

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/268813010>

Composition and Temperature Dependence of Density, Surface Tension, and Viscosity of EMIM DEP/MMIM DMP + Water + 1-Propanol/2-Propanol Ternary Mixtures and Their Mathematical Repre...

ARTICLE in JOURNAL OF CHEMICAL & ENGINEERING DATA · AUGUST 2014

Impact Factor: 2.04 · DOI: 10.1021/je400576e

CITATIONS

2

READS

151

8 AUTHORS, INCLUDING:



Melissa Mazlan

University of Malaya

1 PUBLICATION 2 CITATIONS

SEE PROFILE



Asrina Sairi

University of Malaya

15 PUBLICATIONS 57 CITATIONS

SEE PROFILE



Yatimah Alias

University of Malaya

169 PUBLICATIONS 1,171 CITATIONS

SEE PROFILE



Asrul Farrish Udaiyappan

1 PUBLICATION 2 CITATIONS

SEE PROFILE

Composition and Temperature Dependence of Density, Surface Tension, and Viscosity of EMIM DEP/MMIM DMP + Water + 1-Propanol/2-Propanol Ternary Mixtures and Their Mathematical Representation Using the Jouyban–Acree Model

Wan Melissa Diyana Wan Normazlan,[†] Nor Asrina Sairi,^{*,†} Yatimah Alias,[†] Asrul Farrish Udaiyappan,[‡] Abolghasem Jouyban,[§] and Mehry Khoubnasabjafari^{||}

[†]Chemistry Department, Faculty of Science, University of Malaya, 50603 Kuala Lumpur, Malaysia

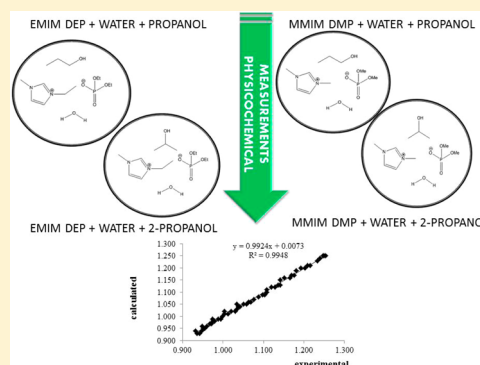
[‡]Chemistry Department, Faculty of Science, University Putra Malaysia, 43400, Serdang, Selangor, Malaysia

[§]Faculty of Pharmacy, Tabriz University of Medical Sciences, 51664, Tabriz, Iran

^{||}Faculty of Medicine, Tabriz University of Medical Sciences, 51664, Tabriz, Iran

S Supporting Information

ABSTRACT: Room temperature ionic liquids (RTILs) have been ascribed as alternative solvents in separation processes or chemical reactions. This research is concerned with the study of density, ρ , viscosity, η , and surface tension, σ , over the mole fractions of (0.1000 to 1.000) mol and temperature from (293.15 to 333.15) K for ternary mixtures of 1-ethyl-3-methylimidazolium diethylphosphate (EMIM DEP)/1,3-dimethylimidazolium dimethylphosphate (MMIM DMP), water, and 1-propanol/2-propanol. As the temperature increased, surface tension and density results for all the multicomponent mixtures show a linear descending trend. Adversely, viscosity results indicate polynomial descending trend. At the whole ranges of temperature, the density, surface tension and viscosity data show a significant gap between mole fractions of ionic liquid. These experimental results have been evaluated and the most prominent polynomial or linear regressions were obtained. The most prominent correlation for density and surface tension for all four systems were obtained using a linear equation. In contrast, the best correlation for viscosity data was obtained using a second order polynomial equation. On the other view, the experimental density and surface tension data decrease linearly with mole fraction of ionic liquid. The Jouyban–Acree model was used to correlate the density, surface tension, and viscosity of the studied mixtures at different temperatures. The accuracy of the model was evaluated and the absolute percentage error (APER) for each correlation was less than 6%.



1. INTRODUCTION

Room temperature ionic liquids (RTILs) possess a number of special characteristics such as nonvolatility, thermal and chemical stable, low vapor pressures, encouraging solubility, and many more^{1,2} which may contribute in their application as separating agents in liquid–liquid extraction processes. These features play a significant role in liquid–liquid extraction,^{3–7} carbon dioxide adsorption,^{8–10} electrochemistry,^{11–13} and other areas.^{14–16}

Azeotropic mixtures such as aqueous solutions of 1-propanol and 2-propanol have many uses in major industries but predominantly they are used as solvent media for an array of separation processes and also in homogeneous and heterogeneous extractive rectification.¹⁷ As it is known, an azeotropic mixture is one tough solution to be separated from one to another. Hence forward, based on the number of paper published,^{18–20} dialkyl-phosphate based ionic liquids are a probable separating agent for the separation process. However, in order to introduce the ionic liquid as an entrainer into both azeotropic aqueous solutions, the demand of fundamental

physicochemical properties such as density, viscosity, and surface tension and their dependency upon composition and temperature of ternary mixture is very important.

Nowadays, studies on separation of organic solvents in mixtures using an ionic liquid have become a great part in the research world.^{21–24} These physicochemicals behavior could significantly effect the mass transfer or energy across the interface. In addition, the study of physicochemical behaviors of RTIL binary or ternary mixtures are more widely used and appropriate compared to pure ionic liquid for some applications. For example, Dong et al.²⁴ reported that mixed electrolytes formed by the combination of ionic liquids and standard liquid electrolyte improved the thermal stability in lithium-ion battery. In another field, Anderson et al.²⁵ studied binary mixtures of two ionic liquids as gas chromatography stationary phases. Based on

Received: June 17, 2013

Accepted: July 7, 2014

Published: July 17, 2014

their observation, the enrichment of the IL-mixture with chloride anion produced a stationary phase with improved dipole-type and hydrogen bond basicity interactions. On the other hand, the separation selectivity of a mixture of alcohols and aromatic analytes was enhanced by tuning the composition of the binary ILs stationary phase mixture.

Physicochemical data for azeotropic systems such as in this work, i.e., 1-propanol + water and 2-propanol + water containing ionic liquids are very important in order for us to have a better understanding in designing separation mechanism. In this study, 1-propanol + water and 2-propanol + water were chosen as the azeotropic mixtures based on the reported observation by Zhang et al.²⁶ at which their result indicated that the used ionic liquid was able to reduce the vapor pressure of water, 1-propanol, and 2-propanol, making it a potential azeotropic mixture to be extracted from one another. Dialkyl phosphate anions are anticipated for practical applications as it can be processed in a single reactor under gentle conditions and still can give a very high yield. On the other hand, this type of ionic liquid is ecological friendly compared to other ionic liquids.²⁷

Hence, in this research we have measured the surface tension, density, and viscosity of the four systems, i.e., 1-ethyl-3-methylimidazolium diethylphosphate (EMIM DEP)/water/1-propanol, 1-ethyl-3-methylimidazolium diethylphosphate (EMIM DEP)/water/2-propanol, 1,3-dimethylimidazolium dimethylphosphate (MMIM DMP)/water/1-propanol, and 1,3-dimethylimidazolium dimethylphosphate (MMIM DMP)/water/2-propanol over the whole concentration range at temperatures from (293.15 to 333.15) K. The trending of density, surface tension and viscosity toward temperature and RTIL compositions of these two types of imidazolium-phosphate based RTILs and primary and secondary alcohol were observed. In an almost similar study, Li and Wang²⁸ measured vapor–liquid (VLE) including 1-propanol + EMIM DEP, 2-propanol + EMIM DEP, water +1-propanol + EMIM DEP, and water +2-propanol + EMIM DEP. The experimental data were then regressed using UNIFAC model with the maximum average relative deviation (ARD) of 2.7%. In their study, EMIM DEP was divided into MMIM DMP. Based on the reported result, both EMIM DEP and MMIM DMP were able to separate and break the azeotropic behavior of alcohols and water mixtures. As an addition, MMIM DMP is a better separating agent for the azeotrope mixture studied than EMIM DEP.

Regardless of all the experimental efforts for physicochemical studies, there are numerous mathematical methods^{29–32} that were reported to compute physicochemical data. The density, viscosity, and surface tension of these ternary mixtures are correlated using the Jouyban–Acree model; and the accuracy of the model is evaluated using absolute percentage error (APER) of the correlated and experimental values.

2. EXPERIMENTAL SECTION

The analytical commercial grade chemicals, i.e., 1-propanol, 2-propanol, 1-ethyl-3-methylimidazolium diethylphosphate (EMIM DEP), and 1,3-dimethylimidazolium dimethylphosphate (MMIM DMP) were supplied by Merck Chemicals and used without further purification. The water content for all four commercial grade chemicals was determined using Karl Fischer. The purity and water content results for all chemicals used are represented in Table 1.

Density, ρ , measurements were carried out using an Anton Paar DMA 4500. The densitometer is precise within $1.0 \cdot 10^{-4} \text{ g} \cdot \text{cm}^{-3}$ and the uncertainty measurement was estimated to be better

Table 1. Purity and Water Content of the Commercial Grade Chemical Used

chemical name	source	reported mole fraction purity	water content
			(w/w %)
1-ethyl-3-methylimidazolium diethylphosphate	Merck Chemicals	0.952	0.34
1,3-dimethylimidazolium dimethylphosphate	Merck Chemicals	0.967	0.28
1-propanol	Merck Chemicals	0.986	0.41
2-propanol	Merck Chemicals	0.985	0.44

Table 2. Mole Fraction of Each Component for EMIM DEP/Water/1-Propanol or 2-Propanol

EMIM DEP/water/1-propanol		
$X_{\text{EMIM DEP}}$	X_{water}	$X_{1\text{-propanol}}$
0.1033	0.4497	0.4470
0.2025	0.3985	0.3990
0.3039	0.3491	0.3470
0.4020	0.3040	0.2940
0.5051	0.2429	0.2520
0.6056	0.1954	0.1990
0.7013	0.1487	0.1500
0.8026	0.0994	0.0980
0.9033	0.0487	0.0480
EMIM DEP/water/2-propanol		
$X_{\text{EMIM DEP}}$	X_{water}	$X_{2\text{-propanol}}$
0.1000	0.4510	0.4490
0.2000	0.4000	0.4000
0.3005	0.3505	0.3490
0.4010	0.3000	0.2990
0.5013	0.2497	0.2490
0.6002	0.2008	0.1990
0.7002	0.1498	0.1500
0.8012	0.0998	0.0990
0.9001	0.0499	0.0500

than $\pm 1.0 \cdot 10^{-3} \text{ g} \cdot \text{cm}^{-3}$. The instrument is equipped with a maximum temperature range of 363.15 K and a minimum of 273.15 K. Calibration of the densitometer was performed at atmospheric pressure using dry air and 1-propanol or 2-propanol.

On the other hand, surface tension, σ , measurements were carried out using KRÜSS Processor Tensiometer K100 using Du Noüy ring method at temperature from (293.15 to 333.15) K. The tensiometer has a surface tension range of (1–1000) $\text{mN} \cdot \text{m}^{-1}$ with 0.01 resolutions. The instrument is equipped with a maximum temperature range of 403.15 K and a minimum of 263.15 K. Calibration of the tensiometer was performed at atmospheric pressure using dry air and pure alcohol (1-propanol or 2-propanol). The reproducibility of the surface tension is 0.50 %. In general, each surface tension value reported was an average of ten measurements.

Viscosity, η , measurements were carried out using Brookfield R/S+ Rheometer. The rheometer has a dynamic viscosity range of (0.002–19) Pa·s. The instrument is equipped with a maximum temperature range of 453.15 K and a minimum of 293.15 K. Calibration of the rheometer was performed at atmospheric pressure using dry air and pure alcohol (1-propanol or 2-propanol). The viscosity was measured with an accuracy less than 1 %.

The mixtures of water, alcohol, and RTIL were prepared by weighing on an AND GR-200 balance covering the complete

Table 3. Mole Fraction of Each Component for MMIM DMP/Water/1-Propanol or 2-Propanol

MMIM DMP/water/1-propanol		
$X_{\text{MMIM DMP}}$	X_{water}	$X_{1\text{-propanol}}$
0.1089	0.4411	0.4500
0.2062	0.3938	0.4000
0.3059	0.3441	0.3500
0.4022	0.2978	0.3000
0.5059	0.2451	0.2490
0.6002	0.1998	0.2000
0.7002	0.1498	0.1500
0.8002	0.0998	0.1000
0.9028	0.0472	0.0500
MMIM DMP/water/2-propanol		
$X_{\text{MMIM DMP}}$	X_{water}	$X_{2\text{-propanol}}$
0.1000	0.4450	0.4450
0.2000	0.4030	0.3970
0.3005	0.3525	0.3470
0.4010	0.3050	0.2940
0.5013	0.2507	0.2480
0.6002	0.1998	0.2000
0.7002	0.1518	0.1480
0.8012	0.0988	0.1000
0.9130	0.0430	0.0440

composition range; the precision in mass fraction being estimated as $\pm 10^{-4}$ g. The experimental mole fractions for each component are represented in Tables 2 and 3. The mixtures were placed into stoppered bottles and stirred for 1 h at room temperature. All the samples prepared and the pure liquids were measured at (298.15 to 333.15) K for density, viscosity, and surface tension reading. The measured values of surface tension, viscosity, and density for pure 1-propanol, 2-propanol, and water over the temperature range of (293.15 to 333.15) K are shown in Table 4. Table 5 represents the surface tension, density and viscosity experimental values of pure ionic liquids used in this work.

3. RESULTS AND DISCUSSION

The density, surface tension, and viscosity data for all the ternary mixtures, EMIM DEP/water/1-propanol or 2-propanol and MMIM DMP/water/1-propanol or 2-propanol, at IL mole fraction from (0.1000 to 0.9000) and at temperature from (293.15 to 333.15) K are tabulated in Tables 6–11.

Density. Figures S1 to S4 (in the Supporting Information) illustrate linear trending for density measurements of all the ternary mixtures as a function of temperature and composition. The results indicate that the density values decrease linearly with temperature and composition of ionic liquid. The density for EMIM DEP + water + 1-propanol is slightly lower than that for EMIM DEP + water + 2-propanol. This shows that the secondary alcohol has a slightly higher density compared to the primary alcohol even in mixture form. The same behavior was observed for MMIM DMP + water + 1-propanol and MMIM DMP + water + 2-propanol ternary mixtures. The density results were then correlated and the test correlation was obtained ($R^2 > 0.99$) using linear equation. This linear trend between density of imidazolium and pyridinium chloroaluminate and temperature shows the same trend that was observed and reported by Siodlak et al.⁴³

The experimental densities of ternary mixtures of EMIM DEP and MMIM DMP with aqueous 1-propanol and 2-propanol at different temperatures as a function of mole fractions are depicted in Figure S5 to S8 (refer Supporting Information). As shown, the density increases with increasing of mole fractions of ionic liquids. This observed result has been quite consistent by the existing result reported by Hofman et al.⁴⁴ According to their observation, the density increased due to stronger intermolecular interactions between the two components in the studied binary mixture.

Surface Tension. In all systems studied, surface tension generally decreased with increasing temperature for any given concentration or mole fractions of IL. Figures S9 to S12 (in the Supporting Information) show the linear trending of surface tension values with the temperature for EMIM DEP + water + 1-propanol/2-propanol and MMIM DMP + water + 1-propanol/2-propanol. This declining trend shows that the molecular

Table 4. Experimental and Literature Values of Density, Surface Tension, and Viscosity of Pure 1-Propanol and 2-Propanol^a

1-propanol							
T	p	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\sigma/\text{mN}\cdot\text{m}^{-1}$		$\eta/\text{mPa}\cdot\text{s}$	
K	atm	exp.	lit.	exp.	lit.	exp.	lit.
293.15	0.987	0.804	0.804 ^b	23.67	23.71 ^c	2.20	2.20 ^b
298.15	0.987	0.8001	0.800 ^b	23.31	23.33 ^d	1.99	1.95 ^b
303.15	0.987	0.796	0.796 ^b	22.81	22.93 ^c	1.73	1.73 ^b
313.15	0.987	0.788	0.789 ^b	22.19	22.15 ^c	1.38	1.38 ^b
323.15	0.987	0.780	0.781 ^b	21.34	21.38 ^c	1.12	1.11 ^b
333.15	0.987	0.764	0.773 ^b	20.66	20.60 ^c	0.91	0.91 ^b
2-propanol							
T	p	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\sigma/\text{mN}\cdot\text{m}^{-1}$		$\eta/\text{mPa}\cdot\text{s}$	
K	atm	exp.	lit.	exp.	lit.	exp.	lit.
293.15	0.987	0.786	0.785 ^b	21.28	21.32 ^c	2.41	2.41 ^b
298.15	0.987	0.781	0.781 ^b	20.94	20.90 ^e	2.06	2.07 ^b
303.15	0.987	0.777	0.777 ^b	20.54	20.53 ^c	1.79	1.79 ^b
313.15	0.987	0.768	0.769 ^b	19.97	19.74 ^c	1.35	1.35 ^b
323.15	0.987	0.759	0.760 ^b	18.87	18.96 ^c	1.03	1.03 ^b
333.15	0.987	0.750	0.750 ^b	18.11	18.17 ^c	0.82	0.81 ^b

^aStandard uncertainties, u , are $u(T) = 0.01$ K, $u(x) = 0.0001$, $u(P) = 1$ kPa, and $u(\sigma) = 0.15$ mN·m⁻¹. Relative standard uncertainties, u_r , are $u_r(\rho) = 0.001$ and $u_r(\eta) = 0.01$. ^bReference 33. ^cReference 34. ^dReference 35. ^eReference 36.

Table 5. Experimental and Literature Values of Density, Surface Tension, and Viscosity of Pure (EMIM DEP) and (MMIM DMP)^a

EMIM DEP							
<i>T</i>	<i>p</i>	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\sigma/\text{mN}\cdot\text{m}^{-1}$		$\eta/\text{mPa}\cdot\text{s}$	
K	atm	exp.	lit.	exp.	lit.	exp.	lit.
293.15	0.987	1.151	1.147 ^b 1.141 ^c	34.78	36.10 ^c	537.03	580 ^f
298.15	0.987	1.146	1.146 ^b 1.149 ^d 1.140 ^c 1.145 ^d 1.149 ^e	34.46	35.58 ^c 37.1 ^e	320.89	410 ^f
303.15	0.987	1.141	1.140 ^b 1.132 ^c 1.142 ^d	34.19	35.43 ^c	284.34	300 ^f
313.15	0.987	1.130	1.134 ^b 1.125 ^c 1.135 ^d	33.54	35.07 ^c	150.96	168 ^f
323.15	0.987	1.116	1.126 ^b 1.118 ^c 1.129 ^d	33.15	34.55 ^c	90.02	101 ^f
333.15	0.987	1.103	1.111 ^c 1.122 ^d	32.81	33.88 ^c	58.02	66 ^f
MMIM DMP							
<i>T</i>	<i>p</i>	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\sigma/\text{mN}\cdot\text{m}^{-1}$		$\eta/\text{mPa}\cdot\text{s}$	
K	atm	exp.	lit.	exp.	lit.	exp.	lit.
293.15	0.987	1.254	1.262 ^b 1.244 ^c	47.78	47.42 ^c	381.22	NA
298.15	0.987	1.250	1.258 ^b 1.242 ^c 1.253 ^e	46.98	47.09 ^c 48.4 ^e	336.74	NA
303.15	0.987	1.247	1.255 ^b 1.234 ^c 1.253 ^g	46.21	47.11 ^c	221.41	NA
313.15	0.987	1.240	1.248 ^b 1.230 ^c 1.246 ^g	44.89	45.11 ^c	178.66	NA
323.15	0.987	1.233	1.242 ^b 1.224 ^c 1.232 ^g	42.67	43.01 ^c	147.95	NA
333.15	0.987	1.230	1.219 ^c 1.232 ^g	41.99	41.88 ^c	133.73	NA

^aStandard uncertainties, u , are $u(T) = 0.01$ K, $u(x) = 0.0001$, $u(P) = 1$ kPa, and $u(\sigma) = 0.15$ mN·m⁻¹. Relative standard uncertainties, u_r , are $u_r(\rho) = 0.001$ and $u_r(\eta) = 0.01$. ^bReference 37. ^cReference 38. ^dReference 39. ^eReference 40. ^fReference 41. ^gReference 42.

interaction between the liquids is weaker. The alcohol hydrogen bond is the main contribution for association between two molecules which are very weakly bound. Therefore, the bond can be easily broken upon increasing temperature leading to lower surface tension values.

Similar to density, the surface tension value for MMIM DMP/water/1-propanol is higher compared to that for MMIM DMP/water/2-propanol. The same goes for the EMIM DEP mixture. MMIM DMP shows a very distinct result unlike EMIM DEP where the gap in the surface tension results between each composition is closer. This shows that the surfactant effect is distinctive in systems containing MMIM DMP rather than EMIM DEP. The experimental results were then correlated, and the best correlation was obtained ($R^2 > 0.99$) using linear equations. This linear trend behavior of surface tension is in agreement with Coutinho et al.⁴⁵ and Zobeydi.⁴⁶

Figures S13 to S16 (in the Supporting Information) depict the surface tension of all four studied mixtures in different temperatures as a function of mole fraction. It shows from the trend that ternary mixtures with EMIM DEP as an ionic liquid have lower surface tension values compared to mixtures with MMIM DMP. This behavior has been proven by Khanjari et al.⁴⁷ where the surface tension varied strongly on the alkyl chain length of quaternary ammonium-based ionic liquids.

Viscosity. Figures S17 to S20 (in the Supporting Information) show the variation in viscosities of EMIM DEP + water + 1-propanol/2-propanol and MMIM DMP + water + 1-propanol/2-propanol with composition and temperature. The viscosity decreases nonlinearly with temperature. At a fixed temperature, the dynamic viscosity values of the ternary mixtures decrease with increasing mole fraction of ionic liquid. Similarly to density and surface tension, the viscosity for EMIM DEP + water + 2-propanol is distinctively lower than that for EMIM DEP + water +

Table 6. Density, ρ ($\text{g}\cdot\text{cm}^{-3}$), of EMIM DEP/Water/1-Propanol and EMIM DEP/Water/2-Propanol at 0.987 atm^a

T	density, $\rho/\text{g}\cdot\text{cm}^{-3}$								
	$X_{\text{EMIM DEP}}$ in EMIM DEP/water/1-propanol								
K	0.1033	0.2025	0.3039	0.4020	0.5051	0.6056	0.7013	0.8026	0.9033
293.15	0.937	0.954	0.973	0.992	1.015	1.041	1.067	1.089	1.108
298.15	0.933	0.950	0.967	0.988	1.009	1.033	1.060	1.084	1.103
303.15	0.930	0.946	0.961	0.982	1.002	1.027	1.053	1.079	1.098
313.15	0.923	0.938	0.952	0.971	0.993	1.013	1.041	1.067	1.088
323.15	0.914	0.929	0.943	0.962	0.981	1.001	1.029	1.055	1.077
333.15	0.904	0.920	0.935	0.950	0.972	0.991	1.018	1.043	1.065
T	$X_{\text{EMIM DEP}}$ in EMIM DEP/water/2-propanol								
K	0.1000	0.2000	0.3005	0.4010	0.5013	0.6002	0.7002	0.8012	0.9001
293.15	0.939	0.958	0.985	1.006	1.033	1.061	1.088	1.118	1.137
298.15	0.936	0.955	0.981	1.002	1.028	1.056	1.082	1.112	1.122
303.15	0.933	0.951	0.977	0.997	1.021	1.050	1.076	1.106	1.116
313.15	0.925	0.944	0.967	0.989	1.012	1.038	1.065	1.095	1.108
323.15	0.918	0.936	0.957	0.979	1.001	1.025	1.053	1.085	1.101
333.15	0.910	0.928	0.949	0.969	0.990	1.012	1.040	1.071	1.092

^aStandard uncertainties, u , are $u(T) = 0.01$ K, $u(x) = 0.0001$, $u(P) = 1$ kPa, and $u(\sigma) = 0.15$ mN·m⁻¹. Relative standard uncertainties, u_r , are $u_r(\rho) = 0.001$ and $u_r(\eta) = 0.01$.

1-propanol. The viscosity decreases quicker for a secondary alcohol than for a primary alcohol. This could be due to the branch alcohol, i.e., 2-propanol becomes a linear alcohol and readily linear alcohol's molecules, i.e., 1-propanol becomes stronger than it usually does. Higher viscosity values were observed for mixtures containing MMIM DMP compared to mixtures containing EMIM DEP. The best correlation values for all mixtures were obtained using a second order polynomial equation ($R^2 > 0.99$). The results corresponded to those of Maik et al.⁴⁸

Figures S21 to S24 (in the Supporting Information) present the viscosity data of ternary mixtures of EMIM DEP + water + 1-propanol/2-propanol and MMIM DMP + water + 1-propanol/2-propanol at different temperatures as a function of mole fraction. At higher mole fraction of ionic liquids, the gaps between each mole fraction of ionic liquids are more obvious and clear compared to those for lower ionic liquids mole fraction. It is understandable that the mobility of the ions is higher when the mixtures have lower viscosity. This phenomenon is in agreement with Laskowska and Domanska⁴⁹ where their study showed a decrease of viscosity with an increase of alcohol content that is significantly strong in dilute solutions of an alcohol in studied ionic liquids.

Data Correlation Using the Jouyban–Acree Model. The experimental density, surface tension, and viscosity data were then correlated using Jouyban–Acree model. The model was provided reasonably accurate results for various physico-chemical properties of the mixtures.⁵⁰

Density. The Jouyban–Acree model for representing the density of ternary mixtures is

$$\ln \rho_{m,T} = x_1 \ln \rho_{1,T} + x_2 \ln \rho_{2,T} + x_3 \ln \rho_{3,T} + x_1 x_2 \sum_{j=0}^2 \left[\frac{A_j (x_1 - x_2)^j}{T} \right] + x_1 x_3 \times \sum_{j=0}^2 \left[\frac{B_j (x_1 - x_3)^j}{T} \right] + x_2 x_3 \sum_{j=0}^2 \left[\frac{C_j (x_2 - x_3)^j}{T} \right] + x_1 x_2 x_3 \sum_{j=0}^2 \left[\frac{D_j (x_1 - x_2 - x_3)^j}{T} \right] \quad (1)$$

where $\rho_{m,T}$, $\rho_{1,T}$, $\rho_{2,T}$, and $\rho_{3,T}$ are the densities of the mixtures and solvents 1, 2, and 3 at temperature T , respectively, and D_j represent the model constants.^{51,52} These model constants are computed by regressing $(\ln \rho_{m,T} - x_1 \ln \rho_{1,T} - x_2 \ln \rho_{2,T} - x_3 \ln \rho_{3,T})$ against $x_1 x_2 / T$, $x_1 x_2 (x_1 - x_2) / T$, $x_1 x_2 (x_1 - x_2)^2 / T$, $x_1 x_3 / T$, $x_1 x_3 (x_1 - x_3) / T$, $x_1 x_3 (x_1 - x_3)^2 / T$, $x_2 x_3 / T$, $x_2 x_3 (x_2 - x_3) / T$, $x_2 x_3 (x_2 - x_3)^2 / T$, $x_1 x_2 x_3 / T$, $x_1 x_2 x_3 (x_1 - x_2 - x_3) / T$, and $x_1 x_2 x_3 (x_1 - x_2 - x_3)^2 / T$ using a no intercept least-square analysis.

The proposed model after excluding nonsignificant model constants ($p > 0.05$), for

(i) EMIM DEP/water/1-propanol mixture:

$$\ln \rho_{m,T} = x_1 \ln \rho_{1,T} + x_2 \ln \rho_{2,T} + x_3 \ln \rho_{3,T} - 34.106 \left[\frac{x_1 x_2}{T} \right] + 35.221 \left[\frac{x_2 x_3}{T} \right] \quad (2)$$

(ii) EMIM DEP/water/2-propanol mixture:

$$\ln \rho_{m,T} = x_1 \ln \rho_{1,T} + x_2 \ln \rho_{2,T} + x_3 \ln \rho_{3,T} + 79.955 \left[\frac{x_1 x_2}{T} \right] + 55.147 \left[\frac{x_2 x_3}{T} \right] - 219.836 \left[\frac{x_1 x_2 x_3}{T} \right] \quad (3)$$

(iii) MMIM DMP/water/1-propanol mixture:

$$\ln \rho_{m,T} = x_1 \ln \rho_{1,T} + x_2 \ln \rho_{2,T} + x_3 \ln \rho_{3,T} - 165.729 \left[\frac{x_1 x_3}{T} \right] + 22.598 \left[\frac{x_2 x_3}{T} \right] + 549.242 \left[\frac{x_1 x_2 x_3}{T} \right] \quad (4)$$

(iv) MMIM DMP/water/2-propanol system:

$$\ln \rho_{m,T} = x_1 \ln \rho_{1,T} + x_2 \ln \rho_{2,T} + x_3 \ln \rho_{3,T} - 7.335 \left[\frac{x_1 x_3}{T} \right] + 57.906 \left[\frac{x_2 x_3}{T} \right] \quad (5)$$

Table 7. Density, ρ ($\text{g}\cdot\text{cm}^{-3}$), of MMIM DMP/water/1-propanol and MMIM DMP/water/2-propanol at 0.987 atm^a

T	density, $\rho/\text{g}\cdot\text{cm}^{-3}$								
	$X_{\text{MMIM DMP}}$ in MMIM DMP/water/1-propanol								
K	0.1089	0.2062	0.3059	0.4022	0.5059	0.6002	0.7002	0.8002	0.9028
293.15	0.965	0.988	1.017	1.044	1.071	1.096	1.123	1.148	1.193
298.15	0.960	0.980	1.010	1.037	1.062	1.087	1.116	1.139	1.182
303.15	0.953	0.974	1.002	1.030	1.054	1.080	1.107	1.131	1.174
313.15	0.942	0.963	0.989	1.016	1.042	1.067	1.094	1.117	1.162
323.15	0.934	0.952	0.976	1.004	1.031	1.056	1.083	1.105	1.150
333.15	0.925	0.941	0.964	0.990	1.020	1.043	1.071	1.094	1.137
T	$X_{\text{MMIM DMP}}$ in MMIM DMP/water/2-propanol								
	$X_{\text{MMIM DMP}}$ in MMIM DMP/water/2-propanol								
K	0.1000	0.2000	0.3005	0.4010	0.5013	0.6002	0.7002	0.8012	0.9130
293.15	0.955	0.975	1.003	1.038	1.069	1.108	1.142	1.176	1.215
298.15	0.951	0.971	0.999	1.034	1.063	1.103	1.136	1.169	1.209
303.15	0.947	0.968	0.995	1.030	1.058	1.098	1.129	1.165	1.203
313.15	0.942	0.960	0.988	1.020	1.051	1.088	1.120	1.152	1.192
323.15	0.936	0.953	0.979	1.012	1.042	1.076	1.108	1.141	1.181
333.15	0.932	0.948	0.974	1.003	1.034	1.067	1.098	1.130	1.171

^aStandard uncertainties, u , are $u(T) = 0.01$ K, $u(x) = 0.0001$, $u(P) = 1$ kPa, and $u(\sigma) = 0.15$ mN·m⁻¹. Relative standard uncertainties, u_r , are $u_r(\rho) = 0.001$ and $u_r(\eta) = 0.01$.

Table 8. Surface Tension, $\sigma/\text{mN m}^{-1}$ of EMIM DEP/Water/1-Propanol and EMIM DEP in EMIM DEP/Water/2-Propanol at 0.987 atm^a

T	surface tension, $\sigma/\text{mN}\cdot\text{m}^{-1}$								
	$X_{\text{EMIM DEP}}$ in EMIM DEP/water/1-propanol								
K	0.1033	0.2025	0.3039	0.4020	0.5051	0.6056	0.7013	0.8026	0.9033
293.15	22.75	24.15	25.37	26.39	27.51	28.47	29.52	30.72	32.39
298.15	22.45	23.77	25.08	26.02	27.12	28.17	29.18	30.43	31.89
303.15	22.14	23.49	24.76	25.76	26.78	27.85	28.84	30.16	31.49
313.15	21.71	23.06	24.16	25.24	26.32	27.33	28.15	29.53	30.87
323.15	21.31	22.63	23.77	24.89	25.89	26.78	27.62	29.03	30.35
333.15	21.07	22.25	23.57	24.49	25.37	26.28	27.22	28.63	29.97
T	$X_{\text{EMIM DEP}}$ in EMIM DEP/water/2-propanol								
	$X_{\text{EMIM DEP}}$ in EMIM DEP/water/2-propanol								
K	0.1000	0.2000	0.3005	0.4010	0.5013	0.6002	0.7002	0.8012	0.9001
293.15	22.34	23.08	24.11	24.92	25.85	26.88	27.92	29.79	31.87
298.15	22.16	22.90	23.90	24.69	25.68	26.60	27.75	29.67	31.58
303.15	22.03	22.74	23.66	24.45	25.42	26.38	27.57	29.44	31.42
313.15	21.76	22.52	23.41	24.19	25.18	26.06	27.09	29.04	31.06
323.15	21.55	22.23	23.05	23.95	24.92	25.74	26.83	28.74	30.77
333.15	21.22	21.92	22.85	23.66	24.72	25.57	26.59	28.24	30.47

^aStandard uncertainties, u , are $u(T) = 0.01$ K, $u(x) = 0.0001$, $u(P) = 1$ kPa, and $u(\sigma) = 0.15$ mN·m⁻¹. Relative standard uncertainties, u_r , are $u_r(\rho) = 0.001$ and $u_r(\eta) = 0.01$.

The calculated densities values using eqs 2 to 5 against the experimental values are depicted in Figures 1 to 4. The correlated data were then compared with the corresponding experimental data by computing the absolute percentage error (APER) using

$$\text{APER} = \frac{100}{N} \sum \frac{|\text{computed} - \text{experimental}|}{\text{experimental}} \quad (6)$$

in which N is the number of data points in each set. The $\text{APER}_{\text{density}}$ for EMIM DEP/water/1-propanol is 0.507 %, 0.427 % for EMIM DEP/water/2-propanol, 0.870 % for MMIM DMP/water/1-propanol, and 0.509 % for MMIM DMP/water/2-propanol.

Surface Tension. The Jouyban–Acree model for representing the surface tension of ternary mixtures is

$$\begin{aligned} \ln \sigma_{m,T} = & x_1 \ln \sigma_{1,T} + x_2 \ln \sigma_{2,T} + x_3 \ln \sigma_{3,T} \\ & + x_1 x_2 \sum_{j=0}^2 \left[\frac{A_j (x_1 - x_2)^j}{T} \right] + x_1 x_3 \\ & \times \sum_{j=0}^2 \left[\frac{B_j (x_1 - x_3)^j}{T} \right] + x_2 x_3 \sum_{j=0}^2 \left[\frac{C_j (x_2 - x_3)^j}{T} \right] \\ & + x_1 x_2 x_3 \sum_{j=0}^2 \left[\frac{D_j (x_1 - x_2 - x_3)^j}{T} \right] \end{aligned} \quad (7)$$

where $\sigma_{m,T}$, $\sigma_{1,T}$, $\sigma_{2,T}$, and $\sigma_{3,T}$ are the surface tensions of the mixture and solvents 1, 2, and 3 at temperature T , respectively, and D_j represent the model constants.⁵² These model constants are computed by regressing $(\ln \sigma_{m,T} - x_1 \ln \sigma_{1,T} - x_2 \ln \sigma_{2,T} - x_3$

Table 9. Surface Tension, $\sigma/\text{mN m}^{-1}$ of MMIM DMP/Water/1-Propanol and MMIM DMP/Water/1-Propanol 0.987 atm^a

T	surface tension, $\sigma/\text{mN}\cdot\text{m}^{-1}$								
	$X_{\text{MMIM DMP}}$ in MMIM DMP/water/1-propanol								
K	0.1089	0.2062	0.3059	0.4022	0.5059	0.6002	0.7002	0.8002	0.9028
293.15	31.84	32.87	34.00	35.11	36.19	37.40	40.41	43.52	45.55
298.15	31.81	32.80	33.93	35.04	36.03	37.29	40.22	43.29	44.90
303.15	32.13	33.15	34.28	35.21	36.28	37.48	40.28	43.41	44.61
313.15	30.77	31.64	32.79	33.70	34.72	35.84	38.48	41.61	42.74
323.15	29.15	29.96	31.16	32.01	33.11	34.08	36.56	39.71	40.51
333.15	28.45	29.27	30.34	31.26	32.26	33.32	35.67	38.68	39.57
T	$X_{\text{MMIM DMP}}$ in MMIM DMP/water/2-propanol								
	0.1	0.2	0.3005	0.401	0.5013	0.6002	0.7002	0.8012	0.913
293.15	29.84	31.03	32.53	34.05	36.40	38.93	40.84	42.94	47.16
298.15	29.80	31.03	32.51	34.04	36.23	38.58	40.40	42.50	46.62
303.15	30.16	31.29	32.74	34.24	36.36	38.65	40.41	42.41	46.39
313.15	28.73	29.81	31.26	32.58	34.57	36.67	38.36	40.16	43.90
323.15	27.10	28.07	29.45	30.71	32.54	34.51	36.17	37.77	41.22
333.15	26.28	27.18	28.55	29.86	31.62	33.38	34.95	36.45	39.56

^aStandard uncertainties, u , are $u(T) = 0.01$ K, $u(x) = 0.0001$, $u(P) = 1$ kPa, and $u(\sigma) = 0.15$ mN·m⁻¹. Relative standard uncertainties, u_r , are $u_r(\rho) = 0.001$ and $u_r(\eta) = 0.01$.

Table 10. Viscosity, $\eta/\text{mPa}\cdot\text{s}$ of EMIM DEP/Water/2-Propanol and EMIM DEP/Water/2-Propanol at 0.987 atm^a

T	viscosity, $\eta/\text{mPa}\cdot\text{s}$								
	$X_{\text{EMIM DEP}}$ in EMIM DEP/water/1-propanol								
K	0.1033	0.2025	0.3039	0.4020	0.5051	0.6056	0.7013	0.8026	0.9033
293.15	47.13	65.39	89.84	117.28	151.28	199.74	248.83	305.27	372.97
298.15	39.45	53.86	73.29	98.03	120.37	151.11	182.76	224.07	273.36
303.15	29.97	44.62	61.48	78.39	100.27	128.36	158.23	197.74	241.36
313.15	22.23	31.71	40.29	49.38	62.35	76.06	93.10	110.22	133.26
323.15	16.07	22.49	28.37	33.28	42.25	51.19	58.24	69.73	80.42
333.15	12.13	17.46	21.61	23.69	30.19	34.77	41.36	47.48	52.04
T	$X_{\text{EMIM DEP}}$ in EMIM DEP/water/2-propanol								
	0.1000	0.2000	0.3005	0.4010	0.5013	0.6002	0.7002	0.8012	0.9001
293.15	41.39	50.05	67.74	101.54	156.95	235.33	336.61	436.52	484.35
298.15	34.20	41.77	54.82	79.18	119.79	176.24	257.14	322.01	351.83
303.15	26.35	31.23	46.19	67.26	108.28	151.76	215.37	278.08	306.72
313.15	17.86	23.23	30.26	41.83	62.37	89.58	124.24	153.24	167.94
323.15	12.64	14.49	20.12	29.25	41.28	55.23	75.66	94.26	101.23
333.15	10.12	12.13	16.61	20.72	28.19	38.71	52.01	61.12	66.53

^aStandard uncertainties, u , are $u(T) = 0.01$ K, $u(x) = 0.0001$, $u(P) = 1$ kPa, and $u(\sigma) = 0.15$ mN·m⁻¹. Relative standard uncertainties, u_r , are $u_r(\rho) = 0.001$ and $u_r(\eta) = 0.01$.

$\ln \sigma_{3,T}$ against x_1x_2/T , $x_1x_2(x_1 - x_2)/T$, $x_1x_2(x_1 - x_2)^2/T$, x_1x_3/T , $x_1x_3(x_1 - x_3)/T$, $x_1x_3(x_1 - x_3)^2/T$, x_2x_3/T , $x_2x_3(x_2 - x_3)/T$, $x_2x_3(x_2 - x_3)^2/T$, $x_1x_2x_3/T$, $x_1x_2x_3(x_1 - x_2 - x_3)/T$ and $x_1x_2x_3(x_1 - x_2 - x_3)^2/T$ using a no intercept least-square analysis.

The proposed model after excluding nonsignificant model constants ($p > 0.05$), for

(i) EMIM DEP/water/1-propanol mixture:

$$\ln \sigma_{m,T} = x_1 \ln \sigma_{1,T} + x_2 \ln \sigma_{2,T} + x_3 \ln \sigma_{3,T} - 608.092 \left[\frac{x_1x_3}{T} \right] - 839.635 \left[\frac{x_2x_3}{T} \right] + 887.654 \left[\frac{x_1x_2x_3}{T} \right] \quad (8)$$

(ii) EMIM DEP/water/2-propanol mixture:

$$\ln \sigma_{m,T} = x_1 \ln \sigma_{1,T} + x_2 \ln \sigma_{2,T} + x_3 \ln \sigma_{3,T} - 632.236 \left[\frac{x_1x_2}{T} \right] - 735.677 \left[\frac{x_2x_3}{T} \right] + 12796.69 \left[\frac{x_2x_3(x_2 - x_3)}{T} \right] + 587.154 \left[\frac{x_1x_2x_3}{T} \right] \quad (9)$$

(iii) MMIM DMP/water/1-propanol mixture:

$$\ln \sigma_{m,T} = x_1 \ln \sigma_{1,T} + x_2 \ln \sigma_{2,T} + x_3 \ln \sigma_{3,T} - 217.262 \left[\frac{x_1x_3}{T} \right] - 312.669 \left[\frac{x_2x_3}{T} \right] - 367.672 \left[\frac{x_1x_2x_3}{T} \right] \quad (10)$$

Table 11. Viscosity, η /mPa·s of MMIM DMP/Water/1-Propanol and MMIM DMP/Water/2-Propanol at 0.987 atm^a

T	viscosity, η /mPa·s								
	$X_{\text{MMIM DMP}}$ in MMIM DMP/water/1-propanol								
K	0.1089	0.2062	0.3059	0.4022	0.5059	0.6002	0.7002	0.8002	0.9028
293.15	34.12	43.14	53.55	72.45	106.23	158.36	217.82	271.38	342.37
298.15	30.43	37.34	47.37	63.63	89.56	132.24	172.33	211.24	264.11
303.15	27.45	33.26	41.84	55.44	75.73	110.11	148.87	168.61	205.15
313.15	21.4	25.77	30.99	40.81	53.38	76.25	109.24	123.28	154.33
323.15	16.12	19.23	24.25	30.24	41.26	58.75	81.12	100.26	126.64
333.15	12.33	15.27	19.74	24.11	32.44	45.24	62.11	84.92	102.32
T	$X_{\text{MMIM DMP}}$ in MMIM DMP/water/2-propanol								
K	0.1000	0.2000	0.3005	0.4010	0.5013	0.6002	0.7002	0.8012	0.9130
293.15	42.35	53.89	63.53	76.22	100.18	123.72	161.76	248.62	355.63
298.15	32.34	41.83	51.28	64.36	82.56	103.92	130.19	202.84	281.72
303.15	25.43	32.83	42.14	54.19	69.14	87.37	109.24	163.64	225.57
313.15	17.24	23.12	30.62	39.23	48.62	62.34	80.52	113.05	153.22
323.15	13.29	17.66	24.88	31.73	38.82	46.55	58.48	79.62	112.64
333.15	10.91	15.12	20.27	26.92	31.66	37.15	44.19	60.73	91.71

^aStandard uncertainties, u , are $u(T) = 0.01$ K, $u(x) = 0.0001$, $u(P) = 1$ kPa, and $u(\sigma) = 0.15$ mN·m⁻¹. Relative standard uncertainties, u_r , are $u_r(\rho) = 0.001$ and $u_r(\eta) = 0.01$.

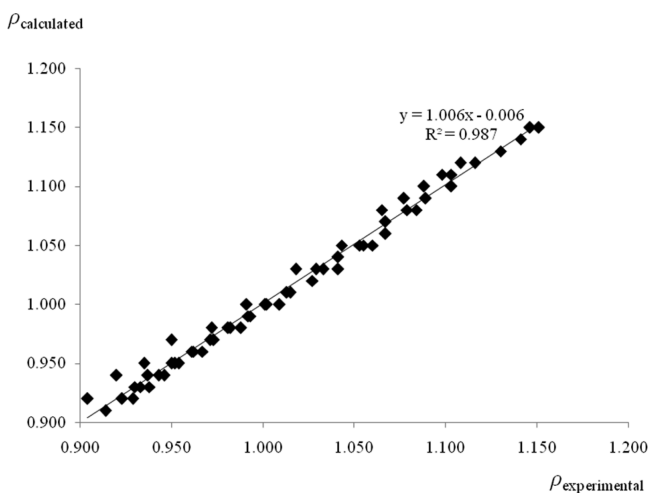


Figure 1. Density values of EMIM DEP/water/1-propanol calculated using eq 2 against the corresponding experimental values.

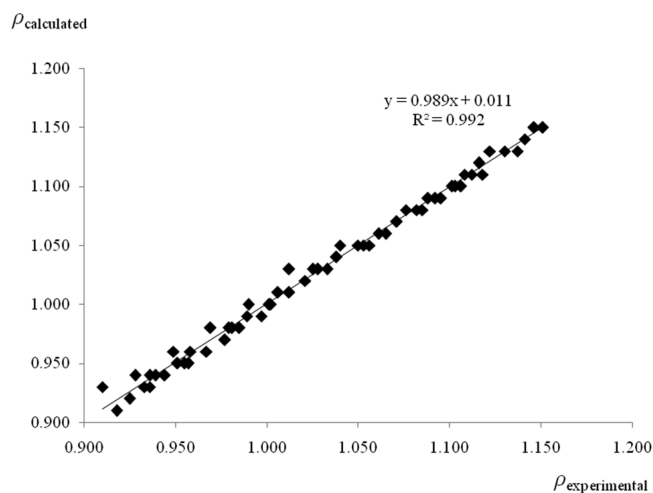


Figure 2. Density values of EMIM DEP/water/2-propanol calculated using eq 3 against the corresponding experimental values.

(iv) MMIM DMP/water/2-propanol mixture:

$$\ln \sigma_{m,T} = x_1 \ln \sigma_{1,T} + x_2 \ln \sigma_{2,T} + x_3 \ln \sigma_{3,T} - 140.120 \left[\frac{x_1 x_3}{T} \right] - 359.944 \left[\frac{x_2 x_3}{T} \right] - 444.851 \left[\frac{x_1 x_2 x_3}{T} \right] \quad (11)$$

The calculated surface tension values using eqs 8 to 11 against the experimental values are depicted in Figures 5 to 8. From eq 6, The $\text{APER}_{\text{surface tension}}$ for EMIM DEP/water/1-propanol is 0.6696 %, 0.245 % for EMIM DEP/water/2-propanol, 1.396 % for MMIM DMP/water/1-propanol, and 1.726 % for MMIM DMP/water/2-propanol.

Viscosity. The Jouyban–Acree model for representing the viscosity of ternary mixtures is

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} + x_3 \ln \eta_{3,T} + x_1 x_2 \sum_{j=0}^2 \left[\frac{A_j (x_1 - x_2)^j}{T} \right] + x_1 x_3 \times \sum_{j=0}^2 \left[\frac{B_j (x_1 - x_3)^j}{T} \right] + x_2 x_3 \sum_{j=0}^2 \left[\frac{C_j (x_2 - x_3)^j}{T} \right] + x_1 x_2 x_3 \sum_{j=0}^2 \left[\frac{D_j (x_1 - x_2 - x_3)^j}{T} \right] \quad (12)$$

where $\eta_{m,T}$, $\eta_{1,T}$, $\eta_{2,T}$, and $\eta_{3,T}$ are the viscosities of the mixture and solvents 1, 2, and 3 at temperature T , respectively, and D_j represent the model constants.^{53,54} These model constants are computed by regressing $(\ln \eta_{m,T} - x_1 \ln \eta_{1,T} - x_2 \ln \eta_{2,T} - x_3 \ln \eta_{3,T})$ against $x_1 x_2 / T$, $x_1 x_2 (x_1 - x_2) / T$, $x_1 x_2 (x_1 - x_2)^2 / T$, $x_1 x_3 / T$, $x_1 x_3 (x_1 - x_3) / T$, $x_1 x_3 (x_1 - x_3)^2 / T$, $x_2 x_3 / T$, $x_2 x_3 (x_2 - x_3) / T$, $x_2 x_3 (x_2 - x_3)^2 / T$, $x_1 x_2 x_3 / T$, $x_1 x_2 x_3 (x_1 - x_2 - x_3) / T$ and

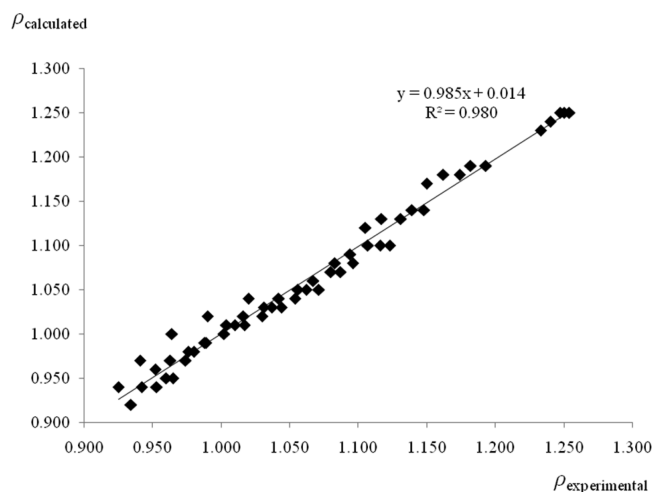


Figure 3. Density values of MMIM DMP/water/1-propanol calculated using eq 4 against the corresponding experimental values.

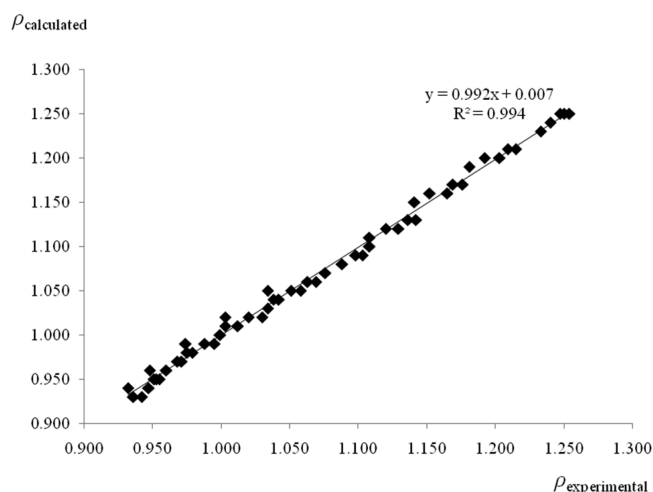


Figure 4. Density values of MMIM DMP/water/2-propanol calculated using eq 5 against the corresponding experimental values.

$x_1x_2x_3(x_1 - x_2 - x_3)^2/T$ using a no intercept least-square analysis.

The proposed model after excluding nonsignificant model constants ($p > 0.05$), for

(i) EMIM DEP/water/1-propanol mixture:

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} + x_3 \ln \eta_{3,T} + 2112.568 \left[\frac{x_1x_2}{T} \right] + 3618.064 \left[\frac{x_2x_3}{T} \right] + 1197.071 \left[\frac{x_1x_3}{T} \right] \quad (13)$$

(ii) EMIM DEP/water/2-propanol mixture:

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} + x_3 \ln \eta_{3,T} + 4314.520 \left[\frac{x_1x_2}{T} \right] + 3778.940 \left[\frac{x_2x_3}{T} \right] - 7994.452 \left[\frac{x_1x_3}{T} \right] \quad (14)$$

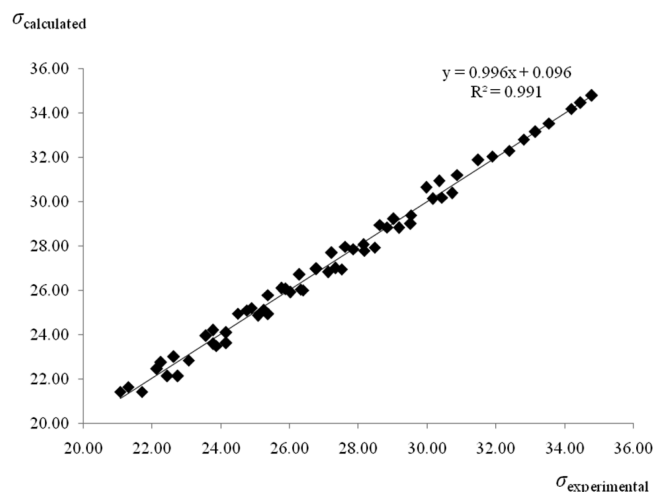


Figure 5. Surface tension values of EMIM DEP/water/1-propanol calculated using eq 8 against the corresponding experimental values.

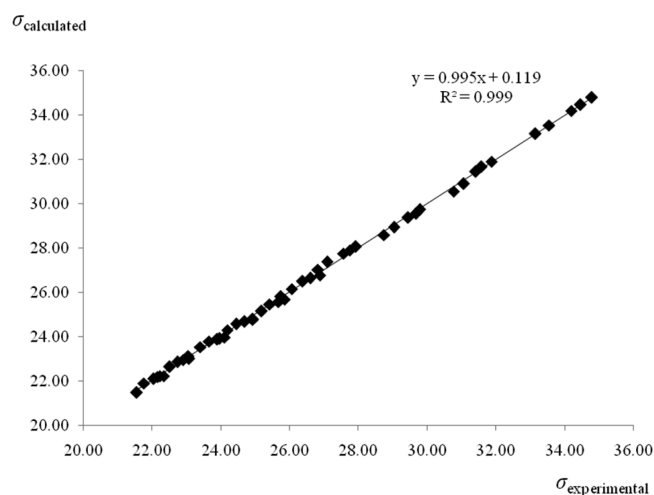


Figure 6. Surface tension values of EMIM DEP/water/2-propanol calculated using eq 9 against the corresponding experimental values.

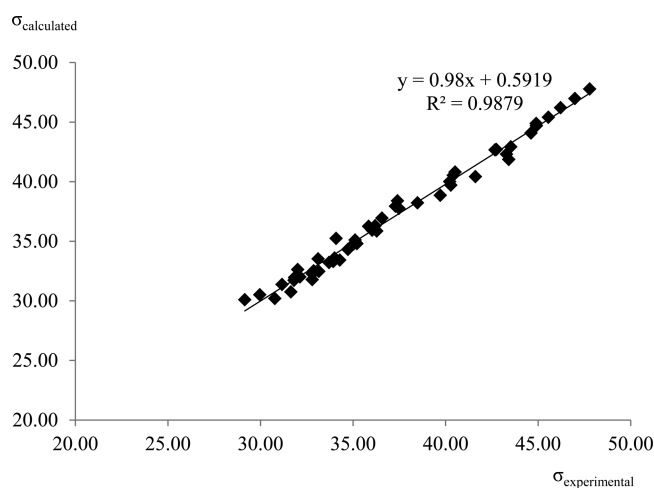


Figure 7. Surface tension values of MMIM DMP/water/1-propanol calculated using eq 10 against the corresponding experimental values.

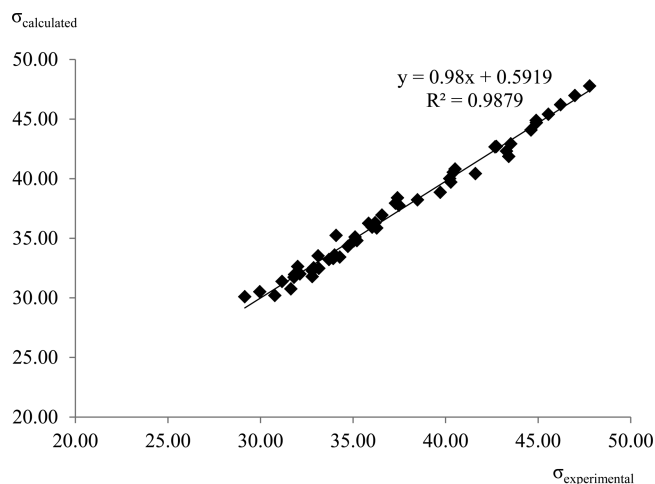


Figure 8. Surface tension values of MMIM DMP/water/2-propanol calculated using eq 11 against the corresponding experimental values.

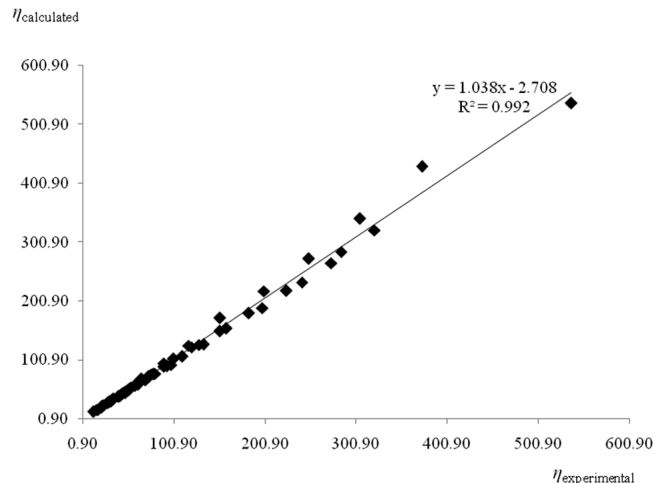


Figure 9. Viscosity values of EMIM DEP/water/1-propanol calculated using eq 13 against the corresponding experimental values.

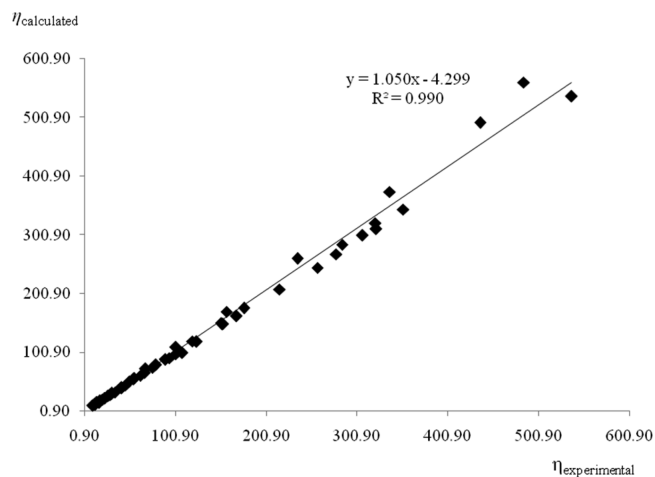


Figure 10. Viscosity values of EMIM DEP/water/2-propanol calculated using eq 14 against the corresponding experimental values.

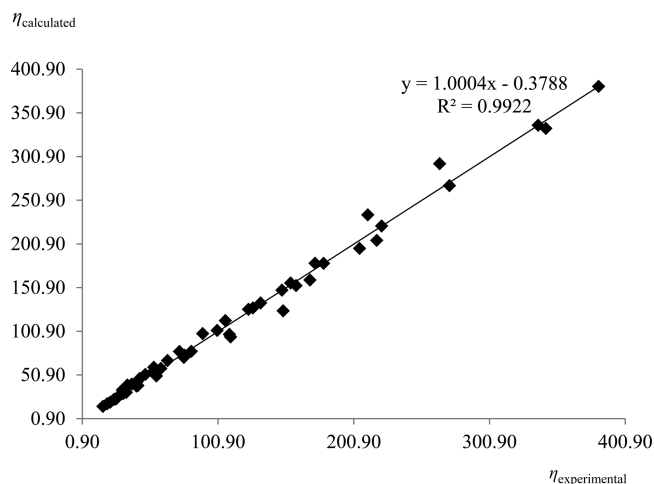


Figure 11. Viscosity values of MMIM DMP/water/1-propanol calculated using eq 15 against the corresponding experimental values.

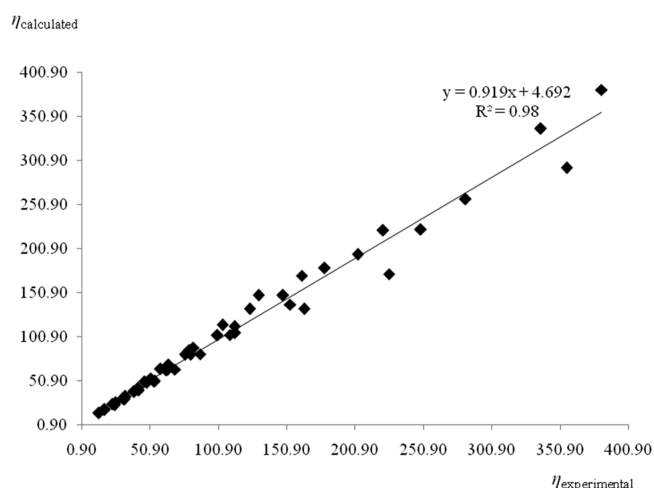


Figure 12. Viscosity values of MMIM DMP/water/2-propanol calculated using eq 16 against the corresponding experimental values.

(iii) MMIM DMP/water/1-propanol system:

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} + x_3 \ln \eta_{3,T} + 2736.178 \left[\frac{x_1 x_2}{T} \right] + 3800.763 \left[\frac{x_2 x_3}{T} \right] - 4081.101 \left[\frac{x_1 x_2 x_3}{T} \right] \quad (15)$$

(iv) MMIM DMP/water/2-propanol system:

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} + x_3 \ln \eta_{3,T} + 1619.312 \left[\frac{x_1 x_3}{T} \right] + 3589.888 \left[\frac{x_2 x_3}{T} \right] \quad (16)$$

The calculated viscosity values using eqs 13 to 16 against the experimental values are depicted in Figures 9 to 12. From eq 6, The $\text{APER}_{\text{viscosity}}$ for EMIM DEP/water/1-propanol is 3.300%, 3.392% for EMIM DEP/water/2-propanol, 5.8480% for MMIM DMP/water/1-propanol and 6.070% for MMIM DMP/water/2-propanol.

4. CONCLUSION

Density, surface tension, and viscosity of ternary mixtures EMIM DEP + water + 1-propanol, EMIM DEP + water + 2-propanol, MMIM DMP + water + 1-propanol, and MMIM DMP + water + 2-propanol have been measured over the entire concentration range at temperatures from (293.15 to 333.15) K. Density and surface tension of ternary mixtures decreased linearly with temperature. However, viscosity for all ternary mixtures decreased nonlinearly with temperature. The ternary mixtures containing primary alcohol or EMIM DEP have lower physicochemical values. The most prominent correlation for density and surface tension for all four ternary mixtures was obtained using a linear equation. In contrast, the best correlation for viscosity data was obtained using a second order polynomial equation. As reported previously, the experimental density, surface tension, and viscosity data for all ternary mixtures studied were also greatly affected by mole fractions of ionic liquids used.

As an addition to this study, the Jouyban–Acree model presented fairly reasonable precise results to calculate the density, surface tension, and viscosity of the four studied mixtures. The overall APER of the studied ternary mixtures are below 0.9 % for density, 1.7 % for surface tension, and 6.1 % for viscosity. Hence, from the APER values, the Jouyban–Acree model is reasonably represents the physicochemical data.

■ ASSOCIATED CONTENT

Supporting Information

Additional figures of the density, surface tension, and viscosity on temperature or mole fraction of ionic liquid. This material is available free of charge via the Internet at <http://pubs.acs.org>.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: asrina@um.edu.my. Tel: +603-79672147. Fax: +603-79677188.

Funding

This research is supported by PPP grant (PV124-2012) and High Impact Research MoE Grant UM.C UM.C/625/1/HIR/MoE/SC/04 from the Ministry of Education Malaysia and University Malaya Centre for Ionic Liquids (UMCiL).

Notes

The authors declare no competing financial interest.

■ REFERENCES

- (1) Reinert, L.; Batouche, K.; Lévêque, J. M.; Muller, F.; Bény, J. M.; Kebabi, B.; Duclaux, L. Adsorption of imidazolium and pyridinium ionic liquids onto montmorillonite: Characterisation and thermodynamic calculations. *Chem. Eng. J.* **2012**, *209*, 13–19.
- (2) Blath, J.; Christ, M.; Deubler, N.; Hirth, T.; Schiestel, T. Gas solubilities in room temperature ionic liquids – Correlation between RTiL-molar mass and Henry's law constant. *Chem. Eng. J.* **2011**, *172*, 167–176.
- (3) Marciniak, A. Influence of anion structure on the liquid–liquid equilibria of 1-Ethyl-3-methyl-imidazolium cation based ionic liquid-hydrocarbon binary systems. *J. Chem. Eng. Data* **2011**, *56*, 368–374.
- (4) Shifflet, M. B.; Niehaus, A. M. S.; Yokozeki, A. Liquid–liquid equilibria in binary mixtures containing chlorobenzene, bromobenzene, and iodobenzene with ionic liquid 1-ethyl-3-methylimidazolium bis-(trifluoromethylsulfonyl)imide. *J. Chem. Eng. Data* **2009**, *54*, 2090–2094.
- (5) Sahandzheva, K.; Tuma, D.; Breyer, S.; Kamps, Á.P.S.; Maurer, G. Liquid–liquid equilibrium in mixtures of the ionic liquid 1-*n*-butyl-3-methylimidazolium hexafluorophosphate and an alkanol. *J. Chem. Eng. Data* **2006**, *51*, 1516–1525.
- (6) Anantharaj, R.; Benerjee, T. Liquid–liquid equilibrium studies on the removal of thiophene and pyridine from pentane using imidazolium-based ionic liquids. *J. Chem. Eng. Data* **2013**, *58*, 829–837.
- (7) Ardila, Y. C.; Machado, A. B.; Pinto, G. M. F.; Filho, R. M.; Maciel, M. R. W. Liquid–liquid equilibrium in ternary systems present in biodiesel purification from soybean oil and castor oil at (298.2 and 333.2) K. *J. Chem. Eng. Data* **2013**, *58*, 605–610.
- (8) Park, Y.; Shin, D.; Jang, Y. N. Park A.H.A.; CO₂ capture capacity and swelling measurements of liquid-like nanoparticle organic hybrid materials via attenuated total reflectance fourier transform infrared spectroscopy. *J. Chem. Eng. Data* **2012**, *57*, 40–45.
- (9) Ahmady, A.; Hashim, M. A.; Aroua, M. K. Experimental investigation on the solubility and initial rate of absorption of CO₂ in aqueous mixtures of methyl-diethanolamine with the ionic liquid 1-butyl-3-methylimidazolium tetrafluoroborate. *J. Chem. Eng. Data* **2010**, *55*, 5733–5738.
- (10) Ramdin, M.; Vlucht, T. J. H.; de Loos, T. W. Solubility of CO₂ in the ionic liquids [TBMN][MeSO₄] and [TBMP][MeSO₄]. *J. Chem. Eng. Data* **2012**, *57*, 2275–2280.
- (11) Rogers, E. I.; Šljukić, B.; Hardacre, C.; Compton, R. G. Electrochemistry in room-temperature ionic liquids: potential windows at mercury electrodes. *J. Chem. Eng. Data* **2009**, *54*, 2049–2053.
- (12) Wibowo, R.; Jones, S. E. W.; Compton, R. G. Investigating the electrode kinetics of the Li/Li⁺ couple in a wide range of room temperature ionic liquids at 298 K. *J. Chem. Eng. Data* **2010**, *55*, 1374–1376.
- (13) O'Mahony, A. M.; Silvester, D. S.; Aldous, L.; Hardacre, C.; Compton, R. G. Effect of water on the electrochemical window and potential limits of room-temperature ionic liquids. *J. Chem. Eng. Data* **2008**, *53*, 2884–2891.
- (14) Iglesias, M.; Gonzalez-Olmos, R.; Cota, I.; Medina, F. Brønsted ionic liquids: Study of physico-chemical properties and catalytic activity in aldol condensations. *Chem. Eng. J.* **2010**, *162*, 802–808.
- (15) Muhammad, N.; Man, Z. B.; Bustam, M. A.; Abdul Mutalib, M. I.; Wilfred, C. D.; Rafiq, S. Synthesis and Thermophysical Properties of low viscosity amino acid-based ionic liquids. *J. Chem. Eng. Data* **2011**, *56*, 3157–3162.
- (16) Jiang, Y.; Xia, H.; Yu, J.; Guo, C.; Liu, H. Hydrophobic ionic liquids-assisted polymer recovery during penicillin extraction in aqueous two-phase system. *Chem. Eng. J.* **2009**, *147*, 22–26.
- (17) Pang, F. M.; Seng, C. E.; Teng, T. T.; Ibrahim, M. H. Densities and viscosities of aqueous solutions of 1-propanol and 2-propanol at temperatures from 293.15 to 333.15 K. *J. Mol. Liq.* **2007**, *136*, 71–78.
- (18) Tian, Y.; Meng, X.; Shi, L. Removal of dimethyl disulfide via extraction using imidazolium-based phosphoric ionic liquids. *Fuel* **2014**, *129*, 225–230.
- (19) Froschauer, C.; Hummel, M.; Gerhard, L.; Schottenberger, H.; Sixta, H.; Weber, H. K.; Zuckersstätter, G. Dialkylphosphate-related ionic liquids as selective solvents for xylan. *Biomacromolecules* **2012**, *13*, 1973–1980.
- (20) Poole, C. F.; Poole, S. K. Extraction of organic compounds with room temperature ionic liquids. *J. Chromatogr. A* **2010**, *1217*, 2268–2286.
- (21) Fan, J.; Fan, Y.; Pei, Y.; Wu, K.; Wang, J.; Fan, M. Solvent extraction of selected endocrine-disrupting phenols using ionic liquids. *Sep. Purif. Technol.* **2008**, *61*, 324–331.
- (22) Pareiro, A. B.; Rodríguez, A. Effective extraction in packed column of ethanol from the azeotropic mixture ethanol + hexane with an ionic liquid as solvent. *Chem. Eng. J.* **2009**, *153*, 80–85.
- (23) Huddleston, J. G.; Willauer, H. D.; Swatoski, R. P.; Visser, A. E.; Rogers, R. D. Room temperature ionic liquids as novel media for 'clean' liquid–liquid extraction. *Chem. Comm.* **1998**, 1765–1766.
- (24) Choi, J. A.; Shim, E. G.; Scrosati, B.; Dong, W. K. Mixed Electrolytes of organic solvents and ionic liquid for rechargeable lithium-ion batteries. *Bull. Korean Chem. Soc.* **2010**, *31*, 3190–3194.
- (25) Quinner, Q.; Baltazar, S.; Leininger, K.; Anderson, J. L. Binary ionic liquid mixtures as gas chromatography stationary phases for

improving the separation selectivity of alcohols and aromatic compounds. *J. Chromatogr. A* **2008**, *1182*, 119–127.

(26) Wang, J.; Wang, D.; Li, Z.; Zhang, F. Vapor pressure measurement and correlation or prediction for water, 1-propanol, 2-propanol and their binary mixtures with [MMIM][DMP] ionic liquid. *J. Chem. Eng. Data* **2010**, *55*, 4872–4877.

(27) Holbrey, J. D.; Reichert, W. M.; Swatoski, R. P.; Brookes, G. A.; Pitner, W. R.; Seddon, K. R.; Rogers, R. D. Efficient halide free synthesis of new, low cost ionic liquids: alkylimidazolium salts containing methyl- and ethyl- sulfate anion. *Green Chem.* **2002**, *4*, 407–413.

(28) Wang, J.; Li, Z. Measurement and modeling of vapor-liquid equilibria for systems containing alcohols, water, and imidazolium-based phosphate ionic liquids. *J. Chem. Eng. Data* **2013**, *58*, 1641–1649.

(29) Valderrama, J. O.; Urbina, F.; Faúndez, C. A. Gas–liquid equilibrium modeling of mixtures containing supercritical carbon dioxide and an ionic liquid. *J. Supercrit. Fluids* **2012**, *64*, 32–38.

(30) Alvarez, V. H.; Saldaña, M. D. A. Thermodynamic prediction of vapor–liquid equilibrium of supercritical CO₂ or CHF₃ + ionic liquids. *J. Supercrit. Fluids* **2012**, *66*, 29–35.

(31) Jiang, H.; Adidharma, H. Thermodynamic modeling of aqueous ionic liquid solutions and prediction of methane hydrate dissociation conditions in the presence of ionic liquid. *Chem. Eng. Sci.* **2013**, *102*, 24–31.

(32) Mattedi, S.; Carvalho, P. J.; Coutinho, J. A. P.; Alvarez, V. H.; Iglesias, M. High pressure CO₂ solubility in N-methyl-2-hydroxyethylammonium protic ionic liquids. *J. Supercrit. Fluids* **2011**, *56*, 224–230.

(33) Pang, F. M.; Seng, C. E.; Teng, T. T.; Ibrahim, M. H. Densities and viscosities of aqueous solutions of 1-propanol and 2-propanol at temperatures from 293.15 to 333.15 K. *J. Mol. Liq.* **2007**, *136*, 71–78.

(34) Ghahremani, H.; Moradi, A.; Torghabeh-Abedini, J.; Hasani, S. M. Measuring surface tension of binary mixtures of water + alcohols from the diffraction pattern of surface ripples. *Der Chem. Sin.* **2011**, *6*, 212–221.

(35) Jiménez, E.; Cabanas, M.; Segade, L.; García-Garabal, S.; Casas, H. Excess volume, changes of refractive index and surface tension of binary 1,2-ethanediol + 1-propanol or 1-butanol mixtures at several temperatures. *Fluid Phase Equilib.* **2001**, *180*, 151–164.

(36) Hoke, B. C.; Chen, J. C. Binary aqueous-organic surface tension temperature dependence. *J. Chem. Eng. Data* **1991**, *36*, 322–326.

(37) Wang, J.; Li, C.; Shen, C.; Wang, Z. Towards understanding the effect of electrostatic interactions on the density of ionic liquids. *Fluid Phase Equilib.* **2009**, *279*, 87–91.

(38) Wang, J.; Zhao, F.; Liu, R.; Hu, Y. Thermophysical properties of 1-methyl-3-methylimidazolium dimethylphosphate and 1-ethyl-3-methylimidazolium diethylphosphate. *J. Chem. Thermodyn.* **2011**, *43*, 47–50.

(39) Ficke, L. E.; Novak, R.; Brennecke, J. F. Thermodynamic and Thermophysical Properties of Ionic Liquid + Water Systems. *J. Chem. Eng. Data* **2010**, *55*, 4946–4950.

(40) Ren, N.; Gong, Y.; Lu, Y.; Meng, H.; Li, C. Surface tension measurements for seven imidazolium-based dialkylphosphate ionic liquids and their binary mixtures with water (methanol or ethanol) at 298.15 K and 1 atm. *J. Chem. Eng. Data* **2014**, *59*, 189–196.

(41) Tenney, C. M.; Massel, M.; Mayes, J. M.; Sen, M.; Brennecke, J. F.; Marginn, E. J. A Computational and Experimental Study of the Heat Transfer Properties of Nine Different Ionic Liquids. *J. Chem. Eng. Data* **2014**, *59*, 391–399.

(42) Kato, R.; Gmehling, J. Activity coefficients at infinite dilution of various solutes in the ionic liquids [MMIM]⁺[CH₃SO₄][−], [MMIM]⁺[CH₃OC₂H₄SO₄][−], [MMIM]⁺[(CH₃)₂PO₄][−], [C₅H₅NC₂H₅]⁺[(CF₃SO₂)₂N][−] and [C₅H₅NH]⁺[C₂H₅OC₂H₄OSO₃][−]. *Fluid Phase Equilib.* **2004**, *226*, 37–44.

(43) Ochędzan-Siodlak, W.; Dziubek, K.; Siodlak, D. Densities and viscosities of imidazolium and pyridinium chloroaluminate ionic liquids. *J. Mol. Liq.* **2013**, *77*, 85–93.

(44) Kavitha, T.; Attri, P.; Venkatesu, P.; Rama Devi, R. S.; Hofman, T. Influence of temperature on thermophysical properties of ammonium ionic liquids with N-methyl-2-pyrrolidone. *Thermochim. Acta* **2012**, *545*, 131–140.

(45) Freire, M. G.; Carvalho, P. J.; Fernandes, A. M.; Marrucho, I. M.; Queimada, A. J.; Coutinho, J. A. P. Surface tensions of imidazolium based ionic liquids: Anion, cation, temperature and water effect. *J. Colloid Interface Sci.* **2007**, *314*, 621–630.

(46) Lemraski, E. G.; Zobeydi, R. Applying parachor method to the prediction of ionic liquids surface tension based on modified group contribution. *J. Mol. Liq.* **2014**, *193*, 204–209.

(47) Ghatee, M. H.; Bahrani, M.; Khanjari, N. Measurement and study of density, surface tension, and viscosity of quaternary ammonium-based ionic liquids ([N₂₂₂(n)]Tf₂N). *J. Chem. Thermodyn.* **2013**, *65*, 42–52.

(48) Królikowska, M.; Lipinski, P.; Maik, D. Density, viscosity and phase equilibria study of {ethylsulfate-based ionic liquid + water} binary systems as a function of temperature and composition. *Thermochim. Acta* **2014**, *582*, 1–9.

(49) Domanska, U.; Laskowska, M. Effect of Temperature and Composition on The Density and Viscosity of Binary Mixtures of Ionic Liquid with Alcohols. *J. Solution Chem.* **2009**, *38*, 779–799.

(50) Jouyban, A.; Fathi-Azarbayjani, A.; Khoubnasabjafari, M.; Acree, W. E., Jr. Mathematical representation of the density of liquid mixtures at various temperatures using Jouyban–Acree model. *Indian J. Chem.* **2005**, *44A*, 1–8.

(51) Jouyban, A.; Maljaeib, S. H.; Khoubnasabjafari, M.; Fathi-Azarbayjani, A.; Acree, W. E., Jr. A global model to predict density of non-aqueous binary solvent mixtures at various temperatures. *Indian J. Chem.* **2012**, *51A*, 695–698.

(52) Jouyban, A.; Fathi-Azarbayjani, A.; Acree, W. E., Jr. Surface tension calculation of mixed solvents with respect to solvent composition and temperature by using Jouyban–Acree model. *Chem. Pharm. Bull.* **2004**, *52*, 1219–1222.

(53) Jouyban, A.; Khoubnasabjafari, M.; Vaez-Gharamaleki, Z.; Fekari, Z.; Acree, W. E., Jr. Calculation of the viscosity of binary liquids at various temperatures using Jouyban–Acree model. *Chem. Pharm. Bull.* **2005**, *53*, 519–523.

(54) Jouyban, A.; Soleymani, J.; Jafari, F.; Khoubnasabjafari, M.; Acree, W. E. Mathematical Representation of Viscosity of Ionic Liquid + Molecular Solvent Mixtures at Various Temperatures Using the Jouyban–Acree Model. *J. Chem. Eng. Data* **2013**, *58*, 1523–1528.