



ACOUSTIC METRICS IN BINARY MIXTURE OF A COMMERCIAL FRAGRANT COMPOUND α -TERPINYL ACETATE WITH BUTANOL-1

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ABSTRACT

The behaviour of molecules within liquid mixtures is qualitatively assessed by studying their acoustic and thermodynamic properties, which involves measuring ultrasonic velocity (u), density (ρ), and viscosity (η) of various binary mixture of different concentration of an ester alpha terpinyl acetate (α -TA) and butanol-1. The values of these experimental parameters were measured, at temperatures 293.15, 298.15, 303.15 and 308.15K. This data was used to calculate other thermodynamic parameters. These derived acoustic parameters were adiabatic compressibility (β_s), acoustic impedance (Z), intermolecular free length (L_f), free volume (V_f), relaxation time (τ), internal pressure (π_i), molar volume (V_m) and Rao's constant, Wada's constant. The influence of mole fraction concentration and temperature variations on several concerned parameters was investigated. The observed values of ultrasonic velocity, density, and viscosity increased with an increase in solute concentration, while they decreased with rise in temperature. The derived acoustical parameters showed a significant relationship with binary mixtures concentration and temperature.

Keywords: Ultrasonic velocity, Viscosity, Density, Alpha Terpinyl acetate, Intermolecular interactions.

1. INTRODUCTION

Humans are fascinated by perfumes, since time memorial. Perfumes are blend of various chemical ingredients. One of the chemicals holds a prominent position which is accountable for the fragrance of the perfume, whereas other chemicals present to provide body to the perfume. Fragrant chemical entities such as Dihydromyrcenol, alpha Campholinic aldehyde, Geranyl and Citronellyl esters etc, [1-8] respond to sensory systems in humans. The fragrances extracts derived from plant source or animal, can also be synthetically prepared in laboratory or in massive scale industrially [9,10]. Esters bearing pleasant odour happens to be choicest chemicals for perfumery, artificial essences, flavouring, and cosmetics [11]. In recent years, a growing amount of interest has been accumulating in the study of liquid mixtures that contain esters as one of their components. Alpha Terpinyl Acetate (α -TA) has been found naturally in various Thymus species grown in Europe. A variety of essential oils e.g. cedar wood oil, pine needle oil, melaleuca oil, and cardamom oil, can be extracted from their sources industrially. This fragrant compound is used in preparing various perfumes. This is commercially important fragrant molecule with sweet, herbaceous floral and lavender odor. It is widely used in soaps, shampoo, antiperspirants, lotions, air fresheners, furnishing care as well as laundry and dishwashing products. It is also used as additive to food, beverages, fruit ice creams, hard candies, baked goods, gelatin puddings and chewing gums, being consumed by end users. α -TA essential oil revealed a high antimicrobial influence towards fungi and dermatophytes. It is a colourless liquid produced from alpha-pinene or alpha-Terpineol and acetic anhydride, has a boiling point of 232°C [12-14].

The constitutional structure and 3D pictorial image of Alpha Terpinyl acetate (α -TA) is given in Fig.-1. It is a monocyclic terpenoid bearing methyl and isopropyl group along with an OH group. It has a chemical name is p-menth-1-en-8-yl acetate.

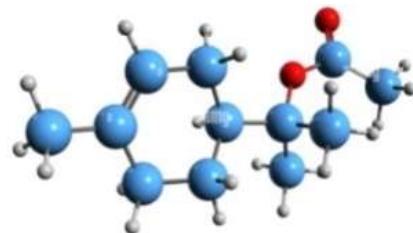
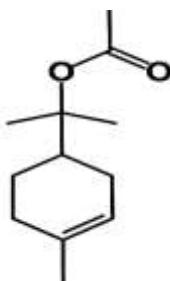


Fig.-1: Constitutional structure and 3D pictorial image of α -Terpinyl acetate

Ultrasonic measurement is a rapidly expanding tool to study the molecular behaviour of binary mixture [15,16]. Where notable progress has been achieved by medical researchers and industrial engineers [17]. Significant advancements have been made in recent years in studying the nature of interactions among solute and solvent of binary liquid mixtures. Ultrasonic research is adequately employed for accurately measuring key experimental properties like density, viscosity, speed of sound along with various derived thermodynamic and acoustical parameters which are of considerable interest in analysing the intermolecular interactions essential for engineering applications.[18-24] Cited literature provides ample data based on the measurement of density and viscosity of liquid mixtures but a combined study of these with ultrasonic velocity is still in juvenile stage [25-29]. Ultrasonic measurements against binary solutions reflects interaction between the components of the mixture [30-32].

EXPERIMENTAL METHOD

In present study, binary solution of alpha terpinal acetate (α -TA) and butanol-1 were prepared, where butanol-1 was taken as an appropriate solvent. Ultrasonic velocity, density and viscosity were measured experimentally in various said mole fraction compositions at varying temperatures 293.15, 298.15, 303.15 and 308.15K. In the entire experimental exercise, AR grade chemicals were used preferably from CDH (India). To assess the purity of the used chemical, their density, viscosity and ultrasonic velocity were compared with cited literature values. Initially stock solution of α - TA and Butanol-1 was prepared using Borosilicate glassware of class-A quality. Subsequently with the help of this solution, binary mixture of various mole fractions were prepared. A Single crystal Interferometer was used to measure the ultrasonic velocity (Model: F-05 Mittal Enterprises, New Delhi; accuracy \pm 0.03%; operating at 2MHz frequency) of prepared binary solution at the given temperature. An automatic water bath was used to maintain desired temperature for the prepared experimental solution. A precaution was taken to clean the 10ml measuring cell of ultrasonic interferometer before starting the experiment to minimize the experimental error. Ostwald viscometer (class-A) was used to record viscosity of the solutions. Digital Stopwatch (\pm 0.01 Sec.) was employed to assess the flow-time of the binary liquids in the viscometer. The density of the given solutions were measured by relative density bottle (class-A) calibrated and the weighing was done through digital weighing scale (\pm 0.1mg). Derived parameters were computed using the given under-mentioned equations.

$$\text{Adiabatic compressibility: } (\beta_s) = 1/(\rho U^2)$$

$$\text{Acoustic Impedance (Z)} = \rho U$$

$$\text{Intermolecular free length: } (L_f) = K (\beta)^{1/2}$$

where K is a temperature dependent constant known as Jacobson constant. $K = (93.875 + 0.345T) \times 10^{-8}$.

$$\text{Free volume: } (V_f) = (M_{\text{eff}} U / K \eta)^{3/2}$$

Where $M_{\text{eff}} = M_1 x_1 + M_2 x_2$ and M_1, M_2 are molecular weights and x_1, x_2 are mole fractions of the constituent components. $K = 4.28 \times 10^{-8}$ is a dimensionless constant which is temperature independent.

$$\text{Relaxation Time } (\tau) = 4\eta / 3\rho U^2$$

$$\text{Internal Pressure: } (\pi_i) = bRT(k\eta/U)^{1/2} \rho^{(2/3)} / M_{\text{eff}}^{7/6}$$

$$\text{Molar volume (V}_m) = (M_1 x_1 + M_2 x_2) / \rho$$

$$\text{Rao's constant: } (R) = (M/\rho)$$

$$\text{Wada's constant: } (W) = (M_{\text{eff}}/\rho)^{-1/7}$$

4. RESULTS AND DISCUSSION

Thermo-acoustical properties of binary solution ably reflect the molecular interactions. The experimental parameter for the binary mixtures showed an increasing trend with increase in mole fraction of alpha terpinal acetate (α -TA), whereas with increase in temperature, there was a decline in the values of the said parameters (Table-1), which probably is due to thermal agitation between the molecules of solute and solvent of binary mixture [34]. In case of derived parameters (Table-2), acoustic impedance (Z) reflected an increase in value with increasing mole fraction of binary solutions, this increase in acoustic impedance can be attributed due to significant interaction between the molecules of the mixture, whereas with a rise in temperature the acoustic impedance reflected a gradual decrease [29].

Another computed parameter, adiabatic compressibility (β_s), recorded a reduction in values with increase in mole fraction of α -TA. On the contrary, this parameter increased with rise in temperature, which can be due to aggregation of solvent molecules enveloping solute molecules [7]. The derived parameter, intermolecular free length (L_f) (Table-2), indicates the approximate space among the constituting atoms of a molecular system and may help in predicting intermolecular interaction between the binary mixture components [35]. The intermolecular free length value showed a decreasing trend with increasing mole fraction of α -TA whereas in each fraction rise in temperature showed enhanced values of intermolecular free length. This may be on account of thermal expansion of liquids [36,37].

Another calculated parameter (Table-3), free volume (V_f) can be defined as an available space for the movement of a molecule center, which experiences repulsion from neighboring molecules. Another important parameter molar volume (V_m) indicates average space occupied per mole of the substance. The enhancement in mole fraction of α -TA increases both free volume and molar volume whereas rise in temperature in each mole fraction results in decrease of free volume yet shows an increase in molar volume. These changes can be based on due to interaction between solvent and solute molecules. Rao's constant (R) and Wada's constant (W) showed enhanced value with the increase in concentration of α -TA as well as temperature [34,38]

Internal pressure (π_i), (Table-4), gives the measure of the cohesion of the molecules, which results due to attractive and repulsive forces between the molecules of a liquid. The internal pressure showed a decreasing trend with increasing mole fraction of α -TA. Similar trend was observed with rise in temperature. This cause may be due to weakening of cohesion forces which reflects a downward trend in internal pressure [29, 39].

The relaxation time (τ), another important formulated parameter which shows time taken by a system to reach equilibrium after being in stage of disturbance. An increase in mole fraction of α -TA results in a stretched relaxation time, possibly due to more time involved in rearranging to a proper state from a random state. With temperature no set trend was observed regarding relaxation time.

Table-1: The measured values of Ultrasonic velocity (U), Density(ρ) and Viscosity, (η) at different mole fractions and temperatures of binary mixtures.

Mole Fraction X_A	Ultrasonic velocity, U(ms ⁻¹), Density, ($\rho \times 10^3$ kg/m ³), Viscosity, η (mPa.s)											
	293.15K			298.15K			303.15K			308.15K		
	U	ρ	η	U	ρ	η	U	ρ	η	U	ρ	η
0.0000	1230	0.8191	2.9500	1208	0.8099	2.5700	1186	0.8083	2.2800	1164	0.7999	2.0300
0.0996	1278	0.8301	1.6235	1241	0.8202	1.5081	1228	0.8115	1.3661	1217	0.8011	1.2887
0.2281	1286	0.8495	1.8934	1250	0.8392	1.8081	1239	0.8301	1.7214	1231	0.8197	1.6012
0.3071	1295	0.8631	1.9983	1261	0.8541	1.9031	1249	0.8451	1.8101	1242	0.8318	1.7139
0.4162	1306	0.8789	2.1533	1272	0.8702	2.0081	1259	0.8611	1.9294	1250	0.8519	1.8094
0.5122	1312	0.8959	2.2208	1281	0.8862	2.1201	1269	0.8761	1.9801	1260	0.8641	1.9012
0.6396	1322	0.9212	2.3006	1291	0.9062	2.1931	1281	0.8962	2.0599	1271	0.8799	1.9509
0.7418	1334	0.9402	2.5021	1302	0.9221	2.3902	1289	0.9099	2.2012	1280	0.8981	2.1108
0.8401	1344	0.9591	2.7587	1312	0.9399	2.6441	1299	0.9292	2.5085	1291	0.9198	2.3971
0.9201	1353	0.9699	3.3091	1325	0.9499	3.1959	1313	0.9399	3.0109	1304	0.9305	2.8902
1.0000	1370	0.9806	3.6965	1336	0.9599	3.5991	1321	0.9497	3.4031	1313	0.9389	3.3106

Table-2- The values of derived parameters acoustic impedance (Z), adiabatic compressibility (β_s), intermolecular free length (L_f), at temperatures of binary mixtures.

Mole Fraction X_A	Acoustic Impedance (Z) gm/m ² s, Adiabatic compressibility (β_s) $\times 10^{-10}$, Intermolecular Free Length (L_f) $\times 10^{-10}$											
	293.15K			298.15K			303.15K			308.15K		
	Z	β_s	L_f	Z	β_s	L_f	Z	β_s	L_f	Z	β_s	L_f
0.0000	1007.9	8.07	0.1751	0978.5	8.46	0.1809	0958.4	8.80	0.1861	0931.8	9.23	0.1923
0.0996	1060.6	7.38	0.1674	1017.6	7.92	0.1750	0996.2	8.17	0.1794	0974.4	8.43	0.1838
0.2281	1092.5	7.12	0.1645	1049.1	7.63	0.1718	1011.1	7.85	0.1758	1009.5	8.05	0.1796
0.3071	1117.1	6.91	0.1620	1077.2	7.36	0.1688	1053.3	7.59	0.1728	1033.9	7.79	0.1767
0.4162	1147.4	6.67	0.1592	1106.9	7.10	0.1658	1084.2	7.33	0.1698	1064.7	7.51	0.1735
0.5122	1175.2	6.48	0.1570	1135.2	6.88	0.1631	1111.7	7.09	0.1671	1088.6	7.29	0.1709
0.6396	1217.2	6.21	0.1537	1169.1	6.62	0.1601	1148.3	6.80	0.1636	1118.5	7.04	0.1678
0.7418	1254.2	5.98	0.1508	1200.7	6.40	0.1573	1172.6	6.61	0.1614	1149.6	6.80	0.1651
0.8401	1289.3	5.77	0.1482	1233.4	6.18	0.1547	1207.3	6.38	0.1585	1187.6	6.52	0.1617
0.9201	1312.7	5.63	0.1463	1258.1	6.01	0.1523	1234.8	6.17	0.1559	1213.7	6.32	0.1591
1.0000	1343.2	5.43	0.1437	1282.2	5.84	0.1503	1254.5	6.03	0.1542	1232.7	6.18	0.1573

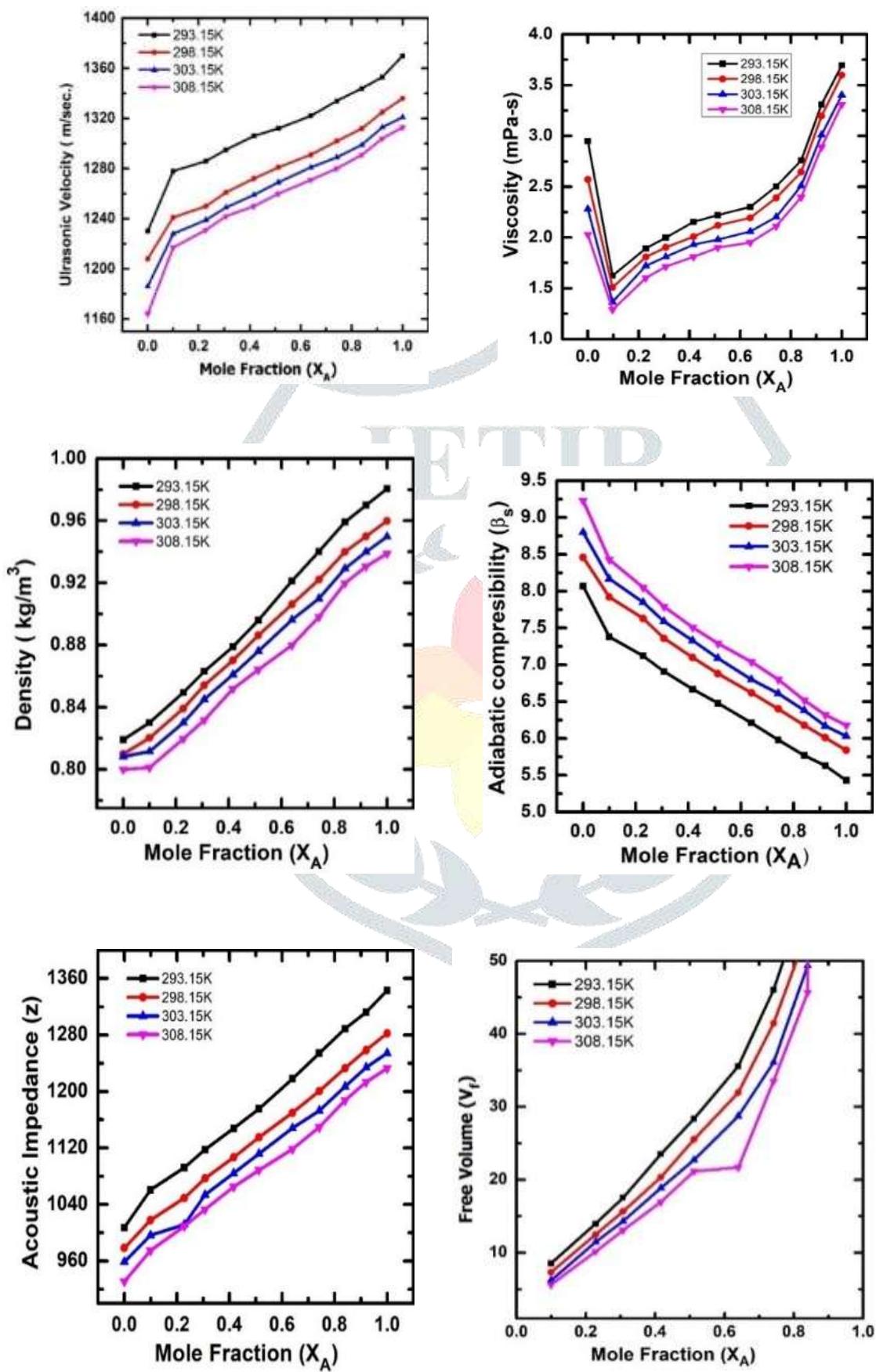
Table-3- The values of derived parameters Free volume (V_f), Molar volume (V_m), Rao's constant (R), at temperatures of binary mixtures.

Mole Fraction X_A	Free Volume (V_f), Molar Volume (V_m) $\times 10^{-3} \cdot m^3$, Rao's Constant (R) $\times 10^3$											
	293.15K			298.15K			303.15K			308.15K		
	V_f	V_m	R	V_f	V_m	R	V_f	V_m	R	V_f	V_m	R
0.0000	08.552	10.398	9.6954	07.327	10.523	9.7467	06.218	10.631	9.7064	05.621	10.771	9.7473
0.0996	13.971	12.005	1.1280	12.494	12.158	1.1305	11.453	12.281	1.1386	10.175	12.449	1.1499
0.2281	17.531	12.935	1.3055	15.655	13.078	1.3091	14.315	13.211	1.3195	13.078	13.422	1.3334
0.3071	23.521	14.215	1.4098	20.361	14.367	1.4121	18.882	14.514	1.4226	16.965	14.661	1.4426
0.4162	28.377	15.258	1.5541	25.537	15.428	1.5559	22.726	15.606	1.5670	21.155	15.813	1.5801
0.5122	35.576	16.523	1.6703	31.955	16.809	1.6752	28.751	16.984	1.6892	21.689	17.301	1.7086
0.6396	46.035	17.523	1.8140	41.445	17.862	1.8295	36.081	18.108	1.8451	33.526	18.347	1.8744
0.7418	59.896	18.422	1.9289	54.207	18.807	1.9509	49.348	19.022	1.9704	45.673	19.216	1.9917
0.8401	86.161	19.236	2.0337	79.252	19.635	2.0587	71.489	19.845	2.0755	66544	20.041	2.0924
0.9201	08.552	10.398	2.1270	07.327	10.523	2.1657	06.218	10.631	2.1731	05.621	10.771	2.1900
1.0000	13.971	12.005	2.2232	12.494	12.158	2.2522	11.453	12.281	2.2678	10.175	12.449	2.2892

Table-4- The values of derived parameters Wada's constant (W), Internal Pressure (π_i), Pax $\times 10^{-6}$, Relaxation Time (τ) $\times 10^{-12}$ Sec.

Mole Fraction X_A	Wada's constant (W) $\times 10^3$, Internal Pressure (π_i) Pax $\times 10^{-6}$, Relaxation Time (τ) $\times 10^{-12}$ Sec.											
	293.15K			298.15K			303.15K			308.15K		
	W	π_i	τ	W	π_i	τ	W	π_i	τ	W	π_i	τ
0.0000	0.0671	2.8464	3.1741	0.0672	2.7061	2.8995	0.0674	2.6121	2.6739	0.0674	2.5114	2.4974
0.0996	0.0781	1.7504	1.5966	0.0782	1.6983	1.5918	0.0794	1.6134	1.4884	0.0794	1.5606	1.1448
0.2281	0.0906	1.5746	1.8385	0.0909	1.5481	1.7969	0.0923	1.5062	1.7811	0.0923	1.4452	1.7187
0.3071	0.0981	1.4660	1.8407	0.0982	1.4397	1.8383	0.1011	1.4009	1.8306	0.1011	1.3526	1.7809
0.4162	0.1084	1.3447	1.9152	0.1085	1.3071	1.9016	0.1099	1.2789	1.8847	0.1099	1.2340	1.8124
0.5122	0.1168	1.2429	1.9201	0.1171	1.2201	1.9138	0.1191	1.1757	1.8713	0.1191	1.1455	1.8478
0.6396	0.1273	1.1321	1.9352	0.1282	1.1063	1.9060	0.1309	1.0684	1.8675	0.1309	1.0312	1.8299
0.7418	0.1357	1.0867	1.9939	0.1371	1.0613	1.9887	0.1394	1.0145	1.9413	0.1394	0.9883	1.9126
0.8401	0.1434	1.0612	2.1231	0.1449	1.0375	2.1190	0.1471	1.0078	2.1031	0.1471	0.9816	2.0848
0.9201	0.1502	0.7832	2.4849	0.1519	0.7670	2.4551	0.1541	0.7426	2.3775	0.1541	0.7252	2.2355
1.0000	0.1571	1.0927	2.6779	0.1589	1.0948	2.6008	0.1611	1.0808	2.5379	0.1611	1.0787	2.4270

Fig 2.: Plots for measured values of density (ρ), ultrasonic velocity (U), viscosity and derived values of acoustic impedance, adiabatic compressibility, and intermolecular free length in terms of mole fractions at different temperatures.



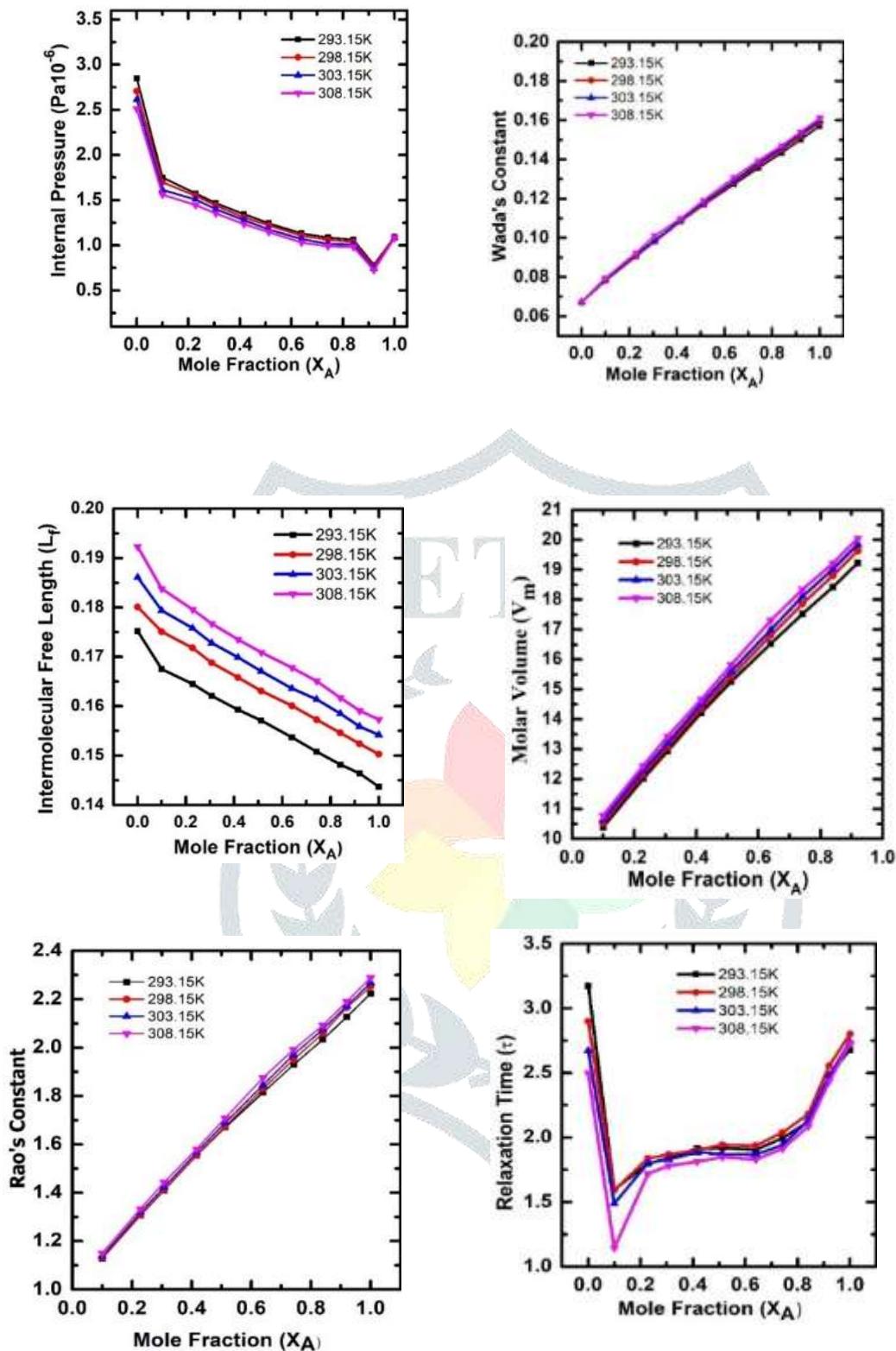


Fig. 3: Plots for derived values of free volume, molar volume, Rao's constant, Wada's constant, internal pressure and Relaxation time in terms of mole fractions at different temperatures.

CONCLUSION

The values of measured parameters ($\rho, \eta, \& U$) and acoustic impedance showed an enhancement with mole fraction. whereas the rise in temperature leads to decline of these values, reflecting a structural change where the molecules are de-clustering apart. The linear increase in β_s and L_f with mole fraction and these values exhibited decrease with rise in temperature. Moreover, rise in relaxation

time and fall in internal pressure both with respect to mole fraction concentration as well as temperature, points towards weak molecular interactions coupled with diminished cohesive forces in the chosen solute and solvent system undertaken.

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CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

All the authors contributed significantly to this manuscript, participated in reviewing/editing, and approved the final draft for publication. The research profile of the authors can be verified from their ORCID IDs, given below:

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