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DEVELOPMENT OF A SIMULATION MODEL FOR A HOLLOW-FIBER MEMBRANE N_2–H_2 SEPARATION SYSTEM

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Abstract. An interactive simulation model to assist the design process of a hollow fiber membrane for gas separation system has been developed. The model is capable of predicting the performance of a multistage hollow fiber membrane permeator, regardless of any operating conditions employed. It takes into account the effect of feed and permeates pressure drop to simulate the multistage membrane process that is more applicable to the gas separation system. Various types of data profile are able to be produced such as permeate flux and feed pressure profile along the hollow fiber length for each membrane module. The simulation model has been tested using data from the real separation system. The results revealed that the model has been successfully predicted the performance of the N_2–H_2 mixtures separation using a commercial size multiple stage hollow fiber membrane permeator. The maximum error is generally not over than 5%. The established model provides a more convenient process in designing a multistage membrane permeator system.

Abstrak. Sebuah model penyelaku interaktif telah dibangunkan bagi membantu mereka bentuk membran gentian geronggang untuk sistem pemisahan gas. Model ini mampu meramal prestasi membran penelap gentian geronggang pelbagai tahap dalam berbagai bentuk keadaan operasi. Ia mengambil kira kesan kejatuhan tekanan suapan dan tekanan tertelap untuk menyelaku proses membran pelbagai tahap yang mana lebih praktikal kepada sistem pemisahan gas. Berbagai jenis profil data dapat dihasilkan seperti fluks tertelap dan profil tekanan suapan sepanjang gentian geronggang bagi setiap modul membran. Model penyelaku ini telah diuji menggunakan data dari proses pemisahan sebenar. Keputusan yang diperolehi mendapati model ini telah berjaya meramalkan prestasi pemisahan campuran gas N_2–H_2 menggunakan membran penelap gentian geronggang pelbagai tahap bersaiz komersial. Ralat maksimum adalah secara purata tidak melebihi 5%. Model yang telah diuji ini memudahkan proses mereka bentuk sistem membran penelap pelbagai tahap.

Key Words: Gas separation, hollow fiber membrane system, modeling and simulation

1.0 INTRODUCTION

Membrane technology for gas separation has been shown to be practical alternative to the conventional separation technique such as absorption, adsorption and cryogenic system. At present, this system is used to remove CO_2

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from natural gas, recovery of helium from natural gas, oxygen enrichment, natural gas dehydration and sweetening, hydrogen recovery in ammonia production, air fractionating from the production of blanketing nitrogen and landfill gas upgrading. The performance of a membrane gas separation system is directly related to the separation mechanism. Gas permeation through a membrane can be defined as a process in which gas molecules dissolves in a membrane skin and then diffuse through the membrane support to be released on the other side of it. This pressure difference is the driving force, which causes a diffusional flow of gas through the membrane.

For evaluating the performance of a membrane gas separation process, it is essential to have a reliable mathematical model that can sufficiently predict these separation mechanisms. Some significant efforts in this area have been made by Stern and Walawender [1], Blaisdell and Kemmeryer [2], Pan and Hadgood [3], Thorman *et al.* [4], Pan [5], Baolin and Guoliang [6], and Kovvali [7]. Generally, constant permeation rates and negligible feed pressure drops along the membrane surface are assumed. In these particular systems, the model is unable to estimate and establish the variation of feed pressure profile as well as the feed and permeate compositions profile along the fiber length. Therefore in this paper, a more reliable mathematical model which takes into account the effect of feed pressure drops, and a proper solution strategy to the model are introduced. The present analysis actually holds in general and is applicable to any binary mixtures and various types of polymer membranes. However the numerical results presented are based on a hydrogen-nitrogen mixtures using a commercial specification of a five-stage membrane permeator system.

2.0 MATHEMATICAL MODELING

The mathematical model developed in this study is similar to that one used by Pan [5]; in addition, it is also taking into consideration the feed and permeate pressure drops. The assumptions utilized in the analysis are as follows:

(1) The feed is on the skin side of the asymmetric membrane.
(2) The feed pressure and permeate pressure vary with the position on the membrane surface [8].
(3) No mixing of permeate fluxes of different compositions occurs inside the porous supporting layer of the membrane.
(4) The porous supporting layer has negligible resistance to permeate fluxes and the diffusion along the pore path is insignificant due to high permeate fluxes.
(5) Feed pressure drop equation was derived from the mechanical energy balance as given by Dodge [9] for flow of gases under isothermal conditions.

(6) The permeate flow inside the fiber is governed by the Hagen-Poiseuille equation as used by Pan [5].

The governing equations for the asymmetric hollow fiber membrane permeator comprises three major equations, namely, material balance equations, permeation rate equations, and feed as well as permeate pressure drops equations. The following formulation is set up in terms of the cross flow pattern. This flow pattern occurs when the flux permeates through the skin side and the porous support of the membrane is in perpendicular direction to the feed flow, as shown in Figure 1.

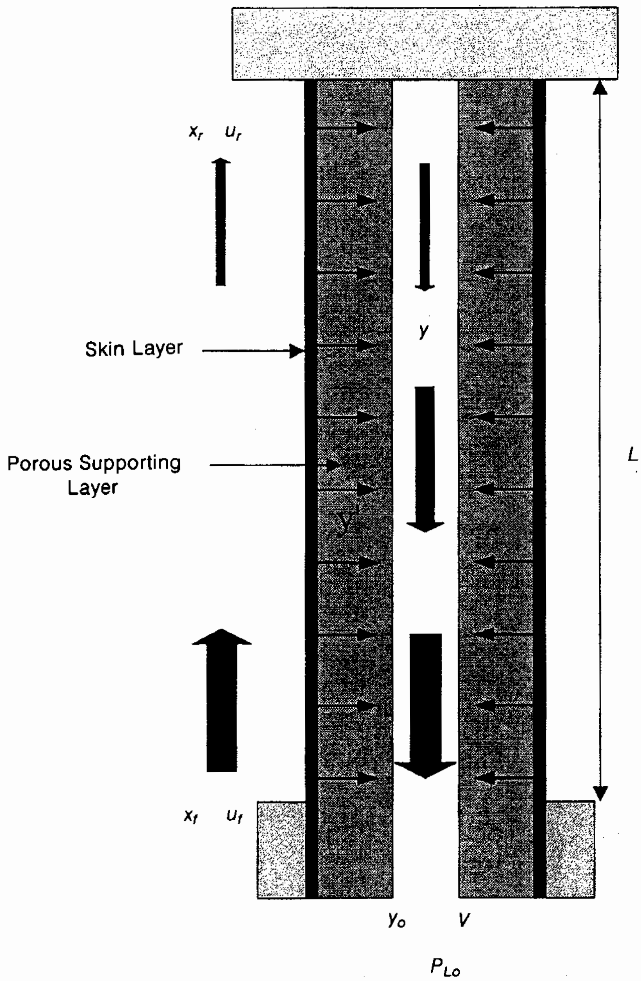

Figure 1 Cross Flow Pattern of Asymmetric Hollow Fiber Membrane Permeator

2.1 Material Balance Equations

The overall material balance for the separation of binary mixtures in a hollow fiber asymmetric membrane permeator is given by

$$\frac{d\theta}{dl_t} = 1 - \frac{u_r}{u_f}, \tag{1}$$

$$\frac{d(\theta y_1)}{dl_t} = x_{1f} - x_{lr}\left(\frac{u_r}{u_f}\right), \tag{2}$$

where $l_t = l_h/L$.

2.2 Permeation Rate Equations

In a crossflow pattern, the concentration of the local permeate stream leaving the membrane active layer (skin), y', is generally different from the bulk permeate stream, y [5]. The total permeate flux of component 1 and 2 can be represented by the equations (3) and (4), respectively:

$$\frac{d(u_f x_l)}{dl_h} = -\pi D_o \ NP_H \ (\Phi_l/l_m)(x_l - \gamma y'), \tag{3}$$

$$\frac{d\{u_f(1-x_l)\}}{dl_h} = -\pi D_o \ NP_H \ (\Phi_2/l_m)\{(1-x_l) - \gamma(1-y') \tag{4}$$

Local permeate concentration, y', is then defined as follows,

$$y' = \frac{1+(\alpha-1)(\gamma+x) - \{[1+(\alpha-1)(\gamma+x)]^2 - 4\gamma\alpha x(\alpha-1)\}^{1/2}}{2\gamma(\alpha-1)}, \tag{5}$$

where α is the selectivity.

2.3 Feed and Permeate Pressure Drops Equation

The derivation of the feed pressure drop equation assumes a compressible gas flowing under pressure in a tube using a mechanical energy balance. After simplifications, it is reduced to

$$\frac{d(P_H)}{dl_h} = -\frac{2zRTFG^2}{P_H R_H \Omega_m \ g_c}, \tag{6}$$

where the friction factor, F, the hydraulic diameter, R_H and the molecular weight of the gas mixtures in the retentate stream, Ω_m are given, respectively, as follows:

$$F = 0.008\,(4R_H)^{-1/3}\,, \tag{7}$$

$$R_H = 0.25\left(\frac{D_{mm}^{\,2}}{ND_o - D_o}\right), \tag{8}$$

$$\Omega_m = \Omega_1 y_1 + \Omega_2(1 - y_2) \tag{9}$$

As the permeation flux is small compared to the axial flow rate, the Hagen Poiseuille equation for flow through the tubes can be used to describe permeate pressure drop in the tube side of the fiber [5]:

$$\frac{d(\gamma)^2}{dl_t} = C_k\left(\frac{\mu_m}{\mu_2}\right)\theta, \tag{10}$$

where

$$C_k = \frac{256RT\mu_2 LU_f}{g_c\,\pi ND_i^{\,4}P_H^{\,2}}. \tag{11}$$

3.0 SOLUTION STRATEGY OF THE MATHEMATICAL MODEL

A numerical integration subroutine, which incorporates the Runge-Kutta Fehlberg and adaptive step size method, has been established using Visual Basic version 5.0. This subroutine is invoked with the following initial conditions to integrate the governing equations.

For $lt = 0$:
$u_o = u_f$, $x_l = x_{1f}$ and $P_H = P_{Ho}$.

The initial values of y' is calculated from the given feed composition using equation (10). The algorithm for the iteration method is described below:

(a) As a first approximation, γ is assumed everywhere to be equal to γ_o .
(b) Then equation (6) and (7) are integrated simultaneously from $l_t = 0$ to $l_t = 1$, giving the profile of feed flow and feed composition (u_f and x_{1f}). Utilizing the values of u_f and x_{1f}, equation (11) is integrated from $l_t = 0$ to $l_t = 1$, giving the profile of feed pressure along the fiber length. This step is repeated until the calculated pressures converge to a satisfied value, that is when the absolute estimate error falls below the tolerance value.
(c) The next step is to integrate equations (4) and (5) from $l_t = 0$ to $l_t = 1$, utilizing the results obtained above and the following initial conditions:

$$\theta = 0 \text{ and } y_{1o} = y'f,$$

after which a $\theta - y - l_t$ relation and the permeate outlet y_{l_o} are obtained.

(d) Utilizing the $\theta - y - l_t$ relation obtained in the step (c), a new $\gamma - l_t$ relation is obtained by integrating equations (15) from $l_t = 1$ to $l_t = 0$ (where $\gamma = \gamma_o$).

(e) With a new $\gamma - l_t$ relation, steps (b) to (d) are repeated, until the calculated y_{10} converges to a satisfied value. The bulk permeate composition y at fiber outlet can be calculated using material balance from the composition of flows of the feed and retentate streams.

4.0 RESULTS AND DISCUSSION

The prediction of N_2–H_2 separation performance of the hollow fiber membrane permeator was based on the data given in Table 1 below.

Table 1 Input Variables for the Calculation Process

Commercial Size Membrane Permeator	
Membrane area	= 920000 cm^2
Number of active fibers	= 16270
Hollow fiber inside diameter	= 0.02 cm
Hollow fiber outside diameter	= 0.06 cm
Membrane module diameter	= 12 cm
Length of membrane module	= 300 cm
Initial Value of Permeation Rates	
Nitrogen permeation rate	= 3.0×10^{-5} cm^3 (STP)/cm^2.s.cmHg
Hydrogen permeation rate	= 1.2×10^{-6} cm^3 (STP)/cm^2.s.cmHg
Operating Conditions	
Feed pressure	= 6308.02 cmHg
Permeate pressure	= 1860.2 cmHg
Volumetric feed flow rate	= 1.3×10^6 cm^3 (STP)/s
Operating temperature	= 313 K
Feed composition	= 60% Hydrogen

4.1 Feed Pressure Profile

Figure 2 shows typical plots of the feed pressure profiles along the membrane surface for feed gas composition 60% hydrogen. As seen from the graph, for each stage, the predicted feed pressure profile decreased along the hollow fiber length toward the feed outlet. This is due to frictional losses taking place when fluid streams flow through the narrow channels of permeator [7]. In this case, feed pressure drops for the succeeding membrane permeator is lesser compared to the first stage; this is shown in Figure 3. The latter is probably due to the fact that a decrease in inlet feed flow rate for each stage reduced the frictional loses caused by the flowing fluids. The total feed pressure drops for the entire stages is about 189.7 cmHg, that is, about 4.3% when compared to the pressure difference between inside and outside of the fiber (6308.02 − 1860.2) = 4447.82 cmHg.

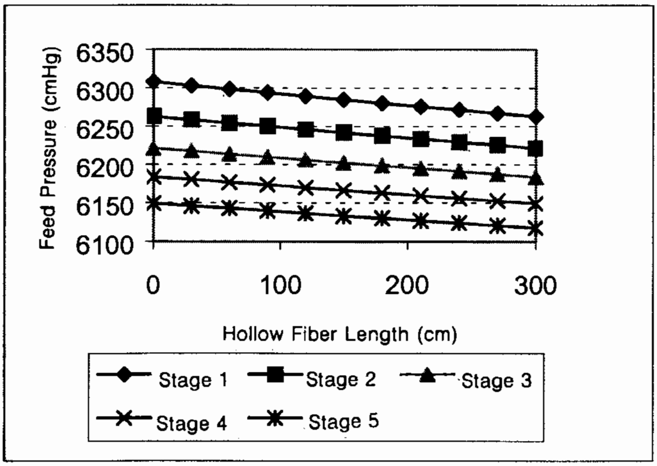

Figure 2 Feed Pressure Profile

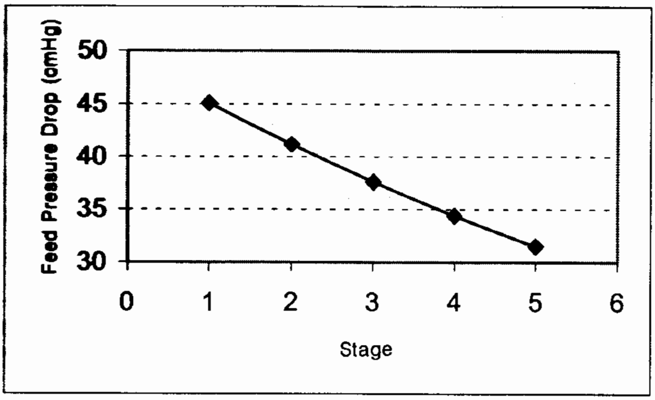

Figure 3 Feed Pressure Drop for Each Stage

Though, it seems to be small in percentage, such feed pressure drops should not be neglected as its effect is significant to the performance of the membrane permeator, specially for the permeate gas recovery. As shown in Figure 4, the plotted graph, which neglects the feed pressure drop has overestimated the

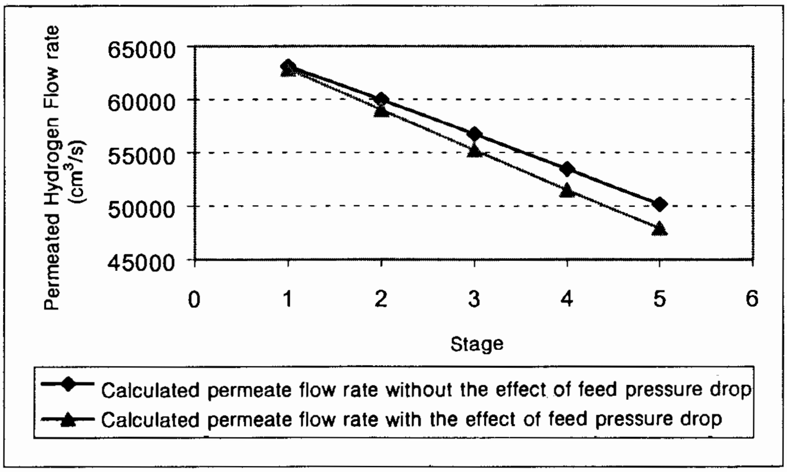

Figure 4 Permeated Hydrogen Flow Rate (cm^3/s)

permeated hydrogen flow rate for each stage.

The difference in the permeated hydrogen flow rates is found to increase with increasing number of stages, with a maximum value of 2216.5 cm^3/s.

4.2 Verification of the Simulation Results

A pilot test data was used to verify the simulation result from this study. The pilot test data was taken from a three-stage commercial size hollow fiber membrane system. The data was also employed by Baolin and Guoliang [6] for verification of their calculation method. However, they do not consider the effect of feed pressure drops in their study. In this study, the permeated gas flow rate simulated using the method described in this paper and the calculated result by Baolin are compared with the values measured from the pilot test data.

Table 2 Comparison of Simulated Hydrogen Permeate Flow Rate with the Measured Values

Stage	Pilot Test Data (m^3(STP)/s)	Calculated Result [6] m^3(STP)/s	Error %	Present Simulated Result m^3(STP)/s	Error %
1	212	201	5.19	205.9	2.88
2	151	160	5.96	152.1	0.73
3	114	120	5.26	105.2	7.72
Overall error			5.47		3.78

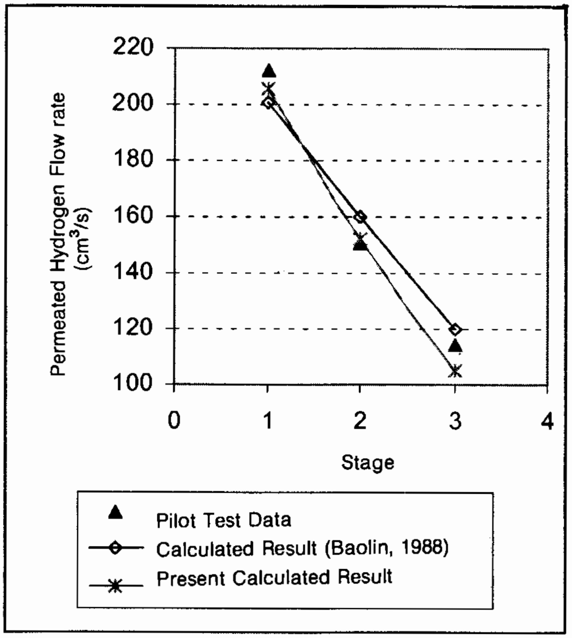

Figure 5 Comparison of Calculated Permeated Hydrogen with the Measured Values

From Table 2 and Figure 5, it can be seen that the simulated results from this study closely coincide with the measured values. The overall error is about 3.78%, as compared to Baolin's, which is about 5.47%.

5.0 CONCLUSION

The improved calculation method has been successfully employed and is capable of predicting the performance of the multistage hollow fiber membrane permeator that is more applicable to the gas separation industry. The method, which takes into account the effect of feed pressure drop, has predicted a better permeated hydrogen flow rate for a large-scale gas separation of N_2–H_2 mixture. The feed pressure drops was found to be significantly increasing with an increasing in the number of stages. This condition has affected the performance of the membrane permeator, specially the permeated gas flow rate, as membrane is a pressure driven system. Thus, the effect of feed pressure drop should not be neglected in estimating the performance of a large-scale multistage membrane permeator system.

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LIST OF SYMBOLS

A	membrane area, (cm^2)
D_o	hollow fiber outside diameter, (cm)
D_{mm}	hollow fiber membrane module diameter, (cm)
F	friction factor
g_c	newton's law conversion factor, ($g.cm/dyne.s^2$)
G	mass flow rate per cross sectional area of the membrane, ($g/s.cm^2$)
L	hollow fiber length, (cm)
l_m	effective skin thickness of the asymmetric membrane, (cm)
l_H	hollow fiber length variable, (cm)
l_t	l_k/L, dimensionless
N	total number of active fiber
P_H	feed-side pressure, (cmHg)
P_L	permeate-side pressure, (cmHg)
Φ/l_m	permeation rate coefficient for component i, [$cm^3(STP)/cm^2.s.cmHg$]
R	universal gas constant, ($cm^3.cmHg/mol.K$)
R_H	hydraulic radius, (cm)
T	absolute temperature, (K)
u_f	feed gas flow rate per unit length of the hollow fiber, [$cm^3(STP)/s.cm$]
u_r	retentate gas flow rate per unit length of the hollow fiber, [$cm^3(STP)/s.cm$]
U_f	feed-side gas flow rate for the entire membrane, [$cm^3(STP)/s.cm$]
x_1	mole fraction of the fast gas in the feed
y'	local permeate concentration on the membrane surface, mol fraction
y	permeate concentration in the bulk permeate stream, mol fraction
y_1	mole fraction of the fast gas in the permeate
Z	gas compressibility factor

Greek letter

α	membrane selectivity
γ	ratio of permeate to feed pressure
μ_2	viscosity of the less permeable component, (cp)
μ_m	viscosity of the gas mixture, (cp)
θ	ratio of permeate to feed gas
Ω_i	molecular weight of component i, (g)
Ω_m	molecular weight of the gas mixture, (g)