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Efficient Carbon Dioxide Capture in Packed Columns by Solvents Blend Promoted by Chemical Additives

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ABSTRACT

This work assessed the carbon dioxide capture performance by different aqueous amines, in comparison to standard amine monoethanolamine (MEA), the standard for the capture process, A continuous operation was implemented, and a packed column served as the scrubber to develop effective sorbents for carbon capture and separation (CCS) procedures. The impact of operating variables such as carbon dioxide loading, feed amine temperature, solvent amine weight concentration, simulated gas flow rate, carbon dioxide feed concentration, amine type, and liquid flow rate was examined. Carbon dioxide concentration; absorption efficiency and overall mass transfer coefficient measurement as an indicator for the effectiveness of the capture process. For each system, the overall amine concentration was maintained at 30 weight percent and the treating a gas mixture containing 15% CO_2 (by volume) mimicking a fossil-derived flue gas. according to the findings of the experimental result show the raising of the operating temperature had no appreciable impact on the effectiveness of CO_2 removal in either combination. On the other hand, the efficiency of CO_2 removal rises with rising amine concentration, volume percentage of amine in the solvent, and liquid flow rate; on the other hand, the efficiency of CO_2 removal falls sharply with increasing gas flow rate. The maximal CO_2 removal coefficients achieved by MEA + DEA and MEA + TEAM aqueous solutions under ideal circumstances are 97.9% and 91.9%, respectively. Additionally, given the experiment's operating conditions, the highest volumetric total gas-phase mass transfer coefficient was found to be 110 and 67.5 (kmol/m^3 h kpa), respectively.

Keywords: chemical absorption, carbon dioxide capture, packed bed column, monoethanolamine, diethylenetriamine.

INTRODUCTION

The world's rapid population growth and significant industrial advancement have led to a notable increase in energy consumption; furthermore, conventional sources of energy, such as petroleum, are accountable for the damage brought to animals, air and water pollution, and other issues when used (AL-aridhee and Moghiman, 2023). The emission of greenhouse gases that contributes most to worldwide warming is carbon dioxide (CO_2) (Procopio et al., 2023; Majeed and Majeed, 2023). Greenhouse gases are the cause of the greenhouse effect phenomenon (Cassia et al., 2018). Greenhouse effect terms refer to the Earth's surface warming as a result of solar energy traveling through the atmosphere mainly unaltered (Hason et al., 2020); the energy is then released again as infrared, and most of it is captured by the water vapor and carbon dioxide in the Earth's atmosphere, surrounding the planet like a blanket (Anderson et al., 2016). As a result of our continued use of fossil fuels, the amount of carbon dioxide in the atmosphere is rising annually, intensifying the planet's greenhouse effect (Waisi et al., 2021). Thus, lowering the amount of carbon dioxide in the atmosphere could be helpful in limiting worldwide warming and its impacts on changing the climate (AL-Mashhadani and Khudhair, 2017; Jaffary et al., 2021). Utilizing carbon capture and storage (CCS) is one way of lowering the amount of carbon dioxide produced by burning fossil fuels (Navarrete Procopio et al., 2023). CCS entails capturing concentrated carbon dioxide streams from specific sources (factories exhaust emissions), followed by safely storing them in a suitable geological reservoir. Drained oil or gas reservoirs, inaccessible coal

seams, and salty aquifers are examples of reservoir geological structures (Bhavsar et al., 2023; Chamgoué et al., 2023). However, there is an urgent need for advancements in the use of carbon dioxide capture, utilization, and storage (CCUS) technology to render it more cost-effective and efficient 1. Chemical looping, 2. Pre-combustion, 3. Oxy-fuel combustion, and 4. Post-combustion methods for carbon utilization have lately gained global attention and advancement and have started to be extensively utilized (Yan and Zhang, 2019). For the purpose of capturing carbon dioxide, a variety of methods are available, including membrane, cryogenic, gas separation, adsorption, and absorption (Majeed and Majeed, 2017). Post-combustion capture (PCC) with the use of the chemical absorption approach, particularly amine solution scrubbing, has been shown as the most advanced and viable technology for reducing carbon dioxide emissions from major fossil-fuel-driven plants (Ling et al., 2020). Because the gaseous carbon dioxide reacts with amine solutions at a high and rapid rate, these methods of separation could selectively remove CO_2 from gas mixtures using absorption scrubbers, which usually include packed bed absorbers (Dhuyool and Shakir, 2023). The current industrial carbon capture units use monoethanolamine (MEA) as a chemical absorbent, which is recognized as the best absorption efficiency (Liu, 2021). Researchers have investigated the blending of various amines to improve the performance of the conventional solvent (MEA) (Lee et al., 2020); Because polyamines include numerous amino groups in a single molecule with rapid reaction kinetics and loadings in terms of CO_2 capturing, thus they have drawn much of interest in CCS processes (Barzagli et al., 2022). MEA has a poor CO_2 loading capacity since its structure only contains one amine group. Three groups of amines in EDTA allow them to load CO_2 more efficiently. Since I-BA and MEA each have one amine group, their loading capacities are equal. Greater loading capacity increases the effectiveness of CO_2 removal from flue gas. DETA: Less biodegradable than MEA, leading to prolonged environmental persistence. More toxic than MEA, posing significant risks to aquatic life if released untreated. Requires careful management during disposal due to hazardous degradation products (Gjernes et al., 2013), I-BA: Moderately biodegradable, with degradation products generally less hazardous than MEA and DETA. Displays moderate toxicity, necessitating attention to environmental exposure levels. Proper disposal methods are required, which may be less intricate compared to MEA and DETA (Nematollahi and Carvalho, 2019). In this work primarily attempts a comparative experiment to compare the efficiency of carbon dioxide absorption through the use of various types of amine solutions using the same experimental setup with various mass blending ratios; the performance is investigated in terms of the total coefficient of mass transfer ($K_G a_v$); sweet carbon dioxide concentration and carbon dioxide capture efficiency (η).

Carbon dioxide absorption efficiency determination

It is represented as the amount of carbon dioxide absorbed by an amine solution under typical usage conditions. The equation for this is as follows (Aroonwilas and Veawab, 2004):

$$\eta = \left(1 - \frac{y_{CO2,out}}{1-y_{CO2,out}} * \frac{1-y_{CO2,in}}{y_{CO2,in}}\right) * 100\ \% \quad (1)$$

where: $\gamma_{CO2.in}$ mole fractions of carbon dioxide gas phase feeding and $\gamma_{CO2,out}$ mole fractions of carbon dioxide gas phase released.

Overall coefficient of mass transfer based on the gas phase ($K_{G\times av}$) determination

The mass transfer coefficient is primarily influenced by three factors: gas phase resistance, liquid phase resistance, and the wetting area between gases and liquids (Nair et al.,2014). Equation 2 is used to compute the $K_{G\times av}$ (Aroonwilas and Tontiwachwuthikul, 1998; Naami et al., 2012; Sheng et al., 2016; Gao et al., 2017):

$$K_{G*a_v} = \frac{G}{P*Z} * \left[\begin{array}{l} ln\left(\frac{y_{CO2,in}*(1-y_{CO2,out})}{y_{CO2,out}*(1-y_{CO2,in})}\right) + \\ + \left(\frac{y_{CO2,in}}{(1-y_{CO2,in})} - \frac{y_{CO2,out}}{(1-y_{CO2,out})}\right) \end{array}\right] \quad (2)$$

where: Z – the packed height of the absorber, G – represents the total inert gas molar flow, and P is the operational pressure.

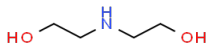
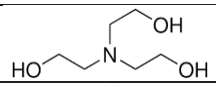
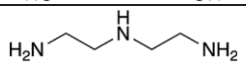
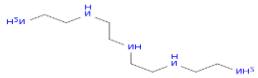
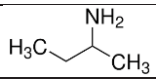
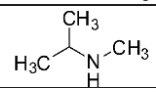
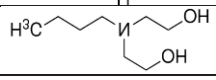
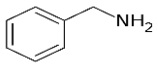
EXPERIMENTAL MATERIALS AND CHEMICALS

The list of compounds used in this experiment and further details on their molecular weight, company name, physical state, and purity are provided below as shown in Table 1. The

Table 1. Experimental materials and chemicals

No	Materials	Chemical symbol	Supplied company	Origin	M.wt g/mol	Purity/State
1	Monoethanolamine	C_2H_7NO	Thomas Baker	India	61.08	97% Liquid
2	Diethylenetriamine	$C_4H_{13}N_3$	Fluka	Switzerland	103.1	99% Liquid
3	N-Butyl diethanolamine	$C_8H_{19}NO_2$			161.2	97% liquid
4	Tetraethylenepentamine	$C_8H_{23}N_5$			189.3	97% liquid
5	Sec-butylamine	$C_4H_{11}N$			73.14	99% liquid
6	Iso-butylamine	$C_4H_{11}N$			73.14	99% liquid
7	Benzylamine	C_7H_9N			107.1	96% liquid
8	Hydrochloric acid	HCl	CDH	India	36.46	35–38% Liquid
9	Carbon dioxide	CO_2	AL-Nahrawan gas company	Iraq	44.01	99.6% purity gas
10	Nitrogen	N_2			28.02	
11	Distilled water	H_2O	Dep Lab.	Iraq	18.015	TDS = 5 ppm PH = 7.1 Cond.= 8µs/cm

Table 2. Experimental amine properties

Substances	Symbol	Structure	Number of an amine group	Loading capacity	Carbone atom	Absorption rate
Monoethanolamine	MEA		1	Medium	2	high
Diethanolamine	DEA		1	Medium	4	medium
Triethanolamine	TEA		1	Medium	6	low
Diethylenetriamine	DETA		3	High	4	high
Tetraethylenpentamine	TEPA		5	Very high	8	low
Sec-butylamine	S-BA		1	Medium	3	medium
Iso-butylamine	I-BA		1	Medium	3	medium
N-butyl-diethanolamine	N-BDEA		1	Low	7	low
Benzylamine	BA		1	Medium	7	Very low
Piperazine	PZ		2	High	4	high

amino solvent is chosen based on the difference in molecule structure, carbon atom number, number of hydroxyl groups, and number of amine groups All these factors affect the solubility in water, diffusion, mass transfer rate, and loading capacity of CO_2. As shown in the Table 2.

EXPERIMENTAL ABSORPTION UNIT SETUP

The absorption unit as shown in Figure 1 was employed in this investigation. The major part of this unit consists mainly of the packed column,

which measures 1.8 m in height and 40 mm in diameter which is divided into five packed stages by a side stream drawn from each stage; all design details of the absorber column are listed in Table 3. The packed column is filled by the inert ceramic packing material structured a randomly packed structure which is a ceramic Intalox saddle type, while the column is made of acrylic; all the design data of Intalox saddle packing are listed in Table 5.

The operation mode was carried out in continuous mode, in which the gas phase was constantly fed into the packed column from its base and followed up while the liquid phase was fed from the top and followed dawn by gravity to its base, this operation mode satisfying the counter-current flow, in which enhancing the chemical absorption of carbon dioxide by perfectly mixing between the gas and liquid phase. the amine solution was heated on the hot plate magnetic stirrer (Dragon Lab, Australia) to the desired operating amine temperature before it was transferred immediately into the packed reactor by diaphragm pump (TYP-2500NH, China); for each test, a 500 ml amine solution is utilized. The feed simulated gas mixture (CO_2 and N_2 gas) was prepared with the aid of using two different gas flow meters (Bass Instrument, Turkey) with a range from 0 to 10 L/min. and an inline CO_2 gas analyzer (BIOGAS 5000, UK,) was used to monitor the carbon dioxide content of the effluent gas.

The simulated gas mixture was initially introduced into the CO_2 analyzer to check its concentration at the beginning of each experiment. the gas mixture flows through a bypass line (by opening HV-008 and closing HV-107) on the absorber to the V-003 when the CO_2 analyzer measures the CO_2 concentration. The CO_2 weight percent is manipulated by changing the flow rate of CO_2 and N_2 through the flow valves FV-104 and FV-005 until the CO_2 inlet concentration is fixed. The operation process of absorption proceeds until no further change in absorption rate (steady state operation), this is confirmed by the reading of carbon dioxide concentration indicated by the gas analyzer. Control experiments are essential in carbon capture studies to establish a baseline for comparison: A standard MEA, is used as a control to establish a baseline performance for CO_2 absorption. regular checks and calibrations of the experimental setup, including gas flow meters, liquid flow meters, and gas analyzers are performed to ensure accurate measurements; and control runs using a non-reactive solvent (e.g., water) may be conducted to detect any systemic errors or leaks in the setup. By incorporating control experiments for more accurate results about the efficacy of amine solvents

Table 3. Design characteristics of the packed bed column

Parameters	Data
Walls material	Acrylic
Column outer diameter (m)	0.05
Column inner diameter (m)	0.04
Column length (m)	1.85
Packing length (m)	1.6
Top stage height (cm)	20
Second stage height (cm)	30
Third stage height (cm)	30
Fourth stage height (cm)	30
Bottom stage height (cm)	50
Column packing	Intalox saddle
Intalox saddle material	Ceramic
Intalox saddle size (mm)	13
Contact surface (m^2/m^3)	480
Bulk density (kg/m^3)	737
Packing factor (m^2/m^3)	660

Table 4. Absorber packing design data (Coulson, J. M., 2001)

Parameters	Value
Column packing type	Intalox saddles
Material	Ceramic
Size	13 mm
Bulk density	737 kg/m^3
surface area	480 m^2/m^3
Packing factor (F_b)	660 m^2/m^3

Table 5. Flag symbol of the absorption unit experimental setup

No	Tag plate	Description
1	V-001	Nitrogen gas cylinder
2	V-002	Carbon dioxide gas cylinder
3	V-003	Outlet gas knock-out drum
4	TK-001	Len amine feed tank
5	TK-002	Washing water tank
6	TK-003	Rich amine tank
7	P-002	Liquid feed pump
8	T-001	Packed bed absorption tower
9	AT-001	Carbon dioxide analyzer

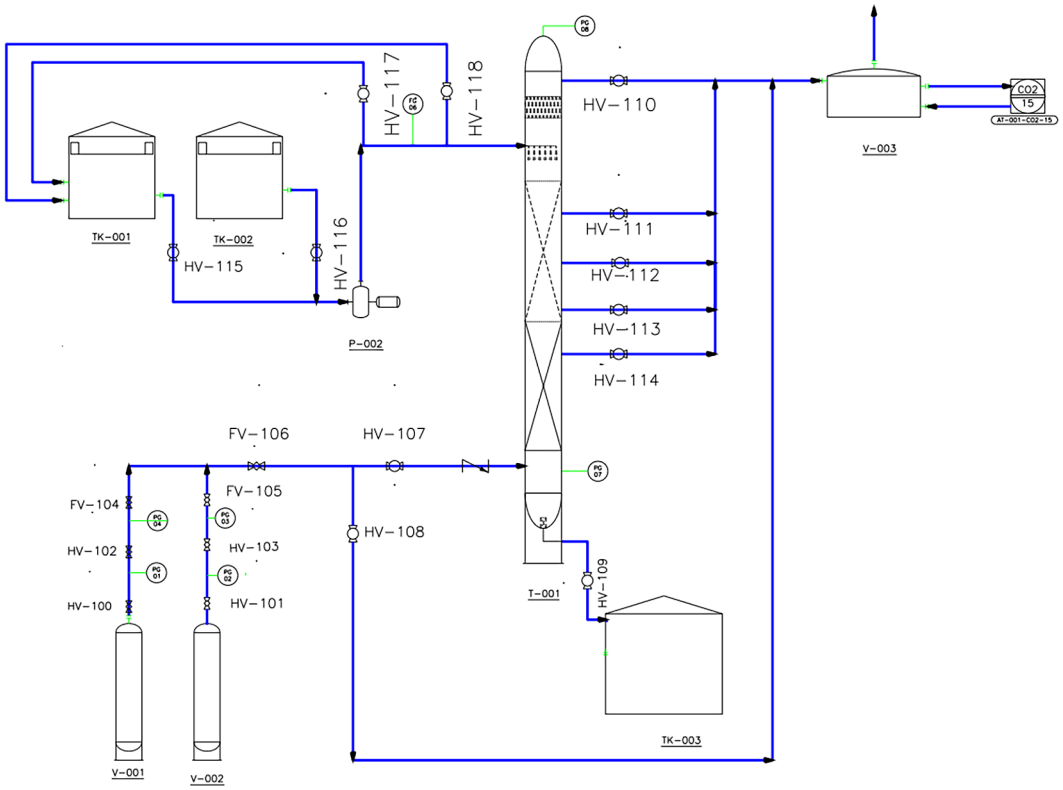


Figure 1. Absorption unit experimental setup

for carbon capture. The experimental conditions was carried out at ambient temperature (25–30 °C) and atmospheric pressure, but an experiment was carried out at different temperatures of amine solution to investigate their impact.

RESULTS AND DISCUSSION

Effect of liquids to gas ratio

The performance test of the absorption of CO_2 from a gas mixture by using MEA as a solvent is done by choosing different liquid-to-gas ratios at operating conditions of 15% inlet CO_2 concentration and 20% MEA solution. The CO_2 profile, removal efficiency, and volumetric mass transfer coefficient along column height are shown in Figures 2, 3 and 4 respectively.

Figure 2 shows that the CO_2 concentration decreases along the column height with different liquid-to-gas ratios. By increasing the liquid-to-gas ratio, the outlet CO_2 mol% decreases at the same height that the CO_2 concentration measures. As a result, CO_2 mol% decreased to 8% at 10 L/G while declined to 11.5% at 2 L/G at 1.6 m of height.

Figure 2 explains the removal efficiency against column height at different liquids to gas ratios. As the contrast CO_2 concentration, removal efficiency increases when liquid-to-gas ratios increase. Figure 3 also shows that 10 L/G gives a higher removal efficiency which is around 50% while a lower removal efficiency is obtained at 2.5 L/G. this is an expected result since removal efficiency is strongly dependent on CO_2 concentration. In conclusion, CO_2 concentration decreases as the liquid-to-gas ratio increases because the absorption performance capacity increases as the liquid flow rate increases. Additionally, when the flow velocity of aqueous amine increases the surface area of the interface per unit volume rises as well, improving absorption capacity. This behavior resulted from amine molecules being more readily available to react with the carbon dioxide molecules at a greater ratio, which increased absorption efficiency (Rajiman et al., 2020), the same finding was also reported by Qing et al., (2011).

Figure 4 indicates the volumetric mass transfer coefficient estimation at various liquid-to-gas ratios along column height. From this figure, the maximum volumetric mass transfer coefficient was obtained at 3.3 L/G which gives a balance

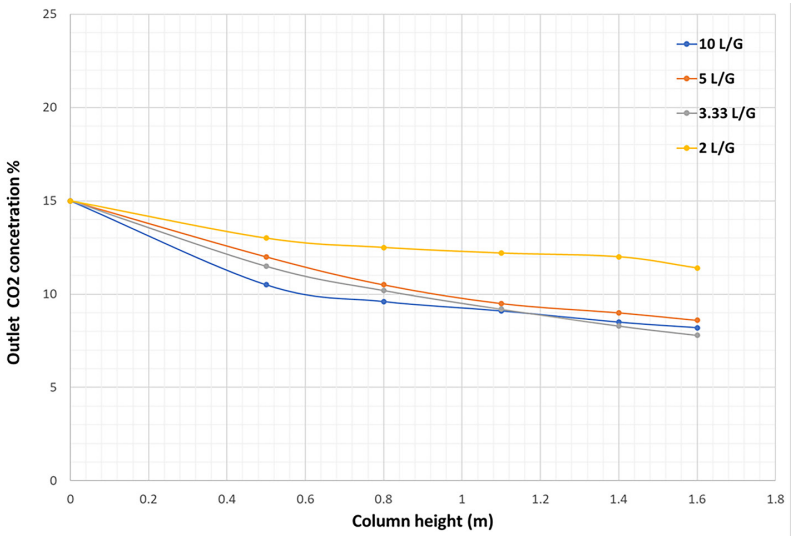


Figure 2. CO_2 concentration at different packing heights

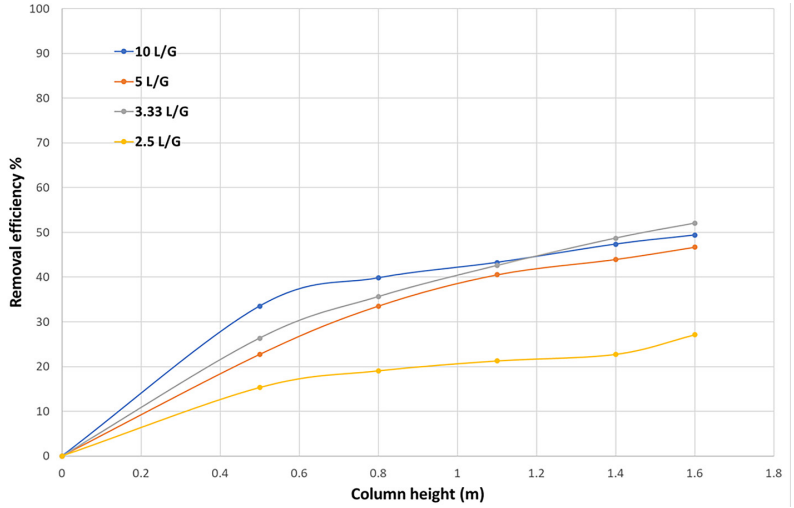


Figure 3. Removal efficiency at the different liquid-to-gas ratios

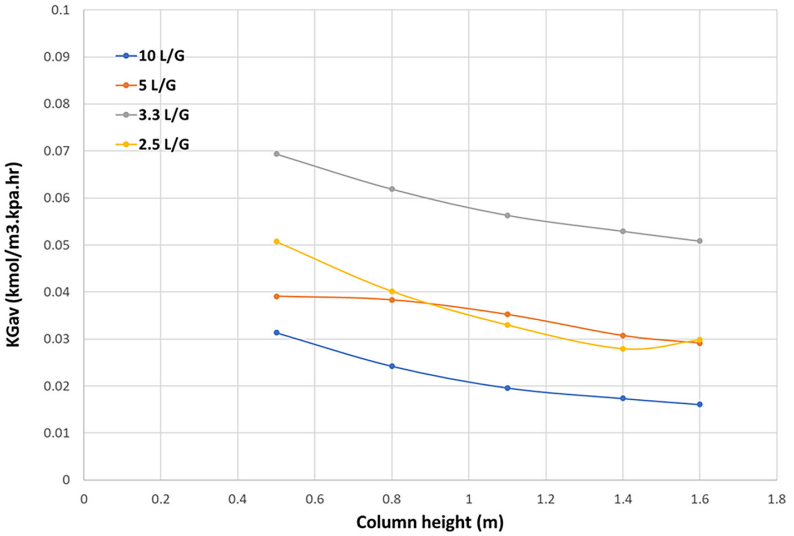


Figure 4. volumetric mass transfer coefficient at various liquid to gas ratios

between hydrodynamic and thermodynamic behavior of chemical absorption. Since the liquid phase boundaries decrease as the droplet circulation rate rises. Consequently, the mass transferring efficiency is improved and the barrier to gas diffusion into the liquid phase is reduced. Due to the minimal increase in effective interfacial area and the negligible droplet size reduction caused by rising liquid flow rates, the growing tendency quickly decreased at higher liquid flow rates. As a result, increasing the liquid flow rate will not improve the mass transfer performance further (Wu et al., 2017). On the other hand, the overall mass transfer coefficient, and carbon dioxide removal efficiency, for all amines dropped as the gas flow rate rose. This is because an increase in the gas flow rate has the potential to Increase the rate at which carbon dioxide renews itself, which raises the bulk partial pressure at the gas/liquid interface (Dhuyool et al., 2024).

Effect of carbon dioxide inlet concentration

The effect of CO_2 inlet concentration has been studied in this section to show the performance of the absorption process at different CO_2 feed mole fractions,10%, 15%, and 20%. The CO_2 concentration in the gas inlet stream appears to have an impact on the removal efficiency, and KGav value. Figure 5, 6, 7 illustrates these values decrease as the CO_2 inlet concentration rises. This pattern is to be caused by the limited diffusion of solvent molecules in the liquid phase. This limited diffusion in the liquid phase leads to a steady absorption of CO_2. Thus, more difficult absorption performance is caused by increased CO_2 feed concentration. While raising the CO_2

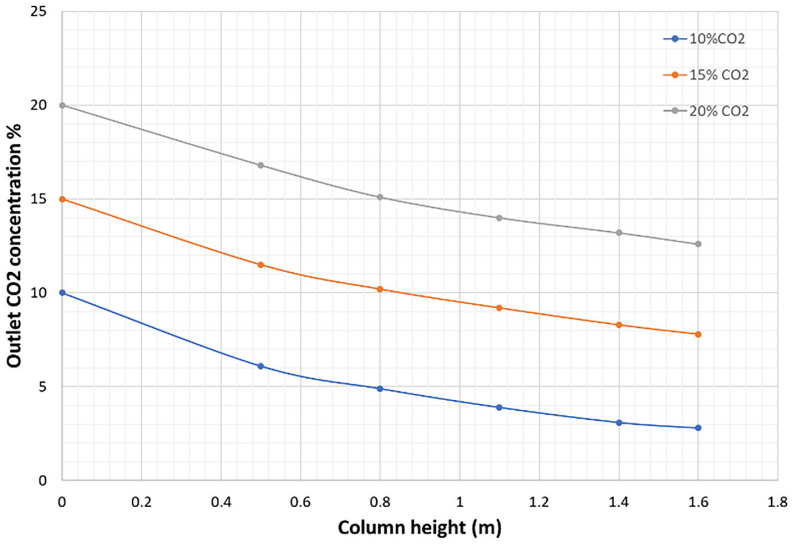


Figure 5. Effect of CO_2 inlet concentration along the column height

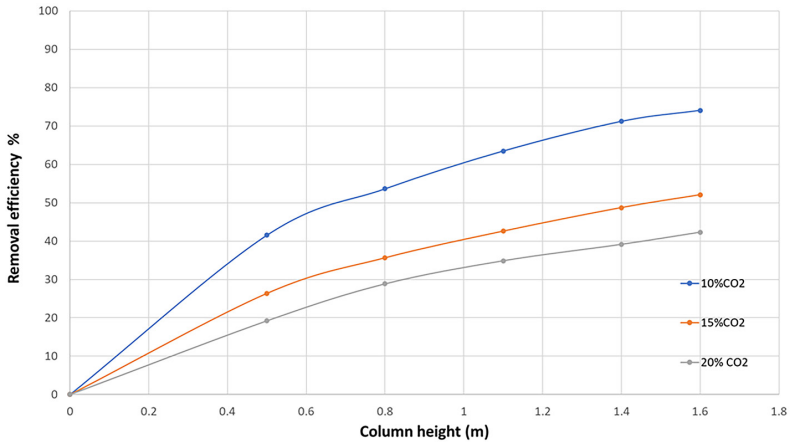


Figure 6. Removal efficiency at different CO_2 inlet concentrations

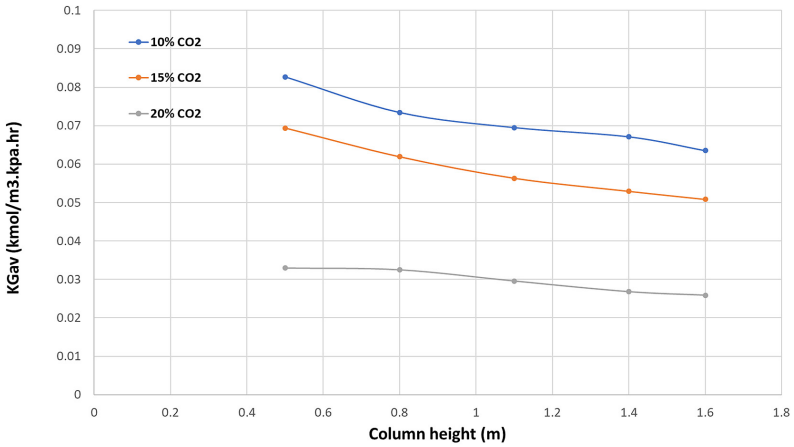


Figure 7. $K_G a_v$ at different inlet CO_2 concentrations

concentration can improve the gas-phase mass transfer drive force and as a result, the mass-transfer process, in the absorption systems used in this study, the resistance of mass-transfer primarily occurs in the liquid side; as a result, an increase in the gas-phase driving force has a negative effect on absorption performance (Sheng et al, 2016; Dhuyool et al., 2024).

MEA concentration's impact on absorption efficiency

Changes in amine concentration effect on absorption efficiency are investigated. Figures 8, 9, and 10 show that a greater solvent concentration causes a rise in mass transfer rate, KGav, and

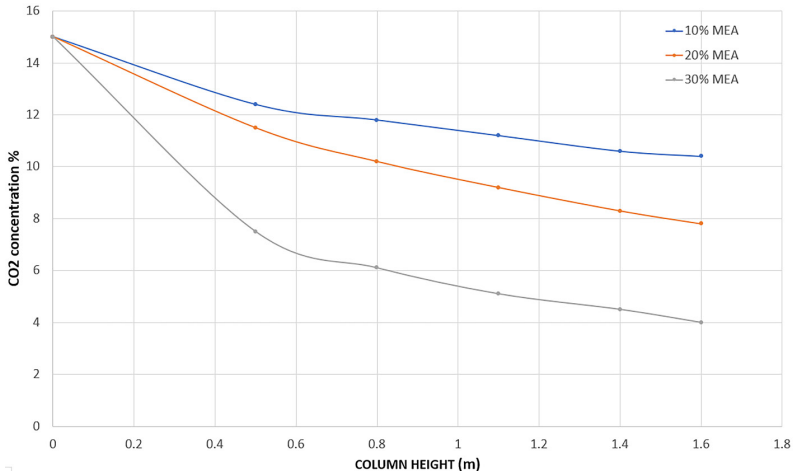


Figure 8. Effect of MEA concentration on CO_2 outlet concentration

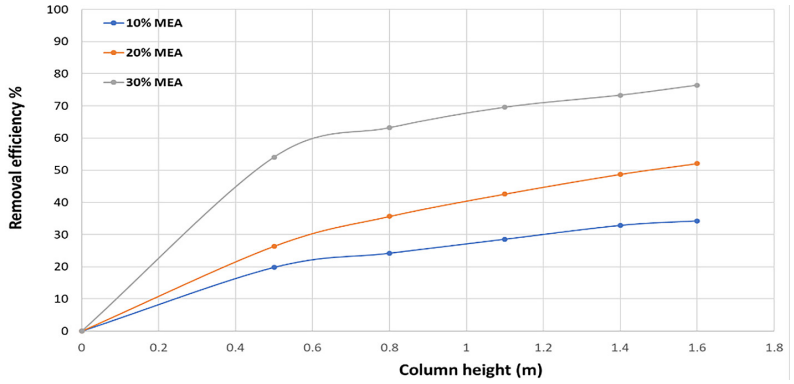


Figure 9. Effect of MEA concentration on removal efficiency

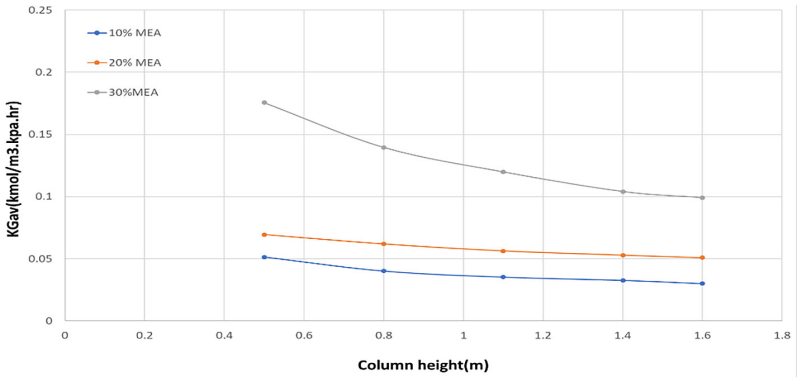


Figure 10. Effect of MEA concentration on KGav

absorption efficiency. This trend can be explained by the fact that when the solvent concentration rises, more solvent molecules are available per unit volume to absorb more CO_2 at the gas-liquid interface. Thus, the rate of the chemical reaction between MEA and CO_2 rises as the concentration of amine in the solution increases (Lin et al., 2019); Since the mixture's absorbing capacity is increased when the number of active sites increases thus an increase in carbon dioxide absorption (Janati et al., 2021). This leads to an improvement in CO_2 absorption performance.

The impact of CO_2 loading on mass transfer performance

The effect of CO_2 loading has been studied in this section to show the performance of the absorption process at different CO_2 loading: 0, 0.15, and 0.3 mol CO_2/mol amine; The CO_2 loading in the amine inlet stream appears to have an impact on the removal efficiency, and KGav value. Figure 11, 12, 13 illustrates these values decrease as the initial CO_2 loading rises. This pattern is to be caused when the solution is fully loaded or at its maximum capacity, which occurs, for example, in MEA solution when the carbon dioxide loading reaches a maximum of about 0.50 CO_2 moles /moles amine, absorption stops occur (Fu et al., 2012); also, the mass transfer driving force of steadily decreases as liquid loading grows throughout the column's length. Less free alkanol amine molecules are accessible to interact with carbon dioxide molecules, which has this effect as a result of the mass transfer being less efficient due to the increased carbon dioxide lean amine loading, as well as the $K_G a_v$ value being decreased (Demontigny et al., 2001).

The impact of lean amine temperature on mass transfer performance

The effect of lean amine temperature on mass transfer performance has been studied in this section to show the performance of the absorption

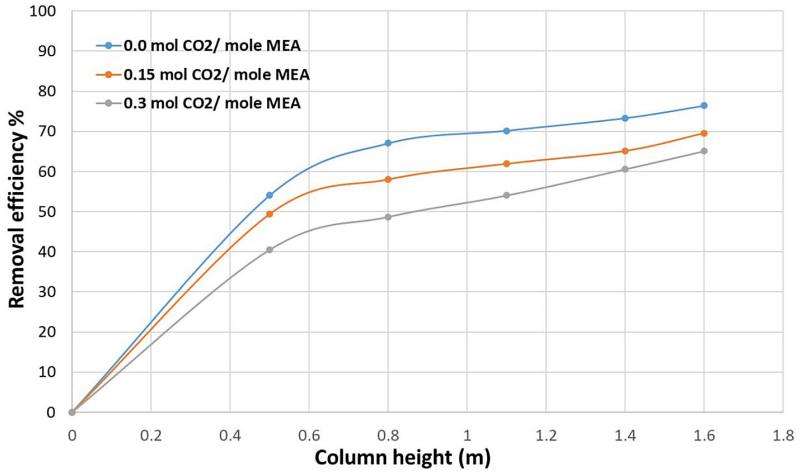


Figure 11. Effect of lean amine loading on CO_2 removal efficiency

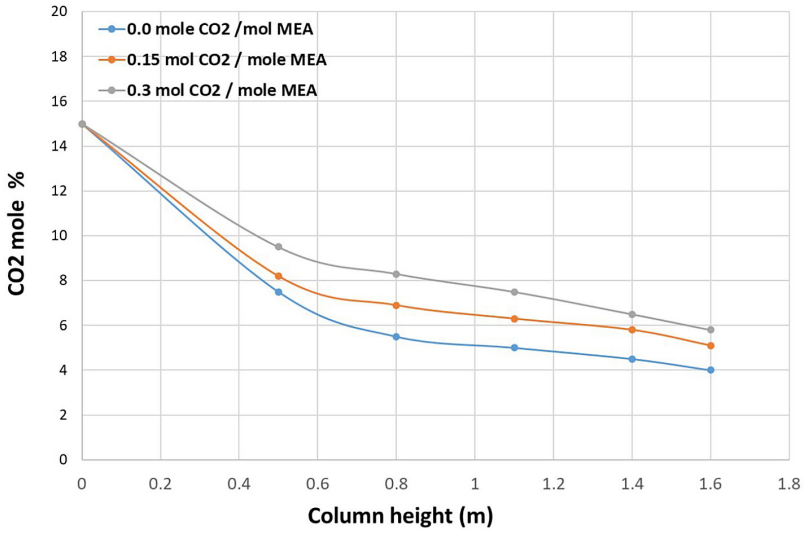


Figure 12. Effect of lean amine loading on CO_2 outlet concentration

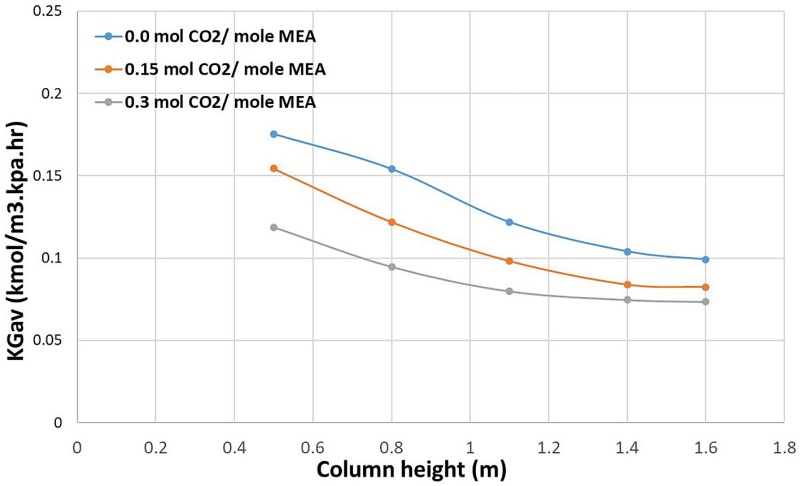


Figure 13. Effect of lean amine loading on $K_G a_v$

process at different lean amine temperatures: 298.15 K, 308.15 K, and 318.15 K. The lean amine temperature appears to have an impact on the removal efficiency, and KGav value. Figure 14, 15, 16 illustrates these values decrease as the lean amine temperature rises. This pattern is because, at high enough temperatures, reverse reactions may occur in the exothermic reaction of absorption of carbon dioxide mechanism; also, carbon dioxide's physical solubility in the absorbent decreases as the temperature rises because of an increase in the carbon dioxide vapor pressure above the solution (Tan et al., 2012). Above that, due to the fact that alkanol amine absorption capacity is temperature dependent, the effect of temperature on absorption capacity plays an important role; it drops when the temperature rises (Gul and Un, 2022).

Investigation of different amines groups on mass transfer performance

The optimum parameter value will be selected based on the operation results above to ascertain the mass transfer coefficient and removal efficiency. These parameters are used as abased operating conditions for testing various amine types, including DEA, TEA, DETA, TEPA, and others.

Based on the results of the preceding experiment, the investigation is conducted under optimal operating conditions. These operating parameters include a 15% CO_2 content in the input gas, a 6 LPM gas flow rate, a 30% weight concentration of amines, and a 20 ml/min liquid flow rate. The performance of different types of amines groups are shown in Figures 17, 18 and

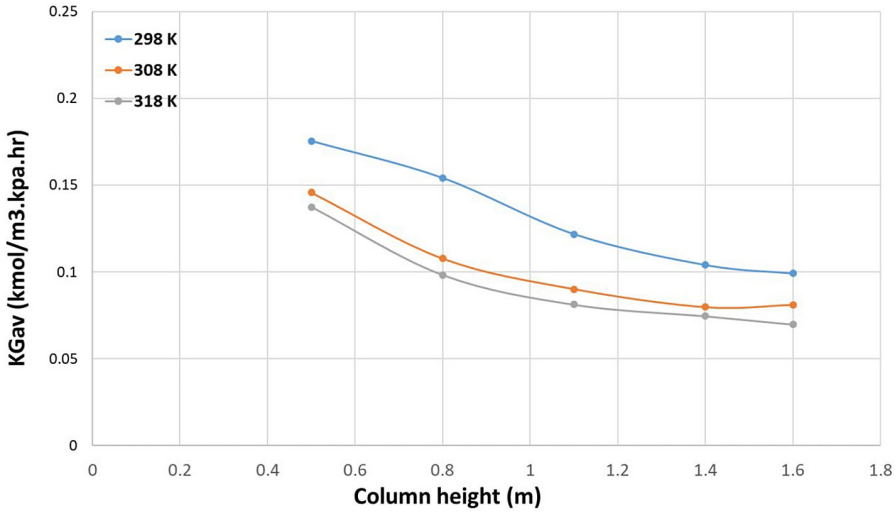


Figure 14. Effect of MEA concentration on CO_2 outlet concentration

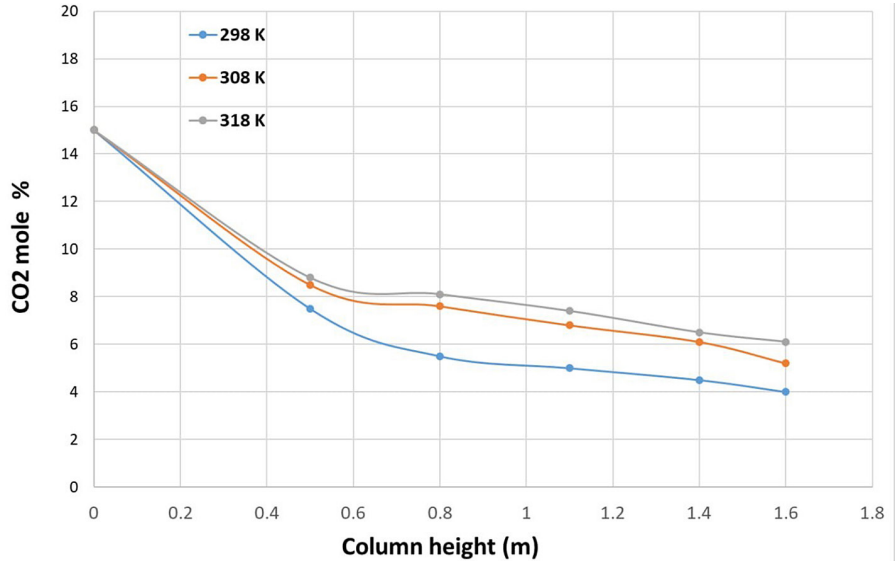


Figure 15. Effect of MEA concentration on CO_2 outlet concentration

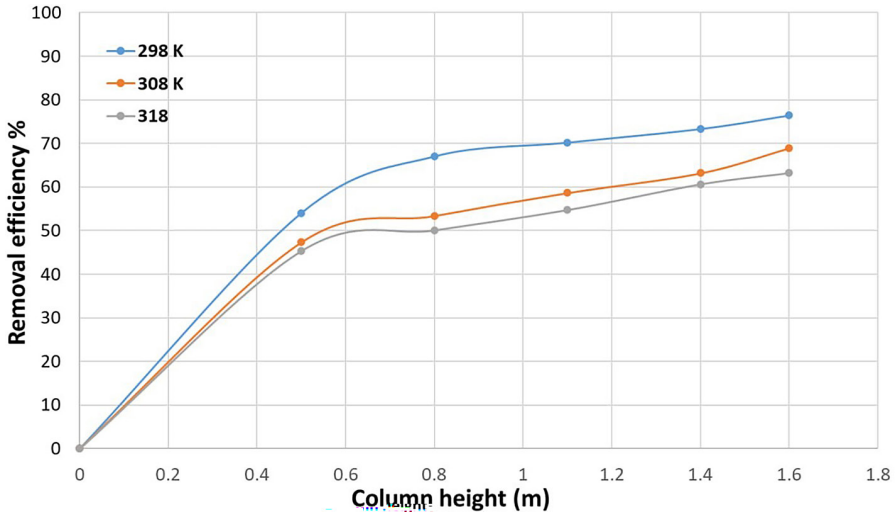


Figure 16. Effect of MEA concentration on CO_2 outlet concentration

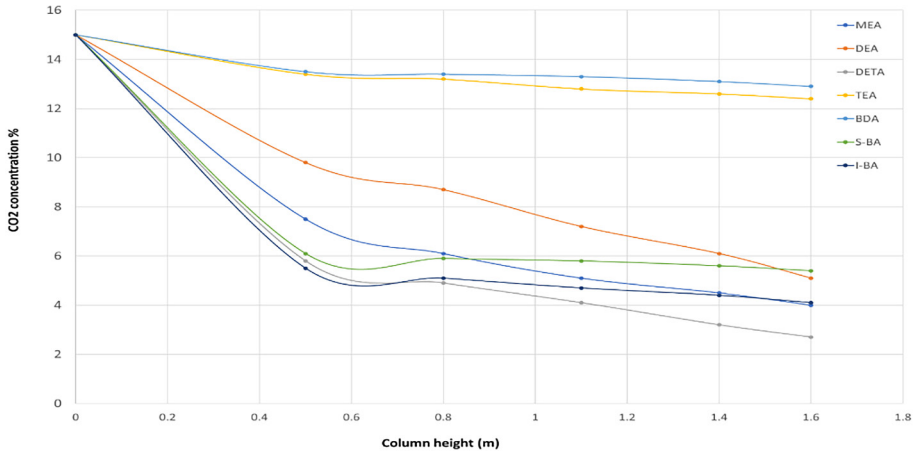


Figure 17. CO_2 % at different amines solution

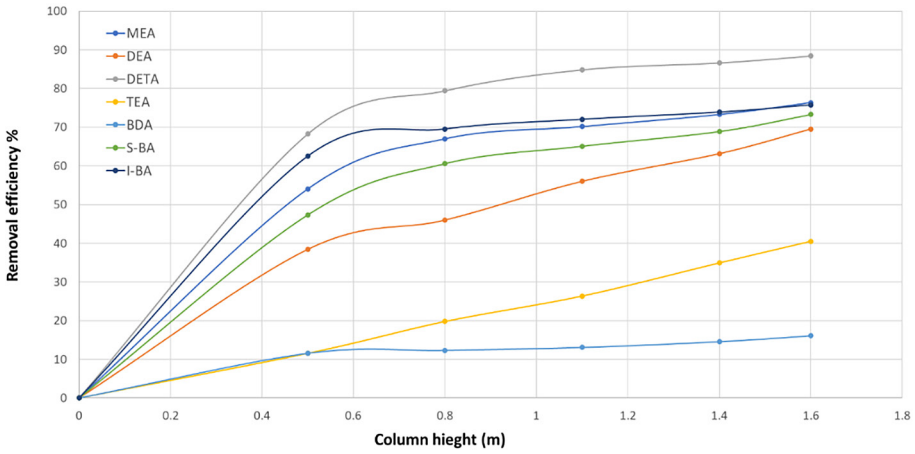


Figure 18. CO_2 removal of different amines solution

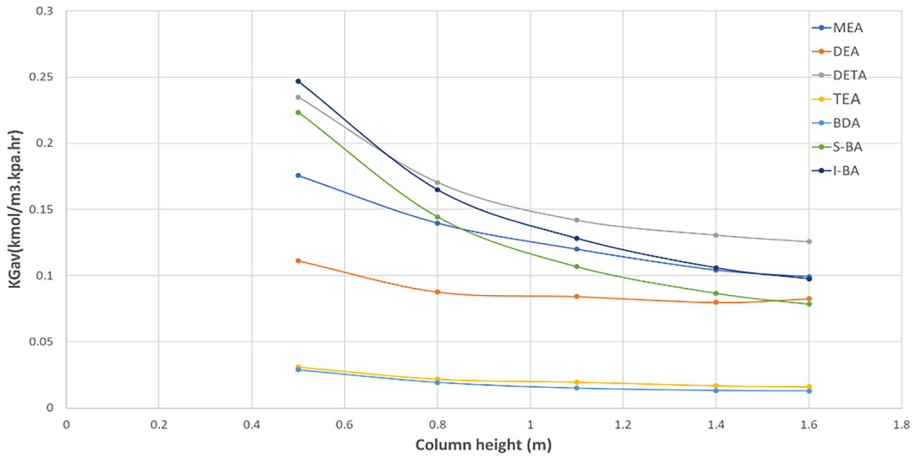


Figure 19. KGav of different amine solvent

19; where the removal efficiency and volumetric mass transfer coefficient are estimated using data provided by measuring CO_2 concentration along column height.

The results show that only DETA and I-BA have higher mass transfer rates and removal efficiency than MEA as a reference solvent used for industrial purposes. At 1.6 m height, the KGav of

DETA is around 0.13 while has a value of 0.95 for both MEA and I-BA. the removal efficiency of EDTA is 85% which is higher than the removal efficiency of MEA and I-BA which is only 76 %. this behavior depends on the number of amine groups and the carbon atomic number. the higher the amine group the higher the solvent capacity to absorb CO_2 such as DETA and I-BA (Chen et al., 2022). The hydrocarbon chain affects the rate of mass transfer due to the effect of the solubility of CO_2 in the amine solvent. the solubility of CO_2 in amine solutions decreases as the carbon group increases in the active compound. and this reduces the mass transfer rate and removal efficiency at smaller heights of the columns. The low performance is shown in TEA and BDA which gives low absorption capacity due to low amine groups relative to the hydrocarbon group. Others amine compounds gives an intermediate behavior in removal efficiency and mass transfer coefficient as shown in Figure 18 and 19.

CONCLUSIONS

Various type of amines was effectively chosen for the carbon dioxide capturing process employing a continuous packed-column scrubber by utilizing the response parameter including carbon dioxide concentration; absorption efficiency and overall mass transfer coefficient measurement as an indicator for the effectiveness of the capture process. The overall mass transfer coefficient and carbon dioxide removal efficiency of all alkanol amine solutions were found to increase with higher amine flow rates and amine concentrations; however, it was found to decrease with increasing inlet carbon dioxide concentration, lean amine loading, inlet amine temperature, and feed gas flow rate. the final loading values decrease in the order BZA/MEA 1/2 > BZA/MEA 1/1 > BZA/MEA 2/1. The obtained result suggests that a DETA solution could be an effective carbon dioxide absorbent. The limitations in the desorption efficiency of DETA and I-Ba are poorer compared to MEA. Energy costs during desorption need further optimization. Due high carbon density, solubility of DETA and I-BA is lower than MEA and this lead to need higher column bed to reach the maximum loading capacity of CO_2. These laboratory studies provide good initial insights into the efficiency and performance of carbon capture processes. However, these findings need to be validated at a larger scale, where factors such as flow dynamics, heat and mass transfer, and reaction kinetics can differ significantly.

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