Ultrasonic study on binary liquid mixtures of propiophenone with anilines and alkyl substituted anilines at T=303.15 to 318.15 K

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Abstract−Densities 'ρ', ultrasonic speeds of sound 'u' of binary mixtures of propiophenone (PPH) with aniline, N-Methylaniline, N,N-dimethylaniline and N,N-diethylaniline were measured over the entire composition range from 303.15 K to 318.15 K and at atmospheric pressure 0.1 MPa. Experimental data of ultrasonic sound were compared and discussed with the computed values of 'u' from various velocity theories like Nomoto's relation (U_{NOM}), impedance relation (U_{IMP}), Van Dael and Vangeel's ideal mix relation (U_{VDV}), Rao's specific velocity relation (U_{RAO}), Junjie's theory (U_{JUN}) and Jouyban-Acree's (U_{JOE}) relation for the above binary mixtures over the entire mole fraction range at the studied temperatures. The results are satisfactory and are in agreement between the theoretical and the experimental values. Further, the molecular interaction parameter (α), average percentage error and Chi-square test values were computed by using the values of experimental and theoretical ultrasonic velocities. The Δu values were correlated with Redlich-Kister polynomial equation to compute the coefficients and the standard deviations of the binary mixtures. The results were analyzed in terms of intermolecular interactions.

Keywords: Ultrasonic Speed of Sound, Molecular Interaction Parameter, Nomato's Relation, VanDeel's Ideal Mixing Relation, Impedance Relation, Junjie's Relation, Rao's Specific Velocity and Jouyban-Acree's Relation for Speed of Sound

INTRODUCTION

Ultrasonic velocity, which is the best tool to measure the mechanical stability of liquids, gives information on the physico-chemical behavior and thermodynamic properties of the complexes of the binary mixtures [1-7]. Ultrasonic velocity and its theoretical evaluation in pure and liquid mixtures and its comparison with experimental data show the behavior of molecular interactions and used to develop theoretical models for liquids. Lageman and Dunbar (1945) initiated using the sound velocity approach for the qualitative estimation of the interactions in liquids. The combined study of ultrasonic velocity and density of solution gives more understanding on the behavior of ideal and non-ideal nature, elastic properties and other co-related parameters of the liquid mixtures. A number of mathematical theories on speed of sound measurements give positive implications to industry and chemical processes. The study on interactions between aromatic and aliphatic ketones with aniline and substituted anilines has been the subject of many research works which are still on.

Literature survey reveals that thermo-acoustical studies were done on binary mixtures of oxalane with anilines and substituted anilines [8], diethylcarbonate with aniline [9], isomeric chlorophenols with N,N-dimethyl aniline [10], IBMK with aniline and N,N-dimethyl aniline [11], N,N-diethyl aniline with aliphatic ketones [12], N-

methyl aniline with methyl isobutyl ketone, 3-pentanone and cyclo alkanones [13], diisopropyl ether or oxolane with N,N-dimethyl aniline or N,N-diethyl aniline [14], etc.

Apparently no study has been reported on comparing the computed values of ultrasonic velocity from various velocity theories such as Nomato's relation [15], Van Dael and Vangeel ideal mixing relation [16,17], Junjie's relation [18], impedance dependence relation, Rao's specific velocity relation [19,20] and Jouyban-Acree's [21] relation for speed of sound with the experimental values of ultrasonic velocity of sound of the Propiophenone with aniline, N-methylaniline, N,N-dimethylaniline and N,N-diethylaniline binary mixtures at temperatures 303.15 K to 318.15 K and to understand the behavior of intermolecular interactions between the molecules. Hence, this study has such importance and it is useful for the chemical and industrial processes.

Propiophenone with aniline, N-methylaniline, N,N-dimethylaniline and N,N-diethylaniline mixtures are having various applications in chemical industries. Propiophenone is used as a component in some perfumes and intermediate in the synthesis of pharmaceutical and other organic compounds [22]. It is used in the synthesis of ketoamphetamines such as cathinone and methcathinone. It can also be converted to synthetic aryl alkenes such as cinnamic acids.

Aromatic anilines and substituted alkyl anilines are used as intermediates to manufacture dyes and vanillin and as a stabilizer for calorimetric peroxidase determination. Aniline has been used as parent substance in the manufacture of several chemical products and intermediates, in the production of isocyanides, rubber chemicals, dyes and pigments, hydroquinone, drugs and agriculture. It is

also used as a solvent and has been used as an antiknock compound for gasoline [23].

In the present paper, the experimental values of densities and ultrasonic velocities of binary liquid mixtures of Propiophenone (PPH) with aniline, N-methylaniline, N,N-dimethylaniline and N,N-diethylaniline were calculated at T=303.15 to 318.15 K. The experimental data of ultrasonic velocities were compared with the values of velocity theories; further, the deviation in molecular interaction parameter (α) values to understand the non-ideal behavior of the liquid mixtures, relative percentage error and Chi-square test values for the goodness of fit were computed. The Δu, values were correlated with Redlich-Kister polynomial equation to compute the coefficients and the standard deviations of the studied binary mixtures.

EXPERIMENTAL SECTION

1. Materials

Propiophenone (PPH), aniline (A), N-methylaniline (MA), N,N-dimethylaniline (DMA) and N,N-diethylaniline (DEA) were purchased from S.D. Fine chemicals Ltd., India and used in the present investigation. Purities of the components are tabulated in Table 1.

Estimated purities of the liquid samples were greater than 99% and water content was found less than 0.003 mass%. Mettler Toledo

Table 1. Provenance and purity of the materials used

Chemicals	CAS number	Source	Mass fraction purity (final)
Propiophenone	93-55-0	S.D fine Chemicals, India	>99%
Aniline	62-53-3	S.D fine Chemicals, India	0.997
N-methylaniline	100-61-8	S.D fine Chemicals, India	0.995
NN-diethylaniline	91-66-7	S.D fine Chemicals, India	0.996
NN-dimethylaniline	121-69-7	S.D fine Chemicals, India	0.995

Table 2. Experimental Vs literature values of density and velocity of pure liquids

Compound	T (K)	Density (ρ)		Ref.	Velocity		Ref.
		Experimental	Literature		Experimental	Literature	
		gm/cm^{-3}	gm/cm^{-3}		m·s^{-1}	m·s^{-1}	
Propiophenone	303.15	1.00450	1.00437	j	1446.41	1440, 1458	i
	308.15	1.00150	1.00600	j	1427.04	1432, 1438	i
	313.15	0.99850			1403.95		
	318.15	0.99550			1380.00		
Aniline	303.15	1.01280	1.01305	a	1615.18	1619.20	a
			1.01284	b		1614.50	c
			1.01280	f		1615.20	f
			1.01350	k		1615.20	k
			1.01290	k		1619.30	m
			1.01300	l		1614.50	m
			1.01290	l			
			1.01280	m			
			1.01390	n			
			1.01310	n			
			1.01318	q		1614.00	q
			1.01317	q		1615.00	q
	308.15	1.00890	1.00870	l	1601.88	1602.95	p
			1.00860	l			
			1.00867	p			
	313.15	1.00490	1.00490	f	1588.29	1582.60	k
			1.00436	k		1580.05	k
			1.00463	k			
			1.00450	k			
			1.00490	k			
			1.00440	l			
			1.00420	l			
	318.15	1.00090			1574.41		

Table 2. Experimental Vs literature values of density and velocity of pure liquids

| Compound | T (K) | Density (ρ) | | | Velocity | | |
| | | Experimental | Literature | Ref. | Experimental | Literature | Ref. |
		gm/cm^{-3}	gm/cm^{-3}		m·s^{-1}	m·s^{-1}	
N-methylaniline	303.15	0.97830	0.97801	a	1545.61	1545.50	a
			0.98172	d		1551.00	d
			0.98170	e			
			0.97820	f		1548.30	f
			0.97828	m		1546.00	m
			0.97823	p		1546.90	p
	308.15	0.97400	0.97424	p	1528.59	1528.15	p
	313.15	0.96960	0.96980	f	1511.63	1512.40	f
			0.96990	f			
			0.96960	f			
	318.15	0.96520			1494.38		
N,N-dimethylaniline	303.15	0.94790	0.94770	l	1468.00	1468.00	o
			0.94800	l			
			0.94767	l			
			0.94840	o			
			0.94800	o			
			0.94833	o			
	308.15	0.94360	0.95173	g	1449.74		
			0.95177	h			
			0.94355	l			
	313.15	0.93930	0.93940	l	1431.27	1431.00	o
			0.93850	l			
			0.93970	o			
	318.15	0.93510			1412.74		
N,N-diethylaniline	303.15	0.92596	0.92600	l	1411.49	1411	a
			0.92529	a		1434	o
			0.92590	k			
			0.92600	k			
			0.92610	o			
	308.15	0.92174	0.92126	a	1404.48	1406	a
			0.92190	k			
			0.92126	k			
	313.15	0.91759	0.91770	l	1397.54	1396	o
			0.91800	l			
			0.91750	o			
	318.15	0.91344			1390.60		

(ME204) balance with the precision of ±0.1 mg was used to prepare the mixtures by mixing weighed amounts of the pure liquids adopting the method of closed system. To avoid air bubbles, mixtures were allowed to stand for some time before every measurement. Average of three repetitions was considered while measuring and recording the values. The purities of the liquids were checked by comparing the values of densities and viscosities with literature data and are given in Table 2, a [32], b [33], c [34], d [35], e [36], f [37], g [38], h [39], i [40], j [41], k [42], l [43], m [44], n [45], o [46], p [47], q [48].

2. Apparatus and Procedure

Anton Paar (DSA 5000 M) oscillating u-tube densimeter was used to calculate the densities and speed of sounds of the pure components and the binary mixtures over the whole composition range with a precision of 0-3 gcm^{-3} in the measurement of densities and 1,000 to 2,000 ms^{-1} in the measurement speed of sound, and it can be operated at temperatures from 273.15 K to 343.15 K with a pressure variation from 0 to 0.3 Mpa at low frequency. Calibration was done at the required temperature using dry air and ultrapure water (millipore). Averages of three consecutive measurements were taken between the temperatures 303.15 K to 318.15 K with an increment of 5 K under atmospheric pressure.

The experimental values of ultrasonic velocity and density of binary mixtures at temperatures from 303.15 to 318.15 K with re-

Table 3. Mole fractions (x₁), densities (ρ), ultrasonic velocities (U) and deviation in velocity (ΔU) values of binary mixtures propiophenone with anilines, N-methylaniline, N,N-dimethylaniline and N,N-diethylaniline at T=303.15 to 318.15 K

X1	303.15 K			308.15 K			313.15 K			318.15 K		
	ρ	U	ΔU	ρ	U	ΔU	ρ	U	ΔU	ρ	U	ΔU
	g cm^{-3}	m·s^{-1}	m·s^{-1}	g cm^{-3}	m·s^{-1}	m·s^{-1}	g cm^{-3}	m·s^{-1}	m·s^{-1}	g cm^{-3}	m·s^{-1}	m·s^{-1}
PPH+aniline												
0	1.0128	1615.18	0.0000	1.0089	1601.88	0.0000	1.0049	1588.29	0.0000	1.0009	1574.41	0.0000
0.0708	1.0122	1603.28	0.2507	1.0083	1589.55	0.2007	1.0044	1575.28	0.1507	1.0005	1560.67	0.1007
0.1463	1.0115	1590.58	0.4746	1.0078	1576.38	0.4246	1.0040	1561.38	0.3746	1.0001	1546.02	0.3246
0.2270	1.0109	1576.98	0.6776	1.0072	1562.28	0.6276	1.0035	1546.52	0.5776	0.9997	1530.34	0.5276
0.3136	1.0101	1562.39	0.8176	1.0065	1547.17	0.7676	1.0029	1530.58	0.7176	0.9992	1513.53	0.6676
0.4066	1.0093	1546.71	0.8810	1.0059	1530.91	0.8310	1.0023	1513.43	0.7810	0.9987	1495.45	0.7310
0.5069	1.0085	1529.78	0.8691	1.0051	1513.38	0.8191	1.0016	1494.95	0.7691	0.9982	1475.96	0.7191
0.6152	1.0076	1511.48	0.7655	1.0043	1494.42	0.7155	1.0009	1474.97	0.6655	0.9976	1454.88	0.6155
0.7327	1.0066	1491.62	0.5566	1.0034	1473.86	0.5066	1.0002	1453.29	0.4567	0.9970	1432.02	0.4067
0.8605	1.0056	1470.00	0.2928	1.0025	1451.47	0.2428	0.9994	1429.70	0.1928	0.9963	1407.15	0.1428
1	1.0045	1446.41	0.0000	1.0015	1427.04	0.0000	0.9985	1403.95	0.0000	0.9955	1380.00	0.0000
PPH+N-methylaniline												
0	0.9783	1545.61	0.0000	0.9740	1528.59	0.0000	0.9696	1511.63	0.0000	0.9652	1494.38	0.0000
0.0939	0.9816	1536.65	0.4331	0.9775	1519.34	0.3831	0.9733	1501.70	0.3331	0.9691	1483.78	0.3031
0.1891	0.9848	1527.55	0.7374	0.9809	1509.97	0.6874	0.9768	1491.65	0.6374	0.9727	1473.06	0.6074
0.2856	0.9878	1518.24	0.9242	0.9840	1500.42	0.8742	0.9801	1481.48	0.8242	0.9762	1462.21	0.7942
0.3834	0.9907	1508.72	1.0003	0.9870	1490.69	0.9503	0.9832	1471.12	0.9003	0.9794	1451.18	0.8703
0.4826	0.9933	1498.98	1.0000	0.9898	1480.73	0.9500	0.9862	1460.54	0.9000	0.9825	1439.92	0.8700
0.5831	0.9958	1489.01	0.9249	0.9924	1470.53	0.8749	0.9889	1449.70	0.8249	0.9855	1428.42	0.7949
0.6852	0.9981	1478.81	0.7796	0.9949	1460.06	0.7296	0.9915	1438.60	0.6796	0.9882	1416.64	0.6496
0.7886	1.0003	1468.34	0.5820	0.9972	1449.33	0.5320	0.9940	1427.24	0.4820	0.9908	1404.61	0.4519
0.8935	1.0024	1457.55	0.3156	0.9994	1438.31	0.2656	0.9963	1415.65	0.2156	0.9933	1392.36	0.1856
1	1.0045	1446.41	0.0000	1.0015	1427.04	0.0000	0.9985	1403.95	0.0000	0.9955	1380.00	0.0000
PPH+NN-dimethylaniline												
0	0.9479	1468.00	0.0000	0.9436	1449.74	0.0000	0.9393	1431.27	0.0000	0.9351	1412.74	0.0000
0.0958	0.9538	1466.28	0.5347	0.9498	1447.82	0.4547	0.9459	1428.87	0.3847	0.9419	1409.80	0.3347
0.1925	0.9599	1464.54	0.8516	0.9560	1445.94	0.7716	0.9522	1426.52	0.7016	0.9485	1406.88	0.6516
0.2900	0.9659	1462.69	1.0359	0.9621	1443.99	0.9559	0.9584	1424.09	0.8859	0.9548	1403.89	0.8359
0.3886	0.9717	1460.71	1.1096	0.9681	1441.91	1.0296	0.9645	1421.54	0.9596	0.9609	1400.80	0.9096
0.4880	0.9774	1458.56	1.1014	0.9739	1439.66	1.0214	0.9704	1418.83	0.9514	0.9670	1397.56	0.9014
0.5885	0.9830	1456.27	1.0199	0.9796	1437.26	0.9399	0.9763	1415.99	0.8699	0.9730	1394.18	0.8199
0.6899	0.9884	1453.86	0.8721	0.9851	1434.74	0.7921	0.9820	1413.02	0.7221	0.9788	1390.69	0.6721
0.7922	0.9937	1451.39	0.6637	0.9906	1432.16	0.5837	0.9876	1409.99	0.5137	0.9846	1387.13	0.4637
0.8956	0.9990	1448.88	0.3619	0.9960	1429.57	0.2819	0.9931	1406.94	0.2119	0.9902	1383.55	0.1619
1	1.0045	1446.41	0.0000	1.0015	1427.04	0.0000	0.9985	1403.95	0.0000	0.9955	1380.00	0.0000
PPH+NN-diethylaniline												
0.0000	0.9260	1411.49	0.0000	0.9217	1404.48	0.0000	0.9176	1397.54	0.0000	0.9134	1390.60	0.0000
0.1173	0.9340	1416.31	0.7228	0.9300	1407.80	0.6728	0.9260	1398.91	0.6228	0.9220	1389.93	0.5728
0.2302	0.9421	1420.61	1.0888	0.9382	1410.71	1.0388	0.9344	1400.00	0.9888	0.9305	1389.10	0.9388
0.3389	0.9502	1424.57	1.2519	0.9464	1413.33	1.2019	0.9427	1400.86	1.1519	0.9389	1388.11	1.1019
0.4437	0.9582	1428.29	1.3115	0.9545	1415.75	1.2615	0.9509	1401.59	1.2115	0.9473	1387.06	1.1616
0.5447	0.9661	1431.79	1.2830	0.9626	1418.00	1.2330	0.9591	1402.21	1.1830	0.9555	1385.96	1.1330
0.6422	0.9739	1435.08	1.1698	0.9705	1420.09	1.1198	0.9671	1402.73	1.0698	0.9637	1384.81	1.0198
0.7363	0.9816	1438.17	0.9749	0.9783	1422.01	0.9249	0.9751	1403.13	0.8749	0.9718	1383.62	0.8249
0.8272	0.9892	1441.08	0.7114	0.9861	1423.80	0.6614	0.9830	1403.45	0.6114	0.9798	1382.39	0.5614
0.9150	0.9968	1443.82	0.3733	0.9938	1425.44	0.3233	0.9908	1403.68	0.2733	0.9877	1381.12	0.2233
1.0000	1.0045	1446.41	0.0000	1.0015	1427.04	0.0000	0.9985	1403.95	0.0000	0.9955	1380.00	0.0000

spective mole fractions (x_1) are reported in Table 3.

3. Theory

The values of ultrasonic velocities of the binary mixtures propiophenone with aniline, N-methylaniline, N,N-dimethylaniline and N,N-diethylaniline at temperatures from 303.15 k to 318.15 k were calculated by using the velocity models, such as Nomoto's relation, Van Dael and Vangeel's relation, impedance relation, Junjie's relation, Rao's specific velocity relations and Jouyban-Acree's relation for speed of sound. Relations are as shown below.

3-1. Nomoto's Relation

Nomoto's empirical formula for the sound velocity U is given by

$$U_{Nom} = \left[\frac{x_1 R_1 + x_2 R_2}{x_1 V_1 + x_2 V_2}\right]^3 \tag{1}$$

where $R_1 = (M_1 u_1^{1/3})/\rho_1$; $R_2 = (M_2 u_2^{1/3})/\rho_2$; $V_1 = M_1/\rho_1$; $V_2 = M_2/\rho_2$

In above equations, x_1 and x_2 are the mole fraction of component liquids. M_1, M_2, u_1, u_2, V_1 and V_2 are the molecular weights, ultrasonic velocity and molar volumes of components of binary liquids mixtures.

3-2. Van Dael and Vangeel's Relation

$$U_{VDV} = \left[\left(\frac{x_1}{M_1 U_1^2} + \frac{x_2}{M_2 U_2^2}\right)(x_1 M_1 + x_2 M_2)\right]^{-1/2} \tag{2}$$

where M_1, M_2 and U_1, U_2 are molecular weights and ultrasonic sound velocities of the compounds 1 and 2, respectively.

3-3. Impedance Relation

$$U_{IMP} = [\Sigma x_i Z_i / \Sigma x_i \rho_i] \tag{3}$$

where X_i is the mole fraction, ρ_i the density of the mixture and Z_i is the acoustic impedance.

3-4. Rao's Specific Velocity Relation

Using the ratio of the temperature coefficient of velocity and expansion coefficient, Rao derived a formula for ultrasonic velocity (U)

$$U_{RAO} = (\Sigma(x_i r_i \rho_i))^3 \tag{4}$$
$$r_i = (u_i^{1/3})/\rho_i$$

where 'r' is Rao's constant or molar sound velocity, which is constant for a liquid at a temperature.

3-5. Junjie's Relation

This relation was derived by Junjie for the ultrasonic velocity of the mixture in terms of the mole fraction, molecular weight and density of the mixture.

$$U_{JUN} = \frac{\Sigma_{i=1}^n x_i V_i}{(\Sigma_{i=1}^n x_i M_i)^{1/2} * (\Sigma_{i=1}^n x_i V_i / \rho_i U_i^2)^{1/2}} \tag{5}$$

where the symbols have their usual meaning

3-6. Jouyban-acree Equation for Velocity

$$\ln u = x_1 \ln u_1 + x_2 \ln u_2 + A_0\left[\frac{x_1 x_2}{T}\right] + A_1\left[\frac{x_1 x_2 (x_1 - x_2)}{T}\right] + A_2\left[\frac{x_1 x_2 (x_1 - x_2)^2}{T}\right] \tag{6}$$

where u is the ultrasonic velocity of the binary liquid mixture and, x_1, u_1, x_2, u_2 are the mole fractions and velocities of component 1 and component 2, respectively. A_0, A_1, and A_2 are the adjustable inter-

action parameters.

3-7. Chi-square Test for Goodness of Fit

The Chi-square value is evaluated for the binary liquid mixtures under study using the formula

$$x^2 = \Sigma_{i=1}^n ((U_{obs} - U_{cal})^2 / U_{cal}) \tag{7}$$

where 'n' is the number of data used,
and U(obs)=experimental values of ultrasonic velocities
U(cal)=computed values of ultrasonic velocities

3-8. Relative Percentage Error

$$\sigma = \left(\frac{1}{n}\right) * \Sigma((U_{obs} - U_{cal})/U_{obs}) * 100\% \tag{8}$$

where 'n' is the number of data used,
and U(obs)=experimental values of ultrasonic velocities
U(cal)=computed values of ultrasonic velocities

3-9. The Molecular Interaction Parameter

$$\propto = \left(\left(\frac{U_{obs}}{U_{cal}}\right)^2 - 1\right) \tag{9}$$

U(obs)=experimental values of ultrasonic velocities
U(cal)=computed values of ultrasonic velocities

RESULTS AND DISCUSSION

The various theoretical computed ultrasonic velocities of propiophenone with anilines, N-methylaniline, N,N-dimethylaniline and N,N-diethylaniline at temperatures 303.15 K to 318.15 K were compared with the experimental values of ultrasonic velocities and presented in Table 4(a)-4(b).

Theoretical ultrasonic velocity studies play a major role when there is no possibility to calculate acoustical and excess acoustic parameters. Due to the presence of dispersion forces, charge transfer, hydrogen bonding, dipole-dipole and dipole-induced dipole interactions, intermolecular interactions take place in the binary mixtures [24], and it happens between the unlike/like molecules in the liquid mixtures [25].

Calculated values of ultrasonic velocities by using various velocity model relations and experimental values of propiophenone with aniline, N-methylaniline, N,N-dimethylaniline and N,N-diethylaniline mixtures over the entire composition range at temperature 303.15 K are represented in Fig. 1(a)-1(d). Deviation in velocity of PPH+A, MA, DMA and DEA binary mixtures at T=303.15 to 318.15 K is shown in Fig. 2(a)-2(d).

The relative percentage errors for all the mathematical velocity models and Chi square values of the binary mixtures at temperatures from 303.15 to 318.15 K are tabulated in Table 5.

From Fig. 3(a)-3(d), it is clear that the relative percentage deviation values are higher and positive for the U_{JUN}, U_{VDV} and U_{NOM} comparatively with other three theories' values. Relative percentage error values are positive in all the six velocity model relations except in Jouyban-Acree relation for PPH+aniline mixture where it is negative at all temperatures. The percentage deviation of velocity values of U_{IMP}, U_{JOU} and U_{RAO}'s was satisfactory and in agreement with the experimental values. Relative percentage error values of U_{JOU} are best suited and they are ranging from −0.0001

Table 4. (a) 'U' values of respective velocity theories for the binary mixtures PPH+aniline & PPH+N-methylaniline at T=303.15 to 318.15 K

	PPH+aniline							PPH+N-MA						
(X1)	Uexp ms^{-1}	Unom ms^{-1}	Uvdv ms^{-1}	Uimp ms^{-1}	Ujun ms^{-1}	Urao ms^{-1}	Ujou ms^{-1}	(X1)	Uexp ms^{-1}	Unom ms^{-1}	Uvdv ms^{-1}	Uimp ms^{-1}	Ujun ms^{-1}	Urao ms^{-1}	Ujou ms^{-1}
T=303.15 K								**T=303.15 K**						
0.0000	1615.18	1615.18	1615.18	1615.18	1615.18	1615.18	1615.18	0.0000	1545.61	1545.61	1545.61	1545.61	1545.61	1545.61	1545.61
0.0708	1603.49	1597.81	1598.19	1603.33	1595.53	1602.83	1603.50	0.0939	1536.73	1534.26	1533.99	1536.07	1533.68	1536.11	1536.67
0.1463	1590.97	1580.56	1581.14	1590.67	1576.59	1589.72	1590.99	0.1891	1527.59	1523.27	1522.76	1526.45	1522.27	1526.52	1527.53
0.2270	1577.55	1563.41	1564.04	1577.11	1558.33	1575.78	1577.55	0.2856	1518.21	1512.62	1511.93	1516.74	1511.36	1516.83	1518.18
0.3136	1563.08	1546.37	1546.91	1562.55	1540.70	1560.92	1563.07	0.3834	1508.58	1502.29	1501.48	1506.96	1500.91	1507.06	1508.59
0.4066	1547.44	1529.44	1529.80	1546.89	1523.67	1545.06	1547.44	0.4826	1498.74	1492.28	1491.40	1497.08	1490.89	1497.19	1498.78
0.5069	1530.50	1512.62	1512.75	1529.98	1507.20	1528.08	1530.50	0.5831	1488.69	1482.56	1481.69	1487.13	1481.28	1487.23	1488.72
0.6152	1512.11	1495.90	1495.81	1511.68	1491.27	1509.88	1512.11	0.6852	1478.42	1473.12	1472.35	1477.08	1472.04	1477.17	1478.44
0.7327	1492.08	1479.30	1479.04	1491.80	1475.84	1490.31	1492.09	0.7886	1467.96	1463.96	1463.35	1466.95	1463.17	1467.02	1467.94
0.8605	1470.25	1462.80	1462.53	1470.12	1460.90	1469.22	1470.25	0.8935	1457.29	1455.06	1454.71	1456.72	1454.63	1456.76	1457.26
1.0000	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41	1.0000	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41
T=308.15 K								**T=308.15 K**						
0.0000	1601.88	1601.88	1601.88	1601.88	1601.88	1601.88	1601.88	0.0000	1528.59	1528.59	1528.59	1528.59	1528.59	1528.59	1528.59
0.0708	1589.71	1583.88	1584.52	1589.59	1581.41	1589.07	1589.76	0.0939	1519.44	1516.98	1516.78	1518.81	1516.37	1518.86	1519.42
0.1463	1576.73	1565.99	1567.06	1576.47	1561.72	1575.46	1576.77	0.1891	1510.08	1505.73	1505.36	1508.96	1504.68	1509.03	1510.04
0.2270	1562.82	1548.21	1549.51	1562.41	1542.75	1561.00	1562.83	0.2856	1500.47	1494.83	1494.32	1499.01	1493.51	1499.12	1500.44
0.3136	1547.82	1530.56	1531.90	1547.33	1524.46	1545.60	1547.82	0.3834	1490.61	1484.26	1483.65	1488.99	1482.81	1489.11	1490.62
0.4066	1531.62	1513.01	1514.26	1531.10	1506.81	1529.16	1531.61	0.4826	1480.54	1474.01	1473.35	1478.88	1472.55	1479.00	1480.56
0.5069	1514.08	1495.59	1496.61	1513.58	1489.77	1511.58	1514.06	0.5831	1470.25	1464.06	1463.40	1468.69	1462.71	1468.81	1470.27
0.6152	1495.03	1478.28	1478.99	1494.62	1473.31	1492.72	1495.01	0.6852	1459.74	1454.40	1453.80	1458.41	1453.26	1458.51	1459.75
0.7327	1474.28	1461.08	1461.48	1474.03	1457.38	1472.46	1474.28	0.7886	1449.04	1445.02	1444.54	1448.04	1444.18	1448.12	1449.02
0.8605	1451.68	1444.00	1444.13	1451.59	1441.97	1450.63	1451.69	0.8935	1438.11	1435.90	1435.62	1437.58	1435.44	1437.63	1438.10
1.0000	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04	1.0000	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04
T=313.15 K								**T=313.15 K**						
0.0000	1588.29	1588.29	1588.29	1588.29	1588.29	1588.29	1588.29	0.0000	1511.63	1511.63	1511.63	1511.63	1511.63	1511.63	1511.63
0.0708	1575.40	1569.28	1570.31	1575.32	1566.52	1574.75	1575.43	0.0939	1501.86	1499.32	1499.28	1501.25	1498.61	1501.30	1501.86
0.1463	1561.70	1550.40	1552.17	1561.48	1545.62	1560.38	1561.70	0.1891	1491.91	1487.39	1487.30	1490.79	1486.19	1490.87	1491.89
0.2270	1547.02	1531.64	1533.91	1546.65	1525.54	1545.11	1547.00	0.2856	1481.71	1475.83	1475.69	1480.23	1474.31	1480.34	1481.69
0.3136	1531.20	1513.02	1515.51	1530.74	1506.22	1528.85	1531.19	0.3834	1471.25	1464.62	1464.44	1469.60	1462.96	1469.72	1471.26
0.4066	1514.12	1494.52	1497.01	1513.62	1487.61	1511.50	1514.12	0.4826	1460.57	1453.75	1453.54	1458.88	1452.08	1459.01	1460.59
0.5069	1495.62	1476.15	1478.42	1495.15	1469.68	1492.96	1495.63	0.5831	1449.66	1443.20	1442.97	1448.07	1441.66	1448.19	1449.67
0.6152	1475.55	1457.91	1459.78	1475.16	1452.39	1473.09	1475.55	0.6852	1438.53	1432.96	1432.74	1437.18	1431.65	1437.28	1438.53
0.7327	1453.68	1439.80	1441.12	1453.46	1435.69	1451.75	1453.68	0.7886	1427.20	1423.01	1422.82	1426.19	1422.05	1426.27	1427.18
0.8605	1429.86	1421.81	1422.48	1429.81	1419.55	1428.76	1429.86	0.8935	1415.63	1413.34	1413.23	1415.12	1412.83	1415.16	1415.64
1.0000	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95	1.0000	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95
T=318.15 K								**T=318.15 K**						
0.0000	1574.41	1574.41	1574.41	1574.41	1574.41	1574.41	1574.41	0.0000	1494.38	1494.38	1494.38	1494.38	1494.38	1494.38	1494.38
0.0708	1560.75	1554.33	1555.74	1560.72	1551.23	1560.09	1560.80	0.0939	1483.94	1481.30	1481.42	1483.34	1480.48	1483.38	1483.97
0.1463	1546.30	1534.39	1536.88	1546.11	1529.04	1544.90	1546.30	0.1891	1473.36	1468.62	1468.83	1472.21	1467.23	1472.29	1473.35
0.2270	1530.80	1514.59	1517.83	1530.46	1507.76	1528.77	1530.78	0.2856	1462.51	1456.34	1456.59	1460.99	1454.59	1461.10	1462.50
0.3136	1514.11	1494.94	1498.59	1513.67	1487.34	1511.60	1514.10	0.3834	1451.40	1444.44	1444.69	1449.69	1442.51	1449.81	1451.41
0.4066	1496.09	1475.43	1479.18	1495.61	1467.72	1493.30	1496.10	0.4826	1440.05	1432.89	1433.13	1438.30	1430.96	1438.43	1440.06
0.5069	1476.59	1456.06	1459.59	1476.13	1448.85	1473.73	1476.60	0.5831	1428.47	1421.68	1421.89	1426.82	1419.90	1426.94	1428.48
0.6152	1455.42	1436.83	1439.86	1455.05	1430.69	1452.79	1455.42	0.6852	1416.66	1410.80	1410.96	1415.25	1409.31	1415.36	1416.66
0.7327	1432.37	1417.75	1419.99	1432.17	1413.18	1430.30	1432.37	0.7886	1404.63	1400.24	1400.34	1403.59	1399.14	1403.67	1404.61
0.8605	1407.27	1398.80	1400.03	1407.25	1396.30	1406.11	1407.27	0.8935	1392.36	1389.98	1390.03	1391.84	1389.38	1391.89	1392.38
1.0000	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00	1.0000	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00

Table 4. (b) 'U' values of respective velocity theories for the binary mixtures PPH+N,N-dimethylaniline & PPH+N,N-diethylaniline at T= 303.15 to 318.15 K

			PPH+N,N-DMA							PPH+N,N-DEA				
(X1)	Uexp ms⁻¹	Unom ms⁻¹	Uvdv ms⁻¹	Uimp ms⁻¹	Ujun ms⁻¹	Urao ms⁻¹	Ujou ms⁻¹	(X1)	Uexp ms⁻¹	Unom ms⁻¹	Uvdv ms⁻¹	Uimp ms⁻¹	Ujun ms⁻¹	Urao ms⁻¹	Ujou ms⁻¹

(Header units use ms^{-1}.)

T=303.15 K (DMA) / **T=303.15 K** (DEA)

(X1)	Uexp	Unom	Uvdv	Uimp	Ujun	Urao	Ujou	(X1)	Uexp	Unom	Uvdv	Uimp	Ujun	Urao	Ujou
0.0000	1468.00	1468.00	1468.00	1468.00	1468.00	1468.00	1468.00	0.0000	1411.49	1411.49	1411.49	1411.49	1411.49	1411.49	1411.49
0.0958	1466.47	1465.84	1465.42	1465.82	1465.70	1465.92	1466.35	0.1173	1416.31	1414.93	1415.01	1415.89	1414.18	1415.55	1416.19
0.1925	1464.70	1463.68	1462.93	1463.65	1463.42	1463.83	1464.61	0.2302	1420.61	1418.38	1418.54	1420.04	1417.04	1419.48	1420.57
0.2900	1462.77	1461.52	1460.53	1461.48	1461.19	1461.72	1462.74	0.3389	1424.57	1421.84	1422.07	1423.97	1420.07	1423.26	1424.65
0.3886	1460.72	1459.36	1458.22	1459.31	1458.98	1459.59	1460.75	0.4437	1428.29	1425.32	1425.59	1427.69	1423.28	1426.91	1428.43
0.4880	1458.56	1457.20	1456.01	1457.15	1456.81	1457.44	1458.63	0.5447	1431.79	1428.80	1429.11	1431.21	1426.66	1430.44	1431.94
0.5885	1456.32	1455.04	1453.89	1454.99	1454.67	1455.27	1456.37	0.6422	1435.08	1432.30	1432.60	1434.56	1430.23	1433.85	1435.22
0.6899	1453.98	1452.88	1451.87	1452.84	1452.55	1453.08	1454.00	0.7363	1438.17	1435.81	1436.09	1437.74	1433.98	1437.14	1438.28
0.7922	1451.56	1450.72	1449.95	1450.69	1450.48	1450.88	1451.52	0.8272	1441.08	1439.33	1439.55	1440.77	1437.92	1440.33	1441.15
0.8956	1449.03	1448.57	1448.13	1448.55	1448.43	1448.65	1448.98	0.9150	1443.82	1442.87	1442.99	1443.66	1442.07	1443.42	1443.85
1.0000	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41	1.0000	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41

T=308.15 K (DMA) / **T=308.15 K** (DEA)

(X1)	Uexp	Unom	Uvdv	Uimp	Ujun	Urao	Ujou	(X1)	Uexp	Unom	Uvdv	Uimp	Ujun	Urao	Ujou
0.0000	1449.74	1449.74	1449.74	1449.74	1449.74	1449.74	1449.74	0.0000	1404.48	1404.48	1404.48	1404.48	1404.48	1404.48	1404.48
0.0958	1448.02	1447.47	1447.06	1447.44	1447.32	1447.55	1447.77	0.1173	1407.80	1406.70	1406.50	1407.33	1406.08	1407.11	1407.69
0.1925	1446.14	1445.20	1444.47	1445.15	1444.94	1445.35	1445.72	0.2302	1410.71	1408.94	1408.60	1410.01	1407.81	1409.65	1410.64
0.2900	1444.11	1442.93	1441.97	1442.87	1442.59	1443.13	1443.62	0.3389	1413.33	1411.17	1410.76	1412.55	1409.69	1412.10	1413.34
0.3886	1441.95	1440.66	1439.56	1440.59	1440.27	1440.89	1441.52	0.4437	1415.75	1413.42	1412.98	1414.95	1411.71	1414.46	1415.80
0.4880	1439.68	1438.39	1437.24	1438.32	1437.98	1438.63	1439.41	0.5447	1418.00	1415.67	1415.24	1417.23	1413.88	1416.74	1418.05
0.5885	1437.32	1436.12	1435.01	1436.05	1435.73	1436.35	1437.28	0.6422	1420.09	1417.93	1417.55	1419.39	1416.20	1418.94	1420.10
0.6899	1434.87	1433.85	1432.87	1433.79	1433.51	1434.05	1435.08	0.7363	1422.01	1420.20	1419.88	1421.44	1418.67	1421.06	1422.00
0.7922	1432.34	1431.58	1430.83	1431.54	1431.32	1431.73	1432.73	0.8272	1423.80	1422.47	1422.25	1423.40	1421.30	1423.12	1423.77
0.8956	1429.69	1429.31	1428.89	1429.28	1429.16	1429.40	1430.10	0.9150	1425.44	1424.75	1424.63	1425.26	1424.09	1425.11	1425.44
1.0000	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04	1.0000	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04

T=313.15 K (DMA) / **T=313.15 K** (DEA)

(X1)	Uexp	Unom	Uvdv	Uimp	Ujun	Urao	Ujou	(X1)	Uexp	Unom	Uvdv	Uimp	Ujun	Urao	Ujou
0.0000	1431.27	1431.27	1431.27	1431.27	1431.27	1431.27	1431.27	0.0000	1397.54	1397.54	1397.54	1397.54	1397.54	1397.54	1397.54
0.0958	1429.04	1428.54	1428.18	1428.50	1428.39	1428.64	1428.84	0.1173	1398.91	1398.17	1397.54	1398.35	1397.68	1398.29	1398.83
0.1925	1426.71	1425.81	1425.16	1425.75	1425.55	1425.98	1426.32	0.2302	1400.00	1398.81	1397.73	1399.11	1397.92	1399.01	1399.95
0.2900	1424.23	1423.08	1422.23	1423.00	1422.74	1423.31	1423.76	0.3389	1400.86	1399.44	1398.09	1399.83	1398.28	1399.71	1400.88
0.3886	1421.61	1420.35	1419.37	1420.25	1419.96	1420.61	1421.19	0.4437	1401.59	1400.08	1398.59	1400.52	1398.75	1400.38	1401.64
0.4880	1418.89	1417.61	1416.60	1417.52	1417.22	1417.89	1418.62	0.5447	1402.21	1400.72	1399.23	1401.16	1399.33	1401.03	1402.25
0.5885	1416.06	1414.88	1413.90	1414.79	1414.50	1415.15	1416.02	0.6422	1402.73	1401.37	1399.98	1401.78	1400.02	1401.65	1402.73
0.6899	1413.14	1412.15	1411.29	1412.07	1411.82	1412.39	1413.36	0.7363	1403.13	1402.01	1400.84	1402.36	1400.83	1402.26	1403.11
0.7922	1410.14	1409.42	1408.76	1409.36	1409.17	1409.60	1410.54	0.8272	1403.45	1402.65	1401.80	1402.92	1401.75	1402.84	1403.43
0.8956	1407.01	1406.68	1406.31	1406.65	1406.54	1406.79	1407.45	0.9150	1403.68	1403.30	1402.84	1403.45	1402.79	1403.40	1403.69
1.0000	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95	1.0000	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95

T=318.15 K (DMA) / **T=318.15 K** (DEA)

| (X1)	Uexp	Unom	Uvdv	Uimp	Ujun	Urao	Ujou	(X1)	Uexp	Unom	Uvdv	Uimp	Ujun	Urao	Ujou
0.0000	1412.74	1412.74	1412.74	1412.74	1412.74	1412.74	1412.74	0.0000	1390.60	1390.60	1390.60	1390.60	1390.60	1390.60	1390.60
0.0958	1409.94	1409.47	1409.15	1409.42	1409.32	1409.58	1409.77	0.1173	1389.93	1389.55	1388.41	1389.26	1389.16	1389.35	1389.88
0.1925	1407.09	1406.20	1405.64	1406.11	1405.94	1406.40	1406.72	0.2302	1389.10	1388.50	1386.54	1387.99	1387.81	1388.15	1389.05
0.2900	1404.08	1402.92	1402.19	1402.82	1402.59	1403.19	1403.62	0.3389	1388.11	1387.44	1384.97	1386.80	1386.54	1387.00	1388.12
0.3886	1400.93	1399.65	1398.81	1399.53	1399.27	1399.96	1400.51	0.4437	1387.06	1386.38	1383.65	1385.67	1385.35	1385.89	1387.09
0.4880	1397.66	1396.38	1395.50	1396.25	1395.98	1396.70	1397.39	0.5447	1385.96	1385.33	1382.58	1384.60	1384.25	1384.82	1385.98
0.5885	1394.29	1393.10	1392.26	1392.98	1392.72	1393.41	1394.24	0.6422	1384.81	1384.27	1381.72	1383.59	1383.23	1383.79	1384.81
0.6899	1390.83	1389.83	1389.09	1389.72	1389.50	1390.10	1391.03	0.7363	1383.62	1383.20	1381.04	1382.62	1382.30	1382.79	1383.59
0.7922	1387.27	1386.55	1385.99	1386.47	1386.30	1386.76	1387.66	0.8272	1382.39	1382.14	1380.54	1381.70	1381.45	1381.83	1382.37
0.8956	1383.58	1383.28	1382.96	1383.23	1383.14	1383.39	1384.03	0.9150	1381.12	1381.07	1380.20	1380.83	1380.68	1380.90	1381.16
1.0000	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00	1.0000	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00
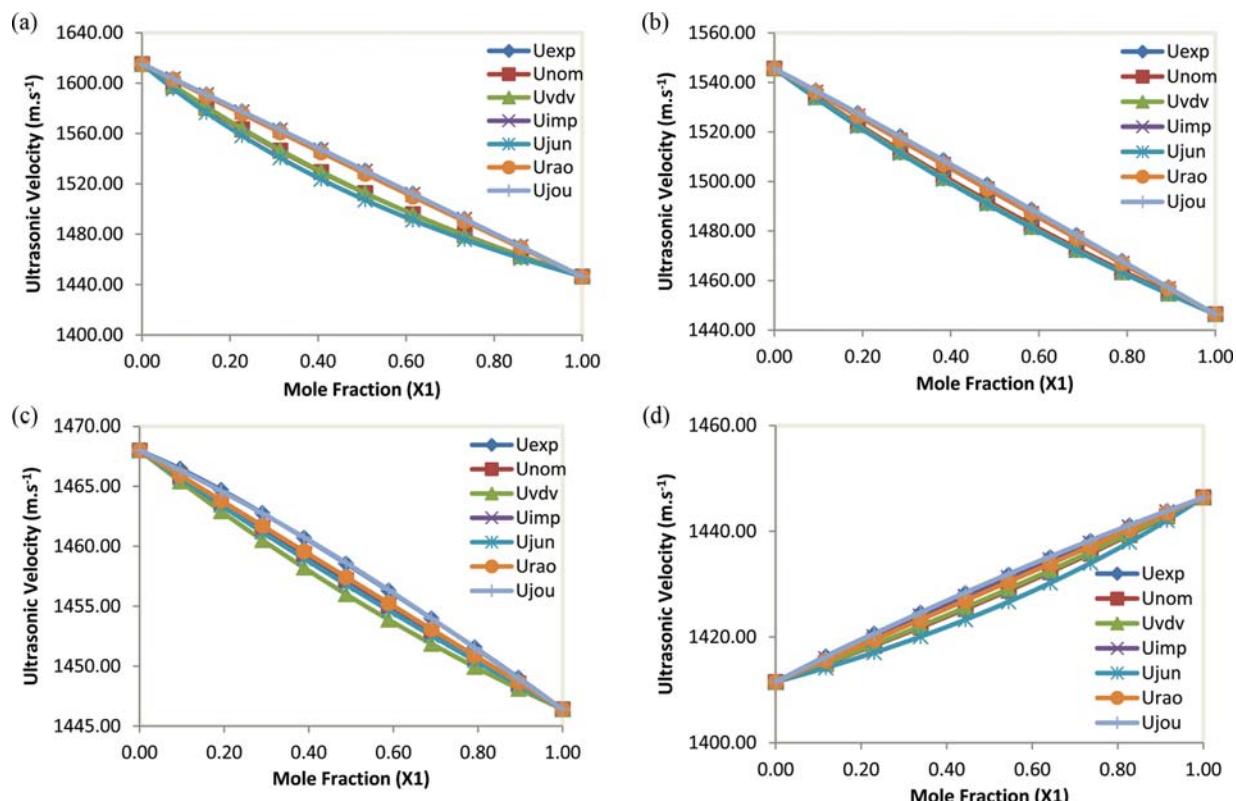
Fig. 1. (a) Experimental Vs theoretical speed of sound values for PPH+aniline at T=303.15 K. (b) Experimental Vs theoretical speed of sound values for PPH+methylaniline at T=303.15 K. (c) Experimental Vs theoretical speed of sound values for PPH+N,N-dimethylaniline at T=303.15 K. (d) Experimental Vs theoretical speed of sound values for PPH+N,N-diethylaniline at T=303.15 K.
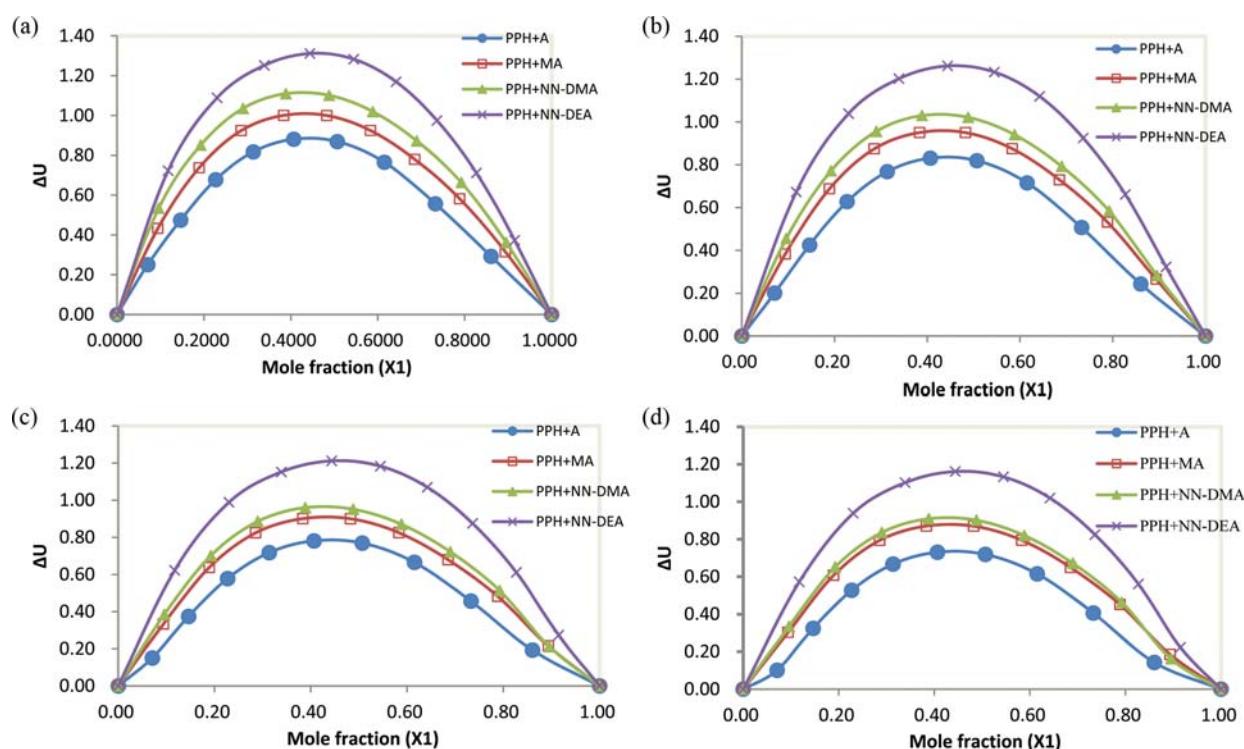

Fig. 2. (a) Deviation in ultrasonic velocity values for all four binary mixtures at T=303.15 K. (b) Deviation in ultrasonic velocity values for all four binary mixtures at T=308.15 K. (c) Deviation in ultrasonic velocity values for all four binary mixtures at T=313.15 K. (d) Deviation in ultrasonic velocity values for all four binary mixtures at T=318.15 K.
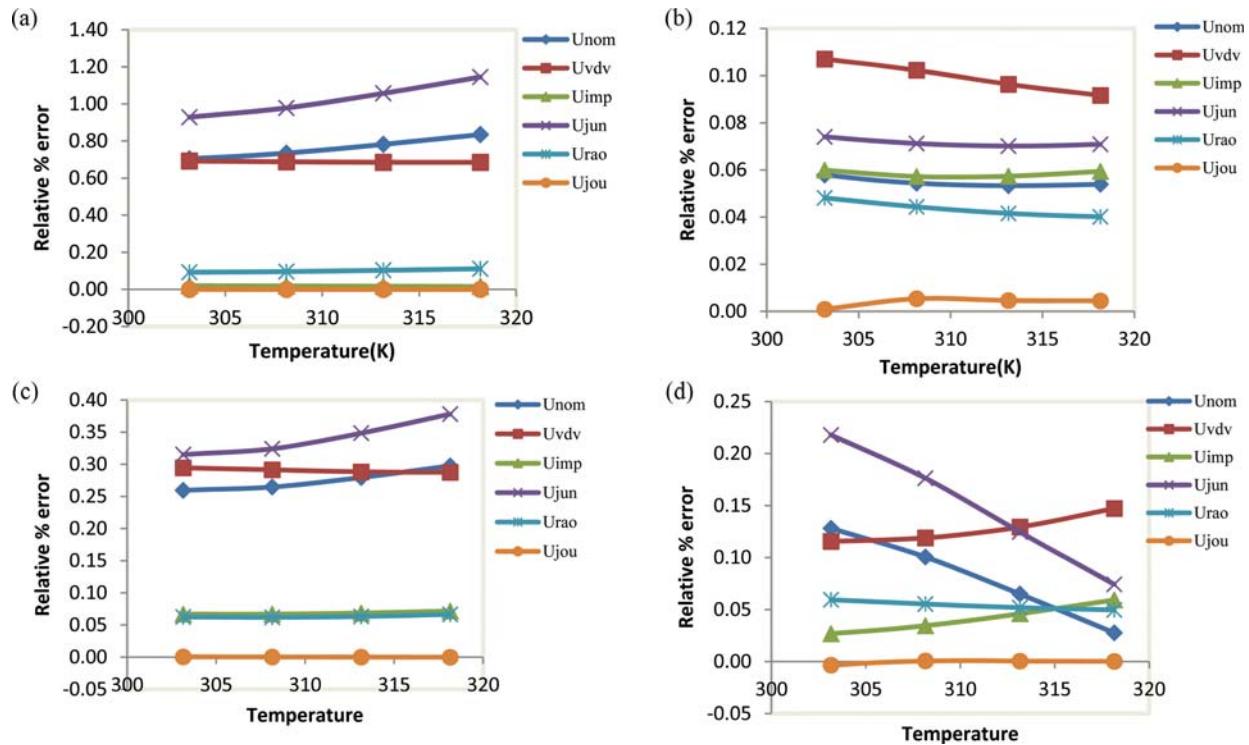
Fig. 3. (a) Relative percentage error values of velocity theories for PPH+aniline at T=303.15-318.15 K. (b) Relative percentage error values of velocity theories for PPH+methylaniline at T=303.15-318.15 K. (c) Relative percentage error values of velocity theories for PPH+N,N-dimethylaniline at T=303.15-318.15 K. (d) Relative percentage error values of velocity theories for PPH+N,N-diethylaniline at T=303.15-318.15 K.

Table 5. Relative percentage error and Chi square values of binary mixtures propiophenone with anilines, N-methylaniline, N,N-dimethylaniline and N,N-diethylaniline at T=303.15 to 318.15 K

| Temp	Relative % error						Chi square					
(K)	Unom	Uvdv	Uimp	Ujun	Urao	Ujou	Unom	Uvdv	Uimp	Ujun	Urao	Ujou
PPH+aniline												
303.15	0.7038	0.6924	0.0197	0.9283	0.0926	−0.0001	1.1447	1.1095	0.0009	1.9938	0.0199	0.0000
308.15	0.7344	0.6881	0.0181	0.9783	0.0960	−0.0004	1.2350	1.0850	0.0008	2.1937	0.0213	0.0000
313.15	0.7822	0.6850	0.0167	1.0570	0.1028	−0.0001	1.3859	1.0633	0.0007	2.5341	0.0242	0.0000
318.15	0.8344	0.6847	0.0154	1.1448	0.1109	−0.0002	1.5598	1.0503	0.0006	2.9408	0.0279	0.0000
PPH+N-methylaniline												
303.15	0.2595	0.2945	0.0668	0.3151	0.0626	0.0005	0.1499	0.1930	0.0099	0.2213	0.0087	0.0000
308.15	0.2646	0.2914	0.0670	0.3241	0.0619	0.0003	0.1543	0.1870	0.0099	0.2315	0.0085	0.0000
313.15	0.2794	0.2883	0.0686	0.3484	0.0633	0.0001	0.1700	0.1809	0.0103	0.2645	0.0088	0.0000
318.15	0.2975	0.2876	0.0718	0.3781	0.0665	−0.0001	0.1902	0.1777	0.0112	0.3074	0.0096	0.0000
PPH+N,N-dimethylaniline												
303.15	0.0579	0.1070	0.0599	0.0741	0.0482	0.0009	0.0072	0.0246	0.0077	0.0118	0.0050	0.0000
308.15	0.0544	0.1023	0.0572	0.0712	0.0444	0.0054	0.0063	0.0224	0.0070	0.0109	0.0042	0.0008
313.15	0.0533	0.0963	0.0574	0.0701	0.0416	0.0046	0.0061	0.0197	0.0070	0.0105	0.0037	0.0008
318.15	0.0539	0.0916	0.0594	0.0709	0.0401	0.0045	0.0062	0.0177	0.0075	0.0106	0.0035	0.0007
PPH+N,N-diethylaniline												
303.15	0.1281	0.1155	0.0268	0.2179	0.0595	−0.0034	0.0346	0.0282	0.0015	0.1004	0.0075	0.0001
308.15	0.1006	0.1189	0.0344	0.1763	0.0554	0.0007	0.0212	0.0297	0.0025	0.0652	0.0065	0.0000
313.15	0.0650	0.1294	0.0460	0.1246	0.0519	0.0004	0.0089	0.0350	0.0045	0.0323	0.0057	0.0000
318.15	0.0277	0.1472	0.0592	0.0743	0.0497	0.0002	0.0017	0.0450	0.0074	0.0115	0.0052	0.0000
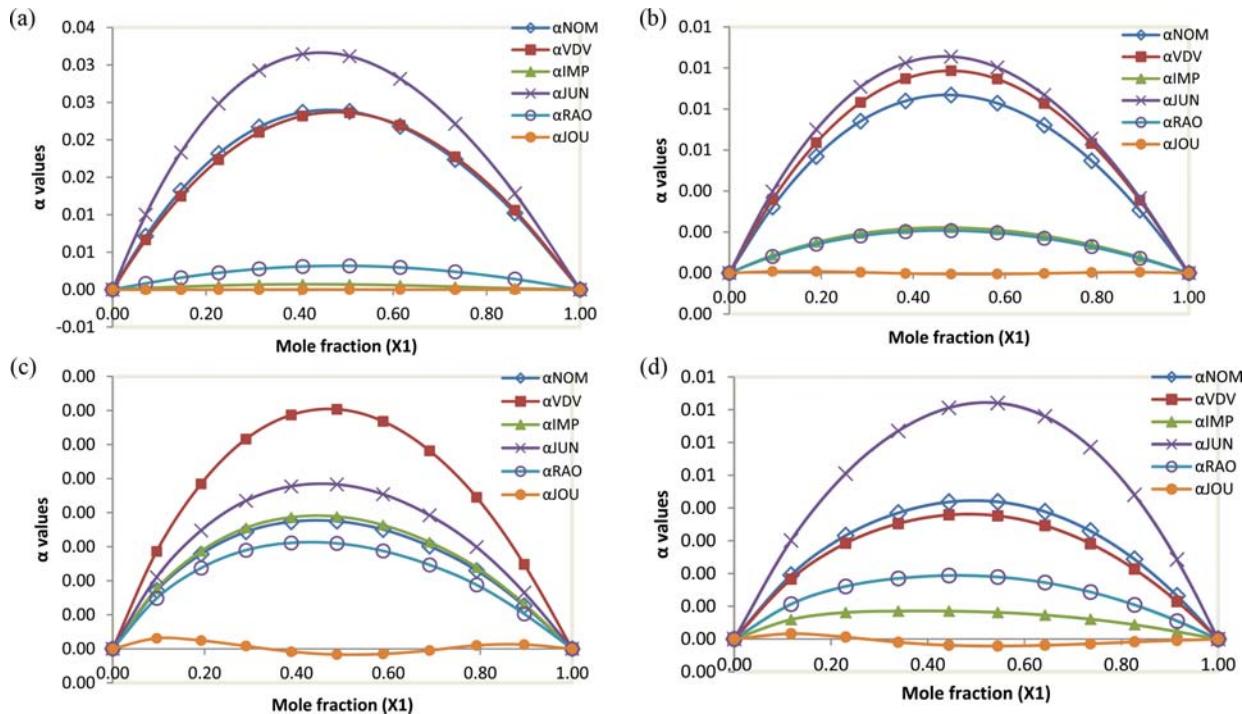
Fig. 4. (a) Molecular interaction parameter values (α) for PPH+aniline at T=303.15 K. (b) Molecular interaction parameter values (α) for PPH+methylaniline at T=303.15 K. (c) Molecular interaction parameter values (α) for PPH+N,N-dimethylaniline at T=303.15 K. (d) Molecular interaction parameter values (α) for PPH+N,N-diethylaniline at T=303.15 K.

to −0.0004 for PPH+A, −0.0001 to 0.0005 for PPH+MA, 0.0009 to 0.0054 for PPH+DMA and −0.0034 to 0.0007 for PPH+DEA.

The U values of Jouyban-Acree, impedance and Rao's specific velocity are closer to experimental values. The algebraic order of average relative percentage error is $U_{JOU}<U_{IMP}<U_{RAO}<U_{VDV}<U_{NOM}<U_{JUN}$. According to Nomoto's theory, volume does not change upon mixing. No interaction takes place when the ratios of specific heats of ideal mixtures and volumes are assumed to be equal. But interactions occur between the molecules by the presence of dispersion forces, charge transfer, hydrogen bonding, dipole-dipole and dipole-induced dipole interactions. The positive deviations indicate the increasing strength of interaction between component molecules of a binary liquid mixture [26,27], while the negative Δu may be due to the structure breaking, resulting in expansion and the speeds of sound through the mixture will be slower [28]. Deviation in ultrasonic velocity values of the studied binary mixtures is tabulated in Table 3.

Deviation in velocity values for all the four binary mixtures is positive, and these values are decreasing with increase in temperature. It indicates that the molecular interactions took place between the unlike molecules in the binary mixtures. The positive deviations in velocity are a result of (a) molecular association and (b) complex formation, whereas negative deviations in velocity are due to molecular dissociation of associated species as a result of addition of inner solvent or an active solvent [29]. Deviation in velocity (Δu) values is maximum at mole fractions 0.4066, 0.3834, 0.3886, and 0.4437 is 0.8810, 1.003, 1.1096 & 1.3115 for PPH+A, PPH+MA, PPH+DMA & PPH+DEA at temperature 303.15 K, respectively. The algebraic order of Δu is PPH+A<PPH+MA<PPH+

DMA<PPH+DEA.

Fig. 4(a)-4(d) shows the molecular interaction parameters (α) values of the four binary mixtures at 303.15 K. Positive values of 'α' are observed in all four binary systems. It indicates the trend of forming good association through strong dipole-dipole/hydrogen bonding interactions [30]. Negative value of 'α' indicates the dominance of dispersion forces arising from the breakage of hydrogen bonds in the associates [24]. The algebraic order of 'α' values is $\alpha_{JOU}<\alpha_{IMP}<\alpha_{RAO}<\alpha_{VDV}<\alpha_{NOM}<\alpha_{JUN}$. Chi-square values are positive for all the studied binary mixtures over the entire composition at temperatures T=303.15 to 318.15 K. Chi-square values are ranging from 0.0000 to 2.9408 for PPH+A, 0.000 to 0.3074 for PPH+MA, 0.000 to 0.0118 for PPH+DMA and 0.000 to 0.1004 for PPH+DEA at T=303.15-318.15 K. There is no significant difference between the experimental and the calculated values, then the Chi square values are closer to zero; therefore, those binary mixtures are in agreement with goodness of fit (Null hypothesis); this type of trend is observed in PPH+MA, DMA and DEA. But in the binary mixture of PPH+A, the Chi-square values are higher than the other three binary mixtures, hence it comes under alternative hypothesis. U_{JOU}, U_{IMP} and U_{RAO}'s Chi-square values are comparatively more satisfactory than the other three models in all the studied binary mixtures. Overall, Jouyban-Acree's relation Chi-square values are in best agreement with the experimental values.

Deviation of ultrasonic velocity values (Δu) was fitted to Redlich-Kister polynomial equation and the coefficients and standard deviations were computed for all the studied binary mixtures.

The values of deviations in velocity (Δu) with respect to the mole fraction at temperatures from 303.15 K to 318.15 K are fitted to the

Table 6. Redlich-Kister polynomial equation & Jouyban-Acree relation's coefficients and standard deviation values for PPH+A, PPH+MA, PPH+DMA & PPH+DEA binary systems at T=303.15 to 318.15 K

Property	Temp/K	A0	A1	A3	A4	σ	A0	A1	A2	σ
PPH+aniline		Δu; Redlich-kister equation coefficients					Jouyban-Acree relation coefficients;			
	303.15	3.4943	−1.0732	0.1869	0.0001	0.0042	2.5314	−0.1023	1.0531	0.00001
	308.15	3.2874	−1.0906	0.2606	−0.8526	0.0042	2.7114	−0.1077	1.0522	0.00002
	313.15	3.0804	−1.1081	0.3345	−1.7054	0.0057	3.0371	−0.0772	4.4855	0.00001
	318.15	2.8733	−1.1256	0.4083	−2.5581	0.0079	3.3967	−0.0613	7.3887	0.00001
PPH+aniline										
	303.15	3.96184	−1.08554	−0.02678	−0.28583	0.00229	1.5015	−0.1444	0.9529	0.00003
	308.15	3.75714	−1.08962	−0.01014	−1.05592	0.00301	1.5368	−0.1624	0.9412	0.00002
	313.15	3.55244	−1.09371	0.00651	−1.82596	0.00405	1.636	−0.1782	0.933	0.00001
	318.15	3.42962	−1.09615	0.0165	−2.28798	0.00474	1.7808	−0.1871	0.934	0.00001
PPH+NN-dimethylaniline										
	303.15	4.3775	−1.0443	−0.5204	0.0352	0.0041	0.996	−0.1886	0.8796	0.00005
	308.15	4.05	−1.0488	−0.5021	−1.1947	0.0045	0.7009	0.6779	0.9241	0.0003
	313.15	3.7635	−1.0528	−0.486	−2.2707	0.0055	0.6773	0.6831	0.9175	0.0003
	318.15	3.5589	−1.0556	−0.4746	−3.0394	0.0064	0.674	0.676	0.913	0.0003
PPH+NN-diethylaniline										
	303.15	5.2178	−0.6876	−1.1854	0.0609	0.0028	1.3261	−0.0948	0.8998	0.00009
	308.15	5.0126	−0.6776	−1.2268	−0.7244	0.0041	1.1792	−0.1879	0.832	0.00004
	313.15	4.8075	−0.6675	−1.2681	−1.5099	0.0056	1.1202	−0.205	0.8218	0.00003
	318.15	4.6023	−0.6574	−1.3095	−2.2954	0.0072	1.1013	−0.2234	0.8083	0.00003

Redlich-Kister Equation [31] of the type:

$$Y^E = x_1x_2\{a0+a1(x_1-x_2)+a2(x_1-x_2)^2+\cdots+a4(x_1-x_2)^4\} \qquad (10)$$

where Y^E is for Δu in the polynomial degree. The values of a0, a1, a2, a3, and a4 are the coefficients of the polynomial equation, and were obtained by the method of least-squares and are given in Table 6 along with standard deviation values. The standard deviations are calculated by using the equation:

$$\sigma(Y^E) = \left\{\frac{\Sigma(Y^E_{obs}-Y^E_{cal})^2}{n-m}\right\}^{1/2} \qquad (11)$$

where n is the total number of experimental points and m is the number of coefficients.

Jouyban-Acree's speed of sound relation interaction parameters and standard deviation values are tabulated and presented in Table 6. Interaction parameter values A_0 and A_2 are positive for all the studied binary mixtures, whereas A_1 values are negative for PPH+ A, PPH+MA and PPH+DEA and mixed results (negative at 303.15 K and positive for T=308.15 to 318.15 K) are observed for PPH+ DMA. Jouyban-Acree speed of sound relation values are in good agreement with the experimental values for all the four binary mixtures.

CONCLUSIONS

We evaluated theoretical ultrasonic velocities in binary liquid mixture containing propiophenone with anilines, N-methylaniline, N,N-dimethylaniline and N,N-diethylaniline by using Nomoto (U_{NOM}), impedance (U_{IMP}), Van-Dael and Vangeel (U_{VDV}), Junjie (U_{JUN}), Rao's specific velocity (U_{RAO}) and Jouyban-Acree's (U_{JOU})

relation for speed of sound models, and compared them with the experimental data of ultrasonic velocity values at T=303.15 to 318.15 K. As per the relative percentage error values, Jouyban-Acree relation, impedance and Rao's specific velocity relations are best suited with the experimental values in all the binary systems. Jouyban-Acree's relation for speed of sound appears to have an edge over the other relations in comparison. Overall, the goodness of fit (Chi-square values) is in good agreement for the models U_{JOU}, U_{IMP} and U_{RAO} comparatively to other three models in all the four binary mixtures. Positive values of interaction parameter in the present systems at all the temperatures clearly indicate the existence of molecular association and complex formation. Deviations in velocity (Δu) values were correlated with the Redlich-Kister polynomial equation and the interaction coefficients and standard deviations of the binary mixtures were computed.

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