STUDY OF TRANSITION METAL-DRUGS COMPLEXES IN MIXED SOLVENT BY P^H – METRIC METHOD.

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ABSTRACT
The interaction of transition metal ions with substituted heterocyclic drug Clarithromycin has been investigated in 70% ethanol-water mixture at 0.1 M ionic strength and 303 K by potentiometric titration. The data obtained is used to estimate the values of proton-ligand stability constant (pK) and metal-ligand stability constant (log K). It is observed that the metal ions form 1:1 and 1:2 complexes with substituted heterocyclic drug.
Keyword: Stability constant, substituted heterocyclic drug.

INTRODUCTION

The studies of metal-ligand complexes in solution of a number of metal ions with carboxylic acids, oximes, phenols etc. would be interesting which throw light on the mode of storage and transport f metal ion in the biological kingdom. The selected substituted heterocyclic drug Clarithromycin is an antibiotic and used to treat various bacterial infections[1]. Numerous researchers [2-5] have studied the formation and stability constant of metal complexes with transition metals. Pandhare [6] have been investigated the study of stability constant of transition metal complexes with 7-Hydroxy Flavones at 30°C in 50 % (V/V) dioxane - water medium and 0.1 M ionic strength. Azharuddin [7] has been investigated the stability constants of binary complexes of some rare earth and nuclear metal ions with some peptides at 0.1 M ionic strength pH-metrically. It is observed that there is formation of 1:1 and 1:2 complexes between metal ions and ligand. Pawar [8] has been studied the determination of stability constants of the complexes of some peptides with d block metal ions in ethanol-water mixtures at 300 K at 0.1 M constant ionic strength pH-metrically. Ebru Mavloglu et al have studied the stability constant of Ni (II), Co (II) ,Cu (II) and Zn (II) complexes of hypoxynthine at physiological condition by potentiometric method [9]. Vyas et. al. have determined the stability constant of binary complexes of d_{10} metal ions Cu (II), Ni (II), Co (II) and Mn (II) with substituted derivatives of coumarin by using Irving –Rossoti method at constant temperature 303 K and 0.1 M ionic strength [10]. After review of literature survey the detail study of complex under identical set of experimental condition is still lacking. It was thought of interest to study the chelating properties of substituted

heterocyclic drugs under suitable condition with lanthanide by pH metrically.

EXPERIMENTAL

Determination of stability constants:

The pH measurements were carried out with equip-tronic EQ-610 pH meter (accuracy ± 0.01 units) using combine glass electrode at 303 K. Pure transition metal nitrates and sulphates (99.9% Pure) were used. All metal nitrates and sulphates available from Sigma Aldrich Chem. Co., U.S.A. Metal solution was prepared in triply distill water and concentration was estimated by standard method. The solution of drug was prepared in solvent .The pH metric reading in 70% ethanol-water mixture were converted to $[H^+]$ value by applying the correction proposed by Van Uitert Haas [11]. The ethanol was purified by the method described by Vogel [12]. The overall ionic strength of solution was constant maintains by adding $NaClO_4$. All the solutions were titrated with standard carbonate free NaOH (0.2 N) solution at constant ionic strength (0.1 M). The titration was carried out in double wall glass jacketed titration cell connect to the constant temperature circulating bath. The temperature of reaction cell is constant by circulating water from Thermostat (0.1^0C). The experimental procedure involved pH metric titrations of solutions of –

1. Free HClO4 (A)
2. Free HClO4 + Ligand (A+L)
3. Free HClO4 + Ligand +Metal ion (A+L+M)

Data obtained from each titration is plotted as pH Vs volume of NaOH added and corresponding volume at successive pH for each set is determined and calculated.

RESULTS AND DISCUSSION

Proton-ligand stability constant (pK)

Substituted heterocyclic drugs may be ionized as acid having replaceable H^+ ion from $^-$OH group. Therefore it is represented as HL,

$$HL \longrightarrow H^+ + L^-$$

he titration data used to construct the curves between volume of NaOH and P^H. They are called acid-ligand titration curves. It is observed from titration curves for all systems ligand start deviating from the free acid curves at P^H =3.5 and deviating continuously up to P^H =12.0. The deviation shows that dissociation of proton in substituted drugs.

The average number of proton associated with the ligand (n_A) was determined from free acid (A) and acid - ligand (A+L) titration curves employing the equation of Irving and Rossotti [13].

$$\overline{n_A} = \gamma - \frac{(V_2 - V_1)(E^0 + N)}{(V_0 + V_1) T^0_L}$$

Where,

V_0 = Initial volume of solution

V_1 =Volume of alkali required during free acid titration.

V_2 = Volume of alkali required during free acid + ligand titration.

N = Normality of sodium hydroxide.

E^0 = Concentration of mineral acid ($HClO_4$)

γ = Number of replaceable hydrogen ion.

T^0_L = Concentration of ligand

The P^K values were determined from formation curves (n_A V/s P^H) by noting the P^H at which n_A=0.5 (half integral method). The accurate values of pK were calculated by point wise calculations which are presented in table-1.

Table-1: Determination of proton-ligand stability constant (pK) of substituted heterocyclic drugs at 0.1 M ionic strength.

System	pK Values	
Clarithromycin	Half integral	Point wise calculation
	8.55	8.95

Metal-ligand stability constant (Log k)

Metal-ligand stability constant of metal chelates with some substituted heterocyclic drug were determined by employing Bjerrum Calvin P^H metric titration method as adopted by Irving and Rossotti. The values of metal ligand stability constant (log K_1 and log K_2) were determined from formation curves (n V/s P^H) by using half integral method. The accurate values were calculated by point wise calculations which are presented in table-2.

Table-2: Determination of metal-ligand stability constant (log K) metal ion with Clarithromycin at 0.1 M ionic strength.

System	Log k_1	Log k_2	Log k_1 – Log k_2	Log k_1 / Log k_2
Ni (II) -Ligand	6.90	4.35	2.55	1.5862
Cu (II) -Ligand	6.85	4.54	2.31	1.5088
Zn (II)-Ligand	6.60	4.28	2.32	1.5420
Fe (II) -Ligand	6.75	4.15	2.60	1.6265
Mg (II) -Ligand	6.60	4.15	2.45	1.5903

CONCLUSION

The result shows the ratio of Log k_1 / Log k_2 is positive and greater than one in all cases. The higher value of ratio (Log k_1 / Log k_2) indicates more stable stepwise complex formation takes place. This implies that there is little or no steric hindrance to the addition of secondary ligand molecule. The difference between log K_1 and log K_2 was sufficiently large indicates the stepwise formation of 1:1 and 1:2 complexes.

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STUDIES THE INFLUENCE OF DIELECTRIC CONSTANT OF MEDIUM ON STABILITY CONSTANT OF L SERINE AND ITS COMPLEXES WITH Pr (III) ION.

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Abstract:

The solution studies of binary complex of Serine with Pr (III) ion have been performed. The protonation constant of Serine and the stability constant of complexes with Pr (III) ion at 303K were investigated. The ionic strength is constant maintained 0.01M by adding $NaClO_4$. The pK and log K values of the complexes were determined in 10% to 50% ethanol –Water medium by the pH metric method. The pK and log K values of the complexes increased as ethanol content increased.

Key word: Dielectric constant, Protonation constant, Stability constant.

Introduction:-

Amino acids are common components of all organisms. Protein of all species made from the amino acids. Protein plays many different biological roles in living systems. Serine compound is one of the naturally occurring proteinogenic amino acids. Serine was first obtained from silk protein, a particularly rich source, in 1865 by Emil Cramer [1]. Serine is important in metabolism in that it participates in the biosynthesis of purines and pyrimidines. Serine plays an important role in the catalytic function of many enzymes.

The lanthanide compounds have a remarkable importance in daily life[2-3].The influence of dielectric constant on complex equilibrium between substituted pyrazilines and lanthanide metal ions in dioxane-water system have been studied[4]. The stability constant of lanthanides and Iminodiacetic acid in water and dioxane-water mixture have been studied by potentiometric method at constant ionic strength[5]. The dissociation constant of Succenic acid, Azelic acid, Acetic acid in methanol-water, Ethanol-water, dioxane-water system at different dielectric constant have been studied[6]. The stability constant of 4-Amino, 3-Napthol Sulphonic acid with Cu(II) metal ions in different dielectric constant of medium using ethanol-water system by pH metry was studied[7]. Reddy [8] et.al. have showed the effect of dielectric constant on the stability constant of Co(II) and Ni(II) with two mercaptoethyl benzimidazole, glycene, ehtelene diamine and salycilic acid in various properties of ethanol-water mixtures at 30^0C and ionic strength is 0.1M. The present paper describes the complexation of Pr(III) with Serine in 0.01M ionic strength. The present work

includes the determination of pK and logK values at various percentage of ethanol-water medium.

Experimental:-

The pH measurements were carried out with equip-tronic EQ-610 pH meter (accuracy ± 0.01 units) using combine glass electrode at 208 K. Pure rare earth nitrate (99.9% Pure) was used. Metal nitrate available from Sigma Aldrich Chem. Co., U.S.A. Metal nitrate was prepared in triply distilled water and concentration was estimated by standard method. All chemical reagent used in this work are A.R. grade. The solution of Serine was prepared in aqueous ethanol. The pH metric readings in different percentage (10%, 20%, 3%, 40%, 50%) of ethanol–water mixture were converted to [H^+] value by applying the correction proposed by Van Uitert Haas. The overall ionic strength of solution was constant maintained by adding $NaClO_4$. All the solutions were titrated with standard carbonate free NaOH (0.2N) solution at constant ionic strength. The titration was carried out in different percentage (10%,20%,3%,40%,50%) of ethanol-water medium. The values of dielectric constant of different percentage of ethanol-water taken form literature [9].

Data obtained from each titration was plotted as pH Vs volume of NaOH added and corresponding volume at successive pH for each set was determined and calculated.

Result and discussion:-

The proton-ligand and meta-ligand stability constant of cortisone and its complexes with Pr(III) metal ions are given in table -1. It could be seen that pK and logK values increased with increase in the percentage of ethanol with decrease in dielectric constant of medium, this is due to the interaction between lanthanide ions and ligand is mainly electrostatic, as well as because of the effect of bulk solvent. The lowering of dielectric constant would increase the electrostatic force of attraction between metal ion and negatively charged ligand to form complex. The values of pK calculated by point wise calculation method. The metal-ligand stability constants were determined by half integral method. Higher values of $logK_1$ and $log K_2$ showed that ligands are stronger chelating agents. The dielectric constant of medium strongly affected on proton–ligand and metal–ligand stability constants because of the fact that at least one of the constituent is charged and other is either charged or has dipole moment. The plots of $logK_1$ and $logK_2$ Vs 1/D for the entire system exhibit a linear relationship.

Table-1 pK and Log K values of Serine and its complexes with Pr (III) in different percentage of Ethanol-water mixtures at 0.01 ionic strength.

% Ethanol	Dieletric constant (D)	1/D	Mole-fraction	Serine	Serine + Pr (III)	
				pK	$LogK_2$	$LogK_1$
10	73.95	0.01352	0.0228	4.8452	4.88	3.07
20	69.05	0.01448	0.0499	5.2967	5.46	3.32
30	63.85	0.01566	0.0825	5.7852	5.79	3.45
40	58.36	0.01714	0.1227	5.9937	6.56	3.60
50	52.62	0.01900	0.1735	6.4129	6.77	4.06

Conclusion:

The proton-ligand and metal-ligand stability constant of complexes increased with increased percentage of ethanol. As the values of dielectric constant of medium decreases, the values of proton-ligand and metal-ligand stability constant of complexes increases. The dielectric constant of medium decreased, as expected from the electrostatic nature of interaction between metal ions and ligands.

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INVESTIGATION OF STABILITY CONSTANT OF COMPLEX OF A NEW SYNTHESIZED SCHIFF BASE LIGANDS OF [5-HYDROXY 3-METHY 1-(2,4-DINITROPHENYL) PYRAZOL 4-YL] (PHENYL) METHANONE AND 4-AMINO ANTIPYRINE SPECTROPHOTOMETRICALLY

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ABSTRACT:

Using the Job's method of continuous variation the stability constant of complex of a new synthesized Schiff base ligand is investigated in 70% dioxane-water mixture maintaining ionic strength constant (0.1 M). The Schiff base was synthesized from [5-hydroxy 3-methy 1-(2,4-dinitrophenyl) pyrazol 4-yl] (phenyl) methanone and 4-Amino Antipyrine. The Formation 1:1 and 1:2 complex is confirmed using isobestic point method spectrophotometrically. Further Comparison of present method and pH-metric method are carried. The results obtained of stability constant are in good agreement.

Keywords: 5-hydroxy 3-methy 1-(2,4-dinitrophenyl) pyrazol 4-yl] (phenyl) methanone, Spectrophotometry, Formation constant, Schiff base.

INTRODUCTION

Interferometry technique is one of the most significant technique to understand the solute-solvent, ion-solvent interaction property of compound in aqueous and non-aqueous medium[1-4]. Meshram et. al. investigate for some substituted Pyrazolines different acoustical properties in binary mixture acetone-water and observed variation of ultrasonic velocity with concentration[5]. Palani have examined the measurement of ultrasonic velocity and density of amino acid in aqueous magnesium acetate at constant temp[6]. By considering size of ion and polarity of solvent ion-ion interaction can be determine and the strength of this interaction is directly proportional to the size of the ions and magnitude of dipole but inversely proportional to the distance between ion and molecules. Voleisines et al. has been studied the structural properties of solution of lanthanide salt by measuring ultrasonic velocity[7]. Tadkalkar et.al. have studied the acoustical properties and thermodynamic properties of citric acid in water at different temperature[8].

After review of literature we observed that there is no study under identical set of experimental condition for Schiff base of Pyrazolone with 4-amino antipyrine. It attract our interest toward the Schiff base of (5-hydroxy -3-methyl-1-(2,4dinitrophenyl)-pyrazol-4-yl)(phenyl)methanone and 4-amino antipyrine acoustical properties study under suitable condition.

EXPERIMENTAL

The Schiff base of (5-hydroxy-3-methyl-1-(2,4dinitrophenyl)-pyrazol-4yl)(phenyl) methanone and 4-amino antipyrine is synthesized by known Technique according to literature process and it will be purified by Vogel's standard method[9]. This novel synthesized Schiff base is used for present study. Solution Schiff Base is prepared in the double distilled Dioxane solvent. The solution of different concentration of Schiff base is prepared. The specific gravity bottle use to determine the densities by relative measurement method with accuracy ± 1×10^{-5} gm/cm^3. The ultrasonic velocities were measured by using ultrasonic interferometer having frequency 3MHz. By circulating water through the double wall measuring cell, made up of steel constant temperature was maintained.

In the present analysis, different acoustic properties such as intermolecular free length (Lf), adiabatic compressibility (bs), apparent molal volume (fv), apparent molal compressibility (fk),relative association (RA), specific acoustic impedance (Z), limiting apparent molal compressibility (f^{0}k), limiting apparent molal volume(f^{0}v), solvation number(Sn) and their constant (Sk, Sv) have been evaluated

RESULTS AND DISCUSSION

In the present study, the acoustical properties are recorded (table-1, table-2 and table-3). From the data obtained we can say that as system concentration increase Intermolecular free length(Lf) decrease and it is observe that ultrasonic velocity are increases. The value of specific acoustic impedance property (Z) increased when there is increase in concentration of Schiff base in dioxane-water system. The value of adiabatic compressibility is decreases with increase in solution concentration it show that there is solute-solvent interaction present. This indicates that there is strong interaction between solvent and solute molecule in solution. The strong interaction between solvent and solute molecule was observed due to increase in the apparent molal volume also increases. It was initiated that decrease in the value of apparent molal compressibility with the increase in concentration of Schiff base; it indicate that weak electrostatic attraction force among the close vicinities of ions.

When the concentration of solution increases there was a decrease in solvation number are detected; it specifies that there are strong coordination bond forms through solvent molecule in primary layer. The value of Sk exhibits negative it indicates the presence of weak ion-ion interactions in Schiff base of benzoyl Pyrazolone system. From table-3, It was detected that the sign of limiting apparent molal volume is positive it indicates that the ion-dipolar interaction between solvent and Schiff base derivative of benzoyl Pyrazolone. Altogether the sign of Sv are positive, directed toward the strong interaction between dioxane and solute molecule. The value of Sk, Sv has been govern from fig. 1 and 2.

Table-1 Ultrasonic velocity, density, adiabatic compressibility ($\square_S$), Specific acoustic impedance (Z) Intermolecular free length (L$_f$).

Concentration moles lit^{-1} (m)	Density (ds)	Ultrasonic velocity	Adiabatic compressibiliy	Intermolecular free length	Specific acoustic

	kg m^{-3}	(Us) m s^{-1}	(β_S) x10^{-10} m^{2}N^{-1}	(L$_f$) x10^{-11} m	impedance (Zx10^6) kg m^{-2} s^{-1}
Schiff base of benzoyl pyrazolone + 10% 1,4 Dioxane					
1x10^{-3}	1019.85	1491	4.41071	4.2239	1.5206
2x10^{-3}	1019.94	1493	4.39851	4.2180	1.5228
3x10^{-3}	1020.02	1496	4.38054	4.2094	1.5259
4x10^{-3}	1020.09	1499	4.3616	4.2003	1.5293
5x10^{-3}	1020.15	1508	4.31256	4.1766	1.5380
6x10^{-3}	1020.2	1513	4.28441	4.1630	1.5431
7x10^{-3}	1020.24	1517	4.25658	4.1494	1.5482
8x10^{-3}	1020.28	1522	4.23235	4.1376	1.5526
9x10^{-3}	1020.32	1525	4.21164	4.1275	1.5565

Table-2 Concentration (m), Relative association (R$_A$), Apparent molal compressibility (ϕ_κ), Apparent molal volume (ϕ_v) , Solvation number (S$_n$)-

Concentration (m) moles lit^{-1}	Apparent molal volume (ϕ_v) m^{3}mole^{-1}	Apparent molar compressibility (ϕ_k) x10^{-10} m^{2}N^{-1}	Relative association (R$_A$)	Solvation number (S$_n$)
1x10^{-3}	0.44660	2.394	0.99935	0.99496
2x10^{-3}	0.45137	2.387	0.99831	0.99211
3x10^{-3}	0.45614	2.377	0.99739	0.98797
4x10^{-3}	0.46091	2.367	0.99639	0.98361
5x10^{-3}	0.46569	2.34	0.99365	0.97246
6x10^{-3}	0.47047	2.324	0.99209	0.96604
7x10^{-3}	0.47526	2.309	0.99052	0.95971
8x10^{-3}	0.47885	2.296	0.98916	0.95419
9x10^{-3}	0.48163	2.284	0.98799	0.94947

Table-3 Limiting Apparent molal compressibility (ϕ^0_κ), Limiting Apparent molal volume (ϕ^0_v), S$_v$ and S$_k$

Ligand	Limiting Apparent molal volume (ϕ^0_v) m^{3}mole^{-1}	Limiting Apparent molal compressibility (ϕ^0_κ)x10^{-10} m^{2}N^{-1}	S$_v$ m^{3}kg$^{1/2}$mole$^{-3/2}$	S$_k$ m^{3}mole^{-2}kg.N^{-1}
Schiff base of pyrazolone	0.4027	2.4162	4.5066	-14.867
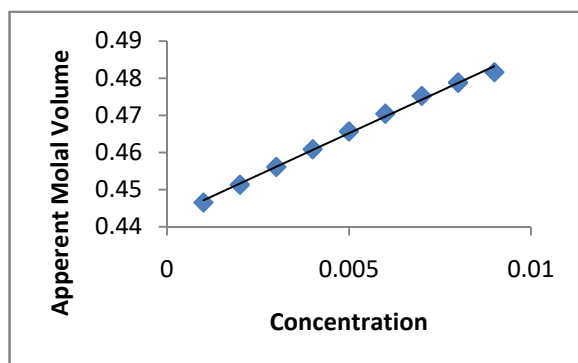
Fig.-1 -Apparent molal volume ($m^3 mole^{-1}$) 10^9 Vs Concentration (mole lit^{-1})
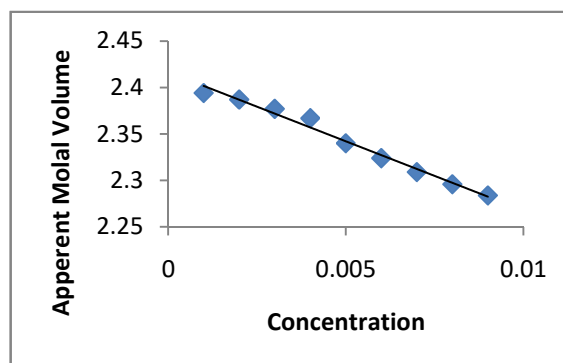
Fig.-2- Aparent molar compressibility10^{-9} ($m^2 N^{-1}$) Vs Concentration (mole lit^{-1})

CONCLUSION

In present study the acoustical properties were intended from experimental data, it shows that there are interface between ion-ion and solvent-solute exists between Schiff base of (5-hydroxy -3-methyl-1-(2, 4dinitrophenyl)-pyrazol-4-yl)(phenyl) methanone and 4-amino antipyrine in 1,4 dioxane-water solvent. And from the investigational statistics it is resolved that there are interaction among the solute and solvent molecule in Schiff base of benzoyl Pyrazolone & dioxane-water systems are strong.

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ULTRASONIC VELOCITY, DENSITY MEASUREMENT OF SUBSTITUTED HETEROCYCLIC DRUG IN DIOXANE-WATER AT 303.15 k

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Abstract

The acoustical properties have been investigated from the ultrasonic velocity and density measurements of substituted heterocyclic drug in mixed medium at 300K. The measurement have been perform to evaluate acoustical parameter such as adiabatic compressibility (βs), Partial molal volume (ϕv), intermolecular free length (Lf), apparent molal compressibility ($\square k$), specific acoustic impedance (Z), relative association (RA), solvation number (Sn).

 Key word: - Ultrasonic velocity, viscosity, adiabatic compressibility, apparent molal volume.

Subject Classification: Physical chemistry.

INTRODUCTION

In the recent years, measurements of the Ultrasonic velocity are helpful to interpreted solute-solvent, ion-solvent interaction in aqueous and non aqueous medium [1-6]. Fumio Kawaizumi[5] have been studied the acoustical properties of complex in water. Jahagirdar et. al. has studied the acoustical properties of four different drugs in methanol and he drawn conclusion from adiabatic compressibility . The four different drugs compress the solvent methanol to the same extent but it shows different solute-solvent interaction due to their different size, shape and structure [6]. Meshram et. al. studies the different acoustical properties of some substituted Pyrazolines in binary mixture acetone-water and observed variation of ultrasonic velocity with concentration[7]. Palani have investigated the measurement of ultrasonic velocity and density of amino acid in aqueous magnesium acetate at constant temperature [8]. The ion-dipole interaction mainly depends on ion size and polarity of solvent. The strength of ion-dipole attraction is directly proportional to the size of the ions, magnitude of dipole. But inversely proportional to the distance between ion and molecules. Voleisines has been studied the structural properties of solution of lanthanide salt by measuring ultrasonic velocity [9]. Syal et.al. has been studied the ultrasonic velocity of PEG-8000, PEG- study of acoustical properties of substituted heterocyclic compounds under suitable condition[10].Tadkalkar et.al. have studied the acoustical and thermodynamic properties of citric acid in water at different temperature[11]. Mishra et.al. have investigated ultrasonic velocity and density in non aqueous solution of metal complex and evaluate acoustic properties of metal complex[13].M. Arvinthraj et.al. have determined the

acoustic properties for the mixture of amines with amide in benzene at 303K-313K .They also determined thermodynamic parameters[14].S.K. Thakur et.al. have studied the different acoustical parameters of binary mixture of 1-propanol and water [14]. Mirikar et.sal. studied the molecular interaction between liquids.[14]

Preliminaries:

$$Adiabetic\ comprressibility(\beta o) = \frac{1}{Uo^2 do}$$

$$Adiabetic\ comprressibility(\beta s) = \frac{1}{Us^2 ds}$$

$$Apparent\ molal\ volume(\phi_v) = \left(\frac{M}{ds}\right) x\ \frac{(d_o - d_s)x10^3}{m\ x\ d_s\ x\ d_o}$$

$$Apparent\ molal\ compressibility(\phi_k) = 1000x\ \frac{(\beta_s d_o - \beta_o d_s)x10^3}{m\ x\ d_s\ x\ d_o} + \frac{\beta_s M}{d_o}$$

$$Specific\ acoustic\ impedance\ (Z) = U_s d_s$$

$$Intermolecular\ free\ length\ (L_f) = K\sqrt{}\ ds$$

$$Relative\ association\ (R_A) = x\ 1\left(\frac{d_s}{d_o}\right)^{1/3}$$

$$Solvation\ number(Sn) = \frac{\phi_k}{\beta_o\left(\frac{M}{d_o}\right)}$$

$$\phi_\kappa = \phi^0{}_\kappa + S_k C$$
$$\phi_\varpi = \phi^0{}_\varpi + \Sigma_\varsigma X$$

Statement of problem:

After review of literature survey the detail study of substituted heterocyclic drug under identical set of experimental condition is still lacking. It was thought of interest to study the acoustical properties of substituted heterocyclic drug under suitable condition.

Material and Method

The double distilled 20% dioxane was used for preparation of different concentration of drug solution. The densities were determined by using specific gravity bottle by relative measurement method with accuracy □ $1x10^{-5}$ gm/cm^3. The ultrasonic velocities were measured by using ultrasonic interferometer having frequency 3MHz.The constant temperature was maintained by circulating water through the double wall measuring cell, made up of steel.

In the present investigation, different properties such as adiabatic compressibility (□s), apparent molal volume (□v), intermolecular free length (L$_f$), apparent molal compressibility (□$_□$), specific acoustic impedance (Z), relative association (R$_A$), solvation number(S$_n$), limiting apparent molal compressibility (□$^□$$_□$), limiting apparent molal volume(□$^□$$_v$) and their constant (S$_k$, S$_v$).

Results and Discussion

In the present investigation, different acoustical properties such as ultrasonic velocity (Us), adiabatic compressibility (□s), intermolecular free length (L$_f$), specific acoustic impedance (Z), are listed in table-1. Partial molal volume (φ$_v$), apparent molal compressibility (φ$_k$), relative association (R$_A$), solvation number (Sn) are listed in table-2. Limiting apparent molal compressibility (φ^0$_\kappa$), limiting apparent

molal volume☐☐$_v$) and their constant (S$_k$, S$_v$) are listed in table-3. It was found that the ultrasonic velocity decreased with the increase in concentration for system (Table-1). Variation of ultrasonic velocity in solution depends upon the increase or decrease of molecular free length after mixing the component. This is based on a model for sound propagation proposed by Eyring and Kincaud[13]. Intermolecular free length increased linearly on increase in concentration of substituted heterocyclic drug (Naloxone) in mixed medium. Hence, decreased in ultrasonic velocity with increase in concentration of drug. It happened because there was significant interaction between ions and solvent molecules suggesting a structure promoting behavior of the added electrolyte. The specific acoustic impedance (Z) increased with the decrease in concentration of drug in aqueous medium. When concentration of electrolyte was increased, the thickness of oppositely charged ionic atmosphere increases due to decrease in ionic strength. This is suggested by decrease in acoustic impedance with concentration in system. It was seen that the intermolecular free length increased with the increase in concentration in system. The intermolecular free length increased due to greater force of attraction between solute and solvent by forming hydrogen bonding. The adiabatic compressibility increased with the increase in concentration of solution. It happened due to collection of solvent molecule around ions, this supporting weak ion-solvent interaction. This indicates that there is significant solute-solvent interaction.

It was observed that apparent molal volume increased with concentration in system. It indicates the existence of strong ion-solvent interaction. It was found that the value of apparent adiabatic compressibility was increased with the increase in concentration of Naloxone in mixed medium. It shows strong electrostatic attractive force in the vicinity of ions. From the data, we were concluded that strong molecular association was found in drug. The value of relative association increased with the increase in concentration in system. It has been found that there was strong interaction between solute and solvent. There were regular increases in solvation number with the increase in concentration; it indicates the solvent molecule forms strong coordination bond in primary layer. It indicates the increase in size of secondary layer of Solvation. The value of S$_k$ exhibits positive. It indicates the existence of ion-ion or solute-solute interactions in naloxone system. The value of S$_k$ exhibits positive, it indicates the strong existence of ion-ion or solute-solute interactions. From table-3, it was found that the value of limiting apparent molal volume was positive. It indicates that the ion-dipolar interaction in naloxone and mixed medium. The positive value of Sv indicates the strong solute-solvent interaction. This value indicates an induced effect of water on solute-solvent interaction. The value of S$_k$ and Sv has been determine from fig. 1 and 2.

Lables of fig. and tables :

Table-1 Ultrasonic velocity, density, adiabatic compressibility (☐$_S$), Specific acoustic impedance (Z) Intermolecular free length (L$_f$).

Concentration moles lit^{-1} (m)	Density (ds) kg m-^{3}	Ultrasonic velocity (Us) m s^{-1}	Adiabatic compressibiliy ($\square_S$) x10^{-10} m^{2}N^{-1}	Intermolecular free length (L$_f$) x10^{-11} m	Specific acoustic impedance (Zx10^6) kg m^{-2} s^{-1}
1x10^{-3}	1030.98	1584.90	3.86029	3.95155	1.634476
2x10^{-3}	1031.28	1579.92	3.88364	3.96348	1.629767
3x10^{-3}	1031.55	1570.56	3.92908	3.98660	1.620519
4x10^{-3}	1031.81	1553.76	4.01350	4.02920	1.603589
5x10^{-3}	1032.07	1539.26	4.08840	4.06662	1.589039
6x10^{-3}	1032.34	1526.36	4.15683	4.10051	1.576088
7x10^{-3}	1032.58	1512.98	4.22998	4.13642	1.562545
8x10^{-3}	1032.76	1498.25	4.31273	4.17670	1.547617
9x10^{-3}	1032.95	1480.12	4.41903	4.22860	1.528889

Table-2 Concentration (m), Relative association (R$_A$), Apparent molal compressibility ($\square_\square$), Apparent molal volume ($\square_v$) , Solvation number (S$_n$)-

Concentration(m) moles lit^{-1}	Apparent molal volume ($\square_v$) m^{3}mole^{-1}	Apparent molar compressibility ($\square_{k\square}\square$x10^{-10}) m^2 N^{-1}	Relative association (R$_A$)	Solvation number (S$_n$)
1x10^{-3}	0.15232	1.75064	1.003724	1.013889
2x10^{-3}	0.16169	1.76076	1.005413	1.019751
3x10^{-3}	0.17418	1.78099	1.008493	1.031467
4x10^{-3}	0.18276	1.81896	1.013999	1.053458
5x10^{-3}	0.18789	1.85259	1.018845	1.072935
6x10^{-3}	0.18971	1.88323	1.023201	1.090679
7x10^{-3}	0.19504	1.91607	1.027767	1.109699
8x10^{-3}	0.20608	1.95336	1.032856	1.131296
9x10^{-3}	0.21361	2.00133	1.039203	1.159078

Table-3 Limiting Apparent molal compressibility ($\square^\square_\square$), Limiting Apparent molal volume

$(\square^{\square}_{v})$, S_v and S_k

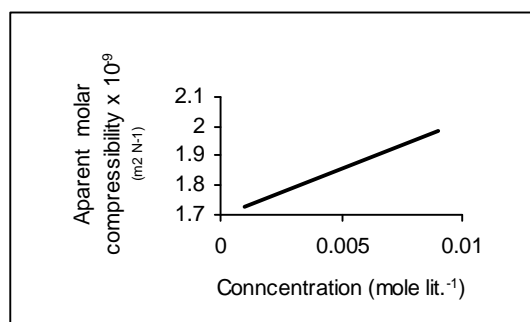| Limiting Apparent molal volume $(\square^{\square}_{v})$ $m^3 mole^{-1}$	Limiting Apparent molal compressibility $(\square^{\square}_{\square})x 10^{-9}$ $m^2 N^{-1}$	S_v $m^3 kg^{1/2}$ $mole^{-3/2}$	S_k $m^3 mole^{-2} kg.$ N^{-1}
0.1492	1.6980	7.1166	31.917

Fig.-1 -Apparent molal volume ($m^3 mole^{-1}$) Vs Concentration (mole lit^{-1})
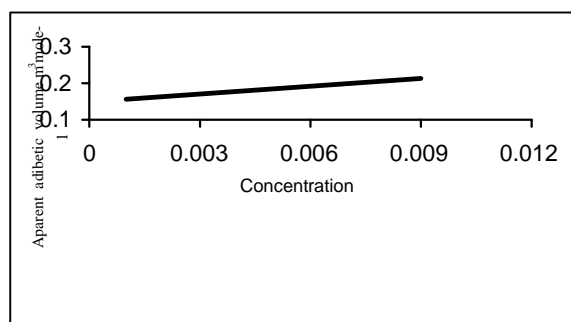

Fig.-2- Aparent molar compressibility10^{-9} ($m^2 N^{-1}$)   Vs Concentration (mole lit^{-1})

Conclusion

The present study shows the experimental data for ultrasonic velocity, density and viscosity at 300.15K for substituted heterocyclic drug in aqueous medium. From experimental data, the acoustical properties were calculated. The solute-solvent interaction and ion-ion / solute-solute interaction existing between drug and aqueous medium were also studied with the help of experimental data. Lastly it has been concluded from the experimental data, that the solute-solvent interaction in drug - aqueous medium systems are weak.

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STUDIES OF STABILITY CONSTANT OF 2-(THIAZOL-4'-YL)-*1H*-BENZIMIDAZOLE-1-ACETIC ACID WITH TRANSION METAL IONS IN % ETHYL ALCOHOL AT 303.15K.

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ABSTRACT

The interaction of transion metal ion with 2-(Thiazol-4'-yl)-*1H*-benzimidazole-1-acetic acid have been investigated by pH metric titration at 0.1 M ionic strength at 303.15k in 70 % Ethanol-Water mixture. The data obtained use to estimate the values of proton-ligand stability constant (P^k) and Metal -ligand stability constant (logK). It is observed that lanthanide ion form 1:1, 1: 2 complexes with all the systems.

INTRODUCTION

The studies of metal-ligand complexes in solution of a number of metal ions with carboxylic acids, oximes, phenols etc. would be interesting which throw light on the mode of storage and transport f metal ion in the biological kingdom.

In the earlier papers extensive data base on metal complexes with substituted heterocyclic drugs was presented.

Narwade et.al.[1] studies the Formation and Stability constant of thorium (IV) complex with some substituted pyrazolines. Mathieu W.A. Steenland et.al.[2] studies stability constant of Cu(II) and Ni(II) complexes of trans –dioxopentaaza macrocycles in aqueous solution by different technique. Hong-Wen Gaoet.al.[3] has studied the stability constant of Cu (II) and Co (II) complexes with CNBAC in water sample spectrophotometrically. Tuba Sismanoglu[4] have studied the stability constant of binary complexes of Nicotinamide with Mn (II) by pH metrically. He also determines change in free energy, change in enthalpy and change in entropy from stability constant at different temperature. Tekade et.al.[5] have been studied complex formation of Cu (II) and Co (II) metal ion complex with substituted isoxazolines. O.Yamauchi et.al.[6] studied stability constant of metal complexes amino acids with charged side chain by pH–metrically. Hayati Sari et.al.[7] studied the stability constant of glyoxime derivative and their Nickel, Copper, Cobalt and Zinc complexes potentiometric and theoretically. AH Naggar et al studied the stability constant of binary and ternary complexes of sulphamethoxazole and glycine with metal ions by potentiometry and conductometry.[8]

After review of literature survey the detail study of complex under identical set of experimental condition is still lacking. It was thought of interest to study the chelating properties of substituted heterocyclic compound under suitable condition with lanthanide by pH metrically.

MATERIAL AND METHOD

pH measurement were carried out with equip-tronic EQ-610 pH meter (accuracy ± 0.01 units) using combine glass electrode at room temperature. Metal ions prepared in triply distill water and concentration estimated by standard method.[9] The solution of drugs prepared in solvent. The pH metric reading in 70% ethyl alcohol – water mixture were converted to [H^+] value by applying the correction proposed by Van Uitert Haas.
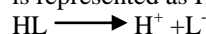
The overall ionic strength of solution was constant and calculated by the equation,

$$\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box\Box C_i Z_i^2$$

The concentration of other ion in addition to Na^+ and ClO_4^- were also taken into consideration.

RESULT AND DISCUSSION

Substituted heterocyclic drugs may be ionized as acid having replaceable H^+ ion from -OH group. Therefore it is represented as HL i.e.

HL $\longrightarrow$ H^+ + L^-

The titration data used to construct the curves between volume of NaOH and $P^{H.}$ They are called acid-ligand titration curves.

It is observed from titration curves for all systems ligand start deviating from the free acid curves at P^H 2.5 and deviating continuously up to P^H =11. The deviation shows that dissociation of proton in substituted drugs.

The average number of proton associated with the ligand (n_A) was determined from free acid and acid - ligand titration curves employing the equation of Irving and Rossotti.[10] The P^k values were determined from formation curves (n_A $_v^s$ P^H) by noting the P^H at which n_A =0.5. The accurate values of pk were calculated by point wise calculations which are presented in table -1.

Table 1: Determination of Proton-Ligand Stability Constant (Pk) of Some Substituted Heterocyclic Drugs at 0.1m Ionic Strrngth.

System	Constant pK	
	Half integral	Point wise calculation
Ligand	6.45	6.495. ± 0.05

Metal -Ligand Stability Constant (Log K)

Metal-ligand stability constant of transition metal ions chelate with some substituted heterocyclic compound were determined by employing Bjerrum calvin P^H metric titration method as adopted by Irving and Rossotti. The formation of chelate between transition metal ions with some substituted heterocyclic compound were indicated by the significant separation starting from pH =2.5 for transition metal ions with ligand. The change of colour from colourless to faint yellow to orange colour complex form.

Table 2: Determination of Metal - Ligand Stability Constant (Logk) of Yb(Iii), Pr(Iii) And Ce(Iii) With Some Substituted Heterocyclic Drugs At 0.1m Ionic Strrngth.

System	Metal ion	$Logk_1$	$Logk_2$	$Logk_1 - Logk_2$	$Logk_1 / Logk_2$
Ligand	Fe(II)	4.70	5.45	0.75	1.1596
	Cu(II)	4.95	5.50	0.55	1.1111
	Co(II)	4.85	5.60	0.75	0.9573
	Ni(II)	4.55	5.75	1.20	1.2637
	Zn(II)	5.30	5.95	0.65	1.1226

The result shows the ratio of $Logk_2$ /$Logk_1$ is positive in all cases. This implies that there is little or no steric hindrance to the addition of secondary ligand molecule. The smaller difference may be due to trans structure.

ACKNOWLEDGEMENTS

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STUDIES OF ACOUSTIC PROPERTIES OF AMINO ACID IN 10% SODIUM BENZOATE AT 303.15K.

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ABSTRACT

Acoustical properties have been measured for aspartic acid in aqueous sodium benzoate at different temperature. The measurement have been perform to evaluate acoustical parameter such as adiabatic compressibility (βs), Partial molal volume (ϕ_v), intermolecular free length (L_f), apparent molal compressibility (ϕ_κ), specific acoustic impedance (Z), relative association (R_A), salvation number (Sn) and also studied the molar polarization.

KEY WORD: Molar polarization, polarizability constant, Ultrasonic velocity, intermolecular free length, relative association.

INTRODUCTION

Amino acids are common components of all organisms. Protein of all species made from the amino acids. Protein plays many different biological roles in living systems. One particularly important function is to serve as the building blocks of proteins. The recent data about viscosity, density, ultrasonic velocity and other physical parameter to study interaction within the system, acoustic parameter and transport properties of aqueous amino acids electrolytes[1] are very helpful to obtain data about various types of interactions in solutions. Mirikar et al have been studied the solution properties of liquid system consisting of polar as well as non polar components find applications in industrial and technology processes.[2]

In the recent years, measurements of the Ultrasonic velocity are helpful to interpreted solute-solvent, ion-solvent interaction in aqueous and non aqueous medium.[3] Fumio Kawaizumi[4] have been studied the acoustical properties of complex in water. Jahagirdar et. al. has studied the acoustical properties of four different drugs in methanol and he drawn conclusion from adiabatic compressibility. The four different drugs compress the solvent methanol to the same extent but it shows different solute-solvent interaction due to their different size, shape and structure.[5] Meshram et. al. studies the different acoustical properties of some substituted Pyrazolines in binary mixture acetone-water and observed variation of ultrasonic velocity with concentration.[6] Palani have investigated the measurement of ultrasonic velocity and density of amino acid in aqueous magnesium acetate at constant temperature.[7] The ion-dipole interaction mainly depends on ion size and polarity of solvent. The strength of ion-dipole attraction is directly proportional to the size of the ions, magnitude of dipole. But inversely proportional to the distance between ion and molecules. Voleisines has been studied the structural properties of solution of lanthanide salt by measuring ultrasonic velocity.[8] Syal et.al. has been studied the ultrasonic velocity of PEG-8000, PEG- study of acoustical properties of substituted heterocyclic compounds under suitable condition.[9] Tadkalkar et.al. have studied the acoustical and thermodynamic properties of citric acid in water at different temperature.[10] Mishra et.al. have investigated ultrasonic velocity and density in non aqueous solution of metal complex and evaluate acoustic properties of metal complex.[11] M. Arvinthraj et.al. have determined the acoustic properties for the mixture of amines with amide in benzene at 303K-313K. They also determined thermodynamic parameters.[12] S.K. Thakur et.al. have studied the different acoustical parameters of binary mixture of 1-propanol and water.[13]

After review of literature survey the detail study of aspartic acid in aqueous sodium nitrate under identical set of experimental condition is still lacking. It was thought of interest to study the acoustical and thermodynamic properties of substituted heterocyclic drug under suitable condition.

EXPERIMENTAL

The constant temperature was maintained by circulating water through the double wall measuring cell, made up of glass. The flow time was also measured by using

digital clock (0.01 Sec).The aspartic acid in aqueous sodium nitrate is used in the present study. The density was determined by using specific gravity bottle by relative measurement method with accuracy $\pm$ 1x10^{-5} gm/cm^3. The ultrasonic velocity was measure by using ultrasonic interferometer having frequency 3MHz (Mittal Enterprises, Model No F-82). The constant temperature is mentioned by circulating water through the double wall measuring cell made up of steel.

In the present investigation different parameters such as adiabatic compressibility (β_s), apparent molal volume (ϕ_v), intermolecular free length (L_f), apparent molal compressibility (ϕ_κ), specific acoustic impedance (Z), relative association (R_A), Solvation number (S_n) were studied.

$$Adiabetic\ comprressibility(\beta o) = \frac{1}{U_o^2 d_o}$$

$$Adiabetic\ comprressibility(\beta s) = \frac{1}{U_s^2 d_s}$$

$$Apparent\ molal\ volume(\phi_v) = \left(\frac{M}{d_s}\right) x \ \frac{(d_o - d_s) x 10^8}{m\ x\ d_s\ x\ d_o}$$

$$Apparent\ molal\ compressibility(\phi_k) = 1000x\ \frac{(\beta_s d_o - \beta_o d_s) x 10^8}{m\ x\ d_s\ x\ d_o} + \frac{\beta_s M}{d_o}$$

$$Specific\ acoustic\ impedance\ (Z) = \ U_s d_s$$

$$Intermolecular\ free\ length\ (L_f) = \ K\sqrt{\Box} ds$$

$$Relative\ association\ (R_A) = \ x\ 1\left(\frac{d_s}{d_o}\right)^{1/3}$$

$$Solvation\ number(Sn) = \frac{\phi_k}{\beta_o\left(\frac{M}{d_o}\right)}$$

RESULTS AND DISCUSSION

In the present investigation, different thermodynamic parameters, such as adiabatic compressibility (β_s), Partial molal volume (ϕ_v), intermolecular free length (L_f), apparent molal compressibility (ϕ_k), specific acoustic impedance (Z), relative association (R_A), salvation number(Sn).

From table-1, these found that ultrasonic velocity increases with increase in temperature. Such an increase in ultrasonic velocity clearly shows that molecular association is being takes place in these mixtures. Variation of ultrasonic velocity in solution depends upon the increase or decrease of molecular free length after mixing the component, based on a model for sound propagation proposed by Eyring and Kincaid.[13] It was found that, intermolecular free length decreases linearly on increasing the temperature of solution. The intermolecular free length decrease due to less force of interaction between solute and solvent by forming hydrogen bonding. This was happened because there is less significant interaction between ions and solvent molecules suggesting a structure promoting behavior of the added electrolyte. This may also indicates that increase in number of free ions showing the occurrence of ionic association due to stronger ion-ion interaction. The value of specific acoustic impedance (Z) increases with increase in temperature. The increase of adiabatic compressibility is decrease with increase in temperature may be due to loss of solvent molecule around ions, this supporting stronger ion-solvent interaction. This indicates that there is not significant solute-solvent interaction. The decrease in adiabatic compressibility

following a increase in ultrasonic velocity showing there by stronger intermolecular interaction.

From table-2, it is observed that apparent molal volume increases with increase in temperature indicates the existence of weak ion-solvent interaction. The values of apparent molal volume are all positive values indicate the presence of weak solute solvent interaction.[14] The value of apparent molal compressibility is decrease with increase in temperature. It shows weak electrostatic attractive force in the vicinity of ions. It can be concluded that weak molecular association is found in solution. The value of relative association increases with increase in temperature of system. It is found that there is strong interaction between solute and solvent.

The Solvation number increase with increase in temperature due to weak solute-solvent interaction. There is regular decrease in solvation number with increase temperature indicates the decrease in size of secondary layer of Solvation. The Solvation number in all system decreases with increase in temperature indicates the solvent molecule forms weak coordination bond in primary layer.

Table-1: Ultrasonic velocity, density, adiabatic compressibility (β_S), Specific acoustic impedance (Z) Intermolecular free length (L_f) at different temperature.

Temperature(K)	Density(ds) Kg m^{-3}	Ultrasonic velocity (Us) m s^{-1}	Adiabatic compressibiliy (β_S) x10^{-10}m^{2}N^{-1}	Intermolecular free length (L_f) x10^{-11}m	Specific acoustic impedance (Zx10^6)kg m^{-2} s^{-1}
298.15	1123.48	1567.12	3.6244	3.8289	1.7606
303.15	1120.98	1574.84	3.5969	3.8144	1.7654
308.15	1116.82	1583.67	3.5702	3.8002	1.7687
313.15	1113.81	1591.52	3.5446	3.7865	1.7727
318.15	1109.97	1601.26	3.5137	3.7699	1.7774

Table-2: Relative association (R_A), apparent molal compressibility (ϕ_κ), Apparent molal volume (ϕ_v), Solvation number (S_n).

Temp. (K)	Apparent molal volume (ϕ_v) m^{3}mole^{-1}	Apparent molal compressibility ϕ_κ x10^{-11} m^2 N^{-1}	Relative association (R_A)	Adiabatic compressibiliy (β_o) x10^{-10}m^{2}N^{-1}	Solvation number (S_n)
298	0.1184	4.31941	1.00710	3.6244	1.03888
303	0.1187	4.29706	1.00695	3.6024	1.039887
308	0.1192	4.27898	1.00691	3.5716	1.043807
313	0.1195	4.26118	1.00686	3.5430	1.045045
318	0.1199	4.23826	1.00655	3.5144	1.049175

CONCLUSION

In the present study mentions the experimental data for ultrasonic velocity, density at different temperature for substituted heterocyclic drug in ethyl alcohol. From experimental data calculated acoustical parameters and studied to explanation solute-solvent interaction and ion-ion / solute-solute interaction are existing between aspartic acid and solvent mixture. From experimental data it can be conclude that weak solute-solvent interaction in all systems.

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