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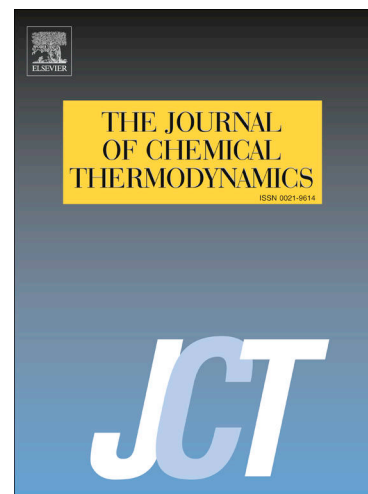
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Highlights

- Absorption performance of CO₂-N₂ in blended aqueous solutions was investigated;
- Increasing CO₂ partial pressure significantly enhances absorption capacity and rate;
- Promotion efficiencies of DEAE and DMA2P were compared;
- Effects of temperature and mass fraction on absorption performance were illustrated.

Abstract

Piperazine (PZ) was used to enhance the absorption of CO₂ in 2-diethylaminoethanol (DEAE) and 1-dimethylamino-2-propanol (DMA2P) aqueous solutions. The mole fraction of CO₂ in N₂-balanced gas mixtures ranged from 0.1 to 0.8. The mass fractions of amine and PZ ranged from 0.300 to 0.500 and from 0.050 to 0.075, respectively. The experiments were performed at 313.2 K. The absorption capacity and absorption rate under a series of conditions were illustrated, and the effect of CO₂ partial pressure on the absorption performance was demonstrated. At a given CO₂ partial pressure, the CO₂ absorbed by the PZ promoted DEAE and DMA2P aqueous solutions increases with the mass fraction of the mixed aqueous solutions, and it increases with the CO₂ partial pressure at a given mass fraction. The absorption capacity of the DMA2P-PZ aqueous solution is slightly higher than that of the DEAE-PZ aqueous solution. In addition, the absorption rate depends on the competitive effects of the mass fractions, solution viscosity and the partial pressure of CO₂.

Keywords: Absorption capacity; CO₂–N₂ gas mixture; PZ; DEAE; DMA2P

1. Introduction

In recent decades, the greenhouse effect and global warming caused by CO₂ emissions have attracted worldwide attention due to the use of large amounts of fossil fuels. The reduction of CO₂ emissions has become a global concern [1-3]. CO₂ capture technologies can be classified as solvent absorption, adsorption, cryogenic separation,

membrane and biological fixation [4-5]. Among these technologies, the alcoholamine absorption method has been widely used for the capture of CO₂ in natural gas, power plant flue gas, syn gas, and hydrogen gas streams [6-11]. Conventional amines such as primary amine-monoethanolamine (MEA), secondary amine-diethanolamine (DEA), tertiary amine-*N*-methyldiethanolamine (MDEA), and sterically hindered amine-2-amino-2-methyl-1-propanol (AMP) have been extensively used in industries for CO₂ removal [12-14]. However, the aforementioned conventional amine aqueous solutions have the disadvantages of high corrosivity and high energy consumption for regeneration. Therefore, the development of new absorbents with high absorption capacity, high reaction rate, low corrosivity and low energy cost in regeneration has attracted more and more attention [15-16].

Recently, 2-diethylaminoethanol (DEAE) and 1-dimethylamino-2-propanol (DMA2P) have attracted more attention in developing new absorbents for CO₂ removal [17-21]. Chowdhury *et al.* [17] investigated the absorption of CO₂ in different tertiary amine absorbents. They also compared the characteristics of these absorbents with those of the conventional absorbent MDEA, indicating that DEAE and DMA2P have higher absorption rates and cyclic capacities, yet lower or equal heat of reaction.

Adding a small amount of primary amine such as MEA to the tertiary amine aqueous solution can achieve better CO₂ absorption performance [22,23]. MEA preserves a high reaction rate of MDEA with CO₂ and a low enthalpy of the reaction of tertiary amine with CO₂, thus leading to a higher absorption rate in the absorber column and a lower regeneration heat in the stripper [22]. In addition to MEA, piperazine (PZ)

can also enhance the absorption of CO_2 in tertiary amine aqueous solutions. PZ is a cyclic diamine that is used as a promoter for CO_2 removal due to its rapid formation of carbamates and higher reactivity and higher CO_2 solubility than MEA [24-28]. Ali *et al.* [26] investigated the solubility of CO_2 in PZ promoted MDEA aqueous solutions and revealed that the addition of PZ can increase the solubility of CO_2 , especially at low CO_2 partial pressure. Kadiwala *et al.* [27] compared the CO_2 absorption performance in MEA and PZ aqueous solutions, and found that PZ has greater absorption capacity and significantly higher reaction rate than MEA. Xu *et al.* [28] determined the effect of PZ concentration on CO_2 loading in MDEA solutions and found that adding a small amount of PZ was beneficial to the CO_2 loading in MDEA aqueous solutions. In recent years, many studies have been focused on the use of accelerators to enhance the absorption of CO_2 in DEAE or DMA2P aqueous solutions [29-32]. However, the absorption performance of N_2 -balanced CO_2 in DEAE-PZ and DMA2P-PZ aqueous solutions have been rarely reported, and the effect of CO_2 partial pressure on absorption performance has not been well documented.

In post-combustion CO_2 capture technology, the CO_2 partial pressure in coal-fired flue gas of the power plant is usually less than 15%, e.g., the CO_2 partial pressure in the inlet flue gas of Huaneng Beijing Thermal Power Plant is 12.56% [33]. Many studies have shown that increasing the CO_2 partial pressure can increase the absorption of CO_2 [34-37]. However, the volumetric mass transfer coefficient of the gas phase first increases and then decreases with the increase of CO_2 partial pressure in the absorption tower [38]. The absorption rate is directly proportional to the CO_2 partial pressure at

the gas-liquid interface, so the higher the CO_2 partial pressure in the flue gas, the higher the CO_2 absorption rate. Increasing the CO_2 partial pressure may also reduce the liquid-gas mole ratio and the liquid-gas reaction is insufficient, resulting in a decrease in the reaction rate [39, 40]. Mazinani *et al.* [39, 40] found that higher CO_2 partial pressure, lower temperature and lower initial absorption can increase the CO_2 loading. However, under lower CO_2 partial pressure conditions, the process of CO_2 capture by amines aqueous solution takes 18-24 hours to reach equilibrium. Prolonged absorption process may cause evaporation of water, which will cause about 5% change in amine concentration. Therefore, a low CO_2 partial pressure is not conducive to the absorption process.

The main purpose of the present study is to determine the effects of partial pressure and solution concentration on the absorption performance of N_2 -balanced CO_2 in DEAE- PZ and DMA2P-PZ aqueous solutions, and to demonstrate the promotion efficiency of PZ. Thus, the time-dependent absorption capacity of N_2 -balanced CO_2 in these aqueous solutions was measured under different CO_2 partial pressures and solution compositions. Moreover, the absorption rates under different operations were also determined by conducting experiments.

2. Experimental

2.1 Materials

DEAE, DMA2P, MEA and PZ were used without further purification. The samples in this study are presented in Table 1 and Fig.1. CO_2 (purity in moles > 0.9999) and N_2 (purity in moles ≥ 0.99999) were purchased (Jinglian Gas Supply Inc. China).

The blended aqueous solutions were prepared by adding doubly distilled water (electrical resistivity $> 15 \text{ M}\Omega \text{ cm}$ at 298 K; Heal Force ROE-100 apparatus). The uncertainty of the electronic balance was $\pm 0.1 \text{ mg}$.

2.2 Apparatus and procedure

The experimental system of analyzing absorption performance was introduced and validated in our previous research [41]. The schematic of the experimental apparatus is shown in Fig. 2. The apparatus includes a high-pressure CO_2 storage tank, a high-pressure N_2 storage tank, two mass flow controllers (MFCs; Beijing Sevenstar Electronics Co., Ltd, Beijing, China; accuracy: $\pm 1\%$), a mass flow meter (MFM; Beijing Sevenstar Electronics Co., Ltd, Beijing, China; accuracy: $\pm 1\%$), an absorption bottle, a constant temperature water bath, a desiccator and a CO_2 analyzer (Sensors Europe GmbH, Germany; accuracy: $\pm 2\%$).

During the experiment, N_2 and CO_2 gases from the high-pressure tanks were transferred to the MFCs to maintain constant flow rates v_{N_2} and v_{CO_2} , respectively, and then the gases were transferred to the mixed gas cylinder to obtain a mixed gas. The mixed gas with a certain volume fraction of CO_2 was inlet into the absorption bottle and was then absorbed by the DEAE-PZ or DMA2P-PZ aqueous solution. N_2 and the unabsorbed CO_2 first flowed into the desiccator, then into the CO_2 analyzer, and finally into the MFM. The volume concentration of CO_2 (C_i) was measured using the CO_2 analyzer, and the flow rate (v_i) was measured by the MFM. C_i , v_i , and the corresponding time t_i were simultaneously recorded by the computer (interval time $\Delta t = 1 \text{ s}$). The absorption was terminated when v_i was almost constant within 100 s. The mass of the

absorbed CO_2 was calculated using the following equation: $273.15 \times 44/22.4 \times [0.9999v_{\text{CO}_2}t/T_0 - \sum v_i C_i \Delta t/T_1]$, where t is the total absorption time and T_0 and T_1 are the temperatures of CO_2 flowing out the MFC and desiccator, respectively. CO_2 loading (α) was calculated using the following equation: $\alpha = n_{\text{CO}_2} / (n_{\text{PZ}} + n_{\text{amine}})$ [42-46], where n_{CO_2} is the mole of loaded CO_2 , n_{PZ} and n_{amine} are the moles of PZ and DEAE or DMA2P in the unloaded aqueous solutions, respectively. In this study, all the experiments were conducted at 313.2 K, and the mole fraction of CO_2 in N_2 -balanced gas mixture ranged from 0.1 to 0.8. The mass fractions of amine and PZ ranged from 0.300 to 0.500 and from 0.050 to 0.075, respectively. The total pressure includes 4 parts, i.e., the vapor pressures of DMA2P and PZ, and the pressures of CO_2 and N_2 derived from the high-pressure gas cylinder (101.3kPa). Generally, the vapor pressures of pure DMA2P and pure PZ are small (approximately kPa), and in the aqueous solutions they get much smaller with the decrease of the mass fractions of DMA2P and PZ, hence we assume that the total pressure remains 101.3kPa during the experiment. The experimental system and detailed operating steps have been introduced and verified in our previous work, and the reliability of the equipment has also been verified in our previous work [41].

3. Results and discussion

The absorption performance of N_2+CO_2 in PZ promoted DEAE and DMA2P aqueous solutions was measured by using the experimental system shown in Fig.2. The absorption capacities including the mass of absorbed CO_2 (m) and CO_2 loading (α) in the blended amine aqueous solutions under different CO_2 partial pressures are presented

in Table 2. Similar to the absorption capacities of pure CO₂ in DEAE-PZ aqueous solutions [29], those of N₂+CO₂ increase with the increase of mass fractions of both DEAE and PZ at given temperature and given CO₂ partial pressure. In addition, by comparison with the references [29], [47] and [48], it is found that the absorption capacity of DEAE and DMA2P aqueous solutions without PZ is consistent with the changes of pressure and concentration. In the absorption process, PZ not only acts as an accelerator to promote the absorption of CO₂, but also can absorb a certain amount of CO₂. The table also shows that the absorption capacity increases rapidly with increasing CO₂ partial pressure, especially in the case of high concentrations. A decrease in CO₂ loading (α) is also observed with increasing amine concentration at given CO₂ partial pressure. Baltar *et al.* [49] found that this behavior was caused by the effects of physically absorbed CO₂. When a low amine concentration is used, an increase of physically absorbed CO₂ leads to a significant increase in CO₂ loading value. However, in the case of high amine concentration, the amount of physically absorbed CO₂ affects CO₂ loading value very slightly. Moreover, the amine in solution cannot completely react with CO₂ and the higher the concentration, the smaller amount of amine can react with CO₂. According to the definition ' α =mol CO₂/mol amines', the CO₂ loading decreases with increasing amine concentration.

Comparison with available literature data is very important. However, the partial pressure and concentration of the data in the literature [50,51] are different from those in our work. Due to inconsistent experimental conditions, we only perform trend comparisons, as shown in Fig.3. From Fig.3, one finds that when the CO₂ partial

pressure ranges from (10 to 101) kPa, the change trend of CO₂ loading in the literature is consistent with the laws in our work and both show a linear relationship. One also finds that when the experimental conditions are close, the obtained experimental results are also close. For example, the CO₂ loading 0.829 (mass fraction of DMA2P is 0.3108 and the partial pressure is 78.31kPa) in literatures [52] is very close to the CO₂ loading 0.80 (the mass fraction of DMA2P is 0.300 and the partial pressure is 80kPa) in our work.

Fig. 4 displays the effect of CO₂ partial pressure on the CO₂ absorption capacity in DEAE-PZ and DMA2P-PZ aqueous solutions. The figure shows that the mass of the absorbed CO₂ (m) and CO₂ loading (α) of N₂-balanced CO₂ increase with the increase in mass fractions of DEAE or DMA2P and PZ at given CO₂ partial pressure. Moreover, at given CO₂ partial pressure and given mass fraction, the mass of the absorbed CO₂ (m) in the DMA2P-PZ aqueous solution is slightly higher than that in the DEAE-PZ aqueous solution. It is very strange that although the absorption performance of CO₂ in DEAE single aqueous solution is slightly better than that in DMA2P single aqueous solution [17], the absorption capacity of DMA2P-PZ aqueous solution is slightly higher than that of DEAE-PZ aqueous solution at given mass fraction and given CO₂ partial pressure. For example, when the mass fraction is $w_{\text{DEAE}}/w_{\text{PZ}} = 0.300/0.050$, $w_{\text{DMA2P}}/w_{\text{PZ}} = 0.300/0.050$ and the CO₂ partial pressure is 80kPa, m (gCO₂/100g CO₂ unloaded aqueous solution) is 9.46 and 10.76, respectively. The figure also shows that the CO₂ partial pressure can significantly affect the mass of the absorbed CO₂ and the CO₂ loading. At a given w_{amine} value, the absorption capacity increases rapidly with the

increase in CO₂ partial pressure. This indicates that the higher the partial pressure, the greater the absorption capacity. This phenomenon was also evident in a previous study by the authors [41].

Fig. 5 and Table 3 display the time dependence of CO₂ absorption capacity in DEAE-PZ and DMA2P-PZ aqueous solutions. At given temperature and given mass fraction, the CO₂ absorption capacity of both blended aqueous solutions increased obviously with the increase of CO₂ partial pressure. For example, in the case of $w_{\text{DEAE}}/w_{\text{PZ}} = 0.300/0.050$, as the CO₂ partial pressure increases from 10 to 80kPa, m (gCO₂/100g CO₂ unloaded aqueous solution) increases from 7.45 to 9.46, and the absorption time decreases from 16000s to 3000s. This indicates that increasing the CO₂ partial pressure could efficiently improve the absorption capacity and significantly reduce the equilibrium time. Baltar *et al.* [49] showed the influence of tertiary amine concentration on CO₂ absorption performance. As aforementioned, they found that the effect of CO₂ absorption at low amine concentrations is related to the physically absorbed of CO₂. In this work, higher CO₂ partial pressure results in higher equilibrium CO₂ solubility, because high CO₂ partial pressure improves the physical absorption of CO₂. Increasing CO₂ partial pressure will lead to higher physically absorbed CO₂ concentration, hence more CO₂ can react with amine in solution.

To quantitatively show the effect of CO₂ partial pressure on the absorption rate of CO₂ in the blended aqueous solutions, a study by Chowdhury *et al.* [17] was followed and the absorption rate ($R \times 10$, g CO₂/100g CO₂ unloaded aqueous solution/min) was set to $0.5m/t_{0.5}$ (where $t_{0.5}$ is the absorption time at which 50% of the absorption capacity

is achieved). The absorption rates under different CO₂ partial pressures and mass fractions of mixed aqueous solutions are presented in Table 4. The table shows that the absorption rate is dependent on the CO₂ partial pressure and the mass fraction, and increases with an increase in CO₂ partial pressure and solution concentration. Moreover, the addition of a small amount of PZ into DEAE and DMA2P aqueous solutions can obviously increase the absorption rate at given CO₂ partial pressure. Fig.6 displays the effect of CO₂ partial pressure on the absorption rate of CO₂ in DEAE-PZ and DMA2P-PZ aqueous solutions. Table 4 and Fig.6 demonstrate that CO₂ partial pressure has a greater effect on the absorption rate than mass fraction. For example, when CO₂ is absorbed in the DMA2P-PZ aqueous solution at $w_{\text{DMA2P}}/w_{\text{PZ}} = 0.300/0.050$, the increase in the CO₂ absorption rate could reach up to 720% when CO₂ partial pressure increased from 10 to 80 kPa. However, the absorption rate only increases by 4% when w_{DMA2P} increases from 0.300 to 0.400 at a w_{PZ} value of 0.050.

However, in the case of high concentration solutions, the absorption rate may decrease with an increase in w_{amine} under low CO₂ partial pressures. In particular, in the cases of $p_{\text{CO}_2} = 80$ kPa and $w_{\text{PZ}} = 0.050$, the CO₂ absorption rate ($R \times 10$, g CO₂/100 g aqueous solution/min) decreases from 0.441 to 0.398 when w_{DEAE} increases from 0.400 to 0.500. Such a phenomenon may be interpreted by the formulation of the mass transfer rate. When gas A is absorbed in an aqueous solution B, the mass transfer rate N may be expressed as follows [32]:

$$N \propto C_A^* \sqrt{D_A k C_B} \quad (1)$$

where C_A^* is the concentration of CO₂, a is the absorbing surface area, D_A is the

diffusion coefficient of gas in the liquid phase, C_B is the concentration of the effective absorbent, and k is the chemical absorption rate constant. D_A is dominated by the viscosity of the aqueous solution and can be expressed as follows [29]:

$$D_A = \frac{14 \times 10^{-5}}{\eta_{water} (1.1 \times L_s / L_w) V_m^{0.6} \eta} \quad (2)$$

where η_{water} is the viscosity of water; η is the viscosity of the aqueous solution; V_m is the molar volume of diffusing substance; and L_s and L_w are the latent heat of vaporization of the solvent and water, respectively. By combining Eq. (1) and (2), the following is obtained:

$$N \propto \left(\frac{k C_B}{\eta} \right) C_A^* \quad (3)$$

For the absorption of pure CO_2 without partial pressure in the DEAE-PZ or DMA2P-PZ aqueous solutions, the absorption rate is only dependent on the competitive effects of concentration and viscosity in blended aqueous solution as C_A^* in Eq. (3) is a constant. However, as C_A^* is not a constant for the absorption of $\text{N}_2\text{-CO}_2$ gas, in addition to concentration and viscosity, the CO_2 partial pressure also affects the absorption rate. At given concentration of the effective absorbent C_B , an increase in CO_2 partial pressure may lead to an increase in C_A^* . Moreover, the mass of the absorbed CO_2 increases, resulting in an increase in solution viscosity. The competitive effects of CO_2 partial pressure, blended aqueous solution concentration, and viscosity on the absorption rate are presented in Fig. 7. The viscosity data in this figure are obtained from the authors' previous research [29]. The figure indicates that when the solution concentration and CO_2 partial pressure are low, the absorption rate increases rapidly

due to the low viscosity. However, at a higher solution concentration and viscosity, the CO₂ absorption rate decreases even under high CO₂ partial pressure. For example, when the total mass fraction of the mixed solution increases from 0.475 to 0.575 and the CO₂ partial pressure is 60kPa, the viscosity of the DEAE-PZ aqueous solution increases from 4.88 mPa·s to 6.40 mPa·s, resulting in a decrease in the CO₂ absorption rate ($R \times 10$, g CO₂/100g CO₂ unloaded aqueous solution/min) from 0.321 to 0.302. Although the change of the absorption rate is not significant, the viscosity did hinder the mass transfer.

To show the superiority of PZ promoted DEAE and DMA2P aqueous solutions, we compared the absorption performance with MEA promoter. Fig.8 and Table 5 show the effects of CO₂ partial pressure on the absorption performance in DEAE-PZ and DMA2P-PZ aqueous solutions compared with MEA promoted DEAE and DMA2P aqueous solution. One finds from this figure that at given mass fraction, DEAE and DMA2P aqueous solutions promoted by PZ have higher absorption capacity and absorption rate for CO₂ than MEA promoted DEAE and DMA2P aqueous solutions. Such phenomenon is also found in other literatures [29-32]. Compared with MEA, as PZ has two functional nitrogen groups, it has nearly twice the absorption rate, double the cyclic CO₂ capacity, 1.5-3 times the mass transfer coefficient, and much less volatility. Moreover, the heat of absorption and the thermal degradation rate of PZ are lower than those of MEA, indicating that PZ has lower regenerative heat duty and higher resistance to degradation and corrosion. Thus, PZ as a promoter of DEAE and DMA2P aqueous solution has a great application potential in the field of CO₂ capture.

4. Conclusions

In this study, the CO₂ absorption performance of DEAE-PZ and DMA2P-PZ aqueous solutions from CO₂-N₂ gas mixture was estimated. The absorption capacity and absorption rate under a series of conditions were illustrated, and the effect of CO₂ partial pressure on the absorption performance was also demonstrated. The results of the present study revealed the following:

- (1) The CO₂ absorption capacities of PZ promoted DEAE and DMA2P aqueous solutions from CO₂-N₂ gas mixture increase with an increase in the blended aqueous solution concentration at given CO₂ partial pressure and increase with an increase in partial pressure of CO₂ at given mass fraction. Moreover, the CO₂ partial pressure has a greater effect on the absorption performance than the solution concentration;
- (2) Under the same mass fraction and CO₂ partial pressure, the absorption capacity of DMA2P-PZ aqueous solution is a slightly higher than that of DEAE-PZ aqueous solution;
- (3) With the increase of CO₂ partial pressure and solution concentration, the CO₂ absorption rate in the PZ promoted amine aqueous solution increases. The absorption rate is dependent on the competitive effects of concentration, viscosity in the blended aqueous solution, and CO₂ partial pressure in the CO₂-N₂ gas mixture. In addition, through the comparison of accelerators, it is found that PZ promoted DEAE and DMA2P aqueous solutions have better absorption performance.

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References

- [1] L.D. Wang, Y.F. Zhang, R.J. Wang, Q.W. Li, S.H. Zhang, M. Li, J. Liu, B. Chen, Environ. Sci. Technol. 52 (2018) 14556-14563.
- [2] C.W. Duan, Y.Z. Cao, L.X. Hu, D. Fu, J.L. Ma, Y. Jeffrey, J. Hazard. Mater. 373 (2019) 141-151.
- [3] M. Owais, M. Jarvinen, P. Taskinen, A. Said, J. CO₂ Util. 31 (2019) 1-7.
- [4] C.L. Huang, C.J. Liu, K.J. Wu, H.R. Yue, S.Y. Tang, H.F. Lu, B. Liang, Energy Fuels 33 (2019) 3380-3389.
- [5] R.J. Wang, S.S. Liu, L.D. Wang, Q.W. Li, S.H. Zhang, B. Chen, L. Jiang, Y.F. Zhang, Appl. Energy 242 (2019) 302-310.
- [6] R. Ramazani, S. Mazinani, A. Hafizi, A. Jahanmiri, Process Saf. Environ. 98 (2015) 325-335.
- [7] M.G. Yildiz, T. Davran-Candan, M.E. Gunay, R. Yildirim, J. CO₂ Util. 31 (2019) 27-42.
- [8] P. Zhang, L.M. Wang, C.J. Lu, M.Y. Li, S.J. Yu, D. Fu, J. Mol. Liq. 288 (2019)

111031-111039.

- [9] Y.Y. Liu, S. Lin, H.F. Lu, C.J. Liu, K.J. Wu, W. Jiang, B. Liang, J. Chem. Thermodyn. 136 (2019) 16-27.
- [10] A. Hafizi, M.R. Rahimpour, S. Hassanajili, Appl. Energ. 169 (2016) 629-641.
- [11] N.N. Xing, B. Dai, X.X. Ma, J. Wei, Y. Pan, W. Guan, J. Chem. Thermodyn. 115 (2017) 171-179.
- [12] M.M.Ghiasi, A.H.Mohammadi, J.Nat.Gas.Sci.Eng. 18(2014) 39-46.
- [13] H.Pahlavanzadeh, S.Nourani, M.Saber, J. Chem. Thermodyn. 43 (2011) 1775-1783.
- [14] J.M.Navaza, D.Gomez-Diaz, M.D. La Rubia, Chem.Eng.J. 146(2009) 184-188.
- [15] G.T.Rochelle, Science 325 (2009) 1652-1654.
- [16] F.Barzagli, S.Lai, F.Mani, Chem.Sus.Chem. 8(2015) 184-191.
- [17] F.A. Chowdhury, H. Yamada, T. Higashii, K. Goto, M. Onoda, Ind. Eng. Chem. Res. 52 (2013) 8323-8331.
- [18] G. Puxty, R. Rowland, A. Allport, Q. Yang, M. Bown, R. Burns, M. Maeder, M. Attalla, Environ. Sci. Technol. 43 (2009) 6427-6433.
- [19] L. Wen, H.L. Liu, W. Rongwong, Z.W. Liang, K.Y. Fu, R. Idem, P. Tontiwachwuthikul, Chem. Eng. Technol. 38 (2015) 1435-1443.
- [20] Y.J. Liang, H.L. Liu, W. Rongwong, Z.W. Liang, R. Idem, P. Tontiwachwuthikul, Fuel 144 (2015) 121-129.
- [21] T.L. Donaldson, Y.N. Nguyen, Ind. Eng. Chem. Fundam. 19 (1980) 260-266.
- [22] T. Chakravarty, U. Phukan, R. Weiland, Chem. Eng. Prog. 81 (1985) 32-36.
- [23] A.L. Kohl, R.B. Nielsen, Gas purification, fifth ed., Gulf Professional Publishing,

Houston, TX, 1997.

- [24] C.H. Yu, H.H. Cheng, C.S. Tan, *Int. J. Greenh. Gas Con.* 9 (2012) 136-147.
- [25] A.V. Rayer, Y. Armugam, A. Henni, P. Tontiwachwuthikul, *J. Chem. Eng. Data* 59 (2014) 3610-3623.
- [26] B.S. Ali, M.K. Aroua, *Int. J. Thermophys.* 25 (2004) 1863-1870.
- [27] S. Kadiwala, A.V. Rayer, A. Henni, *Fluid Phase Equilibr.* 292 (2010) 20-28.
- [28] G.W. Xu, C.F. Zhang, S.J. Qin, W.H. Gao, H.B. Liu, *Ind. Eng. Chem. Res.* 37 (1998) 1473-1477.
- [29] D. Fu, L.M. Wang, C.L. Mi, P. Zhang, *J. Chem. Thermodyn.* 101 (2016) 123-129.
- [30] P.D. Vaidya, E.Y. Kenig, *Ind. Eng. Chem. Res.* 47 (2010) 34-38.
- [31] P.B. Konduru, P.D. Vaidya, E.Y. Kenig, *Environ. Sci. Technol.* 44 (2010) 2138-2143.
- [32] M. Nitta, K. Hayashi, Y. Furukawa, H. Sato, Y. Yamanaka, *Energy Procedia* 63 (2014) 1863-1868.
- [33] B. Huang, S.S. Xu, S.W. Gao, L.B. Liu, J.Y. Tao, H.W. Niu, M. Cai, J.A. Cheng, *Appl. Energ.* 86 (2010) 3347-3354.
- [34] M. Kanakubo, T. Makino, T. Umecky, M. Sakurai, *Fluid Phase Equilibr.* 420 (2016) 74-82.
- [35] P. Singh, D.W.F. Brilman, M.J. Groeneveld, *Int. J. Greenh. Gas. Con.* 5 (2011) 61-68.
- [36] A. Jahangiri, M.N. Hassankiadeh, *Sep. Sci. Technol.* 54 (2019) 1067-1078.
- [37] M.N. Hassankiadeh, A. Jahangiri, *Energy* 165 (2018) 164-178.
- [38] H. Ling, S. Liu, H.X. Gao, H.Y. Zhang, Z.W. Liang, *Sep. Purif. Technol.* 232 (2020)

115957-115968.

- [39] S. Mazinani, A. Samsami, A. Jahanmiri, J. Chem. Eng. Data 56 (2011) 3163-3168.
- [40] S. Mazinani, R. Ramazani, A. Samsami, A. Jahanmiri, B. Van der Bruggen, Fluid Phase Equilib. 396 (2015) 28-34.
- [41] D. Fu, X. Feng, P. Zhang, J. Chem. Thermodyn. 113 (2017) 250-256.
- [42] P. Zhang, X.F. Tian, D. Fu, Energy 161 (2018) 1122-1132.
- [43] K.Fu, P.Zhang, L.M.Wang, X.Y.Huang, D.Fu, J. Mol. Liq. 302(2020) 112518-112528.
- [44] D. Fu, P. Zhang, C.L. Mi, Energy 101 (2016) 288-295.
- [45] D. Fu, P. Zhang, L.M. Wang, Energy 113 (2016) 1-8.
- [46] K.Fu, P.Zhang, D.Fu, J. Chem. Thermodyn. 139 (2019) 105862-105871.
- [47] D. Fu, L.M. Wang, P. Zhang, C.L. Mi, J. Chem. Thermodyn. 95 (2016) 136-141.
- [48] L.M. Wang, X.F.Tian, D. Fu, X.Q.Du, J.H.Ye, J. Chem. Thermodyn. 139 (2019) 105865-105872.
- [49] A. Baltar, D. Gomez-Diaz, J.M. Navaza, A. Rumbo, AIChE J. 66(2020) 16770-16778.
- [50] M.Afkhamipour, M.Mofarahi, Fluid Phase Equilib. 475 (2018) 38-51.
- [51] M. Xiao, W.C. Zheng, L.H. Liu, P. Tontiwachwuthiku, Z.W. Liang, AIChE J. 65 (2019) 1-10.
- [52] M.W.Arsha, H.F.Svendsen, P.Fosbol, N.Von Solms, K.Thomsen, J. Chem. Eng. Data 59(2014) 764-774.

Figure 1 Chemical structures of samples used in this study.

Figure 2 Schematic of the experiments.

Figure 3 Comparison of DEAE aqueous solution and DMA2P aqueous solution (insert)

on the CO_2 loading between experimental data and literature data.

$T=313.2\text{K}$. Symbols: main plot: $\bullet w_{\text{DEAE}}=0.300$; $\circ w_{\text{DEAE}}=0.236$ [51]. Insert

plot: $\bullet w_{\text{DMA2P}}=0.300$; $\circ w_{\text{DMA2P}}=0.3108$ [50]. Lines: trend lines.

Figure 4 Effects of CO_2 partial pressure on the CO_2 absorption capacity and CO_2

loading (insert) in PZ promoted DEAE and DMA2P aqueous solutions.

$T=313.2\text{K}$.

Symbols: $\bullet w_{\text{DEAE}}/w_{\text{PZ}}=0.300/0.050$;

$\blacksquare w_{\text{DMA2P}}/w_{\text{PZ}}=0.300/0.050$. Lines: trend lines.

Figure 5 Time dependence of CO_2 absorption amount in DEAE-PZ and DMA2P-PZ

aqueous solutions at different CO_2 partial pressure at

$w_{\text{DEAE}}/w_{\text{PZ}}=0.300/0.050$ and $w_{\text{DMA2P}}/w_{\text{PZ}}=0.300/0.050$ (insert). $T=313.2\text{K}$.

Symbols: $\bullet p_{\text{CO}_2}=10\text{kPa}$; $\circ p_{\text{CO}_2}=30\text{kPa}$; $\blacksquare p_{\text{CO}_2}=60\text{kPa}$; $\square p_{\text{CO}_2}=80\text{kPa}$. Lines:

trend lines.

Figure 6 Effects of CO_2 partial pressure on the CO_2 absorption rate in DEAE-PZ and

DMA2P-PZ aqueous solutions (insert) at 313.2 K. Symbols: Main plot,

▲ $w_{\text{DEAE}}/w_{\text{PZ}}=0.300/0.000$; ■ $w_{\text{DEAE}}/w_{\text{PZ}}=0.300/0.050$; ● $w_{\text{DEAE}}/w_{\text{PZ}}$
 $=0.300/0.075$; Insert plot, ▲ $w_{\text{DMA2P}}/w_{\text{PZ}}=0.300/0.000$;
 ■ $w_{\text{DMA2P}}/w_{\text{PZ}}=0.300/0.050$; ● $w_{\text{DMA2P}}/w_{\text{PZ}}=0.300/0.075$. Lines: trend lines.

Figure 7 Effects of solution concentration on the apparent absorption rate and solution viscosity. $T=313.2$ K. Symbols: ● $p_{\text{CO}_2}=10\text{kPa}$; ■ $p_{\text{CO}_2}=30\text{kPa}$; ▲ $p_{\text{CO}_2}=60\text{kPa}$; ◆ $p_{\text{CO}_2}=80\text{kPa}$; ○ the viscosities of $w_{\text{PZ}}=0.075$ at different DEAE concentration. Lines: —R—; --- η ---. The viscosity data are taken from Ref. [29].

Figure 8 Effects of CO_2 partial pressure on the absorption capacity and absorption rate (insert) in DEAE-PZ and DMA2P-PZ aqueous solutions comparing with DEAE-MEA and DMA2P-MEA aqueous solutions. $T=313.2$ K. Symbols: ● $w_{\text{DEAE}}/w_{\text{PZ}}=0.300/0.050$; ○ $w_{\text{DEAE}}/w_{\text{MEA}}=0.300/0.050$; ■ $w_{\text{DMA2P}}/w_{\text{PZ}}=0.300/0.050$; □ $w_{\text{DMA2P}}/w_{\text{MEA}}=0.300/0.050$. Lines: trend lines.

Table 1 Sample description

Substance	Source	Purity ^a	CAS No.	IUPAC
DEAE	Aladdin Reagent	$w \geq 0.99$	100-37-8	2-(diethylamino)ethanol
DMA2P	TCI Reagent	$w \geq 0.98$	108-16-7	1-(dimethylamino)propan-2-ol
PZ	Sinopharm Chemical Reagent	$w \geq 0.995$	110-85-0	Piperazine

MEA	Aladdin Reagent	$w \geq 0.995$	141-43-5	2-aminoethanol
CO ₂	Jinglian gas	$x \geq 0.9999$	124-38-9	Carbon dioxide
N ₂	Jinglian gas	$x \geq 0.99999$	<u>7727-37-9</u>	Nitrogen
H ₂ O	Heal force ROE-100 apparatus	Electrical resistivity > 15MΩ cm at $T = 298$ K	7732-18-5	Water

NOTE: ^aStated by the supplier; w denotes mass fraction, x denotes volume fraction.

Table 2 Absorption capacities of CO₂ in blended amine aqueous solutions under different CO₂ partial pressures. $T=313.2$ K and the total pressure of CO₂+N₂ mixture is 101.3kPa.

$m/(\text{gCO}_2 \text{ per } 100\text{g CO}_2 \text{ unloaded aqueous solution})$					$\alpha/(\text{molCO}_2 \text{ per } \{\text{mol amine}+\text{PZ}\})$												
10 kPa	30 kPa	60 kPa	80 kPa	101 kPa	8.1 kPa	8.66 kPa	10 kPa	15 kPa	20.3 kPa	30 kPa	30.4 kPa	46.16 kPa	60 kPa	60.8 kPa	78.31kPa	80 kPa	101 kPa
					0.673 [51]			0.733 [51]	0.787 [51]		0.825 [51]			0.855 [51]			
6.54	7.54	8.21	8.64	9.52[29] [50]			0.58			0.67			0.73			0.77	0.85[50]
7.45	7.96	8.71	9.46	10.24[29]			0.54			0.58			0.63			0.68	0.74[29]
7.86	8.59	9.54	10.1 1	10.87[29]			0.52			0.57			0.63			0.67	0.72[29]
7.97	9.14	10.5 7	10.8 1	12.28[50] [29]			0.53			0.61			0.70			0.72	0.82[50] [29]
8.70	9.75	10.8 1	11.2 0	12.96[29]			0.50			0.55			0.62			0.64	0.74[29]
9.12	10.2 6	11.0 2	11.9 3	13.54[29]			0.48			0.54			0.58			0.63	0.72[29]
9.31	10.8 6	12.9 4	13.1 7	14.80[29]			0.49			0.58			0.67			0.70	0.79[29]
10.2 5	11.5 9	12.9 7	14.0 0	15.15[29]			0.48			0.54			0.61			0.66	0.71[29]

10.6 9	12.1 7	13.4 9	14.4 7	15.40[29]	0.47	0.54	0.60	0.64	0.68[2]
7.50	8.12	9.70	10.2 5	10.79[48]	0.59	0.63	0.76	0.80	0.84[4]
8.40	8.89	10.0 4	10.7 6		0.54	0.58	0.65	0.70	
8.72	9.26	10.3 8	11.0 7		0.52	0.56	0.62	0.67	
					0.476[50]		0.756[50]	0.829[50]	
8.64	10.1 1	11.0 0	12.6 4	13.75[48]	0.51	0.59	0.65	0.74	0.81[4]
9.59	10.7 5	11.7 7	13.5 4		0.49	0.55	0.60	0.69	
10.0 1	10.9 3	12.3 3	13.6 9		0.48	0.52	0.59	0.66	
10.2 3	11.8 7	13.4 7	14.3 2	16.12[48]	0.48	0.56	0.63	0.67	0.76[4]
11.3 0	12.6 3	13.9 4	15.0 7		0.47	0.53	0.58	0.63	
11.6 5	12.8 3	14.1 8	15.3 5		0.46	0.49	0.56	0.61	

Standard uncertainties u are $u(T)=0.1\text{K}$; $u(w)=\pm 0.005$; $u(p)=5\text{kPa}$; $u(\alpha)=0.03\text{molCO}_2$ per mol amine+PZ, $u(m)=0.30\text{gCO}_2$ per 100g CO_2 unloaded aqueous solution.

Table 3 Time dependence of CO_2 absorption amount in DEAE-PZ and DMA2P-PZ aqueous solutions at different CO_2 partial pressure at $w_{\text{DEAE}}/w_{\text{PZ}}=0.300/0.050$ and $w_{\text{DMA2P}}/w_{\text{PZ}}=0.300/0.050$. $T=313.2\text{K}$ and the total pressure of CO_2+N_2 mixture is 101.3kPa.

t/s	m/(g CO_2 /100g CO_2 unloaded aqueous solution)							
	DEAE-PZ				DMA2P-PZ			
	10 kPa	30 kPa	60 kPa	80 kPa	10 kPa	30 kPa	60 kPa	80 kPa
0	0	0	0	0	0	0	0	0
300	0.08	0.25	0.66	1.91	0.11	0.30	0.99	2.27
600	0.33	1.11	2.18	4.09	0.40	1.14	2.60	4.52
900	0.62	2.05	3.89	6.34	0.72	2.07	4.40	6.94
1200	0.93	2.98	5.35	8.04	1.05	3.00	6.06	8.91

1500	1.26	3.87	6.49	8.91	1.39	3.90	7.39	9.98
1800	1.58	4.69	7.31	9.28	1.72	4.74	8.33	10.43
2100	1.90	5.42	7.83	9.43	2.04	5.49	8.92	10.62
2400	2.21	6.03	8.15	9.45	2.37	6.14	9.29	10.71
2700	2.51	6.54	8.34	9.46	2.68	6.70	9.51	10.75
3000	2.80	6.94	8.46		3.00	7.16	9.65	10.75
3300	3.09	7.26	8.54		3.30	7.54	9.75	10.76
3600	3.37	7.49	8.60		3.60	7.84	9.82	
3900	3.64	7.66	8.65		3.90	8.07	9.88	
4200	3.90	7.77	8.68		4.19	8.26	9.92	
4500	4.16	7.86	8.70		4.46	8.40	9.97	
4800	4.42	7.91	8.71		4.73	8.51	10.01	
5100	4.66	7.91			4.99	8.59	10.03	
5400	4.90	7.93			5.24	8.65	10.04	
5700	5.13	7.93			5.47	8.69		
6000	5.35	7.94			5.70	8.73		
6300	5.55	7.94			5.90	8.76		
6600	5.74	7.95			6.10	8.79		
6900	5.92	7.95			6.28	8.81		
7200	6.08	7.96			6.45	8.82		
7500	6.23				6.61	8.83		
7800	6.36				6.76	8.84		
8100	6.48				6.90	8.86		
8400	6.59				7.03	8.87		
8700	6.69				7.15	8.87		
9000	6.77				7.26	8.88		
9300	6.84				7.36	8.89		
9600	6.91				7.45			
9900	6.97				7.53			
10200	7.03				7.61			
10500	7.07				7.68			
10800	7.12				7.75			
11100	7.16				7.81			
11400	7.19				7.86			
11700	7.23				7.91			
12000	7.26				7.96			
12300	7.29				8.01			

12600	7.32	8.05
12900	7.34	8.08
13200	7.37	8.12
13500	7.39	8.15
13800	7.42	8.18
14100	7.44	8.21
14400	7.45	8.24
14700		8.26
15000		8.28
15300		8.31
15600		8.33
15900		8.35
16200		8.37
16500		8.39
16800		8.40
17100		8.40

Standard uncertainties u are $u(T)=0.1\text{K}$; $u(t)=1\text{s}$; $u(w)=\pm 0.005$; $u(p)=5\text{ kPa}$; $u(m)=0.30\text{gCO}_2$ per 100g CO_2 unloaded aqueous solution.

Table 4 The absorption rate (R) of CO₂ in blended amine aqueous solutions.

$T=313.2\text{K}$ and the total pressure of CO₂+N₂ mixture is 101.3kPa.

w_{amine}	w_{PZ}	R×10(g CO ₂ per 100g CO ₂ unloaded aqueous solution/min)			
		10 kPa	30 kPa	60 kPa	80 kPa
DEAE-PZ					
	0	0.019	0.045	0.119	0.177
0.300	0.050	0.055	0.156	0.293	0.437
	0.075	0.066	0.174	0.304	0.456
	0	0.020	0.063	0.151	0.162
0.400	0.050	0.064	0.158	0.298	0.441
	0.075	0.079	0.194	0.321	0.469

	0	0.023	0.055	0.131	0.158
0.500	0.050	0.058	0.166	0.271	0.398
	0.075	0.071	0.183	0.302	0.462
DMA2P-PZ					
	0	0.019	0.021	0.055	0.065
0.300	0.050	0.052	0.157	0.276	0.428
	0.075	0.063	0.180	0.301	0.458
	0	0.021	0.034	0.048	0.054
0.400	0.050	0.071	0.174	0.302	0.447
	0.075	0.077	0.183	0.332	0.474
	0	0.025	0.043	0.048	0.060
0.500	0.050	0.066	0.161	0.284	0.401
	0.075	0.079	0.193	0.348	0.437

Standard uncertainties u are $u(T)=0.1\text{K}$; $u(w)=\pm 0.005$; $u(p)=5\text{kPa}$; $u(R)=0.01\text{g CO}_2$ per 100g CO_2 unloaded aqueous solution/min.

Table 5 Absorption capacities and absorption rate of CO_2 in PZ promoted DEAE and DMA2P amine aqueous solutions compared with MEA promoter under different CO_2 partial pressures. $T=313.2\text{K}$ and the total pressure of CO_2+N_2 mixture is 101.3kPa.

w_{amine}	w_{promoter}	$m/(\text{gCO}_2/100\text{g CO}_2 \text{ unloaded aqueous solution})$				$R \times 10(\text{g CO}_2/100\text{g CO}_2 \text{ unloaded aqueous solution/min})$			
		10kPa	30kPa	60kPa	80kPa	10kPa	30kPa	60kPa	80kPa
DEAE-PZ ^α									

0.300	0.050	7.45	7.96	8.71	9.46	0.055	0.156	0.293	0.437
DEAE-MEA									
0.300	0.050	7.38	7.87	8.33	9.25	0.048	0.136	0.229	0.379
DMA2P-PZ ^a									
0.300	0.050	8.40	8.89	10.04	10.76	0.052	0.157	0.276	0.428
DMA2P-MEA									
0.300	0.050	8.27	8.82	9.37	9.95	0.051	0.118	0.246	0.403

NOTE: ^aThe absorption capacities and absorption rate for DEAE-PZ and DMA2P-PZ are taken from Tables 2 and 4. Standard uncertainties u are $u(T)=0.1\text{K}$; $u(w)=\pm 0.005$ $u(p)=5\text{kPa}$; $u(m)=0.30\text{gCO}_2/100\text{g CO}_2$ unloaded aqueous solution; $u(R)=0.01\text{g CO}_2/100\text{g CO}_2$ unloaded aqueous solution/min.