

Research progress of small molecule carbon dioxide capture agent

Jiayao Jiang

South China University of Technology, Guangzhou, Guangdong, China

1030890233@qq.com

Abstract. At present, the excessive emission of carbon dioxide has threatened people's living environment and future development. More and more countries have begun to pay attention to carbon emission reduction. China has set the dual-carbon strategic goal of "carbon dioxide content reaches its peak" and "carbon neutrality". According to the research on carbon dioxide capture technology by many scholars at home and abroad, it is found that the use of chemical absorbents to capture carbon dioxide is a relatively low cost, less technical difficulty, and can be widely put into industrial production. In this paper, several common chemical absorbents such as hot potassium alkali method, organic amine absorption method, amino acid salt, ionic liquid and phase change absorbent are reviewed. Through the research of various kinds of absorbents by researchers from various countries, their reaction principle, performance advantages and disadvantages, research status are introduced, and the future development direction of carbon dioxide negative emission technology is analyzed.

Keywords: Carbon peak and carbon neutrality; Carbon dioxide capture; Chemical absorbents; Phase change absorbents; Ionic liquids.

Human activities are becoming more common as global industrialization is promoted and economic conditions continue to improve. Over-exploitation and utilization of fossil resources result in a large accumulation and excessive emission of greenhouse gases represented by CO₂, which causes continuous global climate warming, and a variety of extreme weather events and natural disasters occur on a regular basis all over the world. According to this trend, the earth's ecological environment, as well as human survival and development, will face a major crisis. As a result, reducing CO₂ emissions has become a hot topic of increasing concern for people all over the world. The United States, the European Union, Japan, and other countries have established a number of major research plans to support ongoing research on "carbon emission reduction" and to advance the goal of carbon neutrality. China published a white paper on China's Policies and Actions to Address Climate Change in October 2021. General Secretary XI solemnly declared at the United Nations Biodiversity Summit that "China's CO₂ emissions will peak by 2030 and strive to achieve carbon neutrality by 2060" [1]. This statement has set an example for all countries around the world, as well as highlighted the trend that all walks of life in China should develop in the future in the direction of "green" and "low carbon."

Indeed, as early as the 1980s, carbon dioxide capture, utilization, and storage technology (CCUS) aimed at fixed emission sources such as large oil and coal-fired power enterprises began to be incorporated into industrial tail gas treatment processes, where CO₂ was collected by capture technology, compressed, transported to oil mines, and converted to supercritical state, which was then used to improve oil recovery and geologically sealed.

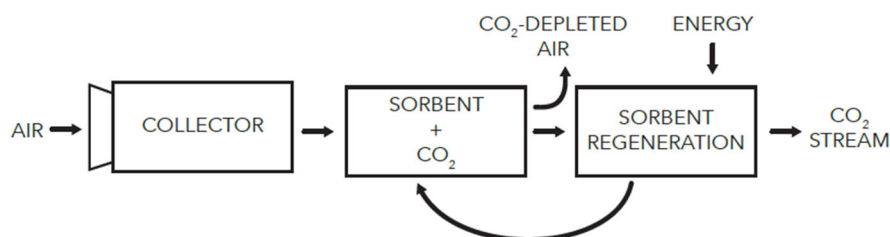


Fig. 1 Direct atmospheric CO₂ capture process diagram

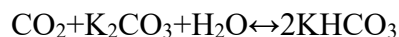
However, because scattered CO₂ emissions account for nearly half of all CO₂ emissions in the atmosphere [2,] capture and storage technology limited to point source emissions can only slow the rate of CO₂ content increase, not reduce it. Negative CO₂ emissions must be realized in order to deal with climate change [3]. The idea of directly capturing CO₂ (DAC) from the atmosphere has been highly concerned by the scientific community and widely supported by the international community among many negative emission technologies that have been proposed or are in the research stage (Fig.1). In theory, there is no significant difference between capturing CO₂ from a point source and capturing dispersed gas directly from the atmosphere; however, the latter requires a more efficient and stable collector to interact with CO₂, and its manufacturing and transportation costs should be kept within an acceptable range. CO₂ capture technology is currently classified as pre-combustion capture, in-combustion capture, and post-combustion capture based on capture time, and physical capture technology and chemical capture technology based on capture methods. Among these, post-combustion capture technology is more practical and has a broader range of applications. Chemical absorption, which uses chemical reagents to absorb CO₂, is the most mature and stable method, with the highest absorption efficiency.

In fact, chemical absorption is a method that uses a chemical solvent to separate the components soluble in the absorbent from the gas phase via chemical reaction selectivity. CO₂, as a chemical substance with acidic and hydrogen bond receptors, will react with alkaline absorbents such as ethanolamine, potash, ammonia, and other mixed solutions to form carbonate or bicarbonate, which will absorb and separate carbon dioxide. When the external environment, such as temperature or pressure, changes, the reaction is reversed, resulting in carbon dioxide desorption and cyclic regeneration of absorbent [4], as well as a significant reduction in gas transportation cost and storage difficulty. However, an absorbent with almost no flaws has yet to be discovered. As a result, there is still a lot of room for research in the selection of chemical absorbents.

Several types of chemical absorbents commonly used in chemical CO₂ capture technology are introduced in this paper, and the current research progress and development prospects of various absorbents are analyzed and summarized, in order to provide some assistance for the future development and application of chemical CO₂ capture technology.

1. Inorganic absorbent

This method primarily employs an alkaline absorbent to react with CO₂ to form carbonate, which is then tightly coupled in the form of an ionic bond. One of the early capture methods was using K₂CO₃ aqueous solution to capture CO₂, also known as the hot potash solution method [5]. The hot potassium carbonation solution process has been widely used in decarbonization processes in natural gas, hydrogen production, and other industries after a long period of development [6]. The following exothermic reaction occurs when carbon dioxide is absorbed into the potassium carbonate/bicarbonate solution:



The specific steps are as follows: a high-concentration potassium carbonate aqueous solution absorbs carbon dioxide to generate potassium bicarbonate at 90~110°C and pressure, and then the generated potassium bicarbonate is decompressed to desorb CO₂ and generate potassium carbonate simultaneously, allowing the absorption liquid to be recycled. Because the temperature of CO₂ absorption by hot potash solution is very close to the temperature of CO₂ release by reverse reaction of absorption solution, this trapping method can significantly reduce the amount of energy consumed for reboiling. Simultaneously, as the concentration of potassium carbonate increases, so does the absorption capacity of the absorption solution, which accelerates the absorption reaction rate.

Among the many absorbents used in chemical absorption methods, the advantage of hot potassium carbonate solution process is that it has low absorption heat, which is more than 50% lower than that of organic amine system, so the energy consumption of solvent regeneration is low, the solvent itself

is not easily degraded, it has strong oxidation resistance and SO_x and NO_x impurity resistance, low volatility and toxicity, and it is low in cost. However, one of the most significant disadvantages is that the rate of absorbing carbon dioxide at normal temperature and pressure is extremely slow, which is referred to as physical absorption. Previous research compared the reaction rates of the hot potassium carbonate solution process and the ethanolamine method and discovered that the ethanolamine solution's carbon dioxide absorption rate was four times greater than that of the hot potassium carbonate solution process, which would reduce production efficiency if used in industrial production. Furthermore, K₂CO₃ is easily precipitated at lower temperatures (30°C), and the lowest working temperature is better than 70°C, so increasing the temperature of the system is one way to accelerate carbon dioxide absorption.

Furthermore, in light of these shortcomings, many researchers have improved the hot potassium carbonate solution process, primarily by adding different rate accelerators (that is, activators) to potassium carbonate reagents, thereby improving reaction efficiency. Organic amines, biological enzymes, and inorganic salts are currently the most common activators. However, biological enzymes have strict requirements for solution acidity and alkalinity, as well as reaction temperature, and are easily inactivated if the conditions are not properly controlled, so they are still in the research stage and will not be used extensively for the time being.

It was discovered that arsenic trioxide had an excellent activation effect, and Le Tsinghua and others investigated the effect of activated carbonate systems such as arsenic trioxide, arsenite, and vanadium pentoxide on enhancing carbon dioxide absorption. However, because arsenite-containing solutions are highly toxic and carcinogenic, causing significant harm to the health of experimenters, their popularity and use are limited [7].

Among the most common organic amine activators are diethanolamine (DEA), aminoacetic acid, 2-aminoethoxyethanol (AMET), triethanolamine (TEA), diethylenetriamine (DETA), methyldiethanolamine (MDEA), piperazine, and sterically hindered amines such as AMP. However, research indicates that when the concentration of diethanolamine exceeds 5%, the activation is insignificant and the product is highly corrosive, so in industrial production, the concentration of diethanolamine is generally controlled at 2.5%~5% [8]. Furthermore, because diethanolamine and aminoacetic acid are both nonionic surfactants, when they coexist in potash solution, they produce a new compound A, which also participates in the carbon dioxide absorption process and eventually produces carbamate, thereby accelerating the absorption rate. The reaction will be accelerated if boric acid is added to promote the hydrolysis of carbamate vinegar [9], which is also known as the double activation catalysis hot potassium carbonate solution process. Wang Jun et al. also investigated the gas purification effects of diethanolamine, aminoacetic acid, and sterically hindered amine. It was discovered through data analysis that all of them can meet the production requirements under increasing system load, and the purification effect when sterically hindered amine is used as activator is obviously better than when the other two types are used, but the volatilization loss of sterically hindered amine is large under high heat load. As a result, if the heat load is high, sterically hindered amine should be used with caution in actual production to avoid increasing production costs [10].

Sangwon Park[11] and others believe that amino acid salts can also serve as activators in place of amines. This is due to the fact that they all share the same amino functional group. Furthermore, amino acid salts have low vapor loss due to ion formation in solution, making them more environmentally friendly than some amines. The absorption rate of glycine activated solution is faster than that of diethanolamine, and arginine can also reduce the equilibrium partial pressure of CO₂ in the composite solution. As a result, in recent years, an increasing number of researchers have begun to investigate the interaction between amino acid salts and K₂CO₃ in solution, and have discovered that adding amino acids to the solution is indeed expected to improve CO₂ loading capacity and absorption rate. However, amino acid salts are only effective when the temperature is above 70°C, so more research is needed.

2. Organic amine absorbent

If chemical absorption method is the most mature and widely used carbon capture technology at present, then organic amine solution is the most widely studied and common absorbent in chemical absorption method, and has been widely used in flue gas and natural gas purification processes. The absorption process is shown in Figure 2.

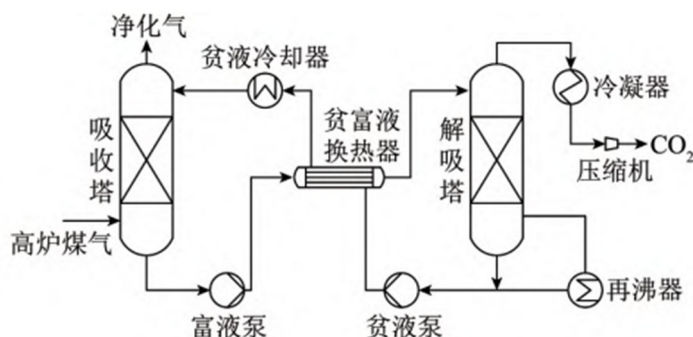


Fig. 2 Flow chart of amine solution absorption

Table 1. Comparison of five commonly used organic amine absorbents and their advantages and disadvantages

absorbefacient	advantage	disadvantage	References
Monoethanolamine (MEA)	High absorption activity; Good absorption capacity; Cheap cost	Large solvent loss and low load capacity; High energy consumption for regeneration (3.6 ~ 4.0 GJ/t in terms of energy consumption per 1t gas absorption); It is easy to degrade and foam, resulting in side reactions to reduce decarbonization efficiency; After reacting with CO ₂ , the product is easy to corrode the equipment.	[14]
N-methyldiethanolamine (MDEA)	Compared with MEA solvent, it has good chemical stability, little possibility of foaming and corrosion during absorption and is not easy to degrade and deteriorate.	The absorption rate is lower than that of MEA solution, and a suitable activator needs to be added. Easily degradable	[15]
Diethanolamine (DEA)	The absorption rate is faster than that of MEA; Lower cost	Easy to degrade; Large energy consumption for regeneration; Strong corrosiveness	[16]
Piperazine (PZ)	The absorption rate and capacity are obviously better than those of MEA.	Low solubility, generally can only act as an activator.	[15]
Sterically hindered amine (AMP)	The theoretical absorption of CO ₂ is obviously higher than that of primary and secondary amines. Low energy consumption for regeneration; Because the structure will produce steric effects, the produced carbamate will be decomposed into carbonate or bicarbonate, which has little influence on the corrosion of equipment.	Expensive; The volatilization loss is large.	[15-17]

Many traditional organic amine solutions exist, and some of the organic amines used as activators in the hot potassium carbonate solution process are also good CO₂ absorbents. The reaction mechanism of organic amines with CO₂ differs depending on the type. For example, the reaction of primary and secondary amines with CO₂ follows the "zwitterionic mechanism," and the reaction is divided into two steps, in which the gas amine first generates zwitterionic intermediate, and then deprotonates to generate ionic carbamate; however, due to a lack of active hydrogen ions, the nitrogen atom of tertiary amine follows the "base catalysis theory," which was proposed by DONALDSON and others, indicating that As a result, different organic amine solvents have different advantages and disadvantages, which can be analyzed and compared in Table 1 with five alcohol amine absorbents.

Although the reaction rate of organic amines is fast and the cost is low, they have the disadvantages of easy degradation, high corrosiveness, and high energy consumption for regeneration. However, the table shows that two amines, such as N-methyldiethanolamine (MDEA) and piperazine (PZ), can complement each other in their advantages and disadvantages, implying that mixed amines can be used as absorbents to reduce reaction energy consumption due to their synergistic effect, which is also the experimental direction of many researchers. ZHANG[14] discovered that a mixed amine absorbent of MEA/MDEA/PZ can reduce regeneration energy consumption by 15.22%~49.92% compared to pure MEA. Through a series of comparative experiments, Xu Zhicheng [18] and colleagues discovered that when 2 mol/L of 1,4-butanediamine (BDA) was mixed with 4 mol/L of N, N-diethylethanolamine (DEEA), the absorption efficiency was excellent, the absorption was stable, and the amount of CO₂ in the rich solution after absorption could reach 97.4 percent of the total absorption. Some researchers have discovered that combining organic amines with other amines, alcohols, ethers, and other substances can cause the absorbent to change phase. When the diethylenetriamine/n-propanol/water phase change system ratio is 3: 3: 4, the absorption capacity increases by 8.5 percent when compared to a single 30 percent ethylene triamine solution [19]. However, the shortcomings are clear: the mechanism is complex, the processes are numerous, and the production cost is relatively high, making it unsuitable for large-scale use in industrial production. Furthermore, high boiling point solution or ionic liquid can be used to reduce the energy consumption of organic amine desorption. In recent years, scholars both at home and abroad have been interested in the phase change absorption formed by combining some functional ionic liquid with an alcoholic amine solution.

3. Amino acid salt absorbents

The amino functional groups in amino acids and ethanolamines are structurally similar. The study of absorbing carbon dioxide with amino acid salts as solutions has also become popular in recent years. According to the findings, amino acid salts that can be used to absorb carbon dioxide primarily include glycine, alanine, proline, and sarcosine, with proline and sarcosine having lower pK_a values and higher apparent rate constants in the experiment and thus being considered better absorption solvents. Because amino acids are mostly neutral, the reaction effect with the acid gas CO₂ may be negligible. Alkaline substances such as LiOH, NaOH, and KOH are frequently used to deprotonate amino acids in order to improve their carbon dioxide absorption rate, and it has been discovered that among these amino acid salts, potassium salt has better activity and solubility [20], so many scholars are committed to researching potassium amino acid as a new carbon dioxide absorbent.

MEA is currently the most commonly used absorbent in industry, so researchers frequently use it as a reference when investigating the properties of new absorbents. As shown in Figure 3, Huang Zhongbin and colleagues discovered that the initial degradation time of MEA was 31 minutes, while that of potassium alanine was 167 minutes, which was approximately four times that of MEA at the same solvent concentration of 2.5mol/L. Adjusting the absorption temperature reveals that the saturated absorption of carbon dioxide by MEA decreases by 11.6 percent with temperature change, whereas potassium alanine only changes by 3.6 percent [21]. Yan Mingwei et al. compared the absorption effects of five absorbents, namely potassium glycinate, potassium glutamate, potassium proline, potassium sarcosine and MEA, under the same CO₂ load, and found that the removal rate

was potassium proline (82.94%) > potassium glycinate (81.8%) > potassium sarcosine (81.33%). All of them are higher than MEA(80.63%), but at the same time, it is found that the removal rates of potassium amino acids change greatly with the increase of carbon dioxide load from 0.2mol/mol to 0.35mol/mol, which shows that amino acid salts are more suitable for the absorption process of carbon dioxide gas at low load.

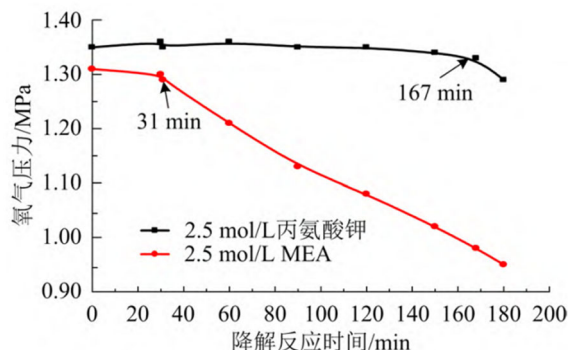
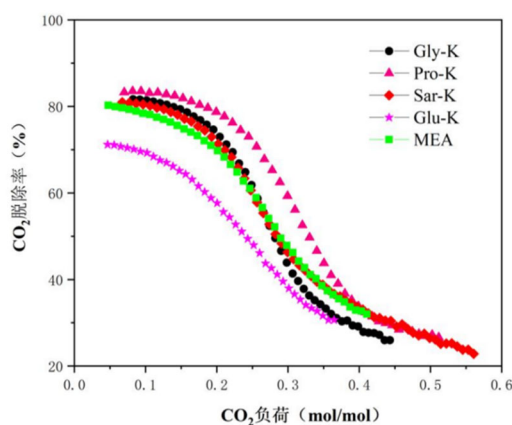


Fig. 3 Enhanced oxidative degradation reaction curve of MEA and potassium alanine [20]



($c=1 \text{ mol/L}$, $T=30^\circ\text{C}$, $V_L=85 \text{ mL/min}$, $V_G=0.25 \text{ L/min}$, $C_{in}=40\%$)

Fig. 4 The change of removal rate of five absorbents under different carbon dioxide loads[21]

When compared to ethanolamine absorbents, amino acid salts have the advantages of good CO_2 capture efficiency under low load, high environmental compatibility, low volatility, degradability, corrosiveness, and toxicity, and their acquisition methods do not rely on fossil resources, which compensates for some shortcomings of traditional organic amine absorbents, but there are still some shortcomings such as high energy consumption, poor thermal stability, and poor toxicity.

4. ionic liquid absorbent

Blanchard et al. reported in Nature in 1999 for the first time that CO_2 had a solubility of 0.6mol/L in conventional ionic liquid 1-butyl-3-methylimidazolium hexafluorophosphate ($[\text{Bmim}][\text{PF}_6]$) at 298.2K, sparking a heated debate and widespread concern among scholars at home and abroad about CO_2 capture by ionic liquid. This leads people to believe that an ionic liquid with adjustable acid-base polarity, coordination of cation and anion, and good thermal stability has a lot of application potential in carbon dioxide absorption and separation. Ionic liquid is an absorbent with a low analytical energy consumption and no heat of vaporization. When different anions and cations adjust the acidity and alkalinity of the liquid, it can precisely identify CO_2 molecules and interact with them via hydrogen bonds, resulting in high-efficiency and high-selectivity separation [22].

Ionic liquids can absorb carbon dioxide based on their anions, which include halogen, boric acid, carboxylic acid, and amino acid ions. Conventional ionic liquid absorbs carbon dioxide primarily

through physical interactions between anion and carbon dioxide, such as van der Waals force and hydrogen bonding, making it more susceptible to physical absorption and easy to desorb. At the same time, while traditional ionic liquids have good thermal stability, their viscosity is high, and the hydrogen bond network structure formed during the absorption process will also increase viscosity, affecting the mass transfer effect. Imidazole ionic liquid, in particular, introduces basic functional groups into its structure, which can significantly improve CO₂ capture capacity. Idazole ionic liquid has received relatively more attention due to its high alkalinity, ease of alkylation, and relative fluidity [23]. The following sections will go over some ionic liquid functions.

Bates et al. [24] designed and developed imidazole ionic liquid 1-(1-aminopropyl)-3-butylimidazole tetrafluoroborate (^[NH₂p-bim][BF₄]) containing primary amine groups in 2002, and discovered that 1 mol of ionic liquid can capture 0.5 mol of CO₂ at 22°C and 0.1 MPa, far outperforming conventional ionic liquid absorption. Since then, many researchers have attempted to introduce amino groups into cations, developing ionic liquids with single and double amino cation functions to study the properties of carbon dioxide capture. SISTLA [25] also compared the properties of various amino acids in anionic ionic liquid, but these failed to solve the ionic liquid's high viscosity problem well. And, because imidazole is a toxic substance, the preparation process of functional ionic liquid with amino group is still not environmentally friendly.

5. Phase change absorbent

In essence, after CO₂ is absorbed by absorbent in liquid state, a two-phase system rich in CO₂ and poor in CO₂ is formed, which has the advantages of low energy consumption, large circulation capacity, and easy separation of substances formed after CO₂ absorption. For example, after absorbing CO₂, inorganic alkaline solutions such as NaOH and KOH generate Na₂CO₃ and K₂CO₃ respectively, but their water solubility is high, so NaOH and KOH are phase change absorbent. When combined with Ca(OH)₂, soluble Na₂CO₃ and K₂CO₃ are transformed into CaCO₃ and precipitated, making subsequent separation and recycling easier. As a result, the combination of Na₂CO₃ and K₂CO₃ with Ca(OH)₂ can be classified as a liquid-solid phase change CO₂ absorbent. CaCO₃ must be calcined at around 900°C to release CO₂, which requires a lot of energy, so its practical application is still limited.

A new type of phase change CO₂ absorbent has gradually entered the researchers' field of vision in recent years. Hossain's group at Jackson State University reported a compound with six urea structural units [27], and discovered that after being mixed with a dimethyl sulfoxide solution of basic tetra-n-butyl ammonium fluoride, the compound can directly capture CO₂ in the air, forming a clathrate complex and precipitating it from the solution, indicating that its CO₂ absorption process has phase change characteristics. After being treated with methanol and water, the obtained complex can release CO₂ and regenerate absorbent (1:1). Custelcean of Oak Ridge National Laboratory reported in 2017 that a small molecule compound (PyBIG) with pyridine and guanidine structure was discovered, which can directly absorb CO₂ in solution state and form an insoluble hydrated carbonate [27], so the absorbent also has phase change characteristics and is very beneficial to subsequent operations such as carbon dioxide capture, transportation, and storage. Without a doubt, this study contributes a significant and novel idea to the research and development of a new generation of CO₂ absorbents. However, the key intermediate used in this study, 2,6-pyridine dicarbaldehyde, is very expensive, and the starting material for the synthesis of this intermediate is 2,6-dimethylpyridine, a by-product of coal tar, which belongs to a type of fossil resource with less content, which must also be included in the consumption of fossil resources and the CO₂ emissions caused by the preparation of absorbent.

In fact, the mixed solution of 2 mol/L 1,4-butanediamine (BDA) with 4 mol/L N,N-diethylethanolamine (DEEA) and some of the ionic liquid-organic amines mentioned above also belongs to phase change absorption because they will all be stratified after absorbing carbon dioxide. When studying the ionic solution-organic amine compound solution, Xu Lingjun and colleagues chose ^[Bmim][BF₄] ionic liquid, which was the first reported and has a relatively skillful synthesis

technique, and the most widely used organic amine MEA, and discovered that the influencing factor of this type of two-phase absorbent to reduce energy consumption lies in the enrichment of reaction products such as carbamate, which is an important condition to prove that this system is a 1 Zhou Haicheng and colleagues synthesized seven different phase change absorption systems, including $[\text{TETA}]\text{Br}$ and $[\text{TETA}][\text{BF}_4]$, and discovered that the $[\text{TETA}]\text{Br}$ -PMDETA- H_2O system had a saturated absorption capacity of 2.631 mol/L of carbon dioxide when the $[\text{TETA}]\text{Br}$ to PMDETA ratio was 3:7 and the temperature was 30°C.

6. Conclusion

The inorganic absorbent represented by the hot potassium carbonate solution process has low degradability and toxicity, as well as low energy consumption for regeneration, of the several chemical absorbents mentioned in this paper. However, in order to improve its reaction rate, additional activators must be added, taking into account industrial production efficiency and economic efficiency. Organic amine solution has the potential to be widely used for industrial carbon dioxide absorption due to its advantages of fast absorption rate and low cost, but its disadvantages of significant energy loss and corrosive reaction products must be addressed. Although amino acid salts have high environmental compatibility and low corrosiveness and toxicity, they do have some drawbacks, including high energy consumption in regeneration and release and a poor recycling effect. Ionic liquid absorbent, which has received a lot of attention in recent years, has a good absorption effect and regeneration ability in terms of chemical properties. In terms of physical properties, many ionic liquids have issues with high viscosity and low environmental compatibility. There is also a type of ammonia absorbent with high absorption capacity in the chemical absorption method, but the volatile nature of ammonia is well known, and it is also not conducive to the carbon dioxide capture process, so this paper does not go into detail about this absorbent. Each type of chemical absorbent has different advantages and disadvantages despite having similar reaction mechanisms. As a result, many researchers both at home and abroad frequently introduce another absorbent or activator to complement one another when improving the performance of a specific type of absorbent. One after the other, compound systems such as hot potassium carbonate solution process-organic amine, hot potassium carbonate solution process-amino acid salt, and ionic solution-organic amine are emerging. The phase change systems formed by these phase change absorbents after compounding are also very beneficial to carbon dioxide capture post-treatment, which has great development potential. Continuous research will provide strong support for the future development of CCUS technology.

Furthermore, the performance of chemical absorbents is, of course, a major factor influencing the work of carbon dioxide capture, but in order to achieve the dual-carbon strategic goal of carbon emission reduction, it is even more important for people to have an independent vision of maintaining ecological health and adhering to green environmental protection, and the relevant departments must carry out the idea of "adhering to sustainable development" to the end, taking into account While improving the efficiency of conventional energy sources, we will aggressively develop renewable energy sources such as solar and wind energy, reducing our reliance on and consumption of fossil fuels. In the future, we must broaden the scope of carbon dioxide utilization and transformation, create a multi-faceted and all-around industrial system, and truly realize negative carbon dioxide emissions.

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