Study on CO$_2$ capture by AAILs-MDEA aqueous solution in packed tower

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Abstract. This study mainly uses N-methyldiethanolamine (MDEA) as the main body of absorption, and uses three kinds of AAILs(AAILs), 1-butyl-3-methylimidazolium lysinate ([Bmim][Lys]), tetramethylammonium glycinate ([N1111][Gly]) and 1-butyl-3-methylimidazolium glycinate ([Bmim][Gly]) as accelerator, respectively. That forms a new blended aqueous solution for CO$_2$ removal. The new AAILs-MDEA aqueous solution absorbent has stronger ability to absorb CO$_2$, faster absorption rate and lower equipment corrosion. When the gas flow rate is 500ml/min and the liquid flow rate is 150ml/min, the best removal efficiency is achieved. At 313.2K, the removal efficiency of MDEA-[N1111][Gly] absorbent can reach 100%; the effect of the three new alkanolamine aqueous solutions on CO$_2$ absorption is: MDEA-[N1111][Gly]>MDEA-[Bmim][Gly]>MDEA-[Bmim][Lys].

Keywords: Packed tower, CO$_2$, AAILs, alkanolamine, gas flow rate.

1. Introduction
In recent years, the increasing population of the world has led to a sharp increase in the consumption of fossil fuels, which is the main factor in the accumulation of CO$_2$ in the global atmosphere [1]. As the world enters the “coal era” and the “natural gas era”, environmental pollution is becoming more and more serious. Global warming, melting glaciers [2], become much more obvious. At present, it is difficult to reduce the CO$_2$ emissions at the root, but the capture and absorption of CO$_2$ can greatly reduce the pollution of CO$_2$ to the atmosphere.

At present, the most widely used absorption method in the world is the Alkanolamine absorption method. It has the advantages of lower cost, better absorption effect, wider use, and mature technology. In this paper, a promoter for the formation of a new complex solution was constructed, which effectively improved the absorption of CO$_2$ by the Alkanolamine method. The Alkanolamine absorption method [3-8] is an alkanolamine as an absorption main agent. MDEA in the tertiary alkanolamine is used in this paper. Compared with other alkanolamines, this solution has the advantages of large absorption, low corrosion to the instrument and easy availability of raw materials.

AAILs is composed of anions and cations which are not easy to volatilize, have low vapor pressure, have a large temperature range, are chemically stable, and can dissolve some insoluble materials and have unique chemical
Properties. It has performed well in the separation and enrichment of CO₂ [9-14]. It can chemically react with CO₂ to form a relatively stable salt, so that the reaction proceeds to the positive reaction direction, and the CO₂ can be absorbed more efficiently. It needs to be compounded with MDEA to obtain a new accelerator for absorbing CO₂. In this paper, three kinds of amino acid functional ionic liquids were mixed with methyldiethanolamine to form a new compound solution for absorbing CO₂.

2. Experiment

2.1. Experiment setup

![Figure 1. Experimental system diagram](image)

The absorption device of this paper is packed tower. It mainly composed of the tower body and the packing in the tower. As shown in Figure 1, the gas flows inlet from the bottom, and the liquid flows inlet from the top. CO₂ and N₂ are supplied by high-pressure steel cylinders, and the flow rate is controlled by a rotameter. After mixing in a gas mixture bottle, the mixture is fed into the packed tower from bottom to top. The compound solution controlled through a liquid flow meter, then passed through a peristaltic pump, absorbed CO₂ in the packed tower. After the gas mixture is absorbed, it enters the CO₂ analyzer for measurement.

2.2. Chemical analysis.

The calculation formula for the removal efficiency of CO₂ is:

\[
\eta_{CO_2} = \left[ \frac{C_0 - C_1}{C_0(1 - C_0)} \right] \times 100\% (l - l)
\]

Where \(\eta_{CO_2}\) is the CO₂ removal efficiency, \(C_0, C_1\) are the concentration of CO₂ at gas inlet and outlet of the packed tower.

The total volumetric mass transfer coefficient is calculated by:

\[
K_{Ga} = \frac{G_1}{(P*Z)(\ln(Y_1/Y_2))} + \frac{1}{Y_2}(4-2)
\]

Where \(K_{Ga}\) is the total volumetric mass transfer coefficient, \(G_1\) is the flow rate of Nitrogen, \(P\) is the total pressure of gas phase, \(Z\) is the height of the packing tower, \(Y_1, Y_2\) are the ratio of CO₂ to nitrogen inlet and outlet the tower gas.

3. Results and discussion

3.1. Influence of liquid flow rate and gas flow rate on the experiment

When the concentration of MDEA is 0.3% and the packing size is 3*3, the Experimental data is as follows:

| Liquid flow rate(ml/min) | 50  | 75  | 100 | 125 | 150 |
|-------------------------|-----|-----|-----|-----|-----|
| CO₂ content (%)         | 6.68| 6.28| 5.47| 5.21| 5.14|
Table 2. The effect of the gas flow rate on the experiment

| Gas flow rate (ml/min) | 500  | 750  | 1000 | 1250 | 1500 |
|-----------------------|------|------|------|------|------|
| CO₂ content /%        | 6.23 | 8.27 | 10.72| 10.95| 11.86|

The experimental conditions are fixed at normal temperature and pressure, and the magnetic stirrer is 1000r/min. It can be seen from Table 1 and 2 that when the gas flow rate is unchanging, the absorption effect of the alkanolamine solution on CO₂ is gradually enhanced with the increase of the liquid flow rate, and the absorption effect is best when the liquid flow rate is 150 ml/min. And when the liquid flow rate is unchanging, the absorption effect is gradually reduced with the increase of the gas flow rate, and the absorption effect is best when the gas flow rate is 500 ml/min.

3.2. The capture efficiency of CO₂ in different compound solutions

The optimal liquid flow rate is 150ml/min and the gas flow rate is 500ml/min. And the ionic liquid volume was 10g, 20g, 30g, the packing size was 3*3, 6*6. The experiment data is as Table 3-3. When the experimental condition is normal temperature and pressure, and when the magnetic stirrer rotates at 1000r/min, It can be seen from Table 3-3 that the three ionic liquids can all promote the absorption of CO₂ by the alkanolamine solution, and the effect of absorbing CO₂ is gradually enhanced with the amount of ionic liquid is increased. When the other conditions are the same and the diameter of the packing changes, the absorption efficiency of the complex solution for CO₂. When the filler diameter is increased, the absorption effect of the compound solution on CO₂ is reduce. It can be concluded that the three AAILs have different effects on the absorption of CO₂ by the aqueous solution of the alkanolamine, where MDEA-[N¹₁₁₁][Gly]>MDEA-[Bmim][Gly]>MDEA-[Bmim][Lys].

Table 3. The capture efficiency of CO₂ in different compound solutions

| Amount of ionic liquid(g) | 10  | 20  | 30  |
|--------------------------|-----|-----|-----|
| Packing size             | 3*3 | 6*6 | 3*3 | 6*6 |
| CO₂ content(%)           |     |     |     |
| [N¹₁₁₁][Gly]            | 0.95| 1.64| 0.19| 0.42| 0   | 0   |
| [Bmim][Gly]             | 1.91| 1.89| 1.24| 1.34| 0.56| 0.94|
| [Bmim][Lys]             | 1.53| 1.89| 0.74| 1.1 | 0.56| 1.17|

3.3. Removal efficiency and total mass transfer coefficient of each compound solution when the concentration of MDEA is 0.3%

Table 4. Removal efficiency and total mass transfer coefficient of MDEA-[Bmim][Lys]

| MDEA-[Bmim][Lys] | η_CO₂ | K_Ga |
|------------------|-------|------|
| 0.05             | 91.2  | 0.0941|
| 0.1              | 95.78 | 0.1211|
| 0.15             | 96.81 | 0.1314|

Table 5. Removal efficiency and total mass transfer coefficient of MDEA-[N¹₁₁₁][Gly]

| MDEA-[N¹₁₁₁][Gly] | η_CO₂ | K_Ga |
|--------------------|-------|------|
| 0.05               | 83.08 | 0.1086|
| 0.1                | 91.37 | 0.1709|
| 0.15               | 100   | 0.1975|
Table 6. Removal efficiency and total mass transfer coefficient of MDEA-[Bmim][Gly]

| MDEA-[Bmim][Gly] | η|CO₂ | KgGα |
|------------------|------------------|-----------------|
| 0.05             | 88.97            | 0.0858          |
| 0.1              | 92.89            | 0.102           |
| 0.15             | 96.81            | 0.1314          |

Figure 2. Effect of the concentration of AAILs on removal efficiency and total mass transfer coefficient

According to Tables 4, 5, 6, it can be concluded that the removal efficiency and total mass transfer coefficient of the new solution composed of different AAILs and aqueous alkanolamine solutions have a same change rule. As the liquid flow rate increases, the removal efficiency and the total mass transfer coefficient of the volume also increase too. It can be seen from Fig. 2 that the removal efficiency of CO₂ and the mass transfer coefficient of total volume increase gradually with the increase of the concentration of AAILs, and the removal efficiency of MDEA-[N_{1111}][Gly] absorbent in the packed column can reach 100% when concentration is 0.15%.

4. Conclusion
In this paper, MDEA was used as the main component of the absorption, and three kinds of AAILs [Bmim][Lys], [Bmim][Gly], [N_{1111}][Gly] were used as promoters to construct a new type of complex absorbent. Through the absorption capacity and absorption efficiency of CO₂ by three new alkanolamine complex solutions, the removal efficiency and total mass transfer coefficient were obtained. The capture effect of the absorbent on CO₂ was verified by using a small packed column. The main conclusions are as follows:

1. The new alkanolamine solution has a good absorption effect on CO₂. The addition of a small amount of AAILs can significantly increase the absorption capacity of CO₂. In this experiment, the tetramethylammonium glycinate has the highest absorption capacity for CO₂.

2. When selecting the optimal intake and intake volume, it can be clearly seen that the absorption rate gradually decreases when the amount of gas flow rate increases gradually and the liquid flow rate is unchanging. That the absorption rate gradually increases when the amount of liquid flow rate increases gradually and the gas flow rate is unchanging.

3. The removal efficiency of CO₂ increases with the increase of the concentration of AAILs, and decreases with the increase of the gas flow rate. At the temperature of 313.2K, The removal efficiency of the absorbent MDEA-[N_{1111}][Gly] can reach 100%. The effect of the three new aqueous alkanolamine solutions on CO₂ absorption is: MDEA-[N_{1111}][Gly]>MDEA-[Bmim][Gly]>MDEA-[Bmim][Lys].

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References

[1] D’Alessandro D M, Smit B, Long J R, et al. Carbon dioxide capture: prospects for new materials[J]. Angewandte Chemie International Edition, 2010, 49: 6058-6082

[2] Zhang X P, Zhang X C, Dong H F, et al. Carbon capture with ionic liquids: overview and progress[J]. Energy & Environmental Science, 2012, 5: 6668-6681

[3] Nahicenovic N, John A. CO2 Reduction and Removal: Measures for the Next Century[J]. Energy, 1991, 16: 1347-1377

[4] Tan L. S., Shariff A. M., Lau K. K., et al. Impact of High Pressure on High Concentration CO2 Capture from Natural Gas by Monoethanolamine/ N-methyl-2-pyrrolidone Solvent in Absorption Packed Column[J]. International Journal of Greenhouse Gas Control, 2015, 34: 25-30

[5] Godini H. R., Mowla D. Selectivity Study of H2S and CO2 Absorption from Gaseous Mixtures by MEA in Packed Beds[J]. Chemical Engineering Research and Design, 2008, 86: 401-409

[6] Navaza J. M., Diaz D. G., Rubia M. D. L. Removal Process of CO2 Using MDEA Aqueous Solutions in a Bubble Column Reactor[J]. Chemical Engineering Journal, 2009, 146(2): 184-188

[7] Dubois L., Thomas D. Screening of Aqueous Amine-based Solvents for Postcombustion CO2 Capture by Chemical Absorption[J]. Chemical Engineering and Technology, 2012, 35(3): 513-524

[8] Chowdhury F. A., Yamada H., Higashi T., et al. CO2 Capture by Tertiary Amine Absorbents: a Performance Comparison Study[J]. Industrial and Engineering Chemistry Research, 2013, 52(24): 8323-8331

[9] Wappel D., Gronald G., Kalb R., et al. Ionic liquids for post-combustion CO2 absorption[J]. International Journal of Greenhouse Gas Control, 2010, 4(3): 486-494

[10] Zhao Y., Zhang X., Zhen Y., et al. Novel alcamines ionic liquids based solvents: Preparation, characterization and applications in CO2 capture[J]. International Journal of Greenhouse Gas Control, 2011, 5(2): 367-373

[11] Wang C., Luo X., Zhu X., et al. The strategies for improving CO2 chemisorption by functionalized ionic liquids[J]. RSC Advances, 2013, 3(36): 15518-15527

[12] Blanchard L. A., Hancu D., Beckman E. J., et al. Green processing using ionic liquids and CO2[J]. Nature, 1999, 399(6731): 28-29

[13] Tao G.-h., He L., Liu W.-s., et al. Preparation, characterization and application of amino acid-based green ionic liquids[J]. Green Chemistry, 2006, 8(7): 639-646

[14] Davis J. H. J. Task-specific ionic liquids[J]. Chemistry Letters, 2004, 33(9): 1072-1077.