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SHORT ARTICLE

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Volumetric Properties for (Ionic Liquid + Methanol or Ethanol or 1-Propanol + Nitromethane) at 298.15 K and Atmospheric Pressure

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ABSTRACT: The densities of the ternary systems {1-butyl-3-methylimidazolium methyl sulfate ($[BMIM]^+[MeSO_4]^-$) + methanol or ethanol or 1-propanol + nitromethane} were measured at the temperature 298.15 K, and the ternary excess molar volumes, V^E_{123}, were calculated . The V^E_{123} values are negative for all mole fractions of the ionic liquid (IL) and increase with increasing mole fraction of IL. The Cibulka equation was used to correlate the ternary excess molar volumes using the Redlich−Kister parameters for the binary systems obtained from the literature. There is a good correlation between the Cibulka equation and the experimental V^E_{123} data.

INTRODUCTION

There is a lack of data on ternary excess molar volumes, V^E_{123}, for ionic liquid (IL) multicomponent systems.[1−8] Gómez et al.[1] determined V^E_{123} for the IL system (ethanol + water + 1,3-dimethylimidazolium methyl sulfate) at several temperatures. Deenadayalu et al.[2] used graph theory to correlate V^E_{123} for (1-ethyl-3-methylimidazolium diethylene glycol monomethyl ether sulfate + methanol + water) and calculated V^E_{123} for ($[MOA]^+$-$[Tf_2N]^-$ + methanol or ethanol + methyl acetate or ethyl acetate)[3,4] at the temperatures (298.15, 303.15, and 313.15) K.

González et al.[6] studied V^E_{123} for the (ethanol + water +1-butyl-3-methylimidazolium methyl sulfate) ternary system at several temperatures. Andreatta et al.[7,8] studied V^E_{123} for the systems [methyl acetate + methanol +1-octyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide] and [ethyl acetate + ethanol + 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide] at the temperature 298.15 K and atmospheric pressure.

In this work, values of V^E_{123} for the ternary IL systems ($[BMIM]^+[MeSO_4]^-$ + methanol or ethanol or 1- propanol + nitromethane) over the entire composition range were calculated from the experimental densities at the temperature 298.15 K and atmospheric pressure. The IL 1-butyl-3-methylimidazolium methyl sulfate ($[BMIM]^+[MeSO_4]^-$) was used. This work is a continuation of our research group's work on thermodynamic properties of ILs.[2−5,9−14]

EXPERIMENTAL SECTION

The solvents were purchased from Aldrich, Merck, and Fluka with mass fraction purity > 0.990 for methanol, ethanol, 1-propanol, and nitromethane and mass fraction purity > 0.970 for the IL. The IL, methanol, ethanol, 1-propanol, and nitromethane were used without any further purification. The water content in the IL was determined using a Karl Fischer coulometer (Metrohm model 797), and the mass fraction water content was found to be < 0.0024. The purity of the solvents was assessed by a comparison of the experimental density value with literature density values where available.[9,15]

The mass balance had a precision of 0.0001 g. The uncertainty in the mole fraction was estimated to be 0.0006.

The ternary mixtures were prepared to obtain a constant ratio of $z = x_3/x_1$, where x_1 is the mole fraction of IL and x_3 is the mole fraction of nitromethane, while varying the mole fraction of methanol or ethanol or 1-propanol (x_2). The densities were measured with an Anton Paar DMA 38 vibrating U-tube densimeter. Ultrapure water supplied by SH Calibration Service GmbH Graz and dried air was used for the calibration of the densimeter at the temperature 298.15 K.[9] The temperature maintenance and control were regulated by a built-in thermostat controller with a temperature uncertainty of ± 0.01 K, and the densimeter was capable of measuring density to ± 0.0001 $g \cdot cm^{-3}$. The uncertainty in the density for the ternary system was 0.002 $g \cdot cm^{-3}$. The experimental uncertainty in V^E_{123} was ± 0.1 $cm^3 \cdot mol^{-1}$. The literature and experimental densities (ρ) are given in Table 1. The structure of the IL is given in Figure 1.

RESULTS AND DISCUSSION

In Tables 2 to 4, the densities, ρ, and the excess molar volumes for the ternary systems, V^E_{123}, are given as functions of composition at the temperature 298.15 K. The V^E_{123} values were calculated from the experimental density values using the following equation:

$$V^E_{123} = \frac{x_1M_1 + x_2M_2 + x_3M_3}{\rho} - \frac{x_1M_1}{\rho_1} - \frac{x_2M_2}{\rho_2} - \frac{x_3M_3}{\rho_3} \quad (1)$$

where x_1, x_2, and x_3 are the mole fractions, M_1, M_2, and M_3 the molecular masses, and ρ_1, ρ_2, and ρ_3 the densities of the pure components IL (1), methanol or ethanol or 1-propanol (2), and

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Table 1. Literature and Experimental Densities of the Pure Compounds at T = 298.15 K

		ρ/g·cm^{-3}	
compound	mass fraction	lit	exptl
$[BMIM]^+[MeSO_4]^-$	0.970	1.20775[a]	1.2080
methanol	0.998	0.78637[b]	0.7863
ethanol	0.998	0.7855[b]	0.7864
1-propanol	0.995	0.79960[b]	0.7976
nitromethane	0.990	1.13117[a]	1.1309

[a] Data from ref 15. [b] Data from ref 9.

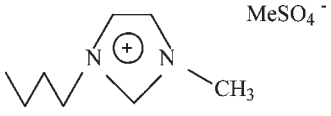

Figure 1. 1-Butyl-3-methylimidazolium methyl sulfate, $[BMIM]^+$-$[MeSO_4]^-$.

Table 2. Densities and Excess Molar Volumes for $[BMIM]^+[MeSO_4]^-$ (1) + Methanol (2) + Nitromethane (3) at T = 298.15 K

		ρ	V^E_{123}
x_1	x_2	g·cm^{-3}	cm^3·mol^{-1}
0.0485	0.7471	0.959	−0.9
0.0903	0.5290	1.048	−0.9
0.1099	0.4269	1.081	−0.9
0.1452	0.2429	1.133	−1.1
0.1771	0.0765	1.164	−0.8
0.1332	0.6173	1.058	−1.6
0.1998	0.4256	1.115	−1.4
0.2390	0.3114	1.138	−1.1
0.2971	0.1467	1.168	−0.8
0.0603	0.8974	0.918	−0.8
0.1591	0.7290	1.042	−1.5
0.2494	0.5752	1.101	−1.5
0.3119	0.4692	1.129	−1.3
0.3705	0.3690	1.149	−1.1
0.4720	0.1965	1.175	−0.6
0.5271	0.1026	1.187	−0.4
0.3706	0.5428	1.129	−1.3
0.4692	0.4211	1.154	−1.1
0.5494	0.3222	1.170	−0.8
0.6151	0.2409	1.180	−0.6
0.7242	0.1053	1.194	−0.2
0.8468	0.1150	1.197	−0.1
0.7200	0.2475	1.184	−0.4

nitromethane (3), respectively, and ρ is the density of the mixture.

Binary excess molar volumes, V^E_m, for (IL + methanol),[11] (IL + ethanol),[11] (IL + 1-propanol),[11] (IL + nitromethane),[15] (methanol + nitromethane),[16] (ethanol + nitromethane),[16] and (1-propanol + nitromethane)[17] have been published in the literature. The binary V^E_m data are relatively small and do not vary greatly with temperature. The Redlich−Kister equation[18] was fitted to the binary literature data to obtain the Redlich−Kister parameters at the temperature 298.15 K. The binary parameters are given in Table 5. The Redlich−Kister equation is given below:

$$V^E_m = x_1x_2\sum_{i=0}^{N} A_i(x_1 - x_2)^i \qquad (2)$$

The ternary excess molar volumes were correlated using the Cibulka equation,[19] which is given below:

$$V^E_{123} = \sum_{i,j=1,2;1,3;2,3} V^E_{ij}(x_i, x_j) - x_1x_2x_3(b_0 - b_1x_1 - b_2x_2) \qquad (3)$$

The Cibulka equation contains three parameters (b_0, b_1, and b_2) corrected over the three binary contributions. The values of b_0, b_1, and b_2 were adjusted to the experimental ternary excess molar volumes and detailed in an earlier paper.[4] The smoothing coefficients and the relative mean standard deviation (rmsd) values, σ, are given in Table 6. Ternary graphs obtained using the Cibulka equation parameters are given in Figures 2 to 4.

For the binary systems (methanol or ethanol or 1-propanol + nitromethane) at the temperature 298.15 K, at all mole fractions of alkanol, V^E_m is negative because of the geometrical fitting of the nitromethane into the remaining alkanol structure.[11] The V^E_m values for (1-propanol + nitromethane) are greater than those

Table 3. Densities and Excess Molar Volumes for $[BMIM]^+[MeSO_4]^-$ (1) + Ethanol (2) + Nitromethane (3) at T = 298.15 K

		ρ	V^E_{123}
x_1	x_2	g·cm^{-3}	cm^3·mol^{-1}
0.0628	0.6726	0.954	−0.8
0.1076	0.4385	1.046	−0.9
0.1263	0.3412	1.080	−1.0
0.1568	0.1824	1.128	−0.8
0.1813	0.0545	1.162	−0.7
0.1640	0.5288	1.050	−1.4
0.2297	0.3403	1.109	−1.1
0.2898	0.1676	1.153	−0.8
0.3109	0.1070	1.166	−0.7
0.0829	0.8588	0.914	−0.8
0.2045	0.6517	1.035	−1.3
0.3024	0.4850	1.096	−1.3
0.3638	0.3807	1.125	−1.1
0.4175	0.2891	1.147	−0.9
0.5020	0.1454	1.175	−0.6
0.2942	0.6370	1.064	−1.3
0.4439	0.4523	1.125	−1.2
0.5383	0.3359	1.151	−0.9
0.6637	0.1808	1.178	−0.4
0.7482	0.0757	1.194	−0.2
0.8775	0.0829	1.197	−0.1
0.7787	0.1862	1.182	−0.2
0.6796	0.2898	1.166	−0.5

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Table 4. Densities and Excess Molar Volumes for $[BMIM]^+[MeSO_4]^-$ (1) + 1-Propanol (2) + Nitromethane (3) at T = 298.15 K

		ρ	V^E_{123}
x_1	x_2	g·cm^{-3}	cm^3·mol^{-1}
0.0979	0.4893	1.004	−0.5
0.1372	0.2842	1.079	−0.6
0.1637	0.1460	1.126	−0.5
0.1836	0.0423	1.160	−0.5
0.0745	0.6116	0.957	−0.4
0.1199	0.3745	1.046	−0.6
0.0539	0.7187	0.915	−0.4
0.0350	0.8172	0.875	−0.2
0.1871	0.4625	1.049	−0.9
0.2495	0.2834	1.108	−0.8
0.2803	0.1947	1.134	−0.6
0.3188	0.0841	1.166	−0.5
0.1454	0.5823	1.005	−1.0
0.1639	0.7208	0.977	−0.8
0.1037	0.8234	0.921	−0.6
0.2412	0.5892	1.036	−0.9
0.3410	0.4193	1.095	−0.8
0.3993	0.3203	1.124	−0.7
0.4477	0.2377	1.146	−0.6
0.5197	0.1153	1.175	−0.4
0.3456	0.5736	1.064	−0.8
0.4964	0.3876	1.124	−0.7
0.5841	0.2794	1.151	−0.6
0.6930	0.1447	1.179	−0.4
0.8948	0.0648	1.197	−0.0
0.8141	0.1492	1.183	−0.1
0.7289	0.2382	1.167	−0.3

for the (ethanol + nitromethane) > (methanol + nitromethane) systems because of the increase in alcohol chain length.[11]

For the binary systems (IL + methanol or ethanol or 1-propanol) at T = 298.15 K, the V^E_m values are negative for all mole fractions of the IL because of the more efficient packing and/or attractive interaction that occurs when the IL and the alkanols are mixed.[11] The excess molar volumes increase in the order (IL + methanol) < (IL + ethanol) < (IL + 1-propanol) because of the increase in alcohol chain length.[11]

For the system (IL + nitromethane), V^E_m is negative at T = 298.15 K for all mole fractions of the IL as a result of packing effects.[15]

The binary V^E_m data for (methanol + nitromethane),[16] (IL + methanol),[11] and (IL + nitromethane)[15] at the temperature 298.15 K are all negative because of the geometrical fitting of the nitromethane molecules into the methanol structure for the first binary system and a more efficient packing and/or attractive interaction between the IL and the methanol or nitromethane molecules for the latter two binary systems. The negative values of V^E_{123} for (IL + methanol + nitromethane) show that the ion−dipole interactions and packing effects among methanol, nitromethane, and the IL dominate over the dissociation of the intermolecular hydrogen bonds in methanol and nitromethane.[3] At high mole fractions of methanol or IL and low mole fractions of nitromethane, the V^E_{123} values are positive, indicating that as the nitromethane mole fraction increases, the minimum of the V^E_{123} value shifts from the binary (IL + nitromethane) system into the middle of the ternary concentration.

The binary V^E_m data for (ethanol + nitromethane)[16] at the temperature 298.15 K exhibit both positive and negative values because of the breakup of the ethanol structure and geometrical fitting of the nitromethane into the ethanol structure, respectively.[16] The binary V^E_m data for (IL + ethanol) at the temperature 298.15 K is negative at all mole fractions of the IL. The ternary V^E_{123} values for (IL + ethanol + nitromethane) are similar to those for (IL + methanol + nitromethane).

The binary V^E_m data for (IL + 1-propanol) at the temperature 298.15 K are negative for all mole fractions of IL as a result of the same effects that occur in the (IL + methanol) binary system. For the binary system of (1-propanol + nitromethane) at the temperature 298.15 K and at all mole fractions of 1-propanol, V^E_m is positive because of the rupture of hydrogen bonds and dispersive interactions between unlike molecules.[17] The binary excess

Table 5. Redlich−Kister Parameters for the Seven Binary Systems at T = 298.15 K

					σ
A_0	A_1	A_2	A_3	A_4	cm^3·mol^{-1}
$[BMIM]^+[MeSO_4]^-$ + Methanol[a]					
4.13	−2.63	0.16	−	−	0.01
$[BMIM]^+[MeSO_4]^-$ + Ethanol[a]					
2.50	−0.93	−0.08	−	−	0.009
$[BMIM]^+[MeSO_4]^-$ + 1-Propanol[a]					
0.95	−0.19	−	−	−	0.009
$[BMIM]^+[MeSO_4]^-$ + Nitromethane[b]					
−1.92	1.34	−0.47	1.58	−1.86	0.01
Methanol + Nitromethane[c]					
−0.70	−0.40	−0.29	−	−	0.003
Ethanol + Nitromethane[c]					
0.05	−0.57	−0.35	−0.59	−	0.005
1-Propanol + Nitromethane[d]					
0.94	−0.26	0.15	−0.41	−	0.001

[a] Experimental data from ref 11. [b] Experimental data from ref 15. [c] Experimental data from ref 16. [d] Experimental data from ref 17.

Table 6. Smoothing Coefficients b_n and Relative Mean Standard Deviations σ for the Cibulka Equation at T = 298.15 K

			σ
b_0	b_1	b_2	cm^3·mol^{-1}
$[BMIM]^+[MeSO_4]^-$ (1) + Methanol (3) + Nitromethane (3)			
35.60	−52.47	− 185.50	0.74
$[BMIM]^+[MeSO_4]^-$ (1) + Ethanol (2) + Nitromethane (3)			
14.68	−29.73	− 133.66	0.74
$[BMIM]^+[MeSO_4]^-$ (1) + 1-Propanol (2) + Nitromethane (3)			
5.86	−9.83	− 86.03	0.31

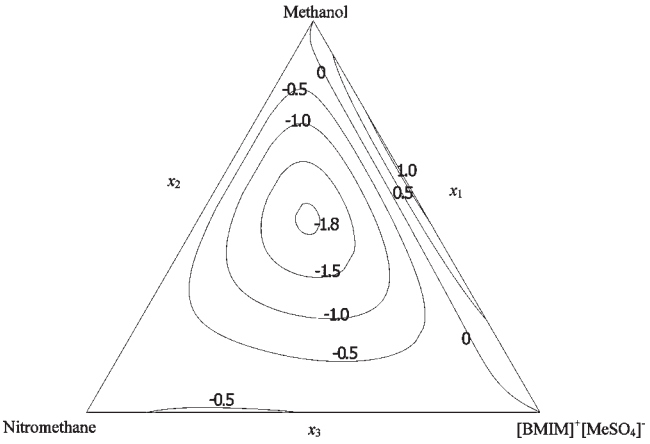

Figure 2. Graph of the excess molar volume, V^E_{123}, of the ternary mixture $[BMIM]^+[MeSO_4]^-$ (1) + methanol (2) + nitromethane (3) at 298.15 K. Solid lines were calculated with the Cibulka equation using the values of the parameters fitted to the binary and ternary data.

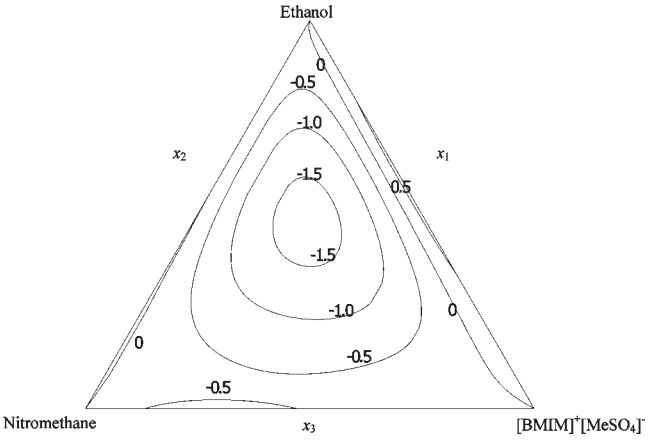

Figure 3. Graph of the excess molar volume, V^E_{123}, of the ternary mixture $[BMIM]^+[MeSO_4]^-$ (1) + ethanol (2) + nitromethane (3) at T = 298.15 K. Solid lines were calculated with the Cibulka equation using the values of the parameters fitted to the binary and ternary data.

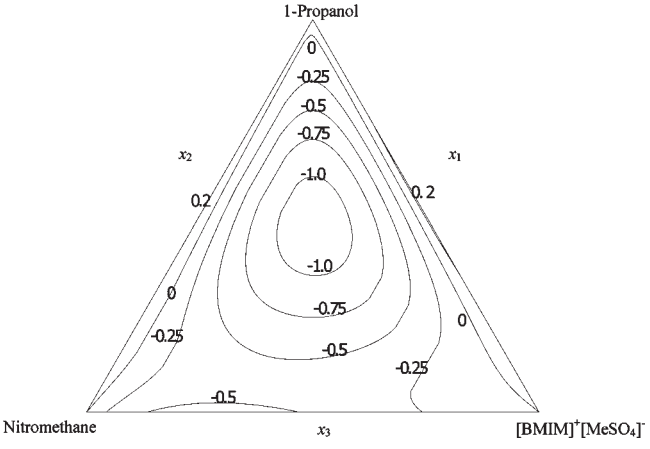

Figure 4. Graph of the excess molar volume, V^E_{123}, of the ternary mixture $[BMIM]^+[MeSO_4]^-$ (1) + 1-propanol (2) + nitromethane (3) at T = 298.15 K. Solid lines were calculated with the Cibulka equation using the values of the parameters fitted to the binary and ternary data.

molar volumes increase in the order (methanol + nitromethane) < (ethanol + nitromethane) < (1-propanol + nitromethane) systems because of the increase in alcohol chain length, which causes a decrease in attractive interaction between the two types of molecules. The negative values of V^E_{123} for (IL + 1-propanol + nitromethane) are due to the same effects has explained previously for the (IL + methanol or ethanol + nitromethane) ternary systems. V^E_{123} increases for the ternary systems in the order (IL + methanol + nitromethane) < (IL + ethanol + nitromethane) < (IL + 1-propanol + nitromethane).

The Cibulka equation correlations indicate that the ternary contribution (three-body effect) is dominant. The molecules of IL, methanol or ethanol or 1-propanol, and nitromethane are engaged in specific interactions, and the ability of methanol or ethanol or 1-propanol to form hydrogen bonds with the IL is increased by the presence of the nitromethane molecules.

CONCLUSION

In this paper, the densities and V^E_{123} have been reported for the systems (IL + methanol or ethanol or 1-propanol + nitromethane) at T = 298.15 K. The V^E_{123} values are negative for all compositions of the IL and show that the ion−dipole interactions as well as the packing effects among the alcohol, nitromethane, and the IL dominate over the dissociation of intermolecular hydrogen bonds in the alcohol and nitromethane. The molecules of IL, methanol or ethanol or 1-propanol, and nitromethane are engaged in specific interactions, and the ability of methanol or ethanol or 1-propanol to form hydrogen bonds with the IL is increased by the presence of nitromethane molecules. The Cibulka equation was used to correlate the ternary data and indicated that the ternary contribution to V^E_{123} is dominant.

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