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Coupling Model for Multi-component Gas Permeation Process

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Abstract. A gas permeation model (Coupling Model) has been developed which has the flexibility to be used for different membrane module configurations. The aim of this work is to predict the performance of a single stage gas separation process using membranes and provide a comprehensive description of process parameters like flow rates, composition, stage cut and stream pressure. The significant feature of this work is the development of computational technique which combines the counter-current flow mode with co-current flow mode. In contrary to other counter-current models (reported in literature), the model reported in this work (Coupling Model) does not require initial conditions to start and also it is independent of any adjustment technique like shooting method. This model is based on real membrane operation and works by the coupling of co-current and counter-current methods. After each iteration, output values of co-current mode become the input values for counter-current mode. This model has the capability to take upto nine components, whereas model reported in literature can handle 4-5 components in a gas mixture to be separated. The results obtained are validated with the published data and will be discussed to elaborate the operation of a gas separation membrane.

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1. Introduction

The separation of gases by perm-selective membranes is an emerging process in the industrial gas separation. Membrane based separations are important in natural gas

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processing, CO_2 capture from flue gases, nitrogen production from air, separation of hydrogen streams in refinery and petrochemical process as well as in ammonia and methanol process streams [1-3]. The feasibility of membrane based separation processes depends on the permselectivity (materials characteristics) along with engineering tools to select appropriate membrane configurations. Generally, gas separation permeators operate in one of the following configurations [7]: countercurrent flow of feed and permeate (shell or tube side feed); co-current flow of feed and permeate (shell or tube side feed); and radial cross flow (shell side feed). Mathematical model (for any type of permeator) is constituted by the equations which govern the transport across the membrane; mass balance equations; pressure drop relations/assumptions for both permeate/feed sides; and boundary conditions that reflect the configuration and operation of the permeator.

A clear understanding of these models and the parameters affecting the membrane process is vital. As this becomes the basis for the designing and optimization of membrane process [9]. The modeling of membrane separation process was initiated by Wander and Stern [9] by studying the effects of different process variables like pressure ratio and stage cut on the cross flow and perfect mixing for binary components. Pan and Habgood [10] extended the work to tertiary components for cross flow mode and provided a parametric analysis of cascade permeation. A lot of information about the theoretical and dimensionless of membrane separation for flow modes like cross-flow, co-current, counter-current and perfect mixing has been found in literature [4-6]. Cooker [11] divided the separation modules in small stages and introduced a tridiagonal approach to analyze the membrane separation phenomena.

This work presents a new modeling approach for membrane separation by introducing the concept of "Coupling Model". This modeling approach combines two flow modes (counter-current and co-current) and both flow modes run simultaneously. The "Coupling Model" does not require any start up condition and also does not require any adjustment technique like shooting method for the system convergence. The results from "Coupling Model" show good agreement with the results reported in literature in less computing time. This model is capable of treating upto nine components, in contrary to other models reported in literature, which can take upto four components in membrane based gas separation process.

2.Gas Permeation Model

Gas permeation model is constituted of various parameters which affect the membrane separation phenomena. A pressure difference/ratio across the membrane, feed

compositions and feed flow rate are considered as operating parameters while membrane type, area and the feed volume are considered as design parameters. The membrane type and composition describe the permeabilities of the components. The combination of both operating and design parameters along with the material balance and mass conservation equations make the basis to compute the desired output values of compositions and flow rates on the retentate and permeated sides [8].

2.1 Model equations

A membrane separation process initiates by introducing the mixture of components at the feed side of the membrane. Based upon the permeability values of components, the membrane lets the more selective component pass through it while retaining the others ones on the retentate side. A driving force (generally pressure difference across the membrane) is needed to precede the separation process using the available membrane area. The Fick's law of diffusion is the principle equation used in the membrane permeation/separation process. The general form of Fick's law has been converted into the form describing the parameters affecting on the separation process. Along with the Fick's law and its modified form, the set of generalized equations (conservation equation, flow equations, stage cut) are integrated in the permeation model [9]. A brief detail of these equations are given as,

Fick's law of diffusion: $$J = -D\frac{\partial C}{\partial d} \qquad (1)$$

Modified Fick's law of diffusion: $J = P_i \frac{(P_u x_i - P_p y_i)}{\delta}$ (2)

Mol Conservation equation: $\sum_i^n X_i = 1 \sum_i^n x_i = 1 \sum_i^n y_i = 1$ (3)

Flow equation: $L_f = L_u + L_p$ (4)

Component balance equation: $X_{if} L_f = x_i L_u + y_i L_p$ (5)

Stage cut: $$\Phi = \frac{L_p}{L_f} (6)$$

Equation (1) is the general form of Fick's law. Equation (2) is the modified Fick's equation describing the parameters affecting the membrane separation phenomena. Equation (3) is the mol

Conservation equation which utilizes at the feed, retentate and permeate side of membrane. Equation (4) and equation (5) are the flow balance and material balance equation used to check the system stability. Stage cut, describing in equation (6) is an important parameter

in the membrane separation. It is the amount of flow passed through the membrane out of the total feed flow. Besides of these equations, the variations in the retentate flow and the retentate compositions can be described by equation (7) and equation (8) respectively. Huges&Acharya [13] describe a method to derive equations (7-12), while a numerical technique such as Euler or RK method has been used to solve above differential equations. Table X shows equations for different flow modes based on the quantities mentioned above.

Variation in retentate flow:

$$\frac{dL_u}{dA} = -\sum_i^n \frac{P_i(P_u\, x_i - P_p\, y_i)}{\delta} \quad (7)$$

Variation in retentate composition:

$$\frac{dx_i}{dA} = \frac{1}{L_u}\left[x_i\left(\sum_i^n \frac{P_i(P_u x_i - P_p\, y_i)}{\delta}\right) - \frac{P_i(P_u x_i - P_p y_i)}{\delta}\right] \quad (8)$$

The permeation equation used in different flow modes are described as,

Cross flow:

$$y_i = \frac{P_i(P_u\, x_i - P_p\, y_i)}{\sum_{j=1}^n Pj\left(P_u\, x_j - P_p\left(\frac{P_u\, P_j x_j\, y_i}{P_p\, P_j\, y_i + P_i(P_u\ x_i - P_p\ y_i)}\right)\right)} = F(\{x_i\},\{P_i\}, P_p, P_u) \quad (9)$$

Co-current flow:

$$y_i = \begin{cases} \frac{L_f\, X_{if} - x_i\, L_u}{L_f - L_u}, \ L_f \neq L_u \\ F(\{x_i\},\{P_i\}, P_p, P_u), \ L_f = L_u \end{cases} \quad (10)$$

Counter-current flow: $$yi = \begin{cases} \frac{q_h\, x_i + L_p\, y_i - L_f\, X_{if}}{q_h + L_p - L_f}, \ q_h + L_p \neq L_f \\ F(\{x_i\},\{P_i\}, P_p, P_u), \ q_h + L_p = L_f \end{cases} \quad (11)$$

2.2 Dimensionless Analysis

Dimensionless analysis is used to develop a similarity among different flow modes. Different dimensionless quantities are described in this regard.

Pressure ratio: ratio of permeate pressure to feed pressure, $\qquad \qquad P_r = \frac{P_p}{P_u}$

Selectivity: ratio of permeability of two components, $\alpha = \frac{P_i}{P_j}$

Flow ratio: ratio of retentate flow to feed flow, $$\overline{q_h} = \frac{L_u}{L_f}$$

Stage cut: ratio of permeate pressure to feed pressure

Flow rate

$$\frac{dL_u}{dA} = -\sum_i^n \frac{P_i(P_u x_i - P_p y_i)}{\delta}$$

$$\frac{d(\overline{q_h})}{d(\overline{A_d})} = -\sum_i^n \gamma_i(x_i - P_r y_i)$$

Composition

$$\frac{dx_i}{dA} = \frac{1}{L_u}\left[x_i\left(\sum_i^n \frac{P_i(P_u x_i - P_p y_i)}{\delta}\right) - \frac{P_i(P_u x_i - P_p y_i)}{\delta}\right]$$

$$\frac{dx_i}{d(\overline{A_d})}$$

Cross Flow mode

$$y_i = \frac{P_i(P_u x_i - P_p y_i)}{\sum_{j=1}^n Pj\left(P_u x_j - P_p\left(\frac{P_u P_j x_j y_i}{P_p P_j y_i + P_i(P_u x_i - P_p y_i)}\right)\right)}$$

$$= \left[\frac{x_i\left(\sum_j^n \gamma_j(x_j - P_r y_j)\right) - \gamma_i(x_i - P_r y_i)}{\overline{q_h}}\right]$$

$$y_i = \frac{\gamma_i(x_i - P_r y_i)}{\sum_{j=1}^n \gamma_j\left(x_j - P_r\left(\frac{\gamma_i x_j y_i}{P_r \gamma_j y_i + \gamma_i(x_i - P_r y_i)}\right)\right)}$$

Co-Current mode

$$y_i = \begin{cases} \dfrac{L_f X_{if} - x_i L_u}{L_f - L_u}, & L_f \neq L_u \\ F(\{x_i\},\{P_i\},P_p,P_u), & L_f = L_u \end{cases}$$

Counter-Current mode

$$yi = \begin{cases} \dfrac{q_h x_i + L_p y_i - L_f X_{if}}{q_h + L_p - L_f}, & q_h + L_p \neq L_f \\ F(\{x_i\},\{P_i\},P_p,P_u), & q_h + L_p = L_f \end{cases}$$

$$y_i = \begin{cases} \dfrac{X_1 - \overline{q_h} x_1}{1 - \overline{q_h}} & \overline{q_h} \neq 1 \\ F(\{x_i\},\{\gamma_i\},P_r), & \overline{q_h} = 1 \end{cases}$$

Initial Conditions

$$x_i(A = 0) = X_{if} \quad ; \quad L_u(A = 0) = L_f$$

$$y_i = \begin{cases} \dfrac{\overline{q_h} x_1 + \Phi y_i}{\overline{q_h} + \Phi - 1}, & \overline{q_h} \neq 1 - \Phi \\ F(\{x_i\},\{\gamma_i\},P_r), & \overline{q_h} = 1 - \Phi \end{cases}$$

$$x_i(\overline{A} = 0) = X_{if} \quad ; \quad \overline{q_h}(\overline{A} = 0) = 1$$

$$\Phi = \frac{L_p}{L_f}$$

Dimensionless area:

$$A_d = \frac{A P_1 P_u}{del\, L_f}$$

Model equation for counter-current mode (equation (12)) suggests that this flow mode has to be started in the backward direction. i.e. from the exit of the membrane to the feed inlet (in equation (12)), $L_{u,o}$ and $x_{i,o}$ represents the exit flow rate and exit composition respectively. Most of the literature follows this method regardless the nature of the equations used in the work [9-12]. In a real time experiment, the counter flow separation started in the same manner as the other modes of flow i.e. from feed to the exit point of the

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membrane. The technique reported in literature encounters two major problems;

(a) outputs of other mode to serve as initial conditions for counter current flow

(b) adjustment of counter mode outputs with the initial feed

The starting point for counter-current flow model is the membrane outlet. So, a value of composition and flow is required at that point to run the counter-current flow towards the feed side of membrane. Such a value is either taken from a trial method or from other modes of separation like cross flow or co-current. Thus, to simulate counter-current mode, other flow modes will have to be started first and then their output becomes the input for this case. The counter-current flow mode runs toward the inlet of membrane by picking the initial conditions from outlet. The accuracy of counter-current flow mode is validated if $L_{u,out} = L_{feed}$ and $x_{out} = x_{feed}$ but sometime this condition is not met because computed result differ with actual one. For the convergence of output result with initial condition, a numerical technique like shooting method is required in most of the published method. An adjustment technique may helpful when the number of components are limited (two or three) but it may become complicated as soon as the number of components increases. Since, for n number of components, we require n+1differential equations (retentate flow rate is the additional differential equation) and the adjustment of each component plus the flow rate value is necessary.

3. Coupling Model

The coupling model is a new technique in membrane based gas separation modeling. In this model, counter-current flow mode is coupled with cross flow or co-current mode. Since, co-current and counter-current are similar except the direction of permeation with respect to feed so, co-current mode is coupled with counter-current mode. The two flow modes run simultaneously without modification of respective permeation equations. The coupling model is described in the following steps,

1) Introducing the initial compositions X_i and initial feed flow L_f at the feed side for co-current.

2) Set input parameters like retentate pressure p_u and permeate pressure p_p across the membrane.

3) Set initial conditions $L_{u,out} = L_{feed}$ and $x_{out} = x_{feed}$ at area A$_{d1}$ = 0.

4) Compute the permeate fraction y_i for co-current case using equation (11).

5) Iterate the area $Ad_2 = Ad_1 + dA_d$ where dA$_d$ is the small area increment.

6) Compute change in flow rate and composition of retentate with differential area using

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equation (7 & 8) respectively. The retentate flow rate and composition after each area increment are calculated as $L_{u2} = L_{u1} - dL_u$ and $x_2 = x_1 - dx_1$ where dL_u and dx_1 are change in retentate flow and composition respectively.

7) Start counter-current method with initial conditions $x_{2\ counter} = x_2$, $A_{d2\ counter} = Ad_2$ and $L_{u2\ counter} = L_{u2}$.

8) Compute the permeate fraction for counter-current case when area is $Ad_{2\ counter} = Ad_2$.

9) Iterate the area in the reverse direction, $Ad_{1\ counter} = Ad_{2\ counter} - dA_d$.

10) Compute the change in the retentate composition and flow rate for counter current case. i.e.

$$x_{1\ counter} = x_{2\ counter} + dx_2 \ ; \qquad L_{u1\ counter} = L_{u2\ counter} + dL_{u1}$$

11) Compute the permeate fraction for counter current case by using equation (12) and repeat step 5-12 till a desired value at retentate or permeate is achieved.

In this methodology, the co-current and the counter current modes run simultaneously so no initial condition is required. The output for the co-current case automatically becomes the input for the counter-current mode after each iteration and the two modes. Moreover, since the initial conditions have been automatically assigned to the counter-current case after each iteration. The process runs in backward direction, so the naturally converging values, with initial conditions of membrane separations are obtained. It is worth mentioning here that in coupling model, feed points for the co-current and counter-current flow mode are not at the same positions. The feed position for the co-current lies at the inlet of the membrane while the feed positions for counter-current lies after one area increment. The feed points for co-current and counter-current mode can be made closer to one position by taking small increment of area.

4. Results and Discussions

The results obtained from "Coupling Model" are validated against the results of some important membrane based industrial gas separations which have been reported in the literature.

4.1. Natural gas processing

A membrane process is applied to separate impurities from natural gas. Gas mixture consists of Hydrogen (H_2), Nitrogen (N_2), Oxygen (O_2) and Methane (CH_4). The membrane used in this process has the least affinity towards methane and therefore it is separated from the mixture [1]. Input parameters for multi-component gas mixture are

presented in Table 4.1.

Table 4.1: Input parameters (process and membrane) for multi-component gas mixture

Parameter	Value	Component.	Mole Fraction [%]	Permeability $X10^7(\frac{cm^3(STP).cm}{s.cm^2.cmHg})$
Permeate Pressure [cm Hg]	50	Hydrogen	0.10	0.2
Feed Pressure [cm Hg]	380	Nitrogen	0.23	0.11
Feed Flow Rate [cm³/sec]	10^6	Oxygen	0.40	0.044
Thickness [cm]	$2.54×10^{-4}$	Methane	0.27	0.013

The graph has been plotted for composition vs stage cut, keeping   a constant pressure difference (pressure ratio) across the membrane $(P_r = \frac{P_p}{P_u} = 0.13)$.
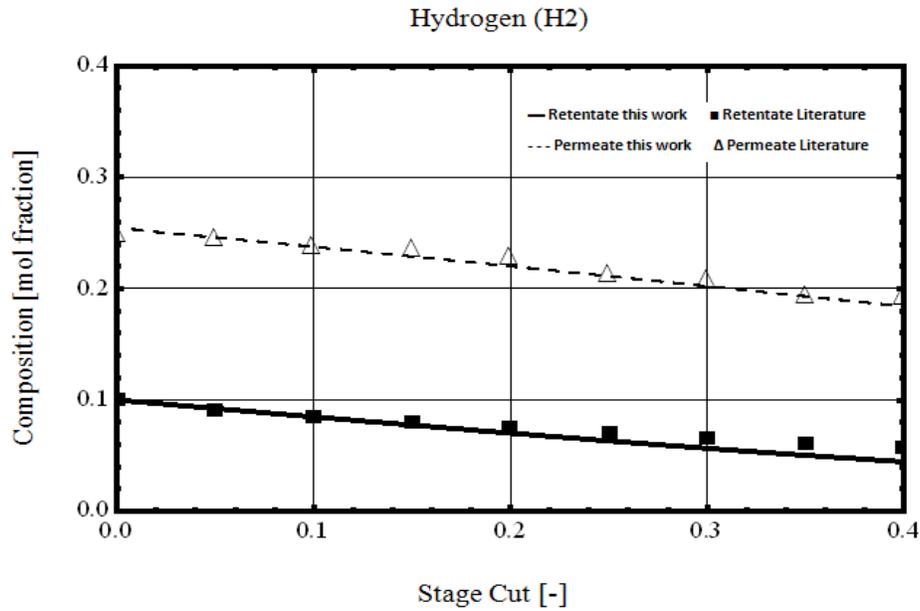
Figure 4.1:Variation in H₂ composition against stage cut keeping pressure ratio constant (Pr = 0.13).

Nitrogen (N$_2$)
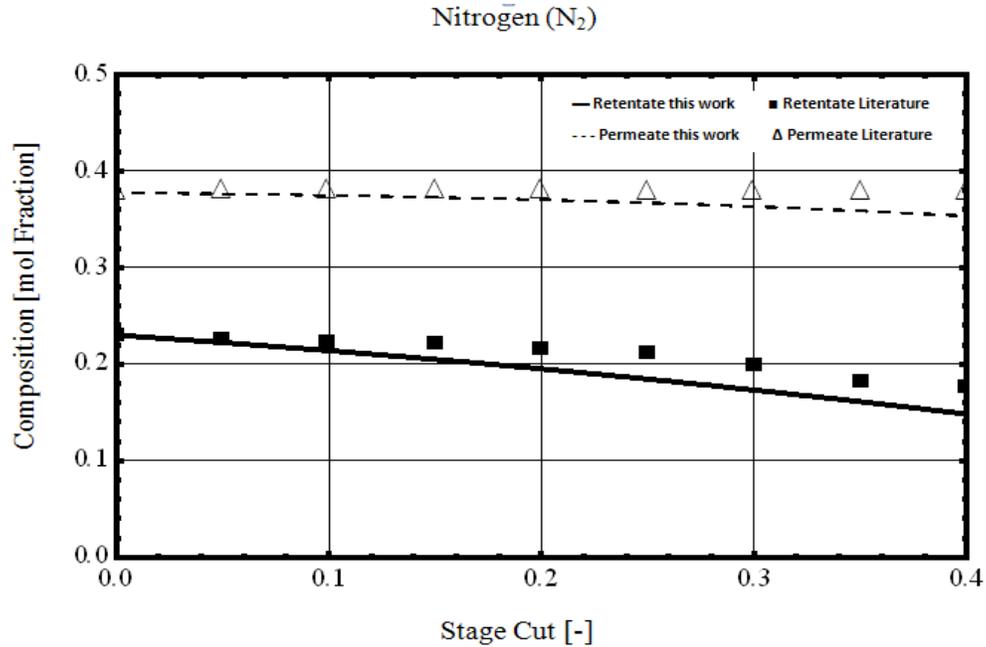
Figure 4.2:Variation in N$_2$ composition against stage cut keeping pressure ratio constant (Pr =0.13)

As evident from Fig. 4.1, the trends for both retentate (solid line) and permeated (broken line) streams are decreasing along stage cut. The variation in gas compositions both on the retentate and permeate side can be attributed to stage cut and pressure ratio. The permeability of hydrogen is greater compared to other components, therefore, it passes through the membrane preferentially. However, hydrogen concentration decreases on the retenate side which results in a decrease in hydrogen concentration on the permeate side as well. A similar situation is observed in case of N$_2$ since it is the 2nd most permeable component in the gas mixture. Oxygenand methane are the least permeable components in this case. Their graphs are shown in figures 4.3 and 4.4. They exhibitdifferenttrends compared to Hydrogen and Nitrogen (Fig.4.1 and Fig. 4.2). Here, concentrations of components in permeate stream increase with as their concentration in retentate stream increases. Since, these are least permeable components so the membrane does not allow these to pass through it. In the continuous permeation (of hydrogen and nitrogen), retentate flow gets enriched with least permeable components. The change in compositions with

pressure ratios is also validated against the published data. The stage cut was kept constant (Φ = 0.4) in this case. The process and membrane parameters were kept constant as given in Table 4.1- 4 2.Pressure ratio is changed by changing the feed pressure.
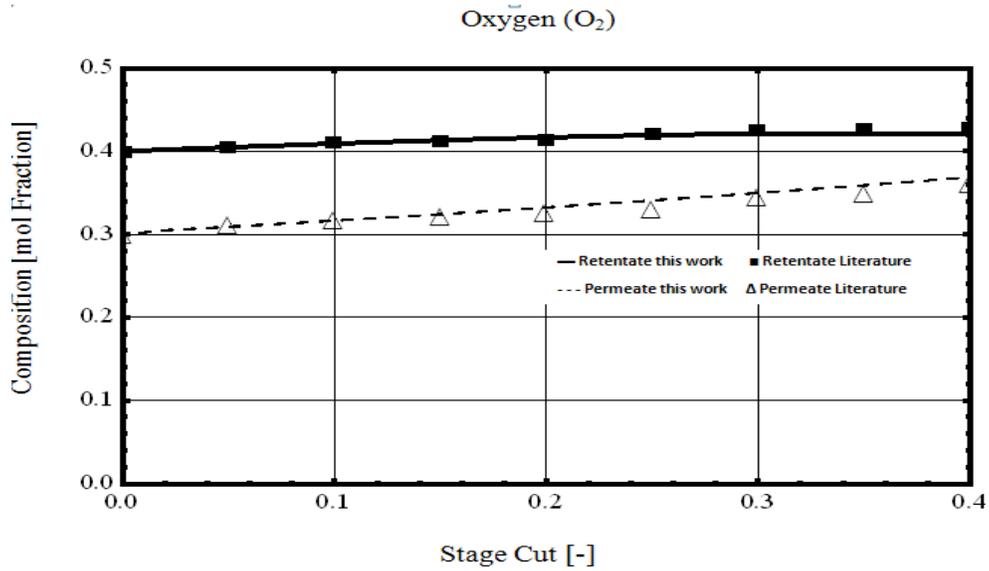
Oxygen (O$_2$)

Figure 4.3:Variation in O$_2$ composition against stage cut keeping pressure ratio constant (Pr =0.13)

Methane (CH$_4$)
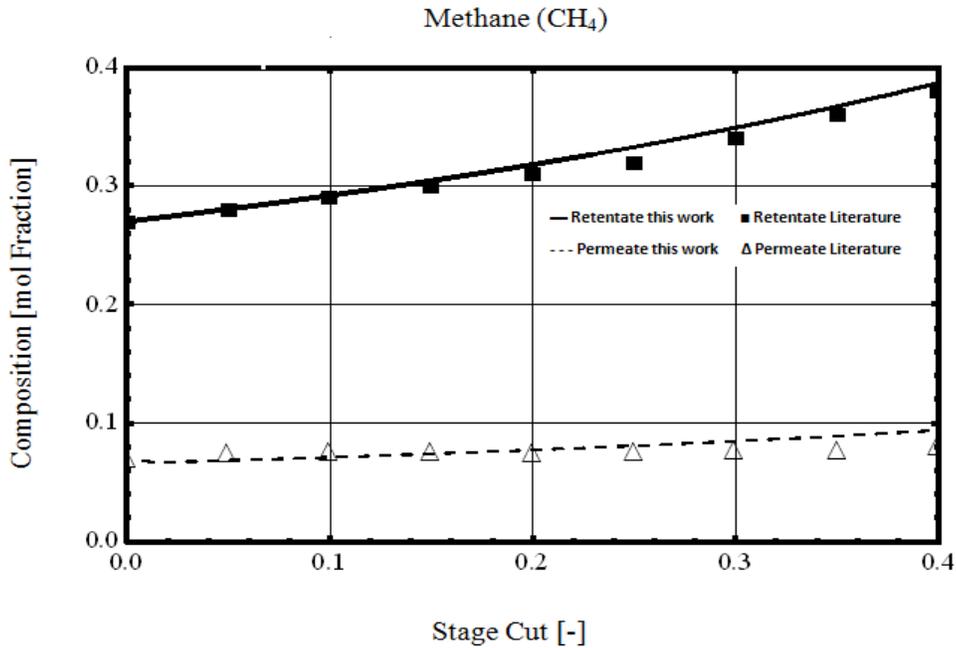
Figure 4.4:Variation in CH$_4$ composition against stage cut keeping pressure ratio constant (Pr = 0.13).

The results validation after changing the pressure ratio is depicted in figures 4.5-4.8. In the figures, lines (both solid and broken) show the results of published data while bullets show the results from coupling model. It can be seen that the results of "Coupling Model" are in good agreement with the published data [1].
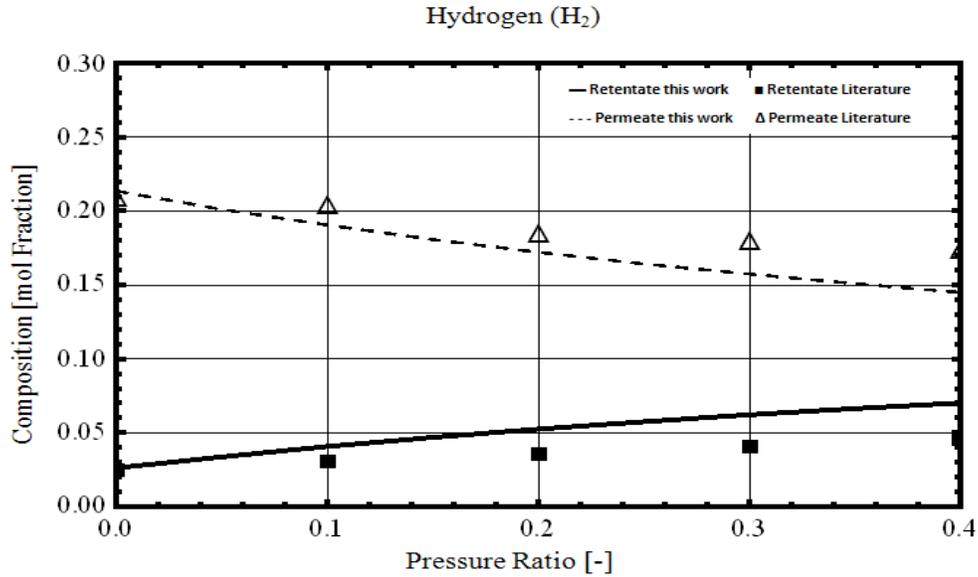
Figure 4.5: Variation in H$_2$ composition against pressure ratio keeping stage cut constant ($\Phi = 0.4$)
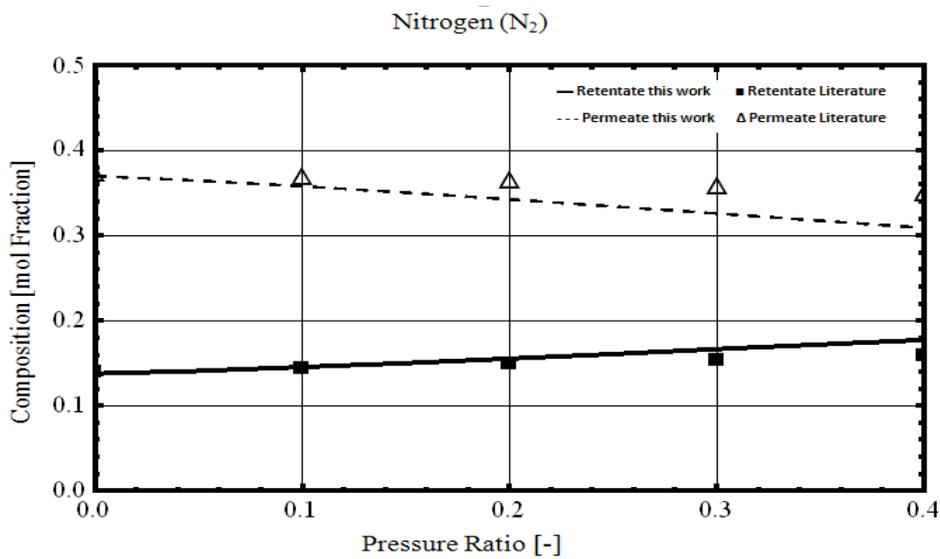
Figure 4.6: Variation in N$_2$ composition against pressure ratio keeping stage cut constant ($\Phi = 0.4$).
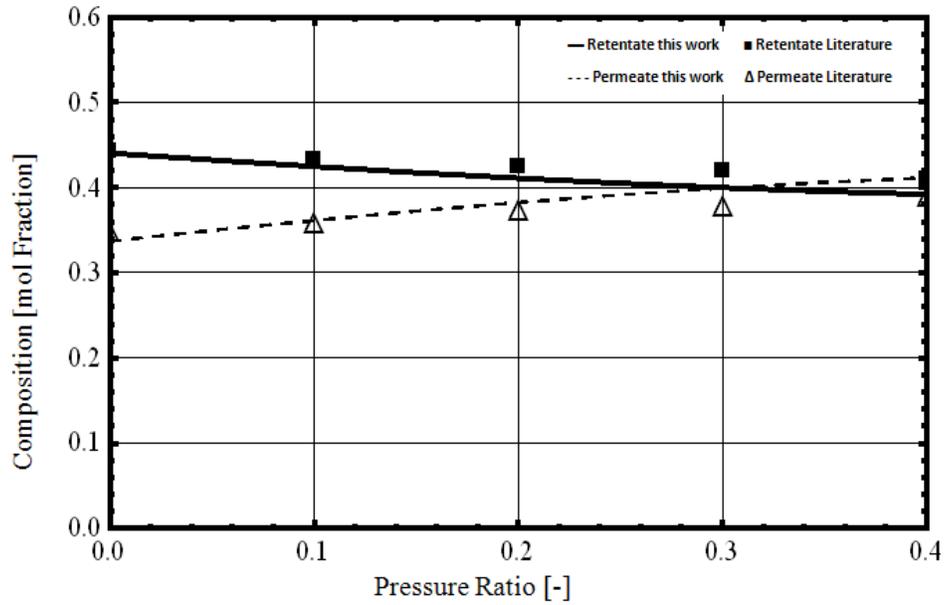
Oxygen (O$_2$)

Figure 4.7: Variation in O$_2$ composition against pressure ratio keeping stage cut constant (Φ = 0.4).
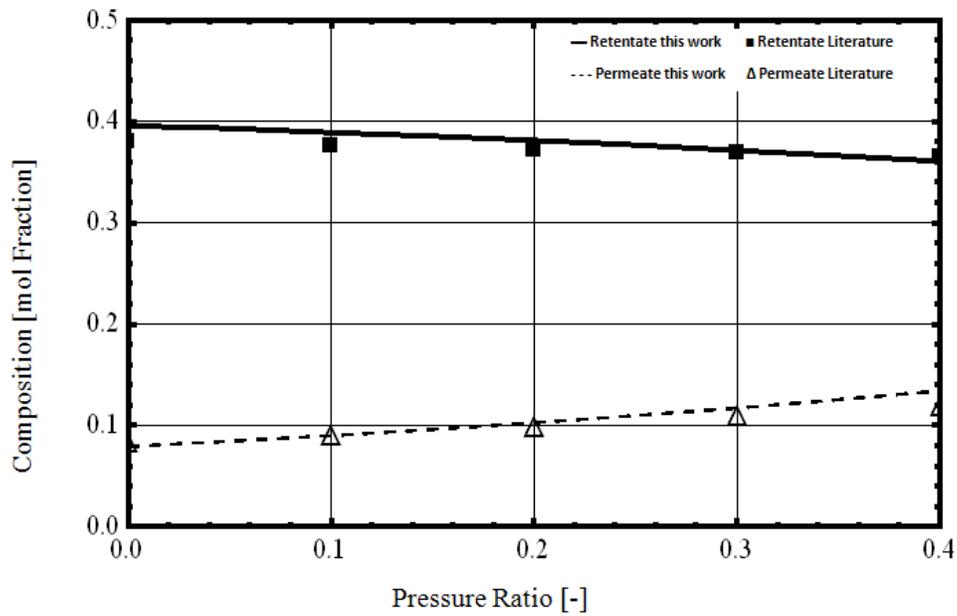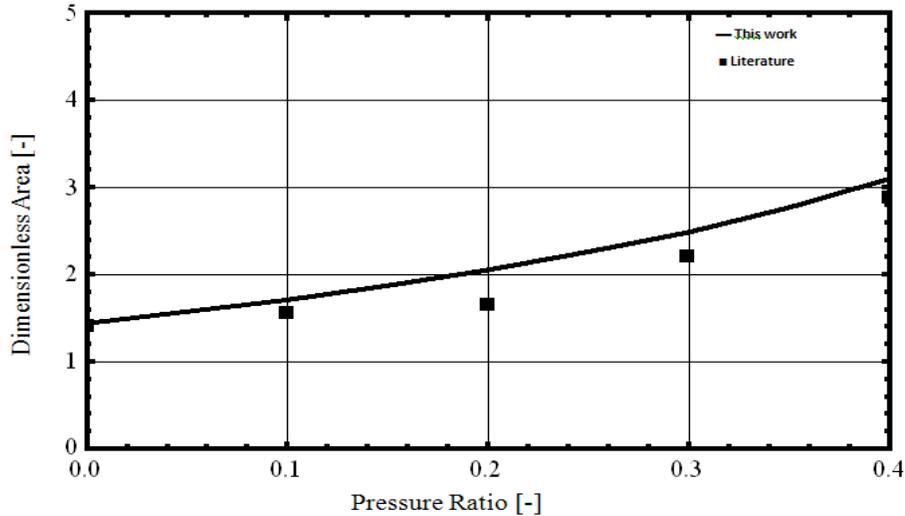
Methane (CH$_4$)

Figure 4.8: Variation in N$_2$ composition against pressure ratio keeping stage cut constant (Φ = 0.4).

The membrane area is an important parameter for gas separation and is

designed in such a way that the maximum separation is done with the minimum area usage. The graph below shows the variation in membrane area with pressure ratio.

Figure 4.9: Area requirement against pressure ratio keeping stage cut constant (Φ =0.4).

It can be seen that with the increase in pressure ratio, the area required for the separation increases. Since the pressure ratio is defined by permeate pressure / feed pressure. Therefore, increase in pressure ratio is due to increase in the permeate pressure or decrease the retentate pressure. In both cases driving force for permeation decreases and more membrane area is required to attain the desired degree of gas separation.

4.2. Hydrogen separation in gas oil de-sulfurization of refinery streams

The results of the "Coupling Model" are validated with the results obtained by the model by Kaldis and the experimental results of gas oil de-sulfurization of refinery stream which generally contain H_2, light hydrocarbons (CH_4, C_2H_6) and H_2S [5]. Taking into account that membrane (polyimide) exhibits the same permeability values for CO_2 and H_2S, H_2S was replaced in by CO_2 mainly for safety and handling reasons. The process and membrane parameters are presented in Table 4.2.

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Parameters	Values		Component	Mole Fraction	Permeance ($\frac{mol}{s-Pa-m^2}$)
Permeate Pressure [bar]	1		Hydrogen [H$_2$]	0.10	9.7304 $\times 10^{-8}$
Feed Pressure [bar]	20		Methane [CH$_4$]	0.167	3.1178 $\times 10^{-8}$
			Ethane[C$_2$H$_6$]	0.043	0.123802 $\times 10^{-8}$
Flow Rate [Nl / hr]	5-30		Carbon Dioxie [CO$_2$]	0.115	0.021414 $\times 10^{-8}$

Table 4.2: Input parameters (process and membrane) for hydrogen separation process

The data is plottted upto 0.6 stage cut while pressure ratio is kept constant. The effect of pressure on residue streams is shown in figures 4.10 to 4.13. A good agreement between calculations (This work and literature) and experimental results can be seen in all figures. With the exception of a slight increase of hydrogen permeate concentration; the other gases in both streams remain almost unaffected by feed pressure. This behavior can be attributed to the fact that the permeabilities of constant gases, such as H$_2$, do not depend on pressure. The latter is not valid for non-ideal gases such as CO$_2$ and C$_x$H$_y$which show pressure dependent permeabilities.
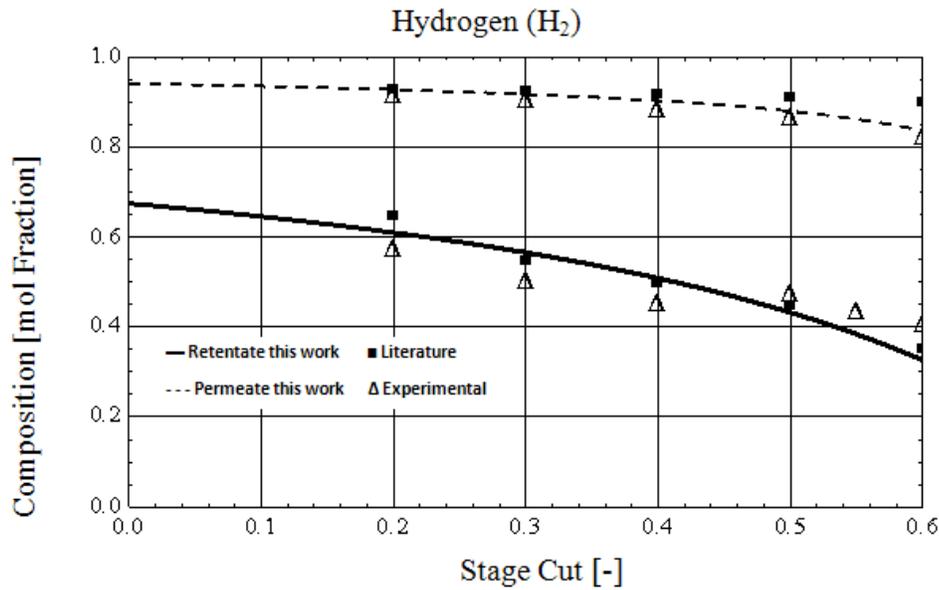
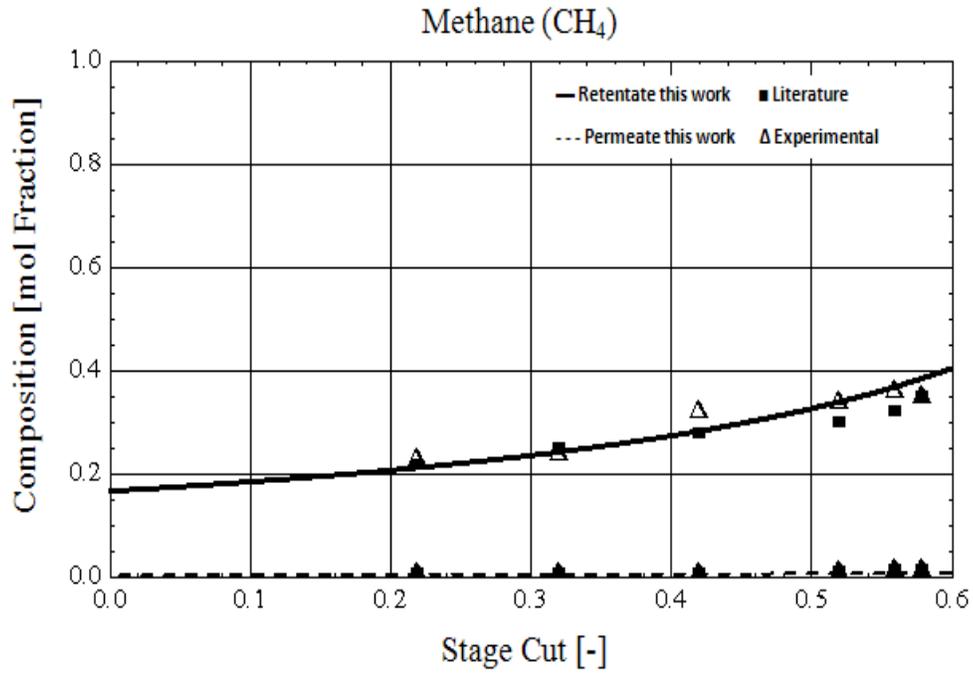**Figure 4.10:**Variation in H$_2$ composition against stage cut.

Methane (CH₄)

Figure 4.11: Variation in Methanecomposition against stage cut.

Ethane (C₂H₆)
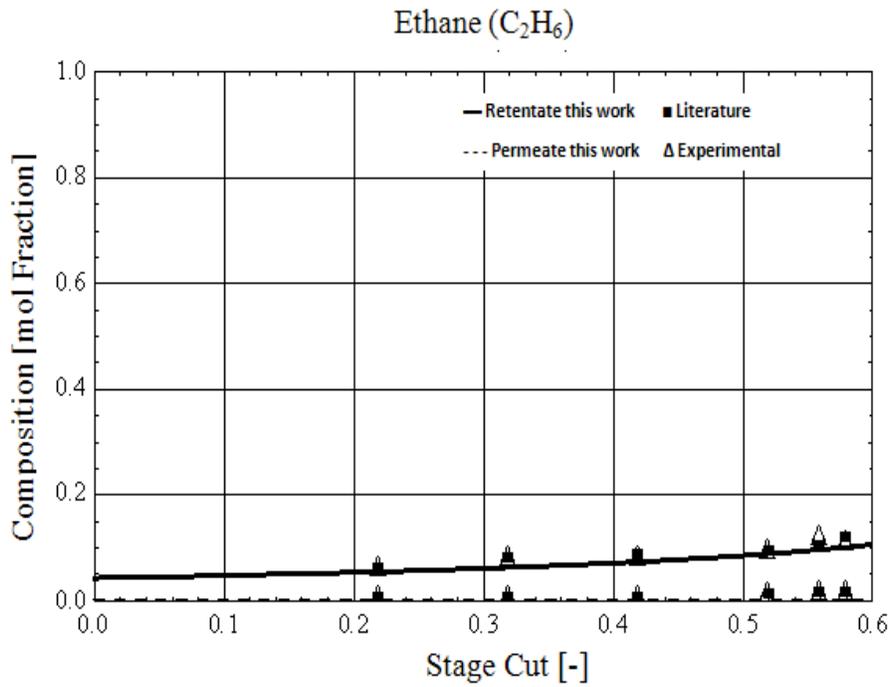
Figure 4.12: Variation in Ethanecomposition against stage cut.
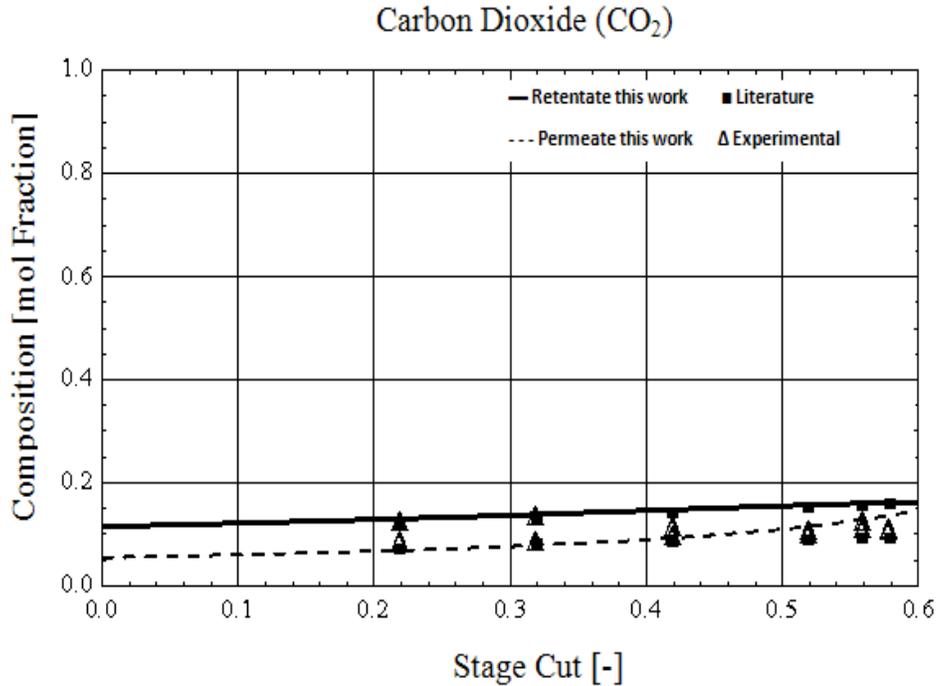
Carbon Dioxide (CO₂)

Figure 4.13: Variation in Ethanecomposition against stage cut.

4.3. Hydrogen treatment

Hydrogen treatment is a common unit operation in refineries. In this process, hydrogen is used to reduce the sulfur, nitrogen, metals and carbon residue content of the feed stock. The process requires substantial amounts of hydrogen gas which is available but needs to be purified by using membrane process. The results of "Coupling Model" have been compared with tri-diagonal model. The process and membrane parameters used in this process are given in Table 4.3.

Component	Feed mole fraction	Permeance [GPU]	Input parameter	Value
H_2	0.650	100	Thickness of Membrane [nm]	10^{-4}
C_2H_4	0.025	3.03	Feed Pressure [bar]	a) 42.4b)76.9
CH_4	0.210	2.86	Permeate Pressure [bar]	a)7.9b)42.4
C_2H_6	0.080	2.00		
C_3H_8	0.035	1.89	Pressure Ratio(modified)=P_f / P_p	a)5.3b)1.8

Table 4.2: Input parameters (process and membrane) for hydrogen separation process
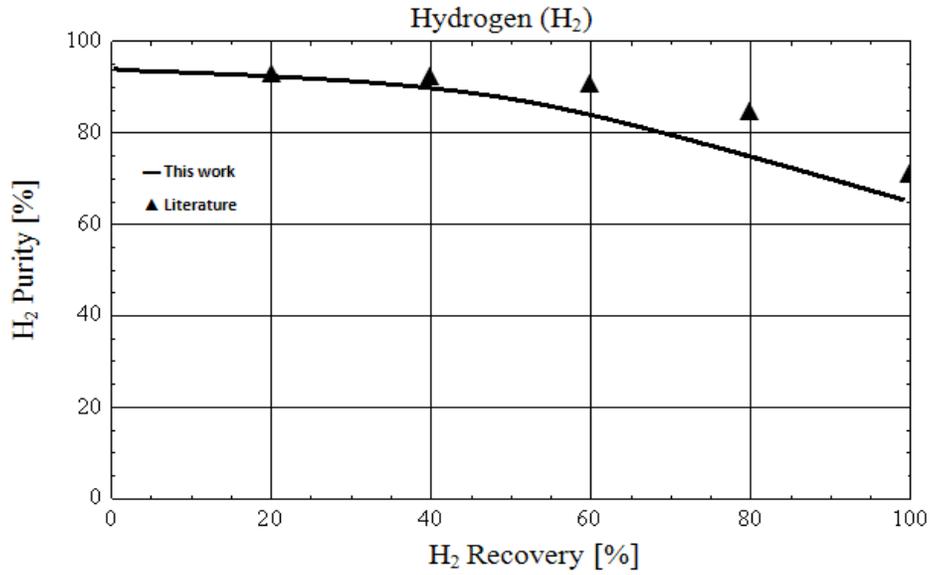
Figure 4.14: Hydrogen recovery versus purity for pressure ratio of 1.8.
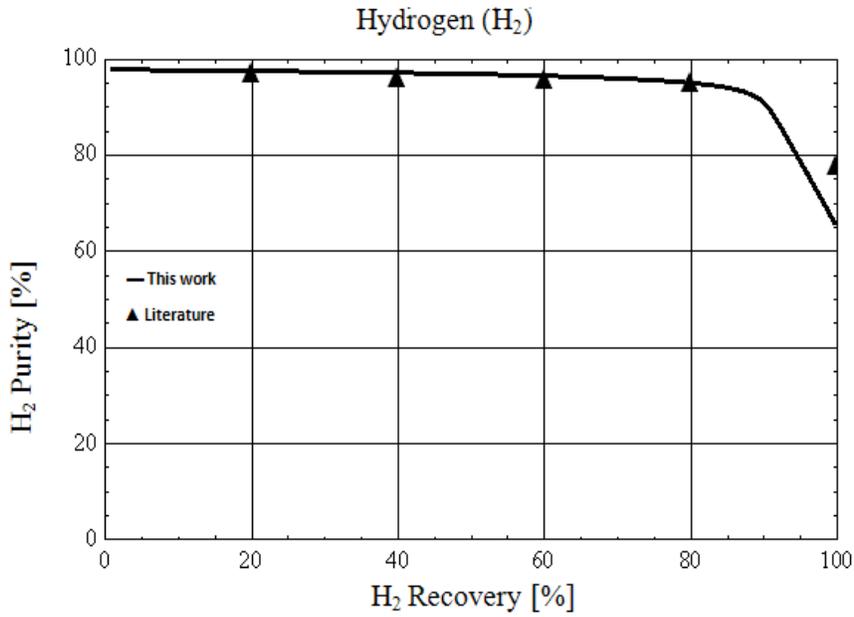

Figure 4.15: Hydrogen recovery versus purity for pressure ratio of 5.3.

The results obtained from coupling model are plotted for two different values of pressure ratio (5.3 and 1.8) and can be seen that these are in good agreement with the published data [7].

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5. Conclusions

In perspective of membrane based gases separation, "Coupling Model" has been developed which combines the co-current and counter-current flow modes. The simulation results show a good agreement between the results already reported in literature and the results obtained from "Coupling Model". It is worth mentioning here that the cases published in literature either used the initial conditions for the counter current cases or depend upon the adjustment methods to converge the values. This method is helpful especially for multi-stage configurations since it eliminates the initial condition (required at every stage for the counter current case) problem. Another important feature of this work is the development of algorithm for the pressure ratio as it is often desired to investigate the composition gradients against pressure ratio while keeping the stage cut constant. The algorithm developed in this program runs only once and the value of composition gradient can be calculated for different pressure values. The permeation model has the capability to handle up to nine components at a time and their separations could be performed in considerably less computing time. The process and membrane parameters (flow rates, pressure differences and permeances) which effect the membrane separation are discussed in detail.

Reference

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