Capturing CO_2 in a Bubble-Column Scrubber Using Blended Amine Solvent

Pao Chi Chen*, Huan Sheng Tseng, Zi Qi Lai and Zi Xiang Liao
Department of Chemical and Materials Engineering ;Lunghwa University of Science and Technology, Taiwan, China.
Email: chenpc@mail2000.com.tw

Keywords: Carbon dioxide, amine, bubble-column scrubber, Taguchi, mass transfer coefficient

Abstract: Carbon dioxide emissions are the major climatic change issue in the world. In order to reduce the CO_2 emissions, several technologies have been explored, in which absorption is found to be a powerful method. The key factors for an effective absorption are the structure of the scrubbers and solvents adopted. Therefore, this study aimed at absorbing carbon dioxide through a bubble column using AMP+MEA blended amine solution as an absorbent. The reason for using MEA+AMP as the absorbent was that its absorption capacity can be improved and the price was cheaper for both. In addition, the scrubbing factor for a bubble-column scrubber was higher. In the experiment, CO_2 absorption was performed by simulating the temperature of the flue gas and the concentration of CO_2 in the coal-fired power plant. The liquid flow rate (A), gas flow rate (B), temperature in the column (C) and solution concentration (D) were chosen as the operating factors. There were 4 factors in total, and 3 levels of each were taken for Taguchi design $L_9(3^4)$. There were a total of 9 groups of experiments. In order to explore the effect of operating variables on the absorption efficiency (E), absorption rate (R_A) and overall mass transfer coefficient (K_Ga), the parameter significance and target operating condition (E=90%; R_A=1x10^{-3} mol/s-L; K_Ga=0.4 s^{-1}) could be obtained by Taguchi design and Taguchi analysis, so as to serve as a reference for future scale-up design. The results showed that E was in 65.79-98.7%; R_A was in 3.54 x10^{-4}-13.9 x10^{-4}mol/s-L; and K_Ga was in 0.1743-0.3950 s^{-1}. After Taguchi analysis, A and B were found to be significant parameters, and the target condition was confirmed to be A1B1C2D3. In addition, the best condition was also discussed in here.

1 INTRODUCTION

Currently, the CO_2 emissions in industry are mainly from coal-fired power plants, steel-making plants, petrochemical industry and cement plants, etc., among which coal-fired power plants are the most concerned. In order to reduce CO_2 emissions, a lot of studies for the capture, storage and reuse of CO_2 were explored, mainly focusing on the post-combustion using absorption method (Yang et al., 2008; Yu et al., 2012), which is to use an alkaline solution to capture CO_2 (Chen et al., 2015). Therefore, the improvement of absorbent efficiency, absorbers and solvent regeneration efficiency has become important in research. How to effectively reduce the cost of electricity becomes the key to the success of CO_2 capture and storage (CCS). According to literature reported, the MEA is widely used in various amines (Chen et al., 2015; Versteeg et al., 1996). However, due to the high energy consumption required using single amine, the development of blended amine was valued (Vaidya and Kenig, 2007; Adeosun et al., 2013; Rinprasertmeechai et al., 2012), such as MEA+AMP, which has development space based on absorptive capacity and lower energy consumption (Choi et al., 2009; Khan et al., 2015; Aroonwilas and Veawab, 2009). A similar viewpoint for lower energy consumption was reported by Gomes et al. (Gomes et al., 2015), who found that diethyl amine was more competitive in price and loading capacity as compared with other amines. However, the reaction rate of blended amines with CO_2 can be expressed below (Versteeg et al., 1996; Xiao et al., 2000; Vaidya and Kenig, 2007):

Chen, P., Tseng, H., Lai, Z. and Liao, Z.
Capturing CO2 in a Bubble-Column Scrubber Using Blended Amine Solvent.
In *Proceedings of the International Workshop on Environment and Geoscience (IWEG 2018)*, pages 76-81
ISBN: 978-989-758-342-1

$$r_0 = [k_1(RR'NH) + K_{OH^-}(OH) + K'(R_2RN)][CO_2] \quad (1)$$

where (RR'NH) and (R_2RN) represent the concentration of MEA and AMP, respectively. This equation shows that the absorption rate is influenced by the concentration of MEA, AMP and OH^-, which also indicates that the CO_2 emitted by the plant can be treated with absorption mode in alkaline solution (Xiao et al., 2000; Chen et al., 2008). In addition, the efficiency of absorption process is usually set to 90%, it is also desirable to have a high mass transfer coefficient to make the scrubber smaller (Weiland and Hatcher, 2011; Idem et al., 2006). Due to the fact the mass transfer coefficient for the use of scrubber is approximately in the range of 0.1-1.0 1/s (Chen et al., 2015; Tontiwachwuthikul et al., 1992; Aroonwilas et al., 1999), it is difficult to achieve both higher efficiency (90%) and higher mass transfer coefficient (0.4 1/s), simultaneously.

In absorption experiment, the packed column, tray column and bubble column were often used. As the operation of the packed column is complicated, and its operating cost is high, while the bubble column is characterized by the merits such as controllable pH value, high mass transfer coefficient, high absorption factor and easy operation. Therefore in this study the absorption experiment is performed using the bubble column with a blended amine solution (MEA/AMP) as the absorbent. This study planned to conduct an experiment to absorb CO_2 by the bubble column with MEA+AMP as a solvent to search for and confirm the target operating condition and parameter significance, and predict the feasibility with absorption efficiency of 90%, absorption rate of 1.0×10^{-3} mol/L s, and mass transfer coefficient of 0.4 1/s. In order to compare, the best condition analysis was also explored.

2 EXPERIMENTAL DESIGN AND PROCEDURE

2.1 Absorption Experiment Design

The experiment aimed at absorbing carbon dioxide using MEA+AMP blended amine in a bubble column. The results were expected to be applied to the absorption of CO_2 emitted by coal-fired power plants. Therefore, the carbon dioxide of flue gas in the coal-fired power plant with the concentration of 15% and temperature of 55°C was simulated to enter the column. Taking liquid flow rate, gas flow rate, liquid temperature, and solution concentration as the condition factors, three levels of each condition factor were taken respectively, i.e. liquid flow rate (A:0.1-0.3L/min), gas flow rate (B:3-9L/min), liquid temperature (C:30-50°C), and solution concentration (D:4-6M) . Theoretically, a total of $81(=3^4)$ experiments needed to be done, but due to the high cost and time-saving in this way, this experiment used the Taguchi experimental design to reduce the groups of the experiment to $L_9\ (3^4)=9$ to save time and experimental costs. The value obtained in the steady state was adopted to obtain the absorption rate, absorption efficiency and overall mass transfer coefficient, and then the statistical software was used to find out the sequence of significance and target condition. Table 1 shows the condition factors and levels, Table 2 shows the orthogonal arrays, and there are 9 groups of experiments under different conditions needed to be conducted.

Table 1: Factors and levels in this work.

Factor	level		
	1	2	3
A(L/min)	0.1	0.2	0.3
B(L/min)	3	6	9
C(°C)	30	40	50
D(M)	4	5	6

Table 2: Orthogonal table, $L_9(3^4)$.

No.	A	B	C	D
No1	1	1	1	1
No2	1	2	2	2
No3	1	3	3	3
No4	2	1	2	3
No5	2	2	3	1
No6	2	3	1	2
No7	3	1	3	2
No8	3	2	1	3
No9	3	3	2	1

The Taguchi method uses the signal and noise ratio (S/N) as the process optimization objective function (Chen et al., 2015; Hvalec et al., 2004). The target value is:

$$\frac{S}{N} = -10 \times \log\left[\frac{1}{n} \times \sum_{i=1}^{n} (y_i - m)^2\right] \quad (2)$$

In addition, the best condition for larger-the-better is:

$$\frac{S}{N} = -10 \times \log\left[\frac{1}{n} \times \sum_{i=1}^{n} \frac{1}{y_i^2}\right] \quad (3)$$

where y_i is experimental data for ith level, n the number of level, and m the target value. Using the two equations and experimental data obtained, the objective condition, best condition and parameter importance could be determined.

2.2 Calculation of Experimental Data

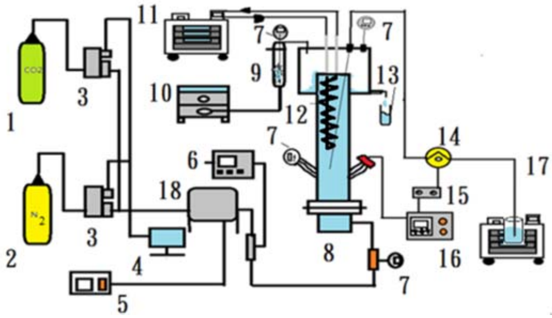
Experimental data including absorption efficiency, absorption rate, and overall mass transfer coefficient are listed in Table 3. In these equations, V_L, Q_y, P_A and T_1 represent the final solution volume, gas flow rate, inlet gas partial-pressure and inlet gas temperature, respectively. In order to determine overall mass transfer coefficient, a two-film model and mass balance are adopted (Chen et al., 2015; c). In addition, it is assumed that the concentration of CO_2 gas in the liquid phase is extremely small, which can be ignored ($C_A \gg HC_{AL}$). Therefore, the overall mass transfer coefficient can be calculated from the following Eq. (6), in which K_Ga can be calculated from known inlet and outlet conditions.

Table 3: Equations evaluated in this work (Chen et al., 2015).

$E = \left(\frac{y_1 - y_2}{y_1}\right) \times 100\%$	4)
$R_A = \left(\frac{1}{V_L}\right)\left(\frac{Q_g P_{A1}}{RT_1}\right)\left[1 - \left(\frac{1-y_1}{y_1}\right)\left(\frac{y_2}{1-y_2}\right)\right]$	5)
$K_G a = \frac{Q_g}{V_L} ln\left[\left(\frac{P_1}{P_2}\right)\left(\frac{T_2}{T_1}\right)\left(\frac{y_1}{y_2}\right)\right]$	6)

2.3 Experimental Devices and Procedures

The devices required for this experiment are shown in Figure 1, including the bubble column, gas-liquid feed system, pH detector, CO_2 detection system, gas heating system and liquid cooling system. In this experiment, the blended amine (AMP/MEA) was used as an absorbent, among which the AMP accounted for 30wt% of the total amine concentration, which was reported in the previous work. In order to achieve the desired concentration, the required blended amine concentration was prepared by using distilled water. Second, the flow rate of carbon dioxide and nitrogen was input based on the proportion of 15% of CO_2, and the gas inlet temperature was maintained at 55°C. The experiment was started after the blended amine was put into the column.



1. CO_2-tank	10. CO_2-meter
2. N_2-tank	11. Cooling system
3. Mass flow controller	12. Coil
4. PC	13. Sampling vessel
5. Heating adjuster	14. Tubing pump
6. Pressure gauge	15. Pump controller
7. Digital thermometer	16. pH controller
8. Bubble column	17. Thermal stat tank
9. Drying vessel	18. Heater

Figure 1: Experimental device.

3 RESULTS AND DISCUSSION

3.1 Operation and Data Calculation at the Steady State Condition

In order to understand the relationship of the outlet concentration of gas, liquid temperature, pH, and inlet pressure against time during operation, the measured value divided by the initial value was defined as X (-) value, which was taken as the Y-axis; the time (t) was taken as the X-axis, as shown in Figure 2; after 1 hour of experiment, it was found that the X values kept constant after 20 minutes, showing that the system reached a steady state. Therefore, all data can be evaluated at the steady state condition. The experimental results may be divided into two parts. Part 1 used the Taguchi method to design 9 groups of experiments, and calculated the data of absorption rate, absorption efficiency and overall mass transfer coefficient as shown in Table 4. Part 2 searched for and confirmed the target operating condition and the sequence of significance based on the results of the data calculated in Part 1 and Taguchi analysis. Table 4 shows the values calculated by Eqs. (4)-(6), where E is in 65.8% - 98.7%; R_A is in 3.54×10^{-4} - 13.9×10^{-4} (mol/s.L); K_Ga is in 0.1743- 0.3950(s^{-1}), and in addition, the pH is in 10.45 - 11.25 in the steady state. These data are comparable with literatures (Chen et al., 2015; Vaidya and Kenig, 2007).

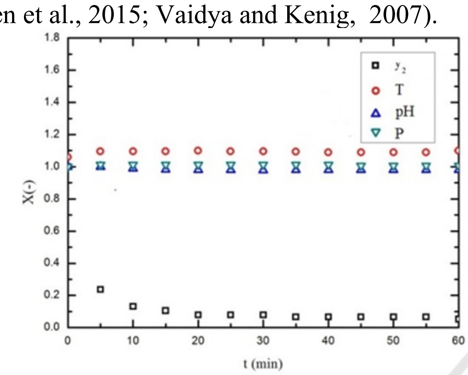


Figure 2: A plot of X vs. t, showing the steady state operation (No.1).

Table 4: Experimental data obtained in here.

NO	E(%)	$R_A(10^4)$ $(mol/s \cdot L)$	$K_G a$ (s^{-1})	pH
1	93.42	3.54	0.1743	10.64
2	82.89	7.3	0.2564	10.45
3	74.03	11.99	0.3525	10.16
4	96.05	4.23	0.2444	11.20
5	65.79	6.62	0.1769	10.43
6	70.51	11.6	0.3058	11.18
7	98.70	4.4	0.3153	10.82
8	78.67	7.35	0.2337	11.25
9	72.73	13.9	0.3950	10.73

3.2 Taguchi Analysis

Substituting the absorption efficiency and target value into Eq. (2) for Taguchi analysis, the results were shown in Table 5. It was found that the parameter significance affecting the absorption efficiency was B>A>C>D, and the combination of the target value was A1B1C2D3. The data showed that B was the main factor. After conducting the Taguchi target analysis for the absorption rate, it was found that the parameter significance affecting the absorption rate was B>D>C>A, and the combination of the target value was A2B3C3D2. It was found that B was the main influence factor. In addition, Taguchi target analysis for the overall mass transfer coefficient showed that the parameter significance affecting the overall mass transfer coefficient was B>D>A>C, and that the best combination was A3B3C2D2. It was also found that factor B was the main influencing factor. Table 6 showed the parameter significance analysis and target conditions obtained through S/N analysis for the three types of calculation data. Regarding the overall influence effect, B was the most significant, followed by D, and it was also found that the impact of C was the least significant.

Alternatively, the best condition and parameter significance through the Taguchi analysis were shown in Table 7. It was found that the parameter significances affecting E (No. 13), R_A(No. 14), and K_Ga(No.15) were B>D>A>C, B>A>C>D and B>A>D>C, respectively. It was also found that B was the main influence factor, while C was the minor factor. The result was similar to that target analysis. Except B, D is the second significant in Target condition, while A is the second significant in the best condition. In addition, the conditions for Nos. 12 and 15 are the same, but the parameter importance is slightly different.

Table 5: S/N ratio analysis for E.

Level	A	B	C	D
1	(-20.24)	(-16.16)	-22.39	-24.75
2	-25.24	-24.06	(-21.06)	-22.27
3	-22.23	-24.93	-24.85	(-21.46)
Delta	5.00	8.77	3.76	3.29
Rank	2	1	3	4
Target	1	1	2	3

Table 6: Target condition and significance.

Target	Significance	Condition
NO.10(E)	B>A>C>D	A1B1C2D3
NO.11(R_A)	B>D>C>A	A2B3C3D2
NO.12(K_Ga)	B>D>A>C	A3B3C2D2

Table 7: The best condition and significance.

The best	Significance	Condition
NO.13(E)	B>D>A>C	A1B1C2D2
NO.14(R_A)	B>A>C>D	A3B3C2D3
NO.15(K_Ga)	B>A>D>C	A3B3C2D2

3.3 Confirmations of the Target Conditions and the Best Conditions

Table 8: Confirmation for target condition and the best condition.

NO	E (%)	$R_A(10^4)$ $(mol/s \cdot L)$	$K_G a$ $(1/s)$	pH
10	(92.11)	12.25	0.5723	10.82
11	97.33	(3.34)	0.2160	10.93
12	82.67	11.86	(0.403)	11.00
13	(94.74)	3.74	0.1924	10.62
14	80.52	(13.48)	0.4415	10.72
15	82.67	11.86	(0.403)	11.00

Table 6 showed the analysis results of the target condition and the best condition obtained by Taguchi analysis. According to the target conditions, three experiments were carried out and the results were shown in Table 8. It was found that No. 10, its E, R_A and $K_G a$, were all up to standard, indicating that the target conditions is obtained in here. In addition, the best condition confirmation showed that No. 13 was not reached the best, while the Nos. 14 and 15 all reached the best as compared with Taghchi experiments listed in Table 4. From both analyses, it was hard to obtain higher E and $K_G a$ simultaneously. Therefore, how to use effective experimental design to obtain the desired conditions became significant. In this study, the condition of A1B1C2D3(No. 10) can satisfy not only a higher E, but also a higher $K_G a$.

4 CONCLUSIONS

A continuous bubble-column scrubber was successfully used to investigate the process variables on the absorption efficiency, absorption rate, and overall mass transfer coefficient. Under Taguchi experimental design, a total of 9 runs were carried out in here. It was found that the system could reach a steady state after 20 minutes, while the final pH was in the range of 10.16-11.25, depending on the operating condition. In addition, the absorption efficiency was found to be in the range of 65.79-98.7%; the absorption rate was in 3.54×10^{-4} - $13.9 \times 10^{-4} mol / s \cdot L$; and the overall mass transfer coefficient was in 0.1743-0.3950 1/s. From Taguchi analysis for target and the best modes, it was found that the gas flow rate (B) was major factors influencing the outcome data, while the influence of the temperature in the column (C) was the minor. In addition, the target condition was found to be A1B1C2D3. Through the confirmation of target conditions, it was known that the E (92.11%), R_A (1.225×10^{-3} mol/s L) and $K_G a$ (0.5723 s^{-1}) all reached the set target value.

REFERENCES

Adeosun A, Goetheer N E H, Abu-Zahra M R M 2013 Absorption of CO2 by Amine Blends Solution: An Experimental Evaluation *International Journal of Engineering And Science* **3** 12

Aroonwilas A, Veawab A 2009 Integration of CO2 capture unit using blended MEA-AMP solution into coal-fired power plants *Energy Procedia* **1** 4315

Aroonwilas A, Veawab A, Tontiwachwuthikul P 1999 Behavior of mass-transfer coefficient of structured packings in CO2 absorbers with chemical reactions 1999 *Ind Eng. Chem. Res.* **38** 2044

Chen P C, Luo Y X, Cai P W 2015 Capture of Carbon Dioxide Using Monoethanolamine in a Bubble-Column Scrubber *Chemical Engineering & Technology* **38** 274

Chen P C, Shi W, Du R, Chen V 2008 Scrubbing of CO2 green-house gases, accompanied by precipitation in a continuous bubble-column scrubber *Ind. Eng. Chem. Res.* **47** 6336

Choi W J, Seo J B, Jang S Y, Jung J H, Oh K J 2009 Removal characteristics of CO2 using aqueous MEA/AMP solutions in the absorption and regeneration process *Journal of Environmcntal Scicnccs* **21** 907

Fan L S 1989 Gas-liquid-solid fluidization engineering *Butterworths USA* Ch. 9 600

Gomes J, Santos S, Bordado J 2015 Choosing amine based absorbents for CO2 capture *Environmental Technology* **36** 19

Hvalec M, Gorsek A, Glavic P 2004 Experimental design of crystallization processes using Taguchi method *Acta Chim. Slov.* **51** 245

Idem R, Wilson M, Tontiwachwuthikul P, Chakma A, Veawab A, Aroonwilas A, Gelowitz D 2006 Pilot plant studies of the CO2 capture perforance of aqueous MEA and Mixed MEA/MDEA solvents at the University of Regina CO2 capture technology development plant and boundary dam CO capture demonstration plant *Ind. Eng. Chem. Res.* **45** 2414

Khan A A, Halder G N, Saha A K 2015 Carbon dioxide capture characteristics from flue gas using aqueous 2-amino-2-methyl-1-propanol (AMP) and monoethanolamine (MEA) solutions in packed bed absorption and regeneration columns *Int. J. Greenh. Gas Control* **32** 15

Rinprasertmeechai S, Chavadej S, Rangsunvigit P, Kulprathipanja S 2012 Carbon dioxide removal from flue gas using amine-based hybrid solvent absorption *World Academy of Science, Engineering and Technology* **64** 410

Tontiwachwuthikul P, Meisen A, Lim J 1992 CO_2 Absorption by NaOH, Monoethanolamine and 2-amino-2-methyl-1-propanol Solutions in a Packed bed *Chem. Eng. Sci*. **47** 381

Vaidya P D, Kenig E Y 2007 Absorption of CO_2 into aqueous blends of alkanolamines prepared from renewable resources *Chem. Eng. Sci.* **62** 7344.

Vaidya P D, Kenig E Y 2007 CO_2-alkanomine reaction kinetics: A review of recent work *Chem. Eng. Technol*. **30** 1467

Versteeg G F, Van Dijck L A J, van Swaaij W P J 1996 On the kinetics between CO_2 and Alkaloamines both in aqueous and non-aqueous solutions. An overview *Chem. Eng. Comm.* **144** 113