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Electrical Conductivity and Viscosity of Aqueous Binary Mixtures of 1-Alkyl-3-methyl Imidazolium Tetrafluoroborate at Four Temperatures[†]

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Dynamic viscosity and specific electrical conductivity on three binary systems of 1-alkyl-3-methyl imidazolium tetrafluoroborate [CnMIM][BF_4] with water covering all ranges of concentrations are presented. Viscosity was measured at 15.0 °C, 25.0 °C, 35.0 °C, and 45.0 °C, while conductivity was measured at those four temperatures for equimolar mixtures only and at 25 °C over the whole composition range. The alkyl chains of the ILs used were ethyl [EMIM][BF_4], butyl [BMIM][BF_4], and hexyl [HMIM][BF_4]. Note that [HMIM][BF_4] is only partially miscible in water at the studied temperatures and atmospheric pressure. Viscosity deviations and molar conductivity were derived from experimental data and fitted to suitable equations. Experimental data were also compared with those previously published, and a good agreement was observed. Transport properties were discussed in terms of Walden's rule, which is partially correct. This paper complements one recently published by us, where density and surface tension at 25 °C for exactly the same systems are presented. The goal of this work is to study the effect on the transport magnitudes of the alkyl chain length in the *n*-alkyl methyl imidazolium cation and to compare the two main transport magnitudes.

Introduction

Ionic liquids (ILs) are molten salts made up of an anion and an organic cation and were discovered in 1914 by Paul Walden, the ethylammoniun nitrate being the first IL, but their systematic study began with the present century.[1] One or both ions are large, and the cation is usually organic and has a low degree of symmetry. For this reason, anion−cation interactions are weak, and hence their melting points are below 100 °C and usually below room temperature (in fact, they are also known as room temperature ionic liquids, RTIL); however, if the size of the cation is too large, the Van der Wall forces will increase the fusion temperature.[1] Different combinations of the multiple anions and cations lead to billions of possible ILs with very different physicochemical properties. So, we can design ionic liquids with the appropriate set of properties according to our purposes. Due to their ionic nature, all ILs have peculiar properties such as good solvents for organic and inorganic compounds including some metal salts, lack of vapor pressure, electrical conductivity, and high thermal, and electrochemical stability. All these characteristics make ionic liquids promising compounds for being used for batteries, organic synthesis, extractions, and alloy electro-deposition, as well as potential "green solvent" replacements for volatile organic compounds (VOCs).[1]

Among the numerous proposed industrial applications of ionic liquids (ILs), their use to improve electrochemical processes is one of the most interesting. To do that, it is necessary to know the transport physical properties (mainly viscosity and electrical conductivity) of the different ILs, both pure and mixed with different solvents, including water. In this communication, we present experimental measurements of dynamic viscosity and specific electrical conductivity on three binary mixtures of 1-alkyl-3-methyl imidazolium tetrafluoroborate [CnMIM][BF_4] with water. Viscosity was measured at 15.0 °C, 25.0 °C, 35.0 °C, and 45.0 °C for all ranges of concentrations and electrical conductivity only for the equimolar mixture, while it was measured for all concentrations at 25.0 °C. The alkyl chain of the ILs used is ethyl [EMIM][BF_4], butyl [BMIM][BF_4], and hexyl [HMIM][BF_4]. Let us note that [HMIM][BF_4] is only partially miscible in water at those temperatures, while for alkyl chains longer than hexyl the resulting ILs are not at all miscible with water at room temperature and atmospheric pressure. It is also important to say that this family of ILs is one of the most popular and suitable for a number of electrochemical applications.[1]

To date, due to the lack of predicting models, the only way to know the physical properties of the IL mixtures is experimentally. However, the measurement of physical properties of ILs mixed with different substances was scarce until 2005.[2] Since then, many papers have been published performing measurements on mixtures of ionic liquids with different kind of solvents such as water, ethanol, aromatic compounds, or other ionic liquids, and the number of published papers is continuously growing.[1] With respect to the two magnitudes measured here and for these or similar IL families, we have found ten papers presenting dynamical viscosity[3−13] and six containing experimental measurements of electric conductivity.[11−16] Such data are crucial to develop any kind of proposed electrochemical applications of IL aqueous mixtures. Aqueous mixtures are especially important because it has been proved that water decreases viscosity exponentially[3] and greatly increases electrical conductivity.[15,16]

This paper complements one recently published by us, where we presented density and surface tension for the same systems as here at 25 °C.[17] The goal of this work is to study the effect of the alkyl chain length of the *n*-alkyl methyl imidazolium cation on the transport magnitudes and also to compare the two

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Table 1. Water Mass Fraction, Molar Mass, Density, Viscosity, and Specific Conductivity at 25.0 °C for [EMIM][BF$_4$], [BMIM][BF$_4$], [HMIM][BF$_4$], and Water[a]

	$10^6\ w$	M / g·mol^{-1}	ρ / g·cm^{-3}	η / mPa·s	η_{lit} / mPa·s	κ / mS·cm^{-1}	κ_{lit} / mS·cm^{-1}
[EMIM][BF$_4$]	220	197.98	1.2799[17]	36.93	37.19[8]	16.3	15.53[15]
[BMIM][BF$_4$]	1040	226.03	1.2012[17]	99.20	100.66[9] 94.26[10] 153.78[12] 106.82[13]	3.52	3.53[15] 5.86[13]
[HMIM][BF$_4$]	273	254.09	1.1454[17]	190.92	174.1[20] 210.4[21]	1.26	1.228[15]
water	–	18.01	0.9970[17]	0.8903	–	0.0264	–

[a] For viscosities and specific conductivities, values in the literature are also included.

transport magnitudes. Note that both magnitudes measure the movement of the ions: in one magnitude it is generated by an electrical field (conductivity) and in the other magnitude by a mechanical one (viscosity). In fact, they can be usually related using Walden's rule.

Experimental Procedure

Dynamic viscosity, η, was measured in an Anton-Paar SVM 3000 Stabinger viscodensimeter, which was thermostatted with a Peltier cell with an uncertainty of ± 0.01 °C. The repeatability in the measurement of the viscosity is ± 0.1 %, and its uncertainty is ± 0.35 %.

Electrical conductivity was measured using a Crison conductivimeter, which has repeatability of ± 1 % and uncertainty better than 5 %. This conductivity uses a fixed 500 Hz ac current. The samples were thermostatted in an external bath with an uncertainty of ± 0.1 °C. Details of the electrical conductivity measurement were given by Prego et al.[18] The ILs were purchased from Solvent Innovation (which has been recently adsorbed by Merck), and the purities are better than 99 % for all ILs except [EMIM][BF$_4$] which is better than 98 %. The water used for the mixtures was Millli-Q grade. Due to the hygroscopic character of the ILs, the chemicals were open from their original tin and mixed with water in a dry chamber with relative humidity grade lower than 10 %. This humidity grade, and the speed of the mixture process, ensures that our original ILs were not contaminated with water as we recently studied quantitatively.[19] Mixtures were bottled and sealed before taking them out of the chamber. The water mass fraction of the three pure ILs is lower than $1 \cdot 10^{-3}$ as certified by the dealer, and they were not further purified. The exact value of original water content is included in Table 1. It is important to note that [HMIM][BF$_4$] is miscible in water at 25.0 °C only for IL mole fraction higher than 0.28 or lower than 0.01.[14]

Results and Discussion

Table 1 shows the molar mass (M), density (ρ) (published by Rilo et al.[17]), viscosity (η), and specific conductivity (κ) of the pure compounds used. Comparing viscosity and specific conductivity data measured here with other data published in the literature,[8−11,13,15,20,21] it can be observed that electrical conductivity data are very similar to that previously published,[15] while a great dispersion exists for available viscosity data, our values being within the published ones.

Table 2. Dynamic Viscosity, η, of All Mixtures Measured at Four Temperatures[a]

x_w	t/°C: 15.0	25.0	35.0	45.0
	η/mPa·s			
	[EMIM][BF$_4$] + H$_2$O			
0.0000	55.5	36.9	26.0	19.17
0.0985	36.2	25.2	18.45	14.01
0.2191	23.88	16.66	12.64	9.90
0.3053	18.26	13.51	10.39	8.23
0.3982	13.26	10.02	7.85	6.33
0.4916	10.29	7.91	6.26	5.09
0.5989	7.28	5.62	4.49	3.68
0.6991	5.43	4.23	3.40	2.81
0.8029	3.18	2.50	2.03	1.696
0.8991	2.32	1.820	1.477	1.200
1.0000	1.137	0.890	0.719	0.597
	[BMIM][BF$_4$] + H$_2$O			
0.0000	173.5	99.2	61.3	40.4
0.0476	134.1	78.7	49.8	33.5
0.1091	108.4	65.6	42.4	29.1
0.1430	89.3	55.1	36.3	25.3
0.2389	58.2	37.7	25.9	18.62
0.3078	45.6	30.2	21.1	15.47
0.4000	32.5	22.1	15.81	11.81
0.5054	21.1	14.72	10.80	8.24
0.6266	13.77	9.82	7.33	5.68
0.6992	9.78	7.07	5.34	4.18
0.7981	6.69	4.89	3.75	2.96
0.9032	3.83	2.79	2.47	1.717
	[HMIM][BF$_4$] + H$_2$O			
0.0000	357.5	190.9	110.3	68.5
0.1101	200.6	113.9	69.4	45.1
0.1679	151.0	89.1	56.2	37.5
0.2812	100.3	61.6	40.3	27.8
0.3581	65.4	41.6	28.1	19.89
0.4987	38.7	25.5	17.72	12.92
0.5075	36.4	24.0	16.76	12.25
0.5730	29.4	19.60	13.83	10.19
0.7200	17.55	10.82	7.76	5.85

[a] Concentration is expressed in mole fraction of the water, x_w.

Table 2 includes the dynamic viscosity data (η) measured for the three systems studied and at four temperatures as a function of mole fraction of water, x_w. In Figure 1(a) to (d), we plot the ln η vs x_w for the data at 15 °C, 25 °C, 35 °C, and 45 °C, respectively. As shown, data for each system form straight lines up to $x_w = 0.1$, following an exponential equation as previously observed in similar systems[3,9,11]

$$\ln \eta = \ln \eta_{\mathrm{IL}} - \frac{x_w}{a} \tag{1}$$

where η_{IL} is the viscosity of pure IL. The best fitting parameters a^{-1} for the three systems are given in Table 3, where the standard deviation, s, is also included. In any case, it is not a perfect exponential behavior because the corresponding fitting parameter is not equal to $a = (\ln \eta_{\mathrm{IL}} - \ln \eta_w)^{-1}$. In Figure 1(b) we include a dotted straight line for each system which represents that perfect exponential behavior. As observed, the linearity is lost for the most diluted mixtures ($x_w > 0.1$, which represents a mass fraction of water of 0.4).

Viscosity deviations for the mixture, $\Delta\eta$, were calculated using the usual equation[4−11]

$$\Delta\eta = \eta - (x_{\mathrm{IL}} \cdot \eta_{\mathrm{IL}} + x_{\mathrm{w}} \cdot \eta_{\mathrm{w}}) \qquad (2)$$

Results vs x_{w} are plotted in Figure 2(a) to (c) for aqueous systems of [EMIM][BF$_4$], [BMIM][BF$_4$], and [HMIM][BF$_4$], respectively. The $\Delta\eta$ curves for the three systems and four temperatures studied are similar, decreasing its absolute value with temperature and increasing it with the alkyl chain length of the IL. They have their minimum skewed to rich-IL concentration, about $x_{\mathrm{w}} = 0.3$. The data have been fitted using a new model with only four free parameters[17,22]

$$\Delta\eta = K \cdot x_{\mathrm{IL}}^{n} \cdot x_{\mathrm{w}} + K' \cdot x_{\mathrm{IL}} \cdot x_{\mathrm{w}}^{n'} \qquad (3)$$

where K, n, K', and n' are fitting parameters, with the exponents n and n' being positive. The resulting curves are included in Figures 2(a) to (c), and the fitting parameter values, with the corresponding standard deviation, s, are given in Table 4.

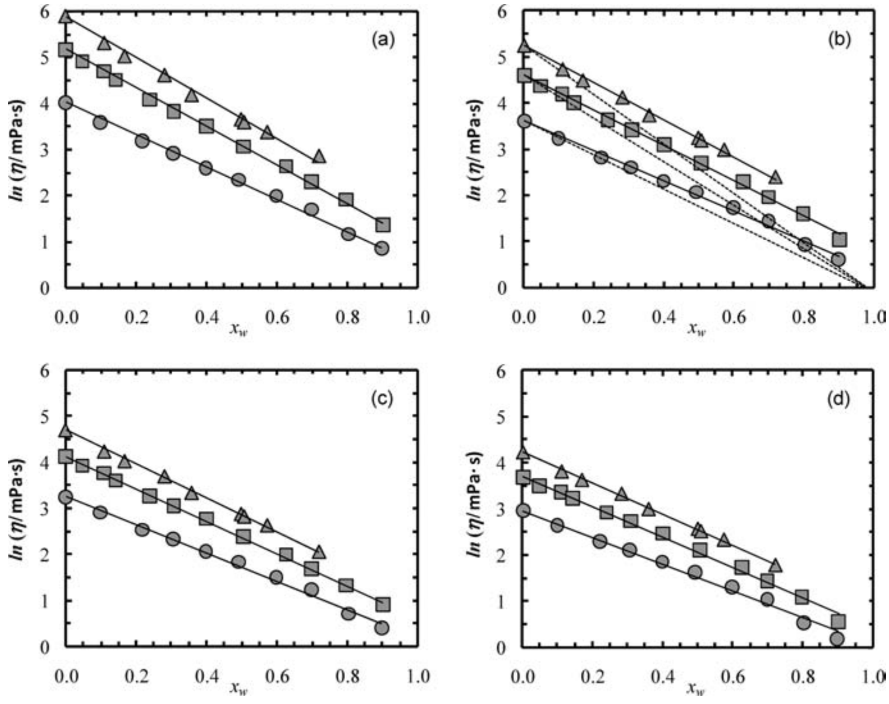

Figure 1. Natural logarithm of dynamic viscosity vs mole fraction of water for the aqueous mixtures of [EMIM][BF$_4$] (dot symbols), [BMIM][BF$_4$] (square symbols), and [HMIM][BF$_4$] (triangle symbols) at (a) 15.0 °C, (b) 25.0 °C, (c) 35.0 °C, and (d) 45.0 °C. Solid lines are the best fit of eq 1 with the parameters given in Table 3. Dotted lines in (b) represent a perfect exponential behavior.

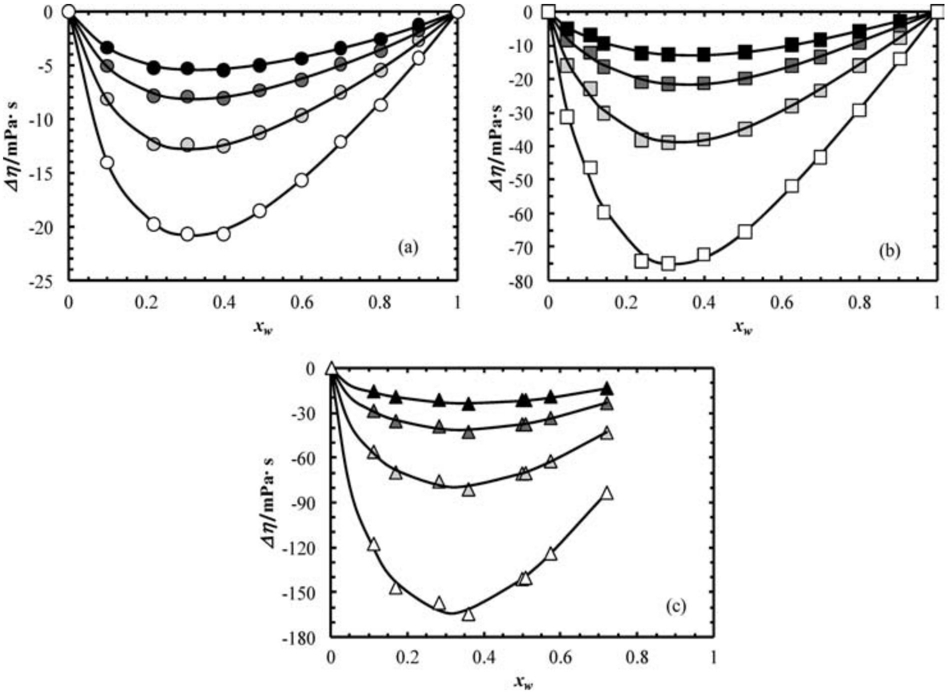

Figure 2. Dynamic viscosity deviations vs the water mole fraction at 15.0 °C (white symbols), 25.0 °C (light gray symbols), 35.0 °C (dark gray symbols), and 45.0 °C (black symbols) for aqueous mixtures of (a) [EMIM][BF$_4$], (b) [BMIM][BF$_4$], and (c) [HMIM][BF$_4$]. Solid lines are the best fit of eq 3 to the data with the parameters given in Table 4.

Table 3. Value of (a^{-1}) for the Best Fit of Natural Logarithm of Viscosity vs the Mole Fraction of Water Following Equation 1[a]

		[EMIM][BF$_4$] + H$_2$O		
t/°C	15.0	25.0	35.0	45.0
a^{-1}/ln [mPa·s]	3.498	3.265	3.071	2.904
s	0.070	0.074	0.079	0.093
		[BMIM][BF$_4$] + H$_2$O		
t/°C	15.0	25.0	35.0	45.0
a^{-1}/ln [mPa·s]	4.164	3.828	3.497	3.286
s	0.062	0.068	0.047	0.078
		[HMIM][BF$_4$] + H$_2$O		
t/°C	15.0	25.0	35.0	45.0
a^{-1}/ln [mPa·s]	4.403	4.043	3.684	3.388
s	0.098	0.055	0.040	0.035

[a] Standard deviation (s) is included.

Table 4. Best Fitting Parameters for Equation 3 Used to Reproduce Viscosity Deviation Data[a]

	t/°C			
	15.0	25.0	35.0	45.0
		[EMIM][BF$_4$] + H$_2$O		
K/mPa·s	−46.99	−45.29	−28.04	−18.88
n	3.01	3.63	3.65	3.76
K'/mPa·s	−46.79	−31.18	−20.87	−14.68
n'	0.60	0.73	0.76	0.79
s	0.185	0.212	0.149	0.106
		[BMIM][BF$_4$] + H$_2$O		
K/mPa·s	−234.62	−106.39	−53.52	−27.82
n	2.81	2.55	2.12	2.04
K'/mPa·s	−155.75	−82.74	−43.81	−27.92
n'	0.66	0.67	0.66	0.68
s	1.67	0.978	0.592	0.371
		[HMIM][BF$_4$] + H$_2$O		
K/mPa·s	−389.20	−161.18	−94.32	−82.44
n	2.42	2.21	1.66	1.41
K'/mPa·s	−297.45	−153.19	−63.71	−14.41
n'	0.50	0.52	0.46	0.20
s	2.81	1.49	0.882	0.568

[a] The corresponding standard deviations, s, are included.

Not published papers reporting η for any aqueous mixture of [HMIM][BF$_4$], one[8] paper of [EMIM][BF$_4$] + water, and five papers reporting viscosity for aqueous solutions of [BMIM][BF$_4$][9−13] appear[23] to exist (but none of them at 15 °C). Comparison of the present measurements with those published before in the literature indicates that the agreement is good within experimental uncertainties. Even for those published by Liu et al.[12] that report a much bigger value for the pure compound (see Table 1), the values for the different aqueous mixtures agree with those published here.

In Table 5, the electrical conductivity data, κ, at 25.0 °C for [EMIM][BF$_4$], [BMIM][BF$_4$], and [HMIM][BF$_4$] aqueous mixtures for all measured compositions (expressed as water mole fraction, x_w, and molarity, c, of the ionic liquid) are shown. Data for the system [EMIM][BF$_4$] + water were published previously.[16] Figure 3 shows the temperature dependence of κ for the equimolar mixture of the three systems studied, and the data appear in Table 6. The three equimolar mixtures follow a linear behavior with temperature in the range measured. From specific conductivity data, κ, molar conductivities, Λ, were extracted following the usual equation[24]

$$\Lambda = \kappa / c \qquad (4)$$

Resulting curves are plotted in Figure 4 against molarity of the ionic liquid. Molar conductivity data have been fitted to an equation derived from Onsager theory[24] that was also used by Stoppa et al.[15]

$$\Lambda = D_1 - D_2\sqrt{c} + D_3 \cdot c \cdot \ln c + D_4 \cdot c \qquad (5)$$

where D_i are fitting constants. As observed in Figure 4, the resulting fits reproduce the data with high quality. Table 7 shows the best D_i fitting parameters for the three systems with the corresponding standard deviation, s.

Only two papers have been found reporting κ vs x_{IL} data in the same systems as those presented here. One was published by us[16] and the other one by Stoppa et al.[15] Agreement is fine in shape, but our data are about 10 % lower in the conductivity maximum than those published by other authors.[15] The origin of discrepancy could be contamination of the ILs as Stoppa et al. suggest, but it is also necessary to take into account that the ac electrical frequency used for measurement is different in both laboratories, fixed at 500 Hz in ours and extrapolated to infinitum in the other one.[15] The influence of the ac frequency used to measure κ in the obtained results for these mixtures has not been published to our knowledge.

Molar conductivity and viscosity were related following Walden's rule[24]

$$\Lambda \cdot \eta = K \qquad (6)$$

where K is constant at a fixed temperature in the original formulation of this rule. To find that product, we have used

Table 5. Electrical Conductivity Data Measured for All Ranges of Concentrations Expressed in Water Mole Fraction, x_w, and Molarity, *M*, at 25.0 °C

[EMIM][BF$_4$] + H$_2$O			[BMIM][BF$_4$] + H$_2$O		
x_w	c/mol·L^{-1}	κ/mS·cm^{-1}	x_w	c/mol·L^{-1}	κ/mS·cm^{-1}
0.0000	6.463	16.3	0.0000	5.309	3.52
0.3665	6.027	41.0	0.2235	5.154	7.85
0.5498	5.610	60.7	0.3705	5.016	13.00
0.6599	5.223	76.4	0.4835	4.858	18.63
0.7332	4.860	81.5	0.6173	4.573	27.50
0.7856	4.507	84.4	0.7167	4.249	36.40
0.8249	4.159	85.2	0.7770	3.963	42.00
0.8555	3.815	84.7	0.8168	3.707	45.50
0.8799	3.477	83.6	0.8458	3.469	48.40
0.8999	3.146	81.8	0.8684	3.241	50.00
0.9166	2.823	79.0	0.8879	3.006	50.80
0.9307	2.508	74.3	0.9031	2.792	50.90
0.9428	2.201	71.0	0.9151	2.599	50.80
0.9533	1.902	65.6	0.9245	2.428	50.50
0.9625	1.610	61.3	0.9322	2.275	49.80
0.9706	1.326	54.3	0.9396	2.117	48.40
0.9778	1.049	47.6	0.9457	1.976	48.10
0.9842	0.778	40.3	0.9520	1.816	47.30
0.9900	0.513	29.5	0.9572	1.677	46.50
0.9952	0.254	17.9	0.9615	1.553	45.40
[HMIM][BF$_4$] + H$_2$O			0.9654	1.434	44.10
			0.9713	1.196	41.50
0.0000	4.507	1.261	0.9801	0.924	36.40
0.1164	4.450	2.02	0.9861	0.677	31.40
0.2364	4.383	3.37	0.9896	0.523	26.80
0.3232	4.322	4.84	0.9930	0.364	21.10
0.3942	4.261	6.27	0.9951	0.257	16.42
0.4548	4.200	7.98	0.9961	0.209	14.31
0.5294	4.108	10.56	0.9974	0.142	10.53
0.5896	4.015	13.08	0.9982	0.100	7.88
0.6520	3.891	16.17	0.9990	0.055	4.75
0.7184	3.710	19.91	0.9996	0.021	2.00

Table 6. Electrical Conductivity Data for the Aqueous Equimolar Mixtures of the Given IL at Four Temperatures

	κ/mS·cm^{-1}		
	[EMIM][BF$_4$]	[BMIM][BF$_4$]	[HMIM][BF$_4$]
t/°C	$x_{IL} = 0.5067$	$x_{IL} = 0.5046$	$x_{IL} = 0.5093$
15.0	40.0	14.04	6.25
25.0	52.6	20.2	9.83
35.0	59.0	26.1	13.82
45.0	69.5	32.5	16.47

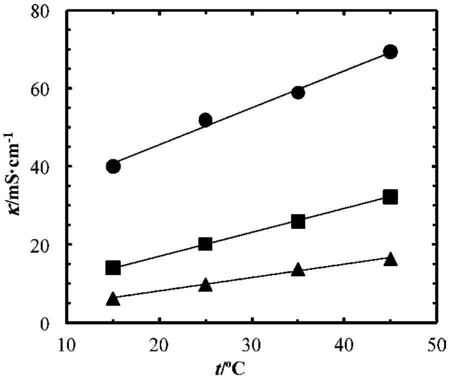

Figure 3. Temperature dependence of the equimolar mixture of aqueous solutions of [EMIM][BF_4] (dot symbols), [BMIM][BF_4] (square symbols), and [HMIM][BF_4] (triangle symbols). Lines are the best fit of a straight line.

Table 7. Best Fitting Parameters of Equation 5 Used to Reproduce Molar Conductivity Data[a]

	[EMIM][BF_4]	[BMIM][BF_4]	[HMIM][BF_4]
D_1	127.10	113.83	122.51
D_2	174.48	145.73	91.52
D_3	−23.35	−16.85	0.01
D_4	93.13	70.17	15.94
s	0.765	0.556	0.086

[a] The corresponding standard deviations, *s*, are included.

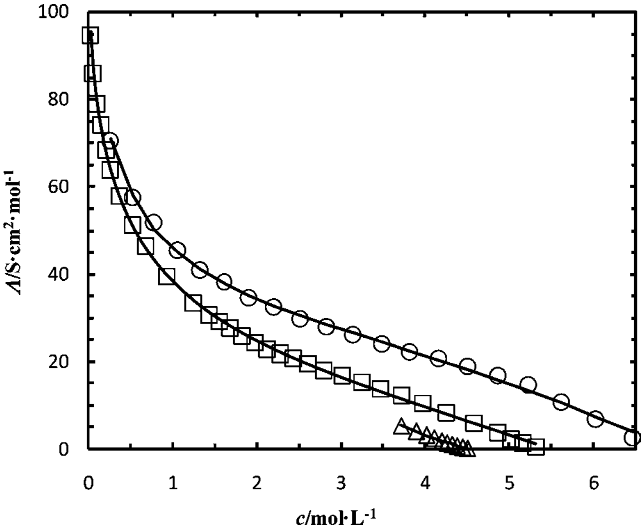

Figure 4. Molar conductivity vs molarity for aqueous solutions of [EMIM][BF_4] (dot symbols), [BMIM][BF_4] (square symbols), and [HMIM][BF_4] (triangle symbols) at 25.0 °C. Lines are the best fit of eq 5 with the parameters given in Table 7.

the exponential fit to η given by eq 1 with the fitting parameter included in Table 3 at 25.0 °C. The used curves are shown in Figure 1(b). In Figure 5, the K constant given in eq 6 is plotted against molarity. The value of K is very similar for the two complete systems analyzed: it presents a minimum for molarity between 1.5 and 5, and it increases for more concentrated solutions. Note that for diluted mixtures $x_w > 0.95$ (i.e., $c/\text{mol}\cdot\text{L}^{-1} < 1.5$) the calculated K is erroneous because ln η does not follow the linear behavior down that concentration, and so we have not included it. It is not possible analyze K quantitatively from its original physical meaning because measured mixtures are not those ideally used to develop Walden's rule,[24] and thus conclusions would be erroneous. However, the coincidence of the curves for the three compounds and its nearly constant value in a broad concentration region reveals that a physical origin for K can exist. This effect was observed also in [BMIM][BF_4] + water.[11]

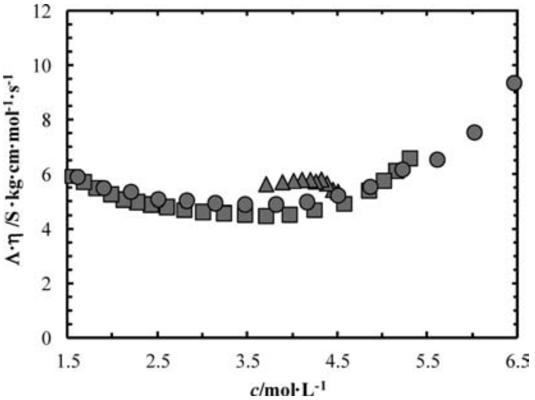

Figure 5. Product of molar conductivity by viscosity vs molarity for aqueous solutions of [EMIM][BF_4] (dot symbols), [BMIM][BF_4] (square symbols), and [HMIM][BF_4] (triangle symbols).

Conclusions

The two mean transport physical properties (dynamic viscosity and electrical conductivity) in aqueous mixtures of three common ionic liquids from the family 1-alkyl-3-methyl imidazolium tetrafluoroborate at four temperatures were measured. From those, viscosity deviations and molar conductivity were extracted. Data have been compared with available previous published ones with good agreement.

Walden's rule was used to relate molar conductivity and viscosity, for which the product must be constant. Results obtained are common for the three systems at the same composition, and the resulting product is constant for a broad range of concentrations but increases its value for the most IL concentrated mixtures.

Literature Cited

(1) Wasserscheid, P.; Welton, T. *Ionic Liquids in Synthesis*, 2nd ed.; Wiley-Verlag: Weinheim, 2008.

(2) Marsh, K. N.; Boxall, J. A.; Lichtenthaler, R. Room temperature ionic liquids and their mixtures-a review. *Fluid Phase Equilib.* **2004**, *219*, 93–98.

(3) Seddon, K. R.; Stark, A.; Torres, M. J. Influence of chloride, water, and organic solvents on the physical properties of ionic liquids. *Pure Appl. Chem.* **2000**, *72*, 2275−2287; ibid. Viscosity and density of 1-alkyl-3-methylimidazolium ionic liquids. *ACS Symp. Ser.* **2002**, *819*, 34−49.

(4) González, B.; Calvar, N.; Gómez, E.; Domínguez, A. Physical properties of the ternary system (etanol + wáter + 1-butyl-3methylimidazolium methylsulphate) and its binary mixtures at several temperatures. *J. Chem. Thermodyn.* **2008**, *40*, 1274–1281.

(5) Mokhtarani, B.; Sharifi, A.; Motaheb, H. R.; Mirzaei, M.; Mafi, M.; Sadeghian, F. Density and viscosity of pyridinium-based ionic liquids and their binary mixtures with water at several temperatures. *J. Chem. Thermodyn.* **2009**, *41*, 323–329.

(6) Gómez, E.; González, B.; Calvar, N.; Tojo, E.; Domínguez, A. Physical properties of pure 1-ethyl-3-methylimidazolium ethylsulphate and its binary mixtures with ethanol and water at several temperatures. *J. Chem. Eng. Data* **2006**, *51*, 2096–2102.

(7) Zafarani-Moattar, M. T.; Majdan-Cegincara, M. Viscosity, density, speed of sound and refractive index of binary mixtures of organic solvent + ionic liquid, 1-butyl-3-methylimidazolium hexafluorophosphate at 298.15 K. *J. Chem. Eng. Data* **2007**, *52*, 2359–2364.

(8) Zhang, S.; Li, X.; Chen, H.; Wang, J.; Zhang, J.; Zhang, M. Determination of physical properties for the binary system of 1-ethyl-3-methylimidazolium tetrafluoroborate + H_2O. *J. Chem. Eng. Data* **2004**, *49*, 760–764.

(9) Malham, I. B.; Turmine, M. Viscosities and refractive indices of binary mixtures of 1-butyl-3-methylimidazolium tetrafluoroborate and 1-butyl-2,3-dimethylimidazolium tetrafluoroborate with water at 298 K. *J. Chem. Thermodyn.* **2008**, *40*, 718–723.

(10) Zhou, Q.; Wang, L.-S.; Chen, H.-P. Densities and viscosities of 1-butyl-3-methylimidazolium tetrafluoroborate + H_2O binary mixtures from (303.15 to 353.15) K. *J. Chem. Eng. Data* **2006**, *51*, 905–908.

(11) Ries, L. A. S.; do Amaral, F. A.; Matos, K.; Martini, E. M. A.; de Souza, M. O.; de Souza, R. F. Evidence of change in the molecular organization of 1-n-butyl-3-methylimidazolium tetrafluoroborate ionic liquid solutions with the addition of water. *Polyhedron* **2008**, *27*, 3287–3293.

(12) Liu, W.; Zhao, T.; Zhang, Y.; Wang, H.; Yu, M. The physical properties of aqueous solutions of the ionic liquid [BMIM][BF4]. *J. Solution Chem.* **2006**, *35*, 1337–1346.

(13) Liu, W.; Cheng, L.; Zhang, Y.; Wang, H.; Yu, M. The physical properties of aqueous solutions of room-temperature ionic liquids based on imidazolium: Database and evaluation. *J. Mol. Liq.* **2008**, *140*, 68–72.

(14) Su, W. C.; Chou, C. H.; Wong, D. S. H.; Li, M. H. Diffusion coefficients and conductivities of alkyl-imidazolium tetrafluoroborates and hexafluorophosphates. *Fluid Phase Equilib.* **2007**, *252*, 74–78.

(15) Stoppa, A.; Hunger, J.; Buchner, R. Conductivities of binary mixtures of ionic liquids with polar solvents. *J. Chem. Eng. Data* **2009**, *54*, 472–479.

(16) Vila, J.; Ginés, P.; Rilo, E.; Cabeza, O.; Varela, L. M. Great increase of the electrical conductivity of ionic liquids in aqueous solutions. *Fluid Phase Equilib.* **2006**, *247*, 32–39.

(17) Rilo, E.; Pico, J.; García-Garabal, S.; Varela, L. M.; Cabeza, O. Density and Surface Tension in binary mixtures of CnMIM-BF_4 Ionic Liquids with water and ethanol. *Fluid Phase Equilib.* **2009**, *285*, 83–89.

(18) Prego, M.; Rilo, E.; Carballo, E.; Franjo, C.; Jiménez, E.; Cabeza, O. Electrical conductivity data of alkanols from 273 to 333 K. *J. Mol. Liq.* **2003**, *102*, 83–91.

(19) Cuadrado-Prado, S.; Domínguez-Pérez, M.; Rilo, E.; García-Garabal, S.; Segade, L.; Franjo, C.; Cabeza, O. Experimental measurement of the hygroscopic grade on eight imidazolium based ionic liquids. *Fluid Phase Equilib.* **2009**, *278*, 36–40.

(20) Sanmamed, Y. A.; González-Salgado, D.; Troncoso, J.; Cerdeirina, C. A.; Romani, L. Viscosity-induced errors in the density determination of room temperature ionic liquids using vibrating tube densitometry. *Fluid Phase Equilib.* **2007**, *252*, 96–102.

(21) Ahosseini, A.; Surto, A. M. Viscosity of imidazolium based ionic liquids at elevated pressures. *Int. J. Thermophys.* **2008**, *29*, 1222–1243.

(22) Cabeza, O.; Pico, J.; Domínguez-Pérez, M.; Rilo, E.; Freire, S.; Varela, L. M. Novel fitting equation for binary mixtures excess thermodynamic magnitudes. ESAT 08 Cannes 129−130 (2008).

(23) National Institute of Standards and Technology, “IUPAC Ionic Liquids Database, IL Thermo (NIST SRD #147)” (2008) http://ilthermo.boulder.nist.gov/ILThermo/mainmenu.uix.

(24) Bockris, J. O’M.; Reddy, A. K. N. *Modern Electrochemistry 1*; Plenum Press: New York, 1998; Ch. 4.

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