



Experiments and Simulations Data for the Absorption of CO₂ into Aqueous Solutions of Monoethanolamine in a Bench Scale Absorption Unit

Experimentos y datos de simulaciones para la absorción de CO₂ en soluciones acuosas de monoetanolamina a escala laboratorio

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Abstract

Experiments and simulations were carried out in a bench scale absorption unit; the runs were made using solutions of monoethanolamine (MEA) at 25 wt%. The gas used was a mixture of air and CO₂. Runs were conducted using three different gas flow rate, in a range between 40 to 190 NL/min, and the flow rate of MEA solution was varied to have different ratios of CO₂:MEA, in a range between 1:2 to 1:5, for each flow rate. The results are compared with those obtained with simulations using Aspen Hysys software with a constant Murphree efficiency value of 24%, and varying it for those values calculated from experiments. In this work it was found that bench scale absorption unit has a percentage of absorption up to 99% for a CO₂:MEA ratio of 1:4.3.

Keywords: CO₂, MEA, absorption, simulations, bench scale.

Resumen

Se realizaron experimentos y simulaciones en una columna de absorción a escala laboratorio, los experimentos se efectuaron utilizando solución de monoetanolamina a 25 wt% y una mezcla de aire y CO₂. Los experimentos se llevaron a cabo usando tres diferentes flujos de gases, en un rango de 40 a 190 NL/min. El flujo de solución de amina fue variando para cada flujo de gas con la finalidad de tener diferentes relaciones CO₂:MEA, en un rango de relaciones de 1:2 hasta 1:5. Los resultados experimentales se comparan con los obtenidos en las simulaciones de los mismos utilizando el software Aspen Hysys con una constante de Murphree igual a 24%, también se emplearon variaciones de esta, ajustando el valor por los calculados en los experimentos. En este trabajo se encontró que la unidad de absorción a escala laboratorio tiene un porcentaje de absorción hasta de 99% para una relación de CO₂:MEA de 1:4.3.

Descriptores: CO₂, MEA, absorción, simulaciones, escala laboratorio.

INTRODUCTION

The removal of CO₂ from gas streams can be achieved by a number of separation techniques including absorption into a liquid solvent, adsorption into a solid, cryogenic separation, permeation through membranes, and chemical conversion. Among these techniques, absorption into a liquid solvent is the most suitable process for removing CO₂ from high-volume flue gas streams.

The commonly used solvents are aqueous solutions of alkanolamines, such as monoethanolamine (MEA), diethanolamine (DEA), diisopropanolamine (DIPA), and methyldiethanolamine (MDEA) (Maddox, 1984; Kohl and Nielsen, 1997). Among these solvents, MEA is the most widely used because it has a faster rate of reaction with CO₂, which allows absorption to take place in a shorter column.

This work focuses on MEA, running both, experiments and simulations, for simulations was used the Aspen HYSYS software which has an estimation method to estimate the Murphree efficiencies in plate columns based on pseudo first order conditions. The estimation method is based on the work of Tomcej (Tomcej *et al.*, 1987), modified later by Rangwala (Rangwala *et al.*, 1992). In a plate column, an efficiency value is estimated for each plate, in a packed column, a packing height of e.g. 1 meter can be defined as one stage with a Murphree efficiency.

Mofarahi *et al.* (2008) have simulated an atmospheric CO₂ absorption and desorption process for the amines MEA, DEA, MDEA and diglycolamine (DGA) using

Matlab with a Kent- Eisenberg equilibrium model. In their calculations, a Murphree efficiency was specified to 35% for each tray, and this is regarded as very optimistic, other authors as Øi, Lars Erik (Øi, L E, 2011) used 10-28% Murphree efficiency for 1 meter of structured packing compared to 35% which was regarded as optimistic in earlier calculations, 15% is a value between typical top and bottom conditions.

In the present work, the disagreement found between experiments and simulations were attributed mainly to the Murphree Efficiency (E_M), since in the first simulations was used a constant E_M value of 24%. Therefore, the objective of this work, was to calculate de E_M from the experiments and used these values in the simulations, achieving a better agreement between them.

EXPERIMENTAL

BENCH SCALE ABSORPTION UNIT

The absorber is made of borosilicate glass Duran 3.3, with internal diameter of 4" and height of 1.70 m, the packing used was 5/8" polypropylene pall ring; the total height of the packing is 1.20 m. There is a tank holding the MEA solution where is pumped to the top of the column, the flow rate of the solution was calibrated manually with a stopwatch and volumetric flask using frequency converter for the pump. There is a Horiba gas analyzer Model ES510. Figure 1 shows a schematic diagram of the absorption unit. The experiments were carried out at Normal conditions of Temperature and Pressure, Normal Liters (NL).

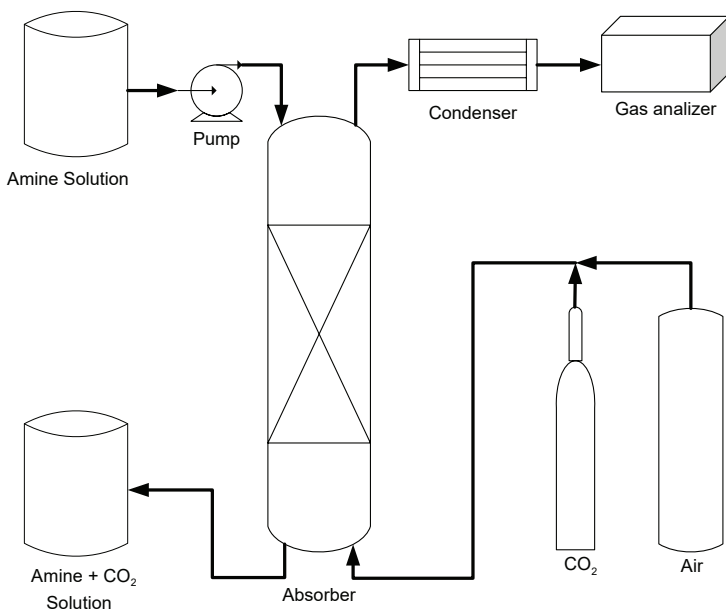


Figure 1. Schematic diagram of gas absorption unit

To start the experiments, the gas was fed from the bottom of the column at 22°C and 34.47 mbar and its composition was measured using the Horiba gas analyzer at the outlet on the top with a temperature of 40°C. The MEA solution is added to the column from the top with a temperature of 28°C and it is leaving the absorber with a temperature of 50°C.

RESULTS AND DISCUSSION

Several experiments were carried out using three different gas flow rates (41.5, 104 and 184 NL/min) with different concentration of air/CO₂. For each gas flow rate, was used a MEA solution 25%wt at different flow rates. At the same time simulations of the experiments were conducted to compare the results. The simulations were done in Aspen Hysys V7.3 using the 'Kent Eisenberg' thermodynamic package of the Amines Property Package, as suggests (Kent and Eisenberg, 1976; Aspen Technology, 2011). Table 1 to 3 show the parameters used during the experiments and the results obtained from the experiment and simulation.

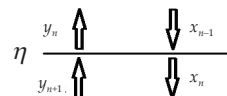
From the results it is possible to see how the %CO₂ drops dramatically when the MEA solution is added and continuous dropping with the increment of MEA solution, as it is expected. However, the results obtained in the simulations do not match with those from the experiments. Looking for an answer for these differences, it was found that the software use efficiency for the structure packing of 24% and it remains always the

same being it independent of the flow and concentration used.

Considering that the efficiency varies with the flow rate and the concentration, the efficiency was calculated for each experiment using the Murphree equation, eq. 1

$$E_M = \frac{y_n - y_{n+1}}{Kx_n - y_{n+1}} \tag{1}$$

where



- E_M = Murphree efficiency
- y = molar gas composition
- x = molar liquid composition
- K = equilibrium constant liquid-gas

The Murphree efficiency (E_M) values obtained for each experiment were then used in the simulations, giving closer results with those for the experiments.

On the other hand, the absorption efficiency was also calculated, for these case, the next equation was used, eq. 2

$$\eta_A = \left[1 - \left(\frac{y_{out}}{1 - y_{out}} \right) \left(\frac{1 - y_{in}}{y_{in}} \right) \right] \times 100 \tag{2}$$

Table 1. Experiments with gas flow rate of 41.6 NL/min

%CO ₂	MEA, ml/min	Mol CO ₂	Mol MEA	%CO ₂ out exp	%CO ₂ out sim
55.55	0.0	1	0.0	55.55	55.55
55.55	490.0	1	1.97	12.6	5.0
55.55	594.0	1	2.40	7.0	4.3
55.55	755.0	1	3.00	5.1	4.0
55.55	874.0	1	3.50	3.5	4.0
55.55	1072.0	1	4.30	1.0	4.0

Table 2. Experiments with gas flow rate of 104 NL/min

%CO ₂	MEA, ml/min	Mol CO ₂	Mol MEA	%CO ₂ out exp	%CO ₂ out sim
12.5	0.0	0.57	0.0	12.5	12.5
12.5	238.0	0.57	1.0	6.0	1.50
12.5	295.0	0.57	1.2	4.8	0.55
12.0	410.0	0.56	1.7	2.9	0.60
12.5	476.0	0.57	1.9	3.0	0.55
12.0	675.0	0.56	2.8	2.2	1.00

Table 3. Experiments with gas flow rate of 184 NL/min

%CO ₂	MEA, ml/min	Mol CO ₂	Mol MEA	%CO ₂ out exp	%CO ₂ out sim
12.5	0.0	1.0	0.0	12.5	12.5
12.5	374.0	1.0	1.5	5.0	2.10
12.2	479.0	1.0	2.0	4.4	0.93
12.5	607.0	1.0	2.5	4.4	0.73
12.5	715.0	1.0	3.0	4.0	0.68
12.5	715.0	1.0	3.0	3.8	0.68
12.2	715.0	1.0	3.0	4.0	0.68
12.2	1231.0	1.0	5.1	3.2	1.21

where

η_A = absorption efficiency

y_{Out} = exit gas molar composition

y_{In} = input gas molar composition

The results obtained using the Murphree efficiency in the simulations as well as the absorption efficiency of the experiments are now shown in Tables 4 to 6.

Table 4. Experiments with gas flow rate of 41.6 NL/min

%CO ₂ in	MEA, ml/min	Mol CO ₂	Mol MEA	%CO ₂ out exp	%CO ₂ out sim	% η_A	% E _M	%CO ₂ sim. with E _M
55.55	490.0	1	1.97	12.6	5.0	88.44	18.3	10.30
55.55	594.0	1	2.40	7.0	4.3	93.96	22.4	5.75
55.55	755.0	1	3.00	5.1	4.0	95.69	23.4	4.64
55.55	874.0	1	3.50	3.5	4.0	97.09	25.7	3.32
55.55	1072.0	1	4.30	1.0	4.0	99.19	33.8	0.83

Table 5. Experiments with gas flow rate of 104 NL/min

%CO ₂ in	MEA, ml/min	Mol CO ₂	Mol MEA	%CO ₂ out exp	%CO ₂ out sim	% η_A	% E _M	%CO ₂ sim. with E _M
12.5	238.0	0.57	1.0	6.0	1.50	56.9	9.50	4.1
12.5	295.0	0.57	1.2	4.8	0.55	65.5	11.00	3.7
12.0	410.0	0.56	1.7	2.9	0.60	78.0	14.60	1.8
12.5	476.0	0.57	1.9	3.0	0.55	78.4	16.00	1.9
12.0	675.0	0.56	2.8	2.2	1.00	83.5	18.20	1.1

Table 6. Experiments with gas flow rate of 184 NL/min

%CO ₂ in	MEA, ml/min	Mol CO ₂	Mol MEA	%CO ₂ out exp	%CO ₂ out sim	% η_A	% E _M	%CO ₂ sim. with E _M
12.5	374.0	1.0	1.5	5.0	2.10	62.12	10.00	3.4
12.2	479.0	1.0	2.0	4.4	0.93	66.88	10.28	3.3
12.5	607.0	1.0	2.5	4.4	0.73	67.78	11.20	3.2
12.5	715.0	1.0	3.0	4.0	0.68	70.83	11.80	3.0
12.5	715.0	1.0	3.0	3.8	0.68	72.34	11.80	3.0
12.2	715.0	1.0	3.0	4.0	0.68	70.00	11.45	2.9
12.2	1231.0	1.0	5.1	3.2	1.21	76.20	15.25	1.8

The results for the comparison between the percent CO₂ from the outside for experiments and simulations are best understood in Figures 2 to 4, where it is possible to see how the standard deviation decrease with simulations using efficiency calculated with Murphree Efficiency.

CONCLUSION

From the results it is possible to see how the values matches better between experiments and simulations using in the software the experimental values of efficiency calculated with Murphree Efficiency. Howe-

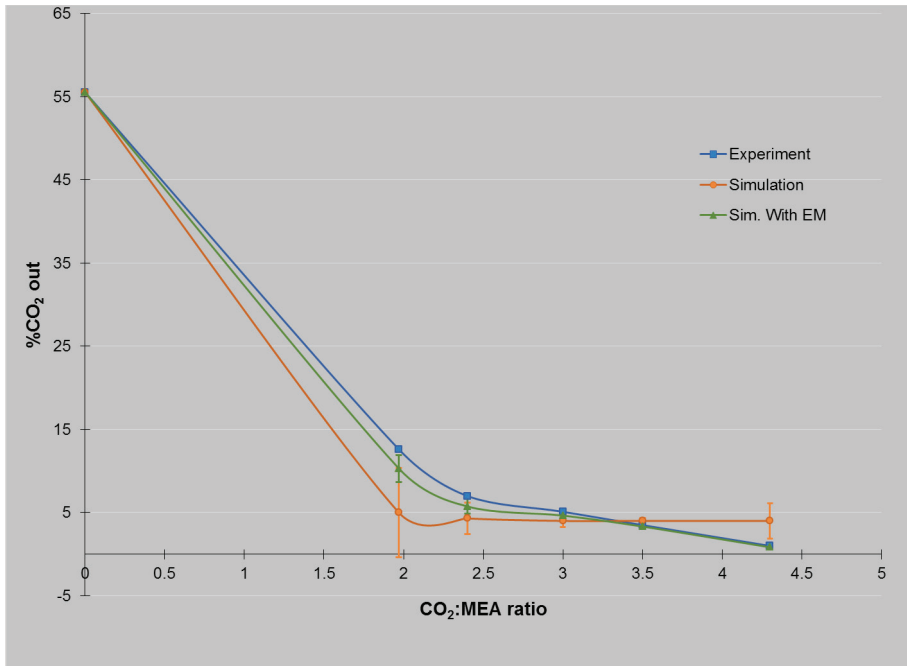


Figure 2. Absorption of CO₂ as function of CO₂:MEA ratio for a gas flow rate of 41.6 NL/min

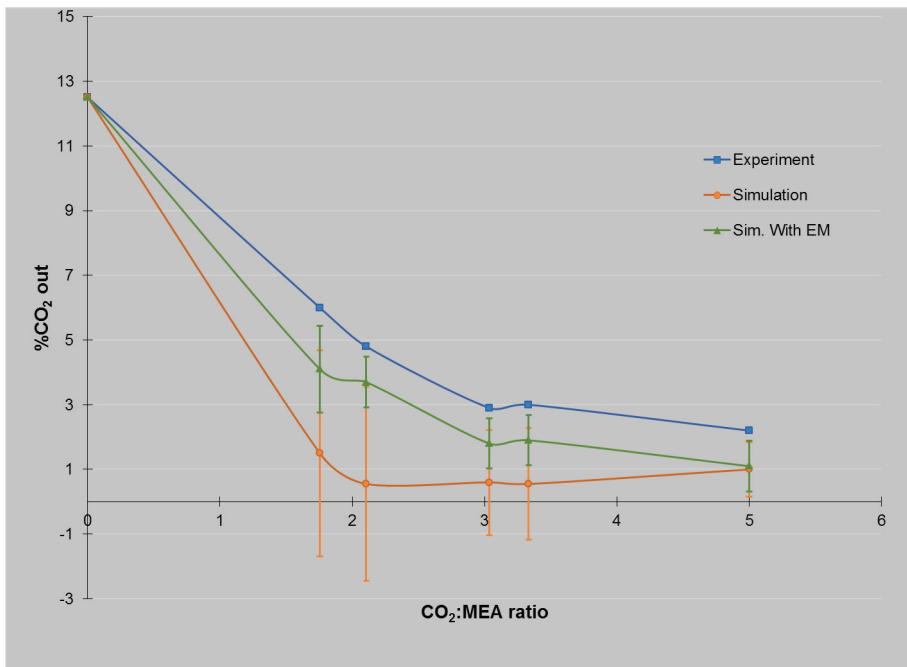


Figure 3. Absorption of CO₂ as function of CO₂:MEA ratio for a gas flow rate of 104 NL/min

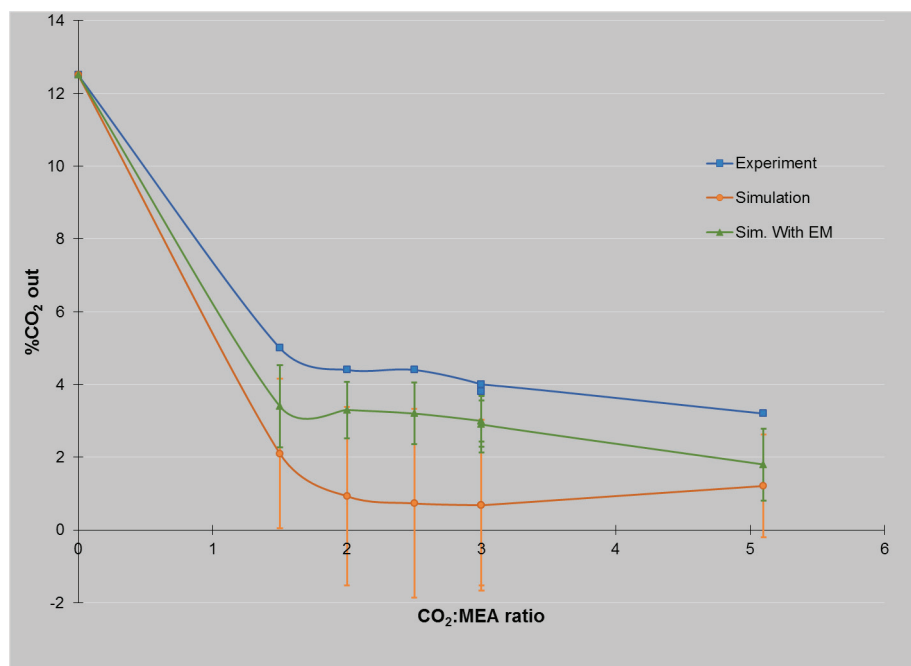


Figure 4. Absorption of CO₂ as function of CO₂:MEA ratio for a gas flow rate of 184 NL/min

ver, there is still a gap between them, but this is only about 1%.

It can be seen that there are better results when the gas flow rate is low (41.6 NL/min) and the concentration of CO₂ is high (55.5%), in these experiments, the percentage of absorption was up to 99% for a CO₂:MEA ratio of 1:4.3 and 88.4% for a ratio of 1:2.

For the case of a gas flow rate of 104 NL/min with CO₂ concentration of 12.5 %, it was possible to have an absorption of 83.5% with a CO₂:MEA ratio of 1:5 and 65.5% for a ratio of 1:2. These values are similar to those obtained using a gas flow rate of 184 NL/min with CO₂ concentration of 12.5%, where the percentage of absorption was 76.2 for a CO₂:MEA ratio of 1:5 and 66.88% for a ratio of 1:2.

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