# Solid Adsorbents for $CO_2$ Capture

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Carbon capture and storage (CCS) is considered to be a promising technology in reducing atmospheric CO2 concentration. Among the CO2 capture technologies, adsorption has grabbed significant attention owing to its advantageous characteristics discovered in recent years. Solid adsorbents have emerged as one of the most versatile CO2 adsorbents.

porous carbon | amine functionalization | physisorption | chemisorption | CO2 capture | activated carbon | Greenhouse effect

## 1. Introduction

### 1.1. Physical and Chemical Properties of $CO_2$

Carbon dioxide ($CO_2$) is a triatomic gas under ambient conditions [1], which is abundant, non-toxic, recyclable, and economical [2]. Moreover, $CO_2$ sublimates from solid-state to gas at −78 °C under atmospheric pressure and is comparatively inert. As a commonly known fact, $CO_2$ gas that naturally occurs in the Earth's atmosphere is of paramount importance to photosynthesis [1]. From an economic point of view, $CO_2$ can be converted into high-value chemical products such as urea, carbonates, and acrylates [3] through catalytic conversion, mineralization, photochemical, or electrochemical reactions, and supercritical $CO_2$ can be also utilized in various industrial fields, including food beverages, refrigerants, transportation fuels, fire extinguishers, polymer synthesis, medical, and exploitation of heavy oil. Solid-state $CO_2$ can be used in artificial rainfall and concrete production [4][5].

### 1.2. Trend of Atmospheric $CO_2$ Concentration and Potential $CO_2$ Emissions Sources

Although the natural carbon cycle controls the $CO_2$ concentration level in the Earth's atmosphere [1], due to both anthropogenic activities and natural emissions, the current atmospheric $CO_2$ concentration reached around 416.5 ppm in mid-2020 [6], which is ~40% greater than the beginning of the industrial revolution (280 ppm) in 1750 [7][8][9], with an average growth rate of 2 ppm per year [9][10]. In other words, the global emission of $CO_2$ was estimated to be more than 36 MT in 2017, which is 18-fold greater than compared to the 1800s [11]. Although it is a consensus that the amount of atmospheric $CO_2$ should not exceed 350 ppm [12], according to the predictions by the International Panel on Climate Change (IPCC), it is expected to reach up to 570 ppm by 2100 [12][13][14]. It is identified that the main causes for the tremendous increase in such atmospheric $CO_2$ concentration are mainly associated with various anthropogenic activities, including vehicular emissions, fossil-fuel power plants, deforestation, chemical processes [15], and waste treatment [16], which have been growing steadily due to rapid industrialization and urban development [15][17]. The natural emission sources, including soil degradation processes and volcanic activities, are also responsible for supplying atmospheric $CO_2$ to some extent [18].

## 1.3. Significant Outcomes Owing to the Trend of Increasing $CO_2$ Emissions

Unfortunately, the non-controllable anthropogenic activities have negatively affected human beings [19] and the entire ecosystem [3][6] by releasing greenhouse gases, including $CO_2$, into the atmosphere. Among the greenhouse gases, $CO_2$ is considered as one of the primary sources, contributing to roughly 64% of the total greenhouse effect [14][20]. The progressive increase in atmospheric $CO_2$ concentration is responsible for climate change, which might adversely impact the global environmental processes, such as the long-term rise in global temperatures, changes in rainfall patterns, rising sea levels [21][22], ocean acidification [23], species extinction, melting of polar ice [9], shrinkage of snow covers [24], and severe weather events, ranging from flash floods [25], hurricanes, freezing winters, severe droughts [22], heat waves [26], urban smog [17], and cold streaks [27]. According to the predictions made by IPCC, the rise in sea level of 3.8 m [14][28] and rise in mean global temperature by 3.7 °C [29][30] are expected by 2100 [24]. Besides, the increasing trend of $CO_2$ in the air might cause various air-borne diseases, which will increase the risk of health complications [31]. The economic loss due to climate change is expected to be 5–20% of the global domestic production [12][28]. Therefore, extensive research projects are currently underway to reduce and control $CO_2$ emissions from power plants, industries, and transportation [32].

## 1.4. Approaches to Reduce Atmospheric $CO_2$ Concentration

Three feasible strategies to reduce $CO_2$ emissions are exhibited by the modified Kaya identity as expressed in equation (1) [28]. They are namely, (i) improving the energy efficiency of coal-fired plants [33][34], (ii) change of the fossil fuels to renewable and carbon-free energy resources [35], and (iii) utilization of carbon capture and storage (CCS) technologies [28][36][37].

$$CD = P\frac{GDP}{P}\frac{E}{GDP}\frac{C}{E} - S_{CO_2}$$

where CD: $CO_2$ emissions, P: Population, GDP: economic development in gross domestic production, E: energy production, C: carbon-based fuels used for energy production, and $S_{CO2}$: $CO_2$ sinks [28].

Apart from the above-mentioned three strategies, enhancing partial pressure in exhaust gas [36], geoengineering approaches including afforestation and reforestation [38], flue gas separation, and carbon mineralization [39] can also be considered. Among the different $CO_2$ mitigation options, IPCC has suggested CCS as a promising technology for achieving a 19% reduction of global $CO_2$ emissions by 2050 [34]. CCS can reduce $CO_2$ emissions (typically 85–90%) from significant stationary point sources such as power plants, cement kilns, and NG wells [40][41]. Nevertheless, CCS is considered a mid-term solution in reducing global warming, climate change, and simultaneously allowing humans to continue using fossil fuels until a renewable and clean energy source is discovered to replace them [34]. CCS is comprised of three significant steps, namely, (i) capture of emitted $CO_2$

from power plants and industrial processing without releasing them into the atmosphere, (ii) transportation of the captured and compressed $CO_2$, and (iii) underground storage of the captured $CO_2$ [26][42][43]. However, the process of $CO_2$ capture, which accounts for 70–80% of the total cost, has proven to be the major barrier for the deployment of CCS [40][44]. Interestingly, in recent years, carbon capture storage and utilization (CCSU) has grabbed significant attention compared to CCS owing to the convertibility of the captured $CO_2$ into commercial products [45][46]. The success of CCS and CCSU technologies are associated with the $CO_2$ adsorption efficiency, ease of handling, manufacturing cost, and renderability of the associated materials [22].

## 1.5. $C_{O2}$ Emission Sources

The $CO_2$ emission sources are the primary candidates for potential applications of CCS or CCSU technologies. Therefore, from a community and industrial point of view, $CO_2$ capture from typical gas streams, including flue gas, biogas, flare gas, syngas, and ambient air, has grabbed significant interest [47]. **Table 1** depicts the summary of the compositions of different gas streams.

**Table 1.** Compositions of different gas streams which act as potential $CO_2$ capture opportunities (Reprinted with permission from ref. [47][48]).

| Component | Cement Rotary Kiln | Dry Atmospheric Air | Biogas Generated from Waste Water Treatment Plant Sludge | Natural Gas Fired Flue Gas | Coal-Fired Flue Gas |
|---|---|---|---|---|---|
| $N_2$ | 59 vol % | 70 vol % | 0–1 vol % | 73–80 vol % | 70–80 vol % |
| $CO_2$ | 19 vol % | 410 ppm | 19–33 vol % | 3–8 vol % | 11–15 vol % |
| $H_2O$ | 13 vol % | - | - | 7–14.6 vol % | 5–12 vol % |
| $O_2$ | 7 vol % | 21 vol % | <0.5 vol % | 4.5–15 vol % | 3–6 vol % |
| $SO_2$ | 5–1200 ppm | - | - | <10 ppm | 200–4000 ppm |
| $SO_3$ | - | - | - | - | 0–20 ppm |
| $NO_X$ | 100–1500 ppm | - | - | 50–70 ppm | 200–800 ppm |
| CO | - | - | - | - | 50–100 ppm |
| $H_2$ | - | 0.5 vol % | - | 5–300 ppm | 5–20 g/m$^3$ |
| Particulate | - | - | - | - | - |

| Component | Cement Rotary Kiln | Dry Atmospheric Air | Biogas Generated from Waste Water Treatment Plant Sludge | Natural Gas Fired Flue Gas | Coal-Fired Flue Gas |
|---|---|---|---|---|---|
| matter | | | | | |
| $H_2S$ | - | - | 100–4000 ppm | - | - |
| Ar | - | 0.9 vol % | - | - | - |
| Xe | - | 0.1 vol % | - | - | - |
| Ne | - | 18 ppm | - | - | - |
| He | - | 5.2 ppm | - | - | - |
| $CH_4$ | - | 1.6 vol % | 60–75 vol % | - | - |
| Kr | - | 1.1 vol % | - | - | - |
| $N_2O$ | - | 0.3 vol % | - | - | - |

## 1.6. $CO_2$ Capture Technologies

**Table 2** depicts the comparison of the leading carbon capture technologies. According to **Table 2**, carbon capture from power plants in industries can be classified as (i) pre-combustion capture, (ii) oxy-fuel combustion, and (iii) post-combustion capture [49] depending on the combustion method and composition of the gas stream [50]. The working conditions such as pressure and temperature differ for each technique [51]. The main factors impacting $CO_2$ capture efficiency are the gas composition, gas stream temperature and energy penalty associated with regeneration [28].

**Table 2.** Comparison of the three main carbon capture technologies.

| CO2 Capture Technology | Advantages | Disadvantages |
|---|---|---|
| Pre-combustion capture | • The concentration of $CO_2$ produced within these processes range from ~15–60% which makes it easy to capture [51] | • When applying to new power plants, the technology is not yet commercialized and requires a high capital investment due to major alternatives to be done into boiler and flue gas systems [28]<br>• Process of gasification and water gas shift reactions are expensive and quite challenging [51]<br>• High energy penalty associated with regeneration of chemical solvents [52] |

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| CO2 Capture Technology | Advantages | Disadvantages |
|---|---|---|
| Oxy-fuel combustion | • Avoids the requirement of chemicals or other means of $CO_2$ separation from flue gas [52] | • Large energy penalty requirement for providing pure oxygen [53]<br>• Absence of complete preparation methods [54]<br>• Pure oxygen is expensive [52]<br>• Limited knowledge regarding the technology [53]<br>• Environmental impacts associated are higher due to energy intensive air separation process [52] |
| Post-combustion capture | • Readily applicable for large-scale in newly built and existing power plants without upgrading and reconstruction [55]<br>• Repairing does not discontinue the procedure of the entire power plant and it can be regulated or managed easily [56]<br>• Shorter time required for creation [57] | • Requirement of huge energy supplies for sorbent regeneration [53]<br>• Requires the separation of impurities from captured $CO_2$ [58]<br>• $CO_2$ in the flue gas is diluted with a concentration ranging from 10–15% which requires high recovery and capital costs and 25–35% additional energy for plant operation [28] |


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## 2. Solid Adsorbents for $CO_2$ Capture

### 2.1. Adsorption Process of $CO_2$

Adsorption is a surface phenomenon that highly depends on surface properties and functionalities [50]. Adsorption of $CO_2$ onto a material occurs through different types of interactions between the gas molecules and the adsorbent. Adsorption can be classified as (i) physisorption or (ii) chemisorption [59]. $CO_2$ adsorption is an exothermic process as reported elsewhere [60][61]. **Figure 1** presents the schematic of the two adsorption processes, while **Table 3** tabulates the differences between physisorption and chemisorption.

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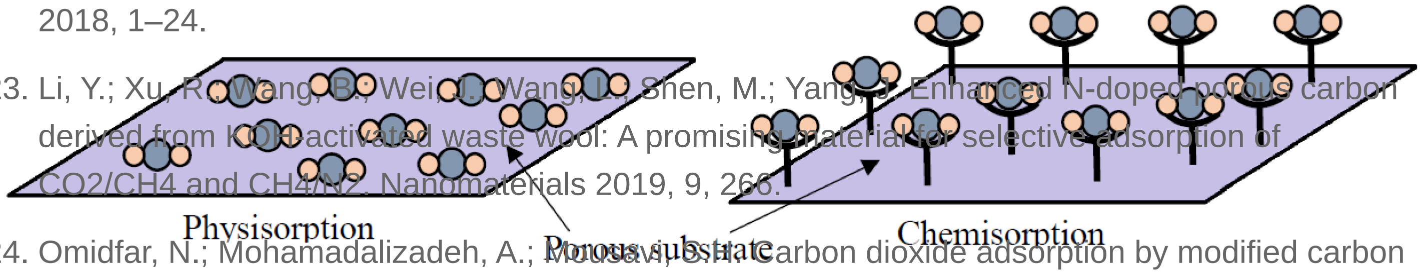


**Figure 1.** Schematic of the interactions between gas molecules and the adsorbent surface during physisorption and chemisorption (Reprinted with permission from ref. [62]).

**Table 3.** Comparison of the $CO_2$ physisorption and chemisorption processes.

| Process | Advantages | Disadvantages |
|---|---|---|
| Physisorption | • More appropriate for high pressure applications [63]<br>• Adsorbent is easily regenerated, and low energy is required for desorption [10]<br>• Relatively stable even past 200 °C [10]<br>• Low cost for adsorbent preparation [64] | • $CO_2$ capture capacity decreases with increasing temperature [15][65]<br>• Low $CO_2$ uptake at low pressures [47]<br>• Low $CO_2$ selectivity for combustion flue gas streams [42]<br>• Adsorption capacity decreases in the presence of water [62] |
| Chemisorption | • High selectivity towards $CO_2$ due to strong interactions between basic species on the adsorbent surface and the acidic $CO_2$ molecule [42][66]<br>• High adsorption capacity at low $CO_2$ partial pressures such as in the ambient air [42][67][68]<br>• Enhanced adsorption capacity in the presence of water [64][69]<br>• Comparatively higher mechanical stability [45] | • Slower than the physisorption process [70]<br>• Functionalization of porous materials with amine groups decreases the $CO_2$ capture capacity due to pore blockage [66][71]<br>• High energy requirement for regeneration of the adsorbent [72]<br>• Low cyclic stability due to amine degradation [66]. Higher cost associated with adsorbent synthesis [64]<br>• Chemisorbents can permanently bind to gases such as $SO_2$ to decrease the capacity of active sites for $CO_2$ capture [72] |

| Process | Advantages | Disadvantages |
|---|---|---|
| | | • Grafted amines volatilize and degrade above 120 °C due to instability at higher temperatures [72]<br>• A corrosive environment could be produced during the regeneration of spent adsorbent due to the presence of amine groups [59] |

## 2.2. Different Regeneration Strategies

The attached $CO_2$ molecules onto the adsorbent surface could be regenerated through the (i) pressure swing adsorption (PSA), (ii) temperature swing adsorption (TSA), (iii) vacuum swing adsorption (VSA), (iv) pressure and vacuum swing adsorption (PVSA), and (v) electric swing adsorption (ESA) processes [26][28][73]. **Table 5** shows the advantages and disadvantages of different regeneration strategies. The regeneration method depends on the chemical and structural properties of a given adsorbent [69].

**Table 4.** Comparison of different regeneration strategies.

| Regeneration Strategy | Advantages | Disadvantages |
|---|---|---|
| Temperature swing adsorption (TSA) | • Simple in operation [69]<br>• Can use low-grade heat from power plants [74] | • Long heating and cooling time periods [69]<br>• Longer desorption time than PSA [28]<br>• Higher energy requirement than PSA [28]<br>• Rapid adsorbent deactivation due to coking at higher temperatures [28] |
| Pressure swing adsorption (PSA) | • Lower energy requirement than TSA [75]<br>• Easy operation [75]<br>• Low capital investment than TSA and VSA [75] | • Compression of the flue gas streams [69]<br>• Dilute gas streams may result in intense energy consumptions during PSA [72] |

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| Regeneration Strategy | Advantages | Disadvantages |
|---|---|---|
| | • Applicability over a wide range of temperatures and pressures [76] | |
| Electric swing adsorption (ESA) | • More economical than TSA and PSA [28]<br>• Independent purge gas flow [69]<br>• Fast heating and cooling rates [69]<br>• Low energy consumption [69] | • Further improvements are required before commercialization [28]<br>• The adsorbents should have good electrical conductivity [69] |
| Vacuum swing adsorption (VSA) | • Applicability in large point sources [69] | • Energy intensive operation |

## 2.3. Criteria for Selecting $CO_2$ Adsorbents

When synthesizing and selecting an effective $CO_2$ adsorbent, the material should be economical and operational simultaneously [74]. Therefore, a prospective $CO_2$ adsorbent should satisfy the following criteria (**Table 5**): (i) $CO_2$ adsorption capacity: The adsorption capacity plays a very decisive role in determining the amount of adsorbent to be inserted into the adsorption column to attain the desired performance [77][78]. (ii) Regenerability: The adsorbent should be fully regenerable and require relatively mild conditions for complete regeneration [78]. (iii) $CO_2$ selectivity: The adsorbent should display substantially high selectivity for $CO_2$ in the co-presence of other species (e.g., N2, methane (CH4), sulfur dioxide ($SO_2$), hydrogen sulfide (H2S), and moisture) [74][79][80], (iv) Adsorption/desorption kinetics: A rapid adsorption/desorption is required for swing adsorption to decrease the cycle time [73][74]; (v) Thermal, chemical, and mechanical stability: During the cyclic regeneration process, the microstructure and morphology of the adsorbent should be retained. Moreover, the adsorbent should withstand harsh operating conditions, including vibration, high temperatures, pressures, and flow rates. Additionally, the amine-functionalized adsorbents should be resistant against oxidizing agents and contaminants such as sulfur oxides (SOX), nitrogen oxides (NOX), water vapor, and heavy metals [11][81], and (vi) Adsorbent cost: The adsorbent should be synthesized using cheap raw materials while adopting a cost-effective and energy-saving synthesis routes [62].

**Table 5.** Threshold values of criteria for selecting an effective $CO_2$ adsorbent (Reprinted with permission from refs. [74][77]).

| Parameter | Requirement |
|---|---|
| CO2 adsorption capacity | 3–4 mmol/g |


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| Parameter | Requirement |
|---|---|
| Regenerability | >1000 cycles |
| CO2 gas selectivity over other gases | >100 |
| Adsorption/desorption kinetics | >1 mmol/g.min |
| Adsorbent cost | $5–15/kg sorbent |

## 2.4. Different Adsorbents for $CO_2$ Capture

Numerous studies on $CO_2$ capture conducted in academic and industrial settings have developed promising adsorbents possessing the requirements demonstrated in **Table 5** [55]. A variety of adsorbents have been discovered and synthesized, including MOFs, zeolites, activated carbons, zeolite imidazolate frameworks (ZIFs), grafted and impregnated polyamines [44], activated alumina, carbonized porous aromatic frameworks (PAFs), covalent organic frameworks (COFs) [82][83], porous organic polymers (POPs) [33], mesoporous silica, carbon nanotubes [84], metal oxides, ionic liquids [85], phosphates [26], and molecular sieves [5].

## 2.5. Importance of Carbon-Based Adsorbents for Effective $CO_2$ Capture

Of the previously mentioned $CO_2$ adsorbents, though zeolites and well-ordered frameworks exhibit high $CO_2$ adsorption capacities at relatively lower pressures [39], the $CO_2$ adsorption performance gradually decreases in the co-presence of moisture [34][86]. Similarly, molecular sieves and silica gel also demonstrate decreased $CO_2$ adsorption performance in the co-presence of moisture [5]. Additionally, the usage of MOFs has been severely limited due to structural collapse upon vacuum treatments [34], contact with acid gases, thermal regeneration [84], and their complex and expensive synthesis procedures [86]. The ionic liquids are also unfavorable for practical applications due to their relatively high operational costs and high viscosity, leading to corrosion-related problems [87].

On the other hand, the application of carbon materials in the day-to-day lives of human beings can be traced back to more than 5000 years when the early humans discovered charcoal formed through the incomplete combustion of wood. Interestingly, many carbon materials have been discovered, such as graphene, fullerene, activated carbons, graphite, carbon foams, biochar carbon nanotubes, and carbon aerogels [88]. The carbon-based materials can be used as appropriate candidates in catalysis, electronics, fuel cells, biology, metal recovery, and gas storage and separation [27][88][89].

Among the aforementioned wide range of applications, carbon-based porous materials can serve as appropriate candidates for $CO_2$ capture due to their advantageous, including low production cost [27], competitive $CO_2$ adsorption performance at a given pressure [39][90], easy synthesis, ease of scaling up [88], wide availability, controllable pore structure, high thermal stability [15], good chemical resistance against alkaline and acidic media [91], fast adsorption kinetics [44], lower regeneration energy requirements [86], high apparent density (0.3 g/cm$^3$) [92] [93], high surface area [94][95], environmental benignity [85], favorable surface chemistry [96], selectivity [66], and


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