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ULTRASONIC STUDY OF BINARY MIXTURES OF DHMOH AND ACETONE AT 288.15-313.15 K

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ABSTRACT

The density (ρ), viscosity (η), and ultrasonic velocity (u) have been measured in the binary solutions of Dihydromyrcenol (DHMOH) with acetone at different temperatures: 288.15−313.15 K to understand the molecular interaction between these solutions. From the measured values, various acoustical parameters such as adiabatic compressibility (κ), acoustic impedance (Z), molecular free length (L_f), molar volume (V_m), free volume (V_f), Relative association (R_A), internal pressure (π_i), and molar sound velocity (U_m) were determined and correlated with concentration at different temperatures. The observed behaviour of these parameters concerning different compositions is discussed in terms of their molecular interactions between the unlike molecules of the binary solutions. The variations in measured and calculated values with concentration and temperature confirmed the presence of molecular interaction in the mixtures and were further confirmed by relative association.

Keywords: Ultrasonic Velocity, Viscosity, Density, Acetone, Intermolecular Interactions

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INTRODUCTION

In recent years many researchers have shown an extensive interest in studying the molecular interactions in binary liquids. The experimental determination of density, viscosity, and ultrasonic velocity offers a more useful tool for understanding the behaviour of the liquid state because of the close connection between the molecular structure and macroscopic properties of the liquid. The physicochemical properties of binary mixtures play a vital role in understanding the molecular structure as well as the other macroscopic properties of the mixtures. These properties of binary mixtures depend upon their composition and find many applications in different chemical, industrial, and biological processes.[1,2] It is for this reason that the investigation of the intermolecular interactions that occur in binary liquid mixtures is very significant. The fundamental mechanisms are still relatively little known at a precise molecular level in the case of a binary mixture of fragrance molecules. In order to study the thermodynamic properties and molecular interaction in the binary mixtures of dihydromyrcenol (*DHMOH, 2,6-dimethyl-7-octen-2-ol*) with acetone, which is the extension of our recent study[2] to cover large range of temperature, the ultrasonic velocity, density and viscosities are measured for all compositions at 288.15, 293.15, 298.15, 303.15, 308.15 K and 313.15 K. Dihydromyrcenol has a stable and powerful lime-like aroma. Due to this reason, it is widely used as a fragrance ingredient in making soaps, detergents, shampoos, lotions, and cleaning agents.[3] The molecular structure of Dihydromyrcenol is shown in Fig.-1. The molecules of acetone have a common carbonyl functional (C=O) group. It is the simplest and smallest ketone having a polar aprotic nature. Acetone is a colourless liquid solvent miscible with water. So, it is widely used in manufacturing industrial products, cosmetics, and personal care products. Thus, the growing demand of acetone and dihydromyrcenol in various products emphasizes its need to study the thermodynamic properties of liquids and their binary mixtures at different temperatures. To our best knowledge, the measurement of density, viscosity, and ultrasonic velocity for the present binary mixture are reported first time. Hence keeping in mind this fact, to study the nature of molecular interaction in the binary mixtures of dihydromyrcenol with acetone, experiments have

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been performed to measure the density, viscosity, and ultrasonic velocity at different temperatures with the help of a circulating water bath (288.15, 293.15, 298.15, 303.15, 308.15, 313.15 K). By using the experimental values of density, viscosity, and ultrasonic velocity, the thermodynamic parameters such as adiabatic compressibility (κ), acoustic impedance (Z), molecular free length (L_f), molar volume (V_m), free volume (V_f), Relative association (R_A), internal pressure (π_i) and molar sound velocity (U_m) have been calculated by using the standard relation[1,2,4-7] discussed in results and discussion section. The variations in the calculated thermodynamic parameters are further discussed in terms of molecular interaction between the components of the binary mixtures over the range of 0.0 to 0.1 mole/lt range. Thus, the present study gives a better understanding of the intermolecular interactions in the present binary system.

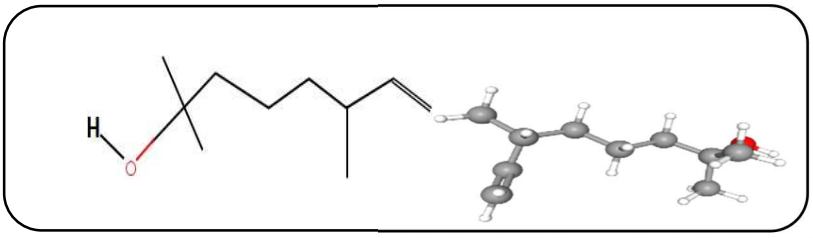

Fig.-1: Structure of Dihydromyrcenol (2,6-dimethyl-7-octen-2-ol) in 2D and 3D

Table-1: The Measured Values of Density, Viscosity, and Ultrasonic Velocity, Along with Values Available in the Publications at 303.15 K

Liquids	Density (ρ) kg/m^3		Viscosity (η) mPa.s		u (ms^{-1})	
	Expt.	Lit.	Expt.	Lit.	Expt.	Lit.
1-propanol	795.42	798.00 [13] 795.71 [18]	1.590	1.610 [13]	1191.2	1189.2 [13] 1189.8 [18]
2-propanol	777.75	776.80 [11,12] 776.50 [18]	1.812	1.800 [11,12]	1127.6	1126.3 [18]
1-butanol	802.80	802.60 [8] 802.00 [9] 804.00 [13]	2.104	2.222 [8] 2.245 [9] 2.054 [13]	1228.8	1228.1 [8] 1228.4 [13]
2-butanol	796.42	798.80 [19] 798.80 [20]	2.352	2.553 [20]	1193.6	1194.0 [19]
Benzene	866.14	867.70 [10] 867.80 [16]	0.568	0.571 [16]	1281.1	1278.3 [10] 1295.2 [16]
Toluene	854.23	853.90 [10] 857.80 [16] 851.60 [17]	0.531	0.526 [16] 0.534 [17]	1284.0	1285.3 [10] 1287.2 [16] 1279.5 [17]
Cyclohexane	769.21	553.50 [10] 767.70 [16]	0.812	0.801 [16]	1232.8	1230.3 [10] 1230.3 [16]
Acetone	780.23	779.00 [17] 780.33 [19]	0.313	0.292 [17] 0.295 [19]	1142.2	1141.2 [17]
1,4 Dioxane	1021.8	1022.7 [15]	1.077	1.088 [14] 1.054 [15]	1348.8	1341.0 [14] 1356.0 [15]

EXPERIMENTAL

The chemical liquid used as a solvent was purchased from S. D. Fine Chemicals, India and Dihydromyrcenol (DHMOH) was from Hina Chemicals, Gujarat (India). It was necessary to weigh the liquids in specially designed weighing bottles with ground glass stoppers before preparing binary mixtures. After that, a stock solution was prepared which is diluted to different desired mole fraction. An electronic balance of sensitivity 0.0001 g was used for weighing appropriate mass of the solute. The densities and viscosities of the liquids were measured at all the experimental temperature values using a specific gravity bottle and a Cannon Fenske viscometer. A stopwatch, with an accuracy of ±0.01 sec, is used to measure the time it takes for liquids to flow through a Cannon Fenske viscometer in a certain amount of time. An electrically operated constant temperature circulating water bath (Mittal Enterprise, New Delhi) with an accuracy of ±0.1°C was used to keep the temperature constant during the experimental observations. The

purity of the solvent was ensured by the comparison of their densities with the literature values. These values very well match the experimental error. For the accuracy in the experimental values, first of all, the calibration of the experimental apparatus was done by the determination of density (ρ), viscosity (η) and ultrasonic velocity (u) for some pure liquids 1-propanol, 2-propanol, 1-butanol, Benzene, Toluene, Cyclohexane, Acetone, and 1,4 Dioxan at 303.15 K are measured and represented in Table-1. The ultrasonic velocities were determined using a single-frequency ultrasonic interferometer of frequency 2 MHz (Model: F-05 Mittal Enterprise, New Delhi). The capacity of the measuring cell is 10 mL and the digital micrometer has the least count of 0.001 mm. The ultrasonic interferometer was also calibrated with the pure liquids. The measured values of 'ρ', 'η', and 'u' of the binary mixtures of DHMOH and acetone as a function mole and different temperatures are shown in Table-II.

The measured values are further used to calculate the derived parameters, such as adiabatic compressibility (κ), acoustic impedance (*Z*), molecular free length (L_f), molar volume (V_m), free volume (V_f), Relative association (R_A), internal pressure (π_i) and molar sound velocity (U_m) at different concentration and temperatures. The expressions used for the calculation of derived parameters, which are available in literature[1, 4-7] are as follows-

Adiabatic compressibility $$\kappa = \frac{1}{\rho u^2} \quad (1)$$

Acoustic impedance $$Z = \rho u \quad (2)$$

Molecular free length

$$L_f = \frac{K_T}{u\rho^{1/2}} \quad (3)$$

where K_T is a temperature-dependent Jacobson constant and given by $K = (93.875 + 0.345T)\times10^{-8}$.

The molar volume of the liquid mixture

$$V_m = \frac{\bar{M}}{\rho} \quad (4)$$

Where $\bar{M}$ is represents the mean molecular weight of the mixture.

Free volume

$$V_f = \frac{\bar{M}u}{k\eta} \quad (5)$$

Where k is a temperature-independent constant, and its value is 4.28×10^{-8} for all liquids[5] and η is experimental viscosity.

Relative association $$R_A = \frac{\rho}{\rho_o}\left(\frac{u_o}{u}\right)^{1/3} \quad (6)$$

Where, ρ_o and u_o are the remeasured values of density and ultrasonic velocity in pure solvent and ρ & u are the measured values of the same parameters in a binary mixture.

On the basis of dimensional analysis[7], the following expression can be used to determine the internal pressure

$$\pi_i = bRT\left[\frac{k\eta}{u}\right]^{1/2}\left[\frac{\rho^{2/3}}{\bar{M}^{7/6}}\right] \quad (7)$$

here, b is a packing factor whose value is taken 2, $k = 4.28\times10^{-8}$ and R is the universal gas constant.

The molar sound velocity

$$U_m = u^{1/3}V \quad (8)$$

Table-2: The Measured Values of Density (ρ), Viscosity (η), and Ultrasonic Velocity (u) of Binary Mixtures in Terms of Moles at Different Temperatures

Conc. (M)	Density, ($\rho\times10^3$ kg/m^3), at different Temperature (K)					
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K
0.0	0.7960	0.7907	0.7851	0.7802	0.7761	0.7706
0.01	0.7966	0.7914	0.7857	0.7808	0.7770	0.7715
0.02	0.7972	0.7920	0.7864	0.7814	0.7775	0.7721

0.03	0.7977	0.7925	0.7870	0.7821	0.7781	0.7726
0.04	0.7984	0.7931	0.7877	0.7827	0.7786	0.7732
0.05	0.7991	0.7938	0.7885	0.7835	0.7794	0.7738
0.06	0.7998	0.7943	0.7891	0.7842	0.7801	0.7744
0.07	0.8005	0.7950	0.7899	0.7850	0.7806	0.7751
0.08	0.8011	0.7957	0.7906	0.7857	0.7812	0.7759
0.09	0.8017	0.7963	0.7914	0.7866	0.7819	0.7764
0.1	0.8024	0.7969	0.7922	0.7874	0.7825	0.7771
	Viscosity, η (mPa.s), at different Temperature (K)					
0.0	0.3284	0.3217	0.3157	0.3132	0.2990	0.2842
0.01	0.3318	0.3251	0.3191	0.3164	0.3024	0.2874
0.02	0.3352	0.3285	0.3225	0.3198	0.3058	0.2908
0.03	0.3389	0.3322	0.3260	0.3235	0.3095	0.2945
0.04	0.3428	0.3361	0.3301	0.3274	0.3134	0.2984
0.05	0.3459	0.3392	0.3332	0.3305	0.3165	0.3015
0.06	0.349	0.3423	0.3363	0.3336	0.3196	0.3046
0.07	0.3518	0.3451	0.3391	0.3364	0.3224	0.3074
0.08	0.3546	0.3479	0.3419	0.3392	0.3252	0.3102
0.09	0.3578	0.3511	0.3451	0.3424	0.3284	0.3134
0.1	0.3606	0.3539	0.3479	0.3452	0.3312	0.3162
	Ultrasonic velocity, u (ms^{-1}), at different Temperature (K)					
0.0	1175.00	1162.00	1153.21	1142.20	1122.00	1110.00
0.01	1182.20	1167.20	1156.30	1145.31	1128.06	1114.41
0.02	1187.80	1171.30	1161.31	1150.11	1135.70	1118.18
0.03	1193.10	1176.30	1166.40	1155.20	1139.12	1123.28
0.04	1198.16	1182.48	1172.81	1160.28	1143.88	1128.48
0.05	1201.80	1185.70	1176.50	1165.12	1149.08	1133.50
0.06	1206.00	1192.84	1182.73	1171.66	1155.28	1136.81
0.07	1212.20	1196.80	1186.40	1176.68	1160.12	1143.90
0.08	1216.60	1204.64	1193.74	1180.44	1164.02	1146.53
0.09	1224.40	1209.00	1199.30	1186.04	1169.56	1153.78
0.1	1232.00	1214.88	1203.11	1193.27	1174.92	1159.20

Table-3: Calculated Values of Different Acoustical Parameters at Various Concentrations and Temperatures

Conc. (M)	Adiabatic compressibility $\kappa \times 10^{-10}$ (m^2/N)					
0.0	9.099	9.366	9.578	9.825	10.235	10.532
0.01	8.982	9.274	9.519	9.764	10.113	10.437
0.02	8.890	9.203	9.429	9.675	9.972	10.359
0.03	8.806	9.119	9.340	9.581	9.904	10.258
0.04	8.724	9.017	9.230	9.490	9.816	10.156
0.05	8.664	8.960	9.163	9.401	9.717	10.058
0.06	8.596	8.848	9.059	9.289	9.605	9.992
0.07	8.501	8.781	8.994	9.201	9.518	9.859
0.08	8.433	8.660	8.876	9.134	9.448	9.805
0.09	8.320	8.591	8.785	9.037	9.349	9.675
0.1	8.210	8.502	8.720	8.919	9.258	9.576
	Acoustical impedance $Z \times 10^5$ (Kg m^{-2} s^{-1})					
0.0	9.353	9.188	9.054	8.911	8.708	8.554

0.01	9.417	9.237	9.085	8.943	8.765	8.598
0.02	9.469	9.277	9.133	8.987	8.830	8.633
0.03	9.517	9.322	9.180	9.035	8.863	8.678
0.04	9.566	9.378	9.238	9.082	8.906	8.725
0.05	9.604	9.412	9.277	9.129	8.956	8.771
0.06	9.646	9.475	9.333	9.188	9.012	8.803
0.07	9.704	9.515	9.371	9.237	9.056	8.866
0.08	9.746	9.585	9.438	9.275	9.093	8.896
0.09	9.816	9.627	9.491	9.329	9.145	8.958
0.1	9.886	9.681	9.531	9.396	9.194	9.008
	Intermolecular free length $L_f \times 10^{-11}$ m					
0.0	6.0912	6.2374	6.3653	6.5056	6.7002	6.8576
0.01	6.0519	6.2068	6.3459	6.4854	6.6603	6.8265
0.02	6.0211	6.1828	6.3157	6.4559	6.6134	6.8008
0.03	5.9924	6.1546	6.2857	6.4246	6.5910	6.7677
0.04	5.9645	6.1201	6.2486	6.3940	6.5615	6.7339
0.05	5.9438	6.1008	6.2259	6.3642	6.5284	6.7015
0.06	5.9206	6.0623	6.1907	6.3258	6.4905	6.6794
0.07	5.8877	6.0396	6.1684	6.2956	6.4613	6.6350
0.08	5.8642	5.9977	6.1278	6.2728	6.4372	6.6164
0.09	5.8247	5.9738	6.0963	6.2396	6.4038	6.5727
0.1	5.7862	5.9426	6.0739	6.1986	6.3722	6.5390
	Relative association, R_A,					
0.0	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000
0.01	0.9987	0.9994	0.9999	0.9999	0.9994	0.9998
0.02	0.9979	0.9990	0.9993	0.9992	0.9978	0.9995
0.03	0.9970	0.9982	0.9986	0.9987	0.9975	0.9986
0.04	0.9965	0.9972	0.9977	0.9980	0.9968	0.9979
0.05	0.9964	0.9972	0.9977	0.9976	0.9963	0.9972
0.06	0.9961	0.9958	0.9967	0.9966	0.9954	0.9970
0.07	0.9953	0.9956	0.9966	0.9962	0.9947	0.9958
0.08	0.9948	0.9943	0.9955	0.9961	0.9943	0.9961
0.09	0.9934	0.9939	0.9949	0.9956	0.9936	0.9946
0.1	0.9922	0.9930	0.9949	0.9946	0.9929	0.9940
	Molar Volume, $V_m \times 10^{-4}$ (m^3/mol)					
0.0	0.7296	0.7345	0.7398	0.7444	0.7484	0.7537
0.01	0.7414	0.7463	0.7517	0.7564	0.7601	0.7655
0.02	0.7532	0.7581	0.7635	0.7684	0.7723	0.7777
0.03	0.7650	0.7700	0.7754	0.7803	0.7843	0.7899
0.04	0.7766	0.7818	0.7872	0.7922	0.7964	0.8020
0.05	0.7883	0.7935	0.7989	0.8040	0.8082	0.8140
0.06	0.7998	0.8054	0.8107	0.8158	0.8200	0.8261
0.07	0.8114	0.8170	0.8223	0.8274	0.8321	0.8380
0.08	0.8231	0.8286	0.8340	0.8392	0.8440	0.8498
0.09	0.8347	0.8404	0.8456	0.8507	0.8558	0.8619
0.1	0.8462	0.8520	0.8571	0.8623	0.8677	0.8737
	Free Volume, $V_f \times 10^{-7}$ (m^3/mol)					
0.0	3.383	3.432	3.490	3.481	3.634	3.859
0.01	3.448	3.487	3.536	3.530	3.693	3.914
0.02	3.505	3.538	3.591	3.584	3.761	3.962
0.03	3.556	3.587	3.644	3.633	3.802	4.011

0.04	3.603	3.639	3.693	3.679	3.845	4.055
0.05	3.656	3.690	3.746	3.737	3.906	4.116
0.06	3.712	3.759	3.811	3.803	3.971	4.166
0.07	3.781	3.818	3.869	3.868	4.036	4.244
0.08	3.843	3.896	3.945	3.925	4.095	4.297
0.09	3.913	3.950	4.005	3.986	4.155	4.367
0.1	3.990	4.019	4.064	4.061	4.222	4.435

RESULTS AND DISCUSSION

The measured values of density, viscosity, and ultrasonic velocity of DHMOH with acetone at temperatures are reported in Table-2. These measured values of fundamental parameters are plotted as a function of molecular concentration and are shown in Figs.-2(a-b) and Fig.-3. Intermolecular interactions are influenced by the strength of the forces that act among the molecules of the solvent and solute. As a result, the intermolecular motion of the molecules is also affected accordingly as well. Attractive forces cause the molecular association in which a solute molecule is encircled by solvent molecules. Various intermolecular forces may be responsible for solute-solvent interactions such as hydrogen bonding, dipole-dipole interactions, and van der Waals forces and form a solvation layer around the solute molecule. These interactions may lead to the modification of the solute for the stabilization in the solvent.

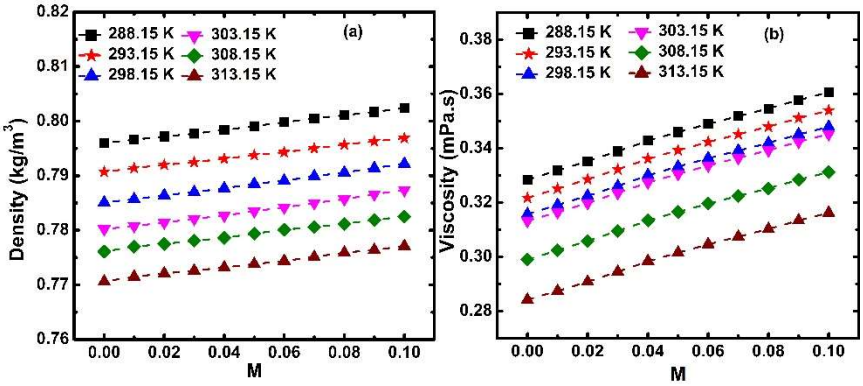

Fig.-2: Measured Values of Density, and Viscosity at Different Molar Concentrations and Temperatures

From the Figure, it can be seen that the measured values of ρ, η and u increase with the increase in molar concentration of DHMOH. From the figures, it can be seen that the values of ρ, η and u increase linearly with concentration confirming an increase in cohesive forces. This may be due to the existence of strong molecular integration. The values of ρ, η and u of the solution increased by 0.9%, 10.2%, and 4.3% at 298.15 K temperature respectively, for the adopted experimental concentration. Further, the measured values of these parameters in the binary mixtures decrease with the increase in temperature. This shows that the intermolecular forces decrease may be due to increase in thermal energy producing agitation in molecule of the binary mixture. As a result, the volume expands, which then increases the free length. These results indicate the presence of intermolecular interaction between the components of binary mixtures. The variation observed in the measured values is different and is not characteristic of an ideal mixture and may be due to the presence of intermolecular interaction in the systems.[2,6]

For a better understanding of the molecular interaction between DHMOH and acetone, as well as the effect of concentration on the molecular interaction, the acoustical parameters such as adiabatic compressibility (κ), acoustic impedance (*Z*), molecular free length (L_f), molar volume (V_m), free volume (V_f), Relative association (R_A), internal pressure (π_i) and molar sound velocity (U_m) have been calculated at all the six different temperatures and given in Table-3. The calculated values of these parameters are plotted as a function of molar concentration at different temperatures and given in Figs.-4(a-b) and Figs.-5(a-f). From the results presented in these figures, it can be seen that adiabatic compressibility (K), free length (L_f) and internal pressure (π_i) are decreased with the molecular concentration which is expected as clear from equations as discussed earlier. It is well known that intermolecular free length is the most important factor that predicts the changes in velocity in mixtures.

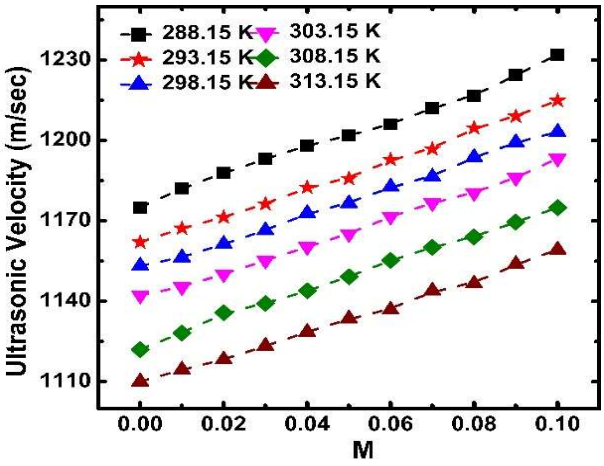

Fig.-3: Measured Values of Ultrasonic Velocity at Different Molar Concentrations and Temperatures

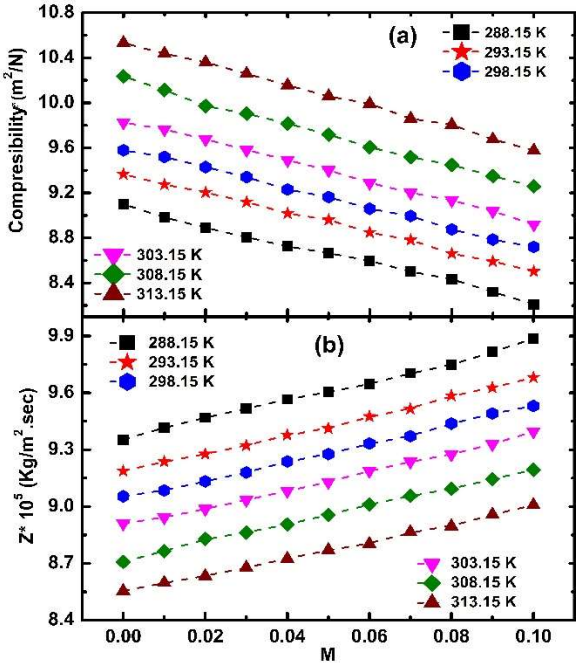

Fig.-4: Variation in Adiabatic Compressibility (κ), and Acoustic Impedance (*Z*) at Different Molar Concentrations and Temperatures

Further, Eyring and Kincaid model.[7,21,22] proposed that ultrasonic velocity increases with the decrease in intermolecular free length and vice-versa when mixing of components. Similar trends have been in the present study for intermolecular free length and ultrasonic velocity. From Tables-2 & 3, it can be seen that measured values of ultrasonic velocity increase and intermolecular free length decreases with the molar concentration. Further, the decrease in internal pressure (π_i) with concentration of DHMOH may be due to the weak dipole-induced-dipole type interaction.[6] Acoustic impedance is characterized by restrictions on the movement of the sound waves. From Table-3 and Fig.-4(b), it can be seen that the value of acoustic impedance decreases with the increase in temperature and increases with the increase in molar concentration. The changes observed in acoustic impedance with temperature and molar concentration further verify the existence of intermolecular interaction between the molecules of binary mixtures. The relative association (R_a) is a measure of the association of the components. For the present binary system, R_a shows an irregular trend. This signifies that like interactions (solute-solute or solvent-solvent) are relatively stronger as compared to unlike interactions. Similar trends are also found in the literature.[1,5,23,24] The molar volume (V_m) and molar sound velocity (U_m) are found to increase with the concentration of DHMOH.

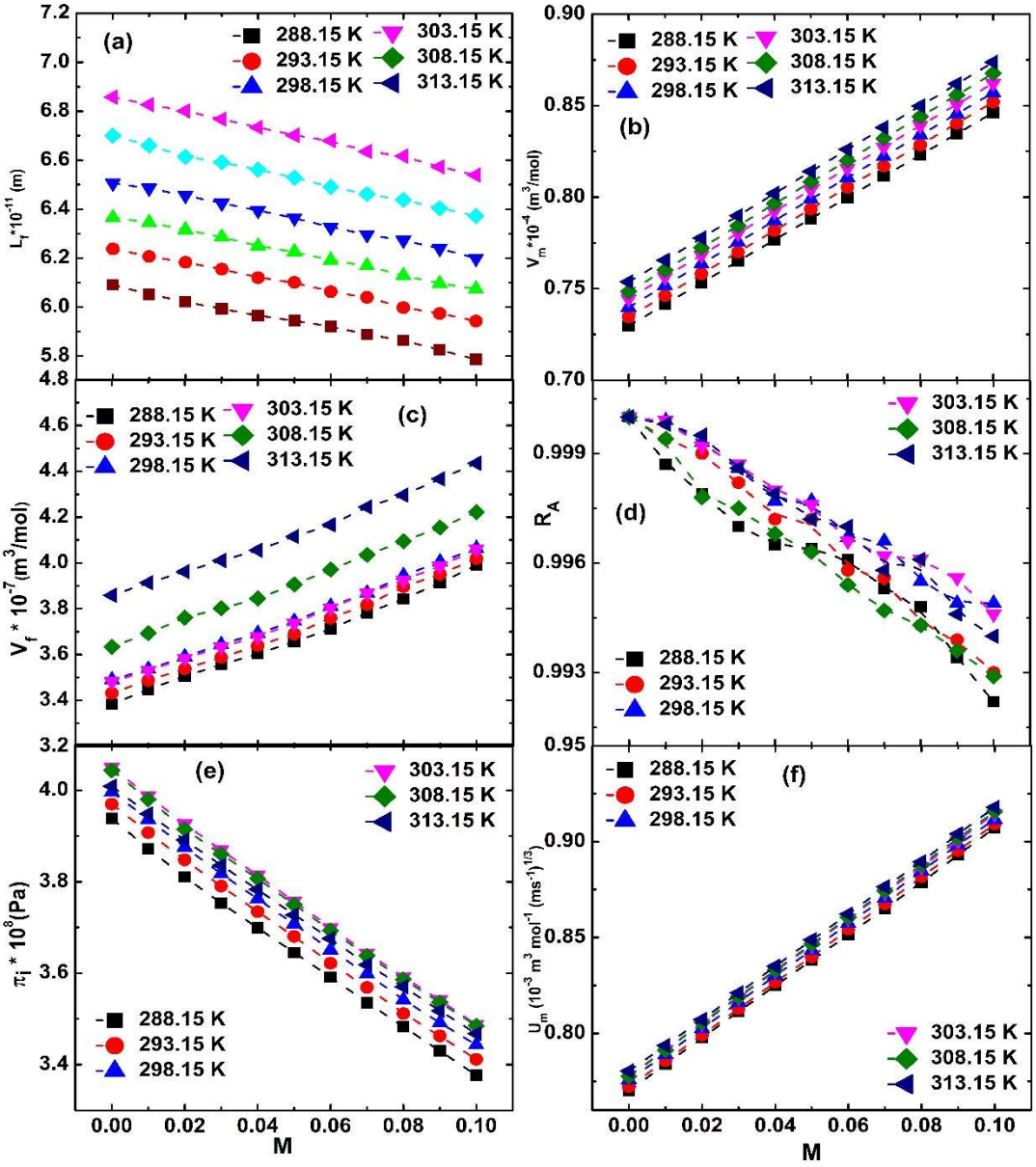

Fig.-5: Variation in Molecular Free Length (L_f), Molar Volume (V_m), Free Volume (V_f), Relative Association (R_A), Internal Pressure (π_i), and Molar Sound Speed (U_m) with Molar Concentration and Different Temperatures (288.15-313.15 K)

CONCLUSION

In this work, we have experimentally determined the density, viscosity, and ultrasonic velocity of sound in the binary mixtures of acetone and DHMOH at said temperatures in the range of 0.0 to 0.1 mole/lt. The measurements of these parameters were found to increase with concentration whereas decrease with temperature. It is also observed that the measured values of ultrasonic velocity showed a significant increment with the concentration of DHMOH whereas the intermolecular free length decreases. On the contrary, ultrasonic velocity showed a reduction in values with the increase in temperature which may be due to the rise in thermal energy of the binary system. The decrease in adiabatic compressibility suggests a formation of a solvation layer around solute molecules which indicates a marked molecular interaction in binary mixtures. The irregular decreasing trend of relative association (R_a) suggests that like interactions are relatively stronger than unlike interactions.

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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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