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Estimation of Thermophysical and thermoacoustical properties of aqueous solutions of 1-butyl-3-ethyl imidazolium trifluoro sulfonate ([C₄mim][CF₃SO₃]) from ultrasonic velocity and density data

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Abstract

Some of the most active academic and scientific research fronts of the last decade are on ionic liquids and their behaviour for industrial purposes. The present work deals with calculation of some useful thermodynamic and thermoacoustical properties of binary mixtures of [C₄mim][CF₃SO₃] + water from speed of sound and density data. Speed of sound and density data were taken from literature. Several relevant properties like thermal expansivity, isothermal compressibility, internal pressure, energy of vaporization and solubility parameter have been evaluated. Thermoacoustical properties have been ascertained from speed of propagation of ultrasound waves in the mixture. Effect of composition and solution temperature has been observed to understand the nature of various interactions involved.

Keywords: Ionic liquids; thermal expansivity; internal pressure; thermophysical

1. Introduction

Recently, ecological concern has rekindled industrial and academic interest in replacing conventional organic solvents due to their volatility, flammability and toxic effects on human health and the environment [1, 2]. Therefore, significant attention is being given to ionic liquids as substitutes [3-5]. Ionic liquids are molten salts, exclusively made up of organic or inorganic anions (mainly based on imidazolium, pyrrolidinium, pyridinium, phosphonium, aluminium and sulfonium ions), are unconventional solvents, gaining widespread recognition and importance from scientific community due to their extraordinary physico-chemical properties like negligible vapour pressure, lesser environmental toxicity, excellent chemical and thermal stability, high latent heat capacity, structural diversity, reusability and recyclability [6-9]. Presently, ionic liquids are being used as greener and environmentally safer reaction media in separation and purification technologies [10-13] removal of hazardous pollutants from liquids and gaseous streams [14-17], electrochemistry [18-20], green synthesis [21-24], green catalysis [25-27], desulphurization of gasoline [28-32] etc. Therefore, keeping in view the sustainable and industrial aspect of ionic liquids, some important thermodynamic properties have been estimated by empirical relationships. Accuracy and precision in density and ultrasonic velocity measurements allow one to calculate many interesting thermodynamic and thermoacoustical properties of industrial and environmental concerns with a high degree of reliability [33-36]. These parameters have great applications in designing different theoretical models of fluid flow, mixing, agitation, heat and mass transfer, wastewater treatment, desalination etc. In addition, these properties provide useful informations about the nature of intermolecular interactions existing in fluids [37-40]. In the present work a number of thermodynamic parameters like thermal expansion coefficient, isothermal compressibility, internal pressure, energy of vaporization, solubility parameter, non-linearity parameter and acoustic impedance, have been evaluated from density and ultrasonic velocity data. Density and ultrasonic velocity data have been taken from literature [41] (not reported in the manuscript). The main aim of this work is to thermophysically characterize the undertaken ionic liquid, [C₄mim][CF₃SO₃] and study its thermodynamic behavior in aqueous solutions which can facilitate its future applications and development.

2. Theoretical formulation: Variation in density and ultrasonic velocity of the mixture as a function of temperature and concentration has been used to calculate different properties of the system under investigation using well known equations.

Thermal expansion coefficient quantifies the extent to which the volume of a fluid changes with the temperature has been computed using following relationship

$$\alpha = \frac{75.6 \times 10^{-3}}{T^{1/9} \cdot u^{1/2} \cdot \rho^{1/3}} \quad 1$$

Where T is temperature in K, u is speed of sound ($\text{m}\cdot\text{sec}^{-1}$) and ρ is the density ($\text{g}\cdot\text{cm}^{-3}$)

Isothermal compressibility (β_T) was estimated from the following expression

$$\beta_T = \frac{1.71 \times 10^{-3}}{T^{4/9} \cdot u^2 \cdot \rho^{4/3}} \quad 2$$

Where u is speed of sound ($\text{m}\cdot\text{sec}^{-1}$), ρ is the density ($\text{g}\cdot\text{cm}^{-3}$) and T is the temperature in K.

Coefficient of thermal expansion (α) and isothermal compressibility (β_T) values were utilized to calculate internal pressure (P_{int}) from the relationship

$$P_{int} = \frac{\alpha T}{\beta_T} \quad 3$$

P_{int} values were applied to estimate energy of vaporization (ΔE_V) and solubility parameter (δ) using the relationships

$$\Delta E_V = P_{int} \cdot V_m \quad 4$$

$$\delta = (P_{int})^{1/2} \quad 5$$

Where V_m is the molar volume of the mixture and has been derived from the expression

$$V_m = \frac{\sum_{i=1}^N x_i m_i}{\rho_{mix}} \quad 6$$

Two acoustical properties viz. parameter of non-linearity (B/A) and acoustic impedance (z) (Eq.9) were deduced from sound velocity data. B/A parameter has been evaluated from Ballou

^[42] (eq.7) and Hartmann ^[43] (eq.8) relationship

$$\frac{B}{A} = -0.5 + \left(\frac{1.2 \times 10^4}{u} \right) \quad 7$$

$$\frac{B}{A} = 2 + \left(\frac{0.98 \times 10^4}{u} \right) \quad 8$$

$$z = \rho u \quad 9$$

Where $u = \text{m}\cdot\text{sec}^{-1}$, $\rho = \text{g}\cdot\text{cm}^{-3}$ for Eqs.(7) and (8) and $\rho = \text{kg}\cdot\text{m}^{-3}$ for Eq.(9)

3. Results and discussion: Literature data of density and speed of sound have been employed to evaluate different thermodynamic properties using well defined relations. Thermal expansion coefficient and isothermal compressibility have been computed from Eqs. (1) and (2) respectively. Internal pressure has been calculated from α and β_T values (Eq.3). P_{int} values were used to estimate Energy of vaporization (Eq.4) and solubility parameter (Eq.6). Ballou (Eq.7) and Hartmann (Eq.8) relationship have been used to compute parameter of non-linearity. Another important acoustical property, acoustic impedance has been calculated from Eq.9. Table 1 enlists the values of α , β_T , P_{int} , ΔE_V , V_m and δ . Thermoacoustical properties (B/A and z parameter) are recorded in Table 2. Temperature and composition dependence of thermal expansivities have been plotted in Fig.2. A dramatic decrease in α values was observed with increasing molalities of [C₄mim][CF₃SO₃]. This decrease may be explained in terms of H-bonding interaction between SO₃⁻ group of aforesaid ionic liquid and H⁺ ions of water. At higher molalities, more SO₃⁻ groups are available to interact with H⁺ ions, leading to volume contraction. Electrostatic interaction between [C₄mim]⁺ cation of IL and OH⁻ terminal of water may be assigned as another contributor. Due to this, the values of isothermal compressibilities are decreasing (Fig.3) with increasing temperature throughout the concentration range of [C₄mim][CF₃SO₃]. Moreover, it is common behavior for imidazolium based ionic liquids, strengthening the fact that ionic liquids do not expand appreciably with the rise in temperature ^[44]. Many workers have reported this kind of unusual volume expansion in case of imidazolium based ILs ^[45-48]. Gautam *et al.* ^[49] reported H-bonding interaction in case of aqueous solutions containing an anionic surfactant, Sodiumdodecylbenzenesulphonate. Nanda *et al.* ^[50] also observed this kind of strong ion-solvent interaction through an unprecedented decrease in α and β_T values with rising temperature and concentration of solute in the mixture. These supporting informations gave us additional confidence to present our results. As usual, the internal pressure values are increasing with increasing concentration of [C₄mim][CF₃SO₃] and solution temperature (Fig.4). This increase in P_{int} values with the increase in molalities of solute component may be attributed to prevalence of columbic forces in ionic liquids ^[51]. At higher concentrations of undertaken ionic liquid, cohesive forces are dominant, which are also responsible for this linear increase in P_{int} values. Existence of van der Wall interactions is an important characteristic of molten salts which is able to describe the reason for the increase in values of ΔE_V (Fig.5) and δ (Fig.6). Thermoacoustical properties like non-linearity parameter and acoustic impedance enable to understand the complex mechanism and phenomenon of clustering, intermolecular spacing, harmonic distortion and acoustic scattering ^[52-55]. As far as our knowledge is concerned, which is based on literature survey, negligible information is available regarding the acoustical properties of these highly structured compounds. The values of B/A are also following the same decreasing trend as we obtained in case of thermal expansivity and isothermal compressibility, from both the relationships (Fig.7 & Fig.8). This decrease is associated with intermolecular free length. The free length decreases on the addition of [C₄mim][CF₃SO₃] due to electrostatic and van der Wall compression ^[56]. Increasing values of acoustic impedance with successive increase in concentration of [C₄mim][CF₃SO₃]

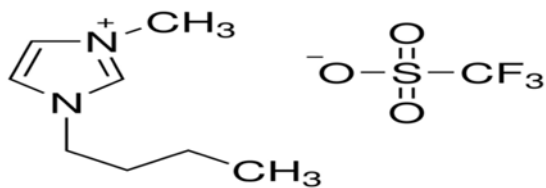
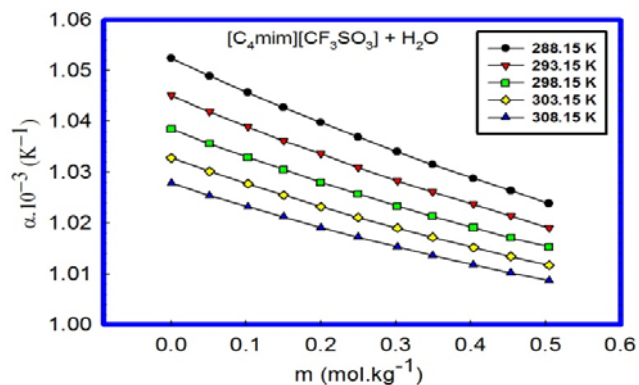
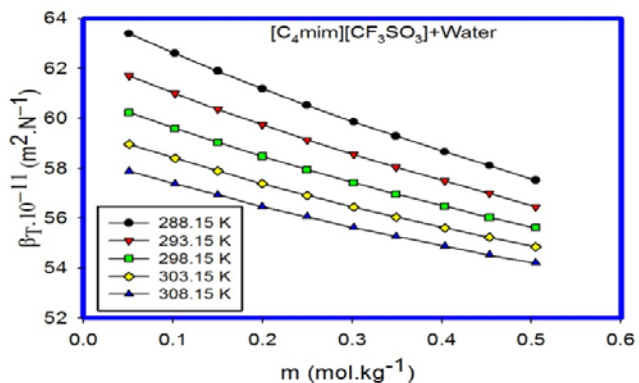
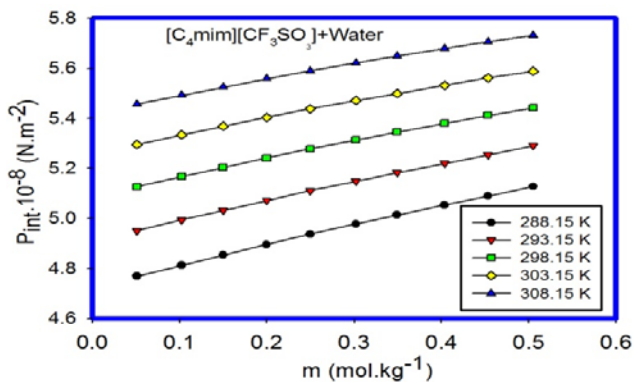
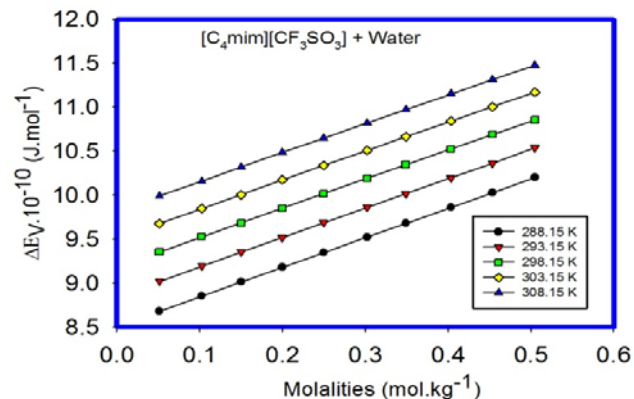
and rising solution temperature (Fig.9) supports the decreasing because of the strong structure making ability of interpretation and revealing that the degree of nonlinearity is SO_3^- ion.

Table 1: Thermal expansivities (α), isothermal compressibility (β_T), internal pressure (P_{int}), molar volume (V_m), energy of vaporization (ΔE_v), and solubility parameter (δ) of $[\text{C}_4\text{mim}][\text{CF}_3\text{SO}_3]+\text{H}_2\text{O}$ from the temperature range 288.15 K to 308.15 K at different concentrations.

m (mol.kg^{-1}) $[\text{C}_4\text{mim}][\text{CF}_3\text{SO}_3]$	$\alpha.10^{-3}$ (K^{-1})	$\beta_T.10^{-11}$ ($\text{m}^2.\text{N}^{-1}$)	$P_{int}.10^{-8}$ (N.m^{-2})	V_m ($\text{cm}^3.\text{mol}^{-1}$)	$\Delta E_v.10^{-10}$ (J.mol^{-1})	$\delta.10^{-4}$ (J.cm^{-3}) ^{1/2}
288.15 K						
0.0510	1.0489	63.39	4.769	18.2149	8.6839	6.9046
0.1023	1.0457	62.60	4.813	18.4016	8.8551	6.9369
0.1499	1.0427	61.89	4.854	18.5757	9.0154	6.9665
0.1996	1.0398	61.19	4.896	18.7587	9.1826	6.9964
0.2496	1.0369	60.52	4.937	18.9442	9.3509	7.0256
0.3019	1.0340	59.86	4.978	19.1386	9.5252	7.0547
0.3489	1.0316	59.29	5.014	19.3145	9.6819	7.0800
0.4034	1.0288	58.66	5.054	19.5197	9.8626	7.1081
0.4532	1.0264	58.11	5.090	19.7079	10.0281	7.1332
0.5045	1.0238	57.53	5.128	19.9024	10.2038	7.1602
293.15 K						
0.0510	1.0419	61.70	4.951	18.2320	9.0237	7.0351
0.1023	1.0389	60.99	4.994	18.4195	9.1957	7.0656
0.1499	1.0362	60.36	5.033	18.5945	9.3556	7.0932
0.1996	1.0336	59.75	5.071	18.7787	9.5207	7.1203
0.2496	1.0309	59.13	5.111	18.9647	9.6900	7.1480
0.3019	1.0283	58.55	5.149	19.1606	9.8637	7.1748
0.3489	1.0261	58.04	5.183	19.3376	10.0197	7.1982
0.4034	1.0237	57.49	5.220	19.5428	10.1988	7.2240
0.4532	1.0214	56.99	5.254	19.731	10.3646	7.2476
0.5045	1.0190	56.46	5.291	19.9270	10.5413	7.2731
298.15 K						
0.0510	1.0356	60.23	5.127	18.2536	9.3566	7.1595
0.1023	1.0329	59.59	5.168	18.4420	9.5281	7.1878
0.1499	1.0305	59.04	5.204	18.6175	9.6865	7.2130
0.1996	1.0280	58.48	5.242	18.8021	9.8529	7.2389
0.2496	1.0257	57.95	5.278	18.9890	10.0194	7.2638
0.3019	1.0233	57.42	5.314	19.1850	10.1926	7.2888
0.3489	1.0213	56.96	5.346	19.3620	10.3481	7.3106
0.4034	1.0191	56.48	5.380	19.5689	10.5262	7.3342
0.4532	1.0171	56.03	5.412	19.7580	10.6911	7.3559
0.5045	1.0153	55.63	5.442	19.9536	10.8557	7.3759
303.15 K						
0.0510	1.0302	58.97	5.296	18.2795	9.6791	7.2766
0.1023	1.0277	58.40	5.335	18.4684	9.8500	7.3030
0.1499	1.0255	57.90	5.369	18.6444	10.0079	7.3264
0.1996	1.0232	57.39	5.405	18.8298	10.1757	7.3511
0.2496	1.0211	56.92	5.438	19.0168	10.3396	7.3736
0.3019	1.0190	56.45	5.472	19.2137	10.5119	7.3966
0.3489	1.0172	56.05	5.500	19.3917	10.6666	7.4165
0.4034	1.0152	55.61	5.532	19.5992	10.8440	7.4383
0.4532	1.0134	55.23	5.562	19.7895	11.0079	7.4582
0.5045	1.0117	54.86	5.589	19.9856	11.1717	7.4765
308.15 K						
0.0510	1.0254	57.89	5.457	18.3093	9.9915	7.3872
0.1023	1.0232	57.38	5.493	18.4983	10.1619	7.4117
0.1499	1.0212	56.94	5.525	18.6749	10.3191	7.4334
0.1996	1.0191	56.47	5.559	18.8604	10.4863	7.4564
0.2496	1.0172	56.06	5.590	19.0482	10.6493	7.4770
0.3019	1.0153	55.63	5.622	19.2458	10.8208	7.4982
0.3489	1.0136	55.27	5.650	19.4241	10.9748	7.5166
0.4034	1.0118	54.88	5.680	19.6322	11.1512	7.5366
0.4532	1.0102	54.53	5.707	19.8230	11.3141	7.5548
0.5045	1.0087	54.21	5.732	20.0208	11.4770	7.5713

Table 2: The non-linearity parameter (B/A) and acoustic impedance (z) of [C₄mim][CF₃SO₃]+H₂O from the temperature range 288.15 K to 308.15 K at different concentrations.

m(mol.kg ⁻¹) [C ₄ mim][CF ₃ SO ₃]	B/A (Eq. 7)	B/A(Eq. 8)	z.10 ⁻³ (kg.s ⁻¹ .m ²)
288.15 K			
0.0510	7.648	8.654	1476.74
0.1023	7.616	8.628	1487.67
0.1499	7.587	8.605	1497.73
0.1996	7.559	8.581	1507.94
0.2496	7.531	8.559	1517.93
0.3019	7.504	8.537	1527.98
0.3489	7.481	8.518	1536.78
0.4034	7.457	8.498	1546.58
0.4532	7.435	8.480	1555.38
0.5045	7.410	8.460	1564.77
293.15 K			
0.0510	7.564	8.586	1555.38
0.1023	7.536	8.563	1500.95
0.1499	7.511	8.543	1510.33
0.1996	7.487	8.523	1519.64
0.2496	7.463	8.503	1529.13
0.3019	7.440	8.484	1538.41
0.3489	7.421	8.468	1546.54
0.4034	7.400	8.452	1555.62
0.4532	7.381	8.436	1563.90
0.5045	7.359	8.418	1572.78
298.15 K			
0.0510	7.491	8.526	1502.48
0.1023	7.467	8.506	1512.11
0.1499	7.446	8.489	1520.76
0.1996	7.425	8.472	1529.64
0.2496	7.405	8.456	1538.24
0.3019	7.385	8.440	1546.94
0.3489	7.369	8.426	1554.54
0.4034	7.352	8.412	1562.87
0.4532	7.335	8.399	1570.55
0.5045	7.322	8.388	1577.76
303.15 K			
0.0510	7.429	8.475	1512.10
0.1023	7.408	8.458	1521.10
0.1499	7.390	8.444	1529.17
0.1996	7.371	8.428	1537.62
0.2496	7.355	8.415	1545.47
0.3019	7.339	8.402	1553.52
0.3489	7.325	8.391	1560.53
0.4034	7.311	8.379	1568.25
0.4532	7.298	8.368	1575.30
0.5045	7.287	8.359	1581.97
308.15 K			
0.0510	7.376	8.432	1519.70
0.1023	7.359	8.418	1528.12
0.1499	7.344	8.406	1535.63
0.1996	7.328	8.393	1543.55
0.2496	7.315	8.382	1550.80
0.3019	7.301	8.371	1558.26
0.3489	7.290	8.362	1564.77
0.4034	7.278	8.352	1571.91
0.4532	7.267	8.343	1578.43
0.5045	7.259	8.337	1584.48

**Fig 1:** Chemical structure of [C₄mim][CF₃SO₃]**Fig 2:** Plot of thermal expansivity vs molalities of [C₄mim][CF₃SO₃] and temperature of the solution**Fig 3:** Plot of β_T vs molalities of [C₄mim][CF₃SO₃] in the temperature range 288.15 K to 303.15 K.**Fig 4:** Dependence of internal pressure values as a function of molalities of [C₄mim][CF₃SO₃] and temperature.**Fig 5:** Energy of vaporization plotted against concentration and the studied temperature range.

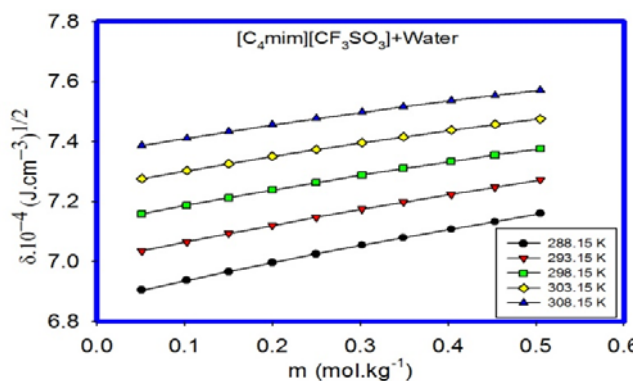


Fig 6. Effect of composition and temperature on the values of solubility parameter.

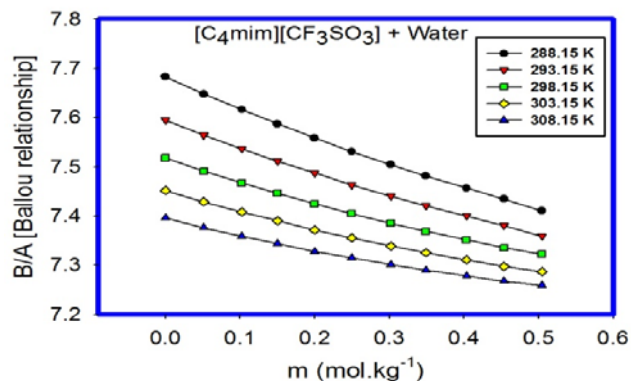


Fig 7: Values of non-linearity parameter from Ballou relationship.

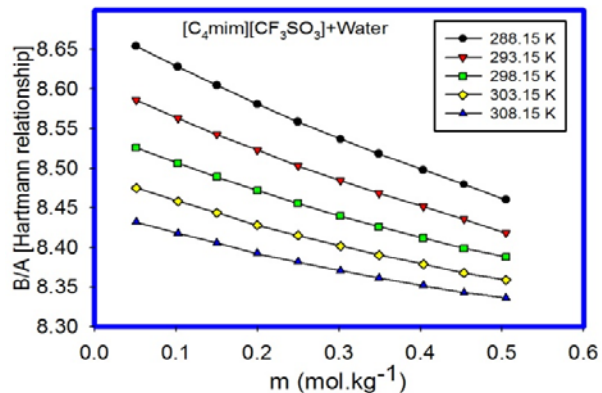


Fig 8: Values of B/A parameter from Hartman relationship.

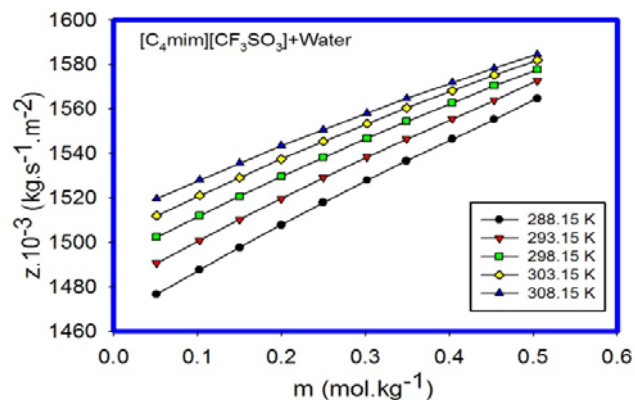


Fig 9: Plot of acoustic impedance vs temperature and composition of $[C_4mim][CF_3SO_3]$.

4. Conclusion: Experimental density and speed of sound data of aqueous $[C_4mim][CF_3SO_3]$ solutions were taken for the evaluation of thermodynamic and acoustical properties. The influence of composition and solution temperature on each property has been carefully analyzed. The study highlights the unusual volume expansion of the mixture due to the strong electrostatic and H-bonding interactions between solute and solvent. The interpretation of the results was also supported by estimating acoustical properties. Study reveals the effect of composition as well as temperature on the estimated properties which is quite significant.

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