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Research Article

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Ultrasonic studies on molecular interactions of $ZnSO_4$ in aqueous solutions of glucose at various concentrations

Neha Sawhney, Mukesh Kumar, Amit Kumar Sharma and Meena Sharma*

Department of Chemistry, University of Jammu (J&K), India

ABSTRACT

The values of density (ρ), ultrasonic velocity (U) and viscosity (η) for various concentrations of $ZnSO_4$ has been measured in various aqueous solutions of glucose at $7MH_z$ and at 303.15 K in order to study the ion–solvent interactions. By using standard relations from measured values of density, ultrasonic velocity (U) and viscosity (η), the desired acoustical and thermodynamic parameters such as adiabatic compressibility (β_{ad}), intermolecular free length (Lf), acoustic impedance (Z), relaxation time (τ), rao's constant (R_M), wada's constant (W), free volume (Vf), gibb's free energy (ΔG) and available volume (V_a) have been calculated. The changes in these parameters with change in concentration have been explained on the basis of solute-solvent interactions and structure modifying tendency of solute in solvent. It is observed that ion–solvent interactions increase with increase in concentration of Zn^{++} ions in aqueous solutions of glucose.

Keywords: Ultrasonic velocity, viscocities, acoustical parameters, adiabatic compressibility, molecular association.

INTRODUCTION

Ultrasonic studies helps in characterising acoustical and thermodynamic behaviour of various liquid mixtures [1-4]. Ultrasonic waves are also known as acoustic waves that are propagating with frequency above 20 $KH_{z.}$ In ultrasonic velocimetry, the propagation velocity of the ultrasonic wave, called the ultrasonic velocity, and is measured by applying wave frequencies in the frequency range between 5 to 10 MHz. Ultrasonic velocity of any medium depends on density and adiabatic compressibility of that medium through a simple mathematical relation called Newton-Laplace equation:

$$U = {}^{1}\!/_{(\eta\beta_{\mathrm{ad}})^{1/2}}$$

The ultrasonic velocity is thus having tendency of providing volumetric and elastic information on solutes in aqueous solution [5]. Ultrasonic methods have been preferred as compared to other techniques (Wyn et al.,1966) like Infrared spectroscopy, Nuclear Magnetic Resonance, etc., as Speed of sound itself is highly sensitive to the structure and molecular interactions present in the liquid mixtures that are related to the binding forces between the constituents of the medium [6]. For estimation of the molecular interactions in solutions qualitatively, the ultrasonic velocity approach was first studied by Lageman [7]. Ultrasonic methods helps for studying molecular interactions and studies on these interactions are significant for investigating their physico-chemical behaviour.

Carbohydrates are one of the most important classes of organic compound that can present abundantly in living nature .Carbohydrates play vital role in variety of areas including biological, industrial applications. The preservation of sugar rich food products are directly related to sugar water interactions. Carbohydrates and metal cations coexist in biological fluids. Carbohydrates metal complexes plays a vital role in chemistry and biology so various studies of interaction between carbohydrates and metal cations have been carried out. Measurement of ultrasonic velocity and related parameters give insight into solution properties. As ultrasonic waves pass through liquid medium they undergo dispersion and disturbing the equilibrium between the molecules. The velocity of ultrasonic waves plays a very important role for interpreting ion-solvent interactions or solute-solvent interactions which determine the chemical structure of solute-solvent molecules. Jacobson's model and Shaaf's model have been extensively used to explain and interpret the results [8,9].

EXPERIMENTAL SECTION

Glucose and Zinc Sulphate (A R grade) were dried under vaccum for 24 hours before use to minimize decomposition due to bacterial contamination. Triply distilled water was used for preparation of various solutions and this was prepared by distilling ordinary water thrice over alkaline $KMnO_4$ in all glass apparatus.

Density Measurement

Densities of various solutions were measured using 5 ml capacity of specific gravity bottle. The temperature is maintained constant by immersing the bottle in Juloba thermostat for 15 minutes. The density can be calculated using the formula

$$\rho_2 = \left({}^{w_2}/_{w_1}\right) \rho_1 \qquad (\text{kgm}^{-3}) \qquad (1)$$

where,
w_1, is the weight of the distilled water.
w_2, is the weight of the experimental liquid.
ρ_1, is the density of water.
ρ_2, is the density of experimental liquid.
Density of water is taken from the literature
Weight can be determined by using digital balance with accuracy of $0.0001 g/m^3$.

Viscosity measurement

The viscosity of the aqueous solutions is measured using an Ostwald's viscometer calibrated with doubly distilled water. The Ostwald's Viscometer with the experimental liquid is immersed in a temperature controlled water bath. The flow of time was measured by a digital stop watch. The viscosity can be calculated using the formula

$$\eta_2 = \eta_1 \left({}^{t_2}/_{t_1}\right) \left({}^{\rho_2}/_{\rho_1}\right) \qquad (\text{kgm}^{-1}\text{s}^{-1}) \qquad (2)$$

where,
η_1, is the Viscosity of water
t_1, is the time of flow of water
ρ_1 , is the density of water.
η_2, is the viscosity of the experimental liquid.
t_2, is the time of flow of the experimental liquid.
ρ_2, is the density of the experimental liquid.

Velocity Measurement

Ultrasonic velocity for different solutions was measured using Single crystal interferometer (Mittal Enterprises, New Delhi) with frequency 7 MHz, Fig.1. Constant temperature was maintained by thermostatic water bath arrangement with accuracy of $\pm 0.01\,^{o}C$. The experimental liquid was taken in a measuring cell and this electronically operated constant temperature bath has been used to circulate water through the outer jacket of this double walled cell. The principle used in the measurement of velocity 'v' is based on the accurate determination of the wavelength 'λ' in the medium. Ultrasonic waves of known frequency 'f ' are produced by a quartz plate fixed at the bottom of the cell. The waves are reflected by a movable metallic plate kept parallel to quartz plate. Standing waves are formed in the medium if the separation between the plates is exactly a whole multiple of sound wavelength. The acoustic resonance give rise to an electrical reaction on the generator driving the quartz plate and the anode current of the generator become maximum.

If the distance is now increased or decreased and the variation is exactly one half wavelengths or multiple of it, anode current again become maximum. By noting the initial and final position of the micrometer for n complete movements (maxima-minima-maxima) of the micro-ammeter needle, one can find out the distance (*d*) moved by the parallel reflector. The wavelength is calculated as

$$\lambda = 2d/n \qquad (3)$$

From the knowledge of wavelength 'λ' the velocity 'v' can obtained by the relation;

$$u = \lambda \, x \, f \qquad (ms^{-1}) \qquad (4)$$

The apparatus used for measuring ultrasonic velocity of liquids can be represented as follows:

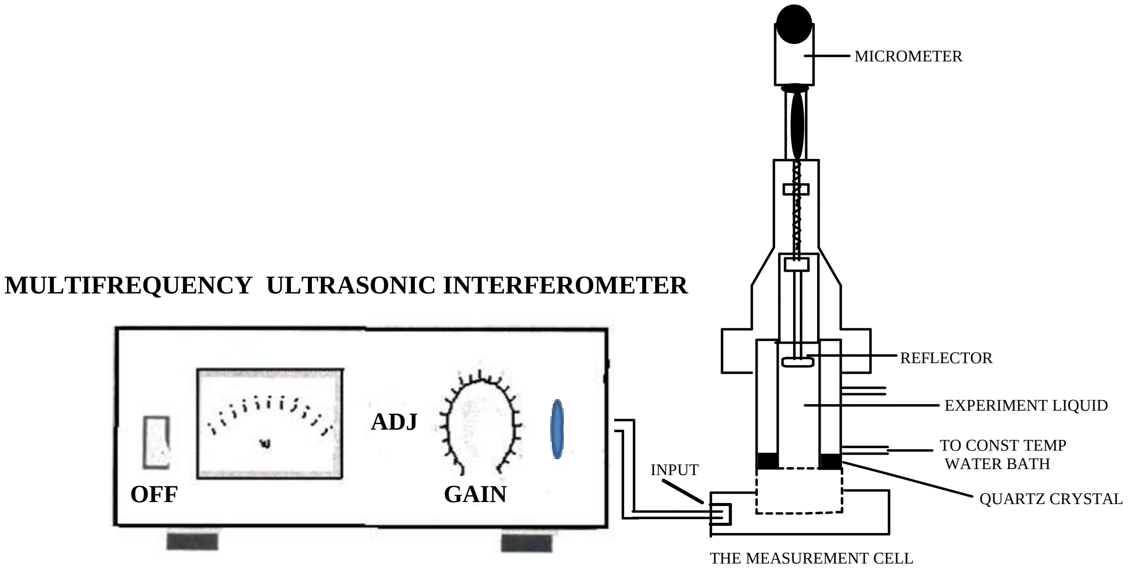

Fig. 1: Schematic representation of apparatus used for measuring ultrasonic velocity of liquids.

RESULTS AND DISSCUSSION

From experimentally determined values of density, ultrasonic velocity and viscosity, various parameters can be calculated such as: adiabatic compressibility (β_{ad}), intermolecular free length (L_f), acoustic impedance (Z), relaxation time (τ), rao's constant (R_M), wada's constant (W), free volume (V_f), gibb's free energy (ΔG) and available volume (V_a) were calculated:

a) Adiabatic compressibility (β_{ad})

Adiabatic compressibility depends on speed of sound and density of medium and thus was calculated by using the equation known as Newton Laplace equation:

$$\beta_{ad} = {}^{1}/_{u^2}\,\rho \; (kg^{-1} ms^{-2}) \qquad (5)$$

Where, u = velocity & ρ = density of given solution

b) Intermolecular free length (L_f)

Intermolecular free length was determined by using the following formula given by Jacobson [10,11]

$$L_f = K_T \, \beta_{ad}^{1/2} \; (m) \qquad (6)$$

Where K_T is temperature dependent constant (= $(93.875+0.375T)x10^{-8}$)

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c) Acoustic impedance (Z)
Acoustic impedance was determined from equation,

$$(\mathrm{kg\ m^{-2}s^{-1}})\ \mathbf{Z} = \mathbf{u} \times \boldsymbol{\rho} \qquad (7)$$

d)The relaxation time (τ)
The relaxation time (τ) [12] was calculated from the relation,

$$\tau = (4/3)\beta_{ad}\eta \qquad (\mathrm{s}) \qquad (8)$$

e) Rao's constant (R_M)
Rao's constant was calculated by using following equation,

$$\mathbf{R_M} = \left(\mathbf{M_{eff}}/\boldsymbol{\rho}\right)\mathbf{u}^{1/3} \qquad (\mathrm{m^{10/3}s^{-1/3}mol^{-1}}) \qquad (9)$$

f) Wada's constant (W)
Wada's constant was calculated by following equation,

$$\mathbf{W} = \left(\mathbf{M_{eff}}/\boldsymbol{\rho}\right)\boldsymbol{\beta}_{\mathbf{ad}}{}^{-1/7} \qquad (\mathrm{m^3/mole(N/m^2)^{1/7}}) \qquad (10)$$

g) Free volume (V_f)
Free volume was calculated by above equation.

$$\mathbf{V_f} = \left(\mathbf{M_{eff}u}/\mathbf{K\eta}\right)^{3/2} \qquad (\mathrm{m^3}) \qquad (11)$$

Where $M_{eff.}$ is the effective molecular weight of solution, which is expressed as $M_{eff} = \Sigma M = m_i x_i$ where, x and m are the mole fraction and molecular weight of the individual component in the mixture respectively. K is the temperature independent constant and its value is 4.28 x 10^9.

h) Gibb's free energy (ΔG)
Gibbs free energy canbe calculated from acoustic relaxation time (τ) as follows,

$$\mathbf{\Delta G} = \mathbf{RT\ ln}\left(\mathbf{kT\tau}/\mathbf{h}\right) \qquad (\mathrm{J\,mol^{-1}}) \qquad (12)$$

Where k is Boltzmann constant, T is the absolute temperature and h is the plank's constant.

i) Available volume (V_a)
The available volume is a direct measure of compactness in the liquid and the strength of attraction between the molecules of a liquid or a liquid mixture. It can be calculated from Schaaf's relation,

$$\mathbf{V_a} = \mathbf{V_m}\left(1 - \mathbf{u}/\mathbf{u_\infty}\right) \qquad (\mathrm{m^3}) \qquad (13)$$

The experimentally determined values of density (ρ), viscosity (η), ultrasonic velocity (U) for liquid mixture at 303.15 K are represented in Table 1.

Table 1: Experimental parameters (U, ρ and) for liquid mixture at 303.15 K.

Conc. of $ZnSO_4$ (mol Kg^{-1})	U (m s^{-1})	$\rho x10^{3}$ (Kg m^{-3})	Ƞx10^{-3} (Kg m^{-1} s^{-1})
	1% glucose solution		
0.00	1511.75	0.9991	0.8114
0.02	1514.3	1.0016	0.8181
0.04	1516.8	1.0062	0.8283
0.06	1517.88	1.0110	0.8353
0.08	1522.3	1.0133	0.8464
0.10	1524.48	1.0159	0.8549
	2% glucose solution		
0.00	1513.03	1.0036	0.8251
0.02	1516.3	1.0064	0.8328
0.04	1519.3	1.0105	0.8410
0.06	1522.3	1.0153	0.8518
0.08	1527.5	1.0174	0.8604
0.10	1531.3	1.0202	0.8696
	3% glucose solution		
0.00	1514.8	1.0070	0.8381
0.02	1518.66	1.0097	0.8460
0.04	1521.86	1.0143	0.8556
0.06	1526.83	1.0190	0.8665
0.08	1530.9	1.0201	0.8754
0.10	1537.3	1.0234	0.8843
	4% glucose solution		
0.00	1519.39	1.0107	0.8516
0.02	1520.66	1.0133	0.8590
0.04	1525.3	1.0177	0.8699
0.06	1530.5	1.0224	0.8811
0.08	1536.5	1.0240	0.8900
0.10	1542.3	1.0277	0.9005
	5% glucose solution		
0.00	1522.3	1.0143	0.8647
0.02	1525.3	1.0163	0.8738
0.04	1529.3	1.0214	0.8853
0.06	1535	1.0261	0.8965
0.08	1538.5	1.0277	0.9041
0.10	1545.8	1.0297	0.9154

In the present analysis viscosity, density and ultrasonic velocity increases with increasing concentrations of solute. The density increase with increasing concentration of solutes proposes a fair strong electrolyte nature of $ZnSO_4$ in which the Zn^{++} ions tends to attract the water molecules. The gradual increase in values of ultrasonic velocity and viscosity with different concentration of $ZnSO_4$ in various glucose solutions confirms the increase of cohesive forces because of strong interactions between solute and solvent molecules [13]. It is observed that the ultrasonic velocity (U) increase with increase in Zn^{++} ions concentrations in various glucose solutions. Molecular association is thus responsible for the observed change. This association is due to Hydrogen bonding between solute and solvent molecules.

Derived parameters such as Adiabatic compressibility (β_{ad}), inter molecular free length (L_f), acoustic impedance (Z), relaxation time (τ) for liquid mixture at 303.15K are represented in Table 2.

The value of adiabatic compressibility (β_{ad}) and intermolecular free length (Lf) decreases and these two parameters shows an opposite behaviour as compared to ultrasonic velocity (U). Experimentally it has been reported that U and L_f vary inversely of each other with change in composition of the mixture [14-16] as in the present system. β_{ad} values decreases with increase in solute concentration, Because in aqueous Zn^{++} ions solution, Zn^{++} ions have tendency to form a core compact structure with the solvent molecules through hydrogen bonding. The decrease in compressibility can be explained due to structural changes of molecules in the mixture resulting to an increase in ultrasonic velocity. With increase in concentration adiabatic compressibility goes on decreasing. A large fraction of water molecule exerts electrostatic force of attraction, which have affinity to attracts the neighbouring molecules results decrease in the effective volume of water.With this increase in ionic concentration electrostatic forces cause the structure to break and the solute surrounded water molecules are more compactly packed, results decrease in adiabatic compressibility (βad), this shows that solute-solute interactions are lesser than solute-solvent interactions. Thus this effect reduces the adiabatic compressibility with increase in the Zn^{++} ions concentration [17-19].

Table 2: Derived parameters (β_{ad}, L_f, Z andτ) for liquid mixture at 303.15K

Conc.of $ZnSO_4$ (mol Kg^{-1})	β_{ad} x 10^{-10} (m^2 N^{-1})	L_f x 10^{-11} (m)	Z x 10^6 (Kg m^{-2} s^{-1})	τ x 10^{-13} (s)
		1%Glucose solution		
0.00	4.3796	4.3425	1.5104	4.7263
0.02	4.3540	4.3297	1.5167	4.7432
0.04	4.3198	4.3127	1.5262	4.7589
0.06	4.2929	4.2992	1.5346	4.7692
0.08	4.2587	4.2821	1.5425	4.7940
0.10	4.2355	4.2704	1.5487	4.8157
		2%Glucose solution		
0.00	4.3526	4.3290	1.5185	4.7764
0.02	4.3216	4.3136	1.5260	4.7867
0.04	4.2873	4.2964	1.5352	4.7953
0.06	4.2500	4.2772	1.5456	4.8151
0.08	4.2127	4.2590	1.5540	4.8209
0.10	4.1802	4.2424	1.5622	4.8349
		3%Glucose solution		
0.00	4.3276	4.3166	1.5254	4.8236
0.02	4.2944	4.3000	1.5333	4.8320
0.04	4.2568	4.2811	1.5436	4.8439
0.06	4.2098	4.2574	1.5558	4.8506
0.08	4.1791	4.2380	1.5645	4.8612
0.10	4.1345	4.2192	1.5733	4.8625
		4%Glucose solution		
0.00	4.2859	4.2957	1.5356	4.8544
0.02	4.2676	4.2866	1.5409	4.8767
0.04	4.2232	4.2642	1.5524	4.8862
0.06	4.1755	4.2401	1.5648	4.8933
0.08	4.1364	4.2202	1.5734	4.8960
0.10	4.0908	4.1968	1.5850	4.8995
		5%Glucose solution		
0.00	4.2551	4.2803	1.5438	4.8939
0.02	4.2291	4.2672	1.5502	4.9151
0.04	4.1861	4.2454	1.5620	4.9290
0.06	4.1363	4.2201	1.5750	4.9320
0.08	4.1110	4.2072	1.5811	4.9434
0.10	4.0642	4.1832	1.5917	4.9480

The free length decreases with increasing solute concentration. The increase of ultrasonic velocity in a solution of different concentration results decrease of intermolecular free lengths as a result of mixing the component [20-22], this indicates a large interaction between solute and solvent molecules and suggests a structure promoting behaviour on the addition of solute.

The acoustic impedance (Z), relaxation time (τ) and Gibb's free energy (ΔG) increase with increase in concentration of solute. The behaviour of acoustic impedance becomes responsible for the propagation of ultrasonic waves, and this behaviour can be explained on the basis of lyophobic interaction between solute and solvent molecules. The relaxation time and Gibb's free energy, both these parameters shows the presence of molecular interaction by the addition of solute concentration.

The relaxation time which is in the order of 10^{-13}second is due to the structural relaxation process [23]. This suggested that these molecules get rearranged due to supportive process [15]. Due to hydrogen bonding between the molecules, unlike molecules approach close to each other and this causes increase in the values of Gibb's free energy.

Derived parameters Rao's constant (R_M) , Wada's constant (W) , free volume (V_f), gibb's free energy (ΔG) and available volume (V_a) for liquid mixture at 303.15K are represented in Table 3.

Table 3: Derived parameters (R_M , W,V_f, ΔG and V_a) for liquid mixture at 303.15K

Conc.of $ZnSO_4$ (mol Kg^{-1})	R_M x 10^{-4} ($m^{10/3}s^{-1/3}mol^{-1}$)	W ×10^{-3} {m^3/mole$(N/m^2)^{1/7}$}	V_f x 10^{-8} (m^3)	ΔG× 10^{-21} ($Jmol^{-1}$)	V_a x 10^{-7} (m^3)
		1%Glucose solution			
0.00	2.0865	3.9494	2.2233	4.5745	10.0273
0.02	2.0939	3.9644	2.2157	4.5895	9.7666
0.04	2.0969	3.9725	2.2022	4.6033	9.4902
0.06	2.0989	3.9788	2.1950	4.6123	9.3735
0.08	2.1133	4.0068	2.1877	4.6340	8.9212
0.10	2.1214	4.0235	2.1793	4.6529	8.7003
		2%Glucose solution			
0.00	2.0968	3.9712	2.2008	4.6186	9.9278
0.02	2.1039	3.9860	2.1954	4.6277	9.5802
0.04	2.1093	3.9980	2.1892	4.6352	9.2543
0.06	2.1115	4.0046	2.1707	4.6524	8.9138
0.08	2.1216	4.0242	2.1674	4.6575	8.3473
0.10	2.1294	4.0401	2.1592	4.6696	7.9324
		3%Glucose solution			
0.00	2.1098	3.9976	2.1837	4.6598	9.7822
0.02	2.1178	4.0138	2.1793	4.6671	9.3666
0.04	2.1214	4.0228	2.1677	4.6773	9.0070
0.06	2.1259	4.0334	2.1560	4.6831	8.4429
0.08	2. 1356	4.0524	2.1522	4.6923	8.0025
0.10	2.1456	4.0721	2.1485	4.6934	7.2854
		4%Glucose solution			
0.00	2.1239	4.0258	2.1715	4.6864	9.3078
0.02	2.1309	4.0404	2.1643	4.7047	9.1887
0.04	2.1357	4.0516	2.1516	4.7137	8.6622
0.06	2.1405	4.0627	2.1396	4.7198	8.0682
0.08	2.1522	4.0850	2.1385	4.7221	7.4021
0.10	2.1596	4.1004	2.1311	4.7251	6.7408
		5%Glucose solution			
0.00	2.1380	4.0542	2.1585	4.7203	9.0256
0.02	2.1468	4.0718	2.1493	4.7384	8.7073
0.04	2.1503	4.0807	2.1341	4.7502	8.2470
0.06	2.1555	4.0925	2.1241	4.7528	7.5911
0.08	2.1662	4.1133	2.1229	4.7624	7.2125
0.10	2.1780	4.1358	2.1170	4.7663	6.3807

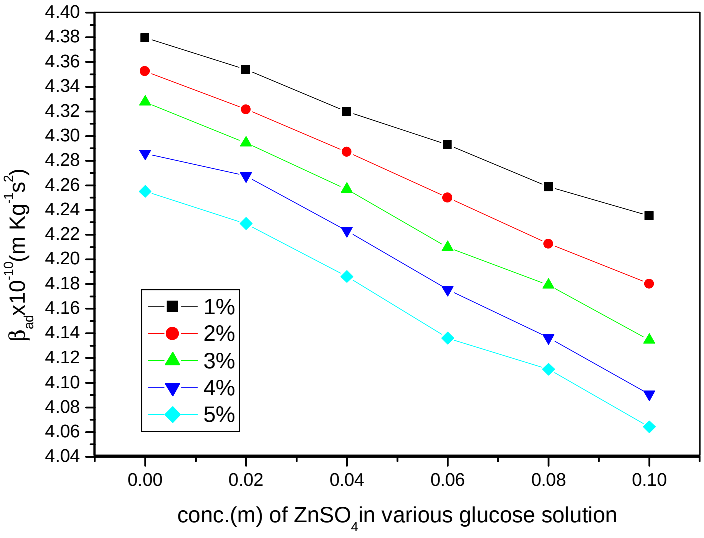

Fig.2. Plot of adiabatic compressibility versus different conc. (m) of $ZnSO_4$ in various glucose solutions at 303.15K. It is found that the adiabatic compressibility goes on decreasing with increase in the concentration of $ZnSO_4$ in different glucose solutions

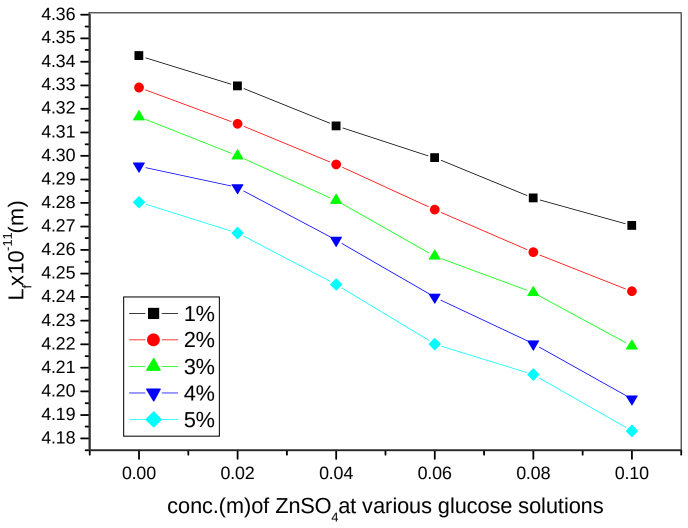

Fig.3. Plot of intermolecular free length versus different conc. (m) of $ZnSO_4$ in various glucose solutions at 303.15K. . It is found that intermolecular free length goes on decreasing with increase in the concentration of $ZnSO_4$ in different glucose solutions

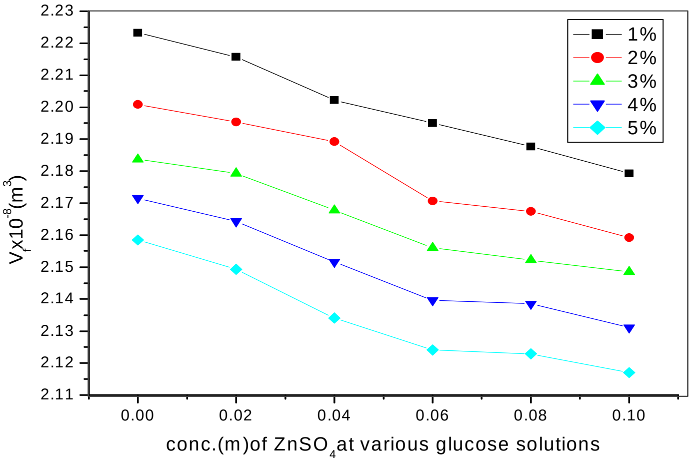

Fig.4. Plot of Free volume versus various conc.(m)of $ZnSO_4$ in various glucose solutions at 303.15K. It is found that free volume goes on decreasing with increase in the concentration of $ZnSO_4$ in different glucose solutions

The values of Rao's constant (R_M) or molar sound velocity and Wada's constant (W) or molar compressibility increases with concentration because of the presence of more number of components and thus leads to a close packing of the medium and thereby increase the interactions between solute and solvent.

The values of free volume (V_f) and Available volume (V_a) decreases with increase in concentration of solute. With the increase in solute concentration, the strength of interaction between the molecules increases, this results decrease in free volume.

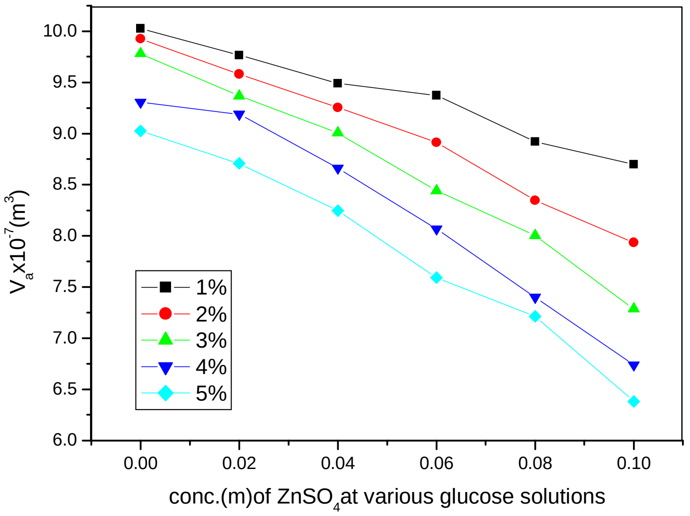

Fig.5. Plot of Available volume versus various conc.(m) of $ZnSO_4$ in various glucose solutions at 303.15 K. It is found that available volume goes on decreasing with increase in the concentration of $ZnSO_4$ in different glucose solutions

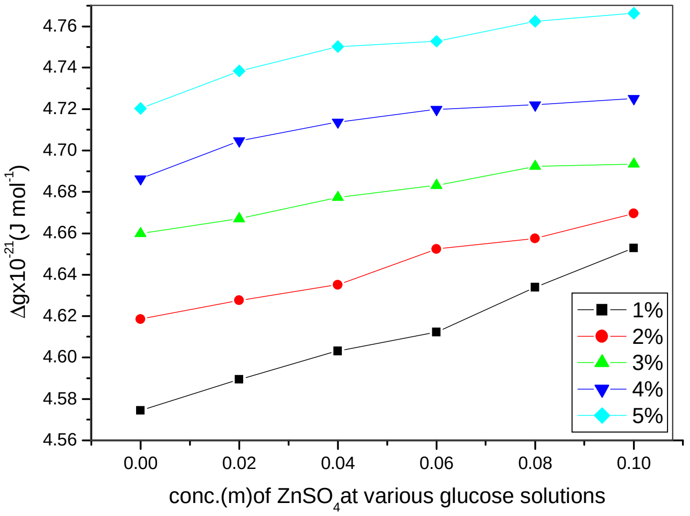

Fig.5. Plot of Gibb's free energy versus various conc.(m) of $ZnSO_4$ in various glucose solutions at 303.15K. It is found that Gibb's free energy goes on increases with increase in the concentration of $ZnSO_4$ in different glucose solutions

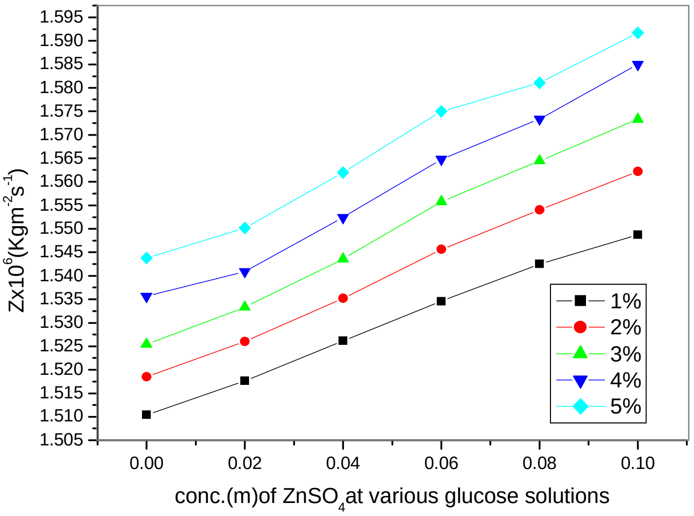

Fig.6. Plot of Acoustic impedance versus various conc.(m) of $ZnSO_4$ in various glucose solutions at 303.15K. It is found that acoustic impedance goes on increases with increase in the concentration of $ZnSO_4$ in different glucose solutions

CONCLUSION

Ultrasonic investigations of molecular interactions in different aqueous solutions of glucose containing various conc. of $ZnSO_4$ have been measured. The experimental parameters give the valuable information regarding ion-solvent interactions in aqueous solutions. Glucose used in aqueous system as solute, modify the structure of water molecules into more arranged structure due to the formation of hydrogen bonding. $ZnSO_4$ added as solute results breaking of hydrogen bonds of solvent system and thus produces free solvent molecules. This results in increase in the value of ultrasonic velocity and decrease in adiabatic compressibility with concentration which shows that solute-solute interactions are smaller than solute- solvent interactions. These structural changes in solutions are further conformed by decreasing values of intermolecular free length with concentration.

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