



Densities and dynamic viscosities of the binary system (water + 1-hexyl-3-methylimidazolium bromide) at different temperatures

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ABSTRACT

Densities and viscosities were measured for pure ionic liquid [C₆mim][Br] (1-hexyl-3-methylimidazolium bromide) and the binary system (water + [C₆mim][Br]) at 0.1 MPa and in the (293.15 to 333.15) K range. The excess molar volume and viscosity deviation were calculated and correlated by Redlich–Kister polynomial expansions. The fitting parameters and the standard deviations were determined.

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1. Introduction

Ionic liquids (ILs) have been recognized as novel solvents, which are liquids over a wide temperature range including room temperature [1,2]. ILs are expected to reduce or eliminate the hazards associated with volatile organic solvents and have been applied in growing applications such as solvents for organic and catalytic reactions, new material productions, solvents for separation and extraction processes, novel electrolytes for electrochemical devices and processes, and enzyme catalysis/multiphase bioprocess operations [1–5].

Generally, there are three ways to use ILs in a biocatalytic process: as a pure solvent, as a co-solvent in aqueous systems, and in a biphasic system [6]. To design any process involving ILs on an industrial scale, it is necessary to know a range of physical properties including viscosity and density. Furthermore, the presence of water in ILs can dramatically affect their physicochemical properties [7–9]. The excess thermodynamic properties are of great importance in understanding the nature of molecular aggregation

that exists in the binary mixtures over the whole composition range [10].

A number of groups have studied the physicochemical properties of aqueous solutions of imidazolium-based ILs [10–14]. Ge *et al.* reported dynamic viscosities, densities for the binary mixtures of (water + [C₄mim][TfO]) over the entire mole fraction range from (303.15 to 343.15) K [10]. Goómez *et al.* reported dynamic viscosities, densities of (water + [C₆mim][Cl]) and (water + [C₈mim][Cl]) over the entire mole fraction range from (298.15 to 343.15) K [11]. Brennecke and Rodriguez reported dynamic viscosities, densities of (water + [C₂mim][EtSO₄]), (water + [C₂mim][TfO]), and (water + [C₂mim][TFA]) over the entire mole fraction range [12]. Shekaari and Mousavi reported osmotic coefficients, mean activity coefficients, vapor pressure data, and excess Gibbs free energies G^E of aqueous solutions of [C_{*n*}mim][Br] ($n = 3, 5, 6$) [15a]. Wang *et al.* measured the densities, conductivities, and polarity indexes of pyrene for aqueous solutions of a series of [C_{*n*}mim][Br] ($n = 4, 6, 8, 10, 12$) and [C₄mim][BF₄] at 298.15 K, and discussed the aggregation behaviors of these aqueous IL solutions [15b]. However, relative few references have been made on the binary systems {water + [C_{*n*}mim][Br] ($n = 1, 2, \dots, m$)} [15a,b], and no reference on the transport properties depending on the whole composition range and/or temperature. Therefore, in this work the densities and dynamic viscosities were measured for pure [C₆mim][Br] and the binary system (water + [C₆mim][Br]) in the (293.15 to 333.15) K range and at atmospheric pressure over the whole

Abbreviations: [C_{*n*}mim]⁺, 1-alkyl-3-methylimidazolium; [C₄Py]⁺, [C₈Py]⁺, 1-butylpyridinium, 1-octylpyridinium; [Pyr]⁺, pyrrolidinium; [EtSO₄][−], ethylsulfate; [TfO][−], trifluoromethanesulfonate; [TFA][−], trifluoroacetate.

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TABLE 1
Densities and viscosities of pure [C₆mim][Br] in the (293.15 to 333.15) K range.

T/K	$\rho/(\text{g} \cdot \text{cm}^{-3})$	$\eta/(\text{mPa} \cdot \text{S})$
293.15	1.2274	6936
298.15	1.2235	3986
303.15	1.2201	2239
308.15	1.2159	1386
313.15	1.2133	851.65
318.15	1.2091	622.34
323.15	1.2057	512.14
328.15	1.2022	412.18
333.15	1.1986	350.69

composition range. Excess molar volumes and viscosity deviations over the entire composition range were obtained from the experimental data and correlated by Redlich–Kister polynomial [16].

TABLE 2
Experimental densities ρ , excess mole volumes V^E , dynamic viscosities η , and viscosity deviations $\Delta\eta$ for the binary system {water (1) + [C₆mim][Br] (2)} in the (293.15 to 333.15) K range.

x_1	$\rho/(\text{g} \cdot \text{cm}^{-3})$	$\eta/(\text{mPa} \cdot \text{S})$	$V^E/(\text{cm}^3 \cdot \text{mol}^{-1})$	$\Delta\eta/(\text{mPa} \cdot \text{S})$	$\rho/(\text{g} \cdot \text{cm}^{-3})$	$\eta/(\text{mPa} \cdot \text{S})$	$V^E/(\text{cm}^3 \cdot \text{mol}^{-1})$	$\Delta\eta/(\text{mPa} \cdot \text{S})$
<i>T = 293.15 K</i>								
0	1.2274	6936	0	0	1.2201	2239	0	0
0.1225	1.2234	3519	0.171	-2567	1.2161	1255	0.183	-709.84
0.2196	1.2204	1578	0.180	-3835	1.2132	695.45	0.186	-1052
0.2978	1.2179	776.62	0.134	-4094	1.2108	395.49	0.135	-1177
0.3480	1.2163	507.80	0.072	-4015	1.209	265.62	0.101	-1194
0.4166	1.2134	313.52	0.022	-3733	1.2062	168.31	0.046	-1138
0.5040	1.2087	173.73	-0.039	-3267	1.2016	98.93	-0.016	-1012
0.5701	1.2039	111.42	-0.068	-2871	1.1968	65.77	-0.039	-897.35
0.6405	1.1969	70.83	-0.074	-2423	1.1899	43.45	-0.044	-762.11
0.7285	1.1843	36.40	-0.076	-1848	1.1773	22.54	-0.035	-586.08
0.8160	1.1637	18.13	-0.066	-1259	1.1569	12.10	-0.022	-400.69
0.9048	1.1236	6.655	-0.051	-654.83	1.1173	4.764	-0.009	-209.29
0.9241	1.1094	4.951	-0.046	-522.7	1.1036	3.619	-0.013	-167.25
0.9440	1.0912	3.480	-0.044	-386.17	1.0854	2.622	-0.005	-123.71
0.9613	1.0709	2.438	-0.039	-267.24	1.0657	1.860	-0.008	-85.75
0.9899	1.0226	1.342	-0.019	-70.00	1.0192	1.047	-0.011	-22.56
1	0.9982	1.304	0	0	0.9956	1.002	0	0
<i>T = 313.15 K</i>								
0	1.2133	851.65	0	0	1.2057	512.14	0	0
0.1225	1.2093	503.66	0.192	-243.74	1.2018	302.69	0.186	-146.78
0.2196	1.2064	320.14	0.201	-344.63	1.1988	180.56	0.217	-219.23
0.2978	1.204	194.10	0.154	-404.12	1.1962	117.64	0.198	-242.15
0.3480	1.2022	143.50	0.123	-412	1.1946	89.65	0.145	-244.45
0.4166	1.1995	97.98	0.060	-399.14	1.192	60.85	0.075	-238.16
0.5040	1.1949	60.18	0.004	-362.57	1.1875	38.59	0.015	-215.71
0.5701	1.1901	41.24	-0.014	-325.26	1.1827	27.69	0.003	-192.79
0.6405	1.1833	28.14	-0.021	-278.45	1.176	19.39	-0.006	-165.07
0.7285	1.1707	15.134	-0.004	-216.56	1.1635	10.75	0.015	-128.69
0.8160	1.1501	8.492	0.026	-148.74	1.1429	7.247	0.056	-87.431
0.9048	1.1107	3.519	0.039	-78.149	1.1044	2.744	0.051	-46.504
0.9241	1.097	2.764	0.036	-62.48	1.0909	2.165	0.048	-37.209
0.9440	1.0795	2.334	0.028	-45.97	1.0736	1.600	0.040	-27.593
0.9613	1.0604	1.469	0.014	-32.12	1.0547	1.547	0.027	-18.796
0.9899	1.0151	0.846	-0.004	-8.402	1.0101	0.695	0.006	-5.016
1	0.9922	0.653	0	0	0.988	0.544	0	0
<i>T = 333.15 K</i>								
0	1.1986	350.69	0	0				
0.1225	1.1946	201.69	0.209	-106.1				
0.2196	1.1915	115.66	0.256	-158.12				
0.2978	1.1890	77.41	0.225	-168.98				
0.3480	1.1874	57.52	0.172	-171.29				
0.4166	1.1848	40.03	0.102	-164.76				
0.5040	1.1803	26.69	0.044	-147.49				
0.5701	1.1758	19.36	0.008	-131.67				
0.6405	1.169	14.63	0.012	-111.74				
0.7285	1.1565	7.929	0.038	-87.624				
0.8160	1.1363	4.762	0.066	-60.146				
0.9048	1.0976	2.184	0.079	-31.624				
0.9241	1.0842	1.733	0.074	-25.316				
0.9440	1.0672	1.301	0.060	-18.778				
0.9613	1.0485	0.982	0.046	-13.039				
0.9899	1.0042	0.588	0.024	-3.4163				
1	0.9832	0.467	0	0				

2. Experimental section

The deionized water was distilled in a quartz still, and its conductance is $0.8\text{--}1.2 \cdot 10^{-4} \text{ S} \cdot \text{cm}^{-1}$. All chemicals used in this study were of reagent grade with the claimed purity more than 99%. *N*-methylimidazole and *n*-C₆H₁₃Br were supplied by Shanghai Jiachen Chemical Ltd. All chemicals used were refined by the traditional methods before use. [C₆mim][Br] was prepared with well-established procedures by our previous works [17a,b]. The resulting after purification was dried under vacuum over CaCl₂ for days at 70 °C. It was further dried with 3 Å molecular sieve for days immediately before use. ¹H NMR (600 MHz, d₆-DMSO) for [C₆mim][Br] agrees well with literature [17c]. The water content after drying, measured by Karl–Fisher titration, was within 0.012 wt%.

3. Apparatus and procedure

The experimental procedures are similar to those used in our previous studies [18,19] and are described briefly as follows. The binary aqueous solutions were prepared by syringing weighed amounts of the pure liquids into stoppered bottles in glove box using a Sartorius CT225D balance with the precision of $\pm 5 \cdot 10^{-5}$ g. The solutions were placed into stoppered bottles and stirred for 2h, and all solutions were made in glass flasks and measurements were carried on one week after preparation to assure complete dissolution and aggregation. All the samples were prepared immediately before the density and viscosity measurements to prevent possible loss of water due to evaporation. Densities of solutions were measured with a KEM oscillating-tube digital densimeter (DA-505) thermostated to better than ± 0.01 K. All measurements were performed at atmospheric pressure. The temperature in the measuring cell was monitored with a digital thermometer. The densimeter was calibrated by double-distilled water and dry air at the temperatures ranging from (283.15 to 323.15) K prior to its use. The experimental uncertainty was less than $\pm 5 \cdot 10^{-5}$ g · cm⁻³. Dynamic viscosities of the IL and its binary solutions were measured using modified Cannon–Ubbelohde suspended level capillary viscometers of (0.4, 0.6, 0.8, 1.5, 2.0, and 3.0) mm in diameters. A thoroughly cleaned and perfectly dried viscometer filled with liquid was placed vertically in a glass sided water thermostat. After attaining thermal equilibrium the efflux times of flow of the liquids were recorded with a digital stop watch with a precision of ± 0.01 s. The capillary viscometers are calibrated and credited by the company, and all the deviations within 0.2%. Triplicate measurements were performed for each composition.

4. Results and discussion

4.1. Pure component

The measured densities and viscosities of pure [C₆mim][Br] in the (293.15 to 333.15) K range are shown in table 1. The measured densities at (293.15 and 303.15) K were in good agreement with the data reported in literature [20], while no viscosity data were referred for pure [C₆mim][Br] in literature by our search. Experimental values of $\ln \rho$ for pure [C₆mim][Br] against T were fitted to the following equation:

$$\ln \rho = a + bT, \quad (1)$$

where ρ is the density. T is the absolute temperature, a and b are fitting parameters. The measured densities can be well described by equation (1), with $R^2 = 0.99927$. The coefficient of thermal expansion for [C₆mim][Br], α_p , is defined by the following equation:

$$\alpha_p = 1/V(\partial V/\partial T)_p = -(\partial \ln \rho/\partial T)_p, \quad (2)$$

where V is the molar volume of pure IL. Then, $\alpha_p = -b = 5.90 \cdot 10^{-4} \pm 0.06$ K⁻¹ for [C₆mim][Br] is obtained from the slope of the fitting line. Generally, the practical IL use was under 373.15 K, with a straight line for $\ln \rho$ versus T in this temperature range, so thermal expansion for each typical IL could consider as a constant. Thermal expansion for [C₆mim][Br] is in the common range for typical ILs [11,12,21], and is much lower than conventional molecular solvents [22].

The viscosities of pure [C₆mim][Br] at the temperatures ranging from (293.15 to 333.15) K were fitted to the Vogel–Fulcher–Tamman (VFT) [23,24]:

$$\eta = AT^{-0.5} \exp\left(\frac{k}{T-T_0}\right), \quad (3)$$

where η is the dynamic viscosity, T is the absolute temperature. A , k , and T_0 are adjustable parameters. The parameter T_0 is the “ideal glass transition temperature”, and it should be slightly below the experimental glass transition temperature T_g [12]. The measured viscosities can be well described by equation (3), with $R^2 = 0.99875$, while $T_0 = 216.2 \pm 13.8$ is slightly higher than the experimental data for [C₆mim][Br] with $T_g = 224$ K [25]. We can consider VFT equation as an empirical equation to correlate the transport properties just as originally proposed, with T_0 close to glass transition temperature.

4.2. Binary system

The dynamic viscosities and densities of the system (water + [C₆mim][Br]) in the (293.15 to 333.15) K range and at 0.1 MPa are shown in table 2. It is clear that their densities and dynamic viscosities decrease with increasing the temperature and the mole fraction of water and that the decreases are more notable for the dynamic viscosities. The measured densities and viscosities of water shown in table 2 were in good agreement with the values reported in the literature [26].

The excess molar volumes and viscosity deviations are calculated from the experimental data by the following equations:

$$V^E = \sum_{i=1}^N x_i M_i (\rho^{-1} - \rho_i^{-1}), \quad (4)$$

$$\Delta \eta = \eta - \sum_{i=1}^N x_i \eta_i, \quad (5)$$

where ρ and ρ_i are the density of the binary system and the density of the pure component, respectively. x_i and M_i represent the mole fraction and the molar mass of the i component. η and η_i are the dynamic viscosity of the binary system and the pure component, respectively.

The excess molar volumes and viscosity deviations for the binary system (water + [C₆mim][Br]) in the (293.15 to 333.15) K range and at 0.1 MPa are also shown in Table 2. The excess molar volumes and viscosity deviations at each temperature were fitted to a Redlich–Kister [16] equation:

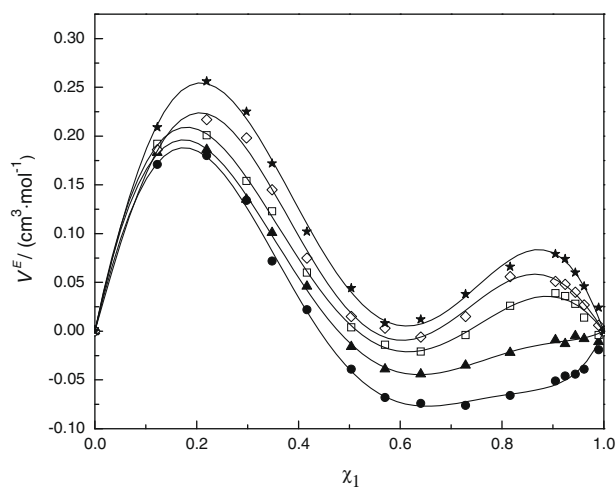


FIGURE 1. Excess mole volumes V^E versus mole fraction of water for the binary system {water (1) + [C₆mim][Br] (2)} at different temperatures: ●, 293.15 K; ▲, 303.15 K; □, 313.15 K; ◇, 323.15 K; ★, 333.15 K. The symbols represent experimental values, and the solid curves represent the corresponding correlations by the Redlich–Kister equation.

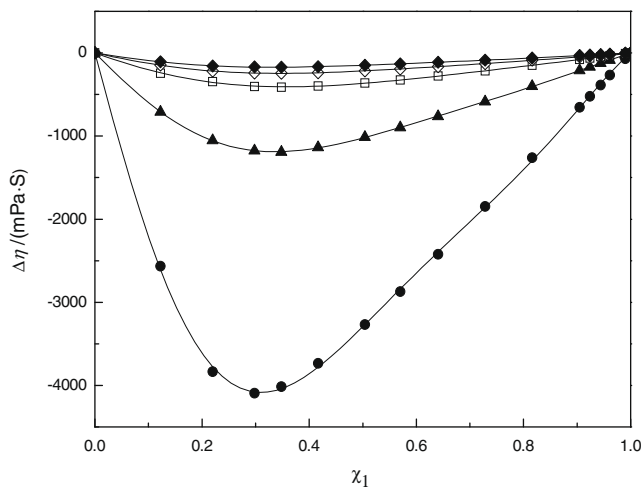


FIGURE 2. Viscosity deviation $\Delta \eta$ versus mole fraction of water for the binary system {water (1) + [C₆mim][Br] (2)} at different temperatures: ●, 293.15 K; ▲, 303.15 K; □, 313.15 K; ◇, 323.15 K; ◆, 333.15 K. The symbols represent experimental values, and the solid curves represent the corresponding correlations by the Redlich–Kister equation.

TABLE 3

Fitted parameters and standard deviations (σ), for binary system {water (1) + [C₆mim][Br] (2)} in the (293.15 to 333.15) K range.

	A_0	A_1	A_2	A_3	A_4	σ
			$T = 293.15\text{K}$			
$V^E/(\text{cm}^3 \cdot \text{mol}^{-1})$	-0.15605	-1.10117	1.9356	-0.57149	-1.29704	0.004
$\Delta\eta/(\text{mPa} \cdot \text{S})$	-13175.19	12770.74	-9949.69	-3536.04	8321.94	27.642
			$T = 303.15\text{K}$			
$V^E/(\text{cm}^3 \cdot \text{mol}^{-1})$	-0.04671	-0.99983	1.85237	-0.35876	-0.70241	0.003
$\Delta\eta/(\text{mPa} \cdot \text{S})$	-4094.44	3385.93	-1719.60	-1216.32	1583.86	2.920
			$T = 313.15\text{K}$			
$V^E/(\text{cm}^3 \cdot \text{mol}^{-1})$	0.02943	-0.99433	2.08156	0.06658	-0.50442	0.004
$\Delta\eta/(\text{mPa} \cdot \text{S})$	-1467.04	1022.36	-328.55	-290.95	229.62	3.171
			$T = 323.15\text{K}$			
$V^E/(\text{cm}^3 \cdot \text{mol}^{-1})$	0.08133	-1.14213	2.63309	0.6285	-1.39228	0.005
$\Delta\eta/(\text{mPa} \cdot \text{S})$	-867.64	634.74	-309.46	-162.24	240.56	0.751
			$T = 333.15\text{K}$			
$V^E/(\text{cm}^3 \cdot \text{mol}^{-1})$	0.16825	-1.28473	2.68025	0.86554	-0.98918	0.005
$\Delta\eta/(\text{mPa} \cdot \text{S})$	-593.54	464.66	-284.26	-98.03	201.35	0.916

$$\Delta Q = x_i x_j \sum_{p=0}^N A_p (x_i - x_j)^p, \quad (6)$$

where ΔQ and x are the excess molar volume or viscosity deviation and the mole fraction, respectively. A_p and N are the fitting parameter and the degree of the polynomial expansion, respectively.

The comparisons of the experimental excess molar volumes V^E and the viscosity deviations $\Delta\eta$ with those obtained from the Redlich–Kister polynomial equation for the binary system are represented in figures 1 and 2, respectively. Table 3 demonstrates the values of the parameters A_p together with the standard deviation σ defined by

$$\sigma = \left\{ \sum_i^n (\Delta Q_{\text{exp}} - \Delta Q_{\text{cal}})^2 / n \right\}^{1/2}, \quad (7)$$

where the subscripts “exp” and “cal” stand for the experimental and the calculated properties, respectively, and n is the number of experimental data points.

A glance of figure 1 reveals that, at higher temperatures, the system (water + [C₆mim][Br]) exhibits the positive excess volume over the whole composition range. The increase in the magnitude of the positive V^E values with increasing temperature can be attributed to the decreasing importance of hydrogen bonding with increasing temperature, which agrees well with the results of other groups [9–12,27,28]. In fact, the positive deviation of the excess volumes indicates that the interactions are weak. However, it displays negative excess volumes at 293.15 K as x_1 exceeds 0.4 to 0.5. We can observe that the excess molar volumes present a minimum at $x_1 \approx 0.5$ to 0.6, and two maximums at $x_1 \approx 0.15$ and $x_1 \approx 0.85$ to 0.9 for this system. For example, note that, for the system (water + [C₆mim][Cl]), the V^E is mostly always negative, with a minimum at $x_1 \approx 0.55$, and the minimum can be due to hydrogen bonds between water molecules and ionic liquids [11]. For binary systems (water + [C₄mim][BF₄]) and (water + [C₄mim][I]), decided by titration calorimetry, eventually for $x_1 \leq 0.5$ to 0.6, IL molecules form clusters of their own kind, as in their pure states [29]. So we can conclude that at $x_1 \approx 0.5$ to 0.6, due to hydrogen bonds between water molecules and ionic liquid, molecules form clusters of their own kind, as in their pure states for (water + [C₆mim][Br]). When $x_1 \approx 0.85$ to 0.9, the maximum was due to the dissociation of the ions forming ionic liquid, the similar maximum appeared in other references [9,11,12]. While the maximum at $x_1 \approx 0.15$ is different from the (water + [C₆mim][Cl]) system [11]. The binary system (water + [C₆mim][Cl]) did not have the maximum at $x_1 \approx 0.15$ [11], and that can be explained that [C₆mim][Cl] has stronger interactions with water, resulting in negative excess volumes [11]. The above phenomenon follows the hydrophilic order of [Br]⁻ and [Cl]⁻, and we assumed that increasing x_1 can gradually form hydrogen bonds for [C₆mim][Br] not like [C₆mim][Cl] [11]. V^E values can be greatly influenced by measured densities and experimental methods (IL aqueous solution is a complicated system in equilibria, and it is hard to control the water content when $0 \leq x_1 \leq 0.4$), and we should take the maximum at $x_1 \approx 0.15$ slightly. For similar binary systems (water + [C₂mim][TfO]) and (water + [C₄mim][TfO]), the V^E versus x_1 has somehow different trend [10,12]. That mixtures of water with [C₂mim][EtSO₄] and [C₂mim][TFA] exhibit mostly negative deviations from ideality, while [C₂mim][TfO] has different trend [12]. They owed the reason that [TfO]⁻ anion is less hydrophilic than [EtSO₄]⁻ or [TFA]⁻, and [C₂mim][EtSO₄] and [C₂mim][TFA] have stronger interactions with water, resulting in negative excess volumes [12]. The mixture of water with [C₄Py][BF₄] or [C₈Py][BF₄] also has positive V^E deviation from ideality with a maximum at $x_1 \approx 0.5$ [9]. The values of excess molar volume were positive for (water + [C₄mim][TfO]) at all temperatures and over the entire range of compositions with two or more maximums [10], while no further discussion was revealed about that. Excess volumes for ((water + [Pyrr][HSO₄]) and (water + [Pyrr][NO₃])) systems at 298.15 K are strongly positive. At the opposite

when the anion is [HCOO]⁻, negative values were observed even for the pyrrolidinium cation [28]. For binary systems (water + [Pyrr][HSO₄]) and (water + [Pyrr][NO₃]), the maximum $x_1 \approx 0.5$ was formed by H-bonds breaking up [28]. The formation of strong H-bonds between water and ions leads to an increase in the volume occupied by ions or molecules. From comparing the V^E for ((water + [C₄Py][BF₄]) and (water + [C₈Py][BF₄])), ((water + [C₆mim][Cl]) and (water + [C₈mim][Cl])), and ((water + [C₂mim][TfO]) and (water + [C₄mim][TfO])) respectively, we can conclude that, V^E values increase in the magnitude with increasing the alkyl chain length of the imidazolium and pyridinium cations for ILs with the same anions [10–12]. In the general, the anion can greatly influence on the excess molar volume for (water + IL) systems, and they can follow the hydrophilic order of anion for the IL with the same cation [10–12].

The comparisons of the experimental viscosity deviations with those obtained from the Redlich–Kister polynomial equation for the system (water + [C₆mim][Br]) are illustrated in figure 2. Negative deviations are observed over the whole temperature and composition ranges and the minimum deviation always appears at $x_1 \approx 0.3$.

The viscosity deviations decrease with increasing of water mole fraction up to $x_1 \approx 0.3$ for (water + [C₆mim][Br]). This behavior becomes reversed for this system at higher mole fractions of water. The similar phenomenon appeared in other (water + IL binary) systems [9–11], with the minimum deviation $x_1 \approx 0.3$ for (water + [C₄Py][BF₄] or [C₈Py][BF₄]) [9], $x_1 \approx 0.2$ for (H₂O + [C₄mim][TfO]) [10], and $x_1 \approx 0.2$ for (water + [C₆mim][Cl] or [C₈mim][Cl]) [11], and no further explanations were revealed in this references [9–11]. We think that the structures of aqueous solutions somehow changed at $x_1 \approx 0.3$, but we did not have direct evidence to approve our views. With the development of molecular dynamics simulation [30,31] and spectra measurements, more information will be obtained about the structure of IL aqueous solution. Increasing the temperature diminishes the viscosity deviations progressively from $T = (293.15$ to $333.15)$ K.

5. Conclusions

The densities and viscosities of the pure [C₆mim][Br] were measured in the (293.15 to 333.15) K range and at atmospheric pressure. The densities and viscosities of the binary system (water + [C₆mim][Br]) in (293.15 to 333.15) K range were also determined over the whole composition range. The excess molar volumes and viscosity deviations for the binary system were calculated from the experimented data. The excess molar volumes of the system (water + [C₆mim][Br]) are positive at higher temperatures, whereas negative deviations from the ideality is observed at 293.15 K as the x_1 exceeds 0.4 to 0.5. Contrarily, the system shows the negative viscosity deviations over the whole temperature and composition ranges.

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