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Studies of acoustical parameters and surface tension of Lithium hydroxide and Lithium bromide in polyethylene glycol (400) electrolyte solution in different concentrations at 303K

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ABSTRACT

The ultrasonic velocity (U) and density (ρ) have been measured in the mixtures of Lithium Hydroxide and Lithium Bromide in Polyethylene glycol (400) solution in different concentrations at 303 K. Ultrasonic velocity has been measured using single frequency interferometer at 2 MHz (Model F-81). From the experimental data, other related thermodynamic parameters, viz adiabatic compressibility, intermolecular free length, and acoustic impedance are calculated. The compressibility of a solvent is higher than that of a solution and it decreases with the increase in concentration of the solution. The abrupt variation of a velocity indicates the formation of complex. Variations of the acoustical parameters in the binary mixtures are studied. The graph between ultrasonic velocity and volume of lithium salts solution in the mixed solution shows some peaks at a particular concentration. The present investigation comprises of theoretical evaluation of acoustic non-linearity parameter, B/A using Hartmann relation, Ballou's empirical relation. A comparative study of B/A values obtained from the aforementioned methods has been made. The results are discussed on the basis of intermolecular interactions present in the liquid mixtures. The non-linear variation of velocity indicates the complex formation between the constituents of the mixture. The results have been discussed in terms of various interactions present in these mixtures.

Key words: Lithium hydroxide-Lithium Bromide-Polyethylene glycol (400)-Acoustical parameters-molecular interactions.

INTRODUCTION

For lithium battery applications, the most common form of electrolyte is a liquid solution of lithium salts dissolved in a suitable polymer solvent. The salt is often of a large anion and the solvent of a mixture having high stabilities. Among the many properties of electrolytes, one of the most important is its physico-chemical and its thermodynamic properties.

Ultrasonic investigations in aqueous solutions in electrolyte and non electrolyte solution with polyethylene glycol provides useful tool in understanding of physic-chemical properties of the interacting components. In recent years, the measurement of ultrasonic velocity has been widely employed in understanding the nature of molecular interactions and physico-chemical properties of the binary liquids [1-5] and to investigate solute-solvent interactions in different proportionate mixtures. These Parameters are required to compute the internal pressure, free volume and other thermodynamic quantities. In many industrial applications, liquid mixtures rather than single component liquid system are used in processing and product formulations. Thermodynamic properties of liquid mixtures have been extensively used to study the departure of a real liquid mixture from ideality. The same authors have studied the ultrasonic velocity and densities in mixtures of polyacrylamide solution in sodium (meta) silicate and potassium silicate solutions in different concentrations at 303 K [6] .Seshagiri and Ramachandra Rao have studied the thermodynamic and transport properties of some lithium salts and they studied the specific interactions between the Lithium salts and solvents [7].

In this study, an attempt has been made to report the experimental value of ultrasonic velocity and density of various compositions of LiOH and LiBr in Polyethylene glycol (400) in lower and higher concentrations at room temperature.

Using ultrasonic velocity and density, the derived acoustical parameters such as adiabatic compressibility (β_{ad}), acoustic impedance (Z), intermolecular free length (L_f) and surface tension (σ) are determined. There is a possibility of complex formation of either stable or unstable type and also it is expected that ultrasonic parameters of a binary mixture will be affected by the concentration of salt solution.

EXPERIMENTAL SECTION

In this present investigation, two electrolytes, Lithium hydroxide and Lithium bromide (AR grade-Merck) were dissolved in distilled water at laboratory temperature to get 1N solution. Polyethylene glycol (400) (PEG) (AR-Grade) is used a solvent. LiOH and LiBr of 0.5N were prepared from the standard solution. LiOH of 0.5N were mixed in PEG (400) solution in different concentration, but the total volume of a binary solution was maintained as 100 ml. Similar method can be followed to prepare the binary solution between LiOH, LiBr and PEG (400) at higher concentration. Ultrasonic velocity and density were measured for LiOH-PEG(400) binary electrolyte solution at different concentration using a ultrasonic interferometer(Mittel Enterprises, New Delhi) with a fixed frequency of 2MHz. Density measurements were carried out using specific gravity bottle at 303K having a volume of 10ml with an accuracy of +0.001kg/$m^{3.}$

RESULTS AND DISCUSSION

Ultrasonic velocities and densities are measured for LiOH and LiBr in PEG (400) in different concentrations are presented in Tables 1-4. Ultrasonic velocity varies in accordance with molecular interactions in solutions. The variation of ultrasonic velocity in a solution depends upon the intermolecular free length (Lf). The ultrasonic velocity increases whereas the free length decreases and vice versa. Presence of ion alters the intermolecular free length. Therefore, ultrasonic velocity of a binary solution will be different from that of the solvent. The minimum value of ultrasonic velocity indicates weakening of the molecular association at these concentrations. The variations of ultrasonic velocity with mole fraction of potassium salt solution at different concentrations are shown in Figure 1. It is observed that the ultrasonic velocity varies non-linearly with concentration and a sudden decrease at a particular concentration shows a complex formation as reported earlier [8-9].

Table 1 Experimental values of ultrasonic velocity (U), acoustic impedance (Z), adiabatic compressibility (β_{ad}), molecular free length (L_f) and surface tension of PEG (400) and Lithium hydroxide binary mixtures at lower concentration

PEG:LiOH	Ultrasonic Velocity (U) ms^{-1}	Acoustic Impedance $(Z)\times10^6$ $kgm^{-2}s^{-1}$	Adiabatic Compressibility $(\beta_{ad})10^{-10}$ $kg^{-1}ms^2$	Molecular Free length (L_f) $10^{-10}m$	Surface Tension (σ) Nm^{-1}
100:00	1565.43	1.7485	3.6532	3.8226	43.58
90:10	1572.25	1.7634	3.8235	3.9108	41.55
80:20	1600.62	1.7950	3.6857	3.8396	42.72
70:30	1652.20	1.8374	3.4835	3.7328	44.49
60:40	1647.76	1.7200	3.4877	3.7350	44.49
50:50	1655.65	1.7511	3.4346	3.7065	44.98
40:60	1662.39	1.7745	3.4041	3.6900	45.48
30:70	1704.66	1.7881	3.2805	3.6224	46.51
20:80	1711.99	1.7907	3.2618	3.6120	46.67
10:90	1732.92	1.8169	3.1759	3.5642	47.65
00:100	1743.38	1.8880	3.0379	3.4859	49.66

From the above observed data, it is clear that the variation of velocity through the mixtures depends upon the value of intermolecular free length (L_f) as suggested by the Eyring and Kincaid [10] according to which the ultrasonic velocity increases/decreases with decreases/increases in the free length, as in the present investigation. The ultrasonic velocity increases whereas the free length decreases and vice versa [11]. Presence of an ion alters the intermolecular free length. There fore, ultrasonic velocity of a solution will be different from that of the solvent. The Fig.1 shows the variation of velocity of LiOH-PEG (400) binary solution at different concentration. It is seen that the velocity increases initially with the increase of concentration and it shows some dips at a composition of 60:40 in the mixture of LiOH (0.5N) and PEG (400). This non-linear variation of velocity indicates the complex formation between the constituents of the mixture [12]. Similar non-linear variation were observed at higher concentration of LiOH with PEG (400) solution.

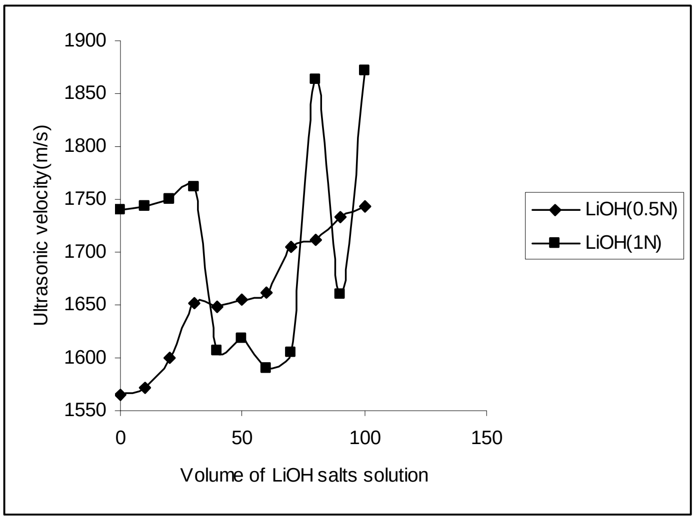

Fig.1 Variation of ultrasonic velocity with concentration of Lithium hydroxide solution in the LiOH-PEG (400) binary mixture

The variations are observed for the LiBr-PEG (400) solution at different concentrations. The linear variations are observed at lower concentration and a non-linear variation are observed at higher concentration of LiBr salts solution.(Fig.2) Hence it may conclude that, some complex molecules are formed at a particular composition of binary mixtures. The above conclusions are similar to the one drawn by Ragouramane and Srinivasa Rao[13]. A similar effect was also observed in solutions of aqueous uranyl chloride, nitrate, strontium iodide, lead acetate and in aqueous solutions of ethylene glycol [14-15].

Table 2 Experimental values of ultrasonic velocity (U), acoustic impedance (Z), adiabatic compressibility (β_{ad}), molecular free length(L_f) and surface tension of PEG (400) and Lithium hydroxide binary mixtures at higher concentration

PEG(400):LiOH	Ultrasonic Velocity (U) ms^{-1}	Acoustic Impedance (Z)×10^{6} kgm^{-2}s^{-1}	Adiabatic Compressibility (β_{ad})10^{-10} kg^{-1}ms^{2}	Molecular Free length (L_f) 10^{-10}m	Surface Tension (σ) Nm^{-1}
100:00	1740.41	1.785	3.222	3.590	46.81
90:10	1742.63	1.772	3.232	3.592	46.58
80:20	1750.20	1.612	3.983	3.991	40.12
70:30	1762.20	1.826	3.102	3.523	48.35
60:40	1606.10	1.660	3.722	3.865	42.25
50:50	1618.90	1.652	3.735	3.864	41.91
40:60	1590.40	1.645	3.808	3.902	41.92
30:70	1604.80	1.556	4.015	4.021	39.14
20:80	1863.20	1.872	2.862	3.385	50.99
10:90	1659.40	1.643	3.665	3.824	42.18
00:100	1872.33	1.843	2.887	3.392	50.47

Acoustic impedance (Z) was calculated using the formula

$$(Z) = (U\rho) \text{ --(1)}$$

The acoustic impedance (Z) also shows the same trend of relative association. The variation of acoustic impedance with concentration is shown in Table 1-4. Acoustic impedance is directly proportional to ultrasonic velocity and it is reciprocal of adiabatic compressibility. In this present study, It increases with concentration of lithium hydroxide and lithium bromide salts solution. The linear variation of acoustic impedance with concentration confirms the presence of molecular association between the solute-solvent molecules. The sudden decrease in impedance at a particular composition may be due to the complex formation in the solution and this may be on the basis of the interaction between the solute and solvent molecules [16]. Such in increasing trends of impedance further support the possibility of molecular interaction between the solute-solvent molecules.

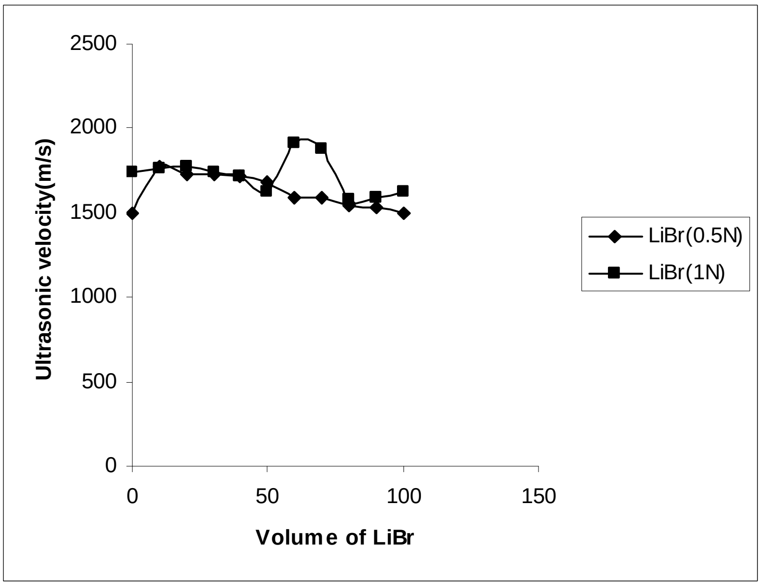

Fig.2 Variation of ultrasonic velocity with concentration of Lithium bromide solution in the LiBr-PEG (400) binary mixture

Table 3 Experimental values of ultrasonic velocity (U), acoustic impedance (Z), adiabatic compressibility (β_{ad}), molecular free length (L_f) and surface tension of PEG (400) and Lithium bromide binary mixtures at lower concentration.

PEG(400):LiBr	Ultrasonic Velocity (U) ms^{-1}	Acoustic Impedance $(Z)\times10^{6}$ $kgm^{-2}s^{-1}$	Adiabatic Compressibility $(\beta_{ad})10^{-10}$ $kg^{-1}ms^{2}$	Molecular Free length (L_f) $10^{-10}m$	Surface Tension (σ) Nm^{-1}
100:00	1497.75	1.5329	4.3502	4.1721	37.37
90:10	1771.30	1.9386	2.9154	3.4135	51.40
80:20	1727.40	1.8958	3.0578	3.4924	49.64
70:30	1726.60	1.8940	3.0510	3.4910	49.58
60:40	1718.88	1.8546	3.1374	3.5407	48.44
50:50	1678.16	1.8140	3.2865	3.6221	46.81
40:60	1591.97	1.7010	3.6921	3.8456	42.75
30:70	1585.30	1.7160	3.6745	3.8334	43.04
20:80	1548.00	1.6532	3.9042	3.9520	40.97
10:90	1531.11	1.6099	4.0265	4.0212	39.68
00:100	1503.00	1.4864	4.2321	4.2310	36.30

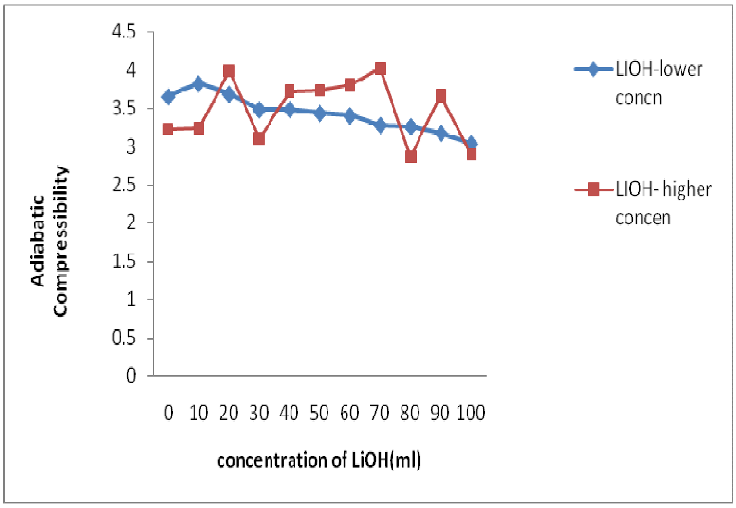

Fig.3 Variation of adiabatic compressibility with concentration of Lithium Hydroxide solution in the LiOH-PEG (400) binary mixture

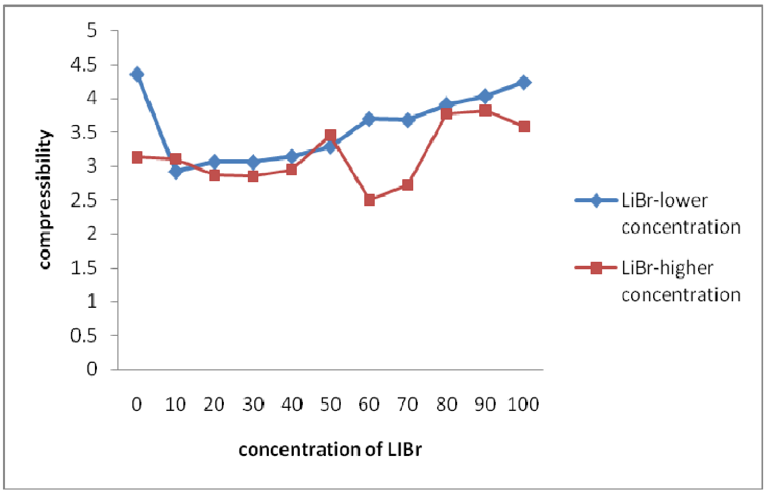

Fig.4 Variation of adiabatic compressibility with concentration of Lithium Bromide solution in the LiBr-PEG (400) binary mixture

The actual deviation depends on the resultant effect. The observed decrease/increase in adiabatic compressibility, intermolecular free length, acoustic impedance and relative association with composition is an evidence of significant interaction between the component molecules in the binary mixtures. Adiabatic compressibility (β_{ad}) of the solution was calculated using the formula-

$$(\beta_{ad})=1/(U^2/\rho) \text{ ---------------------------------------(2)}$$

Where U is the ultrasonic velocity and ρ is the density of the solution. The variation of adiabatic compressibility is shown in Table 1-4. Adiabatic compressibility decreases with increase in the concentration.

Linear variations are observed only at the lower concentration of LiOH and LiBr salts (Fig.3-4) solution and the non linear variations are observed at higher concentrations. It attains a sudden increase at a particular composition and it may be due to departure of solvent molecules around the ions. It means, ion-solvent interaction increases at a given composition. This is again supports the complex formation in the solution at a particular compositions [17].

Table 4 Experimental values of ultrasonic velocity (U), acoustic impedance (Z), adiabatic compressibility (β_{ad}), molecular free length (L_f) and surface tension of PEG (400) and Lithium bromide binary mixtures at higher concentration

PEG(400):LiBr	Ultrasonic Velocity (U) ms^{-1}	Acoustic Impedance $(Z)\times10^6$ $kgm^{-2}s^{-1}$	Adiabatic Compressibility $(\beta_{ad})10^{-10}$ $kg^{-1}ms^2$	Molecular Free length (L_f) $10^{-10}m$	Surface Tension (σ) Nm^{-1}
100:00	1741.56	1.835	3.132	3.531	48.22
90:10	1757.10	1.832	3.105	3.520	48.22
80:20	1776.45	1.991	2.872	3.392	52.02
70:30	1735.61	2.012	2.853	3.371	52.94
60:40	1716.41	1.975	2.945	3.433	51.65
50:50	1619.07	1.784	3.452	3.712	45.31
40:60	1908.21	2.086	2.510	3.171	57.34
30:70	1874.98	1.954	2.731	3.304	53.24
20:80	1576.44	1.672	3.772	3.880	42.00
10:90	1584.52	1.652	3.813	3.902	41.52
00:100	1628.18	1.711	3.584	3.781	43.57

Intermolecular free length (L) were calculated using the formula

$$(L_f) = K\sqrt{\beta ad} \qquad \text{------------------------------------ (3)}$$

Where K is the Jacobson's constant, βad is the adiabatic compressibility of the mixed solution. Intermolecular free length (L_f) decreases with the increase in concentration of the solution. It may be stated that, density and free length are inversely related. The intermolecular free length is decrease with increase in concentration. The inter molecular free length has an inverse behavior. The structural changes are also found to affect the variation of inter molecular free length. An increase in free length produces decrease in ultrasonic velocity. Intermolecular free length (L_f) decreases with the increase in concentration. Table 1-4 shows the variation of free length with concentration of

LiOH and LiBr. It shows linear decrease at lower concentration and non linear variation at higher concentration of LiOH. Similarly, it increase in lower concentration and it shows a non linear variations at higher concentration LiBr. The sudden increase in free length with decrease in velocity at a particular concentration indicates the significant interaction between the solute and molecules due to which structural arrangements of molecules are considerably affected [18-20]. Acoustical parameters are also affecting the surface tension of the binary solution.

Computation of non-linear parameter using B/A ratio

In the last few years [21], a number of theoretical methods have been proposed for estimating the non-linearity parameter (B/A) for pure liquids and liquid mixtures. This parameter has been further correlated with other thermo acoustical parameters, which are used to deduce the available volume and intermolecular free-length of liquid mixtures. Due to the increasing importance of B/A during recent years, an attempt has been made to evaluate B/A of binary mixtures using Hartmann relation and Ballou empirical relation. A comparative study of B/A values obtained from these relations has also been made in order to review the above-mentioned approaches. Furthermore, the results are discussed in terms of the existence of correlation of molecular orientations between the molecules in binary mixtures. General formulation for non-linearity parameter in terms of the acoustical parameters of liquids has been made using the experiment for the sound velocity (U) and introducing the contribution due to acoustic parameters (K) and isothermal acoustic parameter (K”).The expression for B/A has been expressed as[22],

$B/A = 2K+2\gamma K''$

Computations of K and K” require only the knowledge of thermal expansion coefficient, α. Detailed method of calculation is given.

Hartmann and Balizer obtained the following relation for B/A;

$$B/A = 2+\frac{0.98\times10^{4}}{U} \text{ WhereUisinms}^{-1} \text{———————} (4)$$

Empirical relation proposed by Ballou is given [11-12] by

$$B/A = -0.5+\frac{1.2\times10^{4}}{U} \quad \text{-----------------------------} (5)$$

The B/A values for the liquids have been interpreted as the quantity representing the magnitude of the hardness of liquids. The B/A values are concenerned with interactions between the components of the binary systems. The interaction between the components of the binary mixtures is stronger at higher concentration, while it is weaker at lower concentration of salts. The behavior of binary liquid mixtures can be explained in term of 1) physical forces - dispersion 2) chemical forces – dipole-dipole interaction. The former factor increases the intermolecular free length as described by Jacobson. This is turn, causes negative deviation in sound speed and positive deviation in compressibility. On the other hand, the latter factor decreases the intermolecular path lengths leading to a positive deviation in sound speed and negative deviation in compressibility [23]. The ultrasonic velocity is increases, with concentration of LiOH up to a composition 70:30, while at 60:40, it shows a dip. The ultrasonic velocity is again increase with concentration of LiOH. The increase in velocity and decrease in free length shows solvent-solute interactions while the ultrasonic velocity shows a non linear variation at higher concentration of lithium hydroxide.

Table 5 Computation of non-linearity (B/A) parameter of Lithium Hydroxide and Lithium Bromide in PEG (400) electrolyte Solution by Hartmann and Ballizer relation

Concentration of Lithium salts(ml)	Computing of B/A values by Hartman and Ballizer Relation			
	LiOH at Lower concentration	LiOH at Higher concentration	LiBr at Lower concentration	LiBr at Higher concentration
0	8.2603	7.6309	8.5431	7.6271
10	8.2331	7.62237	7.5327	7.5774
20	8.1226	7.5994	7.6732	7.5166
30	7.9315	7.5612	7.6759	7.6464
40	7.9475	8.1017	7.7014	7.7096
50	7.9191	8.0535	7.8397	8.0529
60	7.8951	8.1620	8.1559	7.1357
70	7.7489	8.1067	8.1818	7.2267
80	7.7243	7.2598	8.3307	8.2165
90	7.6552	7.9058	8.4006	8.1848
100	7.6213	7.2341	8.5203	8.1090

Polar molecules can interact with non-polar molecules thro dipole-induced dipole-dipole interactions. When the salts are dissolved in water, the ions are split into Li+, and OH- ions and are strongly banded with water molecules. Similarly, polyethylene glycol is added with water molecules, it enhances the disruption of water structure. It produces electrostatic effects of polymer on the surrounding solvent molecules. In the pure PEG (400) solution, the velocity is low. When, the PEG (400) mixed with lithium salts electrolyte solution, the velocity is increases.

Table 6 Computation of non-linearity (B/A) parameter of Lithium Hydroxide and Lithium Bromide in PEG(400) electrolyte Solution by Hartmann and Ballizer relation

Concentration of Lithium salts(ml)	Computing of B/A values by Ballou Empirical relation			
	LiOH at Lower concentration	LiOH at Higher concentration	LiBr at Lower concentration	LiBr at Higher concentration
0	7.1656	6.3949	7.5120	6.3904
10	7.1323	6.3861	6.2747	6.3294
20	6.9971	6.3564	6.4469	6.2551
30	6.7630	6.3096	6.4500	6.4140
40	6.7826	6.9715	6.4813	6.4913
50	6.7479	6.9124	6.6507	6.9117
60	6.7185	7.0453	7.0378	5.7886
70	6.595	6.9776	7.0696	5.900
80	6.5094	5.9405	7.2519	7.1121
90	6.4247	6.7315	7.3375	7.0732
100	6.3832	5.9091	7.4840	6.8702

Addition of Li salts may increase the interaction between Li+, OH$^-$ and Br $^-$ ion and PEG molecules. In the case of mixed electrolytes, it favored to form more transient lines with dimensional stable morphology due to large cation-anion association effects. Due to the dispersion of ions, there is a strong interaction between solute and solvent molecules and it may be due to hydrogen bonding between the solute and solvent molecules. But at a particular composition, a sudden drop in ultrasonic velocity between the solvent and solute molecules, it may be due to a complex formation. At higher concentration of Li salts electrolytes, more number of ions are formed and non-linear variation were observed due to the structure breaking property of PEG. It may be due to weak interaction between the molecules. The B/A values for the liquids have been interpreted as the quantity representing the magnitude of the hardness of liquids. The B/A values are concenerned with interactions between the components of the binary systems. The interaction between the components of the binary mixtures is stronger at higher concentration, while it is weaker at lower concentration of salts. Table 5-6 shows the variations of the B/A values calculated from Hartmann and Ballou relation and it shows decreased trend with increase in concentration. This data of velocity and densities were taken for computing the various quantities employed in the equation for B/A. The B/A values for the liquids have been interpreted as the quantity representing the magnitude of the hardness of liquids. The B/A values are concenerned with interactions between the components of the binary systems. It is evident from eq. (4) and (5), that Ballous and Hartmann relations depend mainly on the ultrasonic velocity. The interaction between the components of the binary mixtures is stronger at higher concentration, while it is weaker at lower concentration of salts and the accuracy of both the methods limits the usefulness of direct application of these methods to liquid mixture investigation

CONCLUSION

In the present investigation, the measurement of ultrasonic velocity and other acoustical parameters namely adiabatic compressibility, intermolecular free length, acoustic impedance and surface tension have been calculated for Lithium hydroxide and Lithium bromide salts in polyethylene glycol (400) electrolyte solution in different concentrations and it is evident that the presence of ion-solvent interactions leads to strong interactions at lower concentrations and weak interactions at higher concentration of lithium salts of electrolyte solution and consequently promotes the structure breaking characteristics of the aqueous electrolytic solutions. In conclusion, the concentration, nature of the solvent, nature of the solute and its potion play an important role in determining the interactions occurring in the solutions. These conclusions have given scope for further studies on the thermodynamic properties for the system. . On the basis of the discussion, it can be concluded that the variation of computed values of B/A will depends on the ultrasonic sound wave in a given medium which helps in the interpretation of the magnitude of the hardness of the liquid in terms of the non-linearity parameter.

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