

CO₂ Separation Techniques

Subjects: Engineering, Chemical

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Energy plants, industries, as well as other sources of carbon dioxide (CO₂) result in global warming and affect the planet. CO₂ is separated from flue gas during combustion using a variety of advanced separation techniques. There are several techniques involved, such as absorption, adsorption, chemical looping, membrane separation, and cryogenics.

Keywords: adsorption ; absorption ; chemical looping ; CO₂ capture

1. Absorption

The ability of absorption to capture huge quantities of emissions from chemical factories and power plants has gained considerable attention in recent years. Chemical absorption is a reliable technique for CO₂ separation in coal-fired power plants because it is well-suited for existing plants with high operating costs and limited infrastructure ^[1]. The chemical absorption of CO₂ is a commercially viable technology due to its many advantages, including technical efficiency, handling capacity, and sophistication ^[2]. The potential absorbents and processes of absorption CO₂ capture are highlighted in **Table 1**.

Table 1. Summary of absorption-based carbon capture.

Type	Absorbent	Reactive Separator	Operating Conditions P, C, T, G	CO ₂ Capture (%), AC (kg/kg)	Kinetics/Mass Transfer	Ref.
Single solvent	MEA	Flow (SC)	C:8 –16; T:10–40; G:2–10	94, 0.4	C ₂ H ₄ OHNH ₂ (1)+CO ₂ (g)+H ₂ O ↔ C ₂ H ₄ OHNH+3(aq)+HCO–3(aq)	[3][4] [5]
	K ₂ CO ₃	Fixed-bed (Con-O, bench scale)	T:60 G:40 mL/min	99.4, NA	NA	[6][7]
	Ammonia	Sieve plate (CC)	C:10–14; T:25–55 °C	95–99, 1.2	2NH ₃ (g)+CO ₂ (g)↔NH ₂ COONH ₄ (s) NH ₂ COONH ₄ (s)+H ₂ O(g)↔(NH ₄) ₂ CO(s)CO ₃ (s)	[3][4]
	Piperazine	Stirred cell (SC, BS)	P:0.032 T:42 and 0.042	100, 0.32	1st order partial reaction occurs	[8]
	Ionic liquids	Double stirred cell (BS)	T:25–50; P:0.1; A:0.5–1.2	99.11 at 60 °C,	NA	[9] [10] [11] [12]
Mixed Solvents	DEA-K ₂ CO ₃	Split flow (CC, bench scale)	T:115 L:63.66 m ³ /h	99, NA	Promoter selection is very critical. It is a reversible exothermic reaction CO ₂ +K ₂ CO ₃ +H ₂ O ↔ 2KHCO ₃	[13]
	PEI-SiO ₂ Alcohol/amine/water	Packed (bench scale)	L:33.66 m ³ /h	NA, NA	qsensible=CPΔmsolutionΔmCO ₂	[14] [15]
	BDA-DEEA	Packed (CC, BS)	T:40 (absorption) T:90 (desorption) G: 24.78 m ³ /h	46 (HCL), 48 (HCC), 11(HCE) than MEA with 5 M	Carbamate and bicarbonate formations	[16]

Type	Absorbent	Reactive Separator	Operating Conditions P, C, T, G	CO ₂ Capture (%), AC (kg/kg)	Kinetics/Mass Transfer	Ref.
	AMP-PZ	Packed (pilot)	L/G:2.9; packing height=10 m	90, NA	-	[17] [18] [19] [20] [21] [22] [23] [24]

CO₂ is separated from flue gas by absorption using a liquid sorbent [25][26]. It is possible to regenerate the sorbent via a regenerative process or stripping by depressurizing and/or heating. This is the latest and most advanced method for separating CO₂ [27]. Potassium carbonate (K₂CO₃), monoethanolamine (MEA), and diethanolamine (DEA) are examples of common sorbents [28]. MEA is very reactive and absorbs more quickly, and it is quite inexpensive [27]. However, their main drawback is the substantial parasitic energy load in relation to solvent regeneration, which adversely affects the total effectiveness of systems combined with aqueous amine-based absorption processes [29]. DEA and other alkanolamines have also been employed for absorption, although they have comparable defects. Methyldiethanolamine (MDEA), a mixture of MEA and DEA, has been used with moderate success. It has higher CO₂ loading capacity, and degradation and corrosion resistance, as well as cheaper regeneration costs, but lower rates of absorption [30][31][32][33][34][35].

Veawab et al. [36] reported that MEA is the most efficient aqueous alkanolamine for CO₂ absorption, with a performance rate greater than 90%. Additionally, Aaron and Tsouris [37] reviewed various CO₂ capture technologies and determined that MEA absorption is the most viable method for CO₂ capture in CCS. Applying a solvent containing 30% MEA, a 1 t CO₂/h absorption pilot plant was designed and experimentally validated in conjunction with a coal-fired power plant's post-combustion capture technology [38]. In recent times, other adsorbents, including anion-functionalized ionic liquid and piperazine, have attracted a lot of attention [39]. Even though piperazine rapidly reacts compared to MEA, its use in CO₂ absorption is more costly. Due to its higher volatility, it is still in the experimental phase [40]. The risk of amine degradation, which could lead to equipment corrosion, solvent loss, and the formation of volatile degradation compounds, is a significant barrier to the widespread adoption of this technology for the CCS [41][42], while environmental degradation has gone unnoticed.

Furthermore, amine emissions can deteriorate into nitramines and nitrosamines, which are highly toxic to human health and the environment. The chilled ammonia process captures CO₂ using aqueous ammonium salts (including ammonium carbonate) and can regenerate the CO₂ at elevated temperatures and pressures using waste heat, thereby minimizing the downstream compression [43]. There are fewer problems with this process than those caused by amine degradation.

Water's use as a co-solvent, which has higher thermal characteristics than other co-solvents, is one of the key precursors for the high solvent regeneration energy of MEA [44]. In the context of CO₂ absorption, the predicted regeneration energy for 30 wt. % aqueous MEA showed that more than 50% of the total energy was used to heat and vaporize the water co-solvent. The remaining energy was used to reverse the chemical interaction between CO₂ and MEA at the same time [45][46]. Considering this, it was thought that either totally or partially substituting other organic diluents for water as co-solvents could potentially reduce solvent regeneration energy, since they effectively create water-free/water-lean hybrid solvents with poorer thermal properties than water [47][48][49][50][51][52][53]. Instead of vaporizing and heating the co-solvent, comparable to aqueous amines, the regeneration energy will be used more effectively to reverse acid gas chemisorption.

Additionally, hybrid water-free/water-lean solvents have been thoroughly studied in recent years, primarily for their CO₂ capture applications [53][54][55]. They provide a wide range of potentially alluring substitutes to conventional aqueous amines [29][56]. The main objective of water-lean solvents is to preserve the chemical selectivity of water-based solvents, while enabling step gains in efficiency due to the lower specific heats of organics than water [29]. However, two problematic regions refute the claim of their attractiveness. The stated performance of these solvents when scaled up from lab-scale to industrial-scale settings has not been adequately examined due to a lack of availability of a few essential properties. This is predicted given the labor-intensive nature of experimental work, which makes it impossible to expand experimental testing to the broad range of transport and thermophysical parameters needed for precise and representative performance evaluation on an industrial scale. The second issue is that, when carried out on a lab scale, the potentiality of a particular solvent is typically demonstrated using a limited set of parameters, most notably the low enthalpy of absorption and high absorption capacity [57][58][59][60]. These two characteristics are indeed of great concern for chemical absorption procedures, but they are still unsuitable for accurately gauging the potential of the tested solvents for their intended use. However, they ignore significant trade-offs between competing environmental, economic, and operational factors. The results of a straightforward assessment can help direct the development of novel generating solvents [61][62].

However, these difficulties can be overcome if the proper tools or novel process configurations are available. Due to recent developments in computational power and thermodynamic modeling tools, the first issue can be resolved by scaling the data from lab to industrial operating conditions. The most appealing models for this application are molecular equations of state (EoSs) centered on the Statistical Associating Fluid Theory (SAFT) [63][64], due to their strong theoretical background, demonstrated correctness for a range of complex systems, and predictive abilities.

The solution to the second problem, which is to demonstrate the viability of a chosen solvent typically acknowledged using a limited number of requirements, may appear relatively apparent: add more evaluation criteria to the already-existing standard key performance indicators (KPIs). Moreover, making such a preference is more difficult because early design phases may not have access to information on a particular criterion [65]. There must be a justification for why certain criteria should be included or excluded when narrowing the search space among the numerous properties that are available [66][67][68]. The effect of solvent characteristics on economic metrics such as total capital expenditures (CAPEX) and operating expenditures (OPEX) typically serves as the foundation for justification. Through the careful process modeling of hypothetical solvents, Mota-Martinez et al. [69] ranked various solvent characteristics according to how they affected the process' overall economics. Leclaire and Heldebrant [26] recently recommended the use of ideas from green engineering and chemistry to address problems with the advancement of CCUS technologies. They asserted that by applying the 12 + 12 principles of engineering and green chemistry [70], they could indirectly encourage the improvement of chemical processes' economic attractiveness and efficiency, which goes beyond their environmental motivation. Similarly to this, it may be beneficial to consider sustainability, health, and safety issues while assessing the possibility of promising solvents for the removal of acid gas [71][72]. **Figure 1** displays the schematic diagram for the absorption carbon capture process.

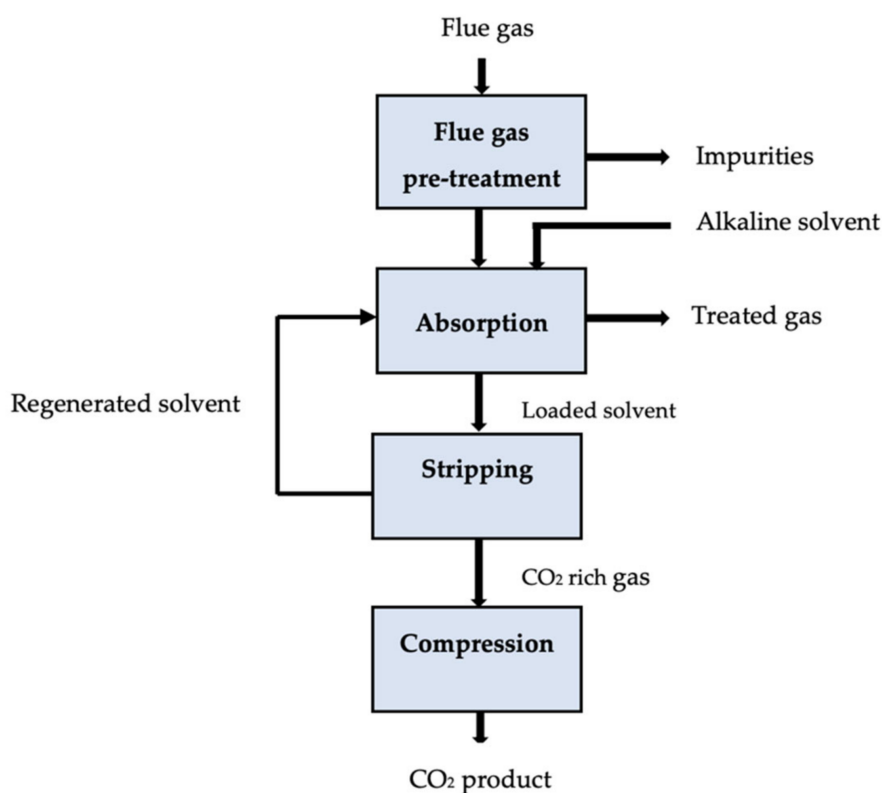


Figure 1. Schematic of an absorption carbon capture process [73].

2. Adsorption

The process of adsorption [26] involves molecules in liquids and gases adhering to solid surfaces by weak van der Waals interactions. Unlike liquid absorbent processes, solid adsorbents bind CO₂ to their surfaces during adsorption. Selection criteria for this sorbent include a large surface area, high regeneration capability, and high selectivity. Common adsorbents include activated carbon, molecular sieves, zeolites, lithium zirconate, and hydrotalcite [74]. **Table 2** highlights the potential adsorbents and adsorption parameters for CO₂ capture.

It is possible to achieve CO₂ adsorption by changing the pressure or temperature of a saturated sorbent. Pressure swing adsorption (PSA) is a commercially applied technology that recovers more than 85% of CO₂ from power plants [75][76]. A solid adsorbent selectively adsorbs CO₂ at high pressures, then the solid desorbs, releasing CO₂ for low-pressure transport (usually atmospheric pressure). The temperature swing adsorption (TSA) releases the CO₂ in the system by increasing its temperature through steam injection or hot air distribution [77]. A CO₂ purity of over 95% and recovery of

over 80% are possible when using CO₂ regeneration, although regeneration is more time-consuming than PSA [78]. It was estimated that the operating costs of a particular TSA process ranged between USD 80 and 150 per tonne of CO₂ captured [79]. Significant attention has been paid to developing CO₂ capture sorbents from agricultural and industrial wastes to lower the overall cost of CO₂ capture. An adsorption carbon capture process is shown in **Figure 2**.

Table 2. Summary of adsorption-based carbon capture.

Adsorbent	Reactive Separator	Operating Conditions P, T, C, G	CO ₂ Capture (%), Ad-C (gCO ₂ /gads)	Kinetics/Mass Transfer	Ref
TEPA-Mg-MOF-74	PBR (LS)	Regeneration temp is 250–300 °C	4–4.9 wt. %, 8.31 mmol CO ₂ /g absorbent, NA	N ₂ adsorption–desorption isotherm	[2]
ZX-APG,	PBR (3-bed, 8-step, VPSA, LS)	T:35; P: 0.007–0.008	85–95, NA, 73–82% CO ₂ purity	Langmuir adsorption isotherm is adopted	[8]
Activated carbon	PBR (1 bed, 3 step, VSA, LS)	Water vapour (H ₂ O): 4.6 mol%, Vf: 44; TDes:100T: 60, ICC:11.2, Bd:0.493, Lg:50, P:0.113, PVP = 3, Trpt:3; SA:921.7, PV:0.37, Tads:35,	69.5, NA	Dual-site Langmuir equation has been adopted	[8]
NPC10	PBR (TSA, LS)	T: 25, P: 0.1, SA: 639	NA, 0.041	Langmuir adsorption isotherm	[8]
Fly ash + PEI + PEG	PBR (LS, TSA)	St: 24 h, P: 0.11, T: 70	4.5 at 85 °C	CO ₂ +2RR'NH ↔ RR'NCOO ⁻ +RR'NH+2CO ₂ +2H ₂ O+RR'NCOO ⁻ ↔ 2HCO ⁻ 3+RR'NH+2	[8]
ZX	MBA (LS, PSA)	Bed dimensions (m): FRR: 0.5; CT: 650; AT: 950; SA: 1873.9; 2b: 0.03, Nm: 36, W: 1.5; L:1.5; Xpth: 0.012 Bd: 0.65, Cs: 1.07, Dp: 3420, ε: 0.31 ks: 0.275	80, NA, 97% purity	Extended Langmuir isotherm was used	[8]
Rayon-HCM	PBR (TSA)		97, 0.2	Langmuir adsorption isotherm adopted	[8] [8]

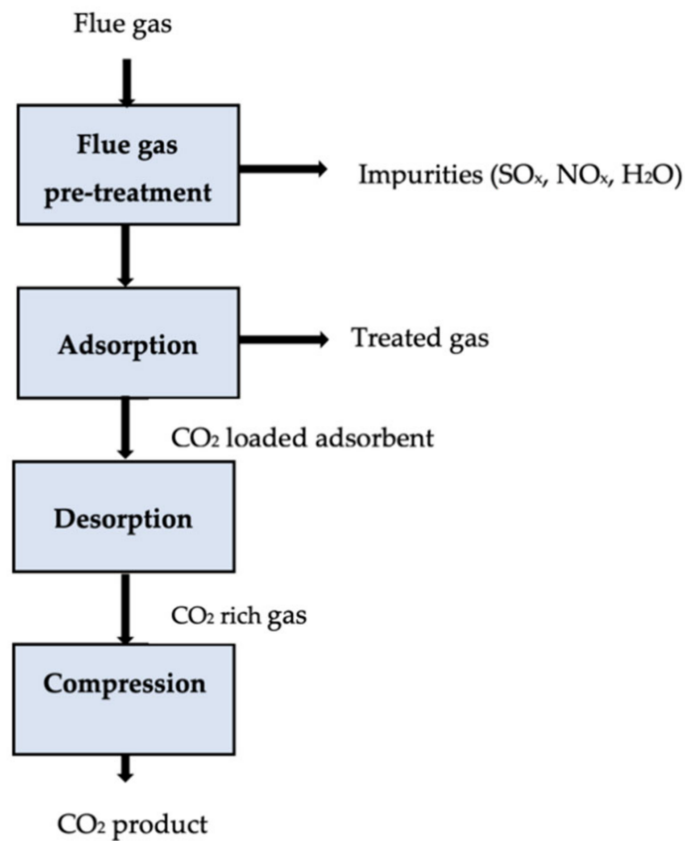


Figure 2. Schematic of an adsorption carbon capture process [73].

3. Chemical Looping Combustion

In contrast to oxy-fuel combustion, which uses pure oxygen for combustion, metal oxides are used as oxygen carriers in combustion. Metal oxides are reduced to metal during the process, while fuels are oxidized to create CO₂ and water. In a subsequent stage, the metal is oxidized and recycled. The removal of water by condensation from the process byproducts is easy, but the separation of pure CO₂ requires no energy. Numerous low-cost metal oxides, including Mn₂O₃, CuO, NiO, and Fe₂O₃, are suitable for this process. The potential sorbents and processes of chemical looping combustion are highlighted in **Table 3**.

Several researchers [87][88][89][90][91] have examined the performance efficiency of various metal oxides in this process. According to Adánez, de Diego [90], a metal oxide can be optimized by using support inert materials, but the selection of an inert material will vary depending on the characteristics of the metal oxide. Chemical looping combustion (CLC) was studied by Lyngfelt, Leckner [92] in a boiler consisting of two fluidized beds. Lyngfelt, Leckner [92] recently reviewed this technology. This process has been demonstrated to be a very promising CO₂ capture technology by both Lyngfelt, Leckner [92] and Adánez, de Diego [90]. The IGCC's CO₂ separation is based on pre-combustion, but Erlach, Schmidt [77] found chemical looping combustion to have a 2.8% higher net plant efficiency than the former method. **Figure 3** illustrates the basic CLC system.

Table 3. An overview of chemical looping combustion-based carbon capture.

Fuel Type	Operating Conditions P, T, C, G	Reactive Separator	CC (%), Purity (%)	Challenges	Kinetics/Mass Transfer	Ref.
Coal, C ₂ H ₅ OH, Isooctane, C ₃ H ₈ and CH ₄ .	T: 200–1200; molar ratios of carbon/CaSO ₄ = 0.5 and carbon/steam = 1	TGA	NA, 93 (with CaSO ₄ at 850–975 °C)	The ΔH _r is dependent on the fuel but not the amount of OC utilized. The yield depends on OC.	Combustion of iso-octane (–5101.58 kJ/mol) with Na ₂ SO ₄ and CaSO ₄ produces without SO ₂ formation between 200 °C and 344.3 °C.	[93] [94]

Fuel Type	Operating Conditions P, T, C, G	Reactive Separator	CC (%), Purity (%)	Challenges	Kinetics/Mass Transfer	Ref.
Syngas, H ₂	XOC: 80–95, HR: 90–99, T: 370–1030	2-stage PBR- CLC	100, NA	PP of O ₂ in reactors; high solid inventories.	The packed bed of OC reduces the need for highly efficient cyclone to reduce costs; boron nitride (BN) used as the dense support material due to high thermal conductivity, low thermal expansion and high thermal stability.	[95] [96] [97] [98]
Coal, kerosene, biomass	Bd: 4.750; Dp: 128 Umf: 0.0129, Φ : 0.64	IFBR	83–99.3% at 800–950 °C, NA	Scale-up, fuel conversion, agglomeration and attrition.	Δ PRC increases linearly with solid flow rate.	[99] [100] [101] [102] [103] [104] [105]
CH ₄ , coal	Iron oxide: 950 °C, FF: 1.18, CO ₂ EF: 10, DT: 5.25	CMBS or RPBR (1 MWth)	>99, >95	Reaction heat exceeds the convective heat-transfer rate to the gas flow.	The reduction kinetics and activation energy parameters are critical to find fuel conversion efficiency, temperature distribution and carbon separation efficiency.	[106] [107]
CH ₄ , syngas	T: 700–975; SITC:20–30; SFRR: 8–10 for CO SFRR:4–12 for H ₂ Fsolids:1.7–2.5	CC-MBR	>99% CH ₄ and 100% syngas conversion. >99.99% H ₂ purity.	The formation of FeO and FeAl ₂ O ₄ indicates further utilization of oxygen in iron-based OC's can be achieved. $-\phi > 1.14$.	At 900 °C, the reduction of Fe ₂ O ₃ to Fe with CO generates 37.7 kJ/mol Fe ₂ O ₃ of heat but its reduction with H ₂ gas needs 61.8 kJ/mol Fe ₂ O ₃ of heat.	[107] [108]

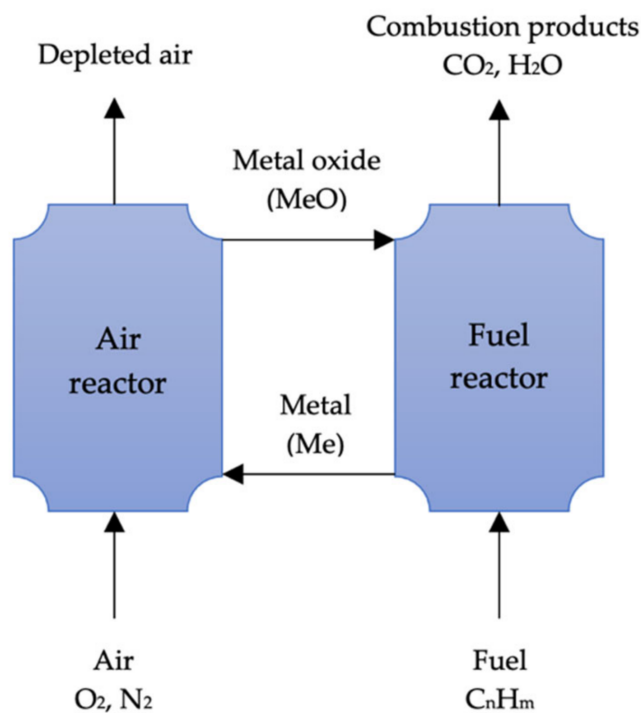


Figure 3. Schematic of chemical-looping combustion (adapted from Yang ^[109]).

4. Membrane Separation

Membrane separation uses a semipermeable membrane or barrier to physically separate CO₂ from other flue gases ^[110]. Membrane separation uses less energy than traditional solvent absorption methods, making it less expensive ^[111]. Membrane separation has successfully been used for selective gas separation in a variety of fields for the past two decades, including natural gas sweetening, air separation, hydrogen production, and biogas upgrading. Researchers are working on developing membrane-based materials to separate CO₂ released by various industries. Furthermore, this technology has produced increased efficiency in terms of both the economy and the environment ^[112]. Scientists have

developed a variety of different membranes for CO₂ separation, including inorganic membranes, polymers, carbon molecular sieve membranes (CMSMs), microporous organic polymers (MOPs), and mixed matrix membranes (MMMs) [113]. The potential sorbents and processes of membrane separation are highlighted in **Table 4**.

In addition, membrane separation technology can also separate gases in CCS processes such as pre- and post-combustion capture. It is generally considered that polymeric membranes are more flexible, durable, and efficient at capturing CO₂ from industrial processes. An upper bound relationship analysis describes how selectivity and permeability are related to CO₂ capture by polymeric membranes [114]. To improve results, glassy and rubbery materials with varying separation principles based on their size and diffusion ability can be used to synthesize polymeric membranes. The condensability and differences in kinetic properties of gas molecules are responsible for gas separation by glassy and rubbery polymers [115]. Considering how difficult it is to examine operating conditions for rapid performance, membranes applied in gas separation systems are typically modeled to determine their working capacity [116]. For optimal results in industrial settings, membrane performance must not be interfered with by flue gas impurities [113]. Researchers were able to separate CO₂ from other gases with an efficiency of 82–88% [117][118]. In fact, despite membrane materials having poor permeability and selectivity [119], it is also problematic to use this extraction method in flue gas with low pressure and CO₂ concentration in flue gas conditions [120]. A membrane carbon capture process is displayed in **Figure 4**.

Table 4. Summary of membrane-based carbon capture.

Membrane	Reactive Separator	Operating Parameters	Challenges	Kinetics/Mass Transfer	Ref.
Dense membranes	Hollow fiber and flat-sheet	S-P, T, P, L _a , pressure ratio of the permeate side to the feed side, pore size and porosity	Lower selectivity at higher permeability	Solution–diffusion; among the mechanisms are Knudsen diffusion and the molecular sieve effect	[121]
Micro-porous Membranes	Hollow fiber and flat-sheet	P, T, pore size and ϵ of the membrane-membrane wettability	Wetting of the membrane	Reaction kinetics depend on solvent	[121]
		Gas flow area	There are other compounds present in the gas stream	Even at high pressures, K _o is controlled by the resistance of the liquid film	[122]
		Liquid flow area	Solvent volatility and limited long-term stability	Pore diffusion depends on membrane support	[123]
Liquid in the membrane pores	Flat-sheet only	Ga, La, VVIS, P, T	Solvent “wash-out” causes the membrane’s stability to decrease	The overall mass transfer coefficient	[124]

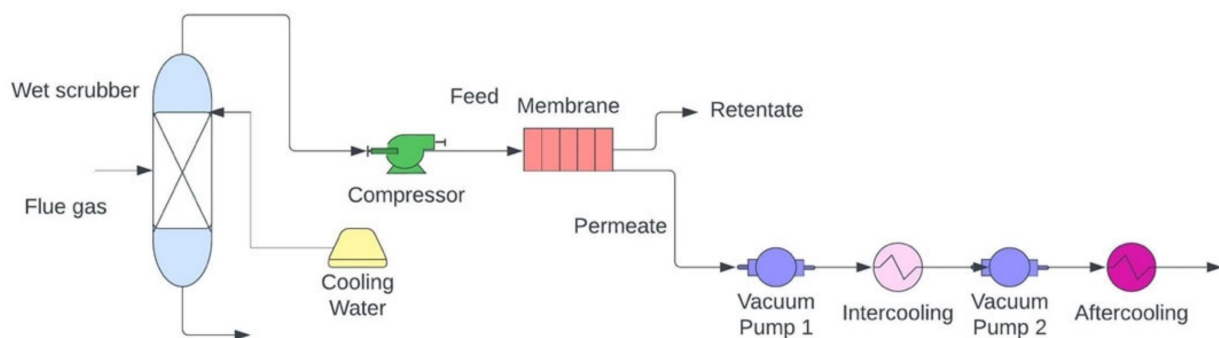


Figure 4. Schematic of membrane carbon capture process (adapted from Wang [125]).

5. Cryogenic Distillation

This process separates CO₂ from gas mixtures by focusing on their boiling points at temperatures ranging from 100 to 135 °C [26][126]. In the presence of high pressures (100–200 atm), solidified CO₂ provides two significant benefits: a lack of solvents and liquefied CO₂ for more convenient transport and injection [127]. It does, however, have some drawbacks that need to be investigated further, as do other processes. When cold and pressurized nitrogen is used as a refrigerant, ice formation compromises equipment safety, causing pressure fluctuations and pipe blockages, as well as increasing the consumption of energy [127][128]. This enhanced CO₂ separation can nullify the need for refrigerant preparation and storage [128][129]. However, CO₂ is separated using cryogenic distillation coupled with biogas upgrading. A comparison of different separation methods for CO₂ capture is shown in **Table 5**.

Table 5. Current status of different separation technologies for CO₂ capture ^{[37][75][76][77][78][125][127][130][131][132][133][134][135][136][137][138][139][140][141][142][143][144][145][146][147][148][149][150][151][152][153][154]}.

Parameter	Chemical Absorption	Physical Absorption	Adsorption	Chemical-Looping Combustion	Membrane Separation	Cryogenic
Separation technique	Amine, chilled ammonia, and amino acid salt solvent.	Rectisol, Selexol, etc. Mostly integrated gasification combined cycle.	Pressure swing adsorption and pressure-temperature swing adsorption.	FeO, CuO, MnO, and NiO	Polymeric, inorganic and mixed membranes.	Cryogenic distillation.
Pros	High reactivity, low cost of the solvent, and low molecular weight result in a high mass-based absorption capacity, and moderate thermal stability and thermal degradation rate.	Highly recommended for separating CO ₂ during pre-combustion processes that operate at elevated CO ₂ partial pressures. Captures CO ₂ selectively from a gas stream without a chemical reaction	Recycling is possible since it is a reversible process. It is possible to achieve high adsorption efficiency (485%). Low waste generation.	Very high CO ₂ concentration. Low-cost oxygen carrier materials. Truly and directly reduces the atmospheric CO ₂ concentration. Viable alternative for CO ₂ capture from mobile and decentralized sources.	No regeneration processes. Less solid waste produced. Less chemical consumption. High efficiency (>95% for single metal).	High capture efficiency (up to 99.9%). Mature technology. For many years, CO ₂ has been recovered in the industry by this method.
Cons	Relatively high maintenance cost.	High energy is required to compress feed gas to a high pressure. Low CO ₂ solubility. Less efficient absorption process. Large equipment sizing.	Requires adsorbent capable of operating at elevated temperatures. The significant amount of energy needed for CO ₂ desorption is high.	Currently, the process is under development, and large-scale operations have not yet been carried out.	Fouling and low fluxes are examples of operational issues. High running costs. Removal (%) decreases with the presence of other metals.	High energy requirement due to refrigeration. High capital expenditure. Need for removal of water, NO _x , SO _x , and other trace components to avoid the freezing and eventual blockage of process equipment. The procedure consumes a significant amount of energy.
CO ₂ concentration (vol.%)	<30.4	>59.3	28–34	3–8	11.8	<90
CO ₂ capture efficiency (%)	95	>90	<85	52–60	90	99.9
CO ₂ capture cost (USD/tonne CO ₂)	26.2	25.1	6.94	16–26	3–10	32.7
CO ₂ purity (%)	99	<99	99.98	>96	95	99.95

Parameter	Chemical Absorption	Physical Absorption	Adsorption	Chemical-Looping Combustion	Membrane Separation	Cryogenic
Status of research and development	SaskPower, Saskatchewan, Canada (Boundary Dam Carbon Capture Project) TransAlta Corporation, Alberta Canada (Project Pioneer Keephills 3 Power Plant) American Electric Power, OH, USA (Mountaineer Power Plant)	Summit Power Group, LLC, Seattle, USA (Texas Clean Energy Project) Don valley, Yorkshire, UK (Don Valley Power Project) Nuon Power, Buggenum, The Netherlands (Integrated gasification combined cycle plant)	Under developmental stage.	Less large-scale demonstration plants.	Schwarze Pumpe power station, Spremberg, Germany (Oxy-fuel technology) CS Energy: Callide Power Plant A, Queensland, Australia (Callide Oxy-fuel Project) OxyCoal, UK (Oxy-fuel technology)	Air Products and Chemicals, Inc., Pennsylvania USA

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