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Prediction of density and viscosity of ternary systems $[C_2q]Br+[C_4q]Br+H_2O$, $[C_2q]Br+[C_6q]Br+H_2O$, and $[C_4q]Br+[C_6q]Br+H_2O$ at different temperatures using their binary subsystems data

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Abstract: The simple equations for prediction of the density and viscosity of mixed electrolyte solutions were extended to the related properties of mixed ionic liquid solutions. The density and viscosity were measured for ternary solutions $[C_2q]Br$(N-ethylquinolinium bromide)+$[C_4q]Br$ (N-butylquinolinium bromide)+H_2O, $[C_2q]Br+[C_6q]Br$(N-hexylquinolinium bromide)+H_2O, and $[C_4q]Br+[C_6q]Br+H_2O$ and their binary subsystems $[C_2q]Br+H_2O$, $[C_4q]Br+H_2O$, and $[C_6q]Br+H_2O$ at 15, 20 and 25 °C, respectively. The results were used to test the predictability of the extended equations. The comparison results show that these simple equations can be used to predict the density and viscosity of the mixed ionic liquid solutions from the properties of their binary subsystems of equal ionic strength.

Key words: Density, viscosity, prediction, N-ethylquinolinium bromide, N-butylquinolinium bromide, N-hexylquinolinium bromide

1 Introduction

The thermodynamic and transport properties of mixed electrolyte solutions play an important role in a variety of fields, such as chemical engineering, separation processes, waste water treatment, pollution control, and oil recovery. A number of research groups have reported the physical properties of binary electrolyte solutions (Ruby and Kawai, 1926; Isono, 1984; Zhang and Han, 1996; Zhang et al, 1997; Königsberger et al, 2008). Ruby and Kawai (1926) reported the density, equivalent conductivity, and relative viscosity of the binary solutions $HCl+H_2O$, $KCl+H_2O$, and $NaCl+H_2O$ at 25 °C. Isono (1984) reported the density, electrolytic conductivity, and viscosity of binary aqueous solutions of alkaline- and rare-earth metal chlorides ($LaCl_3$, Na_2SO_4, $NaNO_3$, NaBr, KNO_3, KBr, and $Cd(NO_3)_2$) at different temperatures. Zhang and Han (1996) reported the density and viscosity of binary solutions $NaCl+H_2O$ and $KCl+H_2O$ at 25 °C. Zhang et al (1997) reported the density and viscosity of the binary solution $CaCl_2+H_2O$ at 25 °C. Königsberger et al (2008) reported the density, viscosity, and heat capacity of binary solution $MgCl_2+H_2O$ in the temperature range of 25-90 °C. However, the study of mixed electrolyte solutions is rarely reported. One of the objectives of the theory of electrolyte solutions is to calculate various properties of mixed electrolyte solutions in terms of the properties of binary solutions. Our research group has made efforts in development of simple equations to predict accurately the properties of mixed solutions based on the available information on binary electrolyte solutions (Hu, 2004; 2005). Many simple equations have been established for thermodynamic properties. For example, several simple equations have been developed for prediction of density of mixed electrolyte solutions, such as the equation of Patwardhan and Kumar (Patwardhan and Kumar, 1986a; 1986b), the rule of Young and Smith (Young and Smith, 1954), and the semi-ideal solution theory (Hu, 2003; Hu et al, 2006a). The equations of Patwardhan and Kumar and the semi-ideal solution theory have both been used together with Eyring's absolute rate theory to establish the simple equations for prediction of viscosity of mixed electrolyte solutions (Hu, 2004). These approaches can all be used to predict the thermodynamic and transport properties of mixed solutions in terms of the properties of their binary subsystems. Recently, Yang et al (2010) have made comparison between prediction results by these equations and experimental data for mixed non-electrolyte solution, mixed electrolyte solution, and

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mixed solution of electrolyte and non-electrolyte.

Ionic liquids (ILs), with melting points generally below 100 °C, are comprised entirely of ions (Brennecke and Maginn, 2001). The ILs have unique properties such as very low vapor pressure, high ionic conductivity, outstanding catalytic property, and high thermal stability. So, ILs have attracted a great deal of attention for their potential use in various fields (Brennecke and Maginn, 2001). Generally, to design an industrial process involving ILs, it is necessary to know a range of their physical properties including density and viscosity. A key strategy in the application of ILs is that ILs are mixed to binary and ternary mixtures to achieve the desired result (Baltus et al, 2004; Lin and Freeman, 2005; Finotello et al, 2008). Therefore, recently the mixtures of ILs have received growing attention (Baltus et al, 2004; Lin and Freeman, 2005; Finotello et al, 2008). Up to now the studies of IL mixtures have focused on their volumetric behavior (Rebelo et al, 2005), their effects on gas solubility (Finotello et al, 2008; Yokezeki and Shifflet, 2007) and on the local environment of solvatochromic probes (Fletcher and Pandey, 2002; Fletcher et al, 2003), and their use as stationary phases in gas chromatography (Baltazar et al, 2008) and as electrolytes in solar cells (Fredlin et al, 2007). Investigations at a molecular level understanding of the structure and dynamics of ILs mixtures have also been undertaken. For example, Xiao et al (2006) have studied the intermolecular dynamics of binary IL mixtures by using optical heterodyne-detected Raman-induced Kerr effect spectroscopy (OHD-RIKES). On the other hand, the presence of water in ILs can greatly affect their physicochemical properties (Seddon et al, 2000; Yang et al, 2007). In addition, with the development of ILs in the colloid and surfactant fields, it has become even more important to study the properties of aqueous solution of ILs (Dong et al, 2008; Wu et al, 2009). Therefore, a number of groups have studied the physicochemical properties of aqueous solution of ILs (Zhang et al, 2004; Ge et al, 2008; Goómez et al,2006; Rodriguez and Brennecke, 2006; Fernándeza et al, 2008; Wang et al, 2007; Shekaari and Mousavi, 2009; Ries et al, 2008). The properties of aqueous solutions of IL mixtures are important not only to technical and industrial applications of ILs, but also to examination of electrolyte theories.

Therefore, in this study the above-mentioned equations developed for prediction of the properties of mixed electrolyte solution were extended to the thermodynamic and transport properties of mixed IL solution. At the same time, the density and viscosity were measured for the ternary solutions $[C_2q]Br+[C_4q]Br+H_2O$, $[C_2q]Br+[C_6q]Br+H_2O$ and $[C_4q]Br+[C_6q]Br+H_2O$ and their binary subsystems $[C_2q]Br+H_2O$, $[C_4q]Br+H_2O$ and $[C_6q]Br+H_2O$ at different temperatures. The results were used to study the predictability of the well-known approaches.

2 Experimental section

All chemicals used in this study were of reagent grade with a claimed purity of >99%. Quinoline, *n*-C_2H_5Br, *n*-C_4H_9Br and *n*-$C_6H_{13}Br$ were supplied by Shanghai Jiachen Chemical Co., Ltd. (China), and used for the synthesis of $[C_2q]Br$, $[C_4q]Br$ and $[C_6q]Br$ ILs. Firstly, these three ILs were synthesized by reacting quinoline with corresponding *n*-alkylbromides for 2 days at 60, 70 and 70 °C, respectively. Then the reaction products were dissolved in ethyl acetate to extract the unreacted quinoline and *n*-alkylbromide. The obtained $[C_2q]Br$, $[C_4q]Br$, and $[C_6q]Br$ are all purple solids at room temperature. Their melting points are 120, 149 and 55 °C, respectively, which are determined by a TA Q2000 DSC (Differential Scanning Calorimeter). These purified ILs were vacuum-dried over $CaCl_2$ for several days at 70 °C before use. Their mass fraction purities were checked by ^1H NMR spectra interpretation using JEOL ECA-600 NMR spectrometer and found to be >99%. These three products have the following spectral properties:

• $[C_2q]Br$ (D_2O, ppm): δ = 9.233 (d, 1 H), 9.001 (d, 1 H), 8.363 (d, 1 H), 8.230 (d, 1 H), 8.155 (t, 1 H), 7.984 (t, 1 H), 7.900 (t, 1 H), 5.024 (m, 2 H), 1.677 (t, 3 H);

• $[C_4q]Br$ (D_2O, ppm): δ = 9.203 (d, 1 H), 8.985 (d, 1 H), 8.315 (d, 1 H), 8.194 (d, 1 H), 8.118 (t, 1 H), 7.971 (t, 1 H), 7.873 (t, 1 H), 4.948 (t, 2 H), 1.971 (m, 2 H), 1.408 (m, 2 H), 0.921 (t, 3 H);

• $[C_6q]Br$ (D_2O, ppm): δ = 9.219 (d, 1 H), 9.043 (d, 1 H), 8.355 (d, 1 H), 8.266 (d, 1 H), 8.170 (t, 1 H), 7.996 (t, 1 H), 7.932 (t, 1 H), 4.974 (t, 2 H), 1.993 (m, 2 H), 1.351 (m, 2 H), 1.206-1.245 (m, 4 H), 0.779 (t, 3 H).

The experimental procedures are similar to those used in our previous study (Hu et al, 2006b) and are described briefly as follows. The binary aqueous solutions were prepared with double-distilled deionized water and the ILs using a Sartorius CT225D balance with a precision of $\pm5\times10^{-5}$g. The ternary solutions were prepared from the binary solutions, and the uncertainty was $\pm5\times10^{-5}$mol·kg^{-1}. These solutions were placed into stoppered bottles and stirred for 2 h. All the solutions were prepared in glass flasks and measurements were carried out one week after preparation to ensure complete dissolution.

Densities of solutions were measured using a KEM oscillating-tube digital densimeter (DA-505) equiped with a thermostat with a precision of ± 0.01 °C (Hu et al, 2006b; Li et al, 2010). The temperature in the measuring cell was monitored with a digital thermometer. The densimeter was calibrated with double-distilled water and dry air (Hu et al, 2006b; Salabat and Alinoori, 2009). The density of water at different temperatures has been reported in the literature (Stokes and Mills, 1965; George and Sastry, 2004). The density of dry air at different temperatures was reported by Lemmon et al (2000). The uncertainty in density measurements was $\pm5\times10^{-5}$ g·cm^{-3}. DSC measurements were performed with a TA Q2000 DSC calorimeter, in the temperature range of 20–200 °C, using a heating rate of 5 °C·min^{-1}and helium as a purge gas. ^1H spectra were collected on a JEOL ECA-600 NMR spectrometer operating at 600.17 MHz. D_2O was used as the solvent and all the NMR experiments were performed at room temperature.

Viscosity was measured using a modified Cannon-Ubbelohde suspended level capillary viscometer (Li et al, 2010). A thoroughly cleaned and perfectly dried viscometer filled with liquid was placed vertically in a glass sided water thermostat (controlled to ±0.01 °C). After the specific temperature was achieved, the efflux time of the liquids

was recorded with a digital stop watch with a precision of ± 0.01 s. Two measurements were performed on each sample. The capillary viscometers are calibrated and credited by the company, and all the deviations were within 0.2%. In our initial measurements, the capillary viscometer was also calibrated with deionized water (0.8903 mPa·s at 25 °C) (Stokes and Mills, 1965).

3 Equations for prediction of density and viscosity of aqueous solutions of ionic liquids

In the following section, the variables with the superscript (o, I) together with the subscript M_iX_i were used to stand for the quantities of component M_iX_i in the binary solution M_iX_i+H_2O (i=1, 2, ⋯, n) having the same ionic strength as that of a mixed solution(i.e. ternary solution), and those without the superscript (o, I) are the corresponding quantities in the mixed solution.

The equation of Patwardhan and Kumar (Patwardhan and Kumar, 1986a; 1986b) can be expressed as

$$\rho = \sum_{i=1}^{n} Y_{M_iX_i} \Big/ \sum_{i=1}^{n} \left(Y_{M_iX_i} \Big/ \rho^{o,I}_{M_iX_i} \right) \tag{1}$$

with $Y_{M_iX_i} = y_{M_iX_i} + m_{M_iX_i} M_{M_iX_i}$, where y, m, ρ, and M is ionic strength fraction, molality, density, and molar mass, respectively.

Hu's equation (Hu, 2004) for the viscosity of a mixed IL solution can be expressed as

$$\ln \eta = \sum_{i=1}^{n} \frac{x_{M_iX_i}}{x^{o,I}_{M_iX_i}} \ln \eta^{o,I}_{M_iX_i} \tag{2}$$

where x is the mole fraction.

4 Comparisons with experimental data

The measured densities and viscosities were used to examine Eqs. (1) and (2), and the procedures are briefly summarized as follows:

1) The measured density and viscosity of the binary solutions is expressed respectively by the following polynomial equations

$$\rho^{o}_{M_iX_i(cal.)} = \sum_{l=0} A_l (m^{o}_{M_iX_i})^{l/2} \tag{3}$$

$$\eta^{o}_{M_iX_i(cal.)} = \sum_{l=0} B_l (m^{o}_{M_iX_i})^{l/2} \tag{4}$$

where $\rho^{o}_{M_iX_i(cal)}$, $\eta^{o}_{M_iX_i(cal)}$ and $m^{o}_{M_iX_i}$ is the calculated density, viscosity, and given molality of the binary solution M_iX_i + H_2O (i = 1, 2, ⋯, n), respectively. A_l (l = 0, 1, 2, 3⋯) and B_l (l = 0, 1, 2, 3⋯) are fitting parameters. The optimum fit was obtained by varying variable l until the values of $\delta^{o}_{\rho,M_iX_i} = \sum_{j=1}^{N} \left(\left| \rho^{o}_{M_iX_i(cal.)} - \rho^{o}_{M_iX_i(exp.)} \right| \Big/ \rho^{o}_{M_iX_i(exp.)} \right) \Big/ N$ and $\delta^{o}_{\eta,M_iX_i} = \sum_{j=1}^{N} \left(\left| \eta^{o}_{M_iX_i(cal.)} - \eta^{o}_{M_iX_i(exp.)} \right| \Big/ \eta^{o}_{M_iX_i(exp.)} \right) \Big/ N$ are all less than 10^{-4}. The values of A_l, B_l, δ^{o}_{ρ,M_iX_i}, and δ^{o}_{ρ,M_iX_i} for the examined binary solutions are shown in Table 1.

Table 1 The parameters for the binary systems [C_2q]Br+H_2O, [C_4q]Br+H_2O and [C_6q]Br+H_2O at different temperatures

Fitting parameters for the binary systems (density or viscosity) at different temperatures						
Parameters	15 °C	20 °C	25 °C	15 °C	20 °C	25 °C
	$\rho^{o}_{[C_2q]Br+H_2O}$			$\rho^{o}_{[C_4q]Br+H_2O}$		
A_0	1.0178	1.03106	1.01119	0.991562	1.0018	0.989773
A_1	−0.24629	−0.413088	−0.188506	0.0880342	−0.0400387	0.0910817
A_2	1.32054	2.08194	1.03227	−0.343202	0.221311	−0.378437
A_3	−3.17164	−4.92593	−2.44625	0.983064	−0.280946	1.0539
A_4	4.24695	6.40421	3.26091	−1.29531	0.225552	−1.34753
A_5	−2.88457	−4.23592	−2.20017	0.866778	−0.0714782	0.878467
A_6	0.777553	1.11649	0.587777	−0.233952	−0.00100146	−0.234039
δ^{o}_{ρ}	3.5×10^{-5}	3.3×10^{-5}	1.5×10^{-5}	3.7×10^{-5}	3.0×10^{-5}	4.8×10^{-5}
	$\rho^{o}_{[C_6q]Br+H_2O}$					
A_0	1.00581	1.00264	1.0061			
A_1	−0.0798252	−0.0677839	−0.115609			
A_2	0.45267	0.445679	0.636022			
A_3	−0.990379	−1.06553	−1.44902			
A_4	1.35359	1.53392	1.94382			

(To be continued)

(Continued)

Fitting parameters for the binary systems (density or viscosity) at different temperatures						
Parameters	15 °C	20 °C	25 °C	15 °C	20 °C	25 °C
A_5	−0.963483	−1.12157	−1.34477			
A_6	0.272756	0.32187	0.370638			
δ_ρ^o	3.9×10^{-5}	2.2×10^{-5}	4.8×10^{-5}			
	$\eta^o_{[C_2q]Br+H_2O}$			$\eta^o_{[C_4q]Br+H_2O}$		
B_0	4.57128	−1.51505	−1.08612	9.61771	9.25816	8.04698
B_1	−16.6622	11.4175	9.87000	−41.5039	−41.2827	−35.4645
B_2	30.3049	−19.1394	−18.2309	75.8043	76.8939	65.4717
B_3	−23.9798	14.4514	15.1185	−60.1681	−62.343	−52.6245
B_4	7.25677	−3.90678	−4.51787	18.0383	19.0096	15.9112
δ_η^o	2.7×10^{-5}	3.4×10^{-5}	2.6×10^{-5}	1.3×10^{-5}	1.1×10^{-5}	2.2×10^{-5}
	$\eta^o_{[C_6q]Br+H_2O}$					
B_0	20.1968	−3.98571	11.2642			
B_1	−93.1508	23.4816	−50.3854			
B_2	169.558	−40.5947	91.2337			
B_3	−134.969	31.5568	−72.1369			
B_4	40.4264	−8.68286	21.5528			
δ_η^o	4.5×10^{-5}	2.8×10^{-5}	1.8×10^{-5}			

2) The compositions ($m^{o,I}_{M_iX_i}$) of the binary solutions having the same ionic strength as that of the mixed solution of given molalities $m_{M_iX_i}$ ($i = 1, 2, \cdots, n$) were determined.

3) Substituting the values of $\rho^{o,I}_{M_iX_i}$ and $\eta^{o,I}_{M_iX_i}$ calculated from Eqs. (3) and (4) into Eqs. (1) and (2) to yield the predictions for the mixed solutions of given $m_{M_iX_i}$ (i = 1, 2, $\cdots$, n), then the predicted values were compared with the corresponding experimental data.

In this paper, the average relative errors between predicted and measured densities (δ_ρ) and viscosities (δ_η) of the mixed solution are as follows (Hu et al, 2006b):

$$\delta_\rho = \sum_{i=1}^{N} \left|\delta_{\rho,i}\right| \Big/ N \tag{5}$$

$$\delta_\eta = \sum_{i=1}^{N} \left|\delta_{\eta,i}\right| \Big/ N \tag{6}$$

with $\delta_{\rho,i} = (\rho_{i,(\text{cal.})} - \rho_{i,(\text{exp.})}) \big/ \rho_{i,(\text{exp.})}$ and

$$\delta_{\eta,i} = (\eta_{i,(\text{cal.})} - \eta_{i,(\text{exp.})}) \big/ \eta_{i,(\text{exp.})}$$

where N is the number of experiments.

5 Results and discussion

Tables 2-4 show the measured densities and viscosities of the binary solutions [C_2q]Br+H_2O, [C_4q]Br+H_2O and [C_6q] Br+H_2O at different temperatures.

Table 2 Density and viscosity of the binary system [C_2q]Br+H_2O at different temperatures

m_B mol·kg^{-1}	ρ_{15} g·cm^{-3}	ρ_{20} g·cm^{-3}	ρ_{25} g·cm^{-3}	η_{15} mPa·s	η_{20} mPa·s	η_{25} mPa·s
0.0500	1.0024	1.0015	1.0003			
0.1000	1.0058	1.0046	1.0037			
0.2000	1.0126	1.0115	1.0103			
0.3000	1.0189	1.0176	1.0166			
0.4500	1.0284	1.0271	1.0258	1.262	1.103	0.980
0.6000	1.0378	1.0363	1.035	1.315	1.155	1.021
0.7500	1.0466	1.0452	1.0437	1.377	1.207	1.067
0.9001	1.0549	1.0534	1.0519	1.442	1.265	1.119
1.0990	1.0660	1.0644	1.0626	1.546	1.351	1.187

Table 3 Density and viscosity of the binary system [C_4q]Br+H_2O at different temperatures

m_B mol·kg^{-1}	ρ_{15} g·cm^{-3}	ρ_{20} g·cm^{-3}	ρ_{25} g·cm^{-3}	η_{15} mPa·s	η_{20} mPa·s	η_{25} mPa·s
0.0500	1.0023	1.0013	1.0001			
0.1000	1.0057	1.0044	1.0031			
0.1999	1.0120	1.0108	1.0091			
0.3000	1.0182	1.0168	1.0149			
0.4499	1.0271	1.0256	1.0239	1.378	1.197	1.055
0.6000	1.0356	1.0341	1.0324	1.482	1.286	1.130
0.7501	1.0439	1.0422	1.0405	1.594	1.377	1.207
0.9001	1.0519	1.0502	1.0484	1.706	1.467	1.284
1.1001	1.0618	1.0600	1.0577	1.884	1.621	1.411

Table 4 Density and viscosity of the binary system $[C_6q]Br+H_2O$ at different temperatures

m_B mol·kg^{-1}	ρ_{15} g·cm^{-3}	ρ_{20} g·cm^{-3}	ρ_{25} g·cm^{-3}	η_{15} mPa·s	η_{20} mPa·s	η_{25} mPa·s
0.0500	1.0024	1.0011	1.0000			
0.1000	1.0053	1.0042	1.0029			
0.1999	1.0111	1.0100	1.0086			
0.2999	1.0169	1.0156	1.0142			
0.4504	1.0251	1.0237	1.0221	1.454	1.267	1.109
0.5989	1.0327	1.0312	1.0294	1.602	1.386	1.208
0.7495	1.0401	1.0384	1.0366	1.769	1.516	1.323
0.8997	1.0467	1.0449	1.0429	1.935	1.666	1.441
1.0998	1.0556	1.0536	1.0515	2.216	1.888	1.632

Tables 5-7 compare measured and predicted densities for the ternary solutions $[C_2q]Br+[C_4q]Br+H_2O$, $[C_2q]Br+[C_6q]Br+H_2O$ and $[C_4q]Br+[C_6q]Br+H_2O$ at different temperatures. The third to fifth columns show that the agreements are good; the $\delta_\rho^{Eq.1}$ values for the ternary solution $[C_2q]Br+[C_4q]Br+H_2O$ at 15, 20 and 25 °C are 2.0×10^{-4}, 2.1×10^{-4} and 1.5×10^{-4}, respectively. The $\delta_\rho^{Eq.1}$ values for the ternary solution $[C_2q]Br+[C_6q]Br+H_2O$ at 15, 20 and 25 °C are 2.8×10^{-4}, 2.7×10^{-4} and 2.2×10^{-4}, respectively. The $\delta_\rho^{Eq.1}$ values for the ternary solution $[C_4q]Br+[C_6q]Br+H_2O$ at 15, 20 and 25 °C are 2.5×10^{-4}, 2.3×10^{-4} and 2.5×10^{-4}, respectively.

Tables 8-10 compare measured and predicted viscosities for the ternary solutions $[C_2q]Br+[C_4q]Br+H_2O$, $[C_2q]Br+[C_6q]Br+H_2O$ and $[C_4q]Br+[C_6q]Br+H_2O$ at different temperatures. The third to fifth columns show that the agreements are also good; the $\delta_\eta^{Eq.2}$ values for the ternary solution $[C_2q]Br+[C_4q]Br+H_2O$ at 15, 20 and 25 °C are 3.3×10^{-3}, 3.6×10^{-3}, and 2.3×10^{-3}, respectively. The $\delta_\eta^{Eq.2}$ values for the ternary solution $[C_2q]Br+[C_6q]Br+H_2O$ at 15, 20 and 25 °C are 3.9×10^{-3}, 3.2×10^{-3} and 2.7×10^{-3}, respectively. The $\delta_\eta^{Eq.2}$ values for the ternary solution $[C_4q]Br+[C_6q]Br+H_2O$ at 15, 20 and 25 °C are 1.7×10^{-3}, 2.1×10^{-3} and 2.6×10^{-3}, respectively.

δ_ρ in Tables 5-7 and δ_η in Tables 8-10 are calculated as follows

$$\delta_\rho = \sum_{i=1}^{N} \frac{\left|\rho_{M_iX_i,(Eq.1)} - \rho_{M_iX_i,(exp.)}\right|}{\rho_{M_iX_i,(exp.)}} \Big/ N \quad \text{(In Tables 5-7)}$$

$$\delta_\eta = \sum_{i=1}^{N} \frac{\left|\eta_{M_iX_i,(Eq.2)} - \eta_{M_iX_i,(exp.)}\right|}{\eta_{M_iX_i,(exp.)}} \Big/ N \quad \text{(In Tables 8-10)}$$

Table 5 Comparisons of measured and predicted densities of the ternary system $[C_2q]Br(B)+[C_4q]Br(C)+H_2O(A)$ at different temperatures

m_B mol·kg^{-1}	m_C mol·kg^{-1}	ρ, g·cm^{-3} exp.	ρ, g·cm^{-3} cal. (by Eq.1)	$\rho_{i,(cal.)}-\rho_{i,(exp.)}$ g·cm^{-3}
		At 15 °C		
0.0200	0.0800	1.0056	1.0057	0.0001
0.0499	0.0501	1.0058	1.0058	0.0000
0.0800	0.0200	1.0061	1.0058	−0.0003
0.0599	0.2401	1.0178	1.0183	0.0005
0.1492	0.1508	1.0183	1.0186	0.0003
0.2392	0.0608	1.0191	1.0188	−0.0003
0.1197	0.4803	1.0361	1.0360	−0.0001
0.2977	0.3023	1.0369	1.0367	−0.0002
0.4775	0.1225	1.0376	1.0373	−0.0003
0.1797	0.7204	1.0525	1.0525	0.0000
0.4453	0.4548	1.0533	1.0534	0.0001
0.7142	0.1859	1.0545	1.0543	−0.0002
			δ_ρ	2.0×10^{-4}
		At 20 °C		
0.0200	0.0800	1.0045	1.0045	0.0000
0.0499	0.0501	1.0047	1.0045	−0.0002
0.0800	0.0200	1.0051	1.0046	−0.0005
0.0599	0.2401	1.0167	1.0170	0.0003
0.1492	0.1508	1.0172	1.0173	0.0001
0.2392	0.0608	1.0179	1.0175	−0.0004
0.1197	0.4803	1.0346	1.0345	−0.0001
0.2977	0.3023	1.0355	1.0352	−0.0003
0.4775	0.1225	1.0362	1.0359	−0.0003
0.1797	0.7204	1.0508	1.0508	0.0000
0.4453	0.4548	1.0517	1.0518	0.0001
0.7142	0.1859	1.0529	1.0527	−0.0002
			δ_ρ	2.1×10^{-4}
		At 25 °C		
0.0200	0.0800	1.0032	1.0032	0.0000
0.0499	0.0501	1.0036	1.0034	−0.0002
0.0800	0.0200	1.0038	1.0036	−0.0002
0.0599	0.2401	1.0150	1.0153	0.0003
0.1492	0.1508	1.0156	1.0158	0.0002
0.2392	0.0608	1.0161	1.0162	0.0001
0.1197	0.4803	1.0331	1.0329	−0.0002
0.2977	0.3023	1.0338	1.0337	−0.0001
0.4775	0.1225	1.0347	1.0345	−0.0002
0.1797	0.7204	1.0491	1.0491	0.0000
0.4453	0.4548	1.0501	1.0501	0.0000
0.7142	0.1859	1.0513	1.0512	−0.0001
			δ_ρ	1.5×10^{-4}

Table 6 Comparisons of measured and predicted densities of the ternary system $[C_2q]Br(B)+[C_6q]Br(C)+H_2O(A)$ at different temperatures

m_B mol·kg^{-1}	m_C mol·kg^{-1}	ρ, g·cm^{-3}		$\rho_{i,(cal.)}-\rho_{i,(exp.)}$
		exp.	cal. (by Eq.1)	g·cm^{-3}
		At 15 °C		
0.0200	0.0800	1.0052	1.0054	0.0002
0.0499	0.0501	1.0058	1.0055	−0.0003
0.0799	0.0201	1.0059	1.0057	−0.0002
0.0595	0.2404	1.0169	1.0173	0.0004
0.1489	0.1511	1.0175	1.0179	0.0004
0.2390	0.0610	1.0186	1.0185	−0.0001
0.1182	0.4810	1.0337	1.0338	0.0001
0.2957	0.3038	1.0355	1.0353	−0.0002
0.4761	0.1237	1.0370	1.0368	−0.0002
0.1769	0.7229	1.0489	1.0484	−0.0005
0.4406	0.4593	1.0512	1.0508	−0.0004
0.7085	0.1915	1.0537	1.0532	−0.0005
			δ_ρ	2.8×10^{-4}
		At 20 °C		
0.0200	0.0800	1.0041	1.0043	0.0002
0.0499	0.0501	1.0046	1.0044	−0.0002
0.0799	0.0201	1.0049	1.0045	−0.0004
0.0595	0.2404	1.0156	1.0160	0.0004
0.1489	0.1511	1.0161	1.0166	0.0005
0.2390	0.0610	1.0174	1.0173	−0.0001
0.1182	0.4810	1.0322	1.0323	0.0001
0.2957	0.3038	1.0340	1.0338	−0.0002
0.4761	0.1237	1.0355	1.0353	−0.0002
0.1769	0.7229	1.0471	1.0466	−0.0005
0.4406	0.4593	1.0495	1.0491	−0.0004
0.7085	0.1915	1.0519	1.0516	−0.0003
			δ_ρ	2.7×10^{-4}
		At 25 °C		
0.0200	0.0800	1.0030	1.0030	0.0000
0.0499	0.0501	1.0034	1.0033	−0.0001
0.0799	0.0201	1.0036	1.0035	−0.0001
0.0595	0.2404	1.0141	1.0146	0.0005
0.1489	0.1511	1.0149	1.0154	0.0005
0.2390	0.0610	1.0161	1.0161	0.0000
0.1182	0.4810	1.0306	1.0306	0.0000
0.2957	0.3038	1.0323	1.0322	−0.0001
0.4761	0.1237	1.0338	1.0339	0.0001
0.1769	0.7229	1.0451	1.0448	−0.0003
0.4406	0.4593	1.0478	1.0474	−0.0004
0.7085	0.1915	1.0506	1.0500	−0.0006
			δ_ρ	2.2×10^{-4}

Table 7 Comparisons of measured and predicted densities of the ternary system $[C_4q]Br(B)+[C_6q]Br(C)+H_2O(A)$ at different temperatures

m_B mol·kg^{-1}	m_C mol·kg^{-1}	ρ, g·cm^{-3}		$\rho_{i,(cal.)}-\rho_{i,(exp.)}$
		exp.	cal. (by Eq.1)	g·cm^{-3}
		At 15 °C		
0.0200	0.0800	1.0052	1.0054	0.0002
0.0499	0.0501	1.0054	1.0055	0.0001
0.0800	0.0200	1.0056	1.0056	0.0000
0.0596	0.2403	1.0168	1.0171	0.0003
0.1493	0.1506	1.0170	1.0175	0.0005
0.2393	0.0606	1.0179	1.0179	0.0000
0.1197	0.4794	1.0335	1.0333	−0.0002
0.2975	0.3019	1.0345	1.0342	−0.0003
0.4768	0.1230	1.0354	1.0350	−0.0004
0.1798	0.7199	1.0481	1.0478	−0.0003
0.4454	0.4545	1.0496	1.0493	−0.0003
0.7142	0.1858	1.0511	1.0508	−0.0003
			δ_ρ	2.5×10^{-4}
		At 20 °C		
0.0200	0.0800	1.0042	1.0042	0.0000
0.0499	0.0501	1.0044	1.0043	−0.0001
0.0800	0.0200	1.0045	1.0044	−0.0001
0.0596	0.2403	1.0153	1.0158	0.0005
0.1493	0.1506	1.0157	1.0162	0.0005
0.2393	0.0606	1.0166	1.0166	0.0000
0.1197	0.4794	1.0319	1.0318	−0.0001
0.2975	0.3019	1.0329	1.0326	−0.0003
0.4768	0.1230	1.0339	1.0335	−0.0004
0.1798	0.7199	1.0462	1.0460	−0.0002
0.4454	0.4545	1.0478	1.0475	−0.0003
0.7142	0.1858	1.0494	1.0491	−0.0003
			δ_ρ	2.3×10^{-4}
		At 25 °C		
0.0200	0.0800	1.0029	1.0029	0.0000
0.0499	0.0501	1.0032	1.0030	−0.0002
0.0800	0.0200	1.0033	1.0031	−0.0002
0.0596	0.2403	1.0139	1.0143	0.0004
0.1493	0.1506	1.0145	1.0146	0.0001
0.2393	0.0606	1.0151	1.0148	−0.0003
0.1197	0.4794	1.0302	1.0301	−0.0001
0.2975	0.3019	1.0313	1.0309	−0.0004
0.4768	0.1230	1.0322	1.0318	−0.0004
0.1798	0.7199	1.0443	1.0440	−0.0003
0.4454	0.4545	1.0460	1.0456	−0.0004
0.7142	0.1858	1.0476	1.0472	−0.0004
			δ_ρ	2.5×10^{-4}

Table 8 Comparison of measured and predicted viscosities of the ternary system $[C_2q]Br(B)+[C_4q]Br(C)+H_2O(A)$ at different temperatures

m_B mol·kg^{-1}	m_C mol·kg^{-1}	η, mPa·s exp.	η, mPa·s cal. (by Eq.2)	$\eta_{i,(cal.)}-\eta_{i,(exp.)}$ mPa·s
		At 15 °C		
0.1197	0.4803	1.452	1.447	−0.005
0.2977	0.3023	1.403	1.397	−0.006
0.4775	0.1225	1.341	1.348	0.007
0.1797	0.7204	1.648	1.650	0.002
0.4453	0.4548	1.576	1.570	−0.006
0.7142	0.1859	1.496	1.493	−0.003
			δ_η	3.3×10^{-3}
		At 20 °C		
0.1197	0.4803	1.259	1.259	0
0.2977	0.3023	1.227	1.219	−0.008
0.4775	0.1225	1.177	1.181	0.004
0.1797	0.7204	1.420	1.424	0.004
0.4453	0.4548	1.374	1.364	−0.01
0.7142	0.1859	1.303	1.305	0.002
			δ_η	3.6×10^{-3}
		At 25 °C		
0.1197	0.4803	1.105	1.107	0.002
0.2977	0.3023	1.074	1.074	0
0.4775	0.1225	1.043	1.042	−0.001
0.1797	0.7204	1.247	1.249	0.002
0.4453	0.4548	1.207	1.199	−0.008
0.7142	0.1859	1.154	1.151	−0.003
			δ_η	2.3×10^{-3}

Table 9 Comparison of measured and predicted viscosities of the ternary system $[C_2q]Br(B)+[C_6q]Br(C)+H_2O(A)$ at different temperatures

m_B mol·kg^{-1}	m_C mol·kg^{-1}	η, mPa·s exp.	η, mPa·s cal. (by Eq.2)	$\eta_{i,(cal.)}-\eta_{i,(exp.)}$ mPa·s
		At 15 °C		
0.1182	0.4810	1.548	1.541	−0.007
0.2957	0.3038	1.459	1.454	−0.005
0.4761	0.1237	1.374	1.370	−0.004
0.1769	0.7229	1.834	1.826	−0.008
0.4406	0.4593	1.686	1.676	−0.01
0.7085	0.1915	1.538	1.535	−0.003
			δ_η	3.9×10^{-3}
		At 20 °C		
0.1182	0.4810	1.340	1.337	−0.003
0.2957	0.3038	1.275	1.267	−0.008
0.4761	0.1237	1.194	1.200	0.006
0.1769	0.7229	1.579	1.578	−0.001
0.4406	0.4593	1.460	1.456	−0.004
0.7085	0.1915	1.345	1.342	−0.003
			δ_η	3.2×10^{-3}
		At 25 °C		
0.1182	0.4810	1.173	1.169	−0.004
0.2957	0.3038	1.113	1.112	–0.001
0.4761	0.1237	1.053	1.057	0.004
0.1769	0.7229	1.372	1.371	–0.001
0.4406	0.4593	1.279	1.273	–0.006
0.7085	0.1915	1.183	1.180	–0.003
			δ_η	2.7×10^{-3}

Table 10 Comparison of measured and predicted viscosities of the ternary system $[C_4q]Br(B)+[C_6q]Br(C)+H_2O(A)$ at different temperatures

m_B mol·kg^{-1}	m_C mol·kg^{-1}	η, mPa·s exp.	η, mPa·s cal. (by Eq.2)	$\eta_{i,(cal.)}-\eta_{i,(exp.)}$ mPa·s
		At 15 °C		
0.1197	0.4794	1.574	1.577	0.003
0.2975	0.3019	1.539	1.541	0.002
0.4768	0.1230	1.512	1.506	–0.006
0.1798	0.7199	1.889	1.887	–0.002
0.4454	0.4545	1.818	1.818	0
0.7142	0.1858	1.754	1.751	–0.003
			δ_η	1.7×10^{-3}
		At 20 °C		
0.1197	0.4794	1.361	1.365	0.004
0.2975	0.3019	1.333	1.335	0.002
0.4768	0.1230	1.310	1.306	–0.004
0.1798	0.7199	1.623	1.624	0.001
0.4454	0.4545	1.564	1.564	0
0.7142	0.1858	1.513	1.506	–0.007
			δ_η	2.1×10^{-3}
		At 25 °C		
0.1197	0.4794	1.195	1.192	–0.003
0.2975	0.3019	1.168	1.169	0.001
0.4768	0.1230	1.150	1.145	–0.005
0.1798	0.7199	1.414	1.408	–0.006
0.4454	0.4545	1.361	1.361	0
0.7142	0.1858	1.320	1.315	–0.005
			δ_η	2.6×10^{-3}

5 Conclusions

The simple equations for prediction of the density and viscosity of mixed electrolyte solutions were extended to the corresponding properties of mixed IL solutions. Their predictabilities have been tested by comparisons with the measured densities and viscosities of the ternary solutions $[C_2q]Br+[C_4q]Br+H_2O$, $[C_2q]Br+[C_6q]Br+H_2O$, and $[C_4q]Br+[C_6q]Br+H_2O$ at different temperatures. The comparison results show that Eqs. 1 and 2 can provide good predictions for the densities and viscosities of the ternary IL solutions from the data of their binary subsystems of equal ionic strength, which indicates that these simple equations can make full use of the information on the binary IL solutions, avoiding much complexity in the calculation of multicomponent thermodynamic and transport properties, and provide good predictions for the multicomponent IL solutions.

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