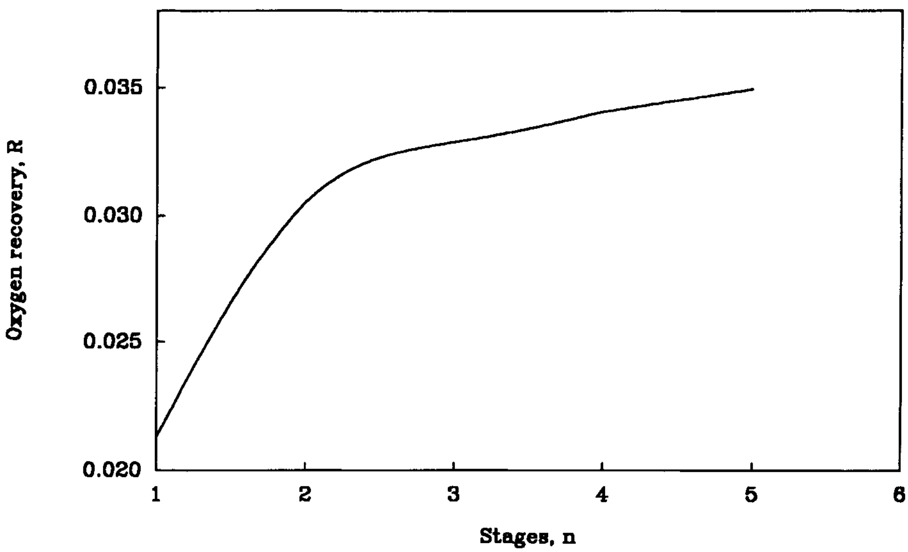

FIG. 9. Oxygen productivity versus permeation stages (p_1 = 7800 kPa, p_{n+1} = 100 kPa, Φ = 0.01).

oxygen recovery, as shown in Fig. 9 where the effect of permeation stages on oxygen recovery is illustrated. It appears that effective utilization of a variable pressure difference between the feed and permeate side can achieve a much better degree of separation which may not be possible to obtain in a conventional permeation scheme.

CONCLUSION

The performance of an internally multistaged permeator for gas separations has been analyzed by using mathematical models based on the perfect mixing case. It has been shown that, in general, a higher degree of separation can be achieved by employing an internally multistaged permeator. A comparison among the permeation stages considered in this study indicated that the effect of the permeation stages on oxygen enrichment is significant when the permeation stages are increased from one to two or three, and its effect decreases gradually if the permeation stages are further increased. A parametric study further revealed that regardless of the number of permeation stages employed, a better degree of separation is only achievable at lower values of overall stage cuts compared to a conventional single-stage permeator.

NOTATION

A membrane area (m^2)
L flow rate (kmol/s)
L_{n+1} permeate flow rate (kmol/s)
p pressure (kPa)
P permeability coefficient ($kmol \cdot m \cdot m^{-2} \cdot s^{-1} \cdot kPa^{-1}$)
R O_2 recovery
x mole fraction of oxygen
x_{n+1} mole fraction of oxygen in the permeate stream

Greek Letters

ϕ individual stage cut
Φ overall stage cut
δ membrane thickness (m)

Subscripts

A oxygen
B nitrogen
f feed
i number of permeation stages

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