Volumetric, Transport and Surface Properties of [bmim][$MeSO_4$] and [emim][$EtSO_4$] Ionic Liquids As a Function of Temperature

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The effect of temperature on density, viscosity, and surface tension of 1-butyl-3-methylimidazolium methylsulfate [bmim][$MeSO_4$] and 1-ethyl-3-methylimidazolium ethylsulfate [emim][$EtSO_4$] ionic liquids (ILs) has been determined. Viscosity values of both ILs above-mentioned have been modeled by the Arrhenius equation and the Vogel−Fulcher−Tammann expression. Density, viscosity, surface tension, and surface excess energy of [bmim][$MeSO_4$] and [emim][$EtSO_4$] decreases as the temperature increases.

Introduction

In recent years, ionic liquids (ILs) have attracted increasing attention as replacements for conventional organic solvents in catalysis, separation processes, electrochemistry and many other fields.[1,2] From a chemical industry point of view, one of the most attractive features of ILs is their ability to be tailor-made for a specific purpose by careful selection of the cation, anion, or both.[2,3] However, given the enormous number of possible cation/anion combinations, IL design would be greatly assisted by theoretical models for estimation of thermophysical properties of intended combinations and their variation with temperature.[4]

Currently, a large quantity of ILs are available commercially or by synthesis.[2] The most commonly studied are the ILs based on imidazolium cations. Although there is a considerable amount of data related with this IL type, properties of 1-butyl-3-methylimidazolium methylsulfate are still scarce.[5,6] Therefore, in this article the density and viscosity from (293.15 to 353.15) K, surface tension from (303.15 to 333.15) K and standard entropy of 1-butyl-3-methylimidazolium methylsulfate ([bmim]-[$MeSO_4$]) and 1-ethyl-3-methylimidazolium ethylsulfate ([emim]-[$EtSO_4$]) ILs have been studied.

To test the quality of our experimental data, Arrhenius and Vogel−Fulcher−Tammann and Eötvös equations were used to model viscosity and surface tension, respectively.

Experimental Section

Water Content. The water content of [bmim][$MeSO_4$] and [emim][$EtSO_4$] (vide infra) were established employing a Mettler Toledo DL31 Karl Fischer titrator and using the one-component technique. The polarizing current for the potentiometric end-point determination was 20 A and the stop voltage 100 mV. The end-point criterion was the drift stabilization (3 $\mu gH_2O \cdot min^{-1}$) or maximum titration time (10 min). The measurement was corrected for the baseline drift, defined as the residual or penetrating water that the apparatus removes per minute. The uncertainty in experimental measurements has been found to be less than 7 %.

Chemicals. Ionic liquids 1-ethyl-3-methylimidazolium ethylsulfate ([emim][$EtSO_4$], ≥95 % purity, from Sigma-Aldrich Chemie GmbH, mass fraction chloride $< 3 \cdot 10^{-5}$), and 1-butyl-3-methylimidazolium methylsulfate ([bmim][$MeSO_4$] ≥ 95 % purity, from Sigma-Aldrich Chemie GmbH, mass fraction chloride $< 3 \cdot 10^{-5}$) were used. The main properties provided by manufacturer of both ILs are shown in Table 1. The water mass fraction was $1 \cdot 10^{-3}$ and $2 \cdot 10^{-3}$ for [bmim][$MeSO_4$] and [emim][$EtSO_4$], respectively. All the ionic liquid samples were dried by heating at 333.15 K for 24 h under reduced pressure. Then, in all experiments every pretreatment (filling and sealing of capillary) was carried out in a controlled atmosphere glovebox under dry nitrogen due to the highly hygroscopic nature of these ILs. Because of this, the ILs moisture was constant along the measurement. The means of three replicate measurements were reported.

Table 1. Properties of [emim][$EtSO_4$] and [bmim][$MeSO_4$] Given by the Manufacturer of These ILs (Sigma-Aldrich Chemie GmbH)

property	[bmim][$MeSO_4$]	[emim][$EtSO_4$]
molar mass/$g \cdot mol^{-1}$	250.32	236.29
electric conductivity/$\mu S \cdot cm^{-1}$	20 (298.15 K)	80 (298.15 K)
flash point/K	375	435
inflammation point/K	703	678
solubility in water	∞	∞
viscosity at room temperature/mPa·s	213.8	122.4
viscosity at 353 K/mPa·s	19.1	14.3
density at room temperature /$g \cdot cm^{-3}$	1.2129	1.2402
density at 353 K/$g \cdot cm^{-3}$	1.1748	1.2011

Table 2. Density, *ρ*, and Molecular Volume, *V*, Values of [bmim][$MeSO_4$] and [emim][$EtSO_4$] As a Function of Temperature

	[bmim][$MeSO_4$]	[emim][$EtSO_4$]
T/K	$\rho/g \cdot cm^{-3}$	$\rho/g \cdot cm^{-3}$
293.15	1.21295	1.24030
298.15	1.20956	1.23692
303.15	1.20627	1.23354
308.15	1.20302	1.23016
313.15	1.19975	1.22679
318.15	1.19650	1.22343
323.15	1.19325	1.22009
328.15	1.19001	1.21675
333.15	1.18678	1.21342
338.15	1.18355	1.21011
343.15	1.18033	1.20680
348.15	1.17713	1.20351
353.15	1.17393	1.20023

Density Measurements. The densities of [bmim][$MeSO_4$] and [emim][$EtSO_4$] were determined from (293.15 to 353.15) K (±0.01 K) at atmospheric pressure using an Anton Paar DMA

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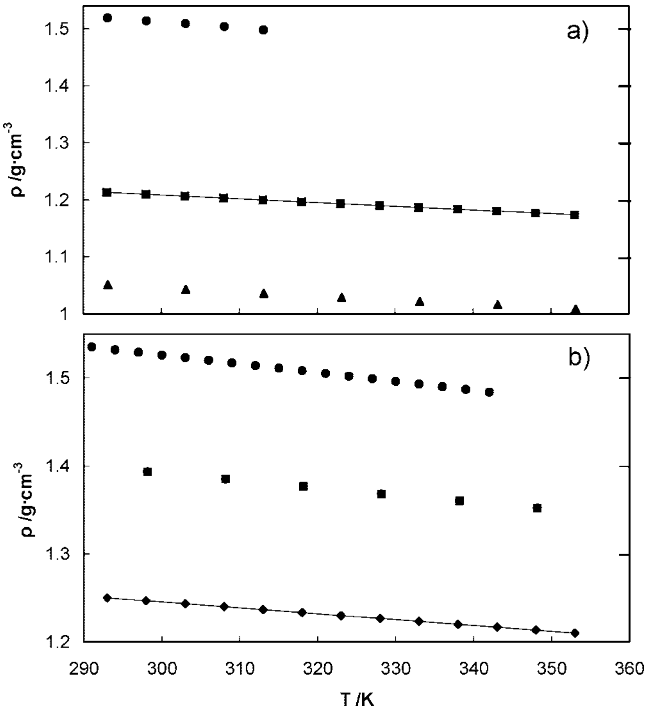

Figure 1. Comparison of experimental densities with those reported in literature. (a) Ionic liquids based on 1-butyl-3-methylimidazolium cation (●, 1-butyl-3-methylimidazolium bis(perfluoroethylsulfonyl)imide;[16] ■, 1-butyl-3-methylimidazolium methylsulfate; ▲, 1-butyl-3-methylimidazolium tricyanomethane.[17] (b) Ionic liquids based on 1-ethyl-3-methylimidazolium cation (●, 1-ethyl-3-methylimidazolium bis[(trifluoromethyl)sulfonyl]imide;[18] ■, 1-ethyl-3-methylimidazolium trifluoromethanesulfonate;[19] ♦, 1-ethyl-3-methylimidazolium ethylsulfate).

Table 3. Viscosity, η, and Surface Tension, γ, Values of [bmim][MeSO$_4$] and [emim][EtSO$_4$] As a Function of Temperature

	[bmim][MeSO$_4$]		[emim][EtSO$_4$]	
T/K	η/mPa·s	γ/mN·m^{-1}	η/mPa·s	γ/mN·m^{-1}
293.15			120.3	
303.15	173.5	42.72	73.7	47.25
313.15	104.2	42.09	48.5	46.78
323.15	66.8	41.60	33.7	46.24
333.15	45.3	41.02	24.5	45.65
343.15	32.2		18.5	
353.15	23.7		14.4	

5000 oscillating U-tube densimeter (Anton Paar GmbH, Graz, Austria). These measurements have been carried out taking into account the viscosity of the sample (viscosity correction). The uncertainty in the experimental measurements was found to be less than $\pm 1 \cdot 10^{-5}$ g·cm^{-3}. To validate the equipment and the method used here, the density of ultrapure water was determined and the measurements are in good agreement with literature values.[7]

Dynamic Viscosities. Dynamic viscosities of [bmim][MeSO$_4$] and [emim][EtSO$_4$] were determined at temperature range of (293.15 to 353.15) K, using an Anton Paar AMVn automated rolling ball microviscometer (Anton Paar GmbH, Graz, Austria).[8] As the principle of measurement is based on falling ball in a capillary, proper validation of the capillaries was achieved at all working temperatures with viscosity standard fluids supplied by Anton Paar GmbH from 1 to 200 mPa·s. The uncertainty in experimental measurements has been found to be less than 0.5 %.

Surface Tension Measurements. The surface tension measurements were performed using a Dataphysics OCA 15 plus pendant drop tensiometer (Dataphysics Instruments GmbH, Filderstadt, Germany), which is based on the pendant drop method.[9] An electronic syringe unit was used to form ionic liquid drops at the lower end of the dosing needle inside a thermal chamber thermostatted using a JULABO F12-EC bath with a temperature stability of ±0.03 K; a temperature range of (303.15 to 333.15) K was available. The drop shape is snapped from the images taken from a charge coupled device (CCD) camera to infer the surface tension using suitable software (SCA 20 from Dataphysics Instruments GmbH). The equipment was validated by fluids with known surface tensions. The uncertainty of surface tension measurements was estimated to be less than ± 0.05 mN·m^{-1}.

Results and Discussion

The density values reported in the literature for pure [bmim][MeSO$_4$][5] and [emim][EtSO$_4$][10–14] are in agreement with data obtained in this work (the difference between experimental data and those reported in literature are 0.18 %,[5] 0.40 %,[10] 0.15 %,[11] 0.06 %[12] and 0.57 %[13]). Given the large influence of impurities content of the commercial ILs abovementioned on their viscosity values,[14] the difference between the experimental data (vide supra) and those values of viscosity reported in literature (where the ILs used were synthesized by each research group) are less than 5 %.[5,11] The values obtained for the surface tension of [bmim][MeSO$_4$] and [emim][EtSO$_4$] are in agreement with the data reported in literature (<0.4 %[5] and <1.7 %,[12] respectively).

Density of [bmim][MeSO$_4$] and [emim][EtSO$_4$]. The density values of both ILs are shown in Table 2. As it can be observed, the density decreases as temperature increases. Given that the density depends mainly on anion mass,[15] the density of [emim][EtSO$_4$] is higher than that of [bmim][MeSO$_4$] in all range of temperature studied. This trend has been shown in figure 1, where ILs based on larger anions sizes present higher density values.[16–19] According to eqs 1 and 2, the density depends linearly on the temperature with a standard deviations (σ) for both ILs less than $4 \cdot 10^{-4}$ g·cm^{-3},[20] which is in agreement with the literature.[21]

$$\rho_{[\mathrm{bmim}][\mathrm{MeSO_4}]}/\mathrm{g \cdot cm^{-3}} = 1.40303 - 6.49 \cdot 10^{-4} \cdot T/\mathrm{K} (\sigma < 4 \cdot 10^{-4} \mathrm{g \cdot cm^{-3}}) \quad (1)$$

$$\rho_{[\mathrm{emim}][\mathrm{EtSO_4}]}/\mathrm{g \cdot cm^{-3}} = 1.43596 - 6.68 \cdot 10^{-4} \cdot T/\mathrm{K} (\sigma < 6 \cdot 10^{-5} \mathrm{g \cdot cm^{-3}}) \quad (2)$$

In eqs 1 and 2, $\rho_{[\mathrm{bmim}][\mathrm{MeSO_4}]}$, $\rho_{[\mathrm{emim}][\mathrm{EtSO_4}]}$, and T are, respectively, the densities of [bmim][MeSO$_4$] and [emim][EtSO$_4$] and temperature.

The volume expansivity of pure ILs, also known as thermal expansion coefficient, α, was calculated from density values. α is described by the relation between molar volume (V_m) or density of the IL and temperature (T) (eq 3).[5,13]

$$\alpha = \frac{1}{V_m}\left(\frac{\partial V_m}{\partial T}\right)_P = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P = -\left(\frac{\partial \ln \rho}{\partial T}\right)_P \quad (3)$$

where the subscript P indicates that this relationship is fulfilled at constant pressure. The α values of [bmim][MeSO$_4$] and [emim][EtSO$_4$] are $5.3 \cdot 10^{-4}$ K^{-1} and $5.5 \cdot 10^{-4}$ K^{-1}, respectively. The α values of [bmim][MeSO$_4$] and [emim][EtSO$_4$] are in agreement with the literature.[5,13]

Viscosity of [bmim][MeSO$_4$] and [emim][EtSO$_4$]. The viscosity values, η, of [bmim][MeSO$_4$] and [emim][EtSO$_4$] are shown in Table 3. As it can be observed, the viscosity decreases as temperature increases. The increase of water content on the viscosity values of the studied ILs[6] and the delocalization of

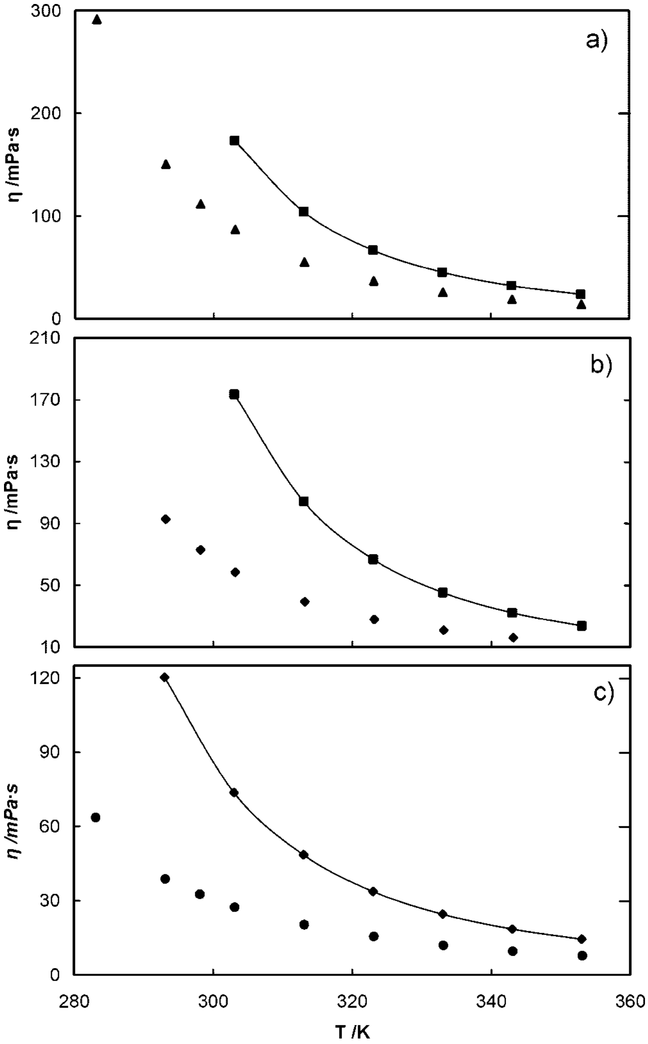

Figure 2. Comparison of experimental viscosities with those reported in literature. (a) Ionic liquids based on 1-butyl-3-methylimidazolium cation (▲, 1-butyl-3-methylimidazolium bis(perfluoroethylsulfonyl)imide;[16] ■, 1-butyl-3-methylimidazolium methylsulfate). (b) Ionic liquids based on 1-ethyl-3-methylimidazolium cation (♦, 1-ethyl-3-methylimidazolium bis[(trifluoromethyl)sulfonyl]imide;[18] ■, 1-ethyl-3-methylimidazolium ethylsulfate). (c) Ionic liquids based on methylsulfate anion (♦, 1-butyl-3-methylimidazolium methylsulfate; ●, 1,3-dimethylimidazolium methylsulfate[23]).

the charge over the higher anions seem to favor lower viscosities. Otherwise, longer alkyl chains on the cation result in an increase in viscosity.[22] These tendencies have been shown in Figure 2, where an IL based on higher anions sizes (Figure 2a,b) and smaller chains on the cation (Figure 2c) present lesser viscosity values.[16,18,23]

To correlate the variation of viscosity of both ILs with the temperature, the well-known Arrhenius equation has been used, eq 4.

$$\eta = \eta_\infty \cdot e^{\left(\frac{E_a}{RT}\right)} \qquad (4)$$

In eq 4, η_∞, E_a, R, and T are the pre-exponential factor (viscosity at infinite temperature), activation energy, universal gas constant, and temperature, respectively. However, particularly for ILs, a better correlation has been found in literature. It is Vogel−Fulcher−Tammann equation (VFT), eq 5.[24–29]

$$\eta = A \cdot e^{\left(\frac{B}{T-T_0}\right)} \qquad (5)$$

where A, B, and T_0 are fit parameters. The fit parameters of eqs 4 and 5 are shown in Table 4.

Surface Tension of [bmim][MeSO$_4$] and [emim][EtSO$_4$]. The surface tension, γ, of [bmim][MeSO$_4$] and [emim][EtSO$_4$] are shown in Table 3. The surface tension decreases as temperature increases. Comparing two works of Pereiro et al. (2006 and 2007) and taking into account the surface tensions of 1,3-dimethylimidazolium methylsulfate and 1-butyl-3-methylimidazolium methyl sulfate reported in literature,[5,23] γ increases as imidazolium cation size decreases. This tendency has been shown in Figure 3, where 1,3-dimethylimidazolium methylsulfate presents higher surface tension than that of [bmim][MeSO$_4$].[23] Here, the same trends are shown (Table 3).

Surface Excess Entropy. Surface excess entropy (S_a) of both studied ILs can be calculated using the slope of the profile of surface tension versus temperature, as shown in eq 6.[30] The values of S_a of [bmim][MeSO$_4$] and [emim][EtSO$_4$] are shown in Table 5.

$$S_a = -(\partial\gamma/\partial T)_P \qquad (6)$$

Surface Excess Energy. Surface excess energy (E_a) can be calculated using the surface excess entropy given by eq 7.[30]

$$E_a = \gamma - T(\partial\gamma/\partial T)_P \qquad (7)$$

The E_a values as a function of temperature are shown in Table 5. The values of E_a for the two ILs are much lower than those of the fused salts ($146\cdot10^{-3}$ at 293.15 K for fused NaNO$_3$). The E_a of [bmim][MeSO$_4$] and [emim][EtSO$_4$] are close to that of organic solvents ($67\cdot10^{-3}$ and $51.1\cdot10^{-3}$ J$\cdot$m^{-2} at 298.15 K for benzene and n-octane, respectively).[30]

For a pure component, the relationship between surface tension and temperature can be described using the empirical constant (k) of Eötvös equation (eq 8), which is related with the polarity, critical temperature (T_c), and molar volume (V_m) of the pure compound.[30]

$$\gamma \cdot \sqrt[3]{V_m^2} = k(T_c - T) \qquad (8)$$

In every studied IL, the empirical constant (k) was calculated from the slope of the linear regression of the product $\gamma\cdot(V_m^2)^{1/3}$ against absolute temperature (the R^2 of each regression is > 0.99). The k values for [bmim][MeSO$_4$] and [emim][EtSO$_4$], shown in Table 5, are closer to that of organic media (for the majority of these compounds k are around $2\cdot10^{-7}$ J$\cdot$K^{-1}) than to those of the fused salts (for NaCl $k = 4\cdot10^{-8}$ J$\cdot$K^{-1}).[31] Therefore, taking in mind the k of Eötvös equation, the [bmim][MeSO$_4$] and [emim][EtSO$_4$] present polarities more similar to those of organic chemicals.

The Interstice Model in the Calculation of Volumetric Properties of [bmim][MeSO$_4$] and [emim][EtSO$_4$] ILs. In this work, an interstice model was used to calculate the intersticial volume of pure [bmim][MeSO$_4$] and [emim][EtSO$_4$] ILs as a function of temperature. This model is a simple algorithm based on four assumptions related with the geometry features of the ionic liquids. A deeper description of employed model is reported in literature,[13] where the average volume of the interstices (ν) was calculated by eq 9.

$$\nu = 0.6791\sqrt{\left(\frac{kT}{\gamma}\right)^3} \qquad (9)$$

where k and T are Boltzmann constant (J K^{-1}) and temperature (K), respectively. According to eq 9, the ν values of [bmim][MeSO$_4$] and [emim][EtSO$_4$] were calculated as function of temperature. Then, the volume of all interstices ($\Sigma\nu$) and molar volume (V_m) are calculated from eqs 10 and 11, respectively.

Table 4. Fit Parameters of Arrhenius and VFT Equations Applied to the Viscosity Values of [bmim][MeSO$_4$] and [emim][EtSO$_4$]

	Arrhenius[a]			VFT[b]			
	η_∞	E_η	σ	T_0	B	A	σ
	10^{-4} mPa·s	kJ·mol^{-1}	mPa·s	K	K	mPa·s	mPa·s
[bmim][MeSO$_4$]	4.48	30.3	0.1	183.33	808.29	0.20	0.04
[emim][EtSO$_4$]	1.33	35.4	0.1	182.10	669.85	0.29	0.05

[a] Equation 4. [b] Equation 5.

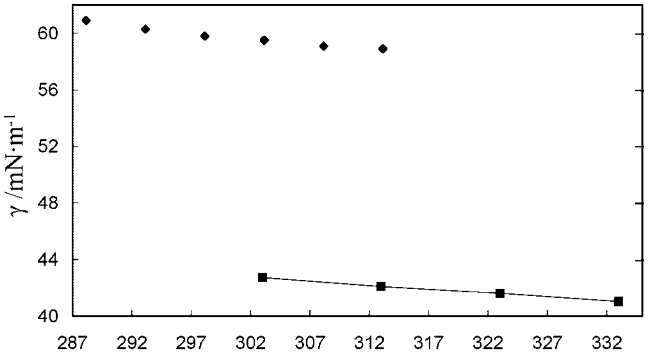

Figure 3. Comparison of experimental surface tensions with those reported in literature (■, 1-butyl-3-methylimidazolium methylsulfate; ♦, 1,3-dimethylimidazolium methylsulfate[23]).

Table 5. Surface Excess Entropy (S_a), Surface Excess Energy (E_a), and k Constant of Eötvös Equation of [bmim][MeSO$_4$] and [emim][EtSO$_4$]

	[bmim][MeSO$_4$]	[emim][EtSO$_4$]
	$S_a/10^6$ J·K^{-1}·m^{-2}	$S_a/10^6$ J·K^{-1}·m^{-2}
	55.9	53.4
T/K	$E_a/10^3$ J·m^{-2}	$E_a/10^3$ J·m^{-2}
303.15	25.77	31.06
313.15	24.58	30.06
323.15	23.54	28.98
333.15	22.40	27.86
	[a]k/J·K^{-1}	[a]k/J·K^{-1}
298.15	$1.22 \cdot 10^{-7}$	$1.44 \cdot 10^{-7}$

[a] Equation 8.

Table 6. Estimation of Volume Expansivity of Pure [bmim][MeSO$_4$] and [emim][EtSO$_4$]

$\alpha/10^4$K^{-1}	[bmim][MeSO$_4$]	[emim][EtSO$_4$]
from equation 3	5.44	5.48
from equation 12	5.98	5.57

$$\sum \nu = 2N\nu \tag{10}$$

$$V_m = V_i + 2N\nu \tag{11}$$

where V_i is the inherent volume. When the temperature increases, if the expansion of IL molar volume only results from the expansion of the interstices, the expression of α, eq 3, will be derived from the interstice theory, eq 12.

$$\alpha = \frac{1}{V_m}\left(\frac{\partial V_m}{\partial T}\right)_p = \frac{3}{2}\cdot\frac{2N\nu}{V_m T} = \frac{3N\nu}{V_m T} \tag{12}$$

As can be seen in Table 6, the α values calculated from eqs 3 and 12 are in good agreement. Therefore, the application of this interstice theory of [bmim][MeSO$_4$] and [emim][EtSO$_4$] ILs is satisfactory.

Conclusions

In this work, the effect of temperature on density, volume expansivity, viscosity, surface tension, surface excess energy, and volume of interstices of [bmim][MeSO$_4$] and [emim][EtSO$_4$] ILs have been determined. Besides surface excess entropy, k of Eötvös equation have been also calculated. The viscosity of both ILs have been modeled by Arrhenius and Vogel−Fulcher−Tammann equations.

As expected, the density, viscosity, surface tension, and surface excess energy of [bmim][MeSO$_4$] and [emim][EtSO$_4$] decrease as the temperature increases.

The Arrhenius and Vogel−Fulcher−Tammann equations fit the viscosity values of [bmim][MeSO$_4$] and [emim][EtSO$_4$] with correlation coefficients (estimated versus experimental values) higher than 0.99 and σ less than 0.1 mPa·s. Therefore, both equations are adequate to model their viscosity values.

Taking into account the k of Eötvös equation and the surface excess energy, the [bmim][MeSO$_4$] and [emim][EtSO$_4$] present polarities and characteristics more closer to those of organic chemicals than to those of funded salts.

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