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Ultrasonic Investigations on the Molecular Interactions Between Citronellol and Butanol-1 in their Binary Mixtures at Different Temperatures

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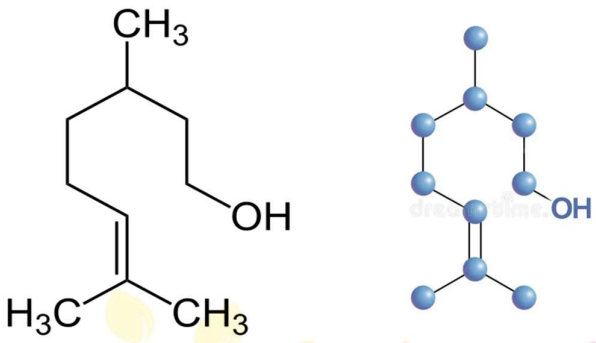
Abstract: Ultrasonic investigation is a valuable approach for determining the nature of molecular interactions in binary mixtures. The Ultrasonic velocity (u), Density (ρ), and Viscosity (η) of a binary mixture of Citronellol, a component of many perfumes and food flavorings for its floral-citrus notes, with Butanol-1 carrying varying molar concentrations, were determined at temperatures 296.15K, 300.15K, 304.15K, 308.15K, and 312.15K. Acoustic characteristics, including adiabatic compressibility (β_s), acoustic impedance (Z), intermolecular free length (L_f), and relaxation time(τ) were obtained. The influence of molar concentration and temperature variations on several concerned parameters was investigated. The observed values of Ultrasonic velocity, Density, and Viscosity increased with increasing solute concentration and decreased with rising temperature. The selected acoustical parameters show a significant relationship between binary mixture concentration and temperature both. Based on the observations, increasing the concentration of Citronellol in the binary mixture leads to molecular association and clustering, resulting in enhanced density and stability. In contrast, rising temperatures disrupt these associations, causing molecular agitation and potential partial dissociation of the solute-solvent complexes.

Keywords: Ultrasonic velocity, Viscosity, Density, Citronellol, Intermolecular interactions.

INTRODUCTION

Perfumes often incorporate essential oils, which are highly concentrated plant extracts used for their aromatic properties. Essential oils are obtained from various parts of plants, including flowers, leaves, bark, and roots through processes such as steam distillation, solvent extraction, and cold pressing [1]. These oils contain a variety of compounds, such as terpenes, aldehydes, and esters which contribute to their distinctive fragrance [2]. Citronellol, a notable product derived from essential oils like citronella, rose, and geranium, is a monoterpenoid alcohol with significant applications in the fragrance industry due to its pleasant floral-citrus aroma. Despite its widespread use in perfumery, citronellol has not yet been extensively studied in acoustical investigations. This gap presents an opportunity to explore its molecular interactions and properties of Citronellol in a suitable solvent, through ultrasonic techniques, which can enhance the understanding of its behavior in various applications. The importance of citronellol extends beyond its fragrance; it also possesses antimicrobial and anti-inflammatory properties, making it valuable further in pharmaceuticals and personal care products. [3]

Citronellol, an acyclic alcohol with the molecular formula $C_{10}H_{20}O$, is prevalent in essential oils in some distinct essential oils [4]. It has a boiling point of 222°C to 224°C and a melting point of −7°C to −4°C, indicating its liquid state under standard conditions. Citronellol is moderately soluble in water but highly soluble in organic solvents like alcohols, ethers, and non-polar solvents, making it valuable in synthesizing fragrances, flavors, and certain pharmaceuticals. Citronellol bears a widespread application. The given image shows the molecular structure of Citronellol.



CITRONELLOL

Solvents play a crucial role in chemical analysis, serving as medium for dissolving, diluting, and extracting substances employed in various analytical techniques. They are selected based on their ability to dissolve specific compounds while maintaining chemical stability and minimal interference with analytes. Butanol-1 (n-butanol), an alcoholic solvent, frequently utilized in liquid-liquid extractions due to its moderate polarity and effectiveness in dissolving both polar and non-polar substances [5]. Its versatility makes it suitable for applications ranging from chromatography to spectrophotometry, where precise solvation properties are essential for accurate analytical measurements. Understanding solvent characteristics and their interactions with analytes is significant for optimizing analytical procedures across scientific disciplines.

Butanol-1, with the molecular formula $C_4H_{10}O$, is a primary alcohol characterized by a four-carbon straight chain bearing a polar hydroxyl group. It has a boiling point of 117.7°C and a melting point of -89.8°C, reflecting its moderate volatility and wide range of liquid state. Butanol-1 is moderately soluble in water and highly soluble in organic solvents such as ethanol, ether, and acetone, making it an effective solvent for both polar and non-polar substances. Its versatility in chemical transformations and solubility properties in various applications make it essential in chemical synthesis, pharmaceutical formulations, coatings, providing specific evaporation rates, and reactivity.

In recent years, the application of ultrasonic technique is an effective tool and has gained considerable interest for analyzing molecular interactions in pure liquids and liquid solutions [6-7]. Key properties such as ultrasonic velocity, viscosity, and density, and their variations with respect to solution composition and temperature, are crucial for advancements in database regarding engineering, chemical processing, and biological production [8-9]. The measurements of the aforesaid primary parameters of ultrasonics support diverse applications, including food processing, material testing, navigational aids, and cleaning processes for glassware and textiles [10]. Hence therefore, the textile, leather, and pharmaceutical industries also benefit significantly from these techniques [11-12].

After thorough investigation of the related acoustical literature, a research experiment was undertaken to study all the thermophysical data of the binary mixtures prepared with Citronellol and 1-butanol, for the assessment of various primary and derived acoustic parameters, to seek an inference about the resulting molecular interaction.

EXPERIMENTAL PROCEDURES

The characteristics of binary mixtures are decided by their composition and have various usage in different chemical, industrial, and biological processes [13, 14]. In this way, the study of intermolecular interactions in binary liquid mixtures is critical. To examine the thermodynamic characteristics and nature of molecular interactions in binary mixtures of Citronellol (3,7-Dimethyloct-6-en-1-ol) with 1-Butanol, ultrasonic velocity, density, and viscosity were assessed in different molar compositions at 296.15K, 300.15K, 304.15K, 308.15K, and 312.15K.

The experimental results were utilized to determine parameters such as adiabatic compressibility **(β_s)**, acoustic impedance (**Z**), intermolecular free length (**L_f**), and relaxation time **(τ)** utilizing the conventional relation [15, 16, 20-21]. These factors facilitate a deeper understanding of the molecular interactions between solute and solvent. Variations observed in the calculated parameters concerning temperature and molar concentration were further analyzed to elucidate the intermolecular interactions occurring between components of the binary mixture. In view to the above introduction the following work was undertaken to study the intermolecular interactions within the binary systems undertaking, which leads to contribute significantly to the field of solution chemistry.

The compounds taken in the experiment were Citronellol and 1-Butanol of AR grade, procured from CDH Fine Chemicals, India. An electronic balance with a sensitivity of ±0.0001 g was employed to weigh the solute required.

A stock solution was prepared employing Class A borosilicate glasses. Subsequently, binary solutions of Citronellol and Butanol-1 were prepared to achieve various molar concentrations required. The densities and viscosities of the liquids were gauged at five different temperatures using a pre-calibrated specific gravity bottle (class A) and an Ostwald viscometer (Class A). The flow duration of the liquids in the viscometer was recorded with an electronic digital stopwatch, accurate to ±0.01 sec. An electrically powered constant temperature circulating water bath (Model number: AP-S-72, Mittal Enterprises, New Delhi) was utilized to maintain a constant temperature during the experimental observations with an accuracy of ±0.1K.

The ultrasonic velocities were ascertained using a single-frequency ultrasonic interferometer operating at 2 MHz (Model: F-05 Mittal Enterprises, New Delhi), with an accuracy of ±0.03%. Prior to initiating the experimental measurements with the ultrasonic interferometer, with a 10 mL capacity measuring cell, was thoroughly cleaned and dried to eliminate maximally any factors that could introduce experimental errors in the measured values.

For precise calibration of the instruments, double-distilled water and certain pure solvents such as 1-butanol, 2-propanol, dichloromethane and methyl acetate were used to determine the density (ρ), viscosity (η), and ultrasonic velocity (u).

The values of ultrasonic velocity, density, and viscosity measurements were employed in computing several other parameters for studying solute-solvent interaction and structural effects. Acoustic characteristics such as adiabatic compressibility (β_s), acoustical impendence (Z), intermolecular free length (L_f), and relaxation time (τ) were calculated using conventional cited formulae. [13, 15-16, 20-21].

Adiabatic Compressibility (β_s)

The molecule's structural changes in the mixture occur due to the electrostatic field between interacting molecules. The structural arrangement of molecules results in a considerable change in adiabatic compressibility, which can be expressed as:

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

U is ultrasonic velocities, and ρ is the density of liquid mixtures.

Acoustic Impedance (Z)

The relation relates the specific acoustic impedance to density and ultrasonic velocity.

$$Z=U. \rho$$

Intermolecular Free Length (L_f)

The free length is the distance covered by sound waves between the surfaces of the neighboring molecules and is related to ultrasonic velocity and density:

$$L_f = K_T. \beta^{1/2} \text{ (Here we put, } K_T = (93.875 + 0.375\ T) \times 10^{-8})$$

Relaxation Time (τ)

Relaxation time is the time it takes for excitation energy to become translational energy, and it is affected by temperature and impurity levels. The relaxation time may be computed using the following equation:

$$\tau = 4\eta / 3\rho. U^2$$

RESULTS AND DISCUSSION

Tables 1(A, B and C) and 2(A, B, C and D) present the recorded ultrasonic velocity, density, viscosity, and associated acoustic parameters for binary systems of Citronellol and butanol-1 at 296.15K, 300.15K, 304.15K, 308.15K, and 312.15K respectively. These are also depicted graphically in Figures 1 to 7.

The measured values of Ultrasonic velocity (u), Density (ρ), and Viscosity (η) of binary mixtures in terms of molar concentration at different temperatures are given below in table-1 (A, B and C).

Table 1A: Ultrasonic velocity u (ms^{-1}) at different Temperatures (K)

Conc. (Moles/L)	296.15 K	300.15 K	304.15 K	308.15 K	312.15 K
0.0	1243.63	1237.16	1230.68	1224.21	1217.73
0.1	1245.04	1238.66	1232.27	1225.89	1219.50
0.2	1247.53	1240.88	1234.22	1227.57	1220.91
0.3	1249.68	1242.98	1236.27	1229.57	1222.86
0.4	1251.98	1245.18	1238.37	1231.57	1224.76
0.5	1253.78	1247.08	1240.37	1233.67	1226.96
0.6	1256.13	1249.28	1242.42	1235.57	1228.71
0.7	1258.18	1251.38	1244.57	1237.77	1230.96
0.8	1260.45	1253.59	1246.73	1239.87	1233.01
0.9	1262.36	1255.60	1248.83	1242.07	1235.30
1.0	1264.58	1257.81	1251.04	1244.27	1237.50

Table 1B: Density ρ (kg/m^3) at different Temperatures (K)

Conc. (Moles/L)	296.15 K	300.15 K	304.15 K	308.15 K	312.15 K
0.0	799.97	799.20	798.42	797.65	796.87
0.1	798.42	796.34	794.25	792.17	790.08
0.2	800.05	797.88	795.70	793.53	791.35
0.3	801.29	798.75	796.21	793.67	791.13
0.4	802.52	799.62	796.71	793.81	790.90
0.5	803.02	800.33	797.63	794.94	792.24
0.6	803.52	801.04	798.55	796.07	793.58
0.7	803.33	801.08	798.82	796.57	794.31
0.8	803.14	801.12	799.09	797.07	795.04
0.9	806.35	803.27	800.18	797.10	794.01
1.0	809.57	805.42	801.27	797.12	792.97

Table 1C: Viscosity, η (Pa.s), at different Temperatures (K)

Conc. (Moles/L)	296.15 K	300.15 K	304.15 K	308.15 K	312.15 K
0.0	2.155	1.992	1.828	1.665	1.501
0.1	2.692	2.523	2.353	2.184	2.014
0.2	2.711	2.545	2.378	2.212	2.045
0.3	2.731	2.565	2.399	2.233	2.067
0.4	2.752	2.586	2.419	2.253	2.086
0.5	2.771	2.605	2.438	2.272	2.105
0.6	2.793	2.627	2.460	2.293	2.126
0.7	2.814	2.648	2.481	2.314	2.147
0.8	2.833	2.667	2.500	2.334	2.168
0.9	2.855	2.689	2.522	2.355	2.189
1.0	2.877	2.710	2.542	2.375	2.208

Figures 1–3 Depict the experimental values of Ultrasonic velocity, Density, Viscosity plotted as a function of molar concentration.

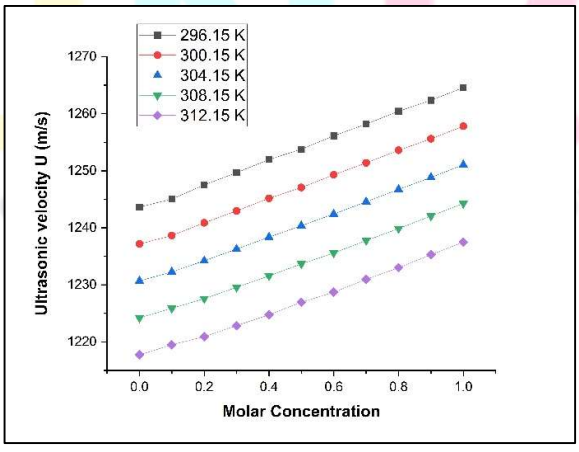


Figure 1: Variance in observed Ultrasonic velocity based on concentration and temperature

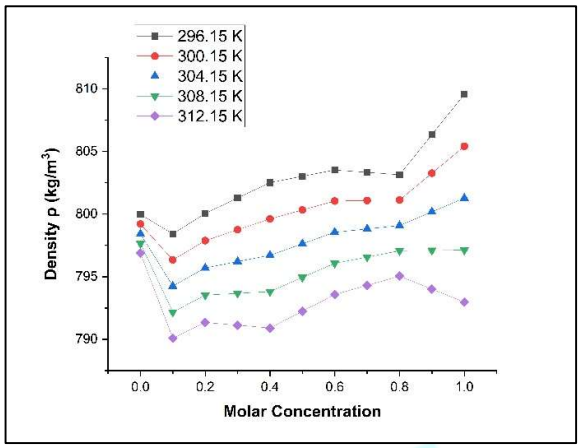


Figure 2: Variance in observed Density based on concentration and temperature

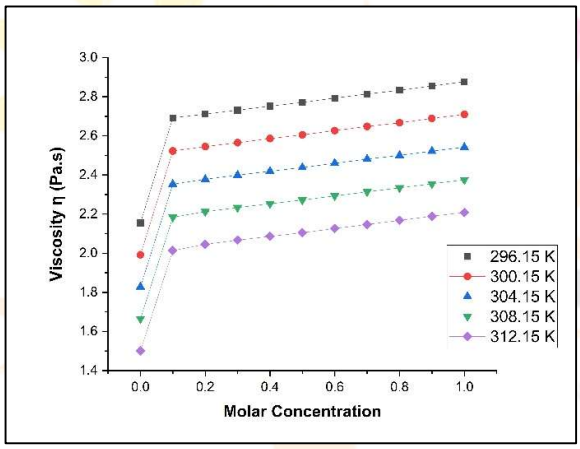


Figure 3: Variance in observed Viscosity based on concentration and temperature

The provided figures (1-3) show that the experimental values of critical parameters rise with an increase in the molecular concentration of Citronellol. Conversely, these experimental values decrease as the temperature increases, indicating a reduction in intermolecular forces due to the increased thermal energy, which causes molecular agitation in the system [14, 21].

Table-1 demonstrates that the measured values of ultrasonic velocity exhibit a linear increase with the rise in solute concentration, suggesting the presence of an intermolecular interaction in the binary system, contributing to this observed behavior [14, 18]. This strong correlation supports the hypothesis that intermolecular interactions play a crucial role in the system's behavior, and the results further suggest that solute concentration directly affects the properties of the binary system. This highlights the importance of understanding and controlling intermolecular interactions in similar systems.

Table 2(A, B, C and D): Computed values of adiabatic compressibility (β_S), acoustic impedance (Z), intermolecular free length (L_f) and relaxation time (τ) at different concentrations and temperatures. The derived parameters, namely β_s, Z, L_f, and τ, presented in Table- 2(A, B C, and D), and are graphically represented in terms of molar concentration (M) in Figures 4 to 7.

Table 2A: Adiabatic compressibility β_S x 10^{-10} (m^2/N) at different Temperatures (K)

Conc. (Moles/L)	296.15 K	300.15 K	304.15 K	308.15 K	312.15 K
0.0	8.080	8.175	8.270	8.365	8.460
0.1	8.076	8.184	8.292	8.399	8.507
0.2	8.028	8.139	8.251	8.362	8.474
0.3	7.987	8.103	8.218	8.334	8.449
0.4	7.946	8.065	8.185	8.305	8.425
0.5	7.918	8.034	8.149	8.265	8.381
0.6	7.884	7.998	8.113	8.228	8.343
0.7	7.860	7.971	8.082	8.194	8.305
0.8	7.834	7.943	8.052	8.161	8.270
0.9	7.778	7.896	8.014	8.131	8.249
1.0	7.720	7.847	7.975	8.103	8.230

Table 2B: Acoustic impedance Z X 10^6 (Kg $m^{-2}s^{-1}$) at different Temperatures (K)

Conc. (Moles/L)	296.15 K	300.15 K	304.15 K	308.15 K	312.15 K
0.0	0.994	0.988	0.982	0.976	0.970
0.1	0.994	0.986	0.978	0.971	0.963
0.2	0.998	0.990	0.982	0.974	0.966
0.3	1.001	0.992	0.984	0.975	0.967
0.4	1.004	0.995	0.986	0.977	0.968
0.5	1.006	0.998	0.989	0.980	0.972
0.6	1.009	1.000	0.992	0.983	0.975
0.7	1.010	1.002	0.994	0.985	0.977
0.8	1.012	1.004	0.996	0.988	0.980
0.9	1.017	1.008	0.999	0.990	0.980
1.0	1.023	1.013	1.002	0.991	0.981

Table 2C: Intermolecular free length, L_f 10^{-11} (m) at different Temperatures (K)

Conc. (Moles/L)	296.15 K	300.15 K	304.15 K	308.15 K	312.15 K
0.0	5.824	5.902	5.979	6.057	6.134
0.1	5.823	5.905	5.987	6.069	6.151
0.2	5.806	5.889	5.972	6.056	6.139
0.3	5.791	5.876	5.961	6.045	6.130
0.4	5.776	5.862	5.949	6.035	6.122
0.5	5.766	5.851	5.936	6.021	6.106
0.6	5.753	5.838	5.922	6.007	6.092
0.7	5.745	5.828	5.911	5.994	6.078
0.8	5.735	5.817	5.900	5.982	6.065
0.9	5.715	5.800	5.886	5.972	6.057
1.0	5.693	5.782	5.872	5.961	6.050

Table 2D: Relaxation time τ x 10^{-9} (sec) at different Temperatures (K)

Conc. (Moles/L)	296.15 K	300.15 K	304.15 K	308.15 K	312.15 K
0.0	2.328	2.171	2.014	1.857	1.699
0.1	2.906	2.753	2.599	2.446	2.292
0.2	2.909	2.762	2.614	2.466	2.318
0.3	2.916	2.771	2.626	2.481	2.336
0.4	2.924	2.781	2.638	2.494	2.351
0.5	2.933	2.790	2.647	2.504	2.361
0.6	2.944	2.801	2.658	2.516	2.373
0.7	2.957	2.814	2.671	2.528	2.385
0.8	2.966	2.824	2.682	2.540	2.398
0.9	2.969	2.831	2.692	2.554	2.415
1.0	2.970	2.835	2.701	2.566	2.432

Figures 4–7 Depict the derived parameters, plotted as a function of molar concentration.

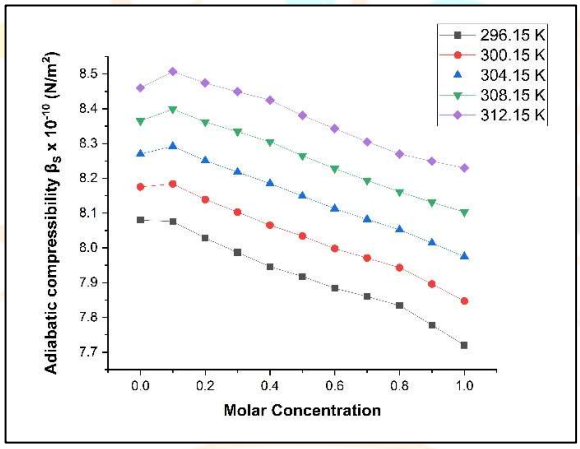


Figure 4: Variance in the calculated parameter of Adiabatic compressibility based on concentration and temperature

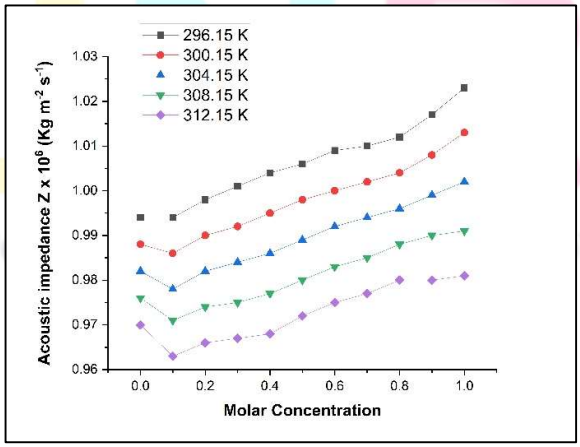


Figure 5: Variance in the calculated parameter of Acoustic impedance based on concentration and temperature

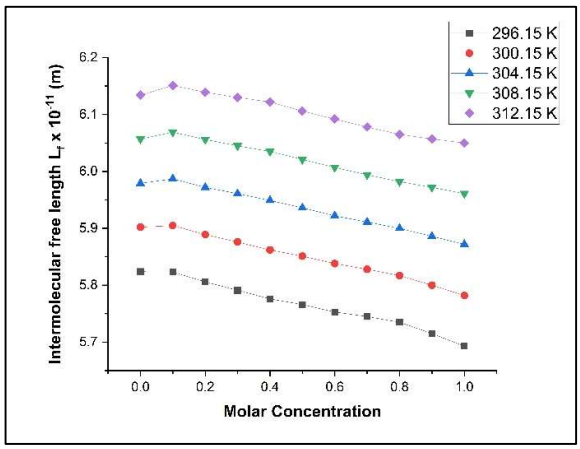


Figure 6: Variance in the calculated parameter of Intermolecular free length based on concentration and temperature

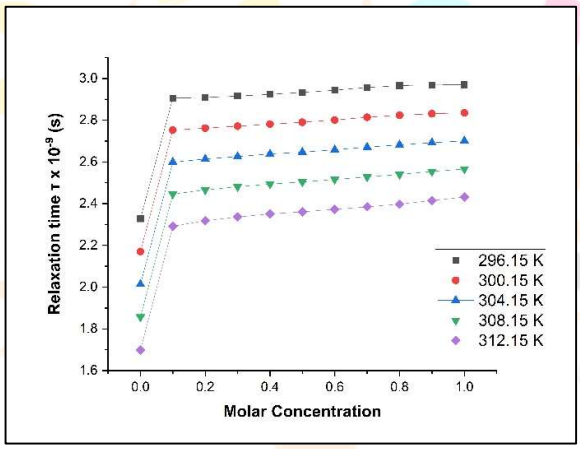


Figure 7: Variance in the calculated parameter of Relaxation time based on concentration and temperature

Adiabatic compressibility increases with temperature, indicating that the mixtures become more compressible as temperature rises. [10] It is consistent with the decreased density and increased molecular spacing at higher temperatures. Conversely, adiabatic compressibility decreases with an increase in the molar concentration of Citronellol. The more vital intermolecular forces and larger molecular size of Citronellol reduce the compressibility of the mixture. [13]

Intermolecular free length increases with temperature, indicating greater average distances between molecules as thermal energy causes them to move apart. Increasing the mole fraction of Citronellol at a constant temperature decreases the intermolecular free length, reflecting stronger intermolecular interactions and closer packing of molecules in citronellol rich mixtures.

The Eyring and Kincaid model [29] proposed that ultrasonic velocity increases with the decrease in intermolecular free length and vice-versa when two components are mixed. Similar trends have been observed in the current study for intermolecular free length and ultrasonic velocity. The declining trend of β_S and L_f suggests a strong interaction between the molecules, causing them to come closer together, resulting in a less compressible nature [18]. This could be attributed to forming a robust envelope around the solute molecules by the solvent, leading to a decrease in compressibility.

Acoustic impedance, which is the resistance to the propagation of ultrasonic waves through the medium, is shown in Table- 2B (Figure 5). Acoustic impedance, a product of density and ultrasonic velocity, decreases with increasing temperature as because of the combined effect of reduced density and ultrasonic velocity at higher temperatures. Again, thereby increasing the molar concentration of Citronellol at a given temperature, increases the acoustic impedance. This is due to the higher density and ultrasonic velocity associated with higher concentration of Citronellol molecules, which leads to higher resistance to ultrasonic wave propagation. [30-31].

Relaxation time, indicates the time for molecules to return to equilibrium after deformation, increases with rising temperature, suggesting that molecular relaxation processes slow down at higher temperatures. An increase in the molar concentration of citronellol results in a longer relaxation time due to the more complex formation and larger molecular structure of Citronellol, which requires more time to return to equilibrium [32]. The increase in relaxation time is related to the structural relaxation process, indicating the presence of molecular interaction between the component molecules. As the temperature rises, excitation energy rises, and relaxation time drops, this is because of decreases in viscosity and density, which allow the system to return to equilibrium more quickly after a disturbance [32].

CONCLUSION

The comprehensive acoustical study of the binary mixture of Citronellol and butanol-1 elucidates the significant impact of concentration and temperature on intermolecular interactions. Significant parameters such as adiabatic compressibility, acoustic impedance, intermolecular free length, and relaxation time reveal correlational changes, highlighting active intermolecular interactions and close molecular packing at higher Citronellol concentrations. A noticeable inverse relationship between adiabatic compressibility and ultrasonic velocity was observed. Furthermore, a decreasing trend of temperature dependent variations in viscosity, density, and acoustic impedance, was also found which might leads to infer an intricate balance between molecular dynamics and environmental conditions. Observational inferences from the experiment reveals that as the concentration of Citronellol increased in the binary mixture, it causes molecules to associate and cluster together, resulting in an increase in density and stability. On the other hand, increasing temperature disturbs these molecular connections, leading to increased movement of molecules and a possible partial separation of the solute-solvent complexes.

These findings contribute to help and understand in a deeper sense of solute-solvent interactions, with practical implications for various concerned industries where precise control of material properties is essential.

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