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Solubility of CO_2 in Nonaqueous Absorption System of 2-(2-Aminoethylamine)ethanol + Benzyl Alcohol

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ABSTRACT: A mixture of 2-(2-aminoethylamine)ethanol (AEEA) with benzyl alcohol (BP) as absorbent was developed to reduce energy consumption in an amine-based CO_2 capture process. The solubility of CO_2 in the AEEA + BP solution was compared with AEEA + diethylene glycol (DEG), AEEA + triethylene glycol (TEG), and AEEA + H_2O. The CO_2 loading (mol CO_2/kg amine) at the same CO_2 partial pressure was in the order AEEA + H_2O > AEEA + BP > AEEA + TEG > AEEA + DEG, and CO_2 loading of all these solutions increased with increasing vapor pressure of CO_2. Besides, the CO_2 loading in AEEA + BP solution significantly decreased with increasing reaction temperature. The desorption rate of AEEA + BP solution was about 1.3 times faster than that of aqueous AEEA solution at the same desorption temperature, and AEEA + BP solution needed 30% time less than AEEA + H_2O solution to reach reaction end point. The AEEA + BP solution showed stable repeatability in four cycles of absorption–desorption tests.
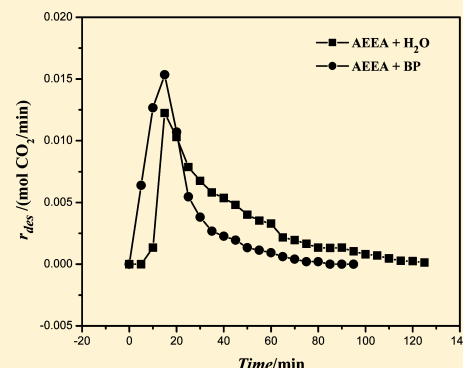
1. INTRODUCTION

CO_2 capture technology in postcombustion continues to be one of the attractive options for the reduction of CO_2 emissions from the fossil fuel combustion process, particularly coal-fired power plants. Among the applicable CO_2 removal processes, chemical absorption using aqueous alkanolamine solutions is the most effective method.[1] The major aqueous alkanolamine solutions applied in the industrial process are monoethanolamine (MEA), diethanolamine (DEA), diglycolamine (DGA), N-methyldiethanolamine (MDEA), and 2-amino-2-methyl-1-propanol (AMP). Recently, attention has been paid to diamines which present a higher absorption capacity than primary and secondary alkanolamines.[2−5] Among them, aqueous 2-(2-aminoethylamine)ethanol (AEEA) solutions (5 to 25) wt % have a CO_2 capacity of (1.35 to 1.12) mol CO_2/mol amine at 296 K and atmospheric pressure.[2,3]

In the modern industry, the heat of regeneration for the stripping out of CO_2 from rich amine solutions in a regeneration process, which consists of sensible heat, reaction heat, latent heat of partial water, and partial latent heat of solvent vaporization, contributes (50 to 80) % of the total energy consumption in a chemical solvent based on the CO_2 separation process.[6] Many researchers have used blended amines, a solution of two or more amines in a variety of compositions, to improve absorption or desorption ability.[7,8] However, regardless of what the compositions of solutions are, the high sensible heat, and the latent heat of water vaporization will inevitably consume excessive energy in the regeneration process. During the regeneration process, a large amount of sensible heat is first utilized to increase the solution temperature to the stripping temperature because of the specific heat capacity of the aqueous solution. Meanwhile,

additional energy is inevitable for the water evaporation, and to promote the desorption reaction.[9−12] On the other hand, the aqueous amine solution processes usually suffer from fast equipment corrosion and amine degradation at high temperature.[9] In brief, the disadvantages of the conventional CO_2 capture processes with aqueous amine solutions are commonly ascribed to water. If water is substituted by other high-boiling-point solvents, a considerable amount of thermal energy can be saved during the solvent heating cycle,[9,13] and the corrosion will also be eliminated.

Recently, nonaqueous solutions have been extensively investigated for acid gas treatment. Usubharatana et al.[14] found that methanol instead of water mixed into MEA solution could enhance the CO_2 absorption. Versteeg et al.[15] studied MDEA mixed with ethanol and methanol to remove CO_2. Luo et al.[16,17] developed a mixture of MEA + triethylene glycol (TEG) and AMP + diethylene glycol (DEG) and AMP + TEG for CO_2 capture. Qi et al.[9] investigated the dynamics of CO_2 absorption and desorption processes in mixed solvents of alkanolamine and poly(ethylene glycol) (PEG). Park et al.[13,18−20] measured the chemical absorption rate of CO_2 with diisopropanolamine (DIPA), triethanolamine (TEA), MEA, and MDEA with polar organic solvents. However, these nonaqueous mixture systems usually have relatively high vapor pressure, and thus are not suitable for CO_2 capture from flue gases. Meanwhile, many nonaqueous solvents have high viscosity which affects the liquid-film coefficient for mass transfer.[21] Ionic liquids (ILs) as novel solvents have been
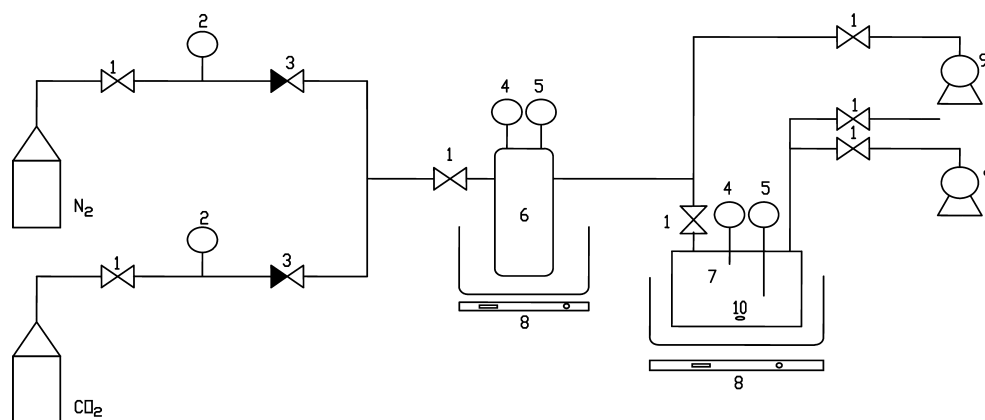
Figure 1. Schematic diagram of vapor–liquid equilibrium apparatus: 1, globe valve; 2, rotameter; 3, check valve; 4, digital pressure gauge; 5, digital thermometer; 6, gas reservoir; 7, reactor; 8, water bath; 9, vacuum pump; 10, rotor.
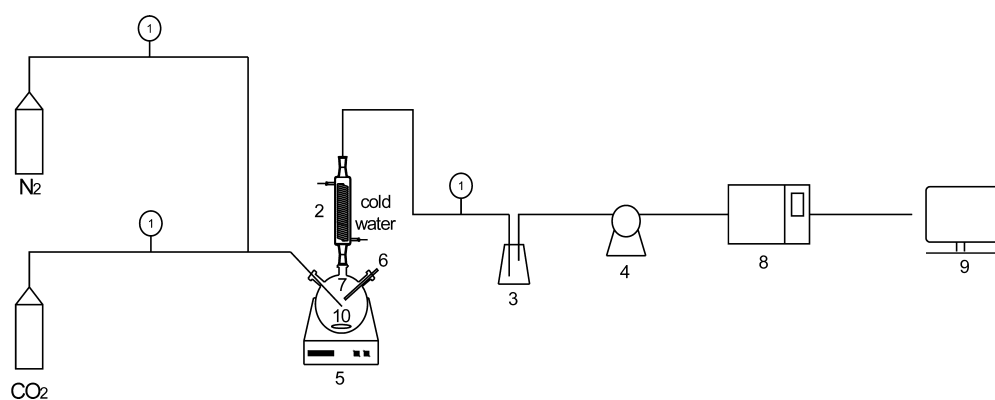

Figure 2. Schematic diagram of cyclic absorption–desorption apparatus: 1, rotameter; 2, condenser; 3, drying bottle; 4, wet gas flow meter; 5, oil bath; 6, thermometer; 7, three-neck flask; 8, gas chromatograph; 9, computer; 10, rotor.

intensively developed for CO_2 capture from flues gases, and they exhibit good solubility and high selectivity to CO_2.[22−24] The mixed solvent of ILs and alkanolamine with efficient and reversible performance for CO_2 capture show more commercial potential. However, the comparatively high viscosity, price, and their unknown toxicity impede ILs for large-scale industrial application.

The objective of this work is to develop a nonaqueous mixture system for CO_2 capture with low stripping temperature, using AEEA as the main chemical absorbent, and benzyl alcohol (BP) with its low viscosity and a boiling point of 478 K as nonaqueous media. To our knowledge, this system is not available in the literature. Besides, the absorption and desorption characteristics of this new system are compared with the aqueous AEEA system, and cycle absorption–desorption tests will also be performed to explore the feasibility of AEEA + BP used in industry.

2. EXPERIMENTAL SECTION

2.1. Materials. Amines, AEEA (minimum purity 99%), BP (minimum purity 98%), DEG (minimum purity 98%), and TEG (minimum purity 99%) were purchased from Aladdin. The CO_2 (99.99%) and N_2 (99.99%) gases were purchased from Guangming Research & Design Institute of Chemical Industry in China.

2.2. The Vapor–Liquid Equilibrium Apparatus. The partial pressures of CO_2 in absorbents and the measure of absorption rate were measured in the vapor–liquid equilibrium

(VLE) apparatus (Figure 1), and the principle and procedures had been reported in our previous work.[25] The internal volumes of the stainless steel gas reservoir and the reactor were 1290 cm^3 and 1105 cm^3, respectively. The gas reservoir and reactor were heated using a water bath. The temperatures of CO_2 and amine solutions were measured with a K-type thermocouple (eastern WRNK-191) and the pressure of the gas was continuously measured using a pressure sensor (Hong Kong North ZTT). The standard uncertainty u for temperature is $u(T) = 1$ K, for CO_2 partial pressure $u(P_{CO_2}) = 0.2$ kPa, for mass fraction of the solution $u(X) = 0.1$ wt %, and for CO_2 loading (mol CO_2/kg amine) $u(\alpha) = 0.01$.

2.3. Cyclic Absorption and Desorption Apparatus. The absorption and desorption apparatus is shown in Figure 2. The apparatus consists of a 500 mL three-neck flask with a rotor, three rotameters, a wet gas flow meter, and an online gas chromatograph (GC-960). The temperature of the experiment was controlled by oil bath, and the standard uncertainty u for temperature is $u(T) = 1$ K. Prior to the introduction of solvent, the oil bath was heated to a desired temperature and the whole system was purged with N_2 to remove air. Then 300 g of solution was fed into the flask. The absorption experiments were performed at 313 K using a CO_2 flow of 300 mL/min. The flow was cut off when the outlet flow rate was 300 mL/min. The desorption experiments were performed at 393 K using N_2 as purge gas at a flow rate of 300 mL/min. The N_2 was cut off when the volume fraction of CO_2 in the outlet gas was less than 0.5%.

3. RESULTS AND DISCUSSION

3.1. Solubility of CO_2 in Pure BP. Figure 3 shows the solubility of CO_2 in pure BP. It can be seen that the solubility is
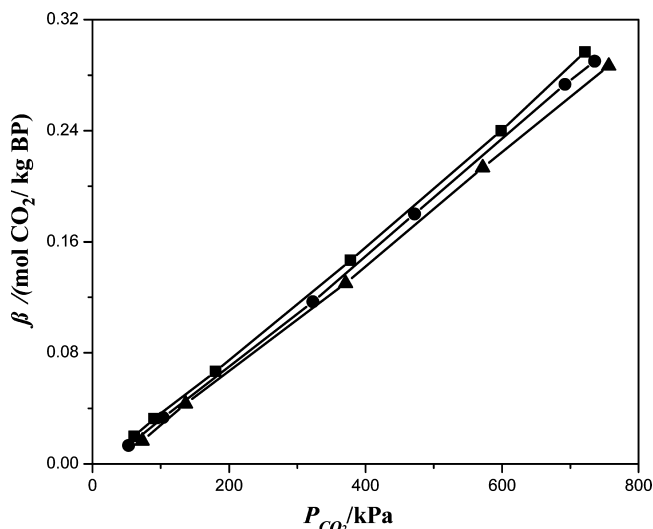

Figure 3. Solubility of CO_2 in pure BP at different temperatures: β, CO_2 loadings, referred to as mol CO_2/kg BP; ■, at 303 K; ●, at 313 K; ▲, at 323 K.

proportional to the partial pressure of CO_2 in the vapor phase, and generally is consistent with Henry's Law:

$$x_B = \frac{P_B}{H_B} \tag{1}$$

where x_B is the solubility of the solute B in the solvent, mol/kg, P_B is the partial pressure in the vapor phase of B, kPa, and H_B is the Henry coefficient, kPa/(mol/kg).

Figure 3 shows that at the same partial pressure of CO_2, the solubility of CO_2 in BP at low temperature is a little higher than that at high temperature, and the CO_2 loading increases with increasing CO_2 partial pressure. The data are presented in Table 1.

Table 2 shows the Henry coefficient of CO_2 in pure BP at different temperatures, proclaiming that H increases with increasing temperature. The linear fit is carried out by ln H on $1/T$, which can be written as follows:

Table 1. Data of CO_2 Solubility in Pure BP Solutions at 303 K, 313 K, and 323 K[a]

303 K		313 K		323 K	
P_{CO_2}/kPa	β	P_{CO_2}/kPa	β	P_{CO_2}/kPa	β
61.00	0.020	53.00	0.013	73.00	0.017
90.30	0.033	103.50	0.033	136.71	0.043
180.32	0.067	322.70	0.117	370.90	0.130
378.04	0.147	471.87	0.180	571.87	0.213
598.83	0.240	692.25	0.273	756.38	0.287
721.27	0.297	735.49	0.290		

[a]CO_2 loading (β) is referred to as mol CO_2/kg BP. The standard uncertainty u for temperature is $u(T)$ = 1 K, for CO_2 partial pressure $u(P_{CO_2})$ = 0.2 kPa, and for CO_2 loading (mol CO_2/kg BP) $u(\beta)$ = 0.001.

Table 2. Henry Coefficient of CO_2 in BP at Different Temperatures[a]

temp/K	H/(kPa/mol/kg)
303	2403.4
313	2465.9
323	2537.9

[a]The standard uncertainty u for temperature is $u(T)$ = 1 K, and for Henry coefficients $u(H)$ = 10 kPa/mol/kg.

$$\ln H = 8.57 - \frac{237.1}{T} \tag{2}$$

Figure 4 shows the comparison of solubility of CO_2 between pure BP and H_2O.[26] Obviously BP has larger physical
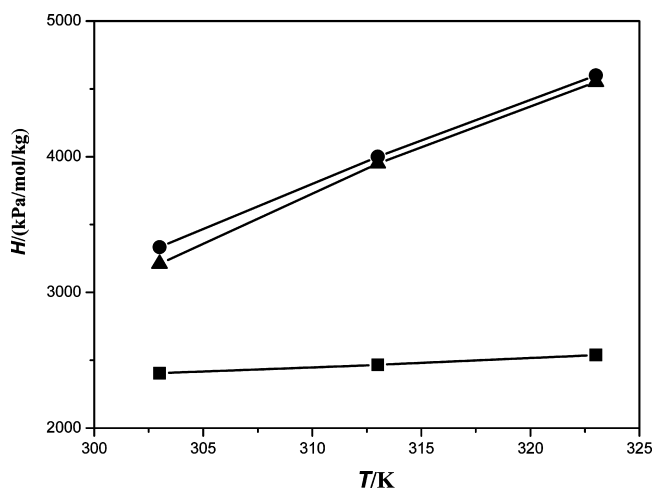

Figure 4. Comparison of Henry coefficients of CO_2 in H_2O and BP: ■, BP in this work; ●, H_2O in Li et al.;[26] ▲, H_2O in this work.

absorption capacity than H_2O, and hence BP as the nonaqueous media in the absorption system will enhance the physical loading of CO_2. The measured Henry coefficients of H_2O in this work are in good agreement with published data within 5 %.

3.2. Solubility of CO_2 and Absorption Rate in Different Amine Solutions. Figure 5 shows the solubility of CO_2 in AEEA + H_2O, AEEA + BP, AEEA + DEG, AEEA + TEG, and the solubility of CO_2 in AEEA + H_2O which was measured by Svendsen et al.[27] The data of CO_2 solubility in these solvents at 313 K are presented in Table 3. We can see in Figure 5 that the solubility of CO_2 in AEEA + H_2O in this work is in good agreement with the study of Svendsen et al.[27] at the CO_2 partial pressure (>2 kPa), the CO_2 loading at the same CO_2 partial pressure is in the order AEEA + H_2O > AEEA + BP > AEEA + TEG > AEEA + DEG, and the CO_2 loadings of these solutions increase with increasing CO_2 partial pressure. It can be explained that in nonaqueous amine solution, CO_2 hydration is completely avoided, and bicarbonate or carbonate formation is negligible.[28] The dissociation of protonated amine and formation of carbamate also do not happen, and only the intermediate zwitterion forms according to the zwitterion mechanism which was first proposed by Caplow[29] and reintroduced by Danckwerts.[30] That is why the CO_2 loading is reduced in nonaqueous solution compared with aqueous solution.
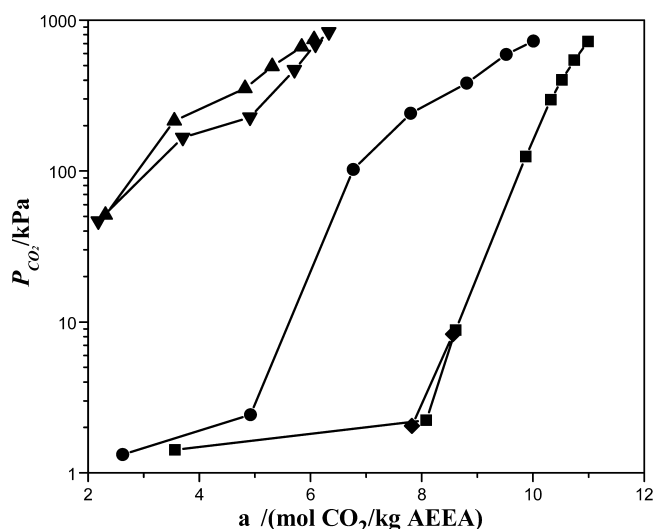
Figure 5. Solubility of CO_2 in solutions of 30 wt % AEEA + 70 wt % H_2O, 30 wt % AEEA + 70 wt % BP, 30 wt % AEEA + 70 wt % DEG, and 30 wt % AEEA + 70 wt % TEG at 313 K. α, CO_2 loadings; ■, AEEA + H_2O in this work; ●, AEEA + BP in this work; ▲, AEEA + DEG in this work; ▼ AEEA + TEG in this work; ◆, AEEA + H_2O in Svendsen at al.[27]

Figure 6. Absorption rate of 30 wt % AEEA + 70 wt % H_2O, 30 wt % AEEA + 70 wt % BP, 30 wt % AEEA + 70 wt % DEG, and 30 wt % AEEA + 70 wt % TEG solutions 300 g at 313 K. ■, AEEA + H_2O in this work; ●, AEEA + BP in this work; ▲, AEEA + DEG in this work; ▼, AEEA + TEG in this work.

That CO_2 loading of AEEA + BP is the highest in the three nonaqueous systems may be due to the difference of their viscosities. The high viscosity of nonaqueous solvent goes against the formation of ions and the chemical absorption of CO_2.[31] The viscosity of BP is relatively low (298 K, 5.037 mPa·s) and has a small difference with that of water (298 K, 0.89 mPa·s), while the viscosities of DEG (298 K, 30 mPa·s) and TEG (298 K, 42 mPa·s) are rather high. The relative low viscosity of BP as the solvent makes CO_2 easier to diffuse in the solution and react with AEEA rather than DEG and TEG.

Figure 6 shows the absorption rate of AEEA + H_2O, AEEA + BP, AEEA + DEG, and AEEA + TEG. In Figure 6, "step 1" indicates that it takes about 14 s for the reactor to reach the maximum pressure after the introduction of CO_2. The time depends on the velocity of CO_2 to be introduced to the reactor. The less time step 1 takes, the more accurate the absorption rate is. The absorption rate is in the order AEEA + H_2O > AEEA + BP > AEEA + DEG > AEEA + TEG. This can be explained because the nonaqueous media mixed into AEEA
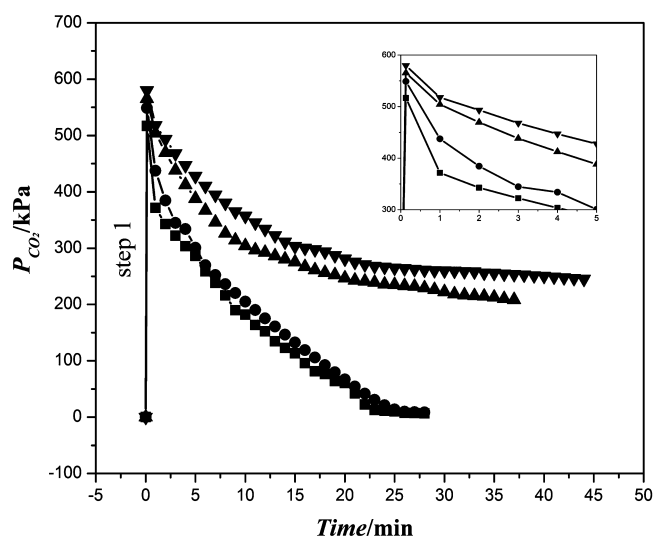
hybrid solvent reduces the diffusivity and solubility characteristics of the solvent, and this results in a reduced CO_2 absorption rate.[14] As stated above, BP blended with AEEA displays better absorption performance than DEG and TEG.

3.3. Effect of Absorption Temperature. Figure 7 shows CO_2 solubility in 300 g of 30 wt % AEEA + 70 wt % BP solutions at 303, 313, and 323 K, and the data are presented in Table 4. It can be seen from Figure 7 that the CO_2 loading is sensitive to the change of CO_2 partial pressure and temperature. The two factors affect the equilibrium of chemical reaction through the change of CO_2 solubility. The decrease of temperature and increase of CO_2 partial pressure lead to higher physical absorption and thus promote the chemical reaction equilibrium moving to CO_2 absorption, while the opposite leads to lower physical absorption and thus promotes the chemical reaction equilibrium moving to CO_2 desorption.[16]

3.4. The Desorption Rate and Cyclic Capacity. Figure 8 shows the stability of 300 g of 30 wt % AEEA+ 70 wt % BP in the four cycles absorption−desorption experiments. We can see in Figure 8 that during the desorption process, about 20%

Table 3. Data of CO_2 Solubility in Solutions of 30 wt % AEEA + 70 wt % H_2O, 30 wt % AEEA + 70 wt % BP, 30 wt % AEEA + 70 wt % DEG, and 30 wt % AEEA + 70 wt % TEG at 313 K[a]

30 wt % AEEA + 70 wt % H_2O			30 wt % AEEA + 70 wt % BP			30 wt % AEEA + 70 wt % DEG			30 wt % AEEA + 70 wt % TEG		
P_{CO_2}/kPa	α	x_1	P_{CO_2}/kPa	α	x_2	P_{CO_2}/kPa	α^1	x_3	P_{CO_2}/kPa	α	x_4
1.42	3.56	1.07	1.32	2.62	0.79	51.17	2.31	0.69	46.61	2.18	0.65
2.23	8.28	2.48	2.43	4.92	1.48	215.01	3.55	1.06	166.38	3.70	1.11
8.82	8.61	2.58	102.34	6.77	2.03	353.02	4.82	1.44	226.87	4.91	1.47
124.63	9.87	2.96	241.56	7.80	2.34	494.61	5.31	1.59	467.92	5.71	1.71
297.29	10.32	3.10	381.9	8.81	2.64	665.29	5.84	1.75	689.92	6.09	1.83
401.55	10.52	3.16	591.13	9.52	2.86	744.42	6.06	1.82	833.29	6.33	1.90
542.9	10.74	3.22	724.47	10.01	3.00						
718.9	10.99	3.30									

[a]CO_2 loading is referred to as mol CO_2/kg AEEA. x_n−x_1, x_2, x_3, and x_4 are moles of CO_2 solute per kilogram of AEEA + H_2O, AEEA + BP, AEEA + DEG, and AEEA + TEG solvent, respectively. The standard uncertainty u for temperature is $u(T)$ = 1 K, for CO_2 partial pressure $u(P_{CO_2})$ = 0.2 kPa, for mass fraction of the solution $u(X)$ = 0.1 wt %, for CO_2 loading (mol CO_2/kg AEEA) $u(\alpha)$ = 0.01, and for mol CO_2/total kg of solvent $u(x_n)$ = 0.01.
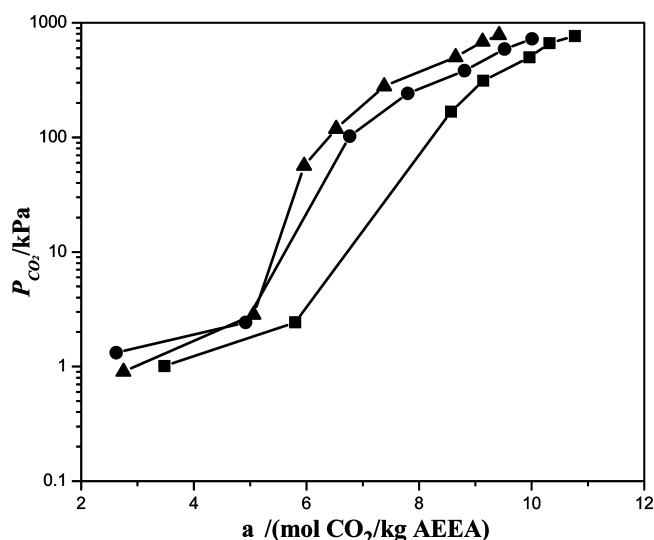
Figure 7. Solubility of CO_2 in aqueous 300 g of 30 wt % AEEA + 70 wt % BP solution at different temperatures: α, CO_2 loadings; ■, at 303 K; ●, at 313 K; ▲, at 323 K.

Figure 8. Absorption and desorption CO_2 loading of 300 g of 30 wt % AEEA + 70 wt % BP in four cycles absorption−desorption experiments. α, CO_2 loadings; n, cycle index; right to left slanted stripes, absorption amount; left to right slanted stripes, desorption amount.

absorbed CO_2 in the solution will not be stripped out, and the desorption amount of the AEEA + BP solutions remain essentially constant during the four cycles absorption−desorption experiments. The reason is that during the absorption process, some absorbed CO_2 forms the intermediate zwitterion which is relatively stable and will not be totally stripped out during the desorption process.

Figure 9 shows the desorption rate of 30 wt % AEEA + 70 wt % H_2O, and 30 wt % AEEA + 70 wt % BP solution at 393 K. It can been seen that the desorption rate of AEEA+ BP is about 1.3 times faster than AEEA + H_2O, and AEEA + BP solution needed 30% less time than AEEA + H_2O solution to reach reaction end point. Because of the substitution of water by BP, a large amount of thermal energy is saved when the solvent is heated to stripping temperature and no latent heat of water vaporization is needed, which accelerates the CO_2 desorption process.

4. CONCLUSIONS

A mixture system of AEEA with BP was developed to reduce energy consumption for CO_2 stripping from amine solution. The CO_2 loading at the same CO_2 partial pressure was in the order AEEA + H_2O > AEEA + BP > AEEA + TEG > AEEA +
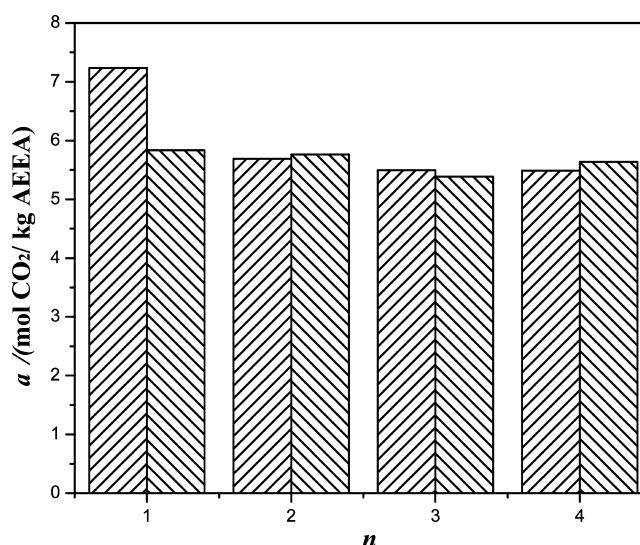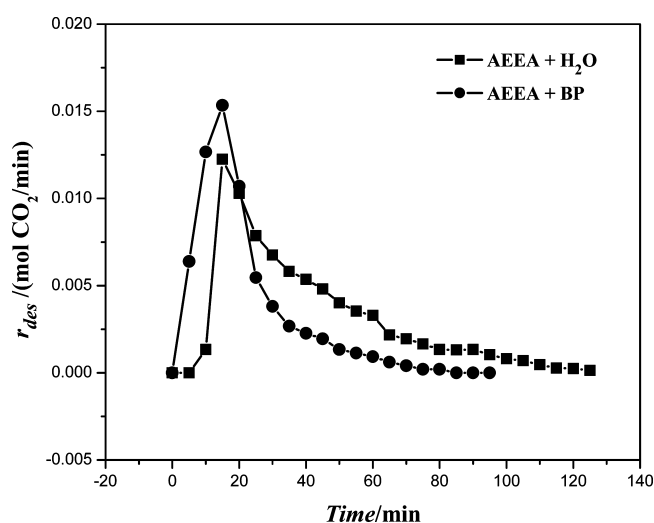

Figure 9. Desorption rate (r_{des}) of 300 g of 30 wt % AEEA + 70 wt % H_2O, and 30 wt % AEEA + 70 wt % BP solutions at 393 K: ■, AEEA + H_2O in this work; ●, AEEA + BP in this work.

Table 4. Data of CO_2 Solubility in 30 wt % AEEA + 70 wt % BP Solution at 303 K, 313 K, and 323 K[a]

303 K			313 K			323 K		
P_{CO_2}/kPa	α	x_2	P_{CO_2}/kPa	α	x_2	P_{CO_2}/kPa	α	x_2
1.01	3.48	1.04	1.32	2.62	0.79	0.90	2.75	0.82
2.43	5.80	1.74	2.43	4.92	1.48	2.84	5.06	1.52
167.79	8.57	2.57	102.34	6.77	2.03	56.64	5.96	1.79
312.8	9.14	2.74	241.56	7.80	2.34	119.06	6.53	1.96
498.61	9.96	2.98	381.9	8.81	2.64	279.66	7.38	2.21
666.2	10.32	3.10	591.13	9.52	2.86	503.08	8.65	2.60
765.61	10.77	3.23	724.47	10.01	3.00	685.87	9.13	2.74
						780.18	9.42	2.83

[a]CO_2 loading is referred to as mol CO_2/kg AEEA. x_2 is moles of CO_2 solute per kilogram of AEEA + BP solvent. The standard uncertainty u for temperature is $u(T)$ = 1 K, for CO_2 partial pressure $u(P_{CO_2})$ = 0.2 kPa, for mass fraction of the solution $u(X)$ = 0.1 wt %, for CO_2 loading (mol CO_2/kg AEEA) $u(\alpha)$ = 0.01, and for mol CO_2/total kg of AEEA + BP $u(x_2)$ = 0.01.

DEG, and the CO_2 loading of these solutions increased with increasing the vapor pressure of CO_2. Moreover, the solubility of CO_2 in AEEA + BP solution significantly decreased with increasing reaction temperature. In the desorption process, the desorption rate of AEEA + BP solution was about 1.3 times faster than that of the AEEA + H_2O solution, and the AEEA + BP solution needed 30 % time less than the AEEA + H_2O solution to reach the reaction end point. As a result significant energy consumption as well as environmental pollution were expected to be avoided in a CO_2 desorption process. The AEEA + BP solution displayed a stable absorption−desorption property in the cycle experiments, which indicated its potential in industrial application. The detailed kinetics of the solution for the mass transfer is under investigation by the authors.

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Notes

The authors declare no competing financial interest.

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