

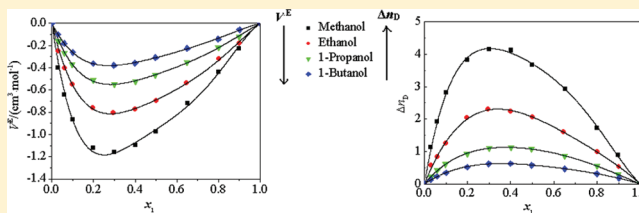
Density, Viscosity, and Refractive Index Properties for the Binary Mixtures of *n*-Butylammonium Acetate Ionic Liquid + Alkanols at Several Temperatures

Yingjie Xu,^{†,‡} Jia Yao,[†] Congmin Wang,[†] and Haoran Li^{*,†}

[†]Department of Chemistry, Zhejiang University, Hangzhou, Zhejiang 310027, China

[‡]Department of Chemistry, Shaoxing University, Shaoxing, Zhejiang 312000, China

ABSTRACT: Densities and viscosities were determined for the binary mixtures of *n*-butylammonium acetate ionic liquid (N4AC) with methanol, ethanol, *n*-propanol, and *n*-butanol at temperatures of (293.15, 298.15, 303.15, 308.15, and 313.15) K under atmospheric pressure. The refractive indices of the above-mentioned binary mixtures were measured at 298.15 K. Excess molar volumes V^E , viscosity deviations $\Delta\eta$, and refractive index deviations Δn_D were obtained from the experimental data and fitted with the Redlich–Kister equation. The correlation results were in good agreement with the experimental data, and optimal fitting parameters were presented. The results were interpreted in terms of interactions and structural factors of N4AC + alkanols mixtures.



1. INTRODUCTION

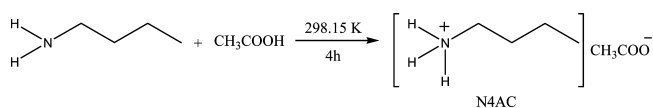
Room-temperature ionic liquids (ILs) are attracting more and more attention due to their unique physicochemical properties and have been applied in many industrial processes, such as organic synthesis,^{1,2} catalytic reactions,^{3–7} CO₂ capture,^{8–10} electrochemistry,^{11,12} and multiphase separations.^{13–18} The thermodynamic properties of the mixtures of ILs with organic molecular liquids are important not only for designing chemical industry separation processes and transport equipment but also for predicting the properties and characteristics of ILs from a theoretical point of view.^{19,20} Recently, the density and viscosity properties of the binary mixtures containing imidazole-,^{20–28} pyridinium-,^{29–32} phosphonium-,^{33,34} and pyrrolidinium-based³⁵ ILs have been widely investigated. However, the experimental data on the density and viscosity properties of new task-specific ILs were rather limited.³⁶

Recently, some simple ammonium ILs have attracted considerable interest in organic synthesis and industry due to their advantages such as easy preparation, cheap cost, and low toxicity. Our group has successfully applied a series of simple ammonium ILs as both acidic catalysts and solvents to produce dialkoxypropanes by cracking reactions,^{37,38} cinnamic acid through the hydrolytic reaction of 1,1,1,3-tetrachloro-3-phenylpropane,³⁹ and unsaturated ketones by the Saucy–Marbet reaction,⁴⁰ eliminating the need for a volatile organic solvent and additional catalyst. Because of their excellent catalytic properties in organic reactions, these physicochemical properties have also attracted the attention of a growing number of scientists. The structural organization of *n*-butylammonium nitrate (N4NO₃) IL aqueous solutions has been investigated using ¹H NMR chemical shifts combined with the local composition model.⁴¹ In spite of the importance of properties of the simple ammonium ILs in different solvent media, only a

small number of density and viscosity properties have been reported in the literature compared with other ILs.^{42–46}

To improve the application of the simple ammonium ILs, it was necessary to study their physicochemical properties with other solvents. In the present study, we chose *n*-butylammonium acetate IL (N4AC) as an example, which is a typical simple ammonium IL and can be used as the acidic catalyst, with its structure was shown in Scheme 1. The densities and

Scheme 1. Molecular Structure and Synthesis of N4AC



viscosities of binary mixtures of N4AC with methanol, ethanol, *n*-propanol and *n*-butanol were measured at temperatures of (293.15, 298.15, 303.15, 308.15, and 313.15) K under atmospheric pressure. The refractive indices were determined at 298.15 K for the above-mentioned binary mixtures. Based on these experimental results, excess molar volumes V^E , viscosity deviations $\Delta\eta$, and refractive index deviations Δn_D were calculated and fitted with the Redlich–Kister equation.

2. EXPERIMENTAL SECTION

Chemicals. Methanol, ethanol, *n*-propanol, *n*-butanol, *n*-butylamine, and acetate acid (analytical reagent grade, with a nominal mass > 99 %) were obtained from Shanghai Chemical Co. Ltd.,

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Table 1. Densities ρ , Viscosities η , and Refractive Indices n_D of the Pure Components at Several Temperatures

component	T/K	$\rho/\text{g}\cdot\text{cm}^{-3}$			$\eta/\text{mPa}\cdot\text{s}$			n_D		
		exptl	lit.	Dr %	exptl	lit.	Dr %	exptl	lit.	Dr %
water	298.15	0.99709	0.997068 ⁴⁸	0.002	0.891	0.890 ³⁵	0.011	1.3329	1.3330 ³⁵	0.007
methanol	293.15	0.79128	0.791218 ³⁶	0.007	0.588	0.5884 ²²	0.068			
	298.15	0.78656	0.786507 ³⁶	0.006	0.557	0.5509 ²²	1.107	1.3265	1.3264 ⁴⁷	0.008
	303.15	0.78183	0.781778 ³⁶	0.006	0.519	0.5154 ²²	0.698			
	308.15	0.77707	0.777028 ³⁶	0.005	0.490	0.4849 ²²	1.052			
	313.15	0.77229	0.772238 ³⁶	0.006	0.462	0.4576 ²²	0.962			
ethanol	293.15	0.78954	0.7890 ²⁹	0.069	1.241	1.216 ⁴⁹	2.056			
	298.15	0.78525	0.7855 ²⁹	0.032	1.128	1.132 ⁴⁹	0.353	1.3594	1.3593 ⁴⁷	0.008
	303.15	0.78093	0.7803 ²⁹	0.081	1.029	1.010 ⁴⁹	1.881			
	308.15	0.77657	0.7760 ²⁹	0.074	0.920	0.907 ⁵⁰	1.433			
	313.15	0.77218	0.7720 ²⁹	0.024	0.842	0.834 ⁴⁹	0.959			
<i>n</i> -propanol	293.15	0.80355	0.8036 ²⁹	0.006	2.229	2.202 ⁴⁹	1.226			
	298.15	0.79955	0.7996 ²⁹	0.006	1.975	1.973 ⁴⁹	0.101	1.3834	1.3833 ⁴⁷	0.007
	303.15	0.79552	0.7956 ²⁹	0.010	1.757	1.733 ⁴⁹	1.385			
	308.15	0.79144	0.7915 ²⁹	0.007	1.568	1.542 ⁵⁰	1.686			
	313.15	0.78734	0.7874 ²⁹	0.008	1.406	1.379 ⁵⁰	1.958			
<i>n</i> -butanol	293.15	0.80970	0.8098 ²⁹	0.012	2.983	2.937 ⁵⁰	1.566			
	298.15	0.80590	0.8060 ²⁹	0.013	2.617	2.569 ⁵⁰	1.868	1.3973	1.3971 ⁴⁷	0.014
	303.15	0.80205	0.8021 ²⁹	0.006	2.306	2.260 ⁵⁰	2.035			
	308.15	0.79817	0.7982 ²⁹	0.004	2.040	1.998 ⁵⁰	2.102			
	313.15	0.79426	0.7943 ²⁹	0.005	1.811	1.784 ⁵⁰	1.513			
N4AC	293.15	0.95961			771.694					
	298.15	0.95644			546.348			1.4426		
	303.15	0.95333			397.170					
	308.15	0.95015			294.586					
	313.15	0.94698			222.241					

Shanghai, China. They were purified by the methods described by our laboratory previously.⁴⁷ The purity of these materials was checked by gas chromatography. The density, viscosity, and refractive index of methanol, ethanol, *n*-propanol, and *n*-butanol were determined and compared with literature values listed in Table 1.

N4AC was prepared from *n*-butylamine and acetate acid according to the procedure reported previously by our laboratory (see Scheme 1).³⁹ Specific processes were as follows: into a 100 mL three-necked flask under vigorous stirring, 0.55 mol of *n*-butylamine was placed, then 0.50 mol of acetate acid was dropped in slowly. The temperature was kept at 25 °C. After the neutralization reaction for 4 h, the excess *n*-butylamine was separated, and then N4AC was obtained and dried under vacuum at 70 °C for at least 48 h before use. The water content was determined to be about 200 ppm by Karl Fischer titration (Mettler Toledo DL32, Switzerland). The structure of N4AC was identified by Fourier transform infrared (FT-IR; Nicolet FT-IR/Nexus470) and ¹H NMR spectroscopy (Bruker, 400 MHz, CDCl₃). IR vibrational frequencies (in cm⁻¹) were as follows: 3039 (⁺N–H stretching vibration), 2961, 2933, and 2875 (C–H stretching vibration), 1633 (C=O stretching vibration), (1557 ⁺N–H bending vibration), and 1467 (C–H bending vibration), which was consistent with the structure of N4AC. The ¹H NMR chemical shifts δ were as follows: 0.94 (t, 3H, $J = 7.36$ Hz, ⁺NH₃CH₂CH₂CH₂CH₃), 1.39 (m, 2H, ⁺NH₃CH₂CH₂CH₂CH₃), 1.63 (m, 2H, ⁺NH₃CH₂CH₂CH₂CH₃), 1.93 (s, 3H, CH₃COO⁻), 2.82 (t, 2H, $J = 7.56$ Hz, ⁺NH₃CH₂CH₂CH₂CH₃), 8.16 (s, 3H, ⁺NH₃CH₂CH₂CH₂CH₃), and the total peak integral in ¹H NMR spectrum was found to correspond for N4AC to a nominal purity higher than 99 %.

Apparatus and Procedure. The binary mixtures of N4AC with methanol, ethanol, *n*-propanol, and *n*-butanol were prepared by using an analytical balance with a precision of $\pm 1.0 \cdot 10^{-5}$ g. The errors in mole fractions of the binary mixtures were less than $\pm 1.0 \cdot 10^{-4}$. All of the samples were prepared immediately before the density, viscosity, and refractive index measurements to avoid the evaporation of the alkanols. The N4AC used in the experiment was not recycled and reused.

The densities of pure materials and binary mixtures were measured with a vibrating-tube densimeter (Anton Paar DMA 5000 M). The uncertainty of density is $\pm 5.0 \cdot 10^{-6}$ g·cm⁻³. The densimeter was calibrated with ultrapure water, which was also listed in Table 1, compared with literature. Two integrated Pt 100 platinum thermometers (uncertainty: 0.01 K) together with Peltier elements provide an extremely precise thermostating of the sample. The overall average relative deviation (Dr %) between density measurements and literature values of alkanols was 0.019 %, according to the data from Table 1.

The viscosities of pure materials and binary mixtures were measured by an Anton Paar AMVn automated microviscometer (reproducibility < 0.5 %, repeatability < 0.1 %),^{30,51} which used the rolling-ball principle. Calibration was carried out using ultrapure water or viscosity standard oils (no. H117; Anton Paar Co). The temperature was controlled by a built-in precise Peltier thermostat within ± 0.01 K. Triplicate measurements of flow times were reproducible within ± 0.02 s. The overall average relative deviation (Dr %) between density measurements and literature values of alkanols was 1.301 %, according to the data from Table 1.

Refractive indices were measured using an Abbe refractometer model WAY-2S, and the temperatures were controlled by a circulating-water bath with the accuracy of ± 0.01 K.

Ultrapure water was used as a reference for calibration. The uncertainty in refractive index was estimated to be $\pm 1.0 \cdot 10^{-4}$. The sample support was rinsed with acetone and dried with a paper towel. The overall average relative deviation (Dr %) between density measurements and literature values of alkanols was 0.009 %, according to the data from Table 1.

3. RESULTS AND DISCUSSION

The experimental values of the density ρ , viscosity η , and refractive index n_D for binary mixtures of N4AC with methanol, ethanol, *n*-propanol, and *n*-butanol at different temperatures under atmospheric pressure are presented in Table 2. The excess molar volumes V^E , viscosity deviations $\Delta\eta$, and refractive index deviations Δn_D were calculated from experimental data according to the following equations:

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2} \quad (1)$$

$$\Delta\eta = \eta - (x_1 \eta_1 + x_2 \eta_2) \quad (2)$$

$$\Delta n_D = n_D - (x_1 n_{D,1} + x_2 n_{D,2}) \quad (3)$$

where x_1 and x_2 are mole fractions of components 1 and 2. ρ_1 , ρ_2 , and ρ in eq 1, η_1 , η_2 , and η in eq 2, and $n_{D,1}$, $n_{D,2}$, and n_D in eq 3 are the densities, viscosities, and refractive indices of pure components 1, 2, and their mixtures, respectively. M_1 and M_2 are molecular weights of components 1 and 2.

The values of V^E , $\Delta\eta$, and Δn_D of the above-mentioned binary mixtures are also listed in Table 2. The results of V^E , $\Delta\eta$, and Δn_D were fitted by the Redlich–Kister polynomial equation:

$$Y^E = x_1 x_2 \sum_{k=0}^M A_k (x_1 - x_2)^k \quad (4)$$

where $Y^E \equiv (V^E, \Delta\eta, \text{ or } \Delta n_D)$, and the coefficients of A_k are adjustable parameters which are obtained by fitting the equations to the experimental values with a least-squares method. M is the degree of the polynomial expansion. The standard relative deviation, s , between the experimental and calculated values was defined in the following equation:

$$s = \left[\sum \left((Y_{\text{exptl}}^E - Y_{\text{calcd}}^E) / Y_{\text{exptl}}^E \right)^2 / N \right]^{1/2} \quad (5)$$

where N is the number of direct experimental data. The values of the parameters A_k together with the standard relative deviation s , for each property Y^E , are given in Table 3.

The excess molar volumes V^E of mixtures versus the mole fraction of N4AC with methanol, ethanol, *n*-propanol, and *n*-butanol at 298.15 K are plotted in Figure 1, which shows that the excess molar volumes are negative over the entire composition range. As can be seen in Figure 1, the absolute values of excess molar volumes ($|V^E|$) follow the sequence: methanol > ethanol > *n*-propanol > *n*-butanol. A minimum value in V^E of four binary mixtures is reached with mole fraction of N4AC near to 0.3.

It is known that the excess molar volumes are the result of several opposing effects. Interactions between like molecules lead to increased V^E values, while negative contributions to V^E arise from interactions between unlike molecules such as ion–dipole and hydrogen bonding or structural effects such as packing.^{21,37,51} N4AC is a typical protic IL and apt to form hydrogen bonds with proton acceptors such as alkanols.

Moreover, the ability to form hydrogen bonding between the alkanols with N4AC follows this order: methanol > ethanol > *n*-propanol > *n*-butanol, and the higher hydrogen bonding interaction leads to larger $|V^E|$ values. On the other hand, the molecular size of the solvents follows this order: methanol < ethanol < *n*-propanol < *n*-butanol, which leads to the most notable packing efficiency between methanol with N4AC and increase of the $|V^E|$ values. To sum up, those are reasons why the $|V^E|$ values for the studied systems follow this order: methanol > ethanol > *n*-propanol > *n*-butanol. A similar phenomenon has been observed by Mokhtarani et al.²¹ and Gonzalez et al.,⁵² the mixtures of alkanols with 1-methyl-3-octylimidazolium nitrate or 1-methyl-3-octylimidazolium chloride also have negative V^E over the entire composition range, and the $|V^E|$ values for the studied systems also decrease with the increase of the alcohol chain length.

It is clear in Figure 2 that the $|V^E|$ values for the binary system of N4AC (1) + methanol (2) increase slightly with the temperature, and a similar case can also be found for the other three binary systems in Table 2. As the temperature increases, the kinetic energy of pure components also increases, which leads to a decrease in the interactions of the pure components.⁵³ The decreased interaction between pure organic molecules results in greater interaction and packing efficiency between alkanols and N4AC, so the contraction in volume increases, and V^E decreases. A similar phenomenon has been observed by Zhou et al.,³⁶ the $|V^E|$ values for binary mixtures of naphthenic acid ILs and ethanol also increase slightly with the temperature.

According to the literature, ILs are generally more viscous than conventional solvents. In most applications, they can be used in mixtures with other less viscous compounds. Therefore, the viscosity of pure ILs and their mixtures with conventional solvents is an important property which is primordial for each industrial process. The viscosity of pure N4AC decreases with the temperature from 771.694 mPa·s at 293.15 K to 222.241 mPa·s at 313.15 K, which are more viscous than pure 1-butyl-3-methylimidazolium hexafluorophosphate²⁰ and 2-hydroxyethylammonium acetate IL,⁴⁶ and less viscous than pure 1-methyl-3-octylimidazolium nitrate²¹ at the same temperature. The viscosities η as well as viscosity deviations $\Delta\eta$ of N4AC with alkanol binary mixtures are listed in Table 2. The viscosity deviations $\Delta\eta$ for binary mixtures of N4AC with methanol at different temperatures are graphically represented in Figure 3, which represents deviations from a rectilinear dependence of viscosity on mole fraction. It can be observed in Figure 3 that the $\Delta\eta$ values are all negative over the whole concentration range for N4AC (1) + methanol (2) mixtures and increase sharply with the temperature. The $\Delta\eta$ values for the other three N4AC (1) + alkanols (2) mixtures have the same negative deviation and similar temperature variation. A minimum value in $\Delta\eta$ is reached with a mole fraction of N4AC near to 0.7 for the four studied binary mixtures.

The refractive index n_D can be used as a measure of the electronic polarizability of a molecule and can provide useful information when studying the interaction between molecules⁵⁴ or their behavior in solution,⁵⁵ and the refractive index deviations Δn_D can be physically interpretable as the deviation of the reduced free volume, which are negatively correlated to V^E values.³⁵ The refractive index deviations of mixtures versus the mole fraction of N4AC with methanol, ethanol, *n*-propanol, and *n*-butanol at 298.15 K are plotted in Figure 4, which presents a positive deviation from ideality over the whole

Table 2. Densities ρ , Viscosities η , Refractive Indices n_D , Excess Molar Volumes V^E , Viscosity Deviations $\Delta\eta$, and Refractive Index Deviations Δn_D for Binary Mixtures at Several Temperatures

x_1	ρ g·cm ⁻³	η mPa·s	n_D	V^E cm ⁻³ ·mol ⁻¹	$\Delta\eta$ mPa·s	Δn_D
xN4AC + (1 - x) Methanol						
T = 293.15 K						
0.0302	0.81476	0.883		-0.386	-22.9648	
0.0597	0.83265	1.237		-0.626	-45.4017	
0.1011	0.85244	1.933		0.844	-76.6241	
0.1992	0.88485	4.825		-1.088	-149.4045	
0.3010	0.90627	11.095		-1.128	-221.6054	
0.4023	0.92099	23.223		-1.063	-287.6087	
0.4995	0.93142	44.755		-0.948	-340.9645	
0.6524	0.94329	115.480		-0.698	-388.2070	
0.7993	0.95152	256.450		-0.423	-360.5209	
0.8988	0.95588	418.491		-0.218	-275.1577	
T = 298.15 K						
0.0302	0.81030	0.820	1.3413	-0.398	-16.2010	0.0113
0.0597	0.82830	1.141	1.3526	-0.641	-32.0110	0.0192
0.1011	0.84833	1.759	1.3665	-0.869	-53.9850	0.0283
0.1992	0.88105	4.283	1.3878	-1.119	-105.0218	0.0382
0.3010	0.90266	9.608	1.4030	-1.160	-155.2396	0.0416
0.4023	0.91752	19.574	1.4144	-1.094	-200.5750	0.0412
0.4995	0.92803	36.676	1.4212	-0.975	-236.4781	0.0367
0.6524	0.94000	92.511	1.4316	-0.719	-264.1411	0.0294
0.7993	0.94831	195.973	1.4365	-0.438	-240.8615	0.0172
0.8988	0.95269	312.521	1.4398	-0.225	-178.5870	0.0090
T = 303.15 K						
0.0302	0.80581	0.763		-0.410	-11.7207	
0.0597	0.82398	1.053		-0.659	-23.1544	
0.1011	0.84420	1.607		-0.892	-39.0185	
0.1992	0.87723	3.828		-1.149	-75.7230	
0.3010	0.89904	8.354		-1.191	-111.5618	
0.4023	0.91403	16.660		-1.121	-143.4457	
0.4995	0.92464	30.448		-0.999	-168.1797	
0.6524	0.93669	69.011		-0.733	-190.2986	
0.7993	0.94507	151.070		-0.443	-166.5114	
0.8988	0.94949	242.558		-0.224	-114.4665	
T = 308.15 K						
0.0302	0.80129	0.712		-0.422	-8.6494	
0.0597	0.81963	0.975		-0.678	-17.0785	
0.1011	0.84005	1.474		-0.918	-28.7527	
0.1992	0.87338	3.437		-1.182	-55.6512	
0.3010	0.89545	7.294		-1.228	-81.7225	
0.4023	0.91053	14.337		-1.153	-104.4784	
0.4995	0.92123	25.586		-1.028	-121.7915	
0.6524	0.93339	53.465		-0.755	-138.9049	
0.7993	0.94183	118.320		-0.456	-117.2549	
0.8988	0.94630	183.852		-0.233	-80.9676	
T = 313.15 K						
0.0302	0.79674	0.666		-0.434	-6.4921	
0.0597	0.81528	0.905		-0.698	-12.8141	
0.1011	0.83589	1.357		-0.944	-21.5504	
0.1992	0.86954	3.099		-1.215	-41.5920	
0.3010	0.89177	6.415		-1.260	-60.8657	
0.4023	0.90704	12.535		-1.187	-77.2372	
0.4995	0.91783	21.649		-1.058	-89.6816	
0.6524	0.93008	47.608		-0.776	-97.6814	
0.7993	0.93859	94.242		-0.468	-83.6588	
0.8988	0.94309	142.668		-0.239	-57.3065	

Table 2. continued

x_1	ρ g·cm ⁻³	η mPa·s	n_D	V^E cm ⁻³ ·mol ⁻¹	$\Delta\eta$ mPa·s	Δn_D
xN4AC + (1 - x) Ethanol						
T = 293.15 K						
0.0319	0.80512	1.644		-0.243	-24.1639	
0.0608	0.81726	2.184		-0.390	-45.8653	
0.0999	0.83176	3.042		-0.533	-75.1921	
0.1977	0.86095	6.773		-0.740	-146.7545	
0.2965	0.88307	13.923		-0.780	-215.7867	
0.4008	0.90146	26.605		-0.752	-283.4227	
0.5005	0.91565	49.217		-0.679	-337.6563	
0.6454	0.93209	113.109		-0.520	-385.3782	
0.8019	0.94597	264.170		-0.308	-354.8651	
0.9009	0.95334	475.323		-0.172	-220.0131	
T = 298.15 K						
0.0319	0.80093	1.488	1.3681	-0.250	-17.0247	0.0058
0.0608	0.81317	1.959	1.3731	-0.401	-32.2936	0.0084
0.0999	0.82777	2.707	1.3804	-0.549	-52.9057	0.0124
0.1977	0.85719	5.903	1.3965	-0.763	-102.9919	0.0204
0.2965	0.87943	11.878	1.4073	-0.803	-150.9288	0.0230
0.4008	0.89794	22.281	1.4152	-0.776	-197.3636	0.0223
0.5005	0.91221	40.200	1.4219	-0.699	-233.8250	0.0207
0.6454	0.92875	90.902	1.4292	-0.535	-262.1090	0.0160
0.8019	0.94274	201.813	1.4360	-0.318	-236.5043	0.0098
0.9009	0.95015	347.838	1.4396	-0.178	-144.4751	0.0052
T = 303.15 K						
0.0319	0.79671	1.349		-0.255	-12.3109	
0.0608	0.80904	1.764		-0.411	-23.3317	
0.0999	0.82374	2.420		-0.563	-38.1960	
0.1977	0.85335	5.176		-0.782	-74.1532	
0.2965	0.87576	10.232		-0.824	-108.2677	
0.4008	0.89440	18.841		-0.795	-140.9562	
0.5005	0.90876	33.212		-0.716	-166.0960	
0.6454	0.92539	72.834		-0.544	-183.8623	
0.8019	0.93948	156.424		-0.321	-162.2535	
0.9009	0.94694	260.752		-0.176	-97.1578	
T = 308.15 K						
0.0319	0.79246	1.226		-0.262	-9.0580	
0.0608	0.80488	1.593		-0.423	-17.1690	
0.0999	0.81969	2.171		-0.579	-28.0957	
0.1977	0.84949	4.557		-0.803	-54.4088	
0.2965	0.87209	8.831		-0.850	-79.1719	
0.4008	0.89081	16.056		-0.816	-102.5615	
0.5005	0.90530	27.757		-0.737	-120.1507	
0.6454	0.92203	59.183		-0.560	-131.2680	
0.8019	0.93622	122.419		-0.332	-113.9790	
0.9009	0.94374	197.777		-0.184	-67.7049	
T = 313.15 K						
0.0319	0.78816	1.117		-0.267	-6.7910	
0.0608	0.80068	1.442		-0.433	-12.8629	
0.0999	0.81560	1.908		-0.595	-21.0788	
0.1977	0.84568	4.013		-0.831	-40.6301	
0.2965	0.86839	7.695		-0.875	-58.8595	
0.4008	0.88718	13.748		-0.835	-75.9076	
0.5005	0.90183	23.371		-0.759	-88.3873	
0.6454	0.91866	48.507		-0.576	-95.3535	
0.8019	0.93294	97.361		-0.340	-81.1720	
0.9009	0.94053	152.815		-0.189	-47.6642	

Table 2. continued

x_1	ρ g·cm ⁻³	η mPa·s	n_D	V^E cm ⁻³ ·mol ⁻¹	$\Delta\eta$ mPa·s	Δn_D
x N4AC + (1 - x) n -Propanol						
$T = 293.15$ K						
0.0315	0.81410	2.862		-0.158	-23.5688	
0.0601	0.82282	3.515		-0.261	-44.9205	
0.0977	0.83338	4.733		-0.361	-72.6985	
0.1980	0.85748	8.602		-0.500	-145.9776	
0.3002	0.877710	17.499		-0.534	-215.7611	
0.4021	0.89470	31.539		-0.508	-280.0752	
0.4970	0.90839	47.475		-0.453	-337.1474	
0.6487	0.92709	108.492		-0.343	-392.8605	
0.7990	0.94252	244.690		-0.206	-372.3518	
0.8981	0.95147	445.203		-0.118	-248.0804	
$T = 298.15$ K						
0.0315	0.81013	2.496	1.3875	-0.160	-16.6015	0.0022
0.0601	0.81891	3.158	1.3912	-0.266	-31.5062	0.0042
0.0977	0.82952	4.116	1.3953	-0.368	-51.0623	0.0061
0.1980	0.85376	7.371	1.4045	-0.511	-102.3873	0.0094
0.3002	0.87410	14.768	1.4122	-0.547	-150.6546	0.0110
0.4021	0.89119	26.079	1.4185	-0.521	-194.7765	0.0113
0.4970	0.90494	38.692	1.4236	-0.465	-233.8146	0.0108
0.6487	0.92374	87.224	1.4305	-0.352	-267.8654	0.0087
0.7990	0.93927	186.830	1.4362	-0.213	-250.1062	0.0055
0.8981	0.94825	326.532	1.4395	-0.120	-164.3432	0.0029
$T = 303.15$ K						
0.0315	0.80612	2.202		-0.160	-11.9914	
0.0601	0.81495	2.773		-0.268	-22.7281	
0.0977	0.82563	3.595		-0.374	-36.8060	
0.1980	0.85000	6.367		-0.521	-73.6795	
0.3002	0.87046	12.513		-0.558	-107.9660	
0.4021	0.88765	22.228		-0.532	-138.5154	
0.4970	0.90148	31.742		-0.474	-166.5192	
0.6487	0.92037	70.415		-0.356	-187.8320	
0.7990	0.93601	148.780		-0.213	-168.9173	
0.8981	0.94503	244.830		-0.117	-112.0465	
$T = 308.15$ K						
0.0315	0.80209	1.955		-0.163	-8.8301	
0.0601	0.81095	2.447		-0.272	-16.7171	
0.0977	0.82170	3.157		-0.381	-27.0489	
0.1980	0.84622	5.493		-0.534	-54.0913	
0.3002	0.86680	10.660		-0.572	-78.8863	
0.4021	0.88410	18.480		-0.547	-100.9044	
0.4970	0.89801	26.400		-0.489	-120.7865	
0.6487	0.91700	56.622		-0.366	-135.0163	
0.7990	0.93273	116.881		-0.221	-118.8125	
0.8981	0.94183	186.414		-0.123	-78.3126	
$T = 313.15$ K						
0.0315	0.79801	1.672		-0.163	-6.6855	
0.0601	0.80691	2.167		-0.274	-12.5116	
0.0977	0.81772	2.828		-0.386	-20.1799	
0.1980	0.84242	4.770		-0.547	-40.3997	
0.3002	0.86311	9.154		-0.586	-58.6172	
0.4021	0.88052	15.721		-0.562	-74.5582	
0.4970	0.89453	22.126		-0.503	-89.1251	
0.6487	0.91360	46.763		-0.374	-98.0204	
0.7990	0.92943	93.136		-0.224	-84.8799	
0.8981	0.93861	144.838		-0.128	-55.0787	

Table 2. continued

x_1	ρ	η	n_D	V^E	$\Delta\eta$	Δn_D
	$\text{g}\cdot\text{cm}^{-3}$	$\text{mPa}\cdot\text{s}$		$\text{cm}^{-3}\cdot\text{mol}^{-1}$	$\text{mPa}\cdot\text{s}$	
$x\text{N4AC} + (1-x)\text{ }n\text{-Butanol}$						
$T = 293.15\text{ K}$						
0.0290	0.81708	3.564		-0.101	-21.7184	
0.0583	0.82415	4.397		-0.181	-43.4125	
0.1003	0.83362	5.588		-0.260	-74.4640	
0.1981	0.85355	9.789		-0.355	-145.4629	
0.2981	0.87149	17.144		-0.372	-214.9970	
0.3990	0.88771	28.334		-0.349	-281.3748	
0.4970	0.90199	50.227		-0.302	-334.7907	
0.6489	0.92184	107.737		-0.221	-394.0326	
0.7984	0.93921	225.041		-0.137	-391.7172	
0.9003	0.94980	389.538		0.056	-305.5476	
$T = 298.15\text{ K}$						
0.0290	0.81328	3.190	1.3999	-0.100	-15.2002	0.0013
0.0583	0.82036	3.806	1.4023	-0.180	-30.5182	0.0024
0.1003	0.82988	4.823	1.4053	-0.262	-52.3082	0.0035
0.1981	0.84990	8.389	1.4115	-0.361	-101.9327	0.0052
0.2981	0.86792	14.412	1.4170	-0.379	-150.2952	0.0062
0.3990	0.88421	23.481	1.4217	-0.356	-196.0928	0.0063
0.4970	0.89853	41.010	1.4256	-0.309	-231.8309	0.0058
0.6489	0.91848	86.028	1.4314	-0.224	-269.3949	0.0047
0.7984	0.93595	170.607	1.4366	-0.142	-266.1509	0.0031
0.9003	0.94658	287.993	1.4399	-0.056	-204.1675	0.0018
$T = 303.15\text{ K}$						
0.0290	0.80945	2.787		-0.100	-10.9738	
0.0583	0.81655	3.318		-0.181	-22.0145	
0.1003	0.82611	4.165		-0.264	-37.7288	
0.1981	0.84622	7.151		-0.366	-73.3708	
0.2981	0.86433	12.181		-0.386	-107.8368	
0.3990	0.88069	19.600		-0.362	-140.2626	
0.4970	0.89509	33.212		-0.314	-165.3332	
0.6489	0.91512	69.137		-0.225	-189.3806	
0.7984	0.93268	138.102		-0.139	-179.4815	
0.9003	0.94337	216.861		-0.052	-140.9575	
$T = 308.15\text{ K}$						
0.0290	0.80557	2.455		-0.099	-8.0713	
0.0583	0.81270	2.908		-0.181	-16.1914	
0.1003	0.82223	3.636		-0.259	-27.7341	
0.1981	0.84252	6.123		-0.373	-53.8657	
0.2981	0.86072	10.388		-0.395	-78.8623	
0.3990	0.87717	16.501		-0.373	-102.2691	
0.4970	0.89162	28.451		-0.323	-118.9783	
0.6489	0.91175	56.308		-0.231	-135.5535	
0.7984	0.92940	109.794		-0.144	-125.8288	
0.9003	0.94016	167.147		-0.057	-98.2847	
$T = 313.15\text{ K}$						
0.0290	0.80167	2.164		-0.098	-6.0475	
0.0583	0.80882	2.592		-0.181	-12.0846	
0.1003	0.81846	3.170		-0.268	-20.7613	
0.1981	0.83878	5.297		-0.378	-40.2175	
0.2981	0.85708	8.894		-0.404	-58.6881	
0.3990	0.87362	14.002		-0.382	-75.8434	
0.4970	0.88815	23.979		-0.333	-87.4808	
0.6489	0.90837	45.886		-0.238	-99.0834	
0.7984	0.92610	87.652		-0.146	-90.3205	
0.9003	0.93693	129.841		-0.058	-70.6120	

composition range for all of these systems and are negatively correlated to V^E values. The magnitude of deviation Δn_D

observed is obtained according this order: methanol > ethanol > *n*-propanol > *n*-butanol. A maximum value in Δn_D of four

Table 3. Fitted Parameters of Equation 4 and Standard Relative Deviation (*s*) for the Binary Mixtures at Several Temperatures

	A_0	A_1	A_2	A_3	<i>s</i>
<i>x</i> N4AC + (1 - <i>x</i>) Methanol					
<i>T</i> = 293.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-3.7112	2.4123	-3.2263	3.3343	0.0565
$\Delta\eta/\text{mPa}\cdot\text{s}$	-1341.4740	-900.0886	-824.6006	-643.7715	0.0493
<i>T</i> = 298.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-3.8194	2.4770	-3.3242	3.4205	0.0560
$\Delta\eta/\text{mPa}\cdot\text{s}$	-931.2759	-588.8279	-485.0483	-337.1464	0.0420
Δn_D	0.1483	-0.0806	0.0811	-0.0846	0.0003
<i>T</i> = 303.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-3.9109	2.5645	-3.3827	3.5523	0.0562
$\Delta\eta/\text{mPa}\cdot\text{s}$	-673.5456	-440.9845	-252.7125	-107.3166	0.0094
<i>T</i> = 308.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-4.0252	2.6490	-3.4878	3.6179	0.0566
$\Delta\eta/\text{mPa}\cdot\text{s}$	-490.8964	-315.5875	-160.9691	-49.1407	0.0116
<i>T</i> = 313.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-4.1393	2.7170	-3.5767	3.7449	0.0573
$\Delta\eta/\text{mPa}\cdot\text{s}$	-356.8749	-200.0164	-109.5879	-62.2541	0.0162
<i>x</i> N4AC + (1 - <i>x</i>) Ethanol					
<i>T</i> = 293.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-2.6755	1.6975	-1.8878	1.4925	0.0441
$\Delta\eta/\text{mPa}\cdot\text{s}$	-1357.0758	-1012.1704	-528.3617	-78.2681	0.0194
<i>T</i> = 298.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-2.7533	1.7639	-1.9618	1.4994	0.0431
$\Delta\eta/\text{mPa}\cdot\text{s}$	-938.2855	-657.0206	-303.8565	-25.9110	0.0187
Δn_D	0.0826	-0.0535	0.0303	0.0002	0.0004
<i>T</i> = 303.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-2.8189	1.8338	-1.9691	1.5533	0.0417
$\Delta\eta/\text{mPa}\cdot\text{s}$	-666.8658	-443.3968	-173.2521	11.2742	0.0205
<i>T</i> = 308.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-2.9001	1.8871	-2.0315	1.5670	0.0418
$\Delta\eta/\text{mPa}\cdot\text{s}$	-482.0299	-301.8621	-105.7770	13.8411	0.0190
<i>T</i> = 313.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-2.9796	1.9561	-2.1032	1.5837	0.0403
$\Delta\eta/\text{mPa}\cdot\text{s}$	-354.5107	-208.6584	-61.4639	-18.0397	0.0187
<i>x</i> N4AC + (1 - <i>x</i>) <i>n</i> -Propanol					
<i>T</i> = 293.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.7910	1.2331	-1.3074	0.8903	0.0373
$\Delta\eta/\text{mPa}\cdot\text{s}$	-1351.8517	-1065.4344	-698.7149	-221.1393	0.0102
<i>T</i> = 298.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.8401	1.2607	-1.3251	0.8939	0.0321
$\Delta\eta/\text{mPa}\cdot\text{s}$	-935.5161	-702.1464	-427.0915	-121.3448	0.0109
Δn_D	0.0429	-0.0181	0.0109	-0.0080	0.0001
<i>T</i> = 303.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.8753	1.3197	-1.2928	0.8808	0.0273
$\Delta\eta/\text{mPa}\cdot\text{s}$	-663.8870	-468.7216	-254.0472	-60.5184	0.0075
<i>T</i> = 308.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.9298	1.3632	-1.3195	0.8295	0.0286
$\Delta\eta/\text{mPa}\cdot\text{s}$	-481.7244	-325.9566	-159.4885	-27.0502	0.0086
<i>T</i> = 313.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.9820	1.4284	-1.3147	0.7450	0.0283
$\Delta\eta/\text{mPa}\cdot\text{s}$	-354.6676	-227.8272	-98.4255	-7.9731	0.0089
<i>x</i> N4AC + (1 - <i>x</i>) <i>n</i> -Butanol					
<i>T</i> = 293.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.2071	0.9332	-0.8927	0.7271	0.0279
$\Delta\eta/\text{mPa}\cdot\text{s}$	-1320.3929	-969.9828	-1127.7459	-914.5164	0.0649
<i>T</i> = 298.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.2329	0.9635	-0.8884	0.6742	0.0373
$\Delta\eta/\text{mPa}\cdot\text{s}$	-915.0510	-644.5952	-732.1942	-598.2069	0.0598
Δn_D	0.0232	-0.0113	0.0093	-0.0014	0.0001

Table 3. continued

	A_0	A_1	A_2	A_3	s
		$T = 303.15 \text{ K}$			
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.2525	1.0058	-0.8437	0.6724	0.0432
$\Delta\eta/\text{mPa}\cdot\text{s}$	-650.8554	-424.4814	-467.1493	-405.0951	0.0653
		$T = 308.15 \text{ K}$			
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.2903	1.0607	-0.8212	0.5117	0.0404
$\Delta\eta/\text{mPa}\cdot\text{s}$	-470.06335	-289.5268	-310.9103	-275.2953	0.0613
		$T = 313.15 \text{ K}$			
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-1.3281	1.0743	-0.8073	0.5393	0.0365
$\Delta\eta/\text{mPa}\cdot\text{s}$	-346.8387	-202.1969	-214.7319	-195.5612	0.0600

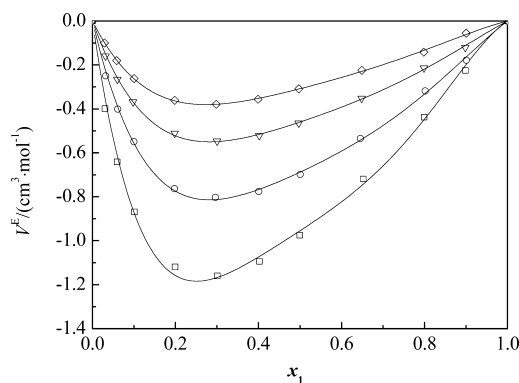


Figure 1. Excess molar volume V^E vs mole fraction for $\{x_1\text{N4AC} + (1 - x_1)\text{ alkanol}\}$ mixtures at 298.15 K: \square , methanol; \circ , ethanol; ∇ , *n*-propanol; \diamond , *n*-butanol. The symbols represent experimental values, and the solid curves represent the values calculated from the Redlich–Kister equation.

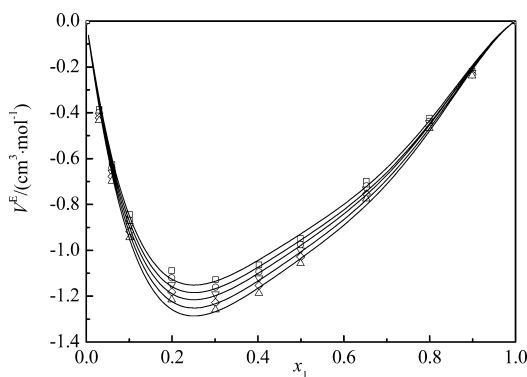


Figure 2. Excess molar volume V^E vs mole fraction for $\{x_1\text{N4AC} + (1 - x_1)\text{ methanol}\}$ mixtures: \square , 293.15 K; \circ , 298.15 K; ∇ , 303.15 K; \diamond , 308.15 K; \triangle , 313.15 K. The symbols represent experimental values, and the solid curves represent the values calculated from the Redlich–Kister equation.

binary systems is also reached with mole fraction of N4AC near $x_1 \approx 0.3$, which is consistent with the V^E . Compared with the maximum absolute values of V^E and Δn_D of the studied systems at 298.15 K, we can find that the high $|V^E|$ values (methanol > ethanol > *n*-propanol > *n*-butanol) correspond to the high Δn_D values (methanol > ethanol > *n*-propanol > *n*-butanol). As it stated above, Δn_D and V^E must be somehow related: if V^E is negative, then there will be less available free volume than in an ideal solution, and photons will be more likely to interact with the molecules or ions constituting the mixture. As a result, light will travel at a weaker velocity in the concerned medium, and its refractive index will be higher than in an ideal solution.³⁵

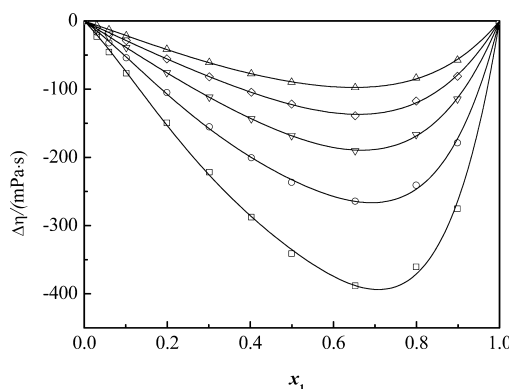


Figure 3. Viscosity deviations $\Delta\eta$ vs mole fraction for $\{x_1\text{N4AC} + (1 - x_1)\text{ methanol}\}$ mixtures: \square , 293.15 K; \circ , 298.15 K; ∇ , 303.15 K; \diamond , 308.15 K; \triangle , 313.15 K. The symbols represent experimental values, and the solid curves represent the values calculated from the Redlich–Kister equation.

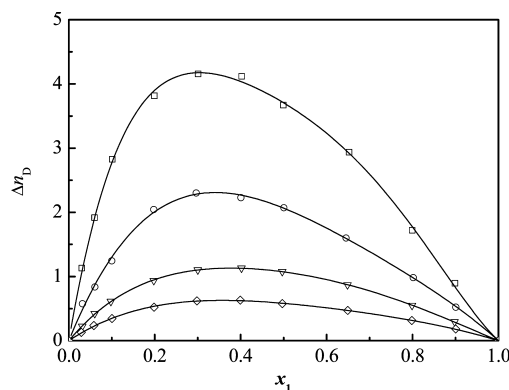


Figure 4. Refractive index deviations Δn_D vs mole fraction for $\{x_1\text{N4AC} + (1 - x_1)\text{ alkanol}\}$ mixtures at 298.15 K: \square , methanol; \circ , ethanol; ∇ , *n*-propanol; \diamond , *n*-butanol. The symbols represent experimental values, and the solid curves represent the values calculated from the Redlich–Kister equation.

4. CONCLUSIONS

In the present research, the experimental densities and viscosities of N4AC and its binary systems with methanol, ethanol, *n*-propanol, and *n*-butanol have been measured at temperatures (293.15, 298.15, 303.15, 308.15, and 313.15) K and atmospheric pressure. The refractive indices of above-mentioned mixtures have been measured at 298.15 K and atmospheric pressure. The excess molar volumes V^E , viscosity deviations $\Delta\eta$, and refractive index deviations Δn_D have been obtained from experimental data and fitted by the Redlich–Kister equation. The estimated coefficients and standard

relative deviation values were also presented. It was found that the excess molar volumes of N4AC + alkanol binary mixtures were negative, and their absolute values increased slightly with temperature and decreased with increasing the alcohol chain length. Meanwhile, the refractive index deviations have positive deviations from ideal solution and also decreased with increasing the alcohol chain length. When the mole fraction of N4AC near $x_1 = 0.3$, both V^E and Δn_D of N4AC + alkanol binary mixtures have extreme points. The viscosity deviations of the studied binary mixtures have negative deviations, and their absolute values decreased sharply as increasing the temperature, with the minima lying nearly at $x_1 = 0.7$.

AUTHOR INFORMATION

Corresponding Author

*E-mail: lihr@zju.edu.cn. Fax: +86 571 87951895.

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REFERENCES

- (1) Welton, T. Room-temperature ionic liquids. Solvents for synthesis and catalysis. *Chem. Rev.* **1999**, *99*, 2071–2083.
- (2) Lee, J. S.; Mayes, R. T.; Luo, H. M.; Dai, S. Ionothermal carbonization of sugars in a protic ionic liquid under ambient conditions. *Carbon* **2010**, *48*, 3364–3368.
- (3) Guan, W.; Wang, C.; Yun, X.; Hu, X.; Wang, Y.; Li, H. A mild and efficient oxidation of 2,3,6-trimethylphenol to trimethyl-1,4-benzoquinone in ionic liquids. *Catal. Commun.* **2008**, *9*, 1979–1981.
- (4) Wang, C.; Guan, W.; Xie, P.; Yun, X.; Li, H.; Hu, X.; Wang, Y. Effects of ionic liquids on the oxidation of 2,3,6-trimethylphenol to trimethyl-1,4-benzoquinone under atmospheric oxygen. *Catal. Commun.* **2009**, *10*, 725–727.
- (5) Wang, Y.; Li, H.; Wang, C.; Jiang, H. Ionic liquids as catalytic green solvents for cracking reactions. *Chem. Commun.* **2004**, *17*, 1938–1939.
- (6) Wasserscheid, P.; Keim, W. Ionic liquids - New “solutions” for transition metal catalysis. *Angew. Chem., Int. Ed.* **2000**, *39*, 3772–3789.
- (7) Xu, X.; Wang, C.; Li, H.; Wang, Y.; Sun, W.; Shen, Z. Effects of imidazolium salts as cocatalysts on the copolymerization of CO₂ with epoxides catalyzed by (salen)(CrCl)-Cl-III complex. *Polymer* **2007**, *48*, 3921–3924.
- (8) Wang, C.; Luo, H.; Jiang, D.; Li, H.; Dai, S. Carbon dioxide capture by superbase-derived protic ionic liquids. *Angew. Chem., Int. Ed.* **2010**, *49*, 5978–5981.
- (9) Wang, C.; Mahurin, S. M.; Luo, H.; Baker, G. A.; Li, H. R.; Dai, S. Reversible and robust CO₂ capture by equimolar task-specific ionic liquid-superbase mixtures. *Green Chem.* **2010**, *12*, 870–874.
- (10) Wang, C.; Luo, H.; Luo, X.; Li, H.; Dai, S. Equimolar CO₂ capture by imidazolium-based ionic liquids and superbase systems. *Green Chem.* **2010**, *12*, 2019–2023.
- (11) Huang, J.; Baker, G. A.; Luo, H.; Hong, K.; Li, Q.; Bjerrum, N. J.; Dai, S. Bronsted acidic room temperature ionic liquids derived from *N,N*-dimethylformamide and similar protophilic amides. *Green Chem.* **2006**, *8*, 599–602.
- (12) Sun, X.; Dai, S. Electrochemical investigations of ionic liquids with vinylene carbonate for applications in rechargeable lithium ion batteries. *Electrochim. Acta* **2010**, *55*, 4618–4626.
- (13) Li, Q.; Zhang, J.; Lei, Z.; Zhu, J.; Zhu, J.; Huang, X. Selection of ionic liquids as entrainers for the separation of ethyl acetate and ethanol. *Ind. Eng. Chem. Res.* **2009**, *48*, 9006–9012.
- (14) Orchilles, A. V.; Miguel, P. J.; Vercher, E.; Martinez-Andreu, A. Ionic liquids as entrainers in extractive distillation: Isobaric vapor-liquid equilibria for acetone + methanol + 1-ethyl-3-methylimidazolium

trifluoromethanesulfonate. *J. Chem. Eng. Data* **2007**, *52*, 141–147.

- (15) Mokhtarani, B.; Gmehling, J. (Vapour + liquid) equilibria of ternary systems with ionic liquids using headspace gas chromatography. *J. Chem. Thermodyn.* **2010**, *42*, 1036–1038.

- (16) Huang, J.; Luo, H.; Liang, C.; Jiang, D.; Dai, S. Advanced liquid membranes based on novel ionic liquids for selective separation of olefin/paraffin via olefin-facilitated transport. *Ind. Eng. Chem. Res.* **2008**, *47*, 881–888.

- (17) Mahurin, S. M.; Lee, J. S.; Baker, G. A.; Luo, H. M.; Dai, S. Performance of nitrile-containing anions in task-specific ionic liquids for improved CO₂/N₂ separation. *J. Membr. Sci.* **2010**, *353*, 177–183.

- (18) Gomez, E.; Dominguez, I.; Calvar, N.; Domínguez, Á. Separation of benzene from alkanes by solvent extraction with 1-ethylpyridinium ethylsulfate ionic liquid. *J. Chem. Thermodyn.* **2010**, *42*, 1234–1239.

- (19) Li, W.; Zhang, Z.; Han, B.; Hu, S.; Xie, Y.; Yang, G. Effect of water and organic solvents on the ionic dissociation of ionic liquids. *J. Phys. Chem. B* **2007**, *111*, 6452–6456.

- (20) Fan, W.; Zhou, Q.; Sun, J.; Zhang, S. Density, excess molar volume, and viscosity for the methyl methacrylate + 1-butyl-3-methylimidazolium hexafluorophosphate ionic liquid binary system at atmospheric pressure. *J. Chem. Eng. Data* **2009**, *54*, 2307–2311.

- (21) Mokhtarani, B.; Sharifi, A.; Mortaheb, H. R.; Mirzaei, M.; Mafi, M.; Sadeghian, F. Densities and viscosities of pure 1-methyl-3-octylimidazolium nitrate and its binary mixtures with alcohols at several temperatures. *J. Chem. Eng. Data* **2010**, *55*, 3901–3908.

- (22) Fan, W.; Zhou, Q.; Zhang, S.; Yan, R. Excess molar volume and viscosity deviation for the methanol + methyl methacrylate binary system at $T = (283.15 \text{ to } 333.15) \text{ K}$. *J. Chem. Eng. Data* **2008**, *53*, 1836–1840.

- (23) Zhang, S.; Li, M.; Chen, H.; Wang, J.; Zhang, J.; Zhang, M. Determination of physical properties for the binary system of 1-ethyl-3-methylimidazolium tetrafluoroborate + H₂O. *J. Chem. Eng. Data* **2004**, *49*, 760–764.

- (24) Mokhtarani, B.; Mojtahedi, M. M.; Mortaheb, H. R.; Mafi, M.; Yazdani, F.; Sadeghian, F. Densities, refractive indices, and viscosities of the ionic liquids 1-methyl-3-octylimidazolium tetrafluoroborate and 1-methyl-3-butylimidazolium perchlorate and their binary mixtures with ethanol at several temperatures. *J. Chem. Eng. Data* **2008**, *53*, 677–682.

- (25) Mokhtarani, B.; Sharifi, A.; Mortaheb, H. R.; Mirzaei, M.; Mafi, M.; Sadeghian, F. Density and viscosity of 1-butyl-3-methylimidazolium nitrate with ethanol, 1-propanol, or 1-butanol at several temperatures. *J. Chem. Thermodyn.* **2009**, *41*, 1432–1438.

- (26) Wang, J. J.; Tian, Y.; Zhao, Y.; Zhuo, K. A volumetric and viscosity study for the mixtures of 1-*n*-butyl-3-methylimidazolium tetrafluoroborate ionic liquid with acetonitrile, dichloromethane, 2-butanone and *N,N*-dimethylformamide. *Green Chem.* **2003**, *5*, 618–622.

- (27) Rebelo, L. P. N.; Najdanovic-Visak, V.; Visak, Z. P.; da Ponte, M. N.; Szydłowski, J.; Cerdeirina, C. A.; Troncoso, J.; Romani, L.; Esperanca, J. M. S. S.; Guedes, H. J. R.; de Sousa, H. C. A detailed thermodynamic analysis of [C(4)mim][BF₄] + water as a case study to model ionic liquid aqueous solutions. *Green Chem.* **2004**, *6*, 369–6381.

- (28) Mohammed, T. Z.; Hemaya, S. Volumetric and Speed of Sound of Ionic Liquid, 1-Butyl-3-methylimidazolium hexafluorophosphate with acetonitrile and methanol at $T = (298.15 \text{ to } 318.15) \text{ K}$. *J. Chem. Eng. Data* **2005**, *50*, 1694–1699.

- (29) Mokhtarani, B.; Sharifi, A.; Mortaheb, H. R.; Mirzaei, M.; Mafi, M.; Sadeghian, F. Density and viscosity of pyridinium-based ionic liquids and their binary mixtures with water at several temperatures. *J. Chem. Thermodyn.* **2009**, *41*, 323–329.

- (30) Sanchez, L. G.; Espel, J. R.; Onink, F.; Meindersma, G. W.; De Haan, A. B. Density, viscosity, and surface tension of synthesis grade imidazolium, pyridinium, and pyrrolidinium based room temperature ionic liquids. *J. Chem. Eng. Data* **2009**, *54*, 2803–2812.

- (31) Gomez, E.; Calvar, N.; Dominguez, A.; Macedo, E. A. Synthesis and temperature dependence of physical properties of four

pyridinium-based ionic liquids: Influence of the size of the cation. *J. Chem. Thermodyn.* **2010**, *42*, 1324–1329.

(32) Xu, Y.; Huang, C.; Li, H. Excess Molar Volume, Refractive index and viscosity of *N*-butylpyridinium tetrafluoroborate ionic liquid + methanol binary system. *J. Chem. Eng. Chin. Univ.* **2011**, *25*, 370–375.

(33) Kilaru, P.; Baker, G. A.; Scovazzo, P. Density and surface tension measurements of imidazolium-, quaternary phosphonium-, and ammonium-based room-temperature ionic liquids: Data and correlations. *J. Chem. Eng. Data* **2007**, *52*, 2306–2314.

(34) Tariq, M.; Forte, P. A. S.; Gomes, M. F. C.; Lopes, J. N. C.; Rebelo, L. P. N. Densities and refractive indices of imidazolium- and phosphonium-based ionic liquids: Effect of temperature, alkyl chain length, and anion. *J. Chem. Thermodyn.* **2009**, *41*, 790–798.

(35) Anouti, M.; Vigeant, A.; Jacquemin, J.; Brigueux, C.; Lemordant, D. Volumetric properties, viscosity and refractive index of the protic ionic liquid, pyrrolidinium octanoate, in molecular solvents. *J. Chem. Thermodyn.* **2010**, *42*, 834–845.

(36) Zhou, Q.; Song, Y.; Yu, Y.; He, H.; Zhang, S. Density and excess molar volume for binary mixtures of naphthenic acid ionic liquids and ethanol. *J. Chem. Eng. Data* **2010**, *55*, 1105–1108.

(37) Jiang, H.; Wang, C.; Li, H.; Wang, Y. Preparation of dialkoxypropanes in simple ammonium ionic liquids. *Green Chem.* **2006**, *8*, 1076–1079.

(38) Wang, C.; Guo, L.; Li, H.; Wang, Y.; Weng, J.; Wu, L. Preparation of simple ammonium ionic liquids and their application in the cracking of dialkoxypropanes. *Green Chem.* **2006**, *8*, 603–607.

(39) Weng, J.; Wang, C.; Li, H.; Wang, Y. Novel quaternary ammonium ionic liquids and their use as dual solvent-catalysts in the hydrolytic reaction. *Green Chem.* **2006**, *8*, 96–99.

(40) Wang, C.; Zhao, W.; Li, H.; Guo, L. Solvent-free synthesis of unsaturated ketones by the Saucy-Marbet reaction using simple ammonium ionic liquid as a catalyst. *Green Chem.* **2009**, *11*, 843–847.

(41) Zhu, X.; Wang, Y.; Li, H. The structural organization in aqueous solutions of ionic liquids. *AIChE J.* **2009**, *55*, 198–205.

(42) Alvarez, V. H.; Mattedi, S.; Pastor, M. M.; Aznar, M.; Iglesias, M. Thermophysical properties of binary mixtures of {ionic liquid 2-hydroxy ethylammonium acetate + (water, methanol, or ethanol)}. *J. Chem. Thermodyn.* **2011**, *43*, 997–1010.

(43) Iglesias, M.; Torres, A.; Gonzalez-Olmos, R.; Salvatierra, D. Effect of temperature on mixing thermodynamics of a new ionic liquid: {2-hydroxy ethylammonium formate (2-HEAF) + short hydroxylic solvents}. *J. Chem. Thermodyn.* **2008**, *40*, 119–133.

(44) Kurnia, K. A.; Taib, M. M.; Mutalib, M. I. A.; Murugesan, T. Densities, refractive indices and excess molar volumes for binary mixtures of protic ionic liquids with methanol at $T = 293.15$ to 313.15 K. *J. Mol. Liq.* **2011**, *159*, 211–219.

(45) Taib, M. M.; Murugesan, T. Densities and excess molar volumes of binary mixtures of bis(2-hydroxyethyl)ammonium acetate + water and monoethanolamine + bis(2-hydroxyethyl)ammonium acetate at temperatures from (303.15 to 353.15) K. *J. Chem. Eng. Data* **2010**, *55*, 5910–5913.

(46) Kurnia, K. A.; Wilfred, C. D.; Murugesan, T. Thermophysical properties of hydroxyl ammonium ionic liquids. *J. Chem. Thermodyn.* **2009**, *41*, 517–521.

(47) Wang, C.; Li, H.; Zhu, L.; Han, S. Excess molar volume of binary mixtures of dehydrolinalool + alkanols at 308.15 K. *Fluid Phase Equilib.* **2001**, *189*, 129–133.

(48) Wang, Y.; Yan, W. Excess Molar Volumes of 1,3-diethyl propanedioate with methanol, ethanol, propan-1-ol, propan-2-ol, butan-2-ol, 2-methyl-propan-1-ol, and pentan-1-ol at $T = (288.15, 298.15, 313.15, \text{ and } 328.15)$ K. *J. Chem. Eng. Data* **2010**, *55*, 4029–4032.

(49) Trenzado, J.; Romano, E.; Segade, L.; Caro, M. N.; González, E.; Galván, S. Densities and viscosities of four binary diethyl carbonate + 1-alcohol systems from (288.15 to 313.15) K. *J. Chem. Eng. Data* **2011**, *56*, 2841–2848.

(50) Sadeghi, R.; Azizpour, S. Volumetric, compressibility, and viscometric measurements of binary mixtures of poly(vinylpyrrolidone) + water, + methanol, + ethanol, + acetonitrile, + 1-propanol, + 2-propanol, and + 1-butanol. *J. Chem. Eng. Data* **2011**, *56*, 167–174.

(51) Yu, Z.; Gao, H.; Wang, H.; Chen, L. Densities, viscosities, and refractive properties of the binary mixtures of the amino acid ionic liquid [bmim][Ala] with methanol or benzylalcohol at $T = (298.15 \text{ to } 313.15)$ K. *J. Chem. Eng. Data* **2011**, *56*, 2877–2883.

(52) Gonzalez, E. J.; Alonso, L.; Dominguez, A. Physical properties of binary mixtures of the ionic liquid 1-methyl-3-octylimidazolium chloride with methanol, ethanol, and 1-propanol at $T = (298.15, 313.15, \text{ and } 328.15)$ K and at $P = 0.1$ MPa. *J. Chem. Eng. Data* **2006**, *51*, 1446–1452.

(53) Zhong, Y.; Wang, H.; Diao, K. Densities and excess volumes of binary mixtures of the ionic liquid 1-butyl-3-methylimidazolium hexafluorophosphate with aromatic compound at $T = (298.15 \text{ to } 313.15)$ K. *J. Chem. Thermodyn.* **2007**, *39*, 291–296.

(54) Hirschfelder, J. O.; Curtiss, C. F.; Bird, R. B. *Molecular theory of gases and liquids*; Wiley: London, 1969.

(55) Israelachvili, J. N. *Intermolecular and surface forces*, 2nd ed.; Academic Press: London, 1992.