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Research Article

NUMERICAL ANALYSIS OF A GAS SEPARATION OF CH_4/CO_2 USING HOLLOW FIBER MEMBRANE MODULE

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ABSTRACT

In this research, an approximated technique is proposed for predicting the performance of membrane gas separation using an asymmetric membrane-based gas separator. The permeation behavior of the high-flux asymmetric membrane varies from that of the traditional symmetric membrane. The advanced mathematical model has been applied in this study for the separation of a binary gas mixture. In the present work, a shell-fed hollow fiber module like counter-current flow pattern is modeled mathematically for CO_2 separation from CH_4. Finite Difference method (FDM) is applied to solving the equations numerically. The models offered separation for a membrane module, for given gas conditions, simulating permeate and residue composition and the stage cut. The different parameters are investigating like a change in the pressure ratio, stage cut and feed flow rates. The numerical approach is helpful as it entails the least effort and computational time due to the fact algebraic equations are used instead of differential equations. The obtained model's data also verified with numerical and experimental results available in the literature.
Keywords: Hollow fiber membrane module, counter-current flow pattern, membrane gas separation, non-porous membranes, carbon dioxide, methane.

1. INTRODUCTION

The increase of industries has resulted in rising carbon dioxide concentration in the environment [13]. The environmental concerns have been raised due to a temperature increase in earth's atmosphere. More than fifty percent of carbon dioxide is produced by power industries and also form non-renewable energy sources [23]. Carbon dioxide, which is discharged from petroleum derivatives, flue gasses from refineries and numerous different sources accounts for 80% greenhouse gases released into the atmosphere [10]. The separation of carbon dioxide is based on physical and chemical methods including absorption, adsorption and membrane technology etc. [12; 14]. Therefore, new alternative technologies are proving to be effective for carbon dioxide capture [21; 23]. However, membrane technology is getting more attention and is

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being applied in various industries for large-scale separation of various gas mixtures [22]. The increase in the use of membrane technology can also be attributed to its low-cost operation and it is environmental friendly too.

Membrane-based gas separation is an effective replacement for common unit operations because of its operational ease, financial viability, low upkeep, small length and low energy consumption [1; 7]. A hollow fiber or spiral wound module is used for gas separation through selective nonporous membrane [18]. Membrane gas separation has been used as an alternative process for adsorption and absorption etc. Oxygen recovery, sweetening of natural gas, hydrogen recovery from steam and carbon dioxide removal from methane processes using polymeric membranes are few of the applications of membrane technology [2; 15]. Hollow fiber membrane has numerous advantages like the area to volume ratio, high packing density, gives the desired mechanical strength to membrane module, mechanical stability and resistance from fouling. Hollow fiber membranes are the cheapest membranes which do not require any material as support [3].

The hollow fiber membrane module consists of two geometries i.e., shell-side feed and bore side feed. In the first type, the feed is entered through shell side which consists of fibers in a closed bundle. The high pressure is created for permeation through the shell to fiber side and retentate is collected at the end of shell side. In the second type, a pressurized feed is entered in fiber bore and permeate is collected at one side of the shell. These designs give the large surface area of membrane to be enclosed in a single shell. The fiber diameter is small but shell thickness is large because of large pressure requirement. The internal diameter is in the range of 50 um and the outer diameter is in the range of 100 – 200 um [4; 5].

The one end of fiber bore is open and feed moves around the fiber tubes. The diameter is usually larger than those in shell-side feed system to minimize the pressure drop. These capillary fibers are used in evaporation, ultrafiltration and gas separation. In this module, feed pressure is limited to 150 psig. The diameter of all the fiber is same in bore side feed modules. Small variation can lead to change in the performance of module [4].

The many advantages of hollow fiber membrane include its manufacture by any method to produce a chemical fiber. The highest area to volume ratio makes hollow fibers as cheapest and are easily available [3; 24]. Due to its Compact size, it does not require any support of material. The module is same as tube heat exchanger and countercurrent shell, but fibers are used as a membrane. To enhance the performance of membrane gas separation the investigation of the procedure should be proficient [11]. These targets are achieved by using the modeling and simulations techniques [25]. The issue of scientific demonstrating of membrane gas separation was first tended to by Weller& Steiner. Boucif developed mathematical models that showed a highly nonlinear behavior. They further studied the boundary value problem experienced in a hollow fiber module with permeate flow in the co-current and counter-current flow with no axial pressure drop. Chern built a model for binary gas mixture with an isothermal operation. Developed equations were solved as a boundary value problem [6]. Rautenbach & Dahm developed a model with the constant pressure of permeate along the length. They solved equations numerically for binary gas mixture in both co-current and counter-current manner [20].

In this study, finite difference method (FDM) is used to find the solution of asymmetric membranes for the separation of CO_2 from CH_4. Several flow conditions are investigated in this numerical method. The obtained resulted are compared with the present literature data and series solution. The operating parameters such as feed pressure, permeate pressure, stage cut, mole fractions of permeate and the reject are studied.

2. MATERIALS AND M ETHODS

2.1. Modeling of Counter-Current flow Pattern

The counter-current flow pattern considers a binary gas mixture for the separation through the membrane, where selectivity of a single component is needed. The permeate composition does not differ from the bulk permeate composition because in asymmetric membranes no support material is used, but bulk values are considered as an average of two local permeate values. This model is solved using algebraic equations in MATLAB and Excel to find the solution. Bo Chen reported that counter-current flow pattern gives better results than the co-current pattern for CO_2 recovery [16]. J. E. Perrin and S. A. Stern also concluded that counter-current flow pattern is very efficient for separation with two permeators [19]. Therefore, the counter flow pattern is used for this model.

2.2. Model Assumptions

The finite difference model is based on the following assumptions [2].

- Model is considered as plug flow.
- Assume system is steady state.
- The thickness of the membrane is considered as uniform.
- The permeability of pure component is kept constant [8].
- The pressure effects of the hollow fiber are negligible.
- Concentration gradients in the radial direction have no effect.
- The permeance of the system is constant at given temperature.

2.3. Model Equations

In a counter-current-flow pattern, the permeate gas flows in opposite direction of feed gas, but in the co-current flow patterns, both gases flow in the same direction. The systematic diagram of counter-current model is given below.

The feed enters a unit with a flow rate L_f and gives two flow rates after the separation from the membrane. The permeate flow rate V_p collected in the opposite direction of reject flowrate L_o.

The mass balance of the system is

$$L_f = L_o + V_p \tag{1}$$

The stage cut of the system is

$$\theta = \frac{V_p}{L_f} \tag{2}$$

Selectivity is defined as a ratio of permeability of component A and component B.

$$\alpha = \frac{P'_A}{P'_B} \tag{3}$$

In a finite difference method, the permeate composition y_p can be calculated form this equation

$$y_p = \frac{-b \pm \sqrt{b^2 - 4ac}}{2a} \tag{4}$$

Where a, b and c are the constants that relate the relationship between selectivity α^* and pressure ratio r.

$$a = 1 - \alpha^* \tag{5}$$

$$b = -1 + \alpha^* + \frac{1}{r} + \frac{x_o}{r}(\alpha^* - 1) \tag{6}$$

$$c = \frac{-\alpha * x_o}{r} \tag{7}$$

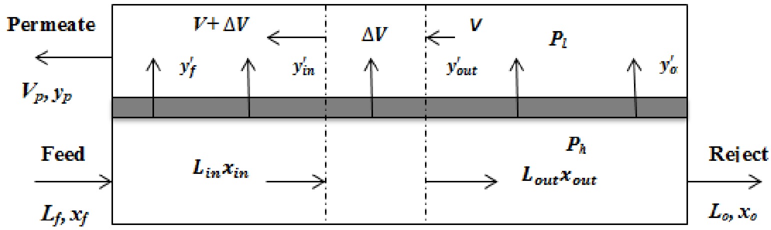

Figure 1. Counter-current flow model

The counter-current model flow diagram is shown in Fig 1. FDM and mass balance is used on both streams with the incremental area ΔA_m.

$$\Delta V = L_{in} - L_{out} \tag{9}$$

Where L_{in} and L_{out} are the flow rates of entering and leaving the system and ΔV is the permeate flow rate.

$$\Delta V y'_{av} = L_{in} x_{in} - L_{out} x_{out} \tag{10}$$

Where $y'_{av} = (y'_{in} + y'_{out})/2$ .Putting L_{out} from equation (9) into (10)

$$\Delta V = \frac{L_{in}(x_{in} - x_{out})}{y'_{av} - x_{out}} \tag{11}$$

Equations (4), (10) and (11) are numerically solved with starts at the feed $x_f = x_{in}$ to find permeate for each specie ΔV and y'_{av} is calculated for each ΔA_m to start x to x_o at the reject side and the bulk composition y_p as a function of x and the starting the calculations at x_o.

The Equation (12), (13) and ΔV are added to obtain the V and y_p is calculated at the feed inlet x_f for each increment ΔA_m.

$$V = \sum \Delta V \tag{12}$$

$$y = \frac{\sum y'_{av} \Delta V}{V} \tag{13}$$

For the counter-current flow area can be calculated as

$$\frac{\Delta V\, y'_{av}}{\Delta A_m} = \frac{\Delta V_A}{\Delta A_m} = \left(\frac{P'_A}{t}\right) * p_h * (x - r'_y) \tag{14}$$

$$r = p_l / p_h$$

Where r is a pressure ratio of feed pressure to the permeate pressure

The average force of driving is

$$(x - r'_y)av = [(x_{in} - r y'_{in}) + (x_{out} - r y'_{out})]/2 \tag{15}$$

To calculate ΔA_m,

$$\Delta A_m = \frac{\Delta V y'_{av}}{\left(\frac{P'_A}{t}\right) \cdot p_h \cdot (x - r y'_y)_{av}} \tag{16}$$

The calculation is started from x_f to obtain the $\sum \Delta A_m$ [9].

2.4. Model Parameters

The parameters of counter-current flow pattern of hollow fiber membrane module are taken from the literature [17].

Table 1. Hollow fiber membrane module configurations

Parameters	Values
Feed Pressure	$3500\ KPa$
Permeate Pressure	$101.325\ KPa$
Permeability of CO_2	3.207×10^{-13} mol/m2.s. Pa
Feed Flow Rate	$50\ mol/s$
Fiber Outer Diameter	$250 \times 10^{-4}\ \mu m$
Fiber inner Diameter	$150 \times 10^{-4}\ \mu m$
Thickness	$125\ \mu m$

3. RESULTS AND DISCUSSIONS

For hollow fiber membrane module, the counter-current flow pattern is used for modeling. The effect of feed pressure, permeate pressure, flow rate and feed content have been investigated and compared with literature results [22].

3.1. Effect of Feed and Permeate Pressure

Fig 2 shows that with an increase in the feed pressure the concentration profile of CO_2 in rejects stream for counter-current flow pattern decreased. With the increase in the feed pressure, the concentration of CO_2 decreased. Due to a continuous increase in the feed pressure, the rate of mass transfer is increased, and then the purity of CO_2 increased in the permeate side. CO_2 values decrease on the reject side because more CO_2 permeates through the membrane. The mole fraction of CH_4 increased on the reject side because mole fraction of CO_2 of the continuous permeated through the membrane. The increase in feed pressure has the best effect for CH_4 because of the increase in the rate of mass transfer more CO_2 permeate. Therefore, the concentration of CH_4 also increased on the reject side.

For counter-current membrane unit design, permeate pressure is a very important parameter. Fig 3 shows that increase in permeate pressure slightly increases the mole fraction of CO_2. The concentration of CO_2 in reject increased due to a reduction in the gradient for counter-current because permeates pressure increased and less CO_2 passed through the membrane. The mole fraction of CH_4 decreased with increase in the permeate pressure. The behavior of CH_4 is opposite because the increase in the permeate pressure creates a less mass transfer and low CO_2 transfer through the membrane. Therefore, purity of CH_4 in reject side decreases.

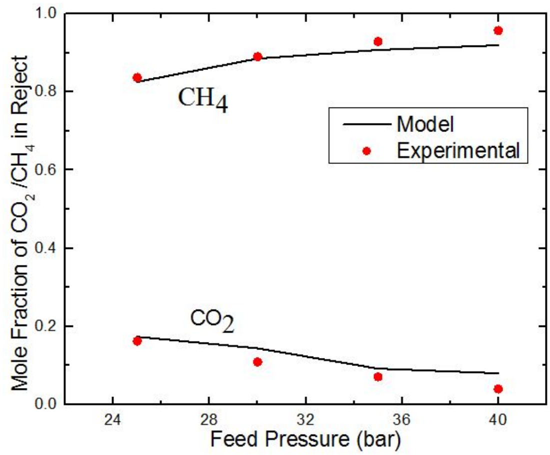

Figure 2. Concentration profiles of CO_2 and CH_4 in reject with feed pressure for counter-current flow pattern.

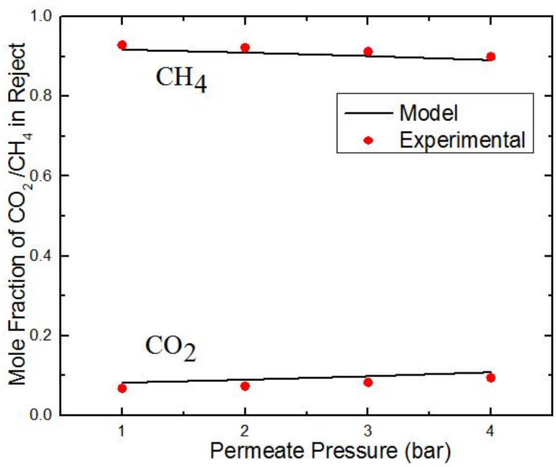

Figure 3. Concentration profiles of CO_2 and CH_4 on reject side with permeate pressure for counter-current flow pattern.

3.2. Effect of feed flow rate

The feed flow rate has a greater effect on moles of CO_2 on the reject side. The influence of feed flow rate on the counter-current pattern is shown in Fig 4. With the increase in the feed flow rate, the contact time of CO_2 concentration with the membrane active surface area is small, and the concentration of CO_2 in the permeate side decreases. Therefore, at a higher flow rate, the concentration of CO_2 increases in the reject. The increase in the feed flow also affects the purity of CH_4 in the reject side. The main theme of this research is achieving higher purity of CH_4 in the reject but when the feed flow rate increases, the CO_2 concentration on the reject side increases too and decrease in the concentration of CH_4 is observed. Fig 4 shows that the concentration of CH_4 is decreased with increases in the feed flow rate.

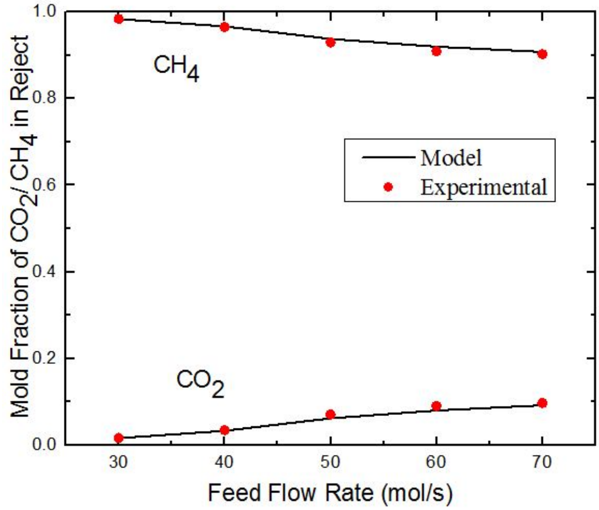

Figure 4. Concentration profiles of CO_2 and CH_4 in the reject with feed flow rate for counter-current flow pattern.

3.3. Effect of CO_2 concentration in the feed

The feed gas is the key parameter for membrane separation process for counter-current flow percentage of impurities. The feed concentration required a specific area to permeate one component through the membrane. When the feed concentration of CO_2 is increased then the amount of gas diffusing through the membrane decreased due to less area. Fig 5 shows that increase in the feed CO_2 concentration results in an increase of the CO_2 concentration in the reject side and a decrease in the concentration of CH_4 on the reject side.

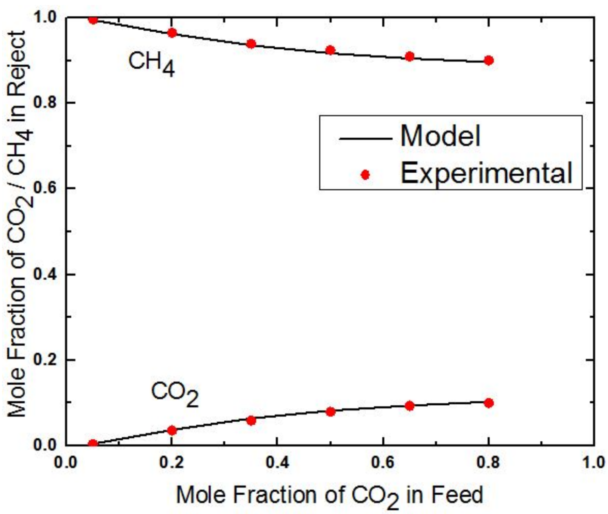

Figure 5. Concentration profiles of CO_2 and CH_4 in the reject with CO_2 feed content for counter-current flow pattern.

Table 2. Comparison between this model data and literature results

Feed Flow rate (mol/s)	Permeate Concentration	Reject Concentration		
		Literature [22]	This Model	%Change
70	0.9178	0.0902	0.09736	5.82
60	0.9106	0.08	0.0853	7.13
50	0.9008	0.062	0.068	4.56
40	0.8886	0.033	0.03467	5.06
30	0.8786	0.0166	0.01667	3.75

Table 1 shows the performance of hollow fiber module like counter-current flow patterns for different flow rates and rejects concentration. The flow rate as a function of stage cut has been investigated for counter-current flow pattern. The data for the performance of asymmetric membranes can be obtained from a mathematical model shown in Table 1. In Table 1, the obtained results from our model are compared with available literature data for separation of CO_2 from CH_4. The results show the better performance of hollow fiber membrane module for the separation of the binary gas mixture. The error in data occurred due to the different model equation are used for the calculations of countercurrent flow pattern. The results demonstrated better performance for high flow rates because it gives maximum purity of gas. The increase in flow rate gave 3% better results in reject values which is better than the values reported in the literature.

4. CONCLUSION

The applied counter-current model has greater advantages than other differential models; it gave valuable results and required less time to simulate. A model is developed to simulate the membrane-based separation of CO_2 from CH_4.

This model is validated with the present literature data and predicts the values of streams at the different flow rates and other various conditions [22]. The counter-current pattern shows that high permeate efficiency for separation of CO_2 from CH_4. This pattern is used to finds the effect of different parameters on the separation of CO_2 from CH_4. It has been found that feed pressure, feed flow rate has direct effect and permeate pressure has opposite effect on the purity of CH_4 in the reject stream. The membrane area is also increased when increases the permeate pressure but decreases with feed pressure. There are no certain values on used properties of the membrane. Therefore, the study of these parameters is impossible here. The results obtained through model are validated using predicting data from the literature for binary gas. The future direction had much more importance with using the multi-component gas system for the separation of CO_2 and compared the results with other solving methods.

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