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# Acoustic and viscosity studies of the binary mixtures of Parabens with alcohols at T= (298.15-323.15) K

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## ABSTRACT

Volumetric and viscometry studies of mixtures containing benzene with 0.01m (molality) of butyl/pentyl paraben in the isopropanol solution were measured in the temperature range 298.15K to 323. 15K.The experimental data have been utilized to calculate the acoustic parameters such as ultrasonic attenuation, relative strength, surface tension, relaxation time and Gibb's free energy. There is a linear decrease in the relative strength of the mixture, surface tension increases in the concentration of benzene in both the systems. The linear variation indicates that the molecules are not very proximal together, so the interaction is diminishing and reveals a weak correlation between them. As temperature increases, there is waning in the values of surface tension suggesting the weak interactions among the constituent molecules. The decrease in relaxation time and Gibb's free energy indicates the reduction in the molecules degree of collaboration and relaxation. The results are discussed in terms of molecular interactions in the mixtures.

**Keywords**: Ultrasonic attenuation, relative strength, surface tension, relaxation time, Gibb's free energy.

## 1. INTRODUCTION

Ultrasonic method is one of the most reliable techniques to apprehend the physico-chemical idiosyncrasy of liquid mixtures. The molecular bonding and the nature of intermolecular interactions can be vividly deciphered by the thorough Study of thermoacoustic parameters [1,2]. Recent studies [3,4] enunciated that the analysis of ultrasonic velocity and their derived parameters with a variation of composition and temperature divulges vital information regarding interactions at an intermolecular level in the mixture**.** Jouyban *et al* [5] applied Jouyban–Acree model to calculate absolute viscosity against temperature and mixture concentrations of binary liquid mixtures. Acoustic speed (U), and viscosity (η) of pure tetrahydrofuran (THF), benzene, toluene, o-xylene, m-xylene, p-xylene, mesitylene as well as binary solutions with THF throughout the composition range and temperatures at 288.15, 298.15, 308.15 and 318.15 K were carried out. Further, deviations in the acoustic parameters were calculated. The THF-aromatic hydrocarbon synergy was compared and the dependence of the number and positioning of the methyl groups was reported [6]. At 308.15 K, Nayeem *et al*. [7] examined the density and acoustic velocity of binary liquid solutions of cyclohexanone with butanol isomers (n-butanol, sec-butanol, and tert-butanol) over the whole composition range. From the experimental data, thermoacoustic parameters and their excess parameters were calculated and the results were attributed to intermolecular

interactions. The conclusions generated from the excess parameters were supported by FTIR spectra. Furthermore, the experimental data were compared to theoretical ultrasonic velocities derived employing various theories.

Manoj Kumar Praharaj [8] derived thermodynamic parameters and their excess parameters from experimentally determined data of binary mixtures of cyclohexane and Nitro-benzene at 2 MHz, 4 MHz, 6 MHz, and 8 MHz at 308.15 K temperature and for different compositions. These calculations elucidate molecular interactions between the composite liquids.

Padmavathi *e tal* [9] calculated acoustical parameters of DMSO with butanol at different temperatures using experimental data obtained from ultrasonic velocity, viscosity and density. The outcome of the results is employed to investigate the nature and extent of interactions between binary molecules. Seema Agarwal [10,11] estimated the excess parameters of 6 liquid mixtures of methanol, ethanol, propanol, butanol, hexanol, and octanol with 1,4-dioxane to interpret the molecular interactions in the system. The acoustic velocity , density and viscosity of the liquid mixture are measured containing benzene with 0.01m of butyl/pentyl paraben in the isopropanol solution in the temperature298.15K - 323.15K. The acoustic parameters are calculated based on the measured data, and these results are discussed in terms of intermolecular interactions.

## 2, EXPERIMENTAL DETAILS

### 2.1 Materials

Butyl/Pentyl *p*-hydroxybenzoates [12-15] were supplied by Frinton Laboratories, Inc., USA whereas isopropanol (IPA) and benzene of AR grade was acquired from M/s Qualigen (India). They were double distilled before use. The specifications of the samples are mentioned in Table.1. The binary mixture of Butyl/Pentyl *p* hydroxy benzoates and isopropanol is employed as solute and non-polar solvent benzene in the preparation of solution.

**Table 1: Specification of the chemical samples**

| Liquid sample | Source | CAS No. | Mass fraction purity (supplier) | Analysis Method |
|---|---|---|---|---|
| Butylparaben | Frinton Laboratories, Inc., USA | 94-26-8 | ≥99% | GC[a] |
| Pentylparaben | Frinton Laboratories, Inc., USA | 6521-29-5 | ≥98% | GC[a] |
| Isopropanol | M/s. Qualigen (India) | 67-63-0 | ≥0.995 | GC[a] |
| Benzene | M/s. Qualigen (India) | 71-43-2 | ≥0.997 | GC[a] |

[a] Gas-Liquid chromatography stated by the supplier

### 2.2.Methods

Liquid mixtures were prepared gravimetrically with sufficient care taken to avoid evaporation for feitures. These compounds were autoclaved and kept on 0.4nm molecular sieves for 72 hours before usage for elimination of excess moisture. Butyl/pentyl paraben were dissolved at 0.01m in isopropanol (IPA), and every concentration of solute was diluted with solvent at various molalities.

Using Mettler Toledo AB135, Switzerland, a digital electronic balance, the mass measurements were performed with an uncertainty of ±0.00001g. The viscosities were measured using an Ostwald viscometer [16]. At each temperature, redistilled water was utilized to calibrate the viscometer. The measurement of viscosity has 0.0001mPa-s error. The flow time has been measured at equilibrium temperature. Electronic stop

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watch having 0.01s precision was used to take the flow measurements. For pure and mixtures mean value was taken in all calculations.

Densities [16] of the pure and mixtures were calculated using 10ml specific gravity bottles in a digital electronic balance. The measured density had an average error of ±0.001kg/m$^3$.

Acoustic velocity with 0.1m/s accuracy was measured using a single-crystal variable path 2MHz ultrasonic interferometer (Mittal Enterprises, New Delhi, India) [16,17] after calibration with standard liquids.

A constant temperature bath procured from Sakti Scientific Instruments Company, India and circulating water from the thermostat were used to maintain the temperature accuracy of 0.1K for all measurements.

### 2.3.Thermoacoustic parameters:

Ultrasonic attenuation ($\alpha$):

$$Ultrasonic\ attenuation, \frac{\alpha}{f^2} = \frac{8\pi^2\eta}{3\rho U^3} \quad mPas^4/Kg \quad .(1)$$

where $f$ indicates frequency of the ultrasonic wave, ρ indicates density in kg/m$^3$, U indicates speed of sound in m/s, η indicates dynamic viscosity in mPa-s

Relative Strength (r):

$$Relative\ Strength, r = 1 - \frac{U}{U_\infty} \ldots\ldots \quad (2)$$

where $U_\infty$ indicates limiting ultrasonic velocity which is equal to 1600m/s.

Surface Tension ($\sigma$):

$$Surface\ Tension, \sigma = 10^{-4} * U^{3/2} * \rho * T^{1/3} \quad N/m \quad \ldots(3)$$

where T indicates absolute temperature in Kelvin

Relaxation Time ($\tau$):

$$Relaxation\ Time, \tau = \frac{4\eta}{3\rho U^2} \quad s \quad \ldots\ldots(4)$$

Gibb's Free Energy ($\Delta G$):

$$Gibb's\ Free\ Energy, \Delta G = K_B T\ ln\left(\frac{K_B T \tau}{h}\right) \quad KJ \quad \ldots.(5)$$

where $K_B$ indicate Boltzmann constant and h is the Planck's constant.

## 3. RESULTS AND DISCUSSION

The liquid mixtures chosen for the present study are Benzene+Butyl paraben+Isopropanol; and Benzene+Pentyl paraben+Isopropanol. The speed of sound, density and viscosity for both systems at various temperatures are shown in tables 2 and 3 respectively.

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**Table.2: Densities, ρ, Ultrasonic Velocities, U, Viscosities, η of ternary system (Benzene +BP+ IPA) at various**

| [a]m(mol.kg$^{-1}$) | ρx10$^{-3}$(kg.m$^{-3}$) | | | | | |
|---|---|---|---|---|---|---|
| | **298.15K** | **303.15K** | **308.15K** | **313.15K** | **318.15K** | **323.15K** |
| 0.0000 | 792.56 | 787.20 | 782.36 | 777.24 | 772.94 | 767.94 |
| 0.1132 | 796.44 | 790.96 | 786.10 | 780.77 | 776.04 | 770.92 |
| 0.2232 | 801.89 | 796.20 | 791.44 | 786.60 | 781.68 | 775.68 |
| 0.3300 | 809.89 | 804.04 | 798.48 | 793.63 | 788.69 | 782.14 |
| 0.4338 | 817.23 | 811.32 | 805.95 | 800.31 | 795.19 | 788.99 |
| 0.5347 | 825.18 | 819.15 | 814.04 | 807.88 | 802.64 | 796.34 |
| 0.6329 | 834.97 | 829.29 | 823.57 | 818.28 | 812.14 | 805.54 |
| 0.7284 | 845.05 | 839.27 | 833.44 | 827.06 | 821.24 | 815.17 |
| 0.8213 | 853.98 | 847.64 | 842.26 | 835.84 | 829.38 | 823.86 |
| 0.9118 | 863.41 | 858.05 | 852.65 | 846.22 | 839.75 | 834.24 |
| 1.0000 | 873.79 | 868.45 | 863.08 | 857.69 | 852.28 | 846.85 |
| [a]m(mol.kg$^{-1}$) | U(m/s) | | | | | |
| 0.0000 | 1160.27 | 1142.38 | 1124.25 | 1105.82 | 1087.33 | 1068.79 |
| 0.1132 | 1160.67 | 1142.77 | 1124.92 | 1107.01 | 1089.01 | 1070.97 |
| 0.2232 | 1165.19 | 1147.07 | 1129.14 | 1108.99 | 1090.75 | 1072.55 |
| 0.3300 | 1172.85 | 1153.70 | 1134.74 | 1115.54 | 1096.30 | 1077.00 |
| 0.4338 | 1184.94 | 1166.21 | 1146.22 | 1125.24 | 1106.27 | 1085.30 |
| 0.5347 | 1198.51 | 1178.25 | 1157.73 | 1137.27 | 1116.89 | 1096.55 |
| 0.6329 | 1213.28 | 1193.27 | 1172.56 | 1149.70 | 1129.92 | 1107.23 |
| 0.7284 | 1229.13 | 1207.67 | 1186.59 | 1165.38 | 1145.32 | 1122.37 |
| 0.8213 | 1246.13 | 1224.48 | 1203.18 | 1181.80 | 1160.54 | 1138.39 |
| 0.9118 | 1268.69 | 1245.91 | 1221.47 | 1201.96 | 1178.58 | 1158.29 |
| 1.0000 | 1299.22 | 1276.30 | 1252.16 | 1230.17 | 1207.38 | 1182.78 |
| [a]m(mol.kg$^{-1}$) | η (mPa-s) | | | | | |
| 0.0000 | 1.8916 | 1.6985 | 1.5558 | 1.3754 | 1.2256 | 1.1100 |
| 0.1132 | 1.7060 | 1.5335 | 1.3933 | 1.2255 | 1.0968 | 0.9871 |
| 0.2232 | 1.3799 | 1.2676 | 1.1822 | 1.0443 | 0.9299 | 0.8619 |
| 0.3300 | 1.2337 | 1.1265 | 0.9964 | 0.8944 | 0.8176 | 0.7556 |
| 0.4338 | 1.1106 | 1.0102 | 0.8638 | 0.7665 | 0.6883 | 0.6231 |
| 0.5347 | 0.9747 | 0.8756 | 0.7814 | 0.7035 | 0.6404 | 0.5820 |
| 0.6329 | 0.8743 | 0.7456 | 0.6976 | 0.6138 | 0.5667 | 0.5438 |
| 0.7284 | 0.7788 | 0.6748 | 0.6323 | 0.5732 | 0.5193 | 0.4914 |
| 0.8213 | 0.6907 | 0.6275 | 0.5734 | 0.5268 | 0.4907 | 0.4632 |
| 0.9118 | 0.6442 | 0.6041 | 0.5609 | 0.5153 | 0.4718 | 0.4539 |
| 1.0000 | 0.6030 | 0.5620 | 0.5370 | 0.5061 | 0.4525 | 0.4361 |

*[a]m states the molalities of benzene in the isopropanol solution of butyl paraben. Standard uncertainties u is* $u_r(m)$ = 0.0002, *u(T)= 0.01%, u(η)* =0.0001mPa-s

Based on the experimentally determined data of U, ρ, η, in the two systems, various thermoacoustic parameters such as ultrasonic attenuation ($\alpha/f^2$), relative strength (r), surface tension (σ), relaxation time (τ) and Gibb's free energy (ΔG) are calculated in the temperature range 298.15K - 323.15K.

**Table.3: Densities, ρ, Ultrasonic Velocities, U, Viscosities, η of ternary system (Benzene +PP+ IPA) at various temperatures and atmospheric pressure, p = 0.1 MPa.**

| [a]m(mol.kg$^{-1}$) | ρx10$^{-3}$(kg.m$^{-3}$) | | | | | |
|---|---|---|---|---|---|---|
| | 298.15K | 303.15K | 308.15K | 313.15K | 318.15K | 323.15K |
| 0.0000 | 789.49 | 785.18 | 780.80 | 776.33 | 771.78 | 767.13 |
| 0.1123 | 796.30 | 791.53 | 787.29 | 782.26 | 777.96 | 773.15 |

| | | | | | | |
|---|---|---|---|---|---|---|
| 0.2217 | 803.68 | 799.02 | 794.48 | 789.56 | 786.56 | 781.58 |
| 0.3281 | 810.50 | 805.72 | 800.87 | 795.94 | 791.93 | 786.83 |
| 0.4317 | 818.00 | 812.91 | 808.15 | 802.83 | 798.03 | 792.85 |
| 0.5326 | 826.20 | 821.15 | 816.03 | 810.85 | 806.61 | 801.30 |
| 0.6309 | 834.40 | 829.72 | 824.50 | 819.41 | 815.37 | 809.96 |
| 0.7267 | 843.03 | 838.06 | 832.77 | 827.42 | 823.02 | 817.56 |
| 0.8200 | 851.66 | 846.34 | 840.98 | 835.58 | 830.13 | 824.63 |
| 0.9111 | 861.62 | 856.24 | 850.83 | 845.39 | 839.91 | 834.39 |
| 1.0000 | 873.79 | 868.45 | 863.08 | 857.69 | 852.28 | 846.85 |
| [a]m(mol.kg$^{-1}$) | U(m/s) | | | | | |
| 0.0000 | 1151.59 | 1135 | 1116.56 | 1098.78 | 1080.91 | 1062.95 |
| 0.1123 | 1161.53 | 1144.51 | 1125.18 | 1105.8 | 1088.35 | 1069.86 |
| 0.2217 | 1171.28 | 1153.25 | 1132.13 | 1112.98 | 1094.83 | 1076.61 |
| 0.3281 | 1179.75 | 1162.2 | 1142.01 | 1121.39 | 1103.75 | 1084.14 |
| 0.4317 | 1190.02 | 1170.07 | 1150.34 | 1130.43 | 1110.53 | 1090.62 |
| 0.5326 | 1200.33 | 1181.78 | 1161.46 | 1140.97 | 1120.59 | 1100.2 |
| 0.6309 | 1212.59 | 1194.79 | 1173.85 | 1152.97 | 1133.26 | 1111.62 |
| 0.7267 | 1227.06 | 1208.04 | 1186.87 | 1165.76 | 1144.84 | 1124.01 |
| 0.8200 | 1242.93 | 1223.61 | 1201.13 | 1179.78 | 1158.6 | 1138.53 |
| 0.9111 | 1266.77 | 1243.53 | 1221.73 | 1200.06 | 1176.56 | 1157.16 |
| 1.0000 | 1299.22 | 1276.3 | 1252.16 | 1230.17 | 1207.38 | 1182.78 |
| [a]m(mol.kg$^{-1}$) | η (mPa-s) | | | | | |
| 0.0000 | 1.6911 | 1.5815 | 1.3843 | 1.2188 | 1.0909 | 0.9825 |
| 0.1123 | 1.2129 | 1.1092 | 1.0219 | 0.9199 | 0.8069 | 0.7474 |
| 0.2217 | 1.0727 | 0.9650 | 0.8886 | 0.8032 | 0.7416 | 0.6926 |
| 0.3281 | 0.8927 | 0.8082 | 0.7615 | 0.7034 | 0.6627 | 0.6182 |
| 0.4317 | 0.8459 | 0.7704 | 0.7416 | 0.6700 | 0.5968 | 0.5673 |
| 0.5326 | 0.7645 | 0.6871 | 0.6376 | 0.5785 | 0.5179 | 0.4981 |
| 0.6309 | 0.7267 | 0.6387 | 0.6059 | 0.5505 | 0.4996 | 0.4900 |
| 0.7267 | 0.6818 | 0.6206 | 0.5635 | 0.5220 | 0.4872 | 0.4600 |
| 0.8200 | 0.6353 | 0.5960 | 0.5540 | 0.5094 | 0.4667 | 0.4492 |
| 0.9111 | 0.6283 | 0.5729 | 0.5423 | 0.5071 | 0.4599 | 0.4448 |
| 1.0000 | 0.6030 | 0.5621 | 0.5371 | 0.5061 | 0.4525 | 0.4360 |

*[a]m states the molalities of Benzene in the Isopropanol solution of Pentyl ParabenStandard uncertainties u is* $u_r(m)$ = 0.0002, $u(T)$= 0.01%, $u(\eta)$ =0.0001mPa-s

In both systems, the ultrasonic velocity of the liquid mixture increased with the increase of benzene concentration. Similarly, the ultrasonic velocity decreases with an increase in temperature. In both the systems, the density of the chosen system increased with the increase of benzene concentration and the density of the mixture decreased with increased temperature. Further, the viscosity of the liquid mixtures decreases with increase in the composition as well as temperatures. Benzene is a non-polar liquid. As the temperature upsurges, molecular dissociation occurs, as indicated by a decline in the acoustic velocities. Benzene molecule has two resonating structures are shown in Fig.1.

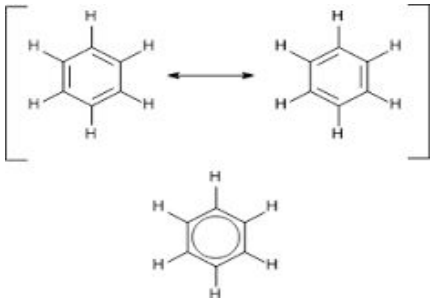

**Fig.1.Resonance Structures of Benzene**

Fig.2 and 3 contain the plot of experimental ultrasonic attenuation($\alpha/f^2$) versus molar concentration at different temperatures.

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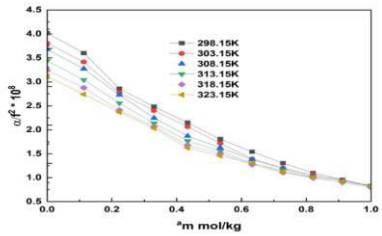


**Fig.2. Variation of Ultrasonic attenuation (dB/m) of Benzene + 0.01m of Butyl Paraben in the Isopropanol solution**

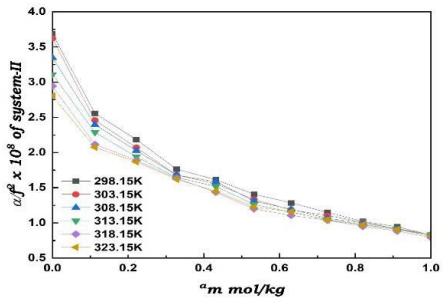


**Fig.3. Variation of Ultrasonic attenuation (dB/m) of Benzene + 0.01m of Pentyl Paraben in the Isopropanol solution**

It is observed that ultrasonic attenuation ($\alpha/f^2$) rapidly decreases with an increase in the molar concentration of benzene in Isopropanol solution of parabens indicating the least stability. Benzene molecule has two resonating structures (Fig.1), which decreases the relaxation time. Reduction in relaxation time reduces the ultrasonic attenuation in this binary liquid system. The nonlinear variation of ultrasonic attenuation in each curve with molar concentration strongly supports the presence of weak intermolecular interaction through hydrogen bonding in the component molecules of this binary liquid system.

From Figs. 4 and 5, it is evident that there is a decrease in the relative strength of the mixture with the increase in concentration of benzene in both the systems. The nonlinear variation indicates that the molecules are not very proximal together, and so the interaction is diminishing, indicating that there is a weak correlation between them.

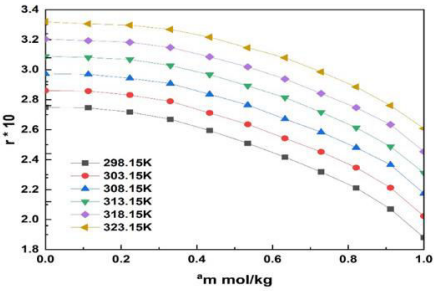


**Fig.4. Variation of Relative strength of Benzene + 0.01m of Butyl Paraben in the Isopropanol solution**

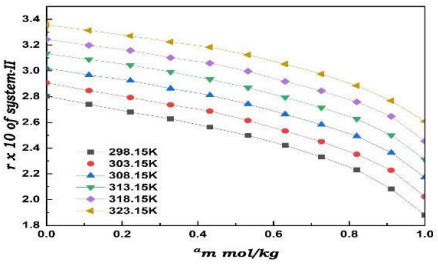


**Fig.5. Variation of Relative strength of Benzene + 0.01m of Pentyl Paraben in the Isopropanol solution**

By observing the Figs.6 and 7, there is a gradual increase in the surface tension values with the increase in the concentration of benzene. But as there is an upsurge in temperature there is waning in the values of surface tension suggesting the weak interactions among the constituent molecules.

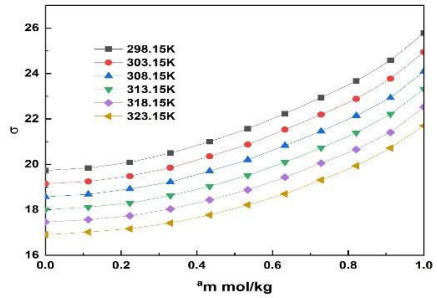


**Fig.6. Variation of Surface tension (N/m) of Benzene + 0.01m of ButylParaben in the Isopropanol solution**

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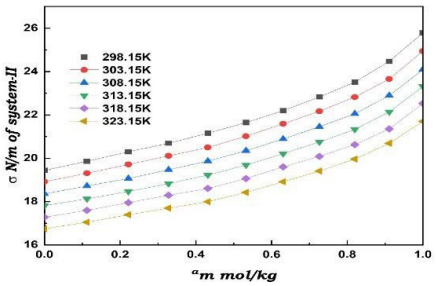


**Fig.7. Variation of Surface tension (N/m) of Benzene + 0.01m of Pentyl Paraben in the Isopropanol solution**

Figs.8 and 9 embellish the variation of relaxation time of systems I and II at various temperatures. The relaxation time decreases upon the increase of benzene concentration. It further reduces with temperature surge. The dispersion of ultrasonic waves in system comprises complete data about the characteristic time of relaxation process that triggers the dispersion. Decrease in relaxation time indicates that degree of collaboration for relaxation of the molecules declines which wanes the bulk of cluster when solute is added to solvent.

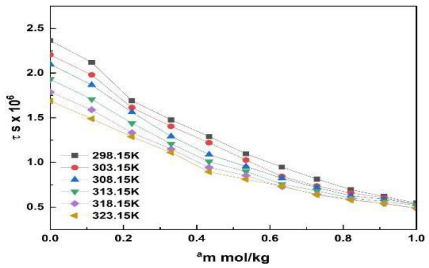


**Fig.8. Variation of Relaxation time(s) of Benzene + 0.01m of Butyl Paraben in the Isopropanol solution**

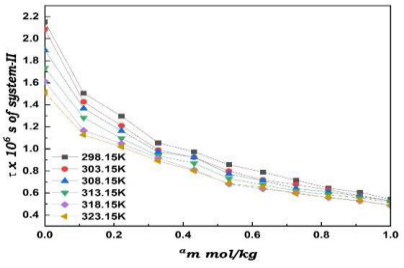


**Fig.9. Variation of Relaxation time (s) of Benzene + 0.01m of Pentyl Paraben in the Isopropanol solution**

The dispersion of atoms or molecules within the substance causes viscosity. The Gibbs free energy may be said as a thermodynamic potential. It measures the efficacy or process-initiating work that can be obtained from a thermodynamic system under constant pressure and temperature. The elastic characteristics of the medium and the induced particle vibrations in the media cause the shift in wavelength of ultrasonic waves. When a system changes from a well defined initial state to a well-defined final state, the Gibbs free energy ΔG equals the work exchanged by the system with its surroundings, minus the work of the pressure forces, during are versible transformation of the system from the same initial state to the same final state. The addition of interacting molecules disrupts the molecular clustering of the other. From Figs. 10 and 11, decreasing of Gibb's free energy indicates rearrangement of molecules will take place longer time. This is due to the existence of weak molecular association in the liquid mixture among different moieties.

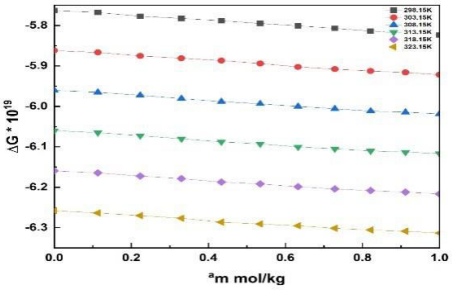


**Fig.10. Variation of Gibb's free energy (KJ) of Benzene + 0.01m of Butyl Paraben in the Isopropanol solution**

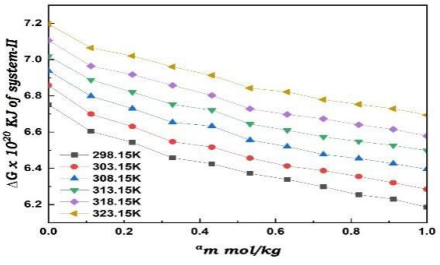


**Fig.11. Variation of Gibb's free energy (KJ) of Benzene + 0.01m of Pentyl Paraben in the Isopropanol solution**

## 4. CONCLUSIONS

Temperature dependence of acoustic velocity, density and viscosity of the liquid mixture containing benzene with 0.01m of Butyl/Pentyl paraben in the isopropanol solution at different concentrations were carried out. The calculated acoustic parameters like ultrasonic attenuation ($\alpha/f^2$), relative strength (r), Surface tension ($\sigma$), Relaxation time ($\tau$) and Gibb's free energy ($\Delta G$) suggests that intermolecular interactions exist in the liquid mixtures.

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**CONFLICTS OF INTEREST**

"The authors declare no conflict of interest."

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