



## A Review of the Natural Gas Purification from Acid Gases by Membrane

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### HIGHLIGHTS

- A comprehensive comparison of CO<sub>2</sub> capture technology.
- Applications of membrane in gas separation.
- Models for a binary gas mixture.
- Industrial aspect of acid gas removal from natural gas by membranes.

### ARTICLE INFO

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### ABSTRACT

This study aims to shed light on natural gas as an important and promising energy source. This energy source is the fastest-growing source in the world due to the increasing global demand. In this paper, the rates of growth in global demand for natural gas according to the latest reports since 1984, as well as the gas specifications required for transport and storage, acid gases, including absorption, desorption, Cryogenic and separation by membranes, are discussed with the advantages and disadvantages of each method. Focusing are presented. In addition to the primary treatment processes that take place on the gas, the most important of which is the removal of acid gases. Processes for removing on the membrane separation process as the most promising process in this field and reviewing all the research that is discussed in details of this process.

## 1. Introduction

Natural gas (NG), as an available and clean energy source, functions a significant role in encountering the growing request for many fields such as power, industry, transport, and others [1]. Based on the Global Energy review 2019, international NG request is predicted to increase by 40% between 2018 and 2050, reaching around 200 quadrillion British thermal units (Btu) by 2050 [2]. As for the participation of natural gas compared to other energy sources, it increased to 24% in 2018, which is one of the fastest growth rates since 1984 [3]. The location, type, and depth of subterranean deposits and the geology of the area are the factors upon which the composition of gas produced from wells depends [4]. Natural gas from the wellhead leaves the reservoir with undesirable impurities such as water, hydrocarbons liquid, and acid gases. Therefore, raw natural gas must be treated to satisfy the pipeline gas specification, environmental limits, safe operating conditions, and the customer's requirements [5]. Corrosion of equipment and pipelines and the toxic nature of the gas result from the presence of carbon dioxide gas and hydrogen sulfide [6]. Natural gas consists mainly of methane at a ratio ranging between 75 to 90 %, in addition to different amounts of ethane, propane, pentane and harmful impurities. The limits of the proportions of components of natural gas needed before pumping gas into the pipeline network or gas liquefaction units are shown in Table 1 [7].

Gas Plants have an important and essential role in the oil and gas industry. The wanted end product, location of the plant, and nature of gas dictate the processes required. So, the main goal of gas processing is to obtain gas that meets specifications including heating value and the recovery of the maximum amount of NGLs (Natural Gas Liquids) and the sales requirements. Getting to the specifications of pipelines is done through several complex processes that take place at the head of the well. These processes include removing natural gas liquids, impurities and harmful compounds, and it is necessary to install equipment for these processes at or near the wellhead. (EIA, 2006). In the first stage, the large particles and sand are mainly removed using scrubbers, then the gas stream is sent to gas-liquid separators where liquid hydrocarbons are separated. The Separator vessel can be of two trends, vertical or horizontal, depending on the liquid-to-gas ratio and gas flow rates. After that, the gas is dried to prevent gas hydrate formation as well as to reduce corrosion. Acid gases are treated in the pre-last stage of the field operations where the acid gas like H<sub>2</sub>S and CO<sub>2</sub> are removed, along with other sulfur impurities. If the gas pressure exiting the acid gas removal unit does not match the design value, then it is entered into the pressure stage. [8, 9].

Several techniques can be employed for PCC. Some of these include absorption, membrane, adsorption and cryogenic separation as shown in Figure 1. Among these unit operations, absorption is the most mature and preferred technology. Membranes, on the other hand, have modular nature and low footprint [14].

### 1.1 Absorption Process

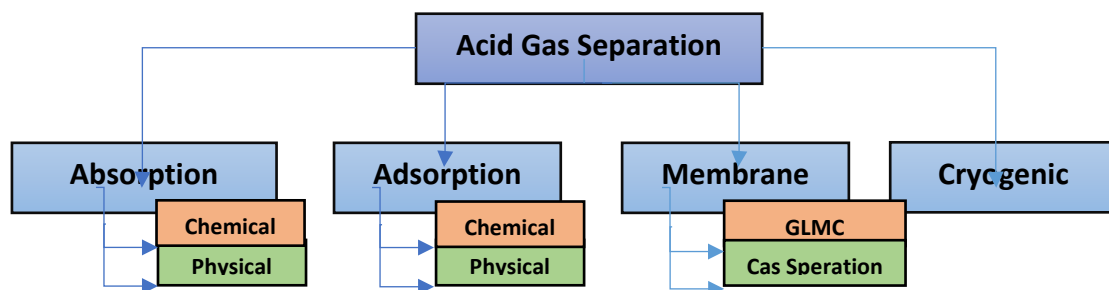
A long-time ago, this method was developed and applied to the commercial level to remove acid gas at natural gas processing and refineries. Through gas/liquid contacting surface area, acid gas is counter-currently contacted and absorbed by a chemical or physical solvent in the absorption tower. The increased contact area is accomplished through various designs, such as trays, packed bed, spray or other internals, for a tower. The initial criteria for choosing a solvent, whether it is physics or chemistry, first includes the acid gas concentration in the gas mixture, temperature, and required separation ratio. Criteria for careful selection between different types of solvents involves high gas solubility, selectivity, environmentally friendly, chemical and thermal stability, inexpensive and available, non-corrosive, low viscosity, non-flammable [15-17]. Operational problems include several issues such as high corrosion of equipment, low acid gas loading capacity, amine degradation, foaming, channeling, flooding, and no selective separation between CO<sub>2</sub> and H<sub>2</sub>S. As for environmental problems, solvent disposal causes environmental risks, also the large energy needed to regenerate the solvent comes from burning fuel that leads to an increase in CO<sub>2</sub> in the ambience. On the other hand, regenerating the solvent consumes a lot of energy. Also, the costs of buildup, operating and maintaining the absorption units are high. Finally, the size of the absorption towers, pumping equipment, piping system and the amount of solvent is large and directly proportional to the amount of CO<sub>2</sub> to be removed [18-20].

### 1.2 Adsorption Process

Depending on the nature of interactions, there are two types of adsorption, namely chemical adsorption and physical adsorption [21]. Important factors in adsorption include; (i) ease of regeneration of adsorbed acid gas, (ii) durability of adsorbent, (iii) selectivity of adsorbent for CO<sub>2</sub>, (iv) adsorption capacity and, (v) stability of adsorbent after several adsorption/desorption cycle [22]. Metal oxides, zeolites, activated alumina, silica gel, mesoporous silicates, ion-exchange resins, activated carbons, and other surface-modified porous media are among the conventional solid adsorbents [23]. The industrial implementation of these adsorbents for sour gas adsorption is restricted because these materials have a low adsorption affinity toward sour gases [24].

**Table 1:** Specifications Limits for Natural Gas Delivery [7]

Major Components	Feed to Pipeline Gas		Feed to LNG Plant
	Minimum	Maximum	
CH <sub>4</sub>	75 Mol%	-	-
C <sub>2</sub> H <sub>6</sub>	-	10 Mol%	-
C <sub>3</sub> H <sub>8</sub>	-	5 Mol%	-
C <sub>4</sub> H <sub>10</sub>	-	2 Mol%	2 Mol% max
Pentanes and heavier	-	0.5 Mol%	0.1 mol% max
Nitrogen and other inert	-	3 Mol%	<1 mol%
Carbon dioxide	-	2 Mol%	< 50 ppmv
Hydrogen sulfide	6	7 mg/m <sup>3</sup>	< 4 ppmv
Total sulfur	115	460 mg/m <sup>3</sup>	< 20 ppmv
Water vapor	60	110 mg/m <sup>3</sup>	< 0.1 ppmv
Oxygen	-	1 Mol%	-



**Figure 1:** CO<sub>2</sub> Separation and Capture Technologies[14]

### 1.3 Cryogenic Process

Cryogenic is the separation process that is used to purify the gas mixture with low-temperature distillation in the range of (-150°C). In the commercial field, the feed gas mixture should contain a high percentage of carbon dioxide (> 90%), as separating the low concentration of carbon dioxide is very expensive. The most important features of this technology are the

production of liquid carbon dioxide with very high purity, where this liquid is ready for the transfer process, also, this technology eliminates the usage of water consumption, chemicals, and corrosion-related issues. [25, 26]. The disadvantages of the method are high energy consumption (especially in reduced gas streams), significant investment, sophisticated equipment, and low concentration inadequacy. Also, the refrigerated separation of carbon dioxide requires the removal of gases, such as water and heavy hydrocarbons, that tend to freeze and prevent heat exchangers [27, 28].

#### 1.4 Membrane Process

The most important definitions of the membrane are; Mulder (1996) suggests that a membrane is a "selective barrier between two phases", Ho and Sirkar (1992) describe it as an "interphase between two bulk phases". Selective barriers for gas separation may be porous or dense membrane; different mechanisms of gas permeation depending on the size of the pores. The transport process in the pores membrane governed by the mean free path of the penetrant and the size of the pores. Based on these factors, several diffusion mechanisms may be present [20]. Convective flow, no separation, occurs when the pores are relatively large-from 0.1 to 10  $\mu\text{m}$ , while the Knudsen flow governs the diffusion if the pore size reduces to the point where it is less than the mean free path of the penetrant molecules. Other techniques may occur when the pore volumes are lower than the mean free path of the separated molecules. They are molecular sieving, surface diffusion, and capillary condensation. All these operations may give rise to separations because of their different molecular differentiation abilities. Though these microporous membranes are themes of enormous research concern, all present industrial gas separations are depending on the high-density polymer membrane. Separation across high-density polymer films takes place by a solution-diffusion technique [20-29].

The intrinsic properties that are the hallmarks of modern membrane technology can be presented as follows:

The concepts of operation are uncomplex.

High selectivity and permeability of various specific components.

Flexibility in control, scale-up and modularize.

Chemicals and waste generation are non-existent.

Low energy consumption in operating at low pressure.

No phase changes.

Compatibility in integrated systems and different environments; [30]

In the literature, there is a lot of research showing advantages and disadvantages, as well as the limits, of the technologies used to capture acid gases from natural gas. Table 2 represents a summary of the overall comparison between the methods widely used in the separation of acid gas at the industrial and commercial levels. Despite the many methods used, the membranes method has proven to compete with absorption, which is the most established method.

## 2. Industrial Applications of Membrane in Gas Separation

The porous and non-porous membranes are used to separate carbon dioxide from natural gas, but in the industrial field, all the commercial membranes are based on the dense polymer membrane. The mechanism of gas separation in this type of membrane depends on a solution-diffusion technique [31]. At present, the membrane technology trade has reached \$500 million in the major applications in natural gas sweetening, air separations, and hydrogen separation [19]. Natural gas purification is anticipated to be the largest commercial membrane gas separation by 2020 [32]. Some criteria primarily affect the choice of the membrane when used for a specific application, such as mechanical efficiency at the operating conditions, stability, productivity and separation efficiency and so on. Four concepts must be carefully examined for the membrane GS process:

1. Design of the system and the module.
2. Configuration of the membrane.
3. Membrane thickness and structure.
4. Membrane material (separation factors, permeability).

The economics of gas separation by membrane depends on both permeability and selectivity. The rate at which any compound permeates through the membrane can be defined as permeability. Selectivity is the ability of a membrane to accomplish a given separation. Selectivity is a key parameter to achieve high product purity at high recoveries. [33, 34].

### 2.1 Gas Membrane Material and Structure

Based on the characteristics of the material, the membranes are classified into polymeric, inorganic, and metallic Membranes. Now, polymeric membranes have a major portion in the industrial field due to the distinguished economy and competitive performance. Polymeric membrane materials can be divided into rubbery and glassy polymer [35]. Polysulfone (PSF), cellulose acetate (CA), polyether sulfone (PESf), polyimide (PI), and polycarbonates (PC) are the common polymeric membranes. polysulfone (PSF) is one of the most widely investigated polymer membrane materials. Polysulfone is widely used to separate gas due to its low price, chemical stability, mechanical strength, high plasticization resistance, and good gas permeability and selectivity [36-38]. Polymeric membranes, especially utilized for Gas Separation, are in general asymmetric or composite and depend on a solution - diffusion conveying technique. These membranes, fabricated as flat sheet or hollow fibers, have a thin, high-density lacing layer on the microporous prop that gives mechanical vigor. Typically, polymeric membranes offer high with but limited selectivity's in comparison to porous inorganic materials because of their low free - volume. They in general, endure a trade-off between permeability and selectivity: as permeability enhanced, selectivity

reduced, and conversely. Presently, only about nine polymer materials are utilized for fabricating at least 85% of the membranes in the field [39].

**Table 2:** A Comprehensive Comparison of CO<sub>2</sub> Capture Technology

Process	Advantages	Disadvantages
Absorption	It is used to effectively remove acid gases over a range of 50 to 100 percentages and is the most popular process.	In physical absorption, the process can be costly When chemical solvents are used and at low pressures, the purification time is long.
Adsorption	The purity of the product is more than 99.5 per cent The units of this process are characterized by easy transportation and installation, especially in the difficult geographical oil sites	Product recovery is lower Comparatively single pure product
Membrane	Relative operating and investment costs, in addition to diversity and simplicity The lack of operational problems when the high recovery of the product and the high pressure Little space is needed, and no environmental impacts	Permeate stream send to recompression unit The purity is not very high
Cryogenic	High recovery The product is of high purity	High energy consumption Small units are not economical
Problems of Operation	Absorption Process	Membrane Process
Working effort	Normal	Higher than normal
Loss of the product	Small amount	The quantity depends on conditions
CO <sub>2</sub> concentration in the product	ppm levels	≥2%
H <sub>2</sub> S concentration in a product	<4 ppm	Probably
Consumption of energy	Very high	Low at low pressures
Operating cost	Moderate	Medium or lower
Maintenance cost	Moderate to low	Low at low pressures
Ease of operation	Relatively complex	Relatively simple
Environment impact	Moderate	Low
Dryness	Wet gas	Dry gas
<b>Problems relating to capital Cost</b>		
Delivery time	Lengthy for big units	Shorter time
Time required to install	lengthy	Short for skid-mounted equipment
Costs of pretreatments	Little	In the case of <i>compression</i> , it is high
Compression use	No	Depend on product specifications

## 2.2 Membrane Configuration

In the industrial field, there are different models which are, hollow fiber membrane, spiral wound membrane and plate frame membrane. The choice between these three modules depends on the following requirements being met:

1. High packing density.
2. Good flow distribution (no dead zones, no channeling).
3. Good mechanical, thermal and chemical stability.
4. Low-pressure drops.
5. Cheap manufacturing.
6. Ease of maintenance and operation.
7. Possibility of membrane replacement.
8. The compactness of the system scale.
9. Possibility of cleaning.

Because the cleaning ability is of less importance in gas separation, the main interest of module design is a high packing density. [36-41]. The hollow fiber model is distinguished from the spiral wound model, at the same size of two models(0.04 m<sup>3</sup>), the effective area is 575 m<sup>2</sup> in the first type while it is 30 m<sup>2</sup> in the second type[42]. Another advantage of the hollow fiber membrane is that the unit can withstand high feeding pressure and low permeate pressure without the need for mechanical supports. [43]. The closeness and uniformity of the fibers inside the shell are the factors that determine the packing density. The packing density ( $\varphi$ ) within the membrane model can be defined as follows:-

$$\varphi = n_{fiber} \times \left(\frac{d_o}{D_i}\right)^2 \quad (1)$$

Where  $n_{fiber}$  is the number of fibers,  $d_o$  is the fiber outer diameter and  $D_i$  is the module inside diameter [4].

The membrane costs ratio of hollow spinning plant operating continuously to the spiral – wound modules is 5 to 20 \$ per m<sup>2</sup> [44]. Asymmetric hollow fibers have tubular forms and offer high fluxes needed for fruitful separations attributed to the capability to decrease the segregating layer to a fluffy skin on the outer surface of the membrane [29]. Because of the low cost, high material density and large surface area per unit volume, hollow fiber membrane modules are extensively utilized in gas separation applications. Furthermore, the asymmetric framework of the hollow fibers gives a reasonable mechanical strength and decreases the membrane resistances versus the transport of components [45].

### 2.3 Hollow Fiber Membrane Module Modeling

Three important aspects of modelling in the asymmetric hollow fiber membrane must be adopted: the transfer of the material across the membrane, the flow through the membrane module, and the styling of the framework. Three sub-models required to complete the description of the HFM model: two sub-models describe flow on both sides of the membrane and the third characterizes the separation mechanism in the membrane and any porous support material as illustrated in Figure 2 [46].

The gas separation model in a membrane was firstly presented by Weller and Steiner (1950). From that time, several models and estimating approaches have been suggested for the different flow patterns and module configurations. Most of these models have focused on binary gas mixtures [47-53] and only a few considered the separation of multicomponent mixtures[54-59]. Many mathematical models have been reported to describe the gas permeation of binary gas mixtures. However, most industrial separation applications involve multicomponent separation. Hence, deriving a mathematical model based on a multi-component mixture is highly important. An overview of the proposed models binary gas mixtures is shown in Table 3 and 4. The model of Pan's [60] has been approved to be a good implementation of multicomponent gas separation in hollow fiber asymmetric membranes. Though, the solution method needs initial assessments of the pressure and concentration behavior within the fiber. To beat the complication of the model, various adjustments, and tactics for a solution have been suggested [54].

Kovvali et al. [61] applied a linear approximation method to introduce the feed and permeate ingredients at definite domains across the fiber length. Coker et al. [62] introduced a model for multicomponent gas separation utilizing a hollow fiber membrane and suggested a stage-wise technique utilizing the 1<sup>st</sup> order finite difference method to improve a group of equations from the differential mass balances. This technique needs an initial assumption of the ingredients rates at every stage. Cocker developed a multicomponent gas separation model for co-current, counter-current, and cross-flow configuration in the hollow fiber model with permeate sweep. The model was used to explore the effect of permeate purging on the separation performance for an air separation unit. Marriott et al. [63] introduced a detailed model for multicomponent gas separation based on the differential balance of mass, momentum, and energy. Also, this model needs the information of diffusion and dispersion coefficients in the fluid phase. It is hard to depict precisely the mass transfer in the porous membrane framework in the asymmetric membrane can barely be elucidated correctly. Consequently, the viability of the model is limited by the uncertainty in obtaining such the type of data needed. The authors reported a good agreement of their model with the experimental data. Chowdhury et al. [54] a new numerical solution tactic has been improved to settle Pan's model for multicomponent gas separation by asymmetric hollow fiber membranes. This numerical solution tactic removes the trial-and-error method needed to fix the boundary value problem. It is no longer needed to guess initial values of concentration, pressure, flow profiles inside the hollow fiber, permitting rapid fulfilment of the model equations. The model foretelling and the numerical method has been reliable with data obtained for many different designs of membrane modules published in the literature. An overview of the proposed models for multi-gas mixtures of feed is shown in Table 5.

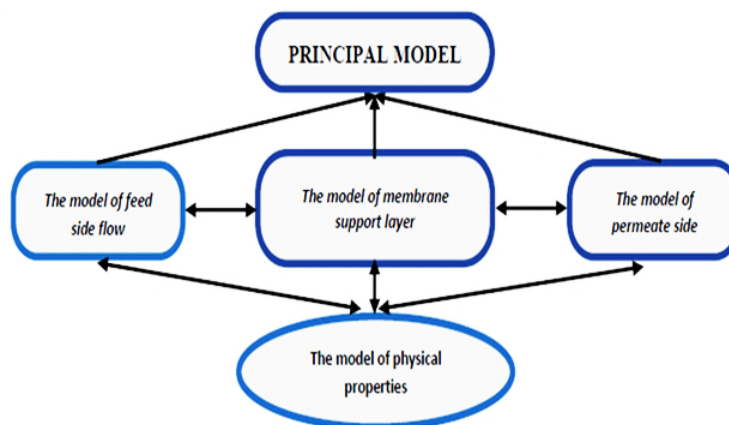


Figure 2: CO<sub>2</sub> Separation and Capture Technologies[14]



**Table 3:** Researchers' Suggested Models for a Binary Gas Mixture

Membrane & Gas Mixture	Model hypotheses	Parametric Study	Performance	Main Remarks	Ref.
Asymmetric HFM CO <sub>2</sub> /CH <sub>4</sub>	<ol style="list-style-type: none"> <li>Steady-state and isothermal.</li> <li>Neglecting the influence of temperature and pressure on permeability</li> <li>Neglecting the pressure drop on both sides of the membrane.</li> <li>Assume that the concentration polarization effect is not important</li> </ol>	<ol style="list-style-type: none"> <li>0-50 % CO<sub>2</sub> in Mixture.</li> <li>20-100bar pressure.</li> <li>Design Configurations Selectivity.</li> </ol>	<ol style="list-style-type: none"> <li>Methane recovery.</li> <li>Membrane area.</li> </ol>	<ol style="list-style-type: none"> <li>Crossflow mathematical model has been suggested to be incorporated with ASPEN HYSYS.</li> <li>Methane recovery decreases with the increase in CO<sub>2</sub>% in feed, while the total area increases for the effective separation.</li> <li>Methane recovery increases directly with the change of feed pressure. Besides, the increase in pressure contributes to reducing the area required for separation.</li> <li>The increase in selectivity increases methane recovery, especially for double stage and single stage with recycling.</li> <li>On the other hand, an increase in selectivity decreases the total membrane area required for effective separation.</li> </ol>	[37]
Polyimide a shell-fed hollow fiber module CO <sub>2</sub> /CH <sub>4</sub>	<ol style="list-style-type: none"> <li>steady-state and isothermal.</li> <li>plug flow in both side of the membrane.</li> <li>The pressure drop in the feed side is negligible.</li> <li>All fibers have a uniform shape.</li> <li>Neglecting the influence of pressure and feed composition on permeability.</li> </ol>	<ol style="list-style-type: none"> <li>The flow rate of feed.</li> <li>The pressure of feed.</li> <li>The permeate pressure.</li> <li>The length of fiber.</li> <li>CO<sub>2</sub> Conc.</li> </ol>	<ol style="list-style-type: none"> <li>Separation efficiency.</li> <li>Optimum process and design condition.</li> </ol>	<ol style="list-style-type: none"> <li>permeation mechanism is solution-diffusion.</li> <li>Impurity concentration is the most important factor in process design.</li> <li>The pressure of the permeate has a great influence on the carbon dioxide concentration in the retentate, and this is evidenced by the results.</li> <li>Recovery of CO<sub>2</sub> decreases with increases in the feed flow rate.</li> <li>The flux of CO<sub>2</sub> increases by pressure increase through a membrane in two flow patterns.</li> <li>To solve the equations the finite difference method was used.</li> </ol>	[38]
Asymmetric HFM CO <sub>2</sub> /CH <sub>4</sub>	<ol style="list-style-type: none"> <li>Newtonian fluids, Laminar flow, Isothermal, steady-state and incompressible flow.</li> <li>Constant permeability</li> <li>Neglecting the effect of concentration on density and diffusion.</li> <li>Darcy law governs the flow in the porous layer.</li> <li>There is diffusion in axial and radial.</li> <li>Saturation in the porous layer.</li> </ol>	<ol style="list-style-type: none"> <li>Porous layer.</li> </ol>	<ol style="list-style-type: none"> <li>Flow regimes.</li> <li>Separation process.</li> </ol>	<ol style="list-style-type: none"> <li>At different values of the Reynolds, number and different values of permeability in the porous layer, the mass transfer equations, Darcy's equation and Navier Stock equation is solved.</li> <li>Simulation by CFD with all assumptions about the model.</li> <li>The presence of the porous layer adds resistance to the transfer of materials across the membrane, also increase the pressure drops.</li> <li>The Sherwood number is reduced at all Re as the resistance of the porous layer is increased.</li> <li>This paper emphasized the importance of embedding the porous layer when modelling the hollow fiber membrane.</li> </ol>	[33]

**Table 4:** Researchers' Suggested Models for a Binary Gas Mixture

Membrane & Gas Mixture	Model hypotheses	Parametric Study	Performance	Main Remarks	Ref.
Flat Carbon molecular sieve membranes (CMSMs) CH <sub>4</sub> / C <sub>2</sub> H <sub>6</sub>	<ol style="list-style-type: none"> <li>Membrane layer is lumped.</li> <li>Isothermal &amp; steady-state.</li> <li>co-current flow pattern.</li> <li>Velocity in the module is constant.</li> <li>Molecular diffusion is negligible.</li> <li>Ideal &amp; incompressible gas behavior.</li> </ol>	<ol style="list-style-type: none"> <li>Effective area.</li> <li>Temperature.</li> <li>Feed pressure.</li> <li>Feed flow rate.</li> </ol>	<ol style="list-style-type: none"> <li>Recovery.</li> </ol>	<ol style="list-style-type: none"> <li>The Runge-Kutta method has been applied to solve the model.</li> <li>In the mentioned model, the sorption-diffusion mechanism has been considered.</li> <li>The error ranged from 0 to 14 per cent between the experimental results and the model results.</li> <li>The recovery changed positively with the effective area, temperature and pressure of the feeder, while the change was inverse with the flow of the feeder.</li> </ol>	[33]
Cellulose Acetate Asymmetric HFM O <sub>2</sub> / N <sub>2</sub> CO <sub>2</sub> /CH <sub>4</sub> He/CH <sub>4</sub>	<ol style="list-style-type: none"> <li>The effect of pressure on permeability is negligible</li> <li>Steady-state and plug flow.</li> <li>In the direction perpendicular to the membrane, there is no concentration gradient</li> <li>The total pressure on both sides of the membrane is constant.</li> </ol>	<ol style="list-style-type: none"> <li>Stage cut<sup>1,2,3</sup>.</li> <li>Feed conc.<sup>2</sup>.</li> <li>Feed pressure<sup>2</sup>.</li> <li>Feed flow rate<sup>3</sup>.</li> </ol>	<ol style="list-style-type: none"> <li>O<sub>2</sub> mol%<sup>1</sup>.</li> <li>Recovery<sup>2,3</sup>.</li> </ol>	<ol style="list-style-type: none"> <li>The membrane gas separation behavior was predicted using the finite difference method</li> <li>The developed model showed an excellent forecast of results</li> <li>With the increase in the feed pressure, the concentration of CO<sub>2</sub> decreased.</li> <li>With the increase in the feed flow rate, the contact time of CO<sub>2</sub> concentration with the membrane active surface area is small, and the concentration of CO<sub>2</sub> in the permeate side decreases.</li> <li>When the feed concentration of CO<sub>2</sub> is increased then the amount of gas diffusing through the membrane decreased due to less area.</li> </ol>	[41],[42]
A plate-and-frame membrane module N <sub>2</sub> /CH <sub>4</sub>	<ol style="list-style-type: none"> <li>Isothermal &amp; steady-state.</li> <li>Ideal gas behavior.</li> <li>Plug flow mode.</li> <li>The constancy of membrane permeability.</li> </ol>	<ol style="list-style-type: none"> <li>Feed flow rate.</li> <li>Stage cut.</li> <li>Feed conc.</li> </ol>	<ol style="list-style-type: none"> <li>Retentate/permeate flux.</li> <li>The concentration of components in retentate/permeate.</li> </ol>	<ol style="list-style-type: none"> <li>In this study, a gas separation membrane module was created in the ACM program and exported to Aspen Plus for further work.</li> <li>In the study, we considered three operating modes of the membrane module: direct flow, countercurrent flow, and crossflow.</li> <li>A comparison of the results has shown that the calculated data are close to the experimental data, confirming that the block can be used for modelling membrane gas separation processes.</li> </ol>	[43]

**Table 5:** Researchers' Suggested Models for a Multi-Gas Mixture

Membrane & Gas Mixture	Model hypotheses	Input variable	Output variable	Main Remarks	Ref.
<b>PDMS Commercial membrane</b> <b>CH<sub>4</sub>, C<sub>3</sub>H<sub>8</sub>, H<sub>2</sub></b>	<ol style="list-style-type: none"> <li>Steady-state and plug flow.</li> <li>Crosscurrent membrane module.</li> <li>Two – dimensional problem.</li> <li>Pressure drop is negligible.</li> <li>The solubility and diffusion in the membrane is a function of feed composition, temperature and pressure.</li> </ol>	<ol style="list-style-type: none"> <li>Temperature.</li> <li>Pressure.</li> <li>Area.</li> <li>Feed composition.</li> </ol>	<ol style="list-style-type: none"> <li>A flow rate of the retentate.</li> <li>Permeability.</li> <li>Coefficient of diffusion.</li> <li>Separation coefficient of propane.</li> <li>Ideal mixing split factor.</li> <li>Solubility.</li> </ol>	<ol style="list-style-type: none"> <li>A mathematical model based on diffusion – solution mechanism.</li> <li>Through the numerical solution method, interpolation by using orthogonal collocation approximates.</li> <li>Solution method accurate and effective for solving the threshold values and have less algebraic equation compared to the limited components method.</li> <li>When examining the effect of temperature, pressure, and feed concentration on dimensionless retentate components flow rate, the deviation between model results and practical results ranged between 4.5% and 40%.</li> </ol>	[55]
<b>Asymmetric HFM</b> <b>H<sub>2</sub>, N<sub>2</sub>, Ar, CH<sub>4</sub></b>	<ol style="list-style-type: none"> <li>Complete mixing mode.</li> <li>Isothermal condition.</li> <li>Fick's law is applicable.</li> <li>The pressure of the permeate side is constant.</li> <li>The permeabilities are pressure independent.</li> <li>Feed side pressure drop is neglected.</li> </ol>	<ol style="list-style-type: none"> <li>Feed flow rate.</li> <li>Pressure.</li> <li>Temperature.</li> <li>Feed composition.</li> <li>Area.</li> </ol>	<ol style="list-style-type: none"> <li>Stage cut.</li> <li>H<sub>2</sub> Recovery.</li> <li>Composition of the permeate.</li> </ol>	<ol style="list-style-type: none"> <li>In this study shortcut, mathematical models are presented for multicomponent systems with cross-flow, co-current and counter-current flow configurations.</li> <li>A simple yet robust method is proposed for solving co-current and counter-current flow modes, which does not require any trial and error (except for counter-current rating problems) or using dimensionless variables and pressure drop calculations can be incorporated into the model easily.</li> <li>The presented models are applicable for systems with any number of components even for binary systems with no modifications required.</li> </ol>	[56]
<b>Asymmetric HFM</b> <b>He, N<sub>2</sub>, CO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub></b>	<ol style="list-style-type: none"> <li>Steady-state and plug flow.</li> <li>The pattern of flow is countercurrent.</li> <li>Diffusion mechanics based on Fick's law.</li> <li>Non-ideal behavior in the gas phase.</li> <li>The permeability of the gas mixture through the membrane is based on the Dual model.</li> <li>Hagen-Poiseuille equation to describe the pressure drop inside the fiber.</li> <li>On both the feed and permeate side the axial diffusion is neglected.</li> </ol>	<ol style="list-style-type: none"> <li>Feed flow rate.</li> <li>Pressure.</li> <li>Temperature.</li> <li>Feed composition.</li> <li>Area.</li> <li>permeate pressure.</li> </ol>	<ol style="list-style-type: none"> <li>Stage cut.</li> <li>H<sub>2</sub> Recovery.</li> <li>Composition of the permeate.</li> </ol>	<ol style="list-style-type: none"> <li>In this paper, a mathematical model has presented and an investigation of a robust and accurate numerical model.</li> <li>In this paper, the numerical approach used with the least computational effort.</li> <li>Using the data in the literature the proposed numerical model was applied.</li> <li>Separation and recovery of helium from multiple components predicted by the model excellently.</li> </ol>	[59]
<b>Asymmetric HFM</b> <b>CO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub></b>	<ol style="list-style-type: none"> <li>Ideal gas in different regions of the membrane.</li> <li>The isothermal process and the effect of temperature is limited only by the density and viscosity of the mixture</li> <li>The permeability of the component is constant.</li> <li>Hagen-Poiseuille equation governs pressure drop in the permeate side.</li> <li>The developed transport model did not take into account the axial dispersion.</li> </ol>	<ol style="list-style-type: none"> <li>Pressure.</li> <li>Feed flow rate.</li> <li>Temperature.</li> <li>Feed composition.</li> <li>permeate pressure</li> </ol>	<ol style="list-style-type: none"> <li>Stage cut.</li> <li>Dimensionless length.</li> <li>CO<sub>2</sub> mol % in the permeate.</li> </ol>	<ol style="list-style-type: none"> <li>In this paper, a model is proposed to simulate co-current hollow fiber membranes, and the proposed model was robust, fast and accurate.</li> <li>The algorithm is based on a modified shooting method with quadratic interpolation, which requires no derivative computations versus classical optimization methods.</li> <li>3.4 seconds is the maximum computational time.</li> <li>The maximum error in permeate pressure was 0.37%.</li> <li>Experimental and other data in the literature were used to validate the method.</li> </ol>	[64]

### 3. Conclusion

An overview was presented on the importance of gas as a clean source of energy and the growing global demand. The different processes for removing acid gases from natural gas are presented and details of each process along with operational conditions are specified. A comparison was made between the four processes, and the advantages and disadvantages were identified in terms of costs, environmental damages and operational conditions. The membrane removal process is the strongest and most promising option among the four methods, as it is environmentally friendly and has a low cost in addition to ease of operation and maintenance. Unlike the earlier published review papers which focus on the relevant research [10-13], and/or address the application of membrane units in the treatment of natural gas in general (dehydration, N<sub>2</sub> removal, etc.) [24-39], this paper has a specific focus on the 'industrial' aspect of 'acid gas removal from natural gas. The paper consolidates the information published in the literature for several commercial membranes, illustrates the advantages and shortfalls of each membrane, and provides a high-level guideline for the selection of commercial membranes. The manuscript provides more details for CO<sub>2</sub> removal membranes compared to H<sub>2</sub>S, noting the industry's lack of interest in H<sub>2</sub>S removal membranes.

#### Author contribution

All authors contributed equally to this work.

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## Data availability statement

The data that support the findings of this study are available on request from the corresponding author.

## Conflicts of interest

The authors declare that there is no conflict of interest.

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