

Studies on Thermo acoustic Parameters in binary liquid mixtures of MIBK with 1-Propanol, 1-Butanol and 1-Pentanol at 303.15K-A new approach by Direct Measurement of Acoustic Impedance

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Abstract

Acoustic Impedance and Ultrasonic Velocities of binary liquid mixtures Methyl Iso Butyl Ketone (MIBK) with 1-Propanol, 1-Butanol and 1-Pentanol have been measured over the wide mixture compositions at $T = 303.15$ K. Acoustic Impedance (Z) has directly been measured by using a new technique based on pulse echo reflectory method, while ultrasonic velocity has been measured by Δt method. These data have been used to compute densities, Intermolecular free length (L_F), molar volumes (V_m), excess intermolecular free length (L_F^E), isentropic compressibility (K_S), excess acoustic impedance (Z^E), excess molar volumes (V_m^E), excess isentropic compressibility (K_S^E). The values of L_F^E , V_m^E , K_S^E are negative over the wide range of composition for all the studied binary mixtures, while the values of Z^E are found positive. The changes in the thermo acoustic parameters as a function of variations in the composition and chain length of alkyl groups in alkanol molecules are discussed in terms of the intermolecular interactions.

Key words: MIBK, 1-Propanol, 1-Butanol, 1-Pentanol, direct acoustic impedance, novel technique, ultrasonic velocity by Δt , intermolecular interactions.

Introduction

In a bid to explore newer approach towards measurement of acoustic parameters in liquid mixtures, authors have proposed a novel technique for investigating molecular associations / interactions in binary and ternary liquid mixtures. The technique involves simultaneous evaluation of both ultrasonic velocity and density by measuring acoustic impedance of liquid mixtures directly at one instance itself. Literature survey shows that acoustic impedance studies have been carried out in different materials by pulse echo reflectory techniques previously. Acoustic impedance of material deposited on one face of a piezoelectric resonator from the measured frequency spectrum of the electrical impedance have been reported earlier¹. Evaluation of acoustic impedance of solid sallow² in various forms and in different directions of crystals were studied by measuring the amplitudes of reflected echoes from the acoustic interface. In this work a novel technique has been presented for the first time for measurement of acoustic impedance in liquid samples on the basis of the reflection coefficient. Literature survey shows that majority of ultrasonic investigations in liquid mixtures have been carried out by way of measurements of ultrasonic velocity in frequency 2 to 3 MHz along with experimentally determined density and viscosity values. But the proposed technique minimizes the innumerable measurements required to be taken for ascertaining exact values of density, as it reads the Z value directly.

In an attempt to standardize the new technique³ three binary liquid mixtures DMSO with benzene, carbon tetrachloride and methanol have been studied; the values of Z obtained from the new proposed technique were compared with those that were obtained from the

conventional method. The results have been discussed in our publication⁴. The efficiency of the new proposed method presented has been further validated in ten pure polar and nonpolar solvents; the work has been published⁵. Molecular interactions between carbonyl carbon group of ketones and aliphatic alcohols play an important role in many industrial and commercial applications. Methyl iso butyl Ketone (MIBK) has strong extractant applications in dewaxing to purify pharmaceuticals, mineral oil, in adhesives and in cellulose based, resin based coatings, in extraction of protactinium-233 from HCl solutions^{6,7}. Suitable physical properties are obtained by blending MIBK with suitable diluents for greater dispersal and more rapid phase disengagement in order to improve upon its extraction efficiency as per its application in industry. In order to investigate and to venture upon new solvation properties of MIBK in presence of alcohols the present work has been undertaken. This work undertakes the study of acoustic behavior of the binary mixtures of 1-propanol, 1-butanol, and 1-pentanol with MIBK.

Material and Methods

In a significantly new approach towards the existing ultrasonic techniques used to study intermolecular interactions in binary and ternary liquid mixtures, a new technique was developed for direct measurement of acoustic impedance in liquids / liquid mixtures in one instant itself based on pulse echo reflectory method.

The authors have been working on the new device for some time and the results obtained have been retrospectively reviewed well in comparison with those found in literature. The design of the fabricated device, the

experimental set up, the underlying theory and principle behind the entire working of the technique, the instrumentation, the structural facts, has been described and explained in detail in earlier publications^{3,4,5}. The proposed technique directly measures the acoustic impedance (Z value) of the liquids ultrasonic velocities (U) in pure solvents and binary mixtures were measured by delay time method (Δt) and as specified in detail in our earlier publications.

Methyl iso butyl ketone (MIBK), used for all the mole fraction concentrations was from E.Merck of 99% purity, 1-propanol, 1-butanol used were from C.D.H. trade mark having purity of 99.5% and 98%, while 1-pentanol used was from S.D. Fine chemicals having purity of 99%. All the solvents used were purified by standard methods before solution preparation⁸. The mixtures were prepared by mixing known masses of pure solvents in air tight weighing bottles and as described earlier.

Results and Discussion

Tables 1, 2 and 3 give the experimental values of echo amplitudes in milli volts received by the probe when in contact with air, water and the test liquid mixtures present in the experimental cell, acoustic impedance values calculated from the echo amplitudes in Ns/m^3 , ultrasonic velocity values in m sec^{-1} measured by Δt method, calculated density values from acoustic impedance and ultrasonic velocity, of binary mixtures as a function of mole fraction (x) of 1-propanol, 1-butanol and 1-pentanol with methyl iso butyl ketone at 303⁰K are given in. The thermodynamic parameters derived⁹ from measured values of acoustic impedance, ultrasonic velocity and calculated density for characterizing the structural environment in the liquid mixtures are

$$\text{Density} = Z/U$$

$$\text{Isentropic Compressibility } K_s = 1/U^2$$

$$\text{Intermolecular Free Length } L_f = K K_s^{1/2}$$

(Where K values are computed from Jacobson's Works)

$$\text{Molar sound velocity } R_m = U/3V$$

$$\text{Molar Volume } V_m = (X_1M_1 + X_2M_2)/m$$

Where U, ρ_m , M_1 , M_2 V are the values for ultrasonic velocity, density of mixture, molecular weights of component 1 and of 2, and molar volumes of the mixtures respectively. Further excess thermodynamic functions such as L_f^E , K_s^E , Z^E and excess molar volume were calculated by using the following formula:

$$A^E = AM - (X_1A_1 + X_2A_2)$$

Where X_1 and X_2 are the mole fractions of the two components in the binary solvent system and A_M , A_1 , A_2 shows the mixture and individual solvent component function properties.

Tables 1 to 6 show a decrease in the values of K_s , L_f , and molar volume V, while an increasing trend is observed in acoustic impedance Z values with the increasing mole fractions of 1-propanol, 1-butanol and 1-pentanol in MIBK binary mixtures. The decrease in molar volume and in K_s , L_f , while opposite trends in Z with x reveal the presence of specific interactions between the components of the mixtures. The trend in increase in values of Z in all the three binary systems was in the order of 1-pentanol > 1-butanol > 1-propanol.

Table-1

Binary mixtures of 1-propanol with methyl iso butyl ketone (MIBK) at 303⁰K

Propanol+ MIBK (mole fraction)	V (air) millivolts	V (water) millivolts	Z (water) Ns/m^3	V (liquid) millivolts	U (expt) m/s	Z liq (expt) Ns/m^3	Density kg/m^3 calc
0	1120	680	1.456	848	1129	812	0.719
0.2258	1130	680	1.456	842	1133	844	0.745
0.4079	1140	680	1.456	840	1136	861	0.758
0.6068	1150	680	1.456	840	1140	871	0.764
0.7556	1140	680	1.456	836	1142	875	0.766
0.8213	1140	680	1.456	836	1145	875	0.764
0.9163	1130	680	1.456	832	1149	878	0.764
1	1120	680	1.456	828	1152	882	0.766

Table-2
Binary mixtures of 1-butanol with methyl iso butyl ketone (MIBK) at 303^oK

Butanol+ MIBK (mole fraction)	V (air) millivolts	V (water) millivolts	Z (water) Ns/m ³	V (liquid) millivolts	U (expt) m/s	Z (liquid) (expt) Ns/m ³	Density kg/m ³ calc
0	1120	680	1.456	848	1129	812	0.719
0.2149	1140	680	1.456	840	1141	861	0.755
0.3498	1140	684	1.456	836	1152	885	0.768
0.4848	1140	684	1.456	832	1159	898	0.775
0.7014	1140	684	1.456	826	1161	919	0.792
0.8457	1150	680	1.456	824	1174	925	0.788
0.9147	1140	680	1.456	820	1177	929	0.789
1	1130	680	1.456	816	1182	934	0.790

Table-3
Binary mixtures of methyl iso butyl ketone (MIBK) with 1-pentanol at 303^oK

Pentanol+ MIBK (mole fraction)	V (air) millivolts	V (water) millivolts	Z water Ns/m ³	V (liquid) millivolts	U (expt) m/s	Z liq (expt) Ns/m ³	Density kg/m ³ calc
0	1120	680	1.456	848	1129	812	0.719
0.2133	1120	680	1.456	834	1135	867	0.764
0.3875	1130	684	1.456	830	1149	898	0.782
0.5232	1130	684	1.456	826	1166	915	0.785
0.6346	1140	684	1.456	824	1178	925	0.785
0.8319	1130	680	1.456	814	1194	943	0.790
0.9037	1130	680	1.456	810	1201	952	0.793
1	1130	680	1.456	808	1210	962	0.795

This is in agreement with the model for sound propagation proposed earlier¹⁰, wherein basis on theoretical explanation, values of ultrasonic velocity is bound to increase if the values of intermolecular free length (L_F) in a system decrease, as a result of mixing of components and vice versa. It therefore implies that L_F is one of the predominating parameter for deciding the nature of variation in ultrasonic parameter. The decrease in L_F leads to decrease in isentropic compressibility K_S as the concentration of 1-alkanol increases in the mixtures¹¹ which indicates presence of specific intermolecular reactions in the liquids. The decrease in intermolecular free length results in increase in ultrasonic velocity and density as specified by Jacobson¹², this can be applied towards the increasing trend in Z values as observed in our experimental finding table 1–6. The mathematical relation for acoustic impedance Z and intermolecular free length show that they should show opposite behavior, this behavior is clearly seen in our experimental results.

R_m , molar sound velocity or Rao's constant is also an important factor in deciding the molecular association in liquid mixtures. In the present work, the values of R_m are found to decrease with mole fractions of x in all the MIBK mixtures, which suggest depolymerization of aggregates of

alkanol molecules. But this may be balanced by the association between dissimilar molecules in the mixtures.

MIBK exhibits both keto and enol forms in equilibrium; by virtue to its chemical structure. It is estimated¹³ that the Kirkwood –Frohlich linear co-relation factor g in MIBK is greater than 1, which implies proximity of α multimerization i.e. presence of parallel dipole alignment in its equilibrium mixture. At lower mole fractions of 1-alkanol, on addition of 1-alkanols in MIBK, may be there occurs release of dipoles in the system due to breaking of dipole-dipole self association in MIBK molecules and there may occur a more preferential hydrogen bonding between oxygen atoms of MIBK and hydrogen atoms of 1-alkanols. This may lead to the formation of some heterogeneous discreet clusters in the mixture.

Excess values of the parameters whether tending to be on the negative side or positive side throw light on nature and strength of the possible interactions present in the system. The magnitude of L_F^E is the result of contributions from several effects. The disruption of alkanol aggregates in the presence of MIBK is due to the breaking up of hydrogen bonds in self associated alkanol molecules making L_F^E negative. Figure 4 depicts the negative change in L_F^E

values in all the concentrations of 1-alkanols. According to literature¹⁴⁻¹⁶, the negative values of excess properties give more accurate information on molecular interactions in liquid mixtures.

It is known that as the number of carbon atoms in the hydrocarbon part of 1-alkanols increase, the polar characteristics of alkanols starts reducing which resist the formation of intermolecular hydrogen bonding between 1-alkanol and MIBK molecules and hence the tendency to form associated complex decreases with the lengthening of carbon chain. During the mixing of any two liquids, by the virtue of their chemical characteristics, there may be likelihood of chemical reaction between the two components and it is possible that as the molecules of components arrange themselves around the molecules of another liquid component, heat is developed and the system entropy increases, this makes the process thermodynamically more viable. This spatial arrangement of one liquid into another component is dependent on the chemical characteristics of the molecular environment in the system like intermolecular hydrogen bonding, dipole – dipole reaction, dipole induced dipole reaction and also due to variation in dielectric constant.

In the present liquid mixture, MIBK due to its chemical nature invariably shows dipole –dipole intermolecular association and therefore when the concentration of 1-alkanol rises in their mixture, this association breaks, giving proximity to more chemically viable intermolecular hydrogen bonding between proton with δ^+ charge (from 1-alkanol) and highly electronegative oxygen of carbonyl from ketonic MIBK. In the process the strongly associated 1-alkanol molecules also lose their intramolecular association. This is very well projected in the values of thermodynamic functions of the mixtures as reported. As per the definition and as per the previously established molecular models intermolecular free length in this mixtures decrease with the increased reactivity in the system. Here two types of molecular reactions are possible, one intermolecular hydrogen bonding between O^{δ^-} (of $>C=O$) of MIBK and H^{δ^+} (of $H-O-R$) of 1-alkanol and there is also a possibility of presence of dipole-dipole reaction between O^{δ^-} (of $>C=O$) of MIBK and R^{δ^+} (of $R-O-H$) of 1-alkanol any time when due to spatial arrangement O^{δ^-} (of $>C=O$) of MIBK and R^{δ^+} (of $R-O-H$) of 1-alkanol lay adjacent to each other. Due to these increased interactions, the reactivity is expected to be on increase which is reflected in the values of L_F in the entire three binary mixtures figure 3. The L_F values in all the three cases are decreasing. It is observed that as the concentrations of 1-alkanol are increased, the values of L_F go on decreasing. It is anticipated that at higher concentrations of 1-alkanol in the mixtures there may be an increase in the concentration of similar kind of molecules in the system. Due to lessening of the intermolecular free spaces in between the molecules of the mixed system, there is a possibility of breaking of weaker intermolecular hydrogen bonds of alkanol molecules (as

specified earlier, intermolecular hydrogen bonding is weaker compared to covalent bonds present in the core molecular structure of 1-alkanols, the strength of hydrogen bond varies between 3-10 kcal mole⁻¹, while that of a covalent bond is of the order of 100 kcal mole⁻¹ readily. The alkanol molecules then would be more liable to be present as discreet, unassociated structures with the presence of dispersive forces between like-like molecules. This may be the possible explanation for decreased magnitude of –ve L_F values at higher concentrations of 1-alkanol in the mixtures. Further as is quite evident from the graphical representation figure 3 the L_F values in all the binary mixtures decrease nonlinearly as the hydrocarbon part of the alkanols go on increasing. This has been reported earlier. This can be explained on the following basis. Higher alkanols have higher molar volume, therefore in the mixture with higher alkanol (having now larger hydrocarbon units) the molar volume of the alkanol part now rises, it now occupies larger spatial arrangement in the molecular core of the binary mixture which now leaves less free intermolecular spaces in between the associated structures present in the system. The magnitude of L_F decreased in the order, 1-pentanol > 1-butanol > 1-propanol in all the three liquid mixtures.

Figure 4 represents the graphical variations in L_F^E in all the binary mixtures which present a clearer picture of the molecular inter actions taking place in the binary systems. As observed L_F^E values in all the binary mixtures are throughout negative. This behavior of L_F^E again indicates the presence of specific intermolecular reactions in the system. The negative values of L_F^E further assist in deciding upon the concentration range in which maximum association in between the reacting components may be possible. The maximum negative value of L_F^E in a mole fraction of the binary mixture is the condition wherein maximum association between the two heterogeneous molecules must have taken place.

Z^E values in all the binary mixtures are throughout positive tables 4 to 6, figure 2. Trends in Z^E values figure 2 reinforce our earlier view that there is the possibility of specific intermolecular reactions in all the mixtures. Positive values of Z^E in all mole fraction concentration further hints to the possibility of the presence of strong attractive forces in between the reacting components of the mixture.

At this point when considering all the liquid mixtures, it is found that the maximum intermolecular association in all the binary mixtures takes place in between ~ 0.4 to ~ 0.6 mole fraction of alkanol concentration range. From figure 2 it is quite evident that as the mole fraction concentration of 1-alkanol increases in the mix, the magnitude of negative value starts decreasing. This can be attributed to the fact that with the increasing concentration of 1-alkanol, the number of similar (like –like) molecules of alkanol increases in the mixture leading to the origination of dispersive kind of forces in the system.

Table-4
Variation of $K_S, L_F, R_m, L_F^E, \beta_S^E, Z^E, V_M, V_M^E$ with mole fractions of 1-propanol at 303⁰K

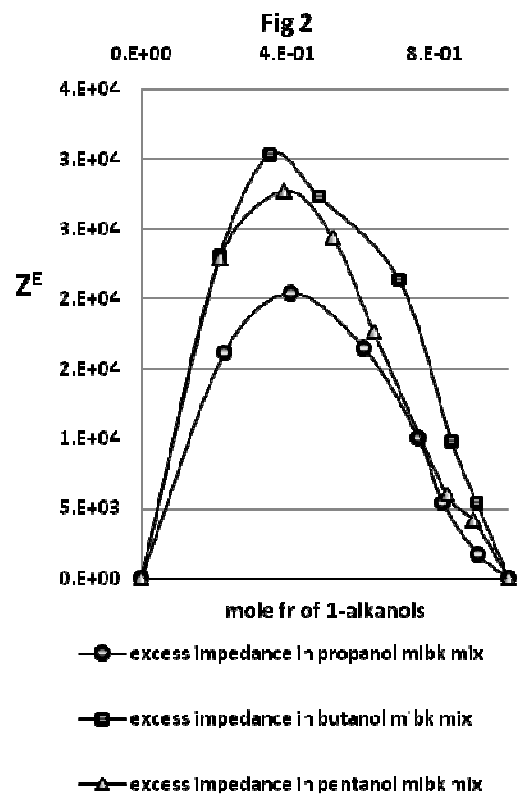
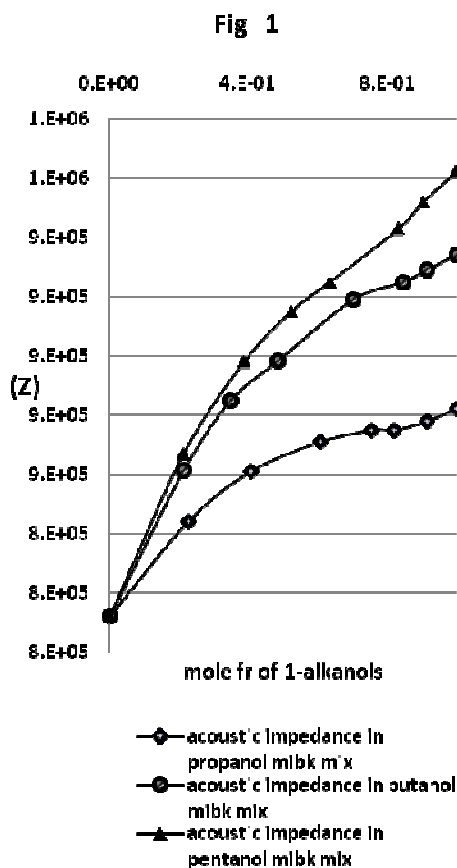
X1	K_S ($m^2 N^{-1}$)	L_F (m)	R_m ($10^{-3} m^3$ $mol^{-1} ms^{-1}$)	V_M ($m^3 mol^{-1}$) $X10^{-4}$	K_S^E ($m^2 N^{-1}$)	L_F^E (m)	V_M^E ($m^3 mol^{-1}$)	Z^E ($N s m^3$)
0.0000	1.0908E-09	6.8551E-11	1.4507	1.3932E+00	0.000E+00	0.000E+00	0.000E+00	0.000E+00
0.2258	1.0457E-09	6.7120E-11	1.2756	1.2236	-2.096E-11	-6.540E-13	-3.227E-06	1.617E+04
0.4079	1.0224E-09	6.6366E-11	1.1543	1.1062	-2.487E-11	-7.811E-13	-3.882E-06	2.040E+04
0.6068	1.0071E-09	6.5868E-11	1.0373	0.9930	-1.892E-11	-5.945E-13	-3.107E-06	1.645E+04
0.7556	1.0008E-09	6.5660E-11	0.9536	0.9123	-9.389E-12	-2.907E-13	-2.123E-06	1.001E+04
0.8213	9.9813E-10	6.5574E-11	0.9209	0.8802	-4.993E-12	-1.506E-13	-1.334E-06	5.400E+03
0.9163	9.9126E-10	6.5347E-11	0.8698	0.8304	-1.725E-12	-4.988E-14	-5.353E-07	1.738E+03
1.0000	9.8404E-10	6.5109E-11	0.8228	0.7849	0.000E+00	0.000E+00	0.000E+00	0.000E+00

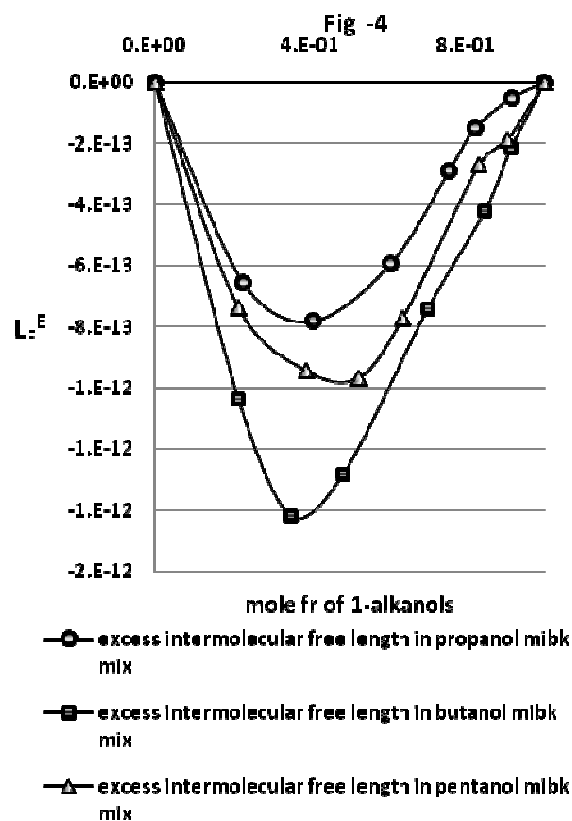
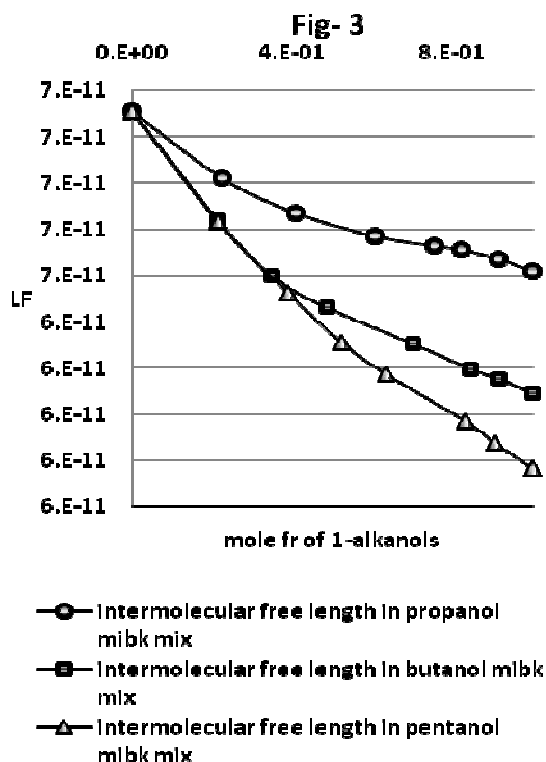
Table-5
Variation of $K_S, L_F, R_m, L_F^E, \beta_S^E, Z^E, V_M, V_M^E$ with mole fractions of 1-butanol at 303⁰K

X1	K_S ($m^2 N^{-1}$)	L_F (m)	R_m ($10^{-3} m^3 mol^{-1}$ ms^{-1})	V_M ($m^3 mol^{-1}$) $X10^{-4}$	K_S^E ($m^2 N^{-1}$)	L_F^E (m)	V_M^E ($m^3 mol^{-1}$)	Z^E ($N s m^3$)
0.0000	1.0908E-09	6.8551E-11	1.4507	1.3932	0.000E+00	0.000E+00	0.000E+00	0.000E+00
0.2149	1.0175E-09	6.6208E-11	1.3095	1.2531	-3.352E-11	-1.035E-12	-4.224E-06	2.310E+04
0.3498	9.8085E-10	6.5004E-11	1.2428	1.1855	-4.522E-11	-1.418E-12	-4.839E-06	3.030E+04
0.4848	9.6033E-10	6.4320E-11	1.1864	1.1295	-4.077E-11	-1.281E-12	-4.302E-06	2.729E+04
0.7014	9.3724E-10	6.3542E-11	1.0876	1.0348	-2.378E-11	-7.406E-13	-3.912E-06	2.140E+04
0.8457	9.2085E-10	6.2984E-11	1.0463	0.9918	-1.346E-11	-4.206E-13	-1.640E-06	9.783E+03
0.9147	9.1455E-10	6.2768E-11	1.0213	0.9673	-6.997E-12	-2.167E-13	-9.537E-07	5.362E+03
1.0000	9.0575E-10	6.2466E-11	0.9917	0.9380	0.000E+00	0.000E+00	0.000E+00	0.000E+00

Table-6
 Variation of $K_S, L_F, R_m, L_F^E, \beta_S^E, Z^E, V_M, V_M^E$ with mole fractions of 1-pentanol at 303^oK

X1	K_S ($m^2 N^{-1}$)	L_F (m)	R_m ($10^{-3} m^3 mol^{-1} ms^{-1}$)	V_M ($m^3 mol^{-1}$) $\times 10^{-4}$	K_S^E ($m^2 N^{-1}$)	L_F^E (m)	V_M^E ($m^3 mol^{-1}$)	Z^E ($N s m^3$)
0.0000	1.0908E-09	6.8551E-11	1.4507	1.3932	0.000E+00	0.000E+00	0.000E+00	0.000E+00
0.2133	1.0162E-09	6.6165E-11	1.3332	1.2781	-2.513E-11	-7.382E-13	-5.436E-06	2.295E+04
0.3875	9.6918E-10	6.4616E-11	1.2802	1.2223	-3.175E-11	-9.418E-13	-6.051E-06	2.776E+04
0.5232	9.3730E-10	6.3544E-11	1.2594	1.1965	-3.213E-11	-9.646E-13	-4.766E-06	2.436E+04
0.6346	9.1773E-10	6.2877E-11	1.2448	1.1787	-2.588E-11	-7.718E-13	-3.379E-06	1.763E+04
0.8319	8.8815E-10	6.1855E-11	1.2113	1.1418	-9.679E-12	-2.690E-13	-1.450E-06	5.971E+03
0.9037	8.7462E-10	6.1383E-11	1.1976	1.1267	-6.541E-12	-1.870E-13	-9.131E-07	4.177E+03
1.0000	8.5883E-10	6.0826E-11	1.1811	1.1084	0.000E+00	0.000E+00	0.000E+00	0.000E+00





Conclusions

Comparing all the values of L_F^E and Z^E in the above binary mixtures, it is found that the excess intermolecular free length in case of 1-butanol mixture is the lowest while its excess acoustic impedance value is the highest at concentration ranging from 0.4 to 0.5 mole fractions. This implies that the intermolecular reactivity in between 1-butanol and MIBK is the maximum. In case of 1-pentanol – MIBK mix, may be due to seemingly long hydrocarbon chain (when compared with 1-butanol structure), there may be presence of steric forces which inhibits the perfect alignment in the spatial molecular structure required for intermolecular hydrogen bonding between the dissimilar molecules, also the dipole-dipole bonding due to long straight chain structure of 1-pentanol may not be that readily possible. In case of 1-propanol MIBK mix, due to smaller molar volume of 1-propanol there may be occurrence of some free intermolecular spaces in between the surfaces of molecules, which must have lead to lesser magnitude in excess values.

Thus based on the above interpretations, 1-butanol – MIBK seemingly appears to be a better binary mixture (based on thermodynamic predictions), in which maximum dispersion of the two components may be predicted.

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