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Ultrasonic and Thermodynamic Investigation of Molecular Interactions in Cinnamaldehyde-Based Binary Liquid Mixtures at 303.15 K

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Abstract: This study presents a comprehensive investigation of molecular interactions in four binary liquid mixtures, namely methanol-cinnamaldehyde, acetone-cinnamaldehyde, p-dioxane-cinnamaldehyde, and cyclohexane-cinnamaldehyde, over the entire mole fraction range. Ultrasonic velocity and density measurements were carried out, and various thermodynamic parameters such as adiabatic compressibility, volume expansivity, excess adiabatic compressibility, and excess volume expansivity were evaluated.

The variation of ultrasonic velocity and density with mole fraction indicates significant changes in molecular packing and intermolecular forces across different systems. Negative values of excess adiabatic compressibility observed in methanol and acetone systems confirm strong heteromolecular interactions dominated by hydrogen bonding and dipole–dipole interactions. In contrast, the p-dioxane system exhibits weak interactions characterized by small negative deviations, while the cyclohexane system shows positive excess values, indicating weak dispersive interactions and structural loosening.

The study reveals that the nature and strength of molecular interactions strongly depend on the polarity and hydrogen-bonding ability of the components. Overall, the results provide valuable insight into non-ideal behavior, structural effects, and interaction mechanisms in binary liquid mixtures, which are essential for understanding solution chemistry and industrial applications.

Index Terms- Ultrasonic velocity, Density, Compressibility, Excess properties, Molecular interactions, Cinnamaldehyde, Binary mixtures, Hydrogen bonding**.**

I. INTRODUCTION

The investigation of molecular interactions in binary liquid mixtures has attracted considerable attention due to its importance in understanding thermodynamic non-ideality, solution structure, and transport behavior in chemical and industrial processes [1]. Intermolecular forces such as hydrogen bonding,

dipole-dipole interactions, dipole-induced dipole interactions, and dispersion forces play a crucial role in determining macroscopic properties of liquid mixtures [2]. Several researchers have employed ultrasonic and thermodynamic studies to probe molecular interactions in liquid systems, as acoustic and volumetric parameters are highly sensitive to structural rearrangements and association phenomena at the molecular level [3]. Measurements of density and ultrasonic velocity, along with derived parameters such as adiabatic compressibility and volume expansivity, have been widely used to elucidate the strength and nature of interactions between unlike molecules in binary mixtures [4].

Binary mixtures composed of polar and non-polar components exhibit diverse interaction mechanisms depending on the polarity, hydrogen-bonding capability, and molecular structure of the constituents. In nonpolar-polar systems, the introduction of a nonpolar solvent tends to weaken the self-associated network of polar molecules, leading to structural modifications governed mainly by dispersion and dipole-induced dipole interactions [6]. Conversely, polar-polar mixtures are often characterized by strong specific interactions such as hydrogen bonding and dipole-dipole attractions, resulting in significant deviations from ideal behavior [7]. Therefore, systematic experimental evaluation of acoustical and thermodynamic properties provides valuable insight into the extent of molecular association, structural effects, and interaction dynamics in such systems [8-10].

The variation of density and ultrasonic velocity with mole fraction provides fundamental information about molecular packing, cohesive forces, and structural organization within the mixtures [11]. Ultrasonic velocity measurements enable the evaluation of adiabatic compressibility, which reflects the resistance of the medium to compression and is directly related to intermolecular free length and interaction strength [12]. Furthermore, derived thermodynamic parameters such as volume expansivity, excess adiabatic compressibility, and excess volume expansivity offer deeper insight into deviations from ideality and the extent of interaction between unlike molecules [13]. Positive or negative deviations in these excess properties reveal whether dispersive, associative, or structural effects dominate the mixture behavior [14].

In this context, we focus on four representative binary liquid mixtures: methanol-cinnamaldehyde, acetone-cinnamaldehyde, p-dioxane-cinnamaldehyde, and cyclohexane cinnamaldehyde over the entire mole fraction range. Methanol is a highly polar protic solvent capable of forming strong hydrogen bonds, while acetone is a polar aprotic solvent dominated by dipole–dipole interactions. p-Dioxane is a weakly polar solvent with limited interaction capability, whereas cyclohexane is a nonpolar solvent governed primarily by dispersion forces. Cinnamaldehyde, being a conjugated aromatic aldehyde with a polar carbonyl group, can participate in dipole interactions and weak hydrogen bonding. The contrasting nature of these components makes these systems suitable models for investigating polar-polar, polar-nonpolar, and nonpolar-nonpolar intermolecular interactions.

Therefore, the objective of the present study is to systematically investigate the nature and strength of molecular interactions in methanol-cinnamaldehyde, acetone-cinnamaldehyde, p-dioxane-cinnamaldehyde, and cyclohexane-cinnamaldehyde binary mixtures by analyzing mole fraction dependent ultrasonic velocity, density, adiabatic compressibility, volume expansivity, and their

corresponding excess parameters, in order to elucidate the interaction mechanisms and structural effects governing these liquid systems.

II. Results and discussion

II. a. Methanol: Cinnamaldehyde (Polar: Non-Polar) System

The variation of ultrasonic velocity, density, adiabatic compressibility, and volume expansivity with mole fraction indicates significant changes in molecular packing and interaction strength across the entire composition range. The ultrasonic velocity increases steadily from 1084.10 m s^{-1} at pure methanol to 1375.20 m s^{-1} at pure cinnamaldehyde, suggesting enhancement in cohesive forces and stronger molecular association with increasing cinnamaldehyde concentration. Simultaneously, density increases from 0.7761 to 0.9858 $\times 10^3$ kg m^{-3}, indicating closer molecular packing (Table 1 & Fig 1).

The adiabatic compressibility decreases continuously, confirming increased resistance to compression and hence stronger intermolecular interactions. The excess adiabatic compressibility values are negative throughout the composition range, indicating strong heteromolecular interactions arising from hydrogen bonding between the -OH group of methanol and the carbonyl group of cinnamaldehyde.

The excess volume expansivity shows small positive deviations, suggesting structural rearrangement and partial breaking of methanol self-associated structure. Overall, the system exhibits strong non-ideal behavior dominated by hydrogen bonding and dipole-dipole interactions.

TABLE 1: METHANOL + CINNAMALDEHYDE (POLAR-NON-POLAR)

Mole fraction (Cm)	Ultrasonic velocity (μ) ms^{-1}	Density (ρ) $\times 10^3$ kg m^{-3}	Adiabatic Compressibility (β) $\times 10^{-10}$ m^2N^{-1}	Volume expansivity (α) $\times 10^{-3}$ k^{-1}	Excess adiabatic compressibility (β^E) $\times 10^{-10}$ m^2N^{-1}	Excess volume expansivity (α^E) $\times 10^{-3}$ k^{-1}	Free length (Lf) $\times 10^{-11}$ m
0.0	1084.10	0.7761	10.955	1.0780	0.0	0.0	6.87
0.1	1110.15	0.7952	10.195	1.2041	-0.1992	0.1085	6.63
0.2	1132.00	0.8325	9.365	1.6148	-0.4684	0.5017	6.35
0.3	1156.18	0.8422	8.891	1.1330	-0.3816	0.0023	6.19
0.4	1188.50	0.8612	8.238	1.3155	-0.4738	0.1673	5.96
0.5	1204.00	0.8841	7.808	1.2576	-0.3430	0.0918	5.80
0.6	1239.35	0.9034	7.220	1.2410	-0.3702	0.0577	5.58
0.7	1258.00	0.9212	6.860	1.2265	-0.1694	0.0256	5.43
0.8	1297.00	0.9410	6.320	1.3810	-0.1486	0.1626	5.22
0.9	1332.00	0.9642	5.871	1.3510	-0.0368	0.0951	5.03
1.0	1375.20	0.9858	5.347	1.2535	0.0	0.0	4.8

II. b. Acetone: Cinnamaldehyde (Polar: Non-Polar) System

In this system, ultrasonic velocity increases from 1138.65 to 1401.55 m s^{-1}, indicating strengthening of intermolecular interactions with increasing cinnamaldehyde concentration. Density also shows a gradual increase, reflecting improved molecular packing.

The adiabatic compressibility decreases across the composition range, suggesting enhanced rigidity of the system due to intermolecular attractions. The excess adiabatic compressibility values are negative throughout, confirming strong interactions between acetone and cinnamaldehyde molecules primarily through dipole-dipole interactions (Table 2 & Fig.1).

The excess volume expansivity values are positive, indicating structural adjustments and dominance of interaction effects over simple mixing behavior. Thus, the system shows moderate to strong non-ideal behavior governed by dipolar interactions.

TABLE 2: ACETONE + CINNAMALDEHYDE (POLAR-NONPOLAR)

Mole fraction (Cm)	Ultrasonic velocity (μ) ms^{-1}	Density (ρ) $\times 10^3$ $kg\ m^{-3}$	Adiabatic Compressibility (β) $\times 10^{-10}$ m^2N^{-1}	Volume expansivity (α) $\times 10^{-3}$ k^{-1}	Excess adiabatic compressibility (β^E) $\times 10^{-10}$ m^2N^{-1}	Excess volume expansivity (α^E) $\times 10^{-3}$ k^{-1}	Free length (Lf) $\times 10^{-11}$ m
0.0	1138.65	0.7792	9.889	1.5900	0.0	0.0	6.53
0.1	1180.00	0.8105	8.863	2.1500	-0.5550	0.5937	6.18
0.2	1205.15	0.8332	8.623	2.2450	-0.3240	0.7224	6.09
0.3	1224.00	0.8532	7.786	2.2000	-0.6900	0.7111	5.79
0.4	1245.50	0.8761	7.378	2.1050	-0.6270	0.6498	5.64
0.5	1270.70	0.8959	6.905	2.2040	-0.6290	0.7825	5.45
0.6	1291.15	0.9160	6.526	2.0850	-0.5370	0.6172	5.30
0.7	1315.20	0.9340	6.194	2.0150	-0.3980	0.6609	5.16
0.8	1347.00	0.9525	5.786	2.0200	-0.3350	0.7016	4.99
0.9	1370.10	0.9697	5.483	1.8050	-0.1670	0.5183	4.86
1.0	1401.55	0.9865	5.179	1.2530	0.0	0.0	4.72

II c. p-Dioxane: Cinnamaldehyde (Weakly Polar: Nonpolar) System

The ultrasonic velocity increases slightly from 1326.40 to 1377.10 m s^{-1}, indicating comparatively weaker interaction strength. Density remains nearly constant, suggesting limited structural changes in the mixture.

Adiabatic compressibility decreases gradually, indicating some degree of interaction between unlike molecules. The excess adiabatic compressibility values are negative but of small magnitude, reflecting weak interactions between p-dioxane and cinnamaldehyde (Table 3 & Fig.1).

Excess volume expansivity values are mostly negative at lower mole fractions and slightly positive at higher compositions, indicating minor structural rearrangements. The system is characterized by weak dipole-induced dipole interactions and limited association effects.

TABLE 3: P-DIOXINE + CINNAMALDEHYDE (POLAR–NON-POLAR)

Mole fraction (Cm)	Ultrasonic velocity (μ) ms^{-1}	Density (ρ) $\times10^3$ kg m^{-3}	Adiabatic Compressibility (β) $\times10^{-10}$ m^2N^{-1}	Volume expansivity (α) $\times10^{-3}$ k^{-1}	Excess adiabatic compressibility (β^E) $\times10^{-10}$ m^2N^{-1}	Excess volume expansivity (α^E) $\times10^{-3}$ k^{-1}	Free length (Lf) $\times10^{-11}$ m
0.0	1326.40	1.0275	5.532	0.9300	0.0	0.0	4.88
0.1	1337.00	1.0260	5.452	0.8540	-0.0613	-0.1034	4.81
0.2	1348.45	1.0245	5.368	0.8535	-0.1266	-0.1413	4.77
0.3	1358.00	1.0255	5.288	0.8700	-0.1879	-0.1579	4.75
0.4	1364.00	1.0265	5.236	0.9460	-0.2212	-0.1136	4.73
0.5	1368.10	1.0270	5.202	1.0125	-0.2365	-0.0795	4.72
0.6	1374.00	1.0245	5.170	1.0705	-0.2498	-0.0539	4.69
0.7	1378.02	1.0290	5.118	1.1920	-0.2831	0.0352	4.69
0.8	1383.00	1.0232	5.110	1.2765	-0.2724	0.0873	4.69
0.9	1385.00	1.0220	5.101	1.2640	-0.2627	0.0424	4.69
1.0	1377.10	0.9865	5.345	1.2540	0.0	0.0	4.8

II. d. Cyclohexane: Cinnamaldehyde (Non-polar:Non-polar) System

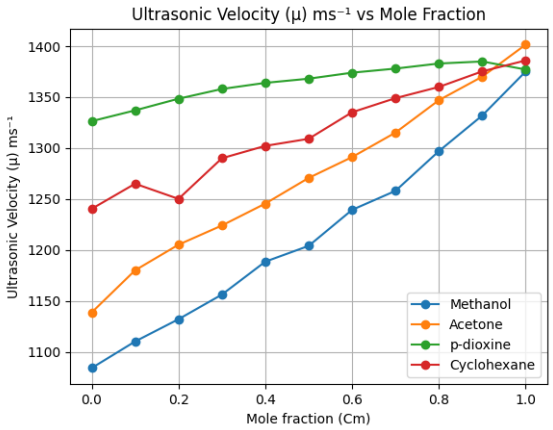
In this system, ultrasonic velocity increases from 1240.35 to 1386.00 m s^{-1}, while density shows a gradual increase with mole fraction. However, the interaction behavior is distinctly different from polar systems. Adiabatic compressibility decreases, but the excess adiabatic compressibility values are predominantly positive, indicating weaker interactions between unlike molecules compared to pure components. This suggests that mixing leads to structural loosening rather than association.

Excess volume expansivity values are positive throughout most of the composition range, confirming expansion in volume and weak dispersive interactions. The system is dominated by dispersion forces and exhibits nearly ideal or weakly non-ideal behavior (Table 4 & Fig. 4).

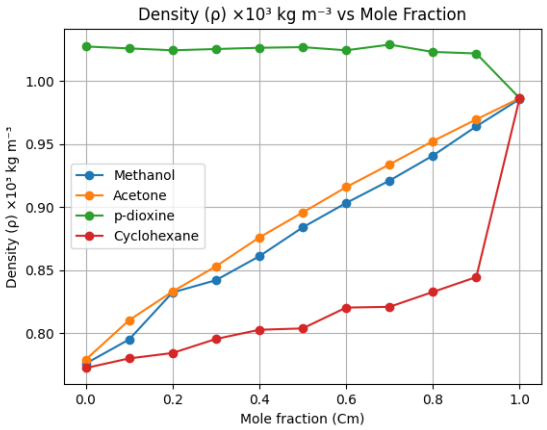
TABLE 4: CYCLOHEXANE + CINNAMALDEHYDE (NON-POLAR–NON-POLAR)

Mole fraction (Cm)	Ultrasonic velocity (μ) ms^{-1}	Density (ρ) $\times10^3$ kg m^{-3}	Adiabatic Compressibility (β) $\times10^{-10}$ m^2N^{-1}	Volume expansivity (α) $\times10^{-3}$ k^{-1}	Excess adiabatic compressibility (β^E) $\times10^{-10}$ m^2N^{-1}	Excess volume expansivity (α^E) $\times10^{-3}$ k^{-1}	Free length (Lf) $\times10^{-11}$ m
0.0	1240.35	0.7726	8.338	1.2300	0.0	0.0	5.99
0.1	1265.00	0.7802	8.010	1.4600	-0.0218	0.2609	5.87
0.2	1250.10	0.7845	8.156	1.4470	0.4304	0.2787	5.93
0.3	1290.00	0.7956	7.553	1.6520	0.1336	0.5446	5.70
0.4	1302.15	0.8029	7.345	1.8400	0.2318	0.7334	5.62
0.5	1309.10	0.8040	7.257	1.8350	0.4500	0.7592	5.59
0.6	1335.15	0.8205	6.837	2.0940	0.3362	1.0491	5.43
0.7	1349.00	0.8211	6.692	2.2300	0.4874	1.2160	5.37
0.8	1360.00	0.8329	6.491	2.3205	0.6026	1.3373	5.29
0.9	1375.30	0.8447	6.259	1.2540	0.6758	0.3016	5.19
1.0	1386.00	0.9867	5.276	0.9215	0.0	0.0	4.77

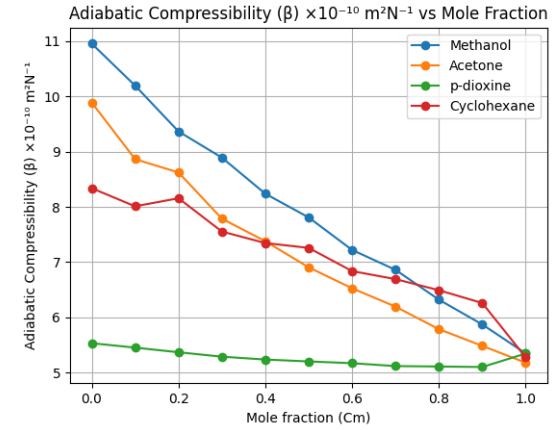
ULTRASONIC VELOCITY (M) MS^{-1}



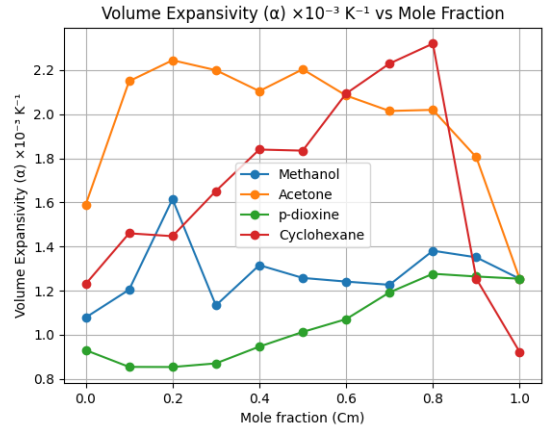
DENSITY (P) $\times 10^3$ KG M^{-3}



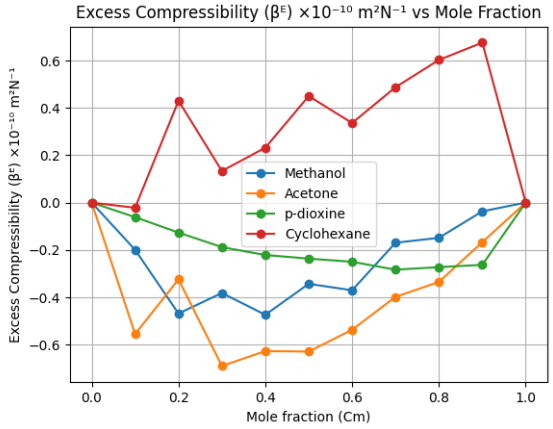
ADIABATIC COMPRESSIBILITY (B) $\times 10^{-10}$ M^2N^{-1}



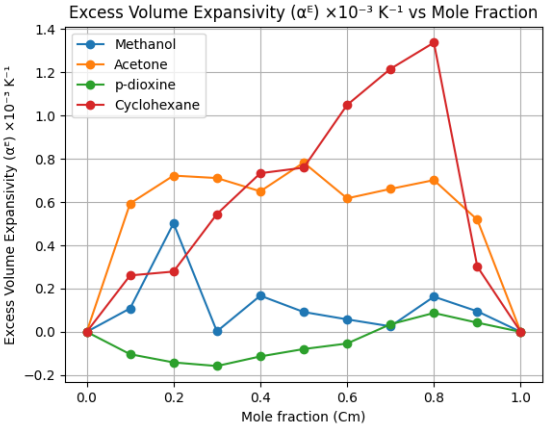
VOLUME EXPANSIVITY (A) $\times 10^{-3}$ K^{-1}



EXCESS COMPRESSIBILITY (B^E) $\times 10^{-10}$ M^2N^{-1}



EXCESS VOLUME EXPANSIVITY (A^E) $\times 10^{-3}$ K^{-1}



FREE LENGTH VS MOLE FRACTION

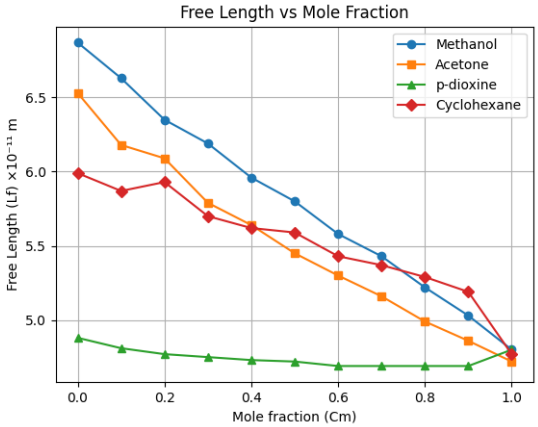


Fig. 1. Variation of Ultrasonic Velocity, Density, Adiabatic Compressibility, Volume Expansivity, Excess Compressibility, and Excess Volume Expansivity with Mole Fraction

III. EXPERIMENTAL

III. a. Materials

Methanol, Acetone, p-Dioxane, Cyclohexane, and Cinnamaldehyde of analytical reagent (AR) grade purity (>99%) were procured from standard commercial suppliers and used without further purification. The purity of the chemicals was confirmed by comparing the experimentally measured densities with standard literature values.

Binary mixtures were prepared by mass using an electronic analytical balance with an accuracy of ±0.0001 g. The mole fraction uncertainty was estimated to be within ±0.0002. Fresh mixtures were prepared prior to each measurement to avoid composition changes due to evaporation.

III.b. Measurements

All measurements were carried out at a constant temperature of (303.15 ± 0.01) K, maintained using a thermostatically controlled water bath. The temperature stability was monitored using a calibrated digital thermometer.

Ultrasonic velocity (U) was measured using a single crystal ultrasonic interferometer operating at a fixed frequency of 2 MHz with an accuracy of ±0.1%. The instrument was calibrated using standard liquids before measurements.

Density (ρ) measurements were performed using a 10 mL specific gravity bottle with an accuracy of ±0.0001 g cm^{-3}. The bottle was thoroughly cleaned and dried before each measurement.

Viscosity (η) was determined using an Ostwald viscometer with a flow time accuracy of ±0.1 s. Each flow time was recorded as the average of three successive measurements.

All experimental measurements were repeated three times, and the average values were reported. The maximum estimated uncertainties were ±0.1% for ultrasonic velocity, ±0.0001 g cm^{-3} for density, and ±0.2% for viscosity. The uncertainty in calculated adiabatic compressibility was estimated to be within ±0.5%.

IV. Governing Equations

The following standard thermodynamic and acoustic relations were used to calculate the derived parameters from experimentally measured ultrasonic velocity, density, and viscosity data at 303.15 K.

IV. a. Adiabatic Compressibility (β)

$\beta = 1 / (\rho U^2)$

Where ρ is the density of the mixture and U is the ultrasonic velocity.

IV. b. Intermolecular Free Length (Lf)

$$Lf = K_T \sqrt{\beta} \; ; \; (K_T = 2.0112 \times 10^{-6})$$

Where K_T is the temperature-dependent Jacobson constant and β is the adiabatic compressibility.

IV. c. Excess Property (Y^E)

$Y^E = Y\text{-mix} - (x_1Y_1 + x_2Y_2)$

Where Y represents any thermodynamic parameter such as β or α; x_1 and x_2 are the mole fractions of components 1 and 2 respectively; Y-mix is the property of the mixture, and Y_1 and Y_2 are the properties of pure components.

IV. d. Volume Expansivity (α)

$$\alpha = \frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)$$

All measurements were carried out at a constant temperature of (303.15 ± 0.01) K, maintained using a thermostatically controlled water bath. The temperature stability was monitored using a calibrated digital thermometer.

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Where Y represents any thermodynamic parameter such as β or α; x_1 and x_2 are the mole fractions of components 1 and 2 respectively; Y-mix is the property of the mixture, and Y_1 and Y_2 are the properties of pure components.

V. d. Volume Expansivity (α)

$$\alpha = \frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)$$

VI. Conclusion

In the present work, molecular interactions in four binary liquid mixtures containing cinnamaldehyde have been systematically investigated using ultrasonic and thermodynamic approaches. The variation of ultrasonic velocity, density, and derived parameters clearly demonstrates the dependence of interaction strength on the nature of the constituent molecules.

The methanol-cinnamaldehyde system exhibits strong non-ideal behavior with negative excess adiabatic compressibility, indicating significant hydrogen bonding and strong heteromolecular interactions. The acetone system also shows pronounced negative deviations, suggesting dominant dipole–dipole interactions. In contrast, the p-dioxane system displays weaker interactions characterized by small deviations, indicating the presence of dipole-induced dipole interactions. The cyclohexane system, being non-polar, shows positive excess values, confirming weak dispersive forces and structural expansion upon mixing.

Overall, the study highlights that polar systems exhibit stronger associative interactions, while non-polar systems are governed by weak dispersive forces. These findings provide important insights into the structure-property relationship and non-ideal behavior of liquid mixtures, which are useful in chemical processing, formulation design, and industrial applications.

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