Research Article

Theoretical Evaluation of Ultrasonic Velocities in Binary Liquid Mixtures Containing Cyclohexanone

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ABSTRACT

Ultrasonic velocities are calculated in binary liquid mixtures of cyclohexanone with n-heptanol, n-octanol and iso-octanol at temperatures, T= (303.15, 308.15, 313.15 and 318.15) K over the entire mole fraction range. The experimental values are compared with the theoretical values of Nomoto and Van Dael-Vangeel. The observed deviation of theoretical values of velocity from the experimental values shows that the molecular interaction is taking place between the unlike molecules in the liquid mixture. The deviation in the variation of U^2 / U^2_{imx} from unity has also been evaluated for explaining the non-ideality in the mixtures. It is observed that the Nomoto's relation provides the best results for the systems cyclohexanone + n-heptanol and cyclohexanone + iso-octanol while Van Dael's relation gives better agreement for the system cyclohexanone + n-octanol.

Keywords: Theoretical velocities, cyclohexanone, n-heptanol, iso-octanol, ultrasoni velocity.

INTRODUCTION

In recent years, Ultrasonic investigations find extensive applications in characterizing of thermodynamic and physico-chemical aspects of binary and ternary liquid mixtures (Troncoso et al., 2001). Theoretical evaluation of ultrasonic velocity in binary liquid mixtures and its correlation to study molecular interaction has been successfully done in recent years (Narendra et al., 2012; Rao et al., 2007; Jaseem et al., 2006; Naidu et al., 2002) using the theoretical relations of (Nomoto, 1958) Van Dael and Vangeel (Subhash C. Bhatia et al., 2010), Impedance dependence (Baluja and Parsania, 1995), Rao's specific velocity (Sravana Kumar and Krishna Rao, 2007) and Junjie's (Azhagiri et al., 2009) equations.

The aim of the present investigation is to compare the sound velocity in three binary mixtures evaluated from various theories and relations. Nomoto's relation based upon the assumption of linearity of sound velocity and upon the additivity of molar volumes. Van Dael-Vangeel evaluated sound velocity in binary liquid mixtures on the basis of additivity of the adiabatic compressibilities. An attempt has been made to compare the merits of the relations for the binary liquid mixtures investigated at different temperatures. The relative applicability of these theories to the present systems have been checked and discussed. The results are explained in terms of intermolecular interactions occurring in these binary systems. The deviation in the variation of U^2 / U^2_{imx} from unity has also been evaluated for explaining the non-ideality in the mixtures.

Experimental

The mass fractions of the liquids, alcohols (obtained from SRL) are as follows: n-Heptanol (0.99), n-octanol (0.990) and iso-octanol (0.990) and that of the liquid, cyclohexanone (obtained from Merck) is 0.99. They were purified by standard procedure (Perrin and Armarego, 1980). Job's method of continuous variation was used to prepare the mixtures of required

proportions. The mixtures were preserved in well-Stoppard conical flasks. After mixing the liquids thoroughly, the flasks were left undisturbed to allow them to attain thermal equilibrium.

The densities of pure liquids and liquid mixtures were measured by using a specific gravity bottle with an accuracy of ± 0.5%. Weights were measured with an electronic balance (Shimadzu AUY220, Japan) capable of measuring up to an accuracy of 0.1mg. An average of 4-5 measurements was taken for each sample.

The ultrasonic velocities were measured by using single crystal ultrasonic pulse echo interferometer (Mittal enterprises, India; Model: F-80X). It consists of a high frequency generator and a measuring cell. The measurements of ultrasonic velocities were made at a fixed frequency of 3MHz. The calibration of the equipment was done by measuring the velocity in carbontetrachloride and benzene. The results are in good agreement with the literature values (Lide, 1995). The ultrasonic velocity has an accuracy of ± 0.5 ms^{-1}. The temperature was controlled by circulating water around the liquid cell from thermostatically controlled constant temperature water bath.

Nomoto established an empirical relation for ultrasonic velocity in binary liquid mixtures as

$$U_N = [(x_1R_1+x_2R_2)/(x_1V_1+x_2V_2)]^3 \qquad (1)$$

Where R is molar sound velocity, x_1 and x_2 are the mole fractions of 1^{st} and 2^{nd} components of the liquid mixture and V is molar volume.

Van Dael-Vangeel Ideal mixing relation

$$U_{imx} = [(x_1/M_1U^2_1 + x_2/M_2U^2_2)(x_1M_1+x_2M_2)]^{-1/2} \qquad (2)$$

Where U_{imx} is the ideal mixing ultrasonic velocity in liquid mixture. U_1 and U_2 are ultrasonic velocities of individual compounds.

RESULTS AND DISCUSSION

The experimental values along with the values calculated theoretically using the relations of Nomoto and Van Dael ideal mixing for the three systems namely n-heptanol, n-octanol and iso-octanol with cyclohexanone at the temperatures of 303.15K, 308.15K, 313.15K and 318.15K are given in Tables 1-3. The caculated percentage deviations at different temperatures are given in the Tables 4-6.

It can be seen from Tables 1-3 that the theoretical values of ultrasonic velocity computed by various theories show deviations from experimental values. The limitations and approximations incorporated in these theories are responsible for it. It is assumed that all the molecules are spherical in shape, which is not true every time. In Nomoto's theory, it is supposed that the volume does not change on mixing. Therefore, no interaction between the components of liquid mixtures has been taken into account.

Similarly, the assumption for the formation of ideal mixing relation is that, the ratio of specific heats of ideal mixtures and the volumes are also equal. Again, no molecular interaction is taken into account. But on mixing two liquids, the interaction between the molecules of the two liquids takes place because of presence of various types of forces such as dispersion forces, charge transfer, hydrogen bonding, dipole-dipole and dipole-induced dipole interactions. Thus, the observed deviation of theoretical values of velocity from the experimental values shows that the molecular interaction is taking place between the unlike molecules in the liquid mixture.

From Tables 1 and 3 it is observed that for the systems cyclohexanone + n-heptanol, cyclohexanone+iso-octanol, there is an agreement between experimental and theoretical values calculated by Nomoto relation followed by Van Dael ideal mixing relation whereas for the system (Table-2) of cyclohexanone + n-octanol, an agreement is present between experimental and theoretical values in Van Dael ideal mixing followed by Nomoto relation at all the four temperatures. The results are in good agreement with conclusions drawn by other researchers (Ali et al., 2000).

The percentage deviations of the ultrasonic velocity are both negative and positive. Such deviations indicate the non-

ideal behavior of liquid mixtures. The ratio U^2 / U^2_{imx} is used as an important tool to measure the non-ideality in the mixtures, especially in those cases where the properties other than sound velocity are not known. A perusal of Tables (4-6) indicates considerable deviations from ideality, which may be due to the existence of week dipole-dipole interactions in liquid mixtures (Anwar Ali et al., 2002).

The deviation of the ratio U^2 / U^2_{imx} from unity and its variation as a function of mole fraction of cyclohexanone is a direct measure of non-ideality of the system as a consequence of association or other type of interactions. The negative values of α □□in all the systems clearly indicate the existence of weak dipole-dipole interactions in mixtures (Santhi et al., 2010).

CONCLUSION

Ultrasonic velocities are calculated experimentally and the results are compared with the values calculated from various theories and relations. It is understood from the results that out of all the theories and the relations discussed above, the Nomoto's relation provides the best results for the systems cyclohexanone + n-heptanol and cyclohexanone + iso-octanol while Van Deal's relation gives better agreement for the system cyclohexanone + n-octanol. The linearity of molar sound velocity and additivity of molar volumes suggested by Nomoto in deriving the empirical relation have been found to be valid for the first and third systems.

Table 1: Experimental and theoretical values of velocities ($m.s^{-1}$) in cyclohexanone + n-heptanol system at different temperatures

Mole fraction, x	U_{exp} (m/s)	U_{Nomoto}	U_{imx}	U_{exp} (m/s)	U_{Nomoto}	U_{imx}
	T = 303.15 K			T = 308.15 K		
0.0000	1312.00	1308.14	1312.00	1298.00	1298.00	1298.00
0.1322	1319.66	1317.91	1323.24	1301.88	1304.31	1305.06
0.2553	1327.83	1327.73	1334.06	1307.09	1310.64	1311.98
0.3702	1336.80	1337.59	1344.48	1313.14	1316.99	1318.76
0.4776	1346.57	1347.50	1354.54	1319.42	1323.36	1325.39
0.5783	1355.98	1357.47	1364.23	1325.56	1329.75	1331.87
0.6729	1365.66	1367.48	1373.59	1331.91	1336.16	1338.20
0.7619	1375.45	1377.53	1382.63	1338.40	1342.59	1344.37
0.8458	1385.43	1387.64	1391.37	1345.32	1349.04	1350.39
0.9250	1396.03	1397.80	1399.82	1352.99	1355.51	1356.27
1.0000	1408.00	1408.00	1408.00	1362.00	1362.00	1362.00
	T = 313.15 K			T = 318.15 K		
0.0000	1278.00	1278.00	1278.00	1255.20	1255.20	1255.20
0.1322	1282.63	1284.99	1286.02	1259.23	1262.86	1264.08
0.2553	1288.06	1292.00	1293.83	1265.54	1270.54	1272.67
0.3702	1294.39	1299.04	1301.42	1272.50	1278.23	1280.99
0.4776	1301.19	1306.10	1308.81	1279.67	1285.95	1289.06
0.5783	1308.07	1313.19	1315.99	1287.25	1293.69	1296.87
0.6729	1314.94	1320.30	1322.97	1294.99	1301.44	1304.44
0.7619	1322.28	1327.44	1329.75	1302.86	1309.22	1311.78
0.8458	1329.98	1334.60	1336.35	1310.93	1317.01	1318.90
0.9250	1338.22	1341.78	1342.76	1319.75	1324.83	1325.80
1.0000	1349.00	1349.00	1349.00	1332.50	1332.66	1332.50

Table 2: Experimental and theoretical values of velocities ($m.s^{-1}$) in cyclohexanone + n-octanol system at different temperatures

Mole fraction, x	U_{exp} (m/s)	U_{Nomoto}	U_{imx}	U_{exp} (m/s)	U_{Nomoto}	U_{imx}
	T = 303.15 K			T = 308.15 K		
0.0000	1340.00	1340.00	1340.00	1322.40	1322.40	1322.40
0.1454	1344.26	1346.70	1345.07	1321.57	1326.33	1322.83
0.2769	1349.92	1353.42	1350.94	1323.62	1330.26	1324.71
0.3963	1356.64	1360.16	1357.38	1327.07	1334.20	1327.66
0.5052	1363.59	1366.93	1364.21	1330.92	1338.15	1331.41
0.6050	1370.38	1373.72	1371.30	1334.93	1342.10	1335.75
0.6967	1377.24	1380.53	1378.56	1338.97	1346.07	1340.53
0.7813	1383.85	1387.36	1385.91	1343.25	1350.04	1345.62
0.8597	1390.39	1394.22	1393.29	1347.69	1354.02	1350.94
0.9324	1397.71	1401.10	1400.66	1352.96	1358.00	1356.42
1.0000	1408.00	1408.00	1408.00	1362.00	1362.00	1362.00
	T = 313.15 K			T = 318.15 K		
0.0000	1298.00	1298.00	1298.00	1279.00	1279.00	1279.00
0.1454	1297.39	1303.04	1300.47	1277.86	1284.34	1281.97
0.2769	1301.37	1308.10	1304.05	1281.58	1289.68	1285.97
0.3963	1306.23	1313.17	1308.46	1286.45	1295.04	1290.71
0.5052	1311.30	1318.25	1313.46	1291.87	1300.39	1295.99
0.6050	1316.14	1323.34	1318.89	1297.32	1305.76	1301.65
0.6967	1321.09	1328.45	1324.62	1302.23	1311.13	1307.58
0.7813	1326.48	1333.56	1330.56	1307.21	1316.50	1313.69
0.8597	1331.39	1338.70	1336.64	1313.05	1321.88	1319.91
0.9324	1337.67	1343.84	1342.80	1319.48	1327.27	1326.19
1.0000	1349.00	1349.00	1349.00	1332.50	1332.66	1332.50

Table 3: Experimental and theoretical values of velocities ($m.s^{-1}$) in cyclohexanone + iso- octanol system at different temperatures

Mole fraction, x	U_{exp} (m/s)	U_{Nomoto}	U_{imx}	U_{exp} (m/s)	U_{Nomoto}	U_{imx}
	T = 303.15 K			T = 308.15 K		
0.0000	1303.20	1303.20	1303.20	1291.20	1291.20	1291.20
0.1445	1310.31	1313.44	1314.11	1294.26	1298.19	1296.90
0.2753	1320.36	1323.73	1325.07	1300.41	1305.20	1303.25
0.3944	1330.80	1334.07	1335.99	1307.81	1312.23	1310.07
0.5033	1341.38	1344.47	1346.81	1315.08	1319.28	1317.21
0.6031	1351.80	1354.92	1357.48	1322.33	1326.35	1324.54
0.6951	1362.21	1365.43	1367.99	1329.09	1333.44	1332.00
0.7800	1372.46	1375.99	1378.31	1335.87	1340.55	1339.52
0.8587	1382.61	1386.61	1388.42	1342.77	1347.68	1347.05
0.9319	1393.38	1397.28	1398.32	1350.40	1354.83	1354.55
1.0000	1408.00	1408.00	1408.00	1362.00	1362.00	1362.00
	T = 313.15 K			T = 318.15 K		
0.0000	1284.20	1284.20	1284.20	1267.00	1267.00	1267.00
0.1445	1285.88	1290.61	1288.96	1268.75	1273.54	1271.95
0.2753	1291.25	1297.04	1294.50	1273.56	1280.08	1277.64
0.3944	1298.00	1303.48	1300.60	1280.04	1286.63	1283.84
0.5033	1305.02	1309.94	1307.09	1286.94	1293.19	1290.41
0.6031	1311.64	1316.41	1313.84	1293.75	1299.75	1297.23
0.6951	1318.30	1322.90	1320.76	1300.27	1306.32	1304.19
0.7800	1324.73	1329.40	1327.79	1306.69	1312.90	1311.25
0.8587	1330.90	1335.92	1334.86	1313.30	1319.48	1318.35
0.9319	1337.49	1342.45	1341.94	1320.75	1326.07	1325.44
1.0000	1349.00	1349.00	1349.00	1332.50	1332.50	1332.50

Table 4: Percentage deviation between experimental and theoretical values of velocities in cyclohexanone + n-heptanol system at different temperatures

Mole fraction, x	% U_{Nom}	% U_{imx}	U^2/U_{imx}^2	$\alpha = [U^2/U_{imx}^2]^{-1}$	% U_{Nom}	% U_{imx}	U^2/U_{imx}^2	$\alpha = [U^2/U_{imx}^2]^{-1}$
	T = 303.15 K				T = 308.15 K			
0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	1.0000	0.0000
0.1322	-0.1325	0.2713	0.9946	-0.0054	0.1867	0.2442	0.9951	-0.0049
0.2553	-0.0076	0.4693	0.9907	-0.0093	0.2721	0.3747	0.9925	-0.0075
0.3702	0.0592	0.5749	0.9886	-0.0114	0.2935	0.4284	0.9915	-0.0085
0.4776	0.0695	0.5917	0.9879	-0.0117	0.2987	0.4528	0.9910	-0.0090
0.5783	0.1096	0.6086	0.9881	-0.0121	0.3157	0.4757	0.9906	-0.0094
0.6729	0.1326	0.5804	0.9885	-0.0115	0.3191	0.4721	0.9906	-0.0094
0.7619	0.1516	0.5223	0.9896	-0.0104	0.3126	0.4458	0.9911	-0.0089
0.8458	0.1595	0.4287	0.9915	-0.0085	0.2766	0.3774	0.9925	-0.0075
0.9250	0.1270	0.2720	0.9946	-0.0054	0.1861	0.2424	0.9952	-0.0048
1.0000	0.0000	0.0000	1.0000	0.0000	-0.0001	0.0000	1.0000	0.0000
	T = 313.15 K				T = 318.15 K			
0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	1.0000	0.0000
0.1322	0.1835	0.2641	0.9947	-0.0053	0.2885	0.3852	0.9923	-8.3492
0.2553	0.3062	0.4481	0.9911	-0.0089	0.3951	0.5638	0.9888	-4.6219
0.3702	0.3593	0.5435	0.9892	-0.0108	0.4501	0.6672	0.9868	-3.4570
0.4776	0.3770	0.5850	0.9884	-0.0116	0.4909	0.7338	0.9855	-2.9121
0.5783	0.3912	0.6052	0.9880	-0.0120	0.5002	0.7476	0.9852	-2.6097
0.6729	0.4076	0.6105	0.9879	-0.0121	0.4981	0.7297	0.9856	-2.4207
0.7619	0.3901	0.5653	0.9888	-0.0112	0.4881	0.6848	0.9864	-2.2932
0.8458	0.3475	0.4792	0.9905	-0.0095	0.4641	0.6079	0.9880	-2.2023
0.9250	0.2667	0.3398	0.9932	-0.0068	0.3844	0.4582	0.9909	-2.1361
1.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	1.0000	0.0000

Table 5: Percentage deviation between experimental and theoretical values of velocities in cyclohexanone + n-octanol system at different temperatures

Mole fraction, x	% U_{Nom}	% U_{imx}	U^2/U_{imx}^2	$\alpha = [U^2/U_{imx}^2]^{-1}$	% U_{Nom}	% U_{imx}	U^2/U_{imx}^2	$\alpha = [U^2/U_{imx}^2]^{-1}$
	T = 303.15 K				T = 308.15 K			
0.0000	0.0000	0.0000	1.0000	0.0000	0.0001	0.0000	1.0000	0.0000
0.1454	0.1814	0.0604	0.9990	-0.0012	0.3599	0.0954	0.9981	-0.0019
0.2769	0.2590	0.0757	0.9985	-0.0015	0.5012	0.0821	0.9972	-0.0016
0.3963	0.2595	0.0547	0.9986	-0.0011	0.5370	0.0446	0.9968	-0.0009
0.5052	0.2447	0.0455	0.9985	-0.0009	0.5433	0.0374	0.9967	-0.0007
0.6050	0.2437	0.0675	0.9987	-0.0013	0.5375	0.0617	0.9969	-0.0012
0.6967	0.2391	0.0960	0.9986	-0.0019	0.5299	0.1162	0.9970	-0.0023
0.7813	0.2539	0.1484	0.9988	-0.0030	0.5052	0.1762	0.9973	-0.0035
0.8597	0.2753	0.2081	0.9988	-0.0041	0.4692	0.2408	0.9976	-0.0048
0.9324	0.2427	0.2113	0.9992	-0.0042	0.3727	0.2554	0.9983	-0.0051
1.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	1.0000	0.0000
	T = 313.15 K				T = 318.15 K			
0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	1.0000	0.0000
0.1454	0.4355	0.2369	0.9973	-0.0047	0.5068	0.3214	0.9962	-0.0064
0.2769	0.5167	0.2060	0.9959	-0.0041	0.6324	0.3423	0.9944	-0.0068
0.3963	0.5311	0.1707	0.9952	-0.0034	0.6675	0.3311	0.9936	-0.0066
0.5052	0.5299	0.1647	0.9950	-0.0033	0.6599	0.3187	0.9933	-0.0063
0.6050	0.5471	0.2087	0.9952	-0.0042	0.6504	0.3337	0.9933	-0.0066
0.6967	0.5569	0.2671	0.9954	-0.0053	0.6832	0.4107	0.9934	-0.0082
0.7813	0.5343	0.3076	0.9956	-0.0061	0.7112	0.4959	0.9937	-0.0098
0.8597	0.5489	0.3943	0.9960	-0.0078	0.6729	0.5226	0.9944	-0.0104
0.9324	0.4616	0.3838	0.9970	-0.0076	0.5907	0.5092	0.9960	-0.0101
1.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	1.0000	0.0000

Table 6: Percentage deviation between experimental and theoretical values of velocities in cyclohexanone + iso-octanol system at different temperatures

Mole fraction, x	% U_{Nom}	% U_{imx}	U^2/U_{imx}^2	$\alpha = [U^2/U_{imx}^2]^{-1}$	% U_{Nom}	% U_{imx}	U^2/U_{imx}^2	$\alpha = [U^2/U_{imx}^2]^{-1}$
	T = 303.15 K				T = 308.15 K			
0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	1.0000	0.0000
0.1445	0.2389	0.2905	0.9942	-0.0058	0.3032	0.2033	0.9959	-0.0041
0.2753	0.2550	0.3566	0.9927	-0.0071	0.3685	0.2190	0.9953	-0.0044
0.3944	0.2459	0.3898	0.9922	-0.0078	0.3377	0.1730	0.9953	-0.0035
0.5033	0.2303	0.4043	0.9920	-0.0080	0.3188	0.1615	0.9953	-0.0032
0.6031	0.2309	0.4202	0.9919	-0.0084	0.3034	0.1671	0.9954	-0.0033
0.6951	0.2364	0.4242	0.9921	-0.0084	0.3270	0.2192	0.9956	-0.0044
0.7800	0.2572	0.4257	0.9924	-0.0085	0.3501	0.2733	0.9960	-0.0054
0.8587	0.2893	0.4202	0.9928	-0.0084	0.3655	0.3187	0.9968	-0.0063
0.9319	0.2799	0.3544	0.9946	-0.0071	0.3280	0.3075	0.9982	-0.0061
1.0000	0.0000	0.0000	1.0000	0.0000	-0.0001	0.0000	1.0000	0.0000
	T = 313.15 K				T = 318.15 K			
0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	1.0000	0.0000
0.1445	0.3685	0.2401	0.9952	-0.0048	0.3769	0.2520	0.9950	-0.0050
0.2753	0.4488	0.2523	0.9946	-0.0050	0.5118	0.3199	0.9936	-0.0064
0.3944	0.4224	0.2004	0.9943	-0.0040	0.5152	0.2974	0.9932	-0.0059
0.5033	0.3774	0.1587	0.9942	-0.0032	0.4853	0.2695	0.9930	-0.0054
0.6031	0.3638	0.1674	0.9942	-0.0033	0.4643	0.2689	0.9930	-0.0054
0.6951	0.3491	0.1868	0.9942	-0.0037	0.4658	0.3021	0.9933	-0.0060
0.7800	0.3524	0.2305	0.9947	-0.0046	0.4750	0.3490	0.9936	-0.0069
0.8587	0.3772	0.2977	0.9956	-0.0059	0.4705	0.3842	0.9944	-0.0076
0.9319	0.3712	0.3332	0.9971	-0.0066	0.4024	0.3548	0.9961	-0.0071
1.0000	-0.0002	0.0000	1.0000	0.0000	0.0000	0.0000	1.0000	0.0000

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