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Acoustical properties of Ternary Mixture of di-(2-ethyl-hexyl) Phosphoric acid in Cyclohexane and Ethanol mixed Solvent at various temperatures

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Abstract

Ultrasonic speed and density have been measured at 303K-318K in ternary system of (di-(2-ethyl hexyl) phosphoric acid) of dehpa with cyclohexane andethanol mixed solvent over entire composition range. The acoustical parameters such as adiabatic compressibility (K_s), free length (L_f), acoustic impedance (Z), Relative association (R_A), wada's constant (W), molar volume (V_m), interaction parameter (χ), surface tension (σ), Rao's constant (R_m) and also excess parameters such as excess adiabatic compressibility (K^E_s), Excess velocity (C^E), Excess free length L^E_f, Excess molar volume (V^E_m)and Excess acoustic impedance(Z^E) are computed for the ternary mixture at different temperatures. The results are discussed in terms of molecular interaction between the components of the ternary mixture. Calculated excess values indicate that both strong and weak molecular interaction prevailing in the system which basically depend upon concentration. The ultrasonic velocities calculated using various theoretical models and validity of these theoretical relations have been discussed in the present system.

Keywords: Ternary mixture, molecular interaction, adiabatic compressibility, acoustic impedance, interaction parameter.

Introduction

Knowledge on thermo acoustic properties of liquid mixture and solutions are more essential for their application in chemical, textile, leather and nuclear industries. Different methods like infra red, Raman effect, di-electric, magnetic resonance are used to know molecular interaction and solution properties. Like other methods, measurement of ultrasonic velocity in liquid mixture and solutions is very useful and convenient tool to recognise the physico-chemical behaviour and molecular interactions. The dependence of acoustical parameters on composition and temperature resulting from molecular interaction between components of liquid mixture. The biological activity of drug molecules, the activation energy of the metabolic process basically depend upon the nature and strength of the intermolecular interactions. The evaluation of excess properties are very useful to ascertain the type and extent of molecular interactions that are present in liquid mixtures. In recent years, considerable emphasis, have been given for the evaluation of ideal and excess thermodynamic quantities of binary and ternary liquid mixtures[1-8]. Increasing use of di-(2-ethyl-hexyl) phosphoric acid (Dehpa), cyclohexane and ethanol in many industrial processes have greatly stimulated the need for extensive information on the acoustic and transport properties of these liquids and their mixtures. Dehpa is a organo phosphorous acid widely used as a characterized extractant in the atomic energy industry. Cyclohexane is a non-polar, hydrophobic hydrocarbon used as oil extractant, paint and varnish remover. It is used in laboratories in analysis and as a standard because of its unique chemical and conformational properties. Ethanol is a good solvent and associated through hydrogen bonding, forms azeotropic mixture with cyclohexane at 337.8K. Moreover ternary mixtures with alcohols as one component is used in industries to avoid the formation of azeotropes[9-10].

In the present study, the measurement on ultrasonic velocity, density and their related acoustical parameters for the system of Dehpa+ cyclohexane +ethanol at 303,308,313 and 318K have been undertaken. The acoustical and thermodynamic parameters and their excess values evaluated from density and sound speed measurements for developing insight into the type and strength of interactions between the component molecules of the ternary mixture[11-14]. Ultrasonic velocities evaluated using Nomoto's relation (NR), free length theory (FLT), Impedance dependence relations (IDR) and Rao's specific sound velocity relation (R) for the ternary mixture under investigation at temperature 303K, have been compared with those obtained from the experimental values to reflect the molecular interactions in liquid mixtures[15-18].

Material and Methods

Organic liquids in this study were of Merck grade chemicals (99.9% purity) and used without further purification. The ultrasonic velocities in liquid mixtures have been measured using an ultrasonic interferometer working at 2 MHz frequency with an accuracy of +2ms^{-1}. An electronically digital operated constant temperature bath has been used to circulate water through the double walled measuring cell made up of steel

containing the experimental solution at the desired temperatures. The density of pure liquid and liquid mixtures was determined with a specific gravity bottle with a 25ml capacity. For preparing various concentrations of ternary mixture, the mole fraction of second component cyclohexane was kept constant ($x_2 \sim 0.2$) and the mole fractions of other two components were varied from 0.0 to 0.8.

Various acoustical and thermodynamic parameters calculated from the measured value of ultrasonic velocity and density using following relations

$$K_s = 1 / \rho C^2 \quad (1)$$
$$L_f = k (K_s)^{1/2} \quad (2)$$
$$V_m = (x_1 M_1 + x_2 M_2) / \rho_{mix} \quad (3)$$
$$R_A = (\rho / \rho_o) (C_o/C)^{1/3} \quad (4)$$
$$Z = \rho C \quad (5)$$
$$\chi = (C^2_{exp}/C^2_{ideal})-1 \quad (6)$$
$$W = V_m (K_s)^{-1/7} \quad (7)$$
$$\sigma = 4.3 \times 10^{-4} \rho C^2 \quad (8)$$
$$R_m = VC^{1/3} \quad (9)$$

Where $C_{ideal} = x_1C_1 + x_2C_2 + x_3C_3$ is ideal mixing velocity.

$$A^E = A_{exp} - A_{id} \quad (10)$$
$$A_{id} = \Sigma X_i A_i \quad (11)$$

Where k is the temperature–dependent constant having the value $(93.875 + 0.375T) \times 10^{-8}$, T, x_i ,A^E, A_{exp} and A_{id} are absolute temperature, the mole fraction of ith component, excess property of any given parameter, the experimental value and the ideal value respectively.

The theoretical values of sound speeds are evaluated using following relations taken from literature.

$$C^{Nom} = \{ x_1R_1 + x_2R_2 + x_3R_3 \} / x_1V_1 + x_2V_2 + x_3V \}^3 \quad (12)$$
$$C^{IDR} = \Sigma x_i Z_i / \Sigma x_i \rho_i \quad (13)$$
$$C^{R} = \Sigma (x_i r_i \rho)^3 \quad (14)$$
$$C^{FLT} = k / L_f (_{mix}) \rho_{exp}^{1/2} \quad (15)$$

Where x_1,x_2,x_3 are mole fractions of the component liquids, R_1,R_2,R_3 the molar sound velocity and V_1,V_2,V_3 are the molar volume. ρ corresponds to density of liquid and Z is the acoustic impedance.

$r_i = (C_i^{1/3} / \rho_i)$ is the Rao's specific sound velocity of the ith component of the mixture.

Results and Discussion

The experimental and literature values of density (ρ) and ultrasonic velocity (C) of pure liquid at 303,308, 313 and 318K are listed in table-1. Measured values of density (ρ), Ultrasonic velocity (C), adiabatic compressibility (Ks), free length (L_f), acoustic impedance (Z), interaction parameter (χ), Relative association (R_A) molar volume (V_m), surface tension (σ) at 303, 308, 313 and 318K for the ternary system of Di(2-ethylhexyl) phosphoric acid + cyclohexane +ethanol are given in table-2. The values of Wada's constant (W) and Rao's constant (Rm) at different temperatures are listed in table-3. The excess parameters K_S^E, C^E, V_m^E , L_f^E , Z^E are listed in table-4 and table-5 at different temperatures. table-6 compares experimental and theoretical values of sound speed and deviations of these theories from experimental sound speeds at temperature 303K. Representative graphs of excess adiabatic compressibility (K_S^E), excess free length (L_f^E), Wada's constant (W), excess velocity (C^E), excess molar volume (V_m^E) and relative association (R_A) as function of concentration and temperature are presented in figures -1 to 6.

Table-1
Comparison of experimental values of density (ρ) and ultrasonic velocity (C) of pure liquids at 303, 308, 318 and 318K with Literature value.

		ρ (kg m-^{3})		C (ms^{-1})	
ORGANIC LIQUIDS	**T/K**	**Exptl**	**Lit**	**Exptl**	**Lit**
DEHPA	303	1009.4	1009.6	1321.6	-
	308	1008.5	1008.2	1302.2	-
	313	1004.6	1004.9	1283.4	-
	318	1002.6	1003.1	1266.0	-
Cyclohexane	303	768.1	767.7[10]	1232.0	1230.3
	308	766.5	-	1212.0	-
	313	765.2	-	1190.0	-
	318	764.3	-	1179.0	-
Ethanol	303	785.1	780.5[13]	1135.0	1130.0
	308	777.9		1126.5	
	313	770.1		1115.2	
	318	762.4		1102.0	

From table-2 it is observed that density and ultrasonic velocity are in increasing trend with the increase in mole fraction of dehpa. As density increases, the number of particles in a given region is increased and this leads to quick transfer of sound

energy and velocity also increases. The increase in ultrasonic velocity in any mixture indicates the association among molecules of a mixture. This result is in accordance with Arul et al [19]. Ethanol is a good solvent that can dissolve both the polar and non-polar components. Cyclohexane is non- polar solvent and show conformational properties. Among three components, dehpa and ethanol are expected to involve in strong interaction due to their polar nature. In addition to this there exists interaction between cyclohexane ring and –OH group of ethanol, which are mostly dispersive in nature. The net result of inter molecular forces in this ternary system is the resultant of all the interactions prevailing between the components. An examination of table-2 shows that L_f and K_s decreases with mole fraction of dehpa and increases with rise in temperature. Increase in temperature, increases the distance between surface of two molecules, thereby increasing the intermolecular distance which results decrease in density and ultrasonic velocity and increase of compressibility. This result agrees with the result shown by Pandey et al[20]. The rapid decrease in K_s with increase in concentration of dehpa clearly indicates the formation of a large number of tightly bound systems. Since velocity and density increase with concentrations, compressibility must decrease with increase in concentration, as they vary inversely with each other.

Table-2
Experimental parameters (ρ, C) and derived parameters (Ks, Z, L_f, R_A, V_m, σ, χ) for dehpa + cyclohexane +ethanol system at 303, 308, 313 and 318K

X_1	C ms^{-1}	ρ $10^{-3}kg\ m^{-3}$	Ks $10^{-10}\ N^{-1}m^2$	Z $10^6 kgm^{-2}s^{-1}$	L_f 10^{-11} m	χ	R_A	V $10^{-5}\ m^3\ mol^{-1}$	σ N/m)
303K									
0.0000	1162.2	0.8120	9.1176	0.9437	6.2655	0.0069	0.8395	6.6914	20.2682
0.1499	1200.0	0.8502	8.1680	1.0202	5.9302	0.0240	0.8696	11.2633	22.2652
0.2343	1218.6	0.8751	7.6952	1.0663	5.7560	0.0290	0.8906	13.6082	23.4505
0.3615	1246.5	0.9032	7.1257	1.1258	5.5390	0.0360	0.9122	17.0769	25.0405
0.5803	1274.3	0.9468	6.5042	1.2065	5.2919	0.0150	0.9493	22.6770	27.1327
0.6629	1278.2	0.9616	6.3651	1.2291	5.2350	-0.0028	0.9631	24.7019	27.6839
0.7828	1285.4	0.9759	6.1989	1.2544	5.1662	-0.0362	0.9756	27.7353	28.3326
308K									
0.0000	1148.1	0.8061	9.4113	0.9254	6.4231	0.0055	0.8335	6.7404	19.7541
0.1499	1183.5	0.8453	8.4460	1.0004	6.0848	0.0211	0.8651	11.3286	21.6812
0.2343	1200.9	0.8706	7.9646	1.0455	5.9089	0.0251	0.8867	13.6785	22.8245
0.3615	1227.0	0.8996	7.3834	1.1038	5.6892	0.0310	0.9097	17.1452	24.3566
0.5803	1254.1	0.9445	6.7318	1.1844	5.4323	0.0115	0.9482	22.7322	26.4249
0.6629	1259.9	0.9596	6.5650	1.2090	5.3646	-0.0023	0.9619	24.7534	27.0333
0.7828	1268.7	0.9749	6.3726	1.2368	5.2854	-0.0212	0.9715	27.7638	27.7542
313K									
0.0000	1134.1	0.7997	9.7223	0.9069	6.5869	0.0049	0.8294	6.7943	19.2399
0.1499	1168.0	0.8395	8.7315	0.9805	6.2422	0.0199	0.8622	7.4069	21.1102
0.2343	1183.8	0.8652	8.2475	1.0242	6.0677	0.0225	0.8846	13.7639	22.2004
0.3615	1208.0	0.8946	7.6601	1.0806	5.8467	0.0268	0.9085	17.2410	23.6610
0.5803	1235.6	0.9404	6.9651	1.1619	5.5751	0.0109	0.9478	22.8313	25.7300
0.6629	1241.7	0.9557	6.7864	1.1866	5.5032	-0.0017	0.9617	24.8544	26.3428
0.7828	1251.7	0.9716	6.5691	1.2161	5.4143	-0.0176	0.9751	27.8581	27.1044
318K									
0.0000	1119.3	0.7935	10.0591	0.8881	6.7594	0.0032	0.8244	6.8474	18.7184
0.1499	1152.0	0.8344	9.0306	0.9612	6.4046	0.0174	0.8587	11.4766	20.5512
0.2343	1166.7	0.8576	8.5663	1.0005	6.2377	0.0187	0.8788	13.8859	21.5309
0.3615	1188.5	0.8904	7.9509	1.0582	6.0095	0.0199	0.9068	17.3224	22.9823
0.5803	1217.9	0.9375	7.1912	1.1417	5.7152	0.0087	0.9470	22.9020	25.1025
0.6629	1225.3	0.9531	6.9883	1.1678	5.6340	-0.0014	0.9609	24.9222	25.7535
0.7828	1237.1	0.9698	6.7376	1.1997	5.5320	-0.0140	0.9746	27.9098	26.5822

The decrease in compressibility with increase in mole fraction of Dehpa clearly reveals that there exists a close association between component molecules of the present ternary system, which results decrease in L_f. A continuous decrease in K_s and L_f is a clear evidence for the existence of strong interactions like dipole-dipole, dipole-induced dipole, hydrogen bonding, formation of

charge transfer complexes etc. This fact is supported by the increase in Relative association R_A figure-6 and acoustic impedance Z table-2 with increase in concentration of dehpa. The higher impedance indicates the presence of bulkier or solvated ion due to interaction between unlike molecules which restricts the free flow of sound waves. From figure-3, table-3 and table-2 the observed increasing trend of Wada's constant, Rao's constant and surface tension with mole fraction of dehpa indicate strong solute-solvent interactions.

The molecules of dehpa are polar and exist as dimmers in pure form, cyclohexane is a closed molecule where as ethanol is a straight chain type molecule. The reduction of ethanol reduces the straight chain type and the system is more and more occupied by ring structured molecules which occupies relatively less space and increases the compactness. The attractive interactions are less as one is strongly polar and other is non-polar and each component maintains their identity. Thus molar volume V_m between the components is in an increasing trend as mole fraction of dehpa increases, as shown in table-2. This reduces the net interactive forces. This fact is supported by increasing negative value of interaction parameter and slow increase in Z and R_A values at higher mole fraction of dehpa. The increase in values of L_f and K_s and decrease in R_A, χ, Z and σ with increase in temperatures clearly reveals that the interactions become weaker with rise in temperature.

In order to know the strength and extent of inter molecular interaction between components of the present mixture,, excess parameters have been calculated and presented in table -4 and table-5. Dispersion forces are responsible for positive deviations where the size of molecules are taken into considerations. Negative deviations indicate dipole-dipole, dipole-induced dipole, charge-transfer interaction and hydrogen bonding between unlike components. Negative deviations of high magnitude lead to complex formation between the hetro-molecules of the mixture through hydrogen bond formation [21,22]. Figure -1 and figure- 2 indicate that K_S^E and L_f^E are negative over the entire mole fraction range and even with the rising of temperature. At about 0.36 mole fraction of dehpa both K_S^E and L_f^E clearly indicate that in the ternary system the hydroxy group of ethanol form strong inter molecular interactions with the positive centre of dehpa. The strong interactions may be due to formation of new hydrogen bonds and strong dipole-dipole interactions between the component molecules. But the less magnitude of negative K_S^E and L_f^E reveals that hydrogen bond formation between unlike molecules is arrested by bulky alkyl groups in dehpa creating steric hindrance and existence of strong dipole-dipole interactions between unlike molecules.

This result agrees with positive variation of Z^E table- 5 over entire mole fraction range and linear variation of W with mole fraction of dehpa figure-3. Similar observations found by other researchers[23,24]. The variation of excess velocity figure-4 shows positive deviation at low concentration and negative deviation at high concentration region of dehpa. It indicates in spite of strong dipole-dipole interaction, weak interaction are still existing in the system and that is mainly due to dispersion forces, which is indicated by decreases of negative K_S^E and L_f^E values at higher mole fraction of dehpa. The excess molar volume V_m^E shows small negative deviation at low concentration region and positive deviation at higher concentration region at all temperatures taken for study figure-5. V_m^E values increase with increase in temperature.

As dehpa is more well structured than ethanol and if a given fraction of ethanol is replaced by same mole fraction of dehpa as in the present case, only less number of dehpa will be existed due to its higher molecular weight (322- 43) than ethanol (46.07). Thus, on increasing the mole fraction, the total number of components in the system is reduced. This leads to observed increase in V_m^E even though strong dipole-dipole interactions are present. The decrease in V_m^E may be due to presence of more number of ring structured molecule at higher concentration region, which occupies comparatively less space. The decrease in values of K_S^E, L_f^E , Z^E and C^E and increase in V_m^E with the increase in temperature indicates the strength of interaction tends to be weaker with rising of temperature due to presence of weak molecular forces and thermal dispersion forces.

Table-3
Rao's constants (R_m) Wada's constants (W) for dehpa+ cyclohexane +ethanol system at temperature 303K- 318K

Mole fraction	W [$10^{-4}m^3$ mol^{-1} ((Nm^{-2})]				Rm [10^{-4}m3mol-^{1}(ms-1)]			
X_1	303K	308K	313K	318K	303K	308K	313K	318K
0.0000	0.4871	0.4892	0.4903	0.4924	7.0337	7.0575	7.0850	7.1091
0.1499	0.8342	0.8352	0.8373	0.8382	11.9690	11.9823	12.0119	12.0308
0.2343	1.0163	1.0174	1.0183	1.0214	14.5349	14.5383	14.5592	14.6172
0.3615	1.2904	1.2883	1.2894	1.2883	18.3783	18.3549	18.3619	18.3483
0.5803	1.7356	1.7314	1.7306	1.7274	24.5852	24.5140	24.4991	24.4574
0.6629	1.8966	1.8923	1.8903	1.8882	26.8079	26.7345	26.7137	26.6684
0.7828	2.1371	2.1318	2.1298	2.1259	30.1560	30.0551	29.9203	29.9602

Table-4
Excess values of adiabatic compressibility (K^E_S), velocity (C^E), molar volume (V^E) for dehpa+ cyclohexane +ethanol system at temperature 303K-318K

Mole fraction	K^E_S X 10-^{10} $N^{-1}m^2$				C^E m/s				V^E 10^{-5} m^3 mol^{-1}			
X_1	303K	308K	313K	318K	303K	308K	313K	318K	303K	308K	313K	318K
0.0000	-0.4435	-0.3550	-0.1437	0.1076	4.0	3.2	2.8	1.5	-2.8084	-2.7927	-2.7634	-2.7337
0.1499	-0.7690	-0.6959	-0.5348	-0.3427	14.2	12.3	11.5	9.9	3.8251	4.0420	4.2184	4.4091
0.2343	-0.8905	-0.8257	-0.6811	-0.4815	17.3	14.8	13.1	10.8	5.2671	5.5561	5.7496	6.4597
0.3615	-0.9304	-0.8770	-0.7596	-0.6063	21.9	18.6	15.9	11.7	6.7868	7.0888	7.3052	7.6042
0.5803	-0.6410	-0.6172	-0.5793	-0.5221	9.5	7.2	6.7	5.3	5.7367	6.0740	6.1233	6.2570
0.6629	-0.4362	-0.4399	-0.4275	-0.4064	-1.8	-1.5	-1.1	-0.8	4.4477	4.6588	4.7338	4.2088
0.7828	-0.1033	-0.1328	-0.1651	-0.3631	-16.6	-13.7	-11.2	-8.8	3.5185	3.5302	3.4611	3.4487

Table-5
Excess values of free length (L^E_f), acoustic impedance Z^E
for dehpa+ cyclohexane +ethanol system at temperature 303K-318K.

Mole fraction	L^E_f X 10^{-11} m				Z^E X10^6kgm^{-2}s^{-1}			
X_1	303K	308K	313K	318K	303K	308K	313K	318K
0.0000	-0.1478	-0.1181	-0.0471	-0.0371	0.0390	0.0369	0.0370	0.0358
0.1499	-0.2484	-0.2221	-0.1667	-0.1003	0.0495	0.0474	0.-461	0.0446
0.2343	-0.2904	-0.2661	-0.2144	-0.1449	0.0584	0.0556	0.0535	0.0477
0.3615	-0.3083	-0.2870	-0.2444	-0.1886	0.0618	0.0583	0.0552	0.0508
0.5803	-0.2128	-0.2020	-0.1871	-0.1657	0.0461	0.0433	0.0423	0.0404
0.6629	-0.1404	-0.1406	-0.1353	-0.1271	0.0323	0.0319	0.0314	0.0311
0.7828	-0.0215	-0.0324	-0.0441	-0.0553	0.0048	0.0073	0.0093	0.0115

Table-6
Application of various theoretical approaches and percentage of deviation
for dehpa + cyclohexane +Ethanol system at 303K..

Mole fraction						ΔC/C %			
X_1	C^{EXP} ms^{-1}	C^{NOM} ms^{-1}	C^{IDR} ms^{-1}	C^{FLT} ms^{-1}	$C^{R\,MS}$	C^{NOM}	C^{IDR}	C^{FLT}	C^R
0.0000	1162.2	1169.2	1157.9	1135.4	1299.6	-0.6000	0.3699	2.3059	-11.8224
0.1499	1200.0	1237.4	1191.3	1151.8	1373.9	-3.1166	0.7250	4.0166	-14.4916
0.2343	1218.6	1258.3	1280.9	1160.1	1428.9	-3.2578	0.7959	4.8005	-17.2575
0.3615	1246.5	1279.4	1233.7	1180.7	1460.7	-2.6393	1.0268	5.2787	-17.1841
0.5803	1274.3	1301.4	1273.2	1225.0	1478.2	-2.1266	0.0863	3.8687	-16.0009
0.6629	1278.2	1306.8	1286.8	1244.9	1472.5	-2.2375	-0.6728	2.6052	-15.2010
0.7828	1285.4	1313.6	1305.8	1280.3	1428.4	-2.1938	-1.5870	0.3967	-11.1249

The experimental values of sound speed along with the calculated values and percentage of deviations for Nomato relation (NOM), Free Length Theory (FLT), Impedance Dependences Relation (IDR), Rao's specific sound velocity relation (R) are presented in Table-6 at temperature 303K.

The large negative deviation observed in Rao's specific sound velocity relation indicates absence of any strong specific interaction and complex formations in the present ternary system, which is supported by the observed negative deviation in Nomoto relation. Positive deviation observed in free length theory indicates that component molecules in the ternary mixture have a more compact arrangement than in component liquids forming the mixture. The observed comparatively more positive deviation at the intermediate concentration range indicate more interactions.

The small positive deviations at the lower mole fraction region and small negative deviations at higher mole fraction region of dehpa observed using impedance dependence relation indicates the interactions become weaker with increasing concentration. Among four theories taken up for prediction of sound velocity, impedance dependence relation is found to yield excellent comparison with the experimental value followed by free length theory[25].

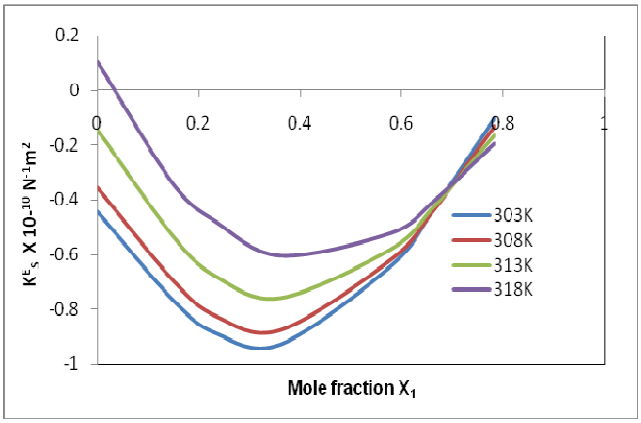

Figure-1
Variation of deviation in isentropic compressibility (K^E_S) with Mole fraction of Dehpa

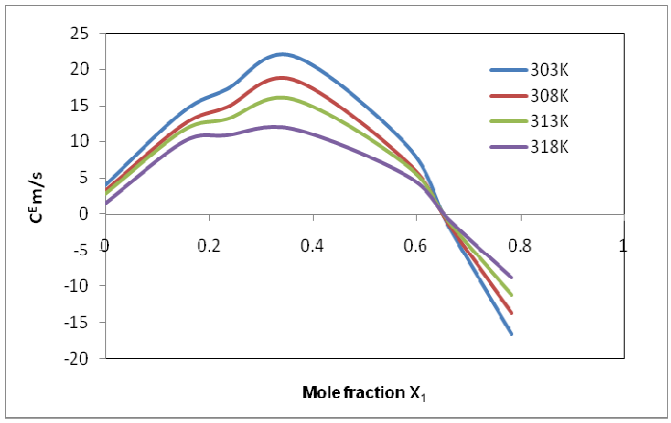

Figure-4
Variation of excess velocity (C^E) with mole fraction of Dehpha

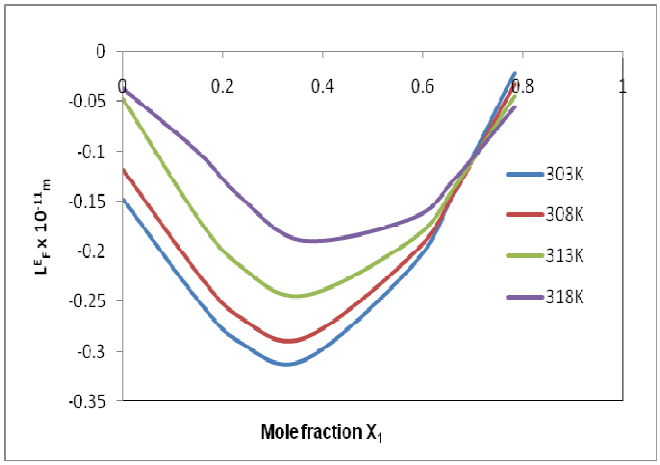

Figure-2
Variation of excess intermolecular free length (L^E_f) with mole fraction of Dehpa

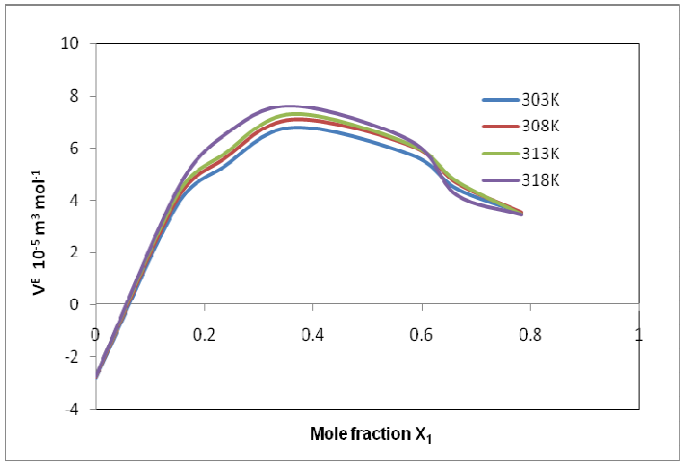

Figure-5
Variation of excess molar volume (V^E_m) with mole fraction of Dehpa

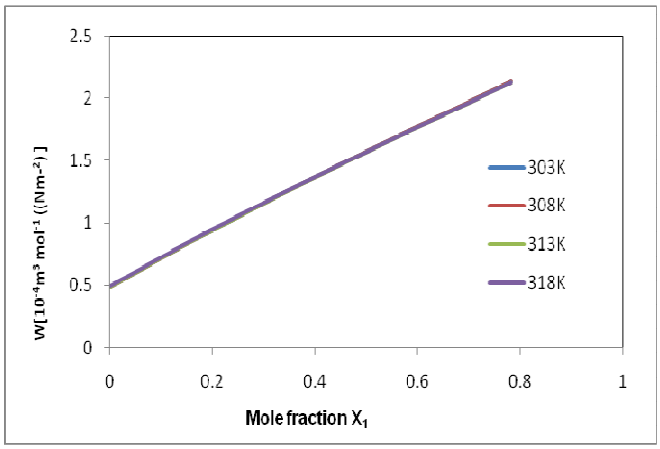

Figure-3
Variation of Wada's constant (W) with mole fraction of Dehpa

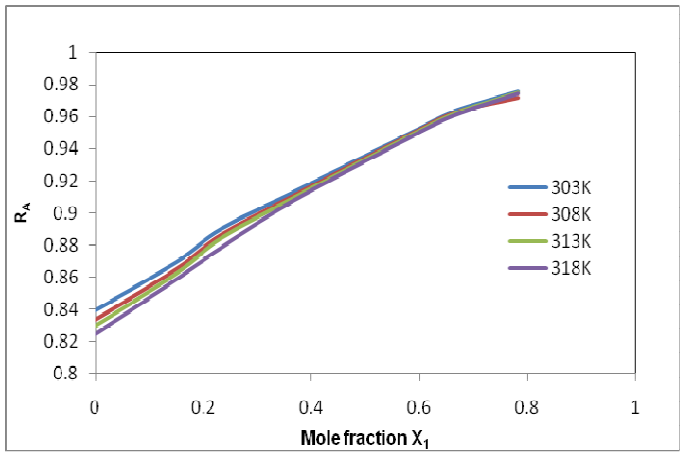

Figure-6
Variation of Relative association (R_A) with mole fraction of Dehpa

Conclusion

Acoustic parameters are sensitive to the molecular interactions present in the ternary system taken for study. The negative values of K_S^E, L_f^E, positive values of Z^E, positive and negative values of V_m^E and C^E in the ternary mixture reveal the presence of strong dipole-dipole interactions between the component molecules in the mixture which basically depend upon concentration of dehpa. The strength of interaction become weaker with increase in concentration and every component maintain their identity. The temperature variation indicates that the strength of inter molecular interaction decreases with rise in temperature. Among four theoretical models, Impedance Dependence Relation (IDR) shows close agreement with experimental ultrasonic speeds followed by Free Length Theory (FLT).

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