Temperature Dependence Measurements of the Density at 0.1 MPa for 1-Alkyl-3-methylimidazolium-Based Ionic Liquids with the Trifluoromethanesulfonate and Tetrafluoroborate Anion

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Experimental density data are reported for four 1-C_n-3-methylimidazolium-based ionic liquids ($n = 2, 4, 6$), three with trifluoromethanesulfonate and one with a tetrafluoroborate anion, at atmospheric pressure and at temperatures in the range of (268 to 356) K. The buoyancy method was employed, using the microbalance of the Krüss K100MK2 tensiometer. At each nominal temperature 40 evaluated buoyancy and temperature readings in two runs were taken in most cases. Averages of density and temperature values obtained at the individual nominal temperatures are presented with an estimated total standard uncertainty less than 0.3 $kg\cdot m^{-3}$ ($2.5\cdot 10^{-4}\ \rho$) and 0.02 K, respectively. The 83 new experimental data points on the density−temperature relation of the four ionic liquids of interest are means calculated from about 3000 individual density readings, which have been altogether taken in the present study. Measurements on two samples of the same stated high purity from different suppliers were performed for two ionic liquids, to demonstrate the influence of the sample origin.

Introduction

The density of ionic liquids, being a thermophysical property required in almost all of their applications, has been extensively studied. Nevertheless, despite of all of the efforts developed by various authors, experimental density data are often discrepant and poorly consistent with each other and in many cases are scarce or even absent.

The results of the most accurate density measurements for common solvents obtained by various authors are often consistent to each other within a standard relative uncertainty of 0.05 %. By contrast, even the best results of the density measurements on [EMIM][CF_3SO_3] (1-ethyl-3-methylimidazolium trifluoromethanesulfonate)[1−5] and [BMIM][CF_3SO_3] (1-butyl-3-methylimidazolium trifluoromethanesulfonate)[1,7,10] obtained by authors using a vibrating-tube densimeter with corrections for viscosity lie in a band of a width of (14 and 10) $kg\cdot m^{-3}$, respectively, which is 1 % and 0.8 % of the measured density value, while the declared standard uncertainties are most often about 0.4 $kg\cdot m^{-3}$ or less. Thus, the density data for pure ILs reported by different authors often do not overlap each other within their stated uncertainties. To establish a base for critical assessment of the reliability of different measurement methods, the data sets obtained by different authors, and different sources of samples, a sufficiently large body of experimental data must be accumulated, obtained by different authors using different methods.

The scatter of the data presented by different authors seems not to correlate markedly with any conceivable attribute of the measurements as described by their authors. There exist cases when values of density obtained by a certain author for several ionic liquids agree well with those of other authors with the exception of one ionic liquid.[4,22−24] In such a case a suspicion arises that the problem rests in that single sample, though of high stated purity, rather than in details of the used measuring procedure. That is why we included into the present density measurements a repeated measurement on [HMIM][BF_4] (1-hexyl-3-methylimidazolium tetrafluoroborate) performed on a sample provided by another supplier than the sample used in our previous study.[22] Similarly, density measurements for [BMIM][CF_3SO_3] were performed on two samples obtained from different suppliers.

The present study is a continuation of the previous work[22] devoted to measurements of the density of 1-alkyl-3-methylimidazolium-based ionic liquids with a tetrafluoroborate anion. Besides the measurements on samples of the same ionic liquid having a different origin, the aim of the present study was to obtain new reliable experimental data for the density of selected ILs using the buoyancy method. Three imidazolium-based ionic liquids with a hydrophilic trifluoromethanesulfonate anion and different length of the alkyl chain, that is, [C_n-mim][CF_3SO_3] with $n = 2$, 4, and 6, were selected. Table S1 in the Supporting Information gives an overview of the literature sources for experimental density data for the ionic liquids of interest. As far as we are aware, no density data are yet available for [HMIM][CF_3SO_3] (1-hexyl-3-methylimidazolium trifluoromethanesulfonate) in the open literature.

Experimental Section

In the present study, the density measurements were performed by the buoyancy method using the microbalance of a Krüss K100MK2 tensiometer having a resolution of 10 μg. Details concerning the apparatus, its calibration, and the measurement procedure are described elsewhere.[22] The sample vessel is placed in a vessel, which is thermostatted externally by a Julabo MB-5 thermostat providing a bath temperature stability of $\pm$ 0.02 K. At temperatures below 298 K, in addition, the external immersion cooler LAUDA ETK 30 is employed. In this way, the sample temperature stability of the order of magnitude of 0.01 K was achieved. The sample temperature is

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measured with a platinum resistance thermometer immersed into the sample. The standard uncertainty of temperature measurements stated by the manufacturer is 0.02 K. Values of the repeatability of the present density and temperature measurement expressed in terms of experimental standard deviation of the density values obtained within one experimental run are typically from (0.1 to 0.2) $kg \cdot m^{-3}$ and from (0.01 to 0.02) K respectively.

The samples of [EMIM][CF_3SO_3] and [HMIM][CF_3SO_3], both with the same specified minimum mass fraction purity of 0.99 and with the mass fraction water content of $w(H_2O) = 1 \cdot 10^{-4}$ and $w(H_2O) = 9.8 \cdot 10^{-5}$, respectively, were obtained from Ionic Liquids Technologies GmbH & Co. (IoLiTec, Denzlingen, Germany). Two samples of [BMIM][CF_3SO_3] with the specified minimum mass fraction purity of 0.99 were used in the reported measurements of which one was supplied by Merck KGaA (Darmstadt, Germany, $w(H_2O) = 2.8 \cdot 10^{-5}$) and the other one by Solvionic SA (Toulouse, France $w(H_2O) = 2.8 \cdot 10^{-5}$). The sample of [HMIM][$BF_4$] (mass fraction purity ≥ 0.99, $w(H_2O) = 5.1 \cdot 10^{-5}$) was obtained from Solvionic SA. The samples were used without further purification except for drying. The measurements were performed in 4 h periods, one period a day. After each 4 h measurement period, the samples were dried by intensive stirring and evaporation under vacuum at a temperature of 353 K for 4 h and than kept under vacuum overnight. The water content of the samples was measured in our laboratory with the coulometric Karl Fischer titrator Mettler Toledo C30.

The measurements were taken at the only one temperature during each 4 h measurement period. At a relative air moisture of (40 to 50) %, the water content of the samples increased from its initial value up to mass fractions from $6 \cdot 10^{-4}$ to $9 \cdot 10^{-4}$ during 2 h of measurements. No unidirectional drift in the density value attributable to the absorption of the air gases and moisture by the sample was observed during any particular measuring period.

Results and Discussion

The present study provides 83 new experimental data points on the density−temperature relation at a pressure of 0.1 MPa for four imidazolium-based ionic liquids, three with trifluoromethanesulfonate and one with a tetrafluoroborate anion. These new experimental density values are means calculated from about 3000 individual density readings, which have been altogether taken in the present study. For each IL sample, a total of about 500 to about 1000 individual buoyancy readings were taken at 13 to 18 temperature points between (268 and 356) K. In most cases, two to three buoyancy force and temperature measurement runs were performed at each thermostat temperature setting, each containing 20 evaluated individual readings as a rule. Table 1 gives averages of the values of temperature and density obtained at the individual nominal temperature points together with the estimate of their type A expanded uncertainty at the 95 % confidence level calculated from the standard deviation of the mean by applying the coverage factor $k = 2$. The last column of the table gives the number of averaged individual data readings. The standard deviations of the density from the fitting function eq 1 range from (0.14 to 0.3) $kg \cdot m^{-3}$ for the studied ILs. The data were fitted to a rational equation:

$$\rho(T) = \frac{\rho_0}{1 + a\tau} \tag{1}$$

where $\tau = T/100$. Table 2 gives coefficients ρ_0 and a of eq 1 used to correlate the experimental data together with the estimates of the uncertainty at the 95 % confidence level for the equation parameters and the root-mean-square deviation (rmsd) of the experimental density values from those calculated

Table 1. Mean Values of the Experimental Readings of Temperature, T, and Density, ρ, with their Expanded Uncertainties at the 95 % Confidence Level, $2u$, Evaluated from the Standard Deviation of the Mean by Applying Coverage Factor $k = 2$, and the Number of the Averaged Data Readings

$T \pm 2u$	$\rho \pm 2u$	
K	$kg \cdot m^{-3}$	no. of data
1-Ethyl-3-methylimidazolium Trifluoromethanesulfonate [EMIM][CF_3SO_3]		
268.45 ± 0.002	1411.49 ± 0.012	23
272.20 ± 0.014	1408.79 ± 0.030	29
276.54 ± 0.002	1404.33 ± 0.019	32
281.04 ± 0.002	1400.26 ± 0.025	39
287.65 ± 0.007	1394.08 ± 0.028	80
292.97 ± 0.002	1389.96 ± 0.016	31
297.94 ± 0.002	1385.93 ± 0.012	26
303.19 ± 0.002	1381.81 ± 0.007	40
308.26 ± 0.003	1377.30 ± 0.010	40
313.30 ± 0.002	1373.19 ± 0.015	40
318.37 ± 0.005	1368.79 ± 0.019	40
323.27 ± 0.002	1364.69 ± 0.017	40
328.15 ± 0.002	1360.37 ± 0.028	40
332.94 ± 0.001	1356.00 ± 0.041	40
338.05 ± 0.002	1352.58 ± 0.023	40
342.99 ± 0.004	1349.05 ± 0.018	40
348.10 ± 0.004	1344.79 ± 0.023	40
355.90 ± 0.003	1339.21 ± 0.016	40
1-Butyl-3-methylimidazolium Trifluoromethanesulfonate [BMIM][CF_3SO_3]		
Sample Supplied by Merck KGaA		
293.37 ± 0.003	1302.68 ± 0.054	41
298.22 ± 0.003	1298.85 ± 0.029	38
303.10 ± 0.002	1295.47 ± 0.014	40
308.06 ± 0.006	1290.59 ± 0.030	40
313.53 ± 0.003	1286.37 ± 0.021	40
318.46 ± 0.007	1282.60 ± 0.015	40
322.88 ± 0.003	1278.95 ± 0.010	40
328.12 ± 0.003	1275.38 ± 0.014	40
332.67 ± 0.002	1271.82 ± 0.007	40
337.79 ± 0.003	1267.55 ± 0.015	40
343.18 ± 0.002	1263.58 ± 0.030	40
348.16 ± 0.002	1259.93 ± 0.025	41
355.85 ± 0.004	1253.95 ± 0.021	40
Sample Supplied by Solvionic SA		
292.94 ± 0.004	1302.17 ± 0.058	40
298.09 ± 0.002	1298.67 ± 0.039	40
303.18 ± 0.003	1293.73 ± 0.007	40
308.19 ± 0.002	1289.35 ± 0.025	40
313.13 ± 0.002	1285.69 ± 0.024	40
318.20 ± 0.002	1281.27 ± 0.011	40
323.13 ± 0.004	1277.33 ± 0.009	40
328.08 ± 0.002	1272.83 ± 0.025	40
332.91 ± 0.002	1269.50 ± 0.028	40
337.95 ± 0.003	1264.90 ± 0.046	40
342.95 ± 0.002	1261.12 ± 0.017	40
1-Hexyl-3-methylimidazolium Trifluoromethanesulfonate [HMIM][CF_3SO_3]		
293.96 ± 0.002	1242.34 ± 0.023	48
298.12 ± 0.004	1239.33 ± 0.011	120
303.20 ± 0.002	1235.32 ± 0.017	60
308.26 ± 0.004	1231.38 ± 0.013	94
313.31 ± 0.003	1227.72 ± 0.017	105
318.32 ± 0.003	1223.93 ± 0.017	60
323.53 ± 0.005	1220.11 ± 0.019	40
328.05 ± 0.007	1216.35 ± 0.021	40
333.22 ± 0.006	1212.82 ± 0.016	40
337.77 ± 0.005	1209.27 ± 0.036	40
342.72 ± 0.005	1205.63 ± 0.024	40
347.48 ± 0.004	1202.25 ± 0.029	40
355.40 ± 0.003	1196.19 ± 0.025	40
1-Hexyl-3-methylimidazolium Tetrafluoroborate [HMIM][BF_4]		
268.65 ± 0.002	1166.57 ± 0.081	57
273.41 ± 0.003	1163.18 ± 0.097	40
277.88 ± 0.002	1160.54 ± 0.023	42
282.47 ± 0.002	1157.37 ± 0.028	30

Table 1. Continue

$T \pm 2u$	$\rho \pm 2u$	
K	$kg \cdot m^{-3}$	no. of data
288.15 ± 0.003	1153.14 ± 0.015	38
292.90 ± 0.002	1149.66 ± 0.027	57
298.06 ± 0.002	1145.96 ± 0.010	80
303.16 ± 0.006	1141.66 ± 0.056	100
308.22 ± 0.002	1139.01 ± 0.021	80
313.33 ± 0.002	1135.27 ± 0.011	80
318.48 ± 0.001	1131.84 ± 0.016	60
323.38 ± 0.003	1128.40 ± 0.016	60
328.20 ± 0.003	1125.24 ± 0.007	60
333.22 ± 0.005	1121.97 ± 0.024	60
337.77 ± 0.004	1118.96 ± 0.018	40
343.75 ± 0.002	1115.56 ± 0.019	40
348.42 ± 0.004	1111.52 ± 0.012	40
356.07 ± 0.004	1106.63 ± 0.032	40

Table 2. Coefficients ρ_0 and a of Equation 1, Their 95 % Confidence Interval Half-Widths, $2u(\rho_0)$ and $2u(a)$, and the Root-Mean-Square Deviation, rmsd, of the Experimental Data from Equation 1

	$\rho_0 \pm 2u(\rho_0)$		rmsd
ionic liquid	$kg \cdot m^{-3}$	$a \pm 2u(a)$	$kg \cdot m^{-3}$
[EMIM][CF_3SO_3]	1693.8 ± 3.0	$(7.455 \pm 0.07) \cdot 10^{-2}$	± 0.32
[BMIM][CF_3SO_3]	1593.6 ± 4.1	$(7.611 \pm 0.10) \cdot 10^{-2}$	± 0.24
[HMIM][CF_3SO_3]	1523.5 ± 2.3	$(7.692 \pm 0.06) \cdot 10^{-2}$	± 0.14
[HMIM][BF_4]	1400.3 ± 2.9	$(7.448 \pm 0.08) \cdot 10^{-2}$	± 0.30

from eq 1. Figures 1 to 4 show deviations of the experimental data points from eq 1.

Our present density values for [EMIM][CF_3SO_3] are closest to those obtained by García-Miaja et al.,[3] and they are approximately by about 4 $kg \cdot m^{-3}$ (i.e., by about 0.3 % relative) greater than the values obtained by other authors (Figure 1). The present experimental values of density itself, the densities calculated from eq 1, and also the density values obtained by other authors are depicted at Figure S1 in the Supporting Information. The results for [EMIM][CF_3SO_3] obtained in ref 4 rank among the solitary cases with a greater deviation from results of other authors mentioned in the Introduction, when the suspicion falls upon the sample.

Our results for [BMIM][CF_3SO_3] obtained with the sample provided by Merck agree within ± 0.1 % with the vibrating-tube densimeter results by García-Miaja et al.,[3] Tokuda et al.,[7]

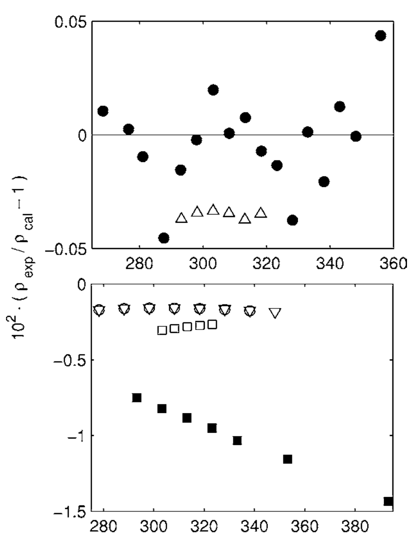

Figure 1. [EMIM][CF_3SO_3]: deviations of experimental density data, ρ_{exp}, of different authors from the values and ρ_{cal}, calculated from the eq 1 as a function of temperature. ▽, Rodríguez and Brennecke;[1] ○, Vercher et al.;[2] △, García-Miaja et al.;[3] ■, Gardas et al.;[4] □, Wong et al.;[5] ●, this work.

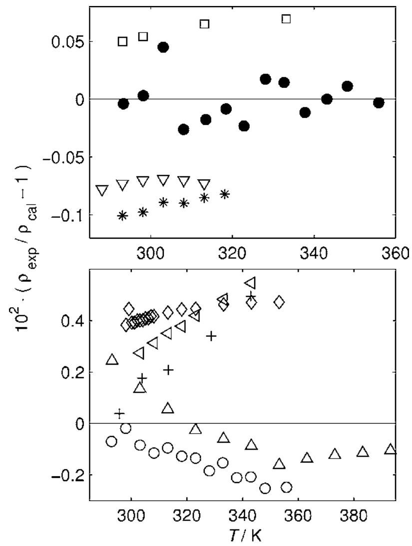

Figure 2. [BMIM][CF_3SO_3]: deviations of experimental density data, ρ_{exp}, of different authors from the values ρ_{cal}, calculated from the eq 1 as a function of temperature. ∗, García-Miaja et al.;[3] +, Fredlake et al.;[6] ▽, Tokuda et al.;[7] △, Gardas et al.;[8] left-pointing triangle, Ge et al.;[9] □, Tariq et al.;[10] ◆, Soriano et al.;[11] ●, this work (the sample supplied by Merck KGaA); ○, this work (the sample supplied by Solvionic SA).

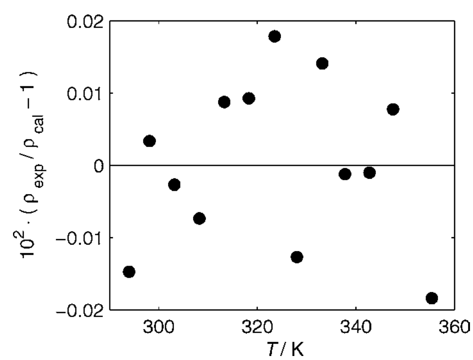

Figure 3. [HMIM][CF_3SO_3]: deviations of experimental density data, ρ_{exp}, from the values, and ρ_{cal}, calculated from the eq 1 as a function of temperature. ●, this work.

and Tariq et al.,[10] evaluated with the account of the correction for viscosity (Figure 2). The temperature dependence of the [BMIM][CF_3SO_3] density obtained by other authors differs from that obtained by us relatively by up to 0.5 % (6 $kg \cdot m^{-3}$)[11] in value and also in slope. The temperature dependence of the density obtained in the present study for the second sample of [BMIM][CF_3SO_3] supplied by Solvionic differs from that for the sample supplied by Merck by up to 3 $kg \cdot m^{-3}$ and has also a slightly different slope. The results obtained for the two samples of the [BMIM][CF_3SO_3] and of the [BMIM][BF_4] suggest that the observed differences between density values obtained by different authors might stem from the differences between samples, though they are declared to be of high purity. The samples should be quantitatively analyzed.

As far as we are aware, no experimental data on the density have been published yet for [HMIM][CF_3SO_3] in open literature. Deviations of the present data on the [HMIM][CF_3SO_3] density from eq 1 fitted to them are depicted in Figure 3.

The deviation plot in Figure 4 shows that the present data on [HMIM][BF_4] density for the sample supplied by Merck agrees with the data by most other authors within a band width of 0.1 %. Our density data obtained previously[22] using the same apparatus and experimental procedure for the sample of [HMIM][BF_4] supplied by IoLiTec are shifted relative to the others by about

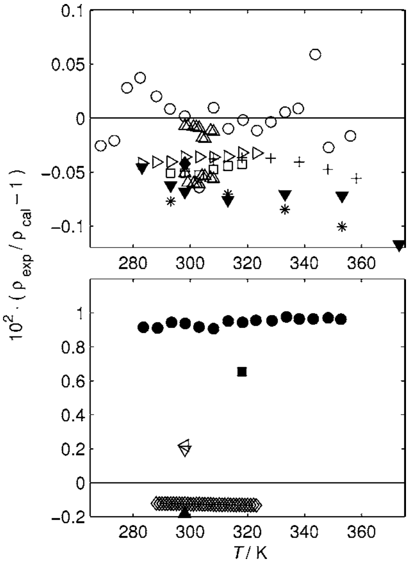

Figure 4. [HMIM][BF_4]: deviations of experimental density data, ρ_{exp}, of different authors from the values, and ρ_{cal}, calculated from the eq 1 as a function of temperature. Left-pointing triangle, Letcher and Reddy;[12] ■, Wagner et al.;[13] ▲, Wagner et al.;[14] ▽, Letcher and Reddy;[15] ◆, Sanmamed et al.;[16] △, Navia et al.;[17] +, Muhammad et al.;[18] □, García-Miaja et al.;[19] ∗, Taguchi et al.;[20] ◆, Rilo et al.;[21] ●, Klomfar et al.;[22] right-pointing triangle, Sanmamed et al.;[25] ▼, Safarov and Hassel;[26] ○, this work.

1 % of the measured density value. Figure S2 in the Supporting Information depicts experimental densities obtained for [HMIM]-[BF_4] by different authors and the temperature dependence of density calculated from eq 1. For illustration, similar information for [EMIM][BF_4] and [BMIM][BF_4] from ref 22 is included in Figure S2 presented in a somewhat briefer form.

Supporting Information Available:

Literature sources for experimental density data for the ionic liquids of interest with the sample water content given as water mass fraction, *w* (Table S1). Temperature dependence of the density at 0.1 MPa for the imidazolium-based ionic liquids of interest. A comparison of the experimental density data obtained by various authors, including the present work for the trifluoromethanesulfonates (Figure S1) and for the tetrafluoroborate (Figure S2). This material is available free of charge via the Internet at http://pubs.acs.org.

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