Excess Volumes, Speeds of Sound, Isentropic Compressibilities, and Viscosities of Binary Mixtures of Acetophenone with Chlorotoluenes and Nitrotoluenes at 303.15 K

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Excess volumes (V^E), ultrasonic sound velocities (u), and viscosities (η) have been measured for the binary mixtures of acetophenone with *o*-chlorotoluene, *m*-chlorotoluene, *p*-chlorotoluene, *o*-nitrotoluene, and *m*-nitrotoluene at 303.15 K. Excess volumes are positive over the entire composition range in all the binary mixtures. Furthermore, from the experimental sound velocity data isentropic compressibility (K_S) and deviation in isentropic compressibility (ΔK_S) from ideal behavior was also computed at the same temperature, and these values are negative in all the binary mixtures. The viscosity data are analyzed on the basis of corresponding states approach. Deviation in viscosities are positive over the entire composition range. The measured data are explained on the basis of intermolecular interactions between unlike molecules.

Introduction

Thermodynamic excess properties of binary liquid mixtures have been very useful in obtaining information on the intermolecular interactions and geometrical effects in the systems.[1,2] Furthermore, thermodynamic properties of binary mixtures, containing components capable of undergoing specific interactions, exhibit significant deviation from ideality arising not only from difference in molecular size and shape but also due to structural changes. A survey of the literature has shown that V^E data for the binary mixtures of acetophenone with 1-alkanols,[3] benzene and substituted benzenes,[4] and chloroethanes and chloroethenes[5] have been reported. It is well-known that the ultrasound technique has a wide range of applications such as in industry and in equipment testing. Molecular association in solution, correlation of sound speed with various parameters, isentropic compressibilities, free volume, etc. can be studied by measurement of speeds of sound in liquid mixtures. Also isentropic compressibility data for the mixtures of acetophenone with 1-alkanols,[3] benzene and substituted benzenes,[6] and chloroethanes and chloroethenes[7] were also reported earlier at 303.15 K. The experimental sound velocity data have been analyzed in terms of free length theory (FLT)[8] and collision factor theory (CFT).[9]

As far as we are aware, no literature data are available on excess volume, sound velocity, and viscosity data of acetophenone with chlorotoluenes and nitrotoluenes. The present study was undertaken to determine the effects of addition of a chloro group and a nitro group in the toluene nucleus that may influence both the sign and the magnitude of excess volume and deviation in isentropic compressibility and deviation in viscosities.

Experimental Section

All the liquids used in the present investigation were of AnalaR grade (S.D Fine Chemicals Ltd., India) purified as described in the literature.[10,11] The purity of the sample was checked by comparing the measured densities with those reported in the literature,[11−16] and these are given in Table 1. Excess volumes (V^E) were measured directly using a single composition loading type dilatometer as described by Prabhakara Rao and Naidu.[17] Four dilatometers with different capacities ranging from 0.2 to 0.8 mole fractions were used to cover the entire composition range. The dilatometer was kept in a thermostat maintained at (303.15 ± 0.01) K. The uncertainity in measured V^E values has been found to be ± 0.003 $\text{cm}^3\cdot\text{mol}^{-1}$.

Ultrasonic sound velocities were measured with a single-crystal interferometer (Mittal Enterprise, New Delhi, M-82) at a frequency of 3 MHz with an uncertainity of ± 0.1 %. The densities of pure liquids were measured using a single-capillary pycnometer (made of Borosil glass), described earlier.[18] The marks on the capillary were calibrated by using triple distilled water. The uncertainity in density measurements was found to be ± 0.0001 $\text{g}\cdot\text{cm}^{-3}$. The viscosities of pure liquids and liquid mixtures were determined by a suspended Ubbelohole-type viscometer, described earlier.[19] The estimated uncertainity in viscosity is ± 0.001 mPa·s. All the measurements were made using a thermostatic bath maintained at (303.15 ± 0.01) K.

Results and Discussion

The measured excess volumes (V^E) data of binary mixtures of acetophenone with chlorotoluenes and nitrotoluenes are given in Table 2. The V^E data for the mixtures of acetophenone with chlorotoluenes and nitrotoluenes are graphically represented in Figures 1 and 2, respectively, along with those binary mixtures of acetophenone with toluene[4] for the mixtures of acetophenone with chlorotoluenes and nitrotoluenes. The V^E values were fitted to an empirical relation proposed by Redlich−Kister:[20]

$$V^E/\text{cm}^3\cdot\text{mol}^{-1} = x_1(1 - x_1)[a_0 + a_1(2x_1 - 1) + a_2(2x_1 - 1)^2] \quad (1)$$

where a_0, a_1, and a_2 are the adjustable parameters and x_1 is the

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Table 1. Comparison of Experimental Densities and Viscosities of Pure Liquids with Literature Values at 303.15 K

	ρ/g·cm^{-3}		η/mPa·s	
liquid	lit$^{11-16}$	exptl	lit$^{11-16}$	exptl
acetophenone	1.01940	1.01943	1.511	1.513
o-chlorotoluene	1.07279	1.07277	0.887	0.884
m-chlorotoluene	1.06524	1.06522	0.780	0.782
p-chlorotoluene	1.06052	1.06054	0.900	0.902
o-nitrotoluene	1.15321	1.15326	1.911	1.908
m-nitrotoluene	1.14766	1.14762	1.777	1.775

mole fraction of acetophenone. The values of parameters were obtained by the least-squares method.

Data for density (ρ), computed from measured excess volumes[21] and experimental sound velocity data (u), are included in columns 2 and 3 of Table 3. Isentropic compressibility (K_S) and the deviation in isentropic compressibility (ΔK_S) are also included in columns 4 and 5 of Table 3. The deviation in isentropic compressibility (ΔK_S) values are graphically represented in Figures 3 and 4 against the volume fraction along with those of acetophenone with toluene, which was reported earlier.[6] The isentropic compressibility (K_S) and the deviation in isentropic compressibility (ΔK_S) were calculated employing in the following relations:

$$K_S = u^{-2}\rho^{-1} \tag{2}$$

$$\rho = (x_1M_1 + x_2M_2)/V^0 + V^E \tag{3}$$

$$\Delta K_S = K_S - (\Phi_1K_{S1} + \Phi_2K_{S2}) \tag{4}$$

$$\phi_1 = x_1{V_1}^0/(x_1{V_1}^0 + x_2{V_2}^0) \text{ and } \phi_2 = 1 - \phi_1 \tag{5}$$

$$V^0 = x_1{V_1}^0 + x_2{V_2}^0$$

where x, M, and V^0 denote the mole fraction, molecular weight, and molar volume of the mixture; K_S, K_{S1}, and K_{S2} denote compressibilities of the mixtures and components 1 and 2, respectively. ΔK_S values indicate the deviation in isentropic compressibility from ideal value, which is assumed to be additive in terms of volume fraction; ϕ_1 and ϕ_2 denote the volume fractions of the components 1 and 2, respectively; and ρ is the density of the mixture.

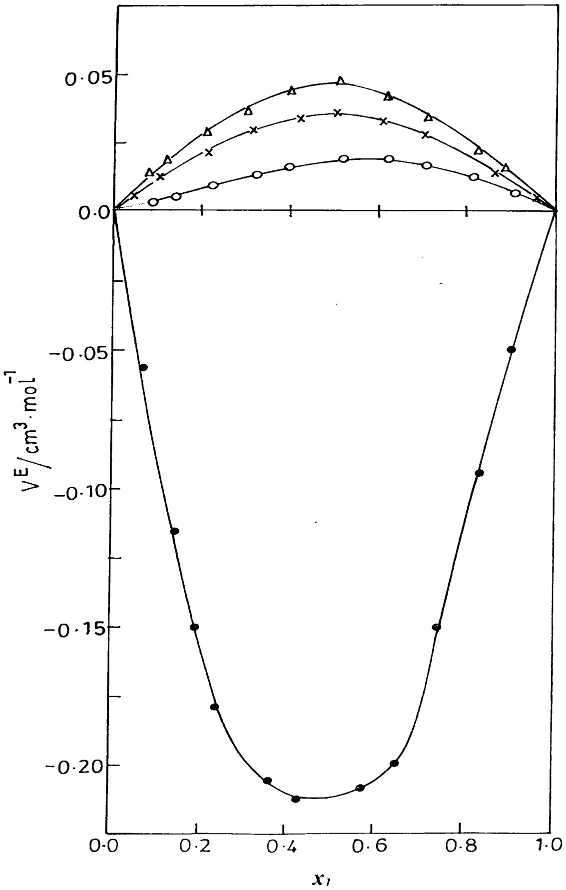

Figure 1. Plots of excess volume V^E vs mole fraction of acetophenone (x_1) + ○, *o*-chlorotoluene; ×, *m*-chlorotoluene; △, *p*-chlorotoluene; ●, toluene at 303.15 K.

The dependence of ΔK_S on volume fraction is expressed by an empirical equation:

$$\Delta K_S/\text{TPa}^{-1} = \phi_1\phi_2[b_0 + b_1(\phi_1 - \phi_2) + b_2(\phi_1 - \phi_2)^2] \tag{6}$$

Table 2. Mole Fraction of Acetophenone (x_1), Experimental and Predicted Excess Volumes (V^E) for the Binary Mixtures of Acetophenone (1) with Chlorotoluenes and Nitrotoluenes (2) at 303.15 K

	V^E	V^E (calcd)		V^E	V^E (calcd)		V^E	V^E (calcd)
x_1	cm^3 mol^{-1}	cm^3 mol^{-1}	x_1	cm^3 mol^{-1}	cm^3 mol^{-1}	x_1	cm^3 mol^{-1}	cm^3 mol^{-1}
Acetophenone (1) + *o*-Chlorotoluene (2)			Acetophenone (1) + *m*-Chlorotoluene (2)			Acetophenone (1) + *p*-Chlorotoluene (2)		
0.0901	0.006	0.006	0.0524	0.010	0.009	0.087	0.016	0.015
0.1510	0.009	0.009	0.1030	0.015	0.015	0.1206	0.0.20	0.020
0.2271	0.013	0.013	0.2141	0.023	0.026	0.2175	0.027	0.030
0.3205	0.016	0.016	0.3119	0.029	0.030	0.3009	0.034	0.036
0.4021	0.018	0.018	0.4266	0.033	0.033	0.4017	0.041	0.040
0.5219	0.020	0.020	0.5009	0.034	0.032	0.5224	0.044	0.040
0.6224	0.020	0.019	0.6174	0.032	0.030	0.6211	0.039	0.037
0.7020	0.018	0.018	0.7050	0.028	0.026	0.7025	0.032	0.032
0.8194	0.015	0.014	0.8519	0.015	0.016	0.8214	0.021	0.022
0.9005	0.009	0.009	0.9520	0.006	0.006	0.8774	0.016	0.016
Acetophenone (1) + *o*-Nitrotoluene (2)			Acetophenone (1) + *m*-Nitrotoluene (2)					
0.0812	0.011	0.011	0.0984	0.018	0.017			
0.1019	0.015	0.014	0.1174	0.021	0.020			
0.2147	0.024	0.026	0.2074	0.030	0.032			
0.3265	0.033	0.035	0.3115	0.039	0.042			
0.4219	0.039	0.039	0.4225	0.047	0.047			
0.5109	0.042	0.039	0.5417	0.050	0.047			
0.6217	0.037	0.035	0.6478	0.043	0.041			
0.7315	0.026	0.026	0.7105	0.035	0.035			
0.7926	0.020	0.020	0.8016	0.025	0.025			
0.8714	0.011	0.012	0.8874	0.014	0.014			

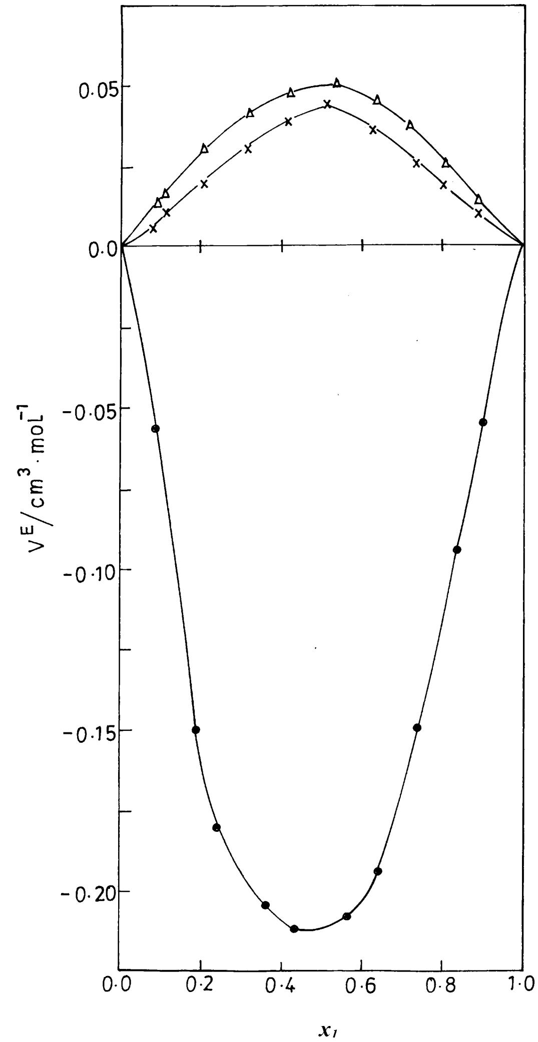

Figure 2. Plots of excess volume V^E vs mole fraction of acetophenone (x_1) + ×, *o*-nitrotoluene; △, *m*-nitrotoluene; ●, toluene at 303.15 K.

where b_0, b_1, and b_2 are the adjustable parameters. These values are calculated by the method of least squares.

The sound velocity data predicted in terms of FLT and CFT along with the experimental results are graphically represented in Figures 5 to 9. The methods and the details of calculations are discussed earlier.[22]

The viscosities of the mixtures[23] were calculated using the equation

$$\eta = k_v \cdot \rho \cdot t \tag{7}$$

where k_v is the viscometer constant and ρ and t are the density and flow time, respectively. The densities of mixtures were calculated from eq 3. Deviation in viscosities ($\Delta \ln \eta$) of the mixtures[23] were obtained by means of the following relation:

$$\Delta \ln \eta/\text{mPa}\cdot\text{s} = \ln \eta_{mix} - x_1 \ln \eta_1 - x_2 \ln \eta_2 \tag{8}$$

where η_{mix} is the viscosity of the mixture; η_1 and η_2 are the viscosities of acetophenone and chlorotoluenes or nitrotoluenes, respectively. Deviations in viscosity versus mole fraction of acetophenone for the mixtures of chlorotoluenes and nitrotoluenes are plotted in Figures 10 and 11. The densities, viscosities, and deviations in viscosities are listed in the Table 3. The experimental values of $\Delta \ln \eta$ are fitted to the Redlich−Kister equation[20] of the type

$$\Delta \ln \eta/\text{mPa}\cdot\text{s} = x_1 x_2[c_0 + c_1(x_1 - x_2) + c_2(x_1 - x_2)_2] \tag{9}$$

where c_0, c_1, c_2, ... are the adjustable parameters.

The sign of excess volume (V^E) of a system depends on the relative magnitude of expansion/contraction on mixing of two liquids. If the factors causing expansion dominate the factors causing contraction, then V^E becomes positive. On the other hand, if the contractive factors dominate the expansive factors, then V^E become negative. The factors that are responsible for expansion in volume are as follows:

(1) Loss of dipolar association (i.e., rupturing of H-bonding of component by the other or breaking up of associates held by

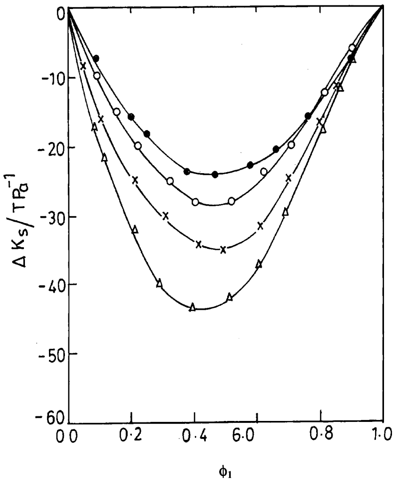

Figure 3. Plots of deviation in compressibility ΔK_S vs volume fraction of acetophenone (ϕ_1) + ○, *o*-chlorotoluene; ×, *m*-chlorotoluene; △, *p*-chlorotoluene; ●, toluene 303.15 K.

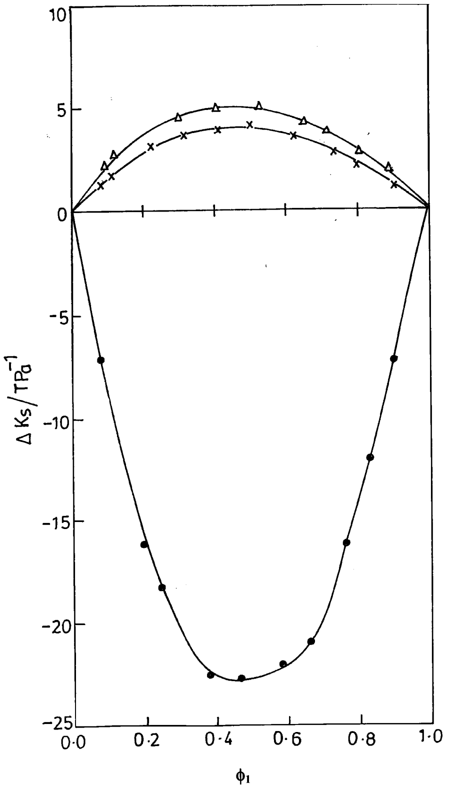

Figure 4. Plots of deviation in compressibility ΔK_S vs volume fraction of acetophenone (ϕ_1) + ×, *o*-nitrotoluene; △, *m*-nitrotoluene 303.15 K.

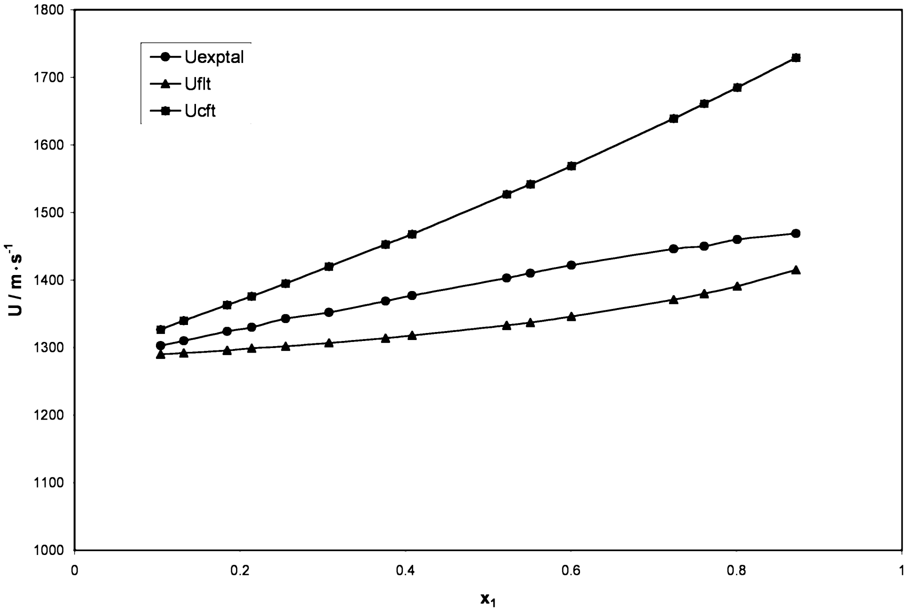

Figure 5. Experimental and correlated sound velocities for the binary mixture acetophenone (1) + *o*-chlorotoluene (2).

Table 3. Volume Fraction (ϕ_1) of Acetophenone, Density (ρ), Sound Velocity (u), Isentropic Compressibility (K_S), and Deviation in Viscosity ($\Delta \ln \eta$) of Acetophenone (1) with Chlorotoluene and Nitrotoluene (2) at 303. 15 K

ϕ_1	ρ / g·cm^{-3}	u / m·s^{-1}	K_S / TPa^{-1}	η_{exp} / mPa·s	η_{calcd} / mPa·s	$\Delta \ln \eta$ / mPa·s	ϕ_1	ρ / g·cm^{-3}	u / m·s^{-1}	K_S / TPa^{-1}	η_{exp} / mPa·s	η_{calcd} / mPa·s	$\Delta \ln \eta$ / mPa·s
Acetophenone (1) + *o*-Chlorotoluene (2)							Acetophenone (1) + *m*-Chlorotoluene (2)						
0.0000	1.07277	1284	565	0.887		0.000	0.0000	1.06524	1280	573	0.782		0.000
0.0900	1.06802	1310	545	0.901	0.896	0.005	0.0520	1.06282	1297	559	0.891	0.886	0.004
0.1508	1.06471	1326	534	0.949	0.948	0.010	0.1022	1.06045	1315	545	0.901	0.901	0.012
0.2269	1.06063	1345	521	0.999	0.994	0.020	0.2127	1.05531	1346	523	1.002	1.000	0.031
0.3202	1.05565	1368	506	1.066	1.060	0.033	0.3101	1.05083	1370	507	1.073	1.072	0.045
0.4018	1.05126	1387	494	1.123	1.121	0.045	0.4246	1.04554	1395	491	1.160	1.159	0.058
0.5216	1.04481	1409	482	1.225	1.223	0.059	0.4988	1.04216	1412	481	1.222	1.220	0.068
0.6221	1.03954	1423	475	1.305	1.303	0.065	0.6154	1.03682	1431	471	1.310	1.305	0.072
0.7017	1.03521	1432	471	1.410	1.405	0.057	0.7033	1.03281	1438	468	1.370	1.369	0.068
0.8192	1.02908	1448	466	1.465	1.460	0.047	0.8508	1.02626	1449	464	1.458	1.451	0.047
0.9004	1.02473	1451	463	1.502	1.500	0.030	0.9516	1.02173	1455	462	1.503	1.497	0.021
1.0000	1.01940	1460	460	1.511		0.000	1.0000	1.01940	1460	460	1.511		0.000
Acetophenone (1) + *p*-Chlorotoluene (2)							Acetophenone (1) + *o*-Nitrotoluene (2)						
0.0000	1.06052	1271	584	0.900		0.000	0.0000	1.15321	2100	196	1.911		0.000
0.0807	1.05717	1304	556	0.903	0.900	0.013	0.0805	1.14236	2017	215	1.910	1.905	0.005
0.1193	1.05554	1314	548	1.011	1.010	0.022	0.1011	1.13962	1997	220	1.903	1.900	0.006
0.2153	1.05143	1343	525	1.058	1.056	0.040	0.2132	1.12454	1889	249	1.857	1.852	0.010
0.2982	1.04804	1371	507	1.118	1.114	0.053	0.3245	1.10951	1803	277	1.817	1.816	0.016
0.3986	1.04382	1406	491	1.198	1.195	0.068	0.4197	1.09673	1737	302	1.781	1.781	0.020
0.5192	1.03881	1419	477	1.286	1.281	0.081	0.5086	1.08482	1681	326	1.747	1.742	0.023
0.6181	1.03483	1430	470	1.352	1.350	0.083	0.6196	1.07006	1622	355	1.701	1.700	0.024
0.6998	1.03154	1440	467	1.400	1.401	0.075	0.7297	1.05544	1568	385	1.652	1.624	0.022
0.8195	1.02673	1448	464	1.469	1.468	0.062	0.7911	1.04736	1541	402	1.624	1.622	0.020
0.8760	1.02446	1453	463	1.496	1.491	0.053	0.8704	1.03673	1508	424	1.582	1.583	0.014
1.0000	1.01940	1460	460	1.511		0.000	1.0000	1.01940	1460	460	1.511		0.000
Acetophenone (1) + *m*-Nitrotoluene (2)													
0.0000	1.14766	2086	200	1.775									
0.0972	1.13506	1987	223	1.748	1.746	0.008							
0.1160	1.13263	1972	227	1.745	1.742	0.009							
0.2051	1.12115	1892	249	1.731	1.730	0.015							
0.3085	1.10782	1808	276	1.713	1.710	0.020							
0.4191	1.09351	1737	303	1.694	1.693	0.026							
0.5383	1.07823	1666	334	1.666	1.664	0.028							
0.6446	1.06475	1608	363	1.640	1.640	0.028							
0.7076	1.05671	1580	379	1.623	1.620	0.027							
0.7994	1.04504	1539	404	1.594	1.591	0.023							
0.8860	1.03403	1503	428	1.564	1.563	0.017							
1.0000	1.01940	1466	460	1.511		0.000							

weaker forces, namely, dipole−dipole or dipole−induced dipole interactions or by van der Waals forces).

(2) The geometry of molecular structure, which does not allow fitting of one component in to other component.

(3) Steric hindrance, which opposes the proximity of the constituent molecules.

The negative V^E values arise due to dominance of the following factors:

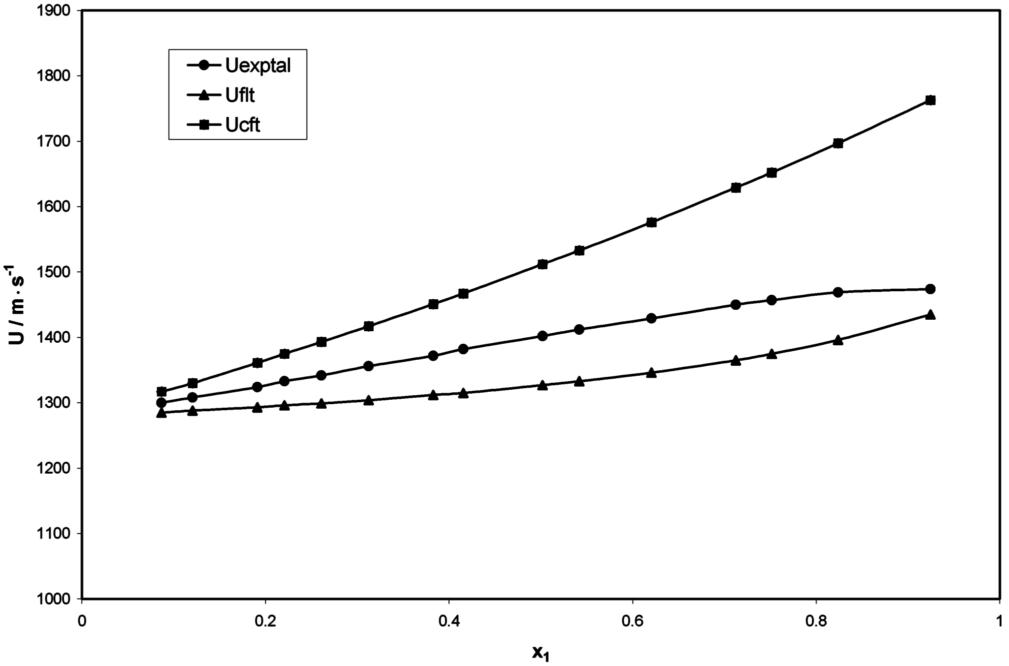

Figure 6. Experimental and correlated sound velocities for the binary mixture acetophenone (1) + *m*-chlorotoluene (2).

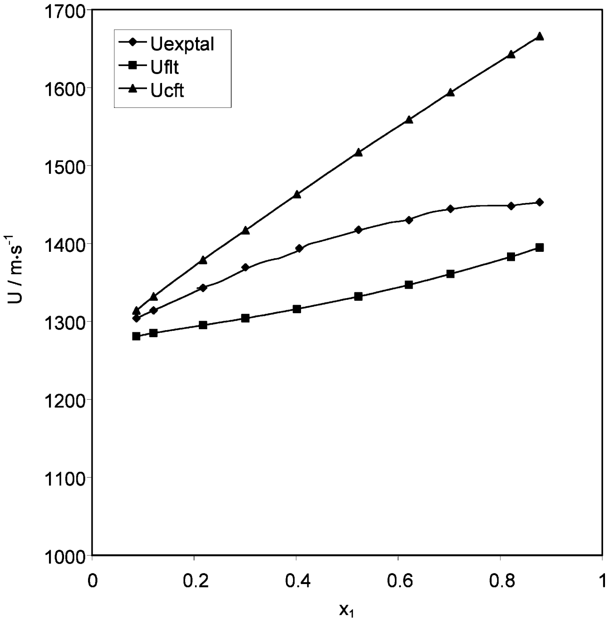

Figure 7. Experimental and correlated sound velocities for the binary mixture acetophenone (1) + *p*-chlorotoluene (2).

(1) Chemical interaction between constituent molecules, such as heteromolecular association through the formation of H-bond, known as strong specific interaction.

(2) Accommodation of molecules of one component into the interstitials of the molecules of the other component.

(3) Geometry of the molecular structure that favors fitting of the component molecules with each other.

The experimental results in the present investigation indicate that the factors which are responsible for expansion in volume are dominant over the entire composition range in all the binary mixtures of acetophenone with chlorotoluenes and nitrotoluenes.

The carbonyl group is highly polar and so has a high percentage of ionic character.[24] There is a negative charge on the carbonyl oxygen atom of acetophenone. Furthermore, negative excess volumes were reported in the literature[4] for the binary mixtures of acetophenone with toluene and chlorobenzene. The weaker interactions in all the binary mixtures may be attributed to the internal interaction between the π-electrons of C═O bond and the π-electrons of the benzene ring.[25] Similar behavior of two adjacent groups, one being phenyl group, has been observed in aromatic ethers and aromatic chlorides. Furthermore, it has been shown that the intramolecular interactions between phenyl and oxygen groups in aromatic ethers[26] and between phenyl and chlorine groups in aromatic chlorides[27] become weaker when the functional groups are separated by at least two methylene groups.

The π-electron density in the derivatives of benzene ring depends on nature of the group that is attached to it and the heteronuclear interaction between component molecules depends on the net electron density in the ring.

Thus, the intermolecular interactions in ortho positions become more than the meta and para isomers due to which polar character will be more in ortho isomer. The algebraic values of V^E for the binary mixtures of acetophenone with chlorotoluenes fall in the order:

p-chlorotoluene > *m*-chlorotoluene > *o*-chlorotoluene

An examination of curves in Figure 1 suggest that V^E data are positive for the binary mixtures of acetophenone with chlorotoluenes. This is due to the factors which are responsible for expansion in volume are dominant over the entire composition range in the binary mixtures.

The V^E data in Figure 2 may be explained if it is assumed that the acetophenone and nitrotoluene mixtures are characterized by interactions between the π-electron cloud of benzene ring with the delocalized π-electron cloud over the nitrogen and oxygen atoms[28] of the nitro group of nitrotoluenes resulting in the formation of weak P-complex.[29]

Since meta substituents are far away as compared to ortho substituents, the polar character decreases, and as expected V^E values are increased. The algebraic excess volume data for the mixtures of acetophenone with nitrotoluene fall in the order:

o-nitrotoluene < *m*-nitrotoluene

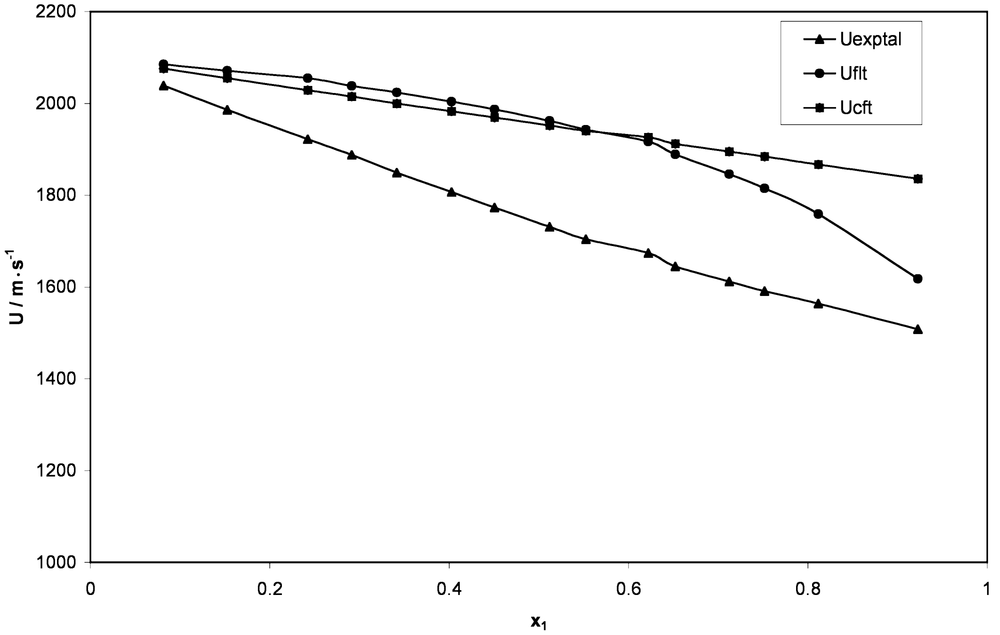

Figure 8. Experimental and correlated sound velocities for the binary mixture acetophenone (1) + *o*-nitrotoluene (2).

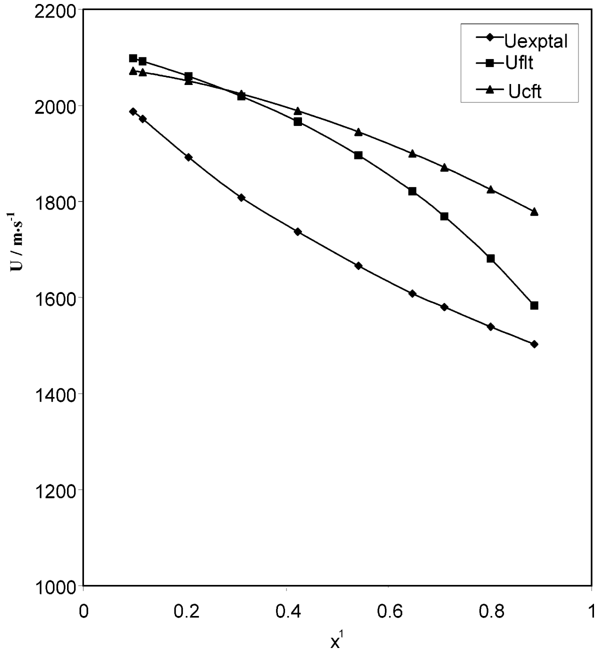

Figure 9. Experimental and correlated sound velocities for the binary mixture acetophenone (1) + *m*-nitrotoluene (2).

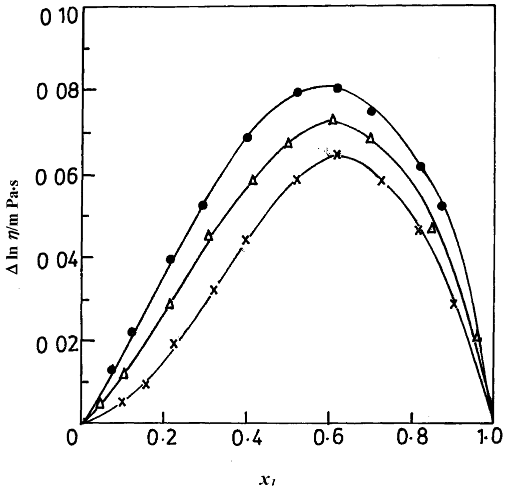

Figure 10. Plots of deviation in viscosity △ ln η vs mole fraction of acetophenone (x_1) + ×, *o*-chlorotoluene; △, *m*-chlorotoluene; ●, *p*-chlorotoluene at 303.15 K.

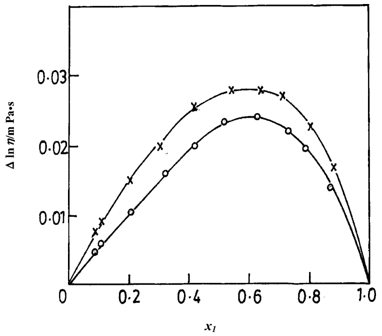

Figure 11. Plots of deviation in viscosity △ ln η vs mole fraction of acetophenone (x_1) + ○, *o*-nitrotoluene; ×, *m*-chlorotoluene; at 303.15 K.

The sign and magnitude of V^E data for the mixtures of acetophenone with nitrotoluenes are smaller than those of mixtures of acetophenone with toluene. This is due to introduction of a nitro group in toluene molecule is influencing the sign and magnitude of V^E to a significant extent.

A comparison of V^E of binary systems containing acetophenone with chlorotoluenes and acetophenone with nitrotoluenes show that ortho-substituted compounds are exhibiting lower V^E values than meta-substituted derivatives probably it is because of lowering of polar character of meta derivatives than ortho derivatives.

An examination of deviation in isentropic compressity (ΔK_S) data in Figures 3 and 4 suggest that the quantity is negative over the entire composition range for the binary mixtures of acetophenone with chlorotoluenes and nitrotoluenes. These may be attributed to the relative strength of effects which influenced the free space between component molecules, described in the literature earlier.[30] The negative ΔK_S values arise from changes of "free volume" in the real mixture and presence of π-electrons in acetophenone resulting in the formation of weak intermolecular complexes[31] leading to positive deviation in sound velocity and negative deviation in isentropic compressibility.

The algebraic ΔK_S values for the systems that contain chlorotoluenes and nitrotoluenes fall in the order:

o-chlorotoluene < *m*-chlorotoluene < *p*-chlorotoluene

o-nitrotoluene < *m*-nitrotoluene

The above order suggests that addition of chloro and nitro groups in the toluene molecule is influencing the sign and magnitude of deviation in isentropic compressibility to a significant extent.

A perusal of Table 3 shows that the values of $\Delta \ln \eta$ are positive over the entire range of composition in all the binary mixtures of acetophenone with chlorotoluenes and nitrotoluenes. The deviation in viscosity of a mixture[32,33] depends on the molecular interactions between the components: mixtures with strong interactions between different molecules show positive viscosity deviations; while for mixtures without specific strong interactions, viscosity deviations are negative.

The positive viscosity in the present investigation suggest that involvement of specific interactions such as formation of H-bonds, change transfer complexes, or strong dipole interactions between component molecules[34] over the entire composition range in all the binary mixtures. The algebraic $\Delta \ln \eta$ values for the systems containing chlorotoluenes and nitrotoluenes fall in the order:

o-chlorotoluene < *m*-chlorotoluene < *p*-chlorotoluene

o-nitrotoluene < *m*-nitrotoluene

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