



## **Ultrasonic studies of molecular interaction of alcohols with non-polar solvents**

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### **ABSTRACT**

*The ultrasonic velocity, density and viscosity for binary liquid mixtures of the n-alcohols namely 1-pentanol, 1-hexanol and 1-heptanol in toluene, carbon tetra chloride, benzene and dioxane at 301K have been measured. From these measured parameters adiabatic compressibility, free length, free volume and their excess values have been measured. The results have been analyzed and interpreted in terms of molecular interactions such as dipole-dipole interaction through hydrogen bonding between solvents and 1-alkanols have been observed. Further the experimental ultrasonic velocities of all these binary systems are compared with theoretical ultrasonic velocity.*

**Key words:** ultrasonic velocity, density, Binary mixtures, dipole- dipole interaction.

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### **INTRODUCTION**

Ultrasonic methods find extensive applications for characterizing aspects of physicochemical behaviour such as the nature of molecular interactions in pure liquids as well as liquid mixtures[1-5]. The study of the solution properties of liquid mixtures consisting of polar as well as non - polar components finds applications in industrial and technological processes. Most of the work on binary mixtures is channelized towards the estimation of thermodynamic parameters like adiabatic compressibility, free length, etc., and their excess values so as to relate them towards explaining the molecular interactions taking place between the components of the binary mixtures. Further, such studies as a function of concentration are useful in gaining insight into the structure and bonding of associated molecular complexes and other molecular processes [5-10]. Ultrasonic velocity measurements have been successfully employed to detect and assess weak and strong molecular interactions, present in binary and ternary liquid mixtures. In this paper, an attempt to investigate the ultrasonic studies of n-alcohol like 1-pentanol, 1-hexanol, 1-heptanol in benzene, carbon tetrachloride, toluene and dioxane binary liquid mixture systems at 301 K are made. We have measured density ( $\rho$ ), ultrasonic velocity (u), viscosity ( $\eta$ ) of mixtures of n-alcohol + benzene + carbon tetra chloride + toluene + dioxane with different mole fractions at a temperature of 301 K. From this data, acoustical parameters like adiabatic compressibility ( $\beta$ ), Inter molecular free length ( $L_f$ ), free volume (Vf), and their excess values are computed. Results are used to explain the nature of molecular interactions between mixing compounds. Further, the experimentally measured ultrasonic velocities at various molar concentrations have been compared with

the theoretically estimated velocities based on empirical, semi empirical and statistical models for the binary systems.

## EXPERIMENTAL SECTION

All the chemicals used in present work were analytical reagent (AR) grade (99.9% pure) and were supplied by SD fine chemicals Ltd India. The liquids were thoroughly distilled to remove dissolved impurities using standard chemical procedures. The purity of the samples was checked by the density measurements and the results were compared with the literature values. Ultrasonic velocities were measured with ultrasonic interferometer (model F80) supplied by Mittal enterprises, New Delhi, operating at a frequency of 2 MHz. It has an accuracy of  $\pm 0.1\%$ . Viscosities of pure compounds and their mixtures were determined using Ostwald's viscometer with an accuracy of  $\pm 0.002\%$ , calibrated with double distilled water. The densities of pure compounds and their solutions were measured accurately using 10 ml specific gravity bottles in Dhona electric balance precisely and the accuracy in weighing is  $\pm 0.1$  mg. Acoustic parameters such as adiabatic compressibility ( $\beta$ ), acoustic impedance (Z), free length ( $L_f$ ), and free volume ( $V_f$ ) were determined using the following relations.

$$\text{Adiabatic compressibility } \beta = \frac{1}{u^2 \rho} \quad \dots(1)$$

$$\text{Intermolecular free length } L_f = K_T \beta \quad \dots(2)$$

$$\text{Free volume } V_f = \left[ \frac{M_{\text{eff}} u}{\eta K} \right] \quad \dots(3)$$

$$\text{Acoustic impedance } Z = u \rho \quad \dots(4)$$

Where KT is the temperature dependent constant having a value of  $199.53 \times 10^{-8}$  in MKS system, K is temperature independent constant whose value is  $4.28 \times 10^9$  in MKS system, b is a factor depending on packing pattern which is 1.78, R is the gas constant and T is temperature in K, U is ultrasonic velocity in m/s,  $\rho$  is the density in Kg/m<sup>3</sup> and Meff is the effective molecular weight, given by Meff =  $\sum x_i m_i$  where x is the mole fraction and m is the molecular weight of the ith component.

## RESULTS AND DISCUSSION

The binary mixture systems taken up for the present study are: 1- Pentanol + toluene, benzene, carbon tetrachloride, dioxane and 1-hexanol+ toluene, benzene, carbontetra chloride, dioxane and 1-heptanol + toluene, benzene, carbon tetrachloride, dioxane). The experimentally determined values of velocity, density and along with the calculated values of free volume ( $V_f$ ), adiabatic compressibility ( $\beta$ ), free length ( $L_f$ ), acoustic impedance (Z) for all the systems at 301 K are reported in Table I & II

**Table 1.** Experimental values of ultrasonic velocity and density of pure liquids (at 301 K) used as a component of binary mixtures reported in this work

Liquids	Ultrasonic velocity(m/s)	Density(Kg/m <sup>3</sup> )
1-Pentanol	1256	814
1-Hexanol	1288	820
1-Heptanol	1207	818
Benzene	1294	874
Toluene	1282	855
Carbontetra chloride	893	1512
Dioxane	1305	1011

In all the three binary systems, the ultrasonic velocity increases with increasing concentration of alcohols. The variation of ultrasonic velocity in a solution depends upon the increase or decrease of intermolecular free length after mixing the components.

**Table 2.** Values of ultrasonic velocity ( $u$  m/s), density ( $\rho$  kg/m<sup>3</sup>) adiabatic compressibility ( $\beta$  m<sup>2</sup> N<sup>-1</sup>) intermolecular free length ( $L_f$  m), acoustic impedance ( $Z$  kgm<sup>-2</sup>s<sup>-1</sup>) and free volume ( $V_f$  m<sup>3</sup>mol<sup>-1</sup>) of binary systems as a function of molar concentration of components "B" at 301 K

X2	u	$\rho$	$\beta \times 10^{10}$	$L_f \times 10^{11}$	$V_f$	$Z \times 10^6$
<b>Pentanol + Toluene</b>						
0	1256	814	7.7874	5.7695	2.17	1022.3
0.1	1262	823	7.6292	5.7106	2.2003	1038.6
0.2	1266	828	7.5353	5.6754	2.1841	1048.2
0.3	1267	834	7.4693	5.6504	2.1623	1056.6
0.4	1270	836	7.4162	5.6303	2.1373	1061.7
0.5	1271	840	7.3693	5.6125	2.1202	1067.6
0.6	1274	842	7.3172	5.5926	2.1051	1072.7
0.7	1278	848	7.22	5.5554	2.0901	1083.7
0.8	1279	848	7.2088	5.551	2.0818	1084.5
0.9	1282	853	7.133	5.5218	2.064	1093.5
1	1282	855	7.1163	5.5153	2.042	1096.1
<b>Pentanol + Dioxane</b>						
0	1403	910	5.5826	4.885	2.3725	1276.7
0.1	1262	840	7.4748	5.6525	2.1257	1060
0.2	1268	862	7.2153	5.5535	2.1363	1093
0.3	1274	885	6.9617	5.4551	2.1472	1127.4
0.4	1281	902	6.756	5.3739	2.1594	1155.4
0.5	1289	930	6.4715	5.2595	2.1734	1198.7
0.6	1287	950	6.355	5.212	2.1705	1222.6
0.7	1300	967	6.119	5.1143	2.1929	1257.1
0.8	1292	967	6.1951	5.146	2.1795	1249.3
0.9	1299	989	5.9921	5.061	2.1921	1284.7
1	1305	1011	5.808	4.9826	2.2027	1319.3
<b>Hexanol + Toluene</b>						
0	1288	820.1	7.3502	5.6052	1.1749	1056.2
0.1	1291	827	7.2552	5.5689	7.1908	1067.3
0.2	1299	835	7.1016	5.5096	5.4215	1084.4
0.3	1302	841	7.0129	5.4751	4.3135	1095.3
0.4	1305	848	6.9258	5.441	3.5965	1106.3
0.5	1301	850	6.951	5.4509	2.9787	1105.8
0.6	1299	852	6.9587	5.4539	2.6976	1106.4
0.7	1297	855	6.9607	5.4547	2.4228	1107.9
0.8	1292	852	7.0282	5.4811	2.1692	1100.9
0.9	1289	854	7.0477	5.4887	1.9818	1100.7
1	1284	855	7.0917	5.5058	1.8247	1098
<b>Hexanol + Benzene</b>						
0	1288	820	7.3502	5.6052	1.8026	1056.2
0.1	1291	828	7.2456	5.5652	1.1765	1068.8
0.2	1293	834	7.1711	5.5365	9.166	1078.4
0.3	1298	842	7.0521	5.4904	7.5172	1092.7
0.4	1303	851	6.9144	5.4365	6.4382	1109.5
0.5	1303	857	6.8688	5.4186	5.4715	1117.1
0.6	1302	861	6.847	5.4099	5.0561	1121.3
0.7	1302	866	6.8103	5.3954	4.6324	1127.4
0.8	1301	865	6.83	5.4032	4.2303	1125.3
0.9	1302	871	6.7807	5.3837	3.9406	1133
1	1301	875	6.757	5.3743	3.6962	1137.8
<b>Heptanol + Toluene</b>						
0	1207	818	8.3893	5.9883	2.202	987.5
0.1	1231	835	7.9018	5.8117	1.583	1027.8
0.2	1245	844	7.652	5.7191	1.2867	1050
0.3	1259	853	7.3991	5.6238	1.0855	1073.6
0.4	1263	855	7.3331	5.5987	9.4022	1079.8
0.5	1268	857	7.2562	5.5693	8.1079	1086.9
0.6	1262	853	7.3621	5.6097	7.4871	1076.1
0.7	1263	852	7.3545	5.6069	6.8523	1076.2
0.8	1261	850	7.3942	5.622	6.3038	1072.3
0.9	1267	852	7.3056	5.5882	5.9064	1080.2
1	1272	853	7.2488	5.5664	5.5679	1084.9

Heptanol + Carbon tetra chloride						
0	1207	818	8.3913	5.9891	2.223	987.3
0.1	1183	905	7.892	5.8081	1.7224	1070.8
0.2	1160	982	7.5657	5.6868	1.4111	1139.2
0.3	1133	1064	7.3214	5.5942	1.1662	1205.5
0.4	1107	1148	7.1082	5.5122	9.8062	1270.8
0.5	1064	1244	7.1006	5.5092	8.0306	1323.6
0.6	1033	1303	7.192	5.5446	7.0226	1345.9
0.7	994	1374	7.3661	5.6113	6.0052	1365.7
0.8	984	1367	7.5551	5.6828	5.3041	1345.1
0.9	941	1443	7.8262	5.7839	4.5612	1357.8

Based on the model for sound propagation proposed by Eyring and Kincaid[11], ultrasonic velocity should increase, if the inter molecular free length decreases and vice versa. This fact was noticed in the present study for all the three binary liquid systems. Same trend was noticed by earlier workers in their liquid mixtures [12-18].

In fact, the molecular association increases ultrasonic velocity ( $u$ ) and acoustic impedance ( $Z$ ), decreases intermolecular free length ( $L_f$ ) and adiabatic compressibility ( $\beta$ ). A reduction in adiabatic compressibility ( $K_s$ ) is an indication that component molecules are held close to each other. The decrease in the values of adiabatic compressibility ( $\beta$ ) and inter molecular free length ( $L_f$ ) with increase in ultrasonic velocity ( $u$ ) further strengthens the strong molecular association between the unlike molecules through hydrogen bonding. Carbon tetra chloride is a non polar liquid and the parameters are same as for benzene. This fact is reflected clearly from the values of ultrasonic velocity ( $u$ ), intermolecular free length ( $L_f$ ), adiabatic compressibility ( $\beta$ ) and acoustic impedance ( $Z$ ) for system.

Alcohols usually exist in polymeric form. In the presence of polar molecules, they dissociate into monomers. With the increase of alcohol, the tendency for intermolecular hydrogen bonding increases. The association is stronger and we expect a larger variation in the parameters. Eventually ultrasonic velocities ( $u$ ) of both the components are nearly equal. In this fact is reflected in increase of ultrasonic velocity ( $u$ ) and hence decrease in inter molecular free length ( $L_f$ ) also it can be observed that adiabatic compressibility values decrease with increasing concentrations of alcohol. Similar results were obtained by earlier workers in their liquid mixtures. [19-21].

Acoustic impedance ( $Z$ ) of a material is the opposition exerted by the medium to displacement of the medium's particles by the sound energy. It is important to measure acoustic impedance because studies have shown that in solvent mixtures when molecular interactions occurs, acoustic impedance exhibits a non-linear variation with increasing mole fraction of solute. This was used as an essential tool to predict molecular level interactions in binary and ternary liquid mixtures. In systems of binary liquid mixtures is always much greater than either of the polar solutes in the inert solvent. Further, the increase in free volume ( $V_f$ ) with rise in concentration of alcohols in all the systems under study, clearly indicate the increasing magnitude of interactions.

**Table 3.** Theoretical values of ultrasonic velocity calculated from Nomoto (NOMO), Impedence, Ideal mixtures relation, Rao's relation and Junjie's relations along with experimental ultrasonic velocity and percentage error for n-alcohols + toluene,benzene,carbontetra chloride,dioxane of binary systems at 301 K

X2	Expt	NOM	IDEAL	JUN	IMP	Rao's	NOMO	IDEAL	JUN	IMP	Rao's
Pentanol + Toluene											
0	1256	1256	1256	1256	1256	1256	0	0	0	0	0
0.1	1262	1258.6	1258.3	1258.3	1259	1259	0.2716	0.2917	0.2954	0.2601	0.2707
0.2	1266	1261.2	1260.7	1260.6	1261	1261	0.383	0.4188	0.4255	0.3628	0.3814
0.3	1267	1263.7	1263.1	1263	1264	1264	0.2576	0.3049	0.3137	0.2312	0.2555
0.4	1270	1266.3	1265.6	1265.5	1267	1266	0.2892	0.3435	0.3536	0.2592	0.2868
0.5	1271	1268.9	1268.2	1268.1	1269	1269	0.1633	0.2201	0.2308	0.1322	0.1608
0.6	1274	1271.5	1270.8	1270.7	1272	1272	0.1941	0.2488	0.2592	0.1644	0.1917
0.7	1278	1274.1	1273.5	1273.4	1275	1274	0.3023	0.3504	0.3595	0.2766	0.3003
0.8	1279	1276.8	1276.3	1276.2	1277	1277	0.1758	0.2126	0.2196	0.1563	0.1742
0.9	1282	1279.4	1279.1	1279.1	1280	1279	0.205	0.2257	0.2297	0.194	0.2041
1	1282	1282	1282	1282	1282	1282	0	0	0	0	0
Pentanol + Dioxane											
0	1403	1403	1403	1403	1403	1403	0	0	0	0	0
0.1	1262	1393.9	1392.2	1393.4	1392	1393	-10.451	-10.317	-10.4133	-10.32	-10.379

0.2	1268	1384.6	1381.6	1383.8	1382	1383	-9.1984	-8.9623	-9.1314	-8.966	-9.071
0.3	1274	1375.2	1371.3	1374.1	1371	1373	-7.9462	-7.6391	-7.8578	-7.644	-7.778
0.4	1281	1365.7	1361.2	1364.4	1361	1363	-6.6106	-6.2629	-6.5093	-6.268	-6.419
0.5	1289	1356	1351.4	1354.6	1351	1353	-5.1959	-4.8373	-5.0901	-4.842	-4.996
0.6	1287	1346.1	1341.7	1344.8	1342	1344	-4.5929	-4.2494	-4.4902	-4.254	-4.400
0.7	1300	1336.1	1332.2	1334.9	1332	1334	-2.7758	-2.4791	-2.686	-2.483	-2.608
0.8	1292	1325.9	1323	1325	1323	1324	-2.6233	-2.3967	-2.5539	-2.399	-2.494
0.9	1299	1315.5	1313.9	1315	1314	1315	-1.2728	-1.1463	-1.2336	-1.147	-1.2
1	1305	1305	1305	1305	1305	1305	0	0	0	0	0
<b>Hexanol + Toluene</b>											
0	1288	1288	1288	1288	1288	1288	0	0	0	0	0
0.1	1291	1287.6	1287	1287.5	1288	1288	0.2882	0.3317	0.2953	0.2876	0.2864
0.2	1299	1287.2	1286.2	1287	1287	1287	0.8741	0.9508	0.8864	0.873	0.871
0.3	1302	1286.7	1285.4	1286.5	1287	1287	1.157	1.2574	1.1729	1.1556	1.1529
0.4	1305	1286.3	1284.8	1286.1	1286	1286	1.4365	1.5508	1.4545	1.4349	1.4319
0.5	1301	1285.9	1284.4	1285.7	1286	1286	1.1492	1.2686	1.1677	1.1476	1.1445
0.6	1299	1285.5	1284.1	1285.3	1286	1286	1.016	1.1308	1.0336	1.0145	1.0115
0.7	1297	1285.1	1283.8	1285	1285	1285	0.8835	0.984	0.8987	0.8821	0.8795
0.8	1292	1284.8	1283.8	1284.6	1285	1285	0.586	0.6628	0.5975	0.585	0.583
0.9	1289	1284.4	1283.8	1284.3	1284	1284	0.3569	0.4003	0.3633	0.3564	0.3553
1	1284	1284	1284	1284	1284	1284	0.0167	0.0167	0.0167	0.0167	0.0167
<b>Hexanol + Benzene</b>											
0	1288	1288	1288	1288	1288	1288	0	0	0	0	0
0.1	1291	1289.6	1284.8	1289.2	1289	1289	0.1282	0.4951	0.1583	0.1418	0.1481
0.2	1293	1291	1282.6	1290.4	1291	1291	0.1583	0.8082	0.2096	0.1818	0.193
0.3	1298	1292.5	1281.4	1291.6	1292	1292	0.3972	1.2464	0.4617	0.4271	0.4417
0.4	1303	1293.8	1281.2	1292.9	1293	1293	0.741	1.7072	0.8115	0.7741	0.7905
0.5	1303	1295.1	1282	1294.2	1295	1295	0.6131	1.6211	0.6839	0.6467	0.6637
0.6	1302	1296.4	1283.8	1295.6	1296	1296	0.4664	1.4369	0.5319	0.4978	0.5141
0.7	1302	1297.6	1286.5	1296.9	1297	1297	0.3664	1.2187	0.4217	0.3932	0.4073
0.8	1301	1298.8	1290.3	1298.3	1299	1298	0.171	0.8242	0.2117	0.191	0.2017
0.9	1302	1299.9	1295.1	1299.6	1300	1300	0.1278	0.4972	0.1499	0.1388	0.1447
1	1301	1301	1301	1301	1301	1301	-0.0282	-0.0282	-0.0282	-0.028	-0.028
<b>Heptanol + Toluene</b>											
0	1207	1207	1207	1207	1207	1207	0	0	0	0	0
0.1	1231	1214.6	1209	1213.8	1214	1213	1.3567	1.8134	1.4194	1.4243	1.4528
0.2	1245	1221.9	1211.8	1220.5	1220	1220	1.8257	2.6332	1.9332	1.9419	1.9919
0.3	1259	1228.9	1215.7	1227.2	1227	1226	2.3735	3.4282	2.5095	2.5211	2.5858
0.4	1263	1235.7	1220.4	1233.8	1234	1233	2.15	3.3606	2.3011	2.3144	2.388
0.5	1268	1242.3	1226.2	1240.4	1240	1239	2.0183	3.2854	2.1712	2.1853	2.2614
0.6	1262	1248.6	1233.1	1246.8	1247	1246	1.078	2.312	1.2219	1.2357	1.3088
0.7	1263	1254.8	1241	1253.2	1253	1252	0.6803	1.7712	0.8031	0.8153	0.8791
0.8	1261	1260.7	1250.1	1259.5	1259	1259	0.0348	0.8779	0.1263	0.1357	0.1843
0.9	1267	1266.4	1260.4	1265.8	1266	1265	0.0556	0.5341	0.1056	0.111	0.1381
1	1272	1272	1272	1272	1272	1272	-0.0368	-0.0368	-0.0368	-0.036	-0.036
<b>Heptanol + Carbontetra chloride</b>											
0	1207	1207	1207	1207	1207	1207	0	0	0	0	0
0.1	1183	1192	1144.2	1185.6	1154	1173	-0.7417	3.303	-0.2005	2.5037	0.8803
0.2	1160	1175.2	1093.3	1162.8	1108	1139	-1.2991	5.7646	-0.2248	4.4932	1.7939
0.3	1133	1156.2	1051.3	1138.2	1069	1107	-2.048	7.2095	-0.4598	5.6843	2.3398
0.4	1107	1134.5	1016.3	1111.8	1034	1074	-2.4837	8.1955	-0.4292	6.5795	2.9566
0.5	1064	1109.5	986.8	1083.1	1004	1043	-4.2761	7.2602	-1.7989	5.6543	2.0035
0.6	1033	1080.4	961.7	1052.1	976.9	1012	-4.5898	6.8999	-1.8458	5.4283	2.0599
0.7	994	1046.2	940.4	1018.2	952.9	981.4	-5.2471	5.3882	-2.4302	4.1367	1.2699
0.8	984	1005.2	922.3	980.9	931.3	951.6	-2.1584	6.267	0.3112	5.3589	3.2879
0.9	941	955.5	907	939.8	911.7	922.5	-1.5459	3.6169	0.1261	3.1098	1.9635
1	894	894	894	894	894	894	0	0	0	0	0

### CONCLUSION

An analysis of these values suggests the presence of strong intermolecular interaction in all the binary mixtures which may be due to hydrogen bond, dipole-dipole, hyperconjugation and charge transfer. It is found that the intermolecular interaction is very strong in alcohol + non-polar solvents and this strong interaction may be attributed to the fact that the interaction due to negative inductive effect dominates over the resonance effect in the mixture. Theoretical ultrasonic velocities of the binary mixtures were estimated using various empirical, semi empirical,

models given by eqns and are presented with experimental values in Table 3. The percentage of deviation of the theoretical ultrasonic velocity values from experimental values are also shown in Table 3. On the whole, all the theoretical models fairly predicted ultrasonic velocities, are reasonably close to the experimental values for these binary mixtures reported in this work, thus showing the validity of these theoretical models for binary mixtures.

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