



Removal of CO₂ from Gas Mixture by Aqueous Blends of Monoethanolamine + Piperazine and Thermodynamic Analysis Using the Improved Kent Eisenberg Model

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Received: 02/08/2018

Accepted: 10/02/2019

Published: 30/03/2019

Abstract

In this study, a system was designed to determine CO₂ solubility in alkanolamine solvents. In what follows, the vapor-liquid equilibrium data of CO₂ solubility in the 12 wt% MEA aqueous solution and PZ + MEA mixture with various concentrations (1+12wt%, 2+12wt%, and 3+12wt%), at different partial pressure of CO₂ (8.44, 25.33, and 42.22 kPa) and various temperatures (303, 313 and 323 K) at a total pressure of 0.83 atm were tested. Also, the variation of CO₂ partial pressure, system temperature, and the effect of adding PZ to MEA solvent were studied. The experimental results showed that by increasing the concentration of PZ and CO₂ partial pressure, CO₂ loading increases and the temperature rise leads to a decrease in CO₂ loading. The values obtained for CO₂ loading in these experiments were in the range of 0.005-0.216 (mol_{CO2}/mol_{Amine}), and the maximum obtained value was for 3wt% of PZ + 12wt% of MEA at 303 K. Finally, the improved Kent-Eisenberg model parameters were fitted by using the MATLAB software and the experimental conditions of this study. Average absolute deviations percentage between the calculated and experimental loading was 24.4%, which indicates that the improved Kent-Eisenberg model is in good agreement with experimental data.

Keywords: CO₂ solubility, MEA aqueous solution, Piperazine additive, improved Kent-Eisenberg thermodynamic model

1 Introduction

Nowadays, one of the biggest environmental issues in the world is the increase in greenhouse gas emissions, and carbon dioxide gas is the most important greenhouse gas due to its highest atmospheric lifetime and having the highest concentration in the atmosphere. This gas traps more energy and heat in the atmosphere which results in severe climate change. Power plants, refineries, and steel and cement industries are the largest CO₂ producers in the world (1). So far, several methods have been proposed for carbon dioxide removal by various researchers. These methods include physical absorption, absorption by chemical solvents, membrane separation and several other processes. The process of gas absorption by chemical reaction of liquid solvents has been widely used in the treatment of acidic gases and gas purification. Among the methods of chemical absorption, the capture of carbon dioxide by alkanolamine solutions can be mentioned which has been studied widely over past decades. Alkanolamines consist of at least one hydroxyl group (OH⁻) and one amine group (RNH₂). The hydroxyl group

increases the solubility of amine in water and reduces the vapor pressure of the solution, while the amine group increases the alkali property of solution (2). Based on the number of radical groups bonded to the nitrogen atom, the amines are divided into three categories:

1. Primary amines such as MEA (Monoethanolamine) and DGA (Diglycolamine).
2. Secondary amines such as DEA (Diethanolamine), DIPA (Diisopropanolamine).
3. Tertiary amines such as MDEA (Methyldiethanolamine) and TEA (Triethanolamine).

In the last few decades, the MEA solvent has been extensively used to absorb CO₂ from natural gases. The advantages of this solvent include low molecular weight, high CO₂ absorption rate, and a very low tendency to absorb hydrocarbons. The major problem with MEA is its limited loading capacity due to the production of carbamate ion and its high energy-consuming recovery (3). Today, organic diamines such as PZ (Piperazine), AEEA (Aminoethylethanolamine), and HMDA are used as additives to alkanolamine because of their high absorption capacity and rapid reaction with CO₂. PZ is the

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most widely known diamine that is used in combination with other amines as an activator to increase the reaction speed. PZ is an organic compound composed of a six-membered ring containing two nitrogen atoms at opposite positions (4). Table 1 presents a summary of the studies conducted over a wide range of temperatures, pressures, and concentrations for blend of MEA and PZ aqueous solutions. To design gas purification systems, it is required to obtain vapor-liquid equilibrium data (VLE) of aqueous alkaline amine. Therefore, providing a thermodynamic model, which can predict the equilibrium behavior of these systems, is advantageous from the operational and economic point of view. So far, extensive studies have been carried out on the thermodynamic models for electrolyte solutions that can be used in gas sweetening industries. Kent and Eisenberg (5) by assuming that the liquid phase was ideal, calculated the solubility of acid gases in amines using a temperature-dependent function for the chemical equilibrium constants. Deshmukh and Mather (6) obtained the solubility of gases in amine systems by applying the Guggenheim equation and the method of Edwards et al. (7) by using activity coefficients. Austgen et al. (8) with the help of the Electrolyte-NRTL Model (9), achieved the energy parameters of interactions among the components of amine systems. Li and Mather (10) by using Clegg-Pitzer model (11) and Kaewsichan et al. (12) by utilizing the Electrolyte-UNIQUAC model predicted the solubility of acid gases in alkanolamine solutions. In the Kent-Eisenberg model, the equilibrium constant for amine reactions (carbamate and protonation reaction) is only dependent on the temperature. However, in Jou et al. (13), the equilibrium constant of amine reaction, in addition to temperature was influenced by the

loading and amine concentration. Hu and Chakma (14) presented a modified model based on the Kent-Eisenberg model for the equilibrium constant of main amine reactions as a function of temperature, partial pressure of the acid gas and amine concentration. In a similar manner, Li and Shen (15), for calculating the CO₂ solubility in a combined system (H₂O+MEA+MDEA), considered the chemical equilibrium constant as a function of the temperature, amine concentration, and CO₂ loading. In this research, the improved Kent-Eisenberg model has been used to predict the CO₂ solubility in PZ and MEA aqueous solutions. In this model, the activity coefficient is not applied in equations to correct the non-ideality of the system and components. Instead, the non-ideality of the species will be considered in the optimization of equilibrium constants coefficients.

2 Experimental

2.1 Materials

Materials used in this work are MEA (>99.5% pure) and anhydrous PZ (>99% pure). These chemicals were purchased from Merck Company. Carbon dioxide (>99.9% pure) and nitrogen (>99.9% pure) were procured from ISFAHAN GAS company. Distilled water was used for preparation of all the solutions.

2.2 Experimental apparatus and methods

Fig. 1 shows the laboratory system used in this work. This system was used by Jahangiri et al. (16) to determine the solubility of CO₂. The procedure is summarized in several steps:

Table 1: Summary of experimental data for the MEA, PZ and blend of MEA–PZ–H₂O–CO₂ system.

Source (author)	Conc. [MEA] (M)	Temp. (K)	Loading(mol CO ₂ /mol Amine)
Dang and Rochelle (17)	2.5 and 5	313 and 333	0.91-0.614
Aboudheir et al. (18)	3-9	293-333	0.1-0.5
Puxty et al. (19)	5	313-333	0-0.5
Aronu et al. (20)	2.5-10	313-393	0.017 - 0.864
Xu and Rochelle (21)	6.82-6.97	373-443	0.303-0.52
Dugas and Rochelle (22)	7-13	313-373	0.2-0.5
Luo et al. (23)	1-5	293-343	0-0.4
Conc. [PZ] (M)			
Bishnoi and Rochelle (24)	0.2-0.6	298-343	0-1.0
Ermatchkov et al. (25)	0.75-3	283-393	0.05-0.95
Aroua and Salleh (26)	0.1-1	293-323	0-0.8
Derks et al. (27)	0.2-0.6	298-333	0.3-1.25
Kamps et al. (28)	1.7-3	313-393	0-0.75
Xu and Rochelle (21)	4.93-8	354-465	0.224-0.451
Dugas and Rochelle (22)	2-12	313-373	0.226-0.411
Conc. [MEA+PZ] (M)			
Dang and Rochelle (17)	0.4+0.6	333	0.06-0.14
	1.9+0.6	333	0.01 - 0.44
	3.8+1.2	313 and 333	0.41- 0.43
Dugas and Rochelle (22)	7+2	313-373	0.242-0.477

- 1- Setting the system to the desired temperature by water bath
- 2- Fill the container with the solvent and place it on the spiral
3. Open the gas valve until the system is full of the gas.
- 4- Close the gas valve and elevate the mercury container by the jack and block the outlet of gas by the water syringe.
- 5- At the start of the absorption, the gas is pressurized in the burette. During this time, the dissolved gas in the

solvent reduces the pressure of system which will be compensated by elevating mercury container in order to perform the test under constant pressure. If the level of fluid in the manometer stays constant for a while, the test is completed, and the volume of dissolved gas is read from the burette. To determine the number of dissolved moles, we can use ideal gas law Eq.1 since the total pressure is almost 1 atm.

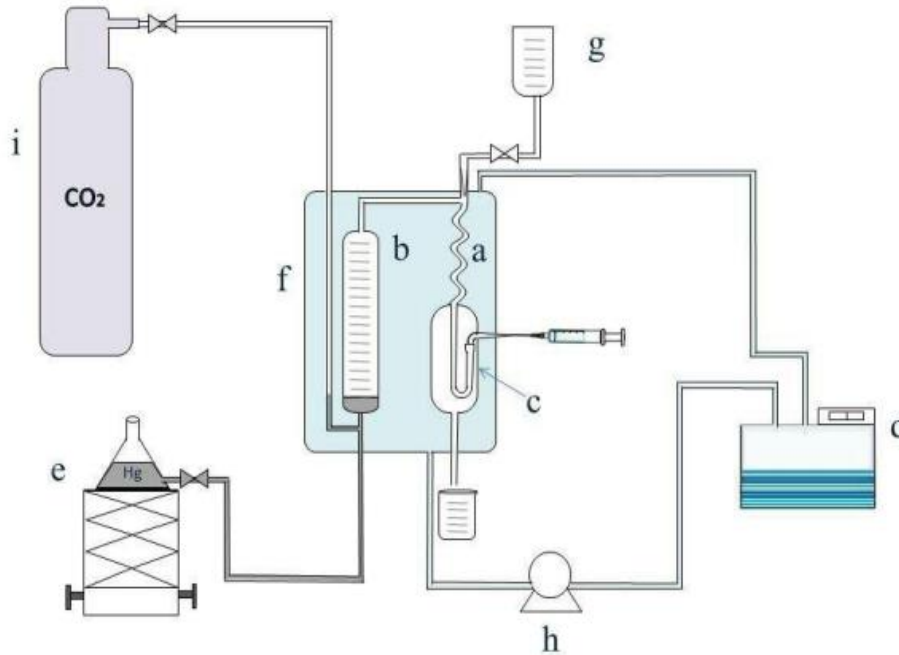


Figure 1: Vapor-liquid equilibrium apparatus for atmospheric pressure (a: Spiral tube, b: Scaling burette, c: Manometer, d: Water bath, e: Mercury jack, f: Cell, g: Solvent Container, h: Circulating Pump, i: CO₂ Capsule)

$$Pv = RT \quad (1)$$

To obtain the moles of solvent consumed, Eq. 2 could be applied.

$$n = \frac{Vd}{M_w} \quad (2)$$

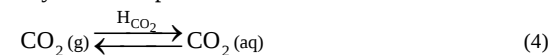
where V is the volume, d is density, and M_w is the molecular mass. Given the moles of CO₂ and the solvent, we are able to obtain the loading values required to determine the solubility of CO₂ in PZ and MEA solutions by using Eq.3.

$$\alpha_{CO_2} = \frac{n_{CO_2}}{n_{MEA} + n_{PZ}} \quad (3)$$

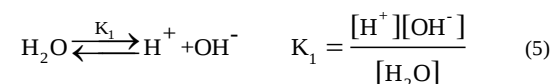
3 Modeling of equilibrium data

The equations describing the main reactions occurring in the MEA-PZ-H₂O-CO₂ system are assumed as follows:

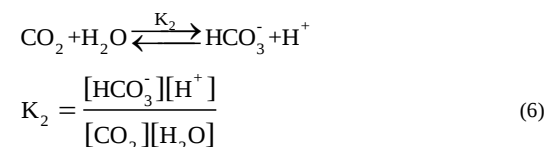
Physical absorption of CO₂:



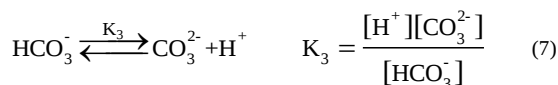
Ionization of water:



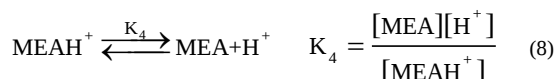
Formation of carbon dioxide:



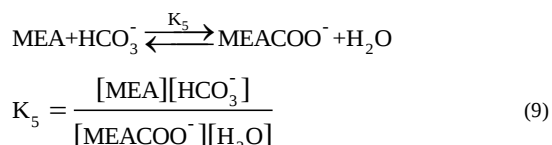
Formation of bicarbonate:



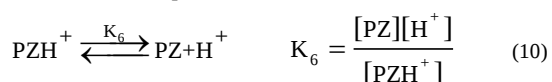
Reversion of the protonation of MEA:



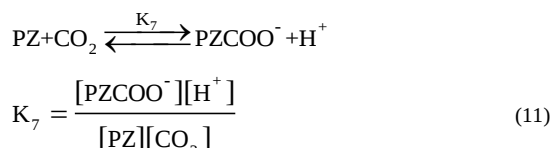
Formation of MEA carbamate:



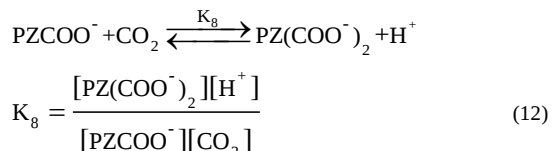
Reversion of the protonation of PZ:



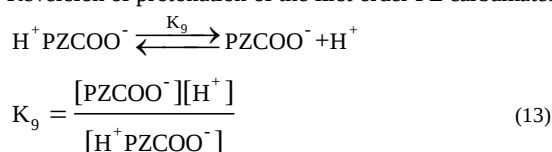
Formation of first order PZ carbamate:



Formation of second order PZ carbamate:



Reversion of protonation of the first order PZ carbamate:



Mass balances and charge balance equations can be written as follows:

Charge balance:

$$[\text{MEA}^+] + [\text{PZH}^+] + [\text{H}^+] = [\text{HCO}_3^-] + 2[\text{CO}_3^{2-}] + [\text{PZCOO}^-] + [\text{MEACOO}^-] + [\text{OH}^-] + 2[\text{PZ(COO}^-)_2] \quad (14)$$

CO₂ balance:

$$\alpha[\text{MEA} + \text{PZ}]_{\text{total}} = [\text{CO}_2] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}] + [\text{PZ(COO}^-)_2] + [\text{PZCOO}^-] + [\text{H}^+ \text{PZCOO}^-] + [\text{MEACOO}^-] \quad (15)$$

MEA balance:

$$[\text{MEA}]_t = [\text{MEA}] + [\text{MEA}^+] + [\text{MEACOO}^-] \quad (16)$$

PZ balance:

$$[\text{PZ}]_t = [\text{PZ}] + [\text{PZH}^+] + [\text{PZCOO}^-] + [\text{H}^+ \text{PZCOO}^-] + [\text{PZ(COO}^-)_2] \quad (17)$$

And according to Henry's law, the vapor phase equilibrium can be expressed as:

$$P_{\text{CO}_2} = H_{\text{CO}_2} \cdot [\text{CO}_2] \quad (18)$$

where P_{CO_2} is the partial pressure of CO₂ and H_{CO_2} is the Henry's law constant for CO₂ in solution.

The chemical equilibrium constant is a function of temperature and is obtained by Eq. 19 where A, B, C, and D parameters for each of the reactions are specified in Table 2.

$$K_i \text{ or } H_{\text{CO}_2} = \exp\left(\frac{A}{T}\right) + B \ln T + CT + D \quad (19)$$

Table 2 Coefficients of the reaction equilibrium constants used in this work

K_i	A	B	C	D	Ref.
K_1	-13446	-22.4	0	140.93	(29)
K_2	-12092	-36.7	0	235.48	(29)
K_3	-12431	-35.4	0	220.06	(29)
K_4	-17.3	0	0.0576	-38.84	(29)
K_5	-1545.3	0	0	2.151	(29)
K_6	3814.4	0	-1.5016	14.119	(30)
K_7	3616	0	0	-8.635	(30)
K_8	1322.1	0	0	-3.654	(30)
K_9	-6066.9	-2.29	0.0036	6.822	(30)
H_{CO_2}	-9624.4	-28.7	0.01441	192.8	(30)

Li and Shen (15) showed the final form of equilibrium constants as follows:

$$F = \exp(a_1 + a_2/T(K) + a_3/T^3(K) + b_1 \alpha_{\text{CO}_2} + b_2/\alpha_{\text{CO}_2} + b_3/\alpha_{\text{CO}_2}^2 + b_4 \ln[m]) \quad (20)$$

Where a_i and b_i are the adjustable parameters which are obtained by using experimental data.

In this study, we assume the equilibrium constants as the function of temperature, the partial pressure of CO₂, and the amine concentration. Adjustable equilibrium constants are related to the K_i values calculated from Eq.19, the parameters of Table 2, and F parameter. These constants are as follows:

$$K'_7 = K_7 F \quad (21)$$

$$K'_9 = K_9 F \quad (22)$$

$$F = \exp(a_1 + a_2/T(K) + a_3/T^3(K) + b_1 \ln P_{\text{CO}_2}(\text{kPa}) + b_2(P_{\text{CO}_2}(\text{kPa})) + c_1[\text{MEA}] + c_2[\text{PZ}]) \quad (23)$$

In Eq. 23, a_i , b_i , and c_i are the adjustable parameters derived from experimental data for a four-component system (MEA-PZ-H₂O-CO₂). However, in this method, only the values of K'_7 and K'_9 parameters are modified by using optimization of the proposed parameters, and for other reactions, the initial equilibrium constants are used. The absolute deviations obtained from the difference between the calculated molar load and the molar load obtained from the experimental results by using Eq.24, and the molar load is calculated using Eq. 25.

$$AD\% = \frac{|\alpha_{\text{calc}} - \alpha_{\text{exp}}|}{\alpha_{\text{exp}}} \times 100 \quad (24)$$

The algorithm used for modeling is shown in Fig. 2.

4 Results

In this study, the solubility of CO₂ in different concentrations of various solutions (%12MEA, %1PZ+%12MEA, %2PZ+%12MEA, and %3PZ+%12MEA) wt%, at different partial pressure (8.44, 25.33, and 42.22) kPa and at various temperatures (303, 313, and 323) K were determined and calculated experimentally. The experimental data and the predicted results from Kent-Eisenberg model are presented in the Table 3. In the improved Kent-Eisenberg model, the non-ideality effect was only considered in the coefficients of F function, and the amine protonation reaction and formation of MEA carbamate were regarded as non-ideal reactions. K'_7 (the equilibrium constant formation of the first order PZ carbamate) and K'_9 (the equilibrium constant

reversion of protonation the first order PZ carbamate) were optimized by the experimental data of CO₂ solubility for PZ + MEA aqueous solutions, and the obtained coefficients are given in Table 4.

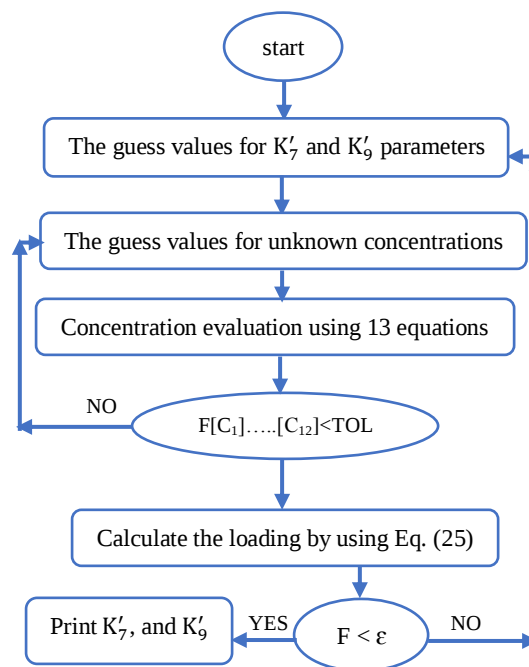


Fig. 2: Calculation algorithm by using the improved Kent-Eisenberg model based on solving simultaneous nonlinear equations

$$\alpha = \frac{[\text{CO}_2] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}] + [\text{MEACOO}^-] + [\text{PZCOO}^-] + [\text{H}^+ \text{PZCOO}^-] + [\text{PZ}(\text{COO}^-)_2]}{[\text{MEA} + \text{PZ}]_t} \quad (25)$$

Table 3 Comparison between the experimental and calculated loading of CO₂ in the aqueous solution of MEA+ PZ at different temperatures.

T/K	P _{CO2} kPa	MEA (12%wt)			MEA(12%wt)+PZ(1%wt)			MEA(12%wt)+PZ(2%wt)			MEA(12%wt)+PZ(3%wt)		
		L·Exp.	L·Calc.	AD%	L·Exp.	L·Calc.	AD%	L·Exp.	L·Calc.	AD%	L·Exp.	L·Calc.	AD%
303	8.44	0.009	0.015	66.6	0.011	0.014	27.2	0.012	0.014	16.6	0.013	0.012	7.7
313	8.44	0.008	0.013	62.5	0.009	0.012	33.3	0.01	0.011	10	0.0115	0.0111	3.4
323	8.44	0.005	0.011	120	0.006	0.01	66.6	0.007	0.01	42.8	0.008	0.009	12.5
303	25.33	0.091	0.139	52.7	0.098	0.128	30.6	0.101	0.119	17.8	0.104	0.111	6.7
313	25.33	0.082	0.111	35.3	0.092	0.103	11.9	0.094	0.095	1	0.097	0.089	8.2
323	25.33	0.07	0.091	30	0.08	0.085	6.2	0.082	0.078	4.8	0.088	0.073	17
303	42.22	0.169	0.232	37.2	0.185	0.214	15.6	0.201	0.199	1	0.216	0.185	14.3
313	42.22	0.153	0.185	20.9	0.163	0.171	4.9	0.179	0.159	11.1	0.194	0.148	23.7
323	42.22	0.128	0.152	18.7	0.138	0.141	2.1	0.153	0.131	14.3	0.162	0.122	24.6

Table 4: Optimized coefficients of K'_7 and K'_9 equilibrium constants

K_i	a_1	a_2	a_3	b_1	b_2	c_1	c_2
K'_7	-2.03	-1.08	-0.017	-3.51	-4.42	-0.82	0.287
K'_9	3.97	-0.072	0.0654	1.93	2.78	1.2	-0.325

4-1 Effect of CO₂ partial pressure, temperature and PZ concentration for the systems of (MEA-PZ-H₂O-CO₂)

Figs. 3 to 6 show the effects of partial pressure, temperature, and concentration parameters.

4-1-1 Effect of partial pressure

As it is shown in the charts, an increase of CO₂ partial pressure enhances CO₂ solubility in the solution. The higher the partial pressure of the CO₂ gas, the greater the solubility.

In similar works, Murshid et al. (31) and Yang et al. (32) observed that CO₂ solubility in AMP + PZ mixture enhances with the increase of CO₂ pressure.

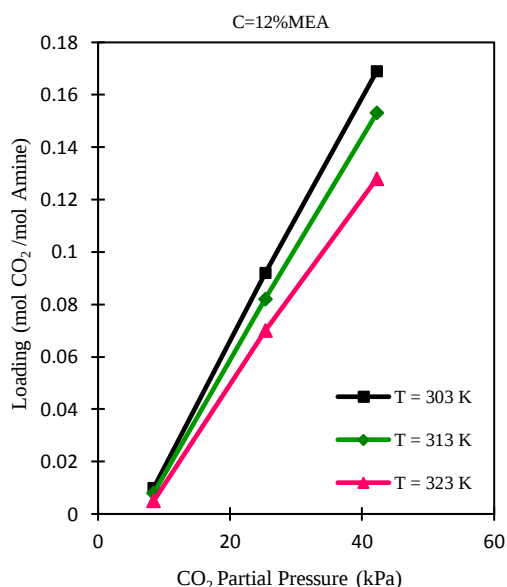


Figure 3: effect of CO₂ partial pressure on the loading value for (MEA-CO₂-H₂O) system at concentration (12%MEA) and different temperature

4-1-2 Effect of temperature

One of the affecting factors on the solubility is temperature. As can be seen, CO₂ solubility in different solvents decreases with temperature rise. This can be justified by the fact that CO₂ molecule is a nonpolar molecule and at low temperatures, it is almost stable. Therefore, its dissolution is physical, and the bond between molecules of solute and solvent is physical. As the temperature of the solution increases, the kinetic energy of its molecules increases, and this results in breaking of the weak bond, and the molecules of dissolved gas which have more energy than the molecules of solvent are removed from the solution which reduces the solubility. It can be said that PZ acts like amine solvents, so for this solvent, the temperature rise reduces the loading. In another similar work, Murshid et al. (31) showed that the solubility of CO₂ in a solution of AMP + PZ decreases with temperature.

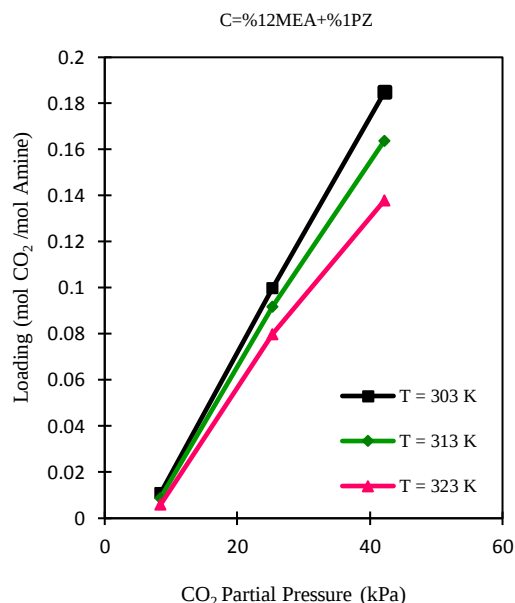


Figure 4 effect of CO₂ partial pressure on the loading value for (MEA-PZ-CO₂-H₂O) system at concentration (12%MEA+1%PZ) and different temperature

4-1-3 Effect of PZ concentration

The increase of loading (numbers on the y axis) is apparent from the increase of PZ concentration at all temperatures and partial pressures. In addition, Chung et al. (33) showed that adding PZ to TEA would increase CO₂ loading. Also, Dash et al. (34, 35), Murshid et al. (31), Yang et al. (32), and Ali and Aroua (36) showed that increasing the concentration of PZ in the solution results in the enhancement of the CO₂ solubility.

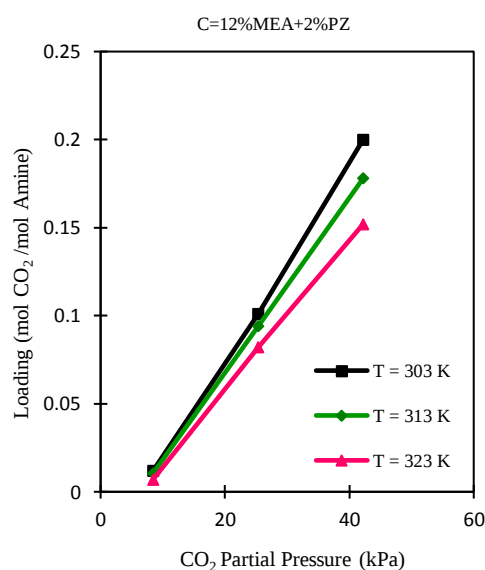


Figure 5 effect of CO₂ partial pressure on the loading value for (MEA-PZ-CO₂-H₂O) system at concentration (12%MEA+2%PZ) and different temperature

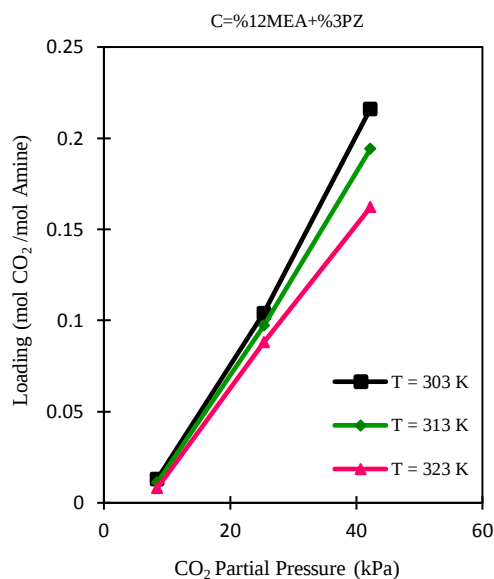


Figure 6 effect of CO₂ partial pressure on the loading value for (MEA-PZ-CO₂-H₂O) system at concentration (12%MEA+3%PZ) and different temperature

5 Conclusions

The results of this research are as follows:

- The increase of pressure or decrease of temperature causes the increase of CO₂ solubility in each of the studied amines.
- By increasing the concentration of PZ in the mixture of MEA + PZ, the solubility of CO₂ increases.
- The experimental results on PZ showed that this amine has a very high potential for CO₂ absorption, and as an activator, it can have a significant effect on the performance of the amine MEA.
- The average absolute deviations (AAD%) which was calculated by the difference between the forecasted value of the model and the experimental results was 24.4%.

Acknowledgement

The authors acknowledge the immense technical support of Jami Institute of Technology, Isfahan, Iran.

Competing interests

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

Authors' contribution

All authors of this study have a complete contribution for sample collection. Corresponding author was responsible for data collection, data analyses and manuscript writing, while an in-depth review was carried out by the co-author.

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