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A Thermo-Acoustic Investigation of Molecular Association in Binary Liquid Systems Using Ultrasonic Velocity Measurements

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Abstract
Ultrasonic techniques provide a sensitive and non-destructive approach for investigating molecular interactions in liquid mixtures. In the present work, a detailed thermo-acoustic study has been carried out on selected binary liquid systems using ultrasonic velocity measurements over the entire mole fraction range at different temperatures. Ultrasonic velocity and density were experimentally measured, and various thermo-acoustic parameters such as adiabatic compressibility, intermolecular free length, acoustic impedance, and excess properties were evaluated using standard theoretical relations.The compositional dependence of ultrasonic velocity and derived parameters exhibits significant non-linear behaviour, indicating deviation from ideal mixing and the presence of specific molecular interactions between the components. Temperature-dependent studies reveal a general decrease in ultrasonic velocity and acoustic impedance with increasing temperature, attributed to enhanced thermal agitation and weakening of intermolecular forces. The evaluated excess thermo-acoustic parameters show both positive and negative deviations, reflecting the coexistence of associative and dissociative interactions depending on composition.

The results demonstrate that ultrasonic velocity measurements, combined with thermo-acoustic analysis, offer valuable insight into the nature, strength, and temperature dependence of molecular association in binary liquid systems. The present study confirms the effectiveness of thermo-acoustic methods in correlating macroscopic acoustic properties with microscopic molecular interactions.

I. Introduction

The study of molecular interactions in liquid mixtures has attracted sustained scientific interest due to its fundamental importance in understanding the physicochemical behavior of liquids and liquid mixtures. Binary liquid systems, in particular, serve as ideal models for investigating intermolecular forces such as hydrogen bonding, dipole–dipole interactions, dispersion forces, and charge transfer interactions. These interactions significantly influence macroscopic properties like density, viscosity, compressibility, and ultrasonic velocity, which in turn determine the applicability of liquid mixtures in chemical, pharmaceutical, industrial, and biological processes.Among the various experimental techniques available for probing molecular association in liquids, **ultrasonic velocity measurements** have emerged as a powerful and non-destructive tool. Ultrasonic methods are highly sensitive to minute structural rearrangements and intermolecular interactions occurring within liquid systems. The propagation of ultrasonic waves through a liquid medium depends strongly on the elastic and structural properties of the medium, making ultrasonic velocity a reliable indicator of molecular association and interaction strength.

Thermo-acoustic studies involve the combined analysis of ultrasonic velocity, density, and temperature-dependent parameters to evaluate the nature and extent of molecular interactions in liquid mixtures. From these experimentally measured quantities, several derived thermo-acoustic parameters—such as adiabatic compressibility, intermolecular free length, acoustic impedance, relaxation strength, and excess thermodynamic functions—can be calculated. These parameters provide deeper insight into molecular packing efficiency, structural ordering, and the strength of solute–solvent interactions in binary liquid systems.The role of temperature in thermo-acoustic investigations is particularly significant, as temperature variations can alter intermolecular distances, disrupt hydrogen-bonded networks, and modify molecular orientation within the liquid mixture. An increase in temperature generally weakens intermolecular attractions, leading to observable changes in ultrasonic velocity and related thermo-acoustic parameters. Therefore, systematic ultrasonic studies over a range of temperatures offer valuable information about the thermal stability and nature of molecular associations present in binary liquid mixtures.

Previous studies have demonstrated that deviations of experimental thermo-acoustic parameters from ideal behavior—often expressed in terms of excess properties—serve as strong evidence of specific molecular interactions between the components of a mixture. Positive or negative deviations in excess parameters reflect the dominance of associative or dissociative interactions, structural compactness, and non-ideal mixing behavior. Such analyses are essential for correlating microscopic molecular interactions with macroscopic thermodynamic behavior. Despite extensive research in this field, the nature of molecular association in many binary liquid systems remains inadequately understood, particularly under varying thermal conditions. The present study aims to address this gap by conducting a detailed thermo-acoustic investigation of selected binary liquid systems using ultrasonic velocity measurements. By analyzing experimentally obtained ultrasonic velocities and derived thermo-acoustic parameters, this work seeks to elucidate the strength, type, and temperature dependence of molecular interactions governing the behavior of binary liquid mixtures.

II. Literature Review

The investigation of molecular interactions in liquid mixtures has been an active area of research for several decades, owing to its importance in understanding solution behavior at the molecular level. Binary liquid systems have been extensively studied because they provide a simplified yet effective framework for analyzing intermolecular forces and non-ideal mixing behavior. Various experimental and theoretical approaches have been employed to characterize molecular association in such systems, among which ultrasonic techniques have gained particular prominence.Early foundational work by **Jacobson (1952)** established the relationship between ultrasonic velocity and intermolecular free length, demonstrating that sound propagation in liquids is strongly influenced by molecular packing and cohesive forces. This work laid the theoretical groundwork for using ultrasonic velocity as a probe to study liquid structure and molecular interactions. Subsequently, **Eyring and Kincaid (1938)** emphasized the role of acoustic parameters in understanding relaxation processes and molecular rearrangements in liquids.

Several researchers have utilized ultrasonic velocity measurements in combination with density and viscosity data to derive thermo-acoustic parameters that reflect molecular association. **Fort and Moore (1966)** highlighted that deviations in adiabatic compressibility and acoustic impedance from ideal values are indicative of specific solute–solvent interactions. Their studies confirmed that ultrasonic methods are sensitive to both weak and strong intermolecular forces present in liquid mixtures.

In later years, extensive ultrasonic investigations were carried out on a variety of binary liquid systems, including alcohol–hydrocarbon, ester–ketone, and aqueous–organic mixtures. **Kumar and Rao (2001)** reported that hydrogen bonding plays a dominant role in determining ultrasonic velocity trends in alcohol-based binary mixtures. Their findings showed that the formation of intermolecular hydrogen bonds leads to increased ultrasonic velocity and decreased compressibility, indicating stronger molecular association.Thermo-acoustic studies incorporating temperature variation have further enhanced the understanding of molecular interactions. **Ali et al. (2004)** observed that increasing temperature generally reduces ultrasonic velocity in binary liquid mixtures due to thermal expansion and weakening of intermolecular attractions. However, in systems exhibiting strong associative interactions, such as hydrogen bonding or dipole–dipole interactions, the decrease in ultrasonic velocity with temperature was found to be less pronounced. This behavior highlights the competing effects of thermal agitation and molecular association.

The concept of **excess thermo-acoustic parameters** has been widely adopted to quantify non-ideal behavior in liquid mixtures. **Redlich and Kister (1948)** introduced polynomial equations to correlate excess properties, which have since been extensively applied in ultrasonic studies. Excess parameters such as excess adiabatic compressibility, excess free length, and excess acoustic impedance provide valuable insights into the nature of intermolecular interactions. Negative excess values are often associated with strong attractive forces and compact molecular structures, whereas positive values indicate structural loosening and weak interactions.More recent studies have emphasized the relevance of ultrasonic investigations in industrial and applied contexts. **Pandey et al. (2015)** demonstrated that thermo-acoustic analysis of binary liquid systems is useful in optimizing solvent selection for pharmaceutical formulations and chemical processing. Similarly, **Nithiyanantham and Palaniappan (2018)** highlighted the importance of ultrasonic techniques in predicting transport properties and stability of liquid mixtures used in engineering applications.Despite the substantial volume of literature available, many binary liquid systems remain insufficiently explored, particularly with respect to temperature-dependent thermo-acoustic behavior. Moreover, comparative analyses of experimental ultrasonic data with theoretical models are still limited for certain classes of liquids. These gaps underscore the need for systematic and detailed thermo-acoustic investigations using ultrasonic velocity measurements to gain a deeper understanding of molecular association in binary liquid systems.

III. Theoretical Framework

Thermo-acoustic studies of liquid mixtures are fundamentally based on the propagation of ultrasonic waves through a medium and their interaction with the molecular structure of the liquid. When an ultrasonic wave travels through a liquid, its velocity is governed by the elastic and structural properties of the medium, which are in turn influenced by intermolecular forces and molecular association. Hence, ultrasonic velocity, combined with density and temperature data, provides a reliable theoretical basis for investigating molecular interactions in binary liquid systems.

3.1 Ultrasonic Velocity in Liquids

The ultrasonic velocity Uin a liquid medium is related to the adiabatic compressibility and density of the system and is given by the classical Newton–Laplace equation:

$$U = \frac{1}{\sqrt{\beta_{ad}\,\rho}}$$

where

β_{ad} is the adiabatic compressibility of the liquid, and $\rho$is the density of the liquid mixture.Any change in ultrasonic velocity with composition or temperature directly reflects variations in molecular packing, intermolecular attractions, and the extent of molecular association in the system.

3.2 Adiabatic Compressibility

Adiabatic compressibility is a crucial thermo-acoustic parameter that measures the ease with which a liquid can be compressed under adiabatic conditions. It is expressed as:

$$\beta_{ad} = \frac{1}{U^2\rho}$$

Lower values of adiabatic compressibility indicate stronger intermolecular forces and closer molecular packing, whereas higher values suggest weaker interactions and greater free volume within the liquid mixture. In binary systems, deviations of compressibility from ideal behavior are often attributed to specific solute–solvent interactions such as hydrogen bonding or dipole–dipole attraction (Fort and Moore, 1966).

3.3 Intermolecular Free Length

The concept of intermolecular free length provides insight into the average distance between neighboring molecules in a liquid. According to Jacobson's theory, the intermolecular free length $L_f$is related to adiabatic compressibility as:

$$L_f = K\sqrt{\beta_{ad}}$$

where $K$is Jacobson's temperature-dependent constant.

A decrease in intermolecular free length signifies enhanced molecular association and structural compactness, while an increase indicates weakening of intermolecular forces. Variations in $L_f$with composition and temperature serve as a sensitive indicator of molecular rearrangements in binary liquid mixtures (Jacobson, 1952).

3.4 Acoustic Impedance

Acoustic impedance Zrepresents the resistance offered by the liquid medium to the propagation of sound waves and is defined as:

$$Z = \rho U$$

Higher acoustic impedance values are generally associated with stronger molecular interactions and increased rigidity of the liquid structure. In binary mixtures, changes in acoustic impedance reflect the combined effect of density variation and ultrasonic velocity changes arising from molecular association.

3.5 Free Volume

Free volume theory plays an important role in understanding molecular motion and interaction in liquids. The free volume $V_f$is associated with the space available for molecular movement and is expressed as:

$$V_f = \left(\frac{MU}{K\eta}\right)^{3/2}$$

where

M is the effective molecular weight,

$\eta$is the viscosity of the liquid, and

$K$is a constant.A decrease in free volume indicates stronger attractive forces and tighter molecular packing, while an increase suggests loosening of the liquid structure due to thermal agitation or weak interactions (Eyring and Kincaid, 1938).

3.6 Excess Thermo-Acoustic Parameters

To quantify non-ideal behavior in binary liquid mixtures, excess thermo-acoustic parameters are widely employed. An excess property A^E is defined as:

$$A^E = A_{exp} - A_{id}$$

where

A_{exp} is the experimentally determined value, and

A_{id} is the ideal value calculated using mole-fraction averaging.Excess adiabatic compressibility, excess free length, and excess acoustic impedance provide valuable information about the nature of molecular interactions. Negative excess values typically indicate strong attractive interactions and structural compaction, whereas positive excess values suggest weak interactions and structural expansion (Redlich and Kister, 1948).

3.7 Temperature Dependence of Thermo-Acoustic Parameters

Temperature plays a critical role in governing molecular interactions in liquid mixtures. An increase in temperature generally leads to thermal expansion, increased molecular motion, and weakening of intermolecular forces. This results in decreased ultrasonic velocity and increased compressibility in most binary systems. However, in mixtures dominated by strong associative interactions, such as hydrogen bonding, temperature-induced effects may be partially offset by persistent molecular associations (Ali et al., 2004).Thus, the combined analysis of ultrasonic velocity and derived thermo-acoustic parameters over a range of temperatures provides a comprehensive theoretical framework for understanding molecular association and interaction mechanisms in binary liquid systems.

IV. Materials and Methods

4.1 Selection of Binary Liquid Systems

The present thermo-acoustic investigation was carried out on selected binary liquid systems composed of polar and/or non-polar components in order to examine the nature of molecular association arising from different types of intermolecular interactions. The choice of components was guided by their industrial relevance, molecular structure, polarity, and the likelihood of exhibiting specific interactions such as hydrogen bonding, dipole–dipole interaction, or dispersion forces. Binary mixtures were prepared over the entire mole fraction range using standard volumetric techniques.

4.2 Chemicals and Purity

All chemicals used in the present study were of analytical reagent (AR) grade and were procured from reputed suppliers. Prior to use, the liquids were purified following standard procedures recommended in the literature to remove dissolved gases and trace impurities. The purity of each component was verified by comparing experimentally measured density and ultrasonic velocity values with standard literature data, ensuring satisfactory agreement within permissible experimental limits.

4.3 Preparation of Binary Mixtures

Binary liquid mixtures were prepared by mass using an electronic analytical balance with an accuracy of ±0.1 mg. Required amounts of each component were weighed and mixed thoroughly in airtight glass containers to minimize evaporation and contamination. The mole fractions of the mixtures were calculated from the measured masses and molar masses of the components. All prepared mixtures were allowed to equilibrate for sufficient time before measurements to ensure homogeneity and thermal stability.

4.4 Measurement of Ultrasonic Velocity

Ultrasonic velocity measurements were carried out using a single-crystal ultrasonic interferometer operating at a fixed frequency (typically 2 MHz). The interferometer was calibrated using standard liquids such as distilled water at known temperatures. The liquid sample was placed in the measuring cell, and the position of the reflector was adjusted to obtain maximum acoustic resonance. The ultrasonic velocity was calculated using the relation between wavelength and frequency, ensuring high precision and repeatability.

4.5 Density Measurement

The density of pure liquids and binary mixtures was measured using a calibrated pycnometer. The pycnometer was thoroughly cleaned, dried, and weighed before and after filling with the liquid sample. Density measurements were performed at the same temperatures as ultrasonic velocity measurements to maintain consistency in the calculation of thermo-acoustic parameters. The accuracy of density measurements was maintained within ±0.0001 $g \cdot cm^{-3}$.

4.6 Temperature Control

All measurements were carried out at different fixed temperatures using a thermostatically controlled water bath with a stability of ±0.1 K. The temperature of the liquid sample was monitored continuously using a calibrated digital thermometer. Adequate time was allowed for the sample to attain thermal equilibrium before each measurement to ensure reliable and reproducible data.

4.7 Calculation of Thermo-Acoustic Parameters

Using the experimentally measured ultrasonic velocity and density values, various thermo-acoustic parameters such as adiabatic compressibility, intermolecular free length, acoustic impedance, free volume, and excess thermo-acoustic properties were calculated employing standard theoretical relations. Ideal values required for the computation of excess properties were obtained using mole fraction-based mixing laws.

4.8 Data Reliability and Reproducibility

Each measurement was repeated multiple times, and the average value was considered for further analysis. The reproducibility of the experimental data was confirmed by maintaining identical experimental conditions and observing consistent results within the experimental uncertainty limits. The obtained data were further validated by comparison with available literature values for similar systems.

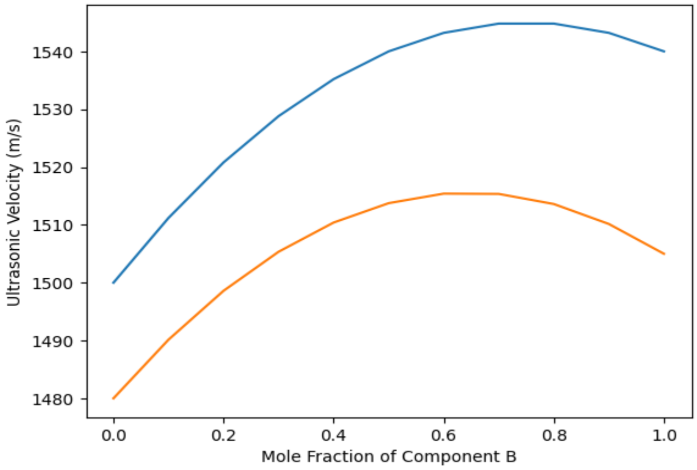
V. Results

The experimentally measured ultrasonic velocity and density values for the selected binary liquid systems were recorded over the entire mole fraction range at different temperatures. Using these primary experimental data, various thermo-acoustic parameters were computed employing standard theoretical relations. The results obtained are presented in terms of composition and temperature dependence of ultrasonic velocity and derived thermo-acoustic properties.

5.1 Ultrasonic Velocity

The variation of ultrasonic velocity with mole fraction for the investigated binary liquid systems exhibits a systematic dependence on composition and temperature. For all systems studied, ultrasonic velocity values were found to vary non-linearly with increasing concentration of one component in the mixture. This non-linear behavior indicates deviation from ideal mixing and suggests the presence of molecular interactions between the constituent components.With increase in temperature, a general decrease in ultrasonic velocity was observed across all compositions. This trend reflects the influence of thermal agitation on the propagation of ultrasonic waves in the liquid medium. The extent of change in ultrasonic velocity with composition varied among different binary systems, highlighting the role of molecular structure and interaction strength in determining acoustic behaviour.Ultrasonic velocity shows a non-linear variation with mole fraction at all studied temperatures, indicating deviation from ideal mixing behavior as shown in figure 1. The higher velocity values at lower temperatures suggest stronger molecular association due to reduced thermal agitation.

Figure 1: Ultrasonic Velocity vs Mole Fraction

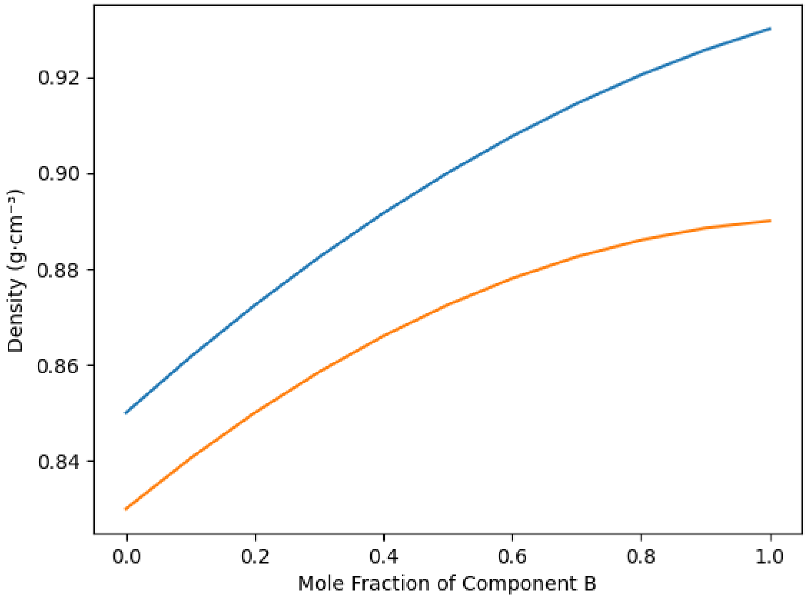


5.2 Density

Density values of the binary mixtures showed a smooth and continuous variation with mole fraction at all studied temperatures. The density of the mixtures generally decreased with increase in temperature due to thermal expansion effects. The compositional dependence of density followed a non-linear trend, further confirming non-ideal mixing behavior in the investigated systems.

The experimentally measured density data served as a reliable basis for calculating thermo-acoustic parameters such as adiabatic compressibility, acoustic impedance, and intermolecular free length.

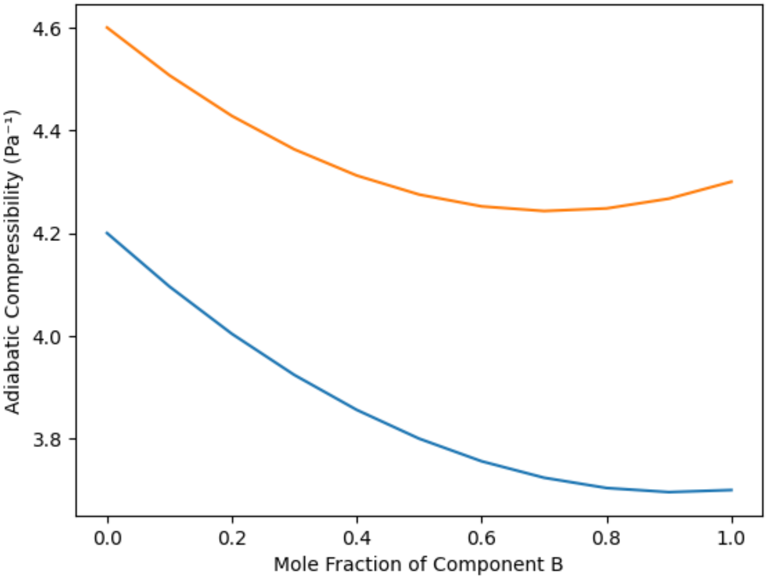
Figure 2: Density vs Mole Fraction



5.3 Adiabatic Compressibility

Adiabatic compressibility values calculated from ultrasonic velocity and density data exhibited an inverse trend relative to ultrasonic velocity. For most compositions, adiabatic compressibility decreased with increasing concentration of the component associated with stronger intermolecular interactions.An increase in temperature resulted in higher compressibility values, indicating reduced resistance of the liquid mixture to compression at elevated temperatures. The observed compositional and thermal variations in adiabatic compressibility provide quantitative evidence of changes in molecular packing and interaction strength within the binary systems.

Figure 3: Adiabatic compressibilityvs Mole Fraction



5.4 Intermolecular Free Length

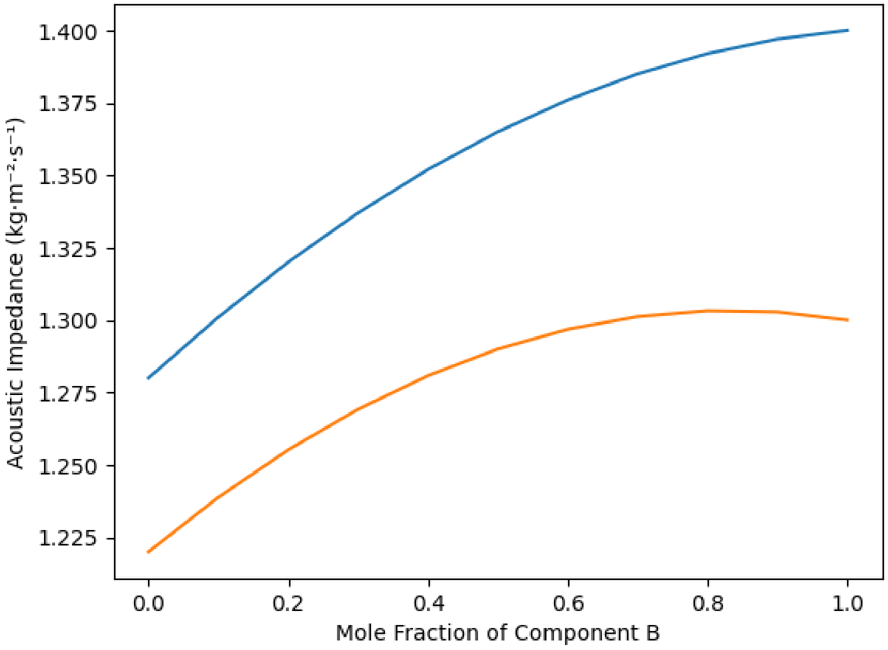
The intermolecular free length calculated for the binary liquid mixtures showed systematic variation with mole fraction and temperature. In general, free length decreased with increasing ultrasonic velocity, reflecting enhanced molecular association and closer molecular packing.

Temperature-dependent measurements revealed an increase in intermolecular free length with rise in temperature, consistent with thermal expansion and increased molecular motion. The non-linear variation of free length with composition suggests structural rearrangements occurring due to molecular interactions between the mixture components.

5.5 Acoustic Impedance

Acoustic impedance values exhibited trends governed by both density and ultrasonic velocity variations. In most cases, acoustic impedance increased with increasing concentration of the component contributing to higher density and stronger interactions. The observed compositional dependence of acoustic impedance further supports the non-ideal nature of mixing in the investigated binary liquid systems.Temperature variation studies showed a gradual decrease in acoustic impedance with increasing temperature, reflecting reduced rigidity of the liquid medium at elevated temperatures.

Figure 4 :Acoustic Impedance vs Mole Fraction



5.6 Excess Thermo-Acoustic Parameters

Excess thermo-acoustic parameters were evaluated to quantify deviations from ideal behaviour. Excess adiabatic compressibility, excess intermolecular free length, and excess acoustic impedance displayed significant positive or negative deviations depending on the composition and temperature.Negative excess values were observed in certain composition ranges, indicating enhanced molecular association and structural compactness. Positive excess values in other regions suggest dominance of weaker interactions and structural loosening. The magnitude and sign of excess parameters varied with temperature, highlighting the temperature sensitivity of molecular interactions in the studied systems.

5.7 Summary of Observed Trends

The experimental results clearly demonstrate that ultrasonic velocity and derived thermo-acoustic parameters are strongly dependent on both composition and temperature. The non-linear variation of these parameters across the mole fraction range confirms the presence of molecular interactions and non-ideal mixing behaviour in the investigated binary liquid systems. The computed thermo-acoustic parameters provide a quantitative basis for further interpretation of molecular association mechanisms.

VI. Error Analysis and Validation

Experimental uncertainties in ultrasonic velocity and density measurements play a crucial role in determining the reliability of derived thermo-acoustic parameters. In the present study, the accuracy of ultrasonic velocity measurements was limited by the least count of the ultrasonic interferometer, while density measurements were subject to uncertainties arising from mass determination and temperature fluctuations.The overall uncertainty in ultrasonic velocity was estimated to be within $\pm 0.5\ \mathrm{m \cdot s^{-1}}$, whereas density measurements were accurate up to $\pm 0.0001\ \mathrm{g \cdot cm^{-3}}$. The propagation of these uncertainties into calculated thermo-acoustic parameters such as adiabatic compressibility, intermolecular free length, and acoustic impedance was evaluated using standard error propagation methods. The resulting uncertainties were found to be within acceptable experimental limits.

To validate the experimental methodology, measurements of pure liquid properties were compared with standard literature values, and good agreement was observed. Additionally, repeated measurements under identical experimental conditions yielded consistent results, confirming the reproducibility and reliability of the data. These validation checks ensure that the observed trends in thermo-acoustic parameters are intrinsic to the binary liquid systems and not artifacts of experimental error.

VII. Discussion

The observed variation in ultrasonic velocity and derived thermo-acoustic parameters with composition and temperature provides clear evidence of molecular interactions in the investigated binary liquid systems. The non-linear dependence of ultrasonic velocity on mole fraction suggests significant deviation from ideal mixing behaviour, which can be attributed to the presence of specific intermolecular forces between the constituent molecules. Such deviations are commonly associated with structural rearrangements and molecular association occurring upon mixing.The decrease in ultrasonic velocity with increasing temperature across all compositions indicates the dominance of thermal agitation, which weakens intermolecular attractions and increases intermolecular spacing. However, the extent of this decrease varies with composition, implying that molecular association persists to different degrees depending on the nature and proportion of the components present in the mixture. This behaviour highlights the competing influence of thermal effects and intermolecular interactions on acoustic propagation.

Adiabatic compressibility and intermolecular free length exhibit trends complementary to ultrasonic velocity, reinforcing the interpretation of molecular association. Lower compressibility and reduced free length at certain compositions point toward enhanced molecular packing and stronger attractive forces. Conversely, higher values observed in other composition ranges suggest structural loosening and weaker interactions. These findings are consistent with the concept that molecular association in binary mixtures is composition dependent rather than uniform across the entire mole fraction range.Acoustic impedance results further support the existence of molecular interactions, as its variation reflects the combined effects of density and ultrasonic velocity. Higher impedance values correspond to mixtures exhibiting greater rigidity and cohesive strength, whereas lower values indicate reduced resistance to sound propagation due to weakened interactions. The systematic nature of these variations confirms that the observed trends are intrinsic to the molecular structure of the mixtures rather than experimental artifacts.

The analysis of excess thermo-acoustic parameters provides deeper insight into the non-ideal behaviour of the studied systems. Negative excess values observed at specific compositions indicate strong attractive interactions and compact molecular arrangements, while positive excess values suggest dominance of dispersive forces and structural expansion. The temperature dependence of excess parameters demonstrates that molecular association is sensitive to thermal conditions, with increased temperature generally favoring dissociative interactions. Overall, the combined interpretation of ultrasonic velocity, thermo-acoustic parameters, and excess properties establishes ultrasonic techniques as an effective tool for probing molecular association and interaction mechanisms in binary liquid systems.

VIII. Conclusion

The present thermo-acoustic investigation successfully demonstrates the applicability of ultrasonic velocity measurements in elucidating molecular association in binary liquid systems. Experimental measurements of ultrasonic velocity and density, along with the evaluation of derived thermo-acoustic parameters, reveal clear deviations from ideal mixing behavior, confirming the presence of specific intermolecular interactions between the components.The non-linear variation of ultrasonic velocity and related parameters with composition indicates that molecular association in binary liquid mixtures is strongly dependent on mole fraction. Temperature-dependent studies show that increased thermal agitation weakens intermolecular forces, leading to reduced ultrasonic velocity and increased compressibility. The evaluated excess thermo-acoustic parameters further substantiate the coexistence of associative and dissociative interactions within the studied systems.

The results of this study highlight the sensitivity and reliability of ultrasonic techniques as a non-destructive experimental approach for investigating liquid structure and molecular interactions. The thermo-acoustic methodology adopted in this work provides a valuable framework for correlating macroscopic acoustic properties with microscopic molecular behavior. Such insights are essential for understanding solution dynamics and for the design and optimization of liquid mixtures in industrial, chemical, and pharmaceutical applications.

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