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Studies on thermodynamic properties of streptomycin aqueous solutions from T= (298.15 to 308.15) K

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ABSTRACT

Densities, viscosities, and the speed of sound in aqueous solutions of aminoglycoside antibiotic -streptomycin sulphate have been measured. The influence of concentration and also the temperature upon some physico-chemical properties such as acoustic impedance, adiabatic compressibility, free length, relaxation time, internal pressure, absorption coefficient, free volume, Rao's constant, Wada's constant, cohesive energy, Gibb's free energy, relative association and Van der Waal's constant have also been studied which help in understanding the molecular interactions occurring in the solution. The obtained results show significant influence of concentration and temperature on the phenomenon of molecular aggregation.

Keywords: Streptomycin, Thermodynamic parameters, Aminoglycoside, Molecular interaction

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INTRODUCTION

The molecular interaction of bioactive drug molecule in the body system and its temperature dependence plays a key role in understanding its action like absorption and transport of drug across the biological membranes. Drug macromolecular interactions are important phenomenon involving a complex mechanism in biophysical chemistry [1]. They are also of the great importance in the development of physical chemistry, biological chemistry, and molecular science. The nature and the degree of molecular interactions of bioactive material in aqueous medium are depending on the configuration of the molecule. The information to understand the complex mechanism involved in molecular interactions can be obtained from physico-chemical properties [2]. Many studies on amino acids in aqueous and aqueous electrolyte solutions show that the physico-chemical properties provide the details of solute-solvent, solute-solute and solvent-solvent interactions in aqueous solutions [3-5]. The information regarding the nature and strength of solute-solvent, solute-solute and solvent-solvent interactions in aqueous binary systems by using physico –chemical parameters are also reported by many researchers [7-9]. Thus, the physico-chemical parameters, which can be derived from the basic parameters like density, viscosity and acoustic velocity etc, have a tremendous potential to understand various molecular interactions.

The drug streptomycin is an aminoglycoside antibiotic and was the first effective treatment for tuberculosis. It is derived from the actinobacterium streptomycies griseus. Despite many adverse effects of this medicine such as ototoxicity, nephrotoxicity, fetal auditory toxicity and neuromuscular paralysis [10, 11], it is on the World Health Organization's (WHO) list of essential medicines. The molecular structure of streptomycin sulphate is as shown in Fig.1 for the ready reference.

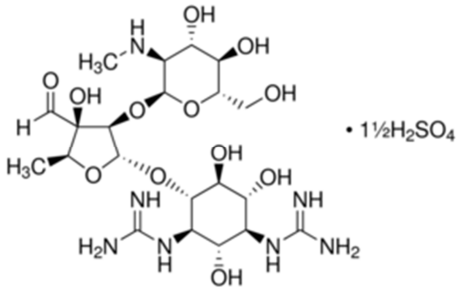

Fig. 1 Molecular structure of streptomycin sulphate

The pharmacodynamics, pharmacokinetics, safety and efficacies of many antibiotic have been widely evaluated [12]. In addition to these studies the physico-chemical data may provide the useful information for understanding the details of the antibiotics. However, a negligible work has been done on physico-chemical studies. Therefore, it is worthwhile to carry out such a study on antibiotic. In the present paper, a systematic study of density, viscosity and ultrasonic velocity of aqueous streptomycin sulphate with different concentration range (0.001 - 0.1) mol kg^{-1} at temperature T= (298.15, 303.15, 308.15) K was carried out and the data was utilized to calculate various thermo-acoustical parameters. The molecular interactions and physico-chemical characteristics of the under-studied antibiotic are also discussed in terms of these parameters.

MATERIALS AND METHODS

The under-studied aminoglycoside antibiotic- streptomycin sulphate CAS No. 3810-74-0, molecular weight of 728.69, Minimum assay 95% was obtained from HIMEDIA Ltd. India. The compound was used as supplied. The solvent double distilled water was used to prepare the fresh solutions. All the solutions were prepared on molality basis by using a monopan electric balance of least count 0.0001g. The densities of solvent and all the freshly prepared solutions were measured by using hydrostatic sinker method at different temperatures T= (298.15, 303.15 and 308.15) K. While measuring the densities, the temperature of experimental liquid was maintained constant by using thermostat U-10 to accuracy 0.1K.The accuracy in the density measurements was ± 0.0001 g/cm^3. The speed of sound at 2 MHz in aqueous solutions of an Aminoglycoside antibiotics streptomycin sulphate of different concentration at different temperatures T= (298.15, 303.15 and 308.15) K were measured by using double walled liquid cell employing pulse- echo overlap technique. Calibration of interferometer was done by measuring speed of sound in double distilled water. The viscosity of binary liquid solutions was measured by using Ostwald's viscometer owing to its versatility. The viscometer was placed in glass jar through which a current of water was maintained constant with the help of thermostat U-10.The jar was properly lagged by asbestos thread leaving suitable window to illuminate and observe the viscometer marks. The constant temperature of water from thermostat is circulated through tubing connected to viscometer. The temperature of water- bath was maintained constant for a long time within an accuracy ±0.1^{O}C. The time of falling of the liquid between the viscometer marks was measured by using an electronic digital timer ET-450A (ECIL) of least count 0.01s. The accuracy of viscosity measurements was ±0.1%.

3. Physical Parameters

The experimental data of density, viscosity and ultrasonic velocity of aqueous Streptomycin sulphate at different concentration and temperatures T= (298.15, 303.15 and 308.15) K is utilized for calculating various thermodynamic parameters using the following empirical relations.

Acoustic impedance (Z)

$$Z = u\rho_s \quad (1)$$

Adiabatic compressibility (β)

$$\beta = 1/u^2\rho_s \quad (2)$$

Where u is ultrasonic velocity and ρ_s is the density of solution

Free length (L_f)

$$L_f = K_1\beta^{1/2} \quad (3)$$

where K_1 is Jacobson, a temperature dependent constant (K_1= (93.875+0.375T) × 10^{-8}

Free Volume (V_f)

$$V_f = \left(M_{eff}u/k\eta\right)^{3/2} \quad (4)$$

where M_{eff} is effective molecular weight, η is viscosity and 'k' is constant equal to 4.28 × 10^9 independent of temperature for all types of liquids.

Internal pressure (π_i)

$$\pi_i = bRT\left(\frac{k\eta}{u}\right)^{\frac{1}{2}}\left(\frac{\rho_s^{\frac{2}{3}}}{M_{eff}^{\frac{7}{6}}}\right) \quad (5)$$

where b stands for the cubic packing factor which is assumed to be '2' for all liquids and solutions, k is temperature independent constant, R is gas constant and T is the absolute temperature.

Relaxation time (τ)

$$\tau = 4/3\beta\eta \quad (6)$$

Absorption coefficient (α/f^2)

$$(\alpha/f^2) = 8\pi^2\eta/3\rho_s u^3 \quad (7)$$

Cohesive energy

$$C.E. = \pi_i \times V_f \quad (8)$$

Gibb's free energy (ΔG)

$$\Delta G = -kTlog(\frac{h}{\tau kT}) \quad (9)$$

where k is the Boltzmann's constant (1.23 × 10^{-23}J.K^{-1}), h the Planck's constant (6.62 ×10^{-34}J.s)

Relative association (R_A)

$$R_A = (\rho_s/\rho_0)\,(u_0/u)^{\frac{1}{3}} \quad (10)$$

where ρ_s, u and ρ_0 , u_0 are respectively the density and ultrasonic velocity of the solution and solvent.

Rao's constant (R_a)

$$R_a = (M_{eff}/\rho_s)\,(u)^{\frac{1}{3}} \quad (11)$$

Molar compressibility or Wada's constant (W)

$$W = \left(\frac{M_{eff}}{\rho_s}\right)\beta^{-1/7} \quad (12)$$

Van der Waal's constant (b)

$$b = V_f\left(1 - \left(\frac{RT}{M_{eff}u^2}\right)\left(1 + \frac{M_{eff}u^2}{3RT}\right)^{1/2}\right) \quad (13)$$

RESULTS AND DISCUSSION

Table 1: Measured parameters of Streptomycin aqueous solutions for five different concentrations at temperatures T= 298.15, 303.15 and 308.15 K

Parameter	Temperature (K)	Value of parameters measured for concentrations (mol.kg^{-1})				
		m=0.0010	0.0050	0.0100	0.0501	0.1003
Density ρ_s 10^3 (Kg.m^{-3})	298.15	0.9976	0.9995	1.0013	1.0131	1.0278
	303.15	0.9967	0.9986	0.9999	1.0117	1.0267
	308.15	0.9955	0.9970	0.9980	1.0098	1.0246
Ultrasonic velocity u (m.s^{-1})	298.15	1496.70	1498.54	1503.75	1510.90	1519.10
	303.15	1509.23	1511.08	1516.29	1523.43	1531.63
	308.15	1518.60	1520.45	1526.02	1532.90	1542.08
Viscosity η 10^{-3} (N.s.m^{-2})	298.15	0.902	0.967	0.986	1.082	1.208
	303.15	0.804	0.852	0.866	0.951	1.052
	308.15	0.729	0.774	0.783	0.849	0.923

Table 2: Derived parameters of Streptomycin aqueous solutions for five different concentrations at temperatures T= 298.15, 303.15 and 308.15 K

Parameter	Temperature (K)	Value of parameters obtained for concentrations (mol.kg^{-1})				
		m=0.0010	0.0050	0.0100	0.0501	0.1003
Acoustic impedance Z 10^6 (Kg. m^{-2}.s^{-1})	298.15	1.493	1.498	1.506	1.531	1.561
	303.15	1.504	1.509	1.516	1.541	1.572
	308.15	1.512	1.516	1.523	1.548	1.580
Adiabatic compressibility β 10^{-10} (N^{-1}m^2)	298.15	4.475	4.455	4.417	4.324	4.216
	303.15	4.405	4.386	4.350	4.259	4.152
Free length L_f 10^{-11} (m)	308.15	4.356	4.339	4.303	4.214	4.104
	298.15	4.351	4.342	4.323	4.277	4.224
	303.15	4.357	4.348	4.330	4.284	4.230
Free volume V_f 10^{-8} (m^3 mol^{-1})	308.15	4.370	4.362	4.344	4.299	4.242
	298.15	1.845	1.673	1.642	1.504	1.345
	303.15	2.221	2.048	2.020	1.843	1.685
Internal pressure π_i 10^9 (Pa)	308.15	2.595	2.387	2.372	2.206	2.065
	298.15	2.726	2.815	2.829	2.882	2.953
	303.15	2.604	2.674	2.682	2.736	2.783
Relaxation time τ 10^{-13} (s)	308.15	2.511	2.580	2.581	2.616	2.639
	298.15	5.382	5.744	5.806	6.226	6.802
	303.15	4.722	4.982	5.023	5.400	5.813
Absorption coefficient α/f^2 10^{-15}	308.15	4.234	4.478	4.492	4.771	5.051
	298.15	7.090	7.559	7.614	8.126	8.830
Cohesive energy	303.15	6.169	6.502	6.532	6.990	7.484
(J mol^{-1})	308.15	5.498	5.807	5.805	6.137	6.459
	298.15	50.306	47.086	46.450	43.344	39.716
Gibb's free	303.15	57.829	54.760	54.171	50.418	46.888
Energy	308.15	65.181	61.587	61.215	57.692	54.517
10^{-21}(J mol^{-1})						
	298.15	4.007	4.246	4.285	4.541	4.866
Relative	303.15	3.648	3.848	3.878	4.149	4.423
Association(R_A)	308.15	3.357	3.569	3.581	3.809	4.026
	298.15	0.9979	0.9994	1.0000	1.0102	1.0230
	303.15	1.0010	1.0025	1.0027	1.0129	1.0261
	308.15	1.0015	1.0026	1.0024	1.0127	1.0255

Table 3: Fluctuations in some constants of Streptomycin aqueous solutions due to variation in concentrations and temperatures

Constants	Temperature (K)	Value of constants obtained for concentrations (mol.kg^{-1})				
		m=0.0010	0.0050	0.0100	0.0501	0.1003
Rao's constant R_a 10^{-4} (m^5 N^{-1})	298.15	2.065	2.068	2.074	2.111	2.156
	303.15	2.073	2.076	2.083	2.120	2.165
	308.15	2.080	2.083	2.091	2.129	2.175
Wada's constant W 10^{-4} (m^4 s^{-1})	298.15	3.910	3.917	3.928	4.005	4.097
	303.15	3.923	3.929	3.942	4.019	4.111
	308.15	3.934	3.941	3.956	4.033	4.127
Van der Waal's constant 10^{-5} cm^3mole^{-1}	298.15	1.524	1.527	1.531	1.562	1.600
	303.15	1.526	1.528	1.533	1.564	1.602
	308.15	1.527	1.530	1.536	1.567	1.605

Table 1 shows the values of measured basic parameters i.e. density, viscosity and ultrasonic velocity of aqueous solutions of streptomycin sulphate for five different concentrations namely 0.001, 0.005, 0.010, 0.0501 and 0.1003 mol kg^{-1} at three different temperatures 298.15, 303.15 and 308.15K. Variations of these parameters against the change in temperature are shown in Fig 2a, b and c.

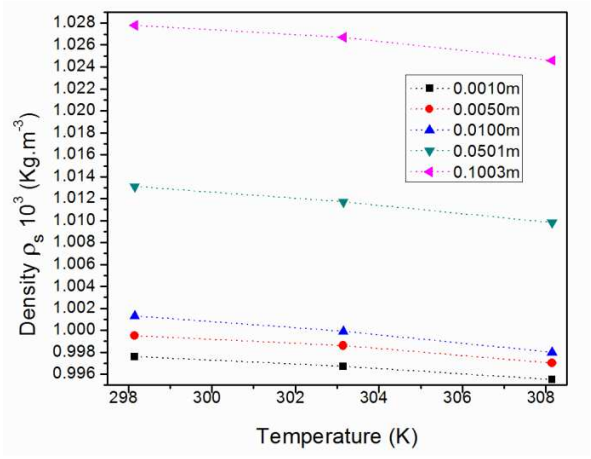

Figure 1a

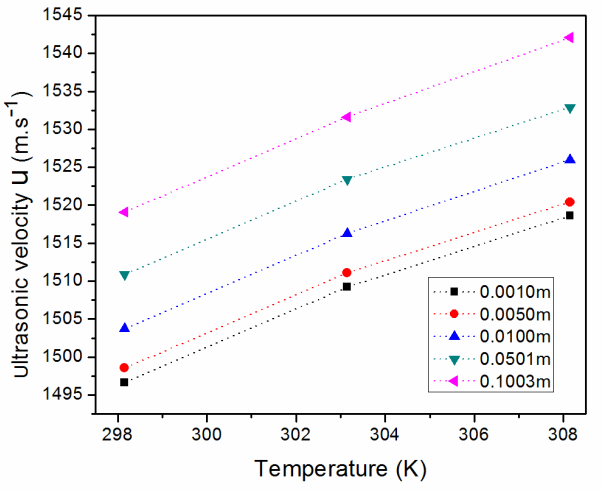

Figure 2b

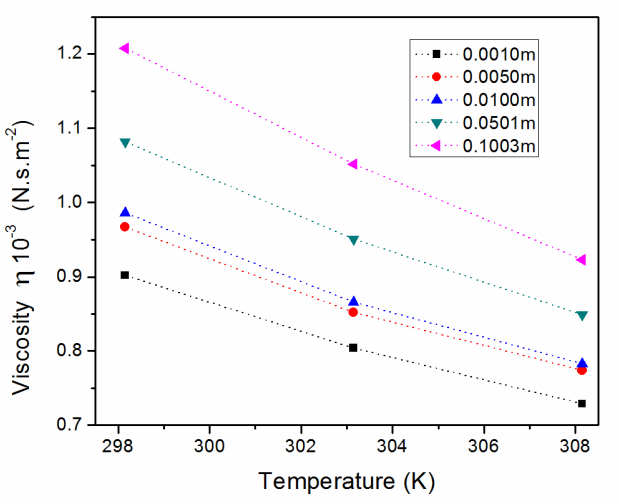

Figure 2c

Fig. 2 Temperature dependence of measured parameters of aqueous streptomycin sulphate for different concentrations 0.001, 0.005, 0.010, 0.0501 and 0.1003 mol kg^{-1} (a) density, (b) speed of sound and (c) viscosity

Fig.2a shows the decrease in density of the aqueous solution of streptomycin sulphate which is almost linear with the rise in temperature for all given concentration. It also shows the increase in density with the increase in concentration. This result is obvious as the volume of the solution increases with the temperature and mass of the solution increase with the concentration. This result also leads to conclusion that it is the composition and not the aggregation that influence the density of the drug. The conclusion is well supported by the earlier study on trimethyl ammonium bromide solutions by Gomez-Diaz et.al. [13]. Fig.2b shows the speed of sound (u) increase with temperature as well as with the concentration. The change in speed of sound in liquid has taken place due to some kinds of aggregation process [14, 15]. According to the literature survey, greater is the association of solute and solvent molecule, greater is the speed of sound in liquid. Thus in the present case of streptomycin sulphate the aggregation process increases with the temperature and also with the concentration. Similar results about the speed of sound in aqueous solutions of various compounds have been already reported [16, 17].

Viscosity (η) is another parameter of the liquid, which is greatly affected by the concentration and the temperature. With the rise in concentration of the solute in solution, the viscosity of the solution in general increases because the increase in concentration increases the number of solute particles in solution. Solvent molecules get attracted towards solute molecule; they strongly interact with one another, results in solute-solvent interactions thereby increasing the viscosity of the solution. This clearly indicates the structure-making tendency of the solute in the solution. Increase in viscosity of the solution may also be due to increase in number of solute particles lying across the fluid streamlines. These particles are subjected to the torsional force due to which they tend to rotate and absorb energy. This absorbed energy corresponds to the increase in viscosity of the solution [18]. The decrease in viscosity of the solution with the rise in temperature occurs as molecules in the solution acquires more and more thermal energy which in turn the motion of molecules increases at the expense of cohesive forces acting between the molecules. Since the solution faces lesser resistance to flow, the decrease in viscosity of the solution will be the result [19]. The variation of viscosity of aqueous streptomycin sulphate as shown in Fig.2c is agreed with these facts.

The solute- solvent interaction can be understood also by knowing physico-chemical parameters. Various physico-chemical parameters such as Acoustic impedance (Z), Adiabatic compressibility (β), Free length (L_f), Free Volume (V_f), Internal pressure (π_i), Relaxation time (τ), Absorption coefficient (α/f^2), Cohesive energy, Gibb's free energy (ΔG) and Relative association (R_A) of aqueous solution of streptomycin sulphate for five different concentrations 0.001, 0.005, 0.010, 0.0501 and 0.1003 mol kg^{-1} at three different temperatures 298.15, 303.15 and 308.15K using the data of Table 1 and formulae given by equations (1) to (9) were calculated. They are shown in Table 2. Variations of these parameters against the change in temperature are shown in Fig 3a- h.

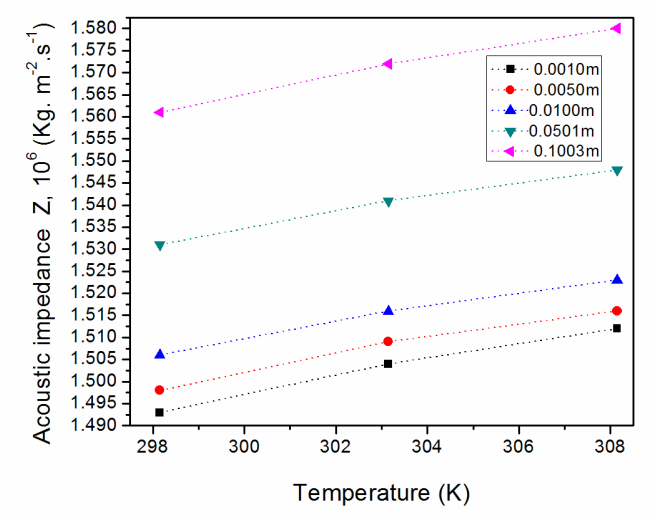

Figure 2a

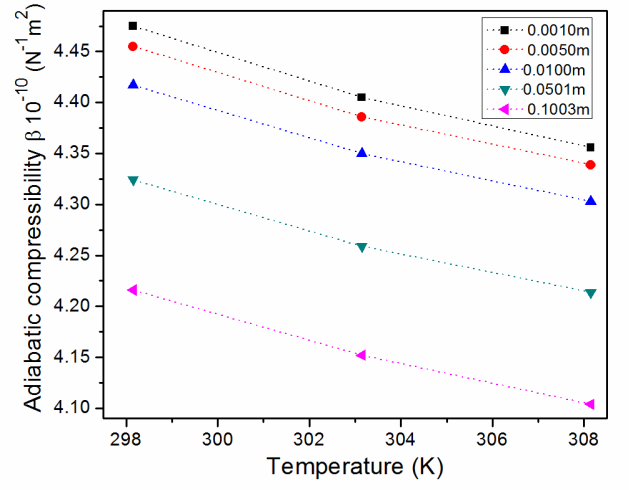

Figure 3b

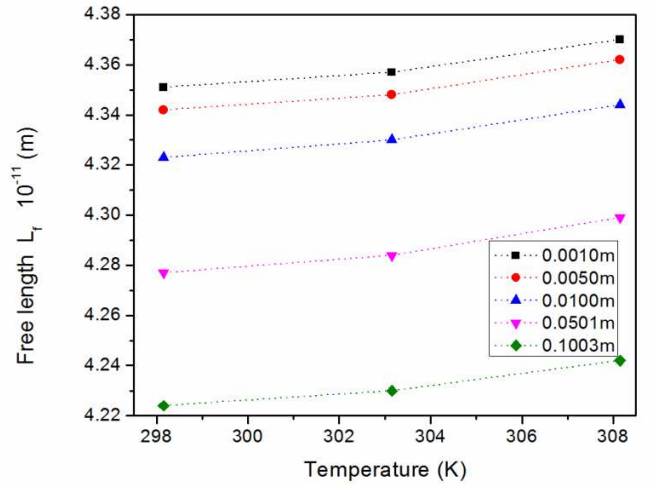

Figure 3c

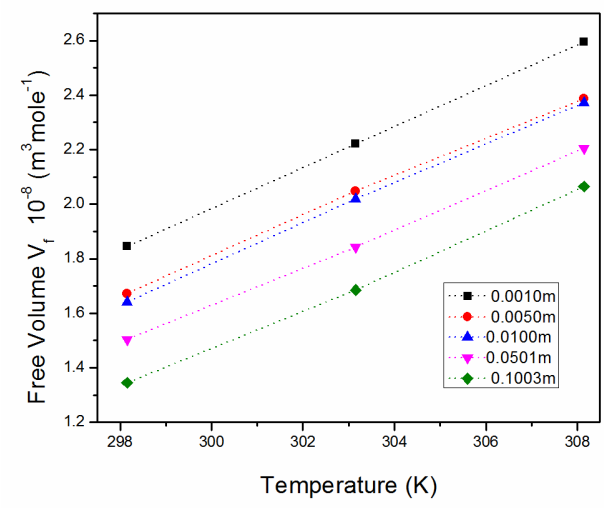

Figure 3d

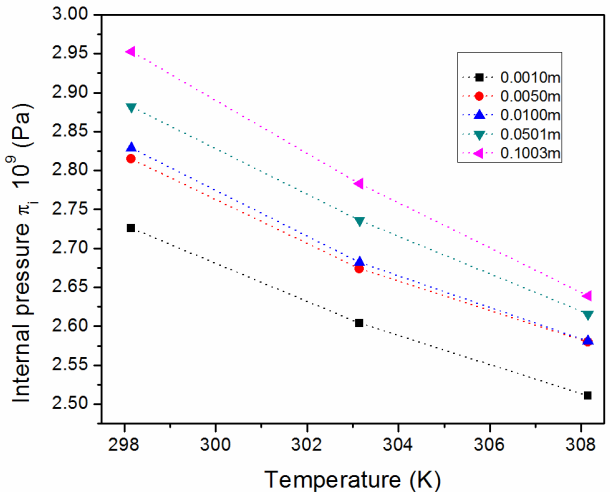

Figure 3e

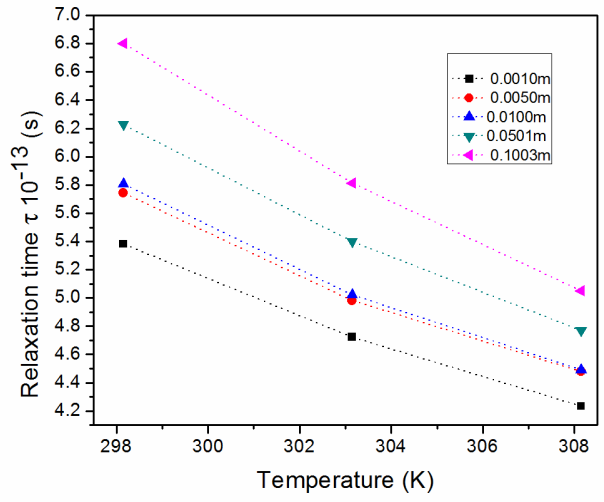

Figure 3f

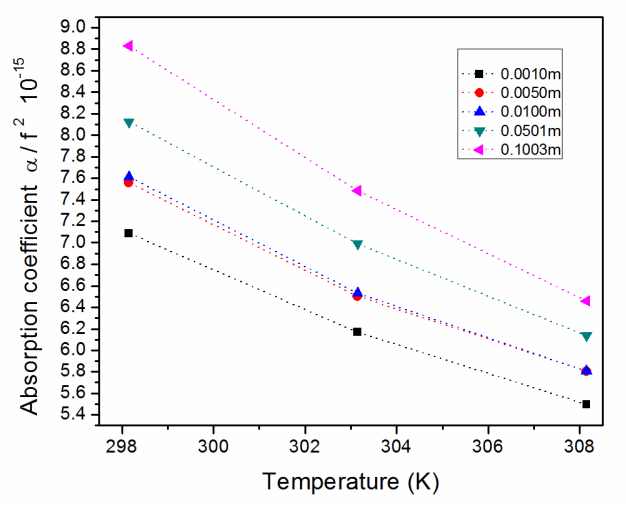

Figure 3g

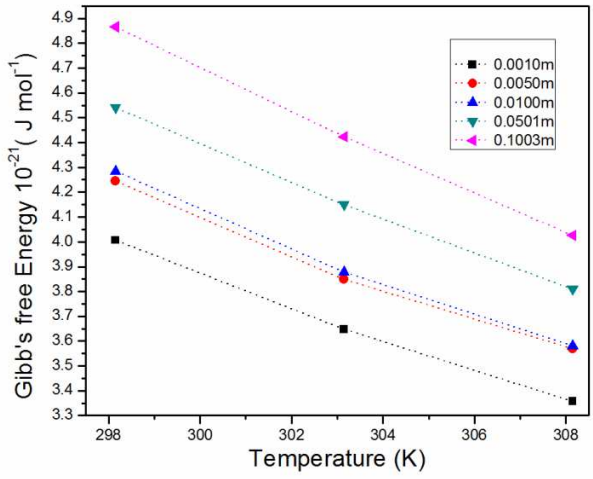

Figure 3h

Fig. 3 Temperature dependence of derived parameters of aqueous streptomycin sulphate for different concentrations (0.001, 0.005, 0.010, 0.0501 and 0.1003) mol kg^{-1} (a) acoustic impedance, (b) adiabatic compressibility, (c) free length, (d) free volume, (e) internal pressure, (f) relaxation time, (g) absorption coefficient and (h) Gibb's free energy

The value of acoustic impedance (Z) of aqueous streptomycin sulphate was found to be of the order of 10^6 (Kg. $m^{-2}.s^{-1}$) (Table 2) and increases slightly and linearly with the increase in temperature. The increase in Z is also observed with increase in concentration (Fig. 3a). The increase in Z with the temperature is obvious as it relates with u and $\rho_s$by Eq.1 and the increase in u is dominant than the decrease in ρ_s. The increasing trend of Z with the increase in concentration indicates that there is a strong association between solute (streptomycin sulphate) and solvent (water) through intermolecular hydrogen bonding. From Fig. 1 it is seen that there are various functional groups such as –OH, $-NH_2$, H-C=O etc, in the molecule of streptomycin sulphate. These groups are responsible for intermolecular hydrogen bonding with water. During association the –OH group may form more and stronger hydrogen bonds with water molecule compared to other active groups that strengthen the dipole-dipole interactions [20, 21].

Fig.3b shows the variation of adiabatic compressibility (β) against temperature of aqueous solution of streptomycin sulphate. It shows that the adiabatic compressibility decreases with the concentration and with the temperature. The decrease in adiabatic compressibility with the concentration indicates the enhancement of molecular association [22] and increase in electro-strictive compression of water around the solute molecule [23, 24]. Thus the decrease in

compressibility with the rise in concentration and the temperature is attributed to the intrinsic ionic compressibility and structural (hydrophobic hydration) factors [25].

The decrease in free length with the rise in concentration (mol kg^{-1}) of aqueous streptomycin sulphate solution is seen from the Fig.3c. This nature indicates that with the rise in concentration (mol kg^{-1}), the solute-solvent interactions and the aggregation become significant. The increase in temperature increases the thermal energy of the solution, which in turn increases the molecular motion and produces disordered structure; result in more free space between the molecules and the increases in volume expansion. This finding leads to the conclusion that the aggregation phenomenon has significant influence on this physical property. Mehra and Malav [26] already reported a similar behavior of free length.

The free volume is another parameter which can be correlated to the structure making and breaking tendency of ions [27]. In Fig. 3d the decrease in free volume with rise in concentration leads to the fact that aqueous solution of streptomycin sulphate has structure making nature whereas increase in free volume with rise in temperature indicates structure breaking nature.

Fig. 3e shows the variation of internal pressure (π_i) against temperature and concentration for aqueous streptomycin sulphate. The increase in internal pressure of aqueous solution with rise in concentration is due to the increase in cohesive forces and solute-solvent interactions; and decrease in internal pressure of the solution with the rise in temperature is due to the thermal agitation of molecules caused by increase in thermal energy of the system which reduces the cohesive forces and decreases the solute-solvent interactions [28, 29]. Increase in internal pressure confirms the structure-making tendency of the solute in solvent, also the presence of solute-solvent interaction and the formation of intermolecular hydrogen bonding between solute and solvent and vice-versa [30].

A variation of relaxation time (τ) with the temperature of streptomycin sulphate at different concentration is as shown in Fig.3f. The graph shows a trend similar to that of viscosity. Structural relaxation processes occurring due to rearrangement of molecule in solutions mainly affects the relaxation time values for solution [31].

Absorption coefficient ($\frac{\alpha}{f^2}$) is the characteristic of the liquid medium and depends upon the external conditions like temperature, pressure, and frequency [22]. It increases with increase in concentration [32]. The behavior of solution of streptomycin sulphate is as shown in Fig.3g. Increase in absorption coefficient observed in the present study with rise in concentration clearly indicates the increase in molecular aggregation and the increase in intermolecular hydrogen bonding resulting in close-packed water structure, which absorbs more sound energy. The decrease in absorption coefficient at higher temperature of the solution occurs due to loosely packed water structure which absorbs less sound energy.

The closer packing of molecule due to hydrogen bonding in the solution is revealed by the Gibb's free energy(ΔG) [26]. In the present work, increase in Gibb's free energy values is observed with rise in concentration (Fig. 3h). Higher values of Gibb's free energy indicate the strong molecular aggregation and minimum free spacing between the molecules due to formation of stronger hydrogen bond. The decrease in Gibb's free energy with the increase in temperature shows that molecules are loosely packed due to weakening of hydrogen bonding between the solute and the solvent and thus lead to the dissociation [33].

As concentration increases, the relative association (R_A) increases slightly. The values of relative association of the streptomycin sulphate at different experimental conditions are shown in Table 2. For the calculations, the values of ρ_0 and u_0 were taken as 996.9, 995.7, 994.0 kg/m^3 and 1496.69, 1509.21, 1518.48 m/s for three temperatures 298.15, 303.15 and 208.15K respectively. The R_A factor suggests that the unlike interactions are predominant over the like interactions. The increase in R_A value with concentration also indicates the strengthening of solute-solvent interactions, which may be due to accumulation of mass rather than enlargement in size of the solute in the solution. The similar observations are reported in earlier studies of some binary mixtures [34]

For the legality of the values of parameters shown in Table 2, some constants like Rao's constant, Wada's constant and Van der Waal's constant of aqueous streptomycin sulphate solution was also determined. The values of these constant are given in Table 3. The orders of these constant is found to be same as compared to the literature data [26,

28]. All these constant shows minor increase in their respective values due to increase in concentration. These variations leads to conclusion that medium is closely packed and favors the increase in solute-solvent interactions.

CONCLUSION

In the present work, the evaluated acoustic parameters confirm the increase in molecular aggregation, solute-solvent interactions, intermolecular hydrogen bonding with increase in addition of solute into the solvent. The effect of the rise in temperature is subtractive. Density has no influence on the molecular aggregation phenomena. The thermodynamic parameter relative association shows solute-solvent interactions are predominant over solute-solute interactions with rise in concentration.

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