

## Vapor pressure of aqueous methyldiethanolamine mixed with ionic liquids



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### ABSTRACT

Vapor pressure of aqueous methyldiethanolamine (MDEA) mixed with various concentrations of 1-butyl-3-methyl-imidazolium tetrafluoroborate ([bmim][BF<sub>4</sub>]) and 1-butyl-3-methyl-imidazoliumdicyanamide ([bmim][DCA]) have been measured and compared with conventional solvents.

A standard response surface methodology (RSM) design, namely central composite design (CCD) has been applied in this work to investigate the effects of temperature and concentration of Piperazine, [bmim][BF<sub>4</sub>] and [bmim][DCA] on the vapor pressure of aqueous 4 kmol m<sup>-3</sup> MDEA.

The vapor pressure data of aqueous 4 kmol m<sup>-3</sup> MDEA mixed with ionic liquids ranging from 0 to 2.0 kmol m<sup>-3</sup> and temperatures ranging from 30 to 80 °C, showed that the ionic liquids reduced the vapor pressure of solvents with the order of [bmim][DCA] > [bmim][BF<sub>4</sub>].

The experimental values are in good agreement with the values predicted by the models. The model results showed that the quadratic model was suitable and sufficient for predicting the vapor pressure of solutions in the investigated ranges of temperature and concentration.

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### 1. Introduction

In conventional methods, aqueous single amines, mixture of specific amines and amines mixed with a salt of an amino acid are used to remove carbon dioxide (CO<sub>2</sub>) and hydrogen sulfide (H<sub>2</sub>S) from natural and synthesis gases through chemical absorption systems [1,2]. Monoethanolamine, diethanolamine and methyldiethanolamine are the common types of alkanolamines that are being used for CO<sub>2</sub> capture through chemical absorption in many industries. However, using of amine technologies for CO<sub>2</sub> capture still have some drawbacks such as waste of solvents due to evaporation in stripping column, thermal degradation of costly reagents and corrosion in the involved equipment. Furthermore solvents with chemical absorption capability need relatively higher regeneration energy than solvents with physical absorption capability [3].

Recently, the unique properties of room temperature ionic liquids (RTILs) such as high thermal, electrochemical and chemical stability, low melting points, tunable gas solubility and selectivity, negligible vapor pressure, recyclability and wide liquid range of temperature have made them an attractive medium for CO<sub>2</sub> and H<sub>2</sub>S absorption [4–6]. Considering the negligible vapor pressure of ionic

liquids in gas sequestration applications, they can be used in such processes for longer times without changes in quantity through vaporization and entering the outlet gas stream from the stripping column. Therefore, ionic liquid can be used as a superior material in such applications. As a first approach, RTILs have been proposed as solvent additives for gas separations [7]. Many researchers have shown the relatively good affinity of imidazolium-based ILs to absorb CO<sub>2</sub> in comparison with other types of ionic liquids [8–12]. However, ionic liquids still have some inherent drawbacks for use in industries. Their high viscosity, low CO<sub>2</sub> loading and absorption rate, and very high cost as compared to conventional solvents, leads the researchers to use another strategy to improve their performance by using them mixed with amines [13–20]. By using these mixtures, many existing drawbacks of standard and task-specific ionic liquids (TSILs) would be avoided while retaining almost all desired properties of them [14].

This work was undertaken to design experiments, build models and evaluate the effect of two common ionic liquids, 1-butyl-3-methyl-imidazolium dicyanamide ([bmim][DCA]) and 1-butyl-3-methyl-imidazolium tetrafluoroborate ([bmim][BF<sub>4</sub>]) in concentrations ranging from 0 to 2 kmol m<sup>-3</sup>(M) on the vapor pressure of aqueous 4 M MDEA. In an industrial application, the BASF Corporation has presented and commercialized activated methyldiethanolamine (MDEA) solvents using MDEA (1.5–4.5 M) mixed with up to 0.8 M Piperazine [21]. Thus, the vapor pressure of IL-MDEA solutions has been compared with the measured PZ + MDEA vapor pressure to approach an industrial perspective.

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To the best of our knowledge, there is no reported vapor pressure data for the aqueous mixture of [bmim][DCA] and [bmim][BF<sub>4</sub>] with MDEA in the open literature. These data are very important for the new suggested ionic liquid-based solvents for CO<sub>2</sub> absorption as discussed in previous work by the authors [17,18].

## 2. Materials

The chemicals used in this study were methyldiethanolamine (MDEA, 98.5%), 1-butyl-3-methyl-imidazolium dicyanamide ([bmim][DCA], 98%) and 1-n-butyl-3-methylimidazolium tetrafluoroborate ([bmim][BF<sub>4</sub>], 98%) purchased from Merck. The Piperazine (PZ, 99.9%) was purchased from Sigma Aldrich. Distilled water was used to dilute the solvents throughout the experiments.

## 3. Apparatus and procedure

The experimental apparatus shown in Fig. 1 was designed, constructed and used for vapor pressure measurements. It was composed of an equilibrium cell (EC) with 40 mL solvent capacity equipped with a temperature transmitter with  $\pm 0.1$  °C accuracy (TI), a magnetic stirrer, a vacuum pump with two pressure transducers with  $\pm 0.01$  kPa accuracy (PT) and two isolating valves (NV and BV) to isolate the solvent and the system during process preparation.

Before solvent preparation, the liquid chemicals were heated up to 10% below boiling temperature at under vacuum and kept constant using temperature control system with  $\pm 0.1$  accuracy to remove existing volatile impurities. The weight of required chemicals has been measured with maximum  $\pm 0.1$  g error. To prevent evaporation of solvents and changing their composition, all samples were prepared without a delay before measurements. For every experiment, the equilibrium cell was filled with the solvent and trapped air bubbles were removed by applying small vibrations before closing the needle valve. To ensure that the same amount of solvent would be used in all experiments, more than 40 mL of solvent was charged and the excess was drained after closing the needle valve (NV). A vacuum pump was used to reduce the pressure down to 5 Pa in order to remove the existing air at the upper part of the equipment while the isolation valve, NV, was kept closed. Then, the system was isolated from the vacuum pump using BV. In next step, NV was opened and the system was kept at a constant temperature to reach equilibrium; that is, when the pressure and solvent temperature remained constant for at least 30 min. All the experiments were done at isothermal conditions at 30, 40, 50, 60, 70 and 80 °C.

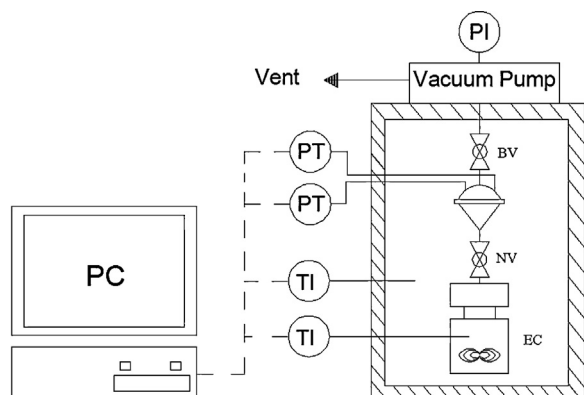


Fig. 1. Experimental setup for vapor pressure measurement. (PT, pressure transmitter; TI, temperature indicator; BV, ball valve; NV, needle valve; EC, equilibrium cell).

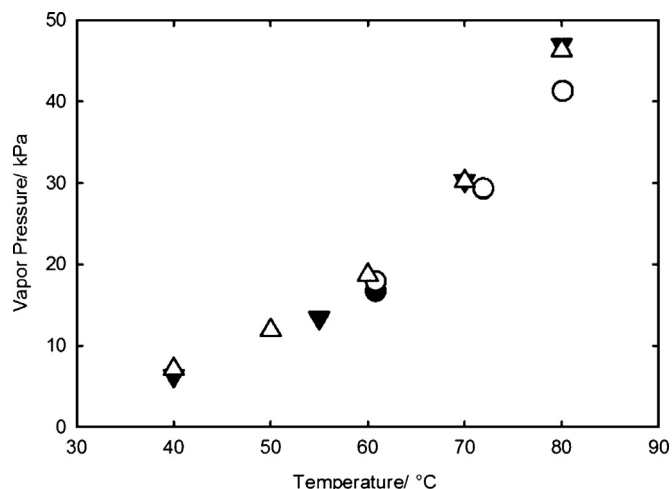


Fig. 2. Vapor pressure of 50 wt.% MDEA at various temperature. ▼, Xu et al. [24]; ○, Raoult's Law [24]; ●, Xu et al. [25]; △, this work.

## 4. Results and discussion

The vapor pressure data for aqueous MDEA are very limited in the literature. However, to confirm the procedure and reproducibility of obtained vapor pressure data, the vapor pressure of 50 wt.% MDEA was measured and compared to published data (Fig. 2).

As shown in Fig. 2, a good consistency was achieved between the data generated in this study and the data from the open literature.

Because the vapor pressure of MDEA, [bmim][BF<sub>4</sub>] and [bmim][DCA] are negligible in comparison with water vapor pressure at the same temperature, as a first approach, it has been assumed that only gaseous water is present in vapor phase. Thus, the vapor pressure data could be calculated from Raoult's Law (Eq. (1)), considering only water mole fraction. These data can be used as reliable criteria to compare with experimental data.

$$P^v = \sum P_i x_i, \quad i = 1 - n \quad (1)$$

where  $P^v$  is total vapor pressure of the solution,  $P_i$  is the partial pressure of the component  $i$  in the mixture (in the solution) and  $x_i$  is the mole fraction of the component  $i$  in the mixture (in the solution).

The calculated data and the data from experiment are listed in Tables 1 and 2. As illustrated in Tables 1 and 2, with higher IL concentration, positive deviation from Raoult's Law has changed to negative at higher temperature. This finding shows that the ionic liquids-MDEA solutions could be used as a promising CO<sub>2</sub> absorption medium with effective operability at the regenerations columns without a big concern about solvent lost.

### 4.1. Design of experiments

A statistical technique called response surface methodology (RSM) uses quantitative data from experiments to measure operating conditions and model equations [22].

The concept of a response surface involves a set of empirical methods assigned to the assessment of relations between the measured responses and a cluster of controlled experimental factors, based on one or more selected criteria [23].

To study the vapor pressure variables of 4 M MDEA + PZ, 4 M MDEA + [bmim][BF<sub>4</sub>] and 4 M MDEA + [bmim][DCA] solutions, a

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