This chapter deals with the simulation aspects as well as scale-up, design and economics of Pebax 2533 membrane into modular configuration to enable their commercial viability. Initially, the basic criteria for the selection of a most suitable module geometry are discussed. Optimized parameters for machine-based manufacture of TFC membrane sheets of large sizes are presented. Design and method of fabrication for spiral wound elements of 0.20 to 0.25 m² effective membrane area are described with illustrations. After the preliminary experiments with three different ultraporous substrates made of PVDF, PES and PSF, the separation of binary CO₂/CH₄ mixtures with Pebax 2533 membrane coated on the best substrate was carried out at the maximum possible pressure of 60 kg/cm² to generate data for the design of a commercial gas permeator.

Mathematical analysis was done to obtain the numerical solution for the plug flow model using a step-wise method, wherein the membrane area was divided into numerous infinitesmal areas. Equations, which represent the material balance across an infinitesmal area were solved to construct a simulation algorithm in a microsoft excel that is capable of giving outputs of membrane area requirement as well as product and permeate compositions for inputs of membrane properties, stage cut, feed capacity, pressure and feed composition. Based on the simulation results, a commercial gas permeator of 100 m³/h capacity was designed for the purification of natural gas to pipeline quality. An economic estimate commercial plant giving a
detailed breakup of capital investment and operating cost is presented at the end of the chapter.

VI.1. SELECTION OF A MEMBRANE MODULE

The heart of any membrane plant is the module i.e., the technical arrangement of membranes. Some important aspects to be considered for module design include packing density, cost-effective manufacture, easy access for cleaning and cost effective membrane replacement. Based on these considerations, modules can be distinguished into four major categories displayed in Fig.VI.1. Characteristics of the modules are compared in Table VI.1.

VI.2. Tubular Module

Tubular membrane is in a hose form on the inside of pressure-tight tubes having 12–24 mm internal diameter [1-3]. If the material of the support tube is impermeable, then a thin porous tube is fitted between the support tube and the membrane. In many cases, several tubular membranes are assembled in a common support block to increase the packing density. Even though the manufacturing costs of these modules are high, they offer excellent resistance to fouling with a low pressure-drop, but are not suitable for high-pressure operations such as in the present case.
VI.3. Hollow Fiber and Capillary Modules

VI.3.1. Capillary Module

Capillary module (not shown in the figure) has a tube bundle of fibers that are arranged parallel to each other with either end attached to a head plate. The active layer is usually on the inside of the capillary, which means that the feed flows inside the capillaries and the permeate is collected at the outside. Dimensions of the fibers are: inner diameter (ID) of 250-1500 µm and outer diameter (OD) of 400-2500 µm [4]. This module has a moderate packing density of 1000 m²/m³ with a higher mass transfer resistance as compared to all other modules, due to the predominant laminar flow. The capillary module offers a maximum pressure-drop of 10 kg/cm², but is unsuitable for high-pressure operations.

VI.3.2. Hollow Fiber Module

Hollow fiber module consists of a pressure vessel containing a bundle of very fine individual fibers of 10-40 µm ID and 80-200 µm OD (see Fig.VI.1). The active layer is usually coated on the outer surface of the fiber, which is in contact with the feed and the permeate flowing inside the fibers. Gill and Bansal [5,6] reported that the counter-current flow is always superior to co-current flow in the module. Compared to the capillary modules, hollow fiber module has a much higher packing density of 10000 m²/m³, which can be produced at a very low cost using the sophisticated equipment that is available. Fabrication of polymeric membranes commonly involves the phase inversion process, wherein polymer solution is extruded through an annular die and contacted with a non-solvent (i.e., water). If an air gap exists between the exit
of the die and the coagulation bath, a thin dense skin layer of the order of 100 nm or less will form on the top of a porous, highly permeable polymer support. In order to assure pin-hole free hollow fibers, various coating and repair techniques can be employed as suggested by Henis and Tripodi [7]. Construction of these modules and their operations are discussed in detail by Kesting and Fritzche [8]. Of late, this module has attracted the attention of many researchers for an effective separation of gaseous mixtures due to its outstanding ability to withstand the pressure drops up to as high as 80 kg/cm² [4].

VI.4. Plate and Frame Module

These modules resemble a filter press stack and flat sheet membranes in rectangular or circular shapes that are assembled along with the feed and product distributors in such a manner that alternate chambers are created for feed flow and other alternate for the permeate flow. These modules are expensive to fabricate and suitable for only low pressure-drop processes such as electrodialysis (ED) and pervaporation (PV).

VI.5. Spiral-wound Module

These are characterized by high packing density (> 900 m²/m³) and a simple design. Essentially, two or more membrane pockets are wound around a centrally located permeate collecting tube with a special mesh used as the spacer. The membrane pocket consists of two membrane sheets with a highly porous material in between, which are glued together along the three edges. The fourth edge of the pocket is connected to the collecting tube. Several such pockets are spirally wound
around the perforated permeate tube with a feed-side spacer placed between the pockets [9]. Usually, several such membrane elements are arranged in one pressure vessel. The feed side flow is strictly axial, while the permeate flows through a porous support inside the pocket along the spiral pathway to the collecting tube [10]. This module was selected for further studies due to the low manufacturing cost, suitability to high-pressure operation and availability of infrastructure as well as technology in India for its fabrication. The detailed procedure for the fabrication of spiral Pebax membrane elements is discussed in Section VI.7.

Table VI.1 Characteristics of Membrane Module Configurations

<table>
<thead>
<tr>
<th>Module Type</th>
<th>Application</th>
<th>Manufacturing Cost</th>
<th>Resistance to Fouling</th>
<th>Pressure Drop</th>
<th>Suitability to High-pressure Operation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hollow Fiber</td>
<td>GS, RO</td>
<td>Very Low</td>
<td>Very Poor</td>
<td>High</td>
<td>Yes</td>
</tr>
<tr>
<td>Spiral Wound</td>
<td>GS, RO, UF, MF</td>
<td>Low</td>
<td>Moderate</td>
<td>High</td>
<td>Yes</td>
</tr>
<tr>
<td>Plate &amp; Frame</td>
<td>ED, PV</td>
<td>High</td>
<td>Good</td>
<td>Low</td>
<td>No</td>
</tr>
<tr>
<td>Tubular</td>
<td>UF, PV</td>
<td>High</td>
<td>Very Good</td>
<td>Low</td>
<td>No</td>
</tr>
<tr>
<td>Capillary</td>
<td>UF, MF</td>
<td>Moderate</td>
<td>Good</td>
<td>Moderate</td>
<td>No</td>
</tr>
</tbody>
</table>

GS: Gas Separation; RO: Reverse Osmosis; UF: Ultrafiltration; MF: Microfiltration
PV: Pervaporation; ED: Electrodialysis
Figure VI.1. Pictorial Representation of Different Types of Membrane Modules
VI.6. MEMBRANE SELECTION AND FABRICATION FOR SCALE-UP

Pebax 2533 membrane was chosen for scale-up and commercialization due to its high permeability and selectivity, excellent mechanical strength and film-forming ability; above all the properties, it can be cast or coated as defect-free ultra-thin TFC films to achieve the maximum flux. However, some of the other membranes developed could not be scaled-up either due to the formation of defects in asymmetric sheet form (cellulose acetate) or due to the unsuitability to high-pressure operation of the hollow fibers (Matrimid). Moreover, all the polyimides including Matrimid were expensive polymers. Due to lack of proper solvent-nonsolvent systems, sulfonated polymers could not be cast as asymmetric films and their lack of adhesion to any of the ultraporous substrates rendered them unsuitable for conversion into TFC membranes. Some of the solvents used for polymer casting, were aggressive and either dissolved the substrates (eg. chloroform for PES) or caused their slow degradation or cracking (eg. m-cresol for Pebax 1657). However, these drawbacks were not encountered with Pebax 2533 membranes, which were cast using isobutanol solvent.

For scale-up into spiral modules, dip-coating technique was employed instead of the normal casting procedure, to obtain an effective Pebax thickness as low as 2 μm on the TFC membrane to derive the maximum possible flux. The conditions used for membrane fabrication by dip coating are presented in Table VI.2. The ultraporous substrate made of PVDF, PES or PSF was first prepared on a non-woven fabric by solution casting from DMF solutions followed by phase inversion in a water bath. After drying, the substrate was dipped in a bath containing 6 % (w/v) Pebax solution.
in isobutanol for 5 min. The solvent was evaporated by heating in an oven at 110°C for 5 min followed by solvent exchange in a hexane bath for 5 min to remove the residual isobutanol solvent present in the membrane matrix.

VI.7. FABRICATION OF SPIRAL GAS SEPARATION ELEMENT OF PEBAX 2533

The spiral wound element (Fig. VI.2) is the most economical configuration in terms of accommodating maximum surface area of the membrane in a given volume (900 m²/m³). The spiral Pebax element fabricated in-house consisted of essentially five components: (i) perforated permeate central tube, which collects the permeate, (ii) permeate spacer, which carries the permeate to the central tube, (iii) membrane, which is the active separation media of 0.2 m² effective area, (iv) feed spacer that separates two membrane leaves, facilitating the flow of feed gas across the membrane, and (v) gas seal (brine seal) to prevent the bypass of feed in membrane housing. Glue was applied over the carrier on three sides. The glue line was at the borders having a width of around 1.5". TFC membrane was folded over in half and inserted into the region where the carrier meets the central tube.
Table VI.2. Optimized Conditions for Scale-up of Membrane by Dip-coating Method

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Optimized Material/Condition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Membrane Material Selected</td>
<td>Pebax 2533</td>
</tr>
<tr>
<td>Solvent for Dissolution of Polymer</td>
<td>Isobutanol</td>
</tr>
<tr>
<td>Concentration of Pebax Solution in Bath</td>
<td>6 % (w/v)</td>
</tr>
<tr>
<td>Substrate used for Dip-coating</td>
<td>Ultraporous PSF/PES/PVDF</td>
</tr>
<tr>
<td>Duration of Dip-coating of Substrate in Bath</td>
<td>5 min</td>
</tr>
<tr>
<td>Coating Thickness</td>
<td>1–2 μm</td>
</tr>
<tr>
<td>Mode of Solvent Evaporation</td>
<td>Heating at 110 °C for 5 min</td>
</tr>
<tr>
<td>Essential Post Treatment</td>
<td>Solvent Exchange in Hexane Bath</td>
</tr>
<tr>
<td>Modular Configuration</td>
<td>Spiral Wound</td>
</tr>
<tr>
<td>Module Dimensions</td>
<td>1.5&quot; × 15&quot;</td>
</tr>
<tr>
<td>Membrane Area per Element</td>
<td>0.20–0.25 m²</td>
</tr>
</tbody>
</table>

The membrane was wound along with the carrier cloth after inserting the feed spacer between the folded elements, thereby separating the two membrane surfaces from each other. In case of a small element, only one membrane envelope is wound as in the present case. Once the diameter is reached, an adhesive tape is wrapped around to keep the element from opening up. The module was then kept aside overnight for the glue to harden or cure. Next day, the sides of the element were trimmed and then the element was finished and readied for testing after connecting to the feed gas cylinder. Figure VI.2(a) represents the design drawing of the miniature spiral Pebax 2533 module having an effective membrane area of 0.2 m². The total length of the permeate central tube is 15", whereas the wounded membrane part is 11".
long. The outer diameter of the permeate tube is 0.75", whereas the maximum diameter of the element is 1.4" at any point.

The module was housed in a stainless steel (SS 316) pressure vessel, which consists of a cylindrical tube of 14" length, 1.5" internal diameter (ID) and a thickness 0.6" with the flange fittings of 5.6" diameter welded at both ends. The gas seal was slipped onto the element and then taped to it. The module was then inserted into the housing. Detachable SS 316 end flanges of matching dimensions were fixed at each end by means of 6 pairs of nuts and bolts. The thickness of each flange was 0.4" such that the total length of the housing is 15.6". Two small O-rings, each of ID equal to outer diameter (OD) of permeate central tube were provided in the internal grooves of the end flanges for a leak-tight arrangement, that prevented the mixing of feed and permeate gases. Figure VI.2(b) shows the arrangement of spacers and TFC membrane sheet on the perforated permeate tube. Figure VI.2(c) is the actual photograph of the module prepared in this work with a pressure vessel.
Figure VI.2. Schematic of Spiral Wound Pebax 2533 Gas Separation Element: (a) Design Drawing, (b) Blow-up and (c) Photograph of Module with Housing
VI.8. SELECTION OF ULTRAPOROUS SUBSTRATE

VI.8.1. Importance of the Support Layer (Substrate)

The support layer is very crucial in determining the performance of TFC membrane. Apart from its inertness, the tightness and pore structure of this layer plays a major role in determining the overall permeability and selectivity of TFC membrane, which consists of three layers as shown in Fig.VI.3. Three different polymeric materials were chosen for making the substrate during upscaling study viz, polyvinylidene fluoride (PVDF), polyethersulfone (PES) and polysulfone (PSF), each having different molecular weight cut off (MWCO).
Figure VI.3. Three-Layered Structure of TFC Membrane within a Spiral Module

Figure VI.4. (a) Miniature Pebax 2533 Spiral Modules with Different Support Layers of PVDF, PES and PSF; (b) Cross-section of a Module of 0.25 m² Membrane Area
VI.8.2. Results with Pebax Modules Coated on Different Substrates

Table VI.3 describes the results with Pebax 2533 membrane cast on three different support layers by machine with a minimum casting thickness in each case in order to achieve the maximum possible permeance without compromising on the selectivity.

Table VI.3. Results with Pebax Spiral Membrane Module Prepared using Different Support Materials (Pressure: 60 kg/cm²)

<table>
<thead>
<tr>
<th>Type of Support</th>
<th>MWCO of Support (Daltons)</th>
<th>Pebax Skin Thickness (μm)</th>
<th>Permeance (GPU)</th>
<th>Pure Gas Permeability (Barrer)</th>
<th>Ideal Selectivity $a' = K_{CO_2} / K_{CH_4}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>CO₂</td>
<td>CH₄</td>
<td></td>
</tr>
<tr>
<td>PVDF</td>
<td>10000</td>
<td>8</td>
<td>7.8</td>
<td>0.36</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>CO₂</td>
<td>CH₄</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>62.4</td>
<td>2.9</td>
<td></td>
</tr>
<tr>
<td>PES</td>
<td>10000</td>
<td>2</td>
<td>29.1</td>
<td>1.25</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>58.2</td>
<td>2.5</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>23.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PSF</td>
<td>5000</td>
<td>2</td>
<td>28.0</td>
<td>0.95</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>56.0</td>
<td>1.9</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>29.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The data presented in Table VI.3 above table show that high permeance could be achieved by decreasing the membrane thickness without compromising on selectivity. Resistance of the support layers appears to play a major role in determining the selectivity of the skin layer. PSF support seems to be the most suitable, since a relatively higher selectivity of 29.5 could be obtained at a permeability of 56.0 Barrers, which is comparable to those obtained using other supports. The study also revealed that even though PSF was the tightest support, it was inert to CO₂ permeation by not forming a resistance layer. The adhesion between Pebax and PSF layers was satisfactory and a 2 μm thick membrane could be cast on this support without the formation of any defects.
The Pebax layer on PVDF support could not be brought down below 8 μm due to defects that can be attributed to the stretching process undergone by the TFC membrane coated on to PVDF support during the machine-based spiral winding. The stretching may cause the thickness of the skin layer at the edges to reduce just a bit, which causes a rise in flux. Maintenance of the separation factor (reproducibility) over a long-term operation was also verified experimentally by testing the spiral module over a period of 4 months at time intervals of one week each.

VI.9. GENERATION OF DESIGN DATA

VI.9.1. Studies with CO2/CH4 Mixtures

Table VI.3 presents pure gas permeability and ideal selectivity data, but in actual practice with multi-component mixtures, the CO2 selectivity is lower due to coupling effects and competitive permeation of other gaseous species. In this regard, experiments with CO2/CH4 binary mixtures were conducted at a pressure of 60 kg/cm², which is usually the pressure at which natural gas is available in oil and gas fields. The effective skin thickness of Pebax 2533 membrane on PSF support was 2 μm. As the feed CO2 concentration varied from 2 to 20 mol %, CO2 permeability increased from 40 to 49 Barrers and selectivity enhanced from 19.2 to 26.7 as shown in Fig.VI.5. It is worth mentioning that average permeability and selectivity over the crucial feed concentration range of 5 % to 2 % CO2 is 42 Barrers and 21, respectively, which is potentially very promising for natural gas purification. It may be recalled that pure CO2 permeability through Pebax 2533 membrane was 56 Barrers with an ideal selectivity of 29.5, which were marginally higher.
VI.9.2. Studies with Pure H₂S Gas

Since H₂S cylinders were not available at a pressure of 60 kg/cm² and mixture gas cylinders containing H₂S destabilize rapidly, experiments were carried out with pure H₂S having a maximum feed pressure of 30 kg/cm² (Fig. VI.6).

Figure VI.5. Results with Spiral Module of Pebax/PSF Composite Membrane for CO₂/CH₄ Gas Mixtures (Feed Pressure: 60 kg/cm²)
The pure H\textsubscript{2}S permeability increased from 122.6 to 146.7 Barrers whereas pure CH\textsubscript{4} only marginally improved from 1.53 to 1.72 Barrers, indicating increasing sorption of the polar H\textsubscript{2}S gas in the membrane with rising pressure. The H\textsubscript{2}S/CH\textsubscript{4} ideal selectivity ranged from 80.1 to 85.3. This study shows that H\textsubscript{2}S could be simultaneously removed from natural gas to an extent greater than even CO\textsubscript{2} using Pebax 2533 membrane.

VI.10. DEVELOPMENT OF THEORY FOR MATHEMATICAL MODELING

VI.10.1. Mathematical Modeling of Gas Permeation

Separation of gases by selective permeation through non-porous polymeric membranes is an important unit operation. Mathematical modeling followed by computer programming of this operation is of utmost significance for predicting the
performance of gas permeators equipped with commercially viable membranes [11]. The separation achieved in a membrane process is not a function of membrane characteristics alone, as it can be considerably enhanced by process design techniques, appropriate selection of operating conditions, flow regimes, etc. [12], which makes the simulation all the more useful and important.

Modeling and simulation of gas permeation has attracted a lot of research interest in the past. Weller and Steiner [13,14] examined the effects of perfect mixing and crossflow regimes on the extent of separation and membrane area requirements. Other researchers [15-17] have thoroughly compared the two commercially important flow models: cocurrent and countercurrent. Since there has been renewed research activity on design considerations like configuration, flow pattern, pressure drop effects, membrane characteristics and channel dimensions, which dominate the module performance [18-21]. However, a detailed review of the literature reveals only simulation results, whereas the actual computer program utilized for carrying out such calculations is lacking.

In the present study, the formulation and working of a computer program in Microsoft Excel based on the plug flow model is presented to determine the membrane area requirement and exit streams' compositions for given Pebax membrane characteristics, feed capacity and operating conditions. For binary mixture separation assuming negligible pressure drop from the point of feed inlet to reject (product) outlet, an excel program would suffice. For multicomponent feed mixtures with the assumption of significant pressure drop at the feed side, programming in 'C' language is essential.
VI.10.2. Derivation of Governing Equations

In the theoretical analysis, mechanistic aspects of transport process are not considered. It is assumed that the membrane transport and permeation properties are known. The goal is then to compute and predict the overall macroscopic separation based on the known local rates of transport or to design the module for the required extent of separation. Additionally, a steady state operation is assumed and no temporal variation inside the membrane module is considered.

Table VI.4. Definitions and Units of Some Terms Constituting the Governing Equations

<table>
<thead>
<tr>
<th>Term</th>
<th>SI Units</th>
<th>CGS Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>Permeation rate ((R))</td>
<td>Moles/sec</td>
<td>std cm(^3)/sec</td>
</tr>
<tr>
<td>Permeation flux ((J))</td>
<td>Moles/sec-M(^2)</td>
<td>std cm(^3)/sec-cm(^2)</td>
</tr>
<tr>
<td>Permeance ((Q))</td>
<td>Moles/sec-M(^2)-pa</td>
<td>std cm(^3)/sec-cm(^2) cmHg</td>
</tr>
<tr>
<td>Permeability ((P))</td>
<td>Moles-M/sec-M(^2)-pa</td>
<td>(std cm(^3))-cm/sec-cm(^2) cmHg</td>
</tr>
</tbody>
</table>
Table VI.4 summarizes important terms and their units in SI and CGS systems. The common unit of permeability is 'Barrer' defined as 1 Barrer = $10^{-10}$ (std cm$^3$) (cm)/(sec·cm$^2$·cmHg).

Rate of permeation of species $i = \frac{\text{Permeability of species}}{\text{effective membrane thickness}} \times [\text{Membrane area}] \times [\text{effective partial pressure difference of } i \text{ across membrane}]

(VI.1)

The rate of permeation is a point function, but the ratio of permeability and membrane thickness is called the permeance; then the above equation can be written as:

Rate of permeation = Permeance $\times$ Membrane area $\times$ Pressure difference \hspace{1cm} (VI.2)

The above equation can also be written as:

Permeation flux = Permeance $\times$ Pressure difference \hspace{1cm} (VI.3)

For a differential area, $dA$, Eq.(VI.2) for the flow rate of the faster permeating CO$_2$ becomes:

$$dr_i = \frac{P_r}{t_m} (dA)(\Delta p_i) = Q_i (dA)(\Delta p_i)$$ \hspace{1cm} (VI.4)

In case of nonporous homogeneous membrane, $(\Delta p_i)$ is the partial pressure difference between the feed gas and the permeate gas and hence,

$$dr_i = Q_i (dA)(P_r x_i - P_p y_i) = Q_i (dA)P_r (x_i - y_i)$$ \hspace{1cm} (VI.5)
Here, the stage pressure ratio, $R$ is given by

$$R = \frac{P_p}{P_f} \quad (VI.6)$$

The relative rates of permeation of two species, $i$ and $j$, can be expressed as:

$$\frac{d r_i}{d r_j} = \frac{Q_i}{Q_j} \frac{\Delta p_i}{\Delta p_j} \quad (V.17)$$

The quantity $(Q_i/Q_j)$ is called the selectivity $(S)$ of species, $i$ over $j$, for the given membrane. The selectivity and permeance are the two most important parameters of the commercial membranes.

![Figure VI.7. Material Balance Across the Membrane Separation Stage](image)

**Figure VI.7. Material Balance Across the Membrane Separation Stage**

The material balance over a typical membrane stage shown in Fig.VI.7 is given as follows:

$$F = L + V; \quad F_Z = L_x + V_y \quad (VI.8)$$

There are three terms used to characterize the membrane stage i.e., stage cut $\theta$, component recovery in permeate, $\phi$ and component recovery in residue, $\phi$. Thus,

$$\theta = \frac{V}{F}; \quad \phi = \frac{V_y}{F_Z}; \quad \phi_i = \frac{L_x}{F_Z} = 1 - \phi$$

$$(VI.9)$$

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Quite often, another term called enrichment factor, $e_i$, is used, which can be defined as:

$$e_i = \frac{y_i}{x_i}$$  \hspace{1cm} (VI.10)

Using these definitions, one can deduce:

$$\theta = \frac{z_i - x_i}{y_i - x_i} \hspace{0.5cm} \phi_i = 1 - \theta_i = e_i \theta_i$$  \hspace{1cm} (VI.11)

**VI.10.3. Cross Flow Model**

The commercial exploitation of membrane-based gas separation technology was accelerated due to the preparation of dense asymmetric membranes, which have a skin on one side with a backing of graded porosity underneath. However, this structure is difficult to model. In practice, it is modeled assuming a nonporous layer, which offers all the permeation resistance backed by a highly open porous structure through which there is no permeation resistance at all. To determine the governing equations for crossflow model, it is necessary to first derive the equations for complete mixing model. Notice that Weller and Steiner [13] were the first to derive the analytical expression for the enrichment of a binary mixture in a single stage membrane process with complete mixing of the feed flow as shown in Fig.VI.8.
In a completely mixed flow, gas phases on either side of the membrane are completely mixed. As a result, there are no special gradients of concentrations and only algebraic equations need to be solved. The mole fraction of the component CO$_2$ on the feed side of the membrane is constant throughout the module and is the same as the residue mole fraction. For a binary system of CO$_2$ (1) and CH$_4$ (2), the relationship between the molar flux ($J$), permeance ($Q$) and membrane area ($A$) is given by [22]:

$$J_1 = Q_1(P_1 - P_2)$$  \hspace{1cm} (VI.12)

where $J_1$ flux of the component CO$_2$. Similarly, flux of the second component CH$_4$ component is given by:

$$J_2 = Q_2[P_1(1 - x) - P_2(1 - y)]$$  \hspace{1cm} (VI.13)
The stage pressure ratio and selectivity are defined as follows:

\[
\frac{P_2}{P_1} = R; \quad \frac{Q_1}{Q_2} = S
\]  
(VI.14)

The permeate composition for a faster permeating component CO\textsubscript{2} can be defined in terms of the flux as:

\[
y = \frac{J_1}{J_1 + J_2}
\]  
(VI.15)

Inserting Eqs. (VI.14) and (VI.15) in Eq. (VI.17) gives:

\[
y = \frac{S(x - Ry)}{S(x - Ry) + [(1 - x) - R(1 - y)]}
\]  
(VI.16)

The above equation can be rearranged to give the quadratic equation of the type:

\[
(S - 1)y^2 + (1 - S) - \left[ \frac{1}{R} - \frac{x(S - 1)}{R} \right]y + \frac{Sx}{R} = 0
\]  
(VI.17)

Equation (VI.17) can be solved for \(y\) to obtain \(x-y\) plot that is similar to the one shown in Fig.VI.9.
From Eq. (VI.8), the material balance over a single stage is given as:

\[ F = L + V \; ; \; F_x = L_x + V_y \]  

(VI.18)

Solving for \( y \), we obtain

\[ y = \frac{F_x}{V} \left( \frac{F - V}{V} \right) x \]  

(VI.19)

The stage cut is then calculated as:

\[ \theta = \frac{V}{F} = \frac{z - x}{y - x} \]  

(VI.20)

The line given by Eq. (VI.20) passes through \((z, z)\) with a slope given by

\[ -\left( \frac{F - V}{V} \right) = \left( \frac{1}{\theta} - 1 \right) \]  

(VI.21)
Equation (VI.5) can be rearranged to obtain an expression for membrane area:

\[
A = \frac{J_1}{Q_i P_i (x - R_y)} = \frac{V_y}{Q_i P_i (x - R_y)} \quad \text{(VI.22)}
\]

For cross flow configuration, the membrane module can be divided into infinite number of small modules with complete mixing case prevalent in each small individual module. This is known as the Weller and Steiner Case-II [13]. Hence, in this study, simulations were performed by dividing the single stage into infinite number of elemental stages and each elemental stage is assumed as a completely mixed flow model [22]. A schematic diagram of differential or step-wise method for cross flow mode is given in Fig.VI.10.

![Figure VI.10. Schematics of Differential or Step-wise Method for Cross Flow Mode](image)

When using a stepwise solution to determine the separator performance, Eqs (VI.23) and (VI.24) were used to determine the average CO₂ concentration in feed and permeate side, respectively:
\[ x_{avg} = \frac{x_i + x_j}{2} \]  
\[ y_{avg} = \frac{y_i + y_j}{2} \]  
\[ (VI.23) \]
\[ (VI.24) \]

The material balance for CO\textsubscript{2} can be rewritten as:

\[ L_i x_i = L_j x_j + \Delta V y_{avg} \]  
\[ (VI.25) \]

The above equation upon rearrangement gives:

\[ L_i (x_i - x_j) = \Delta V (y_{avg} - x_j) \]  
\[ (VI.26) \]

Solving for \(\Delta V\), we get

\[ \Delta V = \frac{L_i (x_i - x_j)}{(y_{avg} - x_j)} \]  
\[ (VI.27) \]

The area of the increment is found from the differential form of flux-permeability relationship by knowing permeate composition and permeate flow as [22]:

\[ \Delta A = \frac{\Delta V y_{avg}}{J_1} \]  
\[ (VI.28) \]
The flux for CO₂ can be calculated in terms of average concentrations:

\[ J_1 = Q_1 \left( P_x \text{avg} - P_y \text{avg} \right) \]  \hspace{1cm} (VI.29)

The total permeate flow rate and CO₂ concentration are given as:

\[ V = \sum \Delta V \]  \hspace{1cm} (VI.30)

\[ y = \frac{\sum \Delta V \text{avg}}{\sum \Delta V} \]  \hspace{1cm} (VI.31)

Total area, \( A \) of the membrane is calculated by the summation of incremental areas

\[ A = \sum \Delta A \]  \hspace{1cm} (VI.32)

**VI.10.4. Process Design Options and Schemes**

Two major factors in process design are product purity and product recovery. The desired product can be either permeate or retentate streams. Examples of the latter are nitrogen generation from air (removal of O₂ from Air), dehydration of natural gas (removal of H₂O from hydrocarbons), sour gas treating (removal of H₂S and CO₂ from hydrocarbons), land fill gas upgrading (removal of CO₂ from CH₄), etc. Examples of the former category are hydrogen recovery from the refinery stream, production of oxygen enriched air, helium recovery, etc. The process design is generally influenced by the separation goal. The permeation process is also influenced strongly by the operating conditions such as temperature, feed pressure, permeate pressure and the scheme of modular arrangement.
VI.10.5. Process Schemes

A single stage system greatly inhibits the ability of the process engineer to meet the pre-selected performance and process criteria. There exists a wide variety of possible membrane process designs as shown in Fig.VI.11 that can be employed to solve a particular gas separation problem. In all the usual process schemes displayed in Fig.VI.11 more than a single stage is employed to improve the product recovery through the use of a recycle loop. Portions of the permeate stream are pressurized and recycled to the feed stream to provide the additional degree of separation that dramatically improves the product recovery and the overall process economics.
Figure VI.11. Different Process Schemes for Enhanced Performance of Gas Permeators
In Fig.VI.11(a), the permeate from stage 1 is compressed and sent to stage 2 to form a cascade arrangement. The reject from stage 2 is recycled to the original feed stream. Figure VI.11(b) shows the permeate from stage 2 being compressed and recycled to the feed inlet point of stage 1. Figure VI.11(c) exhibits a three-stage configuration, wherein the permeate from stage 2 is compressed and sent as the feed to stage 3 with the reject of the last stage getting recycled to the feed inlet stream of stage 2. Therefore, stage 1 becomes a pre-membrane stage to execute bulk removal, while stage 2 is for purification and stage 3 is for recovery. Thus, several process schemes can be employed depending upon the nature of feed and desirable product characteristics. For the current study, a variation of Fig.VI.11(a) is employed with the difference being that the feed is split into different proportions before entering each individual stage with no recycle of stage 2 reject. This scheme is discussed in detail in Section VI.11.

VI.11. CONSTRUCTION, DESCRIPTION AND EXECUTION OF EXCEL SIMULATION PROGRAM FOR THE GENERATION OF DESIGN DATA

VI.11.1. Construction and Description of the Simulation Program

The excel worksheet and program given in Fig.VI.12 have been prepared on the basis of gas separation data obtained with the spiral wound Pebax 2533 membrane (Fig. VI.5). A two stage process scheme is provided underneath the worksheet, which shows the sour natural gas feed F1 coming at a rate of 100 m³/h and getting split into two streams, F2 and F3 in the volumetric ratio of 7:3 for the two individual stages. The ratio
can be changed to 1:1 or any other, depending upon the feed composition, operating conditions and desirable characteristics of the product.

To minimize the hydrocarbon (CH$_4$) losses, permeate P1 of stage 1 is compressed to 60 kg/cm$^2$ and combined with F3 to form F4, which is the feed to stage 2. The permeate P2 is sent to flare. The retentate (reject) is the product in this operation and retentates R1 and R2 from the two stages are combined to form the final product R3, which is sent through a pipeline for storage and utilization. The excel worksheet shows R3 to contain $< 2 \%$ CO$_2$. The worksheet shows data in green, which represent the membrane characteristics, whereas purple represents the operating conditions. In these two colored areas, there are cells in white, which represent the input variables that can be changed as per the requirement. These include the permeability values for CO$_2$ and CH$_4$ as well as the effective thickness of TFC membrane skin. Similarly the operating conditions such as feed capacity, stage cut, feed and permeate pressures are represented in other white cells. The colored cells represent data, which cannot be altered.

Model equations are provided as expressions in these colored individual cells to compute the output in terms of F3, P1, P2, R1, R2, R3 as well as membrane area requirement and CH$_4$ losses. Permeate composition $y$ is a function of stage cut, $\theta$ and is given by a polynomial equation, where $y = K_1 + K_2 \theta + K_3 \theta^2$, which can be solved by regression using the inverse matrices. The values of $K_1$, $K_2$ and $K_3$ for the two stages are provided at the bottom of the excel program.
VI.11.2. Application of Excel Program to Evaluate the Effect of Operating Parameters

The effect of each operating parameter on performance of commercial gas permeator was evaluated by keeping other parameters constant.

VI.11.2.1. Stage Cut

Stage cut ($\theta$) is defined as the quantity of feed allowed to permeate during a particular stage or simply as the ratio of permeate rate to feed rate. The effect of stage cut was evaluated at constant feed capacity of 100 m$^3$/h, feed composition of 95 % CH$_4$ + 5 % CO$_2$ and pressure of 60 kg/cm$^2$. Fig. 6.7.2 (a) represents the variation of membrane area requirement and product (reject) CH$_4$ concentration with the stage cut increasing from 0.01 to 0.25. The fraction 0.01 denotes that 1 % of the feed has permeated during a particular stage whereas 0.25 indicates that 25 % of the feed has permeated. The membrane area requirement increased from 3 m$^2$ to 137 m$^2$ whereas the product CH$_4$ concentration increased from 95.1 % to 99.2 %.

With increasing stage cut more surface area is required to bring about greater quantity of permeation through the membrane. Similarly, more and more quantity of CO$_2$ gets removed through the permeate resulting in increasing concentration of the rejected CH$_4$ gas which is the final product. However, the desired product concentration of at least 98 % CH$_4$ was reached at a stage cut of 0.15 for which the membrane area requirement was 64 m$^2$. These values were considered significant for designing a commercial permeator capable of processing natural gas at the rate of 100 m$^3$/h. The model (excel program) can give erroneous values for large $\theta$ values (> 0.25) since the
CO$_2$ concentration in feed is low (only 5 %). However, for greater feed CO$_2$ concentrations, such as 40 mol %, $\theta$ of 0.5 and above will be accepted by the program.
Figure VI.12. Excel Program for Design of Gas Permeator for CO₂/CH₄ Separation
Figure VI.13. Simulated Effect of Stage Cut on (a) Membrane Area Requirement and Product CH$_4$ Concentration; (b) CH$_4$ Loss and CO$_2$ Permeate Concentration
Figure VI.13(b) depicts the effect of stage cut on % CH₄ loss as well as CO₂ concentration in the permeate. Expectedly, with an increasing stage cut, there was increasing loss of CH₄ in permeate from 0.15 to nearly 8 %. The CO₂ concentration in the second stage permeate initially increased from 52.4 to 60.3 % in the stage cut range of 0.01–0.1, after which CO₂ concentration was dropped to 56.2 % and finally, to just 36.1 % at \( \theta = 0.25 \). Therefore, lower stage cut values must be preferred for industrial applications. It is worth mentioning that the CH₄ loss at an optimized \( \theta \) of 0.15 was only 2.8 %.

VI.11.2.2. Stage Pressure Ratio

The permeate pressure was close to atmospheric pressure, which meant that the stage pressure ratio was equal to feed pressure, which was used for plotting Fig. VI.14. The stage cut was maintained constant at an optimized value of 0.15. With the feed pressure increasing from 10 to 60 kg/cm², membrane area requirement was reduced from 494 to 64 m² due to increasing flux [see Fig. VI.14(a)]. The CH₄ concentration in the product was improved from 97.2 to 98.3 %. Figure VI.14(b) shows that the corresponding CH₄ losses was also reduced from 3.93 to 2.8 % with the permeate CO₂ concentration increasing from 38.6 to 56.2 % due to a better separation efficiency.

VI.11.2.3. Feed Capacity

Figure VI.15(a) shows the effect of feed capacity on membrane area requirement and product concentration at a constant feed composition of 5 % CO₂, pressure of 60 kg/cm² and stage cut of 0.15. For feed capacities ranging from 50 to
500 m$^3$/h, the area requirement increased linearly from 32 to 321 m$^2$. Expectedly, the exit stream compositions and CH$_4$ losses did not undergo any changes with the CH$_4$ concentration in the product remaining constant at 98.3 %, CO$_2$ concentration in permeate at 56.2 % and CH$_4$ losses through the permeate at 2.8 %.

Figure VI.14. Simulated Effect of Feed Pressure on (a) Membrane Area Requirement and Product CH$_4$ Concentration; (b) CH$_4$ Loss and Permeate CO$_2$ Concentration

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Figure VI.15. Simulated Effect of (a) Feed Capacity on Membrane Area Requirement and (b) Feed CO<sub>2</sub> Concentration on Membrane Area and Stage Cut Requirements
Figure VI.15(b) displays the effect of feed CO$_2$ concentration on stage cut and membrane area requirement to achieve a final product concentration of $< 2$ % CO$_2$ as per pipeline specification. The membrane characteristics were kept constant to avoid any confusion. Figure VI.15(b) shows that membrane area required to bring down the CO$_2$ concentration from a range of 5-20 mol % varied from 64 to 141 m$^2$, with the corresponding stage cut increasing from 0.15 to 0.43.

VI.12. DESIGN OF A COMMERCIAL PLANT FOR NATURAL GAS PURIFICATION

VI.12.1. Design Basis

A typical composition of natural gas stream available in an Indian oil and gas field given in Table VI.5 was considered for designing a skid mounted commercial gas separation plant of 100 m$^3$/h capacity. It was assumed that gas was available at a pressure of 60 kg/cm$^2$ and a temperature marginally above the ambient temperature 40°C. The product desirable was a gas mixture containing $< 2$ mol % CO$_2$ and $< 4$ ppm H$_2$S.

Table VI.5. Typical Composition of Natural Gas Considered for Design

<table>
<thead>
<tr>
<th>Feed Component</th>
<th>CH$_4$</th>
<th>C$_2$–C$_3$</th>
<th>C$_4$–C$_5$</th>
<th>C$_6$</th>
<th>CO$_2$</th>
<th>N$_2$</th>
<th>H$_2$S</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mole %</td>
<td>81.4</td>
<td>11.2</td>
<td>2.1</td>
<td>0.1</td>
<td>5.1</td>
<td>0.1</td>
<td>100 ppm</td>
</tr>
</tbody>
</table>

The most significant aspect in membrane plant design is membrane area required for a particular application. From the results obtained using Excel Program, especially Fig.VI.13(a), the membrane area of 64 m$^2$ was required with a stage cut of
0.15 to treat the gas mixture containing 5 % CO₂ to a level of 2 %. The largest commercial spiral module that can be manufactured in India has the dimensions of 8" dia × 40" length and an area of 36 m². Thus, two such modules would provide a total membrane area of 72 m², which suits to the present requirement, since the plant is generally designed to handle 10–20 % greater capacity than the specified one. One pressure housing of the corrosion resistant reinforced plastic (FRP) material would be provided for each module.

VI.12.2. Process Description

The process flow diagram (PFD) of the commercial plant capable of treating 100 m³/h of sour natural gas is provided in Fig.VI.16. The heart of the system is the Pebax 2533 membrane incorporated in the form of two spirally wound modules of 8" diameter and 1 m length that are connected in parallel with respect to each other. Modules are housed in cylindrical FRP pressure vessels. Natural gas, assumed to be free of any higher hydrocarbon condensate, enters the pretreatment section through an isolation valve at the rate of 100 m³/h and a pressure of 60 kg/cm². It is fed initially to a micron filter for the removal of dust particles that could damage the membrane at high pressure. The dust-free gas is then sent to a glycol dehydration plant to remove the moisture, which could swell and plasticize the hydrphilic Pebax membrane. The dry gas then enters the membrane assembly.

Each membrane module generates a product (reject) stream lean in acid gases, which are combined into a single stream containing CO₂ concentration within the acceptable limit (2 mol %) with its pressure close to the feed pressure (~ 59 kg/cm²). Since H₂S is present in low concentrations in the feed, its level in the product may not
reach the pipeline specification of < 4 ppm due to the concentration polarization of other gases near the membrane surface. Hence, the product is sent through an adsorbent bed containing ZnO pellets, which bring down the \( \text{H}_2\text{S} \) concentration to pipeline specification. The product enriched in \( \text{CH}_4 \) and other hydrocarbons is then sent to the storage at the rate of 85 m\(^3\)/h for usage as a fuel. The permeate streams, rich in acid gases, with a pressure around atmospheric (1 kg/cm\(^2\)) are combined and released to flare (burning) at the rate of 15 m\(^3\)/h due to a stage cut of 0.15.

An on-line safety valve (not shown in the figure) with an outlet to flare is provided the to protect commercial plant from a sudden surge in the inlet pressure. Provisions for collection of samples from feed, permeate and product lines are also made. Currently, a compressor for recycle of permeate from the first membrane module (stage 1) to feed inlet of second module (stage 2) is not provided, which would become a vital accessory in future to minimize the hydrocarbon losses. Instead of a ZnO adsorbent bed, an amine absorption column could also be used to constitute the hybrid process, wherein membranes are used for bulk removal of acid gases and amine absorption to execute the final clean-up.
VI.13. ECONOMIC ESTIMATE

Table VI.6 shows that the capital investment required to set up a membrane plant capable of treating 100 m$^3$/h of natural gas containing 5 mol % CO$_2$ is approximately Rs. 21.3 Lakhs. The dimensions and material of construction (MOC) of each item is also given. Pebax of 72 m$^2$ area costs about Rs. 2.5 Lakhs at the rate of Rs 3500/- per square meter due to the sophisticated dip coating method employed. However the pretreatment stage comprising of micron filter and glycol dehydration unit amounts to Rs. 2.5 Lakhs, whereas ZnO adsorbent column for post treatment incurs an expenditure of Rs. 1.3 lakhs.
Table VI.6 Capital Cost Break-up for Commercial Membrane Plant for Treating 100 m³/h of Natural Gas

<table>
<thead>
<tr>
<th>Item</th>
<th>Specification</th>
<th>MOC</th>
<th>Quantity</th>
<th>Total Price (Rs in Lakhs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Membrane Module</td>
<td>8&quot; dia x 40&quot; long Pebax polymer</td>
<td>2</td>
<td>2.5</td>
<td></td>
</tr>
<tr>
<td>Pressure Housing</td>
<td>Cylinder 8.2&quot; ID with End Flanges FRP</td>
<td>2</td>
<td>1.2</td>
<td></td>
</tr>
<tr>
<td>Micron Filter</td>
<td>4&quot; dia x 20&quot; long SS-Mesh in Carbon Steel Cylinder</td>
<td>1</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td>Glycol Dehydration Unit</td>
<td>4&quot; x 24&quot; Carbon Steel Cylinder</td>
<td></td>
<td>1.5</td>
<td></td>
</tr>
<tr>
<td>ZnO Bed</td>
<td>3 Kg adsorbent in 4&quot; dia x 20&quot; long Carbon Steel Cylinder</td>
<td>1</td>
<td>1.3</td>
<td></td>
</tr>
<tr>
<td>Instrumentation</td>
<td>Pressure and Temp. Instruments, Flow Meters, Panel, Wiring, Switches etc.</td>
<td>–</td>
<td>3.8</td>
<td></td>
</tr>
<tr>
<td>Valves</td>
<td>Suitable for 1&quot; Piping Stainless Steel 316</td>
<td>2 Dozens</td>
<td>5.0</td>
<td></td>
</tr>
<tr>
<td>Piping &amp; Fittings</td>
<td>1&quot; dia Piping with SS 316 Flange Fittings Stainless Steel 316 Sufficient for all Connections</td>
<td>5.0</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Grand Total: Rs. 21.3 Lakhs

Instrumentation inclusive of pressure and temperature indicators, flow meters, panel, wiring, switches, etc account for Rs 3.8 Lakhs, whereas valves, piping and fittings cost about Rs. 10.0 Lakhs.

The cost of crude natural gas is Rs. 3.5/- per cubic meter. The operating cost components include the costs for membrane replacement, regeneration of ZnO adsorbent, replacement of consumables used in pretreatment, besides chemicals for maintenance and utilities such as water, steam, etc. The cost for the present scheme excludes the compressor cost for the recycle of permeates. Assuming the membrane
life of 3 years, a periodic replacement of micron filter cartridge, glycol solvent and 
ZnO pellets every 3 months, the cost of replacement comes to about Rs. 6.0 Lakhs in 
3 years. A depreciation of 10 % and rate of interest of 8 % on capital would further 
incur an expenditure of Rs. 11.4 Lakhs, resulting in a total running cost of nearly Rs. 
17.4 Lakhs in 3 years (excluding cost of labor and hydrocarbon losses). The total 
volume of gas treated during the same time frame at the rate of 24 hours/day and 300 
working days in a year round would be $2.16 \times 10^6 \text{ m}^3$. Thus, the operating cost would 
be approximately Rs. 0.80 per cubic meter of natural gas treated.

VI.14. CONCLUDING REMARKS

Amongst the different modular configurations, both hollow fiber and spiral 
wound modules were suitable, but the latter was chosen due to the availability of the 
equipment for casting of sheet membranes by dip coating method as well spiral 
winding. Scale-up of the most promising Pebax membrane into spiral wound modules 
of 0.2-0.25 m$^2$ area were successfully carried out. Different ultraporous substrates 
were prepared for supporting Pebax 2533 layer with PSF substrate, which was found 
to be the best due to its tightness and uniform pore structure. Analyses of gas 
permeator arrangements showed that in actual industrial practice a single stage 
membrane separation may often be inadequate to achieve the target separation at the 
lowest possible cost, and therefore, a multi-stage separation scheme needs to be 
configured.

A step-wise method was carried out to find a solution for a crossflow model, 
wherein the membrane area was divided into numerous infinitesimal areas. For a feed 
rate of 100 m$^3$/h, composition of 95 % CH$_4$ + 5 % CO$_2$ and a pressure of 60 kg/cm$^2$, 

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the simulation program in Microsoft Excel exhibited that the desired product concentration of \( \geq 98\% \) CH\(_4\) could be reached at a stage cut of 15\% with a membrane area requirement of 64 m\(^2\). A commercial gas permeator of the same capacity was designed for the purification of natural gas to pipeline quality. Design based on the simulation results resulted in a process flow diagram consisting of two spiral wound membrane modules of 36 m\(^2\) area, each along with other pretreatment equipment, such as micron filter, glycol dehydration unit and post treatment system like ZnO adsorbent bed. An economic estimate showed that a capital investment of Rs. 21.3 Lakhs was required to set up the commercial plant, which is expected to incur an operating cost of Rs. 0.80 per cubic meter of gas treated. The low operating cost could be attributed to the fact that crude natural gas is already available at a high pressure (55-60 kg/cm\(^2\)) at the source, which provides the driving force for separation without using a compressor.
VI.15. NOMENCLATURE

\[ \begin{align*}
A & \quad \text{Total membrane area} \\
F & \quad \text{Feed volume} \\
J & \quad \text{Molar flux} \\
L & \quad \text{Retentate volume} \\
P_1, P_F & \quad \text{Feed pressure} \\
P_2, P_P & \quad \text{Permeate pressure} \\
\Delta p & \quad \text{Partial pressure gradient} \\
P_i & \quad \text{Permeability of CO}_2 \\
Q & \quad \text{Permeance} \\
r & \quad \text{Rate of flow} \\
R & \quad \text{Stage pressure ratio (downstream / upstream)} \\
S & \quad \text{Membrane selectivity} \\
\theta & \quad \text{Stage cut} = \frac{\text{permeate rate}}{\text{feed rate}} \\
t_m & \quad \text{Membrane thickness} \\
V & \quad \text{Permeate volume} \\
x & \quad \text{Concentration of CO}_2 \text{ in high pressure (feed) side} \\
y & \quad \text{Low pressure side (permeate) concentration CO}_2 \\
z & \quad \text{Concentration of CO}_2 \text{ in feed at the point of inlet}
\end{align*} \]
VI.16. REFERENCES


CHAPTER VI


