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Thermodynamic and Acoustical Properties of Solutions of Tetra Butyl Ammonium Iodide in Various Alcohols at Different Temperatures

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ABSTRACT

Ultrasonic velocities, densities and viscosities of an ionic liquid; tetra butyl ammonium iodide have been measured in different alcohols; methanol, ethanol, 1-propanol and 1-butanol at different concentrations at different temperatures. The concentration and temperature effect have been discussed in terms molecular interactions using various acoustical, apparent and thermodynamic parameters.

Keywords: *Tetra butyl ammonium iodide, ethanol, methanol, 1-propanol, 1-butanol, acoustical properties, apparent properties, thermodynamic parameters, molecular interactions etc.*

INTRODUCTION

Ionic liquids refer tomaterials composed solely of cations and anions [1, 2]. These cations are organic and the anions are inorganic or organic, in which the ions can be designed for specific applications. There are numerous possible combinations of cations and anions, so the interest in ionic liquids is increasing exponentially [3].

The ionic liquids have unique properties such as negligible vapour pressures, low volatility [4-6], high ionic density [7], high ionic conductivity, large electrochemical window [8], high thermal and chemical stability [4, 6],wide temperature range for liquid phase [9,10],high viscosity [11-13]and ability to solvate compounds of wide polarity range [14, 15]. Further, these ionic liquids are generally nontoxic [16, 17] and are used in organic synthesis [18, 19]. These liquids have recyclability [20], and last tunability [21]. These properties give them wide applications in different fields.

The quaternaryammonium ionic liquid was the first ionic liquid reported by Gabriel in 1888 [22]. Later on, in 1914, Walden had synthesized this ionic liquid [23]. Various physic-chemical properties of ionic liquids can be controlled by the anionic or cationic modifications in the ionic liquid's structures. So, various ionic liquids of different physical properties can be prepared depending upon the application of ionic liquids in different fields [24-27].

Due to the presence of hydrophobic and hydrophilic groups in quaternary ammonium ionic liquids, the study of their solutions behaviour is an interesting area [28-31]. The extent of molecular interactions and its nature can be explained qualitatively by the evaluation of some useful thermodynamic and transport parameters in solutions [32,33]. Various workers have studied solutions of tetra alkyl ammonium salts in various solvents at different temperatures [33-37].

In the present work, the acoustical, volumetric and thermodynamic properties of an ionic liquid; tetra-n-butyl ammonium iodide have been studied in different alcohols (methanol, ethanol, 1-propanol and 1-butanol) at different temperatures over a wide range of concentrations.

EXPERIMENTAL

Chemicals

The tetra-n-butyl ammonium iodide (TBAI) was purchased from Datt Chemicals (Bhavnagar, India) and was used as such. The HPL Cgrade solvents, methanol, 1-propanol and 1-butanol supplied by Spectrochem Pvt. Ltd (Mumbai, India) and was purified by procedure reported in literature [38]. The ethanol was supplied by Shree Chalthan Vibhag Khand UdyogS ahakari Mandii Ltd. (Surat, India) and was 99% pure and was used as such.

The structure of tetra-n-butyl ammonium iodide ionic liquid is shown in Fig 1.

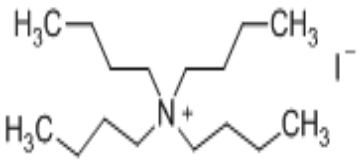


Figure1. *Tetra butyl ammonium iodide structure*

The solutions of tetra-n-butyl ammonium iodide were prepared in different alcohols over a wide range of concentrations. For the preparation of solutions, an electric balance (Mettler Toledo AB204-S) of ±0.1mg accuracy was used.

Apparatus and Procedure

An Anton Paar (DSA 5000 M) instrument was used for the measurements of densities and sound velocities of pure alcohols and solutions of TBAI at different temperatures (298.15, 308.15 and 318.15 K). The accuracies of density and velocity measurements were 0.005kg·m^{-3} and 0.5 m·s^{-1}, respectively.

An Ubbelohde viscometer was used to measure the viscosities of pure solvents and ionic liquids solutions at different temperatures (298.15, 308.15 and 318.15 K) with uncertainty of ± 0.09. The NOVA viscosity bath with an accuracy of 0.5 °C is used to control the temperature. Before viscosity measurements, the pure solvents/ solutions were allowed to attain the desired temperature in the viscosity bath. A Hanhart Gütenbach (made in Germany) stop watch of 0.01s accuracy was used to measure the flow time of pure solvents and solutions. The viscosity was calculated by the equation:

$$\frac{\eta_1}{\eta_2} = \frac{t_1\rho_1}{t_2\rho_2} \quad (1)$$

Where, ρ_1, ρ_2,η_1 and η_2 are the density and viscosity of pure water and solution respectively.

RESULTS AND DISCUSSION

Acoustical Parameters

For the pure alcohols, the experimental sound velocity, viscosity and density at 298.15 K are given in Table 1 along with the theoretical values reported in literature [39-44].

Table1.*Experimental values of density, sound velocity and viscosity of pure solvents at 298.15K.*

Solvents	Density, kg.m^{-3}		Sound velocity,m.s^{-1}		Viscosity.10^3,Pa.s	
Methanol	786.814	(786.636[39*])	1102.63	(1102.54[39*])	0.542	(0.542[40*])
Ethanol	787.043	(786.60[41*])	1148.11	(1145.0[41*])	1.070	(1.079[41*])
1-Propanol	799.606	(799.620[39*])	1205.46	(1205.81[39*])	1.901	(1.915[42*])
1-Butanol	805.998	(805.90[43*])	1239.67	(1239.72[39*])	2.547	(2.520[44*])

(*) The values in parenthesis are from literature.

The measured density, sound velocity and viscosity of solutions of tetra butyl ammonium iodide (TBAI) in different solvents at different temperatures are presented in Table 2, where these values are found to increase with increase in concentration and decrease with increase the temperature.

Table2.*The density, sound velocity and viscosity of TBAI in different solvents at different temperatures.*

Conc., mol/ lit	Density, kg /m^3	Velocity, ms^{-1}	Viscosity .10^3 ,Pa .s	Density, kg /m^3	Velocity, ms^{-1}	Viscosity.10^3 , Pa. s	Density, kg /m^3	Velocity, ms^{-1}	Viscosity.10^3 ,Pa. s
	298.15K			**308.15K**			**318.15K**		
Methanol									
0.10	808.452	1101.17	0.572	797.623	1079.11	0.530	784.676	1052.13	0.453
0.20	820.144	1116.84	0.673	810.975	1092.47	0.624	801.701	1061.1	0.522
0.40	846.921	1135.48	0.865	837.865	1114.01	0.788	828.712	1087.04	0.674
0.60	873.108	1165.64	1.003	863.219	1132.21	0.884	854.257	1101.84	0.773
0.80	895.209	1187.11	1.181	886.396	1153.69	1.003	877.532	1125.23	0.849
1.00	915.035	1204.69	1.399	906.331	1172.66	1.174	897.599	1144.55	1.031
Ethanol									
0.10	806.548	1164.82	1.129	796.755	1132.89	0.831	784.354	1100.32	0.784
0.20	817.822	1176.68	1.333	809.232	1143.49	0.984	800.492	1110.79	0.853
0.40	843.371	1192.09	1.736	834.866	1159.64	1.326	826.233	1127.4	1.139
0.60	867.127	1206.57	2.075	858.752	1174.57	1.705	850.246	1143	1.419
0.80	892.384	1222.96	2.535	884.07	1191.68	2.090	875.644	1160.55	1.723
1.00	917.624	1240.72	3.179	909.413	1209.77	2.538	901.107	1179.31	2.037
1-Propanol									
0.10	815.035	1214.72	2.222	806.981	1181.37	1.796	798.773	1148.68	1.434

0.20	827.467	1220.71	2.464	819.439	1187.39	1.984	811.274	1154.6	1.622
0.40	853.099	1233.98	3.125	845.117	1201.43	2.538	837.017	1169.17	1.953
0.60	875.938	1246.38	3.776	867.997	1214.22	3.019	859.955	1182.63	2.294
0.80	897.618	1256.23	4.595	889.76	1224.66	3.625	881.826	1193.5	2.841
1.00	905.526	1269.12	5.909	913.403	1237.98	4.383	905.526	1207.44	3.368
1-Butanol									
0.10	817.064	1245.86	2.936	809.345	1212.59	2.200	801.494	1179.62	1.861
0.20	827.500	1249.70	3.277	819.808	1216.69	2.527	811.993	1184.26	2.078
0.40	850.087	1258.69	3.974	842.417	1226.32	3.197	834.65	1194.27	2.621
0.60	873.516	1267.87	5.077	865.888	1235.95	4.038	858.146	1204.61	3.137
0.80	901.149	1280.11	6.423	893.549	1248.82	4.964	885.884	1217.94	3.739
1.00	924.866	1288.79	8.476	917.357	1257.96	6.006	909.817	1227.77	4.548

For the understanding of molecular interactions in solutions, some acoustical and apparent properties were also evaluated from experimental data, using equations reported in our earlier publication [45]:

Acoustical impedance (Z):$Z = U\rho$ (2)

Where, ρ is density and U is sound velocity of solution.

Isentropic compressibility (κ_S):$\kappa_s = \frac{1}{\rho U^2}$ (3)

Intermolecular free path length (L_f):

$L_f = K_j \kappa_s^{1/2}$ (4)

Where, the value of Jacobson's constant (K_j) is $((93.875 + 0.375T)\cdot 10^{-8})$.

Molar compressibility(W): $W = \left(\frac{M}{\rho}\right)\kappa_s^{-1/7}$ (5)

Where, the apparent molecular weight (M)of solution is calculated by the equation:

$M = M_1W_1 + M_2W_2$ (6)

whereW_1, W_2, M_1 and M_2 are the weight fractions and molecular weight of solvent and solute respectively.

Relaxation strength (r):

$r = 1 - \left(\frac{U}{U_\infty}\right)^2$ where,

U_∞= 1600 m/s (7)

Rao's molar sound function (R_m):

$R_m = \left(\frac{M}{\rho}\right)U^{1/3}$ (8)

Van der Waals constant (b):

$b = \frac{M}{\rho}\left\{1 - \left[\frac{RT}{MU^2}\right]\left[\sqrt{1 + \left(\frac{MU^2}{3RT}\right)} - 1\right]\right\}$ (9)

Where, the gas constant (R = 8.3143 (J $K^{-1}mol^{-1}$) and T is the absolute temperature.

Solvation number (S_n):

$S_n = \frac{M_2}{M_1}\left[\frac{(100-X)}{X}\right]\left[1 - \frac{K_S}{K_S^\circ}\right]$ (10)

whereM_1, M_2, $\kappa^0_s$and κ_S are molar masses and adiabatic compressibility of pure solvent and solution respectively and$X$ is the number of grams of solute in 100 g of the solution.

Internal pressure (π_i):

$\pi_i = b'RT(K\eta/U)^{1/2}(\rho^{2/3}/M^{7/6})$ (11)

Where, packing factor b' (=2), K is a constant (4.28×10^9).

Free volume (V_f): $V_f = \left(\frac{MU}{K\eta}\right)^{3/2}$ (12)

Molar cohesive energy (MCE):

$MCE = V_m\pi_i$ (13)

Where V_m is the molar volume.

Tables 3 and 4show some of these evaluated parameters.

Table3. *Some acoustical parameters such as intermolecular free path length, acoustical impedance and relaxation strength of TBAI in different solvents at different temperatures*

Conc.mol/lit	298.15K	308.15K	318.15K	298.15K	308.15K	318.15K	298.15K	308.15K	318.15K
intermolecular free path length, m				**Acoustical impedance.10^{-6}, kg.m^{-2} s^{-1}**			**Relaxation strength**		
Methanol									
0.10	6.629	6.810	7.042	0.890	0.861	0.826	0.526	0.545	0.568
0.20	6.489	6.671	6.908	0.916	0.886	0.851	0.513	0.534	0.560
0.40	6.281	6.437	6.633	0.962	0.933	0.901	0.496	0.515	0.538
0.60	6.026	6.239	6.445	1.018	0.977	0.941	0.469	0.499	0.526
0.80	5.844	6.043	6.227	1.063	1.023	0.987	0.450	0.480	0.505
1.00	5.696	5.879	6.053	1.102	1.063	1.027	0.433	0.463	0.488
Ethanol									
0.10	6.274	6.491	6.735	0.939	0.903	0.863	0.470	0.499	0.527

0.20	6.168	6.381	6.604	0.962	0.925	0.889	0.459	0.489	0.518
0.40	5.995	6.194	6.405	1.005	0.968	0.931	0.445	0.475	0.504
0.60	5.842	6.030	6.228	1.046	1.009	0.972	0.431	0.461	0.490
0.80	5.681	5.858	6.044	1.091	1.054	1.016	0.416	0.445	0.474
1.00	5.522	5.689	5.863	1.139	1.100	1.063	0.399	0.428	0.457
1-Propanol									
0.10	5.985	6.185	6.393	0.990	0.953	0.918	0.424	0.455	0.485
0.20	5.911	6.106	6.311	1.010	0.973	0.937	0.418	0.449	0.479
0.40	5.759	5.943	6.136	1.053	1.015	0.979	0.405	0.436	0.466
0.60	5.627	5.802	5.985	1.092	1.054	1.017	0.393	0.424	0.454
0.80	5.515	5.682	5.856	1.128	1.090	1.052	0.384	0.414	0.444
1.00	5.435	5.547	5.712	1.149	1.131	1.093	0.371	0.401	0.431
1-Butanol									
0.10	5.828	6.017	6.215	1.018	0.981	0.945	0.394	0.426	0.456
0.20	5.774	5.958	6.151	1.034	0.997	0.962	0.390	0.422	0.452
0.40	5.656	5.831	6.016	1.070	1.033	0.997	0.381	0.413	0.443
0.60	5.539	5.707	5.882	1.108	1.070	1.034	0.372	0.403	0.433
0.80	5.401	5.560	5.726	1.154	1.116	1.079	0.360	0.391	0.421
1.00	5.296	5.448	5.605	1.192	1.154	1.117	0.351	0.382	0.411

Table4. *Some acoustical parameters such as molar cohesive energy (MCE), Rao's molar sound function (R_m) and Van der Waals constant (b) of TBAI in different solvents at different temperatures.*

Conc., mol/lit	**298.15K**	**308.15K**	**318.15K**	**298.15K**	**308.15K**	**318.15K**	**298.15K**	**308.15K**	**318.15K**
MCE , kJ. mol^{-1}				**$R_m.10^{-4}m^{10/3}\cdot s^{-1/3}\cdot mol^{-1}$**			**b.10^{-5}**		
Methanol									
0.10	29.743	30.017	29.176	6.061	6.129	6.212	10.986	10.643	10.283
0.20	31.912	32.227	31.002	7.897	7.972	8.031	17.590	17.031	16.341
0.40	35.546	35.539	34.463	11.196	11.324	11.438	32.500	31.736	30.799
0.60	37.529	37.091	36.413	14.184	14.326	14.456	49.667	48.102	46.612
0.80	40.424	39.171	37.803	16.961	17.098	17.262	68.157	65.970	64.175
1.00	44.136	42.484	41.715	19.561	19.725	19.912	87.640	85.046	82.847
Ethanol									
0.10	40.598	36.659	37.486	6.195	6.239	6.308	11.939	11.702	11.506
0.20	43.725	39.523	38.675	8.070	8.120	8.173	18.972	18.610	18.250
0.40	49.119	45.145	43.961	11.459	11.545	11.635	34.888	34.393	33.893
0.60	53.017	50.497	48.368	14.519	14.633	14.753	52.617	51.987	51.363
0.80	58.035	55.339	52.726	17.226	17.365	17.508	71.158	70.430	69.691
1.00	64.751	60.726	57.046	19.653	19.808	19.970	90.181	89.334	88.501
1-Propanol									
0.10	55.549	52.515	49.296	6.196	6.220	6.246	12.454	12.167	11.885
0.20	58.088	54.792	52.041	8.028	8.071	8.116	19.507	19.135	18.767
0.40	64.461	61.031	56.210	11.374	11.449	11.528	35.600	35.086	34.571
0.60	70.009	65.738	60.133	14.423	14.525	14.631	53.567	52.912	52.269
0.80	76.680	71.494	66.385	17.201	17.325	17.452	72.536	71.750	70.965
1.00	87.074	78.264	71.914	20.284	19.804	19.950	95.488	90.900	90.009
1-Butanol									
0.10	62.964	57.280	55.322	6.228	6.250	6.274	12.847	12.559	12.271
0.20	66.169	61.053	58.127	8.091	8.131	8.174	20.121	19.751	19.386
0.40	72.004	67.827	64.450	11.517	11.589	11.664	36.766	36.248	35.727
0.60	80.468	75.338	69.652	14.575	14.672	14.775	55.029	54.365	53.717
0.80	89.498	82.557	75.120	17.189	17.307	17.428	73.542	72.760	71.978
1.00	102.359	90.376	82.408	19.623	19.754	19.890	92.649	91.730	90.825

The variation of sound velocity with concentration is also shown graphically in Figure 1. It is observed that at all the studied temperatures, there is linear increase in sound velocitywith concentration in all solutions. Further, for all the solutions, the order of sound velocity is: 1-butanol > 1-propanol > ethanol > methanol. That is, as chain length of alcohol increases, sound velocity increase and decrease with temperature increases.

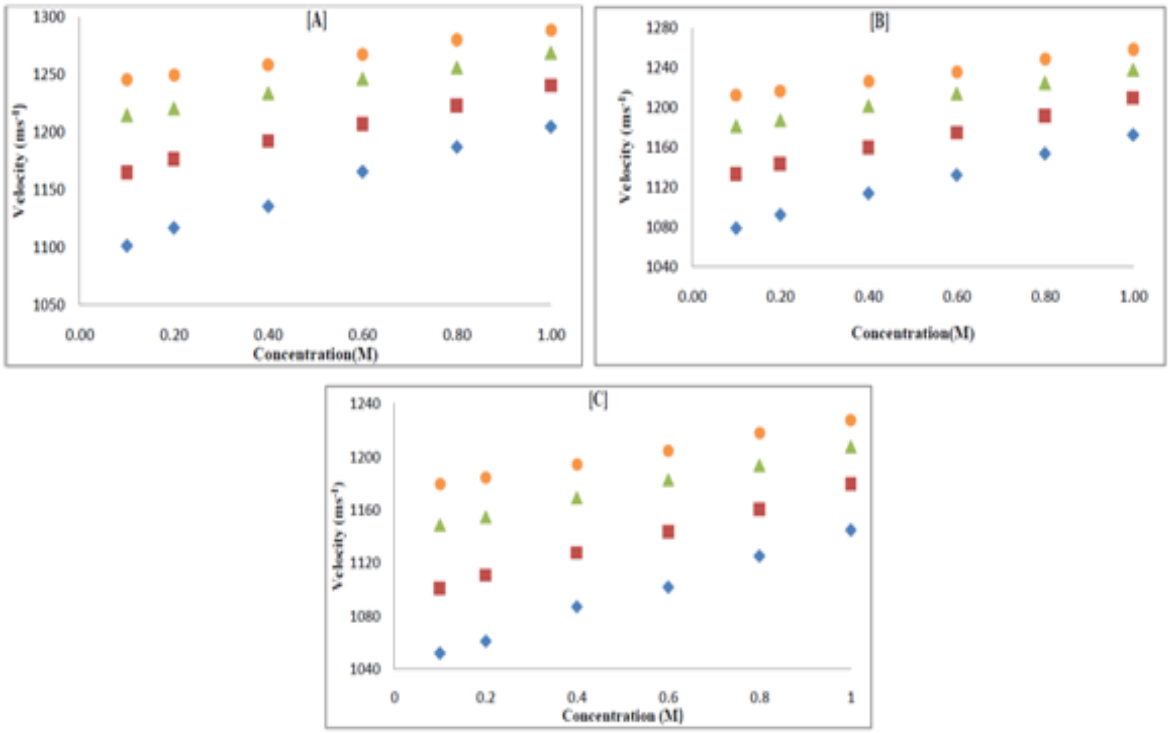


Figure1. *The variation of sound velocity of TBAB in alcohols at [A] 298.15 K, [B] 308.15K and [C] 318.15K. methanol (♦), ethanol (■), 1-propanol (▲) and 1-butanol (●)*

The sound velocity change is related to intermolecular free path length. Table 3shows that as the concentration increases, intermolecular free path length decreases. When concentration is low, molecules are far apart from each other but molecules come close to each other, when concentration increases. When solute and solvent molecules come close to each other the distance decreases i.e. intermolecular free bath length decreases [46], so there will be interaction between these solute and solvent molecules. Thus, when distance between solute and solvent molecules decreases, sound velocity increase. Thus, intermolecular free path length is reverse of sound velocity [47]. Thus, when molecules are closely packed, the sound waves propagate with higher velocity [48,49] due to existence of solute-solvent interactions.

However, when temperature increases, these interactions become weak due to thermal disturbances which causes intermolecular free length to increase as evident from Table 3. This in turn, causes decrease of sound velocity with temperature. As evident from Table 3 that intermolecular free path length decreases with increase in chain length in alcohols in all the concentrations and at all the studied temperatures.

Figure 2 shows the variation of isentropic compressibility with concentration at 298.15 K. It is observed from Figure 2 that isentropic compressibility decreases with concentration. For other temperatures also, variation in isentropic compressibility with concentration is of same nature.

The decrease of isentropic compressibility may be due to accommodation (penetration) of solvent molecules inside the void space of large bulky group Bu_4N^+, which decreases the intermolecular free space and repulsive forces between the ions. This results in compact like structure [46]. As the chain length is bigger in 1-butanol, the decrease of isentropic compressibility is maximum in 1-butanol and minimum in methanol. Thus, interactions in 1-butanol are higher than the rest of the studied alcohols.

As shown in Table 3, the decrease of relaxation strength (r) and increase of acoustical impedance (Z) with concentration also confirm the existence of solvent-solute interactions in studied solutions. Reverse is true with increase in temperature. Table 3 shows that relaxation strength decreases with increase the chain length of alcohols for 1-butanol.

Table 4 shows that when concentration increase, both van der Waals constant and Rao's molar sound function increase for all the solvents and at different temperatures and the order is: 1-butanol > 1-propanol > ethanol > methanol. The order is reverse when temperature increases.

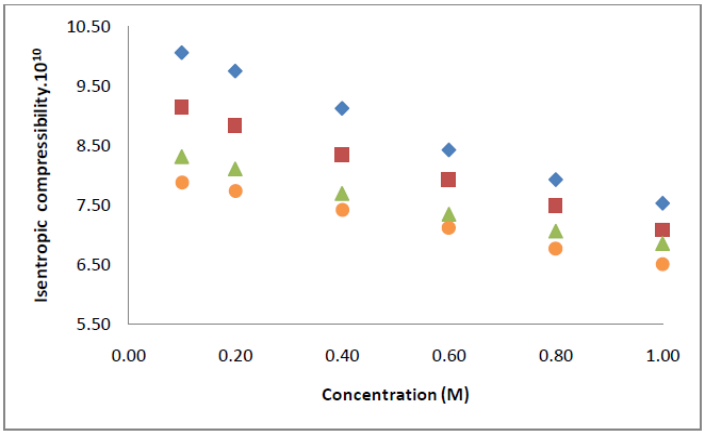


Figure2. *The variation of isentropic compressibility (κ_S) of TBAI with concentration at 298.15K in alcohols, methanol (♦), ethanol (■), 1-propanol (▲) and 1-butanol (●)*

The extent of linear variation of some of the acoustical parameters is given in Table 5 by least square fitting equations and correlation coefficients (γ). It is observed from Table 5 that most of the parameters, correlation coefficients (γ) is greater than 0.9944. The linear change in Rao's molar sound function, van der Waals constant and molar compressibility suggest absence of complex formation in all studied solvents [50].

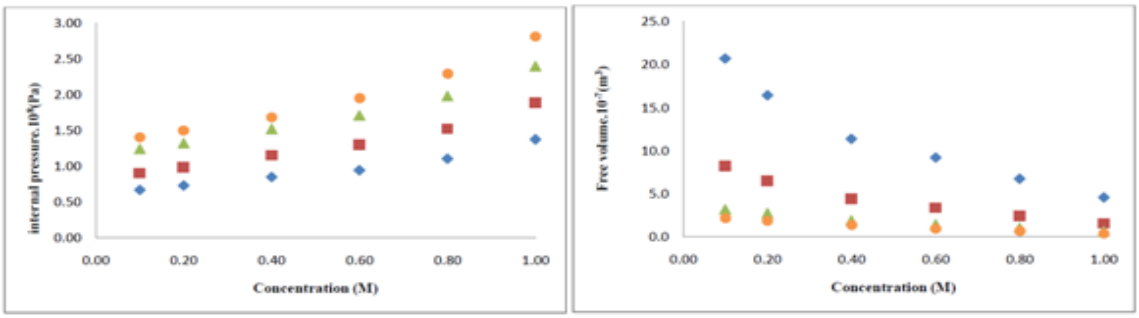
Table5. *The Least square equations and regression coefficients (γ) for some acoustical parameters of TBAI in different solvents at 298.15K*

Parameters / solvents	Methanol	Ethanol	1-Propanol	1-Butanol
U ($m.s^{-1}$)	116.37C + 1091.7γ =0.994	81.699C + 1158.4 γ =0.9976	60.081C + 1209.1 γ = 0.9984	48.616C + 1240.1 γ =0.9967
ρ ($kg\ m^{-3}$)	120.48C+ 797.56 γ =0.997	0.1236C+ 0.7936 γ =0.9999	0.1049C + 0.8082 γ = 0.9802	0.1208C + 0.8033 γ = 0.9986
$\kappa_s .10^{10}$ ($m^2.N^{-1}$)	-3.0021C+ 10.388 γ = 0.9887	-2.2568C + 9.3004 γ = 0.9971	-1.6512C + 8.4178 γ =0.986	-1.551C + 8.0432 γ = 0.9991
$L_f .10^{11}$ (m)	-1.0506C +6.7036 γ = 0.9921	-0.824C + 6.3396 γ = 0.9985	-0.6234C+ 6.0272 γ = 0.9887	-0.6002C + 5.8923 γ =0.9991
$R_m .10^{-4}$ ($m^{10/3}. s^{-1/3}.mol^{-1}$)	14.974C+ 4.9068 γ = 0.9971	14.985C + 5.1114 γ = 0.995	15.499C + 4.9101 γ =0.9985	14.907C + 5.1684 γ = 0.9944
b $.10^{-5}$ ($m^3. mol^{-1}$)	85.266C + 0.3695 γ = 0.9964	87.256C + 1.5435 γ = 0.9981	91.478C + 0.9284 γ =0.9948	89.079C + 2.4683 γ =0.9988
$W.10^3$ ($m^3. mol^{-1}$) ($Nm^2)^{1/7}$	2.845C + 0.9077 γ =0.9975	2.8486C+ 0.9396 γ =0.9957	2.9362C + 0.9042 γ = 0.9987	2.8329C + 0.9483 γ =0.9952
R	-0.1049C+ 0.5354 γ =0.9949	-0.0768C+ 0.4763 γ =0.9977	-0.0583C + 0.4292 γ = 0.9987	-0.0481C + 0.3995 γ = 0.9964

The internal pressure (π_i) is the resultant of the forces of attraction and repulsion between the molecules in a liquid [51]. The variation of internal pressure with concentration in studied solvents is shown in Figure 3 [A] for 298.15K. It is observed that as the concentration of TBAI in all studied solvents increase with increase in concentration.

The increase in internal pressure results due to increase in cohesive attraction between solute and solvent molecules, which causes ordered structural arrangement due to molecular association between solute and solvent molecules [52]. With increase in temperature, ions / molecules move away from each other due to thermal agitation thus reducing the possibility for the interactions [53].

Due to irregular packing of molecules, there exists void space between the molecules which is known as free volume [51]. Figure 3 [B] shows that in all the studied solutions, free volume decreases with increase in concentration indicating thereby interactions between solute and solvent molecules, which causes compact packing [54]. The order is: 1-butanol < 1-propanol < ethanol < methanol. Thus, interactions increase with increase in chain length of alcohols, due to increase the ability of making hydrogen bonds, which decrease with increase the temperature.



[A] [B]

Figure3. *The variation of [A] internal pressure and [B] free volume with concentration of TBAI instudied solvents at 298.15, methanol (♦), ethanol (■), 1-propanol (▲) and 1-butanol (●)*

Comparison of Figures 3 [A] and [B] shows that free volume is inverse of internal pressure. Similar results have been reported by other workers [55]. Thus, due to increase in cohesive forces, internal pressure increases whereas free volume decreases.

Molar cohesive energy is the parameter, which relates the free energy state of the liquid system to the escaping tendency. Table 4 shows that as concentration increases, MCE increases whereas with increase temperature, it decreases. The order in different alcohols is: 1-butanol > 1-propanol > ethanol >methanol. The overall increase of MCE with concentration confirms the enhancement of the structure-forming tendency of TBAI molecules [51].

The solute tendency of structure forming or breaking in the solution can be given by the solvation number. The positive value of the solvation number indicates structure forming tendency, whereas the negative value indicates the structure breaking tendency [56]. From Figure 4, it is observed that for all studied solutions, solvation number values are positive at all temperatures which suggests structure forming tendency of TBAI. This is due to structure forming tendency of solute i.e., TBAI.

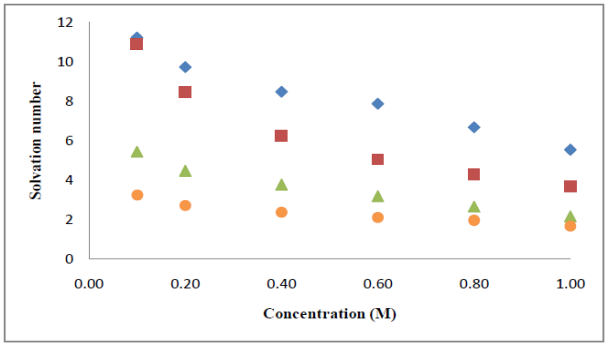


Fig4.*The variation of solvation number with concentration of TBAI in studied solvents at 298.15. methanol (♦), ethanol (■), 1-propanol (▲) and 1-butanol (●).*

Viscosity B Coefficient

The viscosity is transport property of electrolyte solution andit's measurement give information aboutdifferent types of interactions in solution. It depends upon the shape and size of the constituent ions [57].

Table 2 shows that experimental viscosity of all the solutions of TBAI are found to increase with increase in concentration at different temperatures and decreases with increase in temperature. As concentration increases, number of collisions between molecules increases which causes loss of kinetic energy due to which molecules stick together. This causes increase in viscosity [58].

The increase in viscosity with concentration again proves the existence of association between solute and solvent molecules.

The relative viscosities are related to concentration by Jones-Dole equation [59, 60], which also provide useful insights to the extent of ionic solvation.

$$\frac{\eta}{\eta o} = 1 + AC^{1/2} + BC \qquad (18)$$

Where, η and η_0 are the viscosity of the solution and solvent respectively. A is known as Falkenhagen coefficient which reflects solute-solute interactions. The viscosity B coefficient is related to the molar volume of the solvent, size of solute

and is a measure of solute-solvent interactions [61, 62].

Usually A values are small and negative. It is reported that for organic electrolytes, *A* value can be neglected at moderate or high concentrations [63]. So, above equation is simplified as:

$$\frac{\eta}{\eta o} = 1 + BC \qquad (19)$$

From the linear plot of (η/η_o–1) versus concentration, value of coefficient B can be evaluated from slope which is given in Table 6.

In studied solutions, at all the temperatures B values are positive which indicates that the interactions between solute-solvent molecules are strong [58]. Thus, it is again proved that TBAI behaves as structure makers.

However, as temperature increases, *B* values decrease due to decrease in strength of solute-solvent interactions. Further, as the chain length of alcohols increases, B-coefficient increase. Similar results have also been reported by other workers [58, 64, 65].

Table6. *The coefficient B ofJones-Dole equationfor TBAI solutions at different temperatures.*

Solvent	298.15K	308.15K	318.15K
Methanol	1.6433	1.2585	1.1232
Ethanol	2.0988	1.7643	1.3233
1-Propanol	2.4121	1.4912	1.1111
1-Butanol	2.9155	1.6498	1.1489

Apparent Parameters

The behaviour of a solute in a solution can also be predicted by evaluating their apparent properties. So, for studied solutions, apparent molar compressibility and apparent molar volume were evaluated at different temperatures.

The difference between the volume of the solution and the volume of the pure solvent per mole of solute is defined as apparent molar volume (φ_V) which is calculated given by following equation [58]:

$$\varphi_V = \frac{M_2}{\rho} - \frac{1000(\rho-\rho_0)}{m\rho\rho_0} \qquad (14)$$

Where, $M_2$is the molar mass of the solute and *m* is the molality (the number of moles of the solute per kilogram of solvent). ρ and ρ_0 are densities of solution and solvent, respectively. C is the molar concentration. The evaluated values of apparent molar volume are listed in Table 7.

The apparent molar volume depends on type of salt, size of ions, etc. Table 7 shows that, (φ_V) values are positive and large, which is due to large organic ion with large intrinsic volume [66]. Further, large apparent molar volume indicates strong interactions due to hydrogen bonding [67]. It is observed that apparent molar volume is highest for 1-butanol.

Table7. *The apparent molar volume for TBAI solutions in different solvents at different temperatures.*

Concentration M	Apparent molar volume. 10^6 m^3/mole			
	298.15K			
	Methanol	Ethanol	1-Propanol	1-Butanol
0.10	181.890	210.141	260.250	310.332
0.20	238.581	256.127	272.181	310.737
0.40	245.162	259.058	265.738	296.608
0.60	240.271	256.393	262.593	282.456
0.80	240.413	246.620	258.292	261.717
1.00	240.716	236.626	251.733	251.404
308.15K				
0.10	202.252	227.486	262.217	317.795
0.20	239.170	258.250	274.317	315.735
0.40	246.225	261.007	267.743	300.254
0.60	243.794	258.034	264.488	285.439
0.80	241.365	248.083	259.976	264.208
1.00	241.625	237.827	250.393	253.491
318.15K				
0.10	250.479	278.431	264.078	316.808
0.20	239.740	260.334	276.327	316.437
0.40	247.272	262.887	269.646	301.633
0.60	244.642	259.646	266.284	286.928

0.80	242.219	249.488	261.539	265.445
1.00	242.414	238.950	251.785	254.489

The limiting apparent molar volume (φ_V^0) is evaluated by extrapolation of experimental apparent molar volume values using the following equation [68]:

$$\varphi_V = \varphi_V^0 - S_V C^{\frac{1}{2}} \quad (15)$$

Where solute-solute interactions is measured by coefficient S_V and C is the molar concentration.

From the plot of φ_V versus $C^{1/2}$, slope gives value of S_V and intercept gives φ_V^0. These evaluated values are listed in Table 9. The positive φ_V^0 indicates strong solute-solvent interactions [69].

The apparent molar isentropic compressibility (φ_κ) is another parameter which relates thermodynamics and acoustics. This can be obtained from density and sound velocity measurements, using the following [70]:

$$\varphi_\kappa = \left(\frac{\kappa_S \rho_0 - \kappa_{S,0}\rho}{m\rho\rho_0}\right) + \frac{M_2 \kappa_S}{\rho} \quad (16)$$

Where, κ_S and $\kappa_{S,0}$ are the isentropic compressibility of the solution and solvent respectively. M_2 is the molar mass of TBAI. The evaluated values of apparent molar compressibility are listed in Table8.

Table8. *The apparent molar isentropic compressibility of TBAI in different solvents at different temperatures*

ConcentrationM	Apparent molar isentropic compressibility.10^{10} m^5 .N^{-1}.mol^{-1}			
298.15K				
	Methanol	**Ethanol**	**1-Propanol**	**1-Butanol**
0.10	-5.901	-7.396	-4.569	-3.024
0.20	-5.780	-5.920	-3.978	-2.770
0.40	-5.499	-4.960	-3.707	-2.731
0.60	-5.281	-4.494	-3.462	-2.717
0.80	-4.956	-4.293	-3.250	-2.821
1.00	-4.624	-4.157	-2.888	-2.756
308.15K				
0.10	-7.624	-8.086	-5.014	-2.423
0.20	-6.948	-6.512	-4.347	-2.584
0.40	-6.231	-5.465	-4.063	-2.761
0.60	-5.731	-4.949	-3.790	-2.813
0.80	-5.417	-4.726	-3.559	-2.959
1.00	-5.074	-4.569	-3.472	-2.909
318.15K				
0.10	-8.481	-8.076	-5.712	-1.858
0.20	-7.750	-7.232	-4.850	-2.452
0.40	-7.103	-6.051	-4.500	-2.824
0.60	-6.355	-5.478	-4.190	-2.945
0.80	-6.026	-5.221	-3.928	-3.130
1.00	-5.632	-5.044	-3.827	-3.095

The apparent molar isentropic compressibility φ_κ values are negative in all solvents. Further, apparent molar compressibility in studied solvents increases with increase in concentration. The negative apparent molar isentropic compressibility is due to the aggregation of the molecules of solvent around TBAI ions, so the compression resistance will be more than the bulk and this negativity decreases with increase the chain length of alcohols suggesting thereby increase in interactions [71]. Further, apparent molar isentropic compressibility decreases with temperature.

The large organic cation (tetra butyl ammonium) has high intrinsic compressibility as compared to the small monoatomic iodide anion. The iodide ions remain un-solvated, so there is no possibility to have intrinsic compressibility. So, it is assumed that compressibility of the solution is unaffected by poor solvation of iodide ions. Further, there may be possibility of another effect, where solvent molecules can penetrate into the intra-ionic free space. This is an electrostriction effect and causes constriction in the solution volume i.e., more compact. The negative φ_κ is due to the penetration effect which is more than the intrinsic compressibility effect of the tetra-*n*-alkyl ammonium ions [72]. As chain length increases, intrinsic compressibility overrides the penetration effect [73].

The limiting apparent molar isentropic compressibility can be evaluated by the following Gucker's equation [74]:

$$\varphi_\kappa = \varphi_\kappa^0 - S_\kappa C^{\frac{1}{2}} \quad (17)$$

From the linear plots, $\varphi_\kappa^0$and S_κ values can be evaluated from the intercept and slope. The evaluated values of these coefficients are given in Table 9.

Table9. *The coefficients of apparent molar properties in TBAI solutions at different temperatures.*

Solvent	$\Phi_V^0.10^6$ m^3mol^{-1}	$S_V.10^6$ $m^{9/2}mol^{-3/2}$	$\Phi\kappa^0.10^{10}$ $m^5N^{-1}mol^{-1}$	$S_k.10^{10}$ $m^{13/2}N^{-1}mol^{-1}$
298.15K				
Methanol	251.05	-11.405	-7.0618	2.3887
Ethanol	277.06	-34.799	-6.2642	2.1667
1-Propanol	288.26	-34.887	-4.8782	1.8958
1-Butanol	347.67	-91.698	-2.9765	0.2559
308.15K				
Methanol	254.42	-13.529	-8.6506	3.6602
Ethanol	279.99	-36.476	-6.9208	2.4152
1-Propanol	293.2	-39.773	-5.4809	2.1254
1-Butanol	349.96	-107.88	-2.2357	-0.7465
318.15K				
Methanol	255.83	-14.174	-9.6855	4.1244
Ethanol	282.9	-38.2	-7.691	2.7171
1-Propanol	295.78	-40.894	-6.2426	2.5708
1-Butanol	371.57	-115.48	-1.2017	-1.9885

Table 9 show that all the coefficients i.e., φ_V^0, S_V, φ_κ^0andS_κ vary systematically in alcohols from methanol to 1-butanol. Further, for all the studied solvents, φ_V^0values are large and positive whereas φ_κ^0 values are negative. As φ_V^0 is a measure of solute- solvent interaction, the positive value indicates strong solute solvent interactions in studied solutions. Further, φ_V^0 and φ_κ^0 increase with increase the chain length of alcohol which may be due to increase the possibility of hydrogen bonding. The order is: 1-butanol > 1-propanol > ethanol> methanol.

The S_V values are negative and increase with increase the chain length of alcohol and temperature. The negative S_V means that solute-solvent interactions are stronger than solute-solute interactions, S_V is minimum in 1-butanol. This again proves that magnitude of solute-solvent interactions is maximum in 1-butanol and minimum in methanol.

The S_κ values decrease with increase the chain length of alcohols, and increase with increase the temperature.

From acoustic data, some thermodynamic parameters such as enthalpy (ΔH^*), Gibbs free energy (ΔG^*) and entropy (ΔS^*) have also been evaluated using the following equations [63]:

$$\Delta G^* = RTln\left(\frac{\eta V_m}{hN_A}\right) \quad (20)$$

$$\left(Rln\left(\frac{\eta V_m}{hN_A}\right) = \frac{\Delta H^*}{T} - \Delta S^*\right) \quad (21)$$

where R is the gas constant, h is Plank constant, N_A is Avogadro number, V_m is the molar volume, and η is the viscosity.

The enthalpy and entropy can be evaluated from the slope and intercept of the plot of $Rln\left(\frac{\eta V_m}{hN_A}\right)$ versus $1/T$.

The evaluated enthalpy and entropy values are reported in Table 10. It is observed enthalpy values are positive whereas entropy values are negative.

The positive enthalpy suggests endothermic process in studied solutions. The negative entropy is due to more ordered state in solution [63], which again confirms the existence of solute-solvent interactions in studied solutions.

Thermodynamics Parameters

Table10. *The variation of the enthalpy and entropy with concentration in different solvents*

Solvent/ Concentration. M	Methanol	Ethanol	1-Propanol	1-Butanol
Enthalpy kJ.mol^{-1}				
0.10	7.80	13.08	16.10	16.89
0.20	8.85	16.47	15.37	16.85
0.40	8.73	15.48	17.37	15.34
0.60	9.16	13.89	18.48	17.86

0.80	11.87	14.11	17.82	20.17
1.00	10.91	16.38	21.71	23.35
Entropy $J.k^{-1}mol^{-1}$				
0.10	-195.09	-182.56	-178.24	-177.68
0.20	-192.77	-172.53	-181.33	-178.66
0.40	-194.80	-177.75	-176.38	-185.09
0.60	-194.07	-184.33	-173.90	-178.42
0.80	-185.62	-184.66	-177.19	-172.15
1.00	-189.10	-178.05	-165.60	-163.04

Figure 5 shows that Gibb's free energy increase linearly with increase in concentration. Further, for all the solutions, Gibb's free energy values are positive which may be attributed to specific interactions in the solution. [75]. Theincrease in Gibbs free energy with temperature suggests that more time is required for the cooperative process or the rearrangement of molecules in the solution [76]. Comparison of Gibbs free energy values between alcohols shows as chain length increases, Gibbs free energy increases i.e., order is: 1-butanol >1-propanol > ethanol > methanol.

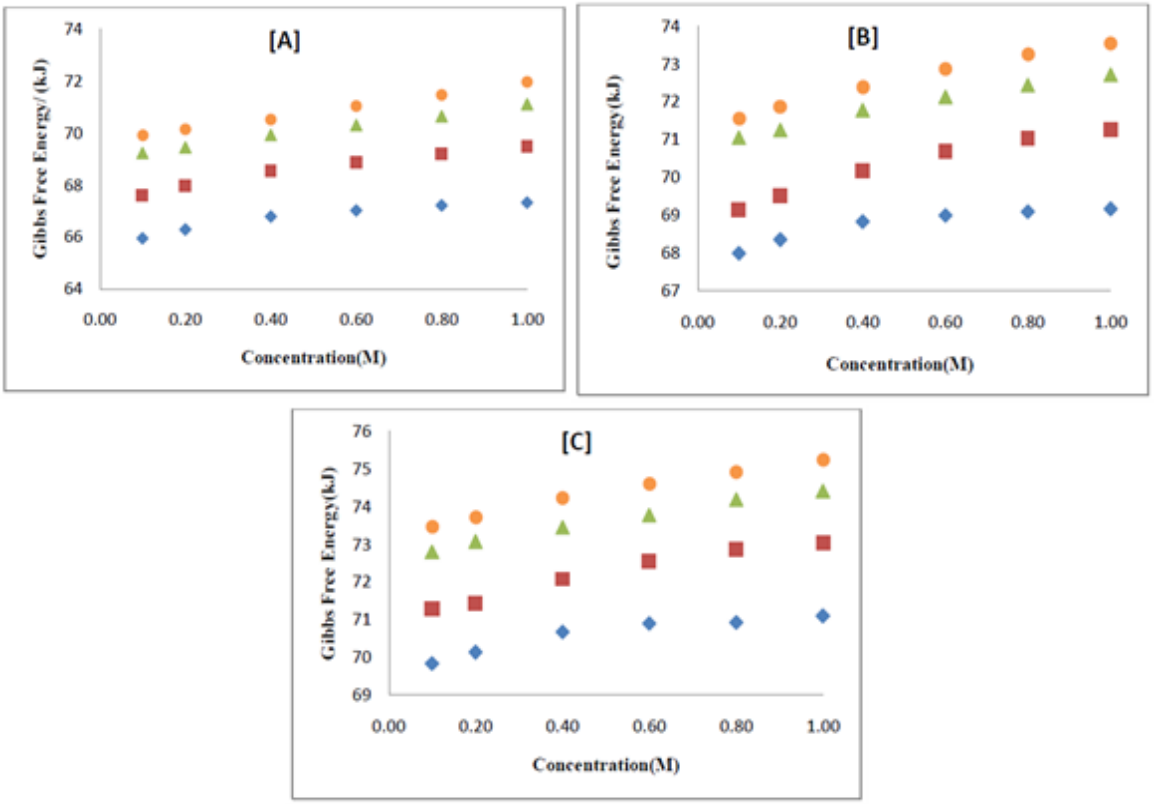


Figure5. *The variation of Gibbs free energy of activation with concentration of TBAI indifferent solvents at [A]298.15 K, [B] 308.15 K and [C] 318.15 K. methanol (♦), ethanol (■), 1-propanol (▲) and 1-butanol (●).*

Conclusion

It is concluded that in all the studied solutions of TBAI, powerful solute-solvent interactions exist and the magnitude of these interactions vary in different alcohols. However, when temperature increases, these interactions become weak due to thermal disturbances. The solute-solvent interactions increase with increase in the chain length of alcohols due to the increase the ability of making hydrogen bonds, which decrease with increase the temperature. The specific interaction between the solute and solvents is again confirmed by negative entropy. The positive enthalpy and Gibbs energy suggest non-spontaneous endothermic process in studied solutions.

Acknowledgments

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