Study of Molecular Interactions in Binary Mixtures of Diethyl Carbonate + Benzene Derivatives at Different Temperatures

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Abstract: Investigation on the molecular interactions between binary mixtures containing diethyl carbonate in combination with nitrobenzene, chlorobenzene and aniline is presented. Ultrasonic velocity and density values were determined for the individual components as well as binary mixtures of the above benzene derivatives with diethyl carbonate at temperatures (293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15) K over the entire composition range. Further, adiabatic compressibility and acoustic impedance values were calculated using the experimental results. In addition to these parameters, the excess parameters like excess adiabatic compressibility, excess acoustic impedance and deviation in ultrasonic velocity were also obtained. Based on all these results, molecular interactions among the selected components were discussed.

Keywords: Ultrasonic velocity, density, molecular interactions, binary mixtures, diethyl carbonate.

INTRODUCTION

Valuable information about molecular structure, packing of the constituent molecules and interactions among them at different conditions of temperatures and concentrations can be obtained by studying changes in ultrasonic velocity, density and other thermo-acoustical parameters. Moreover, accurate experimental data on such parameters of organic compounds and their binary combinations are necessary in many chemical industries and engineering disciplines.

Compare to the other techniques like dielectric relaxation [1], infrared spectroscopy, nuclear magnetic resonance, etc. acoustical or ultrasonic methods are used as complimentary because of sensitive to very low population densities at high energy states, and have the added advantage of being less cost with efficiency.

The study of changes in thermo-acoustical properties of mixtures of organic compounds and the degree of deviation from ideality is an excellent qualitative and quantitative method to derive useful information about structural alignments and intermolecular forces in liquid mixtures. This fact has motivated both the theoretical and the experimental investigations of excess thermodynamic properties of liquid mixtures [2, 3]. Further, dynamic behavior of liquids can be studied by using accurate measurement of energy changes due to scattering [4].

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In the recent past, study of thermodynamic and thermo-acoustic properties of binary systems containing esters of carbonic acid received much attention. In view of the increased significance of esters of carbonic acid, the authors of the present study reported earlier the investigations on binary mixtures containing diethyl carbonate [5]. Interesting results obtained in this study led the authors continue their investigations on diethyl carbonate with simpler benzene derivatives like nitrobenzene, chlorobenzene and aniline. These compounds have been selected in view of their industrial importance summarized below.

Many chemical industries like pharmaceutical industry, agrochemical industry, hydrogen refinery, etc., use diethyl carbonate (DEC) as an extraction solvent and reaction medium. In dyeing industry, it can make the dyeing process uniform and resists fading against sunshine. It is also famous for its action as paint remover in paint industry. DEC also acts as a lubricant of the new refrigerant, hydrofluorocarbon. Aniline is used as precursor in chemical production industries like rubber processing chemicals, drugs, dyes, pigments, herbicides and explosives. About 95% of nitrobenzene produced industrially is used for manufacturing aniline. Also, it is used fin leather dressings, floor and shoe polishes and as a solvent in paint industry.

It is useful in masking unpleasant odors. It finds application as an inexpensive perfume for soaps and in Kerr cells. The major use of chlorobenzene is in rubber dyestuffs and herbicides.

Identifying the industrial importance of diethyl carbonate, some researchers [6, 7] studied molecular

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interactions in binary liquid mixtures containing diethyl carbonate. Based on this background, the binary systems namely, "diethyl carbonate + nitrobenzene", "diethyl carbonate + chlorobenzene" and "diethyl carbonate + aniline" are chosen for the present study. For each of these binary mixtures, the parameters namely, speed of sound, density, excess acoustic impedance (Z^E), excess adiabatic compressibility (β^E) and deviation in speed of sound (ΔU) were determined at (293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15) K over the entire range of composition. The excess/deviation values were correlated with the Redlich-Kister equation. All the results are interpreted in terms of intermolecular interactions.

MATERIALS AND METHODS

All the chemicals used in this study were > 99% pure supplied by Sigma Aldrich Chemical Company. The purity of these chemicals was checked by measuring the density and speeds of sound and comparing these values with those reported in literature. The chemicals were used as such in the present study without carrying out any purification process.

Density and speed of sound of pure liquid components and of liquid mixtures were measured by Anton Paar DSA 5000 density and sound analyzer provided with two Pt 100 platinum thermometer. In view of extreme sensitivity of density towards temperature, a built-in solid state thermostat was equipped with the apparatus, with a readability of 0.001 K. The accuracies in the density and speed of sound reported are 5 x10^{-3} Kgm^{-3} and 0.5 ms^{-1} respectively.

Binary liquid mixtures were prepared by mixing known masses of pure liquids and were stored in airtight-stoppered bottles to minimize losses due to evaporation. For all weighing, an electronic balance with a precision of ± 0.01 mg was used.

RESULTS AND DISCUSSION

The experimental ultrasonic velocity and density values obtained from the measurements of the pure compounds are summarized in Table **1** along with the corresponding literature values [8-12]. A good agreement is observed between experimental and literature values. The experimental ultrasonic velocity and density values for the binary mixture at different temperatures and at atmospheric pressure are summarized in Table **2**. Adiabatic compressibility and acoustic impedance were calculated from our measurements by using the following equations.

Adiabatic compressibility,

$$\beta = 1/\rho U^2 \quad (1)$$

Acoustic impedance,

$$Z = U.\ \rho \quad (2)$$

where, u and ρ are the ultrasonic velocity and density respectively.

The excess properties such as β^E and Z^E have been calculated using the equation

$$Y^E = Y_{mix} - [x_1Y_1 + x_2Y_2] \quad (3)$$

where Y^E is β^E or Z^E, and x represents mole fraction of the component and subscripts 1 and 2 stand for the components 1 and 2.

The excess values for each mixture have been fitted to Redlich-Kister [13] polynomial equation

$$Y^E = X_1.X_2 \sum_{i=0}^{n} A_i \left(X_1 - X_2\right)^i \quad (4)$$

The values of the coefficients A_i were calculated by method of least squares along with the standard deviation σ (Y^E). The coefficient is adjustable parameters for a better fit of the excess functions. The standard deviation values were obtained from

Table 1: Comparison of Experimental Values of Ultrasonic Velocity and Density with Literature Values at 303.15 K

Compound	Ultrasonic velocity, u (m/s)		Density (kg/m^3)	
	Experiment	Literature	Experiment	Literature
Diethyl carbonate	1156.96	--	963.468	963.00[8]
Nitrobenzene	1438.30	1432.8[10]	1192.297	1193.4[9]
Chlorobenzene	1248.92	1252.0[11]	1095.506	1095.0[9]
Aniline	1617.52	1619.3[12]	1012.746	1013.23[12]

Table 2: Experimental Values of Ultrasonic Velocity (U) and Density (ρ) for Diethyl Carbonate + Nitrobenzene, + Chlorobenzene, + Aniline at Different Temperatures

Mole fraction, x_1	Diethyl carbonate + nitrobenzene						
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
	U (m/s)						
0.0000	1474.07	1456.26	1438.30	1420.44	1402.70	1385.13	1367.68
0.0937	1447.27	1428.98	1410.67	1392.44	1374.35	1356.15	1338.01
0.1678	1426.18	1407.66	1389.12	1370.61	1352.09	1333.85	1315.56
0.2715	1396.76	1377.89	1359.07	1340.27	1321.57	1303.04	1284.56
0.3577	1372.21	1353.23	1334.20	1315.29	1296.42	1277.63	1259.01
0.4547	1345.12	1325.91	1306.65	1287.52	1268.35	1249.33	1230.48
0.6149	1300.95	1281.27	1261.57	1241.98	1222.43	1203.00	1183.83
0.6606	1288.49	1268.67	1248.79	1229.07	1209.39	1189.83	1170.60
0.7722	1258.34	1238.14	1217.90	1197.90	1177.88	1158.03	1138.44
0.8907	1227.05	1206.52	1186.02	1165.55	1145.16	1125.06	1105.09
1.0000	1198.01	1177.46	1156.96	1136.62	1116.45	1096.47	1076.66
	ρ (Kg/m^3)						
0.0000	1202.238	1197.263	1192.297	1187.328	1182.355	1177.385	1172.408
0.0937	1179.555	1174.626	1169.588	1164.553	1159.516	1154.472	1149.425
0.1678	1161.679	1156.805	1151.720	1146.631	1141.539	1136.440	1131.336
0.2715	1137.613	1132.465	1127.312	1122.153	1116.988	1111.812	1106.631
0.3577	1117.218	1112.014	1106.804	1101.588	1096.362	1091.128	1085.885
0.4547	1094.887	1089.633	1084.371	1079.102	1073.821	1068.530	1063.228
0.6149	1059.198	1053.894	1048.576	1043.247	1037.908	1032.556	1027.192
0.6606	1049.223	1043.833	1038.529	1033.211	1027.777	1022.327	1016.861
0.7722	1024.734	1019.276	1013.802	1008.310	1002.798	997.262	991.710
0.8907	998.715	993.188	987.638	982.066	976.470	970.851	965.209
1.0000	974.699	969.097	963.468	957.814	952.134	946.428	940.691
	Diethyl carbonate + chlorobenzene						
	U (m/s)						
0.0000	1285.71	1267.44	1248.92	1230.49	1212.19	1194.03	1175.99
0.0840	1278.03	1259.44	1240.65	1221.95	1203.40	1184.95	1166.60
0.1774	1269.70	1250.83	1231.85	1212.88	1194.10	1175.46	1156.89
0.2682	1261.68	1242.55	1223.40	1204.26	1185.27	1166.40	1147.68
0.3591	1253.71	1234.36	1215.05	1195.69	1176.51	1157.46	1138.56
0.4617	1244.74	1225.17	1205.63	1186.10	1166.71	1147.48	1128.40
0.5846	1233.99	1214.19	1194.41	1174.65	1155.04	1135.60	1116.30
0.6290	1230.10	1210.22	1190.34	1170.50	1150.82	1131.30	1111.95
0.7602	1218.65	1198.49	1178.36	1158.30	1138.42	1118.72	1099.17
0.8694	1209.19	1188.83	1168.49	1148.30	1128.29	1108.46	1088.75
1.0000	1198.01	1177.46	1156.96	1136.62	1116.45	1096.47	1076.66

(Table 2). Continued.

	ρ (Kg/m^3)						
0.0000	1106.296	1100.906	1095.506	1090.093	1084.665	1079.226	1073.772
0.0840	1093.551	1088.482	1083.240	1077.982	1072.811	1067.893	1062.919
0.1774	1079.990	1075.030	1069.796	1064.566	1059.358	1054.330	1049.487
0.2682	1067.467	1062.470	1057.263	1051.938	1046.598	1041.639	1036.561
0.3591	1055.470	1050.257	1044.924	1039.576	1034.307	1029.219	1023.897
0.4617	1042.065	1036.828	1031.473	1026.097	1020.901	1015.582	1010.239
0.5846	1026.122	1020.863	1015.482	1010.081	1004.858	999.411	994.039
0.6290	1020.399	1015.145	1009.759	1004.351	999.120	993.662	988.282
0.7602	1003.870	998.694	993.289	987.861	982.408	977.031	971.629
0.8694	990.570	985.281	979.863	974.423	968.857	963.503	958.042
1.0000	974.699	969.097	963.468	957.814	952.134	946.428	940.691
	Diethyl carbonate + aniline						
	U (m/s)						
0.00000	1656.02	1636.79	1617.52	1598.27	1579.1	1559.95	1540.88
0.07302	1613.70	1594.43	1575.11	1555.80	1536.56	1517.56	1498.22
0.16517	1562.65	1543.34	1523.92	1504.55	1485.25	1466.00	1446.83
0.25373	1516.89	1497.74	1478.31	1458.49	1439.08	1419.76	1400.52
0.33866	1475.49	1455.94	1436.36	1416.83	1397.37	1377.98	1358.67
0.44318	1427.55	1407.88	1388.18	1368.52	1348.95	1329.49	1310.13
0.54682	1381.13	1361.32	1341.47	1321.69	1302.02	1282.46	1263.03
0.65461	1335.93	1315.96	1295.96	1276.04	1256.26	1236.61	1217.09
0.75589	1294.35	1274.23	1254.09	1234.06	1214.18	1194.46	1174.87
0.88404	1242.96	1222.58	1202.27	1182.09	1162.07	1142.22	1122.52
1.00000	1198.01	1177.46	1156.96	1136.62	1116.45	1096.47	1076.66
	ρ (Kg/m^3)						
0.00000	1021.401	1017.076	1012.746	1008.41	1004.069	999.721	995.367
0.07302	1018.579	1014.181	1009.775	1005.361	1000.941	996.514	992.080
0.16517	1014.687	1010.192	1005.686	1001.172	996.647	992.115	987.571
0.25373	1010.954	1006.359	1001.755	997.133	992.503	987.864	983.212
0.33866	1007.334	1002.641	997.936	993.220	988.490	983.747	978.991
0.44318	1002.841	998.024	993.191	988.347	983.487	978.613	973.723
0.54682	997.994	993.047	988.083	983.104	978.108	973.097	968.065
0.65461	992.838	987.724	982.646	977.518	972.372	967.207	962.023
0.75589	987.764	982.535	977.289	972.022	966.732	961.426	956.099
0.88404	981.296	975.875	970.438	964.977	959.494	953.985	948.452
1.00000	974.699	969.097	963.468	957.814	952.134	946.428	940.691

$$\sigma(Y^E) = \left[\frac{\sum_{i=1}^{n} (Y^E_{\text{expt}} - Y^E_{cal})^2}{m-n} \right]^{1/2} \quad (5)$$

where, m is the number of experimental points, n is the number of parameters, Y_{expt} and Y_{cal} are the experimental and calculated parameters, respectively. The values of coefficients and standard deviations at different temperatures for the above measured parameters are given in Table **3**.

The excess compressibility is reported to be a quantity which is proportional to the strength of

Table 3: Value of Coefficients, A_i, of the Redlich-Kister Polynomial and Standard Deviations, σ, for Binary Mixtures at T = (293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15) K

Parameter	T/K	A_0	A_1	A_2	A_3	σ
{Diethyl carbonate + Nitrobenzene}						
β^E (10^{-10} $N^{-1}m^2$)	293.15	-1.18628	0.16405	-0.08016	0.04619	0.00093
	298.15	-1.27574	0.18336	-0.05975	0.02052	0.00081
	303.15	-1.37292	0.20048	-0.03573	-0.00387	0.00099
	308.15	-1.47884	0.22737	0.00073	-0.05093	0.00078
	313.15	-1.58812	0.25061	0.04599	-0.10517	0.00097
	318.15	-1.70360	0.26693	0.08773	-0.12727	0.00136
	323.15	-1.83335	0.30288	0.14869	-0.17683	0.00198
Z^E (10^6 Kg $m^{-2}s^{-1}$)	293.15	-0.09750	-0.00717	0.00956	-0.00254	0.00025
	298.15	-0.09869	-0.00801	0.00759	0.00141	0.00019
	303.15	-0.09957	-0.00835	0.00452	0.00367	0.00020
	308.15	-0.10036	-0.00948	0.00035	0.00723	0.00018
	313.15	-0.10180	-0.01008	-0.00376	0.01053	0.00019
	318.15	-0.10345	-0.00965	-0.00776	0.00955	0.00022
	323.15	-0.10465	-0.01073	-0.01342	0.00978	0.00027
ΔU (m/s)	293.15	-14.1143	2.71636	4.10067	-2.84001	0.07941
	298.15	-14.7216	2.42078	7.57047	-1.89416	0.06888
	303.15	-15.3294	2.79696	-1.73622	-0.75324	0.07715
	308.15	-15.8834	2.40258	-5.19703	1.82932	0.03833
	313.15	-16.9577	2.16674	-8.97245	4.88118	0.02974
	318.15	-18.2566	2.84629	-12.36683	4.12045	0.05685
	323.15	-19.1492	2.12971	-17.45136	4.63627	0.11222
	{Diethyl carbonate + Chlorobenzene}					
β^E (10^{-10} $N^{-1}m^2$)	293.15	-0.20304	0.03002	0.04780	0.03341	0.00040
	298.15	-0.22450	0.02856	0.02542	0.03119	0.00048
	303.15	-0.24164	0.03216	0.01546	0.03779	0.00060
	308.15	-0.26354	0.04280	0.01904	0.03236	0.00049
	313.15	-0.29001	0.05507	0.02193	0.00107	0.00098
	318.15	-0.31416	0.04534	-0.01228	0.02395	0.00111
	323.15	-0.33819	0.06362	-0.05605	-0.04050	0.00132
Z^E (10^6 Kg $m^{-2}s^{-1}$)	293.15	-0.03078	-0.00570	-0.00983	-0.00848	0.00007
	298.15	-0.03007	-0.00426	-0.00410	-0.00689	0.00009
	303.15	-0.03094	-0.00504	-0.00118	-0.00630	0.00013
	308.15	-0.03066	-0.00626	-0.00137	-0.00489	0.00007
	313.15	-0.03003	-0.00787	-0.00131	0.00160	0.00016
	318.15	-0.03007	-0.00581	0.00457	0.00032	0.00018
	323.15	-0.03058	-0.00818	0.01242	0.01269	0.00023

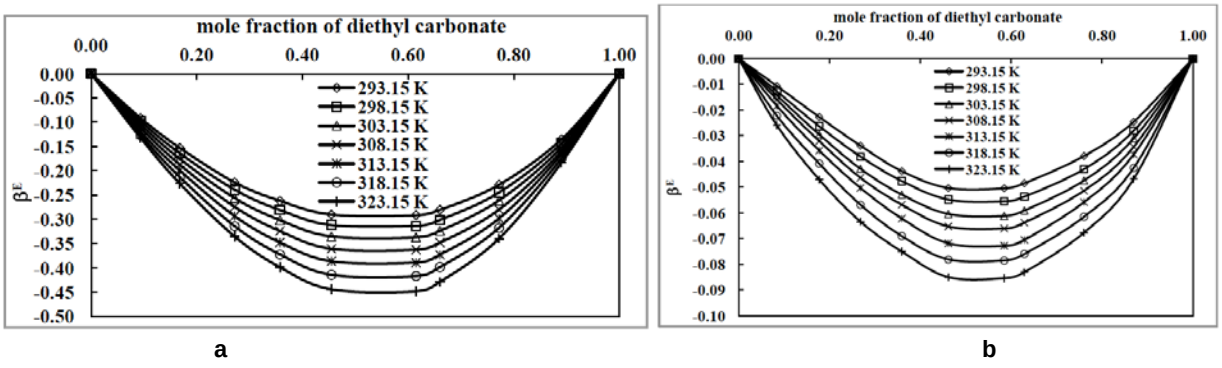
(Table 3). Continued.

	293.15	-1.89063	-0.38619	-2.01288	-0.69555	0.00498
	298.15	-2.80113	-1.01793	-2.58955	-0.16066	0.01254
	303.15	-3.17176	-0.75689	-3.46952	-0.84247	0.01994
ΔU (m/s)	308.15	-4.07101	-0.88498	-4.00149	-1.22564	0.01536
	313.15	-4.96731	-0.89436	-4.05773	-1.63424	0.01695
	318.15	-5.89349	-1.00034	-4.32192	-2.20476	0.02584
	323.15	-6.74481	-1.09255	-5.18975	-2.91245	0.03266
	{Diethyl carbonate + Aniline}					
	293.15	-1.08659	0.31255	-0.05535	-0.03884	0.00178
	298.15	-1.18733	0.33495	-0.06297	-0.03873	0.00178
	303.15	-1.29522	0.36127	-0.07067	-0.03347	0.00195
β^E (10^{-10} $N^{-1}m^2$)	308.15	-1.40968	0.39797	-0.07446	-0.04149	0.00207
	313.15	-1.53316	0.43268	-0.08479	-0.03993	0.00216
	318.15	-1.66582	0.47382	-0.10112	-0.05306	0.00243
	323.15	-1.80978	0.51178	-0.10784	-0.03690	0.00241
	293.15	-0.11004	-0.04051	-0.00766	0.00573	0.00032
	298.15	-0.10925	-0.03948	-0.00708	0.00481	0.00027
	303.15	-0.10871	-0.03893	-0.00727	0.00523	0.00031
Z^E	308.15	-0.10850	-0.03961	-0.00735	0.00535	0.00032
(10^6 Kg $m^{-2}s^{-1}$)	313.15	-0.10833	-0.03964	-0.00758	0.00539	0.00031
	318.15	-0.10808	-0.04003	-0.00641	0.00838	0.00036
	323.15	-0.10800	-0.03947	-0.00716	0.00604	0.00032
	293.15	-100.586	-37.9272	-8.06695	6.98964	0.24156
	298.15	-99.946	-37.0429	-7.42323	6.62878	0.23348
	303.15	-99.524	-36.5856	-7.25354	5.88761	0.24418
ΔU (m/s)	308.15	-99.335	-37.5375	-7.84840	7.23918	0.24409
	313.15	-99.151	-37.5056	-7.72138	6.93625	0.24533
	318.15	-99.033	-38.0965	-6.83319	9.16182	0.27378
	323.15	-98.827	-37.6306	-7.51678	6.73862	0.24531

interaction between unlike molecules [14]. According to Fort and Moore [15], a negative excess compressibility is an indication of strong molecular interaction in the liquid mixtures while a positive value indicates a weak interaction attributable to dispersion forces. Also the magnitude of the excess function depends on the relative strength of interaction.

Figure **1a**, **b**, **c** shows the variations of β^E as a function of mole fraction of diethyl carbonate x_1.It can be seen that the excess adiabatic compressibility values are negative for all the mixtures over the entire range of composition at all temperatures. The variation of β^E with temperature is in a systematic manner. The negative values of $\beta^E$are found to increase with increasing temperature. The more negative value in each binary system is found to occur at x_1 = 0.6 indicating the formation of a stable complex between DEC and three benzene derivatives at this composition. The chemical effect includes charge transfer forces, formation of hydrogen bonds and other complex forming interactions making negative contribution towards β^E [16].

Generally, the excess properties are considered to be the reflecting agents of magnitude of polarity at the site of interactions in the molecules. The negative β^E values decrease in the following order: DEC + Aniline > DEC + nitrobenzene > DEC + chlorobenzene.



a b

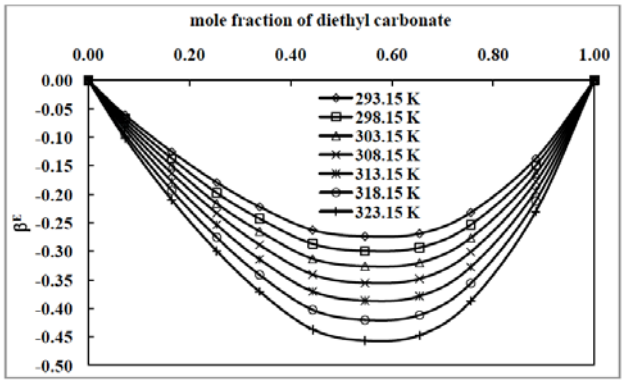


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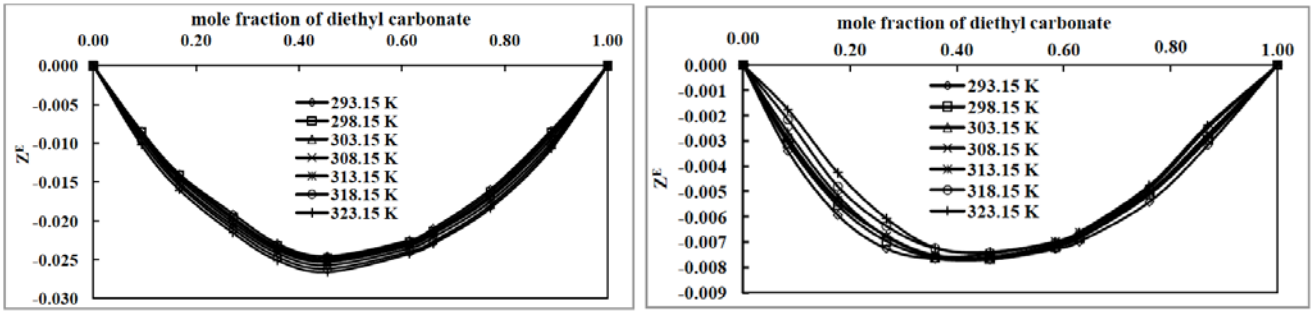
Figure 1: a: Variation of excess adiabatic compressibility (β^E) with mole fraction of diethyl carbonate for diethyl carbonate + nitrobenzene system at different temperatures.
b: Variation of excess adiabatic compressibility (β^E) with mole fraction of diethyl carbonate for diethyl carbonate + chlorobenzene system at different temperatures.
c: Variation of excess adiabatic compressibility (β^E) with mole fraction of diethyl carbonate for diethyl carbonate + aniline system at different temperatures.



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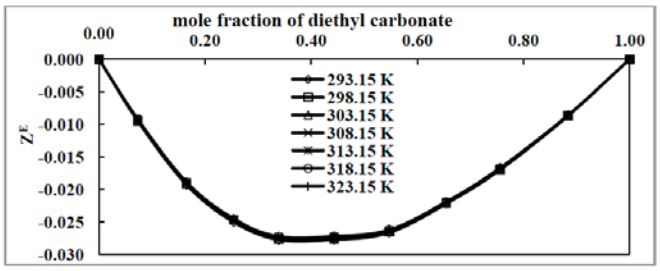

c

Figure 2: a: Variation of excess acoustic impedance (Z^E) with mole fraction of diethyl carbonate for diethyl carbonate + nitrobenzene system at different temperatures.
b: Variation of excess acoustic impedance (Z^E) with mole fraction of diethyl carbonate for diethyl carbonate + chlorobenzene system at different temperatures.
c: Variation of excess acoustic impedance (Z^E) with mole fraction of diethyl carbonate for diethyl carbonate + aniline system at different temperatures.

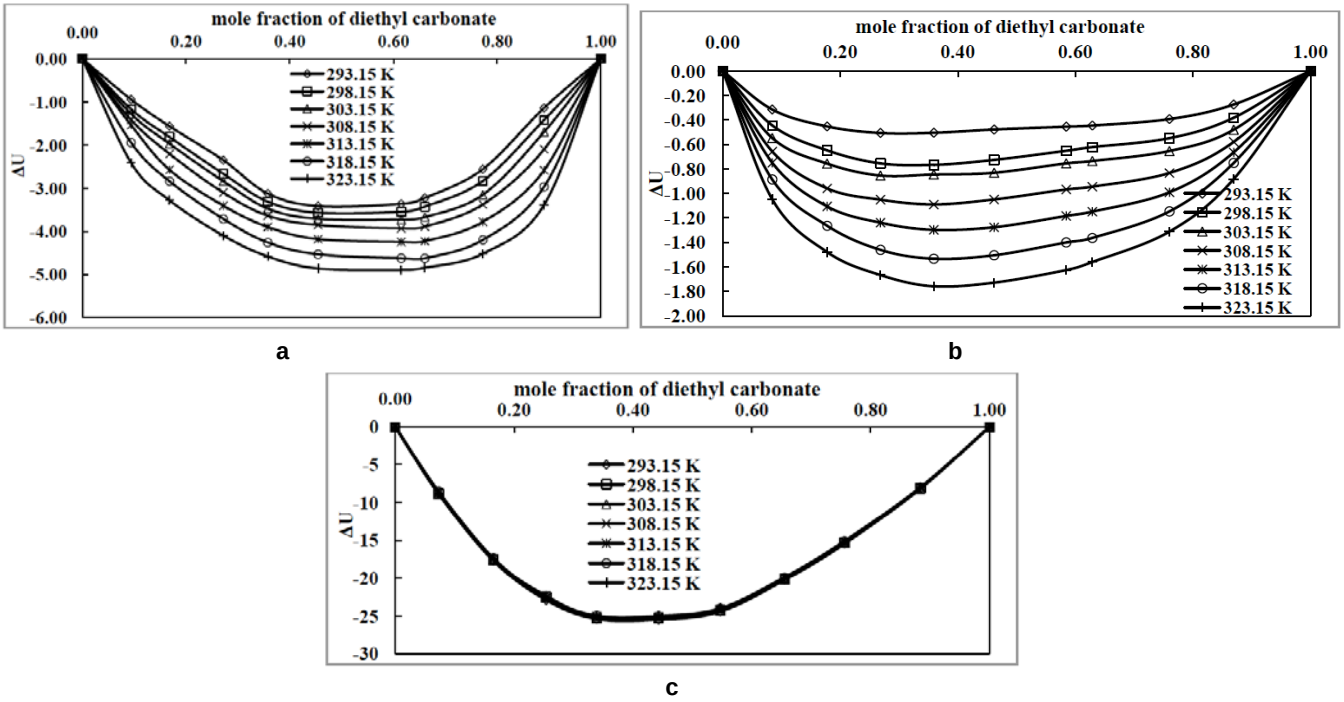

Figure 3: a: Variation of deviation in ultrasonic velocity (ΔU) with mole fraction of diethyl carbonate for diethyl carbonate + nitrobenzene system at different temperatures.

b: Variation of deviation in ultrasonic velocity (ΔU) with mole fraction of diethyl carbonate for diethyl carbonate + chlorobenzene system at different temperatures.

c: Variation of deviation in ultrasonic velocity (ΔU) with mole fraction of diethyl carbonate for diethyl carbonate + aniline system at different temperatures.

Acoustic impedance is important in the determination of acoustic transmission and reflection at the boundary of two materials having different acoustic impedance. It is also useful in the designing of ultrasonic transducers and for assessing absorption of sound in a medium.

The excess acoustic impedance, Z^E as a function of mole fraction of diethyl carbonate $x_1$is presented in Figure **2 a**, **b**, **c**. It is observed that the excess acoustic impedance values are negative for all the mixtures over the entire range of composition at all temperatures. The negative values of Z^E are found to increase with an increase in temperature. The change in Z^E values with temperatureis found to be very small in case of diethyl carbonate + aniline mixture. The negative values of Z^E are in agreement with the negative values of β^E. Based on the literature [17] it can be summarized that more than one type of interaction be present in a given system.

The ΔU values (Figure **3a**, **b**, **c**) exhibit completely negative deviation over the entire composition range for all the three systems studied. The negative ΔU may be due to the structure breaking resulting in expansion and the speeds of sound through the mixture will be slower as suggested by Reddy *et al*. [18]. The nature ΔU *vs.* x_1 curves for all the mixtures in the investigated temperature interval is almost identical. It is also noticeable that the values of the excess speed of sound do not show significant variation with increments in temperature for DEC + aniline system (Figure **3c**).

CONCLUSION

The values of ultrasonic velocities, densities at (293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15) K have been determined over the entire composition range for the binary mixtures of diethyl carbonate with nitrobenzene, chlorobenzene and aniline. From the experimental data excess adiabatic compressibility, excess acoustic impedance and deviation in ultrasonic velocity have been calculated. The excess values and deviations are observed to be negative for all the mixtures at all the temperatures studied. The Redlich-Kister equation has been used to estimate the parameters and standard deviations.

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