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Synthesis, characterization of 2′, 3′-epoxy propyl - N-methyl-2- oxopyrrolidinium salicylate ionic liquid and study of its interaction with water or methanol

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Abstract

Important physico-chemical properties of ionic liquids (ILs) can be manipulated by adjusting the nature of the cation or anion. These properties are exploited in applications such as organic synthesis, catalysis and electrochemical process to mention a few. In this work, the novel pyrrolidone ionic liquid N- (2′, 3′-epoxypropyl)-N-methyl-2-oxopyrrolidinium salicylate $[EPMpyr]^+[SAL]^-$ was synthesized using two step and characterized. The temperature dependent density and speed of sound for ionic liquid, methanol, water, and their corresponding binary mixtures of {IL (1) + methanol or water (2)} were measured over the entire range of mole fractions at temperatures from T = (293.15 to 313.15) K in steps of 5 K, under atmospheric pressure. The calculated thermodynamic properties such as excess molar volume V_m^E, isentropic compressibility k_s, intermolecular free length L_f, and deviation in isentropic compressibility Δk_s, were derived from the investigated density and speed of sound data. The resulting experimental data for excess molar volumes V_m^E, intermolecular free length L_f, and deviation in isentropic compressibility Δk_s, were well fitted to the Redlich–Kister polynomial equation. The effect of temperature and concentration on thermophysical properties were also provided.

Keywords: Density, Speed of sound, N- (2′, 3′-epoxypropyl)-N-methyl-2-oxopyrrolidinium salicylate, Water, Methanol, Redlich–Kister equation.

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1. Introduction

Ionic liquids (ILs) are low melting salts which are a combination of cations and anions; cations are usually found in the organic part of the molecule whilst the anion may be inorganic or organic in nature. These ILs have been developed in the last two decades. Nowadays ILs are an important research area of study, with many researchers focusing on fundamental physical and chemical properties, such as density, ρ, viscosity, η sound velocity, u, low vacuum pressure, Pa, low melting point, high conductivity, S, and solubility.[1] The density, viscosity, and sound velocities are essential for developing industrial process and design.[1-4]

Industrial chemicals are being manufactured by using environment-friendly green solvents ILs, instead of toxic organic volatile solvents.[5-7] In the last decade, concerted attention by the scientific community has significantly improved the nature and potential applications of ILs, in particular, because of their exclusive physico-chemical properties.[8-11] Most of the industrial technological applications of ILs are occurring in mixtures, whilst the number of research groups has increased worldwide.[12-16] Properties such as vapor-liquid equilibrium, liquid-liquid equilibrium and importantly the physico-chemical properties of mixtures are studied. ILs have interesting properties such as negligible vapor pressure,[17] high ionic conductivity.[18] high thermal stability,[19] chemical and electrochemical stability, non-flammability[20] and low or negligible toxicity; these are the potential variables to completely substitute or replace the conventional organic solvents as electrolyte solutions or as co-solvents or additives to improve productivity and performance in industrial applications. The more important physico-chemical properties of ILs such as density, ρ, speed of sound, u, refractive index, n, conductivity, s, polarity and dielectric permittivity's can be adjusted by exchanging cations or anions. These significant properties have been exploited in a several successful applications such as organic synthesis,

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catalysis, separation technology, extraction and electrochemical processes.[1-4] In the modern world ILs are used as environment-friendly green and clean solvents for an excessive diversity of materials:- pharmaceuticals,[21] biomass feedstocks[22-24] and greenhouse gases.[25]

Ionic liquids have materialized as a successful alternative to substitute for traditional toxic volatile organic solvents for separation of aromatic hydrocarbons from liquid mixtures,[26] due to their exclusive properties such as non-flammability, reusable capability, negligible volatilities, high thermal stabilities, non-corrosiveness, to be co-ordinate to a specific application by the combination of altered cation and anion with aliphatic hydrocarbons,[27, 28] Currently ILs have been applicable more in pharmaceutical industries, solvent and anti-solvent for active pharmaceutical ingredient (API),[29,30] thereby improving water solubility of API[31] used to extract biological components from active materials,[32] and as a medium to synthesize pharmaceutically active materials.[33] Ionic liquids are often used for those applications to reduce cost as well as the viscosity of the materials.

Pyrrolidinium based ILs are potentially applicable as an electrolyte in batteries due to its attractive properties, some of the research done using pyrrolidinium based ILs in lithium batteries such as 1-(2-methoxyethyl)-1-methylpyrrolidinium bis-(trifluoro methylsulfonyl) imide are used as a potential alternate for electrolyte components to substitute volatile toxic organic solvents in supercapacitors and lithium batteries.[34,35] Pyrrolidinium dicyanamide ILs are successful candidates for application as electrolytes in electrochemical double layer capacitors (EDLCs).[4-7]

In recent years, the usage of ILs have been increased as potential solvents to extract aromatic hydrocarbons from aliphatic hydrocarbons.[26, 36-40] The thermo-physical properties of N-butyl-N-methyl-2-oxopyrrolidinium bromide was measured and reported at several temperatures from

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(293.15-343.15) K.[41] The present work discloses, the synthesis, characterization and determination of thermo-physical properties of novel N-2′,3′-epoxy propyl-N-methyl-2-oxo pyrrolidinium salicylate and its binary mixtures of water or methanol to understand the molecular interactions which occurs in this solutions. The present work is a part of our investigations on physicochemical properties of ILs with solvents at different temperatures.[41-53]

2. Experimental Section

2.1. Materials

N-methyl-2- pyrrolidone, epichlorohydrin, sodium salicylate, acetonitrile, methanol, acetone, and hexane were purchased from Fluka Chemicals with purity of ≥ 99%. The purity and density of the pure compounds in comparison with literature[54-59] values are presented in Table 1. Ultra-pure deionized water was used in all experiments. The water content using a Metrohm Karl Fishcher coulometer (model KF Titrando) was found to be 0.05% in N-2′, 3′-epoxy propyl-N-methyl-2-oxo pyrrolidinium salicylate $[EPMpyr]^+[SAL]^-$.

2.2. Step 1:- synthesis of N-(2′, 3′-epoxypropyl)-N-methyl-2-oxo pyrrolidinium chloride

The reaction system was set up as follows: A 500 mL three-necked round bottomed flask with a thermometer inlet over cold water flowing condenser was used. Nitrogen gas was flushed into the round bottomed flask 1.0 mol of freshly distilled N-methyl-2-pyrrolidone was mixed with 100 mL of acetonitrile, followed by 1.10 mol of epichlorohydrin. The mixture was now brought to a moderate reflux (90-100) ^oC, then heated under reflux for 48 hours with constant stirring and finally cooled to room temperature. The volatile materials were removed under reduced pressure to give a yellow coloured ionic liquid, N-(2′, 3′-epoxypropyl) -N-methyl -2- oxopyrrolidinium chloride. The structure was confirmed by FTIR, ^{1}HNMR, ^{13}CNMR and Elemental Analysis.

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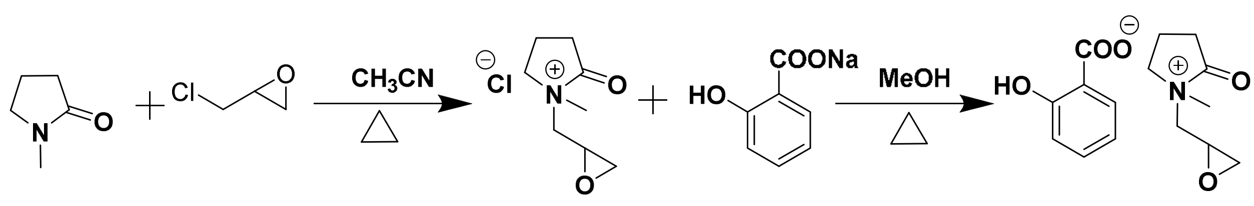
2.3. Characterization of N-(2′, 3′-epoxypropyl)-N-methyl- pyrrolidonium chloride

The $[EPMpyr]^+[Cl]^-$ was characterized by the following technique: NMR (^{1}H and ^{13}C), elemental analysis and FTIR. FTIR ($\nu = cm^{-1}$): 3442, 2995, 1621, 1501, 1403, 1332, 1256, 1113, 967, 856, 756, 679, 561, 479. $[EPMpyr]^+[Cl]^-$ 1H NMR (400 MHz, DMSO): δ 3.48 - 3.51 (m, 1H), 3.30 - 3.32 (t, 2H), 2.76 - 3.29 (s, 1H), 2.61 - 2.62 (s, 3H). 2.26-2.30 (d, 1H) 1.96 - 1.98 (t, 2H) 1.90 - 1.94 (m, 2H) ^{13}C NMR (100 MHz, DMSO): δ 175.03, 51.22, 49.38, 45.72, 45.00, 30.62, 29.50, and 17.59. Elemental Analysis (in %): Theoretical calculation for: $C_8H1_4NO_2$: C, 50.14; H, 7.36; N, 7.31; The values found (in %) are C, 50.45; H, 7.10; N, 7.17.

2.4. Step 2:- synthesis of N-(2′, 3′-epoxypropyl)-N-methyl-2-oxo pyrrolidinium salicylate

The N-(2′, 3′, epoxypropyl)-N-methyl-2-oxopyrrolidinium salicylate was synthesized by dissolving the desire quantity 1.12 mole of sodium salicylate separately in methanol to make a clear solution in round bottomed flask. Then, the above synthesized intermediate IL N-(2′,3′-epoxypropyl)-N-methyl-2-oxopyrrolidium chloride was added to exchange the salicylate anion. The product was purified by a solvent wash with acetone, petroleum ether and hexane to remove unwanted starting materials and sodium chloride, then distilled again at 80 ^{o}C for 48 hrs to get pure moisture free ILs. The product identity was established with FTIR, NMR (proton and carbon) and elemental analysis. Scheme for synthesis of 2′, 3′-epoxy propyl - N-methyl-2-oxopyrrolidinium salicylate given below:



I II

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Scheme for synthesis of 2′, 3′-epoxy propyl - N-methyl-2- oxopyrrolidinium salicylate

2.5. Characterization of N-(2′, 3′-epoxypropyl)-N-methyl- pyrrolidonium salicylate

The $[EPPY]^{+}[SAL]^{-}$ was characterized by the following methods: NMR (^{1}H and ^{13}C), elemental analysis and FTIR. The structure of $[EPMpyr]^{+}$ $[SAL]^{-}$ is as shown in Figure 1. FTIR ($\nu = cm^{-1}$): 3442, 2995, 1621, 1501, 1403, 1332, 1256, 1113, 967, 856, 756, 679, 561, 479.$[EPPYR]^{+}[SAL]^{-}$ 1H NMR (400 MHz, DMSO): δ1.9 - 2.0(M, 2H), 2.15 - 2.3(t, 2H), 2.7 - 2.8(s, 3H), 3.3 - 3.4(m, 3H), 3.5 -3.65(d, 2H), 3.66 - 3.90(m, 1), 4.0 - 4.2(m, 1), 6.75 - 6.85(t, 1H), 6.86 - 7.00(m, 1H), 7.10 - 7.30(t, 1H), 7.40 - 7.60(m, 1H), 7.65 - 7.95(d-d, 1H). 13C NMR (100 MHz, DMSO): δ18, 30, 33, 51, 65, 72, 75, 115, 120, 122, 132, 134, 138, 163 and 178. Elemental Analysis (in %) Theoretical calculation for: $C_{15}H_{21}NO_{4}$: C, 64.50; H, 7.58; N, 5.01; O, 22.91; the values found (in %) are C, 64.95; H, 7.10; N, 5.28; O, 23.36.

2.6. Apparatus and procedure

Anton Parr DSA 5000 M vibrating tube digital densitometer and speed of sound analyzer were used to determine the density and speed of sound of IL and their binary mixtures simultaneously. Temperature and pressure are important parameters to affecting physical properties, and were controlled to ± 0.01 K and 101 kPa respectively. Doubly distilled ultra-pure water was used to calibrate the instrument according to the method of Lagourette et al.[60] The {IL (1) + methanol or water (2)} binary mixture samples were prepared by weighing on a Mettler Toledo AG245, which has a precision of 0.0001 g. The estimated uncertainty in density and speed of sound was less than $\pm 2 \times 10^{-4}$ $g\cdot cm^{-3}$ and ± 0.09 $m\cdot s^{-1}$, respectively.

3. Result and Discussion

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The density ρ, and speed of sound u, are interesting volumetric properties which are important for industrial processes and development. Figures 2 to 5, show the investigated values of density and speed of sound data, the volumetric properties of pure ionic liquid (IL) $[EPMpyr]^+[SAL]^-$ and their binary mixtures with methanol or water, were measured under atmospheric pressure from T = (293.15 - 313.15) K.

Tables 2 and 3, show the experimental values of density, ρ, speed of sound u, excess molar volume V_m^E, isentropic compressibility k_s, deviation in isentropic compressibility Δk_s and intermolecular free length L_f corresponding to several mole fractions of IL systems.

Those systems are formed by ionic liquids and it's binary mixtures of methanol or water, viz. $\{[EPMpyr]^+[SAL]^-$ (1) + methanol (2)}; $\{[EPMpyr]^+[SAL]^-$ (1) + water (2)} at (293.15, 298.15, 303.15, 308.15 and 313.15) K, respectively. All combinations were mixed well to give a homogeneous solution across the entire mole fraction range.

The measured data of ρ and u of pure $[EPMpyr]^+[SAL]^-$, methanol, water, and their binary mixtures are display in Tables 2 and 3 as a function of IL mole fraction (x_1) for entire composition range at temperature from (293.15 to 313.15) K in steps of 5 K under atmospheric pressure. The Figures 6 to 8 were plotted based on the investigated values and these are V_m^E, Δk_s, and L_f as a function of the IL mole fraction at different temperatures of binary mixtures.

Here see the Figures, in water with IL binary mixtures graphs, look like waves, that mean its accelerating due to initially, at the time of mixing pyrrolidonium salicylate IL interact with water to forms slight white precipitates, after shaking it becomes homogeneous liquids in every mole

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fraction of combinations. These are because of anionic effect, here salicylate anion plays a major role in that interaction.

In addition, the curves obtained with the parameters listed in Tables 2 and 3, have also been included. Normally, ILs is completely miscible with solvents, which have more dielectric constants otherwise ILs are not completely miscible.[61-63] The Figures 2 and 3 shows the temperature dependent density values. From the measured data, the density of pure IL was greater then it's starting organic compound. Additionally, the densities of the binary mixture or pure ILs decreases with increase the temperature.

The excess molar volume, V_m^E was calculated from the investigated density data list by using the following equation (1):

$$V_m^E = \sum_{i=1}^{2} x_i M_i (\rho^{-1} - \rho_i^{-1}) \qquad (1)$$

According to Figures 6 (a) and 6 (b), the excess molar volume values are negative for all temperatures over entire composition range, so the volume of the solution was contracted due to the interaction between the IL and their binary mixture of water or methanol and are significant. Furthermore, the Figures 6 (a) and 6 (b) indicates, the greater negative values of excess molar volume occurs when the temperatures increases. Higher temperature has been helpful to reduce the distance between unlike molecules, so the molecules are interacted more strongly. Moreover, Figures 6 (a) and (b) indicates the excess molar volume, minima occurs with water and methanol at x_1 = 0.3026 and at x_1 = 0.2034, respectively. The quasi-clathrates perhaps occurred in the mixture of an IL with organic components are reported in Wang et al.,[64,65] In this case, it may

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happen in our binary mixtures such as $[EPMpyr]^{+}[SAL]^{-}$ with methanol in the nearby $[EPMpyr]^{+}[SAL]^{-}$ at x_1 = 0.2034. Similar results were also investigated for binary mixtures of $[EPMpyr]^{+}[SAL]^{-}$ with water at x_1 = 0.3026. Figures 6 (a) and (b), the excess molar volume graph indicates that 0.2000 and 0.3000 mole fraction of IL has low values and all binary mixture have negative excess molar values. In addition, the value of V_m^E is fully based on the effect of the hydrogen bond, polarity and interstitial accommodation in entire compositions. The packing/filling effect of methanol or water molecules in the interstices of IL, ion-dipole interactions between water and methanol with the pyrrolidonium ring of IL, all contributes to the negative values of V_m^E. The excess molar volume V_m^E, decreases with increasing temperature for both binary systems. The Tables 2 and 3 show that the result of excess molar volume data summaries for binary mixtures of IL with methanol or water. The results suggest the presence of the competing effect. These competing effect could be used to better understand the partial molar volumes of corresponding mixtures at infinite dilution.

Tables 2 and 3, shows that the increasing concentration of IL results in decreases the intermolecular free length, L_f of binary mixture. The Figures 7 (a) and (b), and Tables 2 and 3 indicates that as the speed of sound increases as corresponding decrease in intermolecular free length and the intermolecular free length also increases with increasing temperature. Moreover intermolecular free length explains the greater distances between the surfaces of the two molecules, and this behavior leads to a corresponding decrease in the speed of sound.

Intermolecular free length (L_f) has been calculated from the Eq. (2)

$$L_f = k_j (k_s)^{1/2} \quad (2)$$

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where k_j is the Jacobson's constant and is a temperature dependent constant. Its value is (93.875 + 0.375T) 10^{-8}.

Isentropic compressibility (k_s) defined as Eq. (3)

$$k_s = \rho^{-1}u^{-2} \quad (3)$$

The deviation in isentropic compressibility (Δk_s) can be defined from the isentropic compressibility as illustrated in Eq. (4)

$$\Delta k_s = K_s - \sum_i^2 x_i k_{s,i} \quad (4)$$

This property is related to density and speed of sound by the Newton−Laplace equation: Generally, the speed of sound increases with an increase in mole fraction of the mixture but decreases with temperature. The molar fractions increase linearly with temperatures and decays the isentropic compressibility exponentially. This performance elucidated due to the isentropic compressibility has been well defined as the inverse of the product of the density and square of the speed of sound. The free space was decreased due to the interaction between the molecules in binary mixtures, and in this way contributing to the negative deviation in isentropic compressibility. Figures 8 (a) and (b) display, the negative value of deviation in isentropic compressibility occurs over the entire composition of {$[EPMpyr]^+[SAL]^-$ + methanol or water} at all temperatures. The minimum value of deviation in isentropic compressibility of the binary mixtures was - 40.11 x $10^8 \mathrm{Pa}^{-1}$ and -13.16 x $10^8 \mathrm{Pa}^{-1}$ with methanol and water are occurring at

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x_1 = 0.2034 and 0.1031, respectively. The ideal mixtures are more compressible then these mixtures due to the performance of deviation in isentropic compressibility. In this case, the unlike molecules approach closely and a stronger interaction between methanol or water with $[EPMpyr]^+[SAL]^-$ mixtures that lead to a decrease in compressibility. Normally, the deviation in isentropic compressibility values decreases with an increasing temperature for both binary systems at a several composition of $[EPMpyr]^+[SAL]^-$ as shown in Figures 8 (a) and (b). The compressibility decreases because of the unlike molecules are contiguity, due to the mixture of components have strongly interacted.

4. Correlation of derived properties

The derived properties have been correlated by Redlich-Kister equation[66] as below in Eq. (5):

$$X = x_1 x_2 \sum_{i=1}^{k} A_j (1 - 2x_1)^{i-1} \quad (5)$$

where X is excess molar volumes (V_m^E) , deviation in isentropic compressibility (Δk_s) and intermolecular free length, L_f . The least-square method has been used to determine the fitting parameters A_j values. Table 4 shows that the summarized results.

Composed with the corresponding standard deviations, σ, For the correlation as investigated using Eq. (6).

$$\sigma(X) = \sum_{i=1}^{n} \left[\frac{X_{expt} - X_{calc}}{(N-K)}\right]^{1/2} \quad (6)$$

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where $N$the number of is experimental points and k is the number of coefficients used in the Redlich–Kister equation. The values of V_m^E and Δk_s, as well as the plots of the Redlich–Kister model. Both binary systems of the standard deviations indicate very low values for both excess molar volumes and deviations in isentropic compressibility at all inspected temperatures.

5. Conclusions

In this study, the synthesis, characterization and investigation of important physical parameters of novel pure ionic liquid and their binary mixtures with water or methanol at (293.15 to 313.15) K in steps of 5 K under atmospheric pressure are presented. The physical parameters such as density and speed of sound for pure ionic liquid and their binary mixtures of {$[EPMpyr]^+[SAL]^-$ with methanol or water} were measured. The excess molar volume, V_m^E isentropic compressibility, k_s, deviation in isentropic compressibility, Δk_s and intermolecular free length, L_f were calculated and discussed. The above calculated parameters of excess molar volume, V_m^E and deviation in isentropic compressibility's, Δk_s shows negative values. These indicate strong intermolecular interactions occurring between unlike molecules; the compacting effect is a major role in these binary mixtures because of the strong interaction between pyrrolidonium cation and salicylate anion. The binary combination of IL mixtures has strong attractive interaction, readjustments in structure and packing effect due to the great negative values of Δk_s. The methanol with $[EPMpyr]^+[SAL]^-$ has more effective packing arrangement than water due to their more negative values in Δk_s and V_m^E as well as ion-dipole interactions between methanol and $[EPMpyr]^+[SAL]^-$. The salicylate anion has carboxylate as well as hydroxyl groups so it can

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form hydrogen bonds easily. Acceptable correlations for the excess thermodynamic parameters occurred by fitting with the Redlich-Kister polynomial equation.

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Table 1

Component	Supplier	% Mass purity	T/K	ρ/g.cm^{-3} Exp.	ρ/g.cm^{-3} Lit.
Water			293.15	0.9982	0.9998 [54]
					0.9996
			298.15	0.9971	
			303.15	0.9957	0.9974 [54]
			308.15	0.9941	0.9940 [54]
			313.15	0.9922	
Methanol			29315	0.7914	0.7915 [55]
					0.7912 [56]
					0.7910 [58]
					0.7912 [57]
			298.15	0.7867	0.7868 [55]
					0.7866 [56]
					0.7865 [57]
					0.7866 [59]
	Fluka	≥ 99.0	303.15	0.7820	0.7821 [55]
					0.7818 [56]
					0.7819 [58]
					0.7817 [57]
					0.7819 [59]
			308.15	0.7772	0.7770 [57]
					0.7772 [59]
			313.15	0.7724	0.7726 [55]
					0.7726 [56]
					0.7720 [58]
					0.7722 [57]
					0.7727 [61]
$[EPMPYR]^{+}$ $[SAL]^{-}$			293.15	1.0685	-
			298.15	1.0637	-
			303.15	1.0590	-

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		308.15	1.0542	-
		313.15	1.0495	-

Table 2

Density,(ρ) speed of sound,(u) excess molar volume,(V_m^E) isentropic compressibility (K_s), intermolecular free length,(L_f) and deviation in isentropic compressibility, (Δk_s) with mole fraction of N-(2',3'-epoxypropyl)-N-methyl-2-oxopyrrolidinium salicylate in the binary mixture of {$[EPMpyr]^+[SAL]^-$ (1) + methanol (2)} at (293.15 to 313.15) K and at pressure P = 0.1 MPa.

x_1	$\rho/g.cm^{-3}$	u/ms^{-1}	V_m^E $/cm^3mol^{-1}$	k_s $/10^8Pa^{-1}$	$L_f/10^7m$	Δk_s $/10^8Pa^{-1}$
			T = 293.15 K			
0.0000	0.7914	1119.30	0.000	100.86	2.047	0.00
0.0360	0.8769	1210.33	-0.488	77.84	1.798	-20.50
0.0724	0.9377	1278.51	-0.768	65.24	1.646	-30.56
0.1023	0.9756	1320.95	-0.910	58.74	1.562	-34.96
0.2034	1.0587	1424.91	-1.106	46.52	1.390	-40.11
0.3189	1.1115	1493.34	-1.103	40.34	1.294	-38.21
0.4018	1.1360	1525.85	-1.023	37.81	1.253	-34.95
0.5117	1.1592	1556.94	-0.861	35.59	1.216	-29.48
0.5978	1.1726	1575.19	-0.692	34.37	1.195	-24.67
0.7189	1.1874	1596.43	-0.510	33.04	1.171	-17.53
0.8099	1.1962	1609.51	-0.370	32.27	1.158	-11.94
0.9201	1.2046	1624.27	-0.151	31.47	1.143	-5.04
1.0000	1.2098	1635.20	0.000	30.91	1.133	0.00
			T = 298.15 K			
0.0000	0.7867	1103.13	0.000	104.46	2.102	0.00
0.0360	0.8723	1194.56	-0.502	80.34	1.844	-21.51
0.0724	0.9332	1262.85	-0.791	67.19	1.686	-32.02
0.1023	0.9711	1305.29	-0.936	60.44	1.599	-36.60
0.2034	1.0543	1409.15	-1.137	47.77	1.422	-41.93
0.3189	1.1071	1477.40	-1.134	41.38	1.323	-39.93
0.4018	1.1317	1509.76	-1.051	38.77	1.281	-36.53
0.5117	1.1549	1540.54	-0.885	36.48	1.242	-30.83
0.5978	1.1682	1558.46	-0.712	35.24	1.221	-25.82
0.7189	1.1831	1579.01	-0.530	33.90	1.198	-18.37
0.8099	1.1919	1590.91	-0.391	33.14	1.184	-12.53
0.9201	1.2003	1604.17	-0.160	32.37	1.170	-5.30

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1.0000	1.2055	1613.45	0.000	31.87	1.161	0.00
			T = 303.15 K			
0.0000	0.7820	1086.78	0.000	108.27	2.160	0.00
0.0360	0.8676	1178.57	-0.515	82.97	1.891	-22.58
0.0724	0.9286	1247.02	-0.811	69.25	1.727	-33.56
0.1023	0.9666	1289.44	-0.959	62.22	1.637	-38.33
0.2034	1.0499	1393.18	-1.164	49.07	1.454	-43.84
0.3189	1.1027	1461.27	-1.158	42.47	1.353	-41.72
0.4018	1.1273	1493.52	-1.072	39.77	1.309	-38.16
0.5117	1.1506	1524.09	-0.900	37.42	1.270	-32.22
0.5978	1.1639	1541.79	-0.721	36.14	1.248	-26.99
0.7189	1.1789	1561.88	-0.534	34.77	1.224	-19.21
0.8099	1.1877	1573.56	-0.395	34.00	1.210	-13.11
0.9201	1.1961	1585.61	-0.162	33.25	1.197	-5.54
1.0000	1.2013	1593.94	0.000	32.76	1.188	0.00
			T = 308.15 K			
0.0000	0.7772	1070.51	0.000	112.28	2.292	0.00
0.0360	0.8630	1162.62	-0.533	85.73	1.939	-23.72
0.0724	0.9240	1231.23	-0.837	71.39	1.770	-35.20
0.1023	0.9620	1273.61	-0.987	64.08	1.731	-40.15
0.2034	1.0454	1377.25	-1.194	50.43	1.536	-45.85
0.3189	1.0984	1445.22	-1.186	43.59	1.428	-43.61
0.4018	1.1230	1477.38	-1.097	40.80	1.382	-39.88
0.5117	1.1463	1507.82	-0.919	38.37	1.340	-33.66
0.5978	1.1596	1525.37	-0.734	37.06	1.317	-28.19
0.7189	1.1746	1545.16	-0.539	35.66	1.292	-20.07
0.8099	1.1834	1556.54	-0.397	34.88	1.277	-13.70
0.9201	1.1919	1568.38	-0.162	34.11	1.263	-5.80
1.0000	1.1971	1576.18	0.000	33.62	1.254	0.00
			T = 313.15 K			
0.0000	0.7724	1054.36	0.000	116.46	2.301	0.00
0.0360	0.8583	1146.78	-0.550	88.59	2.036	-24.95
0.0724	0.9194	1215.52	-0.861	73.61	1.856	-36.99
0.1023	0.9575	1257.88	-1.015	66.01	1.732	-42.17
0.2034	1.0410	1361.41	-1.225	51.83	1.535	-48.16
0.3189	1.0940	1429.29	-1.214	44.74	1.426	-45.90
0.4018	1.1186	1461.37	-1.122	41.86	1.379	-42.07
0.5117	1.1419	1491.75	-0.938	39.35	1.337	-35.67
0.5978	1.1553	1509.20	-0.749	38.00	1.314	-30.05
0.7189	1.1703	1528.79	-0.546	36.56	1.289	-21.69
0.8099	1.1792	1539.98	-0.399	35.58	1.272	-15.30
0.9201	1.1877	1551.55	-0.160	34.98	1.261	-6.99
1.0000	1.1929	1559.03	0.000	35.49	1.270	0.00

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Table 3

Density,(ρ) speed of sound,(u) excess molar volume,(V_m^E) isentropic compressibility (k_s), intermolecular free length,(L_f) and deviation in isentropic compressibility (Δk_s) of N-(2',3'-epoxypropyl)-N-methyl-2-oxopyrrolidinium salicylate in the binary liquid mixture of {$[EPMpyr]^+[SAL]^-$ (1) + water (2) } from at (293.15 to 313.15) and at pressure P = 0.1 MPa.

x_1	$\rho/g.cm^{-3}$	u/ms^{-1}	V_m^E $/cm^3mol^{-1}$	k_s $/10^8XPa^{-1}$	$L_f/10^7m$	Δk_s $/10^8XPa^{-1}$
			T = 293.15 K			
0.0000	0.9982	1482.63	0.000	45.57	1.376	0.00
0.0332	1.0633	1620.29	0.034	35.82	1.220	-9.27
0.0630	1.1115	1662.43	-0.384	32.55	1.163	-12.10
0.1031	1.1432	1682.43	-0.616	30.90	1.133	-13.16
0.2067	1.1772	1680.87	-0.828	30.07	1.118	-12.49
0.3026	1.1897	1668.20	-0.849	30.20	1.120	-10.96
0.4170	1.1970	1657.88	-0.747	30.39	1.123	-9.10
0.5059	1.2014	1650.02	-0.756	30.57	1.127	-7.63
0.6171	1.2040	1645.98	-0.582	30.66	1.128	-5.92
0.7118	1.2060	1639.62	-0.493	30.84	1.132	-4.36
0.8123	1.2070	1635.67	-0.291	30.97	1.134	-2.77
0.9029	1.2073	1635.80	-0.039	30.95	1.134	-1.46
1.0000	1.2089	1633.54	0.000	31.00	1.135	0.00
			T = 298.15 K			
0.0000	0.9971	1496.81	0.000	44.76	1.376	0.00
0.0332	1.0607	1620.01	0.047	35.92	1.233	-8.42
0.0630	1.1082	1656.29	-0.365	32.89	1.180	-11.06
0.1031	1.1394	1671.86	-0.589	31.40	1.153	-12.04
0.2067	1.1730	1666.47	-0.797	30.70	1.140	-11.41
0.3026	1.1855	1652.74	-0.819	30.88	1.143	-10.00
0.4170	1.1927	1641.78	-0.720	31.11	1.147	-8.31
0.5059	1.1972	1633.35	-0.736	31.31	1.151	-6.97
0.6171	1.1997	1628.78	-0.570	31.42	1.153	-5.43
0.7118	1.2017	1621.52	-0.490	31.65	1.157	-3.99
0.8123	1.2027	1616.27	-0.291	31.83	1.160	-2.52
0.9029	1.2031	1616.32	-0.042	31.82	1.160	-1.37

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1.0000	1.2046	1612.10	0.000	31.94	1.162	0.00
			T = 303.15 K			
0.0000	0.9957	1509.18	0.000	44.10	1.378	0.00
0.0332	1.0581	1620.83	0.060	35.97	1.245	-7.75
0.0630	1.1048	1649.66	-0.344	33.26	1.197	-10.13
0.1031	1.1355	1660.90	-0.562	31.92	1.173	-11.02
0.2067	1.1688	1651.82	-0.763	31.36	1.162	-10.42
0.3026	1.1812	1637.14	-0.785	31.59	1.167	-9.11
0.4170	1.1884	1625.65	-0.687	31.84	1.171	-7.57
0.5059	1.1929	1616.81	-0.706	32.07	1.175	-6.34
0.6171	1.1954	1611.80	-0.544	32.20	1.178	-4.95
0.7118	1.1975	1604.08	-0.472	32.45	1.182	-3.63
0.8123	1.1985	1598.17	-0.279	32.67	1.186	-2.29
0.9029	1.1989	1598.15	-0.031	32.66	1.186	-1.28
1.0000	1.2003	1592.76	0.000	32.84	1.189	0.00
			T = 308.15 K			
0.0000	0.9941	1519.86	0.000	43.55	1.427	0.00
0.0332	1.0553	1619.78	0.068	36.12	1.259	-7.11
0.0630	1.1014	1642.56	-0.326	33.65	1.215	-9.28
0.1031	1.1316	1649.65	-0.536	32.76	1.238	-9.77
0.2067	1.1645	1637.15	-0.730	32.04	1.224	-9.48
0.3026	1.1769	1621.61	-0.752	32.31	1.229	-8.26
0.4170	1.1841	1609.66	-0.655	32.59	1.235	-6.85
0.5059	1.1885	1600.53	-0.677	32.85	1.240	-5.73
0.6171	1.1911	1595.26	-0.518	32.99	1.242	-4.49
0.7118	1.1932	1587.27	-0.451	33.26	1.247	-3.28
0.8123	1.1943	1580.93	-0.263	33.50	1.252	-2.06
0.9029	1.1946	1580.83	-0.019	33.50	1.252	-1.17
1.0000	1.1961	1574.75	0.000	33.71	1.256	0.00
			T = 313.15 K			
0.0000	0.9922	1528.91	0.000	43.115	1.400	0.00
0.0332	1.0525	1617.85	0.077	36.299	1.303	-6.53
0.0630	1.0979	1634.94	-0.308	34.074	1.263	-8.50
0.1031	1.1277	1638.19	-0.511	33.042	1.225	-9.19
0.2067	1.1603	1622.49	-0.699	32.739	1.220	-8.61
0.3026	1.1726	1606.13	-0.720	33.058	1.226	-7.48
0.4170	1.1797	1593.86	-0.625	33.367	1.231	-6.19
0.5059	1.1842	1584.48	-0.649	33.635	1.236	-5.16
0.6171	1.1868	1578.98	-0.493	33.796	1.239	-4.05
0.7118	1.1889	1570.82	-0.429	34.088	1.245	-2.95
0.8123	1.1900	1564.25	-0.246	34.343	1.249	-1.84
0.9029	1.1903	1564.09	-0.003	34.341	1.249	-1.07
1.0000	1.1919	1557.61	0.000	34.581	1.254	0.00

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Table 4

Coefficients $A_{i,}$ and standard deviations, σ, obtained for the binary systems {[EPMPYR]$^+$[SAL]$^-$ (1) + water or methanol (2)} at different temperatures for the Redlich-Kister equation.

	T/K	A_0	A_1	A_2	A_3	A_4	σ
	{[EPMpyr]$^+$[SAL]$^-$ (1) + methanol (2)}						
V_m^E / (cm^3.mol^{-1})	293.15	-3.513	-2.855	-1.889	-3.213	-3.655	0.02
	298.15	-3.608	-2.903	-2.060	-3.284	-3.688	0.03
	303.15	-3.669	-3.013	-2.098	-3.335	-3.386	0.03
	308.15	-3.749	-3.118	-2.056	-3.479	-4.109	0.03
	313.15	-3.831	-3.207	-2.067	-3.662	-4.260	0.03
L_f	293.15	5.331	-0.980	-6.602	7.909	39.533	0.9
	298.15	5.449	-1.008	-6.782	8.116	40.536	0.9
	303.15	5.571	-1.039	-6.976	8.344	41.567	0.9
	308.15	5.855	-0.963	-6.772	8.302	42.519	0.9
	313.15	5.886	-1.175	-7.752	9.912	44.780	0.9
$\Delta\kappa_S$ / (10^8×Pa^{-1})	293.15	-121.99	-88.49	-47.92	-175.78	-201.65	1.0
	298.15	-127.59	-91.93	-49.65	-184.93	-212.81	1.0
	303.15	-133.35	-95.64	-51.45	-194.88	-224.49	1.1
	308.15	-139.35	-99.81	-53.13	-204.97	-237.44	1.2
	313.15	-147.59	-102.88	-56.59	-207.97	-258.37	1.3
	{[EPMpyr]$^+$[SAL]$^-$ (1) + water (2)}						
V_m^E / (cm3.mol^{-1})	293.15	-2.770	-1.762	-4.611	-2.374	6.514	0.08

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	298.15	-2.685	-1.625	-4.639	-2.283	6.674	0.09
	303.15	-2.561	-1.544	-4.665	-2.226	6.917	0.09
	308.15	-2.444	-1.478	-4.586	-2.192	6.991	0.09
	313.15	-2.330	-1.416	-4.521	-2.193	7.139	0.09
L_f	293.15	4.823	-1.054	-4.048	3.691	28.909	0.60
	298.15	4.922	-1.060	-4.029	3.638	29.277	0.70
	303.15	5.022	-1.062	-4.007	3.591	29.632	0.70
	308.15	5.272	-1.011	-3.669	3.332	29.957	0.70
	313.15	5.283	-1.124	-4.230	3.728	31.158	0.70
$\Delta\kappa_S$ / $(10^8 \times Pa^{-1})$	293.15	-32.53	-19.15	9.05	-102.12	-151.96	0.80
	298.15	-29.73	-17.46	8.36	-93.11	-139.15	0.70
	303.15	-27.09	-15.87	8.22	-85.31	-128.94	0.70
	308.15	-24.57	-14.48	8.12	-76.83	-117.74	0.70
	313.15	-22.17	-12.81	7.86	-72.15	-110.40	0.60

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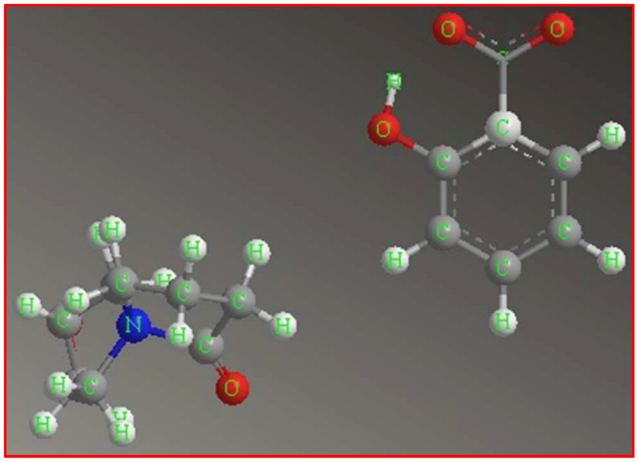

Figure 1. Structure of the ionic liquid $[EPMpyr]^+[SAL]^-$.

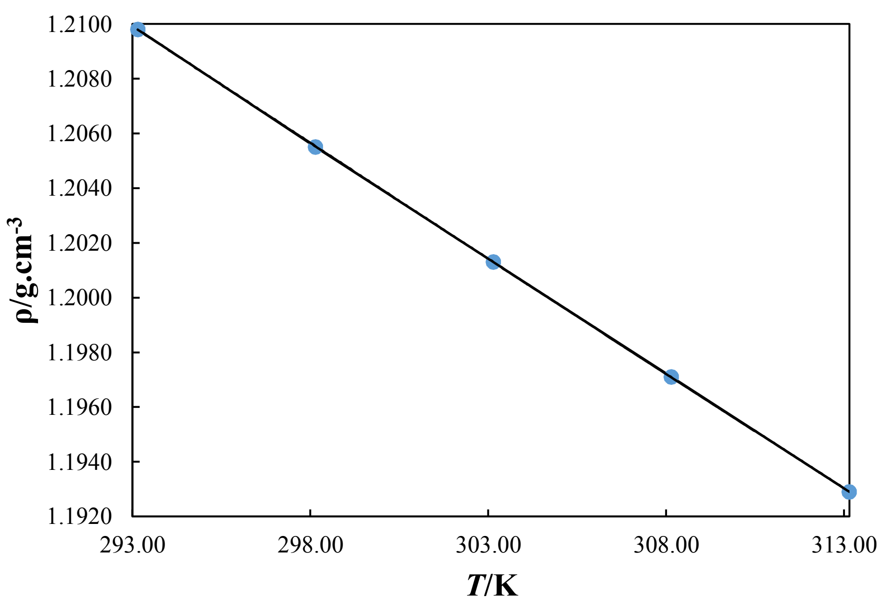

Figure 2. Density, *ρ*, of $[EPMpyr]^+[SAL]^-$ at temperatures from (293.15 to 313.15) K. The solid line represents the smoothness of these data.

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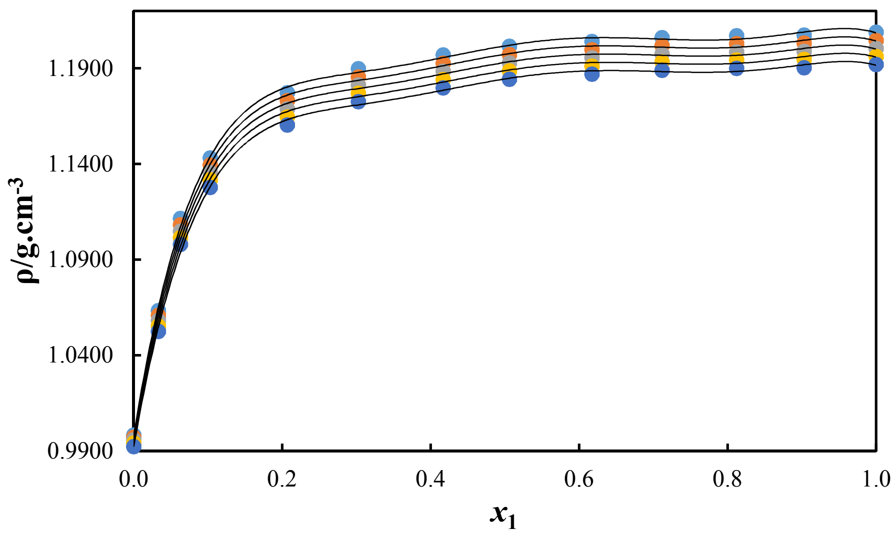
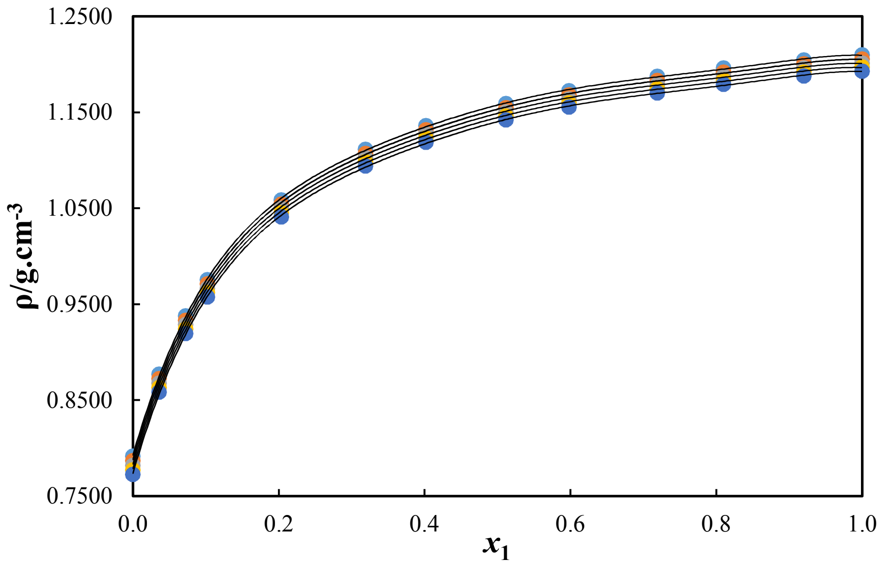


(a)



(b)

Figure 3. Density, ρ, for the mixture of (a) {$[EPMpyr]^+[SAL]^-$ (1) + water (2)} and (b) {$[EPMpyr]^+[SAL]^-$ (1) + methanol (2)} as function of the composition expressed in the mole fraction of {$[EPMpyr]^+[SAL]^-$ at T = 293.15 K (●), T = 298.15 K (●), T = 303.15 K (●), T = 308.15 K (●) and T = 313.15 K (●). The solid line represents the smoothness of these data.

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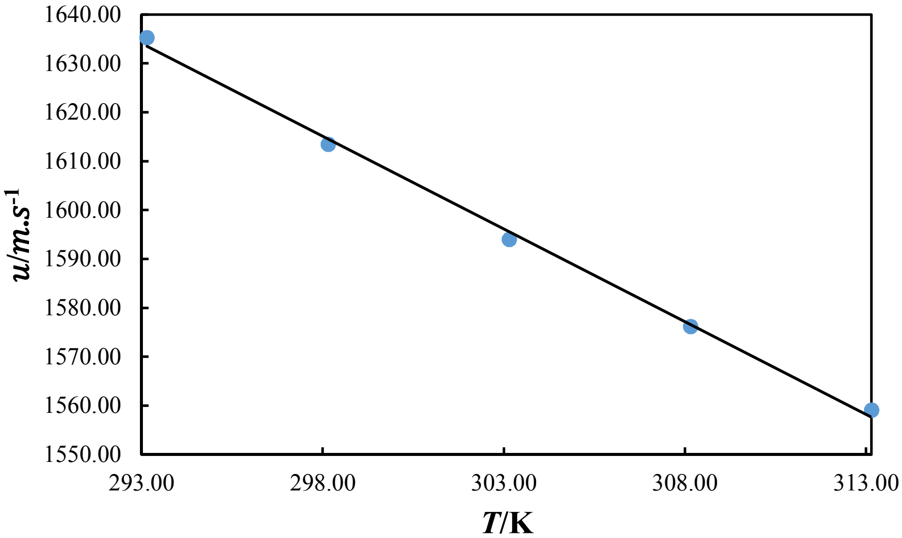
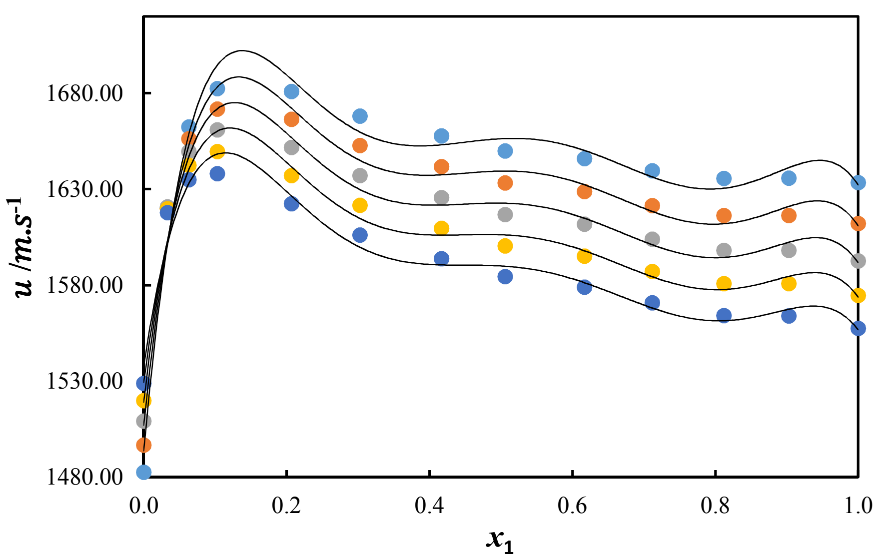


Figure 4. Speed of sound velocity of {[EPMpyr]$^+$[SAL]$^-$ at temperatures from (293.15 to 313.15) K. The solid line represents the smoothness of these data.



(a)

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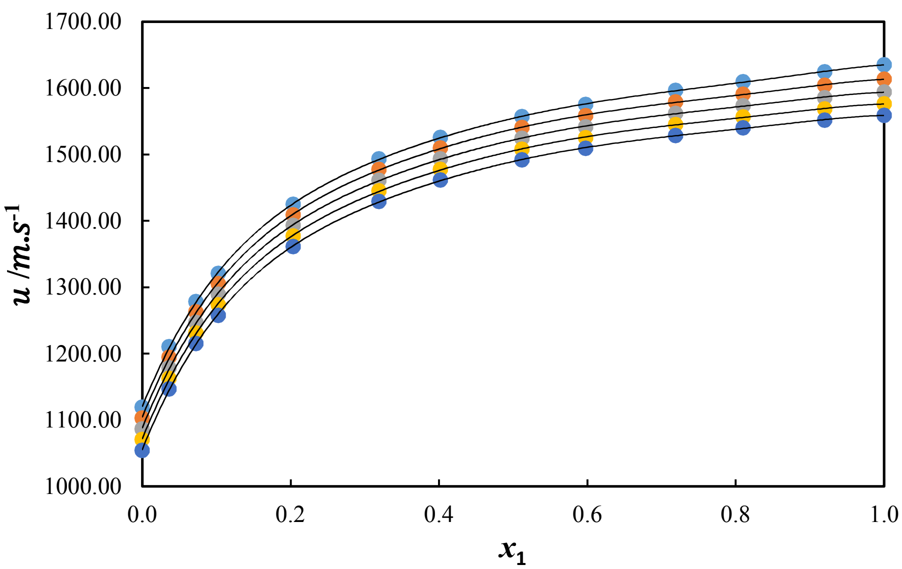
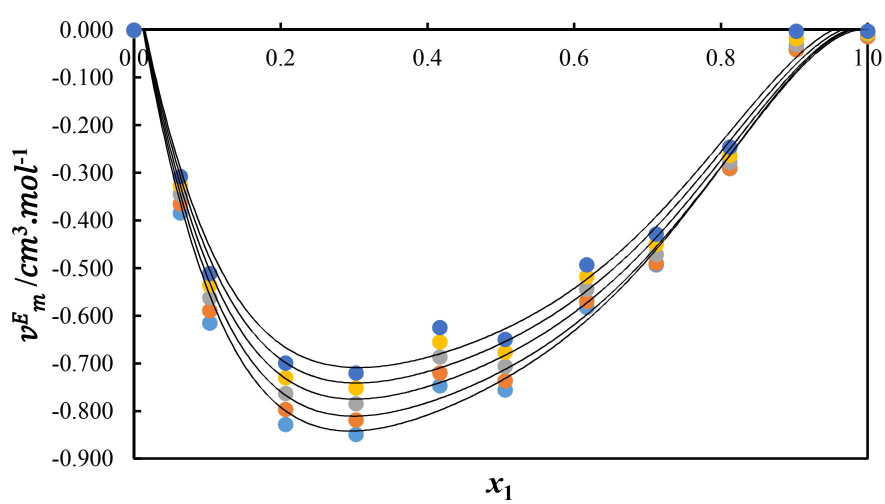


(b)

Figure 5. Speed of sound, *u*, for the mixture of (a) {$[EPMpyr]^+[SAL]^-$ (1) + water (2)} and (b) {$[EPMpyr]^+[SAL]^-$ (1) + methanol (2)} as function of the composition expressed in the mole fraction of {$[EPMpyr]^+[Cl]^-$ at T = 293.15 K (●), T = 298.15 K (●), T = 303.15 K (●), T = 308.15 K (●) and T = 313.15 K (●).The solid line represents the smoothness of these data.



(a)

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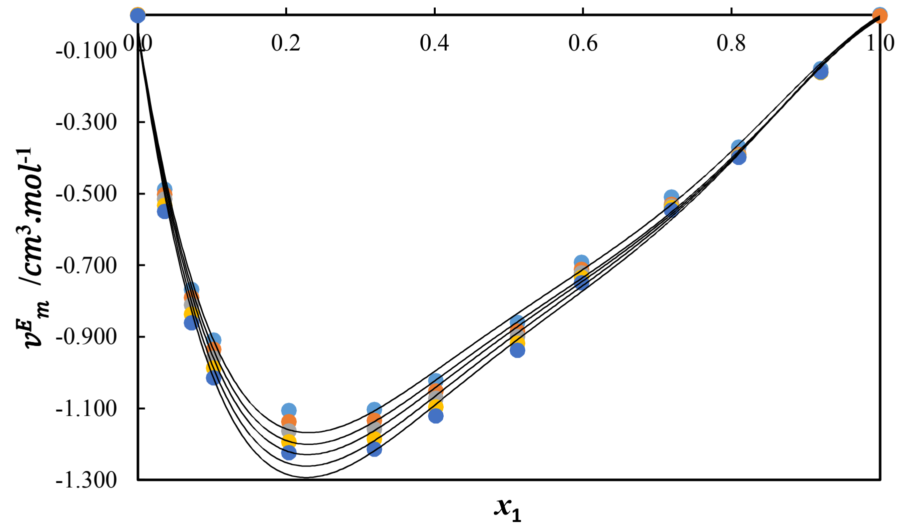
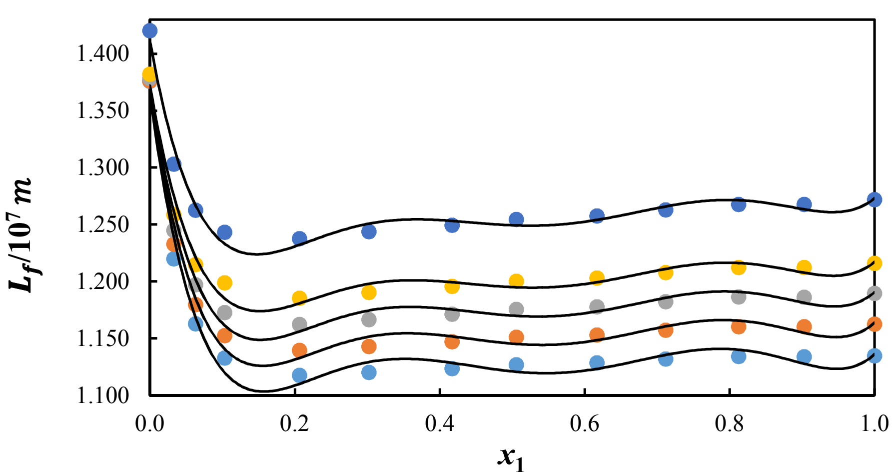


(b)

Figure 6. Excess molar volumes, V^E_m, for the mixture of (a) {$[EPMpyr]^+[SAL]^-$ (1) + water (2)} and (b) {$[EPMpyr]^+[SAL]^-$ (1) + methanol (2)} as function of the composition expressed in the mole fraction of {$[EPMpyr]^+[SAL]^-$ at T = 293.15 K (●), T = 298.15 K (●), T = 303.15 K (●), T = 308.15 K (●) and T = 313.15 K (●).The solid lines were generated using Redlich-Kister curve-fitting.



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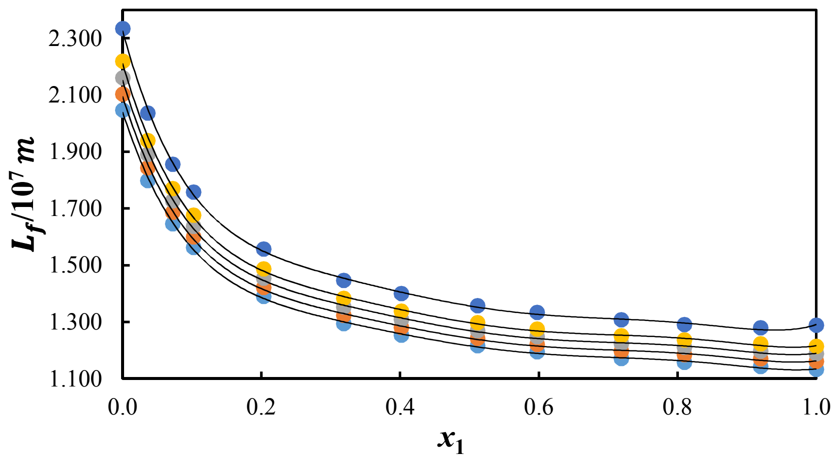
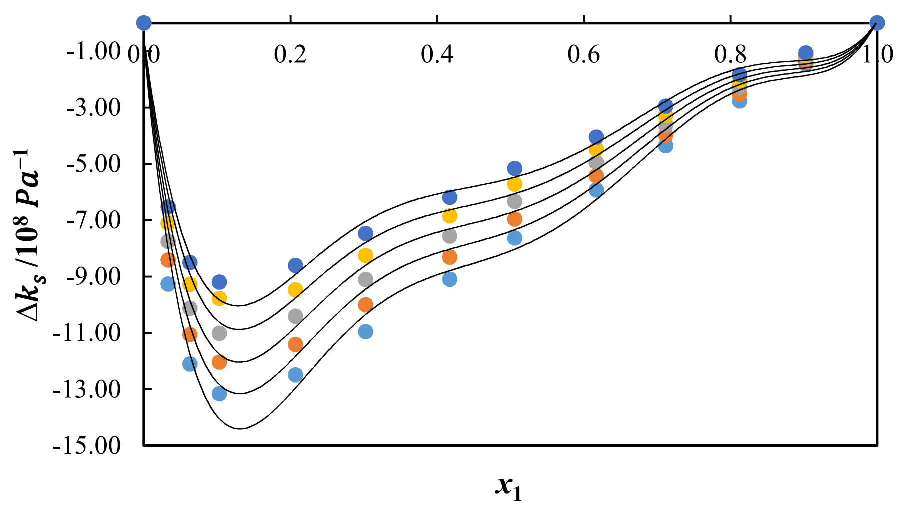
(a)



(b)

Figure 7. Intermolecular free length, L_f, for the mixture of (a) {$[EPMpyr]^+[SAL]^-$ (1) + water (2)} and (b) {$[EPMpyr]^+[SAL]^-$ (1) + methanol (2)} as function of the composition expressed in the mole fraction of {$[EPMpyr]^+[SAL]^-$ at T = 293.15 K (●), T = 298.15 K (●), T = 303.15 K (●), T = 308.15 K (●) and T = 313.15 K (●).The solid lines were generated using Redlich-Kister curve-fitting.



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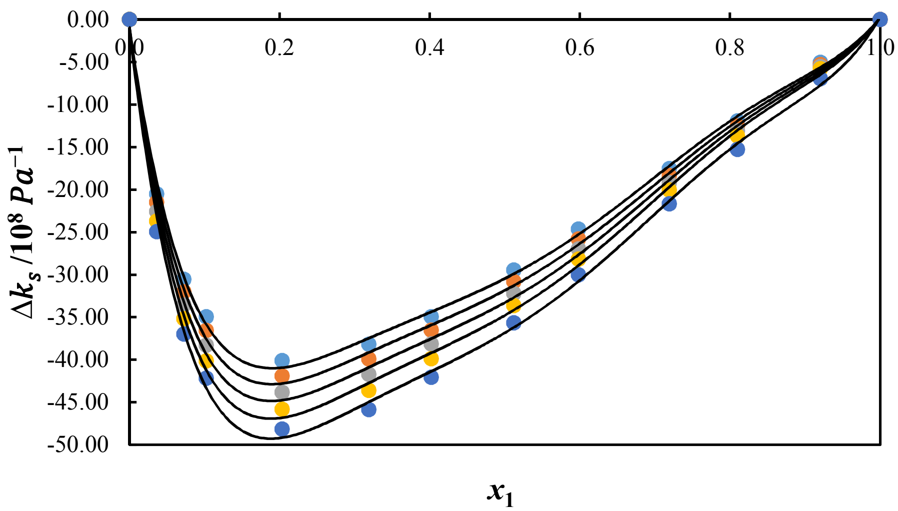
(a)



(b)

Figure 8. Deviation of isentropic compressibility, $\Delta\kappa_S$, for the mixture of (a) {[EPMpyr]$^+$[SAL]$^-$ (1) + water (2)} and (b) {[EPMpyr]$^+$[SAL]$^-$ (1) + methanol (2)} as function of the composition expressed in the mole fraction of {[EPMpyr]$^+$[SAL]$^-$at T = 293.15 K (●), T = 298.15 K (●), T = 303.15 K (●), T = 308.15 K (●) and T = 313.15 K (●). The solid lines were generated using Redlich-Kister curve-fitting.

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Synthesis, characterization of 2′, 3′-epoxy propyl - N-methyl-2- oxopyrrolidinium salicylate ionic liquid and study of its interaction with water or methanol

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Graphical Abstract

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