The Development of Carbon Dioxide Captures and Biochemical Transformation of Carbon Dioxide

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Abstract. In recent years, human activities have led to significant CO\textsubscript{2} emissions. The increase in energy consumption and emissions of greenhouse gases (mainly CO\textsubscript{2}) has led to consequences such as global warming and an accelerated rate of glacial melting, making global environmental development more challenging. Even though the monoethanolamine (MEA) method of capturing carbon dioxide is now widely used in industry, the disadvantages of this method still exist, mainly because of the difficult economic balance. Since CO\textsubscript{2} is inevitable due to human activities, converting the generated CO\textsubscript{2} into high-value clean energy to alleviate the greenhouse effect is a current research hotspot. Therefore, finding a perfect method for capturing CO\textsubscript{2} from industrial and commercial operations as soon as possible is certainly a high priority. This paper provides an overview of the basic principles and practical applications of physical and chemical methods of CO\textsubscript{2} capture and biochemical technology in the conversion of the captured CO\textsubscript{2} into value-added products. The paper describes the current status and challenges faced in the application of carbon capture and storage (CCS) technology worldwide, and finally shows the advantages and prospects of each method. This will lead to the development of a new carbon economy with commercial value, which in turn will facilitate the implementation of CCS on a global scale, ultimately leading to the goal of global carbon neutrality.

Keywords: CO\textsubscript{2}, CCS, MEA, Spirulina treatment, Carbon neutralization.

1. Introduction

In order to mitigate climate change, the Intergovernmental Panel on Climate Change (IPCC) presented a draft decision, the Special Report on CCS (hereinafter referred to as the Report), at the seventh Conference of the Parties in 2001 \cite{1}. Coal has been the optimal way to generate electricity in terms of cost and process compared to oil and gas. CCS separates CO\textsubscript{2} from industrial or associated energy sources. It is seen as one of the options for reducing atmospheric carbon dioxide emissions from human activities. In addition, multiple ways have been explored for converting the captured CO\textsubscript{2} into value-added chemical products, of which the biochemical conversion is a low-energy and economically efficient method for large-scale application. With the growing international concern about global warming, carbon reduction has become an important research topic from a global perspective. In order to achieve medium and long-term CO\textsubscript{2} reduction targets, various CO\textsubscript{2} capture technologies such as absorption, membrane separation, adsorption and low-temperature distillation can be used.

2. Physical methods for Carbon dioxide capture

According to the different principles of CO\textsubscript{2} separation, physical methods can be divided into solvent absorption method, the adsorption method, membrane separation method and low-temperature distillation method.
2.1. Solvent absorption method

In the light of Henry’s law, the monoethanolamine (MEA) scrubbing method has been extensively used to remove and capture CO$_2$ from the flue gas of a combustion process. The reference capture process used is the standard process presented in Fig. 1. Firstly, the post-combustion CO$_2$ capture (PCC) is mixed in the absorber in contact with MEA. Secondly, the CO$_2$-rich MEA solution is subsequently sent to the stripper tower for heating, where the liquid is distributed directly at the top of the tower, and the gas leaves the top of the vessel. Finally, the MEA solution with very low CO$_2$ concentration is circulated to the top of the absorber, thus releasing almost pure CO$_2$ gas.

On the one hand, the advantages of the MEA process are its high absorption rate of CO$_2$, its high absorption capacity, the simplicity of the process, and its low cost, which makes it become the best of all the methods used for large scale CO$_2$ capture. According to Rochelle, among the various CO$_2$ capture physics schemes currently available, the best traditional solution for CO$_2$ emissions from fossil-fuelled power plants is the MEA method, which is currently the most established commercial technology [2]. On the other hand, the MEA method has already been widely used commercially. However, according to Stewart and Hessami, this process has several disadvantages, such as the large size of the equipment required for the reaction and the difficulty of degrading the solvent after the reaction, which causes environmental pollution [3]. However, the main limitation of this process today is the high energy consumption, with the high energy losses in the solvent regeneration phase accounting for about half of the overall operating costs. Therefore, improving the thermal efficiency of solvent regeneration should be the first option for further research in this technology.

Recently, the CO$_2$ capture using triamine solutions has come to the forefront. This new method uses three conventional amines: MEA, 1-dimethylamino-2-propanol (1DMA2P), and piperazine (PZ) formulated into an effective triamine mixture to capture CO$_2$ [4]. The process was evaluated by using different molar ratios of triamine mixtures to capture CO$_2$ to assess the energy consumption and regeneration process costs for each reaction. It was concluded that all the triamine blends tested outperformed the single MEA solution in terms of energy consumption and regeneration process costs. For example, the blends reduced the energy consumption of the reaction by approximately 30 to 50% and increased the recyclable capacity by at least 40%. Obviously, this method is still in the laboratory stage now, but the new method could be a potential alternative to the MEA process mentioned above for industrial CO$_2$ capture. To sum up, MEA is a mature and suitable technology for commercialization, but there are still some difficult problems and challenges to be solved.

![Figure 1. MEA scrubbing process [5]](image)

2.2. Membrane separation method

Membranes have emerged as a promising process, and an energy-efficient, low-cost method of capturing carbon dioxide, and many scholars are now exploring membrane operations to capture carbon dioxide from power plant emissions and other fossil fuel flue gas streams [6]. The principle
of using membrane technology to capture carbon dioxide is to use the difference in permeation rates of membrane materials for different gases to separate the gas one needs. Theoretically, selecting the right membrane material allows carbon dioxide to be separated individually. During the process, the hot gas mixture from the power plant is led to the absorber, where it is cooled to a temperature at which the membrane can work properly, after which the cooled gas mixture is transported to the membrane and separated out using the difference in permeability of the membrane material to the different gases.

The advantages of this technology are low cost, no solvent regeneration and thus environmental pollution, and manageable short-term costs. However, many problems limit the use of membranes, and even with highly selective membranes, the absorption efficiency is not high for flue gases with less than 20% CO$_2$ in the feed mixture [7]. In addition, according to Brunetti et al., membranes need to be heat and chemical resistant and have long-term cost-effectiveness, among other things [8]. In fact, the main drawback of membrane operation is simply the low selectivity of current membrane materials. According to a recent study by Hu et al., the introduction of nanosheets into PIM-PMDA-OH membranes to create new mixed-matrix membranes (MMM): MMMs can significantly improve the selectivity of CO$_2$ [4]. Finally, Hu et al. 7-day test found that MMMs are more resistant to aging than pure polymer membranes, with the permeability of the former decreasing by only about 20%, which indirectly reduces the cost of using MMMs membranes. In conclusion, membrane technology is theoretically superior in capturing CO$_2$ and is less costly in the short term, but it is still immature in the long term.

2.3. Physical adsorption method

Solid adsorbents such as activated carbon, zeolites and some metal oxides have been widely used for the gas separation to treat power plant flue gases [9]. The advantages of these solid adsorbents are that CO$_2$ capture is cost-effective, simple and does not involve the formation of chemical bonds through the interaction between the adsorbent and the adsorbent. It is urgent to develop this new technology to reduce costs and increase efficiency in many power plants. The principle of the solid sorbent method is that gas is removed from one or more components of a gas mixture with the help of intermolecular forces on the surface of a specific solid material. Furthermore, this process is also influenced by temperature, pressure, surface forces and the pore size of the solid adsorbent [10]. Therefore, according to its characteristics, the adsorption process can also be divided into pressure or temperature swing adsorption (PSA/TSA).

According to Stewart and Hessami, one plant already uses alumina as a desiccant to recover carbon dioxide [3]. With this improved system configuration, the overall power consumption has been reduced by 3%, and more than 90% of the CO$_2$ produced has been removed in the overall test results. Although some solid sorbents are capable of capturing CO$_2$ using PSA, such examples are still not commercially available due to their low CO$_2$ adsorption capacity at ambient temperature and pressure, their intolerance to moisture and their mono-gas capture, which indirectly leads to increased costs [11]. In conclusion, solid sorbents have advantages as potential CO$_2$ sorbents, but the disadvantages are still evident when treating the large amounts of CO$_2$ generated by power plants.

In addition to the common solid sorbents used in adsorption technology today, carbon molecular sieves (CMS) have been used in CCS. CMS represents a special class of nanoporous carbon with the advantage of allowing selective adsorption of certain components of a mixture based on differences in size, shape or adsorption equilibrium [12, 13]. The preparation of CMS usually requires various carbonaceous materials to be synthesized. Among the existing methods, there are two ways to manufacture microporous CMS. The first is by controlled pyrolysis of the carbon precursors at the pores, and the other is based on carbon vapor deposition (CVD) production. The latter is a more suitable method for today's technological developments. According to Morali et al., the use of CVD to prepare CMS has higher hydrophobicity, higher resistance to alkalinity and acidity, higher thermal stability and higher molecular selectivity than conventional solid absorbers [14]. However, this
method is a complex process. To summarize, the use of molecular sieves to capture carbon dioxide still has many inconveniences and is still not commercially viable.

2.4. Physical adsorption method

Cryogenic separation technology has been studied for decades as one of the available methods for capturing carbon dioxide. However, this technology has not been widely studied due to the expected high cooling costs and the limited development of industrial technology previously. But, according to Sun et al., an increasing number of review papers on biogas upgrading techniques have been published in recent years [15]. And these scholars have demonstrated that cryogenic separation techniques to extract high purity carbon dioxide and methane are feasible on a laboratory scale. Cryogenic separation techniques for capturing CO₂ have several distinctive merits [16]. And Tuinier et al. use a new process for the capture of cryogenic CO₂ using a dynamically operated packed bed to recover 99% of the CO₂ in the flue gas at a certain cost. Furthermore, with the successful application of a mechanical cooler typical of cryogenic systems: the Stirling Cooler (SC) for CO₂ capture, operating costs are controlled, and environmentally harmful substances are reduced in the reaction [17]. However, there are drawbacks to this approach, such as the whole system needs to take a long time to reach the required low-temperature conditions. Therefore, expensive heat exchangers have to be used to resist the effects of temperature changes, which again increases the cost. To summarize, cryogenic separation technology has been proven to be feasible on a laboratory scale, if it is to be commercialised on a large scale its technology still needs to be continuously developed.

3. Chemical methods for Carbon dioxide capture

3.1. Chemical absorption method

The absorption method refers to the method that CO₂ is absorbed into the bulk phase of another material to achieve CO₂ enrichment. At present, the absorption method is the only technology that can be widely used in commercial deployment, which is usually used to separate CO₂ from other exhaust gases after combustion. The shortcomings corresponding to the early development and wide application of the absorption method are also obvious, such as the unstable cost, the difficulty in temperature control, the low wear resistance, the high heat capacity of the machine, and the low CO₂ selectivity in the absorption process. Therefore, the current research has carried out relevant shortcomings improvement and technical iteration [18]. For example, the ‘phase change’ carbon dioxide capture technology born from the new generation of transformational carbon capture technology effectively solves the high energy consumption problem caused by the need for all heating and regeneration of carbon dioxide absorption liquid in the traditional method, which can greatly reduce the carbon dioxide capture integration cost and has great social and economic benefits. This method reduces the overall cost by reducing the energy consumption of the heating regeneration process of the absorbent. Experts’ witness data show that the regeneration solution volume of the phase change carbon dioxide capture technology can be reduced by 40%-50 %, and the regeneration heat consumption is lower than 2.3 GJ t⁻¹ CO₂, which is more than 45 % lower than the traditional ethanol amine absorbent used earlier [19].

3.2. Chemical adsorption method

The adsorption method refers to selectively absorbing CO₂ molecules onto the surface of another material through weak van der Waals force (physical adsorption) or strong covalent bond force to realize the enrichment of CO₂. The advantage of the adsorption method is the specific adsorption of the adsorbent. Adsorbents that adsorb CO₂ can be regenerated in different ways according to their adsorption mechanism while releasing adsorbed CO₂, which can reduce the cost accumulation caused by raw material loss. Many researchers believe that compared with the absorption method, the adsorption method has the characteristics of low cost, strong thermochemical stability, low machine loss, long service life, low heat capacity, and high CO₂ selectivity. However, this method also shows
shortcomings on the other hand: rotating bed or circulating bed commonly used in variable temperature adsorption systems requires independent adsorption and regeneration working environment, which cannot realize the continuous adsorption-regeneration process of a single fixed bed.

Given the above shortcomings, a rapid temperature swing adsorption rotary direct air carbon dioxide capture system and method are explored and invented to realize the rapid capture of carbon dioxide. Innovation mainly relies on the adsorption and regeneration areas of wheels. The carbon dioxide adsorbent in the regeneration zone releases the adsorbed carbon dioxide gas through heat exchange with the regenerated gas (water vapor to condensate water process, pollution-free) to realize the regeneration of carbon dioxide adsorbent in the regeneration zone. And the wheel rotates continuously during the working period to achieve a continuous adsorption-desorption cycle with high efficiency [20].

In addition to improving the components, the adsorbents can also be improved, such as the carbon dioxide capture technology of solid adsorbents. A CaO-based material has been developed as an advanced adsorbent with a wide temperature threshold. The reactivity of high-temperature CaO-based materials with carbon dioxide is high, and the cost of CaO-based adsorbent capture technology is lower than that of the amine washing technology currently used [21]. In addition, the adsorption enhanced water gas shift (SEWGS) developed by the Energy Research Centre of the Netherlands (ECN) uses CO\textsubscript{2} flushing, which is a typical pre-combustion intermediate temperature gas purification technology. By introducing two steps of co-directional high-pressure CO\textsubscript{2} flushing and reverse low-pressure steam purging in the traditional PSA, the loss of the capture system is greatly reduced [22].

3.3. Membrane absorption method

In the membrane absorption process, the mixed gas and absorption liquid have no direct contact, the microporous membrane has no selectivity and only plays a role in isolating the mixed gas and absorption liquid. The pores in the membrane allow gas molecules to pass through the membrane to the other side without high pressure. As shown in Fig 2, this process mainly relies on the selective absorption of the absorption liquid on the other side of the membrane to separate CO\textsubscript{2} from the mixed gas [23].

![Membrane morphology diagram](image_url)

**Figure.** 2 Schematic diagram of membrane morphology [24]

Following the update of membrane materials, high carbon dioxide selectivity polymer (ionic liquid) has also been innovated. Compared with the most commonly used aqueous amine CO\textsubscript{2} absorbents,
Ionic liquids can avoid the disadvantages of large evaporation loss, easy degradation in heat, non-corrosion resistance and easy wetting of polymer membranes, and have their advantages. Therefore, the Diethanolamine glycine salt is innovatively developed—an ionic liquid prepared by hydroxyl amino acid ammonium salt for carbon dioxide capture and emission reduction. With the change of process conditions, such as the increase of gas and liquid floats, the membrane flux of gas absorbed by the membrane can be directly changed. Ionic liquids are easier to be regenerated, with higher regeneration efficiency and average regeneration speed [25].

### 3.4. Electrochemical process

Winnick first proposed using molten carbonate fuel cell membrane to separate CO$_2$ from the air of the flight cabin and first studied using molten carbonate membrane to separate CO$_2$ from the flue gas of power plants. Molten carbonate fuel cells transmit CO$_2$ through a membrane under a closed circuit (using an external electromotive force). The reaction mechanism is as follows in equation (1) and (2) [26]:

\[
\text{Cathode: } \text{O}_2 + 2\text{CO}_2 + 4e^- = 2\text{CO}_2^-^3 \\
\text{Anode: } \text{H}_2 + 2\text{CO}_2^-^3 = 2\text{CO}_2 + 2\text{H}_2\text{O} + 4e^- 
\]

\[
\text{electrochemical carbon capture}
\]

Figure 3. Electrochemical Carbon Capture process

As shown in Fig 3, the molten carbonate electrochemical cell has several advantages in CO$_2$ separation: the application of molten carbonate in fuel cells has a wide range of technical basis; with the increase of temperature, about 100% of molten carbonate transported CO$_2$, the conductivity is about 1s cm$^{-1}$ at 600$^\circ$C, and the diffusion rate of CO$_2$ is about $10^{-5}$cm$^2$ s$^{-1}$, which makes it suitable for separating CO$_2$ from flue gas of power plant. It is expected to become a competitive CO$_2$ capture technology [27, 28].

However, there are also many shortcomings in the use of fused carbonate electrochemical batteries for CO$_2$ separation from flue gas of power plants: fused carbonate is highly corrosive at high temperatures, and its production and operation are difficult; SO$_2$ in flue gas also poisons the battery, in the high-temperature flue gas environment, there are still problems of electrolyte isolation and electrode degradation [29]. In addition, the molten carbonate electrochemical method also needs to make breakthroughs in the development of carbonate ion solid electrolytes with higher conductivity and further optimize the process. In general, electrochemically mediated CO$_2$ capture and concentration is considered to be an effective solution to overcome the barriers of loss of renewable energy, high cost and loss of active adsorbents. The details are as follows: Potential Regulation of Nucleophilic Electrochemical Fluctuations. The innovation of the electrochemical method is that the applied potential is used to adjust the electrochemical fluctuation of nucleophilicity, which fills the technical gap of air and flue gas mixture at low and medium pressure. In organic redox and transition metal redox, intercalation battery, redox flow battery and bionic inspired design are innovated [30].
4. Biochemical CO$_2$ catalytic conversion methods

As the reaction sites, the reaction conditions, and the molecular size of the reaction product are different, three typical biochemical conversion methods in this article are mentioned, which are easy to build connection and present different medium in process.

4.1. Bacterial-based conversion of CO$_2$

The bacterium used in bacterial catalysis includes methanobacter thermoaggregatives, thermoautotrophic methanobacter, thermoautotrophic methanococcus, salt-tolerant methanobacter ovale, thermoautotrophic, tethanococcus, methanogenes, methanogens petroleum, campylobacter methylus and methyl campylobacter [31]. Methyl campylobacter IMV3011 can catalyze the biotransformation of carbon dioxide to methanol. The reaction principle is that CO$_2$ is reversely reduced to HCOOH, and HCOH is further reacted to generate methanol [32]. But the ability of methane-oxidizing bacterial cells to synthesize methanol is limited by the intracellular reducing equivalent. To solve this problem, the decomposition of poly-β-hydroxybutyric acid (PHB) stored in the cell can generate reducing equivalents, catalyzing the degradation of trichloroethylene for a long time and improving the methanol production capacity.

The initial concentration of nitrogen and copper in the nutrient medium can adjust the accumulation of PHB to improve the ability of methyl campylobacter IMV3011 to reduce carbon dioxide to methanol.

However, heavy-metals-containing catalysts, such as copper sulfate, will affect cell activity and drive frequent cell suspension replacement and increase cost.

4.2. Enzyme-based conversion of CO$_2$

There are several representative chemical methods for the conversion of carbon dioxide to small organic molecules, such as heterogeneous catalysis, electrocatalysis, and photocatalysis. However, the conditions required by these methods are often very harsh [33]. For example, the heterogeneous catalysis method requires high temperature and pressure, while the latter two methods require external electrical energy or light energy, and the conversion rate is not high. Unlike that, enzyme conversion is a valuable method because it has advantages: mild reaction conditions, high conversion rate, and fast reaction rate. Those enzymes include lactate dehydrogenase, malate dehydrogenase, oxaloacetate decarboxylase, multi-enzyme method formate deoxygenase, formaldehyde deoxygenase and alcohol dehydrogenase. The process of Enzyme catalysed CO$_2$ conversion is shown in Figure 3.

In the sol-gel immobilization multi-enzyme method, multi-enzyme method formate deoxygenase, formaldehyde deoxygeanase and alcohol dehydrogenase, three dehydrogenases are used as catalysts. Nicotinamide adenine dinucleotide (NADH) is used as the electron donor, and carbon dioxide is used to catalyze methanol production. The experimental results showed that the methanol conversion rates could reach 92.4 % at 37 °C and pH 7.0 [34]. However, due to the small spatial configuration of the enzyme Changes and the existence of the steric hindrance effect, the enzymatic activity after encapsulation decreased slightly compared with the results of liquid-phase enzymatic reaction [35]. And the sites of multiple reaction steps are the same, and it is difficult to control the sequence of enzyme-catalyzed reactions in each step, resulting in a reduction in methanol production from carbon dioxide.

4.3. Algae-based conversion of CO$_2$

Spirulina belongs to algae, which can be used in the treatment of CO$_2$, the exhaust gas of thermal power plants. The appropriate amount of aeration can promote the growth of spirulina, while excessive aeration inhibits the growth of spirulina [36]. So the place where CO$_2$ is processed is close to the thermal power plant where high-concentration CO$_2$ is generated, and there is no CO$_2$ diffusion link, saving transportation costs. When the continuous air intake device is used to absorb and prepare the exhaust gas, the smaller the air intake flow rate, the higher the CO$_2$ absorption rate of spirulina;
the absorption effect of the series component is better than that of the single component, and the maximum absorption rate can reach 99% [37]. However, the energy consumption to maintain the suitable temperature for spirulina is large, the enzyme activity is easily affected, the reaction rate is slowed down, and a large amount of CO₂, SO₂ and other waste gases are dissolved in water, which makes it difficult to maintain the PH at the suitable PH of spirulina, and the conversion rate of spirulina after processing a large amount of CO₂ The rate dropped significantly.

Microalgae and cyanobacteria are the main types of various algae to convert CO₂ to synthesize single-cell protein [38]. The reaction principle is that Aerobic methanogens convert CO₂ to amino acids and glyceraldehyde-3-phosphate mainly through Kelvin and TCA cycles and then synthesize single-cell protein.

5. Conclusions

In summary, Carbon dioxide capture and storage (CCS), with biochemically converting low-value-added CO₂ into high-value-added chemicals, is one of the most promising CO₂ reduction and conversion measures essential for achieving carbon neutrality, and its significance for quality economic and social development is irreplaceable. However, the world is still using coal combustion as a low-cost way to generate electricity. This is because, according to the above assessment, none of the relevant technological research, whether physical, chemical methods, is perfect and so requires more scientific investment and time to settle. Therefore, there is a growing consensus that achieving complete carbon neutrality is still a long way off for all the world’s people, and it will require a concerted effort to overcome the technical challenges of development.

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