

# Ultrasonic Velocity and Allied Parameters of Uni-Univalent Mixed Electrolytes in Aqueous Solvent

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## **Abstract:**

**Aim:** Ultrasonic velocities of aqueous solutions of uni-univalent mixed electrolytes, have been measured at a constant frequency of 10 M Hz at 298.15 K. From the ultrasonic velocity( $u$ ) and density( $\rho$ ) data of the aqueous solutions of these mixed electrolyte systems, the values of acoustic parameters - viz; adiabatic compressibility ( $\beta_{ad}$ ), apparent molar compressibility ( $\Phi_k$ ), inter molecular free length ( $L_f$ ), specific acoustic impedance ( $Z$ ), hydration number ( $H_n$ ) and relative association constant ( $R_A$ ) have been obtained. The results of the study have been discussed in terms of ion-solvent and ion - ion interaction.

**Keywords :** *ultrasonic velocities, inter-molecular free length, adiabatic compressibility, apparent molar compressibility, specific acoustic impedance and hydration number.*

**Methodology:** The densities ( $\rho$ ), ultrasonic velocities ( $u$ ) and other acoustic parameters of aqueous solutions of the mixed uni-univalent electrolyte systems (keeping  $K^+$  one cation common and KI salt in all the two mixed electrolyte systems) :  $KBr + KI$ ,  $KNO_3 + KI$ , have been determined as a function of fraction of ionic strength ( $y$ ) due to the first electrolyte in the mixture of two electrolytes of constant ionic strength,  $\mu = 1.0$  and  $\mu = 2.0$ .

## I. INTRODUCTION

Mixed electrolytes are very important as they are found in numerous processes in chemical industry. They occur in enormous quantities in the water of ocean and play an important role in the physiological process of body fluids and cell equilibria.

Ultrasonic velocity and absorption measurements in aqueous solutions of electrolyte provide useful information on the structural, physical and chemical aspects of the solution [1,2]. Application of acoustic methods have showed extensive possibility in the fields of solution chemistry, physical chemistry, biochemistry, chemical engineering and process control for a fundamental understanding of many phenomena in solutions and liquid systems [3,4].

It is also possible to estimate the hydration numbers of the electrolyte solutions from the molar compressibility value

at infinite dilution. Metal ions play an important role in chemical and biological systems so the hydration of ions in these systems is a key issue to understand the chemical and dynamical processes [5,6]. Propagation of the sound wave perturbs the equilibrium between solute and solvent molecules and relaxation process are setup leading to excess absorption of the sound wave. Endo et al. [7] studied the ultrasonic velocity of electrolytes of high solubility and showed a decrease in the absorption with increase in concentration. Ravichandran et al. [8] determined ultrasonic velocity of aqueous solutions of  $NH_4Cl$ ,  $NH_4H_2PO_4$ ,  $(NH_4)_2C_2O_4$  at temperature of 303 K and concluded that ultrasonic velocity increases in a non-linear manner. Ultrasonic velocity parameters were used to study interactions in Binary [8-9] and ternary [10] systems, as well as the ion-solvent and inter ionic interaction in aqueous [11], mixed aqueous [12] and non aqueous [13] systems. Bhat and Shiva Kumar [14] studied on acoustic behaviour of potassium thiocyanate in aqueous and various non-aqueous solvents and determined the values of apparent molar compressibility ( $\Phi_k$ ), apparent molar volume ( $\Phi_v$ ) and limiting apparent molar compressibility ( $\Phi_k^0$ ), limiting apparent molar volume ( $\Phi_v^0$ ). The ion - ion and ion- solvent interactions have been discussed in terms of these parameters. Recently Palani and Jayachitra [15] have reported ultrasonic study of ternary electrolyte mixture ( $KCl + MgSO_4 + HgCl_2$ ) at different molalities and temperature with a view to ascertaining the nature of ion-ion and ion-solvent interactions.

The review of literature on acoustical studies of solutions reveal that the ultrasonic velocity data as such do not provide significant information about the nature and relative strength of various types of inter molecular and inter- ionic interactions between components. Hence in present study, their derived parameters of aqueous solutions of mixed electrolytes such as adiabatic compressibility ( $\beta_{ad}$ ), apparent molar compressibility ( $\Phi_k$ ), inter molecular free length ( $L_f$ ), specific acoustic impedance ( $Z$ ), hydration number ( $H_n$ ) and relative association constant ( $R_A$ ) have been calculated to more emphasis on such interactions.

With this aim in view, ultrasonic studies of aqueous solutions of mixed electrolytes have been undertaken in the light of the following aspects:

Determination of ultrasonic velocities at a constant frequency of 10 M Hz and a constant temperature 298.15 K, of the following uni-uni valent mixed electrolytes in aqueous mixture KBr + KI, KNO<sub>3</sub> + KI.

Determination of the values of various acoustic parameters, viz; Adiabatic Compressibility ( $\beta_{ad}$ ), Apparent Molar Compressibility ( $\Phi_k$ ), Inter Molecular Free Length ( $L_f$ ), Specific Acoustic Impedance (Z), hydration Number ( $H_n$ ) and Relative Association Constant ( $R_A$ ).

Ascertaining the nature of ion-solvent and ion-ion interactions in terms of acoustic parameters.

## II. EXPERIMENTAL DETAILS

All the chemicals used were of AR grade and were used as such without further purification. The digital electronic balance (Mettler) with accuracy  $\pm 0.1$  mg, was used for the mass determination. The ultrasonic velocity measurements were recorded on a multi-frequency ultrasonic interferometer (M-83; Mittal Enterprises, New Delhi) at 298.15 K by using a cell of 10 MHz frequency. The solutions of electrolytes were prepared in double distilled water (conductance is  $1 \times 10^{-6}$  ohm<sup>-1</sup>cm<sup>-1</sup>). The densities of solvent (water) and solutions were measured with a 5 ml pycnometer.

The various acoustic parameters were determined as below:

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

$$\Phi_k = 1000 / C \cdot \rho_0 (\beta_{ad} \rho_0 - \beta_{ad} \rho) + M \cdot \beta_{ad} / \rho_0 \quad \text{-----} \quad (2)$$

$$L_f = K (\beta_{ad})^{1/2} \quad \text{-----} \quad (3)$$

$$Z = u \cdot \rho \quad \text{-----} \quad (4)$$

$$R_A = \rho / \rho_0 \cdot (u_0 / u)^{1/3} \quad \text{-----} \quad (5)$$

$$H_n = n_1 / n_2 [1 - \beta_{ad} / \beta_{ad}^0] \quad \text{-----} \quad (6)$$

In the above relationships, u and  $\rho$  are the ultrasonic velocity and density of the solution respectively.  $\rho$ ,  $\rho_0$  and  $\beta_{ad}$ ,  $\beta_{ad}^0$  are the density and adiabatic compressibility of the solution and solvent respectively. M is the molecular

weight of the solute and C is the molar concentration of the solution. K is the Jacobson constant (temperature dependent constant), Z is the acoustic impedance,  $H_n$  is hydration number,  $n_1$  and  $n_2$  are the no. of moles of solvent and solute respectively.

## III. RESULTS AND DISCUSSION

The densities ( $\rho$ ), ultrasonic velocities (u) and other acoustic parameters of aqueous solutions of the mixed uni-univalent electrolyte systems (keeping KI one salt common in all the two mixed electrolyte systems): KBr + KI, KNO<sub>3</sub> + KI, have been determined as a function of fraction of ionic strength (y) due to the first electrolyte in the mixture of two electrolytes of constant ionic strength,  $\mu = 1.0$  and  $\mu = 2.0$ . The data's have been presented in Table 1 and 2.

Table 1& 2 shows that the ultrasonic velocity (u) increases with y in the following mixed electrolyte systems: KBr + KI, KNO<sub>3</sub> + KI. The decrease in ultrasonic velocity (u) with y may be due [9] the structure breaking properties of the second electrolyte in these mixed systems.

The adiabatic compressibility ( $\beta_{ad}$ ) (Table 1 and 2) of all the uni-univalent mixed electrolyte systems is found to be less than that of pure solvent which is in agreement with the previous workers [14]. Adiabatic compressibility,  $\beta_{ad}$  is a measure of intermolecular association or dissociation. It also determines the orientation of the solvent molecules around the solute molecules. Further, it is seen that the  $\beta_{ad}$  of the mixed systems: KBr + KI, KNO<sub>3</sub> + KI,  $\beta_{ad}$  decreases with y. The decrease in  $\beta_{ad}$  with y may be attributed [14,15] to the structure making or breaking properties of the second electrolyte in these mixed systems. Ultrasonic waves are high frequency mechanical waves. Their velocities in medium depend inversely on compressibility of the medium reported by Hykes et al [16]. The rapid decrease of adiabatic compressibility, with decrease of second electrolyte concentration, clearly indicates the formation of a large number of tightly bound systems. Since the velocity increases with decrease in concentration of second electrolyte and the density does so, the compressibility must decrease with decrease in concentration. Such reduction in compressibility has been found in the solution due to solute molecules. The decreased compressibility brings the molecules to a closer packing resulting in a decrease of intermolecular free length.

The inter molecular free length ( $L_f$ ) (Table 1 and 2) decreases with increase in ultrasonic velocity (u) of all the mixed electrolyte systems. This conclusion is supported by the earlier studies [15]. At lower concentration of KI in

the mixture of KBr + KI, KNO<sub>3</sub> + KI, the molecules are not closer and thus inter molecular free length,  $L_f$  is high. But as the concentration of KI in mixed aqueous solution decreases, the molecules come closer and solvent- solvent interaction will exist, thereby decreasing the  $L_f$  and hence internal pressure decreases.

A continuous decrease in  $\beta_{ad}$  or  $L_f$  is a clear evidence for the existence of strong interactions. Such strong interaction may be due to dipole- dipole, ion - dipole, H-bonding, etc

The acoustic impedance ( $Z$ ) (Table 1 and 2) increases with ionic strength of first electrolyte ( $y$ ) in the following mixed electrolyte systems: KBr + KI, KNO<sub>3</sub> + KI, mixed electrolyte systems the acoustic impedance increases with  $y$ . The increase in the value of acoustic impedance ( $Z$ ) with  $y$  supports [21] the structure making property of the first electrolyte in these mixed systems. The values of acoustic impedance,  $Z$  increase with decreasing KI concentration are in agreement with the results of Mehrotra [17,18], which indicates that there is a significant interaction between ion- ion and ion- solvent molecules which considerably affect the structural arrangement. The increase in the values of specific acoustic impedance,  $Z$  with decreasing KI concentration, can be explained on the basis of hydrophilic interaction between the solute – solute and solute- solvent molecules, which increases the intermolecular distance, making relatively wider gaps between the molecules and becoming the main cause of impedance in the propagation of ultrasonic waves.

Table 1 and 2 shows that in the mixed electrolyte systems: KBr + KI, KNO<sub>3</sub> + KI, the relative association constant ( $R_A$ ) decreases with  $y$  (at ionic strength  $\mu = 1.0$  and  $2.0$ ), signifies [14] a decrease in the solvation of the first electrolyte in these mixed systems.

The apparent molar compressibility ( $\Phi_k$ ) has been calculated from ultrasonic velocity ( $u$ ) and density ( $\rho$ ) data (at ionic strength  $\mu = 1.0$  and  $2.0$ ). For a single electrolyte system the concentration-dependence of the apparent molar compressibility ( $\Phi_k$ ) is given by the following relation [19]

$$\Phi_k = \Phi_k^0 + S_k \Phi_c$$

From the above relation it follows that the variation of  $\Phi_k$  with  $\Phi_c$  must be linear. However in the present study of uni-univalent mixed electrolyte systems, the plots of  $\Phi_k$  versus  $y$  are found to be linear. From this it follows that the variation of  $\Phi_k$  with  $y$  is linear and governed by the following relation

$$\Phi_k = \Phi_k^0 + S_k \cdot y$$

A perusal of Table 1 & 2 shows that the values of  $\Phi_k$  are negative in the following mixed electrolyte systems: KBr + KI, KNO<sub>3</sub> + KI, which signify[20] the loss of compressibility of the surrounding water molecules due to strong electrostrictive forces in the vicinity of ions of mixed electrolyte systems causing electrostrictive solvation of the electrolytes.

A perusal of Table 1 and 2 shows that for all the mixed electrolyte systems containing K<sup>+</sup> the values of  $H_n$ , lies between 3-6. These results are supported by the values of  $S_n$  reported in literature [21]. Further the values of  $H_n$  increases with decrease in the concentration of KI.

## CONCLUSION

A survey of authors` scientific investigations in the field ultrasound velocity measurements in electrolyte solutions and various liquid systems is presented, with the help of these surveys and my research it is concluded that various acoustic parameters; viz. specific acoustic impedance ( $Z$ ), apparent molar compressibility ( $\Phi_k$ ) and hydration number ( $H_n$ ) increase, while adiabatic compressibility ( $\beta_{ad}$ ) inter-molecular free-length ( $L_f$ ) and relative association number ( $R_A$ ) decreases with decrease in concentration of KI, taken in the mixed electrolyte system.

At any given concentration, the different acoustic parameters increases or decreases, this indicates the structure distorting nature of K<sup>+</sup> is balanced and subdued by the presence of different anions consistent with the behavior of anion disrupting the hydrogen bonded structure of water.

Table 1: Densities( $\rho$ ), ultrasonic velocities ( $u$ ) and other acoustic parameters of mixed electrolyte systems in aqueous solution as a function of the fraction of ionic strength( $y$ ) due to the first electrolyte in the mixture of two electrolytes at constant ionic strength,  $\mu= 1.0$  at temperature 298.15 K

<i>KBr + KI Mixed electrolyte system</i>									
Conc. of first electrolyte + second electrolyte	Fraction of ionic strength of first electrolyte ( $y$ )	$\rho$ gm c.c <sup>-1</sup>	$u \cdot 10^{-5}$ cm sec <sup>-1</sup>	$\beta_{ad} \cdot 10^{12}$ cm <sup>2</sup> dyne <sup>-1</sup>	$Z \cdot 10^{-4}$ gm.cm <sup>-1</sup> sec <sup>-1</sup>	$L_f$ Å	$\Phi_k \cdot 10^{11}$ cm <sup>2</sup> mol <sup>-1</sup>	$R_A$	$H_n$
0.0 + 1.0	0	1.104	1.4782	41.45	16.32	0.4024	-92.95	1.11	4.11
0.05 + 0.95	0.05	1.103	1.481	41.33	16.34	0.4018	-111.05	1.11	4.26
0.1 + 0.9	0.1	1.101	1.485	41.17	16.35	0.4010	-128.63	1.11	4.44
0.2 + 0.8	0.2	1.097	1.492	40.95	16.37	0.4000	-153.80	1.10	4.70
0.25 + 0.75	0.25	1.095	1.4972	40.74	16.39	0.3989	-176.38	1.10	4.95
0.3 + 0.7	0.3	1.094	1.5001	40.62	16.41	0.3983	-194.48	1.10	5.09
0.5 + 0.5	0.5	1.088	1.5143	40.08	16.48	0.3957	-263.84	1.09	5.74
0.75 + 0.25	0.75	1.08	1.534	39.35	16.57	0.3921	-353.78	1.07	6.62
0.8 + 0.2	0.8	1.079	1.538	39.18	16.60	0.3912	-376.88	1.07	6.83
0.9 + 0.1	0.9	1.075	1.5446	38.99	16.60	0.3903	-399.04	1.07	7.06
0.95 + 0.05	0.95	1.074	1.5481	38.85	16.63	0.3896	-419.14	1.07	7.23
1.0 + 0.0	1	1.072	1.5528	38.69	16.65	0.3887	-436.72	1.06	7.42
Pure solvent	-----	0.996	1.4936	45.01	-----	-----	-----	-----	-----

*KNO<sub>3</sub> + KI Mixed electrolyte system*

Conc. Of first electrolyte + second electrolyte	Fraction of ionic strength of first electrolyte (y)	$\rho$ gm c.c <sup>-1</sup>	$u \cdot 10^{-5}$ cm sec <sup>-1</sup>	$\beta_{ad} \cdot 10^{12}$ cm <sup>2</sup> dyne <sup>-1</sup>	$Z \cdot 10^{-4}$ gm.cm <sup>-2</sup> sec <sup>-1</sup>	$L_f$ Å	$\Phi_k \cdot 10^{11}$ cm <sup>2</sup> mol <sup>-1</sup>	$R_A$	$H_n$
0.0 + 1.0	0	1.1037	1.4782	41.47	16.31	0.4025	-89.60	1.11	4.09
0.05 + 0.95	0.05	1.1010	1.4809	41.42	16.30	0.4022	-97.08	1.11	4.15
0.1 + 0.9	0.1	1.0987	1.4837	41.35	16.30	0.4019	-108.37	1.11	4.23
0.2 + 0.8	0.2	1.0936	1.4894	41.22	16.29	0.4013	-127.70	1.10	4.39
0.25 + 0.75	0.25	1.0909	1.4934	41.10	16.29	0.4007	-142.18	1.10	4.53
0.3 + 0.7	0.3	1.0882	1.4957	41.08	16.28	0.4006	-146.67	1.09	4.56
0.5 + 0.5	0.5	1.0781	1.5065	40.87	16.24	0.3996	-180.77	1.08	4.82
0.75 + 0.25	0.75	1.643	1.5207	40.63	16.18	0.3984	-215.84	1.06	5.11
0.8 + 0.2	0.8	1.0621	1.5236	40.56	16.18	0.3980	-227.58	1.06	5.20
0.9 + 0.1	0.9	1.0560	1.5293	40.49	16.15	0.3977	-236.39	1.05	5.28
0.95 + 0.05	0.95	1.0533	1.5327	40.41	16.14	0.3973	-246.88	1.05	5.38
1.0 + 0.0	1	1.0515	1.5347	40.38	16.14	0.3971	-256.43	1.05	5.42
Pure solvent	-----	0.996	1.4936	45.01	-----	-----	-----	-----	-----

Table 2: Densities( $\rho$ ), ultrasonic velocities( $u$ ) and other acoustic parameters of mixed electrolyte systems in aqueous solution as a function of the fraction of ionic strength( $y$ ) due to the first electrolyte in the mixture of two electrolytes at constant ionic strength,  $\mu=2.0$  at temperature 298.15 K

<i>KBr + KI Mixed electrolyte system</i>									
Conc. Of first electrolyte + second electrolyte	Fraction of ionic strength of first electrolyte ( $y$ )	$\rho$ gm c.c <sup>-1</sup>	$u \cdot 10^{-5}$ cm sec <sup>-1</sup>	$\beta_{ad} \cdot 10^{12}$ cm <sup>2</sup> dyne <sup>-1</sup>	$Z \cdot 10^{-4}$ gm.cm <sup>2</sup> sec <sup>-1</sup>	$L_f$ Å	$\Phi_k \cdot 10^{11}$ cm <sup>2</sup> mol <sup>-1</sup>	$R_A$	$H_n$
0.0 + 2.0	0	1.210	1.4790	37.78	17.90	0.3842	-94.43	1.22	3.91
0.1 + 1.9	0.1	1.207	1.4826	37.69	17.90	0.3837	-102.77	1.21	3.97
0.2 + 1.8	0.2	1.204	1.4870	37.56	17.90	0.3831	-113.11	1.21	4.05
0.3 + 1.7	0.3	1.200	1.4906	37.51	17.89	0.3828	-117.20	1.21	4.08
0.5 + 1.5	0.5	1.195	1.4968	37.53	17.89	0.3820	-135.14	1.20	4.19
0.8 + 1.2	0.8	1.184	1.5102	37.03	17.88	0.3803	-158.14	1.18	4.38
1.0 + 1.0	1	1.180	1.5191	36.72	17.88	0.3788	-185.84	1.18	4.57
1.2 + 0.8	1.2	1.172	1.5262	36.63	17.87	0.3783	-193.50	1.17	4.63
1.5 + 0.5	1.5	1.163	1.5377	36.36	17.86	0.3769	-218.52	1.16	4.81
1.8 + 0.2	1.8	1.153	1.5480	36.19	17.85	0.3760	-236.28	1.14	4.92
1.9 + 0.1	1.9	1.151	1.5544	35.96	17.84	0.3748	-253.88	1.14	5.07
2.0 + 0.0	2	1.147	1.5577	35.93	17.83	0.3746	-256.96	1.14	5.09
Pure solvent	-----	0.996	1.4936	45.01	-----	-----	-----	-----	-----

<i>KNO<sub>3</sub>+ KI Mixed electrolyte system</i>									
Conc. Of first electrolyte + second electrolyte	Fraction of ionic strength of first electrolyte (y)	$\rho$ gm c.c <sup>-1</sup>	$u \cdot 10^{-5}$ cm sec <sup>-1</sup>	$\beta_{ad} \cdot 10^{12}$ cm <sup>2</sup> dyne <sup>-1</sup>	$Z \cdot 10^{-4}$ gm.cm <sup>-2</sup> sec <sup>-1</sup>	$L_f$ Å	$\Phi_k \cdot 10^{11}$ cm <sup>2</sup> mol <sup>-1</sup>	$R_A$	$H_n$
0.0 + 2.0	0	1.210	1.4793	37.77	17.48	0.3841	-94.93	1.22	3.92
0.1 + 1.9	0.1	1.206	1.4841	37.65	17.51	0.3835	-106.58	1.21	4.00
0.2 + 1.8	0.2	1.2000	1.4900	37.54	17.51	0.3829	-113.21	1.21	4.06
0.3 + 1.7	0.3	1.1970	1.4933	37.46	17.63	0.3825	-125.12	1.20	4.12
0.5 + 1.5	0.5	1.1860	1.5045	37.25	17.65	0.3815	-140.14	1.19	4.24
0.8 + 1.2	0.8	1.1680	1.5198	37.07	17.70	0.3805	-152.52	1.17	4.35
1.0 + 1.0	1	1.1570	1.5300	36.92	17.75	0.3798	-164.54	1.15	4.44
1.2 + 0.8	1.2	1.1470	1.5392	36.80	17.84	0.3791	-177.32	1.14	4.52
1.5 + 0.5	1.5	1.1320	1.5575	36.42	17.87	0.3772	-206.48	1.12	4.75
1.8 + 0.2	1.8	1.1150	1.5708	36.35	17.88	0.3768	-215.63	1.10	4.81
1.9 + 0.1	1.9	1.1110	1.5759	36.24	17.90	0.3763	-226.78	1.10	4.88
2.0 + 0.0	2	1.1060	1.5808	36.18	17.90	0.3759	-233.17	1.09	4.92
Pure solvent	----	0.996	1.4936	45.01	-----	-----	-----	-----	-----

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