Experimental studies on removal capacity of carbon dioxide by a packed reactor and a spray column using aqueous ammonia

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Abstract

The absorption of carbon dioxide into ammonia aqueous solution using packed reactor and spray column was investigated. The removal efficiencies were evaluated over ranges of main operating variables, including CO₂ inlet concentration, gas flow rate, liquid flow rate, concentration of ammonia and temperature. Experimental results showed that the suitable reaction temperatures are 40°C and 35°C for spray column and packed reactor respectively, and the mole ratio of NH₃ to CO₂ is a key parameter and plays an important role on CO₂ removal efficiency. Compared with the absorption of CO₂ in the spray column, the CO₂ removal efficiency in the packed reactor shows superior performance.

Keywords: Carbon dioxide; aqueous ammonia; removal efficiency; packed reactor; spray column

1. Introduction

It is well known that carbon dioxide is the major greenhouse gas of which emission should be reduced. Chemical absorption process is generally recognized as the most effective technology, and the monoethanolamine (MEA) scrubbing is widely used in the chemical engineering process of carbon dioxide capture [1, 2]. However, the MEA process suffers the following disadvantages [3], such as low CO₂ loading capacity, high equipment corrosion rate, amine degradation by SO₂, NO₂, HCl and O₂ in the flue gas, high energy consumption during absorption and regeneration.

Some researchers found that ammonia seems to be an alternative and promising absorbent for removing CO₂ from flue gas [4-9]. The absorbent of aqua ammonia is not easy to be degraded and the energy input is much lower than MEA process, and the solution does not have a corrosion problem. Three major acid gases, SO₂, NO₂ and CO₂, will be captured in the aqua ammonia process, which is expected to reduce the total cost and complexity of emission control systems [9]. Bai et al. [4, 6] carried out experimental investigations of the ammonia and MEA capturing CO₂ in a bubble reactor. The tests showed that the ammonia is superior to MEA absorbent in its capacity to absorb and removal CO₂ from flue gas systems. Yeh et al. [10] performed CO₂ absorption and regeneration with aqueous ammonia...
ammonia in a semi-continuous flow reactor. It was found that the energy requirement for the regeneration of NH₃ was less than that for the regeneration of MEA reagent. Diao et al. [11] studied the mechanism and kinetics of the reaction between CO₂ and NH₃ solvent in a sieve-plate tower. Their experiment results showed that the CO₂ removal efficiency reached the highest at 33°C. Niu et al. [12] studied the carbon dioxide absorption into ammonia using a spray column, the experimental results showed that the ammonia concentration played a very important role, when the ammonia concentration was 8%, the removal efficiency of carbon dioxide could realize to 98.4%.

In order to improve the carbon dioxide removal efficiency and reduce the cost, apart from choosing good absorbing liquid, it is very important to select effective reactor and proper operating conditions. The absorption of CO₂ using packed reactor is a well-established technology being used widely in the chemical industrial. However, packed reactor suffer from various operational problems including high gas phase pressure drop, liquid channelling and flooding, disintegration of packing materials caused by high temperature gases, and deposition on packings and clog up of void spaces by solid-laden gases [13]. Thus, many experimental studies focused on the development of innovative configurations of scrubbing and stripping equipment. Kuntz et al. [14] compared the mass-transfer efficiency of spray column and packed reactor for carbon dioxide absorption into MEA, the experimental results showed that the spray column was capable of removing CO₂ from gas stream at a higher rate than the packed column.

In this work, the absorption of carbon dioxide into aqueous ammonia was carried out under various conditions to reveal effects of process parameters, including CO₂ inlet concentration, gas flow rate, liquid flow rate, concentration of ammonia and temperature. And the removal capacity of CO₂ in the packed reactor was compared to that in the spray column.

2. Absorption experiment

The gas absorption experimental setup is shown in Figure 1. The experiments were carried out in spray column and packed reactor respectively. The packed reactor and the spray column were made of stainless steel with 100 mm inner diameter and 500 mm height. In order to keep the reaction temperature, the experiments were carried out under water bath conditions. The water bath was kept at constant temperature by the temperature controller and an electric heater. And also, the aqueous ammonia solution and inlet artificial flue gas of CO₂ and nitrogen mixture were kept at the same temperature with the water bath. The mixture of CO₂ and nitrogen was fed into the packed reactor or the spray column from its bottom.

Figure 1 Schematic diagram of experimental setup of CO₂ and aqueous ammonia absorption
For the spray column, the heated aqueous ammonia solution was sprayed into the column at its upper part. For the packed reactor, the heated aqueous ammonia solution was injected into the packed reactor from its top to the bottom. So, the CO₂ gas and the aqueous ammonia solution were in counter flow pattern, it is beneficial to make CO₂ and aqueous ammonia contact and react thoroughly. As for the spray column reactor, the Sauter Mean Diameters (SMD) of the aqueous ammonia spray were 30/80 μm to 40/80 μm when the pressure of the pump was 0.69 MPa to 1.11 MPa. In the packed reactor, the packing height of the reactor was 400 mm, and the filler of the packed column was ceramic Raschig ring with 8 mm inner diameter.

Effects of different operating and design parameters on CO₂ removal efficiency were investigated. Detailed parameters in experiments are given in Table 1.

Table 1 Experimental parameters

<table>
<thead>
<tr>
<th>Temperature, °C</th>
<th>Ammonia flow rate, L/h</th>
<th>Ammonia concentration, %, w/w</th>
<th>Total gas flow rate, L/min</th>
<th>CO₂ inlet concentration, %, v/v</th>
</tr>
</thead>
<tbody>
<tr>
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<td>8</td>
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<td>10</td>
<td>15</td>
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<td>8-24</td>
<td>8</td>
<td>20</td>
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<td>12</td>
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<td>15</td>
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<td>16</td>
<td>8</td>
<td>10-28</td>
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<td>20</td>
<td>8</td>
<td>8</td>
<td>20</td>
<td>5-15</td>
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3. Results and discussions

The removal efficiency (η) is determined from the difference between the amounts of CO₂ entering and leaving the column, which can be expressed as

$$\eta = \frac{Y_1 - Y_2}{Y_1} \times 100\%,$$  \hspace{1cm} (1)

where $Y_1$ and $Y_2$ stand for the mole ratios of CO₂ to N₂ of the gas phase entering and leaving the absorption column respectively. And the mole ratio of gas phase CO₂, $Y$, can be expressed as

$$Y = \frac{y}{1 - y},$$  \hspace{1cm} (2)

where $y$ is the mole fractions of gas phase CO₂. Finally, the CO₂ removal efficiency can be determined as follows

$$\eta = \frac{y_1 - y_2}{y_1(1 - y_2)} \times 100\%,$$  \hspace{1cm} (3)

where $y_1$ and $y_2$ present the mole fractions of gas phase CO₂ entering and leaving the absorption column respectively.

3.1. Effect of the operating temperature

Figure 2 shows the CO₂ removal efficiency profile under different temperatures in the packed reactor and the spray column. In these cases, the mass concentration of aqueous ammonia solution was 8%, the aqueous ammonia flow rate was 8 L/h, the total gas flow rate was 10 L/min, and the concentration of CO₂ at the inlet was 15%. With the temperature increasing from 20°C to 40°C, experimental results showed that the CO₂ removal efficiency increased from 82.47% to 90.20% in the spray column. Whereas, when the temperature is higher than 40°C, the CO₂ removal efficiency begins to decline. As for the packed reactor, the CO₂ removal efficiency increased from 94.28% to 97.72% when the temperature increasing from 20°C to 35°C, and the CO₂ removal efficiency was reduced when the temperature was higher than 35°C. The temperature plays an important role in diffusion and chemical reactions. The diffusion rate increases when the temperature increasing, which gives rise to the acceleration the absorption
process. However, the increase of the temperature also accelerates the decomposition of the reaction product of ammonium bicarbonate, this gives rise to reduction of the absorption efficiency. Experimental results indicated that the suitable reaction temperatures are 40°C and 35°C for spray column and packed reactor respectively. This phenomenon can be attributed to that the reaction of CO₂ absorption into ammonia aqueous is reversible, the forward reactions are dominant at room temperature, the backward reactions occur at temperatures of around 38°C ~60°C [4,15].

![Figure 2 Effect of temperature in the reactor on CO₂ removal efficiency](image1)

**Figure 2** Effect of temperature in the reactor on CO₂ removal efficiency

![Figure 3 Effect of ammonia volume flow rate on CO₂ removal efficiency](image2)

**Figure 3** Effect of ammonia volume flow rate on CO₂ removal efficiency

### 3.2. Effect of the aqueous ammonia flow rate

The influence of aqueous ammonia solution flow rate on the CO₂ removal efficiency was investigated. Figure 3 shows the CO₂ removal efficiency profile at different aqueous ammonia solution flow rates. In these cases, the mass concentration of aqueous ammonia solution was 8%, the total gas flow rate was 20 L/min, the concentration of CO₂ at the inlet was 15%, and the temperature was 20°C. When the aqueous ammonia flow rate increasing from 8 L/h to 24 L/h, the CO₂ removal efficiency increased from 66.38 % to 73.92% in the spray column. In the packed reactor, the CO₂ removal efficiency increased from 92.54% to 96.01% when the aqueous ammonia flow rate increasing from 8 L/h to 24 L/h.

With the aqueous ammonia flow rate increasing from 8 L/h to 24 L/h, the mole ratios of ammonia to carbon dioxide increased from 4.6 to 13.8, this indicated that there would be more ammonia molecules can participate in the reaction of CO₂ absorption. Besides, the interfacial area per unit volume increases with increasing aqueous ammonia flow rate, which is beneficial to enhance absorption ability.

### 3.3. Effect of the aqueous ammonia concentration

Figure 4 shows the CO₂ removal efficiency profile under different concentrations of aqueous ammonia solution. In these cases, the operating temperature was 20°C, the aqueous ammonia flow rate was 12 L/h, the total gas flow rate was 20 L/min, and the concentration of CO₂ at the inlet was 15%. In the spray column, the CO₂ removal efficiency increased from 38.43 % to 86.07% when the concentration of aqueous ammonia solution increasing from 2% to 16%. In the packed reactor, the CO₂ removal efficiency increased rapidly from 52.73% to 94.28% when the value of concentration of aqueous ammonia solution increasing from 2% to 8%. When the concentration of aqueous ammonia solution was higher than 8%, the CO₂ removal efficiency increased slightly. The CO₂ removal efficiency increased to 99.43% when the ammonia concentration was 16% in the packed reactor.

The mole ratios of ammonia to carbon dioxide increase with the ammonia concentration, this means that increasing ammonia concentration yields a amount of the active ammonia available to diffuse toward the gas-liquid interface and to react with CO₂, this will promote the enhancement factor, which leads to a higher absorption rate.

### 3.4. Effect of the total gas flow rate

The effect of the total gas flow rate on the CO₂ removal efficiency using aqueous ammonia solution is shown in Figure 5. The operating temperature was 20°C, the aqueous ammonia flow rate was 16 L/h, the mass concentration...
of aqueous ammonia solution was 8%, and the concentration of CO₂ at the inlet was 15%. With the total gas flow rate increasing from 10 L/min to 28 L/min, experimental results showed that the CO₂ removal efficiency decreased from 98.29% to 91.37% in packed column. In the spray reactor, the CO₂ removal efficiency decreased from 86.07% to 63.19%. The effect of the gas flow rate on the CO₂ removal efficiency in the spray reactor is greater than that in the packed column.

Increase in the gas flow rate leads to an increase of volumetric overall mass transfer coefficients which gives rise to the absorption rate increasing. However, the mole ratios of ammonia to carbon dioxide decreases from 18.4 to 6.6 as the total gas flow rate increasing from 10 L/min to 28 L/min, this is the main reason of the reduction of removal rate. Thus, the mole ratio of NH₃ to CO₂ flow rate is a key parameter and plays an important role on CO₂ removal efficiency.

3.5. Effect of the inlet concentration of carbon dioxide

The influence of CO₂ inlet concentration on the CO₂ removal efficiency was also investigated. Figure 6 shows the CO₂ removal efficiency profile when the inlet concentration of CO₂ increased from 5% to 15%. According to two-film theory, the gas phase driving force and the gas phase mass transfer coefficient will increase with the increasing CO₂ partial pressure, which is beneficial to enhance absorption rate. Logically, an increase in the CO₂ partial pressure allows more CO₂ molecules to travel from gas bulk to the gas-liquid interface, which would result in higher removal efficiency. Whereas, the mole ratios of ammonia to carbon dioxide decreased from 13.78 to 4.59 as the CO₂ inlet concentration increasing from 5% to 15%, which gave rise to the reduction of removal efficiency. Thus, the CO₂ removal efficiency decreased a little with the inlet concentration of CO₂ increasing both in the packed reactor and spray column.
3.6. Effect of the mole ratio of ammonia to carbon dioxide

According to the experimental parameters, mole ratios of NH$_3$ to CO$_2$ were calculated. Figure 7 shows the effect of mole ratio of NH$_3$ to CO$_2$ on CO$_2$ removal efficiency. It can be seen that the mole ratio of NH$_3$ to CO$_2$ is a key parameter when comparing CO$_2$ removal efficiency at different experimental conditions. With the mole ratio of NH$_3$ to CO$_2$ increasing from 2.3 to 7.7, the CO$_2$ removal efficiency increased from 52.73% to 93.12% in the packed reactor, and the CO$_2$ removal efficiency increased from 38.43% to 86.07% in the spray column. However, under the larger values of the mole ratio of NH$_3$ to CO$_2$, the growth rate of the CO$_2$ removal efficiency is changing slowly, especially when the mole ratio of NH$_3$ to CO$_2$ exceeds 9.2 in packed column.

4. Conclusions

The removal efficiencies for CO$_2$ absorption into aqueous ammonia were investigated in both of a spray column and a packed reactor. The CO$_2$ removal efficiency increases with the concentration of aqueous ammonia solution and the aqueous ammonia flow rate, and decreases with increasing gas flow rate. Experimental results indicated that the temperature plays an important role in CO$_2$ absorption into aqueous ammonia, and the suitable reaction temperatures are 40°C and 35°C for the spray column and the packed reactor respectively. Experimental results also showed that the mole ratio of NH$_3$ to CO$_2$ flow rate is a key parameter and plays an important role on CO$_2$ removal efficiency. Compared with the spray column, the removal efficiency for CO$_2$ absorption into aqueous ammonia in the packed reactor shows superior performance.

Acknowledgment

This research was supported by Beijing Municipal Commission for Science & Technology under Grant No.Z08040902950803.

References