ARTICLE





Response surface modelling of CO₂ capture by ammonia aqueous solution in a microchannel

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Abstract

Post-combustion CO₂ capture is one strategy of greenhouse gases mitigation. Ammonia is a useful option as CO2 absorbent and an alternative to conventional amine-based solutions. This study deals with CO₂ capture by ammonia aqueous solution in a co-current two-phase flow by utilizing a T-shaped microchannel. Three parameters of temperature, gas flow rate, and ammonia concentration were considered as the main parameters affecting the CO₂ capture efficiency. A response surface methodology based on central composite design (CCD) was used to model the CO₂ capture efficiency as output in terms of the aforementioned input variables. CCD suggested a quadratic model to fit the experimental data. The model validation was implemented by ANOVA. All statistic tools including correlation coefficient, P-value, and F-value of the model, and P-value of lack-of-fit confirmed that the prediction model was significant. It was deduced from F-values that the importance of the input variables followed the sequence of ammonia concentration > gas flow rate > temperature. Ammonia concentration was the most effective input variable because there was a direct correlation between ammonia concentration and the number of absorption sites in the liquid phase. Numerical optimization predicted the best output of 96.48% CO₂ capture under the following optimum conditions: temperature of 20.00°C, gas flow rate of 110.59 mL/min, and ammonia concentration of 0.1382 mL/mL (13.82 vol%). The average CO₂ capture of 95.42% obtained at the input conditions indicates the accuracy of the prediction model.

KEYWORDS

ammonia absorbent, central composite design, ${\rm CO_2}$ capture, microchannel

1 | INTRODUCTION

The issue of greenhouse gas emissions and greenhouse effects has become an important subject of increasing concern. CO_2 is the most known greenhouse gas and has been estimated to be responsible for 64% of enhanced global warming.^[1] One beneficial strategy of CO_2 mitigation is post-combustion carbon capture, which has been

widely considered over recent decades.^[2] Of the various alternatives of carbon capture from gas streams including solvent absorption methods, solid adsorption methods, cryogenic methods, membrane-based methods, and biological fixation, solvent-based absorption is the most mature technology.^[3–5] Two mechanisms of chemical and physical absorption may take part in the CO₂ absorption alone or simultaneously to reduce the amount of

 ${\rm CO_2}$. ^[6] Chemical absorption often possesses advantages over physical absorption mainly because of its higher absorption capacity and applicability even in low ${\rm CO_2}$ partial pressures. Chemisorption may also enhance the mass transfer from gas to liquid due to the depletion of ${\rm CO_2}$ in the liquid phase through chemical reaction. ^[7,8] However, the simplicity of solvent regeneration with a less costly pressure-swing process is a notable superiority of physical absorption. ^[9]

The potential for CO₂ capture of many aqueous alkanolamine solutions has been widely studied, [10-15] yet monoethanolamine (MEA) solution remains the most efficient chemisorption solvent due to its high reactivity, thermal stability, and low cost. [16,17] Although MEA solution is frequently being used for the recovery of CO₂ from flue gas, it suffers from some faults with respect to kinetics. $^{[16]}$ Besides, SO_2 , NO_x , and O_2 must be removed from the gas stream prior to coming into contact with the MEA solution due to the degradable feature of MEA through irreversible reactions.^[18] The high energy requirement for MEA regeneration is another shortcoming relating to the real application of this absorbent. [19] Since SO₂, NO_x, and O₂ from stack gas may cause degrading in amine solutions, they should be removed from inlet gas feed to amine-absorption units. Therefore, equipment for removing SO₂, NO_x, and O₂ from stack gas before entering the absorption tower should be made of corrosion-proof materials to prevent corrosion as well as high energy consumption for amine recovery, have made amine-based processes costly. [10,18,19,20] As for the need to develop highly efficient and sustainable CO2 absorption processes, new absorption solvents (and blends) are being investigated. Ammonia-based solvents have attracted an increased amount of attention as a CO2 capture solvent.[19-24] The main advantages of ammonia-based absorption are that there is no need for SO₂, NO_x, and O₂ removal as they do not degrade ammonia, larger CO2 absorption capacity, and lower regeneration energy. [20,21] However, ammonia solution also possesses disadvantages. The main disadvantage is the volatility of ammonia, which requires more ammonia solution make-up during absorption and leads to a high energy cost for process chiller to keep its temperature low. This is the main ammonia is not widely reason why industries.[10,20,22]

By using ammonia solution, both chemical and physical CO_2 absorptions may take place simultaneously. However, physical absorption seems to be dominant. Physical absorption refers to the diffusion of CO_2 molecules from the gaseous film to the gas-liquid interface before dissolving in the liquid film, which can be affected by the gas-liquid contact area. The dissolved CO_2 (CO_2 (CO_2) then reacts with the ammonia molecules to form

 CO_2 -containing ammonia salts, ammonium bicarbonate (NH₄HCO₃), ammonium carbonate ((NH₄)₂CO₃. H₂O), and ammonium carbamate (NH₂COONH₄), according to Equations (1) to (7)^[25,26]:

$$CO_2(aq) + H_2O \rightleftharpoons HCO_3^- + H^+$$
 (1)

$$HCO_3$$
⁻ $\rightleftarrows CO_3$ ²⁻ + H⁺ (2)

$$NH_3(aq) + H_2O \rightleftharpoons NH_4^+ + OH^-$$
 (3)

$$NH_3(aq) + HCO_3^- \rightleftharpoons NH_2COO^- + H_2O$$
 (4)

$$NH_4^+ + HCO_3^- \rightleftarrows NH_4HCO_3(s)$$
 (5)

$$NH_4^+ + NH_2COO^- \rightleftharpoons NH_2COONH_4(s)$$
 (6)

$$2 NH_4^+ + CO_3^{2-} + H_2 \rightleftarrows (NH_4)_2 CO_3.H_2O(s)$$
 (7)

In recent years, there have been a significant number of studies devoted to CO₂ capture by ammonia solutions the laboratory scale by various gas-liquid contactors. [3,27-30] Currently, CO2 chemisorption by traditional gas-liquid contacting devices, such as the spray column,[27] packed bed column,[28] wetted wall column^[29] and bubble column.^[30] faces two major limitations: gas-liquid contact area and mass transfer rate. These limitations can be overcome by high throughput reactors and adding rate promoters to the ammonia solution. [22,31] However, the economic constraints have led to a search for practical solutions that are highly efficient. Today, with the development of microdevices, microreactors have remarkably improved the CO2 absorption efficiency since they provide a high surface-to-volume ratio, short transport paths, and high mass transfer driving forces. [31,32] thereby increasing the rate of reactions, heat, and mass transfer. Up to this point, there have been a limited number of studies on CO₂ absorption by ammonia solution in microchannels. Kittiampon et al^[31] used a T-type microchannel $(0.5 \times 0.5 \times 60 \text{ mm}^3)$ for CO_2 absorption (feed composition: 10 vol% CO₂:90 vol% N₂) by ammonia solution. They studied the effect of temperature, pressure, ammonia concentration, and liquid flow rate on the efficiency of CO₂ capture and reported the best ammonia removal of 96.45%.

Based upon the discussion above, this paper aims to assess the CO_2 capture efficiency of ammonia aqueous solution in a micro co-current flow inside a T-shaped microchannel with an inner diameter of 0.8 mm. Response surface methodology (RSM) was also conducted to seek the best operating conditions maximizing the CO_2 capture efficiency. RSM is a reliable

chemometric method for the behaviour prediction of multivariable systems. There is a growing trend towards the optimization of various controllable systems by experimental design approaches. However, it seems that there have been no reports on the modelling and optimization of CO_2 absorption by aqueous ammonia in a microchannel by RSM. Herein, three-factor central composite design (CCD) was used in terms of temperature (14°C-26°C), gas flow rate (80-140 mL/min), and ammonia concentration 0.08 to 0.16 mL/mL (8-16 vol%) as input variables. The concentration of CO_2 in the gas phase was measured using an online gas chromatograph equipped with a thermal conductivity detector (GC-TCD).

2 | EXPERIMENTAL APPARATUS AND METHODS

2.1 | Materials

Commercial grades CO₂ and N₂ (99.99%) gases were used in this work. Ammonia solution (32%) was purchased from Merck. Distilled water obtained from our lab was used to dilute ammonia solution.

2.2 \mid CO₂ capture process

All of the experiments of CO₂ capture were carried out in a T-shaped quartz microchannel with the inner diameter of 0.8 mm, inlet channel length of 4.5 cm, and mixing channel length of 45 cm and absorption channel length of 45 cm, which is shown in Figure 1.

A schematic diagram of experimental setup used in this work is shown in Figure 2. A gaseous feed composed of 13.8%CO₂: 86.2%N₂ and a liquid feed containing ammonia diluted with deionized water with desired concentration were simultaneously injected into the microchannel via the inlet channels. Prior to injection, the temperature of liquid feed was adjusted by a water bath and a temperature controller. In order to ensure that the inlet temperature is constant, a pump was located near the channel and the tubes were insulated carefully. The

inlet gas flow rate was controlled by a volume flow meter. To measure the CO_2 concentration, the outlet gas was first separated from the outlet liquid by a flash drum and then was passed through a gas washing bottle containing 10% sulphuric acid to prevent ammonia gas from entering the GC. The gas stream was then injected into an online gas chromatograph equipped with a thermal conductivity detector (GC-TCD) and the concentration of CO_2 was calculated in 20-second intervals.

The efficiency of CO₂ absorption was calculated from Equation (8) in which CO_{2in} and CO_{2out} are the concentration of CO₂ in the inlet and outlet of the microchannel, respectively:

$$CO_2 \operatorname{capture}(\%) = \frac{CO_{2in} - CO_{2out}}{CO_{2in}} \times 100$$
 (8)

2.3 | Experimental design and statistical analysis

As the efficiency of CO₂ capture in the continuous systems depends on the temperature, the amount of absorbent, and flow rate of gas stream, an experimental design based on the central composite approach was used to obtain the best plausible answer under controllable experimental conditions. Due to type of gas flow and temperature controllers used for experiments, and by studying previous works, [20-22,26-31] three levels with appropriate ranges were considered. CCD is a response surface methodology and suggests a set of 17 runs (with three replicates of the central point) when investigating three factors with three levels. We used Design Expert Software (version 7.0.0). Table 1 lists the factors and levels of modelling CO₂ capture in this work. CCD offered a second-order polynomial model predicting the CO₂ capture efficiency (dependent output) as a function of independent inputs, as given in Equation (9):

$$Y = b_0 + \sum_{i=1}^{k} b_i x_i + \sum_{i=1}^{k} b_{ii} x_i^2 + \sum_{i=1}^{k} \sum_{i>1}^{k} b_{ij} x_i x_j + \varepsilon$$
 (9)



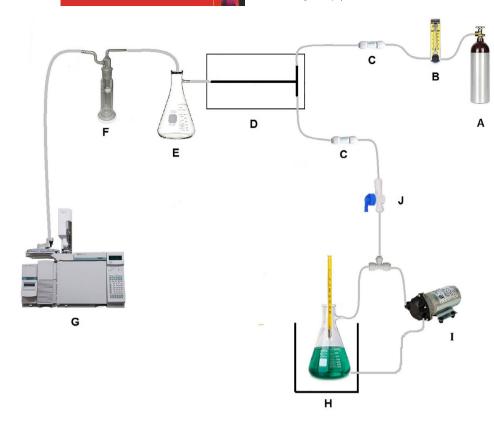


FIGURE 2 The experimental set-up used for the CO₂ capture process. The main components were: (A) gas bottle storage, (B) gas flow meter, (C) non-return valve, (D) microchannel, (E) flash drum, (F) gas washing bottle, (G) gas chromatograph equipped with a thermal conductivity detector, (H) liquid storage surrounded by a water bath, (I) liquid pump, and (J) liquid control valve

TABLE 1 Independent input variables and their coded levels

		Level factor		
Input variable	Symbol	Low (-1)	Medium (0)	High (+1)
Temperature (°C)	A	14	20	26
Gas flow rate (mL/min)	В	80	110	140
Ammonia concentration (mL per mL of solution)	С	8	12	16

where Y is the modelled output, x_i is the dimensionless codded input variable of X_i , and i and j are the index number. b_0 is the offset term and b_i , b_{ii} , and b_{ij} are the coefficients of linear, squared, and mutual interaction effects, respectively. ε is the random error.

The next step of the experimental design is the validation of the results obtained from mathematical modelling. This is often accomplished by ANOVA, which is a powerful statistical package. The regression coefficient of the adjusted and predicted, lack-of-fit, *P*-value, and F-value were used to determine the creditability of the model used and the significance of each input variable.

3 | RESULTS AND DISCUSSION

3.1 | Experiments

First, to determine the best domain of each input variable, a single-level study was performed. Figure 3A shows the effect of the gas flow rate on the CO2 capture efficiency. In this set of experiments, the gas flow rate was changed from 50 to 150 mL/min at the constant liquid flow rate. A temperature of 20°C and ammonia concentration of 15% were set in the tests. It can clearly be seen that the CO₂ capture efficiency was approximately stable by increasing gas flow rate from 50 to 110 mL/min, and then it was decreased from 110 to 150 mL/min. This could be due to the drop in the absorption capacity of liquid phase, the higher gas-to-liquid ratio, and the short contact time between gas and liquid phases.[32] Ammonia loss, which is a result of ammonia stripping, is another reason for the decrease in CO2 absorption at a high gas flow rate. [9,33] Besides, at high gas flow rates, the possibility of effective collisions between CO2 and ammonia molecules may be reduced, thus diminishing the efficiency of the CO₂ capture. According to these results, the levels of gas flow rate as one input variable were determined to be 80 (-1), 110 (0), and 140 (+1).

The effect of temperature on the CO_2 capture efficiency was investigated at various temperatures between $10^{\circ}C$ and $30^{\circ}C$, a gas flow rate of 110 mL/min, and an



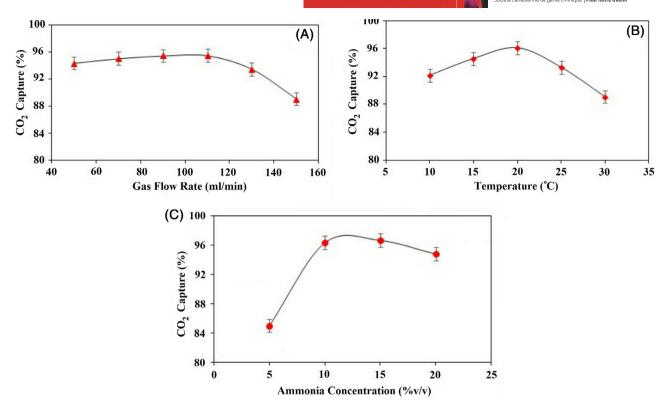


FIGURE 3 Results of one-factor experiments: A, effect of gas flow rate B, effect of temperature; and C, effect of ammonia concentration on the CO₂ capture efficiency

ammonia concentration of 15%. The obtained result is shown in Figure 3B. CO₂ absorption was slightly increased by increasing the temperature from 10°C to 20°C, and then it decreased with more intensity. Generally, absorption is a temperature dependent process. Herein, temperature can affect the physical solubility of CO₂ in aqueous ammonia, the rate constants of reactions between the dissolved CO₂ and ammonia to form CO₂containing ammonia salts, and the diffusion coefficient in the liquid phase. [32] Increasing the temperature decreases the physical solubility of CO2; however, the reaction rates increase.^[26] Increasing the temperature of the liquid phase also facilitates the molecular diffusion in the liquid bulk; therefore, the absorption capacity can be improved.[31] Thus, at temperatures below 20°C, the higher reaction rates and molecular diffusion in the liquid phase were the processes that determined the CO₂ absorption, while the physical solubility of the CO₂ in the aqueous ammonia was the most important process at temperatures higher than 20°C, which had an adverse effect on the CO₂ absorption. In addition, the volatility of ammonia increases with a rising temperature, which is directly related to the absorption capacity of the liquid phase. [25] Therefore, the levels of temperature as the other input variables were chosen as $14(-1)^{\circ}C$, $20(0)^{\circ}C$, and 26 (+1)°C.

The effect of the last input variable, ammonia concentration, on the CO₂ capture efficiency was studied at various values between 0.05 and 0.2 mL/mL (5-20 vol%), a gas flow rate of 110 mL/min, and a temperature of 20°C, and the results are given in Figure 3C. There was a positive correlation between the CO₂ absorption and ammonia concentration when it was between 5% and 12%, and then the CO₂ absorption plunged by further increasing the ammonia concentration from 12% to 20%. This positive correlation can be ascribed to the absorption capacity of the liquid phase, which was increased by increasing the ammonia content. However, ammonia stripping at higher ammonia concentrations can hinder the physical solubility of CO2 in the aqueous phase. Further, some difficulties associated with overusing ammonia can arise, such as microchannel blockage caused by the accumulation of solid ammonium salts. In accordance with the obtained results, the levels of the last input variable of ammonia concentration were determined to be 8 (-1), 12 (0), and 16 (+1) mL per 100 mL of solution.

3.2 | Non-linear modelling and validation by ANOVA

The matrix of the experimental design alongside the observed outputs is presented in Table 2. The CCD fitted



a non-linear model to the observed outputs. All the experiments were repeated five times and the average was considered as the result in order to achieve high accuracy.

Equation (10) is the model suggested for the prediction of CO₂ absorption by aqueous ammonia in terms of coded factors of temperature, gas flow rate, and ammonia concentration:

$$CO_{2} capture (\%) = +95.51 - 1.24A - 1.99B + 5.49C + 0.87AB + 0.59AC + 0.49BC - 3.19A^{2} - 1.61B^{2} - 5.09C^{2}$$

$$(10)$$

where A, B, and C correspond to the coded values of temperature, gas flow rate, and ammonia concentration, respectively. To determine whether the model suggested by CCD is reliable, the data predicted must be analyzed in a statistical manner. ANOVA was used for this purpose and the results are given in Table 3. For a mathematical model to be successful in the prediction of system behaviour, the lack-of-fit should be insignificant, which means that its *P*-value should be greater than .05. Results indicated the *P*-value of lack-of-fit to be .9725; therefore, the model was sufficiently significant and reliable in predicting the output of CO₂ capture efficiency. Also, the model *P*-value of <.0001 confirmed the significance of the model. It should be mentioned that R² is a goodness-

TABLE 2 The matrix of three-factor central composite design in terms of coded inputs showing the observed output

Run no.	A	В	C	Observed output (%)
1	0	0	0	96.20
2	-1	+1	-1	78.84
3	+1	+1	-1	76.57
4	0	-1	0	96.09
5	-1	-1	+1	94.30
6	0	0	-1	85.00
7	0	+1	0	91.55
8	+1	-1	-1	79.87
9	0	0	0	96.12
10	+1	0	0	91.27
11	-1	0	0	93.23
12	+1	-1	+1	90.91
13	-1	-1	-1	85.18
14	-1	+1	+1	89.50
15	0	0	0	94.51
16	+1	+1	+1	89.99
17	0	0	+1	95.70

of-fit measure for linear regression models and adjusted R² is used to compare the goodness-of-fit for regression models that contain different numbers of independent variables. Moreover, predicted R² is used to determine how well a regression model makes predictions. The predicted R² value (0.9871) is high, close to 1, and is in reasonable agreement with the adjusted R² (0.9916). The P-values of all input factors of A (temperature), B (gas flow rate), and C (ammonia concentration) revealed their significance and impression on the output, and all of them are much lower than 0.05. The F-values of A, B, and C factors showed the order of importance as follows: ammonia concentration > gas flow rate > temperature. In the same manner, all the squared and mutual interaction terms are significant model terms. Adequate precision is a parameter comparing the range of predicted outputs at the design points to the average prediction error. Values greater than four represent adequate model discrimination. Therein, the adequate precision of 42.815 indicates an adequate signal. The model exhibited a coefficient of variation (C.V.%) of 0.65%, meaning a small SD relative to the mean.

The accuracy of the model can be found out from the plot of predicted vs observed values, as depicted in Figure 4A. This plot shows that the predicted outputs were in high correlation with the observed outputs, demonstrating an adequate signal for regression model. The normal probability plot of the residuals is shown in Figure 4B. The residuals generally dispersed on a straight line, which indicates the normal distribution of errors. A well-behaved plot of residuals vs predicted outputs is shown in Figure 4C. Obviously, no regular pattern can be found. This reflects that the prediction model is adequate and verifies the assumption that the residuals had a constant variance. Figure 4D illustrates the external residuals against the experimental run order for the CO2 capture efficiency. This plot shows that there was no systematic relationship between the observed outputs and order in which the experimental data were collected. It can be said that the model suggested by CCD successfully passed all the statistical tests.

3.3 | Response surface analysis

3D surface and contour graphs are the graphical representation of the suggested model, as shown in Figure 5, and are used to determine the effectiveness of interactions between independent inputs on the output within the domain considered. Figure 5A,B shows the effects of the mutual interaction of temperature and gas flow rate on the $\rm CO_2$ capture efficiency. The third parameter was set to 0.12 mL/mL (12.00 vol%). The $\rm CO_2$ capture

TABLE 3 ANOVA table for the quadratic model suggested by central composite design

Source	Sum of squares	Degree of freedom	Mean square	F-value	<i>P</i> -value Prob > F
Model	651.68	9	72.41	211.23	<.0001
A-Temperature	15.46	1	15.46	45.11	.0003
B-Gas flow	39.61	1	39.61	115.55	<.0001
C-Ammonia concentration	301.71	1	301.71	880.15	<.0001
AB	6.00	1	6.00	17.50	.0041
AC	2.74	1	2.74	7.99	.0255
ВС	1.93	1	1.93	5.62	.0496
A^2	27.18	1	27.18	79.30	<.0001
B^2	6.96	1	6.96	20.30	.0028
C^2	69.39	1	69.39	202.41	<.0001
Residual	2.40	7	0.34		
Lack of fit	0.57	5	0.11	0.12	.9725
Pure error	1.83	2	0.91		
Cor total	654.08	16			
SD	0.59		\mathbb{R}^2	0.9963	
Mean	89.70		Adj-R ²	0.9916	
C.V.%	0.65%		Pred-R ²	0.9871	
PRESS	8.47		Adeq Precision	42.815	

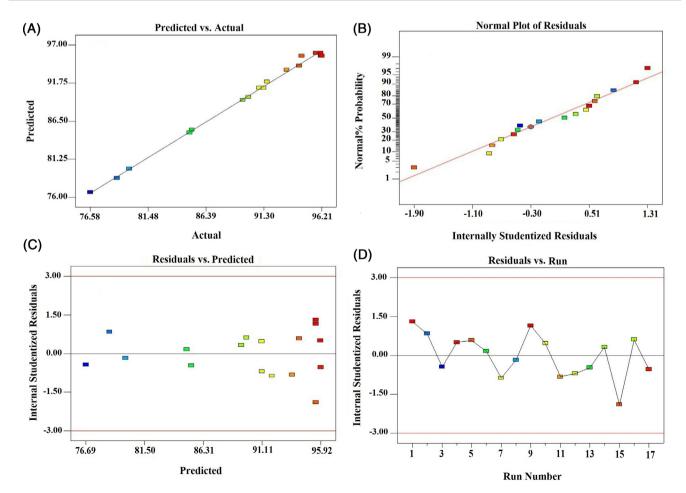


FIGURE 4 Plots of: A, predicted vs actual; B, normal probability; C, residuals vs predicted outputs; and D, residual vs run number

efficiency increased with increasing temperature and gas flow rate, and then it decreased with a further increase in the input variables. This effect is more obvious with a varying gas flow rate, as is clear in the contour plot. At higher gas flow rates, the CO₂ capture efficiency

decreased possibly because of the reduction in the liquid flow rate, which decreased the overall mass transfer coefficient. [32] In addition, at higher gas flow rates, there was not enough contact time between the CO₂ and ammonia molecules, thereby diminishing the opportunity to

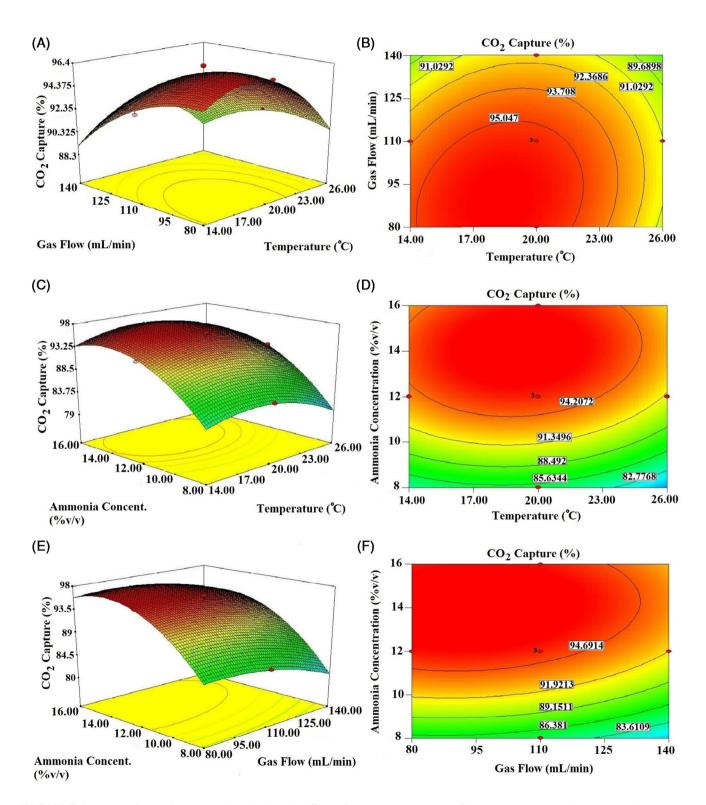


FIGURE 5 3D surface and contour plots showing the effects of: A, B, temperature-gas flow rate; C, D, ammonia concentration-temperature; and E, F, ammonia concentration-gas flow rate



interact with each other. Ammonia stripping also affected the CO_2 capture efficiency for two reasons: reducing the liquid absorption sites and slowing down the mass transfer rate in the gas phase. It should be noted that the CO_2 - NH_3 - H_2O system is a volatile system and there is a gasliquid equilibrium according to Equations (11)-(13)^[25]:

$$CO_2(g) \rightleftarrows CO_2(aq)$$
 (11)

$$NH_3(g) \rightleftarrows NH_3(aq)$$
 (12)

$$H_2O(g) \rightleftarrows H_2O(1)$$
 (13)

Figure 5C,D shows the effect of ammonia concentration and temperature on the $\rm CO_2$ capture efficiency; the third parameter was kept constant in 110 mL/min. The $\rm CO_2$ capture increased and then decreased smoothly when the

temperature was increased from 14°C to 26°C. This result is similar to that observed in Figure 5A,B. The ammonia concentration had a more severe effect on the CO₂ capture efficiency because it is directly related to the absorption capacity. At any temperature, increasing the ammonia concentration from 0.08 mL/mL (8 vol%) to near 0.14 mL/mL (14 vol%) led to an increase in the CO₂ capture efficiency. The higher the ammonia concentration, the more absorbent molecules interacting with the dissolved CO2. However, the CO₂ capture efficiency remained constant by further increasing the ammonia concentration. At higher ammonia concentrations, the CO2 loading capacity will be reduced and this will affect the CO₂ capture efficiency.^[20] The effects of ammonia concentration and gas flow rate on the CO₂ capture efficiency are shown in Figure 5E,F. The third parameter was set to 20°C. It is clear that the maximum value of CO₂ capture efficiency will occur with high ammonia concentration and low gas flow rate. The effects

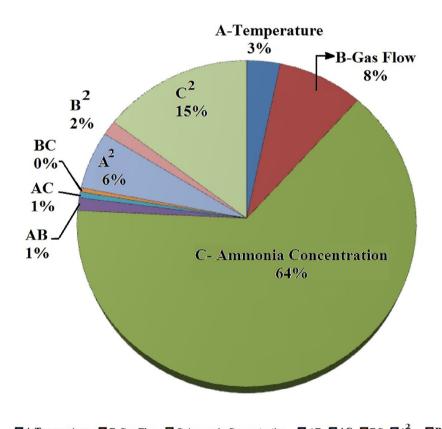


FIGURE 6 The share of the effect of each parameter

effect of each parameter	A-Temperature	■ B-Gas Flow	C-Ammonia Concentration	AB A	AC BC A	■B-	$\Box C^-$

ΓABLE 4 Optimization of individual desirability responses (d_i) to calculate the overall desirability response (D)

Variable	Goal	Lower limit	Lower weight	Upper limit	Upper weight	Importance
Temperature	In the range	14	1	26	1	3
Gas flow	In the range	80	1	140	1	3
Ammonia concentration	In the range	8	1	16	1	3
CO ₂ capture	Maximize	76.57	1	96.20	1	5

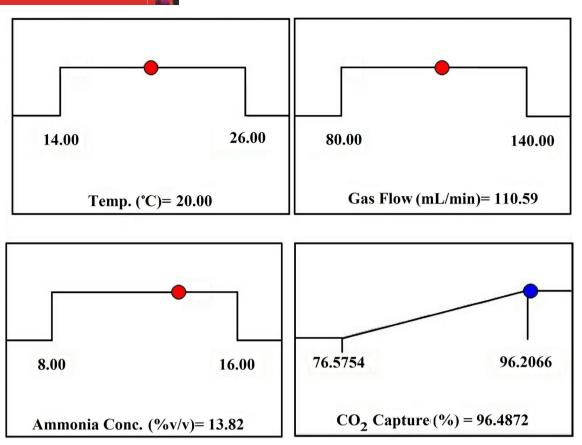


FIGURE 7 Desirability ramp for numerical optimization

TABLE 5 Results of optimization based on predicted and actual values

	Temperature (°C)	Gas flow rate (mL · min ⁻¹)	Ammonia concentration (mL per mL of solution)	CO ₂ capture efficiency (%)
Predicted values	20	110.59	13.82	96.48
Actual values	20	110	14	95.04
Actual values	20	110	13.5	95.81

of these variables on the output were discussed earlier. These results are in accordance with the results obtained from the F-value that demonstrated that the ammonia concentration was the factor that had the most significant effect on the CO_2 capture efficiency, followed by gas flow rate and temperature.

In ANOVA, the sum of the error values (SS_i) for each variable is shown. Based on the sum of the squares of the error, we can obtain the contribution of each parameter to Equation (10) given by the design of the experiments by Equation (14) as follows:

$$PC_i = SS_i / \left(\sum_{i=1}^n SS_i\right) \tag{14}$$

Using Equation (14), we can determine the contribution of each parameter, as shown in Figure 6. As can be seen, the ammonia concentration with 64% has the most effect, followed by the same squared concentration parameter and the gas discharge parameter, respectively.

3.4 | Prediction and verification of the best output

The optimization method developed by Derringer and Suich, [34] based on the prediction model, was used to determine the optimum values of input variables for maximizing the CO₂ capture efficiency. Table 4 shows the desired goals for each input and output variables to be combined into an overall desirability function, and

this function must be maximized. Since the CO₂ capture efficiency was the only output in this study, an importance value of five was set to emphasize the importance of the output and a medium setting of importance field at three left by default for the independent variables.

As shown in Figure 7, the optimum conditions under which the best $\rm CO_2$ capture efficiency can be obtained were found to be a gas flow rate of 110.59 mL/min, a temperature of 20°C, and an ammonia concentration of 0.1382 mL/mL (13.82 vol%). Under these optimum conditions, a $\rm CO_2$ capture efficiency of 96.48% was predicted as the best output.

To verify the adequacy of the model for the prediction of the best CO_2 capture efficiency, two ammonia concentrations close to optimum concentration were examined. An average value of 95.42% was obtained for the CO_2 capture efficiency, which was 1% lower than that predicted. The observed difference between the predicted and observed values of the best output is attributed to the problems with adjusting the input variables at the exact values predicted. Table 5 compares the results of the verification tests with those predicted at optimum conditions. It can be said that the prediction model had sufficient validity to simulate the CO_2 capture efficiency in the microchannel.

4 | CONCLUSIONS

In summary, by utilizing a T-shaped microchannel, a suitable interface was provided between gaseous phase of $CO_2 + N_2$ and liquid phase of $NH_3 + H_2O$ with the aim of CO₂ absorption by ammonia molecules. A response surface methodology based on CCD was implemented to study the effect of temperature (14°C-26°C), gas flow rate (80-140 mL/min) and ammonia concentration (0.08-0.16 mL/mL (8-16 vol%)) on the CO₂ capture efficiency. Among these variables, the ammonia concentration affected the CO₂ capture performance more strongly than the other variables because of the association between ammonia concentration and absorption capacity of the liquid phase. The CO2 capture efficiency was improved at low temperatures, low gas flow rates, and high ammonia concentrations within their defined ranges. A second-order polynomial model fitted well the experimental data and predicted the best efficiency of 96.48% CO₂ capture under the following optimum conditions: gas flow rate of 110.59 mL · min⁻¹, temperature of 20°C, and ammonia concentration of 0.1382 mL/mL (13.82 vol%). At the optimum conditions, by actual values, 95.42% CO₂ capture was observed. This good agreement between the predicted and observed response indicates the accuracy of the prediction model. These

results also demonstrate the superior performance of the used microchannel as a continuous two-phase contacting device. These microdevices have the advantage of ideal mixing, mass and heat transfer. For increasing the capacity for industrial purposes, microchannels should be cascaded in a parallel layout. This has the advantage of no change in the overall efficiency due to no change in the individual channel efficiency.

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