An Effective Iterative Algorithm for Modeling of Multicomponent Gas Separation in a Countercurrent Membrane Permeator

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Abstract: A model is developed for separation of multicomponent gas mixtures in a countercurrent hollow fiber membrane module. While the model's solution in countercurrent module usually involves in a time consuming iterative procedure, a proper initial guess is proposed for beginning the calculation and a simple procedure is introduced for correcting the guesses, hereby the CPU time is decreased essentially. The model's predictions are compared with the experimental data and a good agreement is achieved. By using data taken from a LAB unit in Isfahan, the proposed model is applied to investigate the feasibility of membrane process for hydrogen separation in this unit. It is revealed that a high hydrogen purity and recovery could be achieved in the permeate stream, while the required area increases at higher stage cut or higher permeate pressure.

Keywords: Membrane, Gas Separation, Modeling, Hydrogen Separation

1. Introduction

Gas separations by membranes have received much attention in recent years. The main advantages of membrane process over conventional gas separation processes are: lower energy consumption and lower capital cost, simplicity in operation and their reliability(Brunetti, et al., 2009; David, et al., 2012; David, et al., 2011; Hosseini, et al., 2010; Hosseini, et al., 2008; Kaldis, et al., 2000)Therefore, membrane separation processes can provide an ideal alternative to PSA and cryogenic separation processes conventionally applied in hydrogen purification(David, et al., 2011). Hydrogen recovery by membrane process has been one of the first commercial applications of this technology in the field of gas separation. The major application of hydrogen selective membranes include fuel cell technology and membrane rector processes(Hosseini, et al., 2008). Recovery of hydrogen from hydrocarbon streams in refineries, separation of H_2/N_2 in ammonia synthesis plants and separation of H_2/CO_2 are the other important application of membranes in field of hydrogen separation and purification(Brunetti, et al., 2009; David, et al., 2012; David, et al., 2011; Hosseini, et al., 2010; Hosseini, et al., 2008; Kaldis, et al., 2000). Hydrogen recovery in refineries and H_2/N_2 separation in ammonia plants can be considered as the most important application of hydrogen selective membranes in chemical industries (Brunetti, et al., 2009).

Low selective polymeric materials such as cellulose acetate polymers were primarily used

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in synthesis of hydrogen separation membranes. However, the new polymer membranes with improved flux and selectivity are now available in commercial scale. Polyimides (Ube, Praxair) and brominated polysulfone (Permea) are some examples of these new selective membranes (Baker, 2000).

Most of membrane applications in gas separation mentioned here are involved in multi-component gas streams. Likewise the binary mixtures, the prediction of membrane performance and evaluation of a membrane process are very important. Modeling and simulation of a membrane process is considered as a powerful tool used in achieving this objective. A successful modeling can provide valuable information for the design, optimization and economic evaluation of overall separation process with minimum cost (Kaldis, et al., 2000).

The performance of a membrane module of gas permeation can be modeled based on flow patterns and operating conditions. These models can be classified as countercurrent flow, cocurent flow, cross flow, perfect (complete) mixing and on-side mixing. Calculation methods for binary systems for these models have been introduced by Kammermeyer et al. and other researchers(Shindo, et al., 1985). Shindo et al. developed approximate calculation methods for multicomponent gas streams. Their calculations include all of flow patterns pointed out(Kaldis, et al., 2000). An iterative solution method has been described by Stern et al. for multicomponent systems with perfect mixing. Brubaker and Kammermeyer developed a calculation procedure for separation of ternary and quaternary mixtures in the same flow pattern(Shindo, et al., 1985). Sengupta and Sirkar reported numerical analysis of the separation of ternary mixtures in concurrent and countercurrent permeators(Kaldis, et al., 2000). Pan and Habgood developed calculation methods for multicomponent systems in the cross flow pattern(Shindo, et al., 1985). An iterative solution procedure for multicomponent systems in countercurrent flow pattern was developed byMcCandless (1990). The introduced method in this work is a modified version of the complex method of Box and utilizes a direct search maximization routine .

In this study, a hollow fiber membrane permeator used for separation of a multicomponent gas mixture is modeled and model equations are solved by numerical method. An efficient iterative method was developed to solve the governing equations for a countercurrent pattern. In contrast to the procedure reported by(McCandless, 1990), here

a simple procedure proposed and applied in order to provide new trial values based on result of previous iterations. The experimental data from (Kaldis, et al., 2000)was used to validate the developed model. After model validation, it was applied to Pacol process hydrogen recycle stream in a LAB production unit in Iran which is a mixture of hydrogen and hydrocarbons. The hydrogen purity in permeate stream and the membrane area is predicted for a specified stage cut. The effect of stage cut and permeate pressure on composition of feed and permeate streams is investigated.

2. Modeling of Membrane Permeator

Two membrane modules are commercially used for gas separation applications. These modules are tubular hollow-fiber and spiral-wound module. The hollow fiber module is best described by countercurrent flow pattern as shown in Figure 1. This module is conventionally applied in hydrogen separation processes(Zolandz, 1992). For this reason, the governing equations in this work are developed based on a countercurrent flow pattern.

The following assumptions are taken into account to formulate the model:

- The transport of gases through membrane obeys Fick's law.
- The permeability of each component is independent of the other components in the mixture. In other words, the permeability of each component is considered to be the same as that of pure component.
- The pressure drop through the feed and permeate sides is neglected.
- The flow pattern in feed and permeate sides is considered to be as plug flow.
- The composition of components are considered to be the same at membrane/permeate surface and bulk of permeate stream. This assumption is valid for symmetric membrane or a membrane with ultrathin skin layer and a highly porous support layer.
- The permeability coefficients are assumed to be constant.

The schematic diagram of a differential element for a counter current flow permeator is shown in Figure 2. The overall material balance and the balance for component i over this differential shell are(Shindo, et al., 1985):

$$-dF = dG = dA \sum_{i=1}^{n} \frac{Q_i}{\delta}(p_f x_i - p_p y_i) \qquad (1)$$

$$-d(Fx_i) = d(Gy_i) = dA\frac{Q_i}{\delta}(p_f x_i - p_p y_i) \qquad (2)$$

Q is usually expressed based on barrer or GPU. 1 barrer=10^{-10} cm^3 (STP).cm/cm^2.cm Hg.s and 1GPU=10^{-6} cm^3(STP) /cm^2.cmHg.s.

Solving Eqs. (1) and (2) for dx_i yields to :

$$dx_i = \frac{-dA}{F}\left[\frac{Q_i}{\delta}(p_f x_i - p_p y_i) - x_i\sum_{i=1}^{n}\frac{Q_i}{\delta}(p_f x_i - p_p y_i)\right] \quad (3)$$

The following dimensionless variables are introduced to simplify the equations :

$$s = A\frac{Q_m p_f}{F_f \delta} \qquad (4)$$

$$f = \frac{F}{F_f}, f_o = \frac{F_o}{F_f} \qquad (5)$$

$$\theta = 1 - f_o = \frac{G}{F_f}, r = \frac{p_p}{p_f} \qquad (6)$$

$$q_i = \frac{Q_i}{Q_m} \qquad (7)$$

where, Q_m is the permeability of the base component, usually the most permeable component (hydrogen in this study), F_f and F_o are the flow rates of feed and rejecte stream, respectively.

By writing Eqs. (1) and (3) in terms of dimensionless variables, the following arrangement is obtained(Shindo, et al., 1985):

$$\frac{df}{ds} = -\sum_{i=1}^{n} q_i(x_i - ry_i) \qquad (8)$$

$$\frac{dx_i}{ds} = \frac{1}{f}[x_i\sum_{i=1}^{n} q_i(x_i - ry_i) - q_i(x_i - ry_i)] \qquad (9)$$

Dividing Eq. (9) to Eq. (8) yields to (Shindo, et al., 1985):

$$\frac{dx_i}{df} = \frac{1}{f\sum_{i=1}^{n} q_i(x_i - ry_i)}[x_i\sum_{i=1}^{n} q_i(x_i - ry_i) - q_i(x_i - ry_i)] \qquad (10)$$

and from Eq. (8):

$$\frac{ds}{df} = \frac{-1}{f\sum_{i=1}^{n} q_i(x_i - ry_i)} \qquad (11)$$
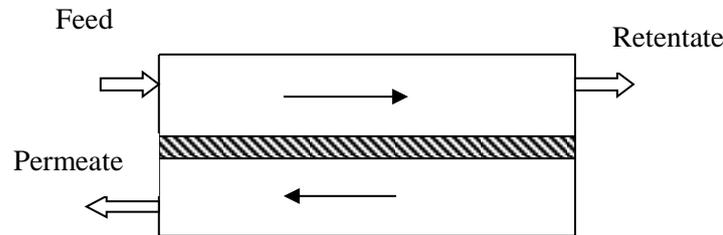

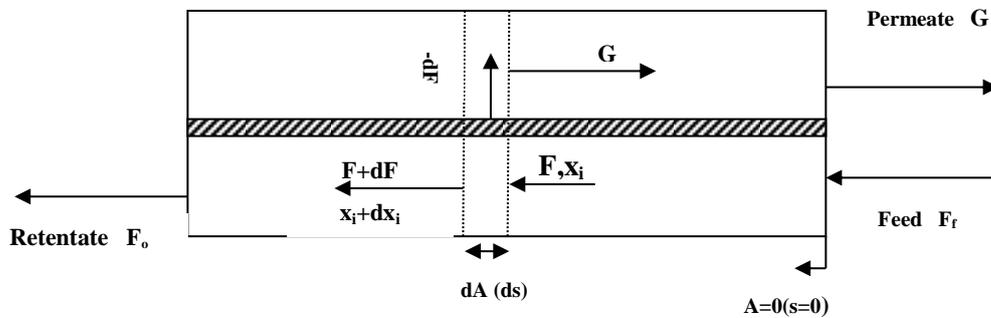**Figure 1.** The countercurrent flow schematic diagram

Figure 2. The Schematic of differential element in a single stage countercurrent permeator and the conditions at two ends

Eqs. (10) and (11) along with the following boundary conditions are solved in this work to obtain the dimensionless membrane area and permeate compositions

$$
\begin{cases}
s=0 \begin{cases} f=1 \\ x_i = x_{if} \\ y_i = y_{ip} \end{cases} \\[2em]
s=s_t \begin{cases} f=1-\theta \\ x = x_{io} \\ yi = y_i' \end{cases}
\end{cases}
\tag{12}
$$

where, y_i values in the Eqs. (10) and (11) should be obtained properly. This is achieved by integrating Eqs. (1) and (2) from an arbitrary point to feed exit point.

Eqs. (13) and (14) are obtained for the flow rate of permeate stream and composition of permeate (y_i), respectively.

$$g = f - (1-\theta) \tag{13}$$

$$y_i = \frac{fx_i - x_{oi}(1-\theta)}{f - (1-\theta)} \tag{14}$$

where, x_{oi} is the composition of components in retentate stream. Eq. (14) is in turn, represents the bulk composition of permeate stream which is assumed to be identical to its local values (the values at the membrane surface in contact with permeate stream). At the fiber closed end ($A=A_t, g=0$) the value of denominator is equal to zero ($f=1-\theta$). Therefore, the bulk values are substituted by the local ones at this point. Obviously, the bulk and local compositions are the same at this point of the fiber. The local values of y_i ($=y_i'$) are calculated by Eq. (15). This equation indicates that the mole fraction of each component is equal to its permeation flux divided by the total permeation flux. Therefore, the values of y_i in Eqs. (10) and (11) are obtained from Eq. (14) expect at the fiber closed end which its local values are computed based on the following procedure:

$$y_i = y_i' = \frac{\dfrac{Q_i}{\delta}(p_f x_i - p_p y_i)}{\displaystyle\sum_{i=1}^{n} \dfrac{Q_i}{\delta}(p_f x_i - p_p y_i)}, g=0(f=1-\theta) \tag{15}$$

Applying Eq. (15) for the base component gives:

$$y_m = \frac{\dfrac{Q_m}{\delta}(p_f x_m - p_p y_m)}{\displaystyle\sum_{i=1}^{n} \dfrac{Q_i}{\delta}(p_f x_i - p_p y_i)} \tag{16}$$

By dividing Eq. (15) to Eq. (16) and some simplification, the following equation is obtained:

$$y_i = \frac{\dfrac{Q_i}{Q_m}x_i}{r\left[\dfrac{Q_i}{Q_m}-1\right]+\dfrac{x_m}{y_m}} \tag{17}$$

The sum of mole fraction of components is equal to unity, namely:

$$\sum_{i=1}^{n} y_i = 1 \tag{18}$$

By substituting Eq. (17) into Eq. (18) the following equation is obtained for calculating y_m:

$$\sum_{i=1}^{n} \frac{\dfrac{Q_i}{Q_m}x_i}{r[\dfrac{Q_i}{Q_m}-1]+\dfrac{x_m}{y_m}} - 1 = 0 \tag{19}$$

Eq. (19) can be solved by the Newton's iterative procedure (Appendix A) and the value of y for the other components is calculated by Eq. (17). Here, the value of x_i and x_m should be specified which are equal to x_{oi} and x_{om}, respectively. The procedure for calculating the value of x_o is explained later.

In order to solve the set of differential equations the forth order Runge-Kutta method was employed. First, it is necessary to guess the values of retentate composition (x_{oi}) and dimensionless membrane area (s_t). It should be noted that, the first guess for dimensionless area is not important in the solution procedure of this model and the obtained solution is not sensitive to the initial guess for this parameter. The procedure of solving the governing equations is summarized in a simple flow chart shown in Figure 3.

In order to reduce the computation time, an appropriate method is applied here for proper initial estimation of x_{oi} values using a mass balance equation based on perfect mixing assumption .Moreover, a simple procedure is applied to provide new x_{oi} values based on the result of previous ones. The comparison of CPU time with that of reported by McCandless (1990)revealed that the present method caused a decrease in computation time. The required computation time is about a few minutes and the number of iterations is of order of 10. The authors believe that   the rapid convergence of the algorithm is due to the developing a procedure for constructing the appropriate initial values. It seems that the selection of initial values has a great effect on the speed of convergence of the algorithm. On the other hand, the model results are correct and have good accuracy.
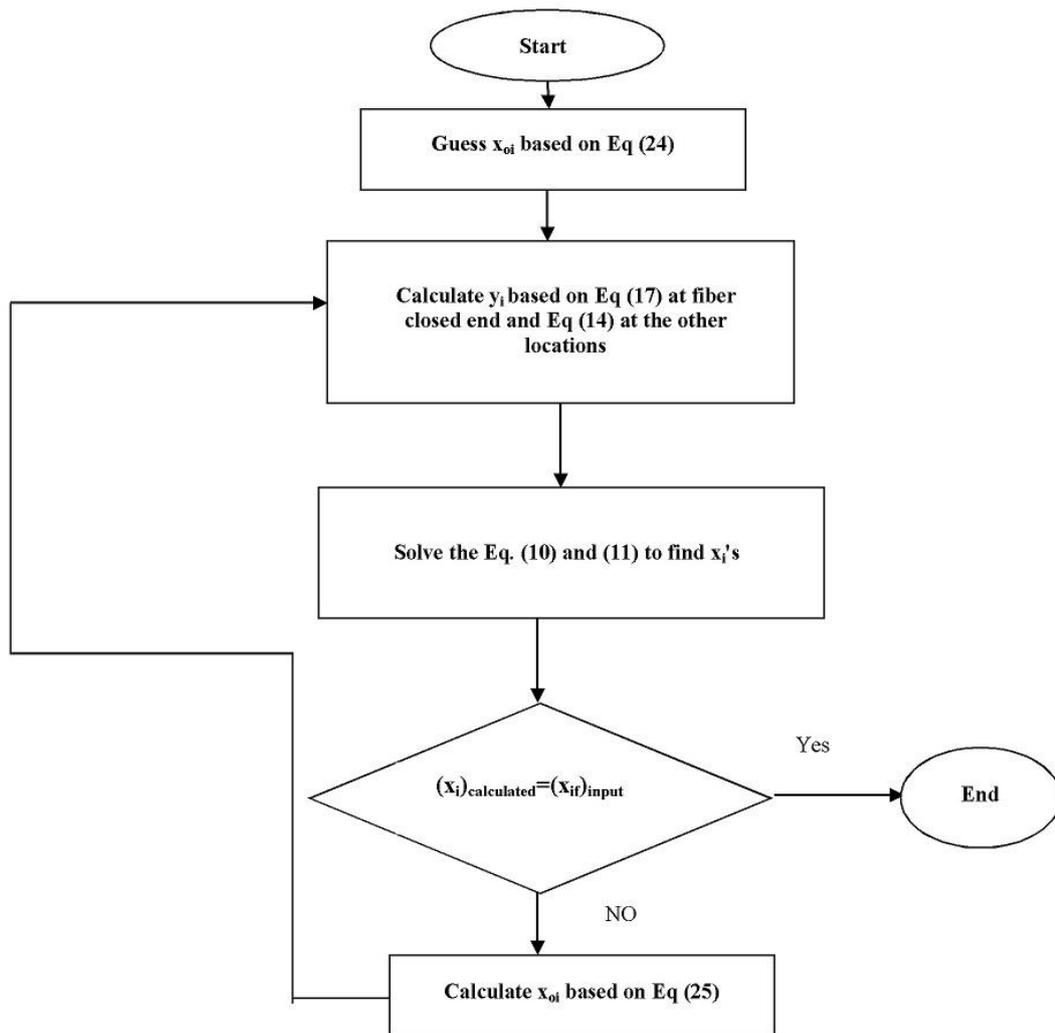
Figure 3. The flow chart of procedure of solving the model equations

The procedure to make initial guess for x_{oi} and the correction of guesses is based on the following equations. It should be noted that the equations are expressed based on dimensionless parameters which are introduced before.

Eq. (20) represents the mass balance for component i based on the perfect mixing assumption.

$$x_{if} - (1-\theta)x_{oi} = q_i s_t (x_{if} - ry_{ip}) \tag{20}$$

Solving this equation for x_{oi} yields to:

$$x_{oi} = \frac{x_{if} - q_i s_t (x_{if} - ry_{ip})}{(1-\theta)} \tag{21}$$

The following equation is valid for any flow pattern:

$$x_{if} - (1-\theta)x_{oi} = \theta y_{ip} \tag{22}$$

where, the terms on the left side of Eq. (20) and (22) are equal, so the value of yip can be expressed as follows:

$$y_{ip} = \frac{q_i s_t x_{if}}{(rq_i s_t + \theta)} \tag{23}$$

By substituting y_{ip} from Eq. (23) into Eq. (21), Eq. (24) is obtained for x_{oi}:

$$x_{oi} = \frac{q_i s_t x_{if} (r-\theta) + \theta x_{if}}{(1-\theta)(rq_i s_t + \theta)} \tag{24}$$

where, the above equation is used for calculating the initial value of x_{oi}'s. The new value of x_{oi}'s is obtained based on the following simple equation:

$$(x_{oi})_{new} = \frac{x_{if}}{(x_i)_{calculated}} (x_{oi})_{old} \tag{25}$$

where, (x_i) calculated is the value of x_i's predicted by the model through solving Eq. (10).

3. Results and Discussions

3.1 . Model Validation

In order to validate the results of the developed model, the experimental data of Kaldis, et al. (2000) was used. In this study, the permeation a multicomponent gas mixture through a membrane permeator was modeled and the model equations were solved by an orthogonal method. The predictions of the mathematical model was compared with experimental results for the separation and recovery of hydrogen from a typical gas oil desulfurization unit for various feed pressures, temperatures and stage cuts. They applied their model to various refinery gas streams of interest as well and the results revealed that high permeate purity (99.95+), and high recovery (0.6–0.9), could be achieved even in a one-stage membrane unit.

Their experiments were carried out by membrane module contained UBE industries polyimide hollow fibers with 400 μm outer and 200 μm inner diameters and with an effective membrane area of 10 cm². Feed stream was contained 67.5% H_2, 16.7% CH_4, 4.3% C_2H_6 and 11.5% CO_2 with flow rates of 5 to 30 Nl/h. The feed gas enters the shell side at high pressure and flows inside the fibers in a counter current mode to the permeate flow. The permeate pressure was kept at 1 bar in all experiments but the feed pressure varied between 5-20 bars. The value of permeability of components has been reported in Table 1. These values are experimental values which have been reported in their work. Kaldis, et al. (2000)conducted their experiments at different temperatures (20-80ºC) and they investigated the effect of temperature on permeate and retentate compositions. The effect of temperature was not studied in this work and the experimental data from (Kaldis, et al., 2000)at 40ºC was selected to validate the predictions of the model which is developed in this study.

Figures. 4-6 show that the model predictions (the solid lines) are in a good agreement with the experimental data. The equations of this model are expressed as dimensionless ones, therefore the calculated values for permeate or retentate mole fractions are independent of feed flow rates. Kaldis, et al. (2000)was considered the permeate pressure drop in developing their model development. As mentioned before, this factor is neglected in the present study. At low permeate (feed) flow rates and high pressures of feed the effect of permeate pressure drop usually can be ignored. This is the reason why the predictions of the model developed in this study are in a agreement with the experimental data.

The results of the error analysis in predicting the component mole fractions are presented in Table 2. The values of error are reported based on the following equation:

Absolute average percent error =

$$\frac{1}{n}\sum_{i=1}^{n}\frac{\left|y_{i\,predicted}-y_{i\,experimental}\right|}{y_{i\,experimental}}\times 100 \qquad (26)$$

3.2. Application of the Model: a Case Study

After the model is validated, it is applied to the hydrogen recycle stream of Pacol unit of Linear Alkyl Benzene (LAB) production process in Iran.

LAB is the most common raw material used in household detergents production. One of the technologies in producing LAB is conversion of linear paraffins into olefins, followed by alkylatiation of benzene in presence of a catalyst.

The Pacol Process is a fixed bed catalytic process designed to selectively dehydrogenate a high purity, normal paraffin feed to the corresponding mono-olefin product(*Iran Chemical Industries Investment CO. (I.C.I.I.C) Esfahan - Iran, LAB complex, unit operating manual, Pacol unit.*).The schematic diagram of this process is illustrated in Figure 7.

The developed model is applied to study the feasibility of using a membrane separation process for purification of recycled hydrogen stream from a Pacol unit. The required data are taken from a LAB production unit. The recycle stream contains 94% H_2, 1% CH_4, 2.5% C_2H_6, 1.4% C_3H_8, 0.6% n-C_4H_{10}, 0.3% n-C_5H_{12} and 0.2% C_6+ at temperature of 90º C and pressure of 2.5 barg. The membrane is considered to be a hollow fiber membrane module. (polyimide membrane, Ube industries) which the value of permeabilities for components is reported in Table3.
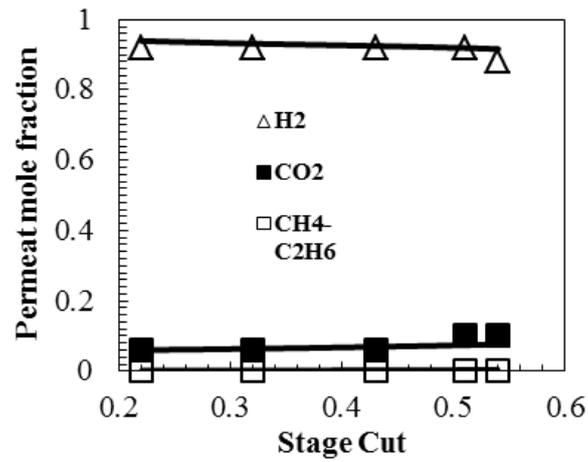
Figure 4. The effect of stage cut on permeate composition, p$_f$=20bar, points: experimental [6], solid lines: modeling
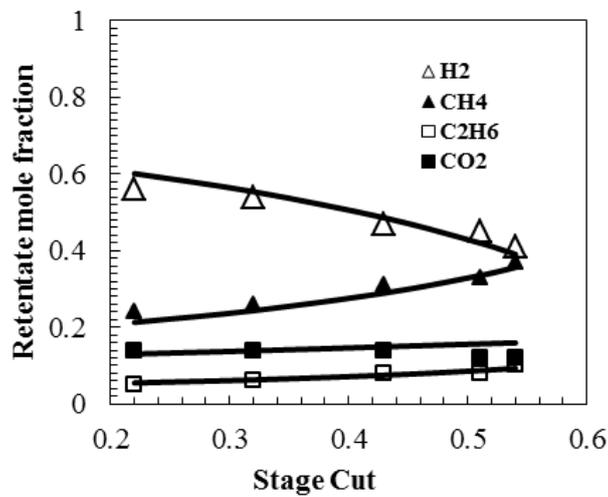

Figure 5. The effect of stage cut on retentate composition, p$_f$=20bar, points: experimental [6], solid lines: modeling
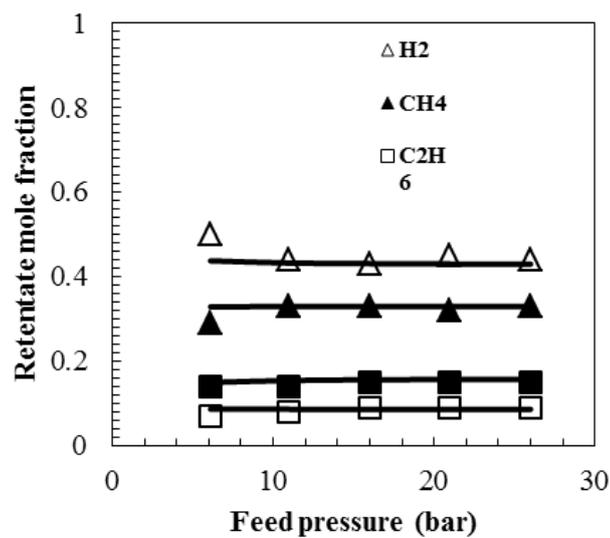

Figure 6. The effect of feed pressure on retentate composition, θ=0.5, points: experimental [6], solid lines: modeling

The feed gas is considered to enter the shell of module and flows inside the fibers in a counter current pattern to the permeate flow. The feed flow rate is 6591.9 Nm³/h. The data was available at a fixed operation (feed) pressure, so the effect of this parameter was not considered. Instead, the permeate pressure is varied between 0.35-1.23 bar in order to investigate the effect of pressure on permeate and retentate streams. The effect of stage cut on composition of these two streams was also investigated.

The effects of stage cut on the permeate and retentate compositions are presented in Figures 8 and 9, respectively. Figure 8 shows that the mole fraction of hydrogen decreases from 90 to 50% with an increase in the stage cut, while the mole fraction of the other components increases slightly. For a given feed flow rate it is obvious that as stage cut (flow rate of permeate stream) increases, the retentate flow rate must be decreased. Besides, the mole fractions of less permeable components increase in this stream.Scince., the membrane is more selective to hydrogen. the mole fraction of hydrogen decreases in retentate stream.

From Figure 9 it is revealed that the permeate stream contains mainly H2 and the amount of hydrocarbons in this stream is significantly low.

Feed stream contains significant amount of hydrogen, the purity of this component is quiet high and relatively insensitive to stage cut. Permeation of gaseous species in glassy polymers (such as polyimides) strongly depends on the diffusion coefficient of these species through the polymer. The diffusion coefficients in glassy materials decrease with increasing molecular size of permeant. Moreover, the sorption or solubility selectivity of a component is an important factor in permeation rate of a component through the polymer material. The sorbtion coefficient of is a measure of energy required for a gas or vapor to be sorbed by the polymer. This coefficient increases with increasing condensability of gas or vapor. Large molecules are normally more condensable than smaller ones. Therefore, larger molecules have larger sorbtion coefficient. Sorbtion selectivity favors larger and more condensable molecules, such as hydrocarbon vapors over permanent gases. However, in glassy polymers the mobility of a component is dominant and permeability decrease with increasing molecular size of permeant. Therefore, the permeation rate of smaller molecules is more than that of larger ones(Baker, 2000).

Hydrogen molecular size is much smaller than that of hydrocarbon (especially C_2-C_6); therefore, it is the most permeable component through the membrane. Consequently, the amount of hydrogen in permeate stream is significantly higher than the other components.

The mole fractions of hydrocarbon components are very close to each other, so these values fall into one line in Figure 9. Hydrogen recovery is about 91% at stage cut of 0.86. This is defined as the ratio of mole of hydrogen in permeate stream to that of in feed stream. The value of total membrane area required for this recovery with a membrane thickness of 200 μm, calculated through Eq. (3) is 3.1×10^6 m².

The variation of dimensionless membrane area with stage cut is presented in Figure 10. It is obvious that the membrane area required for a given separation efficiency increases at higher stage cuts (higher permeate flow rates).

Figures. 11-13 show the effect of permeate pressure on the mole fraction of components in retentate and permeate streams and on dimensionless area, respectively. Figures 11 and 12 show that the permeate and retentate mole fractions are almost unaffected by permeate pressure. The permeance of hydrogen is independent of pressure(Kaldis, et al., 2000) .On the other hand, as mentioned before this component is the most permeable component through the membrane. Therefore, at the specified recovery (91%) the hydrogen concentration remains high and almost unaffected by the permeate pressure. It is obvious that, at a constant stage cut the hydrogen mole fraction must be constant in retantate stream. The permeance of hydrocarbons varies with pressure(Kaldis, et al., 2000). Because, their permeability through membrane is very low, the hydrocarbons concentration in permeate stream is not significant. The amount of these components in feed stream is also negligible. All of this factors lead to an insignificant pressure dependence of their mole fractions in permeate and retentate streams. As said before, the mole fractions of hydrocarbon components are very close to each other, so these values fall into one line in Figure 12. Membrane area required for a specified recovery increases with increasing in permeate pressure. In fact, as permeate pressure is increased the driving force for transport of components decreases. Therefore, at specified recovery or stage cut the membrane area has to be increased.

Table 1. Permeability of components in polyimide membrane
(Kaldis, et al., 2000)

Component	Permeability (cm³(STP)/s.cm².cm Hg)[a]
H_2	$2.9×10^{-4}$
CH_4	$0.037×10^{-4}$
C_2H_6	$0.0064×10^{-4}$
CO_2	$0.93×10^{-4}$

[a] measured at a pressure of 10 bar for H2 and CH4 and 2 bar for CO2 and C2H6; temperature was 40ºC for all gases.

Table 2. The results of error analysis

Component	Absolute average percent error
H_2	3.4
CH_4	13.1
C_2H_6	36.5
CO_2	10.4

Table 3. Polyimide membrane Permeabilities (barrer)a, (Ube industries) [a], 60º C
(Baker, 2000)

Component	Permeability
H_2	50
CH_4	0.4
C_2H_6	0.08
C_3H_8	0.015
C_4H_{10}	-
n-C_5H_{12}	-
C_6+	-

[a] 1 barrer $=10^{\wedge-10}$ cm^3 (STP). cm/cm^2.s.cmHg
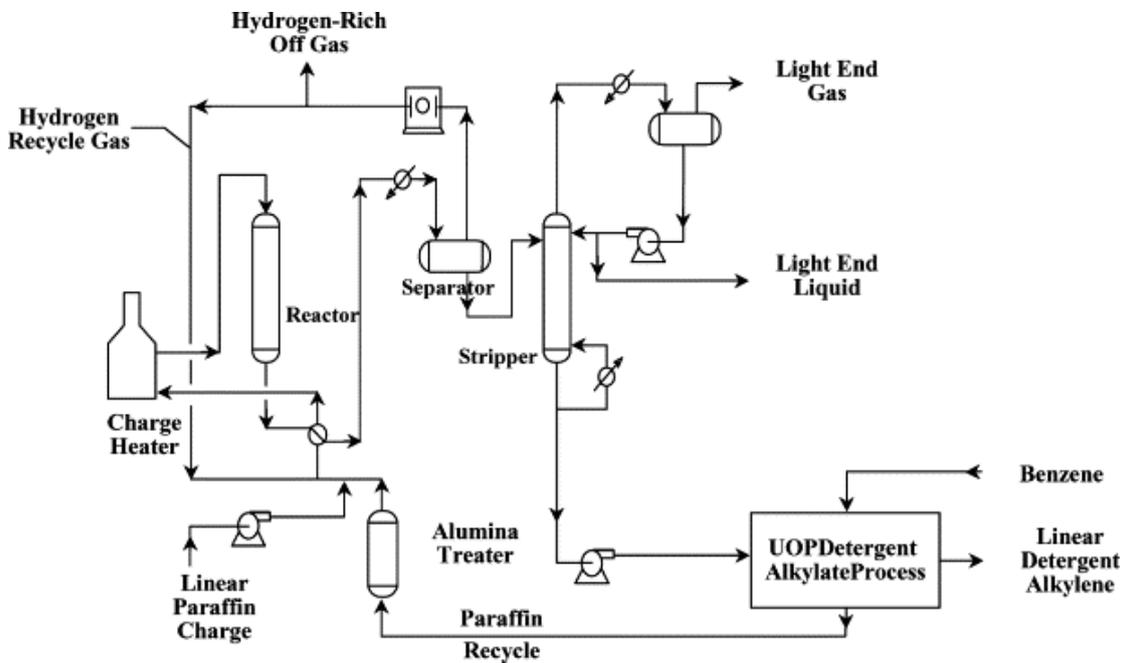

Figure 7. The pacol process schematic diagram
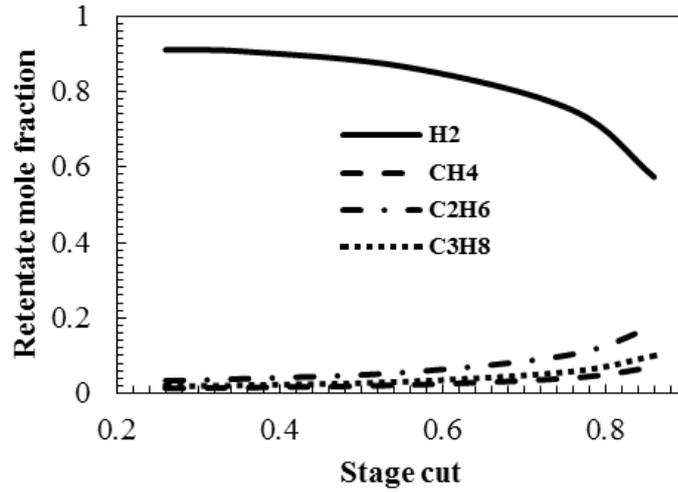
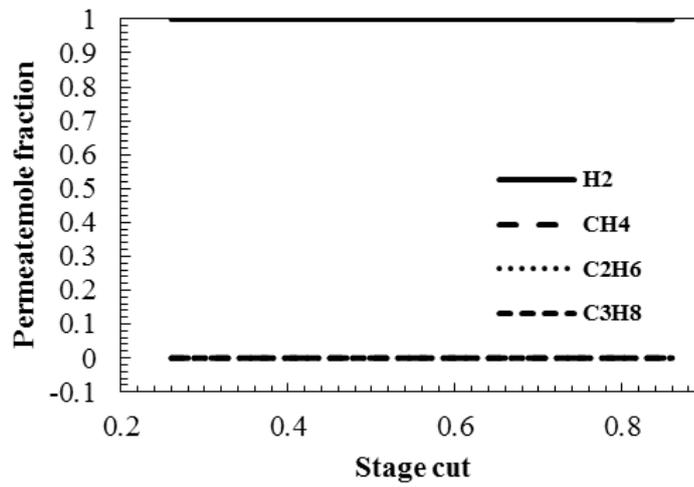**Figure 8.** The effect of satge cut on retentate composition (r = 0.1)

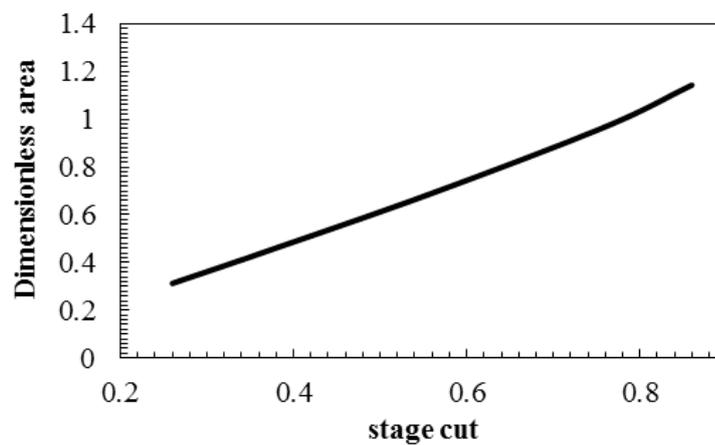**Figure 9**. The effect of satge cut on permeate composition (r = 0.1)

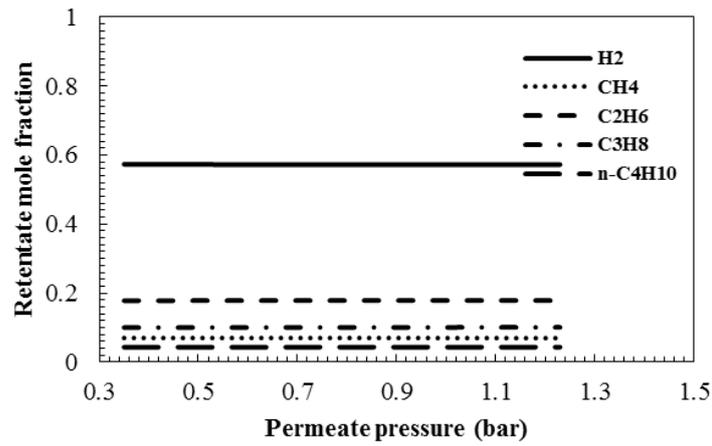**Figure 10**. The effect of stage cut on dimensionless area (r = 0.1)

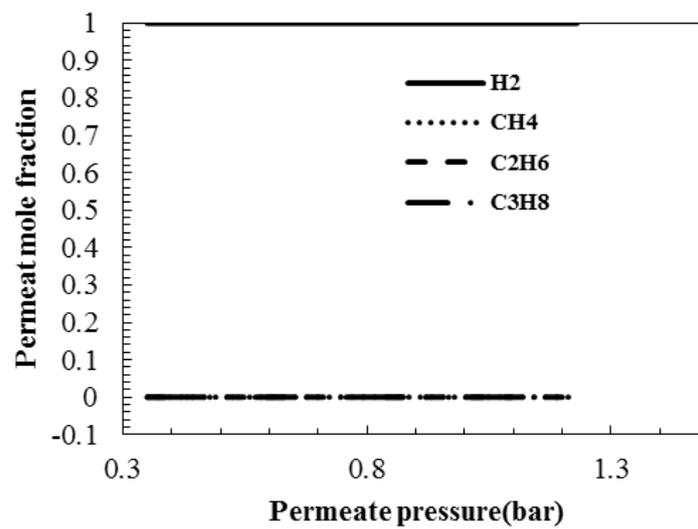**Figure 11**. The effect of permeate pressure on retentate composition ($\theta = 0.86$)

Figure 12. The effect of permeate pressure on permeate composition ($\theta = 0.86$)
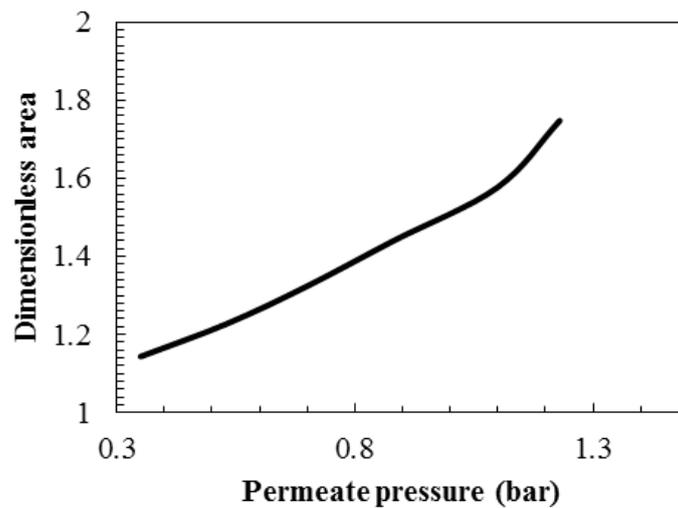

Figure 13 . The effect of permeate pressure on dimensionless area ($\theta = 0.86$)

4. Conclusion

At present study an algorithm was developed for modeling of multicomponent gas separation in a countercurrent membrane permeator. Using the mass balance for component $_i$ based on the perfect mixing assumption, a method is developed to construct proper initial guesses for reject stream mole fractions. Also, a simple procedure is introduced for correcting the guesses. The required CPU time is about a few minutes, and the number of iterations is of order of 10. The model's predictions are compared with the experimental data and a good agreement is achieved. After model validation, it is applied to investigate the feasibility of membrane process for hydrogen separation in a Pacol process. It is revealed that a high hydrogen purity and recovery could be achieved in the membrane permeator (hydrogen recovery is about 91% at stage cut of 0.86), while the required area increases at higher stage cut or higher permeate pressure. support of NIGC for this research.

Nomenclature

A	Membrane area,cm^2
F	Flow rate in feed side,cm^3 (STP)/s
f	Dimensionless feed flow rate,-
G	Flow rate in permeat side,cm^3 (STP)/s
g	Dimensionless permeate flow rate,-
p	Pressure,cmHg
Q	Permeability,Barrer
Q_m	Permeability of the most permeable component,Barrer
r	Pressure ratio,-
s	Dimensionless membrane area,-
x	Mole fraction in feed side,-
y	Mole fraction in permeate side,-

Greek symbols

δ	Membrane thickness, m
θ	Stage cut,-

Subscripts:

f	Feed
i	Feed component
o	Reject stream
p	permeat

Appendix A

A function is defined as shown in Eq. (A-1) and the differentiation of this function is obtained respect to y_m based on Eq. (A-2)

$$f(y_m) = \sum_{i=1}^{n} \frac{\frac{Q_i}{Q_m} x_i}{r[\frac{Q_i}{Q_m} - 1] + \frac{x_m}{y_m}} - 1 \tag{A-1}$$

$$f'(y_m) = \sum_{i=1}^{n} \frac{(\frac{Q_i}{Q_m} x_i)(\frac{x_m}{y_m^2})}{[r[\frac{Q_i}{Q_m} - 1] + \frac{x_m}{y_m}]^2} \tag{A-2}$$

The value of y_m is calculated by following equation:

$$y_m^{(r+1)} = y_m^r - \frac{f(y_{r_m})}{f'(y_{r_m})} \tag{A-3}$$

where, r is the iteration number. The procedure is repeated until the value of y_m no longer changes.

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Graphical Abstract

A model is developed for separation of multicomponent gas mixtures in a countercurrent hollow fiber membrane module. A simple procedure is introduced for correcting the guesses where the CPU time is decreased essentially. The model is adopted in purification process of hydrogen in Pacol unit of linear alkyl benzene (LAB) production process in Iran. A high hydrogen purity and recovery could be achieved in the membrane permeate, while the required area increases at higher stage cut or higher permeate pressure.