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The Open Chemical Engineering Journal

Content list available at: www.benthamopen.com/TOCENGJ/

DOI: 10.2174/1874123101812010067



RESEARCH ARTICLE

Experimental Study of CO₂ Absorption in Potassium Carbonate Solution Promoted by Triethylenetetramine

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Received: February 20, 2018

Revised: April 3, 2018

Accepted: April 21, 2018

Abstract:
Background:

Separation of CO₂ as the major cause of global warming is essential. In this work, potassium carbonate (K₂CO₃) solution was selected as a base solvent for CO₂ absorption due to its ease of regeneration energy, low cost and low environmental impact. However, the absorption rate of CO₂ with K₂CO₃ needs to be improved by adding a suitable promoter. Therefore, the performance of CO₂ in K₂CO₃ solution promoted by triethylenetetramine (TETA) in terms of absorption capacity and absorption rate of CO₂ was studied.

Method:

Experiments were conducted at a total concentration of 2.5 (M) with different TETA mole fractions at temperatures of 303, 313 and 323 K, and CO₂ partial pressure up to 30 kPa using a stirred cell reactor. The effect of CO₂ partial pressure, temperature and concentration of TETA on absorption capacity and absorption rate of CO₂ in K₂CO₃+TETA solution was discussed in detail.

Results:

The CO₂ loading capacity obtained in this work was compared with monoethanolamine (MEA) and a better performance was observed for K₂CO₃+TETA solution. In addition, experimental results revealed that the addition of TETA to K₂CO₃ improved the CO₂ reaction rate. Finally, the response surface methodology was employed to correlate the CO₂ solubility. It was found that the correlated data are in good agreement with the experiment results.

Conclusion:

As an overall conclusion, the solution of K₂CO₃+TETA can be used as a promising absorbent in post combustion CO₂ capture processes.

Keywords: Greenhouse gas, CO₂ capture, Solubility, Absorption rate, Potassium carbonate, Monoethanolamine.

1. INTRODUCTION

The fossil fuel power plants are considered as the main source of greenhouse gases emission, which increase CO₂ concentration in the atmosphere and environmental issues [1, 2]. In order to reduce the CO₂ emission, it is essential to capture CO₂ from fossil fuel power plants [3]. Several technologies such as oxy-fuel combustion, pre combustion and post-combustion are used for CO₂ capture [4]. Among them, post-combustion using a solvent is widely employed as one of the most reliable and economical methods for CO₂ capture [5]. One of the main challenges of this method is the selection of a suitable solvent [6]. To be selected as a solvent, it needs to satisfy several desired properties, including

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high CO₂ loading capacity, low volatility, high absorption rate, high thermal stability and low heat of absorption [7]. Different types of solvents have been studied for the CO₂ capture such as ionic liquids, amino acid salts, inorganic solvents and alkanolamines [8]. The ionic liquids are used as solvents because of some advantages such as high CO₂ capacity and thermal stability. However, they have disadvantages such as high cost and viscosity [9, 10]. Amino acid salt solutions gained interest as a solvent because of low volatility, less toxicity and low thermal stability. The main challenges associated with amino acid salts are high cost and precipitation of carbonates at high concentration [11, 12]. The inorganic solvents such as tri-sodium phosphate and potassium carbonate are another category of solvents with desired characteristics such as low corrosion rate and lower heat of absorption than amines [13]. Alkanolamines such as monoethanolamine (MEA) have been used extensively in many CO₂ capture processes because of its low solvent cost and fast reaction kinetic [14]. However, MEA has several disadvantages such as low CO₂ absorption capacity, thermal or chemical degradation, high regeneration energy requirement and corrosive [15]. Many researchers have tried to find a suitable solvent that minimize the cost and penalty in the power plant efficiency [16]. The solution of potassium carbonate (K₂CO₃) has been received much attention due to its advantages such as low environmental impact, low solvent cost, low absorption heat and low thermal degradation [17, 18]. However, its slow reactivity with CO₂ needs to be improved [19]. To solve this problem, researchers have suggested the addition of promoters to K₂CO₃ in order to enhance the absorption rate. As an example, Cullinane and Rochelle [20], reported the absorption rate of CO₂ in 20-30 wt% potassium carbonate promoted by 0.6 kmol/m³ piperazine (PZ) at temperatures from 313 to 353 K. They concluded that the CO₂ absorption rate was increased by the addition of PZ. Kim et al. [21] investigated the absorption rate of CO₂ in blend solution of 15 wt% potassium carbonate and 7.5 wt% homopiperazine (homoPZ). They observed that K₂CO₃+homoPZ has a higher absorption rate than pure K₂CO₃. Thee et al. [22] used 5 and 10 wt% monoethanolamine (MEA) as a promoter for K₂CO₃. The results revealed that CO₂ absorption rate in K₂CO₃+MEA solution is higher than potassium carbonate. Thee et al. [23] studied the effect of the addition of boric acid to 30 wt% potassium carbonate at 353.15 K. It was found that boric acid has a positive effect on CO₂ absorption rate at blended solution. Shen et al. [24] examined reaction kinetic in a mixture of 35 wt% K₂CO₃ and arginine at temperatures from 313 K to 343 K. It was discovered that arginine can be an effective promoter for potassium carbonate. Thee et al. [25] selected three amino acids, including glycine, proline and sarcosine as potential promoters for potassium carbonate solution and obtained reaction kinetic at temperatures from 313.15 to 353.15 K. Kim et al. [26] evaluated CO₂ absorption rate and capacity of potassium carbonate solution promoted by 2-methylpiperazine at 313, 333 and 353 K. They found that the addition of a small amount of 2-methylpiperazine to potassium carbonate can increase the CO₂ absorption capacity and rate. Bhosale et al. [27] added ethylaminoethanol to potassium carbonate and calculated reaction kinetics at temperatures (303-318 K). It was found that ethylaminoethanol acts as a good promoter. This motivated us to select triethylenetetramine (TETA) as a promoter in order to improve absorption rate of CO₂ in K₂CO₃ solution. TETA with four functional groups, including two primary and two secondary amino groups has advantages compared to other promoters like methyldiethanolamine, 2-((2-aminoethyl)amino)ethanol, diethanolamine, 2-amino-2-methyl-1-propanol and monoethanolamine in terms of fast reaction kinetic, high CO₂ solubility and low regeneration energy. Due to these advantages, TETA was chosen as a promoter for potassium carbonate. Therefore, in this work, absorption capacity and rate of absorption of CO₂ in K₂CO₃+TETA were evaluated using a stirred cell reactor at a temperature range of (303.15 to 323.15) K and pressure up to 30 kPa. The CO₂ absorption capacity of K₂CO₃+TETA as a function of pressure, temperature and concentration was also correlated by response surface methodology. In fact, this work presents new CO₂ solubility data in K₂CO₃+TETA, which are important and necessary for modeling of CO₂ absorption processes in industrial application.

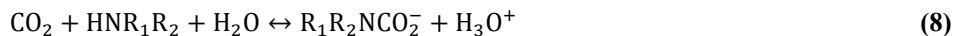
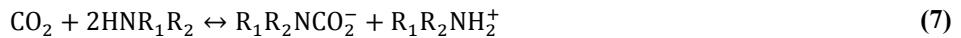
2. REACTION MECHANISM

When CO₂ is absorbed in potassium carbonate solution, the following reactions take place [26, 28]:





In comparison to the potassium carbonate solution, alkanolamines have a high rate of reaction with dissolved CO₂. The general reaction mechanism between CO₂ and amine can be indicated in Eqs. (7, 8), which R₁ and R₂ are carbon side chains [29]:



3. MATERIALS AND METHODS

3.1. Materials

All chemicals used in this study, including potassium carbonate (K₂CO₃), triethylenetetramine (TETA) and monoethanolamine (MEA) with grade >98% pure were purchased from Acros Organics and used without any further purification. The chemical structure of TETA is given in Fig. (1). Experiments were carried out in blended solutions of K₂CO₃ and TETA with the compositions of 2.25 M K₂CO₃ + 0.25 M TETA, 2 M K₂CO₃ + 0.5 M TETA, 1.75 M K₂CO₃ + 0.75 M TETA, 1.5 M K₂CO₃ + 1 M TETA in temperatures 303-323 K and pressures up to 30 kPa.

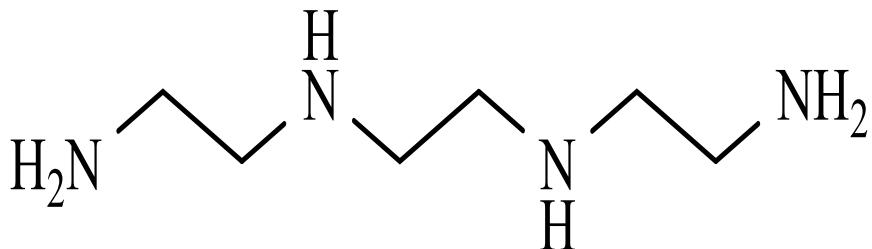


Fig. (1). The chemical structure of TETA.

3.2. Equipment and Experimental Procedure

The CO₂ absorption measurement procedure and equipment used in this study are same as employed in our previous works [30 - 33]. A glass stirred cell reactor was used to measure CO₂ solubility and initial absorption rate of CO₂ in the solution of K₂CO₃ promoted by TETA, as shown in Fig. (2). This reactor is connected to a gas storage tank and a water bath to control the temperature during the experiments. Both stirred cell reactor and gas storage tank are connected with temperature sensor and pressure transmitter. Before running the experiment, the reactor was purged with a vacuum pump. The solvent was placed in the reactor and the solution is allowed to reach a desired temperature. After that, CO₂ was fed to gas storage tank from gas cylinder and was heated using a water bath. Next, CO₂ was transferred to reactor from gas storage tank, and stirrer was switched on. The total moles of CO₂ transferred can be calculated from:

$$n_{\text{CO}_2} = \frac{[P_1 - P_2] V_s}{R T} \quad (9)$$

Where V_s, P₁ and P₂ are the volume, initial and final pressure of CO₂ in the gas storage tank, respectively. The CO₂ partial pressure in the reactor is decreased over time because of the chemical absorption of the CO₂ in solution and the pressure drop in the reactor is monitored and recorded every second by a pressure transmitter. Therefore, the initial absorption rate can be obtained from slope of CO₂ partial pressure changes versus time. The equilibrium partial pressure of CO₂ and the moles of CO₂ remaining in equilibrium cell were obtained by Eqs. 10 and 11, respectively:

$$P_{\text{CO}_2}^e = P_F - P_V \quad (10)$$

$$n_{\text{CO}_2}^r = \frac{V P_{\text{CO}_2}^e}{R T} \quad (11)$$

Absorption capacity of CO₂ in the solution of K₂CO₃+TETA is defined in terms of CO₂ loading α (mole CO₂ absorbed per mole of solvent).

$$\alpha_{\text{CO}_2} = \frac{n_{\text{CO}_2} - n_{\text{CO}_2}^r}{n_{\text{solvent}}} \quad (12)$$

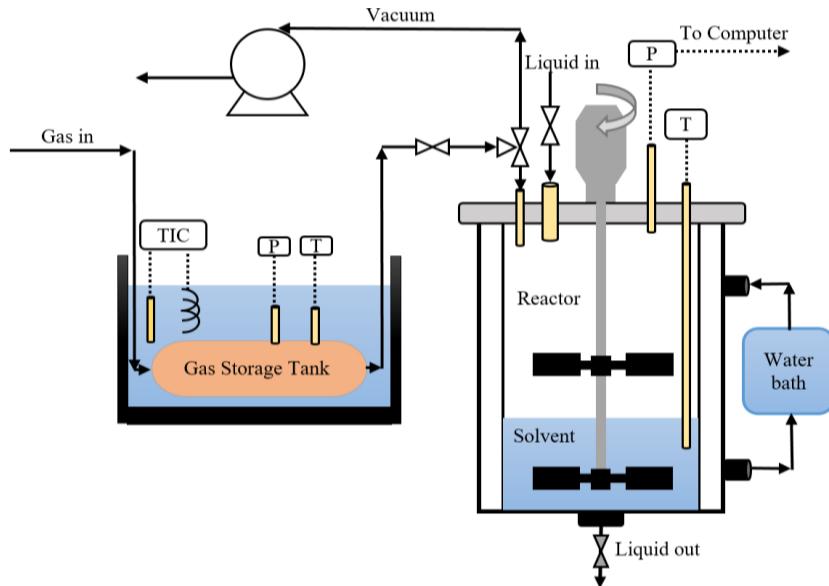


Fig. (2). Schematic diagram of the experimental set up.

As mentioned earlier, the method of CO_2 loading calculation can be found in our previous works. Three repeat runs were taken to check the reproducibility and data presented here represent averages.

4. RESULTS AND DISCUSSION

4.1. CO_2 Absorption Capacity in $\text{K}_2\text{CO}_3+\text{TETA}$ Solution

In order to validate equipment and experimental procedure, the loading capacity of CO_2 in the solution of 2.5 M monoethanolamine was measured at 313 K. Fig. (3) shows a good agreement exists between the CO_2 absorption capacities measured in this work and the values previously reported [34 - 36].

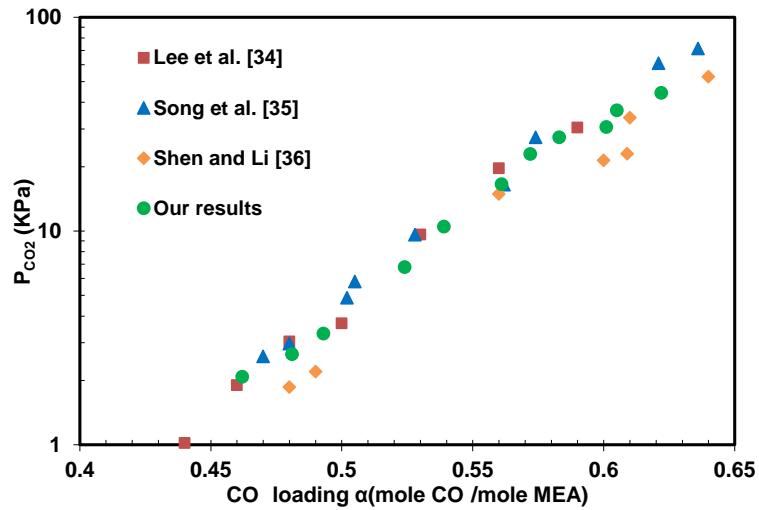


Fig. (3). Solubility of CO_2 in solution of 2.5 M MEA at 313.15 K.

CO_2 solubility data presents useful information for industrial application in design and modeling of CO_2 absorption in CO_2 capture process [24]. In this work, the CO_2 solubility in $\text{K}_2\text{CO}_3 + \text{TETA}$ solution was measured at temperatures 303–323 K, CO_2 partial pressures up to 30 kPa, and results listed in Table 1. As can be observed in Table 1, temperature has a negative effect on the CO_2 loading. In the other words, an increase in the temperature from 303 to 323 K leads to a decrease in loading capacity from approximately 0.8 to 0.65. This reduction can be explained by the fact that the equilibrium would shift in the backward direction with increasing temperature. In order to study the effect of pressure and concentration on the performance absorption capacity of the solvent, CO_2 partial pressure as a function of CO_2 solubility was plotted in Figs. (4–6). It can be seen that $\text{K}_2\text{CO}_3 + \text{TETA}$ solution absorbs more CO_2 at higher pressure which can be due to the increase in the driving force from the gas phase to the interface. According to Figs. (4–6), the absorption capacity also increases with the increase in concentrations of TETA. The highest CO_2 solubility was achieved at TETA mole fraction equal to 0.4. The reason for this behavior could be explained by the fact that TETA as a polyamine has four amine groups Fig. (1) that means TETA can absorb more CO_2 . Thus, a smaller amount of solvent is needed for CO_2 absorption with high efficiency in absorber that leads to a lower cost. The absorption performance of $\text{K}_2\text{CO}_3 + \text{TETA}$ solution was compared with pure K_2CO_3 , at 313.15 K, shown in Fig. (5). It is clear that CO_2 loading of $\text{K}_2\text{CO}_3 + \text{TETA}$ solution is much higher than pure K_2CO_3 in the order: 1.5 M $\text{K}_2\text{CO}_3 + 1$ M TETA > 1.75 M $\text{K}_2\text{CO}_3 + 0.75$ M TETA > 2 M $\text{K}_2\text{CO}_3 + 0.5$ M TETA > 2.25 M $\text{K}_2\text{CO}_3 + 0.25$ M TETA > 2.5 M K_2CO_3 , suggesting high ability of TETA for CO_2 capture.

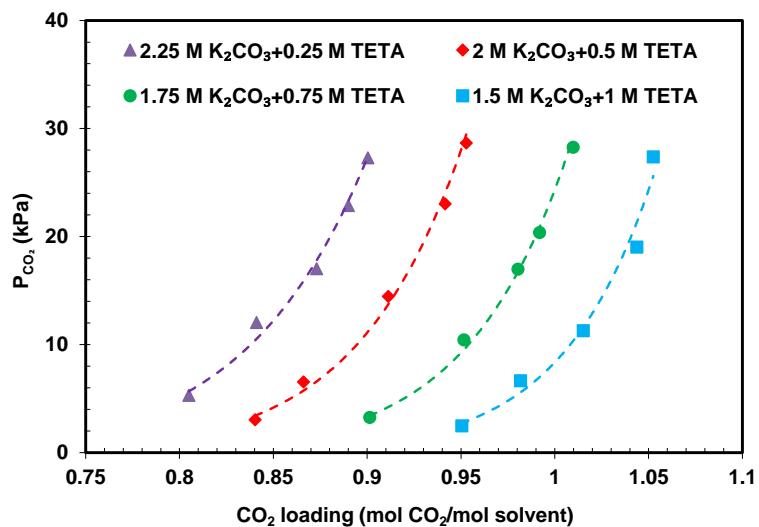


Fig. (4). Loading capacity of CO_2 in $\text{K}_2\text{CO}_3 + \text{TETA}$ solution at 303.15 K.

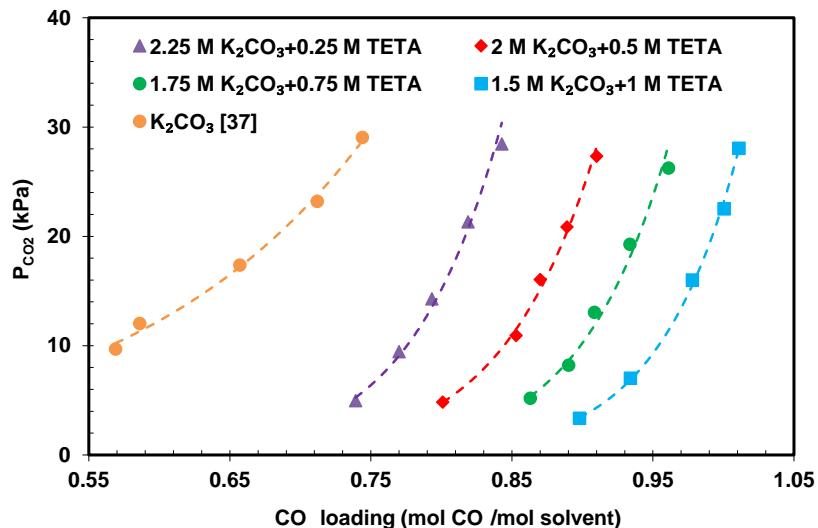
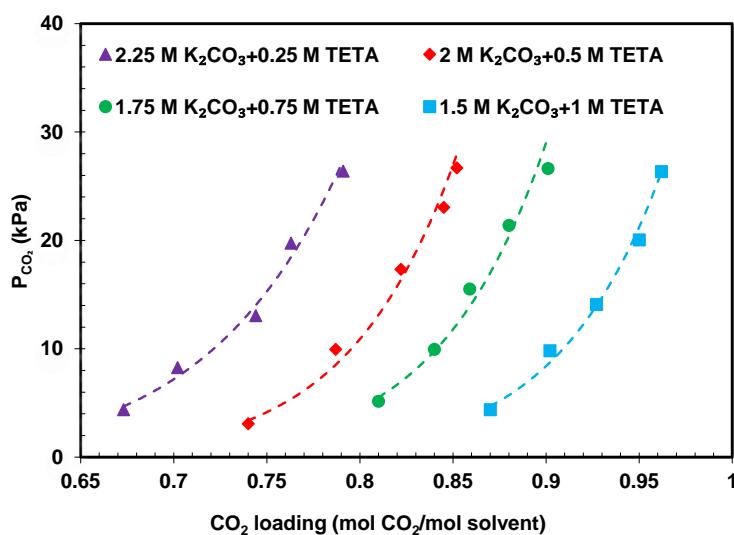


Fig. (5). Loading capacity of CO_2 in $\text{K}_2\text{CO}_3 + \text{TETA}$ solution at 313.15 K.

Table 1. Loading capacity of CO₂ in solution of K₂CO₃+TETA.

2.25 M K ₂ CO ₃ + 0.25 M TETA		2 M K ₂ CO ₃ + 0.5 M TETA		1.75 M K ₂ CO ₃ + 0.75 M TETA		1.5 M K ₂ CO ₃ + 1 M TETA	
P _{CO₂} (kPa)	α	P _{CO₂} (kPa)	α	P _{CO₂} (kPa)	α	P _{CO₂} (kPa)	α
T = 303.15 K							
05.31	0.805	03.05	0.840	03.27	0.901	02.48	0.950
12.04	0.841	06.55	0.866	10.44	0.951	06.65	0.981
17.03	0.873	14.47	0.911	16.99	0.980	11.28	1.015
22.88	0.890	23.02	0.941	20.37	0.991	19.03	1.043
27.29	0.901	28.67	0.952	28.26	1.009	27.37	1.052
T = 313.15 K							
04.95	0.739	04.83	0.801	05.19	0.863	03.35	0.898
09.44	0.769	10.93	0.853	08.21	0.890	07.02	0.934
14.27	0.793	16.05	0.870	13.04	0.908	15.99	0.978
21.32	0.818	20.85	0.889	19.27	0.933	22.52	1.001
28.44	0.842	27.33	0.910	26.25	0.961	28.04	1.011
T = 323.15 K							
04.37	0.673	03.08	0.740	05.14	0.810	04.37	0.870
08.26	0.702	09.94	0.787	09.95	0.840	09.82	0.902
13.05	0.744	17.33	0.822	15.51	0.859	14.09	0.927
19.74	0.763	23.05	0.845	21.37	0.882	20.03	0.951
26.38	0.791	26.69	0.852	26.62	0.901	26.35	0.962

Uncertainties; U (α) = ± 0.001 ; U (T) = ± 0.01 K; U (M): 0.001 mol/l; U (P) = ± 0.1 kPa.

**Fig. (6).** Loading capacity of CO₂ in K₂CO₃+TETA solution at 323.15 K.

In Fig. (7), the solubility of CO₂ in K₂CO₃+TETA solution measured in this work was compared with the solubility of CO₂ in some solvents at 313.15 K. It can be found that the blend of K₂CO₃+TETA has the highest CO₂ loading when compared to MEA and other solvents. This result confirms the high performance of this solvent for CO₂ capture.

4.2. Correlation of Experimental Data

Response surface methodology (RSM) is an efficient method to obtain the sensitivity analysis of a process and optimum condition of a multivariable system [47]. In addition, RSM can be applied for investigation of effect of factors on response, and the relationship between response variables [48]. In this work, the response surface methodology was used to correlate the CO₂ solubility experimental data (α) as a function of temperature (T), pressure (P_{CO₂}) and TETA mole fraction (R) in the solution of potassium carbonate promoted by TETA. Three parameters including pressure, promoter mole fraction and temperature were selected as independent variables, and the validity of the model was

evaluated using analysis of variance ANOVA on response parameter (CO_2 solubility). The effect of these parameters on the CO_2 solubility was studied. The results showed that mole fraction of TETA has the highest impact on CO_2 solubility, while the interaction of pressure and temperature was found to be the least important parameter. In order to check the accuracy of the obtained model, F-value, R-squared and adjusted R-squared were determined using ANOVA analysis. The R-squared and adjusted R-squared for this system have been found to be 0.986 and 0.981, respectively. These high values of R^2 and R^2_{adj} show the significance of model terms. The F-value of the reduced quadratic model was also found to be 85.42 which indicates the validity of model.

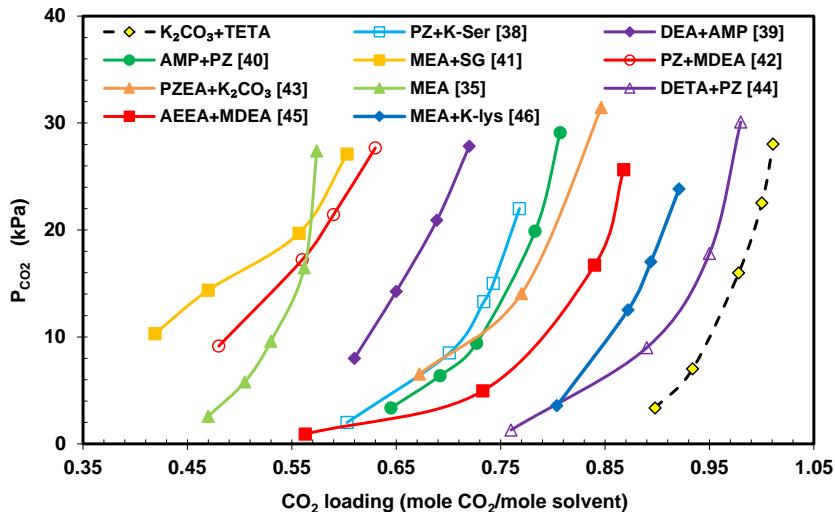


Fig. (7). Loading capacity of CO_2 in $\text{K}_2\text{CO}_3+\text{TETA}$ solution and other solvents at 313.15 K.

$$\alpha = 5.7308 - 0.0261 \times T - 0.0076 \times P + 0.3708 \times R + 4.8684 \times 10^{-5} \times T \times P + 7.5 \times 10^{-4} \times T \times R - 5.3947 \times 10^{-3} \times P \times R + 3.1466 \times 10^{-5} \times T^2 - 5.0457 \times 10^{-5} \times P^2 + 0.0837 \times R^2 \quad (13)$$

The predicted CO_2 absorption capacity by eq. (13) were plotted against experimental data obtained in this work in Fig. (8). It can be clearly seen that the good agreement exists between the values calculated by the eq. (13) for the absorption capacity of CO_2 and the experimental results with the average absolute deviation percent (AAD) equal to 1.19%.

$$\% \text{AAD} = 100 \times \frac{1}{n} \sum_{i=1}^n \left| \frac{(\alpha_{\text{CO}_2})_{\text{exp}} - (\alpha_{\text{CO}_2})_{\text{cor}}}{(\alpha_{\text{CO}_2})} \right| \quad (14)$$

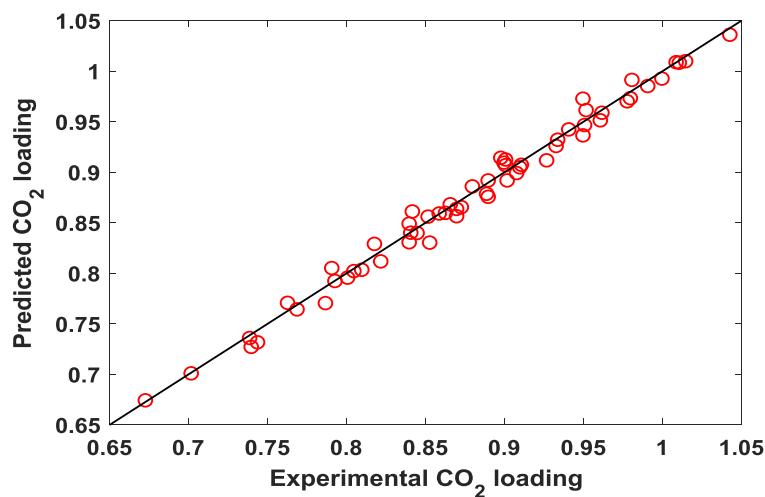


Fig. (8). Comparison of predicted CO_2 solubility using the eq. (13) with experimental data.

4.3. CO₂ Absorption Rate in K₂CO₃+TETA Solution

As previously mentioned, the main challenge of potassium carbonate is its slow rate with carbon dioxide, which leads to the larger absorber size. In this work, TETA was added as a promoter to potassium carbonate in order to enhance the rate of absorption. To check the setup and experimental method, the pressure decay during CO₂ absorption in solutions of pure MEA, TETA and K₂CO₃ at 313.15 K was presented in Fig. (9) and compared with literature data [26, 29]. A good agreement was found between results obtained in this work and the literature. As expected, the absorption rate in TETA solution is much higher than MEA and K₂CO₃ because of its structure feature.

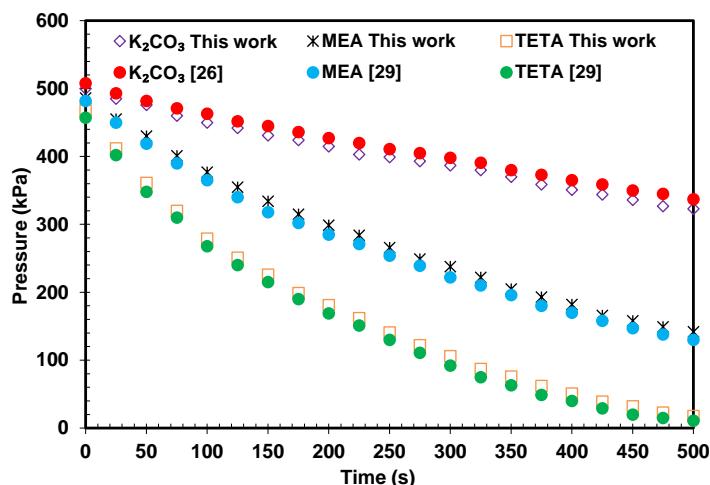


Fig. (9). Comparison of obtained pressure decay in this work with the values reported in the literature at 313.15 K.

The profile of CO₂ partial pressure versus time for the solution of K₂CO₃+TETA at different concentrations and temperatures were illustrated in Figs. (10-12) to investigate the effect of the addition of TETA to potassium carbonate on the absorption rate. The initial slope of the pressure curves over time is an indication of CO₂ absorption rate. It was found that the CO₂ partial pressure decreases with increasing time until a gas-liquid equilibrium is reached. The time required to obtain an equilibrium state is different for each solutions. This pressure reduction in the equilibrium cell shows the amount of gas absorption. According to Figs. (10-12), the addition of small amount of TETA to potassium carbonate has been led to a significant increase in the absorption rate that is favorable for the CO₂ capture process. The structure feature of TETA with two primary amino groups and two secondary amino sites, and also an increase in the rate of reaction of CO₂ with OH⁻ due to increase of PH of solution can be applied to explain this rate enhancement. It also can be observed from Fig. (11) that the mixed absorbent has higher absorption rate compared to single potassium carbonate. This result proves the slow kinetics between potassium carbonate and CO₂.

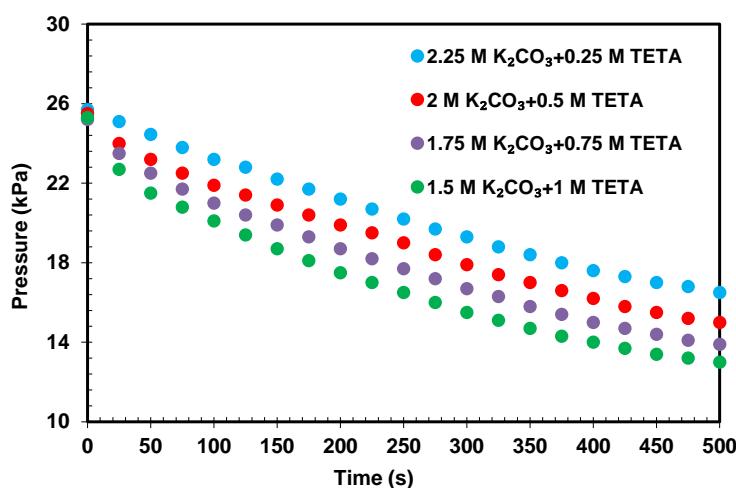


Fig. (10). The effect of addition of TETA to K₂CO₃ on the CO₂ absorption rate at 303.15 K.

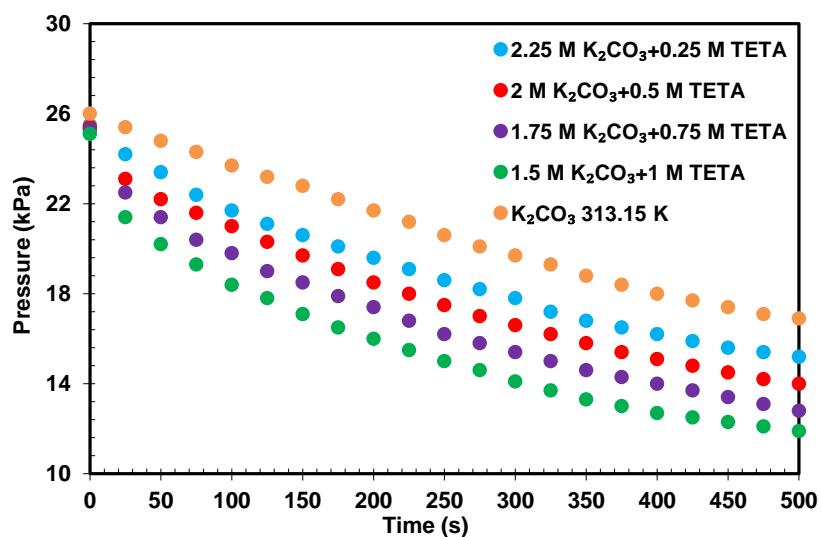


Fig. (11). The effect of addition of TETA to K_2CO_3 on the CO_2 absorption rate at 313.15 K.

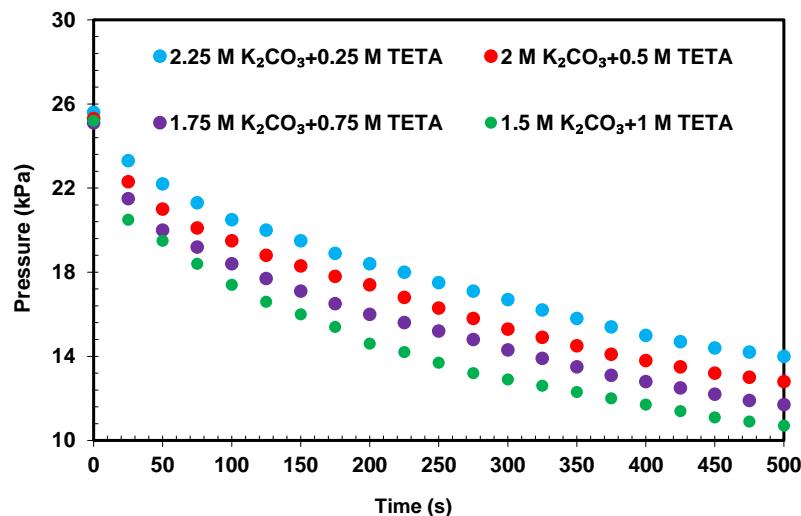


Fig. (12). The effect of addition of TETA to K_2CO_3 on the CO_2 absorption rate at 323.15 K.

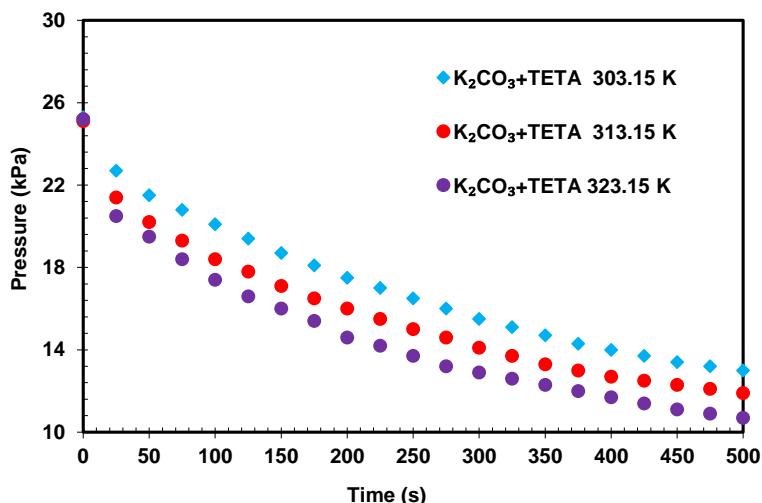


Fig. (13). The impact of temperature on the CO_2 absorption rate in solution of 1.5 M $\text{K}_2\text{CO}_3+1 \text{ M TETA}$.

The influence of the temperature on the initial absorption rate in solution of K_2CO_3 +TETA was investigated and shown in Fig. (13). As expected, higher temperature resulted in an increase in reactivity of CO_2 with solution of K_2CO_3 +TETA. The obtained results shows that TETA has a good ability as a promoter for potassium carbonate and TETA+ K_2CO_3 solution could be an appropriate solvent for CO_2 capture because of its high absorption rate and capacity.

CONCLUSION

In this work, CO_2 solubility and initial absorption rate in K_2CO_3 +TETA solution were investigated at temperatures 303-323 K and the partial pressures up to 30 kPa using a stirred cell reactor. Obtained results revealed that CO_2 absorption capacity of K_2CO_3 +TETA solution increase with the increase in TETA mole fraction, and decreased with the increase in temperature. The results also indicated that the K_2CO_3 +TETA solution has absorption capacity higher than MEA. A correlation was presented to predict CO_2 loading capacity in solution, and predicted values were found in excellent agreement with the experimental results. Therefore, considering these results, K_2CO_3 +TETA solution can be used as an alternative solvent to MEA. However, in order to make better judgment about this solvent, more investigation needs to be performed to determine the corrosion rate and absorption heat of K_2CO_3 +TETA solution.

NOMENCLATURE

AEEA	=	2-((2-aminoethyl)amino)ethanol
MDEA	=	Methyldiethanolamine
K-lys	=	potassium lysinate
K_2CO_3	=	Potassium carbonate
HomoPZ	=	Homopiperazine
PZEA	=	2-(1-piperazinyl)-ethylamine
DEA	=	Diethanolamine
K-Ser	=	Potassium serinate
TETA	=	Triethylenetetramine
AMP	=	2-Amino-2-methyl-1-propanol
DETA	=	Diethylenetriamine
MEA	=	Monoethanolamine
PZ	=	Piperazine
SG	=	Sodium glycinate
P_v	=	Vapor pressure of solution
$P_{\text{CO}_2}^e$	=	The equilibrium partial pressure of CO_2
n_{solvent}	=	The moles of solvent in liquid phase
P_f	=	Final pressure of reactor
$a:$	=	CO_2 solubility (mole CO_2 /mole of absorbent)
V	=	V Volume of reactor
V_s	=	Volume of gas storage tank

CONSENT FOR PUBLICATION

Not applicable.

CONFLICT OF INTEREST

The author (editor) declares no conflict of interest, financial or otherwise.

ACKNOWLEDGEMENTS

Decleared none.

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