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A Theoretical and Experimental Approach to Ultrasonic Velocities, Densities, and Refractive Indices in Benzene and Aniline Mixture

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Abstract

The study investigates the theoretical velocities of binary liquid mixtures comprising Aniline and Benzene under 2MHz ultrasonic waves at temperatures of 303.15K, 308.15K, and 313.15K, examined in relation to mole fraction. Experimental data is compared with various theoretical models, including Nomoto theory (NOM), Ideal mixing relation (IMR), Impedance Relation (IR), Rao's Specific Velocity Method (R), and Junjie's relations (JR). The work reflects the changes in refractive index with varying liquid density across different mole fractions, explaining observed impacts on ultrasonic velocities. To assess the goodness of fit, Chi-square tests and average percentage errors are employed, allowing for an evaluation of the relative applicability of these theories within the studied systems. The study also explores how the thermos-acoustical characteristics of the systems evolve concerning the mole fraction of a common molecule, shedding light on molecular interactions. Furthermore, various parameters for the binary mixture, such as discrepancies in ultrasonic velocity, excess isentropic compressibility, surplus acoustic impedance, excess intermolecular free length, and molar refraction deviation, are computed using experimental data of density, refractive index, and ultrasonic velocities at different mole fractions of benzene in the aniline-benzene mixture. These calculations serve to reflect the intermolecular interactions present in the binary liquid mixture.

Key words : Ultrasonic Velocity, Refractive Index, Binary Mixture, Thermoacoustic Parameters, Aniline, Benzene.

Introduction

In determining the Physio-chemical behaviour of liquid mixtures, speeds of sound data is very important and it has many applications in the industry. A speed of sound investigations has been carrying by many researchers[1-4] from several years. Ultrasonic velocities in liquid mixtures have been calculated and compared with experimental values using various theories[5,6]. Measurement of ultrasonic velocity gives the valuable information about the Physico-chemical behaviour of liquids and their mixtures[7,8]. Comparison of evaluated theoretical velocities with those obtained experimentally is expected to reveal the nature of interaction between component molecules in the mixtures. Such theoretical study is useful in defining a comprehensive theoretical model for a specific liquid mixture. They are also valuable in testing various theories of liquid state. Many researchers compared the experimental values of ultrasonic velocities with theoretically evaluated values for organic liquid mixtures using different theories Nomoto theory[9], Ideal mixing relation[10], Impedance relation[11], Rao's specific velocity theory[12] and Junjie's relation[13]. The goal of the present investigation is to compare the experimentally determined ultrasonic sound velocity in binary liquid mixtures with those with different theories like Nomoto, Ideal mixing relation, Impedance relation, Rao's specific velocity and Junjie's at temperatures 303.15K, 308.15K,313K. In the present Investigation Aniline in Benzene are used as binary liquid mixtures to study the extent of interactions between the dissimilar molecules. The results are explained and discussed in terms of intermolecular interactions occurring in these binary systems. The deviation in the variation of average percentage error, (APE) and Chi-square test for goodness of fit, from unity have also been evaluated to further explain the non-idealist system.

Additionally, the semi-empirical formula, given by Gladstone-Dale (GD), Wiener (W), and Lorentz-Lorenz for predicting the refractive index in binary mixtures has been applied in this study. A thorough examination has been conducted on the experimental and theoretical values of refractive indices at different temperatures. The outcomes have been analyzed through x2 fitting utilizing Origin software and are discussed in detail.

Theory :

(a) The theoretical methods used for analysis of ultrasonic velocities is binary liquid mixture and their equation are as following

1. Nometo established an empirical relation for ultrasonic velocity is binary liquid mixture as:

$$U_{Nom} = \left(\frac{x_1R_1 + x_2R_2}{x_1V_1 + x_2V_2}\right)^3 \quad (1)$$

$where\ R = \frac{Mu^{\frac{1}{3}}}{\rho_1}$ Molecular sound velocities. X_1 and X_2 are the mole fraction of first and second compound of the liquid mixture and V is molar volume.

2. *Impedance dependent relation :*

$$U_{IDR} = \sum XiZi \Big/ \sum Xi\rho i \tag{2}$$

Where X_i is the mole fraction, ρ_i is the density of the mixture and Zi is the acoustic impedance.

3. Rao's specific velocity

$$U_{Rao} = \sum (x_i\, r_i \rho_i)^3 \tag{3}$$

Xi is the mole fraction, ρ_i is the density of the mixture.

4. *Zhang-Junjie relation :*

$$U_{JR} = \frac{x_1 v_1 + x_2 v_2}{(x_1 M_1 + x_2 M_2)^{\frac{1}{2}}} \left[\frac{x_1 v_1}{\rho_1 v_1^2} + \frac{x_2 v_2}{\rho_2 v_2^2}\right]^{-1/2} \tag{4}$$

Where X_1 and X_2 is the mole fractions first and second components of mixture, ρ_1 and ρ_2 is the densities of the constituents components

5. *Van Dael and Bold Vangeel ideal mixing relations :*

$$U_{VDV} = \left(\frac{x_1}{M_1 V_1^2} + \frac{x_2}{M_2 V_2^2}\right)^{-\frac{1}{2}} (x_1 M_1 + x_2 M_2)^{-1/2} \tag{5}$$

Where M_1 and M_2 are molecular weight of constituent's components, v_1 and v_2 are the ultrasonic velocities of individual components. The velocities of the above theories are evaluating the ultrasonic velocities is verified by various first determination, which would help us in finding the appropriate theory applicable the mixture under study. The following are the adopted test determination for the binary under study.

6. *Chi-square test for goodness of fit :*

According to Karl Pearson chi-square value is evaluated for binary liquid mixture under study using the formula

$$\chi^2 = \sum_{i=1}^{n} \frac{(U_{mix\,(Obs)} - U_{mix\,(cal)})^2}{(U_{mix\,(cal)})} \tag{6}$$

Where n is the number of data used.

Average percentage error

$$APE = \frac{1}{n} \sum \frac{[U_{mix\ (Obs)} - U_{mix\ (cal)}]}{(U_{mix\ (Obs)})} * 100 \tag{7}$$

(b) Semi-imperical formula of refrective index of binary mixture

1. Gladstone-Dale (GD) formula for calculating refractive index of binary mixture,

$$(N_m\text{-}1)= \varphi_1(n_1\text{-}1)+ \varphi_2(n_2\text{-}1) \quad (8)$$

2. *Wiener (W) formula is given by,*

$$\left\{\frac{n_m^2 - n_1^2}{n_m + 2n_1^2}\right\} = \left\{\frac{n_2^2 - n_1^2}{n_2^2 + 2n_1^2}\right\} \text{ø} \quad (9)$$

3. L**orentz-Lorenz** formula with principle of molecular polarizability with volume fraction of mixture,

$$\left\{\frac{n_m^2 - 1}{n_m^2 - 2}\right\}\frac{1}{\rho_m} = \left\{\frac{n_1^2 - 1}{n_1^2 - 2}\right\}\frac{w_1}{\rho_1} + \left\{\frac{n_2^2 - 1}{n_2^2 - 2}\right\} \quad (10)$$

Where symbols have their usual meaning.

Furthermore various important parameter for binary mixture in this study has been calculated which include discrepancies in ultrasonic velocity (Δu), excess isentropic compressibility (Δk_S), surplus acoustic impedance (Z^E), excess intermolecular free length (L^E_f), and molar refraction deviation (ΔR_m) were assessed using the provided equations.

$$\Delta u = u_m - (x_1u_1+x_2u_2) \quad (11)$$

In the given equation, Δu represents the change in ultrasonic velocity, u_m signifies the speed in the mixture, and x_1, x_2 denote the mole fractions of the liquids in the mixture. Additionally, u_1 and u_2 stand for the corresponding velocities in the pure liquids, respectively.

$$\Delta k_s = \frac{1}{u_m^2 \rho_m} - \left\{\frac{1}{u_1^2 \rho_1} + \frac{1}{u_2^2 \rho_2}\right\} \quad (12)$$

Where ρ_1 and ρ_2 represents density of individual liquids and ρ_m is density of mixture rest of symbols have their usual meaning.

$$Z^E = (\rho_m u_m) - (x_1\rho_1u_1 + x_2\rho_2u_2) \quad (13)$$

$$L_f^e = \frac{K}{(u_m^2 \rho_m)^{1/2}} - \left[\frac{x_1K}{(u_1^2 \rho_1)^{1/2}} + \frac{x_2K}{(u_2^2 \rho_2)^{\frac{1}{2}}}\right] \quad (14)$$

$$\Delta R_m = R_m^{expt} - R_m^{id} \quad (15)$$

Where $R_{fm}^{expt} = \left(\frac{n_m^2-1}{n_m^2+2}\right)\left(\frac{x_1M_1+x_2M_2}{\rho_m}\right)$ and

$$R_m^{id} = \left[\left(\frac{n_1^2 - 1}{n_1^2 + 2}\right)\frac{M_1}{\rho_1}\emptyset_1 + \left(\frac{n_2^2 - 1}{n_2^2 + 2}\right)\frac{M_2}{\rho_2}\emptyset_2\right]$$

Where M_1,M_2,n_1,n_2,ρ_1,ρ_2, u_1 and u_2 represents the molecular weight, refractive index, density, and ultrasonic velocity, respectively, of the pure components respectively. Also, ρ_m, n_m and u_m represent measured density, refractive index and ultrasonic velocity of the mixtures. K,γ,α, and T denote Jacobson's constant, heat capacity ratio, coefficient of linear expansion and absolute temperature,

respectively.

Results and Discussions

Comparative study of theoretical values of ultrasonic velocities and which are obtained experimentally in the present liquid mixtures is explained in terms of the nature of interaction between the component molecules in the mixture. Theoretical study is useful in building the comprehensive theoretical model for the liquid mixtures. The theoretical values of ultrasonic velocities calculated by using the above all equations. The experimental values of ultrasonic velocity for the binary system, Aniline with Benzene compared with various theoretical values along with the values of average percentage error and chi-square test values at temperatures 303.15K, 308.15K and 313.15K are presented in Table-1,2,3.

Table 1. Represents data sheet of theoretical calculated and experimentally obtain velocity of ultrasonic waves as mole fraction of Benzene increases in mixture at 303.15K

Temperature 303.15 K						
Mole fraction of Benzene	U_{EXT}	U_{NOM}	U_{IR}	U_{RAO}	U_{JN}	U_{IMR}
0	1264	1264.5	1264	1264	1264	1264
0.1007	1281	1286.6	1304.3	1296.5	1284	1278
0.2014	1331	1327	1343	1329.7	1305.6	1326
0.3019	1368	1361	1380.5	1363	1331	1348
0.4023	1402.8	1398.3	1417	1398	1359	1386
0.5023	1440	1433	1452.3	1432	1390	1426
0.623	1472	1468.4	1487	1467	1425.5	1461
0.721	1513	1503.6	151.9	150	1464	1503
0.8017	1545	1539.6	1550.9	1539	1507	1540
0.901	1576.8	1576	1582	1576	1571	1571
1	1612.8	1612	1612.8	1612.8	1612.8	1612.8
APE		0.23	-0.67	0.16	1.85	0.57
χ^2		0.316	1.096	0.373	8.88	0.829

Table 2. Represents data sheet of theoretical calculated and experimentally obtain velocity of ultrasonic waves as mole fraction of Benzene increases in mixture at 303.15K

Temperature 308.15 K						
Mole fraction of Benzene	U_{EXT}	U_{NOM}	U_{IR}	U_{RAO}	U_{JN}	U_{IMR}
0	1256	1256.9	1256	1256	1256	1256
0.1025	1290	1289.8	1296	1289.3	1277	1284
0.2043	1323.6	1328	1334.4	1322.8	1300	1303.6
0.3057	1357.2	1363.7	1372	1357.5	1325	1347
0.4065	1386	1393	1408.6	1392	1354	1378.4

0.5069	1426.4	1428	1444.3	1426	1385.6	1408.5
0.6066	1455.6	1463.6	1478.6	1462	1420.7	1448.9
0.7057	1494	1499	1512.3	1498	1460	1481.4
0.8043	1533.6	1535	1545	1534	1505	1534
0.9024	1567	1571.5	1577	1570.6	1553.8	1562
1	1608	1607.7	1608	1608	1608	1608
APE		-0.25	-0.85	-0.11	1.6	0.54
X^2		0.39	1.62	5.6	0.073	0.838

Table 3. Represents data sheet of theoretical calculated and experimentally obtain velocity of ultrasonic waves as mole fraction of Benzene increases in mixture at 313.15K.

Temperature 313.15

Mole fraction of Benzene	U_{EXT}	U_{NOM}	U_{IR}	U_{RAO}	U_{JN}	U_{IMR}
0	1259.6	1260	1259.6	1259.6	1259.6	1259.6
0.1009	1276	1272	1301.3	1293	1277	1273
0.2017	1304.4	1301.1	1341.8	1328	1293.5	1311
0.3027	1404	1398	1381	1386	1330	1398.6
0.4031	1447	1442.3	1419	1428	1360.3	1438
0.5030	1474	1464	1455.3	1443	1391	1463
0.6030	1502	1498	1491	1481	1429	1499.2
0.7026	1558	1537	1525.3	1532	1465.9	1547
0.8017	1561	1546	1559	1547	1514.8	1556
0.9014	1589	1565.7	1591.6	1585.7	1563.7	1581
1	1623	1622.9	1623	1623	1623	1623
APE		0.56	0.31	0.57	3	0.3
χ^2		0.911	3.498	2.628	24.74	0.33

Table 4. Represents the refractive index vs density data sheet as the mole fraction of benzene increases in the mixture

303.15K		308.15K		313.15K	
Density (ρ)	Refractive Index (R.I.)	Density (ρ)	Refractive Index (R.I.)	Density (ρ)	Refractive Index (R.I.)
1.025	1.583	1.022	1.58	1.014	1.582
1.006	1.522	1.012	1.554	0.998	1.573
0.994	1.501	0.992	1.549	0.995	1.531
0.985	1.492	0.979	1.536	0.978	1.491
0.968	1.488	0.962	1.533	0.97	1.469
0.953	1.481	0.944	1.53	0.959	1.453
0.938	1.479	0.928	1.519	0.953	1.44
0.923	1.472	0.907	1.502	0.931	1.434
0.902	1.434	0.899	1.499	0.908	1.433
0.88	1.431	0.879	1.493	0.883	1.422
0.868	1.423	0.867	1.437	0.861	1.381

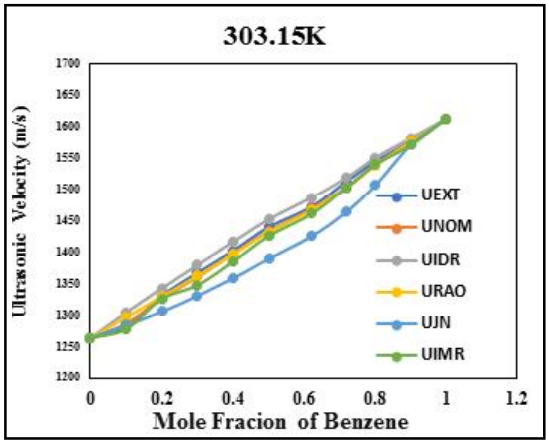

Figure 1 Represents the graph of theoretical and experimentally ultrasonic waves as mole fraction of Benzene increases in mixture at 303.15K (UEXT represents graph of experimental data and UNOM, UIDR, URAO, UJN and UIMR represents theoretical graph by various models discussed in previous section)

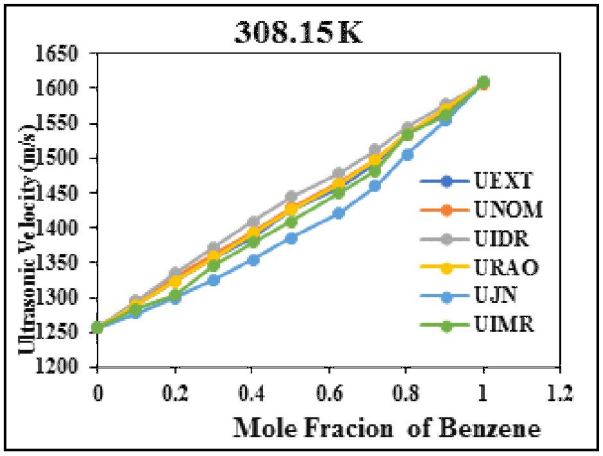

Figure 2 Represents the graph of theoretical and experimentally ultrasonic waves as mole fraction of Benzene increases in mixture at 308.15K (UEXT represents graph of experimental data and UNOM, UIDR, URAO, UJN and UIMR represents the oretical

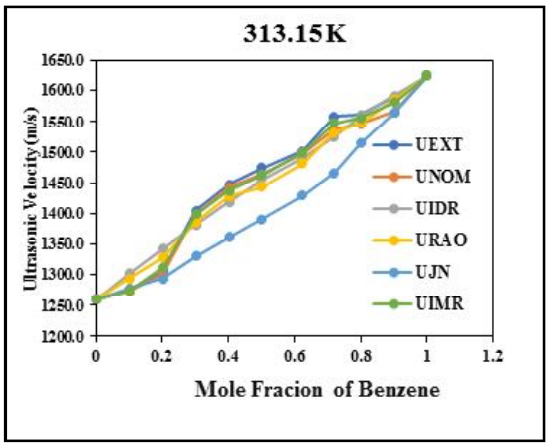

Figure 3 Represents the graph of theoretical and experimentally ultrasonic waves as mole fraction of Benzene increases in mixture at 303.15K (UEXT represents graph of experimental data and UNOM, UIDR, URAO, UJN and UIMR represents theoretical graph by various models discussed in previous section)

There is a deviation between experimental and theoretical values which confirm the existence of molecular interactions. Higher variation is observed at some intermediate concentrations suggesting the existence of strong tendency of association between component molecules as a result of hydrogen bonding. The perusal of the figures reveals good agreement between the experimental and calculated sound velocities, owing to the several assumptions and approximations made in the respective theories. It is observed from the Table-1,2,3 that Nomoto and Rao relation predicts a good agreement of sound

velocity with the experimental values in binary mixtures of Aniline with Benzene. The Junjie Relation and collision factor theory yields a fair estimation of sound velocity for Aniline in Benzene. Thus, from the comparison of experimental velocity values with theoretical models. Interactions between different components of the binary mixture were not taken into account in view of the assumption in Nomoto's theory, that there is no change in the volume on mixing. In case of ideal mixing relation, the ratios of specific heats of ideal mixtures and the volumes are also equal and hence no molecular interactions are considered. However, upon mixing the components, interactions between the molecules occur due to the presence of various types of forces like hydrogen bonding, charge transfer, dispersion forces and interactions of dipole-dipole and dipole-induced dipole. The deviations observed in case of theoretical values of speeds of sound from the experimental values[20-25], thus, indicate that there are molecular interactions between the unlike molecules[26]. The deviations of experimental values from theoretical values calculated using VanDael and Vangeel equation might be due to the compressibility of the component liquids in the present mixtures. The deviations of experimental values and calculated values from impedance relation and Rao's relation simply non-additively of acoustic impedance and Rao's velocity in the liquid mixtures. In case of Junjie's relation large deviations are observed in the system Aniline in Benzene. It is reveals that, the observed deviation of theoretical values of velocity from the experimental values shows that there are molecular interactions takes place. Theoretical results of ultrasonic velocities in liquid mixtures are showed, it is observed that out of all five theories Nomoto's theory gives best results followed by Impedance dependence relation in all the systems studied. Statistical analysis done by using average percentage error and chi-square test. These theories are tested considering percentage deviation and chi square test for goodness of fit. The chi-square value determines the overall validity of the theory. For Aniline + Benzene, IMR, NOM, IR, Rao, JR, relations are applicable for theoretical evaluation of ultrasonic velocity. IDR and IMR are not applicable for theoretical evaluation of ultrasonic velocity. Also having high value of APE in Free length theory as compared to other theories. For Aniline + Benzene, IDR, NOM, IMR, IR and R relation are applicable for theoretical evaluation of ultrasonic velocity. JR is not applicable for theoretical evaluation of ultrasonic velocity. Also having high value of APE in Junjie relation as compared to other theories. Hence the. Junjie relation is not applicable for theoretical evaluation of ultrasonic velocity. For Aniline+ Benzene, IDR, NOM, IMR, and R relation are applicable for theoretical evaluation of ultrasonic velocity. Also having high value of APE in Free length theory as compared to other theories. Hence the Junjie is not applicable for theoretical evaluation of ultrasonic velocity.

Figure 4 illustrates the change in refractive index with varying fluid density, achieved by increasing the mole fraction of benzene at different temperatures. The initial and final measured refractive index values align well with previously documented data. A predominantly linear increase is noted, interspersed with minor Zig-zag fluctuations, as the density of the benzene/aniline mixture undergoes alterations. The parameters presented in the subsequent sections are derived from these experimentally observed values, coupled with the discussion on ultrasonic velocities in the preceding section.

Figure 5 displays the computed disparities in ultrasonic velocity as the benzene fraction rises within the mixture, using standard values derived solely from the mixture fraction, velocities in the pure liquid, and experimentally recorded velocities. The variations are notably distinct across different

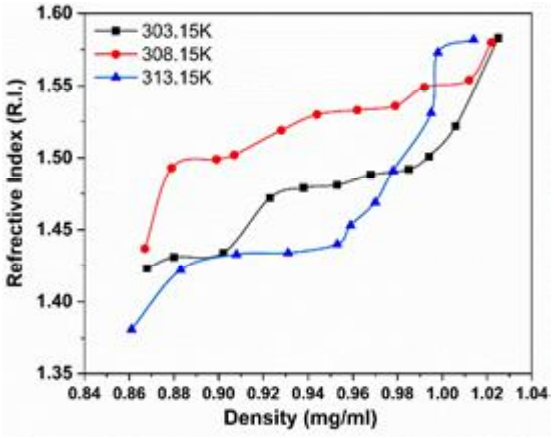

Figure 4 Represents the variation of refractive index as function of density of mixture and the density was varied by increasing mole fraction of benzene

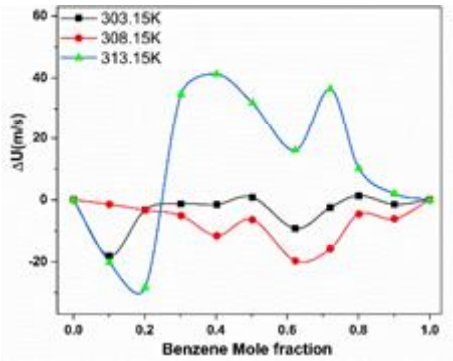

Figure 5 Shows discrepancies in ultrasonic velocity as the fraction of benzene increases in the mixture at various temperature

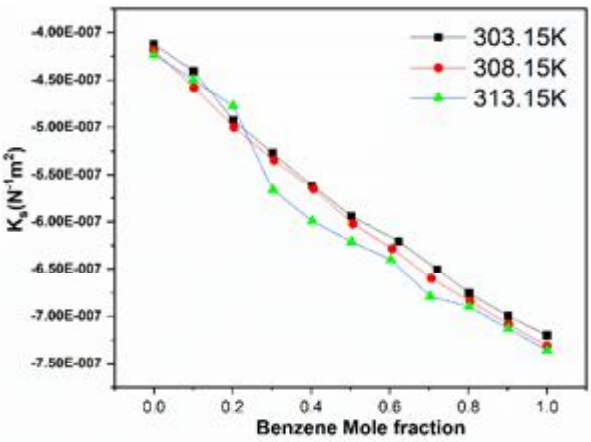

Figure 6 Shows excess isentropic compressibility as the fraction of benzene increases in the mixture at various temperature

temperatures, with the most substantial discrepancies observed at 313.15K. These disparities are likely attributed to the interactions between benzene and aniline in the mixture. This outcome effectively quantifies the effects of hese interactions through the utilization of ultrasonic velocities.

In Figure 6, we examine the fluctuations in excess isentropic compressibility corresponding to the variations in the fraction of benzene within the mixture at different temperatures. The observed trend reveals a systematic and linear reduction in isentropic compressibility across all the temperatures investigated. This implies that as the proportion of benzene increases, the excess isentropic compressibility consistently decreases, and this behavior is consistent across the entire range of measured temperatures.

The significant aspect here is the uniformity of this linear decrease across the temperature range. Regardless of the specific temperature being considered, the isentropic compressibility follows a linear downward trend as the fraction of benzene in the mixture rises. This suggests a robust and consistent relationship between the composition of the mixture and its isentropic compressibility, with a clear and predictable decrease in compressibility associated with an increase in the benzene fraction. The detailed analysis of these trends aids in a comprehensive understanding of the thermodynamic behavior of the mixture across varying compositions and temperatures.

In Figure 7, the surplus acoustic impedance is depicted as a function of the increasing fraction of benzene within the mixture across various temperatures. Notably, the surplus acoustic impedance registers zero at the extremes, corresponding to pure benzene and pure aniline. The deviation from zero is most prominent, particularly at a temperature of 313.15K. This observation implies that the interaction properties between benzene and aniline in the mixture become more conspicuous as the temperature rises, with the maximum deviation occurring at a benzene fraction of 0.6.

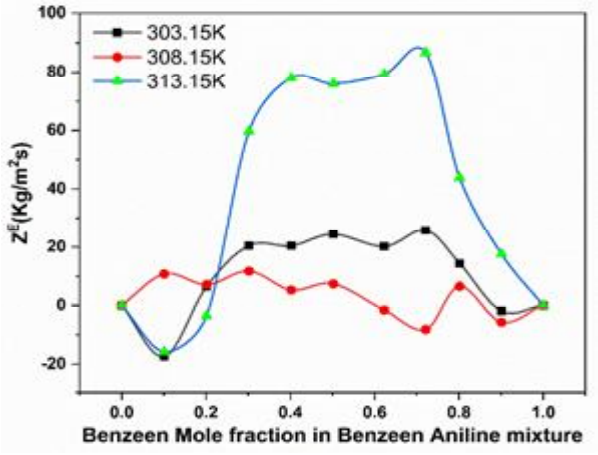

Figure 7 Shows surplus acoustic impedance as the fraction of benzene increases in the mixture at various temperature

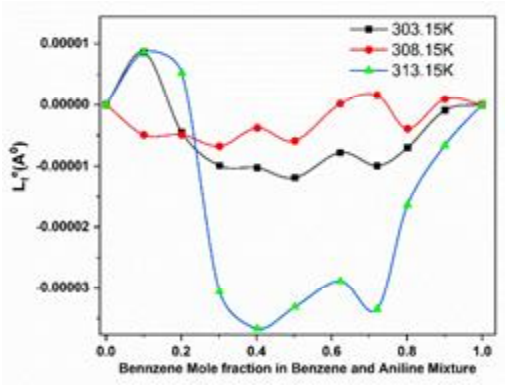

Figure 8 Shows excess intermolecular free length as the fraction of benzene increases in the mixture at various temperature

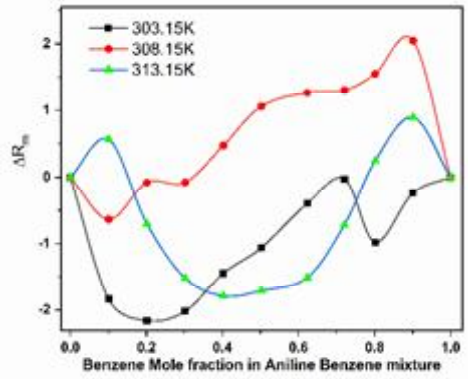

Figure 9 Shows molar refraction deviation as the fraction of benzene increases in the mixture at various temperature

The results suggest that the surplus acoustic impedance serves as a sensitive indicator of the interaction dynamics between benzene and aniline within the mixture. The temperature-dependent variations and the distinctive behavior at specific benzene fractions highlight the intricate interplay between these components. This detailed analysis enhances our understanding of how the surplus acoustic impedance reflects the underlying molecular interactions within the mixture, particularly emphasizing the impact of temperature and benzene fraction on these acoustic properties.

Figure 8 illustrates the excess intermolecular free length as a function of the increasing fraction of benzene in the mixture at various temperatures. The most pronounced variation is observed at 313.15K, and this aligns excellently with the data discussed in previous parameters. As the temperature increases, intermolecular interactions become more apparent, particularly in the mole fraction range of 0.4 to 0.7 for benzene. Conversely, no significant variations in excess intermolecular free length are observed at 303.15K and 308.15K. This underscores the temperature-dependent nature of intermolecular interactions, with the most notable effects occurring within specific temperature ranges.

Figure 9 displays the molar refraction deviation as a function of the increasing fraction of benzene in the mixture at various temperatures. A distinct reversal in trends is evident across different temperatures, as illustrated in the graph. Specifically, at 308.15K, the molar refraction deviation exhibits an almost linear increase with the rise in benzene mole fraction. However, at 303.15K and 313.15K, the variation is more dynamic, initially decreasing and then increasing. This non-linear trend in molar refraction at different temperatures suggests a temperature-dependent nature of intermolecular interactions in the mixture, influencing its refraction properties. The observed variations highlight the intricate and nuanced impact of temperature on the molar refraction behavior within the studied benzene-aniline mixture.

Conclusions

The speeds of sound values for the examined binary systems were both experimentally measured at temperatures of 303.15K, 308.15K, and 313.15K, and evaluated using various theoretical models. Additionally, various parameters for the binary mixture, including discrepancies in ultrasonic velocity, excess isentropic compressibility, surplus acoustic impedance, excess intermolecular free length, and molar refraction deviation, were computed using experimental data of density, refractive index, and ultrasonic velocities at different mole fractions of benzene in aniline-benzene mixture. The experimental speeds of sound values for the studied systems were compared with theoretically evaluated ultrasonic velocities in binary liquid mixtures, and the validity of different theories was assessed based on the experimental speeds of sound values. The study yielded satisfactory results with all the theories. Specifically, the average percentage deviation in the Aniline in Benzene system followed the order IDR < R < NOM < IMR < JR. Additionally, the Chi-square test for goodness indicated the order NOM < R < IMR < IDR < JR for the Aniline in Benzene system. The observed average percentage deviation of theoretical velocity values from experimental values was attributed to the presence of intermolecular interactions in the studied systems. The presence of intermolecular interactions is further substantiated by the various calculated parameters, providing additional justification for this study.

Future Scope :

The future scope of this study includes numerous potential directions for further study. These include outspreading the research to explore other binary liquid mixtures to establish the generalizability of findings, including additional theoretical models and molecular dynamics simulations to enrich understanding at the microscopic level, investigate higher-order interactions in ternary or multicomponent structures, applying the study's insights to optimize industrial processes, mainly in chemical engineering and pharmaceutical manufacturing, investigating the effects of temperature and pressure variations on ultrasonic properties, exploring dynamic properties such as relaxation processes or frequency-dependent behavior to deepen understanding of molecular interactions, and repeatedly refining experimental methods to capture a broader spectrum of activities and phenomena. Addressing these areas of examination will contribute to a more comprehensive understanding of ultrasonic behavior in binary liquid mixtures and its applied implications across scientific and industrial fields.

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