

# Excess parameter studies on tetrahydropyran with 1-hexanol at T = 298.15 to 318.15 K using Anton Paar

K. Anil Kumar<sup>1,\*</sup>, Ch. Srinivasu<sup>2</sup>

<sup>1</sup>Department of Physics, Sri Vani School of Engineering, Chevuturu, A.P., India

<sup>2</sup>Department of Physics, Andhra Loyola College, Vijayawada - 8, A.P., India

\*E-mail address: anilkumarkoneti@gmail.com

## ABSTRACT

Sound velocity, densities of binary mixture of Tetrahydropyran (THP) with 1-hexanol has been measured over the entire range of composition at T = 298.15 to 318.15 K. The excess parameters *viz.*, excess sound velocity, deviations in isentropic compressibility, excess molar volume, excess free length and excess acoustic impedance are deduced from experimental values and discussed intermolecular interactions present in the mixture. At the end all the parameters have been fitted to Redlich-Kister equation and their coefficients are obtained.

**Keywords:** Sound velocity; Densities; Excess parameters; THP; 1-Hexanol; Binary mixtures

## 1. INTRODUCTION

Ultrasonic velocity studies in binary liquid systems which are capable of supporting extensive hydrogen bonding networks have been carried out by many research groups [1-6]. Such studies can provide a lot of information on the molecular interactions. Ultrasonic wave propagation affects the physical properties of the medium and hence, can furnish information on the physics of the liquid and liquid mixtures. THP is used in polymerization processes [7], is a cyclic monoether, an excellent solvent very often used in the manufacture of special chemicals & 1-hexanol is a substance heavily used in the perfume industry. In this paper the sound velocity, densities of binary mixture THP with 1-hexanol have been measured at 298.15 to 318.15 K using

Anton Paar. From the experimental values, excess sound velocity ( $u^E$ ), deviations in isentropic compressibility ( $\Delta K_s$ ), excess molar volumes ( $V_m^E$ ), excess free length ( $L_f^E$ ) and excess acoustic impedance ( $Z^E$ ) for the binary system are estimated using standard equations that are reported by several authors [8-10]. Apart from that the deviations in isentropic compressibility are more related with structural effects and packing phenomena. The systematic investigations of these excess properties are therefore of great importance. The values of  $\Delta K_s$  and  $V_m^E$  which can be measured with great accuracy reflect the degree of deviation from ideality. The deviations from ideal behavior has been widely used for the study of structural variations and molecular interactions of mixtures [29-34].

## 2. EXPERIMENTAL DETAILS

THP, 1-pentanol were purchased from Aldrich chemical Company with purities >0.998, the purities of the purified liquids were checked by measuring their sound velocity, densities [recorded in Table 1] using Anton Paar DSA 5000M at the range of 298.15 to 318.15 K ( $\pm 0.01$  K) and these agreed to within  $\pm 2 \times 10^{-3} \text{ kg m}^{-3}$  with their literature values [11-20]. Sound velocity ( $u$ ), densities ( $\rho$ ) of the pure liquids and their binary mixture were measured using a vibrating U-tube digital density and sound analyzer as explained in the literature [21,22] which is the same as claimed by the manufacturer. The measurements are based on measuring the period of oscillation of a vibrating U-shaped hollow tube filled with the sample. The calibration of the apparatus was carried out with the double distilled, de ionized water before each series of measurements. The mole fraction of mixture was obtained with uncertainty of  $1 \times 10^{-4}$  from the measured apparent masses of the components. All the mixtures were weighed on an electric balance Sartorius, model CP 225D,  $\pm 0.01$  mg. The uncertainties in the density and speeds of sound measurements are  $2 \times 10^{-3} \text{ kg}\cdot\text{m}^{-3}$  and  $0.1 \text{ m s}^{-1}$  respectively.

**Table 1.** Properties of the pure compounds and comparison with literature values at T = 298.15 to 318.15 K.

Compound	T (K)	u (ms <sup>-1</sup> )		$\rho$ (kgm <sup>-3</sup> )	
		Observed	Literature	Observed	Literature
THP	298.15	1269.3	1269.3 [11]	879.4	879.1 [12,13]
	303.15	1246.5	1246.8 [14]	874.3	
	308.15	1223.9	1224.4 [14]	869.3	869.2 [15]
	313.15	1201.6		864.2	864.0 [15]
	318.15	1179.4		859.1	
	298.15	1303.7	1303.0 [16]	816.1	816.2 [17]
1-hexanol	303.15	1286.7	1285.6 [16]	812.5	811.6 [18,19]
	308.15	1269.8		808.8	808.0 [19]
	313.15	1253.1		805.2	804.6 [20]
	318.15	1236.4		801.4	

## 3. THEORY

Using the measured values of data, calculate the various thermo acoustical parameters such as;

$$\text{Isentropic compressibility } K_s = \frac{1}{\rho U^2} \quad (1)$$

$$\text{Molar volume } \bar{V} = \frac{\bar{M}}{\rho} \quad (\text{where } \bar{M} = M_1 X_1 + M_2 X_2) \quad (2)$$

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

(where  $K$  is Jacobson's constant, temperature dependent)

$$\text{Specific acoustic impedance } Z = U \rho \quad (4)$$

The strength of interaction between the component molecules of binary liquid system is well reflected in the excess functions from ideality. The excess thermodynamic properties such as  $\Delta K_s$ ,  $V_m^E$ ,  $L_f^E$ , &  $Z^E$  have been calculated using the following equation;

$$Y^E = Y_{mix} - (x_1 y_1 + x_2 y_2) \quad (5)$$

where  $x_1$  and  $x_2$  are mole fractions of THP and 1-pentanol respectively.

Further, the excess parameters were fitted to Redlich – Kister polynomial equation to estimate the adjustable parameters.

$$Y^E = x_1 x_2 \sum_{i=0}^n a_i (1 - 2x)^i \quad (6)$$

using least-squares regression method, the  $(a_i)$  coefficients are obtained by fitting above equation to the experimental values. The optimum number of coefficients is ascertained from an examination of the variation in standard deviation ( $\sigma$ )

$$\sigma(Y) = \left[ \frac{\sum (Y_{\text{expt}} - Y_{\text{calc}})^2}{N - n} \right]^{1/2} \quad (7)$$

where 'N' is the number of data points and 'n' is the degree of fitting

#### 4. RESULTS AND DISCUSSION

The values of sound velocities and densities for pure liquids are experimentally measured and are compared with the literature values and they are in good agreement with each other as given in the Table 1. The experimental data related to excess sound velocity, deviations in isentropic compressibility, excess molar volume, excess free length and excess acoustic impedance for the binary liquid mixture at different temperatures are given in Tables 2(a), 2(b). Further, parameters  $(a_i)$  and standard deviations  $\sigma(Y)$  for the Redlich-Kister equations are reported in table 3.

According to R J Fort et al. and A Ali et al. the sign and magnitude of  $\Delta K_s$  and  $V_m^E$  play a vital role in assessing the molecular interactions in the liquid mixtures. In general negative values of  $\Delta K_s$  and  $V_m^E$  indicates strong interaction in the mixture which include charge-transfer, dipole-dipole, dipole-induced dipole interactions and interstitial accommodation of the smaller molecules into the spaces created by bigger molecules, while positive signs of these parameters are indicative of weakening of interactions between the component molecules [23,24].

**Table 2(a).** Experimental data ( $u$ ,  $\rho$ ) and derived excess parameters ( $u^E$ ,  $\Delta K_s$ ,  $V_m^E$ ,  $L_f^E$ ,  $Z^E$ ) for THP + 1-hexanol system at 298.15, 303.15 & 308.15 K.

$x_1$	$u$ $\text{ms}^{-1}$	$\rho$ $\text{kgm}^{-3}$	$u^E$ $\text{ms}^{-1}$	$\Delta K_s \cdot 10^{-10}$ $\text{m}^2\text{N}^{-1}$	$V_m^E \cdot 10^{-4}$ $\text{m}^3 \text{mol}^{-1}$	$L_f^E \cdot 10^{-6}$ $\text{m}$	$Z^E \cdot 10^6$ $\text{kgm}^{-2}\text{s}^{-1}$
<b>298.15 K</b>							
0.0000	1303.7	816.1	0.0000	0.0000	0.0000	0.0000	0.0000
0.1335	1299.4	822.2	0.3348	0.0139	0.0040	0.0054	-0.0026
0.2445	1295.7	828.1	0.5321	0.0186	0.0060	0.0072	-0.0036
0.3547	1291.3	834.2	-0.5555	0.0386	0.0078	0.0149	-0.0058
0.4506	1286.1	839.3	-1.4807	0.0535	0.0087	0.0206	-0.0073
0.5620	1283.5	846.8	-1.4660	0.0522	0.0086	0.0202	-0.0072
0.6602	1279.8	853.2	-0.5655	0.0361	0.0074	0.0140	-0.0055
0.7436	1278.9	860.1	0.1643	0.0218	0.0061	0.0085	-0.0039
0.8447	1274.2	866.6	0.1160	0.0165	0.0045	0.0065	-0.0029
0.9073	1272.4	871.7	-0.3791	0.0185	0.0033	0.0072	-0.0027
1.0000	1269.3	879.4	0.0000	0.0000	0.0000	0.0000	0.0000
<b>303.15 K</b>							
0.0000	1286.7	812.5	0.0000	0.0000	0.0000	0.0000	0.0000
0.1335	1282.3	818.4	0.9520	0.0066	0.0039	0.0025	-0.0019
0.2445	1277.8	824.2	1.1719	0.0109	0.0059	0.0041	-0.0028
0.3547	1273.1	830.2	0.1439	0.0310	0.0077	0.0118	-0.0049
0.4506	1267.4	835.2	-0.6550	0.0450	0.0086	0.0172	-0.0063
0.5620	1264.1	842.6	-0.4951	0.0420	0.0085	0.0160	-0.0060
0.6602	1260.0	848.8	0.4406	0.0248	0.0074	0.0095	-0.0043
0.7436	1258.6	855.5	1.0926	0.0109	0.0060	0.0042	-0.0028
0.8447	1253.0	861.9	0.8062	0.0085	0.0044	0.0033	-0.0022
0.9073	1250.6	866.8	0.0855	0.0135	0.0033	0.0052	-0.0022
1.0000	1246.5	874.3	0.0000	0.0000	0.0000	0.0000	0.0000
<b>308.15 K</b>							
0.0000	1269.8	808.8	0.0000	0.0000	0.0000	0.0000	0.0000
0.1335	1265.1	814.7	1.2992	0.0016	0.0039	0.0006	-0.0015
0.2445	1260.1	820.3	1.6866	0.0037	0.0058	0.0014	-0.0021
0.3547	1254.8	826.2	0.7829	0.0229	0.0075	0.0087	-0.0040
0.4506	1248.7	831.0	0.0745	0.0363	0.0085	0.0137	-0.0053
0.5620	1244.9	838.3	0.3070	0.0323	0.0084	0.0122	-0.0050
0.6602	1240.2	844.4	1.2491	0.0143	0.0072	0.0054	-0.0034
0.7436	1238.2	850.9	1.8380	0.0008	0.0059	0.0003	-0.0020
0.8447	1231.8	857.1	1.3626	0.0010	0.0043	0.0004	-0.0015
0.9073	1229.0	861.9	0.4554	0.0088	0.0032	0.0033	-0.0017
1.0000	1223.9	869.3	0.0000	0.0000	0.0000	0.0000	0.0000

**Table 2(b).** Experimental data ( $u$ ,  $\rho$ ) and derived excess parameters ( $u^E$ ,  $\Delta K_s$ ,  $V_m^E$ ,  $L_f^E$ ,  $Z^E$ ) for THP + 1-hexanol system at 313.15 & 318.15 K

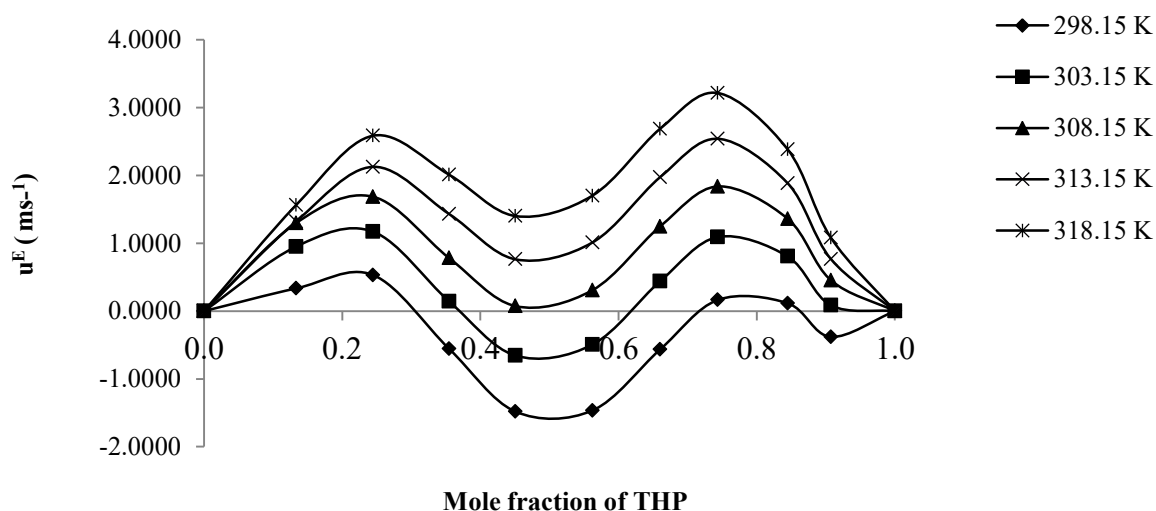
$x_1$	$u$ $\text{ms}^{-1}$	$\rho$ $\text{kgm}^{-3}$	$u^E$ $\text{ms}^{-1}$	$\Delta K_s \cdot 10^{-10}$ $\text{m}^2\text{N}^{-1}$	$V_m^E \cdot 10^{-4}$ $\text{m}^3 \text{mol}^{-1}$	$L_f^E \cdot 10^{-6}$ $\text{m}$	$Z^E \cdot 10^6$ $\text{kgm}^{-2}\text{s}^{-1}$
<b>313.15 K</b>							
0.0000	1253.1	805.2	0.0000	0.0000	0.0000	0.0000	0.0000
0.1335	1247.6	810.9	1.3244	0.0001	0.0038	0.0001	-0.0013
0.2445	1242.4	816.4	2.1273	-0.0037	0.0057	-0.0014	-0.0015
0.3547	1236.7	822.1	1.4323	0.0135	0.0075	0.0051	-0.0032
0.4506	1230.0	826.8	0.7640	0.0268	0.0084	0.0101	-0.0044
0.5620	1225.7	833.9	1.0105	0.0224	0.0082	0.0084	-0.0041
0.6602	1220.4	839.9	1.9751	0.0034	0.0071	0.0013	-0.0024
0.7436	1218.0	846.3	2.5412	-0.0103	0.0058	-0.0038	-0.0011
0.8447	1210.8	852.3	1.8856	-0.0072	0.0043	-0.0027	-0.0009
0.9073	1207.5	857.0	0.7680	0.0042	0.0032	0.0016	-0.0013
1.0000	1201.6	864.2	0.0000	0.0000	0.0000	0.0000	0.0000
<b>318.15 K</b>							
0.0000	1236.4	801.4	0.0000	0.0000	0.0000	0.0000	0.0000
0.1335	1230.4	807.1	1.5657	-0.0046	0.0037	-0.0016	-0.0009
0.2445	1224.8	812.5	2.5864	-0.0123	0.0055	-0.0044	-0.0009
0.3547	1218.7	818.1	2.0116	0.0036	0.0073	0.0014	-0.0025
0.4506	1211.5	822.6	1.4018	0.0167	0.0083	0.0063	-0.0036
0.5620	1206.6	829.6	1.7018	0.0111	0.0081	0.0042	-0.0032
0.6602	1200.8	835.4	2.6890	-0.0090	0.0070	-0.0032	-0.0016
0.7436	1197.9	841.6	3.2161	-0.0225	0.0057	-0.0082	-0.0003
0.8447	1190.0	847.5	2.3857	-0.0162	0.0042	-0.0059	-0.0003
0.9073	1186.1	852.1	1.0831	-0.0011	0.0031	-0.0003	-0.0009
1.0000	1179.4	859.1	0.0000	0.0000	0.0000	0.0000	0.0000

**Table 3.** Parameters ( $a_i$ ) and standard deviations  $\sigma$  (Y) for the Redlich–Kister equations.

parameters	T (K)	$a_0$	$a_1$	$a_2$	$a_3$	$a_4$	$\sigma$ ( $10^2$ )
$u^E$ ( $\text{ms}^{-1}$ )	298.15	-6.5771	1.0846	50.4316	2.1202	-66.6899	58.3196
	303.15	-2.9878	-1.6752	51.8271	7.2411	-63.2176	57.5395
	308.15	0.0828	-3.0559	53.0277	8.1614	-63.4366	60.0364
	313.15	2.8588	-3.2452	56.4096	3.7777	-72.1264	61.8833
	318.15	5.5098	-4.2243	58.7460	3.4022	-75.4325	63.2409
$\Delta K_s$ ( $10^{-10} \text{m}^2\text{N}^{-1}$ )	298.15	0.2229	0.0004	-0.7180	-0.0368	1.0298	0.8898
	303.15	0.1861	0.0329	-0.7707	-0.1008	1.0401	0.9282
	308.15	0.1497	0.0521	-0.8237	-0.1184	1.0910	1.0062
	313.15	0.1116	0.0576	-0.9099	-0.0677	1.2554	1.0822
	318.15	0.0695	0.0760	-0.9913	-0.0677	1.3650	1.1604
	298.15	0.0354	0.0009	-0.0235	-0.0025	0.0432	0.0396

$V_m^E$ ( $m^3 mol^{-1}$ )	303.15	0.0349	0.0009	-0.0230	-0.0030	0.0414	0.0416
	308.15	0.0346	0.0008	-0.0247	-0.0022	0.0450	0.0405
	313.15	0.0340	0.0017	-0.0247	-0.0044	0.0447	0.0391
	318.15	0.0336	0.0013	-0.0265	-0.0038	0.0460	0.0389
$L_f^E$ ( $10^{-6} m$ )	298.15	0.0860	0.0000	-0.2765	-0.0150	0.3972	0.3436
	303.15	0.0710	0.0122	-0.2938	-0.0383	0.3965	0.3515
	308.15	0.0566	0.0196	-0.3115	-0.0442	0.4130	0.3806
	313.15	0.0418	0.0214	-0.3402	-0.0250	0.4696	0.4042
	318.15	0.0261	0.0279	-0.3661	-0.0246	0.5051	0.4287
$Z^E$ ( $10^6 kgm^{-2}s^{-1}$ )	298.15	-0.0301	-0.0001	0.0658	0.0032	-0.0991	0.0883
	303.15	-0.0257	-0.0033	0.0660	0.0096	-0.0948	0.0859
	308.15	-0.0217	-0.0034	0.0668	0.0072	-0.0946	0.0871
	313.15	-0.0181	-0.0042	0.0683	0.0051	-0.0986	0.0890
	318.15	-0.0146	-0.0052	0.0697	0.0055	-0.0990	0.0859

Fig. 1 shows the behavior of excess sound velocity ( $u^E$ ). It exhibits less magnitude of deviations at the entire composition range of THP for the mole fraction 0.2445 to 0.7436 in the mixture at all five temperatures when compare to other mole fractions studied. According to A Ali et al [24] the negative (less magnitude) deviations of  $u^E$  suggests the existence of dispersion forces in the system. Further, the deviations of  $u^E$  are observed to be in opposite trends of  $\Delta K_s$  it suggests specific interactions are exists in the mixture.

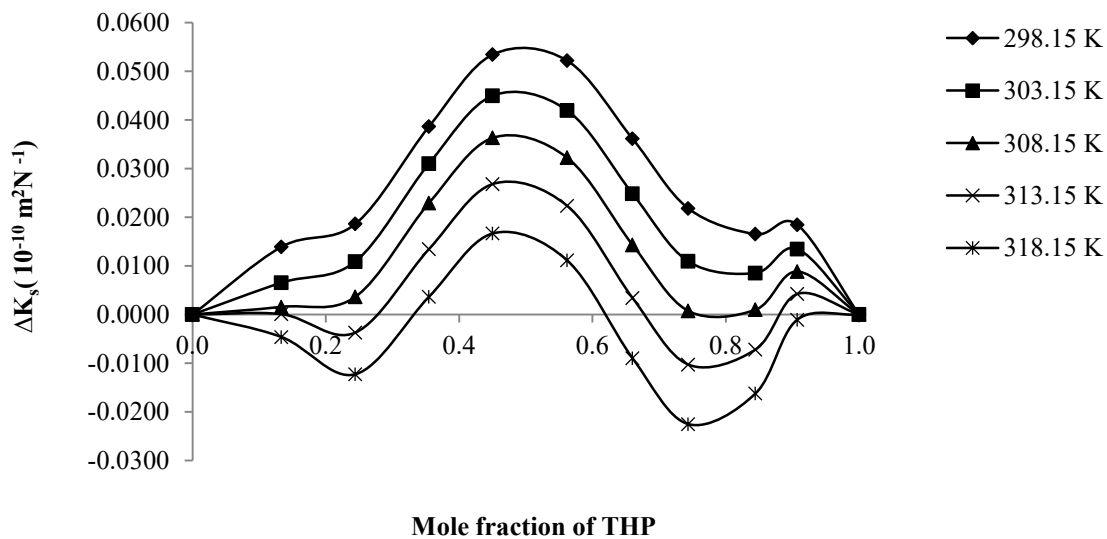


**Fig. 1.** Variation of excess ultrasonic velocity ( $u^E$ ) with mole fraction of THP + 1-hexanol system at different temperatures.

But on close inspection of Table 2(a) 2(b), the values of  $\Delta K_s$  (Fig. 2) are positive at  $T = 298.15, 303.15 \text{ \& } 308.15 \text{ K}$  for the whole mole fraction range of THP and it is negative for the lower and higher concentrations of THP at  $T = 313.15 \text{ \& } 318.15 \text{ K}$  respectively. Where as  $V_m^E$  are positive at all temperatures studied are shown in fig. 3 for the whole mole fraction range of THP. This indicates specific interactions of the following [25]. At the temperatures  $298.15, 303.15 \text{ \& } 308.15 \text{ K}$  for the whole mole fraction of THP an increase of  $\Delta K_s$  denotes weakening of inter-molecular interactions.

The hetero-molecular i.e. hexanol-THP interactions not only disturb the homo molecular (hexanol-hexanol, THP-THP) interactions in components liquids, but also cause re-arrangement in the geometry of the clusters in such a way volume of the cluster increases. It will lead to increase in volume of the cluster i.e. excess molar volume ( $V_m^E$ ) will be positive.

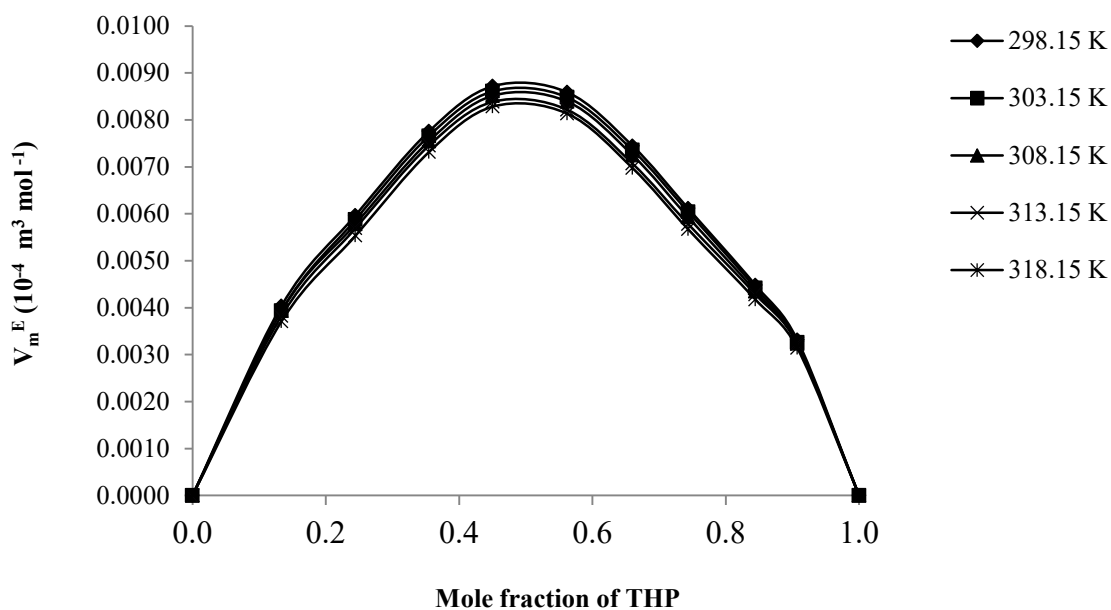
Where as at temperatures  $313.15 \text{ \& } 318.15 \text{ K}$  as mole fraction of THP is  $< 0.2445$  and  $> 0.7436$ , it is observed that  $\Delta K_s$  is negative &  $V_m^E$  is positive. The decrease in  $\Delta K_s$  indicates an attractive hetero molecular interactions leading to an association of molecules but still molar volume increases, the reason for increase in volume may be due to the larger size of molecular clusters (hexanol-hexanol, THP-THP).



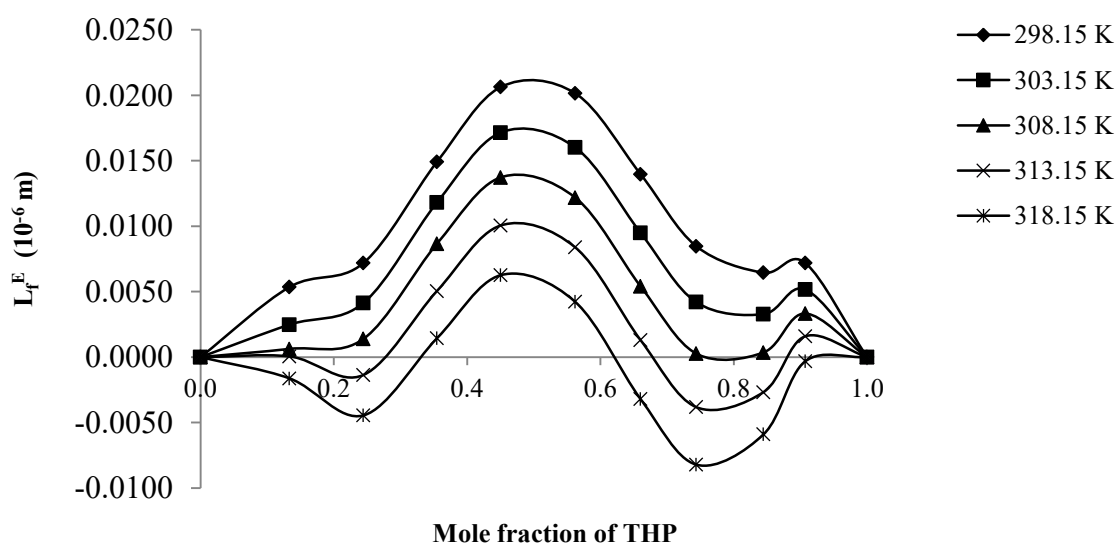
**Fig. 2.** Variation of deviations in isentropic compressibility ( $\Delta K_s$ ) with mole fraction of THP + 1-hexanol system at different temperatures.

The variation of excess intermolecular free length ( $L_f^E$ ) is shown in Fig. 4. The deviation of  $L_f^E$  are well supports the deviation of isentropic compressibility. The sign of excess properties plays a vital role in assessing the compactness or extent of molecular interactions.

The various types of interactions that are operating between the molecules are dispersion forces, which should make a positive contribution to excess values and charge transfer, H-bonding, dipole-dipole interaction and dipole-induced dipole interactions expected to make negative contributions. In the present mixture as  $\Delta K_s$  and  $L_f^E$  are positive and negative suggesting dispersive and attractive forces are present in the mixture [26].



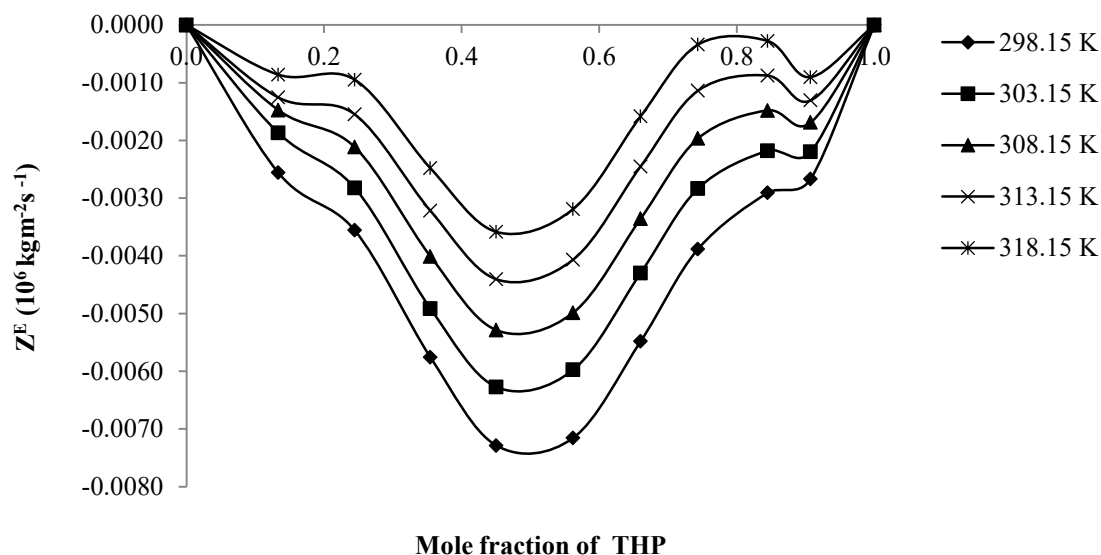
**Fig. 3.** Variation of excess molar volumes ( $V_m^E$ ) with mole fraction of THP + 1-hexanol system at different temperatures.



**Fig. 4.** Variation of excess free length ( $L_f^E$ ) with mole fraction of THP + 1-hexanol system at different temperatures.



Further, the calculated values of excess acoustic impedance (Fig. 5) of binary mixture at all temperatures studied are negative suggests that the rupture of the hydrogen bonded chain of 1-hexanol dominates over that of the hydrogen bond formed between the unlike molecules. The similar observations are reported by the author in his earlier work on 1,4-dioxane with 1-butanol [27,28], this shows weak molecular interactions between the components of the mixture exists.



**Fig. 5.** Variation of excess acoustic impedance ( $Z^E$ ) with mole fraction of THP + 1-hexanol system at different temperatures.

## 5. CONCLUSION

Sound velocity and densities for binary mixture consist of THP with 1-hexanol system is measured at  $T = 298.15, 303.15, 308.15, 313.15$  &  $318.15$  K using Anton-Paar. The calculated excess parameters are discussed and concluded the presence of weak dispersion forces in the mixture.

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