

Solubility and density of carbon dioxide in different aqueous alkanolamine solutions blended with 1-butyl-3-methylimidazolium acetate ionic liquid at high pressure

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ABSTRACT

Using a static high pressure equilibrium cell, the new set of the experimental data is obtained for solubility of carbon dioxide in the aqueous mixtures of different type of alkanolamines such as N-methyldiethanolamine (MDEA), Diethanolamine (DEA), Diisopropanolamine (DIPA), 2-amino-2-methyl-1-propanol (AMP), and 1-butyl-3-methylimidazolium acetate ionic liquid [bmim][acetate]. The solubility of CO₂ in the aqueous MDEA + [bmim][acetate], DEA + [bmim][acetate], DIPA + [bmim][acetate] and AMP + [bmim][acetate] solutions with 0–10 wt% of ionic liquid and 30–40 wt% alkanolamine are carried out at 1 to 40 bar and 323.15 K. Moreover, the temperature dependency of density for these solutions is measured from 293.15 to 343.15 K with 10 K intervals.

The results of the CO₂ solubility are represented by partial pressure of CO₂ against the loading and mole fraction of CO₂. The solubility of the CO₂ in all of the aqueous alkanolamine + ionic liquid solutions decreases with increasing weight percent of ionic liquid. Also, the results show that the maximum decrease of CO₂ loading belongs to addition of [bmim][acetate] to the aqueous MDEA solution. Finally, the results of density of the alkanolamine + ionic liquid solutions present that the density enhances with increasing concentration of [bmim][acetate] and reduces linearly with enhancing temperature.

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1. Introduction

Aqueous alkanolamine solutions are widely used for the removal of acid gases, such as CO₂ and H₂S from industrial, flue and natural gases. The alkanolamines as a solvent are classified into primary, secondary, and tertiary amines such as monoethanolamine (MEA), diethanolamine (DEA), N-methyldiethanolamine (MDEA), respectively. A different category of alkanolamines, which is known as sterically hindered amines, such as 2-amino-2methyl-1-propanol (AMP) has also been suggested as commercially attractive new absorbent [1] so that it is usually blended with the other alkanolamines to achieve maximum solubility of acid gases. The alkanolamine solutions are generally suitable for highly removal of acid gases (CO₂ and H₂S). However, they have the disadvantage of requiring a large expenditure of energy for regeneration. Moreover, corrosion is a major concern when using the alkanolamines, particularly MEA and DGA.

In overall, the alkanolamines present several disadvantages such as volatility, toxicity, degradation, transfer of water into the gas stream during desorption stage and high energy consumption. In addition, using blends of the aqueous alkanolamine systems makes the process

economically expensive. In recent years, from environmental point of view using green solvent such as ionic liquid (IL) allows one to reduce the drawback of usage of the conventional alkanolamine solutions. Ionic liquids are organic salts that are liquids below 100 °C even at room temperature (RTILs) and present the excellent properties such as negligible vapor pressure even at elevated temperatures, thermal and chemical stability, and liquid at a wide range of temperature, nonflammable and tunable nature. Moreover, ILs may be used as a green solvent for absorption of CO₂ and interesting candidate for a variety of applications. Thus, negligible vapor pressure of ILs permits one to replace them with volatile organic compounds (VOCs) which introduce several health, environmental, and economic concerns in numerous industrial applications [2,3].

The solubility of gases especially CO₂ in ILs at various conditions (concentration, temperature and pressure) may be used as a potential application in industrial natural gas treating processes. In the past decade, several various researches have been published the CO₂ solubility data in the different pure ILs so that presently most ILs showed a lower CO₂ loading capacity as compared to the aqueous alkanolamine solutions [4].

The CO₂ solubility in ILs is dramatically increase by incorporating amine functional groups to ILs that is an obvious application of design of task-specific ionic liquids (TSILs) [5–8]. Nevertheless, the use of TSILs for CO₂ absorption shows some drawbacks such as high viscosities at ambient temperature that is even higher with

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absorption of CO₂ [9]. Moreover, the preparation of this type of ILs would require several synthetic steps [5] that limit their commercial viability [10]. Alternatively, the mixed IL-alkanolamine solutions can be used to improve the performance of the CO₂ solubility in absorption processes, because IL such as Imidazolium-based RTILs presents less heat capacity compared to water, i.e. one-third the heat capacity of water ($1.30 \text{ J g}^{-1} \text{ K}^{-1}$ vs $4.18 \text{ J g}^{-1} \text{ K}^{-1}$), or less than one-half on a volume basis ($1.88 \text{ J cm}^{-3} \text{ K}^{-1}$ vs $4.18 \text{ J cm}^{-3} \text{ K}^{-1}$) [11]. The ILs may be used through two strategies that are using the water free alkanolamine with a suitable IL and alternatively a blended of aqueous alkanolamine with IL. Camper et al. [11] mixed 50 mol % of ILs with free water MEA and DEA so that their results showed that the rapid and reversible captures of 1 mol of CO₂/2 mol of MEA and 0.29 mol of CO₂/1 mol of DEA. They mentioned that these types of mixtures are considered as an alternative to the aqueous alkanolamine solutions but it combines the advantages of both ILs and alkanolamines [11].

Using the blend of IL and aqueous alkanolamine solutions allows one to reduce the price and viscosity of solvent compared to the water free IL-alkanolamine mixture that is more suitable for industrial application. In this respect, Chinn et al. in their US patents [12] proposed using the ionic liquids [bmim][BF₄], [bmim][acetate], the MDEA + [bmim][acetate] and the MEA + [bmim][acetate] aqueous solutions as a solvent for the removal of CO₂.

Zhang et al. [13] synthesized the four amino acid ionic liquids: [N1111][Gly], [N2222][Gly], [N1111][Lys] and [N2222][Lys] so that they were mixed with water or N-methyldiethanolamine (MDEA) aqueous solutions to prepare a new type of solvent. The solubility or absorption of CO₂ in these IL + MDEA aqueous solutions was investigated over a range of temperature (298 K–318 K) and low partial pressure of CO₂ (4–400 kPa). The results indicated that the ionic liquid could greatly enhance both the absorption and the absorption rate of CO₂ in MDEA aqueous solution. In the other work by Zhang et al. [14], the absorption

performance of CO₂ in [N1111][Gly] + MDEA + water solution was studied in detail at 25 °C. It was found that MDEA dramatically increases the absorption load of CO₂ in ILs aqueous solutions, while the presence of ILs in the solvent system had little effect on the absorption load of MDEA solutions. Ahmady et al. studied the initial absorption rate [15] and kinetics of the CO₂ absorption [16] in aqueous 4 molar MDEA mixed with various concentrations of [bmim][BF₄] at 303–333 K. They also measured the solubility of CO₂ in the blend of aqueous 4 molar MDEA with [bmim][BF₄], [bmim][acetate] and [bmim][DCA] at 303–333 K and up to 700 kPa [15,17]. Yusoff and coworkers at the two different works [18,19] represented the solubility of CO₂ in the aqueous blended systems of the MDEA + [gua][OTf] and MDEA + [gua][FAP] at 303.15 K, 323.15 K and 333.15 K at the CO₂ partial pressure up to 3000 kPa. They found that addition of these two ILs to the aqueous MDEA present a light decrease of CO₂ solubility.

One of the ILs that show a high solubility of CO₂ through the chemical absorption is the ionic liquids with acetate anions such as the 1-butyl-3-methylimidazolium acetate [bmim][Ac] [12,20–23]. Yokozeki et al. [21] measured solubility of CO₂ in 18 room-temperature ionic liquids (RTILs) at 298 K and pressure up to about 20 bar and demonstrated that the ionic liquids with acetate anions pronounces solubility of CO₂ in comparison to the other ionic liquids.

The published works about the CO₂ solubility in ionic liquids blended with aqueous alkanolamine solutions are very limited so that the most of them are relevant to tertiary alkanolamine such as MDEA. Thus, investigation of gas solubility in ionic liquids is needed for new ionic liquids blended with the different alkanolamines at various conditions. Therefore, in this work a set of new solvents is prepared through blending the [bmim][acetate] as a room temperature ionic liquid (RTIL) with the different aqueous alkanolamines such as MDEA, DEA, DIPa and AMP. The solubility of CO₂ in these new blended solvents are measured at different composition and 323.15 K.

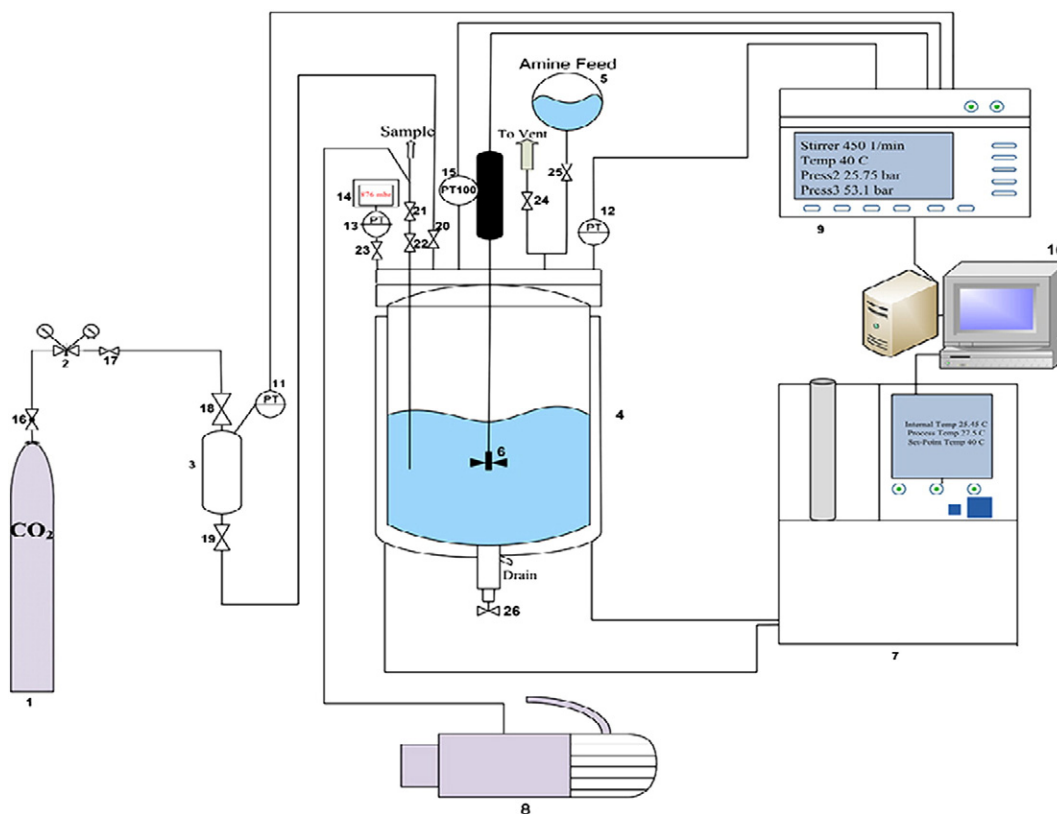


Fig. 1. The schematic diagram of VLE apparatus to measuring the gas solubility in a solvent. (1) CO₂ cylinder; (2) regulator; (3) gas-storage vessel; (4) equilibrium cell; (5) solvent storage vessel; (6) stirrer; (7) circulator; (8) vacuum pump; (9) monitoring unit; (10) PC; (11–13) pressure transducers; (14) pressure indicator; (15) temperature sensor (Pt-100); (16–25) valves; (26) drain valve.

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