

# Gaseous absorption of 2,3,3,3-tetrafluoroprop-1-ene in three imidazolium-based ionic liquids



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## ABSTRACT

The properties of working pairs of refrigerant and absorbent have a great influence on the performance of absorption refrigeration cycle. In this work, the measurements of absorption properties for 2,3,3,3-tetrafluoroprop-1-ene/1-ethyl-3-methylimidazolium tetrafluoroborate ([EMIM][BF<sub>4</sub>]), 2,3,3,3-tetrafluoroprop-1-ene/1-hexyl-3-methylimidazolium tetrafluoroborate ([HMIM][BF<sub>4</sub>]), and 2,3,3,3-tetrafluoroprop-1-ene/1-methyl-3-octylimidazoliumtetrafluoroborate ([OMIM][BF<sub>4</sub>]) between 283.15 and 343.15 K were performed by means of an isochoric saturation method. Experimental data were correlated using NRTL model and Krichevsky-Kasarnovsky (K-K) equation. The studied results show that absorption of gaseous 2,3,3,3-tetrafluoroprop-1-ene increases with a decrease in temperature and an increase in pressure, and absorption diminished in the following order: [OMIM][BF<sub>4</sub>] > [HMIM][BF<sub>4</sub>] > [EMIM][BF<sub>4</sub>].

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## 1. Introduction

Since increasing concerns over a global energy crisis, the absorption refrigeration technology which can be driven by low-quality waste heat has attracted considerable attention [1,2]. In the absorption refrigeration cycle, proper selection of working pairs composed of refrigerant and absorbent is a critical issue, because the performance of the cycle mainly depends on thermophysical properties of working pairs, especially the absorption properties of refrigerant in absorbent. Commonly used working pairs are NH<sub>3</sub>/H<sub>2</sub>O and H<sub>2</sub>O/LiBr, but they have certain disadvantages, such as toxicity, crystallization, corrosion, negative pressure operation, and difficulty for post-desorption separation [3,4].

Ionic liquids (ILs), which consist of organic cations and organic or inorganic anions, are a liquid salt at (or close to) ambient temperature. Owing to their tunable physicochemical properties, extremely low volatility, excellent thermal stability, and environmentally friendly characteristics, ILs were regarded as promising absorbents for absorption refrigeration cycles [2]. In the past few years, several research groups have investigated the absorption

properties of refrigerants in ILs [5–11]. In addition, Kohl et al. [12–16] analyzed the thermodynamic performance of absorption refrigeration systems with refrigerants/imidazolium-based ILs as working pairs. The studied imidazolium-based ILs include 1-ethyl-3-methylimidazolium tetrafluoroborate ([EMIM]BF<sub>4</sub>), 1-butyl-3-methylimidazolium tetrafluoroborate ([BMIM]BF<sub>4</sub>), 1-hexyl-3-methylimidazolium tetrafluoroborate ([HMIM]BF<sub>4</sub>), 1-butyl-3-methylimidazolium hexafluorophosphate ([BMIM]PF<sub>6</sub>), 1-hexyl-3-methylimidazolium hexafluorophosphate ([HMIM]PF<sub>6</sub>), 1-hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([HMIM]Tf<sub>2</sub>N), and 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM]Tf<sub>2</sub>N), the refrigerants are H<sub>2</sub>O, HFC134a, HFC125, HFC143a, HFC152a, HFC32, HFC114, and HFC134. Investigations present great potential and significant application prospects of these novel working pairs. The above studied HFC refrigerants are almost harmless to the atmospheric ozone, but they are considered greenhouse gases under the Kyoto Protocol because of high global warming potential (GWP) values. 2,3,3,3-Tetrafluoroprop-1-ene (HFO1234yf) has the atmospheric lifetime of only 11 days and a 100-year time horizon GWP of 4 relative to carbon dioxide [17], and its thermodynamic properties are quite similar to those of HFC134a. HFO1234yf is therefore considered a “fourth generation” refrigerant in refrigeration industry, and it is

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very important to investigate the HFO1234yf/ILs as potential working pairs in the absorption refrigeration cycle. However, to the author's knowledge, only Liu et al. [18] measured the solubility behavior of HFO1234yf in [HMIM][Tf<sub>2</sub>N].

Based on the types of organic cation, ILs can be divided into imidazolium, pyridinium, tetraalkyl ammonium, tetraalkylphosphonium, and so on. Among different ILs, imidazolium-based ILs have been considered as one of the potential ILs used in the absorption refrigeration system [2]. In this work, the absorption behavior of gaseous HFO1234yf in 1-ethyl-3-methylimidazolium tetrafluoroborate ([EMIM][BF<sub>4</sub>]), in 1-hexyl-3-methylimidazolium tetrafluoroborate ([HMIM][BF<sub>4</sub>]), and in 1-methyl-3-octylimidazolium tetrafluoroborate ([OMIM][BF<sub>4</sub>]) were measured at temperatures ranging from 283.15 to 343.15 K and pressures up to 0.45 MPa by means of isochoric saturation method. Solubility data were correlated by NRTL model and Krichevsky-Kasarnovsky (K-K) equation, and Henry's law constants of HFO1234yf in these three ILs at all isotherms were calculated. Moreover, the effects of cations of imidazolium-based ILs, temperatures, and pressures on the absorption were analyzed.

## 2. Experimental methodology

### 2.1. Materials

HFO1234yf (CAS No. 754-12-1) was provided by Honeywell with a declared mass purity of 99.9%, which was purified 3–5 times by a high-vacuum system and liquid nitrogen (freeze–pump–thaw) before measurements. [EMIM][BF<sub>4</sub>] (CAS No. 143314-16-3), [HMIM][BF<sub>4</sub>] (CAS No. 244193-50-8), and [OMIM][BF<sub>4</sub>] (CAS No. 244193-52-0) were purchased from Sigma-Aldrich with a stated mass purity higher than 97.0%. Fig. 1 shows the structures of the three ionic

liquids. The ionic liquid samples were dried and degassed under vacuum, while simultaneously heated and stirred at a temperature of about 353.15 K for at least 72 h. The final mass fraction of water in the dried samples was measured by Karl Fischer titration (MKC-710), and results show that water content in ILs is below 100 ppm. Table 1 lists the sample descriptions used in the present work.

### 2.2. Experimental technique

A self-designed apparatus based on isochoric saturation method, shown in Fig. 2, was employed to measure absorption properties of HFO1234yf in different ILs. The experimental system has been described in our previous works [19–22], but for the present measurements, some changes were illustrated. The apparatus consists of two different volume stainless steel cells, named equilibrium cell with a calibrated volume of 31.33 cm<sup>3</sup> and gas cell with a calibrated volume of 73.26 cm<sup>3</sup>, respectively. A molecular sieve trap was placed before the gas cell to remove any trace amounts of water from gaseous refrigeration. The absorption behavior of gaseous refrigeration in ionic liquid was performed in equilibrium cell with glass windows. To get a faster equilibrium, the solution inside the cell was stirred by means of a magnetic stirrer. The two stainless steel cells were immersed in a self-made thermostatic bath filled with water, in which the temperature was controlled by LAUDA ECO Gold. The measurement of temperature was performed by a calibrated Pt100 resistance thermometer (Fluke 5608) with an accuracy of 0.02 K. The estimate of combined standard uncertainty of temperature was 0.03 K with level of confidence 0.95 ( $k = 2$ ). Pressure was measured by means of a pressure transducer (KELLER 33X) with a full scale of 3000 kPa, and the combined expanded uncertainty ( $k = 2$ ) in pressure measurement was 2.0 kPa.

For the solubility measurement, a well-known amount of dried ionic liquid was directly charged into equilibrium cell, the mass of ILs was determined by an analytical balance (Mettler Toledo ME204, 220 g full scale) with an uncertainty of 0.002 g, and then the experimental system was kept under vacuum for at least 72 h. After that, gaseous HFO1234yf was added into the gas cell from the gas bottle, the mass of charged gaseous HFO1234yf can be obtained from the initial temperature and pressure of gas cell. Then the interconnecting valve (V4) between gas cell and equilibrium cell was opened, the absorption of gaseous HFO1234yf in the stirred solution was started. Once the pressure of equilibrium cell remained constant, the magnetic stirrer was turned off. After about 1 h of waiting to ensure the achievement of the phase equilibrium, the pressure was recorded. The next step was to change the bath temperature to measure the next equilibrium pressure.

The mole fraction  $x_1$  of dissolved gaseous HFO1234yf in ionic liquid is obtained from  $x_1 = n_1/(n_1 + n_2)$ , where  $n_2$  is the number of moles of ionic liquid. Since the vapor pressures of ionic liquids are near-zero, the gaseous component in the experimental system is assumed as only HFO1234yf. Therefore,  $n_1$ , the number of moles of HFO1234yf dissolved in ionic liquid, can be calculated by Refs. [19–22].

$$n_1 = \left[ \frac{V_{\text{sys}}}{v_{\text{gas}}(T_{\text{ini}}, p_{\text{ini}})} - \frac{V_{\text{sys}}}{v_{\text{gas}}(T_{\text{equilib}}, p_{\text{equilib}})} + \frac{V_{2,\text{cell}} - V_{\text{cell}}}{v_{\text{gas}}(T_{\text{equilib}}, p_{\text{equilib}})} \right] \bigg/ \left[ 1 - \frac{v_{\text{abs,gas}}}{v_{\text{gas}}(T_{\text{equilib}}, p_{\text{equilib}})} \right] \quad (1)$$

where the subscripts of 'ini' and 'equilib' represent initial and

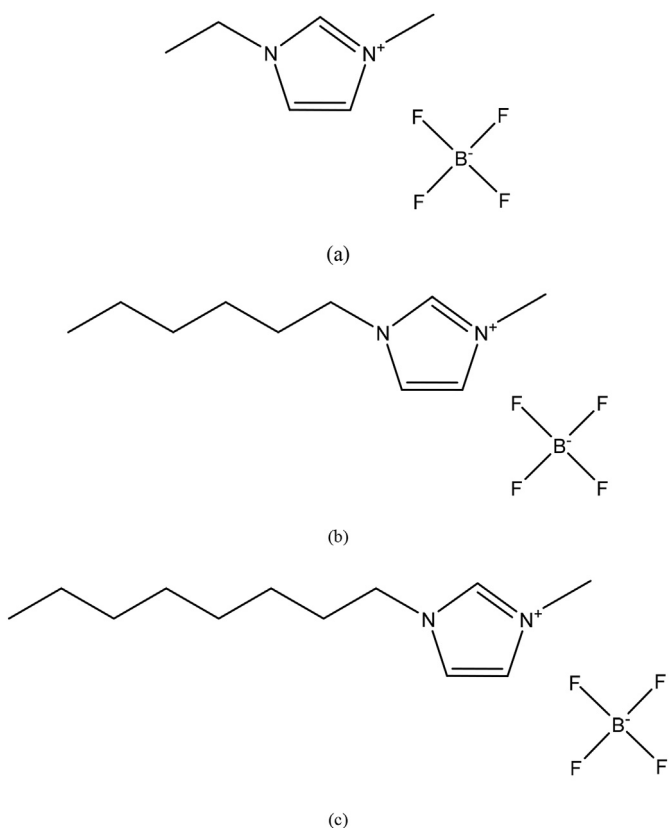


Fig. 1. Structure of ILs: (a) [EMIM][BF<sub>4</sub>], (b) [HMIM][BF<sub>4</sub>], (c) [OMIM][BF<sub>4</sub>].

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