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CO_2 Post-Combustion Capture: A Critical Review of Current Technologies and Future Directions

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ABSTRACT

As the global imperative to CO_2 emissions intensifies, Carbon Capture and Storage (CCS) technologies, particularly CO_2 post-combustion capture, emerge as essential components in the transition towards a sustainable, low-carbon future. This review offers an exhaustive examination of the current state-of-the-art and recent advancements in CO_2 post-combustion capture techniques, including absorption, adsorption, and membrane technologies. It presents an in-depth analysis aimed at understanding their relative efficiencies, challenges, and potentials in contributing to global CO_2 reduction goals. The manuscript methodically evaluates each capture technique, starting with solvent-based absorption, which benefits from extensive research and industrial application but faces challenges such as high energy demand for solvent regeneration and environmental concerns. It then transitions to adsorption-based methods, highlighting their advantages in terms of energy efficiency, alongside the development of novel adsorbents that offer improved capacity and selectivity. Membrane technologies are also explored, with a focus on their potential for low-energy separation and the ongoing innovations aimed at enhancing their permeability and selectivity for CO_2. Finally, cryogenic separation is discussed for its ability to achieve high-purity CO_2 capture through low-temperature processes, addressing specific operational challenges. The review promotes a multidisciplinary approach that combines advancements in chemical engineering, materials science, and environmental policy to accelerate the deployment of efficient, cost-effective CO_2 post-combustion capture technologies. Specifically, it emphasizes the need for continued research/development and enhanced collaboration/innovation within the scientific community and industry players to address the scalability, economic viability, and environmental impact of these technologies, ensuring they can effectively contribute to the decarbonization of the energy sector.

Keywords: Carbon Capture and Storage (CCS); Post-combustion CO_2 capture; CO_2 separation techniques; Absorption; Adsorption; Membranes.

1. Introduction

As the global community confronts the escalating crisis of climate change, the surge in atmospheric CO_2 levels from approximately 340 ppm in 1980 to 421 ppm in 2022 has become a focal point of concern [1]. This dramatic increase is largely attributed to human activities, notably the burning of fossil fuels like coal, oil, and natural gas, particularly in the energy sector[2]. This sector is poised to remain a leading contributor to greenhouse gas emissions, especially CO_2, for the foreseeable future. The International Energy Agency 2021 report revealed a sharp 6% rise in global energy-related CO_2 emissions, reaching 36.3 gigatonnes, a rebound from the COVID-19 pandemic's dampening effect on economic and social activities [3].

In response to this urgent environmental challenge, various international and regional initiatives have been put forth. The European Union, for instance, has committed to a 40% reduction in CO_2 emissions by 2030 compared to 1990 levels, with a long-term goal of achieving a climate-neutral economy by 2050 [3]. The UK and India have set similar ambitious targets for reducing their CO_2 emissions. However, despite these efforts, the current trajectory of CO_2 emissions and associated environmental policies indicate that meeting these goals will require a significant shift in how we manage and mitigate CO_2 emissions [3].

Central to this shift is the development and implementation of Carbon Capture and Storage (CCS) technologies, including Carbon Capture Utilization and Sequestration (CCUS) [4]. These technologies encompass a range of methods for capturing CO_2 emissions at their source, such as power plants and industrial processes, followed by transportation and storage in geological formations [5,6]. While CCS presents a viable solution for bridging the gap between current fossil fuel reliance and a cleaner energy future, it is not without challenges. The most significant of these is the high cost and energy requirements and implementation risks of current CCS methods, particularly in the CO_2 capture phase

[3]. Consequently, the industries that are most responsible for anthropogenic CO_2 emissions have not widely embraced the CCS technologies.

2. CO_2 Capture Techniques

Three principal approaches are currently in use to capture CO_2 at large scales [2]:

- Pre-combustion – Removal of CO_2 before combustion of the fuel source (Coal/Gas), i.e. it involves the conversion of fossil fuels into a synthesis gas, which subsequently undergoes a separation process to extract CO_2 from hydrogen, with hydrogen serving as a clean and versatile fuel source [7].
- Post-combustion – Removal of CO_2 from the flue gas after the fuel source is combusted, i.e. CO_2 is extracted from the exhaust gases emanating from power plants and industrial facilities [7].
- Oxyfuel combustion – Combusting the fuel source with nearly pure oxygen instead of air, i.e. represents a distinct approach, characterized by the combustion of fuel in pure oxygen. This results in a flue gas rich in CO_2 and devoid of nitrogen, simplifying the subsequent sequestration process [7].

Each of these capture methods operates under distinct operational conditions, exhibiting unique advantages and challenges. The pursuit of efficient and cost-effective CCS implementation necessitates continuous innovation, technological advancements, and a holistic understanding of these distinct approaches.

Each of these three strategies have distinct advantages, limitations, and contexts in which they are most effectively applied. These methods are depicted in Fig. 1, providing a visual framework for understanding their categorization under the broad spectrum of CO_2 capture technologies. All of them will be analyzed in depth in the forthcoming sections, providing a comprehensive overview of their operational principles, efficiency, cost-effectiveness, and integration into existing systems.

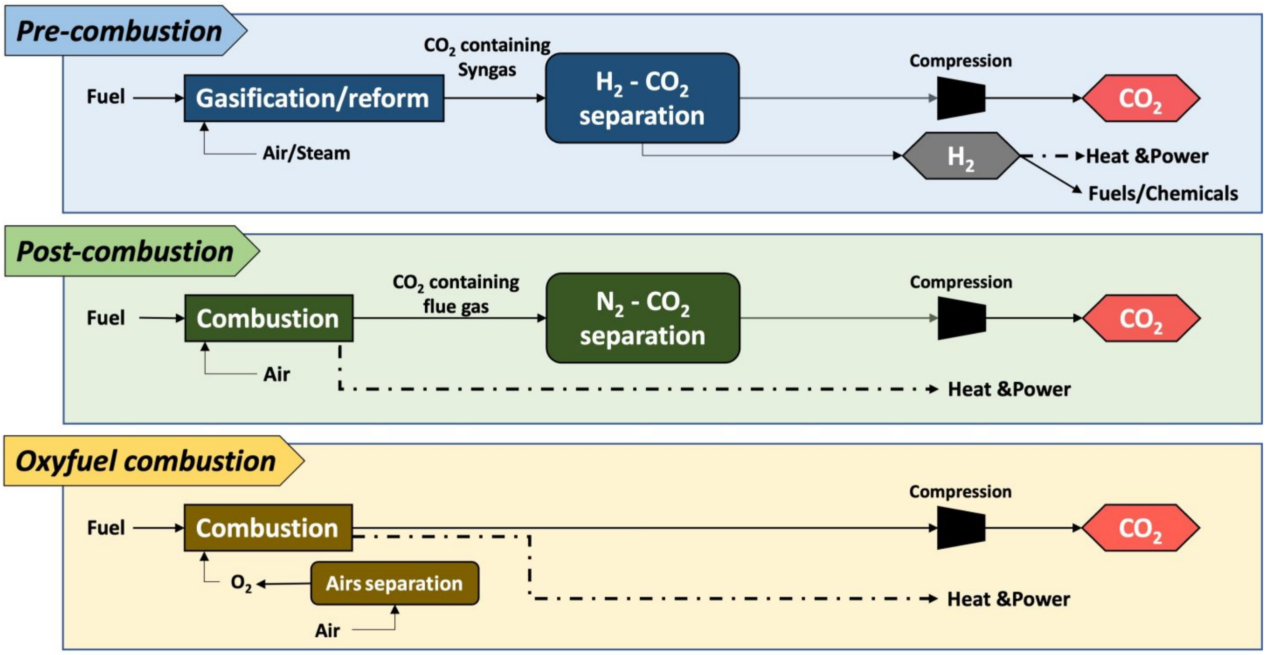

Fig. 1. CO_2 capture technological solutions: (a) pre-combustion; (b) post-combustion; (c) oxyfuel combustion.

Prior to exploring the different CO_2 capture options and their associated technologies, it is imperative to define and comprehend the maturity levels and pertinent terms. This study aims to shed light on the current developmental stage of the technologies being examined. Technology Readiness Level (TRL) is a systematic framework to assess the maturity of complex technologies, progressing sequentially from basic principles observation to successful operational deployment [2] and, thus, serving as a gauge to measure the risk associated with implementing a technology in a specific context. As defined by the US Government Accountability Office (US GAO), the TRL scale ranges from 1 to 9, with each level representing a different stage in the development process (Table 1) [8]. This 9-level TRL scale offers a detailed approach for precise technology assessment, breaking down the development process into specific stages that allow for careful monitoring and evaluation of progress. This granularity is especially useful for complex projects where understanding the exact maturity of each component is crucial. Commercial products typically do not incorporate new technologies unless they achieve a TRL of 8 or 9, indicating maturity and practical applicability [8]. The US GAO reports that the introduction of unproven technologies in product commercialization often leads to delays and budget overruns, while projects utilizing more mature technologies tend to fare better. Although the 9-level TRL system is versatile enough to apply to any technology, including CO_2 capture, a simplified 5-stage (also detailed in Table 1) process has been proposed for describing/categorizing the development and maturity levels in technological plants in broader terms [8]. In particular, the 5-level TRL scale provides a simplified, high-level overview that condenses the stages into broader categories. This approach is more accessible and easier to communicate, making it ideal for strategic planning and high-level decision-making where detailed granularity is less critical.

2.1. Pre-combustion capture

Pre-combustion CO_2 capture technology is a proactive approach to reduce CO_2 emissions by treating fuel before it is burned for power generation. It is typically utilized in power plants that process natural gas or coal [2].

This process begins with the gasification/reforming of the fuel (coal or natural gas) in a low-oxygen environment, producing a syngas predominantly composed of carbon monoxide (CO) and hydrogen (H_2) [9]:

$$Fuel + gasyfing\ (reforming)\ agent \rightarrow CO + H_2 \tag{1}$$

The syngas is then subject to a water-gas shift (WGS) reaction, where steam facilitates the conversion of CO to CO_2, significantly increasing the hydrogen content [10]:

$$CO + H_2O \rightarrow H_2 + CO_2 \tag{2}$$

The advantage of pre-combustion technology lies in the fact that the CO_2 concentration in the syngas stream is considerably higher than that found in post-combustion flue gases, often reaching around 15 - 60% [11,12]. This high concentration, coupled with the high pressure of the syngas stream (2 – 7 MPa), makes CO_2 separation more straightforward and energy-efficient [2,13]. Physical solvents, such as Rectisol and Selexol, are employed due to their efficiency at these higher pressures and CO_2 concentrations, leading to a less energy-intensive separation process compared to the use of chemical absorbents like amine-based solvents required in post-combustion scenarios [14]. Following the separation process, CO_2 undergoes a series of conditioning steps, including the removal of condensates and moisture, the elimination of solid particles, and the adjustment of the gas temperature through cooling or heating. This is done alongside the removal of any unwanted gaseous impurities, after which the CO_2 is compressed in preparation for its final storage. Concurrently, the residual gas, enriched with hydrogen, is utilized as a fuel source in boilers or gas turbines within an Integrated Gasification Combined Cycle (IGCC) system, which is instrumental in electricity generation. Although this pre-combustion method can potentially achieve high carbon dioxide capture efficiencies, up to 80% before combustion, it is often associated with substantial capital and operational expenses, which diminishes the overall economic viability and attractiveness of this CO_2 capture method [3,15]. Specifically, the primary energy cost linked to pre-combustion technology arises from the water-gas shift reaction, i.e. from the need to provide heat for the WGS reaction [16,17].

Information regarding the pre-combustion capture method is summarized in Table 2. The primary areas for advancement and hurdles in this technology revolve around enhancing the gasification phase, the CO_2 separation process (absorption), and implementing novel approaches for syngas cleaning strategies. These efforts aim to reduce energy consumption and overall costs, making pre-combustion a more economically viable CO_2 capture option in the fight against climate change.

Table 3 presents a summary of both commercial and smaller-scale plants around the globe that employ the pre-combustion method for CO_2 capture. It includes commercial operations with CO_2 capturing capabilities exceeding 1000 MT day^{-1} and full-scale demonstration and pilot plants with CO_2 capturing capacities ranging from 100 to 1000 MT day^{-1} [2,18–22].

Table 1. TRL definition.

TRL	Technology Phases	Simplified 5-stage definition	Lifecycle Phase
9	Proven actual system through successful mission operations	9: Commercial Process - Finalization	Facility decommissioning underway or completed
8	System completion and qualification via tests and demonstration	8: Full-Scale Demonstration Plant - Active Service	Facilities operational after commissioning
7	Operational environment demonstration of system prototype	6-7: Pilot Plant - Construction Phase	Investment finalized and construction of facilities ongoing
6	Prototype model or subsystem demonstration in a relevant setting		
5	Validation of components or breadboards within a relevant environment	3-5: Laboratory or Bench-Scale - Developmental Stage	Technology, location, and design under demonstration for viability
4	Laboratory environment validation of components or breadboards		
3	Conceptual proof of function and characteristic through analysis/experimentation		
2	Formulation of technology concept or application	1-2: Conceptual Design - Preliminary Evaluation	Ongoing assessment of various technologies and configurations
1	Observation and reporting of basic principles		

Table 2. Pros and cons of pre-combustion capture.

Advantages/Opportunities	Disadvantages/Challenges
• Extensively used in the industry for over 95 years. • Low emission of CO_2 (92–93% recovery). • Enhanced energy efficiency in separation and compression of CO_2, resulting from decreased gas volume and elevated pressure along with higher CO_2 concentrations. • Lower regeneration energy due to the use of physical solvents for CO_2 separation (mature physical absorption technique) • Flexibility to switch between H_2 production and electricity generation. • Uses less water than post-combustion capture. • Synthesis gas can be used as an alternative turbine fuel. • Hydrogen can be utilized in fuel cells, transportation, and chemical synthesis. • Readily available at the commercial level for the removal of acidic gases.	○ Expensive in terms of capital costs and equipment. ○ Significant energy loss compared to post-combustion capture. ○ Improvements needed for energy recovery efficiency. ○ Requires a chemical plant in front of the turbine. ○ Retrofitting increases cost and complexity, hindering commercialization. ○ High-pressure operation can be challenging. ○ Complex chemical processes may lead to plant shutdowns. ○ Requires cleaned gas stream and costly scrubbing for NOx control. ○ Gasification stage and associated heat transfer need improvement. ○ Cooling of flue gas to CO_2 capture is necessary. ○ Extensive supporting systems required, like air separation units. ○ Efficiency loss in the water-gas shift section. ○ Absorption method requires lower regeneration temperatures; ionic liquids are being explored.

Table 3. Industrial plants using pre-combustion CO_2 capture [2,18–22].

	Country	Separation technology	Plant size MW	Capacity MT day^{-1}	Company	Starting date
TRL 8-9						
	Saudi Arabia	Amines	-	2000	Saudi Aramco	2018
	Canada	Rectisol	240000	41100	North West Redwater	2011
	USA	-	-	23000	Occidental Petroleum	2010
	Australia	-	-	7000	Western Australian Department of Mines and Petroleum	2009
	USA	Amines + membranes	-	2,000	Anadarko Petroleum Corporation	2006
	China	-	39000	9,600	Shenhua Coal Trading	2000
	Canada	Rectisol	-	3,200	Cenovus Energy	2000
	USA	Gas separation	-	3,600	MCN Energy Group	1998
TRL 6-7						
	China	Rectisol	1205	140	Shaanxi Yanchang Petroleum Group	2017
	Spain	Pressurized entrained-flow (Prenflo)	335	100	ELCOGAS S.A.	2011
	Australia	Amine + ammonia + membranes	1.1	740	CO2CRC	2009
	Brazil	MTR membranes	-	300	Petrobras	2009
	USA	MDEA gas separation	50	200	FITampa Electric Power Company	2008

2.2. Post-combustion capture

Post-combustion CO_2 capture technology is acknowledged as a well-established method, primarily involving the direct extraction of carbon dioxide from flue gases emanating from the combustion chambers of thermal power plants [2]. It is noteworthy that these CO_2 separation methods do not interfere with other processes occurring during fuel combustion.

The capture unit is situated downstream of purification systems, which is necessary to isolate of CO_2 from other constituents in the flue gas (SO_2, NO_x, particulate, etc.) [23]; i.e. the flue gas undergoes several pretreatment steps like denitrification, desulfurization, and dust removal before CO_2 capture [24]. Additionally, the flue gases released from post-combustion processes, typically at relatively high temperatures (50 – 130 ˚C) and near atmospheric pressure [25,26], pose a significant design challenge due to their low CO_2 concentration, which ranges from 7 - 15% in coal-fired power plants to around 3 - 5% in gas-fired power plants [2,27]. As a consequence of this limited driving force for CO_2 capture, the implementation of this technology necessitates large-scale processing equipment to handle voluminous amounts of flue gases, thereby escalating capital costs [2].

After the preliminary purification step, the flue to be sent to CO_2 separation unit is primarily constituted by are CO_2, H_2O, and N_2 [3]. In this context, the selection of an appropriate separation technique largely depends on various physicochemical properties of the exhaust gases and process conditions, including temperature, pressure, CO_2 concentration, and the volume of the gas stream. Currently, the most common and commercially viable post-combustion technique is absorption using monoethanolamine (MEA) [28]. However, the need for solvent regeneration and the resulting solvent loss contribute to an increased energy penalty, highlighting the necessity for enhanced solvents for efficient separation. Additionally, adsorption methods, including temperature swing or pressure swing adsorption processes, as well as calcium looping combustion (CLC), are also employed in post-combustion capture [3]. Membrane technology is another viable option, offering advantages like lower capital costs and adaptability for scaling up with existing power plants to minimize energy requirements, carbon footprint, and operational costs [29]. Alternatively, cryogenic technology, which involves compressing and cooling flue gas to separate and condense CO_2 into a high-purity liquid or solid phase, is also an effective method utilized in this context. In this framework, the energy demand for CO_2 capture and separation is considerable and must be factored into the potential costs for electricity production [3,30].

Information regarding the post-combustion capture method is summarized in Table 4. Unlike pre-combustion, post-combustion does not necessitate the use of costly equipment and systems, like syngas separation units and hydrogen turbines. Additionally, post-combustion technology allows for greater adaptability in CO_2 capture, enabling operations without altering the combustion cycle. A further benefit of post-combustion is the ability of the power plant to continue functioning even when the CO_2 capture system is offline.

Table 5 provides an overview of operational facilities employing post-combustion technology for CO_2 capture. The majority of these plants, particularly those with capture capacities exceeding 1000 MT day^{-1}, predominantly utilize amine-based solvents. CO_2 Solutions Inc., located in QC, Canada, distinguishes itself by employing an enzyme-promoted aqueous alkali salt solution, noted for its high efficiency and minimal environmental impact [2,18–22]. While amine-based solvents remain the preferred choice for CO_2 capture in both demonstration and pilot scale plants, alternatives such as ammonia, amino-acid salt-based solvents, membrane systems, solid sorbents, and algal systems are also being explored in various facilities [2,18–22].

Table 4. Pros and cons of post-combustion capture.

Advantages/Opportunities	**Disadvantages/Challenges**
• Suitable for integration with both existing and new coal-fired energy plants. • Ongoing research aims to improve the effectiveness of sorbents and capture mechanisms. • Modifying current designs of power plants for retrofitting is a practical approach. • Enhanced efficiency in converting heat to electricity compared to the pre-combustion approach. • Carbon dioxide emissions are substantially reduced (with an 80–95% recovery rate by adsorption methods and 90–98% by absorption). • Additionally removes nitrogen and sulfur oxides from emissions. • Incorporation of combined processes, such as membrane-pressure swing adsorption, to enhance the capture of CO_2. • Future advancements in coal pulverization technology are expected to reduce carbon emissions and increase operational output.	○ Low CO_2 concentration at standard atmospheric conditions necessitates larger equipment and incurs increased costs. ○ Challenging design of systems capable of handling low CO_2 partial pressure and the high temperatures in flue gases. ○ Low CO_2 concentrations in exhaust gases considerably raise the cost of electricity generation, especially for new facilities or when retrofitting. ○ Sensitivity to impurities such as NO_x and SO_x in the absorption process. ○ Steam removal processes can decrease the performance and capacity of low-pressure turbines. ○ Intensive capture processes require robust system performance, greater circulation capacity, and increased water usage.

Table 5. Industrial plants using post-combustion CO_2 capture [2,18–22].

	Country	Separation technology	Plant size MW	Capacity MT day^{-1}	Company	Starting date
TRL 8-9						
	China	Amines	600	2750	China National Petroleum Company	2018
	South Korea	Amines	300	2740	Korean Electric Power Corporation	2018
	Canada	Aqueous alkali salts with enzymes	-	30	CO_2 Solutions Inc.	2015
	Canada	Amines	115	2740	SaskPower	2014
	USA/Canada	Solvents	240	3830	NRG Energy Inc. / Petra Nova	2010
	India	Amines	-	1940	Indian Farmers Fertiliser Cooperative Limited	2006
	USA	Amines	50	1200	Carbon Dioxide Technology Corporation	1982
TRL 6-7						
	France	Multiple	23	164	Veolia Environmen	2017
	UK	Amines	3	50	RWE nPower	2011
	UK	Amines	5	100	Scottish and Southern Energ	2011
	Norway	Ammonia	630	275	Statoil	2010
	Germany	Amines	7	90	E.ON	2010
	Italy	Amines	660	22	Enel	2010
	Australia	Ammonia	450	4.11	CSIRO	2009
	USA	Ethanol-amino solvent	180	90	AES Corporation	2000

2.3. Oxy-fuel combustion

The oxy-fuel combustion technology represents a departure from conventional methods, as it involves combustion in a mixture of high-purity oxygen and recirculated exhaust gas, rather than air [31,32]. Consequently, the first goal is to minimize N_2 entering the combustion chamber by separating oxygen (to achieve purity above 95%) and N_2 from the air supplied to the boiler [33]. This objective is essential due to the high N_2 content in atmospheric air, which ranges around 79%, and leading to a CO_2 content in boiler flue gases varying between 3% and 15%, depending on the fuel type [34]. Clearly, this would complicate the separation of CO_2 from the other exhaust gas components. Additionally, part of the flue gas is recirculated to lower combustion temperatures, as combustion in pure oxygen is practically unfeasible due to the inability of currently available materials to withstand the extremely high temperatures generated during combustion with pure O_2 (3500 °C) [35]. In this context, studies suggests that the optimal composition of the boiler feed stream should contain 30–35% O_2 and 65–70% CO_2 [36].

The significant interest in oxy-fuel combustion within power systems stems from its key advantages (Table 6): reduced flue gas and nitrogen emissions (NO_x elimination through pure oxygen utilization), improved boiler energy conversion efficiency, increased CO_2 concentration with the potential for direct CO_2 sequestration, and utilization of smaller combustors due to reduced gas volume [37]. More specifically, the resulting flue gas mixture can contain up to 98% CO_2, depending on factors such as fuel type, direct transport and storage of CO_2 with almost zero emissions become feasible [3]. However, the widespread implementation of oxy-fuel combustion faces challenges due to very significant drawbacks (Table 6): the need for a more complex and controlled process, reliance on a significant recycling stream from the flue gas to the combustor to prevent excessively high-temperature combustion, and most importantly the energy-intensive separation unit required for N_2 removal from air to obtain high-purity O_2 [2,3,24,38,39]. In this framework, cryogenic distillation remains the primary method for generating large volumes of high-purity O_2, emphasizing the need for research into new air separation approaches, such as ion-transport and oxygen-transport membranes [40].

Although relatively new, oxy-fuel combustion technology has been explored in various demonstration projects and pilot-scale facilities globally, as summarized in Table 7. Two plants, one in Spain and the other in South Korea, currently boast a capture capacity exceeding 1000 MT day^{-1} [2,18,20–22].

Table 6. Pros and cons of oxy-fuel combustion.

Advantages/Opportunities	Disadvantages/Challenges
• Applicable to current and future power generation infrastructure. • High efficiency in CO_2 recovery, typically in the 90-98% range. • Flexibility in fuel usage, including renewable and waste materials, contributing to carbon neutrality initiatives. • Oxy-fuel combustion alterations can mitigate economic and efficiency impacts. • Enables high-efficiency steam cycles. • Minimal NOx output due to N2 absence in the combustion process (NOx emissions reduced by 60-70% compared to conventional combustion methods). • Cost advantages over other capture methods due to decreased volume of exhaust gases and higher CO_2 concentration (70-95%vol). • Integration simplicity into existing plants due to no requirement for on-site chemical processing. • Smaller process size due to reduced volume of gas production derived by N2 absence.	○ Complexity of combustion with pure oxygen, leading to high temperatures. ○ Substantial oxygen requirements increase capital and operational costs. ○ Significant energy input needed for air separation impacting plant efficiency. ○ Need for development of non-cryogenic air separation techniques. ○ Increased costs due to air separation and flue gas recirculation. ○ Demands for materials resistant to high temperatures of combustion gases. ○ Necessity to validate technology for large-scale operations. ○ Elevated risk of CO_2 leakage.

Table 7. Industrial plants using oxy-fuel combustion [2,18,20–22].

TRL	Country	Plant size MW	Capacity MT day^{-1}	Company
8-9	South Korea	300	3290	Korean Electric Power Corporation
5-6	Spain	323	275	Endesa
2-3	South Korea	-	2740	Korea-CCS 2
2-3	China	-	5480	Shanxi International Energy Group CCUS

2.4. Comparison of CO_2 capture techniques

CO_2 capture from power plant exhausts involves various techniques, including post-combustion capture, pre-combustion capture, and oxy-fuel combustion, as depicted in Fig. 1. Each option offers unique advantages and challenges, impacting factors such as CO_2 concentration, equipment size, and emissions of toxic gas species, as also summarized in Table 8:

- Pre-combustion capture
    - Involves partial oxidation of fossil fuel to produce syngas (CO and H_2O).
    - Syngas is further converted into CO_2 and H_2.
    - CO_2 is captured before combustion.
    - H_2 can be utilized as fuel.
- Post-combustion capture
    - Involves direct removal of carbon dioxide from the flue gas emitted by the combustion chamber of a thermal power plant.
    - Typically employs chemical solvents or adsorbents to capture CO_2.
- Oxy-fuel combustion
    - Involves burning fuel in oxygen instead of air.
    - Results in flue gas predominantly composed of CO_2 and impurities such as SO_x.
    - Can achieve higher CO_2 concentrations compared to pre- and post-combustion capture.
    - In-furnace desulfurization can enable zero gas emissions.
    - Smaller equipment sizes.

Upon reviewing the different CO_2 capture options, their advantages, disadvantages, and techno-economic aspects are reported in Table 9. Although pre-combustion and oxy-fuel capture methods offer distinct advantages, they are unlikely to replace post-combustion capture on a global scale, given that post-combustion CO_2 abatement represents a straightforward approach and serves as the foundation of the current CCS infrastructure. When compared to pre-combustion and oxyfuel, post-combustion stands out as less complex and more adaptable. Pre-combustion, for instance, while efficient, faces challenges like costly regeneration steps and a need for large-scale supplementary equipment. Oxyfuel has a very high capture efficiency but entails complex retrofitting and high costs for air separation (i.e. relevant auxiliary power for separating O_2 from air). Moreover, the high oxygen concentration can alter ash chemistry and cause issues like corrosion, fouling potential leaks, high maintenance costs, and stringent safety management.

Post-combustion capture technology appears to offer a balanced option for CO_2 capture with several compelling advantages. It is highly mature, with the capability to capture CO_2 from a capacity as small as 3 to as large as 5000 MMT in different locations worldwide, indicating a proven track record and scalability. The technology is research-backed, showing potential for increased efficiency and the ability to integrate into existing power plants with relative ease. This retrofitting aspect is crucial, as it allows current plants to upgrade without the need for complete overhauls, thus saving time and resources. While post-combustion does have some disadvantages, such as lower CO_2 separation efficiency due to low CO_2 concentration in flue gas and high energy demands for amine scrubbing, these issues are being addressed through ongoing research and technological improvements. Additionally, the extensive water usage can be seen as a trade-off for its high adaptability and scalability. The economic aspects of post-combustion capture also show promise, despite high capital and operating costs. The large volumes of equipment needed for large plants are offset by the technology capability for small CO_2 level capture, offering versatility for various plant sizes.

Summing up, of these three approaches, CO_2 post-combustion capture offers a viable and tested solution for reducing CO_2 emissions. It is, indeed, currently the most commercially evolved option that is more adaptable to existing infrastructure and have shown promise in reducing the energy penalty associated with CO_2 capture from emission point sources. Its high maturity level, research support for efficiency improvement, and adaptability to existing infrastructures position it as a leading choice among the various carbon capture options. Despite its disadvantages, ongoing enhancements are likely to mitigate these challenges, further solidifying its status as a preferred method in the carbon capture landscape. Building on the understanding that CO_2 post-combustion capture represents a viable, well-tested strategy for reducing emissions, it is clear that this method is at the forefront of commercial application.

To showcase the escalating research focus on Carbon Capture and Storage (CCS) technology, particularly within the realm of post-combustion capture systems, an extensive bibliographic study was undertaken. This involved analyzing the statistics regarding the number and trend of published articles, with 10946 papers on CO_2 capture in the framework of CCS. This analysis was carried out using the "search" and "analyze search results" tools provided by Scopus (Elsevier), and the corresponding graph is depicted in Fig. 2. The results of the literature survey clearly indicate an exponential growth in the number of published papers over the past decade, underscoring the increasing relevance of the topic.

Furthermore, the bibliographic survey was refined to specifically examine "review" type papers, both on CO_2 capture in general and on post-combustion capture in the framework of CCS. Table 10 provides details of all the published review papers. Within this subset, 9 review papers have been published on CO_2 capture in general since 2014, i.e. the techniques available for separating CO_2 from different kind of gaseous streams (i.e. combustion flue gas in the case of post-combustion capture, syngas in the case of pre-combustion capture, etc.). Conversely, 4 papers (highlighted in bold in Table 10) have a more focused perspective specifically on post-combustion capture systems, spanning from 2022 to 2024.

The outcomes of this literature survey clearly indicated that there is both a need and timely relevance for a critical review of the state-of-the-art post-combustion capture techniques. In particular, this review article is specifically focused on delving into diverse methodologies of post-combustion CO_2 capture, dedicating attention to the progress in various CO_2 separation techniques, including absorption, adsorption, and membrane-based methods. A critical vision towards the peculiar features, open challenges, advantages, and disadvantages of each CO_2 post-combustion capture technology will be presented, setting the stage for future research directions. Through a comprehensive examination, the review will shed light on the technological advancements and continuous improvements aimed at addressing the current obstacles.

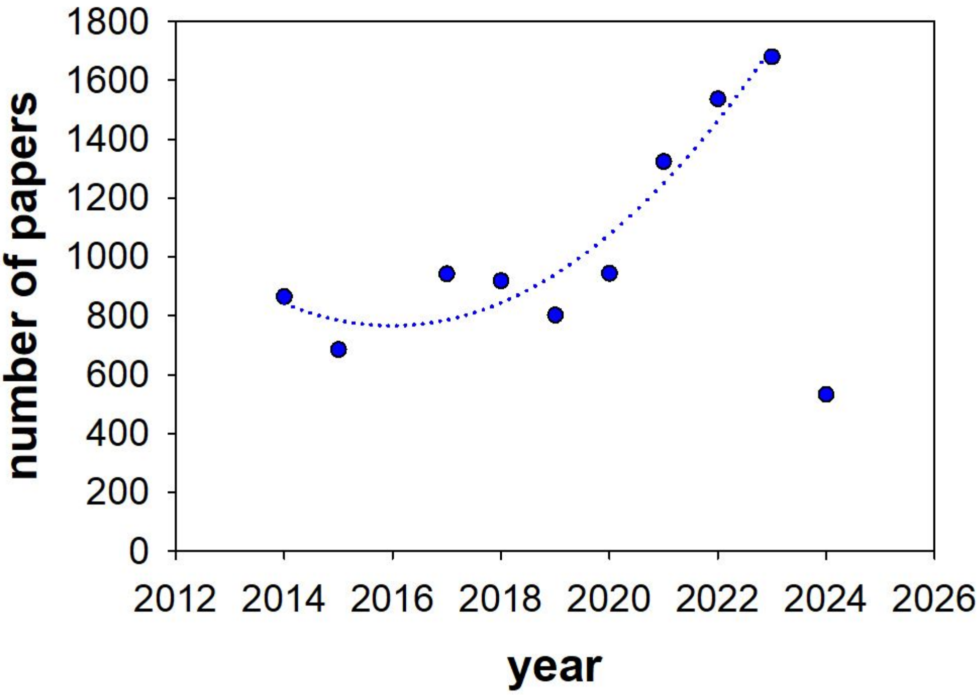

Fig. 2. Time evolution of the number of papers published about CO_2 capture in the framework of CCS.

Table 8. Operating conditions, CO_2 removal efficiency, energy consumption of the different CO_2 capture techniques.

Parameter	**Pre-combustion** [24,41–44]	**Post-combustion** [24,41,44,45]	**Oxyfuel combustion** [24,41,46–48]
CO_2 Concentration	15–40%vol.	4–20% vol.	75–80%vol.
Acid Gases	Sulfur compounds	NO_x, SO_x, COS, H_2S	NOx absent, need of desulfurization
Combustion Medium	Steam/air	Air	High purity oxygen
Equipment Size	Medium size	Large size	Small size
Pressure	High (20–30atm)	Low (0.15– 1atm)	Low (~1 bar)
Temperature	low/high (700 – 1300 °C)	low/high T (400 – 800 °C)	High T (up to 2000 °C)
CO_2 removal efficiency (%vol.)	90	90	>90
Energy Consumption (MJ $kgCO_2^{-1}$)	3.35	4.14	4.05
Cost (USD tCO_2^{-1})	34-63	46-74	52

Table 9. Summary of the main features of the different CO_2 capture options.

Evaluation Criteria	Pre-combustion	Post-combustion	Oxyfuel combustion
Method	Syngas production (CO and H_2), transformed into CO_2 and H_2 by the WGS reaction, with CO_2 being captured from the blend prior to fuel combustion.	CO_2 extraction from the flue gas flow following the burning of high-carbon fuels. The CO_2 content ranges from 10 to 15%, presenting limited separation potential.	Combustion of high-carbon fuel with oxygen of at least 95% purity in the combustion chamber for complete combustion; resulting in flue gas with a low content of CO_2. The CO_2 concentration can reach approximately 95%.
TRL	Extensive application with TRL 9.	Advanced stage with TRL 8, operational in multiple locations.	Operational status with TRL 9.
Advantages	High integration capability with research facilities; modest retrofit requirements for small CO_2 sources.	Greater efficiency in CO_2 capture due to high-pressure operations; beneficial for synthetic gas or hydrogen production.	Exceptionally high CO_2 purity in emissions; minimal pollutants; suitability for a wide operational range.
Disadvantages	Lower efficiency in CO_2 separation for diluted streams; significant energy requirement for compression technologies.	Costly regeneration phase; lower market readiness compared to post-combustion; enhanced support needed for efficiency.	Substantial investment for air separation; challenging scale-up for large-scale application.
Economics	Significant capital and operational costs due to large equipment size for vast flue gas volumes.	Elevated initial costs, particularly for high-capacity operations.	High cost of air separation and modification of the original industrial system.
Maintenance	Complex: need of a total shutdown of the industrial system.	Simple: the industrial system it is connected to does not require shutdown	Complex: the air separation, combustion chamber, and the entire industrial system need to be shut down.

Table 10. Selected review papers focused on CO_2 capture in the framework of CCS.

Source	Year	Focus
Liu et al.[49]	**2024**	**Evaluates current technologies and future challenges in CO_2 capture and mineral storage.** ○ **Emphasis on integrating these technologies with renewable energy sources for enhanced sustainability.**
Obi et al.[50]	**2024**	**Reviews post-combustion CO_2 capture technologies.** ○ **Focus on the efficiency, cost, and energy requirements of absorption, membrane, and adsorption processes.**
Yagmur et al.[51]	2024	Addresses the technological readiness and environmental impacts of various CO_2 capture strategies for CCS. ○ Focus on enhancing energy efficiency and reducing greenhouse gas emissions in industrial applications.
Hekmatmehr et al.[2]	2024	Reviews CO_2 capture technologies for CCS. ○ Emphasis on their readiness levels and suggesting research pathways to address existing challenges.
Kammerer et al.[52]	2023	Review the advancements in CO_2 capturing methods over the last two decades. ○ Focus on their sustainability impact and technological progress.
Joel and Isa[53]	**2023**	**Investigates novel technologies for reducing CO_2 emissions from fossil fuels.** ○ **Emphasis on the intensification technologies for CO_2 capture, with suggestions on how to modify those technologies for suitable application to solvent-based carbon capture.**
Maniarasu et al.[54]	2023	Discusses innovative materials and processes for CO_2 separation and capture. ○ Highlight on the materials and their potential for industrial applications and efficiency improvements.
Alalaiwat and Khan[55]	2023	Examines current technologies and future scenarios for capturing carbon from power plant emissions. ○ Focus on CO_2 separation by absorption and its potential improvements.
Olabi et al.[56]	**2022**	**Discusses the potential, implications and benefits of post-combustion carbon capture technology.**

		○ **Emphasis on its impact on the circular economy.**
Gizer et al. [57]	2022	Explores recent developments in CO_2 capture and utilization. ○ Focus on challenges, materials involved, and directions for future research.
Madejski et al. [58]	2022	Provides a thorough analysis of various CO_2 capture methods. ○ Emphasis on evaluating their application in modern energy technologies for reduced emissions.
Vaz et al. [59]	2022	Examines various technologies for CO_2 capture for CCS. ○ Focus on their application and impact within the energy sectors.
Song et al. [60]	2018	Reviews alternative pathways for efficient CO_2 capture. ○ Emphasis on the potential of hybrid processes to improve capture rates and reduce energy consumption.

3. Post-combustion capture – CO_2 separation technologies

Selecting the right CO_2 separation technology is crucial for the development of every type of post-combustion CO_2 capture system. To achieve this goal, careful consideration must be given to three critical factors: the source of CO_2 emissions (i.e. the type of fuel being used), which influences the presence of trace species and impurities; the composition of the flue gas; the operational conditions of the flue gas, specifically its temperature and pressure (also CO_2 partial pressure) [2]. For example, in separation processes at coal-fired power plants and those utilizing natural gas, gasification, or biomass, the flue gas predominantly comprises CO_2 along with other gases such as N_2, CH_4, H_2, SO_x, NO_x, and O_2 [61]. Hence, the choice of a suitable CO_2 separation method depends on the specific characteristics of the flue gas. Similarly, the effectiveness of the separation process is greatly influenced by the properties of the capture medium, such as its selectivity, affinity, and stability [3].

Within this framework, several technologies are available for CO_2 separation, many of which have been employed globally for industrial purposes over the past 75 years. Within this diverse group, four principal technologies are most used in their broader context: absorption (solvent-based), adsorption (solid sorbents), membranes and cryogenic separation. Fig. 3 provides an overview of these key technologies and media utilized for CO_2 separation. Absorption processes predominantly use solvents, while adsorption techniques employ both natural and synthetic porous solids across different bed configurations. In membrane systems, both organic and inorganic materials, as well as composite membranes formed from their combinations, can be used to enable efficient CO_2 separation. Finally, cryogenic technology uses low-temperature and high-pressure processes to compress and cool the flue gas; CO_2 is condensed into a liquid/solid phase, thus resulting in high-purity CO_2 suitable for subsequent transportation and storage.

In the subsequent sections, an exhaustive review of CO_2 separation techniques is presented, offering detailed comparisons across various capturing media employed in different technological approaches.

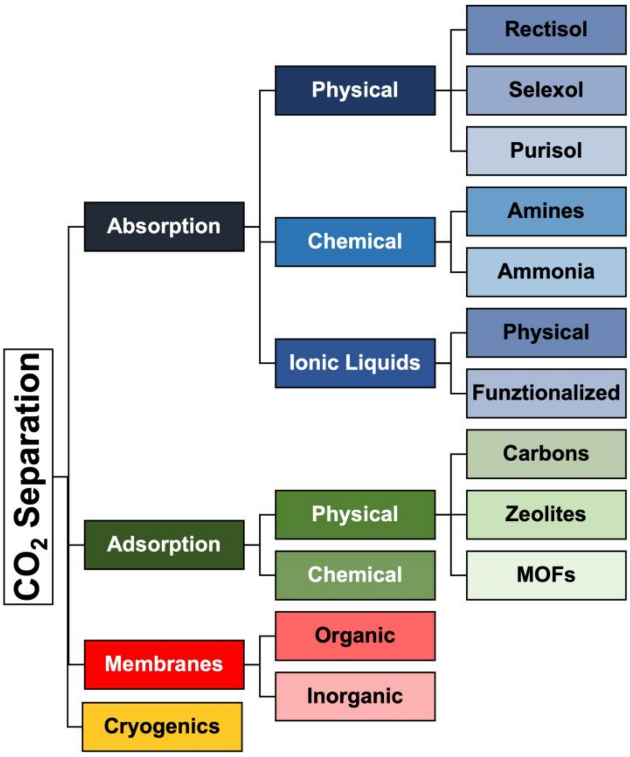

Fig. 3. CO_2 separation techniques along with some commonly used and emerging materials.

3.1. Absorption

Absorption stands as the most established and widely implemented method for CO_2 separation from flue gases, especially prevalent in the petroleum and chemical sectors [24]. This method utilizes a liquid absorbent, or solvent, that can be regenerated through processes such as heating (temperature swing) or decrease of pressure (pressure swing) (Fig. 4) [10,62]. Absorption technologies are categorized into chemical absorption, which relies on acid-base reactions between an alkaline solvent and CO_2, and physical absorption, based on CO_2 solubility in an organic solvent influenced by temperature and pressure [63,64].

The process involves two phases with CO_2 being separated by the solvent in an absorption column, followed by CO_2 extraction in a regeneration column, and recycling the depleted solvent back to the absorber [27]. Absorption occurs at cooler temperatures and higher pressures (i.e. leveraging enhanced gas solubility in liquids), while regeneration happens at higher temperatures and/or lower pressures. The low CO_2 concentrations in post-combustion flue gases (e.g. 3 - 5 %vol. from natural gas turbines; 10 - 15 %vol. from coal-fired power plants) require large absorption towers, increasing capital and operational costs.

For nearly a century, aqueous solutions of alkali metal hydroxides have been used to chemically react with CO_2 in closed environments like submarines and spacecraft, forming insoluble alkali metal carbonates that are subsequently filtered out. However, scaling this method for large sources such as coal-fired power plants is impractical due to the extensive interfacial contact area needed to manage mass transfer resistance at the gas/liquid boundary in absorption columns. This requires large absorbers, contributing significantly to system costs, potentially up to 55% of total capital expenditure [65].

Ionic liquids (ILs), which are composed entirely of ions, are being explored as innovative solvents for CO_2 capture due to their environmentally friendly properties [66]. ILs are often referred to as “green solvents” due to their environmentally friendly properties: i.e. low melting point, allowing them to remain liquid at or near room temperature [67]; low volatility, which reduces the risk of evaporation and solvent loss during the absorption process, contributing to safer and more sustainable operations [67]; good thermal stability, enabling them to withstand the high temperatures required during CO_2 desorption and solvent regeneration phases [67]. These properties collectively make ionic liquids a compelling choice for enhancing the efficiency and environmental performance of post-combustion absorption-based CO2 capture technologies.

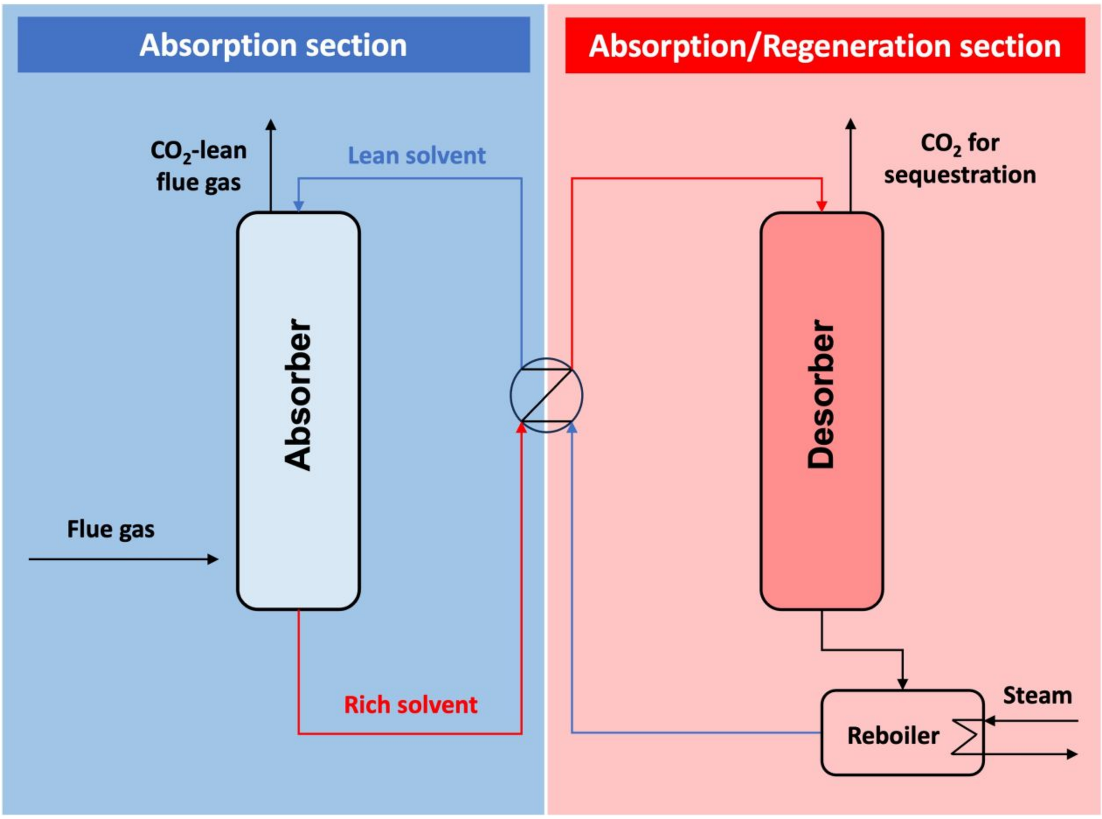

Fig. 4. Scheme of CO$_2$ separation by absorption. CO$_2$ is captured in the absorber (left) and released in the desorber (right).

3.1.1. Chemical absorption

Chemical absorption, a backbone in the gas industry for over 50 years, removes CO$_2$ through solvent scrubbing with recovery efficiencies up to 98% [68]. However, practical large-scale operations typically achieve capture efficiencies between 70% and 95%. During chemical absorption, the solvent chemically interacts with CO$_2$ to form an intermediate compound, through reversible or irreversible chemical reactions. Regeneration occurs by raising the temperature, breaking down the intermediate compound into the original solvent and CO$_2$ [69,70]. The effectiveness of CO$_2$ recovery depends on the reaction dynamics and operational conditions.

The CO$_2$ absorption capabilities and overall performances of different chemical absorbents are reported in Table 11.

3.1.1.1. Amine-based solvents

The solvents commonly employed in absorption-based processes are amines. They capture and release CO_2 through a reversible interaction with CO_2, where the efficiency of CO_2 absorption significantly depends on the chemical composition of the chosen amine [71]. Amines can be classified into three categories based on the number of hydrogen atoms attached to the nitrogen atom [72]:

- primary amines, which contain an $–NH_2$ group (e.g. monoethanolamine - MEA, and 2-amino-2-methyl-1-propanol - AMP);
- secondary amines, with an –NH group (e.g. diethanolamine - DEA, piperazine – PZ, and di-2-propanolamine - DIPA);
- tertiary amines, characterized by an –N group (e.g. methyldiethanolamine - MDEA).

Given the variability in the basic strength of these amines, the binding energy between CO_2 and the amines typically follows the sequence: primary amines > secondary amines > tertiary amines. This hierarchy reflects the inherent differences in their ability to chemically bind CO_2, with primary amines demonstrating the strongest affinity, followed by secondary and tertiary amines, respectively.

The absorption of CO_2 by primary and secondary amines typically follows the zwitterion mechanism [73,74], where CO_2 reacts to form a carbamate, ensuring fast absorption rates. However, this leads to strong C–N bonds in the carbamate, requiring significant energy for regeneration [75]. Conversely, tertiary amines absorb CO_2 through a base-catalyzed hydration mechanism, facilitating the hydrolysis of CO_2 into bicarbonate. While this offers high CO_2 capacity and lower regeneration energy, it comes with slower absorption rates. This trade-off between absorption rate and regeneration energy underscores the challenge of optimizing performance with a single amine type in CO_2 capture processes.

The main features of the different amines include:

- Primary and secondary amines demonstrate high reactivity, enabling the rapid removal of substantial quantities of acid gases [76,77].
- According to stoichiometric calculations, primary and secondary amines capture approximately 0.5 mol CO_2 per mol of amine, while tertiary amines capture about 1.0 mol CO_2 per mol of amine [78].
- Stripping CO_2 from primary and secondary amines during regeneration is more energy-intensive compared to tertiary amines [77,79]. The enthalpy of CO_2 absorption ranges from 80–90 kJ $molCO_2^{-1}$ for primary amines, 70–75 kJ $molCO_2^{-1}$ for secondary amines, and 40–55 kJ $molCO_2^{-1}$ for tertiary amines.
- The reactivity towards CO_2 does align with the amine classification, decreasing from primary to tertiary amines, with reaction rate constants at 25°C being 7000 $m^3\ kmol^{-1}$ for MEA, 1200 $m^3\ kmol^{-1}$ for DEA, and 3.5 $m^3\ kmol^{-1}$ for MDEA [80].

A summary of the most significant physicochemical properties of commonly used amines is presented in Table 12. Ideal characteristics include high CO_2 uptake capacity and thermal stability during amine regeneration, although a high P_{max} (maximum allowable pressure during regeneration) indicating high vapor pressure is generally undesirable. MEA, commonly used in concentrations of 20% to 30% [27], is favored for its cost-effectiveness, efficient kinetics, and rapid mass transfer rates [81–85]. However, MEA-based CO_2 absorption is energy-intensive and costly, requiring approximately 4.2 MJ $kgCO_2^{-1}$, constituting about 36% of the total operational costs [86], with capture costs ranging from 30 to 50 USD MT^{-1} of CO_2 [87]. Piperazine (PZ) and its derivatives offer improved thermal stability and

CO_2 capture capabilities, nearing 1 mmol g^{-1}, compared to MEA, reducing heat losses during absorption-regeneration transitions and enhancing process efficiency [27]. Despite these advantages, PZ higher vapor pressure may complicate practical implementation. It is also noteworthy that, while advanced amines such as PZ and ethyldiethanolamine, offer better chemical stability and resistance to degradation than traditional amines, they come at a higher cost and are more sensitive to other flue gas components like SO_2, NO_x, and fly ash. This necessitates a preliminary treatment to eliminate these impurities from the flue gas before the absorption phase.

According to existing research, amine-based solutions for post-combustion carbon capture face significant challenges: low CO_2 solubility necessitating larger absorption systems and higher costs, as well as the need for pre-cleaning flue gases (removing SO_2, O_2, dust, and hydrocarbons) to maintain absorber column functionality [88]. Additional hurdles include amine thermal degradation, equipment corrosion from degradation by-products, and high energy demands for solvent regeneration, which accounts for about 70% of CO_2 capture facility costs [89]. Moreover, amines released into the atmosphere can undergo photo-oxidation, generating oxidative and thermal breakdown products that pose environmental risks [90]. Of concern are carcinogenic nitrosamines and nitramines formed when amines interact with SO_2, NO_2, and O_2 in flue gases, presenting health hazards. Moreover, another aspect to be considered is the synthesis route of amines, which typically occurs through the interaction between ammonia and ethylene oxide. Yet, ammonia production itself is energy-intensive, contributing significantly to global CO_2 emissions (1.15 [91] to 1.87 [92] $kgCO_2$ $kgNH_3^{-1}$). Thus, the environmental impact of amine-based CO_2 capture extends beyond the capture process to include the initial ammonia production emissions.

To mitigate the high energy demands of regeneration, optimizing the lean solvent loading is a promising approach. For example, increasing MEA concentration from 30 to 40 wt% can reduce energy consumption by approximately 25% [93]. However, higher MEA concentrations may increase corrosion and accelerate solvent degradation rates [91]. Operating the stripper at higher pressures and maintaining a lower temperature of the lean solvent can greatly enhance the efficiency of solvent regeneration, thereby reducing the thermal energy needed for compression by 20% [94]. Nevertheless, the additional cooling water needed to lower the solvent temperature could offset these energy savings [93].

Table 11. CO_2 adsorption performances of different chemical absorbents with corresponding process conditions (P_{CO2} and T).

Solvent	Ads. capacity mmol g^{-1}	P_{CO2} atm	T K	Ref.
2-amino-2-methyl-1-propanol (AMP) (30% wt.)	2.24	1	313	[95]
Diethanolamine (DEA) (40% wt.)	0.95	1	313	[70,95–98]
Methyl diethanolamine (MDEA) (20% wt.)	8.38	1	293	[99,100]
Monoethanolamine (MEA) (30% wt.)	4.09	1	313	[96,97]
Piperazine (PZ) (40% wt.)	2.38	1	313	[101,102]
MDEA/PZ blend (30:20, % wt.)	0.49	1	313	[103,104]
Liquid ammonia (11.7%)	1.91	1	283	[97,105]

Table 12. Properties and characteristic of commonly used amines [27,106].

Amine	Molality (m)	Ads. capacity[1] mmol g^{-1}	$-\Delta H_{abs}$ (kJ mol^{-1})	T_{max}[2] (K)	P_{max}[3] (atm)
Monoethanolamine (MEA)	11	0.66	70	398	2.7
Piperazine (PZ)	8	0.79	64	436	14.3
MEA/PZ Blend	7/2	0.62	80	377	0.7
Methyl diethanolamine (MDEA)/PZ	5/5	0.99	70	393	1.8
MDEA/PZ	7/2	0.8	68	393	1.4
2-Amino-2-methyl propanol (AMP)	5	0.96	73	413	6.1
AMP/PZ Blend	2.3/5	0.7	71	407	4.5
2-Piperidine ethanol	8	1.23	73	400	3.3
2-Methyl piperazine	8	0.93	72	424	9.9
2-Methyl piperazine/PZ	4/4	0.84	70	428	10.3
KGlycinate	6	0.35	69	393	1.08
PZ/AMP	2/4	0.78	72	401	3.4

[1] Ads. capacity is expressed in mmol of CO_2 absorbed per g of water + amine solution.

[2] T_{max} = maximum temperature before thermal degradation of the amine.

[3] P_{max} = highest allowable pressure during regeneration.

3.1.1.1.1. Blended amine-based solvents

Optimal characteristics in amine-based solvents can also be achieved through the strategic blend of sterically hindered amines with primary or secondary amines, complemented by a tertiary amine, thus harnessing the distinct advantages of different types of amines for CO_2 capture. This approach combines the rapid CO_2 absorption rates of primary/secondary amines with the low desorption enthalpy and high CO_2 capacity of tertiary/sterically hindered amines [72]. Structural engineering, including adjustments in functional groups and spatial arrangement, is utilized to tailor these innovative polyamine solvents. The goal is to develop absorbents that match or surpass the performance of current amine blends in efficiency, CO_2 capture capacity, and thermal absorption properties, addressing challenges in solvent-based CO_2 capture technology [107]. This mixed amine strategy enhances the reactivity of less active amines, allowing for lower circulation rates and reduced regeneration costs [108]. It balances fast reaction kinetics with energy-efficient regeneration and high absorption capabilities, offering adaptability for optimizing CO_2 separation processes at low concentrations.

Blending primary/secondary amines with aqueous tertiary amines, such as MEA with MDEA, is a strategy aimed at enhancing performance metrics [72]. This approach involves adding MEA to MDEA solutions to improve CO_2 absorption rates and reduce heat requirements for regeneration compared to using MEA alone [109]. For instance, a MEA/MDEA ratio of approximately 4:1 has been shown to lower lean loading capacity by about 30% compared to pure MEA, indicating potential efficiency and solvent use improvements [72]. Adjusting the MEA/MDEA ratio further enhances reaction kinetics and mass transfer rates, offering a more effective CO_2 capture process. Additionally, blends of MEA with AMP have also been explored to enhance MEA absorption efficiency [110,111]. Within this context, a pilot plant study has been conducted using blends of MEA–MDEA, AMP–DA2MP (DA2MP: 1,5-

Diamino-2-methylpentane), and PZ–MDEA [72]. The AMP–DA2MP blend showcased superior mass transfer and CO_2 absorption efficiency compared to MEA. Additionally, the reaction enthalpy between CO_2 and the AMP–DA2MP blend was lower, resulting in a 13.8% reduction in regeneration energy compared to the use of MEA alone.

Belnding 2-amino-2-methyl-1-propanol (AMP) and piperazine (PZ) is another example of mixed amine solvents [112]. Research indicates that elevating AMP/PZ concentrations can significantly enhance capture efficiency, as it allows operations to proceed well beneath the thresholds for critical viscosity and solubility. Additionally, this blend has been noted for its superior CO_2 absorption rates, reduced heat requirements, and greater thermal stability compared to MEA.

Despite their higher cost compared to single-amine solutions, blended amine solvents show significantly improved performance in laboratory-scale and pilot plant experiments, positioning them as promising alternatives to conventional single amines [72]. Moreover, these blends are compatible with existing amine absorption processes, requiring no major modifications, which facilitates their potential commercialization. However, industrial deployment of blended amine solvents must carefully consider factors like viscosity, volatility, toxicity, degradation, and market price to ensure feasibility, safety, and economic viability for large-scale CO_2 capture applications.

3.1.1.1.2. Non-aqueous amine-based solvents

Using water as a diluent in typical aqueous amine absorbents poses challenges due to its high heat capacity and vaporization enthalpy, which hinder energy-efficient regeneration and contribute to equipment corrosion in amine-based CO_2 absorption. Recent research has therefore explored non-aqueous absorbents as alternatives. Organic molecules like Ethylene Glycol (EG) and Diethylene Glycol, with lower heat capacities and evaporation enthalpies (about half that of water), and requiring reduced stripping temperatures, are emerging as promising substitutes [113].

Shifting to non-aqueous amine-based solvents shows promise in reducing energy demands during regeneration. These solvents focus regeneration energy primarily on breaking CO_2-related products, minimizing contributions from sensible heat and solvent vaporization due to the lower heat capacity and evaporation enthalpy of organic diluents. This adjustment allows non-aqueous amine solvents to potentially achieve substantial energy savings while maintaining CO_2 absorption reactivity akin to aqueous amines [114]. Furthermore, non-aqueous solvents offer operational advantages over traditional aqueous solutions, such as reduced stripping temperatures. This lowers amine losses, prevents thermal and oxidative solvent degradation, and reduces equipment corrosion [115]. These improvements highlight the potential of non-aqueous amine solvents to enhance the efficiency and sustainability of CO_2 capture processes, representing a significant advancement in cleaner industrial technologies.

Recent research on novel non-aqueous absorbents has highlighted the AMP–AEEA–NMP (AEEA: 2-(2-Aminoethylamino)ethanol; NMP: N-Methyl-2-pyrrolidone) solvent, demonstrating a CO_2 uptake capacity of 11.65 mol kg^{-1} [115]. NMP low heat capacity reduces sensible heat, and its low evaporation enthalpy minimizes latent heat during regeneration, requiring approximately 2.09 MJ $kgCO_2^{-1}$ for regeneration, roughly half that of aqueous MEA solutions. Another innovative development is the non-aqueous 2-piperidineethanol–ethylene glycol solvent, which achieves an absorption capacity of 0.97 $molCO_2$ per mole of solvent, forming carbonate species and desorbing CO_2 at a low temperature of 50°C, offering a promising alternative to traditional amine solvents [116]. Barzagli et al. [117,118] compared CO_2 absorption in various aqueous and non-aqueous amine absorbents, including DEGMME (diethylene glycol monomethyl ether), EG–PrOH (Ethylene glycol, EG, with 1-propanol, PrOH) and EG–DEGMME as non-aqueous solvents. Using C13-NMR spectroscopy, they analyzed species distribution and reaction mechanisms in amine/CO_2/solvent systems. Aqueous solutions form carbamate and bicarbonate species upon CO_2 reaction, while non-aqueous solutions form only carbamate, leading to higher absorption rates but lower CO_2 loading compared to aqueous solutions. These findings highlight non-aqueous solvents potential to enhance efficiency and reduce energy requirements in CO_2 capture, signaling a shift towards more sustainable technologies. Additionally, glycol ethers like 2-methoxyethanol (2ME) and 2-ethoxyethanol (2EE) have also emerged as alternatives to water for diluting MEA [119], showing enhanced desorption efficiency and cyclic capacity (1.45 mol kg^{-1}) compared to aqueous MEA, with a 55% reduction in regeneration energy despite increased viscosity during CO_2 absorption.

The significant increase in viscosity observed during CO_2 absorption in non-aqueous amine-based absorbents has been identified as a critical issue for their industrial application, adversely affecting mass transfer and fluid movement. Liu et al. [120] addressed the high viscosity issue in CO_2-saturated non-aqueous amine-based solvents by developing low-viscosity ethylenediamine derivatives for CO_2 capture. These solvents efficiently regenerated at 50˚C to 80˚C. Concerns also arose over significant solvent loss with high volatility alcohols as organic diluents [121]. Some non-aqueous amine-based solvents showed poor regeneration efficiency without N_2 purging. Extensive laboratory-scale research is, therefore, needed to validate the industrial viability of non-aqueous amine solvents for carbon capture processes.

3.1.1.2. Ammonia-based solutions

Exploring alternatives to amine-based scrubbing to address its limitations and inefficiencies, research has delved into non-amine absorbents for post-combustion CO_2 capture. Aqueous ammonia stands out in this regard. The separation process involves a reaction between the flue gas and an aqueous ammonia solution in a wet scrubber, leading to the formation of ammonium bicarbonate (NH_4HCO_3) or ammonium carbonate ($(NH_4)_2CO_3$). Despite the need for heat to decompose the ammonium bicarbonate or carbonate for ammonia solution regeneration, this process is reported to be substantially more energy-efficient, requiring up to 60% less energy compared to the MEA scrubbing process [122]. Furthermore, the energy demand for ammonia-based CO_2 capture is significantly lower, estimated at 1.15 MJ $kgCO_2^{-1}$, in stark contrast to the 4.07 MJ $kgCO_2^{-1}$ energy penalty associated with a 30wt% MEA solution in MEA-based capture processes [123]. As a preparatory step, flue gas is treated to oxidize NO and SO_2 impurities to NO_2 and SO_3, respectively. The scrubbing by-products, mainly ammonium sulfate ($(NH_4)_2SO_4$) and nitrate ((NH_4NO_3), are subsequently utilized as valuable chemicals and fertilizers.

Ammonia-based CO_2 absorption is complex, influenced by factors like concentration, temperature (T), pressure (P), and turbulent conditions [124–126]. Optimizing this process involves fine-tuning parameters to maximize efficiency and minimize costs. Studies have examined how flow rates of liquids and gases, CO_2 loading, and ammonia concentration affect absorption in different reactor setups [127]. Higher CO_2 concentrations with increased flow rates generally reduce absorption efficiency, whereas higher ammonia flow rates or ratios improve efficiency but may lead to solvent leakage. Effective absorption at lower ammonia concentrations depends on maximizing gas-liquid interfacial area. Optimal reaction temperatures (30–40˚C) are critical for achieving high absorption efficiencies with aqueous ammonia [127].

Enhancing the absorption efficiency of aqueous ammonia can be achieved not just refining its physicochemical characteristics but also fine-tuning reaction parameters such as ammonia concentration, flow rate, and the temperature at which the operation is conducted [128–130]. Adding functional additives, such as organic compounds or inorganic salts derived from strong acids and weak bases, can improve absorption efficiency [123,131]. For instance, the addition of merely 1 wt% of amine additives to aqueous ammonia was found to increase CO_2 absorption by 10% [132]. However, it is important to note that absorption efficiency does not markedly increase with higher concentrations of these additives. Physical methods, such as increasing interfacial surface area with solid media or improving mixing with external forces, also boost mass transfer rates [127]. Techniques like hollow fiber membranes [126,133], high gravity technology [134], and spray scrubbing [135,136] can enhance mass transfer coefficients significantly, with hollow fiber membranes improving them by up to 500% compared to traditional packed beds in MEA CO_2 absorption. However, the high costs and flow resistance associated with hollow fiber tubes might restrict their widespread use in gas separation applications.

Ammonia-based CO_2 separation processes, much like amines, encounter significant challenges due to high energy demands and a substantial carbon footprint originating from ammonia production. As one of the most produced chemicals globally, with more than 200 MT $year^{-1}$, ammonia is predominantly manufactured through the Haber-Bosch process. This method involves steam reforming methane to produce hydrogen, which reacts with nitrogen under high pressures (10 to 25 MPa) and temperatures (350 to 550°C) using an iron-based catalyst. Not only does this process account for nearly 1% of global energy consumption, but it also results in significant emissions, with up to 1.87 $kgCO_2$ emitted for every kg of ammonia produced [92]. Additionally, it is estimated that nearly 40% of fertilizer N_2, derived directly from ammonia, is lost to the environment, contributing to atmospheric pollution with dinitrogen and nitrogen oxides [137]. This loss not only signifies a considerable energy waste but also poses environmental risks due to the presence of reactive nitrogen species in the atmosphere and waterways, ultimately affecting the food supply chain. On a positive note, the by-products of ammonia absorption, including NH_4CO_3, NH_4NO_3, and $(NH_4)_2SO_4$, can serve as fertilizers. Therefore, both upstream and downstream factors must be thoroughly considered when evaluating and selecting an appropriate CO_2 capture technology.

3.1.2. Physical absorption

Physical absorption of CO_2 involves its dissolution in a solvent rather than a chemical reaction. According to Henr's law, the solubility of a gas in a liquid is directly proportional to its partial pressure in the gas phase at a constant temperature. Therefore, the solvent capacity to absorb CO_2 is intrinsically linked to the gas partial pressure, with higher pressures enabling greater solubility. This principle introduces a challenge for capturing CO_2 from flue gases of fossil fuel power plants, where CO_2 concentrations are relatively low (around 7 – 15 % for coal-fired plants and 3 - 5% for natural gas plants [2,27]).

Given the principle that gas solubility in a liquid typically diminishes with rising temperatures, effective CO_2 separation via physical absorption in liquid solvents necessitates operations under low temperatures and elevated pressures to ensure higher CO_2 dissolution rates. This step is followed by a desorption phase, conducted at either decreased pressure and/or elevated temperature, facilitating the release and collection of CO_2 from the solvent. Numerous commercial processes designed for physical absorption are utilized for large-scale CO_2 and other impurity separations from flue streams, as detailed in Table 13.

The Selexol process, using mixtures of dimethylethers of polyethylene glycol, excels in removing acid gases such as CO_2 and H_2S. It offers significant benefits including low solvent volatility, low toxicity, excellent thermal stability and chemical resistance, and reduced capital and operational costs in comparison to other physical absorption methods [3]. Additional benefits encompass low corrosion rates due to the absence of heat-stable salt formation, reduced process effluents leading to lesser solvent reclamation and purging needs, and enhanced protection for downstream equipment through the capture of metal carbonyls [2]. Likewise, methanol, the primary solvent used in the Rectisol process, is noted for its stability and lower corrosiveness. Both Selexol and Rectisol are widely used in industrial settings not just for CO_2 capture but also for removing H_2S from flue gases. Yet, a significant limitation of these methods lies in their substantial energy requirements, driven by the heat necessary for conducting the absorption and desorption phases. In contrast, the Purisol process is characterized by its lower energy requirements, whereas the Morphysorb process can achieve a 30% to 40% reduction in operational costs relative to Selexol [80]. Additionally, the Fluor process is made appealing for CO_2 capture due to the high solubility of CO_2 in polypropylene carbonate and the ease of separation facilitated by the weaker solvent-CO_2 bonds, enabling separation at lower pressures [138].

While physical absorption processes for CO_2 capture generally consume less energy than chemical absorption, they face significant limitations in terms of uptake capacity and the range of operating temperatures. Typically, cooling the flue gas to ambient temperature is necessary before absorption can occur, negatively affecting the process thermal efficiency. Therefore, there is a pressing need for the development of physical solvents that are not only more stable but also capable of dissolving higher amounts of CO_2 at elevated temperatures compared to currently available options.

Table 13. Physical absorbents used for commercial CO_2 separation processes [24].

Solvent	Process trade name	Ads. capacity mmol g^{-1}	P_{CO_2} atm	T K
Methanol	Rectisol	0.21	24	243
Dimethyl Ether of Polyethylene Glycol (DEPG)	Selexol	0.14	1	294
N-methyl-pyrrolidone (NMP)	Purisol	0.14	1	298
Propylene Carbonate (PC)	Fluor	0.12	1	298
Dimethyl Carbonate (DMC)	-	0.28	1	298

3.1.3. Ionic liquids

Ionic liquids (ILs) stand out as alternative solvents for post-combustion CO_2 capture due to their unique properties like tunability, extensive liquid range, high thermal stability, low volatility, low toxicity, high polarity and superior CO_2 solubility, making them preferable over traditional volatile organic compounds [139–141]. Moreover, another distinct advantage of ILs is their ability to capture CO_2 directly without the need for a water diluent, simplifying the regeneration process thanks to their low vapor pressure. This characteristic not only simplifies the capture process but also contributes to a reduction in regeneration energy requirements when compared to conventional amine scrubbing methods [142], making ILs a promising option for more energy-efficient and environmentally friendly CO_2 capture technologies.

Blanchard et al. [143] were the pioneers in demonstrating that imidazolium-based ionic liquids can dissolve substantial amounts of CO_2, allowing for the extraction of dissolved products without contamination by the ionic liquid, as it remains insoluble in CO_2. This groundbreaking discovery sparked a surge of scientific interest and research into CO_2 absorption using ILs.

More specifically, ILs generally consist of an organic cation (e.g. Imidazolium, Pyrollidinium, Pyridinium, Ammonium, etc.) matched with either an inorganic or organic anion (e.g. Choride, Bromide, Triflate, Dicyanamide, etc.), showcasing the versatility and wide range of chemical structures possible within this class of materials. The potential combinations of ILs are vast, with their specific properties heavily influenced by the selected anions and cations. Therefore, adjusting these properties to achieve improved CO_2 solubility for efficient capture presents both a significant challenge and an exciting opportunity. This potential for customization makes ILs particularly appealing for CO_2 separation through absorption processes [144,145], as well as through the use of IL-supported and facilitated transport membranes [146].

With reference to the separation mechanism, experimental findings indicate that the interactions between CO_2 and the anionic component of ILs primarily dictate the CO_2 solubility. The role of the cation, while still relevant, is considered secondary [145,147]. These interactions are categorized as Lewis acid-base reactions, where the anion acts as the Lewis base, and CO_2 serves as the Lewis acid. It is observed that ILs with longer cation alkyl side chains tend to exhibit higher CO_2 solubility. This is attributed to a reduction in cation-anion interaction strength and a potential increase in the solvent free volume available for CO_2 accommodation. Moreover, studies, particularly with imidazolium-based ILs, show negligible differences in CO_2 solubility between ILs with hydrogen- versus methyl-terminated anions. This suggests that CO_2-anion interactions do not solely determine CO_2 solubility in ILs, pointing to the influence of other factors [145]. Additional aspects influencing the solubility of CO_2 in ILs include mechanisms such as free volume, where CO_2 molecules find accommodation within the spaces or cavities of the liquid. Furthermore, entropic considerations alongside molecular interactions, such as electrostatic forces, hydrogen bonding, and van der Waals interactions, between the liquid phase entities significantly affect CO_2 solubility. These elements together shape the dynamic and complex nature of how CO_2 dissolves in ILs, highlighting the multifaceted approach needed to enhance ILs for efficient CO_2 capture.

Based on their structure and the nature of interaction with CO_2, ILs can be categorized into both physical ILs, which rely on physical absorption, and functional ILs, which engage in chemical absorption. Significant research into ILs, including conventional, functionalized, and polymerized ILs, for absorption is ongoing [148–161]. ILs CO_2 absorption capacity demonstrates considerable variability [62], as shown in Table 14. Conventional ILs, i.e. lacking chemical functionalization, exhibit a wide range of CO_2 absorption capacities, with specific capacities lower than 1 $molCO_2\ mol^{-1}$ of IL. In contrast, task-specific ILs, i.e. with chemical (amine) functionalization, show an enhanced average absorption capacity, reaching values up to 2 molCO_2 mol^{-1} of IL. Furthermore, ILs that are functionalized with multiple active sites and functionalized IL-based blends, present CO_2 absorption capacities even higher than 2 molCO_2 mol^{-1} of IL [162].

Comparative analyses have also shown that ILs can significantly reduce energy consumption compared to traditional CO_2 absorbents, with selected ILs requiring about 25% and 55% less energy than 30 wt% MEA and 30 wt% MDEA solutions, respectively [163]. Industry-scale simulations (flue gas containing 13 %vol. of CO_2) have shown that transport properties are crucial in selecting an efficient IL for CO_2 separation [164]. Total energy consumption for IL-based CO_2 capture is estimated to be approximately 1.4 MJ kgCO_2^{-1} of CO_2, which is significantly lower than that for amine-based solutions (about 4.07 MJ kgCO_2^{-1} of CO_2). While for amine-based processes the majority of operating costs (70%) is attributed to the solvent regeneration process, for IL-based processes it is tied to electrical expenses for compressors and pumps [164,165]. The economic viability of ILs, therefore, varies with electricity prices, with IL usage being more cost-effective in regions with lower electricity costs, such as China and the US, aligning with the US Department of Energy cost targets [164]. Additionally, the cost of ILs, approximately \$1000 kg^{-1} [166], remains a barrier. Although mass production could reduce prices to around \$40 kg^{-1}, this price is still substantially higher, by 10 to 20 times, than that of commercially available solvents.

Despite their potential superior CO_2 capture properties, hinging on their ability to offer competitive, or ideally superior, features (such as negligible vapor pressure and enhanced selectivity for CO_2 over N_2) compared to existing commercial solvents, further research and development is still needed for their large-scale deployment. Particularly, future investigations should address these commercialization hurdles/challenges of IL-based separation processes: i) rising operational costs due to decreased heat and mass transfer rates, a consequence of increasing solvent viscosity with CO_2

absorption over time [167]; ii) combustibility concerns with certain ILs, which can ignite upon brief exposure to a flame, less than 10 seconds [168,169]; iii) compromised durability and thermal stability throughout the absorption and desorption phases [170]; iv) environmental toxicity in water that may equal or surpass that of traditional solvents [171]; high costs [167]. Specifically, bridging the gap from laboratory to industrial scale necessitates a deeper understanding of system hydrodynamics and transport properties, with current research yet to fully address the design challenges for scaling up IL applications.

3.1.3.1. Physical ionic liquids

Physically absorbed ILs utilize their anionic and cationic structures to absorb and dissolve CO2 through non-chemical interactions, such as hydrogen bonds and van der Waals forces [66], that are relatively weak. This results in lower energy requirements for regeneration but also leads to a modest CO_2 absorption rate, capacity, and selectivity.

The anion is regarded to be crucial in the dissolution of CO_2, while the cation is considered to play a more secondary role. This conclusion is confirmed by both molecular simulation studies and experimental data [172]. For instance, Cadena et al. [173] employed molecular simulations to better understand the high solubility of CO_2 in imidazolium ILs and found that CO_2 predominantly associates with the [PF6] anion, irrespective of the cation. Likewise, in situ ATR-IR spectroscopy data from Kazarian et al. [174] showed favorable interactions between CO_2 and the anions [BF4] and [PF6]. The spectroscopic analysis suggested a Lewis acid-base interaction, with the anion acting as a Lewis base and CO_2 as a Lewis acid. Additionally, the data indicated that the interaction between CO_2 and the [BF4] anion is stronger than that with the [PF6] anion, due to [BF4] being a stronger base. The effect of the anion has been also experimentally investigated by pairing the [bmim] cation with various anions, showing that CO_2 solubility increases in accordance with the different anions [172].

In particular, enhancements in CO_2 solubility within physical ILs can be achieved by fluorinating the anion or by extending the length of the alkyl side chain. Specifically, fluorine-containing ILs generally exhibit higher CO_2 solubilities compared to ILs without fluorine groups. It was found that CO2 solubility increases with the number of fluorine groups in the anion, following this order: [BF4] < [TfO] < [TfA] < [PF6] < [Tf2N] < [methide] < [C7F15CO2] < [eFAP] < [bFAP]. Consistently, Zhang et al. [175] conducted a screening study using COSMO-RS, demonstrating that a longer fluoroalkyl chain in the anion (e.g., [FAP] anion) correlates with higher CO_2 solubilities. It is well-established that the anion is the primary factor in CO_2 dissolution, with the cation playing a secondary role. Nevertheless, cation fluorination (e.g., [C6H4F9mim]) can significantly improve solubility, though to a lesser extent than anion fluorination [172]. Additionally, incorporating long alkyl chains on the phosphonium cation [P66614] can also enhance solubility [172].

One of the most relevant challenges with physical ILs is their high viscosity, which predominantly limits the CO_2 absorption process to mass transfer rather than solubility [142], posing a barrier to their widespread industrial application [176]. The issue of mass transfer limitation in physical ILs can be addressed by modifying the ILs with suitable anions, such as dicyanamide $[DCN]^-$ or tricyanomethanide $[TCM]^-$ [142], or by reducing gas-liquid resistance through the use of supported or encapsulated ILs. These adjustments aim to optimize the properties of ILs for enhanced CO_2 capture, overcoming the inherent drawbacks of physical ILs and potentially paving the way for their more effective and efficient industrial use.

3.1.3.2. Functionalized ionic liquids

Functionalized ILs have been suggested as a significant improvement for CO_2 capture, through the incorporation of reactive and task-specific functional groups that enable chemical absorption, thereby overcoming the limitations associated with physical ILs. The inherent versatility in the structure of

ILs allows for the customization of cation and anion groups to achieve specific properties, such as reduced viscosity, enhanced CO_2 capacity, and decreased reaction enthalpy. Notably, the absorption enthalpy, a critical factor determining the heat required for regeneration, can be effectively controlled by modifying the anion component of the IL. These functional ILs, equipped with chemisorption sites, have demonstrated superior CO_2 absorption performance, positioning them as viable alternatives to traditional amine-based absorbers.

The pioneering approach in structural modifications of ILs involved the functionalization of the imidazolium cation with a primary amine group, marking the creation of the first functionalized IL [177]. This innovation enabled the IL to absorb CO_2 in a 1:2 stoichiometry, similar to MEA, resulting in the formation of ammonium carbamate salts.

Carboxylate-based or acetate-based ILs emerged as leading candidates for CO_2 chemisorption, with [Bmim][acetate] often serving as a reference point for the development of new ILs due to its significant CO_2 capacity and low reaction enthalpy, which is attributed to the formation of carboxylate reaction products. Despite these advancements, challenges such as thermal degradation and poor mass transfer due to high viscosity have not been fully addressed, indicating that while functionalized ILs present a promising route for enhancing CO_2 capture efficiency, further research is needed to resolve these persistent issues.

Amino Acid ILs (AA-ILs) have recently garnered significant interest for their non-toxic biodegradability and notable CO_2 capturing capabilities. Studies have shown that AA-ILs, when functionalized with amine or amino acid groups, exhibit enhanced CO_2 solubility, achieving capacities up to approximately 1.60 molCO_2 per mol of absorbent [178]. The efficiency of CO_2 capture in these ILs is found to vary with the length of the amino acid side chains and is closely linked to the presence of reactive groups, such as amine functionalities [162]. Despite their advantages, AA-ILs face challenges related to their high viscosity, attributed to the formation of hydrogen bond networks within the CO_2-saturated AA-IL solution. This issue can potentially be addressed by strategically positioning the amino groups within the anion component of the IL, thereby diminishing the likelihood of extensive hydrogen bonding [179]. Such structural adjustments could pave the way for reducing the viscosity of AA-ILs, enhancing their practicality for CO_2 capture applications.

Alkylamine Hydroxy Anion ILs (AHA-ILs), particularly those featuring 2-cyanopyrrole anions ([CNPyr]), have emerged as promising candidates for CO_2 chemisorption, offering several advantages including high CO_2 solubility (1:1 stoichiometry), rapid reaction rates, and notable stability [180]. A significant attribute of these ILs is their relatively low viscosity, which facilitates easier handling and processing. The interaction mechanisms underlying CO_2 capture by [DETAH][AHA] illustrate the synergistic roles of the ionic components: the $[DETAH]^+$ cation contributes to a swift CO_2 absorption rate, while the $[AHA]^-$ anion is responsible for the system large absorption capacity and reduced heat requirement for regeneration. A critical aspect of this CO_2 absorption process is the deprotonation of the alkyl chain, a reaction influenced by the anion basicity and the length of the alkyl chain. This mechanism highlights the intricate balance between ionic liquid structure and functionality, underscoring the potential of AHA-ILs in efficient CO_2 capture applications with optimized energy and material efficiencies.

Despite the impressive CO_2 capture capabilities of functional ILs, their practical application on an industrial scale is still constrained by challenges such as high costs and low gas-liquid mass transfer rates, the latter being a consequence of the ILs high viscosity. To overcome these obstacles, strategies beyond mere composition adjustments have been proposed, including the support or encapsulation of ILs and the incorporation of co-solvents, all aimed at reducing viscosity and addressing transport limitations [72]. An innovative approach involves the creation of a biphasic solvent system, which was achieved by diluting a dual-functionalized IL ([DETAH][Tz]) with a 1-propanol–water co-solvent [181]. This method resulted in a system with efficient phase separation, achieving notably low viscosity (2.57 mPa s). Additionally, this biphasic solvent system demonstrated a significant reduction in regeneration heat, being 47.63% lower than that required for MEA, thereby highlighting a promising direction for enhancing the viability of ILs in CO_2 capture through innovative solvent design and optimization.

The challenges of high volatility and energy requirements of amines, alongside the high costs and viscosity of ILs, significantly hinder their broader industrial application in carbon capture. A novel approach to address these limitations involves blending traditional amine solutions with functional ILs, serving as activators to synergistically combine the beneficial aspects of both amines and ILs. This strategy aims to maintain the advantageous properties of each, resulting in an absorbent that experiences a lower regeneration penalty than conventional amine processes, offers improved economic viability, and achieves higher CO_2 absorption rates compared to ILs alone. In this framework, Xiao et al. [182] explored the CO_2 absorption capabilities of several low-viscosity binary absorbents composed of imidazoline ILs and amines. Among these, the blend of MDEA with [BEIM][BF4] demonstrated a high CO_2 capturing capacity, low viscosity, and efficient regenerability. Additionally, the binary solvent system of [bpy][BF4] and MEA was found to provide a 15% energy saving and a 7.44% cost reduction compared to the use of conventional 30 wt% MEA solvent. Furthermore, increasing the concentration of [bpy][BF4] was shown to potentially further decrease the overall energy and operational costs. Thus, the combination of ILs with amines emerges as a compelling solution, promising to deliver both energy savings and cost reductions for CO_2 capture technologies. These findings underscore the potential of IL-amine blends as effective alternatives for enhancing the efficiency and feasibility of carbon capture processes.

Table 14. CO_2 adsorption performances of different ILs with corresponding process conditions (P and T).

ILs	Ads. capacity mol mol^{-1}	P atm	T K	Ref.
Physical				
$[C_{10}Py][NTf_2]$	0.41	20	313	183
$[E_3Py][NTf_2]$	0.40	20	313	183
$[BMIM][PF_6]$	0.60	80	-	143
15-C-5-PL	0.37	10	298	184
Chemical				
[P4442][Cy-Suc]	2.21	1	293	185
[P4442][H-Suc]	1.85	1	293	185
[P4442][Suc]	1.65	1	293	185
$[N_{1111}][Lys] - DMEE$	0.60	1	298	186

3.1.4. Summary - Absorption

Absorption is identified as a prevalent method for CO_2 separation, particularly useful in the petroleum and chemical industries. It involves the use of liquid absorbents or solvents that can be regenerated to capture CO_2. The process is categorized into chemical absorption, which relies on acid-base neutralization reactions, and physical absorption, which depends on the solubility of CO_2 in a liquid solvent. Absorption technologies offer several approaches to CO_2 capture, each with its own set of advantages and limitations. A summary is presented in Table 15.

Chemical absorption is praised for its high recovery efficiencies and the substantial CO_2 capture capacities offered by amines. In particular, amine-based chemical absorption is recognized as the most established method for CO_2 capture, widely implemented in industry for several years. These solvents, through their reactive nature, allow for a significant amount of CO_2 to be absorbed, making the process highly efficient. However, this method is not without its drawbacks. The energy demand for regenerating solvents is high, posing significant operational costs. Additionally, there are environmental and health concerns due to the potential formation of carcinogenic substances from amines, alongside issues related to equipment corrosion and solvent thermal degradation. Innovative developments in the realm of amine-based absorption include strategies like solvent optimization and blending, designed to enhance the efficiency of the process and to reduce the energy consumption associated with solvent regeneration. Likewise, non-aqueous amine solvents also represent a significant improvement over the use of traditional aqueous amine solutions, yet their practicality for industrial use requires thorough evaluation. These non-aqueous systems offer substantial benefits, such as the ability to capture CO_2 with reduced heat requirements for regeneration at lower temperatures, which in turn minimizes amine degradation and equipment wear. However, key challenges that need to be addressed include the rapid increase in viscosity during CO_2 capture, the propensity for high volatility when employing alcohols as organic diluents, and diminished regeneration efficiency in the absence of N_2 purging. Ammonia-based solutions also emerge as an integral part of chemical absorption, offering a more energy-efficient alternative compared to traditional amine scrubbing. This approach leverages the reactive capabilities of ammonia to capture CO_2, forming ammonium bicarbonate and carbonate compounds that can be readily regenerated. Despite its potential for lower energy consumption, the use of ammonia necessitates careful management of its own environmental and operational challenges, including the need for efficient separation and regeneration processes.

On the other hand, physical absorption methods consume less energy than their chemical counterparts and offer substantial benefits in terms of solvent stability and operational cost reductions. Processes such as Selexol and Rectisol are notable for their efficiency and reduced capital and operational expenses. Despite these advantages, physical absorption is limited by its uptake capacity and the range of operational temperatures. Advanced techniques aimed at overcoming these limitations, such as hollow fiber membranes, often come with high costs and flow resistance challenges.

Also, in the framework of absorption, ionic liquids represent a newer class of solvents for CO_2 capture, prized for their tunability, high thermal stability, low volatility, enhanced CO_2 solubility, reduced degradation, and lower heat requirements for regeneration. These features not only make ILs a potent alternative to traditional solvents but also significantly reduce the energy consumption of the capture process. Despite these benefits, the adoption of ILs faces hurdles, including high initial costs and operational challenges such as decreased heat and mass transfer rates, excessive viscosity, combustibility concerns, and environmental toxicity. Molecular simulation emerges as a critical tool in this context, enabling the screening of ILs and facilitating an in-depth understanding of how different cation and anion combinations influence their properties. Currently, the performance of pure ILs falls short when compared directly with traditional amine-based solvents, primarily due to these unresolved challenges. To mitigate the issue of high viscosity, one strategy involves diluting ILs with a co-solvent or blending them with amine solutions, which can also help address cost concerns by reducing the heat required for solvent regeneration. Consequently, a detailed feasibility and economic analysis is crucial prior to considering ILs for industrial-scale carbon capture applications. Moreover, conducting pilot-scale tests on IL-based absorbents is essential to comprehensively assess their efficacy in capturing carbon, ensuring that any proposed solutions are both technically viable and economically sustainable in real-world settings.

In conclusion, while each absorption method offers promising avenues for efficient and effective CO_2 separation processes, they also present unique challenges that must be addressed. Innovations in solvent technologies, alongside strategic optimizations and developments, continue to play a crucial role in advancing the field of CO_2 capture, aiming to mitigate the environmental impact of carbon emissions while ensuring economic viability and operational efficiency.

Table 16 provides a synthesized overview of recent R&D projects focused on CO_2 absorption for post-combustion capture. Specifically, it details project focuses, contractors, durations, and the most relevant operational and economic parameters. These projects, spanning from 2018 to 2026, operate at various TRLs, ranging from lab-scale (TRL 3) to large pilot-scale (TRL 7-8), indicating ongoing efforts to advance CO_2 absorption technologies from experimental stages to practical, scalable applications [2,187].

Table 15. CO_2 absorption processes: advantages and disadvantages.

	Pros	Cons
Chemical		
Amines	○ High recovery efficiencies (up to 98%). ○ High reactivity and substantial CO_2 capture capacities with amines. ○ Potential for efficiency enhancement and energy reduction through optimization and blending	○ High energy demand for solvent regeneration. ○ Amine degradation due to heat and other flue gas components (NOx, Sox, O2) ○ Environmental and health risks from carcinogenic substance formation. ○ Equipment corrosion
Ammonia	○ More energy-efficient alternative to traditional amine scrubbing. ○ Capable of forming easily regenerable compounds. ○ Higher absorption capacity compared to amines.	○ Requires efficient separation and regeneration processes. ○ Associated environmental and operational challenges.
Physical		
Selexol/Rectisol	○ Lower energy requirements. ○ Significant benefits in solvent stability and reduced operational costs.	○ Limited uptake capacity and operational temperature range. ○ High costs and flow resistance with advanced techniques.
Ionic Liquids (ILs)	○ Tunability ○ High thermal stability, low volatility, minimal corrosivity, reduced degradation and superior CO_2 solubility. ○ Significant reduction in energy consumption compared to traditional CO_2 absorbents.	○ High initial costs, though potentially reducible through mass production. ○ Decreased heat and mass transfer rates. ○ High viscosity ○ Combustibility and environmental toxicity.

Table 16. Recent R&D projects focused on CO_2 absorption processes for post-combustion capture [2,187].

Solvent	Focus	Contractor	Duration	CO_2 Loading mol/mol	P atm	T °C	Cost of Solvent \$ kg^{-1}	Capture cost \$ tCO2^{-1}	CO_2 Recovery %vol.	CO_2 Purity %	TRL
Non-aqueous solvent	Capture from cement plant	Research Triangle Institute (RTI)	2023-2025	-	-	<100	-	-	95	-	6 FEED
Amine-based	Capture from steel plant	Univ. of Kentucky Research Foundation	2022-2026	-	-	-	-	50	>95	>95	6 Small Pilot
Solvent	Retrofit NGCC	Univ. of Kentucky Research Foundation	2022-2025	-	-	-	-	-	>99.8	>95	5-6 Bench
Amine-based	Retrofit NGCC power plant	Electric Power Research Institute (EPRI)	2022-2024	-	-	-	6.5	-	95	-	6 FEED[1]
Water-lean solvent	EEMPA solvent for post-combustion	Electric Power Research Institute (EPRI)	2020-2025								6 Small Pilot
Amine-based	Retrofit NGC	Bechtel National, Inc.	2019-2022	0.4-0.49	1.089	53.5	1-2	114.5	80-90	>99	5-7 FEED[1]
Water-lean amine	Develop a novel amine-based	ION Engineering, LLC	2019-2022	0.5-1.0	1	40	-	39-45	-	-	6 Pilot
Amine-based	Retrofit to NGCC	Southern Company Services, Inc.	2019-2022	0.9-1.1	30-60	-	-	-	90	99.9	5-7 FEED[1]
Amine-based	Linde-BASF advanced capture process	University of Illinois	2018-2026	-	1-3.4	30-140	-	-	90	99.8	7-8 Large Pilot
Water-lean solvent	Molecular refinement of water-lean solvents	Pacific Northwest National Laboratory	2018-2021	0.29	1	40	13	-	90	95	3 Lab-scale

[1] FEED = Front-end engineering design

3.2. Adsorption

Adsorption is a process where gas phase molecules are captured by solid surfaces (Fig. 5). It is categorized into physical and chemical adsorption based on the interaction forces [188]. Physical adsorption, or physisorption, involves weak van der Waals forces, characterized by a low heat of adsorption around 10 kJ mol^{-1} [189], allowing for reversible adsorption. Chemical adsorption, or chemisorption, features stronger bonds, similar to heat of reaction values around 100 kJ mol^{-1} [189], involving electron or atom transfer, which is less easily reversible.

Selecting the right adsorbents, whether physical or chemical, is complex and vital, especially under the low CO_2 pressures typical of post-combustion conditions, demanding a balance of economic and performance criteria. The key features for selecting the most appropriate sorbent material are:

- CO_2 adsorption capacity: it defines the sorbent ability to adsorb CO_2 at thermodynamic equilibrium [190,191]. Key for reducing capital costs by minimizing sorbent volume and process equipment size. Optimal materials demonstrate high uptake at low CO_2 pressures, but practical performance is better judged by the working capacity [192,193], representing CO_2 captured in a complete cycle, aiming for 3-4 mmol g^{-1} for competitiveness [194,195].
- CO_2 selectivity: it measures the sorbent preference for CO_2 over other flue gas components, affecting captured CO_2 purity and thus the economics of transport and sequestration [196].
- Tolerance to moisture and impurities: essential for practical operation, sorbents must withstand flue gas contaminants like NO_x and SO_x without significant performance degradation, avoiding the need for costly upstream drying/purification [197,198].
- Adsorption kinetics: Speed at which CO_2 is adsorbed affects system cycle time and economics. Fast kinetics result in efficient capture processes, indicated by sharp CO_2 breakthrough curves [199,200].

- Ease of regeneration: Sorbents should regenerate under mild conditions to lower capture costs, with adsorption heats changing significantly between physical (20-50 kJ mol^{-1}) and chemical adsorbents (60-100 kJ mol^{-1}) [195].
- Stability to repeated cycles: Longevity and recyclability of sorbents are critical for cost-effective operations, favoring materials with durable performance over multiple cycles [195].
- Sorbent costs and synthesis: Economic feasibility and environmental impact of sorbent production are crucial, with a target cost around \$5 kg^{-1} [201] and a process that is scalable and environmentally friendly [64].
- Mechanical/thermal stability: Sorbents must maintain structural integrity under operational conditions to preserve CO_2 capture efficiency and minimize makeup rates, ensuring the process remains economically viable [7].

Even though it is rarely included in the set of required features for an adsorbent, environmental sustainability should also be regarded as a crucial and essential aspect, alongside the above-mentioned commonly considered key features. The development of sustainable adsorbents represents a critical advancement in CO_2 adsorption technologies. Emphasis on adsorbents derived from renewable resources or waste materials can yield significant environmental and economic benefits, thereby aligning with global initiatives aimed at reducing greenhouse gas emissions and mitigating climate change. The utilization of sustainable materials has the potential to markedly reduce the carbon footprint associated with the production and disposal of adsorbents, thereby minimizing their overall environmental impact. This approach is consistent with the objective of developing eco-friendly and economically viable solutions for CO_2 capture. In this context, adsorbents derived from agricultural waste, biochar, and other biomass materials offer a cost-effective solution while promoting waste valorization and resource efficiency. Specifically, materials such as biochar, produced from agricultural residues or forestry by-products, can convert waste into valuable CO_2 capture media, thereby diverting waste from landfills and sequestering carbon, contributing to the reduction of greenhouse gas emissions [202]. The utilization of Life Cycle Assessment (LCA) is crucial for evaluating the environmental impacts of these adsorbent materials from production to disposal, assessing factors such as resource extraction, energy consumption, emissions, and waste generation [203]. LCA comparisons indicate that bio-based adsorbents typically exhibit lower global warming potential (GWP) and reduced energy consumption relative to synthetic adsorbents, as well as lower impacts in terms of acidification, eutrophication, and toxicity [202,204,205].

Among the various challenges facing the large-scale use of adsorption as a primary post-combustion CO_2 capture method, developing an efficient and economically viable sorbent regeneration process is paramount. The regeneration, which involves CO_2 desorption, can be initiated by modifying either temperature or pressure, leading to temperature swing adsorption (TSA) and pressure/vacuum swing adsorption (PSA/VSA) techniques, respectively (Fig. 5). Additionally, a hybrid approach that adjusts both temperature and pressure, known as VTSA/PTSA, can be employed to optimize regeneration. More specifically, TSA involves CO_2 adsorption at lower temperatures followed by regeneration through heating. TSA operates at low pressures (<4 bar), facilitating maintenance and exhibiting tolerance to impurities in the flue gas, such as NO_x, SO_x, and water vapor. This makes TSA a low-risk and short-term implementation option [206]. However, the necessity of high temperatures (150–200°C) for regeneration, contrasting with lower adsorption temperatures (around 50°C), may lead to energy inefficiency due to repeated heating and cooling cycles. Efforts have focused on reducing energy consumption through process optimization and developing advanced sorbents with high adsorption capacity and low regeneration temperatures [7]. Integration into existing power plants is feasible, also exploiting abundant low-grade energy for regeneration. Dilution issues from inert purging gases can be mitigated by using steam/CO_2 or indirect heating [207]. On the contrary, PSA and VSA recover CO_2 by altering pressure, operating at low temperatures and, therefore, eliminating the energy penalties from repeated heating/cooling cycles [208]. However, both of them are energy-intensive processes, especially for achieving vacuum levels below 100 mbar (abs), which are not advisable in industrial applications. A promising solution is, therefore, the combination of TSA and VSA, known as Vacuum Temperature Swing Adsorption (VTSA), which exploits the advantages of both approaches. VTSA reduces the temperature requirements of standard TSA, enabling significant energy savings with proper low-grade waste heat utilization, while enhancing CO_2 purity and recovery compared to standard VSA setups [208].

Regardless of the specific strategy, results reported in the literature demonstrate promising process performances, with energy consumption typically ranging between 2 and 3.5 MJ $kgCO_2^{-1}$ [209–219]. Notably, this falls well below the energy consumption levels of conventional MEA absorption processes, which exceed 4 MJ $kgCO_2^{1}$ [86].

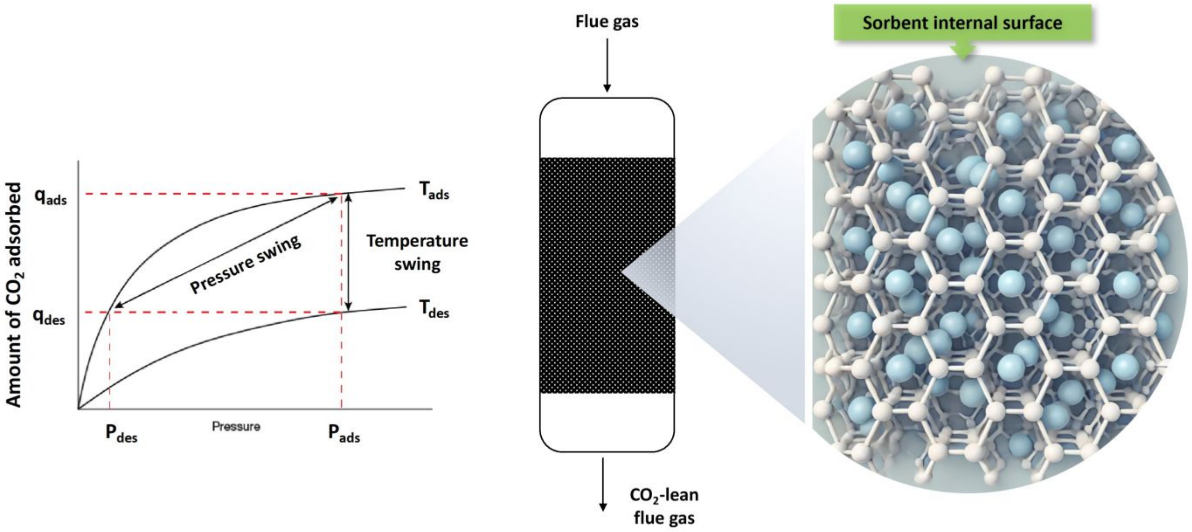

Fig. 5. Scheme of CO_2 separation by adsorption: adsorption/desorption via pressure and temperature swings. The magnified view shows the porous structure of a general sorbent material.

3.2.1. Physical sorbents

Physisorption involves CO_2 molecules being attracted and held onto the adsorbent surface primarily through van der Waals forces, as well as interactions between CO_2 quadrupole moments and the adsorbent ionic and polar sites. Key materials utilized for physisorption include carbon-based sorbents, zeolites, and Metal-Organic Frameworks (MOFs), each offering distinct advantages in terms of adsorption capacity, selectivity, and regeneration potential for CO_2 capture applications.

3.2.1.1. Carbons and carbon-based materials

Carbon, ranking as the sixth-most abundant element globally [220], is the basis for a wide array of cost-effective carbonaceous materials. These include activated carbons (ACs) and various carbon nanomaterials like carbon nanotubes (CNTs), fullerenes, graphene, and carbon nanofilms or fibers, known for their high surface areas and pore volumes [220]. Among these, activated carbons are particularly notable for their affordability and widespread industrial use [221,222]. Despite their comparatively lower adsorption capacity for CO_2 under low pressures than zeolites [221], their standout feature is their exceptional ability to regenerate. This quality ensures that their CO_2 capture efficiency remains stable over numerous cycles of adsorption and desorption, making them efficient and reliable physisorbents [223].

Multiple activation methods have been implemented to produce ACs characterized by enhanced specific surface area (SSA) regulated pore configuration. ACs can be synthesized from a variety of microporous and mesoporous materials, utilizing sources like coal, wood, coke pitch, and various biomass materials such as used coffee beans, coconut shells, sawdust, and olive pits [224]. Independent of their origin, these carbons are known for their substantial surface area and pore volume. They exhibit unique chemical properties, which can impart either acidic or basic qualities to the AC [189,220,225,226]. These characteristics are largely due to the presence of different heteroatoms like oxygen, nitrogen, hydrogen, phosphorus, and sulfur in the carbon structure, which may originate from the raw materials used or the activation technique applied [7]. The CO_2 adsorption effectiveness and selectivity of ACs are significantly influenced by the specific surface functional groups these heteroatoms create, as well as by the dispersed electrons in the carbon architecture [7]. The presence of oxygen-containing groups on AC surfaces tends to confer an acidic property, which intensifies as the oxygen content increases [7]. Conversely, the basic nature of ACs is primarily associated with the π-electron systems in the carbon aromatic rings and the presence of nitrogen-based groups, which contribute to attracting protons [7].

Extensive research has been conducted on activating ACs through different methods to enhance their CO_2 capture capabilities [189,225,227]. These activation processes are broadly classified into physical or chemical categories [228]. The CO_2 adsorption capabilities and overall performance of both physically and chemically activated carbon materials are detailed in Table 17.

Physical activation involves transforming the base material into carbon under low-temperature, oxygen-free conditions, followed by activation using steam, CO_2, O_2, or other gases at elevated temperatures (800–1000˚C) [225,229–231]. This process, especially under an inert atmosphere like nitrogen or helium, primarily removes oxygen-containing groups from the carbon surface, thereby increasing its hydrophobic properties and enhancing Lewis basic sites [7,232]. Chemical activation, on the other hand, employs acids, bases, or salts to modify the raw material, aiming to expand its surface area and introduce or eliminate particular functional groups [7,232]. This often involves incorporating nitrogen-rich functionalities onto the carbon surface through reactions with nitric acid, ammonia, amines, and nitrogen-based compounds [7,233], enhancing the adsorbent's ability to capture CO_2 [234]. Different activating agents, including KOH, NaOH, $ZnCl_2$, H_3PO4, H_2O_2, and K_2CO_3, are used to create porous carbon structures. Among these, KOH stands out as a particularly common choice for crafting ACs due to its effectiveness in generating high specific surface areas and intricate microporous frameworks. The use of KOH can lead to carbons with specific surface areas between approximately 2500 to 3000 m² g^{-1}, alongside an optimal pore size distribution. Moreover, the CO_2 adsorption capacity of KOH-activated carbons typically exceeds 4 mmol g^{-1} at 25°C. The presence of narrow micropores has been identified as a key factor in achieving this exceptional CO_2 capture performance [235]. Despite its effectiveness in enhancing the specific surface area and creating a microporous structure favorable for CO_2 capture, KOH activation encounters two significant challenges. First, the requirement for high temperatures exceeding 600°C during activation is energy-intensive and not cost-effective. Additionally, KOH high corrosiveness, which escalates with increasing temperatures, severely limits its practicality for widespread application. Addressing these issues, Sevilla et al. [236] introduced a chemical activation technique using potassium oxalate ($K_2C_2O_4$), a less corrosive and toxic alternative to KOH. This activation method, also in combination with melamine, particularly when applied to glucose, demonstrated impressive CO_2 adsorption capacities, ranging from 2 to 4.5 mmol g^{-1}. Potassium oxalate stands out not only for its efficiency in CO_2 capture but also for the advantageous pore structure it promotes. Narrow micropores are particularly effective for CO_2 adsorption at pressures below 1 bar, while a combination of micro- and mesopores shows enhanced CO_2 adsorption at higher pressures. Importantly, the presence of ultra-micropores (less than 1 nm) plays a crucial role in achieving high CO_2 adsorption capacities under ambient conditions. Further insights into the mechanisms behind the great CO_2 capture capacity of carbon materials and their selectivity were provided by Qi et al. [237] through first-principles simulations. These studies revealed that CO_2 exhibits a stronger thermal effect within micropores, leading to heightened CO_2 selectivity. This effect becomes increasingly pronounced as pore sizes diminish, highlighting the critical role of ultra-micropores in distinguishing and adsorbing CO_2 effectively.

ACs also stand out among sorbent materials for its regenerative efficiency, requiring minimal energy and lower temperatures for renewal, alongside the widespread accessibility of their source materials and notable thermal resilience [189]. Their ability to adsorb CO_2 is notably enhanced under elevated CO_2 pressures. However, unlike materials such as zeolites, ACs performances in capturing CO_2 is somewhat compromised by humidity and contaminants like NO_x and SO_x, leading to a reduced adsorption capacity under humid conditions compared to dry ones [238,239]. AC physical robustness is challenged during use due to its inherent fragility, with attrition being a common issue [189]. Current research is directed towards enhancing the CO_2 adsorption efficiency and specificity of AC at reduced CO_2 pressures, with a particular interest in utilizing innovative and sustainable materials to develop sorbents with adjustable surface areas and pore structures [227].

Carbon nanomaterials, such as carbon nanotubes (CNTs) and graphene, exhibit significant CO_2 adsorption capacities, rapid kinetics, moisture resistance, and minimal energy needs for regeneration [221]. Their typical properties and adsorption performances are summarized in Table 18. They are characterized by a hierarchical pore structure, combining macro- and micropores, alongside a vast surface area and robust chemical and physical properties [221]. This unique pore architecture is particularly advantageous for CO_2 capture, offering efficient pathways through macropores while leveraging the extensive surface area provided by micropores. Research has intensively explored the development and analysis of CNTs, including both multi-walled (MWCNTs) and single-walled variants (SWCNTs), for their potential in CO_2 sequestration [240,241]. Critical factors influencing CNTs CO_2 adsorption include the proportion of accessible nanotubes, oxygen presence, adsorption sites availability, and the impact of dopants or contaminants [242]. The influence of oxygen on CNTs CO_2 capture capabilities has prompted investigations into introducing and subsequently removing oxygen-rich functional groups to enhance their adsorption efficiency and selectivity [7].

Graphene, a flat single layer of sp2 hybridized carbon atoms arranged in a two-dimensional honeycomb lattice, exhibits notable CO_2 adsorption capabilities [243]. The vast active surface area of graphene has attracted significant research interest as a potential CO_2 sorbent over the past decade. Studies have primarily concentrated on the creation of novel structures, modifications of surface characteristics, and the production of hybrid materials with adjustable porosity [189]. The ability to attach various functional groups or heteroatoms to graphene surface facilitates the customization of its surface properties, all while preserving its inherent structure.

Table 17. CO_2 adsorption performances for physically and chemically ACs with corresponding process conditions (P_{CO_2} and T).

Raw material	Activation	SSA $m^2\ g^{-1}$	P_{CO_2} atm	T K	Ads. capacity mmol g^{-1}	Ref.
	Physical					
Coconut shell	CO_2	371	0.20	298	1.80	244
Almond shell	CO_2	862	1.00	298	2.70	245
Olive stone	CO_2	1215	1.00	298	3.10	245
Date stone	N_2	957	0.25	293	1.66	246
Coffee residue	CO_2	593	1.00	298	2.40	247
Dialdehyde cellulose	CO_2	1241	1.00	298	5.52	248
Cladophora cellulose	CO_2	832	1.00	298	4.97	248
Dialdehyde cellulose	N_2	455	1.00	298	3.21	248
Cladophora cellulose	N_2	500	1.00	298	2.64	248
Almond shell	O_2 (3%)	557	1.00	298	2.10	249
Olive stone	O_2 (3%)	697	1.00	298	2.00	249
	Chemical					
Surgical face mask	KOH	750	1.00	293	2.61	250
Cellulose	KOH	2370	1.00	273	5.80	235
Starch	KOH	2190	1.00	273	5.60	235
Algae	KOH	1940	1.00	273	7.40	251
Yeast	KOH	1348	1.00	298	4.80	252
Rice husk	KOH	1495	0.20/1.00	298	0.72/2.5	253
Sludge	NaOH	179	1.00	298	1.30	254
Wood	H_3PO_4	1889	1.00	303	2.90	255
Palm stone	H_3PO_4	1320	1.00	273	3.10	256
Hazelnut shell	$NaNH_3$	2318	1.00	273	5.91	257
Coconut shell	K_2CO_3	1082	1.00	273	5.12	258
Sub-bituminous coal	K_2CO_3	1773	1.00	273	4.36	259
Rice husk	K_2CO_3	1097	0.25/1.00	295	1.25/2.90	260
Glucose	$K_2C_2O_4$	1690	0.15/1.00	298	1.4/4.5	236

Glucose	$K_2C_2O_4$+melanine	3470	1.00	298	2.5	[236]

Table 18. CO_2 adsorption performances of CNTs and graphene-based adsorbents with corresponding process conditions (P_{CO2} and T).

Sorbent	SSA $m^2\ g^{-1}$	T K	P_{CO2} atm	Ads. capacity mmol g^{-1}	Ref.
CNTs	394	298	0.10	0.52	[261]
MWCNTs	-	293	0.20	0.84	[262]
MWCNTs	407	293	0.15	0.64	[263]
MWCNTs	407	333	0.10	0.34	[263]
SWCNTs	1587	308	0.10	0.50	[264]
Graphene	477	273	0.15	0.70	[265]
Graphene nanoplates	480	298	0.20	0.35	[266]

3.2.1.2. Zeolites and zeolite-like materials

Zeolites are crystalline materials composed of Si, Al, and O, forming TO_4 tetrahedra where T can be either Si or Al. These structures create a network of channels and cavities, offering a high specific surface area and defined porosity [7]. Zeolites stand out as one of the most effective sorbents for CO2 post-combustion capture, thanks to their ability to combine substantial adsorption capacities with rapid kinetics under conditions typical of post-combustion flue gases and biogas (T = 0–100 °C; P_{CO2} = 0.1–1 atm). Zeolites, like activated carbons, demonstrate excellent regenerability; indeed, they maintain their CO_2 adsorption efficiency over numerous adsorption/desorption cycles [267]. An overview of CO_2 adsorption characteristics and performance for different synthetic and natural zeolites is detailed in Table 19.

Zeolites stand out for their unique attributes, such as chemical composition, pore structure, and architecture, which significantly influence their CO_2 adsorption capabilities [267]. The presence of cations within the silicate framework, like Na^+ and Li^+, imparts a negative charge to zeolites, enhancing their ability to adsorb CO_2, a process that varies with the type and quantity of these cations [268]. Furthermore, the Si/Al ratio within zeolites is crucial for their CO_2 adsorption efficiency. Specifically, zeolites with a lower Si/Al ratio, indicating a higher count of extra-framework cations, are generally more effective in capturing CO_2 [269]. This is because aluminum atoms within the structure contribute negative charges that are balanced by additional exchangeable cations, thereby improving adsorption performance.

With reference to the chemical compositions, numerous studies aim to enhance zeolites CO_2 adsorption capacity by replacing their original framework cations with alkali ones, such as Na, Rb, Li, Cs, and K [270]. Zeolites with low silica content are known for their high CO_2 adsorption capacity and selectivity at low pressures, but they generally exhibit poor moisture tolerance. Moisture significantly impacts CO_2 capture efficiency, as water molecules are preferentially adsorbed over CO_2 [271]. This challenge has led to research into hydrophobic zeolites with higher silica content, like MWW zeotype [272] and NaZSM-5 [273], which are less affected by moisture. Similar to carbon-based adsorbents, zeolites can also be adversely affected by trace amounts of NOx or SOx, which can reduce their CO_2 adsorption capacity. With reference to the specific porosity, the pore size of a zeolite directly influences the diffusion rate and selectivity of CO_2. Larger pore sizes facilitate CO_2 diffusion, while smaller pores, close to the kinetic diameter of CO_2, enhance kinetic selectivity, aiding in the separation of CO_2 from other gases [274].

In the domain of synthetic zeolites, geopolymers emerge as synthetic alumino-silicates with inherent and adjustable porosity [275], explored both independently and in conjunction with 13X zeolite for CO_2 sorption [276]. Their CO_2 adsorption capacity and selectivity are notably effective in the low to medium concentration range (up to 20% vol.), attributed to the synergistic effects in composite materials incorporating both 13X and NaA zeolite phases [276]. Additionally, the low-temperature, eco-friendly synthesis process for geopolymers facilitates the production of monoliths or granules that exhibit enhanced mechanical durability and minimal impact on gas diffusion.

Natural zeolites emerge as a sustainable alternative to their synthetic counterparts, which are hindered by high production costs and environmental concerns [277,278]. These naturally occurring minerals are found in abundance across various geological formations worldwide [279,280], offering a cost-effective resource despite their variable composition and potentially lower separation efficiency compared to synthetic zeolites [279]. Their widespread availability and negligible cost make them an attractive option for CO_2 capture from flue gases [200] and biogas purification, despite the trade-offs in purity and performance [281,282]. A recent study [283] also investigates the use of natural tuff from construction and demolition waste as a CO_2 adsorbent, revealing that cation-exchange, especially with Li and Na, significantly enhances its microporosity and surface area, thus also boosting its CO_2 capture capabilities.

Table 19. CO_2 adsorption performances of zeolite and zeolite-like adsorbents with corresponding process conditions (P_{CO2} and T).

Sorbent	Nature	Type	Framework	Surface area m^2 g^{-1}	T K	P_{CO2} atm	Adsorption capacity mmol g^{-1}	Ref.
13X	Synthetic	Na-exchanged X-zeolite	FAU	710	393	0.15	0.70	284
13X	Synthetic	Na-exchanged X-zeolite	FAU	960	298	0.10	0.38	268
NaY	Synthetic	Na-exchanged Y-zeolite	FAU	-	295	1.00	4.06	285
NaY	Synthetic	Na-exchanged Y-zeolite	FAU	542	323	0.20	0.05	286
NaX	Synthetic	Na-exchanged X-zeolite	FAU	534	323	0.20	0.60	286
CaA	Synthetic	Ca-exchanged A-zeolite	LTA	397	323	0.20	0.75	286
CsY	Synthetic	Cs-exchanged Y-zeolite	FAU	842	333	0.10	0.86	287
APG-II	Synthetic	Na-exchanged X-zeolite	FAU	710	393	0.15	0.38	284
Naβ	Synthetic	Na-exchanged β-zeolite	BEA	570	273	0.15	2.30	288
Geopol-G13	Synthetic	Geopolymer	-	50	308	0.20	0.38	276
Na-$G_{1.2}$-Z	Synthetic	13X-Geopolymer composite	-	211	308	0.20	1.5	277
ZAPS	Natural	Erionite	-	426	290	1	2.80	289
ZNT	Natural	Mordenite	-	266	290	1	1.80	289
Tuff	Natural	Tuff	-	36	298	0.15	0.62	200
Li-Tuff	Natural	Li-exchanged tuff	-	155	298	0.10/0.20	0.95/1.09	283
Na-Tuff	Natural	Na-exchanged tuff	-	81	298	0.10/0.20	0.73/0.89	283
NH_4-Tuff	Natural	NH_4-exchanged tuff	-	7	298	0.10/0.20	0.47/0.57	283

3.2.1.3. MOFs and MOF-like materials

Metal-organic frameworks (MOFs), composed of metallic nodes and organic linkers, have emerged as highly promising materials for CO_2 capture following post-combustion processes [290]. Their structure allows for exceptional CO_2 adsorption capabilities, and they exhibit remarkable resilience, maintaining efficiency across numerous adsorption/desorption cycles without significant loss of performance [290]. CO_2 adsorption characteristics and efficiency of various MOFs are summarized in Table 20.

Due to the vast potential for customization through the selection of metallic nodes and organic linkers, MOFs can be engineered to exhibit unique properties, including varied pore sizes and structures [291]. This customization enables the creation of MOFs with exceptionally high surface areas, reaching up to 3000 m^2 g^{-1}, and specific pore sizes tailored for optimal CO_2 adsorption [292]. Particularly effective are MOFs designed with pore sizes that align with the kinetic diameter of CO_2 molecules, coupled with polar functional groups within the pores (e.g., –OH, –N=N–, $–NH_2$, and –N=C(R)–) that can interact with CO_2 quadrupole moment, thereby enhancing adsorption capacity [292]. MOFs are categorized into two primary types: rigid, which maintain a stable framework and permanent porosity similar to zeolites, and flexible (dynamic), which possess an adaptable framework that responds to external stimuli such as guest molecule introduction, temperature, and pressure changes [293].

Studies have shown that pressure and temperature significantly influence CO_2 adsorption in MOFs [292]. Specifically, at low pressure conditions, the capacity for CO_2 adsorption is primarily determined by the heat of adsorption, indicating the importance of interaction strength between CO_2 molecules and the MOF surface. Conversely, at high pressure conditions, the CO_2 adsorption capacity is more reliant on the specific surface area of the MOF, suggesting that the physical availability of adsorption sites becomes the limiting factor for adsorption capacity.

With reference to the thermal stability of MOFs, it is notably improved by increasing the number of linkers attached to each node, due to the stronger bonds formed between nodes and linkers [290]. Utilizing trivalent linker cations with oxyanion terminations, such as Al(3), Zr(3), and Ti(3), known for their high bond enthalpy and dissociation energy, instead of the traditional carboxylate groups and divalent cations, also contributes to enhanced thermal stability [294]. Similarly, the hydrothermal stability of MOFs is significantly influenced by the bond strength between the node and linker, with MOFs featuring bonds with high heat of formation showing greater resistance to moisture. This stability can further be improved by surface modifications, such as introducing hydrophobic functionalities by substituting hydrogen atoms in ligands with fluorine [295]. Mechanical stability, characterized by factors like elastic moduli, rigidity, and compressibility, is dictated more by the physical structure than the chemical properties of MOFs. Increased porosity, although advantageous for CO_2 adsorption, tends to reduce mechanical stability [290,292]. Strategies to enhance MOFs mechanical strength include filling them with solvents, increasing the coordination number of metals, and shortening linkers [290,292].

The CO_2 adsorption capacity and selectivity in MOFs can be improved through various approaches, including adjusting pore sizes and shapes, surface functionalization of pores, and incorporating heterocyclic ligands, amino groups, phenolic hydroxyl groups, alkaline carbonates, and nitro groups [296]. Additionally, combining MOFs with carbon-based substances like graphene-related materials (GRMs) has shown to enhance their adsorption properties [297].

However, the stability of MOFs in the presence of moisture is a critical factor for their practical application in CO_2 capture. Developing MOFs that are resistant to water presents a significant challenge in the field. Hydroxo-functional groups play a crucial role in improving CO_2 capture performance under humid conditions [298]. The μ3-O groups, in particular, form strong interactions with water molecules adsorbed inside the MOF pores, due to water high polarity. This interaction facilitates an effective trapping mechanism for CO_2, enhancing its capture due to the confinement effect within the MOF structure. Sanchez-Gonzalez et al. [299] have introduced an environmentally friendly MOF based on Mg(II), showcasing exceptional storage capabilities. This MOF is not only capable of reversibly adsorbing CO2 but also demonstrates remarkable thermal stability, withstanding temperatures up to 450˚C. This breakthrough underscores the potential of designing MOFs that combine water resistance with high thermal stability, offering promising avenues for advancing CO_2 capture technologies in practical settings.

In addition to their vulnerability to water, MOFs are susceptible to contamination by H2S, SOx, and NOx, even in trace amounts [7]. These substances are adsorbed preferentially, occupying adsorption sites and necessitating their removal from flue gas prior to its entry into the adsorption bed to prevent significant reductions in CO_2 adsorption efficiency.

While MOFs demonstrate significant potential for post-combustion CO_2 capture, their production and application costs remain higher than those of commercially available sorbents [290]. Presently, the development and examination of MOFs, particularly those using costly linkers, are confined to laboratory settings (milligram scale) [64]. The use of expensive and potentially toxic solvents in current MOF synthesis methods poses challenges for upscaling to large-scale, environmentally friendly applications [64]. Consequently, the ability to molecularly tailor MOFs must be balanced with efforts to reduce their synthesis costs and environmental footprint [290].

Table 20. CO_2 adsorption performances of MOFs with corresponding process conditions.

Sorbent	Surface area $m^2\ g^{-1}$	T K	P_{CO_2} atm	Adsorption capacity $mmol\ g^{-1}$	Ref.
HKUST-1	680	298	0.15	1.14	300
MOF-508	323	323	0.10	0.10	301
HKUST-1	1800	298	0.10/1.00	0.93/5.72	302
Fe3O4@HKUST-1	830	298	0.10/1.00	0.45/3.51	302
MOF-2	345	298	1.00	0.80	303
IRMOF-11	2096	298	1.00	1.91	303
MOF-74	816	298	1.00	4.45	303
$Cu_3(BTC)_2$	1781	298	1.00	4.08	303
MOF-505	1547	298	1.00	3.42	303
Cu-BTC	1594	308	0.25/1.00	2/4	304
HKUST-1	-	295	0.10	0.62	305
IRMOF-1	1892	298	0.10	0.08	305
Ni\DOBDC	936	298	0.10	4.07	305
Co\DOBDC	957	298	0.10	2.81	305
ZIF-8	1135	298	0.10	0.12	305
MIL-47	600	298	0.10	0.18	305
UMCM-1	4034	298	0.10	0.09	305

3.2.2. Chemical sorbents

Chemical adsorption of CO_2 utilizes its linear, non-polar molecular structure, which consists of a positively charged carbon atom situated between two negatively charged oxygen atoms. This process depends on the nucleophilic nature of chemical functional groups on the adsorbent surface that can chemically bond with the CO_2 molecule. To address the challenges of low CO_2 adsorption capacity and selectivity, especially at lower CO_2 partial pressures and in the presence of competitive water vapor adsorption, the chemical functionalization of adsorbents has been suggested. Introducing various types of amines into porous support materials is one such strategy to enhance their performance under these conditions [226,306].

When designing amine-functionalized adsorbents for CO_2 capture, a variety of support materials can be employed:

- Mesoporous Silicas [307] - Mesoporous silicas, such as SBA-15 and MCM-41, are known for their well-ordered pore structures, high surface areas, and tunable pore sizes. The uniform pore distribution and large surface area facilitate high amine loading and efficient CO_2 capture. The stability and inert nature of silica make it an ideal support material for long-term use.
- Mesoporous Carbons [308] - Mesoporous carbons, exhibit high surface areas, well-defined pore structures, and excellent thermal and chemical stability and can support high amine loading. Their robust nature allows for repeated use and regeneration in CO_2 capture processes.
- MOFs [309] - MOFs have exceptionally high surface areas and adjustable pore sizes, which can be fine-tuned for specific applications. Their diverse chemistry allows for various amine-functionalization strategies, enhancing their CO_2 adsorption capacities.

- Porous Polymers [310] - Porous polymers, such as hypercrosslinked polymers (HCPs) and covalent organic frameworks (COFs), are characterized by their high porosity, low density, and chemical versatility. These materials can be easily modified with amine groups, offering high CO_2 adsorption capacities. Their lightweight nature and structural flexibility make them attractive for various applications, including gas separation and storage.
- Aerogels [311,312] - Aerogels are highly porous, low-density materials with a three-dimensional network of interconnected nanoparticles. Among this class of materials, silica aerogels are recognized for their ultra-lightweight structure, extremely large surface areas and high porosities, which facilitate gas diffusion and amine loading. Additionally, silica aerogels exhibit excellent thermal stability and low thermal conductivity, making them suitable for diverse environmental conditions and applications requiring thermal management.
- Zeolites [313] – Zeolites can offer well-defined crystalline structure (i.e. precise pore sizes) and ion-exchange capabilities. Their peculiar framework can, therefore, be modified to enhance amine functionalization and improve CO_2 capture efficiency.

Each of these support materials offers unique properties that can be leveraged to improve the performance of amine-functionalized adsorbents. The choice of support material depends on specific application requirements, such as adsorption capacity, selectivity, stability, and ease of regeneration. By carefully selecting and optimizing these materials, researchers can develop highly efficient adsorbents for CO_2 capture and other gas separation processes.

Amine-functionalized adsorbents fall into two categories based on the amine-support interaction: amine-impregnated sorbents, with polymeric amines weakly attached through simple physical mixing of support, amines, and solvents (impregnation method); and amine-grafted sorbents, where small amine molecules or polymeric amines are covalently bonded to the support either during or after synthesis (grafting method). The impregnation method is straightforward, while grafting involves chemically attaching amines to the surface [314]. The CO_2 adsorption capabilities of both types are summarized in Table 21.

Amine-impregnated sorbents often offer greater capture capacities than their grafted counterparts but are hindered by their poor durability through multiple adsorption/desorption cycles due to amine leakage and diffusion challenges, which arise from the greater amine loadings achieved through impregnation [226]. In contrast, amine-grafted sorbents demonstrate enhanced stability through numerous adsorption/desorption cycles, generally achieving faster adsorption rates that can surpass those of some commercial adsorbents like zeolite 13X [315].

Within the domain of amine-functionalized sorbents, significant focus is placed on employing polymeric amines such as polyethylenimine (PEI), polypropylenimine (PPI), polyallylamine (PAA), amino dendrimers, polyaniline, and hyperbranched polyamines. These polymers, featuring amine groups within their structure, are highly sought after for their dense amine content [316]. The preference leans towards polymers enriched with primary and secondary amine groups for their superior efficiency in CO_2 adsorption under both dry and moist conditions [317]. Specifically, PEI-based sorbents, recognized for their high primary amine density, affordability, widespread availability, and stability up to 90˚C, have been the subject of thorough investigation [226].

PEI stands out for its adaptability with numerous supports including siliceous materials (like fumed and precipitated silica, SBA-15, MCM-41, mesoporous siliceous foams, silica gels), carbon-based substances (activated carbons, CNTs, fullerenes, graphene), alumino-silicates (geopolymers), clays, MOFs, alumina, porous polymers, and zeolites [226]. Structurally akin to PEI, PPI is known to boost CO_2 adsorption capabilities but faces commercial hurdles due to monomer synthesis complexities and lengthy polymerization processes [226]. Sorbents supported by PAA often surpass those backed by PEI, offering enhanced oxidative and thermal resilience alongside stronger amine-to-support bonds [226]. Polyaniline, notable for its easy synthesis, nanostructured forms with high surface area, and superior thermal durability, shares these advantages [226]. Dendritic polymers like amino dendrimers and hyperbranched polyamines also deliver outstanding results owing to their unique structure and high density of amine/nitrogen groups [226].

Similar to other CO_2 adsorbents, the efficiency of amine-functionalized materials in capturing CO_2 is significantly affected by flue gas impurities. NO_x and SO_x, in particular, can form stable salts with amines, leading to a considerable reduction in CO_2 adsorption capacity [7]. Additionally, the environmental ramifications of large-scale production and application of amine-functionalized adsorbents require thorough evaluation before their implementation in actual post-combustion CO_2 capture settings [64].

Table 21. CO_2 adsorption performances of amine-impregnated and grafted sorbents with corresponding process conditions (P_{CO2} and T).

Support	Amine (loading %wt)	T K	P_{CO2} atm	Adsorption capacity mmol g^{-1}	Ref.
	Impregnated				
MCM-41	PEI (50)	348	0.10	2.10	318
MCM-41	PEI (50)	348	0.13	2.84	319
SBA-15	PEI (50)	348	0.15	1.95	320
SBA-15	TEPA (66)	358	0.15	2.15	152
Beta-zeolite	TEPA (38)	303	0.10	2.08	321
Zeolite 13X	MEA (25)	348	0.15	0.45	322
Activated carbon	PEI (40)	298	1.00	1.98	323
PE-MCM-41	DEA (76)	298	0.05	3.00	324
Al_2O_3	DETA (40)	330	1.00	1.50	325
PMMA beads	DBU (30)	338	0.10	2.34	326
SBA-15	TEPA (30)+DEA (20)	348	0.05	3.77	327
	Grafted				
Silica gel	AP (1.26)	323	1.00	0.89	328
Silica gel	APTES (20)	298	0.12	0.68	329
PE-MCM-41	TRI (7.90)	323	0.10	1.59	330
SBA-16	AEAPS (3.06)	333	0.15	0.73	331
SBA-15	APTES (2.56)	338	0.10	0.45	332
Zeolite ITQ-6	AP (1.26)	293	0.12	0.67	333
SBA-15	Aziridine polymer (9.78)	348	0.10	4.00	334

3.2.3. Summary - Adsorption

The choice of the most suitable sorbent material for CO_2 capture is influenced by a multitude of factors including adsorption capacity, selectivity, rate of adsorption/desorption, operational temperatures, stability (thermal, mechanical, and cyclic), moisture and impurity tolerance, and production costs. Pros and cons of all the examined sorbents are summarized in Table 22.

Zeolites and carbon-based materials like activated carbons exhibit the highest level of industrial readiness. Zeolites, both in natural and synthetic forms, are applicable for post-combustion CO_2 capture, with their performance dictated by their structural framework (namely Si/Al ration) and the nature and placement of cations within that framework. Natural zeolites offer cost benefits but limited CO_2 capture capabilities (primarily due to their low purity derived from variable chemical composition), whereas synthetic zeolites provide superior CO_2 adsorption at higher costs and are highly sensitive to moisture, necessitating drying steps before use.

Carbon-based adsorbents, excluding nanomaterials such as CNTs and graphene, are generally more affordable and easier to produce on a large scale. Their CO_2 adsorption is predominantly physical, driven by porosity, but the inclusion of heteroatoms can introduce chemisorption properties. Their hydrophobic nature makes them less susceptible to moisture than zeolites, though their CO_2 capture efficiency without functionalization tends to be lower.

MOFs stand out for their adjustable pore structures, exceptional surface area, and porosity, leading to significant CO_2 adsorption. They support a large number of structures, providing remarkable versatility in application. Their adsorption mechanisms involve both sieving effects and chemisorption, facilitated by interactions between CO_2 molecules and the MOFs metallic sites or functional groups. However, like zeolites, MOFs are vulnerable to moisture and impurities, which can compete with CO_2 for adsorption sites, and most MOFs are currently produced only at a laboratory scale or as fine powders when manufactured on a larger scale.

Amine-functionalized sorbents merge the advantages of solid adsorbents and solvents, providing high CO_2 capture capacity and relatively low production costs through the incorporation of nitrogen functionalities. The CO_2 adsorption mechanism varies with the type of amine and support material used, and challenges remain in evenly distributing amines across porous substrates without pore blockage and enhancing thermal and oxidative stability.

With reference to the sorbent regeneration, it can be achieved through temperature or pressure change, each approach carrying distinct advantages and drawbacks. TSA involves CO_2 adsorption at lower temperatures followed by high-temperature regeneration (150-200˚C), operating at low pressures and tolerating impurities like NO_x, SO_x, and water vapor. It integrates easily into existing plants but suffers from energy inefficiency due to repeated heating/cooling cycles. PSA/VSA operate at low temperatures, avoiding these energy penalties but are energy-intensive due to pressurization and vacuum processes, especially below 100 mbar, which is impractical for industrial use. A promising solution is Vacuum Temperature Swing Adsorption (VTSA), combining TSA and VSA benefits to reduce temperature requirements, save energy with low-grade waste heat, and enhance CO_2 purity and recovery.

In conclusion, CO_2 capture through adsorption offers a versatile and promising avenue for mitigating carbon emissions. A diverse array of sorbent materials and regeneration strategies are available, each with its own set of advantages and challenges that must be carefully addressed. Ongoing research and innovation in sorbent materials, optimization processes to tackle energy consumption, purity maintenance, and complexity issues, as well as exploration of hybrid approaches, hold the potential to enhance the efficiency and feasibility of adsorption-based CO_2 capture.

Table 23 provides a synthesized overview of recent R&D projects focused on CO_2 adsorption for post-combustion capture. Specifically, it details project focuses, contractors, durations, and the most relevant operational and economic parameters. Clearly, compared to absorption (Table 16), adsorption technologies are still in the development phase. They are focusing on material optimization and bench-scale testing, indicating a need for further research before they can be implemented on a large scale [2,335].

Table 22. Pros and cons of different classes of sorbents

Adsorbents	Advantages	Disadvantages
Activated carbons	• Broad availability of raw material • Limited costs • High specific surface area • High thermal stability • Moderate resistance to water • Straightforward regeneration process	• Limited CO_2 adsorption capacity • Brittleness • Poor selectivity
Carbon nanomaterials	○ Distinct pore size distribution ○ High specific surface ○ High CO_2 adsorption capacity ○ Chemical and physical stability ○ Fast adsorption kinetics ○ Moisture resistance	○ Costly and complex production ○ Poor selectivity
Synthetic zeolites	• Adjustable textural properties • High surface area and porosity • High CO_2 adsorption capacity • High thermal stability	• Costly production • Sensitivity to moisture/impurities
Natural zeolites	○ Wide availability ○ Low costs	○ Variability in chemical composition affects purity ○ Limited adsorption capacity
MOFs	• Adjustable textural properties • Large surface area and porosity • Notable CO_2 adsorption capacity	• Costly and complex production • Sensitivity to moisture/impurities • Moderate/low thermal stability
Amine-functionalized materials	○ Broad range of supporting structures ○ High CO_2 adsorption capacity ○ Small production cost ○ Effective regeneration	○ Amine thermal/oxidative degradation ○ Pore clogging

Table 23. Recent R&D projects focused on CO_2 adsorption processes for post-combustion capture [2,335].

Sorbent	Focus	Contractor	Duration	CO_2 Loading [mmol g^{-1}]	P [atm]	T [°C]	Cost of manufacturing [$ kg^{-1}]	Capture cost [$ tCO_2^{-1}]	CO_2 Recovery [% vol.]	CO_2 Purity [%]	TRL
Functionalized mixed matrix polymer	Novel adsorbents for MTSA[1]	TDA Research, Inc.	2022-2025	>0.9	0.68	60-90	-	-	>95	-	4 Bench Scale
Amine structured	Novel adsorbents for NGCC	Cormetech, Inc.	2022-2025	-	-	-	-	-	>95	>95	5 Bench Scale
Amine-functionalized resins	Novel adsorbents for VCSA[2] for coal and TSA for NGCC	TDA Research, Inc.	2018-2024	2.5-2.8	0.04-0.07 (PCO2)	-	-	28.9	-	-	4 Bench Scale
Microporous materials	Novel adsorbents development	InnoSepra, LLC	2019-2024	-	0.10-0.12 (P_{CO2})	25-100	-	-	-	-	4 Bench Scale
TiO_2/Al_2O_3 on zeolite 13X	Reduction in CO2 capture cost and energy penalties (PSA)	Rensselaer Polytechnic Institute	2019-2023	0.96-1.2	0.15 (P_{CO2})	20	3.6	30	90	95	4 Bench Scale
SIFSIX-2-Cu-I MOF	Novel adsorbents for VCSA	TDA Research, Inc.	2019-2023	1	30	30	-	30.7-36.4	90	95	3 Bench Scale
Bi-layer laminated MOFs	Optimization of novel adsorbents	Electricore, Inc	2019-2022	1.5-2.5	1-1.1	40-50	100-200	-	90	90	4 Bench Scale

[1]MTSA = microwave assisted thermal swing adsorption; [2]VCSA = Vacuum Concentration Swing Adsorption (VCSA)

3.3. Membranes

Membrane separation offers a method for selectively isolating CO_2 from flue gases using permeable or semipermeable materials through mechanisms such as solution-diffusion transport, molecular sieving, surface diffusion, and Knudsen diffusion (Fig. 6) [64].

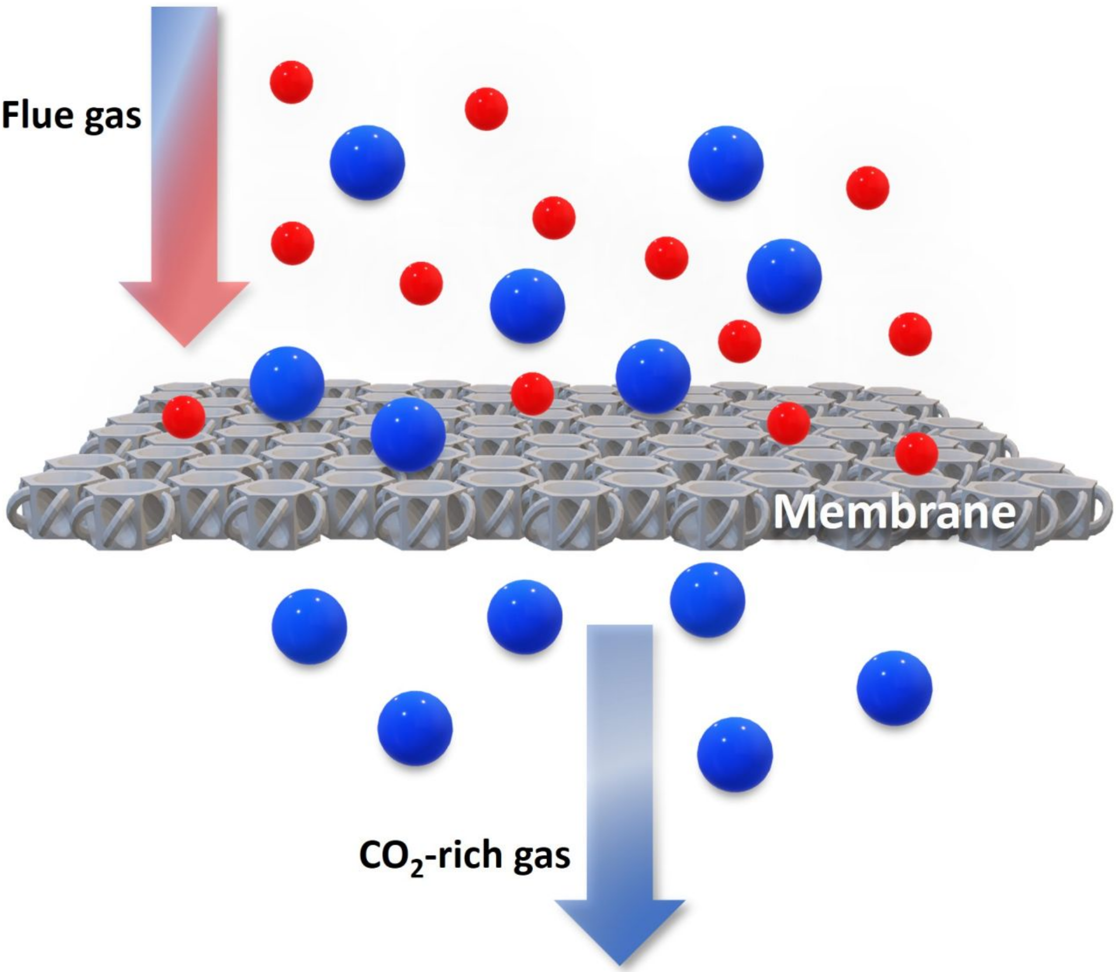

Fig. 6. Scheme of CO_2 separation by membrane.

This technique, driven by pressure and/or concentration gradients, is particularly suited and more energy-efficient for pre-combustion scenarios, where CO2 partial pressure and concentration are higher, compared to post-combustion settings characterized by lower CO2 levels [62,336]. Membrane systems, lacking moving parts, can be seamlessly integrated into existing facilities [64]. Nonetheless, achieving high CO_2 removal purity (> 95% vol.) in a single-stage process is challenging due to limitations in transmembrane pressure ratios and selectivity, necessitating multi-stage configurations for optimal performance [336,337]. In this framework, enhancing CO_2 flux and recovery up to 90% vol. is possible through carefully designed recycling streams within the membrane processes [338]. However, it must be noted that membrane separation cost-effectiveness is more pronounced at smaller scales (typically below 300 MT $year^{-1}$) due to the limitations in the cost benefits of extensive modularization [339]. Indeed, for small-scale post-combustion CO_2 capture facilities processing under 100 t of flue gas daily with recovery rates below 40% vol. CO_2, membrane separation techniques may offer a cost-effective alternative to absorption methods [340]. The effectiveness of membranes in post-combustion carbon capture was also highlighted by Baker et al. [341], especially noting their capacity for significantly reducing energy consumption compared to traditional amine absorption techniques for CO_2 concentrations over 10%.

For a membrane to be effective in CO_2 separation, it must exhibit several critical qualities: high permeability and selectivity for CO_2 molecules, minimal thickness to facilitate efficient gas transfer, and robust physical, chemical, mechanical, and thermal stabilities [342]. These attributes are essential for a membrane to be considered viable for industrial applications, ensuring it can withstand operational conditions while delivering optimal separation performance. The product of selectivity and permeability is defined as the separation factor or separation power, a key indicator of membrane efficiency. Ideally, a membrane should exhibit high separation power. However, selectivity and permeability typically exhibit an inverse relationship, making it challenging to optimize both simultaneously within the same membrane [27]. Adjustments and enhancements through doping, controlling pore size and porosity, along with process engineering, surface treatments, and functionalization, are strategies to improve membrane characteristics. For any type of membranes, transport is driven by a pressure difference or chemical potential difference across the membrane.

The greater this pressure differential and the thinner the membrane, the higher the mass flux or permeability. Thus, there is a push to develop membranes as thin as possible while ensuring they possess sufficient mechanical integrity to sustain the required pressure differential. Besides, for CO_2 separation membranes to reach commercialization, several key challenges must be addressed, such as resistance to plasticization, robustness against thermal and chemical stresses, long-term durability, and cost-effectiveness [343].

From this discussion, it is evident that a critical consideration in membrane processing methods is the trade-off between high material preparation costs and the efficient separation and capture costs. To address this trade-off, several strategies can be employed:

- Material optimization - The development of advanced membrane materials with higher CO_2 selectivity and permeability can reduce the overall membrane area required, thus lowering the initial material costs. Innovations in polymer science and the incorporation of nanomaterials (such as graphene oxide and carbon nanotubes) can enhance membrane performance while potentially reducing costs [344].
- Economies of scale - Scaling up membrane production can lead to cost reductions through economies of scale. Large-scale manufacturing processes can benefit from improved efficiencies and reduced per-unit costs, making high-performance membranes more affordable for industrial applications [345].
- Hybrid systems - Integrating membrane processes with other CO_2 capture technologies, such as absorption or adsorption, can optimize the overall system performance and cost-efficiency. Hybrid systems can leverage the strengths of each technology, reducing the reliance on costly high-performance membranes alone [346].

- Process optimization - Enhancing the operational efficiency of membrane systems through process optimization can minimize energy consumption and operational costs. Advanced control systems and process integration techniques can improve the overall performance and economic viability of membrane-based CO_2 capture [347].
- Sustainable materials - Utilizing more sustainable and cost-effective materials for membrane fabrication can significantly reduce preparation costs. Research into alternative materials, such as bio-based polymers or recycled materials, can provide environmentally friendly and economically attractive options [348].
- Life Cycle Assessment (LCA) - Conducting comprehensive Life Cycle Assessments (LCA) of membrane materials and processes can identify cost-saving opportunities and environmental benefits. LCA can help in selecting materials and designing processes that balance preparation costs with long-term operational efficiency and sustainability.

By employing these strategies, the trade-off between material preparation costs and efficient separation and capture costs in membrane processing can be effectively managed, paving the way for the broader adoption of membrane technologies in CO2 capture applications [349].

A wide variety of membranes have been developed and engineered for CO_2 capture, which can be primarily classified into organic (polymeric) membranes and inorganic (non- polymeric) membranes. These membranes offer a diverse range of solutions customized to meet various capture requirements and efficiencies [350,351].

3.3.1. Organic (Polymeric) membranes

Polymer-based materials stand out among various membrane materials due to their inherent advantages in cost-effectiveness, processability, and diversity [352]. The solution-diffusion and facilitated transport mechanisms are widely recognized and utilized as guiding principles for the development of new polymer designs. Polymers such as polyacetylene, polyaniline, polyetherimides, polycarbonates, poly(phenylene oxide), poly(ethylene oxide), and polysulfone have been explored for post-combustion CO_2 capture applications. By tailoring the polymer preparation and chemical composition, these membranes offer highly adjustable permeability and selectivity. Nevertheless, issues like swelling and plasticization resulting from CO_2 adsorption are significant and must be addressed in the development process. Table 24 provides a summary of newly developed polymeric membrane materials.

The primary mechanism underpinning gas separation in polymeric membranes is the solution-diffusion process [27]. In this process, certain gas molecules are selectively absorbed or adsorbed into the polymer matrix voids and diffuse through the channels between polymer chains. Meanwhile, other gas mixture components are less influenced as they pass through the membrane. This selective permeation is due to the varying strengths and rates at which different gaseous species interact with the polymer matrix, affecting their absorption, adsorption, and diffusion [27]. Consequently, achieving absolute selectivity for a specific gas within a mixture using polymeric membranes is nearly unattainable. There is an inherent compromise between selectivity and flux (or permeability). Polymers with high permeability, such as those with a rubbery or elastomeric open structure, tend to offer high permeability at the expense of selectivity, whereas glassy or semi-crystalline polymers provide higher selectivity but at lower permeability rates. Thus, to reach the desired level of gas enrichment, the gas mixture often requires multiple re-circulations through the membrane system, indicating that polymeric membranes cannot achieve complete CO_2 separation on their own [27].

The challenge in selectively separating CO_2 from N_2-rich mixtures stems from their similar physicochemical properties. Notably, the kinetic diameters of CO_2 (0.33 nm) and N_2 (0.364 nm) are very close, complicating the use of size exclusion as a separation method. Thus, advancements in

CO_2-selective polymeric membranes are expected to focus on two main strategies: increasing CO_2 solubility within the membrane and enhancing the rate of CO_2 diffusion through the polymer matrix. This involves the meticulous design and creation of polymers with specific void spaces or channels, tailored side groups, structures, and chain conformations to boost both selectivity and permeability. Alterations in chain dynamics can modify the void spaces in the polymer matrix, facilitating the diffusion of gaseous species. To improve selectivity and permeability, mixed-matrix membranes, which incorporate various polymers with complementary properties or inorganic adsorbents into polymer matrices, have been investigated [353].

Beyond the fundamental mechanisms of separation, capturing CO_2 with polymeric membranes from power plant emissions introduces additional hurdles [27]: i) the dilute nature of CO_2 in flue gases necessitates a vast membrane area to treat the large volumes emitted; ii) the membrane must also be chemically robust to withstand the different contaminants found in emissions from coal-powered facilities; since H_2O tends to be separated more efficiently than CO_2 by most polymer membranes (due to its smaller kinetic diameter, 0.265 nm), it is necessary to dry the flue gas stream prior to membrane separation.

The thermal and mechanical robustness of polymeric membranes is also of paramount importance. The high temperatures of flue gases exiting power plants exceed the operational temperature limits of polymeric membranes, necessitating cooling of the flue stream to below 100°C to prevent thermal degradation of the membrane polymeric structure [27]. Concurrently, to facilitate CO_2 separation, the flue gas must be pressurized to create a pressure differential across the membrane, enabling CO_2 collection on the permeate side. Advances have been achieved in enhancing both thermal tolerance and separation efficiency. For instance, a polymeric membrane composed of poly(vinyl alcohol) (PVA) and poly(acrylic acid) (PAA) copolymer, integrated with the selective CO_2 carrier 2,3-

diaminopropionic acid hydrochloride (DAPA-HCl), has shown to not only increase CO_2/N_2 selectivity (up to 700) but also improve CO_2 permeability (2×10^{-4} mol m^{-2} kPa^{-1} s^{-1}) under conditions of elevated temperatures (160 ˚C) and pressures (600 kPa) [354]. However, these necessary thermal and mechanical adjustments introduce additional costs and energy demands to the capture process, impacting the overall efficiency of the power plant and the cost-effectiveness of CO_2 capture. A recent study that compared CO_2 capture from post-combustion sources using polymeric membranes against amine absorption methods found that, from an energy perspective, membrane technology is less competitive. Specifically, the energy consumption for capturing CO_2 using polymeric membranes is estimated at around 8 MJ $kgCO_2^{-1}$, whereas amine absorption requires roughly 4 MJ $kgCO_2^{-1}$, highlighting a significant difference in energy efficiency between the two approaches [355].

Table 24. CO_2 capture performances of polymeric membranes used for post-combustion CO_2 capture with corresponding process conditions.

Material	Tickness μm	CO_2 GPU[1]	N_2 GPU[1]	CO_2/N_2 Selectivity	P atm	T ˚C	Ref.
PIM-1	62	1203	5.3	22.9	2	298	[356]
PIM-EATB	180	43	3.2	13.3	1	298	[357]
PIM-Trip-TB	132	74	4.8	15.4	1	298	[357]
TPIM-2	52	38	1.6	23.4	2	298	[358]
PTMSP	60	425	4.1	9.4	2	298	[359]
6FDA-ABS/DAM(5)(DAM)	34	22	0.2	10.3	2	298	[360]
HFBAPP/DAB186C/6FDA	110	1.3	0.4	75.13	20	308	[361]
PIM-SBF	180	77	4.4	17.7	-	-	[362]
PIM-BTrip	160	134	7.4	18.1	-	-	[363]
PIM-TMN-Trip	166	318	21.3	14.9	-	-	[363]
PIM-HMI-Trip	135	327	19.0	17.3	-	-	[363]
TFM-BTrip	176	190	10.4	18.4	-	-	[363]
DTFM-BTrip	112	380	26.8	14.2	-	-	[363]
DM-BTrip	114	193	8.9	21.6	-	-	[363]

[1] Normalized permeability to permeance by using the unit of GPU for a better comparison. GPU = 3.35×10^{-10} mol m^{-2} s^{-1} Pa^{-1}.

3.3.2. Inorganic (non-polymeric) membranes

Inorganic membranes can be fabricated from a range of carbon nanostructures, including carbon nanotubes and hollow fibers, or from various oxides like zeolites, silica, alumina, and metal-organic frameworks (MOFs) [27,352]. Another class includes electrochemical molten membranes, which are primarily composed of ion-conducting eutectics made from alkali-metal carbonates [27,352]. Table 25 summarize the performances of newly developed inorganic membranes.

The effectiveness of gas separation using these inorganic membranes is influenced by different elements such as the heat of adsorption, reflecting the strength of binding between gas molecules and the membrane surface, the compatibility of gas molecule sizes with the membrane pore size, the membrane porosity and tortuosity, the thickness of the membrane active layer, the operating temperature, and the membrane specific surface area [27]. Specifically, the transport of gaseous molecules through these membranes can be categorized into four primary mechanisms: Knudsen diffusion, occurring when the pore size is similar to the molecular diameter of the gas species; surface diffusion, where certain gases exhibit a preferentially higher surface diffusion coefficient; capillary condensation happening within the membrane pores; molecular sieving, which operates on the size exclusion principle. Additionally, the diffusion through intersecting channels, as found in zeolite structures, or between cages in MOFs, is also recognized as a significant transport mechanism [364].

Inorganic membranes typically exhibit greater stability compared to polymeric counterparts, positioning them as preferred options for separating gas mixtures, particularly in severe operating conditions [352]. Indeed, inorganic membranes can be designed as self-supporting structures or as thin films or dispersions applied to inert or catalytically active substrates, which offer mechanical stability and durability. These support materials often consist of porous ceramics like silica, alumina, titania, zirconia, as well as porous glasses or metals such as stainless steel. The chemical and thermal robustness of inorganic materials make them suitable for CO_2 capture from the exhaust gases of power plants. A key benefit is their ability to operate across a broad temperature range, from ambient conditions to over 1000°C, enhancing the thermal management of direct flue gas feeds from power plants and ensuring efficient CO_2 capture. Although inorganic membranes can count on chemical stability and resistance to contaminants in coal-derived flue gases, the presence of water in the flue gas can negatively affect the adsorption and selective transport capabilities of many inorganic membranes. While these materials can endure high operational temperatures, their CO_2 selectivity tends to decrease with increasing temperatures, making the ideal operating temperature for most inorganic and composite membranes below 200°C. For instance, zeolite Y-based microporous membranes exhibit high permeability (10^{-6} mol m^{-2} Pa^{-1} s^{-1}) and CO_2/N_2 selectivity (up to 500) at room temperature, but this performance significantly declines above 200°C, where selectivity shifts to a diffusion-controlled regime, and the disparity between CO_2 and N_2 diffusion coefficients sharply decreases [365].

Adjusting the adsorption characteristics of the membrane through surface engineering [366] and functionalization, particularly with surface amine groups [367], can enhance its CO_2 adsorption capacity and surface diffusion. Likewise, employing micro- and nano-structuring techniques can refine the pore size and porosity distribution, optimizing the membrane for selective transport of specific gases, such as CO_2. However, the challenge of achieving high selectivity and permeability (or gas flux) remains a significant barrier to the advancement of microporous inorganic membranes.

Table 25. Inorganic membranes used for post-combustion CO_2 capture.

Material	Inlet %CO_2 in N_2	CO_2 permeance GPU	CO_2/N_2 selectivity	P atm	T ˚C	Ref.
Na-Y	100	450	100	3.00	303	[368]
SAPO-34	15	4120	170	1.00	243	[369]
SAPO-34	100	3500	32	2.41	295	[370]
ZSM-55	100	864	44	3.00	298	[371]
SSZ-13	100	954	32	3.03	298	[372]
AIPO-18	100	1880	45	3.03	298	[373]
IRMOF-1	87.4	614	410	5.05	298	[374]
ZnTCPP	100	2070	33	1.00	308	[375]
CAU-1	90	3995	22.7	1.00	298	[376]

3.3.3. Summary - Membranes

Membrane separation technology provides a nuanced approach to CO_2 capture, leveraging the unique properties of different membrane materials, polymeric (organic) and inorganic (non-polymeric), to achieve selective gas separation. Their capture mechanism relies on the ability to exploit solution-diffusion transport, molecular sieving, surface diffusion, and Knudsen diffusion mechanisms, driven by pressure and/or concentration gradients This efficacy of this separation method hinges on straightforward operation, which typically does not involve chemical reactions, moving parts, use hazardous chemicals or significant temperature variations to separate CO_2. Moreover, membrane technology is recognized for its scalability and compact/modular design, which can be linearly scaled up or down to meet specific needs while maintaining a minimal footprint. Both polymeric and inorganic membranes offer unique advantages and face distinct challenges in CO_2 capture applications. A summary is provided in Table 26.

Polymeric membranes are praised for their cost-effectiveness, versatility, customizable properties and ease of processing, attributes that make them particularly appealing for post-combustion CO_2 capture applications. These membranes operate principally through solution-diffusion processes, selectively absorbing or adsorbing CO_2 into the polymer matrix. Despite their widespread use, polymeric membranes confront significant hurdles, such as susceptibility to swelling and plasticization from CO_2 adsorption, which can compromise their structural integrity and separation efficiency. Additionally, the inherent trade-off between selectivity and flux in these materials complicates efforts to achieve optimal gas separation performance. The challenge is further intensified when separating CO_2 from N_2 (i.e. from flue gases that are N_2-rich mixtures) due to their similar physicochemical properties, necessitating sophisticated polymer designs to enhance both CO_2 solubility and diffusion rates.

In contrast, inorganic membranes are recognized for their superior stability and resilience, qualities that make them well-suited for operation under severe conditions. These membranes can withstand a broad spectrum of temperatures, offering enhanced thermal management for direct flue gas feeds and ensuring effective CO_2 capture from power plant exhausts. However, their application is not without challenges; the presence of water vapor in flue gases can impair their adsorption and transport capabilities, and the quest for high selectivity and permeability remains a formidable barrier to their widespread adoption. Despite these obstacles, inorganic membranes robustness against chemical and thermal stresses presents a compelling case for their use in CO_2 separation endeavors.

As the field evolves, the continuous refinement of membrane materials and separation mechanisms will undoubtedly enhance their efficacy and feasibility for large-scale CO_2 capture. Specifically, the review has highlighted a critical trade-off in membrane processing methods between high material preparation costs and efficient separation and capture costs. To manage this trade-off, strategies include optimizing materials, leveraging economies of scale, integrating with other CO_2 capture technologies, optimizing processes, using sustainable materials, and conducting Life Cycle Assessments (LCA). By implementing these strategies, membrane technologies can achieve broader adoption for CO_2 capture and, with advancements towards higher Technology Readiness Levels (TRLs), are poised to become crucial components in sustainable and efficient carbon management solutions.

Table 27 provides a synthesized overview of recent R&D projects focused on CO_2 absorption for post-combustion capture. Specifically, it details project focuses, contractors, durations, and the most relevant operational and economic parameters. These projects encompass a quite wide range of TRLs, reflecting both early-stage research and advanced development phases. By focusing on performance metrics and cost efficiency, they demonstrate significant potential for scalable and effective CO_2 capture solutions [2,335].

Table 26. Membrane types: advantages and disadvantages.

	Pros	Cons
Polymeric (organic)	○ Cost-effective, versatile, and easily processable. ○ Adjustable permeability and selectivity through polymer design.	○ Susceptible to swelling and plasticization from CO_2. ○ Inherent compromise between selectivity and permeability. ○ Challenges in separating CO_2 from N_2 due to similar physicochemical properties.
Non-polymeric (inorganic)	○ Superior stability and suitable for severe conditions. ○ Can operate across a broad temperature range. ○ Robust against thermal and chemical stresses for power plant exhaust CO_2 capture.	○ Water vapor in flue gases can impair adsorption and selective transport. ○ Challenges in maintaining high selectivity and permeability at high temperatures. ○ Achieving high selectivity and permeability simultaneously is challenging.

Table 27. Recent R&D projects focused on membrane-based processes for CO_2 post-combustion capture [2,335].

Type	Focus	Contractor	Duration	CO_2 selectivity [-]	P_{CO2} normalized flux [GPU]	T [°C]	Cost of membrane material [$ m^{-2}]	Cost of installation and manufacturing [$ m^{-2}]	Capture cost [$ $tCO2^{-1}$]	CO_2 Recovery [%vol.]	CO_2 Purity [%]	TRL
Mixed matrix	Achieve high CO2 permeance	State University of New York	2019-2023	0.3 (H_2O) 50 (N_2) 0.5 (SO_2)	1500-2000	60	-	-	30	-	95	4 Bench-scale
Polymeric	Improve capture efficiency	The Ohio State University	2019-2023	1 (H_2O) 170 (N_2)	3500	57-77	20	40	40-41.5	>60-90	>95	4 Bench-scale
Polymeric	Retrofit polymeric membrane capture system	Membrane Technology and Research, Inc.	2019-2022	0.3 (H_2O) 50 (N_2) 0.5 (SO_2)	1000	30	10	50	57.64	90	>96	5-7 FEED[1]
Polymeric	Large pilot polymer membrane system	Membrane Technology and Research, Inc.	2018-2026	0.3 (H_2O) 50 (N_2) 0.5 (SO_2)	1000	30	-	50-100	-	70-75	99	6 Large pilot

[1] FEED = Front-end engineering design

3.4. Cryogenic separation

Cryogenic technology involves the extraction of CO_2 from other components through multiple stages of compression and cooling at very low temperatures (–100 ˚C to –135 ˚C) and high pressures (101 bar to 203 bar), resulting in high-purity liquid CO_2 suitable for transportation and storage (Fig. 7) [24]. Specifically, the process is carried out using equipment such as compressors, multi-stage heat exchangers, Joule-Thomson valves, and cold traps. Technologies used in cryogenic separation are primarily classified into three groups [377]: i) traditional cryogenic separation methods, such as liquid-vapor distillation, extractive distillation technologies, packed bed, external cooling loop; ii) innovative cryogenic separation methods, including solid vapor desublimation-based separation, cryocoolers (Stirling coolers), successive release and recompression, and centrifugal separation of condensed contaminants; iii) hybrid technologies that integrate both traditional and innovative techniques.

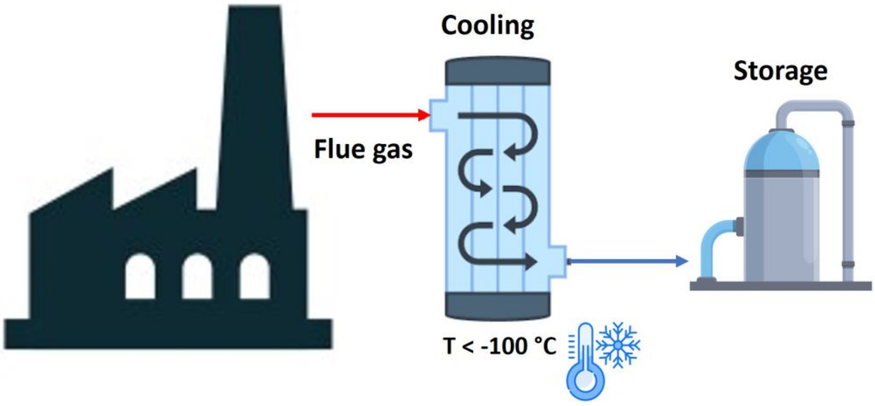

Fig. 7. Scheme of CO_2 cryogenic separation.

In post-combustion applications, cryogenic separation methods typically involve either CO_2 desublimation on heat exchanger surfaces followed by heating and pressurizing to liquefy CO_2 [378] or using packed beds for desublimation followed by regeneration with a fresh gas stream at elevated temperatures for CO_2 recovery [379]. The effectiveness in CO_2 capture and energy consumption of different types of cryogenic separation approaches are reported in Table 28.

Tuinier et al. [380] demonstrated that dynamically operated packed beds at a 600 MW coal-fired power plant achieved cost competitiveness with higher CO_2 avoidance costs at USD 126.50 tCO_2^{-1} avoided compared to amine scrubbing (USD 54.50 tCO_2^{-1} avoided) and membrane processes (USD 120 tCO_2^{-1} avoided), particularly when utilizing low-cost Liquefied Natural Gas (LNG) for cold energy supply [380]. Additionally, the cryogenic packed bed process can simultaneously separate H_2O and CO_2 from flue gas based on differences in their dew and sublimation points, thereby preventing operational issues such as clogging and large pressure drops. An effective strategy to reduce energy consumption in cryogenic CO_2 capture involves reusing waste cold energy from industrial sources like LNG through an external cooling loop. Jensen et al. [381] found that an external cooling loop cryogenic CO_2 capture process for a 550 MW coal-fired power plant achieved high purity liquid CO_2 (99.2% vol.) with low energy consumption (0.74 MJ $kgCO_2^{-1}$) using an internal CF4 refrigeration cycle to manage heat transfer from melting CO_2 to desublimating CO_2, supplemented by an external cooling loop using natural gas or other refrigerants. Anti-sublimation CO_2 capture, which involves frosting and defrosting CO_2 at atmospheric pressure in a low-temperature evaporator, is another efficient cryogenic process. Clodic et al. [382] estimated that anti-sublimation CO_2 capture from flue gas in a conventional pulverized coal-fired power boiler consumed 1.25 MJ $kgCO_2^{-1}$ for 90% CO_2 removal, representing a 21% lower energy penalty compared to MEA-based CO_2 capture.

The main benefits of cryogenic separation over other separation methods include: i) no need for chemicals or solvents, which eliminates recurring consumable costs; ii) no requirement for water supply or treatment; iii) removal of the solvent recovery step; iv) immediate elimination of water from the downstream inlet separator, preventing corrosion; v) provision of CO_2 at higher pressures, beneficial for enhanced oil recovery or sequestration; vi) potential production of natural gas liquids as valuable by-products; vii) no risk of foaming; viii) suitability for cold climates; and ix) cost-effectiveness for treating flue gases with high CO_2 concentrations. On the contrary, the main drawbacks of cryogenic technology are: i) potential process blockages, due to ice formation in CO_2 purification units or solid CO_2 formation on heat exchanger surfaces; ii) high energy demands for regeneration (due to extremely low temperatures and high pressures); iii) elevated CO_2 capture costs, attributed to significant pressure drops during operation and the need for costly steps to remove water from flue gas [383]; iv) need to remove water and other components like SO_x and NO_x before cooling in order to prevent blockages [2].

Table 29 presents a summary of recent R&D projects dedicated to CO_2 cryogenic separation for post-combustion capture. It outlines the specific focuses of each project, the contractors involved, the most significant operational and economic parameters. It is important to underline that the concentration of CO_2 in the source gas significantly impacts the temperature needed for effective condensation; a small decrease in CO_2 percentage can reduce the required temperature by approximately 20°C, highlighting the high energy demands at lower CO_2 concentrations [384]. Therefore, in terms of cost-effectiveness, cryogenic processes are more suitable when CO_2 concentrations and stream pressures are sufficiently high, which is typically the case in pre-combustion and oxyfuel combustion scenarios. Indeed, as clearly inferable from Table 29, commercial cryogenic CO_2 capture (TRL > 6) is only available for highly concentrated flue gases [377]. On the contrary, post-combustion cryogenic is still in the research and development testing phases [377].

Table 28. CO_2 capture performances of different cryogenic separation techniques with corresponding process conditions.

Type	Inlet $\%CO_2$ in N_2	Cold Energy Source	CO_2 recovery %	CO_2 purity %	Ref.
Packed bed	10	LNG*	99.5	-	385
External cooling loop	14	CF4[b] refrigerant and LNG	93.3	99.2	381
CryoCell	20-35	Chiller	34.0	-	386
Stirling cooler	13	Stirling cooler	85.0	-	387
Anti-sublimation	10	Mixed refrigerant	90.0	-	382

[a] LNG = Liquidfied Natural Gas; [b] CF4 = Carbon Tetrafluoride

Table 29. Recent R&D projects focused on membrane-based processes for CO_2 capture [2,335].

Type	Focus	Contractor	P [atm]	T [°C]	Efficiency [%]	TRL
Conventional distillation	CO_2/CH4	Buckeye, US	>20	<-20	>98	9
Hybrid cryogenics	Natural gas	ExxonMobil, US	39 - 42	-60 to -85	>90	6 - 7
Sprex	Natural gas	OTAL, France	70	-60 to -70	>90	6 - 7
Gas hydrate	Combustion flue gas	LREGH, China	10 - 25	> 0	-	6 - 7
Packed bed	Combustion flue gas	-	1	- 150	99	1 - 2
Condensed centrifugal	Combustion flue gas	-	> 31	-40 to -58	> 90	1 - 2
Stirling coolers	Combustion flue gas	-	1	- 90 to - 120	96	1 - 2

3.5. CO_2 separation technologies – Summary and comparison

In the quest for effective CO_2 separation in post-combustion scenarios, i.e. where CO_2 is captured after fossil fuels have been burned, an array of different technologies is available, each offering unique benefits and facing specific challenges. A comprehensive understanding and comparison of these technologies is crucial for selecting the most appropriate method for specific applications, considering factors such as CO_2 source, flue gas composition, and operational conditions. Table 30 outlines the main pros/cons and the key characteristics of the CO_2 separation techniques discussed in this review paper. In particular, it also provides a comparative analysis of the energy consumption costs associated with the different separation technologies.

Absorption

CO_2 capture via absorption processes utilizes gas dissolution in a liquid medium, divided into chemical absorption, involving solvent-chemical reactions with CO_2, and physical absorption, where CO_2 dissolves based on solubility. Each method provides a unique set of advantages, tailored to different industrial requirements and environmental goals.

Chemical Absorption - Chemical absorption is characterized by its use of solvents, such as amines like MEA and DEA, that react chemically with CO_2 to form a compound, facilitating its removal from gas streams. This method, standing as the most time-tested technique, is highly efficient, especially for capturing CO_2 from large point sources like power plants and industrial facilities. The energy consumption for chemical absorption is typically in the range of 3.5-4.0 MJ $kgCO_2^{-1}$. This high energy demand can significantly offset the environmental benefits of CO_2 capture, making it less attractive unless waste heat or other energy recovery methods are employed.

- Advantages

  - High Capture Efficiency: the high recovery efficiencies and the ability to achieve up to 98% CO_2 capture establish it as a cornerstone for industrial applications.
  - Adaptability: the versatility and efficiency enhancements possible through solvent optimization also count among its strengths. Indeed, the process can be tailored to specific industrial applications by choosing solvents with properties optimized for particular flue gas compositions and conditions.
- Disadvantages
  - Energy Intensive: The need to regenerate solvents (usually by heating) makes chemical absorption energy-intensive, potentially offsetting some of the environmental benefits of CO_2 capture.
  - Solvent Degradation and toxicity: Long-term use can lead to solvent degradation, especially in the presence of impurities like oxygen and sulfur compounds, requiring careful management and occasional replacement of the absorbing medium. Environmental and health risks can also arise from potential toxic by-products.
  - Operational Costs: Complex systems and continuous solvent regeneration increase operational expenses.

Physical Absorption - It involves the dissolution of CO_2 in a liquid solvent without chemical reactions. Processes like Selexol and Rectisol are notable for their efficiency and reduced capital and operational expenses. The energy consumption for physical absorption ranges between 1.0-2.0 MJ $kgCO_2^{-1}$, depending on the specific process and solvent used. This lower energy requirement is due to the absence of chemical reactions and the less intensive solvent regeneration process.

- Advantages

  - Lower Energy Requirements: physical absorption does not require significant energy for solvent regeneration, making it a more energy-efficient option.
  - Simplicity and Safety: The process is less complex and involves fewer hazardous chemicals, reducing operational risks and simplifying maintenance.
  - Flexibility: Physical absorption systems can be easily scaled and adapted to various operational sizes and conditions, offering broad applicability across industries.
- Disadvantages
  - Lower CO_2 Capture Rates: The efficiency of physical solvents is generally lower than that of chemical absorption, especially at low CO_2 concentrations.
  - Sensitivity to Temperature and Pressure: The efficiency of CO_2 capture via physical absorption is highly dependent on the temperature and pressure conditions, requiring precise control to optimize performance.
  - Requirement for Pre-treatment: flue gases may need to be pre-treated to remove contaminants that can affect the solubility of CO_2 in the absorbent.

Ionic Liquids - Defined by their unique properties, such as low volatility, high thermal stability, and significant CO_2 solubility, ILs offer a promising alternative to traditional absorption methods. Their tailor-made chemical structures allow for the customization of physical and chemical properties, aligning with specific requirements for CO_2 separation processes. The energy consumption for processes involving ILs varies based on the specific ionic liquid used and its interaction with CO2. Generally, the energy requirement is around 2.0-3.0 MJ $kgCO_2^{-1}$. ILs can be engineered to minimize regeneration energy, but their higher initial manufacturing costs and operational challenges can affect the overall efficiency.

- Advantages

  - Selective Absorption: ILs can be engineered to possess high selectivity for CO_2, even in the presence of contaminants or other gases.
  - Thermal Stability: ILs exhibit exceptional thermal stability, which allows them to operate efficiently across a wide range of temperatures without degradation.
  - Minimal Environmental Impact: Due to their low volatility, ILs have a minimal environmental footprint in terms of emissions. Their stability and recyclability further contribute to reducing the overall environmental impact of CO_2 capture processes.
  - Versatility: The structural versatility of ILs enables the design of solvents with specific functionalities, such as enhanced CO_2 solubility or reduced viscosity
- Disadvantages
  - High Manufacturing Costs: The synthesis of ILs can be complex and costly, particularly for those designed with specific functionalities, thus currently limiting their widespread adoption for CO_2 capture.
  - Regeneration Energy Requirements: While ILs are efficient in capturing CO_2, the energy required for their regeneration, particularly for ILs that chemically react with CO_2, can be significant.
  - Operational Challenges: The viscosity of some ILs can pose challenges in process design and operation, affecting mass transfer rates and requiring specialized equipment to manage fluid dynamics effectively.
  - Compatibility and Corrosion: Compatibility with existing infrastructure and potential corrosiveness towards materials used in CO_2 capture equipment are concerns that need to be addressed.

Adsorption

Adsorption processes, based on the of use surface properties of solid materials, are crucial for their versatility, efficiency, and scalability in capturing CO_2 from gas streams. Adsorption can be further divided into two main categories based on the nature of the interaction between the adsorbent material and CO_2: physical adsorption (physisorption) and chemical adsorption (chemisorption). Each category brings its distinct advantages and challenges, tailoring to different operational needs and environmental objectives.

Physical Adsorption - It involves the establishment of weak interactions, such as van der Waals forces, between the adsorbate (CO_2) and the adsorbent material. This process is characterized by its low energy requirements for both adsorption and regeneration, making it an appealing option for large-scale applications. Physical adsorption, using materials such as activated carbons or MOFs, has an energy requirement of approximately 1.5-2.0 MJ $kgCO_2^{-1}$, which is relatively lower with respect to the other separation techniques due to the less intensive regeneration process.

- Advantages
    - Energy Efficiency: Physisorption typically requires less energy for adsorbent regeneration compared to chemisorption, due to the weaker interactions between CO_2 and the adsorbent.
    - Versatility: A wide range of materials, including activated carbons, zeolites, and Metal-Organic Frameworks (MOFs), can be employed as adsorbents, offering flexibility in process design.
    - Rapid Dynamics: The process features fast adsorption and desorption kinetics, allowing for efficient cyclic operations in response to fluctuating process conditions.
- Disadvantages
    - Lower Capacity at Low Pressures: Physisorption is generally more effective at higher pressures; at lower pressures common in flue gas applications, its capacity diminishes.

  - Sensitivity to Impurities: The presence of moisture and other impurities in the gas stream can significantly impact the adsorption capacity and selectivity of physical adsorbents.
  - Operational Limitations: Achieving high selectivity and capacity might require operating at lower temperatures, which can add complexity to integration with industrial processes.

Chemical Adsorption - It involves the formation of stronger chemical bonds between CO_2 and the adsorbent. This results in higher selectivity and capacity for CO_2 capture, particularly at low pressures, making chemisorption suitable for capturing dilute CO_2 streams. Chemical adsorption, while having higher selectivity and capacity, requires more energy for regeneration with respect to the physical adsorption, resulting in energy consumption in the range of 2.5-3.0 MJ $kgCO_2^{-1}$.

- Advantages
  - High Selectivity and Capacity: Chemisorption captures CO_2 efficiently even at low concentrations due to the strong chemical affinity between the adsorbent and $CO_{2.}$
  - Stable Operation: The strong bonding ensures a stable CO_2 capture process, reducing the likelihood of breakthrough and leakage.
  - Potential for Integration: Certain chemisorptive materials can be regenerated using heat or pressure swings, allowing for potential integration with industrial processes that provide waste heat.
- Disadvantages
  - Higher Energy Requirements for Regeneration: The strong chemical bonds formed during chemisorption necessitate higher energy inputs to regenerate the adsorbent, increasing operational costs.

  - Process Complexity: The complexities associated with regeneration, potential deactivation by impurities, and higher costs compared to physical adsorbents complicate their widespread adoption.

Membrane Separation

Membrane-based methods stand out for their efficiency and versatility. Membranes are typically categorized into two main types: organic and inorganic, each offering unique advantages and facing distinct challenges in the context of CO_2 capture. The energy requirement for membrane separation is typically around 1.0-2.5 MJ $kgCO_2^{-1}$, depending on the type of membrane material used and the operational conditions.

Organic Membranes - Organic membranes, primarily composed of polymeric materials, are widely recognized for their flexibility, scalability, and relatively low manufacturing costs. Their ability to selectively permeate CO_2 over other gases such as nitrogen makes them particularly useful in post-combustion CO_2 capture processes.

- Advantages
  - Cost-Effectiveness: Organic membranes are generally cheaper to produce than inorganic ones, making them cost-effective for large-scale applications.
  - Versatility: The chemical composition of polymeric membranes can be tailored to enhance selectivity and permeability for CO_2, allowing for customization to meet specific process requirements.
  - Scalability: Their flexibility and ease of fabrication support the scaling of processes from pilot to industrial levels without significant increases in cost.
- Disadvantages

  - Chemical Stability: Organic membranes can be susceptible to chemical degradation when exposed to impurities in flue gases, such as SO_x and NO_x, limiting their lifespan.
  - Thermal Stability: High temperatures commonly encountered in gas streams can compromise the structural integrity of polymeric membranes, reducing their effectiveness over time.
  - Plasticization: The presence of high CO_2 concentrations can lead to plasticization, where the membrane swells and loses its selective properties, reducing its overall performance.

Inorganic Membranes - Inorganic membranes, made from materials such as zeolites, silica, or metal oxides, offer exceptional thermal and chemical stability, making them suitable for high-temperature applications. Their robust nature allows for operation in harsher conditions compared to organic membranes.

- Advantages
  - High Thermal and Chemical Stability: Inorganic membranes can withstand higher temperatures and harsh chemical environments, making them ideal for applications involving hot gas streams or corrosive gases.
  - Long Lifespan: Due to their durability, inorganic membranes typically have a longer operational life, reducing the need for frequent replacements and thereby lowering long-term operational costs.
  - High Selectivity: Certain inorganic membranes, such as those made from zeolites, offer very high selectivity for CO_2, enabling efficient separation even in the presence of other gases.
- Disadvantages

  - Cost: The manufacturing and processing costs of inorganic membranes are generally higher than those for organic membranes, which can be a barrier to widespread adoption.
  - Brittleness: The rigid structure of inorganic materials can lead to brittleness, making them more susceptible to physical damage during handling and operation.
  - Complex Fabrication: The production of inorganic membranes often involves more complex and energy-intensive processes, which can limit their scalability and increase production costs.

Cryogenic Separation

Cryogenic technology involves the extraction of CO_2 from other components through multiple stages of compression and cooling at very low temperatures (–100 ˚C to –135 ˚C) and high pressures (100-203 bar), resulting in high-purity liquid CO_2 suitable for transportation and storage. The energy consumption for cryogenic methods ranges from 0.8 MJ $kgCO_2^{-1}$ when using waste cold energy from industrial sources, such as LNG, up to 10 MJ $kgCO_2^{-1}$ for conventional setups. Although cryogenic separation can achieve high-purity CO_2 and offers advantages such as no chemical usage and high CO_2 recovery rates, the high energy demand remains a major challenge.

- Advantages
  - No chemicals or solvents: Eliminates recurring consumable costs.
  - No water requirement: Reduces operational complexity.
  - High Purity CO_2: suitable sequestration.
- Disadvantages
  - Potential Blockages: Risk of ice or solid CO_2 formation.
  - High Energy Demands: Requires significant energy for cooling and compression.
  - Costly pre-treatment: Need to remove water and impurities to prevent blockages.

Table 30. CO_2 separation techniques: advantages, disadvantages and main features [2,17,168,169,388–392].

	Type	Advantages/Disadvantages	CO_2 capture efficiency %	Energy demand MJ $kgCO_2^{-1}$
Absorption	Chemical (e.g. MEA, DEA, etc.)	**Pros**: • Widely commercial. • High CO_2 capture efficiency. • Reversible processes allow for solvent regeneration. • Adaptable to specific industrial applications. **Cons**: o Energy-intensive regeneration. o Potential for solvent degradation. o Higher operational costs.	90 - 99	4.0 - 6.0
	Physical (e.g. Selexol, Purisol, etc.)	**Pros**: • Lower energy requirements for regeneration. • Simpler and safer operational processes. • Flexible and easily scalable. **Cons**: o Lower CO_2 capture rates. o Efficiency highly dependent on temperature and pressure. o May require pre-treatment of gases.	90 - 98	2.0 - 4.5
	Ionic Liquids (ILs) (e.g. Imidazolium-based ILs, AA-ILs, etc.)	**Pros**: • High selectivity for CO_2 absorption. • Exceptional thermal stability. • Minimal environmental impact due to low volatility. **Cons**: o High manufacturing costs. o Significant energy may be required for regeneration. o Operational challenges related to viscosity and compatibility.	95 - 99	1.5 – 2.0
Adsorption	Physical (ACs, zeolites, MOFs, etc.)	**Pros**: • Energy-efficient regeneration. • Fast adsorption and desorption kinetics. • Wide range of applicable materials. **Cons**: o Reduced capacity at low pressures. o Sensitivity to impurities.	80 - 95	2.0 – 3.5

		○ May need lower temperatures for optimal performance.		
	Chemical (amine-impregnated/grafted materials, etc.)	**Pros**: • High selectivity for CO_2 absorption. • Exceptional thermal stability. • Minimal environmental impact due to low volatility. **Cons**: ○ Higher energy for regeneration. ○ Material degradation over time. ○ More complex system design.	70 - 95	1.5 – 4.0
Membranes	Organic (Polymeric)	**Pros**: • Cost-effective production. • Chemical customization for selectivity. • Scalable and flexible for various applications. **Cons**: ○ Susceptible to chemical degradation. ○ Structural integrity affected by high temperatures. ○ Potential for plasticization.	80 - 90	0.5 – 6.0
	Inorganic (Non-polymeric)	**Pros**: • High thermal and chemical stability. • Long lifespan reduces replacement frequency. • Very high selectivity for CO_2. **Cons:** ○ Generally higher manufacturing costs. ○ Water vapor in flue gases can impair adsorption and selective transport. ○ Challenges in maintaining high selectivity/permeability at high temperatures. ○ Achieving high selectivity/permeability simultaneously is challenging.		
Cryogenics		**Pros**: • High separation efficiency. • Mature technology. **Cons:** ○ Very energy-intensive process because it operates at extremely low temperatures and high pressures. ○ Need for moisture pre-removal.	> 95	0.8 - 10

- Solidified CO_2 may accumulate on the surface of the heat exchanger.

4. Challenges and future perspectives

This review thoroughly examines the current landscape and future prospects of CO_2 post-combustion capture technologies, providing a critical lens through which the nuanced methodologies are examined. The review underscores absorption, adsorption, and membrane separation as fundamental technologies pivotal to enhancing CO_2 capture efficiency from post-combustion gas streams. These methodologies, each bearing distinct advantages and facing unique challenges, collectively contribute to the broader strategy of Carbon Capture and Storage (CCS), a crucial effort in mitigating the escalating crisis of global greenhouse gas emissions.

ABSORPTION - Solvent-based absorption techniques have long served as the backbone of post-combustion capture strategies, prized for their operational feasibility and seamless integration into existing infrastructures. Despite their widespread implementation, these techniques face considerable challenges that underscore an urgent need for transformative advancements in solvent chemistry. This review clarifies the critical issues at hand, notably the high energy requirements associated with solvent regeneration and the looming environmental impacts of solvent use and disposal. These challenges not only compromise the efficiency and sustainability of the capture process but also elevate operational costs and environmental risks. The quest for more sustainable and energy-efficient solvents has catalyzed a shift towards exploring novel chemical pathways and alternative solvent systems. Among the promising candidates are blended amine-based solvents and ionic liquids, which have emerged as frontrunners due to their potential to offer significantly lower regeneration energies coupled with reduced toxicity profiles. However, the transition to these novel solvents is not without its challenges. The development of amine-based and ionic liquid solvents necessitates a deeper understanding of their physicochemical interactions with CO_2, as well as their stability, capacity, and selectivity under varied operating conditions. Moreover, economic considerations, including the cost of synthesis, scalability, and the lifecycle analysis of these solvents, remain pivotal factors that will ultimately determine their viability and adoption on an industrial scale. In light of these considerations, the future of solvent-based absorption techniques hinges on the successful navigation of the complex interplay between innovation in solvent chemistry, environmental sustainability, and economic feasibility.

ADSORPTION - Adsorption, distinguished by its potential for low-energy operation and high selectivity, emerges as a promising avenue towards energy-efficient CO_2 capture. The development of novel adsorbent materials, such as MOFs, offers substantial improvements in adsorption capacity and stability. These cutting-edge materials are praised for their remarkable adsorption capacity and stability, which are crucial for efficient CO_2 capture. Nevertheless, although these developments represent significant progress, they also introduce challenges that require careful scrutiny. A primary concern lies in the scalability of these advanced adsorbents. Although novel proposed sorbents demonstrate exceptional laboratory-scale performance, replicating this efficiency on an industrial scale remains a daunting task. Within this context, it is, indeed, evident that the primary scientific hurdles largely revolve around creating new materials with improved CO_2 absorption capabilities through eco-friendly and scalable synthesis methods. Specifically, the control of their chemical and physical properties at the molecular level is crucial, facilitated by a deeper insight into the structure/function dynamics. Furthermore, to tackle the economic and environmental challenges associated with adsorbent production, there should be a significant emphasis on using sustainable raw materials, like agricultural and food waste. This challenge is also intensified by the energy-intensive nature of the adsorbent regeneration step, a pivotal step of the process that significantly influences the overall sustainability and cost-effectiveness of the adsorption method. Indeed, regeneration, which involves removing the captured CO_2 from the adsorbent to make it reusable, often requires substantial energy, thus diminishing the low-energy advantage of the adsorption process. Addressing these issues necessitates a dual approach. First, there is a pressing need to enhance the robustness of adsorbent materials. This involves developing adsorbents that can maintain their structural integrity and performance over many cycles of CO_2 capture and regeneration, even under the harsh conditions typical of industrial applications. Second, optimizing thermal management strategies during the regeneration step is crucial. Innovative solutions that reduce the thermal energy required for regeneration, such as the implementation of novel swing processes or the integration of renewable energy sources, could play a significant role in overcoming the current limitations. In light of these considerations, the field of adsorption-based CO_2 capture is at a crossroads. The path forward will undoubtedly involve tackling the intricacies of material scalability and the energetics of the regeneration process. Through targeted research and development efforts, there is an opportunity to refine adsorption technologies, making them not only more efficient and selective but also more compatible with the demands of large-scale, sustainable industrial applications.

MEMBRANES - Membrane technologies introduce a groundbreaking phase in CO_2 post-combustion capture, providing a low-energy option as an alternative to traditional separation techniques, enabled by advances in materials science. These advancements aim to produce membranes characterized by exceptional CO_2 permeability and selectivity, essential qualities for effective CO_2 separation. However, the application of membrane technologies in the context of CO_2 capture presents a unique set of challenges that must be navigated to unlock their full potential. A principal challenge is the optimization of the trade-off between selectivity and flux. High selectivity towards CO_2 is crucial for effective separation from flue gases, yet this often comes at the cost of reduced gas flow through the membrane, leading to lower overall process efficiency. Achieving an optimal balance requires not just advancements in membrane material composition but also innovative membrane module designs that can enhance gas flow without compromising selectivity. Moreover, the durability of membrane materials under the stringent conditions encountered in post-combustion capture processes represents a significant hurdle. Flue gases contain a variety of impurities, such as sulfur and nitrogen oxides, that can degrade membrane materials over time, reducing their effectiveness and lifespan. The development of membranes that can withstand these harsh conditions while maintaining high performance is crucial for the practical application of membrane technologies in industrial settings. Beyond material and design challenges, the scalability of membrane technologies poses another layer of complexity. Translating laboratory-scale successes to commercial-scale operations requires not only materials that are cost-effective to produce at scale but also membrane systems that can be integrated into existing infrastructure with minimal modifications. This necessitates a multidisciplinary approach that combines insights from chemistry, engineering, and economics to develop solutions that are not only technologically sound but also commercially viable.

CRYOGENICS - Cryogenic separation presents a compelling option for CO_2 capture by leveraging extremely low temperatures to achieve high-purity CO_2 separation. This technique is particularly advantageous due to its ability to avoid chemical usage, thus eliminating issues related to solvent degradation and toxicity. Additionally, cryogenic separation can deliver CO_2 at high pressures, which is beneficial for processes such as enhanced oil recovery and sequestration. However, the high energy demands for cooling and compression remain significant challenges. The integration of waste cold energy from industrial sources, such as LNG, can mitigate some of these energy costs, making cryogenic separation more viable. Despite its advantages, the practical application of cryogenic separation in post-combustion scenarios is still limited and requires further research to optimize its energy efficiency and scalability.

The integration of different CO_2 separation methods is also a promising strategy to develop more economical and efficient capture processes tailored to different requirements. This hybrid approach can capitalize on the unique advantages of each method while mitigating their individual limitations, thus offering a flexible and robust solution to carbon capture challenges. Combining these methods can, indeed, significantly enhance the overall efficiency and cost-effectiveness of carbon capture systems. For instance, hybrid membrane-cryogenic [346] and absorption-membrane [393] processes have demonstrated reduced energy requirements and high CO_2 recovery rates compared to traditional methods, like chemical absorption, making them competitive alternatives. However, integrating different capture technologies also introduces technical and economic challenges. The complexity of managing multiple processes requires sophisticated control systems and thorough understanding of the interactions between different components. Ensuring seamless integration is critical for maintaining system stability and performance, necessitating advanced process designs and control strategies. Additionally, the materials used in these hybrid systems must exhibit high durability and compatibility to withstand varied operational environments without significant degradation. Future research should focus on optimizing these hybrid systems to balance performance and cost. Advanced simulation and modeling techniques can aid in fine-tuning the integration of different methods, minimizing energy consumption, and improving overall process efficiency. Moreover, conducting comprehensive techno-economic analyses will help identify the most cost-effective configurations and operational strategies, ensuring that the benefits of hybrid systems are realized in practical applications. Innovations in material science and process engineering will play, then, a crucial role in enhancing the performance and economic viability of combined capture methods. Additionally, real-world demonstrations and pilot projects are essential to validate the performance and reliability of these integrated systems on a larger scale, paving the way for their industrial deployment. By addressing these challenges and leveraging the synergistic benefits of combining different capture methods, more economical and efficient carbon capture solutions will be developed.

Concluding, the path to advancing CO_2 post-combustion capture technologies is filled with complex challenges but also boundless opportunities. As this review suggests, the future of CO_2 post-combustion capture does not lie in a single technology but rather in an integrated and holistic approach that leverages the collective strengths of diverse methodologies. Navigating the complex landscape of material science innovations, process optimizations, and scalability challenges will require concerted efforts from the global research community, industry stakeholders, and policymakers. This integrated approach holds great promise for advancing carbon capture technologies and contributing to global efforts in mitigating climate change.

5. Conclusions

This review meticulously explores the state-of-the-art in CO_2 post-combustion capture technologies, providing a detailed examination of the advancements, limitations, and performance characteristics of absorption, adsorption, and membrane separation methods. Through a critical lens, the manuscript delineates the significant strides made in each technology while also highlighting their distinct roles within the broader framework of Carbon Capture and Storage (CCS) to mitigate global greenhouse gas emissions effectively.

ABSORPTION - The review underscores the dominant position of solvent-based absorption in the current CO_2 capture landscape, driven by its operational feasibility and adaptability to existing infrastructures. Despite its widespread adoption, this method efficiency is tempered by high energy demands for solvent regeneration (4.0 - 6.0 MJ $kgCO_2^{-1}$). Recent advancements have focused on developing novel solvent systems, including blended amine solutions and ionic liquids, demonstrating potential for reduced regeneration energies and lower environmental impacts. These innovations represent significant progress in enhancing the sustainability and economic viability of absorption technologies.

ADSORPTION - The analysis reveals that adsorption techniques, characterized by their potential for low-energy operation and high selectivity, have seen substantial advancements through the development of novel adsorbent materials (e.g. carbon-based and zeolite-like materials, MOFs, etc.). These materials have been shown to offer superior adsorption capacity and stability, crucial for efficient CO_2 capture. The review notes that while these developments mark a significant leap forward, the practical application of such advanced adsorbents at an industrial scale remains a challenge, reflecting the need for ongoing research and development in adsorbent materials.

MEMBRANE - Membrane separation has been identified as a promising avenue for energy-efficient CO_2 separation, with significant advancements in materials science contributing to the development of membranes with enhanced CO_2 permeability and selectivity. The review highlights the progress made in tailoring membrane compositions to achieve optimal separation performance, though it acknowledges that balancing selectivity with flux and ensuring membrane durability under post-combustion conditions are ongoing challenges.

CRYOGENICS – Cryogenic separation is recognized as a promising method for achieving high-purity CO_2 capture using very low temperatures. This approach eliminates the requirement for chemical solvents and yields CO_2 at high pressures, which is beneficial for enhanced oil recovery and sequestration. While this method faces high energy demands for cooling and compression, the use of waste cold energy from industrial sources can help reduce these costs. Further research is, therefore, needed to enhance the energy efficiency and scalability of cryogenic separation for large-scale applications.

Concluding, the review articulates a comprehensive overview of the current achievements within CO_2 post-combustion capture technologies, highlighting the advancements across absorption, adsorption, and membrane separation methods. Each technology has evolved significantly in the last decades, reflecting concerted efforts to address the efficiency and environmental impact of CO_2 capture. However, integrating and fine-tuning these technologies to boost their efficacy, scalability, and sustainability is imperative. The path to realizing the full potential of these technologies in the framework of CCS is marked by the imperative for constant innovation, cross-disciplinary collaboration, and a holistic understanding of their integration within the global framework of climate change mitigation.

Nomenclature

AC	Activated carbon
AEAPS	N-(2-aminoethyl)-3-aminopropyltrimethoxysilane
AEEA	2-(2-Aminoethylamino)ethanol
AMP	2-Amino-2-methyl-1-propanol
AP	Aminopropyl
APG	Aminopropyl gel
APTES	3-aminopropyl-triethoxysilane
BEA	Beta zeolite
CCS	Carbon Capture and Storage
CNTs	Carbon nanotubes
CO_2	Carbon Dioxide
DBU	Diazabicyclo-[5.4.0]-undec-7-ene
DEA	Diethanolamine
DEPG	Dimethyl Ether of Polyethylene Glycol
DETA	Diethylenetriamine
DIPA	Di-2-propanolamine
DMC	Dimethyl Carbonate
EDA	Ethylenediamine
EG	Ethylene Glycol
FAU	Faujasite
H2S	Hydrogen Sulfide

IEA	International Energy Agency
IGCC	Integrated Gasification Combined Cycle
ILs	Ionic Liquids
LNG	Liquefied Natural Gas
LTA	Linde Type-A zeolite
MCM-41	Mobil Composition of Matter No. 41
MEA	Monoethanolamine
MDEA	Methyl Diethanolamine
MOF	Metal Organic Framework
MT	Metric Tons
MMT	Million Metric Tons
MWCNT	Multi-walled Carbon Nanotube
NH4	Ammonium
NH4HCO3	Ammonium Bicarbonate
NH4NO3	Ammonium Nitrate
NOx	Nitrogen Oxides
NMP	N-methyl-2-pyrrolidone
PAA	Polyallylamine
PC	Propylene Carbonate
P_{CO2}	CO_2 Partial Pressure
PEI	Polyethylenimine
PE-MCM-41	Pore-expanded MCM-41

PMMA	Polymethylmethacrylate
PPI	Polypropylenimine
PSA	Pressure Swing Adsorption
PZ	Piperazine
SBA-15	Santa Barbara Amorphous No. 15
SBA-16	Santa Barbara Amorphous No. 16
SO2	Sulfur Dioxide
T	Temperature
TEPA	Tetraethylenepentamine
TRL	Technology Readiness Level
TRI	(3-aminopropyl)-trimethoxy-silane
TSA	Temperature Swing Adsorption
VSA	Vacuum Swing Adsorption
VTSA	Vacuum Temperature Swing Adsorption
$y_{feedCO2}$	CO_2 Molar Ratio in the Feed Stream

Author biographies

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Federica Raganati received her degree and PhD in Chemical Engineering from the University Federico II of Naples (Italy) in 2010 and 2014, respectively. She is a researcher at the Institute of Sciences and Technologies for Sustainable Energy and Mobility (STEMS – CNR). Her main research topics are the set-up of innovative fluidized bed reactor configurations (e.g. sound-assisted fluidized beds), the reduction of CO_2 emissions through post-combustion capture processes and the energy storage through thermochemical processes.

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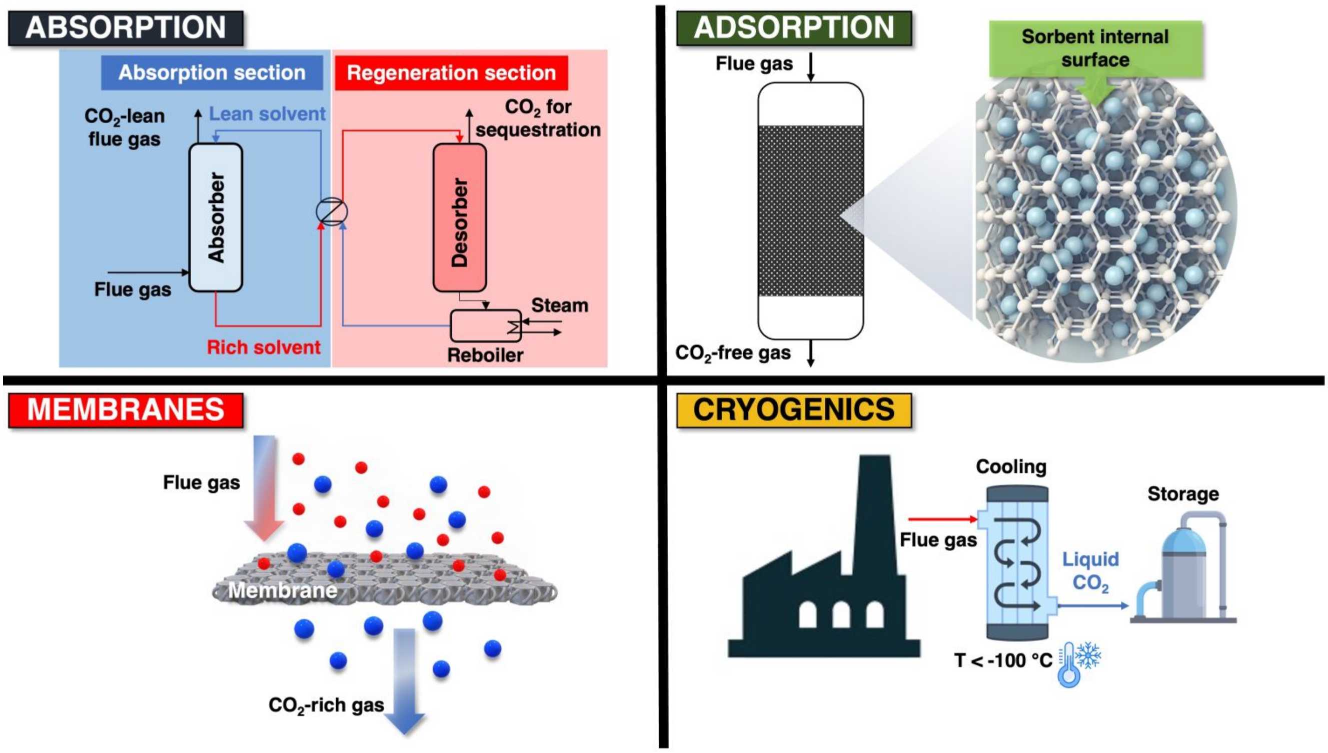
TOC graphic


ABSORPTION
Absorption section
Regeneration section
CO_2-lean flue gas
Lean solvent
CO_2 for sequestration
Absorber
Desorber
Flue gas
Steam
Rich solvent
Reboiler
ADSORPTION
Flue gas
Sorbent internal surface
CO_2-free gas
MEMBRANES
Flue gas
Membrane
CO_2-rich gas
CRYOGENICS
Cooling
Storage
Flue gas
Liquid CO_2
T < -100 °C