



A Comprehensive Review of CO₂ Capture and Separation Methods

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ABSTRACT

As the global demand for energy continues to grow rapidly, the use of fossil fuels in power plants is leading to an increase in CO₂ emissions. In order to meet the climate change target set at the 21st Conference of the Parties (COP 21), effective strategies are necessary to reduce industrial emissions. One of the most promising methods for accomplishing this is carbon capture and storage (CCS), which involves capturing and separating CO₂ generated in thermal power plants. This review explores different approaches and recent advancements in CO₂ capture and separation techniques. Carbon capture can be divided into two main categories based on whether CO₂ separation is required in the gas stream. The study compares novel methods like oxy-combustion and chemical looping combustion with traditional post-combustion and pre-combustion carbon capture methods. It also examines the current state of technology, limitations associated with each approach, and commonly used techniques for separating CO₂ from gas mixtures. The review suggests further research and investigation to improve the capture system, considering factors such as technological maturity, economic viability, and the need for a better understanding of combustion systems.

Keywords: CO₂ capture; clean energy technologies; emission reduction; allam cycle; post-combustion; pre-combustion; flue gas treatment

1. INTRODUCTION

The rapid growth of industrialization in the past century has resulted in a significant demand for power. The most common method of power generation involves the use of fossil fuels, which leads to the emission of CO₂, a major greenhouse gas component. In fact, approximately 65% of GHG emissions come from different power industries and the fossil fuel-based energy sector [1]. Given the increasing global concerns about climate change caused by GHGs, there is a pressing need for research focused on reducing these emissions. Meeting the goals set at the COP-21 agreement, held in Paris in 2015, requires substantial reductions in CO₂ emissions to keep the global average temperature rise below 2°C, with efforts aimed at limiting it to 1.5°C [2]. To address emissions, research efforts are underway to explore the use of renewable resources as alternatives to fossil fuels. One interesting approach involves converting CO₂ into organic compounds through photocatalytic reduction, enabling the production of renewable fuel feedstock. This process allows for the utilization of traditional fossil fuels as renewable energy sources. For example, Zhou et al. [3] demonstrated the use of the semiconductor ZnS for photocatalytic reduction of CO₂ to formate (HCOO⁻), while Sharma et al. [4] utilized sulfide-based photocatalysts, such as Cu₃SnS₄, for the reduction of CO₂ to CH₄. Although these methods show promise, no technology has fully replaced conventional fuels yet. While nuclear and renewable energy sources are expected to play a significant role in low-carbon power generation, safety, and other concerns limit their complete substitution for fossil fuels [5]. Therefore, it remains essential to find ways to use conventional fuels for power generation while reducing CO₂ emissions. One solution that offers the potential for reducing emissions is Carbon Capture and Storage (CCS) technology. This process involves capturing CO₂ emitted from power plants and storing it in a secure location to prevent its release into the environment. The International Energy Agency (IEA) has projected that CCS should contribute to 17% of CO₂ abatement by 2035, ensuring cost-effectiveness [6]. To achieve this goal, ongoing research efforts worldwide are focused on developing new technologies. However, the primary challenge for the widespread adoption of CCS is its high cost, which increases electricity prices. Currently, the estimated cost of capturing CO₂ using established technologies is at least \$60 per metric ton of CO₂, making CCS economically unattractive. This high cost discourages investments in the energy market. Consequently, researchers in developed countries are striving to reduce the cost of carbon capture to around \$20 per metric ton of CO₂ [6]. Furthermore, it is crucial to consider the environmental impact of these methods, as reducing CO₂ emissions may inadvertently lead to increased emissions of other pollutants that can affect the environment [7]. In terms of carbon capture technologies, there are two main categories: those that involve separation of CO₂ and those that do not. This review focuses on examining these processes, assessing the current state of carbon capture technologies, and identifying areas that require further research.

2. CARBON CAPTURE WITH SEPARATION

The process of separating CO₂ from a gas mixture is a critical requirement in this context and can occur either prior to or following combustion. In the case where the gas stream, containing carbon dioxide, is formed before combustion, it is referred to as precombustion carbon capture. In such instances, the gas stream primarily consists of CO₂ and H₂. Conversely, if the gas stream is formed after combustion, it is known as post-combustion carbon capture,

with CO₂ and N₂ being the main constituents. Numerous technologies are currently employed or being developed to separate CO₂ from gas mixtures, and these separation techniques are generally applicable to both precombustion and post-combustion processes.

2.1 PRECOMBUSTION CARBON CAPTURE

This method presents an alternative to directly combusting fuel in a combustor [8]. It involves converting the fuel into a combustible gas, which is then used for power generation. Before combustion, CO₂ is separated and stored from the gas generated by fossil fuels [9]. A schematic diagram of the process is illustrated in Figure 1 [10].

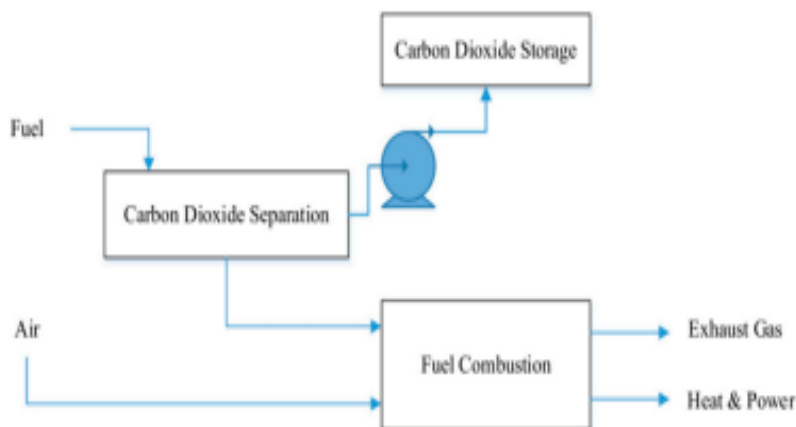
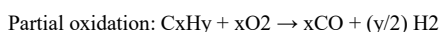
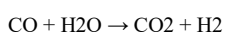


Figure 1. Schematic diagram of the pre-combustion carbon capture process [10].

Initially, synthesis gas (syngas), which is primarily a mixture of H₂ and CO with a trace of CO₂, is produced from fossil fuel. This can be achieved through steam reforming by adding steam to the fuel [11]. Another method is supplying pure oxygen after separating it from the air and introducing it to the fuel, known as partial oxidation for liquid or gaseous fuels and gasification for solid fuels [11]. The reactions for these processes are as follows [11]:



The syngas produced is then converted from CO to CO₂ through a water-gas shift reaction [11]:



The products of the water-gas shift reaction remain under high pressure, facilitating the removal of CO₂ at ambient temperature. The remaining gas consists mainly of hydrogen with minimal impurities and is used to generate power in a combined cycle power plant. The high pressure (typically 2-7 MPa) and high concentration of CO₂ (15-60% by volume) before the CO₂/H₂ separation stage result in lower energy requirements for CO₂ separation and compression compared to post-combustion carbon capture [6]. To mitigate the energy penalty caused by air separation and reforming or gasification processes, Sorption Enhanced Water Gas Shift (SEWGS) technology can be employed, integrating the water-gas shift reaction and CO₂ separation [12]. SEWGS increases the conversion rate of CO by removing CO₂ from the product of the water-gas shift reaction, resulting in additional CO₂ emission reduction [13]. While the process is similar for any fossil fuel, using fuels other than natural gas requires additional refining stages to address higher contaminant production [11]. Currently, the main research focus of precombustion carbon capture is on implementing this method in Integrated Gasification Combined Cycle (IGCC) power plants. An IGCC layout is shown in Figure 2. Oxygen is separated from air in a cryogenic air separation plant [14]. The oxygen is then passed to a gasifier where coal is gasified at high pressure and temperature, producing syngas. After cooling and preliminary cleaning, the syngas undergoes a water-gas shift reaction in a water-gas reactor, converting it to H₂S, H₂, and CO₂. Several cleaning steps follow to remove sulfur, mercury, water, and other impurities, resulting in syngas consisting mainly of CO₂ and H₂. The CO₂ is then captured, and the hydrogen is used for power generation. Physical solvents are commonly used for CO₂ separation from syngas in commercially developed technologies.

2.2 POST-COMBUSTION CARBON CAPTURE

This technique allows for the capture of CO₂ in existing power plants without requiring significant modifications to the plant. This gives it an advantage over other CCS (Carbon Capture and Storage) processes in terms of easier retrofitting [28-30]. It is considered the simplest method for capturing CO₂, involving the removal of CO₂ from the exhaust flue gases of power plants. Typically, these flue gases exit at atmospheric pressure and contain a relatively low concentration of CO₂, as indicated in Table 1. The low concentration of CO₂ poses a challenge because the driving force for capture is insufficient [31]. To handle the large volume of flue gases, large-sized equipment, and high capital costs are necessary. Therefore, identifying a cost-effective approach

to capture CO₂ from flue gas is crucial. Additionally, the flue gas contains various contaminants such as SO_x, NO_x, and fly ash, which further increase the cost of separation using existing technologies [32].

Method	Concentration of CO ₂ (Vol. %)
Coal-fired Boiler	14
Natural gas-fired boiler	8
Natural gas combined cycle	4
Natural gas partial oxidation	40
Coal oxygen combustion	>80

Table 1. Amount of CO₂ in flue gases of power plants [31].

Separating CO₂ from flue gas presents several challenges. Equipment must be designed to withstand the high temperature of the flue gas, and the gas needs to be cleaned before CO₂ separation can occur.

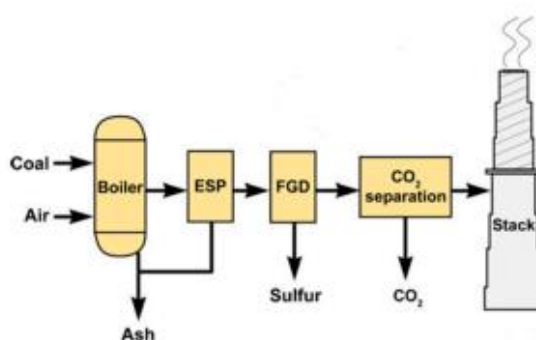


Figure 2. Schematic diagram of a simplified flue gas cleanup process for post-combustion carbon capture [32].

Merkel et al. [32] proposed a flow process to clean up the gas, as illustrated in Figure 2. The hot exhaust gas from the boiler undergoes treatment in an electrostatic precipitator (ESP) to remove large particulates. Then, a flue gas desulfurization unit (FGD) removes sulfur products. Post-combustion carbon capture technology is designed to treat the outlet gas from the FGD. At this stage, the gas mixture contains approximately 10-14% CO₂, predominantly mixed with nitrogen. Figure 4 depicts a schematic of a coal-fired power plant utilizing solvent-based absorption post-combustion carbon capture. In this setup, coal is pulverized and combusted with air to generate heat. The heat is used to produce steam, which, in turn, drives three different steam turbines operating at various pressures. The low-quality exhaust steam is condensed in a condenser and returned to the boiler. The exhaust flue gas from the boiler goes through a cleaning process to remove sulfur, ash, NO_x, and other impurities. After the final stage of cleaning, the gas is directed to the CO₂ capture process. When natural gas is used as fuel, the complexity of the system is significantly reduced. Figure 5 illustrates a typical layout of a post-combustion carbon capture combined cycle power plant that employs natural gas as fuel. Natural gas is combusted with compressed air, and the resulting product is expanded through a gas turbine to generate power. The exhaust from the gas turbine remains at a high temperature, which is utilized to produce steam for additional power generation through a steam turbine. The cooled flue gas then enters the CO₂ capture process. Figure 5 depicts a solvent-based CO₂ capture system using MEA (Monoethanolamine). MEA scrubbing removes CO₂ from the flue gas in the absorber column, leaving clean gas for exhaust. The MEA is subsequently purified in the stripper column for reuse in the absorber column. CO₂ is captured from the stripper column and compressed for storage. MEA is the most commonly used method for separating CO₂ from flue gas, although other technologies are also employed for this purpose.

2.3 CO₂ SEPARATION TECHNOLOGY

Numerous separation technologies exist for the separation of CO₂ from gas mixtures, and these technologies can be broadly categorized into five sectors: absorption, adsorption, clathrate hydrate process, membrane technology, and calcium looping carbon capture [10]. In the absorption process, research is primarily focused on the development and enhancement of various solvents to improve their performance in capturing CO₂. Different solvent systems, such as aqueous amine solutions like monoethanolamine (MEA) and advanced solvents like ammonia-based systems, are being studied to increase their CO₂ absorption capacity and selectivity. Additionally, efforts are being made to optimize the operating conditions and improve the energy efficiency of absorption processes. Adsorption technology emphasizes the exploration of new materials and the modification of existing ones to enhance their CO₂ adsorption capabilities. Researchers are investigating novel adsorbents, including metal-organic frameworks (MOFs), zeolites, and activated carbon, with tailored properties to selectively capture CO₂ from gas mixtures. The focus is on improving the adsorption capacity, kinetics, and stability of these materials to make them more efficient and economically viable. The clathrate hydrate separation process involves the formation of hydrate crystals with

CO₂ molecules entrapped within a lattice structure. Research in this area is focused on exploring different thermodynamic promoters and additives to enhance the formation and stability of CO₂ hydrates. By optimizing the operating conditions and understanding the kinetics of hydrate formation and dissociation, scientists aim to improve the efficiency of this separation process. Membrane technology utilizes selectively permeable membranes to separate CO₂ from gas mixtures based on differences in molecular size, solubility, or diffusion rates. Ongoing research involves the development of advanced membrane materials, such as polymer membranes, mixed matrix membranes, and ceramic membranes, with improved CO₂ separation performance. Efforts are also being made to enhance membrane stability, selectivity, and permeability for better process efficiency. Calcium looping carbon capture is a promising technology that utilizes calcium oxide (CaO) as a sorbent to capture CO₂ from gas streams. The process involves the cyclic reaction between CaO and CO₂ to form calcium carbonate (CaCO₃), which can be subsequently calcined to release pure CO₂. Researchers are focusing on optimizing the reaction conditions, sorbent properties, and reactor designs to enhance the carbon capture capacity, sorbent stability, and overall process efficiency. Research and development in these five sectors are crucial for advancing CO₂ separation technologies and improving their performance in terms of efficiency, selectivity, and cost-effectiveness. Continued efforts in these areas are essential for enabling large-scale deployment of carbon capture and storage (CCS) technologies to mitigate greenhouse gas emissions.

2.4 ABSORPTION

The method of using solvents to absorb CO₂ and separate it from a gas stream has been employed on an industrial scale for over 50 years [48]. This absorption process can be classified into two types: physical absorption and chemical absorption, as shown in **Figure 6**. In chemical absorption, the solvent reacts with CO₂ to form chemical compounds, which are later separated to obtain pure CO₂. In physical absorption, the solvent does not chemically react with CO₂ but physically absorbs it. This process is based on Henry's law, which states that the amount of gas dissolved in a solvent is proportional to the gas's partial pressure in equilibrium with the solvent [48].

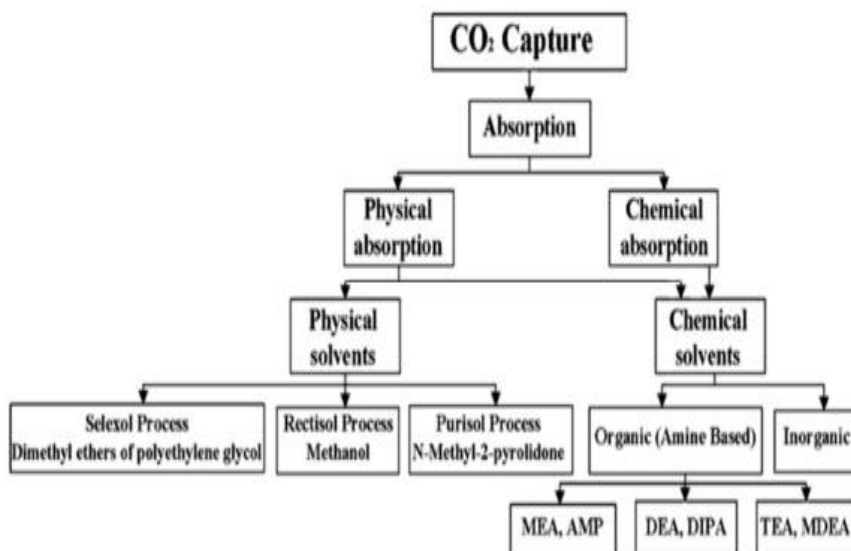


Figure 3. Classification of absorption processes for CO₂ capture [49].

Chemical absorption of CO₂ occurs in two stages. In the first stage, the gas stream is brought into contact with the solvent in a counterflow, where CO₂ is absorbed by the solvent. The solvent is then regenerated by heating in a stripping column to desorb CO₂, which is collected as pure CO₂ from the top of the column [10]. The regenerated CO₂-lean solvent is recycled back to the absorber [50]. The first stage of the process operates optimally at high pressures and low temperatures, while the second stage performs best at low pressures and high temperatures [10]. Chemical absorption is particularly suitable for capturing CO₂ at relatively low pressures, making it advantageous for post-combustion processes that utilize amine or carbonate solvents [48]. In physical absorption, organic or inorganic physical solvents are used that do not chemically react with CO₂. This process relies on Henry's law for vapor-liquid equilibrium. Physical absorption shows better performance than chemical absorption at higher partial pressures of CO₂, such as in Integrated Gasification Combined Cycle (IGCC) applications [11]. Physical solvents require less energy for regeneration, which is another advantage [52]. However, physical absorption processes have a downside: the solvent's capacity is highest at low temperatures, necessitating cooling of the gas stream prior to absorption, which reduces overall efficiency [52]. Commercially used physical absorption processes include Selexol, Rectisol, and Purisol [11]. A comprehensive comparison using Aspen Plus revealed that Selexol was more energy-efficient for CO₂ capture, thanks to its lower energy consumption for solvent regeneration and simpler process configuration [49]. Research has also shown that applying low-temperature CCS in an IGCC could yield even higher net efficiency compared to the Selexol process [53]. In post-combustion carbon capture, the partial pressure of CO₂ in the flue gas stream is typically very low. Therefore, research efforts for this process primarily focus on identifying suitable solvents. Numerous studies have been conducted on different processes and solvents to identify cost-effective absorption methods.

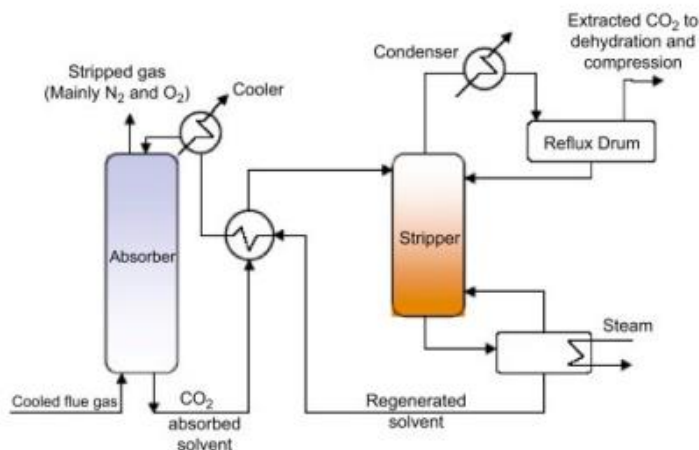


Figure 4. Schematic diagram of a CO₂ absorption plant [51].

2.5 ADSORPTION

Adsorption is a separation process that involves removing a component from a mixture using a solid surface. In contrast to absorption processes, adsorption relies on the formation of physical or chemical bonds between the solid adsorbent surface and CO₂, driven by intermolecular forces [31]. The adsorption process can absorb single or multiple layers of the gas, depending on factors such as pore size, temperature, pressure, and surface forces [54]. In adsorption, a column is initially filled with the adsorbent material, and the gas stream containing CO₂ is passed through the column. CO₂ adheres to the solid surface of the adsorbent until the adsorbent becomes saturated. The CO₂ is subsequently desorbed from the adsorbent through different cycles to achieve CO₂ adsorption [31]. There are four main regeneration cycles used for single-bed CO₂ adsorption: pressure swing adsorption (PSA), temperature swing adsorption (TSA), electrical swing adsorption (ESA), and vacuum swing adsorption (VSA). In TSA, the adsorbent's temperature is raised to break the chemical bonds and release CO₂. However, this method is costlier due to the additional energy required for heating the adsorbent bed and the time-consuming heating and cooling cycles for regeneration [48]. ESA, on the other hand, utilizes low-voltage electric current to heat the adsorbent rapidly using the Joule effect, enabling fast regeneration. However, ESA requires high-grade electrical energy compared to the low-grade heat energy used in TSA [55]. Pressure swing adsorption involves reducing the pressure of the adsorbent to achieve desorption, while vacuum swing adsorption is a specialized form of PSA used when the feed gas pressure is close to ambient pressure. VSA minimizes the additional energy required for achieving high pressure in PSA by using a partial vacuum downstream of the feed stage to draw the low-pressure feed gas [48]. These cycles can be combined with each other, offering flexibility in the adsorption process. Plaza et al. [56] presented a VSA process model using Aspen Plus for post-combustion carbon capture. A schematic of the regeneration process is shown in Figure 5. Pressure swing operation is advantageous when the partial pressure of CO₂ is high, while temperature swing adsorption is more suitable for low CO₂ concentrations in the gas stream. PSA becomes less efficient with low CO₂ concentrations [58]. Adsorption processes are preferred due to their high adsorption capacity at normal pressure and temperature, long-term stability, low regeneration cost, high adsorption rate, and lower energy requirements [59].

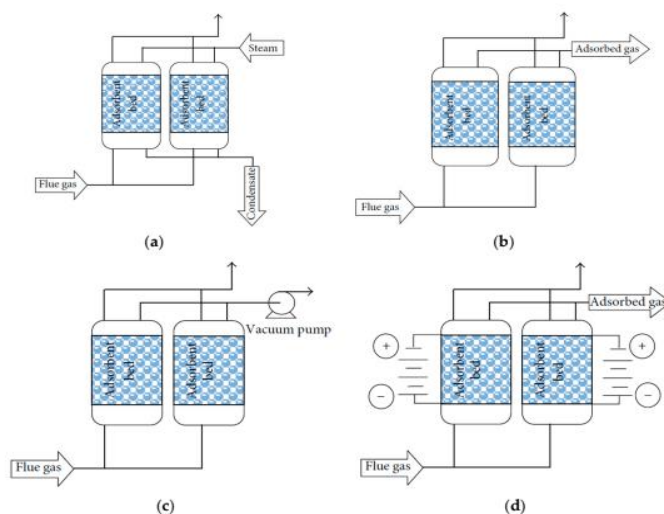


Figure 5. Schematic diagram of different adsorption regeneration cycles: (a) TSA (b) PSA (c) VSA (d)ESA [57].

The focus of research in adsorption processes is finding suitable sorbents to effectively separate CO₂ from gas streams. Various substances, such as zeolites, activated carbons, molecular sieves, hydrotalcite, and metal-organic framework materials, have been investigated for this purpose [60]. Garcia et al. demonstrated that the partial pressure of CO₂ is a significant factor when using activated carbon as an adsorbent [18]. Sorption-enhanced water gas shift (SEWGS) combines CO₂ adsorption with the water gas shift reaction, offering a more economical and energy-efficient alternative to amine scrubbing in absorption processes [61]. Hydrotalcite-based materials are particularly suitable for high-temperature adsorption, showing improved results when used in a sorption-enhanced WGS reactor for enhanced carbon capture [62].

2.6 MEMBRANE TECHNOLOGY

Membranes are barriers made of various materials that selectively separate different substances from mixtures through different mechanisms [31]. They can be composed of organic or inorganic materials and operate based on the solution-diffusion process. In non-facilitated membranes, the permeate, including CO₂, dissolves into the membrane and diffuses through it. The amount of CO₂ dissolved per unit volume is proportional to the partial pressure of CO₂ [63]. Non-facilitated membrane separation technology is particularly useful in pre-combustion capture, where the partial pressure of CO₂ is relatively high. Membranes can be utilized in different configurations such as spiral wounds, flat sheets, and hollow fiber modules. They can be selective or non-selective for specific acidic gases [64]. In carbon capture, membrane technology can be classified into two categories: gas separation membranes and gas absorption membranes. In a gas separation membrane system, the gas mixture containing CO₂ is introduced at high pressure into a membrane separator, typically consisting of parallel cylindrical membranes. CO₂ selectively permeates through the membrane, and it is recovered at a lower pressure on the other side of the membrane. In a gas absorption system, a microporous solid membrane is employed to separate CO₂ from the gas stream. Gas absorption systems exhibit high CO₂ removal rates due to minimized issues such as flooding, foaming, channeling, and entrainment. Furthermore, the equipment required for gas absorption systems is more compact compared to membrane separators [53]. The two systems are illustrated in Figure 6.

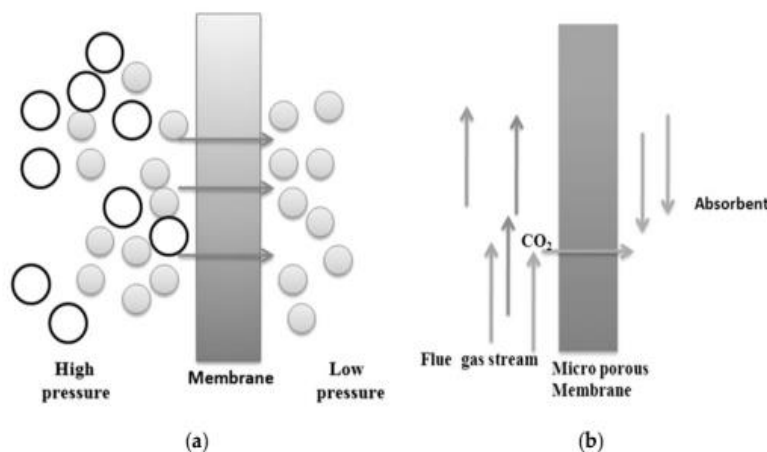


Figure 6. Principle of (a) gas separation membrane and (b) gas absorption membrane

Membrane technology offers several advantages, including the absence of problems such as weeping, entrainment, foaming, and flooding commonly encountered in packed column operations. Membranes also provide a higher surface area and better control of liquid and gas flow rates [65]. However, membranes exhibit reduced effectiveness at lower CO₂ concentrations and become impractical when the CO₂ concentration in the gas stream falls below 20% [66]. Therefore, they are not well-suited for post-combustion capture processes. One drawback of membranes is their limited lifetime, requiring periodic replacement. Additionally, membrane fibers present higher mass transfer resistance. For optimal performance, membrane pores should be fully occupied by the gas phase. When the liquid phase enters the membrane pores, mass transfer resistance increases, making the membrane economically unjustifiable. This phenomenon is known as membrane wetting. The desired condition is to completely fill the membrane pores with gas, but it is not always achievable. During overextended operational periods, membrane pores can become partially or fully wet [64]. Numerous studies have investigated the effect of wetting on mass transfer through different types of membranes and absorbents. Wetting significantly reduces absorption efficiency, even at low levels. Using ionic liquids as absorbents can improve efficiency by approximately 15% to 20% compared to pure water in counter-current and co-current flows [66].

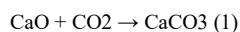
2.7 CLATHRATE HYDRATE PROCESS

Gas hydrates or clathrate hydrates, as described in [72], are ice-like crystal compounds formed by water molecules and various substances such as CO₂, N₂, H₂, and O₂. These small gas molecules are trapped inside water molecule cavities. The concentrations of gases within the hydrate crystals differ from their original concentrations in the gas mixture [73]. The concept of selectively separating CO₂ by forming hydrate crystals between the solid hydrate crystal phase and the gaseous phase is based on thermodynamics. The minimum pressure required to form hydrates at 273.9 K is 5.56 MPa. However, the pressure of syngas after the water gas shift reaction is typically 2-7 MPa, while post-combustion flue gas is close to atmospheric pressure.

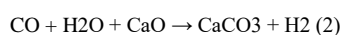
Therefore, compressing the gas stream is necessary to enhance hydrate formation rates [74]. The addition of promoters has been studied to reduce the equilibrium conditions for hydrate formation. One extensively studied promoter is tetrahydrofuran (THF), as mentioned in [75]. The equilibrium of hydrate formation decreases with the addition of THF at any temperature. Increasing the THF concentration leads to a decrease in hydrate formation pressure up to an optimum concentration of approximately 1 mol% THF. This allows for CO₂ separation without significant compression of flue gas. Moreover, the addition of 3.2 mol% propane to a CO₂/O₂ mixture can reduce the equilibrium pressure for hydrate formation by 50%, as reported in [76]. For precombustion fuel gas mixtures, the addition of tetra-n-butyl ammonium bromide (TBAB) reduces the hydrate phase equilibrium condition. The hydrate formation condition decreases with an increase in TBAB concentration up to the stoichiometric condition. Beyond that, the phase equilibrium increases with further TBAB concentration [77]. In [78], Park et al. showed that quaternary ammonium salts, including tetrabutylammonium fluoride (TBAF), can capture up to 95% of CO₂ from an integrated gasification combined cycle (IGCC) plant using a single step of hydrate formation. TBAF demonstrated better results than TBAB, although with lower gas uptake. Furthermore, recent studies have examined the impact of reactor types. Zheng et al. [79] investigated the effect of bed volume and reactor orientation on hydrate formation in precombustion carbon capture. They found that a horizontal orientation outperformed a vertical configuration. Low water saturation was also found to be preferable for enhancing hydrate formation.

2.8 CALCIUM LOOPING TECHNOLOGY

The calcium looping carbon capture system is an alternative method for capturing CO₂ from a gas stream, as described in reference [80]. It involves a direct reaction between CO₂ and CaO, resulting in the formation of solid calcium carbonate (CaCO₃), which can be easily separated from other gases. The main reversible reaction in this process is:



The forward reaction, known as the carbonation reaction, is exothermic, while the reverse reaction, called the calcination reaction, is endothermic. The carbonation reaction initially proceeds at a fast rate but eventually slows down [81]. Since the calcination reaction is endothermic, it requires a significant amount of heat supplied at high temperatures. This heat is often generated by the oxy-combustion of coal or natural gas within the calcination reactor [33]. After CO₂ is recovered from the calcination reactor, it is compressed and stored. The calcium looping process can be applied to both precombustion and post-combustion carbon capture. In precombustion carbon capture, the following key reaction occurs in the gasifier:



Using the calcium looping process in precombustion carbon capture offers advantages such as increasing the destruction rate of tar when hydrogen is used as a fuel and enhancing the conversion rate from CH₄ and CO to H₂ by removing CO₂ from the gas mixture [80]. While the main application of the calcium looping process is in post-combustion carbon capture [82], where limestone captures CO₂ from exhaust flue gases in a power plant using a circulating fluidized bed carbonator. The sorbent is then transferred to a calciner operating at higher temperatures. After regeneration, the sorbent is cycled back to the carbonator. The calciner utilizes oxyfuel combustion of coal or natural gas to generate the required heat.

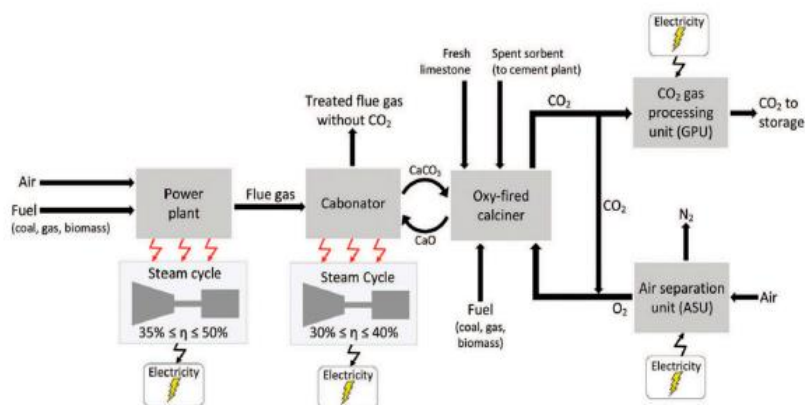


Figure 7. A schematic diagram of post-combustion carbon capture using Calcium looping [82].

3. CARBON CAPTURE WITH WATER CONDENSATION

This approach is relatively new in power generation, where pure oxygen is used for combustion instead of supplying air to the combustion chamber. Consequently, the combustion products primarily consist of CO₂ and steam. The CO₂ in the mixture can be captured by condensing the steam, eliminating the need for separate CO₂ separation technologies discussed earlier. As a result, this method is more economically feasible. When oxygen is produced using an Air Separation Unit (ASU), the process is referred to as oxy-combustion carbon capture. Another method of supplying oxygen is through the use of metal oxide in a process called chemical looping combustion [85,86].

3.1. OXY-COMBUSTION CARBON CAPTURE

In this method, the fuel is burned using nearly pure oxygen instead of air. The resulting flue gas consists primarily of a mixture of water and carbon dioxide. In a conventional power plant, fuel is combusted in the presence of air, and the nitrogen in the air acts as a temperature moderator. However, in oxyfuel combustion, since there is no nitrogen present in the combustor, the flame temperature can become excessively high. To control the temperature within the desired range, recycled CO₂ is introduced into the combustor along with pure oxygen. Another approach to regulating the flame temperature is by injecting steam into the combustion chamber [87]. After the combustion process, water is separated from the product through condensation. The captured CO₂ is then purified and compressed to a supercritical condition for transportation or reuse in the cycle. Figure 8 provides a flow sheet illustrating the concept of oxyfuel combustion.

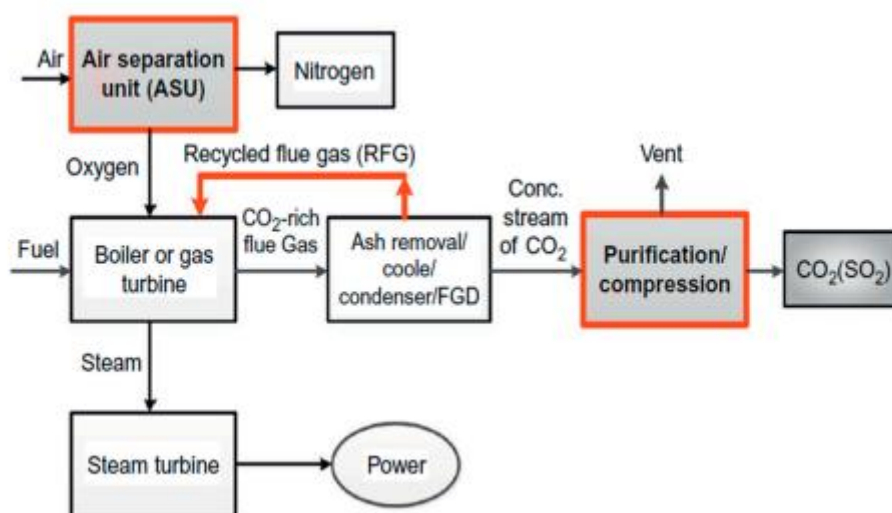


Figure 8. Flowsheet of oxy-fuel combustion technology for power generation with CO₂ capture [5].

Oxy combustion, which involves burning fuel in nearly pure oxygen instead of air, presents different reaction pathways and combustion characteristics compared to conventional air-fuel combustion [88]. Extensive research is required to thoroughly understand and utilize this method due to its unique combustion characteristics. Oxyfuel combustion offers several advantages over conventional combustion methods. In conventional air firing systems, a significant amount of nitrogen is present, which consumes a considerable amount of heat before being released into the environment. However, in oxy-combustion, the absence of bulk nitrogen in the combustion environment reduces or eliminates the production of NO_x. Moreover, there are no significant pollutants in the combustion products. This makes oxyfuel combustion a more cost-effective method compared to other carbon capture technologies. However, one of the main challenges is the high operational cost associated with producing high-purity oxygen and compressing CO₂ after combustion [89]. The production of oxygen with high purity at a reasonable cost remains a major hurdle in oxyfuel combustion. Wu et al. [90] highlighted different research on oxygen separation methods for oxyfuel combustion. They suggested that membrane methods are more economical and simpler compared to cryogenic methods, while adsorption technology is not yet suitable for large-scale implementation. Chemical looping air separation is considered a highly promising and efficient technique for oxyfuel combustion. Ongoing research aims to improve understanding and optimize the oxy-combustion process, including boiler design and a deeper understanding of combustion procedures. Compact boiler designs can reduce the cost of power generation, decrease flue gas volume and associated heat loss, and reduce emissions of SO_x and NO_x while improving combustibility. Oxy combustion has initially been applied to industrial processes and later explored for CO₂ generation for oil recovery. The incorporation of recycled flue gas was subsequently introduced to produce a high-purity CO₂ stream for oil recovery and mitigate the environmental impact of fossil fuel power plants [92, 93]. Oxy combustion can be applied to coal and natural gas, classified as oxy-pulverized coal processes, oxy-coal-fired boiler processes, CO₂-based cycles, or water-based cycles [94]. Comparisons with other systems have shown that oxy-fuel systems have 1-5% less efficiency loss compared to post-combustion capture, with pressurized systems gaining approximately 3% more efficiency. The ASU may require more power in a pressurized system, but it saves significant power during CO₂ compression. The absorptivity and emissivity of flue gases increase in oxyfuel combustion due to higher partial pressures. The ratio of the recycle stream depends on the fuel type, heat recuperator arrangement, and recycling strategy. The oxy-fuel environment has no significant influence on the devolatilization process of solid fuel, but it does result in longer ignition delays compared to conventional systems. Researchers have investigated the effect of recycled CO₂ on the combustion environment. The incorporation of CO₂ instead of N₂ in the combustion environment can decrease the burning velocity or speed of flame propagation [97]. Studies have shown contrasting results regarding the flame speed in an oxy-fuel environment compared to an air-fuel environment, potentially attributed to different experimental setups [97, 98]. Water vapor has also been found to affect flame propagation speed, with an increase in steam concentration leading to a decrease in flame velocity [99]. The presence of CO₂ in the oxyfuel environment strengthens the radiation effect of CH₄, and the maximum flame temperature is influenced by factors such as the presence of CO₂, H₂O, and pressure [88, 101]. Limited research has been conducted on ignition, flame stability, and flame extinction in an oxy-combustion environment. Ignition delay is typically longer in an oxy-fuel environment due to the participation of CO₂ in chemical reactions, higher heat capacity, and different collision efficiency [102, 103]. The flammability of oxy-methane flames is also influenced by the addition of CO₂ and steam, affecting the upper and lower flammability limits

[104]. A novel approach to power generation, known as the Allam cycle, has been proposed by Allam et al. [107]. This cycle utilizes oxyfuel combustion and operates with pressurized supercritical CO₂ as the working fluid. The key advantage of the Allam cycle lies in the high heat capacity of high-pressure CO₂ compared to low-pressure CO₂, eliminating the need for water vaporization and condensation in the cycle. In the Allam cycle, fuel is burned in pure oxygen in a high-pressure combustor, generating a feed stream with pressures ranging from 200 to 400 bar. This stream is expanded in a single turbine with a pressure ratio of 6 to 12. The heat from the high-temperature turbine exhaust is transferred to a high-pressure recycled CO₂ stream in a recuperator. The recycled stream is then sent back to the combustor to control the turbine inlet temperature. The authors reported theoretical thermal efficiencies of 59% for natural gas fuel and 52% for coal fuel, while inherently capturing CO₂. A 50 MWth demonstration plant utilizing the Allam cycle with natural gas as fuel has been recently completed in La Porte, Texas, and plans are underway for a commercial 300 MW plant to showcase the advantages of this cycle [114]. As CO₂ capture is integrated within the cycle, no additional measures are required, making it a potentially cost-effective alternative to conventional power plants employing CCS. Ongoing research aims to optimize the parameters of the Allam cycle. Key factors affecting its efficiency include turbine inlet temperature and pressure, turbine outlet pressure, temperature difference on the hot side of the primary heat exchanger, and the performance of the air separation unit within the cycle.

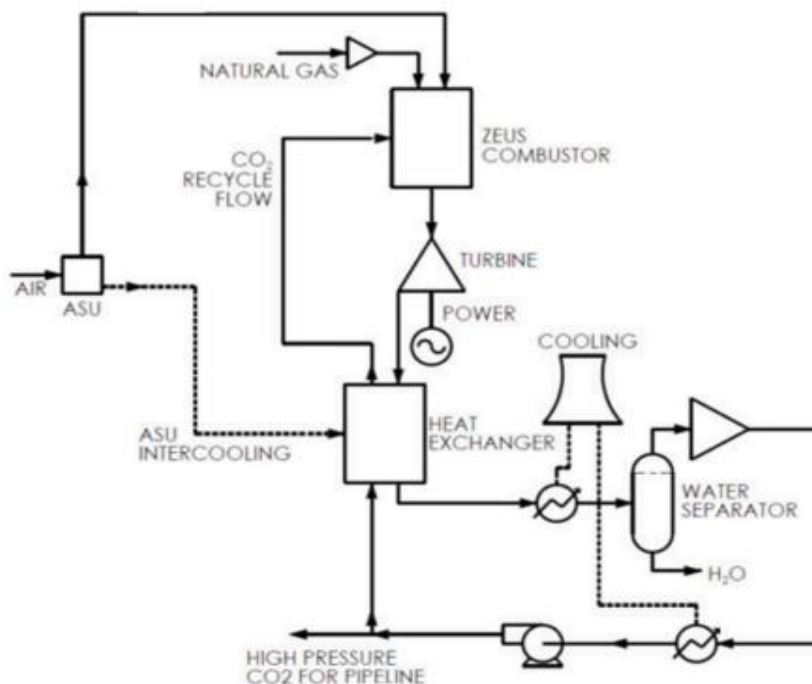


Figure 9. Schematic of the Allam power cycle operating on natural gas as fuel [107].

4. COMPARISON OF THE METHODS

Carbon capture processes that require the separation of CO₂ have been developed over time and have reached a certain level of maturity, with established full-scale applications. Numerous experimental and numerical modeling studies have been conducted on these processes. One advantage of post-combustion capture is its easy integration with existing power plants. However, the concentration and partial pressure of CO₂ in flue gases are typically low, necessitating additional energy and costs to achieve the minimum required concentration for transportation and storage. Chemical absorption processes for separation can lead to solvent degradation and severe corrosion of equipment, resulting in high costs for solvents and equipment, increasing the cost of electricity production by around 70% [118]. Research is ongoing to develop new solvents and reduce the cost of carbon capture. The large size of the equipment also contributes to high capital and operating costs in this method. Pre-combustion carbon capture is predominantly used in process industries, and there are full-scale CCS plants in some industries that utilize this method [11]. The gas mixture in this process contains a higher amount of CO₂ compared to conventional flue gas mixtures. Due to the higher pressure and lower gas volume, less energy is required in pre-combustion capture compared to post-combustion capture. However, there is still a significant energy penalty associated with pre-combustion capture. It is mainly used in integrated gasification combined cycle (IGCC) technology, which requires a large auxiliary system, leading to high capital costs compared to other systems.

On the other hand, carbon capture processes that do not require separation are relatively new in power generation, and there are no full-scale operational plants based on these processes. Some pilot-scale operations and subscale demonstration plants are under development, particularly using oxyfuel combustion [10, 30, 94]. One promising example is the 50 MWth demonstration power plant built in Texas by Net Power using the Allam cycle concept, aiming for near-zero emissions. This method offers advantages such as equipment size reduction, compatibility with various types of coal, and no need for an onsite chemical plant. However, it requires a large amount of high-purity oxygen, necessitating an energy-intensive Air Separation Unit (ASU) for oxygen production. Membrane-based technology for air separation may offer competition to cryogenic ASU through greater integration into the power

cycle [133]. The ASU and CO₂ compression units used in this process significantly decrease net power output. Furthermore, there are technical uncertainties that require further research to understand full-scale operation. Nonetheless, since no additional cost is required for CO₂ separation, this process remains promising for low-cost electricity production while achieving near-zero emissions. The process of Calcium Looping Combustion (CLC) for carbon capture is still in the preliminary stage and has not been implemented commercially. Further research is needed to fully harness its potential. One advantage of CLC is the absence of flame, resulting in no thermal NO_x production, and the outlet stream from the air reactor is environmentally benign [93]. Developing appropriate oxygen carriers for CLC could make it more attractive compared to other processes. Bituminous coal is considered for coal-based power plants due to its extensive use in power production [30]. The Selexol process is considered for pre-combustion carbon capture in an IGCC GE-type gasifier. For chemical looping combustion, ilmenite and nickel aluminum oxide were used as oxygen carriers for coal-based and natural gas-based power production, respectively [134, 135]. When coal is used as a fuel, the CLC exhibits the same efficiency as the base combustion technology using pulverized coal without any capture. Reduction in efficiency is the highest in pre-combustion carbon capture. Post-combustion and oxy-combustion carbon capture show an almost similar drop in efficiency. An interesting observation in this comparison is the efficiency of the Allam cycle [30]. When natural gas is used as fuel, the pre-combustion carbon capture shows a 14% drop in the efficiency from the reference powerplant whereas the post-combustion carbon capture shows an 8% drop. The traditional oxy-combustion process exhibits an efficiency of 44.7%. Chemical looping combustion indicates only a 4% drop in efficiency from the reference plant. The Allam cycle shows an extraordinary performance whose efficiency happens to be over 3 percentage points higher than that of the reference combined cycle without CO₂ capture [30]. From the efficiency comparison of, it may be concluded that the chemical looping combustion and the Allam cycle are expected to be the leading technologies in the near future for fossil fuel-based power generation. The 50 MWth Allam cycle provides the basis for the deployment of large-scale facilities. Currently, 300 MW natural gas-fired plants are under development. The chemical looping method is not yet technologically ready to implement on an industrial basis. The method is still in the investigation stage. More experimental data are necessary before large-scale commercialization [134,135]. Conventional carbon capture process results in a reduction of efficiency. More fuel is burnt per unit of electricity production due to this inefficiency which leads to more production of CO₂. Also, the processes used for capturing carbon dioxide may affect the environment in different ways other than direct emission of CO₂. For example, different substances used for separating and capturing CO₂ may have undesired effects on the human body and environment. Using a solid sorbent covered with coating was experimented with to reduce the formation of dust from the substance [136]. This could also reduce the capacity of the substance to capture carbon dioxide. Also, stripping of organic solvent from membranes and sorbents is suggested to prevent undesired odor. Before employing carbon capture, it should be ensured that reducing CO₂ is not being achieved at the cost of other environmental impacts. Life cycle assessment of the plants is necessary to properly understand the environmental impacts of the carbon capture methods [7,137,138,139,140,141]. Schreiber et al. [7] used the life cycle assessment (LCA) methodology for post-combustion carbon capture using MEA whose impact on the environment and human health was investigated for five power plants. The global warming potential (GWP), human toxicity potential (HTP), acidification potential (AP), photo oxidant formation potential, and eutrophication potential (EP) were considered as impact categories. As expected, GWP was much lower with MEA compared to the power plants without capture whereas HTP was three times higher with MEA plants. Schreiber et al. [7] concluded that upstream and downstream processes such as emissions from fuel and material supply, waste disposal, and wastewater treatment influence the environmental impact measures for power plants with carbon capture. Viebahn et al. [137] revealed about a 40% increase in AP, EP, and HTP when post-combustion carbon capture was implemented in a power plant. A similar result was found by Veltman et al. [138]. They showed that a power plant with post-combustion capture yields a 10 times increase in toxic impacts on freshwater compared to a plant without capture. Impacts on other categories were negligible. Degradation of MEA resulted in the emission of ammonia, acetaldehyde, and formaldehyde. Cuellar et al. [139] compared the life cycle environmental impacts of carbon capture and storage with carbon capture and utilization. GWP with utilization was much greater than that with storage. The highest reduction of GWP was found for pulverized coal and IGCC plants employing the oxyfuel capture method as well as combined cycle gas turbine plants equipped with a post-combustion capture technology. Pehnt et al. [140] showed that a conventional power plant operating on coal with post-combustion carbon capture would result in an increase in the environmental impact in almost all categories except GWP. Solvent degradation and energy penalty due to the CO₂ capture process are the main reasons for this increase. Precombustion capture showed a decrease in all the environmental impact categories compared to a conventional power plant. They identified oxyfuel combustion as the most potential process to reduce all the environmental impact categories if co-capture of other pollutants can be achieved. Nie et al. [141] investigated the comparative environmental impacts of post-combustion and oxy-fuel combustion carbon capture. Their analysis showed that almost all environmental impact categories except GWP would increase with post-combustion carbon capture. The same is true for oxyfuel combustion except for GWP, AP, and EP. However, the amount of increase of these impact categories was found to be less in oxyfuel combustion compared to the post-combustion carbon capture. No LCA analysis was found for chemical looping combustion and the newly proposed Allam cycle based on oxyfuel technology.

5. CONCLUSION

Despite the adverse environmental impact of fossil fuels, they continue to dominate the power generation sector. This review provides a comprehensive discussion of various technologies aimed at reducing emissions from fossil fuel sources. Retrofitting existing power plants with post-combustion carbon capture technology, which involves separating CO₂, is considered the most suitable option. Integrated gasification combined cycle (IGCC) plants are well-suited for pre-combustion carbon capture. Post-combustion carbon capture can be implemented without extensive modifications to operational power plants. The separation of CO₂ from the gas stream in these processes requires additional energy, resulting in increased electricity prices. Different technologies are available for CO₂ separation, with pre-combustion processes being less costly due to the higher partial pressure of CO₂. The absorption process is a mature technology but requires attention to equipment corrosion and solvent regeneration costs. Large-scale implementation of adsorption processes is limited by low CO₂ adsorption capacity and gas influence on adsorbents. The development of new adsorbents is necessary to overcome these limitations. Membrane technology offers lower energy consumption but is less effective at low CO₂ concentrations. Further research is needed to

understand membrane behavior at higher capacities. Clathrate hydrate formation shows promise for CO₂ separation, but the development of suitable additives or promoters is required to enhance competitiveness. Carbon capture with water condensation is economically viable due to simpler design, higher plant efficiency, and favorable life cycle environmental aspects compared to other capture methods. Life cycle assessments demonstrate that oxyfuel combustion has a smaller environmental impact compared to other methods. However, further research is required to fully understand the unique combustion characteristics and optimize its competitiveness. Oxyfuel combustion-based near-zero emission power cycles, such as the Allam cycle, require additional research and experimental data for validation. Chemical looping combustion is a promising technology for emissions reduction, but the availability of suitable oxygen carriers and proper reactor design are critical for its success. Life cycle assessments of these new methods are also necessary to assess their environmental impact accurately. Continued research and development in these areas are essential to address the challenge of global climate change effectively.

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