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MEMBRANE TECHNOLOGY SYMPOSIUM ARTICLE

A Review of the Applications of Membrane Separation Technology in Natural Gas Treatment

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ABSTRACT

While membrane technology is still young and has great potential for further improvements, it has been proven economical and technically efficient in many applications such as treatment of natural gas. In this paper a brief history of membrane gas separation technology is presented, and the theoretical aspects of the process are discussed. The article also introduces applications of membranes in the natural gas industry, discusses different system designs and the effects of operating parameters on the performance of membrane systems, and compares the technical and economical aspects of membrane processes with conventional technologies.

Key Words. Membrane; Gas separation; Natural gas treatment

INTRODUCTION

Sour gas mainly contains methane, carbon dioxide, nitrogen, higher hydrocarbons, and small amounts of hydrogen sulfide, helium, oxygen, argon, and water vapor. The composition of natural gas varies from one location to another and its quality highly depends on the concentration of the contaminants. Removal of acid gases (CO_2 and H_2S) and water vapor is essential before feeding the natural gas to a pipeline. All these three contaminants are corrosive, and hydrogen sulfide is toxic as well. To meet pipeline requirements, the contaminants must comply with such concentration specifications as $<2\%$ CO_2, <4 ppm H_2S, and <0.1 g/m^3 H_2O.

Available techniques for natural gas treatment include membrane, absorption, adsorption, and cryogenic distillation. Membrane processes have been

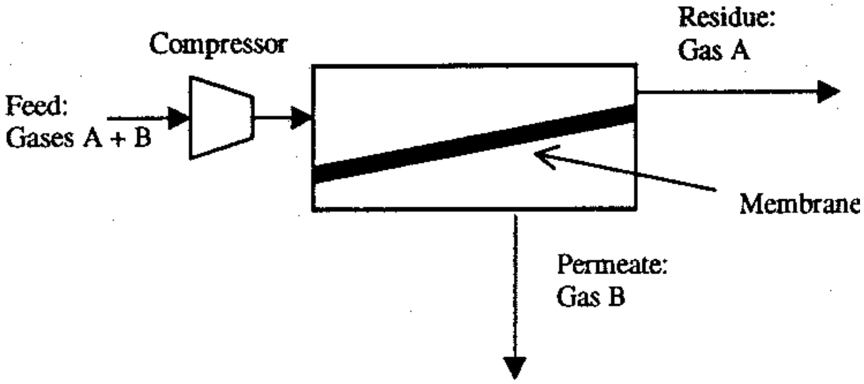


FIG. 1 A simplified illustration of a gas separation membrane system.

proven to be technically and economically superior to the competing technologies in many industrial applications. This superiority is due to many advantages that membrane technology benefits from, including low capital investment, simplicity and ease of installation and operation, low maintenance requirements, low weight and space requirements, and high process flexibility.

Figure 1 illustrates the simplest membrane gas separation process possible. As shown in this figure, a gas mixture is fed into the membrane at high pressure. The fast diffusing gas permeates through the membrane and is enriched in the permeate side. The slower gas is concentrated in the retentate or residue stream.

BRIEF HISTORICAL BACKGROUND

The gas separation properties of membranes have been realized for more than a century. The early documented reports root back to the works of Mitchell, Fick, and Graham of the mid-nineteenth century. In 1831, Mitchell measured the rates of escape of ten gases through natural rubber balloons (1). At approximately the same time, Fick developed his famous laws of diffusion by studying gas transport across a nitrocellulose membrane (2). A few decades later in 1866, Thomas Graham observed the separation of gases using natural rubber via Knudsen diffusion (3). In 1920, H. A. Daynes recognized the relation between time lag and diffusion coefficient by studying the nonsteady-state transport behavior of gases through a membrane (2).

Despite many experimental works, the progress of membrane separation techniques was very slow in the early stages. More than a century separates the early works of Mitchell and the first large-scale application of membranes, which was the concentration of uranium 235 from 0.17 to 3% (4).

The major problem with the early membranes was their insufficient selectivities and low fluxes. The first breakthrough came about with the introduction of asymmetric membranes by Loeb and Sourirajan (5). They successfully produced a membrane with a very thin dense layer and a relatively thick porous sublayer. The dense layer was responsible for the separation, while the porous sublayer provided mechanical strength to the selective layer with minimum resistance to the permeation of components. Figure 2 shows the cross section of an asymmetric membrane with the dense selective layer and the porous sublayer.

While Loeb–Sourirajan membranes proved efficient in water desalination, they were not suitable for gas separation because the membranes lost their separation properties after drying. Vos and Burris (6) solved the problem by adding surfactant to the water to reduce the interfacial tension between the membrane walls and water molecules. The improved drying process was a major step in the introduction of membranes to the gas separation industry.

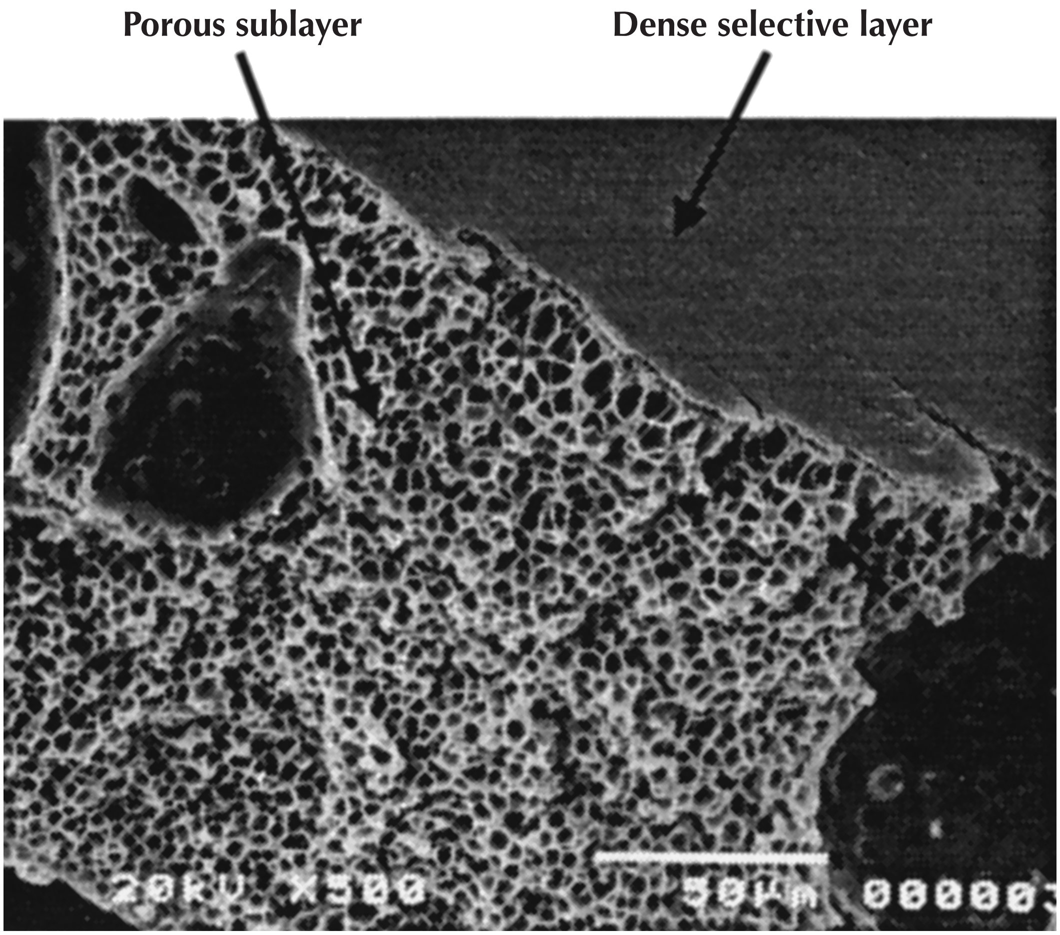


FIG. 2 Cross section of an asymmetric gas separation membrane with the dense selective layer and the porous sublayer.

Other major progresses in the preparation of gas separation membranes include the development of composite membranes by Ward et al. (7) and the coating technique by Henis and Tripodi (8, 9). Composite membranes allowed for the separate optimization of the selective and supporting layers. Using this technique, both separation factor and permeance of a membrane could be enhanced. The coating technique, on the other hand, increased the separation factor by plugging the large pores and defects of the selective layer using materials such as silicone rubber. These accomplishments eventually led to the introduction of the PRISM membrane system by Monsanto (now Permea). It was a hollow fiber system spun from polysulfone and was used for hydrogen recovery in the ammonia process.

Since the early 1980s many researches focused on fundamental and industrial improvement of membrane systems. The results of these efforts can be realized in many industrial-scale separation processes in which membrane systems are used. These applications could be categorized in three principal classes: hydrogen recovery, natural gas and landfill gas treatment, and oxygen and nitrogen enrichment from air.

PRINCIPLES OF MEMBRANE GAS SEPARATION

Membrane gas separation is a pressure-driven process in which a gas mixture is fed into the membrane module under high pressure, typically between 10 to 200 bars. The separation's driving force is the partial pressure difference of the gases in the feed and permeate. Membrane separation is considered a nonequilibrium process because it is based on the relative permeation rate of the feed components. If a membrane system is allowed to go to equilibrium, permeation would continue until the pressure and concentration of the gases on both sides are equal.

The two main parameters defining the performance of a membrane are separation factor and permeance. In a binary system consisting of gases "a" and "b" with gas "a" as the faster permeating gas, separation factor is defined as the concentration ratio of "a" to "b" in the permeate over the concentration ratio of "a" to "b" in the feed:

$$\alpha_{\mathrm{a/b}} = \frac{y_{\mathrm{a}}/y_{\mathrm{b}}}{x_{\mathrm{a}}/x_{\mathrm{b}}}$$

where α is the separation factor, y_{a} and y_{b} are the concentrations of gases "a" and "b" in the permeate, and x_{a} and x_{b} are the concentrations of gases "a" and "b" in the feed. Permeance is defined as the volume of the feed passing through a unit area of membrane at unit time and under unit pressure:

$$P = v/At\Delta p$$

where P is permeance ($\mathrm{cm^3/cm^2 \cdot s \cdot cmHg}$); v is volume of gas ($\mathrm{cm^3}$); A is area ($\mathrm{cm^2}$), t is time (s); and Δp is pressure difference (cmHg).

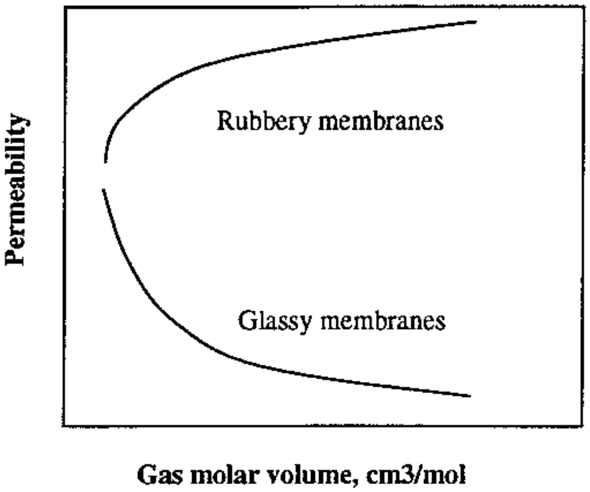


FIG. 3 Comparison of permeation rates of gases through glassy and rubbery membranes.

Polymeric membranes can be categorized under two distinct groups: glassy membranes and rubbery ones. Glass transition temperatures of rubbery membranes are lower than the ambient, while those of the glassy ones are higher. The two groups behave differently toward permeation of gases. Figure 3 gives a general idea of permeation rates of different gases through glassy and rubbery membranes. As shown in this figure, glassy membranes are more permeable toward smaller molecular size gases, and rubbery ones allow condensable gases to permeate more easily.

APPLICATIONS OF MEMBRANE PROCESSES IN THE NATURAL GAS INDUSTRY

Of particular interest in this paper is the applications of membranes in natural gas treatment. The commercial use of membrane systems for removal of acid gases started in 1984 with the installation of the SACROC unit by Cynara, a subsidiary of Dow (10). Membrane processes have also found applications in offshore installations, where membrane systems can offer major savings by weight reduction of the gas to be transported to the shore. Verghese (11) summarizes the industrial installation of membranes for natural gas treatment.

Amine absorption dominates the natural gas treatment processes. In recent years, membrane technology has impacted the market by its many advantages over amine processes, including:

- Smaller and lighter systems, especially for offshore platforms.
- Simultaneous removal of carbon dioxide, hydrogen sulfide, and water vapor.
- Small or no need for human supervision and maintenance.

- Reduced energy consumption.
- No fire or explosion hazards.
- Lower capital and operating costs.
- Ability to treat gas at the wellhead.

The high gas pressure at the wellhead is ideal for membrane processes.

Removal of Acid Gases (CO_2 and H_2S)

Carbon dioxide can be found in natural gas in various concentrations, typically between 7 and 40%. In case of enhanced oil recovery this concentration can rise up to 80%. Removal of carbon dioxide to less than 2% is essential for reducing the risk of pipeline corrosion. Most commercial membranes offer a separation factor of 20 or higher for CO_2 over methane. However, a number of laboratory-scale experiments report significantly higher separation factors, which are indicative of a great potential for improving the CO_2 removal membranes. Table 1 compares the separation factor of some membranes toward carbon dioxide over methane found in laboratory-scale experiments. Membrane modules are of hollow fiber or spiral wound designs. CO_2 preferentially concentrates in the permeate side, leaving the methane-rich residue at approximately the same pressure as the feed.

Cook and Losin (21) reported the installation of a membrane system to treat 30 MMscfd of natural gas containing 11% carbon dioxide. They compared a membrane system with an amine/glycol system and concluded that the total operating costs for both systems were the same at 0.13 $/MMscf. They also recognized the advantages of the membrane system such as flexibility for expansion and turndown, and space savings.

TABLE 1
Separation Factor of Selected Membranes toward Carbon Dioxide over Methane

Polymer	Separation factor	Reference
Polyimide	67	12
Polyimide	50	13
Polycarbonate	30	14
Polyimide (film)	50–70	15
Polyethersulfone	50	16
Radel A PSF	32.3	17
Polyaniline	336	18
Ultem (polyetherimide)	45	19
Polyimide	70	20

Because CO_2 removal membranes are also selective to hydrogen sulfide, there is no need for an additional membrane stage to separate the latter gas. Hydrogen sulfide is both corrosive and toxic, and it would be advantageous to remove it at the wellhead. Commercial membranes such as cellulose acetate show relative selectivities of around 50 for H_2S with respect to methane, and 2 with respect to carbon dioxide. Membrane units can reduce the H_2S content of natural gas to around 100 ppm. Final polishing can then be achieved with a zinc oxide bed. Because of the toxicity of hydrogen sulfide the concentrated gas must be treated before releasing it into the atmosphere. In a study conducted by Alexander and Winnick (22), a novel membrane system is introduced for removal of hydrogen sulfide from natural gas, and recovery of elemental sulfide and hydrogen, using an electrochemical membrane separator. In an economic comparison with available technologies they showed that the treating cost for 1000 m^3 of feed is \$5.38 for the membrane system versus \$9.25 for the conventional technology.

While membranes are good for bulk removal of acid gases, they are inferior to, or must be combined with, other processes when the acid gases are present at low concentration. That is because, at small concentrations, the partial pressure of acid gases, and therefore the driving force of the process, decreases. A number of membrane hybrid systems have received attention in the literature for acid gas removal from natural gas. The membrane/potassium carbonate system at the SACROC installation discussed earlier is an example of such hybrids (10). PRISM, from Permea, has also been combined with traditional amine and cryogenic processing units (23). Baldus and Tillmann (24) also show that a membrane/cryogenic hybrid with a membrane separation factor of 20 is advantageous over a membrane alone system. They show that a membrane system alone is economical and efficient if the separation factor of the membrane is 50 or greater.

Dehydration

Most often natural gas is saturated with water vapor. The presence of water in the natural gas stream may cause formation of solid methane hydrates, which can block the pipelines or lead to formation of carbonic acid and S^{2-} ions in the presence of CO_2 and H_2S, respectively. The latter substances corrode pipelines. Membranes remove water vapor along with carbon dioxide and hydrogen sulfide in natural gas upgrading applications. Typical separation factors of membranes toward water vapor are in the range of 500 with reference to methane. Therefore, membranes can efficiently remove water vapor to low ppm levels (~5–20 ppm) even at low partial pressures.

Membrane technologies compete with the glycol absorption process for natural gas dehydration. The main disadvantages of the glycol process are loss of glycol due to contamination with aromatic hydrocarbons, and maintenance requirements. Membrane processes do not suffer from the above problems. However, they have the disadvantage of methane loss in the 5 to 6% range compared with the acceptable level of 2 to 3%.

Higher Hydrocarbons Removal

The presence of condensable hydrocarbons in natural gas streams can sometimes lead to condensation in pipelines. Traditionally, the whole stream is compressed and cooled to induce condensation of the heavier hydrocarbons. This process is expensive and cumbersome. Membranes can reduce the condenser's duty by allowing only the condensable gases to enter the condenser, resulting in significant savings in the condensation cost.

Rubbery membranes are ideal for the removal of higher hydrocarbons. In this process, condensable hydrocarbons preferentially permeate through the membrane and accumulate in the permeate side. The transport mechanism in this case is different from other membrane gas separation processes. The heavier hydrocarbons have a higher condensation tendency. With the choice of the proper rubbery membrane, these hydrocarbons form a thin film over the membrane surface, restricting the accessibility of the membrane to noncondensable gases such as nitrogen. The higher hydrocarbons permeate through the flexible polymer chains leaving the noncondensable gases behind. Therefore, unlike most of the membrane separation processes, removal of higher hydrocarbons is more efficient at lower temperatures, where their condensability is enhanced.

Nitrogen Recovery

Nitrogen is also found in different concentrations in natural gas. Because of low selectivities of existing membranes toward N_2 over CH_4, this technology is not viable for this separation. The main technology available for nitrogen/methane separation is adsorption.

Applications in Enhanced Oil Recovery

In addition to natural gas treatment, membranes have found a variety of applications in enhanced oil recovery (EOR). In EOR, CO_2 is injected into an oil-bearing reservoir at high pressure. The carbon dioxide dissolves the oil and carries it to the production well. The produced gas contains CO_2, CH_4, and other hydrocarbons. It is desired to recover both the natural gas and carbon dioxide contents of this stream. The volume of produced gas and it CO_2 content increase with time, making it a difficult application for conventional amine processes. The modularity of membranes allows for capacity expansion as needed, a flexibility not available with other technologies. In a typical application of membrane systems to EOR, the carbon dioxide concentration in the product is reduced from 70 to 10%. The permeate is then enriched in CO_2 to around 95%.

A membrane/DEA hybrid is a well-established design for EOR applications at optimum cost. In this design a membrane is used to remove the bulk of the carbon dioxide before feeding the stream into a conventional DEA system for final polishing. According to a study conducted by Ryan (25), a combination of these two processes is more economical than each process alone.

Presently, membranes and membrane hybrids are showing significant growth and attract more attention in both natural gas and EOR applications. In addition, removal of carbon dioxide from NGL using membranes resulted in commercial adoption of this process. All of the above applications have proven to be economically more justifiable than competing techniques.

MEMBRANE SYSTEM DESIGN AND ECONOMY

Module Configurations

Industrial membrane modules usually come in three major configurations: plate and frame, spiral wound, and hollow fiber. Each configuration has its advantages and disadvantages. For example, hollow fibers provide the highest surface area per volume but are difficult to clean, while the opposite is valid for the plate and frame configuration.

A plate-and-frame system replicates the conventional filtration setup. The flat sheet membranes are stacked on top of each other, in the form of envelopes, separated by low flow resistance separators. The feed flows over the outside of the membrane envelopes and permeate is collected inside.

In spiral-wound modules, two flat-sheet membranes are sealed together to form an envelope enclosing a separator in between. The separator prevents the two membranes from collapsing and provides mechanical strength, while exhibiting minimum resistance toward the flow of permeate. One or more of these envelopes are wound around a cylindrical collector. In this configuration, feed flows outside the envelopes and permeate is collected inside and is removed through the central collector.

In the hollow-fiber configuration the membrane is shaped into thin tubes with a hollow inside. Thousands of these fibers are enclosed in a pressure vessel with an overall look of a conventional heat exchanger. Feed can flow on the outside or inside of the fibers and permeate is collected from the other side. The most common gas separation modules are either the spiral-wound or hollow-fiber configurations.

Module Design

The cost of membrane systems is greatly influenced by their design. Membrane modules can be put together in one stage or multistage designs.

Each stage consists of a bank of membrane modules arranged in parallel or series. For relatively low flow rate applications, a single-stage membrane system provides the most economical result. As the size of the process and/or the value of desired compound increases, more complex designs are needed.

Single-Stage Membrane System

In a single-stage design, feed is treated in a once-through mode and is split into the permeate and retentate streams. The latter streams are the final products of the separation. An example of a single-stage design is adapted from Ref. 26 and is shown in Fig. 4. In this example a stream with 93% methane and 7% carbon dioxide is fed into the system at high pressure. The permeate is collected at close to atmospheric pressure with 36.6% CO_2 content. The retentate exits the module with negligible pressure drop and contains 98% CH_4. This operation corresponds to an overall separation factor of 7.6.

The effects of feed pressure, permeate pressure, feed flow rate, and product purity in a single-stage membrane system are shown in Fig. 5 (26). In this example a one-stage system as above is considered. The objective is to remove carbon dioxide by collecting this gas at the permeate side, and to produce methane at higher purity at the retentate side of the module. As shown in this figure, increasing feed pressure and/or decreasing permeate pressure increase methane recovery and reduce membrane area requirements. This can be explained by the fact that by increasing the feed pressure and/or decreasing the permeate pressure, the partial pressure difference of the fast-permeating compound, carbon dioxide in this case, increases. Therefore, the membrane area required to treat a certain amount of gas as well as the amount of methane lost to the permeate side decrease. Increasing the feed flow rate does not significantly affect the methane recovery but inversely affects the membrane area re-

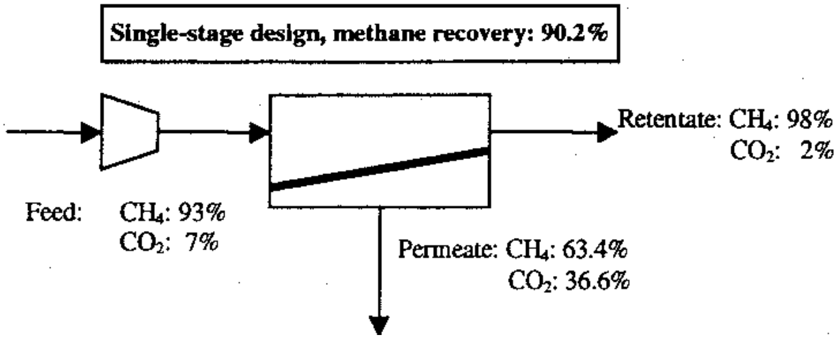


FIG. 4 Single-stage membrane system for treatment of natural gas (reproduced from Ref. 26).

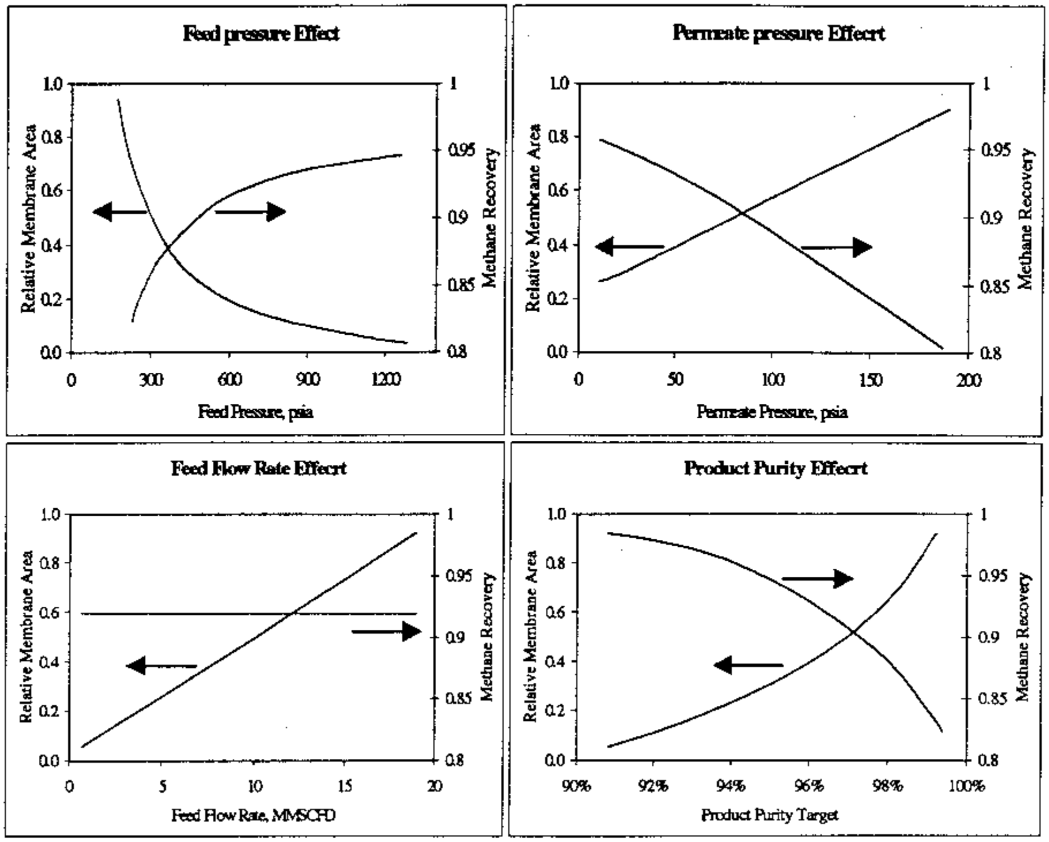


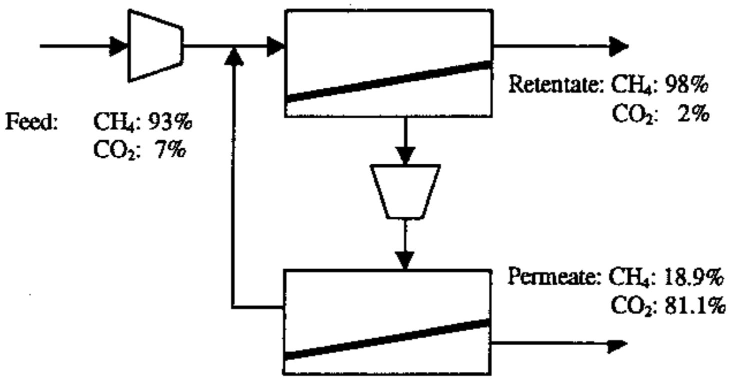
FIG. 5 Effects of selected operating parameters on the performance of a CO_2 removal membrane (reproduced from Ref. 26). Feed composition: 90–97% CH_4 (90% for product purity effect).

quirement. Finally, higher purity targets increase the membrane area requirement and decrease methane recovery. That is because, as more carbon dioxide permeates through the membrane, its partial pressure decreases and, as discussed above, more membrane area would be required. At the same time, the partial pressure of methane increases as more CO_2 permeates through the membrane. Therefore, its permeation rate increases, resulting in higher overall methane loss.

Multistage Membrane Systems

Product purity and/or recovery can be improved by incorporating additional membrane stages into a system. Many different arrangements could be assumed for a multistage membrane system. Different designs must be carefully weighed against their final product cost. In this paper the performances

Two-stage design, methane recovery: 98.7%



Three-stage design, methane recovery: 94.6%

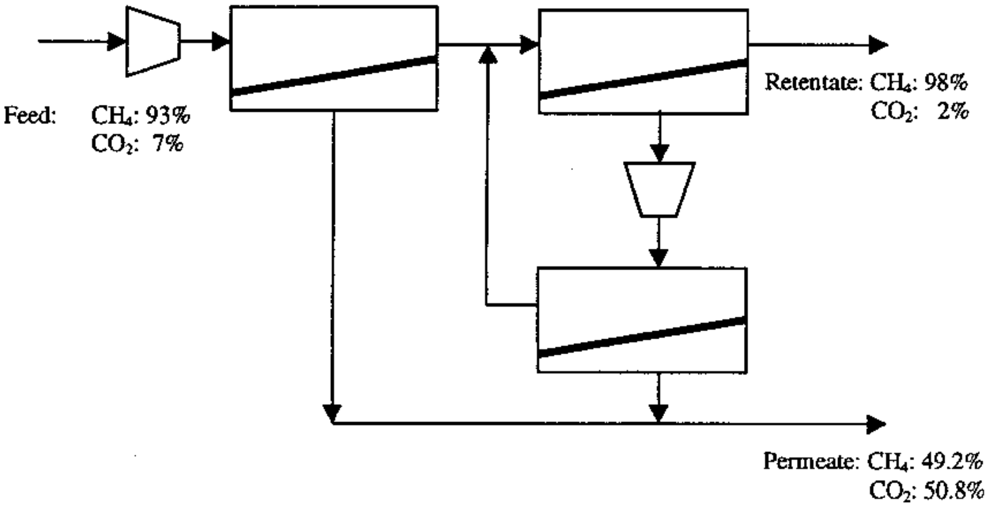


FIG. 6 Two- and three-stage membrane system for treatment of natural gas (reproduced from Ref. 26).

of two- and three-stage membrane systems are shown in Fig. 6 for the same separation as above.

In the two-stage design, methane is recovered as retentate at the same concentration as in the single-stage design. In the two-stage design the CO_2-rich permeate from the first stage is recompressed and fed into the second module. The permeate from the second membrane is highly concentrated in carbon dioxide. Retentate from the second stage, which is depleted in CO_2, is recycled back to the first stage for further treatment. The two-stage design increases methane recovery by reducing the concentration of this gas in the final permeate stream. At the same time, carbon dioxide is collected at a higher concentration compared with a single-stage design, which can be interpreted into an overall smaller amount of methane loss to the waste stream. The two-stage design provides the user with a higher separation factor of 57, and higher methane recovery compared with a single-stage system.

By adding a third membrane module to the above arrangement, methane recovery improves even further. The three-stage design is shown in Fig. 6. In this design the third membrane is located before the other two to remove the bulk of CO_2 from the stream before feeding it to the previous system. Three-stage design is most effective when the concentration of carbon dioxide in the feed is relatively high. In this design, permeate from the first membrane is highly concentrated in carbon dioxide. The retentate, on the other hand, is depleted in CO_2, but still contains significant amount of this gas. The latter stream is then treated as described in the two-stage design example. The overall separation factor in this case is 14, which is higher than the single-stage example but smaller than the two-stage one. The main advantage of a three-stage membrane system is that it reduces the duty of the recycle compressor by removing the bulk of carbon dioxide at high purity at the beginning of the process. This design also reduces the membrane area requirement. A cost and performance comparison between these three processes is shown in Table 2 (26).

Membrane system designs are not limited to the above examples. More sophisticated designs are also available to meet certain separation objectives (26–29). The choice of system design depends on many factors such as product price, environmental regulations, space availability, and membrane characteristics.

The economic comparison between membrane systems and competing technologies indicates that membranes are most cost-effective in applications where high carbon dioxide concentration, e.g., 10–70%, and/or low flow rates are encountered. Babcock et al. (27) compared single and multistage membrane systems with amine treatment for a wide range of operating conditions.

TABLE 2
Process Comparison between Single-, Two-, and Three-Stage Membrane Designs for Treatment of Natural Gas (adapted from Ref. 26)

Membrane design	Methane recovery (%)	Membrane area	Compression
Single-stage	90.2	0.80	—
Two-stage	98.2	1.0	1.0
Three-stage	94.6	0.45	0.5

They concluded that membranes are economical compared to the amine process over a wide range of feed flow rates and acid gas concentrations. Figure 7 compares the process cost of amine and membrane processes at a feed flow rate of 37.2 MMscfd and carbon dioxide concentrations ranging from 5 to 90%. In general, the process cost ($/Msfd feed) for the amine process increases almost linearly with carbon dioxide concentration; it goes through a maximum and decreases at higher CO_2 concentrations for the membrane system. Also, the cost increases with feed flow rate for the amine process while it decreases for membranes. Except at low CO_2 concentrations and low flow rates, a multistage membrane system shows significant superiority to an amine process. In the same study the performances of the two technologies at different feed pressures were compared, and it was concluded that membranes were superior to the amine process at higher pressures and concentrations. The two techniques were equally viable at low pressure (<300 psig) and low CO_2 concentration (<15%).

A similar study conducted by Purgason et al. (30) compared the performance of a single-stage membrane with DEA and membrane/DEA hybrid systems. The results are shown in Table 3. This study concluded that membrane system cost 24% less than the DEA process and 15% less than the hybrid system.

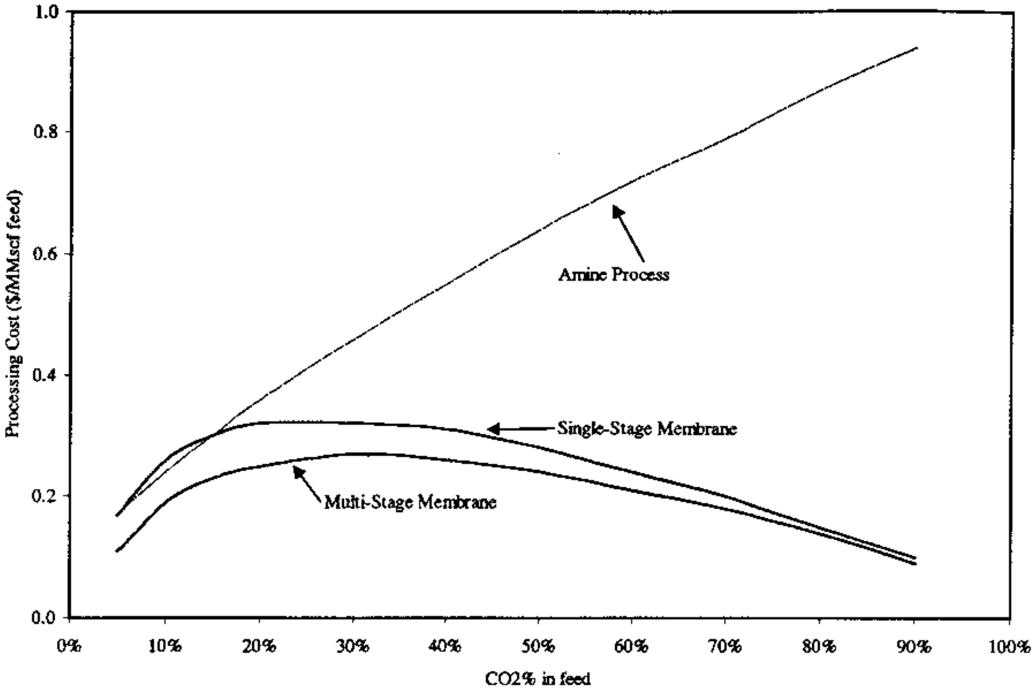


FIG. 7 Cost comparison of amine process and single- and multistage membrane designs (reproduced from Ref. 27).

TABLE 3
Comparison of Amine and Membrane Processes for Natural Gas Treatment (adapted from Ref. 30)

	DEA amine	Membrane	Membrane/DEA hybrid
Relative capital cost	1.0	0.26	0.72
Relative operating cost	1.0	1.51	1.14
Relative net present cost at 15%	1.0	0.76	0.89

SUMMARY

The above discussions can be summarized in the following points.

1. Pressure difference alone does not define the final product purity and flow rate. Permeate pressure (or pressure ratio) also plays a significant role.
2. Single-stage membrane systems are efficient as bulk separator. Efficiency decreases as purity closer to 100% is targeted. For higher purities and lower costs, a multistage system or a hybrid system must be carefully weighed against a single-stage system.
3. There are inherent trade-offs in membrane designs between membrane cost, compressor cost, and product loss and purity. Smaller product loss is always accompanied by higher membrane module and compressor costs.
4. In most applications, membrane hybrid systems are more economical for achieving high product purity compared with stand-alone processes.

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