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Acoustical Studies of Molecular Interactions in the Solution of Anti-Malarial Drug

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Abstract In the present study ultrasonic velocity (U), density (ρ) and viscosity (η) have been measured at frequency 1 MHz in the binary mixtures of chloroquine phosphate with water in the concentration range (0.1 to 0.0125 %) at 303 K,308 K,313 K using multi frequency ultrasonic interferometer. The measured value of density, ultrasonic velocity, and viscosity have been used to estimate the acoustical parameters namely adiabatic compressibility (βa), relaxation time (τ), acoustic impedance (z), free length (Lf), free volume (Vf) and internal pressure (Πi), Wada's constant (W) to investigate the nature and strength of molecular interaction in the binary mixture of chloroquine phosphate hydrochloride with water. The obtained result supports the complex formation, molecular association by intermolecular hydrogen bonding in the binary liquid mixtures.

Keywords: *chloroquine phosphate, free volume, acoustical parameters, ultrasonic velocity*

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1. Introduction

Ultrasound refers to sound waves of such a high frequency that it cannot be heard. High resolution ultrasound imaging has been used for determination of melanoma invasion depth in vivo for preoperative staging purposes [1,2]. Now a day's Ultrasonic technology is employed in a wide range of applications in medicine, biology, industry, material science, agriculture, oceanography, sonochemistry research etc. due to its non-destructive nature [3-10]. These waves have also been used to extract and release intracellular enzymes such as invertase, to promote enzyme release, enhance productivity in biological processes [11] etc. In field of agriculture, ultrasound waves have been utilized extensively in chemical additives (fertilizers and plant protection preparations) for improving the production yield of food produced. In materials chemistry, ultrasound waves have been useful in the preparation of biomaterials, protein microspheres, in the modification of polymers and polymer surfaces etc. [12-17]. Much work has been done in solutions of polymers [18-23], amino acids [24,25] and other electrolytes [26-33]. However, little work has been done for solutions of solid organic compounds [34-38].

Ultrasonic offers the most exciting and fascinating field of scientific research among the researchers since the ultrasonic and other related thermo acoustic parameters provide useful information regarding the structure of molecules, molecular order, molecular packing, inter and intra -molecular interactions [39,40] etc. Ultrasonic study of liquid - liquid mixture has gained much importance during the last two decades in assessing the nature of molecular interaction and investigating the physiochemical behavior of this system [41,42]. The review of literature reveals that lot of work has been done to investigate ultrasonic measurement of pure liquid and liquid mixture at different environment, but less effort has been made to investigate ultrasonic studies in binary mixture i.e. chloroquine phosphate with water. Thus, in the present work, acoustical studies of have been studied in water at different temperatures over a wide range of chloroquine phosphate concentrations. The thermodynamic parameters: adiabatic compressibility, acoustic impedance, relaxation time, free length, free volume, internal pressures have been calculated. The variation of these parameters with percentage concentration was found to be useful in understanding the nature of interactions between the components.

2. Materials and Methods

Chloroquine phosphate used in the present work was of analytical reagent (AR) grade with a minimum assay of 99.9%, used without purification. Different concentrations of solution were prepared by adding sufficient amount of solvent water to chloroquine phosphate. The ultrasonic velocities (U) have been measured in ultrasonic interferometer Mittal Model-F-05 with an accuracy of 0.1%. The viscosities (η) of binary mixtures were determined using Ostwald's viscometer by calibrating with doubly distilled water with an accuracy of ±0.001 PaSec. The densities (ρ) of these binary solutions were measured accurately using 25 ml specific gravity bottle in

an electronic balance, precisely and accurately. The basic parameter U, η, and ρ were measured at various concentrations (0.0125 % to 0.1%) and temperatures (303, 308 & 313 K). The various acoustical parameters were calculated from U, η & ρ value using standard formulae.

On using ultrasonic velocity, density and viscosity the following acoustical parameters like adiabatic compressibility (β_a), intermolecular free length (L_f), relaxation time (τ), free volume (V_f), internal pressure (Π_i), acoustic impedance (Z), surface tension(S), attenuation (α/f^2), Rao's constant (R), molar volume (V_m),cohesive energy (CE) were calculated by applying the known expressions [43].

3. Result and Discussion

The measured values of ultrasonic velocity, density and related thermo-acoustical parameters like adiabatic compressibility (β_a), intermolecular free length (L_f), relaxation time (τ), free volume (V_f), internal pressure (Π_i), acoustic impedance (Z), Wada's constant (W), ultrasonic attenuation (α/f^2), Rao's constant (R), molar volume (V_m),cohesive energy (CE) of chloroquine phosphate with water at 303K, 308 K, and 313 K temperatures in different concentrations are shown in Table 1 and Table 2.

Table 1(a). Solution of chloroquine phosphate in water at 303 K.

Concentration (%)	Density (Kgm^{-3})	Viscosity $x10^{-3}$ (Nsm^{-2})	Ultrasonic Velocity (m/s)	Adiabatic compressibility $x10^{-10}$ (m^2 /N)	Intermolecular free length $x10^{-11}$ (m)	Free Volume $x10^{-3}$ (m^3 mol^{-1})	Rao's constant
0.0125	987.52	0.7913	1525	4.311	4.11957	111.911	5.9525
0.025	998.96	0.8192	1527	4.293	4.11096	106.451	5.9465
0.05	1000.1	0.8481	1533	4.254	4.09225	101.653	5.9471
0.1	1004.4	0.9173	1535	4.225	4.07828	90.547	5.9243

Table 1(b). Solution of chloroquine phosphate in water at 308 K.

Concentration (%)	Density (Kgm^{-3})	Viscosity $x10^{-3}$ (Nsm^{-2})	Ultrasonic Velocity (m/s)	Adiabatic compressibility $x10^{-10}$ (m^2 /N)	Intermolecular free length $x10^{-11}$(m)	Free Volume $x10^{-6}$ (m^3 mol^{-1})	Rao's constant
0.0125	988.36	0.7349	1515	4.408	4.2019	3.9152	5.994
0.025	990	0.7743	1520	4.372	4.1846	3.6382	5.991
0.05	990.6	0.7939	1528	4.324	4.1615	3.532	5.998
0.1	992.08	0.8047	1531	4.300	4.1502	3.227	5.993

Table 1(c). Solution of chloroquine phosphate in water at 313 K.

Concentration (%)	Density (Kgm^{-3})	Viscosity $x10^{-3}$ (Nsm^{-2})	Ultrasonic Velocity (m/s)	Adiabatic compressibility $x10^{-10}$ (m^2 /N)	Intermolecular free length $x10^{-11}$(m)	Free Volume $x10^{-2}$ (m^3 mol^{-01})	Rao's constant
0.0125	985.4	0.6395	1507	4.468	4.2670	151.31	6.002
0.025	987.96	0.6763	1510	4.439	4.2530	139.55	5.990
0.05	988.76	0.7041	1518	4.389	4.2289	132.44	5.996
0.1	991.16	0.7147	1521	4.361	4.2155	129.86	5.985

Table 2(a). Solution of chloroquine phosphate in water at 303 K.

Concentration (%)	Internal pressure $x10^3$ ($Nm^{-2)}$	Acoustic Impedance $x10^3$ (Kg^{-1} m^2 S^{-1})	Relaxation time $x10^{-6}$ (S)	Ultrasonic attenuation $x10^{-15}$ (s^2m^{-1})	Wada's constant $x10^1$	Cohesive energy $x10^3$ (KJ/Mole)	Molar volume $x10^{-3}$ (m^3/mol)
0.0125	16.227	1.521	2.4476	5.892	1.1259	8.3911	517.1
0.025	16.516	1.525	2.5442	6.067	1.1250	8.5289	516.4
0.05	16.785	1.533	2.6579	6.200	1.1253	8.6579	515.8
0.1	17.495	1.542	2.8946	6.651	1.1214	8.9855	513.65

Table 2(b). Solution of chloroquine phosphate in water at 308 K.

Concentration (%)	Internal pressure $x10^3$ ($Nm^{-2)}$	Acoustic Impedance $x10^6$ (Kg^{-1} m^2 S^{-1})	Relaxation time $x10^{-6}$ (S)	Ultrasonic attenuation $x10^{-15}$ (s^2m^{-1})	Wada's constant $x10^1$	Cohesive energy $x10^3$ (KJ/Mole)	Molar volume (m^3/mol)
0.0125	4.7089	1.497	2.2228	5.632	1.1327	2.4575	521.9
0.025	4.8255	1.505	2.3614	5.866	1.1322	2.5145	521.1
0.05	4.8007	1.514	2.4482	5.917	1.1333	2.5425	520.8
0.1	16.5413	1.519	2.619	6.250	1.1325	8.6013	520

Table 2(c). Solution of chloroquine phosphate in water at 313 K.

Concentration (%)	Internal pressure $x10^3$ ($Nm^{-2)}$	Acoustic Impedance $x10^6$ (Kg^{-1} m^2 S^{-1})	Relaxation time $x10^6$ (S)	Ultrasonic attenuation $x10^{-10}$ (s^2m^{-1})	Wada's constant	Cohesive energy $x10^3$ (KJ/Mole)	Molar volume $x10^{-3}$ (m^3/mol)
0.0125	15.039	1.485	1.9082	4.995	1.134	7.8502	523.5
0.025	15.4742	1.492	2.0313	5.237	1.132	8.0791	522.1
0.05	15.7569	1.501	2.1387	5.361	1.133	8.2167	521.7
0.1	15.8841	1.508	2.1851	5.398	1.131	8.2676	520.5

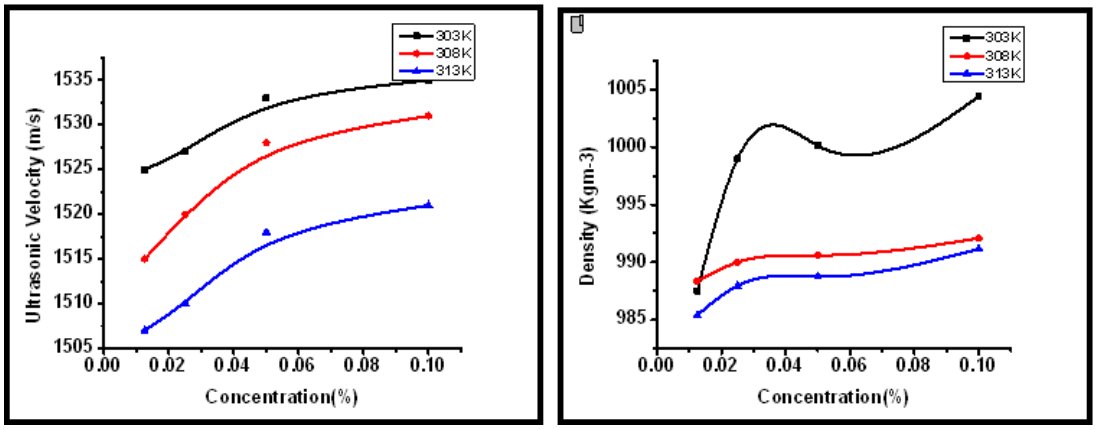

Figure 1. Variation of Ultrasonic velocity and Density with concentration and temperature.

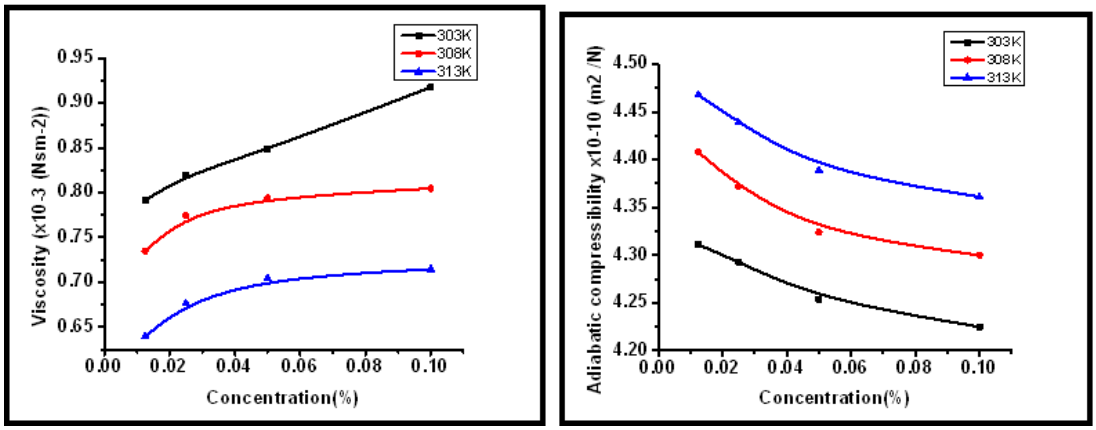

Figure 2. Variation of viscosity and adiabatic compressibility with concentration and temperature.

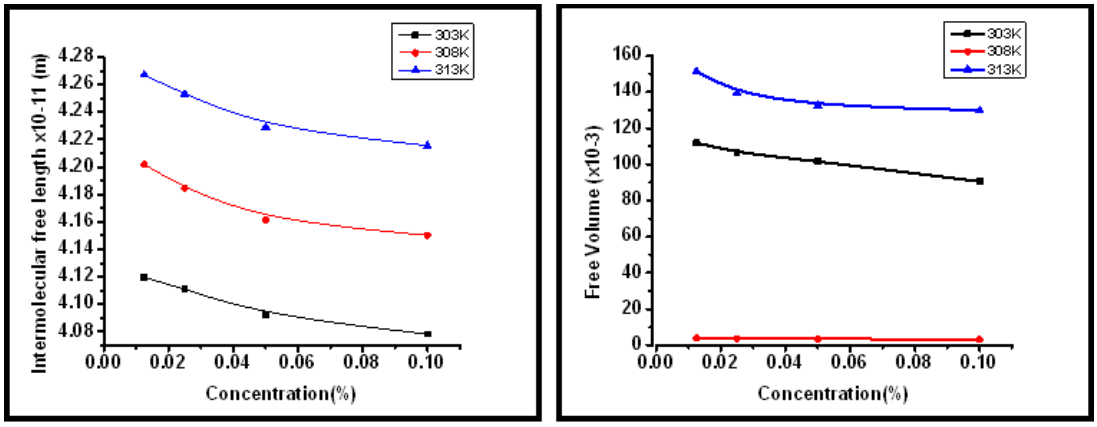

Figure 3. Variation of Intermolecular free length and Free volume with concentration and temperature.

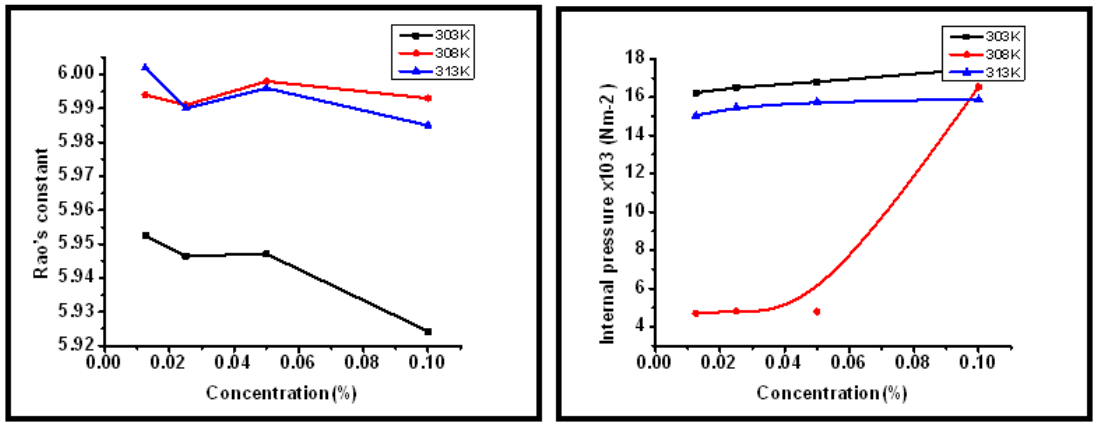

Figure 4. Variation of Rao's constant and Internal pressure with concentration and temperature.

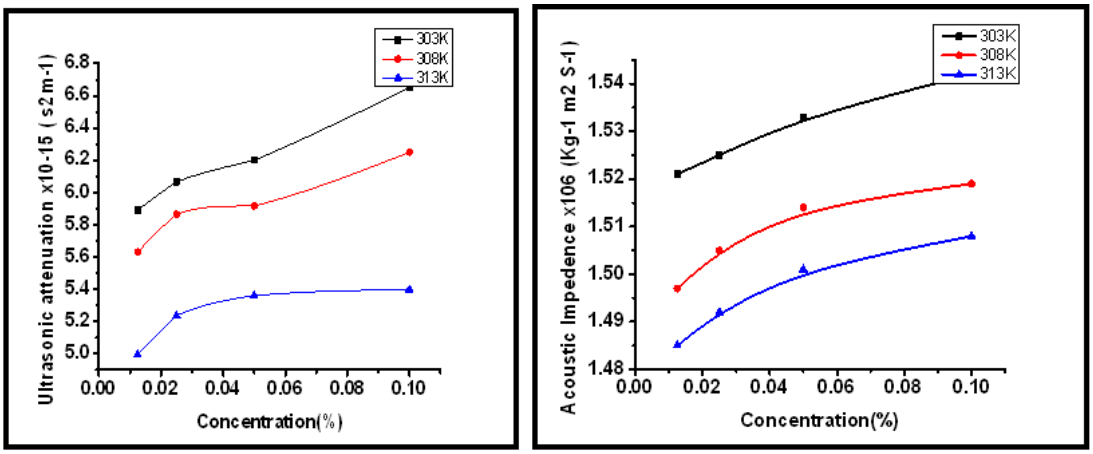

Figure 5. Variation of Ultrasonic attenuation and Acoustic impedance with concentration and temperature

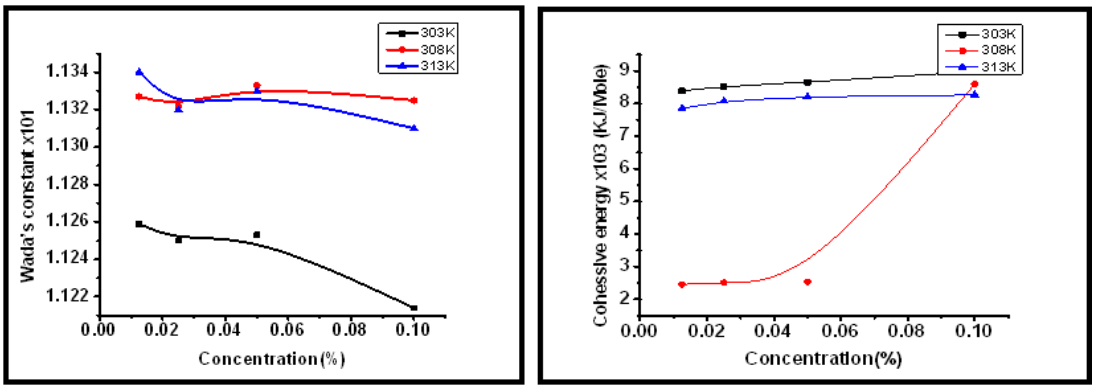

Figure 6. Variation of Wada's constant and cohesive energy with concentration and temperature

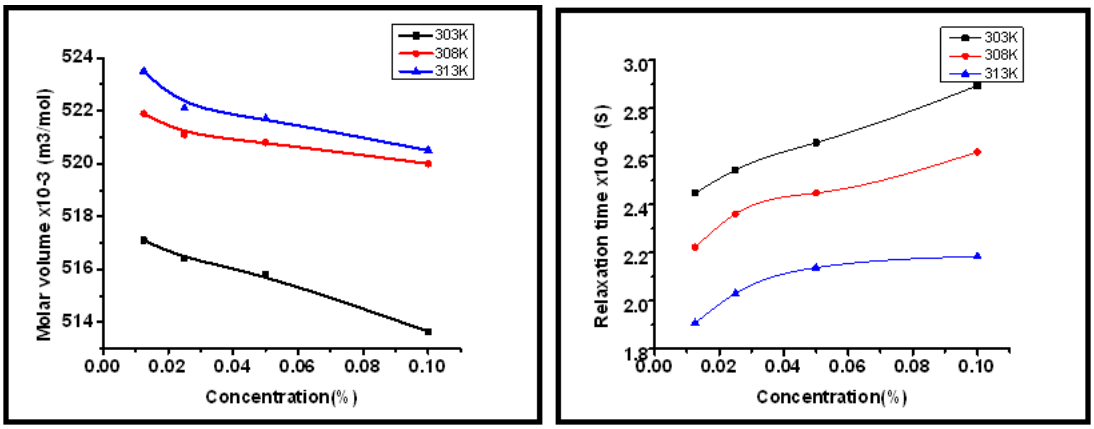

Figure 7. Variation of molar volume and Relaxation time with the concentration and temperature

The variation of acoustical parameters with concentrations and temperature is shown graphically in Figure 1 to Figure 7.

It is observed that ultrasonic velocity and acoustic impedance show nonlinear increasing variation with increase in molar concentration. This indicates that the complex formation and intermolecular weak association which may be due to hydrogen bonding. Thus complex formation can occur at these molar concentrations between the component molecules. Adiabatic compressibility (βa) shows an inverse behavior compared to the ultrasonic velocity. Adiabatic compressibility decreases with increase in concentration of chloroquine phosphate. The decrease in compressibility implies that there is an enhanced molecular association in the system with increase in solute concentration. The opposite trend of ultrasonic velocity and adiabatic compressibility indicate that the association among interacting chloroquine phosphate and water molecules. In the present system of aqueous chloroquine phosphate, free length varies nonlinearly with increase in molar concentration which suggests the significant interaction between solute and solvent due to which structural arrangement is also affected. Relaxation time decreases with increase in concentration. Nonlinear trend of density with concentration indicates the structure-making and breaking property of solvent due to the formation and weakening of H-bonds. The free volume increases and internal pressure decreases with increases in molar concentration indicate the association through hydrogen bonding. It shows the increasing magnitude of interaction between the chloroquine phosphate and water.

4. Conclusion

The ultrasonic study of the liquid mixtures serves as a probe to detect the molecular association arising from the hydrogen bonding between the molecules of chloroquine phosphate and water. The non-linear variation of thermo acoustical parameters with concentration reveals the complex formation between the component molecules. In the present paper the ultrasonic velocity velocity(U), density (ρ) and viscosity (η) and acoustical parameters viz. adiabatic compressibility, intermolecular free length, relaxation time, acoustic impendence, attenuation, Rao's constant, molar volume, cohesive energy, Wada's constant have been measured at different concentrations. The parameters indicate that there is a strong molecular interaction between unlike molecules as the concentration of drug solution increases and the interaction decreases as temperature increases.

Acknowledgements

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