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CO_2 capture: a concise, comprehensive overview of recent research trends

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Abstract

Due to the growing threat of climate change, the pressing need for carbon dioxide capture has become a global priority in the development of innovative technologies. Multipronged approaches and multifarious research efforts are underway to efficiently capture carbon dioxide (CO_2) from emission sources, ambient air, and indoor air. Currently, absorption is the dominant industrial-scale process, using different solvents and their blends to lower the energy intensity of solvent desorption and regeneration. However, adsorption is emerging as a promising alternative due to its energy efficiency, eco-friendliness, and potential for large-scale applications. High-performance sorbents with large surface areas and bio-based materials exhibit high CO_2 loading and selectivity in fixed-bed and fluidized-bed systems. Cryogenic CO_2 capture systems, which do not require solvents or membranes, are optimized for energy through process integration. Researchers are investigating different membrane materials in hollow fiber membrane contactors for enhanced CO_2 capture efficiency. Membranes that can selectively filter CO_2 from gas mixtures are also being explored. Furthermore, hybrid technologies integrating different CO_2 capture approaches are being developed to reduce costs and boost overall performance to curb rising atmospheric CO_2 levels.

Keywords: *absorption, adsorption, climate change, cryogenic distillation, direct-from-air capture, hybrid separation systems, hybrid separation technologies, membrane systems*

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1. Introduction

High levels of atmospheric carbon dioxide (CO_2), contributing to complex weather events that are draining the global economy, are being addressed by global research to reduce CO_2 emissions into the atmosphere and capture, utilize, and store the CO_2 emanating from different sources. CO_2 capture is the largest part of the total carbon capture, utilization, and storage (CCUS) cost [1, 2]. CO_2 capture is primarily intended for sources such as fossil fuel-based power plants and industries including steel, cement, petroleum processing, and petrochemicals, given their large-scale contributions to CO_2 emissions in the atmosphere. An analysis is available on the major industrial sectors and countries with CO_2 capture potential [3]. Bibliometric analysis of CO_2 capture technologies, carried out by employing VOSviewer software (version 1.6.15) [4] and reviewing the patents registered for the technologies with patent holders, grants us a deep understanding of the technologies and current research gaps, which will facilitate planning future research [4]. Advances in renewable energy sources [5] and energy policies and commitments of governments and corporate boards are being proposed to boost the advancement and commercialization of CO_2 capture technologies.

The basic methods for carbon capture are (i) pre-combustion (fuel gasification with oxygen from the air separation unit and CO_2 separation from syngas) capture, (ii) the oxy-fuel process (fuel combustion with oxygen from the air separation unit and partial recycling of the flue gas), and (iii) post-combustion (conventional fuel combustion with air) capture [6, 7]. Post-combustion capture is widely researched because of its flexibility and easy integration with existing power generation and industrial units. The absorption process, being simple, is commonly used for large-scale CO_2 capture. Traditionally, sodium or potassium hydroxide solution and monoethanolamine (MEA) solvents are used. Despite CO_2 being easily absorbed in these solvents, subsequent desorption and solvent regeneration processes are energy-intensive and challenging. Also, the decomposition of solvents during thermal regeneration forms undesirable byproducts, affecting the overall effectiveness and economics of the process. Adsorption, an energy-efficient and environmentally friendly process with high capacity and selectivity, can overcome these challenges [8–10] without generating secondary waste streams [11]. Advances in synthesis and cost-effectiveness of activated porous carbon adsorbents from biomass as precursors are comprehensively reviewed [12, 13] for their costs and CO_2 adsorption capacity and performance. Cryogenic distillation and membrane processes are efficient when the gas streams are rich in CO_2 [14]. Capturing CO_2 with microalgae [15] can help recycle it as a carbon source which can be useo grow biomass for bioenergy and other products.

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The CO_2 capture literature [2, 16–18] reveals research efforts related to developing and improving technologies, such as (a) absorption–physical sorption with solvents, such as Selexol, Rectisol, and Purisol, and chemical sorption with solvents [19] such as alkali, amines, amine blends, ionic liquids [20], deep eutectic solvents, and ammonia; (b) adsorption, using adsorbent beds (fixed or fluidized) of carbon, zeolites, silica, metal–organic frameworks, amine-impregnated adsorbents, or polymers and regeneration cycles, such as pressure swing, temperature swing, steam, and moisture; (c) membrane separation [1], e.g., gas separation using membrane materials, such as polyamide, polyphenylene oxide, and polydimethylsiloxane and gas absorption using membrane materials, such as polypropylene, and PTFE and ceramic bed systems; (d) cryogenic separation in cryogenic distillation or cryogenic packed beds [21] and (e) chemical or solvent looping. Each of these technologies has achieved different levels of perfection and technological readiness, with some advantages and limitations related to suitability for post-combustion CO_2 capture, efficiency, energy intensiveness, environmental impacts, footprint, economics, ease of incorporation into the operating facilities, etc.

Research is also underway that involves capturing CO_2 directly from ambient air (DAC) to supplement carbon capture efforts from point source emissions. Several DAC technologies are being developed, each with unique features and some limitations [22]. These technologies include the solid adsorbent-based approach using materials, such as zeolites [23] and activated carbon [24]; the liquid absorbent-based approach using solvents, such as aqueous amine blends [24]; and membrane-based systems using high surface-area-to-volume membrane contactors for CO_2 absorption with sodium hydroxide [25]. Efficient CO_2 capture and its conversion into usable organic compounds is a vital and challenging mission [26] to mitigate climate change-related challenges.

2. Methodology

An extensive literature search was conducted which involved scanning Scopus journals and Google Scholar for an overview of the research trends in CO_2 capture in this decade. The research for post-combustion CO_2 capture continues to rely on basic processes, such as absorption, adsorption, membrane-based processes, and cryogenics, summarized in **Table 1**. However, several advances are taking place in process configurations, operating conditions, and materials, enhancing CO_2 capture selectivity, energy efficiency, and productivity. Further, direct-from-ambient-air CO_2 capture is being explored through absorption, adsorption, and direct atmospheric cryogenic processes. This paper synthesizes a concise yet comprehensive overview of the current research trends in post-combustion and direct-from-air CO_2 capture using absorption, adsorption, membrane, and cryogenic systems. Unlike the review, this overview provides a broad summary of research areas, highlighting key aspects without an in-depth critical assessment of research outcomes, comparing approaches, or identifying research gaps.

3. Absorption

Solvent-based absorption is currently the most technologically mature CO_2 capture strategy and has been successfully implemented commercially for many years [27–30] using MEA as the predominant solvent. Also, the MEA-based post-combustion CO_2 capture [31, 32] is among the most widely employed technologies for retrofitting natural gas combined cycle power plants. An amine-based carbon capture path, using MEA, diethanolamine (DEA), and triethanolamine, is the prime CO_2 technology employed to capture CO_2 from industrial flue gases, with the advantages of high CO_2 capture capacity and an accelerated absorption rate but the disadvantage of being energy-intensive. Thus, new amine solvents with high capture efficiency and low energy consumption are being developed [33]. Blended amine solvents used to capture CO_2 have gained attention due to the advantages of rapid absorption kinetics of primary and secondary amines and lower regeneration energy requirements for tertiary amines over MEA [34]. Electrochemically mediated amine regeneration for efficient desorption of amines and artificial intelligence applications in optimizing post-combustion CO_2 capture technologies are researched [35]. Process simulation and optimization of the chemical absorption process are conducted using amine solutions and process modeling for post-combustion CO_2 capture from the flue gas sources with a high CO_2 concentration, such as from the coal chemical industry (40–60 vol% CO_2), tail gas after blast furnace gas, and oil field-associated gas combustion. A simulation model, encompassing the absorption and desorption system, using MEA, MEA and methyl diethanolamine, and 2-amino-2-methyl-1-propanol blends is believed to predict the CO_2 capture system's key performance indicators, such as CO_2 loading, CO_2 capture rate, energy consumption, and the operating parameters. By optimizing solvent usage, a 90% CO_2 absorption rate and 95% product pureness are claimed [36], along with lower energy consumption. 2-(ethylamino)–ethanol-1-dimethylamino-2-propanol (1DMA2P), a tertiary amine composite solvent, is reported [37] to have an excellent mass transfer performance. The composite solvent has the potential to substitute traditional amines because of its ability to capture high amounts of CO_2 from flue gases at lower capital and operating costs than the other amine solvents.

For low-energy-intensive regeneration, a superior-performing solvent, 2-amino-2-methyl-1-propanol (AMP), a hindered form of MEA, one of the least expensive sterically hindered amines, and a readily available and easy-to-process single reactive amino group compound with a lower regeneration temperature than MEA, DEA, and methyl diethanolamine and superior chemical, thermal stability, and degradation rates of about half the values of MEA was employed [38]. Additionally, it induced less corrosion than MEA because of its excellent absorption capacity. However, its reaction rate with CO_2 is lower than that with MEA and DEA, necessitating the employment of a rich AMP solution and mixing with piperazine. Solvent selection in the absorption process is crucial for CO_2 capture, as it directly affects the absorption efficiency, equipment size, and regeneration energy usage and, hence, capital and operating costs. The interaction intensity and diffusivity of amine blends were analyzed using molecular dynamic simulation, given that high intermolecular interaction intensity promotes CO_2 absorption and intramolecular interaction intensity affects CO_2 desorption during regeneration. A decreasing order of interaction intensity was observed in N-Diethylethanolamine (DEEA), MEA-DEEA, piperidine, and 1-methylpiperazine-piperazine, indicating that DEEA can increase the CO_2 absorption rate in pure and composite amine systems. The intramolecular interaction intensity made it simpler to regenerate DEEA, piperidine, piperazine, and 1-MPZ than MEA [33]. A tri-solvent amine system of DEEA, along with piperazine (PZ) and 4-amino-1-methylpiperidine (PD), was

Table 1 • Summary of CO_2 capture approaches.

Process	Approaches and materials	Remarks
Absorption	Physical absorption using solvents such as alcohols, glycols, and acrylates. Chemical absorption using absorbents, such as alkali hydroxides, alcohol amines and their blends, composite solvents, ionic liquids, deep eutectic solvents, nanofluids, proprietary solvents, and ammonia.	Developed and used at a commercial scale. Ongoing investigations for solvents and systems design to enhance CO_2 capture selectivity and energy efficiencies.
Adsorption	Adsorbent beds—fixed, fluidized, or moving beds with non-carbonaceous adsorbents, such as modified silica-based and zeolites-based metal–organic frameworks; carbonaceous adsorbents, such as amine-impregnated, bio-based, polymer-derived and -doped adsorbents, aerogels, activated C, C molecular sieves, C nanotubes, and graphene. MOF combined with membranes and proprietary adsorbents.	Developed at different stages of lab scale and pilot scale. Ongoing investigations for novel materials and their combinations to enhance CO_2 capture selectivity at larger scales.
Membrane-based	Hollow fiber membrane—multi-stage systems used for membrane separation, e.g.,gas separation using membrane materials, such as polyamide, PVDF, polyphenylene oxide, polydimethylsiloxane, and modified polymers, and graphene. Membrane absorption—gas absorption using membrane materials, such as polypropylene, and PTFE and ceramic, zeolite, MOF-based composite and mixed-matrix, ionic liquid–glass, and carbon molecular sieves.	Developed essentially at lab scale and also at pilot scale. Ongoing investigations underway for different membrane materials and systems for the following processes: Membrane separation—enhancing selectivity for diffusion, molecular sieve, solubility, and reaction selectivity. Membrane absorption—enhancing gas–liquid separation selectivity Hybrid systems, such as combining membranes with liquid absorbents, such as MEA or liquid ammonia and membrane cryogenics.
Cryogenics	Cryogenic distillation. Cryogenic packed beds.	Developed and used at a commercial scale. Ongoing investigations to enhance CO_2 capture selectivity and energy efficiencies through process intensification.
Direct from air capture (DAC)	Absorption in aqueous solutions, such as amine blends, alkalis, and membrane contactor systems. Adsorption on solid sorbents, such as zeolites, activated C, cellulose-based, porous C, and synthesized amine-functionalized metal–organic frameworks, nitrogen-rich functional groups and modified biochar-based adsorbents.	Developed at lab scale and pilot scale. Ongoing investigations for large specific surface areas and adjustable pore sorbent systems and direct atmospheric cryogenic C capture

promising for CO_2 removal from coal-based power plants [39] and was found to be superior to MEA in boosting the CO_2 absorption efficiency and lowering CO_2 desorption energy requirements [36].

In another study, a comparative techno-economic analysis and rigorous process simulation were carried out on amine-based solvents, viz. MEA, MEA-methyldiethanolamine (MDEA), piperazine (PZ)-MEA and PZ-MDEA blend for CO_2 capture from a steel plant's flue gases to investigate the absorption synergy of various amine solvent blends [40]. A detailed energy evaluation showed that a solvent with low reaction enthalpy and heat capacity and high reactivity and absorption capacity can minimize the energy cost of the process. New, highly efficient blended amine solvents for capturing CO_2 are investigated to determine their full potential. Multiple capture levels were evaluated for the commercially available amine-based system as the basis for the capture technology at the Hunter plant. This case study revealed that the implemented CO_2 capture system, utilizing the commercially available Mitsubishi Heavy Industries (MHI) KM-CR Process with proprietary KS-1 TM solvent, was technically feasible [41]. The residual acid gases, SO2 and SO3, were removed from the flue gases by scrubbing with caustic soda solution to enhance the CO_2 capture system's efficiency. The expenditure of retrofitting an operating power plant with a CO_2 absorption mechanism depends on the power plant type, location, flue gas composition, capture technology, capture quantity, available utilities, etc. The techno-economic evaluation of a state-of-the-art MEA-based post-combustion CO_2 capture system for a 750 MW combined cycle gas turbine power plant demonstrated that the advanced process configurations evaluation should be based on the overall energy requirements and cost savings rather than only on energy savings, given potential trade-offs involved at the system level [42]. Polyamines, especially those with primary and secondary amines, such as triethylenetetramine (TETA) and diethylenetriamine (DETA), can handle high CO_2 loads with high capture rates. The high regeneration energy consumption is addressed by amine-based biphasic absorbents [43]. While biphasic absorbents constituting tetraethylenepentamine, 1-ethylimidazole, and H2O (TEH) and other multiple amino groups of TETA showed remarkable absorption rates and CO_2 loads, the carbamate generated in absorption substantially increased the CO_2-lower phase's viscosity and also formed precipitation. This made it tough to transfer the enriched phase for regeneration [44]. Specific solvents are added to polyamine biphasic absorbents to improve their phase separation performance and flow properties. A nanofluid, prepared by adding nanoscale TiO2 and SiO2 powder in N, N-dimethylpropylamine-polyethylene glycol dimethyl ether biphasic solvent, showed improved CO_2 absorption and reduced energy requirements for desorption [45].

Catalytic solvent regeneration [46] lowered the regeneration temperature, accelerated the regeneration rate, and reduced energy consumption. A skillfully prepared catalyst, α-Fe_2O_3, showed encouraging two-phase solvent regeneration for triethylenetetramine and 2-(diethylamino) ethanol admixture, a 2.4 times increase in CO_2 stripping, and a 40% reduction in heat duty. The proton transfer mechanism suggested for low-energy-intensive solvent regeneration [46] has potential for industrial applications. Single-atom N-Dimethylformamide solutions, a kind of supported metal or nonmetal catalyst in a single-atom dispersion state [47], are developed with iron and manganese-based single-atom ethanol solutions. Density functional theory simulation and experiments have revealed that the copper, iron, and boron single atoms in the solution promote CO_2 absorption, but manganese is ineffective. Significantly, copper single atom also boosts CO_2 desorption at low temperatures. Copper- and boron-based single-atom [48] solutions' excellent CO_2 sorption efficiency and substantially lower desorption energy compared to those of conventional amine systems make them potential candidates for commercial-scale applications. The traditional alternative solvents developed to lower energy consumption generally reduce CO_2 absorption efficiency. This is addressed by developing non-toxic compatible solvents [49] with low regeneration energy that accelerate CO_2 capture with the help of stable enzymes in operating conditions and have a long life besides post-process sustainability. A liquid-infused surface (LIS) approach is employed for reactive liquids to develop low-energy separation schemes consisting of a chemically functionalized microtextured and thermally conductive solid substrate that retains a high surface area-to-volume ratio geometry to trap and immobilize a tens-of-micrometers-thick liquid layer for reaction and separation for an economical CO_2 capture [50]. When amines are used as reactive liquids, the LIS class is called "solid with infused reactive liquid (SWIRL) amine". SWIRL-tetraethylenepentamine (TEPA) displayed stability and high post-combustion CO_2 capture capacity at flue gas temperatures in a power plant, avoiding energy-intensive temperature swings [51]. Water vapor increased the CO_2 capacity of SWIRL-TEPA without sacrificing its stability.

Korea Electrical Power Corporation tested proprietary solvents, KoSol-5 and KoSol-6, high-efficiency CO_2 absorbents in a 10 MW pilot plant CO_2 capture process and an advanced process and found a reduction in energy consumption equivalent to 10% of the prevailing power generation efficiency [52, 53]. Over 10,000 hrs of operation, the KoSol-6 system demonstrated excellent functioning with a 90% CO_2 capture rate and required 39% less energy than MEA [54]. Four operating modes were taken up, with advanced options progressively added to the basic process; these included intercooler, lean vapor compression [55], and thermal vapor recompression. The key performance parameters, viz. CO_2 absorption and desorption capabilities, CO_2 removal rate, energy consumption, corrosivity, durability against oxidative degradation, and operating and long-term stability were evaluated for MEA, PZ, and PZ-MDEA blend solvents and performances were compared for their ability to capture CO_2 from cement plant emissions. In the aqueous ammonia solution and chilled ammonia process used to capture CO_2 from flue gases generated during ammonia manufacturing, ammonium carbonate, ammonium bicarbonate, and ammonium carbamate were formed in the tower in addition to ammonium salts of NOX and SOX. Integrating this technology with urea synthesis will significantly lower corrosion, energy consumption, and cost [56] compared to the traditionally employed amine solution.

Ionic liquids, i.e., room-temperature molten salts, have good ionization properties and self-assembly capabilities [57]. Ionic liquids, especially amino acid ionic liquids (AAILs), play an important role in CO_2 capture [58, 59]. This class of solvents has shown superior characteristics for CO_2 absorption and displayed significant sorption capacity and selectivity, insignificant volatility, and the notable advantage of not requiring thermal regeneration to help achieve the desired objective of substantially low specific energy consumption. "The catalytic effect of environmentally friendly imidazole series ionic liquids [60] was evaluated for CO_2 desorption rate, amount of CO_2 released, and heat duty and the PF6 catalyst exhibited excellent catalytic activity" for these parameters with good stability, making homogeneous PF6 catalyst a future candidate for use in desorbing the captured CO_2 with lower heat duty. The evaluation of the appropriateness of the PF6 catalyst for secondary and tertiary amine setups also demonstrated promising results. AAILs, the green work fluids prepared from amino acids and their derivatives, have displayed outstanding biodegradability, biocompatibility, and functionalized design. The quantitative relationship of the amino groups in AAILs with CO_2 and CO_2 capture energy requirements is key to the molecular design of perfect AAILs. A thermodynamic model [61] was proposed for chemical reactions at different stoichiometric ratios of CO_2 and AAIL. "The solvent cyclic capacity and regeneration energy of AAILs in the pressure swing and temperature swing CO_2 capture systems were also estimated". It has been found that more amino groups in AAILs do not necessarily increase cycles of repeated solvent use and require higher energy for regeneration. AAILs, which are screened to determine the best performance with regard to high cyclic capacity and low energy consumption, can absorb CO_2. Six AAILs, which were added as split-phase regulators in TEH to form a new liquid–liquid two-phase absorption system, TEH-AAILs, displayed excellent sorption results [62, 63]. To disrupt the intricate hydrogen-bonding network in the system and decrease lower-phase viscosity, H2O was substituted [64] with 1-propanol, a weak hydrogen-bonding acid–base solvent. AMP addition to TETA-1-1DMA2P-methyl-1-propanol blend reduced lower-phase viscosity [65] because of hydroxyl groups in 1-propanol and AMP. The effects of various concentrations of AAILs on CO_2 loading and phase separation performance were studied to design and optimize the biphasic absorbents for CO_2 capture. Based on the outstanding CO_2 capture capacity, low viscosity, and low energy requirement, 1-ethyl-3-methylimidazolium bis[trifluoromethylsulfonyl]imide ([emim][Tf2N]) was singled out as the ionic liquid solvent. An absorption column with two sections with intercooling in between was proposed to optimize the temperature profile and improve energy requirements to address the constraints of lower sorption capacity and increased viscosity [66] under reduced operating pressures and temperatures. An integrated ionic liquid and sorption process design method [67] for gas separation employing a rigorous rate-based process model was demonstrated for pre-combustion carbon capture. A computer-aided approach [68] for the optimal design of ionic liquids for

CO_2 absorption, using a simple and reliable data-driven mathematical model to predict CO_2 solubility, was demonstrated for a post-combustion carbon capture case study.

Deep eutectic solvents (DESs), first prepared by Abbott et al. in 2003, are essentially eutectic mixtures of certain stoichiometric ratios of choline salts and coordination agents, such as metal salts, metal salt hydrates, or hydrogen bond donors [69]. These are non-toxic, biodegradable, low-cost, and easy to synthesize with atom utilization efficiencies of 100% and higher absorption capacity [70] with the benefits of the traditional ionic liquids kept intact. However, their high viscosities sharply reduced CO_2 sorption performance [71]. Adding water to DESs lowers their viscosity but at the cost of absorption capacity. The viscosity can be decreased by functionalizing DES [72, 73]. Encapsulation of the DESs' choline chloride/urea by a porous shell by microfluidics resulted in microcapsules with uniform particle size and waterproofing and a substantial rise in specific surface area and CO_2 absorption capacity. The simultaneous addition of carbonic anhydrase converted the water content in DESs into bicarbonate and enhanced the absorption performance. Jointly encapsulated DES and carbonic anhydrase with microfluidics are potential solvents [74] for efficient CO_2 capture because of their high absorption capacity, rate, selectivity, and ability to desorb CO_2 from microcapsules. From a series of DESs, it was found that cyclodextrins and MEA-based DES displayed high absorption capacity in temperatures ranging from 30 to 110 °C, a low regeneration temperature, and excellent cycling ability [75] for CO_2 capture due to the circular hydrogen bond formed with hydroxyl of MEA and cyclodextrins.

From an energy systems perspective, "DAC technologies are categorized [76] as high-temperature aqueous solutions (HT DAC) and low-temperature solid sorbent (LT DAC) systems". Based on techno-economic analyses of advanced DAC technologies, it is inferred that LT DAC systems are more suitable because of their lower energy costs and waste heat use potential from some other sources or renewable energy sources. A DAC plant is investigated to optimize energy through pinch analysis and thermal integration of flows [77]. In the absorption cycle of the proposed process, CO_2 in the air comes into contact with the aqueous KOH solution absorbent and K_2CO_3 solution, and the reaction product formed is then reacted with $Ca(OH)_2$ to regenerate and recycle the solvent in the desorption cycle. $CaCO_3$ is calcined at high temperatures with natural gas and O_2 from the air separation unit to dissociate CO_2. CaO is reacted with water to regenerate $Ca(OH)_2$ [78]. System optimization can increase heat recovery and substantially reduce hot and cold utilities' requirements. Thus, improved CO_2 capture can be achieved with optimal energy consumption.

4. Adsorption

Due to its simplicity and low energy-intensiveness, adsorption is one of the well-established techniques to capture CO_2 at an industrial scale. The literature divulges that several researchers are working to enhance CO_2 selectivity and the capacity of adsorbents. Solid materials capable of reversibly adsorbing CO_2 have been at the center of intensive research efforts for more than three decades, and encouraging results are reported regarding their stability and low energy consumption when operating the adsorption systems compared to the more widely adopted absorption systems using liquid amines. Based on the working temperatures, "solid CO_2 sorbents are broadly classified [79] into three groups: (i) low-temperature (<200 °C) sorbents, such as zeolite and activated carbon; (ii) intermediate-temperature (200–400 °C) sorbents, such as MgO and layered double hydroxides derived mixed oxides; and (iii) high-temperature sorbents (>400 °C), such as CaO and lithium silicates". Solid oxide sorbents, such as MgO and CaO, have become apparent as potential substitute CO_2 sorbent materials due to their low price, strong cyclic stability, and high CO_2 adsorption. CO_2 adsorption on MgO and NaCl [80] surfaces is modeled and validated with experimental results and a hybrid scheme is proposed [81]. CO_2 removal under moist conditions has also been investigated, considering that Mg (OH)2 has high reactivity in capturing CO_2 [81]. MgO is cost-effective with a lower regeneration temperature than CaO [82] at an operating temperature of 250–450 °C. Impregnation of MgO, a very standard CO_2 sorbent used at intermediate temperatures, with alkali salts, such as $NaNO_3$, generally improves its CO_2 sorption capacity. A CO_2 capture performance study using commercial MgO adsorbents upgraded with a ternary mixture of $LiNO_3$, $NaNO_3$, and KNO_3 for post-combustion CO_2 capture at a high residence time in a fixed-bed reactor revealed that high $LiNO_3$ content in the ternary mixture lowered the CO_2 capacity [83]. In contrast, high ratios of $NaNO_3$ and KNO_3 resulted in a higher CO_2 capacity. It was inferred that molten salt migration and aggregation of MgO particles probably led to a decline in CO_2 uptake over the sorption–desorption cycles.

Robust porous "metal–organic frameworks (MOF), Zn-Ox-Mtz, with one-dimensional channels, and narrow windows" have been identified [84] as up-and-coming sorbents with an excellent capacity to capture CO_2 from flue gases and natural gas with high selectivity. Challenging molecular sieving effects can be achieved with these adsorbents' high surface areas and customized pore features. Carbonaceous and non-carbonaceous adsorbents, such as zeolites, activated carbons, silica, zeolite, MOF [85], organic aerogels, and nano-structured carbon-based adsorbents, such as graphene [86–91] and porous organic polymers [92], are investigated for their CO_2 capture rates and purity and energy requirements. Carbon nanofiber composites—gCNFs—formed by using graphene-based materials in the electrospun CNF as nanofillers are reported to exhibit outstanding CO_2 adsorption properties, which improved further by incorporating amine-modified gCNFs [93]. The evaluation of MOF and zeolite adsorbents in different operating configurations of a vacuum-pressure swing adsorption system revealed that over 90% CO_2 recovery with the same level of purity was obtained for MOF against over 80% for zeolite adsorbent [94]. A fixed-bed adsorption system, consisting of packed MOF sorbents, and temperature swing steam desorption achieved over 85% CO_2 removal from a natural gas combined cycle power plant flue gas [95], which could be increased to over 98% by combining MOF with a membrane capture system. Integration of a four-stage low-temperature vacuum swing adsorption system, together with the removal of water vapors and NOX from the flue gases, is described [96] for capturing post-combustion CO_2 from a natural gas-combined power plant flue gas that can be retrofitted on the existing low CO_2 emission facilities for over 90% CO_2 purity with 15% recovery. A thermodynamic model of a multistage vacuum-pressure swing adsorption cycle [97] was proposed to analyze energy efficiency under different operating parameters.

The synthesis of different types of aerogels—an upcoming class of composites with noticeable features, such as a large surface area, designable pore size, and ample active sites for high CO_2 capture performance—is reviewed, and the future scope of hybrid aerogels is highlighted [98]. Amine-impregnated polymer aerogels [99] for CO_2 removal, considered a pioneering development, were synthesized using an oil–water emulsion polymerization method, freeze-drying, and impregnation with an MEA solution to improve CO_2 adsorption. Reportedly, 90% of aerogels' CO_2 capture capacity was achieved within 10 min, exhibiting their immense potential as extremely efficient sorbents for CO_2 capture. Their stable regeneration capacity makes them potential candidates for greater industrial applications. CO_2 capture inside porous silica was evaluated for a methane steam reforming process at the operating pressures and temperatures, employing Grand Canonical Monte Carlo molecular simulations, and it was inferred that stronger pore–fluid interactions between the silica pores and the adsorbed CO_2 resulted in higher adsorption inside silica pores than that between homogeneous carbon slit pores and the adsorbed CO_2. "The presence of structured layers due to more molecular packing confirms possible liquid-like and vapor-like phase coexistence of the supercritical bulk phase CO_2 under confinement" [100].

Aminosilane-modified hierarchical silica adsorbents [101] with high thermocyclic stability have shown encouragingly high-capacity and fast CO_2 capture at low pressures directly from the atmospheric air and indoors, even at low concentrations. The surface morphology and adjustable porous structure of different functional groups on the polymeric backbone provide high selectivity for CO_2 capture, making them outstanding adsorbents [102, 103]. The amine-functionalized styrene–divinylbenzene copolymer adsorbent [104] reportedly captured the highest CO_2 at 27 °C and near atmospheric pressure and, in the desorption cycle in batch and continuous experiments, 92–94% CO_2 was recovered at 56 °C. Coal fly ash-bound limestone-derived adsorbents, prepared by mixing limestone, calcined lime, and hydrated lime with coal fly ash using the extrusion–spheronization method, improved CO_2 capture stability and elutriation in limestone circulation in a fluidized bed-based calcium looping system. The ternary Ca-Al-O and Ca-Si-O phases offer structural stability to these sorbents and enhance their cyclic CO_2 capture performance because of the combined symbiotic effect of pore structure modification by hydration pretreatment and anti-sintering ability enhancement. Hydrated lime, with its pore structure and strong binding properties [105], is reportedly better than limestone as a calcium precursor for making CaO-based adsorbent pellets. A Li_4SiO_4-based sorbent was more efficient than calcium looping systems for post-combustion CO_2 capture in a natural gas combined cycle plant [106]. The polymer-derived and K-Ti co-doped Li_4SiO_4-based adsorbent showed a positive synergistic effect for low CO_2 concentrations with a high adsorption capacity, fast adsorption rate, lower regeneration temperatures, and highly stable cycling performance [107]. The effective and stable CO_2 capture results make this sorbent a promising candidate for commercial applications.

Various process schemes, heat recovery plans, and fueling options for different CO_2 capture scenarios in a lime plant were computed using Aspen Plus®software. They were then analyzed, and steam cycles were simulated using EBSILON®Professional. A techno-economic assessment using ECLIPSE software and optimization of the indirectly heated carbonate looping (IHCaL) process were explored. The heat recovery strategy for combustion flue gases was the key enabling factor in deploying the IHCaL technology [108]. Solid sorbents in fixed-bed and fluidized-bed adsorbers [109] were demonstrated at a 1 MW coal-based power plant [110] and a 150 kg CO_2/day cement plant [111]. A mathematical model was proposed and validated with a 5 tpd CO_2 pilot plant's operating data to predict the fluidized bed performance using amine-impregnated porous material sorbent [10]. The fluidized bed process, with its larger reaction zone, lower sorbent requirements, and three operations—adsorption, desorption, and drying carried out while the sorbent is in circulation—is potentially suitable for large-scale CO_2 sources.

Porous solid sorbents, classified as physisorbents and chemisorbents with large specific surface areas and adjustable pore conditions, have demonstrated good prospects as a CO_2 capture medium for DAC application [112, 113]. Given the high operating costs of DAC resulting from low CO_2 concentrations of 410 ppm in the ambient air, advanced sorbents with high performance, low cost, and simple engineering designs for large-scale applications are being developed [114]. A low-carbon and low-energy-intensive process to efficiently adsorb CO_2 for DAC applications, employing an MEA-incorporated liquid-like adsorbent, was investigated [115]. The synthesized amine-functionalized metal–organic frameworks [116] reportedly exhibit notable CO_2 capture capacity under damp conditions for DAC applications. Zeolite 13X-APG [117] was examined to improve the understanding of open adsorption for DAC and scaling, performance-affecting parameters, and design optimization were established through sensitivity analyses. Improving the CO_2–sorbent reactions and sorbent regeneration processes can help make CO_2 adsorption competitive with other technologies. It is important to consider chemical interactions besides physical properties. A review of the bio-based adsorbents' synthesis, activation, and their applications in post-combustion CO_2 capture processes is portrayed using current process modeling and machine learning trends are discussed to develop such advanced sorbents and design adsorption systems [11]. The bio-based activated adsorbents, derived from agricultural waste materials, with their low ash content, environmental friendliness, and optimized process economics, are more attractive for CO_2 capture [118]. The preparation and functioning of high-porosity, high-selectivity, low-cost, stable, and renewable biochar-based sorbents are reported [119, 120] for their physicochemical characteristics, such as surface functional groups, alkali earth metals, surface basicity, polarity, aromaticity, and hydrophobicity. Although virgin biochar can be used as it is for CO_2 adsorption, its properties can be enhanced by modifying it with suitable functional groups. Nitrogen-rich functional groups improve the selectivity and adsorption performance of biochar-based adsorbents. Eco-friendly and easy-to-make cellulose-based porous carbon materials, which possess outstanding mechanical and physical characteristics, are proposed as CO_2 adsorbents in solid, cellulose-based films and aerogel forms [121].

The advantages of CO_2 adsorption over other CO_2 capture methods include simple design and operation, low maintenance, energy efficiency, and better performance. The regeneration of adsorbent material and desorption of the captured CO_2 is also straightforward, especially in the temperature swing adsorption process. Further, given that less energy-intensive CO_2 desorption is efficiently achieved by heating the adsorbent under atmospheric conditions, the capture system involves low capital and operating costs. The Finite Difference Method, a one-dimensional model, showed that for a specified operating range and adsorbent material, optimal CO_2 adsorption from flue gases was accomplished at elevated temperatures, high purge flow rates, and lower adsorbent bed lengths [122].

5. Cryogenic distillation

From an environmental viewpoint, cryogenic CO_2 capture is better than other CO_2 capture methods, given that no chemical solvents or membrane materials requiring periodic swap/replacement are used. However, specially designed separation equipment is needed to address the freezing issue of CO_2. The energy-intensiveness issue is addressed by "a cryogenic distillation-based CO_2 capture method [123] from natural gas with high ethane content, such as shale gas or oilfield-associated gas, to use the azeotropic characteristics of CO_2 and ethane mixture to avoid CO_2 freeze-out using propane precooled mixed refrigerant system and reduce energy consumption through process integration with natural gas liquefaction and ethane recovery". CO_2 separation and ethane recovery are then carried out by extractive distillation. The concept of direct atmospheric cryogenic carbon capture and a broad outline of the research scenario is presented [22]. Cryogenic distillation offers the advantages of producing pure CO_2 in solid, liquid, or gaseous form with easy scale-up for large capacities. Given the high energy costs of desublimating CO_2, DAC could be considered for locations with ultra-low temperatures and strong and consistent wind patterns to offset the process's energy costs. There is a scope for further research and technological developments to improve the feasibility of DAC.

6. Membranes

Membrane technology [124], due to its distinct features such as its low carbon footprint, solvent-free, energy efficiency, easily scalable modular design, and low capital and operating costs, is an appealing CO_2 capture approach. However, fabricating highly selective and permeable defect-free membranes requires precision for their large-scale commercial applications [125]. Hollow fiber membranes with a high surface area-to-volume ratio, packing density, and flow are effective in CO_2 capture. PTFE hollow fiber membrane has the greatest inner fiber diameter of 1.499 mm and good performance [126]. The CO_2 source, process conditions, feed gas composition and quantity, desired separation level, and cost determine membrane material selection. For example, a membrane with high selectivity is generally favored for high-purity product requirements, and highly CO_2-permeable membranes are used to process large quantities of gases. A single-stage membrane system was used to compare various membrane systems for CO_2 enrichment. Single- and two-stage membrane systems were simulated for the techno-economic feasibility evaluation for CO_2 capture from fossil fuel-based advanced supercritical and natural gas combined cycle power and cement plants. The results revealed that a single-stage membrane system could enrich CO_2 from flue gases from advanced supercritical power and cement plants to 60 vol% CO_2 in the permeate stream. For further enrichment for very lean O_2-containing captured CO_2 streams for CO_2 utilization in chemicals or fuel manufacture, a second-stage membrane unit or hybrid systems, such as membrane absorption or membrane cryogenics [127], are needed. Enriching the flue gases from a natural gas combined cycle power plant containing 5 vol% CO_2 to 40 vol% in the captured stream was reportedly challenging. CO_2 enrichment of low-concentration CO_2 from point sources, <1% in the CO_2-N2 mixture, to over 99% purity was obtained via a three-stage system using poly[bis((2-methoxyethoxy)ethoxy)phosphazene] (MEEP)-based membrane. The techno-economic evaluation of CO_2 capture for the various capture routes, using a model, revealed the capture costs as chemical absorption < MEEP-based membrane < pressure swing adsorption < cryogenic < Pebax-based membrane technologies [128]. Molecular-level studies give a better understanding of hybrid systems design. Integrating membranes with water-lean solvents in a hybrid system enhances CO_2 separation [129]. Additionally, incorporating a CO_2 separation membrane in the DAC system reduces the solvent evaporation losses. An exhaustive review on the different aspects of zeolite membranes [23], such as syntheses, design, physicochemical characteristics, strength, flow and mass transfer mechanisms, CO_2 capture selectivity, stability, operating parameters, and performance, is presented.

Hollow fiber membrane contactors (HFMCs) with thirty times more interfacial area [130] due to the highest surface area per volume are projected as a promising recourse to the conventional absorption–desorption columns, helping to reduce the traditional gas absorbers' size by tenfold and offering a remarkable CO_2 capture performance. However, the inherent wetting phenomenon substantially reduces the separation efficiency. The properties of HFMC systems based on the membrane types, such as porous, nonporous, dense, and composite membranes, are outlined [131], along with their respective advantages and disadvantages and scale-up, engineering, and economic feasibility features. Specific additives or membrane surface modifications of the polypropylene and polysulfone membranes showed improved wetting resistance. Polyvinylidene fluoride membranes that have high wetting resistance can be easily prepared, but their performance on a large scale is yet to be established. "The CO_2 loading capacity of the MDEA solution, promoted by potassium lysinate (KLys), was experimentally measured, and the solution's CO_2 removal efficiency was investigated for a CO_2-N_2 gas mixture employing computational fluid dynamic simulations in HFMC". The CO_2 loading capacity was found to be positively correlated with KLys concentration in the solution and negatively correlated with temperature, and the MDEA + KLys solution had a higher CO_2 removal efficiency than MDEA or MEA solutions [132]. Given that nano-absorbents depict substantially increased mass transfer and heat transfer [133] during CO_2 capture while retaining high selectivity and absorption capacity in the CO_2 chemisorption process, a zeolitic imidazolate framework-8 (ZIF-8) with high specific surface area and a lot of active surface charges was blended with the MDEA + KLys solution to improve the CO_2 removal efficiency further. The CO_2 absorption efficiency of ZIF-8 nano-absorbent is better than that of other nano-absorbents, such as carbon nanotubes.

Pure MOF membranes, MOF-based composite membranes, and MOF-covalent organic framework membranes were investigated [134] with a particular focus on computational studies for post-combustion (CO_2/N_2), pre-combustion (CO_2/H_2), and natural gas sweetening (CO_2/CH_4) CO_2 capture. "The key advances and future directions in the development, modeling, and testing of early MOF membranes, ionic liquid/MOF membranes, ultrathin MOF membranes, MOF-covalent organic framework membranes, MOF glass membranes, MOF-based mixed-matrix membranes", are highlighted [134], along with potential opportunities and challenges associated with the computational modeling of MOF-based membranes and carbon capture applications. Transport membranes, carbon molecular sieving membranes, mixed matrix membranes, composite membranes, and poly(ionic liquids)-based membranes are discussed [135]. Electrochemical CO_2 capture with bipolar membranes as the main component is considered advantageous, efficient, more suitable, and more sustainable than non-electrochemical methods [136], with low floor space requirements and easy modular scale-up. Thin-film composites and scalable surface-engineering nanotechnologies employed to modify membrane surfaces, such as “atomic layer deposition, chemical vapor deposition, plasma treatment, direct fluorination, ion/electron beam treatment, ozone treatment, and surface-initiated polymerization”, are reviewed for their mechanisms, nanostructures, and CO_2 capture enhancement [137].

The CO_2 capture performance of a ceramic membrane contactor coupled with a catalyst-aided solvent (metatitanic acid powder) and aqueous MEA regeneration pathway [138] increased the CO_2 absorption rate due to the catalyst’s nanofluid effect in causing a concentration change in the liquid film by reducing the mass transfer resistance. Aqueous ammonia can absorb CO_2 in a PTFE HFMC at ambient temperatures and with an increasing liquid flow rate to obtain an equivalent performance to the MEA solvent [139]. However, long-term stability tests revealed that the absorption of aqueous ammonia was sustained in the first hour of the operation and then steadily deteriorated with time. This was attributed to the precipitation of ammonium salts on the membrane surface of the shell side, causing membrane fouling and membrane wetting. Composite membrane systems, such as thin-film composite, facilitated transport membrane (FTM), and mixed-matrix membranes appear to resolve these issues with HFMC. For example, FTMs overcome the wetting resistance, but there is a trade-off between permeability and selectivity. The long-term stability tests on mixed-matrix membranes showed a constant CO_2 flux and good capture performance. Coating varied materials in the membrane matrix further improves membrane characteristics and overcomes other problems. This led to the development of new materials for highly efficient CO_2 capture in HFMCs. Combining membrane technology with liquid absorbents, such as MEA, is a hybrid technology. Given the superiority of ammonia over other liquid absorbents, such as lower regeneration energy, higher efficiency, and integration potential with other CO_2 separation methods, aqueous ammonia can effectively absorb CO_2 in HFMCs [139].

7. Conclusions

Carbon removal is a necessity and no longer a “nice-to-have”, offering business opportunities to implement an essential pathway to a sustainable future. CO_2 capture is the key component of a comprehensive CO_2 reduction strategy constituting capture, utilization, and storage (CCU). Post-combustion CO_2 capture is primarily intended for sources such as power plants and steel and cement production using fossil fuels which contribute to large-scale CO_2 emissions into the atmosphere. Different approaches have been extensively researched for their flexibility, ease of integration into existing facilities, and significant potential to advance decarbonization efforts.

For the absorption process, commonly used at the industrial scale, different solvents and their blends with additives are investigated to lower the desorption energy consumption. Catalytic solvent regeneration is explored to lower overall CO_2 capture costs. The adsorption process is energy-efficient and eco-friendly. Researchers are examining high interfacial area sorbents and bio-based sorbents for selectivity and high CO_2 loading, which are required for large-scale operations in fixed-bed and fluidized-bed configurations. The Kawasaki CO_2 Capture (KCC) process tackles the energy-intensive nature of traditional solvent-based absorption processes by employing amine-impregnated sorbents in fixed-bed and moving-bed systems and successfully demonstrating bench (5 t-CO_2/d) [140] and pilot (40 t-CO_2/d) [141] scales at a coal-fired power plant of Kansai Electric Power Company, Inc. The compact KCC systems are capable of capturing a large quantity of CO_2 by varying the adsorbent circulation rate for large-scale applications and desorbing with low-temperature waste heat (around 60 °C), enhancing CO_2 capture productivity. Thus, commercialization of the KCC technology appears imminent.

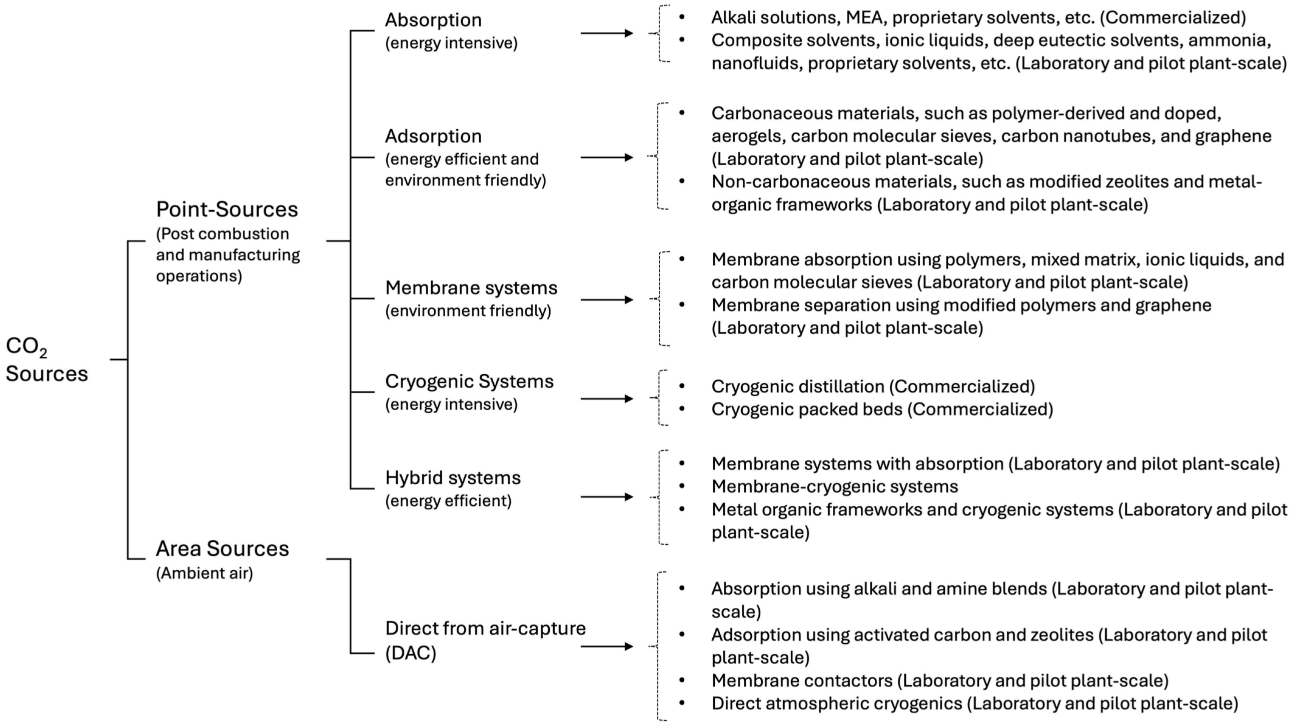

Figure 1 • CO_2 capture pathways.

Process intensification can lower energy requirements for cryogenic CO_2 capture, which does not involve solvents or membranes. For an effective capture of CO_2 and its enrichment for commercial applications, hollow fiber polymeric and ceramic membrane materials are being researched for the contactors. Efforts are underway to perfect fabricating methods for highly selective and permeable defect-free membrane systems. A novel, scalable technique has been developed [142] to create atom-thin, ultra-strong, porous graphene membranes that selectively filter CO2 from gas mixtures. This breakthrough can overcome the long-standing hurdle of producing large-area membranes with high CO_2 selectivity. The method appears promising, as it will drastically cut the membrane costs, enhance membrane quality, and boost performance, facilitating large-scale commercial applications. Advances in CO_2 capture pathways [143], including cost-effective hybrid systems, are nearing commercialization. Membrane–cryogenic hybrid CO_2 capture systems leverage the synergy between these two processes with the advantages of 98% CO_2 capture and low energy consumption [144]. Advances in membrane materials, refined process design, and optimized operating parameters could improve system performance further. CO_2 capture pathways—commercialized and at different stages of laboratory and pilot plant scale development—are exhibited in **Figure 1**. Alongside efficient and cost-effective capture, converting CO_2 into valuable green organic compounds is a crucial and challenging task [26] towards decarbonization and mitigating climate change-related issues.

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