

Review

# A Compact Review of Current Technologies for Carbon Capture as Well as Storing and Utilizing the Captured CO<sub>2</sub>

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**Abstract:** With the consequences of climate change becoming more urgent, there has never been a more pressing need for technologies that can help to reduce the carbon dioxide (CO<sub>2</sub>) emissions of the most polluting sectors, such as power generation, steel, cement, and the chemical industry. This review summarizes the state-of-the-art technologies for carbon capture, for instance, post-combustion, pre-combustion, oxy-fuel combustion, chemical looping, and direct air capture. Moreover, already established carbon capture technologies, such as absorption, adsorption, and membrane-based separation, and emerging technologies like calcium looping or cryogenic separation are presented. Beyond carbon capture technologies, this review also discusses how captured CO<sub>2</sub> can be securely stored (CCS) physically in deep saline aquifers or depleted gas and oil reservoirs, stored chemically via mineralization, or used in enhanced oil recovery. The concept of utilizing the captured CO<sub>2</sub> (CCU) for producing value-added products, including formic acid, methanol, urea, or methane, towards a circular carbon economy will also be shortly discussed. Real-life applications, e.g., already pilot-scale continuous methane (CH<sub>4</sub>) production from flue gas CO<sub>2</sub>, are shown. Actual deployment of the most crucial technologies for the future will be explored in real-life applications. This review aims to provide a compact view of the most crucial technologies that should be considered when choosing to capture, store, or convert CO<sub>2</sub>, informing future researchers with efforts aimed at mitigating CO<sub>2</sub> emissions and tackling the climate crisis.



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**Keywords:** CCSU; CCU; CCS; CO<sub>2</sub>; BECCS; carbon capture; post-combustion; pre-combustion; oxy-fuel combustion; chemical looping; DAC

## 1. Introduction

From 2011 to 2020, average global temperatures increased by 1.1 °C compared to pre-industrial level in 1850–1900. By 2021–2040, it is likely that the 1.5 °C level will be reached or even exceeded if our annual emissions remain the same. A rise up to even 2 °C is possible if we do not take action on a larger scale [1]. With the rise in global temperatures, we will face an imbalance in our unique ecological balance. Impacts become visible through extreme weather, rising sea levels, and ecological disruptions [2]. Total greenhouse gas (GHG) emissions, typically from carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), fluorinated gases (FG), and nitrous oxide (N<sub>2</sub>O), are the fundamental drivers of global temperature rise. The CO<sub>2</sub> emissions are, in fact, the largest of all these GHGs [3]. The Global Carbon Project estimates global CO<sub>2</sub> emissions for 2022 at 37.1 (±2) Gt. Emissions were expected to hit 37.5 Gt in 2023, a 1.1% increase compared to 2022 [3]. Reducing these emissions has become one of our greatest challenges. To tackle the rising temperatures, the European Union declared 2019 an ambitious goal to have net-zero emissions by 2050 and decoupled

economic growth from resource usage [4]. However, reducing emissions is difficult because the underlying causes are deeply embedded in complex infrastructure, which includes many sectors, such as energy, transportation, production, and agriculture. These sectors have been developed and optimized over decades. Thus, transitioning to something more sustainable is difficult, and long-term strategies can be costly. In addition, political and economic issues easily complicate GHG mitigation. The public interest and the industry's capabilities must be in harmony. Otherwise, this often causes prolonged negotiations and delayed implementation of policies and regulations. In addition, many developing nations, particularly China and India, are experiencing rapid economic development, leading to a large demand for coal as the main energy resource [5]. As more and more industrial output is produced to meet demand from such countries, a massive ramp-up in CO<sub>2</sub> emissions can be seen. For example, China produced 11.4 Gt CO<sub>2</sub> in 2022, 30.73% of all global CO<sub>2</sub> emissions. Industrial countries, such as the USA and the European Union, emitted 2022, 5.1, and 2.8 Gt CO<sub>2</sub>, respectively. Yet, concerning the CO<sub>2</sub> emissions per capita, China released 6.9 Gt CO<sub>2</sub> less into the atmosphere than the USA last year [3].

The main goal of reducing GHG emissions, particularly CO<sub>2</sub> emissions, should be finding urgent solutions to curb emissions without disrupting economic growth and industrial practices in industrial and developing countries. We should invest in reliable, cost-effective, and already-established strategies. More specifically, the focus should be on technologies that can be easily scaled [6], have high and efficient CO<sub>2</sub> fixation capabilities [7], have a low energy penalty for each ton of CO<sub>2</sub> captured [8,9], and have low investment and operating costs [10,11]. Hereby, it heavily depends on the industry sector whether a technology is considered efficient [12]. For high-temperature processes, waste heat can be applied to carbon capture processes, lowering the additional energy demand [13]. In contrast, technologies with low energy consumption are interesting for power-generating industries such as power plants [14–16].

This work provides a compact overview of possible CO<sub>2</sub> capture technologies, and the primary techniques used, like pre- and post-combustion, oxy-fuel combustion, and chemical looping for capturing flue gas CO<sub>2</sub>. The application of direct air capture to capture airborne CO<sub>2</sub> is also presented. The technologies will be discussed in terms of their advancements and limitations. In addition to carbon capture, carbon storage (CCS), utilization (CCU), and bio-based technologies (BECCSUs) will be assessed in terms of feasibility and today's application. The review will not discuss other important factors in CO<sub>2</sub> reduction, e.g., limiting deforestation processes or indirect carbon capture methods. Nevertheless, those factors play an important role and contribute to the big picture.

## 2. Current Challenges of Reducing CO<sub>2</sub> from Flue Gases

To reduce emissions, capturing CO<sub>2</sub> from flue gases, e.g., power plants or chemical production plants, plays an important role. It is even bigger if the power plant is fairly big and generates energy for many production sites or households. However, in addition to political challenges, there are a number of key challenges in CO<sub>2</sub> capture from flue gas, identified in the literature [17–21], which are summarized below. The main goal of making the process of carbon capture and storage more practical should be to address these challenges.

### 2.1. High Energy Consumption and Carbon Penalty

A big challenge in CO<sub>2</sub> capture from flue gases is the high energy demand associated with the capture process. Much of the technology employed today for CO<sub>2</sub> capture is based on solvent-based systems, which consume large amounts of energy in both the absorption and regeneration steps. The energy used in this process is generally produced by the

combustion of extra fossil fuels, which increases the overall CO<sub>2</sub> emissions of the facility and causes what can be described as a “carbon penalty”. In other words, the energy needed for CO<sub>2</sub> capture can diminish the benefit of capturing the gas if it raises the emissions. Thus, it becomes very significant to lower the energy intensity of CO<sub>2</sub> capture processes to make the technology more effective and efficient at net emission reduction.

### *2.2. High Initial Capital Costs or Operational Costs*

The deployment and operation of CO<sub>2</sub> capture systems come with significant capital investments in the beginning and associated continuous operational costs. Infrastructure developments, including gas treatment systems, capturing units, and storage facilities for CO<sub>2</sub>, require a huge financial investment. That is why the higher cost may greatly influence the feasibility of CO<sub>2</sub> capture projects and hinder large-scale applications. In addition to the initial capital costs, there are also continuing operating costs due to energy requirements, e.g., solvent regeneration, maintenance of complex systems, and monitoring of CO<sub>2</sub> emissions. These financial strains represent a strong disincentive to the widespread adoption of CO<sub>2</sub> capture technologies unless full economic incentives, such as governmental funding or carbon credit programs, are provided.

### *2.3. Low CO<sub>2</sub> Purity in Flue Gases and Inefficient Capture Rates*

The quite low CO<sub>2</sub> concentration in flue gases poses a significantly greater challenge to the efficiency of CO<sub>2</sub> capture. The flue gas basically contains all sorts of impurities, like NO<sub>x</sub>, SO<sub>x</sub>, moisture, and soot, which complicate the process of capture. These components lower the rate of capture and lower the purity of the captured CO<sub>2</sub>, thereby making it difficult to use or store it afterward. Therefore, it is of great importance to develop new technologies for the removal of these additional substances to improve the overall efficiency of the CO<sub>2</sub> capture systems. Advanced membrane or filtration technologies can be used to remove unwanted impurities from the gas stream before the actual capture of CO<sub>2</sub>. This will not only produce high-purity CO<sub>2</sub> but also increase the efficiency of the whole process.

### *2.4. Solvent Degradation and Environmental Concerns*

Another problem that exists in CO<sub>2</sub> capture is the degradation and environmental effects of solvents involved in the process. Solvents, like amines, undergo chemical changes during the process of absorbing CO<sub>2</sub> and should be replaced or renewed after certain periods of time. Partial degradation of these solvents can further cause the release of harmful byproducts, either inflicting environmental damage or requiring costly disposal measures. Many of the solvents used can contaminate soil and water if not disposed of properly. The development of less harmful and stable solvents that degrade less easily is an important step toward sustainable CO<sub>2</sub> capture.

### *2.5. Scalability and Infrastructure Requirements*

Scalability of technologies designed for CO<sub>2</sub> capture is a big challenge. They must be flexible enough to be used in small-scale industrial facilities and large-scale power generation plants. While CO<sub>2</sub> capture has been demonstrated effectively in pilot plants, the question of how to scale up these technologies to an industrial level without losing efficiency or driving up costs remains very relevant.

In addition, infrastructure for the storage or processing of large amounts of captured CO<sub>2</sub> is required. The general transportation of CO<sub>2</sub> via pipelines to its place of storage or being used in other processes requires substantial infrastructure development. Moreover, the wide-scale deployment of CO<sub>2</sub> capture technologies will need massive financial investment in infrastructure and logistical solutions to ensure CO<sub>2</sub> is stored or utilized in a safe and efficient manner.

## 2.6. Uncertain Long-Term Storage Solutions

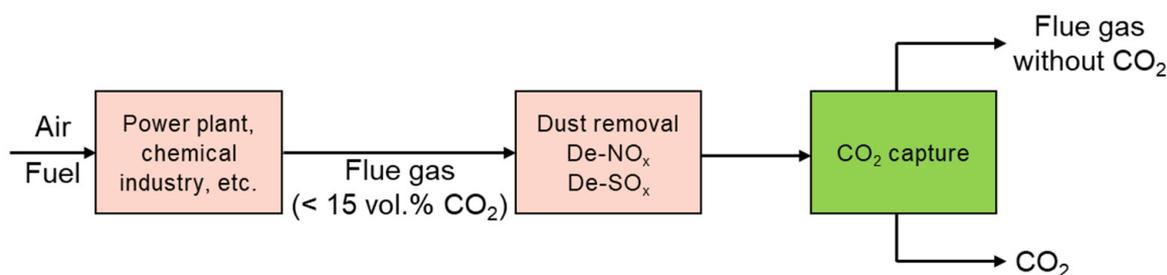
After capturing, the CO<sub>2</sub> has to be stored somewhere long-term. This is one of the uncertain aspects of CCS technologies. Geological storage in deep saline aquifers or depleted oil/gas fields seems to be promising, but many questions remain unanswered about the long-term safety of such storage and the possibility of CO<sub>2</sub> leakage over time. This poses some real risks, as CO<sub>2</sub> may escape from underground storage sites for decades or many centuries. In addition, the effectiveness and stability of CO<sub>2</sub> storage reservoirs are still under study. Establishing and maintaining public confidence in CO<sub>2</sub> storage will require much stricter regulation, continued monitoring, and transparent reporting to show that storage sites remain secure over time.

## 3. CO<sub>2</sub> from Flue Gases

Power plants burn fuel, e.g., coal, to produce primary electricity and some heat. Fuel combustion with air results in a flue gas with a low concentration of CO<sub>2</sub> because of a high proportion of nitrogen gas from the air. However, for coal, combustion can also produce other components that occur in the flue gas stream, such as nitrogen dioxide (NO<sub>2</sub>), sulfur dioxide (SO<sub>2</sub>), and soot particles. To capture the CO<sub>2</sub> from the resulting flue gas stream, a gas purification step can be required. This ensures that the subsequent carbon capture steps are neither damaged nor rendered inefficient. Typical flue gas streams of natural gas or coal-fired power plants consist mostly of about 8% and 14% CO<sub>2</sub> by volume, respectively. In the cement industry, the kiln off-gas can have a concentration of CO<sub>2</sub> of up to 33% by volume. For other applications, such as partial oxidation of natural gas or coal combustion with pure O<sub>2</sub>, CO<sub>2</sub> concentration can reach up to 40% or >80% by volume, respectively [22].

### 3.1. Post-Combustion Route (PCCC)

Post-combustion carbon capture (PCCC) of CO<sub>2</sub> is a conventional approach that is mature for many industrial applications (Figure 1).



**Figure 1.** Simplified scheme for the post-combustion route for carbon capture (based on [17]).

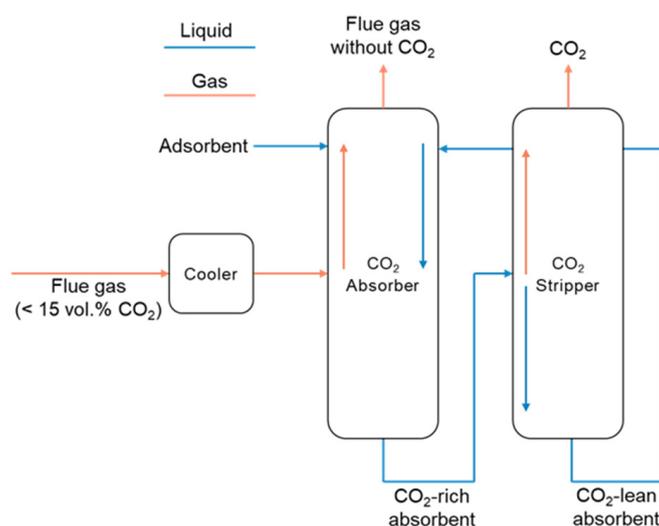
In this method, CO<sub>2</sub> is captured from the flue gases produced after combustion, as typically found in facilities like power plants. Even though this is a very promising application, the concentration of CO<sub>2</sub> in flue gases can be very low (less than 15 vol.% for coal-based power plants). Thus, the economic capture of CO<sub>2</sub> remains very challenging from a technical perspective. Such a dilute concentration also diminishes the thermodynamic driving force of the capture process [23]. One of the greatest selling points of the post-combustion route is its ability to retrofit existing power plants, making it a stepping stone towards a greener path without new infrastructure. This flexibility makes post-combustion capture a preferable option for numerous existing facilities seeking a process to lower their CO<sub>2</sub> emissions. A current operational example of a retrofitted post-combustion coal-fired power plant is the Boundary Dam Power Station, owned by SaskPower in Canada. It is the world's first integrated and continuously used post-combustion CCS facility in a coal-fired power plant [24]. The plant consists of five units with a total power capacity of 531 MW. In 2014,

unit #3, with a power capacity of 120 MW, was retrofitted to capture around 1 Mt CO<sub>2</sub> per year. The captured CO<sub>2</sub> is then used for enhanced oil recovery (EOR) or stored underground in deep saline aquifers [24,25]. Future possibilities for CCS application include retrofitted coal-fired power plants in countries with high CO<sub>2</sub> emissions, highly installed coal-fired power plant capacities, and additional storage opportunities near such facilities. In 2023, these conditions were investigated for several Asian countries because of their high portion of the global CO<sub>2</sub> emissions. The study concludes that India and China have the highest CO<sub>2</sub> removal possibility using retrofitted power plants. Of the 520 power plants screened in all countries, 305 (59%) passed the screening. These coal-fired power plants have a total capacity of 378 GW; if 90% CO<sub>2</sub> capture is implemented, 1.55 Gt CO<sub>2</sub> per year can be captured and stored in designated areas around the power plants [26].

In addition to the already established methods, there are numerous other techniques for post-combustion CO<sub>2</sub> capture. Among these are emerging approaches that show promising potential for greater cost efficiency.

### 3.1.1. Absorption via Fluid Absorbents

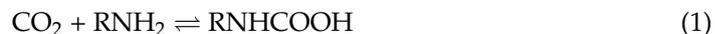
The absorption technology is a widely used and common separation method for industry applications in the petroleum, coal, natural gas power plants, and chemical industries for CO<sub>2</sub> separation [20]. The process can be divided into two main types of absorption: chemical or physical absorption. First, the flue gas stream must be cooled down. Subsequently, it is inserted into an absorber column where the CO<sub>2</sub> is absorbed into the absorbent. The lean flue gas is then released, and the CO<sub>2</sub>-rich absorbent liquid is transferred into a stripper column, where the solvent is regenerated by heat or pressure change. The released pure CO<sub>2</sub> is then captured and prepared for further storage or utilization. Conversely, the sorbent is transferred back into the absorber column for another cycle (Figure 2) [19].



**Figure 2.** Simplified flowsheet of CO<sub>2</sub> absorption via fluid absorbents (based on [19]).

The most common method of capturing CO<sub>2</sub> from flue gases is chemical absorption. More specifically, the property of some solvents to selectively absorb and react with CO<sub>2</sub> and release it upon regeneration [27]. The process is well-established in the natural gas industry and is also used to generate food- and beverage-grade CO<sub>2</sub> [28]. Due to their ability to establish a strong interaction with CO<sub>2</sub>, many aqueous amines, including monoethanolamine (MEA), methyl diethanolamine (MDEA), and diethanolamine (DEA), have emerged as the reference solvents for CO<sub>2</sub> absorption [29]. The removal of CO<sub>2</sub> from diluted flue gas streams becomes efficient because of the chemical absorption [30].

Amine-based CO<sub>2</sub> absorption utilizes the principle of chemical reactions where the reaction between the amine (e.g., MEA) and the dissolved CO<sub>2</sub> leads to the formation of carbamate compounds (Equation (1)) [31]. The detailed mechanism, including the formation of zwitterions and side reactions with water, is, for simplicity, not shown but can be seen in the literature [32,33]. If heat is applied, the carbamate formation is reversed, and the CO<sub>2</sub> and amine are released.



Although this technique is widely used, one major disadvantage is the high energy demand for solvent regeneration. Standard amine-based systems have an energy consumption rate from 2.5 to 3.5 GJ/ton of captured CO<sub>2</sub>. The implementation of heat integration techniques has shown the potential to reduce this energy demand by a further 0.5 GJ per ton [18]. Moreover, using amine-based absorbents such as MEA leads to corrosion in the columns [34]. Due to a certain amount of oxygen in the flue gas, the solvent can also decompose into corrosive products, reducing the solvent volume and harming the environment and people [35].

Apart from the previous solvents for chemical absorption, several other liquid sorbents can be used, as seen in the literature. These include amine blends [36,37], aqueous alkali metal solutions [38], nanofluids (suspended nanoparticles) [39], and chilled aqueous ammonia [40].

Furthermore, the use of ionic liquids (ILs) is considered [41–43]. ILs are ionic organic compounds with melting points below 100 °C and advantages like an extremely low vapor pressure, good electrical conductivity, as well as thermal/chemical stability. In the case of carbon capture, they are favorable due to their high capacity and fast absorption. They can absorb the CO<sub>2</sub> physically, or, in the case of functionalized ILs, a chemisorption with CO<sub>2</sub>-philic sites on the compounds is also possible and the topic of recent research [43].

Solvents for physical absorption work differently than solvents for chemical absorption. In such solvents, CO<sub>2</sub> dissolution is not controlled by chemical reactions. The efficacy of dissolving CO<sub>2</sub> depends primarily on the solubility of CO<sub>2</sub> in the solvent, which follows Henry's law and is influenced by partial pressures [44]. In industry, several solvents are used for physical absorption of acidic gases (CO<sub>2</sub> and H<sub>2</sub>S), such as chilled methanol in the Rectisol process [45–48], dimethyl ether of polyethylene (DMPEG) in the Selexol process [49,50], propylene carbonate in the Fluor process [51], and *N*-methyl-2-pyrrolidone (NMP) in the Purisol process [52]. The Rectisol process is more efficient for carbon capture than the Selexol or Purisol process due to the higher solubility of CO<sub>2</sub> in methanol at high pressures and low temperatures. The Selexol process can be efficient. However, due to the usage of NMP, it is more expensive than methanol [47]. A challenge of these solvents is the selectivity for CO<sub>2</sub>, which must be high to only absorb CO<sub>2</sub>. If there are contaminants such as H<sub>2</sub>S in the flue gas, they can be absorbed as well [46].

As a current application example, Shell has built the Shell CANSOLV CO<sub>2</sub> Capture System in 2014, integrated into the previously mentioned coal-fired power plant in Estevan, Canada. The capture system uses the DC-103 proprietary solvent [53,54].

### 3.1.2. Adsorption on Suitable Materials

In contrast to the absorption carbon capture, where a liquid dissolves CO<sub>2</sub>, adsorption is the deposition of CO<sub>2</sub> on a solid surface. The adsorption can happen in two different ways: The first is chemical adsorption (chemisorption), where the CO<sub>2</sub> creates new covalent bonds with the surface. Second, the physical adsorption (physisorption) creates no new bonds between the solid and CO<sub>2</sub> on the surface. Physisorption is based on weak Van der Waals interactions, which influence the energy demand for the regeneration process. The

regeneration energy demand for physisorption is lower than for chemisorption due to the interaction of the gas molecules and the solid surface [55,56].

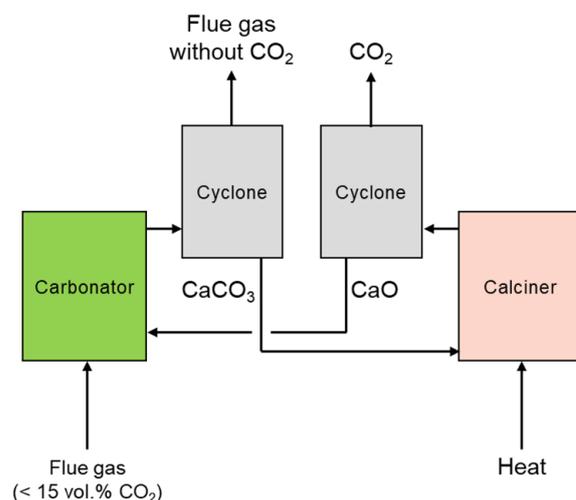
While the processes of CO<sub>2</sub> adsorbing vary only with the used material, the desorption process in terms of material regeneration can be performed in different ways. It can be achieved with heat in a temperature swing adsorption (TSA) [57], with pressure differences in a pressure swing adsorption (PSA) or vacuum swing adsorption (VSA) [58,59], with a combination of temperature and pressure in temperature vacuum-pressure swing adsorption (TVPSA) [60] or with electricity in an electric swing adsorption (ESA) [61–63].

As for solid sorbent materials, many current investigations on different materials have been carried out in the literature. Physical adsorbents can be divided into carbonaceous materials and non-carbonaceous materials. Carbonaceous sorbents are graphene [64,65], activated carbon (AC) [66,67], carbon molecular sieves (CMSs) [68,69], and carbon nanotubes (CNTs) [70,71]. Non-carbonaceous sorbents, on the other hand, are materials such as metal-organic frameworks (MOFs) [72–74], natural or synthetic zeolites [74,75], and silica-based materials [76,77]. Solid sorbents for chemisorption differ from those with physisorption because of the chemical reactions on the surface. This results in an energy-intensive regeneration to break the newly formed bonds and causes sorbent degeneration. Possible and studied materials are amine-based materials, metal salts, or hydrotalcite [78–81]. Due to the usage of different materials, regeneration processes, and CO<sub>2</sub> concentrations in the flue gas, the energy demand is estimated to range from 1.5 to 4.5 MJ/kg CO<sub>2</sub> [7].

Current examples of carbon capture with solid adsorbents are the demonstration plant in Lashburn (Canada) operated by Svante Inc., which uses its VeloxoTherm™ process based on CO<sub>2</sub> adsorption [82].

### 3.1.3. Calcium Looping (CaL)

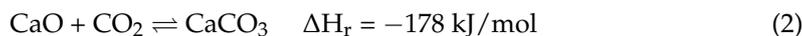
Another emerging chemisorption solid carbon capture process traps CO<sub>2</sub> via a carbonization step of calcium, called calcium looping or calcium cycling (Figure 3). Intended as a post-combustion carbon capture technology, this approach was first proposed in 1999 [83].



**Figure 3.** Simplified flowsheet of CO<sub>2</sub> capture via calcium looping (based on [84]).

The principle behind this carbon capture technique is the reversible reaction of CaO forming CaCO<sub>3</sub> with CO<sub>2</sub> in an exothermic reaction (Equation (2)). First, the flue gas is inserted in the carbonator reactor, which has temperatures ranging from 600 to 700 °C. The resulting carbonate is transferred in a cyclone to separate the solid from the cleaned flue gas. Furthermore, in the calciner, with temperatures from 900 to 950 °C, the CaCO<sub>3</sub> is then regenerated back to CaO, and the pure CO<sub>2</sub> is captured [85]. The reaction to form the

carbonate is rapid, while the reversal reaction is slower due to the CO<sub>2</sub> diffusion through the CaCO<sub>3</sub> layer formed on the free CaO surfaces [86].



An obvious challenge of this process is the energy required to reach the high temperatures for the endothermic calcination step. However, some of the heat generated from the carbonation step can be used to lower the energy requirement of the calcination. Another challenge is the loss of activity due to the fast sorbent decay after about 20 to 30 cycles, which leads to sintering processes in the calcination step [87]. The advantages of this carbon capture technique are the use of cheap material, e.g., limestone [88], and relatively low-efficiency penalty ranging from 7 to 8% [89,90]. In the case of retrofitted coal-fired power plants, it is also possible to achieve power efficiency penalties down to 2.75% [91]. Moreover, heat for the calciner can be generated by burning fuel together with pure oxygen. Due to the temperature range and the CaO/CaCO<sub>3</sub> equilibrium, the possibility of in situ carbon capture would be achievable while generating additional heat for power generation, thus increasing the plant's energy output [92].

For example, integration into high-temperature processes like cement or steel can be favorable. In 2015, a technical analysis with process modeling of a cement plant with integrated calcium looping carbon capture was performed. The plant has several benefits due to the handling of limestone and lime, raw products of the cement process. Lime (CaO) can be purged from the CaL process, replacing limestone in the kiln feed. The heat needed for the calcination in the CaL process can be obtained by burning solid fuels, like petcoke, a material which is also used in the cement plant, with pure oxygen. Furthermore, the exothermic reaction in the carbonator is used for steam generation to generate electricity via steam turbines. The latter can then be utilized for air separation to obtain pure oxygen for combustion and CO<sub>2</sub> capture, purification, and compression. Overall, the power yield of this integrated plant is calculated to be 426.66 kWh per ton produced clinker, taking the power consumption of all steps into account. Carbon capture efficiency can reach up to 94.7% [93].

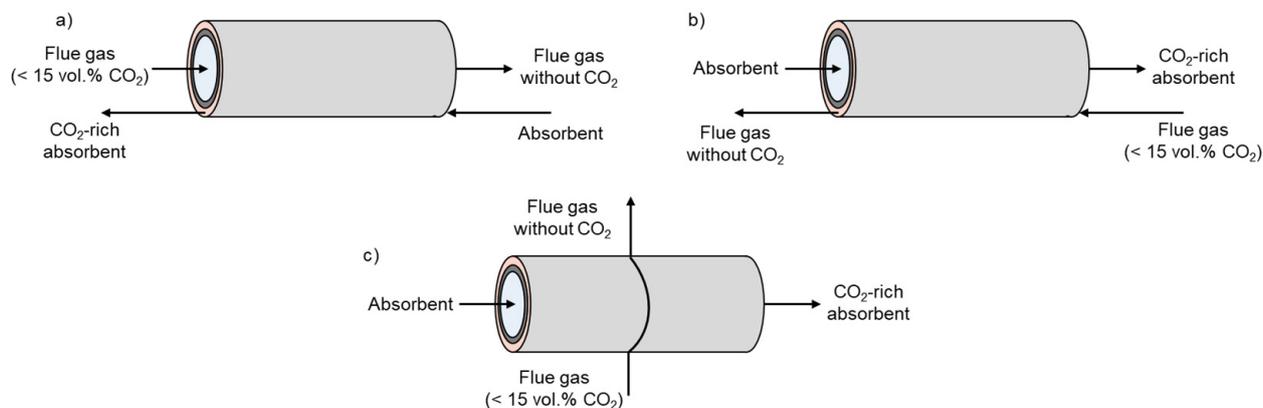
Generally, it is estimated that the cost for calcium looping is around USD 29–50/ton of CO<sub>2</sub>, which is 50% cheaper than carbon capture with amines [90].

Currently, the project CaLby2030 (started in 2022) aims to develop a commercial scale of calcium looping carbon capture in high-temperature industries such as the cement or steel industry, which account for more than 5 Gt of CO<sub>2</sub> emissions per year. The demonstration plants are planned for Germany, Sweden, and Spain. They claim to demonstrate >99% CO<sub>2</sub> capture rates [94]. For example, the La Pereda Hunosa power plant, a former 50 MW coal-fired power plant in the Asturias region in Spain, has been retrofitted to burn biomass and solid recovered fuel since 2023. Since 2012, an experimental CaL carbon capture facility called La Pereda Pilot has been operating with a small amount of flue gas (2000 N m<sup>3</sup>/h) from the power plant. The calciner is powered by a 2 MW<sub>th</sub> combustion of biomass pellets with enriched oxygen contents of around 37 vol.%. The facility achieves up to 99.5% carbon capture efficiency [95,96].

#### 3.1.4. Membrane Separation

Using membranes to separate components from a mixture is a well-established and commercialized method. Current applications include natural gas or biogas purification [97,98], water purification [99], or reverse osmosis for water desalination [100]. Using membrane technology over other established technologies, such as distillation, offers some important advantages. These are the low energy consumption due to the absence of extensive heating or cooling processes (but for the regeneration of the sorbent), the possibility of

easy linear scaling of the working system due to the modularity, and the incorporation of currently used technologies [101,102]. In the case of carbon capture from flue gases, much progress has been made over the last decades [20,28,103]. Three possible ways to operate a hollow fiber membrane, for example, in a continuous carbon capture setup, are shown in Figure 4.



**Figure 4.** Simplified flowsheet of CO<sub>2</sub> absorption via membrane separation using a hollow fiber membrane: (a) flue gas stream inside the lumen side, (b) fluid absorbent inside the lumen side, and (c) transverse flow of flue gas stream (based on [102]).

The flue gas stream can flow parallel to the liquid on the shell side, parallel to the liquid inside the lumen side, and transverse to the liquid stream on the lumen side. In the case of the liquid absorbent stream flowing through the fiber lumen, absorption performance is significantly improved by 150 to 180%. This can be explained by the better phase contact between gas and liquid. Furthermore, a parallel flow operation is 20% more effective than a possible co-current operation [104].

The selection of membrane material is highly important, and studies of different materials can be found in the literature. Examples are mixed matrix membranes (MMMs) [105–107], polymers of intrinsic microporosity membranes (PIMs) [108], metal–organic-frameworks (MOFs) [109,110], ceramics [111,112], composites [113,114], or hollow fibers of different plastic materials [115]. Based on the material and process used, these membranes can be operated with or without a sorbent liquid on the other side of the membrane to absorb the CO<sub>2</sub> as an additional driving force. Well-established sorbents such as amines, aqueous alkali metal solutions, or ionic liquids are possible. A special type of membrane is the liquid membrane (LM), which can have a support (e.g., polyethylene) combined with a liquid (e.g., ionic liquids) or exist without any support. Bulk liquid membranes (without support) consist of a non-miscible liquid that acts as the membrane phase [116]. More detailed information can be found in the literature [117–120]. Disadvantages concerning mass transfer can occur depending on the material used for the membrane. The most prominent challenges in choosing membrane materials are the mass transfer resistance and membrane wetting due to the mostly aqueous liquid used for absorption [121]. Furthermore, for membrane separation of flue gases, other significant challenges occur. Due to the low concentration of CO<sub>2</sub> of around 10–15 vol.%, the driving force for separation is decreased because membrane separation depends on pressure differences. With low partial pressures of the CO<sub>2</sub> in the gas stream, the separation is challenging, resulting in inefficient separation for flue gas carbon capture [122]. Applications with higher partial pressures are more favorable, like pre-combustion carbon capture. Another challenge is the membrane selectivity for a specific component. In most cases, choosing the right material is a trade-off between permeability and selectivity [119]. In addition, impurities, like NO<sub>x</sub> or SO<sub>x</sub>, if not removed, can damage the membrane [122]. Despite these challenges, membrane

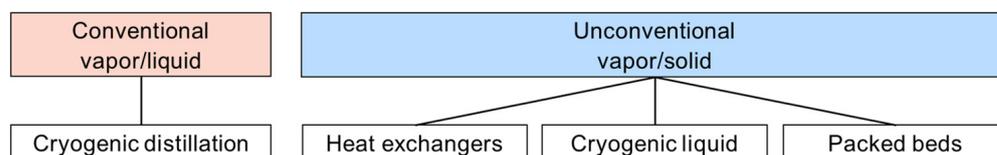
carbon capture is a low-cost method. For a CO<sub>2</sub>/N<sub>2</sub> containing diluted flue gas feed with 1% CO<sub>2</sub> in a membrane separation process, an achieved 99.8% purity of the CO<sub>2</sub> gas stream after separation was calculated. The results claim costs of USD 50.1/ton of CO<sub>2</sub> and an energy demand of 0.5 GJ per ton of CO<sub>2</sub> [123].

Several projects have utilized membrane carbon capture processes for post and pre-combustion [124]. A current example is the construction of a pilot plant for membrane separation at the Dry Fork Power Station in Wyoming (USA) by the company Membrane Technology and Research Inc. (MTR). They construct modular membrane modules using their Polaris™ polymeric membranes. The pilot plant aims to capture 55,000 tons of CO<sub>2</sub> per year. The construction was finished in October 2024. They claim to be the largest application of membrane carbon capture to date [125].

### 3.1.5. Cryogenic CO<sub>2</sub> Separation

The technology of cryogenic carbon capture is based on the simple concept of separating components based on their different physical properties, such as boiling point or, in the case of CO<sub>2</sub>, desublimation properties. Therefore, carbon capture uses flue gas streams with highly diluted CO<sub>2</sub>. Cryogenic separation can be difficult and energy-intensive due to the low concentration of CO<sub>2</sub> and the presence of other components like H<sub>2</sub>O. As the latter can clog the system at these low temperatures [20,126], the technology performs better with highly concentrated CO<sub>2</sub>. This can be achieved with different combustion setups (e.g., using the pre-combustion route) or additional carbon capture technologies like PSA to increase the CO<sub>2</sub> concentration in the gas stream [18].

Based on the used phase equilibria, this approach has two possible pathways (Figure 5). The conventional way uses the vapor/liquid equilibrium (VLE), and the unconventional way uses the vapor/solid equilibrium (VSE).



**Figure 5.** Techniques for cryogenic CO<sub>2</sub> capture in the conventional and unconventional pathways (based on [102]).

The conventional method, cryogenic distillation, is a complex and energy-intensive process due to the special properties of CO<sub>2</sub>. The column must operate at a higher pressure because of the triple point of CO<sub>2</sub> at 5.11 bar to achieve a vapor/liquid separation. Due to the low CO<sub>2</sub> concentration in the flue gas, the efficiency is low, and the energy demand is high [127]. The energy required for a conventional cryogenic distillation for a feed with 70% CO<sub>2</sub> by volume is estimated at around 1472 kJ/kg CO<sub>2</sub> [128]. For diluted CO<sub>2</sub> streams like flue gases, the energy demand is estimated to be from around 2160 to 2376 kJ/kg [35,129]. The resulting energy requirement for cryogenic distillation is therefore highly dependent on the concentration in the feed gas mixture.

In the unconventional way, there are three main technologies. The usage of heat exchangers, cryogenic liquids, or packed beds. The usage of heat exchangers was first developed in 2002 [130]. In this approach, the CO<sub>2</sub> desublimates on the fins of a heat exchanger, which is cooled by a blend of refrigerants (e.g., from methane to butane). The refrigerants evaporate to create the necessary cooling of around −120 °C. The system consists mostly of more than one heat exchanger in a cascade and can be operated with diluted flue gas streams but must be dried to avoid water ice forming on the fins. An advantage of this approach is the possibility of getting high purities of CO<sub>2</sub> after a sublimation step of 99.9%. The total electric energy consumption of this cooling method with an

initial CO<sub>2</sub> concentration of around 15% is calculated to be from around 541 to 1119 kJ/kg CO<sub>2</sub>, depending on the efficiency of the refrigerating system. It counts for all electrical systems such as fans, pumps of the cooling tower, and compressors of the refrigerating system [131,132]. It is, therefore, compatible with other carbon capture technologies and lower than the conventional way using cryogenic distillation.

Another interesting concept in the field of heat exchangers is the usage of Stirling coolers. The system consists of three coolers where the input stream is first precooled to remove any H<sub>2</sub>O in the gas stream and a second cooler for the CO<sub>2</sub> freezing step. The solids formed during the second step are transported to a third cooler to create cryogenic storage for the collected CO<sub>2</sub>. At a flow rate of 1.5 L/min and a CO<sub>2</sub> concentration of 13 vol.% in the flue gas, the energy consumption over the whole system was 1.5 MJ/kg CO<sub>2</sub>. A capturing rate of 96% of CO<sub>2</sub> was achieved [133,134]. This concept receives more and more interest in the literature due to the high efficiency of Stirling coolers, high reliability, and compact size, among other reasons [135].

Another way of cryogenic carbon capture is the usage of cryogenic liquids like isopentane. The flue gas containing CO<sub>2</sub> is passed through the liquid in a bubble column and the dissolved CO<sub>2</sub> forms solids. The slurry liquid can then be filtered to obtain the pure CO<sub>2</sub> out of the liquid. The technique aims to create a closed loop to achieve a continuous carbon capture method [136]. An energy penalty of 0.74 MJ<sub>el</sub>/kg CO<sub>2</sub> or, in other words, 15% of the plant's power output, is estimated through simulation for a coal-fired power plant with cryogenic carbon capture using cryogenic liquids [137].

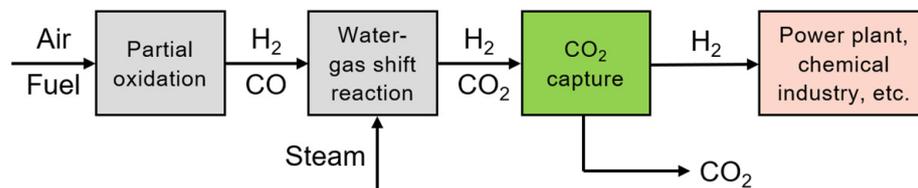
The third approach uses a packed bed with a cold material to desublimates the CO<sub>2</sub> onto the surface. The packed beds are integrated into a column, and the flue gas moves through the column until every packed bed is covered in CO<sub>2</sub> solids. Then, the column must be regenerated to obtain the pure CO<sub>2</sub> [128,138]. An alternative to this process is using just one packed bed in a cycle of CO<sub>2</sub> desublimation onto the surface and subsequently transporting the packed bed out of the column into a regeneration unit to sublimate the CO<sub>2</sub>. Afterward, the packed bed can be transported back into the column, and the capturing starts again [139]. The energy requirement for multiple beds at an initial CO<sub>2</sub> concentration of 10 vol.% is estimated to be 1.8 MJ<sub>el</sub>/kg CO<sub>2</sub> for cooling down the packed beds to −70 °C. However, the process can recover >99% of the CO<sub>2</sub> from the flue gas [140]. For only one single bed operated in a cycle, energies were calculated for carbon capture systems of three examples: an oil-fired boiler, a combined cycle gas turbine (CCGT), and for biogas upgrading. For the oil-fired boiler, the energy consumption was calculated to be 1.20 MJ/kg (332 kWh/ton), for the CCGT, 2.99 MJ/kg (831 kWh/ton), and, for the biogas upgrading, 0.95 MJ/kg (263 kWh/ton) [139].

These techniques are still under development. In March 2024, Carbon America announced a facility with a cryogenic carbon capture technology (FrostCC) that compresses and expands the flue gas with recuperative heat integration. The facility has been operating for more than 1000 h and has a capture capacity from 500 tons up to 1000 tons of CO<sub>2</sub> per year, with a capture rate of 99% [141].

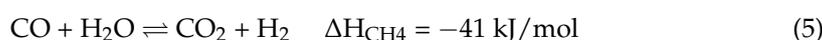
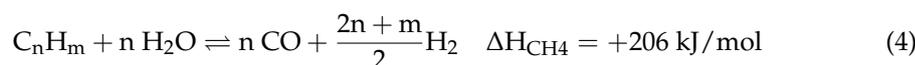
### 3.2. Pre-Combustion

In the last chapter, the post-combustion route was discussed, along with several techniques. However, instead of changing the approach for the carbon capture method from diluted CO<sub>2</sub> streams like flue gases, the combustion route can also be altered. The pre-combustion route is another important route in carbon capture technologies (Figure 6). Here, a commonly used fuel (e.g., coal) reacts with air (or pure oxygen) to produce syngas after a partial oxidation step (Equation (3)). A steam-reforming step (Equation (4)) can also be employed instead of partial oxidation, or in parallel; it is advantageous to use

exothermic partial oxidation to heat the endothermic steam reforming. In parallel, it is called auto-thermal reforming [142].

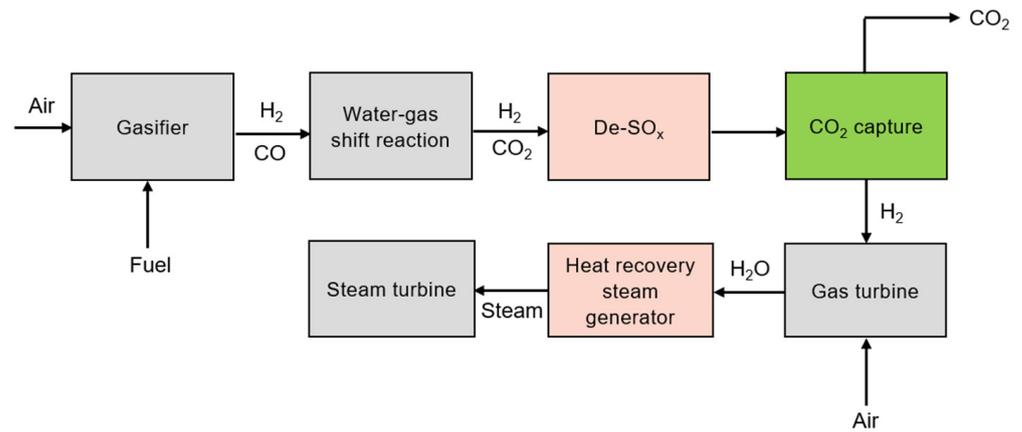


**Figure 6.** Simplified flowsheet of the pre-combustion route (based on [17]).



When used alone, the reforming step typically produces about 43% H<sub>2</sub>, 11% CO, 21% H<sub>2</sub>O, and 6% CO<sub>2</sub> [143]. Following the step of syngas production, a water–gas shift reaction (Equation (5)) is necessary to transform any of the CO into CO<sub>2</sub>. Afterward, the stream consists mainly of CO<sub>2</sub> and H<sub>2</sub>. The syngas has to be cooled down to prevent damage or loss of efficiency in the following steps. Additionally, it has to be cleaned up from impurities such as hydrochloric acid (HCl), hydrogen sulfide (H<sub>2</sub>S), carbonyl sulfide (COS), and trace metals like mercury (Hg) [144]. Due to the usage of only oxygen and not air, the partial pressure of CO<sub>2</sub> is much higher than in a diluted flue gas stream. Therefore, the CO<sub>2</sub> capture is easier and more cost-efficient. As the main reason for efficiency loss in the pre-combustion approach is the energy-intense water–gas shift reaction, this is of high importance [19]. However, despite this challenge, producing H<sub>2</sub> for power generation in, e.g., fuel cells, or the production of value-added chemicals, is an advantage [145].

To achieve a higher efficiency and counteract the efficiency loss of the water–gas shift reaction, an integrated gasification combined cycle can be added (Figure 7). The pure H<sub>2</sub> stream after the carbon capture is combusted with air in a gas turbine to generate heat and electricity. The waste heat is then recovered and converted into steam, which produces electricity in a steam turbine [146]. The principle behind a combined gas and steam turbine is called the Brayton–Rankine cycle [147]. Due to the possibility of additional carbon capture, the interest in integrated gasification combined cycle (IGCC) plants has increased in the 21st century [148]. There are also variations of the concept using other fuels and process design, but the integrated cycle stays for all approaches. For example, the biomass-based integrated gasification combined cycle (BIGCC) or the natural gas combined cycle (NGCC). For the NGCC, the natural gas is used directly in the gas turbine without the pre-combustion design.



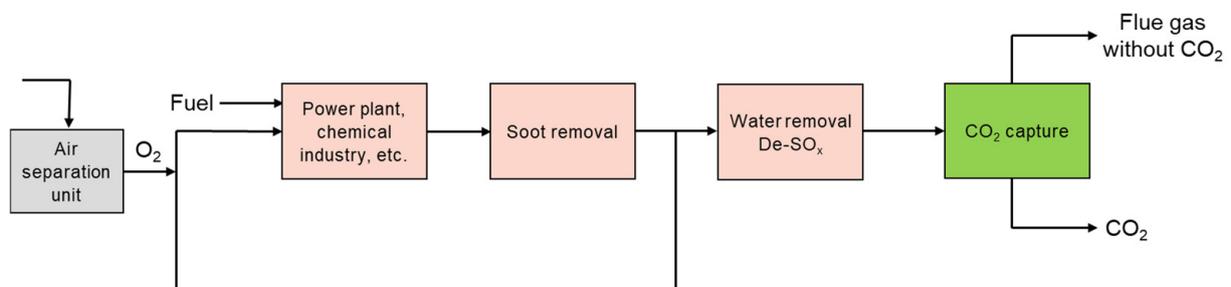
**Figure 7.** Simplified flowsheet of an integrated gasification combined cycle (based on [149]).

Currently, there are several IGCC power plants around the world operating with net electric power ranging from 127 to 2400 MW and efficiencies of up to 48% [148]. For example, the IGCC power plant unit of the Nakoso Thermal Power Station, run by Jōban Joint Power Co., Ltd. in Iwaki (Japan). It operates with an electric power of 250 MW and an efficiency of 48%. The application of carbon capture techniques is not completely clear from the source. A gas clean-up is described via MDEA and gypsum recovery (for desulfurization), but additional storage or utilization of the CO<sub>2</sub> is not further mentioned [150]. As a second example, the Jazan IGCC Complex is operated by Saudi Aramco in Jazan Economic City, Saudi Arabia. The electric power output is claimed to be 3800 MW. Additional CCSU technologies are not presented [151].

Due to the higher efficiency and the possibility of capturing CO<sub>2</sub> with a higher partial pressure, the cost of carbon capture for pre-combustion and additional IGCC technology is estimated to be around USD 77.0 (GBP 60.4) per ton of CO<sub>2</sub>. Capturing CO<sub>2</sub> via pre-combustion is cheaper in comparison to the post-combustion and oxy-fuel technology, which costs around USD 162.7 (GBP 124.7) and USD 263.2 (GBP 206.6), respectively [10].

### 3.3. Oxy-Fuel Combustion

Oxy-fuel combustion is another combustion method apart from pre-combustion, where effective carbon capture technologies can be applied (Figure 8). Pure oxygen is used for combustion, which is obtained by mostly cryogenic air separation or can be obtained during water electrolysis. After burning the fuel and soot removal, the flue gas is split and recycled back into the combustion chamber. This is because temperature control can be achieved in the combustion chamber. Furthermore, flue gas is more concentrated with CO<sub>2</sub>, which makes separation much easier [19,152].



**Figure 8.** Simplified flowsheet of the oxy-fuel combustion route (based on [19]).

Oxy-fuel combustion was first postulated in the 1980s as a method for combustion, achieving high purities of CO<sub>2</sub> in the recycling flue gas of ~95% after water removal [153].

Two main applications of possible oxy-fuel process designs exist, i.e., coal-fired power plants, pulverized-coal-fired oxy-fuel combustion (oxy-PC) [154], and the usage of circulation fluidized beds (oxy-CFBs) [155]. As for the oxy-PC, the pulverized fuel is used in certain boilers, which have been extensively reviewed in the literature [154,156–158]. The oxy-CFB method, on the other hand, employs another approach utilizing a different boiler system with limestone as an additional component. An advantage of oxy-CFB with limestone is that in situ sulfur capture can be achieved [159,160]. Oxy-fuel combustion has the advantage of reduced NO<sub>x</sub> emissions during the combustion compared to combustion with air due to the suppression of thermal NO<sub>x</sub> formation as part of the temperature control. Moreover, due to the flue gas recycling, parts of the NO<sub>x</sub> become decomposed after re-entering the combustion zone [158,161]. In the case of oxy-CFB, however, the presence of limestone can negate this advantage and increase NO<sub>x</sub> emissions depending on the temperature [155]. The cost of oxy-fuel combustion can be high due to the necessary air separation. However, an energy and economic assessment in the literature shows that when used in coal-fired power plants with heat-integration management, the CO<sub>2</sub>-avoiding cost can reach USD 43.24/ton of CO<sub>2</sub>. A challenging drawback is the reduced net power efficiency of 10.41% for the power plant, due to energy-intensive steps like the air separation [162].

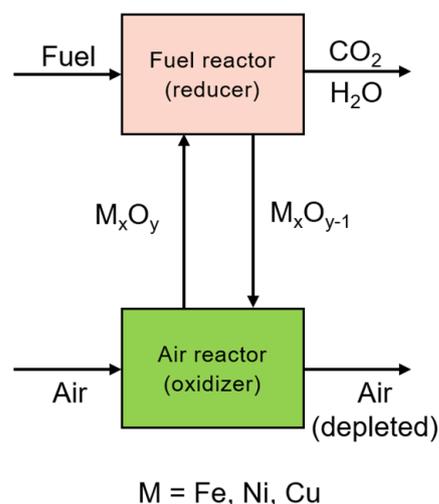
Using oxy-fuel combustion for ships is evaluated with a techno-economic analysis to reduce CO<sub>2</sub> emissions by onboard carbon capture. However, onboard O<sub>2</sub> production is one of the biggest challenges due to higher energy demand and reduced engine effectiveness. It is also evaluated that ships can obtain liquified O<sub>2</sub> from harbors, avoiding the on-board O<sub>2</sub> production, which increases efficiency but requires additional storage capacity on ships [163].

Currently, there are only a few operating oxy-fuel power plants worldwide. Many of them are no longer operating [124]. One oxy-fuel plant is the NET Power demonstration plant in La Porte, Texas (USA). The project started in 2018, operating with an output power of 50 MW. Currently, there is a planned scale-up to 300 MW, with a carbon capture capacity of 850 Mt of CO<sub>2</sub> per year, starting in 2027/28 [164,165]. As for 2024, China plans to construct an oxy-fuel combustion project based in the cement sector, which they claim to be the world's largest project of its kind. The plant aims for an annual carbon capture of 0.2 Mt CO<sub>2</sub> [166].

The Fraunhofer UMSICHT Institute takes a different approach. They combine a biogas plant, which produces mainly CH<sub>4</sub> and CO<sub>2</sub>, with an oxy-fuel combustion plant. Due to the usage of biomass, carbon capture, and oxy-fuel, the project name is BO<sub>2</sub>CCS. Furthermore, they want to utilize the O<sub>2</sub> produced during electrolysis for H<sub>2</sub> production and create a market for this unused by-product [167].

### 3.4. Chemical Looping Combustion (CLC)

An emerging technology for carbon capture is chemical looping combustion (CLC) (Figure 9). It was first patented in 1954 and intended as a production method for pure CO<sub>2</sub> [168]. Here, the air and fuel are separated from each other in the combustion process. The metals/metal oxides, which are transferred between the fuel reactor (reducer), and the air reactor (oxidizer) act as oxygen carriers. In the air reactor, the metals/metal oxides are loaded with new oxygen atoms due to the reaction with air. Then, the metal oxides react in the fuel reactor with the solid or fluid fuel and release some of the oxygen for combustion to form CO<sub>2</sub> and water, which can then be separated after condensation of water. A pure CO<sub>2</sub> stream for additional storage or utilization is achieved. The metal oxides used can be based on different metals like Fe, Ni, or Cu [169]. Due to the usage of air instead of pure oxygen, the need for energy-intensive air separation is obsolete. Furthermore, a dilution of the CO<sub>2</sub> stream with air is not possible.



**Figure 9.** Simplified flowsheet of chemical looping combustion route (based on [170,171]).

Equation (6) shows the simplified oxidation reaction in the air reactor between the metal oxides and oxygen (air). Equation (7) depicts the simplified reduction reaction in the fuel reactor with gaseous, liquid, or solid fuel. In the case of solid fuel, additional steam or CO<sub>2</sub> is necessary to fluidize the fuel [169]. In reality, the oxidation or reduction of the metal oxides is not ideal due to effects such as phase transformations or surface and bulk oxygen defects [172].

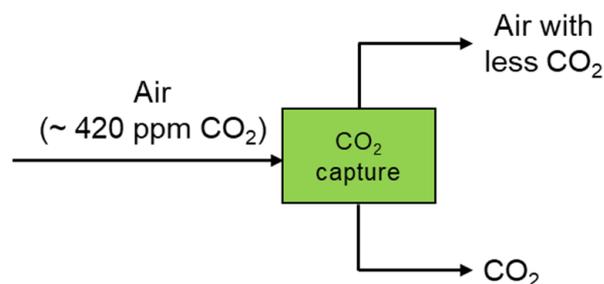


Specific and important properties are necessary for the selection of metal oxides as oxygen carriers due to the cycling process and high temperatures needed for CLC. These properties are, for example, a high melting point, high resistance to attrition and agglomeration, high reactivity with oxygen and the fuel, as well as high oxygen transport capacities [170]. The selection is of great importance, and many investigations of different metal oxides can be seen in the literature [173–177]. The carbon capture cost highly depends on the materials used and the process design, but it is estimated to be around USD 39 (EUR 37) per ton of CO<sub>2</sub> for a power plant with a net electric power of 630 MW and a capture rate of over 90% [178]. Currently, possible metal oxides are, for example, Fe<sub>2</sub>O<sub>3</sub>, Mn<sub>3</sub>O<sub>4</sub>, CuO, NiO, CoO, or FeTiO<sub>3</sub> (ilmenite) [178]. The choice of the oxygen carrier used in a CLC process also depends on environmental aspects such as the processing of the material, recycling of used oxygen carriers, handling of plant waste as well as material loss during processing and operation. Considering these challenges, CLC carbon capture processes can be optimized regarding sustainability [179].

Currently, no commercial plants use CLC for carbon capture, but there are some projects that explore further up-scaling. For example, the TU Vienna investigated and successfully operated a 100 kW pilot plant with CLC over three years. Now, they are up-scaling the process, aiming for a demonstration plant with an electric power of 10 MW [178]. Furthermore, a consortium between China and Europe (CHEERS) is constructing and testing a chemical looping combustion demonstration plant near Chengdu, China. It is currently in the testing phase after start-up. For the oxygen carrier, the plant uses ilmenite (FeTiO<sub>3</sub>). The plant will have an electric power output of 3 MW with a carbon capture rate of 96% [180]. As for 3 MW power plants using CLC, there are also two demonstration plants under construction in Ulsan (South Korea) [181] and at Ningxia University in China [182].

#### 4. Direct Air Capture (DAC) Approach to Capture Airborne CO<sub>2</sub>

In addition to capturing CO<sub>2</sub> from point sources like power plants or chemical industries, another approach is to lower the CO<sub>2</sub> content in the atmosphere is by directly capturing airborne CO<sub>2</sub> from the atmosphere (Figure 10).



**Figure 10.** Simplified flowsheet of direct air capture.

The approach to capture CO<sub>2</sub> regardless of the CO<sub>2</sub> emission source has gained more and more interest in recent years, despite the low concentration of CO<sub>2</sub> in the air of currently around 420 ppm [183]. In a projection by the International Energy Agency (IEA) for reaching the net-zero emission goal by 2025, the DAC removal target is ~980 Mt CO<sub>2</sub> per year [184]. The benefit of directly removing CO<sub>2</sub> from the atmosphere with additional carbon storage technologies is an important alternative to the general approaches of capturing CO<sub>2</sub> at fixed locations [185]. Due to the very low concentration of CO<sub>2</sub>, a large air volume must be moved through an additional carbon capture technology, which makes this technology expensive, with up to USD 1065/ton of CO<sub>2</sub>. Recent developments in DAC systems could reduce that cost to less than USD 100/ton of CO<sub>2</sub> [186]. The cost is highly determined by the capturing system and the energy cost. Depending on the method, the energy usage can range from 3.4 to 4.8 GJ per ton of CO<sub>2</sub> (thermal) and from 0.55 to 1.12 GJ per ton of CO<sub>2</sub> (electrical) [186].

Furthermore, the compatibility and profitability of DAC systems depend on government policy and funding [187]. Policies like carbon credits or a carbon tax could motivate industries to invest more in DAC systems for decentralized carbon capture. This is even more relevant for sectors that cannot capture their CO<sub>2</sub> emissions at point sources. While carbon credit prices are not well-regulated and standardized, a carbon tax is easier to implement [188]. For example, a tax higher than the amount of the carbon capturing cost could create a market for such technologies. In that case, industries can avoid paying the tax by investing in cheaper solutions like carbon capture. The DAC company Skytree suggested a carbon tax of more than USD 200/ton. Together with added value to the captured CO<sub>2</sub>, selling it to other industries can turn DAC systems profitable because the revenue is greater than the capturing cost. [188]. As a current example, the carbon tax in Sweden for 2024 is USD 131.73 (SEK 1450) [189]. While such policies like a carbon tax can reduce fossil energy consumption and lower carbon emissions, they can also have negative aspects that should be taken into account as well. For example, a tax that is too high can lead to social–economic consequences like industries relocating to countries with lower carbon tax [190,191].

While policy measures can make DAC systems profitable, technological solutions are required to reduce the high energy penalties even further thus improving the efficiency of DAC systems. Of the several available carbon capture techniques, absorption and adsorption are the most promising because they can capture CO<sub>2</sub> even from very dilute streams [192]. When adsorption processes are used, temperature swing adsorption, compared to pressure swing adsorption, is thermodynamically more effective for ultra-dilute systems, whereas pressure swing adsorption is better for bulk gas separations [193].

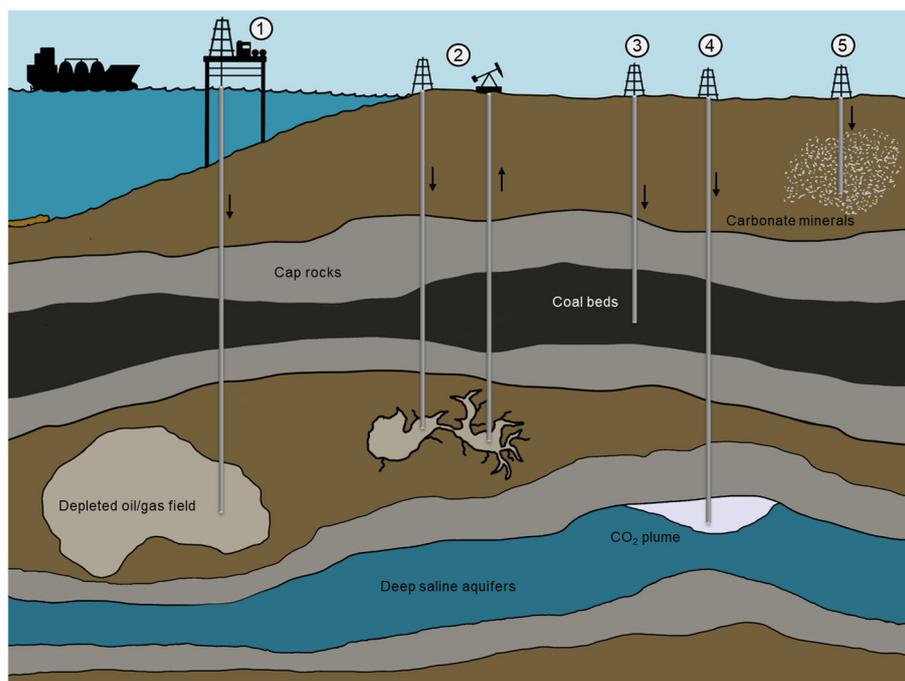
Previously discussed materials, such as MOFs or zeolites, can be used as solid sorbent material. An example is the zeolite Faujasite, which concentrates CO<sub>2</sub> from 400 ppm (0.04%) to 95% in a temperature vacuum swing adsorption (TVSA) [194]; the only downside is the high regeneration temperatures needed to desorb the CO<sub>2</sub> from the surface. Commercial utilization of the DAC system with adsorption is tested by Climeworks with their Orca facility in Iceland. The system can capture up to 4000 tons of CO<sub>2</sub> per year and uses geothermal energy produced by the Hellisheidi geothermal power plant to regenerate its sorbents. They also cooperate with Carbfix to store their captured CO<sub>2</sub> underground [195]. Since 2022, Climeworks has constructed a new DAC plant in Iceland, which will be about ten times bigger than the Orca plant and has a capturing capacity of up to 36,000 tons of CO<sub>2</sub> per year. The plant is currently in operational ramp-up [196].

As for absorption with liquid sorbents, amine-based sorbents or aqueous alkali metal solutions can be used due to their efficiency in absorbing CO<sub>2</sub> from dilute streams [185,197]. However, absorption has some drawbacks, such as the liquid amine-based sorbents degradation and a lower rate of CO<sub>2</sub> absorption compared to alkaline solutions because of present oxygen, high energy demand, and environmental hazards [198]. Hydroxide-based aqueous solutions have an even higher energy demand for regeneration with temperatures of up to 900 °C [21]. However, another step can be implemented to solve the problem of the high regeneration energy for a hydroxide-based aqueous solution. The dissolved CO<sub>2</sub> in a NaOH solution forms a carbonate, which can react with additional Ca(OH)<sub>2</sub> to form CaCO<sub>3</sub>. The solid CaCO<sub>3</sub> precipitates. After filtration and calcination, pure CO<sub>2</sub> and CaO are obtained. The CaO can be transferred back into the solution to form again Ca(OH)<sub>2</sub>. Using this cycle, a total energy penalty of 2.6 GJ/ton of CO<sub>2</sub> can be achieved [199]. The company 1PointFive, in cooperation with Carbon Engineering, uses this technology for its DAC systems and is currently constructing a DAC plant in Ector County, Texas (USA). The plant is expected to be commercially operational in mid-2025 and is designed to capture up to 0.5 Mt CO<sub>2</sub> per year. The captured CO<sub>2</sub> will then be stored in underground saline formations [199].

## 5. Storing the Captured CO<sub>2</sub> (CCS)

A crucial part of carbon capture and storage (CCS) technologies is storing CO<sub>2</sub> underground after capturing it from a point source or the atmosphere to achieve net-zero or net-negative emissions of CO<sub>2</sub> to prevent further climate change [1].

After the CO<sub>2</sub> has been captured, it is compressed and liquefied for better transport via pipelines, ships, or trucks. At temperatures and pressures greater than 31.1 °C and 7.38 MPa, the CO<sub>2</sub> is in its supercritical state, which is favorable for transportation or injection. In its supercritical state, CO<sub>2</sub> still behaves like a gas but has a much higher density than in its gaseous state. Depending on temperature and pressure, the density can range from 200 to 900 kg/m<sup>3</sup> [200]. There are different possible approaches for storing the CO<sub>2</sub> (Figure 11). It can be either stored physically in geological formations, e.g., deep saline aquifers [201], or in depleted oil and gas reservoirs [202], used for enhanced oil recovery (EOR) [203], injected into mineral formations to mineralize the CO<sub>2</sub> to carbonates, trapping it chemically (CCMC) [204], or in unconventional storage location like coal beds or gas hydrates.



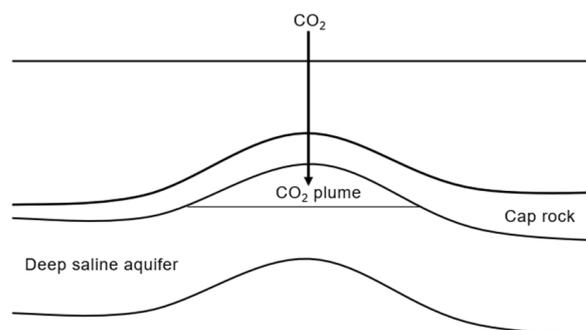
**Figure 11.** Simplified possibilities for carbon storage solutions: (1) depleted oil/gas fields, (2) enhanced oil recovery, (3) coal beds, (4) deep saline aquifers, and (5) carbonate materials (based on [205]).

Moreover, when bio-based fuels (biomass) are used for combustion processes and the resulting  $\text{CO}_2$  is then captured and combined with storage, it is called BioCCS or BECCS [19]. A big advantage of using biomass is that  $\text{CO}_2$  from the atmosphere is fixated in vegetation and is, therefore, removed from the atmosphere. The released  $\text{CO}_2$  after combustion can then be captured and stored, which is necessary to achieve real net-negative  $\text{CO}_2$  emissions (NETs) [206]. In the technical summary of the IPCC Special Report on Climate Change and Land (SRCCL), BECCS is stated as one of the most effective migration activities in the special topic of land use, which has the potential to lower  $\text{CO}_2$  emissions by 0.4 to 11.3 Gt  $\text{CO}_2$  per year between 2020 and 2050 [207].

### 5.1. Deep Saline Aquifers

Deep saline aquifers are underground fractured spaces filled with water that flows at a very low velocity, in the order of a couple of centimeters per year. They are highly saline, making them unsuitable for extraction and use for industry or human consumption. They are widely available and have a high potential for global carbon storage [200]. It is estimated that at least 1000 Gt of  $\text{CO}_2$  or more can be stored in those [208]. However, it should be considered that not all deep aquifers are suitable for long-term carbon storage, as most can have some routes to the surface, enabling leakage over long-term periods. Favorable deep aquifers are located at depths of 800 to 3000 m below the surface and are covered with a low permeable rock (cap rock), preventing diffusion to the surface [209]. Generally, for trapping  $\text{CO}_2$  inside geological formations, several mechanisms are identified in the literature, including structural and stratigraphic trapping, dissolution trapping, residual trapping, and mineral trapping [210].

Structural and stratigraphic trapping relies on the previously mentioned impermeable or low permeable cap rocks, preventing  $\text{CO}_2$  in an upward migration leakage. The  $\text{CO}_2$  is sequenced in a supercritical state into the aquifer, forming a  $\text{CO}_2$  plume. This plume can be held in place if the aquifer has a concave-down geometry at the injection site (Figure 12). The  $\text{CO}_2$  can also flow slowly to such geometry near the injection site [210,211].

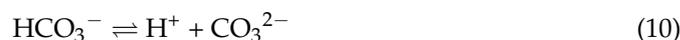


**Figure 12.** Concave-down geometry of a deep saline aquifer with cap rock sealing (based on [211]).

Possible leakage can occur due to the mobile CO<sub>2</sub> phase. If the cap rock has faults or fractures, the CO<sub>2</sub> can easily escape into shallow aquifers or abandoned wells, for instance, and, over a period of time, back to the surface [211]. A deeper understanding of the subsurface conditions, such as cap rock integrity and potential pathways for CO<sub>2</sub> migration back to the surface, is important to ensure the safety of storage sites. This applies also to other geological formations [212,213].

Furthermore, the CO<sub>2</sub> can be stored via residual or capillary trapping. The literature considers it a fast and safe method for long-term carbon storage [214–216]. After the CO<sub>2</sub> is injected and forms a plume, capillary forces of the porous rocks can snap off parts of the plume, creating isolated bubbles or ganglia and trapping it inside the pores, making the CO<sub>2</sub> immobilized and unable to escape for hundreds of years. After immobilization, the CO<sub>2</sub> will slowly be dissolved or mineralized [217,218]. The advantage of immobilized CO<sub>2</sub> due to capillary trapping is substantial for storage security [219].

Solubility trapping works by the slow process of CO<sub>2</sub> dissolution from the injected CO<sub>2</sub> plume into the saline water and is considered the most effective storage mechanism, because the forming CO<sub>2</sub>–brine mixture has a higher density and, therefore, sinks to the ground, removing the CO<sub>2</sub> from the top [218]. The CO<sub>2</sub> is first only dissolved as a gas into the brine solution. This applies to 99% of the total dissolved CO<sub>2</sub>. Subsequently, carbonic acid will be formed (Equation (8)), which will then dissociate into hydrogen carbonate (Equation (9)) and further into carbonate (Equation (10)) [220].



The density difference results in a natural convection and is only controlled by molecular diffusion [220]. It has been simulated that, for a 30-year injection of 1 Mt of CO<sub>2</sub> per year in a saline aquifer, roughly 8% of the injected CO<sub>2</sub> will be dissolved after 200 years if natural convection is the only mechanism. To optimize and accelerate this process, an additional brine injection after the CO<sub>2</sub> injection period is presented, resulting in a 50% dissolution of the CO<sub>2</sub> after the 200-year period [220].

Additionally, mineral trapping can be a consequence of solubility trapping, because the formed carbonic acid may react with minerals inside the brine solution or mineral formations surrounding the saline aquifer. A major advantage of mineral capture is that the CO<sub>2</sub> is bound chemically and precipitates from the solution, allowing for long-term storage. However, the reaction of the CO<sub>2</sub> with the minerals in the water can be fast compared to other traps or very slow, up to thousands of years, depending on the minerals present [218]. It also depends on the pH value of the brine solution, which can be modified to enhance the mineralization processes [221].

Storing CO<sub>2</sub> or, more specifically, acid gases, in deep saline aquifers was first operated in the 1990s by several gas and oil producers in the Alberta basin, Canada. Due to regulations and the uneconomic separation of acid gases (containing H<sub>2</sub>S and CO<sub>2</sub>), mixtures with gas contents ranging from 85% H<sub>2</sub>S and 15% CO<sub>2</sub> to 2% H<sub>2</sub>S and 98% CO<sub>2</sub> were injected into deep saline aquifers. By the end of 2002, nearly 1.5 Mt CO<sub>2</sub> and 1 Mt H<sub>2</sub>S were injected successfully in this region [222]. The first commercial project dedicated to only injecting CO<sub>2</sub> on a bigger scale into deep saline aquifers started in 1996. The CO<sub>2</sub> was captured at the Sleipner gas field in the North Sea. By 2004, the project had successfully stored nearly 8 Mt of CO<sub>2</sub> [222]. Currently, they claim to have stored around 20 Mt CO<sub>2</sub> [223] with a carbon removal of around 0.8 Mt CO<sub>2</sub> per year (as of 2016) [224].

### 5.2. Depleted Oil and Gas Reservoirs (DOGR)

Another way to store CO<sub>2</sub> in geological formations is in depleted oil and gas reservoirs. It is considered a safer option than deep saline aquifers because of the proven ability to store hydrocarbons, such as natural gas, over many years [202]. The IPCC Special Report on Carbon Dioxide Capture and Storage estimates the storage capacity of depleted oil and gas reservoirs in a range from 675 to 900 Gt CO<sub>2</sub> [208]. The reservoirs are mostly sealed with impermeable rocks, which provide a sealed storage location [202]. Due to the presence of (brine) water or residual oil in these reservoirs, previously discussed trapping mechanisms can also be applied, including structural trapping, capillary trapping, solubility trapping, and mineralization trapping [225]. Despite the possible leakage pathways, such as faults or fractures in the cap rock, well-integrity is a high-risk leakage pathway for this storage location [226]. Wellbores consist of steel tubes embedded in cement, while old abandoned wells are sealed with cement plugs. Corrosion can cause fractures in the cement or even the steel due to the acidic nature of CO<sub>2</sub> allowing CO<sub>2</sub> to escape into shallow formations or even into the atmosphere. In addition, old wells may have outdated wellbore standards and little information about the reservoir geology, posing a potential leakage threat. Furthermore, most of the old wells were not designed to handle CO<sub>2</sub> [226,227]. If enhanced oil/gas recovery has been used in these reservoirs, the infrastructure for handling supercritical CO<sub>2</sub> and data on potential leakage risks are available, facilitating CO<sub>2</sub> storage activities [228].

The company Wintershall Dea, a leading member of the Greensand consortium, plans to store from 0.5 to 1 Mt CO<sub>2</sub> per year by 2025 and from 4 to 8 Mt CO<sub>2</sub> per year by 2030 in the old offshore oil field Nini West (Danish North Sea) [229]. Another example is Beach Energy's Moomba CCS project, which stores CO<sub>2</sub> in depleted gas reservoirs near the Moomba Gas Plant, Australia. They achieved their first injection in 2024 and can store up to 1.7 Mt CO<sub>2</sub> per year [230].

### 5.3. Enhanced Oil Recovery (EOR)

Enhanced oil recovery is closely related to the prior discussed method to store CO<sub>2</sub> in depleted oil/gas reservoirs. The difference is that EOR is used at active wells, maximizing the yield of oil or gas from the site. The decline in new oil and gas field discoveries over the last few decades has highlighted EOR as an important strategy for the future [231]. Specifically, it is a tertiary oil recovery method used to extract residual oil from oil fields that cannot be recovered by conventional primary or secondary methods. In numbers, the leftover oil is around two-thirds of all original oil in place (OOIP) [232].

There are three possible EOR strategies, whereas only one can be used to store CO<sub>2</sub> underground. The first is thermal EOR, in which heated steam or hot water is injected into the well to lower the viscosity of the oil and enhance the oil extraction. In situ heat generation from the combustion of tiny amounts of oil within the oil reservoir can also be applied [233]. However, this technique of in situ heat generation is not easy to implement

due to the complex nature of combustion [231]. Another method is the chemical EOR, where chemicals such as polymers or surfactants are injected to enhance oil properties like viscosity or lower interfacial tension. For polymers, naturally derived polymers like xanthan gum or synthetically based polymers like polyacrylamides can be used. For surfactants, cationic, anionic, nonionic, or zwitterionic surfactants, like the anionic alkyl benzene sulfonate, can be used [234–236].

The last EOR method and practical for CO<sub>2</sub> storage is the CO<sub>2</sub>-EOR. For this, CO<sub>2</sub> with a purity from around 90 to 98% [237] is pumped in its supercritical state into the oil fields to extract the remaining oil, whereas most of the CO<sub>2</sub> stays deep underground [203]. By injecting the CO<sub>2</sub>, it becomes miscible with the oil, lowering its viscosity and improving the volumetric sweep and displacement efficacies. Consequently, oil production is enhanced [238,239].

In applications where miscible CO<sub>2</sub> flooding is not practical, e.g., due to technical requirements, CO<sub>2</sub> can also be injected at lower pressures to act as an immiscible fluid, displacing the oil in the reservoir throughout the porous rock structure. The oil is then transported to the well for extraction. Knowledge of the rock wettability and interfacial tensions between CO<sub>2</sub> and oil is crucial for this progress [240].

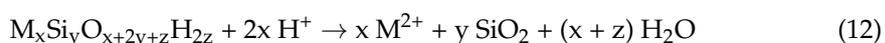
In 2010, the project at the Petrobras Santos Basin Pre-Salt Oil Field (Brazil) started, which is currently the largest CCS facility on the planet with a yearly carbon capture of over 10 Mt CO<sub>2</sub> and enhanced oil recovery for storage. As for 2022, they claim to have 40.8 Mt of CO<sub>2</sub> reinjected since the beginning of the project [241].

#### 5.4. Mineralizing the CO<sub>2</sub> Underground

Besides large geological formations, such as saline aquifers and depleted oil/gas reservoirs, CO<sub>2</sub> can be injected directly into mineral formations to carbonize the CO<sub>2</sub> to carbonates and immobilize it for long-term storage. If the operating humidity is less than 0.2 relative humidity, the process is known as the dry route. In contrast, the water route is the mineralization of CO<sub>2</sub> under wet conditions [84]. To form carbonates with acid gases like CO<sub>2</sub>, the rocks should be rich in calcium and magnesium or iron silicate [242], like in mafic or ultramafic lithologies [208]. The term mafic describes ferromagnesium minerals of all kinds, for example, basalt [243]. Basalt formations are widely available. Most of the ocean floor and more than 5% of the continents are basaltic [208]. The mineralization also highly depends on the pressure of the gas stream and time, resulting in slow kinetics and low conversions in most cases [84]. They can form carbonates in combination with CO<sub>2</sub> according to this general Equation (Equation (11)). In the water route, the CO<sub>2</sub> forms carbonic acid (Equations (8)–(10)), which improves the dissolution of the minerals and the carbonation process [84].



In contrast to the direct carbonization route of minerals, the indirect route includes two steps. In the first step, the minerals are dissolved with an acid (Equation (12)) and then converted to carbonates with CO<sub>2</sub> in alkaline conditions (Equations (13)–(14)). It has higher carbonation efficiencies and purity of the by-products compared to the direct route [84].



Other rock types such as sandstone, carbonate, and shale can also be used for CO<sub>2</sub> mineralization methods. Mineralization processes generally depend highly on temperature, permeability, rock type, porosity, and mineral precipitation. A deeper understanding of these factors for a specific storage location strengthens the effectiveness of carbon storage [244].

Commercial carbon storage via mineralization is being carried out by the company Climeworks and its DAC facility Orca. By injecting the captured CO<sub>2</sub> into the basaltic rock formation in Iceland, the CO<sub>2</sub> is transformed chemically into carbonates [245].

### 5.5. Coal Seams

An alternative approach to carbon storage could be the adsorption of CO<sub>2</sub> in unmineable coal seams. Estimations of global capacity are uncertain and range from 3 to 200 Gt of CO<sub>2</sub> [246]. Due to the coalification process, these coal seams contain methane, mostly adsorbed on the surface of the coal. By injecting CO<sub>2</sub> into these coal seams, the coal bed methane (CBM) can be replaced and extracted, which is called enhanced coalbed methane recovery (CO<sub>2</sub>-ECBM) [247]. This process is analogous to the CO<sub>2</sub>-EOR process. Owing to the higher affinity of CO<sub>2</sub> for adsorption on the coal in comparison to methane, with ratios ranging from 2:1 up to 10:1 for low-rank coal, the CO<sub>2</sub>-ECBMR method has the potential to serve as a viable carbon storage method [248]. A field test in Japan concluded that permeability is a key factor for technical feasibility, allowing economical and large-scale operations [249]. The effects of different parameters on the permeability, such as temperature, pressure, stress changes in the coal bed, and different coal ranks, are currently investigated in the literature [250–253]. Ongoing applications are rare, but successful projects still in operation include the San Juan Basin (NM, USA), Fenn Big Valley (AB, Canada), or Uinta Basin (UT, USA). The project in San Juan Basins started in 1996, and the project in the Fenn Big Valley started in 1998, having maximum injection rates of 0.1 and 0.02 Mt CO<sub>2</sub> per year, respectively. The Uinta Basin project, which started in 2005, operates at a significantly higher maximum rate, injecting 0.9 Mt of CO<sub>2</sub> per year [35].

### 5.6. Global Status of CCS Facilities

Currently, there are about 50 operational CCS projects with a CO<sub>2</sub> capturing capacity of 51 Mt CO<sub>2</sub> per year. A total of 44 are in construction, adding an additional 51 Mt CO<sub>2</sub> capture capacity. Furthermore, 534 more facilities are in development. In total, all facilities will have a carbon capture capacity of around 416 Mt CO<sub>2</sub> per year, which is an average growth rate of 32% since 2017 [254].

The issue of global capacity for CO<sub>2</sub> storage is of significant importance. However, due to the absence of sufficient data for individual storage locations and the complexity of calculating the size of different storage possibilities, it is currently challenging to determine an exact global capacity for CO<sub>2</sub> storage. Several studies on this topic are present in the literature, and it is possible to derive estimations from them. However, their validity depends on the scale in question. Regional estimations are mostly more robust and accurate than global ones. By 2007, global CO<sub>2</sub> storage was ascertained to be up to 200,000 Gt. For Europe and the USA alone, it was up to 2499 Gt and 3747 Gt, respectively [255,256]. In the Global Status of CCS 2021 report, they estimated a higher storage capacity for the USA of 8061.8 Gt [254]. It is highly dependent on the given data and calculation methods, which can differ year by year. Nevertheless, it does show the potential of CO<sub>2</sub> storage possibilities, especially because of our annual emission rate of 37.5 Gt (2023) [3]. Yet storage capacity is not infinite and is only a medium-term solution. The importance of techniques to avoid CO<sub>2</sub> emissions on a larger scale should be the main goal. During this transition phase, CO<sub>2</sub> storage seems like an acceptable solution.

## 6. Utilizing the Captured CO<sub>2</sub> (CCU)

Another possible approach after CO<sub>2</sub> capture besides storing is the utilization of CO<sub>2</sub> as a valuable product. A report by the International Energy Agency (IEA) states that, in 2015, the market for CO<sub>2</sub> consumption was around 230 Mt CO<sub>2</sub> per year. Five years later, in 2020, it was expected to be 250 Mt CO<sub>2</sub> per year, with a rising trend. The three largest CO<sub>2</sub> consumers are the USA (33%), China (22%), and Europe (16%). By sectors, the fertilizer industry, which produces urea, accounts for 57% of total CO<sub>2</sub>, the oil industry 34%, including EOR, the food and beverage sector with 3% each, metal fabrication with 2%, and others with 4% [257]. There are many CO<sub>2</sub> utilization possibilities that are presented in the literature [258–260]. For example, CO<sub>2</sub> utilization can be used to add value to products like chemicals or fuels. CO<sub>2</sub> can also act as a C<sub>1</sub> building block for the synthesis of chemicals such as formic acid [261], methanol [262,263], methane [264], polycarbonates [265,266], syngas for industry [267], and urea [268]. It is important to note that every utilization method only captures CO<sub>2</sub> in the short term, leading to a net-zero but not a net-negative emissions scenario. After a period of time, the utilized CO<sub>2</sub> will be released back into the atmosphere and thus must be captured again. Despite this fact, the recycling of already available CO<sub>2</sub> instead of creating new CO<sub>2</sub> by burning fossil fuels is an important factor for a circular economy and a sustainable future.

### 6.1. Direct Utilization of CO<sub>2</sub>

The direct utilization of CO<sub>2</sub> as gas or in a supercritical state plays an important role in industries such as food, beverages, oil, and welding applications as shielding gas [269]. For example, in food processing, supercritical CO<sub>2</sub> can be used as a blowing agent but also for essential oil or caffeine extraction, and for preservation or freeze drying [270]. Bottled water, as an example of beverages, reached 2023 a market size of USD 267.15 billion [271], with sparkling (carbonated) water accounting for USD 29.71 billion in 2020 [272]. With other carbonated beverages like beer, soft drinks, and sparkling wine not even included, the demand for CO<sub>2</sub> in the beverages industry is high and keeps growing.

### 6.2. Bulk Chemicals

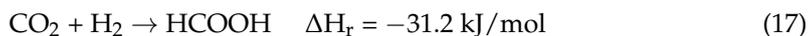
Various bulk chemicals can be produced using CO<sub>2</sub> from capturing processes, enhancing the economic value of CO<sub>2</sub> as industrial feedstock [269]. Many projects with this purpose exist, like the EU Horizon 2020 funded project RECO<sub>2</sub>DE, which aims to produce value-added additives for the cement industry, like CaCO<sub>3</sub>, different acids, or glycine from cement industry flue gas CO<sub>2</sub> [273].

#### 6.2.1. Formic Acid

Formic acid is an important and valuable bulk chemical for industry, which has many fields of application such as pharmaceuticals, textiles, or agriculture [274]. Current research considers formic acid additionally as a liquid organic hydrogen carrier (LOHC) due to its high H<sub>2</sub> capacity of 53 g H<sub>2</sub>/L [275]. Global production of formic acid via hydrolysis of methyl formate was around 770,000 tons in 2014 [274]. Formic acid is conventionally produced by reacting methanol with CO (Equation (15)) in an exothermic reaction and following endothermic hydrolysis (Equation (16)) [276,277].



Utilizing captured CO<sub>2</sub> represents a different approach to the production of formic acid by directly reacting CO<sub>2</sub> with H<sub>2</sub>, preferably sustainable H<sub>2</sub> (Equation (17)) [278].



The CO<sub>2</sub> hydrogenation is thermodynamically not favored in contrast to the hydrolysis of methyl formate. Hence, a catalyst for direct hydrogenation should be utilized. In the literature, homogenous catalysts are the most studied systems for direct CO<sub>2</sub> hydrogenation [279], first described in 1976 by a combination of VIII transition metals and a base [280]. Currently, more advanced catalysts like noble metals with pincer ligands [281] or catalysts with non-platinum metals like Fe or Co [282,283] are showing high activity in basic solutions. However, the use of basic agents to remove the acidic product produces significant amounts of salt waste [279]. Research on catalysts in acidic conditions has already been carried out to counteract this [284]. A challenge for this pathway is the lower activity for catalysts used under acidic conditions like Ru complexes with bipyridine ligands [285].

### 6.2.2. Urea

The production of sustainable urea is highly important as it is the most important nitrogenous fertilizer [286]. The industrial process for the production of urea via the Bosch-Meiser process [253] proceeds by the reaction of NH<sub>3</sub> with CO<sub>2</sub> to form ammonium carbamate (Equation (18)) and further urea (Equation (19)). The main issue is currently the fossil fuel-based production of NH<sub>3</sub> by steam reforming CH<sub>4</sub> and additional water–gas shift reactions to produce CO<sub>2</sub> and H<sub>2</sub> [287]. A key role in turning urea production sustainable is the sustainable production of H<sub>2</sub> via electrolysis for NH<sub>3</sub> production and captured CO<sub>2</sub> utilization as feedstock [288].



A techno-economic and environmental analysis of a urea production plant was performed in 2024. The plant consists of an air separation unit for N<sub>2</sub>, a water electrolysis unit for H<sub>2</sub>, a carbon capture unit for CO<sub>2</sub>, an ammonia synthesis unit for NH<sub>3</sub>, and a urea synthesis unit for final urea production. The carbon capture unit uses MEA absorption to capture 90% CO<sub>2</sub> from cement flue gas. The study reports that, for a plant with an annual urea production of 220,000 tons of urea, an energy consumption of around 8.18 million GJ per year (2272 GWh), where the electrolysis consumes roughly half of the total energy demand. The energy demand is in the same range as the conventional process. The global warming potential is significantly lower at 326 kg CO<sub>2</sub>-eq per ton of urea. This is due to the purging of some NH<sub>3</sub> during the process, not capturing CO<sub>2</sub> in the flue gas stream, and the released, unreacted CO<sub>2</sub> after urea production. However, the produced green urea would cost USD 571/ton and would therefore be 62.2% more expensive than fossil fuel-based urea (USD 351.25/ton). With carbon credit systems or lower electricity prices, the cost of urea could be lowered to USD 417.72/ton or even USD 200/ton [268].

Research is also ongoing to substitute NH<sub>3</sub> with only N<sub>2</sub>. By using frustrated Lewis pairs (FLPs) on rice-like InOOH nanocrystals, a reaction of N<sub>2</sub> and CO<sub>2</sub> can produce urea electrochemically at a yield rate of 6.85 mmol/h per gram [286]. To provide more context, using 1 kg of a catalyst, daily urea production would be 9.87 kg. Despite current low production rates, replacing NH<sub>3</sub> with N<sub>2</sub> may be a possible pathway to more sustainable urea. However, potential environmental issues by handling indium, e.g., production and waste management, in large quantities, should be taken into consideration.

### 6.3. Fuels

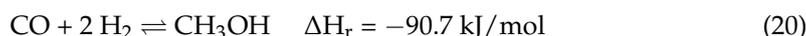
Besides bulk chemicals, the production of fuels via captured CO<sub>2</sub> is an important topic for a net-zero emission future. Next to single components such as methanol and methane used as fuels, the possibility for syngas production via reversed water–gas shift reaction (Equation (5)) can be achieved. The only drawback is the need for hydrogen as feedstock because it is also a part of syngas. The hydrogen should be derived from renewable sources to transform such syngas production sustainably. If achieved in the industry, syngas can be used to serve as a renewable feedstock for fuel production or ammonia production, among others.

As an example, the European Horizon 2020 project SUN-to-LIQUID produces Fisher-Tropsch fuels via syngas from the thermochemical conversion of captured CO<sub>2</sub> and water [289].

#### 6.3.1. Methanol

Methanol, with a current production capacity of 181 million tons in 2024 [290], is an important chemical for fuel applications due to its high energy content of 726.3 kJ/mol [291]. It can be mixed with conventional fuels without technical modifications up to 85% [292]. Besides formic acid, methanol is also considered a good liquid organic hydrogen carrier for H<sub>2</sub> transportation and serves as an important C<sub>1</sub> building block for industry [293].

Conventionally, methanol is produced by a reaction of CO and H<sub>2</sub> (Equation (20)) via Cu/ZnO catalyst, both derived from fossil-based syngas. For CO<sub>2</sub> utilization, methanol can be directly synthesized via hydrogenation of CO<sub>2</sub> (Equation (21)) [294,295].



The direct hydrogenation of CO<sub>2</sub> with H<sub>2</sub> to form methanol is thermodynamically favorable, but kinetics are a major drawback of this reaction. Industrial applications are therefore currently not economically feasible without high active catalysts. The conventional route is additionally more attractive because of the methanol yield and high equipment/hydrogen production cost. If large scale production of renewable hydrogen can be achieved, processes for direct hydrogenation, such as methanol production, can become more favorable [296,297].

Methanol is also an important starting material for several other products, such as formaldehyde, fuel additives (MTBE), acetic acid, methacrylate, and several other chemicals [292]. It can also be used to produce dimethyl ether (DME), another important fuel alternative, via a dehydration reaction (Equation (22)) [269,295].



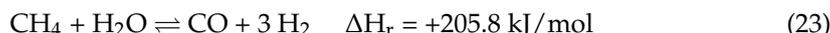
Due to all the advantages of producing methanol from captured CO<sub>2</sub> and renewable H<sub>2</sub> sources, it is considered as an important key factor for transitioning from fossil-based to renewable energy usage [294].

Current example is the company Carbon Recycling International (CRI), which has been operating the George Olah Plant in Iceland since 2012. They claim to be the world's first e-methanol plant, with a production of 214 Mt methanol per year from captured CO<sub>2</sub> [298].

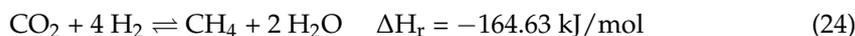
#### 6.3.2. Methane

Methane (CH<sub>4</sub>) is in high demand as a fuel, for heat generation in buildings, and as a feedstock for the chemical industry. Currently, CH<sub>4</sub> is used in steam reforming for syngas

production (Equation (23)), resulting in H<sub>2</sub> and CO for further processes. This is also the most common method for H<sub>2</sub> production [295].



To produce CH<sub>4</sub> from CO<sub>2</sub>, direct hydrogenation can be applied (Equation (24)). The so-called Sabatier reaction is named after the French chemist Paul Sabatier [269,295].



The reaction can be carried out using precious metal catalysts, like Ru or Pd, which can be costly and are limited in their reserves. A different approach is the usage of non-noble metal catalysts like Co. These catalysts have lower activity, but experiments have shown that by combining them with, e.g., porous molecular sieves like KIT-6 as a support, the activity and selectivity can be increased [299]. Moreover, Ni catalysts can be used, which show high activity and selectivity and are also much cheaper than precious metal catalysts [299].

It is also possible to produce methane biologically via hydrogenotrophic methanogens by feeding the microorganisms with captured CO<sub>2</sub> and H<sub>2</sub> [300].

A recent study investigated a compressed natural gas (CNG) pilot plant in Poland that continuously produces CH<sub>4</sub> via hydrogenation of CO<sub>2</sub> from flue gas, which is captured with amines. The H<sub>2</sub> needed for the process was produced by water electrolysis from renewable energy. The plant uses a two-stage catalytic reactor filled with a Ni/Al<sub>2</sub>O<sub>3</sub> catalyst. The plant carried out a 99.4% conversion of the CO<sub>2</sub> at a gas flow of 8.8 kg/h with temperatures in the reactors of about 300 °C and 9.3 bar. The produced CNG has a CH<sub>4</sub> content of ~94.6% and can thus be injected into the local gas grid. However, the study also describes that catalyst wetting was a problem during long-term hydrogenation, and therefore, a decrease in CH<sub>4</sub> production was observed [301].

#### 6.4. Algae Biomass

Another big part of CO<sub>2</sub> utilization is the use of algae fed with CO<sub>2</sub> from flue gases for biomass cultivation. The biomass can then be transformed into other value-added products, such as fuels [302–304]. For example, algae like *Chlorella* sp. can be cultivated in an outdoor photobioreactor (55 m<sup>2</sup>) to produce 1 kg dry biomass from 4.4 kg CO<sub>2</sub> with a daily production rate ranging from 14.6 to 28.1 g/m<sup>2</sup>. The results in this experiment were generated in summer, which can differ from experiments in winter due to seasonal or weather-related changes in sunlight. The impurities in the used flue gas (produced by natural gas combustion), such as NO<sub>x</sub> and CO with contents up to 45 mg/m<sup>3</sup> and 3 mg/m<sup>3</sup>, respectively, had no negative influences on the growth [305].

A commercial example is the company Algaecytes, which claims to have built the world's largest commercial-scale microalgae production plant located in Dessau (Germany) to harvest essential omega-3 fatty acids. The plant with an operation start planned for 2024 consists of an enclosed facility with LED lights inside to achieve 24/7 production [306].

## 7. Conclusions

This work has reviewed a selection of both established and emerging CO<sub>2</sub> capture technologies, as well as storage and utilization options. Processes like absorption using MEA are already well-established in industrial practices, and emerging technologies like calcium looping and membrane carbon capture are being tested on a bigger scale. Techniques like cryogenic carbon capture, initially considered as an uneconomic method of capturing CO<sub>2</sub> from flue gases, are currently competitive with other carbon capture technologies.

This shows that every technology has potential if it is sufficiently researched and tested in practice. Furthermore, direct air capture is no longer a concept; it has crossed over into practical experimentation and commercialization.

Several use cases for carbon utilization are reviewed for important products like methanol or formic acid, which are C<sub>1</sub> building blocks for the chemical industry. Implementing such production chains for a sustainable future regarding a cyclic economy and CO<sub>2</sub> as a new alternative carbon source instead of oil or natural gas is discussed. The potential of applied carbon storage is presented, and storage capacities for mid-term storage to achieve net-zero or net-negative emissions are shown.

In total, with all facilities operating, under construction, or in development, a global carbon removal capacity of 416 Mt CO<sub>2</sub> per year was reached in 2024. This is, compared to our annual CO<sub>2</sub> emissions of 37.5 GT CO<sub>2</sub> per year, only 1.11%. However, given the rapid growth over the past few years and the ongoing research in novel technologies, carbon capture technologies will be more implemented in current applications.

It is obvious that all the discussed technologies need continued research and development to improve their performance, reduce their cost, and enable them to be more universally applicable. Real-life applications of some of the technologies discussed in this article, like storage solutions and enhanced capture strategies, are currently being tested and up-scaled for experimental and pilot scenarios or even demonstration plants.

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