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Article

Performance of Water-Lean Solvent for Postcombustion Carbon Dioxide Capture in a Process-Intensified Absorber: Experimental, Modeling, and Optimization Using RSM and ML

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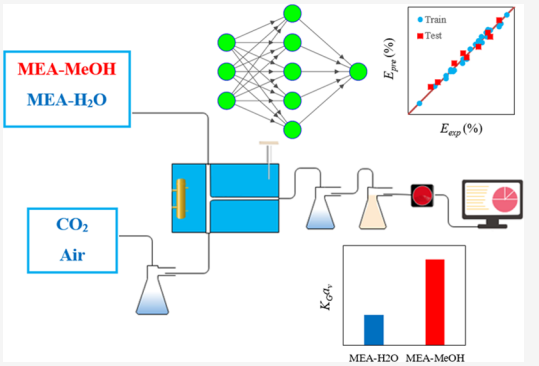
ABSTRACT: In recent years, the absorption of carbon dioxide by water-lean solvents has received special attention. In this study, carbon dioxide absorption was performed in a microfluidic device using a water-lean monoethanolamine solution. The effect of different operating conditions, including inlet solvent flow, solvent concentration, and temperature, on CO_2 removal efficiency, overall mass transfer coefficient, and mass transfer flux were investigated. Response surface methodology (RSM) was used to analyze and optimize the responses. The maximum removal efficiency of 92.24% and mass transfer coefficient of 155.12 kmol/m^{3}hrkPa were achieved at the solvent concentration of 30 wt %, 40 °C, and solvent flow of 9 mL/min. For amine-methanol solvent in these conditions, the overall volume mass transfer coefficient was 18.09% higher than the aqueous solvent. The mass transfer coefficient obtained in this study was significantly higher than the values reported for packed towers. Furthermore, the artificial neural network (ANN) method as a branch of machine learning (ML) models was utilized for modeling the CO_2 removal efficiency of the water-lean monoethanolamine solvent. The number of neurons and different transfer functions have been optimized in MLP and RBF models to select optimum ANN. The results show that the MLP model with a tangent sigmoid transfer function showed the best performance with an RMSE value of 0.35103, which indicates that the utilized ML method predicts the CO_2 removal efficiency of water-lean amine solution to satisfactory levels.



1. INTRODUCTION

Energy consumption has increased with the development of industry since the 20th century. Currently, fossil fuels supply most of the world's energy demand, which leads to an increase in greenhouse gas emissions. Fossil fuel power plants, especially coal, have the highest levels of carbon dioxide emissions. The flue gas produced in power plants is produced at atmospheric pressure. Environmental problems related to the emission of pollutant gases due to the combustion of fossil fuels include air pollution and the harmful effects of greenhouse gases, especially global warming.[1] Therefore, it is essential to remove carbon dioxide from the atmosphere or rather to prevent its release. To this end, researchers are looking for a solution to reduce carbon dioxide emissions, and to avoid this global hazard, carbon capture and sequestration (CCS) are considered a vital strategy. One of these strategies is postcombustion capture by absorption. Absorption is widely used due to its high efficiency, low energy consumption, and environmental friendliness.[2] The standard method is chemical absorption with alkanolamines, which is commercialized, and the separation process is relatively complete and efficient. The advantages of alkanolamines include a high reaction rate, high efficiency, reasonable price, and chemical stability. The disadvantages of alkanolamines include high energy consumption, low amine absorption capacity, sensitivity to oxygen, and corrosion in equipment.[3] Among alkanolamines, monoethanolamine (MEA) is a crucial absorbent for removing carbon dioxide, which has advantages such as high reactivity, low solvent cost, low molecular weight, and high absorption capacity based on weight. Adding an amine to a physical solvent such as water and methanol increases the carbon dioxide uptake capacity several times, so the use of water-lean solvents has been developed.[4,5] Today, the use of water-lean solvents in the carbon dioxide absorption process due to high absorption rate, double absorption qualities, and lower energy demand in the regeneration has increased.[6] Recently, more attention has been paid to the water-lean solvent of monoethanolamine-methanol because it has a faster rate of carbon dioxide uptake and a higher mass transfer rate than

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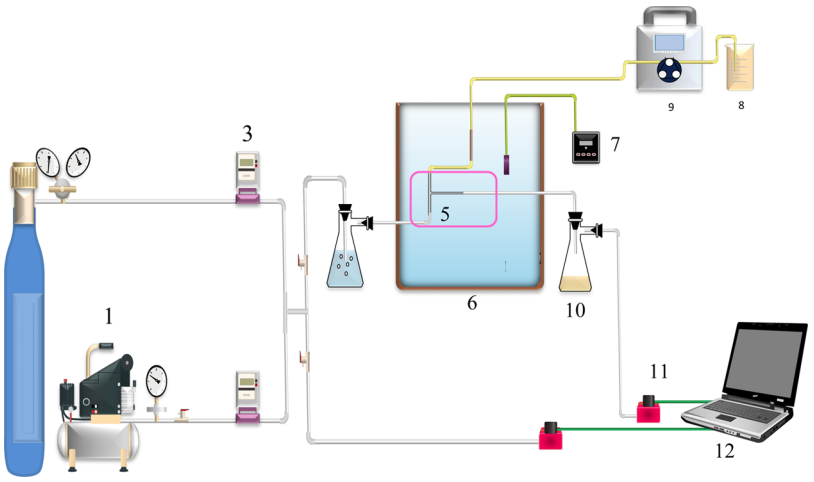

Figure 1. Schematic diagram of experimental setup including (1) air compressor, (2) carbon dioxide cylinder, (3) mass flow controller, (4) humidifier, (5) microchannel, (6) water bath, (7) temperature controller, (8) fresh solvent tank, (9) solvent pump, (10) separator, (11) CO_2 measuring sensor, and (12) recorder.

aqueous monoethanolamine.[7] Gao et al. studied the absorption of carbon dioxide by the monoethanolamine-methanol solution in a packed tower. They stated that solvent concentration, temperature, liquid flow, and carbon dioxide concentration in the inlet gas stream are effective in the carbon dioxide absorption process.[8] Sama et al. investigated the overall mass transfer coefficient and mass transfer flux in the aqueous and nonaqueous solutions of monoethanolamine in a packed column.[7]

The process intensification by microchannel in the carbon dioxide absorption process is one of the new methods, that has been paid great attention to in recent years. A device in which chemical reactions occur in ducts smaller than 1 mm is called a microreactor. One of the most important characteristics of microreactors is their high surface-to-volume ratio. Therefore, more performance can be expected from them. Shrinkage of the reactor is associated high heat and mass transfer rates for a multiphase system and rapid mixing.[9] The performance of the microchannel reactor in the gas absorption process has been evaluated by TeGrotenhuis et al. They showed that microchannel technology could remove more than 90% of carbon dioxide by diethanolamine solution in less than 10 s.[10] In another study, Chen et al. examined the process of carbon dioxide uptake by monoethanolamine in the microchannel. Their research showed that the amount of carbon dioxide in the output decreased to 300 ppm. In this study, the effects of monoethanolamine concentration, temperature, pressure, and microchannel dimension ratio were investigated.[11] Lam et al. showed that microchannel has a high absorption efficiency due to its high surface-to-volume ratio, short mass transfer time and heat transfer, and high reaction rate.[12] Napada et al. argued that absorption operation in the packed tower has high operating costs, high maintenance, and high risk in terms of safety points. Therefore, in their study, to increase the efficiency of carbon dioxide absorption from flue gases, they proposed a microchannel. For this purpose, an ammonia solution was used for absorption in the microchannel. The results showed that a solution of 10 wt % ammonia at a temperature of 30 °C and pressure of 300 kPa has an absorption efficiency of about 96.45%.[13] In another study, water was used as a green solvent to absorb carbon dioxide from the microchannel. Under optimum operating conditions, the highest carbon dioxide uptake was 70.9%. Adding monoethanolamine to water increased the absorption efficiency by 88%.[14] In our previous study, the authors have applied methyl diethanolamine-piperazine-methanol solvent in a microchannel to investigate the effect of operating conditions on the performance of the proposed water-lean solvent.[15]

Recently, Pashaei et al. have applied artificial neural network (ANN) and response surface methodology (RSM) methods for modeling of postcombustion carbon capture process using alkanolamine solvent to recognize the process variables and optimal conditions.[16] Moreover, Hosseinpour et al. evaluated and classified the various ML approaches utilized in the postcombustion process (PCC). They focused on simulation, optimization, and solvent management in PCC by the ML technique.[17]

Therefore, in this research, a microchannel has been used for the carbon dioxide absorption process from simulated flue gas. Then, for the first time, the performance of the water-lean solution of monoethanolamine-methanol in the microfluidic device has been compared. The effects of different operating conditions, including inlet solvent flow, solvent concentration, and temperature, on CO_2 removal efficiency, overall mass transfer coefficient, and mass transfer flux were investigated. Response surface methodology by a three-level full factorial design was used to analyze and optimize the responses. Furthermore, the ANN method as a powerful tool for conducting complex computations that facilitate the training of computer algorithms to perform specific tasks with exceptional precision was utilized for modeling the CO_2 removal efficiency of the water-lean monoethanolamine solvent. The ANN architectures, transfer function, and number of neurons were optimized to select the optimum ANN.

2. THEORY

2.1. Chemical Reaction. In the CO_2 absorption process by alkanolamine, physical and chemical absorption will occur at the same time. First, CO_2 penetrates the gas−liquid interface via physical absorption. Then, the penetrated CO_2 reacts in the liquid film via chemical absorption.[18] The chemical reaction of CO_2 in the liquid phase follows the zwitterion reaction mechanism.[19,20]

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$$CO_2 + 2MEA \leftrightarrow MEACOO^- + MEAH^+ \quad (1)$$

$$CO_2 + MEA \overset{K1}{\leftrightarrow} MEAH^+COO^- \quad (2)$$

$$MEAH^+COO^- + B \overset{KB}{\leftrightarrow} MEACOO^- + BH^+ \quad (3)$$

Here, B can represent monoethanolamine, methanol, or water.[21] The overall reaction of CO_2 with monoethanolamine is a reversible equilibrium reaction.[22]

2.2. Response Definition. To calculate the overall volumetric mass transfer coefficient based on gas phase (K_Ga_v), removal efficiency (E), and volumetric molar transfer flux ($N_{CO_2}a_v$), eqs 4−7 were used.[23]

$$K_Ga_v = \frac{F}{ZP}\left\{\ln\left(\frac{Y_{in}}{Y_{oyt}}\right) + (Y_{in} - Y_{out})\right\} \quad (4)$$

$$Y = \frac{y}{1-y} \quad (5)$$

$$E = \frac{Y_{in} - Y_{out}}{Y_{in}} \times 100 \quad (6)$$

$$N_{CO_2}a_v = \frac{F}{Z}\cdot(Y_{in} - Y_{out}) \quad (7)$$

In the above equations, F is the inert gas flow (kmol/m^2·hr), P is the pressure (kPa), Z is the microchannel length (m), and Y_{in} and Y_{out} are the molar ratios of CO_2 in the inlet and outlet gas stream, respectively.

3. MATERIALS AND METHODS

3.1. Materials. In this study, methanol with a high purity of 99% from Shiraz Petrochemical Company of Iran, monoethanolamine with a high purity of 99% from Shazand Petrochemical Company of Iran, and 98% sulfuric acid from Kimia Pars Iran Company were used.

3.2. Experimental Method. Figure 1 shows the diagram of the experimental setup. For each test, the bath temperature was controlled according to the test design. Also, the solvent with specific concentrations according to the test design is prepared and entered the microchannel according to the specified flow. As gas and liquid pass through the microchannel, the gas mixture reacts with the solvent, and the effluent stream enters the separation chamber for separation. Subsequently, the carbon dioxide concentration in the gas phase was measured by a sensor, and the data was recorded. The microreactor used in the experiment was T-shaped and had a length of 29 cm and an inner diameter of 0.8 mm with a circular cross section. Air is mixed with carbon dioxide to simulate flue gas. Absorption of carbon dioxide by the aqueous solution of monoethanolamine and the nonaqueous monoethanolamine-methanol solution in the microchannel with different operating conditions, including 3−9 mL/min inlet fluid flow, 10−30 wt % solvent concentration, and 20−40 °C temperature, has been studied. In this study, a three-level full factorial design was used to investigate the effect of operating parameters on the carbon dioxide absorption process. The experiments were conducted at atmospheric pressure, a CO_2 inlet gas concentration of 15 vol %, and an inlet gas flow of 300 mL/min. All experiments were repeated three times. More details were described in our previous study.[15]

4. RESULTS AND DISCUSSION

4.1. Experimental Design. The response surface three-level factorial method was used to design experiments and statistically analyze the results. The reason for using this method was the high accuracy of this technique. Three variables of temperature, solvent concentration, and solvent flow in three levels were investigated according to Table 1. In this study, the CO_2 absorption process by aqueous and nonaqueous MEA solution was investigated in terms of the overall mass transfer coefficient based on gas phase (K_Ga_v), removal efficiency (E), and volumetric molar flux ($N_{CO_2}a_v$). The number of tests for the nonaqueous monoethanolamine-methanol solution was 27. The same number of tests have been performed for the aqueous monoethanolamine solution. Table 2 shows the test results for the aqueous and nonaqueous solutions of monoethanolamine under different operating conditions.

Table 1. Range and Levels of Variables

			levels		
variables	unit	symbol	level 1	level 2	level 3
MEA concentration	wt %	A (MEA Conc.)	10	20	30
temperature	°C	B (T)	20	30	40
fluid flow	mL/min	C (QL)	3	6	9

4.2. Analysis of Variance. To check the rationality of the model in the prediction of E, K_Ga_v, and N_Aa_v, the predicted data must be statistically analyzed by analysis of variance (ANOVA). In order to avoid lengthening the paper, only the results related to the removal efficiency and mass transfer coefficient are presented in Tables 3 and 4. According to the ANOVA table, the P-value of the models was <0.0001, which shows the validity of the models. For the removal efficiency of MEA−MeOH solvent, the value of F obtained for the temperature variable was 324.64, which shows that the most influential variable on the removal efficiency is temperature. This is due to the nonlinear dependence of the reaction rate on temperature. The value of F for the solvent concentration was 249.96, which indicates that the variable is influential in removal efficiency. The value of F for the liquid flow is 108.93, which is the least effective variable for the removal efficiency. According to Table 4 for the mass transfer coefficient of nonaqueous MEA solution, it can be seen that the order of importance of the parameters is $B > A > C$.

Using regression analysis for the data, the following quadratic equations were obtained to calculate the removal efficiency and overall mass transfer coefficient for the aqueous and water-lean MEA solutions

$$E_{(MEA\text{-}MeOH)} = 91.15 + 1.53A + 1.75B + 1.01C - 0.4650AB + 0.3683AC - 0.3317BC + 0.8022B^2 - 0.3311C^2 \quad (11)$$

$$E_{(MEA\text{-}H_2O)} = 88.22 + 2.67A + 2.09B + 1.30C - 0.6742AB - 0.4558BC - 0.9533A^2 + 0.4633B^2 \quad (12)$$

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Table 2. Test Results for Aqueous and Water-Lean Monoethanolamine Solutions

				aqueous			water-lean		
run	MEA conc. (wt %)	T (°C)	Q_L (mL/min)	E (%)	K_Ga_v (kmol/m^{2}h kPa)	$N_{CO_2}a_v$ (kmol/m^{3}h)	E (%)	K_Ga_v (kmol/m^{2}h kPa)	$N_{CO_2}a_v$ (kmol/m^{3}h)
1	10	20	3	81.21	103.54	711.36	92.66	158.31	811.62
2	20	20	3	85.11	117.24	745.56	89.96	140.23	788.05
3	30	20	3	86.89	124.65	761.09	90.32	142.28	791.12
4	10	30	3	82.37	107.31	721.53	93.7	167.15	820.76
5	20	30	3	87.67	128.25	767.94	88.5	132.3	775.19
6	30	30	3	88.47	132.13	774.94	93.36	164.12	817.78
7	10	40	3	87.07	125.17	762.73	89.53	137.72	784.22
8	20	40	3	90.32	142.28	791.12	93.64	166.64	820.26
9	30	40	3	90.84	145.53	795.74	89.2	135.95	781.34
10	10	20	6	82.42	114.58	739.5	91.06	144.36	796.29
11	20	20	6	86.46	122.8	757.37	91.87	152.44	804.77
12	30	20	6	88.72	133.44	777.18	89.84	139.54	787
13	10	30	6	83.57	111.48	732.06	91.19	147.82	798.82
14	20	30	6	87.4	126.99	765.61	92.35	155.92	808.94
15	30	30	6	90.55	143.71	793.21	90.26	141.94	790.63
16	10	40	6	88.25	131.04	773.01	95.23	183.18	834.16
17	20	40	6	90.84	145.53	795.74	92.88	160.13	813.61
18	30	40	6	91.54	150.16	801.9	91.43	149.36	800.9
19	10	20	9	84.28	114.04	738.26	93.49	165.27	818.92
20	20	20	9	87.59	127.85	767.25	94.71	177.58	829.89
21	30	20	9	90.91	145.93	796.29	88.73	133.5	777.22
22	10	30	9	86.12	121.34	754.39	89.97	140.26	788.1
23	20	30	9	90.02	141.76	790.33	93.4	164.67	818.32
24	30	30	9	92.05	153.73	806.31	88.11	130.36	771.82
25	10	40	9	88.78	133.78	777.72	87.07	125.51	762.73
26	20	40	9	91.31	148.56	799.86	94.56	175.66	828.35
27	30	40	9	92.24	155.12	808	92.9	161.8	813.76

Table 3. Analysis of Variance for Removal Efficiency of Aqueous and Water-Lean MEA Solution

	water-lean					aqueous				
source	sum of square	d_f	mean square	F-value	P-value	sum of square	d_f	mean square	F-value	P-value
model	125.98	9	14.00	82.85	<0.0001	252.55	9	28.06	75.07	<0.0001
A-MEA conc.	42.23	1	42.23	249.96	<0.0001	128.75	1	128.75	344.43	<0.0001
B-T	54.85	1	54.85	324.64	<0.0001	78.54	1	78.54	210.12	<0.0001
C-Q_L	18.40	1	18.40	108.93	<0.0001	30.29	1	30.29	81.03	<0.0001
AB	2.59	1	2.59	15.36	0.0011	5.45	1	5.45	14.59	0.0014
AC	1.63	1	1.63	9.64	0.0064	0.0184	1	0.0184	0.0492	0.8270
BC	1.32	1	1.32	7.81	0.0124	2.49	1	2.49	6.67	0.0194
A^2	0.4392	1	0.4392	2.60	0.1253	5.45	1	5.45	14.59	0.0014
B^2	3.86	1	3.86	22.86	0.0002	1.29	1	1.29	3.45	0.0808
C^2	0.6578	1	0.6578	3.89	0.0650	0.2604	1	0.2604	0.6967	0.4155
residual	2.87	17	0.1689			6.35	17	0.3738		
cor total	128.85	26				258.90	26			

$$K_Ga_{v(\text{MEA-MeOH})} = 146.94 + 10.61A + 12.27B + 7.19C + 3.84AC + 7.04B^2 + 0.4633B^2 \quad (13)$$

$$K_Ga_{v(\text{MEA-H}_2\text{O})} = 131.70 + 12.34A + 9.62B + 6.45C - 3.10A^2 + 2.66B^2 + 0.4633B^2 \quad (14)$$

Figure 2 compares removal efficiencies for experimental data and model predictions. According to the predicted values of R^2 and adjusted R^2, there is a good correlation between the model prediction and the experimental data. The predicted R^2 has a reasonable forecast of the data because its difference with adjusted R^2 is less than 0.2.

4.3. Main Factors. *4.3.1. Effect of Solvent Concentration.* Chemical absorption is a determining factor in the CO_2 absorption process. Therefore, increasing the amine concentration in the solvent increases the chemical absorption and consequently increases the absorption rate. Increasing the solvent concentration increases the number of active absorption sites and increases the removal efficiency. Figure 3 shows the changes in removal efficiency, overall volumetric mass transfer coefficient, and volumetric molar flux for aqueous and water-lean monoethanolamine solution at the temperature of 30 °C and liquid flow of 6 mL/min.

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Table 4. Analysis of Variance for Mass Transfer Coefficient of Aqueous and Water-Lean MEA Solution

	water-lean					aqueous				
source	sum of square	d_f	mean square	*F*-value	*P*-value	sum of square	d_f	mean square	*F*-value	*P*-value
model	6233.77	9	692.64	71.06	<0.0001	5322.86	9	591.43	45.17	<0.0001
A-MEA conc.	2026.09	1	2026.09	207.86	<0.0001	2740.96	1	2740.96	209.34	<0.0001
B-T	2708.97	1	2708.97	277.92	<0.0001	1664.65	1	1664.65	127.14	<0.0001
C-Q_L	931.11	1	931.11	95.52	<0.0001	747.68	1	747.68	57.10	<0.0001
AB	20.25	1	20.25	2.08	0.1676	10.16	1	10.16	0.7757	0.3907
AC	177.25	1	177.25	18.18	0.0005	31.14	1	31.14	2.38	0.1415
BC	12.44	1	12.44	1.28	0.2742	26.73	1	26.73	2.04	0.1712
A^2	28.76	1	28.76	2.95	0.1040	57.74	1	57.74	4.41	0.0510
B^2	297.46	1	297.46	30.52	<0.0001	42.38	1	42.38	3.24	0.0898
C^2	31.43	1	31.43	3.22	0.0903	1.42	1	1.42	0.1083	0.7461
residual	165.70	17	9.75			222.59	17	13.09		
cor total	6399.48	26				5545.45	26			

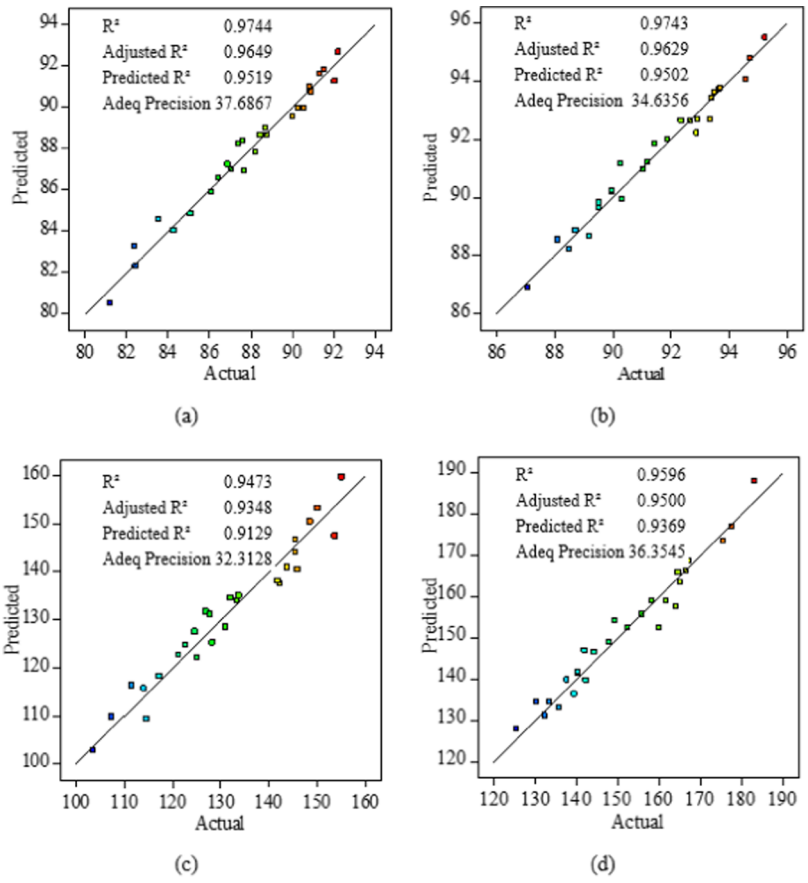

Figure 2. Parity plot of experimental and predicted values for removal efficiency of (a) aqueous and (b) water-lean MEA solution and mass transfer coefficient of (c) aqueous and (d) water-lean MEA solution.

As shown in Figure 3, with increasing concentration for both solvents, the removal efficiency, mass transfer coefficient, and molar flux increase. The higher the solvent concentration during the test, the more active sites of the solvent and the more solvent molecules interact with CO_2. As a result, the overall mass transfer coefficient, removal efficiency, and volumetric mass transfer flux increase. For the monoethanolamine-methanol solution at a concentration of 30 wt %, the removal efficiency was about 2.81%, the mass transfer coefficient was 14.2%, and the molar flux was 3.10% more than the aqueous solution of monoethanolamine. According to the results, it would be inferred that methanol improves the mass transfer phenomenon due to the superior physical absorption of CO_2. Moreover, methanol has a lower viscosity than water, which enhances the CO_2 diffusion coefficient in the solution. Hence, the mass transfer of MEA−MeOH solution increases, in accordance with the previous studies in packed beds.[22,24,25]

4.3.2. Effect of Temperature. Figure 4 displays the effect of the temperature on removal efficiency, mass transfer coefficient, and molar flux for the aqueous and water-lean solutions of monoethanolamine at a concentration of 20 wt % and a liquid flow of 6 mL/min. Based on the figure, with increasing temperature from 20 to 40 °C, the mass transfer

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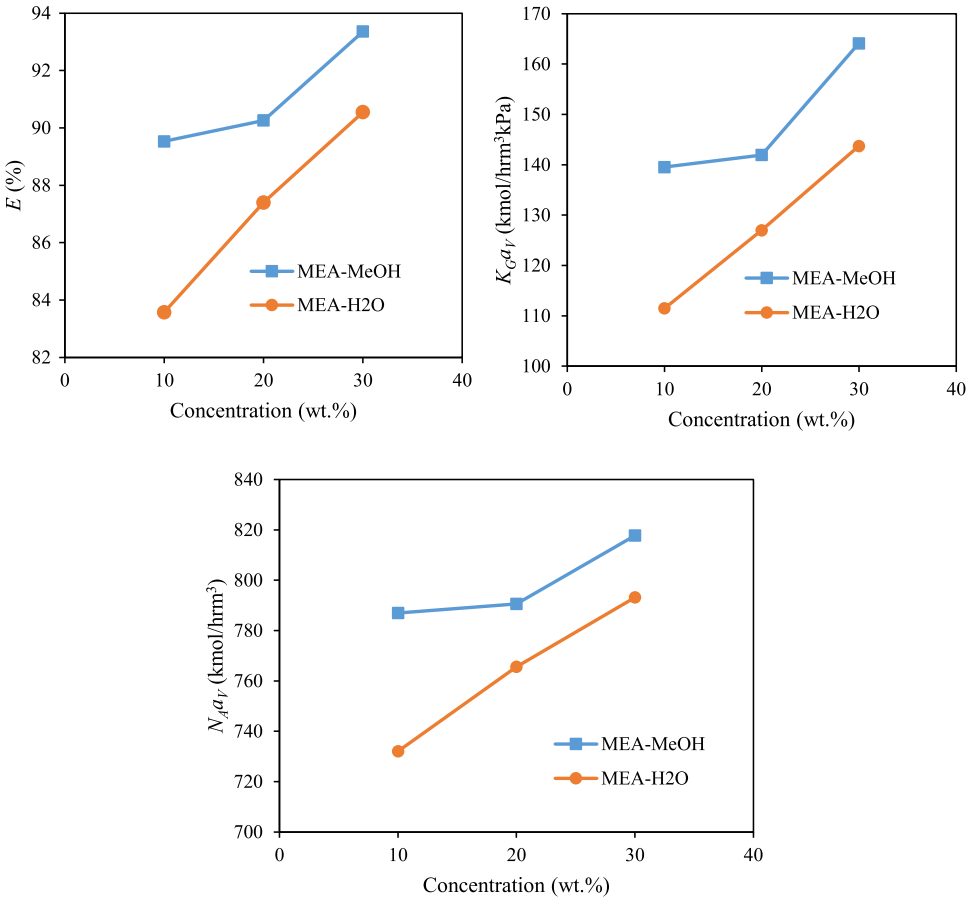

Figure 3. Effect of solvent concentration for the aqueous and water-lean solutions of monoethanolamine at 30 °C and liquid flow of 6 mL/min on $K_G a_v$, E, and $N_{CO_2} a_v$.

coefficient in nongaseous solution increases by 18.8%, increasing from 140.23 to 166.64 kmol/m^2.hr.kPa. Temperature is an essential parameter in absorbing carbon dioxide by the amine solvent. As the temperature increases, the solvent's viscosity decreases, and as a result, the diffusion coefficient in the solvent increases. Physical and chemical absorption occur simultaneously. In physical absorption, increasing temperature causes a decrease in absorption, but in chemical absorption, increasing temperature according to Arrhenius relation causes a constant increase in reaction rate exponentially.[26]

A comparison of results for the aqueous and water-lean solutions of monoethanolamine at 40 °C reveals that the removal efficiency of the hybrid MEA−methanol solvent was 2.80% higher than that of the aqueous solution. This enhancement for mass transfer coefficient and molar flux was 11.45% and 3.08%, respectively. Such improvements have been reported for the CO_2 absorption performance of DEA−MeOH solvent.[5]

4.3.3. Effect of Solvent Flow. The effect of liquid flow on the removal efficiency, mass transfer coefficient, and molar flux for the aqueous and water-lean solutions of monoethanolamine is shown in Figure 5. For this study, the temperature was 30 °C and the solvent concentration was 20 wt %. As shown in the figure, as the fluid flow increases from 3 to 9 mL/min for both solutions, the removal efficiency, the overall volumetric mass transfer coefficient, and the volumetric mass transfer flux increase. Increased fluid flow means more fresh solvent is available for carbon dioxide absorption. As the liquid flow rises, the gas−liquid mass transfer interface rises. The higher the interfacial area, the higher the removal efficiency and mass transfer flux. Moreover, increasing the fluid flow increases the fluid velocity in the microchannel, resulting in more turbulence. For both solvents, the turbulence results in better mixing of liquid and gas, reduces the thickness of the liquid film in the microchannel, and according to two film theory, enhances the mass transfer coefficient. For the monoethanolamine-methanol solution at a liquid flow of 6 mL/min, the removal efficiency is about 2.42%, the overall transfer coefficient is 10.04%, and the molar flux is 3.00% higher than the aqueous solvent of monoethanolamine. According to the higher physical solubility of methanol, the solvent flow has a greater impact on the mass transfer phenomenon in the water-lean solvent.

Analysis of the results shows that during the experiment, with increasing liquid concentration, temperature, and liquid flow, the removal efficiency, overall mass transfer coefficient, and volumetric mass transfer flux increased.

4.4. Interaction Effects. The significance of the interaction of parameters was investigated by the computed P-value in the ANOVA table. If the P-value is less than 0.05, the interaction is significant. The temperature variable was an essential operational variable in this study. It was the most significant effect among the operational variables. Table 3 shows the analysis of variance for the removal efficiency response. According to the table, the interaction of temperature and concentration, temperature and liquid flow, and concentration and liquid flow in the model is significant. Figure 6(a) shows that increasing the temperature from 20 to 30 °C

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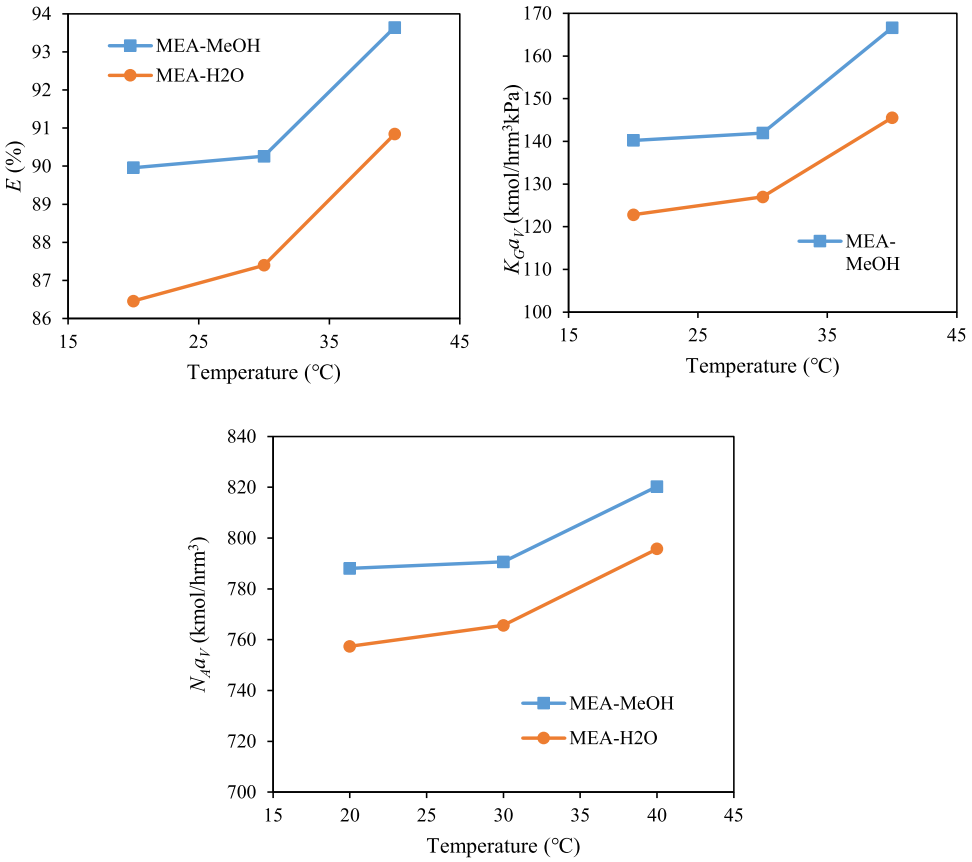

Figure 4. Effect of temperature for the aqueous and water-lean solutions of monoethanolamine at a solvent concentration of 20 wt % and liquid flow of 6 mL/min on $K_G a_v$, E, and $N_{CO_2} a_v$.

leads to a slight increase in carbon dioxide removal efficiency of about 1%. However, increasing the temperature to 40 °C increases the removal efficiency drastically and increases the removal efficiency from about 90 to 93.5%. Furthermore, the interaction of the amine concentration and fluid flow is significant. The value of F for this interaction is 9.64. Figure 6(b) shows the diagram of the interaction between the liquid concentration and liquid flow. With increasing fluid flow from 3 to 6 mL/min and then from 6 to 9 mL/min, the removal efficiency increases uniformly. This increase is about 2%, and the removal efficiency increases from about 87 to 89%. It means that at higher flow rates, the removal efficiency was much greater than in the lower levels.[27] The value of F for the interaction of liquid flow and temperature is 7.81. The calculated value of F was less than that of F for the other interactions; therefore, the least interaction is related to this state. Figure 6(c) shows that with increasing temperature in different fluid flows, the removal efficiency increases slightly and by a maximum of 1%. The interactions for the overall volumetric mass transfer coefficient and the volumetric mass transfer flux have similar behavior due to the similarity of the responses to the removal efficiency response.

4.5. Optimization. To maximize the removal efficiency and mass transfer coefficient, the optimization was done by adjusting the variables. The desirability function was utilized, which differs from 0 for fully unfavorable to 1 for completely satisfactory.[27,28] The single-response optimization results show that in the following operating conditions: concentration of 30 wt %, temperature of 40 °C, and liquid flow of 9 mL/min, the removal efficiency and mass transfer coefficient for both aqueous and water-lean systems show the highest value. Figure 7 shows a comparison between the aqueous and water-lean solutions. For the monoethanolamine-methanol solvent in these conditions, the removal efficiency is about 2.99%, the overall volume mass transfer coefficient is 18.09%, and the volumetric mass transfer flux 3.24% higher than the aqueous solvent of monoethanolamine. The higher the Henry's law constant of CO_2 in MeOH than H_2O, the higher the solubility and mass transfer performance.[24] However, methanol has a higher vapor pressure than water. Therefore, there is a higher solvent loss in the absorber due to vaporization. This issue must be considered in the designing stage by implementing proper operating strategies in the CO_2 capture plant.

4.6. Artificial Neural Network. Deep learning algorithms use layered structures called neural networks to mimic human behavior based on data analysis. The design of this layered structure is derived from the structure of the human brain. Neural networks can be trained in a manner similar to the behavior of the human brain. Neural networks can model nonlinear problems. Diverse types of neural networks differ from each other in terms of structure, data flow, number and type of neurons in layers, number of layers, and other things. In this study, multilayer perceptron (MLP) and radiation basis function (RBF) methods were employed for the CO_2 absorption process modeling. Levenberg−Marquardt (LM) training algorithms were used to develop an MLP-ANN.[29] The performance of Pure-line (PL), tangent sigmoid (TANSIG), and log sigmoid (LOGSIG) transfer functions have been assessed for the MLP method. The data are divided into two parts, training and test, 70% of the data was assigned to the

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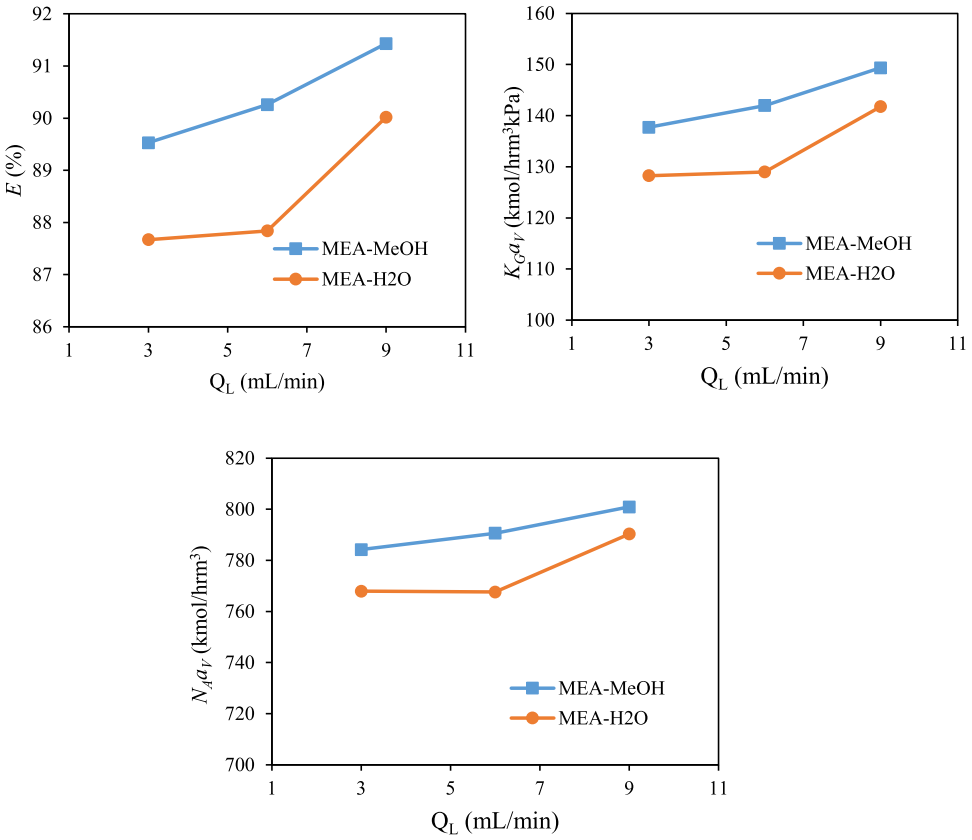

Figure 5. Effect of liquid flow for the aqueous and water-lean solutions of monoethanolamine at 30 °C and solvent concentration of 20 wt % on $K_G a_v$, E, and $N_{CO_2} a_v$.

training phase and 30% to the test phase. To evaluate the accuracy and adaptability of the model, we performed statistical tests. The parameters that were used were the root-mean-square error (RMSE), mean absolute percent error (MAPE), and the coefficient of determination (R^2). The optimal model was selected based on the highest R^2, and the lowest MAPE and RMSE.

$$\text{RMSE} = \sqrt{\frac{\sum_{i=1}^{n} (E_{\text{exp}} - E_{\text{pre}})^2}{n}} \tag{15}$$

$$\text{MAPE}\ (\%) = \frac{\sum_{i=1}^{n} \left| \frac{E_{\text{exp}} - E_{\text{pre}}}{E_{\text{exp}}} \right|}{n} \times 100 \tag{16}$$

$$R^2 = 1 - \frac{\sum_{i=1}^{n} (E_{\text{exp}} - E_{\text{pre}})^2}{\sum_{i=1}^{n} (E_{\text{exp}} - \bar{E}_{\text{exp}})^2} \tag{17}$$

where E_{exp} is the removal efficiency experimental data and E_{pre} is the ANN predicted data.

The number of hidden neurons in each structure is of great importance. If fewer neurons were selected, this leads to underfitting, whereas choosing a large number of neurons may lead to overfitting, high variance, and increase the time required for network training. As shown in Figure 8, the number of hidden neurons in the MLP with different transfer functions and RBF model was changed from 1 to 8. To determine the ideal number of neurons, the minimum RMSE was considered as the target. As shown in the figure, for MLP-PL, the number of neurons has no effect on RMSE. However, for the others, by increasing the number of neurons RMSE reduces. This reduction was sharper for the MLP model to reach minimum. In the higher number of neurons, more than minimum, the RMSE rises due to overfitting. The minimum RMSE was related to the TANSIG transfer function. Hence, the optimum number of neurons for the MLP model with TANSIG transfer function was 5 with RSME and R^2 of 0.3510 and 0.9992, respectively.

Figure 9 displays a parity plot that compares the results from the ANN model against data. A diagonal $y = x$ line is drawn as a guide to designate which predictions are in agreement with the actual values. The figure shows the close correlation between the optimized MLP-ANN model results and the experimental removal efficiency of the water-lean MEA solution for training and testing data. The results show that the network is well trained. The coefficient of determination for all data 0.9992 has been calculated. It shows that the trained network predicts removal efficiency with good accuracy. More statistical analysis of the optimized ANN model is presented in Table 5. The mean absolute percent error for all data was 0.30762. In general, the results show that the utilized MLP-ANN model predicts the CO_2 removal efficiency of water-lean amine solution to satisfactory levels.

4.7. Comparison with Other Contactors. There are several gas−liquid contactors for absorbing CO_2 from flue gas. In this research, a microchannel has been used. The microchannel made good contact between the liquid and the gas stream. Table 6 shows the results of various packed bed contactors that are usually used to separate CO_2 from the gas mixture. Removal efficiency, overall mass transfer coefficient, and volumetric mass transfer flux in the microchannel system

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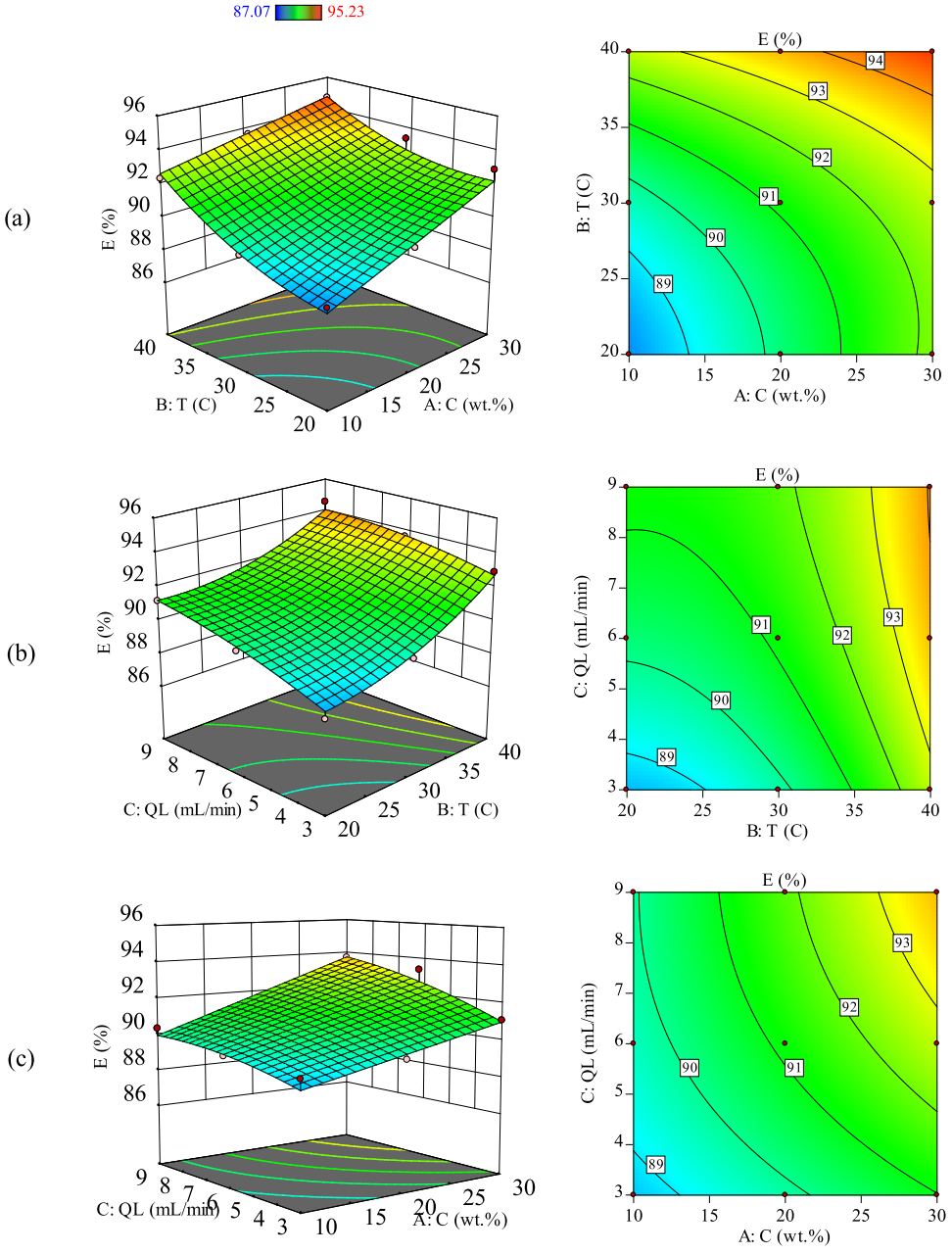

Figure 6. Interaction of operating variables for monoethanolamine-methanol: (a) Temperature−concentration, (b) temperature−liquid flow, and (c) concentration−liquid flow.

are significantly higher than those in other systems. The $K_G a_v$ values of this study are consistent with the values reported for microchannels. The resulting $K_G a_v$ is about 40 times higher than the reported values for packed towers.

Absorption in the microchannel takes place in a short time and path. Due to the good contact between liquid and gas, the rate of mass transfer and heat transfer in it is higher. The amount of absorptive consumed in the microchannel is less compared to that in other systems. It has lower operating and maintenance costs and low risk.[19]

4.8. Limitations of the Study. CO_2 absorption by the aqueous and nonaqueous solution of monoethanolamine was carried out in the microchannel with different operating conditions, including inlet fluid flow of 3−9 mL/min, solvent concentration of 10−30 wt %, and temperature of 20−40 °C. The application of obtained equations by RSM and the proposed model by ANN have been limited to the above-mentioned operating conditions.

5. CONCLUSIONS

In this study, process intensification of carbon dioxide absorption in a T-shaped microchannel was performed. The performance of two different mixtures of the aqueous and water-lean solutions of monoethanolamine in different operating conditions, including inlet fluid flow, solvent concentration, and temperature, was measured. Response surface methodology by three-level full factorial design was used to analyze and optimize the responses. In the analysis of the results, it was found that increasing the liquid concentration, increasing the temperature, and increasing the liquid flow increase the removal efficiency, the overall mass transfer coefficient, and the mass transfer flux.

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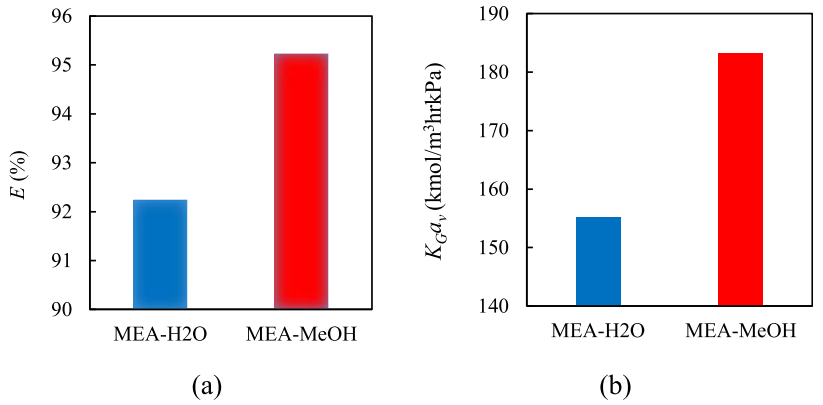

Figure 7. Comparison of maximum (a) removal efficiency and (b) mass transfer coefficient for aqueous and water-lean monoethanolamine solution at a concentration of 30 wt %, 40 °C, and liquid flow of 9 mL/min.

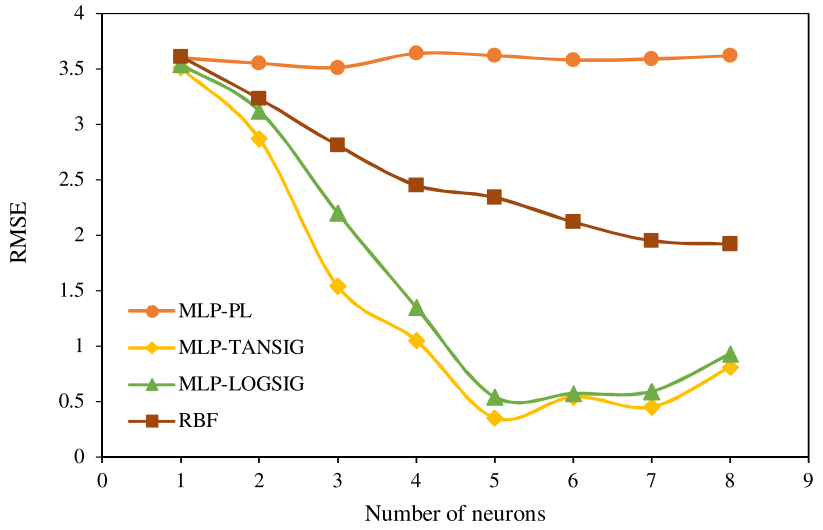

Figure 8. Effect of the number of neurons for RBF and MLP model structures with various transfer functions.

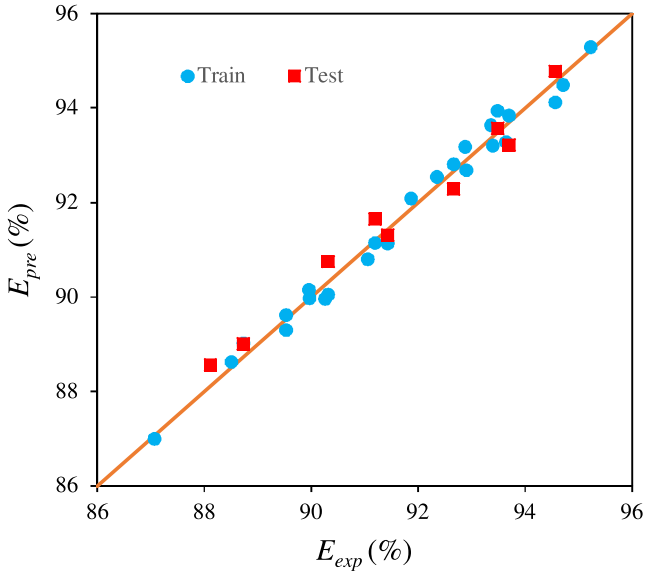

Figure 9. Parity plot of experimental and ANN predicted values for removal efficiency of the water-lean MEA solution.

The maximum removal efficiency and mass transfer coefficient have been obtained in the following operating conditions: concentration of 30 wt %, temperature of 40 °C, and liquid flow of 9 mL/min for both aqueous and water-lean systems. For monoethanolamine-methanol solvent in these conditions, the removal efficiency was about 95.23%, and the overall volume mass transfer coefficient was 18.09% higher than the $K_G a_v$ of aqueous solvent.

Table 5. Statistical Analysis of Optimized ANN Model

	train	test	all data
R^2	0.9993	0.9987	0.9992
RMSE	0.35103	0.36428	0.35877
MAPE	0.29138	0.31419	0.30762

Furthermore, the ANN method was utilized for modeling the CO_2 removal efficiency of the water-lean monoethanolamine solvent. The ANN architectures, transfer function, and number of neurons have been optimized to select the optimum ANN. The number of neurons and different transfer functions have been optimized in MLP and RBF models to select optimum ANN. The results show that the MLP model with a tangent sigmoid transfer function had the best performance with an RMSE value of 0.35103. The mean absolute percent error for all data was 0.30762. In general, the results show that the utilized MLP-ANN model predicts the CO_2 removal efficiency of water-lean amine solution to satisfactory levels.

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Table 6. Comparison of Microchannel and Packed Towers for CO_2 Capture

system type	experimental conditions	removal efficiency (%)	$N_{CO_2}a_v$ (kmol/m^3 h)	K_Ga_v (kmol/m^3 h kPa)
packed tower[24]	pressure: 100 kPa temperature: 10−12 °C solvent: methanol and MEA 15−30 wt % inlet CO_2 conc.: 6.6−8.13 vol %	41.35−92.31		3.3−0.2
packed tower[26]	pressure: 100 kPa temperature: 30−40 °C solvent: methanol and MEA wt % 30 inlet CO_2 conc.: 15 vol %	90 (fixed)		3.5−0.5
packed tower[22]	pressure: 100 kPa temperature: 25 °C solvent: solution of 5 mol MEA in methanol inlet CO_2 conc.: 6.6−8.13 vol %		14.2−48.5	6.4−3.5
packed tower[30]	pressure: 100 kPa temperature: 35−45 °C solvent: methanol and MEA wt % 15−30 inlet CO_2 conc.: 5−15 vol %	41.20−95.00	9.8−19.75	0.95−4.85
microchannel[13]	pressure: 141−401 kPa temperature: 10−30 °C solvent: ammonia 4−10 wt % inlet CO_2 conc.: 10 vol %	72.2−96.6	781.9−781.9	32.4−115.6
microchannel[14]	pressure: 1.7 bar temperature: 40−50 °C solvent: water inlet CO_2 conc.: 40−60 vol %	4.8−70.9		
microchannel in this study	pressure: 87 kPa temperature: 20−40 °C solvent: methanol solution and MEA 10−30 wt % inlet CO_2 conc.: 15 vol %	95.2−87.1	834.1−762.7	125.5−183.1

Comparison between packed tower and microchannel system showed that the removal efficiency and overall mass transfer coefficient in the microfluidic system are significantly higher than those in other systems.

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Notes

The authors declare no competing financial interest.

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