

Investigation of CO₂ Solubility in Blends of AMP and HMDA Solvents: Thermodynamic Modeling Based on the Deshmukh-Mather Model

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ABSTRACT: Among the various methods for CO₂ removal from the atmosphere, CO₂ absorption is considered as the most applicable one. Although several studies have been performed on CO₂ absorption by using chemical solvents, there are not many studies on using improved absorbents. Therefore, the aim of this study was to measure the solubility of CO₂ in a utilized aqueous blends of 2-amino-2-methyl-1-propanol (AMP) and hexamethylenediamine (HMDA). In the present research, the equilibrium set up for measuring CO₂ solubility in aqueous solvents was assembled. CO₂ solubility experiments were carried out over a various range of CO₂ partial pressures (8.44, 25.33, and 42.22 kPa) temperatures (303, 313, and 323 K) and solvent concentration (0.4, 1.8, 2, and 3 M). The results showed that increasing CO₂ partial pressure or decreasing the temperature would lead to an increase in CO₂ loading. Furthermore, with increasing HMDA concentration, CO₂ loading reached a maximum value before it followed a slight decrease. The experimental results were fitted to the thermodynamic Deshmukh Mather model and the required parameters in this model were generated. The results showed that there is a good agreement between theoretical and experimental results of CO₂ loading.

KEYWORDS: Carbon dioxide; AMP; HMDA; Deshmukh Mather Model; Correlation.

INTRODUCTION

Carbon dioxide as a leading greenhouse gas plays a crucial role in global warming. It is reckoned that the rising level of CO₂ in the atmosphere could be in charge for environmental irregularities such as extreme weather events and ocean acidification [1]. Usually, a considerable amount of carbon dioxide is produced by the plants that work based on fossil fuels and is released in the

atmosphere. Natural gas refineries, still mills, cement production industries and numerous large and small industries influence the amount of carbon dioxide in the atmosphere [2,3]. So far, different methods such as chemical absorption, physical absorption, refrigerating methods, membrane separation and biological absorption are developed to remove carbon dioxide from the air [4-6].

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Among the abovementioned methods, chemical absorption processes are the most common method to remove carbon dioxide from the air. In the chemical absorption of carbon dioxide, solvents could dissolve large amounts of carbon dioxide in comparison to other gasses. Therefore, it is easily possible to separate carbon dioxide by chemical absorption processes. Alkanolamines solvents are the most common solvents used for the removal of carbon dioxide from the air or a mixture of gases. Some common amines are monoethanolamine (MEA), diethanolamine (DEA), and methyl-diethanolamine (MDEA) [7, 8]. Recently, a new class of amines like 2-amino-2-methyl-1-propanol (AMP), hexamethylenediamine (HMDA), and piperazine (PZ) are introduced which claims to have better functioning compared to conventional solvents [9-14]. Recently, ionic liquids (ILs) have been proposed as a potential replacement solvent for amines in the carbon capture processes [15-16]. The results of recent studies show that these have the potential to remove carbon dioxide with high efficiency. Different vigorous mathematical models such as electrolyte-NRTL, UNIQUAC-NRF, LSSVM, MLP-ANN, RBF-ANN and ANFIS models were furthermore developed to predict or correlate the level of carbon dioxide removal from different gas mixtures with alkanolamines solvents [15, 17-19]. The most important models which could easily predict the CO₂ solubility data are Deshmukh-Mather [18] and Kent-Eisenberg [20-22] models. In this research, the aim was to assess the effect of influential parameters such as temperature, CO₂ partial pressure and solvent concentration on CO₂ solubility in a blend of AMP/HMDA and then correlation of Deshmukh-Mather model binary parameters based on the experimental data.

EXPERIMENTAL SECTION

Experiment Object

In this study, the impact of key factors such as temperature, CO₂ partial pressure and solvent concentration were examined on carbon dioxide removal efficiency. It is while AMP/HMDA solvents were selected in the experiment to provide us with the determinative parameter which was named CO₂ loading. It has to be mentioned that throughout this research, the term CO₂ loading (Eq. (1)) is used to refer to the mole of CO₂ per mole of solvent. In order to investigate the above-mentioned factor, an apparatus was assembled which will be fully discussed in the following sections.

$$\alpha = \frac{\text{molCO}_2}{\text{molSolvent}} \quad (1)$$

Materials

In order to carry out the experiments, AMP and HMDA solvents were purchased with purity of 95% and 99% respectively, both supplied by Merck Company and a certain amount of distilled water for preparing aqueous solution. CO₂-N₂ gas was prepared by SEPAHAN, industrial and medical gasses production, Company which is located in Isfahan. This project was undertaken to assemble an experimental set up which could facilitate CO₂ solubility process. Accordingly it was assembled which is illustrated in Fig. 1.

In order to conduct the experiment in a constant temperature, first the equilibrium cell was filled with water at the desired temperature. The gas was then allowed to enter the system to pass the spiral tube. Meanwhile the inlet gas was closed and the mercury level was brought up with the application of the mercury jack. Immediately afterwards the u-shaped tube was closed with the usage of a drop of water. Consequently the stream of gas was trapped in the system before the absorption process takes place. Later on, the valve of the solvent container was opened which injected a certain amount of solvent into the system. As the solvent flowed down the spiral tube, it came into contact with the gas available and finally the absorption process was initiated. During the process, the solvent was saturated with this gas which caused a gas pressure drop. This pressure variation was compensated with an elevation in the level of the mercury of burette. As a result, first the amount of gas which was dissolved into the solvent was obtained based on the height difference of mercury in the burette. Secondly the height of mercury in the burette converted to the dissolved CO₂ mole by using ideal equation of state. Thirdly CO₂ loading was calculated according to Eq. 1. The experiments were all carried out in different range of temperature, concentration and partial pressure and the results were all registered. It should be noted that each experiment was repeated three times and the averaged data was used to reduce the experimental errors. The error bars of all experimental data obtained from the statistical analysis have been shown in Fig. 2.

THEORETICAL SECTION

Correlation of experimental data with the Deshmukh mather model

In this study, the Deshmukh Mather model was used to investigate CO₂ loading. The gas pressure was equal

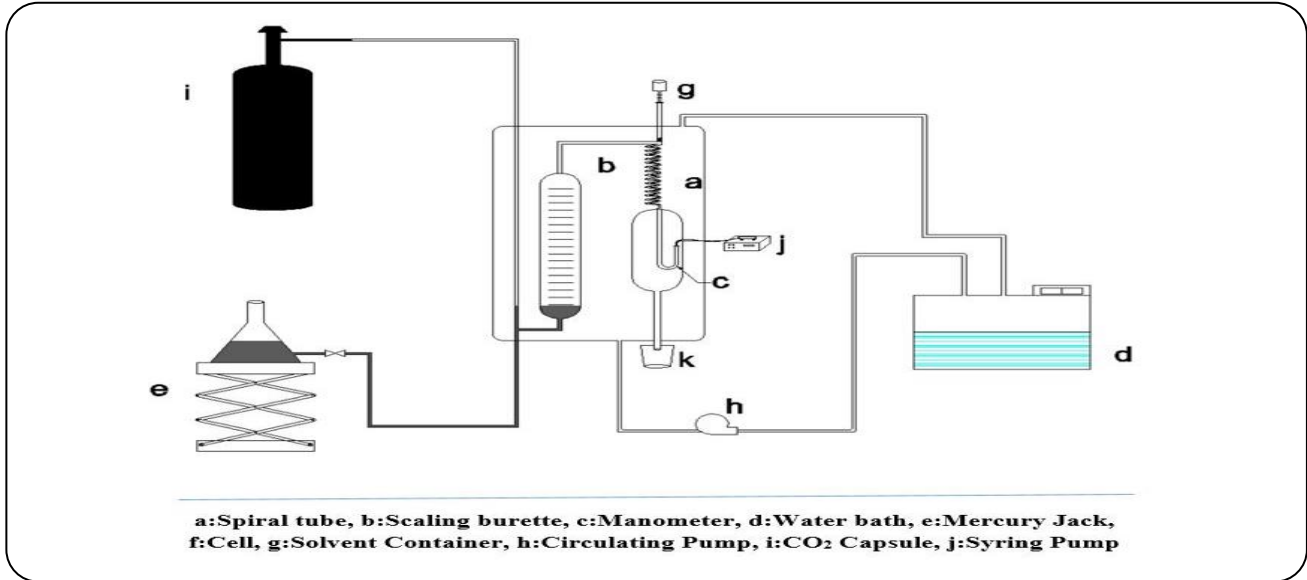


Fig. 1 set up for measuring the solubility of gases in liquid.

to the barometric pressure of about 0.82 atm. In the Deshmukh-Mather model, the activity coefficient of the solute species (Eq. 2) is calculated by the Guggenheim theory [23]. It has to be noted that, there is no assumption taken into account based on the ideality of the liquid phase. The equations of the model are explained as follows:

$$\ln \gamma_i = \frac{AZ_i^2 \sqrt{I}}{1 + B\sqrt{I}} + 2 \sum \beta_{i,j} m_j \quad (2)$$

$$\beta_{ij} = a_{ij} + b_{ij}T + c_{ij}T^2$$

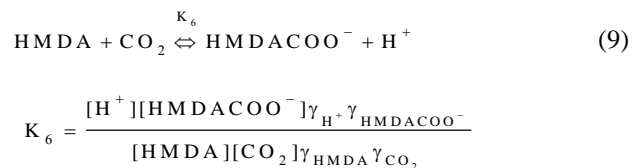
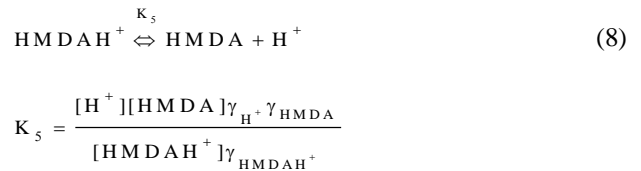
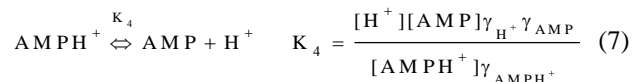
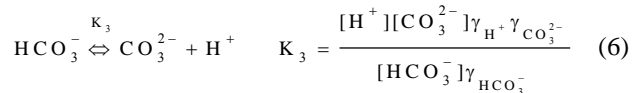
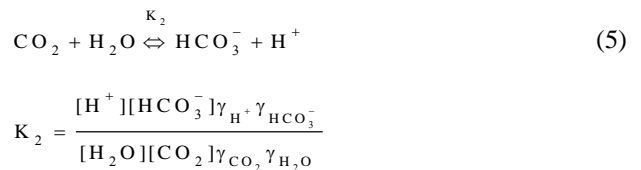
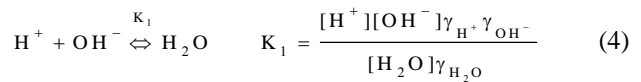
$$I = \frac{1}{2} \sum m_j Z_j^2$$

In Eq. 2, Z_j and m_j refer to the electrical charges and concentrations of the relevant species, respectively. B as a function of temperature and dielectric constant of the solvent is proposed by Pitzer and equals to 1.2. A as a temperature-dependent parameter is calculated according to Eq. (3) [24].

$$A = 1.306548 + \frac{0.1328238 \times 10^{-1}}{T} - \frac{0.3550803 \times 10^{-4}}{T^2} + \frac{0.3381968 \times 10^{-7}}{T^3} \quad (3)$$

Reaction scheme

An equilibrium solution of CO₂ in the equilibrium aqueous system of AMP+HMDA+H₂O was governed by the following set of equations:



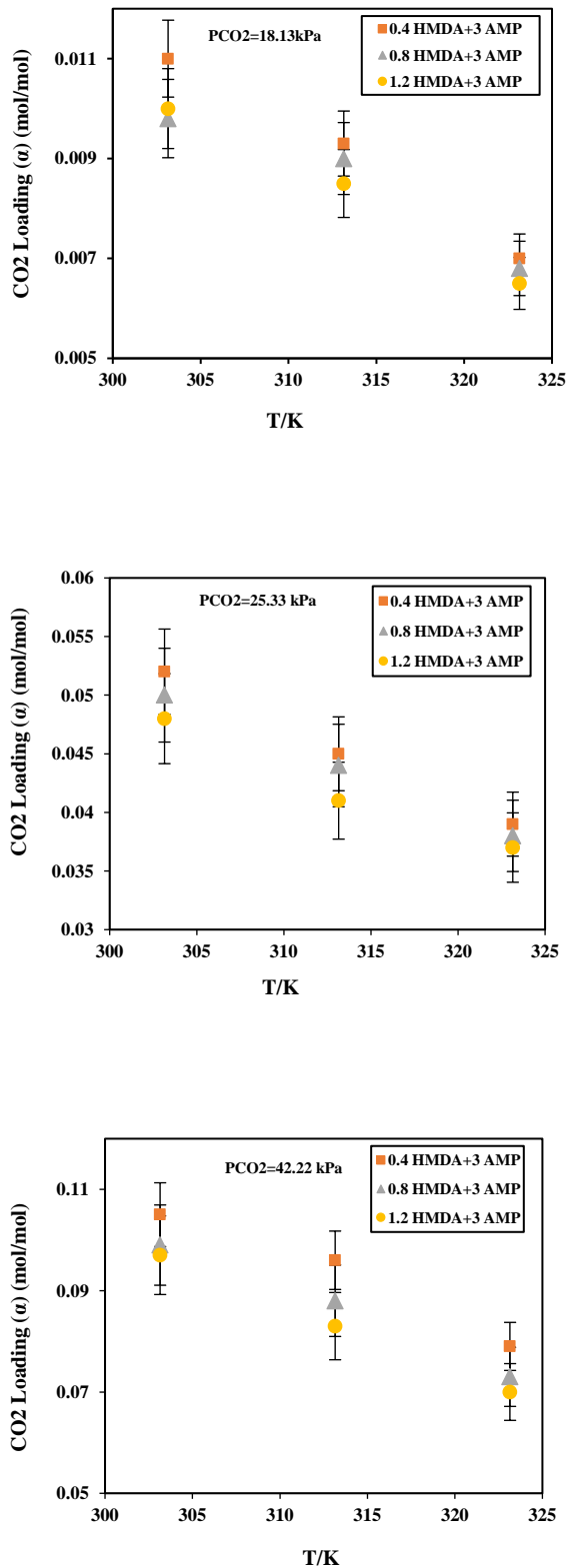


Fig. 2: The average error bars of experimental data obtained from statistical analysis.



$$K_7 = \frac{[\text{H}^+][\text{HMDA}(\text{COO}^-)_2]\gamma_{\text{H}^+}\gamma_{\text{HMDA}(\text{COO}^-)_2}}{[\text{HMDACOO}^-][\text{CO}_2]\gamma_{\text{HMDACOO}^-}\gamma_{\text{CO}_2}}$$



$$K_8 = \frac{[\text{H}^+][\text{HMDACOO}^-]\gamma_{\text{H}^+}\gamma_{\text{HMDACOO}^-}}{[\text{HMDACOOH}]\gamma_{\text{HMDACOOH}}}$$

It has to be noticed that, the carbamate ion formation reaction is ignored in the presence of AMP among the above reactions. It is for the reason that, AMP-Carbamate ion has a low concentration in the mixture of AMP and HMDA, which could be omitted as a result [25]. In addition to the previous equations, the following set of conditions must also be satisfied:

Charge balance:

$$[\text{H}^+] + [\text{AMPH}^+] + [\text{HMDAH}^+] = [\text{HCO}_3^-] + \quad (12)$$

$$[\text{OH}^-] + 2[\text{CO}_3^{2-}] + [\text{HMDACOO}^-] + 2[\text{HMDA}(\text{COO}^-)_2]$$

CO₂ mass balance:

$$\alpha([\text{AMP}]_i + [\text{HMDA}]_i) = [\text{CO}_2] + \quad (13)$$

$$[\text{HCO}_3^-] + [\text{CO}_3^{2-}] + [\text{HMDACOO}^-] + [\text{HMDACOOH}] + [\text{HMDA}(\text{COO}^-)_2]$$

Amine mass balance:

$$[\text{AMP}]_i = [\text{AMP}] + [\text{AMPH}^+] \quad (14)$$

HMDA mass balance:

$$[\text{HMDA}]_i = [\text{HMDA}] + [\text{HMDAH}^+] + \quad (15)$$

$$[\text{HMDACOO}^-] + [\text{HMDACOOH}] + [\text{HMDA}(\text{COO}^-)_2]$$

The concentration of carbon dioxide in the liquid phase can be estimated from Henry's law:

$$P_{\text{CO}_2} = H_{\text{CO}_2}[\text{CO}_2] \quad (16)$$

The dependency of the equilibrium constant, K_i , as well as the Henry's constant, H , with temperature was expressed as:

Table 1: The amounts for equilibrium constant used in Eq.(17) [11].

Parameter	a _i	b _i	c _i	d _i	T(K)
K ₁	-13445.9	-22.4773	0	132.899	273-498
vK	-12092.1	-36.7816	0	231.465	273-498
rK	-12431.7	-35.4819	0	216.05	273-498
+K	-2556.2	0	0	11.555	273-498
sK	-7051.0	0	0	-5.537	303-333
vK	4746.82	0	0	-7.16	303-333
vK	13986.36	0	0	-19.035	303-333
sK	-7986.0	0	0	-1.847	303-333
H _{CO2}	-8477.7-	-21.957	0.005781	163.80	273-333

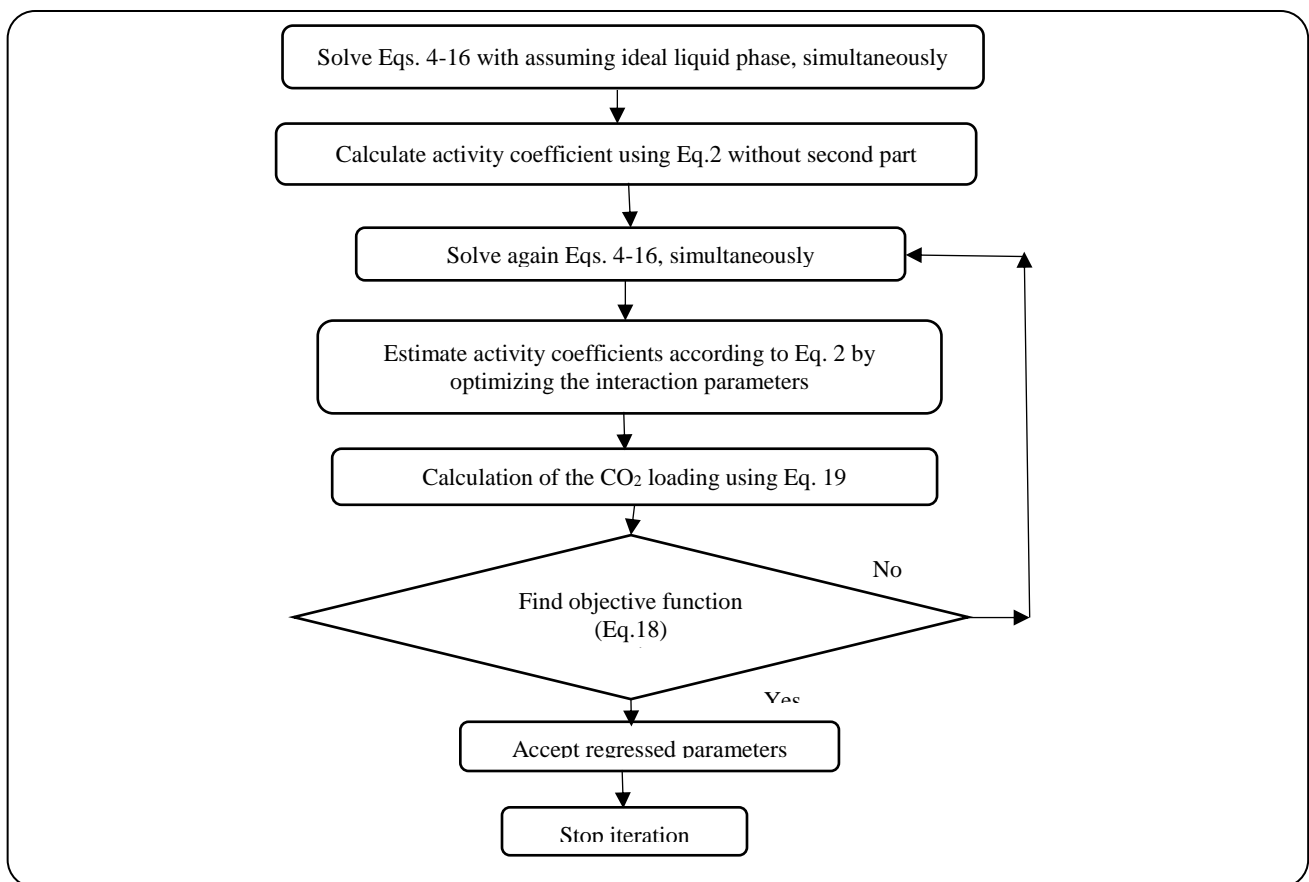
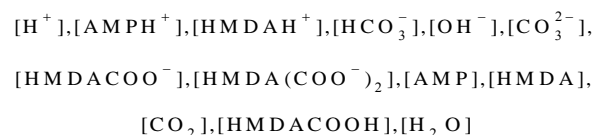


Fig. 3: Flowchart of activity coefficient parameters calculation in Deshmakh Mather model.

$$\ln K_i \text{ (or } H_{CO_2}) = \frac{a_i}{T} + b_i \ln(T) + c_i T + d_i \quad (17)$$

Where a_i–d_i were constants. Values of these constants were taken from the literature [9, 12, 26], as given in Table 1.

The unknowns which exist in the Equation are expressed below [10]:



The following algorithm (Fig. 3) in MATLAB software which has been employed in the present research provides a method to calculate the species activity

coefficients. It has also been used to determine the amounts for the binary interaction parameters in an aqueous system of AMP-HMDA-H₂O-CO₂.

The set of nonlinear algebraic equations to determine the concentration components and model parameters was obtained simultaneously by employing a few defined algorithms and functions such as 'Fsolve' and 'Fmincon' simultaneously. In the MATLAB software, 'Fsolve' and 'Fmincon' functions were used to solve the nonlinear equations and optimize the model parameters, respectively. During the optimization procedure, our objective function was defined in a way to minimize the average absolute deviation percentage which was expressed as Eq. (18).

$$RMSE = \sqrt{\frac{\sum_{i=1}^n (\alpha_{(calc)_i} - \alpha_{(exp)_i})^2}{n}} \times 100 \quad (18)$$

RESULTS AND DISCUSSION

The experiment of CO₂ solubility was conducted in aqueous AMP/HMDA solution in different operational conditions. The object was to determine the amounts of CO₂ loading. The experiments results were used for determination of Deshmukh-Mather model parameters. In the following, the experimental results were illustrated according to various operational conditions and the effects of these parameters on CO₂ loading were investigated. Fig. 4 displays the effect of temperature on CO₂ loading.

It is apparent that there has a decrease in CO₂ loading as temperature rises. Since the experiments are exothermic, this decreasing trend seems logical. Also, this phenomenon is due to the higher vapor pressure that results from the higher kinetic energy of the molecules in the liquid at high temperatures.

On the other hand, increasing the partial pressure of CO₂ resulted in higher CO₂ loading. In higher partial pressure of CO₂ over (AMP+HMDA) system, HMDA forms many carbamate species upon reaction with CO₂ in an aqueous medium. The presence of the HMDA carbamate protonated HMDA carbamate, and HMDA carbamate may increase the overall stability of the carbamate species, resulting in more amine being tied up for every mole of CO₂ absorbed. The highest value of CO₂ loading was obtained at the highest partial pressure (i.e. 42.22 kPa) and lowest temperature (i.e. 303K). Fig. 5 shows the influence

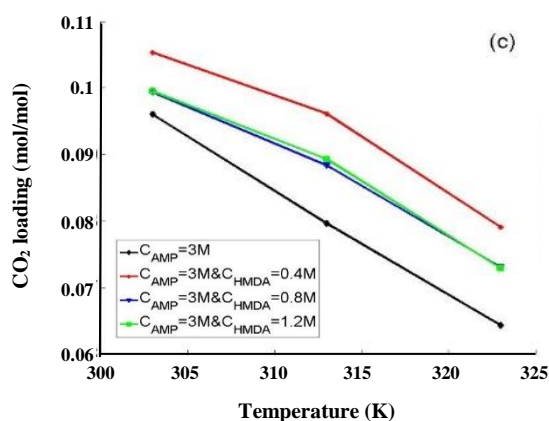
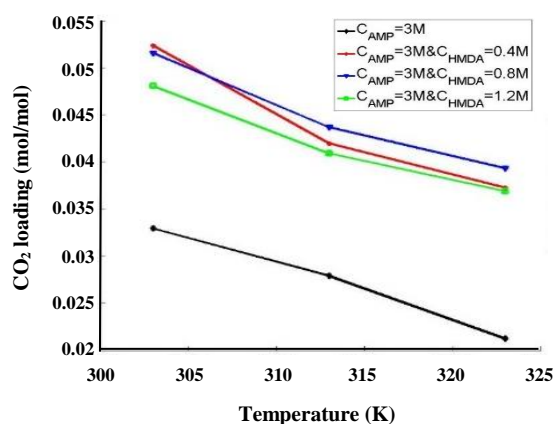
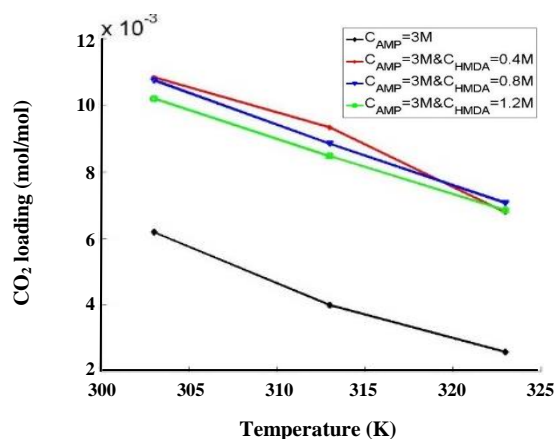


Fig. 4: The effect of temperature on CO₂ loading in different concentrations of AMP+HMDA. a: P_{co2}=8.44kPa, b: P_{co2}=25.33kPa, c: P_{co2}=42.22kPa.

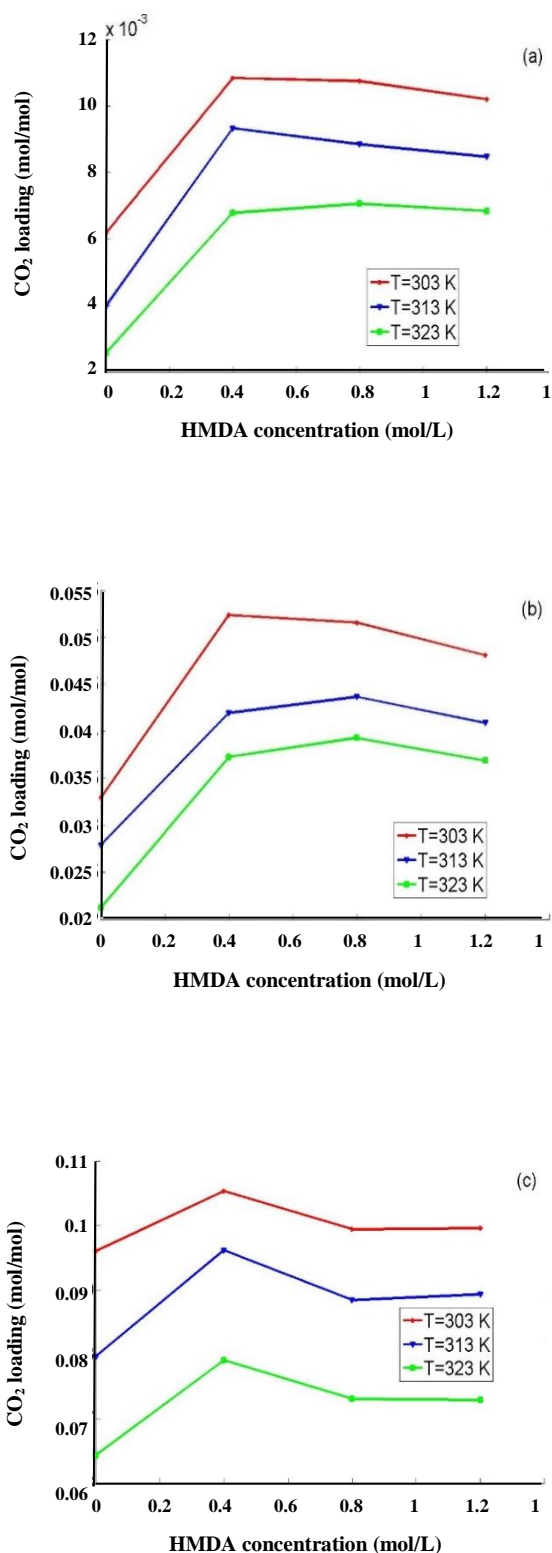


Fig. 5: The effect of adding HMDA to AMP 3M on CO₂ loading
 a: P_{CO2}=8.44 kPa, b: P_{CO2}=25.33kPa, c: P_{CO2}=42.22kPa.

of adding HMDA to AMP 3M on the CO₂ loading. In Fig.5 CO₂ loading decreases with raising the solvent concentration which seems irrational.

It has been found that the addition of small amounts of PZ to an aqueous solution of AMP significantly enhances the rate of absorption of CO₂. The solvent capacity has been found to increase with total amine in the CO₂ partial pressure range of 8-43 kPa, which is the typical condition for CO₂ partial pressure in the power plant flue gas streams, indicating improved CO₂ capacity of blends of AMP and PZ solvents. These results were in agreement with Dash et al. research [27].

On the other hand, according to Eq. 1, the increase in the denominator (solvent moles) is much more than the numerator (CO₂ moles) during the experiment. For this reason, there has been a decrease in CO₂ loading with concentration increase. This enhancement has been also seen in the previous works [2, 25, 8] on AMP/HMDA mixtures which assert the accuracy of the matter.

On the basis of the calculated activity coefficients with the binary parameters, CO₂ loading was calculated using Eq. (19).

$$\alpha_{calc} = \frac{[CO_2] + [HCO_3^-] + [CO_3^{2-}]}{[AMP] + [HMDA]} \quad (19)$$

The experimental and calculated data of the CO₂ loading for different concentrations of HMDA, as well as the corresponding AAD% were presented in Table 2.

The contradictory behavior of the HMDA at low and high CO₂ partial pressure may explain why neither of the Deshmukh-Mather models can provide an accurate estimation for the entire range of partial pressures. It seems that the Deshmukh-Mather is capable of capturing the positive role of the HMDA in the mixture.

In particular, the corresponding optimized parameters used in the Kent-Eisenberg and Deshmukh Mather models were provided in Table 3. It should be noted here that the initial guess in our Deshmukh-Mather model was obtained from the results of Tong et al. and Jahangir and Hasankiade [10, 28].

CONCLUSIONS

This study was divided into two sections. In the first section solubility of CO₂ was investigated by using an AMP-HMDA mixture at different operational conditions including partial pressure, solvent concentrations and

Table 2: Amounts of experimental and calculated CO₂ loading in a blend of AMP+HMDA+H₂O.

Concentration mol/L	Temp.K	P _{CO₂} = 8.44 kPa			P _{CO₂} = 25.33 kPa			P _{CO₂} = 42.22 kPa		
		α_{Exp}	α_{Calc}	ϵ_{RMSE}	α_{Exp}	α_{Calc}	ϵ_{RMSE}	α_{Exp}	α_{Calc}	ϵ_{RMSE}
3AMP + 0.4 ← HMDA	303	0.011	0.014	0.28	0.052	0.041	1.16	0.105	0.068	3.74
3AMP + 0.4 ← HMDA	313	0.009	0.011	0.14	0.042	0.032	0.98	0.96	0.053	4.26
3AMP + 0.4 ← HMDA	323	0.007	0.009	0.19	0.037	0.026	1.11	0.79	0.044	3.55
3AMP + 0.8 ← HMDA	303	0.001	0.012	0.14	0.052	0.037	1.51	0.099	0.061	3.86
3AMP + 0.8 ← HMDA	313	0.009	0.010	0.07	0.044	0.029	1.49	0.088	0.048	4.05
3AMP + 0.8 ← HMDA	323	0.007	0.008	0.07	0.039	0.023	1.59	0.073	0.039	3.41
3AMP + 1.2 ← HMDA	303	0.010	0.011	0.08	0.048	0.033	1.51	0.099	0.055	4.45
3AMP + 1.2 ← HMDA	313	0.009	0.009	0.02	0.041	0.026	1.49	0.089	0.043	4.60
3AMP + 1.2 ← HMDA	323	0.007	0.007	0.02	0.037	0.021	1.57	0.073	0.035	3.76

Table 3: Values for binary interaction parameters of AMP-HMDA-H₂O-CO₂ system.

Parameters	a _{ij}	b _{ij}	c _{ij}
HMDACOO ⁻ -HMDAH	823.9100	199.9000	0.8630
AMPH ⁺ -CO ₂	234.9300	-654.2300	0.0190
AMPH ⁺ -HMDA	753.2100	-64.4800	0.2180
AMPH ⁺ -AMP	985.9000	-256.6300	0.1180
AMPH ⁺ -HCO ₃ ⁻	8.5783×10 ³	-87.8700	0.1220
AMP-HCO ₃ ⁻	1.499×10 ³	-29.6900	0.0210
AMP-HMDAH ⁺	1.499×10 ³	-39.6900	-0.0360
CO ₂ -HMDAH ⁺	5.3600	-98.4670	-0.1850
HMDA-HCO ₃ ⁻	27.5100	-54.0600	0.0790
HMDAH ⁺ -HCO ₃ ⁻	137.5300	-36.9300	0.2320
HMDAH ⁺ -HMDA	164.7200	-17.4000	-0.0030
AMPH ⁺ -CO ₃ ²⁻	1.9808×10 ³	-6.8900	0.0200
AMP-H ⁺	73.3000	-102.4000	0.2900
HMDAH ⁺ -CO ₃ ²⁻	122.3800	-10.5400	1.0000×10 ³
HMDA-CO ₃ ²⁻	147.7200	-120.4000	0.3820
HMDACOO ⁻ -HMDA	1.4999×10 ³	73.1000	0.1400
HMDACOO ⁻ -AMPH	128.5900	-286.7200	1.2340

temperature. CO₂ loading in AMP/HMDA solution increases with a decrease system temperature and an increase in CO₂ partial pressure. CO₂ loading followed a slight decrease after it reached the maximum value with increasing HMDA concentration. In the second section, all the above-mentioned conditions were used to the correlation of the Deshmukh-Mather model parameters. Then the results of both sections were compared with each other. It was remarkably found that the model results for CO₂ loading were in a good agreement with the experimental results. Also, it should be noted that the results of the present research could be employed for prediction of CO₂ solubility in different operating conditions an industrial applications. However, further studies on the current topic were therefore recommended with more focus on finding solvents with promising characteristics.

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