



# Hybrid CO<sub>2</sub> capture processes consisting of membranes: A technical and techno-economic review

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## ABSTRACT

Because of the greenhouse effect, there is a pressing need to restrict and reduce CO<sub>2</sub> emissions. Post-combustion capture technology is a type of widely used technologies for CO<sub>2</sub> capture. Compared to the standalone CO<sub>2</sub> capture processes such as absorption and cryogenic separation, hybrid CO<sub>2</sub> capture processes demonstrate improved separation efficiency and capacity for the overall performance. Membrane separation is a great candidate for process hybridization with other CO<sub>2</sub> capture processes. Three categories of hybrid processes consisting of membrane technology, i.e., in-series, parallel and integrated configurations, have been applied for CO<sub>2</sub> capture. This paper mainly reviews the recent research progresses on the process development as well as the techno-economic analyses of the hybrid processes corresponding to these configurations. Furthermore, the perspectives on future directions of hybrid CO<sub>2</sub> capture processes are discussed to facilitate its research and practical applications.

## 1. Introduction

As the main component of greenhouse gas, carbon dioxide (CO<sub>2</sub>) emissions have increased rapidly. From the latest data, global annual CO<sub>2</sub> emissions increased by 4.8 % from 33.3 GtCO<sub>2</sub> in 2020 to 34.9 GtCO<sub>2</sub> in 2021 [1]. Most of the CO<sub>2</sub> in the atmosphere originates from human activities, including electricity and heat generation, agricultural practices, factories and other economic activities [2]. As of 2016, worldwide CO<sub>2</sub> emissions amounted to approximately 32.3 Gt, of which a major portion comes from the combustion of fossil fuels [3]. The massive emission of CO<sub>2</sub> has caused the aggravated greenhouse effect, which has a devastating impact on the environment and the natural habitats. Considering the current technological maturity of renewable energy supply for large-scale applications, although coal-fired power generation is one of the main sources of greenhouse gas emissions, it will continue to be crucial in the foreseeable future. Therefore, CO<sub>2</sub> capture is an essential approach to mitigate greenhouse gas emissions [4]. One of the

potentially scalable technologies to reduce CO<sub>2</sub> emissions is CO<sub>2</sub> capture and storage (CCS). It is predicted that CCS can contribute about 19 % of the required emission reduction by 2050 [3].

The conventional CO<sub>2</sub> capture processes include absorption [5,6], adsorption [7,8], cryogenic distillation [9,10] and membrane technology, etc. [11]. Each of these processes has its advantages and limitations. With the development and application of new membrane materials and absorbents, membrane synergistic capture process gradually attracts attention. Hybrid CO<sub>2</sub> capture processes using membranes offer many advantages over conventional processes, such as lower energy consumption, smaller equipment size, high flexibility and easy operation and maintenance. Therefore, membrane-hybridized CO<sub>2</sub> capture processes have demonstrated great potential for practical applications.

In recent years, there have been several reviews of CO<sub>2</sub> capture technology focusing on the materials of absorbents [12,13], adsorbents [12–15] and membranes [16–18] as well as the CO<sub>2</sub> capture processes [12,13,19,20]. Among them, some recent reviews discuss adsorbents and

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introduce the development of ionic liquids [13], carbon-based adsorbents [14] and porous materials [15] as the next-generation CO<sub>2</sub> capture adsorbents. With the progress in carbon emission reduction and carbon capture, there have been emerging needs for more choices of different carbon capture technologies [21,22]. A hybrid process consisting of different carbon capture technologies can be more efficient than a single technology for certain applications. However, summary and discussions on the hybrid processes are quite limited. Bhattacharyya discusses the design of hybrid membrane-solvent-processes for post-combustion CO<sub>2</sub> capture in a short review [20]. While complementing the recent reviews, this review highlights the development of membrane-hybridized CO<sub>2</sub> capture processes for post-combustion carbon capture. Compared with the traditional standalone process, the advantages of the hybrid process are not only reflected in the CO<sub>2</sub> recovery efficiency and low energy consumption, but also in the economics.

In this review, first, traditional CO<sub>2</sub> capture technologies are briefly introduced. Next, recent progresses on hybrid CO<sub>2</sub> capture processes involving membrane technology are elaborated. The hybrid CO<sub>2</sub> processes reported in the literatures are classified into three types, i.e., the in-series, parallel and integrated arrangements. After expounding the economic analysis of the standalone and hybrid CO<sub>2</sub> capture processes, the prospects and challenges of the hybrid CO<sub>2</sub> capture processes are discussed. This review not only presents the fundamentals of the practical CO<sub>2</sub> capture processes but also sheds light on future research directions of hybrid CO<sub>2</sub> capture technologies.

## 2. Conventional CO<sub>2</sub> capture methods

Each CCS technology has its merits and drawbacks depending on the nature of the capture process. The comparison between different technologies as well as their technology readiness levels (TRL) are tabulated in Table 1. The TRL is categorized as following: TRL1-Concept, TRL2-Formulation, TRL3-Proof of concept (lab tests), TRL4-Lab prototype, TRL5-Lab scale plant, TRL6-Pilot scale, TRL7-Demonstration, TRL8-Commercial refinement required, and TRL9-Commercial [23].

Chemical absorption, with its high CO<sub>2</sub> capture efficiency, scalability, and process maturity, is currently carried out in million-ton industrialized applications. And it is the current CO<sub>2</sub> capture technology with high technology readiness level (TRL9). CO<sub>2</sub> is mainly captured through its reaction with the solvent to form carbonate or bicarbonate. CO<sub>2</sub> can be desorbed from the solvent through heating [24]. Amine-based solvents are the most widely used absorbents, including monoethanolamine

**Table 1**  
Comparison of conventional separation technologies.

Technology	Advantages	Disadvantages	Technology readiness level (TRL)
Absorption	a. Highly selective b. High capture efficiency	a. High energy consumption for regeneration b. Highly corrosive	<b>TRL 9</b> Mature
Cryogenic	a. Easy to transport	a. High investment and operation costs b. Strongly influenced by humidity	<b>TRL 9</b> Mature
Adsorption	a. Thermal and chemical stability b. Low energy consumption for regeneration	a. High investment and operation costs b. Complex processes	<b>TRL 8</b> Sorbent technologies are generally less developed than solvents
Membrane	a. Low complexity b. Low energy consumption c. Environmentally friendliness	a. Relatively low capture CO <sub>2</sub> purity b. Incurance of high ancillary power consumption	<b>TRL 8</b> Pilot-scale for CO <sub>2</sub> capture from flue gas

(MEA), *N*-methyl diethanolamine (MDEA), and piperazine (PZ), etc. [5, 6]. However, the high regeneration energy consumption (>3 GJ/tCO<sub>2</sub>) is the major limit for its wide and practical application [25]. The regeneration energy consumption in this type of CO<sub>2</sub> capture process accounts for about 80 % of the total operating cost [26]. In addition, the absorption method has other drawbacks, for instance, poor economy for feed with low CO<sub>2</sub> concentration, solvent degradation and oxidation, and high equipment corrosion caused by the absorbents in particular at high temperatures [27].

The adsorption method is an alternative technology to capture CO<sub>2</sub> from flue gas with broad prospects. The most common ones are pressure swing adsorption (PSA) and temperature swing adsorption (TSA) [28]. PSA consists of high-pressure adsorption and low-pressure desorption to separate and recover CO<sub>2</sub> [29]. TSA is conducted by low-temperature CO<sub>2</sub> absorption and high-temperature CO<sub>2</sub> desorption to capture CO<sub>2</sub> [30]. Separation is achieved by the adsorption and desorption of gases on a solid surface. Hence, it can provide remarkable energy savings and mitigate the limitation of the standard amine-based absorption capture [7,8]. However, the application of CO<sub>2</sub> capture by adsorption is restricted by its high operating and capital costs and complex process control resulting in a TRL of TRL8 [31].

The cryogenic CO<sub>2</sub> capture technology utilizes the difference in liquefaction temperature of different gases to separate CO<sub>2</sub> from flue gas [32]. This process usually involves phase transitions to produce a binary/tertiary system to remove undesired components from a gas mixture. The advantage of the cryogenic method is to achieve higher CO<sub>2</sub> purity (99.9 %) and recovery (99.9 %) than the other separation technologies [10]. However, cryogenic separation is less employed commercially due to the high capital and operation costs [9]. Employing cold energy sources (e.g., liquid natural gas) could be a feasible route to reduce the energy requirement of the cryogenic separation.

Membrane separation technology is another promising CO<sub>2</sub> capture technology with the advantages such as simple operation, modular structure, small footprint and no harmful by-product emissions [33]. The current technological maturity of membrane separation technology is still in the developmental stage (TRL8). Two parameters of the membrane separation process are of the utmost importance, i.e., permeability and selectivity. The former mainly affects separation efficiency, and the latter determines the CO<sub>2</sub> concentration in the permeate gas (CO<sub>2</sub> purity). Ideally, the membranes used in CO<sub>2</sub> capture processes should have both high CO<sub>2</sub> permeability and selectivity. However, overcoming the permeability-selectivity trade-off is one great challenge for membrane development [34]. In addition, the membrane separation performance is closely related to the permeate/feed pressure ratio and selectivity, making it difficult to achieve high CO<sub>2</sub> recovery and purity simultaneously [11]. Yang et al. pointed out that the minimal selectivity is a function of the permeate/feed pressure ratio [24]. When the permeate/feed pressure ratio is close to 0, selectivity needs to be higher than 300 to achieve 95 % CO<sub>2</sub> purity and 90 % CO<sub>2</sub> recovery. The product purity is therefore limited by the CO<sub>2</sub> concentration and the trade-off between product purity and recovery.

Membrane technology can effectively concentrate CO<sub>2</sub> at moderate concentration but struggle to achieve a high purity. Therefore, it would be more effective to employ membranes for bulk CO<sub>2</sub> removal in a hybrid process. On the other hand, absorption (or adsorption, cryogenic) could achieve the required purity but has a significant equipment footprint in capturing CO<sub>2</sub>. Despite the complexity of these processes, the CO<sub>2</sub> capture cost of membrane separation is still competitive with other separation technologies.

## 3. Hybrid CO<sub>2</sub> capture processes

A hybrid process consists of at least two conventional technologies (absorption, adsorption, membrane, and cryogenic, etc.). Among the hybrid systems, the combination of absorption and membrane technology is the most studied. The liquid absorbents in the absorption-

membrane process usually include primary amine (e.g., MEA), secondary amine (e.g., PZ, diethanolamine (DEA)), tertiary amines (e.g., triethanolamine (TEA)), and blended amines (e.g., MDEA, *N,N*-diethylethanolamine (DEEA)-MEA) [35–39]. The energy requirement of CO<sub>2</sub> capture using different alkanolamines in absorption-membrane is low compared to conventional standalone absorption [35]. Besides, the adsorption-membrane method consists of adsorption processes such as pressure swing adsorption (PSA) or vacuum swing adsorption (VSA) and membrane units [40,41]. For example, Jaschik et al. constructed a hybrid system of a four-column VSA unit and a membrane unit that could improve CO<sub>2</sub> capture in terms of energy consumption and absorbent productivity, reducing regeneration energy consumption to 1.54–1.56 GJ/tCO<sub>2</sub> [42]. Belaisaoui et al. found that post-combustion CO<sub>2</sub> capture using membrane-cryogenic hybrid technology could achieve a target of 86 % CO<sub>2</sub> recovery and 90 % CO<sub>2</sub> purity at low energy consumption [43]. The hybrid processes have shown great potential for CO<sub>2</sub> capture with good CO<sub>2</sub> capture performance and greater flexibility in the selection of process operating parameters. Compared to the standalone CO<sub>2</sub> capture process, the hybrid system reduces the energy penalty for solvent regeneration and the replacement costs of degraded solvents.

The hybrid CO<sub>2</sub> capture processes can be categorized into three main configurations, i.e., in-series, parallel and integrated arrangements. For the hybrid process, the in-series arrangement is a prevailing way to couple different CO<sub>2</sub> capture processes due to its unique CO<sub>2</sub> capture performance, which recently became a hot research topic. Therefore, the in-series arrangement is stressed in this review. Besides, the integrated arrangement, which includes the well-studied membrane contactor process, is another elaborated process.

### 3.1. In-series hybrid processes

The in-series arrangement is the most used configuration in the hybrid process. The membrane unit is usually arranged in series with another carbon capture technology (e.g., absorption, adsorption and cryogenic, etc.). The order in which membrane units is arranged usually depends on the function of the membrane process. Usually, membrane processes are divided into two categories: membrane as a pre-treatment unit and as a post-treatment unit.

#### 3.1.1. Membrane as pre-treatment

For the first type of in-series hybrid processes, the membrane serves as a pre-treatment unit before another carbon capture technologies. Generally, the purpose is to improve the CO<sub>2</sub> concentration that enters the subsequent unit (absorber or cryogenic system). In Table 2, the CO<sub>2</sub> capture performance of the membrane unit as a pre-treatment unit is summarized. For example, as illustrated in Fig. 1(a), using a membrane unit for CO<sub>2</sub> pre-concentration lightens the burden of the absorption system and reduces the amount of absorbent used [38,39,44,45]. This reduces both the necessary liquid to gas ratio in the absorber (L/G, the ratio of the total mass flow rate of amine solution to the total mass flow rate of gas) and the reboiler duty and hence realizing energy savings [46]. For example, Gilassi et al. proposed a hybrid membrane and enzymatic absorption process that can significantly reduce the solvent circulation rate in the absorption and desorption units, and the energy

consumption for CO<sub>2</sub> regeneration [27]. Compared to the standalone process, the L/G ratio was reduced by 26 %, 43 %, and 65 % for intermediate CO<sub>2</sub> capture of 30 %, 50 %, and 70 %, respectively [27]. Colin et al. used MEA as a solvent hybridized with an H<sub>2</sub> selective membrane for CO<sub>2</sub> post-combustion capture. This hybrid process was less costly than the absorption process using MEA [47]. Similarly, the energy consumption of CO<sub>2</sub> captured by different commercial amines combined with a single-stage membrane was examined under a similar arrangement. The total power consumed by cooler, pump, regenerator reboiler and regenerator condenser was obtained using MEA (13.1 MW), while MDEA (17.2 MW) was evaluated to have the highest total power consumption [48]. Cormos et al. assessed the CO<sub>2</sub> capture performance of coal-based super-critical power plants using an absorption-membrane process by varying the location of the membrane units [16]. The design parameters of the CO<sub>2</sub> capture process for a natural gas combined cycle (NGCC) power plant were optimized under this arrangement [37]. The optimal values for the feed/permeate pressure ratio and flow rate ratio are 10:1 and 50 %, respectively, which reduces the solvent regeneration load of the hybrid process by more than 20.7 % compared to the conventional chemical absorption process [37]. Steffen et al. reported an absorption-membrane process for CO<sub>2</sub> capture in the oxidative coupling of methane (OCM) process, which was experimentally validated in a small-scale plant [49,50]. In addition, the hybrid process using PZ-activated MDEA can reduce column height by 25 % and save 20 % of energy, but at the same time double the product loss compared to the standalone absorption process [50]. To sum up, the in-series arrangement has a wide range of application scenarios, such as OCM, NGCC power plants, enhanced oil recovery (EOR), and coal-based super-critical power plants [37,38,49,51].

In the membrane-cryogenic hybrid process, as shown in Fig. 1(b), CO<sub>2</sub> from flue gas could be initially concentrated via the membrane. Then a high concentration CO<sub>2</sub> stream is cryogenic-separated via phase change with decreased energy consumption. It has been reported that membrane-cryogenic hybrid processes reduce CO<sub>2</sub> capture costs with minimal environmental impact because of the small membrane area and reduced energy consumption for compression [54,55]. In 2012, Belaisaoui et al. first proposed to combine membrane and cryogenic processes and verified its feasibility [43]. The hybrid process can achieve 90 % CO<sub>2</sub> purity and 85 % CO<sub>2</sub> recovery through simulation and analysis with an energy consumption lower than 2 GJ/tCO<sub>2</sub> [56]. When the membrane was used as a pre-treatment process in the membrane-cryogenic hybrid process for CO<sub>2</sub> capture from flue gases in coal-fired (13.6 % CO<sub>2</sub>) and blast furnace (23.2 % CO<sub>2</sub>), CO<sub>2</sub> capture energy consumption was 1.65 GJ/tCO<sub>2</sub> and 1.99 GJ/tCO<sub>2</sub>, respectively, and the capture costs were 36.14 \$/tCO<sub>2</sub> and 28.81 \$/tCO<sub>2</sub>, respectively. Moreover, it has the potential to be scaled up industrially [57]. Dragan et al. proposed a modeling and optimization framework for combined PSA and membrane processes for gas separations [58]. Under this arrangement, performance enhancements including increase in overall hydrogen recovery (2 %), a significant reduction in bed size (up to 46 %), increased adsorbent production efficiency (up to 184 %), captured CO and CO<sub>2</sub> concentrations (up to 76 % combined CO and CO<sub>2</sub>) could be achieved [58].

**Table 2**  
Membrane as a pre-treatment unit.

Process	Gas Composition	Recovery rate (%)	Purity (%)	Energy consumption (GJ/tCO <sub>2</sub> )	Ref
Membrane-Absorption	4.2%CO <sub>2</sub> +81.7%N <sub>2</sub>	5–30	–	3.8–4.1	[34]
Membrane-Absorption	–	–	96.3	2.41	[35]
Membrane-Absorption	12.1%CO <sub>2</sub>	90.6	84.7	1.6	[44]
Membrane-Cryogenic	13.7%CO <sub>2</sub> +72.9N <sub>2</sub>	95	70	1.11	[49]
Membrane-Cryogenic	15%CO <sub>2</sub> +85%N <sub>2</sub>	90.0	–	1.7	[52]
Membrane-Cryogenic	40%CO <sub>2</sub> +60%CH <sub>4</sub>	98.2	98.0	0.8 GJ/tCH <sub>4</sub>	[53]
Membrane-Cryogenic	13.5–23.2%CO <sub>2</sub>	90.0	95.0	1.65–1.99	[53]
Membrane-Cryogenic	15–30%CO <sub>2</sub>	90	94	3	[43]

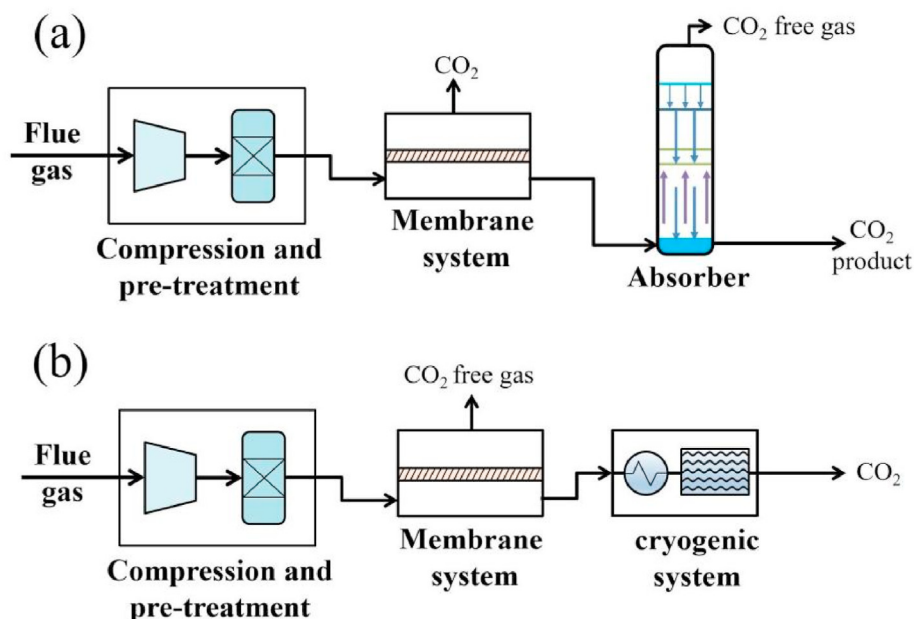


Fig. 1. Schematic diagram of in-series arrangement: (a) the membrane system is placed before the cryogenic system and (b) the membrane system is placed before the cryogenic system.

### 3.1.2. Membrane as post-treatment

When membrane units are placed after other separation units, the main purposes are to increase CO<sub>2</sub> capture ratio, reduce amine emissions or perform CO<sub>2</sub> desorption using specific membrane materials. The CO<sub>2</sub> capture performance of the membrane unit as a post-treatment unit is summarized in Table 3. In the first case, the flue-gas first goes through the absorption unit to remove a fraction of the CO<sub>2</sub>, whereas the rest is stripped in the subsequent selective membrane, as shown in Fig. 2(a). Freeman et al. found that in an absorption-membrane hybrid system, about half of the CO<sub>2</sub> in the flue gas was removed by the absorber, followed by additional separation by the membrane contactor to achieve 90 % total removal of CO<sub>2</sub> [59]. Meanwhile, the absorber operated at a higher lean-loading state and also benefited from the downstream membrane to mitigate fugitive amine emissions. As shown in Fig. 2(b), the cryogenic-membrane hybrid process can be used in CO<sub>2</sub>-enhanced oil recovery (CO<sub>2</sub>-EOR), and the energy consumption of the whole system was less than 0.85 GJ/tCO<sub>2</sub> [60]. This configuration can also be used in the OCM process. Simulation was used to evaluate the feasibility of the hybrid membrane-absorption system on an industrial scale. The results show that the hybrid process has more economic advantages than the standalone absorption process [46].

This arrangement can be used in the Rectisol process, where the CO<sub>2</sub> is separated from the absorption unit, followed by a high CO<sub>2</sub> purity permeate stream through a CO<sub>2</sub>-selective gas separation membrane [61]. In addition to the above hybrid process (absorption-membrane, cryogenic-membrane), the combination of the adsorption process (PSA,

VSA) and membrane separation has also been used for CO<sub>2</sub> capture [42, 62].

In particular, as shown in Fig. 2(c), Ataievarjovi et al. proposed a new approach by solvent absorption and membrane desorption, which significantly decreased the energy consumption (0.433 GJ/tCO<sub>2</sub>) by about 75 % compared with the gas stripping method [63]. Among them, CO<sub>2</sub> is absorbed by solvents, and then desorbed by a pervaporation (PV) membrane [63]. In this method, solvent absorption and membrane desorption were used for CO<sub>2</sub> capture under medium or high CO<sub>2</sub> partial pressure (3–4 MPa), such as the integrated gasification combined cycle (IGCC) [35].

### 3.2. Parallel hybrid processes

In the parallel arrangement, the flue gas leaving the power plant is split and treated by different systems in parallel. As shown in Fig. 3, Zhang et al. proposed a hybrid system for CO<sub>2</sub> capture in a parallel arrangement for NGCC power plants combining amine absorption with membrane technology [64]. As shown in Fig. 4, the effectiveness of membrane processes and feasibility of hybrid processes combining membrane permeation and conventional amine absorption processes were investigated by Kundu et al. This process was used to obtain high CO<sub>2</sub> purity (>80 %) [65]. Chowdhury et al. evaluated the parallel arrangement by varying the number of absorption and membrane trains [66]. They found that the parallel configuration consumes about 14 % more energy under the best-case scenario than the standalone MEA system [66]. Compared with the in-series arrangement, the main advantages of the parallel arrangement are to reduce the absorber size to half of the original one in the in-series arrangement as well as the capital cost. The gas treated by the absorber is less in volume and has a higher loading difference, so the absorber and direct contact cooler can be much smaller. Reboiler duty could also be decreased. In a short summary, the hybrid process could benefit from the cost savings in two ways: a reduction in regeneration energy in the in-series case and a reduction in capital costs in the parallel case.

### 3.3. Integrated hybrid processes

The major problem of conventional CO<sub>2</sub> capture technology is the

Table 3  
Membrane as a post-treatment unit.

Process	Gas Composition	Recovery rate (%)	Purity (%)	Energy consumption (GJ/tCO <sub>2</sub> )	Ref
Absorption-Membrane	CO <sub>2</sub> +80%N <sub>2</sub>	77	–	–	[55]
Absorption-Membrane	CO <sub>2</sub> +H <sub>2</sub>	90	95	–	[57]
Adsorption-Membrane	15% CO <sub>2</sub> +85%N <sub>2</sub>	99	–	–	[58]
Cryogenic-Membrane	>90%CO <sub>2</sub>	99	96	0.85	[61]

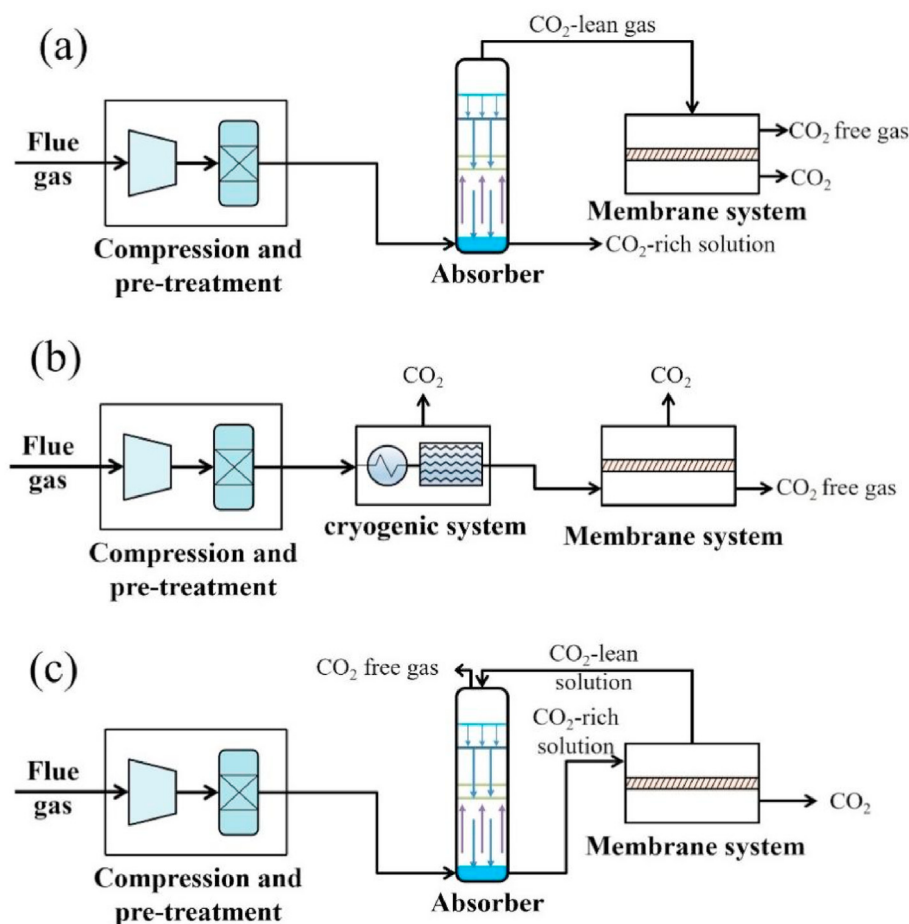


Fig. 2. Schematic diagram of the in-series arrangement: (a) the absorption-membrane system, (b) the cryogenic-membrane system and (c) the absorption-membrane system with membrane desorption.

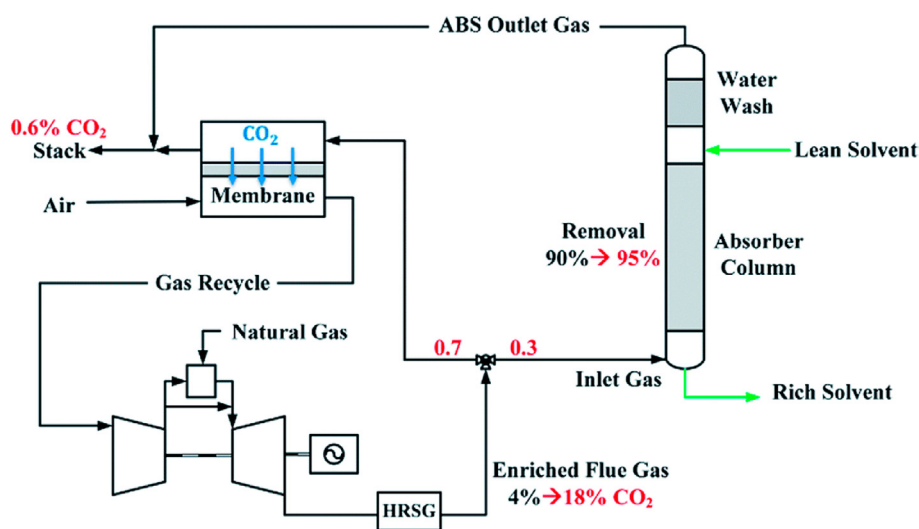


Fig. 3. Process diagram of an NGCC plant with membrane-amine hybrid carbon capture. Reprinted with permission from Ref. [64]. Copyright (1972) Royal Society of Chemistry.

high energy consumption used for solvent recovery and secondary separation. By combining the solvent absorption and membrane separation to form an integrated process, the integrated absorption-membrane process may achieve the economic and technical feasibility.

### 3.3.1. Membrane contactor

With the development of membrane technology, membrane contactor has been used as an alternative to a typical packed column for CO<sub>2</sub> separation. With the feed gas and absorbents flowing on both sides of the membrane, the membrane contactor integrates absorption and

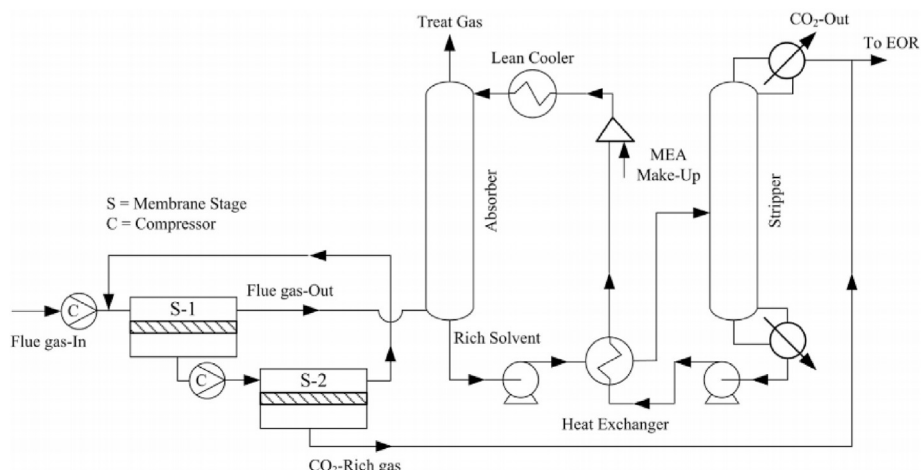


Fig. 4. A hybrid membrane and amine processes to treat flue gas of cement plant. Reprinted with permission from Ref. [65]. Copyright (2014) Elsevier.

membrane separation into one process. Membranes which act as the medium to separate the gas and liquid phases are the core part in this process. They not only physically separate the two phases but also significantly affect the mass transfer of the components. Membrane contactors have the advantages such as low energy consumption, known contact area, easy scale-up and improved selectivity. Compared to a packed column, when the overall mass transfer of the two processes are similar, the exchange area in the membrane contactor would be lower and the volume would be smaller [67]. For the optimization of the overall system, the design of membrane modules and the control of operating conditions should also be considered. In general, CO<sub>2</sub> absorption performances of membrane contactors depend upon: membranes,

absorbents, module design and operating conditions, as illustrated in Fig. 5.

**3.3.1.1. Membranes.** Membrane contactors currently used in industry include both polymeric membranes and ceramic membranes. Ideally, the membrane used in a membrane contactor should be both hydrophobic and porous so that it can prevent membrane pore wetting and allow CO<sub>2</sub> to pass through efficiently. To achieve the goals, the selection of membrane materials as well as various surface modifications of the membrane surface and the introduction of biocatalytic membranes have been explored.

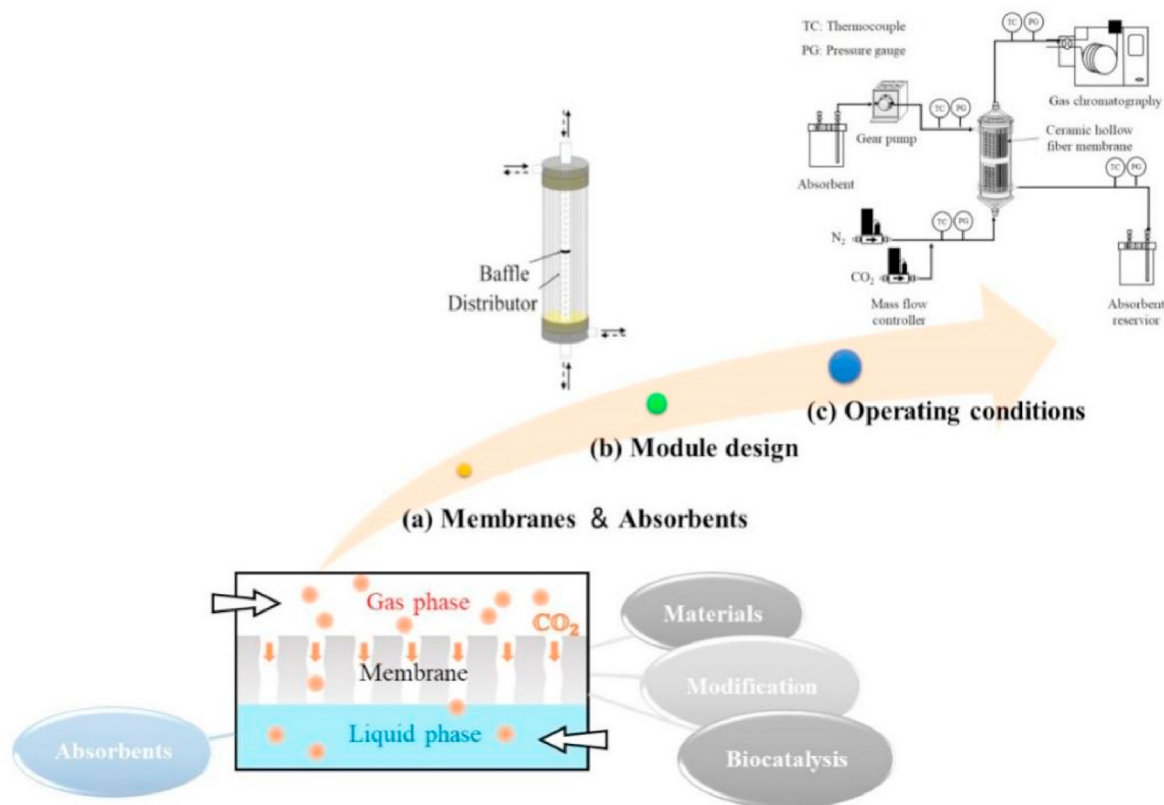


Fig. 5. Illustration of CO<sub>2</sub> absorption in membrane contactors. (b) Is reprinted with permission from Ref. [68]. Copyright (2019) Elsevier. (c) Is reprinted with permission from Ref. [69]. Copyright (2020) Elsevier.

### (1) Selection of materials

Many studies focus on the exploration of suitable materials to make membranes hydrophobic and stable for membrane contactors. Considering both hydrophobicity and chemical stability, polymeric materials, such as polypropylene (PP), polyvinylidene fluoride (PVDF) and polytetrafluoroethylene (PTFE), are the most used to prepare the desired membranes [70,71]. Other promising novel polymers for membrane contactors include poly (vinylidene fluoride-co-hexafluoro propylene) (PVDF-HFP) with the fluorine-rich HFP moiety to enhance hydrophobicity [72] and cyclic olefin polymers (COP) with a rigid bridged ring structure to prevent crystallization [73]. In addition, ceramic membranes are also used which can prevent the absorbent from corroding the membranes. The widely used materials in ceramic membranes are kaolin clay or alpha alumina [74,75].

During the CO<sub>2</sub> capture process, the amine solvents and their absorption by-products swell or corrode the membrane materials as the reaction proceeds, which would change the inherent properties of membranes and hence causing membrane wetting and degradation of membrane performance [76]. To resolve this issue, membrane surface modifications were employed to improve membrane properties and separation performances.

### (2) Surface-modified membranes

High hydrophobicity is essential for membranes used in membrane contactors to hinder the membrane pores from being wetted. Modification of polymeric membranes by surface coating, blending additives in dope solution and surface grafting is prevalent to improve membrane hydrophobicity.

Rosli et al. [77] reported the surface coated PVDF membrane with a moisture-proof protective layer formed by low-density polyethylene-functionalized silica nanoparticles (NPs). With the increase of membrane hydrophobicity, an increase in liquid entry pressure (LEP) from 4.75 bar to 13.55 bar was achieved. According to Chen et al. [78], the incorporation of hydrophobic silica in membrane could lead to the formation of a bilayer hollow fiber membrane. Its outer layer has a porous structure with large finger-like macrovoids, and the inner layer has a thin and dense spherulite stacking structure. The purposely designed structure contains a highly porous outer layer for gas permeation and a hydrophobic inner layer to prevent pore wetting. Similar to modified silica, metal nanoparticles (e.g., TiO<sub>2</sub>, CaCO<sub>3</sub>) [79,80], zeolitic imidazole framework (ZIF) nanosheets [81] and other inorganic additives such as lithium chloride (LiCl) [82] can be incorporated into the

organic membrane matrix to realize superhydrophobic modification. Furthermore, changing the additive concentration can control the hydrophobicity and uniformity of the membranes [82,83].

Grafting hydrophobic functional groups is also a commonly used membrane modification method. Due to the presence of hydroxyl groups (–OH) on the surface of ceramic membranes, ceramic membranes are usually hydrophilic in nature which need to be modified with agents like fluoroalkylsilanes (FAS) [69,84]. Through the reaction between hydroxyl groups of the membrane and silane groups of FAS compounds, the membrane hydrophobicity gradually increased with the increase of soaking time until the desired hydrophobicity was obtained (Fig. 6) [84]. Guo et al. [85] grafted ceramic hollow fiber membranes using hexadecyltrimethoxysilane (C16) ethanol solution. Compared to the commonly used modifier, FAS, C16 is cheaper and less toxic. After grafting, the water contact angle of the membrane increased dramatically from 40° to 132°, realizing the transformation from being hydrophilic to hydrophobic.

### (3) Biocatalytic membranes

The amine-based solvents are the most established absorbents in industry, but they have disadvantages such as amine degradation, swelling polymeric membranes, corrosiveness and toxicity [76]. In recent years, the use of enzymes like carbonic anhydrase (CA) to catalyze the CO<sub>2</sub> capture process with green benign solvents (water) has become an alternative technology [86–88]. CA can catalyze the hydration reaction of CO<sub>2</sub> in an aqueous solution to generate bicarbonate, but as a biocatalytic enzyme, the direct use of free enzyme will make its activity greatly affected by the environment. The strategy to overcome this problem is to fix it on membrane surface.

After immobilizing the enzyme on the membrane surface, the CO<sub>2</sub> absorption flux was increased by about 165 % compared with the non-biologically catalyzed PVDF membrane [86]. The biocatalytic membrane contactor using only distilled water as the absorbent achieved a larger overall mass transfer coefficient compared to the membrane contactor using common amine-based absorbents or aqueous sodium hydroxide solutions [87]. It is worth noting that the activity of biocatalytic enzymes is greatly affected by external conditions, like pH (especially after absorbing CO<sub>2</sub> gas) and temperature [88], so the enzymes in the membranes prepared in the study are generally covalently immobilized with polydopamine (PDA) and polyethyleneimine (PEI).

**3.3.1.2. Absorbents.** The choice of absorbent is another crucial influencing factor in CO<sub>2</sub> capture operations. The absorbents used in industry should preferably be environmentally friendly, easy to regenerate, low

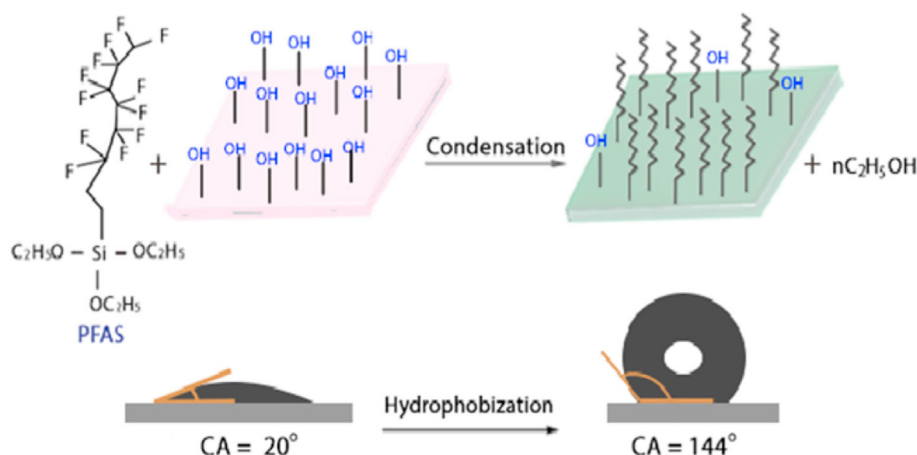


Fig. 6. Scheme of the grafting process by PFAS compounds. Reprinted with permission from Ref. [84]. Copyright (2021) Springer Nature.

toxicity and harmless. At the same time, since the absorbents are mostly thermally regenerated in the carbon capture process, they also need to be thermally stable. Finally, the contact with membrane materials and feed gas requires the absorbents to have high chemical stability and compatibility [89]. Considering the above criteria, the following sections will discuss different types of absorbents used in membrane contactors.

### (1) Amine-based solvents

The absorption of CO<sub>2</sub> by alkanolamines in membrane contactors has been extensively studied. Amine-based solvents can be divided into primary amines, secondary amines and tertiary amines according to the number of hydrogen atoms substituted on nitrogen atoms in the amine molecule [90]. At present, a variety of amine solutions, including primary amines (MEA) and secondary amines (DEA), have been employed in the process of CO<sub>2</sub> capture [91]. In contrast, there is more area for improvement of the tertiary amine absorbent and blended amine-based solvent for practical application.

MDEA is a commonly used cost-effective tertiary amine absorbent, and by dispersing nanoparticles in the solvent can further improve the CO<sub>2</sub> removal rate. Cao et al. [92] found that dispersing 0.5 wt% carbon nanotube NPs into 5 %, 10 %, and 20 % MDEA solution could increase CO<sub>2</sub> removal by 5.32 %, 3.88 %, and 2.70 %, respectively. Moreover, according to Cao et al. [93], a novel tertiary amine, 1-Dimethylamino-2-propanol (1DMA2P), has better CO<sub>2</sub> absorption properties than MDEA.

Alternatively, the use of mixed amine absorbent may be more effective than the monoamine solution. However, the interactions between the amine components are different. Hence, the ratio of different components and the corresponding concentrations are particularly important. Magnone et al. [94] found that compared with the MDEA solution, the CO<sub>2</sub> absorption flux of the mixed solution of MDEA and DEA increased by more than 5 times. The viscosity values of the absorbent are also worth noting. Bernhardsen et al. [95] reported the overall mass transfer coefficient ( $K_{ov}$ ) value decreases with the increase of the viscosity in the mixed absorbent.

### (2) Ionic liquids

Ionic liquids (ILs) are usually composed of organic cations and organic or inorganic anions with melting temperatures below 100 °C which have low energy consumption for regeneration. They are one of the best alternative liquid absorbents to amines [96,97]. However, due to the ionic nature of ILs, most ILs have high viscosity, which slows down CO<sub>2</sub> diffusion and prolongs the production cycle. This kinetic limitation can be overcome with a large liquid-gas interfacial area using hollow fiber membrane contactors [98,99]. The selectivity of ILs for CO<sub>2</sub> capture can be improved by changing the functional group. Imidazolium-based ILs are widely used due to their relatively low viscosity, and the ease of synthesis [100–102]. Swati et al. [101] found that the combination of 1-octyl-3-methyl imidazole tetrafluoroborate [OMIM][BF<sub>4</sub>] and PTFE membrane has good long-term stability. However, the potential harm to the environment and unclear toxicity of ILs need to be further investigated for ILs-based absorbents [103].

### (3) Amino acids

Amino acid salts have attracted researchers' attention in the field of CO<sub>2</sub> absorption because of their low corrosiveness and toxicity, excellent stability, fast reaction rate with CO<sub>2</sub>, and low volatility [104,105]. Gao et al. [106] added potassium glycinate to the MEA solution and the energy consumption of the blended solution is lower than that of pure MEA solution. Under the same conditions, potassium glycinate/MEA solution

has better CO<sub>2</sub> absorption performance than MEA solution.

When there are different kinds of amino acid salts in the blended absorbents, the absorption mechanism is more complicated than that of single amino acid. For the absorption process, the interactions exist not only between amino acids and CO<sub>2</sub>, but also between different components of the absorbent. Gusnawan et al. [107,108] formulated a green biosolvent rich in 18 amino acids with soybean and moringa. The following conclusions were found: 1) the presence of L-arginine has the most significant effect on the flux of CO<sub>2</sub> in soy-based solvents; 2) the presence of hydrophobic amino acid salts reduces the CO<sub>2</sub> absorption activity in the hydrophobic membrane contactor; 3) this biosolvent has comparable CO<sub>2</sub> absorption flux and efficiency to the traditional MEA solvent, reaching 31.2 mol m<sup>-2</sup> s<sup>-1</sup> and 97 %, respectively. Therefore, the potential of biosolvent-based absorbents is well-illustrated.

### (4) Other absorbents

Aqueous ammonia can also react with CO<sub>2</sub> and be used as chemical absorbents. The most prominent advantage is its low regeneration cost. The reactivity of aqueous ammonia is slightly lower than that of MEA. However, at a suitable flow rate, the CO<sub>2</sub> absorption performance of ammonia solution can reach 98 % of that of MEA solutions [109]. A disadvantage of ammonia is the ammonia slip problem [110]. It will not only aggravate solvent consumption, but also cause ammonium contamination.

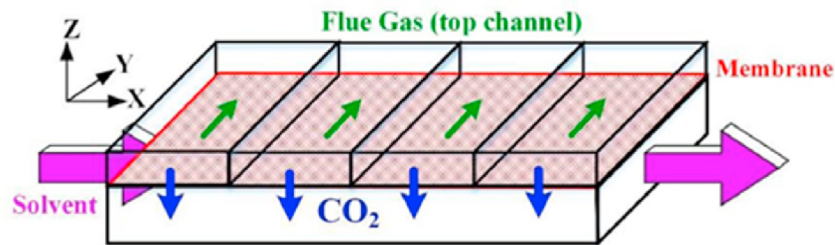
Alkali salts and water are CO<sub>2</sub> absorbents with less toxicity and lower cost than amine solvents. However, the reaction rate between them and CO<sub>2</sub> is low, especially at room temperature and low CO<sub>2</sub> partial pressure [111]. Adding accelerating agents such as metal oxide nanoparticles to the basic fluid to form nanofluids (NFs) may be helpful. The introduction of NPs increases the absorption area of the absorbents and improves the CO<sub>2</sub> absorption rate. At a low liquid flow rate of around 10 ml/min, the CO<sub>2</sub> absorption efficiency of ZnO NFs was 130 % higher than that of distilled water [112]. CO<sub>2</sub> is desorbed at elevated temperature, and then the NPs can be recycled, which solves the problem of solvent waste. To prevent the aggregation of these NPs in the absorbent, ultrasonic irradiation could be conducted [113,114].

**3.3.1.3. Membrane module design.** The membrane module is the most important part in the equipment of CO<sub>2</sub> absorption membrane contactor. The structural shape and configuration of the module seriously affect the gas-liquid mass transfer rate. Membrane modules can be categorized into two types, i.e., flat-sheet membrane modules and hollow fiber membrane modules.

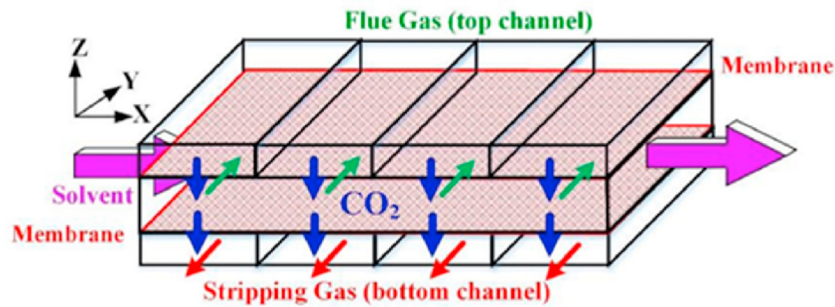
#### (1) Flat-sheet membrane modules

Flat-sheet membrane is known for its simple fabrication. To improve the mass transfer inside the membrane module, Pan et al. [115] studied the design of the membrane module structure and compared three structural modes of the PTFE flat-sheet membrane: pure absorption (A), parallel absorption and stripping (P-AS) and contiguous absorption and stripping (C-AS), as shown in Fig. 7(a–c). The results showed that the parallel-type and contiguous-type membrane contactor significantly improved the adsorption flux compared with the A type, with enhancement ratios of 1.16–1.61 and 1.55–2.21, respectively. Constantinou et al. [116] designed three kinds of flat membrane modules with different structures: single-channel PTFE membrane contactor, nickel mesh contactor and eight-channel PTFE membrane contactor. The PTFE material is not easy to wet, and the eight-channel gas-liquid exchange area is larger. Thus, the average CO<sub>2</sub> flux of the eight-channel PTFE contactor is the highest with the value of 80 mol m<sup>-2</sup> s<sup>-1</sup>.

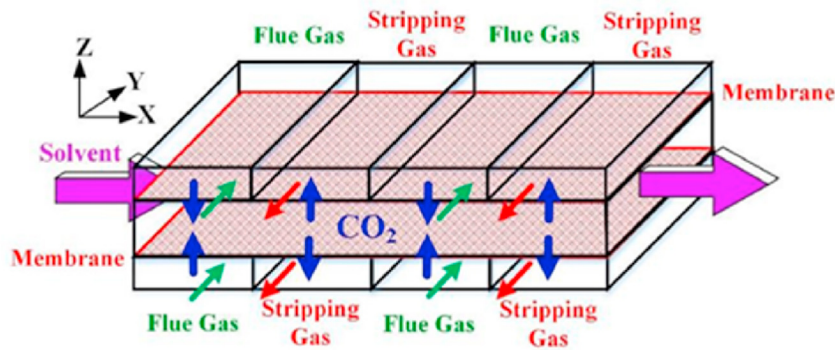
Flat sheet membrane can also be made into spiral wound membrane



(a) A type operation



(b) P-AS type operation



(c) C-AS type operation



(d)



(e)

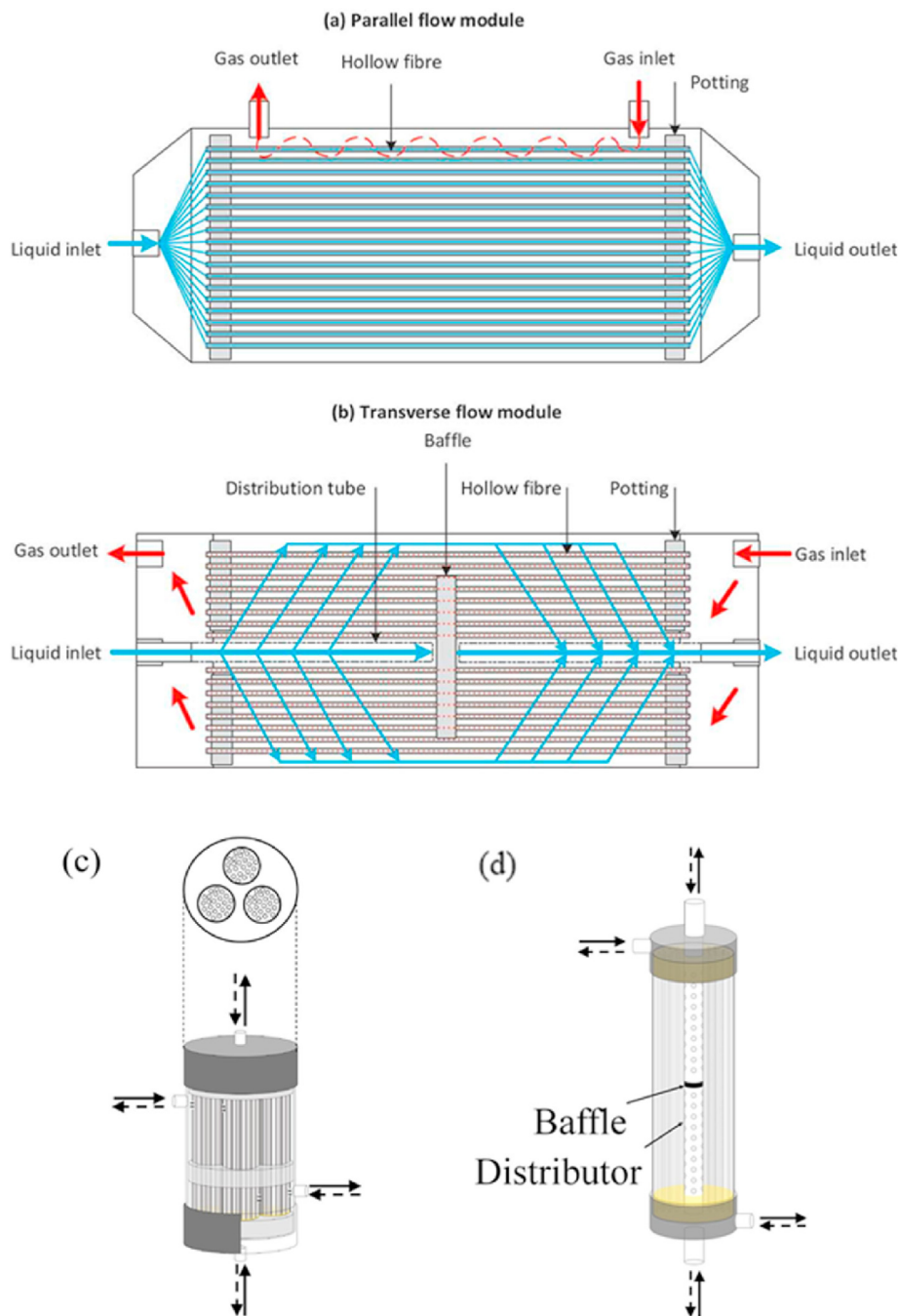
**Fig. 7.** Schematics of (a) pure absorption (A), (b) parallel-type absorption and stripping (P-AS), (c) contiguous-type absorption and stripping (C-AS) membrane contactors. Reprinted with permission from Ref. [115]. Copyright (2017) Elsevier. Empty and spiral wired annulus channels of concentric circular membrane contactors: (d) empty channel and (e) helix channel. Reprinted with permission from Ref. [117]. Copyright (2022) MDPI.

module. Ho et al. [117] designed a concentric membrane module which contains helices with different pitches installed on the outside of the tube (Fig. 7(d and e)). The MEA solution absorbent flowed in the cavity of the spiral concentric tube assembly to generate eddy currents, while the  $\text{CO}_2/\text{N}_2$  gas mixture flowed on the side of the tube. Adding helices can reduce the concentration polarization effect and increase the  $\text{CO}_2$  absorption flux.

## (2) Hollow fiber membrane modules

Although the production process of flat membrane modules is

relatively simple, hollow fiber membrane modules are the most common type among membrane contactors. In practical applications, hollow fiber membrane modules are more efficient than flat membrane modules because of their high surface-to-volume ratio and larger gas-liquid contact area. In addition, they have lower energy consumption and good self-support characteristics. The usual mode of operation is that the  $\text{CO}_2$ -containing feed gas passes through the lumen side of the hollow fibers, while the solvent flows on the shell side of the module.  $\text{CO}_2$  permeates through the membrane and is absorbed by the solvent. The  $\text{CO}_2$ -rich solvent is desorbed, regenerated and recycled [90]. There is little difference in  $\text{CO}_2$  absorption flux between countercurrent and cocurrent flow. Gas-liquid contact performance may be better in countercurrent



**Fig. 8.** Illustration of two most commonly applied flow mode in membrane contactors: (a) parallel flow and (b) cross flow. Reprinted with permission from Ref. [118]. Copyright (2021) Taylor & Francis. (c) 3-parallel connection module, (d) cross-flow advanced module. Reprinted with permission from Ref. [68]. Copyright (2019) Elsevier.

flow [68]. However, the average residence time of the fluid needs to be increased. At present, compared with parallel flow, the commonly used 3 M hollow fiber membrane contactor cross-flow device is equipped with baffle and distribution pipes to solve the above problems (Fig. 8(a and b)) [118]. In the report by Lee et al. [68], as depicted in Fig. 8(c and d), baffles were added to the module to make the gas and liquid phases contact vertically or at specific angles, rather than in the horizontal direction. The baffle minimizes bypassing on the shell side of the module and improves mass transfer characteristics by creating fluid flow perpendicular to the membrane surface. The CO<sub>2</sub> absorption performance of the novel cross-flow module is higher than that of 3-parallel modules with almost the same membrane area.

Normally high L/G will lead to high energy consumption. In most applications, an excess of absorbents is used to achieve a high performance. Chan. et al. [119] proposed a new idea to reduce L/G by increasing the length of the membrane module. Increasing the length from 1 m to 4 m reduces L/G by 70 % and flux by only 16 %. Total energy consumption reduced by 30 % compared to conventional processes.

**Table 4**  
Summary of CO<sub>2</sub> capture performance by membrane contactors.

Membrane	Contact angle (°)	Absorbent	liquid flowrate (mL·min <sup>-1</sup> )	Feed gas (.)	Gas flowrate (mL·min <sup>-1</sup> )	CO <sub>2</sub> flux ( × 10 <sup>-4</sup> mol m <sup>-2</sup> s <sup>-1</sup> )	Ref.
PP	–	potassium glycinate (1 M)	750–1500	CO <sub>2</sub> /N <sub>2</sub> (5–15/95–85)	5000	1.25–3.6	[53]
ceramic	126.3 ± 2.08	MEA 30 wt %	8.33	CO <sub>2</sub> /N <sub>2</sub> (12/88)	–	80	[68]
ceramic	125.11	MEA 30 wt %	–	CO <sub>2</sub> /N <sub>2</sub> (13/87)	30–60	40.8	[69]
PVDF + 5 % PTFE	92 ± 1	DEA (1 M)	50	CO <sub>2</sub> /N <sub>2</sub> (19/81)	100	20.20 ± 0.24	[71]
PVDF-HFP	155	PG 30 wt %	100	pure CO <sub>2</sub>	70	180	[72]
COP + Span 80	100	MEA (1 M)	2500	CO <sub>2</sub> /N <sub>2</sub> (15/85)	140	1.6	[73]
Kaolin/Al <sub>2</sub> O <sub>3</sub>	142	water	100	pure CO <sub>2</sub>	–	1800	[74]
Al <sub>2</sub> O <sub>3</sub>	123 ± 1.86	MEA 30 wt %	9	pure CO <sub>2</sub>	–	45.75	[75]
Silica/PVDF	120.0 ± 0.8	AMP (1 M)	100	CO <sub>2</sub> and N <sub>2</sub>	100, respectively	2.32	[77]
PVDF + 4 % SiO <sub>2</sub>	147.2 ± 2.0	DEA (1 M)	50	CO <sub>2</sub> /N <sub>2</sub> (19/81)	100	31.2	[78]
fTiO <sub>2</sub> /PVDF	125	MEA (5 M)	0.4 m s <sup>-1</sup>	pure CO <sub>2</sub>	–	127	[79]
PVDF + 3 % SA-CaCO <sub>3</sub>	151.3 ± 0.9	water	150	pure CO <sub>2</sub>	100	22.8	[80]
PVDF/(ZIF-8-ZIF-L)	134.5	DEA (1 M)	0.25 m s <sup>-1</sup>	pure CO <sub>2</sub>	1000	30.2	[81]
PVDF + 8%PA	78.6 ± 0.4	distilled water	70	CO <sub>2</sub> /N <sub>2</sub> (19/81)	130	13.1	[82]
SiO <sub>2</sub> /PVDF	155 ± 3	MEA (0.1 M)	60	CO <sub>2</sub> /H <sub>2</sub> S (40/60)	200	3.8	[83]
kaolin- FAS C8	160	water	–	CO <sub>2</sub> /CH <sub>4</sub>	–	–	[84]
α-Al <sub>2</sub> O <sub>3</sub>	132	MEA (5 M)	–	0.41 mol CO <sub>2</sub> /mol Absorbent	200–400	11.7	[85]
Biocatalytic PVDF	59.7 ± 2.2	water	0.25 m s <sup>-1</sup>	pure CO <sub>2</sub>	–	25	[86]
Biocatalytic PP	78	Water and 100 mM buffer with 5.4 mg/L <sub>reactor</sub> CA immobilized on MNPs	34	CO <sub>2</sub> /N <sub>2</sub> (15/85)	100	–	[87]
Biocatalytic PP	–	Water and 100 mM buffer	26	CO <sub>2</sub> /N <sub>2</sub> (15/85)	100	2.9	[88]
PTFE	–	1DMA2P (2 M)	0.024 m s <sup>-1</sup>	CO <sub>2</sub> partial pressure 15 kPa	0.239 m s <sup>-1</sup>	9	[93]
α-Al <sub>2</sub> O <sub>3</sub>	–	MEA, DEA, MDEA, AMP, PZ	5	CO <sub>2</sub> /N <sub>2</sub> (15/85)	50	1.16–6.98	[94]
PP	–	MDEA 10 wt % + [Bmim][BF <sub>4</sub> ] 20 wt %	15–55	CO <sub>2</sub> /N <sub>2</sub> (34.5/65.5)	70	6	[100]
PP	110	[OMIM][BF <sub>4</sub> ] 50 wt %	–	CO <sub>2</sub> /N <sub>2</sub> (45/55)	–	6.62	[101]
PTFE	142	[EMIM][EtSO <sub>4</sub> ] 50 wt %	–	CO <sub>2</sub> /N <sub>2</sub> (45/55)	–	6.53	[101]
PP	–	K <sub>2</sub> CO <sub>3</sub> 5 wt % + L-Proline (0.1 M)	30	CO <sub>2</sub> /N <sub>2</sub> (21.8/78.2)	300	7.5	[104]
PTFE	–	MEA (1 M) + PG (1 M)	0.09 m s <sup>-1</sup>	CO <sub>2</sub> /N <sub>2</sub> (15/85)	0.0496 m s <sup>-1</sup>	35	[106]
Si/PVDF	126	Soybean Biosolvent (1 M)	0.5	CO <sub>2</sub> /N <sub>2</sub> (12/88)	15	0.2–1.52	[108]
PP	–	ZnO nanoparticle 0.15 wt %	50	CO <sub>2</sub> /N <sub>2</sub> (21/79)	300	9500	[112]
PP	–	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -L NP 0.025 wt %	75	CO <sub>2</sub> /N <sub>2</sub> (22/78)	100	5.5	[113]
PP	–	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> NP 0.075 wt %	75	CO <sub>2</sub> /N <sub>2</sub> (21/79)	100	5.66	[114]
PTFE	–	propylene carbonate	0.0431–0.1347 m s <sup>-1</sup>	CO <sub>2</sub> /N <sub>2</sub> (40/60)	0.3234 m s <sup>-1</sup>	14–21	[115]
PTFE	–	NaOH (2 M)	1.66–2.56	CO <sub>2</sub> /N <sub>2</sub> (20/80)	160–354	80	[116]
PP	–	de-ionized water	100–2000	pure CO <sub>2</sub>	4000	–	[118]
PTFE	–	MDEA and PZ 45 wt %	71	CO <sub>2</sub> /N <sub>2</sub> (6.4/93.6)	0.68	7.44	[119]
PTFE	–	DMEA (2 M)	0.012–0.069 m s <sup>-1</sup>	CO <sub>2</sub> partial pressure 15, 20 kPa	0.159–0.359 m s <sup>-1</sup>	8–12	[120]
ceramic	51.4	MEA 5 wt %	200–450	CO <sub>2</sub> /N <sub>2</sub> (20/80)	8100–13,500	65.22	[121]

**3.3.1.4. Operating conditions.** The overall performance of membrane contactors depends largely on operating parameters. Gas and liquid flow rates affect the mass transfer by changing the thickness of the boundary layer, while changes in feed concentration or temperature affect the driving force of mass transfer and membrane wetting. The presence of membranes leads to additional mass transfer resistance between the gas and liquid phase. To reduce the additional resistance, it is necessary to increase the mass transfer rate between the boundary layers of the solvent and gas phase.

Increasing the liquid flow rate within a certain range can improve the absorption flux and overall mass transfer coefficient. The CO<sub>2</sub> absorption flux increases with the increase of gas velocity and CO<sub>2</sub> partial pressure [53,120]. The turbulence of the gas phase is intensified, resulting in a decrease in the thickness of the gas-phase boundary layer, which is favorable for the diffusion of CO<sub>2</sub> molecules from the gas phase to the gas-liquid interface.

The increased mass transfer resistance caused by membrane wetting is another obstacle to improve CO<sub>2</sub> removal efficiency. Changing

operating conditions can also mitigate this problem. For example, during the CO<sub>2</sub> removal process, the gas diffuses on the surface of the membrane, and then forms uniform bubbles through the pores of the membrane, which dissolve, diffuse and react with the absorbent. As the gas flow increases, the pressure on the membrane shell side increases, which forces the in-tube absorbent out of the pores of the membrane. This will obviate membrane wetting and increase the CO<sub>2</sub> mass transfer rate and removal efficiency [121].

A list of CO<sub>2</sub> capture performance by membrane contactors is summarized in Table 4.

### 3.3.2. Other integrated processes

A laboratory-scale integrated cryogenic-membrane hybrid CO<sub>2</sub> separation system designed by Song et al. for conventional biogas upgrading is shown in Fig. 9 [122–124]. The separation performances of two kinds of hollow fiber membranes (polyimide and polysulfone) were tested at different operating temperatures. Membrane treatment integrated with cryogenic separation could help enhance CO<sub>2</sub> concentration initially thus reducing cryogenic separation's energy consumption via enhancing liquefaction and desublimation temperature.

## 4. Economical aspects of hybrid processes

The economic evaluation of the hybrid CO<sub>2</sub> capture processes is essential to demonstrate the feasibility for wide application and accelerate its acceptance in the industry. A techno-economic assessment was conducted to understand the capital investment and operating expenditures. As each chemical industry emits flue gases containing different CO<sub>2</sub> concentrations, it is necessary to understand capability of each CO<sub>2</sub> capture process. There is a need to understand the economics of different CO<sub>2</sub> capture technologies and the factors that affect them.

Typically, the economic efficiencies vary with different capture methods which influence the choice of capture technology. Therefore, when comparing different flue gas separation technologies, the impacts of capital expenditure (CAPEX) and operating expenditure (OPEX) on the cost of CO<sub>2</sub> capture need to be identified. Among them, CAPEX mainly includes compressor pumps, expanders and so on, and the OPEX includes the electric energy required to operate compressors and pumps. This paper focuses on the economic analysis for different CO<sub>2</sub> capture processes.

The commonly used post-combustion CO<sub>2</sub> capture technologies, absorption method, adsorption method, membrane method, cryogenic method, absorption-membrane method, adsorption-membrane method, and cryogenic-membrane method are compared in the economic analysis and hereafter abbreviated as A, AD, M, C, A-M, AD-M, and C-M, respectively. The CO<sub>2</sub> capture cost of the seven technologies under different CO<sub>2</sub> capture scenarios is depicted in Fig. 10 [36,38,41,42,44,52, 57,60,125–127]. The CO<sub>2</sub> capture cost was converted from the regeneration energy consumption. Compared to the conventional technologies (i.e., A, AD, M and C), the hybrid processes offer reduced CO<sub>2</sub> capture costs for CO<sub>2</sub> inlet concentration ranging between 10 % and 50 % by approximately 20–50 USD/tCO<sub>2</sub>. The cost evaluation shows that the electricity cost with hybrid membrane–cryogenic CO<sub>2</sub> capture is 6 % more expensive than the plant operated with absorption by MEA [128]. The CAPEX of the standalone capture process is generally lower than that of the hybrid capture process, with the CAPEX of the hybrid capture process being in the range of 35–65 USD/tCO<sub>2</sub>. This could be attributed to the increased cost of purchasing additional compressors and vacuum pumps. In addition, the concentration of CO<sub>2</sub> captured is an essential factor that influences the OPEX of the hybrid process. Gilassi et al. [27] found that the OPEX cost increases significantly as the concentration of CO<sub>2</sub> captured increases. When the intermediate CO<sub>2</sub> capture is set to 30 %, the capture cost is 36.52 USD/tCO<sub>2</sub> at the expense of an electricity usage of 124.82 MW [27].

Compared to a standalone absorption process, 4.73 % of energy savings could be identified with an in-series hybrid configuration,

whereas the parallel configuration offered 6.11 % savings in capital cost from the reduction in equipment size [129]. When the CO<sub>2</sub> recovery rate increases, the productivity of the membrane modules decreases, thus increasing the capital cost of the membrane system [130]. The National Energy Technology Laboratory (NETL) report shows an approximately linear relationship between the percentage of CO<sub>2</sub> captured and the total investment cost [131,132]. The total amine cost slowly decreases as inlet CO<sub>2</sub> concentration of the absorber tower increases while the membrane cost increases rapidly. The lowest overall mixed amine/membrane design cost was achieved at the inlet CO<sub>2</sub> concentration of 12.1 % [36]. When the membrane selectivity is high, the CO<sub>2</sub> capture cost is lower.

The influencing factors and their effects on each CO<sub>2</sub> capture technology are different. Briefly, the generic influences on total operating expenses (TOE) for hybrid CO<sub>2</sub> capture systems include the pressure drop, the CO<sub>2</sub> fraction in the feed and CO<sub>2</sub> permeability of the membrane. (1) The pressure drop in the membrane system affects plant's energy balance, as it is related to energy consumption. Moreover, the total membrane area required is also slightly affected by the pressure drop. Nevertheless, the slight reduction in the membrane area requirements for systems with increased pressure drop is at the expense of a higher energy penalty. Therefore, using membrane module designs with reduced pressure drop is preferable to increase the overall competitiveness of the hybrid system. (2) The CO<sub>2</sub> permeation rate of membrane: the total area required for the separation is related to the CO<sub>2</sub> permeation rate through the membrane, which decreases with increasing CO<sub>2</sub> permeation rate. This indicates that any improvement in the membrane's CO<sub>2</sub> permeation rate affects the CAPEX of the plant. The above two influences mainly explain the effect of membranes in the CAPEX of hybrid CO<sub>2</sub> capture technologies. (3) The fraction of CO<sub>2</sub> in the feed: the amine consumption quickly decreases as the inlet CO<sub>2</sub> concentration to the absorber tower increases. At the same time, the membrane cost increases rapidly, making the total cost slightly higher than that of the standalone absorption process [128].

To select the most suitable technology for the CO<sub>2</sub> capture process, several factors must be considered, including the range of CO<sub>2</sub> concentrations, CO<sub>2</sub> capture capacity, energy consumption and CAPEX. A comparative analysis of the standalone CO<sub>2</sub> capture processes and the hybrid processes is shown in Table 5 [36,38,41,42,44,52,57,60, 125–127]. Values 1–8 indicate the ranking of a specific parameter of the CO<sub>2</sub> capture process, with the value being the smaller the better. The lower the total score, the greater the process's potential for industrial applications. Among them, A-M and C-M technologies are relatively more advantageous when considering CO<sub>2</sub> application scenarios, CO<sub>2</sub> capture capacity, energy consumption and CAPEX conditions. Similarly, Jackson et al. also used this comparison method to select the most suitable solvent for algae cell milking [133].

## 5. Conclusions and outlook

With the incorporation of the energy efficient membrane technology, the hybrid processes have demonstrated relatively low energy consumption for CO<sub>2</sub> capture. Moreover, the combination of various CO<sub>2</sub> capture technologies as well as the different arrangements of these processes (e.g., in-series, parallel and integrated) offer great flexibility in process design and enable the treatment of a wide range of feed gas conditions. This would also allow researchers and engineers to customize and optimize the CO<sub>2</sub> capture process via process hybridization. However, on the other hand, the enhanced flexibility is a double-blade sword as it also increases the complexity of the whole process and causes difficulty for process design. Due to the high complexity of the hybrid processes, it is challenging to determine the decisive factor that affects the CO<sub>2</sub> capture performance of the process. Therefore, more studies on the investigation of hybrid CO<sub>2</sub> capture processes and process optimization are necessary. In addition, the comparisons between different hybrid processes in both technical and economic aspects are insufficient. Additional techno-economic analyses would provide more in-depth

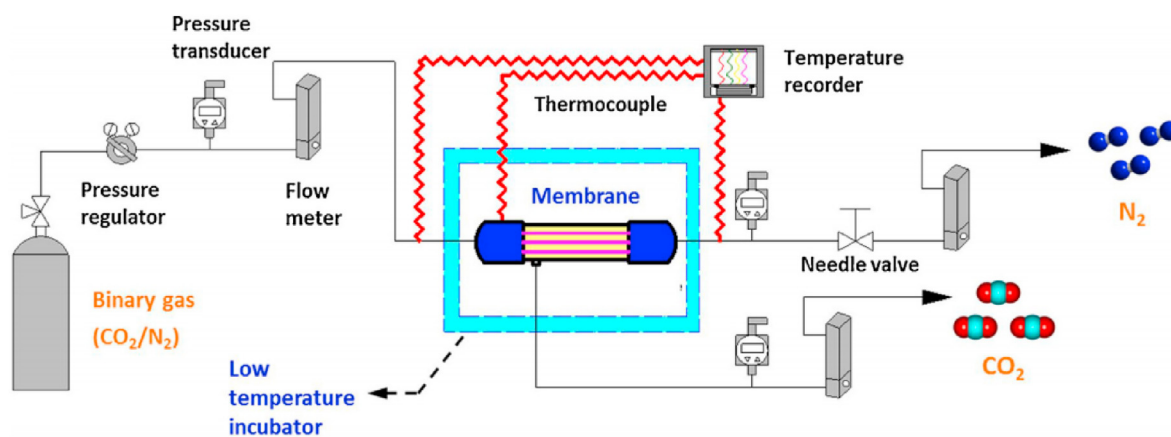


Fig. 9. Schematic of the novel cryogenic-membrane hybrid CO<sub>2</sub> separation setup. Reprinted with permission from Ref. [122]. Copyright (2018) Elsevier.

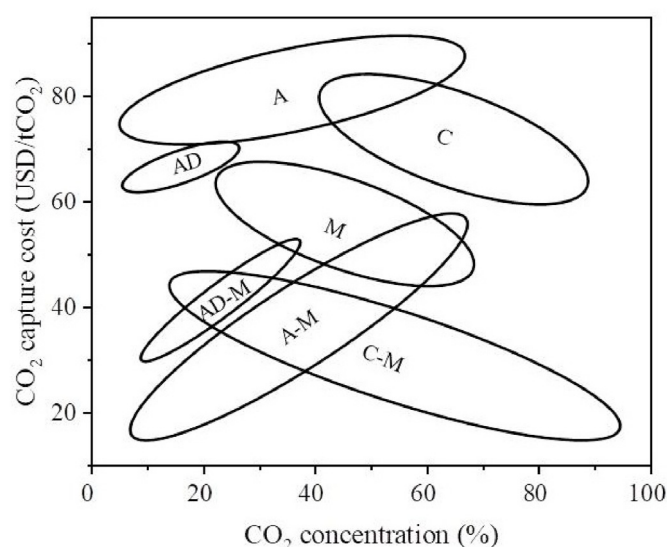


Fig. 10. The CO<sub>2</sub> capture cost of different CO<sub>2</sub> capture processes.

insights on the feasibility and design of these hybrid processes.

In comparison with the in-series or parallel hybrid processes, the membrane contactor-based CO<sub>2</sub> capture process is a more integrated process which provides a large contact area per unit volume with a low investment cost and relatively low energy consumption. To some extent, the membrane contactors overcome the drawbacks of the process complexity of the hybrid processes discussed above and demonstrate favorable economic efficiency. On the other hand, the performance of membrane contactors largely depends on the membranes. Membrane

wetting is the main obstacle for performance improvement which results in increased mass transfer resistance of the separation process. In addition, the long-term stability of the membranes in the solvent environment is another critical issue. It is still uncertain whether the membranes can survive the stringent operating conditions of flue gas treatment or maintain the separation performance in real industrial applications (e.g., power plants). More attention should be paid to the investigations of membrane performance under actual process conditions. Besides, most of the current studies on membrane contactor-based CO<sub>2</sub> capture processes focus on absorption of CO<sub>2</sub> from feed gas. Relatively few studies investigate the stripping process for CO<sub>2</sub> capture or solvent desorption which can be explored for future research. Furthermore, taking the advantage of information technology, more attention could be paid to machine learning-based material development and process optimization of CO<sub>2</sub> capture processes to accelerate its industrial application [134–136].

#### Author contributions

Meng-Chao Yu: Conceptualization, Writing - Original Draft, Visualization; Li-Ju Bai: Conceptualization, Writing - Original Draft, Visualization; Stefania Moiola: Writing - Review & Editing; Paitoon Tontiwachwuthikul: Writing - Review & Editing; Tatiana V. Plisko: Writing - Review & Editing; Alexandr V. Bilyukevich: Writing - Review & Editing; Ying-Nan Feng: Supervision, Conceptualization, Writing - Review & Editing, Project administration, Funding acquisition; Helei Liu: Supervision, Conceptualization, Writing - Review & Editing, Project administration, Funding acquisition.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Table 5

Comparison of standalone and hybrid CO<sub>2</sub> capture processes.

Process	Range of CO <sub>2</sub> concentrations	CO <sub>2</sub> capture capacity	Energy consumption	CAPEX	Total score
Absorption	3	3	7	3	16
Adsorption	7	7	5	2	21
Cryogenic	6	5	6	1	18
Membrane	4	6	4	4	18
Absorption-membrane	1	1	2	8	12
Adsorption-membrane	5	4	3	6	18
Cryogenic-membrane	2	2	1	7	12

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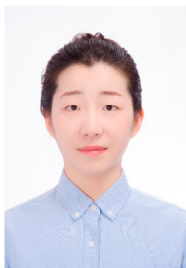
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