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# ULTRASONIC INVESTIGATION OF AQUEOUS DEXTRAN AT DIFFERENT TEMPERATURES AND FREQUENCIES

**Subhraraj Panda*[1] and Achyuta Prasad Mahapatra[2]**

[1]Department of Physics, Centurion University of Technology and Management, Jatni Campus Bhubaneswar-752050, Odisha, India.
[2]*KBRC Degree College* Cuttack, Odisha, India.

***Corresponding Author: Subhraraj Panda**
Department of Physics, Centurion University of Technology and Management, Jatni Campus Bhubaneswar-752050, Odisha, India.

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## ABSTRACT

The propagation of Ultrasonic waves in aqueous solution of dextran polymers depends on its viscoelastic behaviour and density which significantly affected with change in temperature and frequency. Therefore the application of low intensity ultrasound, acting as a high frequency dynamic mechanical deformation applied to a polymer, can monitor the changes in acoustic and thermodynamic properties associated with it. In this paper, values of density, viscosity and ultrasonic velocity and related acoustic parameters such as adiabatic compressibility (β), acoustic impedance (Z), relaxation time (τ), intermolecular free length ($L_f$) and Gibb's free energy (ΔG), have been determined and the solute-solvent interactions for the aqueous solution of dextran (1%) have been studied at five different temperatures i.e. 30 $^0$C, 35 $^0$C,40 $^0$C,45 $^0$C, 50 $^0$C and four different frequencies i.e. 1MHz, 5MHz, 9MHz, 12MHz. The variation of thermo acoustic parameter with different temperature and frequency leads to the analysis of molecular motion and various types of inter-molecular interaction and their strength of the constituent between solute (dextran) and solvent (distilled water). The results have been interpreted in the light of structural rearrangement occurs in the aqueous dextran solution.

**KEYWORDS:** Aqueous dextran solution, ultrasonic velocity, adiabatic compressibility (β), acoustic impedance (Z), relaxation time (τ), intermolecular free length ($L_f$), Gibb's free energy(ΔG).

## INTRODUCTION

A lot of pharmaceutical and chemical industries use different kinds of polymer solution and liquid mixtures in their processes,[1,2] for this reason it is necessary to pay attention to the thermodynamic studies and modelling test and development to optimal industrial designs and simulations. The interactions between different molecules are responsible for non-ideal mixing properties of solvents, and these interactions influence the physio-chemical properties. The practical importance of solution rather than single component liquid systems has gained much importance during the last two decades in assessing the nature of molecular interactions and investigating the physio-chemical behaviour of such systems.[3]

In our present work we have calculated thermo acoustic parameters like adiabatic compressibility, intermolecular free length, relaxation time, acoustic impedance, and Gibb's free energy of a novel polymer dextran of molecular weight 70,000 at five different temperature i.e. 30 $^0$C, 35 $^0$C, 40 $^0$C, 45 $^0$C, 50 $^0$C and four different frequencies i.e. 1MHz, 5MHz, 9MHz, 12MHz.in solvent distilled water at concentration of 1% dextran.

We have chosen a polymer dextran as a solute with water as a solvent. This is the only polymer which is water soluble. Dextran and their derivatives find wide applications in various industries particularly in pharmaceutical sector[4,5] the fast increasing of these poly-glucosans for medical[6,7,8] industrial and research[9] purposes motivated to carry out investigation of thermo acoustic parameter of dextran by ultrasonic technique.

## MATERIALS AND METHODS

### Experimental Details

Freshly prepared distilled water has been used as solvent for preparing dextran solution. Dextran of molecular weight 70,000 used as solute, is of analytical reagent (AR) grade, manufactured by HI Media Laboratories Private Limited, India.

### (i) Density Measurement

The densities of the solution are measured using a 25ml specific gravity bottle. The specific gravity bottle with

the 1% aqueous solution of dextran is immersed in a temperature controlled water bath and density is measured at five different temperatures (30 $^0$C, 35 $^0$C, 40 $^0$C, 45 $^0$C & 50 $^0$C) and calculated using standard equation.[10]

**(ii) Viscosity measurement**

The viscosity of solution is measured using an Oswald's viscometer calibrated with distilled water. The Oswald's viscometer with the polymer solution is immersed in a temperature controlled water bath. The time of flow was measured using a racer stop watch with an accuracy of 0.01 second at above mentioned temperatures and calculated using standard equation.[10]

**(iii) Velocity Measurement**

The ultrasonic interferometer used for measuring velocity have been calibrated by taking some standard liquid. The velocity of aqueous solution of Dextran (1%) have been measured at four different frequencies i.e. 1MHz, 5MHz, 9MHz, 12MHz. using multi-frequency ultrasonic interferometer (Model M-84) supplied by M/s Mittal Enterprises, New Delhi. The measuring cell of interferometer is a specially designed double walled vessel with provision for temperature constancy. An electronically operated digital constant temperature bath (Model SSI-03spl) supplied by M/s Mittal Enterprises, New Delhi, operating in the temperature range -10 $^o$C to 85 $^o$C with an accuracy of ± 0.1$^o$C has been used to circulate water through the outer jacket of the double walled measuring cell containing the experimental liquid. The velocity of aqueous solution of Dextran (1%) have been measured at above mentioned temperatures**.**

**Theoretical Aspect**

**(i) Adiabatic Compressibility (β)**

The adiabatic compressibility is the fractional decrease of volume per unit increase of pressure, when no heat flows in or out. It is calculated from the speed of sound and the density (ρ) of the medium by using the equation of Newton Laplace as:

$$\beta = 1/U^2.\rho \text{ ---------- (1)}$$

**(ii) Acoustic impedance (*Z*)**

The specific acoustic impedance is given by,

$$Z = U.\rho \text{ ---------- (2)}$$

Where 'U' and 'ρ' are velocity and density of Dextran solution.

**(iii) Relaxation time (τ)**

Relaxation time is the time taken for the excitation energy to appear as translational energy and it depends on temperature. The relaxation time can be calculated from the relation,

$$\tau = \frac{4}{3}\frac{\eta}{\rho U^2} \text{ ---------- (3)}$$

**(iv) Intermolecular free length ($L_f$)**

The intermolecular free length is the distance between the surfaces of the neighbouring molecules. It is calculated by using the relation:

$$L_f = \frac{K_T}{U\rho^{1/2}} \text{ ---------- (4)}$$

Where $K_T$ is the temperature dependent constant.

**(v) Gibb's free energy (ΔG)**

The Gibb's free energy is calculated by using the relation

$$\Delta G = kT \ln (kT\tau/h) \text{ ---------- (5)}$$

Where, τ is the viscous relaxation time, '*T*' is the absolute temperature; '*k*' is the Boltzmann's constant and '*h*' is the Planck's constant.

## RESULTS AND DISCUSSION

The density and viscosity of aqueous dextran at temperature 303K, 308K, 313K, 318K and 323K are represented in Table-1.

**Table 1: The experimental values of density (ρ) and viscosity (η) at 303K, 308K, 313K and 318K of aqueous solution of Dextran.**

| T in kelvin | Density(ρ) Kg.m$^{-3}$ | Viscosity(η) 10$^{-3}$ N.s.m$^{-2}$ |
|---|---|---|
| 303 | 1000.827 | 0.991 |
| 308 | 999.212 | 0.890 |
| 313 | 996.998 | 0.809 |
| 318 | 994.597 | 0.748 |
| 323 | 991.621 | 0.713 |

**Table 2: Values of ultrasonic velocity (U) and adiabatic compressibility (β) at 303K, 308K, 313K, 318K and 323K of aqueous solution of Dextran (1%) at different frequencies.**

| T in kelvin | Velocity(U) m.s$^{-2}$ | | | | Adiabatic compressibility(β) (10$^{10}$ m$^2$ N$^{-1}$) | | | |
|---|---|---|---|---|---|---|---|---|
| | 1MHz | 5MHz | 9MHz | 12 MHz | 1MHz | 5MHz | 9MHz | 12 MHz |
| 303 | 1520.00 | 1516.00 | 1514.00 | 1519.00 | 1.521 | 1.517 | 1.515 | 1.520 |
| 308 | 1528.47 | 1523.00 | 1521.00 | 1526.00 | 1.527 | 1.522 | 1.520 | 1.525 |
| 313 | 1538.33 | 1532.75 | 1528.00 | 1535.00 | 1.534 | 1.528 | 1.523 | 1.530 |
| 318 | 1545.27 | 1540.00 | 1538.00 | 1542.00 | 1.537 | 1.532 | 1.530 | 1.534 |
| 323 | 1552.00 | 1548.00 | 1543.50 | 1550.00 | 1.539 | 1.535 | 1.531 | 1.537 |

**Table 3: Values of Acoustic impedance (Z) and Relaxation time (τ) 303K, 308K, 313K, 318K and 323K of aqueous solution of Dextran (1%) at different frequencies.**

| T in kelvin | Acoustic impedance (Z) ( $\times 10^4$ kg·m$^2$·s$^{-1}$ | | | | Relaxation time(τ)( x $10^{10}$ )s | | | |
|---|---|---|---|---|---|---|---|---|
| | 1MHz | 5MHz | 9MHz | 12 MHz | 1MHz | 5MHz | 9MHz | 12 MHz |
| 303 | 1.521 | 1.517 | 1.515 | 1.520 | 5.712 | 5.742 | 5.757 | 5.720 |
| 308 | 1.527 | 1.522 | 1.520 | 1.525 | 5.085 | 5.122 | 5.135 | 5.101 |
| 313 | 1.534 | 1.528 | 1.523 | 1.530 | 4.570 | 4.604 | 4.632 | 4.590 |
| 318 | 1.537 | 1.532 | 1.530 | 1.534 | 4.200 | 4.229 | 4.240 | 4.218 |
| 323 | 1.539 | 1.535 | 1.531 | 1.537 | 3.978 | 3.999 | 4.022 | 3.988 |

**Table 4: Values of Intermolecular free length ($L_f$) and ultrasonic Gibb's free energy (ΔG) 303K, 308K, 313K, 318K and 323K of aqueous solution of Dextran (1%) at different frequencies.**

| T in kelvin | Intermolecular free length($L_f$)($10^{-10}$ m) | | | | Gibb's free energy( ΔG)(x$10^{-20}$ K.J.mol$^{-1}$) | | | |
|---|---|---|---|---|---|---|---|---|
| | 1MHz | 5MHz | 9MHz | 12 MHz | 1MHz | 5MHz | 9MHz | 12 MHz |
| 303 | 4.126 | 4.137 | 4.142 | 4.129 | 233.04 | 234.00 | 234.48 | 233.28 |
| 308 | 4.142 | 4.157 | 4.163 | 4.149 | 218.43 | 219.76 | 220.24 | 219.03 |
| 313 | 4.156 | 4.171 | 4.184 | 4.165 | 204.97 | 206.33 | 207.50 | 205.78 |
| 318 | 4.178 | 4.192 | 4.197 | 4.186 | 195.15 | 196.45 | 196.95 | 195.96 |
| 323 | 4.201 | 4.212 | 4.224 | 4.206 | 190.73 | 191.73 | 192.86 | 191.23 |

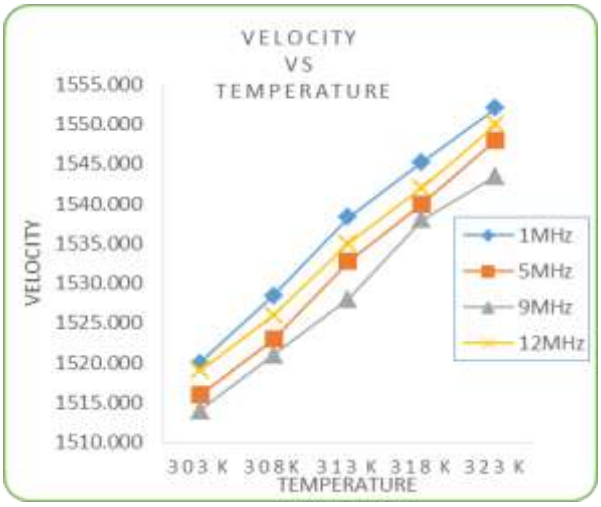


**Fig. 1: Variation of ultrasonic velocity with temperature.**

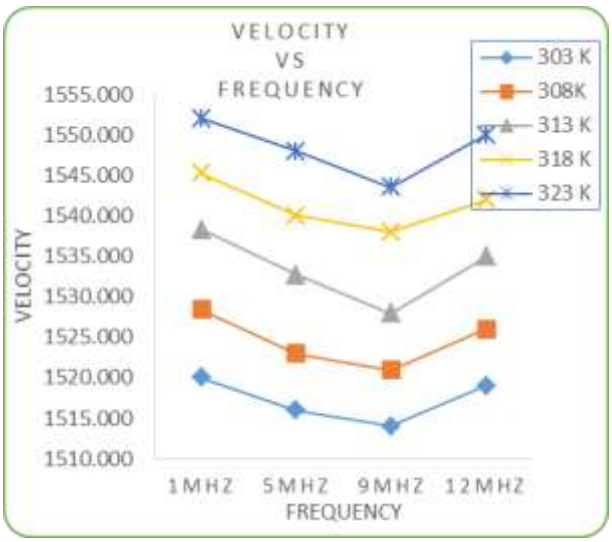


**Fig. 2: Variation of ultrasonic velocity with frequency.**

Measurements of density and viscosity in liquid solutions gives some reliable information in the study of molecular interaction. The viscosity and density gives the strength of molecular interaction between the interacting molecules. Dispersion, induction and dipolar forces which are operated by the density and viscosity, these forces are responsible for the existence of specific interactions in the interacting molecules leading to the decrease with increase in temperature indicates thermal energy of the system. This causes an increase in volume and hence decrease in density and viscosity as in table-1.

Figure 1 contains the plot of ultrasonic velocity versus molar concentration. It is observed that ultrasonic velocity increases with increase in temperature indicating association in the constituent. The electrons surrounding the nucleus in the atom of each dextran molecule are symmetrical, distributed round the nucleus. According to London these electrons are in continuous and rapid motion with respect to the nucleus due to increase in temperature and hence it is possible that at any instant, the electron density round the nucleus may be concentrated in the other region of the same molecule. This results in an unsymmetrical distribution of the electron density round the nucleus and hence temporary dipole are induced the permanent dipoles are present in the water molecules. Thus there is dipole-induced dipole interaction between constituents.[11,12]

The stronger dipole-induced dipole interaction between the unlike molecules are responsible for the association. Interstitial accommodation, induction and London dispersion forces in dextran molecules may leads association in the constituents.

The relaxation time as well as the intermolecular free lengths are properties of liquid solutions which mainly affect the ultrasonic velocity. The velocity decreases

with the increase of frequency up to 9MHz but at higher frequency (12MHz) the result is reversed due to highly vibration of the molecules at their equilibrium position. . This is confirmed by the fact that both relaxation time increase slowly figure-10 and free length with the increase of frequency figure-8. Since the association of the interacting molecules varies with the frequency of the ultrasonic wave.[13]

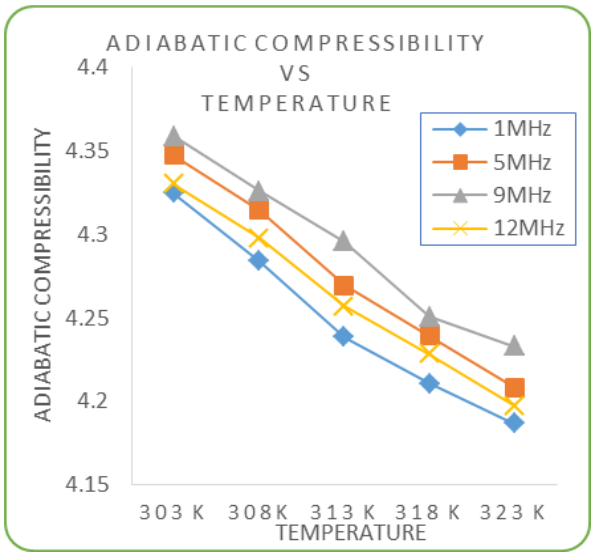


**Fig. 3: Variation of adiabatic compressibility with temperature.**

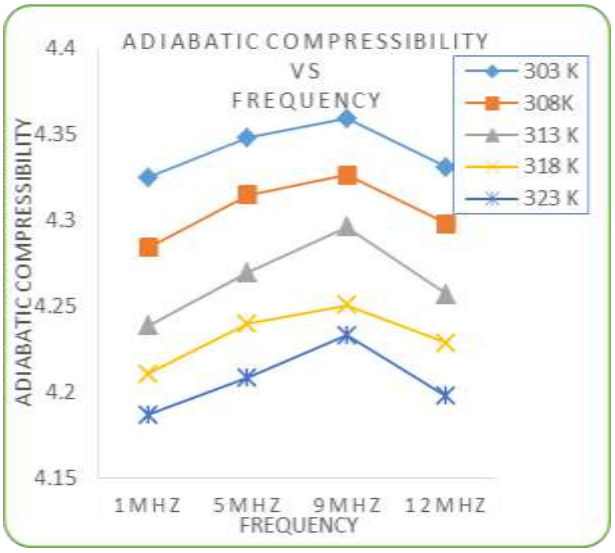


**Fig. 4: Variation of adiabatic compressibility with frequency.**

Adiabatic compressibility decreases with increase in temperature figure3 which indicates strong interaction between solute and solvent molecules. It is a well-known fact that when a solute dissolves in a solvent some of the solvent molecules are attached to the ions (produced from the solute) because of ion-solvent interactions. Since the solvent molecules are oriented in the ionic field (i.e. electrostatic fields of ions) the solvent molecules are more compactly packed in the primary salvation shell as compared to the packing in the absence of the ions, this is the reason why the solvent is compressed by the introduction of ions. Thus, the electro static field of the ions causes compression of the medium giving rise to a phenomenon called electrostriction. Since the solvent molecules are compressed they do not respond any further application of pressure so the solution becomes harder to compress i.e. the compressibility decreases with temperature.[14]

The adiabatic compressibility increases (Figure 4), suggests minimum interaction between dextran and water molecules. Adiabatic compressibility is given by the relation, $\beta = 1/U^2.\rho$. Which shows that as frequency increases, velocity decreases at a particular density and hence adiabatic compressibility increases and at higher frequency the interaction is reversed. This also confirms the fact that free length increases (figure-8), with the increase in frequency.[15]

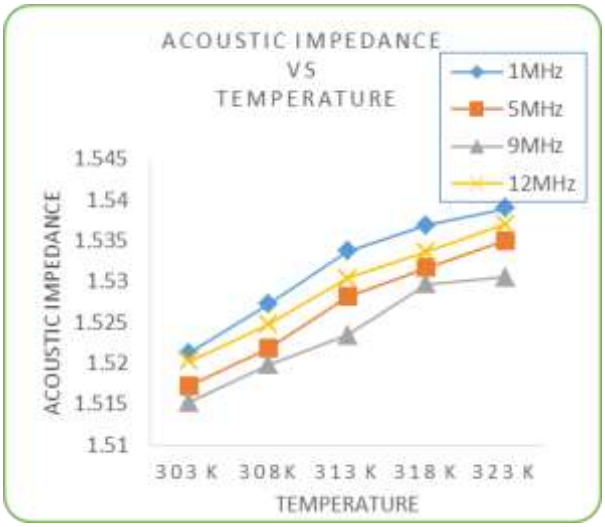


**Fig. 5: Variation of acoustic impedance with temperature.**

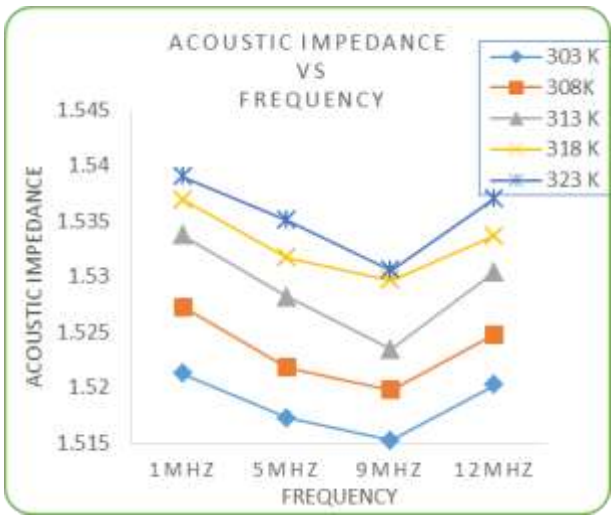


**Fig. 6: Variation of acoustic impedance with frequency.**

Acoustic impedance is the ratio of the effective sound pressure at a point to the effective particle velocity at that

point. The pressure is measured by the totality of the forces of dispersion, repulsion, ionic and dipolar.[16] In the present Investigation, acoustic impedance increases with increase in temperature [fig-5]. This indicates the possibility of strong molecular interaction between the components of the mixture.

Acoustic impedance decreases with the increase in frequency. This suggests the possibility of weak interaction between solute and solvent molecules. Acoustic impedance is also given by the product of the ultrasonic velocity and density Z = Uρ and is used for assessing the absorption of sound in a medium.

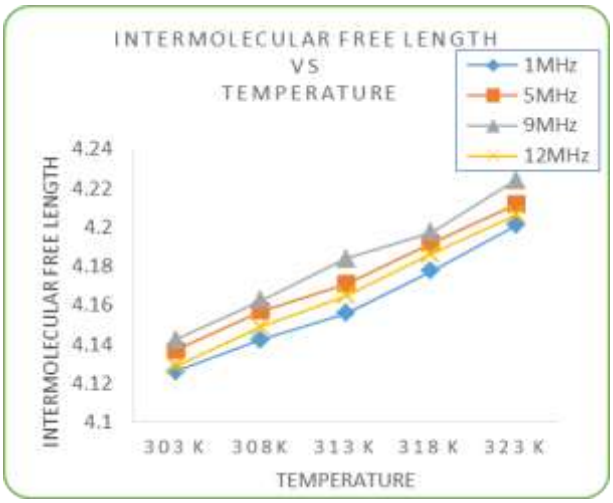


**Fig. 7: Variation of intermolecular free length with temperature.**

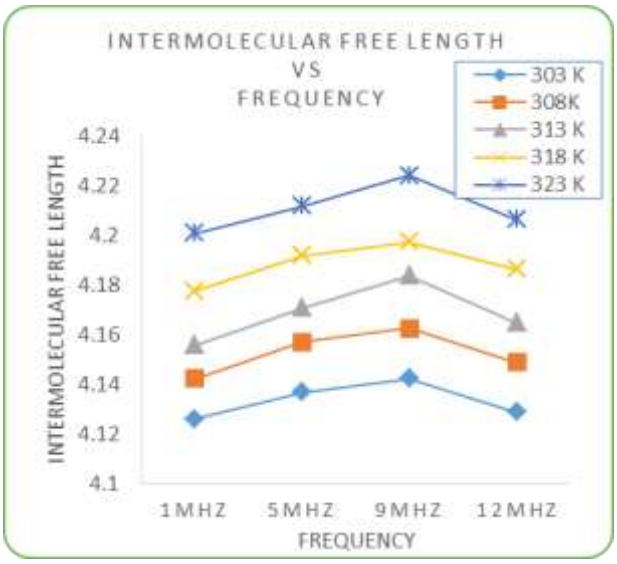


**Fig. 8: Variation of intermolecular free length with frequency.**

Inter molecular free length increases(Fig.7) with increase in temperature [] as the temperature increases it leads to the less order structured and more spacing between the molecules due to increase in thermal energy of the system which increases in volume expansion and hence increase in inter molecular free length.

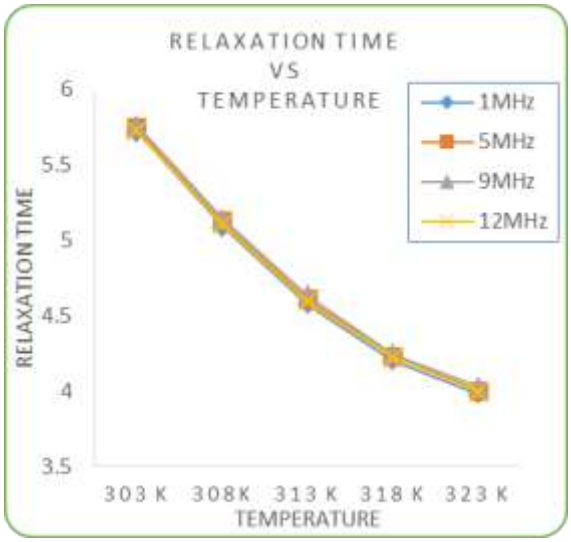


**Fig. 9: Variation of relaxation time with temperature.**

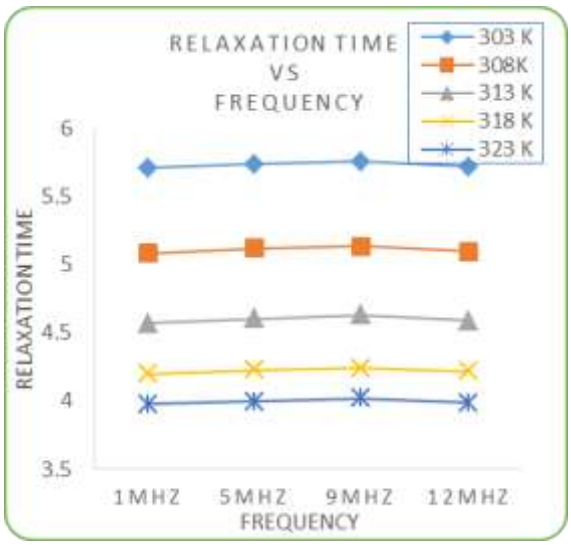


**Fig. 10: Variation of relaxation time with frequency.**

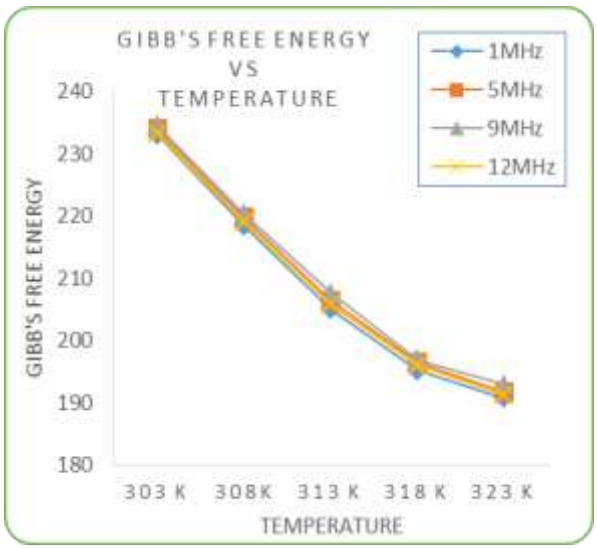


**Fig. 11: Variation of Gibb's free energy with temperature.**

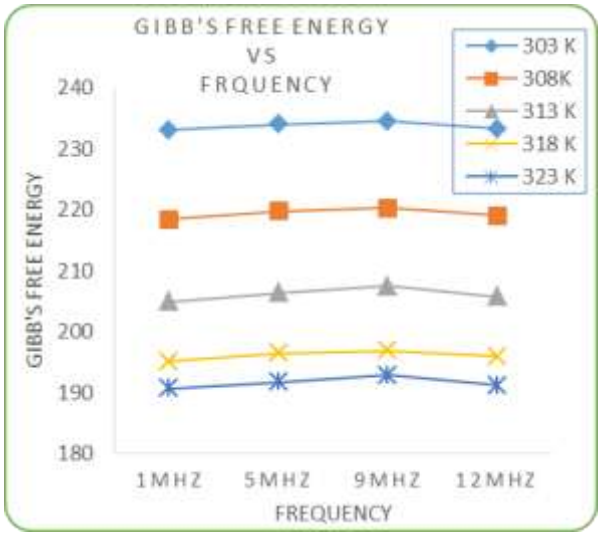


**Fig. 12: Variation of Gibb's free energy with frequency.**

The viscous relaxation time and Gibbs free energy both decreases as temperature increases as shown Fig.9 & fig.11 respectively. With increase in temperature excitation energy increases and hence relaxation time decreases. Further as the kinetic energy of the molecule increases, it takes long time for rearrangement of molecule and this suggests a decrease in Gibb's free energy.[17]

The viscous relaxation time and Gibbs free energy increases with the increase in frequency gradually. An increasing value of viscous relaxation time and Gibbs free energy suggests that the closer approach of unlike molecules is due to hydrogen bonding. The increase in Gibbs free energy suggests shorter time for the rearrangement of molecules in the solution slowly as figure-12. When frequency increases, the energy imparted to the molecules obviously expedites the rearrangement procedure slowly.[18]

## CONCLUSION

From the experimental measured parameters i.e. density, viscosity and ultrasonic velocity of the aqueous solutions of dextran at different temperatures, thermo-acoustic parameters like, adiabatic compressibility (β), acoustic impedance (Z), relaxation time (τ), intermolecular free length (Lf) and Gibb's free energy (ΔG), have been calculated at different temperature and frequency. The results show that the specific solute-solvent interactions play an important role for explaining acoustic parameters. However, any deviation from the usual behaviour is probably due to characteristic structural changes in the respective system.

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