Carbon Dioxide Separation Technologies

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Carbon dioxide (CO_2) emissions from burning fossil fuels play a crucial role in global warming/climate change. The effective removal of CO_2 from the point sources or atmosphere (CO_2 capture), its conversion to value-added products (CO_2 utilization), and long-term geological storage, or CO_2 sequestration, has captured the attention of several researchers and policymakers.

CCUS carbon dioxide Gas hydrates Net-zero CO2 Separation

1. Introduction

Energy is the primary source of greenhouse-gas (GHG) emissions, with a share of around 76% (mainly CO₂ emissions). Though COVID-19 triggered an exceptional decrease in global GHG emissions in 2020, the largestever annual rise in CO₂ emissions witnessed a CO₂ rise from 31.5 Gt to 36.6 Gt in 2021 $\frac{1}{2}$. To achieve the COP26 targets established for net zero, carbon-capture utilization and storage (CCUS) technologies could be the bottleneck. CCUS is predominantly employed to capture CO₂ produced from different industrial sources, such as steel plants, power plants, chemical industries, and thermal-electric power plants. The most conventional approaches for carbon capture are precombustion, postcombustion, and oxyfuel combustion methods ^[2]. Although there has been extensive research in direct air-capture approaches, the high capital cost has been a significant challenge to deploying this technique. In addition, the development of CO₂ separation techniques (to separate CO₂ from flue or fuel-gas mixtures) has also gained a significant attraction for the economical deployment of carboncapture technologies. In this direction, adsorption, absorption, microbial, membrane separation, and environmentally friendly techniques such as 'gas hydrate-based' separation and biological processes have grown significantly around the world ^{[2][3][4]}. The adsorption, absorption, and cryogenic distillation processes are the most mature CO₂ separation methods with high separation efficiency. CO₂ capture through Gas hydrate-based and membrane separation methods is highly effective due to their low energy consumption. However, these technologies also have drawbacks, mainly temperature requirements, energy intensity, and CO2 concentration dependency [3][5][6][7][8][9]. The industrial applications of CO2 capture through different methods have numerous drawbacks, restricting this process from being used commercially.

2. CO₂ Separation/Capture Technologies

Research and technological advancements have created multiple novel carbon dioxide (CO₂) separation methods. The uncontrollable release of notorious anthropogenic GHGs, especially carbon dioxide, has caused major

environmental issues. The harmful effect of CO_2 has motivated the development of technologies dedicated to achieving net-zero emission goals while evaluating their efficacy, economics, and environmental impacts ^[10]. The flue gas properties (such as composition, temperature, and pressure conditions) are also important parameters for selecting the appropriate process of CO_2 separation ^[11]. The current scenario requires investments to mitigate carbon emissions effectively (CO_2 capture and sequestration) ^[12]. Some primary CO_2 capture techniques investigated globally are adsorption, absorption, chemical looping combustion, membrane separation, microbial/algal separation, hydrates-based separation, and cryogenic distillation method ^{[2][3]}. **Figure 1** compares the advantages and disadvantages of different CO_2 capture/separation techniques mentioned above. As shown in **Figure 1**, absorption, adsorption, and membrane-based separation methods offer high separation efficiency. Though cryogenic separation is the most mature technique, the process is highly energy intensive.

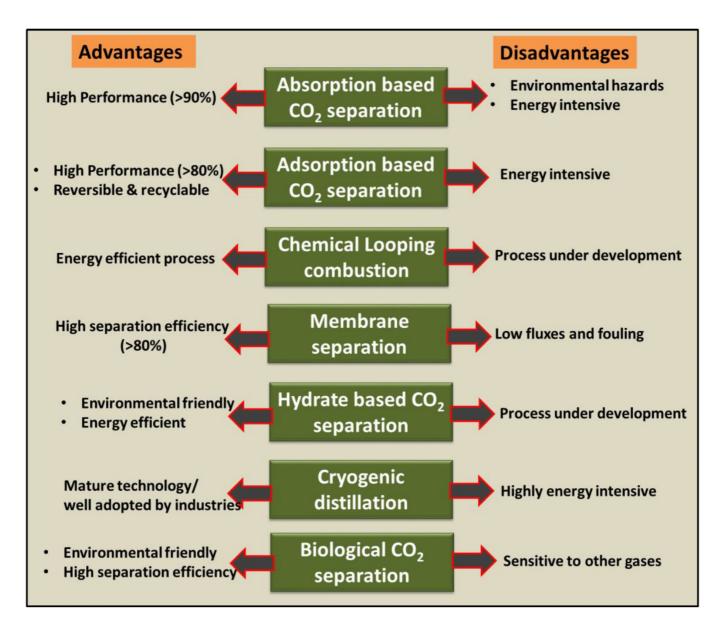


Figure 1. Comparison of different separation technologies for their advantages and disadvantages.

2.1. Absorption

The absorption method is widely applicable in the petroleum, coal, and natural-gas industries for separating CO_2 [13]. Removing CO_2 from a gas stream using different absorbents (physical and chemical) has been used in the industrial sector for over 50 years. The absorption method is broadly divided into two types: physical and chemical absorption [14][15]. Chemical absorption is the process by which a solvent absorbs CO_2 and produces chemical compounds. These chemical components are later reused by removing the absorbed CO_2 through different techniques. However, if the solvent is chemically inert, it does not interact with CO_2 . CO_2 is chemically absorbed in two steps: the treated gas is initially introduced into counter-flowing interaction with the solvent stream. The solvent absorbs CO_2 from the flue gas stream during this phase. As the solvent warms up, CO_2 is desorbed in a stripping column, further migrating to the top of the column, where pure CO_2 is recovered, compressed, and stored [14][16][17].

On the other hand, physical solvents do not undergo any reaction with CO_2 , making them more desirable for CO_2 separation processes. Henry's law of equilibrium in vapor–liquid mixtures governs the physical absorption process. It states that the relative gas pressure in equilibrium with the solvent at any given temperature is directly proportional to the amount of a gaseous phase dissolved in a unit volume of the solvent. Since the physical absorption process is pressure dependent, it performs better than chemical absorption at higher partial pressures of CO_2 , such as in an integrated gasification combined cycle (IGCC) power plants [14][18][19]. The coal, natural gas, and petroleum industries extensively use absorption techniques to segregate CO_2 [13]. Kim and Yang [20] studied the competence of hollow-fiber membranes in CCS with various aqueous absorbents. The capability of the hollow polytetrafluoroethylene (PTFE) membrane filter was measured at varying temperatures. They conclude that the absorption rate of CO_2 increases with the temperature rise. Sensitive absorbents such as 2-amino-2-methyl-l-propanol (AMP) and monoethanolamines (MEA) are vastly active agents to achieve augmented rates of CO_2 absorption. **Figure 2** shows the different categories of absorption medium broadly categorized into physical and chemical absorption. The physical and chemical process, selexol process, selexol process, amine-based process, and inorganic chemical process.

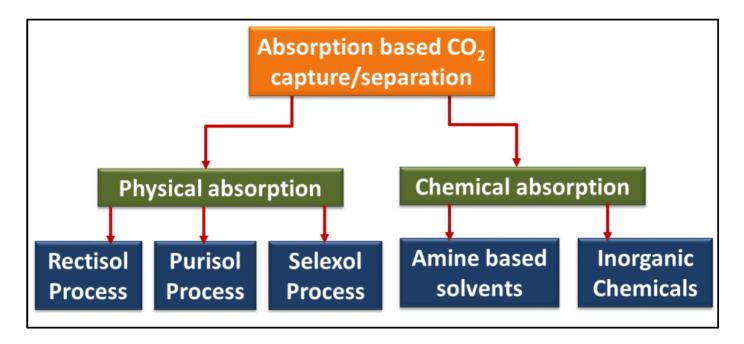


Figure 2. Absorption-based CO₂ capture/separation processes.

In recent years, research has been undertaken on CO_2 capture from fossil-fuel emission sources ^{[21][22]}. The CO_2 absorption in aqueous media is evaluated by the equation ^[23]:

$$CO_2 + 2H_2O \leftrightarrow HCO_3^- + H_3O^+$$

Absorption of CO_2 into aqueous solvent was initially done to purify gases such as synthetic gas, hydrogen, and natural gas ^{[22][24]}. However, research that originated on CO_2 capture from fossil-fuel sources has been done with absorption ^{[21][22]}. CO_2 absorption through the membrane is the combination of gas absorption and membrane separation processes. This method perceived a remarkable perspective in the last decade for capturing CO_2 from flue gas streams ^[25].

The absorption process for the large implementation of CCS is the perhaps amine based which may cause equipment corrosion, solvent loss, and the production of volatile degradation constituents ^{[26][27]}. The release of nitramines and nitrosamines from the degradation of amine emissions can cause potential damage to human health ^[28]. Kozak et al. ^[29] presented a chilled-ammonia process (CAP) to capture CO₂ from flue gases produced by different industries. They suggested that this process required low energy for the regeneration of CO₂ at increased pressure and temperature, reducing the downstream compression, and is more environmentally friendly than the amine processes. Furthermore, the capacity of solvents in absorption was found to be better at lower temperatures, which necessitates the cooling of the solvent before the process and reducing the efficiency of the process ^[19]. Although absorption is the utmost developed CO₂ separation process due to its high efficiency and low cost, it has certain environmental drawbacks due to the disposal of the absorbent after use ^[2].

2.2. Adsorption

Burning fossil fuels has led to the inexorable emission of greenhouse gases (GHG) and responsible for global warming. Carbon dioxide escape can be prevented by capturing it before it gets released into the environment. One such method that has been gaining popularity is the adsorption of CO_2 on adsorbent material at high temperatures. CO_2 adsorption at high temperatures is a significant CO_2 separation method. Adsorption is a physical process in which a solid sorbent is used to fix the CO_2 onto its surface. The adsorption process reduces energy consumption and cost during CO_2 separation. Adsorbents can be used to capture single or multilayer gases depending on the absorbent's temperature, pore size, surface force, and pressure ^{[2][30][31]}. The process employs an adsorbent with a nanoporous surface to precisely adsorb CO_2 from the flue gas. Regeneration of the adsorbent is done by creating a vacuum environment around the adsorbent or by providing heat ^[32]. Generally used adsorbent materials are molecular sieves, zeolites, activated carbon, calcium oxides, lithium zirconate, and hydrocalcites ^[2]. **Table 1** enlists different physical and chemical adsorbents used in the postcombustion capture of CO_2 .

Table 1. Chemical and physical adsorbents efficiency for postcombustion CO_2 [11][33].

Sorbent	Operating Pressure (kPa)	Operating Temperature (K)	CO ₂ Capture Capacity (mol CO ₂ /kg Sorbent)		
CHEMICAL ADSORBENTS					

Sorbent	Operating Pressure (kPa)	Operating Temperature (K)	CO ₂ Capture Capacity (mol CO ₂ /kg Sorbent)		
Mesoporous (MgO) ^[34]	101	298	1.8		
CaO nanopods ^[11]	101	873	17.5		
CaO derived from nano- CaCO3 ^{[11][34]}	101	923	16.7		
CaO-MgAl ₂ O ₄ (spinel nanoparticles) ^[34]	101	923	9.1		
Nano CaO/Al ₂ O ₃ ^[34]	101	923	6.0		
Lithium–Silicate ^[34]	-	993	8.18		
CaO [34]	100	873	17.3		
PHYSICAL ADSORBENTS					
Activated Carbon [35]	110	303	1.58		
NiO-ACs [35]	101	298	2.227		
Na-Y ^{[<u>36]</u>}	101.32	273	4.9		
NaKA ^[35]	101.32	373	3.88		
MWNT ^[<u>37</u>]	101	303	1.7		
CNT at (Cu ₃ (btc) ₂) [36]	1818	298	13.52		
MOF-177 ^[38]	4545	298	33.5		
Pd-GNP Nanocomposite [39]	1111	298	4.5		

Adsorption is broadly categorized into chemical and physical adsorption processes. Chemical adsorption or 'chemisorption' is driven by chemical reactions at the contact surface. Metal salts and metal oxides are compounds that constitute most chemical adsorbents. 'Physiosorption' or physical adsorption does not affect the chemical structure of the adsorbent during adsorption. Inorganic porous materials such as zeolites, hydrotalcite, and activated carbons (AC) are widely used physical adsorbents ^{[30][37][40][41]}. Activated carbon is an economical material with a large surface area and flexible pore structure when treated with activating agents. However, effective CO_2 separation through AC is possible when the AC possesses weak binding energy with carbon dioxide ^[42]. Zeolites are hydrophilic, yet strong CO_2 adsorption agents. However, upon interaction with water, the strength of the links between the interconnected substances reduces, decreasing the adsorption capability of the zeolites. Applying metal–organic frameworks (MOFs) as adsorbents is a new approach. Metal ions or ion clusters are the essential components of MOFs, amalgamated by organic linkers and bridging agents to form stable coordination bonds. MOFs have advantages such as ease of synthesis and design with large porosity and modified pore features. Silica, a non-carbonaceous material, has a large surface area, small pore size, and high mechanical stability. Materials made of mesoporous silica use amine-based compounds to trap CO_2 ^{[40][42][43][44]}. An effective

adsorbent should have the following properties: (i) good mechanical strength, (ii) high sorption kinetics, (iii) high selectivity, and (iv) stable adsorption capacity ^[27].

The various pathways for carrying out the adsorption process are $\begin{bmatrix} 13 \end{bmatrix}$:

- 1. Pressure swing adsorption (PSA);
- 2. Temperature swing adsorption (TSA);
- 3. Electrical swing adsorption (ESA);
- 4. Vacuum Swing Adsorption (VSA).

The recovery of CO₂ captured during the adsorption process through processes such as pressure-temperature swing adsorption (PTSA), vacuum swing adsorption (VSA), pressure swing adsorption (PSA), and temperature swing adsorption (TSA) processes where PSA and TSA are the most widely used techniques. PSA adsorbs CO₂ onto a solid adsorbent surface at fluctuating pressures between maximum and minimum permissible pressure limits. TSA is the process of CO₂ recovery through variations of temperature using hot air or steam. The PSA method is favorably implemented in industrial applications due to its high recovery efficiency (85%) and lower application cost than TSA. However, TSA is observed to be 95% effective in recovering CO₂ from adsorbed surfaces, although it has a longer regeneration time than PSA. The requirement of high temperature during TSA and high energy during PSA methods are the most significant drawbacks of these two methods ^{[6][7][45][46]}. Yong et al. ^[47] reviewed the various adsorbents at high temperatures. They studied material from carbon-based adsorbents with high adsorption capacity for CO₂ separation at surrounding temperature and pressure to other agents such as zeolites, metal oxide sorbents, and hydrotalcite-like compounds (HTIcs). It is of the utmost importance to understand that the choice of adsorbents depends on the operating conditions of the process. MgAl₂O₄, CaO– and nano CaO/Al₂O₃ are the most effective chemical adsorbents. The regeneration of chemical adsorbents is complex, even though they have high adsorption capacity and selectivity ^[30].

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