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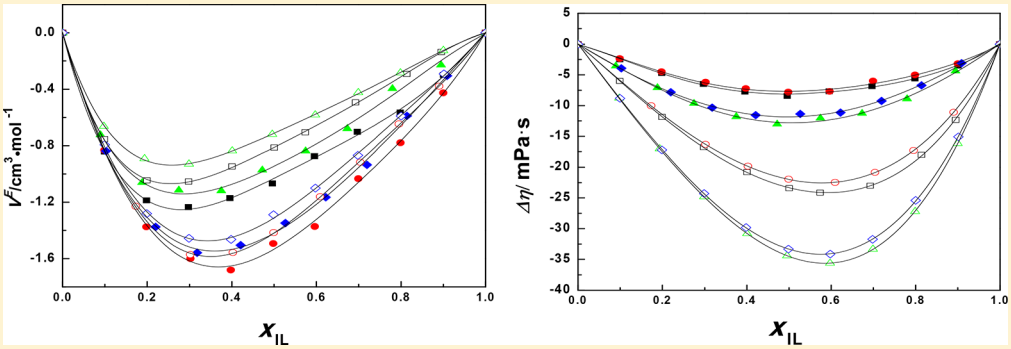
Excess Molar Volume and Viscosity Deviation of $[C_2mim][NTf_2]$/$[C_4mim][NTf_2]$ + DMC/DEC

Qingshan Liu,*[,†,‡] Liwei Zhao,[†] Qige Zheng,[‡] Lin Mou,[‡] and Pengfei Zhang[‡]

[†]College of Land and Environment, and [‡]College of Science, Shenyang Agricultural University, Shenyang 110866, People's Republic of China

Supporting Information



ABSTRACT: Two hydrophobic ionic liquids (ILs) 1-alkyl-3-methylimidazolium bis[(trifluoromethyl)sulfonyl]imide ($[C_2mim][NTf_2]$ and $[C_4mim][NTf_2]$) were synthesized. The binary mixtures of the two ILs were prepared with dimethyl carbonate (DMC) or diethyl carbonate (DEC) over the entire mole fraction range, respectively. The density and viscosity of the binary mixtures were determined by an automated DMA 5000 M and a Lovis 2000 ME Anton Paar Rotational Stabinger Viscometer at temperatures ranging from 288.15 to 328.15 K. The uncertainties of the density and viscosity are $u_r(\rho) = 0.00120$ and $u_r(\eta) = 0.01$. The excess molar volumes and viscosity deviations were calculated for the four binary mixtures by the measurement values. The effects of the extension of the alkyl chain of the cation and anion on the above properties are discussed. The effects of the methyl introduction on the cation was also discussed with the literature. The excess molar volumes and viscosity deviations were fitted versus mole fraction according to the Redlich−Kister equation. The thermal expansion coefficients of the systems were calculated according to the density values.

1. INTRODUCTION

Because of the outstanding physicochemical properties, such as low melting temperature, good solvation, negligible vapor pressure, high electrical conductivity, good thermal stability, and designability, etc.,[1−3] ionic liquids (ILs) have been applied in industrial and academic areas.

Because of the hydrophobicity, the hydrophobic ILs have attracted much more concern from industry and the academic community. As the hydrophobic group, the bis(trifluoromethylsulfonyl)imide($[NTf_2]^-$) anion has been used for synthesizing the ILs. This type of ILs has exhibited much more unique properties, such as negligible vapor pressure at ambient temperatures and excellent thermal stability. So, the systematic study of this type of ILs physicochemical properties has been probed by some groups, especially the imdazolium type ILs.[4−15]

However, viscosity is a limiting factor in the application of ionic liquids,[16] which reduces transport capabilities and retards the chemical processes that occur in these solvents. It is essential for increasing the potential application of ILs in industry and academia to study the physicochemical properties of the binary mixtures in more detail. For $[C_2mim][NTf_2]$ and $[C_4mim][NTf_2]$ ionic liquids, the density, viscosity, and conductivity et al. have been studied by some groups.[6,7,9,11,13] The binary mixtures have also been studied with the traditional solvents by some groups.[4,5,8,10,12,15] The traditional solvents are ethyl acetate and methanol,[4] *N*-methyl-2-pyrrolidone and ethanol,[16] triton,[8] propylene carbonate,[10] and 1-propanol and 1-butanol.[15]

Dimethyl carbonate (DMC) and diethyl carbonate (DEC), as a class of security substitutes for hazardous chemicals, have also attracted much more attention by scholars both at home and abroad. DMC and DEC have also been used in many fields as environmentally benign chemicals, such as lithium batteries, water emulsifiable formulations, synthesis of chemicals, and as solvents for extractive, natural, synthetic resin, or polymers, etc. The binary mixtures of DMC or DEC with other solvents have also been studied by some groups.[17−27]

In 2011,[28] we studied the electrical conductivity of pyridinium type ILs with traditional solvents. The solvents are methanol, ethanol, acetonitrile, and propylene carbonate. The

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Table 1. Provenance, Mass Fraction Purity, CAS No., Water Content, and Br^- Content of the Used Materials[a]

chemical name	provenance	CASRN	purification method	water content	Br^- content	final mass fraction
$[C_2mim][Br]$	Lanzhou Institute of Chemical Physics	65039-08-9	none			≥0.99[b]
$[C_4mim][Br]$	Lanzhou Institute of Chemical Physics	85100-77-2	none			≥0.99[b]
DMC	Aladdin	616-38-6	none			>0.99[b]
DEC	Aladdin	105-58-8	none			>0.99[b]
$Li[NTf_2]$	Solvay (ZhenJiang) Chemicals Co., LTD	90076-65-6	none			>0.999[b]
$[C_2mim][NTf_2]$	synthesized	174899-82-2	water wash	276×10^{-6}[c]	$<50 \times 10^{-6}$[d]	>0.99[e]
$[C_4mim][NTf_2]$	synthesized	174899-83-3	water wash	324×10^{-6}[c]	$<50 \times 10^{-6}$[d]	>0.99[e]

[a]$Li[NTf_2]$, lithium bis[(trifluoromethyl)sulfonyl]imide; $[C_2mmim][NTf_2]$, 1-ethyl-3-dimethylimidazolium bis[(trifluoromethyl)sulfonyl]imide; $[C_4mmim][NTf_2]$, 1-butyl-3-dimethylimidazolium bis[(trifluoromethyl)sulfonyl]imide. [b]As stated by the supplier. [c]Determined by Karl Fischer method. [d]Determined by $AgNO_3/HNO_3$ solution. [e]Determined by NMR spectra.

system exhibited a substantially higher conductivity after the addition of the solvents in ILs.

Recently, we have also prepared the four binary mixtures of 1-alkyl-2,3-methylimidazolium bis[(trifluoromethyl)sulfonyl]-imide ($[C_2mmim][NTf_2]$ or $[C_4mmim][NTf_2]$) with DMC or DEC.[29] The density and viscosity were measured in the whole composition. The excess molar volume and viscosity deviation were calculated according to the density and viscosity.

On the basis of the above, the four binary mixtures of $[C_2mim][NTf_2]$ or $[C_4mim][NTf_2]$ have been prepared with DMC or DEC. The density and viscosity of the systems have been determined in the whole composition. The excess molar volume and viscosity deviation were calculated and fitted by the Redlich−Kister equations to help better understand the types of interactions involved in the solutions. The effect of the extension of the alkyl chain and methyl group introduction of the cation on these properties is also discussed with literature values.[29] The thermal expansion coefficients were also calculated according to the density values of the solution. This work is a contribution to enlarge the existing database of these properties needed to optimize the structural and interaction parameters of group-contribution models.

2. EXPERIMENTAL SECTION

2.1. Materials. The materials of 1-ethyl-3-methylimidazolium bromide ($[C_2mim][Br]$), 1-butyl-3-methylimidazolium bromide ($[C_4mim][Br]$), and lithium bis[(trifluoromethyl)-sulfonyl]imide ($Li[NTf_2]$) were used for the ionic liquids synthesis. The provenance and mass fraction purity of the used materials are listed in Table 1.

2.2. Preparation of ILs $[C_2mim][NTf_2]$ and $[C_4mim][NTf_2]$. $[C_2mim][NTf_2]$ and $[C_4mim][NTf_2]$ were synthesized in the distilled water and dichloromethane by the traditional ion exchange reaction.[30,31] The 30 g of $[C_2mim][Br]$ (or $[C_4mim][Br]$) was dissolved with distilled water and placed in a 250 mL flask. The 50 mL of dichloromethane was also added into the 250 mL flask. The equivalent amount of $LiNTf_2$ salt was also added into the flask at room temperature and stirred for 1 h. The bottom solution was washed several times with distilled water for more than 10 times. The Br^- were detected by $AgNO_3/HNO_3$ solution (the content was estimated to lower than 50×10^{-6} mass fraction). The compounds are colorless liquid, and the final product was finally dried in high vacuum (<1mbar) for 48 h at 353 K before the binary mixtures preparation. The final products of the ILs $[C_2mim][NTf_2]$ and $[C_4mim][NTf_2]$ were characterized by ^1H NMR spectroscopy (see the Figures S1 and S2 of the Supporting Information) and the purities were estimated better than 99% by mass according to NMR spectra. The structures of $[C_2mim][NTf_2]$, $[C_4mim][NTf_2]$, DMC, and DEC are shown in Figure 1.

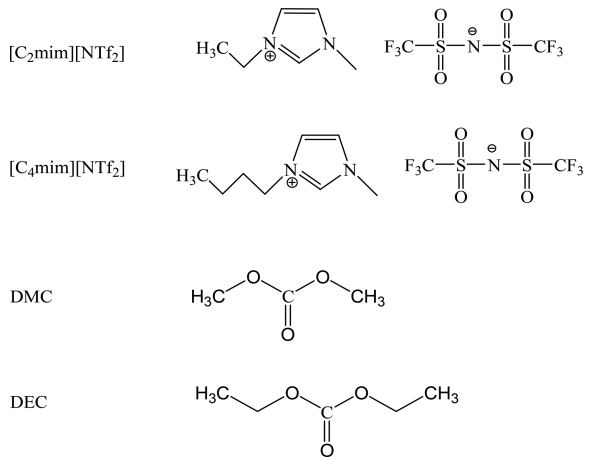

Figure 1. Structures of $[C_2mim][NTf_2]$, $[C_4mim][NTf_2]$, DMC, and DEC.

2.3. Water Content. The water content of $[C_2mim][NTf_2]$ and $[C_4mim][NTf_2]$ was determined by a Karl Fischer coulometric titrator (C20S, Mettler Toledo). The water content is 276×10^{-6} and 324×10^{-6} of the mass fraction $[C_2mim][NTf_2]$ and $[C_4mim][NTf_2]$, respectively. These values are also listed in Table 1.

2.4. Binary Mixtures Preparation. The binary mixtures of $[C_2mim][NTf_2]$ or $[C_4mim][NTf_2]$ + DMC or DEC were prepared by the weighing amounts of $[C_2mim][NTf_2]$ or $[C_4mim][NTf_2]$ and DMC or DEC, covering the whole composition range of the mixture, using a JJ224BC electronic digital balance (uncertainty 1×10^{-4} g), and the uncertainty of the mole fraction is better than 0.0002. A 3 mL sample was used to measure the properties.

2.5. Density and Viscosity. The density and viscosity values of the binary mixtures (including the pure $[C_2mim][NTf_2]$, $[C_4mim][NTf_2]$, DMC, and DEC) were measured from 288.15 to 328.15 K by an automated DMA 5000 M and a Lovis 2000 ME Anton Paar Rotational Stabinger Viscometer with cylinder geometry. The principle is based on a modified Couette according to a rapidly rotating outer tube and a relative slowly rotating inner measuring bob. The viscometer−densimeter was calibrated before the properties measurements with degassed water (heating and cooling) and standard oil. The values were recorded every 5 K, the temperature was controlled to be ±0.02 K, and each value is the average of the three times and the uncertainty is better than 0.00120 for density at 0.68

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Table 2. Compared Values of Density, ρ/g·cm^{-3}, and Viscosity, η/mPa·s, of [C_2mim][NTf_2], [C_4mim][NTf_2], DMC, and DEC with Literature Values at Atmospheric Pressure (p = 101.3 kPa)[a]

		ρ		η	
	T/K	exp	lit.	exp	lit.
[C_2mim] [NTf_2]	298.15	1.51839	1.518057[4](0.022), 1.518251[5](0.009), 1.51834[6](0.003), 1.51837[7](0.001)	33.52	32.49[4](3.073), 33.36[5](0.477)
	303.15	1.51334	1.512880[4](0.030), 1.513232[5](0.007), 1.511324[8](0.133)	28.35	27.68[4](2.363), 28 × 10[5](0.882), 27.1[9](4.409)
	308.15	1.50830	1.507841[4](0.030), 1.508204[5](0.006), 1.506269[8](0.135)	24.15	23.29[4](3.561), 23.93[5](0.911)
	313.15	1.50328	1.502822[4](0.030), 1.503162[5](0.008), 1.50333[6](−0.003), 1.503330[7](−0.003)	20.76	20.16[4](2.890), 20.66[5](0.482), 19.4[9](6.551)
	318.15	1.49827	1.497823[4](0.030), 1.498194[5](0.005), 1.496215[8](0.137)	17.90	17.48[4](2.346), 17.98 [5](−0.447)
	323.15	1.49328	1.492845[4](0.029), 1.493330[5](−0.003), 1.493332[7](−0.003), 1.491207[8](0.139)	15.93	15.34[4](3.704), 15.80[5](0.816), 14.90[9](6.466)
[C_4mim] [NTf_2]	293.15	1.44084	1.43927[10](0.109), 1.43889[11](0.135), 1.4425[12](−0.115), 1.44142[13](−0.040)	61.32	62.08[10](−1.239), 63.8[13](−4.044)
	298.15	1.43601	1.43430[10](0.119), 1.43410[11](0.133), 1.4377[12](−0.118), 1.43664[13](−0.044)	48.93	50.05[10](−2.289), 50.9[13](−4.026)
	303.15	1.43121	1.42940[10](0.126), 1.42931[11](0.133), 1.4329[12](−0.118), 1.43186[13](−0.045), 1.4289[14](0.1614)	39.84	41.24[10](−3.514)
	308.15	1.42643	1.42457[10](0.130), 1.42454[11](0.132), 1.4280[12](−0.110), 1.4241[14](0.1633), 1.4270[15](−0.040)	32.91	
	313.15	1.42166	1.41961[10](0.144), 1.41978[11](0.132), 1.4232[12](−0.108) 1.42234[13](−0.048), 1.4194[14](0.1590)	27.56	28.28[10](−2.612)
	318.15	1.41691	1.41467[10](0.158), 1.41504[11](0.132), 1.4185[12](−0.112), 1.4146[14](0.1630), 1.4174[15](−0.035)	23.33	
	323.15	1.41217	1.40965[10](0.178), 1.41031[11](0.132), 1.4137[12](−0.108), 1.41287[13](−0.050), 1.4099[14](0.1607)	19.97	20.64[10](−3.355), 20.5[13](−2.654)
DMC	288.15	1.07639	1.07716[18](−0.072)	0.669	
	293.15	1.06986	1.06969[17](0.016), 1.07018[18](−0.030), 1.069923[19](−0.006), 1.06954[20](0.030), 1.069834[21](0.002) 1.0696[22](0.024) 1.06990[23](−0.004)	0.626	0.623[17](0.479), 0.6173[20](1.390), 0.624[21](0.319), 0.62[22](0.958), 0.623[23](0.479)
	298.15	1.06327	1.06311[17](0.015), 1.06326[18](0.001), 1.063337[19](−0.006), 1.06295[20](0.030), 1.063247[21](0.002), 1.0628[22](0.044), 1.06333[23](−0.006),1.0622[24](0.101),	0.587	0.579[17](1.363), 0.5771[20](1.687), 0.585[21](0.341), 0.58[22](1.193), 0.584[23](0.511), 0.534[24](9.029)
	303.15	1.05665	1.0568[14](−0.0142), 1.05635[17](0.028), 1.05623[18](0.040), 1.056719[19](−0.007), 1.05635[20](0.028), 1.056632[21](0.002), 1.0562[22](0.043), 1.0566[24](−0.005)	0.552	0.548[17](0.725), 0.5403[20](2.120), 0.550[21](0.362), 0.50[22](9.420), 0.549[23](0.543), 0.503[24](8.877)
	308.15	1.05000	1.0502[14](−0.0190), 1.04985[18](0.014), 1.050071[19](−0.007), 1.049983[21](0.002), 1.0498[22](0.019), 1.05007[23](−0.007), 1.0500[24](0.000)	0.520	0.519[21](0.192), 0.47[22](9.615), 0.517[23] (0.577), 0.474[24](8.846)
	313.15	1.04329	1.0435[14](−0.0201), 1.04310[17](0.018), 1.04304[18](0.024), 1.043388[19](−0.009), 1.04301[20](0.027), 1.043307[21](−0.002), 1.0425[22](0.076), 1.04339[23](−0.010)	0.498	0.486[17](2.410), 0.4807[20](3.474), 0.491[21](1.406), 0.45[22](9.639), 0.488[23](2.008)
	318.15	1.03657	1.0368[14](−0.0222), 1.03651[18](0.006), 1.036687[19](−0.011), 1.036590[21](−0.002), 1.0356[22](0.094)	0.472	0.465[21](1.483), 0.43[22](8.898)
	323.15	1.02983	1.0300[14](−0.0165), 1.029939[19](−0.011), 1.02959[20](0.023), 1.029843[21](−0.001), 1.0284[22](0.139)	0.450	0.4270[20](5.111), 0.442[21](1.778), 0.38[22](15.556)
	328.15	1.02303	1.023246[19](−0.021), 1.023050[21](−0.002)	0.429	0.420[21](2.098)
DEC	293.15	0.97491	0.97480[23](0.011), 0.97483[25](0.008), 0.97500[26](−0.009)	0.812	0.809[23](0.369), 0.803[25](1.108), 0.800[26](1.478), 0.795[27](2.094)
	298.15	0.96930	0.96921[23](0.009), 0.96915[25](0.015), 0.96897[26](0.034), 0.9691[27](0.021)	0.757	0.750[23](0.925), 0.757[25](0.000), 0.746[26](1.453), 0.749[27](1.057)
	303.15	0.96366	0.96358[23](0.008), 0.96339[25](0.028), 0.96239[26](0.132)	0.708	0.702[23](0.847), 0.700[25](1.130), 0.695[26](1.836), 0.699[27](1.271)
	308.15	0.95800	0.95793[23](0.007)	0.664	0.658[23](0.904)
	313.15	0.95232	0.95226[23](0.006), 0.95210[25](0.023), 0.95232[26](0.000)	0.625	0.618[23](1.120), 0.616[25](1.440), 0.610[26](2.400), 0.623[27](0.320)
	323.15	0.94086	0.93978[26](0.115)	0.556	0.541[26](2.610)

[a] $\delta\% = 100(Y_{exp} - Y_{lit.}/Y_{exp})$, the deviations, δ, of the experiment with literature values of density and viscosity are listed behind the literature values. Standard uncertainties: $u(T)$ = 0.05 K, $u(p)$ = 5 kPa, $u_r(\rho)$ = 0.00120, $u_r(\eta)$ = 0.01. [C_2mim][NTf_2], 1-ethyl-3-dimethylimidazolium bis[(trifluoromethyl)sulfonyl]imide; [C_4mim][NTf_2], 1-butyl-3-dimethylimidazolium bis[(trifluoromethyl)sulfonyl]-imide; DMC, dimethyl carbonate; DEC, diethyl carbonate.

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Table 3. Experimental Values of Density, ρ/g·cm^{-3}, of ILs + Solvents at Different Mole Fractions from 288.15 K to 328.15 K at Atmospheric Pressure (p = 101.3 kPa)[a]

x_{IL}	T/K = 288.15	T/K = 293.15	T/K = 298.15	T/K = 303.15	T/K = 308.15	T/K = 313.15	T/K = 318.15	T/K = 323.15	T/K = 328.15
					[C_2mim][NTf_2] + DMC				
0.0000	1.07639	1.06986	1.06327	1.05665	1.05000	1.04329	1.03657	1.02983	1.02303
0.0989	1.19915	1.19310	1.18703	1.18095	1.17486	1.16875	1.16263	1.15650	1.15034
0.1983	1.28285	1.27711	1.27135	1.26559	1.25983	1.25407	1.24830	1.24254	1.23676
0.2973	1.34257	1.33702	1.33147	1.32591	1.32035	1.31481	1.30926	1.30372	1.29819
0.3958	1.38769	1.38228	1.37685	1.37144	1.36603	1.36063	1.35523	1.34985	1.34447
0.4960	1.42397	1.41862	1.41328	1.40796	1.40266	1.39736	1.39208	1.38681	1.38155
0.5954	1.45255	1.44728	1.44202	1.43678	1.43155	1.42634	1.42114	1.41592	1.41075
0.6967	1.47687	1.47162	1.46642	1.46124	1.45607	1.45093	1.44579	1.44067	1.43557
0.7982	1.49752	1.49237	1.48723	1.48209	1.47698	1.47188	1.46680	1.46174	1.45669
0.9004	1.51450	1.50939	1.50428	1.49919	1.49412	1.48906	1.48402	1.47900	1.47400
1.0000	1.52855	1.52347	1.51839	1.51334	1.50830	1.50328	1.49827	1.49328	1.48831
					[C_2mim][NTf_2] + DEC				
0.0000	0.98049	0.97491	0.96930	0.96366	0.95800	0.95232	0.94660	0.94086	0.93508
0.0991	1.09045	1.08501	1.07954	1.07407	1.06856	1.06305	1.05753	1.05198	1.04642
0.1982	1.17930	1.17395	1.16860	1.16323	1.15786	1.15248	1.14711	1.14172	1.13632
0.3024	1.25464	1.24939	1.24411	1.23883	1.23356	1.22828	1.22301	1.21774	1.21246
0.3979	1.31221	1.30699	1.30178	1.29656	1.29136	1.28615	1.28095	1.27576	1.27058
0.4982	1.36222	1.35704	1.35187	1.34670	1.34155	1.33641	1.33127	1.32613	1.32101
0.5963	1.40491	1.39978	1.39464	1.38951	1.38440	1.37930	1.37420	1.36911	1.36403
0.6992	1.44262	1.43750	1.43238	1.42728	1.42219	1.41712	1.41206	1.40701	1.40198
0.7987	1.47505	1.46995	1.46486	1.45977	1.45471	1.44966	1.44462	1.43960	1.43459
0.9000	1.50375	1.49866	1.49358	1.48851	1.48346	1.47842	1.47341	1.46841	1.46342
1.0000	1.52855	1.52347	1.51839	1.51334	1.50830	1.50328	1.49827	1.49328	1.48831
					[C_4mim][NTf_2] + DMC				
0.0000	1.07639	1.06986	1.06327	1.05665	1.05000	1.04329	1.03657	1.02983	1.02303
0.0898	1.17803	1.17207	1.16607	1.16005	1.15402	1.14798	1.14193	1.13585	1.12976
0.1882	1.25071	1.24510	1.23948	1.23385	1.22822	1.22258	1.21694	1.21130	1.20565
0.2749	1.29536	1.28995	1.28455	1.27914	1.27373	1.26832	1.26292	1.25751	1.25210
0.3754	1.33426	1.32902	1.32378	1.31855	1.31332	1.30809	1.30288	1.29767	1.29246
0.4720	1.36215	1.35702	1.35190	1.34678	1.34167	1.33658	1.33149	1.32640	1.32133
0.5745	1.38580	1.38077	1.37574	1.37071	1.36570	1.36070	1.35571	1.35073	1.34575
0.6730	1.40416	1.39914	1.39418	1.38923	1.38429	1.37937	1.37445	1.36955	1.36466
0.7799	1.41988	1.41497	1.41007	1.40518	1.40031	1.39544	1.39059	1.38574	1.38090
0.8948	1.43465	1.42979	1.42493	1.42008	1.41526	1.41045	1.40567	1.40088	1.39612
1.0000	1.44566	1.44084	1.43601	1.43121	1.42643	1.42166	1.41691	1.41217	1.40745
					[C_4mim][NTf_2] + DEC				
0.0000	0.98049	0.97491	0.96930	0.96366	0.95800	0.95232	0.94660	0.94086	0.93508
0.1025	1.08684	1.08145	1.07605	1.07065	1.06523	1.05980	1.05435	1.04888	1.04340
0.2198	1.17790	1.17270	1.16748	1.16226	1.15703	1.15180	1.14657	1.14133	1.13607
0.3178	1.23657	1.23147	1.22635	1.22123	1.21612	1.21099	1.20588	1.20077	1.19565
0.4211	1.28586	1.28083	1.27580	1.27077	1.26574	1.26071	1.25569	1.25068	1.24568
0.5268	1.32709	1.32213	1.31716	1.31220	1.30725	1.30230	1.29736	1.29244	1.28750
0.6220	1.35833	1.35342	1.34849	1.34358	1.33867	1.33378	1.32890	1.32402	1.31916
0.7193	1.38568	1.38079	1.37590	1.37104	1.36617	1.36132	1.35648	1.35166	1.34684
0.8139	1.40825	1.40338	1.39852	1.39368	1.38884	1.38401	1.37919	1.37439	1.36958
0.9088	1.42849	1.42365	1.41881	1.41397	1.40918	1.40439	1.39960	1.39485	1.39010
1.0000	1.44566	1.44084	1.43601	1.43121	1.42643	1.42166	1.41691	1.41217	1.40745

[a]Standard uncertainties are $u_r(\rho)$ = 0.00120, $u(T)$ = 0.05 K, $u(p)$ = 5 kPa, $u(x)$ = 0.0002.

confidence level, the uncertainty is better than 0.01 for viscosity at 0.95 confidence level. For density, the uncertainty is obtained from literature.[32] For viscosity, the uncertainty is mainly from the machine (the values are 1.59 mm and 1.8 mm capillary are 0.007 and 0.005, respectively, for a steel ball). The water content should also be considered for the uncertainty determination. However, the contents are less than 400 × 10^{-6} mass fraction before measurement. According to Jacquemin[33] et al. and our previous work,[34] the water content change can be ignored before and after measurement of density and viscosity by the Lovis 2000 ME Anton Paar Rotational Stabinger viscometer. So, the values can be ignored during measurements.

The density and viscosity of the pure compounds [C_2mim][NTf_2], [C_4mim][NTf_2], DMC, and DEC from this study and available from the literature are shown in Table 2.[4−15,17−27] The measured values of the binary mixtures are listed in Tables 3 and 4. All of the values of the density and viscosity were measured at atmospheric pressure (p = 101.3 kPa).

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Table 4. Experimental Values of Dynamic Viscosity, η/mPa·s, of ILs + Solvents at Different Mole Fractions from 288.15 K to 328.15 K at Atmospheric Pressure (p = 101.3 kPa)[a]

x_{IL}\T/K	T/K = 288.15	T/K = 293.15	T/K = 298.15	T/K = 303.15	T/K = 308.15	T/K = 313.15	T/K = 318.15	T/K = 323.15	T/K = 328.15
					[C_2mim][NTf_2] + DMC				
0.0000	0.669	0.626	0.587	0.552	0.520	0.498	0.472	0.450	0.429
0.0989	1.579	1.451	1.348	1.250	1.162	1.086	1.016	0.954	0.898
0.1983	2.913	2.637	2.400	2.195	2.019	1.865	1.729	1.608	1.501
0.2973	4.862	4.332	3.884	3.507	3.177	2.904	2.661	2.452	2.268
0.3958	7.561	6.635	5.878	5.241	4.706	4.247	3.831	3.512	3.224
0.4960	11.21	9.698	8.484	7.472	6.640	5.946	5.358	4.857	4.421
0.5954	16.89	14.58	12.53	10.80	9.382	8.189	7.249	6.332	5.588
0.6967	22.56	19.02	16.66	14.52	12.62	10.96	9.578	8.381	7.456
0.7982	30.07	25.12	21.25	18.37	16.22	14.22	12.43	10.90	9.610
0.9004	38.55	31.87	26.71	22.66	19.42	17.21	15.16	13.29	11.68
1.0000	49.41	40.36	33.52	28.35	24.15	20.76	17.90	15.93	14.26
					[C_2mim][NTf_2] + DEC				
0.0000	0.871	0.812	0.757	0.708	0.664	0.625	0.589	0.556	0.525
0.0991	1.938	1.769	1.623	1.495	1.389	1.290	1.204	1.121	1.049
0.1982	3.368	3.032	2.746	2.498	2.288	2.102	1.939	1.794	1.667
0.3024	5.628	4.987	4.452	3.992	3.607	3.278	2.993	2.746	2.528
0.3979	8.533	7.450	6.552	5.819	5.196	4.672	4.223	3.839	3.509
0.4982	12.38	10.66	9.269	8.127	7.192	6.410	5.749	5.184	4.704
0.5963	17.23	14.65	12.59	10.93	9.572	8.445	7.525	6.739	6.075
0.6992	24.14	20.32	17.65	15.41	13.37	11.60	10.14	8.895	7.875
0.7987	31.13	25.96	21.91	18.79	16.59	14.56	12.72	11.15	9.813
0.9000	39.07	32.29	27.04	22.93	19.65	17.36	15.35	13.53	11.93
1.0000	49.41	40.36	33.52	28.35	24.15	20.76	17.90	15.93	14.26
					[C_4mim][NTf_2] + DMC				
0.0000	0.669	0.626	0.587	0.552	0.520	0.498	0.472	0.450	0.429
0.0898	1.605	1.473	1.365	1.263	1.173	1.095	1.022	0.958	0.899
0.1882	3.207	2.886	2.611	2.376	2.176	2.002	1.849	1.719	1.588
0.2749	5.396	4.762	4.235	3.797	3.428	3.113	2.837	2.600	2.394
0.3754	9.186	7.930	6.922	6.100	5.404	4.822	4.332	3.943	3.590
0.4720	14.30	12.12	10.39	8.998	7.885	6.959	6.189	5.540	4.994
0.5745	22.53	18.87	16.30	14.00	11.98	10.30	8.913	7.757	6.772
0.6730	32.06	26.30	21.86	18.59	16.17	14.01	12.10	10.53	9.191
0.7799	44.52	35.81	29.38	24.46	20.58	17.54	15.20	13.21	11.54
0.8948	61.53	48.74	39.46	32.86	27.73	23.58	19.93	17.01	14.93
1.0000	78.02	61.32	48.93	39.84	32.91	27.56	23.33	19.97	17.41
					[C_4mim][NTf_2] + DEC				
0.0000	0.871	0.812	0.757	0.708	0.664	0.625	0.589	0.556	0.525
0.1025	2.120	1.928	1.764	1.618	1.492	1.385	1.286	1.198	1.117
0.2198	4.415	3.925	3.517	3.168	2.873	2.614	2.391	2.194	2.025
0.3178	7.583	6.526	5.739	5.085	4.539	3.977	3.591	3.259	2.978
0.4211	13.26	11.18	9.492	8.179	7.219	6.414	5.815	5.216	4.710
0.5268	20.12	17.25	14.77	12.60	10.76	9.287	8.069	7.105	6.138
0.6220	28.36	23.42	19.58	17.03	14.69	12.63	10.92	9.492	8.288
0.7193	38.93	31.68	26.16	21.88	18.70	16.35	14.18	12.28	10.66
0.8139	50.88	40.79	33.25	27.57	23.20	19.64	16.89	14.60	12.69
0.9088	64.94	51.44	41.43	34.06	28.32	23.69	20.05	17.26	15.00
1.0000	78.02	61.32	48.93	39.84	32.91	27.56	23.33	19.97	17.41

[a]Standard uncertainties are $u(T)$ = 0.05 K, $u(p)$ = 5 kPa, $u(x)$ = 0.0002, $u_r(\eta)$ = 0.01.

3. RESULTS AND DISCUSSION

The density and viscosity of the ILs [C_2mim][NTf_2], [C_4mim][NTf_2], DMC, and DEC are listed in Table 2. The deviations of the experiment and literature values are calculated and listed in Table 2. From Table 2, for [C_2mim][NTf_2], the absolute density deviations are less than 0.03% in the literature[4−7] except for those in ref 8 (but the absolute deviations are still less than 0.14%); the absolute viscosity deviations are less than 6.6% in the literature.[4,5,9] For [C_4mim][NTf_2], the absolute density deviations are higher than 0.04% and less than 0.18% in the literature;[10−15] the absolute viscosity deviations are higher than 1.2% and less than 4.1% in the literature.[10,13] For DMC, the absolute density deviations are less than 0.10% in the literature[17−24] except one point, 0.139%;[22] the absolute viscosity deviations are less than 10% in the literature[17,20−24] except for one point, 15.556%.[22] For DEC, the absolute density deviations are less than 0.03% in the literature[23,25−27] except for two points, 0.132% and 0.115%;[26]

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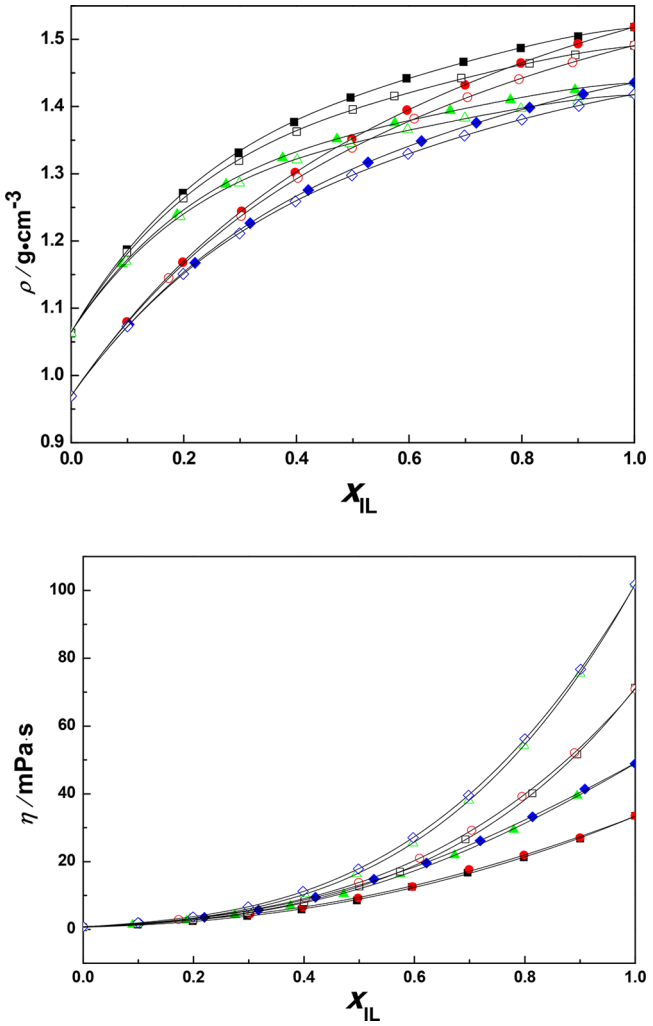

Figure 2. Plot of density, ρ/g·cm^{-3}, and viscosity, η/mPa·s, vs molar fraction, x_{IL}, of binary mixtures at 298.15 K at p = 101.3 kPa: ■, [C_2mim][NTf_2] + DMC; ●, [C_2mim][NTf_2] + DEC; ▲, [C_4mim][NTf_2] + DMC; ♦, [C_4mim][NTf_2] + DEC; □, [C_2mmim][NTf_2] + DMC; ○, [C_2mmim][NTf_2] + DEC; △, [C_4mmim][NTf_2] + DMC; ◇, [C_4mmim][NTf_2] + DEC.

the absolute viscosity deviations are less than 2.4% in the literature.[23,25−27]

In Figure 2, the density and viscosity of the four binary mixtures were plotted over the entire mole fraction at 298.15 K and compared with our previous work (research on [C_2mmim][NTf_2] + DMC, [C_2mmim][NTf_2] + DEC, [C_4mmim][NTf_2] + DMC, and [C_4mmim][NTf_2] + DEC systems).[29] From Figure 2, for the density, for the same ILs, the values of DMC systems are higher than those of the DEC systems; for the same solvents, the values change to smaller after the introduction of a methyl group on the 2-position or methylene group on the side chain of the ILs cation. For the viscosity, the tendency is in contrast. For the same ILs, the values of the DMC systems are lower than those of the DEC systems; for the same solvents, the values change to larger after the introduction of a methyl group on the 2-position or methylene group on the side chain of the ILs cation.

From Tables 3 and 4, the density and viscosity decrease with the temperature increase and concentration decrease of the ILs. According to the viscosity, the binary mixtures are viscous liquids and non-Newtonian fluids. On this basis, all of the shear rates have been listed in Table S1 in the Supporting Information.

3.1. Excess Molar Volume. According to the following equation, the excess molar volumes, V^E can be determined:

$$V^E = \frac{x_1M_1 + x_2M_2}{\rho} - \frac{x_1M_1}{\rho_1} - \frac{x_2M_2}{\rho_2} \quad (1)$$

herein, V^E is the excess molar volume; ρ is the density of the binary mixture; x_1, M_1, ρ_1, x_2, M_2, and ρ_2 are the mole fraction, molar mass, and density of components 1 and 2, respectively. The calculated values of the four binary mixtures are listed in Table 5.

The V^E, against x_i plots of the four binary mixtures are shown in Figure 3 from 288.15 to 328.15 K. From Figure 3, it can be seen that the values are negative over the whole composition range and all of the curves are asymmetrical. According to the references[26,35] and our previous research results,[29] we conclude that there are three main factors that may affect the V^E values, namely the specific factors between molecules (such as hydrogen bonds, charge-transfer complexes, breaking of hydrogen bonds), the physical intermolecular forces (generally including electrostatic forces between charged particles and between permanent dipoles, induction forces between a permanent dipole and an induced dipole, and forces of attraction and repulsion between nonpolar molecules) and the structural characteristics of the component. In the studied mixtures, hydrogen bonding and ion-dipole forces are expected to be the dominant attraction, however, they usually weaken with the increase of temperature, which will lead to the opposite trend of V^E. Besides, the negative excess volumes observed may be related to a relatively more packed arrangement of the molecules and ions in the mixture at high temperature.

In Figure 3, the absolute values, $|V^E|$, increase with the temperature increasing. This points to the existence of stronger interactions of ILs with solvents than before the temperature increase. The same result has been found in the literature.[35] To compare the values with [C_2mmim][NTf_2] and [C_4mmim][NTf_2],[29] the V^E, against x_i plots of the eight binary mixtures are shown in Figure 4 at 298.15 K. From Figure 4, the order of the absolute values, $|V^E|$, is [C_4mmim][NTf_2] + DMC < [C_2mmim][NTf_2] + DMC < [C_4mim][NTf_2] + DMC < [C_2mim][NTf_2] + DMC < [C_4mmim][NTf_2] + DEC < [C_2mmim][NTf_2] + DEC ≈ [C_4mim][NTf_2] + DEC < [C_2mim][NTf_2] + DEC in the whole mole fraction. For DEC and DMC, the absolute values of the mixtures containing DEC are higher than those of the mixtures containing DMC. This may occur because with the increases of the chain carbonate, the polarity lessens, and the intermolecular attraction decreases; dilution process is less endothermic, and $|V^E|$ values that are higher are those of the mixtures containing DEC. The same result has also been found in ref 24. After the introduction of the methylene group on the side chain of ILs cation 1-position, the absolute values of V^E are lower than those without the methylene group for the same solvent. This may be because the introduction of the methylene group leads to the increase of steric effect, which weakens the binding ability of ILs with solvents (DEC or DMC) and makes the interaction worse. The same result has been found after the introduction of the methyl group on the ILs cation 2-position for the same solvent. The absolute values are lower than those without methyl introduction. The steric effect can also be used to explain why the absolute values decrease after the introduction of the methyl group on the ILs

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Table 5. Excess Molar Volume, V^E/g·cm^{-3}, and Viscosity Deviations, $\Delta\eta$/mPa·s for the Binary Mixture of ILs + Solvents at Different Mole Fractions from 288.15 K to 328.15 K at Atmospheric Pressure (p = 101.3 kPa)[a]

	V^E/cm^3·mol^{-1}									$\Delta\eta$/mPa·s								
T/K:	288.15	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	288.15	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15
x_{IL}	[C_2mim][NTf_2] + DMC																	
0.0989	−0.7615	−0.7999	−0.8415	−0.8848	−0.9304	−0.9799	−1.0304	−1.0828	−1.1384	−3.912	−3.106	−2.497	−2.052	−1.696	−1.417	−1.180	−1.028	−0.8993
0.1983	−1.0768	−1.1307	−1.1883	−1.2478	−1.3108	−1.3785	−1.4479	−1.5204	−1.5964	−7.419	−5.866	−4.716	−3.868	−3.186	−2.650	−2.198	−1.911	−1.670
0.2973	−1.1146	−1.1723	−1.2342	−1.2969	−1.3636	−1.4348	−1.5081	−1.5847	−1.6655	−10.30	−8.107	−6.494	−5.309	−4.368	−3.618	−2.992	−2.601	−2.273
0.3958	−1.0559	−1.1115	−1.1706	−1.2311	−1.2951	−1.3636	−1.4336	−1.5069	−1.5837	−12.40	−9.718	−7.744	−6.314	−5.167	−4.271	−3.539	−3.066	−2.680
0.4960	−0.9691	−1.0157	−1.0675	−1.1200	−1.1782	−1.2388	−1.3018	−1.3680	−1.4373	−13.63	−10.64	−8.438	−6.868	−5.601	−4.602	−3.758	−3.272	−2.868
0.5954	−0.7911	−0.8302	−0.8744	−0.9196	−0.9675	−1.0190	−1.0720	−1.1241	−1.1826	−12.80	−9.705	−7.666	−6.304	−5.208	−4.374	−3.600	−3.336	−3.077
0.6967	−0.6415	−0.6675	−0.7017	−0.7371	−0.7743	−0.8149	−0.8562	−0.8999	−0.9453	−12.06	−9.287	−6.870	−5.398	−4.362	−3.654	−3.035	−2.854	−2.609
0.7982	−0.5160	−0.5379	−0.5651	−0.5877	−0.6155	−0.6428	−0.6720	−0.7033	−0.7347	−9.502	−7.220	−5.623	−4.369	−3.160	−2.450	−1.952	−1.906	−1.857
0.9004	−0.2727	−0.2845	−0.2986	−0.3105	−0.3239	−0.3370	−0.3517	−0.3684	−0.3851	−6.004	−4.532	−3.529	−2.921	−2.376	−1.531	−1.004	−1.099	−1.202
	[C_2mim][NTf_2] + DEC																	
0.0991	−0.7540	−0.7917	−0.8319	−0.8730	−0.9151	−0.9601	−1.0072	−1.0565	−1.1091	−3.743	−2.962	−2.380	−1.952	−1.602	−1.330	−1.100	−0.9584	−0.8370
0.1982	−1.2574	−1.3142	−1.3747	−1.4363	−1.5014	−1.5695	−1.6415	−1.7168	−1.7954	−7.124	−5.619	−4.505	−3.689	−3.031	−2.514	−2.081	−1.809	−1.580
0.3024	−1.4654	−1.5316	−1.6002	−1.6694	−1.7429	−1.8203	−1.9009	−1.9862	−2.0747	−9.920	−7.784	−6.212	−5.075	−4.159	−3.436	−2.831	−2.459	−2.150
0.3979	−1.5468	−1.6107	−1.6807	−1.7512	−1.8256	−1.9034	−1.9846	−2.0698	−2.1607	−11.65	−9.097	−7.240	−5.887	−4.812	−3.964	−3.254	−2.834	−2.481
0.4982	−1.3703	−1.4289	−1.4925	−1.5571	−1.6263	−1.6979	−1.7727	−1.8511	−1.9321	−12.67	−9.854	−7.810	−6.352	−5.172	−4.246	−3.464	−3.031	−2.664
0.5963	−1.2623	−1.3155	−1.3722	−1.4291	−1.4894	−1.5529	−1.6179	−1.6862	−1.7574	−12.58	−9.743	−7.702	−6.260	−5.096	−4.186	−3.386	−2.984	−2.640
0.6992	−0.9493	−0.9899	−1.0344	−1.0783	−1.1257	−1.1742	−1.2261	−1.2807	−1.3368	−10.67	−8.143	−6.014	−4.624	−3.715	−3.103	−2.552	−2.410	−2.253
0.7987	−0.7175	−0.7466	−0.7789	−0.8098	−0.8432	−0.8782	−0.9144	−0.9535	−0.9928	−8.508	−6.438	−5.014	−3.995	−2.832	−2.146	−1.695	−1.685	−1.682
0.9000	−0.3916	−0.4073	−0.4249	−0.4404	−0.4583	−0.4756	−0.4949	−0.5158	−0.5365	−5.486	−4.115	−3.204	−2.656	−2.152	−1.387	−0.8190	−0.8627	−0.9566
	[C_4mim][NTf_2] + DMC																	
0.0898	−0.6503	−0.6860	−0.7229	−0.7618	−0.8026	−0.8475	−0.8931	−0.9401	−0.9901	−6.013	−4.606	−3.565	−2.819	−2.257	−1.834	−1.504	−1.246	−1.055
0.1882	−0.9569	−1.0069	−1.0601	−1.1159	−1.1739	−1.2361	−1.3006	−1.3680	−1.4389	−12.02	−9.164	−7.076	−5.571	−4.441	−3.589	−2.926	−2.406	−2.038
0.2749	−1.0031	−1.0567	−1.1143	−1.1740	−1.2359	−1.3022	−1.3706	−1.4417	−1.5168	−16.54	−12.55	−9.642	−7.556	−5.996	−4.825	−3.919	−3.216	−2.703
0.3754	−1.0092	−1.0622	−1.1187	−1.1775	−1.2378	−1.3031	−1.3697	−1.4399	−1.5128	−20.52	−15.48	−11.81	−9.200	−7.275	−5.835	−4.721	−3.834	−3.213
0.4720	−0.8729	−0.9209	−0.9718	−1.0251	−1.0793	−1.1384	−1.1987	−1.2614	−1.3273	−22.88	−17.16	−13.02	−10.10	−7.924	−6.313	−5.073	−4.124	−3.451
0.5745	−0.7514	−0.7930	−0.8366	−0.8824	−0.9287	−0.9787	−1.0296	−1.0828	−1.1372	−22.58	−16.62	−12.06	−9.127	−7.144	−5.745	−4.691	−3.907	−3.412
0.6730	−0.6170	−0.6431	−0.6787	−0.7150	−0.7526	−0.7926	−0.8333	−0.8758	−0.9208	−20.66	−15.17	−11.26	−8.404	−6.150	−4.701	−3.753	−3.061	−2.666
0.7799	−0.3482	−0.3704	−0.3953	−0.4221	−0.4472	−0.4736	−0.5003	−0.5277	−0.5545	−16.47	−12.15	−8.909	−6.732	−5.201	−4.063	−3.099	−2.463	−2.132
0.8948	−0.2059	−0.2162	−0.2282	−0.2407	−0.2529	−0.2671	−0.2819	−0.2957	−0.3114	−8.354	−6.196	−4.385	−2.848	−1.773	−1.134	−0.9957	−0.9068	−0.6939
	[C_4mim][NTf_2] + DEC																	
0.1025	−0.7644	−0.7981	−0.8373	−0.8789	−0.9214	−0.9664	−1.0134	−1.0628	−1.1148	−6.659	−5.086	−3.931	−3.101	−2.478	−2.001	−1.634	−1.348	−1.139
0.2198	−1.2576	−1.3152	−1.3752	−1.4368	−1.5011	−1.5689	−1.6404	−1.7146	−1.7919	−13.42	−10.19	−7.830	−6.142	−4.879	−3.932	−3.197	−2.629	−2.212
0.3178	−1.4316	−1.4936	−1.5581	−1.6257	−1.6947	−1.7660	−1.8425	−1.9234	−2.0072	−17.81	−13.52	−10.33	−8.060	−6.374	−5.208	−4.226	−3.467	−2.913
0.4211	−1.3806	−1.4414	−1.5043	−1.5697	−1.6373	−1.7056	−1.7795	−1.8571	−1.9394	−20.10	−15.12	−11.55	−9.009	−7.025	−5.555	−4.351	−3.516	−2.926
0.5268	−1.2367	−1.2914	−1.3481	−1.4080	−1.4690	−1.5325	−1.5988	−1.6690	−1.7393	−21.39	−15.44	−11.36	−8.724	−6.891	−5.528	−4.500	−3.678	−3.282
0.6220	−1.0680	−1.1157	−1.1649	−1.2159	−1.2672	−1.3220	−1.3783	−1.4374	−1.5001	−20.50	−15.03	−11.14	−8.022	−6.032	−4.750	−3.813	−3.139	−2.739
0.7193	−0.8596	−0.8959	−0.9346	−0.9767	−1.0167	−1.0599	−1.1042	−1.1509	−1.1997	−17.43	−12.66	−9.249	−6.973	−5.158	−3.650	−2.768	−2.244	−2.006
0.8139	−0.5375	−0.5617	−0.5879	−0.6157	−0.6415	−0.6687	−0.6956	−0.7247	−0.7524	−12.78	−9.271	−6.716	−4.989	−3.710	−2.908	−2.209	−1.758	−1.578
0.9088	−0.2784	−0.2908	−0.3036	−0.3160	−0.3320	−0.3473	−0.3599	−0.3768	−0.3936	−6.042	−4.360	−3.105	−2.210	−1.648	−1.413	−1.205	−0.9389	−0.8696

[a]Standard uncertainties are $u(T)$ = 0.05 K, $u(p)$ = 5 kPa, $u(x)$ = 0.0002, $u_r(V^E)$ = 0.0030, $u_r(\Delta\eta)$ = 0.01.

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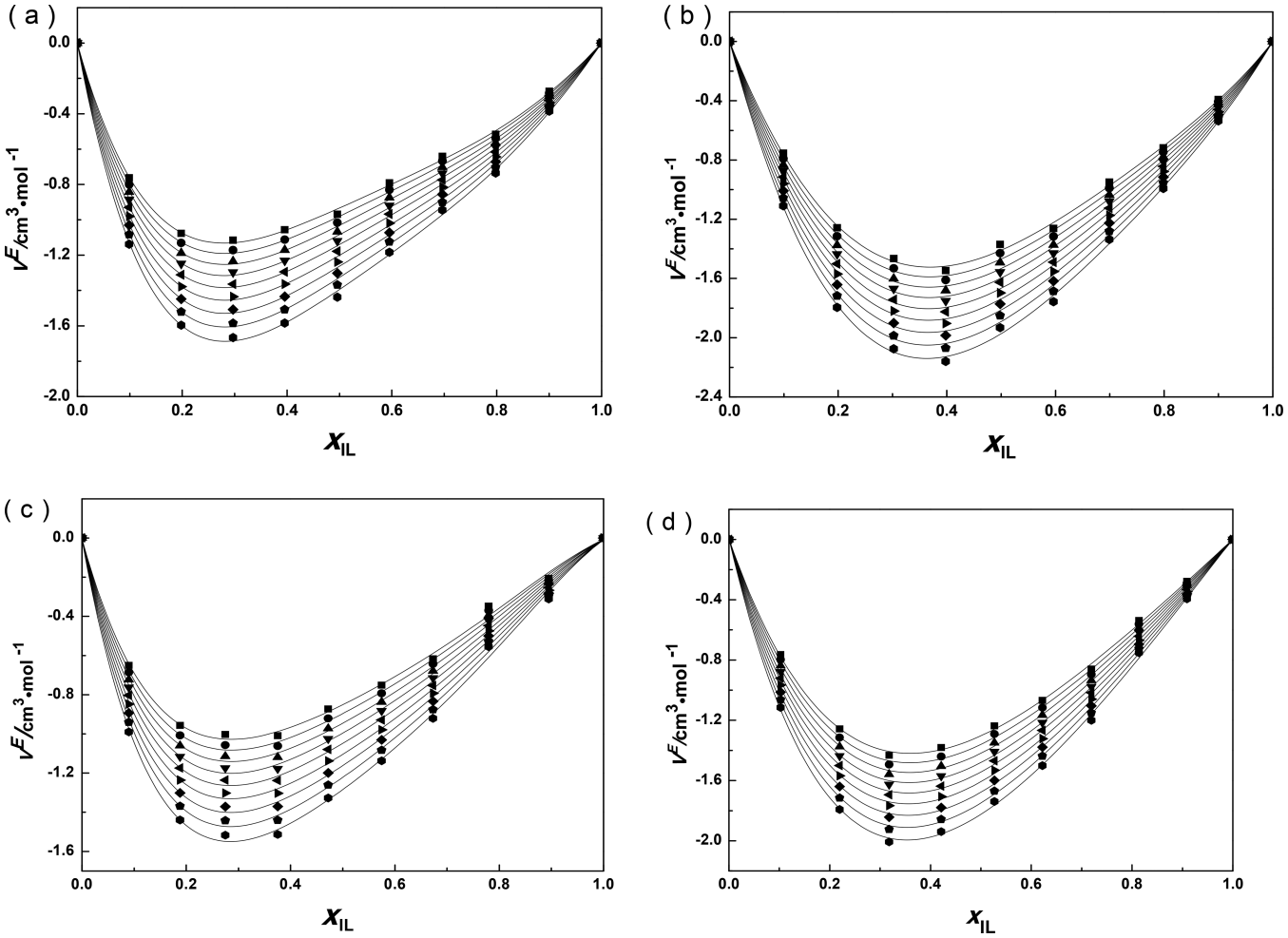

Figure 3. Plot of excess molar volumes, V^E/cm^3·mol^{-1}, vs molar fraction, x_{IL}, of binary mixtures from 288.15 K to 328.15 K at p = 101.3 kPa. (a) [C_2mim][NTf_2] + DMC; (b) [C_2mim][NTf_2] + DEC; (c) [C_4mim][NTf_2] + DMC; (d) [C_4mim][NTf_2] + DEC: ■, 288.15 K; ●, 293.15 K; ▲, 298.15 K; ▼, 303.15 K; ◀, 308.15 K; ▶, 313.15 K; ♦, 318.15 K; ⬟, 323.15 K; ⬢, 328.15 K.

cation. The minimum excess molar volumes can be shown at the mole fractions $x \approx 0.30$ for DMC type binary mixtures, $x \approx 0.38$ for DEC type binary mixtures, respectively. From Figure 4, for DMC type binary mixtures, the mole fractions are slightly higher than the mole fractions $x \approx 0.25$ of the [C_2mmim][NTf_2] or [C_4mmim][NTf_2] system;[29] for DEC type binary mixtures, the mole fractions are slightly higher than the mole fraction, $x \approx 0.35$ of the [C_2mmim][NTf_2] or [C_4mmim][NTf_2] system.[29] This should be because the introduction of the methyl group leads to the increase of steric effect, which weakens the binding ability of ionic liquids with solvents (DEC or DMC) and makes the interaction worse. Therefore, it can reach a stable state at a lower concentration, and the $|V^E|$ reaches the maximum easily.

3.2. Viscosity Deviation. According to the following equation, the viscosity deviation, $\Delta\eta$ can also be determined:

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

where $\Delta\eta$ is the viscosity deviation; x_1, η_1, x_2, and η_2 are the mole fraction and viscosity of components 1 and 2, respectively. The calculated values of the viscosity deviation are also listed in Table 5.

Like those of the excess molar volumes, the viscosity deviations, $\Delta\eta$, versus mole fraction, x_i, of the four binary mixtures are shown in Figure 5 from 288.15 to 328.15 K. In Figure 5, as with the excess molar volumes, the values are negative over the whole composition range and all of the curves are asymmetric. In Figure 5 the absolute values decrease with increasing temperature. To compare the values with those of our previous work,[29] the data $\Delta\eta$ of [C_2mmim][NTf_2] and [C_4mmim][NTf_2] against x_i plots of the eight binary mixtures are also shown in Figure 6 with this work at 298.15 K. Unlike those of the excess molar volumes, the minimum viscosity deviations can be seen at the mole fraction $x \approx 0.50$ for the four binary mixtures, which are slightly less than those of the four binary mixtures of [C_2mmim][NTf_2] or [C_4mmim]-[NTf_2].[29] From Figure 6, the order of the absolute values, $|\Delta\eta|$, is [C_2mim][NTf_2] + DEC < [C_2mim][NTf_2] + DMC < [C_4mim][NTf_2] + DEC < [C_4mim][NTf_2] + DMC < [C_2mmim][NTf_2] + DEC < [C_2mmim][NTf_2] + DMC < [C_4mmim][NTf_2] + DEC < [C_4mmim][NTf_2] + DMC in the whole mole fraction. From Figure 6, the absolute values are increased with the methyl group and methylene group introduction on the cation of the ILs. The absolute values increase from DEC to DMC for the same ionic liquid. The results are opposite with excess molar volumes. Similar to the excess molar volumes, the viscosity is also related to the molecular interaction between the components of mixtures as well as of the size and shape of molecules. According to the literature,[36] intermolecular interactions strongly influence the

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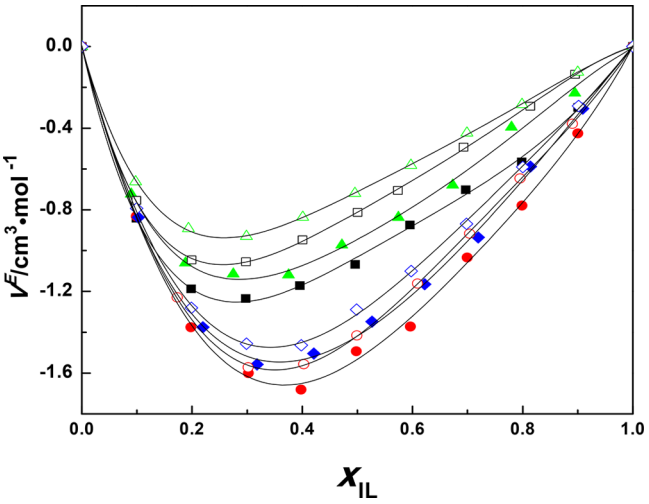

Figure 4. Plot of excess molar volumes, V^E/cm^3·mol^{-1}, vs molar fraction, x_{IL}, of binary mixtures at 298.15 K at p = 101.3 kPa. ■, [C_2mim][NTf_2] + DMC; ●, [C_2mim][NTf_2] + DEC; ▲, [C_4mim][NTf_2] + DMC; ◆, [C_4mim][NTf_2] + DEC; □, [C_2mmim][NTf_2] + DMC; ○, [C_2mmim][NTf_2] + DEC; △, [C_4mmim][NTf_2] + DMC; ◇, [C_4mmim][NTf_2] + DEC.

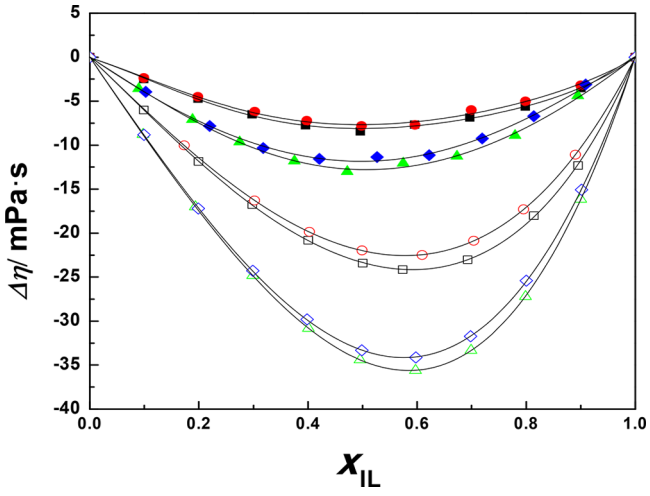

Figure 6. Plot of viscosity deviation, $\Delta\eta$/mPa·s, vs molar fraction, x_{IL}, of four binary mixtures at 298.15 K at p = 101.3 kPa. ■, [C_2mim][NTf_2] + DMC; ●, [C_2mim][NTf_2] + DEC; ▲, [C_4mim][NTf_2] + DMC; ♦, [C_4mim][NTf_2] + DEC; □, [C_2mmim]-[NTf_2] + DMC; ○, [C_2mmim][NTf_2] + DEC; △, [C_4mmim][NTf_2] + DMC; ◇, [C_4mmim][NTf_2] + DEC.

$\Delta\eta$ such as anion−cation interactions in the pure IL. The IL has high viscosity because the movement of molecules from one equilibrium position to the next is strongly disturbed by neighboring molecules. The anion−cation interactions in the

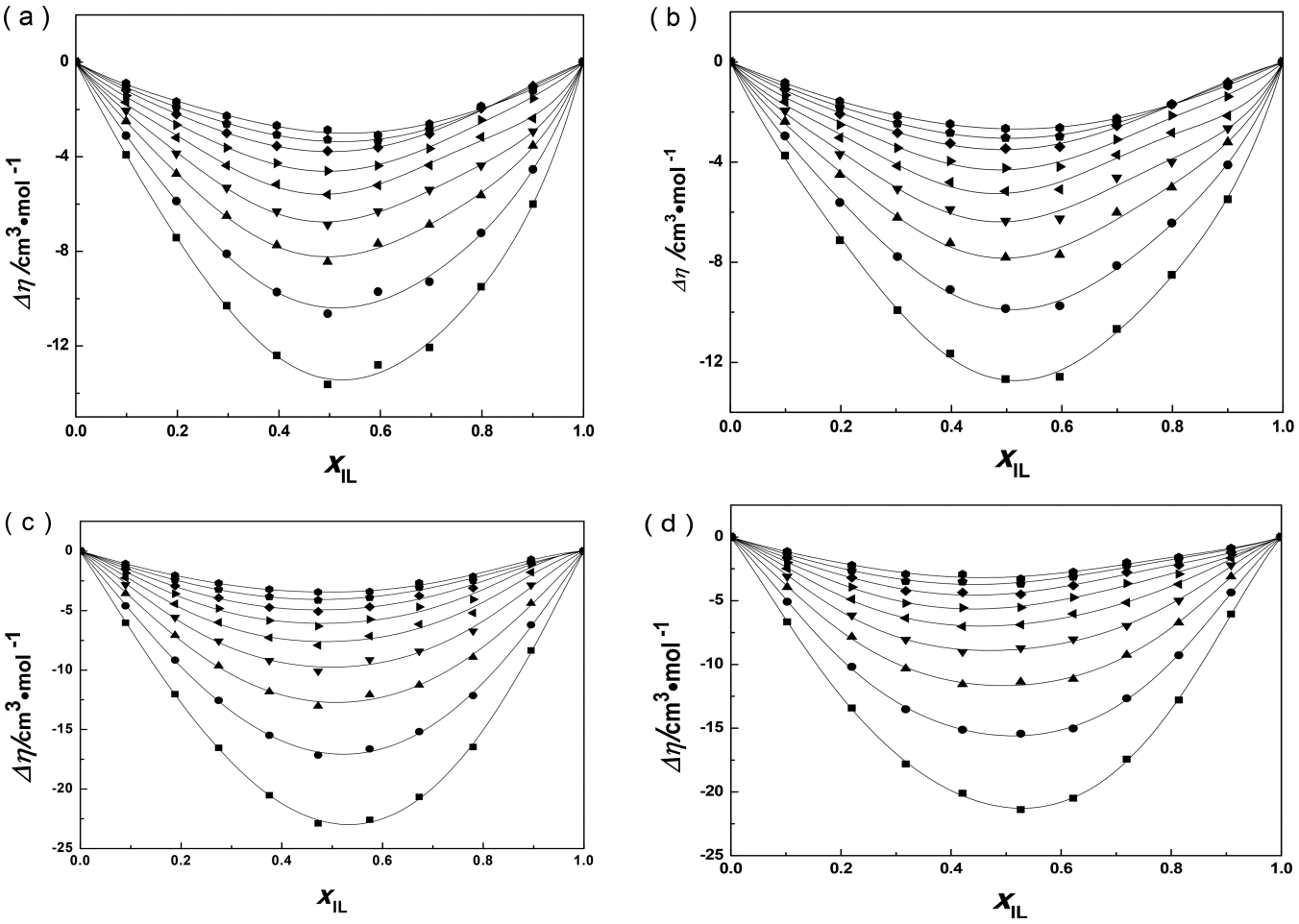

Figure 5. Plot of viscosity deviation, $\Delta\eta$/mPa·s, vs molar fraction, x_{IL}, of four binary mixtures from 288.15 K to 328.15 K at p = 101.3 kPa. (a) [C_2mim][NTf_2] + DMC; (b) [C_2mim][NTf_2] + DEC; (c) [C_4mim][NTf_2] + DMC; (d) [C_4mim][NTf_2] + DEC: ■, 288.15 K; ●, 293.15 K; ▲, 298.15 K; ▼, 303.15 K; ◀, 308.15 K; ▶, 313.15 K; ◆, 318.15 K; ⬟, 323.15 K; ⬢, 328.15.

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Table 6. Fitted Parameter Values by Equations 4, 5 for Excess Molar Volume, V^E/g·cm^{-3}, and Viscosity Deviations, $\Delta\eta$/mPa·s, of the Binary Mixture ILs + Solvents[a]

	V^E/cm^3·mol^{-1}									$\Delta\eta$/mPa·s								
T/K:	288.15	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	288.15	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15
									[C_2mim][NTf_2] + DMC									
A_0	−3.733	−3.919	−4.124	−4.335	−4.561	−4.802	−5.049	−5.305	−5.577	−53.16	−41.22	−32.45	−26.37	−21.62	−18.14	−15.05	−13.33	−11.79
A_1	2.639	2.819	2.970	3.122	3.282	3.447	3.618	3.811	4.003	−5.959	−2.746	0.5448	1.671	2.157	0.5042	−0.1271	−1.588	−2.145
A_2	−3.226	−3.364	−3.526	−3.665	−3.822	−3.984	−4.155	−4.351	−4.540	−1.104	−0.5998	−0.0209	0.5907	2.078	4.472	5.028	2.912	1.030
A_3	1.110	1.109	1.161	1.269	1.361	1.498	1.617	1.698	1.822	−13.21	−11.40	−12.59	−11.62	−8.787	−0.6330	2.880	2.826	1.365
$10^2\sigma$	1.800	1.860	1.900	1.870	1.950	1.960	2.000	2.080	2.110	21.74	22.06	16.59	16.51	19.93	10.69	4.880	7.240	9.400
									[C_2mim][NTf_2] + DEC									
A_0	−5.637	−5.874	−6.131	−6.389	−6.664	−6.952	−7.250	−7.563	−7.891	−50.12	−38.97	−30.68	−24.77	−20.20	−16.82	−13.87	−12.17	−10.67
A_1	3.189	3.325	3.464	3.606	3.758	3.916	4.084	4.262	4.454	−4.100	−2.049	0.6684	2.084	2.750	1.461	0.6864	−0.3274	−0.8336
A_2	−1.286	−1.379	−1.476	−1.560	−1.650	−1.743	−1.855	−1.978	−2.097	1.580	2.120	2.113	1.675	2.933	4.860	5.520	3.348	1.202
A_3	−0.9464	−0.9160	−0.8854	−0.8280	−0.7943	−0.7390	−0.6912	−0.6577	−0.6116	−10.16	−7.402	−8.770	−9.723	−7.752	−1.028	2.909	2.195	0.4807
$10^2\sigma$	2.830	2.860	2.910	2.950	2.960	3.010	3.020	3.020	3.080	23.72	19.40	24.31	27.13	26.10	15.93	7.240	4.060	2.580
									[C_4mim][NTf_2] + DMC									
A_0	−3.403	−3.582	−3.778	−3.983	−4.191	−4.415	−4.644	−4.884	−5.133	−91.70	−68.26	−51.14	−39.44	−30.86	−24.67	−19.88	−16.23	−13.86
A_1	2.631	2.783	2.915	3.052	3.196	3.356	3.526	3.702	3.894	−13.47	−6.621	−1.670	0.3459	1.861	2.042	2.380	2.045	0.5609
A_2	−2.095	−2.212	−2.341	−2.479	−2.622	−2.777	−2.941	−3.098	−3.272	14.48	9.781	7.569	8.178	8.609	8.473	6.886	5.310	4.914
A_3	1.565	1.621	1.716	1.818	1.940	2.061	2.174	2.306	2.432	4.559	−0.6859	−2.484	0.7425	2.256	3.866	2.167	1.399	3.947
$10^2\sigma$	2.790	2.740	2.750	2.750	2.800	2.890	2.980	3.070	3.220	20.74	19.34	25.56	29.31	31.98	30.10	18.07	11.69	10.92
									[C_4mim][NTf_2] + DEC									
A_0	−5.212	−5.441	−5.676	−5.923	−6.173	−6.432	−6.708	−6.997	−7.297	−85.67	−63.18	−47.31	−36.07	−27.99	−22.10	−17.63	−14.41	−12.42
A_1	3.048	3.186	3.315	3.438	3.593	3.732	3.894	4.072	4.257	−10.11	−3.274	0.6456	3.835	5.304	7.029	6.429	5.106	3.268
A_2	−1.145	−1.202	−1.293	−1.387	−1.489	−1.603	−1.696	−1.808	−1.918	15.06	9.876	7.670	5.930	5.239	4.048	3.214	2.835	2.305
A_3	0.2020	0.2122	0.2786	0.3746	0.4125	0.5138	0.6176	0.6803	0.7662	13.27	5.674	2.214	−1.881	−3.562	−7.824	−7.028	−4.674	−2.931
$10^2\sigma$	2.210	2.240	2.260	2.290	2.290	2.340	2.400	2.450	2.580	24.86	24.17	22.66	19.38	12.76	11.69	13.85	12.18	14.03

[a]Standard uncertainties are $u(T)$ = 0.05 K, $u(p)$ = 5 kPa.

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Table 7. Thermal Expansion Coefficients, $10^4\alpha/K^{-1}$, of ILs + Solvents at Different Mole Fractions According to Equation 5 in the Temperature Range 288.15 K to 328.15 K[a]

x_{IL}	T/K = 288.15	T/K = 293.15	T/K = 298.15	T/K = 303.15	T/K = 308.15	T/K = 313.15	T/K = 318.15	T/K = 323.15	T/K = 328.15
					[C_2mim][NTf_2] + DMC				
0.0000	12.4	12.4	12.5	12.6	12.7	12.8	12.8	12.9	13.0
0.0989	10.2	10.2	10.3	10.3	10.4	10.4	10.5	10.6	10.6
0.1983	8.96	9.00	9.05	9.09	9.13	9.17	9.21	9.26	9.30
0.2973	8.27	8.30	8.34	8.37	8.41	8.44	8.48	8.51	8.55
0.3958	7.78	7.81	7.84	7.87	7.91	7.94	7.97	8.00	8.03
0.4960	7.44	7.47	7.50	7.53	7.56	7.59	7.61	7.64	7.67
0.5954	7.16	7.19	7.21	7.24	7.26	7.29	7.32	7.35	7.37
0.6967	6.97	7.00	7.02	7.05	7.07	7.10	7.12	7.15	7.17
0.7982	6.81	6.83	6.86	6.88	6.91	6.93	6.95	6.98	7.00
0.9004	6.67	6.69	6.71	6.74	6.76	6.78	6.81	6.83	6.85
1.0000	6.61	6.63	6.65	6.67	6.70	6.72	6.74	6.76	6.79
					[C_2mim][NTf_2] + DEC				
0.0000	11.6	11.7	11.8	11.8	11.9	12.0	12.0	12.1	12.2
0.0991	10.1	9.86	9.73	9.68	9.64	9.60	9.65	9.60	9.65
0.1982	9.33	9.11	8.99	8.94	8.90	8.85	8.89	8.85	8.89
0.3024	8.77	8.56	8.44	8.40	8.35	8.30	8.34	8.29	8.33
0.3979	8.38	8.19	8.07	8.02	7.98	7.93	7.96	7.92	7.95
0.4982	8.08	7.88	7.77	7.72	7.68	7.63	7.66	7.62	7.65
0.5963	7.83	7.64	7.53	7.48	7.44	7.40	7.42	7.38	7.40
0.6992	7.63	7.44	7.33	7.29	7.24	7.20	7.22	7.18	7.20
0.7987	7.46	7.28	7.17	7.12	7.08	7.04	7.06	7.02	7.04
0.9000	7.32	7.14	7.03	6.99	6.94	6.90	6.92	6.88	6.90
1.0000	6.61	6.63	6.65	6.67	6.70	6.72	6.74	6.76	6.79
					[C_4mim][NTf_2] + DMC				
0.0000	12.4	12.4	12.5	12.6	12.7	12.8	12.8	12.9	13.0
0.0898	10.3	10.3	10.4	10.4	10.5	10.5	10.6	10.7	10.7
0.1882	9.03	9.08	9.12	9.16	9.20	9.24	9.29	9.33	9.37
0.2749	8.34	8.37	8.41	8.44	8.48	8.52	8.55	8.59	8.63
0.3754	7.87	7.90	7.93	7.96	8.00	8.03	8.06	8.09	8.12
0.4720	7.49	7.52	7.54	7.57	7.60	7.63	7.66	7.69	7.72
0.5745	7.22	7.24	7.27	7.30	7.32	7.35	7.38	7.40	7.43
0.6730	7.03	7.05	7.08	7.10	7.13	7.16	7.18	7.21	7.23
0.7799	6.86	6.89	6.91	6.93	6.96	6.98	7.01	7.03	7.06
0.8948	6.71	6.74	6.76	6.78	6.81	6.83	6.85	6.88	6.90
1.0000	6.61	6.63	6.65	6.68	6.70	6.72	6.74	6.77	6.79
					[C_4mim][NTf_2] + DEC				
0.0000	11.6	11.7	11.8	11.8	11.9	12.0	12.0	12.1	12.2
0.1025	10.0	10.1	10.1	10.2	10.2	10.3	10.3	10.4	10.5
0.2198	8.91	8.95	8.99	9.03	9.07	9.12	9.16	9.20	9.24
0.3178	8.41	8.45	8.48	8.52	8.55	8.59	8.62	8.66	8.70
0.4211	7.78	7.81	7.84	7.87	7.90	7.93	7.96	8.00	8.03
0.5268	7.46	7.49	7.52	7.54	7.57	7.60	7.63	7.66	7.69
0.6220	7.21	7.24	7.26	7.29	7.32	7.34	7.37	7.40	7.43
0.7193	7.01	7.03	7.06	7.08	7.11	7.13	7.16	7.18	7.21
0.8139	6.86	6.89	6.91	6.94	6.96	6.98	7.01	7.03	7.06
0.9088	6.72	6.74	6.76	6.79	6.81	6.83	6.86	6.88	6.90
1.0000	6.61	6.63	6.65	6.68	6.70	6.72	6.74	6.77	6.79

[a]Standard uncertainties are $u_r(\alpha) = 0.0012$, $u(T) = 0.05$ K, $u(p) = 5$ kPa, $u(x) = 0.0002$.

IL are obviously weakened, and the viscosity value decreases rapidly when organic solvents are added to the IL.

3.3. Redlich–KisterPolymomial Equation. Usually, the Redlich–Kister equation was used for the fitting of the excess molar volumes or viscosity deviations. The equation is below:

$$Y = x(1 - x)\sum_{i=0}^{3} A_i(2x - 1)^i \quad (3)$$

where Y is the excess molar volume, V^E, or viscosity deviation, $\Delta\eta$; x is the mole fraction of ILs; A_i are the adjustable parameters and determined using the least-squares methods.[37−39] For most ILs system,[24,36−39] the A_i values were taken from 0 to 3; four parameters are suitable and convenient. The adjustable parameters are listed in Table 6.

In each case, the standard relative deviation between experimental and calculated data wase used to determine the optimum number of the adjustable parameters. It was calculated according to the following equation:

$$\sigma(Y) = \left[\sum (Y_{exp} - Y_{cal})^2/(n - m)\right]^{1/2} \quad (4)$$

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where Y is the experimental and calculated values of the excess molar volume or viscosity deviation; n is the number of the experimental points over the whole concentration; and m is the number of the coefficients of the Redlich−Kister equation. The standard relative deviations are also listed in Table 6. From Table 6, it can be seen that all of the standard relative deviations are less than 0.033 $cm^3 \cdot mol^{-1}$ for density and 0.32 mPa·s for viscosity, respectively. According to the literature,[40] the standard relative deviations are in the permissible range. So, the four parameters are suitable and convenient for the excess molar volumes and viscosity deviations fitting by the Redlich−Kister equation.

3.4. Thermal Expansion Coefficient. According to the literature, the thermal expansion coefficient of the mixtures can be obtained according to the following equation:

$$\alpha_p/(K^{-1}) = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right) \quad (5)$$

where α is the thermal expansion coefficient. The values are listed in Table 7. As seen in Table 7, the values of the thermal expansion coefficient decrease with the increase of the concentration of the ILs and increase with increasing temperature. All of the values are in the range of 6×10^{-4} to 13×10^{-4} K^{-1}.

4. CONCLUSIONS

In this work, the densities and viscosities of four binary mixtures were measured by traditional methods from 288.15 K to 328.15 K. The uncertainties of the density and viscosity are $u_r(\rho) = 0.00120$ and $u_r(\eta) = 0.01$. The excess molar volumes and viscosity deviations were calculated from the experimental values. The density decreases with the cation alkyl chain length increase for the same ILs or solvents at the same mole fraction. However, the viscosity tendency is in contrast with the density. Meanwhile, the interionic interaction, especially hydrogen bonding, has a significant influence on the viscosity of ILs. The excess molar volumes and viscosity deviations are negative in the whole concentration. The orders of the absolute values are $[C_4mmim][NTf_2]$ + DMC < $[C_2mmim][NTf_2]$ + DMC < $[C_4mim][NTf_2]$ + DMC < $[C_2mim][NTf_2]$ + DMC < $[C_4mmim][NTf_2]$ + DEC < $[C_2mmim][NTf_2]$ + DEC ≈ $[C_4mim][NTf_2]$ + DEC < $[C_2mim][NTf_2]$ + DEC for excess molar volumes and $[C_2mim][NTf_2]$ + DEC < $[C_2mim][NTf_2]$ + DMC < $[C_4mim][NTf_2]$ + DEC < $[C_4mim][NTf_2]$ + DMC < $[C_2mmim][NTf_2]$ + DEC < $[C_2mmim][NTf_2]$ + DMC < $[C_4mmim][NTf_2]$ + DEC < $[C_4mmim][NTf_2]$ + DMC for viscosity deviations in the whole mole fraction.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.jced.8b00591.

^{1}H NMR spectrus of $[C_2mim][NTf_2]$, $[C_4mim][NTf_2]$, and shear rates of ILs + solvents at different mole fractions from 288.15 K to 328.15 K at atmospheric pressure (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: liuqingshan@dicp.ac.cn.

ORCID

: 0000-0002-0949-8066

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Notes

The authors declare no competing financial interest.

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