Vaporization Thermodynamics of Pyrrolidinium, Pyridinium, and Imidazolium-Based Ionic Liquids Bearing the Bis(fluorosulfonyl)imide [FSI] and the Bis(trifluoromethylsulfonyl)imide [NTf₂] Anions

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Abstract

New experimental vapor pressures in the range 407 K to 460 K and vaporization enthalpy of the ionic liquids (IL) *N*-alkyl-*N*-methyl-pyrrolidinium bis(fluorosulfonyl)imide ionic liquids have been measured using quartz crystal microbalance. The absolute vapor pressures and vaporization enthalpies were compared with analogous pyrrolidinium-based ILs with the bis(trifluoromethanesulfonyl)imide anion. The evaluated difference in vaporization enthalpy of ILs with bis(fluorosulfonyl)imide and bis(trifluoromethanesulfonyl)imide anions allowed for estimation of corresponding property for a wide set of ILs with bis(fluorosulfonyl)imide anion. The results are relevant to chemical engineering calculations of processes involving ILs as reaction and separation media.

Keywords Enthalpy of vaporization · Ionic liquid · Vapor pressure

1 Introduction

The irreversible trend of modern technologies towards renewable energies requires safe storage and delivery of the generated energy. Lithium-ion batteries are among the most effective technical solutions today. They are widely used in portable

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electronic devices, full-electric vehicles, etc. Ionic liquids are of great interest as a new class of electrolyte materials for lithium-ion batteries due to their thermal stability and negligible (even at elevated temperatures) vapor pressures [1, 2]. Ionic liquids based on the bis(fluoromethanesulfonyl) imide [FSI]-anion and bis(trifluoromethanesulfonyl)imide [NTf₂]-anion have so far been among the most prominent material candidates because they generally have a high conductivity and relatively low viscosity [3].

Each specific application requires specific physicochemical properties of ILs, and they can be tailored, provided the general principles governing ILs families are known. It is therefore important to study structure–property relationships to reduce the experimental effort required to produce materials with the desired properties. This paper extends our previous studies on the structure–property relations in ILs families and deals with vapor pressure measurements on *N*-alkyl-*N*-methyl-pyrrolidinium bis(fluorosulfonyl)imide ionic liquids (see Fig. 1).

2 Experimental Section

2.1 Materials

The samples of 1-propyl-1-methylpyrrolidinium bis(fluorosulfonyl)imide ([C₃C₁Pyrr][FSI]) 1-butyl-1-methylpyrrolidinium bis(fluorosulfonyl)imide ([C₄C₁Pyrr][FSI]) used in this work were of commercial origin (see Table S1). Prior to the experiments, samples were subjected to vacuum evaporation at 413 K and 10^{−5} Pa for more than 24 h to reduce possible traces of solvents and moisture. Samples were additionally conditioned inside of the vacuum chamber at the highest temperature of the vaporization experiment within 12 h. This additional purification allowed for removing of residual traces of water and volatile impurities as well as for collecting the amount of the vaporized IL required for the FTIR analysis.

2.2 Measurements of Vaporization Enthalpies by the Quartz Crystal Microbalance (QCM)

Absolute vapor pressures and the standard molar enthalpies of vaporization of the [C_{*n*}C₁Pyrr][FSI] were determined using the QCM method [4]. In the QCM method

Fig. 1 Structures of *N*-alkyl-*N*-methyl-pyrrolidinium bis(fluorosulfonyl)imide ionic liquids studied in this work

a sample of an IL is placed in an open chamber (Langmuir evaporation) inside of the isothermal stainless steel cylinder and exposed to high vacuum (10^{-5} Pa) with the whole open surface of the loaded compound. Directly above the measuring cavity containing the sample the QCM sensor is placed. Liquid sample fully covers the bottom of cavity and due to low mass loss during the study the surface area of liquid sample is left constant. Throughout the vaporization into vacuum, a certain amount of sample is deposited on the quartz crystal surface. The change of the vibrational frequency Δf is directly related to the mass deposition Δm on the crystal according to the Sauerbrey's equation [5] for the case $\Delta f \ll f$:

$$\Delta f = -C \cdot f^2 \cdot \Delta m \cdot S_C^{-1}, \quad (1)$$

here f is the fundamental frequency of the crystal (6 MHz), S_C is the surface of the crystal, and C is a constant [6]. The resolution of acquisition device is at level of 0.01 Hz, at the same time preliminary studies had shown the long-term stability of recorded frequency at level of 0.5 Hz. Therefore, during each isotherm the time of vacuum exposure was controlled to achieve the frequency change at level of 100 Hz. The measured frequency change rates (df/dt) can be used for calculation of absolute vapor pressures p_{sat} according to equation:

$$p_{\text{sat}} = K' \frac{df}{dt} \sqrt{\frac{T}{M}}, \quad (2)$$

where the $K' = (9.5 \pm 1.1) \times 10^{-6} \text{ Pa} \cdot \text{s} \cdot \text{kg}^{1/2} \cdot \text{Hz}^{-1} \cdot \text{K}^{-1/2} \cdot \text{mol}^{-1/2}$ is the empirical constant containing all parameters of the Sauerbrey equation as well as parameters specific for the geometry of the experimental setup [6]. The K' -value for our apparatus was evaluated with the help of reliable vapor pressure data on imidazolium and pyridinium-based ILs compiled in reference [6]. Using the frequency change rate df/dt measured by the QCM and the K' -values the standard molar enthalpy of vaporization, $\Delta_1^{\text{g}} H_{\text{m}}^{\circ}(T)$ at experimental temperatures was obtained by:

$$R \ln(p_{\text{sat}}/p^{\circ}) = -\frac{\Delta_1^{\text{g}} G_{\text{m}}^{\circ}(T_0)}{T_0} + \Delta_1^{\text{g}} H_{\text{m}}^{\circ}(T_0) \left(\frac{1}{T_0} - \frac{1}{T} \right) + \Delta_1^{\text{g}} C_{\text{p,m}}^{\circ} \left(\frac{T_0}{T} - 1 + \ln \left(\frac{T}{T_0} \right) \right), \quad (3)$$

where $\Delta_1^{\text{g}} G_{\text{m}}^{\circ}(T_0)$ is the Gibb's energy change during vaporization of the studied ILs; T_0 appearing in Eq. 3 is an arbitrarily chosen reference temperature. The value $\Delta_1^{\text{g}} C_{\text{p,m}}^{\circ} = C_{\text{p,m}}^{\circ}(\text{g}) - C_{\text{p,m}}^{\circ}(\text{liq})$ is the difference between the molar heat capacities of the gaseous, $C_{\text{p,m}}^{\circ}(\text{g})$, and the liquid phase, $C_{\text{p,m}}^{\circ}(\text{liq})$, respectively. The vaporization enthalpy $\Delta_1^{\text{g}} H_{\text{m}}^{\circ}(298 \text{ K})$ at the reference temperature is calculated according to the Kirchhoff's equation:

$$\Delta_1^{\text{g}} H_{\text{m}}^{\circ}(298.15 \text{ K}) = \Delta_1^{\text{g}} H_{\text{m}}^{\circ}(T_{\text{av}}) + \Delta_1^{\text{g}} C_{\text{p,m}}^{\circ}(298.15 - T_{\text{av}}), \quad (4)$$

where T_{av} is the average temperature of the temperature range of the QCM study. In order to detect and avoid any possible effect of impurities on the measured frequency loss rate (df/dt), a typical experiment was performed in a few consequent series with increasing and decreasing temperature steps. Every series consisted of

9 to 10 temperature points of mass loss rate determination. Several runs have been performed to test the reproducibility of the results. The study was finished when the enthalpy of vaporization, $\Delta_1^g H_m^o$ (298 K), obtained in the sequential runs by adjusting Eqs. 3 and 4 to the temperature dependent rates (df/dt) agreed within the assessed experimental uncertainty of about $\pm 1 \text{ kJ}\cdot\text{mol}^{-1}$. In order to confirm the absence of decomposition of IL under the experimental conditions, the residual IL in the crucible and the IL-deposit on QCM were analyzed by ATR-IR spectroscopy. Primary experimental results of the QCM study is given in Table S1 in the Supporting Information.

3 Results and Discussion

3.1 Absolute Vapor Pressures

Absolute vapor pressures and vaporization enthalpies of the $[C_nC_1\text{Pyrr}][\text{FSI}]$ have been measured for the first time. Ionic liquids are widely applied in the modern catalytic processes like Solid Catalyst with Ionic Liquid Layer (SCILL) [7, 8] or Supported Ionic Liquid Phase (SILP) [8, 9]. For the catalyst to work stably, it is necessary to know the vapor pressure of an IL at any temperature. The temperatures 373 K and 473 K seem to be a reasonable choice to assess the vapor pressure as a measure of a possible long-term uptake of the IL during catalytic applications. As can be seen from Table 1, the extremely low vapor pressure values of $[C_nC_1\text{Pyrr}][\text{FSI}]$ at these temperatures indicate that even at elevated temperatures a negligible mass uptake of the IL can be expected in various catalytic or separation applications.

The absolute vapor pressure data on ILs are still seldom available in the literature and it is interesting to compare the general vapor pressure levels within the families of pyrrolidinium and imidazolium-based ILs with fluorinated anions [FSI] and [NTf₂] (see Fig. 2).

It is apparent from Fig. 2 that the vapor pressures of the $[C_nC_1\text{Pyrr}][\text{FSI}]$ is the lowest of ILs taken into comparison. Vapor pressures of ILs with [NTf₂] ions are of 10 to 100 times higher than those for the [FSI]-ILs. So, just removing two CF₂ groups from anion significantly decreases the absolute vapor pressures for studied ILs. That can be due to the decrease in charge delocalization in the [FSI] anion in comparison to [NTf₂].

Table 1 Absolute vapor pressures of $[C_nC_1\text{Pyrr}][\text{FSI}]$ at 373 K and at 473 K

IL	$10^6 p_{\text{sat}}, \text{Pa}$ 373 K	$10^3 p_{\text{sat}}, \text{Pa}$ 473 K
$[C_3C_1\text{Pyrr}][\text{FSI}]$	0.034	0.317
$[C_4C_1\text{Pyrr}][\text{FSI}]$	0.030	0.453

Fig. 2 Chain-length dependence of absolute vapor pressures at $T=423.15$ K for homologous series [C_nC₁Pyrr][NTf₂] (circles, ○), [C₂mim][NTf₂] (diamond, ◆) from [10], and [C_nC₁Pyrr][FSI] (stars, ☆) this work

3.2 Standard Molar Vaporization Enthalpy from Vapor Pressure Measurements

The standard molar enthalpies of vaporization $\Delta_1^g H_m^o(T_{\text{av}})$ derived from the temperature dependence of the vapor pressures measured with the QCM (see Table 2, column 4) are referenced to the average temperature T_{av} (see Table 2, column 3), which is the middle of the temperature range under study.

It is commonly recognized that reliable experimental data on enthalpies of vaporization for ILs are very scarce. For this reason, the development of a simple or sophisticated method for predicting or correlating enthalpies of vaporization is highly desirable. An example of the successful structure–property correlation within a homologous series is the chain length dependence of the $\Delta_1^g H_m^o(298 \text{ K})$ values (see Tables S3–S5). New experimental result on $\Delta_1^g H_m^o(298 \text{ K})$ of [C_nC₁Pyrr][FSI] allows for structure–property analysis in the series of pyrrolidinium-based ILs with the [FSI] and [NTf₂] containing anion. In Table 3 we compared vaporization enthalpies of [C_nC₁Pyrr][FSI] and [C_nC₁Pyrr][NTf₂] with the chain-length $n=3$ and 4. It is evident that the enthalpies of vaporization of ILs with the comparable chain length with the [FSI]-anion are systematically about 3 kJ·mol^{−1} higher compared to the analogous ILs with the [NTf₂]-anion.

This observation could be used as a simple “rule of thumb” to quickly assess $\Delta_1^g H_m^o(298 \text{ K})$ -values for ILs with the same [FSI] anion, but, e.g. with the pyridinium and imidazolium cations. For example, a reliable $\Delta_1^g H_m^o(298 \text{ K})$ -set of pyridinium-based ionic liquids [C_nPy][NTf₂] (see Table 4) was measured and evaluated in our recent work [13]. Using this data set and the average contribution

Table 2 Thermodynamics of vaporization of the pyrrolidinium-based ILs $[C_nC_1\text{Pyrr}][\text{FSI}]$

IL	T-range K	T_{av}	$\Delta_1^g H_m^o(T_{\text{av}})$ $\text{kJ}\cdot\text{mol}^{-1}$	$\Delta_1^g C_m^o(T_{\text{av}})^a$	$\Delta_1^g C_{p,m}^o{}^b$ $\text{J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1}$	$\Delta_1^g H_m^o(298\text{ K})^c$ $\text{kJ}\cdot\text{mol}^{-1}$
1	2	3	4	5	6	7
$[C_3C_1\text{Pyrr}][\text{FSI}]$	407.6–460.0	433.3	136.7 ± 1.1	81.7 ± 1.5	– 43	142.5 ± 1.6
$[C_4C_1\text{Pyrr}][\text{FSI}]$	407.5–450.0	429.7	140.6 ± 1.0	81.4 ± 1.5	– 51	147.3 ± 1.7

^aThe standard Gibbs energies of vaporization were evaluated using the calibration coefficient developed in our recent work [6]

^bHeat capacity differences between liquid and gas phases were calculated according to empirical equation $\Delta_1^g C_{p,m}^o = C_{p,m}^o(\text{liq}) \times (-0.26 \pm 0.05) + (68.7 \pm 37.0)$ [11]. The need heat capacities of ILs in liquid state were evaluated according to an empirical procedure suggested by Ahmadi et al. [12] to be $C_{p,m}^o(\text{liq}) = 430.2\text{ J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1}$ for $[C_3C_1\text{Pyrr}][\text{FSI}]$ and $462.0\text{ J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1}$ for $[C_4C_1\text{Pyrr}][\text{FSI}]$.

^cAdjusted to 298.15 K using $\Delta_1^g C_{p,m}^o$ -values from column 6. The final uncertainties of vaporization enthalpies adjusted to the reference temperature $T = 298\text{ K}$ are expressed as expanded uncertainties $U(\Delta_1^g H_m^o)$ at the 0.95 level of confidence with $k = 2$. These uncertainties combine the experimental uncertainty and the uncertainty of the temperature adjustment. Uncertainties in the temperature adjustment of vaporization enthalpies from T_{av} to the reference temperature are estimated to account with 20 % to the total adjustment

Table 3 Comparison of enthalpies of vaporization $\Delta_1^g H_m^o(298\text{ K})$ for $[C_nC_1\text{Pyrr}][\text{FSI}]$ measured in this work and $[C_nC_1\text{Pyrr}][\text{NTf}_2]$ from [10] (in kJ mol^{-1})

IL	$n = 3$	$n = 4$
$[C_nC_1\text{Pyrr}][\text{FSI}]$	142.5^a	147.3^a
$[C_nC_1\text{Pyrr}][\text{NTf}_2]$	140.3^b	143.9^b
Δ^c	2.2	3.4
Average Δ	2.8 ± 0.6	

^aMeasured in this work (see Table 2)

^bEnthalpies of vaporization derived from the chain-length dependence of experimental data given in Table S3

^cDifference between vaporization enthalpies of $[C_nC_1\text{Pyrr}][\text{FSI}]$ and $[C_nC_1\text{Pyrr}][\text{NTf}_2]$

$\Delta = 2.8 \pm 0.6\text{ kJ mol}^{-1}$ developed in Table 3, the $\Delta_1^g H_m^o(298\text{ K})$ -values for the $[C_n\text{Py}][\text{FSI}]$ series were calculated (see Table 4).

Another suitable example is calculation of the vaporization enthalpies of $[C_n\text{mim}][\text{FSI}]$ (see Table 4) with the $[C_n\text{mim}][\text{NTf}_2]$ data [11] on the $\Delta_1^g H_m^o(298\text{ K})$ -values (see Table S5). These new empirical values could be used to validate theoretical results from the literature. In fact, a careful literature search has shown that experimental studies of the enthalpies of vaporization of ILs with the $[\text{FSI}]$ anion are absent in the literature. The few theoretical enthalpies of vaporization that were found for comparison are listed in Table 5.

Results of molecular dynamics (MD) simulations using a polarizable many-body force field have been reported [14] on pyrrolidinium and imidazolium ILs with $[\text{FSI}]$ and $[\text{NTf}_2]$. As can be seen from Table 5, the vaporization enthalpies of $[C_3C_1\text{Pyrr}][\text{FSI}]$ and $[C_3C_1\text{Pyrr}][\text{NTf}_2]$ agree with the QCM results within the experimental

Table 4 Calculation of vaporization enthalpies $\Delta_1^g H_m^o$ (298 K) of pyridinium and imidazolium ILs connected to the [FSI] anion with help of reliable data on [C_nPy][NTf₂] and [C_nmim][NTf₂] and contribution Δ from Table 3 (in kJ mol⁻¹)

IL	$\Delta_1^g H_m^o$ (298 K)	IL	$\Delta_1^g H_m^o$ (298 K) ^c
Pyridinium ILs			
[C ₂ Py][NTf ₂]	131.1 ^a	[C ₂ Py][FSI]	133.9
[C ₃ Py][NTf ₂]	134.8 ^a	[C ₃ Py][FSI]	137.6
[C ₄ Py][NTf ₂]	138.5 ^a	[C ₄ Py][FSI]	141.3
[C ₅ Py][NTf ₂]	142.2 ^a	[C ₅ Py][FSI]	145.0
[C ₆ Py][NTf ₂]	145.9 ^a	[C ₆ Py][FSI]	148.7
Imidazolium ILs			
[C ₂ mim][NTf ₂]	122.9 ^b	[C ₂ mim][FSI]	125.7
[C ₃ mim][NTf ₂]	126.9 ^b	[C ₃ mim][FSI]	129.7
[C ₄ mim][NTf ₂]	130.9 ^b	[C ₄ mim][FSI]	133.7
[C ₅ mim][NTf ₂]	134.9 ^b	[C ₅ mim][FSI]	137.7
[C ₆ mim][NTf ₂]	138.9 ^b	[C ₆ mim][FSI]	141.7

^aEnthalpies of vaporization derived from the chain-length dependence of experimental data given in Table S4

^bEnthalpies of vaporization derived from the chain-length dependence of experimental data given in Table S5

^cCalculated as the sum of the enthalpy of vaporization given in column 2 and the contribution Δ from Table 3

Table 5 Comparison of experimental and theoretical vaporization enthalpies $\Delta_1^g H_m^o$ (298 K) of pyrrolidinium and imidazolium ILs combined with [FSI] and [NTf₂] anions (in kJ·mol⁻¹)

Method	[C ₃ C ₁ Pyrr][FSI]	[C ₃ C ₁ Pyrr][NTf ₂]	[C ₂ mim][FSI]	[C ₂ mim][NTf ₂]	[C ₄ C ₁ Pyrr][FSI]
MD [14]	143.9	137.3	134.6	123.9	–
γ_1^∞ based [15]	153.1	–	140.4	–	155.6
Experiment	142.5 ± 1.6 ^a	140.9 ± 1.8 ^b	125.7 ± 3.0 ^c	123.7 ± 1.3 ^d	147.3 ± 1.7 ^a

The extended uncertainties in this table are given as extended uncertainties with $k=2$ and confidence level 0.95

^aExperimental data on $\Delta_1^g H_m^o$ (298 K) were taken from Table 2

^bExperimental $\Delta_1^g H_m^o$ (298 K) was taken from Table S3

^cEmpirical $\Delta_1^g H_m^o$ (298 K) was taken from Table 4

^dExperimental $\Delta_1^g H_m^o$ (298 K) was taken from Table S5

uncertainties. Also the MD result for [C₂mim][NTf₂] of 123.9 kJ·mol⁻¹ is in very good agreement with the experiment (123.7 ± 1.3 kJ·mol⁻¹). Surprisingly, the MD result for [C₂mim][FSI] of 134.6 kJ·mol⁻¹ does not agree with our empirical value (125.7 ± 3.0 kJ·mol⁻¹), even taking into account a very large uncertainty ascribed to our estimate. However, our new results on enthalpies of vaporization could be useful for re-parameterization the MD method.

Another set of the theoretical vaporization enthalpies of [C₃C₁Pyrr][FSI], [C₄C₁Pyrr][FSI], and [C₂mim][FSI] were obtained with a Gas-Chromatographic (GC) method. In this method the activity coefficients at infinite dilution γ_1^∞ of a

volatile solute (typically an industrially relevant solvent such as benzene, methanol, acetone, denoted with a subscript 1) in an ionic liquid as a solvent (denoted with a subscript 2) are conveniently measured using gas-chromatography [16]. Usually, a set of γ_1^∞ -values for 20 to 40 solutes of different polarity is correlated with the solubility parameters of solutes, which leads to the vaporization enthalpy of the solvent (ionic liquid) under study. In our experience, the reliability of the GC method depends crucially on the right choice of solutes [17]. Rabhi et al. [15] determined γ_1^∞ -values for 53 solutes in $[\text{C}_3\text{C}_1\text{Pyrr}][\text{FSI}]$, $[\text{C}_4\text{C}_1\text{Pyrr}][\text{FSI}]$, and $[\text{C}_2\text{mim}][\text{FSI}]$. The significant spread of the correlated data led to a significant overestimation of the ILs vaporisation enthalpies, as shown in Table 5.

4 Conclusions

The experimentally determined absolute vapor pressures of pyrrolidinium-based ILs with the bis(fluorosulfonyl)imide anion are 10 times lower than that of analogous pyrrolidinium-based ILs with the bis(trifluoromethanesulfonyl)imide anion. Accordingly, the enthalpies of vaporization of the [FSI] containing ILs are higher. Simple additive rule was developed to get reasonable vaporization enthalpies of pyridinium and imidazolium-based ILs bearing the [FSI] anion. The evaluated values are useful for chemical engineering calculations for the processes involving ILs as catalysis, reaction media and separation phase.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s10765-021-02946-2>.

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