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CO_2 Removal from Gas Mixtures by Aqueous Solutions of MEA and (MEA+AEEA) and Results Comparing Using the Modified Kent-Eisenberg Model

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Abstract

Considering the growing importance of alkanolamine aqueous solvents in gas refineries or other powerhouses, it is essential to achieve the appropriate solution for CO_2 absorption. This requires to produce systematic vapor-liquid equilibrium data in a wide range of temperature, CO_2 partial pressure and different alkanolamines concentration. In this research with the application of equilibrium pilot plant in local atmospheric pressure, CO_2 solubility data have been reported in MEA solvent and its blend with AEEA in temperatures (303, 313 and 323) K, CO_2 partial pressures of (8.44, 25.33 and 42.22) kPa, concentrations of 12 wt% for MEA and (12+1, 12+2 and12+3) wt% for (MEA+AEEA). The measured solubility was then predicted by the theoretical model of modified Kent Eisenberg. The constant parameters of the apparent equilibrium for the porotonation and carbamate reaction in the Modified Kent Eisenberg model were optimized with the MATLAB software. It was conclude that CO_2 solubility values in all the studied experiments increased with increasing CO_2 partial pressure while increasing temperature and solvent concentration decreased the solubility. The comparison between the CO_2 absorption into the MEA solvent alone and AEEA activated MEA shows that (MEA+AEEA) blend in compare to the single MEA has a higher CO_2 loading. Also %AAD values for the solubility of MEA and (MEA+AEEA) were found to be 3 and 17.28 respectively.

Keywords: CO_2 solubility, MEA, AEEA, Modified Kent–Eisenberg model, correlation

1 Introduction

Industrialization and rapid population growth during the previous century have led to further contaminations on planet earth. CO_2 is one of the major gases released from chimneys of factories and exhaust of vehicles and is regarded among the critical industrial concerns (1). CO_2 can be collected from gas mixtures through several different methods including chemical-physical absorption, selective absorption by means of solid absorbent, and membrane separation (2). Among these technologies, chemical absorption using aqueous alkanolamine solution is the most developed and reliable one. Among the alkanolamine solutions, MEA is the most popular and common solvent among the available solvents and one of the most conventional amines used for CO_2 absorption from natural gases or mixture of gases. Compared to other amine solvents. This solvent has the highest absorption rate of acidic gases. Low molecular weight, low absorption of hydrocarbons, and high alkalinity are among its other advantages (2). However, certain important challenges exist in utilization of MEA like high corrosion rate of equipment, high energy consumption in recycling, and requirement of large volume of absorption. Hence, it is greatly essential to find a new solvent compound with low energy demand and acceptable cost-effectiveness. For higher CO_2 solubility, activators such as AEEA, PZ, and HMDA can be used as well. AEEA is a di-amine containing a first type and a second type amine group. This composition causes AEEA to have a good potential for CO_2 absorption. In (CO_2-AEEA-H_2O) system, CO_2 simultaneously reacts with both amine groups present in the molecular structure of AEEA. Daniel Bonenfant et al. (3) demonstrated that AEEA has the greater capacity of CO_2 absorption compared to MEA whereas its CO_2 desorption capacity is lower than MEA. Also in another study (4), the researchers analyzed the absorption capacities of CO_2 and H_2S in AEEA solution and its mixture with MDEA and TEA solvents. Zoghi et al. (5) measured absorption capacity of CO_2 gas at high partial pressures and reported effect of addition of AEEA as activator to MDEA solution. Mondal & Bajpai (6) studied CO_2 solubility in combination of DEA and AEEA solutions. Their results indicated that increase in molar fraction of AEEA in the solvent mixture led to an increase in CO_2 solubility, the same trend was observed with increase of partial pressure. It is essentially necessary to have knowledge about gas – liquid

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equilibrium of acidic gases in alkanolamines. Therefore, a practical thermodynamic model is required; the respective model is supposed to be capable of comprehensively and accurately predicting solubility under different conditions of temperature, pressure, amine concentration, and acidic gas loading. The most notable instances of such models include: Kent and Eisenberg, Deshmukh – Mather, electrolyte-NRTL derived from Chen & Evans and extended UNIQUAC (7). In this research, the performance of MEA and AEEA mixture in the CO_2 absorption was investigated at different operating conditions. Also, the Kent Eisenberg thermodynamic model was used based on the experimental data. The model parameters were optimized using a rigorous optimization method by minimizing an objective function.

2 Materials and Methods

2.1 Materials

Sample solutions of MEA (purities > 99.5 mass %) and AEEA (purities > 98.0 mass %) were obtained from Merck Co. The mixture of CO_2 (purity > 99.9 mol %), and nitrogen (N_2) (purity > 99.6 mol %) was purchased from ISFAHAN GAS Co; and all of the solutions were prepared with deionized water.

2.2 Apparatus and Experimental Procedure

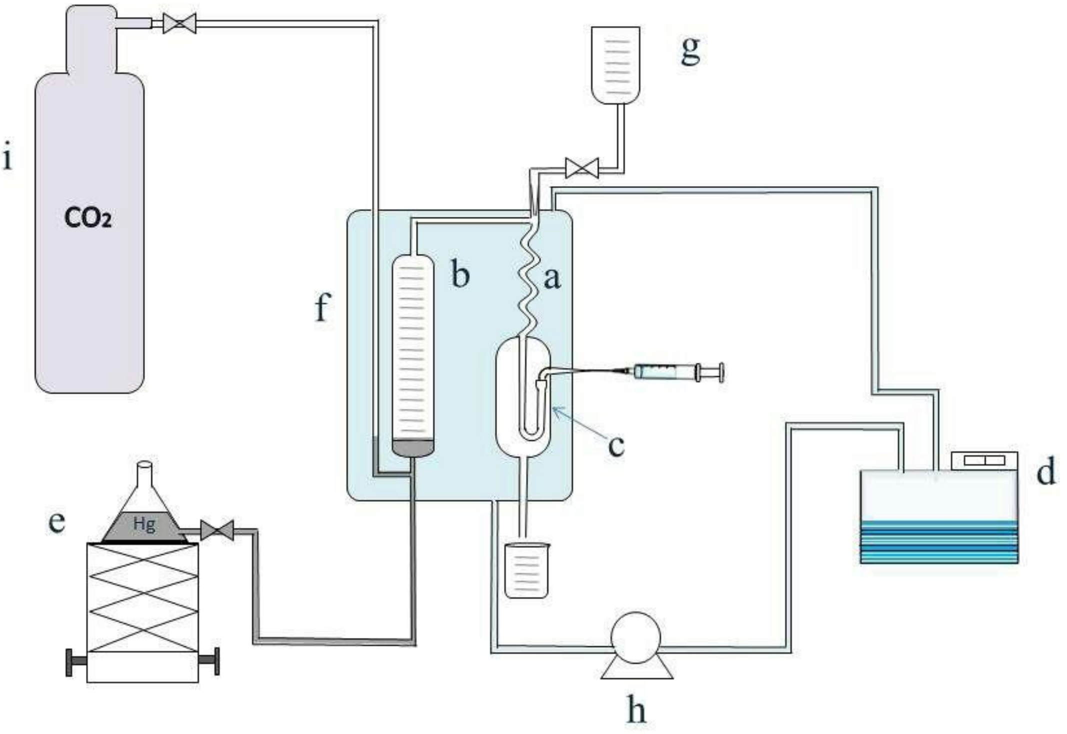
The solubility system used in the present project for conducting the experiments is analogous to the stems utilized by jahangiri et al. (8-11) for acquiring solubility of CO_2. The respective system consists of the following elements as illustrated in Figure 1.

The research method is explained as follows; water temperature is fixed at a specific value, then the mercury vessel attached to the gas burette is lowered by means of the jack. The capsule valve is opened to fill the apparatus by the tested gas. Subsequently, the gas inlet valve is closed and the gas will be pressurized inside the burette by moving the mercury vessel upwards by means of the movable jack and blocking the gas outflow by water. Then, the valve of burette containing solvent is opened so as to let the solvent enter the spiral tube. The internal pressure of apparatus will be reduced proportional to the amount of gas dissolved into the solvent. The pressure drop is compensated by raising the mercury vessel in order to carry out the experiment at constant pressure. Pressure is adjusted using the height variations of the solution in two arms of the U-shaped tube. The molar volume of the dissolved gas is computed via the equation of state of ideal gas (Equation (1)):

$$P\upsilon = RT \tag{1}$$

And through Equation (2), number of CO_2 moles will be determined by dividing the recorded volume in the laboratory to the acquired molar volume:

$$n = \frac{V}{\upsilon} \tag{2}$$



a:Spiral tube, b:Scaling burette, c:Manometer, d:Water bath, e:Mercury Jack, f:Cell, g:Solvent Container, h:Circulating Pump, i:CO_2 Capsule

Figure. 1 Schematic diagram of the solubility apparatus

Equation (3) is used to calculate the number of moles of the solvents consumed in a multi-component mixture:

$$n_t = \frac{Vd}{M_W} \qquad (3)$$

where “V” is volume, d is density, and Mw is the molecular mass. “n_t” is total number of models consumed from the mixture of solvents and it can be written. In the equations above, x_i, d_i, and $(M_w)_i$ respectively denote molecular percentage of solvent “i” in the mixture, density of solvent “i”, and molecular mass of solvent “i”. Having values of CO_2. This procedure is resumed until certain volume of gas is consumed and the consumed volume is read from the scaling burette. The CO_2 loading can be determined using Equation (4):

$$\alpha_{co_2} = \frac{mol_{co_2}}{mol_{a\min e}} \qquad (4)$$

3 Modified Kent-Eisenberg Model

Kent & Eisenberg (12) developed a simple thermodynamic model for prediction of equilibrium data in (Amin + H_2O + CO_2) systems using apparent equilibrium constants. In the respective model, activity coefficients of all substances present in equilibrium reactions is assumed equal to 1 and non-ideality of system is incorporated into the equilibrium constants and are modified as tuning parameters (13). The gaseous phase is assumed as ideal taking into account the values obtained for compressibility coefficient, and of course, low overall pressure of 1 atm (14).

3.1 Model Framework

3.1.1 Physical and chemical equilibria

The absorption of CO_2 into an amine solution includes both phase and chemical equilibria. The gas phase CO first dissolves into the aqueous phase:

$$CO_2(g) \overset{H_{CO_2}}{\rightleftarrows} CO_2(aq) \qquad (5)$$

The dissolved CO_2 undergoes a series of chemical reactions and forms various ionic species. For (MEA+AEEA) blends, the following reactions are considered:

$$Dissociation\ of\ water:\ H_2O \overset{K_1}{\rightleftarrows} H^+ + OH^- \qquad (6)$$

$$Dissociation\ of\ carbon\ dioxide:\ CO_2 + H_2O \overset{K_2}{\rightleftarrows} HCO_3^- + H^+ \qquad (7)$$

$$Dissociation\ of\ bicarbonate\ ion:\ HCO_3^- \overset{K_3}{\rightleftarrows} CO_3^{2-} + H^+ \qquad (8)$$

$$Dissociation\ of\ protonated\ MEA:\ MEAH^+ \overset{K_4}{\rightleftarrows} MEA + H^+ \qquad (9)$$

$$Formation\ of\ carbamate\ MEA:\ MEA + HCO_3^- \overset{K_5}{\rightleftarrows} MEACOO^- + H_2O \qquad (10)$$

$$Dissociation\ of\ monoprotonated\ AEEA:\ AEEAH^+ \overset{K_6}{\rightleftarrows} AEEA + H^+ \qquad (11)$$

$$Dissociation\ of\ diprotonated\ AEEA:\ {}^+HAEEAH^+ \overset{K_7}{\rightleftarrows} AEEAH^+ + H^+ \qquad (12)$$

$$\begin{aligned} Formation\ of\ carbamates:\ & AEEA + CO_2 \overset{K_8}{\rightleftarrows} AEEACOO_P^- + H^+, \\ & AEEA + CO_2 \overset{K_9}{\rightleftarrows} AEEACOO_S^- + H^+ \end{aligned} \qquad (13)$$

$$\begin{aligned} Dissociation\ of\ protonated\ carbamates:\ & {}^+HAEEACOO_P^- \overset{K_{10}}{\rightleftarrows} AEEACOO_P^- + H^+ \\ & {}^+HAEEACOO_S^- \overset{K_{11}}{\rightleftarrows} AEEACOO_S^- + H^+ \end{aligned} \qquad (14)$$

$$Formation\ of\ dicarbamate:\ AEEACOO_P^- + AEEACOO_S^- + 2CO_2 \overset{K12}{\rightleftarrows} 2\,{}^-OOCAEEACOO^- + 2H^+ \qquad (15)$$

Jacobsen et al.(15) proposed the governing equations of AEEA-CO_2-H_2O system using NMR equations. Based on the respective equations, there exist diverse ion components in the solution. AEEA is composed of two groups of primary and secondary amines. Nevertheless, it reacts similar to monoamines. Its difference is in gas absorption capacity and also larger number of its components that formed in comparison with monoamines. According to the test performed by Jacobsen & Ma’mun, 14 components have the possibility to be present in the aqueous solution of system.

$$AEEA, AEEAH^{+}, {}^{+}HAEEAH^{+}, AEEACOO_{P}^{-}, AEEACOO_{S}^{-}, {}^{+}HAEEACOO_{P}^{-}, {}^{+}HAEEACOO_{S}^{-}, {}^{-}OOCAEEACOO^{-}$$
$$, CO_{2}, HCO_{3}^{-}, CO_{3}^{2-}, H_{2}O, H^{+}, OH^{-}$$

In addition to the above equations, the following equations also apply to the system:

$$Electoroneutrality:$$
$$[MEAH^{+}]+[AEEAH^{+}]+[H^{+}]+2[{}^{+}HAEEAH^{+}]=[HCO_{3}^{-}]+2[CO_{3}^{2-}]+[AEEACOO_{P}^{-}]+[AEEACOO_{S}^{-}]+[MEACOO^{-}]+[OH^{-}]$$
$$+2[{}^{-}OOCAEEACOO^{-}]$$

$$CO_{2} balance:$$
$$\alpha[MEA+AEEA]_{total}=[CO_{2}]+[HCO_{3}^{-}]+[CO_{3}^{2-}]+[AEEACOO_{P}^{-}]+[AEEACOO_{S}^{-}]+[{}^{+}HAEEACOO_{P}^{-}]+[{}^{+}HAEEACOO_{S}^{-}]+$$
$$[MEACOO^{-}]+[{}^{-}OOCAEEACOO^{-}]$$

$$MEA\ balance:$$
$$[MEA]_{t}=[MEA]+[MEAH^{+}]+[MEACOO^{-}]$$
$$AEEA\ balance:$$
$$[AEEA]_{t}=[AEEA]+[AEEAH^{+}]+[AEEACOO_{P}^{-}]+[AEEACOO_{S}^{-}]+[{}^{+}HAEEACOO_{P}^{-}]+[{}^{+}HAEEACOO_{S}^{-}]+[{}^{+}HAEEAH^{+}]$$
$$+[{}^{-}OOCAEEACOO^{-}]$$

$$The\ vapor-liquid\ equilibrium\ for\ CO_{2}\ \text{(Henry Law)}:$$
$$P_{CO_{2}}=H_{CO_{2}}.[CO_{2}]$$

where H_{CO2} and P_{CO2} are the Henry's constant and partial pressure of CO_2, respectively. Chemical equilibrium constant is taken as a function of temperature and expressed as in Equation (16). The values of parameters A, B, C, and D for each reaction are specified in Table (1).

$$H_{CO_2} = ex(\frac{A}{T} + BlnT + CT + D) \qquad (16)$$

In the modified Kent-Eisenberg model, by adding a function in terms of partial pressure, concentration, and temperature as a function of K_8 and K_{10} equilibrium constants, non-ideality effect can be somehow represented in the values of functions K'_8 and K'_{10}. It must be noted that in this method only values of parameters K'_8 and K'_8 are modified using optimization of proposed parameters, and, for the rest of reactions, the same initial equilibrium constants are used. The objective function used here has been selected based on difference of values of the calculated molar load and the molar load acquired from experimental results. The value of molar load is calculated via Equation (17). The stages of solving Kent – Eisenberg model are depicted in Figure 2.

$$K'_8 = K_8\,F,\ K'_{10} = K_{10}\,F \qquad F = exp\,(a_1 + a_2/\,T(K) + a_3/T(K) + b_1\,ln\,(P_{CO_2}\,(kpa)) + b_2\,(P_{CO_2}\,(kpa)) + c_1[MEA] + c_2[AEEA])$$

Table 1: Henry's constant and equilibrium constant parameters used in the Kent–Eisenberg model for reactions (5) – (15)

K_i	A	B	C	D	Ref.
K_1	-13445.9	-22.4773	0	140.932	(16)
K_2	-12092.1	-36.7816	0	235.482	(16)
K_3	-12431.7	-35.4819	0	220.067	(16)
K_4	-17.3	0	0.05764	-38.846	(16)
K_5	-1545.3	0	0	2.151	(16)
K_6	-5865.15	0	0	0.9609	(17)
K_7	-5074.99	0	0	4.7738	(17)
K_8	4208.91	0	0	-31.136	(17)
K_9	17375.05	0	0	-63.297	(17)
K_{10}	18119.88	0	0	-76.993	(17)
K_{11}	-25591.42	0	0	63.6	(17)
K_{12}	-2169.54	0	0	2.743	(17)
H_{CO_2}	-8477.711	-21.9574	0.005780	155.1699	(17)

$$\alpha_{Calc} = \frac{[CO_2]+[HCO_3^-]+[CO_3^{2-}]+[MEACOO^-]+[AEEACOO_P^-]+[AEEACOO_S^-]+[^+HAEEACOO_P^-]+[^+HAEEACOO_S^-]+[^-OOCAEEACOO^-]}{[MEA+AEEA]_t} \quad (17)$$

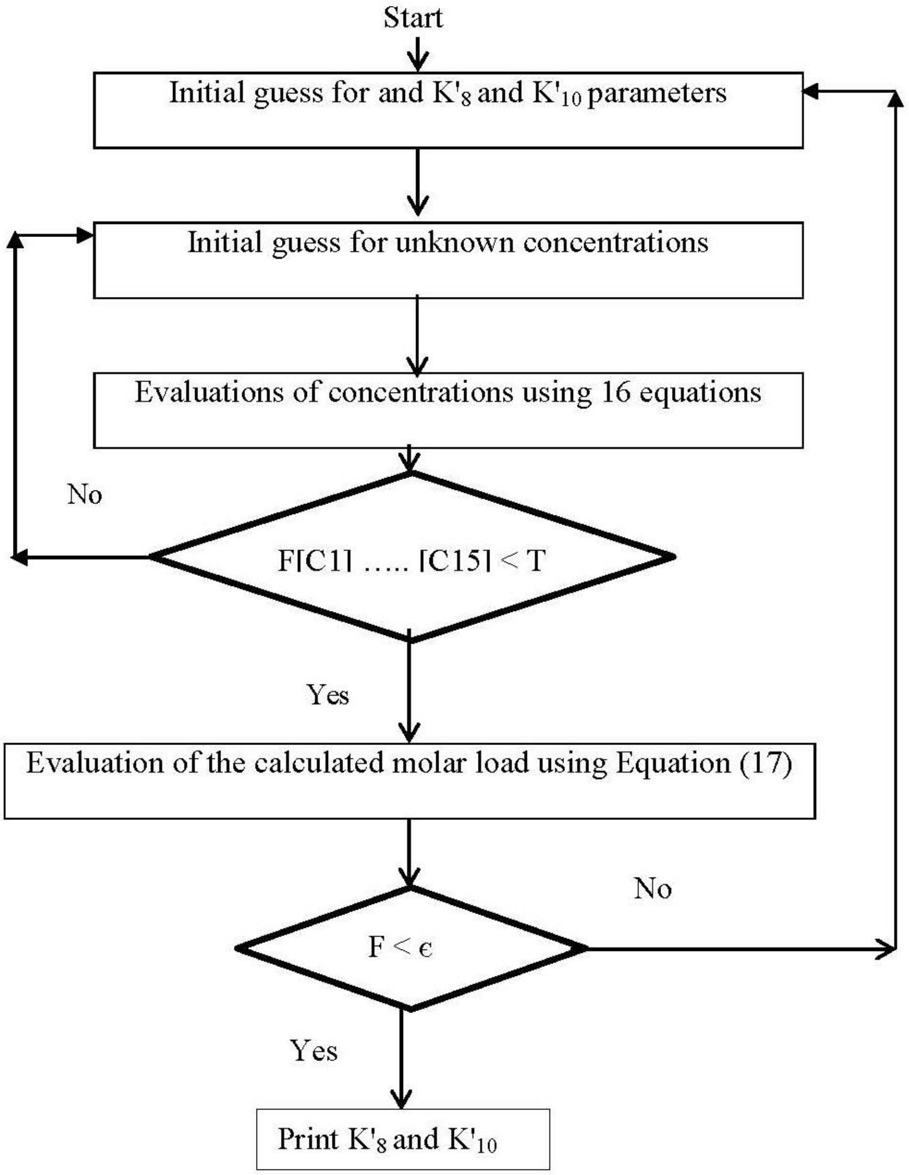


Figure 2: Computational algorithms using Kent-Eisenberg model based on simultaneous solution of non-linear equations

4 Results and Discussion

K'_8 (equilibrium constant of amine protonation reaction) and K'_{10} (equilibrium constant of carbamate production) were optimized by the experimental date of CO_2 solubility in MEA solution. The acquired coefficients are included in Table (2). Average absolute error of model is determined via the equation (18):

$$\%AAD = \frac{1}{n}\sum_{i=1}^{n}\left|\alpha_{calculated} - \alpha_{experimental}\right| \times 100 \qquad (18)$$

According to Equation (18), average absolute error of Kent-Eisenberg model compared to experimental data was predicted equal to 17.28% for (MEA-AEEA-CO_2-H_2O) system. Comparison of the measured loading values and calculated loading values using the modified Kent-Eisenberg model and absolute error percentage for each of the data are provided in Table (3).

In the present research, solubility data of CO2 absorption by MEA and (MEA+AEEA) were measured at different temperatures, partial pressure, and concentrations by CO2 absorption system at local atmospheric pressure. The results were reported in Table (3) and Figure (3 (A, B, C)). The diagrams were similar to one another for different temperatures, partial pressure, and concentrations (each one individually). And for the same reason, illustration of all diagrams is skipped here.

Table 2: Optimized coefficients of K'₈ and K'₁₀ equilibrium constants

K_i	**a₁**	**a₂**	**a₃**	**b₁**	**b₂**	**c₁**	**c₂**
K'_8	1.0575	0.1942	0.0185	2.3147	-4.3255	1.5214	-0.789
K'_{10}	4.1444	4.4854	-1.0996	4.9750	3.3813	3.5043	-3.866

Table 3: Experimental and calculated loading of CO_2 in the aqueous solution of MEA and MEA+AEEA at different operating conditions

CO₂ Loading (α)											
Exp.	Calc.	AD%	Exp.	Calc.	AD%	Exp.	Calc.	AD%	Exp.	Calc.	AD%
MEA (12 wt %)			MEA (12 wt %) , AEEA (1 wt %)			MEA (12wt%) , AEEA (2wt%)			MEA (12 wt %) , AEEA (3 wt %)		
T=303 K, P_{CO2}=8.44 kPa											
0.015	0.043	2.81	0.012	0.367	35.54	0.012	0.368	36.94	0.013	0.393	38.04
T=313 K, P_{CO2}=8.44 kPa											
0.012	0.034	2.15	0.009	0.367	35.79	0.011	0.038	37.12	0.011	0.395	38.39
T=323 K, P_{CO2}=8.44 kPa											
0.006	0.028	2.16	0.005	0.366	36.14	0.004	0.038	37.76	0.003	0.394	39.16
T=303 K, P_{CO2}=25.33 kPa											
0.142	0.129	1.31	0.103	0.06	4.35	0.105	0.056	4.95	0.107	0.052	5.48
T=313 K, P_{CO2}=25.33 kPa											
0.125	0.102	2.35	0.091	0.047	4.38	0.09	0.044	4.63	0.089	0.041	4.84
T=323 K, P_{CO2}=25.33 kPa											
0.085	0.083	0.21	0.085	0.038	4.68	0.079	0.036	4.32	0.073	0.0333	3.97
T=303 K, P_{CO2}=42.22 kPa											
0.27	0.216	5.44	0.182	0.01	8.29	0.195	0.092	10.27	0.216	0.086	12.99
T=313 K, P_{CO2}=42.22 kPa											
0.23	0.17	6.02	0.172	0.078	9.32	0.184	0.073	11.12	0.186	0.068	11.81
T=323 K, P_{CO2}=42.22 kPa											
0.172	0.138	3.4	0.161	0.064	9.76	0.145	0.059	8.62	0.135	0.055	7.98
%AAD= 3.17			***%AAD*** = 16.47			***%AAD*** = 17.3			***%AAD*** = 18.07		

4.1 Effect of temperature for the systems of (MEA-AEEA-H_2O-CO_2)

Effects of temperature variations on CO_2 loading values are illustrated in Figure (3(A)). It is clear that loading trend declines with increasing temperature. This decline is reasonable taking into account the exothermic reaction because gas dissolution in liquid is normally exothermic. Accordingly, the temperature increase applied to the solution causes a change in the system and shifts the system toward the reactants so as to reduce this change. As a result, equilibrium concentration of gaseous phase will increase and gas solubility will decrease.
Chao Guo et al. (18) Mondal & Bajpai (6) and Kim & Sevendsen (19) demonstrated that AEEA acts like amine solvents, and as a result, temperature increase for this solvent will lead to reduction of molar load.

4.2 Effect of partial pressure for the systems of (MEA-AEEA-H_2O-CO_2)

Figure (3(B)) indicates that CO_2 solubility in the solution increases with increasing partial pressure at any temperature. This trend signifies that gas concentration will rise in liquid phase and hence, solubility will increase. For instance, Mondal & Bajpai (6) and also Najafloo et al. (20) in their research works for AEEA solution showed that CO_2 solubility is improved by increasing the partial pressure. In Table (3), for the (MEA + AEEA) mixture at partial pressure of 8.44, the loading value calculated by the model was largely different from the value acquired from experimental data. It can be therefore asserted that the model does not provide good prediction for this solution at low partial pressure.

4.3 Effect of concentration for the systems of (MEA-AEEA-H_2O-CO_2)

Reviewing the experimental results provided in Table (3) and Figure (3(C)), it is observed that addition of AEEA to MEA at temperatures of 303 K and 313 K results in increase of CO_2 loading (it increases less at temperature of 313 K), and also at the temperature of 323 K, increase of loading is observed at the temperature of 323 K with addition of AEEA in the first step and then a reduction of loading happens. This is indicative of the fact that solubility value declines at very high temperatures of AEEA. It can be also inferred that AEEA at high temperatures has a high absorption up to a certain concentration and the decline after the respective value might be attributed to release of carbamate ion at high temperatures.

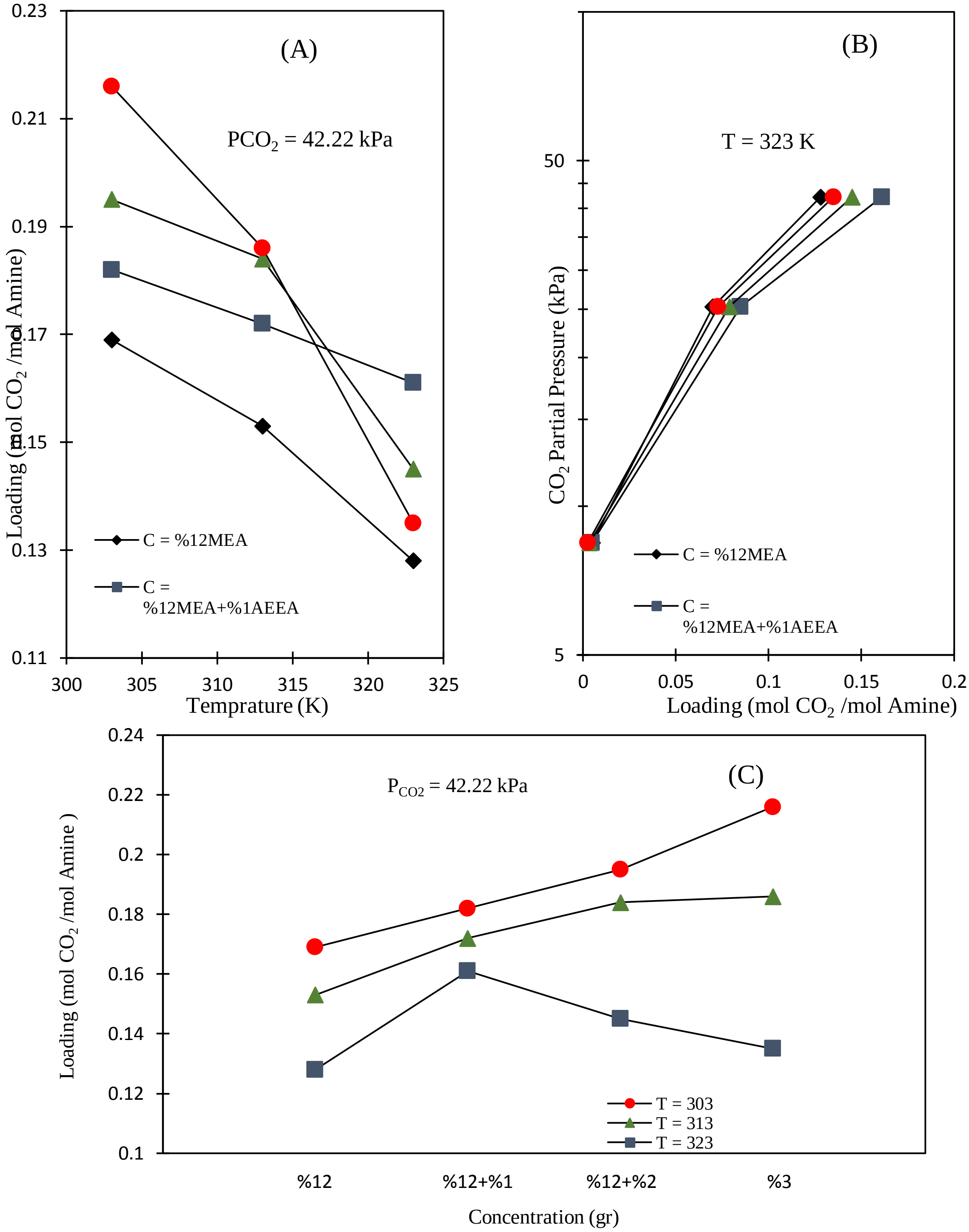


Figure 3: effect of operating conditions on CO_2 loading for ($MEA\text{-}AEEA\text{-}CO_2\text{-}H_2O$) system based on: (A): Temperature, (B) CO_2 Partial pressure, (C): Solvent concentration

5 Conclusions

In the present research, first the measured data of CO_2 solubility and (MEA+AEEA) mixture were experimentally determined and calculated at weight concentrations of 12% for MEA and weight concentrations of (12+1), (12+2) and (12+3) wt% for (MEA+AEEA) mixture at temperatures of 303, 313 and 323 K, and partial pressures of 8.44, 25.33, and 42.22 kPa Then, the vapor – liquid equilibrium data were modeled using modified Kent – Eisenberg model.

The conclusions derived from the present research can be summarized as below:

* With increasing concentration of AEEA in ($MEA\text{-}AEEA\text{-}CO_2\text{-}H_2O$) system, solubility of CO_2 gas increases at temperatures of 303 and 313 K but decreases at temperature of 323 K. The reason is due to release of carbamate ion.

* The experimental data of CO_2 gas solubility in (MEA+AEEA) mixture was modeled by means of Kent-Eisenberg model as well and the value of average

absolute error for (MEA-CO_2-H_2O) and (MEA-AEEA-CO_2-H_2O) system were predicted equal to 3.17 and 17.28, respectively.

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