

Gaseous absorption of fluorinated ethanes by ionic liquids



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ABSTRACT

Solubilities of 1,1-difluoroethane (R152a), pentafluoroethane (R125), fluoroethane (R161), 1,1,1-trifluoroethane (R143a) and 1,1,1,2-tetrafluoroethane (R134a) in trihexyltetradecylphosphonium bis(2,4,4-trimethylpentyl) phosphinate ([P(14) 666][TMPP]) from 302.2 K to 343.6 K and up to 1.14 MPa are presented in this work. Effects of fluorine atoms of fluorinated ethanes, the cations and anions of ionic liquids on the solubility behaviors are investigated by comparing the solubilities of fluorinated ethanes and ethane in different ionic liquids. Moreover, the partial molar enthalpies and partial molar entropies of solutions at infinite dilution limit for R161, R143a, R125, R134a and R152a in [P(14) 666][TMPP] are obtained. The experimental results are successfully correlated with a modified Krichevsky–Kasarnovsky equation.

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1. Introduction

Ionic liquids are a class of liquid salts near ambient temperature. Most of them have negligible vapor pressure, high thermal and chemical stability, and high solubilities for gases [1]. Recently, lots of work have been carried out on the solubility behaviors of hydrofluorocarbons (HFCs) in ionic liquids which are important for applying ionic liquids + HFCs as working pairs for the absorption refrigeration cycles [2–4] or using ionic liquids as solvents for separation of hydrofluorocarbon mixtures [5]. Most of these work are on the solubilities of HFCs in imidazolium-based ionic liquids [6–22]. There are little experimental data for the solubilities of HFCs in phosphonium-based ionic liquids although phosphonium-based ionic liquids have some advantages such as high thermal stability and low cost. Shiflett et al. [9] reported the solubilities of R134a in trihexyltetradecylphosphonium 1,1,2-trifluoro-2-(perfluoroethoxy) ethanesulfonate ([P14666][TPES]) and tributyltetradecyl phosphonium 1,1,2,3,3,3-hexafluoropropanesulfonate ([P14444][HFPS]). Sousa et al. [23,24] studied the solubilities of trifluoromethane, difluoromethane and fluoromethane in trihexyltetradecylphosphonium bis(trifluoromethylsulfonyl) amid ([P14666][Tf₂N]), trihexyltetradecylphosphonium chloride ([P14666][Cl]), tributylmethylphosphonium methylsulfate ([P4441][C₁SO₄]) and tributylethylphosphonium diethylphosphate ([P4442][(C₂)₂PO₄]).

In this work, we measured the solubilities of R161, R143a, R125, R134a and R152a in [P14666][TMPP] which has high solubilities for small hydrocarbons and carbon dioxide [25,26]. High solubility can

help to improve the efficiency of the absorption refrigeration cycles and of the separation process of hydrofluorocarbon mixtures. We hope that [P14666][TMPP] also has high solubilities for these five fluorinated ethanes. The solubility measurements were carried out from 302.2 K to 343.6 K and up to 1.14 MPa with an isochoric saturation method. The interactions between the five fluorinated ethanes and ionic liquids were investigated. A modified Krichevsky–Kasarnovsky equation was used to correlate the experimental results.

2. Experimental

2.1. Materials

R161 was purchased from Zhejiang Province Chemical Industry Research Institute with a purity ≥ 99.95 wt%. R143a, R125, R152a, R134a were purchased from Zhejiang Sinoloong Refrigerant Co., Ltd. with purities ≥ 99.9 wt%. [P(14) 666][TMPP] was purchased from Strem Chemicals, Incorporated with a purity ≥ 95.0 wt%. Information provided by supplier shows that [P(14) 666][TMPP] contains 2140 ppm chloride and 6510 ppm bromide ions. The ionic liquids were purified by drying it at 393 K for 48 h under vacuum condition. The chemicals used in this study are summarized in Table 1.

2.2. Solubility apparatus

Solubilities of R143a, R125, R152a, R134a and R161 in [P(14) 666][TMPP] were measured using an isochoric saturation method as shown in Fig. 1. The principle is loading some known quantity of solvent and gas into the equilibrium cell. The pressure in the

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Table 1
The chemicals used in this work.

Chemical name	Supplier	Purity in mass fraction	Purification method	Water content in mass fraction
[P14666][TMPP] ^a 1,1,1, 2-Tetrafluoroethane (R134a)	Strem Chemicals, incorporated Zhejiang Sinoloong Refrigerant Co., Ltd.	≥95.0% ≥99.9%	Vacuum drying None	0.01% ^b
1,1,1-Trifluoroethane (R-143a)	Zhejiang Sinoloong Refrigerant Co., Ltd.	≥99.9%	None	–
Pentafluoroethane (R-125)	Zhejiang Sinoloong Refrigerant Co., Ltd.	≥99.9%	None	–
1,1-Difluoroethane (R-152a)	Zhejiang Sinoloong Refrigerant Co., Ltd.	≥99.9%	None	–
Fluoroethane (R-161)	Zhejiang Province Chemical Industry Research Institute	≥99.95%	None	–

^a Trihexyltetradecylphosphonium bis(2,4,4-trimethylpentyl) phosphinate.

^b Measured with Metrohm 831 Karl Fischer Titrator.

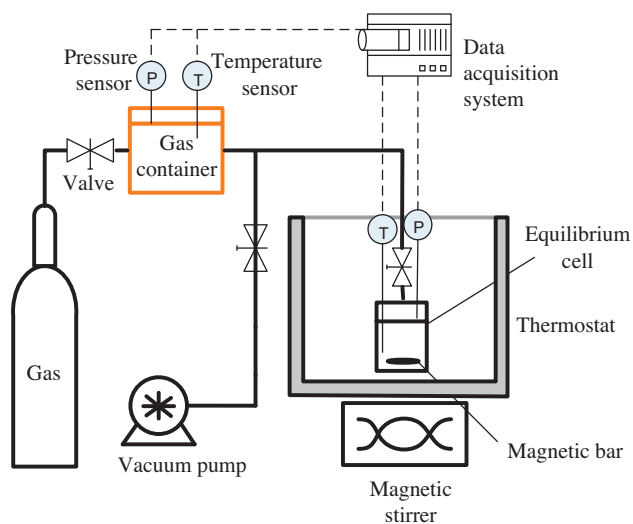


Fig. 1. Schematic of the experimental apparatus.

equilibrium cell will decrease at fixed temperature due to the dissolution of gas in the solvent. The change in the mass of gas yields the solubility of gas in the solvent. A detailed description of apparatus and experimental procedure were given in our previous papers [21,22], we just give a brief introduction in this work. The apparatus mainly consists of the equilibrium cell, the gas reservoir, the magnetic stirrer and the temperature and pressure sensors. The gas reservoir was made of copper to keep the temperature of gas in it uniform and stable. The volume of the equilibrium cell and the gas container were calibrated by gravimetric method using water as working liquid (Mettler Toledo ME4002 balance, uncertainty is 0.02 g). Temperature inside the equilibrium cell and the gas reservoir were measured with platinum resistant thermometers (Fluke 5608) with an expanded uncertainty 0.02 K (coverage factor $k=2$, 0.95 level of confidence). The temperature of the equilibrium cell is controlled with the thermostat. The temperature stability is less than ± 0.04 K/30 min. The expanded uncertainty of the temperature in the equilibrium cell is less than 0.1 K ($k=2$). Pressure sensors (Keller 33X) with an expanded uncertainty 0.3 kPa ($k=2$) were used to measure the pressure in the equilibrium cell and the gas reservoir. The magnetic stirrer is used to accelerate the dissolution of gas in ionic liquid.

2.3. Calculation of gas solubility

The mole number of gas dissolved in the ionic liquid is calculated from

$$n_g^1 = V_{GC}(\rho_i - \rho_f) - \rho_g(V_{EC} - V_1) \quad (1)$$

where V_{GC} , V_{EC} and V_1 are the volume of the gas container, the equilibrium cell and ionic liquid in the equilibrium cell, respectively; ρ_i and ρ_f are the molar densities of solute in the gas container before and after loading the solute into the equilibrium cell; ρ_g is the molar density of solute in the vapor phase in the equilibrium cell when equilibrium is reached. ρ_i , ρ_f and ρ_g were obtained from the REFPROP 9.1 [27]. V_1 was obtained from the mass and density of ionic liquid. The mass of ionic liquid in the equilibrium cell was measured with a Mettler Toledo ME 204 balance with an uncertainty 0.2 mg. Densities of [P(14) 666] [TMPP] have been reported previously, they can be expressed as a linear function of temperature [25]:

$$\rho/g \text{ cm}^{-3} = -5.9125 \times 10^{-4}(T/K) + 1.0655 \quad (2)$$

Solubility of the gas in the ionic liquid can be expressed as mole fraction

$$x = \frac{n_g^1}{n_1 + n_g^1} \quad (3)$$

where n_1 is the mole number of ionic liquid.

The relative expanded uncertainty in mole fraction can be obtained from

$$U_r(x) = \frac{ku(x)}{x} = k \left[\left(\frac{\partial x}{\partial V_{GC}} \right)^2 u(V_{GC})^2 + \left(\frac{\partial x}{\partial \rho_i} \right)^2 u(\rho_i)^2 + \left(\frac{\partial x}{\partial \rho_f} \right)^2 u(\rho_f)^2 + \left(\frac{\partial x}{\partial \rho_g} \right)^2 u(\rho_g)^2 + \left(\frac{\partial x}{\partial V_{EC}} \right)^2 u(V_{EC})^2 + \left(\frac{\partial x}{\partial V_1} \right)^2 u(V_1)^2 + \left(\frac{\partial x}{\partial n_1} \right)^2 u(n_1)^2 \right]^{1/2} \quad (4)$$

where $u(x)$ is the standard uncertainty in mole fraction; $u(V_{GC})$, $u(V_{EC})$, $u(V_1)$, $u(\rho_i)$, $u(\rho_f)$, $u(\rho_g)$ and $u(n_1)$ are the standard uncertainty in V_{GC} , V_{EC} , V_1 , ρ_i , ρ_f , ρ_g and n_1 , respectively. The relative expanded uncertainty in mole fraction $U_r(x) = U(x)/x$ including random error is estimated to be less than 4% ($k=2$).

3. Results and discussion

Solubilities of R143a, R125, R152a, R134a and R161 in [P(14) 666][TMPP] were measured at temperatures from 302.2 K to 343.6 K and at pressures from 0.055 MPa to 1.14 MPa as shown in Tables 2 and 3. The solubility measurements were repeated three times using the same sample, no significant difference were found. The solubilities increase with the increasing of pressure and decrease with the increasing of temperature. In order to investigate

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