Densities and Viscosities of Pure 1-Methyl-3-octylimidazolium Nitrate and Its Binary Mixtures with Alcohols at Several Temperatures

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This paper reports measurements of densities and viscosities for the binary system of 1-methyl-3-octylimidazolium nitrate and different alcohols in the temperature range of (283.15 to 333.15) K. The alcohols are ethanol, 1-propanol, and 1-butanol. The excess molar volume and viscosity deviation are calculated for the above systems, and the Redlich−Kister polynomial equation is used to fit the excess molar volume and viscosity deviation. The excess molar volume and viscosity deviation have negative deviations from ideal solution. The excess molar volumes increase with increasing the alcohol chain length.

Introduction

In recent years, there has been an increasing interest to study the physical properties of ionic liquids (ILs).[1] ILs show interesting properties such as negligible vapor pressure, nonflammability, chemical stability at high temperatures, and excellent solubility in organic and inorganic compounds. The properties of the ILs can be adjusted by altering the cation or anion, the chain length of the alkyl rests, and the substituent. Therefore, up to 10^{18} possible ILs are theoretically designable.[1] It is then possible to design a suitable IL for a specific process by selection of a certain cation or anion.

The applications of these new solvents in industry are very limited because of the lack of their physicochemical information either in pure forms or as binaries with organic or inorganic compounds.

1-Methyl-3-octylimidazolium nitrate [(Omim)(NO_3)] is a halogen-free IL and thus more environmentally friendly than the halogenated ILs.[2] There are limited data on the physical properties of (Omim)(NO_3) in literature. Seddon et al. reported the viscosities of pure (Omim)(NO_3) at different temperatures.[3] They observed that the presence of the chloride ion has a large effect on the physical properties of this IL.[4] The presence of water or alcohol in ILs has a large effect on the physical properties of the IL.[5,6] The measurement of the density and viscosity of different binaries of ILs and alcohols have been investigated by some researchers. Gonzalez et al.[7] determined the densities and viscosities of 1-methyl-3-octylimidazolium chloride [(Omim)Cl] binaries with methanol, ethanol, and 1-propanol. Sibiya and Deenadayalu[8] have reported the excess molar volume and isentropic compressibility of trioctylmethylammonium bistrifluoromethylsulfonylimide binaries with methanol, ethanol, and 1-propanol.

In the present study, as a continuance of our previous studies on the physical properties of ILs,[9−11] experimental data for density and viscosity of (Omim)(NO_3) at different temperatures are presented. The new experimental data for density and viscosity of (Omim)(NO_3) and alcohols (ethanol, propanol, or butanol) have been measured at different temperatures. To our knowledge, the experimental data for the binaries of this IL and alcohol have not been reported in literature. The excess molar volume and viscosity deviations are calculated, and the Redlich−Kister polynomial equation is used to correlate these data.[12]

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Experimental Section

(Omim)(NO_3) was synthesized in our laboratory from (Omim)Cl according to the procedure in literature.[13] (Omim)Cl was prepared according to the method described in literature.[14]

In this procedure, $NaNO_3$ (51 g, 0.6 mol) is added to a solution of (Omim)Cl (115.4 g, 0.5 mol) in dichloromethane and stirred for 24 h at the room temperature. The suspension is

Table 1. Density ρ and Viscosity η of Pure (Omim)(NO_3) at Several Temperatures

T/K	ρ/g·cm^{-3}	η/mPa·s
283.15	1.0740	2231.3
288.15	1.0705	1435.9
293.15	1.0673	954.6
298.15	1.0642	664.1
303.15	1.0611	466.0
308.15	1.0580	337.4
313.15	1.0549	249.7
318.15	1.0519	189.3
323.15	1.0489	147.1
328.15	1.0459	115.1
333.15	1.0429	88.4
338.15	1.0418	69.6
343.15	1.0388	56.8

Table 2. Comparison of Experimental Viscosities η with the Values Reported by Seddon et al.[6] for Pure (Omim)(NO_3) at Different Temperatures

T/K	η/(mPa·s)		
	experimental	literature	100 Δ
283.15	2231.3	2918	23.53
293.15	954.6	1238	22.89
303.15	466.0	563	17.23
313.15	249.7	288	13.29
323.15	147.1	159	7.47
333.15	88.4	95.80	7.69
343.15	56.8	62.00	8.26

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Table 3. Density ρ, Viscosity η, Excess Molar Volume V^E, and Viscosity Deviation $\Delta\eta$ of the Binary System of Ethanol (1) + (Omim)(NO_3) (2) at Several Temperatures

x_1	ρ / g·cm^{-3}	η / mPa·s	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s	ρ / g·cm^{-3}	η / mPa·s	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s
		T = 283.15 K				T = 288.15 K		
0.128	1.0651	969.7	−0.108	−976.3	1.0616	659.1	−0.119	−593.2
0.266	1.0530	375.5	−0.232	−1262.7	1.0496	265.9	−0.255	−788.5
0.357	1.0430	215.6	−0.283	−1219.7	1.0396	162.1	−0.317	−761.7
0.455	1.0300	101.8	−0.358	−1114.9	1.0267	76.2	−0.415	−707.0
0.554	1.0140	48.2	−0.506	−947.8	1.0107	38.6	−0.571	−602.5
0.662	0.9916	24.8	−0.756	−730.3	0.9883	20.6	−0.829	−465.6
0.767	0.9613	11.6	−1.003	−509.4	0.9579	9.9	−1.073	−325.7
0.868	0.9157	5.1	−1.001	−290.7	0.9120	4.5	−1.042	−186.2
		T = 293.15 K				T = 298.15 K		
0.128	1.0584	464.3	−0.123	−368.3	1.0553	333.2	−0.130	−246.0
0.266	1.0464	198.3	−0.276	−502.7	1.0433	150.3	−0.297	−337.4
0.357	1.0365	124.5	−0.358	−489.8	1.0334	97.1	−0.378	−330.3
0.455	1.0235	59.7	−0.463	−461.1	1.0204	47.5	−0.483	−314.9
0.554	1.0075	31.5	−0.625	−394.9	1.0043	26.1	−0.641	−270.7
0.662	0.9850	17.2	−0.884	−306.2	0.9818	14.5	−0.904	−210.6
0.767	0.9544	8.6	−1.110	−214.7	0.9512	7.5	−1.144	−148.0
0.868	0.9081	4.0	−1.066	−123.0	0.9048	3.5	−1.093	−85.1
		T = 303.15 K				T = 308.15 K		
0.128	1.0521	246.4	−0.131	−160.1	1.0489	185.1	−0.137	−109.3
0.266	1.0401	116.4	−0.316	−225.9	1.0368	89.0	−0.322	−158.9
0.357	1.0300	73.4	−0.402	−226.6	1.0267	58.4	−0.411	−158.9
0.455	1.0169	38.3	−0.508	−216.1	1.0136	31.1	−0.522	−153.2
0.554	1.0006	21.8	−0.659	−186.6	0.9972	18.4	−0.680	−132.6
0.662	0.9779	12.4	−0.918	−145.7	0.9743	10.8	−0.929	−103.8
0.767	0.9469	6.5	−1.152	−102.9	0.9431	5.7	−1.158	−73.6
0.868	0.9003	3.1	−1.114	−59.2	0.8963	2.8	−1.118	−42.5
		T = 313.15 K				T = 318.15 K		
0.128	1.0458	143.0	−0.138	−74.9	1.0427	112.2	−0.165	−53.1
0.266	1.0337	69.0	−0.333	−114.6	1.0306	55.9	−0.365	−83.3
0.357	1.0236	48.3	−0.437	−112.6	1.0203	39.8	−0.453	−82.2
0.455	1.0105	25.8	−0.557	−110.6	1.0071	21.8	−0.569	−81.8
0.554	0.9940	15.7	−0.714	−96.1	0.9904	13.5	−0.720	−71.3
0.662	0.9708	9.5	−0.949	−75.4	0.9671	8.7	−0.966	−55.8
0.767	0.9393	5.0	−1.154	−53.8	0.9354	4.5	−1.177	−40.2
0.868	0.8920	2.5	−1.083	−31.1	0.8880	2.3	−1.115	−23.3
		T = 323.15 K				T = 333.15 K		
0.128	1.0397	81.4	−0.167	−47.0	1.0336	53.6	−0.182	−23.6
0.266	1.0274	45.8	−0.373	−62.3	1.0213	32.0	−0.408	−33.0
0.357	1.0172	33.2	−0.469	−61.6	1.0110	23.9	−0.520	−33.2
0.455	1.0038	18.6	−0.591	−61.9	0.9974	14.1	−0.637	−34.4
0.554	0.9871	11.8	−0.748	−54.2	0.9804	9.3	−0.788	−30.5
0.662	0.9637	7.8	−0.999	−42.3	0.9568	6.2	−1.040	−24.1
0.767	0.9318	4.0	−1.201	−30.8	0.9244	3.3	−1.240	−17.8
0.868	0.8839	2.1	−1.124	−17.9	0.8761	1.8	−1.159	−10.4

filtered to remove the NaCl salt. The organic phase is then washed several times with a small volume of water until no further AgCl is precipitated in the aqueous phase when a concentrated $AgNO_3$ solution is added. The solvent is removed under vacuum, and the resulting IL is stirred with activated charcoal for 12 h. Removing the solvent gives 115.7 g of (Omim)(NO_3) with a 90 % yield.

The structure of the synthesized (Omim)(NO_3) was checked with NMR spectroscopy.[15] ^{1}H NMR (500 MHz, $CDCl_3$): δ/ppm 0.81 (3H, J = 6.8 Hz, t), 1.19 to 1.26 (10H, m), 1.83 (2H, m), 3.96 (3H, s), 4.18 (2H, J = 7.4 Hz, t), 7.26 (1H, s), 7.49 (1H, s), 9.76 (1H, s). ^{13}C NMR (125 MHz, $CDCl_3$): δ/ppm 14.4, 22.9, 26.6, 29.3, 29.4, 30.6, 32.0, 36.6, 50.5, 122.7, 124.3, 137.9.

Since the physical properties of ILs are sensitive to impurities and water content, (Omim)(NO_3) was dried and degassed for 24 h at 343.15 K under vacuum, and was kept in bottle under argon gas. The mass fraction of water in the dried product was $1.01 \cdot 10^{-3}$. The water content of IL was determined by using a 684 Karl Fischer coulometer. The mass fraction of chloride ion, determined by the potentiometry titration method, was less than $1 \cdot 10^{-4}$. Ethanol (CAS No. 64-17-5) and 1-propanol (CAS No. 71-23-8) were purchased from Merck Company. 1-Butanol (CAS No. 71-36-3) was supplied by Fluka. The water mass fractions of ethanol, 1-propanol, and 1-butanol, determined by using the Karl Fischer coulometer, were $2.4 \cdot 10^{-6}$, $3.8 \cdot 10^{-6}$, and $5.2 \cdot 10^{-6}$, respectively.

The mixtures of alcohol and IL were prepared by mass using a laboratory balance with the precision of $\pm 10^{-4}$ g. The experimental uncertainty for mass was $\pm 1 \cdot 10^{-4}$. The mixtures were placed into stopper bottles and stirred for 5 min. All of the samples were prepared immediately before the density and viscosity measurements were made to prevent the evaporation of the alcohol.

Table 4. Density ρ, Viscosity η, Excess Molar Volume V^E, and Viscosity Deviation $\Delta\eta$ of the Binary System of 1-Propanol (1) + (Omim)(NO_3) (2) at Several Temperatures

x_1	ρ / g·cm^{-3}	η / mPa·s	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s	ρ / g·cm^{-3}	η / mPa·s	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s
		T = 283.15 K				T = 288.15 K		
0.107	1.0648	1090.8	−0.035	−902.2	1.0614	733.2	−0.062	−549.4
0.258	1.0489	487.4	−0.077	−1169.2	1.0454	355.6	−0.085	−710.6
0.364	1.0351	217.6	−0.104	−1202.8	1.0316	164.2	−0.119	−750.0
0.470	1.0185	113.9	−0.154	−1070.4	1.0151	85.8	−0.189	−676.6
0.556	1.0022	65.3	−0.212	−927.3	0.9989	52.0	−0.265	−587.1
0.671	0.9750	28.8	−0.329	−707.7	0.9716	23.8	−0.381	−450.5
0.756	0.9490	16.2	−0.408	−530.9	0.9455	13.7	−0.449	−338.8
0.866	0.9029	7.8	−0.401	−294.2	0.8990	6.8	−0.404	−188.0
		T = 293.15 K				T = 298.15 K		
0.107	1.0583	510.9	−0.077	−341.9	1.0551	364.6	−0.081	−228.7
0.258	1.0423	257.7	−0.120	−451.3	1.0391	191.4	−0.137	−301.9
0.364	1.0285	126.9	−0.156	−481.1	1.0253	99.7	−0.179	−323.4
0.470	1.0119	75.4	−0.230	−431.7	1.0086	59.9	−0.255	−293.0
0.556	0.9956	42.1	−0.302	−383.1	0.9923	34.4	−0.328	−261.6
0.671	0.9683	19.9	−0.414	−295.8	0.9648	16.8	−0.437	−203.1
0.756	0.9420	11.7	−0.474	−223.1	0.9384	10.1	−0.495	−153.6
0.866	0.8953	5.9	−0.423	−124.2	0.8916	5.1	−0.443	−85.7
		T = 303.15 K				T = 308.15 K		
0.107	1.0520	267.2	−0.082	−149.1	1.0488	201.2	−0.085	−100.3
0.258	1.0360	146.6	−0.163	−199.6	1.0329	113.4	−0.187	−137.4
0.364	1.0221	78.7	−0.213	−218.4	1.0190	63.0	−0.248	−152.2
0.470	1.0054	48.5	−0.283	−199.3	1.0022	39.7	−0.320	−140.0
0.556	0.9889	28.6	−0.352	−179.3	0.9856	24.0	−0.384	−126.7
0.671	0.9613	14.4	−0.458	−140.2	0.9579	12.4	−0.488	−99.7
0.756	0.9350	8.8	−0.527	−106.4	0.9314	7.6	−0.547	−76.0
0.866	0.8879	4.5	−0.466	−59.6	0.8843	3.9	−0.490	−42.7
		T = 313.15 K				T = 318.15 K		
0.107	1.0458	153.0	−0.108	−70.2	1.0427	120.0	−0.130	−49.2
0.258	1.0297	86.9	−0.203	−98.7	1.0265	65.9	−0.211	−74.9
0.364	1.0158	51.1	−0.261	−108.2	1.0124	42.1	−0.265	−78.8
0.470	0.9989	32.9	−0.342	−100.2	0.9956	27.6	−0.357	−73.4
0.556	0.9823	20.4	−0.413	−91.3	0.9789	17.4	−0.436	−67.4
0.671	0.9545	10.8	−0.523	−72.4	0.9512	9.5	−0.565	−53.7
0.756	0.9279	6.7	−0.589	−55.3	0.9244	5.9	−0.625	−41.3
0.866	0.8806	3.5	−0.518	−31.3	0.8768	3.1	−0.547	−23.4
		T = 323.15 K				T = 333.15 K		
0.107	1.0397	95.3	−0.134	−36.2	1.0336	58.4	−0.150	−20.7
0.258	1.0235	53.4	−0.235	−56.1	1.0173	36.5	−0.269	−29.4
0.364	1.0093	35.0	−0.289	−59.1	1.0030	25.0	−0.332	−31.6
0.470	0.9923	23.4	−0.373	−55.1	0.9858	17.3	−0.433	−30.0
0.556	0.9756	15.1	−0.458	−50.9	0.9690	11.6	−0.525	−28.3
0.671	0.9477	8.4	−0.593	−40.8	0.9409	6.5	−0.679	−23.3
0.756	0.9209	5.2	−0.660	−31.6	0.9138	4.2	−0.748	−18.2
0.866	0.8731	2.8	−0.578	−18.0	0.8656	2.3	−0.653	−10.5

In this study, the Anton Paar DMA-5000 digital densitometer is used for measuring the density with an experimental uncertainty of less than $\pm$ 5·10^{-5} g·cm^{-3}. The kinematic viscosity is measured using an Anton Paar automated microviscometer (AMVn), which measures viscosities based on the falling ball principle. The diameter of the applied capillary for the viscosity measurement is 3 mm for the viscosities lower than 80 mPa·s and 4 mm for the viscosity in the range of (80 to 2500) mPa·s. The experimental uncertainty for (3 and 4) mm capillaries are ($\pm$ 0.1 and $\pm$ 0.25) mPa·s, respectively. The temperature is controlled in the viscometer and densitometer by a built-in Pt100 thermometer. The uncertainties in this range of temperatures are $\pm$ 0.1 K.

Results and Discussion

The experimental densities and viscosities of pure (Omim)(NO_3) are reported in Table 1. As Table 1 shows, the pure density and viscosity are reduced by increasing temperature. The observed viscosities of (Omim)(NO_3) are compared with those reported by Seddon et al.[3] in Table 2. The difference is defined as follows:

$$\Delta = (\eta_{ref} - \eta_{exp})/\eta_{ref} \quad (1)$$

where η_{ref} and η_{exp} are the values of viscosity in literature and in the experiment, respectively. As seen in Table 2, the experimental viscosity data are lower than the values reported by Seddon et al. The difference may be due to the existence of impurities in IL. According to the information reported by Seddon et al.,[3] the mass fraction of chloride ion in their IL was 0.028 while it was less than 1·10^{-4} in the present work. As Seddon et al. have mentioned, the presence of even low concentrations of chloride in the IL substantially increases the viscosity. For example by addition the chloride ion from (0.01 to 2.2) mol·kg^{-1}, the viscosity of (Omim)(NO_3) increases from (1238 to 8465) mPa·s.[4] In addition, the water content of (Omim)(NO_3) in this work is greater than from

Table 5. Density ρ, Viscosity η, Excess Molar Volume V^E, and Viscosity Deviation $\Delta\eta$ of the Binary System of 1-Butanol (1) + (Omim)(NO_3) (2) at Several Temperatures

x_1	ρ / g·cm^{-3}	η / mPa·s	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s	ρ / g·cm^{-3}	η / mPa·s	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s
		T = 283.15 K				T = 288.15 K		
0.168	1.0558	771.0	−0.017	−1086.2	1.0525	532.4	−0.053	−662.9
0.261	1.0439	435.1	−0.044	−1214.9	1.0406	310.8	−0.089	−751.2
0.358	1.0298	241.2	−0.091	−1192.7	1.0264	179.3	−0.126	−743.8
0.458	1.0127	126.4	−0.148	−1084.8	1.0092	98.8	−0.169	−681.0
0.570	0.9896	59.4	−0.214	−902.3	0.9861	47.4	−0.235	−571.9
0.676	0.9626	29.9	−0.267	−695.7	0.9591	24.7	−0.296	−442.9
0.754	0.9384	18.7	−0.308	−533.2	0.9350	15.7	−0.344	−340.1
0.848	0.9022	10.4	−0.303	−332.0	0.8987	9.0	−0.330	−212.1
		T = 293.15 K				T = 298.15 K		
0.168	1.0493	378.7	−0.071	−416.1	1.0462	276.5	−0.076	−276.4
0.261	1.0374	229.4	−0.103	−476.9	1.0342	171.5	−0.117	−319.9
0.358	1.0231	137.0	−0.140	−476.9	1.0199	105.1	−0.161	−322.2
0.458	1.0059	74.2	−0.190	−444.6	1.0027	59.3	−0.214	−301.8
0.570	0.9828	38.5	−0.261	−373.7	0.9795	31.8	−0.285	−255.3
0.676	0.9558	20.6	−0.331	−290.7	0.9524	17.4	−0.362	−199.5
0.754	0.9315	13.4	−0.366	−223.8	0.9280	11.4	−0.388	−153.9
0.848	0.8951	7.8	−0.348	−139.9	0.8916	6.8	−0.368	−96.4
		T = 303.15 K				T = 308.15 K		
0.168	1.0430	206.6	−0.080	−181.5	1.0399	157.7	−0.104	−123.4
0.261	1.0311	132.2	−0.138	−212.8	1.0280	103.5	−0.167	−146.4
0.358	1.0168	80.6	−0.192	−219.4	1.0136	64.5	−0.219	−152.9
0.458	0.9994	47.9	−0.240	−205.7	0.9962	39.3	−0.268	−144.6
0.570	0.9761	26.4	−0.312	−175.3	0.9728	22.3	−0.341	−124.0
0.676	0.9490	14.9	−0.379	−137.6	0.9456	12.8	−0.409	−97.9
0.754	0.9245	9.9	−0.413	−106.5	0.9211	8.7	−0.441	−75.8
0.848	0.8880	5.9	−0.389	−66.9	0.8844	5.2	−0.414	−47.9
		T = 313.15 K				T = 318.15 K		
0.168	1.0369	122.8	−0.133	−85.3	1.0338	97.4	−0.156	−60.4
0.261	1.0249	78.5	−0.200	−106.5	1.0218	63.4	−0.227	−77.0
0.358	1.0104	52.2	−0.248	−108.8	1.0073	42.9	−0.279	−79.3
0.458	0.9930	32.6	−0.301	−103.6	0.9898	27.4	−0.330	−76.0
0.570	0.9694	19.0	−0.358	−89.5	0.9662	16.3	−0.394	−66.1
0.676	0.9421	11.2	−0.427	−71.0	0.9387	9.8	−0.463	−52.7
0.754	0.9176	7.6	−0.473	−55.2	0.9142	6.8	−0.509	−41.1
0.848	0.8808	4.6	−0.441	−35.0	0.8772	4.0	−0.472	−26.2
		T = 323.15 K				T = 333.15 K		
0.168	1.0308	73.2	−0.165	−49.4	1.0247	48.9	−0.192	−24.9
0.261	1.0187	51.6	−0.241	−57.5	1.0126	35.5	−0.287	−30.1
0.358	1.0041	35.7	−0.301	−59.3	0.9979	25.5	−0.360	−31.7
0.458	0.9865	23.2	−0.357	−57.2	0.9801	17.2	−0.411	−31.3
0.570	0.9629	14.1	−0.425	−50.0	0.9561	10.9	−0.474	−27.9
0.676	0.9353	8.7	−0.491	−40.0	0.9284	6.8	−0.548	−22.7
0.754	0.9106	6.0	−0.530	−31.3	0.9035	4.8	−0.586	−17.9
0.848	0.8736	3.6	−0.503	−20.0	0.8664	2.9	−0.580	−11.6

the work of Seddon et al.[3] ($0.22 \cdot 10^{-3}$), which can produce a discrepancy in viscosity values.

The experimental densities and viscosities of (Omim)(NO_3) + ethanol, 1-propanol, and 1-butanol binary systems are reported in Tables 3 to 5, respectively. The excess molar volume and viscosity deviations are calculated from the following equations:

$$V^E = \sum_{i=1}^{N} x_i M_i [1/\rho - 1/\rho_i] \quad (2)$$

$$\Delta\eta = \eta - \sum_{i=1}^{N} x_i \eta_i \quad (3)$$

where ρ and ρ_i are the densities of the mixture and the i component, respectively; x_i represents the mole fraction of the i component; M_i is the molecular weight of the i component; η and η_i are the viscosities of mixture and the i component, respectively.

As Tables 3 to 5 show, the excess molar volume and viscosity deviations are negative in the whole range of concentration. This behavior can be attributed to the packing effect and ion−dipole interaction of alcohol molecules with the IL.[16] The molar volume of (Omim)(NO_3) is greater than the molar volume of the alcohol, and the difference between these molar volumes imply that the alcohol molecules fit in the available volume of IL upon mixing.[16] Also, the negative value of the excess molar volume shows that the ion−dipole interactions between alcohol and IL are dominating.[16]

The excess molar volume and viscosity deviation are fitted by the Redlich−Kister equation:

$$\Delta Q_{ij} = x_i x_j \sum_{P=0}^{M} B_P (x_i - x_j)^P \quad (4)$$

where ΔQ_{ij} is excess molar volume or viscosity deviation, x is the mole fraction, B_P is the fitting parameter, and M is the degree

Table 6. Fitted Parameters of Equation 4 and Standard Relative Deviation (*s*) for the Binary System of Ethanol (1) + (Omim)(NO_3) (2) at Several Temperatures

	B_0	B_1	B_2	B_3	s
		$T = 283.15$ K			
V^E/(cm^3·mol^{-1})	−1.6621	−2.8162	−5.9804	−4.6360	0.007
$\Delta\eta$/(mPa·s)	−4274.05	3567.55	−2973.63	1632.68	0.040
		$T = 288.15$ K			
V^E/(cm^3·mol^{-1})	−1.9063	−3.1359	−5.9541	−4.3650	0.007
$\Delta\eta$/(mPa·s)	−2683.13	2157.94	−1627.30	810.66	0.027
		$T = 293.15$ K			
V^E/(cm^3·mol^{-1})	−2.1208	−3.2636	−5.7663	−4.3320	0.005
$\Delta\eta$/(mPa·s)	−1755.83	1350.68	−924.26	423.24	0.029
		$T = 298.15$ K			
V^E/(cm^3·mol^{-1})	−2.1917	−3.2116	−5.9557	−4.6848	0.007
$\Delta\eta$/(mPa·s)	−1183.38	870.97	−555.53	242.89	0.014
		$T = 303.15$ K			
V^E/(cm^3·mol^{-1})	−2.2769	−3.0956	−5.9432	−5.0820	0.006
$\Delta\eta$/(mPa·s)	−814.26	577.03	−304.97	90.33	0.009
		$T = 308.15$ K			
V^E/(cm^3·mol^{-1})	−2.3402	−3.1797	−5.8716	−4.8438	0.003
$\Delta\eta$/(mPa·s)	−578.41	399.23	−187.70	33.89	0.011
		$T = 313.15$ K			
V^E/(cm^3·mol^{-1})	−2.4900	−3.2135	−5.3472	−4.4138	0.004
$\Delta\eta$/(mPa·s)	−419.38	280.32	−108.78	−4.50	0.018
		$T = 318.15$ K			
V^E/(cm^3·mol^{-1})	−2.5197	−3.1081	−5.7840	−4.6892	0.007
$\Delta\eta$/(mPa·s)	−308.65	199.05	−60.42	−24.20	0.018
		$T = 323.15$ K			
V^E/(cm^3·mol^{-1})	−2.6290	−3.2449	−5.6823	−4.5164	0.007
$\Delta\eta$/(mPa·s)	−227.48	135.60	−101.24	69.26	0.021
		$T = 328.15$ K			
V^E/(cm^3·mol^{-1})	−2.7349	−3.2273	−5.7734	−4.8350	0.005
$\Delta\eta$/(mPa·s)	−174.63	99.77	−73.29	50.83	0.023
		$T = 333.15$ K			
V^E/(cm^3·mol^{-1})	−2.8105	−3.1744	−5.7529	−4.8511	0.007
$\Delta\eta$/(mPa·s)	−128.63	66.20	−43.59	31.14	0.025

Table 7. Fitted Parameters of Equation 4 and Standard Relative Deviation (*s*) for the Binary System of 1-Propanol (1) + (Omim)(NO_3) (2) at Several Temperatures

	B_0	B_1	B_2	B_3	s
		$T = 283.15$ K			
V^E/(cm^3·mol^{-1})	−0.6849	−1.3875	−2.3226	−1.3658	0.009
$\Delta\eta$/(mPa·s)	−4025.50	3087.50	−2867.16	2008.69	0.023
		$T = 288.15$ K			
V^E/(cm^3·mol^{-1})	−0.8529	−1.8428	−2.1968	−0.2465	0.009
$\Delta\eta$/(mPa·s)	−2536.32	1818.69	−1641.40	1223.83	0.028
		$T = 293.15$ K			
V^E/(cm^3·mol^{-1})	−1.0062	−1.7821	−2.2289	−0.3674	0.008
$\Delta\eta$/(mPa·s)	−1641.52	1101.05	−978.40	764.20	0.026
		$T = 298.15$ K			
V^E/(cm^3·mol^{-1})	−1.1063	−1.7708	−2.2473	−0.5310	0.007
$\Delta\eta$/(mPa·s)	−1114.58	706.66	−642.66	535.93	0.027
		$T = 303.15$ K			
V^E/(cm^3·mol^{-1})	−1.2205	−1.6467	−2.2996	−0.9977	0.009
$\Delta\eta$/(mPa·s)	−760.74	445.97	−388.03	341.68	0.028
		$T = 308.15$ K			
V^E/(cm^3·mol^{-1})	−1.3610	−1.5817	−2.2598	−1.2939	0.006
$\Delta\eta$/(mPa·s)	−536.37	294.36	−241.03	217.84	0.027
		$T = 313.15$ K			
V^E/(cm^3·mol^{-1})	−1.4526	−1.7920	−2.5003	−0.9445	0.009
$\Delta\eta$/(mPa·s)	−386.07	204.00	−164.56	147.34	0.022
		$T = 318.15$ K			
V^E/(cm^3·mol^{-1})	−1.5211	−2.0879	−2.7568	−0.4554	0.009
$\Delta\eta$/(mPa·s)	−286.53	154.38	−108.88	77.76	0.007
		$T = 323.15$ K			
V^E/(cm^3·mol^{-1})	−1.6041	−2.0213	−2.9598	−0.9004	0.007
$\Delta\eta$/(mPa·s)	−216.28	111.44	−77.14	54.33	0.005
		$T = 328.15$ K			
V^E/(cm^3·mol^{-1})	−1.6993	−2.2124	−3.1108	−0.8510	0.008
$\Delta\eta$/(mPa·s)	−160.24	71.37	−91.62	98.69	0.021
		$T = 333.15$ K			
V^E/(cm^3·mol^{-1})	−1.8519	−2.2880	−3.2632	−1.0212	0.008
$\Delta\eta$/(mPa·s)	−117.43	45.11	−52.15	58.12	0.015

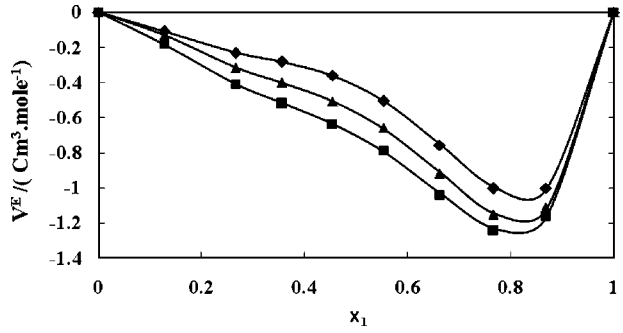

Figure 1. Plot of the excess molar volumes V^E against mole fraction for the binary system of ethanol (1) + (Omim)(NO_3) (2) at different temperatures: ♦, 283.15 K; ▲, 303.15 K; ■, 333.15 K. The solid lines represent the corresponding correlation by the Redlich−Kister equation.

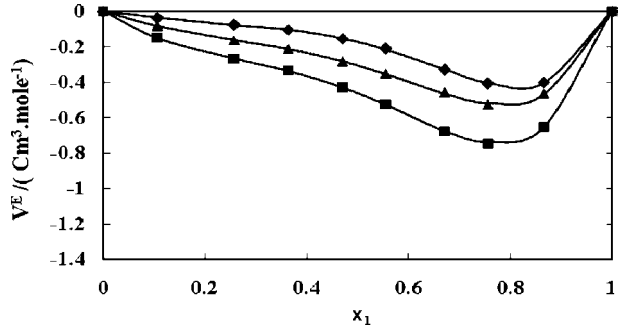

Figure 2. Plot of the excess molar volumes V^E against mole fraction for the binary system of 1-propanol (1) + (Omim)(NO_3) (2) at different temperatures: ♦, 283.15 K; ▲, 303.15 K; ■, 333.15 K. The solid lines represent the corresponding correlation by the Redlich−Kister equation.

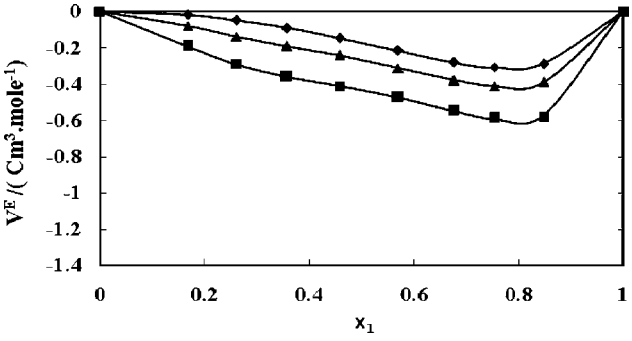

Figure 3. Plot of the excess molar volumes V^E against mole fraction for the binary system of 1-butanol (1) + (Omim)(NO_3) (2) at different temperatures: ♦, 283.15 K; ▲, 303.15 K; ■, 333.15 K. The solid lines represent the corresponding correlation by the Redlich−Kister equation.

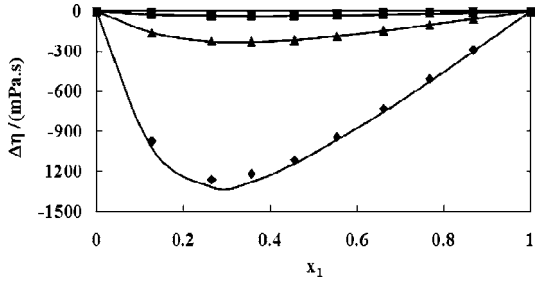

Figure 4. Plot of the viscosity deviation $\Delta\eta$ against mole fraction for the binary system of ethanol (1) + (Omim)(NO_3) (2) at different temperatures: ♦, 283.15 K; ▲ 303.15 K; ■, 333.15 K. The solid lines represent the corresponding correlation by the Redlich−Kister equation.

of the polynomial expansion. A third-order polynomial is found to be optimum for the properties of the alcohol (1) +

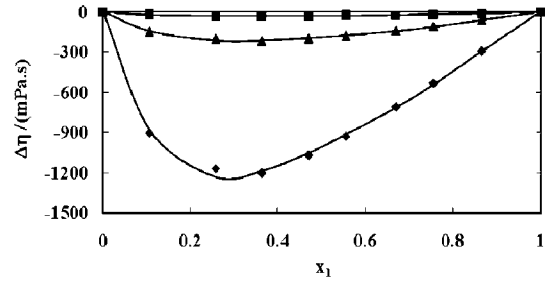

Figure 5. Plot of the viscosity deviation $\Delta\eta$ against mole fraction for the binary system of 1-propanol (1) + (Omim)(NO_3) (2) at different temperatures: ♦, 283.15 K; ▲, 303.15 K; ■, 333.15 K. The solid lines represent the corresponding correlation by the Redlich−Kister equation.

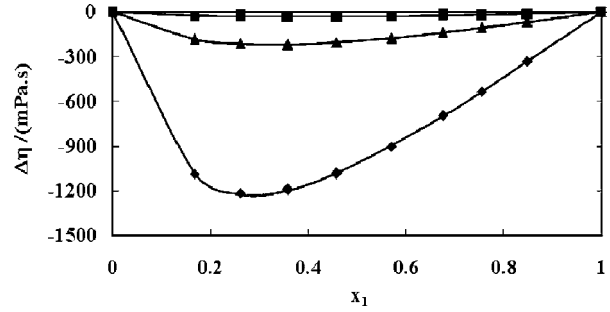

Figure 6. Plot of the viscosity deviation $\Delta\eta$ against mole fraction for the binary system of 1-butanol (1) + (Omim)(NO_3) (2) at different temperatures: ♦, 283.15 K; ▲, 303.15 K; ■, 333.15 K. The solid lines represent the corresponding correlation by the Redlich−Kister equation.

(Omim)(NO_3) (2) binary system. The fitting parameters are reported in Tables 6 to 8. The standard relative deviations (*s*) in these tables are defined by the following equation:

$$s = \left[\sum_{i}^{n}(z - z_{cal})/z)^2/(n_d - n_p)\right]^{1/2} \quad (5)$$

where z and z_{cal} are the values of experimental and calculated property, respectively, n_d is the number of data, and n_p is the number of parameters in the Redlich−Kister equation.

The excess molar volume and viscosity deviation can be fitted by the Redlich−Kister equation with a reasonable agreement. The comparison of the excess molar volumes with those calculated by the Redlich−Kister equation at different temperatures for the ethanol (1) + (Omim)(NO_3) (2) system is shown in Figure 1. As the figure shows, the excess molar volume and viscosity deviation are decreasing with increasing the temperature. Similar behaviors are observed for the excess molar volume for the systems of propanol (1) + (Omim)(NO_3) (2) and butanol (1) + (Omim)(NO_3) (2), which are presented in Figures 2 and 3, respectively. As can be seen from Figures 1 to 3, the excess molar volumes for the binary system of ethanol (1) + (Omim)(NO_3) (2) are more negative than those for the other two systems. This behavior is also observed in the other isotherms. The excess molar volumes are increasing with increasing the alcohol chain length. This implies that the ion−dipole interactions and packing effects are stronger in ethanol systems than in the other alcohol systems. This behavior is identical to the results reported by Gonzalez et al.[7] for the (Omim)(Cl) binaries with methanol, ethanol, and 1-propanol. The comparison of experimental densities and viscosities for (Omim)(Cl) reported by Gonzalez et al.[7] with those in the present research for (Omim)(NO_3) indicates that the densities of (Omim)(Cl) are lower than the densities of (Omim)(NO_3),

Table 8. Fitted Parameters of Equation 4 and Standard Relative Deviation (*s*) for the Binary System of 1-Butanol (1) + (Omim)(NO_3) (2) at Several Temperatures

	B_0	B_1	B_2	B_3	s
		$T = 283.15$ K			
V^E/(cm^3·mol^{-1})	−0.6914	−1.2902	−0.9828	−0.4972	0.008
$\Delta\eta$/(mPa·s)	−4064.12	3227.81	−2522.15	1396.60	0.005
		$T = 288.15$ K			
V^E/(cm^3·mol^{-1})	−0.7653	−1.1005	−1.4459	−0.9894	0.009
$\Delta\eta$/(mPa·s)	−2564.31	1944.68	−1441.40	794.20	0.004
		$T = 293.15$ K			
V^E/(cm^3·mol^{-1})	−0.8571	−1.2607	−1.5285	−0.6451	0.002
$\Delta\eta$/(mPa·s)	−1671.40	1207.69	−820.78	433.83	0.005
		$T = 298.15$ K			
V^E/(cm^3·mol^{-1})	−0.9599	−1.2854	−1.5151	−0.7767	0.006
$\Delta\eta$/(mPa·s)	−1138.75	792.44	−514.00	268.01	0.004
		$T = 303.15$ K			
V^E/(cm^3·mol^{-1})	−1.0700	−1.1679	−1.4899	−1.2802	0.007
$\Delta\eta$/(mPa·s)	−780.72	519.12	−293.96	129.90	0.003
		$T = 308.15$ K			
V^E/(cm^3·mol^{-1})	−1.1835	−1.1684	−1.6411	−1.3077	0.008
$\Delta\eta$/(mPa·s)	−550.42	347.85	−176.93	68.52	0.003
		$T = 313.15$ K			
V^E/(cm^3·mol^{-1})	−1.2802	−1.0847	−1.8828	−1.4906	0.009
$\Delta\eta$/(mPa·s)	−397.54	247.06	−108.25	19.15	0.008
		$T = 318.15$ K			
V^E/(cm^3·mol^{-1})	−1.4100	−1.1338	−2.0348	−1.4850	0.006
$\Delta\eta$/(mPa·s)	−293.02	174.38	−64.09	−1.66	0.009
		$T = 323.15$ K			
V^E/(cm^3·mol^{-1})	−1.5161	−1.1793	−2.0824	−1.6111	0.004
$\Delta\eta$/(mPa·s)	−217.81	120.65	−82.70	57.15	0.007
		$T = 328.15$ K			
V^E/(cm^3·mol^{-1})	−1.6344	−1.2064	−2.1793	−1.8515	0.007
$\Delta\eta$/(mPa·s)	−164.52	85.00	−59.15	44.59	0.008
		$T = 333.15$ K			
V^E/(cm^3·mol^{-1})	−1.7241	−1.0546	−2.4135	−2.4631	0.007
$\Delta\eta$/(mPa·s)	−120.32	55.81	−31.33	21.43	0.005

while their viscosities are greater than the viscosities for (Omim)(NO_3).

The viscosity deviations are compared with those calculated by the Redlich−Kister equation at temperatures of (283, 303, and 313) K in Figures 4, 5, and 6 for the systems of alcohol (1) + (Omim)(NO_3) (2). The viscosity deviation decreases around $x_1 = 0.3$ and increases afterward. These behaviors are observed in all of the isotherms.

The experimental densities for the binary systems of alcohol (1) + (Omim)(NO_3) (2) are lower than the densities which were reported previously for the binary systems of alcohol (1) + (Bmim)(NO_3) (2).[11] The behavior, however, is the reverse of the behavior for the viscosity data. It can be then concluded that the length of the cation alkyl's chain in the IL's structure causes a decrease in the density of the IL while it causes an increase in its viscosity.

Conclusion

In the present research, the experimental densities and viscosities of (Omim)(NO_3) and its binary systems with ethanol, 1-propanol, and 1-butanol are measured. The excess molar volume and viscosity deviation are calculated for the binary systems at different temperatures. The third-order polynomial of the Redlich−Kister equation is used for fitting the excess molar volume and viscosity deviations. The results show that the third-order Redlich−Kister equation can fit these data with a reasonable agreement. The effect of alcohol chain length on the excess molar volume is investigated. The results reveal that the excess molar volumes increase with increasing the alcohol chain length. It shows that as the alkyl chain length increases from butyl group to octyl group, the densities of alcohol (1) + IL (2) binary systems decrease, while the values of viscosity increase.

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