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## Absorption Rate of Carbon Dioxide from Gas Mixture

Jafar Ghani Majeed

Department of Materials Engineering, College of Engineering, Al-Mustansiriyah University, Baghdad, Iraq

**Abstract:** Absorption of CO<sub>2</sub> gas from CO<sub>2</sub>/Air mixture into aqueous sodium hydroxide solution has been achieved using packed column in pilot scale at constant temperature (T) of 25±1°C. The aim of the present work was to improve the Absorption rate of this process, to find the optimal operation conditions, and to contribute to the using of this process in the chemical industry. Absorption rate (RA) was measured by using different operating parameters: gas mixture flow rate (G) of 360-540 m<sup>3</sup>/h, carbon dioxide inlet concentration (CCO<sub>2</sub>) of 0.1-0.5 vol. %, NaOH solution concentration (CNaOH) of 1-2 M, and liquid holdup in the column (VL) of 0.022-0.028 m<sup>3</sup> according to experimental design. The measured RA was in the range of RA = 3.235 – 22.340 k-mol/h. Computer program (Statgraphics/Experimental Design) was used to estimate the fitted linear model of RA in terms of (G, CCO<sub>2</sub>, CNaOH, and VL), and the economic aspects of the process. R-squared of RA model was 91.7659 percent, while the standard error of the estimate shows the standard deviation of the residuals to be 1.7619. The linear model of RA was adequate, the operating parameters were significant except the liquid holdup was not significant, and the interactions were negligible.

**Keywords:** Packed column; CO<sub>2</sub> absorption; NaOH solution; Absorption rate.

### 1. Introduction

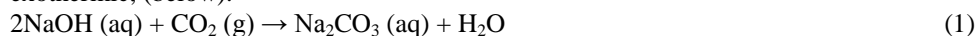
Carbon dioxide is one of the more frequent found gases on the earth. It is a main product in combustion processes and the natural metabolism of living organisms. Carbon dioxide discharged by the consumption of fossil fuel for power and heat production is said to occupy nearly 35- 40 % of the amount of discharged CO<sub>2</sub> all over the world [1-3]. Environmental issues caused by exhaust greenhouse gases (GHG) and toxics have become global problems. Through the past studies of five decades, increased GHG levels in atmosphere is believed to cause global warming, in which CO<sub>2</sub> is the largest contributors. International Panel on Climate Change (IPCC) predicts that the CO<sub>2</sub> content in atmosphere may contain up to 570 ppm CO<sub>2</sub>, causing an increase in mean global temperature around 1.9°C and an increase mean sea level of 38 m [4]. Also accompanied is species extinction.

Indoor CO<sub>2</sub> levels usually vary between 400 and 2000 ppm (parts per million). Outdoor CO<sub>2</sub> levels are usually 350 - 450 ppm. Heavily industrialized or contaminated areas may periodically have a CO<sub>2</sub> concentration of up to 800 ppm. The levels of outdoor CO<sub>2</sub> are higher in areas where traffic is very heavy.

CO<sub>2</sub> must not be confused with carbon monoxide (CO), a very toxic gas that is a by-product from poor combustion in i.e. cars and fireplaces. Carbon monoxide is dangerous at very low concentrations (25 to 50 ppm). In industrial environments where process generated CO<sub>2</sub> dominates (or CO<sub>2</sub> not generated by people), for example in breweries, packaging industry, freezer storages etc, the maximum Permitted CO<sub>2</sub> concentration according to most standards is as high as 5000 ppm during an 8-hour working period. You will not find such high levels in a home or in an office environment where people are the main source of carbon dioxide.

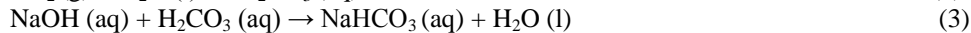
Several methods have been proposed to remove CO<sub>2</sub> from the air, or from flue gases, such as chemical absorption, physical absorption, membrane separation, biochemical methods, and the catalytic conversion method. In addition to these methods, the absorption of carbon dioxide in an alkaline solution with crystallization has also been adopted to explore the removal of carbon dioxide from waste gas [5]. These usually involve using a variant of the Kraft process. Scrubbing processes may be based on sodium hydroxide [6-8]. The CO<sub>2</sub> is absorbed into solution, transferred to lime via a process called causticization and released in a kiln.

Some researchers outlined a specific methods of air capture [9-13]. CO<sub>2</sub> is absorbed by an alkaline NaOH solution to produce dissolved sodium carbonate. The absorption reaction is a gas liquid reaction, strongly exothermic, (below):

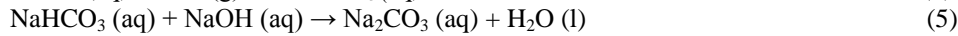


The mechanism of reaction between the acidic oxide and the alkali depends on the concentration of the alkali solution.

(a) When the alkali (NaOH) solution is very dilute ( $\text{pH} < 8$ ), carbon dioxide first reacts with water to form carbonic acid ( $\text{H}_2\text{CO}_3$ ) slowly. The acid thus formed then reacts with the alkali to give sodium hydrogen carbonate ( $\text{NaHCO}_3$ ):



(b) When the alkali solution is a fairly concentrated one ( $\text{pH} > 10$ ), carbon dioxide directly reacts with it to form the bicarbonate, which further reacts with the alkali to form sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) as the main product by complete neutralization:



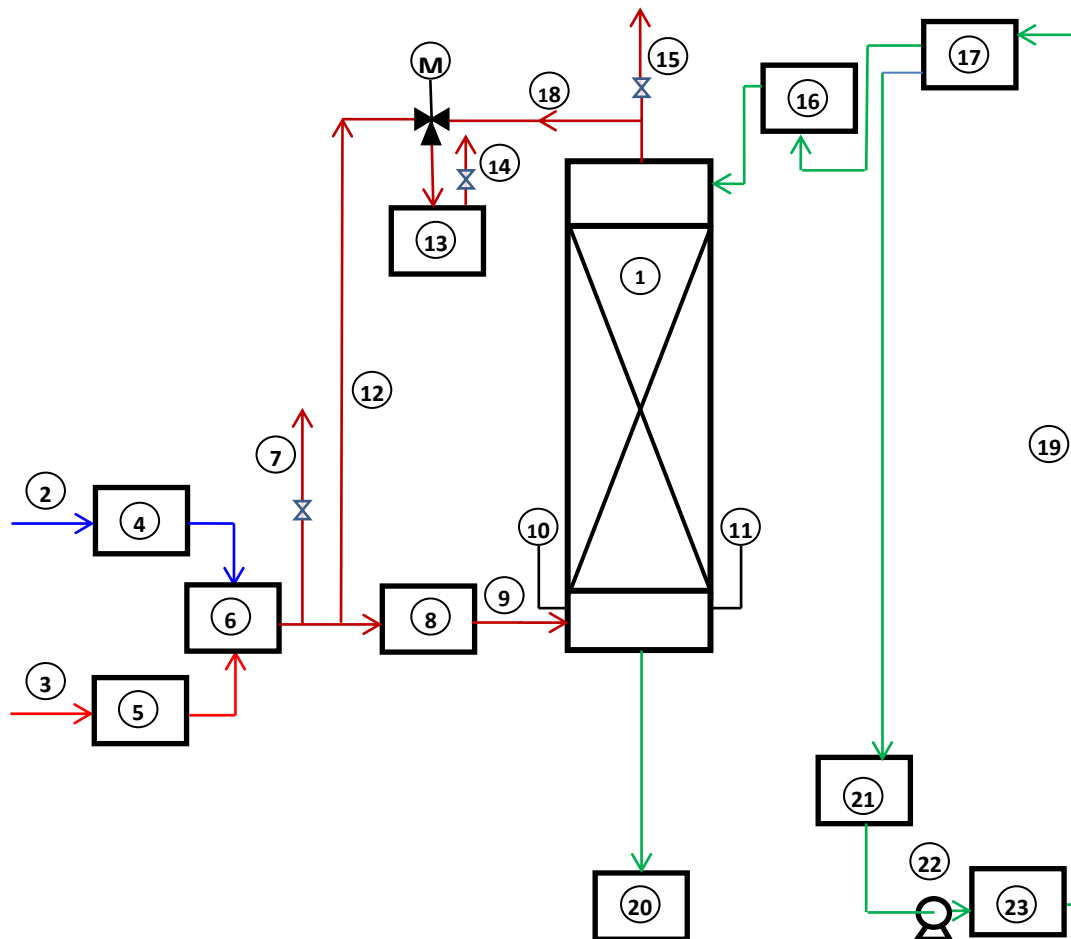
Thus, only when the concentration of the alkali solution is quite low, the reaction proceeds via the formation of carbonic acid. But the acidic oxide is not completely neutralized in this case.

## 2. Experimental Work

### 2.1. Experimental Apparatuses

The main equipment of the experimental apparatus as shown in Figure 1 is the packed column (1), with the size of 2 m height and 0.2 m diameter. The main complementary apparatus and pipe lines are as follows: Compressed air (2), carbon dioxide gas (3), flow meter for compressed air (4), flow meter for  $\text{CO}_2$  gas (5), mixing gas chamber (6), gas mixture vent (7), thermostat (8), gas mixture into packed column (9), temperature indicator (10), pH indicator (11), gas mixture to  $\text{CO}_2$  analyzer (12),  $\text{CO}_2$  analyzer (13), gas vent (14), gas mixture out (15), flow meter for aqueous NaOH solution (16), head tank (17), outlet gas mixture to  $\text{CO}_2$  gas analyzer (18), aqueous sodium hydroxide solution into packed column (19), product tank (20), aqueous sodium hydroxide solution feed tank (21), feed pump (22), thermostat (23).

**Figure-1.** Schematic diagram of the experimental apparatus for  $\text{CO}_2$  gas absorption from  $\text{CO}_2/\text{Air}$  gas mixture into aqueous NaOH solution in packed column.



## 2.2. Operating Parameters

The preliminary experiments were carried out to absorb of CO<sub>2</sub> gas from gas mixture into aqueous NaOH solution in pilot scale packed column using experimental apparatus as shown in Figure 1 to find the proper operating parameters would be used in this work. Operating variable parameters were as follows:

- \* Gas mixture flow rate ( $G$ ): 360-540 m<sup>3</sup>/h.
- \* CO<sub>2</sub> gas inlet concentration ( $C_{CO_2}$ ): 0.1-0.5 vol. %.
- \* Aqueous NaOH solution concentration ( $C_{NaOH}$ ): 1-2 M.
- \* Liquid temperature ( $T$ ): 25±1°C.
- \* Liquid holdup in the column ( $V_L$ ): 0.022-0.028 m<sup>3</sup>.

## 2.3. Absorption of CO<sub>2</sub> from CO<sub>2</sub>/Air Gas Mixture

Experiments of absorption of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture into aqueous NaOH solution have been carried out using the mentioned various operating parameters by using experimental apparatus as shown in Figure 1, according to experimental design plan seen in Table 1. The flow rates of compressed air (2) and CO<sub>2</sub> gas (3) from gas cylinder were measured by flow meters (4) and (5) respectively, and enter the mixing chamber (6). The gas mixture (9) enters the packed column (1) through the thermostat (8) from lower part. The gas mixture via the gas line (12) enters CO<sub>2</sub> gas analyzer (13) to measure CO<sub>2</sub> inlet concentration in the gas mixture. Aqueous NaOH solution from NaOH solution feed tank (21) by feed pump (22) enters the upper part of the column through the liquid feeding line (19). The thermostat (23) is maintained the desired liquid temperature constant in packed column (1) during the all experiments runs. One part of gas mixture from the top of column through the gas mixture line (18) was taken into CO<sub>2</sub> gas analyzer (13) to measure outlet CO<sub>2</sub> gas concentration, while the other part of gas mixture discharge to atmosphere via the gas mixture line (15). In the column, the liquid temperature measured by temperature gage (10), while the pH of the liquid is measured by digital pH-meter (11). The resultant reaction product Na<sub>2</sub>CO<sub>3</sub> solution from downer part of column sent to solution product tank (20).

Table-1. Experimental design plan for absorption of CO<sub>2</sub> into aqueous NaOH solution.

Run No.	Gas mixture flow rate ( $G$ )	CO <sub>2</sub> gas inlet concentration ( $C_{CO_2}$ )	NaOH solution concentration ( $C_{NaOH}$ )	Liquid holdup in the column ( $V_L$ )
	(m <sup>3</sup> /h)	(vol. %)	(M)	(m <sup>3</sup> )
1	450	0.3	1.5	0.025
2	540	0.5	2	0.022
3	360	0.5	1	0.028
4	360	0.1	1	0.022
5	360	0.5	2	0.022
6	540	0.5	2	0.028
7	360	0.1	1	0.028
8	450	0.3	1.5	0.025
9	360	0.1	2	0.028
10	450	0.3	1.5	0.025
11	360	0.1	2	0.022
12	540	0.1	1	0.028
13	360	0.5	2	0.028
14	360	0.5	1	0.022
15	540	0.1	2	0.022
16	540	0.5	1	0.022
17	540	0.1	1	0.022
18	540	0.1	2	0.028
19	540	0.5	1	0.028

## 2.4. Definition of CO<sub>2</sub> Absorption Rate

The CO<sub>2</sub> inlet flow rate ( $G_{in}$ ) is calculated using the following equation:

$$G_{in} = [(Q_{air} \times D_{air}/M_{air}) / (1 - Y_{CO_2, in})] Y_{CO_2, in} \quad (6)$$

Where,

$G_{in}$  = CO<sub>2</sub> inlet flow rate, k-mol/h.

$Q_{air}$  = Air volumetric flow rate, m<sup>3</sup>/h.

$D_{air}$  = Density of air, kg/m<sup>3</sup> at standard conditions.

$M_{air}$  = Molecular weight of air, kg/k-mol.

$Y_{CO_2, in}$  = CO<sub>2</sub> mole fraction at inlet of packed column, vol. %.

The CO<sub>2</sub> outlet flow rate ( $G_{out}$ ) is calculated using the following equation:

$$G_{out} = [(Q_{air} \times D_{air} / M_{air}) / (1 - Y_{CO_2, out})] Y_{CO_2, out} \quad (7)$$

Where,

$G_{out}$  = CO<sub>2</sub> outlet flow rate, k-mol/h.

$Y_{CO_2, out}$  = CO<sub>2</sub> mole fraction at outlet of packed column, vol. %.

The CO<sub>2</sub> absorption rate ( $R_A$ ) is the difference between the inlet and outlet CO<sub>2</sub> flow rates [14 -17]:

$$R_A = G_{in} - G_{out} \quad (8)$$

Where,

$R_A$  = CO<sub>2</sub> absorption rate, k-mol/h.

### 3. Results and Discussion

#### 3.1. Absorption Rate Calculation

The absorption of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture into aqueous sodium hydroxide (NaOH) solution were carried out according to experimental design plan in Table 1 with the variation of gas mixture flow rate ( $G$ ), CO<sub>2</sub> gas inlet concentration ( $C_{CO_2}$ ), aqueous NaOH solution concentration ( $C_{NaOH}$ ), and liquid holdup in the column ( $V_L$ ).

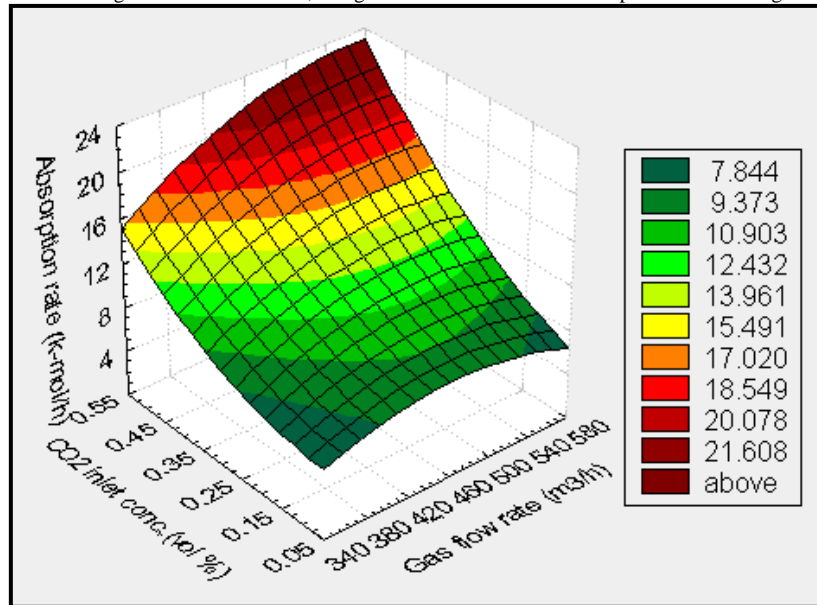
Absorption rate ( $R_A$ ) was calculated by using equation (4). The  $R_A$  of CO<sub>2</sub> gas from gas mixture CO<sub>2</sub>/Air was in the range of  $R_A = 3.235 - 22.340$  k-mol/h. The results of  $R_A$  are summarized in Table 2:

**Table-2.** Data base of experimental design and results of CO<sub>2</sub> gas absorption from gas mixture CO<sub>2</sub> /Air into aqueous NaOH solution.

Run No.	Gas mixture flow rate ( $G$ )	CO <sub>2</sub> gas inlet concentration ( $C_{CO_2}$ )	NaOH solution concentration ( $C_{NaOH}$ )	Liquid holdup in the column ( $V_L$ )	Absorption rate
	(m <sup>3</sup> /h)	(vol. %)	(M)	(m <sup>3</sup> )	(k-mol/h)
1	450	0.3	1.5	0.025	12.350
2	540	0.5	2	0.022	21.204
3	360	0.5	1	0.028	14.260
4	360	0.1	1	0.022	3.235
5	360	0.5	2	0.022	16.616
6	540	0.5	2	0.028	22.340
7	360	0.1	1	0.028	7.543
8	450	0.3	1.5	0.025	13.564
9	360	0.1	2	0.028	10.454
10	450	0.3	1.5	0.025	11.895
11	360	0.1	2	0.022	7.430
12	540	0.1	1	0.028	5.550
13	360	0.5	2	0.028	14.650
14	360	0.5	1	0.022	13.414
15	540	0.1	2	0.022	9.450
16	540	0.5	1	0.022	18.731
17	540	0.1	1	0.022	7.308
18	540	0.1	2	0.028	11.368
19	540	0.5	1	0.028	20.515

The effects of operating parameters on absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture could be seen as in the following Figures:

**Figure-2.** Effect of CO<sub>2</sub> gas inlet concentration, and gas mixture flow rate on Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air mixture.



**Figure-3.** Effect of NaOH concentration, and gas mixture flow rate on Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air mixture.

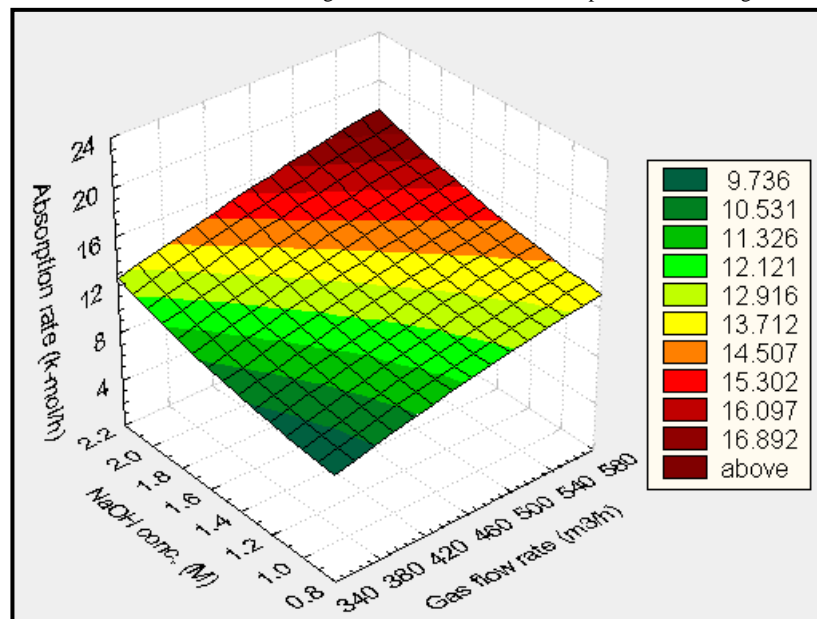


Figure-4. Effect of liquid holdup, and gas mixture flow rate on Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air mixture.

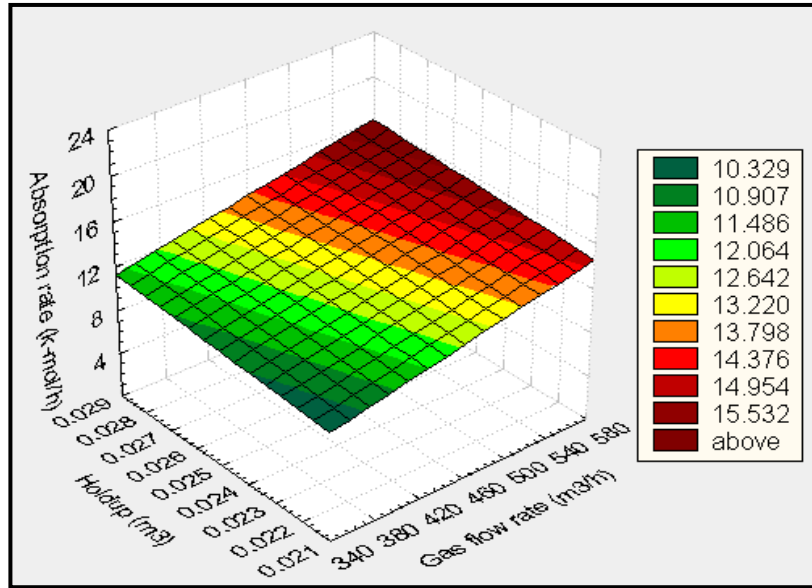


Figure-5. Effect of aqueous NaOH concentration, and CO<sub>2</sub> inlet concentration on Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air mixture.

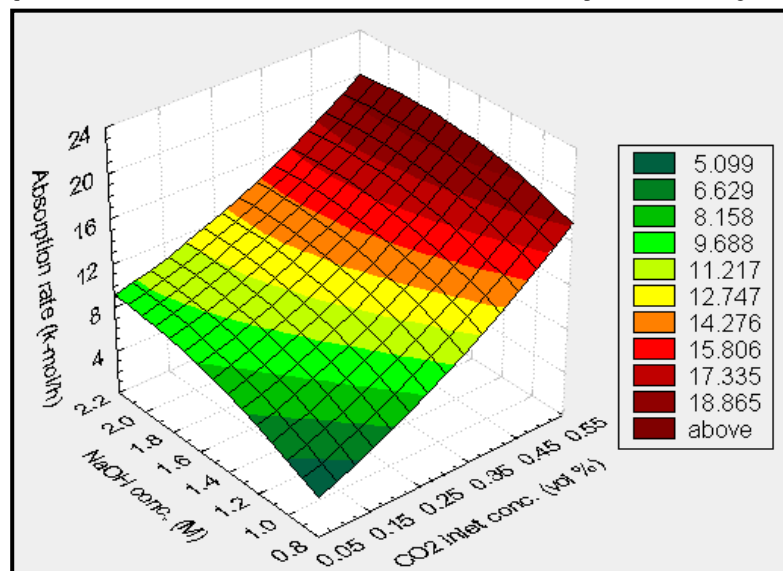


Figure-6. Effect of aqueous NaOH concentration, and liquid holdup on Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air mixture.

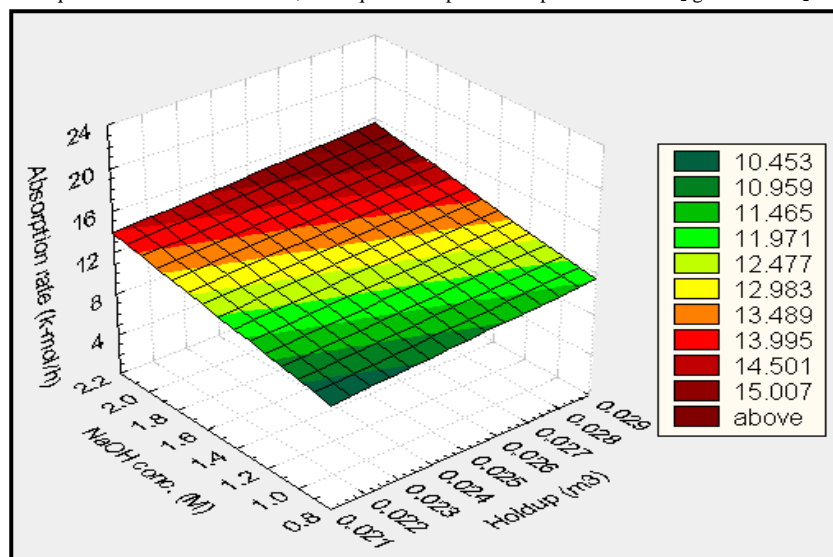
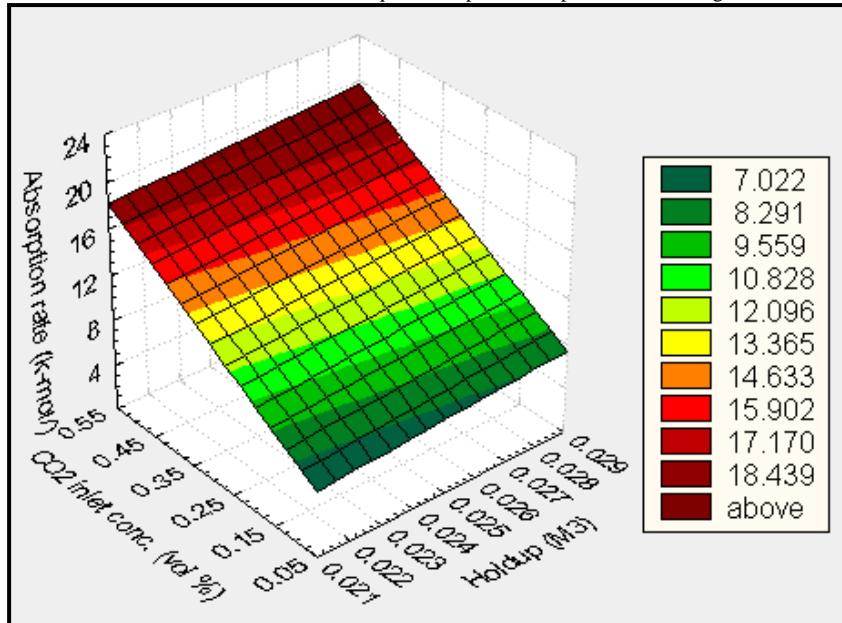


Figure-7. Effect of CO<sub>2</sub> inlet concentration, and liquid holdup on Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air mixture.



The 3 D sequential Graphs/Box plot and Block plot for Absorption rate ( $R_A$ ) with independent parameters: gas mixture flow rate ( $G$ ), inlet CO<sub>2</sub> gas concentration ( $C_{CO_2}$ ), aqueous NaOH solution ( $C_{NaOH}$ ), and liquid holdup ( $V_L$ ) as in the following figures:

Figure-8. 3 D sequential Graph/Box plot for Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air mixture with operating parameters.

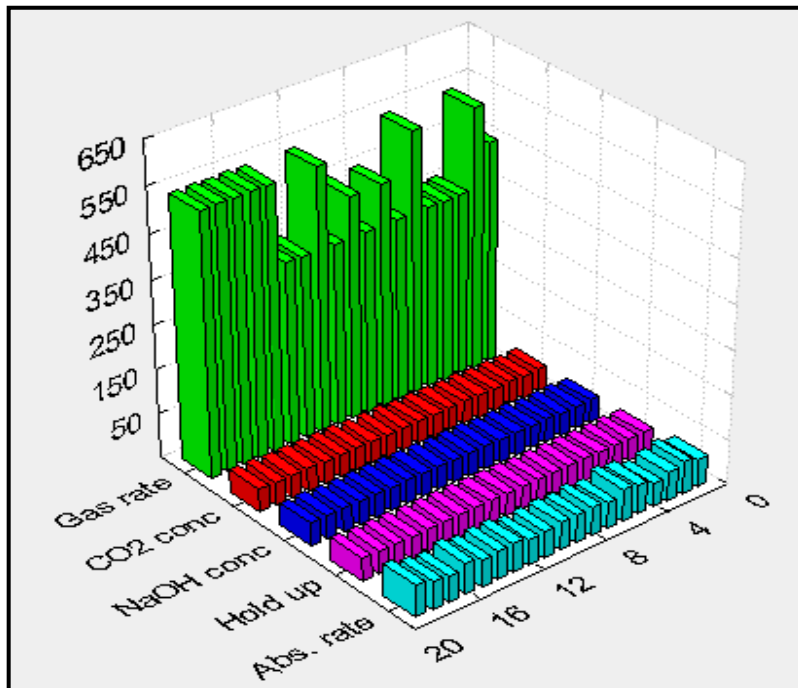
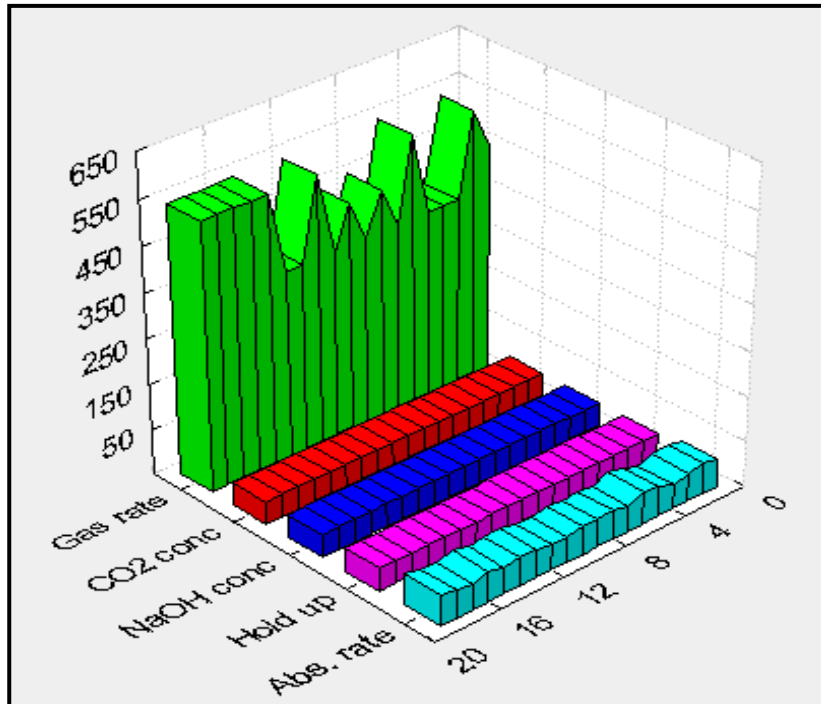


Figure-9. 3 D sequential Graph/Block plot for Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air mixture with operating parameters.

From Figure 2 – Figure 9, the Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture ( $R_A$ ) is increased with the increase in gas mixture flow rate ( $G$ ), inlet CO<sub>2</sub> gas concentration ( $C_{CO_2}$ ), aqueous NaOH solution ( $C_{NaOH}$ ), and liquid holdup ( $V_L$ ). The biggest effect of operating parameters on Absorption rate was  $C_{CO_2}$ , and smallest one was  $V_L$ .

### 3.2. Correlation Model of Absorption Rate

Computer program (Statgraphics/Experimental Design) was used to estimate the fitted linear model of absorption rate ( $R_A$ ) for CO<sub>2</sub> gas absorption from CO<sub>2</sub>/air gas mixture into aqueous NaOH solution in packed column in terms of operating parameters:  $G$ ,  $C_{CO_2}$ ,  $C_{NaOH}$ , and  $V_L$ . The output shows the results of fitting a multiple linear regression model to describe the relationship between absorption rate and 4 independent variables.

The equation of the fitted model is:

$$R_A = -12.877 + 0.020 * G + 24.810 * C_{CO_2} + 2.87 * C_{NaOH} + 193.583 * V_L \quad (9)$$

Since the P-value in the ANOVA Table 3 is less than 0.05, there is a statistically significant relationship between the variables at the 95.0% confidence level.

The R-Squared statistic indicates that the model as fitted explains 91.7659 % of the variability in Absorption rate. The adjusted R-squared statistic, which is more suitable for comparing models with different numbers of independent variables, is 89.4133 %. The standard error of the estimate shows the standard deviation of the residuals to be 1.7619. This value can be used to construct prediction limits for new observations by selecting the Reports option from the text menu. The mean absolute error (MAE) of 1.17437 is the average value of the residuals. The Durbin-Watson (DW) statistic tests the residuals to determine if there is any significant correlation based on the order in which they occur in your data file. Since the P-value is greater than 0.05, there is no indication of serial autocorrelation in the residuals at the 95.0% confidence level.

In determining whether the model can be simplified, notice that the highest P-value on the independent variables is 0.2085, belonging to Liquid holdup. Since the P-value is greater or equal to 0.05, that term is not statistically significant at the 95.0 % or higher confidence level. Consequently, you should consider removing Liquid holdup from the model.

Table-3. ANOVA

Parameter	Estimate	Standard Error	T Statistic	P-value
Constant	-12.8765	4.54644	-2.83221	0.0133
CO <sub>2</sub> concentration	24.810	2.20238	11.2651	0.0000
Gas flow rate	0.0200444	0.00489417	4.09557	0.0011
Liquid holdup	193.583	146.825	1.31846	0.2085
NaOH concentration	2.8695	0.880951	3.25727	0.0057

The validity range for the model in equation (9) is:

$$360 \leq G \leq 540 \text{ m}^3/\text{h}$$

$$0.022 \leq V_L \leq 0.028 \text{ m}^3$$

$$1 \leq C_{NaOH} \leq 2 \text{ M}$$

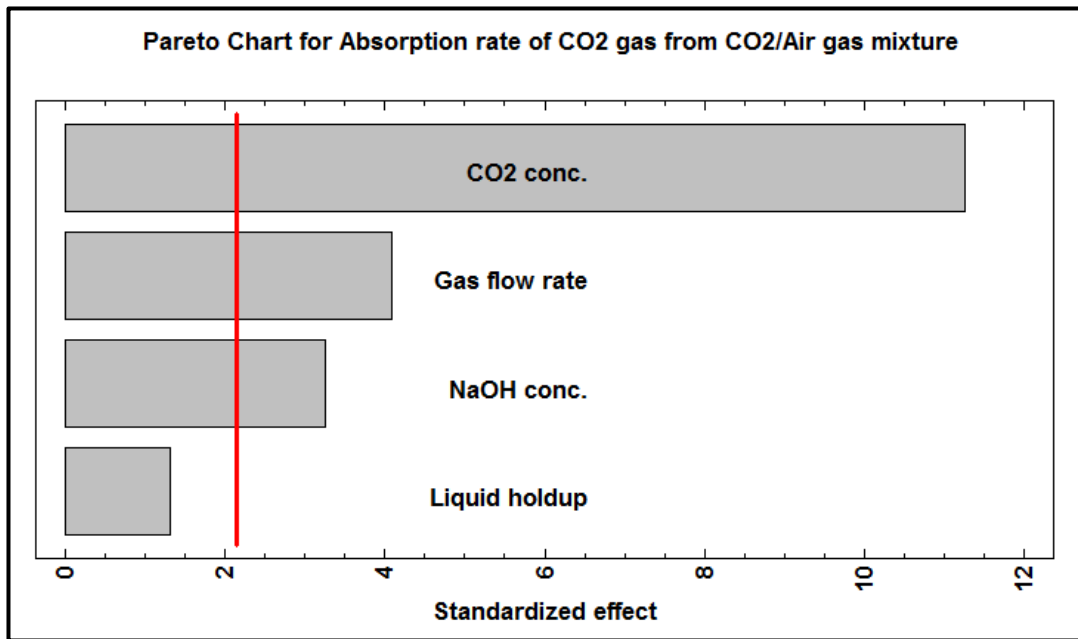
$$0.1 \leq C_{CO_2} \leq 0.5 \text{ vol. \%}$$

The accuracy of the  $R_A$  model is  $\mp 1.7619 \%$ .

The linear model in equation (9) is adequate, and the operating parameters were significant except  $V_L$  and were in ordered of  $C_{CO_2} > G > C_{NaOH} > V_L$ . The interactions of operating parameters were negligible.

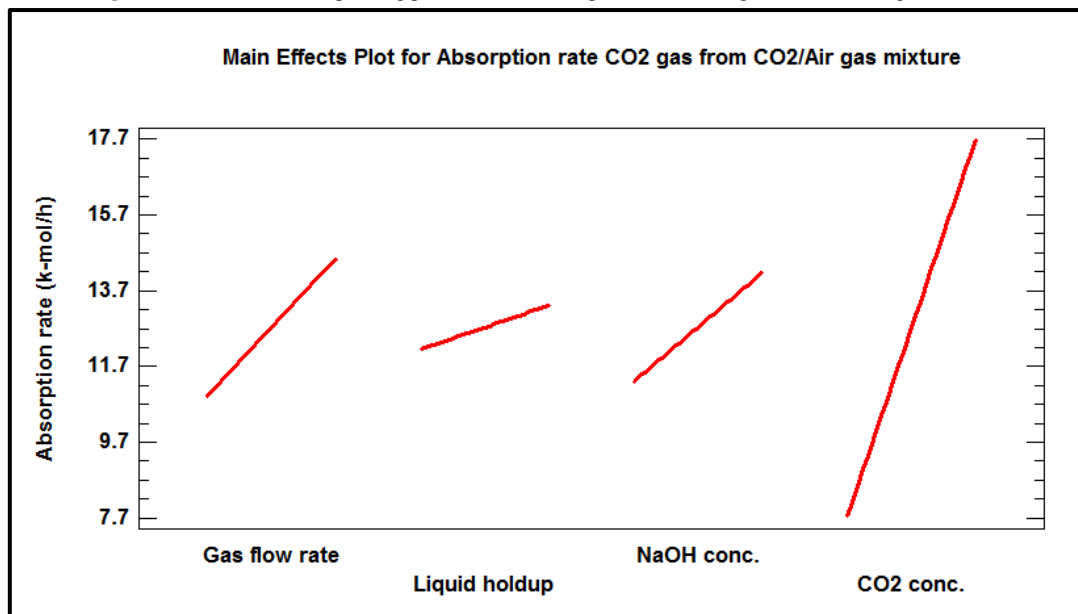
The Pareto Chart of Absorption rate ( $R_A$ ) of  $CO_2$  gas from  $CO_2$ /Air gas mixture could be seen in Figure10:

Figure-10. Pareto chart for Absorption rate of  $CO_2$  gas from  $CO_2$ /Air gas mixture.

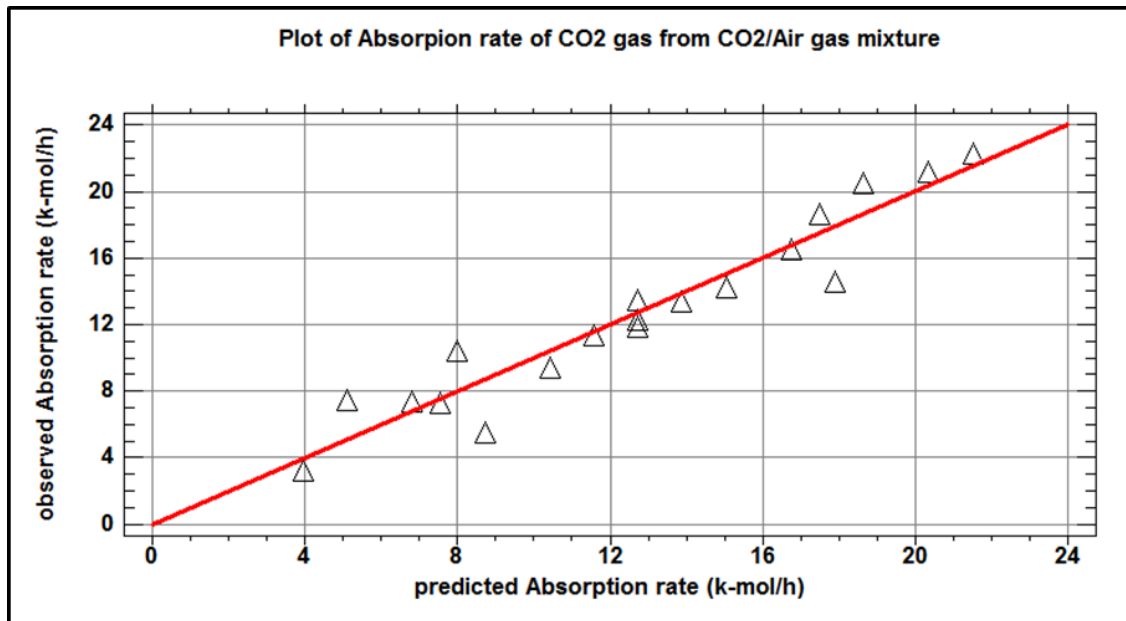


The main effects of operating parameters on Absorption rate of  $CO_2$  gas from gas mixture  $CO_2$ /Air is seen in following Figure 11:

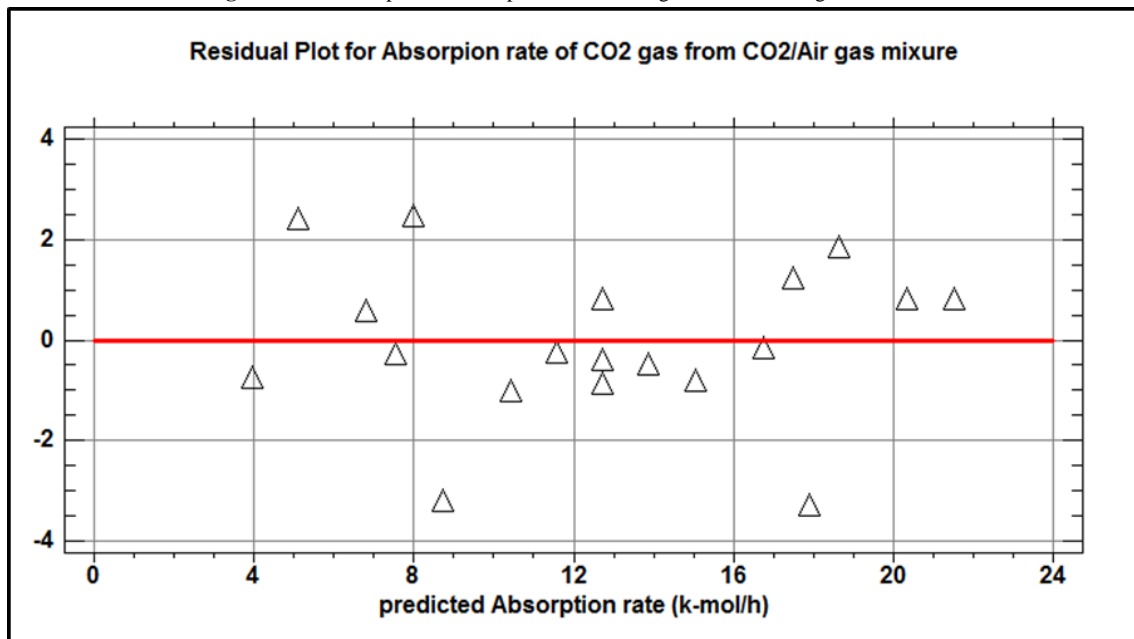
Figure-11. Main effects of operating parameters on Absorption rate of  $CO_2$  gas from  $CO_2$ /Air gas mixture.



The observed and predicted Absorption rate of  $CO_2$  gas from  $CO_2$ /Air gas mixture represents in following Figure 12:

Figure-12. Observed vs. predicted Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture.

The residual plot for Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture would be seen in Figure 13:

Figure-13. Residual plot for Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture.

### 3.3. Optimal Response

The optimum operating parameters for present work were obtained using the computer program to analyze the experimental results. The goal of optimizing was to maximize the Absorption rate ( $R_A$ ) of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture. The results of optimizing were summarized in Table 4:

Table-4. Optimum operating parameters and optimum Absorption rate of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture.

Operating parameters	Low value	High value	Optimum value
Gas mixture flow rate (m <sup>3</sup> /h):	360	540	540
CO <sub>2</sub> gas inlet conc. (vol. %):	0.1	0.5	0.5
NaOH solution conc. (M):	1	2	2
Liquid hold up in column (m <sup>3</sup> ):	0.022	0.028	0.028
Absorption rate of CO <sub>2</sub> gas from CO <sub>2</sub> /Air gas mixture (k-mol/h):			21.5119

## 4. Conclusions

\* Absorption of CO<sub>2</sub> gas from CO<sub>2</sub>/Air gas mixture into NaOH solution was carried out in pilot scale packed column. The Absorption rate ( $R_A$ ) of CO<sub>2</sub> gas was measured at various operating conditions ( $G$ ,  $C_{CO_2}$ ,  $C_{NaOH}$ , and  $V_L$ ) according to experimental design. The measured Absorption rate was in the range of  $R_A = 3.235 - 22.340$  k-mol/h. The  $R_A$  could be improved and increases by increasing in the  $G$ ,  $C_{CO_2}$ ,  $C_{NaOH}$ , and  $V_L$ .

\* A computer program (Statgraphics/Experimental Design) was used to estimate the linear fitted model of Absorption rate ( $R_A$ ) in terms of operating conditions ( $G$ ,  $C_{CO_2}$ ,  $C_{NaOH}$ , and  $V_L$ ). The linear fitted model of  $R_A$  was adequate, and operating parameters were significant, while the interactions were negligible. The accuracy of Absorption rate model is  $\pm 1.7619$  %.

\* Using the same computer program the optimum operating conditions were obtained with values of  $G = 540$  m<sup>3</sup>/h,  $C_{CO_2} = 0.5$  vol. %,  $C_{NaOH} = 2$  M, and  $V_L = 0.028$  m<sup>3</sup>. The optimum Absorption rate ( $R_A$ ) was in value of **21.5119 k-mol/h**.

\* On the base of results of measured Absorption rate, we conclude to scaling up the size of the pilot plant used in present work by **3-4** times to commercial size plant and using the optimum operating parameters obtained in future work.

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