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Ultrasonic Studies and Excess Properties of Binary Mixtures of Ethylmethylketone and Chlorobenzene At 308.15K

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ABSTRACT: The binary mixtures of Ethylmethylketone with Chlorobenzene containing different ultrasonic properties have been studied at temperature 308.15k at a fixed frequency of 2 M.Hz. The ultrasonic related like parameters velocity (U), density (ρ), viscosity (η) and computed values of deviation in viscosity ($\Delta\eta$), molar volume (V), excess molar volume (V^E), adiabatic compressibility (β_{ad}), deviation in adiabatic compressibility ($\Delta\beta_{ad}$), intermolecular free length (L_f), excess intermolecular free length (L_f^E), acoustic impedance (Z), excess acoustic impedance (Z^E), excess velocity (U^E), Rao's constant (R) and Wada's constant (W) were determined. The emergence of a new close packed structure in the intermediate composition ranges of binary liquid systems has been observed clearly from these values. Hydrogen bonded intermolecular complexes might be formed between ethyl methyl ketone and the solute molecules having definite stoichiometric proportion of the system. These results are in qualitative agreement with the observations made by earlier investigators.

KEYWORDS: Ultrasonic Velocities, Excess Properties, Thermodynamic Parameters, Acoustic impedance, Intermolecular free length, Wada's and Rao's constants.

1.INTRODUCTION

Properties of liquid-liquid mixtures are thermodynamically very important as a part of studies of thermodynamic, acoustic and transport aspects. The intermolecular forces of liquids in a mixture shows a considerable effect on the physical and chemical properties [1-5]. Ultrasonic measurements are very useful in chemical & food processing; material testing under water ranging and cleaning. These studies are also extensively used in textile industry, leather industry and pharmaceutical industry. In recent years sound velocity measurements are very useful in the technology of polymers to understand polymer solvent and polymer interactions, structure of polymers. The properties of mixing and excess properties are provided to be increasingly useful and it is a very challenging area in thermodynamics and many people are being attracted to it. Modern chemical industry requiring more precise and sophisticated methods of calculation will certainly benefit from these developments.

Thermodynamic and physicochemical studies of binary liquid mixtures are considerable theoretical and technological importance as they provide wide range of solvents of varying compositions and properties. Ultrasonic studies provide a wealth of information about the liquid state. It explains the many of the properties of individual liquids and combination of the liquids. The propagation of ultrasonic waves in liquids and liquid Mixtures provide an information on the nature of the inter molecular and intra molecular interactions in these liquid systems. Extensive studies of ultrasonic velocity in liquids, liquid mixtures and their interpretation in the light of molecular structure has gained much importance during the last three decades [6-10].

Ultrasonic velocity and related data of liquid mixtures are found to be the most powerful tool in testing the theories of the liquid state. In addition to ultrasonic velocity data can be utilized to deduce some useful properties of liquid mixtures which are not easily accessible by other means.

The negative excess values indicate the strong interactions and the positive values show weak interactions between the components liquid. Ultrasonic velocity and densities were reported [11] for the binary liquid mixtures of

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trichloromethane with methyl ethyl ketone, diethyl ketone, cyclo hexane and 1,4-dioxane and excess thermodynamic parameters were discussed in terms of the difference in molecular structure and intermolecular forces between unlike components.

II. RELATED WORK

A wealth of data accumulated for the purpose of this research study has appeared in excellent reviews by Herzfeld and Litovitz [12] Nozdrev [13] and also the volumes edited by Mason [14] and Flugg [15], Bergmann [16] Beyer and Letchar [17]. In all these studies the measured ultrasonic parameters of prime importance are the ultrasonic velocity, density and viscosity which lead to the estimation of derived quantities like compressibility, excess thermodynamic properties like β_{ad}^E , Excess intermolecular free length, excess molar volume, excess free volume, excess internal pressure, excess free energy of activation in liquids and liquid mixtures.

S. Anuradha et.al [18] has measured densities and ultrasonic velocity at 299K for binary mixture of acetonitrile (ACN) with acetone/ethyl methyl ketone/methyl isobutyl and acetophenone over entire composition range. From this centropic compressibility (K_s), intermolecular free length (L_f) and their deviations, excess compressibility (L_E^s) and excess intermolecular free length (L_F^E) have been calculated and interpreted in terms of intermolecular interactions. Excess compressibility, excess volume and excess intermolecular free length in chloro benzene + 2-octanol mixtures were studied by Agrwal and Sarma [19]. The excess values were explained in terms of molecular interactions in nature. The negative excess values indicate the strong interactions and the positive values show weak interactions between the components liquid.

III. EXPERIMENTAL TECHNIQUES

Single crystal Ultrasonic pulse echo Interferometer (model M-82) manufactured by Mittal Enterprises working at 2MHz was used to measure the sound velocity of a liquid or liquid mixture. The ultrasonic velocity had an accuracy ± 0.5 m/s. To determine viscosity of liquid, Ostwald Viscometer is used and a stop watch of accuracy in milli seconds is selected to note the time of flow of the liquid. Specific gravity bottle is employed to determine density of liquid and the weight of the liquid is determined by using single pan electronic balance capable of measuring up to 0.5mg.

In the present study the binary mixtures were prepared by Ethylmethylketone and chlorobenzene with different molefractions. Thermostat of high accuracy is used to maintain constant Temperature throughout the experiment.

The adiabatic compressibility (β_{ad}) has been determined by using the experimentally measured ultrasonic velocity (U) and density (ρ) by using the following formula:

$$\beta_{ad} = \frac{1}{\rho U^2} \quad (2.1)$$

The mean molar volume (V) of a binary liquid mixtures at a given mole fraction is given by:

$$V = \frac{M}{\rho} \quad (2.2)$$

The Excess volume (V^E) at a given mole fraction is the difference between mean molar volume and the average volume is calculated using the given formula :

$$V^E = V - (V_1 X_1 + V_2 X_2) \quad (2.3)$$

Where V_1 and V_2 are the mean molar volumes of pure liquids 1&2 respectively.

The Excess compressibility (β_{ad}^E) at a given mole fraction is the difference between adiabatic compressibility (β_{ad}) and the sum of the fractional contributes of the two liquids is :

$$\beta_{ad}^E = \beta_{ad} - (\beta_{ad} X_1 + \beta_{ad} X_2) \quad (2.4)$$

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Where β_{ad1} and β_{ad2} are the individual β_{ad} values of pure liquid in the binary mixtures at that Temperature. The Rao's (R) constant has been calculated using the given formula:

$$R = V U^{\frac{1}{3}} \quad (2.5)$$

And Wada's Constant (W) also calculated as :

$$W = \frac{M}{\rho \beta_{ad}^{\frac{7}{1}}} \quad (2.6)$$

Inter Molecular free length $L_f(A^0)$ is calculated as follows:

$$L_f = K(\beta_{ad})^{1/2} \quad (2.7)$$

Where K is Jacobson's constant which is temperature dependent parameter.

The excess viscosity (η^E) at a given mole fraction is the difference between viscosity (η) and the sum of the fraction contribution of the two liquids are given by Fort and Moore is:

$$\eta^E = \eta_{mix} - (\eta_1 X_1 + \eta_2 X_2) \quad (2.8)$$

Where η_{max} , η_1 , η_2 are the individual viscosity values of pure liquid in the binary mixture and viscosity of mixture.

The excess mean free length (L_f^E) at a given mole fraction is the difference between mean free length and sum of the fraction contribution of the two liquid are given by:

$$L_f^E = L_{f_{mix}} - (L_{f1} X_1 + L_{f2} X_2) \quad (2.9)$$

Where $L_{f_{mix}}$, L_{f1} and L_{f2} are the individual mean free length values of pure liquid in the binary mixtures and mean free length of mixture. Specific acoustic Impedance (Z) is also calculated using the below relation:

$$Z = \rho U \quad (2.10)$$

Excess acoustic impedance (Z^E) is difference between the ideal acoustic impedance and acoustic impedance of the pure components i.e. $Z^E = Z_{real} - Z_{ideal}$ (2.11)

Finally the Excess velocity (U^E) is difference between the ideal velocity and velocity of the pure components.

$$U^E = U_{real} - U_{ideal} \quad (2.12)$$

A) THEORETICAL CONSIDERATIONS: NOMOTO EQUATION

Rao found experimentally that, for pure liquids, the ratio of temperature coefficients of sound velocity U and molar volume V remains almost constant:

$$[(1/U)(dU/dT)] / [(1/V)(dV/dT)] = -3 \quad (2.13)$$

Where T is the absolute temperature. After Integrating the above equation we obtains the Rao's constant as:

$$VU^{1/3} = \text{constant} = M/\rho U^{1/3} = R \quad (2.14)$$

Where M is molecular weight and ρ is density. The constant R is called the molar sound velocity or Rao's constant. It was found to be additive i.e it can be calculated as a sum of increments from the atoms or atom groups in the molecule and from the chemical bonds. On assuming the additivity of molar sound velocity (R) and no volume change on mixing, Nomoto established the following relation for a liquid mixture is:

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$$R = M/\rho U^{1/3} \quad (2.15)$$

Where U and ρ are determined experimentally and M is the mean molecular weight in a binary liquid mixtures that calculated as:

$$M = (X_1M_1 + X_2M_2) \quad (2.16)$$

Where M_1 and M_2 are molecular weights of constituent components.

Simple manipulation yields the following relation (20):

$$U_{\text{Nomoto}} = [(X_1R_1+X_2R_2) / (X_1V_1+X_2V_2)]^3 \quad (2.17)$$

B) THE VAN DAEL AND VANGEEL EQUATION

The ideal mixing theory advanced by Van Dael and Vangeel in the light of assumptions made by Blandamer and Waddington, yield the following relation for adiabatic compressibility $(\beta_{\text{ad}})_{\text{imx}}$ is:

$$(\beta_{\text{ad}})_{\text{imx}} = \phi_1\gamma_1/\gamma_{\text{imx}} (\beta_{\text{ad}})_1 + \phi_2 \gamma_2 / \gamma_{\text{imx}} (\beta_{\text{ad}})_2 \quad (2.18)$$

Where ϕ_1, ϕ_2 are the volume fraction of species 1 and 2, γ_1 and γ_2 are ratios of specific heats of the respective species. This relation holds good if the mixture is ideal and if $\gamma_1 = \gamma_2 = \gamma_{\text{imx}}$. Using the additional assumption that $V_1 = V_2$ the above equation can be transformed in to a linear combination of mole fraction X_1 and X_2 then,

$$(\beta_{\text{ad}})_{\text{imx}} = X_1(\beta_{\text{ad}})_1 + X_2(\beta_{\text{ad}})_2 \quad (2.19)$$

On the basis of this equation, Van Dael obtained the relation for ultrasonic velocity in liquid mixtures as

$$1/(X_1M_1+X_2M_2)*1/U_{\text{imx}}^2 = X_1/M_1U_1^2 + X_2/M_2U_2^2 \quad (2.20)$$

Where U_{imx} is the ideal mixing ultrasonic velocity in liquid mixture, U_1 and U_2 are ultrasonic velocities in species.

IV. RESULTS AND DISCUSSION

Various types of interactions which are possible and which can operate in the binary liquid mixtures containing ethylmethylketone are hydrogen bonded complex formation, that can produce negative deviation in excess molar volumes, excess intermolecular free length and deviation in adiabatic compressibility. This is may be fact that O-H hydrogen bonded complex formation.

The variation of ultrasonic velocity with the mole fraction of ethylmethylketone decreases with increasing with mole fraction of chlorobenzene are given in the Table 1. Similarly the density, Viscosity, and acoustic impedance are also decreases with increasing mole fraction (X). It is observed that the converse on the behavior of adiabatic compressibility and inter molecular free length from the Table 1. This may be due to self-association of the solvent molecules and very weak dipole- induced dipole interaction between the component molecules. The variation of ultrasonic velocity in a liquid mixture depends upon the increase or decrease of inter molecular free length after mixing the components. On the basis of a model, for sound propagation proposed by Eyring and Kincaid ultrasonic velocity should increase if the intermolecular free length decreases or vice versa as a result of mixing of components. This is in fact that these observations in the present investigations for the liquid system.

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The adiabatic compressibility (β_{ad}) and inter molecular free length (L_f) are also calculated at the Temperature 308.15 K and given in Table 1. The graph is plotted between adiabatic compressibility (β_{ad}), inter molecular free length (L_f) are taken on Y-axes and mole fraction (X) is on X-axis. This variation of adiabatic compressibility and mean free length as a function of mole fraction can be shown in Fig.1. Both are increases with increasing the molefraction. The existence of minimum free length is an indication that, the structural readjustment in the liquid mixture is in the direction of less compressible phase or closer packing of molecules. This idea has been further verified from binary mixture of propylene carbonate with Tetra Hydro Furan, 1, 4 dioxane and acetone by D. S Wankhede et al [21]. These are increasing with mole fractions. The deviation in adiabatic compressibility can be explained by taking into consideration of the loss of dipolar association and different in size and shape of the component molecules, which are lead to decrease in velocity and increase in compressibility.

Excess thermodynamic parameters like excess molar volume, excess adiabatic compressibility, excess intermolecular free length, excess velocity, excess acoustic impedance, deviation in viscosity, Rao, and Wada's constants were calculated and listed in Table 2. The excess molar volume, excess adiabatic compressibility and excess mean free length were shows the similar behavior i.e. these are increases upto X=0.5312 then increases with continuous increasing molefraction (X). But it is converse on excess acoustic impedance, excess velocity, deviation in viscosity which are increases upto X= 0.5312 then decreases with continuous increasing the molefraction (X). Rao's and Wada's constants are gradually decreases with continuous increasing the mole fraction (X). The negative sign due to contractive factors dominate the expansive factors. This is similar to S.Shakal Ahmed et al [22] measured excess value (V^E), viscosity (η) of binary mixtures of 1-butylamine with ethanol heptanol.

The excess volumes of mixing have been evaluated both as a function of composition to examine the possibility of structural adjustment. The results indicate that there is a volume contraction as mixing. This can be attributed mainly to the specific interactions between unlike molecules of the systems. It indicates these systems results are molecular complex formation.

The closer packing of molecules due to hydrogen bonds formation between hetero molecules is bound to have greater influence on the overall compressibility of the systems. In this study of binary liquids mixtures of ethyl methyl ketone +chlorobenzene it was observed that hetero molecular interactions are strong. The deviation in $\Delta\eta$ can be constructed from Table 2 which are positive increased to mole fraction of x= 0.5312 then decreases with continuous increasing mole fraction of X. According to Fort and Moore, these deviations in viscosity needs S become more positive as the strength of interaction increases. The deviation in viscosity gives a qualitative estimation of the strength of inter molecular interactions.

The increase in deviation in viscosity may be due to specific interactions between unlike components such as hydrogen bond formation and charge transfer complex and the decrease in deviations may be due to the difference in size and shape of the component molecules and the loss of dipolar association in pure component may be contribute. The similar studies were carried for binary mixtures of tetra hydroforum with methanol by Shahla Parveen et al [23].

The molar excess volume (V_m^E) values are decreasing up to mole fraction X = 0.5312 from where increases with continuous increasing of mole fraction. V_m^E indicates that the association between the unlike molecules predominates over dissociation of ethyl methyl ketone with chloro benzene. The results were interpreted by SheoPrakash et al [24]. The negative value of V_m^E for some liquid systems are explained by Prigogine [25] in terms of different size of molecules of dipole-dipole interactions between them.

The molar sound velocity (R) and molar compressibility (w) are the two important parameters, which are widely used for the study of physico-chemical behavior of liquid and liquid mixtures. The parameters (R) and (w) are called Rao's constant and Wada's constant. Fig.(2) shows the graph between Rao's constant and Wada's constant as a function of mole fraction (X). Which shows the similar behavior i.e. both are decreases with increasing molefraction (X). The content of positive deviations in U over the entire composition range for all the three systems studied and suggested that significant interactions are positive in these mixtures. Similar studies were observed in binary liquid mixtures namely, carbon tetra chloride by R. Kumar et al [26]. From the Table 2 it is clear that, Z^E and U^E values are positive deviations indicate the increase strength of interactions between component molecules of liquid mixtures. The above trends are in support of the

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finds for the variation of $\Delta\beta_{ad}$ and L_f^E with compositions. This is due to the strong interaction through formation of hydrogen bonding between the components [27-30]. Theoretical calculation of ultrasonic velocity using Nomoto relation followed by VamDaemixing, ideal mixing ultrasonic velocity, percentage of deviation in Nomoto relation, percentage of deviation in ideal mixing, ratio of Nomoto relation to ideal mixing were calculated and listed in Table 3. From the Table, it reveals that theoretical values are agree good with experimental values.

Table-1: Variation of ultrasonic Velocity, density, Viscosity, molar Volume, adiabatic compressibility, mean free length and acoustic impedance of Ethylmethylketone with Chlorobenzene mixtures at Temperature 308.15K with mole fraction

Mole fraction X	U m/s	ρ gm/cc	η cp	V $\text{cm}^3\text{mol}^{-1}$	$\beta_{ad} \times 10^{12} \text{m}^2\text{N}^{-1}$	$L_f 10^{-10}\text{m}$	Z
0.0000	1233	1.0905	0.7512	103.22	60.32	0.4905	1344.59
0.1847	1224	1.0488	0.6865	100.20	63.64	0.5038	1283.72
0.3616	1216	1.0049	0.6217	97.45	67.30	0.5180	1222.00
0.5312	1207	0.9574	0.5557	95.12	71.69	0.5347	1155.63
0.6938	1186	0.9026	0.4854	93.61	78.72	0.5603	1070.79
0.8500	1167	0.8457	0.4145	92.44	86.82	0.5884	986.99
1.0000	1155	0.7888	0.3441	91.42	95.03	0.6156	911.06

Table-2: Variation of excess molar volume, excess adiabatic compressibility, deviation in viscosity, excess intermolecular free length, excess velocity, excess acoustic impedance, Rao's and Wada's constant of Ethylmethylketone with Chlorobenzene mixtures at Temperature 308.15K with mole fraction

Mole fraction X	$V^E \text{cm}^3\text{mol}^{-1}$	$\Delta\beta_{ad} 10^{12} \text{m}^2\text{N}^{-1}$	$\Delta\eta$ cp	$L_f^E 10^{-10}\text{m}$	Z^E	U^E m/s	R	W
0.0000	0.0000	0.0000	0.0000	0.0000	0.00	0.00	5137	2976
0.1847	-0.8400	-3.0885	0.0105	-0.0098	19.22	5.41	4975	2867
0.3616	-1.5000	-5.5750	0.0177	-0.0177	34.19	11.21	4828	2766
0.5312	-1.8277	-7.0647	0.0208	-0.0222	41.32	15.43	4701	2676
0.6938	-1.4200	-5.6800	0.0167	-0.0170	26.98	7.42	4600	2599
0.8500	-0.7500	-3.0037	0.0093	-0.0084	10.88	0.30	4517	2530
1.0000	0.0000	0.0000	0.0000	0.0000	0.00	0.00	4452	2470

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Table 3: Ultrasonic velocity(expt.), Ideal mixing Ultrasonic velocity, Nomoto relation values (theo.), deviations in Nomoto, deviation in ideal mixing, ratio of Nomoto to ideal mixing values of ethylmethylketone with Chlorobenzene mixtures at 308.15K

Mole fraction X	U_{exp} ms ⁻¹	U_{Nomoto} ms ⁻¹	U_{imx} ms ⁻¹	% U_N	% U_{imx}	U^2/U^2_{imx}
0.0000	1233.00	1233.00	1233.00	0.000	0.0000	1.0000
0.1847	1224.00	1219.72	1219.72	-0.3493	-2.5221	1.0524
0.3616	1216.00	1206.56	1206.56	-0.7761	-3.9781	1.0846
0.5312	1207.00	1193.51	1193.51	-1.1177	-4.4859	1.0961
0.6938	1186.30	1180.57	1180.57	-0.4837	-3.3449	1.0704
0.8500	1167.00	1167.73	1167.73	0.0626	-1.6625	1.0341
1.0000	1155.00	1155.00	1155.00	0.000	0.0000	1.0000

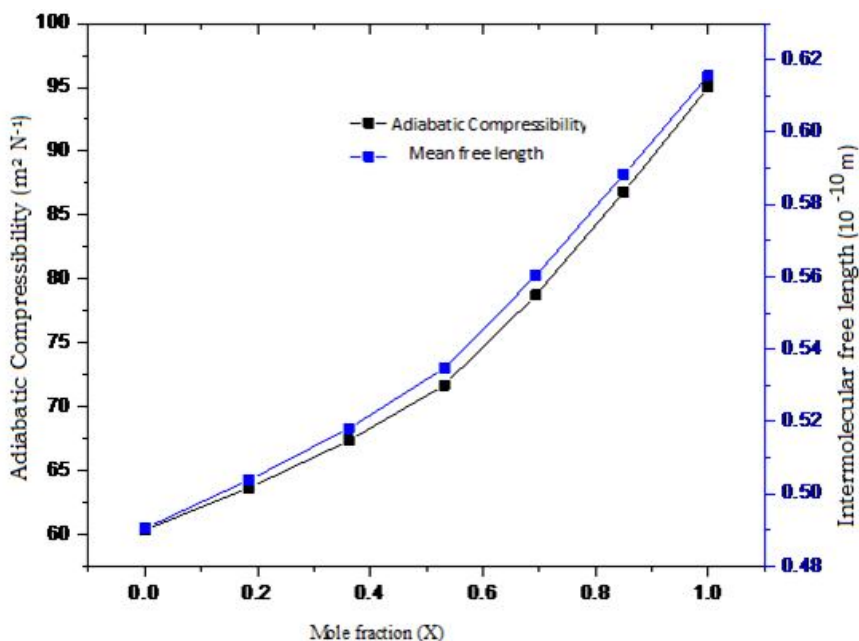


Fig. 1. Variation of adiabatic compressibility and intermolecular free length with mole fraction of Ethylmethylketone with Chlorobenzene at Temperature 308.15K

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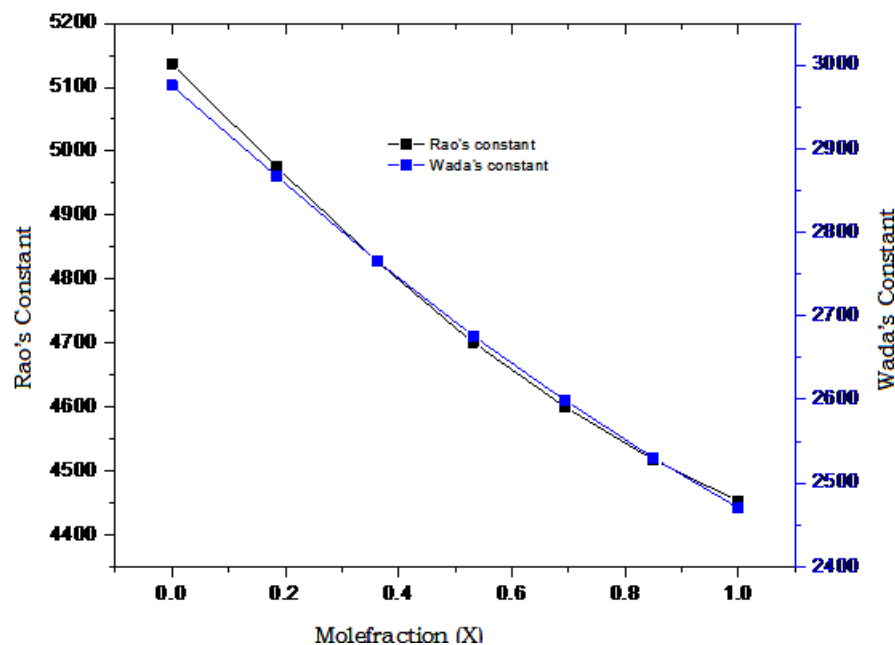


Fig. 2 . Variation of Rao's constant and Wada's constant with mole fraction of Ethylmethylketone with Chlorobenzene at Temperature 308.15K

V. CONCLUSION

The ultrasonic velocity (U), adiabatic compressibility (β_{ad}), molar volume (V), viscosity (η), intermolecular free length (L_f) and their excess parameters were studied in the binary systems containing ethyl methyl ketone with chloro benzene had concluded that Velocity versus composition exhibits non-linearly variation, viscosity versus composition reaches broad maximum, excess Intermolecular free length exhibits a minimum deviation in adiabatic compressibilities and excess molar volumes and excess free lengths are typically over the entire composition range reaching minimum excess value at some intermediate concentration which depends on the nature of solute while the deviation in viscosities, excess acoustic impedance and excess velocity show maximum for this system. Hydrogen bonded intermolecular complexes might be formed between ethyl methyl ketone and the chloro benzene having definite stoichiometric proportion. These results are in qualitative agreement with the observations made by earlier investigators.

It may be concluded that there is good agreement between experimental and theoretical velocities of Nomoto relation in all the three binary systems. Thus, the linearity of molar sound velocity and additivity of molar volumes, as suggested by Nomoto in deriving the empirical relation have been truly observed in the aforementioned binary liquid mixtures. The success of Nomoto's relation in predicting the experimental ultrasonic velocities for polar-polar liquid mixtures has also been emphasized by others.

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