A Critical Review About the Challenges, Opportunities and Future Directions of Membrane Technology for Natural Gas Purification

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ABSRTACT:

Natural gas, a crucial and rapidly expanding energy resource, requires effective purification to minimize environmental hazards and meet quality standards for pipeline transmission and distribution. Among the various purification technologies, membrane technology stands out as an attractive option due to its cost-effectiveness, low energy requirements, and straightforward fabrication process. It offers significant advantages over traditional methods such as adsorption and cryogenic processes.In particular, membrane-based gas separation using hollow fibers (HF) has seen considerable advancement in recent decades. Hollow fiber membranes are advantageous due to their high specific surface area, reduced maintenance needs, and minimal pre-treatment requirements. However, their application is sometimes limited by their low tensile strength, which makes them susceptible to damage under high-pressure conditions.Braid reinforced hollow fiber (BRHF) membranes address this limitation by incorporating a reinforcing braid to enhance mechanical strength and durability. This reinforcement allows BRHF membranes to withstand higher feed pressures and more aggressive feed conditions without compromising structural integrity. With tensile strengths reaching up to 170 MPa, BRHF membranes represent a promising solution for natural gas purification, offering both enhanced performance and longevity.This review discusses various materials used in fabricating gas separation membranes, including inorganic, organic, and mixed matrix membranes (MMM). It also highlights the potential of BRHF membranes in overcoming the challenges associated with high-pressure applications and aggressive feed conditions, suggesting that they could significantly advance natural gas purification technology.

Keywords:Membrane Technology; Natural Gas Separation; Hollow Fiber Membrane; Braid Reinforced Membrane.

1. INTRODUCTION

Natural gas, a fossil energy source formed deep beneath the Earth's surface, primarily consists of methane (CH_4)—a compound made up of one carbon and four hydrogen atoms. In addition to methane, natural gas contains smaller amounts of other hydrocarbons, known as natural gas liquids (NGLs), and non-hydrocarbon gases such as water vapor and carbon dioxide (CO_2). This versatile energy source is widely used for various applications, including heating, cooking, electricity generation, and as a vehicle fuel.Given the environmental impact of natural gas usage, purification is crucial. The conditioning process removes undesirable components such as acid gases (CO_2 and hydrogen sulfide, H_2S) and water vapor. Effective purification is essential not only to prevent corrosion but also to mitigate the environmental hazards associated with CO_2 emissions. Excess CO_2 contributes to climate change, which poses risks to human health and can lead to severe consequences like flooding.In response to these challenges, research into membrane-based gas purification technologies has gained significant traction. These advanced methods, particularly for separating CO_2 from natural gas, offer a more environmentally friendly and energy-efficient approach to purification.

Table 1. Composition of raw natural gas and pipeline specifications [1,2].

Components	Formula	Composition (mol%)	Maximum pipeline specification	Composition
Methane	CH_4	70–90	Methane	75-nonemol%
Ethane	C_2H_6	0–20	Ethane	10mol%
Propane	C_3H_8	0–20	Propane	5mol%
N-Butane	C_4H_{10}	2.54	N-Butane	2mol%
Carbon dioxide	CO_2	0.1–5	Carbon dioxide	2–3mol%
Nitrogen	N_2	0–5	Nitrogen	3mol%
Oxygen	O_2	0–0.2	Oxygen	0.01mol%
Hydrogen sulphide	H_2S	0–5	Hydrogen sulphide	0.25–0.3g/100scg
Rare gases	Ar,He,Xe,Ne	trace	Water vapor	4.0–7.0lb/MMscf

Various technologies, including metal-organic frameworks (MOFs), carbon nanotubes, mesoporous silica, and carbon molecular sieves, are available for industrial-scale natural gas purification to remove CO2. These technologies, which include adsorption, absorption, and membrane separation, each have distinct advantages and drawbacks. Among these, membrane separation has emerged as a practical option due to its cost-effectiveness, safety, low energy consumption, and minimal need for supervision. However, polymeric membranes often face a trade-off between selectivity and permeability, leading to Robeson's Upper Bound. To address this issue, inorganic fillers like zeolites and MOFs are introduced into polymer matrices to create mixed matrix membranes (MMMs). MOFs, with their 3D networks and porosity, are particularly promising due to their compatibility with polymers and strong interactions with polymer chains.

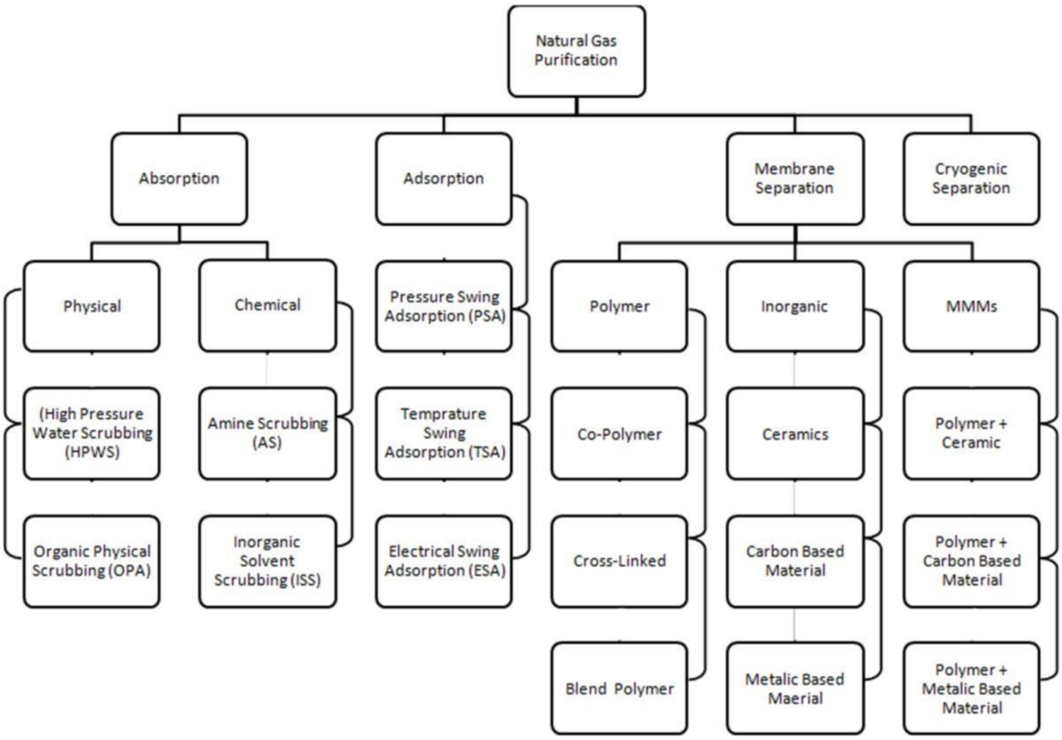
In recent years, hollow fiber (HF) membranes have gained commercial interest for various applications, including water purification, bio-separations, and gas separation, due to their high selectivity and large surface areas. However, HF membranes made by immersion-precipitation techniques often suffer from low mechanical strength and can be damaged under high pressure or airflow. To improve their durability for high-pressure applications, reinforcing HF membranes with a stronger tubular braid or coating a separation layer on such braids has proven effective. The concept of braid-reinforced hollow fiber (BRHF) membranes, introduced by Cooper et al. and further developed by Hayano et al. and Lee et al., involves embedding braided or fibrous materials into membranes to enhance their mechanical strength. This review focuses on how these membrane technologies, particularly MMMs and BRHF membranes, can improve separation performance in natural gas purification, and discusses the impact of polymer, braid, and spinneret types on membrane performance.

2. General Processes of Gas Purification

Natural raw gas primarily consists of methane (CH4) along with lighter gases like butane (C4H10), propane (C3H8), and ethane (C2H6), as well as corrosive gases such as hydrogen sulfide (H2S) and carbon dioxide (CO2) [1]. Traditional methods for removing these corrosive gases include reactive absorption, solid bed absorption, and physical absorption, which are widely used in the industry. While these techniques offer several benefits, they also face challenges such as high operational and capital costs. A critical step in natural gas purification is the extraction of CO2, which must be reduced to less than 2% to prevent corrosion and damage to pipelines. Consequently, CO2 separation technologies are a major focus of research [21]. Selecting the appropriate technology depends on balancing economic and efficiency considerations for specific applications. Natural gas is refined through various stages using absorption, adsorption, cryogenic separation, and membrane technology [20]. These methods are effective for CO2 separation, but high concentrations of contaminants like H2S may require a pre-upgrade stage. Among these methods, membrane separation stands out for its energy efficiency and lower processing costs [22]

Flowchart 1. Natural gas purification technologies [23]. Reprinted/adapted with permission from Ref. [23]. Copyright 2022, Elsevier.

2.1 Absorption

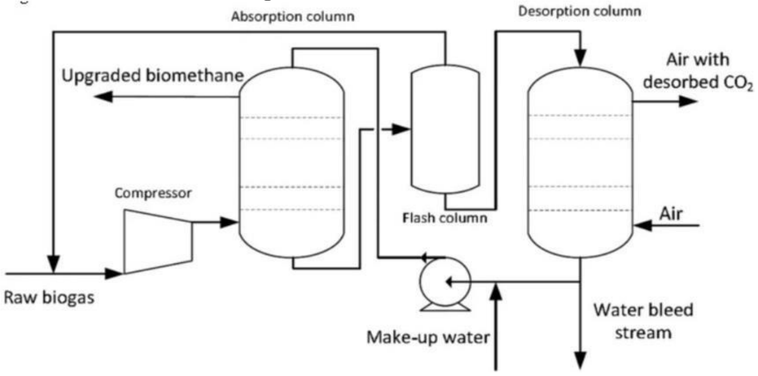


Principle: Absorption relies on the solubility of different gas components in a liquid solvent. In a packed column, raw gas interacts with the solvent, where CO_2 has higher solubility than CH_4, leading to CH_4-rich gas exiting the column.

Types: Absorption is categorized into physical (organic and high-pressure water scrubbing) and chemical (inorganic solvent and amine scrubbing) methods.

2.2 High-Pressure Water Scrubbing

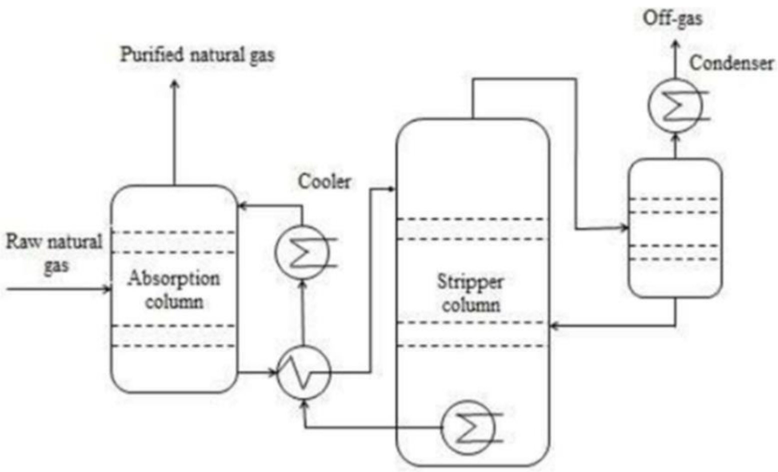
Process: High-pressure water scrubbing is used to remove CO_2 and H_2S from raw gas by exploiting the



higher solubility of these gases in water compared to CH_4. The gas is introduced at 10 bar pressure, and water flows counter-currently to absorb CO_2.

Efficiency: The process is efficient, with high CH_4 recovery (>97%), but requires significant energy for water regeneration, leading to higher operational costs.

2.3 Chemical or Amine Scrubbing

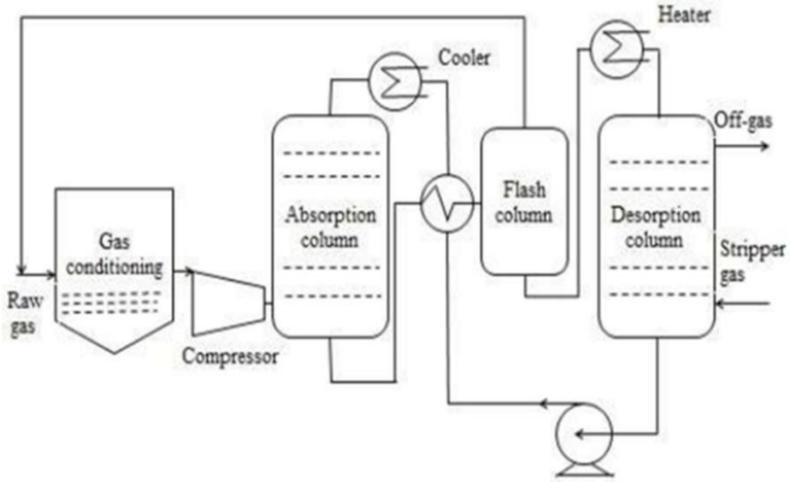


Process: Involves a reversible chemical reaction between amine solvents (like MDEA, MEA, DEA) and acidic gases (CO_2, H_2S). A mixture of MDEA and piperazine (AMDEA) is often used for enhanced CO_2 absorption.

Operation: The process includes an absorber where CO_2 is absorbed from natural gas and a stripper where CO_2 is released from the amine solution using heat.

Challenges: The process requires substantial heat for amine regeneration, and issues like corrosion, contamination, and waste treatment complicate operations.

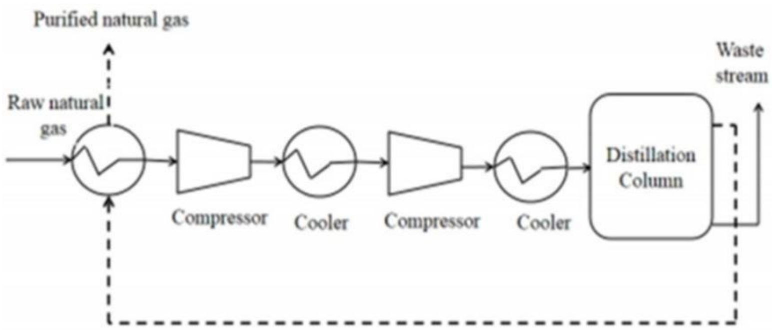
2.4 Organic Physical Scrubbing (OPS)



Process: Similar to water scrubbing but uses organic solvents like PEG, NMP, or methanol. These solvents have higher CO_2 solubility than water, reducing solvent recirculation volume.

Efficiency: OPS is more efficient in CO_2 removal than water scrubbing but involves higher costs and energy requirements for solvent regeneration.

2.5 Cryogenic Separation

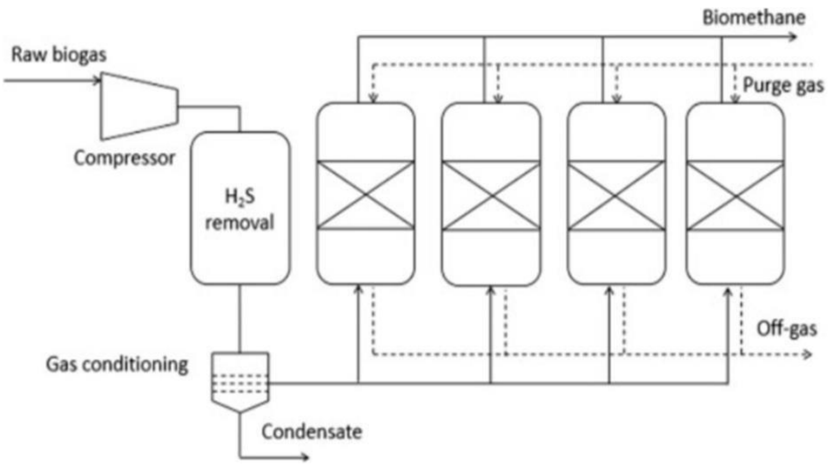


Principle: Utilizes the different boiling points of gases to separate CO_2 and CH_4 under high pressure (80 bar) and low temperature (-170°C).

Stages: Involves multiple stages, including cooling, liquefying, and removing CO_2. The process is energy-intensive and costly due to the use of complex equipment like distillation columns and compressors.

Applications: Useful in producing liquefied natural gas (LNG) and liquefied biomethane (LBM).

2.6 Pressure Swing Adsorption (PSA)



Process: A dry technique where gas is separated by adsorption onto porous solids with high specific areas. CO_2 is adsorbed at a faster rate than CH_4, allowing CH_4 to pass through.

Phases: The PSA system operates in four phases: pressurization, feed, blow down, and purge. Multiple columns are used to minimize CH_4 loss and energy consumption.

Adsorbents: Common adsorbents include activated carbons, zeolites, and silica gels. H_2S must be removed before PSA to avoid damaging the adsorbents.

2.7 Membrane Separation

History: The development of synthetic membranes for gas separation began in the 1800s, with significant advancements in the 20th century, leading to commercially viable membranes with high selectivity and permeability.

Principle: Membranes act as barriers, allowing specific gases to pass through based on differences in pressure, concentration, and other factors. The process is modeled by pore-flow and solution-diffusion models.

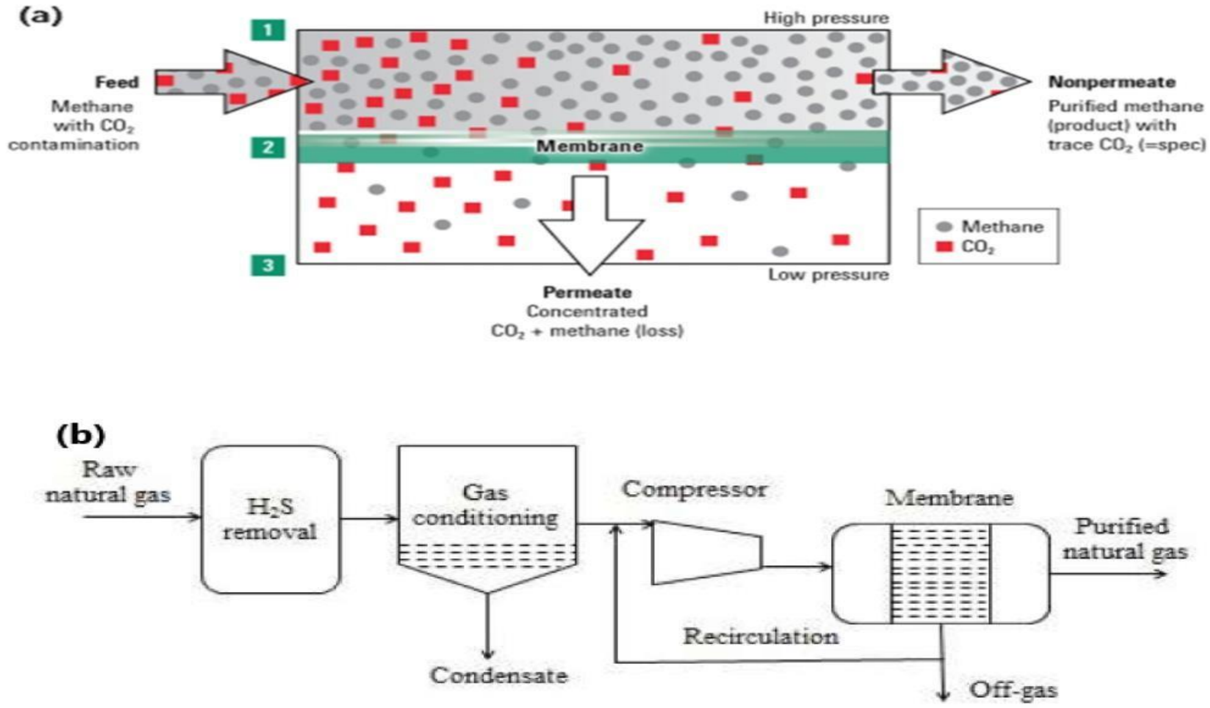
Advantages: Membrane separation is energy-efficient, cost-effective, and can achieve high CO_2 purity with efficient CH_4 recovery.

2.8 Membranes for Gas Purification

Application: Membrane-based technology is widely used in natural gas purification, where CO_2 permeates through the membrane while CH_4 is retained.

Efficiency: This method is advantageous for low gas flow and high CO_2 content, offering better selectivity, lower energy consumption, and efficient CH_4 recovery (up to 96%).

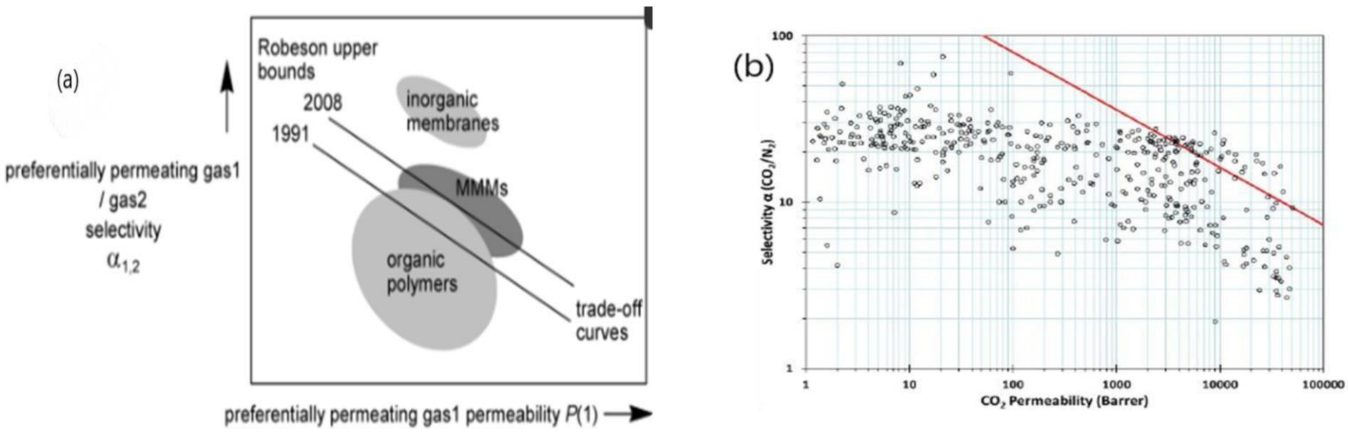
Types: Common membranes include polymeric, inorganic, and mixed matrix membranes, chosen based on their selectivity and permeability.



3. MEMBRANE MATERIALS

In the field of membrane-based gas separations, the primary focus is typically on the permeability (productivity) and selectivity (efficiency) of the membrane for a specific gas separation process. To achieve high permeability and selectivity, various membrane materials have been explored and classified into three main types: polymeric, inorganic, and mixed matrix membranes. The selection of appropriate membrane materials is crucial not only for achieving higher permeability ratios but also for yielding superior permeabilities. The chemistry of the membrane material plays a significant role and must be tailored to the specific separation process. Additionally, asymmetric membrane configurations are particularly advantageous for industrial applications.

3.1 POLYMERIC MEMBRANES



Polymeric membranes, made from organic materials such as cellulose acetate (CA), polysulfone (PSF), polydimethylsiloxane (PDMS), polycarbonate (PC), and polyimide (PI), are the most widely used in commercial applications. These membranes are valued for their ease of fabrication, high selective permeation, and strong mechanical properties. For example, a CH_4 purity of 98% was achieved using a polyvinyl amine/polyvinyl alcohol blend membrane. The first commercialized polymeric membrane for gas purification was the CA membrane, which effectively removes CO_2 and H_2S. Cellulose acetate is cost-effective due to the abundant availability of cellulose and its excellent separation properties. However, CA membranes have limitations, such as susceptibility to plasticization (plasticization pressure $\approx$ 8 bar), where the –OH functional group facilitates CO_2 dissolution in the membrane matrix, lowering gas mixture selectivity below ideal levels. Polydimethylsiloxane (PDMS) is another promising material due to its higher gas permeability compared to other synthetic polymers. The permeability of CH_4 and CO_2 in PDMS is notably higher due to its side chain configurations and compositions. However, PDMS membranes suffer from low separation factors and mechanical strength. Despite extensive research and the synthesis of many polymers, only a few have reached the commercial market. Most commercial membranes are polymer-based with low permeability and high selectivity. However, low permeability limits their suitability for large-scale gas treatment, such as flue gas treatment. The fabrication of membranes with both high permeability and selectivity is challenging due to the trade-off between these properties. This trade-off was first highlighted by Robeson and is evident in the well-known log-log plot where gas pair (CO_2/N_2) selectivity is plotted against the permeability of the more permeable gas (CO_2).For instance, a suitable membrane for capturing CO_2 from a flue gas power plant would require a polymer with at least 1000 barrer permeability and a CO_2/N_2 selectivity above 30. Currently, only a few polymers approach these targets. Among newly synthesized polymers, thermally rearranged polymers and polymers of intrinsic microporosity are the most promising. However, issues such as physical aging and costly multistep synthesis need to be addressed before they can be used in industrial processes.In natural gas separation, state-of-the-art polymeric membranes can be economically competitive with conventional technologies regarding operating and capital costs. Despite showing promising results in gas separation, polymeric membranes have significant drawbacks, including low selectivity, necessitating multi-stage separation systems, which increase capital costs. Moreover, polymeric membranes often deteriorate under extreme conditions of high pressure and temperature, mainly due to chain swelling in the presence of corrosive feed components. Other challenges include compaction, aging, and plasticization. Membranes with extremely high permeability are typically accompanied by low gas pair selectivity.

Table 2. Commercial membrane materials as well as their selectivities for impurity removal from natural gas [78]. Reprinted/adapted with permission from Ref. [78]. Copyright 2022, Elsevier.

Components likely to be permitted	Preferential polymeric material category	Polymers utilized	Selectivities over methane
H2S	Rubbery	ether-amideblockco-polymer	20–30(%)[a]
CO2	Glassy	Polyimide,CA,perfluoropolymer	10–20(%)[a]
N2	Rubbery	Siliconrubber	0–3(%)[a]
	Glassy	Perfluoropolymer	2–3(%)[a]
C3+hydrocarbons	Rubbery	Siliconrubber	5–20(%)[a]

α selectivities are typical of those that are measured with high pressure containing natural gas.

Table3. Characteristics, disadvantages and types of polymeric membranes.

Characteristics	(1) Polymer is flexible and soft in a rubbery state while it is hard and rigid in a glassy state. (2) When compared to rubbery membranes, glassy membranes have high glass transition temperature (Tg) and glassy membranes also have high selectivityCO_2/CH_4[79].
Disadvantages	(1) While handling Carbondioxide, they might experience plasticization problems. (2) Swelling of the polymer network in the membrane will occur and also segmental mobility increases when the membrane is exposed to CO_2 which in turn results in an increase in permeability of all the components of gas[80]. (3) Because of this phenomenon, components of gas having characteristics of low permeability will experience high permeability hence the membrane selectivity decreases [70].
Examples	Cellulose acetate, poly sulfones, poly dimethyl siloxane, poly ether sulfone, poly ethylene, polyimide, polyether, poly pyrrolones etc

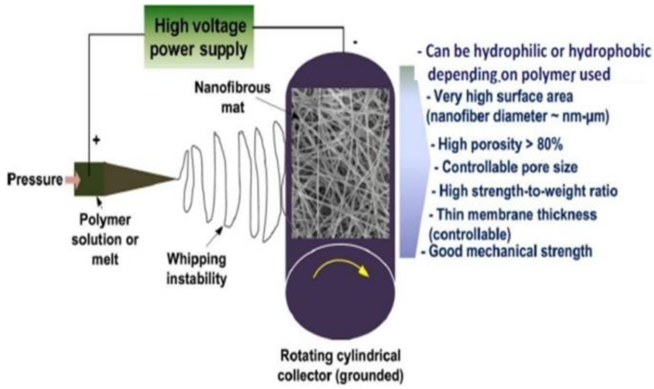
3.2 INORGANIC MEMBRANES

Inorganic membranes are considered more advantageous than conventional polymeric membranes due to their greater thermal stability, chemical resistance, and mechanical strength. These membranes are typically made using materials such as zeolites, carbon molecular sieves (CMS), metal-organic frameworks, and ceramics. Compared to polymeric membranes, inorganic membranes exhibit higher selectivity and gas fluxes. For example, CMS and zeolites have higher selectivity and diffusivity than polymeric membranes, with excellent selectivity due to well-defined shape and size discrimination, leading to a narrow pore size distribution. Most inorganic membranes exceed the Robeson upper bound in terms of selectivity and permeability. Inorganic membranes have several advantages, including solvent resistance under high-pressure conditions and stability at high temperatures. However, they also have drawbacks, such as high fabrication and operational costs, low surface area per unit volume, and difficulties in transforming them into modules with large surface areas for industrial use. The fabrication of inorganic membranes is challenging and requires continuous monitoring due to their delicate structure. Despite their excellent gas separation properties, rigid materials like zeolites and CMS face challenges in forming continuous, defect-free membranes suitable for practical applications. Because both polymeric and inorganic membranes have limitations, researchers have been motivated to develop new membrane materials, leading to the creation of mixed matrix membranes (explained in Section 4). In general, H_2S can negatively affect the performance of the membrane, making its pre-removal necessary. The process for purifying natural gas with membrane technology involves removing oil droplets, water, and aerosols by a filter before the gas enters the membrane unit. A system capable of removing both CO_2 and H_2S from raw gas, while also tracing impurities using different membranes, is needed. Compared to single-stage processes, multi-stage processes have lower operating and investment costs while providing high CH_4 purity. For example, Xiao et al. (2015) identified that using a multi-stage process can improve CH_4 recovery from 80% to 99.5%.

4. General Membrane Fabrication Procedures:

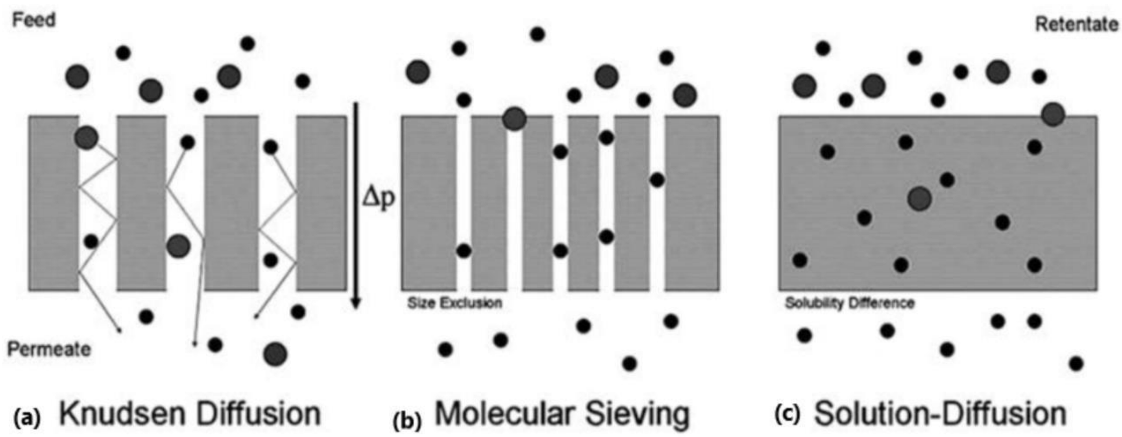
Phase Inversion: The most common method due to its scalability and flexibility. It involves transforming a polymer solution into a solid phase, typically through immersion in a coagulation bath or by thermally induced phase separation.

4.1 Electrospinning:



This technique creates micro/nanofibers by applying strong electric fields to a polymer solution or melt, resulting in high surface area and porosity, making it a viable alternative to phase inversion for certain applications.

4.2 Main Permeation Mechanism:



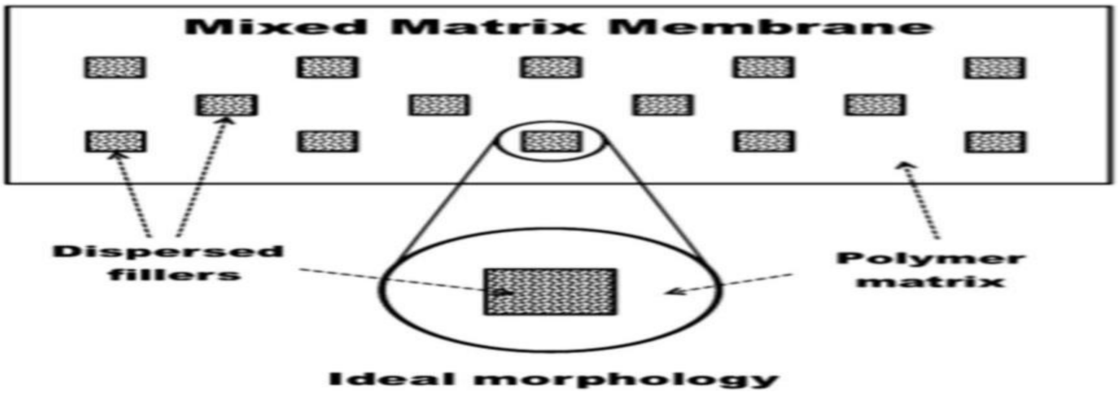
Membrane separation is based on different transport mechanisms depending on the membrane's morphology:

1. **Knudsen Diffusion**: Occurs in porous membranes with small pores.

2. **Solution Diffusion**: Common in polymeric membranes and involves solubility, diffusion, and desorption processes.

3. **Molecular Sieving:** Used in zeolites and CMS membranes, where gas permeation is controlled by the ratio of molecular size to micropore diameter.

4.3 Mixed Matrix Membranes (MMMs):



Composition: MMMs combine inorganic fillers and polymer matrices to enhance mechanical strength, separation performance, and cost-efficiency.

Fillers: These can be inorganic, organic, or both, influencing the membrane's permeability and selectivity. Common fillers include zeolites, carbon nanotubes (CNT), metal-organic frameworks (MOFs), and more.

Fabrication Techniques: The document details various methods of incorporating fillers into polymer matrices, such as surface modifications and sonication, to improve adhesion and prevent defects.

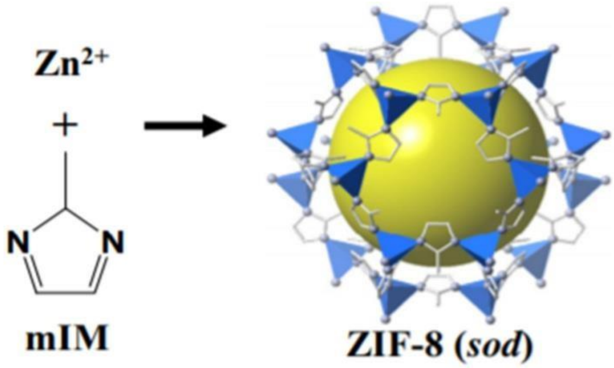
Table 5. Promising MMMs for purification of natural gas.

Material	P_{CH4}	P_{CO2}	$\alpha_{CO2/CH4}$	**References**
Pure Matrimid	0.21	7.29	34.71	[9,158]
Matrimid+MOF-5	0.45	20.20	44.89	[9,138]
Matrimid+CMS	0.24	12.60	52.5	[9]
PurePSf	0.22	6.30	28.64	[159,160]
PSf+AlPO	1.30	51.00	39.3	[160,161]
PureABS	0.12	2.87	24.10	[162]
ABS+AC-2	0.41	20.50	50.10	[163]

Table 6. Gas separation performance of MMMs in comparison to pristine polymeric membranes.

Polymer	Filler used	Filler loading (wt%)	Gaseous pair	Pure polymeric membrane		Matrix membranes	
				Permeability (GPU)	Selectivity	Permeability (GPU)	Selectivity
Polysulfone	ZIF-8	1	CO2/CH4	21.4	19.5	31.3	13.5
Polysulfone	MIL-125(Ti)	20	CO2/CH4	9.3	22	29.1	29.5
Matrimid®	SAPO-34	20	CO2/CH4	4.3	34	6.8	67
6FDA-ODA	UiO-66	7	CO2/CH4	25.8	20.2	43.3	56.9
PDMS	4A	50	H2/CH4	1200	0.8	13,700	14.7
Pebax1657	ZIF-8	8	CO2/CH4	130	9	450	15
Polyethersulfone	SAPO - 34	20	CO2/CH4	0.9	32.2	2.1	40.5
Matrimid®	ZIF-8	10	H2/CH4	34	32	25	50
6FDA-durene	ZIF-8	42	CO2/CH4	256	19.4	779	20.8
Pebax1657	SAPO - 34	50	CO2/CH4	110	18	320	18

4.4 **Zeolite Imidazolate Frameworks (ZIFs) in MMMs:**



Advantages: ZIFs, a subclass of MOFs, offer benefits like high selectivity, structural stability, and gas adsorption capabilities, making them promising fillers for MMMs.

Applications: ZIFs show potential for gas separation and storage, particularly for CO_2, due to their strong metal-ligand interactions and thermal stability.

5. SEPARATION PERFORMANCE OF MIXED MATRIX MEMBRANES:

1. **Permeability and Free Volume**: In MMMs, permeability is influenced by solubility (S) and diffusivity (D). The diffusion coefficient of penetrants is affected by the free volume in the polymer matrix. The equation provided (Equation 1) indicates that increased free volume can enhance the diffusion of penetrants, as demonstrated by the increase in permeability of N2 with higher loading of FS (Fumed Silica) in a PTMSP/FS MMM.

2. **Solubility and Interaction**: The solubility coefficient of penetrants in MMMs depends on the interaction between the filler and the polymer. Functional groups on fillers and polymers can interact with gases, increasing their solubility. The relationship is described by the van't Hoff equation (Equation 2), where increased interaction between penetrants and functional groups leads to higher solubility and, consequently, increased gas permeability. For instance, increased silica loading in a poly(amide-6-b-ethylene oxide) and silica MMM enhanced CO2 solubility due to strong interactions between CO2, SiO2, and the polyimide block in PEBAX.

6. Future Prospects and Concluding Remarks

Membrane-Based Gas Separation: Among various gas separation methods, membrane-based processes are highlighted for their simplicity, environmental friendliness, and ease of use. Research has shown that spinning parameters play a crucial role in fabricating defect-free membranes.

Nanoparticles and Membrane Performance: Nanoparticles, particularly ZIFs (Zeolitic Imidazolate Frameworks), are noted for their superior CO2 adsorption capacity compared to other MOFs. Hollow Fiber (HF) membranes benefit from a high specific surface area and lower maintenance needs, although high pressure can pose a challenge.

Braid Reinforced Hollow Fiber Membranes (BRHF): To address mechanical strength issues in HF membranes, braid reinforcement is utilized. Braid reinforced membranes have shown promise in various applications, including wastewater treatment through Membrane Bioreactor (MBR) technology. Research in this area has been increasing, revealing a growing trend in using BRHF membranes for Microfiltration (UF), Reverse Osmosis (RO), and Nanofiltration (NF).

Future Research Directions and Concluding:

1. **Gas Separation Applications**: Investigate the use of BRHF membranes in gas separation, particularly for natural gas purification. Their high mechanical strength and ability to handle high feed pressures make them suitable for this application.

2. **Fabrication Techniques**: Explore advanced fabrication techniques such as grafting and blending to improve BRHF membranes. Current research is limited in this area.

3**. ZIF-Based Fillers:** Conduct studies on incorporating ZIF-based fillers into BRHF membranes to assess performance improvements in gas separation applications.

4. **Diverse Applications**: The potential applications of BRHF membranes are vast, and future research could expand their use into various fields beyond those currently explored.

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