SIMULATION OF AN INTERNALLY MULTI-STAGED PERMEATION MODULE OPERATING WITH COCURRENT FLOW PATTERN

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Introduction

Membrane separation plays an important role in many modern industrial processes and often offers significant advantages over the conventional separation processes. Membranes used in gas separations, however, suffer either from low selectivities or low permeabilities. Therefore, considerable attention has been paid to the development of more efficient separation schemes using currently available membrane materials to achieve the desired degree of separation. Numerous separation schemes and detailed modeling studies have been reported in the literature by Pan and Habgood for permeate purging, Ohno et al. and Perrin and Stern for asymmetric permeators, Hwang and Thorman for membrane columns, Sidhoum et al. for internally staged permeators and Li et al. for internally multistaged permeators. Among these innovative separation schemes, the internally staged permeation systems have attracted considerable interest. Figure 1 shows an internally multistaged permeation module which splits the overall driving force, i.e. pressure difference between feed and permeate side, into several parts as follows: \( p_1 > p_2 > p_3 \ldots > p_{n-1} > p_n > p_{n+1} \). Thus, the internally multistaged permeator can be operated without interstage recompression. The whole purpose of using such an arrangement is to optimize the use of pressure energy for the enrichment of desired product(s).

The internally staged permeator can be viewed as a conventional simple cascade with the exception that each individual permeator is coupled locally; therefore, the permeate stream from the preceding stage can be locally fed into the next stage. By contrast, in a conventional simple cascade process the permeators are connected in series and the permeate streams are collected externally and then fed into the next stage as feed. If compositions of the feed and permeate flow are changing along the flow path (usually the case in a hollow-fibre permeator), the better separation achieved in the internally staged permeator becomes obvious. Sidhoum et al. conducted a comparative study of these two separation schemes and indicated that the internally staged permeator is the better one, especially when the ideal separation factor of the gases involved is low.

An analysis of an internally multistaged permeator based on cocurrent flow, a more practical flow pattern in membrane process design, is presented. The example of oxygen enrichment from air is chosen to illustrate the performance of the permeator, and the simulation results generated by the cocurrent flow model are compared to those based on the perfect mixing model.

Development of Model

The development of the cocurrent flow model for an internally multistaged permeation module is based on the 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.
3) Negligible gas-phase concentration gradient exists in the permeation direction, i.e. there is no concentration polarization.
4) Negligible pressure drop of the feed, nonpermeate, and permeate streams occurs along the flow path in each stage.

With the above-listed assumptions and the flow conditions illustrated in Fig. 1, for a binary gas mixture of A/
B, such as O₂/N₂, the permeation equations for the cocurrent flow pattern are given by:

\[
\frac{d(L'_ix'_i)}{dA'_i} = \frac{P_A}{\delta} \{p_{i-1}x'_{i-1}R_{i-1} + p_{i+1}x'_{i+1}R_i - p_{ix'_i}(R_{i-1} + R_i)\}, \quad (R_{i-1} = 0 \text{ when } i = 1)
\]

\[
\frac{d(L'_i(1-x'_i))}{dA'_i} = \frac{P_B}{\delta} \{p_{i-1}(1-x'_{i-1})R_{i-1} + p_{i+1}(1-x'_{i+1})R_i - p_{ix'_i}(R_{i-1} + R_i)\}, \quad (R_{i-1} = 0 \text{ when } i = 1)
\]

\[
R_i = \frac{dA'_i}{dA'_1}
\]

\[
i = 1, 2, 3, \ldots, n
\]

where \(i\) represents the number of permeation stages in the permeator.

Also, the permeation equations for the bottom compartment of the permeator are:

\[
\frac{d(L'_{n+1}x'_{n+1})}{dA'_{n+1}} = \frac{P_A}{\delta} (p_nx'_n - p_{n+1}x'_{n+1})R_n
\]

\[
\frac{d[L'_{n+1}(1-x'_{n+1})]}{dA'_{n+1}} = \frac{P_B}{\delta} \{p_n(1-x'_n) - p_{n+1}(1-x'_{n+1})R_n\}
\]

where

\[
R_n = \frac{dA'_n}{dA'_1}
\]

In addition to the permeation equations, material balances over the boundaries shown by the dashed loop in Fig. 1 are:

Overall:

\[L_f = \sum_{i=1}^{n} L'_i + L'_{n+1}\]  \(\text{(6)}\)

**A:** \[L_f = \sum_{i=1}^{n} L'_i x'_i + L'_{n+1} x'_{n+1}\]  \(\text{(7)}\)

**B:** \[L_f(1-x_f) = \sum_{i=1}^{n} L'_i (1-x'_i) + L'_{n+1} (1-x'_{n+1}) \]  \(\text{(8)}\)

and \(i = 1, 2, 3, \ldots, n\).

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

\[
\Phi_i = \frac{L_f - \sum_{i=1}^{n} L'_i}{L_f - \sum_{i=1}^{n} L'_{i-1}}, \quad L'_{i-1} = 0 \text{ when } i = 1
\]

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

\[
\Phi = \prod_{i=1}^{n} \Phi_i = \frac{L_f - \sum_{i=1}^{n} L'_i}{L_f} = \frac{L'_{n+1}}{L_f}
\]

\(i = 1, 2, 3, \ldots, n\)

Rearranging Eqs. (1) to (5) leads to the following expressions:

\[
\frac{dx'_i}{dA'_1} = \frac{1}{L'_i} \left( P_A C_i - x'_i dL'_i/dA'_1 \right)
\]

\[
\frac{dL'_i}{dA'_1} = \frac{1}{\delta} \left( P_A C_i + P_B C_2 \right)
\]

where

\[
C_1 = p_{i-1}x'_{i-1}(R_{i-1} + p_{i+1}x'_{i+1}) - p_{ix'_i}(R_{i-1} + R_i)
\]

\[
C_2 = p_{i-1}(1-x'_{i-1})R_{i-1} + p_{i+1}(1-x'_{i+1}) - p_{ix'_i}(R_{i-1} + R_i)
\]

\[
\frac{dx'_{n+1}}{dA'_{n+1}} = \frac{1}{L'_{n+1}} \left( \frac{P_A}{\delta} (p_nx'_n - p_{n+1}x'_{n+1})R_n - x'_{n+1} dL'_{n+1}/dA'_{n+1} \right)
\]

\[
\frac{dL'_{n+1}}{dA'_{n+1}} = \frac{R_n}{\delta} \left[ P_A (p_nx'_n - p_{n+1}x'_{n+1}) + P_B (p_n(1-x'_n) - p_{n+1}(1-x'_{n+1})) \right]
\]

\(i = 1, 2, 3, \ldots, n\)

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

Equations (11) to (16) can be simultaneously integrated using an appropriate numerical technique based on the following boundary conditions:

1. At the permeator inlet, i.e. \(A'_1 = 0\)

\[x'_1 = x_0 x'_i = x_i (i = 2, 3, \ldots, n+1),\]

\[A'_i = 0 (i = 2, 3, \ldots, n)\]

\[L'_1 = L_f L'_i = 0 (i = 2, 3, \ldots, n+1)\]

2. At the permeator outlet, i.e. \(A'_{n+1} = A_1\)

\[x'_i = x_0 (i = 1, 2, 3, \ldots, n+1)\]

\[A'_i = A_i (i = 2, 3, \ldots, n)\]

The initial permeate conditions of \(x_i (i = 2, 3, \ldots, n+1)\) in
Fig. 2  Effect of overall stage cut on oxygen enrichment for four different permeation stages: cocurrent flow and perfect mixing ($p_1 = 7800$ kPa, $p_{n+1} = 100$ kPa)

Table 1. Operating conditions used in simulation

| Feed Rate, ($\text{kmol s}^{-1}$) | $2.05 \times 10^{-5}$ |
| Feed composition, (mole fraction): | $\text{O}_2$: 0.21, $\text{N}_2$: 0.79 |
| membrane thickness, (m) | $3.81 \times 10^{-5}$ |
| Permeability × $10^{3}$, $(\text{kmol m}^{-2} \cdot \text{s}^{-1} \cdot \text{kPa}^{-1})$: | $\text{O}_2$: 2.94, $\text{N}_2$: 0.86 |

the permeator inlet are determined from the ratio of Eqs. (1) and (2), and Eqs. (4) and (5), respectively, which are evaluated at stage inlet (when $L'_i = 0$, $i = 2, 3...n + 1$). Eqs. (11) (when $i = 2, 3...n$) and (15) are indeterminate at the inlet conditions. This problem has been overcome by applying L'Hôpital's rule. The values of each stage cut and pressure ratio across each membrane have been kept the same as under such conditions that the permeator may maximize the degree of separation.

Results and Discussion

In this study, simulation of an internally multistaged permeator was made for the case of oxygen enrichment using air as feed. Published values of permeabilities for an ethyl cellulose membrane were used in the simulation. The operating conditions employed in this study and permeability values used for all the components involved are listed in Table 1. The feed pressure, $p_1$, was maintained at 7800 kPa, while the permeate pressure, $p_{n+1}$, was kept at 100 kPa in all cases. The effects of flow pattern on oxygen enrichment in the permeate stream and membrane area requirements were examined and are presented in the following paragraphs.

Oxygen enrichment curves for two flow patterns, i.e. perfect mixing and cocurrent flow, are illustrated in Fig. 2, which shows that cocurrent flow is generally a better flow pattern regardless of the number of permeation stages employed. Also, it can be seen from the figure that the number of permeation stages in the permeator is increased, the oxygen enrichment is improved, especially in the lower range of overall stage cuts. However, the effect of permeation stages on oxygen enrichment is significant only when their number is increased from one permeation stage to two or three; thereafter, the enhancement effect gradually decreases. For instance, at an overall stage cut of 0.01 and one permeation stage in the permeator with cocurrent flow pattern, the oxygen enrichment achieved was about 47% compared to 66%, 72%, and 76% for two and four permeation stages respectively at the same value of overall stage cut.

Comparison of membrane area requirements for different values of overall stage cuts and different numbers of permeation stages as generated by two flow patterns is illustrated in Fig. 3. As could be expected, the membrane area requirements increase with increasing number of permeation stages in the permeator. It can also be seen that the area requirements for cocurrent flow and for perfect mixing are almost the same. This indicates that in the case of cocurrent flow pattern, better separation achieved is due to more effective utilization of available membrane area compared to the perfect mixing case.

Conclusions

Performance of an internally multistaged perme-
ation module for gas separations has been analyzed using mathematical models based on cocurrent flow and perfect mixing. Simulation results obtained using the problem of oxygen enrichment from air indicated that, in an internally multistaged permeator the effect of the number of permeation stages on oxygen enrichment is significant when it is increased from one to two or three, and its effect decreases gradually with further increase in number of stages. A parametric study further revealed that the performance of the internally multistaged permeator with cocurrent flow yields better separation results than those obtained from the perfect mixing case.

**Nomenclature**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>membrane area</td>
<td>[m$^2$]</td>
</tr>
<tr>
<td>$L$</td>
<td>flow rate</td>
<td>[kmol·s$^{-1}$]</td>
</tr>
<tr>
<td>$I_{pr1}$</td>
<td>permeate flow rate</td>
<td>[kmol·s$^{-1}$]</td>
</tr>
<tr>
<td>$p$</td>
<td>pressure</td>
<td>[kPa]</td>
</tr>
<tr>
<td>$P$</td>
<td>permeability coefficient</td>
<td>[kmol·m$^{-2}$·s$^{-1}$·kPa$^{-1}$]</td>
</tr>
<tr>
<td>$R_1$</td>
<td>differential area ratio defined in Eq. (3)</td>
<td></td>
</tr>
<tr>
<td>$x$</td>
<td>mole fraction of oxygen</td>
<td></td>
</tr>
<tr>
<td>$X_{pr1}$</td>
<td>mol fraction of oxygen in permeate stream</td>
<td></td>
</tr>
<tr>
<td>$\phi$</td>
<td>individual stage cut, defined in Eq. (9)</td>
<td></td>
</tr>
<tr>
<td>$\Phi$</td>
<td>overall stage cut, defined in Eq. (10)</td>
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</tr>
<tr>
<td>$\delta$</td>
<td>membrane thickness</td>
<td>[m]</td>
</tr>
</tbody>
</table>

**Subscripts**

- $A$ = oxygen
- $B$ = nitrogen
- $f$ = feed
- $i$ = number of permeation stages

**Superscripts**

- * = local values
- - = initial permeation conditions

**Literature Cited**