

Excess Volume of Binary Mixtures of Water (Methanol or Ethanol) with an Ionic Liquid [EEIM][DEP] or [BEIM][DEP] at 1 atm and (293.15 to 333.15) K

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Abstract: Densities were measured for the binary mixtures of water (methanol or ethanol) and an ionic liquid (IL) 1,3-diethylimidazolium diethylphosphate [EEIM][DEP] or 1-n-butyl-3-ethylimidazolium diethylphosphate [BEIM][DEP] at 1 atm and (293.15 to 333.15) K using a vibrating-tube densimeter. The molecular volume, standard entropy, and lattice energy of the two ILs were estimated with the Glasser theory. The excess volumes are negative for all binary mixtures studied in the whole composition range, and reach to the maximum at the mole fraction of IL being around 0.3. Besides, they decrease with increasing temperature for the aqueous solution of ILs, whilst a reverse trend is found for the IL solutions of methanol or ethanol at any concentration. The excess molar volumes were correlated successfully by a five-parameter polynomial equation as a function of temperature and mole fraction of IL with average absolute relative deviation (ARD) of density within 0.02%.

Keywords: Density, excess volume, ionic liquid, water, methanol, ethanol.

1. INTRODUCTION

Ionic liquids (ILs), as a novel class of organic molten salts at ambient temperatures, show the characteristics of both electrolyte and non-electrolyte. And their ionic attribute is highly dependent on the solvent property and interaction with the solvent, giving rise to varying non-ideality of solution behavior. To reveal the non-ideality of the IL solutions from the aspect of excess volume, density property is measured for binary ILs solutions at different conditions. As a continuation study on the thermodynamic properties of ILs [1–3], we measured the density of binary mixtures of water (methanol or ethanol) and an IL ([EEIM][DEP] or [BEIM][DEP]) at 1 atm in the temperature range of (293.15 to 333.15) K. The volume properties of the ILs were discussed in terms of the Glasser's theory. The excess molar volumes (V^E) are correlated as a function of temperature and composition by an empirical polynomial equation. The solution behavior of these IL-containing mixtures was discussed in terms of the variation of excess molar volume with temperature, composition and the constituents of the mixtures.

2. EXPERIMENTAL

2.1. Materials

The chemicals used are N-ethylimidazole and N-butylimidazole (≥ 99.5 wt.%, Zhejiang Kaile Reagents

Company, China), triethylphosphate (≥ 99.5 wt.%, Tianjin Guangfu Reagents Company, China) and aether (≥ 99.5 wt.%, Beijing Yili Reagents Company, China), which were used as received. Methanol and ethanol were supplied by Beijing Chemical Plant with nominal purity above 99.7 wt.%, degassed ultrasonically before use, and stored over the activated fresh molecular sieves of type 4 Å (Union Carbide) in tightly sealed glass bottles. The ILs [EEIM][DEP] and [BEIM][DEP] were synthesized and purified in our laboratory according to the method described in the literature [4], and their purities are better than 99 wt.% as determined by the NMR analysis. Water content in the ILs is below 0.032% as determined by the Karl Fisher titrator (type CBS-1A).

2.2. APPARATUS AND PROCEDURE

The densities of pure ILs and their binary mixtures with water (methanol or ethanol) at different temperatures were measured using the digital vibrating tube densimeter (DMA 4500M, Anton Paar Co. Ltd., Austria). The binary mixtures with different mole fractions of ILs were prepared by weighting with an electronic balance (type AR 2130, Ohaus Co. Ltd., USA) with a mass precision of ± 0.001 g. The nominal uncertainty of the experimental density is $\pm 1 \times 10^{-5}$ g·cm⁻³. The temperature of the densimeter measuring cell is better than 0.002 K and the precision of measurement is ± 0.01 K with a built-in thermostat. The accuracy of the density data is verified by comparing the experimental data of water with the reference

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Table 1: Density ρ (g·cm⁻³) of Pure ILs at 1 atm and (293.15 to 333.15) K

Ionic liquid	T/K								
	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15
[EEIM][DEP]	1.12932	1.12596	1.12260	1.11924	1.11588	1.11251	1.10915	1.10580	1.10247
[BEIM][DEP]	1.08660	1.08321	1.07984	1.07650	1.07315	1.06980	1.06645	1.06310	1.05976

values [5] from 293.15 to 333.15 with the average relative deviation within 0.02%.

3. RESULTS AND DISCUSSION

3.1. Density and Derived Properties of Pure ILs

The densities of pure ILs [EEIM][DEP] and [BEIM][DEP] at 1 atm were measured in the temperature range of (293.15 to 333.15) K, and listed in Table 1. The density of the pure components studied here follows the order of ethanol≈methanol<water <[BEIM][DEP]< [EEIM][DEP] in the fixed other conditions. The density of ILs is generally higher than that of molecular solvents due to the strong electrostatic interaction among ions of the ILs. Besides, the density of ILs decreases with the increase of the length of the alkyl substitute to the imidazolium ring, implying that the electrostatic interaction plays a predominant role since a shorter alkyl substitute means a smaller ionic size and a lower steric hinderance for the closest approaches among ionic species and thus a stronger Columbic interaction albeit the dispersion interaction is a little weaker for the ILs with shorter alkyl substitutes.

Density results for the pure ILs were used to derive other thermodynamic properties such as the coefficient of thermal expansion, molar volume, standard entropy and lattice energy. The coefficient of thermal expansion α of a liquid is defined by the following equation:

$$\alpha = \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_p = - \left(\frac{\partial \ln \rho}{\partial T} \right)_p \quad (1)$$

where V and ρ is the molar volume and density of the IL, respectively. The variation trend of $\ln \rho$ vs. T for the two ILs studied is presented in Figure 1. As shown in the figure, the values of $\ln \rho$ decrease slightly in a linear way with the increasing temperature. The α values were obtained from the slope of the linear regression of $\ln \rho$ against T , being about $6.02 \times 10^{-4} \text{ K}^{-1}$ and $6.25 \times 10^{-4} \text{ K}^{-1}$, respectively, for [EEIM][DEP] and [BEIM][DEP]. The coefficient of thermal expansion of the ILs is quite low, and increases with the length of the

alkyl substitutes to the imidazolium ring as a result of the decreasing electrostatic interaction among ionic species in the ILs involved.

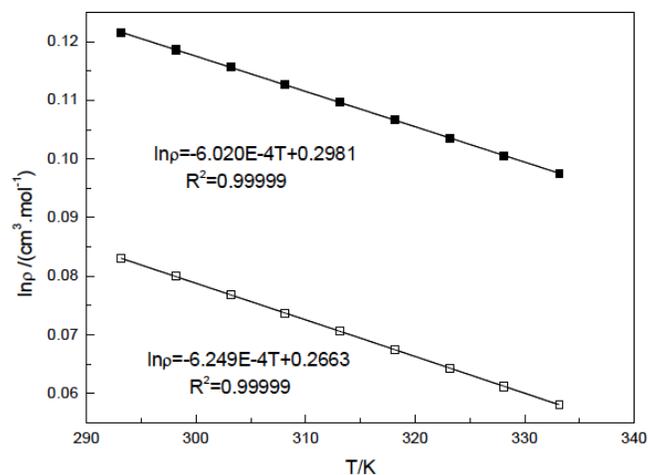


Figure 1: Plot of $\ln(\rho/\text{g}\cdot\text{cm}^{-3})$ against T/K for ILs: (■), [EEIM][DEP]; (□), [BEIM][DEP].

From the experimental density data, the molecular volume of the ILs, V_m , can be estimated using the following equation.

$$V_m = M / (N_A \cdot \rho) \quad (2)$$

where M is the molar mass, N_A is the Avogadro constant, ρ is the density. The V_m values thus determined for [EEIM][DEP] and [BEIM][DEP] at 298.15 K are 0.4106 and 0.4698 nm³, respectively. By comparing the V_m values of these ILs, the mean contribution of a methylene group (-CH₂-) to the molecular volume is about 0.0289 nm³, which is close to the literature value 0.0293 nm³ derived from ILs [MMIM][DMP] and [EMIM][DEP] [6], 0.0280 nm³ from n-alcohols, 0.0272 nm³ from n-amines and 0.0267 nm³ from n-paraffins [7].

The standard entropy, S° , can be calculated in terms of the molecular volume and a good linear relationship proposed by Glasser [7]. The linear correlation constants in Eq. (3) are appropriate for the ILs with large organic cations.

Table 2: The Molecular Volume, V_m , Standard Entropy, S° , and Lattice Energy, U_{POT} , Derived from Density of [EEIM][DEP] and [BEIM][DEP] at 298.15 K

Ionic liquid	$M/(\text{g}\cdot\text{mol}^{-1})$	$\rho/(\text{g}\cdot\text{cm}^{-3})$	$V_m/(\text{nm}^3)$	$S^\circ/(\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1})$	$U_{\text{POT}}/(\text{kJ}\cdot\text{mol}^{-1})$
[EEIM][DEP]	278.29	1.12596	0.4106	541.3	419.5
[BEIM][DEP]	306.34	1.08321	0.4698	615.1	405.6

$$S^\circ(298)/(J\cdot\text{mol}^{-1}\cdot\text{K}^{-1}) = 1246.5(V_m/\text{nm}^3) + 29.5 \quad (3)$$

The S° values calculated by Eq. (3) for the two ILs are listed in Table 2, from which the mean entropy contribution per methylene group (-CH₂-) to the standard entropy is determined as 36.0 J·mol⁻¹·K⁻¹. This is comparable to the literature value 36.5 J·mol⁻¹·K⁻¹ derived from ILs [MMIM][DMP] and [EMIM][DEP] [6], 35.1 J·mol⁻¹·K⁻¹ from [C_nmim][NTf₂], 33.9 J·mol⁻¹·K⁻¹ from [C_nmim][BF₄] [7] and 34.1 J·mol⁻¹·K⁻¹ from [C_nmim][AlCl₄] [8].

Density data is also useful for the estimation of lattice energy (U_{POT}) of ILs. According to the Glasser's theory, the lattice energy depends only on the chemical formula, ionic charges and molecular volume of the materials involved, and neither relies on any other structural information. For the present case of ionic liquids of MX type with charge ratio 1:1, the Coulombic interaction is the principal contribution to the lattice energy, and the U_{POT} values can be estimated by Eq. (4).

$$U_{\text{POT}}/(\text{kJ}\cdot\text{mol}^{-1}) = 1981.2(\rho/M)^{1/3} + 103.8 \quad (4)$$

As shown in Table 2, the lattice energies thus determined for [EEIM][DEP] and [BEIM][DEP] are 419.5 kJ·mol⁻¹ and 405.6 kJ·mol⁻¹, respectively. Obviously, the lattice energy of ILs is much lower than that of the fused inorganic salts, for example, the lattice energy of fused CsI which has the least crystal energy among all the alkali halides is about 602.5 kJ·mol⁻¹ [9]. The low lattice energy may be the underlying reason for forming ILs at room temperature [8].

3.2. Density of Binary Mixtures

The density data for the binary mixtures at the mole fraction of IL, x_{IL} , being about 0.2, 0.4, 0.6, and 0.8, were measured at the same pressure and temperature as that for the pure components, and listed in Table 3.

3.3. Excess Molar Volume of Binary Mixtures

The excess molar volume, V^{E} , is an important thermodynamic property to represent the non-ideality of

a solution, which is defined as the difference of the molar volume between the real mixture (v_m) and the ideal solution ($v_{m,\text{id}}$) at the same temperature, pressure and composition i.e.

$$V^{\text{E}} = V_m - V_{m,\text{id}} \quad (5)$$

The V_m and $V_{m,\text{id}}$ values can be calculated as follows:

$$V_m = \frac{x_{\text{IL}}M_{\text{IL}} + x_{\text{S}}M_{\text{S}}}{\rho_{\text{mix}}} \quad (6)$$

$$V_{m,\text{id}} = \frac{x_{\text{IL}}M_{\text{IL}}}{\rho_{\text{IL}}} + \frac{x_{\text{S}}M_{\text{S}}}{\rho_{\text{S}}} \quad (7)$$

where M_{S} and M_{IL} are the mole masses of solvent and IL, respectively. x_{S} and x_{IL} represent the mole fractions of solvent and IL, respectively. ρ_{S} , ρ_{IL} and ρ_{mix} refer to the densities of solvent, IL and the mixture, respectively.

Based on the experimental data of density, as listed in Tables 1 and 3, the excess molar volume for the binary mixtures at different conditions were calculated by Eqs. 5 to 7. The results show that the excess volumes are negative for all the systems studied at any temperature and concentration, indicating a negative deviation from the ideal solution behavior. Besides, the excess volume is relatively low being in the range of (0.9 to 1.9) cm³·mol⁻¹ for all the binary mixtures studied, and reaches the maximum at the mole fraction of IL being around 0.3. For the convenience of illustration, plots of excess molar volume against the mole fraction of IL at different temperatures for the binary mixtures of [EEIM][DEP] with ethanol and water are presented in Figures 2 and 3, respectively. As shown in Figure 2, the excess molar volume of the binary mixture ([EEIM][DEP] + ethanol) increases with increasing temperature at fixed other conditions. This trend holds for all binary mixtures of (IL+ alcohols) studied here and in good agreement with those reported elsewhere for other ILs mixtures with different alcohols [10, 11].

Table 3: Density Data Measured for the Binary Mixtures at 1 atm, Different Temperatures and Mole Fractions of ILs

x_{IL}	Density ρ (g·cm ⁻³) at the following temperatures, T/K								
	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15
[EEIM][DEP] + water									
0.199	1.12833	1.12469	1.12101	1.11730	1.11357	1.10981	1.10604	1.10223	1.09841
0.401	1.13438	1.13084	1.12729	1.12373	1.12017	1.11661	1.11303	1.10946	1.10588
0.598	1.13353	1.13012	1.12668	1.12323	1.11979	1.11635	1.11291	1.10947	1.10603
0.809	1.13121	1.12787	1.12451	1.12114	1.11777	1.11439	1.11101	1.10764	1.10427
[BEIM][DEP] + water									
0.200	1.09214	1.08850	1.08482	1.08112	1.07738	1.07359	1.06971	1.06595	1.06210
0.400	1.09338	1.08985	1.08629	1.08273	1.07915	1.07557	1.07199	1.06839	1.06479
0.599	1.09125	1.08780	1.08433	1.08085	1.07737	1.07388	1.07042	1.06697	1.06352
0.807	1.08885	1.08547	1.08210	1.07872	1.07534	1.07196	1.06856	1.06517	1.06178
[EEIM][DEP] + methanol									
0.200	1.01179	1.00762	1.00375	1.00024	0.99647	0.99271	0.9889	0.98508	0.98126
0.401	1.07411	1.07052	1.06692	1.06333	1.05974	1.05616	1.05257	1.04893	1.04522
0.601	1.10255	1.09906	1.09556	1.09208	1.08861	1.08515	1.08170	1.07827	1.07483
0.800	1.11860	1.11520	1.11177	1.10834	1.10493	1.10152	1.09812	1.09474	1.09137
[BEIM][DEP] + methanol									
0.200	0.99468	0.99095	0.98718	0.98340	0.97956	0.97588	0.97213	0.96836	0.96459
0.401	1.04466	1.04111	1.03754	1.03397	1.03041	1.02686	1.02331	1.01976	1.01621
0.600	1.06680	1.06334	1.05987	1.05641	1.05295	1.04951	1.04607	1.04263	1.03921
0.802	1.07901	1.07563	1.07224	1.06886	1.06546	1.06207	1.05868	1.05529	1.05193
[EEIM][DEP] + ethanol									
0.201	0.97488	0.97115	0.96739	0.96362	0.95986	0.95609	0.95232	0.94854	0.94475
0.402	1.04854	1.04496	1.04136	1.03778	1.03420	1.03062	1.02699	1.02325	1.01947
0.603	1.08752	1.08403	1.08053	1.07705	1.07358	1.07012	1.06667	1.06323	1.05980
0.803	1.11249	1.10907	1.10565	1.10223	1.09881	1.09540	1.09200	1.08861	1.08524
[BEIM][DEP] + ethanol									
0.200	0.96142	0.95771	0.95397	0.95023	0.94649	0.94274	0.93899	0.93523	0.93145
0.400	1.02217	1.01862	1.01505	1.01148	1.00790	1.00432	1.00071	0.99705	0.99328
0.600	1.05405	1.05056	1.04707	1.04360	1.04014	1.03668	1.03324	1.02980	1.02637
0.801	1.07366	1.07029	1.06690	1.06351	1.06011	1.05671	1.05331	1.04993	1.04655

In contrast to the binary mixtures of IL plus alcohols, the excess volumes of the (IL + water) mixtures decrease as the temperature increasing, as shown in Figure 3, implying that the non-ideality of the mixture is higher at lower temperature and decreases as the temperature increasing. This variation trend may be attributed to the strong dielectric property of water and its decreasing dielectric behavior as temperature increasing, for example, the dielectric constant of water decreases from 80.1 at 293K to 63.8 at 333K [5]. The

high dielectric property of water makes the ionic liquid partially dissociated into ions, leading to a high asymmetrical interaction between neutral water molecules and charged particles of the IL component, and accordingly a high non-ideality of the mixture and the excess volumes. This means that an aqueous solution of ILs usually shows a larger deviation from the ideal solution behavior than an alcoholic solution of ILs, which is justified by the fact that the values of V^E for the (IL + water) mixtures are always larger than those for

the (IL + alcohol) mixtures at the same temperature and concentration. Besides, as the temperature increases the ILs component is apt to present as neutral molecules rather than as ionic species due to the decreasing dielectric constant of water, leading to a lower non-ideality and excess volume.

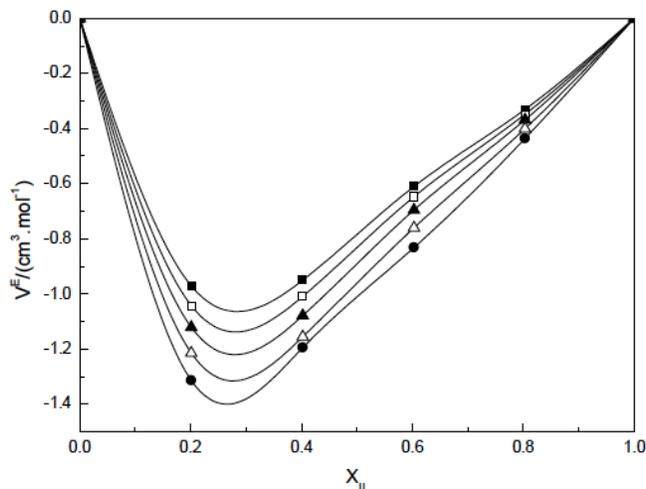


Figure 2: The experimental and correlative excess molar volume for the binary mixture ([EEIM][DEP] + ethanol) at different mole fractions of IL and temperatures: (■), 293.15 K; (□), 303.15 K; (▲), 313.15 K; (△), 323.15 K; (●), 333.15 K; (—), calculated values from Eq. (8).

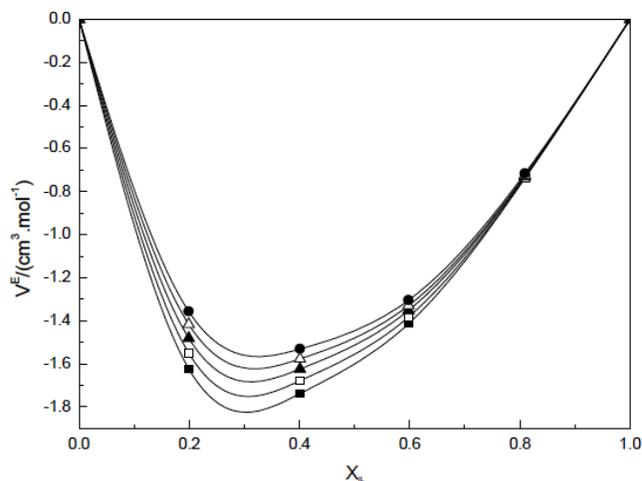


Figure 3: The experimental and correlative excess molar volume for the binary mixture ([EEIM][DEP] + water) at different mole fractions of IL and temperatures: (●), 293.15 K; (△), 303.15 K; (▲), 313.15 K; (□), 323.15 K; (■), 333.15 K; (—), calculated values from Eq. (8).

For a specific ionic liquid, the solvent property has an important influence on the non-ideality and excess molar volume of the IL-containing binary solutions, as an illustrative example, Figure 4 presents the excess molar volumes against the mole fraction of IL for the binary mixtures of [EEIM][DEP] with water, methanol and ethanol at 298.15K. It is seen that among the three

binary mixtures, the aqueous solution of IL shows the highest excess molar volume and the ethanol solution of IL shows the lowest one at the same temperature and mole fraction of IL. This trend is in agreement with the dielectric constant [5] order at 298.15K, viz. water (78.3) > methanol (32.7) > ethanol (24.5). As a result, the aqueous solution of IL is similar to an electrolyte solution, while the alcoholic solution of IL is more like a non-electrolytic organic mixture. In effect, the excess volume is a reflection of the compromising effects among molecular sizes and various interactions for a specific mixture, and is dependent on the temperature and composition. The negative excess volume may be associated with the strong ion-dipole and dipole-dipole interactions between IL and the solvent molecules, as well as the interstitial effect in molecular packing due to the differences in the size and shape of the component molecules, by which the volume of the liquid mixture is apt to be contracted.

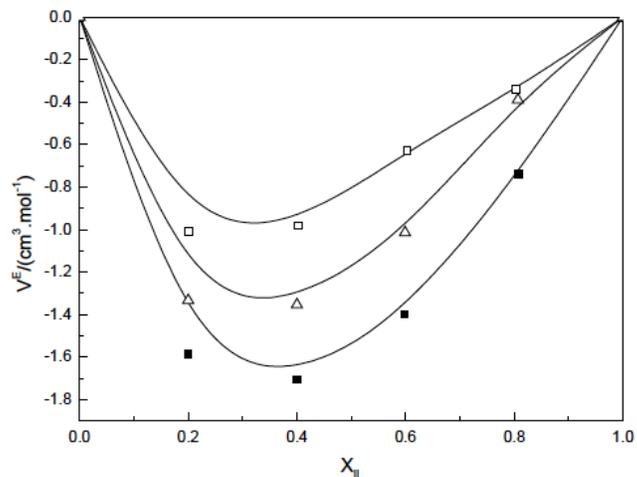


Figure 4: Excess molar volume for the binary mixtures of [EEIM][DEP] with different solvents at 298.15K: (□), [EEIM][DEP] + ethanol; (△), [EEIM][DEP] + methanol; (■), [EEIM][DEP] + water.

3.4. Correlation of the Density Data

In view of the complex variation of the excess molar volume with temperature and IL-content for a binary mixture, the experimental values of V^E are fitted to the following equation (8) by the least-square method.

$$V^E = x_{IL} (1 - x_{IL}) \sum_{i=0}^4 A_i (1 - 2x_{IL})^i (1 + BT + CT^2) \quad (8)$$

Where x_{IL} is the mole fraction of IL, T is the absolute temperature in K. The best fit was obtained when five adjustable parameters were used in Eq. 8. The resulting parameters A_0 , A_1 , A_2 , B , C , and the average

Table 4: The Regressed Parameters of Eq. (8) Along with the Deviation in ARD for the Binary Systems

Correlation results	IL + solvents binary systems		
	IL + water	IL + methanol	IL + ethanol
[EEIM][DEP] IL			
A_0	-0.00079	-0.00032	-0.00034
A_1	-0.00047	-0.00030	-0.00037
A_2	-0.00033	-0.00020	-0.00028
B	73.29247	12.86897	-9.84410
C	-0.15506	0.12047	0.13907
ARD(V^E)	1.5930%	1.7324%	0.7284%
ARD(ρ)	0.0175%	0.0146%	0.0043%
[BEIM][DEP] IL			
A_0	-0.00227	-0.00051	-0.00048
A_1	-0.00125	-0.00046	-0.00051
A_2	-0.00113	-0.00043	-0.00059
B	26.54501	12.87334	-4.51104
C	-0.05934	0.06443	0.08369
ARD(V^E)	1.1104%	2.4621%	2.9749%
ARD(ρ)	0.0104%	0.0182%	0.0129%

$$ARD = \frac{1}{n} \sum_{i=1}^n |V^{E(\text{cal})} / V^{E(\text{exp})} - 1.0|$$

relative deviations of excess molar volume between the experimental and correlated values, ARD (V^E), for each binary system are listed in Table 4. The experimental density data were accurately correlated by using Eqs. (5) through (8) with the highest correlative deviation in terms of ARD (ρ) within 0.02%, which is close to the experimental uncertainty of the densimeter used here. The goodness of Eqs. (5) through (8) for representing density and excess molar volume of the IL-containing binary mixtures can be clearly seen from Figures 2 through 4, as well as the very low ARD values as listed in Table 4.

4. CONCLUSIONS

The density data for the binary mixtures of an IL of [EEIM][DEP] or [BEIM][DEP], and a solvent of water, methanol or ethanol, were measured at 1 atm in the temperature range of (293.15 to 333.15) K using densimeter. The excess volumes are all negative in the whole temperature and concentration range studied, indicating a negative deviation from the ideality, and the variation of excess volume with temperature shows two opposite trends for the (IL + water) mixtures and the (IL + alcohol) mixtures. Moreover, the excess volume as well as the density data can be correlated

accurately by a five-parameter empirical polynomial equation.

ACKNOWLEDGEMENTS

The authors are grateful to the financial support from National Natural Science Foundation of China (21076005).

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Received on 30-08-2014

Accepted on 08-09-2014

Published on 18-09-2014

DOI: <http://dx.doi.org/10.6000/1929-5030.2014.03.03.2>