

Study of Solute-Solvent Interaction of Hydrazone Schiff's bases in N, N dimethyl formamide solution at 303K by measurement of Ultrasonic velocity, Density and Viscosity

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Abstract- Molecular interaction of hydrazone schiff's bases, 1-(1-hydroxynaphthalen-2-yl)ethanone – 2 - chlorobenzoylhydrazone (H_2L^1), 1-(1-hydroxynaphthalen-2-yl)ethanone – 4 - chlorobenzoylhydrazone (H_2L^2) was studied from the evaluated values of acoustical parameters like, specific acoustical impedance (Z), isentropic compressibility (K_s), Intermolecular free path length (L_f), Relaxation strength (r), Rao's molar sound function (R_m), Molar compressibility (W), Relative association (R_A), Relaxation time (τ), Ultrasonic attenuation (α/f^2), Free volume (V_f), Internal pressure (π), Apparent molar compressibility (ϕ_K), Apparent molar volume (ϕ_V), Solvation number (S_n). These acoustical properties are determined from the evaluated values of density (ρ), viscosity (η) and ultrasonic velocity (U) of hydrazone schiff's bases in DMF solvent at 303K with vares series of concentration. In the present work it is observed that, in DMF solvent, solute-solvent interactions are predominant.

Keywords: Acoustical parameters, Hydrazone Schiff's bases, N, N dimethylformamide, Solute-solvent interaction.

1. INTRODUCTION

Study of certain physical properties of materials is now well understood by means of examining several acoustic and thermodynamical parameters from determining values of ultrasonic velocity, density and viscosity of liquids [1], liquid mixtures [2, 3], solution of salts [4, 5], polymers [6], amino acids [7, 8], biological molecules [9, 10] etc. The study of molecular interaction plays a vital role in understanding structural behavior of solutions. Thus studies in solutions put forward extensive applications in the field of chemical and food industries [12, 13]. Several researchers study ultrasonic measurements for solutions of organic and inorganic compounds. Hydrazone Schiff's bases have been intensively investigated in several research fields because of its triatomic structure $>C=N-N$ and their chelating capability. Hydrazones have wide application in the field of biology, medicine, analytical chemistry, catalysis and optics [14]. From the literature survey, it is found that molecular interaction of Schiff's bases in different solvents at different temperature and concentration have been extensively studied by using ultrasonic velocity, density and viscosity [15-17]. However, study of acoustical properties of hydrazone Schiff's bases is very scanty. In the view of various applications of hydrazone Schiff's bases in

different fields, in the present work we studied molecular interactions of their solution in N, N dimethylformamide of various concentration by determining density, ultrasonic velocity and viscosity at 303K.

2. EXPERIMENTAL

2.1. Materials and methods

The hydrazone Schiff's bases, 1-(1-hydroxy naphthalen-2-yl)ethanone – 2 – chlorobenzoyl hydrazone (H_2L^1), 1-(1-hydroxynaphthalen-2-yl) ethanone – 4 – chlorobenzoylhydrazone (H_2L^2) used in this study was synthesized and recrystallized in our laboratory [18-19]. The structures of present synthesized hydrazone schiff's bases are shown in fig. 1. The solvent N, N dimethyl formamide (DMF) used in the present work is of AR grade and purified according to literature method [20].

2.2. Ultrasonic velocity, density and viscosity measurement

Ultrasonic interferometer (Mittal Enterprise, New Delhi, Model No.F-81), of frequency 2MHz was used to determine sound velocity (measured frequency of given instrument is 1.9912×10^6 Hertz), Pycnometer of borosil glass is used to determine density with accuracy of 0.1Kg/m^3 . It

was calibrated by measuring density of distilled water. The density of DMF was evaluated with

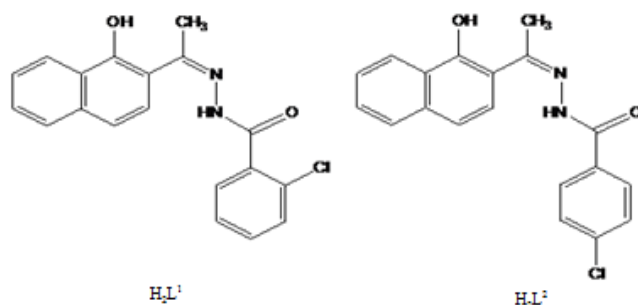


Fig. 1. Structure of 1-(1-hydroxynaphthalen-2-yl)ethanone – 2 - chlorobenzoylhydrazone (H_2L^1) and 1-(1-hydroxynaphthalen-2-yl)ethanone – 4 - chlorobenzoylhydrazone (H_2L^2)

respect to density of water. Precalibrated Ostwald's viscometer was used to determine viscosity of Schiff's bases with the help of digital stopclock with accuracy of 0.01 sec. Elite thermostat waterbath was used to maintain temperature with accuracy of 0.1K.

Theoretical equations

Various acoustical parameters are evaluated from the determined values of density (ρ), viscosity (η) and ultrasonic velocity (U) by using following standard equations,

Specific acoustical Impedance (Z)

$$Z = U\rho \quad (1)$$

Isentropic compressibility (K_s)

$$K_s = 1/U^2\rho \quad (2)$$

Intermolecular free path length (L_f)

$$L_f = K_j K_s^{1/2} \quad (3)$$

Where, K_j $\{(93.875+0.375T) \times 10^{-8}\}$ is the temperature dependent Jacobson constant.

Relaxation Strength (r)

$$r = 1 - (U/U_\infty)^2 \quad (4)$$

Where, $U_\infty = 1.6 \times 10^5 \text{ cmsec}^{-1}$

Rao's molar sound function (R_m)

$$R_m = (M/\rho)U^{1/3} \quad (5)$$

The apparent molecular weight (M) of the solution can be calculated according to following equation

$$M = M_1 W_1 + M_2 W_2$$

Where, W_1 and W_2 are weight fractions of solvent and solute respectively. M_1 and M_2 are the molecular weights of the solvent and compounds respectively.

Molar Compressibility (W)

$$W = (M/\rho) K_s^{-1/7} \quad (6)$$

Relative Association (R_A)

$$R_A = \rho/\rho_0 (U_0/U)^{1/3} \quad (7)$$

Where, U , U_0 and ρ , ρ_0 are ultrasonic velocities and densities of solution and solvent respectively.

Relaxation Time (τ)

$$\tau = \frac{4\eta}{3\rho U^2} \quad (8)$$

Ultrasonic Attenuation (α/f^2)

$$\alpha/f^2 = \frac{8\pi^2\eta}{\rho U^3} \quad (9)$$

Free Volume (V_f)

$$V_f = [M_{\text{eff}}U/K\eta]^{3/2} \quad (10)$$

Where, $K = 4.28 \times 10^9$

Internal pressure (π)

$$\pi = bRT [K\eta/U]^{1/2} [\rho^{2/3}/M^{7/6}] \quad (11)$$

Where, b is packing factor and its value is 2, R is gas constant ($8.314 \text{ JK}^{-1}\text{mole}^{-1}$) and T is absolute temperature.

Apparent Molar Compressibility (Φ_k)

$$\Phi_k = [(\rho_0 K_s - \rho K_s^\circ) 1000/C\rho_0] + [K_s^\circ M_2/\rho_0] \quad (12)$$

Where, M_2 is molecular weight of solute, ρ_0 and K_s° are density and isentropic compressibility of pure solvent, C is the concentration of solution in molarity.

Apparent Molar Volume (Φ_v)

$$\Phi_v = [M/\rho] - [1000(\rho - \rho_0) / \rho C] \quad (13)$$

Solvation number (S_n)

$$S_n = M_2/M_1 [1 - k_s / k_s^\circ] [(100 - X)/X] \quad (14)$$

Where, X is number of gram of solute in 100gm of solution, M_1 and M_2 are molecular weights of solvent and solute respectively.

3. RESULTS AND DISCUSSION

The density (ρ), viscosity (η) and ultrasonic velocity (U), of DMF and solutions of 1-(1-hydroxynaphthalen-2-yl)ethanone-2-chloro

benzoylhydrazone (H_2L^1), 1-(1-hydroxy naphthalene-2-yl)ethanone-4-chlorobenzoyl hydrazone (H_2L^2) in DMF were measured at 303K and are reported in Table 1.

Table 1. Density (ρ), ultrasonic velocity (U) and viscosity (η) of Schiff's bases H_2L^1 and H_2L^2 solutions in DMF at 303K.

Conc. M Mol dm ⁻³	Density(ρ) Kg m ⁻³	Velocity (U) ms ⁻¹	Viscosity (η) mPa s	Density(ρ) Kg m ⁻³	Velocity (U) ms ⁻¹	Viscosity (η) mPa s
	H_2L^1			H_2L^2		
0.000	941.5	1439.3	0.78102	941.5	1439.3	0.78102
0.001	942.3	1440.5	0.78343	942.1	1440.2	0.78189
0.002	942.8	1441.0	0.78901	942.8	1441.1	0.78451
0.003	943.4	1441.8	0.79244	943.4	1441.9	0.78929
0.004	944.0	1442.6	0.79795	943.8	1442.4	0.79491
0.005	944.5	1443.2	0.81178	944.3	1443.0	0.80790

Table 2. Acoustical properties of hydrazone schiff's bases in DMF at 303K.

Conc.(M) Mol dm ⁻³	Z × 10 ⁶ (Kg m ⁻² s ⁻¹)	Ks × 10 ⁻¹⁰ (Pa ⁻¹)	L _f × 10 ⁻¹¹ M	r	R _A	W × 10 ⁻³ (m ³ Pa ^{-8/3} mol ⁻¹)	R _m × 10 ⁻⁴ (m ^{10/3} s ^{-1/3} mol ⁻¹)
0.000	1.3551	5.12716	4.69847	0.1907	1.0000	1.64914	8.76625
H_2L^1							
0.001	1.3573	5.11427	4.69256	0.1894	1.00002	1.64855	8.76244
0.002	1.3585	5.10801	4.68969	0.1888	1.0002	1.64862	8.76226
0.003	1.3601	5.09910	4.6856	0.1879	1.00028	1.64842	8.76063
0.004	1.3618	5.09021	4.68151	0.1870	1.00036	1.64822	8.75900
0.005	1.3631	5.08329	4.67833	0.1863	1.00040	1.64810	8.75791
H_2L^2							
0.001	1.3568	5.11749	4.69404	0.1897	1.00001	1.64875	8.76369
0.002	1.3586	5.10730	4.68937	0.1887	1.00013	1.64865	8.76246
0.003	1.3602	5.09839	4.68527	0.1878	1.00021	1.64845	8.76083
0.004	1.3613	5.09270	4.68266	0.1872	1.00028	1.64845	8.76045
0.005	1.3626	5.0857	4.67947	0.1866	1.00040	1.64859	8.76033

Table 3. Acoustical properties of hydrazone Schiff's bases in DMF at 303K.

Conc. (M)	τ (10 ⁻¹⁰ sec)	α/f^2 (10 ⁻¹⁴ s ² m ⁻¹)	V _f × 10 ⁷ (m ³)	$\pi \times 10^{10}$ (Nm ⁻²)	$\Phi_k \times 10^7$ (m ⁵ N ⁻¹ mol ⁻¹)	Φ_v (m ³ mol ⁻¹)	S _n
0.000	5.33922	2.19451	1.76581	1.5604			
H_2L^1							
0.001	5.34224	2.19392	1.76127	1.5628	-17.241	-848.9862	0.33212
0.002	5.3737	2.20608	1.74353	1.5679	-13.11137	-689.4354	0.49346
0.003	5.38765	2.21057	1.73435	1.5710	-12.80073	-671.3306	0.72319
0.004	5.41565	2.22083	1.71853	1.5762	-12.64021	-662.0763	0.95238
0.005	5.50202	2.25531	1.67651	1.5896	-12.04154	-635.2567	1.13086
H_2L^2							
0.001	5.33509	2.19144	1.76488	1.5612	-12.93474	-636.8747	0.24919
0.002	5.34231	2.19304	1.75873	1.5633	-13.4657	-689.4354	0.51173
0.003	5.36549	2.20133	1.74492	1.5678	-13.03646	-671.3306	0.74141
0.004	5.39766	2.21376	1.72804	1.5731	-11.7452	-609.2389	0.88818

0.005	5.4784	2.24594	1.68826	1.5856	-11.32649	-593.0315	1.06677
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It is evident from Table 1 that, at 303K in DMF with increase in concentration, density, viscosity and ultrasonic velocity of both Schiff's bases H_2L^1 and H_2L^2 increases linearly. At particular temperature and concentration, wealth information regarding molecular interaction present in solution was given by determining its acoustical parameters. In the present investigation from Table 2, it is observed that with linear increase in ultrasonic velocity and density, there is increase in acoustic impedance (Z) and decrease in adiabatic compressibility (K_s) values with concentration as shown in fig. 1 and 2 respectively, which supports strong solute-solvent interactions [21].

Further it is clear from fig. 3 and values in Table 2 that the intermolecular free path length (L_f) values decreases as concentration increases, which suggest that the distance between solute and solvent molecules decreases due to increases in solute-solvent interactions, which causes increase in ultrasonic velocity.

It was supported from decrease in observed values of relaxation strength (r) and increase in values of relative association (R_A) with concentration indicate solute-solvent interaction dominates over solvent-solvent interaction as given in Table 2. Properties like molar sound function (R_m) and molar compressibility (W) observed to be increases with concentration for both Schiff's bases in DMF. This linear increase in these acoustical properties indicates absence of complex formation in system [22], as shown in Table 2.

Both ultrasonic attenuation (α/f^2) and relaxation time (τ) properties are depends upon ρ , η and U and temperature [23]. From fig. 4, 5 and Table 3 it is observed that both these values increased linearly with concentration of Schiff's bases, this increase supports structure making capacity of solute and absence of any solute-solute interactions. The increase of internal pressure (π) and decrease of free volume (V_f) values in Table 3 indicates the increase in cohesive forces [24]. At a particular temperature and pressure free volume (V_f) of solute is depends upon the internal pressure of liquid in which it dissolved. With decrease in free volume, internal pressure increases and vice versa, this again confirms existence of solute-solvent interactions in the system as shown in fig. 6, 7.

Solvation number (S_n) is another parameter which confirms interactions occurring in

the solution [25]. For both Schiff's bases H_2L^1 and H_2L^2 , the S_n values are positive and increases with concentration as shown in Table 3 and fig. 8. This positive value suggest structure forming tendency of studied compounds in DMF, this again proves solute-solvent interaction dominate in solution. In the present study, central moiety, solvent and molecular weight of both Schiff's bases are same, only the position of -Cl group is responsible for magnitude of molecular interactions.

Existence of solute-solvent interactions in solutions was further confirmed by apparent molar volume (ϕ_v) and apparent molar compressibility (ϕ_k) as shown in Table 3.

Further, the apparent molar compressibility (ϕ_k) of the solutions is fitted to the following Gucker's relation (15) [26],

$$\phi_k = \phi_k^\circ + S_k\sqrt{C} \quad (15)$$

Where, ϕ_k° and S_k are the limiting apparent molar compressibility at infinite dilution and interaction parameter respectively. From the plot of ϕ_k verses \sqrt{C} , ϕ_k° and S_k values were calculated from the intercept and slope of the plot respectively. These values are listed in Table 4.4a. The apparent molar volume (ϕ_v) is also related to concentration by Masson's equation (16) [27],

$$\phi_v = \phi_v^\circ + S_v\sqrt{C} \quad (16)$$

Where, ϕ_v° is the limiting apparent molar volume at infinite dilution and S_v is the solute-solvent interaction parameter. From the intercept and slope of the plot of ϕ_v verses \sqrt{C} , the ϕ_v° and S_v values were evaluated and are reported in Table 4. The type of intermolecular interaction in solutions can also be confirmed by evaluating the constants of the following Bachem's relation (17) [28],

$$K_s = K_s^\circ + AC + BC^{3/2} \quad (17)$$

Where, A and B are constants and C is molar concentration of solutions.

The constant A and B have been determined from the intercept and slope of the plots $(k_s - k_s^\circ)/C$ Vs $C^{1/2}$ and are also given in Table 4.

As shown in Table 4, values of A, ϕ_k° and ϕ_v° are negative whereas B, S_k and S_v values are positive for studied compounds. The negative values of ϕ_k° , ϕ_v° and A indicates the predominance of solute-solvent interactions in the studied solution. In a solution, when solute causes electrostriction, compressibility decreases which is reflected by negative values of ϕ_k° . This is again confirmed by the positive values of S_k and S_v [29].

Table 4. Coefficients of equations (15), (16) and (17) for Schiff's bases H₂L¹ and H₂L² at 303K temperature.

Ligands	$A.10^{10}$ $N^{-1}m^2Lmol^{-1}$	$B.10^{10}$ $N^{-1}m^2mol^{3/2}L^{3/2}$	$\phi_k.10^7$ $m^5N^{-1}mol^{-1}$	$S_k.10^7$ $m^5L^{1/2}$ $N^{-1}mol^{-3/2}$	ϕ_v m^3mol^{-1}	S_v $L^{1/2}m^3mol^{-3/2}$
H ₂ L ¹	-14.93291	93.732865	-19.87063	118.90448	-963.52660	4944.10856
H ₂ L ²	-11.32240	39.763735	-15.01195	47.34989	-720.23027	1513.69944

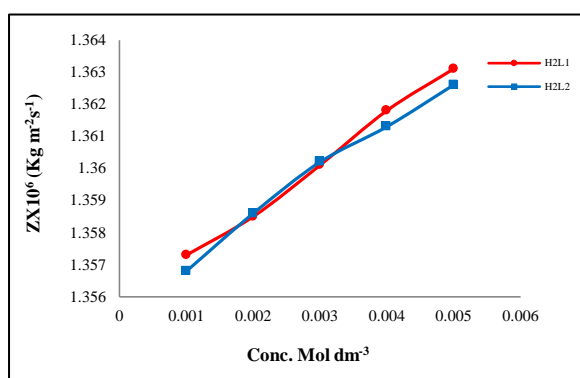


Fig. 1. Acoustic impedance Vs Conc. at 303K

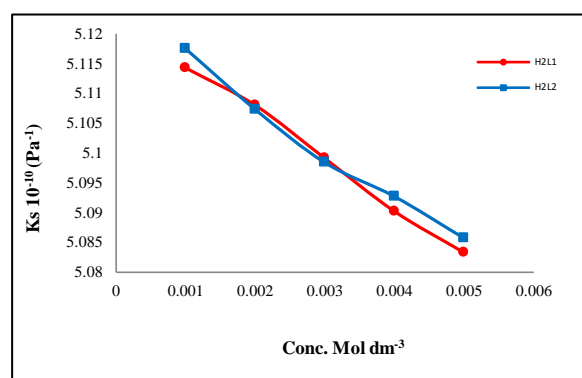


Fig. 2. Adiabatic compressibility Vs Conc.at303K

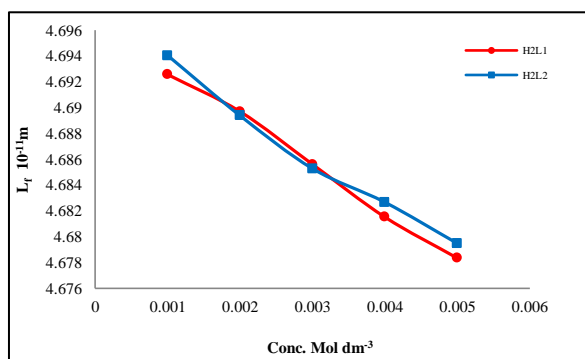


Fig. 3. Intermolecular free path length Vs Conc. at 303K

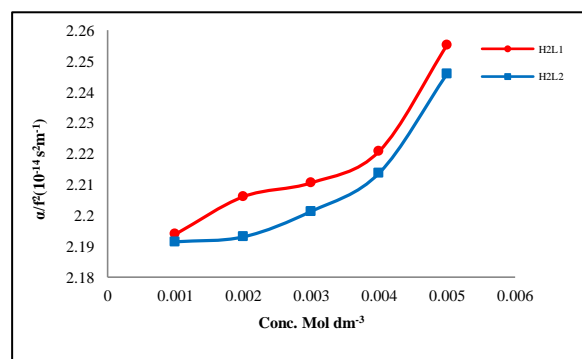


Fig. 4. Ultrasonic attenuation Vs Conc. at 303K

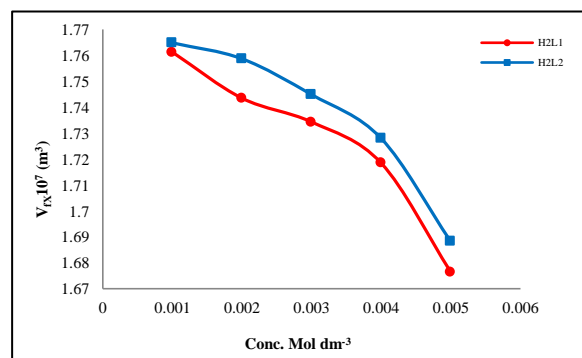
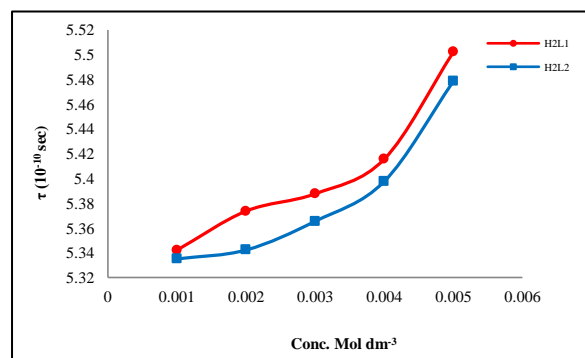


Fig. 5. Relaxation time Vs Conc. at 303K

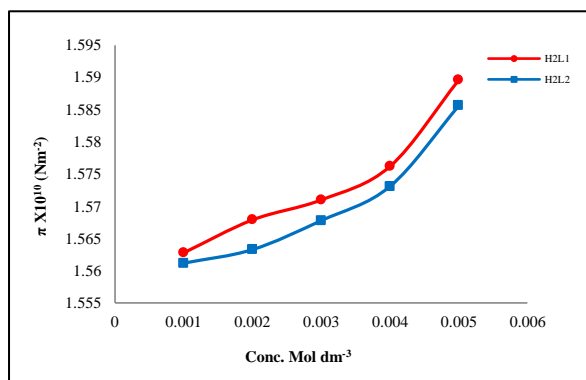


Fig. 6. Free volume Vs Conc. at 303K

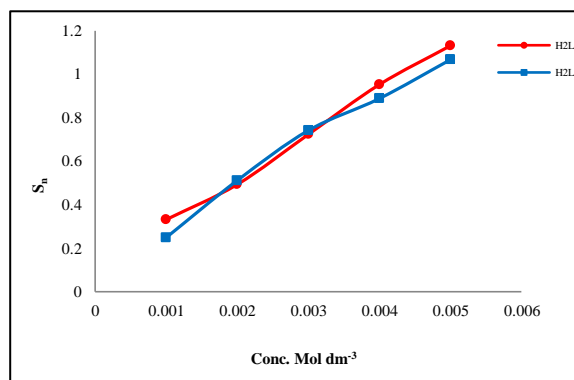


Fig. 7. Internal pressure Vs Conc. at 303K

Fig. 8. Solvation number Vs Conc. at 303K

4. CONCLUSION

The positive solvation suggests structure forming tendency of studied compounds in DMF solutions. This structure forming tendency of studied compounds is also confirmed by apparent molar properties. Overall, in the studied solution, solute-solvent interaction is more predominant over solute-solute interaction. These interactions in

solutions at constant temperature are affected by nature of solvent, concentration and structure of compounds. From the given study it shows that not only polarity, electronegativity, inductive effect of substituted groups affect the interaction but different position of groups also affects interaction in the same solution.

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