

ORIGINAL ARTICLE

Experimental study on the effect of process parameters on postcombustion CO₂ capture using rotating packed bed technology

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Abstract

Environmental issues are the most discussed in today's industrialized world, and this has always been associated with the emission of greenhouse gases. CO₂ emissions, especially from power plants, are considered the major greenhouse gas source. Capturing CO₂ from large emission sources is necessary to address the worst impacts of climate change, including more frequent and severe droughts, heat waves, and intermittent rainfall. Conventional post-combustion CO₂ capture has always been questioned because of its high impact on energy costs. Rotating packed bed technology has shown the potential to reduce the cost of CO₂ capture. This paper presents an experimental study on an in-house designed rotating packed bed absorber for the capture of CO₂ from a flue gas stream. Effects of process parameters such as rotating speed, liquid-to-gas ratio, and monoethanolamine (MEA) concentration on CO₂ capture efficiency and overall volumetric mass transfer coefficient were studied. The study showed that as rotor speed increased, the CO₂ capture efficiency increased, this can be attributed to higher rotor speed, resulting in more droplet flow, indicating an increase in the area of contact between the flue gas and the MEA solvent. Also, as the concentration of MEA increased CO₂ capture efficiency increased, since an increase in concentration results in an increase in the rate of reaction. Finally, the study showed that a rotating packed bed absorber is an efficient unit for CO₂ capture since it enhances mass transfer performance.

KEYWORDS

MEA solvent, postcombustion CO₂ capture, process Intensification, rotating packed bed

1 | INTRODUCTION

The emission of CO₂ especially from power plants which contribute about 41% of the emission to the environment has become an important concern in achieving a net-zero

target by 2050.¹ Global carbon dioxide emissions from combustion and industrial processes grew by 0.9% in 2022, reaching its all-time level of 36.8 Gt, necessitating the need for capture from large point sources. Studies have shown that a typical 500 MWe coal-fired power

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plant released about 8000–10,000 tons of CO₂ per day and the same capacity gas-fired power plant emits about 4000 tons of CO₂ per day.² The high concentration of CO₂ has a devastating effect on environment due to global warming, so eliminating CO₂ is crucial to address the threat to the environment. Decarbonization of the global energy system is necessary to achieve a net zero target by 2050. According to Metz et al.³ implementation of CCS is projected to contribute about 20% towards the overall reduction in CO₂ emission by 2050. Various technologies that have been developed for the capture of CO₂, can be broadly categorized as precombustion, postcombustion, and oxyfuel combustion method.

Postcombustion carbon capture (PCC) is the most mature CO₂ abatement option as demonstrated by commercial deployment projects such as the Petro Nova carbon capture and sequestration plant and the boundary dam carbon capture plant.⁴ Many researchers have looked at different alternatives to improving the performance of conventional PCC to reduce the cost of its commercial deployment such as investigating different solvents, using plant configurations and process intensification^{2,5–15}

Deployment of post-combustion carbon capture at a commercial scale must be cost-effective, since if it is costly, it will lead to increase in energy costs. Approaches to cutting capital and operating costs include process intensification options. Rotating packed bed (RPB) technology is a process intensification option for CO₂ capture, as proposed by Ramshaw¹⁶ and Ramshaw and Mallinson.¹⁷ Indicating that mass transfer is intensified by almost 100% as a result of the rotation of the bed. Burn et al.¹⁸ and Burns and Ramshaw¹⁹ classified the flow in the RPB as pore, film, and droplet flows. Burns and Ramshaw¹⁹ reported that at low rotational speed, there is a severe liquid maldistribution with rivulet flow occupying the packing leaving most of the packing starved of liquid. Zahir et al.²⁰ reported that liquid dispersion in an RPB is highly dependent on solvent-packing contact angle and rotational speed of the packing and at optimum conditions liquid dispersion almost double thereby increasing the absorption of CO₂ by eight times.

The use of RPB for CO₂ capture was reported by many researchers and it found that RPB has the potential to boost mass transfer for the CO₂ capture process.^{2,10–14} Operating costs could easily be reduced by strategies such as intercooling and heat integration, among others. However, they reduce plant flexibility and make operation and control more difficult.²¹ Experimental studies on RPB for CO₂ capture using *N*-methyldiethanolamine enhanced with addition of carbonic anhydrase resulted in 2–5-fold increase in degree of CO₂ absorption

compared to amine without biocatalyst as reported by Wojtasik-Malinowska et al.²² Wen et al.²³ reported the use of controllable cross-sectional areas for intensified CO₂ capture in RPB and conclude that it is viable to adopt packing with constant cross-sectional area.

According to research carried out by Joel et al.,^{12,13} an intensified absorber and stripper are significantly more compact than conventional packed bed (PB) absorber and stripper, with a size reduction of about 12 times and 9.63 times, respectively. Despite the reported advantages of using a RPB for CO₂ absorption, its commercial implementation is not in sight. In this paper, an experimental study on the effect of processing parameters for CO₂ absorption was done to have insight into the best operating conditions that will result in smaller-sized columns with optimum CO₂ absorption rate.

2 | EXPERIMENTAL SECTION

2.1 | Materials

The materials used for this experiment included MEA solution and CO₂–N₂ mixture. An aqueous solution of MEA was prepared at 30, 50, 70, and 90 wt.% with distilled water, and CO₂ and N₂ gas mixtures were prepared for the experimental studies. Concentrated MEA was supplied by a local supplier with a purity of 99%, while the CO₂ and N₂ gases were supplied by African Oxygen (AFROX) Ltd.

2.2 | Experimental procedure

Figure 1 shows the experimental setup of the RPB absorber for the CO₂ capture process with lean-MEA representing solvent that is going into the RPB for the absorption of CO₂ while the rich-MEA representing solvent that has capture CO₂ in it. The packing for the RPB is stainless steel expamet as shown in Figure 2 with the detailed geometric sizes of the packing. Table 1 shows the RPB packing specifications. Flue gas from coal-fired power plants typically contains between 10% and 15% CO₂ as such a simulated one, similar in composition was used as the basis for these experimental studies. In this absorption experiment, flue gas from a coal-fired power plant is represented with a gas mixture containing 14% CO₂ and 86% N₂. The gas mixture is made to travel inward from the packing's outer layer and enters the RPB through the gas inlet. The absorbent, made of different concentrations of 30, 50, 70, and 90 wt%, is simultaneously injected into RPB from the liquid inlet, distributed through the liquid outlet, and flowed out. The mixed gas and the absorbent are in counter-current contact in RPB to remove

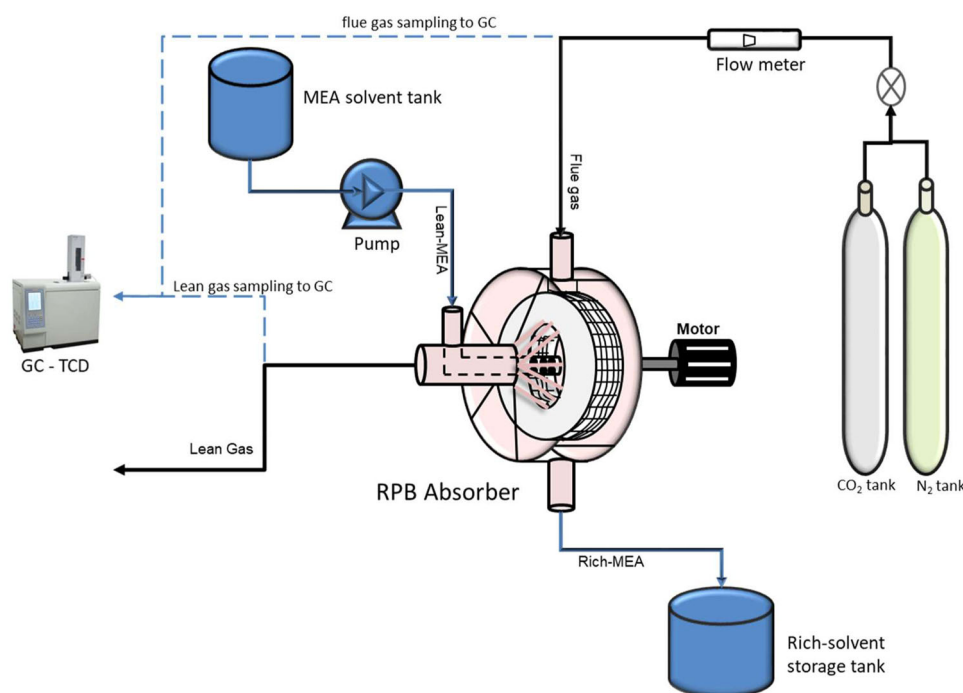


FIGURE 1 Experimental set-up for CO₂ capture.

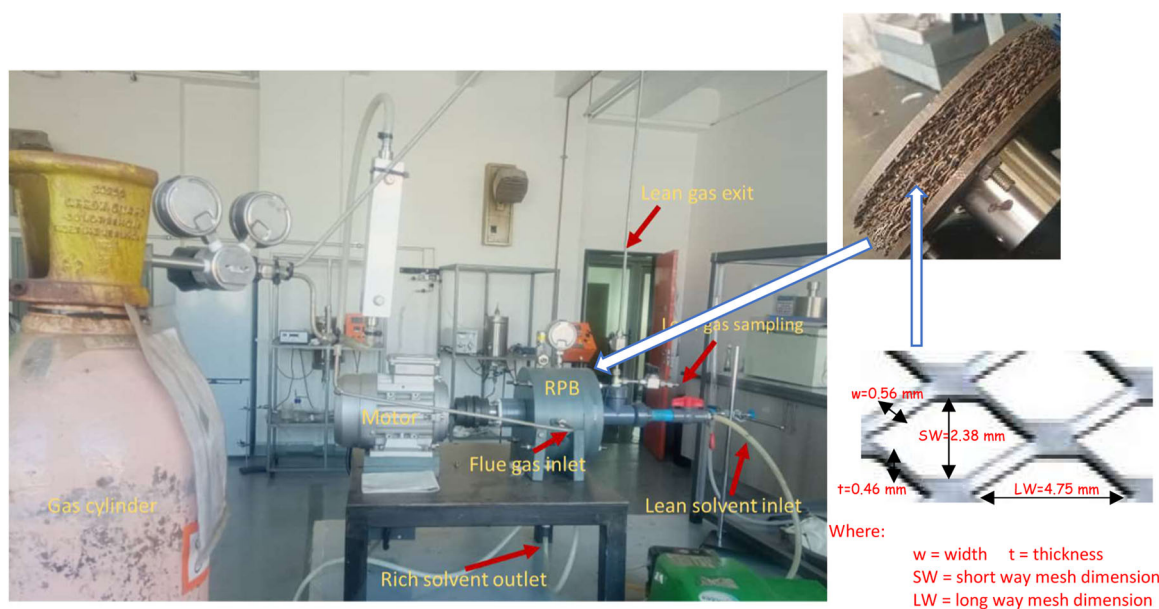


FIGURE 2 Experimental set-up for rotating packed bed absorber.

CO₂. A GC-TCD (SRI 8610C Gas chromatography) is used to determine the CO₂ concentration of the gas before entering the RPB and after exiting the RPB. The key operating conditions of the absorption experiment are listed in Table 2. The surface area per unit volume of the packing is calculated using Equation (1), where the volume is calculated using Equation (2), the total surface area is calculated using Equation (3) and the packing cross-sectional

area is calculated using Equation (4). Voidage of the packing is calculated using Equation (5).

$$\text{Surface area per unit volume} = \frac{\text{Total surface area}}{\text{Volume}}, \quad (1)$$

$$\text{Volume} = \pi \left(\frac{d_o^2}{4} - \frac{d_i^2}{4} \right) Z, \quad (2)$$

TABLE 1 Rotating packed bed specification.

Item	Value
Inner diameter of the packing, cm	2.8
Outer diameter of the packing, cm	13.0
Axial height of the packing, cm	2.2
Specific surface area of the packing, m ² /m ³	1095
Voidage	0.87

TABLE 2 Operating conditions for experimental studies (CO₂ inlet concentration, 14%; temperature, 26°C, pressure, 1 atm).

Parameter	Range
Monoethanolamine solvent concentrations, wt. %	30–90
Liquid to gas ratio, v/v	0.01–0.03
Rotational speed, Hz	0–40

$$\begin{aligned} \text{Total surface area} &= \frac{4(w + t)}{LW \cdot \sin \theta} \\ &\times \text{bed cross sectional area} \\ &\times \text{number of sheet,} \end{aligned} \quad (3)$$

$$\text{Bed cross sectional area} = \pi \left(\frac{d_o^2}{4} - \frac{d_i^2}{4} \right), \quad (4)$$

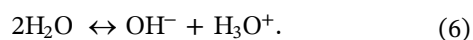
$$\text{Voidage } (\varepsilon) = 1 - \frac{\rho}{\rho_{ss}}. \quad (5)$$

where d_o and d_i = outer and inner diameter of packing, w = width of mesh, Z = axial depth of packing, LW = long way mesh dimension, ρ = packed bed density, ρ_{ss} = stainless steel density.

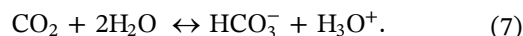
2.3 | Mechanism of CO₂ absorption with MEA

The mechanisms underlying the reactions between carbon dioxide and monoethanolamine have been elucidated using two different approaches: the zwitterion process and the termolecular mechanism.^{24,25} Following Aboudheir et al.,²⁴ the authors propose a series of reactions that can be used to characterize the reaction between carbon dioxide and monoethanolamine in an aqueous solution (Equations 6–15).

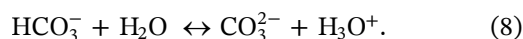
Ionization of water:



Dissociation of dissolved CO₂ to carbonic acid:



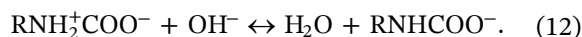
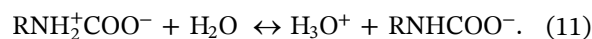
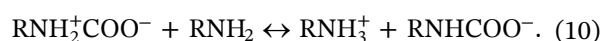
Bicarbonate dissociation as:



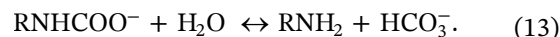
Zwitterion formation from the reaction of MEA with CO₂:



Deprotonation of the zwitterion and carbamate formation:



Carbamate reversion to bicarbonate (hydrolysis reaction):



Dissociation of protonated MEA:



Bicarbonate formation:



According to Yu et al.,²⁶ molecular diffusion, physical dissolution, and chemical reaction are the three processes that predominate in the pseudo-first-order reaction of CO₂ absorption by MEA. Aroonwilas et al.²⁷ observed that the chemical reaction between CO₂ and MEA occurs very rapidly, and once the CO₂ is physically absorbed into the liquid phase, it begins to transform into other components. The penetration of CO₂ from the gas phase to the gas-liquid interphase can be considered to be the principal obstacle in the mass transfer mechanism.

2.4 | Theoretical method of analysis

Mass transfer performance in RPB is usually evaluated by assessing the overall volumetric mass transfer coefficient ($K_G a$). The $K_G a$ is of great importance as it serves as a

guiding parameter for the design of RPBs. Equation (16) is used to calculate the $K_G a$.²⁸

$$K_G a = \frac{G}{\pi P Z (r_{\text{out}}^2 - r_{\text{in}}^2)} \int \frac{1}{(y_{\text{CO}_2} - y_{\text{CO}_2}^*)} d \left(\frac{y_{\text{CO}_2}}{1 - y_{\text{CO}_2}} \right) \quad (16)$$

where P = total pressure, Z = axial height of packing, r_{in} and r_{out} = inner and outer radii, G = gas flowrate, y_{CO_2} and $y_{\text{CO}_2}^*$ = mole fraction and equilibrium mole fraction of CO_2 in the gas phase.

Since the reaction between CO_2 and MEA is fast, the equilibrium mole fraction ($y_{\text{CO}_2}^*$) can be assumed to be zero. Therefore, $K_G a$ (Equation 17) can be obtained by integrating Equation (16).

$$K_G a = \frac{G}{\pi P Z (r_{\text{out}}^2 - r_{\text{in}}^2)} \times \left[\ln \frac{y_{\text{in}}(1 - y_{\text{out}})}{y_{\text{out}}(1 - y_{\text{in}})} + \left(\frac{1}{1 - y_{\text{in}}} - \frac{1}{1 - y_{\text{out}}} \right) \right] \quad (17)$$

where y_{in} and y_{out} are the inlet and outlet mole fractions of CO_2 in the gas phase respectively.

Again, CO_2 capture efficiency is an important quantity that determines the performance of the RPB absorber, and this can be evaluated using Equation (18).

$$\text{efficiency } (\eta) = \left[1 - \frac{y_{\text{out}}(1 - y_{\text{in}})}{y_{\text{in}}(1 - y_{\text{out}})} \right] \times 100. \quad (18)$$

3 | RESULTS AND DISCUSSION

3.1 | Effect of rotational speed on CO_2 capture efficiency and $K_G a$

The RPB's energy needs are influenced by the rotor speed, which also influences the level of capture. To maximize the energy needed to maintain the speed with respect to the capture level, it is crucial to establish the relationship between rotational speed and capture levels.

To understand the relationship between the rotational speed, capture level and K_G , the rotor speed was varied from 0 Hz to 40 rpm. The range was chosen for this experimental study considering the motor's maximum rotor frequency of 50 and 0 Hz was chosen as the lowest rotational speed so as to understand the effect of rotation

when the bed is stationary (no rotation) as compared to when the rotation takes effect. 30 wt% MEA concentration was chosen for this study and an inlet (synthesized flue) gas CO_2 concentration of 14% was used.

Figure 3 shows the effects of varying rotor speeds on CO_2 capture levels for 30 wt% lean MEA concentrations operating at a temperature of 26°C and a pressure of $101,325 \text{ N/m}^2$. The results show that there is a positive correlation between rotor speed and the degree of CO_2 capture. According to the results of Burns et al.,^{18,29} the rotation of the absorber has been shown to enhance mass transfer by promoting a combination of droplet and film flow. Also, at higher rotational speeds thickness of the liquid film decreases, resulting in improved molecular transfer from the gas phase to the liquid phase according to Burns et al.¹⁹ Moreover, when the rotor speed is increased, the liquid maldistribution problem is effectively solved, resulting in a larger wetted area. Consequently, this increase in wetted area contributes to the improvement of mass transfer. To understand the contribution of the rotation on the capture efficiency, the experiment was done with no rotation of the bed while the solvent was passed through the packing using a liquid distributor. Figure 3 shows that at 0 Hz motor frequency the capture efficiency was 7.67% but when the frequency was increased to 10 Hz the capture efficiency increased to 34.62% indicating that the rotation of the bed favored an increase in the absorption CO_2 . Also, it was observed that as the rotational speed increased the overall volumetric mass transfer coefficient increased which agrees with findings by Joel et al.¹² and Liu et al.³⁰ that an increase in absorption results in an increase in the overall mass transfer coefficient due to increase in mass transfer driving force. It was observed that there is 14.45 times increase in overall

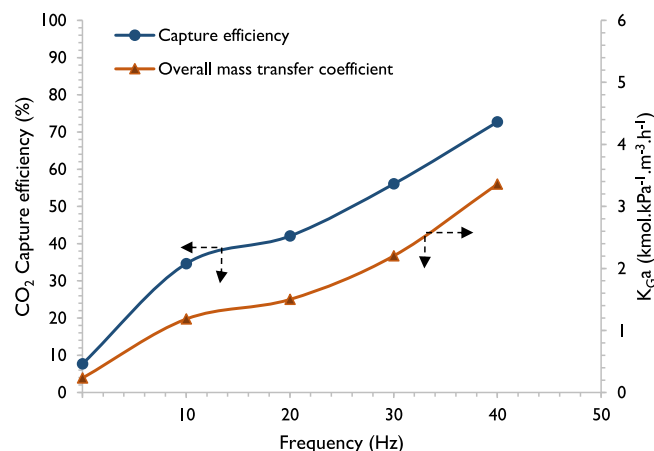


FIGURE 3 Effect of rotational speed on the CO_2 capture efficiency and overall volumetric mass transfer coefficient. Reactions conditions: $L = 0.10 \text{ L/min}$, $G = 9.5 \text{ L/min}$, $y_{\text{CO}_2, \text{in}} = 14\%$, $C_{\text{MEA}} = 30 \text{ wt\%}$, $P = 101,325 \text{ N/m}^2$, $T = 26^\circ\text{C}$.

volumetric mass transfer coefficient at 40 Hz motor speed compared to when there is no rotation of the motor. This study confirms the findings of Zahir et al.²⁰ that the contact angle between solvent and packing and the rotation speed of the packing have a significant influence on the liquid dispersion in the packing; for example, an eightfold increase in CO₂ absorption was observed when the liquid dispersion was doubled. In addition, the shear forces cause greater micro-mixing with increasing speed, which leads to a decrease in the effective boundary layer thickness and an increase in the mass transfer coefficient on the liquid side.²²

3.2 | Effect of MEA concentration on CO₂ capture efficiency and $K_G a$

Higher capture rates and a greater propensity for equipment corrosion are caused by an increase in the concentration of lean MEA. To find the concentration that provides an optimum capture efficiency and with lower tendency for corrosion, it is important to have a thorough understanding of this connection. Also, increasing the MEA concentration leads to an increase in the viscosity of the solvent and this will invariably increase the thickness of the film thereby decreasing the mass transfer driving. Lean MEA solvent flow rate of 0.1 L/min, at 26°C and a motor speed of 20 Hz were chosen. Lean MEA concentrations of 30, 50, 70, and 90 wt% were used in this investigation.

Figure 4 illustrates the effect of different MEA concentrations on the level of CO₂ capture corresponding to the input conditions shown in Table 2. The capture efficiency shows an upward trend as the concentration of

MEA increases. This behavior is due to a higher level of CO₂ absorption brought on by an increase in hydroxide ions per unit volume. This result is consistent with the findings of Freguia and Rochelle,³¹ who showed that the rate of reaction of a first-order pseudo reaction depends on the concentration of MEA. Higher concentration of MEA leads to a higher reaction rate. Furthermore, it can be deduced from the results of Figure 4 that an increase in the concentration of MEA leads to a corresponding increase in the overall volumetric mass transfer coefficient. This finding that a higher MEA concentration leads to higher capture is consistent with the reports of Jassim et al.³² who found that at a higher MEA concentration, CO₂ penetration is lower due to improved absorption kinetics.

3.3 | Effect of liquid-to-gas ratio on CO₂ capture efficiency and $K_G a$

Flue gas flow rate is an important consideration in calculating the size of the absorption column when designing RPB absorbers, and this experimental study is vital in meeting the CO₂ emission removal target. For this study, input conditions in Table 2 having a constant rotor speed of 20 Hz were used. 50 wt% MEA concentration was selected, and the flue gas flow rate was kept constant at 9.5 L/min while the lean MEA flow rate was varied from 0.1 to 0.32 L/min.

Figure 5 depicts how the liquid-to-gas ratio affected the effectiveness of CO₂ capture. It was shown that the CO₂ capture efficiency improves along with the liquid-to-gas ratio, which is related to an increase in the amount of

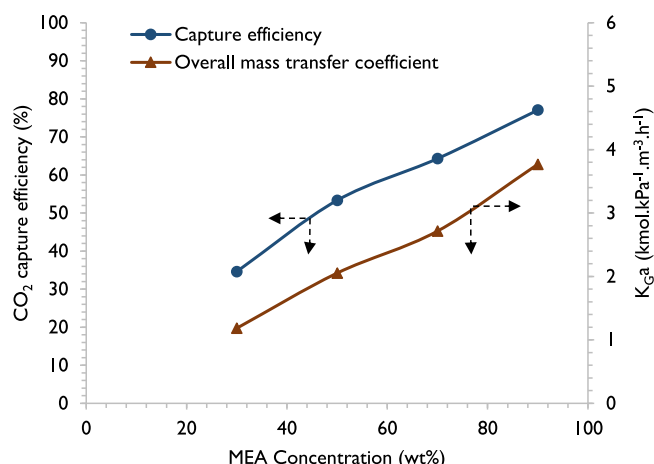


FIGURE 4 Effect of monoethanolamine concentration on CO₂ capture efficiency and overall volumetric mass transfer coefficient. Reactions conditions: $L = 0.10$ L/min, $G = 9.5$ L/min, $y_{\text{CO}_2, \text{in}} = 14\%$, $P = 101,325$ N/m², $T = 26^\circ\text{C}$ motor speed = 20 Hz.

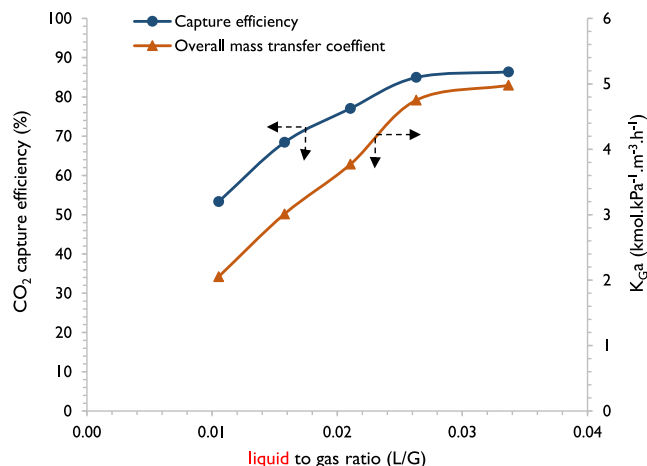


FIGURE 5 Effect of liquid to gas ratio on CO₂ capture efficiency and overall volumetric mass transfer coefficient. Reactions conditions: $L/G = 0.01$ – 0.03 , $y_{\text{CO}_2, \text{in}} = 14\%$, $P = 101,325$ N/m², $T = 26^\circ\text{C}$ motor speed = 20 Hz.

solvent that reacts with CO₂ in the flue gas. The amount of Hydroxide ions that can react with CO₂ will rise as the amount of lean solvent increases. Figure 5 shows that once the liquid-to-gas ratio reaches 0.027 l/l, subsequent increases in the ratio have little to no effect on the amount of CO₂ that can be captured. This might be because the solvent was adequate to capture the most CO₂ available or because the bed was flooded due to the high solvent flow rate in the bed. The effect of liquid-to-gas ratio on the overall volumetric mass transfer coefficient was also observed in Figure 5, showing an increase in the overall mass transfer coefficient as the liquid-to-gas ratio increases. This is consistent with the findings of Otitoju et al.³³ and Zhan et al.³⁴ that increasing the flow rate of lean solvent leads to an increase in CO₂ capture efficiency. It also agrees with Joel et al.¹² that increasing the flue gas flow rate leads to a decrease in capture efficiency.

4 | CONCLUSION

In this study, an experimental investigation of RPB absorber was conducted to study the effects of various process variables on the absorption of CO₂ from flue gas streams. The effect of rotation speed on CO₂ capture efficiency and overall volumetric mass transfer coefficient was investigated by varying the rotation speed from 0 to 40 Hz. The study showed a 9.5-fold improvement in CO₂ capture efficiency at 40 Hz compared to no rotation, and the overall volumetric mass transfer coefficient also improved by 14.5 times. The study on the effect of MEA concentration on CO₂ capture efficiency and overall volumetric mass transfer coefficient was conducted at a solvent flow rate of 0.1 L/min and a speed of 20 Hz, varying the concentration from 30 to 90 wt%. It was found that the capture efficiency and the overall volumetric mass transfer coefficient increase with increasing concentration MEA, which can be attributed to an increase in the rate of reaction as concentration increases. The effect of liquid to gas ratio was also studied and it was found that as the L/G ratio increases the CO₂ capture efficiency and overall mass transfer coefficient also increase. Finally, the study showed that an RPB absorber has the potential for use in the capture of CO₂ from flue gas and its process parameters studied will help in CO₂ capture plant design and operation.

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