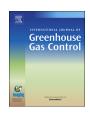
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Comparative kinetics of carbon dioxide (CO₂) absorption into EAE, 1DMA2P and their blends in aqueous solution using the stopped-flow technique



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Keywords: CO₂ capture EAE/1DMA2P Kinetic model Mechanism ABSTRACT

The amine blends as the potential carbon dioxide ($\rm CO_2$) capture absorbents have been received much attention in recent years, but this amines interaction mechanism remains unclear. In the present work, the kinetics of $\rm CO_2$ absorption into aqueous solutions of a secondary amine (Ethylethanolamine, EAE), a tertiary amine (1-Dimethylamino-2-propanol, 1DMA2P) and their blend were investigated by using a stopped-flow setup. The kinetic data of aqueous EAE solution can be interpreted well by the zwitterion mechanism, and the reaction rate of 1DMA2P can be fitted well using the base-catalyzed hydration mechanism. Experimental results showed that the pseudo first-order rate constants (k_0) of EAE/1DMA2P is much higher than the calculated values of individual EAE and 1DMA2P using the parallel reaction mechanism. It has been accepted that the tertiary amine (i.e. 1DMA2P) can attract the $\rm CO_2$ from the zwitterion to release the free EAE molecules or the 1DMA2P as a base join the reaction of the $\rm CO_2$ and EAE. Then, the reaction pathway was proposed and the kinetic model with acceptable AARD% values of 6.642 was developed. Therefore, it can be concluded that the addition of EAE into 1DMA2P with low energy consumption can importantly enhance the absorption performance to solve the contradiction between low absorption heats and high $\rm CO_2$ absorption rates.

1. Introduction

Post-combustion carbon dioxide (CO2) capture (PCC) by chemical absorption using alkanolamines has been considered to be one of the economic technologies for potential carbon dioxide (CO₂) removal from power plant flue gases for environmental reasons (Rao and Rubin, 2002; da Silva and Booth, 2013). Alkanolamines, such as monoethanolamine (MEA), diethanolamine (DEA), and methyldiethanolamine (MDEA), which are typical primary, secondary and tertiary amines, have been applied in industrial application for a number of years (Svendsen et al., 2011; Liang et al., 2015c). However, investigated individual alkanolamines encounter a big challenge of contradiction between fast kinetics and low energy consumption for solvent regeneration (Liang et al., 2015b; Manzolini et al., 2015; Chen et al., 2018). Amine blends including primary/secondary amines (carbamate formers) and tertiary amines (preferential bicarbonate producers) have attracted many attention and been considered to be the potential or superior absorbents in the development of PCC technology (Idem et al., 2006; Ling et al., 2019).

Recently, 1-Dimethylamino-2-propanol (1DMA2P), a new tertiary amine, has received much attention for its better CO₂ capture

performance (Kadiwala et al., 2012; Zhang et al., 2016; Liu et al., 2017). Liu et al. (2017) reported that the equilibrium CO₂ solubility of 1DMA2P was competitive with that of AMP and the absorption heat of 1DMA2P (-31.67 kJ/mol) was lower than the values of MDEA (-54.6 kJ/mol). Kadiwala et al.(2012) illustrated that 1DMA2P exhibited higher CO₂ absorption kinetics than that of MDEA. Liang et al.(2015a) investigated the solubility, absorption heat and mass transfer of CO2 absorption into aqueous 1DMA2P solution, and they also found that 1DMA2P showed better mass transfer performance and lower absorption heat. However, 1DMA2P has a low CO2 absorption kinetics due to the characteristics of tertiary, but the introduction of rate activator could improve the CO2 absorption rate. Secondary amines have attracted lots of attentions due to the larger CO₂ capacity, lower energy requirement for solvent regeneration and comparative CO2 absorption rate compared to those of primary amines. Recently, N-Ethylethanolamine (EAE), which can be obtained from renewable resources such as agriculture products orresidues, represents a potential absorbent for CO2 removal. Li et al.(2007) measured the CO2 absorption rateinto aqueous EAE solution by using a stopped-flow technique, indicating a higher CO2 absorption kinetics compared with DEA. Also, Vaidya and Kenig (2007) found that the addition of EAE can significantly enhance

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the absorption kinetics of N,N-diethylethanolamine (DEEA) solution, showing EAE is a better effective activator. Additionally, it also has been reported that EAE has a smaller energy consumption, a lower corrosion rate and a higher pKa value than those of MEA by Bhosale et al. (2015). The above investigations indicated aqueous 1DMA2P and EAE solutions are both promising alternative absorbents for ${\rm CO_2}$ capture.

Owing to the growing concern and potential CO2 capture performance of blended amines, sound fundamental understanding of the CO₂ absorption kinetics, mass transfer and reaction heat is an important step towards developing absorbents with fast kinetics, high capacity and low energy consumption for solvent regeneration, Recently, Kierzkowska-Pawlak (2015a) studied the kinetics of a CO2 reaction into DEEA solution activated with N-(2-aminoethyl)ethanolamine (AEEA) at 303 K using a stirred cell reactor and found that the experimental kinetics was under pseudo-first-order regime. Xu et al. (2013) investigated the CO₂ absorption kinetics into aqueous solution of 1,4-butanediamine (BDA) and DEEA and concluded that overall reaction of blended BDA/DEEA solution can be regarded as a reaction between DEEA and CO2 in parallel with the reaction of BDA and CO2. Also, Konduru et al. 10 reported the similar observation by using DEEA and piperazine (PZ) mixtures. Wang et al. (2017) investigated the kinetics of CO₂ absorption into PZ/ AEEA solution in a laboratory wetted wall column (WWC), whereas Sema et al.(2012) measured the absorption rate of CO2 in to the blended MDEA/MEA solution by using a laminar jet absorber. In addition, the CO2 absorption kinetics of blended amine solution such as MDEA/AMP, MEA/AMP, MEA/DEA, DEEA/1,6-hexamethyl diamine (HMDA), DEEA/N,N-dimethylbutylamine (DMBA) and DEEA/EAE have also been investigated (Sakwattanapong et al., 2009; Sutar et al., 2013; Xiao et al., 2017). Therefore, a comprehensive study on the CO₂ absorption kinetics appears essential and desirable.

It should be noted that a precise knowledge of the reaction mechanism for the CO₂ absorption into amine solution is also essential. The zwitterion mechanism was originally proposed by Caplow (1968), and the single step termolecular mechanism reported by Crooks and Donnellan (1989) and represented by da Silva and Svendsen (2006) have been accepted and applied by many researchers (Li et al., 2007; Ma'mun et al., 2007a; Ma'Mun et al., 2007b; Henni et al., 2008a; Bougie and Iliuta, 2009; Zheng et al., 2013; Chen et al., 2017; Wang et al., 2017). However, Donaldson and Nguyen (1980) reported that the tertiary amines cannot directly react with CO2 and acted as a base can catalyze the hydration of CO2 to produce bicarbonates, and then the base-catalyzed hydration mechanism was initially proposed to explain the CO₂ absorption process. Subsequently, Blauwhoff et al.(1983), Barth et al.(1982), and Blauwhoff and Swaaij (1985) also validated this mechanism using TEA and MDEA solution. Additionally, the base-catalyzed hydration mechanism has been widely used for the description of the CO2 absorption into tertiary amines, such as DEEA (Sutar et al., 2013), DMEA (Jiang et al., 2018), 1DMA2P (Kadiwala et al., 2012) and 1DEA2P (Liu et al., 2014a). For the blended amines, there is no uniform reaction to describe the CO2 absorption into the blended amines. Ramachandran et al.(2006) investigated the kinetics of absorption of CO₂ in loaded MDEA and MEA solutions, which can be explained by the modified termolecular mechanism with consideration of the contribution of hydroxide ions. Dubois and Thomas (Dubois and Thomas, 2009) reported the CO2 absorption into mixed MEA/MDEA/PZ system without clear mechanism, and the experimental results showed that PZ was a very positive activator to enhance the absorption performance. Later, the Kinetic data of aqueous mixtures of DEEA and PZ were experimentally by using a stirred cell reactor, and were interpreted using a homogeneous activation mechanism by the investigated reaction with consideration of a reaction of CO2 with PZ parallel with the reaction between CO₂ and DEEA(base-catalyzed hydration mechanism). Subsequently, kinetics of CO2 absorption with aqueous BDA blended with DEEA using a wetted wall column was studied by Xu et al. (2013), and the overall reaction can be regarded as a reaction between CO2 and

DEEA in parallel with the reaction of CO₂ with BDA (both using the single step termolecular mechanism) with the interaction reaction rate (r_{CO2-int}) of zero. The similar result has been reported by Kierzkowska-Pawlak (2015b), who investigated the kinetics of CO₂ absorption into DEEA/AEEA solution. In the work of Kierzkowska-Pawlak and Kruszczak (Kierzkowska-Pawlak and Kruszczak, 2017), the CO₂ absorption kinetics of N-methyl-1,3-propane-diamine (MAPA)/DEEA is also interpreted using the a reaction between CO₂ and MAPA (single step termolecular mechanism) in parallel with the reaction of CO₂ with DEEA base-catalyzed hydration mechanism, and the MAPA can act as an effective activator in DEEA solutions. Thus, the reaction mechanism of EAE and 1DMA2P is also important to be comprehensively studied.

There is no literature reporting the effect of adding EAE into aqueous 1DMA2P solution. This work focuses on the kinetic data and mechanism of the $\rm CO_2$ reaction with 1DMA2P, EAE and their mixtures in order to understand the mechanism of $\rm CO_2$ absorption in aqueous 1DMA2P/EAE solution. In the present work, the $\rm CO_2$ absorption kinetic data of $\rm 20-80$ mol/m³EAE, $\rm 100-200$ mol/m³ 1DMA2P and their mixtures was determined by using the stopped-flow apparatus. The experimental kinetic data of $\rm CO_2$ absorption into EAE, 1DMA2P and EAE/1DMA2P were then evaluated using the existing and developed models mechanism and interpreted corresponding mechanism.

2. Reaction mechanisms and kinetics models

2.1. EAE-CO₂-H₂O system

The zwitterion mechanism, originally proposed by Caplow(1968) using a number of amines, is widely accepted for the description of the reaction between primary/secondary amines and CO_2 . The CO_2 absorption into aqueous EAE solution explained by zwitterion mechanism includes two steps: the zwitterion formation and zwitterion deprotonation (i.e. carbamate formation), as presented below:

Zwitterion formation:

$$RR'NH + CO_2 \stackrel{(k_2 - EAE)(k-1)}{\leftrightarrow} RR'N^+HCO_2^-$$
 (1)

Deprotonation of zwitterion by a base B:

$$RR^{'}N^{+}HCO_{2}^{-} + B^{\overset{(k_{Base})(k_{-Base})}{\longleftrightarrow}}RR^{'}NCO_{2}^{-} + BH^{+}$$
(2)

Applying the pseudo-steady state principle for the intermediate zwitterion, the overall forward reaction rate of ${\rm CO_2}$ in aqueous EAE solution can be written as:

$$r_{\text{CO}_2-\text{EAE}} = k_{0-\text{EAE}}[\text{CO}_2] = \frac{[\text{EAE}][\text{CO}_2]}{1/k_{2-\text{EAE}} + k_{-1}/(k_{2-\text{EAE}}(\sum k_{Z-\text{Base}}[\text{B}])}$$
 (3)

Where

$$k_{Z-B} = \frac{k_{Z-EAE}k_{Z-Base}}{k_{-1}} \tag{4}$$

Then, the Eq. (3) can be expressed as:

$$r_{\text{CO}_2-\text{EAE}} = \frac{k_{2-\text{EAE}}[\text{EAE}][\text{CO}_2]}{1 + k_{2-\text{EAE}}/(k_{Z-\text{EAE}}[\text{EAE}] + k_{Z-\text{H}_2\text{O}}[\text{H}_2\text{O}])}$$
(5)

2.2. 1DMA2P-CO₂-H₂O system

Donaldson and Nguyen(1980) firstly proposed that the tertiary amines cannot directly react with CO_2 and act a base can catalyze the hydration of CO_2 to produce bicarbonates. Then, the CO_2 absorption into 1DMA2P can be explained by the follow representative reactions:

$$R_2R'N+CO_2 + H_2 \stackrel{k_2}{O} \leftrightarrow R_2R'NH^+ + HCO_3^-$$
 (6)

In addition, other reactions also occur simultaneously in the aqueous 1DMA2P solutions:

$$CO_2 + H_2 O \leftrightarrow H^+ + HCO_3^-$$
 (7)

$$CO_2 + OH^- \leftrightarrow HCO_3^-$$
 (8)

$$R_2R^{'}N+H^+\leftrightarrow R_2R^{'}NH^+ \tag{9}$$

$$HCO_3^- \leftrightarrow H^+ + CO_3^{2-} \tag{10}$$

$$H_2 O \leftrightarrow H^+ + OH^- \tag{11}$$

The totalreaction rate of CO₂ absorption into aqueous 1DMA2P solution by applying the base-catalyzed hydration mechanism (Donaldson and Nguyen, 1980) can be expressed as:

$$r_{\text{CO}_2-1\text{DMA2P}} = k_{0-1\text{DMA2P}} [\text{CO}_2] = (k_{2-1\text{DMA2P}} [\text{1DMA2P}] + k_{B-OH} [\text{OH}^-] + k_{B-H_2O} [\text{H}_2O]) [\text{CO}_2]$$
 (12)

It has been mentioned above that the OH^- concentration can be ignored in the amine solution. Additionally, the term of $k_{B-H2O}[H_2O]$ can be neglected due to the extremely slow reaction rate of CO_2 with H_2O in Eq. (6). Then, the Eq. (12) can be simplified as:

$$r_{\text{CO}_2-1\text{DMA2P}} = k_{0-1\text{DMA2P}}[\text{CO}_2] = k_{2-1\text{DMA2P}}[1\text{DMA2P}][\text{CO}_2]$$
 (13)

2.3. EAE-1DMA2P-CO₂-H₂O system

Based on the investigations in the literatures (Kierzkowska-Pawlak, 2015a), the $\rm CO_2$ absorption kinetics of secondary amine is much faster than that of tertiary amine. If the 1DMA2P solution can contribute to the $\rm CO_2$ absorption into aqueous blended EAE/1DMA2P solution, the $\rm CO_2$ absorption into aqueous EAE/1DMA2P solution can be considered that the reaction of $\rm CO_2$ and EAE is in parallel with the $\rm CO_2$ reaction between 1DMA2P.

Based on the experimental results, the absorption rate of aqueous EAE/1DMA2P solution is much higher than the simple sum of individual 1DMA2P and EAE solutions. Vaidya and Kenig and other researchers (Vaidya and Kenig, 2009; Konduru et al., 2010) also reported that tertiary amine can attract the $\rm CO_2$ from the zwitterion due to the higher electronegativity of N atom attached to tertiary amine. Then, the $\rm CO_2$ may be transferred from EAE zwitterion to 1DMA2P and the zwitterion-type complex could be formed, as presented:

$$RR'N^{+}HCO_{2}^{-} + R_{2}R'N \leftrightarrow R_{2}R'NCO_{2}^{-} + RR'NH$$
 (14)

The $R_2R'N$ in this reaction can be regarded as the base B in the Eq. (6). The complex of $R_2R'NCO_2^-$ is an unstable intermediate and the homogeneous hydrolysis reaction of water and the zwitterion-type complex could occur to form bicarbonate (Yu et al., 1985).

$$R_2R'NCO_2^- + H_2 O \leftrightarrow R_2R'NH^+ + HCO_3^-$$
 (15)

Therefore, the total CO_2 reaction rate in the formulated amine solution, by combining Eq. (5) and Eq. (13), is can be written as:

$$r_{\text{CO}_2-1\text{DMA2P/EAE}} = k_{0-1\text{DMA2P/EAE}}[\text{CO}_2]$$

$$= \frac{k_{2-\text{EAE}}^A[\text{EAE}][\text{CO}_2]}{1 + k_{2-\text{EAE}}^A/(k_{Z-\text{EAE}}^A[\text{EAE}] + k_{Z-1\text{DMA2P}}^A[\text{1DMA2P}])} + k_{2-1\text{DMA2P}}^A[1\text{DMA2P}][\text{CO}_2]$$
(16)

3. Experimental section

3.1. Materials

1-Dimethylamino-2-propanol (1DMA2P) with a purity of $\geq 98~\%$ was obtained from TCI (Shanghai) Development Co., Ltd. China, and Ethylethanolamine (EAE) with a purity of $\geq 99~\%$ was supplied by Thermo Fisher Scientific Co., Ltd. China. Commercial grade CO $_2$ and air ($\geq 99.9~\%$) was purchased from Changsha Rizhen Gas Co, China. All materials were used without further purification

 ${\rm CO_2}$ solution was prepared by bubbling the commercial grade ${\rm CO_2}$ through water in a glass reactor for at least half an hour. It should be noted that the amine concentration was at least 10 times more than the ${\rm CO_2}$ concentration which was diluted by using water in the aqueous solution to satisfy the pseudo first-order conditions. The amine solutions with desired concentration were prepared by diluting the reagent grade amines using water.

3.2. Measurement of kinetics data

The stopped-flow apparatus is applied for the direct determination of the pseudo first-order kinetics data of aqueous EAE, 1DMA2P and EAE/1DMA2P solutions. The stopped-flow setup (model SF-61DX) was manufactured by Hi-Tech Scientific Ltd. (U.K.). It includes four main parts: a conductivity-detection cell, a microprocessor, a sample handling unit, and an A/D converter. A water bath (HANUO, HX20 Shanghai Hanuo Instruments Co., Ltd., China) is used to control the system temperature with \pm 0.1 K by heating the sample-flow circuits. Two internal syringes loaded with saturated CO2 solution and amine solution were pushed by the pneumatically air supply pushes to ensure the adequate mixing of these two solutions. The observed pseudo first-order reaction rate constant is obtained from the fitted exponential equation by using the "Kinetasyst" software. A detailed description of the stopped-flow apparatus and the operational procedure can be found in Henni et al. (2008a) and Liu et al. (2014a). Experiment was repeated ten times at desired concentration and temperature for each sample.

4. Results and discussion

4.1. CO₂ absorption kinetics of EAE

The reaction kinetics of CO₂ absorption into aqueous EAE solutions were determined using stopped-flow apparatus over a range of concentrations from 20 to 80 mol/m³ at various temperatures ranging from 293.15 K-313.15 K. The observed pseudo first-order rate constants (k_0) obtained from this work were also compared to those of Li et al.(2007), as presented in Fig. 1. It can be found that there is a good agreement between the k_0 values obtained from this work and those from the literature with an AARD of 7.6 %, which is in an acceptable range of less than 10 %. As shown in Fig. 1, both the EAE concentration and temperature have significant influences on the observed pseudo first-order rate constants (k_0) as expected. In addition, the approximate reaction order can be fitted by power law relationship from the effect of EAE concentration and temperature on the observed pseudo first-order rate constants (k_0) for each temperature. The reaction orders were 1.987, 1.985, 2.024, 1.971 and 1.958 for 293.15 K, 298.15 K, 303.15 K, 308.15 K and 313.15 K in terms of EAE concentration in unit of mol/L. The reaction between CO2 and EAE can be explained by the zwitterion

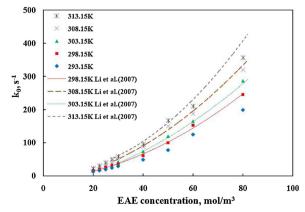


Fig. 1. The k_0 of EAE as function of concentration at different temperatures.

 Table 1

 Kinetics constants obtained from zwitterion mechanism.

Т, К	$k_{2\text{-EAE}}$, m ³ /(kmol·s)	$k_{Z\text{-EAE}}$, m ⁶ /(kmol ² ·s)
293.15 298.15 303.15 308.15 313.15	8.655×10^{3} 1.018×10^{4} 1.354×10^{4} 1.652×10^{4} 1.991×10^{4}	3.631×10^{4} 4.571×10^{4} 5.295×10^{4} 6.317×10^{4} 7.053×10^{4}

mechanism reported by the researchers. Then, the observed pseudo first-order rate constants (k_0) could be described by Eq. (5) to evaluate the accuracy of the model. The influence of $k_{H2O}[{\rm H_2O}]$ can be neglected in the present work. Table 1 presents the kinetics constants (i.e. $k_{2\text{-EAE}}$ and $k_{Z\text{-EAE}}$) obtained from the above two mechanisms using *Microsoft Excel solver tool function*. It can be clearly observed that the kinetics constants increase with the increasing temperature, and can be fitted to the Arrhenius expression (Eq. (17)).

$$k = Aexp(-E_a/RT) (17)$$

where A represents the pre-exponential factor, E_a and R are the activation energy and universal gas constant, respectively. Then, the $k_{2\text{-EAE}}$ and $k_{Z\text{-EAE}}$ can be expressed as:

$$k_{2-\text{EAE}} = 5.970 \times 10^9 \exp(-3947/T)$$
 (18)

$$k_{\rm Z-EAE} = 1.181 \times 10^9 \exp(-3037/T)$$
 (19)

Based on the Eqs. (19) and (20), the activation energy values are equal to 32.81 and 25.25 kJ/mol, and the activation energy in terms of $k_{2\text{-EAE}}$ is very close to the value of 32.65 kJ/mol reported by Li et al. (2007).To compare the contribution of $k_{2\text{-EAE}}$ and $k_{Z\text{-EAE}}$ on the pseudo first-order rate constants (k_0), both rate constants cannot be negligible in the CO₂ absorption process. Also, it is clear that the both the zwitterion formation step and the zwitterion deprotonation step can affect the total CO₂ reaction rate.

In addition, the observed pseudo first-order rate constants (k_0) were predicted according to the zwitterion mechanism (Eq. (5)), with the combination of Eqs. (18) and (19). Fig. 2 shows a comparison of the experimental pseudo first-order rate constants (k_0) with the calculated data from Eq. (5), resulting in the acceptable average absolute relative deviation (AARD%) of 6.073. The calculated values of the pseudo first-order rate constant (k_0) obtained from zwitterion mechanism show the exactly results. Therefore, it can be concluded that the CO₂ absorption into aqueous EAE can be well explained by the zwitterion mechanism.

It is essential to comprehensively evaluate the EAE as a potential alternative absorbent by comparing its CO₂ absorption performance with MEA and other secondary amines (DEA, MAE, 1A2P and 3A1P).

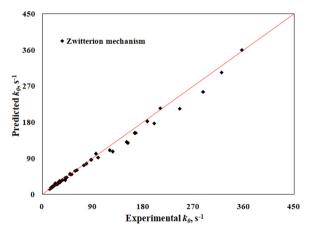


Fig. 2. Crossplot between experimental and predicted $k_{\rm 0}$ values for aqueous EAE solution.

Table 2 Second order reaction rate constant (k_2) and dissociation constant (pKa) of MEA, DEA, MAE, EAE, 1A2P and 3A1P at 298 K.

Amine	k_2 , m ³ ·mol ⁻¹ ·s ⁻¹	рКа
MEA DEA MAE EAE 1A2P	5.573 (Ali, 2005) 0.815 (Ali, 2005) 5.381 (Ali et al., 2002) 10.18 4.314 (Henni et al., 2008b)	9.45 (Hamborg and Versteeg, 2009) 8.88 (Hamborg and Versteeg, 2009) 9.93 (Goto et al., 2014) 9.99 (Goto et al., 2014) 9.46 (Goto et al., 2014)
EAE	10.18	9.99 (Goto et al., 2014)

Then, this comparison and evaluation was presented in terms of the second order reaction rate constant (k_2) and dissociation constant (pKa). As expected, it can be seen that the second order reaction rate constants (k_2) increase with the increasing pKa values. By linearly plotting the $\log_{10}(k_2)$ and pKa, the Brønsted line for DEA, MAE, EAE, 1A2P and 3A1P has showing R² of 0.924, which is acceptable due to the existence of steric hindrance (Liu et al., 2019b). Based on the Table 2, it can be found that EAE with high absorption rate and equilibrium CO₂ solubility (Liu et al., 2019a) is a promising absorbent.

4.2. CO₂ absorption kinetics of 1DMA2P

The CO_2 absorption kinetics into aqueous 1DMA2P solutions was also studied using the stopped-flow technique over temperature range of 293.15 K–313.15 K and 1DMA2P concentration range of 100–200 mol/m³. Fig. 3 demonstrates the observed pseudo first-order rate constants (k_0) plotted against 1DMA2P concentration at different temperatures. It can be observed that the observed pseudo first-order rate constant (k_0) increases with the increasing 1DMA2P concentration as well as the system temperature. In addition, these increases are more significant at both high concentration and temperature.

The reaction orders with respect to aqueous 1DMA2P solution obtained by fitting the k_0 values using a power law equation were between 0.74 and 0.87 over the temperature range of 293.15–313.15 K in terms of 1DMA2P concentration in unit of mol/L. Based on the base-catalyzed hydration mechanism, the second order rate constant ($k_{2\text{-}1DMA2P}$) values were therefore fitted to Eq. (16) and tabulated in Table 3. The plotted $\ln k_2$ value versus 1/T was linear using Arrhenius equation (Eq. (20)), and the temperature dependency of the Arrhenius equation for 1DMA2P-CO₂-H₂O can be given as:

$$k_{2-1DMA2P} = 3.065 \times 10^{12} exp(-7557/T)$$
 (20)

The activation energy (E_a) for the base-catalyzed hydration mechanism was calculated to be 62.83 kJ/mol, which was close to 62.55 kJ/mol reported by Kadiwala et al.(2012) for 1DMA2P in aqueous solution. Furthermore, the second order rate constant $(k_{2\cdot 1DMA2P})$ values are also compared to the values from Kadiwala et al.(2012) with an

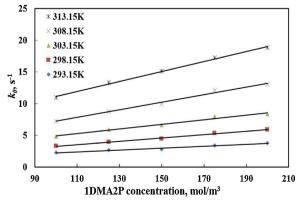


Fig. 3. Observed pseudo first-order rate constants (k_0) of 1DMA2P.

Table 3 Second order reaction rate constant (k_2) of aqueous 1DMA2P solution

Т, К	k_2 , m ³ /(kmol·s)
293.15	19.438
298.15	30.208
303.15	44.560
308.15	69.153
313.15	100.542

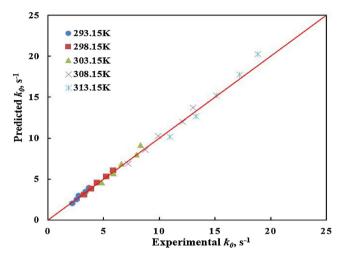


Fig. 4. Crossplot between experimental and predicted k_0 values for aqueous 1DMA2P solution.

acceptable AARD of 9.1 %. This a litter higher AARD is due to the much lower 1DMA2P concentration. Substituting Arrhenius equation (Eq. (20)) in Eq. (13) fits the experimental data for pseudo first-order rate constant (k_0), as shown in Fig. 4. The parity plot in Fig. 4 represents that the calculated values were in good agreement with the determined values with AARD% of 4.406. On the basis of the results obtained, it can be concluded that the base-catalyzed hydration mechanism can provide good prediction of the kinetics data in the tested amine concentration range.

Furthermore, the second order rate constant $(k_{2\text{-}1DMA2P})$ at 298 K, pKa value at 298 K as well as the CO_2 absorption heat $(\triangle \mathrm{H}_{\mathrm{abs}})$ of 1DMA2P was compared with some investigated tertiary amines reported in the literature, as tabulated in Table 4A. It can be seen that the 1DMA2P shows a relative high second order rate constant $(k_{2\text{-}1DMA2P})$ at 298 K as well as the pKa value, and especially presents a lowest CO_2 absorption heat $(\triangle \mathrm{H}_{\mathrm{abs}})$, indicating the low energy requirement for solvent regeneration. Thus, the EAE with higher absorption rate and 1DMA2P with low CO_2 absorption heat was blended for the evaluation.

4.3. CO_2 absorption kinetics of EAE/1DMA2P solution

The effect of the addition of EAE into aqueous 1DMA2P solutions as a potential activator was investigated in terms of CO_2 absorption rate using the stopped-flow technique. Fig. 5 illustrates the changes in the

pseudo first-order rate constant (k_0) in relation to both the EAE concentration $(10-20 \text{ mol/m}^3)$ and 1DMA2P concentration $(100-200 \text{ mol/m}^3)$ at different temperature. It is obvious that the increase in EAE concentration can significantly enhance the pseudo first-order rate constant (k_0) of aqueous EAE/1DMA2P solution compared to the increase by increasing the 1DMA2P concentration, due to the translation of CO₂ from EAE zwitterion to 1DMA2P with the release of free active MEA molecules.

The values of the pseudo first-order rate constants (k_0) for 100, 125, 150, 175 and 200 mol/m³ 1DMA2P with variation of the EAE concentration from 10 to 20 mol/m³were also correlated using the empirical power law relationship to calculated the reaction orders. For example, the effects of EAE concentration on the pseudo first-order rate constants (k_0) of 125 mol/m³ 1DMA2P solution are presented in Fig. 6. The determined reaction orders are the number between 0.91 and 1.55 for 25 lines for the fixed concentration of 1DMA2P solution (100 - 200 mol/m³), and those are between 0.11 and 0.59 for the fixed concentration of EAE solution (10-20 mol/m³). It can be concluded that the EAE is the main contributor to the absorption rate of the blended EAE/1DMA2P solution. Then, the experimental absorption rate data were interpreted and fitted with the proposed mechanisms and kinetic model (Eq. (16)). The generated rate constants of $k_{2-\text{EAE}}^A$, $k_{Z-\text{EAE}}^A$, $k_{Z-1DMA2P}^A$ and $k_{2-1DMA2P}^A$ are tabulated in Table 4B, and the comparison of the reaction rate constants (k) at different temperatures among EAE, 1DMA2P and blended EAE/1DMA2P solutions were also presented in Fig. 7. It is obvious that the rate constant values of $k_{2-\text{EAE}}^A$, $k_{Z-\text{EAE}}^A$ and $k_{2-1\text{DMA2P}}^{A}$ for EAE/1DMA2P solution obtained from the proposed model are much higher than those in individual aqueous EAE solution and 1DMA2P solution, respectively. This phenomenon can be explained by the fact that the tertiary amine (i.e. 1DMA2P) can attract the CO₂ from the zwitterion to release the free EAE molecules (Eqs. (14) and (15)) or the 1DMA2P as a base can participate in the CO₂ absorption into EAE solution (Eq. (2)). Thus, the total CO2 reaction rate increases significantly. In addition, due to the much lower concentration of EAE compared to that of 1DMA2P in EAE/1DMA2P solution, the $k_{2-1\text{DMA2P}}^{A}$ is enhanced significantly with the increase of the total CO₂ reaction rate, leading to the higher k2-1DMA2P of EAE/1DMA2P solution than that of 1DMA2P solution. Furthermore, the experimental pseudo first-order rate constants (k_0) of EAE/1DMA2P (m mol/L + n mol/L) is much higher than the sum of k_0 data of EAE (m mol/L) and 1DMA2P (n mol/ L). So, it can be concluded the absorption rate can be importantly improved by addition of EAE into 1DMA2P solution. Thus, EAE acts as an effective activator in aqueous 1DMA2P solution and the absorbent EAE/1DMA2P is attractive for the enhancement of CO₂ capture.

All the kinetic constants were found to be increased with the increasing temperature and then were estimated using the Arrhenius equation, and the calculated A and Ea in the Eq. (17) for various reaction kinetic constants are given in Table 4B. Additionally, the comparison of activation energy (Ea) values obtained from individual EAE and 1DMA2P, EAE/1DMA2P, MEA(Ali), MEA/DEEA and MEA/DMEA (Jiang et al., 2018) was performed in order to evaluate the behavior of EAE and 1DMA2P in the blended amine solution (Table 5). As expected, the activation energy (Ea) values for the three constants (i.e. $k_{2-\rm EAE}^A$, $k_{2-\rm EAE}^A$, and $k_{2-\rm 1DMA2P}^A$) for the EAE/1DMA2P solution became lower compared with those for the individual EAE and 1DMA2P solution, indicating the fast reactions for EAE and 1DMA2P in the blended amine

Table 4A Second order reaction rate constant (k_2) and dissociation constant (pKa) of MDEA, DMEA, DEEA, EAEADMA2P and 1DEA2P.

Amine	k_2 , m^3 ·kmol $^{-1}$ ·s $^{-1}$	pKa	-∆H _{abs} , kJ/mol
MDEA	12 (Liu et al., 2014b)	8.56 (Hamborg et al., 2007)	54.6 (Rho et al., 1997)
DMEA	27.2 (Liu et al., 2014b)	9.20 (Xiao et al., 2016)	48.6 (Xiao et al., 2016)
DEEA	79.8 (Liu et al., 2014b)	9.74 (Xiao et al., 2016)	47.0 (Xiao et al., 2016)
1DMA2P	30.2	9.20 (Xiao et al., 2016)	31.7 (Liu et al., 2017b)
1DEA2P	91.2 (Liu et al., 2014b)	9.91 (Xiao et al., 2016)	45.7 ± 3.7 (Liu et al., 2017c)

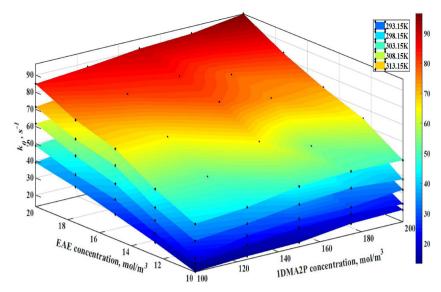


Fig. 5. Observed pseudo first-order rate constants (k_0) of 1DMA2P/EAE solution.

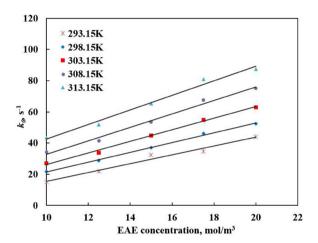


Fig. 6. The effects of EAE concentration on the observed pseudo first-order rate constants (k_0) of 125 mol/m³ 1DMA2P solution.

 ${\bf Table~4B} \\ {\bf Reaction~rate~constants~of~CO_2~absorption~into~aqueous~1DMA2P/EAE~solution~for~proposed~model.}$

Т, К	$k_{2-\text{EAE}}^{A}$ m ³ /(kmol·s)	$k_{Z-{ m EAE}}^{A}$ ${ m m}^{6}/({ m kmol}^{2}\cdot{ m s})$	$k_{Z-1\mathrm{DMA2P}}^{A}$ $\mathrm{m}^{6}/(\mathrm{kmol}^{2}\cdot\mathrm{s})$	$k_{2-1\mathrm{DMA2P}}^{A}$ m ³ /(kmol·s)
293.15 298.15 303.15 308.15 313.15	2.077×10^{4} 2.273×10^{4} 2.446×10^{4} 2.593×10^{4} 2.811×10^{4}	7.581×10^{4} 9.141×10^{4} 1.072×10^{5} 1.193×10^{5} 1.386×10^{5}	5.294×10^{3} 7.284×10^{3} 9.009×10^{3} 1.162×10^{4} 1.336×10^{4}	26.386 41.187 56.337 79.724 109.614

solution. This phenomenon can also be found in the investigation of Jiang et al. (2018), who added the MEA into DEEA and DMEA solution.

In addition, the pseudo first-order rate constant (k_0) values for EAE/1DMA2P system were also predicted using the proposed reaction rate/kinetic model at various temperatures, EAE concentrations and 1DMA2P concentrations. The experimental pseudo first-order rate constants (k_0) compared with the predicted values are plotted in Fig. 8 as a parity chart with AARD% of 6.642 for the proposed model. Thus, it can be concluded that the kinetic model developed in the present work is favourably acceptable and provides accurate results.

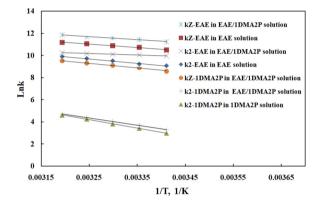


Fig. 7. Reaction rate constants of CO_2 absorption into aqueous EAE, 1DMA2P and EAE/1DMA2P solutions as a function of 1/T.

Table 5Activation energy *Ea* for different reaction rate constants in various amine solutions.

Rate constant	System	Activation energy (Ea), kJ/mol
$k_{2-\text{EAE}}$	EAE	32.81
$k_{\mathrm{Z-EAE}}$	EAE	25.25
$k_{2-\mathrm{EAE}}^{A}$	EAE/1DMA2P	11.25
$k_{Z-\mathrm{EAE}}^{A}$	EAE/1DMA2P	22.51
$k_{Z-1\text{DMA2P}}^{A}$	EAE/1DMA2P	34.64
k _{2-1DMA2P}	1DMA2P	62.83
$k_{2-1\mathrm{DMA2P}}^{A}$	EAE/1DMA2P	53.60
$k_{2-\text{MEA}}$	MEA	46.60
$k_{2-\text{MEA}}$	MEA/DEEA	43.15
$k_{2-\mathrm{MEA}}$	MEA/DMEA	37.33

5. Conclusion

The kinetics data of CO_2 absorption into aqueous EAE, 1DMA2P and EAE/1DMA2P solutions have been experimentally determined by using the stopped-flow technique at the temperature range from 293.15 to 313.15 K. The observed pseudo first-order rate constants (k_0) were obtained for $20-80 \text{ mol/m}^3$ EAE, $100-200 \text{ mol/m}^3$ 1DMA2P and EAE/1DMA2P with concentration of $10-20 \text{ mol/m}^3 + 100-200 \text{ mol/m}^3$. It can be observed that the pseudo first-order rate constants (k_0) as well as the overall reaction rates of CO_2 for the three amine systems depend

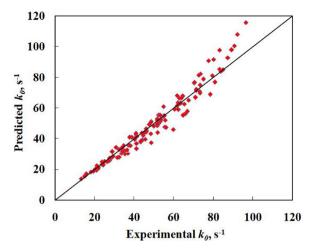


Fig. 8. Crossplot between experimental and predicted k_0 values for aqueous EAE/1DMA2P solution.

on the amine concentration and temperature, and the secondary amine exhibits the high reaction than that of 1DMA2P.

The probable reaction pathway of CO_2 absorption into EAE solution was interpreted by the zwitterions mechanism. The AARD% value of 6.073 fitted using Eq. (5) represented the good agreement between the predicted data and the experimental values, indicating the applicability of the zwitterions mechanism. In addition, the second rate constant (k_2) was correlated from the pseudo first-order rate constants (k_0) of the 1DMA2P solution using base-catalyzed hydration mechanism, and was found to depend of temperature with relating to the Arrhenius expression. The experimental values were found to be in accordance with the correlated pseudo first-order rate constants (k_0) , with the AARD% value of 4.406 and the activation energy (E_a) was calculated to be 62.83 kJ/mol for 1DMA2P system.

Furthermore, the kinetic performance of CO_2 absorption into the blended EAE/1DMA2P solution was also studied to evaluate the enhancement behavior and find the suitable mechanisms to explain the CO_2 absorption pathway. The experimental results showed that the overall reaction rate of CO_2 and EAE/1DMA2P is higher than the sum of individual EAE and 1DMA2P solution. Then, the determined data were then fitted using Eq. (16) with the AARD% of 6.642. It can be concluded that the addition of EAE can enhance the overall CO_2 reaction rate with 1DMA2P, which can be explained by the fact that 1DMA2P can attract the CO_2 from the zwitterion to release free EAE molecules or the 1DMA2P as a base can join the reaction of CO_2 and EAE.

Therefore, EAE acted as an effective activator can be added into the aqueous 1DMA2P solution to enhance the overall reaction rate, and EAE/1DMA2P can be considered as a potential absorbent for the application in the CO_2 capture process.

Author contribution statement

The corresponding author of Dr. Zhiwu Liang is responsible for ensuring that the descriptions are accurate and agreed by all authors. Hongxia Gao is responsible for the conceptualization, resources, and writing-original draft. Nan Wang is responsible for Data Curation and investigation. Jigang Du is responsible for the investigation and experiments. The corresponding author of Dr. Xiao Luo is responsible for writing- review & editing and gives Supervision.

Declaration of Competing Interest

The work described has not been published previously, and is not under consideration for publication elsewhere. In addition, its publication is approved by all authors (Hongxia Gao, Nan Wang, Jigang Du, Xiao Luo, Zhiwu Liang) and tacitly or explicitly by the responsible authorities where the work was carried out, and that, if accepted, it will not be published elsewhere in the same form, in English or in any other language, including electronically without the written consent of the copyright-holder.

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