

Review

A Review on Hollow Fiber Membrane Contactors for Carbon Capture: Recent Advances and Future Challenges

Rouzbeh Ramezani ¹, Luca Di Felice ¹ and Fausto Gallucci ^{1,2,*}

¹ Sustainable Process Engineering, Department of Chemical Engineering and Chemistry, Eindhoven University of Technology, De Rondom 70, 5612 AP Eindhoven, The Netherlands

² Eindhoven Institute for Renewable Energy Systems (EIRES), Eindhoven University of Technology, De Rondom 70, 5612 AP Eindhoven, The Netherlands

* Correspondence: f.gallucci@tue.nl

Abstract: Energy need is predicted to increase by 48% in the next 30 years. Global warming resulting from the continuously increasing atmospheric CO₂ concentration is becoming a serious and pressing issue that needs to be controlled. CO₂ capture and storage/use (CCS/CCU) provide a promising route to mitigate the environmental consequences of CO₂ emission from fossil fuel combustion. In recent years, hollow fiber membrane contactors are regarded as an advanced technique with several competitive advantages over conventional technologies such as easy scale-up, independent control of flow rates, more operational flexibility, absence of flooding and foaming as well as high interfacial area per unit volume. However, many factors such as the membrane material selection, proper choice of solvent, and membrane module design are critical to success. In this regard, this paper aims at covering all areas related to hollow fiber membranes, including membrane material, membrane modification, membrane surface modification, shape, solvent characterization, operating parameters and costs, hybrid process, membrane lifetime, and energy consumption as well as commercially available systems. Current progress, future potential, and development of pilot-scale applications of this strategy are also assessed carefully. Furthermore, pore wetting as the main technical challenge in membrane contactor industrial implementation for post- and pre-combustion CO₂ capture processes is investigated in detail.

Keywords: CO₂ capture; microporous membrane; membrane contactor; gas separation; hollow fiber



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1. Introduction

Economic development and global population growth have enormously contributed to excessive energy consumption worldwide [1]. Fossil fuels are recognized as a major energy source for the near-to-middle term, although their combustion undoubtedly generates large quantities of carbon dioxide into the atmosphere [2]. The greenhouse gas emissions depending on energy conversion are estimated to increase by approximately 30% by 2040 [3]. This massive release of CO₂ due to human activities has caused severe environmental consequences, such as rising sea levels and global warming, and its impact on climate change has become increasingly apparent in recent years [4]. Therefore, it is crucial to implement suitable and reliable processes to manage CO₂ emissions from various industrial gas streams. Post-combustion capture, pre-combustion capture, and oxyfuel combustion are three main strategies to decrease CO₂ emissions from the use of fossil fuels [5]. Currently, the technologies for CO₂ capture from gas mixtures mainly include chemical absorption, physical adsorption, and cryogenic, membrane-based, and calcium looping processes [6,7]. Of all carbon capture approaches, absorption using amine solutions in a packed column is the most common deployed separation technique available due to its high separation purity and fast absorption rate [8]. However, operating costs and energy consumption are still high [9]. Aside from the cost, amine scrubbing suffers from several operational

limitations such as entrainment, foaming, and flooding [10]. In this regard, a promising alternative to conventional absorption technology is membrane gas-liquid contactors, where acid gases are absorbed into selective absorption liquid via mass transfer through a porous, non-selective membrane [11]. Today, membrane technology is accepted in various industries thanks to its technical achievements, such as efficiency and operational simplicity, high selectivity, permeability for the transport of specific components, low energy requirement, stability under different process environments, easy control, easy scale-up with high flexibility, and compatibility between different membrane operations in integrated separation processes [12,13]. The development of an efficient membrane separation process normally includes (i) materials science and engineering; (ii) development of membranes materials; (iii) membrane synthesis and modification; (iv) module design and manufacturing; (v) process engineering and integration of membrane processes in industrial processes; and (vi) cost analysis, economic analysis, and environmental and safety assessments. The membrane industry by itself shows a promising future, which is already illustrated by its growth rate (stable at around 10% for several years now), diversity of applications, and the various feasible developed membrane-based processes such as membrane contactors [14]. Membrane contactors combine the intrinsic merits of membrane separation technology with those of chemical absorption [15]. Membrane modules are generally designed as low-cost hollow fibers, which allow a high interfacial area (up to $10,000 \text{ m}^2/\text{m}^3$) significantly greater than most traditional absorbers' columns (at best $80\text{--}800 \text{ m}^2/\text{m}^3$), which results in high overall mass transfer rates [13]. This approach was first utilized by Qi and Cussler [16,17] in 1985 for CO_2 removal using a microporous polypropylene membrane. Figure 1 shows a schematic diagram of the gas transport process inside the membrane module.

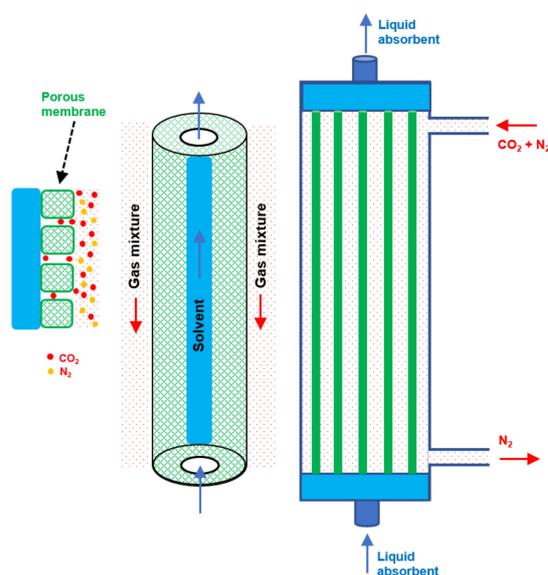


Figure 1. Schematic diagram of a single hollow fiber membrane contactor and membrane contactor module.

A liquid absorbent is fed to one side of the membrane while a gas mixture is supplied to the opposite side. Due to its modularity, CO_2 from the gas mixture can diffuse from the bulk gas phase to the surface of the membrane, transport through the membrane pores, and reach the liquid phase. Compared with the membrane-based gas separation technique, the materials that are incorporated in membrane contactors generally have no separation selectivity [18]. Instead, a porous membrane is employed as a nondispersive barrier that provides the separation between the gas and the liquid phases while significantly enhancing the effective contact surface between two phases without dispersing one phase into the other [19]. Different phases on each side of the membrane can be regulated independently,

which increases the process's flexibility. Existing resistance to the mass transfer is offered by the existing phase(s), the membrane, or a combination of both. To ensure a high gas flux across the membrane, the membrane needs to be highly porous. Additionally, one main requirement for a successful gas separation membrane is the compatibility of the absorption liquid and the membrane material [20]. Polyimide (PI), polyethersulfone (PES), polysulfone (PS), polytetrafluoroethylene (PTFE), polyvinylidene-fluoride (PVDF), and polypropylene (PP) are the most widely applied membrane materials while 2-amino-2-methyl-1-propanol (AMP), methyldiethanolamine (MDEA), monoethanolamine (MEA) and diethanolamine (DEA) have been widely considered as liquid absorbents for CO₂ absorption in membrane contactors.

According to Falk-Pedersen et al. [21], the weight and dimensions of the membrane contactor can be reduced by 66% and 72%, respectively, compared to conventional absorption columns. Other unique advantages of membrane hollow fiber contactors include reduced capital costs, operational flexibility, easy scale-up, and lower energy consumption [22]. Thanks to the advantages of hollow fiber membranes, this technique is a viable strategy for different applications including gas separation, food processing, azeotropic mixture separation, medicine, and water treatment, as shown in Figure 2 [23,24].

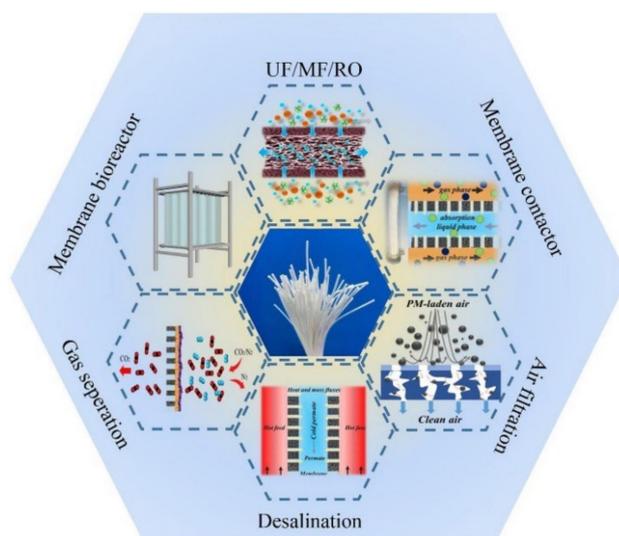


Figure 2. Hollow fiber membranes for different applications. Reprinted with permission from Ref. [24]. 2020, Elsevier.

Membrane crystallizers, gas-liquid membrane contactors, membrane distillation, membrane extractors, and membrane emulsifiers are the main membrane-based processes that might be applied for advanced separation. Gas-liquid membrane contactors have been occasionally mentioned in the literature for the separation of acid gases from different sources such as natural gas, industrial gas, and effluent gas from domestic processes. From this perspective, this review is focused on the development of various types of hollow fiber membrane contactors for CO₂ removal from the point of view of the membrane material, membrane modification, composite membrane, surface modification, membrane module shape, solvent development, operating parameters, hybrid process, and membrane lifetime performance.

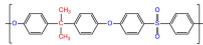
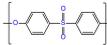
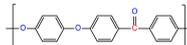
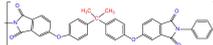
To the best of our knowledge, detailed and comprehensive reviews that cover all relevant properties of hollow fiber membranes have not been reported. This review aims at providing recent research developments in the field of membrane contactors-based carbon capture, as well as a reference for researchers to understand the technological progress further. First, the most important membrane materials employed for the application of gas-liquid membrane contactors, as well as their properties, synthesis procedures, and modification are introduced. Then, a special section features the membrane structure,

including the shape of developed membranes, additives implemented in the membrane matrix, etc. Next, a section is devoted to the solvent type that is used as an absorption liquid. Understanding the operating parameters and their influence on the performance of the membrane process is discussed separately. Gas-liquid membrane contactors as a hybrid process as well as their merits are listed in a different section with in-depth discussion. The final section is assigned to the cost of operation as well as the challenges and lifetime of gas-liquid membrane contactors to provide valuable insights for large-scale practical operations.

2. Membrane Materials

From the point of view of material types, membranes are commonly classified into polymeric and inorganic membranes [25]. Up to now, polymeric materials have been favored for CO₂ capture due to their low manufacturing costs and good performance as well as physicochemical properties, which can be regulated much easier than inorganic membranes. Table 1 shows the chemical structures of common polymers used in membrane contactors. The membranes employed for gas-liquid membrane contactors are typically microporous with symmetric or asymmetric structures. To ensure a successful gas absorption process in hollow fiber membrane contactors for long-term operation, the properties of membrane materials should be well studied. The materials need to be chemically and thermally compatible with the liquid absorbents and operating conditions.

Table 1. Chemical structures of typical polymers for membrane contactors.

Polymer	Chemical Structure	Polymer	Chemical Structure
Polysulfone (PSF)		Polypropylene (PP)	
Polyethersulfone (PES)		Polyvinylidene (PVDF)	
Polyether ether ketone (PEEK)		Polytetrafluoroethylene (PTFE)	
Polyetherimide (PEI)		Polymethyl pentene (PMP)	
Polyethylene (PE)		Polydimethylsiloxane (PDMS)	

Generally, a membrane material with a high contact angle with the solvent is preferable as it decreases the potential for pore wetting, resulting in high mass transfer rates and avoiding mixing between liquid and gas.

Thermal extrusion and phase inversion are the most widely employed techniques for membrane fabrication. Depending on the membrane fabrication method and specific conditions, the contact angle, porosity, and pore size of the membrane can be controlled, and lead to an improved CO₂ absorption flux and removal efficiency. Polymers such as polyetherimide (PEI), polysulfone (PSF), PVDF, PTFE, polyethylene (PE), and PP have been the most popular choices in the literature for CO₂ capture applications. Membranes that have been recently used in the gas separation process are listed in Table 2 along with their preparation method and their characteristics. PEI and PSF membranes have good chemical and thermal stability and dissolve well in common organic solvents. Owing to their great hydrophobicity and superior thermal and chemical stability, PTFE membranes have become very common in the field of gas separation [26]. PVDF membranes show high chemical resistance in contact with corrosive chemicals and have a moderate melting temperature. Low price, excellent processability, high melting temperature, widely adjustable

mechanical properties, and high resistance to water and chemical environments have made PP membranes one of the most widely used membranes in the literature [27].

A performance comparison between PP and PTFE membranes for CO₂ absorption was performed by deMontigny et al. [28]. Two amine solutions including MEA and AMP were also utilized as liquid absorbents. The authors concluded that PTFE exhibits a better resistance than PP when wetting is a matter of importance. Their long-term studies showed that PP membrane performance reduces over time when tested with MEA and AMP solutions, while the PTFE membrane maintains its initial level of performance. This poor performance of the PP membranes compared to PTFE can be explained by the fact that PTFE had a porosity of 50%, while the PP membranes had a porosity of 35%. PEEK membranes have the advantages of high resistance to chemicals and superior mechanical properties as well as a good performance at capturing CO₂ with low CO₂ concentration. Li et al. [29] scaled up PEEK hollow fibers for the CO₂ absorption process. Their measurements revealed that a CO₂ removal rate greater than 90% can be obtained with a large PEEK membrane module. However, the synthesis of porous PEEK membranes is challenging owing to the limited solubility of PEEK in common solvents and the necessity of extreme operating conditions in the melt-spinning process. Furthermore, the mass transfer coefficient in the absorption step was found to be an order of magnitude greater than that of conventional column contactors.

Similarly, the effect of the pore size of PTFE membranes on CO₂ removal was studied by Chen et al. [30]. It was concluded that CO₂ recovery increased with a decrease in the pore size of the PTFE membrane from 1.3 µm to 0.1 µm. The smaller pore size means a decrease in the wetting of the membrane pore. Therefore, it is expected that a pore with a smaller diameter would reveal less wetting with chemical absorbents, which leads to higher CO₂ recovery. Furthermore, an asymmetric PTFE membrane indicated a better CO₂ recovery than a symmetric membrane.

To improve the PP membrane materials that resist wetting, Franco et al. [31] prepared a plasma-treated PP membrane and examined its wetting performance in contact with an MEA solution. They found that untreated PP becomes degraded while the plasma-treated PP membranes are less susceptible to wetting and degradation when coupled with MEA and showed a 30% higher CO₂ mass transfer rate than untreated PP membranes. Since PP materials have a cheaper price than PTFE, treated PP material will remain competitive with PTFE. Li et al. [26] studied the effect of porosity and pore size on the performance of PTFE hollow fibers, and their results revealed that the increase in membrane porosity and pore size in the non-wetting zone improved CO₂ removal efficiency. A larger pore size is beneficial to reduce the mass transfer resistance of the membrane phase. However, it increases the risk of membrane wetting during the absorption process. According to their results, the CO₂ removal efficiency increased as pore size increased from 0.15 µm to 0.18 µm. With a further increase in pore size from 0.18 µm to 0.27 µm, CO₂ removal efficiency decreased, which could be caused by pore wetting.

Hedayat et al. [32] evaluated PVDF and PSF membrane contactors for simultaneous absorption of H₂S and CO₂ from a gas mixture similar to natural gas. Based on their results, less membrane wetting in dense layer PVDF membrane by MDEA solution was observed in comparison with micropores of PSF membrane. Fabrication of porous PEI membranes via wet phase inversion technique using different amounts of polymer in a spinning solution was performed by Bakeri et al. [33,34]. Their investigation outcomes revealed that with a proper selection of polymer concentration in PEI, a greater CO₂ absorption rate than commercial PVDF hollow fiber membrane can be obtained. A thermally-induced phase separation method was applied by Ghasem et al. [35] for the fabrication of PVDF. Membrane performance was tested in gas–liquid membrane contactor modules used in the absorption of CO₂ from CO₂/CH₄ gas mixture in an aqueous 0.5 M NaOH solution. According to their results, CO₂ absorption flux decreases with increasing polymer concentration in the dope solution. The reason for this is that as polymer concentration increases, the outer skin layer of the membranes became denser and thicker. PP and PVDF membranes were suggested

by Lin et al. [36] for CO₂ capture in MDEA blended with PZ solution. The authors claimed that the effect of the viscosity of solvent on the wetting ratio is more significant than that of the contact angle between the membrane and absorbent. Moreover, PVDF showed a greater performance than PP membrane from the point of view of lower pore wetting. Fashandi et al. [37] evaluated the potential of poly(vinyl chloride) (PVC) membranes for the first time for CO₂ absorption in a membrane contactor unit. Low cost, good processability, and acceptable chemical resistance are advantages of PVC as a hydrophobic polymer. Figure 3 shows SEM images of the cross-section and surface of the PVC membrane. The reduction in hollow fiber membrane wall thickness, optimum pore size, and optimum orientation of PVC chains resulted in a good CO₂ absorption performance. According to their result, hollow fiber membranes prepared from PVC polymer enjoy advantages including the hydrophobic nature of PVC, the presence of macrovoids within the PVC structure, the optimum orientation of PVC chains, optimum wall thickness, and the corrugated inner surface of PVC membranes.

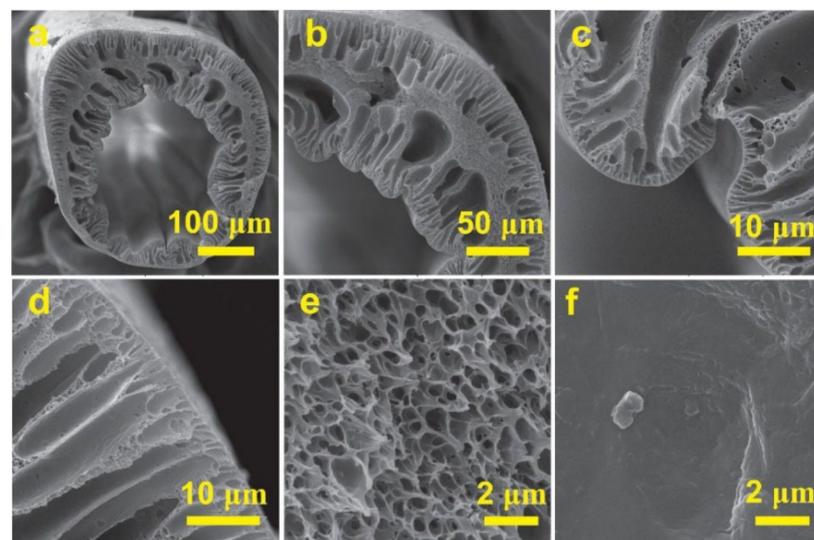


Figure 3. SEM images of PVC hollow fiber membrane, (a) HFM cross-section, (b) magnified HFM cross-section, (c) inner edge of HFM, (d) outer edge of HFM, (e) magnified porous section and (f) outer surface of HFM. Reprinted with permission from Ref. [37]. 2016, Elsevier.

A few researchers studied CO₂ absorption using dual-layer membrane contactors due to their advantages such as higher packing density, self-supporting structure, and self-containing vacuum channel. Figure 4 shows the conceptual schematic of dual-layer flat and hollow membranes. Chen et al. [38] tested dual-layer PVDF in membrane contactors with DEA as an absorbent for CO₂ absorption. It was found that the outer layer of the membrane shows a large finger-like porous structure, while the inner layer is composed of PVDF and hydrophobic modified SiO₂. Their CO₂ absorption measurements showed that dual-layer membrane exhibits higher CO₂ flux and better long-term stability compared to single-layer.

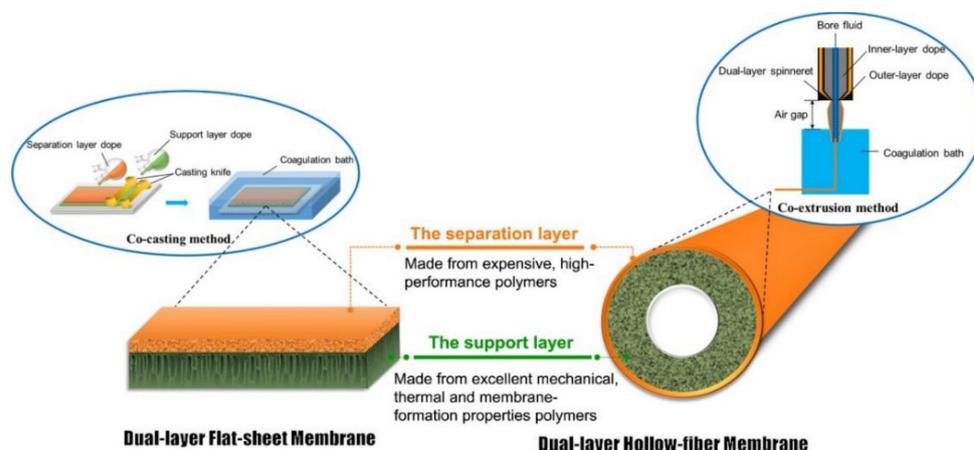


Figure 4. The conceptual schematic of dual-layer flat-sheet and hollow fiber membranes and fabrication methods. Reprinted with permission from Ref. [39].2018, Elsevier.

The stability of the membrane process is critical in the corrosive environment of amine solvents. Although polymeric membranes presented good performance for CO₂ capture, they still suffer from low thermal and chemical stability in contact with aggressive solvents in high-temperature environments, which limits their application for long-time processes in industrial scales where harsh conditions are commonly employed [40]. To overcome such limitations of polymeric membranes, ceramic hollow fiber membranes that showed better mechanical and thermal stability as well as excellent porosity and permeability by controlling the sintering temperature were developed [41]. However, like other types of membranes, these ceramic membranes are not free of shortcomings. They are fragile and expensive, and their preparation is time-consuming and difficult [42]. Lee et al. [43] fabricated hydrophobic modified ceramic hollow fiber membranes by the phase inversion technique and observed that CO₂ removal efficiency can be obtained up to 90% with 30% MEA solution. In another work [44], the same authors examined the effect of temperature on the performance of ceramic membrane contactors. According to their observation, the solvent temperature had no significant effect on CO₂ removal efficiency, while gas temperature showed a greater impact on CO₂ removal efficiency. As the gas temperature increases from 20 to 35 °C at a gas flow rate of 0.75 Nm³/h, CO₂ removal efficiency increases and reaches a maximum value of 71%. With further increases in gas temperature to 50 °C, CO₂ removal efficiency decreases. Koonaphapdeelert et al. [45], for the first time, conducted CO₂ desorption experiments from MEA solution at a temperature of 363 K in a ceramic hollow fiber membrane contactor. The finding of the study proved that even in the region of an ordinary column indicating loading or flooding, the membrane contactors could operate very well. Magnone et al. [46] implemented a ceramic membrane contactor for CO₂ absorption with several single and blended amines. They fabricated ceramic hollow fiber membranes by a phase inversion technique. It was found that the highest absorption flux can be determined by aqueous MEA and DEA solutions. CO₂ separation from biogas using a ceramic membrane contactor was carried out by Lee et al. [47]. The authors reported that their developed ceramic membranes with 96% CO₂ removal efficiency can replace the polymeric membranes or packed columns. Kong et al. [48] assessed SO₂ absorption in a hydrophilic ceramic membrane contactor and concluded that the multichannel ceramic membranes revealed a more compact configuration and better application potential for SO₂ absorption compared to a single-tube membrane contactor.

Table 2. Summary of different membrane materials with their properties studied in the literature.

Material	Pore Size	Porosity	Preparation Method	CO ₂ Flux × 10 ⁻⁴ mol/(m ² ·s)	Ref.
PTFE	N.A.	50%	Commercial, Sumitomo Polymer	0.1	[28]
PTFE	0.15–0.27 μm	44–58%	Paste extrusion-stretching-sintering	0.7–1.3	[26]
PP	0.2 μm	60%	Commercial, Accurel Membrana	N.A.	[49]
PP	0.03 μm	40%	Phase inversion technique	0.5–2.6	[50]
PTFE	0.11–1.3 μm	33–52%	Stretching and heating methods	0.8–1.8	[30]
PP	0.1 μm	20%	Commercial, Parsian Co.	N.A.	[51]
PVDF, PSF	N.A.	50%	Commercial, Ecofine Co.	N.A.	[32]
PTFE	0.48 μm	52%	Commercial, Markel Corporation	3–12	[52]
PP	0.2 μm	50%	Commercial, Memtec	0.4–1.2	[31]
PTFE	2.5 μm	34%	Stretching	N.A.	[15]
PEI	3.9–9.3 nm	72%	Wet phase inversion method	2–2.7	[33]
PS	N.A.	43%	Commercial, VWR Eurolab	N.A.	[53]
PP, PPO	0.27 μm	50%	Commercial, Parker Filtration	N.A.	[19]
PVDF	78–222 nm	67–85%	Arkema	0.5–4	[54]
PVDF	50–470 nm	32–45%	Thermally induced phase separation	1–3	[35]
PP	200 nm	50%	N.A.	1–5	[55]
PSF	N.A.	50%	Commercial, Airrane Co.	N.A.	[56]
PVC	0.89 μm	75%	Phase inversion technique	0.2–1	[37]
PP, PVDF	0.2 μm	40–50%	Commercial, Pall Co.	0.5–3	[36]
PMP	N.A.	30–40%	Commercial, Celgard	N.A.	[57]
PEEK	N.A.	59–71%	Phase inversion technique	0.1–0.3	[29]
Ceramic	68 nm	46%	Phase inversion technique	0.2–8	[43]
Ceramic	N.A.	40%	Phase inversion technique	N.A.	[45]

Table 2. Cont.

Material	Pore Size	Porosity	Preparation Method	CO ₂ Flux $\times 10^{-4}$ mol/(m ² .s)	Ref.
Ceramic	0.9 μ m	49%	Phase inversion technique	5–6.5	[46]
Ceramic	300 nm	54%	Phase inversion spinning	7.5	[47]
Ceramic	N.A.	N.A.	Phase inversion and sintering	0.18	[58]
Ceramic	100 nm	N.A.	Phase inversion spinning	0.1	[48]

The summary of this section is as follows:

- The success of membrane gas absorption over other processes will largely depend on the type of membrane materials and fabrication method used.
- At high temperatures, ceramic hollow fiber membranes have better mechanical and thermal stability than polymeric membranes.
- PTFE offers better resistance than PP when wetting is a matter of importance.
- The plasma-treated membranes exhibited a lower degradation rate and a higher mass transfer rate compared to untreated membranes.
- Pore wetting significantly reduces the CO₂ mass transfer rate, as the mass transfer coefficient through liquid-filled pores is orders of magnitude lower than that if the pores are gas-filled.
- Membrane materials with a high contact angle with the solvent could be resistant to pore wetting.
- A membrane with small pores will resist wetting, but there is a trade-off as smaller pores will also reduce the rate of CO₂ mass transfer.
- From a wetting point of view, hydrophobic membrane materials are preferred due to their relatively high contact angle.
- PP and PE membranes can be chemically degraded during long-term operation in contact with an amine-based solvent.

3. Membrane Modification

The low mass transfer resistance, small pore size, good wetting resistance, and high surface porosity are the essential requirements of membranes, resulting in a high absorption performance through membrane contactors in a long-term stable operation. Using thermal or stretching techniques to prepare PTFE, PE, and PP membranes leads to a symmetrical structure with low porosity and large pore size. On the other hand, PVDF also has the limitation of difficulty in fabrication and small critical surface tension, which restricts its application. As a result, modification of the membranes should be considered to improve their properties and morphology. Modification of polymers through the introduction of organic and inorganic additives, surface treatment, surface coating, and blending with other polymers has been evaluated to develop new membranes which offer higher performance. The opportunity to prepare better membranes for a specific application such as gas–liquid membrane contactors has been recently studied by several researchers (See Table 3), and thus some remarkable results have been reported regarding the preparation and development of membranes.

Table 3. Characteristics of the different membrane materials studied in the literature.

Material	Additive	Pore Size	Preparation Method	CO ₂ Flux × 10 ⁻⁴ mol/(m ² ·s)	Ref.
PVDF, PEI	PEG	0.14–0.35 µm	Spinning process	3.5–4	[59]
PVDF	LiCl, Glycerol, (PEG-400), Methanol, Phosphoric acid	0.036–4.42 µm	Wet spinning process	4.03	[60]
PVDF	LiCl	0.014–0.43 µm	Wet-spinning Process.	1.61	[61]
PEI	Ethanol, Glycerol, Acetone	9.02–26.84 nm	Phase inversion method	1–8	[62]
PSF	Glycerol	47–373 nm	Wet spinning process	2.9	[63,64]
PSF	Glycerol, PEG200, Ethanol, Acetic acid	3–10.52 nm	Phase inversion method	0.98	[65]
PVDF	LiCl	3.96 nm	Wet spinning process	4.1	[66]
PVDF	Glycerol, phosphoric acid, ethanol, PEG-400	5.22–9.62 nm	Commercial: Arkema	7.8	[67]
PVDF	phosphoric acid, LiCl	5.66–9.46 nm	Wet spinning method	5.4	[68]
PVDF	Glycerol	0.07–0.1 µm	Dry–wet phase inversion method	3–8	[69]
PEI, PVDF	Glycerol	0.09–0.05 µm	Wet spinning method	1.5	[70]
PEI	MMT	44–331 nm	Wet phase inversion method.	2.35	[71]
PVDF	Water, Glycerol, Phosphoric acid	9.25–20 nm	Commercial: Elf Autochem	3–7	[72]
PVDF	LiCl, glycerol	5–25 nm	Commercial: ARKEM	N.A.	[73]
PEI	Water, Glycerol, Acetic acid, Ethanol, Methanol	101–140 nm	Wet-spinning method	0.9–1.7	[34]
PES	Water, methanol, ethanol, glycerol, acetic acid, acetone	160–630 nm	Commercial: Arkema	0.5–4.5	[74]
PVDF	Ethylene glycol	1 µm	Dry-jet wet phase inversion	N.A.	[75]
PVDF	PVP, LiCl	N.A.	Phase inversion technique	N.A.	[76]
PVDF	LiCl and water	N.A.	Commercial	1.2	[77]
PES	o-xylene	0.2 µm	Commercial, BASF	0.1–4	[78]

Table 3. Cont.

Material	Additive	Pore Size	Preparation Method	CO ₂ Flux × 10 ⁻⁴ mol/(m ² ·s)	Ref.
PVDF	LiCl + phosphoric acid	17–53 nm	Dry-jet wet spinning phase inversion	1.31	[79]
PSF	PVP	117–129 nm	Non-solvent phase inversion method	2.5–5	[80]
PSF	Ethanol	19–24 nm	Commercial	1–4	[81]
PVDF	Methanol	0.35–0.48 μm	Commercial Arkema	N.A.	[82]
PVDF	LiCl	N.A.	Phase inversion	1.6	[83]
PVDF, PSF	Glycerol	6.1–9.6 nm	Wet spinning method	N.A.	[84]
PEI	SMM	20–640 nm	Commercial	3.5	[85]
PVDF	SMM	158–650 nm	Dry–wet phase inversion process	5.4	[86,87]
PSF	SMM	250–268 nm	Dry–wet phase inversion process	2.5	[88]
PVDF	DDS, MTS	N.A.	Alkali treatment	N.A.	[89]
PEI	SMM	77–280 nm	Dry–wet phase inversion process	3.2	[90,91]
PVDF	SMM	90–300	Dry–wet phase inversion process	6.8	[92]

3.1. Role of Additives

One of the attractive methods to enhance the wettability resistance of membranes, as well as their properties, is using salt additives in the casting solutions. Generally, additives are water soluble, such as water-soluble polymers, e.g., polyvinyl pyrrolidone (PVP), polyethylene glycol (PEG), inorganic salts such as LiCl, or non-solvents such as glycerol, ethanol, acetone, methanol, phosphoric acid, and acetic acid.

To modify the membrane morphology and structure, several researchers have used additives in the casting solution. For instance, Naim et al. [59] compared the performance of modified PEI and PVDF membranes in terms of morphology in aqueous DEA solution for the CO₂ stripping process at 353 K. They added non-solvent additive PEG into the polymer dope. A higher contact angle, effective surface porosity, and gas permeation were observed for PVDF compared to PEI. Moreover, it was reported that the addition of PEG to PVDF and PEI has a significant effect on stripping flux. Based on their results, at the liquid velocity of 0.7 m/s, stripping flux increased up to 90% and 110% with the addition of PEG to PVDF and PEI, respectively, which can be explained due to the enhancement of effective surface porosity that allowed more surface contact for gas and liquid. In another work, the same authors [60] assessed the effect of the addition of various non-solvent additives on the viscosity, pore size, and surface porosity of PVDF membrane as depicted in Figure 5. Phosphoric acid (PA), methanol, PEG-400, glycerol, and lithium chloride were selected as additives. It is observed that the effect of additives on pore size and surface porosity is significant. For instance, the surface porosity of PVDF increases from 2 to 1690 (1/m) with the addition of PEG400. PVDF-LiCl and PVDF-PA presented the highest viscosity, while PVDF-PEG400 indicated the lowest. In summary, the pore size of evaluated membranes follows the order: PVDF > PVDF-Glycerol > PVDF-methanol > PVDF-PA > PVDF-PEG400 > PVDF-LiCl. Furthermore, their experimental results revealed that the PVDF-PEG400 membrane has the highest stripping flux among other additives. Figure 6 shows the cross-sectional morphology of the inner and outer structure of the non-additive PVDF as well as PVDF membranes with additives.

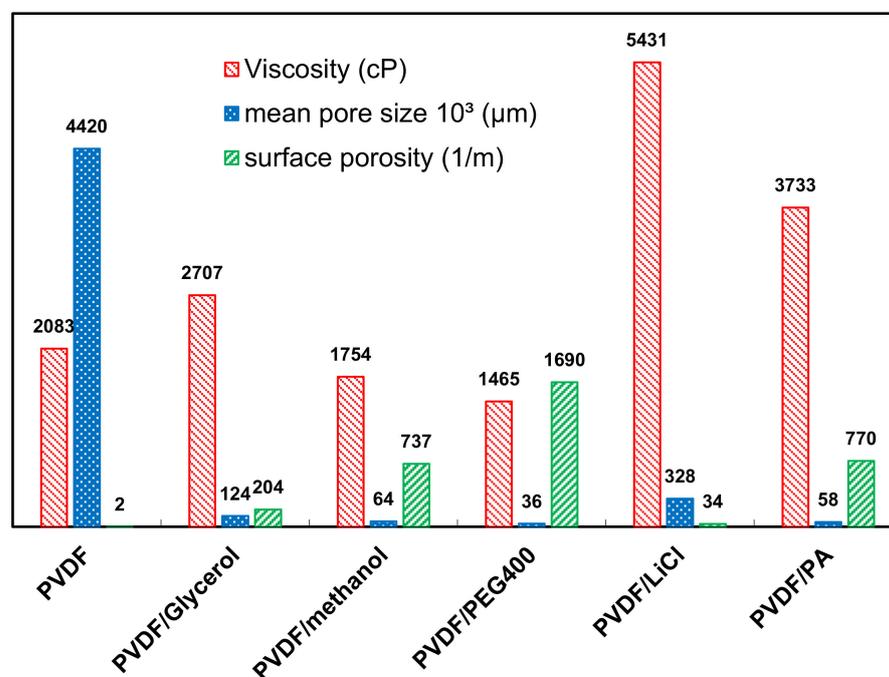


Figure 5. Comparison of PVDF membrane with modified PVDF membrane by various additives in terms of viscosity, pore size, and surface porosity. Reprinted with permission from Ref. [60]. 2012, Elsevier.

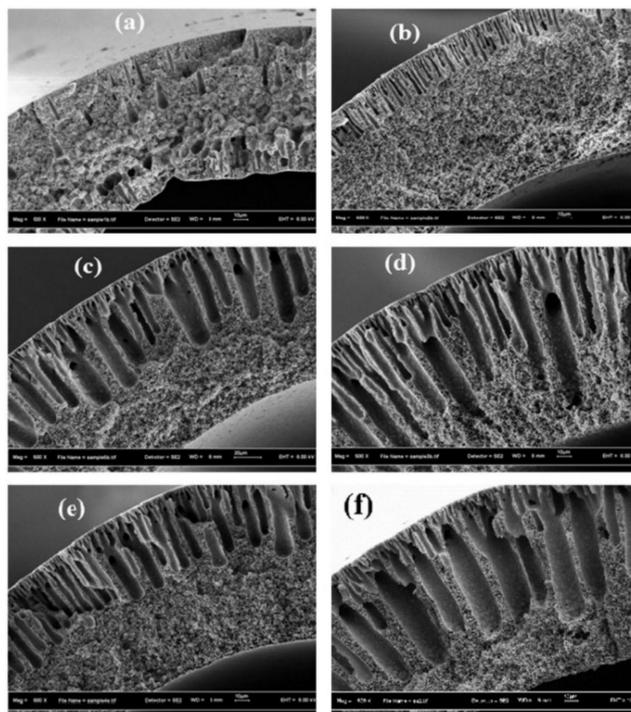


Figure 6. FESEM micrograph of PVDF membrane with various non-solvent additives: (a) LiCl, (b) phosphoric acid, (c) PEG-400, (d) methanol, (e) glycerol, (f) plain. Reprinted with permission from Ref. [60]. 2012, Elsevier.

The formation of small drop-shaped cavities was observed near the inner surface of PVDF-LiCl due to its high viscosity. Moreover, the longer finger-like structure was formed for polymer dope solutions with low viscosity such as PVDF-PEG400, PVDF-methanol, and PVDF-glycerol due to rapid liquid–liquid demixing.

Rahbari-Sisakht et al. [62] selected ethanol, glycerol, and acetone as additives in the spinning dope during the fabrication of PEI hollow fiber membranes. Based on gas permeation results, the largest and smallest pore size was obtained with additives of ethanol and glycerol, respectively. Additionally, PEI-glycerol indicated the highest CO₂ absorption flux when compared to other additives. In another work, Rahbari-Sisakht et al. [63,64] fabricated PSF hollow fiber membranes via a wet-spinning process, whereas 4 wt% glycerol was used as the additive. As a result, a thick sponge-like layer and a thin finger-like layer were obtained in the membrane structure, which resulted in a faster CO₂ absorption rate. Mansourizadeh et al. [66] prepared porous hydrophobic PVDF membranes with a developed structure for CO₂ absorption. They have found that the addition of 4 wt% LiCl resulted in a very small mean pore size (3.9 nm) with high surface porosity. PEG-400, ethanol, phosphoric acid, and glycerol were suggested by Mansourizadeh et al. [67] as non-solvent additives in the polymer dope for PVDF membrane fabrication. According to their observation, the membranes developed by using phosphoric acid, glycerol, and PEG-400 exhibited spongelike structures, which provided high wetting resistance.

Mansourizadeh et al., in another work [70], conducted CO₂ absorption experiments using modified PEI and PVDF membranes. An amount of 6 wt% glycerol was used as an additive. As a result, developed PEI and PVDF membranes exhibited much higher CO₂ flux than pure membranes due to improved structure with good hydrophobicity and low resistance. Furthermore, the developed PVDF membrane showed 35% higher CO₂ flux than PEI. It was also found that the compatibility of glycerol with the PVDF solution is better than the PEI solution.

An effort was made by DashtArzhandi et al. [71] to improve the hydrophobicity, wetting resistance, and structure of PEI hollow fiber membrane by incorporating hydrophobic clay particles into the polymer matrix. They investigated the effects of incorporating hy-

drophobic MMT clay particles as an additive into the polymer matrix in different loadings on the membrane properties. It was shown that modified PEI membranes possess better mechanical stability, smaller pore size, and higher porosity than PEI membranes without MMT. Bakeri et al. [34] successfully applied five compounds including acetic acid, glycerol, ethanol, methanol, and water in their study for structure modification of the PEI membrane. Their characterization results on the structure of PEI revealed that the PEI modified with water shows a sublayer with a sponge-like structure, while the addition of other additives to PEI exhibits a sublayer with finger-like macrovoids due to their lower viscosity. According to their viscosity results, the values of viscosity of PEI with water, methanol, ethanol, glycerol, and acetic acid are 2095, 826, 1014, 1341, and 1049 cP, respectively. To further improve the performance of the PVDF membrane, Pang et al. [79] proposed the idea of using a mixture of two additives including LiCl and phosphoric acid with different concentrations. According to their results, the highest CO₂ flux was achieved by the addition of 8 wt% phosphoric acid, while the PVDF membrane without additives offered the lowest CO₂ flux. This positive effect of the additive on the CO₂ flux can be explained by the fact that the wetting resistance of the membrane decreases with the increase in non-solvent additive in the polymer dope. Although the addition of a mixture of phosphoric acid + LiCl to PVDF showed a positive effect on CO₂ flux, its influence was found to be lower than single phosphoric acid.

In flat sheet membranes, the incorporation of higher loading of ZIF-8 is possible and beneficial for enhanced gas separation performance. In this regard, the flat sheet mixed matrix membrane with 0.5 wt% loading of ZIF-8 was used by Nordin et al. [93], and a 19% enhancement in CO₂/CH₄ selectivity was observed compared to the neat PSF membrane. Nabian et al. [80] employed a PSF flat membrane contactor with polyvinyl pyrrolidone (PVP) as an additive in CO₂ absorption. It was reported that the porosity and hydrophobicity of the PSF membrane increased with the addition of PVP while a reduction in mass transfer resistance was observed. As a result, the CO₂ absorption flux of the PSF + PVP flat membrane was shown to be 169% greater than the flat PSF membrane without PVP.

3.2. Surface Modifying Macromolecules

High hydrophobicity is one of the fundamental properties of membranes utilized in membrane contactors to ensure that the membrane pores remain dry for effective gas transport. Various surface modification methods have been used to enhance surface hydrophobicity, such as UV-assisted grafting, low-temperature plasma treatment, redox-initiated graft polymerization, ion beam irradiation, and photochemical grafting. Membrane surface modification using surface modifying macromolecules (SMM) in the casting or spinning dope has been applied by many researchers as a promising choice to increase the hydrophobicity of membranes. For example, Rahbari-Sisakht et al. [86,87] studied the effects of the SMM concentration on the morphology of SMM-blended PVDF hollow fibers and the performance of the membranes fabricated in membrane contactor applications for CO₂ capture. Surface modifying macromolecules with 0, 2, 4, and 6 wt% were tested as additives in the spinning dope. The surface-modified PVDF membranes showed a larger pore size, higher gas permeance, effective surface porosity, contact angle, and overall porosity but lower critical water entry pressure compared to the PVDF hollow fiber membrane without SMM. Furthermore, by increasing SMM concentration in the spinning dope, the CO₂ absorption flux increased significantly.

In another work, Rahbari-Sisakht et al. [62] fabricated a novel surface-modified polysulfone hollow fiber membrane via a dry-wet phase inversion process using surface-modifying macromolecules. Their results showed that surface-modified membranes offer higher performance compared to plain polysulfone membranes. Moreover, they found that with the membrane prepared from SMM in the spinning dope, a maximum CO₂ flux was achieved, which was almost 76% more than the membrane without SMM. Bakeri et al. [90,91] used surface-modifying macromolecules to enhance the surface hydrophobicity of the PEI mem-

brane. They compared the performance of the SMM-modified membrane with PEI hollow fiber without SMM modification. According to their results, the surface-modified membrane exhibited a large pore size and higher effective surface porosity compared to polyetherimide without SMM. Additionally, the surface-modified PEI membrane indicated a higher contact angle than the PEI membrane without SMM due to the higher surface hydrophobicity of the former membrane. It was also concluded that the surface-modified PEI membrane has a higher CO₂ absorption flux than the PEI membrane without SMM due to the larger pore size and higher surface porosity of the former membrane. Mansourizadeh et al. [92] used surface-modifying macromolecules to improve the hydrophobic PVDF membrane properties for CO₂ absorption in gas–liquid membrane contactors. They investigated the effect of SMM concentration in the polymer dope, air gap distance, and bore fluid composition on the structure and performance of the membranes. The authors observed that by increasing SMM in the polymer dope, the membranes present smaller mean pore sizes, higher permeability, and surface hydrophobicity.

3.3. Composite Membranes

A major issue in gas–liquid membrane contactors is polymeric membrane wetting by liquid absorbent which will result in significant resistance to mass transfer as shown in Figure 7 [94]. Furthermore, polymeric membranes are limited to short-term and mild operating conditions such as low alkalinity, acidity, and low-temperature applications [95]. Therefore, the chemical and mechanical stabilities as well as non-wetting characteristics of polymeric membranes need to be improved. In this regard, many researchers proposed selecting composite membranes with dense skin layers as they combine the advantages of both polymeric and inorganic membranes [96]. Several criteria must be fulfilled for an ideal dense skin layer, including great chemical and thermal stabilities, high CO₂ permeability, and negligible mass transfer resistance [97].

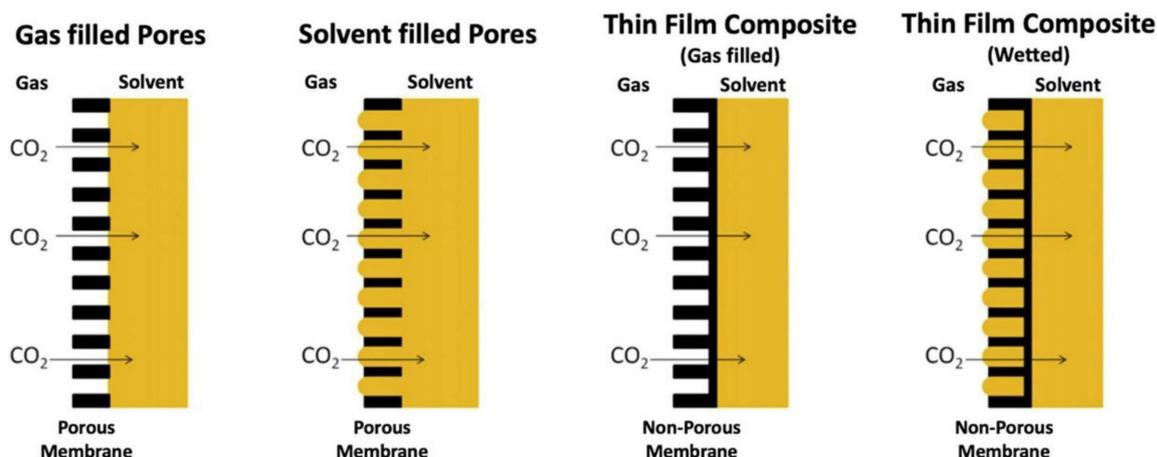


Figure 7. CO₂ absorption through a gas-filled porous membrane contactor and a thin film composite membrane contactor with both dry and wetted porous support. Reprinted with permission from Ref. [94]. 2015, Elsevier.

Dibrov et al. [98] developed thin film composite PTMSP membranes for desorption by a novel two-layer coating technique. The authors revealed that composite membranes with a thin PTFE layer deposited on a cermet substrate ensure a stable CO₂ flux for 100 h during regeneration from a 50 wt% MDEA solution at 100 °C and 30 bar. A comparison between the performance of porous PTFE membrane and nonporous Teflon-PP composite membrane was conducted by Dai et al. [99]. According to their investigation, the Teflon-PP composite membrane offers better stability while the PTFE membrane exhibited higher CO₂ flux. However, both membranes showed good thermal stability at 100 °C for 6 h. Lin et al. [100] employed a high-temperature steam-induced hydrolysis technique to prepare a

fluorinated OH/SiO₂-TiO₂/PES composite hollow fiber membrane. It was found that SiO₂ and TiO₂ can significantly affect the surface hydrophobicity of composite membranes. The contact angle and hydrophobicity of the composite membrane increased significantly compared with the original PES membrane. Amirabedi et al. [96] used techniques of blending the inorganic particles with the dope solution before membrane fabricating and coating the inorganic particles on the surface of the prepared membrane for the fabrication of a PP composite hollow fiber membrane. The resulting composite membranes indicated a higher contact angle (145–168°) than the PP membrane (124°). This trend can be attributed to the incorporation of methyl functional groups with lower surface free energy and an increase in surface roughness. The mechanical stability results showed that the incorporation of the inorganic particles on the surface and in the cross-section of the PP composite membrane improved stress compared to the PP membrane. Furthermore, an effective improvement in the non-wetting property of the membranes was observed. Wu et al. [101] tested a superhydrophobic PVDF composite flat-sheet membrane which was prepared via a non-solvent-induced phase inversion method. Based on their results, PVDF composite membrane exhibited strong anti-wettability to MEA solution and long-term stability even after 20 days. Xu et al. [102] prepared composite dense hollow fiber membranes by dispersing aminosilane-modified ZIF_8 nanocrystals into a PDMS matrix deposited on a porous PVDF substrate as shown in Figure 8. Their fabricated composite membrane presented a significant hydrophobicity with a high contact angle of 130°. Furthermore, CO₂ absorption flux in MEA solution using fabricated composite membrane was obtained equal to 3.8×10^{-3} (mol/(m²·s)) which is higher than other dense membranes such as PVDF (0.2×10^{-3} mol/(m²·s)), PEI (1.3×10^{-3} mol/m²·s), and PTFE (1.1×10^{-3} mol/(m²·s)). CO₂ absorption flux indicated an approximate 80% reduction in PVDF while composite PVDF exhibited only showed an 11% drop after 10 days of testing, indicating its good stability. Similarly, Fosi-Kofal et al. [103] evaluated a PVDF composite hollow fiber membrane prepared by embedding CaCO₃ nanoparticles in the polymer matrix. They detected better wettability resistance and higher surface roughness with more abundant and narrower finger-like pores, permeation rate, and porosity for composite membranes compared to the neat membrane. The key messages of the membrane modification section can be summarized as follows:

- Permeability, wetting resistance, and pore size are important factors, which can seriously affect the performance of the membranes.
- By controlling the phase inversion rate of the spinning dope using the appropriate amount of non-solvent additives, an improved membrane structure can be achieved.
- Membranes that are composed of microporous support and an ultrathin dense layer could prevent membrane wetting, due to the dense thin film on the liquid side of the membrane, which prevents the liquid breakthrough.

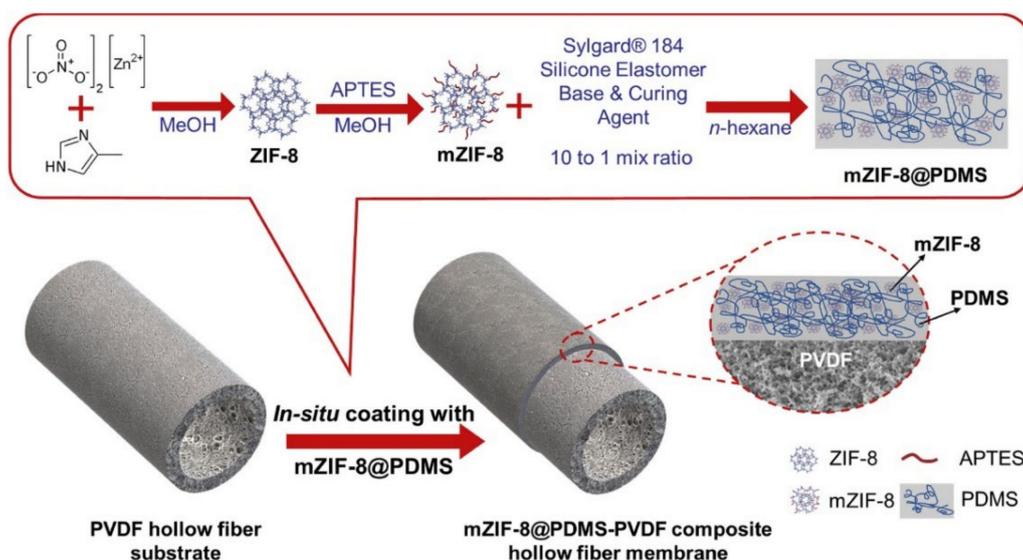


Figure 8. Schematic diagram of the construction of the mZIF_8-based composite dense HF membrane. Reprinted with permission from Ref. [102]. 2019, Elsevier.

4. Module Shape

The gas–liquid membrane contactors have been tested for CO_2 absorption in different module shapes and configurations. Depending on the mode of operation, contactors can be classified into two modes including parallel and cross-flow, as shown in Figure 9. Compared to parallel flow, cross-flow enjoys various advantages including lower pressure drop on the shell side, higher mass transfer coefficients, well-defined flow conditions on both phase fluid sides, and less mass transfer resistance due to the remixing effect [104]. Yang et al. [105] proved that cross-flow membrane contactors offer better absorption performance than parallel flow. Constantinou et al. [106] studied CO_2 absorption in a single-channel PTFE membrane contactor, a nickel mesh contactor, and an eight-channel PTFE membrane reactor. The PTFE membrane used in the experiments consisted of 20 μm thick pure PTFE supported on an 80 μm thick PP layer. It was found that higher CO_2 removal can be obtained when the PP supporting layer was on the gas side rather than on the liquid side, due to less mass transfer resistance of the gas phase. Furthermore, the eight-channel PTFE contactor with 71% removal efficiency showed higher performance compared to the nickel mesh contactor with about 30% efficiency. DeMontigny et al. [28] revealed that the mass transfer rate in the counter-current mode is 20% higher compared to the co-current mode.

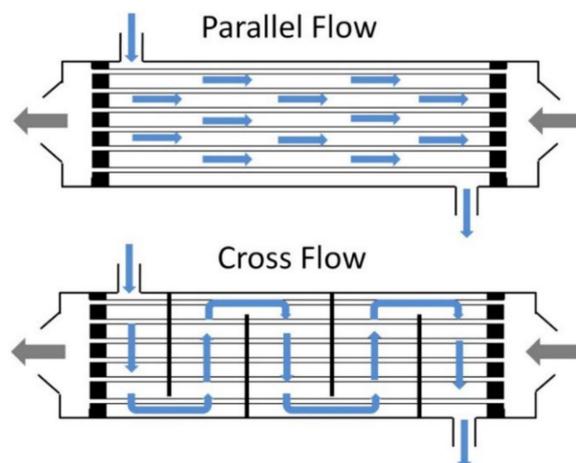


Figure 9. Parallel flow and cross-flow arrangements in hollow fiber membrane modules. Reprinted with permission from Ref. [104]. 2018, Elsevier.

From a geometry point of view, gas–liquid membrane contactors can be classified into two classes: flat sheet and cylindrical membrane modules. A cylindrical membrane module is divided into three types of hollow fibers, capillaries, and tubes [8]. Large gas–liquid contact areas, high surface-to-volume ratio, self-supporting characteristics, and ease of scale-up are the main advantages of hollow fiber membranes compared to flat-sheet membranes [97]. An ideal membrane module design should enable facile process integration and operation, reduce hydrodynamic pressure loss, maximizes effective membrane area, minimizes concentration polarization, and have a cheap price [107].

Although most of the current studies focus on CO₂ capture by hollow fiber membrane contactors, a number of researchers studied CO₂ absorption and desorption using flat-sheet membrane contactors [108–113]. The main advantage of flat-sheet membranes is the ease of membrane fabrication and module assembly. Constantinou et al. [114] studied the feasibility of a flat membrane microstructured contactor (see Figure 10) for CO₂ absorption, and 76% CO₂ removal efficiency was reported by using the eight-channel PTFE contactor.

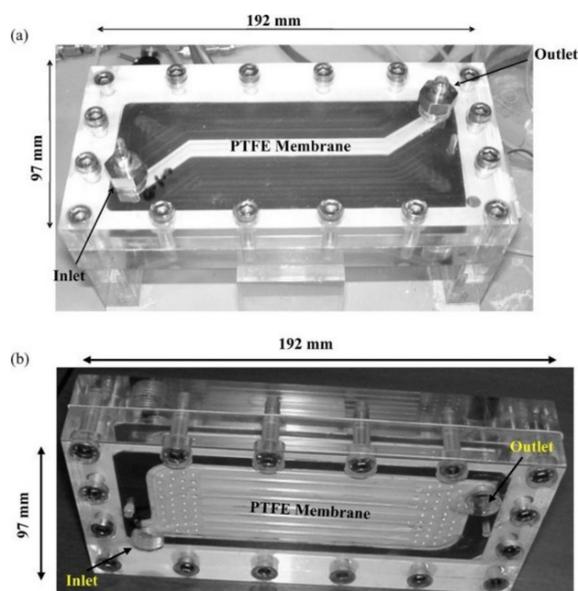


Figure 10. Membrane microstructured contactors, with (a) PTFE single channel contactor and (b) eight-channel PTFE contactor. Reprinted with permission from Ref. [114]. 2014, American Chemistry Society.

5. Absorbents for Membrane Contactors

The selection of an absorbent is a key factor in hollow fiber membrane contactors, as the absorbent's properties directly affect the performance of the process from the capture efficiency and economic aspect point of view [115]. Furthermore, the wetting phenomena in membranes depend on the interactions between the solvent and the membrane [116]. To make the hollow fiber membrane approach more competitive on an industrial scale, liquid solvents as one of the main parts of this technology should ideally possess the following criteria: (a) high CO₂ absorption capacity leading to a smaller plant footprint and less solvent flow; (b) easily regenerable, resulting in reduced energy consumption for regeneration, thereby reducing process cost; (c) fast reactivity with CO₂ to increase absorption efficiency and reduce liquid mass transfer resistance; (d) good physical and chemical compatibility with membrane materials to improve the long-term stability of the membrane module; (e) non-toxic, which is favorable to environmental aspects; (f) low viscosity to minimize the mass-transfer resistance; (g) high surface tension results in a lower potential to wet membrane pores [117]. Among these criteria, viscosity along with surface tension has the greatest effect on the pore wettability in a hollow fiber membrane contactor [20]. To date, chemical absorbents are the most widely utilized absorbents in CO₂ separation processes. The molecular structures of some common solvents for CO₂ absorption are given in Table 4.

Table 4. Molecular structure of different chemical absorbents used for CO₂ capture.

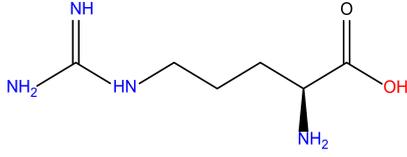
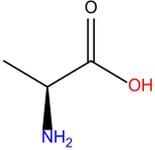
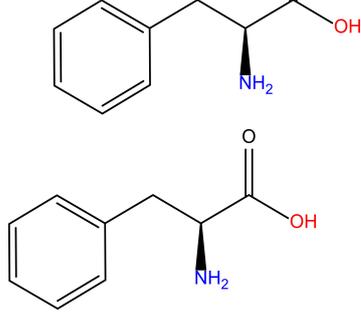
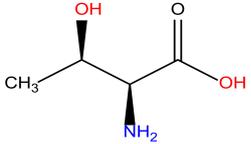
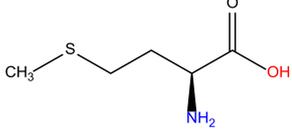
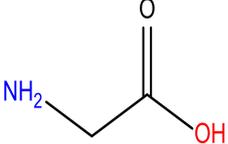
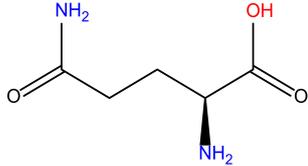
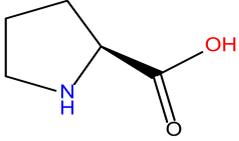
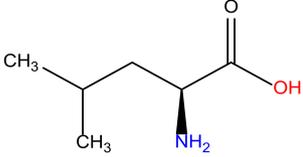
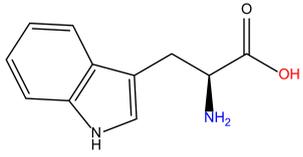
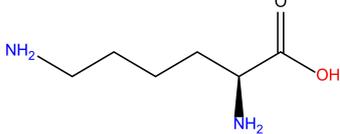
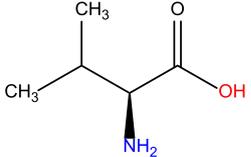
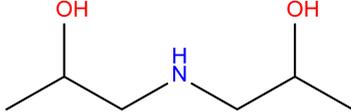
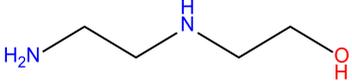
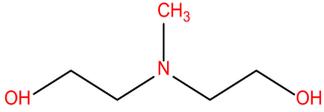
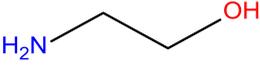
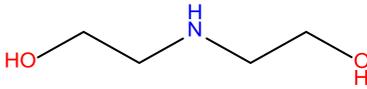
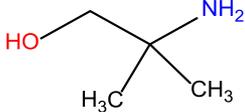
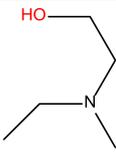
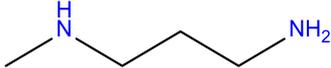
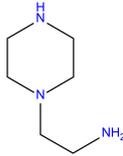
Absorbent	Molecular Structure	Absorbent	Molecular Structure
Arginine (Arg)		Alanine (Ala)	
Phenylalanine (Phe)		Threonine (Thr)	
Methionine (Met)		Glycine (Gly)	
Glutamine (Glu)		Proline (Pro)	
Leucine (Leu)		Tryptophan (Try)	
Lysine (Lys)		Valine (Val)	
Diisopropanolamine (DIPA)		(2-Aminoethyl)ethanolamine (AEEA)	
Methyldiethanolamine (MDEA)		Monoethanolamine (MEA)	
Diethanolamine (DEA)		Piperazine (PZ)	

Table 4. Cont.

Absorbent	Molecular Structure	Absorbent	Molecular Structure
2-amino-2-methyl-1-propanol (AMP)		Diethylethanolamine (DEEA)	
Methylamino)propylamine (MAPA)		2-(1-piperaziny)-ethylamine (PZEA)	

For the initial screening of membrane-solvent combinations, solvents that are used in traditional absorption columns and contactors are in favor since they have proven performance for CO₂ absorption processes, and therefore thermodynamic data are readily available for the absorption phenomena. Various absorbents such as cyclic amines, primary, secondary, and tertiary amines, amino acids, ionic liquids, and inorganic solvents were presented in the literature for gas absorption in hollow fiber membrane contactors. A summary of recent publications using different types of solvent for CO₂ separation is provided in Table 5.

Table 5. Summary of various absorbents applied in hollow fiber membrane contactors.

Solvent Category	Liquid Absorbent	Concentration	Gas Mixture	CO ₂ Flux ×10 ⁻⁴ mol/(m ² ·s)	Membrane Material	Ref.
Amino acid salt	Proline, alanine, sarcosine, glycine	1 kmol/m ³	CO ₂ /N ₂	N.A.	PP	[118]
Amino acid salt	Arginine, serine, threonine, alanine	0.5–1 kmol/m ³	CO ₂ /N ₂	0.8–2	PVDF	[119]
Amino acid salt	Glycine + MEA	1 kmol/m ³	CO ₂	N.A.	PP	[120]
Amino acid salt	Arginine, glycine	1 kmol/m ³	CO ₂ /CH ₄	5–8	PVDF	[121]
Amino acid salt	Lysine	1 kmol/m ³	CO ₂ /CH ₄	N.A.	PP	[122]
Amino acid salt	Sarcosine	1 kmol/m ³	CO ₂ /N ₂	2–2.5	PP	[123]
Amino acid salt	Glycine	1–3 kmol/m ³	CO ₂ /N ₂ /O ₂	5.5–6.1	PP	[124]
Amino acid salt	Glycine + PZ	1 kmol/m ³	CO ₂ /N ₂	N.A.	PP	[125]
Amino acid salt	Sarcosine	1.5 kmol/m ³	CO ₂ /CH ₄	N.A.	PP	[126]
Amino acid salt	Sarcosine, glycine	0.5 kmol/m ³	CO ₂ /CH ₄	17–19	PVDF	[127]
Amino acid salt	Glycine	1 kmol/m ³	CO ₂	1–3.5	PP	[128]
Alkanolamines	MDEA + MEA, DEA + AMP	30 wt%	CO ₂ /N ₂	N.A.	PP	[129]
Alkanolamines	MEA, DEA, MDEA, AMP	10 wt%	CO ₂	N.A.	PP	[130]
Alkanolamines	MEA	30 wt%	CO ₂	5	PP	[131]
Alkanolamines	MDEA + PZEA	1 kmol/m ³		5–7.2	PP	[132]
Alkanolamines	EDA, PZEA	1 kmol/m ³	CO ₂ /CH ₄	N.A.	PP	[133]
Alkanolamines	DEAB	2 kmol/m ³	CO ₂	0.3–0.4	PTFE	[134]
Alkanolamines	MDEA + PZ	N.A.	CO ₂ /N ₂	N.A.	PP	[135]
Alkanolamines	MDEA + PZ	2.5 kmol/m ³	CO ₂ /N ₂	N.A.	PP	[136]
Alkanolamines	DMEA	2 kmol/m ³	CO ₂ /N ₂	0.1–0.15	PTFE	[137]
Alkanolamines	AMP + PZ	1.1 kmol/m ³	CO ₂ /N ₂	2.5–3.08	PVDF	[138]
Alkanolamines	MEA, AMP, DEA	1 kmol/m ³	CO ₂ /N ₂	1–4	PVDF	[139]
Alkanolamines	MEA	1 kmol/m ³	SO ₂ /CO ₂	0.45	PP	[140]
Alkanolamines	MEA	5 kmol/m ³	CO ₂	N.A.	PTFE	[141]
Alkanolamines	MEA	30 wt%	CO ₂ /N ₂	N.A.	PTFE	[142]
Alkanolamines	MEA + DMEA	2 kmol/m ³	CO ₂ /N ₂	0.01–0.03	PTFE	[143]
Alkanolamines	DEEA + PZ	2 kmol/m ³	CO ₂ /N ₂	0.016–0.03	PTFE	[144]
Alkanolamines	1DMA2P	2 kmol/m ³	CO ₂ /air	0.011	PTFE	[145]
Ammonia	NH ₃	5 wt%	CO ₂ /N ₂	N.A.	PP	[146]
Inorganic solvent	K ₂ CO ₃ + proline	5 wt%	CO ₂ /N ₂	3.5–5.2	PP	[147]
Inorganic solvent	K ₂ CO ₃ + 2MPZ	0.5 kmol/m ³	CO ₂ /CH ₄	N.A.	PP	[148]

Table 5. Cont.

Solvent Category	Liquid Absorbent	Concentration	Gas Mixture	CO ₂ Flux ×10 ⁻⁴ mol/(m ² ·s)	Membrane Material	Ref.
Inorganic solvent	K ₂ CO ₃ + SarK	0.12 kmol/m ³	CO ₂ /H ₂ S	2.4	PVDF	[149]
Inorganic solvent	K ₂ CO ₃ + PZ	5 wt%	CO ₂ /N ₂	N.A.	PP	[150]
Inorganic solvent	K ₂ CO ₃ + MDEA	3 kmol/m ³	CO ₂ /N ₂	0.005–0.01	pp	[151]
Inorganic solvent	K ₂ CO ₃	1 kmol/m ³	CO ₂ /N ₂	8–10	PP	[152]
Inorganic solvent	K ₂ CO ₃ + DEA	N.A.	CO ₂ /CH ₄	N.A.	PP	[153]
Inorganic solvent	K ₂ CO ₃ + Gly-K, Lys-K, Arg-K	1.3 kmol/m ³	CO ₂ /CH ₄	9–19	PP	[154]
Ionic liquid	[emim][EtSO ₄]	N.A.	CO ₂ /N ₂	0.3	PP	[155]
Ionic liquid	[Bmim][BF ₄]	10–50 wt%	35% CO ₂	1.5–10	pp	[156]
Ionic liquid	[Bmim][BF ₄] + MEA	40 wt%	CO ₂ /SO ₂ /N ₂ /O ₂	0.4	PP	[157]
Ionic liquid	[Bmim][BF ₄] + MDEA	25 wt%	CO ₂ /N ₂	4.5–7	PP	[158]
Ionic liquid	[Emim][BF ₄]	0.5 kmol/m ³	Pure CO ₂	N.A.	PTMSP	[159]
Ionic liquid	[Emim][BF ₄], [apmim][BF ₄]	1–5 kmol/m ³	CO ₂ /N ₂	0.1–8	PP	[160]
Ionic liquid	[Emim][Ac]	N.A.	CO ₂ /N ₂	N.A.	PVDF	[161]

5.1. Alkanolamines

Well-known amines such as primary amines (MEA and DGA), secondary amines (DEA and DIPA), tertiary amines (MDEA and TEA), diamines (PZ and AEEA), and sterically hindered amines (AMP), have been widely tested for the CO₂ absorption process from flue gas. Each type of amine exhibits its pros and cons in the treatment of acidic gas. For instance, MEA has the advantages of high reactivity with CO₂ and high absorption efficiency at low CO₂ partial pressure, while MDEA takes advantage of low energy consumption in the regeneration process and large absorption capacity [162].

A PTFE membrane was selected by Khaisri et al. [141] to study membrane stability in CO₂ desorption membrane contactors using a CO₂-loaded MEA solution at a temperature of 373 K. They found that a high membrane porosity has a significant effect on solvent desorption performance. It was observed that an increase in the membrane porosity from 23% to 40% resulted in an increase in the CO₂ desorption flux by roughly 260%. Yang et al. [140] evaluated the effect of SO₂ on the CO₂ capture in a PP membrane contactor using MEA solution and they observed an increase in MEA loss per ton of captured CO₂ with an increasing concentration of SO₂ in the absorption–desorption loop system. It was reported that enhancing MEA loss from 1.34 kg to 6.67 kg led to a decrease in the CO₂ mass transfer rate from 0.45×10^{-4} to 0.15×10^{-4} (mol/(m²·s)) and CO₂ removal efficiency from 95% to 30%. This trend can be explained by the promotional effect of SO₂ on the degradation of MEA by the formation of sulfate. Membrane-wetting studies were conducted by Rongwong et al. [139] in a PVDF membrane with three different amines to gain a better understanding of the role of solvents on membrane wetting. According to their obtained data, the CO₂ flux of MEA, AMP, and DEA continuously decreased by roughly 26%, 39%, and 78%, respectively, after 12 days of operation. The results can be explained due to the enhancement in the membrane mass transfer resistance due to membrane wetting. Furthermore, it was pointed out that MEA solution mixed with sodium glycinate resulted in stable CO₂ absorption flux, indicating that the membrane wetting was negligible.

Until now, no single amine has been found to exhibit satisfactory performance in both absorption and desorption. The mixed solvents are an attempt to capitalize on the advantages of each single amine, and thus represent an attractive method for solvent performance improvement [163]. A mixture of MDEA and PZEA solution as a CO₂ absorbent in the membrane contactor was suggested by Zhang et al. [132]. In another work by Zhang et al. [143], CO₂ absorption experiments using a combination of MEA and DMEA solution were carried out in a PTFE membrane contactor with different heights. It was concluded that both desorption rate and absorption capacity can be improved by mixing MEA and DMEA solutions, indicating a synergistic impact in CO₂ absorption into MEA + DMEA. Furthermore, it was claimed that an optimum membrane height should be determined to enhance the efficiency of the process. They concluded that membrane contactor height has a negative effect on CO₂ flux, which could be explained by the fact that the free concentration of absorbent decreases along with the contactor. Nakhjiri et al. [133] investigated the separation process of CO₂ from CO₂/CH₄ gaseous stream using two amines including ethylenediamine (EDA) and 2-(1-piperazinyl)-ethylamine (PZEA) in a PP membrane. Based on their observation, PZEA separated about 92% of CO₂ that existed in the feed gas while the removal efficiency using EDA was about 86%. DEAB as a novel tertiary amine was utilized by Cao et al. [134] for CO₂ absorption in a PTFE membrane contactor system and the results showed a greater CO₂ absorption rate for DEAB compared to AMP and MDEA solutions, which makes DEAB an attractive choice for CO₂ absorption. Additionally, it was reported that the overall mass transfer coefficient and CO₂ absorption performance decrease by 80% and 49% when membrane wetting ratios are 50% and 5%, respectively.

Wang et al. [129] used three blend solutions including MDEA + MEA, MDEA + PZ, and AMP + DEA for CO₂ absorption from CO₂/N₂ mixture in a polypropylene membrane contactor. Paul et al. [130] studied capturing CO₂ using different aqueous single and blended alkanolamine solutions in a hollow-fiber membrane contactor. They used the

aqueous solutions of MEA, DEA, MDEA, and AMP, as well as aqueous blends of MEA + MDEA, MEA + AMP, DEA + MDEA, and DEA + AMP as absorbents. Among the single solutions, the aqueous solution of MEA showed the highest CO₂ absorption flux. Their results revealed that except for DEA + AMP, other blend solutions indicated a higher absorption rate as the concentration of MEA or DEA increased. Gao et al. [144] proposed a blend solution of PZ and DEEA as a new chemical solvent for CO₂ absorption and examined its performance using PTFE hollow fiber membrane contactor at 313 and 353 K. Their experimental outcomes demonstrated that PZ + DEEA solution exhibits a cyclic capacity of 0.8 mol CO₂/L and an absorption rate of about 32×10^{-5} (mol/(L·s)).

5.2. Amino Acids

Although amines have several advantages, they suffer from some drawbacks such as corrosion of process equipment, high solvent regeneration energy requirement, thermal and oxidative degradation, and high vapor pressure [164]. To overcome these limitations, alternative solvents based on amino acid salts have been studied, and so far, these have presented potential benefits over conventional chemical absorbents such as no environmental or toxic issues, less volatility, high absorption capacity, and fast reactivity with CO₂ [165]. Additionally, they are more stable toward oxidative degradation than most amines and have better resistance to thermal degradation [166]. Glycine, arginine, alanine, glutamine, proline, sarcosine, lysine, serine, leucine, valine, threonine, methionine, asparagine, taurine, and histidine are some of the important amino acids used for CO₂ absorption in the literature [167]. One of the distinct properties of amino acid salts is their high surface tension, resulting in fewer pore-wetting problems in hollow fiber membrane contactors [167]. Therefore, the combination of amino acid salts with membrane contactors is expected to be an effective technique to address the issues associated with using alkanolamines.

He et al. [118] assessed the CO₂ absorption performance of several amino acid salts in terms of surface tension and CO₂ absorption flux in a polypropylene membrane contactor. Figure 11 shows a comparison between the surface tension of MEA solution and amino acid salts at different temperatures. It can be seen from this figure that amino acid salts have greater surface tension than MEA solution at all temperatures, which makes them attractive candidates in membrane absorption modules from a lower membrane wetting point of view. Additionally, potassium salts of taurine and glycine showed higher surface tension values than other amino acids leading them to be considered proper choices in membrane contactors. Furthermore, their comparison demonstrated that among studied amino acid salts, potassium arginate exhibited the highest CO₂ absorption flux in the PP membrane contactor, while potassium threoninate presented the lowest absorption flux. Their observation is consistent with the result from Gusnawan et al. [119] that potassium arginate indicated the maximum CO₂ absorption flux compared to potassium salts of threonine, alanine, and serine in a polyvinylidene fluoride membrane contactor.

Lu et al. [120] proposed glycine salt mixed with MEA solution for CO₂ absorption in a PP hollow fiber membrane contactor. The authors concluded that the addition of MEA to amino acid salt of glycine has a positive effect on the overall mass transfer coefficient, and reaches a capture efficiency of more than 90%. The CO₂ removal from a gaseous mixture containing 80% CH₄ and 20% CO₂ in a PVDF hollow fiber was explored by Nakhjiri et al. [121]. The authors selected potassium salts of arginine and glycine as liquid absorbents. It was found that potassium glycinate and potassium arginate can remove CO₂ by up to 62% and 95%, respectively. In another study by Nakhjiri et al. [122], the authors evaluated CO₂ absorption using potassium salt of lysine and they found that potassium lysinate has a removal efficiency of CO₂ of about 91% and can be considered an effective absorbent for natural gas sweetening. Masoumi et al. [123] employed potassium salt of sarcosine for CO₂ capture at high CO₂ partial pressure using a membrane contactor for the first time and observed that potassium sarcosinate has a better absorption flux than DEA and MDEA solutions.

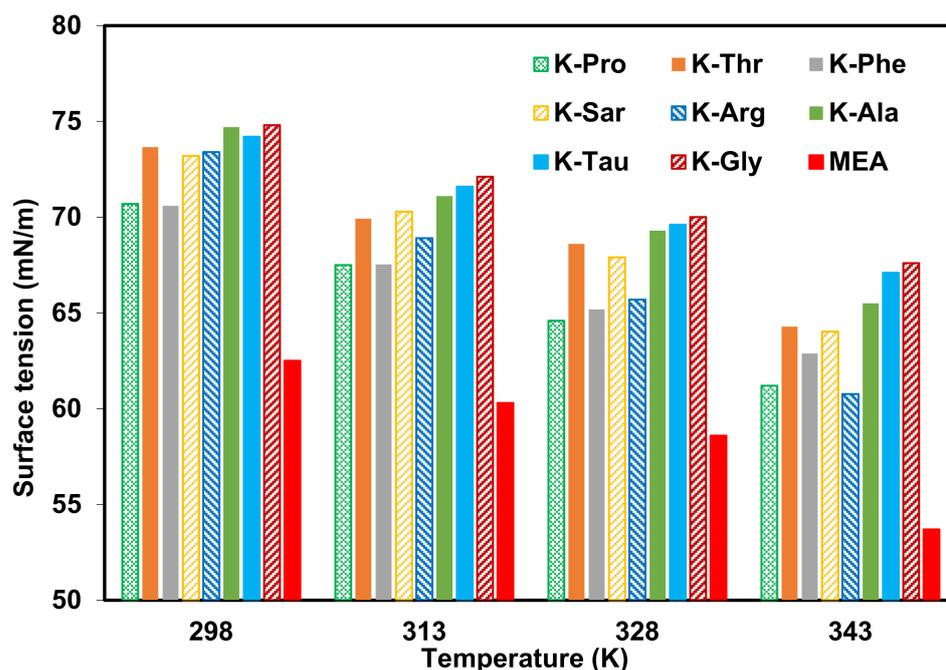


Figure 11. Surface tension of amino acid salt solutions and MEA as a function of temperature. Reprinted with permission from Ref. [118]. 2016, American Chemical Society.

Yan et al. [124] conducted CO₂ absorption experiments from flue gas using three absorbents including MEA, MDEA, and potassium glycinate in pilot-scale hollow fiber membrane contactors. To study the wettability of the PP membrane by aqueous potassium glycinate solution, the authors also measured the surface tension of absorbents. Their investigation results revealed that potassium glycinate has a lower potential for membrane wetting after a continuously steady operation for 40 h due to its higher surface tension than MDEA and MEA. Zheng et al. [125] and Lu et al. [168] prepared a novel CO₂ absorbent by mixing glycine and piperazine and applied it to a PP membrane contactor to absorb CO₂ from a CO₂/N₂ mixture. The authors claimed that the addition of piperazine to glycine increases the overall mass transfer coefficient by up to 30%, and the efficiency can be determined by more than 90%.

The separation of CO₂ from CH₄ using potassium sarcosinate was carried out by Simon et al. [126], and its performance was compared to MEA solution. Based on their results, potassium sarcosinate offers a higher CO₂/CH₄ selectivity (120) compared to MEA with a value of 60 under the same conditions. Likewise, Rahim et al. [127] studied the feasibility of using potassium and sodium salts of glycine and sarcosine in a PVDF hollow fiber gas–liquid membrane contactor. The finding of the study showed that potassium sarcosinate has a better absorption performance compared to potassium glycinate due to its high CO₂ loading capacity, while potassium glycinate exhibited better stripping efficiency as glycine is more acidic than potassium sarcosinate. Their long-term experiments indicated that potassium glycinate is more stable than potassium sarcosinate. Furthermore, it was concluded that potassium salts of amino acids show better desorption performance while higher performance in absorption was determined by sodium salt of amino acids.

5.3. Ionic Liquids

Due to their special characteristics such as negligible vapor pressures, which essentially minimize the opportunity for solvent release into the atmosphere, ionic liquids are considered green solvents [169]. These types of solvents carry the advantage of being easy to be recycled, having a low melting point, non-flammability, remarkable thermal stability, and ease of regeneration [170]. Qazi et al. [155] evaluated the membrane-wetting condition in a PP membrane contactor with ionic liquid 1-ethyl-3-methylimidazolium ethylsulfate

([emim][EtSO₄]). By considering their results, a drop of 40% in CO₂ removal efficiency was observed when the membrane condition changes from non-wetted to 5% membrane wetting, which means membrane wetting has a noteworthy effect on CO₂ separation efficiency due to an enhancement in the mass transfer resistance. Moreover, for both wetted and non-wetted membranes, CO₂ removal efficiency is favored by an increase in membrane porosity. The absorption performance and energy-consuming of [bmim][BF₄] blended with MEA solution in a membrane contactor and a stripping column were studied by Yang et al. [157]. Their experimental outcomes showed that the CO₂ removal efficiency of about 96% can be obtained using 30 wt% MEA + 40 wt% [bmim][BF₄] solution. Moreover, thermal energy at the desorber column was found to be equal to 5.14 and 8.18 GJ per ton of captured CO₂ for [bmim][BF₄] + MEA and MEA solutions, respectively. It was also reported that MEA solution loss for the mixed system (1.16 kg per ton of captured CO₂) is lower than that for the single MEA solution (3.55 kg per ton of captured CO₂). The authors claimed that [bmim][BF₄] + MEA solution requires 487 kJ for mechanical energy in pumping solvent while the MEA system needs about 482 kJ due to its lower viscosity. Similarly, Bazhenov et al. [159] tested a flat-sheet PTMSP membrane contactor for CO₂ desorption using [Emim][BF₄] to examine the compatibility between ionic liquids and membranes. The CO₂ stripping measurements were conducted at a transmembrane pressure of 10 bar and a temperature of 303 K. They concluded that as stripping temperature increases from 303 K to 323 K, CO₂ stripping flux in the membrane contactor increases up to 30%. Furthermore, their long-term studies (40 days) indicated that PTMSP membranes are impermeable to [Emim][BF₄] at a trans-membrane pressure of 40 bar. Their results also showed that the solvent-membrane interaction is determined by the solvent surface tension regardless of molecular size and viscosity of ionic liquids. Two ionic liquids including [bmim][BF₄] and [apmim][BF₄] were utilized by Lu et al. [160] for CO₂ separation in a PP membrane contactor. Their results revealed that [apmim][BF₄] presents greater performance than [bmim][BF₄] in terms of CO₂ absorption capacity and absorption rate. Gomez-Coma et al. [161] implemented the CO₂ capture in a PVDF hollow fiber membrane contactor with [emim][Ac] ionic liquid due to its high CO₂ solubility. According to their observation, mass transfer of the membrane contactor is significantly dependent on the water content in the [emim][Ac] ionic liquid. This effect was explained due to the change in viscosity and the molecular interactions that water promotes in [emim][Ac].

5.4. Deep Eutectic Solvents (DES)

Despite the advantages of ionic liquids discussed in the previous section, their limitations in CO₂ capture processes, such as high viscosity during CO₂ absorption, low CO₂ capacity, and their high price, unfortunately impair the process efficiency in a large-scale application [171]. Furthermore, there are growing concerns regarding the toxicity of some conventional ionic liquids. Therefore, it is critically important to develop novel absorbents, which are more advantageous than ionic liquids. In this regard, deep eutectic solvents as a new class of ionic liquids have attracted lots of attention and were proposed by several researchers due to their ease of synthesis, cost-effectiveness compared to ionic liquids, and eco-friendly nature as well as their lack of by-product generation [172]. However, like other categories of absorbents, low CO₂ uptakes of deep eutectic solvents need to be resolved. This section provides a brief description of works that have been undertaken on using deep eutectic solvents in CO₂ capture units. Deep eutectic solvents are prepared by mixing two components including hydrogen bond acceptors (HBAs) and hydrogen bond donors (HBDs) [173].

Trivedi et al. [174] tested a combination of EDA and [MEA-Cl] as a deep eutectic solvent and reported that their synthesized material exhibited gravimetric CO₂ uptake of around 33.7 wt%, which is higher than all ionic liquids, which have a maximum uptake of 18.5 wt% CO₂. The authors explained that this excellent performance of [MEA-Cl][EDA] can be attributed to different polarities and basicities of the deep eutectic solvents originating from the HBA functional groups. Gu et al. [175] synthesized a series of hydrophobic

functional deep eutectic solvents based on polyamine hydrochloride as an HBA and thymol as an HBD. According to their finding, [TEPA]Cl-thymol could efficiently capture CO₂ even at low partial pressures. The CO₂ capacity of [TEPA]Cl-thymol was high, up to 1.355 mol CO₂/mol DES at 40 °C and 101.3 kPa.

Supported DES membranes have been evaluated in recent years; they combine highly processable and flexible polymeric support impregnated with DES for CO₂ capture [176]. Vatanpour et al. [177] proposed the hydrophilic DES ethaline, which is a mixture of choline chloride and ethylene glycol in a PES/dimethyl acetamide polymer solution. Their results showed that DES can be used as hydrophilic additives to improve membranes' antifouling properties. Craveiro et al. [178] showed that PTFE membranes containing deep eutectic solvents based on choline chloride offer high selectivity for CO₂/CH₄, with values above the Robeson upper bound indicating their good potential for CO₂/CH₄ separations. In another work, Jiang et al. [179] found that the addition of deep eutectic solvents to casting solution in PES membrane fabrication could improve membrane porous structure, which contributed to a significantly increased permeability and a high selectivity.

5.5. Inorganic Solvents

Since the absorption of CO₂ using inorganic solvents is effective at high temperatures and pressure, this process has gained widespread acceptance for CO₂ removal from natural gas [180]. The low environmental impacts, high chemical stability, high CO₂ absorption capacity, negligible volatility, better resistance to the presence of sulfur oxides and nitrogen oxides, and slow degradation are the main benefits of inorganic absorbents [181]. However, this category of absorbents suffers from slow reaction kinetics with CO₂ at prevailing low temperatures resulting in larger equipment to achieve the same removal [182]. The addition of amine promoters such as amines, amino acids, and cyclic amines to potassium carbonate (K₂CO₃) solution was suggested as an efficient technique to enhance its absorption rate. An experimental investigation of the simultaneous separation of H₂S and CO₂ from biogas using K₂CO₃ mixed with potassium sarcosinate was conducted by Jin et al. [149]. It was concluded that the addition of potassium sarcosinate increases both CO₂ and H₂S absorption flux. They also observed that CO₂ absorption flux reduces by approximately 30% after 90 min of operation due to partial wetting of PVDF membrane, while it showed little effect on H₂S absorption flux. Niknam et al. [147] used proline as a promoter for K₂CO₃ solution in a PP membrane contactor and compared its performance with K₂CO₃ promoted with MEA solution. According to their observation, proline and MEA have the same positive effect on CO₂ removals. Furthermore, their results revealed that membrane wetting up to 10% significantly decreases CO₂ recovery in K₂CO₃ + proline solution from 90% to 45%. In another work, Izaddoust et al. [150] added piperazine to a K₂CO₃ solution for CO₂ separation in a PP membrane and discovered that 90% CO₂ recovery can be determined by the addition of the 0.03 M PZ. Additionally, their results revealed that aqueous K₂CO₃ + PZ solution has better performance than single K₂CO₃ in terms of membrane wetting. This can be explained by the fact that at low wetting conditions, a fast reaction rate in the stagnant phase can prevent a sharp reduction in recovery. The potentialities of potassium salts of arginine, lysine, and glycine as promoters in K₂CO₃ solution were explored by Li et al. [154] in a PP membrane contactor. Based on their results, K₂CO₃ promoted by potassium glycinate exhibits a more efficient performance than the other solvents with the highest removal efficiency and CO₂ absorption flux.

Ammonia is another attractive solvent due to its low regeneration energy demands compared to MEA solution, low thermal and oxidative degradation, cheap price, and high CO₂ absorption capacity. Therefore, the feasibility of ammonia-based CO₂ absorption processes using hollow fiber membrane contactors was assessed by several researchers in the literature. For example, the membrane contactor approach was tested for CO₂ capture using 1 to 5 wt% ammonia solution by Makhloufi et al. [146]. The authors performed CO₂ absorption experiments using two different types of hollow fiber membrane contactors including a composite membrane contactor and a microporous membrane contactor.

According to their results, the composite membrane contactor showed no performance loss after more than 6 months of use, while the CO₂ removal efficiency in ammonia using microporous membrane contactor decreases over time due to ammonium salt precipitation within the porous structure as well as on the lumen side of the microporous fibers.

The important conclusions of this section are as follows:

- The choice of proper absorbent plays a critical role in membrane gas absorption as it can strongly influence mass transfer, efficiency, and operating cost.
- High surface tension and good compatibility with the membrane are the most important criteria for selecting an absorbent.
- Proline is the most promising amino acid for bulk absorption of CO₂ with faster reaction kinetics than other amino acids while glycine exhibits the highest surface tension.
- Better absorption and desorption performance can be expected when a mixture of two solvents is used.
- Potassium salt of amino acids has greater absorption properties than sodium salt of amino acids.
- Amino acid salts are a better choice than amines for use in membrane contactors from the surface tension point of view.
- Absorbent properties and their interaction with membrane material are essential factors in determining the extent of wetting.
- The interaction between chemical solvent and membrane pores leads to a change in the internal pore structure, which enhances membrane wetting.

6. Operating Parameters

In addition to the type of membrane and absorption liquid, also the process parameters such as the temperature of absorption and desorption, liquid flow rate, and feed pressure, play an important role and contribute to the overall process performance. The operating conditions must be carefully adjusted to optimize the membrane performance while membrane wetting is avoided. The most important parameters which influence the performance of gas–liquid membrane contactors are listed in Table 6.

Table 6. Operational parameters affecting the performance of a membrane contactor.

Membrane Parameters Affecting Membrane Performance	
Fiber inner radius (μm)	Membrane area (m^2)
Membrane thickness (μm)	Number of fibers
Module inner diameter (m)	Membrane porosity
Membrane length (m)	Membrane tortuosity
Packing density	
Operating Parameters Affecting Membrane Performance	
Gas temperature (K)	Gas flow rate (L/h)
Liquid temperature (K)	Liquid flow rate (L/h)
Operating pressure (MPa)	Liquid concentration

Here, recent publications which studied the influence of operating parameters on the performance of membrane contactors are discussed briefly. Mohammadi et al. [183] showed that when half of the membrane thickness is filled with glycerol, CO₂ removal decreases by about 40%. They also investigated the effect of the membrane porosity-to-tortuosity ratio on CO₂ removal and concluded that CO₂ removal is enhanced from 66% to 71% when the membrane porosity-to-tortuosity ratio increases from 0.1 to 0.9. The porosity has a significant effect on the tortuosity of the membrane and the mass transfer of CO₂. There is a direct relationship between the effective diffusion coefficient of CO₂ inside the membrane side and membrane porosity. It means that enhancing the porosity of the membrane results in a significant increase in the membrane diffusivity and therefore, the mass transfer of CO₂ inside the membrane side increases, which leads to an increase in CO₂ removal efficiency

under non-wetting conditions. It is worth quoting that in comparison to the non-wetting mode of operation, in partial wetting conditions, penetration of liquid into the pores causes a decrease in the effective diffusion coefficient of CO_2 and hence decreases the removal efficiency. In addition, it was observed that increasing the glycerol concentration from 10 wt% to 35 wt% causes a significant enhancement in CO_2 removal while a further increase up to 50 wt% in concentration decreased CO_2 removal due to an increase in the viscosity of the solution, which leads to an increase in the resistance of mass transfer and a reduction in the fluid mobility. Lin et al. [100] tested PES composite hollow fiber membranes for CO_2 capture under different gas and liquid flow rates. According to their observation, higher CO_2 flux and separation efficiency can be achieved as the flow rate of the liquid increases from 2 to 16 L/h. This positive effect can be explained by the fact that more CO_2 is absorbed per unit volume of absorbent at a high flow rate, which is sufficient to ensure that the amount of fresh absorbent is sufficient. In other words, a higher solvent flow rate increases the driving force for CO_2 due to the greater concentration difference. Moreover, the authors indicated that too high a gas flow rate results in a lower separation of CO_2/CH_4 . It was described that a rise in gas flow rate leads to minimal gas residence time in the tube, which allows more CO_2 to pass from the contactor without absorption, resulting in reduced CO_2 removal efficiency. A similar finding has been reported by Mesbah et al. [148].

The effect of fiber inner diameter was studied by Qazi et al. [155], and a significant drop in removal efficiency was determined by increasing the fiber inner diameter. The reason is that as the fiber inner diameter increases, gas volume increases, and thus more CO_2 will be available for the same amount of liquid, which decreases removal efficiency. The authors also assessed the effect of module length and showed that a longer membrane length causes a considerable increase in absorbent residence time in the module and mass transfer surface area, which improves the process efficiency. However, an optimized value for membrane module length should be used for optimum separations as pressure drop can be observed when membrane length increases and also CO_2 concentration drops progressively along membrane length, which has a negative impact on the driving force for separation. It was also found that there is an optimized value for the liquid flow rate. Although a high liquid flow rate has a positive effect on removal efficiency, it also increases the potential for pore wetting. This behavior was attributed to the increase in pressure with an increase in liquid flow rate, thus resulting in higher transmembrane pressure and increasing membrane pore wetting. Hence, it is imperative to maintain the operation in a reasonable liquid velocity range to ensure higher CO_2 capture efficiency. In another work, the impact of the number of fibers on CO_2 removal efficiency inside hollow fiber membrane contactors was evaluated by Nakhjiri et al. [122]. Based on their results, a rise in the total number of fibers from 100 to 600 positively encourages gas–liquid mass transfer interface and as a result improves the gas/liquid contact zone inside the hydrophobic membrane contactor, which leads to enhancement in the CO_2 removal from 40% to 92%.

Chen et al. [184] pointed out that CO_2 absorption flux increases with the rising of liquid temperature from 298 K to 318 K. This is mainly attributed to the fact that with an increase in liquid temperature, both the chemical reaction rate and diffusion coefficients of species in the liquid increase. Furthermore, the authors investigated the effect of CO_2 partial pressure on absorption flux and concluded that the CO_2 absorption flux rises up quickly as the CO_2 partial pressure increases. It was reported that as the amount of CO_2 in the inlet gas increases, more CO_2 molecules will travel through the gas bulk and the porous membrane to contact and react with solvent at the gas–liquid interface. As a result, a higher mass transfer driving force and CO_2 absorption flux are achieved. Cao et al. [185] claimed that the membrane wetting decreases slightly with increasing CO_2 partial pressure. This phenomenon can be explained by the fact that a higher concentration of CO_2 can consume more amounts of reactive absorbent molecules. Therefore, more protonated absorbent and bicarbonate can be generated at the higher CO_2 concentration, resulting in higher surface tension with the increased CO_2 loading. As a result, the membrane wetting could be mitigated at the higher CO_2 concentration in the gas mixture. Dai et al. [99] examined

the influence of operating pressure on CO₂ flux in a PTFE membrane contactor and showed that CO₂ flux greatly increases from 0.5 to 4×10^{-4} mol/(m²·s) when operating pressure increases from 1 bar to 20 bar due to the higher CO₂ driving forces. In addition, it was found that increasing operating pressure is more effective in enhancing CO₂ flux in the porous PTFE membrane contactor compared to the nonporous membrane due to less mass transfer resistance in the porous membrane. The effect of module packing density in PTFE membrane contactors was studied by Ghobadi et al. [186]. The authors pointed out that a rise in packing density from 10 to 50% can improve the CO₂ separation efficiency by 20%, as given in Figure 12. This can be explained by the fact that an increase in the packing density results in a higher membrane surface area over the available module volume and also a more ordered fiber arrangement inside the modules.

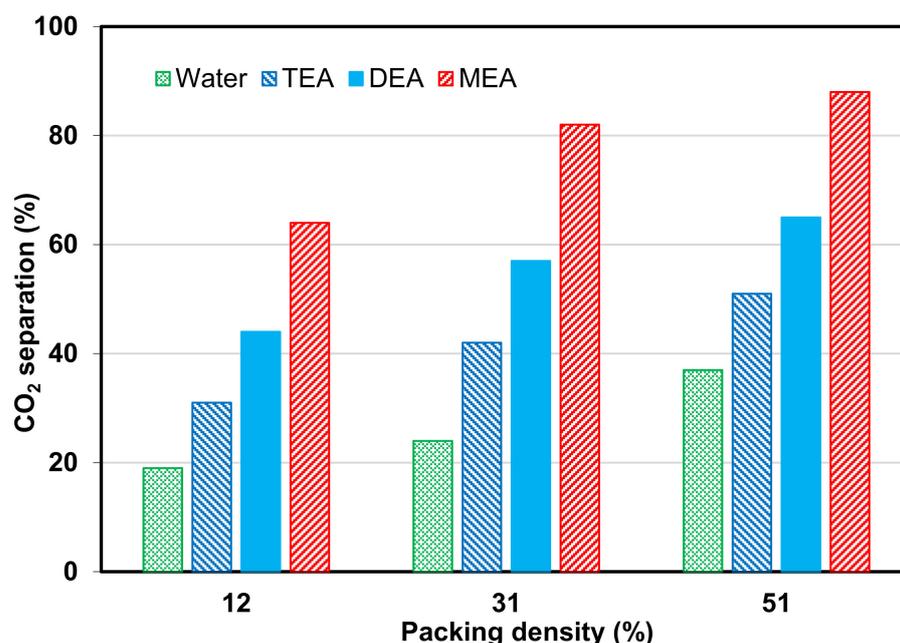


Figure 12. CO₂ separation as a function of module packing density for different absorbents. Reprinted with permission from Ref. [186]. 2017, Elsevier.

The key messages of this section are as follows:

- An increase in operating pressure, liquid temperature, and packing density favor CO₂ flux.
- Membrane wetting becomes more serious with the increase in solvent flow rate.
- Higher gas flow rates can improve CO₂ absorption flux, but at the cost of reducing CO₂ removal efficiency.
- Improving CO₂ absorption performance by increasing the liquid flow rate to a very high level may not be ideal because of the risk of membrane wetting.
- The module with a smaller diameter hollow fiber membrane can accommodate a much greater interfacial area per unit volume, which makes it a more efficient absorber unit.
- Increasing module length, number of fibers, and porosity provide better CO₂ separation performance.

7. Membrane-Based Hybrid Processes

One of the main challenges of existing CO₂ capture approaches is the energy penalty. The energy consumption for regeneration of the MEA-based absorption process varies from 2.5 to 3.5 GJ/ton CO₂ depending on the source of CO₂, which is believed to be responsible for approximately 80% of the total energy consumption [7]. This high energy needs to be decreased to around 1 GJ/ton CO₂ while keeping CO₂ removal efficiency above 90% [187]. In this regard, the integration of two technologies (see Figure 13) was represented in the

literature as an effective way to overcome the issue of the energy penalty [188]. Substitution of the conventional energy-consuming process by an energy-saving membrane process or their combination, which is called the hybrid process, is principally economical. The specific feature of the hybrid process is its synergy outcome from the integration of two-unit operations. The combination of the conventional process with membrane separation technology improves the capture efficiency of the process while the energy consumption is minimized accordingly. The membrane-based hybrid process may be classified into two types including the membrane-cryogenic process and the membrane-chemical absorption process. To select a suitable hybrid process, several factors should be taken into account such as pressure and temperature of the gas stream, CO₂ concentration, the availability of the capture equipment, as well as desired product purity, as the energy consumption is significantly affected by chosen hybrid process type.

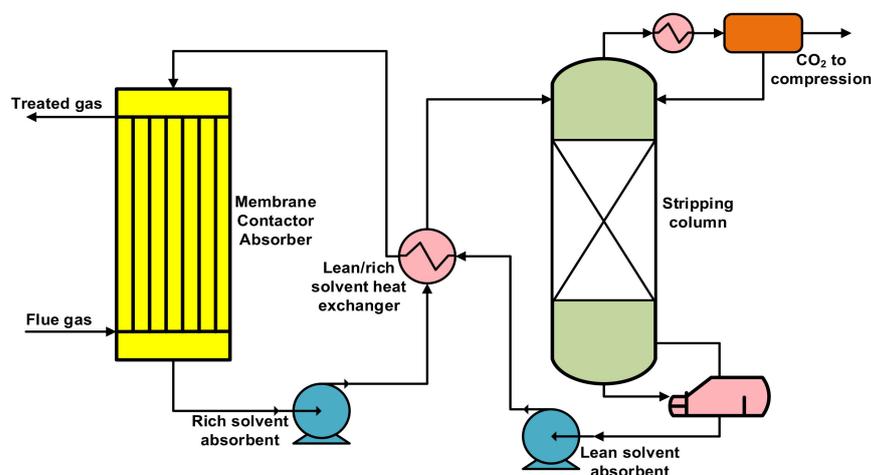


Figure 13. Schematic diagram of the membrane contactor–stripper hybrid process for CO₂ absorption.

Yeon et al. [189] studied the performance of a hybrid process that includes a membrane contactor and packed tower as an absorber and a thermal stripper, respectively, for CO₂ capture from flue gas. They used a porous PVDF hollow fiber module with MEA and TEA as absorbent solutions. Their investigation results showed that the integration of membrane and stripper as a hybrid process leads to a higher capture efficiency than the conventional absorption column. The CO₂ absorption rate per unit volume of the membrane contactor was obtained in their work to be 2.7 times greater than that of the packed column. The authors also measured operating power consumption and concluded that the electricity consumption in the membrane contactor (0.78 GJ/ton CO₂) is lower than the packed column unit (1.16 GJ/ton CO₂). Likewise, Kosaraju et al. [57] assessed asymmetric poly (4-methyl-1-pentene) (PMP) hollow fiber membranes with an ultrathin, dense skin layer for an absorption and desorption loop system, and MEA as an absorption liquid for the separation. Their long-term test results revealed a decrease in CO₂ absorption performance after 55 days. A hybrid membrane-liquefaction system was proposed by Anantharaman et al. [190] for CO₂ absorption. A techno-economic analysis was carried out by the authors and the results indicated that the cost of CO₂ avoided (48 €/ton CO₂) is about 9% more cost-efficient than the conventional MEA-based absorption process. Lee et al. [191] evaluated the performance of a ceramic hollow fiber membrane contactor module integrated with a stripping column for CO₂ capture. It was found that the hybrid process can absorb more than 90% of CO₂ with high stability for more than 60 h. Moreover, their results showed the overall gas mass transfer coefficient in a ceramic hollow fiber membrane contactor could be improved by 1.8 times compared to a conventional packed column. It can therefore be concluded that the ceramic hollow fiber membrane contactor is a promising alternative to the polymeric membrane contactor and packed column systems. Wang et al. [192] examined CO₂ desorption from 20 wt% MEA solution in a hollow fiber membrane contactor combined

with a packed column. According to their results, the performance of the stripper increases as the membrane's length increases and membrane diameter decreases. Jiang et al. [193] compared CO₂ separation in four different configurations including one-stage membrane, two-stage membrane, chemical absorption, and membrane-chemical absorption. Based on their observation, the CO₂ purity in the two-stage membrane design was found to be 46.2% greater than the one-stage membrane. This promotion of CO₂ removal efficiency can be explained that using two PI membrane modules leads to more separation of the feed gas flow and a rise in the CO₂ concentration of the permeate gas resulting in an efficiency of around 99%. Furthermore, the combination of membrane and chemical absorption showed a reduction in regeneration heat and absorbent flow rate.

Burdyny et al. [194] utilized a hybrid cryogenic and membrane process in oxy-fuel combustion. They reported that hybrid configuration enhanced the efficiency by 0.9% as well as reducing the size of the cryogenic equipment and distillation columns, which will benefit the industry substantially. A hybrid process including PP hollow fiber membrane contactor as an absorber and a packed column as a stripper was used by Yan et al. [124] for CO₂ capture from flue gas. Several amine solutions such as MEA, MDEA, and potassium glycinate were tested as the solvent, and their performance in CO₂ capture was compared. It was found that the process has a capture efficiency of more than 90% with a lower potential of membrane wetting when potassium glycinate is employed as an absorbent. In another work, Yan et al. [195] studied the effect of the combination of the membrane with the stripper column on CO₂ absorption. Aqueous MEA solution and PP were used as absorbent and hollow fiber, respectively. It was concluded that in the case of using a fresh membrane, the membrane separation system showed an overall mass transfer coefficient about 2.9 times higher than that of the packed column while having a lower absorption cost. However, this improvement could reduce when all the membrane pores were completely wetted. Belaissaoui et al. [196] explored the potential of membrane separation combined with a cryogenic process for post-combustion CO₂ capture. Based on their results, the hybrid configuration needs lower energy in comparison to conventional CO₂ absorption for a CO₂ concentration of 15–30 mol%. This improvement was in the range of 12 to 25%, which makes the use of this technology an attractive strategy. Kundu et al. [197] conducted a study on the membrane-absorption system for CO₂ removal and investigated its performance from an energy requirement perspective. It was revealed that the heat duty of the reboiler in a hybrid process can be significantly saved due to an increase in the membrane area. The authors also claimed that the membrane technology exhibits less energy demand and can save up to 35–55% energy compared to the MEA system.

The major conclusions are drawn as follows:

- The hybrid process offers a high degree of flexibility, with respect to the capture ratio and/or final CO₂ purity.
- The improvement to either membrane or cryogenic technologies will improve the hybrid system as well.
- Combining membranes and absorption technologies could result in significant energy savings by reducing the steam required for amine regeneration.
- The hybrid process showed a higher CO₂ removal efficiency than the conventional absorption tower.
- The membrane contactor hybrid process was proved to be economic by evaluation through CO₂ recovery cost and operating power consumption.
- The membrane-absorption unit can improve the absorption process by generating 400–1500% greater mass transfer area per unit volume leading to smaller equipment sizes.
- Compared to standalone methods, hybrid processes showed superiority not only in CO₂ recovery and energy penalty but also in installation investment.

8. Long-Term Stability of Membranes

The membranes' pores in most hollow fiber membrane contactors are gradually wetted over time, which will lead to a sharp increase in the membrane mass transfer resistance and a remarkable reduction in CO₂ absorption fluxes. It is well known that any reaction between the absorbent and membrane material could possibly change the membrane matrix and surface morphology. Therefore, to make gas–liquid membrane contactor technology more competitive on an industrial scale, the long-term stability of membranes should be improved. The long-term stable operation of the gas–liquid membrane contactor requires that the pores of the membrane remain completely gas-filled over the long operational time.

Dai et al. [99] conducted a long-term CO₂ absorption test of PTFE membrane in contact with [Bmim][TCM] as an absorbent at 20 bar in a 14-day test. It was found that the CO₂ flux through the PTFE membrane decreased by 50% after 7 days of operation due to the morphological rearrangements and pore wetting by the absorbent. The authors observed that the PTFE membrane surface was partially destroyed by absorbent after two weeks of testing at 20 bar. Jin et al. [149] studied the effect of operation time on CO₂ and H₂S flux in a PVDF membrane contactor and observed that the CO₂ absorption flux decreases significantly after 5 h while the H₂S flux changes over time were negligible. Similar results were found by Rajabzadeh et al. [198]. They showed that the PVDF membrane contactor presents a performance reduction of more than 80% after 60 h of operation. Furthermore, it was concluded that membranes with lower porosity and pore diameter were stable for a longer time, compared to membranes with larger porosity and pore diameter, which were completely wetted during operation.

Xu et al. [102] fabricated a composite membrane with a dense skin layer by dispersing ZIF-8 nanocrystals into a PDMS matrix deposited on porous PVDF and studied its performance throughout 15 days of operation. The authors revealed that the developed membrane remains constant at its high CO₂ flux even after 15 days of the test, indicating a significant improvement in its long-term stability. Zhang et al. [199] evaluated the long-term stability of a PEI hollow fiber membrane over 60 days. According to the results of the long-term stability test, the membrane was able to maintain a relatively stable performance with a CO₂ absorption flux of about 1.4×10^{-2} mol/(m²·s), indicating lower wetting issues. Amirabedi et al. [96] compared the chemical stability of neat PP and composite membranes with MEA solution. It was found that the composite membrane exhibited a more stable non-wetting property during the long-term process compared to a neat membrane due to the coating of nanoparticles on the membrane surface. Furthermore, the neat membrane showed a significant reduction in CO₂ flux after 30 days of testing. This trend can be attributed to the penetration of MEA into the membrane pores. The long-term stability of PP, PMP, and PTFE membranes was investigated by Chabanon et al. [200], and it was reported that the PP membrane has the highest sensitivity toward wetting conditions among other membranes. PTFE and PMP showed better resistance to wetting.

The key messages of this section are as follows:

- The membrane wetting phenomena increase the mass transfer resistance and limit the long-term process stability.
- Membrane material and absorbent compatibility are important to secure long-term absorption performance.
- The long-term absorption performance of the membrane contactors is predominantly related to the hydrophobicity of the hollow fibers and subsequently the membrane wetting.

9. Challenges in Membrane Contactors

Membrane fouling in membrane contactors has received less attention than membrane wetting. Nevertheless, membrane fouling should not be neglected as the typical flue gas contains contaminants including fine particulates, aerosols that originate from SO₂, and water vapor, which reduces CO₂ removal efficiency in the membrane contactor process [201]. The fine particles in the flue gas may severely impact the performance of the membrane or

even destroy membrane material, which has become an important limitation for the industrial applications of membrane contactors [202]. The availability of suitable membranes with the desired performance appropriate for an intended application is another concern. Although a wide range of membrane products are available in the market, each product is only suited for limited applications. This could be the limiting factor in the application of that technology in practice [203].

The presence of the membrane in the gas–liquid contactors adds additional resistance to mass transfer, which results in a reduction in the overall mass transfer rate. This membrane resistance can be more important if the membranes are wetted by liquid solvents. Membrane wetting has become the main concern for the practical application of membrane contactors, which results in economically unviable operations. The hollow fiber membranes are generally packed randomly in parallel alignment into a shell. The non-uniform fiber distribution can cause severe fluid channeling and bypassing on the shell side of the module which results in the decline of the mass transfer process. These underperformances are often attributed to the disadvantages in the configuration that cause limitations to its application in the industry [116].

Another major challenge with membrane contactors is the selection of an ideal membrane–absorbent combination. The most important requirements of absorbent in membrane gas absorption applications are that the long-term use of absorbent should not damage the membrane either physically or chemically and that the membrane–absorbent combination should have sufficiently high critical entry pressure to avoid wetting. It is difficult to maintain the membrane performance in long-term operation. Most membranes do not have the resilience in practical industry conditions and quickly fail, which is one of the biggest challenges for their potential applications in industrial practice. The regeneration process in membrane contactors has the ability to minimize the temperature required for the solvent regeneration process; however, the additional energy cost to maintain the reduced partial pressure in the gas side by sweeping gas and/or vacuum must be considered. It is important to remark that energy duty contributions will be different depending on the membrane material and their operational limits.

10. Conclusions

Membrane contactors are considered one of the most promising technologies in gas–liquid absorption processes. This review summarized the recent advances in membrane contactor-based gas separation. It was found that the success of a membrane contactor process will strongly depend on the type of membrane material chosen and its fabrication technique. The material must be thermally and chemically compatible with the absorbent and it must be processable to ensure it can be manufactured at scale for a moderate cost. A membrane material with a high contact angle with the solvent is preferable as it decreases the potential for pore wetting. The ceramic hollow fiber membranes showed better mechanical and thermal stability compared to polymeric membranes, which makes them suitable candidates for high-temperature and long operating applications. The addition of non-solvent additives into the polymer dope, surface treatment, surface coating, and the combination of two polymers have been suggested to modify the membrane morphology and structure. The composite membranes with dense skin layers exhibited strong anti-wettability to absorbents and long-term stability even for several days. Compared to flat-sheet membranes, hollow fiber membranes have the advantages of large gas–liquid contact areas, high surface-to-volume ratio, self-supporting characteristics, and ease of scale-up. Chemical absorbents play a critical role in the absorption and desorption processes in membrane contactors. Good chemical compatibility, fast CO₂ reactivity, low regeneration energy, and high surface tension were reported to have the greatest influence on the capture cost and so were usually prioritized. In this regard, the latest developments in absorbents have been discussed, and their advantages and limitations were pointed out. The operating conditions, among other parameters, have a strong influence on the efficiency of the separation and can lead to an economical strategy. Therefore, the effects of

important operating parameters such as gas and liquid flow rate, membrane area, module length, membrane porosity, fiber diameter, operating pressure, number of fibers, module packing density, absorbent concentration, and temperature on the CO₂ separation in the HFMC were comprehensively investigated. The integration of two technologies of the conventional process with membrane separation was represented as an effective way to overcome the issue of the energy penalty. Finally, to make gas–liquid membrane contactor technology more competitive on an industrial scale, the long-term stability of the membrane should be improved.

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Nomenclature

ArgK	Potassium arginate
AEEA	(2-Aminoethyl)ethanolamine
AMP	2-amino-2-methyl-1-propanol
CH ₄	Methane
CO ₂	Carbon dioxide
DGA	Diglycolamine
DEAB	Diethylamino-2-butanol
DMEA	Dimethylethanolamine
DEEA	Diethylethanolamine
DIPA	Diisopropanolamine
DEA	Diethanolamine
EDA	Ethylenediamine
GlyK	Potassium glycinate
K-Pro	Potassium proline
K-Thr	Potassium threoninate
K-Phe	Potassium phenylalanine
K-Arg	Potassium arginate
K-Ala	Potassium alaninate
K-Tau	Potassium taurinate
K ₂ CO ₃	Potassium carbonate
LysK	Potassium lysinate
LiCl	Lithium chloride
MDEA	Methyldiethanolamine
MEA	Monoethanolamine
N ₂	Nitrogen
NaOH	Sodium hydroxide
NH ₃	Ammonia
PI	Polyimide
PES	Polyethersulfone
PS	Polysulfone
PTFE	Polytetrafluoroethylene
PVDF	Polyvinylidene fluoride
PP	Polypropylene
PEI	Polyetherimide
PSF	Polysulfone

PE	Polyethylene
PPO	Poly(phenylene oxide)
PMP	Poly(4-methyl-1-pentene)
PDMS	Polydimethylsiloxane
PVP	Polyvinyl pyrrolidone
PEG	Polyethylene glycol
PA	Phosphoric acid
PEG-400	Polyethylene glycol
PZ	Piperazine
PZEA	2-(1-piperazinyl)-ethylamine
SO ₂	Sulfur dioxide
SarK	Potassium sarcosinate
TEA	Triethanolamine
1DMA2P	1-Dimethylamino-2-propanol
2MPZ	2-methylpiperazine
[emim][EtSO ₄]	1-ethyl-3-methylimidazolium ethylsulfate
[Bmim][BF ₄]	1-butyl-3-methylimidazolium tetrafluoroborate
[Emim][BF ₄]	1-Ethyl-3-methylimidazolium tetrafluoroborate
[apmim][BF ₄]	1-(3-aminopropyl)-3-methyl-imidazolium tetrafluoroborate
[Emim][Ac]	1-ethyl-3-methylimidazolium acetate

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