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Statistical factorial design analysis for parametric interaction and empirical correlations of CO₂ absorption performance in MEA and blended MEA/MDEA processes

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ABSTRACT

A principle of statistical factorial design was applied to characterize behaviour of mass-transfer performance in the carbon dioxide (CO_2) absorption process using monoethanolamine (MEA) and blended MEA/methyldiethanolamine (MDEA) solutions. Over a hundred experiments were carried out using a bench-scale absorber packed with structured packing. The absorption performance was analyzed in terms of overall mass-transfer coefficient (K_Ga_e). The results show that CO_2 loading of the solution is the most influential process parameter on K_Ga_e , followed by alkanolamine concentration, CO_2 partial pressure in feed gas, temperature, and liquid circulating rate, respectively. The temperature interacts with all tested process parameters. A mixing ratio between MEA and MDEA in the blended solution significantly affects K_Ga_e . Second-order correlations for the prediction of K_Ga_e were developed for both MEA and MEA/MDEA systems. A relative packing factor was derived as a function of packing surface area and eventually integrated into the developed mass-transfer correlations for the purpose of scale-up.

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1. Introduction

Capture of carbon dioxide (CO₂) from power plant flue gas streams for greenhouse gas mitigation can be technically implemented by a number of gas separation technologies. Gas absorption into liquid solvent is the most attractive because of its maturity in gas treating services and its compatibility with low CO₂ partial pressure in flue gas. Such capture technology can be integrated to the power plants as a flue gas post-treatment unit after the treatments of particulate matters (PMs) and sulphur dioxide (SO₂). However, a critical issue for this absorption-based ${\rm CO_2}$ capture technology is its significant energy requirement for solvent regeneration and CO₂ stripping, making the process energy-intensive and costly. To reduce the energy requirement, the improvement of design and operation strategy together with the use of energyefficient absorption solvents, such as aqueous solutions of blended monoethanolamine (MEA) and methyldiethanolamine (MDEA), must be applied using the knowledge of mass-transfer performance for CO₂ absorption.

Table 1 summarizes all published studies on mass-transfer performance reported in terms of overall mass-transfer coefficient ($K_G a_e$) in CO₂-alkanolamine systems. These studies have mainly

focused on the individual effect caused by each process parameter on $K_G a_e$ under specific operating conditions. There have been no investigations on the interactions between parameters. No studies have yet demonstrated whether parametric interactions, which occur when a variation in one process parameter results in a change in another parameter's effect on absorption performance, occur in CO_2 –alkanolamine systems, and if they do occur, they have not yet been characterized. Because of the nature of the CO_2 absorption process, in that several process parameters (e.g. temperature, CO_2 partial pressure and CO_2 loading) vary constantly and simultaneously during the operation, knowledge of such parametric interactions is very important for accurate design and cost-effective operation of the absorption process.

This work reveals the relationships between process parameters and CO_2 absorption performance of single MEA and blended MEA/MDEA systems as well as the interactions between such parameters. The parametric effects were investigated by conducting a series of absorption experiments of which the testing matrix was set according to the statistical factorial design principle. The experimental results were correlated into factorial-design based correlations as a function of key parameters that include liquid circulating rate, CO_2 partial pressure in feed gas, alkanolamine concentration, temperature, and CO_2 loading of solution. The developed correlations provide knowledge of the degree of interaction between parameters that causes variation in the parametric effects.

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Table 1 Published studies on overall mass-transfer coefficient $(K_G a_e)$ for CO_2 -alkanolamine systems

Alkanolamine	Test conditions					Reference
	Liquid flow rate (m ³ /(m ² h))	CO ₂ partial pressure (kPa)	Amine concentration (kmol/m³)	Temperature (°C)	CO ₂ loading (mol/mol)	
Monoethanolamine (MEA)	120–180 GPM 1500–6000 lb/(ft ² h) 0.2–7.0 L/s 50–150 10 GPM/ft ² 7.6–22.9 7–16	- 8.1-12.2 Up to 324.2 - 2026.5 Up to 15.3 Up to 10	Up to 3.3 3.0-3.3 0.45-13.5 - 3.0 3.0-5.2 3.0-9.0	Up to 53 43-49 20-37.5 20 24-46 20-37 25	Up to 0.20 0.14-0.18 0.00-0.40 - 0.15-0.43 - 0.20-0.35	[1] [2] [3] [4] [5] [6] [7]
Diethanolamine (DEA)	980-2460 lb/(ft ² h) 0.2-7.0 l/s 50-150 5-20 GPH	4.1–32.4 Up to 324.2 – 10.1–90.2	1.0-4.0 0.45-13.5 - 2.4-3.4	25–55 20–38 20	- 0.00-0.40 - -	[8] [3] [4] [9]
2-Amino-2-methyl-1-propanal (AMP)	4.9–14.6 5.9–14.6	Up to 15.4 Up to 15.2	1.1, 2.0, 3.0 1.1, 2.0, 3.0	24 24	0.15 -	[10] [6]

2. Absorption experiments

2.1. Experimental apparatus

Fig. 1 shows a schematic diagram of the bench-scale gas absorption apparatus by which the CO_2 absorption experiments were conducted. The gas absorption system consists of an absorber, a CO_2 supply, a nitrogen (N_2) supply, two calibrated mass flowmeters, a 20 L solvent reservoir, a 20 L solvent receiver, a variable-speed gear pump, a heated circulating bath, an infrared gas analyzer, and a handheld thermocouple thermometer. The absorber was 20 mm in diameter, packed with DX-type structured packing (Sulzer Brothers Limited, Winterthur, Switzerland), and made of acrylic plastic to allow visual observation of liquid flowing phenomena. Using three removable column sections, the height of the absorber can be varied from 0.165 to 0.825 m. The supplies of CO_2 (99.50%) and N_2

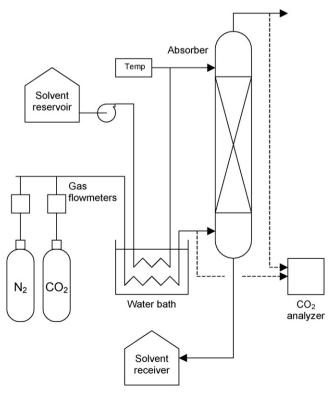


Fig. 1. Schematic diagram of experimental apparatus.

(99.95%) were from gas cylinders purchased from Praxair Canada Inc. The flow rates of CO₂ and N₂ gases entering the absorber were measured by the mass flowmeters (Model GFM 17, Aalborg Instruments & Controls Inc.). Two 20L carboys served as reservoirs for supplying and receiving the aqueous alkanolamine solution used in the experiments. The gear pump (C-75211-10), purchased from the Cole-Parmer Instrument Company, was used to feed the alkanolamine solution to the absorber. The heated circulating bath (Model 7305, Poly Science, Niles, IL), with ±0.05 °C stability, was used for controlling the temperatures of both the CO₂ containing gas stream and alkanolamine solution entering the absorber. The infrared gas analyzer (Model 302WP, Nova Analytical System Inc., Hamilton, Ontario) was used to measure CO₂ concentrations of the gas entering and leaving the absorber. The analyzer can measure CO_2 concentrations of up to 20.0 vol.% with an accuracy of $\pm 2\%$ of full-scale reading. The handheld thermocouple thermometer with ±0.1% accuracy (Model 600-104, Barnant Company, Barrington, IL) was used to measure and ensure the temperature of amine solution entering the absorber.

2.2. Experimental procedure

The alkanolamines used in this work were MEA and MDEA. The MEA and MDEA with purities of $\geq 99\%$ were purchased from Fisher Scientific (Fair Lawn, NJ) and Sigma–Aldrich Ltd. (Oakville, Ontario), respectively. Details on the experimental procedure for CO₂ absorption and sample analysis can be found in our previous works [11,12]. In this work, the CO₂ concentrations of gas entering and leaving the absorber as well as CO₂ content (loading) of amine solution were measured and recorded as the primary experimental data. All experiments were carried out under atmospheric pressure. Gas flow rate was in the range of 0.43–0.60 m³/(m² s). Mass balance of each experimental run was determined and ensured that it was within $\pm 10\%$.

2.3. Mass-transfer coefficient

The absorption performance of a packed column is commonly presented as the overall mass-transfer coefficient ($K_G a_e$), which can be determined by using the following differential equation [11]:

$$K_{\rm G}a_{\rm e} = \frac{G_{\rm I}}{P(y_{{\rm CO}_2,{\rm G}} - y_{{\rm CO}_2}^*)} \left(\frac{{\rm d}Y_{{\rm CO}_2,{\rm G}}}{{\rm d}Z}\right)$$
 (1)

where $G_{\rm I}$ is inert gas velocity in kmol/(m² h); P is total pressure of the system in kPa; Z is column height in m; $y_{\rm CO_2,G}$ and $y_{\rm CO_2}^*$ are

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