MOLECULAR INTERACTIONS AND EXCESS THERMODYNAMIC PROPERTIES OF MIXED SOLUTIONS OF ZINC SULPHATE AND ZINC NITRATE AT 303K BY ULTRASONIC METHOD

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ABSTRACT: The ultrasonic velocity, density and viscosity were measured in aqueous mixtures of zinc sulphate (Heptahydrate) and zinc nitrate (Hexahydrate) at 308 K. Acoustic parameters, such as specific acoustic impedance, free length, isotropic compressibility, molar compressibility, molar sound velocity, free volume, available volume, relaxation time, excess parameters and the deviation from the experimental values has been evaluated. The value of ultrasonic velocity decreases and comes to a minimum and then increases as the mole fraction of zinc nitrate increases in the mixed salt solution. The excess adiabatic compressibility of zinc sulphate and zinc nitrate mixed solution with water was positive at all concentrations, while it is negative at a mole fraction of 0.7945. Variation of these parameters with concentration indicates the complex formation between the constituents of mixture. The results were interpreted as per the intermolecular interactions.

Key Words: Ultrasonic velocity, compressibility, excess thermodynamic properties, zinc sulphate-zinc nitrate mixed solution.

INTRODUCTION

The ultrasonic method plays an important role in understanding the physic-co-chemical behavior of liquids. There has been an increasing interest in the study of molecular interactions between components of liquids mixtures (Ravichandran, S and Ramanathan.K, 2006; Ramanathan K and Ravichandran S, 2004; Tabhane A and Patki B.A., 1985). The spectroscopic method (Foster, 1969) has been used to study the molecular complexes formed between interacting molecules. Non-spectroscopic techniques have also been very widely used for such studies.

Molecular interactions give more details about the structure of the liquids. Madhu Roestogi et.al., (2002) correlate the velocity with structural changes as the concentration of CCl_4 is varied. The velocity gives information about the bonding between the molecule and formation of complexes at various temperatures through molecular interactions. The thermodynamic and transport properties of binary liquids have been studied by various workers. Acharya et.al., (2003) have measured the velocity and density from which L_f , was calculated in the binary liquid mixtures having DIBK (Di-isobutyl ketone) with various polar liquids at 303.16K and found that L_f varies with the concentration.

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Deshmukth et.al.,(2002) studied the molecular interaction between acrylophenone and its complexes with ligands and the results are interpreted in terms of solute-solvent and solute-solute interactions. Karunanidhi and Subramaniam (2002) have calculated the compressibility and impedance which explains the molecular interactions in the case of water + acetone and Butanol + acetone. Subramaniya Naidu and Ravindra Prasad (2002) have examined the free length and free volume to explain the strength of molecular interactions. Ragouramana (1999) have calculated the velocity and absorption studied in the solution of Benzoic acid, ohydroxybenzoic acid with dimethyl sulphoxide at 303.15K. This indicates that possibility of association between monomer solute and solvent molecule interaction. Viscosity (Yadava and Yadava, 1987., Gopal et al., 1978) dielectric constant (Yadava. and Yadava, 1978, Yadava. and Yadava 1979) and Refractive index measurements (Singh and Bhat 1982) have also been used to investigate molecular interactions between varieties of interacting molecules. In this work, the ultrasonic velocities and densities of binary mixture of zinc sulphate and zinc nitrate in water at 308 K have been measured in different concentrations.

MATERIALS AND METHODS

In the present investigation zinc sulphate (AR Grade) of molecular weight 287.54g/mol was taken and dissolved in one liter of distilled water to get 1N solution. Similarly, zinc nitrate (AR Grade) of molecular weight 297.47 g/mol dissolved in one liter of distilled water to get 1N solution. The above two solutions prepared separately and the solution is filtered. The above solutions are mixed in different ratio such as 90:10, 80:20 ...10:90 and the results are interpreted.

The velocity of the mixed salt solution of zinc sulphate and zinc nitrate at various concentrations is measured with the help of ultrasonic interferometer (Mittal enterprises-F-80 model) at a standard frequency of 2 MHz with an accuracy of ± 0.1 m/s. Density of the mixture is measured with the help of specific gravity bottle whose capacity is 25ml and the viscosity of the mixture can be measured accurately by "Oswald's viscometer" whose capacity is 10ml with a precision of ± 0.1 kgm⁻³ and 0.1% respectively. Distilled water has been used as a standard liquid with time flow been measured with an accuracy of 0.1 sec. The measurements have been made at room temperature of 303K.

RESULTS AND DISCUSSION

The ultrasonic velocity and other acoustical parameters are calculated by using as usual formula and the values are given in Table1 and 2. It is seen that, the ultrasonic velocity decreases with the increase of concentration of zinc nitrate. It attains a minimum at a mole fraction of 0.6928. The increase in the concentration of Zinc nitrate weakens the molecular forces and hence the abrupt change in velocity is obtained at a mole fraction of 0.7945. This non-linear variation of velocity with increase in concentration indicates the complex formation between the constituents of mixture (Ravichandran. and Ramanathan, 2010)

The two salts are dissolved in water to form $So_4^{-}Zn^{2+}$ and NO_3 ion. During the solvation in water, the cations are formed; because of these ions have great tendency to form complexes with several molecules or ions. The cations are relatively very small in size and have higher effective nuclear charge. Thus they have a high positive charge density which facilitates the acceptance of lone pairs of electrons from other molecules ions. Addition of zinc nitrate may loosen the cohesive forces holding the solvent which is the reason for reduction in ion solvent interaction resulting in lowering of internal pressure in aqueous solutions of Zn nitrate with increase in concentration. This may be mainly due to characteristics of nitrate ions in water. This fact is confirmed from Raman studies also. The variation of density with the concentration of zinc nitrate is shown in Table.1. It was inferred that as the concentration of nitrate increases the density creases up to the concentration of 30:70 at a mole fraction of 0.6928. But at the concentration of 20:80, the density shows a sharp increase which again confirms the structural rearrangement of molecules (Nikam et al., 2000).



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Table 1 Ultrasonic velocity(U), density(ρ), specific acoustic impedance(Z), adiabatic compressibility(β_{ad}), intermolecular free length(L_f), effective molecular weight(M_{eff}), internal pressure(Π_i) in the binary solution of zinc sulphate and zinc nitrate in different concentration at 303K.

Molefraction of (X ₁) ZnSo ₄	Molefraction of (X ₂) Zn(No ₃) ₂	U (m/s)	ρ (kg/m ³)	Z (kg/m²s)	$\beta_{ad} x 10^{-10}$ $(kg^{-1}ms^{2})$	L _f (Å)	ηx10 ⁻³ (Ns/m ²)	M _{eff} (g)	П _і 10 ⁻⁵ Ра
0.903	0.0970	1604	1176	1886304	3.305	0.3636	1.597	288.50	1.393
0.8054	0.1946	1600	1170	1872000	3.339	0.3655	1.529	289.47	1.362
0.7071	0.2929	1597	1168	1865296	3.357	0.3664	1.497	290.45	1.346
0.6081	0.3919	1595	1166	1859770	3.371	0.3672	1.427	291.43	1.311
0.5085	0.4915	1590	1164	1850760	3.398	0.3687	1.562	292.42	1.372
0.4082	0.5918	1585	1163	1843355	3.423	0.3700	1.603	293.42	1.391
0.3072	0.6928	1573	1161	1826253	3.481	0.3732	1.621	294.42	1.408
0.2055	0.7945	1581	1164.4	1840916	3.436	0.3707	1.631	295.43	1.397
0.1031	0.8969	1577	1159.3	1828216	3.468	0.3725	1.632	296.45	1.396

Table 2 Molar volume (V), molar compressibility(W), molar sound velocity (R), available volume(V_a), free volume(V_f), relaxation time(τ), classical absorption co-efficient(α/f^2)_{class}, surface tension(σ) and attenuation (α) constant in the binary solution of zinc sulphate and zinc nitrate in different concentration at 303K.

Molefraction of (X1) ZnSo4	Molefraction of (X ₂) Zn(No ₃) ₂	V (m³/mol)	R (m ³ /mole) (m/s) ^{1/3}	W (m ³ /mole) (Kg ⁻¹ ms ²)	V _a (m ³ /mole)	V _f (m³/mole)	τ 10 ⁻¹³ (s)	$(\alpha/f^2)_{class} \\ x10^{-15}$	σ (N/m)	α x10 ⁻¹⁵ (Nep/ m)
0.903	0.0970	0.2453	2.871	5.547	0.2459	0.0176	7.350	8.652	47605.2	2.754
0.8054	0.1946	0.2475	2.895	5.589	0.2475	0.0188	6.807	8.390	47154.2	2.672
0.7071	0.2929	0.2487	2.907	5.612	0.2482	0.0195	6.701	8.274	46952.9	2.635
0.6081	0.3919	0.2499	2.920	5.636	0.2491	0.0210	6.414	7.030	46796.9	2.525
0.5085	0.4915	0.2511	2.931	5.657	0.2496	0.0184	7.074	8.773	46509.2	2.794
0.4082	0.5918	0.2524	2.943	5.679	0.2500	0.0176	7.316	9.100	46235.5	2.898
0.3072	0.6928	0.2536	2.949	5.693	0.2493	0.0173	7.522	9.429	45632.4	3.003
0.2055	0.7945	0.2537	2.956	5.706	0.2507	0.0173	7.472	9.319	46115.2	2.968
0.1031	0.8969	0.2557	2.976	5.743	0.2520	0.0173	7.547	9.437	45478.7	3.006

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Acoustic impedance is almost reciprocal of adiabatic compressibility. Compressibility decreases with the increase of concentration, whereas the acoustic impedance increases. In this case, the acoustic impedance decreases with increase of concentration of Zinc Nitrate; whereas compressibility increases till the mole fraction of 0.6928. The sudden increase in impedance at a mole fraction of 0.7945 of zinc nitrate may be due to the complex formation in the solution and this may be on the basis of the interation between solute and solvent complex (Ishwara Bhat. and Shree Varaprasad, 2003). The variation of impedance with the concentration of nitrate is inferred that dips are formed which again showing the same result as that of the velocity ,thus indicating the formation of complex which may be due to the solute-solvent interactions as per the results interpreted earlier (Karunanithi, Subramanian, Aruna.1999).

The variation of adiabatic compressibility with the concentration of nitrate is given in Table1. Compressibility is inversely proportional to the velocity. It is primarily the compressibility that changes with structure. This leads to a change in ultrasonic velocity. The greater the attractive forces among the molecules of a liquid, the smaller will be compressibility. It shows the reverse effect as that of impedance which produces the peaks which again confirms the formation of complex molecule which is due to the ion-solvent interactions (Varma and Surendrakumar, 2000). Intermolecular free length is the distance between the surfaces of the neighboring molecule. From the observed value, it is seen that as the concentration of zinc nitrate increases L_f showing the same effect as that of compressibility, reaches its maximum at the concentration of 0.6928 and then decreases which indicates that there is a significant interactions present in between the molecules due to the dipole induced dipole or dipole-dipole interaction leads structural rearrangement (Ravichandran and Ramanathan, 2010; Govindappa.et al 1990) as described earlier. Surface tension is directly proportional to the density and velocity it shows the result as that of velocity, conforming at the mole fraction of 0.7945 the complex formed has high surface tension between SO_4^{2-} & $(NO_3)^{2-}$ due to the presence of water molecules (Govindappa et al 1990). The viscosity decreases with the increase in the concentration of nitrate and reaches its minimum at mole a fraction of 0.3919 and increases on further increasing the concentration of nitrate, which is due to the less cohesive force between them (Kannappan and Hahi, 2002). Maxima (Sri Devi et. Al, 2004) in the viscosity of a binary mixture may be due to the formation of intermolecular association.

Table1 shows the variation of the internal pressure with the increase in the concentration of Zn nitrate. The primary effect of dissolving Zn nitrate lowers the compressibility of the solvent molecules. The lowering of compressibility results in the increase of ultrasonic velocity and hence Π_i increases with the concentration of zinc nitrate. The internal pressure decreases up to the mole fraction of 0.3919 and then increases reaching the maximum at 0.6928 and again decreases showing the complex formed has high internal pressure (Tabhane et al 1999; Kannappan and Rajendran 1992) and the interaction is strong as described earlier. Attenuation value decreases up to the concentration of 0.3919 and again increases reaching the maximum at the mole fraction of 0.6929. It decreases showing the same result as that of viscosity confirming that the complex formed has high attenuation value (Muraliji et. al., 2004) as said earlier.

Free volume represents the average volume in which the centre of the molecules can move inside the cell due to the repulsion of surrounding molecule. As this parameter is inversely proportional to the viscosity, it shows that at the mole fraction of 0.3919 this parameter shows its maximum value (Tabhane et.al, 1999) and on again increasing the concentration its value seems to be decreasing which shows that cohesive force varies with the changes in the solute concentration described earlier ²⁷. It also describes in detail about the extent of complication in the mixture at the mole fraction of 0.3919. Acoustic relaxation time depends upon viscosity and adiabatic compressibility. It reaches its minimum at the mole fraction of 0.3019 and then increases with the concentration of zinc nitrate (Muraliji et. al., 2004) and shows a steep increase as the concentration of nitrate increases.

EXCESS PARAMETERS

The values of the β_{ad}^{E} of a mixed salts solution is shown in the Table 3. Its value at first shows some fluctuations and reaches it's maximum at the mole fraction of 0.6928 and shows a steep decrease at the concentration of 0.7945, attains a negative value indicating that the complex formed has strong interactions between them (Madhu Roestogi et al 2002) as interpreted earlier.

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Table-3 Computed values of excess compressibility (β^E), excess free length (L_f^E), excess viscosity (η^E) and excess surface tension (η^E) in the binary solution of zinc sulphate and zinc nitrate in different concentration at 303K.

Mole Fraction of X1(ZnSo4)	Mole fraction of X ₂ Zn(No ₃) ₂)	β ^E x10 ⁻¹² (kg ⁻¹ ms ²)	L ^E 10 ⁻⁵ (Å)	η ^E x10 ⁻³ (Ns/m ²)	σ ^ε 10 ⁴ (N/m)
0.903	0.0970	1.496	2.459	1.8846	4.7998
0.8054	0.1946	2.898	5.00	1.8081	4.7709
0.7071	0.2929	2.456	4.266	1.7310	4.7417
0.6081	0.3919	1.590	2.830	1.6534	4.7123
0.5085	0.4915	1.981	3.523	1.5754	4.6829
0.4082	0.5918	2.260	4.011	1.4967	4.6532
0.3072	0.6928	5.892	0.102	1.4176	4.6233
0.2055	0.7945	-0.891	-1.390	1.3379	4.5932
0.1031	0.8969	0.089	0.038	1.2576	4.5629

The variation of L_f^E of zinc nitrate and zinc sulphate solution is shown in Table 3. It attains both positive and negative values showing that the interaction is varying from weak to strong .Excess free length increases with the increase in concentration of zinc nitrate, then decreases to a minimum of -1.390. The excess values of compressibility and free length are negative at the particular concentration. The reduction incompressibility and free length as well as the negative excess values clearly indicate the existence of molecular interactions. The observed β_{ad}^E and L_f^E values which are negative at a mole fraction of 0.7945 of Zn nitrate, such that the effect of interaction due to formation of hydrogen bonded complexes between unlike molecules on compressibility dominates the physical forces which contribute to negative β_{ad}^E and L_f^E values. Similar negative excess compressibility and excess free length values are reported by Ramamurthy and Sastry (Ramamoorthy and Sastry1985; Eswari Bai et al., 2004). The negative excess free length indicates that the sound wave needs to cover a larger distance. This may be attributed to dominant nature of interactions between unlike molecules. Srivastava and Dubey (1985) also arrived at a similar conclusion on the basis of excess values of compressibility. This view supports the present investigation.

These parameters shows the non-linear variation with the concentration of zinc nitrate, and all the values are positive indicating the weak interactions exist between them (Kannappan & Hahi, 2002) Where its value varies from 4.8 to 4.563 in the linear manner and there has been no notable variation in between the molecules. According to Fort and Fort and Moore (1965) stated that the excess viscosity tends to become more positive as the strength of interaction increases. The excess viscosity with the mole fraction of zinc nitrate is shown in the Table.4. The excess viscosity variation gives a qualitative estimation of the strength of intermolecular interactions. The excess viscosities may be generally explained by considering the following factors.

1. The difference in size and shape of the component molecules cause the loss of dipolar association in pure component may contribute to a decrease in velocity and

2. Specific interactions between unlike components such as hydrogen bond formation and charge transfer complexes may cause for increase in viscosity in mixtures than the pure components.

The former effect produces negative deviation in excess viscosity, and latter effect produce positive deviation in excess viscosity. The excess viscosity is generally considered as a result of the above two major effects. In the present study, it can be observed that, the excess viscosity values are positive. It is maximum at the lower concentration, and it decreases with the increase of mole fraction of zinc nitrate.



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These parameters indicate that the weak interactions exist between them (Kannappan & Hahi, 2002) where its value varies from 1.885 to 1.258 in the linear manner and there is no notable variation in between the molecules.

CALCULATION OF THEORETICAL VELOCITY AND ITS DEVIATION

The theoretical evaluation of sound velocity in liquid mixtures is of considerable interest. The theoretical evaluation of sound velocity based on molecular models in liquid mixtures has been used to correlate with the experimental findings (Table 4). The comparison of theoretical and experimental results also provides better understanding about the validity of the various thermodynamics, empirical and statistical theories. The various theories, which are used to calculate the ultrasonic velocity in the multicomponent mixture, are free length theory, Nomoto's relation and Impedance dependent relation.

Mole fraction	Mole fraction of	U _{exp} (m/s)	U _{FLT} (m/s)	U _{Nomato} (m/s)	U _{IDR} (m/s)	Percentage of Deviation (%)		
of(X ₁) (ZnSo ₄)	$\begin{array}{c} (X_2) \\ (Zn(No_3)_2) \end{array}$					FLT	Nomato	IDR
0.903	0.0970	1604	1603.51	1603.99	1604.0	0.0306	0.0006	0
0.8054	0.1946	1600	1599.95	1599.98	1600.0	0.0031	0.0013	0
0.7071	0.2929	1597	1596.98	1597.00	1597.0	0.0013	0	0
0.6081	0.3919	1595	1594.97	1595.00	1595.0	0.0019	0	0
0.5085	0.4915	1590	1589.98	1590.00	1590.0	0.0013	0	0
0.4082	0.5918	1585	1585.30	1585.29	1585.3	0	0.0006	0
0.3072	0.6928	1573	1573.00	1573.00	1573.0	0	0	0
0.2055	0.7945	1581	1581.00	1580.99	1581.0	0	0.0006	0
0.1031	0.8969	1577	1576.99	1577.00	1577.0	0.0006	0	0

Table-4 Theoretical velocities and percentage of deviation in the binary solution of zinc sulphate and zinc nitrate in different concentration 303K.

FREE LENGTH THEORY

Jabobson (1952) introduced the concept of determination of ultrasonic velocity in pure liquids and liquid mixtures known as free length theory. Further he related the ultrasonic velocity in pure liquid to the free length L_f by the relation,

$$UL_{f} \rho^{1/2} = K$$
(1)

Where k is temp dependent Jacobson's constant.

In the case of liquid mixtures, the equation (1) can be written as

$$U_{FLT} = K \setminus L_{fmix} \rho^{1/2} \cdots$$

Where L_{fmix} and ρ are the free length and density of the mixtures.

NOMOTO'S RELATION

Assuming the linear dependence of the molar sound velocity (R) on concentration in mole fraction (X1 and X2) and the additively of molar value V_{m} , Nomoto's (1958) established an empirical formula for ultrasonic velocity in binary liquid mixtures as

 $U_{\text{Nomoto}} = (x_1 R_1 + x_2 R_2 / x_1 V m_1 + x_2 V m_2)^3 \dots \dots \dots \dots (3)$

(2)

In case of multicomponent mixtures, the above equation (3) can be written as

 $U_{\text{Nomoto}} = \sum x_i R_i / \sum x_i V m_i \text{ where } i=1 \text{ to } n....(4)$

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IMPEDANCE DEPENDENT RELATION

Ultrasonic velocity in pre liquids and liquid mixtures depends on impedance known as impedance dependent relation (1958) given by the equation

 $U_{IDR} = \sum x_i Z_i / \sum x_i d_i$ where i=1 to n

Where x_i is the mole fraction and Z_i is the acoustical impedance of the solution. From the Table 3, the percentage of deviation from the theoretical velocity is calculated by use of various theories such as free length theory (FLT), Nomato's theory and impedance dependant relation (IDR). In the case of free length theory the values ranges from 0.00064 to 0.03055. In the Nomato's theory the values varies from 0.00062 to 0.00125, however in the case of Impedance dependent relation the percentage of deviation is "0". Thus it is seen that the IDR theory is in close agreement with that of its theoretical velocity (Arumugam et al., 1998) when compared to other theories.

DISCUSSION

The ultrasonic velocity of the solution derived from zinc sulphate and zinc nitrate decreases with increasing the mole fraction of zinc nitrate up to 0.6928. Since, zinc sulphate can furnish only 2 ions whereas zinc nitrate can furnish 3 ions. Ultrasonic velocity is expected to be largely influenced by the addition of zinc nitrate. Each ion, either Zn^{++} , SO_4^{--} or NO^{3-} can better organize solvents around them. The ultrasonic velocity is to decrease with increase in the concentration of any ionic spaces. At the mole fraction of zinc nitrate 0.7945, there is a increase in velocity. It illustrates, organization of zinc nitrate themselves rejecting more water of hydration. But at a mole fraction of 0.8969, there is a decrease in velocity. Hence, at this mole fraction, the organized ions at 0.7945 must segregate. So this study illustrates that the complete dissolution of zinc nitrate up to the mole fraction of 0.6928. But association of ions begins at mole fraction of 0.7945. Suppression of association begins at mole fraction of 0.8969. This suppression could be possible by charging individual neutral ionic clusters.

The density of mixture also decreases with increasing the mole fraction of zinc nitrate up to 0.6928. So this clearly supports complete dissolution of zinc nitrate, thus weakening the force between free water molecules by adding zinc nitrate. At the mole fraction of 0.7945, since there is a association of ions partly rejecting water at hydration, the density increases. In other words, they release the water and enhance the hydrogen bonding interaction. The density of the solution is decreased at the mole fraction of 0.8969, since the association is suppressed by charging neutral clusters. The water is to be taken for salvation. Hence, there may be weak hydrogen bonding interaction in the water thus reducing its density. The data and free length also supports the above view. With increase of mole fraction of zinc nitrate, there is decreasing the hydrogen bonding interaction in water. Hence, the free length is increases up to the mole fraction of 0.6928 of zinc nitrate but at 0.7945, water is released, so there is more hydrogen bonding interaction. This results in decrease in free length. The viscosity of the solution increases with increase in the mole fraction of zinc nitrate up to 0.6928. Since, water is being used more and more for hydration. The free water has reduced hydrogen bonding interaction. Hence, a decrease in viscosity is absorbed.

CONCLUSION

The ultrasonic study of the aqueous solution of Zinc sulphate and Zinc nitrate confirmed the presence of strong ion-dipole interactions. The present study of 1N aqueous zinc sulphate with zinc nitrate mixed salt solution indicates the process of ion-association and formation of a specific complex formation at X_{ZnSo4} : $X_{Zn(NO3)}$ ratio of 20:80. The present study reveals interesting structural instability in their solution state. Further, study may give more details about complex ion formation.

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