



Special Issue Article

Intensification of the Mass Transfer of the Green CO₂ Capture Employing the Ternary Solution of Alkanolamine-Amino Acid

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ABSTRACT

The increased demand of the world for energy and its reliance on fossil fuels ultimately contribute to the surge in the levels of carbon dioxide in the atmosphere. To achieve a green, efficient carbon capture, a novel multi-component amine-amino acid solvent including methyldiethanolamine (MDEA), diisopropanolamine (DIPA), and Arginine (ARG) was designated for the CO₂ absorption in a T-microreactor. The potential absorption of the aqueous solutions of the desired mixed amines has been assessed through the CO₂ absorption percentage (AP) and the total volumetric gas-phase mass transfer coefficient (TGMTC) over a wide range of the gas flow rates (60-240 mL/min), solvent flow rates (2-6 mL/min), under the three mixing concentrations of MDEA: DIPA: ARG (28:8:4), (28:6:6), and (28:4:8) wt%. The research findings demonstrate an increment of 31% in the absorption percentage of CO₂ by reducing DIPA to 4 wt% and raising the concentration of arginine to 8 wt% in the ternary amine solutions. Additionally, the highest mass transfer coefficient of 38.06 (kmol/m³.h.kPa) was achieved utilizing the aqueous solution of MDEA+DIPA+ARG (28+4+8) wt%.

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1. Introduction

The increase in the world's energy demand and reliance on fossil fuels ultimately contribute to the increase of CO₂ in the atmosphere and global warming. With the aim of capturing CO₂, the developed absorption technologies are classified into three sections; pre-

combustion, post-combustion, and oxyfuel combustion [1]. The chemisorption of alkanolamines is mentioned among researchers as the most developed post-combustion technology for CO₂ capture [2], which has brought the best efficiency in the gas treatment industry. Despite the shortcomings

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of each amine, an alternative absorbent, methyldiethanolamine (MDEA), is widely employed as a tertiary alkanolamine for the removal of CO₂, as it has the desirable chemical and thermal stability, high loading capacity, and low corrosivity [3]. However, since the mid-1980s, MDEA-based mixed alkanolamine solvents have been proposed to further improve the MDEA absorption performance, including its relatively slow kinetics of the CO₂ absorption [4]. Regarding the benefits of diisopropanolamine, DIPA, as the secondary alkanolamine, including the effective removal of acid gases, no corrosive effects, thermal stability, less carbamate formation during reaction with CO₂, and less heat and energy requirement during regeneration, it has been proposed for CO₂ absorption experiments [5]. Furthermore, it has been reported that the reaction rate of DIPA is higher as compared to the aqueous solution of MDEA [6]. The challenge of utilizing three-amine solvents has rarely been detailed in recent decades. Dubois et al. [7] compared the rate of CO₂ absorption using a system consisting of MEA, MDEA, and PZ in a specific compact column. A high rate of CO₂ capture was achieved for an aqueous solution of MEA+PZ (15+30) wt%. MEA-DIPA-AMP solvents with various ratios of (15-10-5), (10-10-10), (7.5-7.5-7.7), and (5-15-10) wt% were employed by Song-Mo Kim et al. to assess the mechanism of the CO₂ absorption by Song-Mo Kim et al. [8]. The results indicated that the CO₂ loading ratio increases in the order of AMP \gg DIPA > MEA.

Regardless of the type of the solvent, the liquid-gas contactor is one of the most important issues in optimizing the mass transfer and energy consumption. Recently, the new intensification technology, known as microreactors or microchannels, has gained considerable attention. They have numerous

merits including a short length of diffusion, high concentration gradient, high surface-to-volume ratio, strong controllability, and small sample requirement [9]. Various studies have been carried out in the last few years regarding the employment of the microreactor technology for the CO₂ absorption. The experimental assessment of the separation of CO₂ employing the aqueous 40 wt% MDEA-piperazine solvent indicated that at a solvent flow rate of 9 ml/min of solvent, an operating temperature of 40 °C, and input concentration of 10 vol.% of CO₂, the highest value of the total gas phase mass transfer coefficient, TGMTC, of 202.50 kmol/m³h.kPa was achieved [10]. Morais et al. [11] utilized an aqueous MEA solution in a Y-junction microreactor to evaluate CO₂ capture from a gas mixture including CO₂+CH₄+N₂ chemically and physically. The liquid-side mass transfer coefficient, K_{La}, in the designed Y-microreactor, was found to be 24.68 s⁻¹, a significantly higher value than that of the typical two-phase contactor devices. The results of the CO₂ mass transfer into water-lean MDEA+PZ+methanol showed that the concentration of the selected solvent in the range of 30-50 wt%, as the second significant operating variable, enhances the values of CP, TGMTC, and the mass transfer flux by 25, 58, and 23% respectively [12]. Besides, concerning the CO₂ mass transfer into the blended aqueous solution of DEA-amino acid, it is investigated that the utilization of arginine solutions with more amine functional groups would significantly increase the values of the CO₂ removal efficiency and TGMTC, to 95.98% and 73.76 kmol/m³.h.kPa in a T-microchannel [13].

Regarding prior investigations, whereas mixed alkanolamine blends perform superiorly for the CO₂ absorption under the lower partial pressures of CO₂ than that of single-

component alkanolamine solvents, they are not resistant to the solvent loss and oxidative degradation from an environmental standpoint. With the aim of carbon dioxide capture in a T-shaped microreactor, a multi-component solvent using MDEA, DIPA, and arginine has been used in this study. Moreover, the values of AP (%) and TGMTC ($\text{kmol/m}^3 \cdot \text{h} \cdot \text{kPa}$) were experimentally investigated in a range of the

operating variables of the concentration of the solution, solvent flow rate, and gas flow rate.

2. Experimental

2.1. Material and Methods

All chemicals and their properties are summarized in Table 1. The solutions were prepared with 60 wt% distilled water (40 wt% amine mixed solution).

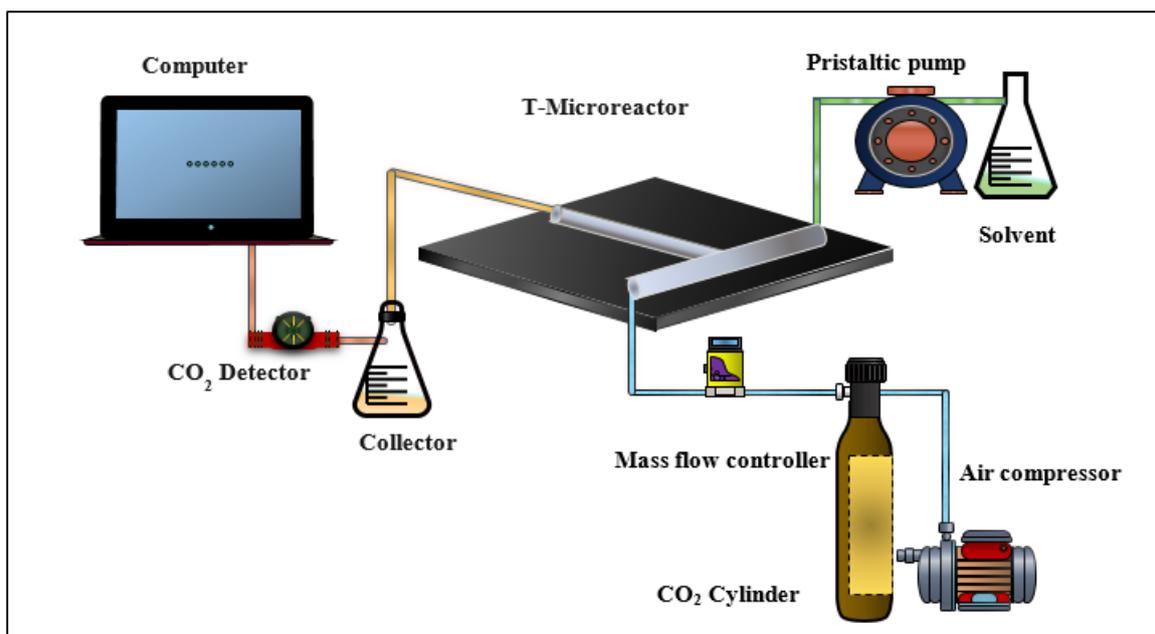
Table 1. The details of the used chemicals in the CO₂ capture experimental tests.

Component	Chemical formula	Molar mass ($\text{gr} \cdot \text{mol}^{-1}$)	Solubility in water ($\text{g} \cdot \text{L}^{-1}$)
Methyldiethanolamine (MDEA)	$\text{C}_5\text{H}_{13}\text{NO}_2$	119.164	Miscible
Diisopropanolamine (DIPA)	$\text{C}_6\text{H}_{15}\text{NO}_2$	133.191	870 @ 20 °C
L-Arginine (ARG)	$\text{C}_6\text{H}_{14}\text{N}_4\text{O}_2$	174.204	14.87 @ 20 °C
Carbon dioxide gas (CO ₂)	CO ₂	44.01	1.45 @ 25 °C

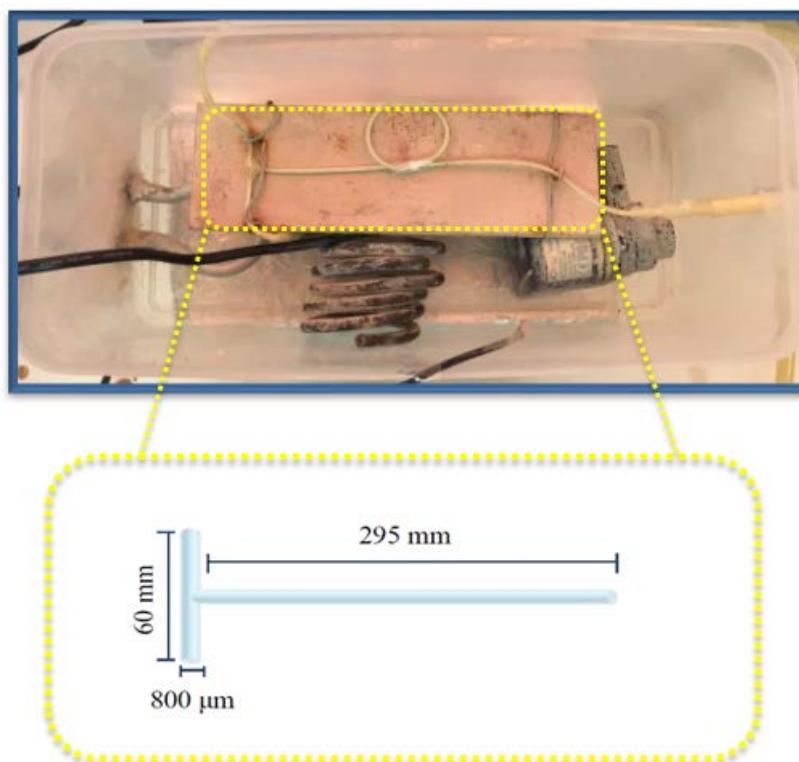
2.2. Experimental Setup

The CO₂ capture experiments have been conducted utilizing a microreactor featuring a T-form junction layout, and fabricated from polymethylmethacrylate, PMMA, with a diameter of 0.8 mm and length of 29.5 cm, as illustrated in Fig. 1. For the CO₂ absorption tests, the microreactor was positioned in a water bath. The bath temperature was regulated employing a thermostatic recirculating water bath, which maintained the bath temperature at 45 °C. The volumetric flow rate of the inert gas and CO₂ were measured and fed to the microreactor by two

mass flow controllers MFCs, (GPC series, BREEZENZ, Apasco, Iran) from an air compressor and CO₂ cylinder, respectively. From the other side of the microchannel, the entering solvent stream was adjusted via a peristaltic pump (BT-100-1 F, China). Subsequently, the gas and liquid phases contacted each other at the T-junction of the microreactor. The output mixed stream of the microreactor was collected in a bottle, then the concentration of CO₂ was determined using a CO₂ sensor (CM-0123 20%, COZIR-WR sensor, Germany) connected to a computer.



(a)



(b)

Figure 1. (a) The chematic, and (b) the actual and dimensions of the experimental test facility for the CO₂ capture process.

For CO₂ absorption experiments, CO₂ and a blended solution of MDEA + DIPA + ARG were used. Owing to industrial constraints, the aqueous mixture of MDEA and DIPA with amino acid ARG had the total concentrations

of 40 wt% of amine. ARG, as a chemical promotor, was added to MDEA and DIPA portions to intensify the CO₂ capture percentage to that of the aqueous MDEA+DIPA. The operating ranges of the inlet gas

containing CO₂ (Q_g), and also blended amine solution (Q_l) were determined as 60-240 mL/min and 2-6 mL/min respectively, for the effective mass transfer zone in the slug annular two-phase flow pattern [14]. Each experiment

was carried out at 45°C with the constant atmospheric pressure. Table 2 summarizes the operational parameters controlling the absorption process.

Table 2. The range of the operating conditions of CO₂ capture in the microreactor.

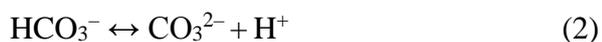
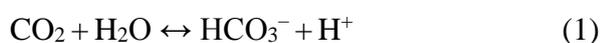
Parameter	Content
Solvent	Aqueous solution of MDEA+DIPA+ARG
Temperature (°C)	45
Pressure	Atmospheric
CO ₂ concentration (vol.%)	15
Gas flow rate (mL.min ⁻¹)	60-240
solvent flow rate (mL.min ⁻¹)	2-6
MDEA concentration (wt.%)	28
DIPA concentration (wt.%)	4-8
L-Arginine concentration (wt.%)	4-8

3. Theories

3.1. Mechanism of the Chemical Reaction

The following reactions have been used to describe the CO₂ absorption into the amine-amino acid aqueous multi-component solution, MDEA-DIPA-ARG:

The formation of carbonate and bicarbonate;



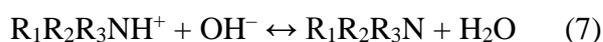
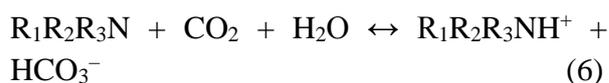
Producing R₂NH⁺CO₂⁻ from the CO₂ reaction with DIPA (R₂NH⁺);



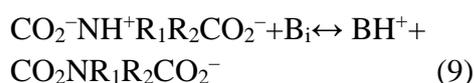
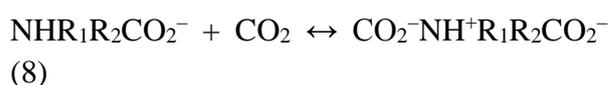
Forming stable carbamate from the zwitterion;



MDEA (R₁R₂R₃N) and CO₂ react in the liquid phase via the base-catalyzed hydration;



The amino group of ARG (NHR₁R₂CO₂⁻) forms an intermediate complex.



It is noted that within the aqueous solution of MDEA, there exists a lack of the production of any carbamate compounds through the direct reaction of CO₂ with a tertiary amine. This

absence stems from the fact that there is no hydrogen atom existing that can be displaced or substituted by CO₂ during this particular process. Additionally, some literature has reported on the direct reaction of DIPA with carbon dioxide [15]. Amino acids play imperative roles through the formation of zwitterion in the basic environment, despite their low rate of the CO₂ uptake [16]. Lastly, the zwitterion is deprotonated in the basic media, including R₂NH, H₂O, and OH⁻ in the aqueous amine-amino acid solution [17].

3.2. Definition of TGMTC (kmol/m³.h.kPa) and AP (%) as the Responses

A comprehensive analysis of the total volumetric gas phase mass transfer coefficient, TGMTC (K_{Gav}), and the absorption percentage (AP) has been performed to assess the efficiency of the absorbed CO₂ gas performance, as a specific and complete criterion [18]. To gauge the extent of CO₂ capture into the multi-component amine-amino acid solvent, one can establish the volumetric mass transfer flux (N_{Aav}), which is characterized based on two-film hypothesis, as depicted in Fig. 2. Within this theory lies the assumption that both the liquid and vapor phases consist of thin films alongside bulk regions. All resistance to mass transfer is concentrated within the thin boundary layer adjacent to the phase interface [19]. Therefore, under the steady-state condition, the CO₂ transfer takes place from its gaseous phase through a static film within its aqueous phase due to the driving force ($y_{A,G} - y_A^*$).

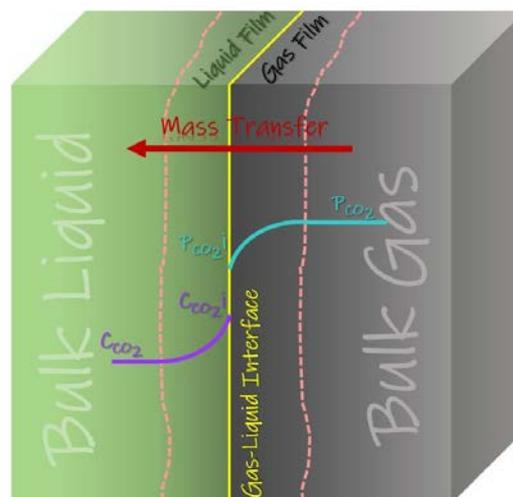


Figure 2. The schematic graph of the two-film mass transfer theory of the CO₂ absorption [20].

Regarding the overall mass balance at the microreactor inlet and outlet, the volumetric mass flux of CO₂ is defined based on the total pressure (P) of the system and the driving force ($y_{A,G} - y_A^*$), as follows [21]:

$$N_A a_V = K_G a_V P (y_{A,G} - y_A^*) \quad (10)$$

In this case, y_A^* , $y_{A,G}$, and a_V express the equilibrium mole fractions of component A at the gas-liquid interface, the mole fractions of component A in the gas stream, and interfacial area per volume (m^2/m^3) respectively. Considering the CO₂ reacts with the solvent in any length of the element (dh), the overall differential mass balance has been formulated as demonstrated below:

$$N_A a_V dh = G_I dY_A \quad (11)$$

In the above relation, the mole ratio of component A and the molar flow rate of the inert gas are denoted by Y_A and G_I respectively. By combining relations (10) and (11), it is plausible to establish a correlation, capable of calculating the total volumetric gas-phase mass transfer coefficient (TGMTC) in terms of K_{Gav} (kmol/m³.h.kPa):

$$K_G a_V = \frac{G}{ZP} \left[\ln \left(\frac{Y_{A, \text{inlet}}}{Y_{A, \text{outlet}}} \right) + (Y_{A, \text{inlet}} - Y_{A, \text{outlet}}) \right] \quad (12)$$

In the above correlation, $Y_{A, \text{inlet}}$ and $Y_{A, \text{outlet}}$ are the relative CO_2 molar ratio in the entry and exit gas of the absorption bed, Z symbolizes the length of the T-microreactor (m), P represents the pressure of the system (kPa), and G is the flow rate of the inert gas (kmol/h.m^2).

The CO_2 absorption percentage (AP) is calculated using Eq. (13). The terms “inlet C_{CO_2} ” and “outlet C_{CO_2} ” refer to the concentration of CO_2 at the entrance to and exit of the microchannel.

$$\text{AP}(\%) = \left(\frac{\text{Inlet } C_{\text{CO}_2} - \text{Outlet } C_{\text{CO}_2}}{\text{Inlet } C_{\text{CO}_2}} \right) \times 100 \quad (13)$$

4. Results and Discussion

4.1. Effect of the Concentration of Amine-amino Acid (wt%)

The values of the CO_2 absorption percentage (AP) and the total volumetric gas-phase mass transfer coefficient (TGMTC) for various mixing concentrations of secondary amine-amino acid, including MDEA: DIPA: ARG (28:8:4), (28:6:6), and (28:4:8) wt% have been illustrated in Fig. 3. As seen, increasing the concentration of DIPA in the mixture constantly declines the responses. Specifically, under a constant $Q_l=4 \text{ mL/min}$ and $Q_g=180 \text{ mL/min}$, with rising concentration of DIPA at its highest level of 8 wt% in the solvent, MDEA+DIPA+ARG (28+8+4) wt%, the absorption percentage of 72.44 %, has been measured in the experiments. In turn, at the lowest concentration of 4 wt% of DIPA, it is possible to achieve the AP value of 82.38%. In this situation, the value of TGMTC has risen by 31%.

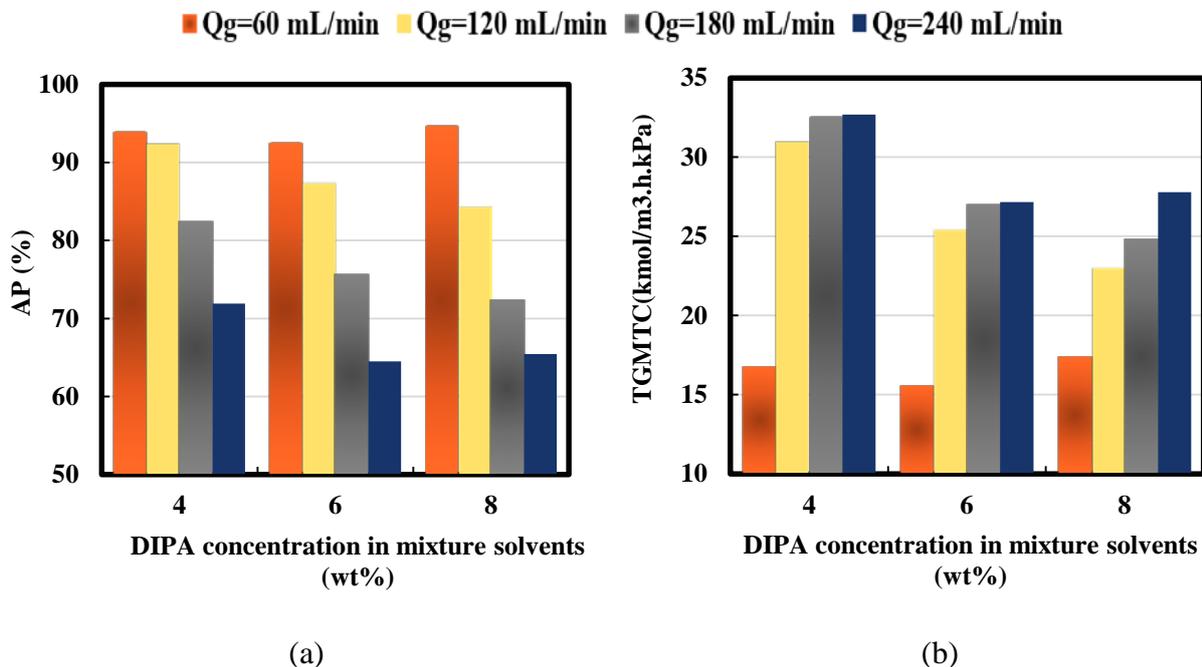


Figure 3. The effects of the concentration of DIPA and ARG (wt%) on; a) the absorption percentage (%), and b) the total gas-phase mass transfer coefficient ($\text{kmol/m}^3.\text{h.kPa}$).

Finally, the values of the AP have showed reductions as follows; MDEA:DIPA:ARG

(28:4:8) wt% > (28:6:6) wt% > (28:8:4) wt% with absorption percentage of (AP=82.38%) >

(AP= 75.61%) > (AP=72.44%) respectively. This behavior can be attributed to the fact that as the total concentration of the solvent is kept constant at 40 wt.%, the reduction in the weight fractions of DIPA leads to an increase in the concentration of arginine. Arginine with a similar structure to MEA and DEA, undergoes a direct and reversible reaction with CO₂ by creating a zwitterion intermediate. This intermediate is then deprotonated by the bases within the solution, producing a stable carbamate. Hence, it is proved that the higher rate of reaction in the MDEA solution is due to carbamate [22]. Furthermore, arginine possesses a greater number of active amine groups, as compared

to DIPA, so the rising concentration of ARG indicates a greater quantity of free active absorbent molecules and a higher capability for the CO₂ absorption [23]. All these depict the superiority of arginine over DIPA for CO₂ capture in the designed T-microreactor.

4.2. Effect of the Solvent Flow Rate-Q_i

Figs. 4a, b depict the influence of the solvent flow rate on the measured values of AP and TGMTC for the ternary amine solutions of MDEA: DIPA: ARG. From the figure, it is evident that as the inlet solvent flow rate increased within the range of 2-6 mL/min, both the absorption percentage and TGMTC followed an amplifying trend.

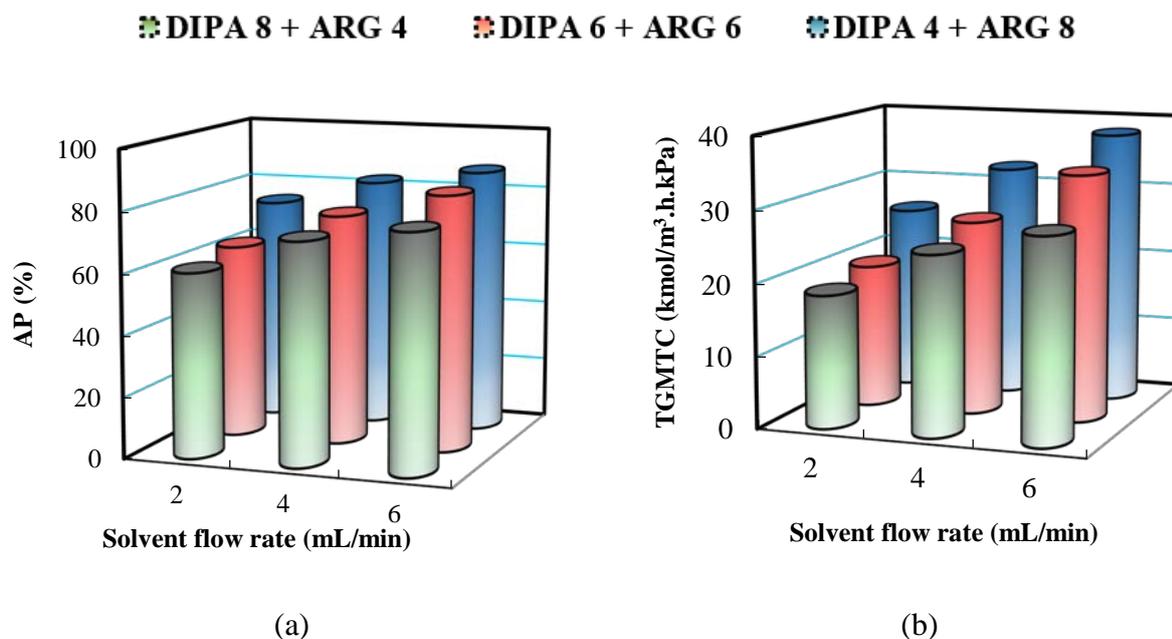


Figure 4. The values of the a) absorption percentage, and b) TGMTC versus the solvent flow rate at T=45°C for the various concentrations of DIPA-ARG (wt%).

In the case of AP, increasing the solvent flow rate results in a higher concentration of DIPA and L-arginine within the microchannel. Increasing the content of amine-amino acid in the mixture leads to increased chemical reactions with CO₂, as well as a large interfacial area between the solvent and gas streams. As a result, less carbon dioxide would

be present in the output of the microreactor [24].

In the case of TGMTC, the increasing inlet solvent flow rate changes the obtained results for MDEA: DIPA: ARG solutions. To further elaborate, the total gas-phase mass transfer coefficients have increased from 18.40 to 28.18, 19.90 to 34, and 25.81 to 38.06

($\text{kmol/m}^3 \cdot \text{h} \cdot \text{kPa}$) with the concentrations of (28:8:4), (28:6:6), and (28:4:8) wt% respectively. The two-phase surface renewal rate and the intensity of the vortices in the liquid slugs may both be improved by increasing the solvent flow rate. This increases the convection velocity and bubble moving speed, leading to the acceleration of the gas-liquid mass transfer and TGMTC [25].

4.3. Effect of the Feed Gas Flow Rate- Q_g

The relationship between the volumetric flow rate of the gas phase with both the CO_2 absorption percentage and TGMTC is illustrated in Figs. 5a, b. As seen, at a given solvent flow rate of 4 mL/min, rising the gas flow rate from 60 to 240 mL/min has led to a decrease in the values of the AP but an increase in the obtained values of the TGMTC. More specifically, the values of AP for DIPA+ARG (4+8) wt% follow a falling trend from 93.94 % to 71.91 %, as the volumetric flow rate of the gas stream increases from 60 to 240 mL/min. The decrease in the capture efficiency is resulted from reducing the gas retention time due to the increase in the gas velocity [26].

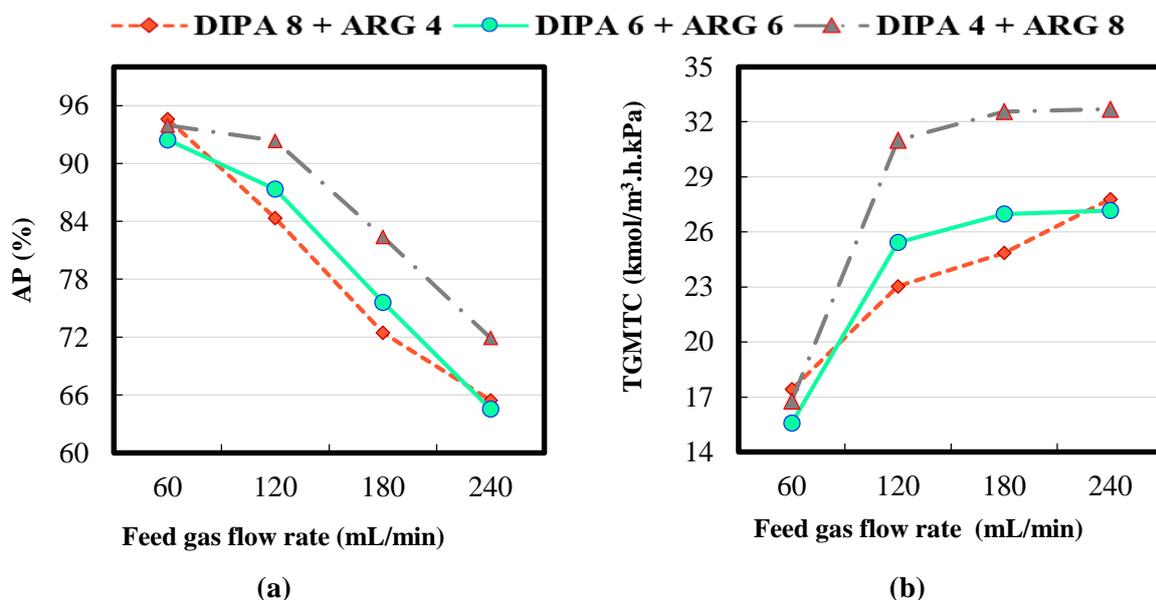


Figure 5. The result of increasing the inlet gas flow rate on a) AP (%), b) TGMTC ($\text{kmol/m}^3 \cdot \text{h} \cdot \text{kPa}$) for the various concentrations of DIPA-ARG (wt%) at $T=45\text{ }^\circ\text{C}$ and $Q_L=4\text{ mL/min}$.

On the other side, According to Fig. 5b, for the same concentration of the solvent, the values of the TGMTC have experienced a rising trend from 16.78 to 32.69 $\text{kmol/m}^3 \cdot \text{h} \cdot \text{kPa}$ by changing the gas-phase flow rate from 60 to 240 mL/min. The clear reason for the intensification can be traced back to Eq.12, in which the $(Y_{\text{inlet}} - Y_{\text{outlet}})$ term declines by rising the gas flow rate. It seems that the interface turbulence would happen by rising the flow

rate of the gas stream, resulting in an increase in the bubble length, and a rise in the specific surface area. The internal circulation of the liquid slug then gets easier. The enhancement of the mass transfer in the region where the gas and liquid phases are separated is the next outcome. This increase in the shear force between the liquid slug and the microreactor's wall can also be accomplished by reducing the thickness of the liquid boundary layer,

promoting convective circulation within the liquid slug, and shortening the length of the slug [25].

4.4. Comparisons of Different gas-liquid contactors

Next, a thorough comparison of the solvent used in the microreactor is made with other solvents in other gas-liquid contactors to examine their mass transfer potential for scaling-up, as shown in Figs. 6a, b.

In particular, The TGMTC values of the several gas-liquid contactors, including the falling film, spray column, packed bed, and rotating packed bed contactors are compared in Fig. 6a to the current T-mixer microreactor, in

which CO₂ capture was performed utilizing the blended solution of MDEA+DIPA+ARG.

According to the figure, the T-shaped microreactor is shown to have a higher efficiency than the traditional CO₂ capture devices, since even under different operating conditions, it constitutes a considerable portion of the TGMTC value during the CO₂ absorption process. As an illustration, a maximum K_{GAV} limit of 5.2 kmol/m³.h.kPa has been assigned to the conventional rotating packed bed contactor. In this investigation, the TGMTC value of 38.06 kmol/m³.h.kPa, which is significantly higher than the typical value found in the typical liquid-gas contactors by at least seven orders of magnitude, was achieved.

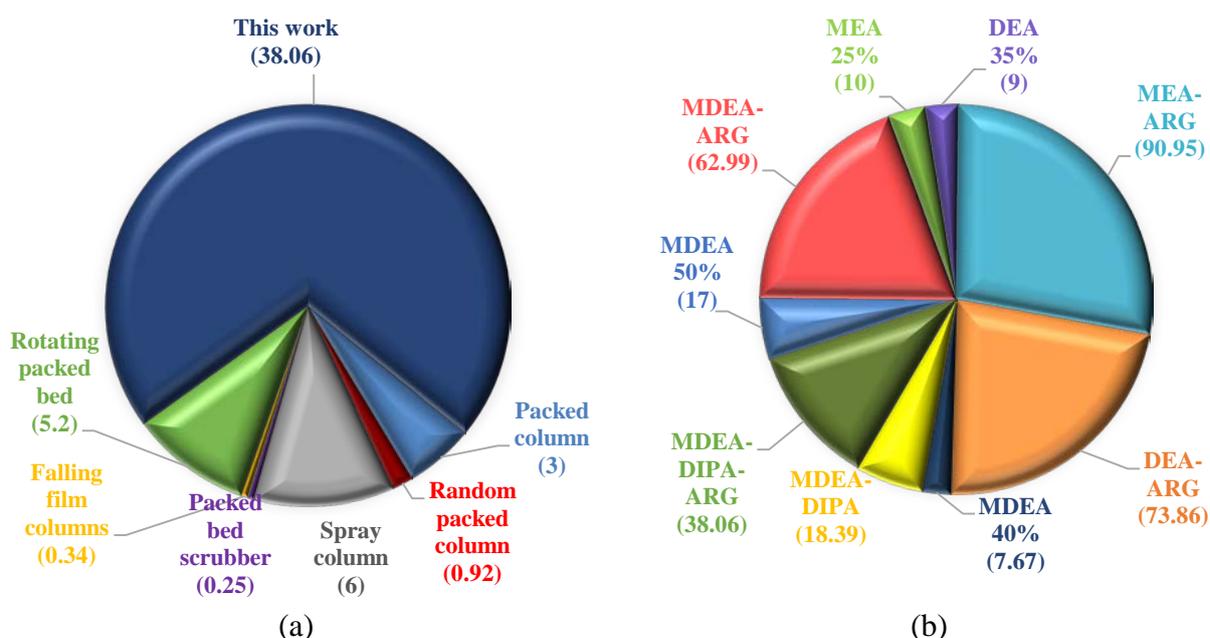


Figure 6. The comparison of the maximum K_{GAV} obtained from a) various gas-liquid mass transfer contactors, and b) different solvents employed in microreactors [27-34]

Finally, the results reveals that the mass transfer efficiency of carbon dioxide absorption is equally influenced by the choice of solvent and the type of micro-contactor. This work provides a comparative analysis of the mass transfer coefficients that were found for the different solvents, used in the microreactor, as shown in Fig. 6b. Based on

the figure, the highest TGMTC values are arranged in an ascending order of: MDEA 40% (7.67) < DEA 35% (9) < MEA 25% (10) < MDEA 50% (17) < MDEA: DIPA (18.39) < MDEA: DIPA: ARG (38.06) < MDEA: ARG (62.99) < DEA: ARG (73.86) < MEA: ARG (90.95) kmol/m³.h.kPa. The blended green amine-amino acid binary solutions produce

more K_{GAV} than the ternary blended solvent that compromises MDEA-DIPA-ARG, despite having the highest density and viscosity. Nonetheless, the advantages of the secondary commercial amine DIPA in combination with the green arginine amino acid should not be disregarded.

5. Conclusion

To inhibit the emission of acidic CO_2 , in the current work, the effect of a ternary mixed aqueous solution of MDEA, DIPA, and arginine amino acid was experimentally investigated in terms of the carbon dioxide absorption percentage (AP, %) and the total mass transfer coefficient of the gas phase (TGMTC, $kmol/m^3 \cdot h \cdot kPa$). A T-shaped microreactor with $D = 800 \mu m$ and $L = 295 mm$ was utilized to intensify the gas-liquid mass transfer. The results illustrated that by rising the concentration of DIPA to the highest level (8 wt%) under a constant $Q_l = 4 mL/min$ and $Q_g = 180 mL/min$, the absorption percentage of 72.44 % has been measured in the experiments. In turn, at the lowest concentration of DIPA (4 wt%), it is possible to achieve the AP value of 82.38%. In this situation, the total gas phase mass transfer coefficient, TGMTC, has risen by 31%. Besides, both the absorption percentage and TGMTC pursue amplified trends by raising the flow rate of the inlet solvent in the range of 2-6 mL/min. For the ternary MDEA: DIPA: ARG solution, at a constant liquid flow rate of 4 mL/min and gas flow rate of 180 mL/min, AP is reduced as follows; (28:4:8) wt% = 82.38% > (28:6:6) wt% = 75.61% > (28:8:4) wt% = 72.44%. The maximum mass transfer coefficient of $38.06 kmol/m^3 \cdot h \cdot kPa$ was obtained for MDEA+DIPA+ARG (28+4+8) wt%, despite varying the operating conditions, which was more than seven times the value found in the earlier research having employed traditional CO_2 absorption devices. Finally, in

the studied ternary solution of akanolamine-amino acid, all experimental mass transfer results illustrated the considerable effect of arginine on the secondary alkanolamine, DIPA, for CO_2 capture.

Acknowledgments

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