

Review

# Review of Carbon Capture and Methane Production from Carbon Dioxide

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**Abstract:** In the last few decades, excessive greenhouse gas emissions into the atmosphere have led to significant climate change. Many approaches to reducing carbon dioxide (CO<sub>2</sub>) emissions into the atmosphere have been developed, with carbon capture and sequestration (CCS) techniques being identified as promising. Flue gas emissions that produce CO<sub>2</sub> are currently being captured, sequestered, and used on a global scale. These techniques offer a viable way to encourage sustainability for the benefit of future generations. Finding ways to utilize flue gas emissions has received less attention from researchers in the past than CO<sub>2</sub> capture and storage. Several problems also need to be resolved in the field of carbon capture and sequestration (CCS) technology, including those relating to cost, storage capacity, and reservoir durability. Also covered in this research is the current carbon capture and sequestration technology. This study proposes a sustainable approach combining CCS and methane production with CO<sub>2</sub> as a feedstock, making CCS technology more practicable. By generating renewable energy, this approach provides several benefits, including the reduction of CO<sub>2</sub> emissions and increased energy security. The conversion of CO<sub>2</sub> into methane is a recommended practice because of the many benefits of methane, which make it potentially useful for reducing pollution and promoting sustainability.



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## 1. Introduction

Energy plants, industries, as well as other sources of carbon dioxide (CO<sub>2</sub>) result in global warming and affect the planet [1]. The temperature of the Earth's surface has risen by 0.8 °C in tandem with an increase in CO<sub>2</sub> concentration of 280 to 400 ppm [2]. In the last century, the CO<sub>2</sub> level has increased slightly to 408.8 ppm [3], with projections of 600 to 700 ppm, raising average surface temperatures by 4.5–5 °C [4]. Unstable financial, technological, and sociological advances, along with natural and human developments, are all important factors affecting the CO<sub>2</sub> concentration [5]. In addition to CO<sub>2</sub>, other greenhouse gas (GHG) emissions have significantly increased in recent years, such as sulfur hexafluoride (SF<sub>6</sub>), perfluorocarbons, hydrofluorocarbons (HFC), nitrous oxide (N<sub>2</sub>O), and methane (CH<sub>4</sub>) [1,6]. According to the International Panel on Climate Change, GHG emissions should be lessened by 50–80% to prevent our planet from disintegrating [2]. Almost 190 countries met in Paris in December 2015 (Conference of Parties—COP 21) to agree on limiting global temperatures to 2 °C by the end of the 21st century. COP 21 proposed several techniques, including increasing energy efficiency, utilizing renewable fuels, and implementing geoengineering techniques such as afforestation and, more specifically, developing carbon capture and storage (CCS) technologies [3,4]. Therefore, carbon capture and storage (CCS) is generally acknowledged as a potential, if not suitable, method to minimize CO<sub>2</sub> emissions globally [7,8].

An estimated 60% of CO<sub>2</sub> emissions are attributed to many stationary sources, including industrial plants, fossil fuel power plants and thermoelectric cement plants, iron and

steel mills, gas processing industries, refineries, and power plants. These industries serve a variety of sectors such as the industrial sector, transportation, and electricity generation [9]. According to the above analysis, stationary emission sources will continue to be the greatest drivers of greenhouse gases (GHGs) resulting from the burning of fossil fuels for several decades to come. The production of electricity is responsible for 25% of all emissions, while forestry and agriculture account for 24% [10]. Thermal power plants typically use coal as fuel, because it is cheap and readily available, yet it is considered a major emission source, with emissions approaching 2249 lbs/MWh [11]. As a result, new and improved technologies such as CCS for swift CO<sub>2</sub> removal from the atmosphere to tackle global warming have received a lot of attention lately. Figure 1 illustrates the various energy sectors contributing the most to CO<sub>2</sub> emissions.

The type of combustion process utilized, as well as the volume of CO<sub>2</sub>, absorbed, separated, and transported for reuse or storage, are the primary determinants of CCS [4]. CCS can reduce CO<sub>2</sub> emissions from power plants to limit global warming to 2 °C [4]. Post-combustion is the most advanced of the three major CCS technologies, followed by pre-combustion and oxy-fuel combustion. In terms of CO<sub>2</sub> capture, coal gasification is the best option; however, it cannot be used in gas-fired power plants. It makes use of most of the existing power plant infrastructure as a retrofit technology. There has been a demonstration of the ability to recover 300 t/day of CO<sub>2</sub> on a small scale [4].

During oxy-fuel combustion, pure oxygen replaces air, thereby eliminating the requirement for flue gas de-NO<sub>x</sub> and the incorporation of 80–98 °C CO<sub>2</sub> in the exhaust, resulting in a more effective and reliable CO<sub>2</sub> removal method. Although the process requires a lot of energy, corrosion is aided by SO<sub>2</sub> concentration due to the increased energy consumption [12]. Gasification under low O<sub>2</sub> pressure in pre-combustion CCS produces syngas (CO + H<sub>2</sub>) from the fuel pretreatment [4]. In the presence of high CO<sub>2</sub> concentrations in fuel gas, separation is less difficult than with other methods, resulting in the formation of H<sub>2</sub> as a transport medium for energy. This helps to offset some of the technology's operational costs [12].

The purpose of this paper is to provide an overview of the state of carbon capture systems, both conventional and the emerging, and present CO<sub>2</sub> as a feedstock for producing methane (CH<sub>4</sub>). It further concentrates on improving the concept and fostering research on CO<sub>2</sub> capture and utilization to circumvent the storage issues with the current CCS technology, and make sustainability a priority for future generations.

There are currently no studies that address all the gaps; however, this paper covers CO<sub>2</sub> capture, sequestration, utilization, and CO<sub>2</sub> methanation.

## 2. Methodology

The focus of this research on the literature was intended to identify the benefits and drawbacks of current carbon capture systems and porous carbon-based adsorbents for CO<sub>2</sub> adsorption.

In addition, the challenges and prospects for CO<sub>2</sub> conversion into methane were discussed. The manuscript's section on methodology describes how the literature was found, gathered, and then arranged by identifying the knowledge gaps that exist in earlier studies. Using scientific databases and a variety of search engines, such as NCBI, Google Scholar, Scopus, Science Direct, Web of Science, and major publishers, an in-depth analysis of the articles and literature from peer-reviewed journals was carried out. The search for relevant literature was constrained to studies from the years 2000 to 2022 to draft this manuscript. The limited literature search for this manuscript's preparation was mostly focused on studies between the years 2000 and 2022. For this paper's main theme, several pertinent keywords were employed in the literature search. These keywords include zeolite, activated carbon, graphene, silica, and metal-organic framework, as well as CO<sub>2</sub> capture and conversion. Research articles with the most pertinent material were selected after the articles were evaluated and the information from the abstracts was reviewed. To prevent translation challenges, articles published in languages other than English were omitted.

### 3. CO<sub>2</sub> Capture Technologies

CCS technology relies on CO<sub>2</sub> capture. Three methods for trapping and sequestering CO<sub>2</sub> are envisaged, based on the settings of a fossil fuel-based power plant, gas steam pressure, and CO<sub>2</sub> partial pressure. Among the carbon capture technologies are pre-combustion, oxy-fuel combustion, and post-combustion [13].

#### 3.1. Pre-Combustion Capture Method

This method involves capturing CO<sub>2</sub> before combustion, rather than after combustion, thus making this method more feasible. Gasifiers and catalytic reactors are involved. In a gasifier with low oxygen pressure, the following equations (Equations (1)–(3)) demonstrate how pure syngas (CO, H<sub>2</sub>) is produced. Moreso syngas is enriched by passing it through a shift reactor with a steam, as shown in Figure 1.



Natural gas could also be applied for steam reforming via this method; between 700 and 850 °C, an endothermic reaction occurs, culminating in the conversion of CH<sub>4</sub> and H<sub>2</sub>O and the generation of syngas (CO, H<sub>2</sub>). An exothermic reaction occurs after partial oxidation, in which CH<sub>4</sub> burns in oxygen to create CO<sub>2</sub> (Equations (4) and (5)) [14].



A key element of pre-combustion is the hydration of gas separated from air, which is the most promising technology today [15].

The CO<sub>2</sub> captured in this technique produces hydrogen fuel, which is used to generate electricity in several industries while emitting little CO<sub>2</sub> [16]. Fertilizers and hydrogen are produced through pre-combustion capture [17]. CO<sub>2</sub> is generated from this process in concentrations ranging from 5 to 60%, making it easy to capture; however, the water gas shift reactions and gasification are costly and difficult to operate [18].

#### 3.2. Oxy Fuel Combustion Capture Method

A system for air separation during oxy-fuel combustion capture is used to obtain pure oxygen, as flue gas and coal are fed into the oxy-fuel boiler [19–21]. Coal is burned at a fixed temperature and pressure. Figure 1 illustrates how the flue gases, lacking oxygen and nitrogen, are only made up of CO<sub>2</sub> and water vapors. Additionally, this reduces the quantity of flue gas required to remove NO<sub>x</sub> and sulfur [22,23]. Condensing water vapor maintains the boiler's temperature. Dehydrated CO<sub>2</sub> is compressed and delivered to a storage facility or an industry where it is utilized in beverage carbonation, as a solvent, and as a fire extinguisher [24]. Flue gas is recycled in its natural state to maintain the boiler's internal temperature [20]. When compared to other carbon capture technologies, it is the most efficient and adaptable [25]. There are, however, some obstacles to overcome. The air separation unit (ASU) uses cryogenic distillation, which consumes a great deal of energy to produce essentially pure oxygen. The integrated gasification combined cycle (IGCC) can be replaced by this method, which converts CO to CO<sub>2</sub>.

A major drawback of this method is that it demands more energy to separate oxygen from the air [4,26].

#### 3.3. Post Combustion Capture Method

After burning carbonaceous materials (such as biomass) or fossil fuels, CO<sub>2</sub> is removed from the flue gas using a viable adsorbent such as amine [4,27]. Currently, it is the most effective method of capturing CO<sub>2</sub> from natural gas plants. As separating agents, it

employs amine solutions including potassium carbonate ( $K_2CO_3$ ) mono-ethanolamine (MEA), methyl-di-ethanolamine (MDEA), and di-ethanolamine (DEA) [14]. The novel sorbent piperazine collects contaminants efficiently because of its high volatility [28]. A chemical reaction deposits  $CO_2$  in the solution, which is then separated by flowing high-temperature (100–200 °C) steam over it.

An equation representing the reversible absorption–regeneration reaction is shown below [14].



However, this method has some drawbacks, such as a high parasitic load, high flue gas temperatures, and the inability to operate at low  $CO_2$  levels. Figure 1 illustrates a direct post-combustion mechanism. Gas-fired power plants use about 4%  $CO_2$ , while coal-fired power plants utilize 7–14%, increasing electricity costs by 32% and 65%, respectively, in gas-fired and coal power plants [4]. As a result, cutting costs while improving system performance is still a subject of great interest [29]. Membrane technology, including ceramic or polymeric membranes, marine algae, and cryogenic distillation, can all be used for post-combustion  $CO_2$  capture. This has many benefits, including the ability to retrofit and eliminate the need for a cryogenic separator and shift reactor. In post-combustion,  $CO_2$  is usually absorbed or desorbed using a solution containing alkanol amine [30–32].

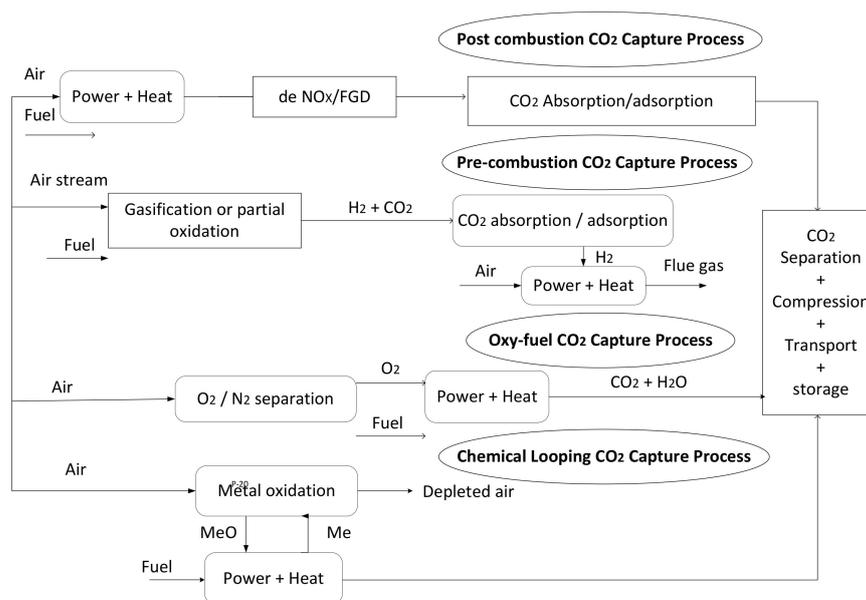


Figure 1.  $CO_2$  capture technologies [33].

#### 4. Various Combustion Technologies for $CO_2$ Capture

A comparison of the three  $CO_2$  capture technologies is illustrated in Table 1. Coal gasification uses pre-combustion, while oxy-fuel combustion and post-combustion are applied to coal- and gas-fired plants.  $CO_2$  capture using post-combustion technology is presently the most advanced [28,34]. According to Gibbins and Chalmers [35], three technologies were compared in terms of the costs of both gas-fired and coal-fired power plants (Table 2).  $CO_2$  mitigation for coal-fired power plants was the cheapest using pre-combustion technology, while oxy-fuel and post-combustion technologies were comparable. For post-combustion capture, the costs per tonne of avoided  $CO_2$  were 50% lower for gas-fired plants than for the two other technologies. Moreover, the most inefficient method of  $CO_2$  capture is post-combustion  $CO_2$  capture, having an energy cost of roughly 8% for coal-fired plants and 6% for gas-fired plants [36].

**Table 1.** Comparison of various CO<sub>2</sub> capture technologies [37].

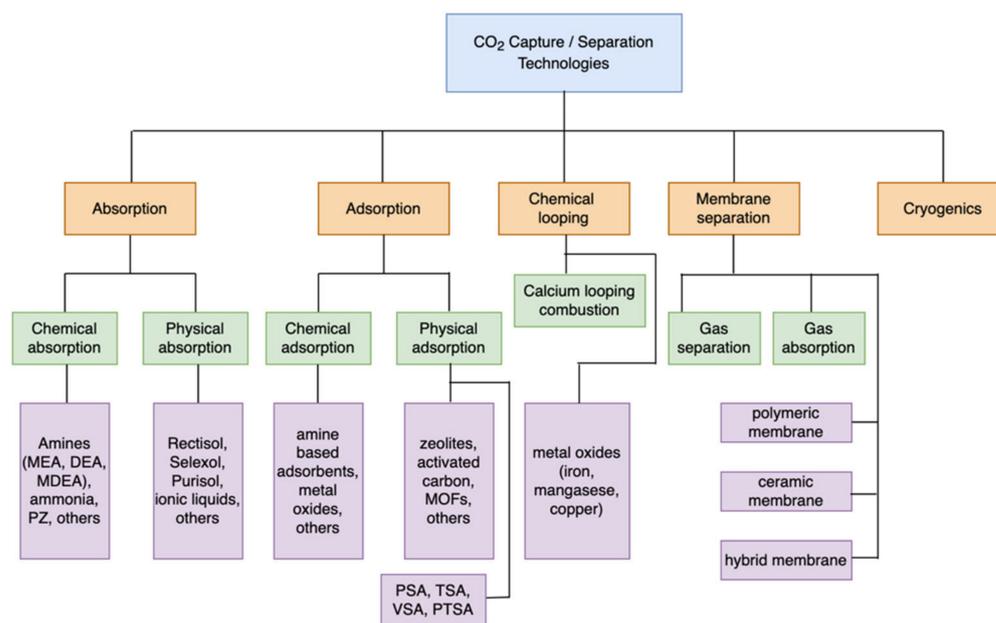
Parameter	Post-Combustion	Pre-Combustion	Oxy-Fuel Combustion
Application area	Can be utilized with current coal combustion plants.	Integrated gasification combined cycle and turbines, which can effectively use H <sub>2</sub> -rich syngas.	Synergy between novel cycle and integrated gasification combined cycle has been employed.
Pros	Small concentrations of CO <sub>2</sub> can be captured. Possible to retrofit to existing plants.	Lower energy penalty than post combustion methods. The pressure and temperature of the regeneration process can be altered.	The CO <sub>2</sub> capture efficiency is 100%. Absence of hazardous NOx.
Cons	High costs for operation and regeneration. Excessive solvent losses.	Syngas treatment and drying before CO <sub>2</sub> capture. Capital costs and investment costs are high.	High operational and capital costs. Annexing to existing plants is difficult.
CO <sub>2</sub> concentration (vol.%)	4–14	15–40	75–80
CO <sub>2</sub> capture efficiency (%)	85–90	85–90	90–100
CO <sub>2</sub> purity (%)	99.6–99.8	95–99	87.0–94.8
Temperature and pressure	Flue gas must be cooled, and pressure is dependent on the CO <sub>2</sub> capture process.	High pressure and low temperature (depending on the technique utilized).	Cryogenic temperatures are used to separate oxygen. The flue gas is recycled to lower the temperature because oxy-fuel combustion produces high temperatures.
Equipment size	Large size equipment required with high investment.	Medium equipment.	The equipment is of low size.
Combustion medium	Air is used.	Steam/air is required for gasification to generate CO <sub>2</sub> .	High-purity oxygen for combustion.
Start of the art	Amine scrubbing plants (reaction with monoethanolamine) are in practice. Currently, power plants use this technology.	Integrated gasification combined cycle and ammonia production plants are running currently.	Efficient CO <sub>2</sub> separation.
Acid gases	Contains H <sub>2</sub> S, COS, NOx, and SOx.	Sulfur compounds need to be removed.	NOx is not present; however, gas desulfurization is necessary.

**Table 2.** Comparison of the costs of various capture methods [35,37]. Costs exclude storage and shipping costs, but include CO<sub>2</sub> compression to 110 bars.

Fuel Type	Parameter	Capture Technology			
		Pre-Combustion	Post-Combustion	Oxy-Fuel Combustion	No Capture
Gas-fired	Capital cost (USD/kW)	1180	870	1530	500
	Cost of CO <sub>2</sub> avoided (USD/t CO <sub>2</sub> )	112	58	102	-
	Cost of electricity (c/kWh)	9.7	8.0	10.0	6.2
	Thermal efficiency (% LHV)	41.5	47.4	44.7	55.6
Coal-fired	Capital cost (USD/kW)	1820	1980	2210	1410
	Cost of CO <sub>2</sub> avoided (USD/t CO <sub>2</sub> )	23	34	36	-
	Cost of electricity (c/kWh)	6.9	7.5	7.8	5.4
	Thermal efficiency (% LHV)	31.5	34.8	35.4	44.0

## 5. CO<sub>2</sub> Separation Techniques

CO<sub>2</sub> is separated from flue gas during combustion using a variety of advanced separation techniques [4]. There are several techniques involved, such as absorption, adsorption, chemical looping, membrane separation, and cryogenics [18,38,39]. Figure 2 displays a flowchart outlining the methods and techniques for CO<sub>2</sub> capture/separation technologies.



**Figure 2.** Separation techniques for CO<sub>2</sub> capture [40].

### 5.1. Absorption

The ability of absorption to capture huge quantities of emissions from chemical factories and power plants has gained considerable attention in recent years. Chemical absorption is a reliable technique for CO<sub>2</sub> separation in coal-fired power plants because it is well-suited for existing plants with high operating costs and limited infrastructure [41]. The chemical absorption of CO<sub>2</sub> is a commercially viable technology due to its many advantages, including technical efficiency, handling capacity, and sophistication [42]. The potential absorbents and processes of absorption CO<sub>2</sub> capture are highlighted in Table 3.

CO<sub>2</sub> is separated from flue gas by absorption using a liquid sorbent [3,43]. It is possible to regenerate the sorbent via a regenerative process or stripping by depressurizing and/or heating. This is the latest and most advanced method for separating CO<sub>2</sub> [44]. Potassium carbonate (K<sub>2</sub>CO<sub>3</sub>), monoethanolamine (MEA), and diethanolamine (DEA) are examples of common sorbents [45]. MEA is very reactive and absorbs more quickly, and it is quite inexpensive [44]. However, their main drawback is the substantial parasitic energy load in relation to solvent regeneration, which adversely affects the total effectiveness of systems combined with aqueous amine-based absorption processes [46]. DEA and other alkanolamines have also been employed for absorption, although they have comparable defects. Methyldiethanolamine (MDEA), a mixture of MEA and DEA, has been used with moderate success. It has higher CO<sub>2</sub> loading capacity, and degradation and corrosion resistance, as well as cheaper regeneration costs, but lower rates of absorption [47–52].

Veawab et al. [53] reported that MEA is the most efficient aqueous alkanolamine for CO<sub>2</sub> absorption, with a performance rate greater than 90%. Additionally, Aaron and Tsouris [54] reviewed various CO<sub>2</sub> capture technologies and determined that MEA absorption is the most viable method for CO<sub>2</sub> capture in CCS. Applying a solvent containing 30% MEA, a 1 t CO<sub>2</sub>/h absorption pilot plant was designed and experimentally validated in conjunction with a coal-fired power plant's post-combustion capture technology [55]. In recent times, other adsorbents, including anion-functionalized ionic liquid and piperazine, have attracted a lot of attention [56]. Even though piperazine rapidly reacts compared to MEA, its use in CO<sub>2</sub> absorption is more costly. Due to its higher volatility, it is still in the experimental phase [28]. The risk of amine degradation, which could lead to equipment corrosion, solvent loss, and the formation of volatile degradation compounds, is a significant barrier to the widespread adoption of this technology for the CCS [57,58], while environmental degradation has gone unnoticed.

Furthermore, amine emissions can deteriorate into nitramines and nitrosamines, which are highly toxic to human health and the environment. The chilled ammonia process captures CO<sub>2</sub> using aqueous ammonium salts (including ammonium carbonate) and can regenerate the CO<sub>2</sub> at elevated temperatures and pressures using waste heat, thereby minimizing the downstream compression [59]. There are fewer problems with this process than those caused by amine degradation.

Water's use as a co-solvent, which has higher thermal characteristics than other co-solvents, is one of the key precursors for the high solvent regeneration energy of MEA [20]. In the context of CO<sub>2</sub> absorption, the predicted regeneration energy for 30 wt. % aqueous MEA showed that more than 50% of the total energy was used to heat and vaporize the water co-solvent. The remaining energy was used to reverse the chemical interaction between CO<sub>2</sub> and MEA at the same time [60,61]. Considering this, it was thought that either totally or partially substituting other organic diluents for water as co-solvents could potentially reduce solvent regeneration energy, since they effectively create water-free/water-lean hybrid solvents with poorer thermal properties than water [62–68]. Instead of vaporizing and heating the co-solvent, comparable to aqueous amines, the regeneration energy will be used more effectively to reverse acid gas chemisorption.

Additionally, hybrid water-free/water-lean solvents have been thoroughly studied in recent years, primarily for their CO<sub>2</sub> capture applications [68–70]. They provide a wide range of potentially alluring substitutes to conventional aqueous amines [46,71]. The main objective of water-lean solvents is to preserve the chemical selectivity of water-based solvents, while enabling step gains in efficiency due to the lower specific heats of organics than water [46]. However, two problematic regions refute the claim of their attractiveness. The stated performance of these solvents when scaled up from lab-scale to industrial-scale settings has not been adequately examined due to a lack of availability of a few essential properties. This is predicted given the labor-intensive nature of experimental work, which makes it impossible to expand experimental testing to the broad range of transport and thermophysical parameters needed for precise and representative performance evaluation on an industrial scale. The second issue is that, when carried out on a lab scale, the potentiality of a particular solvent is typically demonstrated using a limited set of parameters, most notably the low enthalpy of absorption and high absorption capacity [72–75]. These two characteristics are indeed of great concern for chemical absorption procedures, but they are still unsuitable for accurately gauging the potential of the tested solvents for their intended use. However, they ignore significant trade-offs between competing environmental, economic, and operational factors. The results of a straightforward assessment can help direct the development of novel generating solvents [76,77].

However, these difficulties can be overcome if the proper tools or novel process configurations are available. Due to recent developments in computational power and thermodynamic modeling tools, the first issue can be resolved by scaling the data from lab to industrial operating conditions. The most appealing models for this application are molecular equations of state (EoSs) centered on the Statistical Associating Fluid Theory (SAFT) [78,79], due to their strong theoretical background, demonstrated correctness for a range of complex systems, and predictive abilities.

The solution to the second problem, which is to demonstrate the viability of a chosen solvent typically acknowledged using a limited number of requirements, may appear relatively apparent: add more evaluation criteria to the already-existing standard key performance indicators (KPIs). Moreover, making such a preference is more difficult because early design phases may not have access to information on a particular criterion [80]. There must be a justification for why certain criteria should be included or excluded when narrowing the search space among the numerous properties that are available [81–83]. The effect of solvent characteristics on economic metrics such as total capital expenditures (CAPEX) and operating expenditures (OPEX) typically serves as the foundation for justification. Through the careful process modeling of hypothetical solvents, Mota-Martinez et al. [84] ranked various solvent characteristics according to how they affected the process' overall

economics. Leclaire and Heldebrant [43] recently recommended the use of ideas from green engineering and chemistry to address problems with the advancement of CCUS technologies. They asserted that by applying the 12 + 12 principles of engineering and green chemistry [85], they could indirectly encourage the improvement of chemical processes' economic attractiveness and efficiency, which goes beyond their environmental motivation. Similarly to this, it may be beneficial to consider sustainability, health, and safety issues while assessing the possibility of promising solvents for the removal of acid gas [86,87]. Figure 3 displays the schematic diagram for the absorption carbon capture process.

Table 3. Summary of absorption-based carbon capture.

Type	Absorbent	Reactive Separator	Operating Conditions P, C, T, G	CO <sub>2</sub> Capture (%), AC (kg/kg)	Kinetics/Mass Transfer	Ref.
Single solvent	MEA	Flow (SC)	C:8 –16; T:10–40; G:2–10	94, 0.4	$C_2H_4OHNH_2(1) + CO_2(g) + H_2O \leftrightarrow C_2H_4OHNH_3^+(aq) + HCO_3^-(aq)$	[88–90]
	K <sub>2</sub> CO <sub>3</sub>	Fixed-bed (Con-O, bench scale)	T:60 G:40 mL/min	99.4, NA	NA	[91,92]
	Ammonia	Sieve plate (CC)	C:10–14; T:25–55 °C	95–99, 1.2	$2NH_3(g) + CO_2(g) \rightleftharpoons NH_2COONH_4(s)$ $NH_2COONH_4(s) + H_2O(g) \rightleftharpoons (NH_4)_2CO(s)CO_3(s)$	[88,89]
	Piperazine	Stirred cell (SC, BS)	P:0.032 T:42 and 0.042	100, 0.32	1st order partial reaction occurs	[93]
	Ionic liquids	Double stirred cell (BS)	T:25–50; P:0.1; A:0.5–1.2	99.11 at 60 °C,	NA	[94–97]
Mixed Solvents	DEA-K <sub>2</sub> CO <sub>3</sub>	Split flow (CC, bench scale)	T:115 L:63.66 m <sup>3</sup> /h	99, NA	Promoter selection is very critical. It is a reversible exothermic reaction $CO_2 + K_2CO_3 + H_2O \leftrightarrow 2KHCO_3$	[98]
	PEI-SiO <sub>2</sub> Alcohol/amine/water	Packed (bench scale)	L:33.66 m <sup>3</sup> /h	NA, NA	$q_{sensible} = \frac{C_p \Delta m_{solution}}{\Delta m_{CO_2}}$	[99,100]
	BDA-DEEA	Packed (CC, BS)	T:40 (absorption) T:90 (desorption) G: 24.78 m <sup>3</sup> /h	46 (HCL), 48 (HCC), 11(HCE) than MEA with 5 M	Carbamate and bicarbonate formations	[101]
	AMP-PZ	Packed (pilot)	L/G:2.9; packing height=10 m	90, NA	-	[102–109]

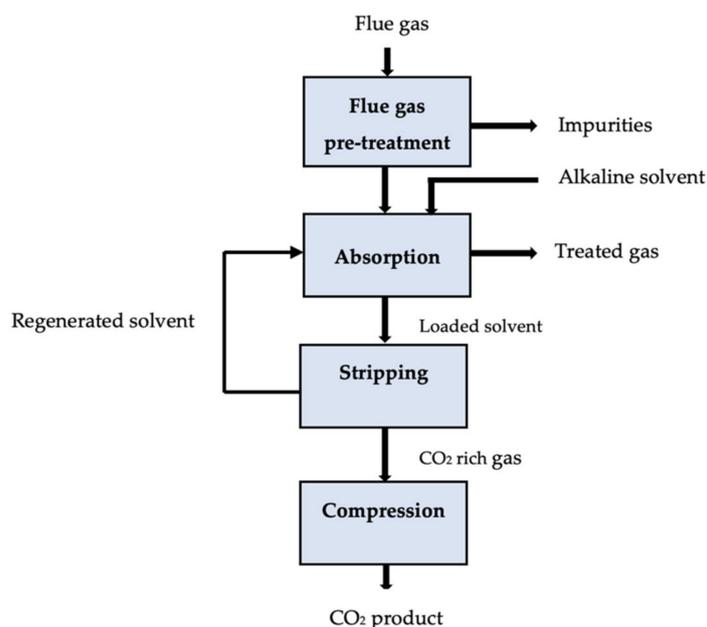


Figure 3. Schematic of an absorption carbon capture process [110].

### 5.2. Adsorption

The process of adsorption [43] involves molecules in liquids and gases adhering to solid surfaces by weak van der Waals interactions. Unlike liquid absorbent processes, solid adsorbents bind CO<sub>2</sub> to their surfaces during adsorption. Selection criteria for this sorbent include a large surface area, high regeneration capability, and high selectivity. Common

adsorbents include activated carbon, molecular sieves, zeolites, lithium zirconate, and hydrotalcite [27]. Table 4 highlights the potential adsorbents and adsorption parameters for CO<sub>2</sub> capture.

It is possible to achieve CO<sub>2</sub> adsorption by changing the pressure or temperature of a saturated sorbent. Pressure swing adsorption (PSA) is a commercially applied technology that recovers more than 85% of CO<sub>2</sub> from power plants [111,112]. A solid adsorbent selectively adsorbs CO<sub>2</sub> at high pressures, then the solid desorbs, releasing CO<sub>2</sub> for low-pressure transport (usually atmospheric pressure). The temperature swing adsorption (TSA) releases the CO<sub>2</sub> in the system by increasing its temperature through steam injection or hot air distribution [113]. A CO<sub>2</sub> purity of over 95% and recovery of over 80% are possible when using CO<sub>2</sub> regeneration, although regeneration is more time-consuming than PSA [114]. It was estimated that the operating costs of a particular TSA process ranged between USD 80 and 150 per tonne of CO<sub>2</sub> captured [115]. Significant attention has been paid to developing CO<sub>2</sub> capture sorbents from agricultural and industrial wastes to lower the overall cost of CO<sub>2</sub> capture. An adsorption carbon capture process is shown in Figure 4.

**Table 4.** Summary of adsorption-based carbon capture.

Adsorbent	Reactive Separator	Operating Conditions P, T, C, G	CO <sub>2</sub> Capture (%), Ad-C (gCO <sub>2</sub> /gads)	Kinetics/Mass Transfer	Ref.
TEPA-Mg-MOF-74	PBR (LS)	Regeneration temp is 250–300 °C	4–4.9 wt. %, 8.31 mmol CO <sub>2</sub> /g adsorbent, NA	N <sub>2</sub> adsorption–desorption isotherm	[109]
ZX-APG,	PBR (3-bed, 8-step, VPSA, LS)	T:35; P: 0.007–0.008	85–95, NA, 73–82% CO <sub>2</sub> purity	Langmuir adsorption isotherm is adopted	[116]
Activated carbon	PBR (1 bed, 3 step, VSA, LS)	Water vapour (H <sub>2</sub> O): 4.6 mol%, Vf: 44; TDes:100T: 60, ICC:11.2, Bd:0.493, Lg:50, P:0.113, PVP = 3, Trpt:3; SA:921.7, PV:0.37, Tads:35	69.5, NA	Dual-site Langmuir equation has been adopted	[117]
NPC10	PBR (TSA, LS)	T: 25, P: 0.1, SA: 639	NA, 0.041	Langmuir adsorption isotherm	[118]
Fly ash + PEI + PEG	PBR (LS, TSA)	St: 24 h, P: 0.11, T: 70	4.5 at 85 °C	$\text{CO}_2 + 2\text{RR}'\text{NH} \leftrightarrow \text{RR}'\text{NCOO}^- + \text{RR}'\text{NH}_2^+ \text{CO}_2$ $+ 2\text{H}_2\text{O} + \text{RR}'\text{NCOO}^- \leftrightarrow 2\text{HCO}_3^- + \text{RR}'\text{NH}_2^+$	[119]
ZX	MBA (LS, PSA)	Bed dimensions (m): FRR: 0.5; CT: 650; AT: 950; SA: 1873.9; 2b: 0.03, Nm: 36, W: 1.5; L:1.5; Xpth: 0.012 Bd: 0.65, Cs: 1.07, Dp: 3420, ε: 0.31 ks: 0.275	80, NA, 97% purity	Extended Langmuir isotherm was used	[120]
Rayon–HCM	PBR (TSA)		97, 0.2	Langmuir adsorption isotherm adopted	[121,122]

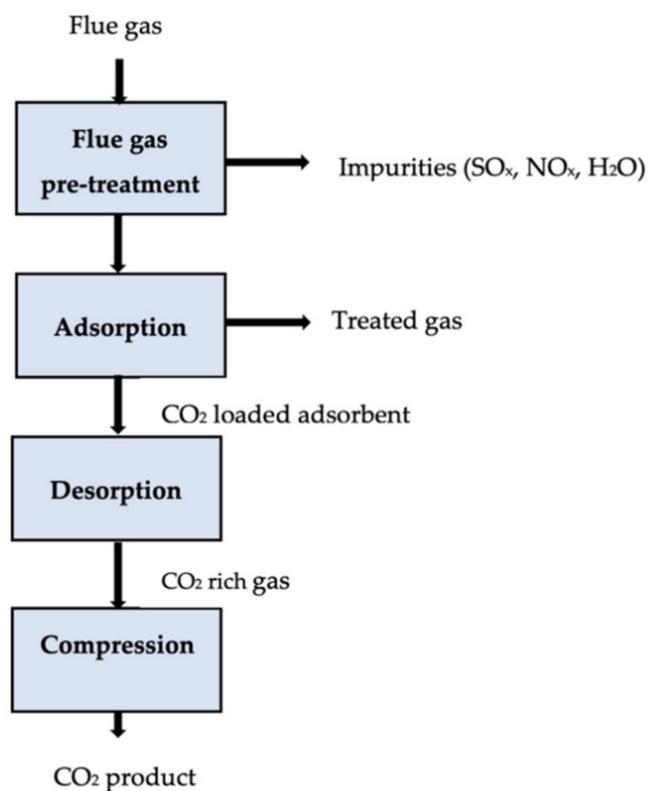


Figure 4. Schematic of an adsorption carbon capture process [110].

### 5.3. Chemical Looping Combustion

In contrast to oxy-fuel combustion, which uses pure oxygen for combustion, metal oxides are used as oxygen carriers in combustion. Metal oxides are reduced to metal during the process, while fuels are oxidized to create CO<sub>2</sub> and water. In a subsequent stage, the metal is oxidized and recycled. The removal of water by condensation from the process byproducts is easy, but the separation of pure CO<sub>2</sub> requires no energy. Numerous low-cost metal oxides, including Mn<sub>2</sub>O<sub>3</sub>, CuO, NiO, and Fe<sub>2</sub>O<sub>3</sub>, are suitable for this process. The potential sorbents and processes of chemical looping combustion are highlighted in Table 5.

Several researchers [123–127] have examined the performance efficiency of various metal oxides in this process. According to Adánez, de Diego [126], a metal oxide can be optimized by using support inert materials, but the selection of an inert material will vary depending on the characteristics of the metal oxide. Chemical looping combustion (CLC) was studied by Lyngfelt, Leckner [128] in a boiler consisting of two fluidized beds. Lyngfelt, Leckner [128] recently reviewed this technology. This process has been demonstrated to be a very promising CO<sub>2</sub> capture technology by both Lyngfelt, Leckner [128] and Adánez, de Diego [126]. The IGCC’s CO<sub>2</sub> separation is based on pre-combustion, but Erlach, Schmidt [113] found chemical looping combustion to have a 2.8% higher net plant efficiency than the former method. Figure 5 illustrates the basic CLC system.

Table 5. An overview of chemical looping combustion-based carbon capture.

Fuel Type	Operating Conditions P, T, C, G	Reactive Separator	CC (%), Purity (%)	Challenges	Kinetics/Mass Transfer	Ref.
Coal, C <sub>2</sub> H <sub>5</sub> OH, Isooctane, C <sub>3</sub> H <sub>8</sub> and CH <sub>4</sub> .	T: 200–1200; molar ratios of carbon/CaSO <sub>4</sub> = 0.5 and carbon/steam = 1	TGA	NA, 93 (with CaSO <sub>4</sub> at 850–975 °C)	The ΔHr is dependent on the fuel but not the amount of OC utilized. The yield depends on OC.	Combustion of iso-octane (–5101.58 kJ/mole) with Na <sub>2</sub> SO <sub>4</sub> and CaSO <sub>4</sub> produces without SO <sub>2</sub> formation between 200 °C and 344.3 °C.	[129,130]

Table 5. Cont.

Fuel Type	Operating Conditions P, T, C, G	Reactive Separator	CC (%), Purity (%)	Challenges	Kinetics/Mass Transfer	Ref.
Syngas, H <sub>2</sub>	XOC: 80–95, HR: 90–99, T: 370–1030	2-stage PBR-CLC	100, NA	PP of O <sub>2</sub> in reactors; high solid inventories.	The packed bed of OC reduces the need for highly efficient cyclone to reduce costs; boron nitride (BN) used as the dense support material due to high thermal conductivity, low thermal expansion and high thermal stability.	[131–134]
Coal, kerosene, biomass	Bd: 4.750; Dp: 128 Umf: 0.0129, Φ: 0.64	IFBR	83–99.3% at 800–950 °C, NA	Scale-up, fuel conversion, agglomeration and attrition.	ΔP <sub>RC</sub> increases linearly with solid flow rate.	[135–141]
CH <sub>4</sub> , coal	Iron oxide: 950 °C, FF: 1.18, CO <sub>2</sub> EF: 10, DT: 5.25	CMBS or RPBR (1 MWth)	>99, >95	Reaction heat exceeds the convective heat-transfer rate to the gas flow.	The reduction kinetics and activation energy parameters are critical to find fuel conversion efficiency, temperature distribution and carbon separation efficiency.	[142,143]
CH <sub>4</sub> , syngas	T: 700–975; SITC:20–30; SFRR: 8–10 for CO SFRR:4–12 for H <sub>2</sub> Fsolids:1.7–2.5	CC-MBR	>99% CH <sub>4</sub> and 100% syngas conversion. >99.99% H <sub>2</sub> purity.	The formation of FeO and FeAl <sub>2</sub> O <sub>4</sub> indicates further utilization of oxygen in iron-based OC's can be achieved. –φ > 1.14.	At 900 °C, the reduction of Fe <sub>2</sub> O <sub>3</sub> to Fe with CO generates 37.7 KJ/mol Fe <sub>2</sub> O <sub>3</sub> of heat but its reduction with H <sub>2</sub> gas needs 61.8 KJ/mol Fe <sub>2</sub> O <sub>3</sub> of heat.	[143,144]

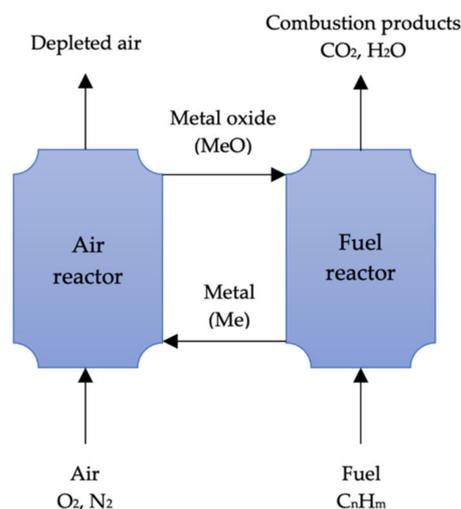


Figure 5. Schematic of chemical-looping combustion (adapted from Yang [145]).

#### 5.4. Membrane Separation

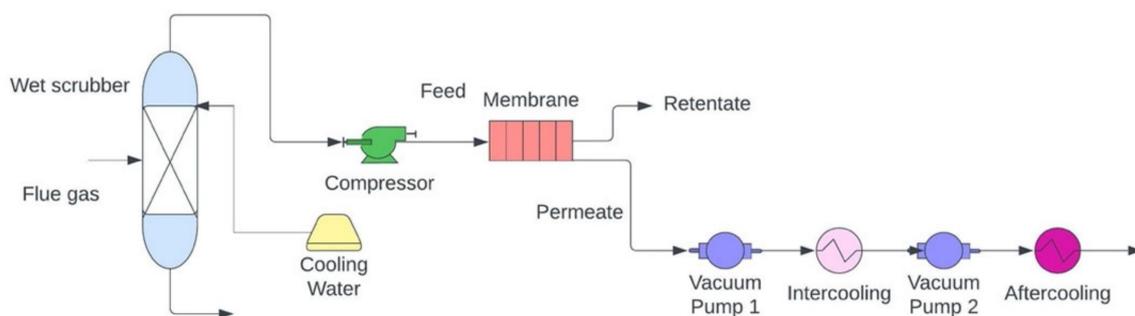
Membrane separation uses a semipermeable membrane or barrier to physically separate CO<sub>2</sub> from other flue gases [146]. Membrane separation uses less energy than traditional solvent absorption methods, making it less expensive [147]. Membrane separation has successfully been used for selective gas separation in a variety of fields for the past two decades, including natural gas sweetening, air separation, hydrogen production, and bio-gas upgrading. Researchers are working on developing membrane-based materials to separate CO<sub>2</sub> released by various industries. Furthermore, this technology has produced increased efficiency in terms of both the economy and the environment [148]. Scientists have developed a variety of different membranes for CO<sub>2</sub> separation, including inorganic membranes, polymers, carbon molecular sieve membranes (CMSMs), microporous organic polymers (MOPs), and mixed matrix membranes (MMMs) [149]. The potential sorbents and processes of membrane separation are highlighted in Table 6.

In addition, membrane separation technology can also separate gases in CCS processes such as pre- and post-combustion capture. It is generally considered that polymeric membranes are more flexible, durable, and efficient at capturing CO<sub>2</sub> from industrial processes. An upper bound relationship analysis describes how selectivity and permeability are related to CO<sub>2</sub> capture by polymeric membranes [150]. To improve results, glassy and rubbery materials with varying separation principles based on their size and diffusion

ability can be used to synthesize polymeric membranes. The condensability and differences in kinetic properties of gas molecules are responsible for gas separation by glassy and rubbery polymers [151]. Considering how difficult it is to examine operating conditions for rapid performance, membranes applied in gas separation systems are typically modeled to determine their working capacity [152]. For optimal results in industrial settings, membrane performance must not be interfered with by flue gas impurities [149]. Researchers were able to separate CO<sub>2</sub> from other gases with an efficiency of 82–88% [153,154]. In fact, despite membrane materials having poor permeability and selectivity [155], it is also problematic to use this extraction method in flue gas with low pressure and CO<sub>2</sub> concentration in flue gas conditions [156]. A membrane carbon capture process is displayed in Figure 6.

**Table 6.** Summary of membrane-based carbon capture.

Membrane	Reactive Separator	Operating Parameters	Challenges	Kinetics/Mass Transfer	Ref.
Dense membranes	Hollow fiber and flat-sheet	S-P, T, P, L <sub>a</sub> , pressure ratio of the permeate side to the feed side, pore size and porosity	Lower selectivity at higher permeability	Solution–diffusion; among the mechanisms are Knudsen diffusion and the molecular sieve effect	[157]
Micro-porous Membranes	Hollow fiber and flat-sheet	P, T, pore size and ε of the membrane–membrane wettability	Wetting of the membrane	Reaction kinetics depend on solvent	[157]
		Gas flow area	There are other compounds present in the gas stream	Even at high pressures, K <sub>o</sub> is controlled by the resistance of the liquid film	[158]
		Liquid flow area	Solvent volatility and limited long-term stability	Pore diffusion depends on membrane support	[159]
Liquid in the membrane pores	Flat-sheet only	G <sub>a</sub> , L <sub>a</sub> , VVIS, P, T	Solvent “wash-out” causes the membrane’s stability to decrease	The overall mass transfer coefficient	[160]



**Figure 6.** Schematic of membrane carbon capture process (adapted from Wang [161]).

### 5.5. Cryogenic Distillation

This process separates CO<sub>2</sub> from gas mixtures by focusing on their boiling points at temperatures ranging from 100 to 135 °C [6,43]. In the presence of high pressures (100–200 atm), solidified CO<sub>2</sub> provides two significant benefits: a lack of solvents and liquefied CO<sub>2</sub> for more convenient transport and injection [37]. It does, however, have some drawbacks that need to be investigated further, as do other processes. When cold and pressurized nitrogen is used as a refrigerant, ice formation compromises equipment safety, causing pressure fluctuations and pipe blockages, as well as increasing the consumption of energy [37,162]. This enhanced CO<sub>2</sub> separation can nullify the need for refrigerant preparation and storage [162,163]. However, CO<sub>2</sub> is separated using cryogenic distillation coupled with biogas upgrading. A comparison of different separation methods for CO<sub>2</sub> capture is shown in Table 7.

**Table 7.** Current status of different separation technologies for CO<sub>2</sub> capture [16,37,54,111–114,161,164–187].

Parameter	Chemical Absorption	Physical Absorption	Adsorption	Chemical-Looping Combustion	Membrane Separation	Cryogenic
Separation technique	Amine, chilled ammonia, and amino acid salt solvent.	Rectisol, Selexol, etc. Mostly integrated gasification combined cycle.	Pressure swing adsorption and pressure–temperature swing adsorption.	FeO, CuO, MnO, and NiO	Polymeric, inorganic and mixed membranes.	Cryogenic distillation.
Pros	High reactivity, low cost of the solvent, and low molecular weight result in a high mass-based absorption capacity, and moderate thermal stability and thermal degradation rate.	Highly recommended for separating CO <sub>2</sub> during pre-combustion processes that operate at elevated CO <sub>2</sub> partial pressures. Captures CO <sub>2</sub> selectively from a gas stream without a chemical reaction	Recycling is possible since it is a reversible process. It is possible to achieve high adsorption efficiency (485%). Low waste generation.	Very high CO <sub>2</sub> concentration. Low-cost oxygen carrier materials. Truly and directly reduces the atmospheric CO <sub>2</sub> concentration. Viable alternative for CO <sub>2</sub> capture from mobile and decentralized sources.	No regeneration processes. Less solid waste produced. Less chemical consumption. High efficiency (>95% for single metal).	High capture efficiency (up to 99.9%). Mature technology. For many years, CO <sub>2</sub> has been recovered in the industry by this method.
Cons	Relatively high maintenance cost.	High energy is required to compress feed gas to a high pressure. Low CO <sub>2</sub> solubility. Less efficient absorption process. Large equipment sizing.	Requires adsorbent capable of operating at elevated temperatures. The significant amount of energy needed for CO <sub>2</sub> desorption is high.	Currently, the process is under development, and large-scale operations have not yet been carried out.	Fouling and low fluxes are examples of operational issues. High running costs. Removal (%) decreases with the presence of other metals.	High energy requirement due to refrigeration. High capital expenditure. Need for removal of water, NO <sub>x</sub> , SO <sub>x</sub> , and other trace components to avoid the freezing and eventual blockage of process equipment. The procedure consumes a significant amount of energy.
CO <sub>2</sub> concentration (vol.%)	<30.4	>59.3	28–34	3–8	11.8	<90
CO <sub>2</sub> capture efficiency (%)	95	>90	<85	52–60	90	99.9
CO <sub>2</sub> capture cost (USD/tonne CO <sub>2</sub> )	26.2	25.1	6.94	16–26	3–10	32.7
CO <sub>2</sub> purity (%)	99	<99	99.98	>96	95	99.95
Status of research and development	SaskPower, Saskatchewan, Canada (Boundary Dam Carbon Capture Project) TransAlta Corporation, Alberta Canada (Project Pioneer Keephills 3 Power Plant) American Electric Power, OH, USA (Mountaineer Power Plant)	Summit Power Group, LLC, Seattle, USA (Texas Clean Energy Project) Don valley, Yorkshire, UK (Don Valley Power Project) Nuon Power, Buggenum, The Netherlands (Integrated gasification combined cycle plant)	Under developmental stage.	Less large-scale demonstration plants.	Schwarze Pumpe power station, Spremberg, Germany (Oxy-fuel technology) CS Energy: Callide Power Plant A, Queensland, Australia (Callide Oxy-fuel Project) OxyCoal, UK (Oxy-fuel technology)	Air Products and Chemicals, Inc., Pennsylvania USA

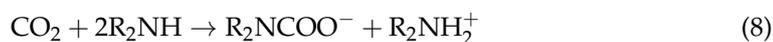
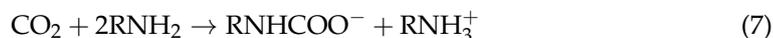
## 6. CO<sub>2</sub> Capture Using Dry Solid Sorbents

The selective separation of CO<sub>2</sub> based on interactions between gases and solids is required for CO<sub>2</sub> capture using a dry adsorbent [188]. In packed columns, universal dry adsorbents are typically utilized, including activated carbon and molecular sieves [189]. The surface tension and pore size of the adsorbent, as well as the process temperature and partial pressure, are critical factors in a dry adsorption process [190]. Adsorption and desorption cycles are repeated throughout the process (regeneration).

The following are the several adsorption types: (i) pressure swing adsorption (PSA) [4–6]; (ii) temperature swing adsorption (TSA) [191,192], which combines two processes—low-temperature adsorption followed by desorption or regeneration by raising the pressure; (iii) electric swing adsorption (ESA) [193], which entails adsorption and desorption by altering the electricity supply while a low-voltage current flows through the adsorbent; and (iv) vacuum swing adsorption (VSA) [194]. Additionally, due to their low energy needs and relative simplicity, adsorption-based technologies such as pressure/vacuum swing adsorption (PVSA) have been extensively researched [195–197].

To effectively treat high volumes of combustion emissions from numerous sources, it is imperative to increase a dry adsorbent's CO<sub>2</sub> capture selectivity and adsorptive capacity. The CO<sub>2</sub> adsorption can be enhanced and stabilized by adding functional groups to the surface of the adsorbent material that has a high affinity for CO<sub>2</sub> to react with it. CO<sub>2</sub> can then be selectively adsorbed using the adsorbent's sizeable specific surface area and pore structure [198–203].

The addition of different amine groups to solid materials used as CO<sub>2</sub> capture sorbents is anticipated to increase polarization and CO<sub>2</sub> capture to achieve high selectivity and capture performance. These adsorbents have several benefits, including the potential eradication of corrosion issues and reduced costs of energy for regeneration. Under anhydrous conditions, the reactions of CO<sub>2</sub> with amine functional groups result in ammonium carbamates, as follows [204]:



The reversible nature of the adsorption process and the possibility to increase adsorption efficiency by altering the composition of adsorbent materials make this process important. Therefore, by choosing a suitable adsorbent material, the CO<sub>2</sub> adsorption efficiency can be improved. Activated carbons [205,206], zeolites [207], hollow fibers, and alumina are currently the major commercially available adsorbents. Each material has a unique surface area, pore structure, and surface functional groups, and their application areas are very specific. The following section discusses and provides descriptions of some typical carbonaceous and non-carbonaceous dry sorbents for CO<sub>2</sub> capture.

### Non-carbonaceous adsorbents

Non-carbonaceous adsorbents include zeolites, metalorganic frameworks, silica, etc. The CO<sub>2</sub> uptake capacity of these materials is high; however, they are expensive and highly sensitive to moisture. As a result, zeolites can capture CO<sub>2</sub> molecules of 0.33 nm due to their suitable channel diameters (0.3–1.0 nm) [208].

#### 6.1. Adsorbents Based on Zeolite

Adsorbents with a natural structure, such as zeolites, are made up of an interlocking structure of AlO<sub>4</sub> and SiO<sub>4</sub> tetrahedrons that share atoms of oxygen [209]. Small pore size and high porosity are responsible for zeolite's excellent CO<sub>2</sub> adsorption at temperatures as low as 30 °C.; however, their CO<sub>2</sub> capture capacity rapidly declines above this temperature and is almost non-existent above 200 °C [209]. Zeolite's high hydrophilicity significantly reduces its CO<sub>2</sub> adsorption capacity when the gas contains water. In this case, high temperatures are required for the regeneration [210], resulting in significant energy losses as a result of the CO<sub>2</sub> regeneration [200].

The CO<sub>2</sub> selectivity of zeolites remains low despite their ability to separate gases [211–213]. The moisture content of the gas is a constraint on zeolites' capacity to adsorb CO<sub>2</sub> from flue gas. Water competes for adsorption with CO<sub>2</sub>, reducing the amount of CO<sub>2</sub> adsorbed by Y-type zeolites (CaY and NiY) [214]. By contacting an ion and a dipole at low desorption temperatures, physisorption occurs, resulting in a linear orientation of the CO<sub>2</sub> molecule [209,215].

Additionally, zeolites exhibit excellent chemical reactivity, recyclability, high stability, excellent recyclability, and structural diversity, which make them a promising candidate for the CO<sub>2</sub> adsorption [216]. This structure consists of a network of streams into which gas molecules are adsorbable, ranging from 0.5 to 1.2 nm in size [200,209,217]. Flue gas mixtures (CO<sub>2</sub>, H<sub>2</sub>O, CO) are not suitable for zeolites because they are not CO<sub>2</sub> selective, along with their limited capacity to adsorb at high temperatures [209,218,219].

Zeolite contains exchangeable cations including K<sup>+</sup>, Mg<sup>2+</sup>, and Na<sup>+</sup> that act as a balance to the negative charge that results from a SiO<sub>4</sub> tetrahedron replacing an AlO<sub>4</sub> tetrahedron. Specifically, the charge density, cation distribution, and size of the zeolites influence their adsorption and separation properties. Zeolite particles have a large pore size that allows CO<sub>2</sub> to diffuse into them. The adsorption of CO<sub>2</sub> molecules by zeolites is increased by the presence of cations in their structure via molecules and the adsorbent's interaction electrostatically [220]. CO<sub>2</sub> adsorption has been studied mostly for the zeolites 13X and 5A. Using a type 13X zeolite, Moura, Bezerra [221] investigated the effect of cation exchange on CO<sub>2</sub> adsorption, concluding that zeolites rich in alkali-cations, such as Li<sup>+</sup> and Na<sup>+</sup>, could effectively adsorb CO<sub>2</sub>. The order of decreasing adsorption capacity was Li<sup>+</sup> > Na<sup>+</sup> > NH<sub>4</sub><sup>+</sup> > Ba<sup>2+</sup> > Fe<sup>3+</sup>. According to Walton [222], it is believed that the ionic charge of zeolites, the ionic radius, the shielding effects, as well as the nuclear charge are the main causes of the observed decline in CO<sub>2</sub> adsorption capacity. According to Calleja, Jimenez [223], zeolites 13X and 5A have CO<sub>2</sub> adsorption capacities of 3–25 wt. % and 2–12 wt. %, respectively, at a CO<sub>2</sub> partial pressure of 15% and a CO<sub>2</sub> pressure of 100%.

## 6.2. Adsorbents Based on Metal-Organic Frameworks

The molecular organic framework (MOF) is a crystalline material containing metal ions or clusters of metal ions combined with organic molecules (ligands) [9,43]. As the MOF has been adapted to a variety of chemical processes and applications, over 20,000 MOFs have now been created, each with distinct geometries, pore sizes, and functions [224–228].

Having large mesopores, high surface areas (approximately 10,000 m<sup>2</sup>/g), and an adaptability to different geometries, MOFs are ideal chemisorbents for CO<sub>2</sub> capture [228,229]. Although MOFs have some advantages when it comes to CO<sub>2</sub> capture at high temperatures, they also have some drawbacks. For example, moisture is absorbed while CO<sub>2</sub> is being captured, the manufacturing process is difficult, and the MOFs are not very durable [200].

In a study by Szczeńsiak and Choma [230], copper-based MOFs (Cu-BTC), with benzene-1,3,5-tricarboxylate, had an uptake capacity for CO<sub>2</sub> of 9.59 mmol/g adsorbent and a surface area of 1760 m<sup>2</sup>/g at 1 atm at 273 K. The CO<sub>2</sub> capacity of most MOFs decreases noticeably as the temperature increases during the CO<sub>2</sub> capture phase. At 298 K, 313 K, and 328 K, the temperatures for CO<sub>2</sub> capture, Aarti et al. [231] reported 4 mmol/g adsorbent for Cu-BTC-PEI-2.5, 2.61 mmol/g adsorbent for Cu-BTC-PEI-2.5, and 1.66 mmol/g adsorbent for Cu-BTC-PEI-2.5, respectively, due to MOFs' structural stability's degradation with temperature.

There has been a breakthrough in developing microporous coordination polymers (MCPs) to decrease the production cost of MOFs. As the name implies, MCPs are composed of repeating channels of metal ions linked by ligands whose function is to link ions. MCPs have porous structures because of their inherited organic functionality [232]. The magnesium-based MOF Mg-MOF-74 was found to have better CO<sub>2</sub> adsorption with 8.61 mmol/g adsorbents at 1 bar partial pressure and 298 K [233]. Magnesium dioxo-benzenedicarboxylate (Mg/DOBDC) is an MCP composed of Mg<sup>2+</sup> ions connected to 2,5-dioxide-1,4-benzene-dicarboxylate [232]. Researchers have claimed that the high performance can be attributed to the improved ionic behavior of the magnesium oxide Mg-O bond when CO<sub>2</sub> partial pressures are as low as 0.1 bar. As opposed to MgO, exothermic car-

bonation does not form magnesium carbonate ( $\text{MgCO}_3$ ) from  $\text{Mg}/\text{DOBDC}$ . MCP networks are inflexible, and prevent insertion into the  $\text{MgO}$  bonds likely due to their inflexibility. Due to the highly ionic nature of the  $\text{Mg-O}$  bonds in  $\text{Mg}/\text{DOBDC}$ , the material can capture  $\text{CO}_2$  reversibly. Low  $\text{CO}_2$  partial pressures can be improved by these techniques [232].

The ability of many early MOF adsorbents to adsorb large amounts of  $\text{CO}_2$  was demonstrated. To be used as  $\text{CO}_2$  adsorbents, MOFs must be stable in an aqueous medium. Mahdipoor, Halladj [234] reported an MOF  $\text{CO}_2$  adsorbent containing iron-based on poly terephthalate (BDC). Observations revealed that the amino-functionalized MOF MIL 101(FE) is water- and ethanol-stable, and capable of capturing 13 mmol/g  $\text{CO}_2$  per adsorbent. Additionally to the physical  $\text{CO}_2$  adsorption by the amino MIL 101(FE) MOF's structure, the chemisorption of  $\text{CO}_2$  by the MIL 101(FE) MOF structure affects the total  $\text{CO}_2$  adsorption by the MOF [234].

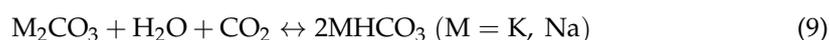
### 6.3. Mesoporous Silica Materials

Mesoporous silica has also been proposed as a viable candidate material for capturing  $\text{CO}_2$  because of its large surface area and high capacity to be synthesized with a variety of pore hole sizes [235]. The modification of mesoporous silica for  $\text{CO}_2$  capture has been proven to be an effective method for developing adsorbents, despite its low adsorption capacity [235]. The possibility of chemically treating their surface OH groups to promote their  $\text{CO}_2$  selectivity and adsorption capacity may be further investigated to process flue gases at low pressures using  $\text{CO}_2$  [235].

Sánchez-Zambrano, Lima Duarte [236] investigated the chemical modifications of mesoporous silica treated with 3-aminopropyl triethoxysilane (APTES), and then impregnated it with polyethyleneimine (PEI) to find out how the modifications affected kinetic mechanisms, site energy distributions, and  $\text{CO}_2$  adsorption under post-combustion conditions [10,43]. As determined by microcalorimetry, the functionalization process resulted in new adsorption sites. When amine groups were added to the support, physisorption accounted for the vast majority of adsorption, and  $\text{CO}_2$  capture capacity and selectivity increased as the temperature increased (50 and 75 °C). Due to sites that could not be restored due to strong chemical bonds formed by adsorption products, using a turbomolecular vacuum pump was the only way to regenerate after adsorption at 25 °C with  $\text{CO}_2$ . In the first stage of  $\text{CO}_2$  capture, these sites became available for regeneration, but only at higher temperatures [236]. Although mesoporous silicas are highly effective adsorbents for  $\text{CO}_2$  capture due to their large pores with a tunable size, high surface area, and mechanical and thermal stability, they do not possess sufficient  $\text{CO}_2$  adsorption abilities to be of any use, especially at a 1-atmosphere pressure [235].

### 6.4. Alkali Metal-Based Materials

Alkali metal carbonates, including those made of Na, K, and Al, have been reported to be efficient dry adsorbents for  $\text{CO}_2$  capture from flue gas operating below 473 K in relatively moderate conditions [237,238]. Various inorganic supports, including alumina, silica, ceramics, zirconia, and carbon materials, are added with alkali metal carbonates during this procedure; Equation (9) explains how moisture and  $\text{CO}_2$  react, facilitating  $\text{CO}_2$  adsorption, and how decarbonization (Equation (10)) regenerates the adsorbent (10) [38]:



141 kJ/mol and  $\Delta H = -135$  kJ/mol for  $\text{M} = \text{K}$  and  $\text{Na}$ , respectively



As shown in Equation (8), alkali-metal bicarbonates are typically formed when  $\text{CO}_2$  and  $\text{H}_2\text{O}$  react with carbonate sorbents between 333 and 383 K, which then regenerate alkali-metal carbonates at 373–473 K, releasing  $\text{CO}_2$ . Theoretically,  $\text{Na}_2\text{CO}_3$  and  $\text{K}_2\text{CO}_3$  have  $\text{CO}_2$  adsorption capacities of 41.5 and 31.8 wt. %, respectively.

The use of lithium-based materials such as lithium-based silicate ( $\text{Li}_4\text{SiO}_4$ ) and lithium-based zirconate ( $\text{Li}_2\text{ZrO}_3$ ) for direct  $\text{CO}_2$  capture from flue gas at temperatures of 700–900 K is another promising method for capturing  $\text{CO}_2$  [239].  $\text{Li}_4\text{SiO}_4$ , specifically, is a promising  $\text{CO}_2$  captor because of its low volume change during  $\text{CO}_2$ -adsorption-desorption and its high  $\text{CO}_2$  sorption capacity of 36.7% [240].

In a study conducted by Kato et al.,  $\text{Li}_4\text{SiO}_4$  and  $\text{Li}_2\text{ZrO}_3$  were examined at low  $\text{CO}_2$  concentrations, i.e., 50 ppv. They found that  $\text{Li}_4\text{SiO}_4$  was 30 times more capable of absorbing  $\text{CO}_2$  than  $\text{Li}_2\text{ZrO}_3$ . Additionally, zirconia materials are more expensive than silica materials [241]. Recently, Seggiani et al. [242] reported that  $\text{Li}_4\text{SiO}_4$ , with the addition of 30%  $\text{Na}_2\text{CO}_3$  or  $\text{K}_2\text{CO}_3$ , demonstrated a  $\text{CO}_2$  sorption capacity of 23 wt. % at an ideal sorption temperature of 853 K and low  $\text{CO}_2$  partial pressure of 0.04 bar, equating to a  $\text{Li}_4\text{SiO}_4$  conversion of almost 80%.  $\text{Li}_8\text{SiO}_6$  was proposed by Durán-Muoz et al., as a substitute dry adsorbent for  $\text{CO}_2$  capture. It demonstrated a high sorption capacity of roughly 51.9 wt. % over a wide temperature range, with an efficiency of 71.1% [243].

It is technically and economically desirable to use alkali-metal-based materials to capture  $\text{CO}_2$  post-combustion at low temperatures and in low concentrations, since they do not require additional cooling processes; although, the long-term stability and sustained performance of such adsorbents under real flue gas conditions must be addressed.

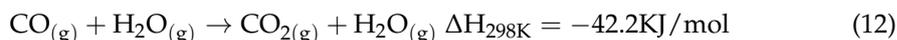
#### 6.5. Alkaline Metal-Based Ceramics

In addition to alkaline ceramics, binary metal oxides are also referred to as alkaline oxides because they contain more than one alkaline element. Metal-based alkaline ceramics include  $\text{Li}_5\text{AlO}_4$ ,  $\text{Li}_2\text{CuO}_2$ ,  $\text{Li}_2\text{ZrO}_3$ , and  $\text{Li}_4\text{SiO}_4$ . Notably, these alkaline metal ceramics can be regenerated at elevated temperatures. Consequently, they are ideal for adsorption and desorption cycles, such as those associated with post-combustion capture. The robustness of alkaline metal ceramics allows them to be made without refractory supports, resulting in a simpler synthesis method [244]. Alkaline ceramics are synthesized in a variety of ways, depending on the desired type. When synthesizing zirconates, co-precipitation, sol-gel, or soft chemistry methods are preferred for achieving the desired morphology of an adsorbent. A sorbent needs to have a morphology with a large surface area and a small particle size to capture  $\text{CO}_2$  to its maximum capacity [244]. The sol-gel method can be used to create a mesoporous structure for silicates that is advantageous [244].

These materials, including zirconate  $\text{Na}_2\text{ZrO}_3$ , are important for process intensification [245], owing to their catalytic activity [244]. Following  $\text{CO}_2$  chemisorption, the carbonated derivate,  $\text{Na}_2\text{CO}_3$ , is formed. Equation (11) depicts the reaction pathway for  $\text{Na}_2\text{ZrO}_3$  [244,245].



Sutton, Kelleher [246] described alkaline metal-based ceramics as catalysts of the water gas shift reaction (WGSR).



Compared to other ceramics made of alkaline metal, lithium cuprate ( $\text{Li}_2\text{CuO}_2$ ) has demonstrated the ability to adsorb  $\text{CO}_2$  over a wide temperature range (120 to 690 °C), and it retains its efficacy as a  $\text{CO}_2$  adsorbent at low  $\text{CO}_2$  partial pressures [247,248]. At  $\text{CO}_2$  concentrations less than 15%, the alkaline metal zirconate ( $\text{Li}_2\text{ZrO}_3$ ) exhibits excellent  $\text{CO}_2$  selectivity and  $\text{CO}_2$  acceptor performance in flue gas streams [244]. In summary,  $\text{Li}_2\text{CuO}_2$  and  $\text{Li}_2\text{ZrO}_3$  are two  $\text{CO}_2$  adsorbents that have been subjected to extensive research and have been demonstrated to be promising dependable  $\text{CO}_2$  adsorbents [244].

Much of the research on alkaline ceramics in recent years has focused on  $\text{CO}_2$  capture at high temperatures. The  $\text{CO}_2$  adsorption capacities of two alkaline yttrium oxides ( $\text{NaYO}_2$  and  $\text{LiYO}_2$ ) at high temperatures were recently evaluated. Since lithium ions are not as close together as sodium ions are in an octahedron,  $\text{LiYO}_2$  has a greater capacity for  $\text{CO}_2$

capture than NaYO<sub>2</sub>. However, both ceramics were capable of adsorption/desorption for at least ten cycles [249]. Table 8 lists the basic CO<sub>2</sub> adsorption capacities of the main adsorbent types that are commonly addressed in the literature.

**Table 8.** The CO<sub>2</sub> adsorption capacity of the most common adsorbents [250–252].

Adsorbent	Examples	Temp. (K)	Press. (atm)	SBET(m <sup>2</sup> g <sup>-1</sup> )	Capacity (mmol g <sup>-1</sup> )
Zeolite-based	NaY, NaX, 13X, ZIF-70, ZIF-69	273–384	1	15–1730	≤5.4
Amine-based	Silica monolith/TEPA, MCM-41/PEI	298–384	1	16–367	≤5.9
MOF-based	MOF-53, MOF-177	198–304	1–96	270–4500	≤48.7
Calcium-based	Ca(OH) <sub>2</sub> , CaO	195–348	1	-	≤11.6
Alkali ceramic-based	Na <sub>2</sub> ZrO <sub>3</sub> , Li <sub>2</sub> ZrO <sub>3</sub>	500–600	1	-	≤6.5
Carbon-based	Activated. Carbon BPL, MAXSORP, Activated carbon	195–348	1	1150–3250	≤8

### Carbonaceous Adsorbents

Even though carbon is the only component of carbonaceous materials, they offer several advantages, such as high thermal/chemical stability, heat and electrical conductivity, bio-affinities, elasticities, and strengths [253–255]. Being lightweight, having a very high specific surface area, and having a large pore volume make them especially suitable for applications involving the storage of gases or adsorption [256,257]. Additionally, they offer benefits for CO<sub>2</sub> capture: (i) they are not moisture-sensitive; (ii) they are reasonably priced; (iii) their desorption/adsorption temperatures are below 373 K.; (iv) they may be employed at atmospheric pressure; and (v) their energy usage is minimal. Each of these factors has had an impact on recent research in this field.

#### 6.6. Activated Carbons

The textural characteristics and surface groups of carbon-based adsorbents are highly correlated with CO<sub>2</sub> adsorption capability [258,259]. Due to their materials' wide range of pore sizes, from micropores to macropores, activated carbons are not suitable for selective gas adsorption. Adsorption temperatures of less than 25 kJ/mol are typically seen in pristine carbon-based adsorbents, which have weak CO<sub>2</sub> affinities [260]. At 298 K and 0.1 pressure, the standard CO<sub>2</sub> adsorption capacity of activated carbon is ~5 wt. % [261,262].

By modifying the activation and preparation conditions, the pore structures of activated carbons can be easily regulated [263,264]. Additionally, the activated carbon's surface functional groups can be easily modified utilizing a variety of treatment methods [265,266]. Incorporating different basic groups into activated carbon has been extensively researched for improving the CO<sub>2</sub> affinity by enhancing the CO<sub>2</sub> adsorption capacity [267,268].

NiO-loaded activated carbons (NiO-ACs) were synthesized by Jang et al., utilizing a post-oxidation technique that involved nickel electroless plating at 573 K in an air stream. The NiO-AC samples' ability to adsorb CO<sub>2</sub> increased as oxidation duration increased. The maximal CO<sub>2</sub> adsorption capacity was 49.9 cm<sup>3</sup>/g, above the 41.2 cm<sup>3</sup>/g capacity of unaltered activated carbon at 298 K and 1 bar. They reported that the acid-base characteristics of the NiO caused it to serve as an electron donor on the carbon surface, increasing the CO<sub>2</sub> adsorption, which acts as an electron acceptor [269].

#### 6.7. Graphene

A graphene derivative known as graphene oxide (GO) can be created using different functional groups on the edges and basal planes [270]. Researchers have extensively studied modifying the surfaces of GO with functional groups for applications including gas storage, separation, conversion of energy, and synthesizing newly developed GO-like derivatives with lightweight frameworks [271–273].

Thermally exfoliated graphene nanoplates were reported by Meng et al. as being innovative, highly effective sorbents for CO<sub>2</sub> capture. At 298 K and 30 bar, the produced graphene nanoplates demonstrated remarkable capture efficiencies of 248 wt. %. The

graphene nanoplates' wider inter-layer spacing and substantial inner void volume were attributed to the higher CO<sub>2</sub> capture capacity [273].

#### 6.8. Ordered Porous Carbons

Since they are widely used as electrode materials, catalyst supports, and other types of materials, ordered porous carbon materials have drawn a lot of study interest [274–276]. There are numerous ways to make ordered porous carbons, including (i) direct synthesis utilizing organic self-assembly, which uses a mixture of carbon precursors and blocks co-polymers as soft templates, and (ii) nano-casting, employing silica materials as structure-controlling hard templates [277,278].

Yoo et al., examined the impact of the phenolic resins' carbonization temperature on the total pore volumes and the specific surface areas of ordered nanoporous carbons (ONCs). Due to its greater specific surface area and smaller micropore size distribution, ONC carbonized at 1173 K had the highest CO<sub>2</sub> adsorption capacities at 298 K (15.8 wt. % at 1 pressure and 68.5 wt. % at 30 bar) [279].

#### 6.9. Activated Carbon Fibers (ACFs)

ACFs are attractive adsorbent materials because of their numerous micrometer porosities, nanostructures, and other characteristics, including narrow pore size distributions and large specific surface areas [280,281]. Compared to granular and powdered adsorbents, ACFs are more flexible due to their fibrous structure [282].

For large-scale CO<sub>2</sub> capture testing, Thiruvengatchari et al. created huge honeycomb-shaped carbon fiber composite (HMCFC) adsorbents (the adsorbent mass in one column was 4.486 kg). For a simulated flue gas with 13% CO<sub>2</sub>, 5.5% O<sub>2</sub>, and the remaining N<sub>2</sub> at 293 K, the average CO<sub>2</sub> adsorption capacity was 11.9 wt. %. Additionally, they demonstrated that the thermal decomposition process and combined vacuum improve the CO<sub>2</sub> collection efficiency of the HMCFC adsorbents [283].

### 7. Research Progress in Converting CO<sub>2</sub> into Valuable Fuels

One of the most stable compounds with carbon in its greatest valence state is CO<sub>2</sub>. Due to its low electron affinity, electrophilic reactions are challenging. Thus, a nucleophilic assault on the carbon atom is necessary for the conversion of CO<sub>2</sub>. As is well known, the dissociation energy required to rupture the C=O bond within a CO<sub>2</sub> molecule is greater than 750 kJ mol<sup>-1</sup> [284]. From a thermodynamic perspective, this is an uphill reaction. To provide the necessary energy for such a reaction to be completed, high temperatures, high pressures, or extremely effective catalysts are frequently required.

To date, a range of methods have been employed to reduce CO<sub>2</sub>, including the thermal catalysis [285–289], photocatalysis [290–293], electrocatalysis [294–297], and photoelectrochemical (PEC) reactions [298–301], in which heat, light, or electricity are utilized to supply the reaction's necessary energy. Eight electrons are required for each CO<sub>2</sub> molecule to convert into a hydrocarbon molecule completely. Due to the large number of compounds that are created during the reduction process, the purification procedure is complicated, and the yield of the desired products is decreased. Table 9 presents a clear comparison of the performance of different CO<sub>2</sub> conversion methods employed in recent years. Extremely concentrated and efficient enzymatic processes were added to the aforementioned reduction technologies to boost the efficiency and accuracy of the CO<sub>2</sub> conversion [302,303]. These reactions were inspired by natural photosynthesis. The schematic of the most modern CO<sub>2</sub> chemical conversion techniques is shown in Figure 7.

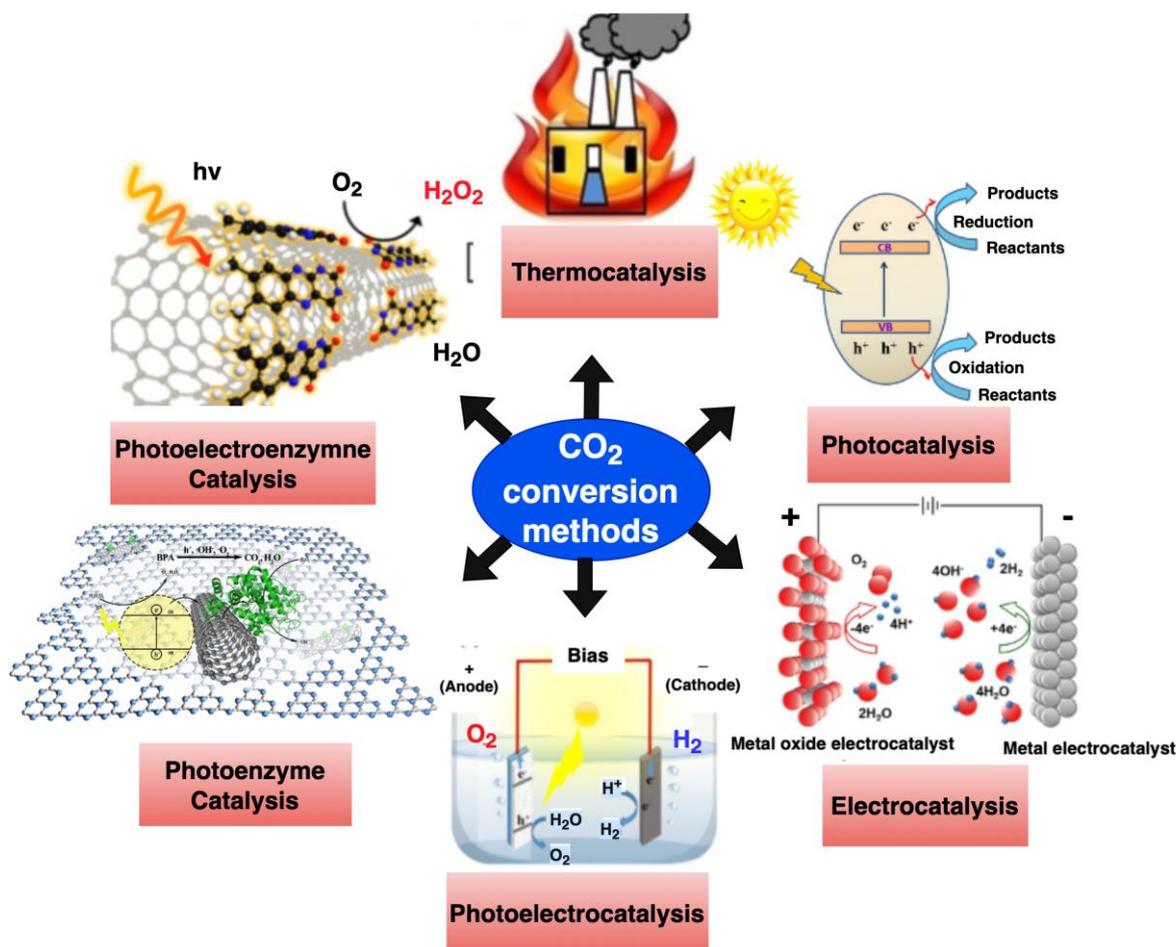


Figure 7. A schematic of the CO<sub>2</sub> chemical conversion methods [304].

Table 9. Performance evaluation of various CO<sub>2</sub> reduction systems.

Materials	Method of CO <sub>2</sub> Conversion	Efficiency	Reference
CuO + Cu <sub>2</sub> O	Photo electrocatalysis	CH <sub>3</sub> OH, 95%, 85 mM at −0.2 V vs. standard hydrogen electrode (SHE) after 1.5 h	[305]
C <sub>60</sub> polymer film	Photocatalysis	HCOOH, 239.46 μM after 2 h	[306]
Ni-Ru/Al <sub>2</sub> O <sub>3</sub>	Thermal catalysis	CO <sub>2</sub> conversion: 82.7% CO <sub>2</sub> selectivity: 100%	[307]
Polyoxometalates (POMs)	Electrocatalysis	38.9 mA cm <sup>−2</sup>	[308]
Co-Pi/Fe <sub>2</sub> O <sub>3</sub>	Photoelectro/enzymatic catalysis	HCOOH, 6.4 μM h <sup>−1</sup>	[309]
Plain graphite rod	Photo/enzymatic catalysis	HCOOH, 15.49 μM mg Enzyme <sup>−1</sup> min <sup>−1</sup>	[310]

As a result of the use of different CO<sub>2</sub> conversion strategies, such as thermal catalysis, photocatalysis, electrocatalysis, photoenzymatic catalysis, photoelectroenzymatic catalysis, and photoelectrochemical (PEC), a range of yields and products were obtained. Table 10 the numerous value-added products made using different CO<sub>2</sub> utilization strategies and the catalysts used throughout the conversion process.

**Table 10.** List of catalysts utilized in CO<sub>2</sub> conversion processes to manufacture valuable products.

Catalysts	Products	Methods	Reference
CuInS <sub>2</sub> thin film	Methanol	Photoelectrochemical	[311]
Sulfur modified copper	Formate	Electrochemical	[312,313]
Indium	Formate and acetate	Electrochemical	[314]
W <sub>18</sub> O <sub>49</sub>		Photocatalytic	[315]
p-type GaP	Methanol	Photoelectrochemical	[316]
Zn <sub>2</sub> GeO <sub>4</sub> nanoribbon	Methane	Photocatalytic	[317]
Bi <sub>2</sub> WO <sub>6</sub> nanoplate	Methane	Photocatalytic	[318]
HNb <sub>3</sub> O <sub>8</sub> nanobelt	Methane	Photocatalytic	[319]
TiO <sub>2</sub>	Methanol and methane	Photocatalytic	[320]
PET supported TiO <sub>2</sub>	Carbon monoxide	Photocatalytic	[321]
Nickel (Ni)	Methane	Thermal (Reforming)	[322]

## 8. Technologies for CO<sub>2</sub> Capture at Various Technological Readiness Levels (TRL)

The global concern over the effects of industrial operations, such as chemical processes, on the environment has increased interest in green technologies. This issue has fueled significant growth in research and new technologies across many fields since the turn of the 21st century. Due to their significant contributions to high greenhouse gas emissions, the power generation, agricultural, and chemical industries stand out numerically in developing green technologies. It is crucial to track how new technologies are being developed and how old ones are being improved. This allows for future projections for a given area of Research, Development and Innovation (RDI).

A tool called technological maturity analysis, created by NASA in 1990 and modified by The Electric Power Research Institute (EPRI), can rank existing technologies by highlighting the stage they are at. This system is called the Technology Readiness Level (TRL) system [323]. TRL has a straightforward nomenclature (TRL1, TRL2, TRL3, etc.) and enables analysis of the stages of technological development. As illustrated in Table 11, these stages advance progressively, and each one is necessary for the next.

**Table 11.** Stages of TRL development as defined by EPRI. Source: adapted from Freeman and Bhowan (2011) [323].

	TRL Level	Technology Mature Level
Demonstration	6	Integrating pilot testing into an appropriate environment
	7	Full-functioning prototype, miniature demonstration
	8	Commercial implementation and full-scale deployment
	9	Standard trade services
Development	3	Component-level proof-of-concept evaluation
	4	A laboratory setting for system validation
	5	Validation of a subsystem in an environment
Research	1	Observation of fundamental ideas, initial conception
	2	Application-based formulation

TRL analysis assesses current technologies, aids in decision-making, and provides for forecasts of time, cost, and environmental implications. Politicians and businesspeople use it as a tool today across the globe, notably in RDI.

Carbon capture, storage and use (CCSU) technologies are typically expensive technologies, hence it is crucial to conduct a technological maturity analysis to identify potential solutions that can direct RDI investments by analyzing the TRL levels the technology can achieve [324].

The International Energy Agency (2020) [325] conducted a survey of opinions on CO<sub>2</sub> capture systems based on TRL levels. Table 12 presents information from this review's four categories of technologies—adsorption, absorption, membrane separation, and chemical capture.

**Table 12.** TRL assessments for adsorption, absorption, membrane separation, and chemical capture technologies (2020) conducted by the International Energy Agency (2020) [325].

Technology Category	TRL Level	Considerations
Absorption	9	It is one of the most advanced technologies. As a result of the research time, this technology has been applied to small and large power plants, fuel converters, and industrial production facilities.
Adsorption	9	The technology is used in natural gas and ethanol processing, where CO <sub>2</sub> can be captured in large plants. There are many possible applications for this technology. One of its main advantages is its simplicity of operation.
Membrane separation	6–7	Among existing separation technologies, it is considered the most effective and is relatively new. In terms of advances, they depend on existing separation technologies. A small number of its commercial applications have been developed, while most are in demonstration or development phases.
Chemical capture	4–6	Due to the time and research intensity involved, the capture involving chemical reactions is provided in the TRL. Its magnitude is explained by the requirement for extensive pilot-scale testing given that it is still relatively new.

TRL studies have been carried out using information from technology suppliers. Data from Kearns et al. (2021) [326] were used as a comparative source.

The main drawback of the TRL scale, according to Freeman and Bhowan (2011) [323], is the absence of requirements at each stage of development to move on to the next stage. Specifically, the amount of effort needed to move from TRL1 to TRL2 and beyond is unknown.

## 9. The Use of CO<sub>2</sub> as a Feedstock for Fuel and Chemical Production

Finding alternatives to fossil fuels is viewed as being of great priority globally due to the increasing reliance on them and the depletion of resources. In general, it has become crucial to find a sustainable solution for transforming CO<sub>2</sub>, a toxic greenhouse gas that contributes to global warming, into a renewable carbon supply. CO<sub>2</sub> can be directly converted into several useful chemicals through endergonic or exergonic processes [327]. CO<sub>2</sub> can be used to meet the demands of different industries, including those for beverages, food, and chemicals [328]. Because of the financial and environmental benefits, technologies that allow CO<sub>2</sub> to be converted into value-added products are continuously being researched.

The valence state of CO<sub>2</sub> can be altered, in contrast to physical processes [329]. This procedure can be utilized to create chemical feedstock (carbonates, plastics, polymers [330]) as well as energy forms (syngas, methanol, ethane, methane). There are three types of chemical conversions that can be distinguished: thermochemical, electrochemical (photo-electrochemical [331]), and biological processes that involve enzymes [332].

There is a thermodynamic hurdle in CO<sub>2</sub> conversion due to its high stability [333]. Hydrogen is a key element in many processes that convert CO<sub>2</sub> into value-added chemicals. It should be produced from renewable energy sources to have an environmentally benign effect. Syngas (intermediate products) is produced as a byproduct of the reforming process that converts waste materials into useful fuels and chemicals. It often contains significant amounts of hydrogen and carbon monoxide, as well as small amounts of water and carbon dioxide [334]. Through the pyrolysis or gasification of biomass or natural gas conversion, respectively, reforming can occur in a solid state and with or without a gaseous state into syngas. Significant amounts of CO<sub>2</sub> released from various industrial facilities, such as fossil fuel-fired power stations, can be used as feedstock in diverse CO<sub>2</sub> recycling processes. The primary obstacle limiting the development of large-scale applications for biofuel is the availability of source feedstocks (namely, CO<sub>2</sub> and H<sub>2</sub>). Methane (CH<sub>4</sub>), methanol (CH<sub>3</sub>OH), and dimethyl ether (CH<sub>3</sub>OCH<sub>3</sub>) are just a few of the many biofuels that can be made from CO<sub>2</sub>. This path makes it possible to create a wide range of fuels for both stationary and mobile applications.

### 9.1. Production of Chemicals

A wide variety of fine compounds can be made from CO<sub>2</sub>, in addition to synthetic fuels. The most significant uses are urea alkylene carbonates (a few kt year<sup>-1</sup>), polycarbonates (4 Mt year<sup>-1</sup>), acrylic acid and acrylates (10 Mt year<sup>-1</sup>), polyurethane (≈18 Mt year<sup>-1</sup>), inorganic carbonates (≈60 Mt year<sup>-1</sup>), and urea (≈160 Mt year<sup>-1</sup>) [335]. The greatest market for the use of carbon dioxide is urea, a key fertilizer [335,336]. It is also frequently utilized as a feedstock in producing fine chemicals, polymers, medicines, and inorganic compounds such as urea resins and melamine [337,338].

Other chemicals that can be generated from CO<sub>2</sub> capture are also beneficial in various sectors, including lubricants, polymers, agrochemicals, pharmaceuticals, coatings, and catalytic processes. These chemical classes include organic carbonates such as diphenyl carbonate (DPC), dimethyl carbonate (DMC), diallyl carbonate (DAC) and diethyl carbonate (DEC), as well as cyclic carbonates such as styrene carbonate (SC), cyclohexene carbonate (CC), propylene carbonate (PC) and ethylene carbonate (EC), and even polycarbonates such as bisphenol polycarbonate (BPA-PC) and poly(propylene carbonate) [339,340].

This process faces difficulties since it requires a large amount of catalyst inventory and operates at high temperatures and pressures. Additionally, this procedure faces additional challenges in separating the catalyst from the products [339,340]. Commercially accessible Al-based catalysts are frequently utilized, but they are not eco-friendly in manufacturing polycarbonates from the reaction of CO<sub>2</sub> with epoxides. In this context, an alternate method with a lot of potential for producing polycarbonates from CO<sub>2</sub> and olefins is the oxidative carboxylation approach [341]. Another chemical made from the interaction of CO<sub>2</sub> and cyclic amines, such as azetidines and aziridines, or the N-analogues of epoxides, is polyurethane [341,342].

Formic acid is another significant chemical that can be created using CO<sub>2</sub>. The hydrogenation of CO<sub>2</sub> into formic acid has lately attracted some study interest because of the benign reaction conditions, the absence of byproduct formation, its ability to store hydrogen in liquid form, and the ease of formic acid's decomposition into hydrogen and CO<sub>2</sub> [337,343].

The biological use of CO<sub>2</sub> provides an additional route for producing biodiesel and numerous commodity chemicals produced from biomass (used in food, silage, biogas, and fertilizer) [344]. This method has the benefits of a faster development rate, a shorter growth cycle, no competition with other plants for land, and the creation of various valuable by-products. To remove contaminants such as heavy metals, NO<sub>x</sub>, and SO<sub>x</sub> that are harmful to microalgae growth, the captured CO<sub>2</sub> should be filtered before being fed into a photobioreactor [345].

Using CO<sub>2</sub> as a technological fluid without converting it into chemicals has found applications outside of EOR in a variety of industries, such as the air conditioning (as a coolant), food-preservation, dry-washing, solvent, and beverage industries [335,343,346]. Generally, EOR consumes 50 Mt year<sup>-1</sup> CO<sub>2</sub>, compared to the 8 Mt year<sup>-1</sup> CO<sub>2</sub> used by the beverage and food industries [347].

Overall, the proposed laboratory-scale solutions are still far from being commercialized for industrial use, even though there is a huge market for converting captured CO<sub>2</sub> into chemicals and fuels. This is partly due to the exorbitant manufacturing costs of the materials under investigation thus far, which are also not chemically stable, and in part to the generally low CO<sub>2</sub> conversion rates and total yields of the primary products. As a result, they do not satisfy the criteria for widespread deployment. Furthermore, knowledge of the mechanisms underlying the chemical reactions involved in the transformations of CO<sub>2</sub> is still in its infancy. Evaluations of the requirements and factors of the procedure have also been neglected in this discipline.

### 9.2. Production of Fuels

The most effective method of utilizing CO<sub>2</sub> is its conversion to fuels. Alkanes, methane, methanol, syngas, and other compounds can be produced from the captured CO<sub>2</sub>. In addition to transportation, power plants, and fuel cells, the fuel produced can be used in many different

industries [335]. The number of methods for using CO<sub>2</sub> to produce fuels is enormous. Since CO<sub>2</sub> is a molecule with a steady thermodynamic state, its use requires a lot of heat and catalysts to achieve high fuel yields [348]. The hydrogenation and dry reforming of methane (DRM) are the two main processes for producing fuel from captured CO<sub>2</sub> [349].

The possibility of creating fuel, storing H<sub>2</sub>, recycling CO<sub>2</sub>, and resolving the difficulty of storing electric energy makes CO<sub>2</sub> hydrogenation a very attractive method of using CO<sub>2</sub> [343]. DRM is also regarded as one of the most significant routes for the Fischer–Tropsch (FT) process' production of methanol and numerous other liquid fuels [341,350,351]. The source of hydrogen from fossil fuels seems to be a concern in the hydrogenation of CO<sub>2</sub> into methane [352], methanol [353], carbon monoxide [352], and formic acid [354], as this can result in a rise in the atmospheric emissions of CO<sub>2</sub>.

However, renewable energy sources, such as solar, wind, and biomass, can be used as a substitute for fossil fuels to further reduce CO<sub>2</sub> emissions during hydrogenation [355]. Recently, the “e-gas” created by the German Audi Motor Company using CO<sub>2</sub> hydrogenation produced 1000 Mt year<sup>−1</sup> of methane [356].

The volumetric gas density of methane is low, which makes it an unsuitable fuel for use in vehicles. Additionally, its global warming potential (GWP) is 30 [348]. Methane is readily available, thus producing more will not be profitable for CO<sub>2</sub> capture (methane is plentiful in landfill gas, coal gas, shale gas, and natural gas). It appears that a preferable mechanism is CO<sub>2</sub> hydrogenation to methanol [357]. However, it is extremely difficult to activate C–H bonds with the currently used (mostly Cu-based) catalysts for the manufacture of methanol, and the catalysts that have been tried so far are not particularly profitable [358–360].

Methanol has numerous uses in organic solvents, combustion engines, plastics, and paints [348]; nevertheless, only 0.1% of CO<sub>2</sub> emissions are reduced by its synthesis [361]. One of the most significant methods for using CO<sub>2</sub> is the reverse water–gas shift (RWGS) reaction, which converts CO<sub>2</sub> into CO because CO is a starting material for the FT reaction, which produces methanol and hydrocarbon fuels [359]. Despite this, the RWGS reaction's endothermic nature and the low conversion at moderate temperatures provide the two biggest challenges to the implementation of large-scale methanol production from CO<sub>2</sub> using the FT process. Another major obstacle is the development of active catalysts that can speed up the reaction rate and enhance the yield.

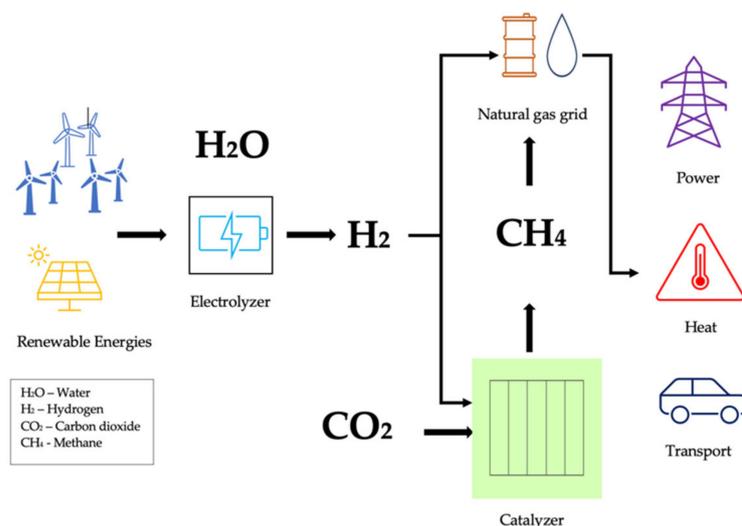
DRM has recently gained a lot of study interest in terms of utilizing CO<sub>2</sub> to produce syngas [340,362,363]. According to Ofélia de Queiroz et al. [364], DRM often produces syngas with a greater purity than partial oxidation and steam reforming. Additionally, the DRM process only produces 2% of unreacted methane, which is less than steam reforming, allowing it to be used at remote natural gas locations to produce liquid fuels, which are more transport-friendly than gaseous fuels [341]. The DRM reaction has been thoroughly examined for Rh, Ir, Ru, Ni–Co, and Ni supported on lanthanum oxide, alumina, and silica [341]. Despite major advancements in the design of catalysts with high activity and ideal stability for DRM, finding a good catalyst for this reaction still poses a significant difficulty, especially at high operation temperatures when deactivation by coke formation is inevitable (>700 °C) [365–369].

Another appealing method that can lessen the formation of coke and maintain the stability of catalysts at high temperatures is the oxidative dehydrogenation of light alkanes to alkenes (ODA), which uses CO<sub>2</sub> as a soft oxidant instead of the usual dehydrogenation oxidant, O<sub>2</sub> [370–374]. Additionally, by eliminating hydrogen via the RWGS process, CO<sub>2</sub> enhances the light alkanes' oxidative dehydrogenation equilibrium conversion [375]. However, it is important to monitor the temperature since too much heat might lead to the olefins overoxidizing, which produces carbon oxides and reduces selectivity [376]. The redox cycle and active oxygen species are both formed by CO<sub>2</sub>. The active site type, the reduction capability of the metal, and the supporting material all affect how CO<sub>2</sub> functions in the ODA and how this reaction works [377]. The catalysts studied thus far have little stability, while having a high initial activity.

The aforementioned discussion makes it clear that creating novel catalysts with high catalytic activity under a variety of reaction conditions, coke resistance, and long-term structural and chemical stability is the main obstacle to using captured CO<sub>2</sub> as a feedstock for the creation of synthetic fuels.

#### 9.2.1. Production of Methane (CH<sub>4</sub>) Based on CO<sub>2</sub> (Methanation), Challenges and Prospects

Chemical feedstocks such as CO<sub>2</sub> can be used to transform renewable energy. One promising approach to combating CO<sub>2</sub>-induced climate change is CO<sub>2</sub> methanation for value-added products, where water electrolysis is a potential energy storage technique for generating H<sub>2</sub> with renewable energy and would contribute to the creation of a carbon-based cycle that is sustainable. Due to their inherent intermittency, renewable energy sources are currently limited by the need for scalable storage methods [378]. Therefore, the most practical and easiest method of storing significant volumes of intermittent energy generated from renewable sources for extended periods is the generation of synthetic natural gas or liquid fuels. Power to gas (PtG) is one concept that has gained a lot of attention over the years (Figure 8) [379]. In this method, CO<sub>2</sub> interacts with H<sub>2</sub>, which is created by water electrolysis using renewable solar or wind energy to generate CH<sub>4</sub> as a substitute for natural gas. In Copenhagen, a commercial-scale PtG plant with 1.0 M2 of capacity was successfully operating in 2016, exploiting the change of the energy system towards a sustainable system [380]. Five projects, using CO<sub>2</sub> methanation at a commercial or pilot plant size with capacities varying from 25 kW to 6300 kW, were implemented in Germany between 2009 and 2013 [381]. In fact, natural gas, or methane, is Germany's main source of heat and a significant contributor to natural gas supplies. Due to their robust dynamic properties, natural gas power plants now generate a larger portion of Germany's electricity than the country's present coal-fired power plants do [382]. Because it has a higher H:C ratio than its conventional counterpart, its use in automobiles instead of gasoline minimizes CO<sub>2</sub> emissions. The following reactions in Table 13 take place in the methanation reactor [383]:



**Figure 8.** Schematic illustration of “power to gas” (PtG) technology (adapted from Younas [380]).

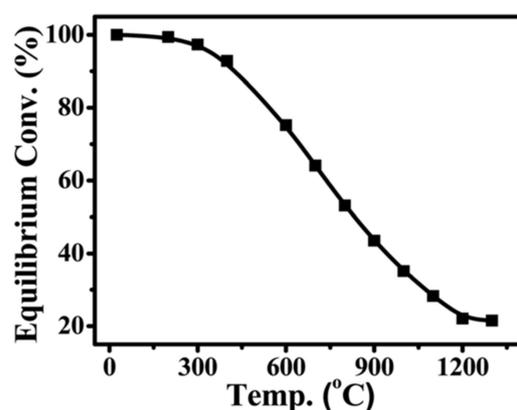
**Table 13.** CO<sub>2</sub> methanation: main reactions and side reactions [384].

Reaction Equation	Response Type	$\Delta H_R^{298K}/\text{kJ}\cdot\text{mol}^{-1}$
$\text{CH}_4 \rightleftharpoons \text{C} + 2\text{H}_2$	Methane pyrolysis	−75
$\text{CO}_2 + 2\text{H}_2 \rightleftharpoons \text{C} + 2\text{H}_2\text{O}$	CO <sub>2</sub> reduction	−90
$\text{CO} + \text{H}_2 \rightleftharpoons \text{C} + \text{H}_2\text{O}$	CO reduction	−131
$2\text{CO} \rightleftharpoons \text{C} + \text{CO}_2$	Boudouard reaction	−172
$\text{CO}_2 + 4\text{H}_2 \rightleftharpoons \text{CH}_4 + 2\text{H}_2\text{O}$	CO <sub>2</sub> methanation	−165
$2\text{CO} + 2\text{H}_2 \rightleftharpoons \text{CH}_4 + \text{CO}_2$	reverse dry reforming	−247
$\text{CO} + 3\text{H}_2 \rightleftharpoons \text{CH}_4 + \text{H}_2\text{O}$	CO methanation	−206
$\text{CO}_2 + \text{H}_2 \rightleftharpoons \text{CO} + \text{H}_2\text{O}$	reverse water gas shift	41

It is difficult to apply CO<sub>2</sub> because of its inertness, which prevents it from being converted into chemicals with added value. The use of specific catalysts can, however, help to resolve this problem [385]. According to Park, Kwak [386], utilizing a double-layered TiO<sub>2</sub>/Cu-TiO<sub>2</sub> catalyst instead of a typical TiO<sub>2</sub> catalyst resulted in a two-fold improvement in the yield of CH<sub>4</sub> production from CO<sub>2</sub> (film catalyst). To clean syngas in ammonia factories, carbon oxides were also hydrogenated to methane. Additionally, this might result in carbon-neutral fuel (methane) [327]. The conversion of CO<sub>2</sub> to methane can also occur biologically, for example, using methanogens. Methane-producing organisms are produced via the anoxic enrichment of waste-activated sludge (methanogens). The effectiveness of methane generation was improved by almost 70-fold when the organism's activated cultures were used [387].

Due to the increasing demand for storing renewable energy and mitigating global warming, CO<sub>2</sub> methanation has gained renewed attention due to the French chemist Paul Sabatier's discovery in 1902 [388]. Storing energy from renewable resources such as wind and solar, the efficient conversion of biogas to biomethane, and the conversion of CO<sub>2</sub> into chemical feedstocks and fuels are all made possible by the Sabatier reaction [389,390].

Figure 9 illustrates the exothermic nature of CO<sub>2</sub> methanation, which has a high equilibrium conversion between 25 and 400 °C [391,392]. By using the right catalysts, CO<sub>2</sub> methanation can achieve 99% CH<sub>4</sub> selectivity, avoid product separation, and circumvent the challenges of dispersed product distribution. Because of this thermodynamic characteristic, CO<sub>2</sub> methanation is more important in terms of energy effectiveness and economic viability.



**Figure 9.** A plot of equilibrium CO<sub>2</sub> conversion in methanation at various temperatures (based on literature data) [393,394].

### 9.2.2. Challenges

In CO<sub>2</sub> methanation, deactivating metal catalysts is a major challenge. There are two different ways that methanation catalysts can be deactivated: (a) chemically and (b) physically.

In contrast to Co/Al<sub>2</sub>O<sub>3</sub>, which deactivated quickly in the same amount of time, the Co/ZrO<sub>2</sub> catalysts showed a greater CO<sub>2</sub> methanation activity and practically consistent

performance even after 300 h on stream. Through thermogravimetric analysis and hydrothermal ( $\text{H}_2\text{O}$ ) treatment verification experiments, the deactivation of the  $\text{Co}/\text{Al}_2\text{O}_3$  catalyst was further researched. An excessive amount of  $\text{CoAl}_2\text{O}_4$  was produced because of the addition of extra  $\text{H}_2\text{O}$  to the reaction system, which hastened the deactivation of the  $\text{Co}/\text{Al}_2\text{O}_3$  catalyst. As a result, the product  $\text{H}_2\text{O}$  encourages the production of the inactive phase  $\text{CoAl}_2\text{O}_4$ , which causes  $\text{Co}/\text{Al}_2\text{O}_3$  catalysts to deactivate quickly. One of the causes of deactivation is the deposition of carbon, but the primary cause is the creation of the  $\text{CoAl}_2\text{O}_4$  spinel structure in the inactive phase.

Active metal sintering and carbon deposition are the main causes of physical deactivation. Ni/YSZ catalysts were synthesized by Kesavana et al. [395] using several techniques. Thin coatings of carbon are generated on NiO particles with a spherical shape, and graphitic laminations are formed on NiO particles that are exposed to surfaces on the Ni/YSZ catalyst created by the impregnation procedure. When NiO/ $\text{Ni}^{2+}$  oxidation with a high  $\text{CO}_2:\text{H}_2$  ratio was used, the Ni/YSZ(EDTA) catalyst displayed outstanding stability, and operant XAS demonstrated that it was not deactivated. Increasing the  $\text{H}_2/\text{CO}_2$  ratio or adding steam can prevent carbon from depositing on the catalyst because hydrogen reacts with the carbon deposits to keep them from deactivating the catalyst.

Increased metal dispersion via strong metal–support contact, the addition of catalyst promoters, the development of novel synthesis techniques, and other typical techniques are used to reduce metal sintering [396]. When the active Co particles are separated by graphite-like carbon, metal sintering is averted effectively. In a study by Li et al. [397], NiO–MgO@ $\text{SiO}_2$  catalysts were successfully prepared and maintained their activity after 100 h on stream. Therefore, particles with the right size are beneficial to  $\text{CH}_4$  formation.

### 9.2.3. Prospects

Increasing atmospheric  $\text{CO}_2$  concentrations are believed to be one of the primary drivers of climate change. Although  $\text{CO}_2$  capture technologies are quite advanced, using captured  $\text{CO}_2$  remains a significant difficulty that will require extensive future research. In the short to medium term, the creation of new life cycle assessments (LCA), techno-economic tools, and benchmark assessments will make it possible to evaluate  $\text{CO}_2$  conversion pathways consistently and transparently. Current barriers to furthering  $\text{CO}_2$  conversion include energy and water usage, the need for costly catalysts, and problems with gas infrastructure. Cost-effective  $\text{CO}_2$  conversion techniques are especially needed, and their pilot-scale testing is essential for their commercial application. A thorough introduction to the various  $\text{CO}_2$  conversion processes aids in comprehension of the mechanisms and the selection of suitable methods for using captured  $\text{CO}_2$ . It is preferable to use the prospective integrated approaches to close the current gaps for widespread industrial applications. Additionally, it is advantageous to demonstrate research and development (R&D) initiatives for  $\text{CO}_2$  conversion and analyze  $\text{CO}_2$  utilization markets to determine the likelihood of the full-scale implementation of these technologies.

Economic viability, whether it is attained through technical progress or legislative changes, is crucial to promoting the development of the industrial feasibility of  $\text{CO}_2$  conversion technology. Because of this, future key research objectives should be concentrated on  $\text{CO}_2$  conversion and CCS legislation, policy, and assessments, as well as the integration of  $\text{CO}_2$  use with other strategies to minimize costs and energy consumption, especially at a larger scale. Public education and publicity on the capture, sequestration and methanation of  $\text{CO}_2$  (CSCM) should be highlighted, and global collaborations need to be further strengthened in the meantime, to increase the general public's awareness of the environmental repercussions. Additionally, it should be highlighted that CCS is necessary to achieve our climate goals, and that  $\text{CO}_2$  is not a substitute for it. To keep the rise in the global mean temperature below  $1.5^\circ\text{C}$ , governments should strengthen their commitment to CSCM and play a critical role in supporting its deployment (e.g., through laws, financial options, and tax incentives). The encouragement of private sector funding for larger-scale demonstration programs and the commercialization of  $\text{CO}_2$  technologies is another successful approach.

The market for CO<sub>2</sub>-based products may soon be impacted by regulations and policies as CO<sub>2</sub> becomes a resource that diverse sectors of the global economy want.

The following are general directions for future CO<sub>2</sub> methanation research:

1. To increase the H/C ratio of catalyst surface and facilitate C–C coupling for high-value-added products;
2. To promote CO<sub>2</sub> adsorption and activation by enhancing the oxygen vacancies and support basicity;
3. To investigate potential novel catalytic materials and enhance the stability of the catalyst;
4. To develop more effective catalysts for CO<sub>2</sub> hydrogenation at low temperatures and with little energy consumption;
5. To analyze the process intensification and optimization of CO<sub>2</sub> conversion technologies, which are crucial to understanding how various operating parameters interact, improve process effectiveness, and reduce costs.

## 10. Conclusions

The above sections provide a comprehensive overview of different CO<sub>2</sub> separation and capture technologies. The ideal separation techniques to lower CO<sub>2</sub> emissions are also highlighted, along with their benefits and drawbacks, in this review paper. Additionally, dry solid sorbents, with a focus on emerging adsorbents, were addressed. A discussion of the challenges involved in converting CO<sub>2</sub> into value-added products was also conducted, and future perspectives were laid out.

This study leads to the conclusion that the CO<sub>2</sub> capture and separation (CCS) problem cannot be solved by a single method. Multiple technologies covered in this paper would need to be incorporated to solve this complex problem. To solve the technological problems in the CCS, it is necessary to scale-up innovative technologies from a laboratory to an industrial scale. This requires further research on new physical and chemical sorbents and procedures with improved cost-effectiveness and efficiency.

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