Effect of Water on Solubility of Carbon Dioxide in (Aminomethanamide + 2-Hydroxy-*N*,*N*,*N*-trimethylethanaminium Chloride)

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In this study, the densities of (aminomethanamide + 2-hydroxy-*N*,*N*,*N*-trimethylethanaminium chloride + water) and the solubilities of CO_2 in (aminomethanamide + 2-hydroxy-*N*,*N*,*N*-trimethylethanaminium chloride) with different varying mole fractions of water at temperatures of (303, 308, and 313) K are reported. The common name for (aminomethanamide + 2-hydroxy-*N*,*N*,*N*-trimethylethanaminium chloride) is reline. The densities are linear (AAD % is 0.22 %) with respect to reline mass fraction at each temperature. The logarithms of Henry's law constants were correlated using the two-suffix Margules model as a function of reline mole fraction with an AAD of 3.44 %. The results showed that CO_2 solubility in reline decrease with an increase water content. Hence water can serve as an antisolvent to strip CO_2 dissolved in reline. The absorption of CO_2 in (aminomethanamide + 2-hydroxy-*N*,*N*,*N*-trimethylethanaminium chloride + water) at low pressures is found to be endothermic at water content. However, the absorption becomes exothermic if the water content increases to a mole fraction greater than 0.769.

Introduction

Most of the observed increase in globally averaged temperatures in the twentieth century is due to the increase in atmospheric concentration of the anthropogenic greenhouse gas carbon dioxide. For the purpose of diminishing the quantity of CO_2 contained in the atmosphere, current CO_2 capture systems typically utilize amine-based absorption agents, such as monoethanolamine, diethanolamine, and 2-amino-2-methyl-L-propanol.[1] The theories of absorption have been well documented,[2,3] and thermophysical properties required for process design have been reported extensively, for example, CO_2 solubility,[4−6] diffusivity,[5,7] density,[8−10] viscosity,[8] and heat capacity.[11,12]

Although the above-mentioned agents react quickly and have a high capacity for CO_2, they are typically corrosive and possibly environmentally malign. Furthermore, a temperature swing absorption-stripping process is usually employed to recover the amine solution that in itself requires much energy. There is still a need to find absorbents able to capture CO_2 without these disadvantages. The potential of ionic liquids as green solvents has received much attention in recent years.[13] They have the advantages of being nonflammable, having low vapor pressure, and wide liquid range. Several researchers have reported CO_2 solubility in ionic liquids including imidazolium-based types such as 1-butyl-3-methylimidazolium tetrafluoroborate ([Bmim][BF_4])[14−16] and 1-butyl-3-methylimidazolium hexafluorophosphate ([Bmim][PF_6]).[14,15,17−19] Nevertheless, Jastorff et al.[20,21] evaluated the eco-toxicological risk profiles[22] of some conventional ionic liquids and suggested that while these ionic liquids are of low volatility the environmental impact in terms of toxicity remains uncertain.

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Recently, Abbott and co-workers developed a class of ionic liquids known as "deep eutectic solvent (DES)",[23−25] by mixing a quaternary ammonium salt and a hydrogen bond donor, both of which may have high melting points, to form a eutectic mixture with a substantially lower melting point. For example, the melting points of aminomethanamide and 2-hydroxy-*N*,*N*,*N*-trimethylethanaminium are 408 K and 578 K, but the freezing point of a eutectic mixture made of these two components mixed by the mole ratio equal to 2:1 is 285 K. These DESs are easy to prepare and relatively cheap. Many of their constituents are present in organisms naturally and are biodegradable thus posting a lower environmental risk. They have been used to promote the chemical reaction,[26] the synthesis of porous zeolites,[27] and the electrolytic solvent for electrodepostion,[28] used as solvents for electrolytes in a dye-sensitive solar cell.[29] Currently, Li et al.[30] measured CO_2 solubilities in DESs composed of various mole ratios of aminomethanamide and 2-hydroxy-*N*,*N*,*N*-trimethylethanaminium at pressures >1 MPa and from these extrapolated their Henry's law constants. It shows that these DESs are capable of absorbing CO_2.

One important characteristic of DESs is that both constituents are water soluble. If water is added, the solvent power of the DES will be reduced. Hence water acts as a good antisolvent to drive the solute out from rich solutions. Thus a temperature swing absorption-stripping process may be avoided and energy expenditure of CO_2 capture reduced. In this study, we investigated the CO_2 solubility and Henry's law constants in (aminomethanamide + 2-hydroxy-*N*,*N*,*N*-trimethylethanaminium chloride + water) {DES (aminomethanamide + 2-hydroxy-*N*,*N*,*N*-trimethylethanaminium chloride) provided by ScioniX[31]} as a function of water contents at a pressure of 0.1 MPa. These data are essential for the evaluation and development of a concentration swing absorption-stripping process for capturing CO_2.

10.1021/je900078k CCC: $40.75
Published on Web 04/01/2009

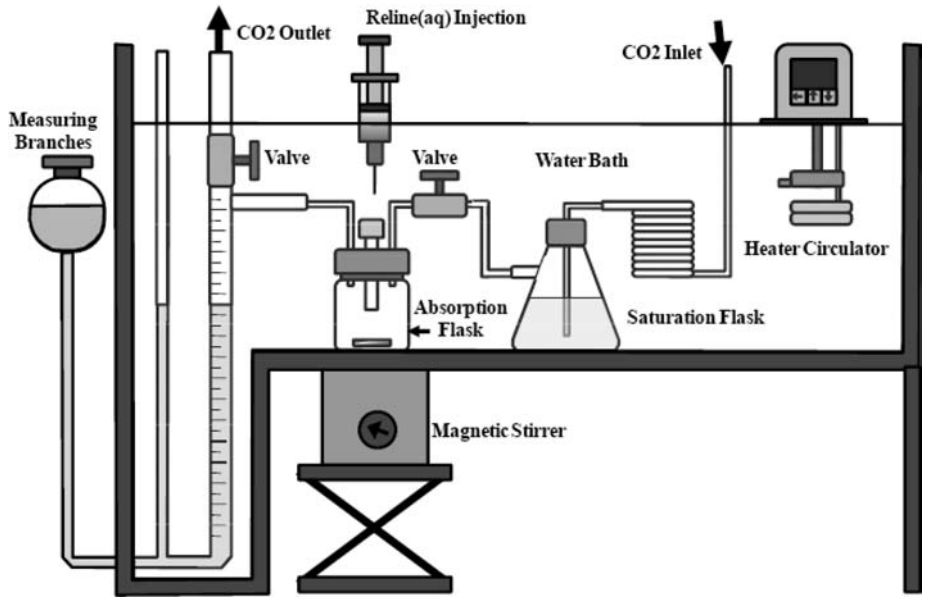

Figure 1. Schematic of the CO_2 solubility apparatus.

Table 1. Density of Reline with Different Water Content

	reline/mass %										
T/K	99.44	98.81	96.1	95.06	90.71	85.4	68.5	66.1	44.9	22.7	H_2O
293.15		1.2022		1.1931	1.1834	1.1709	1.1350	1.1296	1.0838	1.0397	0.9980
303.15	1.1953	1.1968	1.18919	1.1873	1.1779	1.1656	1.1300	1.1246	1.0792	1.0360	0.9957
308.15	1.1925	1.1939	1.18670	1.1845	1.1752	1.1630	1.1274	1.1221	1.0768	1.0339	0.9942
313.15	1.1899	1.1909	1.18422	1.1817	1.1725	1.1603	1.1248	1.1195	1.0743	1.0318	0.9924
323.15	1.1848	1.1852	1.17918	1.1763	1.1671	1.1551	1.1195	1.1142	1.0692	1.0271	0.9884
333.15	1.1798	1.1796	1.17415	1.1709	1.1618	1.1497	1.1142	1.1088	1.0637	1.0220	0.9837
343.15	1.1747	1.1741	1.16917	1.1655	1.1565	1.1443	1.1087	1.1034	1.0581	1.0163	0.9781
353.15	1.1697	1.1687	1.16390	1.1601	1.1511	1.1389	1.1030	1.0976	1.0522	1.0102	0.9717

Experimental Section

Chemicals. The deep eutectic solvent reline is supplied by ScioniX.[31] It is a mixture of aminomethanamide (commonly known as urea) and 2-hydroxy-*N*,*N*,*N*-trimethylethanaminium chloride (commonly known as choline chloride) with a mole ratio of 2 urea-to-1 choline chloride, and this term will be used to define the mixture herein after. The reported purity is greater than 98 %. Reline was dried at 343 K for 48 h in an evacuated oven and stored in the drybox before use. The deionized water was produced by a Barnstead Thermodyne, model Easy Pure 1052. All aqueous reline solutions were degassed ultrasonically with a Branson, model 3510, at a temperature of 313 K with a Ulvac, model GVD-050A, for a time of 1 h. The CO_2 was supplied by BOC Lien-Hwa Ind. Gases Co., Ltd., with a mole fraction purity of greater than 0.999.

Measurement of CO_2 Solubility. The experimental procedure and the method of analysis have been reported by Li and Lai.[5] The principle is similar to that reported by Haimour and Sandall.[32] The apparatus is made up of a saturation flask, an absorption flask with a magnetic stirrer, and a set of burets with branches filled with mercury as shown in Figure 1. The system was immersed in a water bath at a constant temperature that was monitored by a thermometer (CROPICO model 3002 PT100) with an uncertainty of ± 0.01 °C. The whole apparatus was purged by CO_2 for a time of 1 h before the experiment. Then the inlet and outlet valves were closed. A known volume of reline with the known mole fraction of water, as determined by Mettler Toledo Karl Fisher titrator model DL31, was injected into the absorption flask. CO_2 and reline with the specific water were mixed with a magnetic stirrer. The volume change of the system can be determined by the levels of the mercury in the buret. The system was determined to be in equilibrium after the mercury level remained unchanged for a time of 2 h. All the experiments

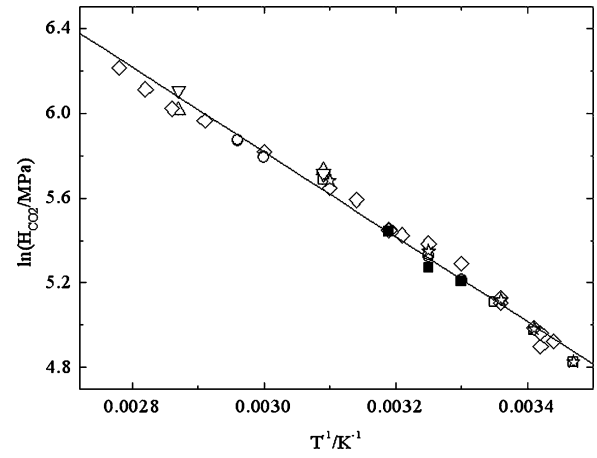

Figure 2. Henry's law constant of CO_2 in water as a function of temperature: □, Gibbs and Van Ness;[34] △, Schulze and Prausnitz;[35] ▽, Zawisza and Malesinska;[36] ◇, Versteeg and van SwaaiJ;[7] ○, Zheng et al.;[37] ☆, Dalmolin et al.;[38] ■, this work; —, calculated by eq 5.

Table 2. Solubility and Henry's Law Constant of CO_2 in Reline with Different Water Content

303 K			308 K			313 K		
reline/wt %	$10^4\ x_{CO_2}$	H_{CO_2}/MPa	reline/wt %	$10^4\ x_{CO_2}$	H_{CO_2}/MPa	reline/wt %	$10^4\ x_{CO_2}$	H_{CO_2}/MPa
0.00	5.2 ± 0.2	183 ± 10	0	4.6 ± 0.3	194 ± 6	0	4.0 ± 0.2	230 ± 10
47.4 ± 0.5	8.3 ± 0.2	114 ± 3	43.6 ± 0.1	7.4 ± 0.2	128 ± 4	48.26 ± 0.07	7.5 ± 0.8	116 ± 3
79.4 ± 0.2	13.9 ± 0.5	68 ± 3	69.2 ± 0.2	11.2 ± 0.3	85 ± 2	74.59 ± 0.03	13.4 ± 0.2	69.0 ± 0.9
86.2 ± 0.3	17 ± 1	58 ± 4	85.43 ± 0.04	17.2 ± 0.5	55 ± 1	87.01 ± 0.03	18.9 ± 0.7	49 ± 2
91.85 ± 0.06	21 ± 2	45.7 ± 0.3	90.91 ± 0.04	20.4 ± 0.6	47 ± 1	91.82 ± 0.08	24.5 ± 0.5	37.5 ± 0.8
95.58 ± 0.02	23 ± 1	41 ± 3	95.03 ± 0.08	24.8 ± 0.3	38.0 ± 0.6	96.74 ± 0.05	29.0 ± 0.6	31.9 ± 0.6
99.10 ± 0.05	27 ± 1	36 ± 2	98.94 ± 0.03	29.8 ± 0.6	31.8 ± 0.5	98.87 ± 0.03	33 ± 4	29 ± 3

at the same conditions were repeated between 3 and 5 times to ensure reproducibility within 3 %.

At constant temperature and pressure, the volume of CO_2 absorbed can be determined by the volume change of the system and the volume of the solution

$$\Delta V_{CO_2} = V_{Initial} + V_{Injected} - V_{Final} \quad (1)$$

where ΔV_{CO_2} is the volume of CO_2 absorbed; $V_{Initial}$ and V_{Final} are the initial and final volume of the gas phase at 0.1 MPa;

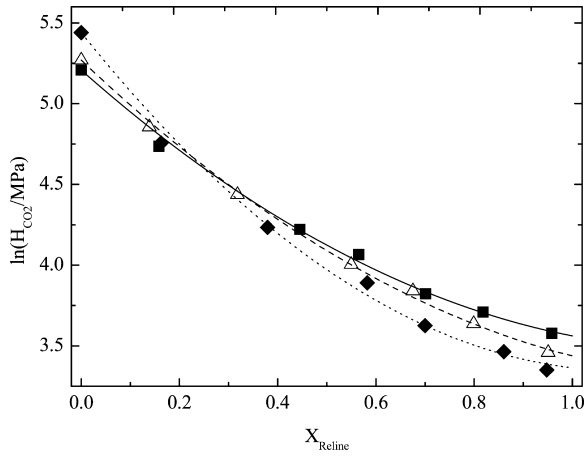

Figure 3. Henry's law constant of CO_2 in reline as a function of mole fraction: ■, 303 K; △, 308 K; ◆, 313 K; (lines) —, 303 K; - - -, 308 K; ·····, 313 K; correlated by eq 6.

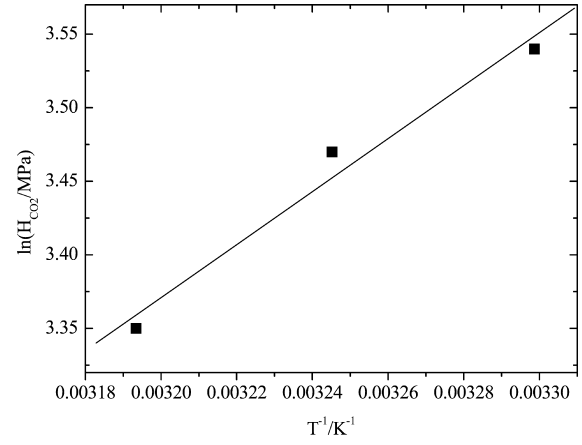

Figure 4. Henry's law constant of CO_2 absorption in pure reline as a function of temperature: ■, reline, 303 K, 308 K, and 313 K; —, correlated by eq 8.

$V_{Injected}$ is the injected volume of reline with different contents which can be expressed as the following relation

$$V_{Injected} = \frac{W_{Injected}}{\rho_{Solvent}} \quad (2)$$

where $W_{Injected}$ is the mass of injected samples and $\rho_{solvent}$ measured by Anton Paar Stabinger (model SVM 3000) is the density of reline with different water content. The temperatures were kept the constant to ± 0.02 K by the thermostat. The uncertainty in density was ± 0.0002 g·cm^{-3}. The mole of CO_2 absorbed (Δn_{CO_2}) was calculated by eq 3

$$\Delta n_{CO_2} = \frac{P_{CO_2}\Delta V_{CO_2}}{RT} \quad (3)$$

where P_{CO_2} is CO_2 partial pressure (0.1 MPa) and T is the experimental temperature.

Results and Discussion

The density of water was measured in the same temperature range to validate the experimental procedure. The average absolute deviation percentage (AAD %) of the calibration differed from literature data[33] by about ± 0.02 %.

Therefore, the density data of reline with water in the temperature range from (293 to 353) K were measured and listed in Table 1. The densities determined are linear with respect to reline mass fraction at each temperature. The data can be well represented by

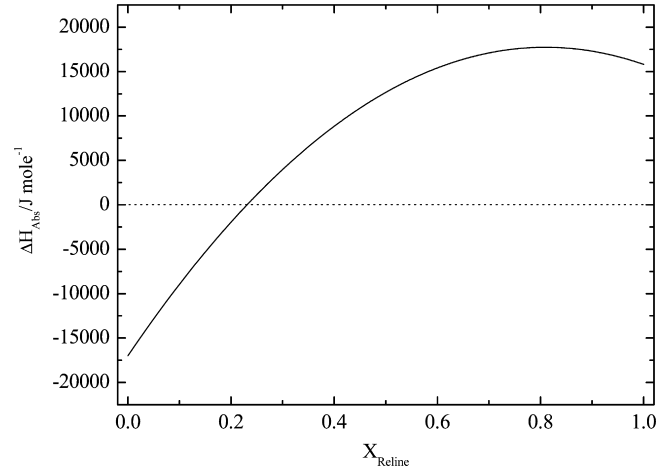

Figure 5. Enthalpy of CO_2 absorption in reline as a function of mole fraction: —, calculated by eq 9; ····, y-axis = zero.

$$\rho_{\text{mixture}} = w_{\text{reline}}\rho_{\text{reline}} + w_{\text{water}}\rho_{\text{water}} = w_{\text{reline}}(a_0 + a_1T + a_2T^2) + w_{\text{water}}\rho_{\text{water}} \quad (4)$$

with an AAD of ± 0.22 %. In eq 4, w_{reline} is the mass fraction of reline and w_{water} is the mass fraction of water. The parameters determined by regression are as follows: $a_0 = 1.4$ g·cm^{-3}, $a_1 = -7.903 \cdot 10^{-4}$ g·cm^{-3} K^{-1}, and $a_2 = 3.853 \cdot 10^{-7}$ g·cm^{-3}·K^{-2}.

To ensure the validity of the experimental procedure and method, the solubilities of CO_2 in pure water were measured. The Henry's law constants at temperatures of (303, 308, and 313) K were found to be (183 ± 10) MPa, (194 ± 6) MPa, and (230 ± 10) MPa. These results are compared with the literature data (Gibbs and Van Ness, 1971;[34] Schulze and Prausnitz, 1981;[35] Zawisza and Malesinska, 1981;[36] Versteeg and van Swaaij, 1988;[7] Zheng et al., 1997;[37] and Dalmolin et al., 2006[38] in Figure 2. On the basis of the available data of CO_2 solubility in water, Versteeg and van Swaaij[7] proposed the following correlation

$$\ln(H_{CO_2,\text{water}}/\text{MPa}) = 10.843 - \frac{16994/\text{J}\cdot\text{mol}^{-1}}{RT} \quad (5)$$

where R is the gas constant. The AAD of CO_2 solubilities in water obtained by this work compared to eq 5 is 2.42 %.

Since the density data were analyzed, the solubilities and Henry's law constants of CO_2 in reline with different water contents at temperatures of (303, 308, and 313) K are listed in Table 2. A two-suffix Margules model proposed by O'Connell and Prausnitz[39] was used to correlate the Henry's law constants with

$$\ln H_{CO_2,\text{solvent}} = x_{\text{reline}} \cdot \ln H_{CO_2,\text{reline}} + x_{\text{water}} \cdot \ln H_{CO_2,\text{water}} - a_{13} \cdot x_{\text{reline}} \cdot x_{\text{water}} \quad (6)$$

and the results shown in Figure 3. The overall AAD of eq 6 is 3.44 %. The so-called interaction term a_{13} is temperature dependent and given by

$$a_{13} = 21.96 - \frac{6374}{T/\text{K}} \quad (7)$$

Using eq 6, the Henry's law constant of CO_2 in pure reline was extrapolated, and its temperature dependence was correlated with eq 8. The results are shown in Figure 4

$$\ln(H_{CO_2}/\text{MPa}) = -2.392 + \frac{14974/\text{J}\cdot\text{mol}^{-1}}{RT} \quad (8)$$

The enthalpy of absorption of CO_2 ($\Delta H_{\text{Abs,solvent}}$) in reline with different water contents can be estimated by taking the derivative of $\ln H_{CO_2,\text{solvent}}$ with respect to $1/T$ to give

$$\Delta H_{\text{Abs,solvent}}/\text{J}\cdot\text{mol}^{-1} = b_0 + b_1x_{\text{reline}} + b_2x_{\text{reline}}^2 \quad (9)$$

where the parameters are given: $b_0 = -2044$ J·mol^{-1}, $b_1 = 10319$ J·mol^{-1}, and $b_2 = -6374$ J·mol^{-1}. The results, shown in Figure 5, demonstrate a crossover point at the mole fraction of reline of about 0.231, above which the absorption is endothermic and below which the absorption is exothermic.

Conclusions

In this study, the densities of reline/water mixtures and the solubilities of CO_2 in reline with different water contents at temperatures of (303, 308, and 313) K were measured. The results showed that CO_2 solubility in reline will decrease with an increase in water content. Hence, water can serve as an antisolvent to drive out CO_2 dissolved in reline. According to eq 9, the enthalpy of absorption of CO_2 in reline at low pressure is endothermic. This usually implies lack of solvation between solute and solvent and large difference between cohesive energy density of the solute and that of the solvent.[40] However, using data at high pressure (> 1 MPa), Li et al.[30] found that the absorption of CO_2 is exothermic. So there must be a crossover point of temperature dependence from low to high pressure. According to eq 9, the absorption of CO_2 is endothermic at high reline and low water concentrations. If water content increases to beyond 76.9 %, the absorption becomes exothermic. These interesting temperature and concentration dependences make possible the development of green and energy-saving absorption and stripping processes for CO_2 capture.

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Received for review January 19, 2009. Accepted March 2, 2009.

JE900078K