Acoustic properties of binary liquid mixtures of carbon tetrachloride with benzene and substituted benzenes

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ABSTRACT

The ultrasonic velocities in binary liquid mixtures of carbon tetrachloride with benzene, toluene, m-xylene, ethyl benzene and chloro benzene have been measured at entire mole fraction range at 303.15K, using ultrasonic interferometer of frequency 2 MHz. Isentropic compressibilities K_S, deviations of K_S from ideal mole fraction mixture law ΔK_S, available volumes V_a, Lennard - Jones potential repulsive term exponents n, molecular interaction terms á etc. have been calculated therefrom. The values of acoustic parameters are explainable by considering the varying extent of molecular interactions and the difference in free volumes between unlike molecules. Observed ΔK_S have been analyzed in the light of Flory's theory of binary liquid mixtures. Ultrasonic velocities evaluated from Nomoto's and Ideal mixture relation due to Van Deal *et al.,* have been compared with observed values.

Key words: Ultrasonic velocities, Isentropic compressibilities, Nomoto's relation, Molecular interactions, Flory's theory.

INTRODUCTION

The present work on acoustic properties of binary liquid mixtures of carbon tetrachloride with benzene and substituted benzenes is in continuation with our earlier ultrasonic studies[1-3]. Recently, the thermodynamics and acoustic study[1-10] of binary liquid mixtures have got considerable importance in industry and engineering. The properties of liquid mixtures are very important as some of the polymers which are insoluble in pure solvent can be dissolved in solvent mixtures[11-13]. Hence, considerable attention has been paid to the study of polymer solutions in mixed solvents. More over, an additional support of acoustic study to the thermodynamic and transport studies has proved a powerful tool to deal with molecular interactions in binary liquid mixtures and developing theoretical models. Densitometric and viscometric studies[14] of binary mixtures under present study have shown evidence of a weak molecular interaction between

unlike molecules. Except for carbon tetrachloride + benzene, ΔK_S have been found negative in sign at entire mole fraction range for all other binary mixtures under present investigation. Plot of ΔK_S as the function of difference of boiling points between carbon tetrachloride as first component and benzene, toluene, ethyl benzene and m-xylene as second components of binary mixtures predict the regular varying degree of molecular interactions between hetero molecules. Ultrasonic velocities of binary mixtures calculated from Nomoto's relation[15] and Ideal mixture relation due to Van Deal and Vangeel[16] deviate at all mole fraction range with varying degree of extent from observed values. This gives an evidence of hetero molecular interactions. Other derived parameters like molecular interaction terms (α), Lennard-Jones potential repulsive term exponents (n) and available volumes (V_a) deal with nature and strength of molecular interactions at different composition of binary mixtures. The analysis of observed data of ΔK_S of few binary

mixtures in the light of Flory's theory [17-18] shows its partial success on account of its assumptions and limitations.

MATERIAL AND METHODS

Carbontetrachloride (A.R.,C.D.H.), benzene (A.R,C.D.H), toluene (A.R.,C.D.H), m-xylene (A.R.,C.D.H), ethylbenzene (A.R,C.D.H) and chlorobenzene (A.R.,C.D.H.) were purified by the method mentioned elsewhere[14]. After triple distillation they were stored properly in bottles. The purity of these liquids were checked by density measurement and comparison with literature values[19].

Measurement of Ultrasonic Velocity

Ultrasonic velocity of pure liquids and binary liquid mixtures of different composition was measured using an ultrasonic interferometer (F-81, Mittal enterprises, New Delhi) of frequency 2 MHz with an accuracy of ± 0.037 % previously used by us (1-3). The measurement was carried out at 303.15K with the help of thermostat operating with an accuracy of ± 1%.The working of ultrasonic interferometer was cheeked for its accuracy by meassuring ultrasonic velocity in liquids of known velocity enlisted in literature[20].

Binary liquid mixture of carbon tetrachloride with benzene, toluene, m-xylene, ethyl benzene and chlorobenzene of different composition were prepared by mixing known masses of pure liquids in air tight and narrow mouth ground glass stoppered bottles to minimize the leakage of volatile liquids.

RESULTS AND DISCUSSION

The values of ultrasonic velocity (u), isentropic compressibility (K_S) and deviation of K_S from mole fraction mixture law (ΔK_S) and other derived acoustic parameters like available volume (V_a) , Lennard- Jones potential repulsive term exponent (n) and molecular interaction term (α) along with Nomoto's velocity and Ideal mixture velocity due to Van Deal and Vangeel have been recorded in Table 1 . These parameters were calculated using following relations:

$$K_S = U^{-2} \rho^{-1} \qquad …(1)$$

ρ is the density determined already (12).

$$\Delta K_S = K_S(obsd) - x_1 K_S(1) - x_2 K_S(2) \qquad …(2)$$

where , K_S (1) and K_S (2) and K_S (obsd) are isentropic compressibilities of pure components and binary mixtures respectively.

$$V_a = M/\rho \ [\ 1 - U/U\infty] \qquad …(3)$$

M is molar mass and $U\infty$ is equal to 1600 m s[-1].

$$n = [\ 6V/V_a - 13] \qquad …(4)$$

V is molar volume.

$$\alpha = [(U/U_{im})^2 - 1] \qquad …(5)$$

U_{im} is ultrasonic velocity from Van Deal and Vangeel empirical relation

$$\frac{1}{[x_1 M_1 + x_2 M_2] U^2_{im}} + \frac{x_1}{M_1 U_1^{\,2}} + \frac{x_1}{M_2 U_2^{\,2}} \quad …(6)$$

M_1 and M_2 are molar masses of first and second components of binary mixture. U_1 and U_2 are their ultrasonic velocities.

Another empirical formula for velocity in binary mixtures is due to Nomoto on the assumption of linear dependence of molar sound velocity i.e. Rao's constant R.

$$U_{Nomoto} = \frac{x_1 R_1 + x_2 R_2}{x_1 V_1 + x_2 V_2} \qquad …(8)$$

$$R = (M/\rho)^{1/3} \qquad …(9)$$

Observed values of ultrasonic velocities (U) and isentropic compressibilities (K_S) have been expressed by equation :

$$U = A_0 + A_1 x_1 + A_2 x_1^2 + A_3 x_1^3 \qquad …(9)$$

$$Ks = B_0 + B_1 x_1 + B_2 x_1^2 + B_3 x_1^3 \qquad ….(10)$$

ΔK_S has been expressed by following equation containing three variable constants characteristic of binary mixtures.

Table 1: Ultrasonic properties of binary mixtures for mole fractions x_1 of carbon tetrachloride at 303.15K

x_1	Ultrasonic Velocity/ m s^{-1}	Nomoto's Velocity/ m s^{-1}	Ideal mixture Velocity/ m s^{-1}	$10^{10}Ks$ / pa^{-1}	$10^{10}\Delta Ks$/ Pa^{-1}	V_a/ 10^{-6} m^3 mol^{-1}	α	n
x_1 carbon tetrachloride + x_2 benzene								
0.0000	1290	1289.9	1290	6.90710	0.0000	17.3951	0.0000	17.96
0.1068	1213.4	1242.5	1227.2	7.13751	0.1620	21.8900	-0.0223	11.83
0.2353	1128	1188.0	1162.4	7.50096	0.4433	27.0091	-0.0583	7.33
0.3173	1085.3	1154.6	1126	7.66097	0.5509	29.6499	-0.0709	5.65
0.4148	1040	1116.3	1086.8	7.87136	0.6989	32.5144	-0.0842	4.14
0.5334	994.6	1071.5	1044.1	7.99829	0.7500	35.4783	-0.0925	2.85
0.6092	968	1044.0	1019.2	8.10077	0.8040	37.2510	-0.0979	2.18
0.8216	932	970.9	957.9	7.85823	0.4257	39.9869	-0.0533	1.37
0.9143	924	940.8	934.3	7.66431	0.1725	40.7331	-0.0219	1.20
1.0000	914	913.9	914	7.54661	0.0000	41.5775	0.0000	0.99
x_1 carbon tetrachloride + x_2 toluene								
0.0000	1286	1286.0	1286.0	7.04989	0.0000	21.0825	0.0000	17.57
0.1041	1234.9	1246.9	1231.5	7.07290	-0.0287	24.2618	0.0055	13.29
0.2074	1189.1	1208.3	1182.4	7.09280	-0.0601	27.0266	0.0113	10.36
0.3118	1150.3	1169.2	1137.1	7.06529	-0.1395	29.2692	0.0233	8.34
0.4188	1109.8	1129.2	1094.6	7.08814	-0.1698	31.5601	0.0279	6.58
0.5168	1079.2	1092.6	1058.7	7.06042	-0.2462	33.1955	0.0391	5.43
0.6213	1037.9	1053.8	1023.2	7.17866	-0.1798	35.4453	0.0289	4.07
0.7103	1000.7	1020.7	995.0	7.34162	-0.0611	37.4449	0.0114	3.01
0.8029	971.2	986.4	967.4	7.40894	-0.0398	38.9098	0.0078	2.26
0.9151	937.3	945.1	936.2	7.49464	-0.0098	40.5274	0.0023	1.48
1.0000	914	914.0	914.0	7.54661	0.0000	41.5775	0.0000	0.99
x_1 carbon tetrachloride + x_2 m-xylene								
0.0000	1302	1301.9	1302.0	6.89457	0.0000	23.1114	0.0000	19.19
0.1198	1241	1260.6	1238.9	7.02176	0.0490	27.1464	0.0033	13.74
0.2493	1203.2	1214.4	1177.3	6.88689	-0.1702	29.1887	0.0444	11.19
0.3468	1173.8	1178.6	1134.8	6.81090	-0.3098	30.6698	0.0699	9.52
0.4310	1146.7	1146.9	1100.5	6.77045	-0.4052	31.9637	0.0857	8.17
0.5186	1117.6	1113.2	1067.0	6.76223	-0.4705	33.3300	0.0970	6.90
0.6270	1078	1070.5	1028.2	6.79836	-0.5050	35.0662	0.0992	5.39
0.7100	1042	1036.9	1000.4	6.92728	-0.4302	36.7242	0.0848	4.20
0.8585	983.2	975.0	954.2	7.13986	-0.3145	39.1336	0.0617	2.56
1.0000	914	914.0	914.0	7.54611	0.0000	41.5775	0.0000	0.99
x_1 carbon tetrachloride + x_2 ethyl benzene								
0.0000	1306	1306.0	1306	6.81730	0.0000	22.6845	0.0000	19.65
0.1210	1252.2	1263.5	1241.5	6.85434	-0.0512	26.1533	0.0173	14.60
0.2100	1221.1	1231.5	1198.0	6.80995	-0.1605	27.9387	0.0389	12.33
0.3215	1191.3	1190.3	1147.7	6.67086	-0.3809	29.3829	0.0774	10.48
0.4008	1168.3	1160.4	1114.4	6.60410	-0.5055	30.4684	0.0990	9.23
0.5010	1134.1	1121.6	1074.9	6.59147	-0.5912	32.1059	0.1131	7.60

0.6512	1077.4	1061.7	1020.8	6.67212	-0.6201	34.7073	0.1139	5.36
0.7100	1052.2	1037.7	1001.1	6.75410	-0.5810	35.8551	0.1046	4.52
0.8500	988.3	979.0	957.0	7.04760	-0.3896	38.6023	0.0664	2.69
0.9161	959.2	950.6	937.6	7.19641	-0.2890	39.7339	0.0466	1.98
1.0000	914	914.0	914.0	7.54661	0.0000	41.5775	0.0000	0.99

x_1 carbon tetrachloride + x_2 chlorobenzene

0.0000	1238.4	1238.4	1238.4	5.93146	0.0000	23.1406	0.0000	13.54
0.1214	1191.9	1197.4	1187.2	6.11353	-0.0140	26.0482	0.0079	10.52
0.2295	1151.7	1161.3	1145.1	6.27668	-0.0255	28.4688	0.0115	8.41
0.3541	1126.8	1120.1	1100.1	6.46656	-0.0369	29.7175	0.0491	7.29
0.4571	1073.7	1086.3	1065.5	6.62486	-0.0449	33.0207	0.0154	5.24
0.5552	1042.3	1054.5	1084.5	6.77952	-0.0488	34.7848	0.0151	4.21
0.6646	1010.4	1019.3	1002.0	6.95298	-0.0520	36.6181	0.0168	3.28
0.7730	978.5	984.9	971.7	7.14288	-0.0371	38.3801	0.0140	2.44
0.8798	948.9	951.3	943.6	7.33678	-0.0158	40.0259	0.0112	1.74
0.9742	924.4	921.9	920.2	7.50117	-0.0038	41.3470	0.0091	1.20
1.0000	914	914.0	914.0	7.54661	0.0000	41.5775	0.0000	0.99

Table 2: Fitting constants and the standard deviation σ for the various binary liquid mixtures of carbon tetrachloride at 303.15 K

System	A_0/ms^{-1}	A_1/ms^{-1}	A_2/ms^{-1}	A_3/ms^{-1}	σ/m.s^{-1}
Carbon tetrachloride + benzene	1290	-767.96597	497.27902	-72.49233	2.90620
Carbon tetrachloride + toluene	1286	-475.46708	142.06503	-41.16845	4.06777
Carbon tetrachloride + m-xylene	1302	-457.56019	312.76798	-245.04937	5.27414
Carbon tetrachloride + ethyl benzene	1306	-425.54218	270.41667	-239.44523	4.35027
Carbon tetrachloride +chlorobenzene	1238.4	-356.62622	-3.67551	37.98036	6.09493
System	$10^{10}B_0$/pa^{-1}	$10^{10}B_1$/Pa^{-1}	$10^{10}B_2$/pa^{-1}	$10^{10}B_3$/pa^{-1}	$10^{10}\,\sigma(K_s)$/pa^{-1}
Carbon tetrachloride + benzene	6.90710	2.74552	-0.40338	-1.76527	2.90620
Carbon tetrachloride + toluene	7.04989	-0.19717	0.76614	-0.03889	4.06777
Carbon tetrachloride + m-xylene	6.89457	0.50089	-2.99967	3.17140	5.27414
Carbon tetrachloride + ethyl benzene	6.81730	0.17496	-2.83935	3.40747	4.35027
Carbon tetrachloride + chloro benzene	5.93146	1.49790	-0.02495	0.14557	6.09493

System	$10^{10}C_0$/pa^{-1}	$10^{10}C_1$/Pa^{-1}	$10^{10}C_2$/pa^{-1}	$10^{10}\sigma(\Delta K_s)$/pa^{-1}
Carbon tetrachloride + benzene	3.09143	0.65594	-1.52054	0.05804
Carbon tetrachloride + toluene	-0.81065	0.13301	1.31083	0.05272
Carbon tetrachloride +m-xylene	-1.81754	-1.32519	0.67757	0.05452
Carbon tetrachloride + ethyl benzene	-2.36720	-1.68950	0.83970	0.04033
Carbon tetrachloride + chlorobenzene	-0.19613	-0.06029	0.07980	0.00390

$$\Delta K_s = x_1 x_2 [C_0 + C_1(x_1-x_2) + C_2(x_1-x_2)^2] \qquad ...(11)$$

Sets of fitting constants A_0, A_1, A_2, and A_3, B_0, B_1, B_2 and B_3 and C_0, C_1 and C_2 have been evaluated by the method of least squares. All these constants have been given in Table 2. Standard deviations (σ) for ,U K_s and ΔK_s have also been recorded in the last column of Table2. ó can be obtained from following equation.

$$\sigma = [\Sigma(Y_{obsd}-Y_{calcd})^2/(n-m)]^{1/2} \qquad ...(12)$$

Where n and m are number of observations and total number of fitting constants. Y represents U, K_S and ΔK_S .

ΔK_S equivalent to K_S have been compared with excess isothermal compressibility K_T calculated from statistical theory of binary liquid mixtures by Flory *et al.,* [17-18].

Excess isothermal compressibility is given by the equation (15).

$$K_T^E = \frac{3\tilde{V}^2}{P*}[\ (\tilde{V}^{1/3}-1)^{-1}-3]-[\emptyset_1\tilde{V}_1 K_T(1)+\emptyset_2\tilde{V}_2 K_T(2)] \ ...(13)$$

Where, $K_T(1)$ and $K_T(2)$ are isothermal compressibilities of first and second components respectively. The characteristic pressure P* has been calculated using following equation.

$$P* = \emptyset_1 P_1^* + \emptyset_2 P_2^* - \emptyset_1 \theta_2 X_{12} \qquad ...(14)$$

$\theta_2 X_{12}$ is a pair-interaction parameter. It has been evaluated at mole fraction $x_1 = 0.2, 0.5$ and 0.8
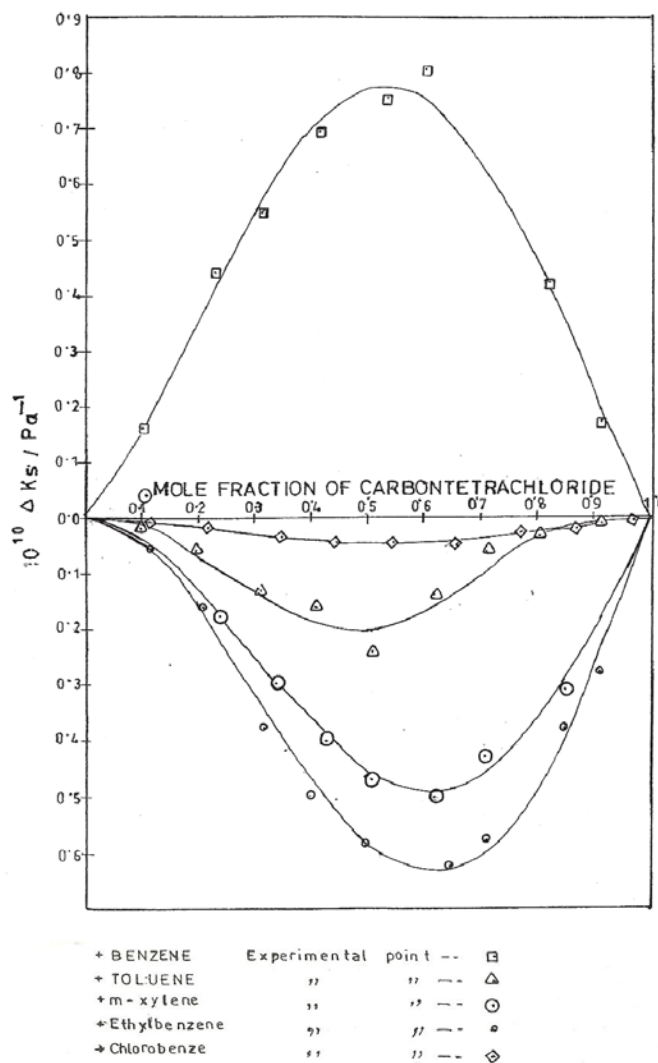
Curves calculated from the equation 1 (Table 2) at 303.15 K

Fig. 1: Excess isentropic compressibility for carbontetra chloride

from excess volumes V^E calculated from observed values of density(14).

Reduced volume V has been defined as

$$\widetilde{V} = \frac{V}{V^*} \qquad \ldots(15)$$

V and V* are molar and characteristic volumes of pure liquids respectively.

$$\emptyset_2 = 1 - \emptyset_1 = \frac{x_2 V_2^*}{x_1 V_1^* + x_2 V_2^*} \qquad \ldots(16)$$

K_T is calculated from relation

$$\gamma = \frac{\alpha}{K_T} \qquad \ldots(17)$$

α and γ are coefficient of thermal expansion and isothermal pressure coefficient of liquid respectively.

$$P^* = \gamma T \, \widetilde{V}^{\,2} \qquad \ldots(18)$$

Parameters V, α, γ, P*, V*, , T* and T for pure liquids at 303.15 K has been placed in Table 3.

Due to unavailability of excess molar heat capacity (C_p) of binary mixtures concerned, Flory based K_T^E values could not be converted into K_s by the method used by Holder and W. Halley[21]. K_T values (Flory) and observed ΔK_s values at $x_1 = 0.2$, 0.5 and 0.8 for binary mixtures of carbon tetrachloride with benzene, toluene and m-xylene have been recorded in Table 4. Due to unavailability of parameters for ethyl benzene and chlorobenzene, we could not be
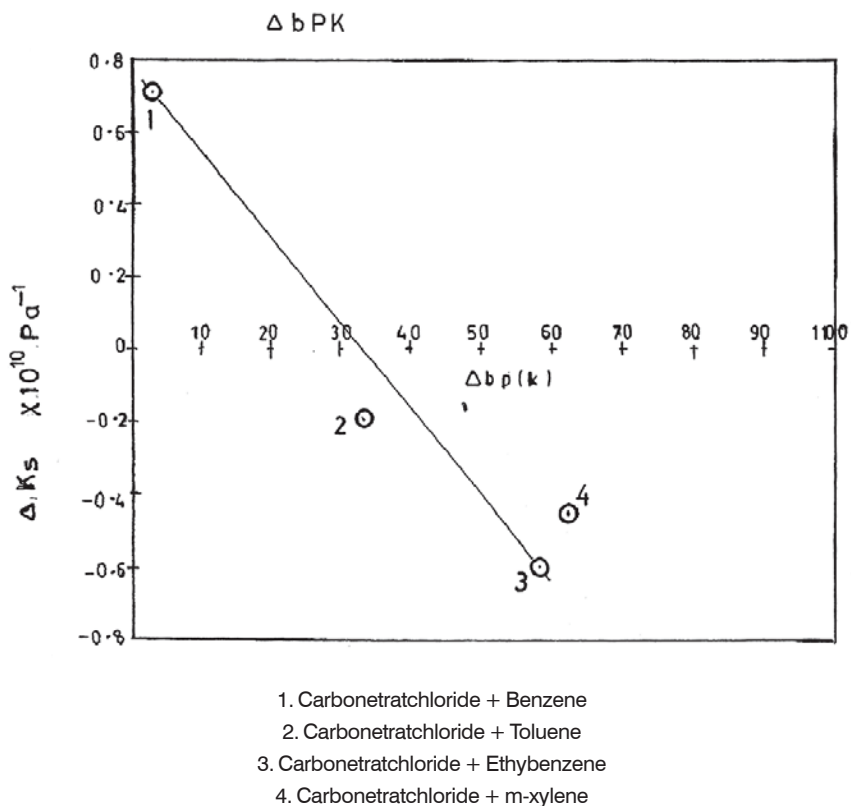

1. Carbonetratchloride + Benzene
2. Carbonetratchloride + Toluene
3. Carbonetratchloride + Ethybenzene
4. Carbonetratchloride + m-xylene

Fig. 2: Plot of D Ks Vs D bp (k) for equimola mixture

able to calculate theoretical values of K_T^E (Flory) for these binary systems. Flory's theory predicts the negative sign of K_T^E. As we notice, negative sign of ΔK_S for carbon tetrachloride + toluene and + m-xylene but positive sign for carbon tetracholoride + benzene predicts the partial success of Flory's theory despite the fact that it deals with mixture of such molecules which differ in shape and size however, with certain limitation and approximations. Let us now analyze the experimental values of ultrasonic velocity of binary mixtures given in Table 1 in the light of theoretical values computed by theories of Nomoto and Van Deal and Vangeel. Both Nomoto's and ideal mixture velocity due to Van Deal *et al.*, deviate appreciably from experimental velocity at almost entire mole fraction range. This may be due to limitations and approximations incorporated in these theories Nomoto's velocity expression is based on the assumption that molecules are spherical and there is no change in volume upon mixing of two liquids, which is possible only in ideal mixtures where there is no molecular

interaction. Similarly, the assumption of Van Deal and Vangeel of same ratio of specific heats of components both in pure and combined state and unchanged volumes of components, lead to error in computation of ultrasonic velocity. All these facts and discussions lead to the fact that the molecular interaction is present in all binary mixtures caused by dispersive, dipole-dipole, dipole-induced dipole interactions, charge transfer, hydrogen bonding etc.

As we notice the data given in Table1, ultrasonic velocity decreases and isentropic compressibility increases with increasing mole fraction (x_1) in all five binary mixtures. ΔK_S values for remaining four binary mixtures except carbon tetrachloride + benzene are found to be negative in sign at entire mole fraction range.

10^{10} ΔK_S .Pa for our equimolar binary mixtures of carbon tetrachloride + benzene, +toluene, + m-xylene, + ethylbenzene and + chlorobenzene at 303.15K are 0.7728, - 0.2026, - 0.4543,- 0.5918 and -0.0490 respectively. Thus,

Table 3: Parameters for the pure component liquids at 30.3 15K

Liquid	V/cm³ mol⁻¹	α×10³/K⁻¹	γ/cal. cm⁻³K⁻¹	V*/cm³. mol⁻¹	V*/cm³. mol⁻¹	T*K	P*/cal. cm⁻³	
Carbon-tetrachloride	97.68	1.240	0.263	1.2986	75.23	4748	137	0.06386
Benzene	89.95	1.233	0.292	1.2975	69.33	4730	149	0.06408
Toluene	107.424	1.085	0.278	1.2684	84.69	5046	135.5	0.06007
m-xylene	124.094	1.008	0.270	1.2527	99.06	5249	128.4	0.05775

Table 4: Comparison of calculated and observed excess compressibility at 303.15K

System	x_1	$\theta_2 x_{12}$ (cal cm⁻³)	$-K_T^E \times 10^{10}$/ pa⁻¹ (Calculated)	$\Delta K_S \times 10^{10}$/ pa⁻¹ (Observed)
Carbon tetrachloride + benzene	0.2	0.8973	0.898067	0.344075
	0.5	1.2501	0.869112	0.772857
	0.8	1.1908	0.840779	0.470015
Carbon tetrachloride + toluene	0.2	0.7703	0.889071	-0.066969
	0.5	0.8285	0.834502	-0.202662
	0.8	0.6192	0.784465	-0.041431
Carbon tetrachloride + m-xylene	0.2	1.3191	0.885929	-0.124560
	0.5	1.0196	0.827425	-0.454385
	0.8	3.1914	0.757060	-0.378996

ΔK_S varies in following order

Carbon tetrachloride + benzene > + chlorobenzene> + toluene> + m-xylene>+ethlybenzene.

Comparative plots of ΔK_S versus mole fraction of carbon tetrachloride in all five binary mixtures have been shown in fig. 1.

The molecular interaction terms for carbon tetrachloride + benzene are negative in sign at entire mole fraction range with least values at x_1=0.60. It shows the dominance of dispersive forces. On the other hand α are found positive in sign with varying magnitude at entire mole fraction range in all remaining mixtures. Maximum values of á are found in the vicinity of x_1 = 0.5 to 0.6 in following order,

Carbon tetrachloride + benzene, <+chlorobenzene, < + toluene,< + m-xylene,< + ethyl benzene

The Lennard -Jones potential is given by relation :

$$\phi\,(r) = [\,-A/r^6 + D/r^n] \qquad \ldots(19)$$

n is the individual exponent for each liquid describing the magnitude of repulsive forces therein. It may vary from one liquid to other and with composition of binary mixtures. r is intermolecular distance. As we see, in all binary mixtures n decreases from their respective values to 0.99 with increasing mole fraction (x_1). This shows the dominance of repulsive forces over attractive forces in carbon tetrachloride rich zone of binary mixtures.

Available volumes $(10^6 V_a/m^3 mol^{-1})$ increases respectively from 17.3951, 21.0825 , 23.1114 , 22.6845 and 23.1406 to 41.5775 with increasing mole fraction (x_1) in binary mixtures of carbon tetrachloride with benzene, toluene, m-

xylene, ethyl benzene and chloro benzene.

Negative ΔK_S may result from two contributions arising from interaction and free volume factors. Interactions due to dipole-dipole and dipole-induced dipole type enhances the solvent structure in mixture[22]. Solvent structure of substituted benzenes like toulenes, m-xylene, ethyl benzenes and chloro benzene is ruptured by the addition of nonpolar carbon tetrachloride, which may result expansion upon mixing. But, the specific interactions between unlike molecules is larger than the expansion caused by the breaking of dipolar order in pure liquid molecules. In case of carbon tetrachloride + benzene usual dispersive interaction dominate over specific interactin there in. Benzene and substituted benzenes act as sacrificial π donors towards carbon tetrachloride as sacrificial σ - acceptor. Such type of EDA interactions result in production of negative values of ΔK_S. Progressively stronger EDA interaction resulting from increasing π - electron density due to inclusion of methyl and ethyl groups to benzene nucleus results in more and more negative value of ΔK_S as shown already for equimolar mixtures. Least negative values of ΔK_S in case of chlorobenzene may be due to presence of electronegative chlorine atom. Moreover, size of molecules involved in mixture formation may be other dominating factor in deciding the sign and magnitude of ΔK_S.

Thacker and Rowlinson[23], Reddy *et at.*,[24] and Fort and Moore[25] have used the difference in boiling points of components as a measures of strength of interaction. In fig 2, the values of ΔK_S for equimolar mixtures have been plotted vs the difference (Δbp.K) in boiling points of second component and carbon tetrachloride. The plot is almost linear with some scatter. It shows again the increasing strength of interaction of carbon tetrachloride with benzene and substituted benzenes.

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