Mathematical Modeling of Helium Recovery from a Multicomponent Fuel Gas with Polymeric Membrane

Muhammad Ahsan and Arshad Hussain

***Abstract*—A mathematical modeling is performed to recover helium from fuel gas using polymeric membrane. This study implements a numerical model used in gas separation for hollow fiber membrane modules. Helium recovery from fuel gas is a process of great importance, but there is not yet simple and fast model applied to recover helium from multicomponent gas mixture. The numerical technique presented in this study shows reliable investigation of helium permeation with minimal effort in a membrane module. The presented model has been validated against available data from the literature. The results obtained by using presented model show good similarity with literature data. The model is extended to multistage membrane separation with different variation in permeate pressure. To our knowledge, this is first dependable numerical study for the recovery of helium from multicomponent gas mixture using countercurrent flow pattern and multistage membrane permeation. Operating conditions and membrane system structures are easy to optimize using this model due to its simplicity and algebraic nature.**

***Index Terms*—Mathematical modeling, helium recovery, countercurrent flow, multistage permeation.**

I. Introduction

Membrane gas separation has been used progressively in the process industry during the last three decades. Development of the asymmetric membranes is the main this, which combines high selectivity and high permeability. Fritzsche and Narayan [1] given a short review on the development of membrane technology. The main competing separation processes for membrane gas separation are cryogenic distillation and physical adsorption. High recovery and high purity products are obtained by these two separation processes. For large scale separation processes cryogenic distillation is more effective and efficient. Membrane separation processes normally give products of intermediate purity up to 96%. Beside this numerous studies have shown that for low to medium level production membrane separation can be a good alternate [2], [3]. Membrane systems are preferred in offshore installations due to their less area requirements and low weight. For economical processes can be designed by combining membrane process with cryogenic distillation or physical adsorption [4]. The main objective of chemical processes is to decrease the number of possible process alternatives to a number appropriate for mathematical modeling and simulation. At this stage it is not needed to do mathematical modeling and simulation of all possible processes choice. It is essential to rely on that mathematical model which is fast, accurate and requires less computation effort. Such model also required less information as compared to complicated model, in which generally detailed information is not available. At this level it is also vital to use such mathematical model which is also appropriate for optimization purposes. In this way a user can obtain realistic results speedily by using different operating conditions and structures. Limited reliable mathematical models for membrane gas separation are available in literature. A spiral wound membrane modules design model was given by Hogsett *et al.* [5]-[7]. Some model has a tendency to overestimate the module performance and is also stated to less accurate for certain cases of stage cut and area [8]. Approximate analytic solutions have been derived by several researchers for binary mixtures and counter-current permeators.

This model is testified to be very sensitive to initial estimates and to inaccurate from convergence problems [9]. An approximate solution, effective for binary mixtures have been derived in a research paper by Krowidi *et al.* [10]. Their model is equated with the series solution [7] and shows to provide improved results than the series solution. But, the model suggested by Krowidi *et al.* suffers from the same difficulties as the two prior models. It is limited to binary mixtures and the model equations are highly non-linear and complex. The mathematical model for the boundary value problem is complex and required substantial computational efforts. Furthermore, concentration profiles and prior initial guesses of the pressure along the fiber length are needed. Subsequently the membrane modules available commercially have a preset length of fibers; in such case this mathematical model leads to significant mathematical complications because of the trial-and-errors involved. To overwhelm the numerical difficulties in solving the boundary value problems, numerous changes and different numerical methods have been suggested to solve the model equations [11]-[13]. These models purpose in introducing additional assumptions and simplifying the governing equations that leads to estimated solutions with less computational time and effort. In the case of binary gas mixtures the governing equations are solved by asymptotic solutions in a cross flow approach depended on the Navier–Stokes equations. But, these methods do not actually ease the numerical problems but somewhat at low permeabilities the results look to be inaccurate [14].

A mathematical model was developed for multicomponent gas separation, which needs previous information of dispersion and diffusion coefficients in the fluid phase and mass transfer coefficient in the porous substrate [15], [16]. These factors are unluckily not normally obtainable, and the

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structure of membranes can barely be defined precisely. A model was developed by Chowdhury *et al.* [13] established on Pan's formulation [17], and the numerical solution was expressed as an initial value problem. But, the overall material balance for co-current flow was used as an approximation for that of countercurrent flow for easy integration into AspenPlus. Researchers implemented a numerical model for the solution of permeation problems using diverse discretization methods, in the environment of finite difference model [18], [19]. Makaruk and Harasek [20] presented a model for multicomponent permeation established on the finite difference Gauß-Seidel method, and the solution was stabilized by adjusting a relaxation factor in case of problems with convergence. Katoh *et al.* [21] developed a tanks-in-series model to consider non-ideal mixing flows and an applied relaxation model as a steady computational procedure to solve the governing differential equations. The numerical technique of is a combination of improved Powell hybrid algorithms, the L'Hospital's rule, the Gear's BDF method, and secant method. The equations are simplified in such a way that the Gear's BDF method alone is suitable for solving the equations [22]. A numerical model for high-flux, asymmetric hollow fiber membrane was proposed, which is developed on Pan's original formulation [23], but the governing equations were simplified following the method of Sengupta and Sirkar [24].

In this research work, a mathematical model is implemented for the recovery of helium from fuel gas, which is based on Pettersen's formulation [25], the initial guesses are estimated by using Paterson approximation [26] instead of logarithm mean.

The mathematical model is implemented to countercurrent flow configuration with various operating conditions and found very accurate for helium recovery from multicomponent gas mixtures. The model results are validated with data available in literature and its accuracy has been show. The mathematical model is used to study dynamic performance of different membrane module configurations for helium enrichment and recovery from fuel gas. Several operational modes for counter-current flow pattern are also studied to increase the helium enrichment and recovery in permeate stream.

II. Mathematical Modelling

The objective of the research described here has been to formulate and implement a fast and simple mathematical model for gas separation by membrane separation. This part shows the formulation of the mathematical model and the assumptions used in it. In solution-diffusion theory for gas separating membranes [27], in a non-porous membrane Fick's law can be used to express local permeation of a gas The membrane area needed for a specific separation value can be calculated on several stages of accuracy. A fugacity coefficient was used to explain the non-ideal behavior in the gas phase [28]-[30]. The following model dependent on differential equations was presented for the separation of a binary mixture in a hollow fiber module [8]:

$$dn_i = \frac{\boldsymbol{P_i} dA(p_{f,i} - p_{p,i})}{d} \quad (1)$$

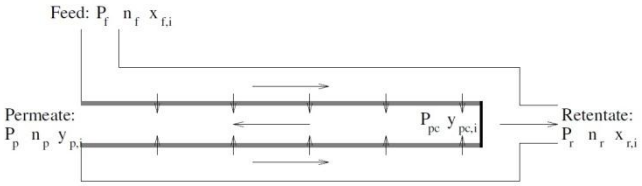

Fig. 1. Schematic of hollow fiber membrane module.

$$\frac{d\bar{F}}{d\bar{l}} = -R\{(x - \delta y) + s[(1 - x) - \delta(1 - y)]\}; \quad (2)$$

$$\bar{F}\frac{dx}{d\bar{l}} = -R\{(1 - x)(x - \delta y) - sx[(1 - x) - \delta(1 - y)]\}; \quad (3)$$

The dimensionless membrane area, *R* is expressed as

$$R = \frac{AP_f Q_1}{n_f} \quad (4)$$

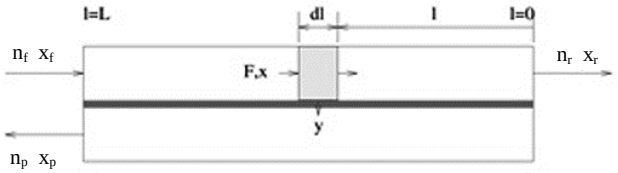

Fig. 2. Cross section element for the mass and material balance.

This model is dependent on the suppositions of trivial axial diffusion in permeate and feed side, minor pressure losses at the feed side and plug flow at permeate and feed side. Membrane permeation in rate governed process equilibrium is never reached. Rate governed methods perhaps normally be defined by associations between a flux and a driving force. In a heat exchanger, which is also a case of a rate-governed chemical engineering unit operation, the integral relation between driving force and flux and is defined by the renowned equation

$$Q = UA\Delta T_{lm} \quad (5)$$

An integral relation among differences in partial pressure and flux across the membrane can be expressed as:

$$\int dn_i = Q_i A \Delta P_{lm,i} \quad (6)$$

Here Q_i is the permeation constant for component *i*, [flow rate/ (area. pressure)] and $P_{lm,i}$ is expressed by equation (7), considering counter-current flow in the module

$$\Delta P_{lm,i} = \frac{(p_{f,i} - p_{p,i}) - (p_{r,i} - p_{pc,i})}{\ln\frac{(p_{f,i} - p_{p,i})}{(p_{r,i} - p_{pc,i})}} = \frac{(p_f x_{f,i} - p_p y_{p,i}) - (p_r x_{r,i} - p_{pc} y_{pc,i})}{\ln\frac{(p_f x_{f,i} - p_p y_{p,i})}{(p_r x_{r,i} - p_{pc} y_{pc,i})}} \quad (7)$$

For membrane module the component material balances are shown by equation (8).

$$n_f x_{f,i} = n_P y_{P,i} + n_r x_{r,i} \quad (8)$$

$$\int dn_i = n_P y_{P,i} \quad (9)$$

Assuming ideal gas the partial pressure in the closed end of the hollow fibers is given by

$$p_{pc,i} = P_{pc} y_{pc,i} \quad (10)$$

Mathematical modeling of the pressure drop inside the hollow fibers is a difficult assignment. For the pressure change inside the hollow fibers, a methodology adopted by several researchers is to use a Hagen-Poiseuille equation in differential form to define [29]-[31].

$$P\frac{dP_p}{dl} = \frac{128\mu P_S T}{\pi N D_i^4 T_S} n_p(l) \quad (11)$$

$$P_{pc} = P_p \quad (12)$$

The permeate composition at the closed end of the hollow fibers can be formulated [32]:

$$y_{pc,i} = \frac{1+(\alpha-1)(\delta_r+x_r)}{2\delta_r(\alpha-1)} - \frac{\sqrt{(1+(\alpha-1)(\delta_r+x_r))^2-4\delta_r\alpha(\alpha-1)x_r}}{2\delta_r(\alpha-1)} \quad (13)$$

Established on the reasons published by Weller and Steiner [6], it can be presented that the resulting expression must hold between the components:

$$\frac{y_{pc,i}}{y_{pc,i+1}} = \frac{Q_i(x_{r,i}-\delta_r y_{pc,i})}{Q_{i+1}(x_{r,i+1}-\delta_r y_{pc,i+1})}; i = 1, \dots, nc-1 \quad (14)$$

For the binary case this equation reduces to Giglia's expression. Equations (6), (9) and (12) jointly give

$$n_f\theta y_{p,i} = Q_i A \Delta P_{i,lm}; i = 1, \dots, nc \quad (15)$$

Here $\Delta P_{i,lm}$ defined by equation (7). Introducing the stage cut changes the material balance, equation (8), dimensionless.

$$x_{f,i} = \theta y_{p,i} + (1-\theta)x_{r,i}; i = 1, \dots, nc-1 \quad (16)$$

Lastly the sum of the mole fractions must be equal to one.

$$\sum_{i=1}^{nc} x_{f,i} = 1 \sum_{i=1}^{nc} x_{r,i} = 1 \sum_{i=1}^{nc} y_{p,i} = 1 \sum_{i=1}^{nc} y_{pc,i} = 1 \quad (17)$$

The above mathematical model can be redeveloped by implementing the Paterson approximation to the logarithmic mean [33], with the purpose of reducing 3*nc*+2 algebraic equations to one implicit algebraic equation with one unknown — the stage cut.

III. Results and Discussion

The proposed mathematical model has been applied to the data reported in literature. The operating parameters and module design considered for the comparison and validation of the membrane systems are summarized in Table I [34].

Fig. 3 shows the concentration of helium in permeate stream against stage cut. As the membranes permeate helium selectively, any degree of helium removal from the residue stream may be obtained, by increasing the portion of the total feed stream gas that permeates the membrane. The ratio of total permeate flow to total feed flow is known as the stage cut. The helium losses in the residue stream can be cut to a very low level by operating the membrane separation step at high stage-cut. If the process is run at high stage-cut, substantial amounts of the less permeable gases present in the feed stream will permeate the membrane. In this case, the composition of the permeate stream may be only marginally richer in helium than the feed stream, and it is preferred to return the permeate stream to the feed stream. Fig. 4 describes the dimensionless membrane area required for a specific value of helium in permeate stream. We can observe that case 2 is giving high value of helium in permeation stream as compared to case 1 for the same value of dimensionless membrane area. Fig. 5 shows the recovery of helium in permeate streams at various stage cuts for different cases. We can observe that high recovery of helium is obtained at high stage cut. The recovery value of helium in both cases is overlapping with the value reported in literature at stage cut≈0.2. In both cases 97% or more helium recovery is obtained in permeate stream at 0.35 value of stage cut.

TABLE I: Operating Parameters for Membrane Systems

	Case 1	Case 2
Membrane material and type	Ethyl cellulose hollow fiber	Ethyl cellulose hollow fiber
Flow configuration	Countercurrent, shell side feed	Countercurrent, shell side fee
Feed flow rate (lbmol/h)	721	87
Feed composition	Helium (He): 0.4% Nitrogen(N_2):16.9% Hydrocarbons (C_1-C_{4+}): 82.70%	Helium (He): 2.5% Nitrogen(N_2): 27.7% Hydrocarbons (C_1-C_{4+}): 69.8%
Feed pressure (psia)	365	383
Permeate pressure (psia)	18	18
Temperature (K)	301	322
Selectivities	He/N_2: 4.6 He/CH_4: 1.8 N_2/CH_4:0.4	He/N_2: 4.6 He/CH_4: 1.8 N_2/CH_4:0.4

Fig. 6 illustrates the relationship between the stage cut and dimensionless membrane area. The membrane area required to perform the helium separation increases with the stage-cut. In a specific situation, capital and operating cost considerations may impose an upper limit on stage-cut. Nevertheless, it is envisaged that the process of the membrane gas separation will often be advantageous when run at comparatively high stage-cut, such as at least 0.2 or more.

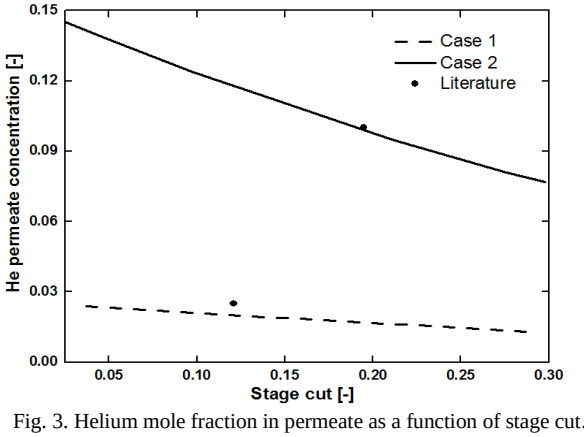

Fig. 3. Helium mole fraction in permeate as a function of stage cut.

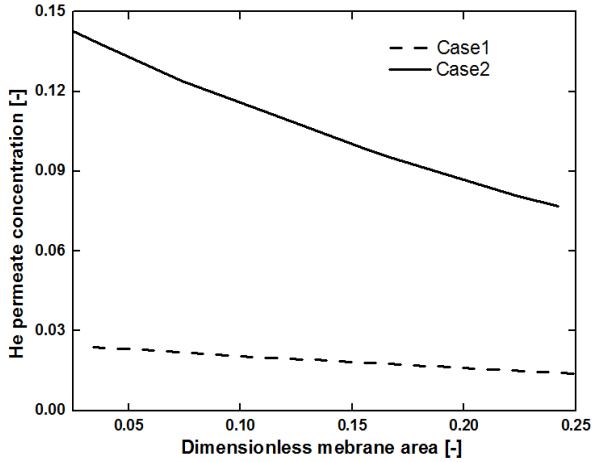

Fig. 4. Helium mole fraction in permeate as a function of dimensionless membrane area.

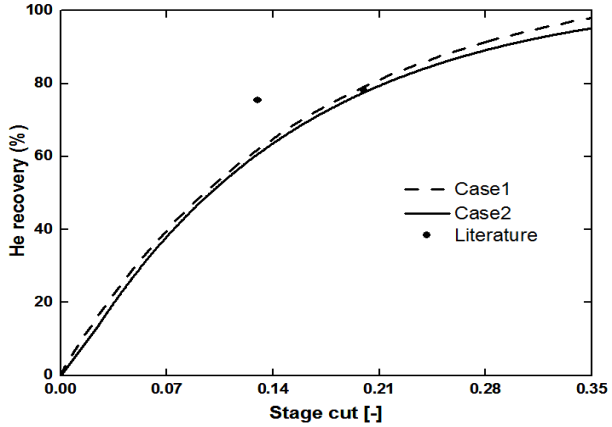

Fig. 5. Helium recovery in permeate against stage cut for different case.

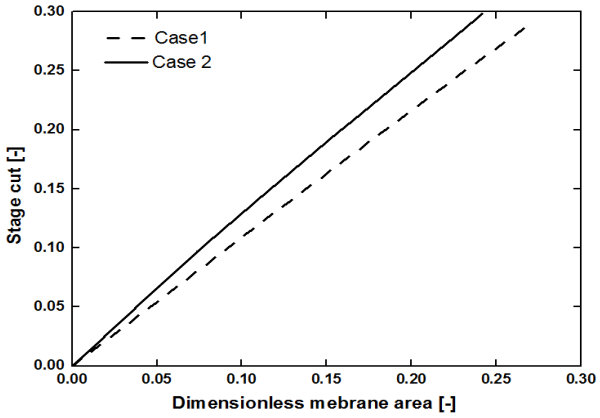

Fig. 6. Stage cut vs. dimensionless membrane area.

Fig. 7 shows two membrane separators in four different operating modes with different permeate pressure configurations. The permeate pressure in the permeator is assumed to be uniform, but different for each permeator (either 18 psia or 25 psia). In Mode A, the feed gas wit lower permeate pressure enters the unit and the residue is treated by the unit with a higher permeate pressure. This is the intermediate operating mode as shown by the permeate concentration and helium recovery shown in the figure. Reverse feed flow is applied to operate mode B, this mode is appeared as most efficient one. In mode C permeator is operated with the same feed and residue concentrations with different feed flow rates, has the least permeation and recovery value of helium. Feed stream with high flow rate entered in low permeate pressure separator. Mode D, operates high flow stream with high permeate pressure separator. Feed stream with high flow rate entered in high permeate pressure separator. This mode is also an intermediate operating mode like mode A. The process of the membrane gas separation can reach significant decreases in helium loss with a single membrane separation stage. More stages or steps may optionally be used as preferred to increase the concentration of helium in permeate or otherwise adjust the composition of permeate or residue streams.

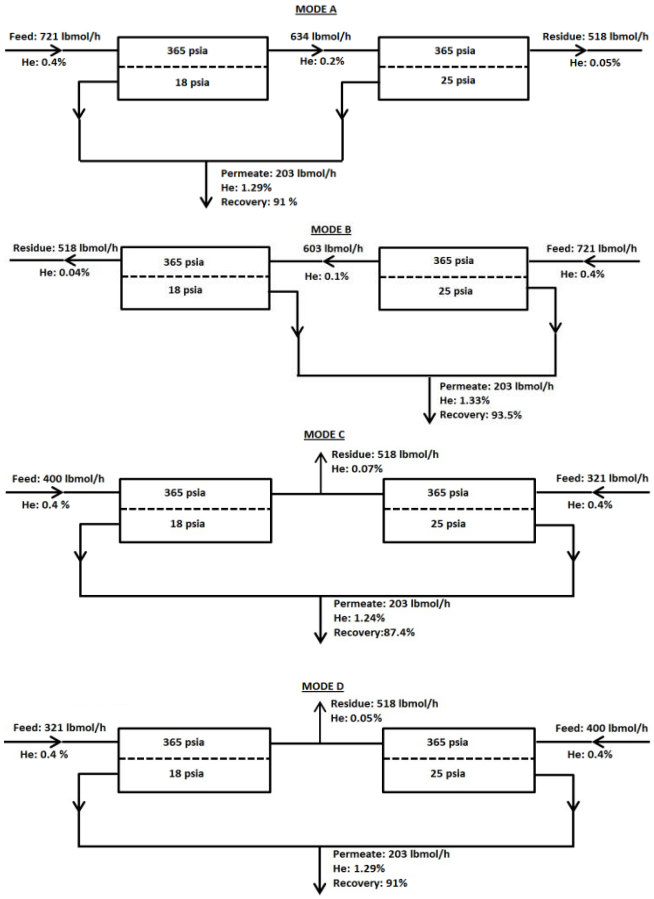

Fig. 7. Effect of permeate-pressure variation on permeation in multistage membrane separator.

IV. Conclusion

The numerical model proposed in this research work has advantages over other models commonly used; it requires least computational effort and time with better solution stability. Additionally, the computational complexity does not increase as the number of separation stages increase. The model predicted the helium separation and recovery from multiple components gas mixture with different flow rates and cases quite adequately. In a comparison of the model and literature data for helium recovery, it was found that the membrane can recover 99.9% helium in the permeate stream at high stage cut. It was also shown that these membranes can be used in recover helium in the permeate stream from the single-stage operation. Further enrichment recovery of helium is possible with multi-stage separation. In helium recovery, the model is used for predicting the membrane gas separation performance and the required dimensionless membrane area. Countercurrent flow pattern is used to predict the permeate mole fraction and the dimensionless membrane area requirement for helium gas separation. In the

development of a practical permeation process it is essential to reduce the membrane area requirements in order to decrease the cost of the process. The countercurrent flow pattern predicted the high permeate mole fraction with minimum membrane area. This flow pattern is used to study the effect of different variables on separation of helium from gas mixture.

These variables included flow rate and composition of the feed, the pressure difference across the membrane and the total membrane area. In helium recovery from fuel gas mathematical model presented here for calculating the performance of separators with the asymmetric membrane has been proved by the available literature data using a membrane separator. The model is applicable to both single and multi-stage separation.

Different modes are compared at different permeation pressure to predict a most efficient operating mode. The two separators in series operation can be used to increase the overall performance for helium recovery, especially when a high concentration of the helium is needed. Mathematical model results are validated for multicomponent gas mixture using available literature data. Comparison of the model and literature data shows that the model results presented in this study can be used for accurate prediction of separation and recovery of helium.

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NOMENCLATURE

A membrane area
D_i inner diameter of the hollow fibers
D_o outer diameter of the hollow fibers
d effective membrane thickness
F local feed rate
$\tilde{F}$ dimensionless local feed rate $\tilde{F} = F/n_f$
N number of hollow fibers pr. module, $N = A/(\pi D_o L)$
n flow rate
nc number of components
L fiber length
l length coordinate
$\tilde{l}$ dimensionless length coordinate, $\tilde{l} = l/L$
P_i permeability constant for component i
$\boldsymbol{P}$ pressure
p_i partial pressure of component i
Q_i overall permeability constant, defined as $Q_i = P_i/d$
R permeation factor defined by equation (4)
T temperature
x_i molar fraction of component i at high pressure side
y_i molar fraction of component i at low pressure side
αi membrane selectivity, defined as $\alpha_i = Q_i/Q_{i=nc}$
δ trans-membrane pressure ratio, defined as $\delta = Pp/Pf$
θ stage cut (cut ratio), defined as $\theta = n_p/n_f$
μ viscosity of gas mixture inside the hollow fibers
g trans-membrane pressure ratio, defined as $g = Pp/Pf$
$\Delta P_{lm,i}$ logarithmic mean partial pressure difference, defined by Equation (7).

Subscripts:

f feed inlet on the high pressure side
r residue outlet on the high pressure side
s reference condition, e.g. Ts = 273K and Ps = 1.013 bar.
p permeate outlet on the low pressure side
pc closed end of the hollow fibre
i component i, $i \in [1, 2, 3, \ldots, nc]$. Component 1 is the fastest permeating component.

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