Sound Velocities in Aqueous Solutions of n-Butylamine between 303.15 and 323.15 K: Experiment and Theory

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Abstract

Density (ρ) and sound velocity (u) have been measured for aqueous solutions of n-butylamine (W + NBA) in the whole range of composition at an interval of 5 K. Deviation in Sound Velocity (Δu), Isentropic Compressibility (K_s), Excess Isentropic Compressibility (K_s^E), Specific Acoustic Impedance (Z), Rao's Constant (R) and Wada's Constant (W) have been calculated from measured u and ρ. Δu versus x_2 curve is negative and with the increment of temperature, Δu decreases; K_s values of W + NBA are all positive whereas K_s^E values are all negative. All the above outcomes are interpreted in terms of molecular interaction especially hydrogen bonding and hydrophobic hydration between water and n-butylamine. Moreover, experimental data for u correlated with some theoretical equations. The relations are Nomoto's Relation (u_N), Impedance Relation (u_{IR}), Rao's specific velocity method relation (u_R), Van Deel's ideal mixing relation (u_{IMR}) and theoretical sound velocity according to Free Length Theory (u_{FLT}). The validity of these relations with experimental values has been tested by measuring standard percentage deviation ($\sigma\%$) and average percentage error (APE).

Keywords: *Sound velocity, Isentropic compressibility, Hydrogen bonding, Hydrophobic hydration, Theoretical relations.*

1. Introduction

Sound velocity provides valuable information about the physicochemical behavior of binary liquid mixtures. In 1945, Langemann and Dunbar showed qualitatively that the molecular association in liquids depends upon sound velocity [1]. Being related to the binding forces between the components of the medium, it is connected with the structure and molecular interactions existing in the liquid systems [2,3]. The derived properties such as adiabatic compressibility, excess isentropic compressibility, etc. provide a better understanding of the molecular scenario of liquids [4–8]. The sound velocity of the liquid mixture has significant applications in various industrial technological processes as it is useful to detect and evaluate molecular interactions [9–11].

The interaction between water and amines is quite complex. Both amines and water (W) are self-associated liquids through hydrogen bonding. Water forms trimer and larger aggregates of molecules by a cooperative interaction [12] and amine such as *n*-butylamine (NBA) are self-

associated to form non-cyclic trimer [13]. NBA possesses a hydrophilic amino (-NH$_2$) group as well as a hydrophobic alkyl group. The nature of the interaction of these two groups toward water is completely different. The hydrophilic amino (-NH$_2$) group interacts with water through H-bonding and disrupts normal water structure, while the alkyl group promotes the structure of water molecules through the hydrophobic hydration. It is believed that both effects take place in water-rich region.

Earlier, we reported excess molar volumes, viscosities, and refractive index of aqueous solutions of isomeric butylamines [14–16]. In the present communication, we are reporting some results related to sound velocities of W+NBA system in terms of experimental and theoretical approaches along with various acoustic and thermodynamic parameters. Moreover, to the best of our knowledge, they are scanty literature on this system. This investigation may serve as background work in formulating models for more complicated aqueous systems like surfactants, emulsions and biopolymers.

For the theoretical computation of sound velocity in liquid and liquid mixtures, several relations, semi-empirical formulas and theories are available in the literature. Among them, Nomoto's Relation (u_N), Impedance Relation (u_{IR}), Rao's specific velocity method relation (u_R), Van Deel's ideal mixing relation (u_{IMR}) and Free Length Theory (u_{FLT}) are utilized to calculate sound velocities and then compared with experimental values [9,17–23].

2. Experimental section

2.1 Materials

n-butylamine (NBA) (purity = 99.5 %, mol. Wt. = 73.14 and B.P. = 78 ^{0}C) was procured from Sigma-Aldrich Chemical Co. Ltd and thrice distilled water was used for the preparation of an aqueous solution of *n*-butylamine. NBA was used without further treatment except keeping under molecular sieves (4$\overset{.}{A}$) for 2-3 weeks prior to use.

2.2. Apparatus

The aqueous solutions at different compositions were made by volume using a pipette with an accuracy of ±0.01 cm^3. Densities and sound velocities were measured by using the Anton Paar density meter (DSA 5000 M) with precision up to ±10^{-6} g.cm^{-3} and 0.1 m/s, respectively. The instrument was adjusted and calibrated with air and bi-distilled water at 293.15K maintaining the temperature constant up to ±0.01 K (density of air =0.001177 g.cm^{-3} and density of water = 0.998205 g.cm^{-3}).

2.3. Preparation of solutions

In order to measure sound velocity and density, binary solutions of water and NBA at different compositions were prepared by the volumetric method. Special caution was taken to prevent evaporation and also the introduction of moisture into the experimental samples.

The deviation in sound velocity, Δu, is derived according to the following equation :

$$\Delta u = u - (x_1 u_1 + x_2 u_2) \tag{1}$$

Where, x_i and u_i are mole fraction and sound velocity of ith component, respectively.

The isentropic compressibility, K_s, is computed directly from the measured values of speed of sound and density using the Newton-Laplace equation:

$$K_s = -\frac{1}{V_m}\left(\frac{\delta V_m}{\delta P}\right)_s = \left(\frac{1}{\rho u^2}\right) = \left(\frac{V_m}{Mu^2}\right) \tag{2}$$

Excess isentropic compressibility, K_s^E, is given by:

$$K_s^E = K_s - (\emptyset_1 K_{s1} + \emptyset_2 K_{s2})$$ (3)

In the above equation, K_{Si} and $\emptyset_i$ are isentropic compressibility and volume fraction of ith component, respectively. Where, $\emptyset_i = \frac{x_i V_i}{\sum x_i V_i}$.

Specific acoustic impedance, Z, is determined from the equations:
$$Z = u\rho$$ (4)

Rao's constant and Wada's constant is also a measure of interaction existing in the solution.

Rao's constant, R, is calculated by using the following equation:
$$R = u^{\frac{1}{3}} V = u^{\frac{1}{3}} \frac{M}{\rho}$$ (5)

Wada's constant, Wa, is calculated by the following equation:
$$Wa = \overline{M} K_s^{-1/7} \Big/ \rho$$ (6)

In order to correlate measured and some derived properties, the following general polynomial equations have been used:

$$Y = \sum_{i=0}^{n} a_i x_2^i$$ (7)

Here, Y represents density, ρ, u, K_s, Z and a_i is the fitting coefficient and x_2 is the mole fraction.

The excess or deviation parameters have been correlated by the Redlich-Kister polynomial equation of the form:

$$Z^E = x_2(1 - x_2) \sum_{i=0}^{n} A_i (2x_2 - 1)^i$$ (8)

with the standard deviation, σ, following

$$(Z^E) = \left[\sum \frac{(z_{exp}^E - z_{cal}^E)}{n - p} \right]^{1/2}$$ (9)

Here, Z^E represents Δu and K_s^E; Z_{exp}^E and Z_{cal}^E are the experimental and calculated values of Z^E respectively and A_i is the i-th fitting coefficient, n the number of measurements, p the number of coefficients and the other terms have their usual significance.

2.4. Correlative models for sound velocity, u

2.4.1. Nomoto's Relation (u_{NR})

On assuming the additivity of molar sound velocity (R) and no volume change on mixing, Nomoto established the following relation for the ultrasonic velocity of binary liquid mixtures :

$$u_{NR} = \left[\frac{(x_1 R_1 + x_2 R_2)}{(x_1 V_1 + x_2 V_2)} \right]^3 \tag{10}$$

2.4.2. The Impedance Relation (u_{IR})

Impedance is the product of ultrasonic velocity (u) and the density (ρ) of a liquid mixture. Hence, the impedance relation predicts the ultrasonic velocity of the given mixture by simply using the values of impedance (Z_i) and the density (ρ) values. Impedance relation is given as

$$u_{IR} = \frac{\sum x_i Z_i}{\sum x_i \rho_i} \tag{11}$$

Where x_i is mole fraction , ρ_i is the density of the mixture and Z_i is the acoustic impedance.

2.4.3. The Rao's Specific Velocity Method Relation (u_R)

Rao's specific velocity method is given as

$$u_R = \left(\sum x_i r_i \rho \right)^3 \tag{12}$$

Where x_i is the mole fraction, u_i is the sound velocity and ρ_i is the density of the mixture. R_i is the Rao's specific sound velocity, which is given by $r_i = u_i^{\frac{1}{3}}/\rho_i$ and Z_i is the acoustic impedance.

2.4.4. Ideal Mixing Relation based on Van Deal and Vangeel Theory

Van Deal and Vangeel proposed the ideal mixing theory in the light of assumptions made by Blandamer and Wadding, yield the following relation for adiabatic compressibility $(\beta_{ad})_{imix}$

$$(\beta_{ad})_{imix} = \emptyset_1 \frac{\gamma_1}{\gamma_{imix}} (\beta_{ad})_1 + \emptyset_2 \frac{\gamma_2}{\gamma_{imix}} (\beta_{ad})_2 \tag{13}$$

Where $\emptyset_1$ and $\emptyset_2$ are the volume fractions of the liquids 1 and 2, γ_1 and γ_2 are the ratios of specific heats of the respective liquids. This relation holds good if the mixture is ideal and if $\gamma_1 = \gamma_2 = \gamma_{imix}$. Using the additional assumption that $V_1 = V_2$ the above equation can be transformed into a linear combination of mole fractions x_1 and x_2,

$$(\beta_{ad})_{imix} = x_1 (\beta_{ad})_1 + x_2 (\beta_{ad})_2 \tag{14}$$

On the basis of this equation, Van Deal and Vangeel obtained the relation for ultrasonic velocity in liquid mixtures as

$$u_{IMR} = \left[\frac{1}{x_1 M_1 + x_2 M_2} \right]^{\frac{1}{2}} \left[\frac{x_1}{M_1 u_1^2} + \frac{x_2}{M_2 u_2^2} \right]^{-\frac{1}{2}} \tag{15}$$

Where u_1 and u_2 are the ultrasonic velocities of the pure liquid components.

2.4.5. Relation based on Free Length Theory

Jacobson deduced an empirical relation for ultrasonic velocity (u_{FLT}) making use of intermolecular free length (L_f) and density (ρ) as

$$u_{FLT} = [K/ L_{fmix.}\rho_{exp}^{\frac{1}{2}}] \tag{16}$$

Where K is temperature-dependent called Jacobson's constant and the value of K at the working temperatures of the experiment were calculated (MKS units) and they are given below.

Temperature (K)	303.15	308.15	313.15	318.15	323.15
Value of K	2.075×10^{-6}	2.094×10^{-6}	2.113×10^{-6}	2.132×10^{-6}	2.151×10^{-6}

The Standard Percentage Deviation; $\sigma\%$ for all mole fractions:

$$\sigma\% = \left| \left[\frac{\sum\left\{ \frac{100(u_{exp}-u_{cal})}{u_{exp}} \right\}^2}{n} \right]^{\frac{1}{2}} \right| \tag{17}$$

Where n represents a number of data points [7].

2.4.6. The average percentage error (APE)

APE was calculated for the predicted refractive index data by the following equation

$$APE = \frac{1}{n}\sum\left[\frac{(u_{mix})_{exp}-(u_{mix})_{cal}}{(u_{mix})_{cal}} \right] \times 100 \tag{18}$$

3. Results and discussion

Density (ρ) and sound velocity (u) were measured for the binary solutions of W + NBA in the whole range of composition at an interval of 5 K. Deviation in Sound Velocity (Δu), isentropic compressibility (K_s), excess isentropic compressibility (K_s^E), specific acoustic impedance (Z), Rao's constant (R) and Wada's constant (W) were calculated from measured density (ρ) and sound velocity (u). Experimental data for u have been correlated with some theoretical equations. The relations are Nomoto's Relation (u_N), Impedance Relation (u_{IR}), the Rao's specific velocity method relation (u_R), Van Deel's ideal mixing relation (u_{IMR}) and theoretical sound velocity according to Free Length Theory (u_{FLT}). Their validity was verified by measuring $\sigma\%$ and APE.

3.1. Density (ρ)

The densities, (ρ) of the system W + NBA are shown in Table 1 and Figure 1(a) at different temperatures which are required to calculate some derived properties from sound velocity data. This time, we measured ρ by Anton Paar densitymeter (DSA 5000 M) instead of a pycnometer [14]. Though we changed the instrument, our present values agreed well with previous data. It can be seen from Figure 1(a) that the density decreases rapidly as NBA is added to water however with increasing concentration of NBA, the rate of fall of density decreases. At lower concentrations of NBA, the rate

of change of density with temperature appears to be much lower than the rate at higher concentrations of NBA [14].

Table 1: Densities, ρ / kg.m^{-3} of W + NBA system for different molar ratios at different temperatures.

x_2	Densities, ρ / kg.m^{-3}				
	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
1.0000	727.3	722.6	717.6	712.7	707.8
0.8996	741.0	736.0	731.1	725.7	720.3
0.7991	754.1	749.3	744.5	739.7	733.9
0.6991	767.1	762.4	757.6	752.8	748.0
0.6014	782.7	778.1	773.8	768.7	762.3
0.4977	798.4	793.8	789.2	784.5	779.7
0.3970	818.1	813.7	809.2	804.7	800.1
0.2990	841.3	837.1	832.8	828.4	824.0
0.1992	875.0	871.1	867.1	863.1	859.0
0.0983	919.2	915.9	912.4	908.9	905.3
0.0000	995.7	994.1	992.2	990.3	988.1

3.2. Sound Velocity (u)

The sound velocities (u) of the systems W + NBA at different temperatures for different molar ratios are shown in Table 2 and plotted in Figure 1(b). The sound velocity of water is usually high. Sound velocity in water at 35^0C and 45^0C has been measured by plotting given data against temperature (The Engineering ToolBox, www.engineeringtoolbox.com). With the addition of amine in water, sound velocity decreases gradually from the water-rich region to the amine-rich region.

3.3. Deviation in Sound Velocity (Δu)

Deviation in sound velocities, (Δu) of W + NBA system at different temperatures are calculated according to Equation 1 and shown in Table 2 and plotted in. Figure 1(c). The values of Δu are fitted to the Redlich-Kister polynomial (Equation 8). The co-efficient of this equation along with Standard Deviations (Equation 9) are listed in Table 7. On examination of Δu versus x_2 curve following characteristics are observed:

1) For W + NBA Δu in Δu versus x_2 curve is negative except at 303.15K temperature where at initial stage Δu value rises slightly.
2) With the increment of temperature, Δu value decreases.
3) For all the lines, they give minima and these minima shift from $x_2 \approx 0.2$ to $x_2 \approx 0.4$ with the rise in temperature.

Positive Δu values indicate the experimental values are greater than those of the ideal one, i.e. to propagate sound waves and feel fewer obstacles. Whereas negative values of Δu indicate more obstacles to propagating sound waves in the system which is observed in W + NBA system than that of ideal values. With the addition of butylamine, rowdiness due to intermolecular interaction increases. Hence, the value of Δu decreases. As the temperature increases, the stability of H bonding between amine and water decreases. So, we observe the shifting of minima from lower temperature to higher temperature in the water-rich region.

Table 2: Sound velocities, u / m.s^{-1} and deviation in sound velocities, Δu / m.s^{-1} of W + NBA system for different molar ratios at different temperatures.

x_2	u	Δu	u	Δu	u	Δu	u	Δu	u	Δu
	303.15 K		308.15 K		313.15 K		318.15 K		323.15 K	
1.0000	1221.10	0.00000	1212.56	0.00000	1204.62	0.00000	1196.79	0.00000	1188.14	0.00000
0.8996	1247.27	-2.53436	1237.79	-5.23538	1228.32	-8.56655	1218.84	-11.80588	1209.37	-14.19714
0.7991	1272.63	-5.90731	1262.16	-11.36110	1251.55	-17.63524	1241.26	-23.27549	1230.60	-28.42957
0.6991	1289.97	-17.15731	1280.83	-23.03510	1270.04	-31.28324	1260.42	-37.83649	1250.49	-43.82557
0.6014	1312.60	-22.45974	1301.58	-31.93118	1288.56	-44.16207	1278.85	-52.35191	1265.37	-63.42000
0.4977	1331.97	-32.73757	1321.83	-43.14791	1310.91	-55.13917	1299.71	-66.46058	1288.97	-76.41158
0.3970	1361.85	-31.64770	1350.65	-44.88432	1339.42	-58.99214	1327.22	-72.90763	1316.74	-84.17458
0.2990	1391.73	-29.78590	1375.99	-49.28144	1360.66	-69.24738	1350.24	-82.93421	1344.50	-90.99486
0.1992	1432.24	-17.80872	1418.41	-37.14475	1404.71	-57.27110	1391.64	-75.18777	1379.70	-91.01029
0.0983	1482.11	3.21397	1473.23	-12.94185	1463.55	-30.85835	1453.01	-47.84226	1441.31	-65.00386
0.0000	1507.00	0.00000	1516.00	0.00000	1526.00	0.00000	1534.00	0.00000	1541.00	0.00000

Table 3: Isentropic compressibility, k_s / $10^{-10} Pa^{-1}$ and excess isentropic compressibility, K_s^E / $10^{-10} Pa^{-1}$ of W + NBA system for different molar ratios at different temperatures.

x_2	K_s	K_s^E	K_s	K_s^E	K_s	K_s^E	K_s	K_s^E	K_s	K_s^E
	303.15 K		308.15 K		313.15 K		318.15 K		323.15 K	
1.0000	9.22113	0.00000	9.41230	0.00000	9.60323	0.00000	9.79619	0.00000	10.00816	0.00000
0.8996	8.67493	-0.45181	8.86841	-0.44532	9.06623	-0.43425	9.27614	-0.41334	9.49249	-0.40506
0.7991	8.18744	-0.82618	8.37706	-0.81852	8.57498	-0.80231	8.77482	-0.78669	8.99765	-0.76718
0.6991	7.83418	-1.04232	7.99541	-1.05690	8.18287	-1.04499	8.36108	-1.04518	8.54925	-1.05443
0.6014	7.41549	-1.29475	7.58627	-1.29229	7.78327	-1.26332	7.95475	-1.26310	8.19303	-1.21492
0.4977	7.05997	-1.42422	7.21001	-1.43222	7.37365	-1.42627	7.54609	-1.41530	7.71902	-1.42217
0.3970	6.59048	-1.60139	6.73672	-1.59973	6.88811	-1.59247	7.05482	-1.57437	7.20874	-1.58639
0.2990	6.13660	-1.66170	6.30959	-1.61487	6.48586	-1.56417	6.62104	-1.55996	6.71344	-1.61380
0.1992	5.57125	-1.63676	5.70588	-1.60007	5.84434	-1.55862	5.98244	-1.52429	6.11549	-1.50576
0.0983	4.95234	-1.28158	5.03068	-1.25297	5.11675	-1.21494	5.21140	-1.17729	5.31743	-1.12751
0.0000	4.42240	0.00000	4.37713	0.00000	4.32787	0.00000	4.29145	0.00000	4.21924	0.00000

3.4. Isentropic Compressibility (K_s)

Isentropic Compressibility (K_s) of W + NBA system was calculated by using Equation 2, tabulated in Table 3 and plotted in Figure 1(d). K_s have been fitted to the general polynomial Equation 7. And the coefficients and regression values are given in Table 6. With close examination of the K_s versus x_2 curves, the following characteristics have been observed.
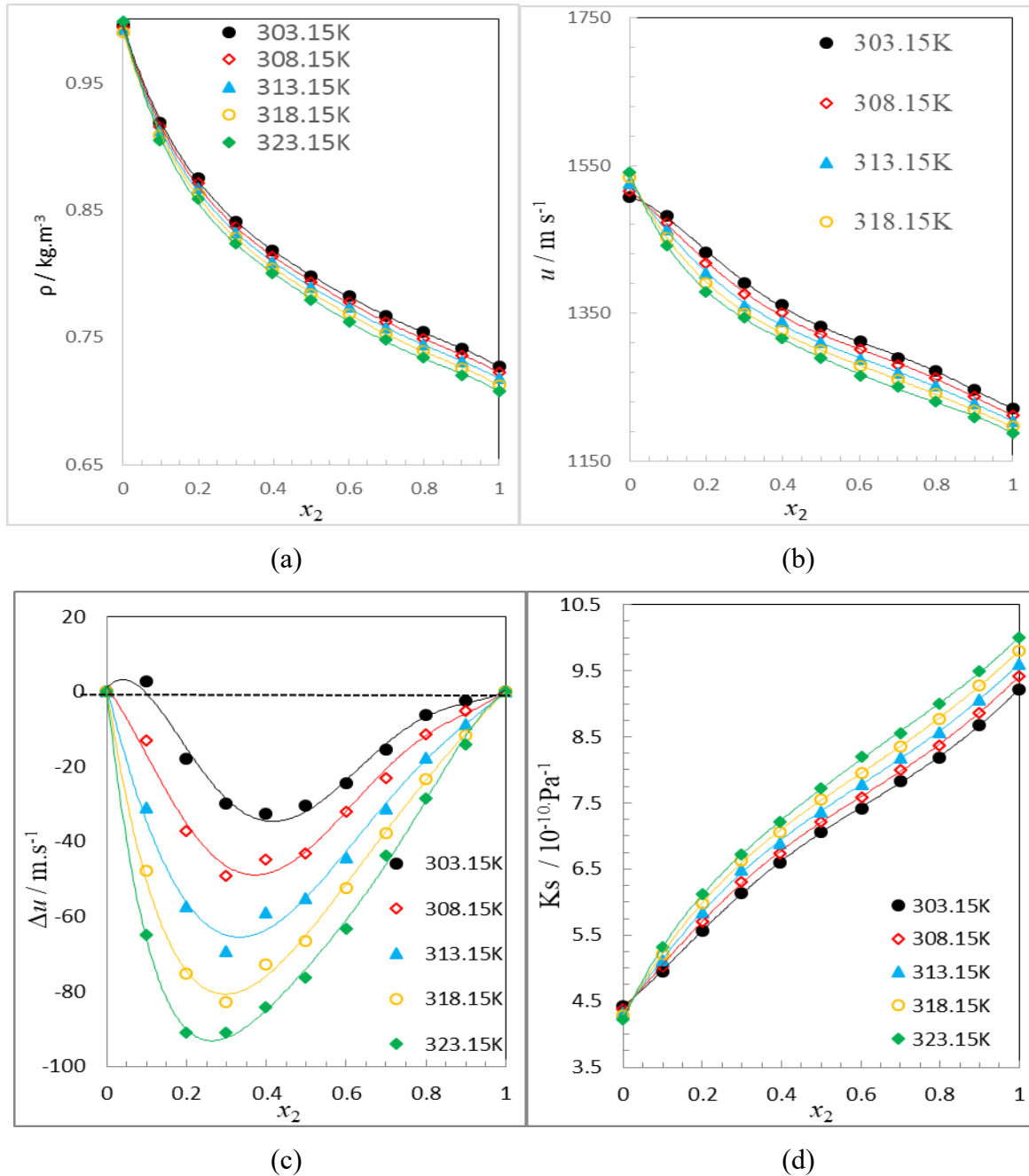

(a)

(b)

(c)

(d)

Figure 1: (a) Densities, ρ / kg.m^{-3}, (b) Sound velocities, u / m.s^{-1}, (c) Deviation in sound velocities, Δu / m.s^{-1} and (d) Isentropic compressibility, K_s / $10^{-10}\,Pa^{-1}$ of W + NBA system for different molar ratios at different temperatures. Solid lines represent polynomial fitting values.

1) Isentropic Compressibility values of W + NBA are all positive and gradually increase.
2) Initially, K_s lines are twisted and after crossing this point K_s lines are separated and rise gradually up to the whole mole fraction in amines.
3) K_s values increase with the rise of temperature.

3.5. Excess Isentropic Compressibility (K_s^E)

Excess Isentropic Compressibility (K_s^E) of W + NBA was calculated at different temperatures by using Equation 3, tabulated in Table 3 and plotted in Figure 2(a). K_s^E values have been fitted to the Redlich-Kister Equation 8. And the coefficients and standard deviations ($\sigma\%$) are given in Table 7. On examination, K_s^E versus x_2 curves give the following characteristics:

1) K_s^E values are all negative.
2) Temperature effect is prominent at $x_2 \approx 0.1$ to $x_2 \approx 0.4$. It is negligible in extreme water-rich and amine-rich regions.

K_s^E is nothing but the pressure dependence volume. The observed K_s^E is negative. It indicates that these are less than ideal values. It is possible when the molecules comprising the systems are in a closer packing situation. With the addition of NBA to water, the solute can form hydrogen bonding of the type N – H------ O (between the hydrogen atom of - NH_2 group of butylamine and the oxygen atom of – OH group of a water molecule) leading to a contraction in volume which should result in negative K_s^E values. In this volume contraction, the following factors are responsible:

1) Interstitial accommodation of molecules of one component into the structural network of the other component.
2) Favorable geometrical fittings.
3) Size differences of molecules.
4) Hydrophobic hydration is particularly important for these systems.

Considering the structural features and related properties of water and butylamines, high negative K_s^E values are observed. Hydrophobic hydration plays the most important role which is in accord with the work of Saleh et. al [14].

3.6. Specific Acoustic Impedance (Z)

Specific Acoustic Impedance (Z) of W + NBA system was calculated at different temperatures by using Equation 4, tabulated in Table 4 and plotted in Figure 2(b). Z values have been fitted to the general polynomial Equation 7. The coefficients and regression values are given in Table 6. It is observed that the Z versus x_2 curve decreases with the addition of solute (butylamine). This behavior of acoustic impedance becomes responsible for the propagation of ultrasonic waves, and this behavior can be explained on the basis of hydrophobic interaction between water and butylamine. This behavior indicates the presence of strong molecular interaction by the addition of solute to water [24].

3.7. Rao's Constant (R) and Wada's Constant (Wa)

Rao's Constant (R) and Wada's Constant (Wa) of W + NBA system were calculated at different temperatures according to Equations 5 & 6 and listed in Table 5. And these have been plotted in Figures 2(c, d) respectively. R and Wa values have been fitted to the general polynomial Equation 7. And the coefficients and regression values are given in Table 6. It is observed that the lines increase

with the addition of butylamines to water up to the whole composition. And the curves increase with a slight curvature.

(a)

(b)

(c)

(d)
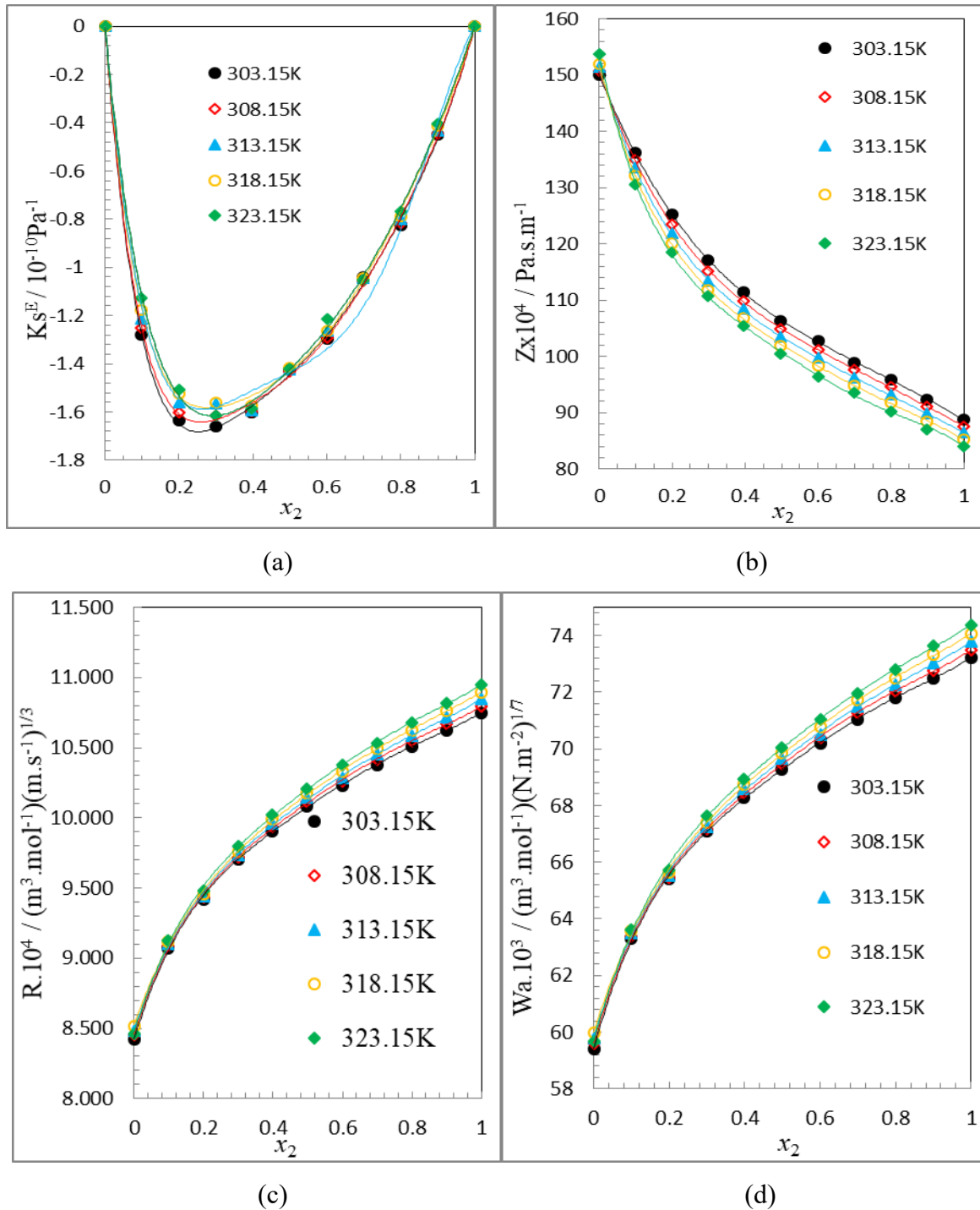
Figure 2: (a) Excess isentropic compressibility, $K_s^E / 10^{-10} Pa^{-1}$, (b) Specific acoustic impedance, Z / Pa.s.m^{-1}, (c) Rao's constant, R.10^4 / $(m^3.mol^{-1})(m.s^{-1})^{\frac{1}{3}}$ and (d) Wada's constant, Wa. 10^3 / $(m^3/mol)(N.m^{-2})^{\frac{1}{7}}$ of W + NBA system for different molar ratios at different temperatures. Solid lines represent polynomial fitting values.

Table 4: Specific Acoustic Impedance, Z / Pa.s.m^{-1} of W + NBA system for different molar ratios at different temperatures.

x_2	Z / Pa.s.m^{-1}				
	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
1.0000	88.8106	87.6196	86.4435	85.2952	84.0965
0.8996	92.4216	91.0976	89.7969	88.4476	87.1085
0.7991	95.9731	94.5788	93.1791	91.8119	90.3137
0.6991	98.9526	97.6489	96.2225	94.8905	93.5388
0.6014	102.7372	101.2746	99.7088	98.3001	96.4579
0.4977	106.3414	104.9273	103.4533	101.9604	100.5067
0.3970	111.4176	109.9028	108.3884	106.7999	105.3514
0.2990	117.0893	115.1816	113.3137	111.8570	110.7883
0.1992	125.3233	123.5594	121.8086	120.1143	118.5180
0.0983	136.2415	134.9281	133.5359	132.0616	130.4791
0.0000	150.0475	150.6995	151.4158	151.9044	153.8026

The values of Rao's Constant (R) or Molar Sound Velocity and Wada's Constant (Wa) or Molar Compressibility increase with concentration because of the presence of more molecules and thus leading to close packing of the medium. This thereby increases the interaction between solute and solvent [24].

3.8. Theoretical correlative model for Sound Velocity

In order to correlate the experimental sound velocity (u) value, different theoretical relations like Nomoto's Relation (u_N) (Eqn. 10), The Impedance Relation (u_{IR}) (Eqn. 11), The Rao's Specific Velocity Method (u_R) (Eqn. 12), Ideal mixing relation based on Van Deal and Vangeel theory (u_{IMR}) (Eqn. 15) and relation based on Free Length Theory (u_{FLT}) (Eqn. 16) were used. The correlating ability of each relation was tested by calculating standard percentage deviation ($\sigma\%$), and average percentage error (APE) according to Equations 17 and 18 respectively. Experimental and different theoretical sound velocities (u_{exp}, u_N, u_{IR}, u_R, u_{IMR} and u_{FLT}) at different temperatures were listed in Table 8 and plotted in Figure 3 for W + NBA system. The $\sigma\%$ and APE values for W + NBA system at different temperatures for different theoretical sound velocities has been presented in Tables 9, $\sigma\%$ values for W + NBA system are plotted in Figure 4. APE values for W + NBA system are shown in Figure 5. If the experimental value is more than the accepted value, we get a positive value of APE and a negative APE indicates that the experimental value is less than the accepted value. It is observed from the tabulated values that the theoretical values of ultrasonic velocities evaluated by the above-mentioned relations show deviations from the experimental values. The reason for the deviation may be the limitations and approximations incorporated in these theories. In Nomoto's Relation, the effect of volume change due to mixing was ignored i.e., the interaction between the molecules was not taken into the interpretation. In free length theory, it was anticipated that the molecular shape is spherical but it was not correct in all cases. In the case of ideal mixing relation, it was assumed that the ratio of specific heats and volumes are equal. Again molecular interactions were ignored. Also in the Impedance Relation (u_{IR}) and the Rao's Specific Velocity Method (u_R) no molecular interactions were considered. Among them, sound velocities according to the Impedance Relation (u_{IR}) are the closest to the experimental values.

Upon mixing two liquids, the interaction between the molecules of the two liquids take place because of the presence of various types of forces such as dispersion forces, hydrogen bonding, dipole-dipole, dipole-induced dipole interactions and hydrophobic interaction. Thus, the observed deviation of theoretical values of velocity from the experimental values shows the molecular interactions between the unlike molecules in the liquid mixture.

Table 5: Rao's constant, R.10^4 / $(m^3 \cdot mol^{-1})(m \cdot s^{-1})^{\frac{1}{3}}$ and Wada's constant, Wa. 10^3 / $(m^3/mol)(N \cdot m^{-2})^{\frac{1}{7}}$ of W + NBA system for different molar ratios at different temperatures.

x_2	R	W	R	W	R	W	R	W	R	W
	303.15 K		308.15 K		313.15 K		318.15 K		323.15 K	
1.0000	10.749	73.218	10.793	73.478	10.845	73.778	10.896	74.074	10.945	74.360
0.8996	10.625	72.494	10.670	72.759	10.715	73.018	10.766	73.320	10.819	73.625
0.7991	10.510	71.822	10.548	72.045	10.587	72.271	10.627	72.505	10.680	72.814
0.6991	10.379	71.055	10.419	71.286	10.454	71.496	10.494	71.729	10.534	71.963
0.6014	10.231	70.187	10.263	70.373	10.286	70.505	10.328	70.755	10.378	71.046
0.4977	10.080	69.293	10.112	69.483	10.143	69.667	10.175	69.852	10.208	70.050
0.3970	9.909	68.288	9.936	68.445	9.963	68.606	9.989	68.757	10.020	68.939
0.2990	9.706	67.086	9.718	67.159	9.732	67.240	9.758	67.395	9.797	67.622
0.1992	9.422	65.399	9.434	65.469	9.446	65.544	9.461	65.630	9.479	65.737
0.0983	9.072	63.309	9.087	63.400	9.101	63.486	9.115	63.566	9.126	63.636
0.0000	8.422	59.402	8.452	59.586	8.486	59.792	8.518	59.984	8.464	59.659

Table 6: Coefficients, a_i, of Equation 7, expressing densities, ρ / kg.m^{-3}, sound velocities, u / m.s^{-1}, isentropic compressibilities, k_s / $10^{-10}\, Pa^{-1}$, specific acoustic impedance, z / pa.s.m^{-1} and their regration indices, R^2 of W + NBA system for different molar ratios at different temperatures.

Property	T (K)	a_0	a_1	a_2	a_3	a_4	a_5	R^2
ρ / kg.m^{-3}	303.15	0.9952	- 0.9462	2.2567	- 3.4620	2.7667	-0.8833	0.9999
	308.15	0.9936	- 0.9684	2.3277	- 3.5711	2.8424	-0.9017	0.9998
	313.15	0.9917	- 0.9892	2.3927	- 3.6677	2.9081	-0.9182	0.9998
	318.15	0.9897	- 1.008	2.4500	- 3.7557	2.9704	-0.934	0.9998
	323.15	0.9972	- 1.1741	3.2437	- 5.4360	4.5717	-1.4952	0.9996
u / m.s^{-1}	303.15	1507.9	- 156.73	- 1851.2	4992.1	- 5011.7	1741.1	0.9995
	308.15	1516.9	- 428.35	- 762.44	3116.8	- 3540.3	1310.4	0.9995
	313.15	1526.9	- 734.32	594.02	476.18	- 1188.2	530.32	0.9994
	318.15	1534.4	- 1004.8	1821.6	- 1933.8	959.35	-180.06	0.9998

	T(K)							σ
	303.15	1540.8	- 1302.4	3485.4	- 5811.9	4909.3	-1633.7	0.9998
K_s / $10^{-10} Pa^{-1}$	303.15	4.4194	4.9449	8.2363	- 26.004	25.646	-8.0224	0.9999
	308.15	4.3732	6.6909	2.8836	- 18.973	22.043	-7.6067	0.9999
	313.15	4.3231	8.7252	- 4.7855	- 5.0783	10.369	-3.9507	0.9998
	318.15	4.2895	10.461	- 10.944	5.2014	2.6398	-1.8511	0.9999
	323.15	4.2222	13.244	- 25.599	39.805	- 33.005	11.348	0.9999
Z / $Pa.s.m^{-1}$	303.15	150.06	- 159.81	201.66	- 115.2	- 5.1693	17.267	0.9999
	308.15	150.71	- 189.31	323.63	- 332.76	172.08	-36.719	0.9999
	313.15	151.41	- 221.54	467.42	- 612.79	421.41	-119.48	0.9999
	318.15	151.85	- 250.1	598.49	- 872.53	654.93	-197.39	0.9999
	323.15	153.64	- 303.07	875.45	- 1489.1	1263.4	-416.39	0.9998

Table 7: Coefficients, A_i, of Equation 8, expressing deviation in sound velocities, Δu / $m.s^{-1}$, excess isentropic compressibilities, K_s^E / $10^{-10} Pa^{-1}$, and standard deviation, (σ), Eqn. 9 of W + NBA system for different molar ratios at different temperatures.

Property	T(K)	A_0	A_1	A_2	A_3	A_5	σ
Δu / $m.s^{-1}$	303.15	-122.174	78.998	50.789	104.655	223.124	-444.620
	308.15	-164.683	134.636	-56.445	180.984	239.599	-475.217
	313.15	-214.924	167.421	-146.072	339.232	217.646	-558.759
	318.15	-259.228	216.602	-166.200	314.840	75.894	-403.113
	323.15	-302.245	204.379	-116.269	497.399	-175.280	-405.268
K_s^E / $10^{-10} Pa^{-1}$	303.15	-5.7770	3.2338	-4.0010	1.5374	-3.3356	3.9363
	308.15	-5.7999	3.0466	-3.3355	1.2156	-3.8560	4.5782
	313.15	-5.7555	3.1165	-2.7276	-0.0151	-4.2901	5.9267
	318.15	-5.7345	1.9719	-4.9871	2.6207	0.5359	4.5991
	323.15	-5.6927	3.6462	-3.6153	-2.0346	-1.2556	6.8048

In statistics, the Standard Percentage Deviation ($\sigma\%$) is used to measure quantitatively the amount of variation or dispersion of a set of data values. A low $\sigma\%$ indicates that the data points tend to be close to the mean. It is observed that $\sigma\%$ values and APE values of sound velocities according to the Impedance Relation (u_{IR}) are the closest.

(a)

(b)

(c)

(d)

(e)

Figure 3: Experimental and different theoretical sound velocities, ($u_{expt.}$, u_N, u_{IR}, u_R, u_{IMR} and u_{FLT}) in $m.s^{-1}$, of W +NBA system for different molar ratios at different temperatures; (a) 303.15 K, (b) 308.15 K, (c) 313.15 K, (d) 318.15 K, and (e) 323.15 K.

Figure 4: Standard percentage deviation, $\sigma\%$ for different theoretical sound velocities, (u_N, u_{IR}, u_R, u_{IMR} and u_{FLT}) of W + NBA system at different molar ratios and different temperatures; (a) 303.15 K, (b) 308.15 K, (c) 313.15 K, (d) 318.15 K, and (e) 323.15 K.

Figure 5: Average percentage error, APE for different theoretical sound velocities, (u_n, u_{ir}, u_r, u_{imr} and u_{flt}) of W + NBA system at different molar ratios and different temperatures.
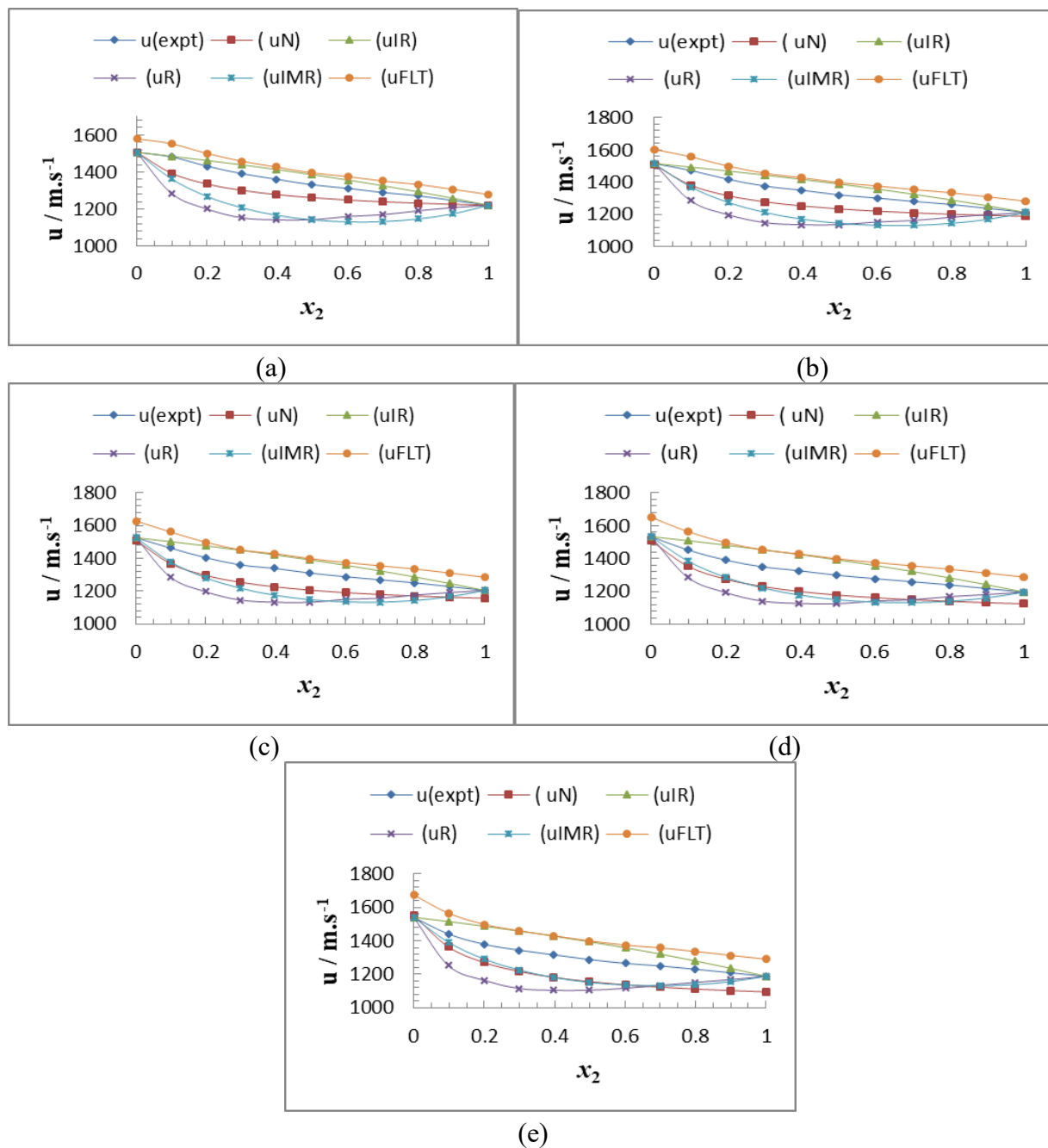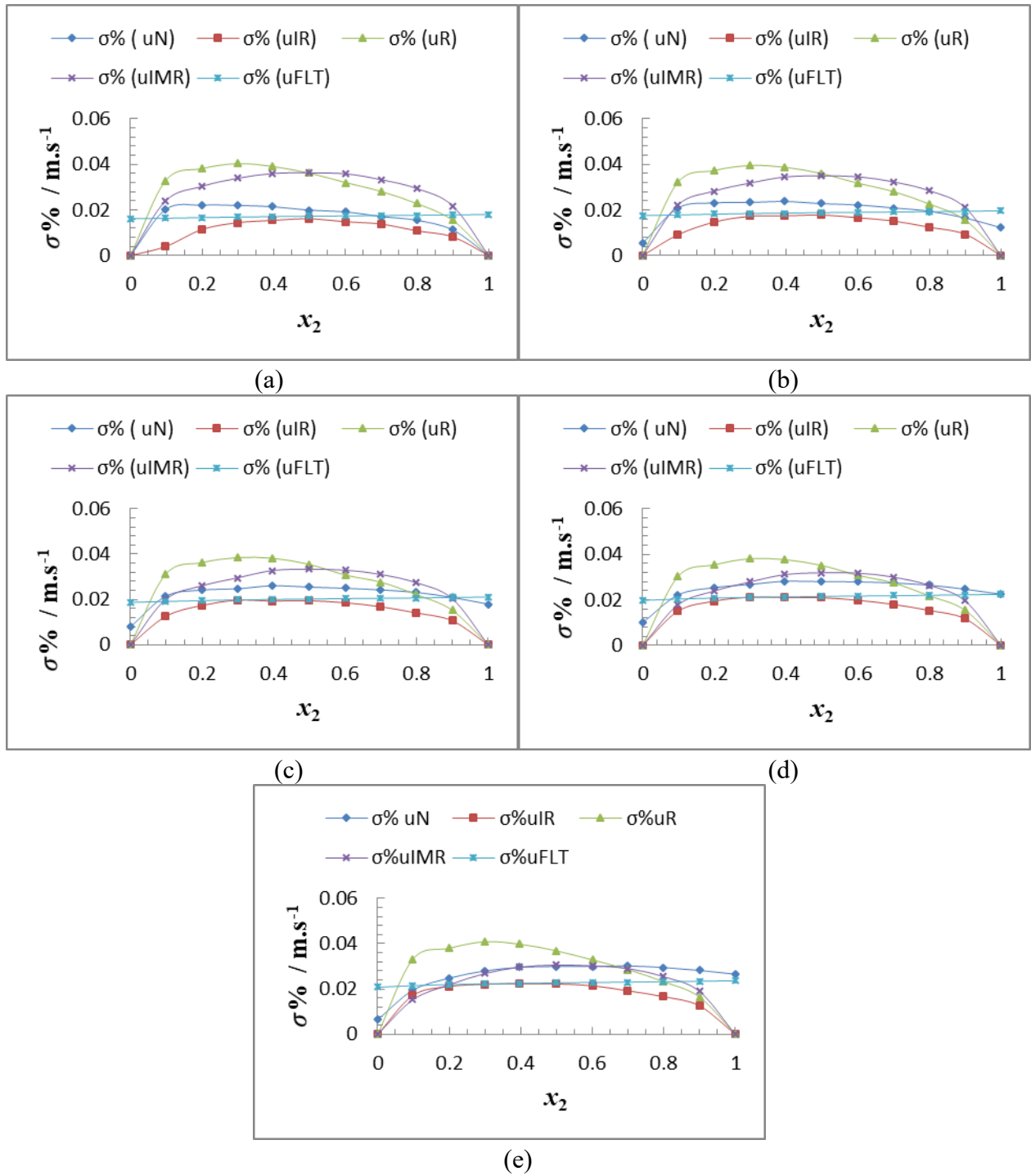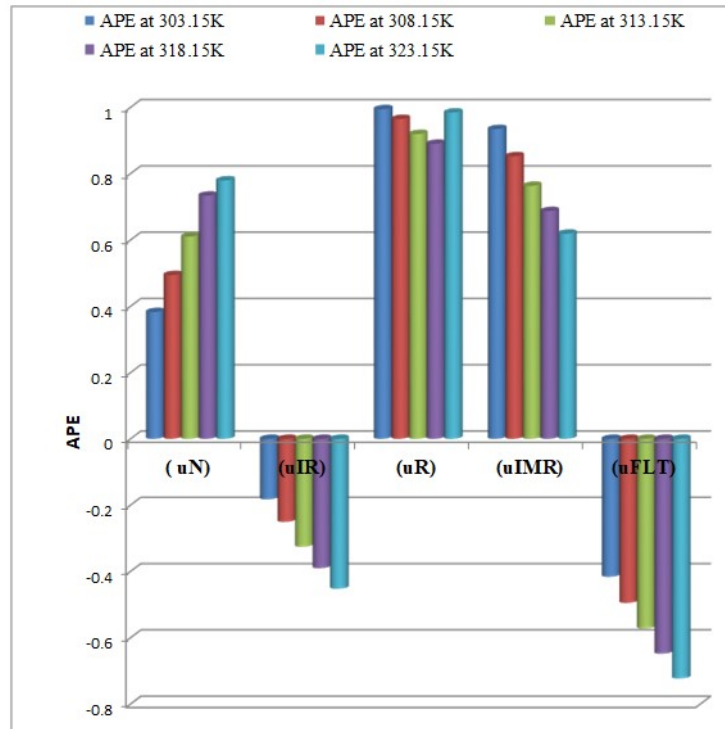
Table 8: Experimental and different theoretical sound velocities, ($u_{expt.}$, u_n, u_{ir}, u_r, u_{imr} and u_{flt}) in $m.s^{-1}$, of W +NBA system for different molar ratios at different temperatures.

x_2	303.15 K	308.15 K	313.15 K	318.15 K	323.15 K
			$u_{expt.}$		
1.0000	1221.10	1212.56	1204.62	1196.79	1188.14
0.8996	1247.27	1237.79	1228.32	1218.84	1209.37
0.7991	1272.63	1262.16	1251.55	1241.26	1230.60
0.6991	1289.97	1280.83	1270.04	1260.42	1250.49
0.6014	1312.60	1301.58	1288.56	1278.85	1265.37
0.4977	1331.97	1321.83	1310.91	1299.71	1288.97
0.3970	1361.85	1350.65	1339.42	1327.22	1316.74
0.2990	1391.73	1375.99	1360.66	1350.24	1344.50
0.1992	1432.24	1418.41	1404.71	1391.64	1379.70
0.0983	1482.11	1473.23	1463.55	1453.01	1441.31
0.0000	1507.00	1516.00	1526.00	1534.00	1541.00
			u_N		
1.0000	1221.10	1189.20	1157.06	1126.15	1095.11
0.8996	1226.34	1194.98	1163.35	1132.88	1102.87
0.7991	1232.64	1201.92	1170.92	1140.98	1112.23
0.6991	1240.31	1210.38	1180.15	1150.86	1123.66
0.6014	1249.65	1220.69	1191.41	1162.92	1137.65
0.4977	1262.42	1234.81	1206.85	1179.48	1156.90
0.3970	1279.07	1253.24	1227.03	1201.16	1182.20
0.2990	1301.71	1278.36	1254.59	1230.83	1216.99
0.1992	1336.17	1316.69	1296.79	1276.38	1270.76
0.0983	1394.35	1381.73	1368.71	1354.36	1363.84
0.0000	1507.00	1508.66	1510.28	1509.08	1552.17
			u_{IR}		
1.0000	1221.10	1212.56	1204.62	1196.79	1188.14

0.8996	1258.99	1252.95	1247.58	1242.06	1236.12
0.7991	1294.30	1290.54	1287.52	1284.09	1280.49
0.6991	1327.10	1325.41	1324.53	1322.98	1321.41
0.6014	1357.11	1357.28	1358.30	1358.45	1358.61
0.4977	1386.96	1388.95	1391.84	1393.63	1395.38
0.3970	1414.16	1417.78	1422.34	1425.59	1428.69
0.2990	1439.08	1444.18	1450.23	1454.80	1459.05
0.1992	1463.04	1469.53	1477.00	1482.80	1488.09
0.0983	1485.91	1493.72	1502.51	1509.47	1515.68
0.0000	1507.00	1516.00	1526.00	1534.00	1541.00
			, u_R		
1.0000	1221.10	1212.56	1204.62	1196.79	1188.14
0.8996	1208.98	1199.23	1192.04	1182.10	1168.57
0.7991	1191.30	1183.04	1176.55	1169.51	1151.69
0.6991	1170.57	1163.06	1157.20	1150.64	1134.63
0.6014	1160.98	1154.22	1150.67	1142.75	1117.25
0.4977	1143.55	1137.25	1132.42	1126.70	1105.46
0.3970	1142.46	1137.05	1133.07	1128.19	1104.38
0.2990	1153.64	1149.29	1146.31	1142.28	1115.69
0.1992	1201.20	1198.55	1197.23	1194.78	1164.75
0.0983	1285.19	1285.16	1286.44	1286.40	1252.18
0.0000	1507.00	1516.00	1526.00	1534.00	1541.00
			u_{IMR}		
1.0000	1221.10	1212.56	1204.62	1196.79	1188.14
0.8996	1175.50	1170.73	1166.47	1161.86	1156.39
0.7991	1147.26	1145.14	1143.52	1141.26	1138.11
0.6991	1133.20	1133.05	1133.43	1132.94	1131.58
0.6014	1131.56	1132.95	1134.89	1135.81	1135.86
0.4977	1142.75	1145.50	1148.85	1151.03	1152.35
0.3970	1167.51	1171.44	1176.00	1179.28	1181.70
0.2990	1207.42	1212.43	1218.15	1222.44	1225.86
0.1992	1269.26	1275.41	1282.34	1287.69	1292.16
0.0983	1363.72	1371.16	1379.46	1386.02	1391.64
0.0000	1507.00	1516.00	1526.00	1534.00	1541.00
			u_{FLT}		
1.0000	1279.69	1282.37	1285.54	1288.66	1290.75
0.8996	1307.11	1309.06	1310.83	1312.41	1313.82
0.7991	1333.69	1334.83	1335.62	1336.55	1336.88
0.6991	1351.86	1354.57	1355.35	1357.18	1358.49
0.6014	1375.58	1376.52	1375.11	1377.02	1374.65
0.4977	1395.88	1397.94	1398.97	1399.49	1400.29
0.3970	1427.19	1428.41	1429.39	1429.11	1430.46
0.2990	1458.50	1455.21	1452.06	1453.89	1460.62
0.1992	1500.96	1500.08	1499.07	1498.47	1498.86
0.0983	1553.22	1558.05	1561.86	1564.55	1565.79
0.0000	1579.31	1603.28	1628.50	1651.76	1674.09

Table 9: Standard percentage deviatin, $\sigma\%$ and average percentage error, APE for different theoretical sound velocities, (u_n, u_{ir}, u_r, u_{imr} and u_{flt}) in $m.s^{-1}$, of W + NBA system at different molar ratios and different temperatures.

x_2	$\sigma\%$ (u_N)	$\sigma\%$ (u_{IR})	$\sigma\%$ (u_R)	$\sigma\%$ (u_{IMR})	$\sigma\%$ (u_{FLT})
			303.15 K		
1.0000	0.00000	0.00000	0.00000	0.00000	0.01803
0.8996	0.01125	0.00820	0.01543	0.02173	0.01784
0.7991	0.01547	0.01085	0.02282	0.02943	0.01767
0.6991	0.01713	0.01384	0.02815	0.03331	0.01755
0.6014	0.01914	0.01482	0.03198	0.03585	0.01739
0.4977	0.01992	0.01612	0.03619	0.03629	0.01727
0.3970	0.02145	0.01542	0.03909	0.03600	0.01708

0.2990	0.02198	0.01442	0.04033	0.03390	0.01689
0.1992	0.02212	0.01144	0.03815	0.03033	0.01665
0.0983	0.02026	0.00396	0.03292	0.02406	0.01637
0.0000	0.00000	0.00000	0.00000	0.00000	0.01623
APE	0.38256	-0.18253	0.99460	0.93445	-0.41621
308.15 K					
1.0000	0.01225	0.00000	0.00000	0.00000	0.01965
0.8996	0.01651	0.00937	0.01561	0.02109	0.01944
0.7991	0.01947	0.01245	0.02267	0.02848	0.01926
0.6991	0.02091	0.01519	0.02813	0.03235	0.01911
0.6014	0.02221	0.01658	0.03171	0.03456	0.01896
0.4977	0.02278	0.01778	0.03602	0.03495	0.01882
0.3970	0.02374	0.01742	0.03875	0.03446	0.01861
0.2990	0.02330	0.01724	0.03950	0.03180	0.01844
0.1992	0.02309	0.01467	0.03730	0.02827	0.01816
0.0983	0.02087	0.00914	0.03217	0.02222	0.01782
0.0000	0.00542	0.00000	0.00000	0.00000	0.01757
APE	0.49407	-0.25044	0.96505	0.85248	-0.49492
313.15 K					
1.0000	0.01797	0.00000	0.00000	0.00000	0.02110
0.8996	0.02089	0.01061	0.01524	0.02033	0.02089
0.7991	0.02312	0.01405	0.02219	0.02740	0.02070
0.6991	0.02422	0.01680	0.02768	0.03109	0.02055
0.6014	0.02494	0.01854	0.03077	0.03293	0.02040
0.4977	0.02549	0.01949	0.03557	0.03341	0.02022
0.3970	0.02605	0.01930	0.03822	0.03277	0.02001
0.2990	0.02475	0.01968	0.03851	0.02955	0.01985
0.1992	0.02415	0.01736	0.03628	0.02601	0.01954
0.0983	0.02145	0.01253	0.03119	0.02004	0.01914
0.0000	0.00791	0.00000	0.00000	0.00000	0.01874
APE	0.61007	-0.32560	0.91970	0.76328	-0.57222
318.15 K					
1.0000	0.02250	0.00000	0.00000	0.00000	0.02243
0.8996	0.02468	0.01170	0.01546	0.01959	0.02222
0.7991	0.02646	0.01537	0.02184	0.02642	0.02202
0.6991	0.02742	0.01803	0.02745	0.03005	0.02185
0.6014	0.02792	0.01980	0.03078	0.03175	0.02170
0.4977	0.02803	0.02097	0.03520	0.03194	0.02152
0.3970	0.02818	0.02098	0.03770	0.03110	0.02130
0.2990	0.02677	0.02119	0.03806	0.02788	0.02111
0.1992	0.02536	0.01941	0.03541	0.02387	0.02080
0.0983	0.02211	0.01501	0.03025	0.01781	0.02035
0.0000	0.00997	0.00000	0.00000	0.00000	0.01981
APE	0.73381	-0.39023	0.89017	0.68732	-0.64813
323.15 K					
1.0000	0.02656	0.00000	0.00000	0.00000	0.02366
0.8996	0.02821	0.01262	0.01648	0.01898	0.02345
0.7991	0.02949	0.01663	0.02326	0.02548	0.02325
0.6991	0.03022	0.01922	0.02860	0.02906	0.02307
0.6014	0.02995	0.02143	0.03284	0.03021	0.02293
0.4977	0.02995	0.02229	0.03695	0.03058	0.02272
0.3970	0.02958	0.02233	0.03979	0.02965	0.02248
0.2990	0.02798	0.02212	0.04088	0.02679	0.02224
0.1992	0.02476	0.02109	0.03795	0.02183	0.02196
0.0983	0.01946	0.01715	0.03311	0.01527	0.02148
0.0000	0.00649	0.00000	0.00000	0.00000	0.02078
APE	0.77961	-0.45206	0.98442	0.61888	-0.72271

4. Conclusion

Sound velocity and density of W + NBA system were measured and various derived parameters, viz., Δu, K_s, K_s^E, Z, R and Wa (Wada's Constant) were calculated. Here, it is found that with the addition of amine in water, sound velocity decreases gradually and Δu versus x_2 curve is negative except at 303.15 K temperature. It is also observed that K_s values are positive whereas K_s^E

lines are negative. The variation of these properties with composition and temperature is explained through hydrogen bonding and hydrophobicity i.e. with the strength between constituents due to molecular interaction of the studied binary mixture. Without these five theoretical relations namely, Nomoto's Relation (u_N), The Impedance Relation (u_{IR}), The Rao's Specific Velocity Method (u_R), Ideal mixing relation based on Van Deal and Vangeel theory (u_{IMR}) and relation based on Free Length Theory (u_{FLT}) have been tested. The experimental and theoretical values of sound velocities correlated statistically through the standard percentage deviation ($\sigma\%$) and average percentage error (APE). From our analysis sound velocities according to the Impedance Relation (u_{IR}) is the best among the five relations.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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