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Advance in Post-Combustion CO_2 Capture with Alkaline Solution: A Brief Review

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Abstract

CO_2 emission from combustion flue gases has become the primary factor in global warming. Chemical methods of absorbing CO_2 from combustion flue gases have been widely used in engineering, because of its simple and convenient operation, high absorption efficiency and economic value. This paper analyses the domestic and foreign researches on chemical methods of absorbing CO_2 from combustion flue gases, and summaries the effect of the reactors structure, absorbent type and operating conditions on absorption efficiency. This article also forecasts present developmental directions, and expects to have a certain reference to later research and development.

Key word: CO2, reactors structure, absorbent type, operating conditions, absorption efficiency

1. Introduction

The global warming and greenhouse effect have become serious global environmental issues. The main reason of issues is the carbon dioxide produced in the energy utilization and emited directly by human beings[1]. The volume percentage of content of carbon dioxide in the atmosphere has increased from 2.84×10^{-4} before the industrial revolution to 3.56×10^{-4}[2]. Therefore, the stability, safety and environment acceptability of CO_2 capture and storage (CCS) technologies have been paid worldwide attention. These technologies include the chemical absorption and adsorption methods[3], membrane separation[4] and chemical looping combustion[5], underground storage technology[6], terrestrial vegetation[7] and marine microalgae fixation[8]. The chemical method has been widely used due to its advantages of simple operation, high absorption efficiency, high economic value and mature technology etc [9]. The chemical method is one of the most widespread methods of removing CO_2 from combustion flue gases in the industry process at present. The CO_2 absorption efficiency in the tower is affected by two

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factors, residence time and absorption rate[10]. There are two aspects to reduce the project cost and increase absorption efficiency. One is to select proper absorbent,and another is to improve the absorption equipment. In this paper, the progress of chemical method were summarized and analyzed, indcluding reactor structure effect on absorption performance, absorbent type effect on the absorption performance, and operating conditions effect on the absorption efficiency.

2. Advantages of CO_2 chemical absorption

CO_2 absorption technology has been widely used due to its high efficiency, low energy consumption, environmental friendly etc. Among them, biological, physical and chemical methods are the most representative. As to the biological method, CO_2 is fixed and absorbed by photosynthesis of plants, alga and photosynthetic bacteria. CO_2 is fixed by photosynthesis with no energy consumption which is a kind of energy conservation. But its capacity and absorption efficiency is low. Physical method is utilizing organic solution to absorb and remove the acid gases (CO_2, SO_2, H_2S etc.) with the pressurized condition in the CO_2 absorption from combustion flue gases. There is no chemical reaction in the whole process. The key of this method is to determine the characteristics of absorbent. Compared with the biological method, the physical method could get high CO_2 absorption efficiency, but the costs of production and requirement of absorbent is higher. In the chemical absorption method, CO_2 is removed by the chemical reaction of absorbent with CO_2 from flue gases. This method has the advantages of high absorption efficiency, high economic value, simple operation and mature technology and so on, but the causticity and renewable energy consume of the absorbent obstruct its development. The chemical absorption method is important in the CCS technology, but the absorption performance is required to be improved.

3. Reactor structure effect on absorption performance

Domestic and foreign researchers utilized many kinds of reactor, and obtained different CO_2 absorption efficiencies by chemical method. The CO_2 absorption properties in different reactors structure by different researchers are summarized in Table 1.

Table 1. Comparison of typical reactors for CO_2 absorption

Reactor type	Experimental parameters					References
	Concentration of absorbent	Flowrate of absorbent	Concentration of CO_2	Flowrate of flue gas	Operating temperature	
Higee	28%	/	19%	/	/	[13]
	15%	0.6L/min	12%	0.4m³/min	25℃	[14]
	0.5mol/L	/	1.5%	/	/	[15]
Packed tower	/	/	/	/	25℃	[18]
	0.02-0.50mol/L	/	0.03%	/	25℃	[19]
	3.15mol/L	166752m³/h	/	/	/	[21]
Spray tower	2-6%	1.02-3.06m³.h⁻¹	5-15%	61-214m³.h⁻¹	15-55℃	[23]
	/	/	/	45m³/h	/	[24]
	0.8-10%	120-200ml/min	7-15%	7.6-24.7L/min	28-28℃	[25]

The objects of all researchers are different, and the parameters are not comparable, but we still can summarize the properties of reactors. Higee combines chemical absorption and ultra-weight technology to enhance the mass transfer performance, improve absorption efficiency and reduce costs. Higee has the advantages of high mass transfer efficiency, small size, large capacity, low fluid pan-point and energy consumption[11,12].Packed tower enhances the mass transfer performance and increases the tower production capacity mainly by increasing surface area and porosity, or by changing the gas-liquid two-phase approach way to improve the gas-liquid two-phase turbulent intensity, which can improve the quality of two-phase effect. Packed bed has the advantages of simple manufacture, wide range of material adaptability, adaptable, low pressure drop and high heat and mass transfer efficiency, but its flow and distribution is irregular[16,17,20,22].Researchers used different methods to improve the absorption properties of the spray tower by changing its liquid flow. Spray tower is widely used in desulfurization and denitration out of power plant flue gases for its simple structure, small air resistance.Moreover it does not blocked the system when it handled with the matter with particles or sludge mixture.

Researchers continue to explore various new towers to remove CO_2 from combustion flue gases more effectively. Researchers still devote themselves to exploring new towers. Franz[26], Burges & Calderbank[27], An et al.[28], Serizawa[29] researched the characteristics of bubble column, Tan et al.[30-32]researched the characteristics of gas purification in the plate tower. Lin et al.[33] used NaOH to absorb CO_2 in rotate packed bed. Yan et al.[34] researched the hydrodynamics of the plate bubble column. Han et al.[35] compared the packed bed, spray tower with bubble tower from the point of the mass transfer efficiency, possibility of scaling and so on. There is no fixed pattern in choosing tower in engineering application for its properties. We need to constantly explore new towers to obtain high CO_2 absorption efficiency from combustion flue gases and improve mass transfer efficiency.

4. Absorbent type effect on the absorption performance

Chemical absorption method refers to use liquid solution (generally is alkaline solution) to selectively remove combustion flue gas component which is easily soluble in absorbent by chemical reactions. Removing CO_2 from combustion flue gases by chemical method usually uses ethanol amine, ammonia, alkaline solution as absorbents. In recent years, some domestic and foreign researchers have deeply researched amine solution, ammonia, alkaline solution as absorbents to absorb CO_2 from combustion flue gases from the aspects of the mechanism of it, mass transfer coefficient, absorption efficiency, and so on. This paper analyses and compares them separately, as shown in Table 2, and summaries the kinetics of ammonia absorbing CO_2 from combustion flue gases, as shown in Table 3.

Table 2. Types of absorbent effect on the absorption properties

Absorbent type	Reaction expression	Absorption efficiency	References
Amine solution (MEA,DEA etc.)	$CO_2+2R_1R_2N \Leftrightarrow R_1R_2NH^+_2 + R_1R_2NCOO$	61%-90%	[36-38]
Ammonia solution	$NH_4OH+CO_2 \Leftrightarrow (NH4)_2CO_3+H_2O$ $(NH4)_2CO_3+CO_2+H_2O \Leftrightarrow 2NH_4HCO_3$	78%-98%	[39-45]
Alkali solution (NaOH, KOH etc.)	$H_2O+CO_2 \rightarrow HCO_3^-+H^+$ $OH^-+CO_2 \rightarrow HCO_3^-$ $HCO_3^-+OH^- \rightarrow CO_3^{2-}+H_2O$	92%-99%	[33, 50-54]

Table 3. Kinetics of ammonia absorbing CO_2

Reactor	Reaction rate constant	Activation energy J/mol	Reference
/	$\lg k=11.13-2535/T$	48567	[46]
Wetted-wall tower	$\lg k=10.2+2280/T$	43668	[47]
Sieve-tray tower	$\lg k=5.38-1395/T$	26730	[42]
Stirrer	$\lg k=11.09-2506/T$	48000	[48]
Disc tower	$\lg k=11.23-2550/T$	/	[49]

MEA has the advantages of absorbing low concentration CO_2 from combustion flue gases for its small molecular weight and large ability to absorb acid gases, absorbent can be recycled under certain conditions, but the absorption efficiency is not high in the circular process, and absorption ability of absorbent also decreased for some irreversible reaction reasons. Degradation products in rich fluid may cause system corrosion which increases operating costs. Before removing CO_2 from combustion flue gases, desulfurization and denitration are required, and CO_2 absorption capacity is low. Compared with MEA solution absorbing CO_2, ammonia has advantages of higher absorption capacity, removing pollutants in combustion flue gases combined, and having less corrosion to equipment. The product can be used to produce fertilizer; the renewable energy consumption is low. However, absorbent is easily volatile and the products are unstable, which are the bottlenecks in the development of ammonia used in absorbing CO_2 from combustion flue gases. NaOH solution has been concerned by many researchers, because it can easily react with CO_2 as alkali solution, and high absorption efficiency.And also because it is alkali solution which has strong corrosion, resulting in higher operating cost in engineering application.

Furthermore, the product of NaOH solution reaction with CO_2 is not recyclable, absorbent is non-renewable and other factors limited its development.

In the research of using chemical method to absorb CO_2 from combustion flue gases, absorbent kinds trend to diversification.Table 2 and Table 3 only list part of researches of using alcohol amine, ammonia, NaOH solution to absorb CO_2 from combustion flue gases. Haghnegahdar et al.[55] used $Ca(OH)_2$ to absorb CO_2 in the jet stream bed. Espen et al.[56]researched the process of the mixture of alkaline solution and organic solvent enhanced absorb CO_2 experiment. Constantly exploring new absorbent is also a effective measure to enhance the heat and mass transfer between the absorbent and CO_2, and improve the absorption efficiency.

5. Operating conditions effect on the absorption efficiency

5.1Absorbent concentration

Fig. 1 selected seven references' datum, which are not comparable with each other. Because each reference adopts reactor type, operating conditions and absorbent type are not same. However, we can find a trend from figure1that CO_2 absorption efficiency increases with the concentration of absorbent increasing, then keep stabilized. When the absorbent concentration is low, increasing absorbent concentration, CO_2 absorption efficiency increment is large. However, when the absorbent concentration is high, CO_2 absorption efficiency increment is not obvious. These phenomenons are determined by reversible equilibrium conditions and gas-liquid two-phase mass transfer conditions. In the view of chemical dynamics, increasing absorbent concentration is equivalent to increasing the reactant concentration, resulting in response moving to the positive direction, improving the reaction rate and CO_2 absorption efficiency. Thus, in different operating conditions, it is feasible to improve CO_2 absorption efficiency by increasing the absorption concentration.

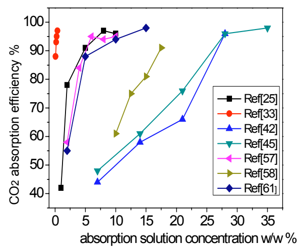

Fig. 1. Absorbent concentration effect on absorption efficiency

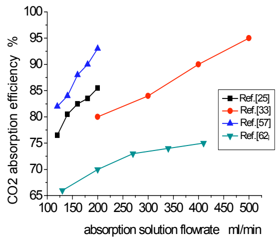

Fig. 2. Aabsorbent flowrate effect on absorption efficiency

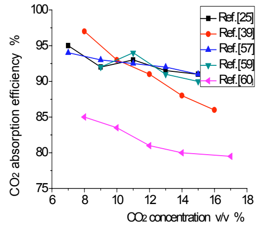

Fig. 3. CO_2 concentration effect on absorption efficiency

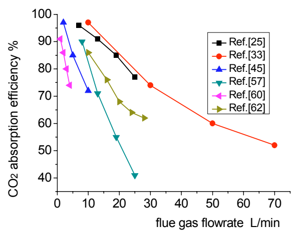

Fig. 4. Flue gas flow rate effect on absorption efficiency

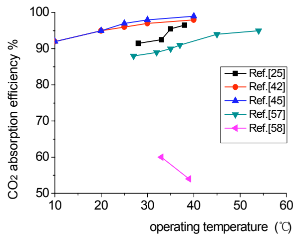

Fig. 5. Operating temperature effect on absorption efficiency

5.2 Absorbent flow rate

As can be seen from Fig. 2, within the changing scope, CO_2 absorption efficiency is also increasing as the absorbent flow rate increases. As the shown figure, CO_2 absorption efficiency increment is more than 20%.Increasing the absorbent volume flow rate is equivalent to increasing gas-liquid two-phase area of unit volume, enhancing gas-liquid mass transfer rate, improving the CO_2 absorption efficiency. While increasing the absorbent flow rate makes the droplets residence time cut down, reducing the CO_2 absorption efficiency. Synthesizing two aspects of effect, CO_2 absorption efficiency increases with absorbent flow rate increasing. Therefore, in certain experimental conditions, it is feasible to improve CO_2 absorption efficiency by increasing the absorbent flow rate, but the absorbent utilization still should be taken into account.

5.3 CO_2 concentration

It can be seen from Fig. 3, CO_2 volume concentration changes in the range of 5% to 20%. As the CO_2 concentration increases, CO_2 absorption efficiency decreases, but the declination is small. This is mainly for two reasons: on the one hand, as the CO_2 concentration increases, the unit volume of gas-liquid contact time is shorten in the tower; on the other hand, increasing CO_2 concentration promotes the reaction rate of CO_2 absorption in a certain extent. Synthetically analysing these two factors, we know CO_2 absorption efficiency decreases slowly with the CO_2 concentration increasing.

5.4 Flue gases flow rate

As can be seen from Fig. 4, in the flue gas flow rate change scope, CO_2 absorption efficiency decreases with the flue gas flow rate increasing, and the reduction of the CO_2 absorption reaches about 50%.Increasing flue gas flow rate is equivalent to reducing the gas-liquid two-phase residence time in the tower, resulting in slowing down the reaction rate of CO_2 absorption; and increasing the flue gas flow rate will carry spray absorbent out from the tower, leading to the loss of absorbent. Therefore, increasing flue gas flow rate will reduce the CO_2 absorption efficiency.

5.5 Operating temperature

Fig. 5 reflects the operating temperature effects on the CO_2 absorption efficiency. It can be seen from the figure, CO_2 absorption efficiency increases with the operating temperature increasing.CO_2 can react with absorbent at different temperature and pressure. Generally speaking, the higher the operating temperature is, the faster the chemical reaction rate is, the higher the mass transfer efficiency and the CO_2 absorption efficiency are. The figure shows the optimum operating temperature is in the range of 28-32℃. Appropriately increasing the operating temperature is also an effective method of improving CO_2 absorption efficiency.

CO_2 absorption efficiency is not only determined by absorbent concentration, absorbent flow rate, flue gases flowrate,CO_2 concentration and operating temperature, but also by tower height, swirl, nozzle type and other factors.

6. Conclusion

Chemical methods have become the primary means of absorbing CO_2 in the combustion flue gases, owing to its advantages of economy and technology. On the current status of research, the author believes that the following issues need further exploration:

(1) Combine removal of other pollutants in the flue gas system: SO_X, NO_X, HCI and other acid gases, as well as Hg, As and other trace heavy metals in the combustion flue gases removal mechanism.

(2) Absorbent renewable energy, product recycling and thermal stability study.

(3) Build the mathematical model of alkaline solution to absorb CO_2 in flue gases.

(4) Combine the absorption of CO_2 with the re-use

(5) explore new means to strengthen the absorption, such as physical-chemical method.

In a word, deeper explore alkaline solution absorbing CO2 in the combustion flue gases has a certain significance to control greenhouse-effect.

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