Effect of fiber variation on the performance of countercurrent hollow fiber gas separation modules

J. Lemanski, G.G. Lipscomb *

Chemical and Environmental Engineering Department, University of Toledo, Toledo, OH 43606-3390, USA

Received 8 July 1999; received in revised form 25 August 1999; accepted 26 August 1999

Abstract

A theoretical and experimental study of the effects of variable fiber properties on countercurrent hollow fiber gas separation module performance is presented. Variations in ID, slow gas permeance, and selectivity are considered. Variability in any of these properties is detrimental to performance. The drop in performance increases as either property variation or product purity increase, but permeate mixing can improve performance. Fibers with higher permeation to feed rate ratios may stop producing a retentate product and may consume product from other fibers. The theory agrees well with experiments for the production of nitrogen from air. ©2000 Elsevier Science B.V. All rights reserved.

Keywords: Hollow fiber membrane; Gas separation; Theory

1. Introduction

Industrial hollow fiber membranes are produced by either extruding a polymer melt or polymer solution through a tube-in-orifice spinneret. Although fiber size and transport properties can be highly sensitive to variations in processing conditions [1], the effects of fiber property variations on module performance have not been fully quantified. Previous work has addressed the effects of variable fiber inner diameter (ID) [2–5] and wall thickness [5] for liquid applications including membrane processes for extraction, stripping, absorption, and dialysis. These works suggest that variations in fiber ID can reduce performance by 50% or more at low flows while wall thickness variations can increase or decrease performance, depending on the fraction of solute transferred.

Two recent works have considered the effects of fiber size and transport properties on hollow fiber gas separators. Rautenbach et al. [6] evaluated the effects of a Gaussian distribution of ID, skin thickness, and selectivity on the performance of a countercurrent hollow fiber air separator. Recovery of the nitrogen enriched retentate decreased by up to 30% with increased variation and increased retentate purity. For a given percent variation, fiber ID variations produced the largest decrease in recovery.

Lemanski et al. [7] evaluated the effects of permeate mixing and Gaussian variations in fiber ID, permeance, and selectivity on the performance of a cross-flow hollow fiber air separator for which the nitrogen enriched retentate stream is the desired product. As variation increased, recovery and flow rate decreased by as much as 70 and 80%, respectively, for
higher purity products. For a given percent variation, ID variations produced the largest performance decreases. As product purity increased, fibers with the highest permeation to feed rate ratios stopped producing and consumed product for modules with ID and permeance variations. Permeate mixing improved performance for modules with ID and permeance variations.

Here we theoretically quantify the effects of variation in fiber ID, permeance, and selectivity on the performance of a countercurrent hollow fiber module for binary gas separations. This work compliments the previous study of Rautenbach et al. by demonstrating experimentally and theoretically that some fibers can stop producing gas and actually consume the product produced by others. Furthermore, changes in both product recovery and product flow rate are reported and the effect of permeate mixing is evaluated.

2. Theory

In this work we consider variations in ID, slow gas permeance, and ideal separation factor. A Gaussian distribution of each fiber property is assumed,

\[ g(\phi) = \frac{1}{\sigma \sqrt{2\pi}} \exp\left[-\frac{(\phi - \bar{\phi})^2}{2\sigma^2}\right] \]

(1)

where \( \phi \) is the value of the fiber property, \( \bar{\phi} \) the mean value, and \( \sigma \) the standard deviation. The fraction of fibers for which the property value falls in the interval \([\phi, \phi + d\phi]\) is equal to \( g(\phi) d\phi \). For a bundle of fibers with a single variable material property, the average flow per fiber is given by

\[ \bar{f} = \int_{\phi_{\text{min}}}^{\phi_{\text{max}}} f(\phi) g(\phi) d\phi \]

(2)

where \( f(\phi) \) is the flow in fibers for which the material property has a value of \( \phi \), \( \phi_{\text{max}} \) the maximum value of the material property, \( \phi_{\text{min}} \) the minimum value, and the overbar indicates an average value.

One would expect Eq. (1) to be least accurate for variations in separation factor since actual distributions are likely skewed toward values less than the mean. However, the analysis described here could be modified to accommodate any distribution. Additionally, it is unlikely that a single fiber property would vary independently of others; for example, processing conditions that cause variations in ID could produce permeance variations as well. The goal of this work is to elucidate how variations in each property affect performance but the analysis could be modified to account for simultaneous property variations by replacing the single integral in Eq. (2) with multiple integrals over each property variation and substituting a joint probability distribution for \( g \).

The performance analysis is based on the following assumptions:

1. A binary, ideal gas is fed to the fiber lumens.
2. Lumen pressure changes are described by the Hagen–Poiseuille equation, and the magnitude of the lumen pressure drop is much smaller than the feed pressure.
3. Shell pressures are constant, with the shell-side permeate flowing countercurrent to the lumen-side retentate.
4. Concentration polarization and axial diffusion are negligible.
5. Properties of a single fiber are constant, i.e. concentration and length independent.
6. Operation is isothermal and steady-state.
7. Module tubesheet thickness is small relative to active fiber length.

Fig. 1 illustrates a typical hollow fiber gas separation module. Lumen-side and shell-side mass and momentum balances for each fiber yield the following dimensionless performance equations [8]:

\[ \frac{d\Theta_r}{dz} = \frac{d\Theta_p}{dz} = -(J_1 + J_2) \]

(3)

\[ \frac{d(x \Theta_r)}{dz} = \frac{d(y \Theta_p)}{dz} = -J_1 \]

(4)

\[ \frac{d\Pi^2}{dz} = -N^p \Theta_r \]

(5)

where \( z \) is the dimensionless fiber length (actual length/total active length), \( \Theta_r \) the dimensionless retentate molar flow rate (actual retentate flow/feed flow), \( \Theta_p \) the dimensionless permeate molar flow rate (actual permeate flow/feed flow), \( x \) the retentate mole fraction of the fast gas (i.e. the faster permeating species), \( y \) the permeate mole fraction of the fast gas, and \( \Pi \) the dimensionless retentate pressure (actual pressure/feed pressure). \( J_1 \) and \( J_2 \) are the dimension-
less molar permeation rates of the fast and slow gases, respectively, and are given by

\[ J_1 = \alpha N^h (x - \gamma y) \]  
\[ J_2 = N^h[(1 - x) - \gamma(1 - y)] \]

where \( \alpha \) is the ideal separation factor (ratio of fast gas permeance \( Q_1 \) to slow gas permeance \( Q_2 \)) and \( \gamma \) the dimensionless permeate to retentate pressure ratio (shell pressure/actual lumen pressure). Typically the lumen pressure drop is much smaller than the feed pressure, so that \( \gamma \) is approximately independent of \( z \).

The dimensionless groups \( N^p \) and \( N^h \) are given by

\[ N^p = \frac{256 \mu R_g T Z F}{\pi (ID)^4 p_f^2} \]  
\[ N^h = \frac{\pi (OD) Z Q_2 p_f}{F} \]

where \( \mu \) is the retentate viscosity, \( R_g \) the ideal gas constant, \( T \) the temperature, \( Z \) the total active fiber length, \( F \) the fiber molar feed rate, \( ID \) the fiber inside diameter, \( p_f \) the feed pressure, and \( OD \) the fiber outside diameter. Permeance \( (Q) \) is the ratio of the intrinsic, specific permeability to the effective membrane thickness based on the permeation area calculated using the fiber OD. \( N^p \) represents a dimensionless viscosity while \( N^h \) represents a dimensionless product of membrane area and permeance.

To solve these equations one must specify fiber properties (OD, ID, Z, \( \alpha \), \( Q_2 \)), a fluid property (\( \mu \)), operating conditions (\( T \), \( p_f \), \( \gamma \)), a feed rate for each fiber (\( F \)), and boundary conditions for the dependent variables (\( \Theta_r \), \( \Theta_p \), \( x \), \( y \), \( \Pi \)). In practice it is simpler to specify an outlet pressure for each fiber than a feed rate, since all fibers experience the same pressure drop. The feed rate (or in dimensionless form \( N^h \)) becomes an additional variable that is calculated simultaneously with the other unknowns. The boundary conditions for Eqs. (3)–(9) are:

\[ \Theta_r(z = 0) = 1 \]  
\[ \Theta_p(z = 1) = 0 \]  
\[ x(z = 0) = x_f \]  
\[ y(z = 1) = \frac{J_1}{J_1 + J_2} \bigg|_{z=1} \]  
\[ \Pi(z = 0) = 1 \]  
\[ \Pi(z = 1) = 1 - \Delta \Pi \]
where \( x_f \) is the lumen feed composition and \( \Delta \Pi \) the dimensionless lumen pressure drop (actual pressure drop/feed pressure).

Note that Eqs. (3)–(15) apply for each fiber in a bundle. For a bundle of fibers, mass balances for individual fibers are linked by the permeate composition, \( y \), since all fibers can contribute to the permeate. The value of \( y \) depends on the extent of radial mixing in-between fibers. Two limiting cases for mixing are considered here: no radial mixing and perfect radial mixing. One would expect the mixing that occurs in real modules to fall between these two limits. The degree of radial mixing in commercial devices is a function of fiber packing uniformity, port placement, the presence of baffles, and other design factors.

For the no-mixing case, the permeate from each fiber remains adjacent to that fiber, so each fiber in the bundle performs independently and no additional equations are needed to predict bundle performance. For the perfect-mixing case, the permeate from all fibers is well mixed so, for a given axial position, the permeate composition is the same for all fibers. This composition, \( \tau(z) \), is calculated by replacing the boundary condition given by Eq. (13) with

\[
\tau(z = 1) = \frac{\int_{\phi_{\text{min}}}^{\phi_{\text{max}}} J_1(\phi, z) F(\phi) g(\phi) \, d\phi}{\int_{\phi_{\text{min}}}^{\phi_{\text{max}}} [J_1(\phi, z) + J_2(\phi, z)] F(\phi) g(\phi) \, d\phi} \bigg|_{z=1}
\]  

(16)

and by using a common value for \( y \), \( y(\phi, z) = \tau(z) \), in the mass balances for each fiber.

Overall bundle performance is quantified by calculating the average flow rate, average recovery, and average composition of the product from all fibers. We assume a high purity retentate is the desired product, but similar expressions could be derived if the permeate was the desired product.

The average retentate flow exiting the fiber bundle is given by

\[
\overline{f_r} = \int_{\phi_{\text{min}}}^{\phi_{\text{max}}} \Theta_r(\phi, z) F(\phi) g(\phi) \, d\phi \bigg|_{z=1}
\]  

(17)

where \( \Theta_r(\phi, z) \) is the dimensionless molar retentate flow rate from those fibers for which the varying material property equals \( \phi \) at location \( z \). The average retentate recovery is given by

\[
\overline{\epsilon_r} = \frac{\overline{f_r}}{\int_{\phi_{\text{min}}}^{\phi_{\text{max}}} F(\phi) g(\phi) \, d\phi}
\]  

(18)

where the denominator is the average feed flow entering the fiber bundle. The fast gas composition in the product produced by combining the retentate flows from each fiber is given by

\[
\overline{x_r} = \frac{\int_{\phi_{\text{min}}}^{\phi_{\text{max}}} x(\phi, z) \Theta_r(\phi, z) F(\phi) g(\phi) \, d\phi |_{z=1}}{\overline{f_r}}
\]  

(19)

where the numerator is the average fast gas flow leaving the fiber bundle. Note that one associates ‘better’ performance with higher values of \( \overline{f_r} \) and \( \overline{\epsilon_r} \) for a given \( \overline{x_r} \).

Since an analytical solution does not exist for Eqs. (3)–(19), a numerical approximation was obtained. Eqs. (3)–(15) were solved using a finite difference technique. Second-order central differences were used to represent each derivative at interior nodes, and second-order forward and backward differences were used to represent derivatives at \( z = 0 \) and \( z = 1 \), respectively. The resultant set of non-linear algebraic equations was solved for the unknown values of \( \Theta_r, \Theta_p, x, y \) at each node and \( F \) at each fiber inlet using a full multi-variate Newton–Raphson algorithm. A grid-refinement analysis was used to evaluate numerical error.

Eqs. (16)–(19) were evaluated using Gauss–Hermite quadrature [9]. This method was developed to evaluate integrands including \( e^{-x^2} \) as in Eq. (1), and converts the integrals to finite sums. For example, Eq. (17) is approximated using

\[
\overline{f_r} \approx \frac{1}{\sqrt{\pi n}} \sum_{i=1}^{n} w_i \left( \Theta_r F \right)_{|z=1}
\]  

(20)

where \( n \) is the number of quadrature points, the subscript \( i \) indicates the value evaluated at a specific quadrature point (i.e. a particular value of \( \phi \)), and \( w_i \) are the weight factors. Values for \( \phi_i \) and \( w_i \) are tabulated as a function of \( n \). Increasing \( n \) improves the accuracy of the simulation.

Using the Gauss–Hermite quadrature method requires extension of the integration limits to \( \pm \infty \). We expect this will have little effect on the solution for
fiber property distributions that possess small \( \sigma \). Moreover, it also allows us to avoid assigning ambiguous values for \( \phi_{\text{min}} \) and \( \phi_{\text{max}} \).

3. Results

The results presented here are for \( \bar{\sigma} = 8 \) and \( \gamma = 0.1 \) which are representative of current commercial air separators and are independent of average values for the other fiber properties. Five quadrature points and 101 spatial nodes were chosen for all simulations based on a mesh refinement analysis. Increasing \( n \) from 3 to 5 and the number of nodes from 51 to 101 gave values for \( \bar{\sigma} \) and \( \bar{f}_r \) that differed by less than 1% over the entire \( \tau_r \) range considered.

In the following discussion, percent variation refers to the ratio of the standard deviation to the mean value of a property distribution expressed as a percentage \( (\sigma / \bar{\phi} \times 100\%) \). Relative retentate flow is the ratio of the retentate flow rate for a module with a variable fiber property to the flow from a module with no variation \( \left( f_r / \bar{f}_r (\sigma = 0) \right) \). Module performance refers to the value of the relative retentate flow or recovery for a given retentate purity. Better performance is associated with higher relative retentate flow or recovery at fixed retentate purity.

Fig. 2 compares the effects of a 15% variation in \( Q_2 \) and \( \alpha \) and a 10% variation in ID on performance for the no mixing case. Results for a 15% ID variation are not shown because retentate recoveries were negative over the entire retentate composition range for the quadrature point corresponding to the smallest ID fibers \( \text{(ID/ID}_0 = 0.57) \). The implications of a negative recovery are discussed below.

A small performance decrease is predicted for 15% variations in \( Q_2 \) or \( \alpha \). A much larger performance decrease is predicted for a 10% ID variation. For example, at \( \tau_r = 0.005 \), both \( \bar{e}_r \) and \( \bar{f}_r \) decrease by 10% or less for a module with 15% \( Q_2 \) or \( \alpha \) variations. However, for a module with 10% ID variation, \( \bar{e}_r \) and \( \bar{f}_r \) decrease by 35 and 50%, respectively. This implies the membrane area and work required to produce a 99.5% \( \text{N}_2 \) stream will be 50 and 100% larger, respectively, than in the absence of size variations!

The large influence of size variation on performance compared to variations in \( Q_2 \) and \( \alpha \) can be rationalized by examining the performance equations given in the previous section. Eqs. (6), (7) and (9) indicate that permeation rates for a particular fiber vary linearly with \( Q_2 \) and \( \alpha \). Thus a 10% change in either fiber property should produce a similar change in performance. Eqs. (5) and (8) indicate that for a given pressure drop, the fiber flow rate is proportional to \( \text{ID}^4 \). Thus a 10% change in fiber ID could produce a 46% change in flow rate. As a result, gas flows in smaller fibers will be lower than in larger fibers and smaller fibers will produce a higher purity retentate at a lower recovery than larger fibers.

![Graph](image-url)

Fig. 2. The effect of variations in ID, \( Q_2 \), and \( \alpha \) on performance with no permeate mixing: (a) retentate recovery as a function of retentate fast gas mole fraction and (b) relative retentate flow rate as a function of retentate fast gas mole fraction. The lines in both figures are: solid line, no variation; dash, 15% \( \alpha \) variation; dot, 15% \( Q_2 \) variation; dash-dot, 10% ID variation.
What happens at the product end when a fiber stops producing a retentate product? At the point of zero recovery, the lumen pressure drops below the module outlet pressure, inducing a flow from the product end into the fiber. This backflow of product retentate will completely permeate as well and reduce the product flow rate. To simulate this behavior, the component mass balance (Eq. (4)) at \( z = 1 \) is replaced by the boundary condition that the retentate composition is equal to the mixing-cup composition of the product from all producing fibers calculated using Eq. (19).

Fig. 3 illustrates typical changes in retentate recovery, pressure, and composition along the module length for fibers that correspond to each of the five quadrature points. These results correspond to a module with 10% ID variation and no permeate mixing producing a product with \( \tau_p = 0.005 \).

Fig. 3(a) illustrates how retentate recovery varies with axial distance. Positive values indicate a flow from the feed end to the product end while negative values indicate a flow in the opposite direction. Note that the permeate always flows from the product end to the feed end. Therefore fibers operate in a countercurrent mode when recoveries are positive but in cocurrent mode when recoveries become negative. The point of zero retentate flow, where the retentate has completely permeated, corresponds to \( \Theta_r = 0 \).
Fig. 3, this occurs at $z = 0.73$ for the smallest fiber and at $z = 0.99$ for the next smallest fiber. Only fibers with ID/ID > 0.86 contribute to the product since recoveries are negative for smaller fibers.

Fig. 3(b) illustrates how retentate total pressure varies with axial distance. For the three larger producing fibers, the pressure decreases monotonically from inlet to outlet. However, for the two smallest fibers, the pressure passes through a minimum at the point where $\Theta_1 = 0$ then increases to the outlet pressure. The presence of a pressure minimum is required for flow to occur from both ends of the fiber.

Fig. 3(c) illustrates how composition varies with axial distance. For all fibers, the fast gas composition drops rapidly near the feed end but the rate of change decreases with axial distance. For the three largest fibers, the minimum composition occurs at the outlet. For the two smallest fibers though, the composition passes through a minimum value then increases to $x_f$ at the outlet. The region near the outlet is magnified to illustrate this in Fig. 3(d). Note that the point at which the minimum occurs does not correspond to the point where $\Theta_1 = 0$, nor does $x = 0$ when $\Theta_1 = 0$ as observed for cross-flow modules [7]. This behavior arises from the cocurrent contacting that occurs in the non-product producing fibers. At the product end of a non-producing fiber, the retentate product that flows back into the fiber completely permeates to produce a permeate sweep with a fast gas composition equal to that of the retentate product. This sweep prevents the retentate composition from dropping to zero as observed in cross-flow. Moreover, the sweep alters the ratio of fast to slow gas permeation rates to produce a minimum in the fast gas composition prior to the point of zero retentate flow.

Fig. 4 illustrates the effect of percent variation in ID and permeate mixing on module performance. As the variation in ID increases, module performance decreases. For $x_f = 0.005$, a 5% ID variation decreases recovery and flow rate by 10 and 15%, respectively, for the no mixing case, and by 35 and 50%, respectively, for a 10% ID variation and no mixing.

Permeate mixing improves performance for a module with a given ID variation. For $x_f = 0.005$, a 10% ID variation decreases recovery and flow rate by 20 and 35%, respectively, for a module with mixing, compared to 35 and 50%, respectively, for a module with no mixing.

Fig. 4(c) illustrates the effect of permeate mixing on the permeate concentration profiles for a module with 10% ID variation producing a retentate with $x_r = 0.005$. The thinner lines show profiles for each quadrature point in a module with no mixing, and the thicker line shows the profile for all fibers in a module with mixing. As one would expect based on the retentate concentration profiles in Fig. 3(c), $y$ is higher at a given location for larger fibers because $x$ is higher for larger fibers. Therefore, mixing will improve performance for the two larger fibers and reduce performance for the three smaller fibers, since mixing decreases $y$ for the two larger fibers and increases $y$ for the three smaller fibers. For the module and operating conditions represented, the result is an increase in overall performance compared to the no mixing case.

Fig. 5 illustrates the effects of variation in $Q_2$ and permeate mixing on performance. The same qualitative performance effects were observed as for the ID variation cases. As $Q_2$ variation increases, performance decreases. For $x_f = 0.005$, a 15% $Q_2$ variation decreases recovery and flow rate by 5 and 10%, respectively, for the no mixing case, and by 40 and 60%, respectively, for a 30% $Q_2$ variation and no mixing. Permeate mixing improves performance for a given $Q_2$ variation. At $x_f = 0.005$, a 30% $Q_2$ variation decreases recovery and flow rate by 25 and 40%, respectively, for a module with mixing, compared to 40 and 60%, respectively, for a module with no mixing. Fibers with the highest permeation to feed rate ratios stop producing product and start consuming retentate as the retentate purity increases. For a module with 30% $Q_2$ variation and no permeate mixing, fibers with $Q_2/Q_f = 1.86$ stop producing when $x_f = 0.0052$; those with $Q_2/Q_f = 1.41$ stop when $x_f = 0.004$.

Fig. 6 illustrates the effects of percent variation in $\alpha$ and permeate mixing on module performance. Similar to the ID and $Q_2$ variation cases, performance decreases as the percent variation in $\alpha$ increases; at $x_f = 0.005$, a 15% $\alpha$ variation decreases recovery and flow rate by 3 and 5%, respectively, and by 25 and 40%, respectively, for a 30% $\alpha$ variation.

In contrast to the ID and $Q_2$ variations, fibers do not stop producing as purity increases and permeate mixing has no effect on performance. Apparently, a Gaussian $\alpha$ variation does not produce permeation to feed rate ratios that differ enough for some fibers to stop producing product.
Fig. 4. The effect of variations in ID on performance: (a) retentate recovery as a function of retentate fast gas mole fraction, (b) relative retentate flow rate as a function of retentate fast gas mole fraction, and (c) permeate fast gas mole fraction profiles for a module with 10% ID variation producing a product with $x_r = 0.005$. The lines in Figs. (a) and (b) are: solid line, no variation; long dash, 5% variation/well mixed; dot, 5% variation/no mixing; dash–dot, 10% variation/well mixed; short dash, 10% variation/no mixing. The lines in Fig. (c) are: long dash, ID/ID$_0$ = 0.71; dot, ID/ID$_0$ = 0.86; solid line, ID/ID$_0$ = 1; dash–dot, ID/ID$_0$ = 1.14; short dash, ID/ID$_0$ = 1.29; thick line indicates mixed permeate composition experienced by all fibers.

A qualitative summary of the results presented in this section is given in Table 1. In this table, performance change indicates the relative magnitude of the decrease in recovery and retentate flow that occurs for the same percent variation in the fiber properties listed while sensitivity to permeate mixing indicates the relative magnitude of the increase in recovery and retentate flow that accompanies permeate mixing.

### Table 1

<table>
<thead>
<tr>
<th>Property</th>
<th>Performance change</th>
<th>Sensitivity to permeate mixing</th>
</tr>
</thead>
<tbody>
<tr>
<td>ID</td>
<td>Large</td>
<td>Moderate</td>
</tr>
<tr>
<td>$Q_2$</td>
<td>Moderate</td>
<td>Moderate</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>Moderate</td>
<td>None</td>
</tr>
</tbody>
</table>
The apparatus simulates a single module that contains fibers with widely varying permeances. Both the separator and the tube share a common inlet, outlet, and pressure drop, like fibers with different permeances would in a single module.

A valve (V1 in Fig. 7) was installed at the end of the tube to control the fraction of total feed entering the tube. Varying the position of the valve corresponds to varying the number of low permeance fibers present. All of the data presented are for a single valve position.

At this position, approximately 10% of the total feed enters the tube. This implies that the module simulated by the separator and tube system contains greater than 10% low permeance fibers since fibers with lower permeation rates receive less feed than fibers with higher permeation rates.

System performance data were obtained by feeding pressurized air to the common inlet and adjusting valve V2 at the common outlet to control the feed rate. Flow rates (Sierra Instruments, Monterey, CA and Omega...
Engineering, Stamford CT), concentrations (Engineering Systems and Designs, Newark DE), and pressures were measured at the locations illustrated in Fig. 7. System performance was characterized by calculating the overall retentate recovery \( (F_5/F_1) \) using the nomenclature in Fig. 7) and separator recovery \( F_3/(F_3 + F_4) \) as a function of retentate concentration \( (C_1) \).

System performance was also predicted using the theory. Instead of a continuous Gaussian variation, a discrete distribution of fiber properties was used. All of the fibers in the air separator were assumed to possess the average values of ID, \( Q_2 \), and \( \alpha \). Since these values, and values for OD and Z, were not published for the separator, the required parameters were estimated from air separation performance data obtained with valve V1 closed. Values of \( \alpha \) and the product \( ODQ_2 \) (needed to calculate \( N^h \) ) for the separator were estimated from Eqs. (3)–(7) and (10)–(13) by minimizing the sum of the squared difference between predicted and experimental recoveries over a range of retentate compositions. The value of the ratio \( Z/ID^4 \) (needed to calculate \( N^p \) ) for the separator was estimated from Eqs. (5), (14) and (15) by minimizing the sum of the squared difference between predicted and experimental pressure drops over a range of retentate recoveries. The parameters estimated for the separator are \( \alpha = 5.6 \), \( ODQ_2 = 4.3 \times 10^{-14} \) mol Pa\(^{-1} \) s\(^{-1} \), and \( Z/ID^4 = 7.8 \times 10^{15} \) m\(^{-3} \). For the tube, \( Q_2 \), and therefore \( N^p \), were assumed to be zero because changes in flow rate and concentration over the tube length were not measurable with the meters used. Due to the presence of valve V1, Eq. (5) was not used to calculate the tube flow rates as a function of pressure drop because pressure losses in the tube were found to be insignificant compared to the valve loss. Instead, the pressure drop for the valve and tube were assumed to be proportional to the square of the tube flow rate [10]. The proportionality constant, the valve flow coefficient, was obtained from the manufacturer’s literature.

Fig. 8 illustrates how recovery varies with product composition for the air separator and tube system. Experimental data and theoretical performance predictions are shown for both the system and the separator operating at \( p_f = 4.1 \times 10^5 \) Pa and \( \gamma = 0.24 \). Mass balance errors were less than 5% for the experimental data. For high retentate recoveries the retentate fast gas (O\(_2\)) composition decreases as the recovery decreases. However, as the retentate recovery decreases past 0.32 the retentate composition passes through a minimum value of 0.067. Further decreases in recovery lead to
an increase in composition as the retentate produced by the separator becomes a smaller portion of the total retentate produced by the system. The retentate from the tube possesses the same fast gas composition as the feed which, when mixed with the declining retentate flow from the separator, produces this minimum.

At the lowest system recovery measured, the separator recovery was $-0.04$ indicating the module had stopped producing gas and a portion of the retentate produced by the tube was flowing back into the module. This back flow into the separator was verified by reversing the mass flow meter at the separator outlet and by measuring the composition at the separator outlet and finding it equal to the tube product composition.

The agreement between experiment and theory is good. The minimum retentate composition and associated system recovery are predicted accurately as is the increase in product $O_2$ composition with further decrease in system recovery. Additionally, at the lowest system recovery measured, the predicted module recovery of $-0.042$ is in good agreement with the experimental value of $-0.040$. These results help validate the theory presented here and the performance decline predictions, including the possibility for complete permeation by fibers in a module.

5. Conclusions

An analysis of fiber property variation effects on countercurrent hollow fiber gas separator performance is presented. The analysis utilizes standard mass and momentum balances and gas permeation expressions. Gaussian variations in ID, slow gas permeance, and selectivity are explicitly considered.

Performance decreases as the level of variation increases. ID variations decrease performance most for a given level. Permeance and selectivity produce similar but less pronounced performance decreases.

Permeate mixing can improve module performance in the presence of ID and permeance variations. However, permeate mixing does not affect performance when selectivity variations are present.

At higher product purities, some fibers can stop producing and consume a fraction of the product from other fibers. This phenomenon is predicted to occur only in the presence of ID and permeance variations, and was observed experimentally in an apparatus designed to mimic a module with permeance variations.

We believe the results presented here could prove useful in several ways. First, when making strategic decisions about the development of new membrane processes, one should consider the limitations imposed by fiber variability. Typically, process models based on uniform fiber properties are used to estimate performance in economic evaluations. However, the performance decline due to fiber variability may be significant and should be considered, especially for high purity applications. Second, fiber variability must be kept below target levels to ensure modules will perform as desired. One could use the analysis presented here to determine the targets and guide plant efforts for process improvement. For example, if a change in separation factor variability has little effect on performance but reducing permeance variability improves it, one might consider process modifications that reduce permeance variability even if separation factor variability increased. Third, one could use this analysis to evaluate alternative module designs and systems for the production of high purity products. Fiber variability greatly reduces the performance of a single countercurrent stage but the use of multiple stages, cascades, or other novel internal contacting patterns [8] might offer improved performance. The
6. List of Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
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<tr>
<td>( f )</td>
<td>product flow rate, (mol s(^{-1}))</td>
</tr>
<tr>
<td>( F )</td>
<td>feed flow rate (mol s(^{-1}))</td>
</tr>
<tr>
<td>( g )</td>
<td>fiber property distribution function</td>
</tr>
<tr>
<td>ID</td>
<td>fiber inside diameter (m)</td>
</tr>
<tr>
<td>( J )</td>
<td>dimensionless component molar permeation rate</td>
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<tr>
<td>( n )</td>
<td>number of Gauss–Hermite quadrature points</td>
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<tr>
<td>( N^p )</td>
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<tr>
<td>( N^h )</td>
<td>dimensionless group defined by Eq. (9)</td>
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<tr>
<td>OD</td>
<td>fiber outside diameter (m)</td>
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<tr>
<td>( p )</td>
<td>total pressure (Pa)</td>
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<tr>
<td>( Q )</td>
<td>permeance (mol m(^{-2}) Pa(^{-1}) s(^{-1})), ratio of intrinsic permeability to effective membrane thickness based on the permeation area calculated using the fiber OD</td>
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<tr>
<td>( R_g )</td>
<td>ideal gas constant (Pa m(^3) mol(^{-1}) K(^{-1}))</td>
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<tr>
<td>( T )</td>
<td>retentate temperature (K)</td>
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<tr>
<td>( w_i )</td>
<td>Gauss–Hermite quadrature weight factor</td>
</tr>
<tr>
<td>( x )</td>
<td>mole fraction of faster permeating gas component in retentate</td>
</tr>
<tr>
<td>( y )</td>
<td>mole fraction of faster permeating gas component in permeate</td>
</tr>
<tr>
<td>( z )</td>
<td>ratio of actual active fiber length to total active fiber length</td>
</tr>
<tr>
<td>( Z )</td>
<td>total active fiber length (m)</td>
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<tr>
<td>( \alpha )</td>
<td>ideal separation factor, ratio of fast gas permeance to slow gas permeance</td>
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<tr>
<td>( \gamma )</td>
<td>ratio of permeate pressure to retentate pressure</td>
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<tr>
<td>( \Delta )</td>
<td>change in</td>
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<td>( \Theta )</td>
<td>recovery, ratio of actual molar flow rate to feed rate</td>
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<tr>
<td>( \mu )</td>
<td>retentate viscosity (Pa s)</td>
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<td>( \Pi )</td>
<td>ratio of actual total pressure to feed pressure</td>
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<td>( \sigma )</td>
<td>standard deviation</td>
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<td>( \phi )</td>
<td>arbitrary fiber property</td>
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Subscripts and superscripts

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<thead>
<tr>
<th>Subscript</th>
<th>Superscript</th>
<th>Description</th>
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<tbody>
<tr>
<td>f</td>
<td>feed</td>
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</tbody>
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max | maximum value |
min | minimum value |
p | permeate value |
r | retentate value |
1 | value for faster permeating gas component |
2 | value for slower permeating gas component |
\( \bar{\text{}} \) | average value |

Acknowledgements

The authors acknowledge partial support of this work by the National Science Foundation through grant CTS-9408414 and MG Generon for the modules used.

References