

## Research Article

# Acoustic and Volumetric Properties of Mixture of (*N,N*-Dimethylacetamide + Ethyl Acrylate) with 1-Butanol or *iso*-Butanol or *t*-Butanol at 308.15 K

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Densities,  $\rho$ , and ultrasonic speeds,  $u$  of mixtures of 1-butanol or *iso*-butanol or *t*-butanol with equimolar mixture of (*N,N*-dimethylacetamide + Ethyl acrylate) over the entire composition range have been measured at  $T = 308.15$  K. Using the experimental results, deviation in ultrasonic speed,  $\Delta u$ , deviation in isentropic compressibility,  $\Delta k_s$ , excess molar volume,  $V_m^E$ , excess intermolecular free length,  $L_f^E$ , and excess acoustic impedance,  $Z^E$ , have been calculated. The variation of these properties with composition of the mixtures has been discussed in terms of molecular interactions in these mixtures. The deviation/excess properties have been fitted to Redlich-Kister type polynomial and the corresponding standard deviations have been calculated. Negative values of  $V_m^E$ ,  $\Delta k_s$ , and  $L_f^E$  and positive values of  $\Delta u$ , and  $Z^E$  are observed over the entire composition range. The observed negative and positive values of deviation/excess properties are attributed to the strong interactions between the unlike molecules of the mixtures. Further theoretical values of sound velocity in the mixtures have been evaluated using various theories and compared with experimental sound velocities to verify the applicability of such theories to the systems studied. Theoretical ultrasonic velocity data has been used to study molecular interactions in the systems investigated.

## 1. Introduction

The measurement of speed of sound in liquids enables for the determination of some useful acoustic and thermodynamic parameters that are found to be very sensitive to molecular interactions. Acrylic esters are important industrial chemicals that are widely used as precursors in the production of technically important special type polymers. Alkanol molecules are polar and self-associated through hydrogen bonding of their hydroxyl groups [1]. *N,N*-dimethylacetamide is commonly used as a solvent for fibres or in the adhesive industry. Ethyl acrylate and *N,N*-dimethylacetamide molecules are polar having dipole moments 1.96 D [2] and 3.72 D [2] at 298.15 K, respectively. A survey of the literature indicates that Francesconi and Comelli [3], Gonzalez and Ortega [4], and Liao et al. [5] reported

density and viscosity data for binary mixtures of esters with alkanols. Recently some researchers have reported the ultrasonic, volumetric, and viscometric studies of esters with alcohols [6–8]. *N,N*-dimethylacetamide as one component and +2-propoxyethanol, +2-isopropoxyethanol [9] and +aliphatic alcohols [10, 11], and +benzene, +toluene, +ethyl benzene [12] as other components.

Literature survey indicates that there have been no studies on these equimolar mixtures. As a part of our continuing research on ultrasonic, volumetric, and viscometric properties of binary mixtures/equimolar mixtures of amides/alkanols/acrylic esters [13–17], here, we report densities,  $\rho$ , ultrasonic speeds,  $u$  of 1-butanol, *iso*-butanol, *t*-butanol with equimolar mixture of (*N,N*-dimethylacetamide + Ethyl acrylate) over the entire composition range at  $T = 308.15$  K. From

TABLE 1: Comparison of experimental values of ultrasonic speed,  $u$ , and density,  $\rho$ , of pure liquids with the corresponding literature values at 308.15 K.

Compound	$u$ (m·s <sup>-1</sup> )		$\rho$ (kg·m <sup>-3</sup> )	
	Present work	Literature	Present work	Literature
<i>N,N</i> -Dimethylacetamide	1416.23	1417.13 [18]	927.4	927.8 [19]
Ethyl acrylate	1119.12	1118.70 [6]	903.8	904.9 [6]
1-butanol	1207.76	1208.80 [20]	798.0	798.12 [21] 801.91 [22]
<i>iso</i> -butanol	1175.20	1175.20 [7]	789.6	789.7 [23]
<i>t</i> -butanol	1083.60	1084.30 [7]	769.9	770.19 [24]

TABLE 2: Experimental values of ultrasonic speed,  $u$ , and density,  $\rho$ , of all the systems with mole fraction,  $x_1$ , or volume fraction,  $\Phi_1$ , of 1-butanol/*iso*-butanol/*t*-butanol at 308.15 K.

$x_1$	$\Phi_1$	$u$ (m·s <sup>-1</sup> )	$\rho$ (kg·m <sup>-3</sup> )	$x_1$	$\Phi_1$	$u$ (m·s <sup>-1</sup> )	$\rho$ (kg·m <sup>-3</sup> )	$x_1$	$\Phi_1$	$u$ (m·s <sup>-1</sup> )	$\rho$ (kg·m <sup>-3</sup> )
(DMA + EA) + 1-butanol				(DMA + EA) + <i>iso</i> -butanol				(DMA + EA) + <i>t</i> -butanol			
0.0000	0.0000	1260.95	919.62	0.0000	0.0000	1260.95	919.62	0.0000	0.0000	1260.95	919.62
0.1003	0.1005	1255.20	908.95	0.1186	0.1186	1251.60	907.21	0.1429	0.1422	1240.75	903.86
0.2031	0.2028	1249.38	897.99	0.2282	0.2277	1242.93	895.16	0.2647	0.2630	1225.30	889.23
0.3059	0.3066	1244.50	886.79	0.3379	0.3375	1234.37	882.61	0.3867	0.3826	1204.41	873.03
0.4056	0.4055	1239.11	875.39	0.4387	0.4373	1225.81	870.53	0.4924	0.4866	1186.90	857.74
0.5056	0.5067	1233.68	863.68	0.5411	0.5398	1216.98	857.66	0.5965	0.5905	1168.95	841.81
0.6061	0.6072	1228.05	851.53	0.6387	0.6372	1208.24	844.74	0.6857	0.6784	1153.70	827.60
0.7065	0.7092	1222.98	838.80	0.7375	0.7343	1199.64	831.32	0.7752	0.7683	1137.40	812.67
0.8043	0.8059	1217.00	826.11	0.8342	0.8296	1190.30	817.10	0.8498	0.8421	1122.60	799.94
0.9029	0.9038	1211.86	812.52	0.9098	0.9047	1183.30	805.36	0.9306	0.9260	1102.00	784.74
1.0000	1.0000	1207.76	798.00	1.0000	1.0000	1175.20	789.61	1.0000	1.0000	1083.60	769.90

the experimental results, various properties have been calculated and presented as follows: deviation in ultrasonic speed,  $\Delta u$ , deviation in isentropic compressibility,  $\Delta k_s$ , excess molar volume,  $V_m^E$ , excess intermolecular free length,  $L_f^E$ , and excess acoustic impedance,  $Z^E$ . The experimental values of  $u$  and  $\rho$  of pure liquids at  $T = 308.15$  K along with their literature [6, 7, 18–24] values are presented in Table 1. These results are in good agreement with the reported data.

The present work deals with the study of ultrasonic and volumetric properties to investigate the molecular interactions between the unlike molecules of the mixtures of 1-butanol, *iso*-butanol, and *t*-butanol with equi molar mixture of (*N,N*-dimethylacetamide + Ethyl acrylate) over the entire composition range at  $T = 308.15$  K (see Table 2 for experimental values).

## 2. Experimental Section

Analytical Reagent grade (A.R) of EA (mass fraction purity 0.99) was obtained from KEMPHASOL Company, Mumbai, India. *N,N*-dimethylacetamide (mass fraction purity 0.99) and isobutyl alcohol (mass fraction purity 0.99) are of Graded Reagent (G.R) and 1-butanol (mass fraction purity 0.99) and *t*-butanol (mass fraction purity 0.99) of (AR grade) obtained from LOBA Chemicals, Mumbai, India, are used in the present investigation and are further purified by standard

methods [25]. Equimolar mixture of DMA and EA is prepared. This solution is in turn used to prepare the liquid mixtures with 1-butanol or *iso*-butanol or *t*-butanol so that the entire composition range is covered (i.e., 0–100% of the alkanols). Mixtures are prepared by mass in air tight bottles. The mass measurements are performed with a METTLER TOLEDO (Switzerland make) ABB5-S/FACT digital balance with an accuracy of  $\pm 0.01$  mg. The uncertainty in the mole fraction is  $10^{-4}$ . The ultrasonic velocity of pure liquids and their mixtures has been measured by using a multifrequency ultrasonic interferometer (M-82 Model) supplied by Mittal enterprise, New Delhi, at a fixed frequency of 2 MHz with an accuracy of  $\pm 0.2\%$ . In the present study, the constant temperature water bath (digital electronic) supplied by Concord Instruments Co. Ltd., Chennai (RAAGA type), has been used. The instrument can maintain temperature to  $\pm 0.01$  K as per its specifications.

Densities of pure liquids and their mixtures have been determined by using a 5 cm<sup>3</sup> two stem double walled Parker & Parker type pycnometer [26]. This pycnometer is calibrated with triply distilled water. The pycnometer filled with air bubble free experimental liquids was kept in a transparent walled constant temperature bath for 20 to 30 min to attain thermal equilibrium. The positions of the liquid levels in the two arms are recorded with the help of traveling microscope. At least three to four measurements are performed, from which an average value of density of the experimental liquid is

TABLE 3: Coefficients  $A_i$  of Redlich-Kister type polynomial Equations (5) and (6) and the corresponding standard deviation,  $\sigma$ , of all the systems.

	$A_1$	$A_2$	$A_3$	$A_4$	$A_5$	$\sigma$
(DMA + EA) + 1-butanol						
$\Delta u$ ( $\text{m}\cdot\text{s}^{-1}$ )	-1.34	1.08	-5.59	4.16	-10.45	0.22
$V_m^E$ ( $\times 10^{-5}$ ) ( $\text{m}^3\cdot\text{mol}^{-1}$ )	-0.125	0.052	-0.009	0.064	-0.046	0.0004
$\Delta k_s$ ( $\times 10^{-10}$ ) ( $\text{Pa}^{-1}$ )	-0.475	0.110	-0.009	-0.032	0.180	0.0033
$L_f^E$ ( $\times 10^{-10}$ ) (m)	-0.014	0.002	0.003	0.0003	0.003	0.0001
$Z^E$ ( $\times 10^6$ ) ( $\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ )	0.019	-0.004	0.006	-0.003	-0.019	0.0001
(DMA + EA) + <i>iso</i> -butanol						
$\Delta u$ ( $\text{m}\cdot\text{s}^{-1}$ )	9.9	1.04	0.37	0.08	-11.06	0.14
$V_m^E$ ( $\times 10^{-5}$ ) ( $\text{m}^3\cdot\text{mol}^{-1}$ )	-0.255	0.090	-0.067	0.086	-0.048	0.0004
$\Delta k_s$ ( $\times 10^{-10}$ ) ( $\text{Pa}^{-1}$ )	-0.890	0.181	-0.068	-0.090	0.255	0.0023
$L_f^E$ ( $\times 10^{-10}$ ) (m)	-0.027	0.005	-0.003	0.002	0.004	0.0001
$Z^E$ ( $\times 10^6$ ) ( $\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ )	0.038	-0.005	0.002	-0.012	0.003	0.0001
(DMA + EA) + <i>t</i> -butanol						
$\Delta u$ ( $\text{m}\cdot\text{s}^{-1}$ )	51.65	-8.53	86.99	-52.88	-106.74	0.44
$V_m^E$ ( $\times 10^{-5}$ ) ( $\text{m}^3\cdot\text{mol}^{-1}$ )	-0.45	0.016	-0.063	0.237	-0.155	0.0005
$\Delta k_s$ ( $\times 10^{-10}$ ) ( $\text{Pa}^{-1}$ )	-2.470	0.603	-1.486	0.867	1.522	0.0057
$L_f^E$ ( $\times 10^{-10}$ ) (m)	-0.074	0.016	-0.051	0.039	0.047	0.0002
$Z^E$ ( $\times 10^6$ ) ( $\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ )	0.073	0.002	0.080	-0.067	-0.086	0.0004

determined. The reproducibility in the measured parameter density is 3 in  $10^5$  parts.

### 3. Results and Discussion

Using measured data of ultrasonic speed ( $u$ ) and density ( $\rho$ ), acoustic parameters such as the isentropic compressibility ( $k_s$ ), acoustic impedance ( $Z$ ), and the intermolecular free length ( $L_f$ ) have been determined from the following equations:

$$\begin{aligned}
 k_s &= \frac{1}{u^2 \rho}, \\
 Z &= u\rho, \\
 L_f &= Kk_s^{1/2},
 \end{aligned} \tag{1}$$

where  $K$  is Jacobson temperature dependent constant and is equal to  $K = (93.875 + 0.375T) \times 10^{-8}$ , where  $T$  is absolute temperature.

The deviation in ultrasonic speed in the mixtures has been calculated using the relation

$$\Delta u = u - (x_1 u_1 + x_2 u_2), \tag{2}$$

where “ $x$ ” is the mole fraction of the liquid mixture and the subscripts 1 and 2 stand for alkanols and equimolar mixture, respectively. Pertinent to the calculation of deviation and excess parameters, equimolar mixture is considered as one component of the liquid mixture [27].

The deviation in isentropic compressibility has been calculated from the following equation [28]:

$$\Delta k_s = k_s - (\Phi_1 k_{s1} + \Phi_2 k_{s2}). \tag{3}$$

Since  $k_s$  is not additive on mole fraction, it is additive on volume fraction. Hence, such values are calculated using volume fraction  $\Phi_i = x_i V_i / \sum x_i V_i$ .

The excess functions such as  $V_m^E$ ,  $L_f^E$ , and  $Z^E$  have been calculated using

$$\begin{aligned}
 V_m^E &= V_m - (x_1 V_{m1} + x_2 V_{m2}), \\
 L_f^E &= L_f - (x_1 L_{f1} + x_2 L_{f2}), \\
 Z^E &= Z - (x_1 Z_1 + x_2 Z_2).
 \end{aligned} \tag{4}$$

The values of deviation in ultrasonic velocity, excess molar volume, excess free length, and excess acoustic impedance have been fitted to a Redlich-Kister [29] type polynomial equation

$$Y^E = x(1-x) \sum A_i (1-2x)^{i-1}, \tag{5}$$

where  $Y^E$  is  $\Delta u$ ,  $V_m^E$ ,  $L_f^E$ , and  $Z^E$ . The subscript “ $i$ ” in the summation of the above equation takes values from 1 to 5.

The values of  $\Delta k_s$  have been fitted to Redlich-Kister type polynomial with volume fraction instead of mole fraction in the polynomial:

$$\Delta k_s = \Phi(1-\Phi) \sum A_i (1-2\Phi)^{i-1}. \tag{6}$$

The values of coefficients  $A_i$  ( $A_1$ ,  $A_2$ ,  $A_3$ ,  $A_4$ , and  $A_5$ ) in the above equations ((5) and (6)) have been determined using the least square method and are compiled in Table 3 along

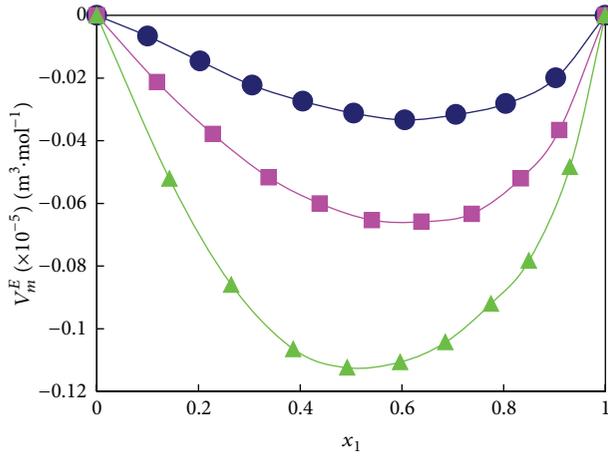


FIGURE 1: Variation of excess molar volume  $V_m^E$  with mole fraction  $x_1$  of (●), 1-butanol/(■), *iso*-butanol/(▲), and *t*-butanol with equimolar mixture of (DMA + EA).

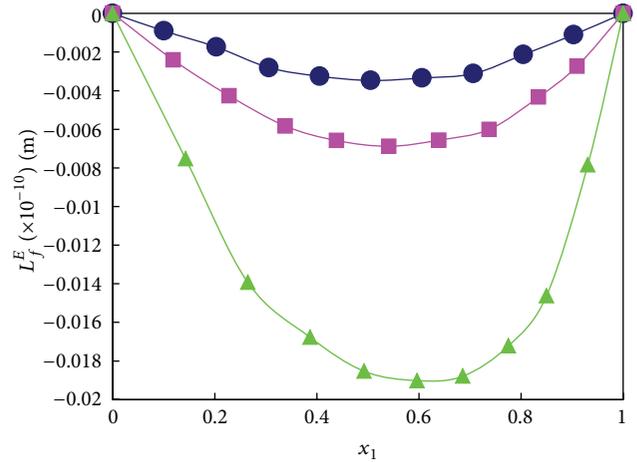


FIGURE 3: Variation of excess free length  $L_f^E$  with mole fraction  $x_1$  of (●), 1-butanol/(■), *iso*-butanol/(▲), and *t*-butanol with equimolar mixture of (DMA + EA).

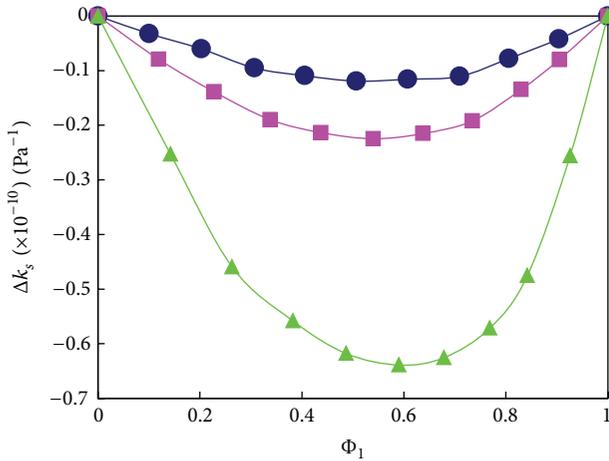


FIGURE 2: Variation of deviation in isentropic compressibility,  $\Delta k_s$ , with volume fraction,  $x_1$ , of (●), 1-butanol/(■), *iso*-butanol/(▲), and *t*-butanol with equimolar mixture of (DMA + EA).

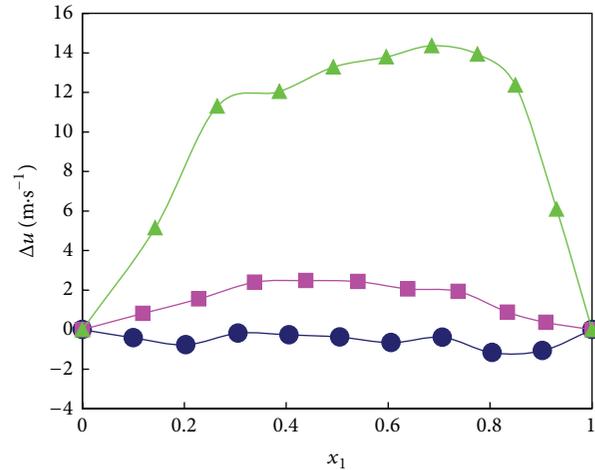


FIGURE 4: Variation of deviation in ultrasonic velocity,  $\Delta u$ , with mole fraction,  $x_1$ , of (●), 1-butanol/(■), *iso*-butanol/(▲), and *t*-butanol with equimolar mixture of (DMA + EA).

with the standard deviations  $\sigma(Y^E)$  calculated using the expression

$$\sigma(Y^E) = \left[ \sum \frac{(Y_{\text{exp}}^E - Y_{\text{cal}}^E)^2}{(m - n)} \right]^{1/2}, \quad (7)$$

where “ $m$ ” is the total number of experimental points (= 11) and “ $n$ ” is the number of coefficients (= 5) in (5).

The variations of these properties with mole fraction of 1-butanol/*iso*-butanol/*t*-butanol with equi molar mixture of (DMA + EA) are presented in Figures 1, 2, 3, 4, and 5. Negative values of  $V_m^E$ ,  $\Delta k_s$ , and  $L_f^E$  and positive values of  $\Delta u$   $Z_E$  have been observed in the present investigation mixtures. The factors that are mainly responsible for the expansion of molar volume, that is, positive values of  $V_m^E$ , are as follows [30, 31]. (i) Breaking one or both of the components in a solution, that is, loss of dipolar association between the molecules (dispersion

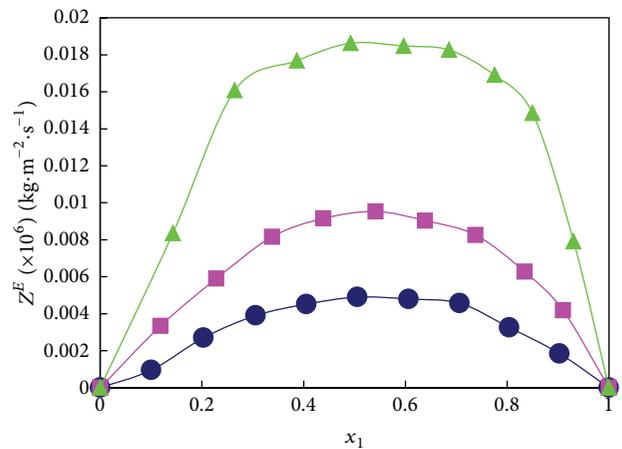


FIGURE 5: Variation of excess acoustic impedance,  $Z^E$ , with mole fraction,  $x_1$ , of (●), 1-butanol/(■), *iso*-butanol/(▲), and *t*-butanol with equi molar mixture of (DMA + EA).

forces). (ii) The geometry of molecular structures which does not favor the fitting of molecules of one component into the voids created by the molecules of other component. (iii) Steric hindrance of the molecules. The negative values of  $V_m^E$  [32] are due to strong specific interactions such as (iv) association of molecules through the formation of hydrogen bond (or) association due to dipole-dipole interactions and (v) accommodation of molecules due to considerable differences in molar volumes. The variation of excess molar volume in the present investigation is negative over the entire mole fraction range [33]. As we know, the molecules of alkanols are self-associated through hydrogen bonding in pure state [1]. Mixing of (DMA + EA) with alkanols would induce mutual dissociation of hydrogen bonded structures present in the pure alkanols with subsequent formation of new hydrogen bonds (O...H-O-) between carbonyl group (-C=O) of amide/acrylate molecules and hydroxyl group (-OH) of alcohol groups leading to a contraction in volume and decrease in isentropic compressibility of the mixtures. The magnitude of negative  $V_m^E$ ,  $\Delta k_s$ , and  $L_f^E$  values follows the order 1-butanol < *iso*-butanol < *t*-butanol which indicates the order of the interactions between the components of the mixtures. In general, the negative values of deviation in isentropic compressibility indicate strong and specific interactions such as H-O and -dipole interactions; on the other hand, the positive values of deviation in isentropic compressibility indicate weak interactions and dispersion forces operating between the molecules of the components of the mixtures [34, 35].

Further, it is interesting to note that interaction is more in (DMA + EA) + *t*-butanol mixture than in (DMA + EA) + *iso*-butanol mixture. This is due to the presence of three -CH<sub>3</sub> groups at the  $\alpha$ -carbon atom of *t*-butanol, which increase the electron density at oxygen atom to greater extent than that in *iso*-butanol, which has two -CH<sub>3</sub> groups at  $\beta$ -carbon atom, resulting in the strong interaction in *t*-butanol mixture. Hence, the interaction between (DMA + EA) and alkanols increases when the -OH group is attached to more number of -CH<sub>3</sub> groups. The variation of  $\Delta u$  and  $Z^E$  with composition of mixture is displayed in Figures 4 and 5, respectively. From these figures,  $\Delta u$  and  $Z^E$  values are positive and become more positive with increasing of -CH<sub>3</sub> groups attached to -OH group of alkanols. It has been also observed that the strength of interaction in  $\Delta u$  and  $Z^E$  is the same as that of  $V_m^E$ ,  $\Delta k_s$ , and  $L_f^E$  for all the systems. Furthermore, DMA and alkanol molecules having large dipole moments, causing dipole-dipole and dipole-induced dipole interactions, also exist between the liquid mixtures, which is also favorable to strong interactions between the liquid molecules.

In the present study, theoretical sound velocities have been evaluated by considering (DMA + EA) as one component and alkanols as the other component in the mixture. Such an evaluation of theoretical sound velocity is useful to verify the applicability of various postulates of the theories of liquid mixtures and to arrive at some useful inferences regarding the strength of molecular interactions between component liquids in some cases. The theoretical values of ultrasonic velocity obtained using various theories along with the experimental velocity are summarized in Table 4.

Nomoto [36] established the following relation with sound velocity based on the assumption of the linearity of the molecular sound velocity and the additivity of molar volume:

$$u_N = \left\{ \frac{(\sum x_i R_i)}{(\sum x_i V_i)} \right\}^3, \quad (8)$$

where  $x_i$  is the mole fraction,  $R_i = u_i^{1/3} V_i$  is the molar sound velocity,  $V_i$  is the molar volume, and  $u_i$  is the sound velocity of the  $i$ th component.

Van Dael [37] obtained the ideal mixture relation

$$\sum \left( \frac{x_i M_i}{u_i^2} \right) = \left\{ \frac{1}{\sum x_i M_i} \right\} \left\{ \frac{1}{u_V} \right\}^2, \quad (9)$$

where  $M_i$  is the molecular weight of  $i$ th component in the liquid mixture.

Impedance dependence relation used by Baluja and Parshania [38] is given as

$$u_{\text{imp}} = \frac{\sum x_i Z_i}{\sum x_i \rho_i}, \quad (10)$$

where  $Z_i$  is the acoustic impedance and  $\rho_i$  is the density of the  $i$ th component of the mixture.

Junjie's [39] equation is given as

$$u_{\text{jun}} = \left\{ \frac{\sum x_i V_i}{(\sum x_i M_i)^{1/2}} \right\} \left\{ \sum \left( \frac{x_i V_i}{\rho_i u_i^2} \right) \right\}^{-1/2}. \quad (11)$$

Jacobson's [40] equation is given as

$$u_J = K \rho^{-1/2} L_f^{-1}, \quad (12)$$

where  $K$  is Jacobson temperature dependent constant and is equal to  $K = (93.875 + 0.375T) \times 10^{-8}$ ,  $T$  is absolute temperature, and  $L_f$  is the ideal free length of the mixture given by  $L_f = (x_1 L_{f1} + x_2 L_{f2})$ .

Rao's (specific sound velocity) [41] relation is given as

$$u_R = \left( \sum x_i r_i \rho \right)^3, \quad (13)$$

where  $r_i = u_i^{1/3} / \rho_i$  is the Rao's specific sound velocity of the  $i$ th component of the mixture.

The experimental data have been fitted to a polynomial; it describes the ultrasonic velocity data quantitatively as well as qualitatively even in the specific interaction predominant region where nonideal behavior of the system is noticed.

The polynomial equation is [42]

$$f(x) = u(x) = \sum a_k x^k, \quad (14)$$

where  $k$  in the summation assumed values from 0 to 3,  $x$  is the mole fraction of the 1-butanol/*iso*-butanol/*t*-butanol, and  $a_k$ , is constant coefficient to be determined using numerical methods. The values of sound velocities (after determining the coefficients in the above polynomial equation by applying least squares method) have been compiled in Table 4.

TABLE 4: Theoretical values of ultrasonic velocity from (8) to (13) and polynomial equation (14) with mole fraction,  $x_1$ , of 1-butanol/*iso*-butanol/*t*-butanol at 308.15 K.

$x_1$	$u_N$	$u_V$	$u_{Imp}$	$u_{Jun}$	$u_J$	$u_R$	$f(x)$
(DMA + EA) + 1-butanol							
0.0000	1260.95	1260.95	1260.95	1260.95	1260.95	1260.95	1260.67
0.1003	1255.98	1250.91	1256.26	1253.98	1255.26	1268.06	1255.39
0.2031	1250.80	1241.86	1251.32	1247.20	1249.35	1274.03	1249.90
0.3059	1245.54	1233.97	1246.24	1240.80	1244.53	1277.70	1244.34
0.4056	1240.36	1227.39	1241.18	1234.95	1239.01	1277.67	1238.92
0.5056	1235.09	1221.79	1235.95	1229.44	1233.78	1275.13	1233.48
0.6061	1229.70	1217.13	1230.55	1224.28	1228.09	1269.55	1228.03
0.7065	1224.24	1213.42	1224.99	1219.50	1222.94	1259.95	1222.63
0.8043	1218.83	1210.69	1219.42	1215.21	1217.06	1247.97	1217.44
0.9029	1213.30	1208.79	1213.63	1211.26	1211.82	1230.96	1212.31
1.0000	1207.76	1207.76	1207.76	1207.76	1207.76	1207.76	1207.37
(DMA + EA) + <i>iso</i> butanol							
0.0000	1260.95	1260.95	1260.95	1260.95	1260.95	1260.95	1260.80
0.1186	1251.29	1243.76	1252.07	1247.65	1251.71	1270.84	1251.79
0.2282	1242.24	1229.87	1243.58	1236.16	1242.88	1276.04	1243.14
0.3379	1233.07	1217.71	1234.81	1225.39	1234.37	1277.62	1234.19
0.4387	1224.55	1207.97	1226.49	1216.12	1225.73	1275.28	1225.73
0.5411	1215.78	1199.36	1217.78	1207.32	1217.03	1268.83	1216.91
0.6387	1207.33	1192.32	1209.22	1199.49	1208.21	1258.38	1208.31
0.7375	1198.68	1186.27	1200.29	1192.12	1199.62	1244.89	1199.42
0.8342	1190.11	1181.37	1191.27	1185.43	1190.35	1225.46	1190.55
0.9098	1183.35	1178.21	1184.02	1180.58	1183.23	1206.56	1183.51
1.0000	1175.20	1175.20	1175.20	1175.20	1175.20	1175.20	1174.99
(DMA + EA) + <i>t</i> -butanol							
0.0000	1260.95	1260.95	1260.95	1260.95	1260.95	1260.95	1261.46
0.1429	1235.74	1219.73	1239.24	1224.71	1240.85	1268.19	1240.77
0.2647	1214.20	1190.14	1219.89	1197.39	1225.26	1267.26	1223.11
0.3867	1192.60	1164.74	1199.70	1172.89	1204.43	1257.64	1204.83
0.4924	1173.86	1145.68	1181.49	1153.72	1186.88	1242.11	1188.10
0.5965	1155.38	1129.25	1162.89	1136.55	1168.95	1221.42	1170.45
0.6857	1139.53	1116.84	1146.38	1123.07	1153.72	1200.01	1154.14
0.7752	1123.62	1105.80	1129.26	1110.60	1137.42	1174.50	1136.45
0.8498	1110.35	1097.59	1114.56	1100.98	1122.62	1151.22	1120.54
0.9306	1095.96	1089.66	1098.15	1091.31	1102.03	1119.18	1101.96
1.0000	1083.60	1083.60	1083.60	1083.60	1083.60	1083.60	1084.75

The root mean square deviation (RMSD) ( $\sigma$ ) corresponding to sound velocity values calculated using the polynomial equation (14) from their experimental values has been evaluated using the relation

$$\sigma = \left\{ \sum \frac{(u_E - u_P)^2}{n} \right\}^{1/2}, \quad (15)$$

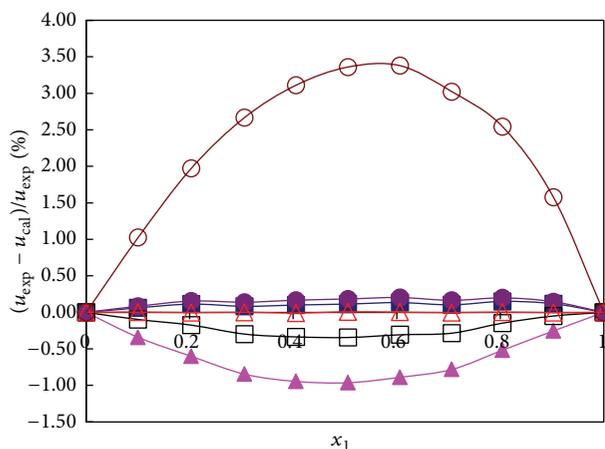
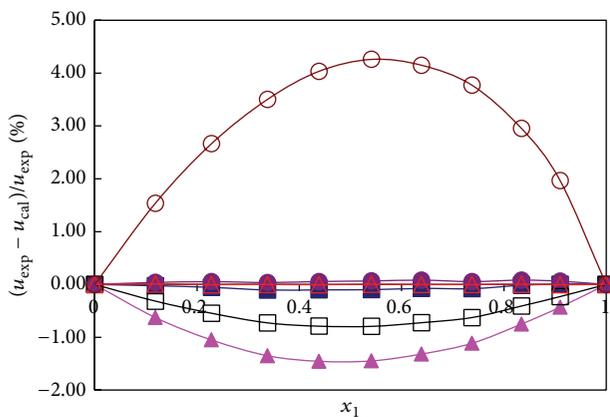
where  $u_E$  is the experimental sound velocity,  $u_P$  is the calculated sound velocity from the polynomial equation  $f(x)$ , and  $n$  is the number of mole fractions at which experimental and theoretical velocities have been determined. These standard

deviations which are small and coefficients obtained from (14) are presented in Table 5.

The percentage of deviations of theoretical velocities from the experimental ultrasonic velocity values are plotted in Figures 6, 7, and 8 for all the systems investigated. Data from Table 5 and from Figures 6, 7, and 8 reveal that sound speed computed from Jacobson's relation exhibits more satisfactory agreement with the experimental values. The interdependence of intermolecular free length and velocity is given by Jacobson relation. The small percentage deviation of Jacobson relation provides the better estimate of the ultrasonic velocity values.

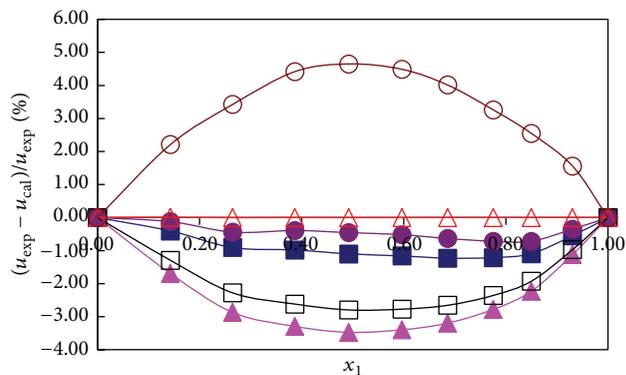
TABLE 5: Parameters and root mean square deviation (RMSD),  $\sigma$ , of ultrasonic velocity;  $u$  calculated using polynomial equation  $f(x)$ .

Name of the System	$a_0$	$a_1$	$a_2$	$a_3$	$\sigma$ (m·s <sup>-1</sup> )
(DMA + EA) + 1-butanol	1260.67	-52.10	-5.54	4.34	0.324
(DMA + EA) + <i>iso</i> -butanol	1260.80	-74.44	-13.50	2.13	0.178
(DMA + EA) + <i>t</i> -butanol	1261.46	-146.66	19.82	-49.87	1.190

FIGURE 6: Percentage deviations of theoretical ultrasonic velocities with mole fraction,  $x_1$ , of 1-butanol with equimolar mixture of (DMA + EA); Nomoto (■), van Dael (▲), impedance (●), Junjie (□), Jacobson (△), Rao's (○).FIGURE 7: Percentage deviations of theoretical ultrasonic velocities with mole fraction,  $x_1$ , of *sec*-butanol with equimolar mixture of (DMA + EA); Nomoto (■), van Dael (▲), impedance (●), Junjie (□), Jacobson (△), Rao's (○).

## 4. Conclusions

- (i) Ultrasonic velocities,  $u$ , and densities,  $\rho$ , of mixtures of 1-butanol or *iso*-butanol or *t*-butanol with equimolar mixture of (*N,N*-dimethylacetamide + Ethyl acrylate) over the entire composition range have been measured at  $T = 308.15$  K.
- (ii) Using the experimental results, deviations in ultrasonic velocity,  $\Delta u$ , and isentropic compressibility,  $\Delta k_s$ ,

FIGURE 8: Percentage deviations of theoretical ultrasonic velocities with mole fraction,  $x_1$ , of *t*-butanol with equimolar mixture of (DMA + EA); Nomoto (■), van Dael (▲), impedance (●), Junjie (□), Jacobson (△), Rao's (○).

excess molar volume,  $V_m^E$ , excess intermolecular free length,  $L_f^E$ , and excess acoustic impedance,  $Z^E$ , have been calculated and these properties with composition of the mixtures have been discussed in terms of molecular interactions in these mixtures.

- (iii) The deviation/excess properties have been fitted to Redlich-Kister type polynomial and the corresponding standard deviations have been calculated.
- (iv) Negative values of  $V_m^E$ ,  $\Delta k_s$ , and  $L_f^E$  and positive values of  $\Delta u$  and  $Z^E$  are observed over the entire composition range. The observed negative and positive values of deviation/excess properties are attributed to the strong interactions between the unlike molecules of the mixtures.
- (v) The strength of interaction in the mixtures follow the order (DMA + EA) + *t*-Butanol > *iso*-Butanol > 1-Butanol.
- (vi) Further theoretical values of sound velocity in the mixtures have been evaluated using various theories and have been compared with experimental sound velocities to verify the applicability of such theories to the systems studied. Theoretical ultrasonic velocity data has been used to study molecular interactions in the systems investigated.

## Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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