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Density, Surface Tension, and Viscosity of Ionic Liquids (1-Ethyl-3-methylimidazolium diethylphosphate and 1,3-Dimethylimidazolium dimethylphosphate) Aqueous Ternary Mixtures with MDEA

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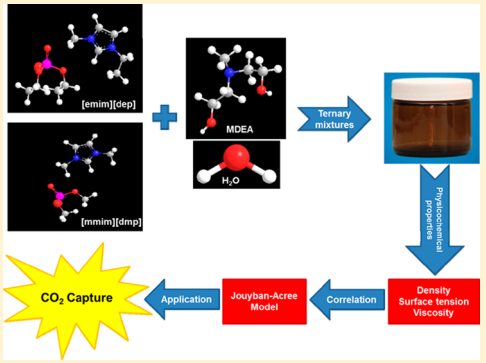
ABSTRACT: The density, surface tension, and viscosity of ionic liquids (1-ethyl-3-methylimidazolium diethylphosphate and 1,3-dimethylimidazolium dimethylphosphate) and ternary mixtures with aqueous MDEA, were measured over the whole concentrations range at various temperatures (293.15−343.15) K by using Anton Paar DMA 4500 densimeter, Kruss Processor Tensiometer K100, and R/S+ Brookfield rheometer. The experimental density and surface tension decrease linearly with mole fractions of ionic liquids. Data on viscosity demonstrates a temperature-dependence behavior, that decreased nonlinearly with temperature. Evaluations of the measured physicochemical properties were completed. The best correlation for density and surface tension data were linear fitting, while the viscosity data was obtained by polynomial regression. ILs mole fraction also influenced the experimental density, surface tension, and viscosity. The Jouyban−Acree model was used to correlate the physicochemical properties of the mixtures and pure compounds at different temperatures. The absolute percentage error (APER) for each correlations was less than 8 %, proving that this model accurately represents the physicochemical properties data.



1. INTRODUCTION

In recent years, the capture of CO_2 from flue gas has gained a lot of interests by manufacturers due to international efforts for the reduction of greenhouse gas emissions. Capturing of CO_2 from flue gas presents an additional challenge, even though CO_2 separation processes have been extensively utilized in industry.[1] Various CO_2 capture technologies have received widespread attention particularly physical absorption,[2,3] chemical absorption,[4,5] and adsorption.[6,7] Meanwhile, in screening feasible ILs in capturing CO_2, many factors have to be considered such as cost, stability, corrosiveness, toxicity, and environmental impact.[8,9] Ionic liquids (ILs) with dialkylphosphate anions are biodegradable, less toxic, and greener than other ILs[8] and probable for practical applications because they can be produced in a one-pot reactor under mild conditions with very high yield.[9]

Chemical absorption with aqueous alkanolamine is the most appropriate for CO_2 capture.[4] However, there are some drawbacks which negatively affect the technology efficiency such as high equipment corrosion rate, high energy consumption, and a large absorber volume needed.[10] Thus, ILs with methyldiethanolamine (MDEA) aqueous mixtures have been introduced to overcome these weaknesses.

The application of ILs for CO_2 uptake was improved by blending ILs with mixed amines such as MDEA aqueous solution. Consequently, the addition of ILs increased the CO_2 absorption rate and decreased the vapor pressure of amines.[11] Zhang et al.[12] successfully found functionalized ILs with MDEA aqueous solutions had high CO_2 absorption rate and large uptake capacity. The ILs with MDEA aqueous solutions were more suitable for the industrial device than pure IL.[11] High concentrated aqueous solutions of IL-MDEA with high temperature are efficient for the absorption under low pressure; meanwhile, a lower temperature is recommended for high pressure absorption.[13] However, Ahmady et al.[14] reported that all used ILs in their research reduced the CO_2 loading in aqueous MDEA solutions. The CO_2 loading in all IL+MDEA mixtures increases with increasing CO_2 partial pressure and decreases with increasing temperature.

The determination of physicochemical properties of a substance is crucially important in the design of various physical and chemical processes,[15,16] especially for ILs aqueous mixtures. ILs have an excellent ability to replace organic solvents in chemical processes.[17,18] However, further research remains to be completed in consideration of achieving the precise physicochemical characterization of ILs mixture systems.[19] Therefore, various physicochemical properties of ILs have been studied such as densitiy,[16,19−21] surface tension,[22] and viscosity.[20]

Ternary mixtures of organic solvent−IL−water system have been studied to achieve lower viscosity of solution, which directly lower energy requirements for absorption processes of CO_2.[23,24]

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Knowledge of interface properties, especially the surface and interfacial tension, and its relationship to the ILs chemical nature is crucial to choose the right IL for a specific application.[22] In this study, physical properties of ternary system MDEA−IL−water for density, surface tension, and viscosity are reported.

2. EXPERIMENTAL SECTIONS

2.1. Chemicals. 1-Ethyl-3-methylimidazolium diethylphosphate ([emim][dep], minimum mass purity ≥ 98 %), 1,3-dimethylimidazolium dimethylphosphate ([mmim][dmp], minimum mass purity ≥ 98 %) and pure *N*-methyldiethanolamine (MDEA, purity ≥ 98 %) were purchased from Merck. All chemicals were of analytical grade and used without further purification.

2.2. Sample Preparation. Different mole fractions for aqueous ternary mixtures of [emim][dep]- MDEA-H_2O and [mmim][dmp]-MDEA-H_2O, systems were prepared as tabulated in Table 1.

Table 1. Compositions of ILs, MDEA, and water in Ternary Mixtures[a]

system	mole fraction X_{IL}	X_{MDEA}	X_{H_2O}
(i) [emim][dep] + MDEA + H_2O			
0.1	0.0986	0.1944	0.7070
0.2	0.2091	0.5412	0.2497
0.3	0.3666	0.3676	0.2658
0.4	0.4188	0.2162	0.3650
0.6	0.5973	0.2911	0.1116
0.7	0.6825	0.2899	0.0276
0.8	0.8097	0.1903	0.0000
0.9	0.9134	0.0866	0.0000
1	1.0000	0.0000	0.0000
(ii) [mmim][dmp] + MDEA + H_2O			
0.1	0.0774	0.2789	0.6438
0.2	0.1998	0.4346	0.3657
0.3	0.3247	0.6054	0.0699
0.4	0.3753	0.3888	0.2359
0.5	0.4772	0.2393	0.2835
0.6	0.6367	0.3295	0.0338
0.7	0.6866	0.3126	0.0008
0.8	0.8238	0.1565	0.0197
0.9	0.9305	0.0695	0.0000
1	1.0000	0.0000	0.0000

[a]Standard uncertainties, u are $u(T)$ = 0.01 K, $u(x)$ = 0.0001, and $u(P)$ = 1 kPa.

2.3. Apparatus and Procedure. *Density.* Density measurement of the aqueous ternary mixtures was carried out with densimeter DMA 4500 (Anton Paar, Austria) at various temperature [(293.15 to 343.15) K]. The apparatus is precise within $1.0 \cdot 10^{-4}$ g·cm^{-3}, and the expanded uncertainty of the measurements was estimated to be better than 0.001 g·cm^{-3}. Calibration of the densitometer was performed at atmospheric pressure using dry air and pure water (supplied).

Surface Tension. Surface tension measurement was carried out using KRŰSS Processor Tensiometer K100 using Du Noüy ring method in various temperatures from (293.15 to 343.15) K. The tensiometer has a surface tension range of (1−1000) mN·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. In general, each surface tension value reported was an average of ten measurements. Calibration of the tensiometer was performed by determining the surface tensions of distilled water and pure MDEA and comparing with literature data.

Viscosity. As for the viscosity study, concentrations for aqueous ternary systems of ILs range from (0.1 to 1.0) M were determined. The viscosity measurements were carried out using R/S+ rheometer (Brookfield, USA). The rheometer is a controlled stress (or controlled torque) instrument and was calibrated using standard viscosity oil. Temperature of the solution was maintained within ± 0.1 K. The viscosity was measured with an accuracy less than 3 %. All measurements for each sample were performed in triplicate, and the values were reported as an average. The measurement was taken at atmospheric pressure and temperature range between (293.15 and 343.15) K, with 10 K increment.

3. RESULTS AND DISCUSSION

The validation of data using MDEA was carried out to ensure the accuracy of measurement used in this work. These experimental data of pure compound and comparisons with the literature reviews[24−28] were presented in Table 2.

Table 2. Comparison of Measured Densities, Surface Tensions, and Viscosities with Literature Values for MDEA Pure Components at 0.1 MPa[a]

	density, ρ/g·cm^{-3}		surface tension, γ/mN·m^{-1}		viscosity, η/mPa·s	
T/K	exptl.	lit.	exptl.	lit.	exptl.	lit.
303.15	1.0338	1.03356[b]	37.33	37.29[d]	56.3000	57.50[b]
		1.03410[c]		37.31[e]		57.57[f]
313.15	1.0267	1.02640[b]		N/A	34.4474	35.00[b]
		1.02652[c]				34.78[f]
323.15	1.0189	1.01901[b]		N/A	22.9255	22.00[b]
		1.01887[c]				21.98[f]
333.15		N/A	35.47	35.58[d]	14.9923	N/A

[a]Standard uncertainties, u are $u(T)$ = 0.01 K, $u(x)$ = 0.0001, $u(P)$ = 1 kPa and $u(\gamma)$ = 0.15 mN·m^{-1}. Relative standard uncertainties, u_r are $u_r(\rho)$ = 0.001 and $u_r(\eta)$ = 0.03. [b]Data from ref 22. [c]Data from ref 24. [d]Data from ref 23. [e]Data from ref 25. [f]Data from ref 26.

The measured densities, ρ, surface tensions, γ and viscosities, η for the aqueous ternary mixtures of [emim][dep] and [mmim]-[dmp] with MDEA at T = (293.15, 301.15, 303.15, 313.15, 323.15, 333.15, and 343.15) K over the whole composition range are listed in Tables 3 and 4.

Density. Figures 1 and 2 present the experimental densities of ternary mixtures of [emim][dep] and [mmim][dmp] with MDEA. The findings indicate that ILs ternary mixtures demonstrate temperature-dependence behavior and liquid density decreases linearly with temperature increment. The density of all the compositions always increases with the ILs mole fractions, and decreases with temperature, in agreement with Ciocirlan and co-workers.[29]

The accuracy of the density data were further evaluated by correlation with temperature either by linear, exponential, power or polynomial equations. The best correlation was obtained by linear equation (R^2 = 0.99). Soriano et al.[30] and Jacquemin et al.[31] also reported the linear behavior with temperature being for pure ILs.

Figures 3 and 4 present the experimental densities of ternary mixtures of [emim][dep] and [mmim][dmp] with MDEA in different temperature as a function of mole fraction. It is obvious

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Table 3. Experimental Densities (ρ), Surface Tensions (γ), and Viscosities (η) for Ternary Mixtures at Different Temperatures for [emim][dep]−MDEA−H_2O at 0.1 MPa[a]

	T/K	293.15	301.15	303.15	313.15	323.15	333.15	343.15
	X_i							
				[emim][dep]−MDEA−H_2O				
ρ/g·cm^{-3}	0.1	1.0912	1.0837	1.0815	1.0723	1.0623	1.0501	1.0348
	0.2	1.1004	1.0933	1.0913	1.0814	1.0721	1.0597	1.0438
	0.3	1.1145	1.1074	1.1058	1.0966	1.0863	1.0744	1.0594
	0.4	1.1230	1.1163	1.1142	1.1051	1.0960	1.0846	1.0689
	0.6	1.1290	1.1223	1.1201	1.1124	1.1031	1.0920	1.0768
	0.7	1.1353	1.1287	1.1269	1.1191	1.1101	1.0987	1.0817
	0.8	1.1399	1.1331	1.1314	1.1238	1.1147	1.1034	1.0878
	0.9	1.1460	1.1388	1.1369	1.1292	1.1201	1.1086	1.0923
	1.0	1.1530	1.1452	1.1429	1.1343	1.1245	1.1134	1.0977
γ/mN·m^{-1}	0.1	37.61	37.41	37.36	37.15	36.94	36.78	36.63
	0.2	37.21	37.05	37.01	36.81	36.62	36.47	36.29
	0.3	36.52	36.35	36.31	36.13	35.91	35.68	35.47
	0.4	36.31	36.13	36.09	35.89	35.69	35.48	35.25
	0.6	36.01	35.85	35.81	35.62	35.42	35.24	35.05
	0.7	35.76	35.63	35.60	35.42	35.24	35.08	34.89
	0.8	35.58	35.45	35.41	35.25	35.08	34.91	34.72
	0.9	35.43	35.30	35.26	35.12	34.96	34.78	34.57
	1.0	35.29	35.15	35.12	34.98	34.83	34.64	34.43
η/mPa·s	0.1	41.4339	29.6484	27.0616	17.8278	12.1118	8.7245	6.5644
	0.2	72.0000	52.3037	48.2755	31.9723	22.5031	17.8281	13.5040
	0.3	96.8000	67.1318	63.1407	40.2889	25.6371	19.2430	14.0211
	0.4	114.3000	78.1407	69.6294	48.3817	32.2020	23.7283	16.3701
	0.6	165.3006	112.7113	103.4241	68.0464	42.3307	28.1679	21.6359
	0.7	207.5924	135.2498	120.1645	75.7333	52.2512	35.6182	26.9558
	0.8	258.1360	162.8320	141.8077	81.9080	55.5458	38.8716	23.6362
	0.9	327.8395	201.5538	180.5538	100.5176	61.2818	41.3168	28.5453
	1.0	577.2231	320.2527	281.2820	151.9150	89.0253	58.1505	40.1934

[a]Standard uncertainties, u, are $u(T)$ = 0.01 K, $u(x)$ = 0.0001, and $u(P)$ = 1 kPa, and $u(\gamma)$ = 0.15 mN·m^{-1}. Relative standard uncertainties, u_r, are $u_r(\rho)$ = 0.001 and $u_r(\eta)$ = 0.03.

that the density increases with increasing of mole fraction of ILs. The density of [emim][dep] mixture is less denser compare to [mmim][dmp] mixture. This is in agreement with Pinkert et al.[32] that larger ions result in less denser ILs. We found no previous data as a function of mole fraction for [emim][dep] or [mmim][dmp] or their mixtures with water.

Surface Tension. Figures 5 and 6 show the effects of temperature and composition on surface tension of ternary mixtures of [emim][dep] and [mmim][dmp] with MDEA. To establish the accuracy of the experimental data obtained in this work, surface tension of pure MDEA was determined at (313.15 and 333.15) K, and compare with literature[25,27] which were tabulated in Table 2.

Surface tensions for the whole compositions of aqueous ternary mixtures were obtained at (293.15, 301.15, 303.15, 313.15, 323.15, 333.15, and 343.15) K. The surface tension results are given in Tables 3 and 4 and plotted as a function of temperature in Figures 5 and 6. It is observed that the surface tension results show a linear dependence with a temperature in the composition (mole fractions) considered. The accuracy of the surface tensions data was further assessed by correlating them with temperature by means of a linear equation, resulted good correlations (R^2 = 0.99). A linear variation of surface tension with temperature for ionic liquid is in agreement with Wang et al.[33]

Figures 7 and 8 present the surface tension of ternary mixtures of [emim][dep] with MDEA and [mmim][dmp] with MDEA in different temperature as a function of mole fraction. Ternary mixtures of [emim][dep] with MDEA have lower surface tension than ternary mixtures of [mmim][dmp] with MDEA. This is in agreement with Tariq et al.,[34] who reported the surface tension of the mixtures depends strongly on the alkyl chain length of the ILs. ILs with longer alkyl chains producing mixtures with lower surface tension values at any given composition. Surface tension of [emim][dep] mixture is lower than [mmim][dmp] mixture which have longer alkyl chains.

The surface tension decreases with the increasing of ILs mole fraction for [emim][dep]- MDEA- H_2O. However, the surface tension increases with the decreasing of ILs mole fraction for [mmim][dmp], probably because of high energy binding of [emim][dep] compared to [mmim][dmp]. The surface tensions for these ILs decrease proportionately as the alkyl chain size increases, which are related to the anion influence.[35]

Viscosity. The result of density measurements at the temperature range from (293.15 to 343.15) K of aqueous ternary mixtures of [emim][dep] with MDEA and [mmim]-[dmp] with MDEA are summarized in Tables 2 and 3. As expected, for every measured composition of ternary mixtures, the viscosity decreases with increasing temperature. Nevertheless, this decrease is considerably greater than densities[36] and surface tensions data. In Figures 9 and 10, the results indicate that viscosity of ternary mixtures demonstrate temperature-dependence behavior, reveals the viscosities are more influenced by temperature,[29] which decreases nonlinearly with temperature.

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Table 4. Experimental Densities (ρ), Surface Tensions (γ), and Viscosities (η) for Ternary Mixtures at Different Temperatures for [mmim][dmp]−MDEA−H_2O at 0.1 MPa[a]

	T/K	293.15	301.15	303.15	313.15	323.15	333.15	343.15
	X_i							
				[mmim][dmp]-MDEA-H_2O				
ρ/g·cm^{-3}	0.1	1.1071	1.1006	1.0986	1.0919	1.0852	1.0766	1.0663
	0.2	1.1284	1.1221	1.1207	1.1124	1.1049	1.0958	1.0829
	0.3	1.1486	1.1421	1.1401	1.1295	1.1201	1.1088	1.0967
	0.4	1.1673	1.1606	1.1587	1.1481	1.1396	1.1287	1.1145
	0.5	1.1863	1.1799	1.1778	1.1683	1.1602	1.1489	1.1367
	0.6	1.2048	1.1971	1.1948	1.1871	1.1793	1.1689	1.1561
	0.7	1.2185	1.2109	1.2091	1.1996	1.1918	1.1813	1.1691
	0.8	1.2398	1.2336	1.2321	1.2238	1.2157	1.2052	1.1915
	0.9	1.2473	1.2408	1.2392	1.2313	1.2226	1.2120	1.1987
	1.0	1.2525	1.2461	1.2441	1.2368	1.2274	1.2172	1.2034
γ/mN·m^{-1}	0.1	40.43	40.29	40.24	40.01	39.75	39.38	39.04
	0.2	40.97	40.82	40.79	40.58	40.34	39.96	39.60
	0.3	41.97	41.75	41.68	41.41	41.08	40.69	40.33
	0.4	42.97	42.71	42.63	42.35	42.01	41.58	41.21
	0.5	43.59	43.28	43.19	42.85	42.55	42.13	41.72
	0.6	44.16	43.91	43.82	43.53	43.21	42.78	42.39
	0.7	44.76	44.41	44.32	43.93	43.56	43.15	42.81
	0.8	45.24	44.94	44.85	44.45	43.98	43.61	43.25
	0.9	45.77	45.52	45.44	45.06	44.63	44.26	43.86
	1.0	46.23	45.95	45.87	45.56	45.12	44.81	44.45
η/mPa·s	0.1	46.6395	29.1966	25.2690	16.4867	11.4700	8.2360	6.9800
	0.2	85.6484	53.8526	48.4783	30.1093	20.4690	13.8367	10.6540
	0.3	94.1090	61.7733	55.8309	35.5910	25.4991	17.2703	13.2983
	0.4	105.8037	69.8091	62.4302	40.4197	27.7074	19.0643	14.1079
	0.5	121.8330	81.3819	73.5440	46.1001	32.5985	22.8632	16.4012
	0.6	149.4780	99.8808	91.1669	58.8255	40.4290	27.9771	20.2820
	0.7	163.3437	109.6022	99.7410	63.1879	41.3600	28.9264	21.1065
	0.8	180.3391	117.9302	106.8806	68.5343	42.3108	30.9817	21.7677
	0.9	202.5040	128.0987	115.2849	72.2165	45.1552	30.3650	22.1353
	1.0	229.5487	138.7974	121.6571	77.6727	47.2535	33.6860	23.9578

[a]Standard uncertainties, u, are $u(T)$ = 0.01 K, $u(x)$ = 0.0001, and $u(P)$ = 1 kPa and $u(\gamma)$ = 0.15 mN·m^{-1}. Relative standard uncertainties, u_r, are $u_r(\rho)$ = 0.001 and $u_r(\eta)$ = 0.03.

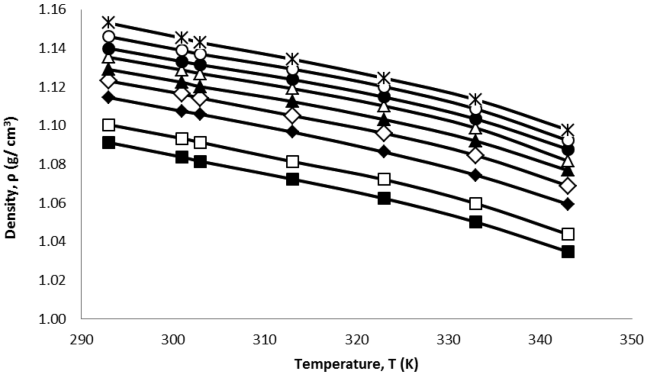

Figure 1. Experimental densities, ρ, of ternary mixtures [emim][dep]−MDEA−H_2O at different mole fractions as a function of temperature. ■, 0.1 X_{IL}; □, 0.2 X_{IL}; ♦, 0.3 X_{IL}; ◊, 0.4 X_{IL}; ▲, 0.6 X_{IL}; Δ, 0.7 X_{IL}; ○, 0.8 X_{IL}; ●, 0.9 X_{IL}; *, 1.0 X_{IL}.

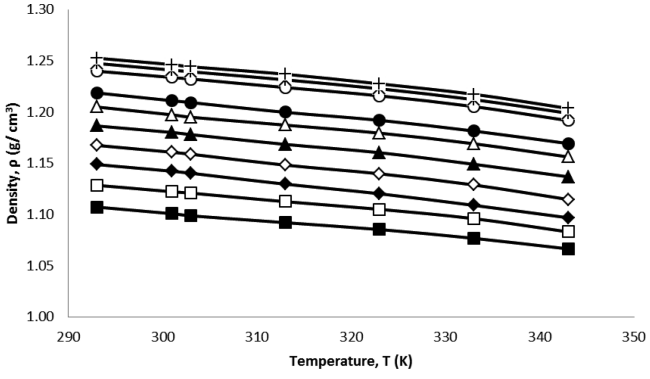

Figure 2. Experimental densities, ρ, of the ternary mixtures [mmim]-[dmp]−MDEA−H_2O at different mole fractions as a function of temperature. ■, 0.1 X_{IL}; □, 0.2 X_{IL}; ♦, 0.3 X_{IL}; ◊, 0.4 X_{IL}; ▲, 0.5 X_{IL}; Δ, 0.6 X_{IL}; ○, 0.7 X_{IL}; ●, 0.8 X_{IL}; *, 0.9 X_{IL}; +, 1.0 X_{IL}.

The accuracy evaluation was done by correlating the results with temperature, and the best correlation was obtained with polynomial equation (R^2 = 0.99). The results corresponding to Baroutian et al.[37] in measurement of physicochemical properties of binary and ternary blends of palm oil + palm biodiesel + diesel fuel at different temperature.

Figures 11 and 12 present the viscosity of ternary mixtures of [emim][dep] with MDEA and [mmim][dmp] with MDEA at different temperatures as a function of mole fraction. At lower ILs mole fraction, viscosity significantly lower, however the value were noticeable at higher mole fraction of ILs. The observation in decrease of viscosity with an increasing of MDEA and water

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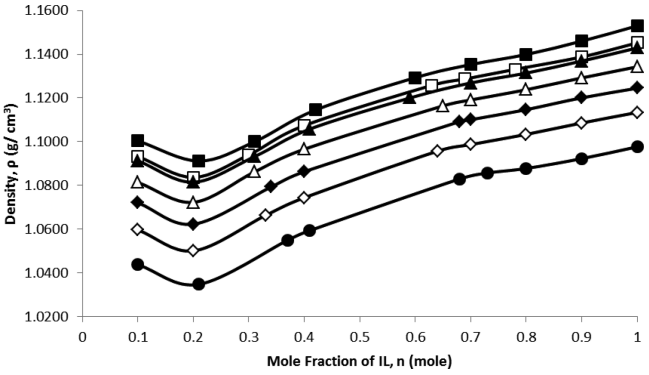

Figure 3. Experimental densities, ρ, of ternary mixtures [emim][dep]−MDEA−H_2O at different temperature as a function of mole fractions. ■, 293.15 K; □, 301.15 K; ▲, 303.15 K; Δ, 313.15 K; ♦, 323.15 K; ◊, 333.15 K; ●, 343.15 K.

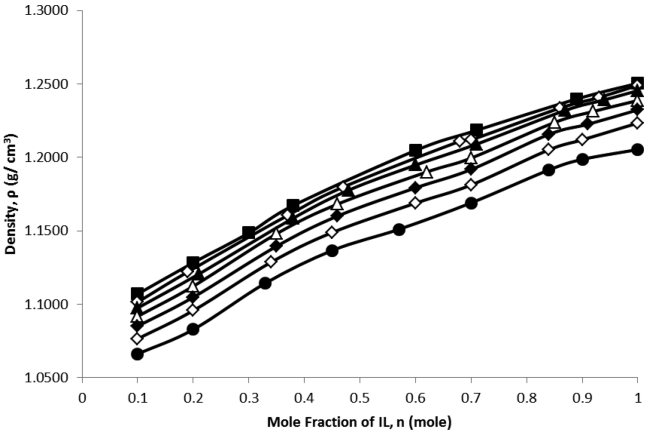

Figure 4. Experimental densities, ρ, of the ternary mixtures [mmim]-[dmp]−MDEA−H_2O at different temperatures as a function of mole fractions. ■, 293.15 K; □, 301.15 K; ▲, 303.15 K; Δ, 313.15 K; ♦, 323.15 K; ◊, 333.15 K; ●, 343.15 K.

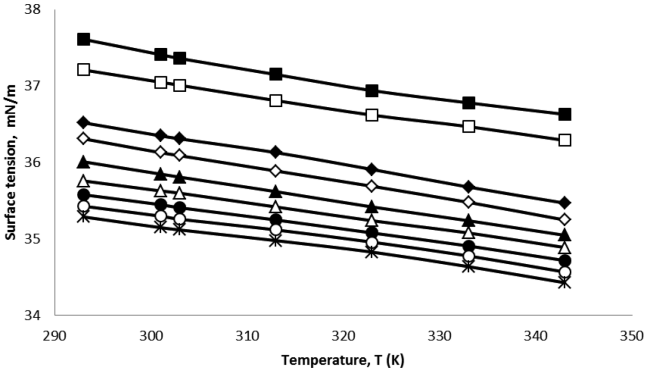

Figure 5. Experimental surface tension, γ, of ternary mixtures [emim][dep]−MDEA−H_2O at different mole fractions as a function of temperature. ■, 0.1 X_{IL}; □, 0.2 X_{IL}; ♦, 0.3 X_{IL}; ◊, 0.4 X_{IL}; ▲, 0.6 X_{IL}; Δ, 0.7 X_{IL}; ○, 0.8 X_{IL}; ●, 0.9 X_{IL}; *, 1.0 X_{IL}.

content is particularly strong in dilute solutions in ILs mixture. Ion-dipole interactions and/or hydrogen bonding between the cation of the IL and water will take place. Conclusively, this weakening of the strong hydrogen bonding interactions between the cation and anion of the IL leads to a higher mobility of the ions and a lower viscosity of the mixture.[38]

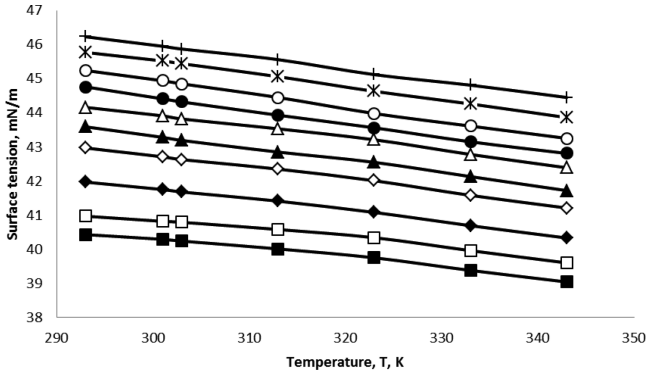

Figure 6. Experimental surface tension, γ, of the ternary mixtures [mmim][dmp]−MDEA−H_2O at different mole fractions as a function of temperature. ■, 0.1 X_{IL}; □, 0.2 X_{IL}; ♦, 0.3 X_{IL}; ◊, 0.4 X_{IL}; ▲, 0.5 X_{IL}; Δ, 0.6 X_{IL}; ○, 0.7 X_{IL}; ●, 0.8 X_{IL}; *, 0.9 X_{IL}; +, 1.0 X_{IL}.

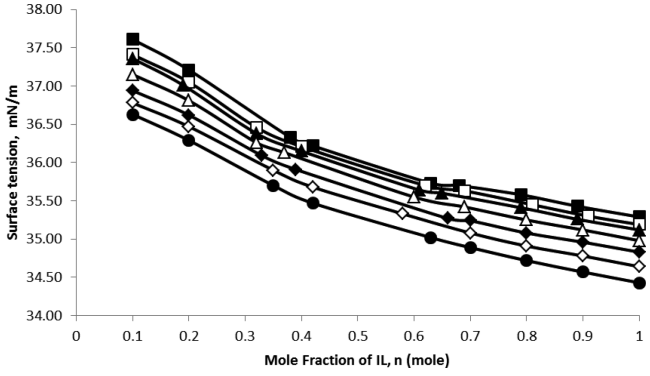

Figure 7. Experimental surface tension, γ, of ternary mixtures [emim][dep]−MDEA−H_2O at different temperatures as a function of mole fractions. ■, 293.15 K; □, 301.15 K; ▲, 303.15 K; Δ, 313.15 K; ♦, 323.15 K; ◊, 333.15 K; ●, 343.15 K.

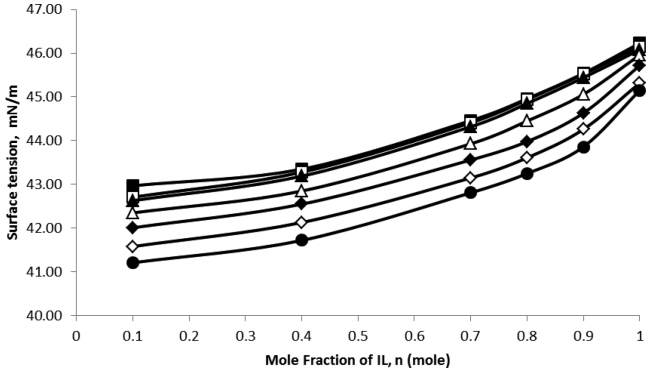

Figure 8. Experimental surface tension, γ, of ternary mixtures [mmim][dmp]−MDEA−H_2O at different temperatures as a function of mole fractions. ■, 293.15 K; □, 301.15 K; ▲, 303.15 K; Δ, 313.15 K; ♦, 323.15 K; ◊, 333.15 K; ●, 343 K.

Figure 13 shows the comparison of the surface tension data between mixtures [emim][dep]−MDEA−H_2O and MEA−MDEA−H_2O[39] at different mole fractions as a function of temperature. The similarities in the decreasing of surface tension data between the series were observed. Fu et al.,[39] who reported that the increase of CO_2 loading tends to increase the surface tensions at given amine concentration for the CO_2 loaded MEA-MDEA aqueous solutions.

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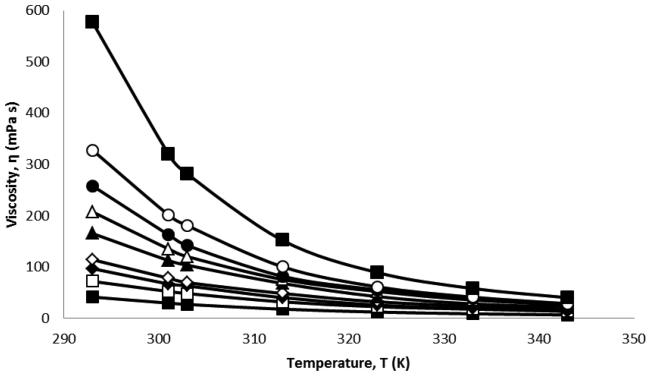

Figure 9. Experimental viscosities, η, of ternary mixtures [emim][dep]−MDEA−H_2O at different mole fractions as a function of temperature. ■, 0.1 X_{IL}; □, 0.2 X_{IL}; ♦, 0.3 X_{IL}; ◊, 0.4 X_{IL}; ▲, 0.6 X_{IL}; Δ, 0.7 X_{IL}; ○, 0.8 X_{IL}; ●, 0.9 X_{IL}; *, 1.0 X_{IL}.

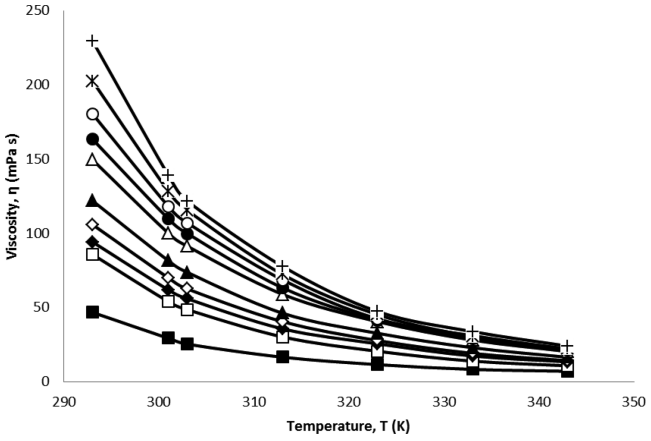

Figure 10. Experimental viscosities, η, of the ternary mixtures [mmim][dmp]−MDEA−H_2O at different mole fractions as a function of temperature. ■, 0.1 X_{IL}; □, 0.2 X_{IL}; ♦, 0.3 X_{IL}; ◊, 0.4 X_{IL}; ▲, 0.5 X_{IL}; Δ, 0.6 X_{IL}; ○, 0.7 X_{IL}; ●, 0.8 X_{IL}; *, 0.9 X_{IL}; +, 1.0 X_{IL}.

Data Correlation Using the Jouyban−Acree Model. *Density.* The Jouyban−Acree model for representing the density of ternary solvents 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 mixture and solvents 1, 2, and 3 at temperature T, respectively, and D_j represent the model constants.[40,41] 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_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-squares

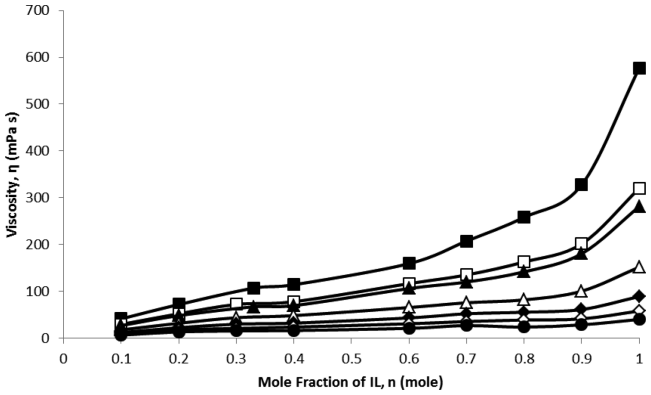

Figure 11. Experimental viscosities, η, of ternary mixtures [emim]-[dep]−MDEA−H_2O at different temperatures as a function of mole fractions. ■, 293.15 K; □, 301.15 K; ▲, 303.15 K; Δ, 313.15 K; ♦, 323.15 K; ◊, 333.15 K; ●, 343.15 K.

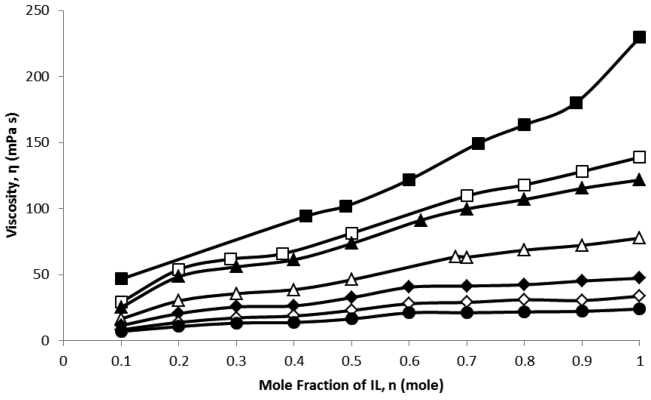

Figure 12. Experimental viscosities, η, of ternary mixtures [mmim]-[dmp]−MDEA−H_2O at different temperatures as a function of mole fractions. ■, 293.15 K; □, 301.15 K; ▲, 303.15 K; Δ, 313.15 K; ♦, 323.15 K; ◊, 333.15 K; ●, 343.15 K.

analysis. The obtained models after excluding nonsignificant model constants ($p > 0.05$), for [emim][dep]−MDEA−H_2O is

$$\ln \rho_{m,T} = x_1 \ln \rho_{1,T} + x_2 \ln \rho_{2,T} + x_3 \ln \rho_{3,T} + 123.812\left[\frac{x_1x_2}{T}\right] - 431.999\left[\frac{x_1x_2(x_1 - x_2)}{T}\right] + 299.190\left[\frac{x_1x_3}{T}\right] - 135.921\left[\frac{x_2x_3}{T}\right] - 1303.665\left[\frac{x_2x_3(x_2 - x_3)}{T}\right] \quad (2)$$

and for [mmim][dmp]−MDEA−H_2O is

$$\ln \rho_{m,T} = x_1 \ln \rho_{1,T} + x_2 \ln \rho_{2,T} + x_3 \ln \rho_{3,T} + 92.957\left[\frac{x_1x_2}{T}\right] + 159.705\left[\frac{x_1x_2(x_1 - x_2)}{T}\right] + 39.817\left[\frac{x_1x_3}{T}\right] + 106.868\left[\frac{x_2x_3}{T}\right] \quad (3)$$

The calculated density values using eqs 2 and 3 against the experimental values are presented in Figures 14 and 15. The accuracy of the calculations was evaluated using absolute percentage error (APER) computed by

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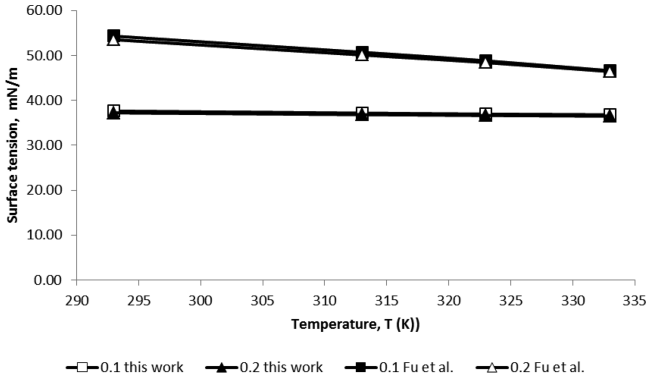

Figure 13. Comparison of experimental surface tension, γ between ternary mixtures [emim][dep]−MDEA−H_2O and MEA−MDEA−H_2O (Fu et al.[39]) at different mole fractions as a function of temperature.

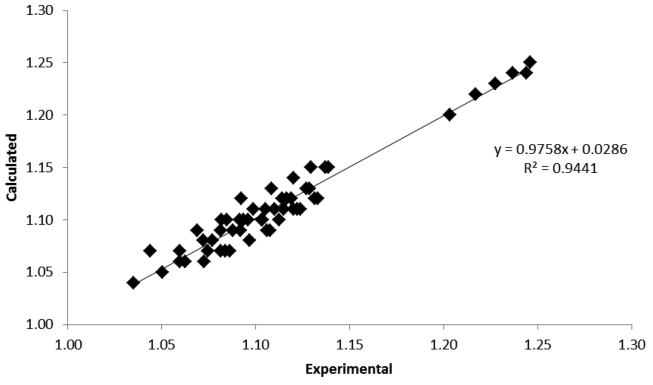

Figure 14. Density values of [emim][dep]−MDEA−H_2O calculated using eq 2 against the corresponding experimental values.

$$\text{APER} = \frac{100}{N}\sum \frac{|\text{Computed} - \text{Experimental}|}{\text{Experimental}} \quad (4)$$

The N in eq 4 is the number of data points in the data set. The APER for [emim][dep]−MDEA−H_2O is 0.8310 % in eq 2 and 1.2183 % for [mmim][dmp]−MDEA−H_2O in eq 3.

Surface Tension. The Jouyban−Acree model for representing the surface tension of ternary solvents is

$$\ln \gamma_{m,T} = x_1 \ln \gamma_{1,T} + x_2 \ln \gamma_{2,T} + x_3 \ln \gamma_{3,T} + x_1x_2 \sum_{j=0}^{2}\left[\frac{A_j(x_1 - x_2)^j}{T}\right] + x_1x_3 \times \sum_{j=0}^{2}\left[\frac{B_j(x_1 - x_3)^j}{T}\right] + x_2x_3 \sum_{j=0}^{2}\left[\frac{C_j(x_2 - x_3)^j}{T}\right] + x_1x_2x_3 \sum_{j=0}^{2}\left[\frac{D_j(x_1 - x_2 - x_3)^j}{T}\right] \quad (5)$$

where $\gamma_{m,T}$, $\gamma_{1,T}$, $\gamma_{2,T}$, and $\gamma_{3,T}$ are the surface tensions of the mixture and solvents 1, 2, and 3 at temperature T, respectively, and D_j represent the model constants.[42] These model constants are computed by regressing $(\ln \gamma_{m,T} - x_1 \ln \gamma_{1,T} - x_2 \ln \gamma_{2,T} - x_3 \ln \gamma_{3,T})$ 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, $x_2x_3(x_2 - x_3)/T$, $x_2x_3(x_2 - x_3)^2/T$, $x_1x_2x_3/T$, $x_1x_2x_3(x\_ - x_2 - x_3)/T$, and $x_1x_2x_3(x_1 - x_2 - x_3)^2/T$ using a no intercept least-squares analysis. The

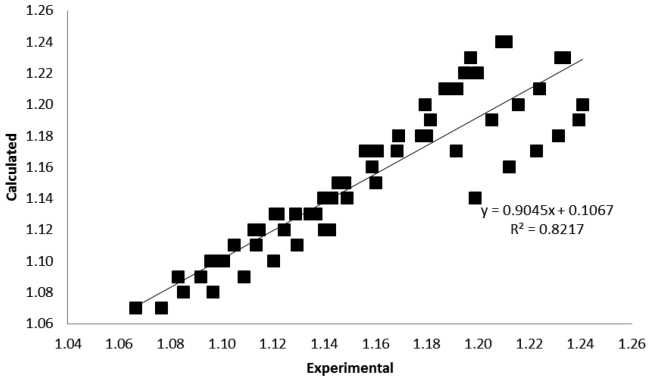

Figure 15. Density values of [mmim][dmp]−MDEA−H_2O calculated using eq 3 against the corresponding experimental values.

obtained models after excluding nonsignificant model constants ($p > 0.05$), for [emim][dep]−MDEA−H_2O is

$$\ln \gamma_{m,T} = x_1 \ln \gamma_{1,T} + x_2 \ln \gamma_{2,T} + x_3 \ln \gamma_{3,T} - 184.940\left[\frac{x_1x_2}{T}\right] + 97.129\left[\frac{x_1x_2(x_1 - x_2)}{T}\right] - 240.749\left[\frac{x_1x_3}{T}\right] + 268.683\left[\frac{x_1x_3(x_1 - x_3)}{T}\right] - 608.197\left[\frac{x_2x_3}{T}\right] + 574.476\left[\frac{x_2x_3(x_2 - x_3)}{T}\right] \quad (6)$$

and for [mmim][dmp]−MDEA−H_2O is

$$\ln \gamma_{m,T} = x_1 \ln \gamma_{1,T} + x_2 \ln \gamma_{2,T} + x_3 \ln \gamma_{3,T} - 171.909\left[\frac{x_1x_2}{T}\right] + 167.099\left[\frac{x_1x_2(x_1 - x_2)}{T}\right] - 106.504\left[\frac{x_1x_3}{T}\right] - 500.764\left[\frac{x_2x_3}{T}\right] + 511.022\left[\frac{x_2x_3(x_2 - x_3)}{T}\right] \quad (7)$$

The calculated density values using eqs 6 and 7 against the experimental values are presented in Figures 16 and 17. The accuracy of the calculations was evaluated using absolute percentage error (APER) computed by eq 4, where N is the number of data points in the data set. The APER for [emim][dep]−MDEA−H_2O is 0.4858 % in eq 6 and 0.9032 % for [mmim][dmp]−MDEA−H_2O in eq 7.

Viscosity. The Jouyban−Acree model for representing the viscosity of ternary solvents is

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} + x_3 \ln \eta_{3,T} + x_1x_2 \sum_{j=0}^{2}\left[\frac{A_j(x_1 - x_2)^j}{T}\right] + x_1x_3 \times \sum_{j=0}^{2}\left[\frac{B_j(x_1 - x_3)^j}{T}\right] + x_2x_3 \sum_{j=0}^{2}\left[\frac{C_j(x_2 - x_3)^j}{T}\right] + x_1x_2x_3 \sum_{j=0}^{2}\left[\frac{D_j(x_1 - x_2 - x_3)^j}{T}\right] \quad (8)$$

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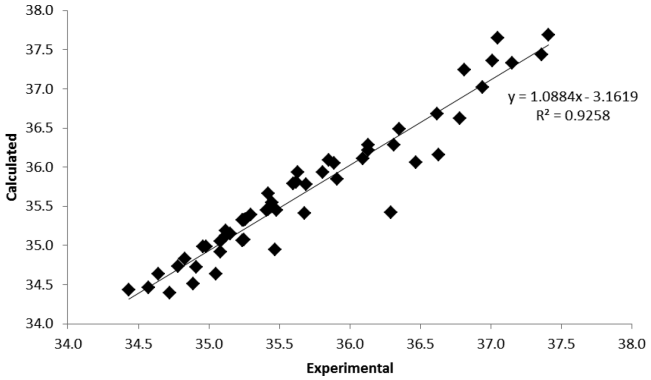

Figure 16. Surface tension values of [emim][dep]−MDEA−H_2O calculated using eq 6 against the corresponding experimental values.

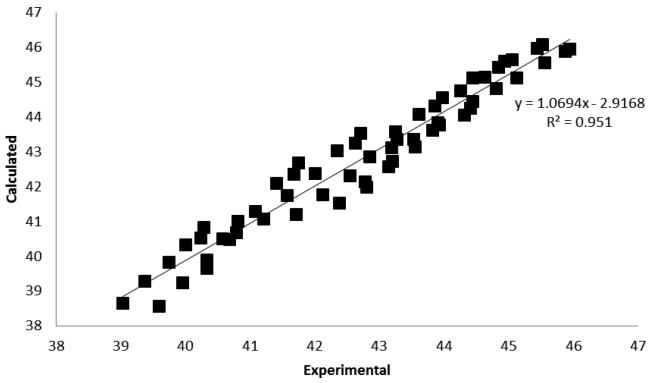

Figure 17. Surface tension values of [mmim][dmp]−MDEA−H_2O calculated using eq 7 against the corresponding experimental values.

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 mixed solvents with respect to solvent composition and temperature by using Jouyban−Acree model.[43,44] 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_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-squares analysis. The obtained models after excluding nonsignificant model constants ($p > 0.05$), for [emim][dep]−MDEA−H_2O is

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} + x_3 \ln \eta_{3,T} + 630.494\left[\frac{x_1x_2}{T}\right] - 2015.538\left[\frac{x_1x_2(x_1 - x_2)}{T}\right] + 2973.430\left[\frac{x_1x_3}{T}\right] + 2354.561\left[\frac{x_1x_3(x_1 - x_3)}{T}\right] - 3066.373\left[\frac{x_2x_3}{T}\right] - 10617.4\left[\frac{x_2x_3(x_2 - x_3)}{T}\right] \quad (9)$$

and for [mmim][dmp]−MDEA−H_2O is

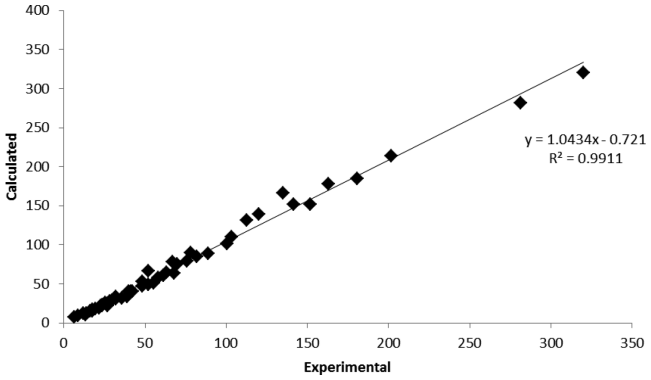

Figure 18. Viscosity values of [emim][dep]−MDEA−H_2O calculated using eq 9 against the corresponding experimental values.

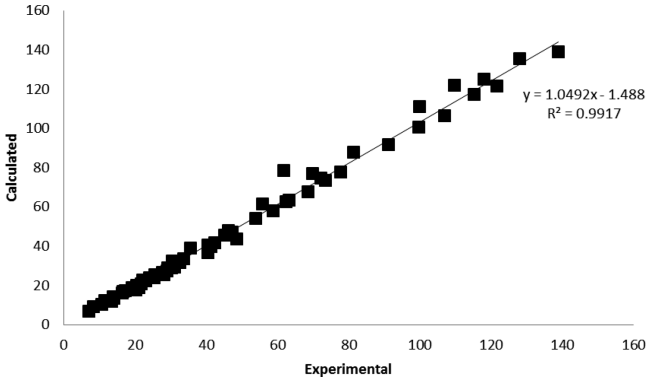

Figure 19. Viscosity values of [mmim][dmp]−MDEA−H_2O calculated using eq 10 against the corresponding experimental values.

$$\ln \eta_{m,T} = x_1 \ln \eta_{1,T} + x_2 \ln \eta_{2,T} + x_3 \ln \eta_{3,T} + 71.289\left[\frac{x_1x_2}{T}\right] + 2581.633\left[\frac{x_1x_2(x_1 - x_2)}{T}\right] - 1651.581\left[\frac{x_1x_3}{T}\right] + 1541.061\left[\frac{x_1x_3(x_1 - x_3)}{T}\right] - 2048.221\left[\frac{x_2x_3}{T}\right] - 2800.601\left[\frac{x_2x_3(x_2 - x_3)}{T}\right] \quad (10)$$

The calculated density values using eqs 9 and 10 against the experimental values are presented in Figures 18 and 19. The accuracy of the calculations was evaluated using absolute percentage error (APER) computed by eq 4, which N is the number of data points in the data set. The APER for [emim][dep]−MDEA−H_2O is 7.0278 % in eq 9 and 4.4109 % for [mmim][dmp]−MDEA−H_2O in eq 10.

4. CONCLUSION

Aqueous ternary mixtures of [emim][dep] and [mmim][dmp] with MDEA were prepared and their physicochemical properties namely densities, ρ, surface tensions, γ, and viscosities, η, in the temperature range from (293.15 to 343.15) K were measured and reported. The ternary mixtures demonstrate temperature-dependent behaviors. Densities and surface tensions of mixtures decreased linearly with temperature, meanwhile viscosities decreased nonlinearly. The accuracy assessment of densities, surface tensions, and viscosities was completed by correlation. The best correlation for density and surface tension data are

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linear equation, and polynomial equation for viscosity. The experimental density, surface tension and viscosity also influenced by mole fractions of ILs. This article enlarges widely the experimental physicochemical properties information about ILs−MDEA−H_2O systems as it includes the behavior at various temperatures commonly found in engineering process. The proposed models provided reasonable accurate results to calculate the density, surface tension and viscosity of ILs−MDEA−H_2O mixtures with respect to their composition and temperature. The overall APER for density, surface tension and viscosity of [emim][dep]−MDEA−H_2O and [mmim][dmp]−MDEA−H_2O are below than 8 %, revealing that the Jouyban−Acree model is able to accurately represent the physicochemical properties data.

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Notes

The authors declare no competing financial interest.

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