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Studies on acoustical parameters, thermodynamical properties and theoretical evaluation of morpholine with *ortho* xylene in isopropanol at 303K, 308K and 313K using ultrasonic sound

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Ultrasonic velocity (U), density (ρ) and viscosity (η) have been measured for morpholine with *ortho*-xylene in isopropanol at 303K, 308K & 313K. From these experimental values, the adiabatic compressibility (β), acoustic impedance (Z), free length (L_f), free volume (V_f), internal pressure (π_i), cohesive energy (CE), Lenard Jones Potential (LJP) and viscous relaxation time (r) have been calculated and their importance are discussed. Formation constant (K) values of the compounds have been determined by using a modified Bhat equation. The thermodynamical parameters have also been computed and the complexes are thermodynamically stable as noticed from their free energy of formation values. The theoretical values of ultrasonic velocity have been calculated at respective temperatures using the Nomoto's relation (U_{NOM}), Impedance relation (U_{IMP}), Ideal mixing relation (U_{IMR}) and Junjie's relation (U_{JUN}) and these values are further confirmed by Chi-Square test analysis. The molecular interaction parameter has been calculated by using the experimental and theoretical ultrasonic velocity values.

Keywords: Formation constant, Nomoto's relation, Acoustic impedance, Xylene, Morpholine, Viscous relaxation time

Literature survey shows that acoustical, thermodynamic parameters and excess parameters are very useful to identify the nature and strength of molecular interactions in the liquid mixtures of binary as well as ternary systems. Also it is noted that most of the previous studies were concentrated on xylene family (*ortho*, *meta* and *para*) with some other compound of binary system. Hence, the present study focused on ternary system involving morpholine with *ortho* xylene in isopropanol which reported about the interactions of solute and solvent and in particular electron donor (morpholine) and acceptor (*ortho* xylene). Further the hetero cyclic compound, morpholine which forms strong charge-transfer-complex because of amine group exhibited by it. A large body of *in vivo* studies has demonstrated morpholine's potential to not only increase potency but also provide compounds with desirable drug-like properties and improved pharmacokinetics. The literature study describes the medicinal chemistry/ pharmacological activity of morpholine derivatives on various therapeutically related molecular targets, attempting to highlight the importance of the morpholine ring in drug design and frequently used heterocycle in medicinal chemistry and a privileged structural component of bioactive molecules. This is mainly due to its contribution to a plethora of biological activities as well as to an improved pharmacokinetic profile of such bioactive molecules[1-15]. This anomalous character of morpholine and its derivatives motivates the author to undertake the present study. O-xylene, another compound used in this current investigation is a pungent, volatile organic compound which is widely used in printing process and behaves as electron acceptor meanwhile morpholine acts as donor. In fact, the third one in this ternary system which is under study is isopropyl alcohol which is widely used as solvent and polar in nature. Some researchers[16-19] calculated ultrasonic velocities based on the theories such as Nomoto's relations [20], Impedance relation[21], Junjie[22], Ideal mixing relation and which were correlated with the experimental ultrasonic velocity along with molecular interactions.

Experimental Section

All chemicals used in the present study were analytical reagent grade. A single crystal variable path ultrasonic interferometer (Model F81) operating at 2 MHz frequency supplied by Mittal Enterprises, New Delhi, India was used in the measurement of ultrasonic velocity of liquid mixtures with the accuracy of ± 0.01 ms^{-1}. The density (ρ) of solutions was measured using specific gravity bottles of capacity 10mL.Viscosity was measured by Ostwald's viscometer, in which the flow time for solutions was measured through a digital stop clock of accuracy ± 0.01s. The temperature was maintained at 303,308 and 313 K during the measurement of ultrasonic velocity, density and viscosity values.

Theory and Calculations

Extending author's previous studies and applying formulae to the present system, the calculation part is provided in the supplementary section[23-26].

Results and Discussion

The experimental values of ultrasonic velocity, density and viscosity of *o*-xylene (acceptor) and morpholine (donor) in isopropanol medium at 303K, 308K and 313K are given in Table 1–4 and Table 5 along with calculated acoustical parameters

Table 1 — Ultrasonic velocity (U), Density (ρ), Viscosity (η), Adiabatic compressibility(β), Free length (L_f) , Acoustic Impendance(Z), Internal Pressure (π_i) and formation constant(K) values for morpholine with *o*-xylene in isopropanol at 303K

Concn ×10^{-3}M	U m/s	ρ kg m^{-3}	η cP	β Kg^{-1}ms^{2}	L_f Å	z kg^{-2}s^{-1}	π_i atm	K dm^{3}mol^{-1}
1	1431.65	876.7	0.7261	5.57	1.47	1.26	2.156	-720.4
2	1472.11	906.1	0.8161	5.09	1.4	1.33	2.215	-384.8
3	1496.21	926.2	0.9264	4.82	1.37	1.39	2.254	-530.4
4	1525.86	952.6	1.0265	4.51	1.32	1.45	2.291	-446.9
5	1551.74	978.1	1.1546	4.25	1.28	1.52	2.223	-923.1
6	1589.57	998.5	1.312	3.96	1.24	1.59	2.245	-1039
7	1628.42	1024.1	1.4764	3.68	1.19	1.67	2.216	-540.3
8	1656.94	1042.6	1.5823	3.5	1.16	1.73	2.236	-871.1
9	1691.54	1065.4	1.6885	3.28	1.13	1.8	2.243	-274
10	1738.36	1087.3	1.8763	3.04	1.09	1.89	2.244	-546

Table 2 — Cohesive energy(CE), viscous relaxation time(r), free volume(V_f), leonard jones potential(LJP) values for morpholine with o-xylene in isopropanol at 303K

Concn (M) ×10^{-3}	CE kJmol^{-1}	VRT(r)×10^{-10} s	V_f ×10^{-12} m^3/mol	LJP
1	1138.83	5.015	0.102	3.292
2	1215.88	5.727	6.680	3.304
3	1275.61	6.089	5.659	3.317
4	1317.27	6.347	4.997	3.334
5	1373.21	6.712	4.296	3.351
6	1436.39	7.078	3.677	3.359
7	1492.81	7.435	3.194	3.364
8	1522.96	7.504	2.955	3.368
9	1545.89	7.551	2.765	3.372
10	1596.64	7.773	2.459	3.372

Table 3 — Ultrasonic velocity (U), Density (ρ), Viscosity (η), Adiabatic compressibility(β), Free length (L_f) , Acoustic Impendance(Z), Internal Pressure (π_i) and formation constant(K) values for morpholine with *o*-xylene in isopropanol at 308K

Concn ×10^{-3}M	U m/s	ρ kg m^{-3}	η cP	β Kg^{1}ms^{2}	L_f Å	z kg^{-2}s^{-1}	π_i atm	K dm^{-3}mol^{-1}
1	1419.26	875.1	0.6911	5.67	1.48	1.24	1.16	-571.1
2	1451.45	894.2	0.7625	5.31	1.43	1.3	1.22	-818.4
3	1491.63	926.4	0.8106	4.85	1.37	1.38	1.27	-889.8
4	1532.42	949.2	0.9261	4.49	1.32	1.45	1.36	-1036
5	1575.25	968.7	1.0211	4.16	1.27	1.53	1.43	-306.6
6	1596.11	988.1	1.1459	3.97	1.24	1.58	1.52	-1316
7	1639.25	1015.4	1.2956	3.66	1.19	1.66	1.63	-1297
8	1681.42	1039.5	1.3658	3.4	1.15	1.75	1.68	-1744
9	1726.72	1056.1	1.4119	3.18	1.11	1.82	1.7	-814
10	1761.92	1079.7	1.569	2.98	1.07	1.9	1.8	-926

Table 4 — Cohesive energy(CE), viscous relaxation time(r), free volume(V_f), leonard jones potential(LJP) values for morpholine with o-xylene in isopropanol at 308K

Concn (M) $\times10^{-3}$	CE $kJmol^{-1}$	VRT(r)$\times10^{-10}$ s	$V_f \times10^{-12}$ m^3/mol	LJP
1	1116.56	5.315	0.108	4.426
2	1188.83	5.432	7.242	4.480
3	1194.97	5.676	6.883	4.531
4	1249.99	5.780	5.869	4.445
5	1285.84	6.191	5.284	4.439
6	1344.32	6.506	4.533	4.436
7	1397.76	6.344	3.924	4.410
8	1405.99	6.074	3.766	4.404
9	1403.23	6.381	3.729	4.379
10	1453.65	5.515	3.281	4.366

Table 5 — Ultrasonic velocity (U), Density (ρ), Viscosity (η), Adiabatic compressibility(β), Free length (L_f) , Acoustic Impendance(Z), Internal Pressure (π_i) and formation constant(K) values for morpholine with *o*-xylene in isopropanol at 313K

Concn $\times10^{-3}$M	U m/s	ρ kg m^{-3}	η cP	β $Kg^{-1}ms^2$	L_fÅ	z $kg^{-2}s^{-1}$	π_i atm	K $dm^{-3}mol^{-1}$
1	1391.6	869.6	0.6724	5.94	1.52	1.21	1.15	-690.7
2	1426.2	881.5	0.7104	5.58	1.47	1.26	1.18	591.6
3	1455.9	899.7	0.8276	5.24	1.42	1.31	1.27	-638.2
4	1486.2	929.2	0.9158	4.87	1.37	1.38	1.36	-436.7
5	1509.4	942.5	1.0161	4.66	1.34	1.42	1.43	-700.7
6	1539.4	962.6	1.1268	4.38	1.3	1.48	1.51	-1740
7	1581.6	987.1	1.2361	4.05	1.25	1.56	1.59	-1040
8	1616.5	1011.2	1.3561	3.78	1.21	1.63	1.67	-548.3
9	1642.8	1027.1	1.4579	3.61	1.18	1.69	1.74	-951
10	1675.2	1046.5	1.5616	3.41	1.15	1.75	1.8	-886

such as adiabatic compressibility, free volume, internal pressure and cohesive energy with various concentrations ranges from 0.001 to 0.01M. From the tables, it is observed that ultrasonic velocity (U) increases with the concentration which conforms that there was a solvent stronger interaction between donor and acceptor systems. The velocity is increasing to prove the strong solute-solute interactions. It was implied that the strength of molecular interactions becomes high where the velocity maximum occurs. The increase in velocity also proved strong complex formation through intermolecular hydrogen bond and dipole-dipole interaction. Adiabatic compressibility β indicates the formation or dissociation of intermolecular bond. β and L_f values Table 1–4 and Table 5) indicates the reverse trend that the strong interaction between morpholine and *ortho* xylene. From the adiabatic compressibility (β) and free length (L_f) explained that the increasing close packing and tightly bound of the system[27]. These acoustical parameters also proved to detect the intermolecular interaction in ternary liquid mixtures. The internal pressure is also showed to analyze the force of attraction and force of repulsion in the ternary liquid mixtures[28]. The change of free volume (V_f) is totally reversed. The CE (Table 2, Table 4 and Table 6) increases as well as the intermolecular forces also increases[29]. The magnitude of CE also proves that the presence of strong dipole-dipole interaction in the intermolecular hydrogen bonding for ternary system. LJP increases with the concentration revealed that there was a extent of complexation of the system with all temperatures. The LJP values suggest the molecular interactions are strong in the present system because the values are found to be increases at all the temperatures while the concentration gets increases (Table 2, Table 4 and Table 6). The values of internal pressure and free volume are seems to be opposite trend with one another which means the internal pressure increases whereas free volume decreases at all the working temperatures (Table 1, Table 3 and Table 5). This tendency supports the close packing of molecules exhibited by the present system which is under investigation. The acoustic impedance values (Table 2, Table 4 and Table 6) argue for the presence of strong interaction. The internal pressure values increase with increase in concentration at irrespective of temperature which supports for the degree of complex formation. VRT, one of the valuable parameters, is time gap between ground state and excited state of the molecules which interpret the same type of interactions are observed in current study. The formation constant or stability constant K is the most significant one, because, it's values decide the stability of the complex. The formation constant K value is determined by using Kannappan equation[30] which may be helpful to assess the complex formation too. The free energy of formation and entropy change values are determined by using K values which also strongly support for the charge transfer complex formation since the negative values are recorded (Table 7) at 303K,308K and 308K. The entropy values advocate for the orderliness of the molecules in the present system. So this tendency, envisages the versatile character of morpholine ring which easily access the other two compounds that leads to facile synthetic routes in which amine group is easily introduced.

Theoretical values of ultrasonic speeds in ternary liquid mixtures determined using Nomoto's relation, Impedance dependence relation, ideal mixture relation and Junjie's relation along with experimental values at 303K, 308K and 313K are provided in Table 8, Table 9 and Table 10 and their respective percentage deviation are tabulated (Table 11, Table 12 and Table 13). Table 14 reveals the values of molecular interaction parameter at 303K, 308K and 313K, the values are found to be (except one or two) <1 which infers that there is a moderate type of interaction is predicted in the present system. Comparison of absolute average percentage deviation with related to experimentally measured ultrasonic velocities and values of Chi-square test at 303K, 308K and 313K are provided in Table 15. This additional study has been actually carried out as the supplement to the experimental study.

Table 6 — Cohesive energy(CE), viscous relaxation time(r), free volume(V_f), leonard jones potential(LJP) values for morpholine with o-xylene in isopropanol at 313K

Concn (M) $\times10^{-3}$	CE $kJmol^{-1}$	VRT(r)$\times10^{-10}$ s	$V_f\times10^{-12}$ m^3/mol	LJP
1	1114.58	5.234	0.094	1.951
2	1168.41	5.355	7.843	1.760
3	1242.55	5.906	6.434	1.473
4	1284.92	6.145	5.700	1.027
5	1328.66	6.400	4.992	0.844
6	1378.93	6.727	4.403	0.562
7	1414.88	6.845	3.991	0.245
8	1453.67	7.010	3.588	-0.067
9	1483.17	7.123	3.298	-0.264
10	1512.22	7.224	3.063	-0.500

Table 7 — The values of thermodynamical properties for morpholine with *o*-xylene in isopropanol at 303K, 308K & 313K

Concn	Delta G kJmol-1			Delta G* kJmol-1			Delta S Jmol-1		
(M)	303K	308K	313K	303K	308K	313K	303K	308K	313K
0.001	22.460	22.792	23.207	-7842.8	-8009.7	-8186.7	-113.69	-112.38	-111.15
0.002	22.538	22.881	23.276	-7851.5	-8019.7	-8194.4	-113.71	-112.41	-111.18
0.003	22.738	22.800	23.555	-7873.7	-8010.7	-8225.5	-113.79	-112.39	-111.28
0.004	22.835	22.954	23.669	-7884.5	-8027.9	-8237.9	-113.82	-112.44	-111.32
0.005	22.994	23.017	23.785	-7902.0	-8034.9	-8250.7	-113.88	-112.46	-111.36
0.006	23.157	23.211	23.927	-7919.8	-8056.4	-8266.2	-113.94	-112.53	-111.41
0.007	42.359	23.329	23.976	-9441.8	-8069.4	-8271.6	-118.96	-112.58	-111.42
0.008	42.406	23.269	24.044	-9444.6	-8062.8	-8279.0	-118.97	-112.55	-111.45
0.009	42.411	23.169	24.090	-9444.9	-8051.7	-8283.9	-118.97	-112.52	-111.46
0.01	42.495	23.289	24.130	-9449.9	-8065.0	-8288.2	-118.99	-112.56	-111.48

Table 8 — Theoretical values based on Ideal mixture relation (IMR), Nomoto's relation (NOM), Junjie's relation (JUN) and Impedance relation (IMP) for morpholine with o-xylene in isopropanol at 303K along with experimental velocity(U)

Concentration x10^{-3}M	U m/s	IMR	NOM	JUN	IMP
1	1431.65	1313.37	1337.19	1334.13	1337.69
2	1472.11	1295.57	1335.64	1329.55	1336.73
3	1496.21	1283.22	1334.08	1325.01	1335.05
4	1525.86	1274.94	1332.53	1320.49	1334.63
5	1551.74	1269.76	1330.98	1316.02	1333.63
6	1589.57	1266.1	1329.43	1311.57	1333.00
7	1628.42	1266.74	1327.88	1307.78	1332.41
8	1656.94	1268.6	1326.34	1302.78	1331.86
9	1691.54	1271	1324.79	1298.44	1331.36
10	1738.36	1283	1323.25	1294.12	1330.28

Table 9 — Theoretical values on based Ideal mixture relation (IMR), Nomoto's relation (NOM), Junjie's relation (JUN) and Impedance relation (IMP) for morpholine with *o*-xylene in isopropanol at 308K along with experimental velocity(U)

Concn $\times10^{-3}$M	U m/s	IMR	NOM	JUN	IMP
1	1419.26	1393.22	1395.32	1257.57	1409.39
2	1451.45	1368.14	1376.58	1192.47	1397.90
3	1491.63	1344.63	1361.86	1163.58	1385.43
4	1532.42	1322.56	1350.00	1151.79	1371.85
5	1575.25	1301.81	1340.23	1149.18	1356.98
6	1596.11	1282.28	1332.05	1151.87	1340.65
7	1639.25	1263.89	1325.10	1157.75	1322.63
8	1681.42	1246.56	1319.13	1165.60	1302.64
9	1726.72	1230.23	1313.93	1174.68	1280.34
10	1761.92	1214.84	1309.37	1184.49	1255.31

Table 10 — Theoretical values based on Ideal mixture relation (IMR), Nomoto's relation (NOM), Junjie's relation (JUN) and Impedance relation (IMP) for morpholine with *o*-xylene in isopropanol at 313K along with experimental velocity(U)

Concn $\times10^{-3}$M	U m/s	IMR	NOM	JUN	IMP
1	1391.6	1363.52	1369.65	1303.89	1377.50
2	1426.2	1336.87	1350.73	1250.52	1362.39
3	1455.9	1311.87	1334.51	1216.16	1346.62
4	1486.2	1288.40	1320.45	1193.30	1330.14
5	1509.4	1266.33	1308.14	1177.87	1312.92
6	1539.4	1245.56	1297.28	1167.47	1294.89
7	1581.6	1226.00	1287.62	1160.62	1276.00
8	1616.5	1207.56	1278.99	1156.31	1256.19
9	1642.8	1190.18	1271.21	1153.88	1235.39
10	1675.2	1173.80	1264.18	1152.85	1213.52

Table 11 — Percentage deviation of theoretical values of ultrasonic velocity from the values of experimental velocities for morpholine with *o*-xylene in isopropanol at 303K

Concn $\times10^{-3}$M	U_{exp} m/s	IMR	NOM	JUN	IMP
1	1431.65	8.26	6.60	6.81	6.56
2	1472.11	11.99	9.27	9.68	9.20
3	1496.21	14.24	10.84	11.44	10.77
4	1525.86	16.44	12.67	13.46	12.53
5	1551.74	18.17	14.23	15.19	14.06
6	1589.57	20.35	16.37	17.49	16.14
7	1628.42	22.21	18.46	19.69	18.18
8	1656.94	23.44	19.95	21.37	19.62
9	1691.54	24.86	21.68	23.24	21.29
10	1738.36	26.19	23.88	25.56	23.47

Table 12 — Percentage deviation of theoretical values of ultrasonic velocity from the values of experimental velocities for morpholine with *o*-xylene in isopropanol at 308K

Concn $\times 10^{-3}$M	U_{exp} m/s	IMR	NOM	JUN	IMP
1	1419.26	1.83	0.70	11.39	1.69
2	1451.45	5.74	3.69	17.84	5.16
3	1491.63	9.85	7.12	21.99	8.70
4	1532.42	13.69	10.48	24.84	11.90
5	1575.25	17.36	13.86	27.05	14.92
6	1596.11	19.66	16.01	27.83	16.54
7	1639.25	22.90	19.31	29.37	19.16
8	1681.42	25.86	22.53	30.68	21.55
9	1726.72	28.75	25.85	31.97	23.91
10	1761.92	31.05	28.75	32.77	25.69

Table 13 — Percentage deviation of theoretical values of ultrasonic velocity from the values of experimental velocities for morpholine with *o*-xylene in isopropanol at 313K

Concn $\times 10^{-3}$M	U_{exp} m/s	IMR	NOM	JUN	IMP
1	1391.6	2.02	1.58	6.30	1.01
2	1426.2	6.26	5.29	12.32	4.47
3	1455.9	9.89	8.34	16.47	7.51
4	1486.2	13.31	11.15	19.71	10.50
5	1509.4	16.10	13.33	21.96	13.02
6	1539.4	19.09	15.73	24.16	15.88
7	1581.6	22.48	18.59	26.62	19.32
8	1616.5	25.30	20.88	28.47	22.29
9	1642.8	27.55	22.62	29.76	24.80
10	1675.2	29.93	24.54	31.18	27.56

Table 14 — Molecular interaction parameter(α) $\chi_u/10^{-2}$ for morpholine with *o*-xylene in isopropanol in relation to experimental velocity at 303K, 308K and 313K

Concn	303K		308K		313K	
$\times 10^{-3}$M	U_{exp} m/s	α	U_{exp}	α	U_{exp}	α
1	1431.65	0.1882	1419.26	0.0377	1391.6	0.0416
2	1472.11	0.2911	1451.45	0.1255	1426.2	0.1381
3	1496.21	0.3595	1491.63	0.2306	1455.9	0.2316
4	1525.86	0.4324	1532.42	0.3425	1486.2	0.3306
5	1551.74	0.4935	1575.25	0.4642	1509.4	0.4207
6	1589.57	0.5762	1596.11	0.5494	1539.4	0.5275
7	1628.42	0.6526	1639.25	0.6822	1581.6	0.6642
8	1656.94	0.7059	1681.42	0.8194	1616.5	0.7920
9	1691.54	0.7712	1726.72	0.9700	1642.8	0.9052
10	1738.36	0.8358	1761.92	1.1035	1675.2	1.0368

Table 15 — Comparison between Absolute average percentage deviation and Values of Chi-square test

System	Temp.	AAPD				Values of Chi-square test			
		IMR	NOM	JUN	IMP	IMR	NOM	JUN	IMP
Morpholine+ *o*-Xylene+ Isopropyl alcohol	303K	18.615	15.395	16.393	15.182	78.76	53.81	62.40	52.00
	308K	17.669	14.922	25.573	14.830	89.91	59.51	159.41	65.89
	313K	17.193	14.205	21.695	14.636	79.66	51.10	114.12	59.24

Conclusion

In this present probe, the acoustical & thermodynamical parameters were calculated or computed based on the measured parameters like ultrasonic velocity, density and viscosity for the ternary system namely morpholine with *o*-xylene in isopropanol at 303K, 308K and 313K. The values of free energy of formation support for the thermodynamically stable nature of the complex. The K values at all the working temperatures under consideration as well as entropy values are evident for the charge transfer complex formation. The acoustical properties pave the way for close packing of the molecules and the presence of strong dipole-dipole interactions. Among all the four theories which under consideration for the present study, calculated theoretical values are compared with the measured values at 303K, 308K and 313K for the *ortho* xylene-morpholine system, Impedance relation is found to be suitable and show the minimum deviation at 305K and 308K, whereas at 313K, Nomoto's relation shows a good agreement with experimental values. It is also reflected by the values of Chi-square test. The molecular interaction parameter also proves the presence of moderate interaction between the solute and solvent.

Supplementary Information

Supplementary information is available in the website http://nopr.niscpr.res.in/handle/123456789/ 58776.

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